Neutrons and Magnetism

II. Inelastic scattering, dynamical scattering and polarimetry

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Plan

- Inelastic scattering
- Crystal fields
- Spin waves
- Generalized susceptibility
- Critical scattering
- Dynamical scattering
- Polarimetry

The cross-section

$$\frac{d^2\sigma}{d\Omega\cdot dE}=\frac{k'}{k}\left(\frac{m_n}{2\pi\hbar^2}\right)^2\sum_{\zeta,s}p_\zeta p_s\sum_{\zeta',s'}\left|\left\langle \boldsymbol{k}',s',\zeta'\middle|\hat{V}(\mathbf{r})\middle|\boldsymbol{k},s,\zeta\right\rangle\right|^2\delta\left(\hbar\omega+E_\zeta-E_{\zeta'}\right)$$

The *matrix element*, which contains all the physics.

This is also *time-dependent*, and contains the *dynamics* of the target.

The kinetic energy of a thermal neutron is about the same as the energy of interatomic magnetic vibrations, therefore neutrons can create or annihilate magnetic waves.

G. L. Squires, *Introduction to the theory of thermal neutron scattering*, Dover Publications, New York, 1978
W. Marshall and S. W. Lovesey, *Theory of thermal neutron scattering*, Oxford University Press, Oxford, 1971
S. W. Lovesey, *Theory of neutron scattering from condensed matter*, Oxford University Press, Oxford, 1986

Neutron inelastic scattering

Magnetic fluctuations are governed by a wave equation:

$$H\psi = E\,\psi$$

The Hamiltonian is given by the physics of the material.

Given a Hamiltonian, *H*, the energies *E* can be calculated.
(this is sometimes very difficult)

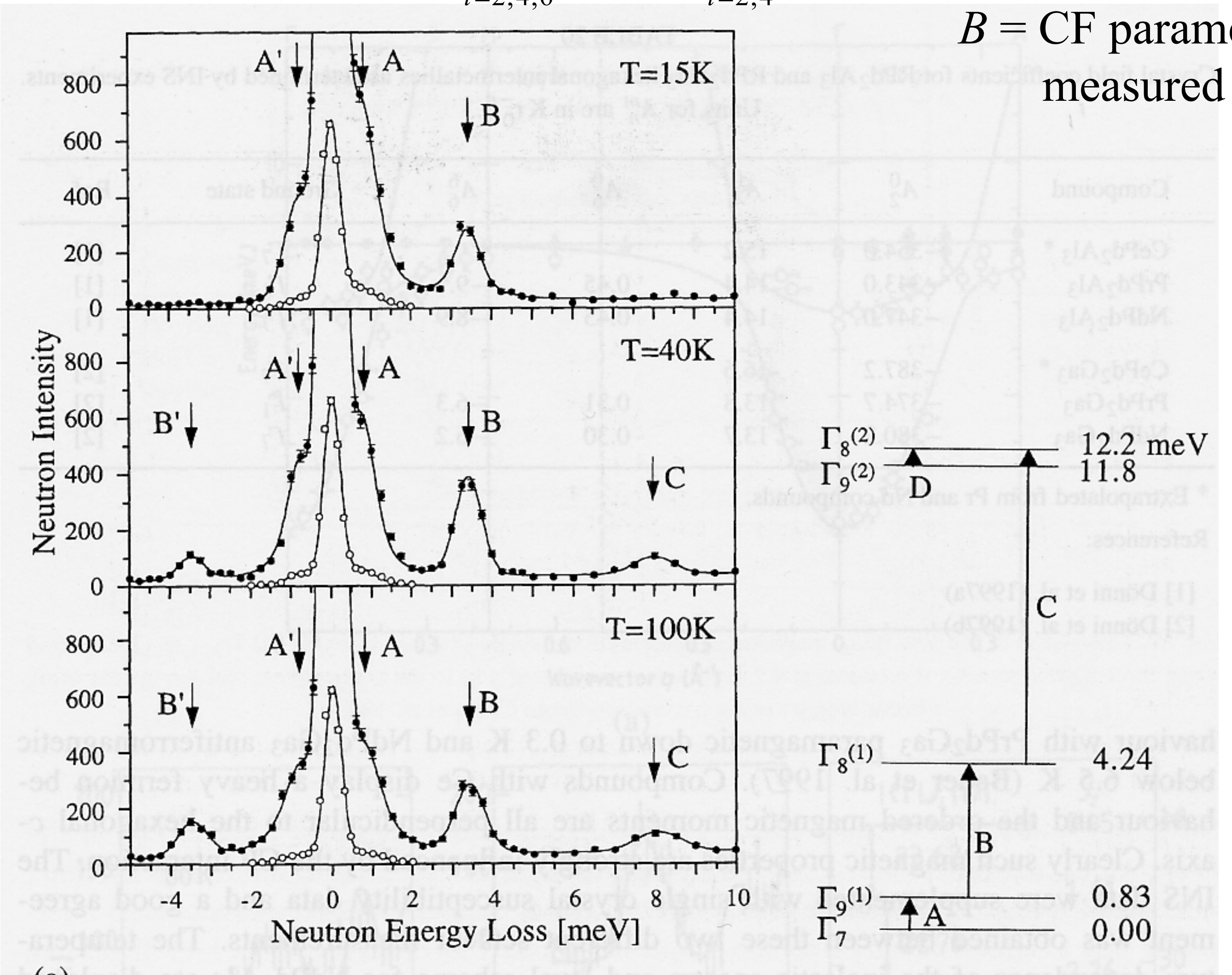
Neutrons measure the energy, *E*, of the magnetic fluctuations, therefore probing the free parameters in the Hamiltonian.

Crystal fields in $NdPd_2Al_3$

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

O = Stevens parameters
(K. W. Stevens, Proc. Phys. Soc A**65** (1952) 209)

B = CF parameters, measured by neutrons

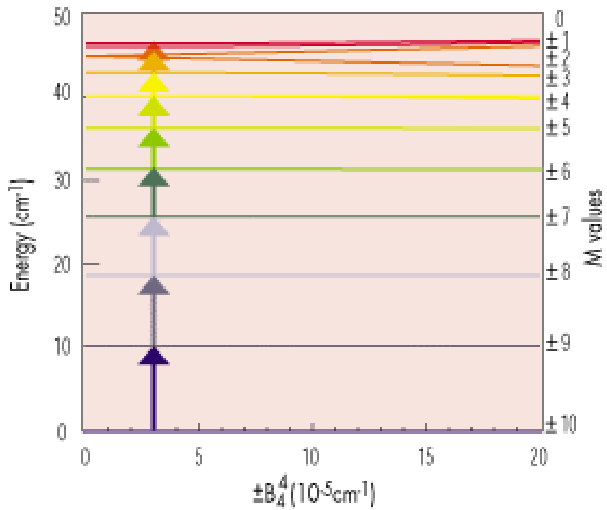


A. Döni *et al.*, J. Phys.: Condens. Matter **9** (1997) 5921

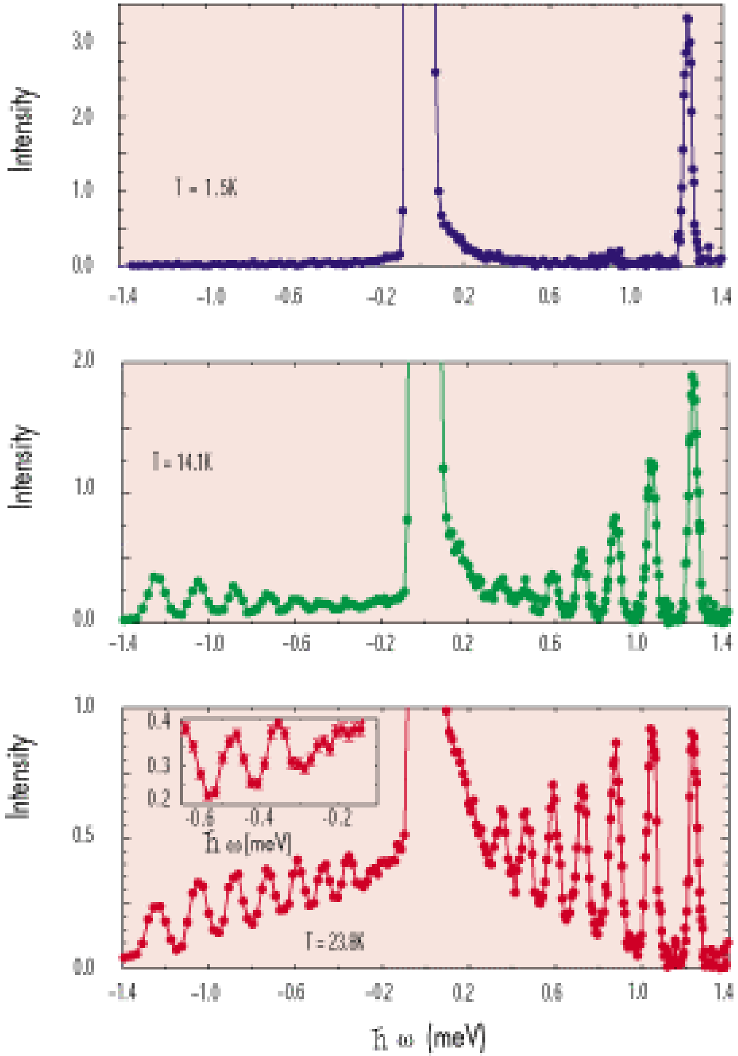
O. Moze., *Handbook of magnetic materials* vol. 11, 1998 Elsevier, Amsterdam, p.493

Quantum tunneling in Mn_{12}-acetate

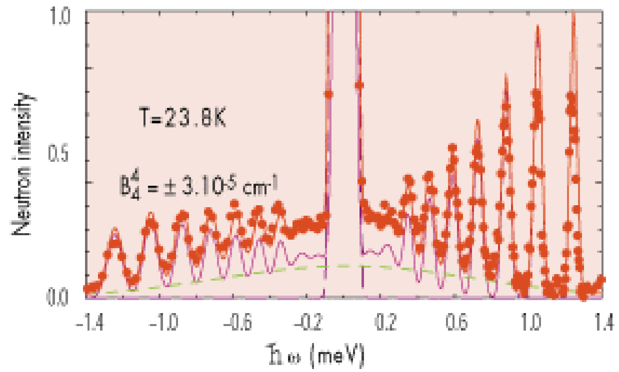
$$H = \sum_{l=2,4,6} B_l^0 O_l^0 + \sum_{l=2,4} B_l^4 O_l^4$$



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

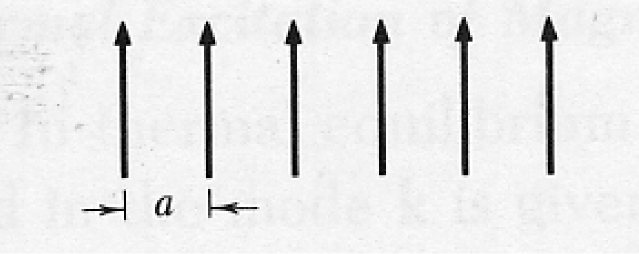
Spin waves and magnons

A simple Hamiltonian for spin waves is:

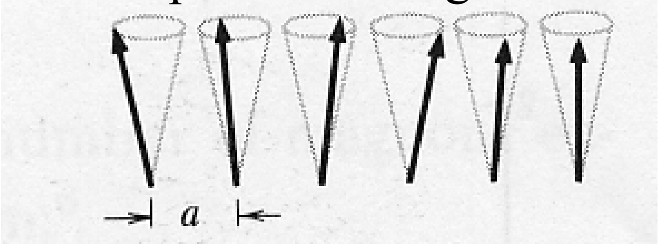
$$H = -J\sum_{i,j} \mathbf{s}_i \mathbf{s}_j$$

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

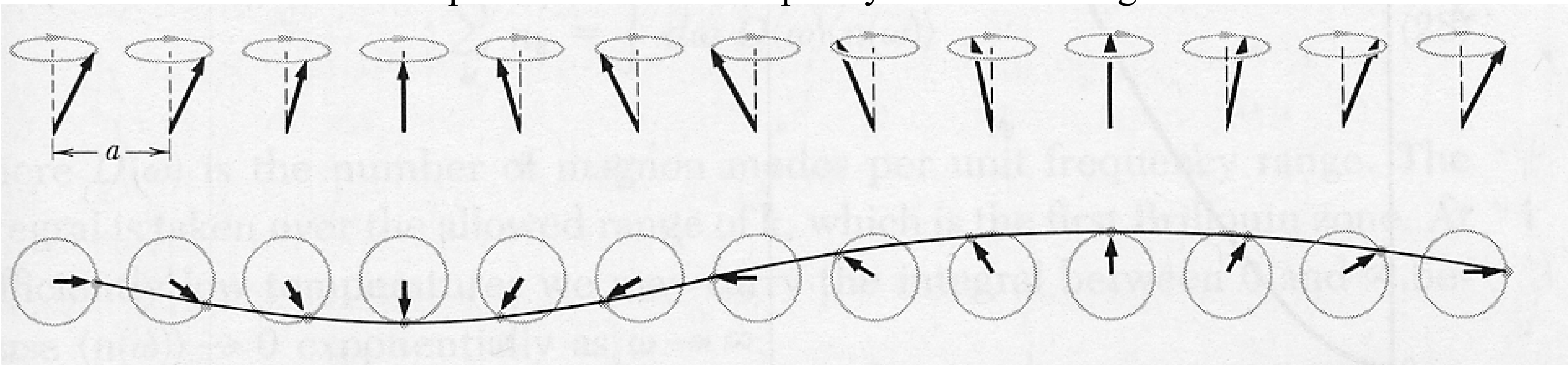
Take a simple ferromagnet:



The spin waves might look like this:



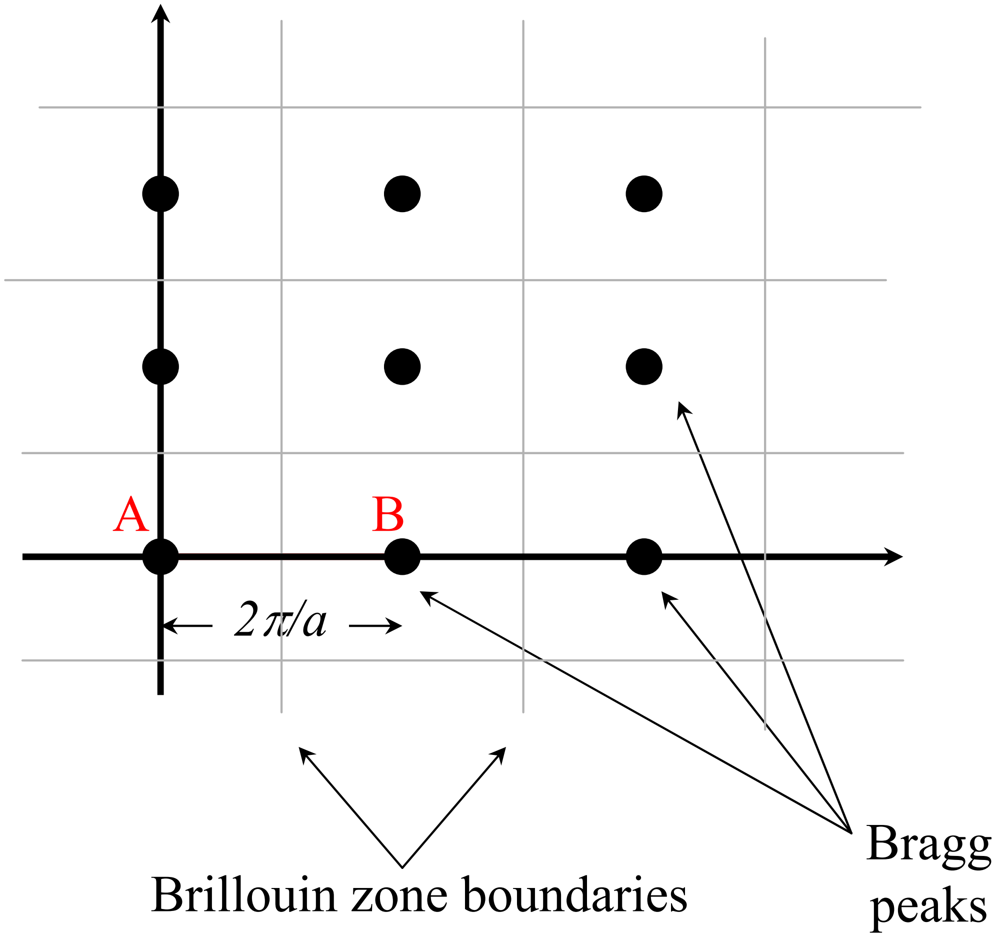
Spin waves have a frequency and a wavelength



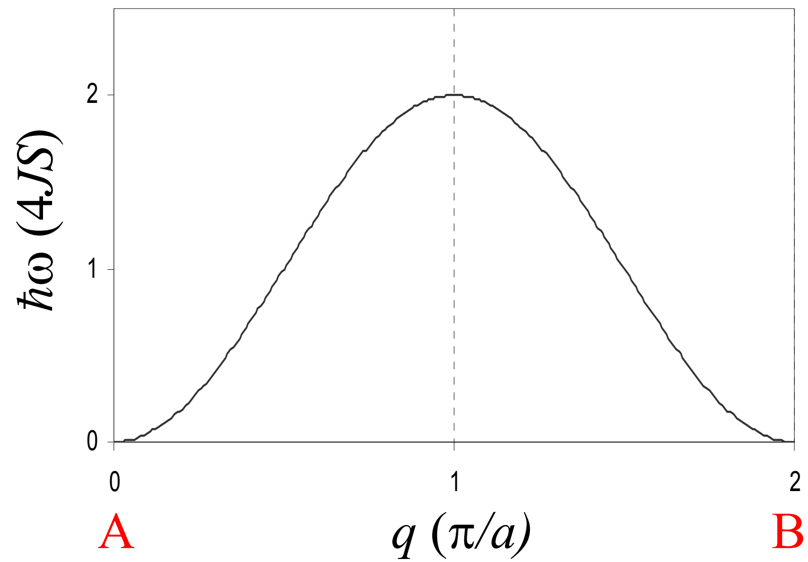
The frequency and wavelength of the waves are directly measurable with neutrons

Magnons and reciprocal space

Reciprocal space



Spin wave dispersion



$$\hbar\omega = 4JS(1 - \cos qa)$$

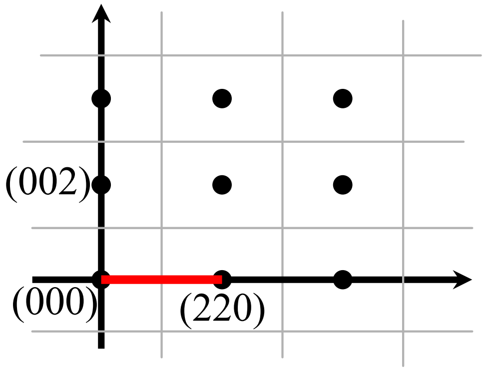
$$= Dq^2 \quad (\text{for } qa << 1)$$

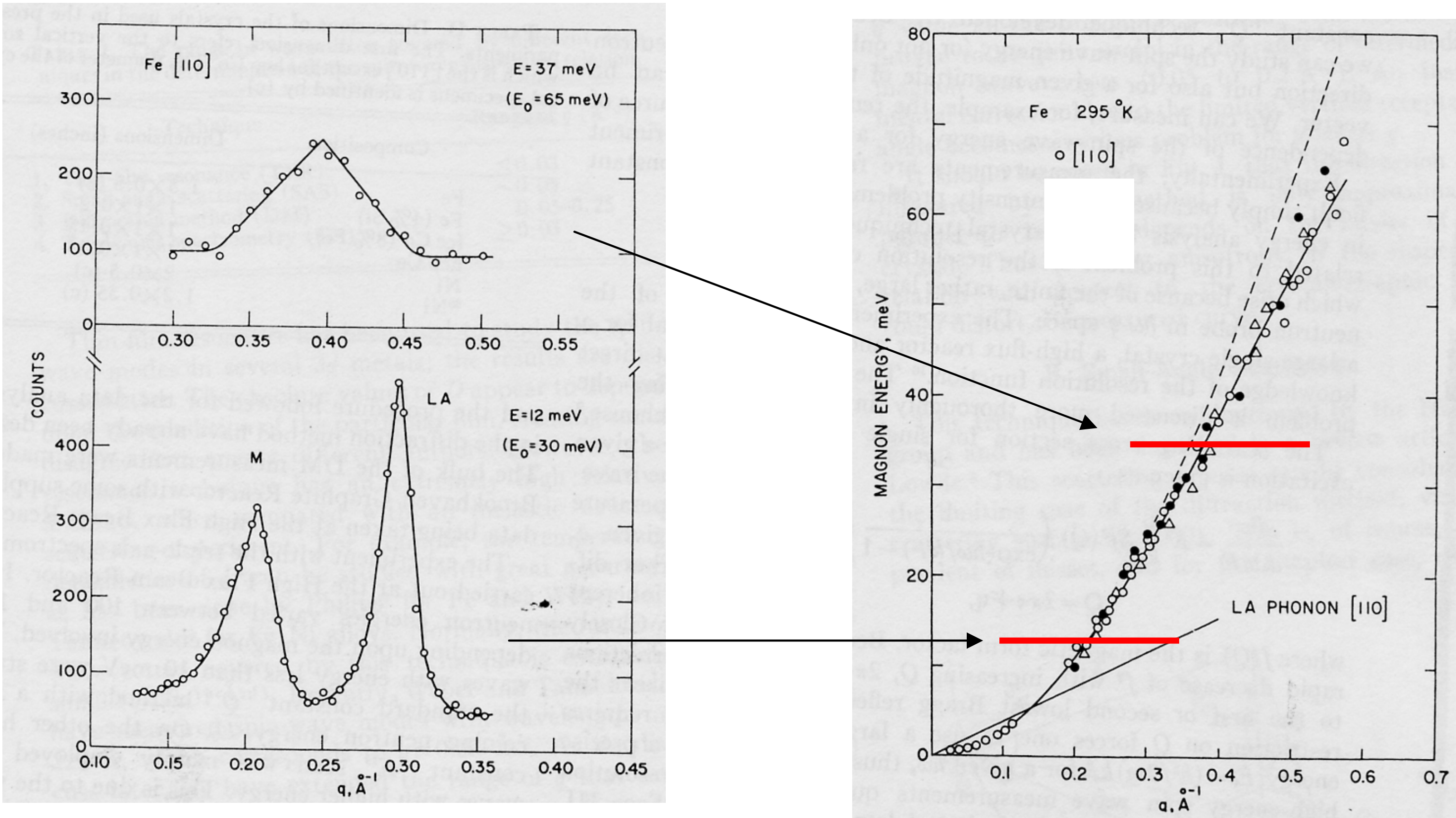
$$D = 2JSa^2$$

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

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

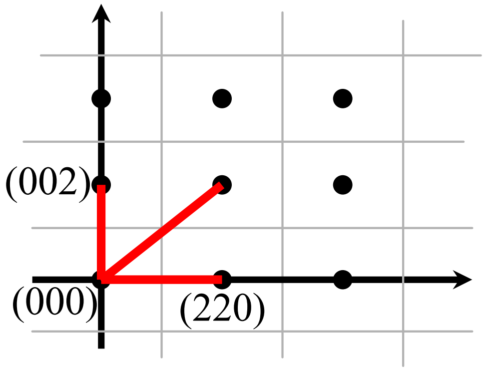
Magnons in crystalline iron

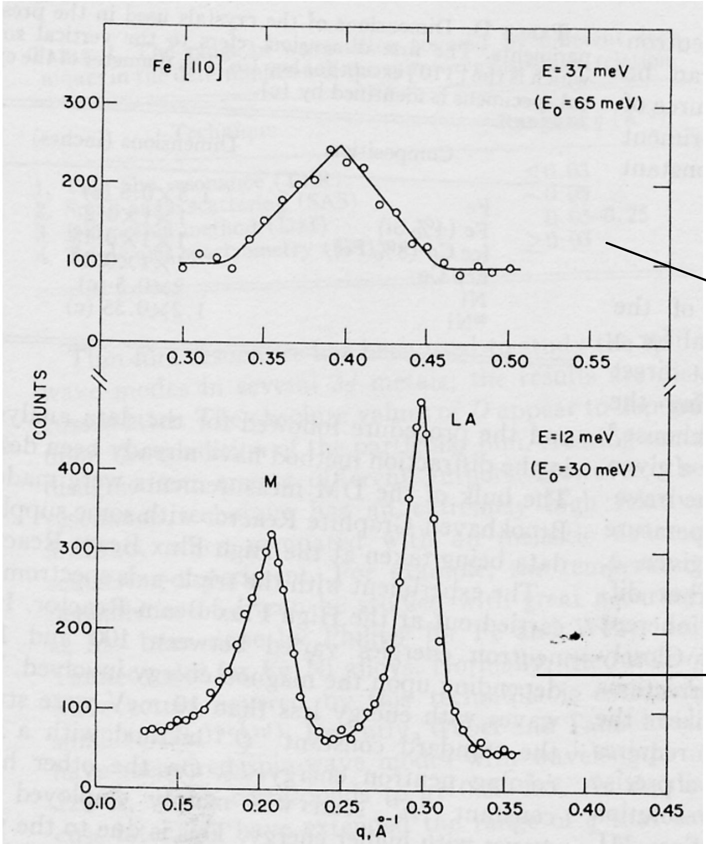


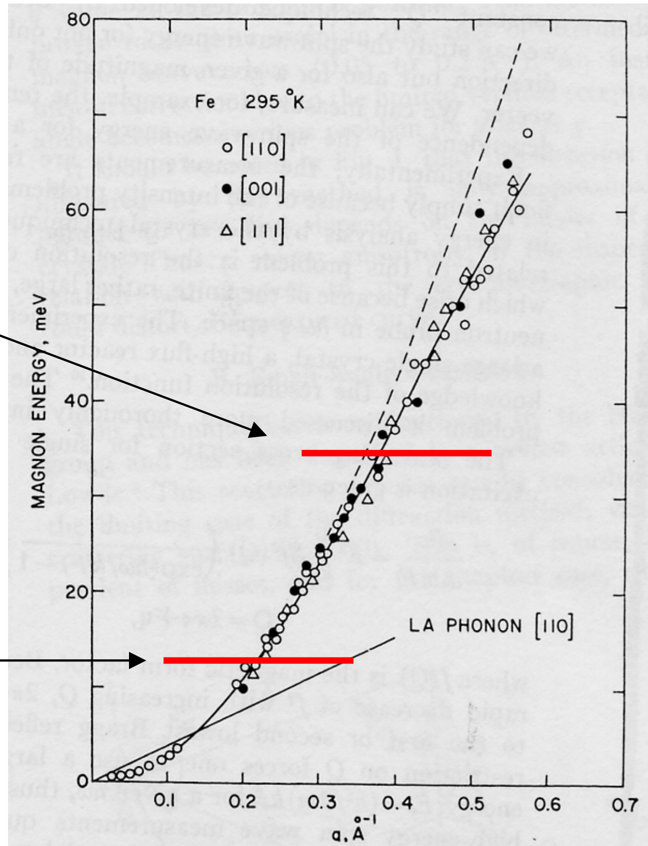


G. Shirane *et al*., J. Appl. Phys. **39** (1968) 383

Magnons in crystalline iron



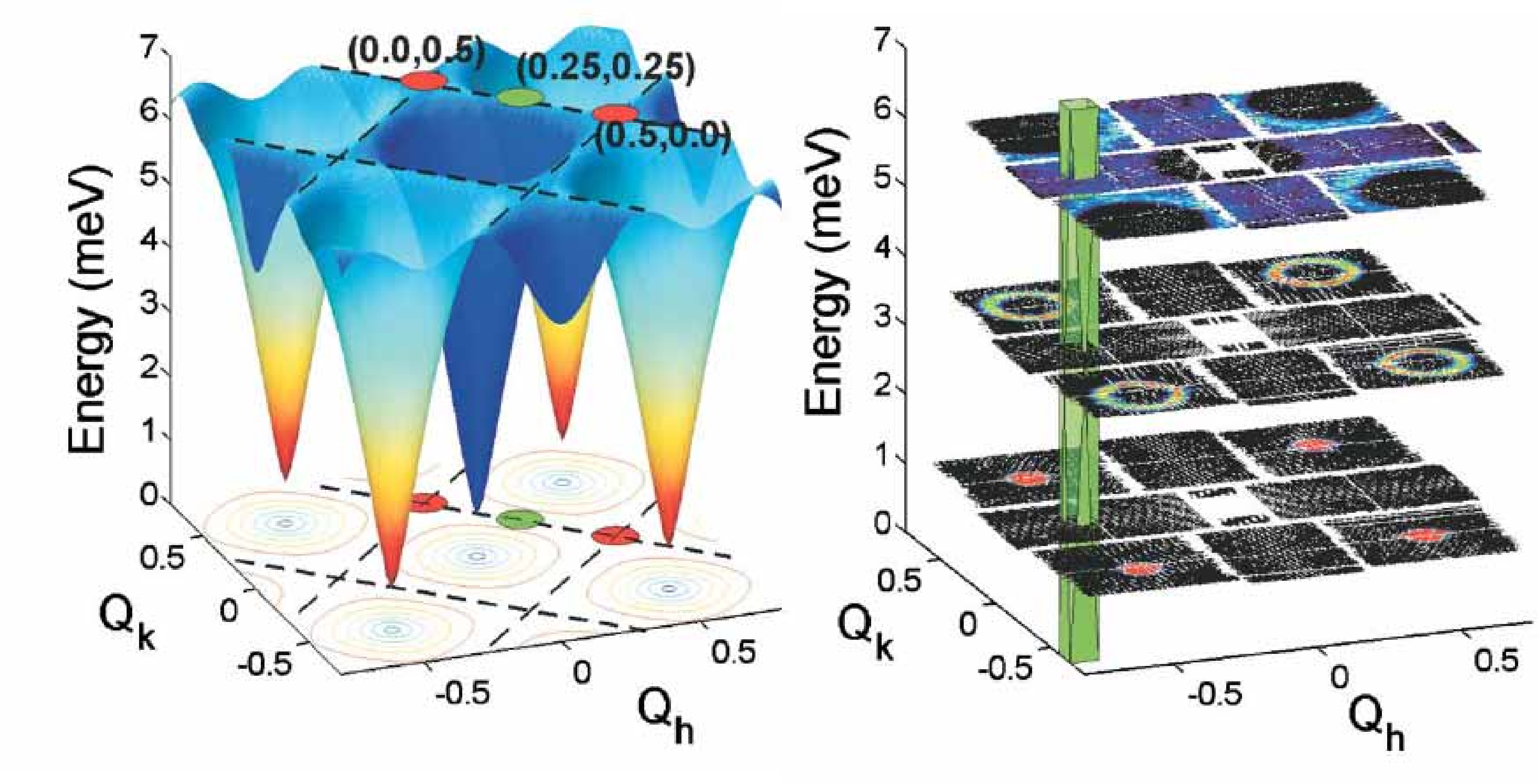




G. Shirane *et al*., J. Appl. Phys. **39** (1968) 383

Spin-waves in Rb_2MnF_4

A quasi-two dimensional antiferromagnetic system



T. Huberman *et al.*, Phys. Rev. B **72** (2005) 014413

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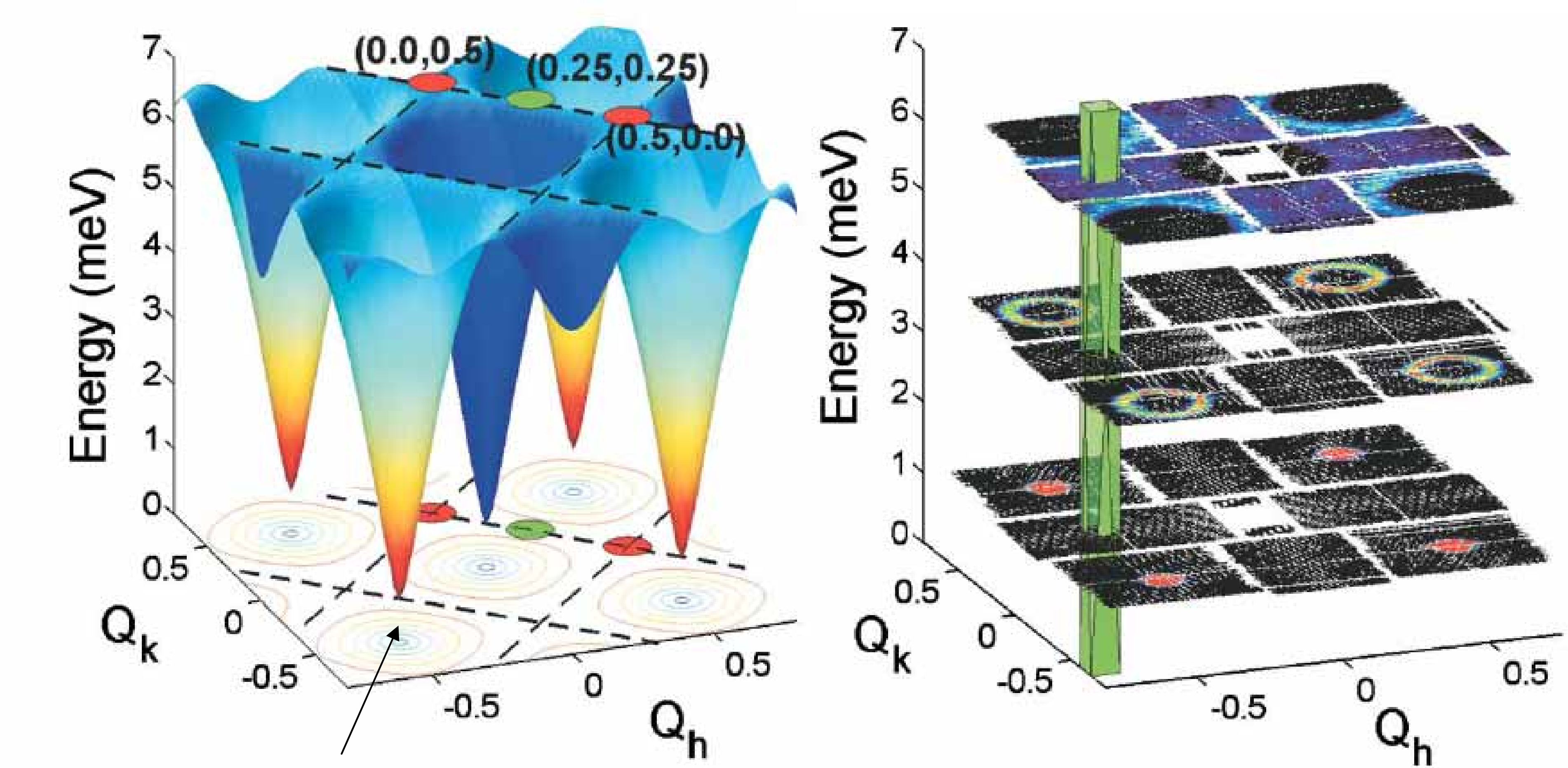
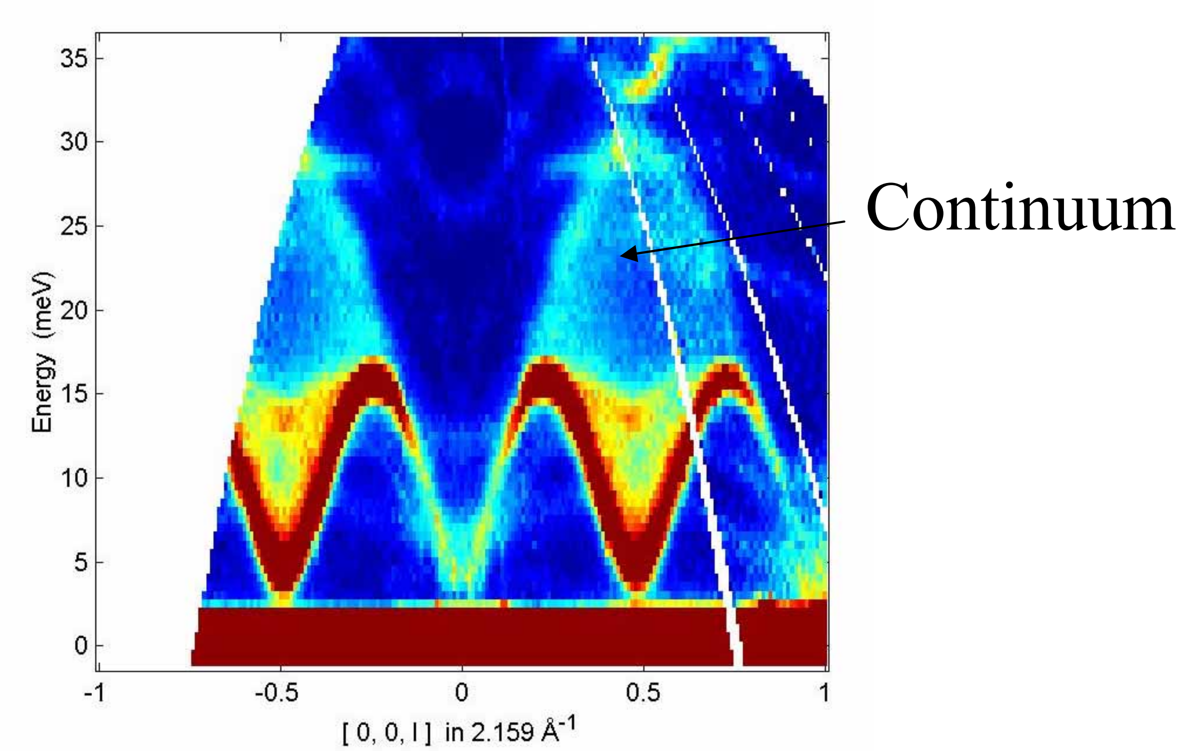


Gap in dispersion due to magnetic anisotropy

T. Huberman *et al.*, Phys. Rev. B **72** (2005) 014413

The Spin-Peierls system, CuGeO3

Initially thought to be a 1D antiferromagnet



B. Lake *et al.*, Nature Mater. **4** (2005) 329

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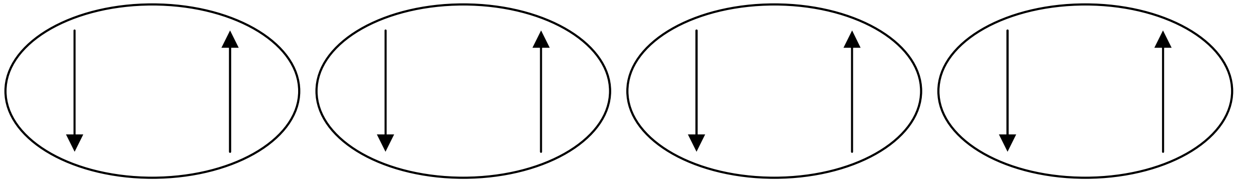
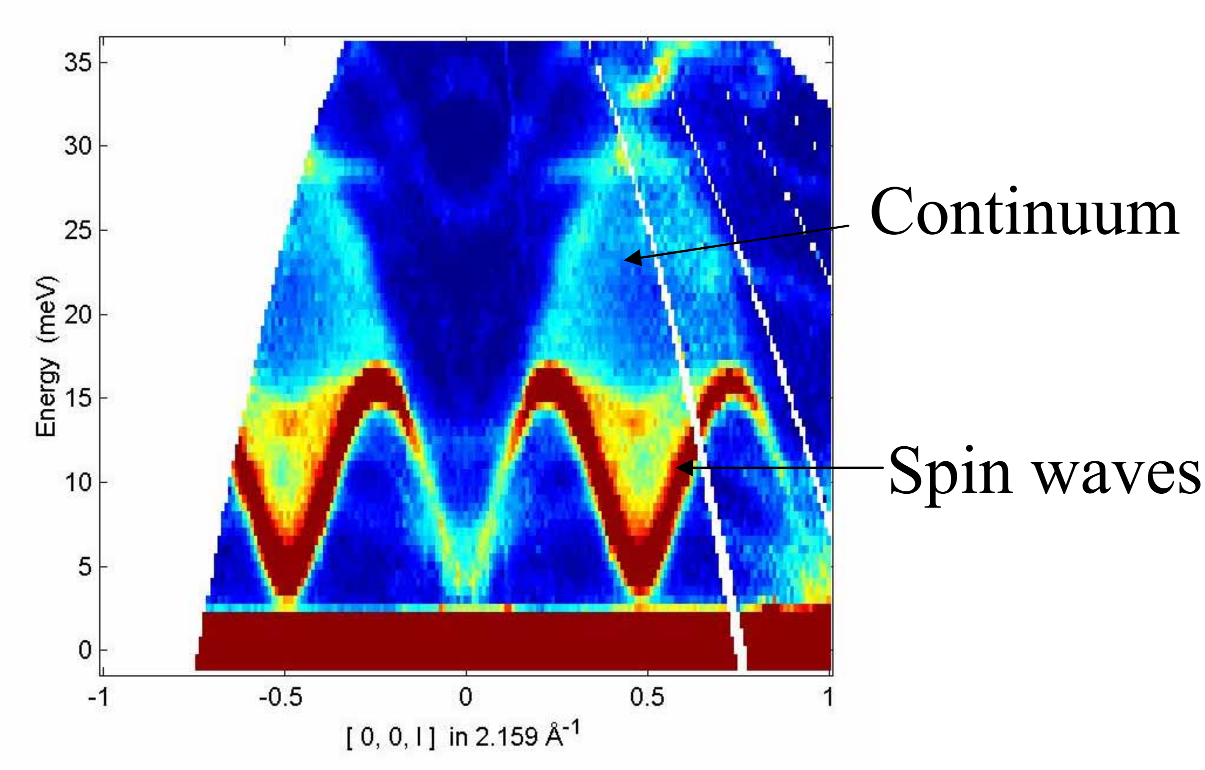
Initially thought to be a 1D antiferromagnet
Magnetic moments couple up, or *dimerize*





B. Lake *et al*., Nature Mater. **4** (2005) 329

The Spin-Peierls system, $CuGeO_3$

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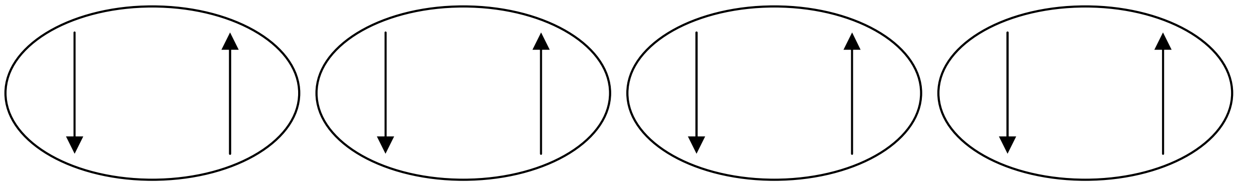
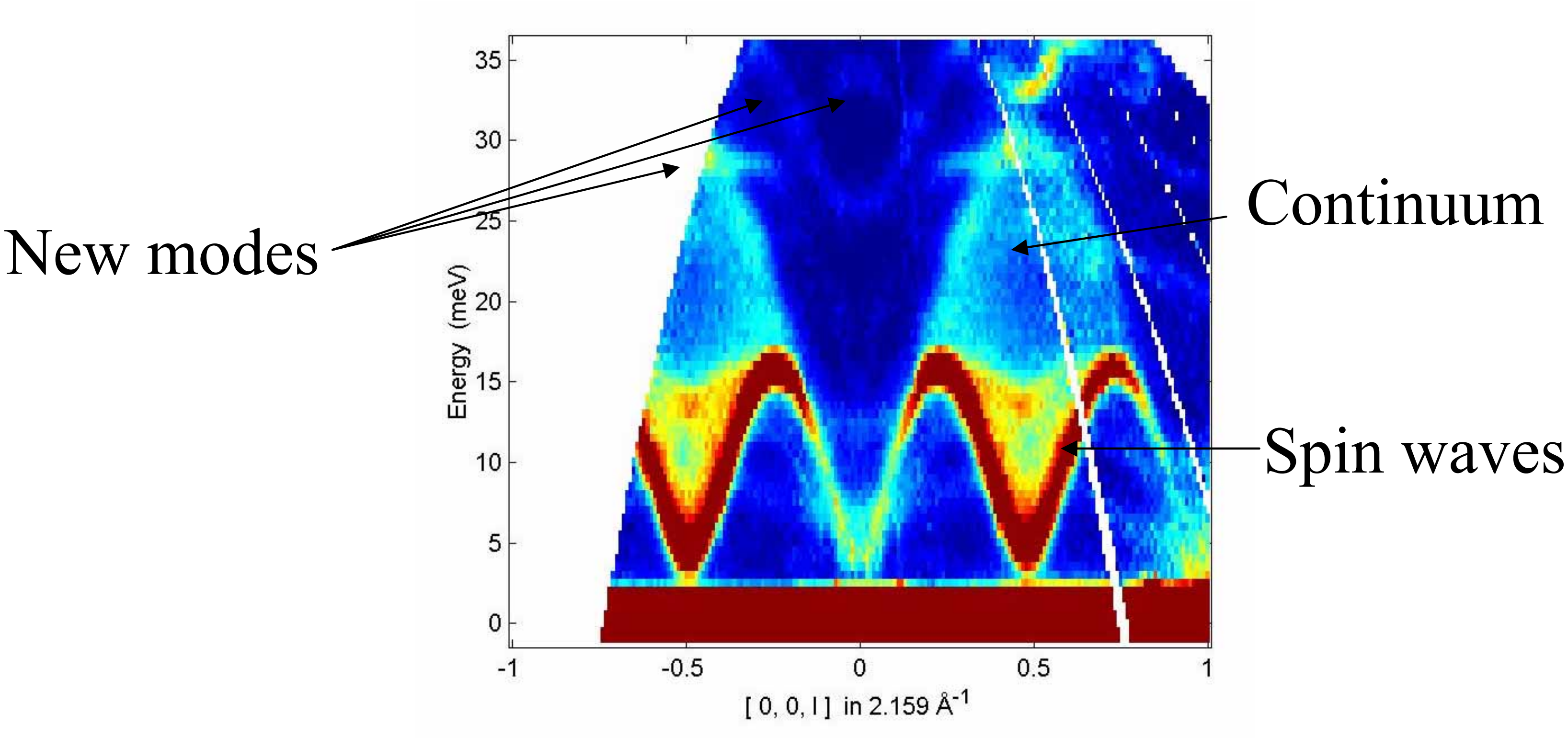
Magnetic moments couple up, or *dimerize*

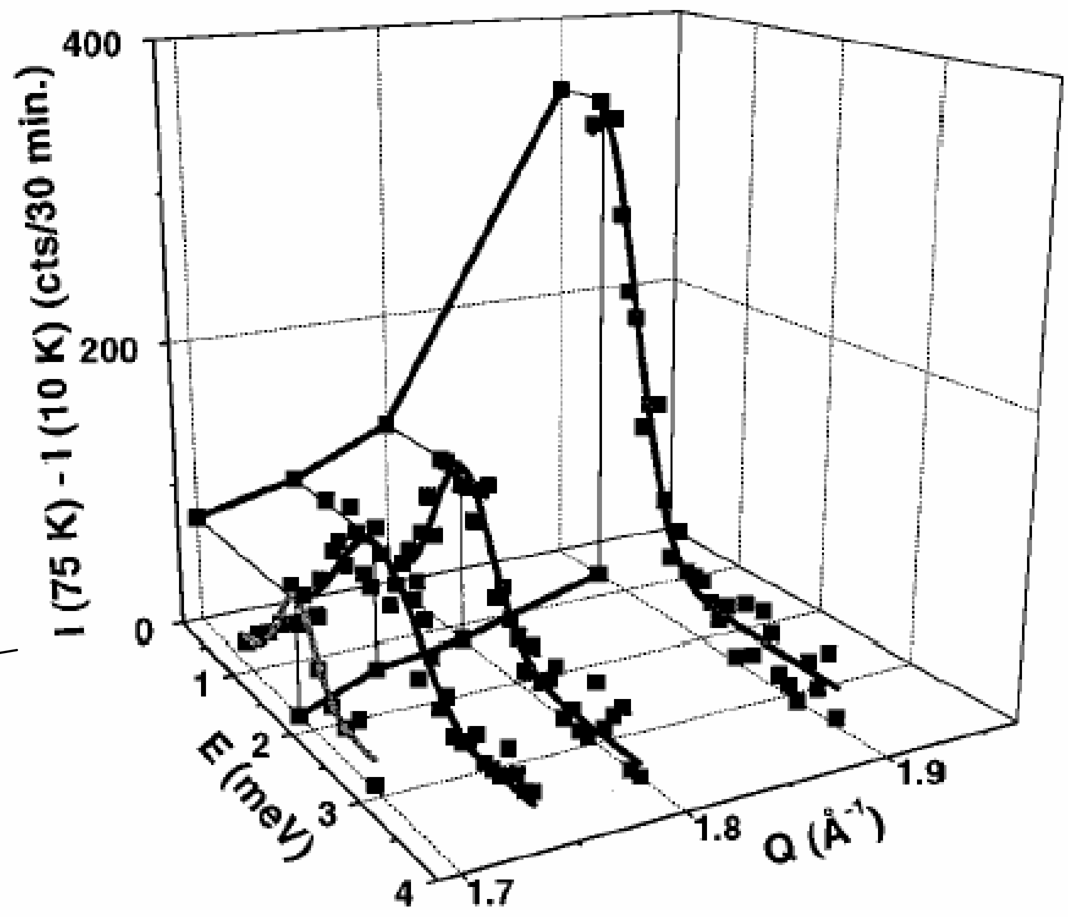




B. Lake *et al*., Nature Mater. **4** (2005) 329

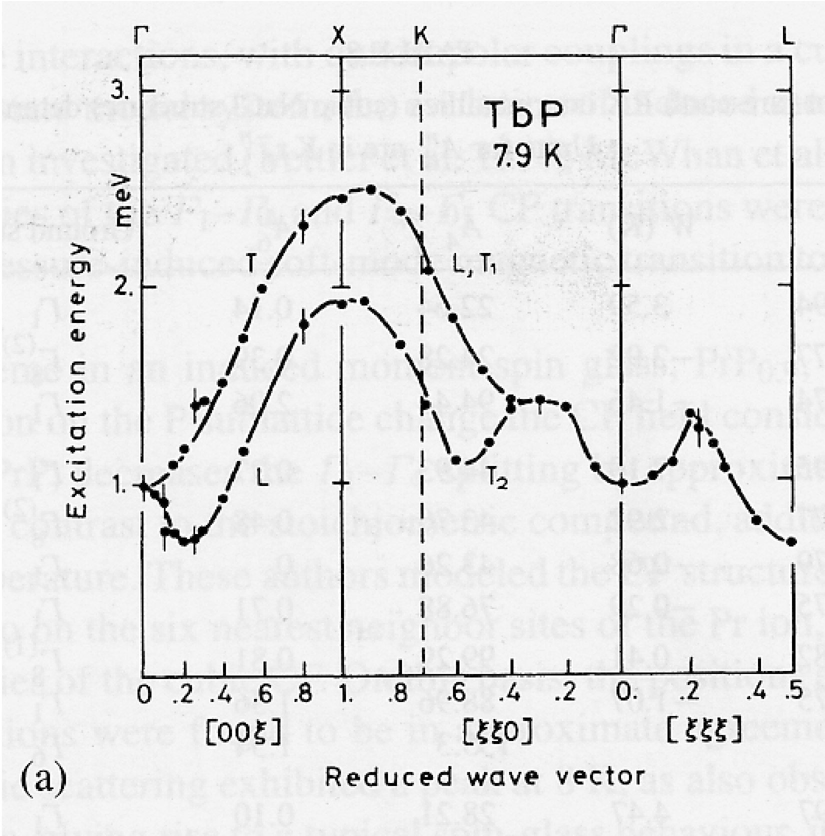
Spin-waves in a multilayer of Dy/Y

Growth direction



A. Schreyer *et al.*, J. Appl. Phys. **87** (2000) 5443

Dispersive crystal fields in TbP



A. Loidl *et al*., *CEF effecits in f-electon magnetism* (1982) eds. R.P. Guertin *et al*., Plenum New York.

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

Other magnetic Hamiltonians

Model magnetic systems (one, two and three dimensions)
Superconductivity
Giant and colossal magnetoresistance
Quantum magnetic fluctuations
Heavy fermion materials
Overdamped excitations in amorphous materials
Multiple magnon scattering
Slow relaxation in spin glasses
Fluctuations in Fractals and percolation theory
etc. etc. etc.

Neutron scattering and magnetic susceptibility

The magnetization is a time-dependent quantity.
It can be presented as a susceptibility

$$\mathbf{M}(t) \propto \chi(\omega) H_0 e^{-i\omega t} + \chi^*(\omega) H_0^* e^{i\omega t}$$

where the susceptibility is complex

$$\chi(\omega) = \chi'(\omega) + i\chi''(\omega)$$

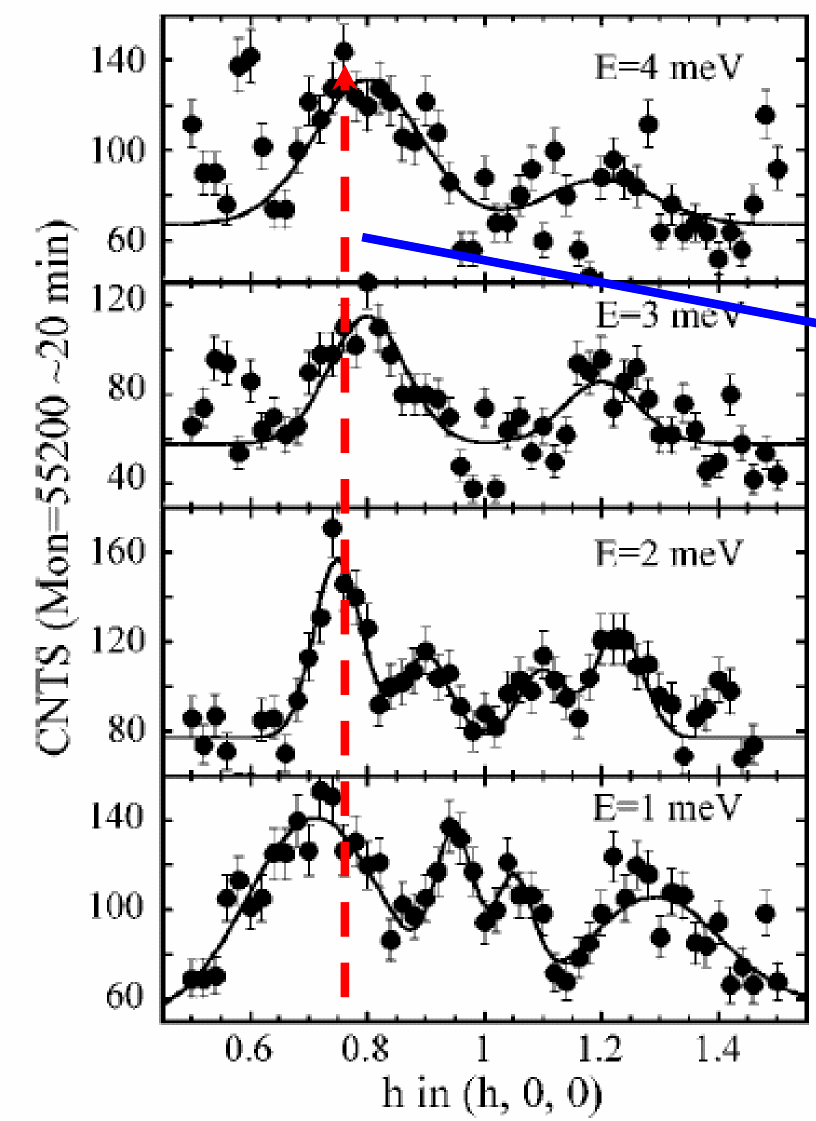
We have already seen that the neutron cross section is related to the magnetization.
It must therefore be related to the susceptibility:

$$\frac{d^2\sigma}{d\Omega dE} \propto \frac{k'}{k} \sum_{\alpha} \frac{\chi''(\mathbf{Q},\omega)}{\left(1\text{-}e^{-\beta\hbar\omega}\right)}$$

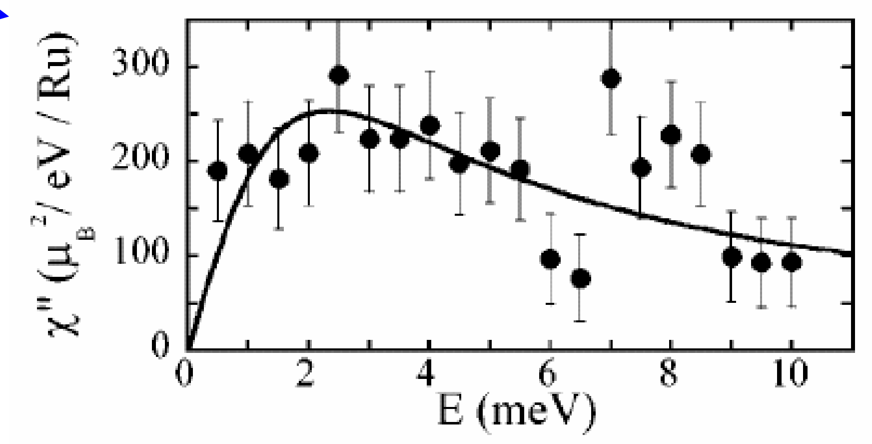
$\chi(\mathbf{Q},\omega)$ is a *generalized* susceptibility
Neutron scattering can therefore be directly related to magnetometry measurements.

Spin excitations in $Sr_3Ru_2O_7$

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



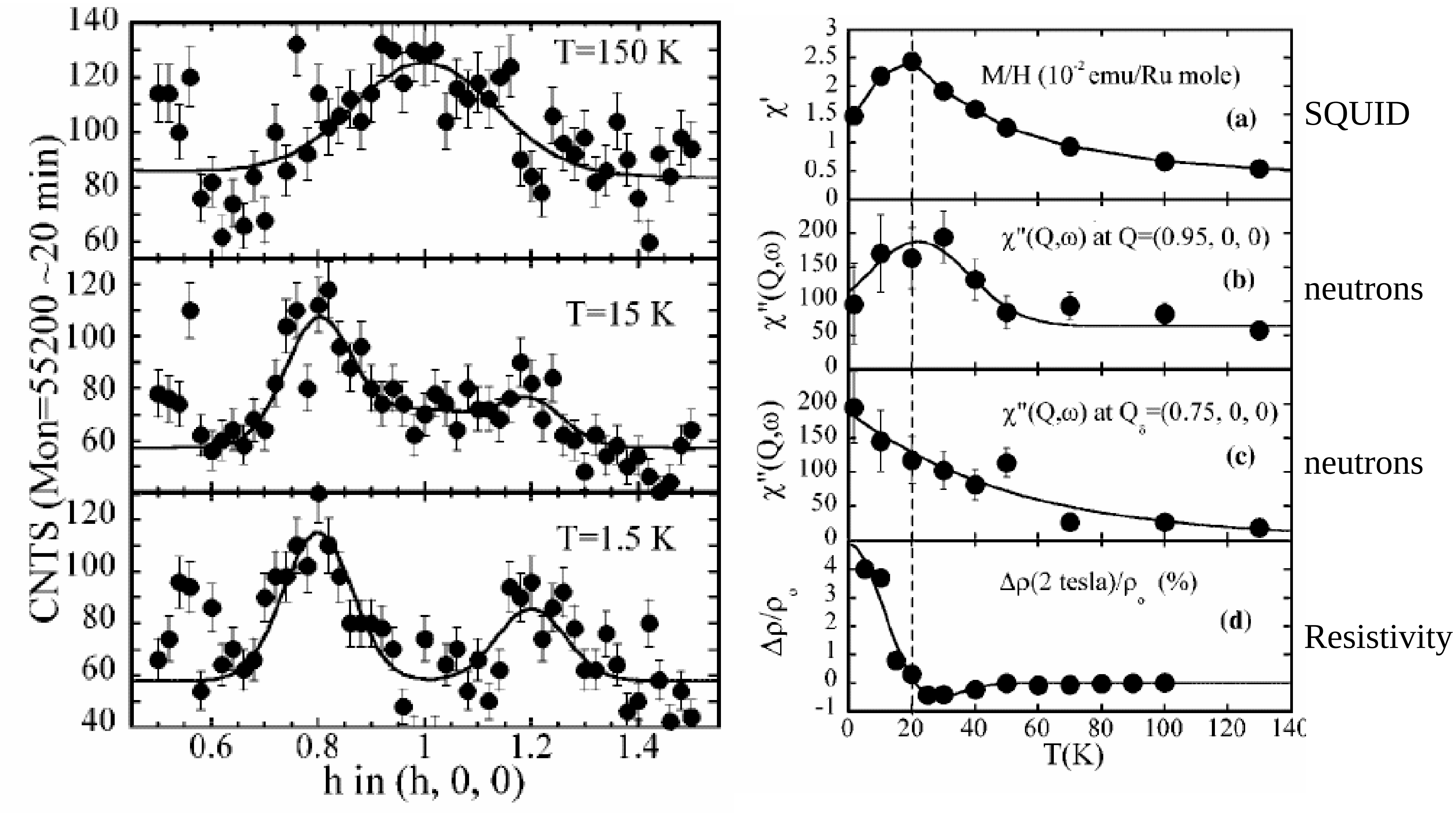
Inelastic neutron scattering at 1.5K



Susceptibility

L. Capogna *et al*., Phys. Rev. B. **87** (1998) 143

Temperature dependence of the spin excitations in $Sr_3Ru_2O_7$



L. Capogna *et al.*, Phys. Rev. B **87** (1998) 143

Neutrons and bulk susceptibility

If neutrons measure the generalized susceptibility,
it must be possible to convert between bulk susceptibility measurements and neutron cross-sections.
This can be done using the Kramers-Krönig relation:

$$\int d\omega \frac{\chi''(\mathbf{Q},\omega)}{\omega} = \pi\chi'(\mathbf{Q},\omega)$$

Integrate the neutron scattering over all energies:

$$\frac{d\sigma}{d\Omega} = \int \hbar d\omega \left(\frac{d^2\sigma}{d\Omega d\omega}\right)$$

$$\propto k_B T \sum_\alpha \chi'_{\alpha\alpha}(\mathbf{Q},0)$$

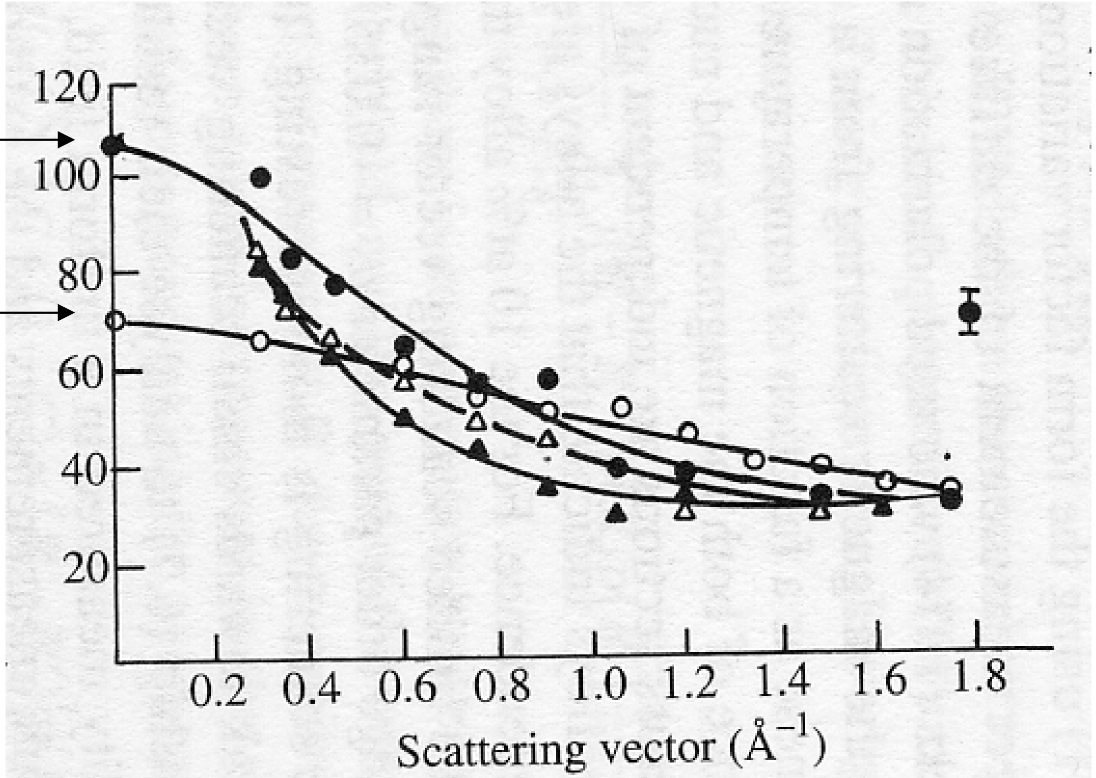
Bulk susceptibility measures the real part of the susceptibility.
Bulk susceptibility averages over all the sample, which is equivalent to **Q** = 0.

$$i.e. \ \frac{d\sigma}{d\Omega}(\mathbf{Q}=0) \propto k_B T \chi'$$

Bulk susceptibility can be put as a point on a neutron scattering plot!

Diffuse scattering in $Cu_{0.95}Mn_{0.05}$

Points taken from bulk susceptibility



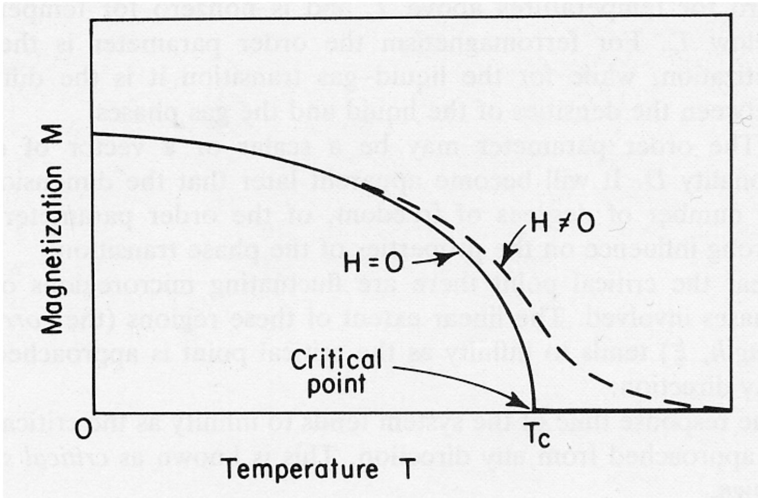
● 293 K
○ 35 K
△ 15 K
▲ 4.2 K

N. Ahmed and T. J. Hicks, Solid State Comm. **15** (1974) 415

T. J. Hicks, *Magnetism in disorder*, 1995, Clarendon, Oxford

Magnetic phase transitions

The magnetic structure of a simple ferromagnet as a function of temperature



M. F. Collins, *Magnetic critical scattering*, 1989, Oxford University, Oxford

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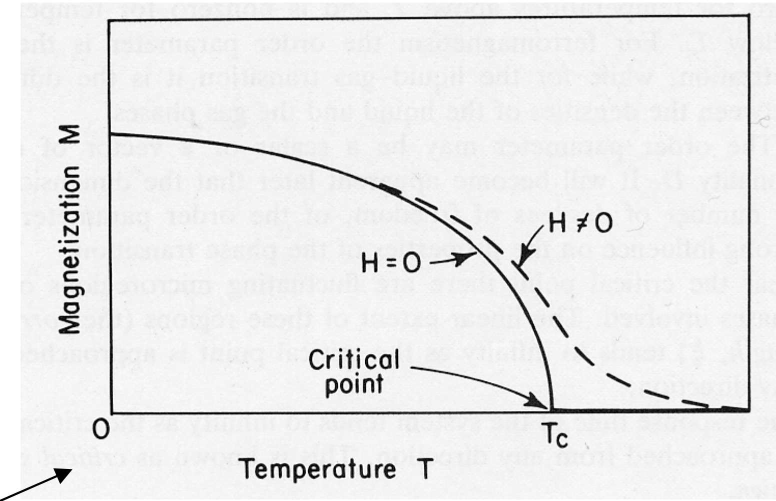
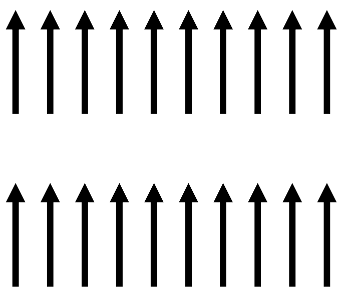


Time-average
$(\hbar\omega = 0)$

Snapshot

$(\int_{-\infty}^{\infty}\hbar\omega)$



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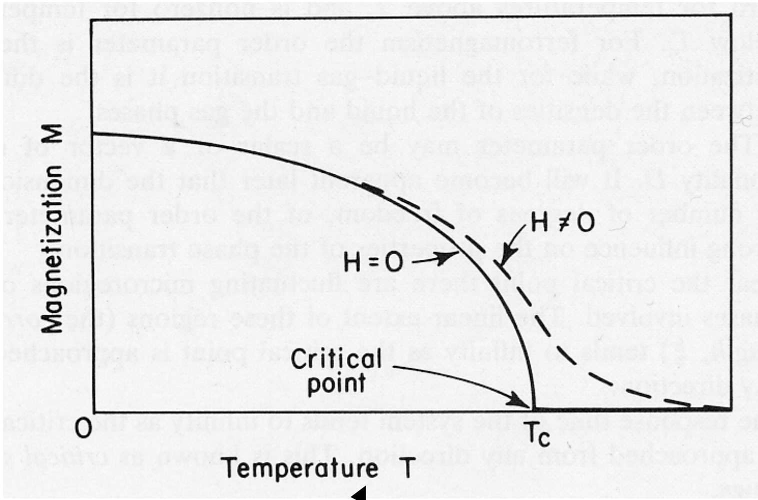
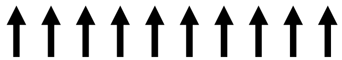


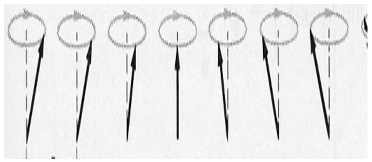
Time-average

($\hbar\omega = 0$)



Snapshot

($\int_{-\infty}^{\infty} \hbar\omega$)



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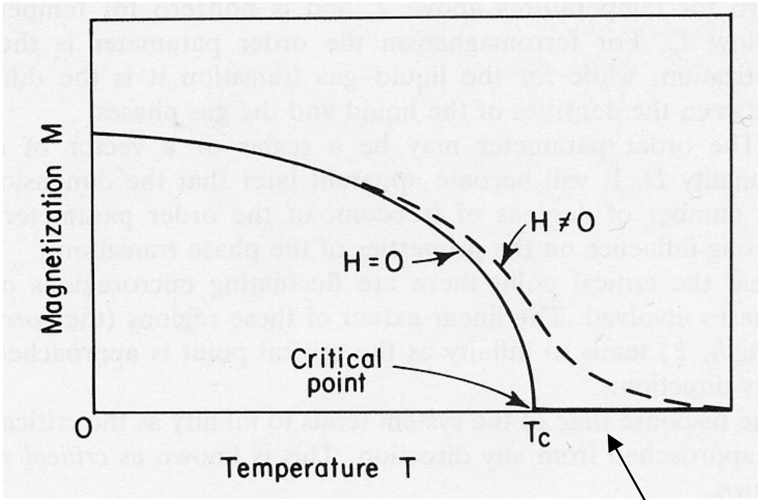

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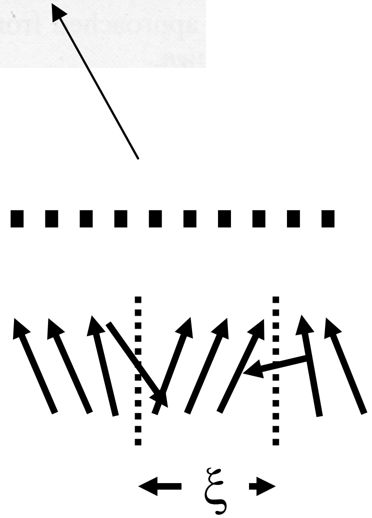

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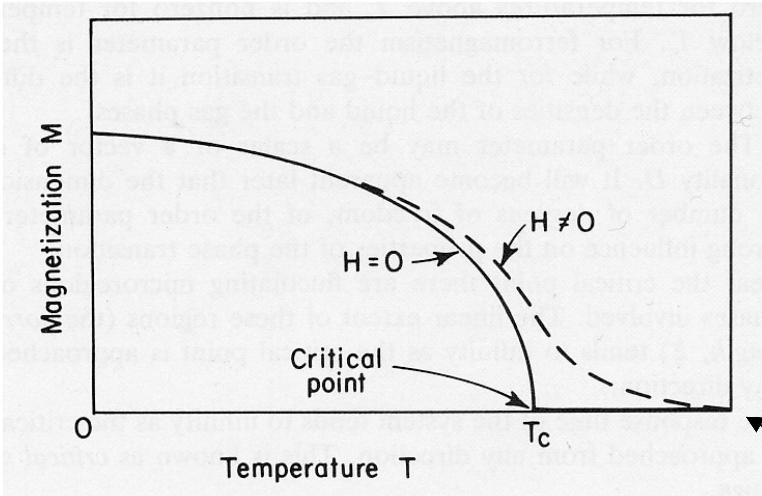

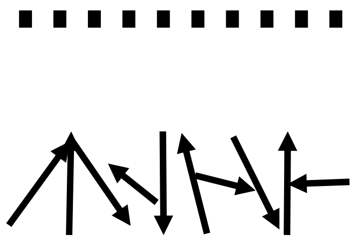

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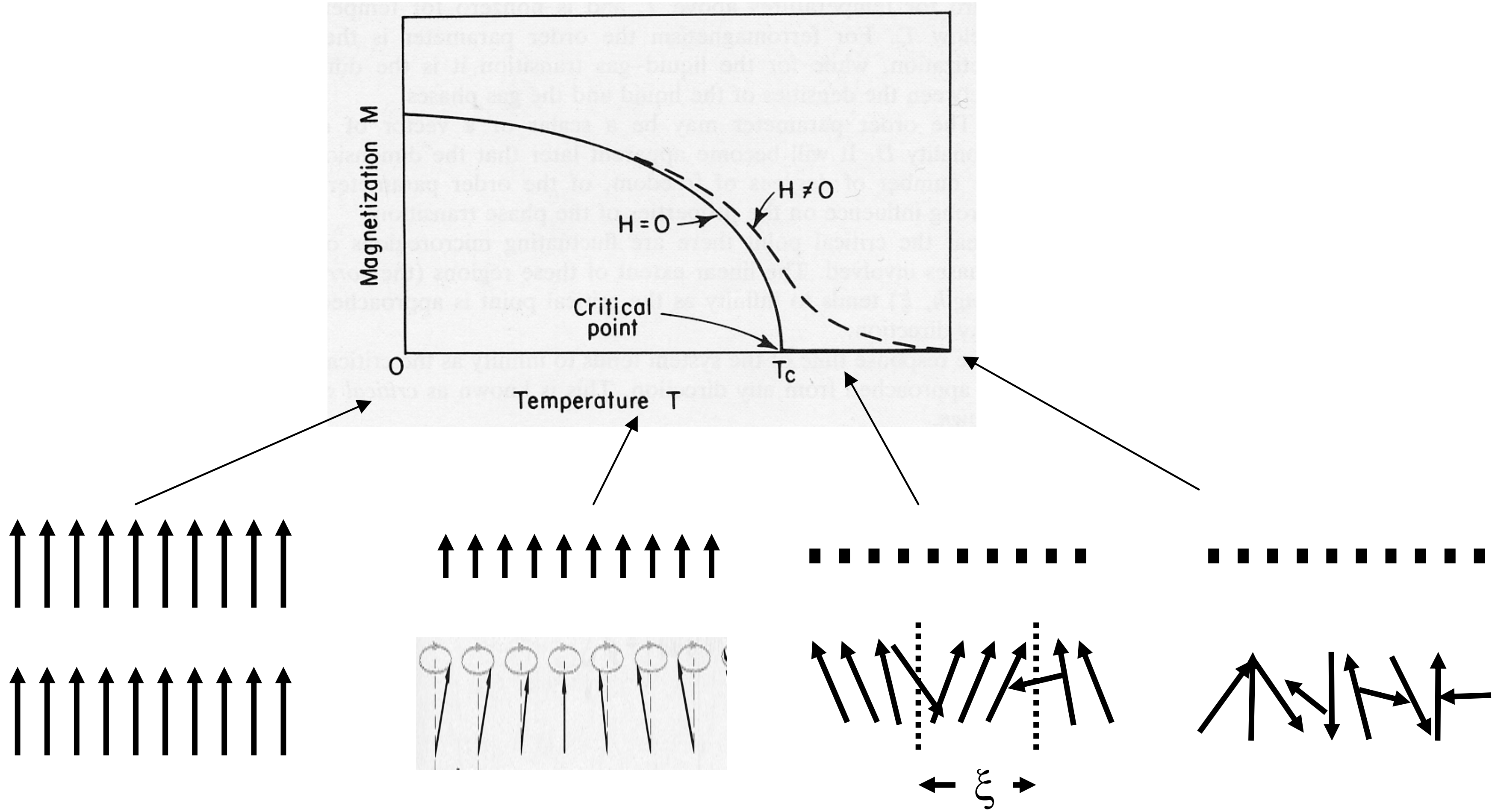


Time-average $(\hbar\omega = 0)$

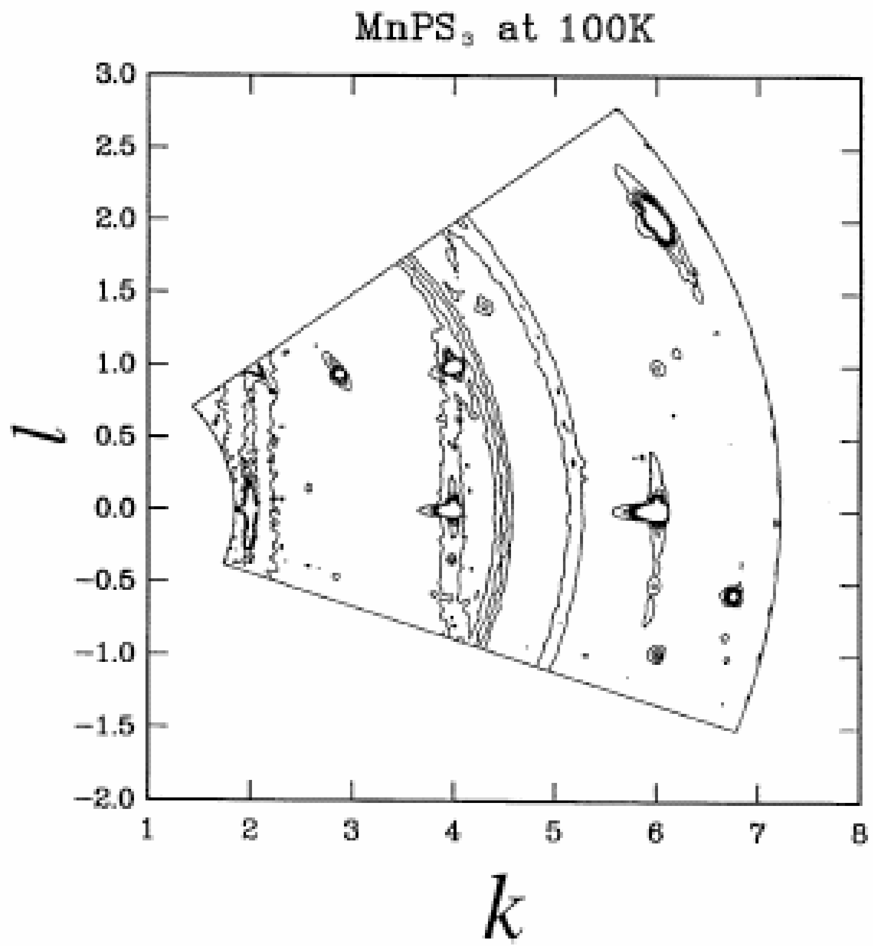
Snapshot $\left(\int_{-\infty}^{\infty} \hbar\omega\right)$

Renormalization theory and universality:

$$M(T) \propto (T-T_N)^{-\beta} \qquad \xi(T) \propto (T-T_N)^{-\nu}$$

M. F. Collins, *Magnetic critical scattering*, 1989, Oxford University, Oxford

Magnetic critical scattering in $MnPS_3$



A.R. Wildes *et al*., J. Magn. Magn. Mater. **87** (1998) 143

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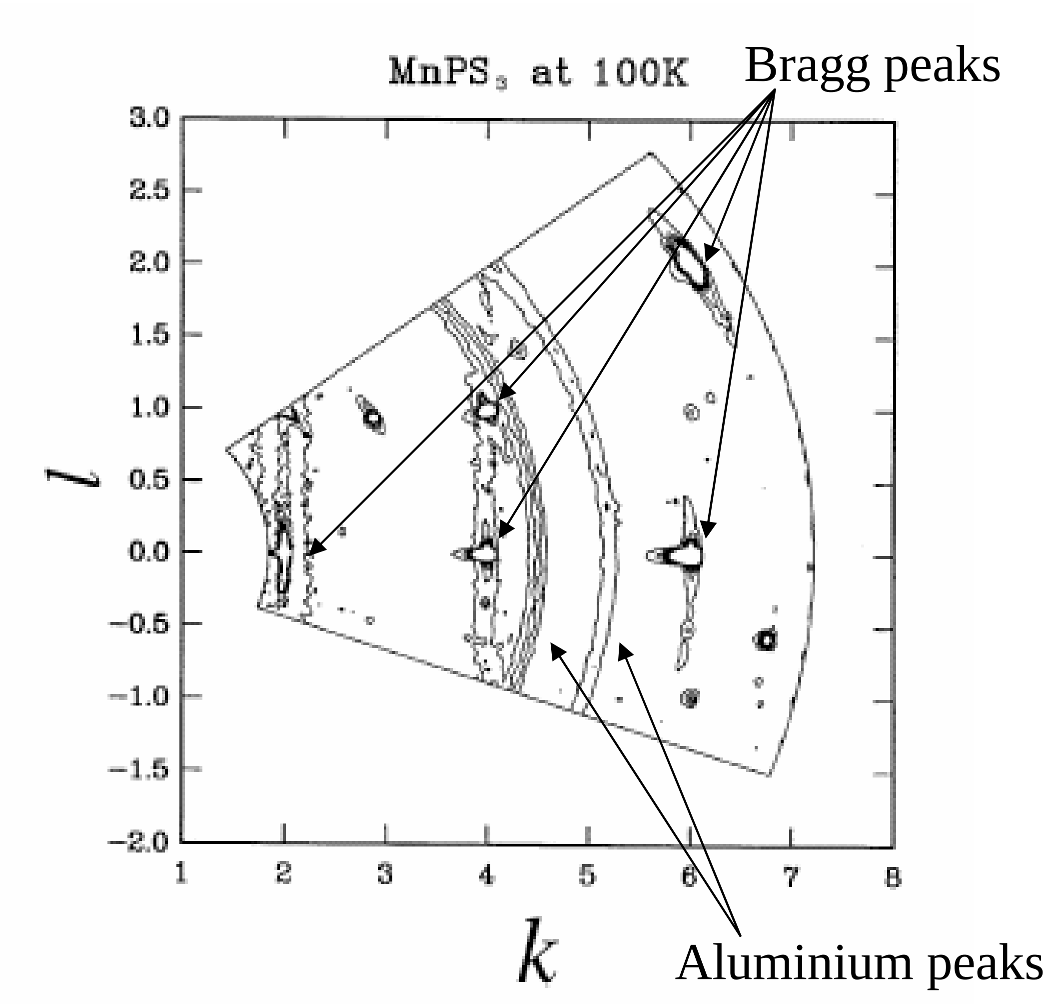

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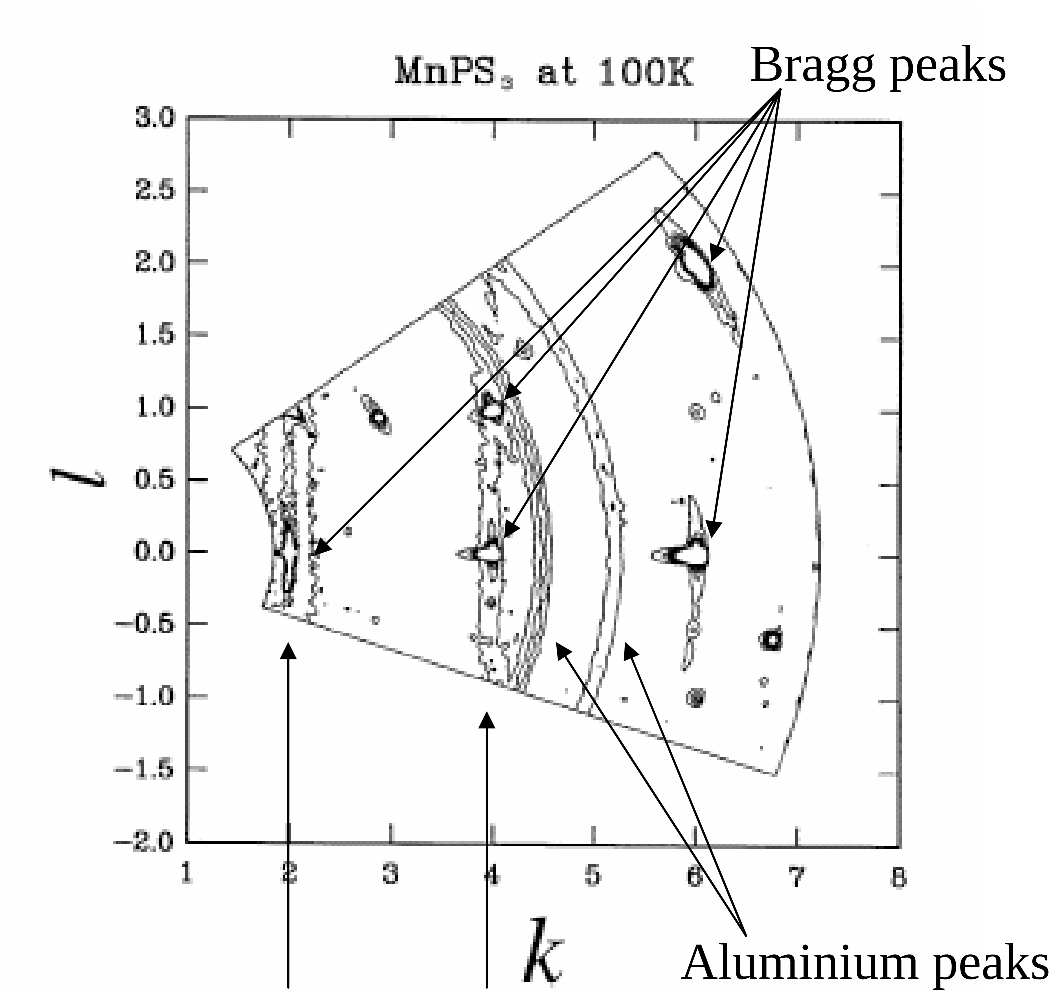

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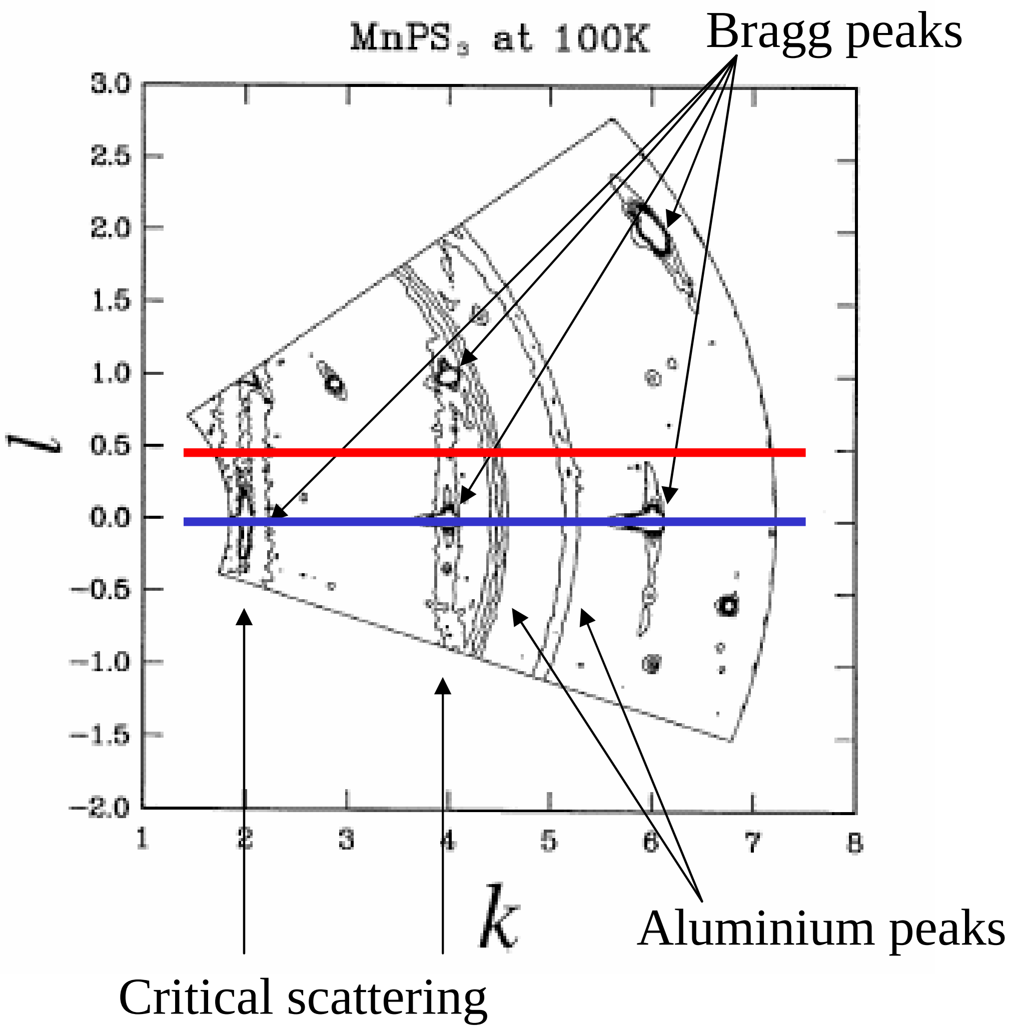
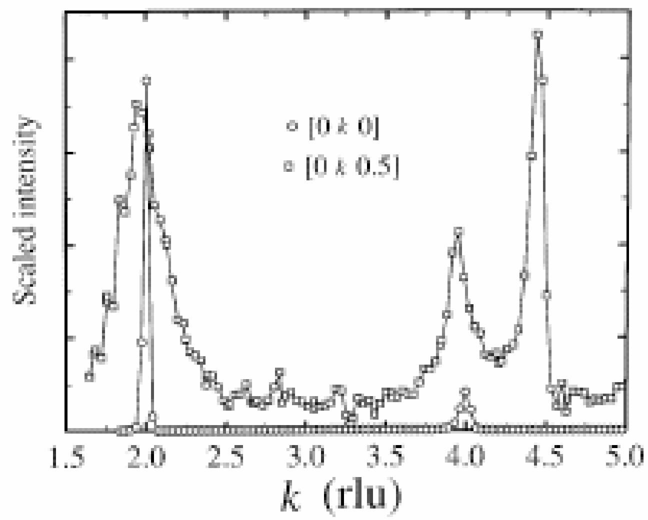




The widths of the peaks show that the rods are scattering from critical fluctuations.

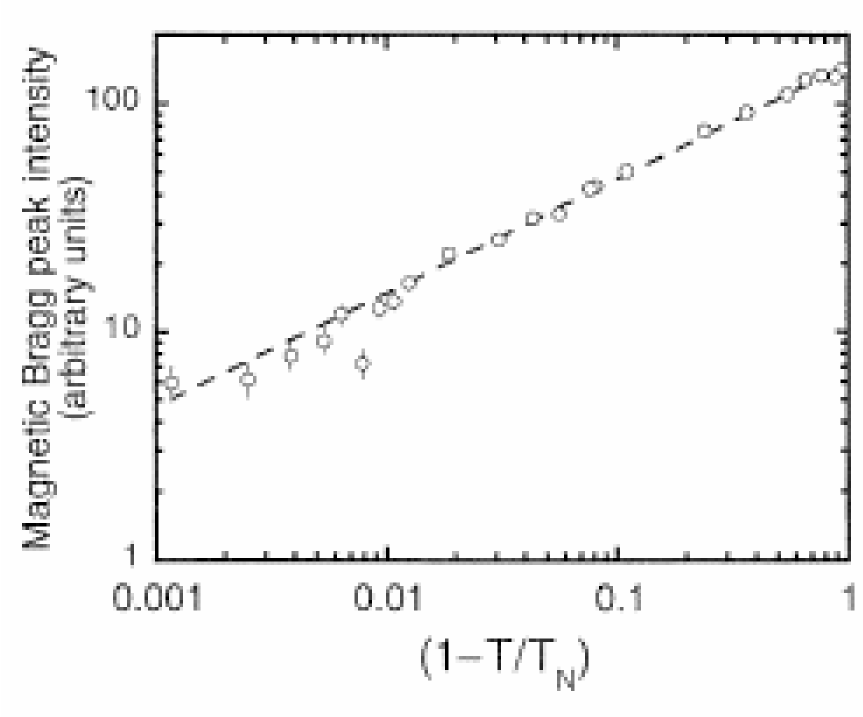
A.R. Wildes *et al*., J. Magn. Magn. Mater. **87** (1998) 143

The magnetization of $MnPS_3$

The intensity of magnetic Bragg scattering, which is *elastic*, gives the Magnetization

$$\frac{d\sigma_{magnetic}}{d\Omega} = \left\langle \mathbf{M}^*_\perp(\mathbf{Q}) \right\rangle \left\langle \mathbf{M}_\perp(\mathbf{Q}) \right\rangle$$

$$\propto M^2$$

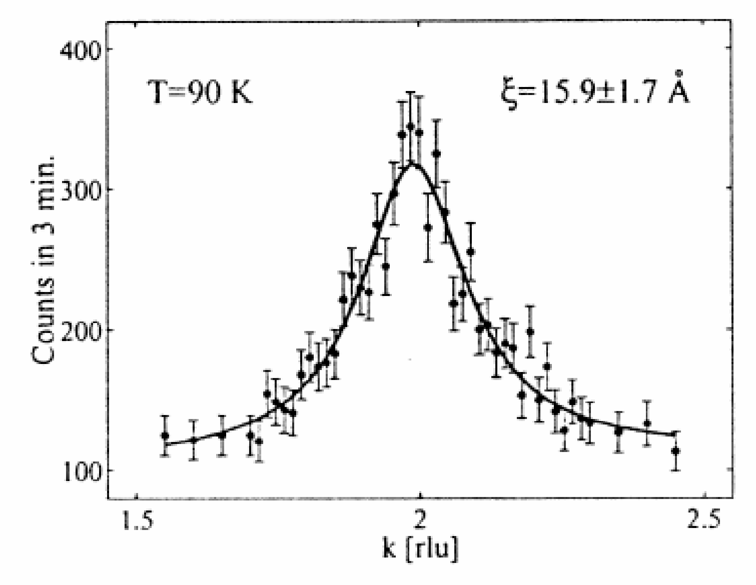


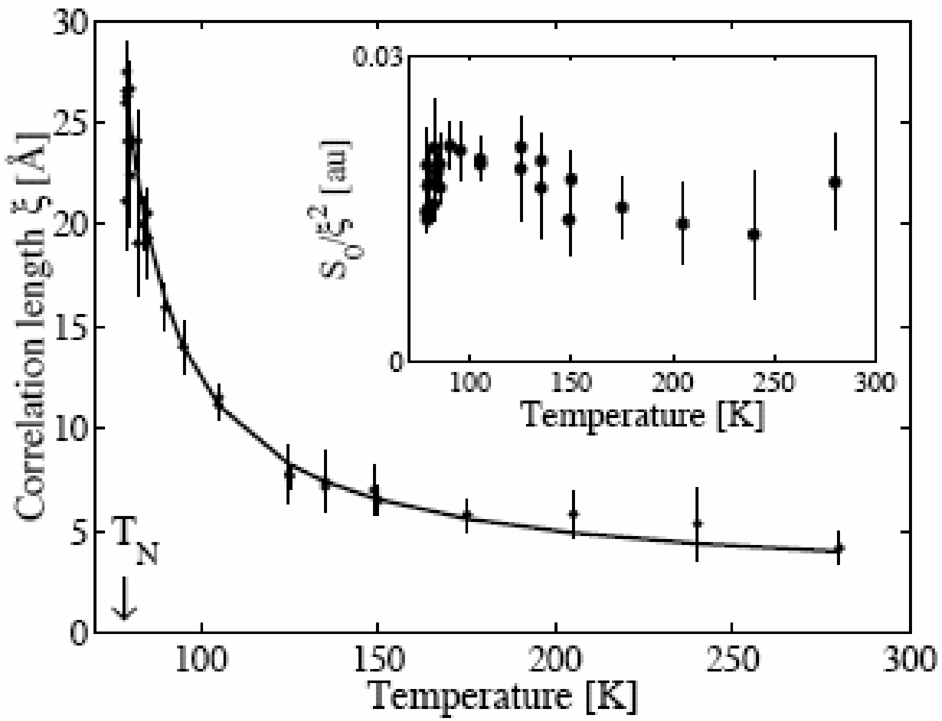
$$T < T_N, \;\; M \sim (T_N - T)^{-0.25}$$

A.R. Wildes *et al*., J. Phys.: Condens. Matter **10** (1998) 6417

The correlation length of $MnPS_3$

The width of magnetic quasielastic scattering, integrated over energy, gives the magnetic correlation length





$$T > T_N, \quad \xi \sim e^{b\left(T_{KT}/(T-T_{KT})\right)^{1/2}}$$

H. M. Rønnow *et al.*, Physica B **2760278** (2000) 676

Reflectivity and glancing incidence reflection

If the neutron has a *plane wave function*, if the interaction is *weak*, then the wave equation can be solved using first order perturbation theory, i.e. Fermi's Golden Rule

The two assumptions form the first *Born approximation*

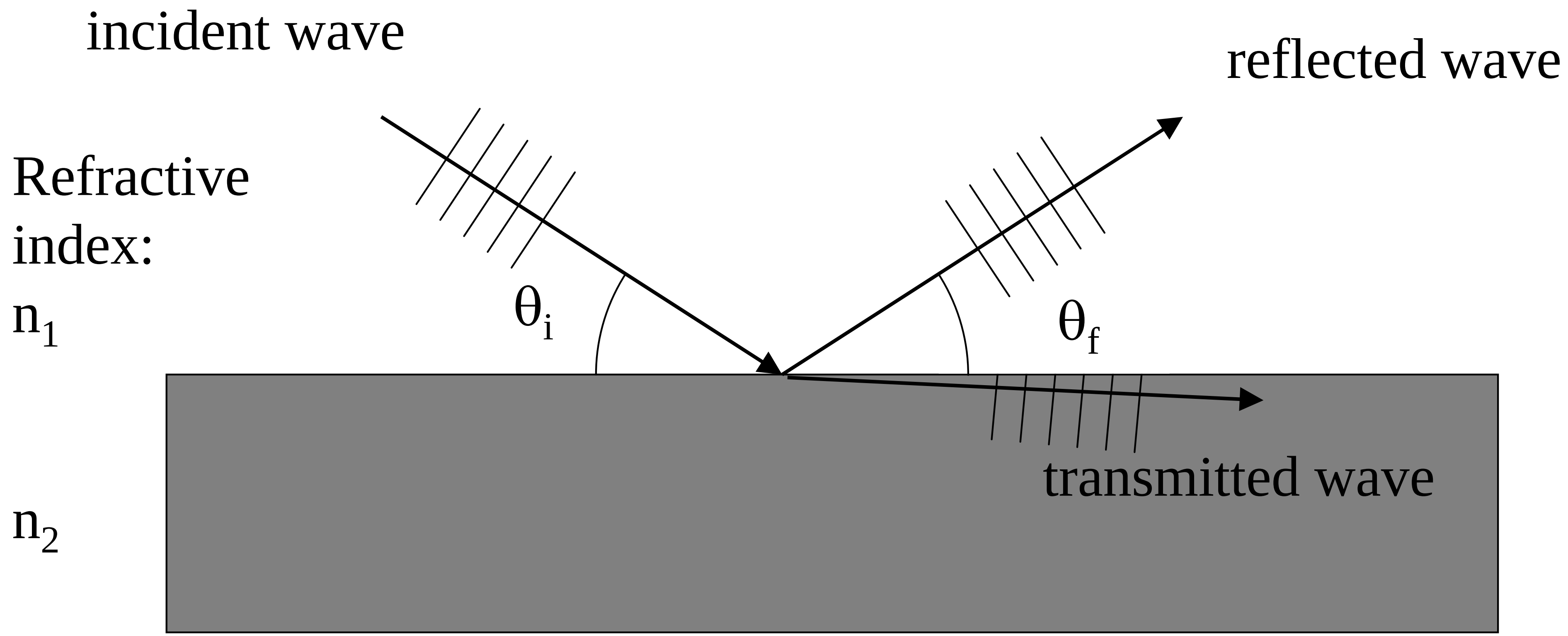
The assumptions do not hold when the sample acts like a neutron *mirror*, which it does when measuring the scattering at *glancing angles*.

Then, the interaction is *strong*, and another, another theory needs to be developed

This theory, known as a *dynamical* theory, is in development for neutrons and x-rays.

Reflectivity = Final intensity/Incident intensity

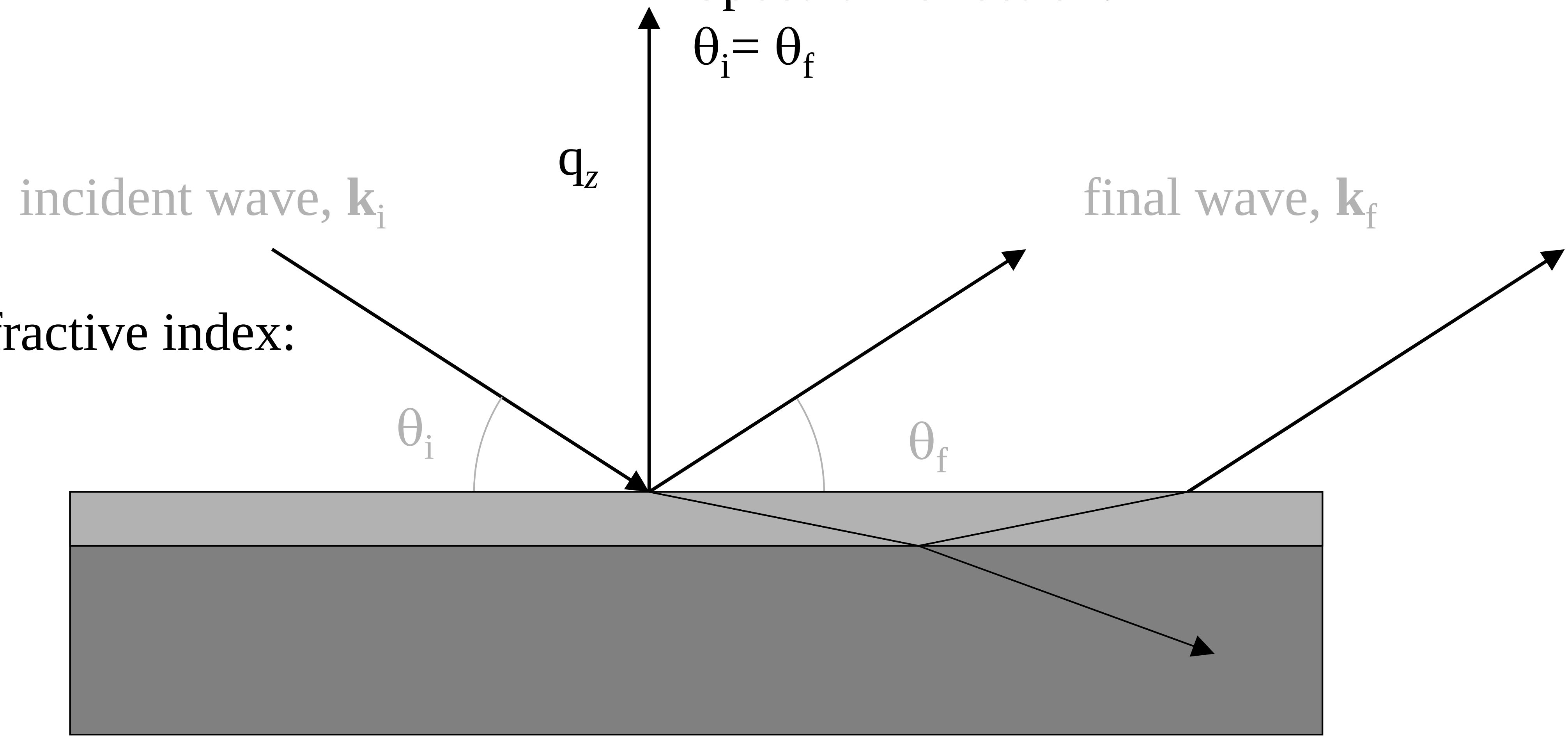
$$R = \frac{I_f}{I_i}$$



Specular Reflectivity

Specular reflection:

$\theta_i = \theta_f$



Refractive index:

n_1

n_2

n_3

$$n_j^2 = 1 - \frac{\lambda^2}{\pi}\rho_j - i\frac{\lambda}{4\pi}\mu_j$$

ρ = the scattering length density

μ = the linear absorption coefficient

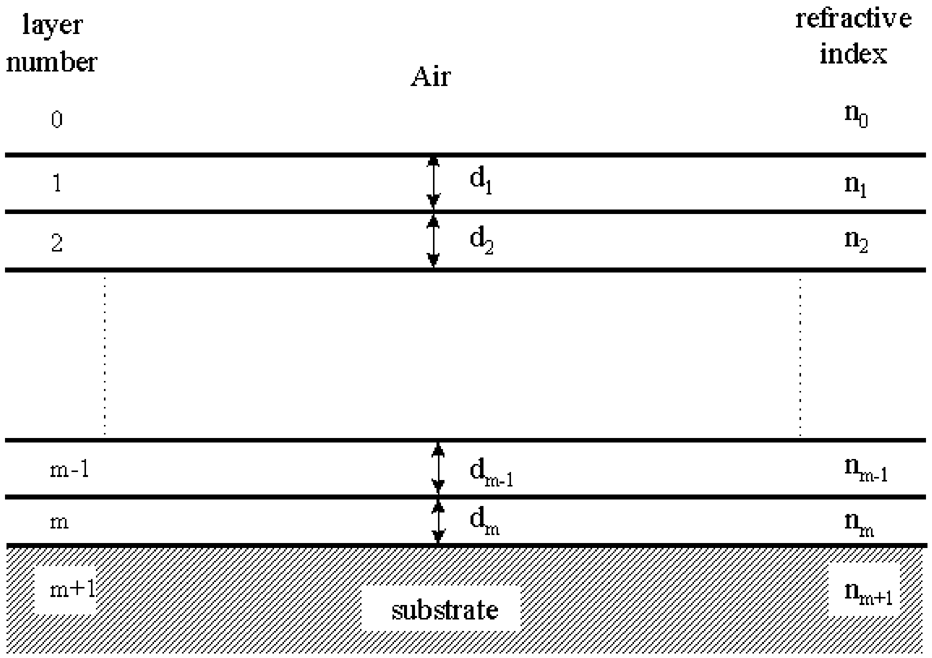
Modeling the Data

$$n_j^2 = 1 - \frac{\lambda^2}{\pi}\rho_j - i\frac{\lambda}{4\pi}\mu_j$$

$$\rho_j = N_j \left|\hat{V}_j\right|$$

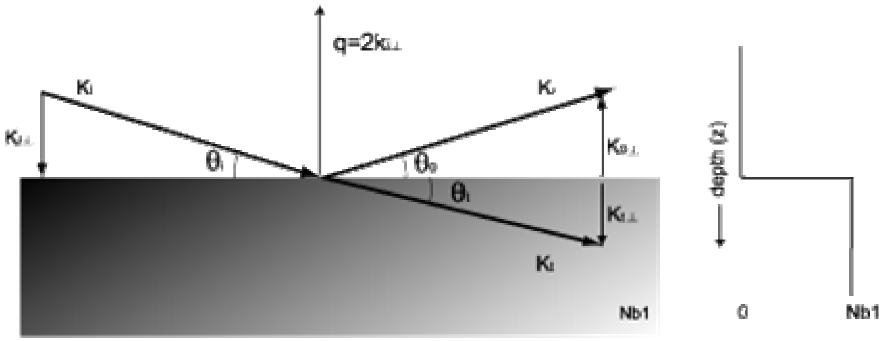
$$\left|\hat{V}_j\right| = \left|b_j + B_j \hat{\mathbf{I}}_j \cdot \hat{\boldsymbol{\sigma}} - \gamma\mu_N \mathbf{B}_j \cdot \hat{\boldsymbol{\sigma}}\right|$$

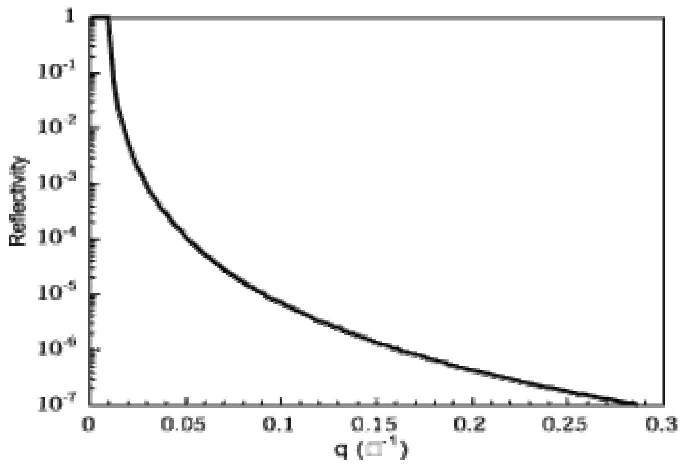
Magnetic!



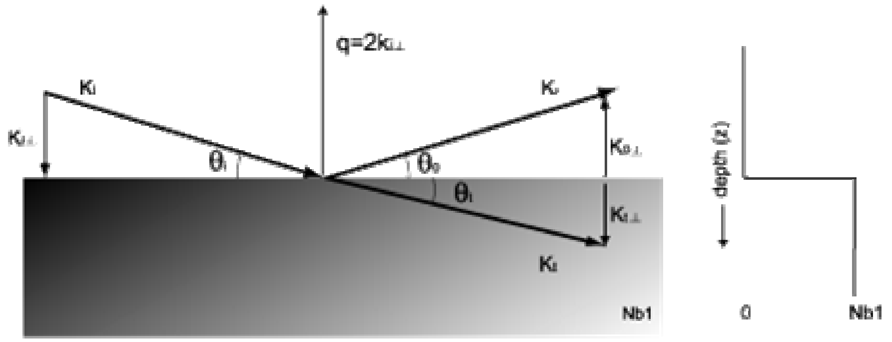
Use this to calculate the Fresnel coefficients at each interface
Arrive at an equation that can be fitted to the data.
May be done using Dynamical theory of scattering
(e.g. W. H. Zachariasen, *Theory of X-ray diffraction in crystals*, 1945, Dover, New York)

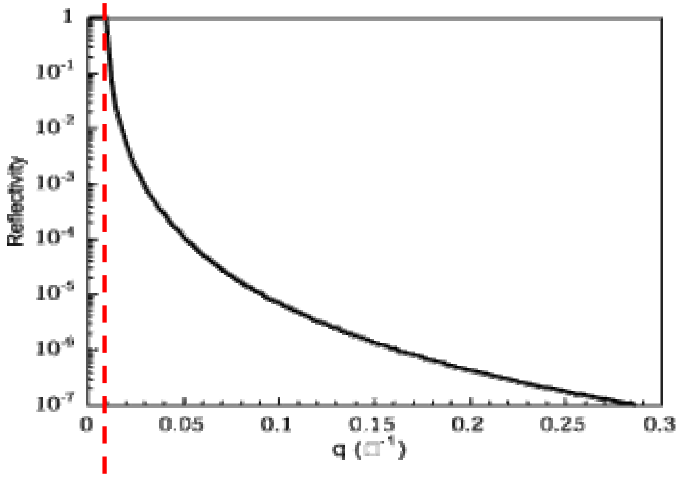
Specular Reflectivity





Specular Reflectivity

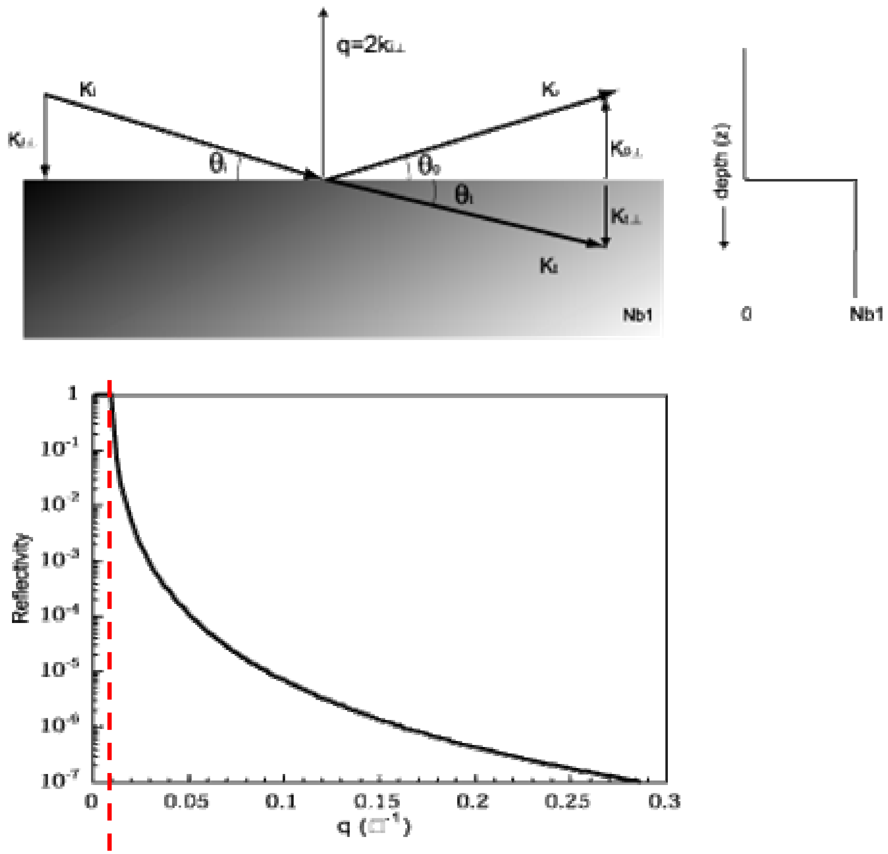


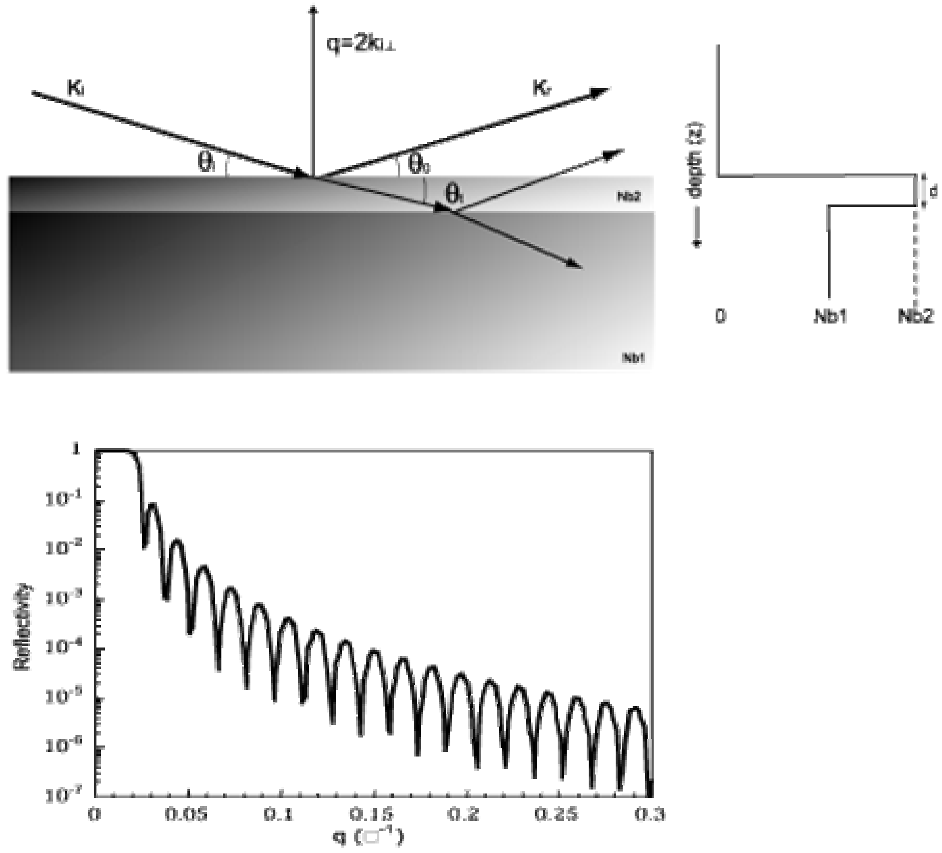


Total reflection | Reflectivity (~ q_z^{-4})

Critical edge

Specular Reflectivity

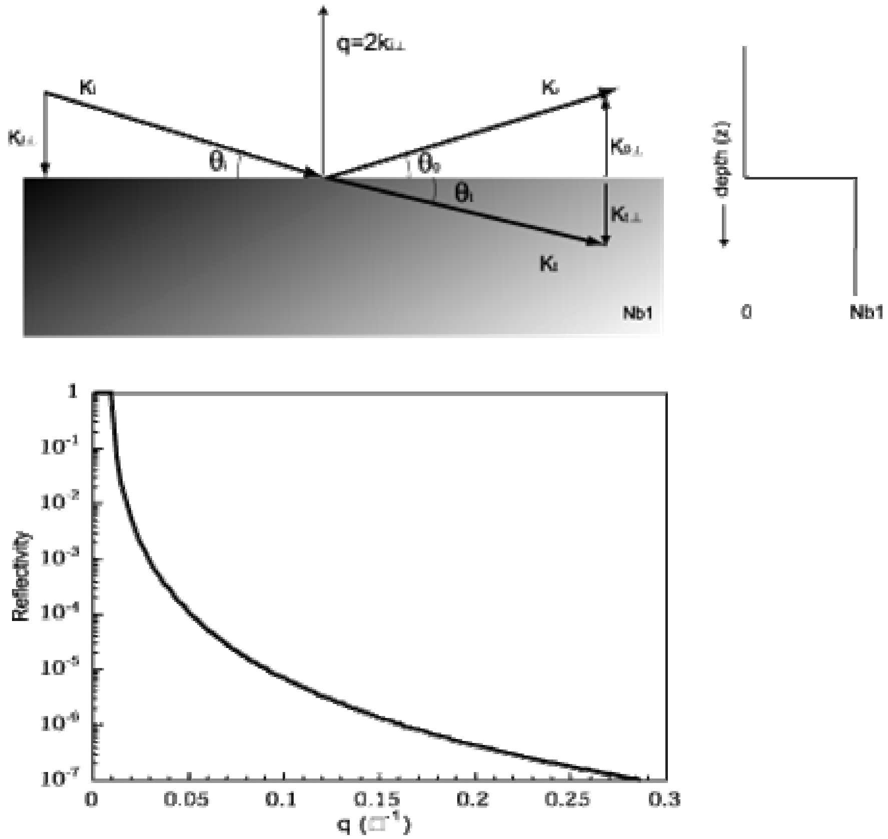


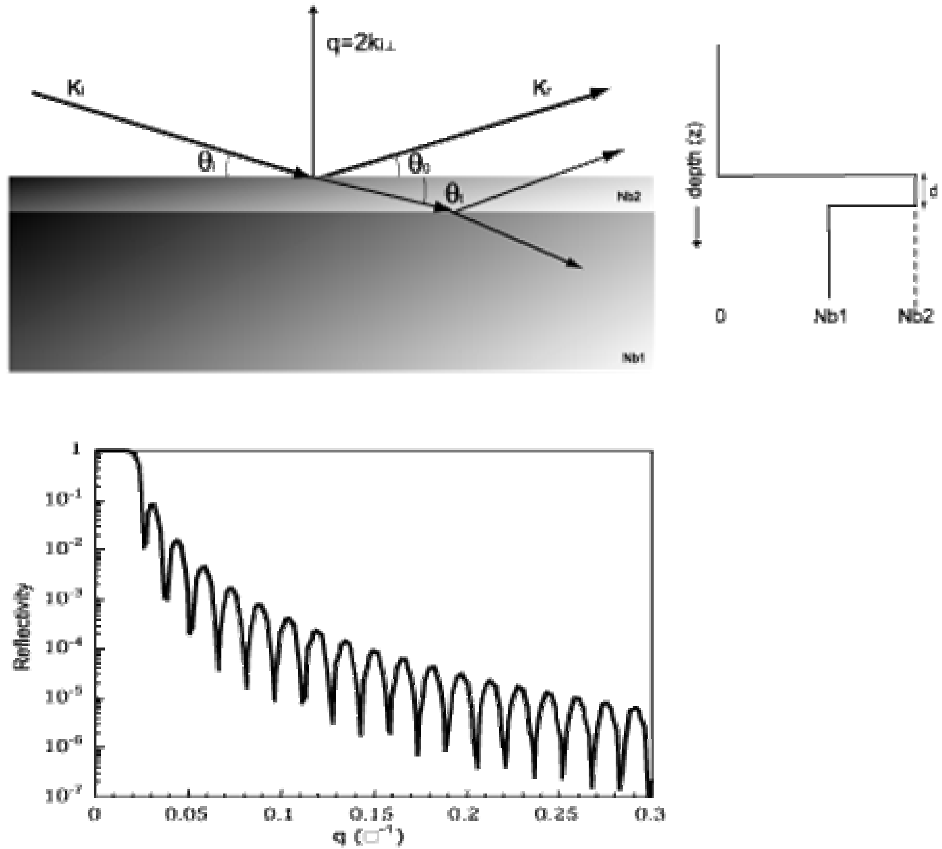


Total reflection | Reflectivity (~ q_z^{-4})

Critical edge

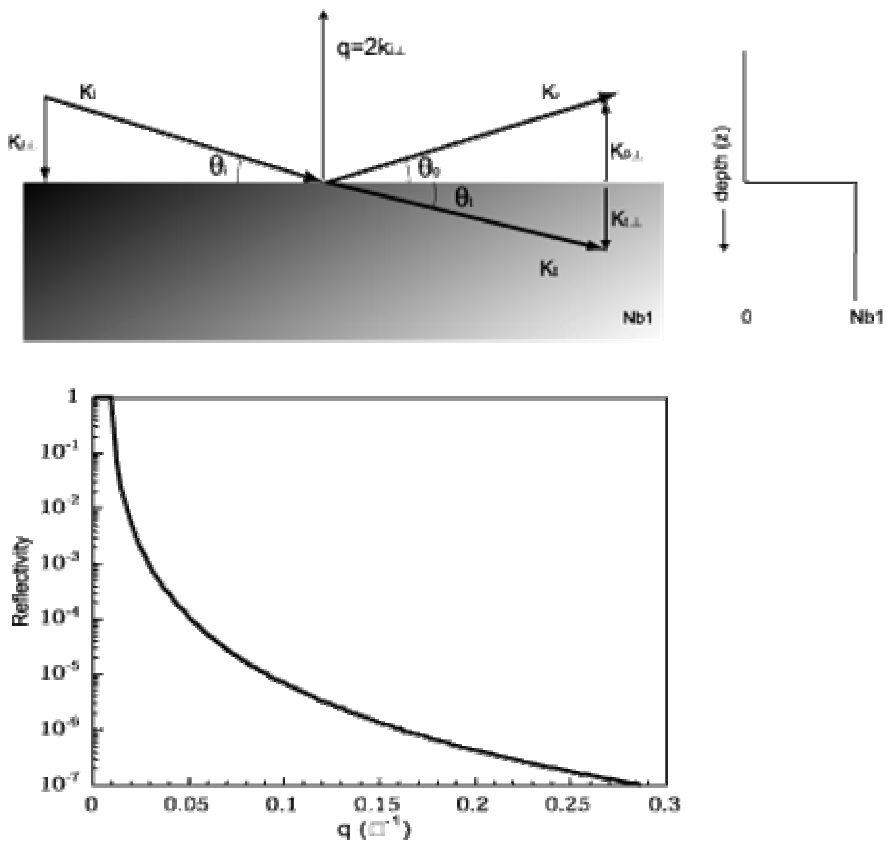
Specular Reflectivity

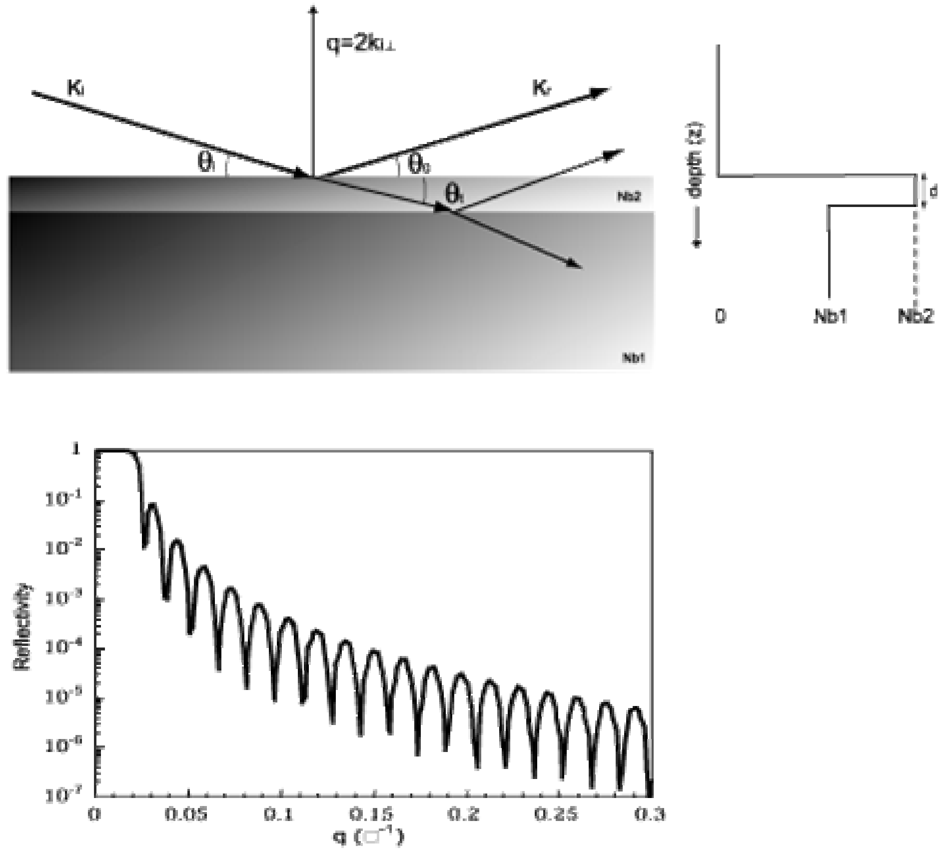




Specular reflectivity can derive the scattering length density and absorption as a function of depth

Specular Reflectivity

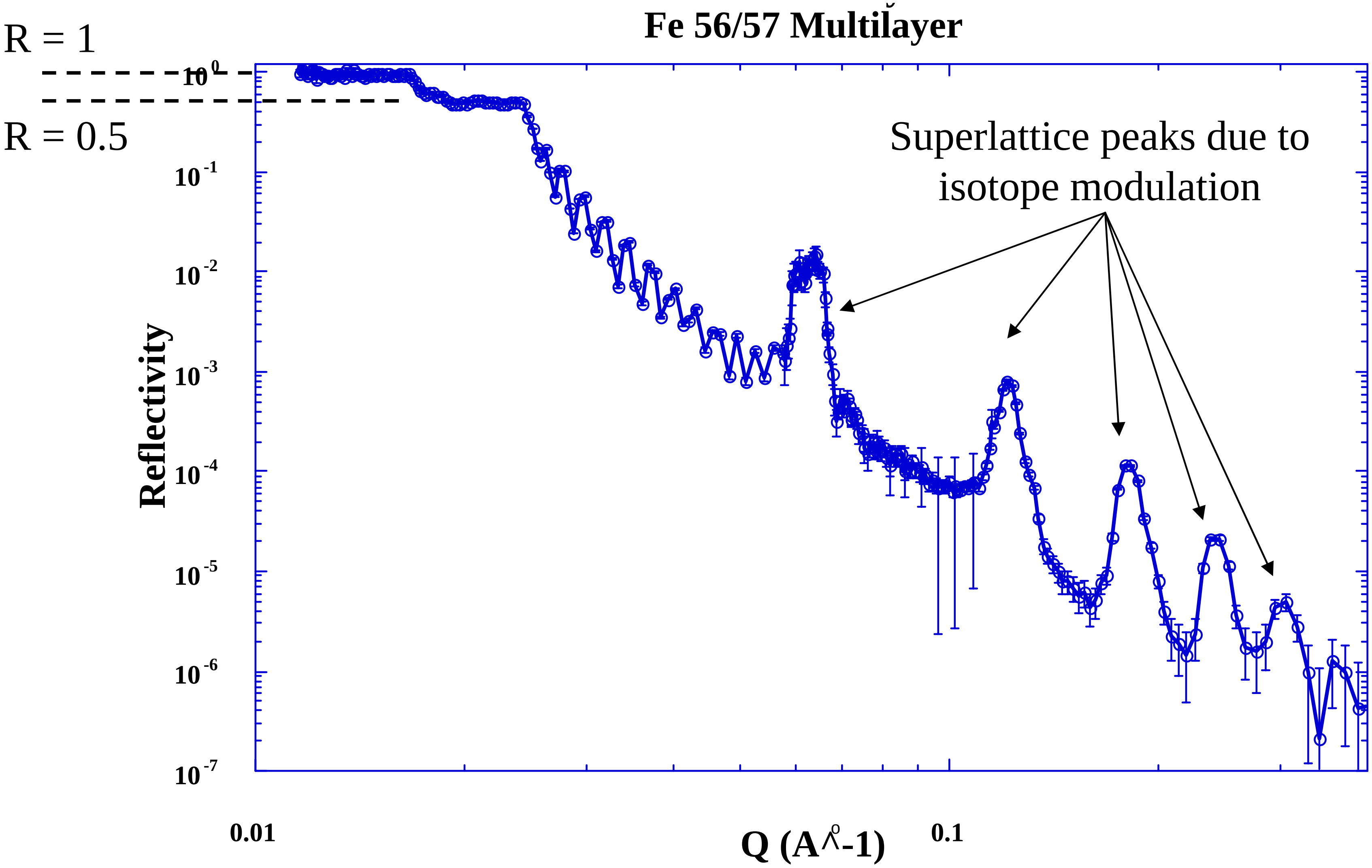




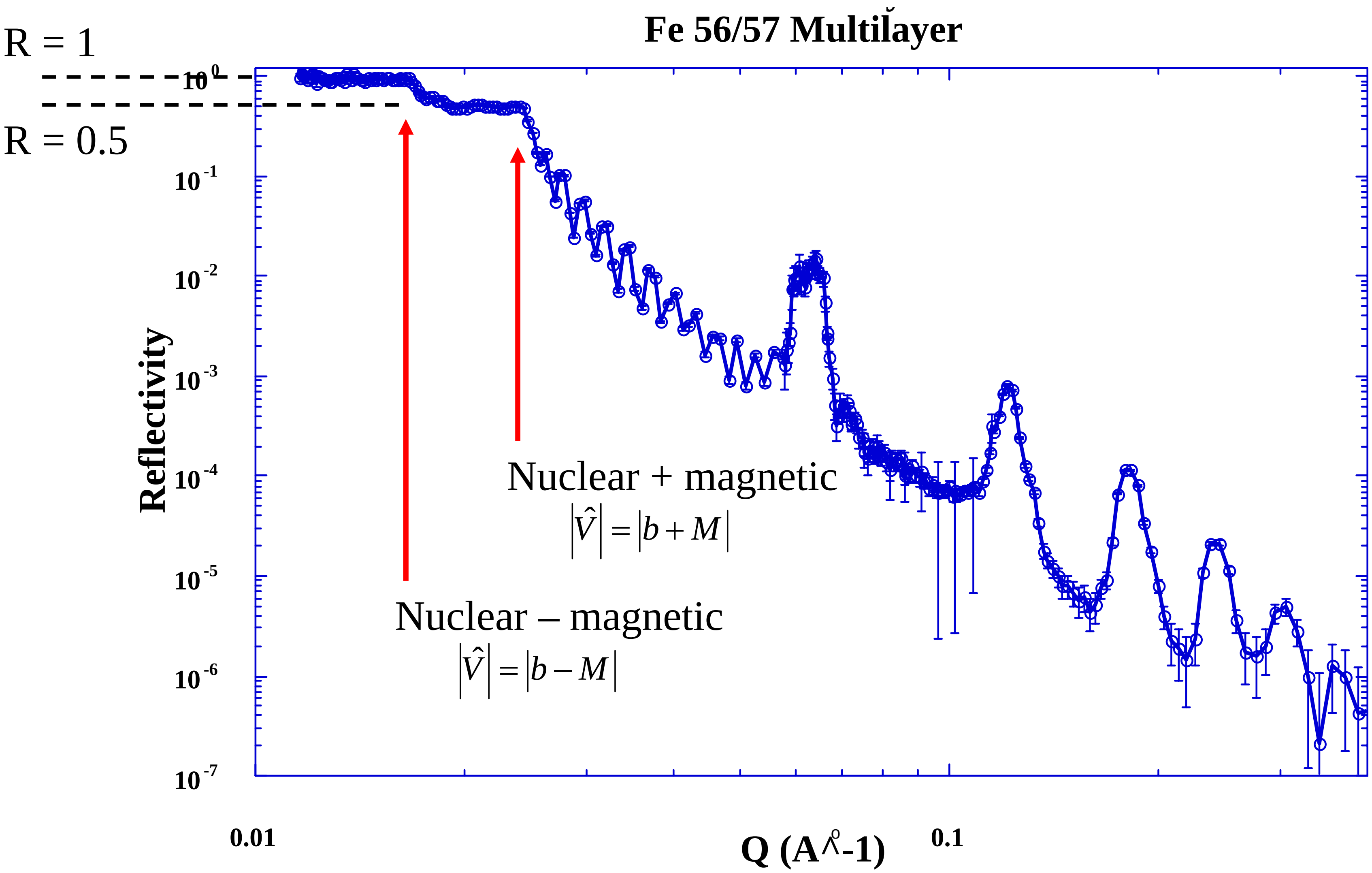
Specular reflectivity can derive the scattering length density and absorption as a function of depth

Specular reflectivity can't give information about in-plane structure

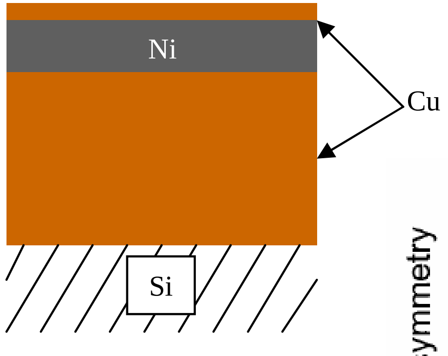
Refractive indices for magnetic materials



Refractive indices for magnetic materials

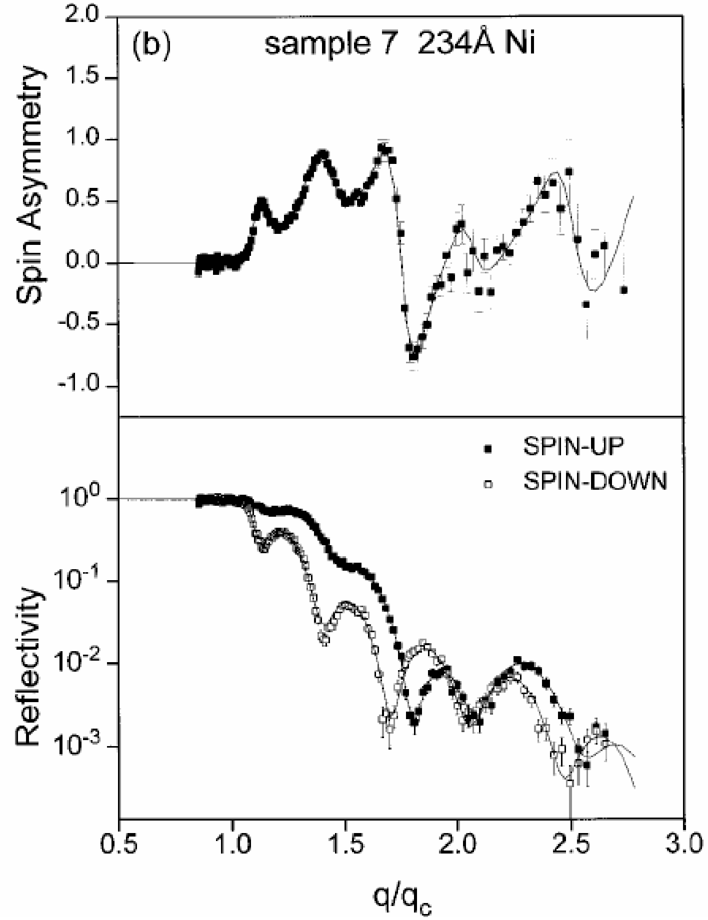


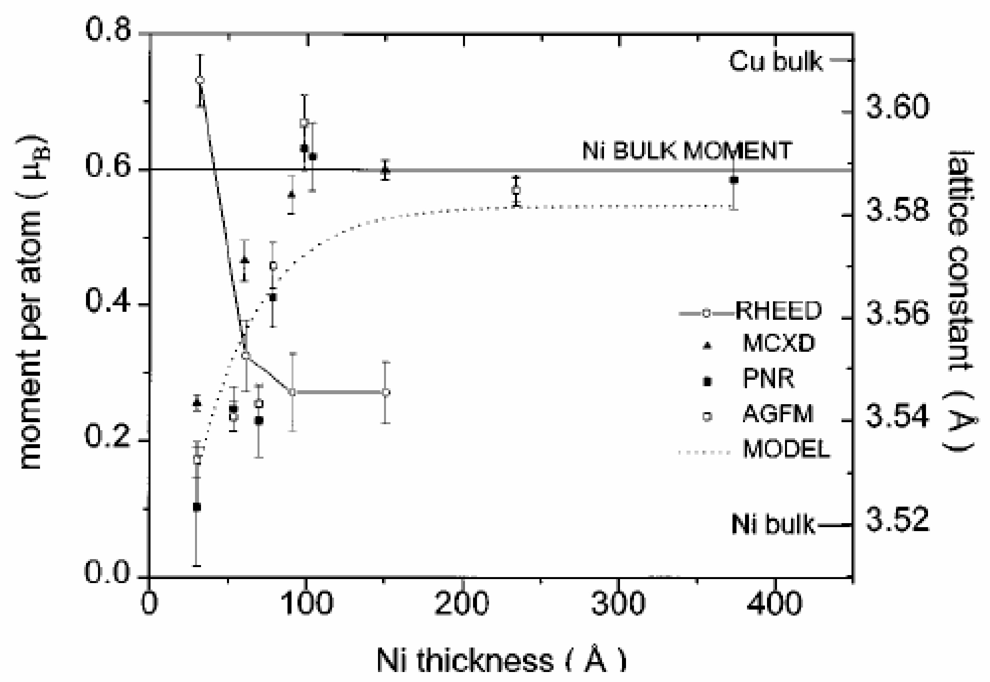
Ferromagnetic thin films, Ni in Cu



$$\text{Spin asymmetry} = \frac{R^+ - R^-}{R^+ + R^-}$$

From this, we can get the nickel moment as a function of film thickness

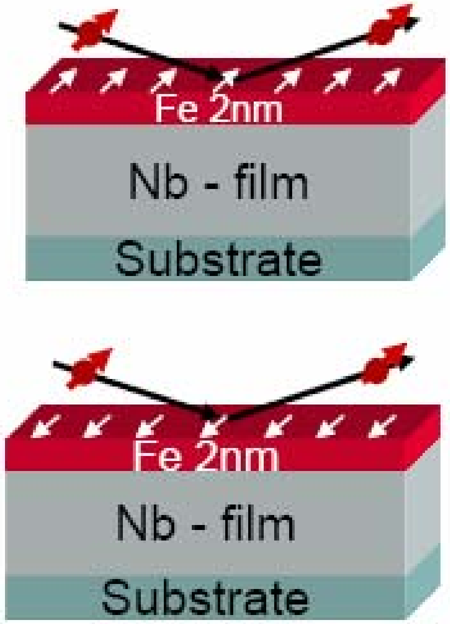


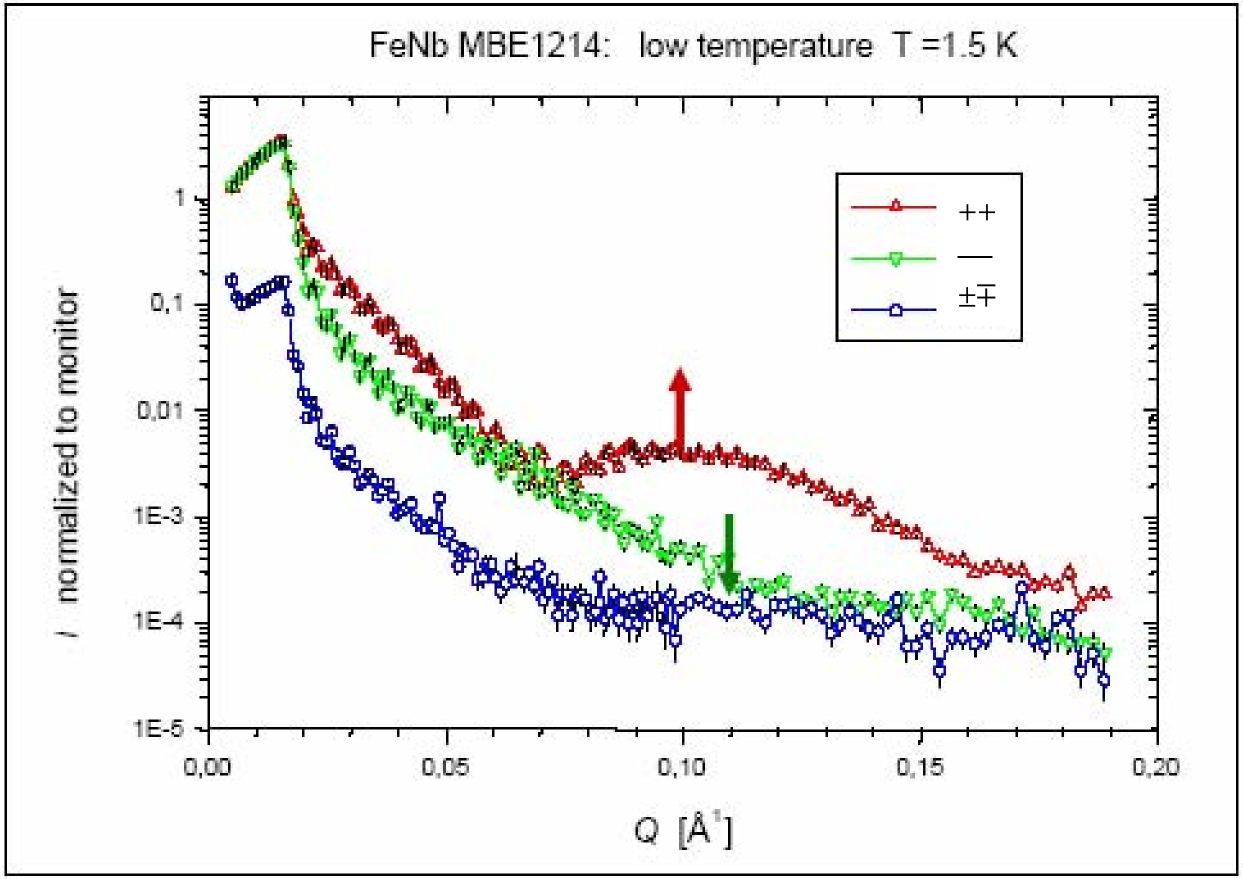


S. Hope *et al*., Phys. Rev. B **55** (1997) 11422

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

Neutron polarization analysis, Fe on Nb





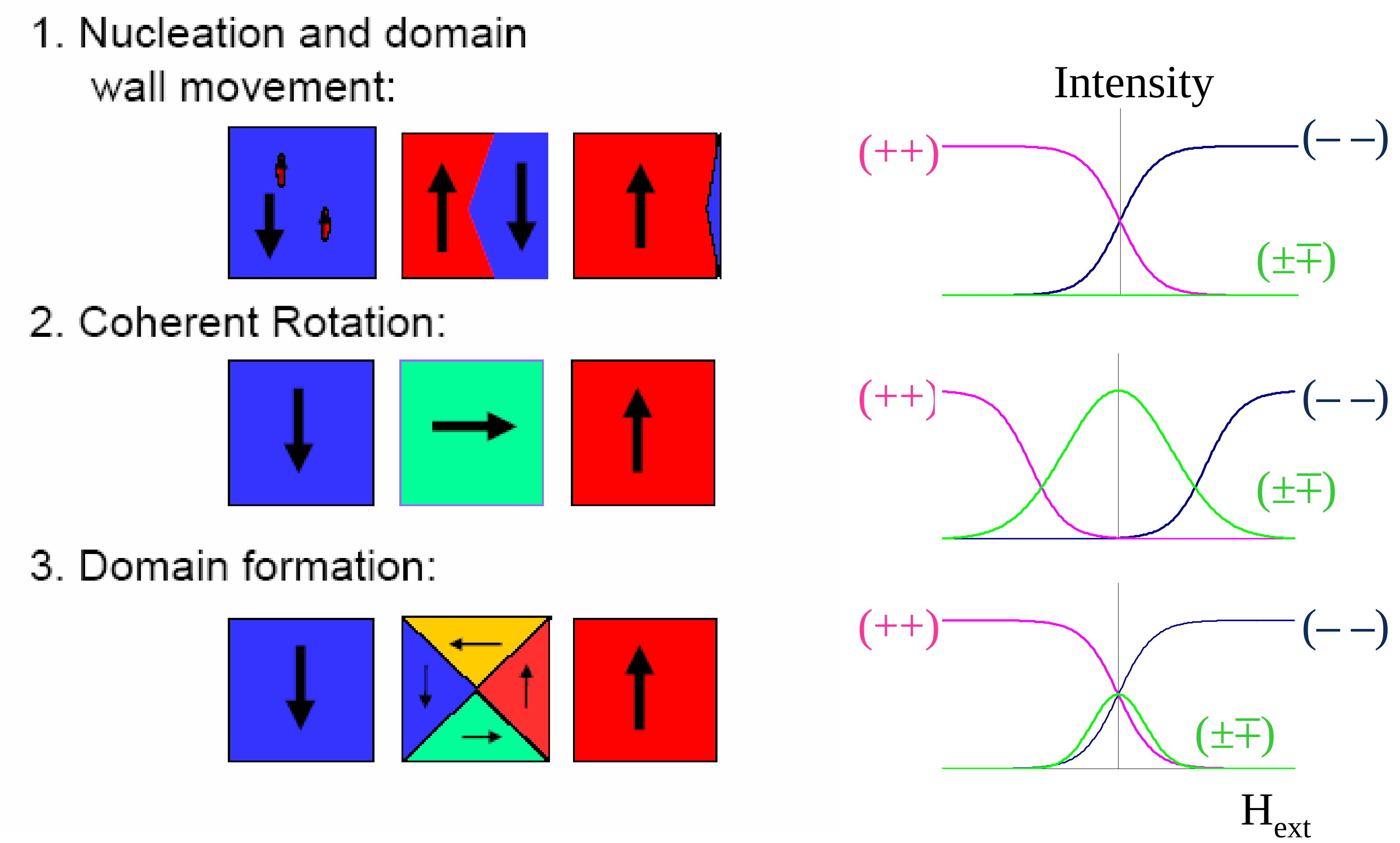
F. Radu and V. K. Igatovich, Physica B **267-268** (1999) 175

Determining how Magnetism Rotates

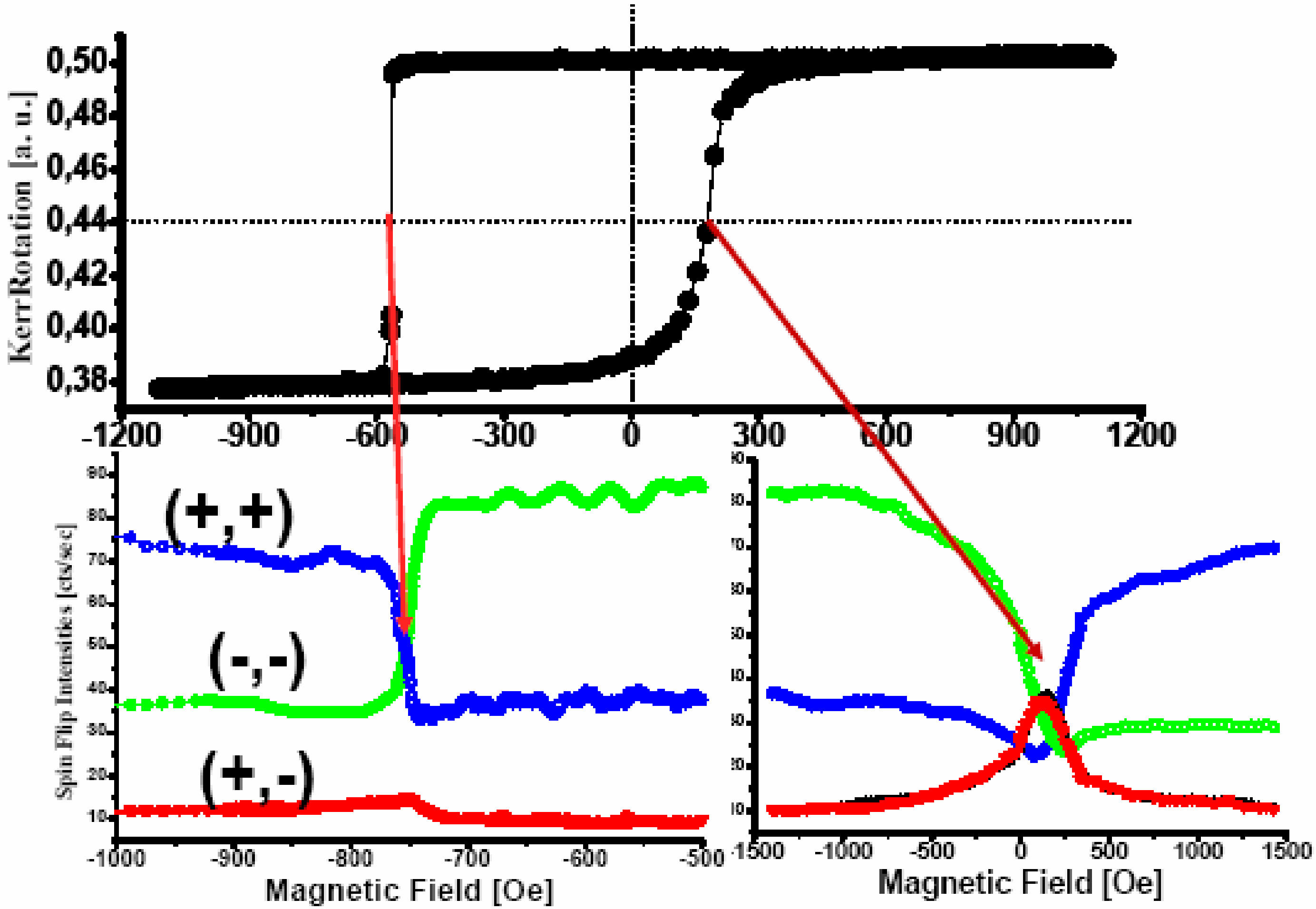
Recall: for Born approximation, **P⊥Q**

$$\frac{\mathrm{d}\sigma^{\pm\pm}}{\mathrm{d}\Omega} = \left| \int \left(b(\mathbf{r}) \mp M_{\perp z}(\mathbf{r}) \right) e^{i\mathbf{Q}\cdot\mathbf{r}} d\mathbf{r} \right|^2$$

$$\frac{\mathrm{d}\sigma^{\pm\mp}}{\mathrm{d}\Omega} = \left| \int M_{\perp y}(\mathbf{r}) e^{i\mathbf{Q}\cdot\mathbf{r}} d\mathbf{r} \right|^2$$



PNR from Co/CoO layer

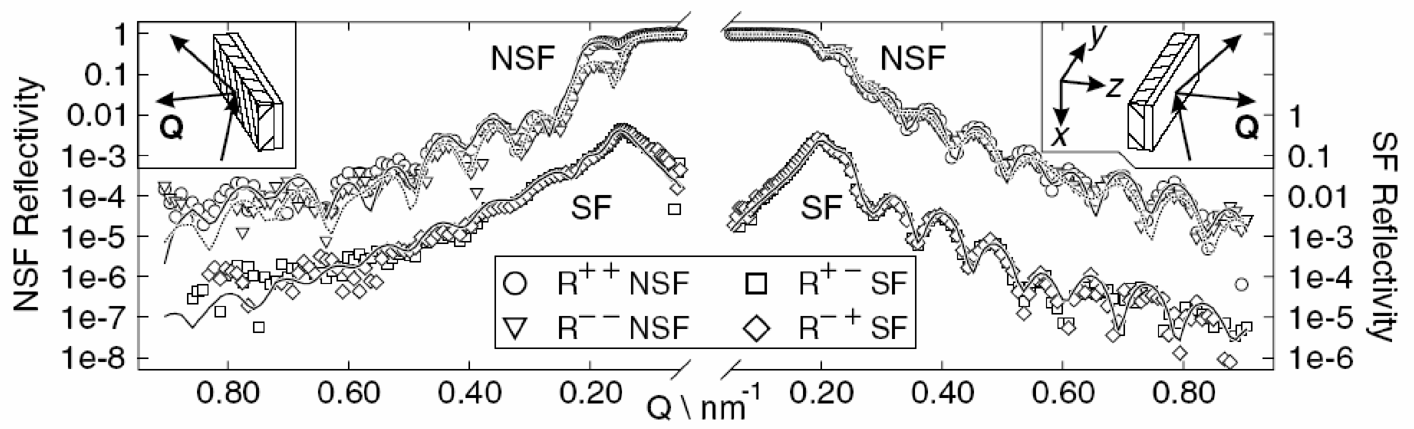


Nucleation

Domain formation

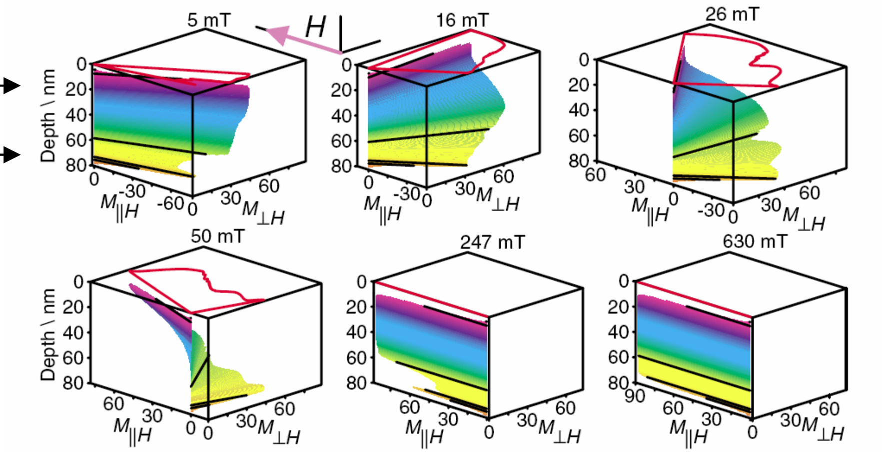
F. Radu *et al.*

Magnetic structures: induced twist in FePt/NiFe



FePt

NiFe

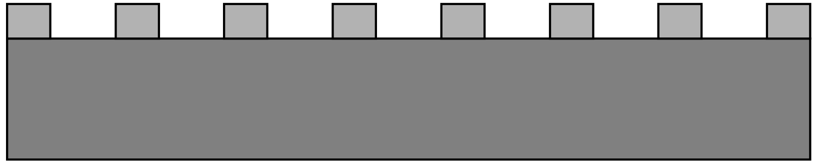


K. V. O'Donovan *et al.*, Phys. Rev. Lett. **88** (2002) 067201

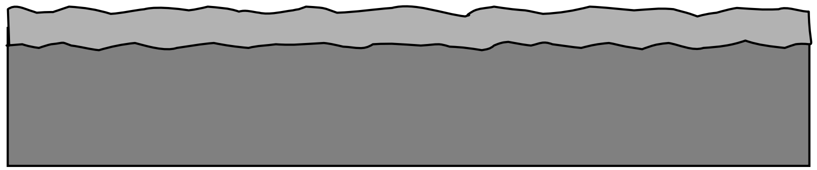
Off-specular scattering

Specular reflection tells nothing about:

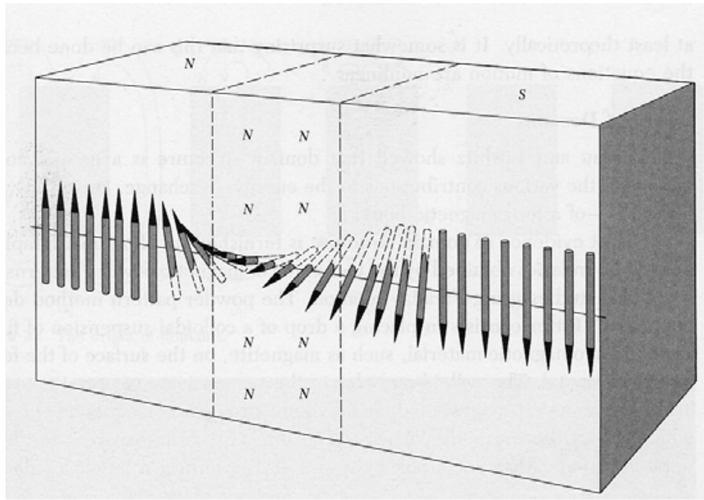
Lateral structures



Roughness, magnetic and/or nuclear



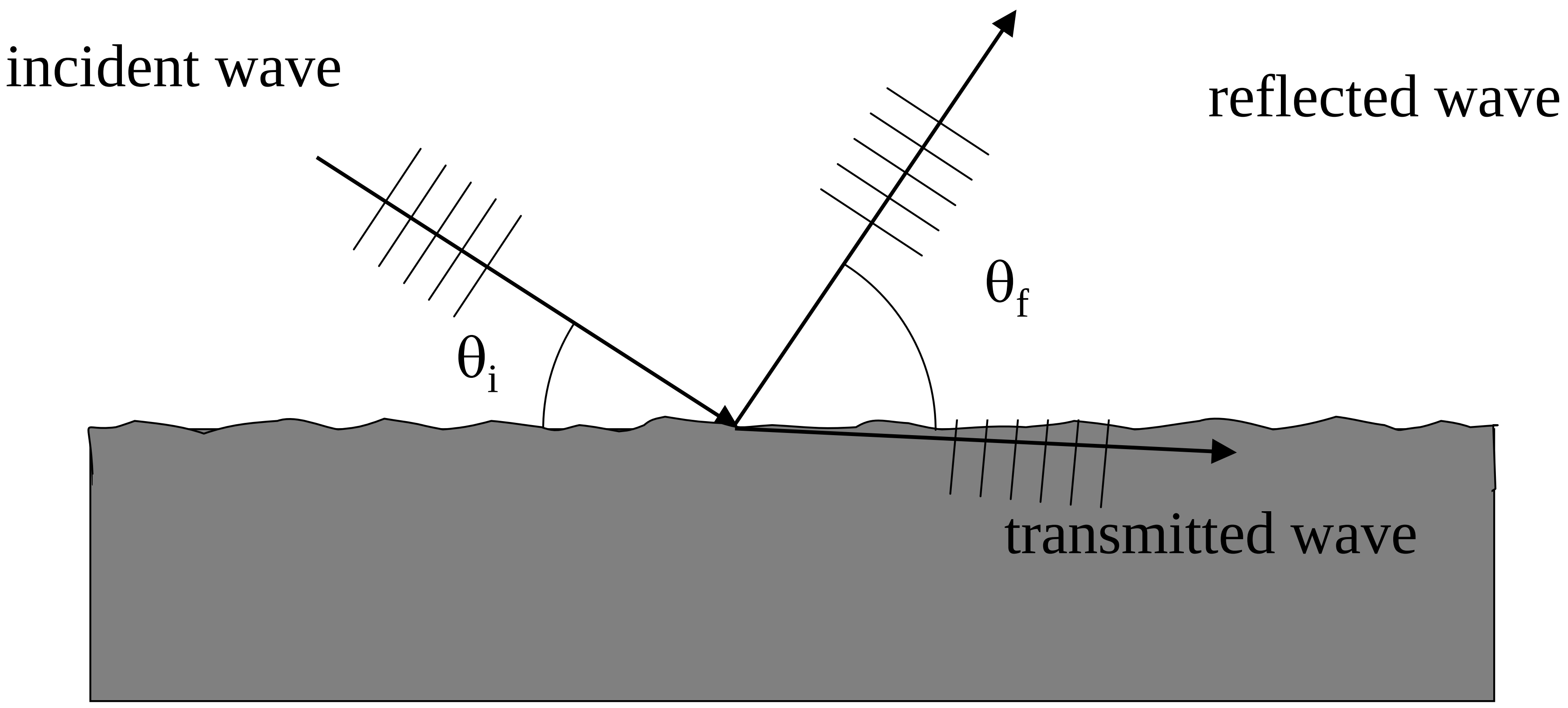
Bloch, Néel, and domain walls



For this we need to measure off-specular scattering

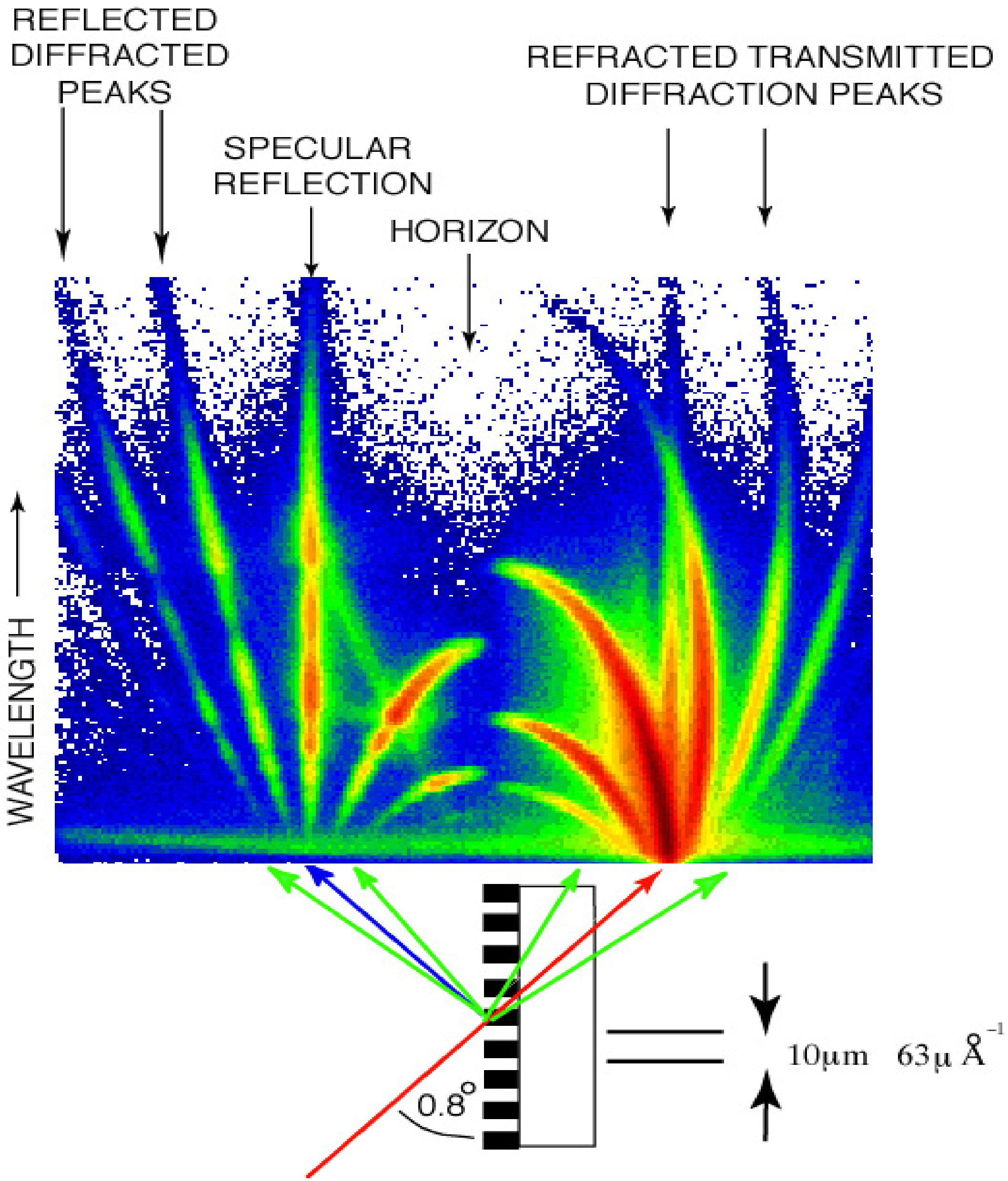
Off-specular scattering

$$\theta_i \neq \theta_f$$



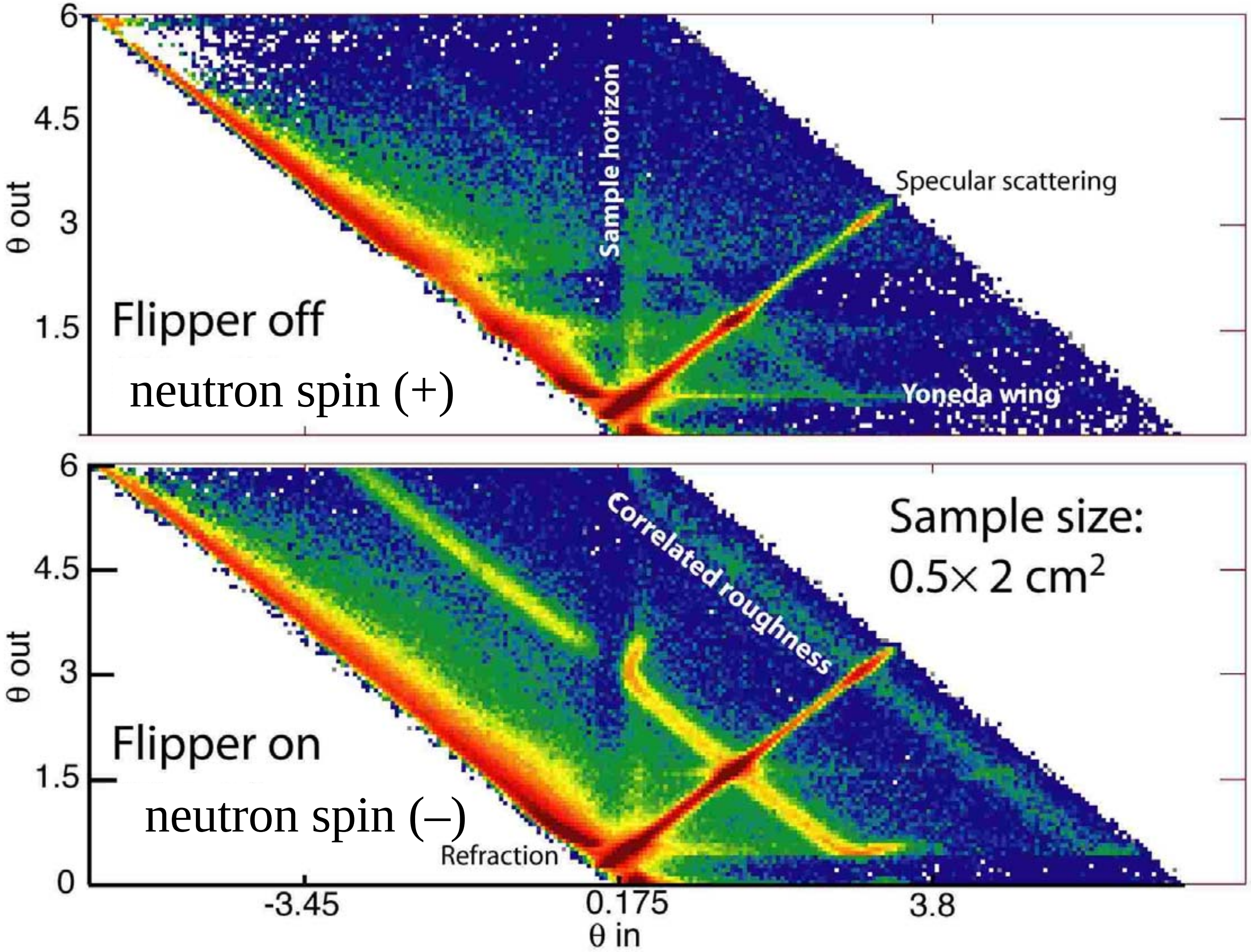
Analysis, however, is hard - particularly for polarization analysis

Off Specular Reflectivity from a Ni Grating



Correlated Magnetic Roughness in a Co/CoO multilayer

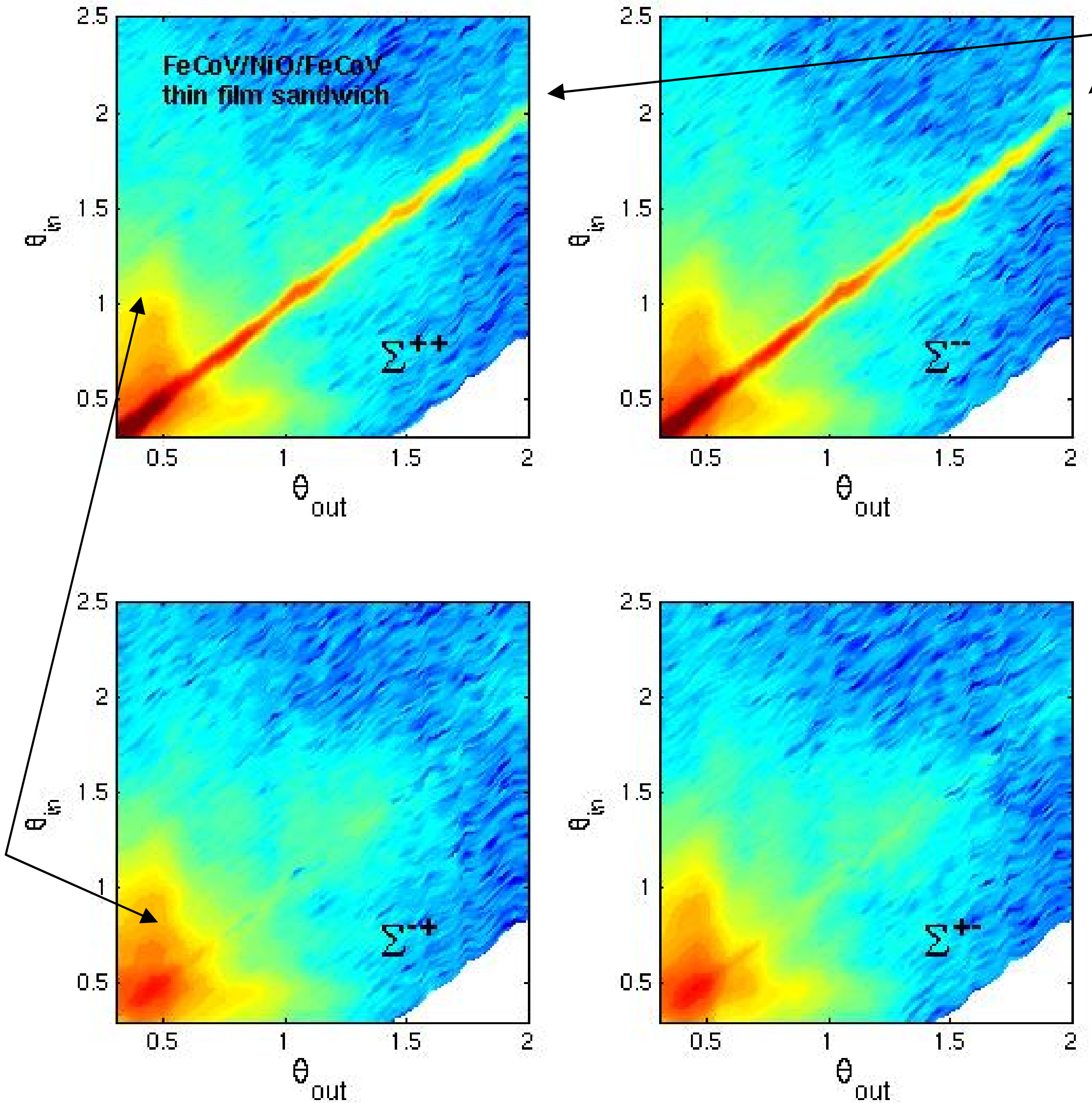
Measured with polarized neutrons, $\left|\hat{V}^{\pm}\right| = \left|b \pm \gamma\mu_N B\right|$



F. Radu *et al.*

Scattering from domain walls: FeCoV/NiO/FeCoV sandwich

Specular scattering,
$\theta_i = \theta_f$
$\Sigma^{\pm\pm} >> \Sigma^{\pm\mp}$



Off-specular scattering
$\theta_i \neq \theta_f$
$\Sigma^{\pm\pm} \approx \Sigma^{\pm\mp}$

Ch. Schanzer and V. R. Shah
K. H. Andersen *et al.*, submitted to Physica B

The phase problem

Return to the Born approximation. Recall:

$$\int V_m(\mathbf{r}) e^{i\mathbf{Q}\cdot\mathbf{r}} \cdot d\mathbf{r} = \mathbf{M}_\perp(\mathbf{Q})$$

If we could take the inverse Fourier transform of $\mathbf{M}_\perp(\mathbf{Q})$, we could unambiguously solve the magnetic structure.

The problem is that the cross-section is given by:

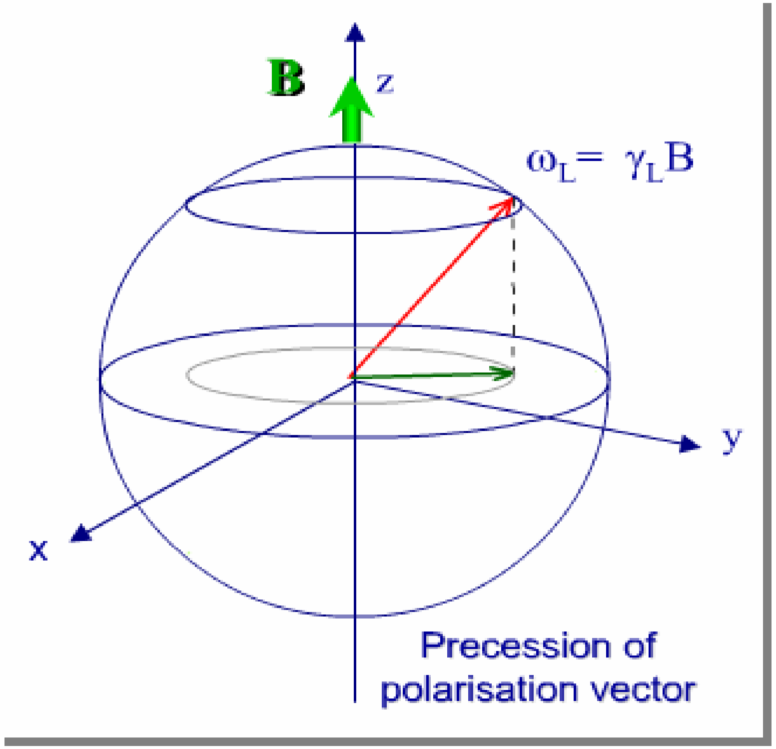
$$\frac{d\sigma_{magnetic}}{d\Omega} = \langle \mathbf{M}_\perp^*(\mathbf{Q}) \rangle \langle \mathbf{M}_\perp(\mathbf{Q}) \rangle$$

We therefore measure the *amplitude* of $\mathbf{M}_\perp(\mathbf{Q})$, but lose the *phase*.

Normally a model is fitted to the data, but what can be done if two models give equally good fits?

Polarimetry

The previous discussion of neutron polarization analysis was over-simplified. The neutron spin is not really ‘flipped’ by a magnetic field, it *precesses*



The previous discussion dealt with the *projection* of the neutron spin along the initial polarization axis

Bragg Scattering with Polarization Analysis

$$\vec{P}_f \sigma = \begin{bmatrix} \vec{P}_i NN^* & \text{nucl} \\ -\vec{P}_i(\vec{M}_\perp . \vec{M}_\perp^*) + \vec{M}_\perp(\vec{P}_i . \vec{M}_\perp^*) + \vec{M}_\perp^*(\vec{P}_i . \vec{M}_\perp) - i\left(\overleftrightarrow{M}_\perp^* \times \overleftrightarrow{M}_\perp\right) & \text{mag} \\ + N\vec{M}_\perp^* + N^*\vec{M}_\perp - i\left(N\vec{M}_\perp^* - N^*\vec{M}_\perp\right) \times \vec{P}_i & \text{nucl-mag int} \end{bmatrix}$$

σ	total scattering cross-section of Bragg peak
N	nuclear structure factor
$\vec{M}_\perp$	magnetic interaction vector $\propto \vec{Q} \times \vec{M} \times \vec{Q}$ where $\vec{M}$ is the magnetic structure factor
$\vec{P}_i, \vec{P}_f$	incident, final polarization vector

M. Blume, Phys. Rev. **130** (1963) 1670
S. V. Maleev *et al*., Sov. Phys. Solid State **4** (1963) 2533
T. J. Hicks, Adv. Phys. **46** (1996) 243

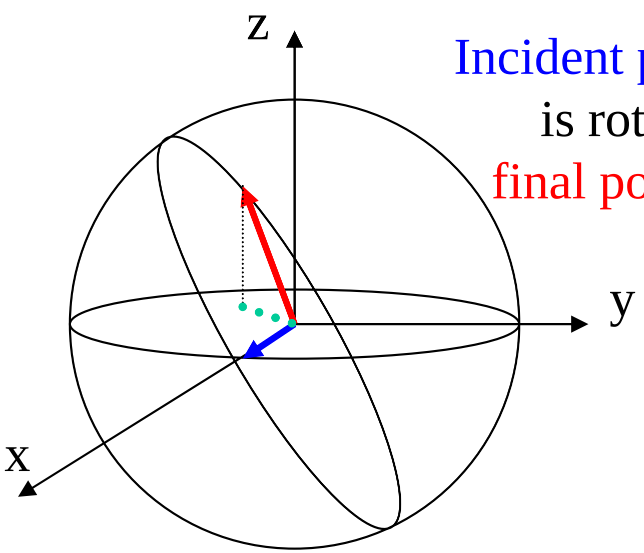
The polarization tensor

Measuring the rotation of polarization due to the scattering at the sample gives a unique solution for complex magnetic structures

This is a measurement of the polarization tensor:

$$\vec{P}_f = \mathbf{P}\vec{P}_i + \vec{T}, \text{ where } \mathbf{P} = \begin{bmatrix} P_{xx} & P_{xy} & P_{xz} \\ P_{yx} & P_{yy} & P_{yz} \\ P_{zx} & P_{zy} & P_{zz} \end{bmatrix}$$

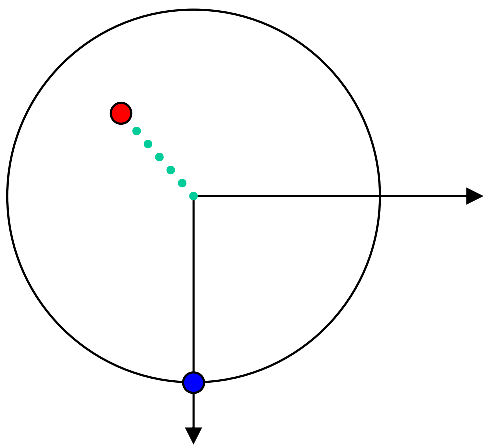
It is often shown as a stereographic projection:



Incident polarization

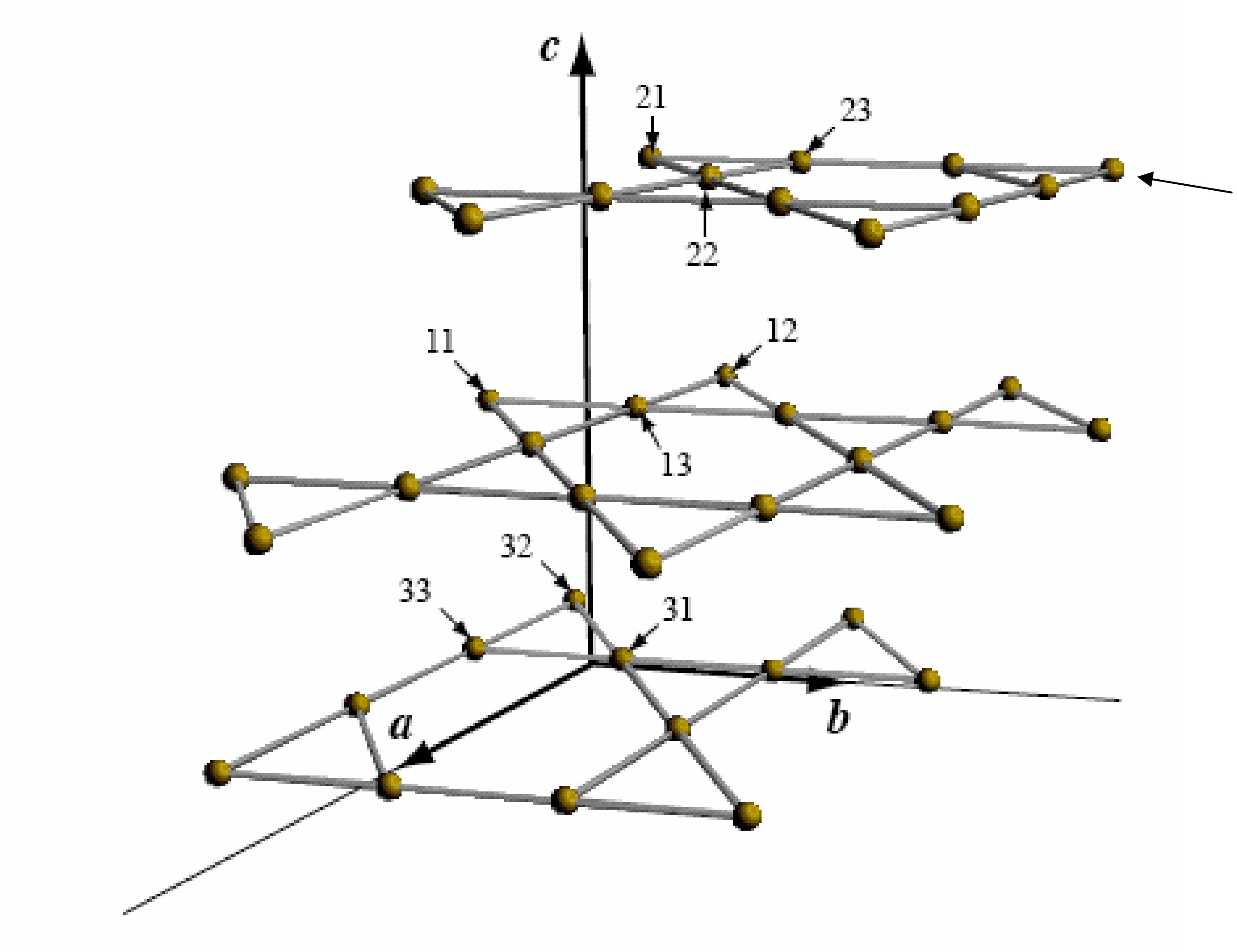
is rotated to

final polarization



The magnetic structure of potassium Jarosite

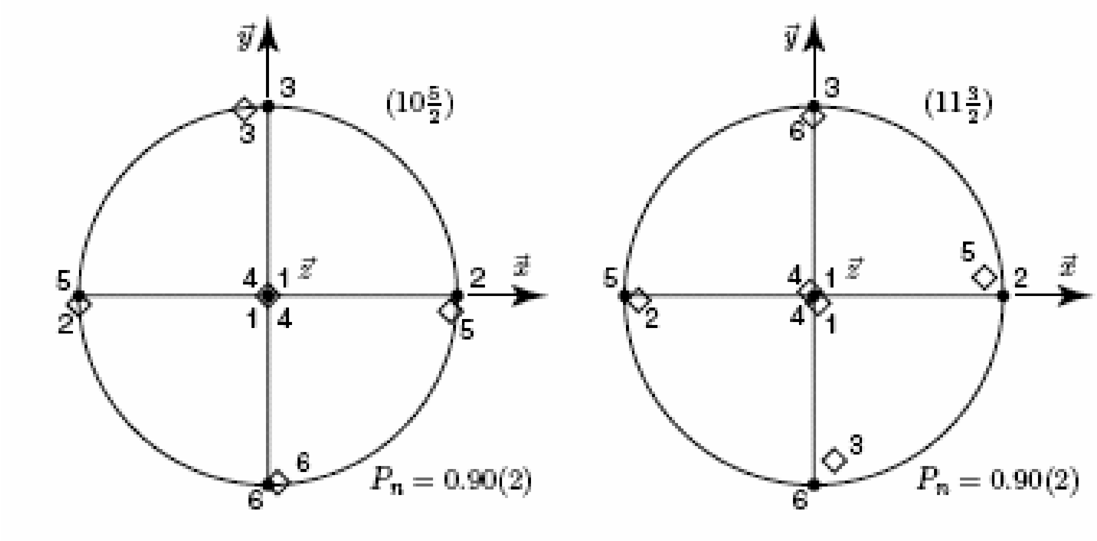
The magnetic iron atoms lie on a Kagome lattice



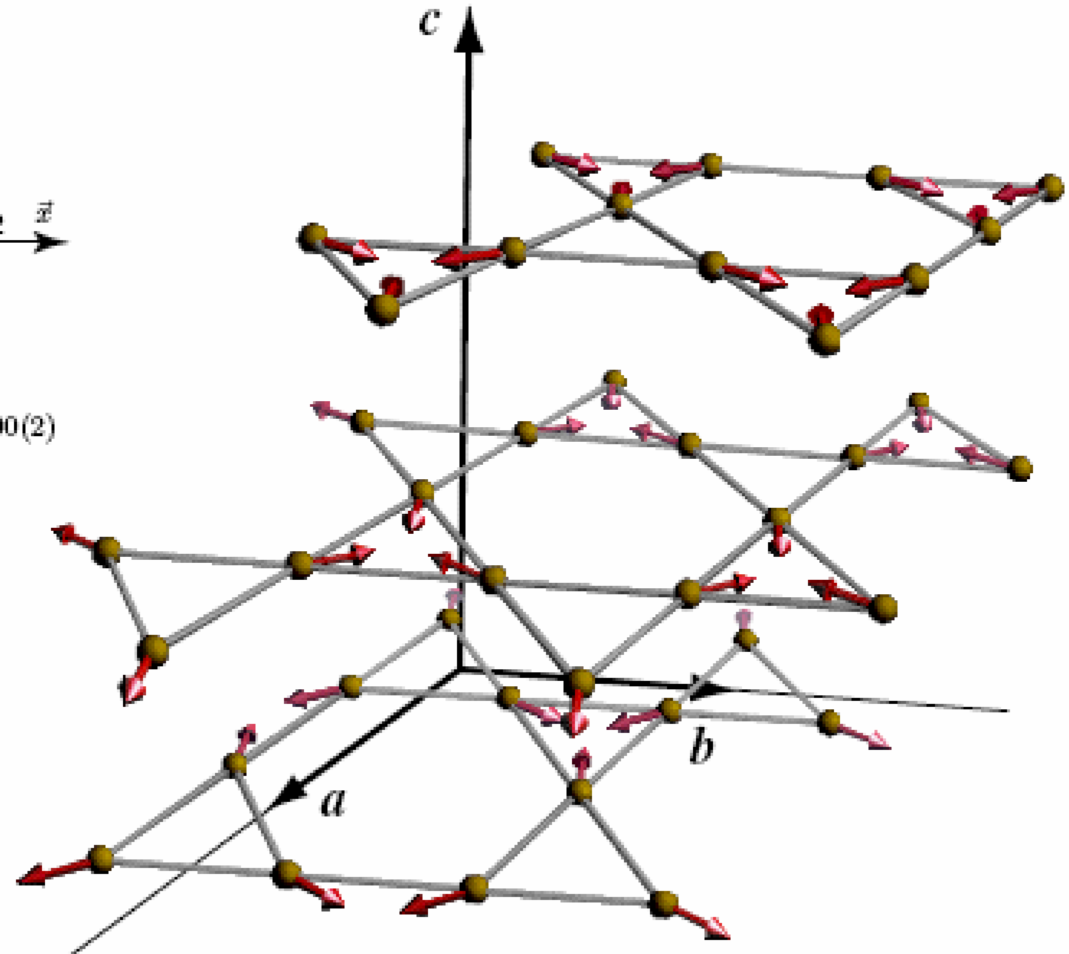
The magnetic structure of potassium Jarosite

The rotation of the polarization around various Bragg peaks…

leads to an unambiguous magnetic structure



- ● Incident polarization
- ◇ Final polarization



A. Harrison *et al.*, in preparation

Neutron polarimetry currently works only for materials with no spontaneous moment
i.e. antiferromagnets

Conclusions

Neutrons interact with the magnetic induction of the sample.

Scattering is the most common neutron method for looking at magnetism.
Elastic scattering probes magnetic structures.
Inelastic scattering probes magnetic dynamics.

Polarization analysis is a particularly powerful tool for studies of magnetic materials.

The information obtained is
COMPREHENSIVE
(in theory, it covers all length and time scales, limited only by the wavelength)

and frequently UNIQUE.
(e.g. no other way exists to measure non-collinear ferromagnetic structures)

Neutrons show whether the magnetic properties of a material are novel, and frequently provide the information required to find *why* they are novel.