In memory of **Sergey V. Maleev**
(1931 – 2021)
Maleev-Blume Equations;
Dyson -Maleev Representation

Spin waves in full-polarized state of Dzyaloshinskii-Moriya helimagnets: polarized SANS study

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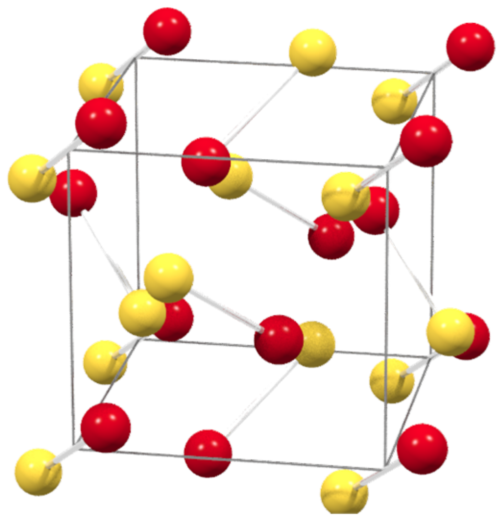
Gatchina, St-Petersburg 188300, Russia

Outline

- Introduction: a bit of history of MnSi
- Spin fluctuations in $Mn_{1-x}Fe_xSi$ by polarized SANS
- Spin waves in $Mn_{1-x}Fe_xSi$ compounds and in others
- Others: $Fe_{1-x}Co_xSi$ and Cu_2OSeO_3
- Conclusions

Crystal structure B20



Examples
MnSi, FeSi, CoSi
$Mn_{1-y}Fe_ySi$, $Mn_{1-y}Co_ySi$, $Fe_{1-x}Co_xSi$

MnGe, FeGe, CoGe
$Mn_{1-y}Fe_yGe$, $Fe_{1-y}Co_yGe$, $Mn_{1-y}Co_yGe$

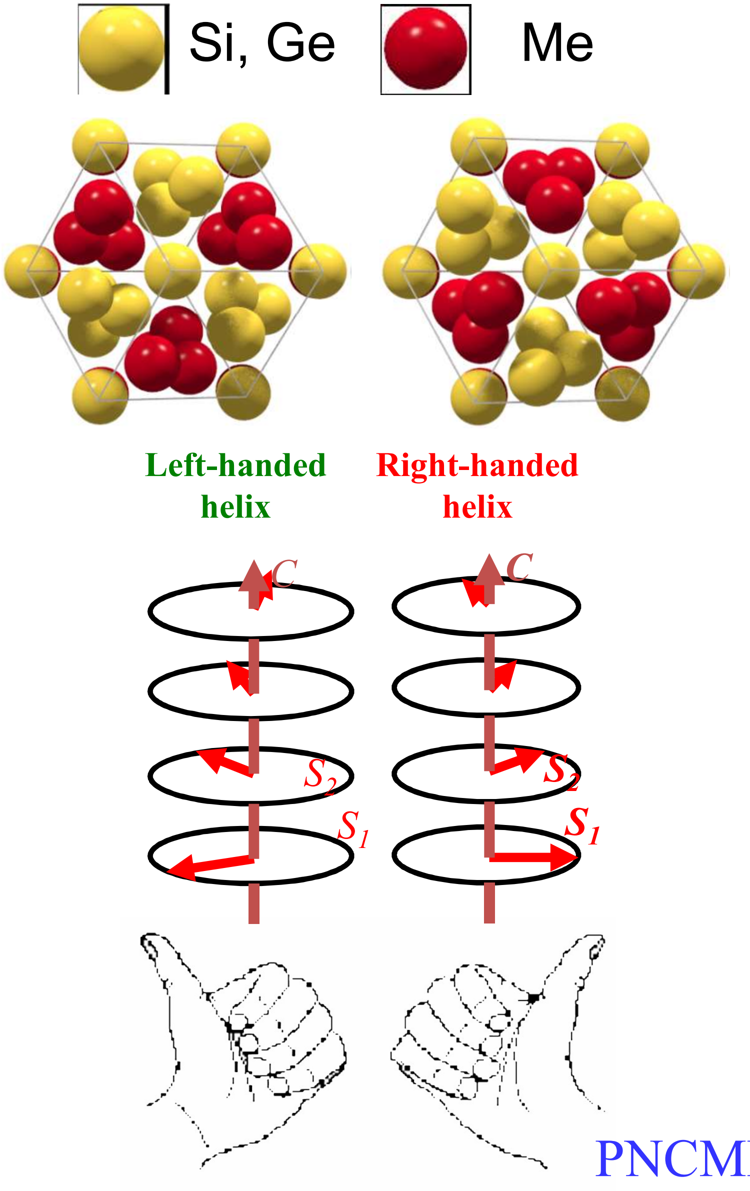
and Cu_2OSeO_3 ; $Co_8Zn_8Mn_4$, etc

Chirality of the crystal structure: non-superposable by pure rotation and translation.

- B20-type cubic
- Space group $P2_13$, $a \approx 4.6$ Å
- 4 Me and 4 Si atoms are inside a unit cell with positions

(u,u,u), (1/2+u,1/2-u,u), (1/2-u,-u,1/2+u), (-u,1/2+u,1/2+u)
with $u_{Mn} = 0.138$ and $u_{Si} = 0.845$

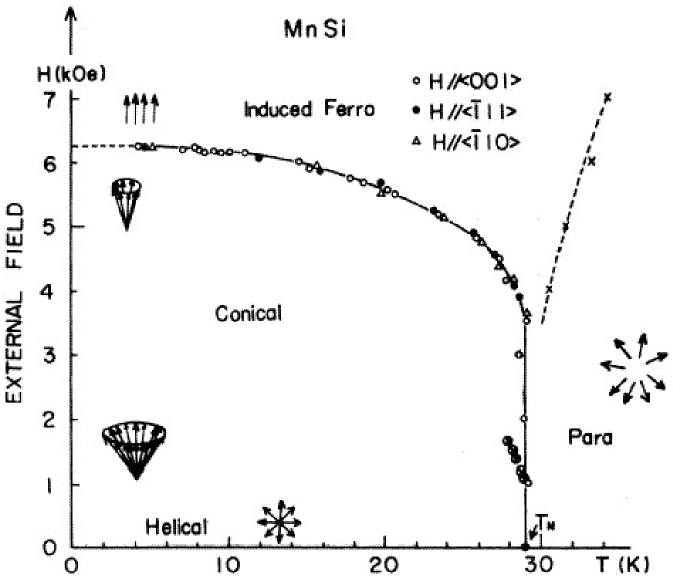
Constituting interactions in magnetic system of B20 compounds



$$W(q) = E_{EX} + E_{DM} + E_{AE} =$$
$$= (A/2)\ (q^2 + \kappa_o^2)\ \mathbf{S}_q^{\ 2} +$$
$$+ D\ (\mathbf{q}\ [\mathbf{S}_q \times \mathbf{S}_{-q}]) + E_{AE}$$

[1] O. Nakanishia, A. Yanase, A. Hasegawa, and M. Kataoka, Solid State Commun. 35, 995 (1980).
[2] P. Bak and M. H. Jensen, J. Phys. C 13, L881 (1980).

(H-T) phase diagrams and Bak-Jensen model



$$W(q) = E_{EX} + E_{DM} + E_{AE} + E_h =$$
$$= (A/2)\ (q^2 + \kappa_o^2)\ \mathbf{S}_q^{\ 2} +$$
$$+ D\ (\mathbf{q}\ [\mathbf{S}_q \times \mathbf{S}_{-q}]) + E_{AE} + E_h$$

1) $k = S\ D/\ A$ the helix wave vector
2) $A\ k^2 = g\ \mu_B\ H_{C2}$ the critical field of transition to the fully polarized state for MnSi

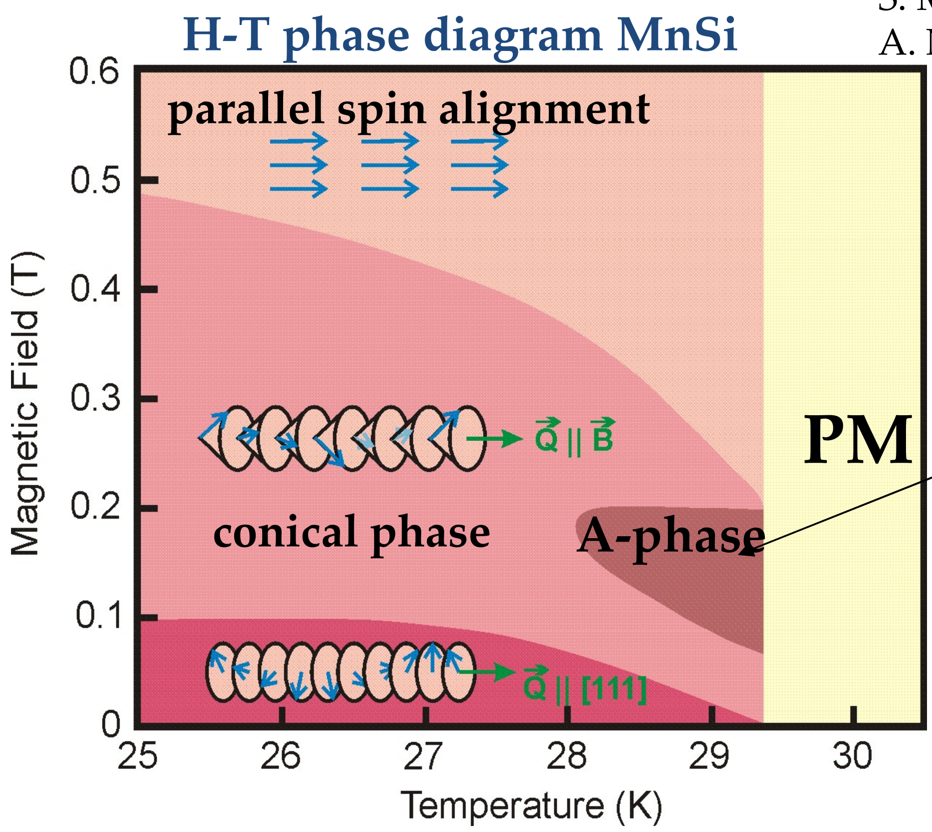
[1] Y. Ishikawa, G. Shirane, J.A. Tarvin, M. Kohgi, Phys.Rev.B **16** (1977) 4956.
[2] O. Nakanishia, A. Yanase, A. Hasegawa, and M. Kataoka, Solid State Commun. 35, 995 (1980).
[3] P. Bak and M. H. Jensen, J. Phys. C 13, L881 (1980).

1) $A\ = g\ \mu_B\ H_{C2}/\ k\ = 50$ meV A²

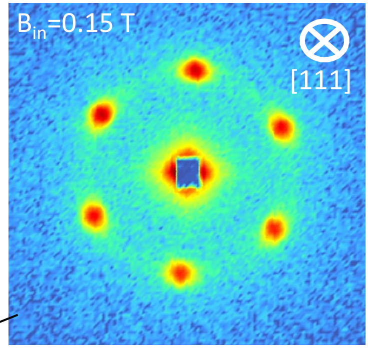
2) $S\ Da = A\ ka = \ 8$ meV A²

Skyrmion lattice in MnSi

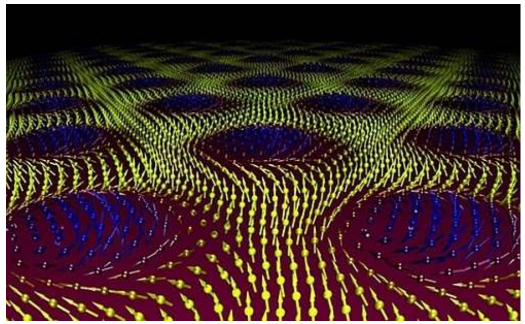
S. Mühlbauer, B. Binz, F. Jonietz, C. Pfleiderer, A. Rosch, A. Neubauer, R. Georgii, P. Böni, Science 323 (2009) 915.



Q - space



r - space



Ferromagnetic exchange J
Dzyaloshinskii-Moriya constant **D**,
Spiral wave vector k = D / J

$$E = \frac{1}{2} \sum_{i \neq j} \left(-J_{ij} \mathbf{s}_i \cdot \mathbf{s}_j + \mathbf{D}_{ij} \cdot [\mathbf{s}_i \times \mathbf{s}_j] \right)$$

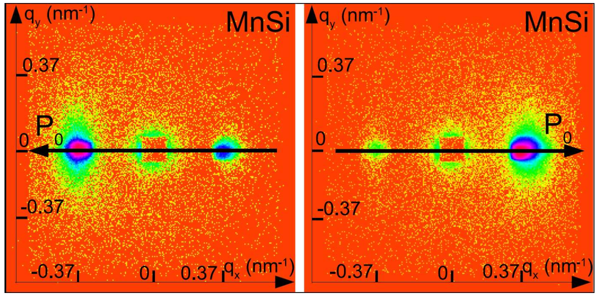
$$\mathbf{M}(\mathbf{r}) = \mathbf{M}_f + \sum_{j=1}^{3} \left(\mathbf{m}_{\mathbf{Q}_j} e^{i\mathbf{Q}_j \cdot \mathbf{r}} + \text{c. c.} \right)$$

Spin Chirality in MnSi: half polarized SANS

Etalon sample MnSi $u_{Mn} = 0.135$, $u_{Si} = 0.845$

The elastic cross section on the magnetic spiral has the form:

$$\sigma_{el}(Q) = [r\, F(Q)/2]^2 \{ S^2 (1+(\mathbf{qc}))^2 + + 2\langle C \rangle (\mathbf{q}\ \mathbf{P_0})(\mathbf{q}\ \mathbf{c})\ \} \delta(\mathbf{Q}\text{-}\mathbf{k})$$

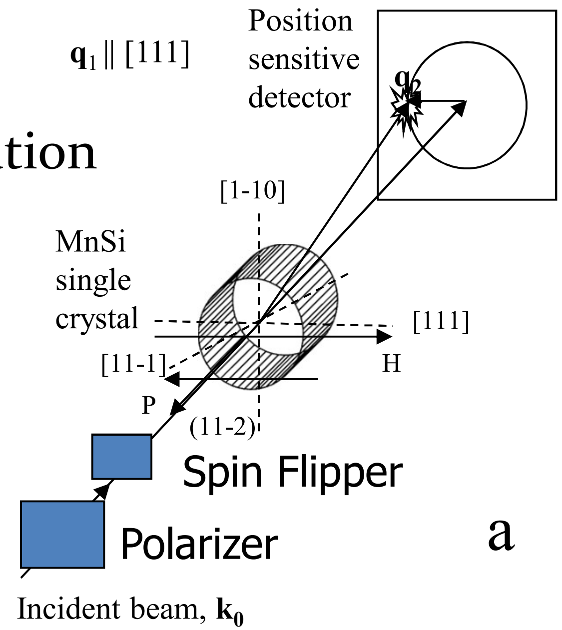


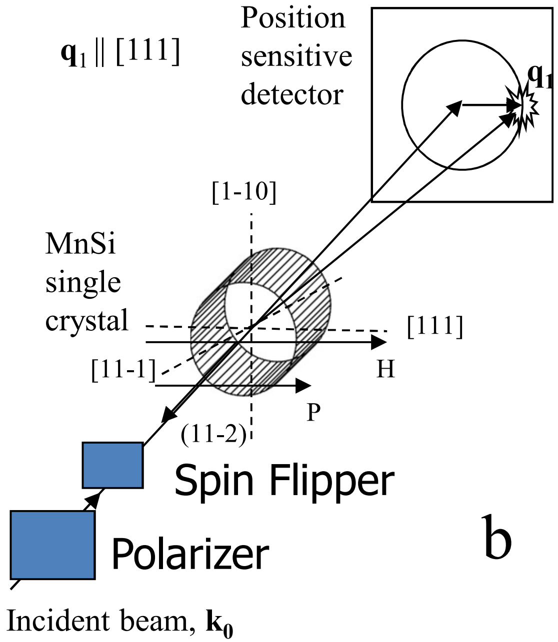
$\mathbf{C} = [\mathbf{S}_1 \times \mathbf{S}_2]$ and

$\mathbf{q}$ is the unit vector of Q

$\mathbf{P_o}$ is the incident polarization

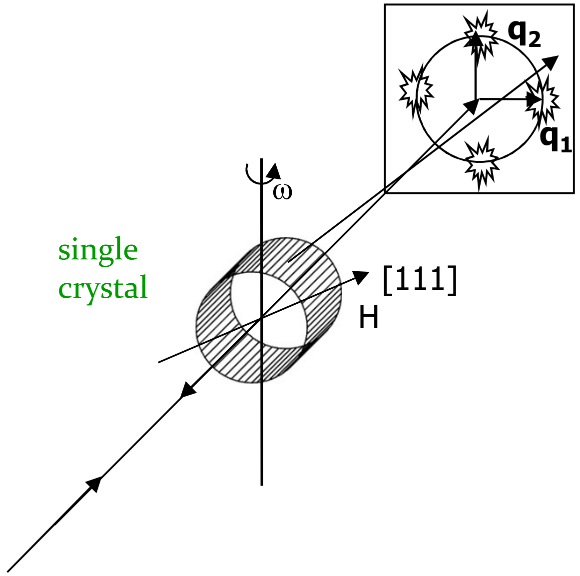
M. Tanaka, H. Takayoshi, M. Ishida, Ya. Endoh, J. Phys. Soc. J. 54, 2970 (1985).



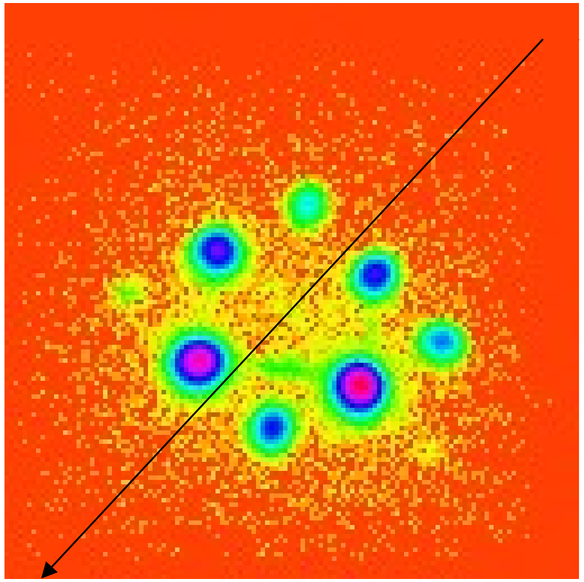


The thermal phase transition in MnSi

Small angle neutron diffraction experiment on MnSi single crystal performed at SANS2@GKSS/HZG (resp. H. Eckerlebe)



T =10 K



Screen shot of the neutron diffraction picture from MnSi

S.V. Grigoriev, S.V. Maleyev, A.I. Okorokov, Yu.O. Chetverikov, R. Georgii, P. Boni, D. Lamago, H. Eckerlebe, K. Pranzas, Phys. Rev.B **72** (2005) 134420.

Neutron cross section at $T > T_C$

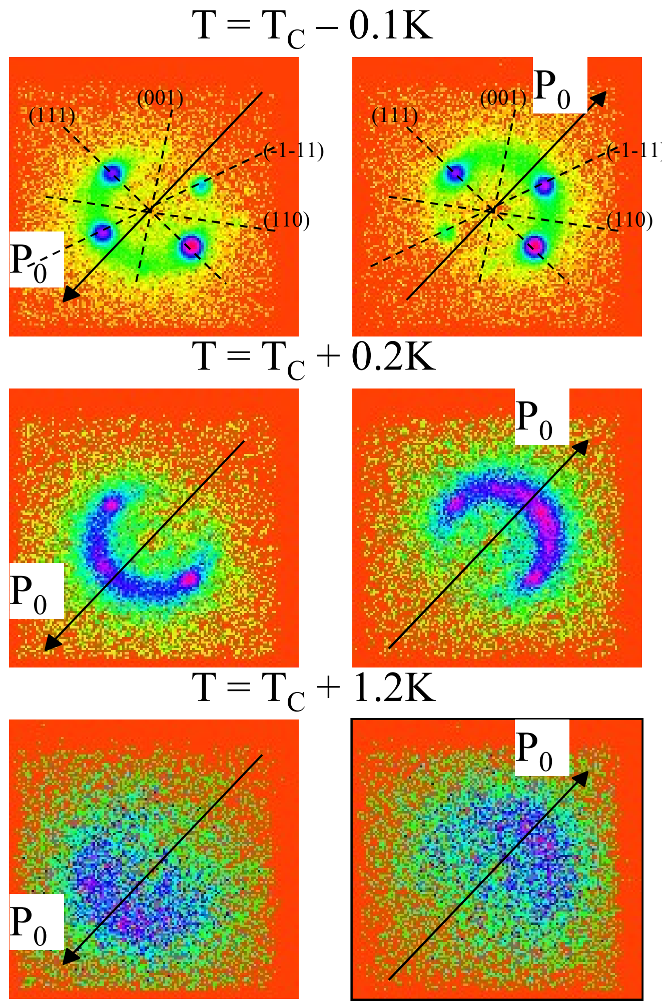
Neutron cross section (S.V. Maleev)

$$\frac{d\sigma}{d\Omega} = \frac{[rF(\mathbf{q})]^2 T}{[B(q+k)^2+\kappa^2]} \frac{k^2+q^2+\kappa^2-2k\mathbf{q}\mathbf{P}_0}{[(q-k)^2+\kappa^2+(|U|k^2/2)(\hat{q}^4-1/3)]}$$

Polarization of the scattered neutrons

$$P_s = \frac{\sigma(\mathbf{P}_0)-\sigma(-\mathbf{P}_0)}{\sigma(\mathbf{P}_0)+\sigma(-\mathbf{P}_0)} = -\frac{2kqP_0\cos\varphi}{q^2+k^2+\kappa^2}$$

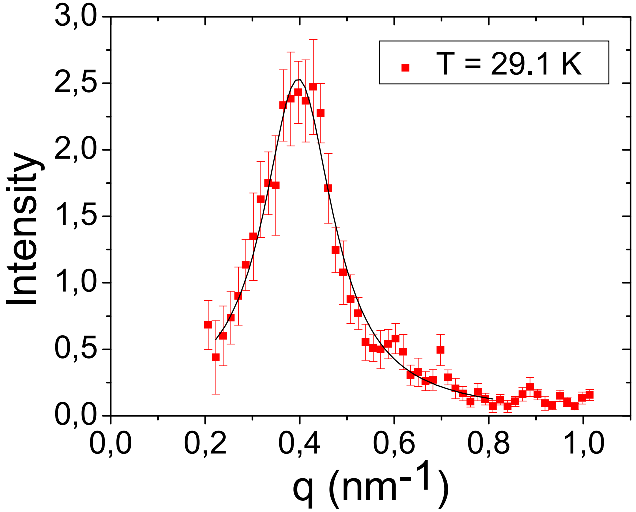
Phys.Rev.B **72** (2005) 134420
Phys.Rev.B **81** (2010) 144413

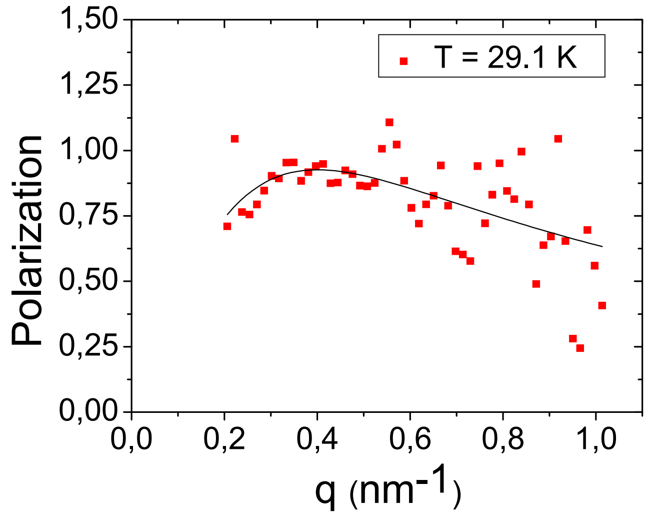


Data analysis: q-dependence

$$\frac{d\sigma}{d\Omega} = \frac{[rF(\mathbf{q})]^2 T}{[B(q+k)^2+\kappa^2]} \frac{k^2+q^2+\kappa^2-2k\mathbf{q}\mathbf{P}_0}{[(q-k)^2+\kappa^2+(|U|k^2/2)(\hat{q}^4-1/3)]}$$

$$P_s = \frac{\sigma(\mathbf{P_0})-\sigma(-\mathbf{P_0})}{\sigma(\mathbf{P_0})+\sigma(-\mathbf{P_0})} = -\frac{2kqP_0\cos\varphi}{q^2+k^2+\kappa^2}$$



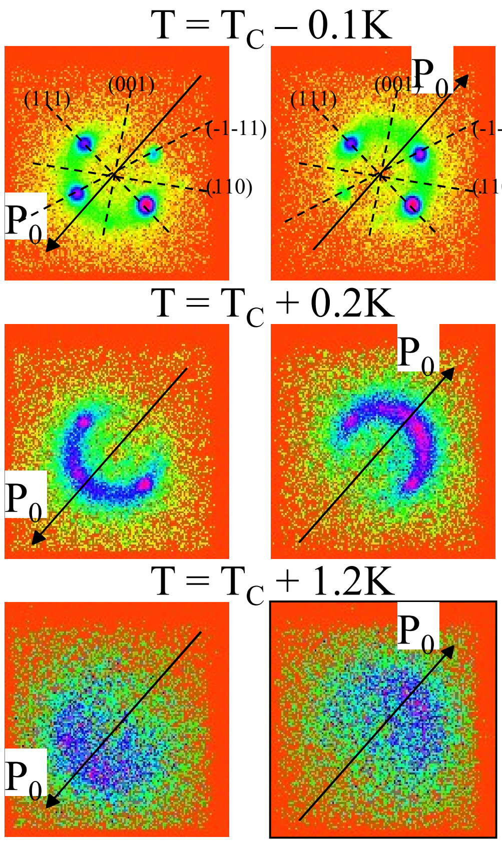


Intensity of the scattering at **q** || **P**$_o$ I(q) = (I(q,P$_o$) + I(q,-P$_o$))

Phys.Rev.B **72** (2005) 134420

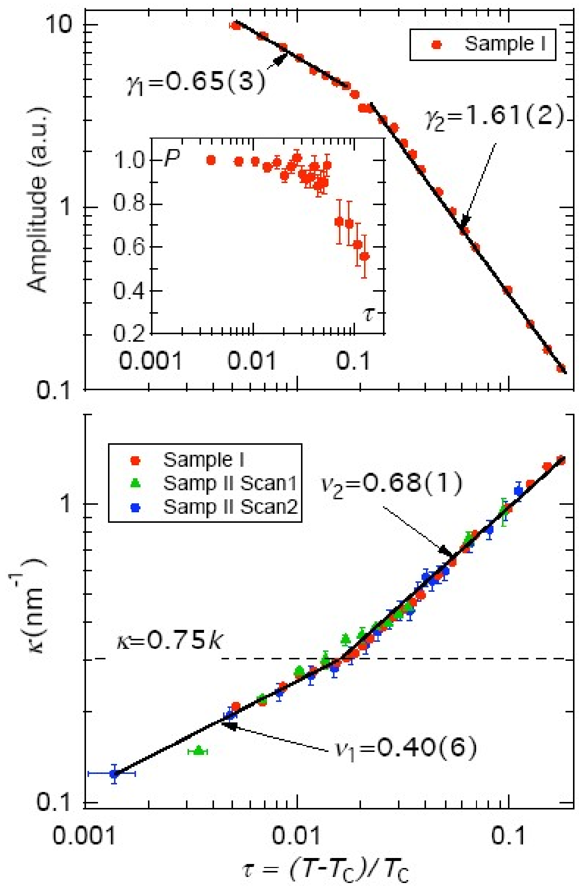
Polarization of the scattering at **q** || **P**$_o$ at T = T$_C$ + 0.3 K.

Data analysis: temperature dependence



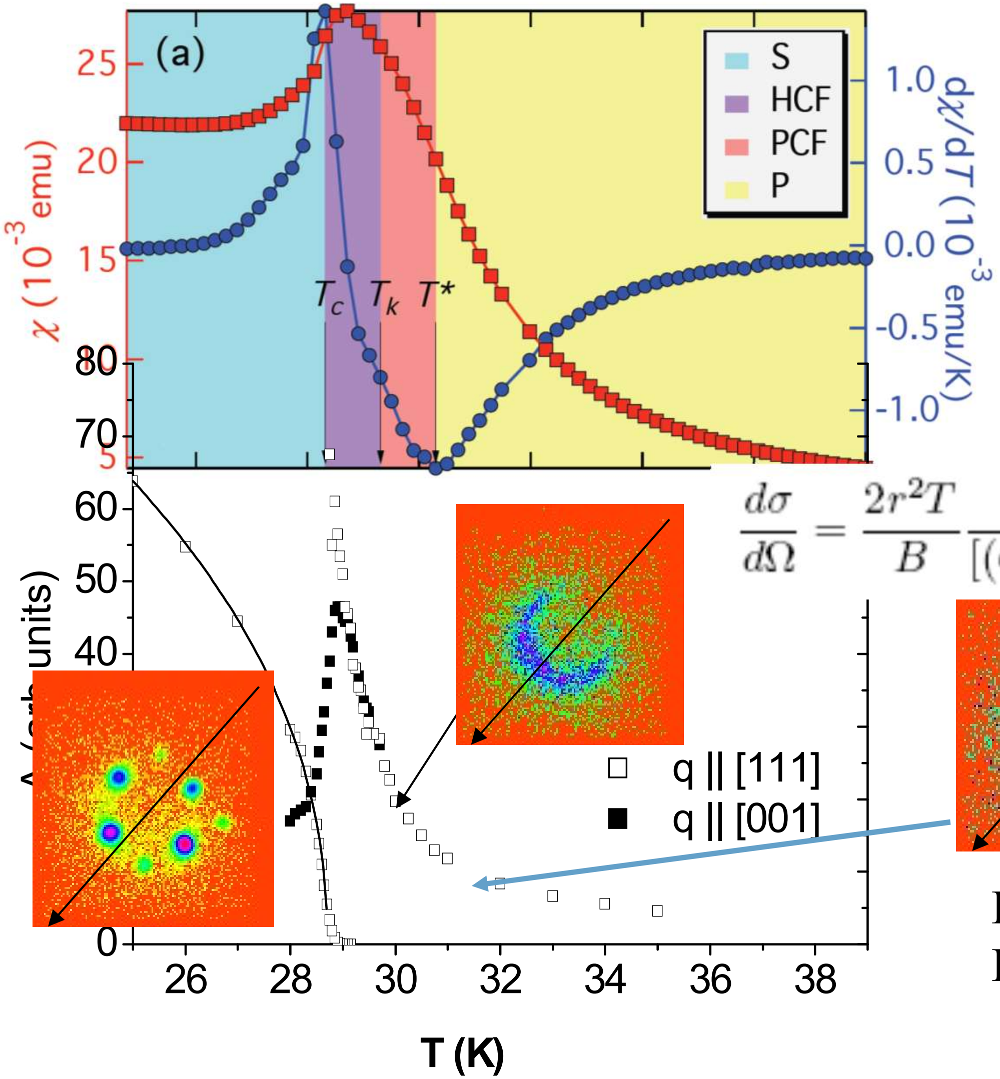
Polarized SANS maps

Phys.Rev.B **72** (2005) 134420
Phys.Rev.B **81** (2010) 144413



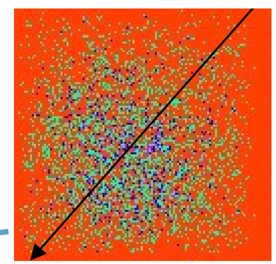
τ - dependence of (a) amplitude of scattering nd (b) inverse correlation length.

Thermal phase transition in MnSi



Thermal phase transition in MnSi is characterized by two temperatures: ordering temperature Tc and T_{DM}

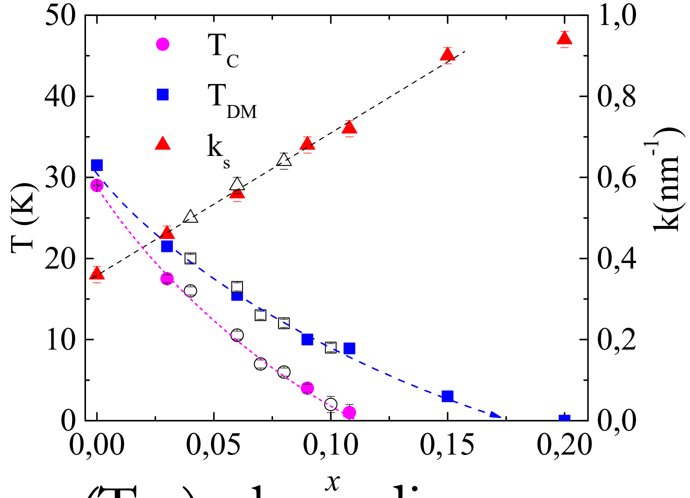
$$\frac{d\sigma}{d\Omega} = \frac{2r^2T}{B}\frac{Q^2+k^2+\kappa_1^2+2k(D/|D|)(\mathbf{Q}\cdot\mathbf{P}_0)}{[(Q+k)^2+\kappa_1^2][(Q-k)^2+\kappa_1^2]+k^2\kappa_A^2(\mathbf{Q})}$$



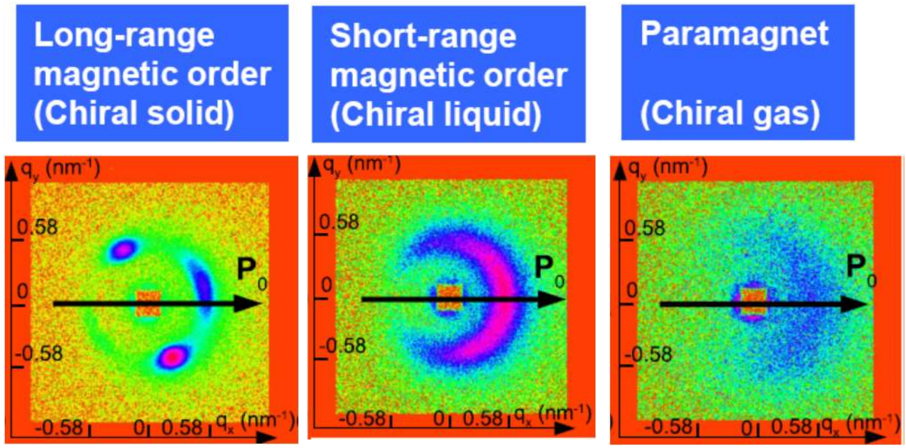
Phys.Rev.B **72** (2005) 134420
Phys.Rev.B **81** (2010) 144413

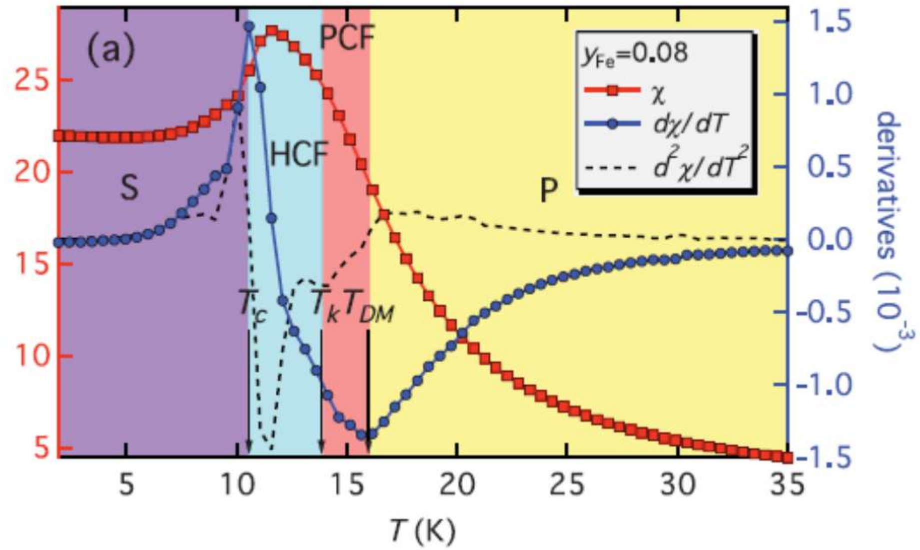
Thermal phase transition in $Mn_{1-x}Fe_xSi$

Thermal phase transition in $\mathbf{Mn_{1-y}Fe_ySi}$ is characterized by two temperatures: ordering temperature Tc and T_{DM}



(T-x) phase diagram





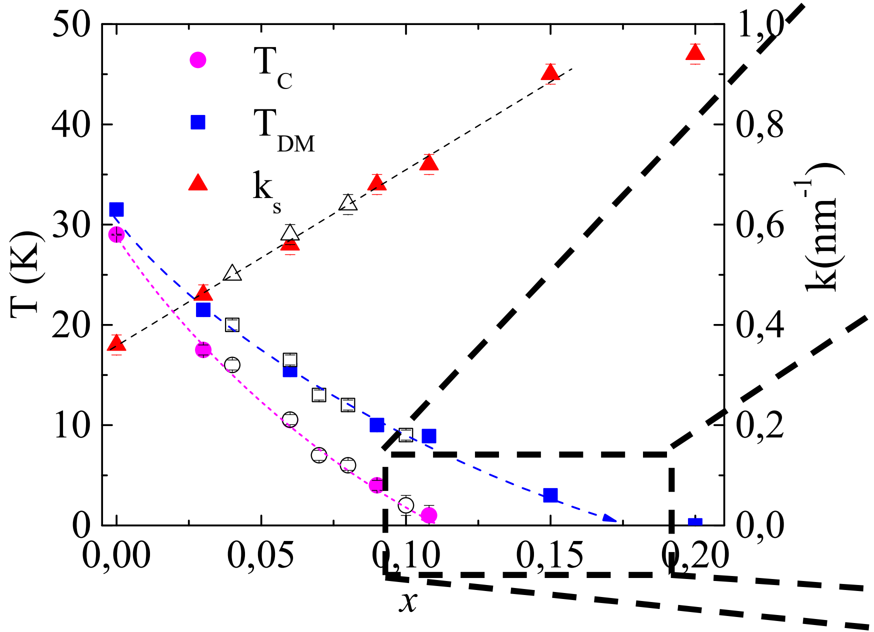
Correlation between magnetic susceptibility and polarized neutron scattering data is established in $Mn_{1-x}Fe_xSi$.

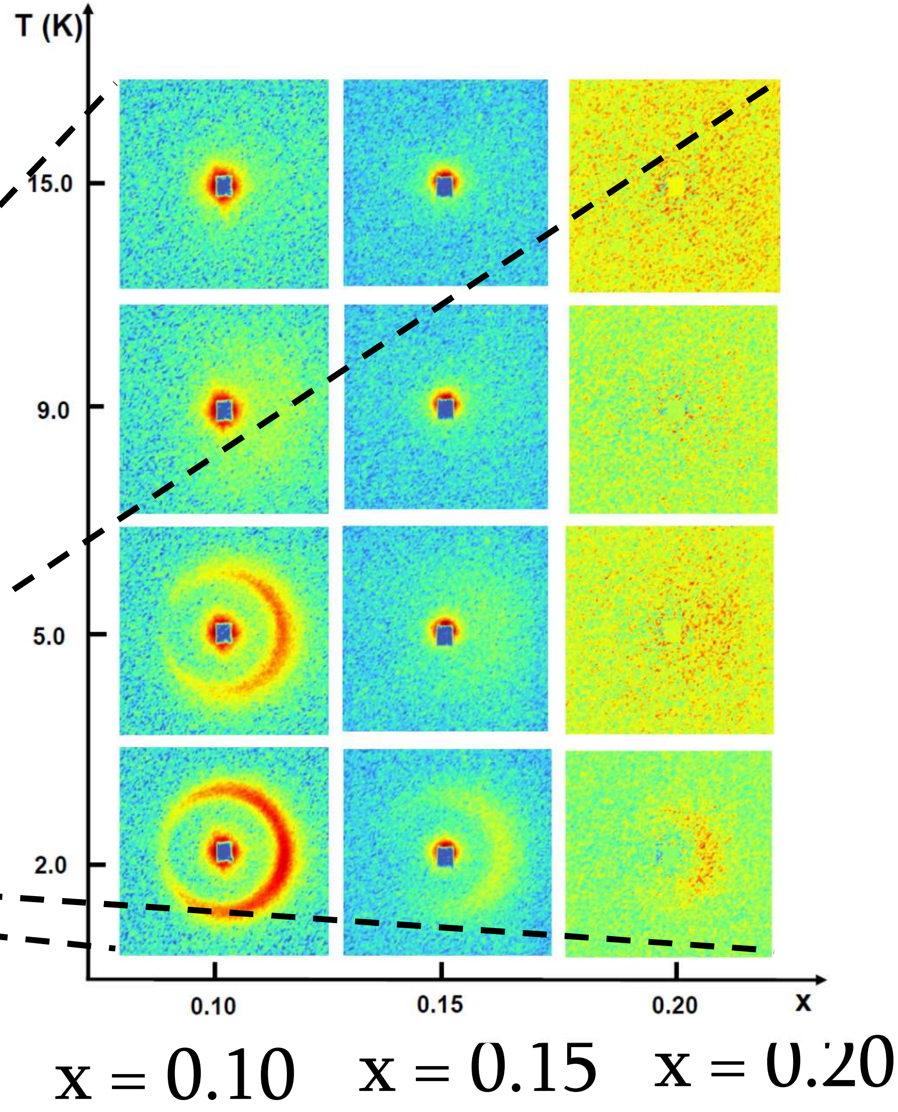
The extrema of the $\partial\chi/\partial T$ derivative may be used for identification of the magnetic phases with long-range and short-range magnetic order.

Phys.Rev.B **83** (2011) 224411

SANS measurements in $Mn_{1-x}Fe_xSi$ Beyond the Quantum Phase Transition

The polarized small-angle neutron scattering (SANS) was performed at D22@ILL (resp. Ch. Dewhurst). **$Mn_{1-y}Fe_ySi$** compounds with x = 0.10, x = 0.15 and x = 0.20.

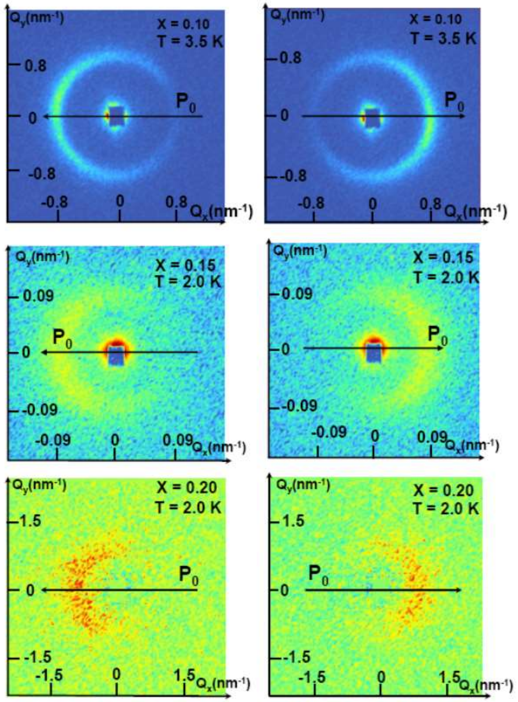


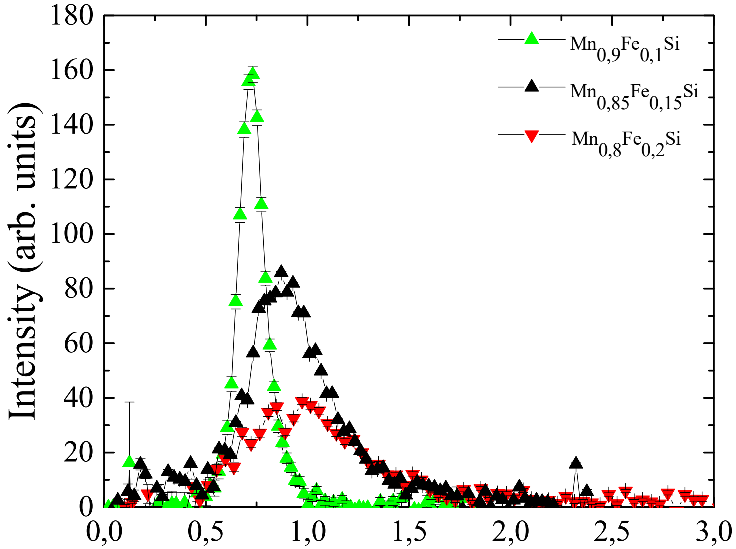


S. V. Grigoriev, O. I. Utesov, N. M. Chubova, C. D. Dewhurst, D. Menzel, S. V. Maleyev, JETP **132**, 588–595 (2021)

Short-range order beyond the QCP

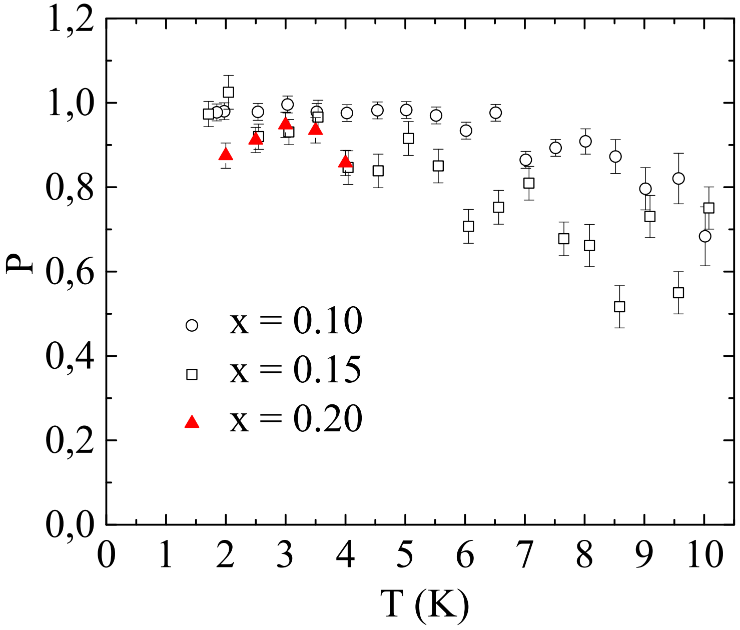
Analysis of the polarized SANS data gives
(i) the position of the peak ≡ helix wavevector k;
(ii) the width of the peak ≡ the coherent length of the helix fluctuation;
(iii) polarization of the peak ≡ the chirality of the fluctuation.





$$\frac{d\sigma}{d\Omega} = \frac{[rF(\mathbf{q})]^2 T}{[B(q+k)^2+\kappa^2]} \frac{k^2+q^2+\kappa^2-2k\mathbf{q}\mathbf{P}_0}{[(q-k)^2+\kappa^2+(|U|k^2/2)(\hat{q}^4-1/3)]}$$

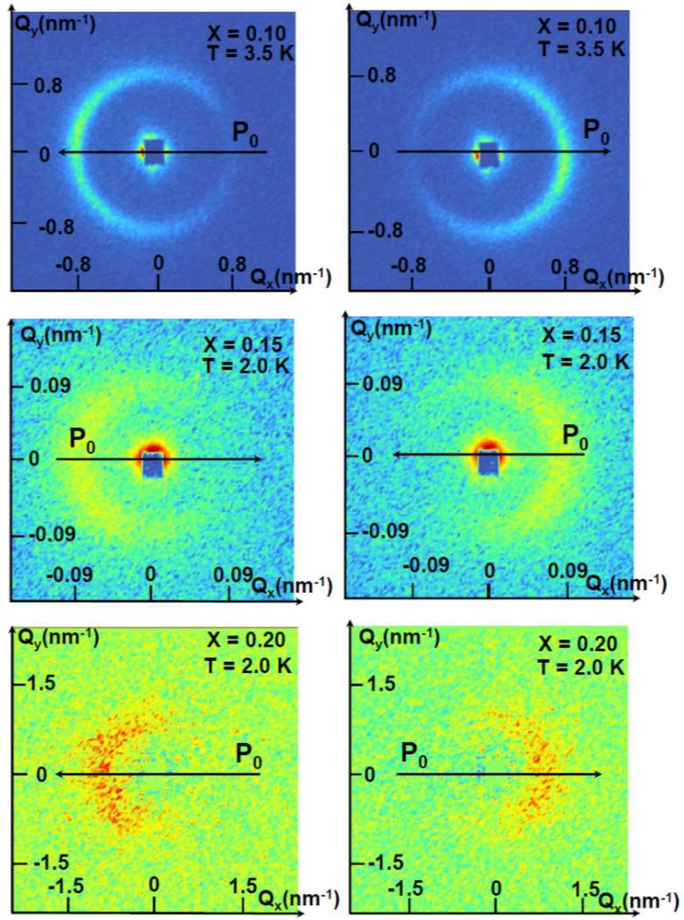
Polarization of scattered neutrons



x = 0.10

x = 0.15

x = 0.20

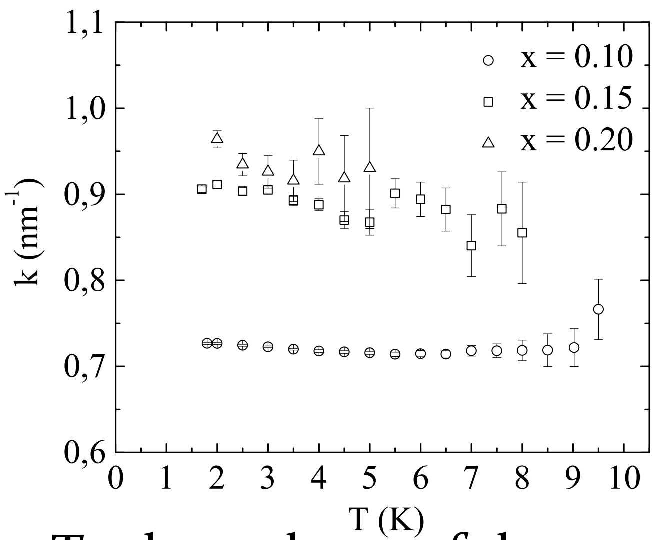


T - dependence of P for
x= 0.10, x = 0.15 and x = 0.20.

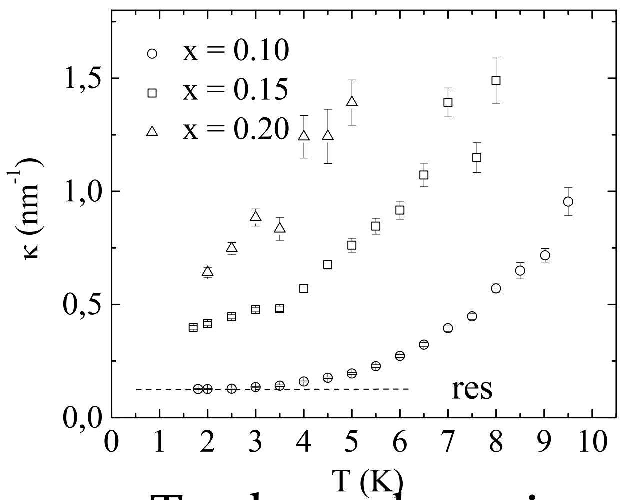
$$P_s = \frac{\sigma(\mathbf{P_0}) - \sigma(-\mathbf{P_0})}{\sigma(\mathbf{P_0}) + \sigma(-\mathbf{P_0})} = -\frac{2kqP_0\cos\varphi}{q^2 + k^2 + \kappa^2}$$

SANS intensity: scattering profiles.

Helical wave vector k



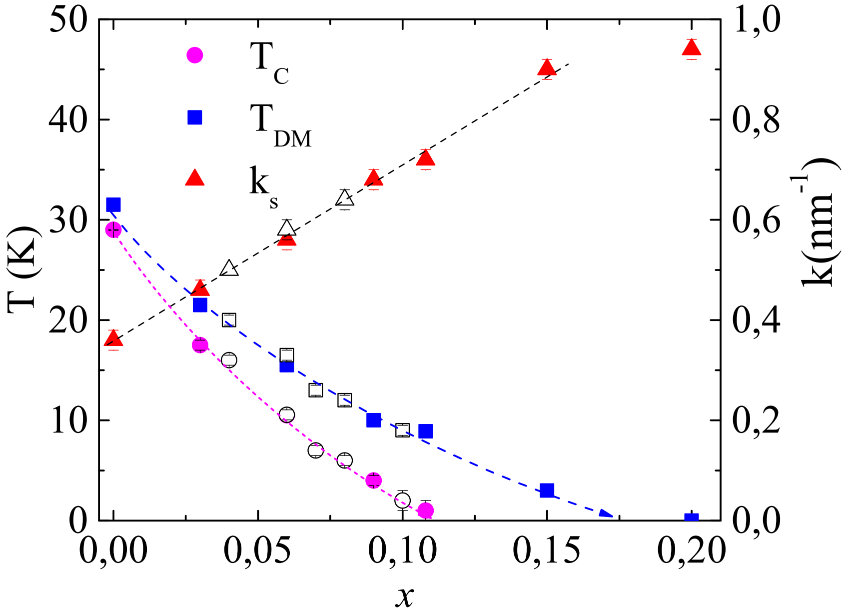
T - dependence of the wave vector k for x= 0.10, 0.15 and 0.20.



T – dependence inverse correlation length for x= 0.10, 0.15 0.20.

$$\frac{d\sigma}{d\Omega} = \frac{[rF(\mathbf{q})]^2 T}{[B(q+k)^2+\kappa^2]} \frac{k^2+q^2+\kappa^2-2k\mathbf{q}\mathbf{P}_0}{[(q-k)^2+\kappa^2+(|U|k^2/2)(\hat{q}^4-1/3)]}$$

Conclusions and questions:



(T-x) phase diagram of $\mathbf{Mn_{1-y}Fe_ySi}$

Conclusions

- Ordering temperature Tc approaches zero at x = 0.11
- Temperature T_{DM} approaches zero at x = 0.17
- Polarization is close to 1 at x < 0.17 means that Dzyaloshinskii-Moriya interaction is finite and provides chiral rotation of spins within magnetic fluctuations.

Question

- What is the value of ferromagnetic exchange interaction?
- Decreases together with Tc or with T_{DM} ?

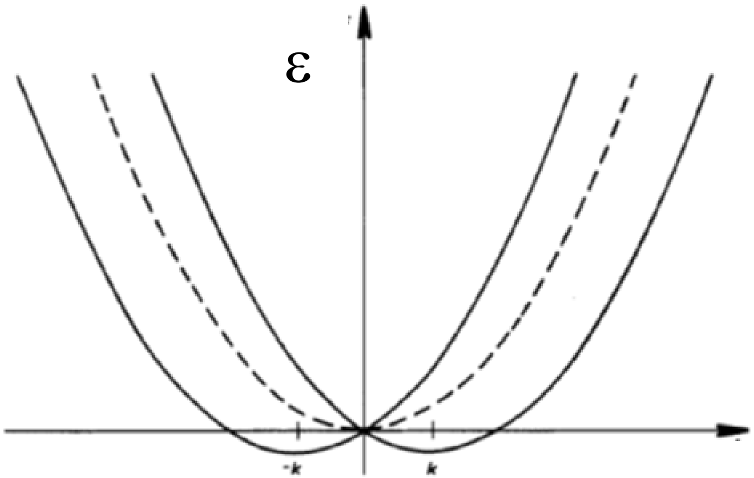
Disorder induced by the antiferromagnetic bonds and DM interaction that destabilizes the magnetic order additionally.

Spin-waves in ferro- and heli- magnets

Mitsuo Kataoka, J.Phys.Soc.Jap. 56 (1987) 3635

- Dynamics in the - "field-induced ferromagnetic" – fully polarized state $H > H_{C2}$

Ferromagnet	Helimagnet ($H > H_{C2}$)
$\varepsilon_q = Aq^2 + g\mu H$	$\varepsilon_q = A(q-k)^2 + g\mu (H - H_{C2})$



Bak-Jensen model

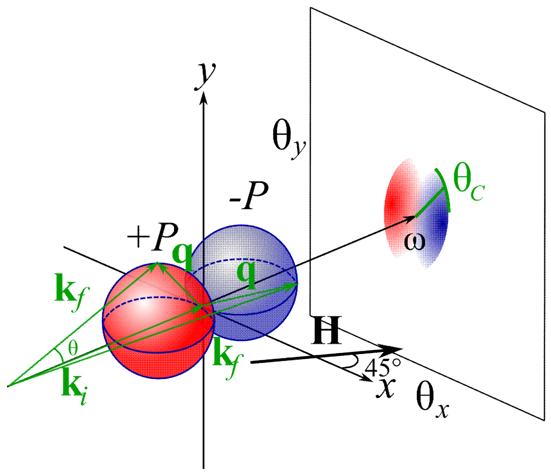
$W(q) = E_{EX} + E_{DM} + E_{AE} + E_h =$
$= (A/2)\ (q^2 + \kappa_o^2)\ \mathbf{S}_q^{\ 2} +$
$+ D\ (\mathbf{q}\ [\mathbf{S}_q \times \mathbf{S}_{-q}]) + E_{AE} + E_h$

1) $k = S\,D/\,A$
2) $A\,k^2 = g\,\mu_B\,H_C$

Small angle neutron scattering on spin waves in ferromagnets: kinematics

Conservation energy law	(1)	$\hbar\omega = E' - E = \left(\hbar^2/2m\right)\left(k'^2 - k^2\right) = \varepsilon_q$
Conservation impulse law	(2)	$\vec{q} = \vec{k}' - \vec{k}$
Dispersion of SW in ferromagnets:	(3)	$\varepsilon_q = Aq^2$
Substituting (3) to (1) and	(4)	$k'^2 = k^2 - \alpha q^2$
accounting for (2) we obtain	(5)	$q^2 = k'^2 + k^2 - 2k'k\cos\theta$
the following solution:		

$$k' = \left(\frac{\alpha}{\alpha - 1}\right) k \left[\cos\theta \pm \sqrt{\cos^2\theta - \left(1 - \alpha^{-2}\right)}\right]$$



$$\theta_0 = \alpha^{-1} = \hbar^2/2Dm$$

A.I. Okorokov, V.V. Runov, B.P. Toperverg, A.D. Tretyakov, E.I.Maltsev, I.M.Puzeii, V.E.Mikhailova, JETP Lett. 43 (1986) 503.

Small angle neutron scattering on spin waves in helimagnets: kinematics

Conservation energy law (1) $\hbar\omega = E' - E = \left(\hbar^2/2m\right)\left(k'^2 - k^2\right) = \varepsilon_q$

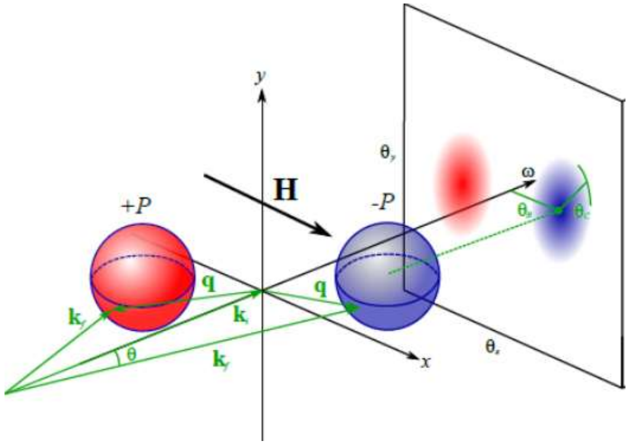
Conservation impulse law (2) $\vec{q} = \vec{k}' - \vec{k}$

Dispersion of SW in helimagnets: (3) $\varepsilon_q = A(\mathbf{q} - \mathbf{k}_s)^2 + g\mu\,(H - H_{C2})$

Substituting (3) to (1) and accounting for (2) we obtain equation (4) and solution: (4) $k'^2 = k^2 - \alpha(A(\mathbf{q} - \mathbf{k}_s)^2 + g\mu\,(H - H_{C2}))$

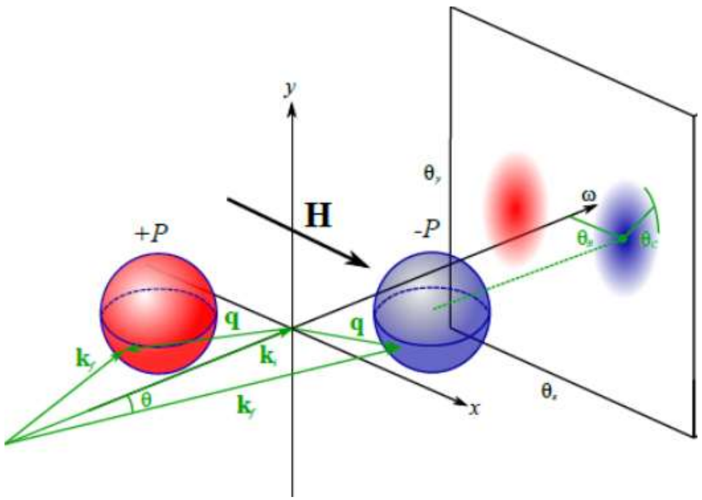
$$(\tilde{\omega} - \theta_0)^2 + (\theta_x - \theta_s)^2 + \theta_y^2 = \theta_c^2,$$

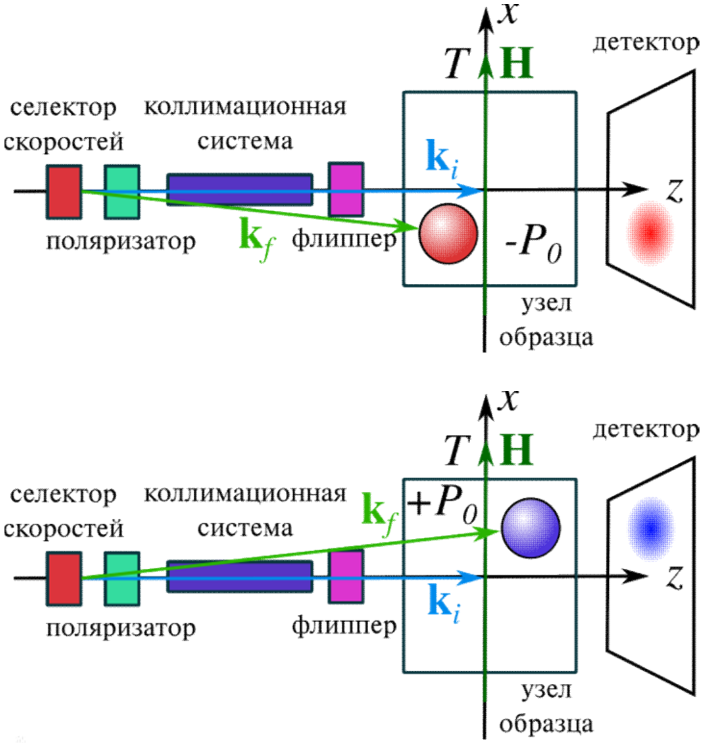
$$(\tilde{\omega} + \theta_0)^2 + (\theta_x + \theta_s)^2 + \theta_y^2 = \theta_c^2,$$



S. V. Grigoriev, A. S. Sukhanov, E. V. Altynbaev, S.~A. Siegfried, A. Heinemann, P. Kizhe, and S. V. Maleyev Phys. Rev. B 92 220415(R) (2015)

Small Angle **Polarized** Neutron scattering on magnons in MnSi



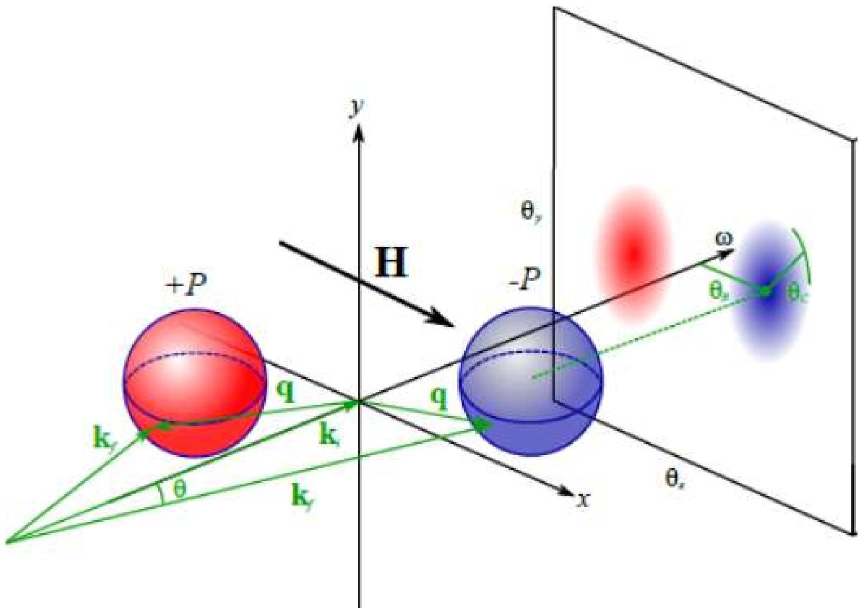


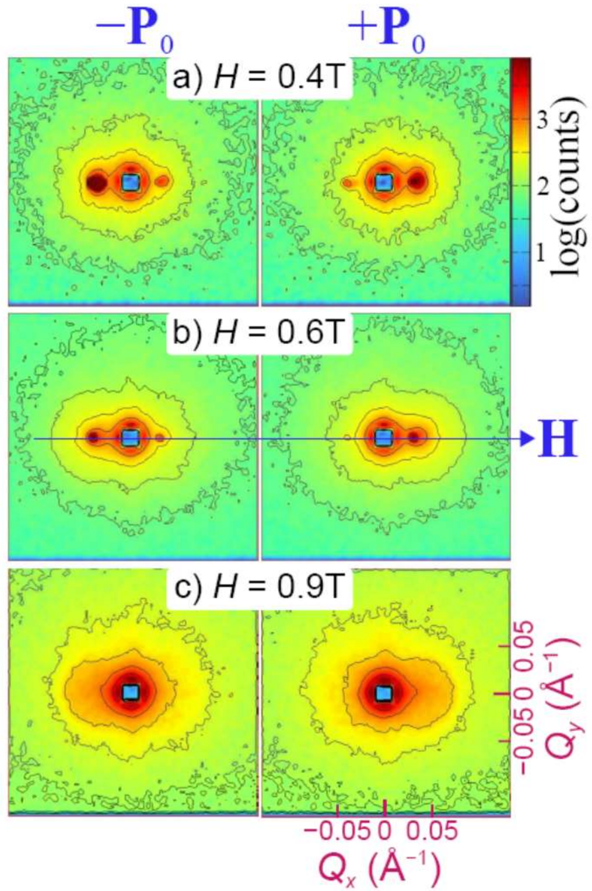
$$\sigma(\mathbf{q},\omega) = (r_m S)^2 F(\mathbf{q})^2 (\frac{k_f}{k_i})^2 \times$$

$$\times\{[1 + (\mathbf{em})^2 + 2(\mathbf{em})(\mathbf{eP}_0)]n_q \delta(\omega - \epsilon_q) +$$

$$+[1 + (\mathbf{em})^2 - 2(\mathbf{em})(\mathbf{eP}_0)](n_q + 1)\delta(\omega + \epsilon_q)\},$$

Experimental setup: SANS machines
SANS1@MLZ, PA20@LLB, D11 @ILL



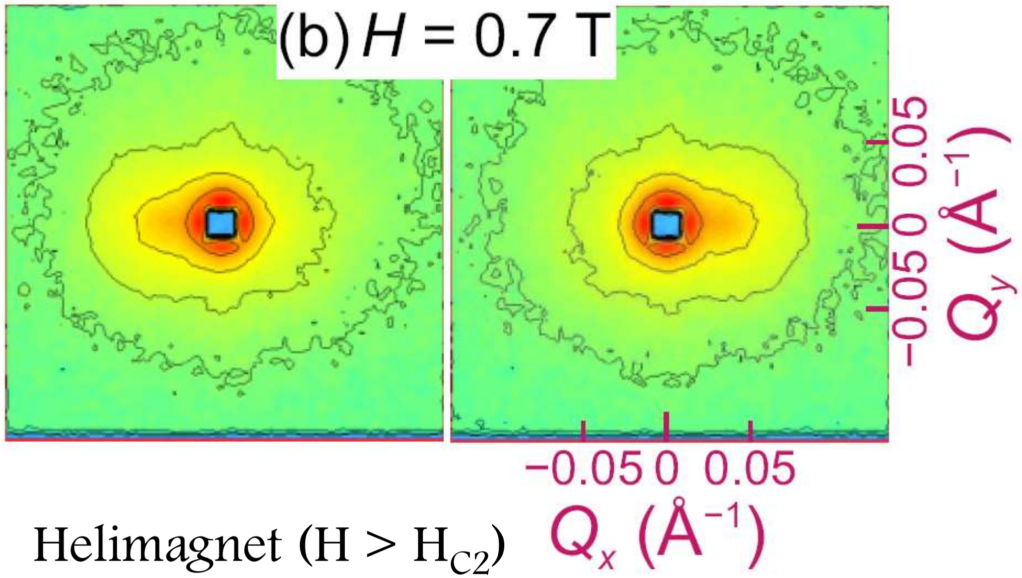


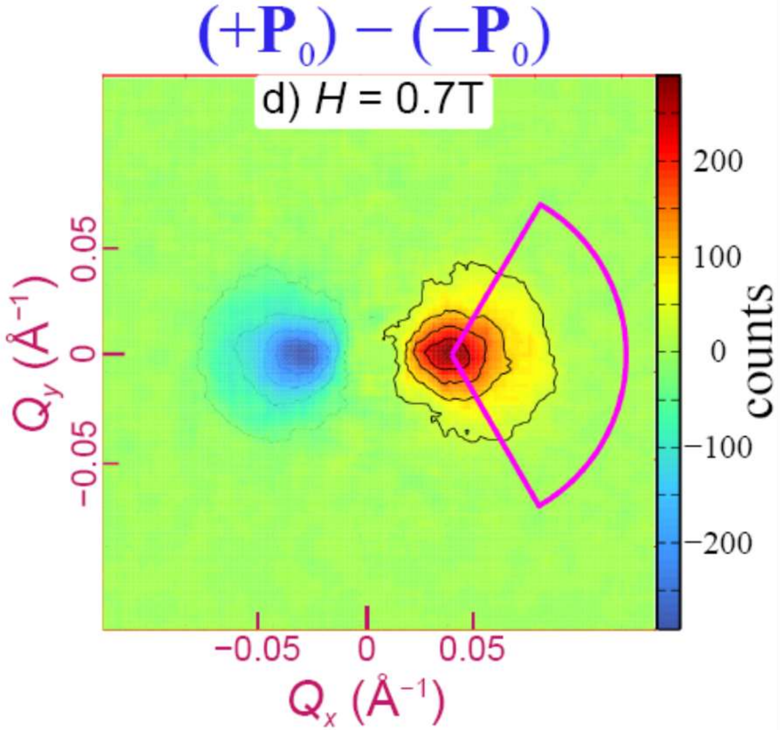
$$\sigma(\mathbf{q},\omega) = (r_m S)^2 F(\mathbf{q})^2 (\frac{k_f}{k_i})^2 \times$$

$$\times\{[1 + (\mathbf{em})^2 + 2(\mathbf{em})(\mathbf{eP}_0)] n_q \delta(\omega - \epsilon_q) +$$

$$+[1 + (\mathbf{em})^2 - 2(\mathbf{em})(\mathbf{eP}_0)](n_q + 1)\delta(\omega + \epsilon_q)\},$$

Small Angle **Polarized** Neutron scattering on magnons in MnSi





Helimagnet ($H > H_{C2}$)

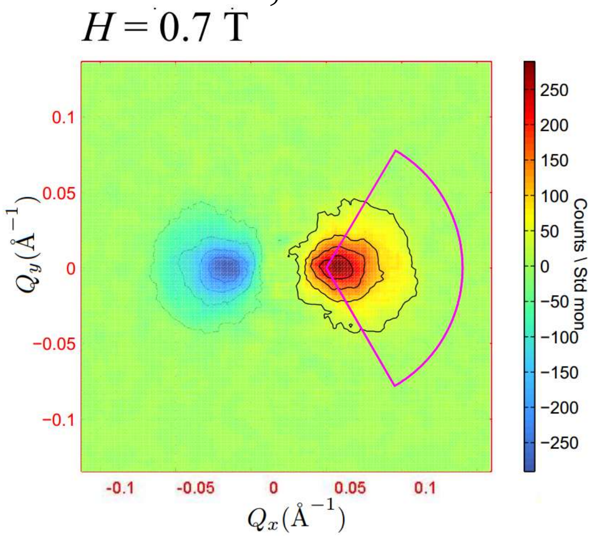
$\varepsilon_q = A(q - k)^2 + g\mu\ (H - H_{C2})$

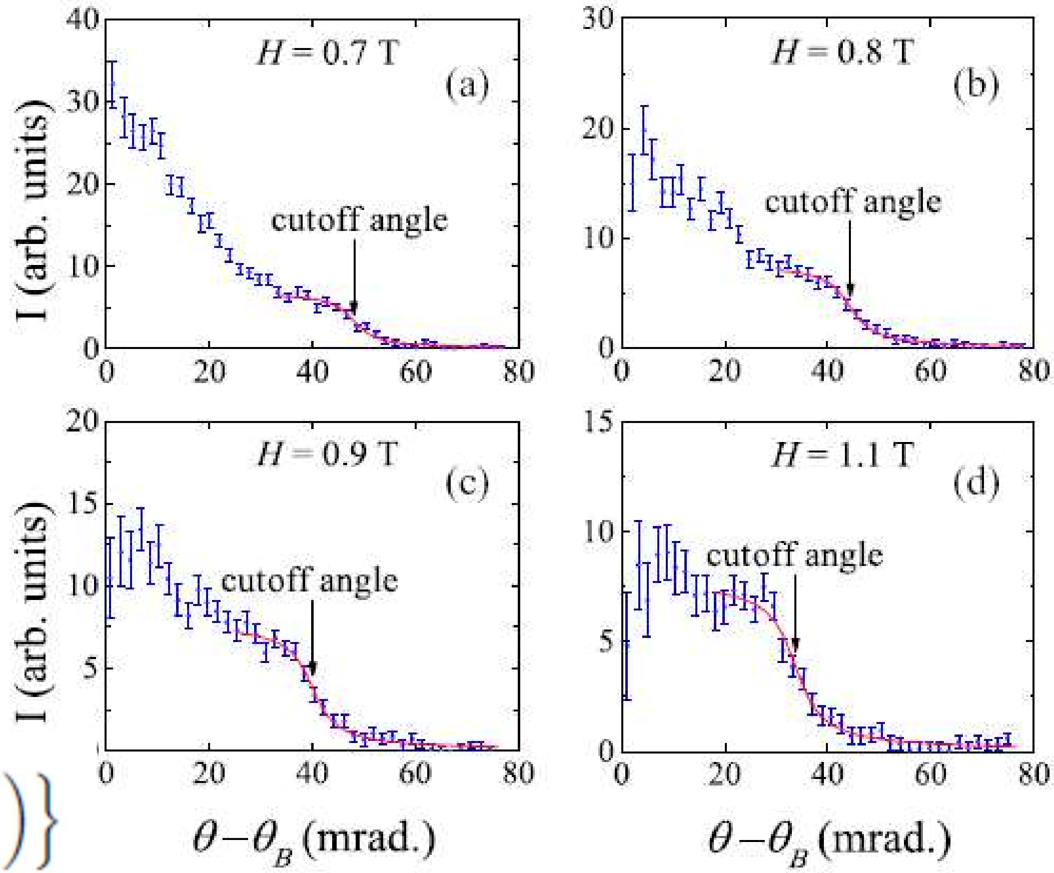
$$\Delta\sigma(\mathbf{q},\omega) = \sigma(\mathbf{q},\omega,+P_0) - \sigma(\mathbf{q},\omega,-P_0) =$$

$$= 4(r_m S)^2 F(\mathbf{q})^2 (\frac{k_f}{k_i})^2 (\mathbf{em})^2 n_q [\delta(\omega - \epsilon_q) - \delta(\omega + \epsilon_q)].$$

Field-evolution of anti-symmetric part of neutron scattering in MnSi

MnSi, T = 15 K



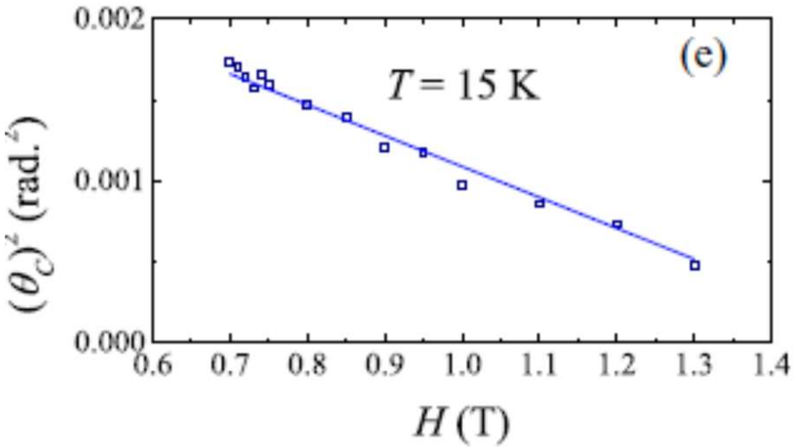


$$I(\theta) = I_0 \left\{ \frac{1}{2} - \left(\frac{1}{\pi} \arctan \left[\frac{2(\theta - \theta_C)}{\delta} \right] \right) \right\}$$

Spin wave stiffness of MnSi by polarized SANS

MnSi

Field dependence of cut-off angle

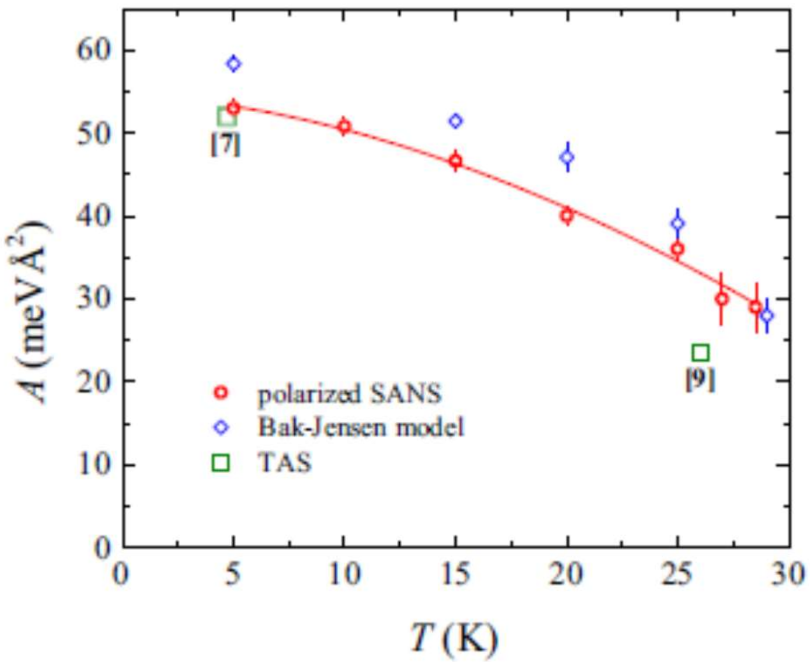


$$\theta_c^2 = \theta_0^2 - \theta_0 \frac{g\mu_B(H - H_{c2})}{E_i}$$

$$\theta_0 = \frac{\hbar^2}{2Am_n}$$ $A = \hbar^2/(2m_n\theta_0)$

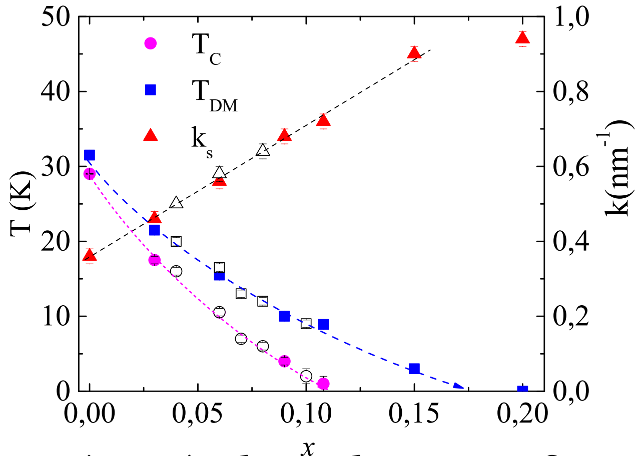
A = 48 meV A^2
T = 15 K (H> H_{C2})

Temperature dependence of spin wave stiffness

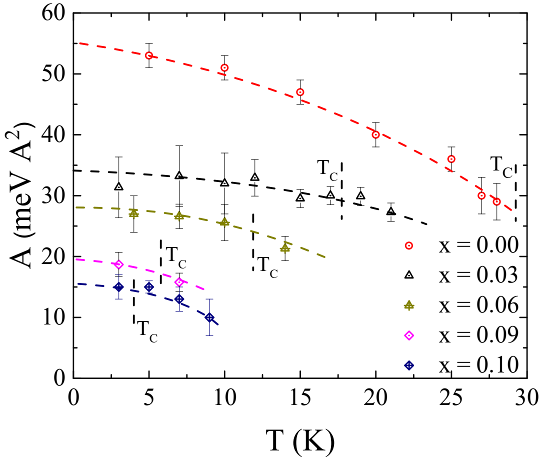


$$g\mu_B H_{C2} = Ak_s^2$$

Spin wave stiffness measurements in $Mn_{1-x}Fe_xSi$

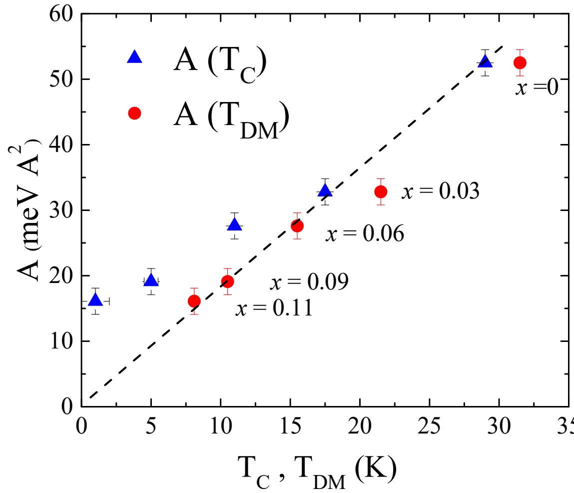


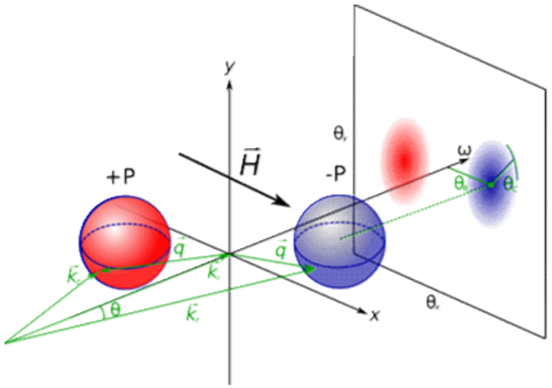
(T - x) phase diagram of **$Mn_{1-y}Fe_ySi$.**



Spin-wave stiffness of **$Mn_{1-y}Fe_ySi$.**

SW stiffness A vs **T_C , T_{DM}** in **$Mn_{1-y}Fe_ySi$.**





Гелимагнетик (H > H_{C2})

$\varepsilon_q = A(q - k)^2 + g\mu\,(H - H_{C2})$

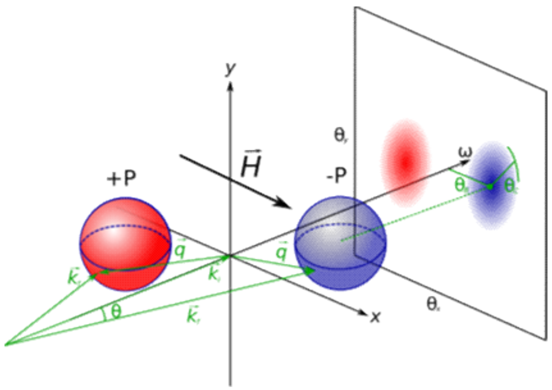
S. V. Grigoriev, E. V. Altynbaev, S.-A. Siegfried, K. A. Pschenichnyi, D. Menzel, A. Heinemann, and G. Chaboussant, Phys. Rev. B 97 (2018) 024409.

Conclusion (almost)

We have experimentally proven the validity of the spin-wave dispersion relation for helimagnets with the DM interaction in the full-polarized state (Mitsuo Kataoka, J.Phys.Soc.Jap. 56 (1987) 3635):

$$\varepsilon_{\mathrm{q}} = A(\mathbf{q} - \mathbf{k}_s)^2 + g\mu\,(H - H_{C2})$$

Small-angle polarized neutron scattering is shown to be a method to study spin waves in helimagnets.

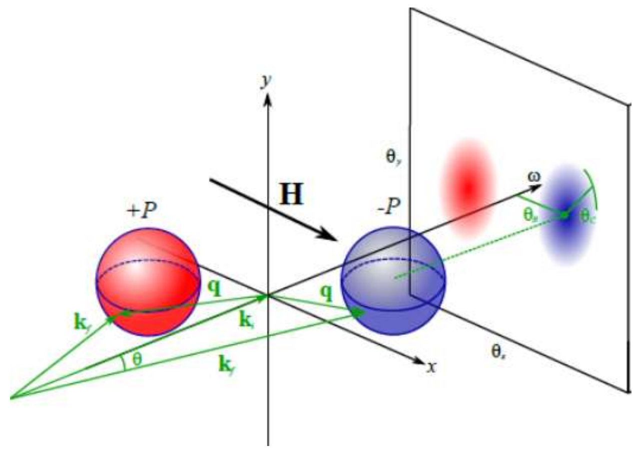


S. V. Grigoriev, A. S. Sukhanov, E. V. Altynbaev, S.-A. Siegfried, A. Heinemann, P. Kizhe, and S. V. Maleyev, Phys. Rev. B 92, 220415(R) (2015)

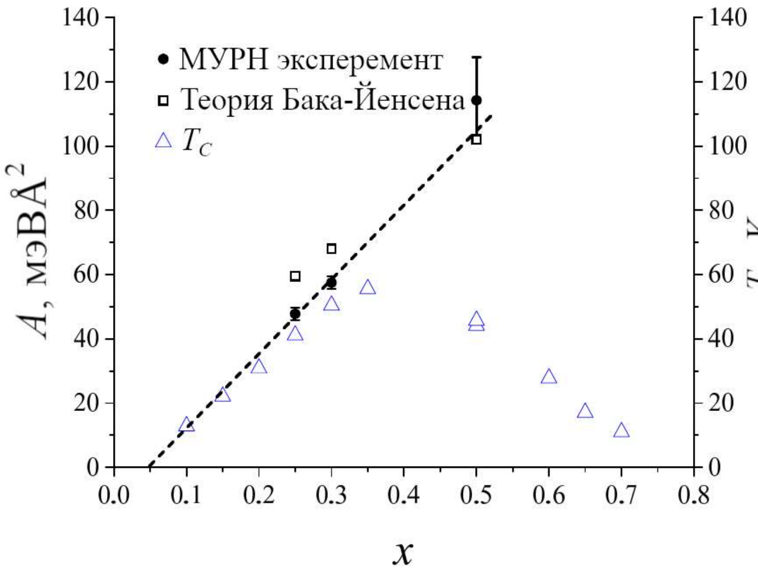
Taku J. Sato, Daisuke Okuyama, Tao Hong, Akiko Kikkawa, Yasujiro Taguchi, Taka-hisa Arima, Yoshinori Tokura, Phys. Rev. B,84(14),(2016),144420

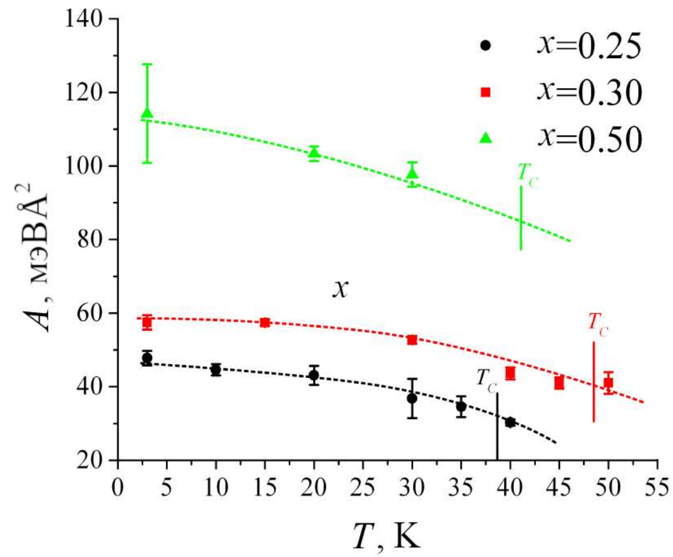
SANS experiments aiming to measure the spin-wave stiffness had been performed at

MnSi [Phys. Rev. B 92, 220415(R) (2015)],
$Mn_{1-x}Fe_xSi$ [Phys. Rev. B **97,** 024409 (2018)],
FeGe [Phys. Rev. B **95** , 134415 (2017)],
$Mn_{1-x}Fe_xGe$ [JMMM **459** 159-164 (2018)],
$Fe_{1-x}Co_xSi$ [JETP Letters, Vol. 107, No. 10, pp. 640–645 (2018)]
$Fe_{1-x}Co_xSi$ [Physical Review B vol. 100 N. 9 pp. 094409]
Cu_2OSeO_3 [Phys. Rev. B **99** (2019) 054427]

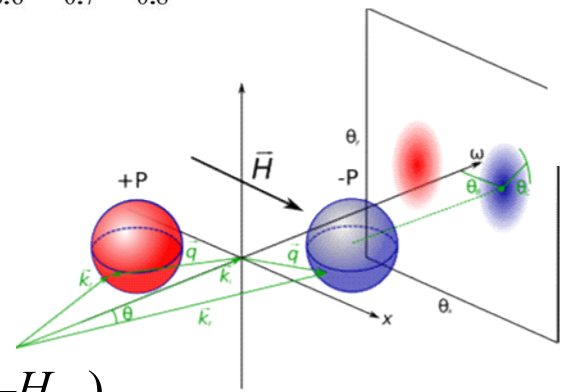


Spin wave stiffness measurements in $Fe_{1-x}Co_xSi$





(T - x) phase diagram of
$Fe_{1-x}Co_xSi$.

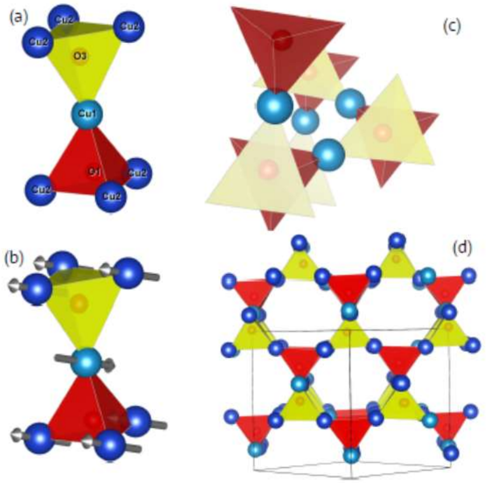


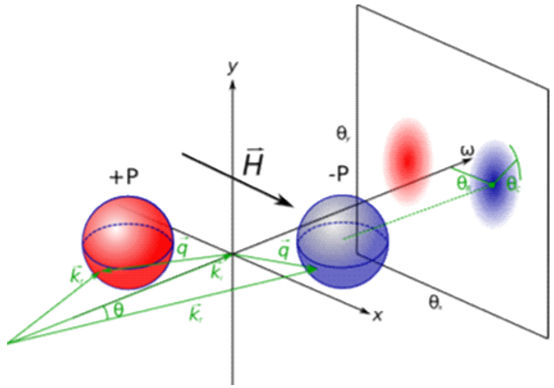
$$\varepsilon_q = A(\mathbf{q} - \mathbf{k}_s)^2 + g\mu\,(H - H_{C2})$$

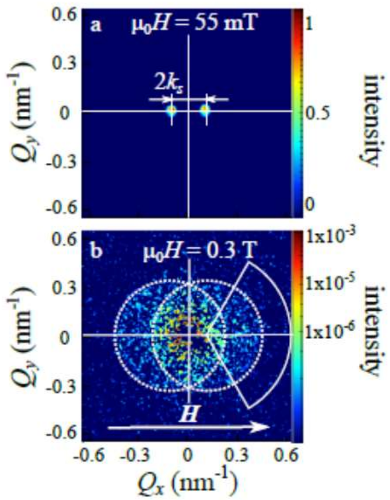
$Fe_{1-x}Co_xSi$ [JETP Letters, Vol. 107, No. 10, pp. 640–645 (2018)]
$Fe_{1-x}Co_xSi$ [Physical Review B vol. 100 N. 9 pp. 094409]

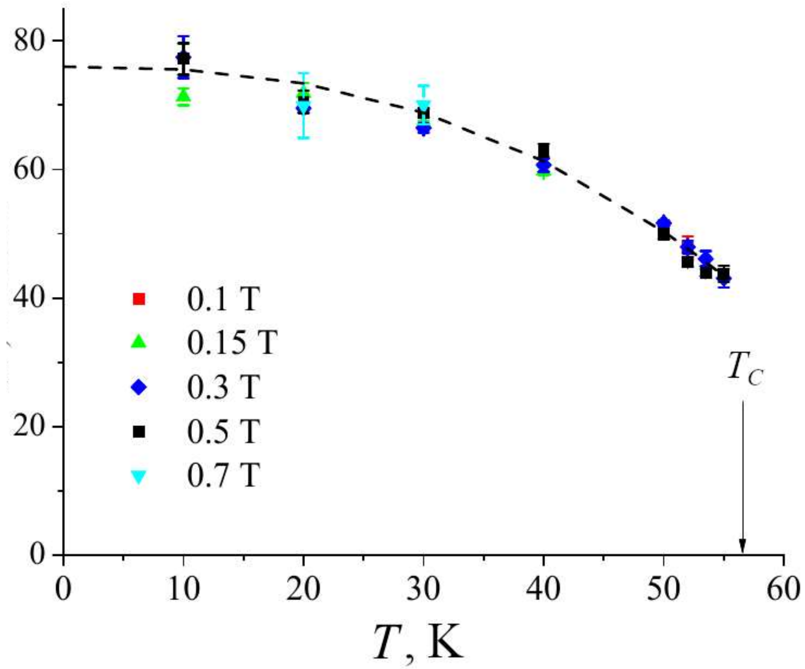
Spin wave stiffness measurements in $\mathbf{Cu_2OSeO_3}$

Crystal structure of $\mathbf{Cu_2OSeO_3}$









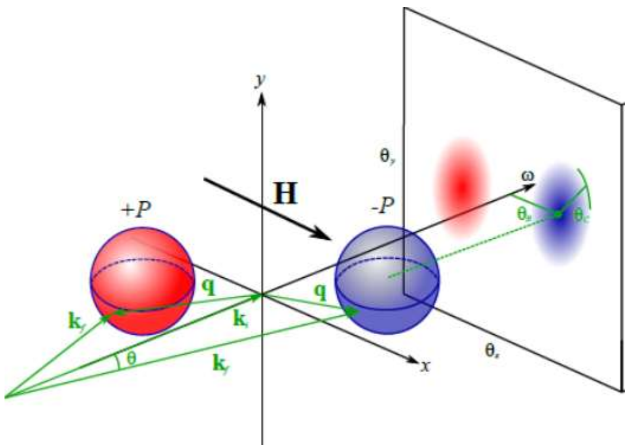
$$\varepsilon_q = A(\mathbf{q} - \mathbf{k}_s)^2 + g\mu\,(H - H_{C2})$$

Phys. Rev. B **99** (2019) 054427

Spin-wave stiffness of $\mathbf{Cu_2OSeO_3}$

SANS experiments aiming to measure the spin-wave stiffness had been performed at

MnSi [Phys. Rev. B 92, 220415(R) (2015)],
$Mn_{1-x}Fe_xSi$ [Phys. Rev. B **97,** 024409 (2018)],
FeGe [Phys. Rev. B **95** , 134415 (2017)],
$Mn_{1-x}Fe_xGe$ [JMMM **459** 159-164 (2018)],
$Fe_{1-x}Co_xSi$ [JETP Letters, Vol. 107, No. 10, pp. 640–645 (2018)]
$Fe_{1-x}Co_xSi$ [Physical Review B vol. 100 N. 9 pp. 094409]
Cu_2OSeO_3 [Phys. Rev. B **99** (2019) 054427]



$$\varepsilon_q = A(\mathbf{q} - \mathbf{k}_s)^2 + g\mu\,(H - H_{C2})$$

For all these compounds the ratio
$A = g\,\mu_B\,H_{C2}/k^2$
gives the value of spin-wave stiffness
A close to that **measured independently using small angle polarized neutron scattering**.

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ILL — Neutrons for Science

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A. Heineman

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