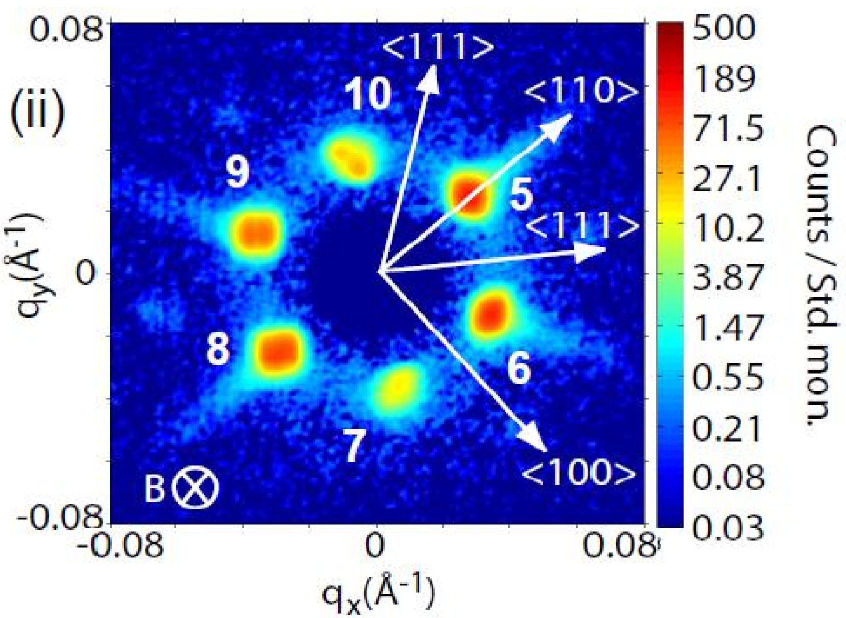
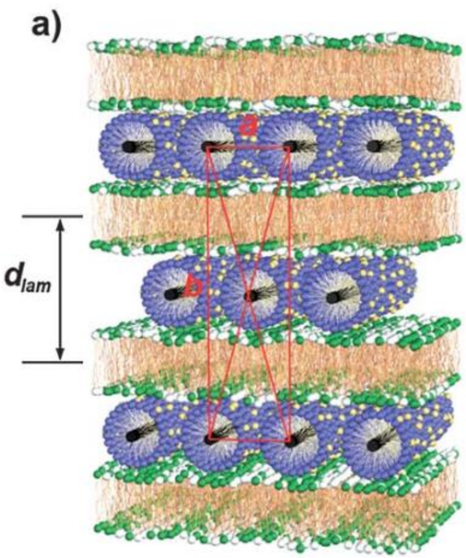
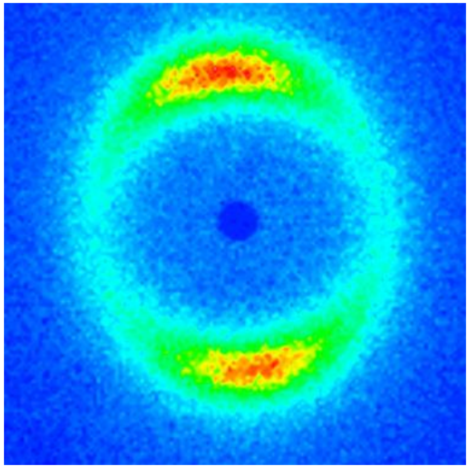
FRM II
Forschungs-Neutronenquelle
Heinz Maier-Leibnitz

MLZ
Heinz Maier-Leibnitz Zentrum

Physics with Neutrons II, SS 2016







Lecture 7, 13.6.2016

MLZ is a cooperation between:

JÜLICH FORSCHUNGSZENTRUM

Helmholtz-Zentrum Geesthacht
Zentrum für Material- und Küstenforschung

TUM
Technische Universität München

Plan for SS 2016

- VL1: Repetition of winter term, basic neutron scattering theory
- VL2: SANS, theory and applications
- VL3: Neutron optics
- VL4: Reflectometry and dynamical scattering theory
- VL5: Diffuse neutron scattering
- VL6: Magnetic scattering cross section
- VL7: Magnetic structures and structure analysis
- VL8: Polarized neutrons and 3d-polarimetry
- VL9: Inelastic scattering on magnetism
- VL10: 4.7.2016 (8:30!!) Phase transitions and critical phenomena as seen by neutrons
- VL11: Spin echo spectrocopy

Exam: Please register until 30.6.2016

Reminder: Cross section for magnetic neutron scattering

Magnetic neutron scattering

Nuclear neutron scattering

Interaction: Fermi pseudopotential $$V(r) = \frac{2\pi\hbar^2}{m} b\delta(r)$$

- Scalar funtion
- Point like (delta function)
- For an incoming plane wave: s-wave scattering
- Spherical symmetry
- FT of delta function is constant

Magnetic neutron scattering

Magnetic neutron scattering:

Interaction: Magnetic moment of neutron interacts with local magnetic field

$$\hat{U} = \hat{\mu} \cdot \vec{H} \qquad \hat{U} = \hat{\mu} \cdot \vec{H} = \gamma \mu_N \hat{\sigma} \cdot \vec{H}$$

$$\vec{H} = \nabla \times \left(\frac{\vec{\mu}_e \times \vec{R}}{|\vec{R}|^3} \right) - \frac{e}{c} \frac{\vec{v}_e \times \vec{R}}{|\vec{R}|^3}$$

Spin of electrons

Orbital momentum

- Vector (dipole-dipole) interaction
- Extended range, not point like
- No s-wave scattering
- No spherical symmetry
- Magnetic moments: Unpaired electrons

Magnetic neutron scattering

Leaving away the maths

- For spin only scattering (neglect orbital momentum)
- For unpolarized neutrons (average over polarization states)
- For identical magnetic ions with localized moments

Master formula

$$\boxed{\begin{aligned}\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\mathrm{d}E'}\right) &= (\gamma r_0)^2\frac{k'}{k}F^2(\vec{Q})e^{-2W(\vec{Q})}\sum_{\alpha,\beta}(\delta_{\alpha,\beta}-\frac{Q_\alpha Q_\beta}{Q^2})S^{\alpha\beta}(Q,\omega)\\ S^{\alpha\beta}(Q,\omega) &= \sum_{j,j'}e^{i\vec{Q}(R_j-R_{j'})}\sum_{\lambda,\lambda'}p_\lambda\langle\lambda|S_{j'}^\alpha|\lambda'\rangle\langle\lambda|S_j^\beta|\lambda'\rangle\delta(\hbar\omega+E_\lambda-E_{\lambda'})\end{aligned}}$$

Magnetic neutron scattering

Master formula

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right) = (\gamma r_0)^2 \frac{k'}{k} F^2(\vec{Q}) e^{-2W(\vec{Q})} \sum_{\alpha,\beta} (\delta_{\alpha,\beta} - \frac{Q_\alpha Q_\beta}{Q^2}) S^{\alpha\beta}(Q,\omega)$$

$$S^{\alpha\beta}(Q,\omega) = \sum_{j,j'} e^{i\vec{Q}(R_j - R_{j'})} \sum_{\lambda,\lambda'} p_\lambda \langle\lambda|S^\alpha_{j'}|\lambda'\rangle\langle\lambda|S^\beta_j|\lambda'\rangle \delta(\hbar\omega + E_\lambda - E_{\lambda'})$$

Dipole-dipole interaction: Angular dependence

Neutron polarization factor $(\delta_{\alpha,\beta} - \dfrac{Q_\alpha Q_\beta}{Q^2})$

⇨ Only moments perpendicular to Q contribute to magnetic scattering

Don't confuse with polarized neutrons!

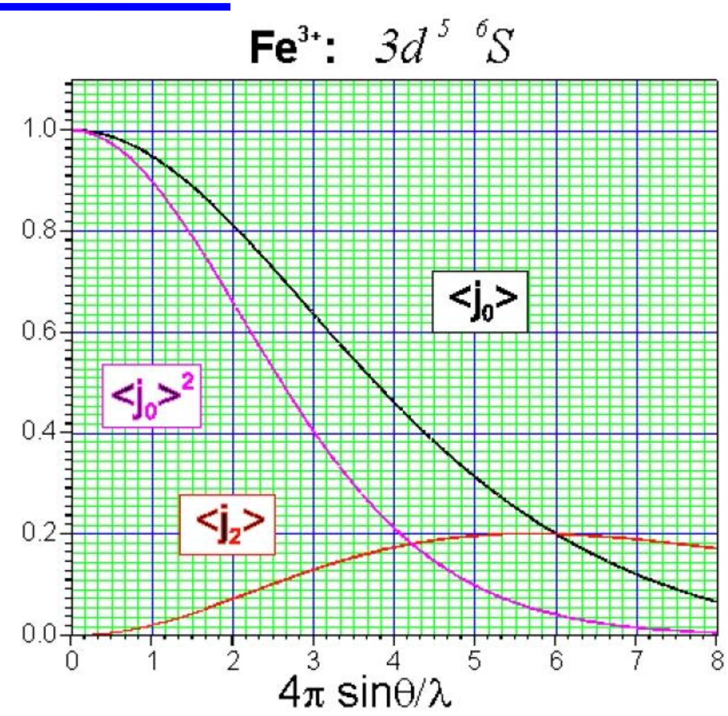
Master formula

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right) = (\gamma r_0)^2 \frac{k'}{k} \underline{F^2(\vec{Q})} e^{-2W(\vec{Q})} \sum_{\alpha,\beta} (\delta_{\alpha,\beta} - \frac{Q_\alpha Q_\beta}{Q^2}) S^{\alpha\beta}(Q,\omega)$$

$$S^{\alpha\beta}(Q,\omega) = \sum_{j,j'} e^{i\vec{Q}(R_j - R_{j'})} \sum_{\lambda,\lambda'} p_\lambda \langle\lambda|S^\alpha_{j'}|\lambda'\rangle\langle\lambda|S^\beta_j|\lambda'\rangle \delta(\hbar\omega + E_\lambda - E_{\lambda'})$$

Dipole-dipole interaction: Magnetic form factor

- Fourier transform of electron cloud
- Useful to discriminate magnetic/nuclear scattering
- Check the tables for each ion!



Magnetic neutron scattering

For the case of orbital momentum + spin

$$\mu = -\mu_b(L + 2S)$$

Effective angular momentum operator

$$\hat{S}_j^\alpha = \frac{1}{2} g \hat{J}_j^\alpha$$

Landé splitting factor

$$g = 1 + \frac{J(J+1) - L(L+1) + S(S+1)}{2J(J+1)}$$

➡ Approximation for small Q, spin+orbital momentum

Magnetic scattering function

$$S^{\alpha\beta}(Q,\omega) = \sum_{j,j'} e^{i\vec{Q}(R_j - R_{j'})} \underline{\langle S_{j'}^{\alpha}(0)\ S_{j'}^{\beta}(t)\rangle} e^{-i\omega t} \mathrm{d}t$$

<u>Spin correlation funtion:</u>

Correlation of magnetic moment at site j, time t=0 and site j', time t=t

➡ <u>Fourier transform measured with neutrons!</u>

Magnetic neutron scattering

Why neutrons are so unique:

Fluctuation dissipation theorem

$$S^{\alpha\beta}(Q,\omega) = \frac{N\hbar}{\pi}\left(1 - e^{-\frac{\hbar\omega}{k_b T}}\right)^{-1} \mathrm{Im}\chi^{\alpha\beta}(\vec{Q},\omega)$$

Neutrons directly measure generalized susceptibility tensor

$$M^{\alpha}(\vec{Q},\omega) = \chi^{\alpha\beta}(\vec{Q},\omega) H^{\beta}(\vec{Q},\omega)$$

Magnetic structures

Paramagnets

By defition no correlation between spins $\langle \hat{S}_0^\alpha \rangle \langle \hat{S}_l^\beta \rangle = 0$

Consider only l=0 (self correlation) ⇨ Incoherent scattering

$$\langle \hat{S}_0^\alpha \rangle \langle \hat{S}_0^\beta \rangle = \delta_{\alpha\beta} \langle \hat{S}_0^\alpha \rangle \langle \hat{S}_0^\beta \rangle = \delta_{\alpha\beta} \langle (\hat{S}_0^\alpha)^2 \rangle = \frac{1}{3} \delta_{\alpha\beta} \langle \hat{\boldsymbol{S}} \rangle^2 = \frac{1}{3} \delta_{\alpha\beta} S(S+1)$$

Non-zero only for α=β

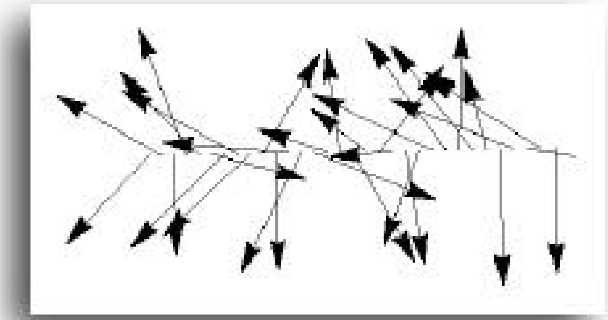
$$\sum_{\alpha,\beta} \left(\delta_{\alpha\beta} - \frac{Q_\alpha Q_\beta}{Q^2} \right) = \sum_\alpha \left(1 - \left(\frac{Q_\alpha}{Q} \right)^2 \right) = 2$$

Paramagnetic scattering (isotropic in Q)

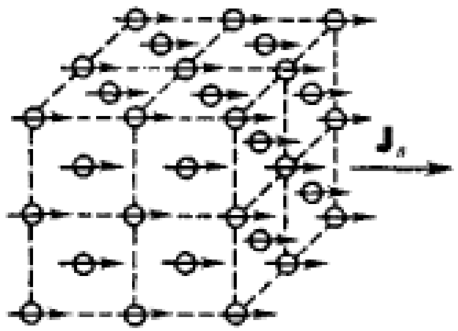
$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \frac{2}{3} N (\gamma r_0)^2 e^{-2W(\boldsymbol{Q})} F^2(\boldsymbol{Q}) S(S+1)$$

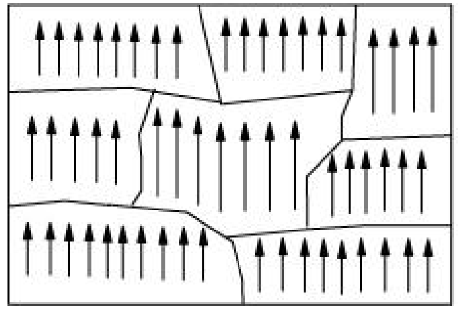
Scales with (S(S+1), S^2

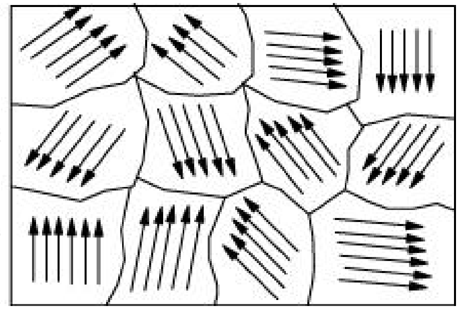
Scales with magnetic form factor F(Q)

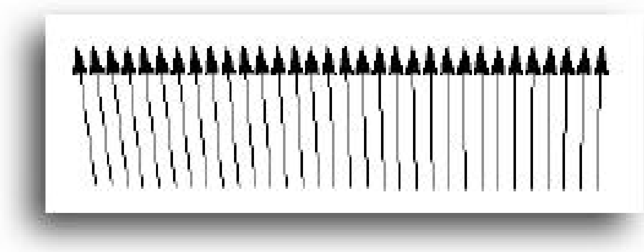


Ferromagnets









Consider a single domain sample, sind along z $\langle \hat{S}_l^x \rangle = \langle \hat{S}_l^y \rangle = 0; \quad \langle \hat{S}_l^z \rangle \neq 0$

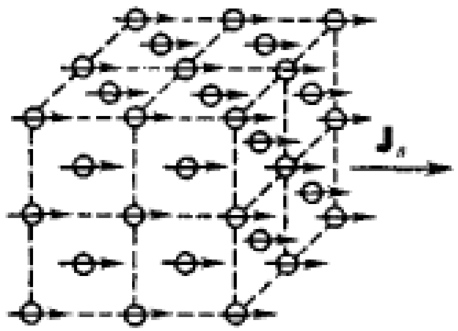
- Bravais properties of the ferromagnet $$\frac{d\sigma}{d\Omega} = (\gamma r_0)^2 e^{-2W(\boldsymbol{Q})} F^2(\boldsymbol{Q}) \left(1 - \left(\frac{Q_z}{Q}\right)^2\right) \langle \hat{S}^z \rangle^2 \sum_{\boldsymbol{l}} e^{\imath \boldsymbol{Q} \cdot \boldsymbol{l}}$$

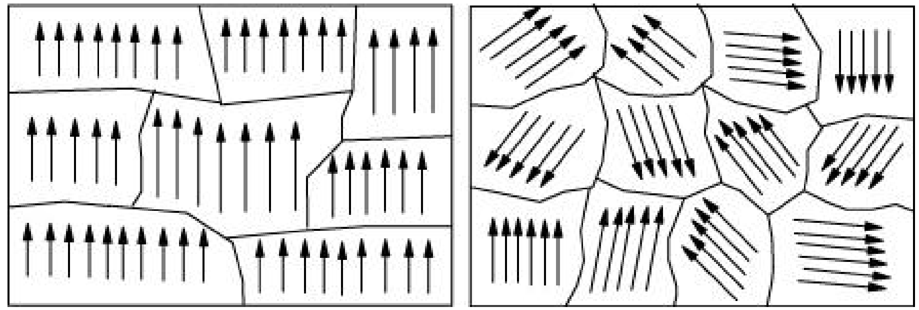
- Unit vector along magnetization direction z $$\frac{Q_z}{Q} = \frac{\boldsymbol{Q} \cdot \boldsymbol{e}}{Q} = \frac{\boldsymbol{\tau} \cdot \boldsymbol{e}}{\tau}$$

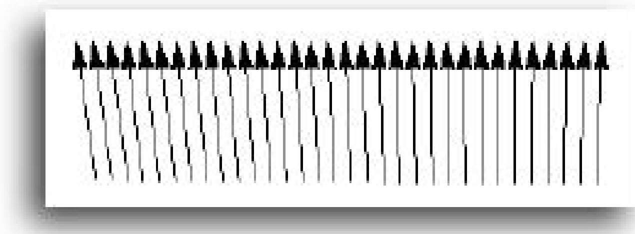
Use lattice sum identities

$$\boxed{\frac{d\sigma}{d\Omega} = N \frac{(2\pi)^3}{v_0} (\gamma r_0)^2 e^{-2W(\boldsymbol{Q})} F^2(\boldsymbol{Q}) \langle \hat{S}^z \rangle^2 \sum_{\boldsymbol{\tau}} \langle 1 - \left(\frac{\boldsymbol{\tau} \cdot \boldsymbol{e}}{\tau}\right)^2 \rangle \, \delta(\boldsymbol{Q} - \boldsymbol{\tau})}$$

Ferromagnets





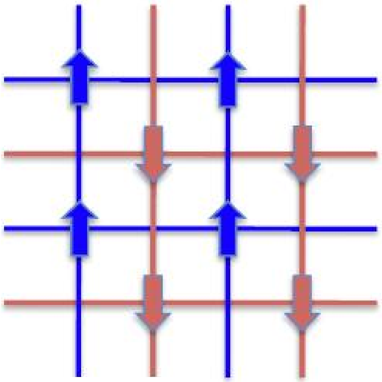


$$\boxed{\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = N\frac{(2\pi)^3}{v_0}(\gamma r_0)^2 e^{-2W(\boldsymbol{Q})} F^2(\boldsymbol{Q})\langle \hat{S}^z\rangle^2 \sum_{\boldsymbol{\tau}} \langle 1 - \left(\frac{\boldsymbol{\tau}\cdot\boldsymbol{e}}{\tau}\right)^2\rangle\, \delta(\boldsymbol{Q}-\boldsymbol{\tau})}$$

- Scattering appears at reciprocal lattice vectors
- Scattering proportional to square of the magn. moment
- Strong temperature dependence close to transition temp.
- Intensity folows magn. Form factor F(Q)
- Depends on relative direction of S and Q

Alignment with field ⇨ vary polarization factor!

Antiferromagnets



- Split into two sublattices A and B on site d
- Treat as non-Bravais lattice with two atoms per unit cell (Sublattice A and B, with spin up/down

$$\frac{d\sigma}{d\Omega} = (\gamma r_0)^2 e^{-2W(\boldsymbol{Q})} F^2(\boldsymbol{Q}) \left(1 - \left(\frac{Q_z}{Q}\right)^2\right) \langle \hat{S}^z \rangle^2 \sum_{\boldsymbol{l}} e^{\imath \boldsymbol{Q}\cdot\boldsymbol{l}} \sum_{\boldsymbol{d}} \sigma_{\boldsymbol{d}} e^{\imath \boldsymbol{Q}\cdot\boldsymbol{d}}$$

With $\sigma_{\boldsymbol{d}} = \pm 1$ for both sublattices A and B

- Use lattice sum indentities

Antiferromagnet

$$\frac{d\sigma}{d\Omega} = N_m \frac{(2\pi)^3}{v_{0m}} (\gamma r_0)^2 e^{-2W(\boldsymbol{Q})} \times \sum_{\boldsymbol{\tau}_m} |S_m(\boldsymbol{\tau}_m)|^2 \langle 1 - \left(\frac{\boldsymbol{\tau}_m \cdot \boldsymbol{e}}{\boldsymbol{\tau}_m}\right)^2 \rangle \delta(\boldsymbol{Q} - \boldsymbol{\tau}_m)$$

$$S_m(\boldsymbol{\tau}_m) = \langle S^z \rangle F(\boldsymbol{\tau}_m) \sum_{\boldsymbol{d}} \sigma_{\boldsymbol{d}} e^{\imath \boldsymbol{\tau}_m \cdot \boldsymbol{d}}$$ Magnetic structure factor

Antiferromagnets

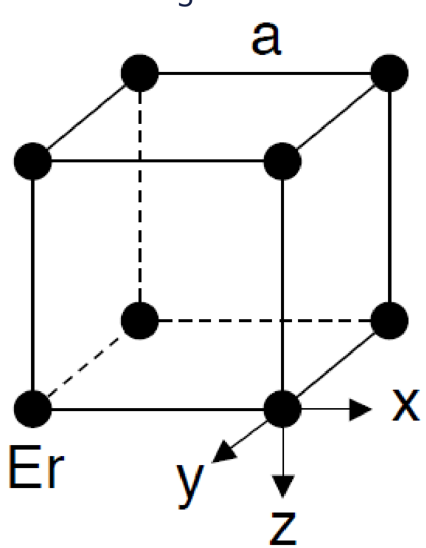
Antiferromagnet

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = N_m \frac{(2\pi)^3}{v_{0m}} (\gamma r_0)^2 e^{-2W(\boldsymbol{Q})} \times \sum_{\boldsymbol{\tau}_m} |S_m(\boldsymbol{\tau}_m)|^2 \langle 1 - \left(\frac{\boldsymbol{\tau}_m \cdot \boldsymbol{e}}{\boldsymbol{\tau}_m}\right)^2 \rangle \delta(\boldsymbol{Q} - \boldsymbol{\tau}_m)$$

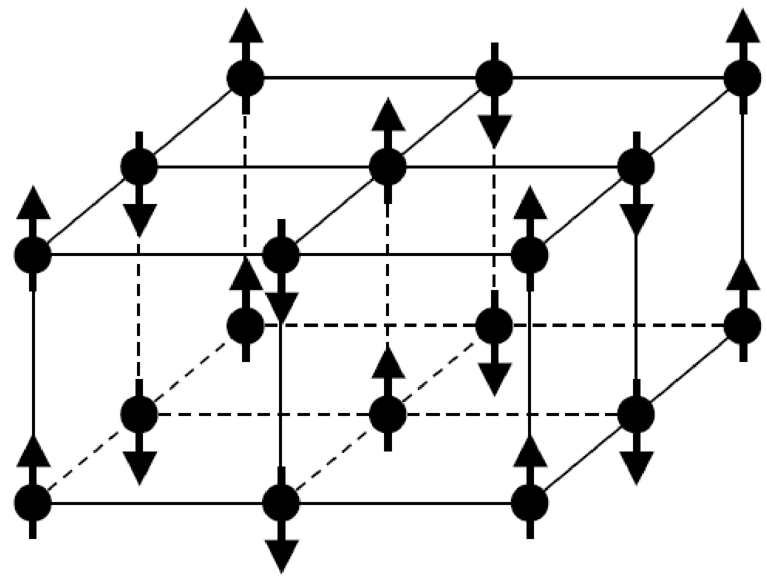
$$S_m(\boldsymbol{\tau}_m) = \langle S^z \rangle F(\boldsymbol{\tau}_m) \sum_{\boldsymbol{d}} \sigma_{\boldsymbol{d}} e^{\imath \boldsymbol{\tau}_m \cdot \boldsymbol{d}}$$

Magnetic structure factor

Example $ErPd_3$



Nuclear unit cell



Magnetic unit cell ⇨ Doubling of x and y direction

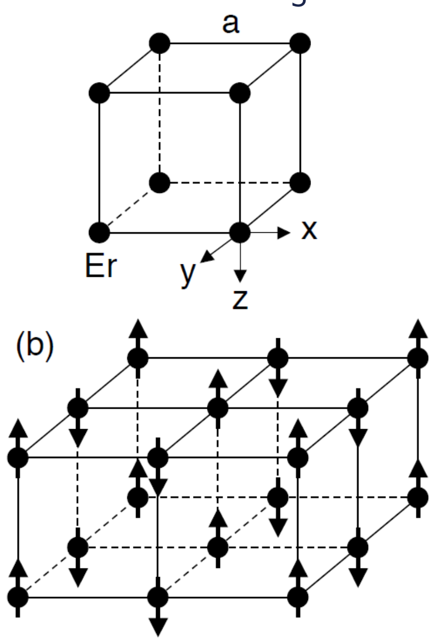
Antiferromagnets

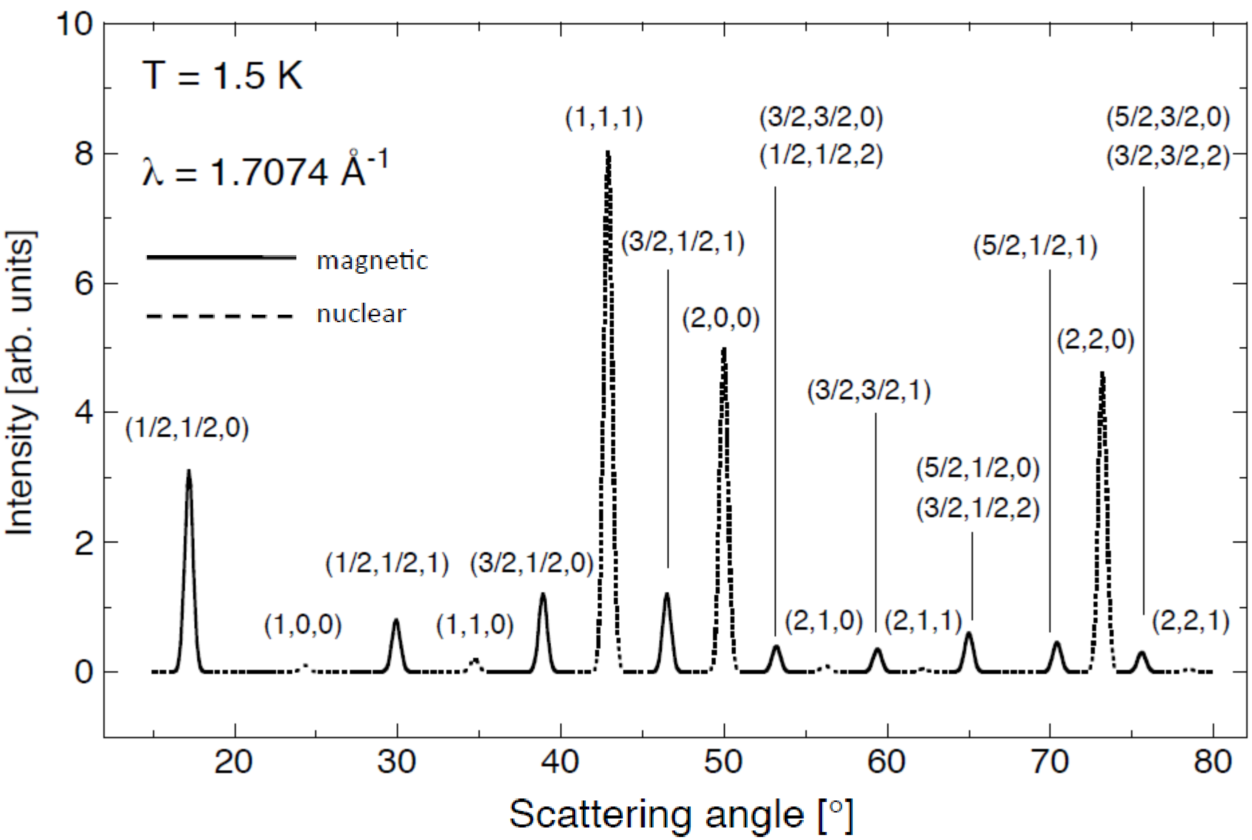
Antiferromagnet

$$\frac{d\sigma}{d\Omega} = N_m \frac{(2\pi)^3}{v_{0m}} (\gamma r_0)^2 e^{-2W(\boldsymbol{Q})} \times \sum_{\boldsymbol{\tau}_m} |S_m(\boldsymbol{\tau}_m)|^2 \langle 1 - \left(\frac{\boldsymbol{\tau}_m \cdot \boldsymbol{e}}{\boldsymbol{\tau}_m}\right)^2 \rangle \delta(\boldsymbol{Q} - \boldsymbol{\tau}_m)$$

$$S_m(\boldsymbol{\tau}_m) = \langle S^z \rangle F(\boldsymbol{\tau}_m) \sum_{\boldsymbol{d}} \sigma_{\boldsymbol{d}} e^{\imath \boldsymbol{\tau}_m \cdot \boldsymbol{d}}$$ Magnetic structure factor

Example $ErPd_3$

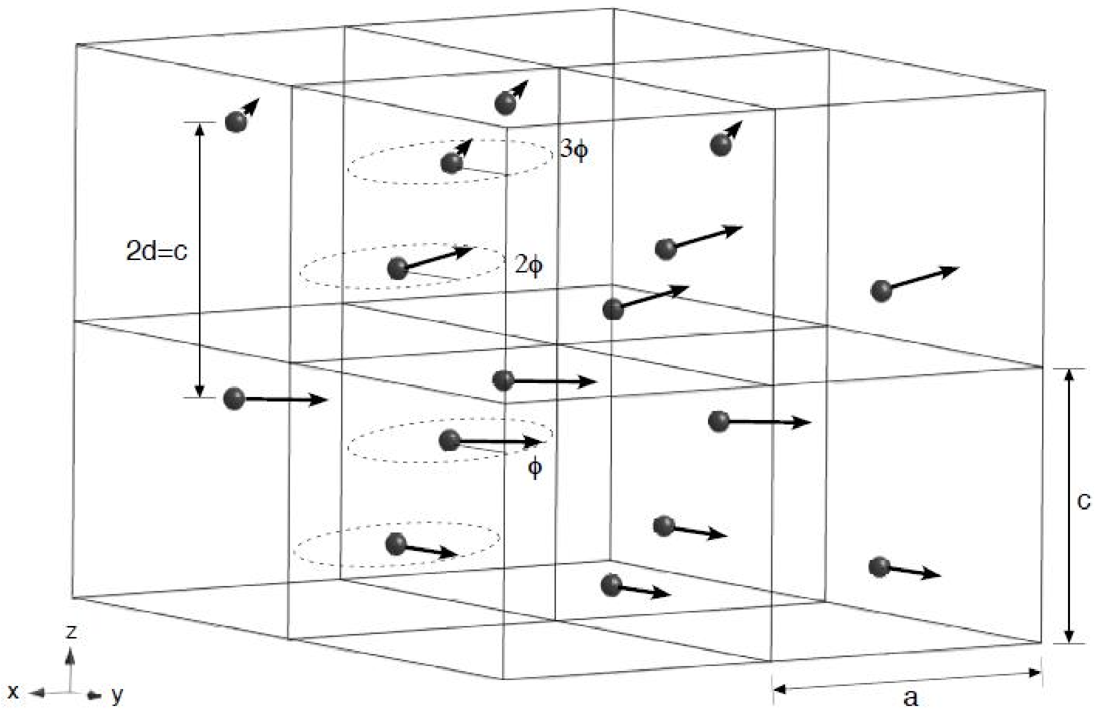




Magnetic Bragg peaks at half integer position

Spiral Magnetic Structures

Holmium below T=133K



$$\langle \hat{S}_l^x \rangle = \langle \hat{S} \rangle \cos(\boldsymbol{P}^* \cdot \boldsymbol{l})$$

$$\langle \hat{S}_l^y \rangle = \langle \hat{S} \rangle \sin(\boldsymbol{P}^* \cdot \boldsymbol{l})$$

$$\langle \hat{S}_l^z \rangle = 0$$

with $\boldsymbol{P}^* = 2\pi/\boldsymbol{P}$

Spin rotate by an angle Φ between adjacent lattice planes

Define propagation vector along spiral axis $\boldsymbol{P} = \frac{2\pi}{\phi} d\, \boldsymbol{e}_z$

Length of spiral can be commensurate (e.g. 3c or 5c) or incommensurate

Spiral Magnetic Structures

$$\langle \hat{S}_l^x \rangle = \langle \hat{S} \rangle \cos(\boldsymbol{P}^* \cdot \boldsymbol{l})$$

$$\langle \hat{S}_l^y \rangle = \langle \hat{S} \rangle \sin(\boldsymbol{P}^* \cdot \boldsymbol{l})$$

$$\langle \hat{S}_l^z \rangle = 0$$

- Insert into master formula
- Spins along x in thr basal plane (l=0)

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = (\gamma r_0)^2 e^{-2W(\boldsymbol{Q})} F^2(\boldsymbol{Q}) \sum_{\boldsymbol{l}} e^{\imath \boldsymbol{Q}\cdot\boldsymbol{l}} \langle \hat{S} \rangle^2 \times \left[\left(1 - \left(\frac{Q_x}{Q} \right)^2 \right) \cos(\boldsymbol{P}^* \cdot \boldsymbol{l}) - \frac{Q_x Q_y}{Q^2} \sin(\boldsymbol{P}^* \cdot \boldsymbol{l}) \right]$$

Rewrite cos as exp. function

vanishes

- Use lattice sum identity

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \frac{N}{2} \frac{(2\pi)^3}{v_0} (\gamma r_0)^2 e^{-2W(\boldsymbol{Q})} F^2(\boldsymbol{Q}) \langle \hat{S} \rangle^2 \left(1 - \left(\frac{Q_x}{Q} \right)^2 \right) \times \sum_{\boldsymbol{\tau}} \left(\delta(\boldsymbol{Q} + \boldsymbol{P}^* - \boldsymbol{\tau}) + \delta(\boldsymbol{Q} - \boldsymbol{P}^* - \boldsymbol{\tau}) \right)$$

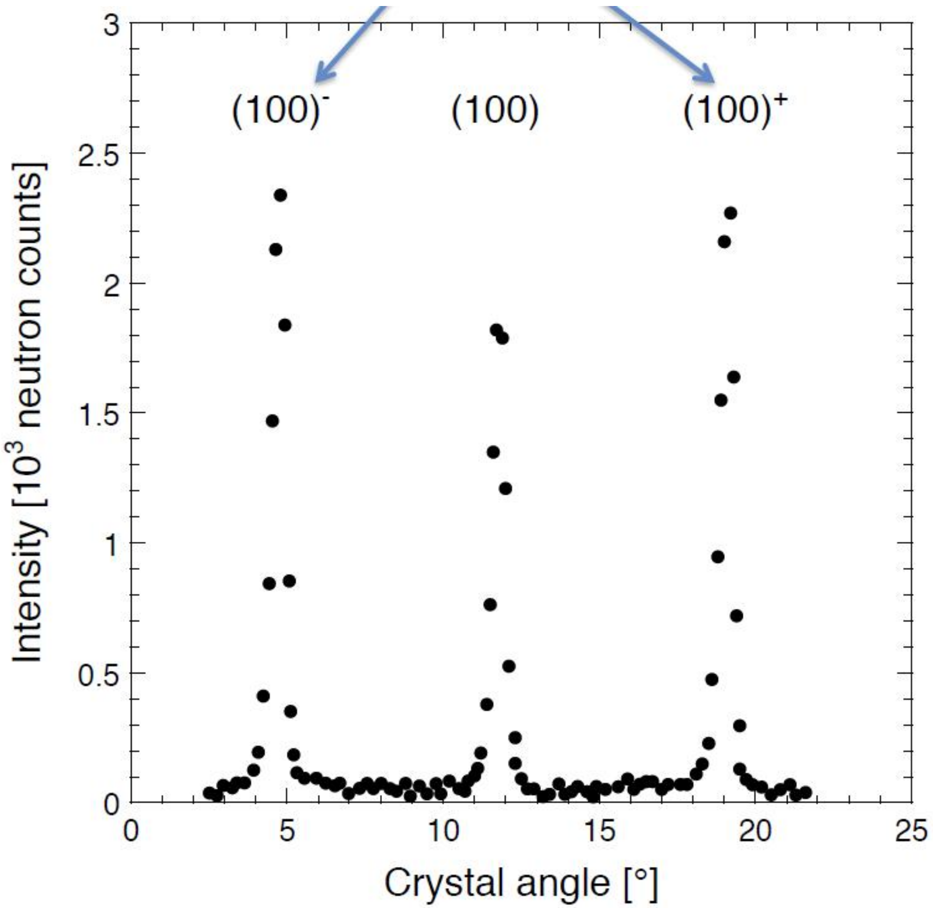
Intensity at (in)commensurate peaks which flank the nucear Bragg peak

Spiral Magnetic Structures

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \frac{N}{2}\frac{(2\pi)^3}{v_0}(\gamma r_0)^2 e^{-2W(\boldsymbol{Q})} F^2(\boldsymbol{Q})\langle\hat{S}\rangle^2 \left(1 - \left(\frac{Q_x}{Q}\right)^2\right) \times \sum_{\boldsymbol{\tau}} \left(\delta(\boldsymbol{Q} + \boldsymbol{P}^* - \boldsymbol{\tau}) + \delta(\boldsymbol{Q} - \boldsymbol{P}^* - \boldsymbol{\tau})\right)$$

Intensity at (in)commensurate peaks which flank the nucear Bragg peak

incommensurate “satellite” reflections

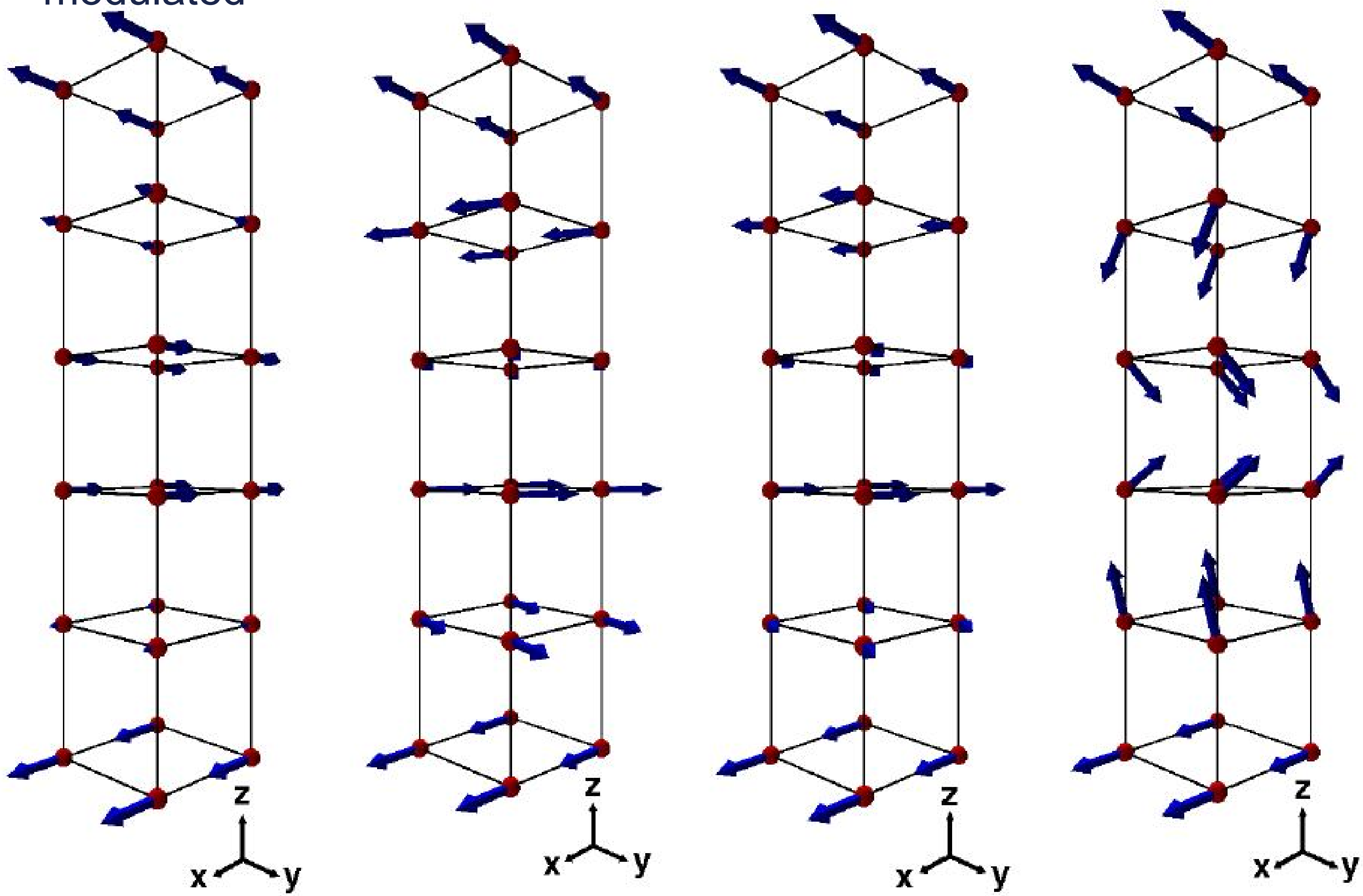


Holmium at T=77 K
(Koehler et al., 1966)

Spiral Magnetic Structures

There is a zoo of incommensurate structures:

Amplitude modulated | Circlular helix | Elliptic helix | Cycloid



- And of course the corresponding AF representation of each
- Typically generated by frustration or Dzyaloshinsky-Moriya interaction

Example: $Ba_2CuGe_2O_7$

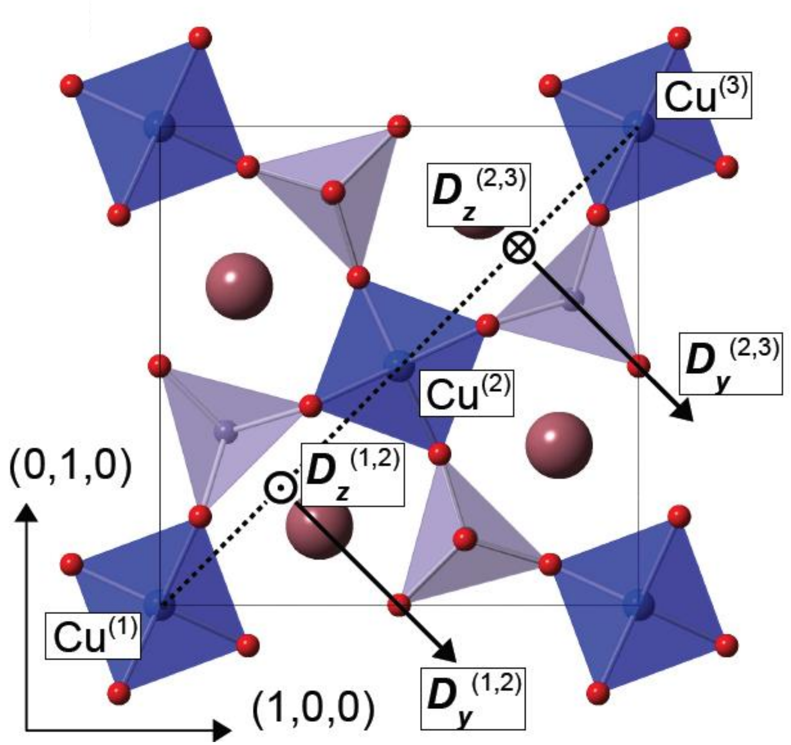
Non-centrosymmetric tetragonal AF, space group $P42_1m$

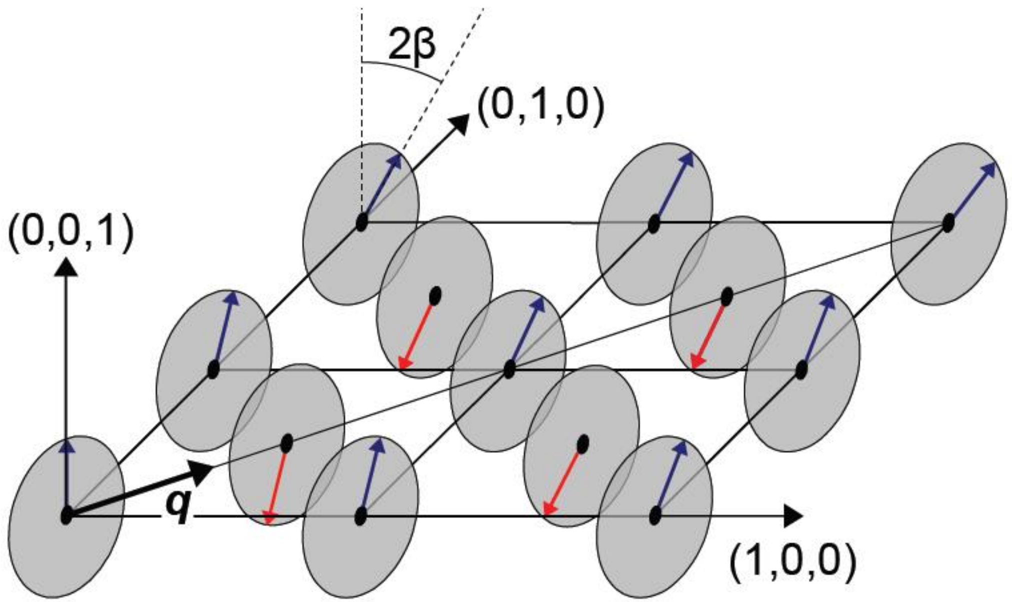
$H=J(\boldsymbol{S}_1\cdot\boldsymbol{S}_2)+\boldsymbol{D}\cdot(\boldsymbol{S}_1\times\boldsymbol{S}_2)$

Two components of Dzyaloshinskii-Moriya vector $\boldsymbol{D}_z$ and $\boldsymbol{D}_y$

T_N=3.2K, S=1/2

Almost AF planar cycloid (DM), pitch 200 Å



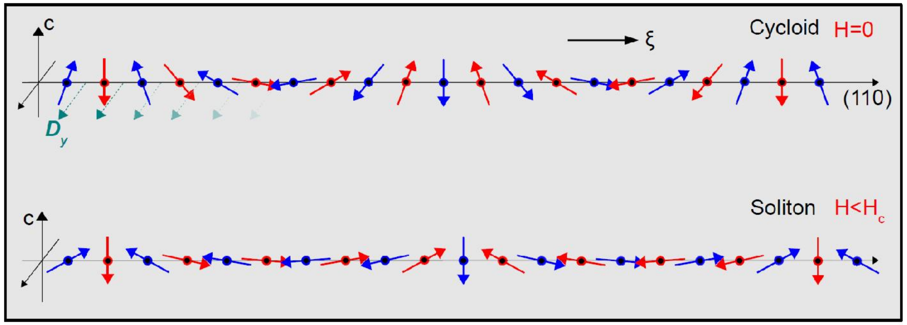


A. Zheludev et al., *Phys. Rev. B,* **57**, 2971 (1998)

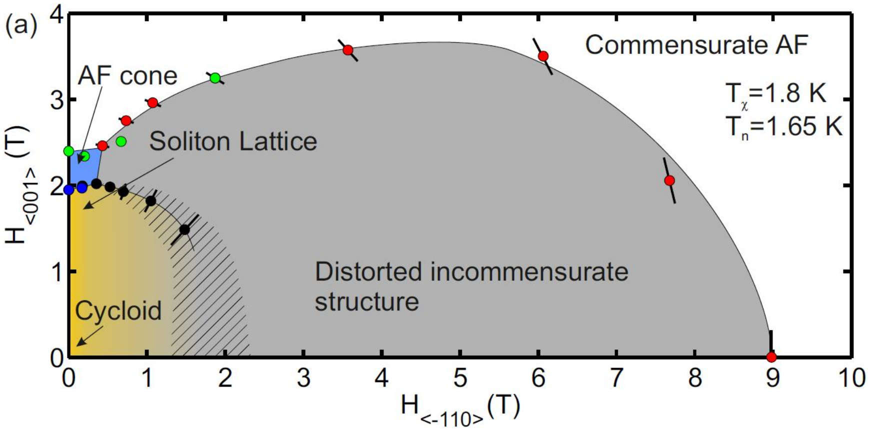
A. Zheludev et al., *Phys. Rev. B,* **59**, 11432 (1999)

Example: $Ba_2CuGe_2O_7$

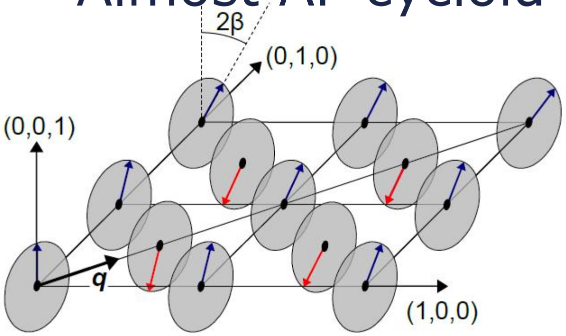
Cycloid to soliton distortion



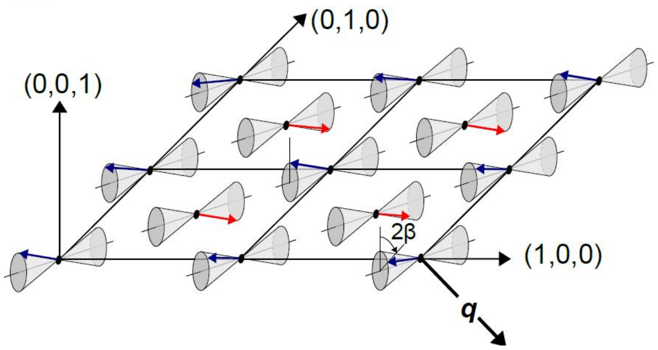
Phase diagram



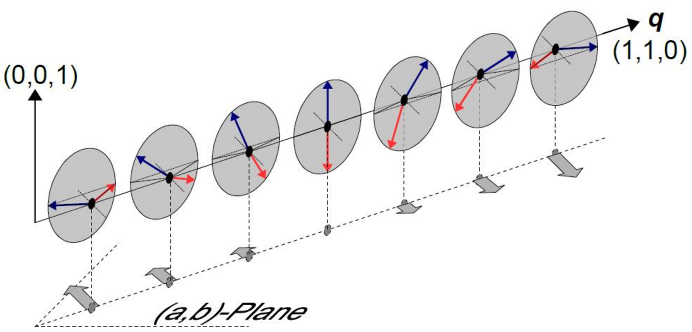
Almost AF cycloid



AF-cone



Influence of $\boldsymbol{D_z}$



S. Mühlbauer *et al.*, PRB **84** 180406(R) (2011)
S. Mühlbauer et al., PRB in press (2012), arXiv.1203:3650

Zero Field Magnetization

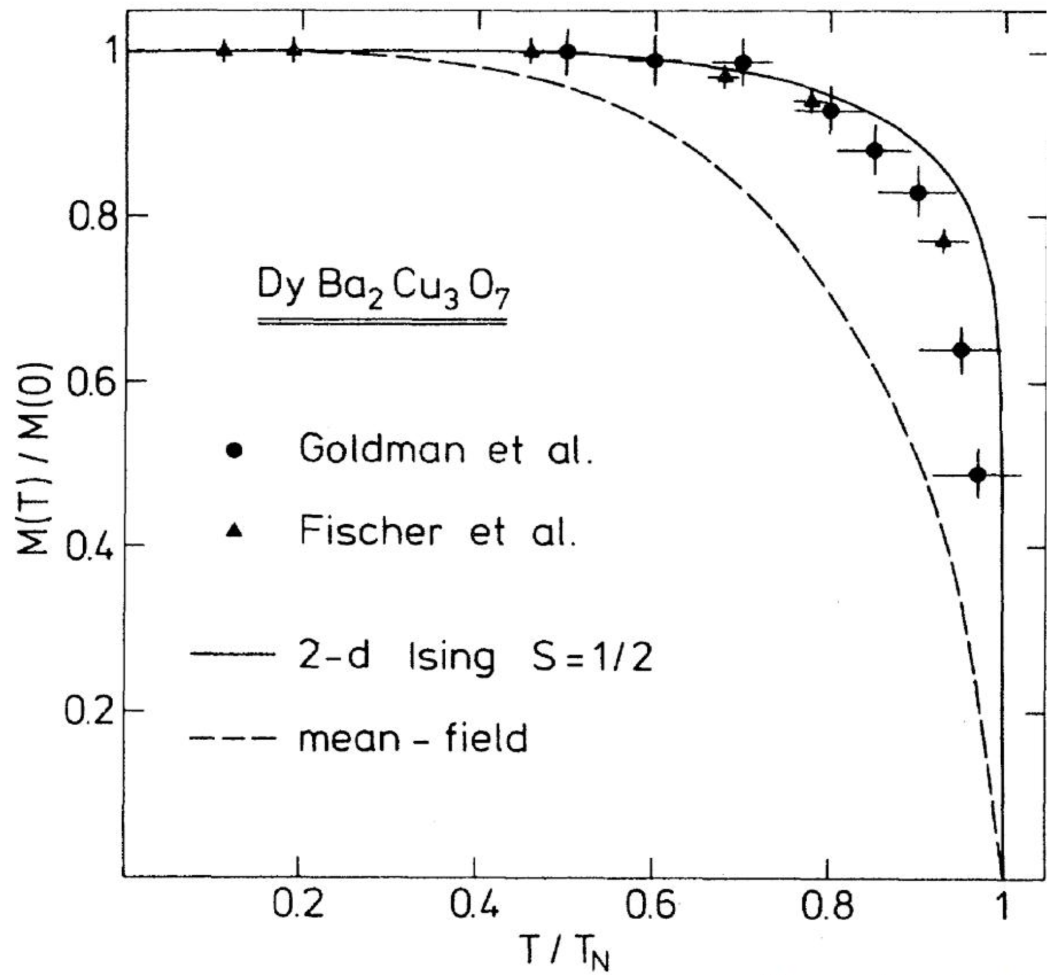
Magnetic intensity proportional to square of expectation value of spin operator $\langle \hat{S}^2 \rangle$

Magnetic neutron diffraction probes moments perpendicular to Q

Related to magnetic moment

$$\boldsymbol{\mu} = g\mu_B \langle \hat{\boldsymbol{S}} \rangle$$

Magnetic neutron diffraction probes the absolute moment and its direction



Allenspach et al., 1989