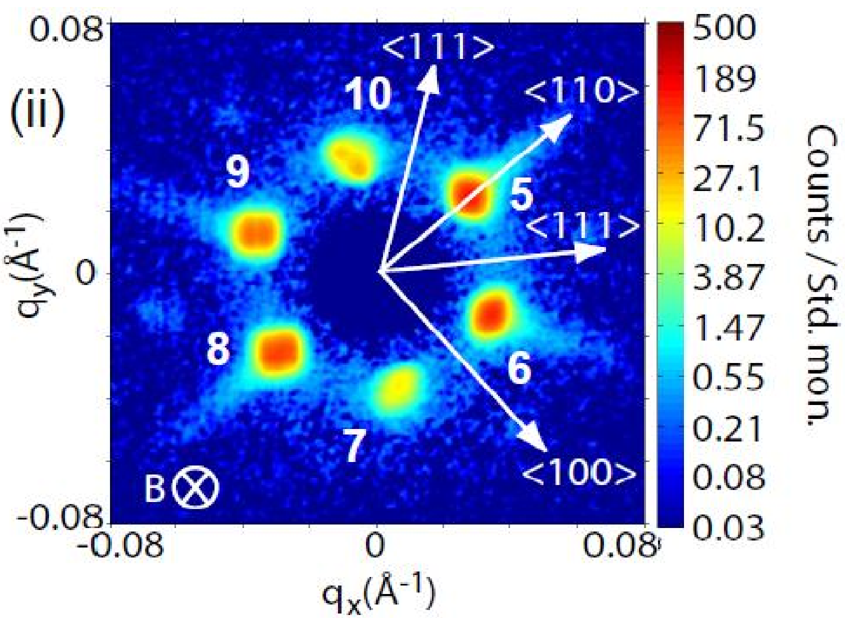
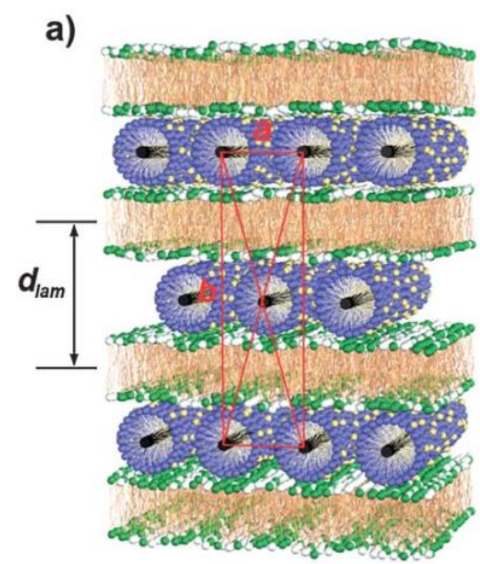
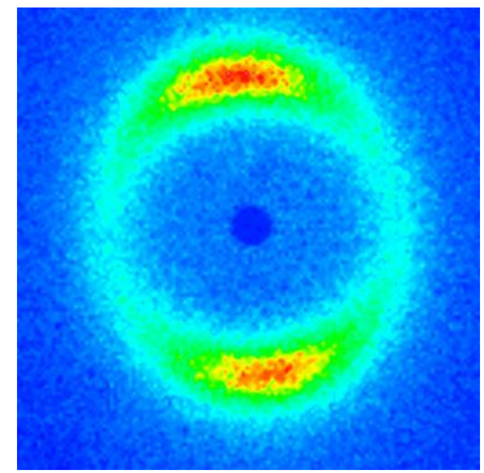
FRM II
Forschungs-Neutronenquelle
Heinz Maier-Leibnitz

MLZ
Heinz Maier-Leibnitz Zentrum

Physics with Neutrons II, SS 2016







Lecture 10, 4.7.2016

MLZ is a cooperation between:

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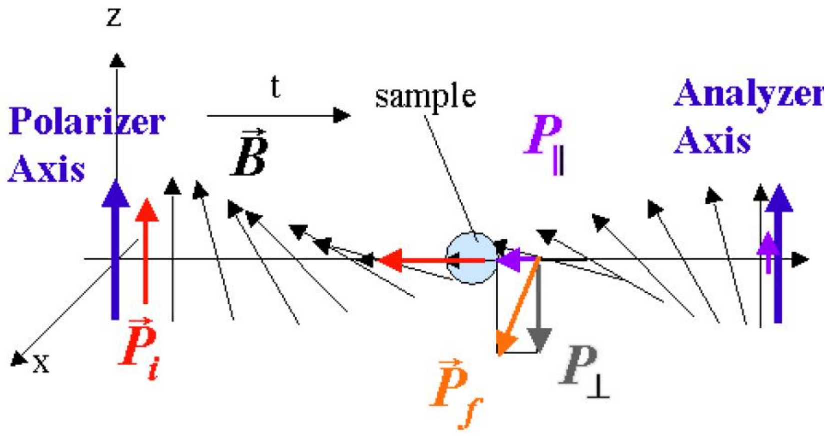
Plan for SS 2016

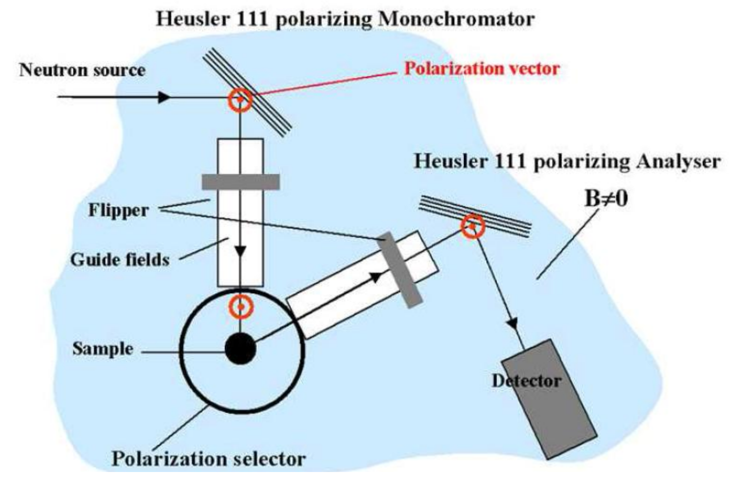
- VL1: Repetition of winter term, basic neutron scattering theory
- VL2: SANS, theory and applications
- VL3: Neutron optics
- VL4: Reflectometry and dynamical scattering theory
- VL5: Diffuse neutron scattering
- VL6: Magnetic scattering cross section
- VL7: Magnetic structures and structure analysis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

Reminder:
Polarized Neutrons &
3D Polarimetry

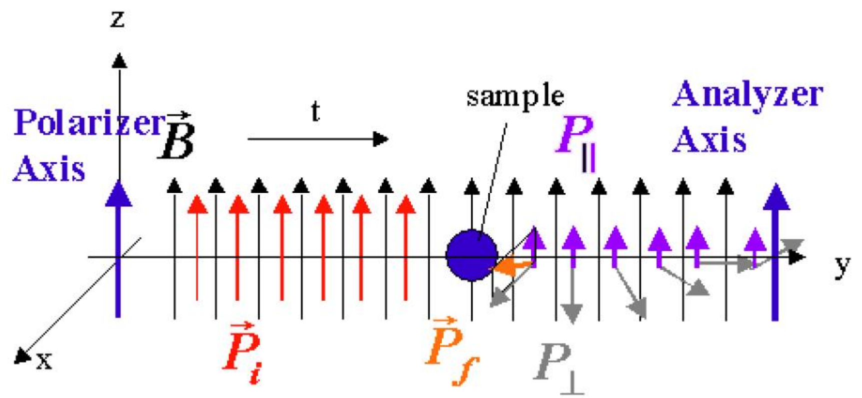
Uniaxial polarization analysis

Adiabatic rotation of polarization allows to measure three components (x,y,z)





Assume that scattering process at the sample rotates the polarization



- Spin precesses in guide field
- Only component parallel to z is measured

Information is lost!

Pol. effects: Summary

	Non spin flip	Spin flip	Polarization dependent
Nuclear coherent	1	0	no
Nuclear incoherent spin (single isotope)	1/3	2/3	no
Nuclear incoherent isotope (I=0)	1	0	no
Paramagnetic scattering	$1/2(1-Q^2)$	$1/2(1-Q^2)$	no
FM, collinear, P perp. to Q, M ||Q	1 nuclear coh. + magnetic (b+p vs. b-p)	0 nuclear incoh.	yes
FM, non-collinear, P perp. to Q	<1 nuclear coh. + magnetic	>0 nuclear incoh. + magnetic	yes
FM, collinear, P||Q, M perp. to Q	Useless configuration, external field problem, No magnetic signal nuclear coh.	0 nuclear incoh.	no
AF, collinear, P||Q M perp. to Q	Only nuclear for P perp. to M	<1	yes
AF, non-collinar	<1	>0	yes

Neutron polarization: Things to consider

- Reflectivity of polarizing monochromators is weak (have to be kept single domain). The polarized intensity typically amounts 20-30% of the unpolarized beam.
- Guide field necessary, difficulties with magnetic field at the sample region
- All flippers / polarizers /analyzers have finite efficiency! Corrections needed, difficult for small signals due to leakage from one channel to the other. ^{3}He analyzers are time-dependent.
- Four channels instead of one need to be measured!
- Only the projection of the polarization on the quantization axis (z-axis, guide field) can be measured

3D polarimetry: Theory

Polarization is a vector that may turn at the scattering process!

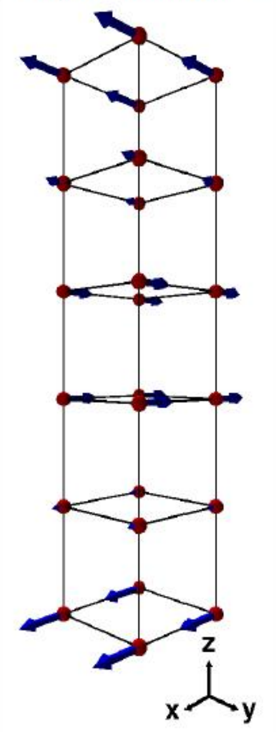
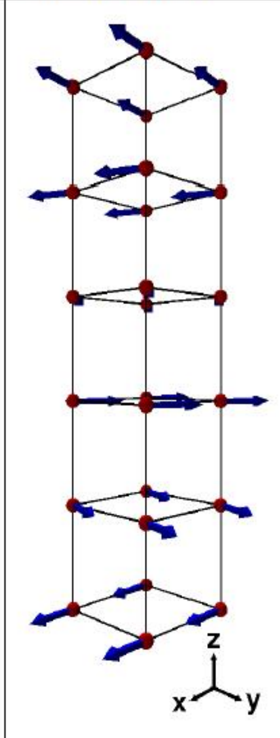
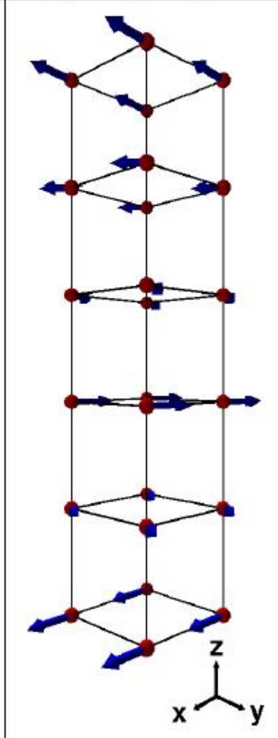
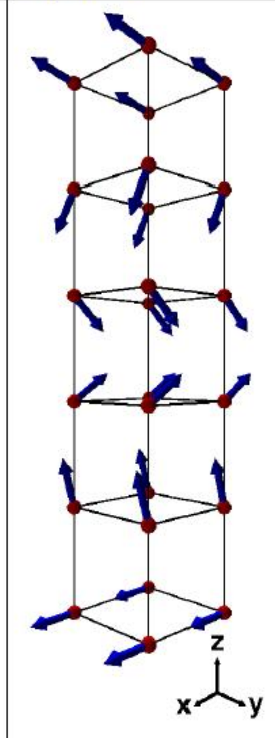
Rewrite as tensor! $\boldsymbol{P}' = \tilde{P}\boldsymbol{P}_0 + \boldsymbol{P}''$

Final polarization — Rotation of polarization — Generation of polarization

$$\sigma \tilde{\boldsymbol{P}} = \begin{pmatrix} (N-M^y-M^z) & iI^z & -iI^y \\ -iI^z & (N+M^y-M^z) & M_{mix} \\ iI^y & M_{mix} & (N-M^y+M^z) \end{pmatrix} \qquad \sigma \boldsymbol{P}'' = \begin{pmatrix} C \\ R^y \\ R^z \end{pmatrix}$$

Item	correlation functions	description
N	$\frac{k_f}{k_i}\langle N_{\boldsymbol{Q}} N^{\dagger}_{\boldsymbol{Q}}\rangle_\omega$	nuclear contribution
$M^{y/z}$	$(\gamma r_0)^2 \frac{k_f}{k_i}\langle M^{y/z}_{\perp\boldsymbol{Q}} M^{\dagger y/z}_{\perp\boldsymbol{Q}}\rangle_\omega$	y- and z-components of the magnetic contribution.
$R^{y/z}$	$(\gamma r_0)\frac{k_f}{k_i}\langle N^{\dagger}_{\boldsymbol{Q}} M^{y/z}_{\perp\boldsymbol{Q}}\rangle_\omega + \langle M^{\dagger y/z}_{\perp\boldsymbol{Q}} N_{\boldsymbol{Q}}\rangle_\omega$	real parts of the nuclear-magnetic interference term.
$I^{y/z}$	$(\gamma r_0)\frac{k_f}{k_i}\langle N^{\dagger}_{\boldsymbol{Q}} M^{y/z}_{\perp\boldsymbol{Q}}\rangle_\omega - \langle M^{\dagger y/z}_{\perp\boldsymbol{Q}} N_{\boldsymbol{Q}}\rangle_\omega$	imaginary parts of the nuclear-magnetic interference term.
C	$i(\gamma r_0)^2\frac{k_f}{k_i}(\langle M^{y}_{\perp\boldsymbol{Q}} M^{\dagger z}_{\perp\boldsymbol{Q}}\rangle_\omega - \langle M^{z}_{\perp\boldsymbol{Q}} M^{\dagger y}_{\perp\boldsymbol{Q}}\rangle_\omega)$	chiral contribution
M_{mix}	$(\gamma r_0)^2\frac{k_f}{k_i}(\langle M^{y}_{\perp\boldsymbol{Q}} M^{\dagger z}_{\perp\boldsymbol{Q}}\rangle_\omega + \langle M^{z}_{\perp\boldsymbol{Q}} M^{\dagger y}_{\perp\boldsymbol{Q}}\rangle_\omega)$	mixed magnetic contribution or magnetic-magnetic interference term

3D polarimetry: Example

Name	Ampl. modulated	Circular helix	Elliptical helix	Cycloid
Figure				

Different types of incommensurate spin "spirals"

Can be distinguished only using 3D polarimetry!

$\boldsymbol{Q}_+ = (0,0,1+k)$				
$\boldsymbol{S^k}$	$\frac{a\hat{e}_x}{2}$	$\frac{(a\hat{e}_x - ia\hat{e}_y)}{2}$	$\frac{(a\hat{e}_x - ib\hat{e}_y)}{2}$	$\frac{(a\hat{e}_x - ia\hat{e}_z)}{2}$
$\boldsymbol{M_{\perp Q}}$	$(0, \frac{a}{2}, 0)$	$(0, \frac{a}{2}, -\frac{ia}{2})$	$(0, \frac{a}{2}, -\frac{ib}{2})$	$(0, \frac{a}{2}, 0)$
M^y	$(\gamma r_0)^2 \frac{a^2}{4}$	$(\gamma r_0)^2 \frac{a^2}{4}$	$(\gamma r_0)^2 \frac{a^2}{4}$	$(\gamma r_0)^2 \frac{a^2}{4}$
M^z	0	$(\gamma r_0)^2 \frac{a^2}{4}$	$(\gamma r_0)^2 \frac{b^2}{4}$	0
M^{mix}	0	0	0	0
C	0	$-(\gamma r_0)^2 \frac{a^2}{2}$	$-(\gamma r_0)^2 \frac{ab}{2}$	0
$\frac{d\sigma}{d\Omega}$	$(\gamma r_0)^2 \frac{a^2}{4}$	$(\gamma r_0)^2 \frac{a^2}{2}(1 + P_0^x)$	$(\gamma r_0)^2 (\frac{a^2+b^2}{4} + P_0^x \frac{ab}{2})$	$(\gamma r_0)^2 \frac{a^2}{4}$
P_{ij}	$\begin{pmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix}$	$\begin{pmatrix} -1 & 0 & 0 \\ -1 & 0 & 0 \\ -1 & 0 & 0 \end{pmatrix}$	pol. tensor provided in table 2.3	$\begin{pmatrix} -1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix}$

	$\boldsymbol{Q}_- = (0,0,1+k)$
P_{ij}	$\begin{pmatrix} \frac{-(\alpha^2+1)P_0^x - 2\alpha}{\alpha^2+1+2\alpha P_0^x} & 0 & 0 \\ -\frac{2\alpha}{\alpha^2+1} & \frac{\alpha^2-1}{\alpha^2+1} & 0 \\ -\frac{2\alpha}{\alpha^2+1} & 0 & \frac{1-\alpha^2}{\alpha^2+1} \end{pmatrix}$

3D polarimetry: Setup

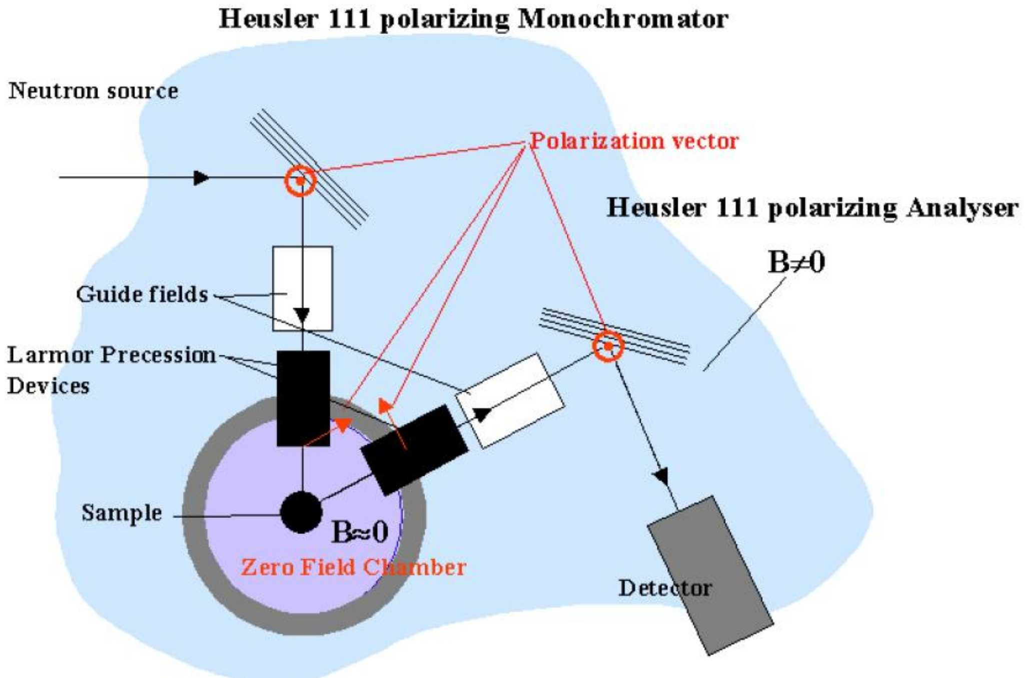
Mu-pad of Cryopad:

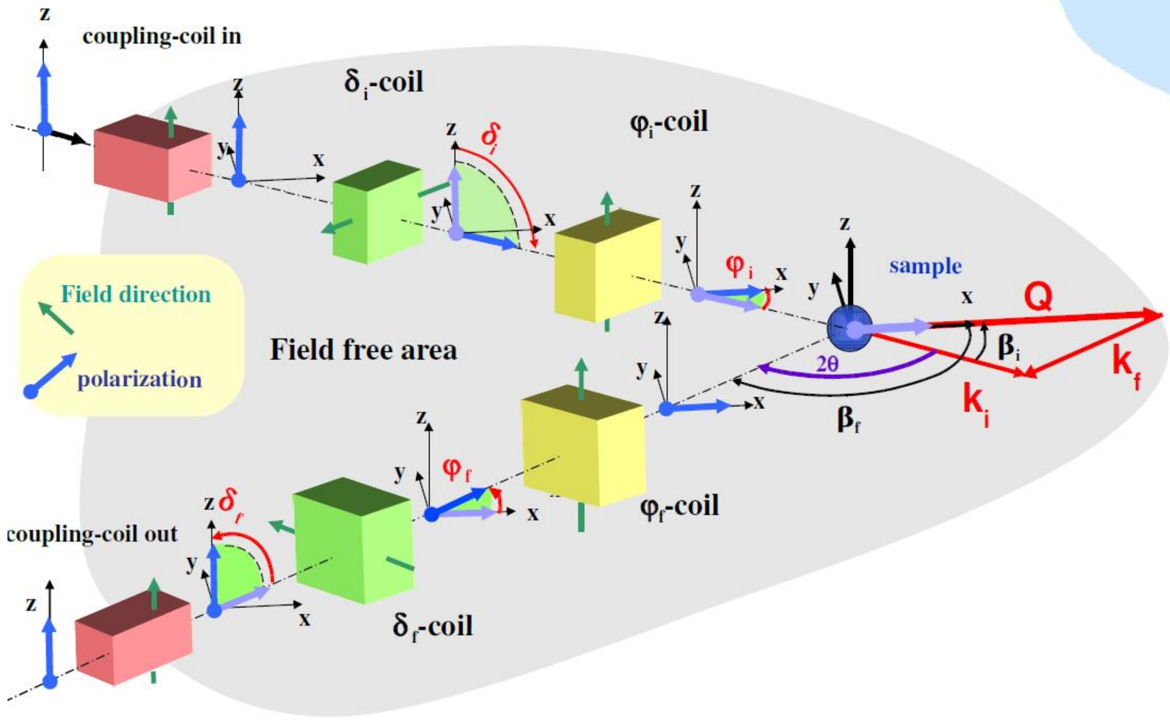
Mu-metal or Meissner shield:

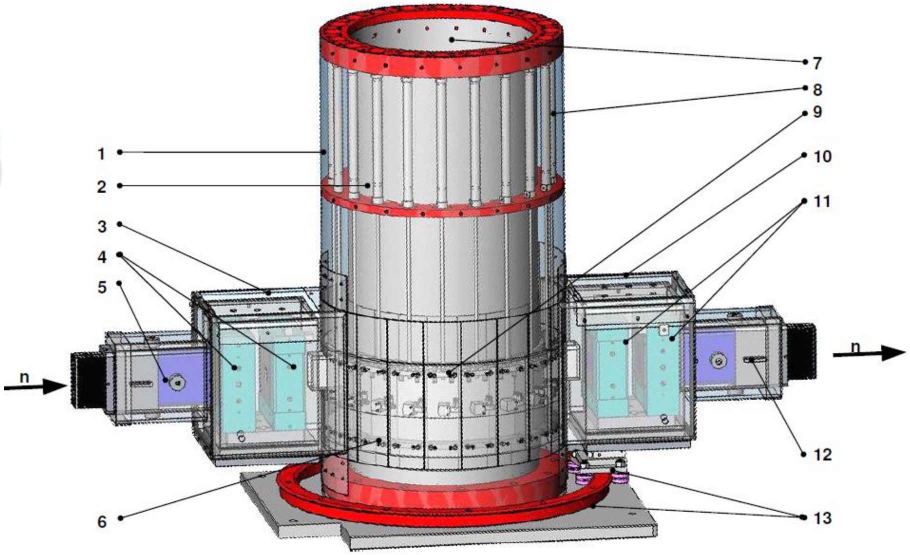
- Zero field chamber to maintain spin direction

Spin precession devices:

- Turn the spin adiabatically to the desired direction and back to the analyzer axis.







3D polarimetry: Summary

Uni-axial polarization analysis vs. 3D polarimetry

Uni-axial polarization analysis

$$\check{P} = \begin{pmatrix} \check{P}_{xx} & & \\ & \check{P}_{yy} & \\ & & \check{P}_{zz} \end{pmatrix}$$

Measurement of the diagonal terms only

Slow, but helps solving most typical magnetic structures

Magnetic field at the sample is possible

Complicated due to leakage caused by finite efficiencies of flipper and analyzer

3D polarimetry

$$\mathrm{P}_{ij} = (P_{i0}\tilde{P}_{ji} + P''_j)/|\boldsymbol{P}_0|$$

Measurement of the 9 elements of the polarization matrix

Even slower, but helps solving almost all magnetic structures

Complicated for FM samples

Complicated due to leakage caused by finite efficiencies of flipper and analyzer

No magnetic field at the sample allowed

This is the last measurement to be done!

Magnetic diffraction

Measurement strategy for magnetic diffraction

Identification of magnetic peaks:
- ⇨ Strong temperature dependence close to T_c
- ⇨ Polarization factor
- ⇨ Q-dependence (form factor)

Solving the magnetic structure
- ⇨ Use symmetry arguments and crystal symmetry!
- ⇨ Use field to align domains/magnetization, use different polarization factors

If more than one structure remains
- ⇨ Use classical uniaxial polarization analysis, measure flipping ratio $(b+p)^2$, $(b-p)^2$,

If more than one structure remains
- ⇨ Use 3-D polarimetry

Spin waves: Magnetic excitations seen by neutrons

Spin waves

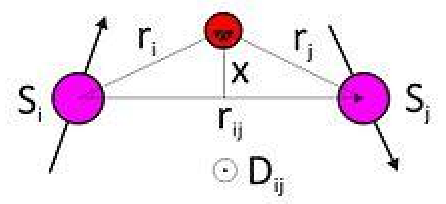
Magnetic interactions for spins on a (Bravais) lattice:

Exchange interaction | Single ion anisotropy | Zeemann + anisotropic g-factor

$$\hat{H}_{spin} = \sum_{j \neq j'} \sum_{\alpha\beta} J_{jj'}^{(\alpha\beta)} \hat{S}_j^{(\alpha)} \hat{S}_{j'}^{(\beta)} + \sum_{j\alpha} D_{j\alpha} \left(\hat{S}_j^{(\alpha)} \right)^2 - \sum_{j\alpha\beta} \mu_B H^{(\beta)} g_j^{(\alpha\beta)} \hat{S}_j^{(\alpha)}$$

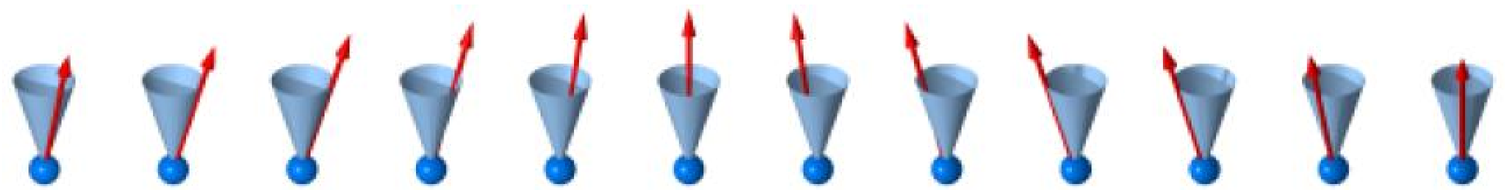
Dzyaloshinskii-Moriya (no inversion on bond j, j')

$$\hat{H}_{DM} = \sum_{j \neq j'} \boldsymbol{D}_{jj'} \left(\hat{\boldsymbol{S}}_j \times \hat{\boldsymbol{S}}_{j'} \right)$$



Dipolar interactions

$$\hat{H}_D = \sum_{j \neq j'} \frac{(g\mu_B)^2}{r_{jj'}^3} \left[\hat{\boldsymbol{S}}_j \hat{\boldsymbol{S}}_{j'} - 3 \left(\hat{\boldsymbol{S}}_j \hat{\boldsymbol{r}}_{jj'} \right) \left(\hat{\boldsymbol{S}}_{j'} \hat{\boldsymbol{r}}_{jj'} \right) \right]$$

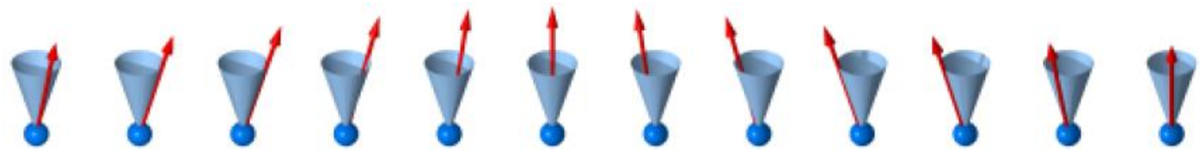


Spin waves

Spin waves (physical picture)

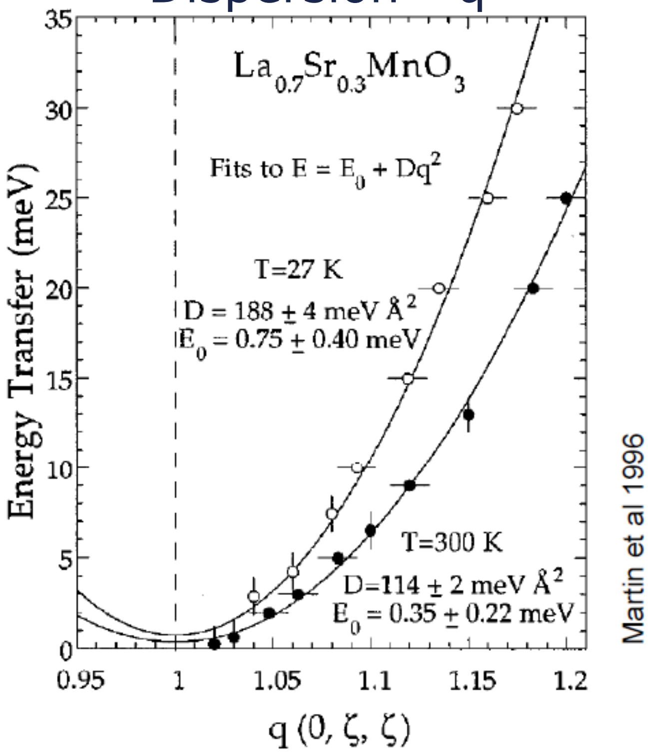
Collective excitation of the order parameter

Involves oscillation of spin components transverse to the ordered moment

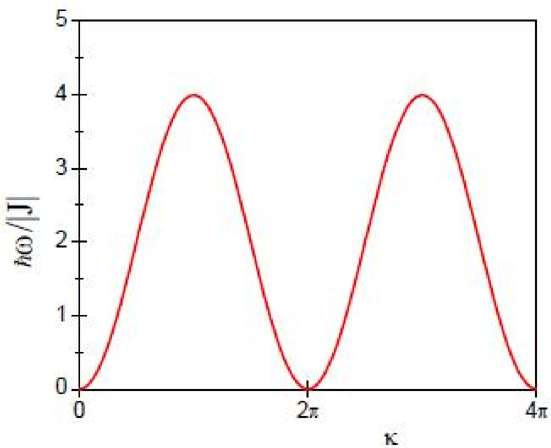


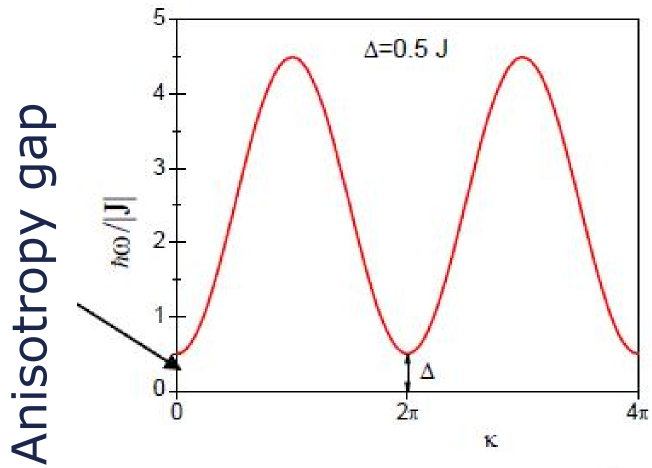
Quantized excitation: Magnon (spin wave) ⟺ phonon

Dispersion ~q²



Example: linear chain Heisenberg FM





$$\hat{H} = -|J| \sum_j \hat{S}_j \hat{S}_{j+1}$$

$$\hbar\omega = 2|J|S\left[1-\cos(\kappa a)\right]$$

$$\hat{H} = -|J| \sum_j \hat{S}_j \hat{S}_{j+1} - \Delta \sum_j \left(S_j^z\right)^2$$

$$\hbar\omega = 2|J|S\left[1-\cos(\kappa a)\right] + \Delta$$

Spin waves (magnon)

Spin waves: How to treat them by neutrons

Similar to Phonons: Use QM picture with raising and lowering operator

Bose-Einstein occupany $\langle n_{\boldsymbol{q}} \rangle = \left(\exp\left(\frac{\hbar\omega(\boldsymbol{q})}{k_B T} \right) - 1 \right)^{-1}$

Inelastic cross-section for <u>single magnon</u> scattering (FM)

Polarization factor

Form factor

DW factor

$$\left(\frac{d^2\sigma}{d\Omega\, dE'} \right) = (\gamma r_0)^2 \frac{k'}{k} \frac{(2\pi)^3}{V_0} \frac{1}{2} S\left(1 + \hat{q}_z^2\right) \left| \frac{1}{2} g F(\boldsymbol{q}) \right|^2 e^{-2W_q} \times$$

$$\times \sum_{\boldsymbol{\tau},\boldsymbol{\kappa}} \left[\delta(\boldsymbol{\kappa} - \boldsymbol{q} - \boldsymbol{\tau}) \delta(\hbar\omega_\kappa - \hbar\omega) \langle\langle n_\kappa + 1 \rangle\rangle + \delta(\boldsymbol{\kappa} + \boldsymbol{q} - \boldsymbol{\tau}) \delta(\hbar\omega_\kappa + \hbar\omega) \langle\langle n_\kappa \rangle\rangle \right]$$

Magnon creation

Magnon annihilation

Spin waves

Ferromagnetic spin waves (magnons)

Considering only nearest neighbour interactions on a cubic lattice

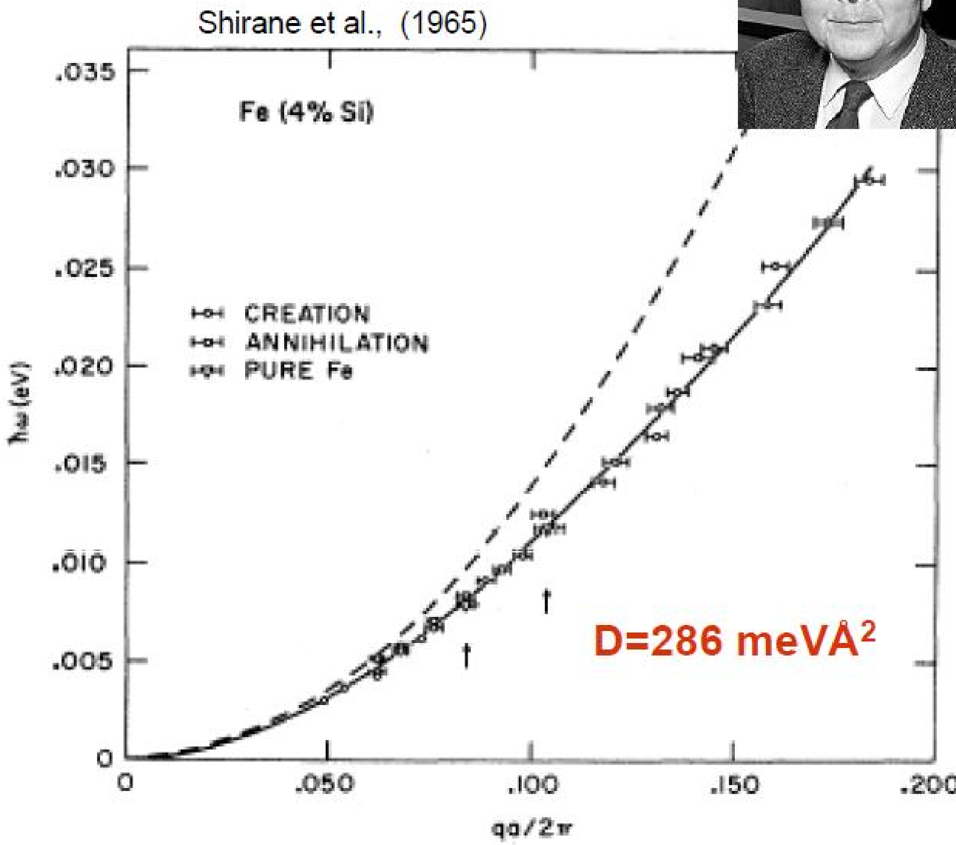
$$J(\boldsymbol{\kappa}) = -|J|\sum_{\rho} e^{i\kappa\rho} = -n|J|\gamma(\boldsymbol{\kappa}) \qquad \gamma(\boldsymbol{\kappa}) = \frac{1}{n}\sum_{\rho} e^{i\kappa\rho} \approx 1 - \frac{1}{2}(a\kappa)^2$$

Number of nn
cubic: n=6

$$\hbar\omega_{\kappa} = 2nS|J|\left[1-\gamma(\boldsymbol{\kappa})\right] \underset{\kappa\to 0}{\approx} 2|J|Sna^2\kappa^2$$

$$\boxed{\hbar\omega_{\kappa} \underset{\kappa\to 0}{\approx} D\kappa^2 \qquad D = 2|J|Sna^2}$$

Spin stiffness



Neutrons directly measure the spin wave stiffness/dispersion

Spin waves

Antiferromagnetic (AF) spin waves (magnons)

Again, split up into <u>two ferromagnetic</u> sublattices

$$\hat{H} = \underbrace{\sum_{\alpha}\sum_{l,m} J^{\alpha}_{lm}\, \hat{S}^{(\alpha)}_{l}\hat{S}^{(\alpha)}_{m}}_{\text{Inter-sublattice interactions}} + \underbrace{\sum_{\alpha}\sum_{l,l'} J'^{\alpha}_{ll'}\, \hat{S}^{(\alpha)}_{l}\hat{S}^{(\alpha)}_{l'}}_{\text{Interactions on the "red" sublattice}} + \underbrace{\sum_{\alpha}\sum_{m,m'} J'^{\alpha}_{mm'}\, \hat{S}^{(\alpha)}_{m}\hat{S}^{(\alpha)}_{m'}}_{\text{Interactions on the "blue" sublattice}}$$

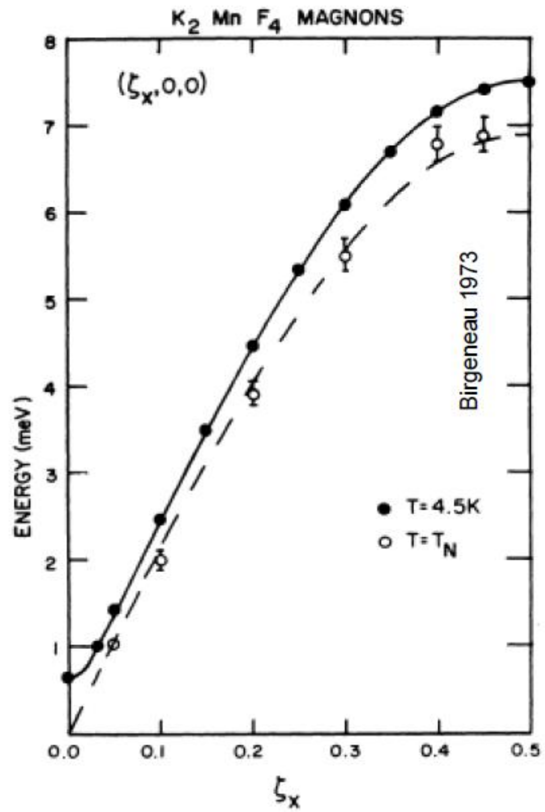
With groundstates $\hat{S}^{z}_{l}\left|0\right\rangle = S\left|0\right\rangle$ $\quad \hat{S}^{z}_{m}\left|0\right\rangle = -S\left|0\right\rangle$

Heisenberg AF

$$\boxed{(\hbar\omega_{\boldsymbol{\kappa}})^2 = 4S^2\left(\left[J(0)\right]^2 - \left[J(\boldsymbol{\kappa})\right]^2\right)}$$

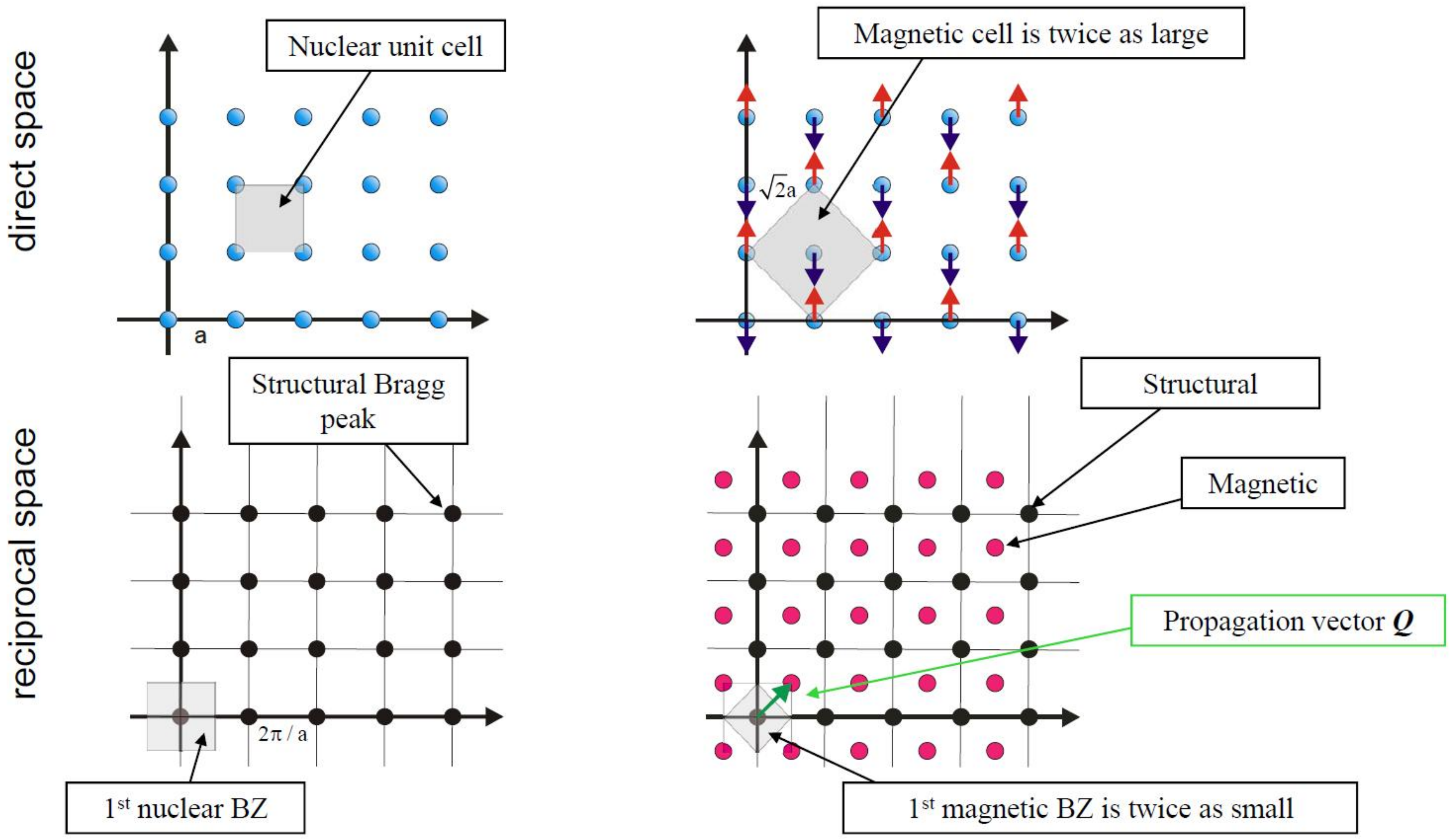
Nearest neighbour interactions on a cubic lattice (AF)

$$\boxed{\hbar\omega_{\boldsymbol{\kappa}} \underset{\kappa\to 0}{\approx} v\kappa \qquad \underbrace{v}_{\text{Spin wave velocity}} = 2JSa}$$



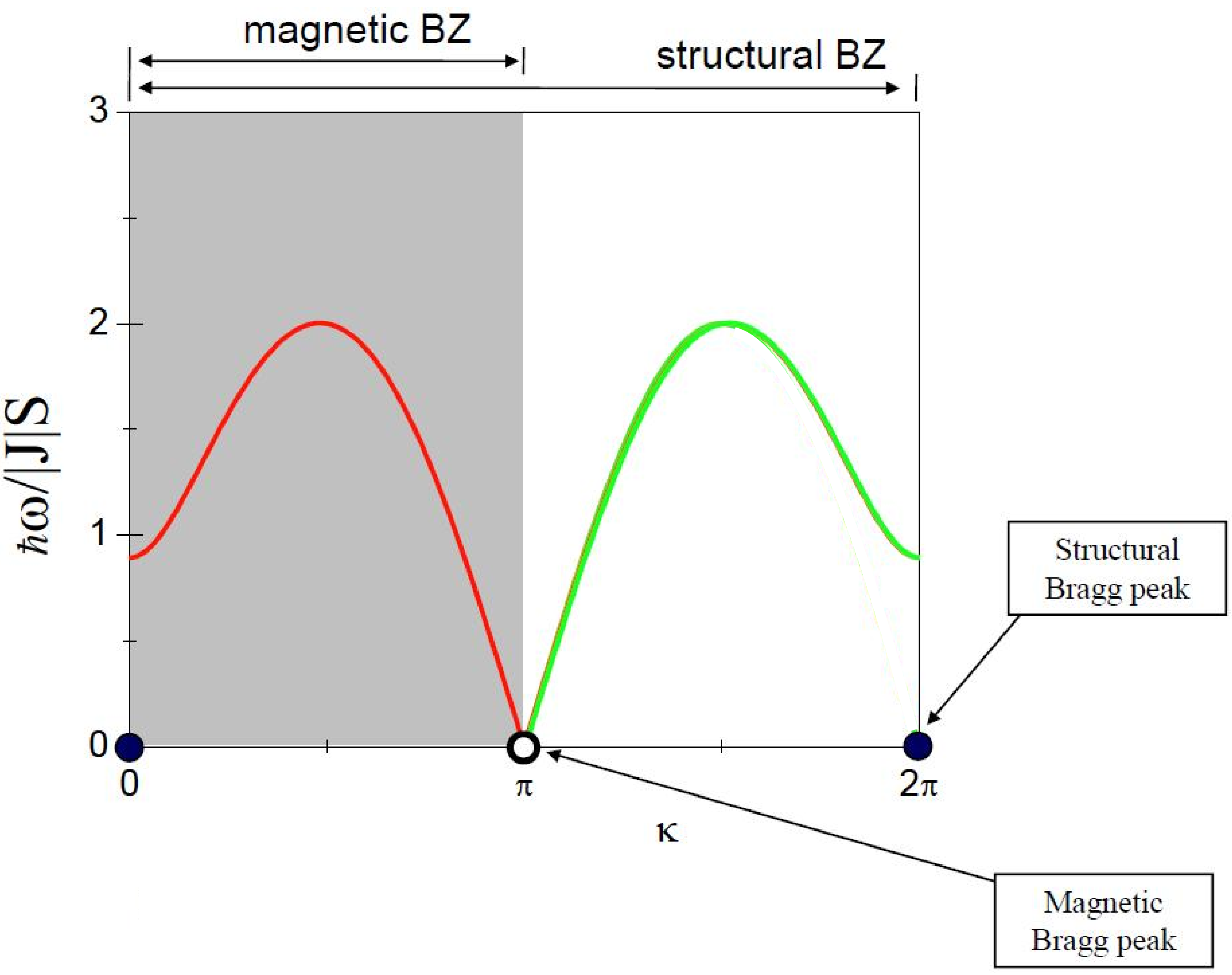
Spin waves

Remember unit cell doubling for AF structures!



Spin waves

Example: linear chain AF



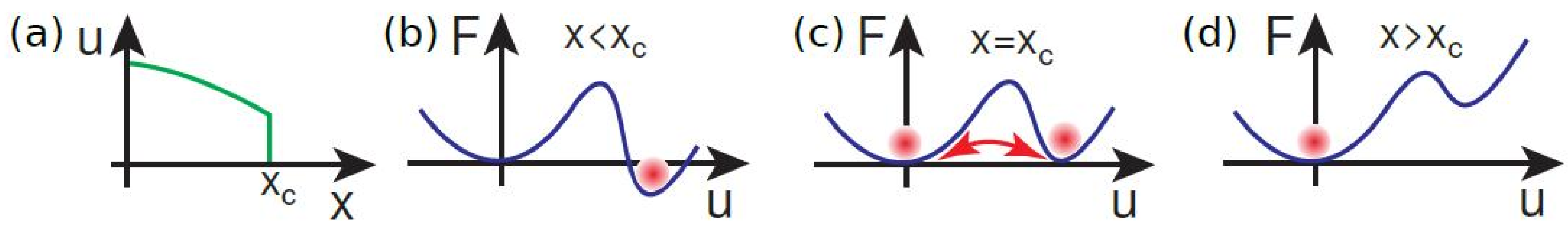
Phase Transitions: Seen by Neutrons

Phase Transitions

Define an order parameter which is zero in the disordered phase and finite in the ordered phase (magnetization, particle density...)

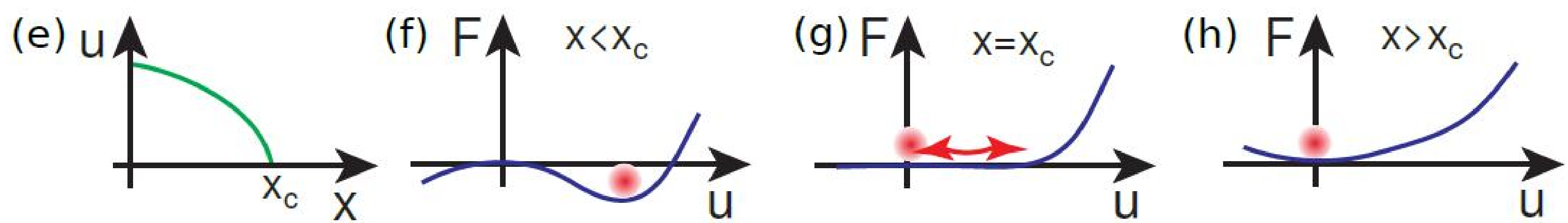
Discriminate two different kind of phase transitions (Ehrenfest classification):

first-order



Jump of the OP, accompagnied by latent heat
Transition from one local minima of the free energy to another

second-order

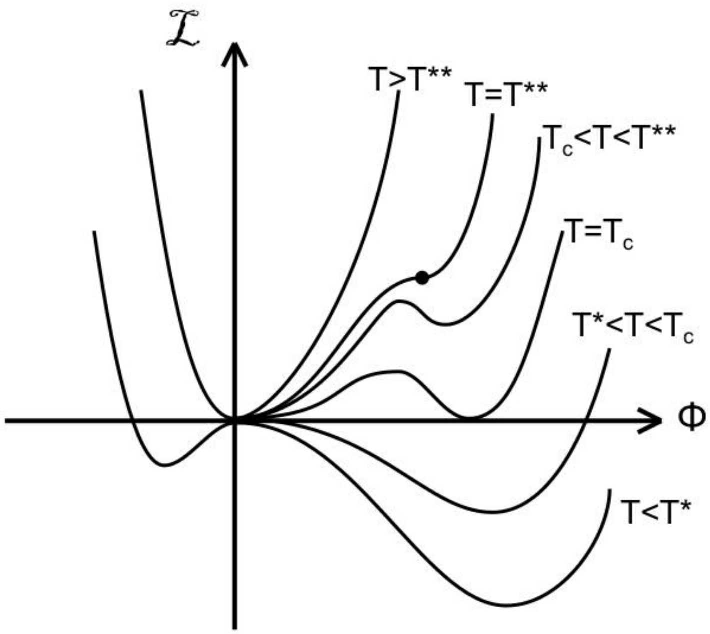


Continuous transition of the OP, no latent heat, jump of specific heat
Flat landscape of the free energy

<u>Abundance of critical fluctuations of the OP</u>
<u>Typically, Landau Theory/mean field theory is used</u>

Phase Transitions

Two major descriptions:



Landau theory:
Expansion of the OP close to T_c (OP small)

$$F = F_{n0} + \alpha|\psi|^2 + \frac{\beta}{2}|\psi|^4 + \frac{1}{2m^*}\left|\left(\frac{\hbar}{i}\nabla - \frac{e^*}{c}\mathbf{A}\right)\psi\right|^2 + \frac{h^2}{8\pi}$$

Mean field theory:
Complicated many body interactions (FM spins) simplifield to a mean field

System with n spins $\hat{H} = -\sum_j g\mu_b \hat{\vec{S}}_j \vec{B} - \sum_{i,j} J_{ji} \hat{\vec{S}}_j \hat{\vec{S}}_i$

Average spin: $\langle \hat{\vec{S}} \rangle = \frac{1}{N}\sum_{i=0}^{N} \hat{\vec{S}}_i$

Expectation value to each spin corresponds to $\langle \hat{\vec{S}} \rangle$

Hamiltonian $\boxed{\hat{H} = -\sum_j g\mu_b \hat{\vec{S}}_j \left(\vec{B} + \frac{1}{g\mu_b} J_j \langle \hat{\vec{S}} \rangle\right)}$

Phase Transitions

Critical fluctuations: Universal scaling laws (Kadanoff 1970's)

➡ Depend only on

Dimensionality of the OP (symmetry group)
Dimensionality of the system
Short range or long range interactions

➡ No microscopic details!

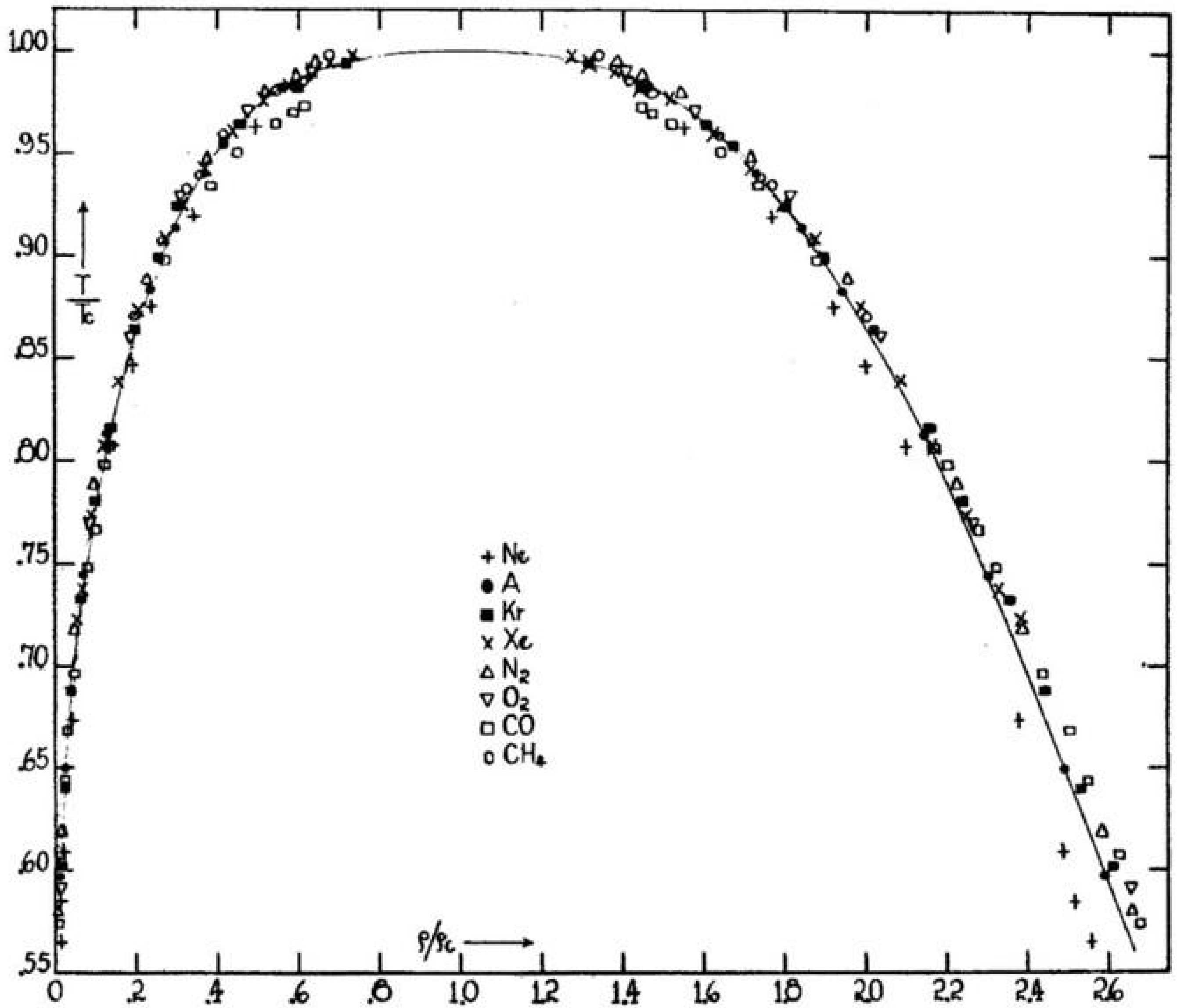
$$\tau = \frac{T - T_c}{T}$$

Quantity	T<T_c (ordered)	T>T_c (disordered)	T=T_c
Spec. heat	$\lvert \hbar\omega\beta \rvert \ll 1$	$C_V = \tau^{-\alpha}$	$C_V = \lvert h \rvert^{-\varepsilon}$
Order parameter	$\phi = \lvert \tau \rvert^{\beta}$		$\phi = \lvert h \rvert^{-\delta} \operatorname{sign}(h)$
Compressibility (susceptibility)	$\chi_T = \lvert \tau \rvert^{-\gamma'}$	$\chi_T = \tau^{-\gamma}$	
Correlation length	$\xi = \lvert \tau \rvert^{-\nu'}$	$\xi = \tau^{-\nu}$	$\xi = \lvert h \rvert^{-\mu}$
Correlation function			$G(r,0) = \lvert r \rvert^{-(d-2+\eta)}$

- Landau theory: $\alpha = 0 \quad \beta = \frac{1}{2} \quad \gamma = 1 \quad \delta = 3 \quad \varepsilon = 0 \quad \mu = \frac{1}{3} \quad \nu = \frac{1}{2} \quad \eta = 0$

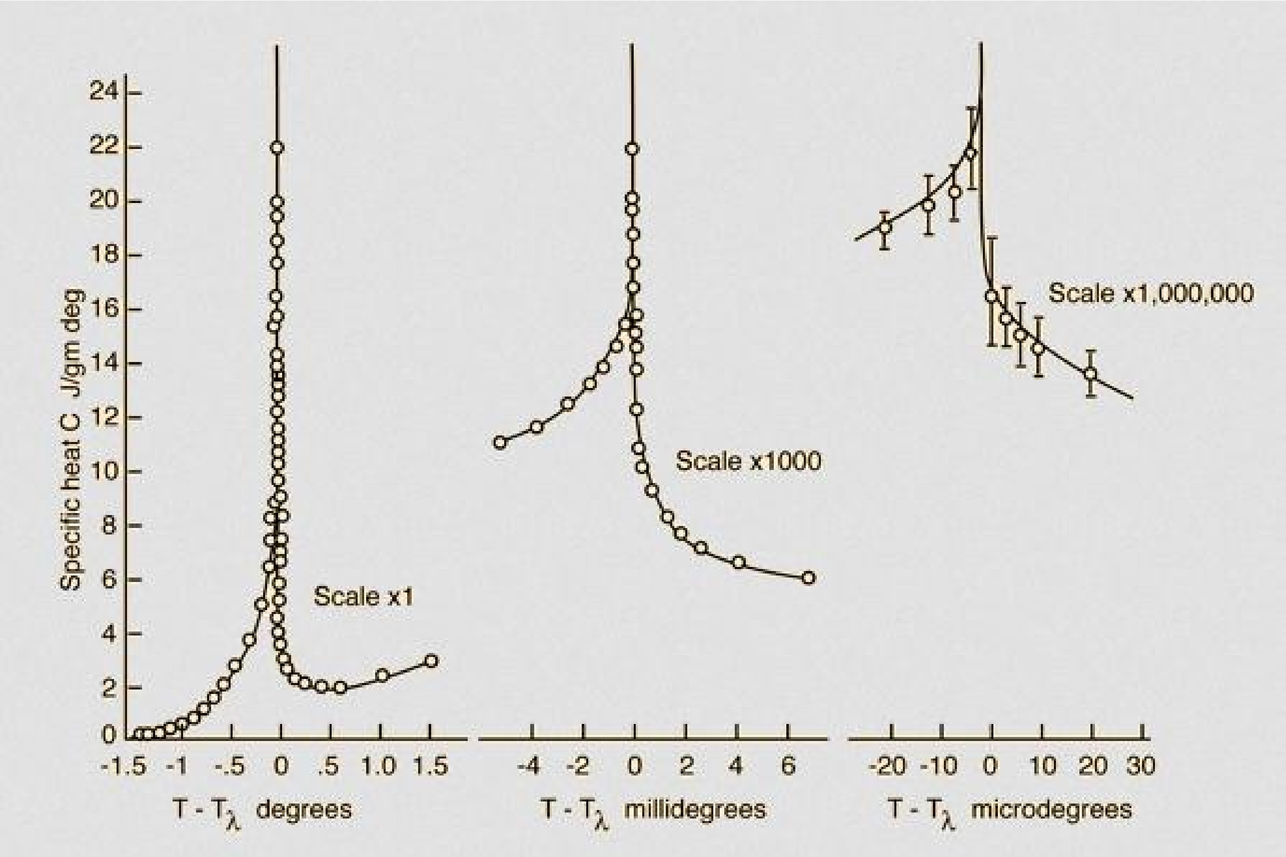
Phase Transitions

Example: Universal scaling in the liquid-gas transition for eight different liquids



Phase Transitions

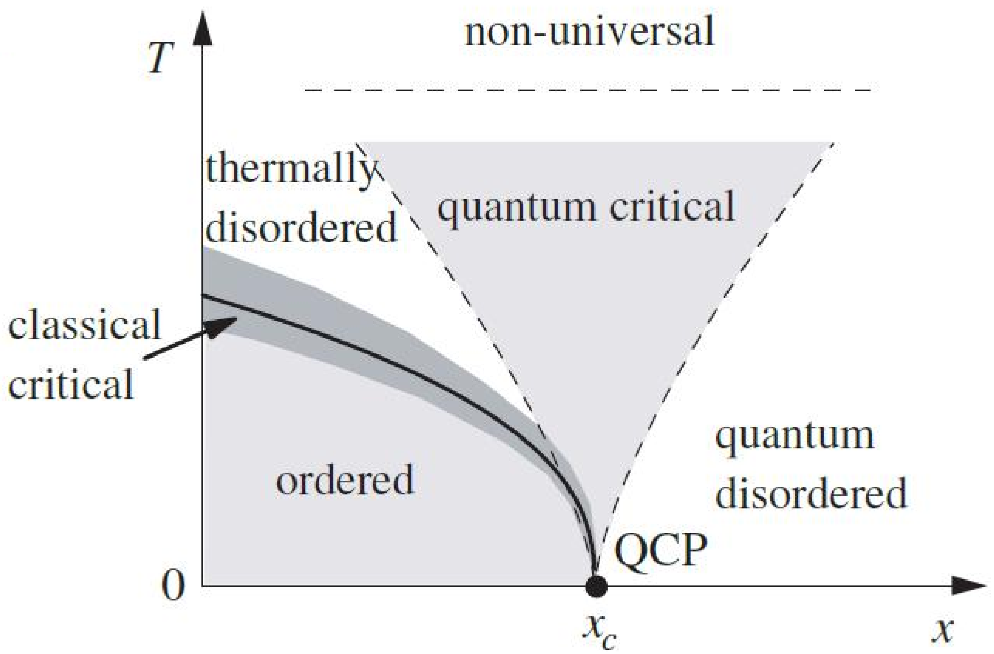
Example: Universal scaling of the lambda transition of Helium



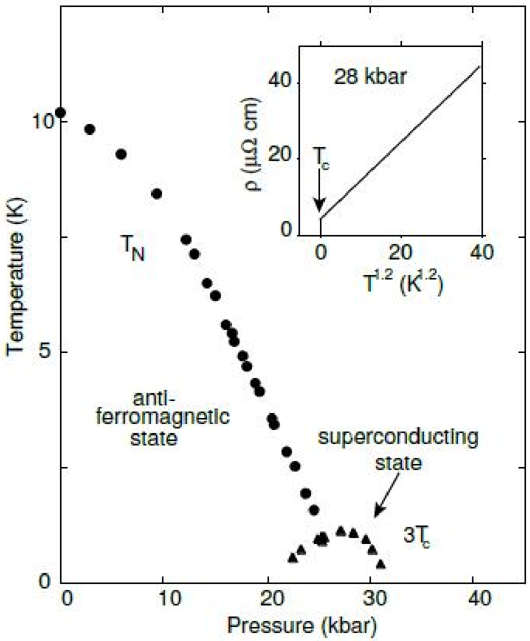
(Quantum)Phase Transitions

Phase transitions at T=0: Quantum phase transitions

Supression of a phase transition at finite T using an control parameter



Superconductivity
in$CePd_2Si_2$



Quantum fluctuations replace classical fluctuations

Often new physics emerges!

Phase Transitions

Why neutrons are perfect for investigations of (magnetic)phase transitions?

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

Neutrons directly measure generalized susceptibility tensor

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

Measurement of fluctuations as a function of momentum and energy transfer (space and time correlations)

2 examples from the neutron world:

- Ferromagnetic fluctuations in iron
- Brazovskii transition in the helimagnet MnSi

Critical Scattering Fe

Iron: Archetypal ferromagnet, model system to study fundamental properties of continuous phase transitions

Simple Heisenberg model fails to predict the dynamical scaling:

- Linewidth $\Gamma = Aq^z$ with critical exponent z and constant A
- Mismatch (in particular at low q) due to long range dipolar exchange + short range isotropo FM exchange

$\boxed{\text{Isotropic 3D Heisenberg: z=5/2} \\ \text{Dipolar interactions: z=2}}$

Hamiltonian $$H = \sum_q \left[-J_0 + Jq^2a^2 + Jg\frac{q^\alpha q^\beta}{q^2}\right] S^\alpha_{-\mathbf{q}} S^\beta_{\mathbf{q}},$$

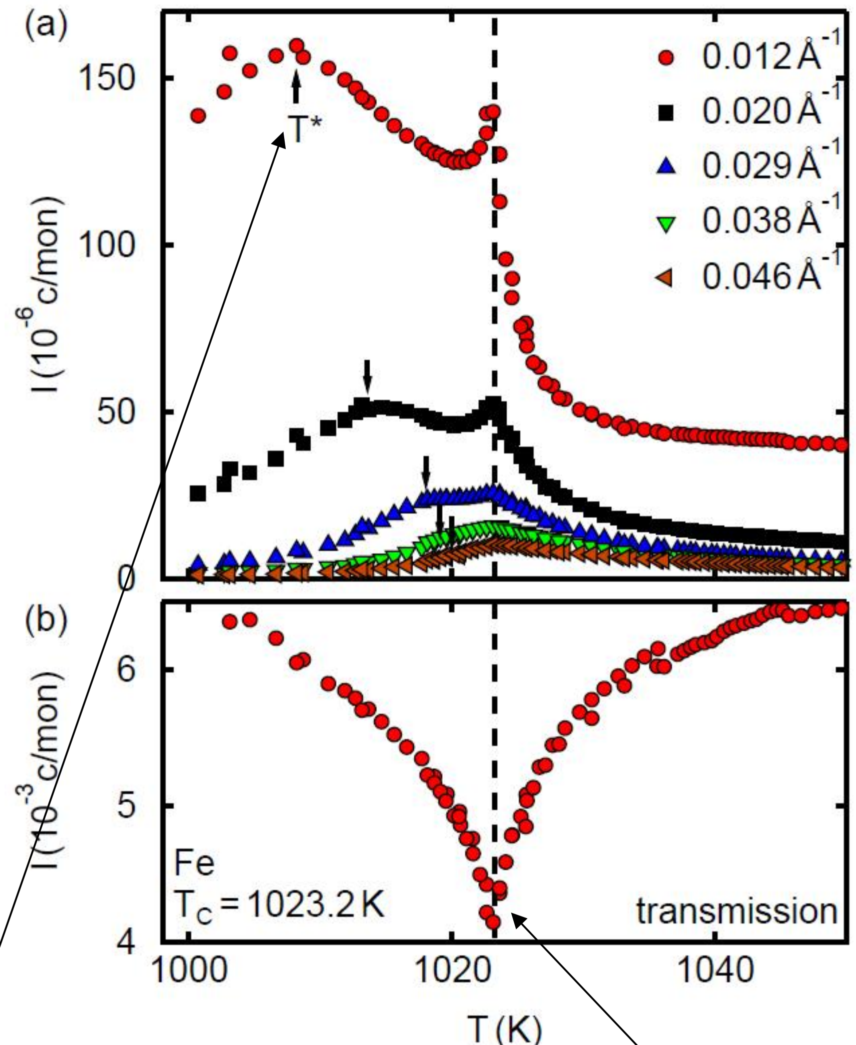
Susceptibility $$\chi_t = (\boldsymbol{q},T) = \frac{\Psi}{\kappa^2(T) + q^2} \text{ and } \chi_l = (\boldsymbol{q},T) = \frac{\Psi}{\kappa^2(T) + q^2 + q_D^2}$$

Linewidth of fluctuations $$\Gamma^\alpha(q,\kappa,g) = Aq^z\gamma^\alpha(x,y)$$ Measurement with NRSE

Critical Scattering Fe

RESEDA in MIEZE mode (SANS):
transverse fluctations around ***Q***=0

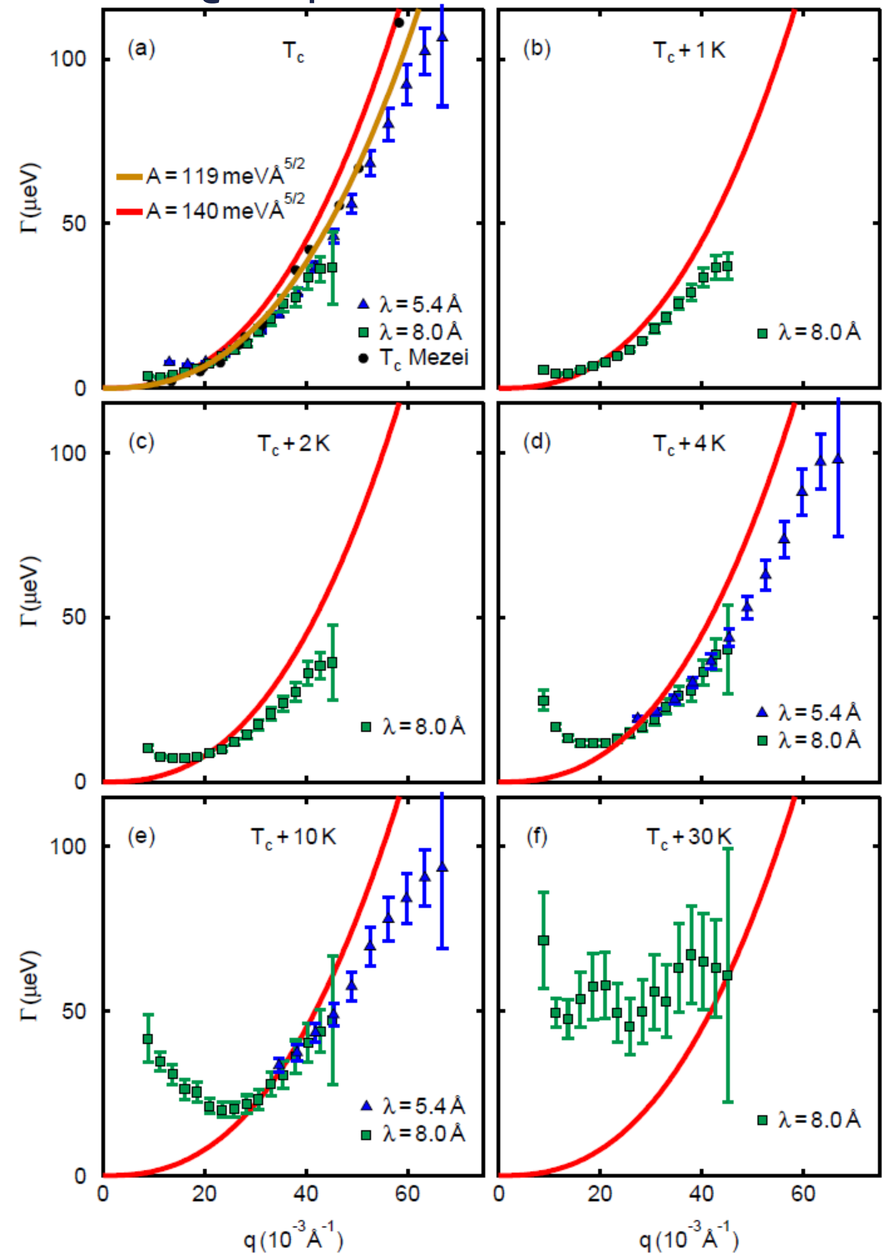
Critical scattering



FM domains

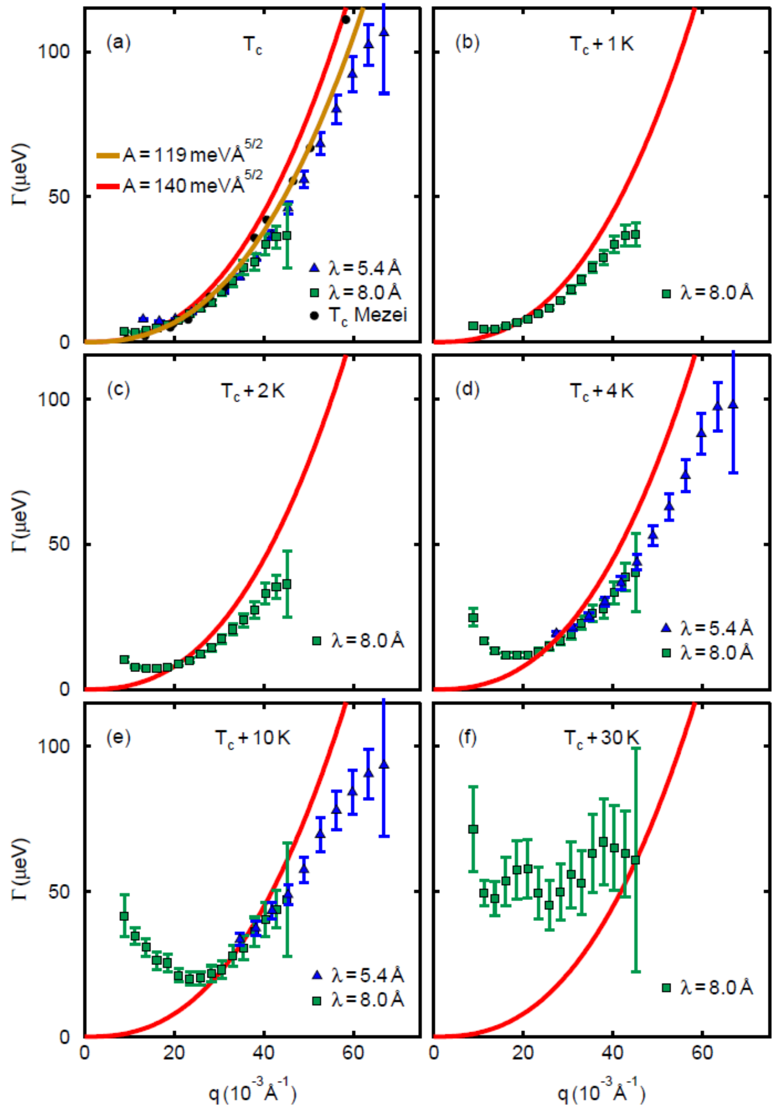
Critical opalescence

Q-dependence linewidth



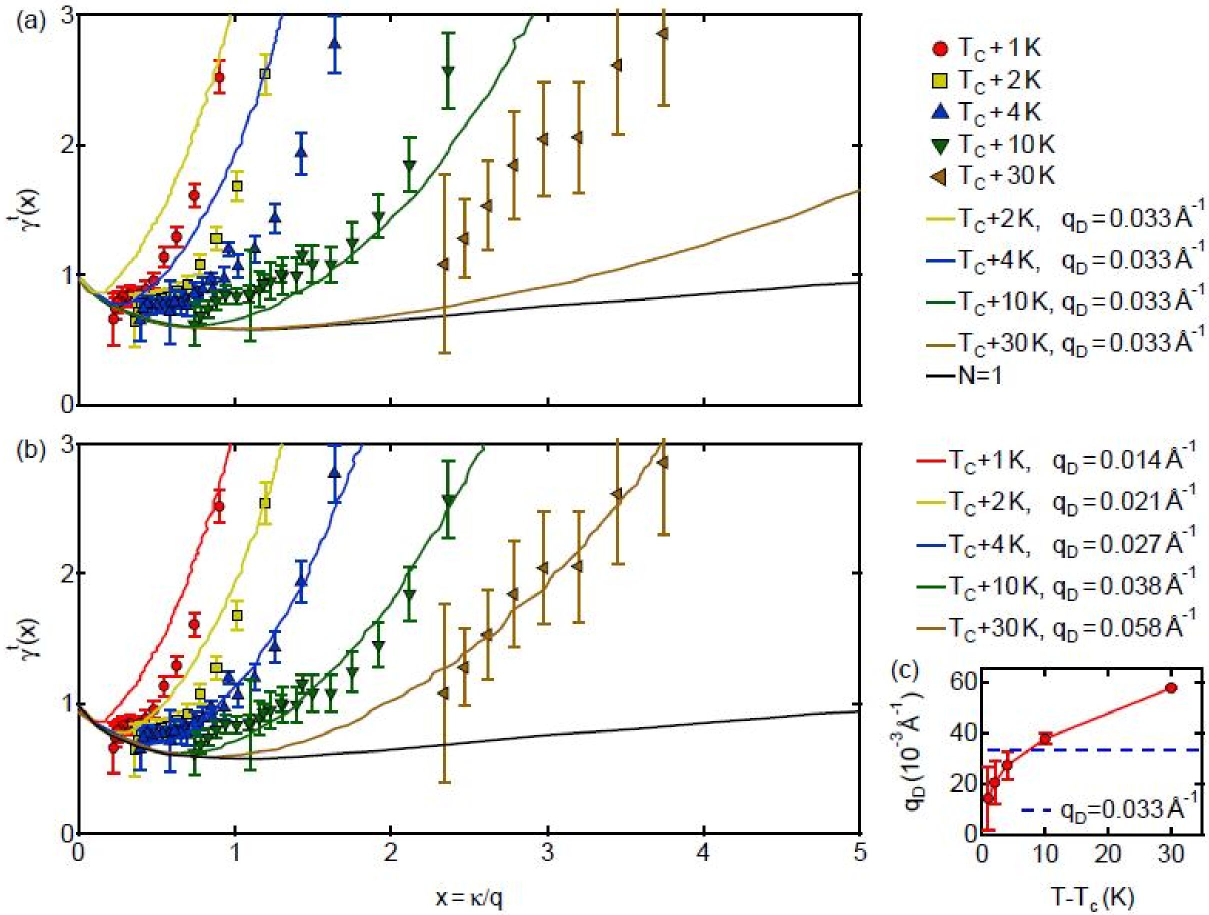
Critical Scattering Fe

Q-dependence linewidth



Deviations al low ***q***
from critical scaling z=5/2

Scaling functions



$\boldsymbol{q_d}$: dipolar interactions

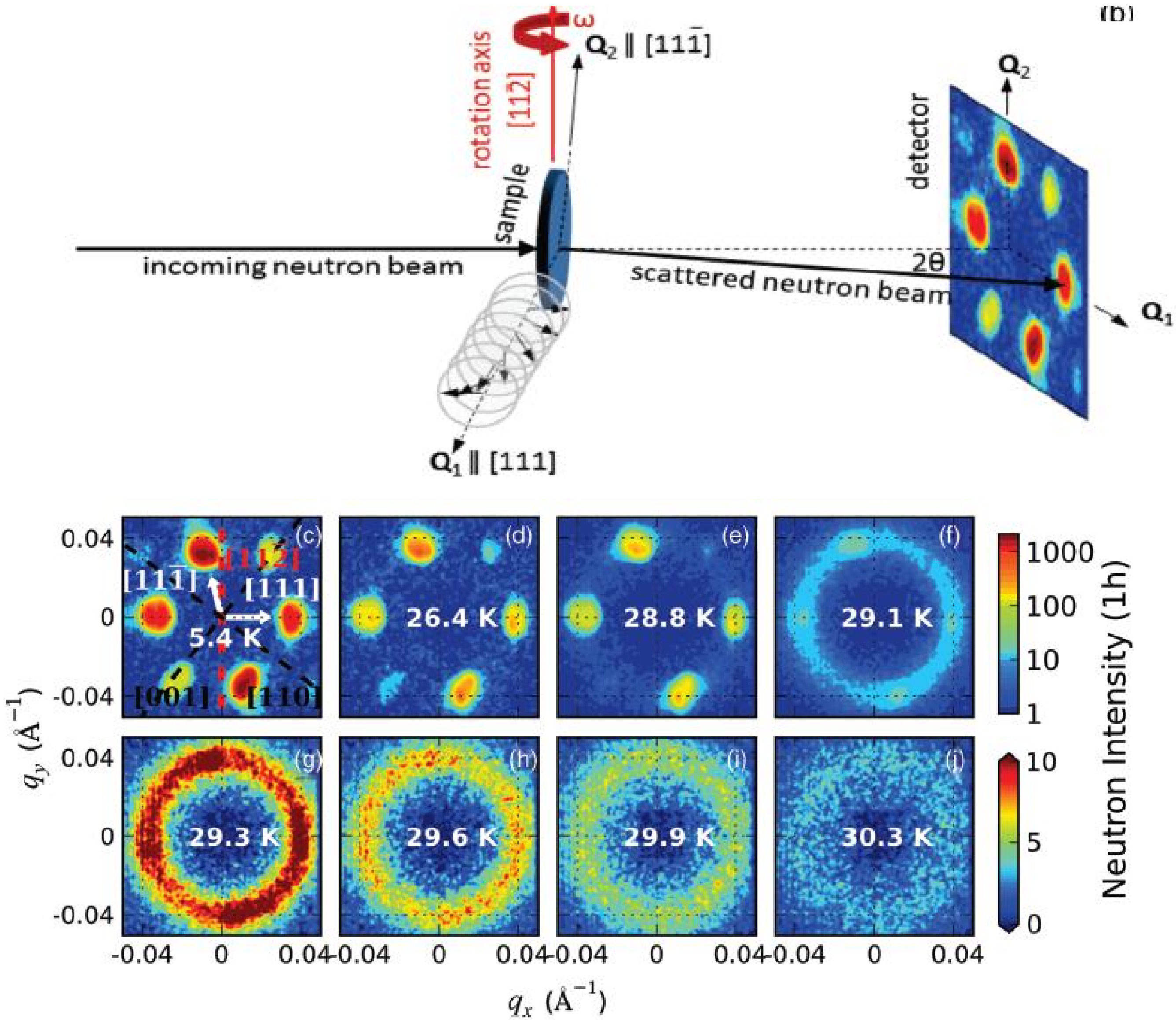
$\boldsymbol{q_d}$: should be temperature independent?

Damping due to conduction electrons?

Brazovskii Transition

Ferromagnet: Critical fluctuations around $\boldsymbol{q}=0$

Helimaget: Critical fluctuations around $\boldsymbol{q}=\boldsymbol{k}$



M. Janoschek et al., *Phys. Rev B* ***87***, *134407 (2013)*

More phase space for critical fluctuations

Interaction of critical fluctuations

Otherwise 2nd order phase transition is driven 1st order

Brazovskii Transition

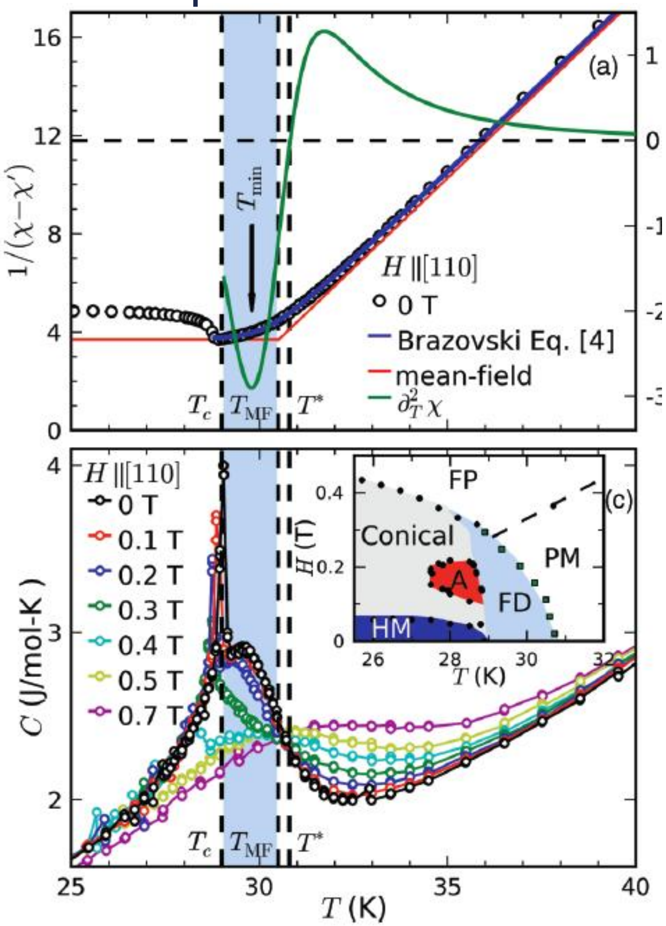
FRM II Forschungs-Neutronenquelle Heinz Maier-Leibnitz

MLZ Heinz Maier-Leibnitz Zentrum

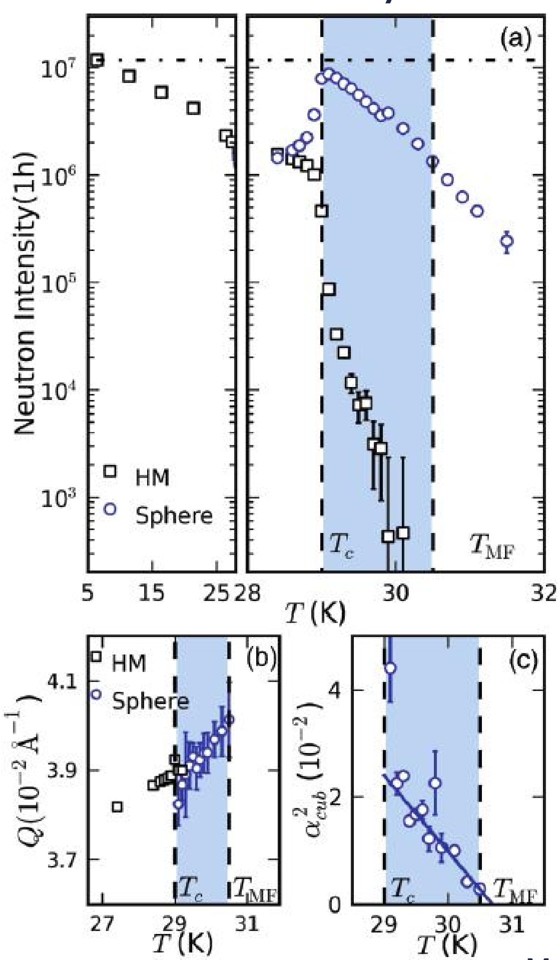
Ferromagnet: Critical fluctuations around $\boldsymbol{q}$=0

Helimaget: Critical fluctuations around $\boldsymbol{q}$=k

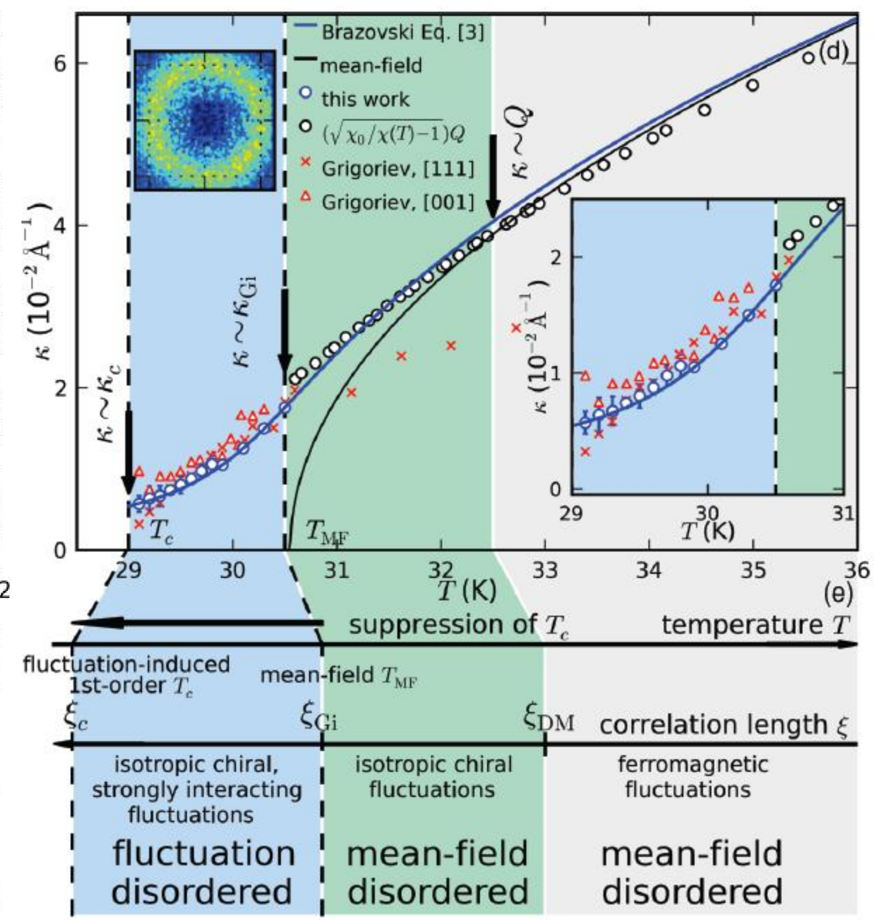
susceptibility
specific heat



intensity



inverse linewidth



M. Janoschek et al., *Phys. Rev B* ***87****, 134407 (2013)*

Neutron prove the realization of the Brazovskii scenario in the helimagnet MnSi