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PSI


Oksana Zaharko :: Laboratory for Neutron Scattering and Imaging, PSI, Switzerland

# *Exotic spin states in frustrated antiferromagnet $MnSc_2S_4$*

ESS-ILL User Meeting, Lund, 6 October, 2022

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# Main collaborators

*Shang Gao (PhD 2017) and Guratinder Kaur (PhD 2021) (PSI Villigen, Switzerland)*

*Diego Rosales and Flavia Gomez Albarracin (La Plata, Argentina)*

*Vladimir Tsurkan (Augsburg, Germany; Chisinau Moldova)*

## *Local contacts at Neutron Scattering Facilities:*

*T. Fennell, J. White, G. S. Tucker, B. Roessli, R. Sibille, LNS, SINQ, Villigen*

*J. Su, P. Cermak, A. Schneidewind, FRM2, Munich*

*E. Ressouche, S. Raymond, M. Boehm, P. Steffens, F. Bourdarot, ILL, Grenoble*

## *Publications:*

*S. Gao et al. Nature Physics 13, 157 2017*

*S. Gao et al. Nature 583, 37 2020*

*D. Rosales et al. PhysRevB 105, 224402 2022*

*K. Guratinder et al. PhysRevB 105, 174422 2022*

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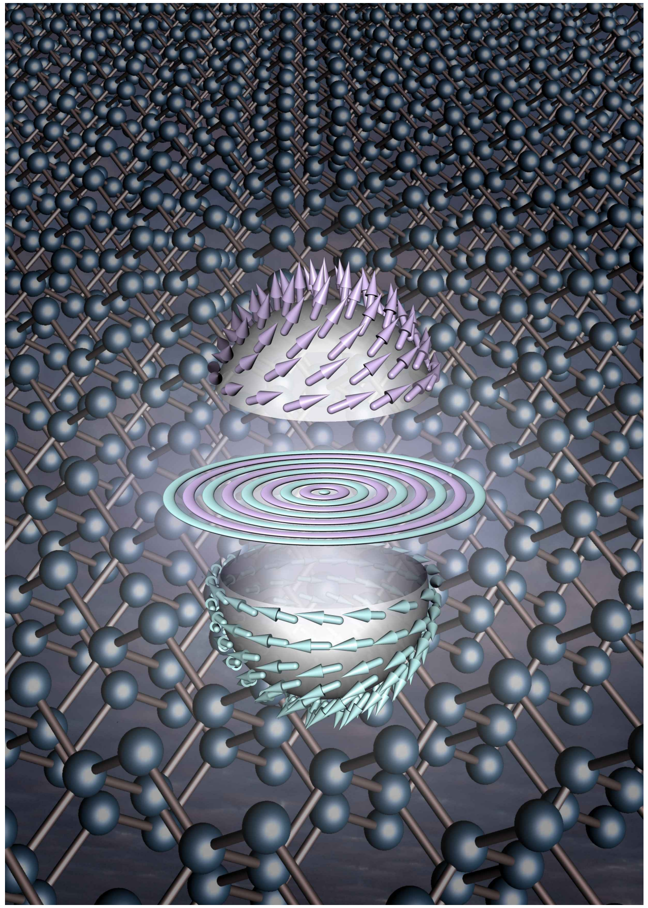
# Outline

Spiral spin liquids

Multi-step ordering in $MnSc_2S_4$

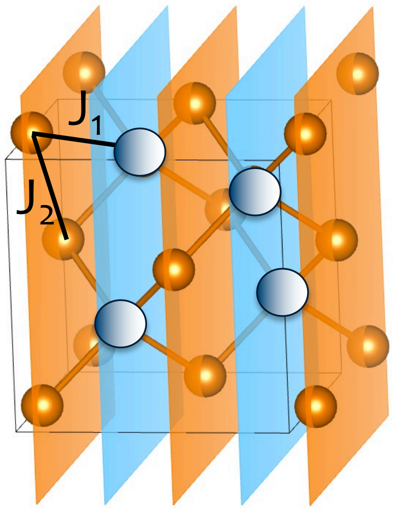
Exchange Hamiltonian of $MnSc_2S_4$

3k-structure and fractional AF skyrmion lattice



*Diego Rosales, Private Collection*

# Spiral spin liquid on diamond lattice




$$\boldsymbol{H} = J_1 \sum_{<ij>} \boldsymbol{S}_i \boldsymbol{S}_j + J_2 \sum_{<<ij>>} \boldsymbol{S}_i \boldsymbol{S}_j$$

D. Bergman et al. Nature Physics 3, 487(2007)

*J2 – AF, introduces frustration (fcc)*

*J1 - can increase frustration*

*two fcc sublattices*

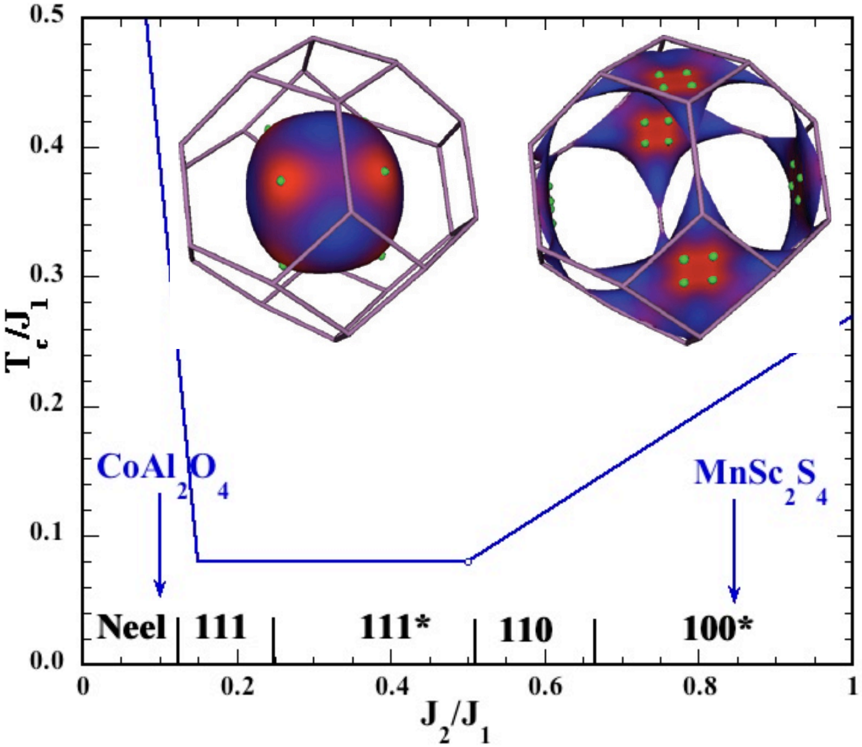
*T=0 : Ordering vectors lie on continuous surface in momentum space*

massive spiral degeneracy

*spins can distort from GS1 to GS2 with no E cost*

*entropy stabilizes LRO at finite T by lifting degeneracy of the free energy along the spiral surface*

order-by-disorder



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# Spin spirals on bipartite lattices

*T. A. Kaplan Phys. Rev. 116, 888(1959)*

$$\boldsymbol{H} = J_1 \sum_{<ij>} \boldsymbol{S}_i \boldsymbol{S}_j + J_2 \sum_{<<ij>>} \boldsymbol{S}_i \boldsymbol{S}_j$$

## Lattices

*square* — *C. L. Henley PRL 62, 2056 1989*

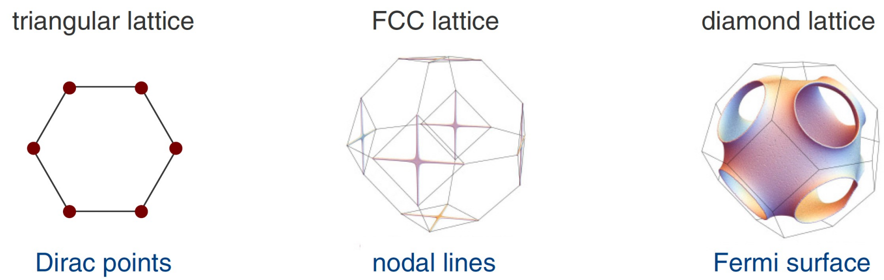
*diamond* — *S. Gao et al. Nature Physics 13, 157 2017 $MnSc_2S_4$*

*honeycomb* — *A. Mulder et al. PRB 81, 214419(2010)*
*S. Gao et al. PRL 128, 227201(2022) $FeCl_3$*

*Correspondence between*

*Spin spiral surface of frustrated AF with*

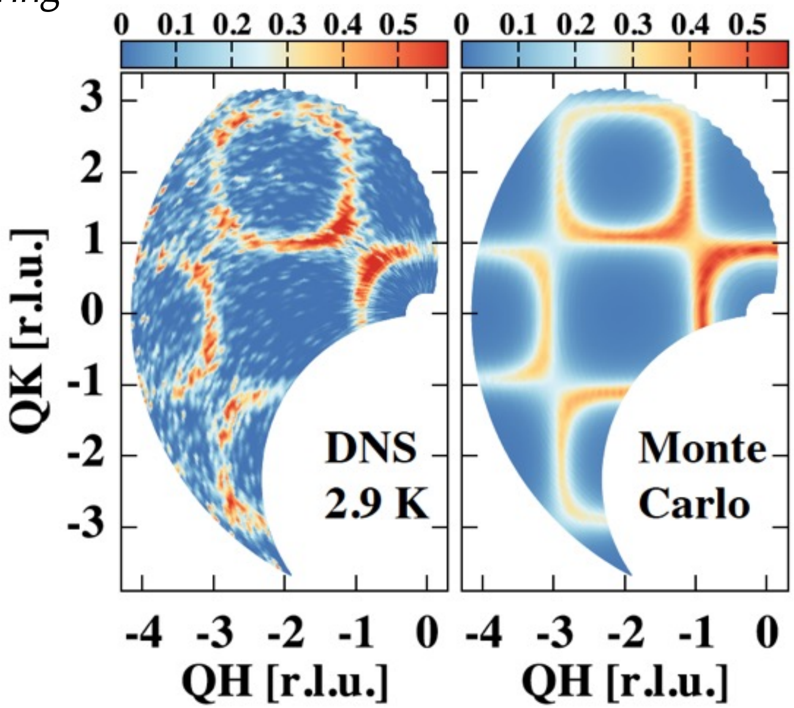
*Fermi surface of noninteracting fermion systems*

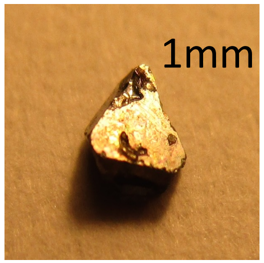



*J. Attig , S. Trebst PRB 96, 085145(2017)*

# AF spinel MnSc2S4 and spiral spin liquid

*Neutron diffuse scattering*




*Classical MC calculation*

*8x10x10x10 atoms S=5/2*

*J1-J2 Heisenberg Hamiltonian*

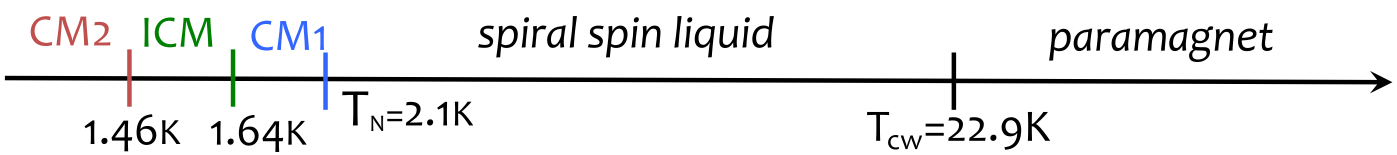
*|J2/J1|=0.85*

*T/|J1|=0.55*

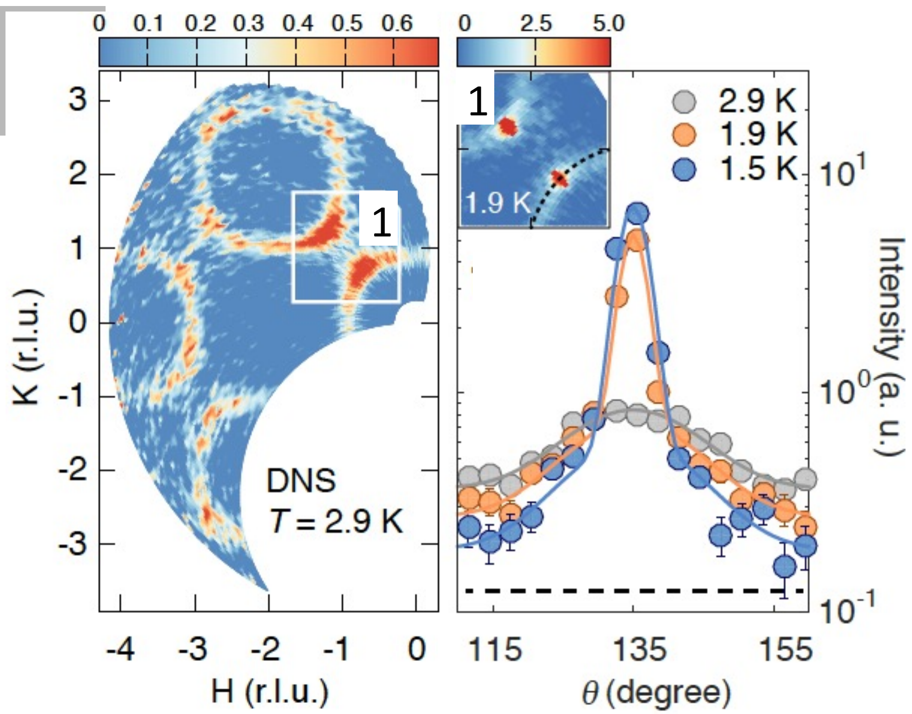
*J1 FM, J2 AF*

*small AF J3= -0.1J1*

*induces LRO* *k=¾ ¾ 0*



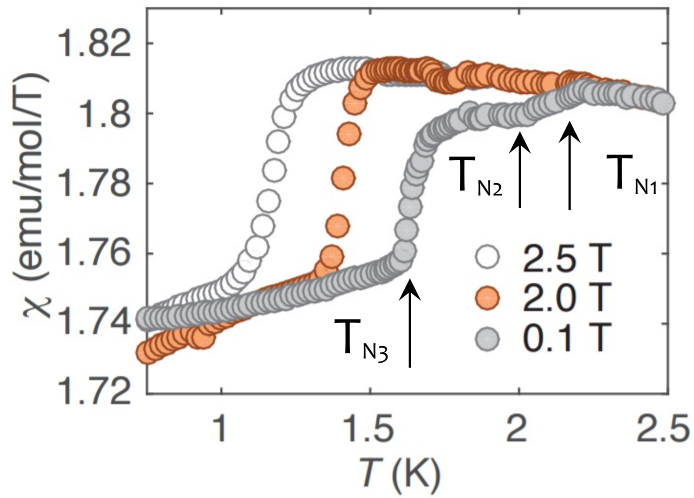
# AF spinel $MnSc_2S_4$ and multistep ordering at H=0

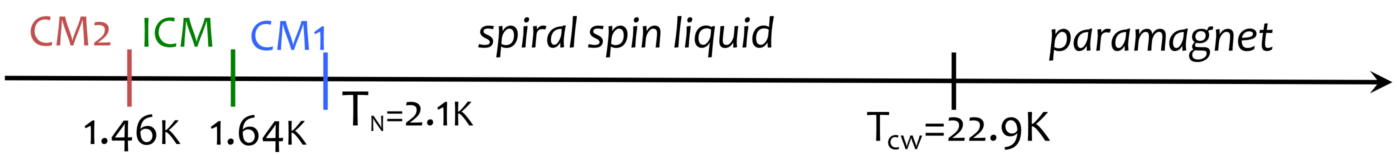



$T < T_N$ $k = ¾\ ¾\ 0$

$T_2 < T < T_1$ $k = ¾ \pm \delta\ ¾ \mp \delta\ 0$

$T < T_2$ $k = ¾\ ¾\ 0$



# AF spinel $MnSc_2S_4$ and multistep ordering

*Neutron single crystal diffractometer ZEBRA*

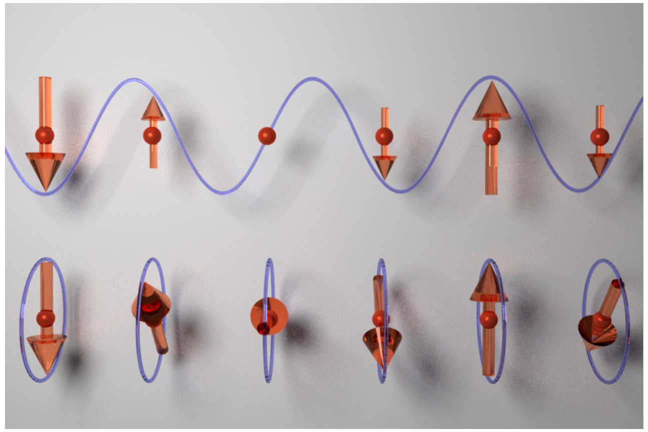
CM1 *amplitude modulated*

*transversal to k*

$M_{1\text{-}10}$ *for k=(¾ ¾ 0)*

CM2 *helical structure transversal to k*

$M_{1\text{-}10}$ $M_{001}$ *for k=(¾ ¾ 0)*

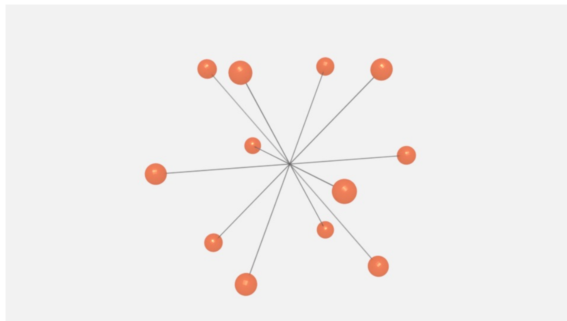


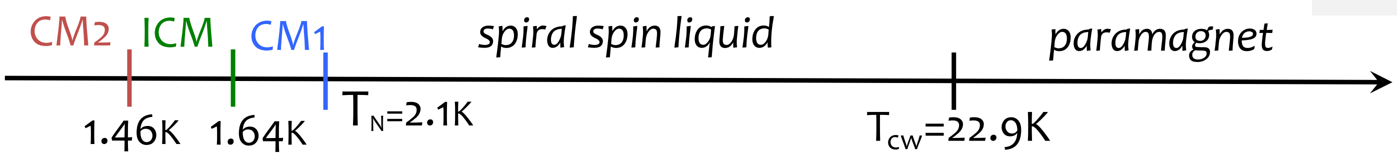
*Cubic, Fd-3m, $Mn^{2+}$ in 8a (1/8 1/8 1/8)*

*k=(¾ ¾ 0) the star has 12 arms, –k ≠k*

*12 domain of single-k*

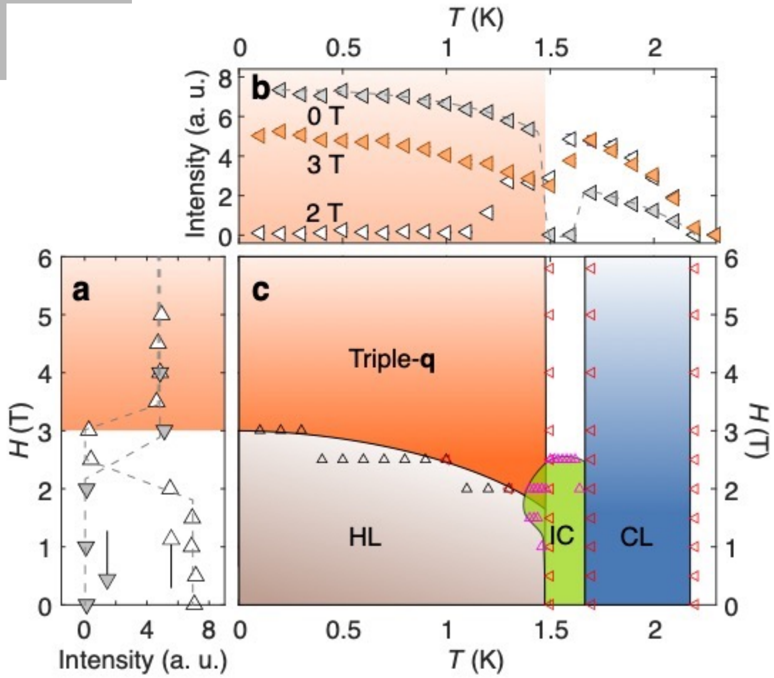
*4 domains of triple-k*



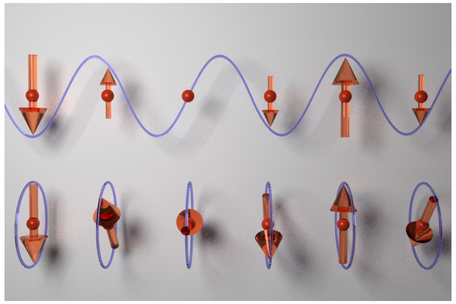


# AF spinel MnSc2S4 and magnetic field

*Phase diagram H//001*




*Magnetic field favors states with highest magnetic susceptibility χ*



*Collinear domains with spins ⊥H*

*Helical domains with the spin-plane ⊥H*

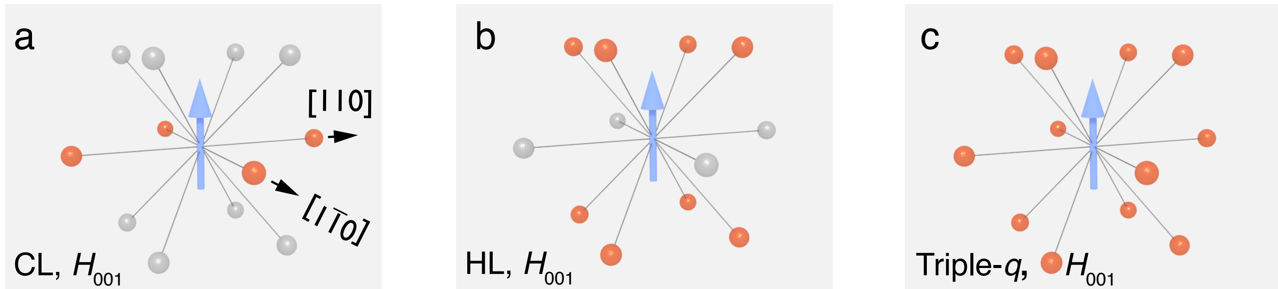
*Triple-k structure:*

*Σ (k1 + k2 + k3) = 0*

*k1 = [¾ ¾ 0] k2 = [-¾ 0 -¾] k3 = [0 -¾ ¾]*

*and –k1 –k2 –k3*

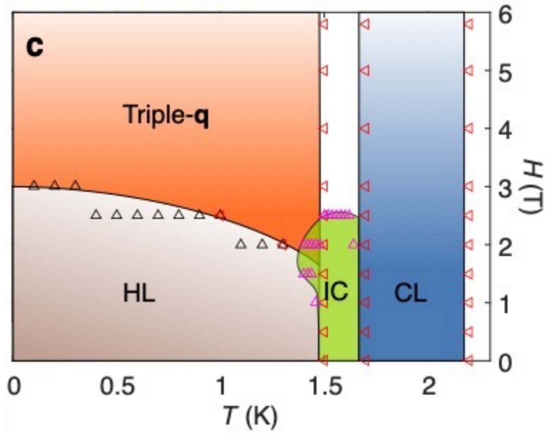
*Expected magnetic reflections*

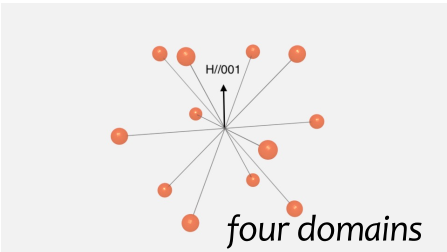


# AF spinel MnSc2S4 and experimental (H,T) phase diagrams

H // 001

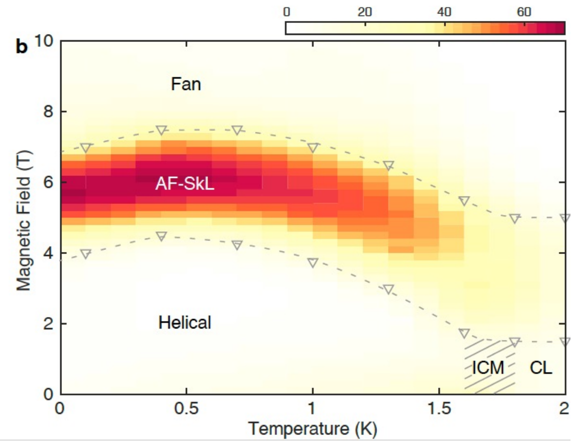
*ZEBRA*

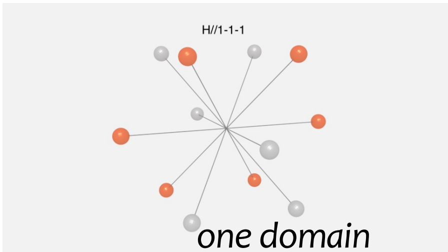



H // 111

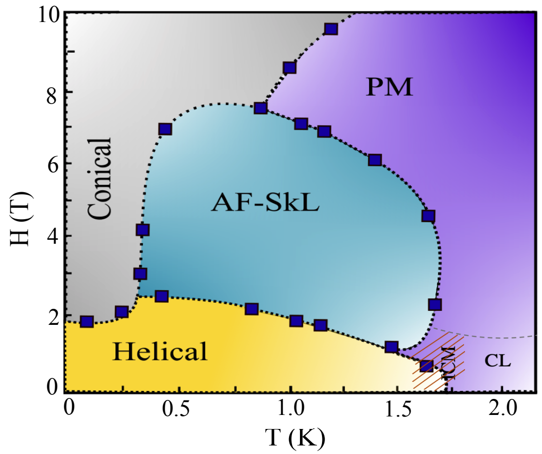
D23

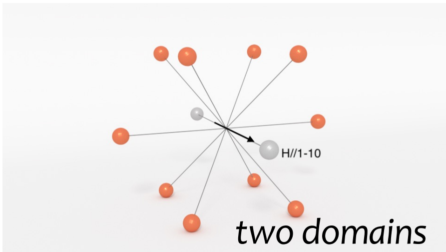



H // 110

*ZEBRA*



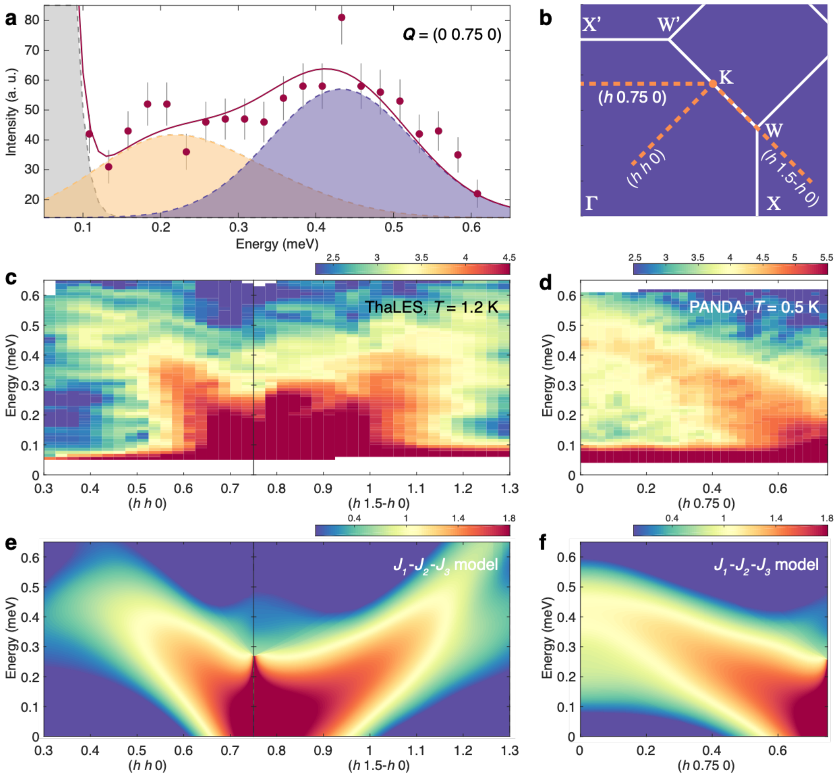



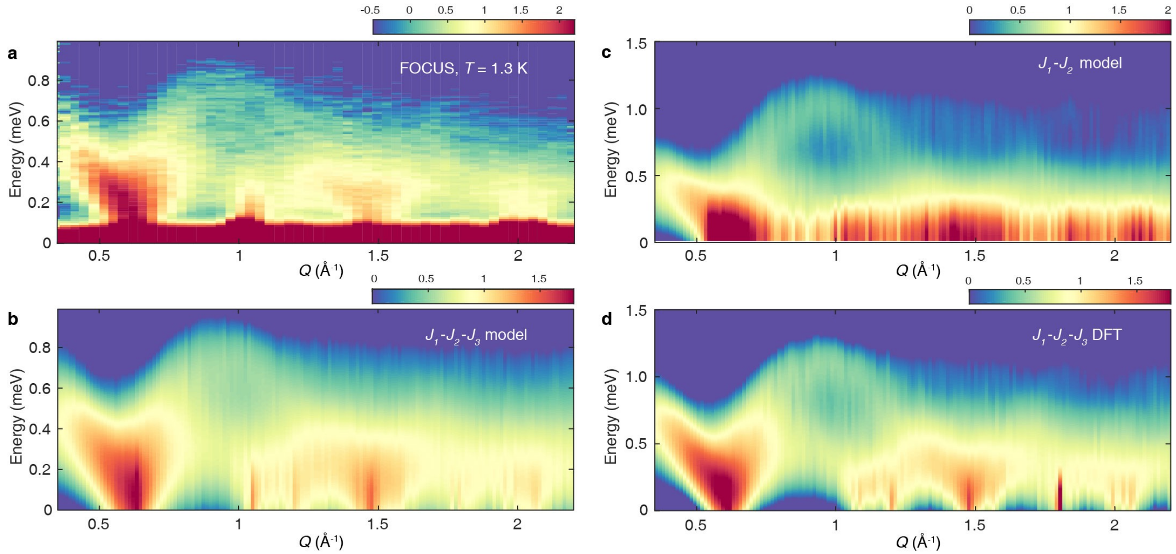
# AF spinel MnSc2S4: INS

$$\mathcal{H} = \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + 3\, J_{||} \sum_{ij \in NN} (\mathbf{S}_i \cdot \hat{r}_{ij})\,(\mathbf{S}_j \cdot \hat{r}_{ij})$$

$$+ A_4 \sum_{i,\alpha=x,y,z} (S_i^{\alpha})^4 - g\mu_B\, \mu_0 \boldsymbol{H} \sum_i \mathbf{S}_i$$

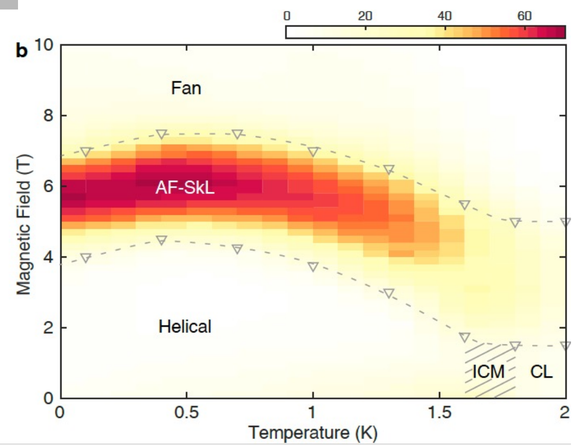
$J_1$= -0.31, $J_2$= 0,46, $J_3$= 0.087 K

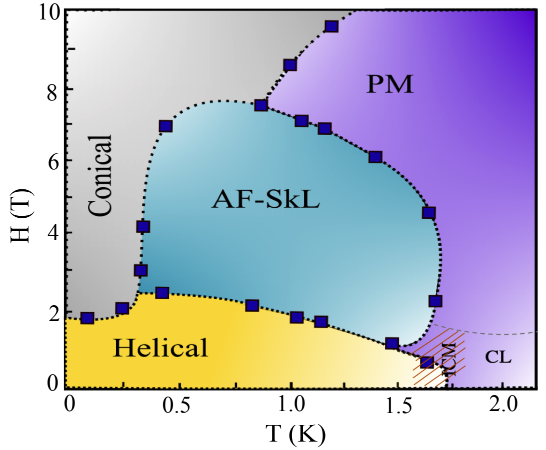


# AF spinel MnSc2S4: Monte-Carlo study

$$\mathcal{H} = \sum_{ij} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j + 3\,J_{||}\sum_{ij\in NN}(\mathbf{S}_i\cdot\hat{r}_{ij})(\mathbf{S}_j\cdot\hat{r}_{ij})$$
$$+A_4\sum_{i,\alpha=x,y,z}(S_i^\alpha)^4 - g\mu_B\,\mu_0\boldsymbol{H}\sum_i\mathbf{S}_i$$



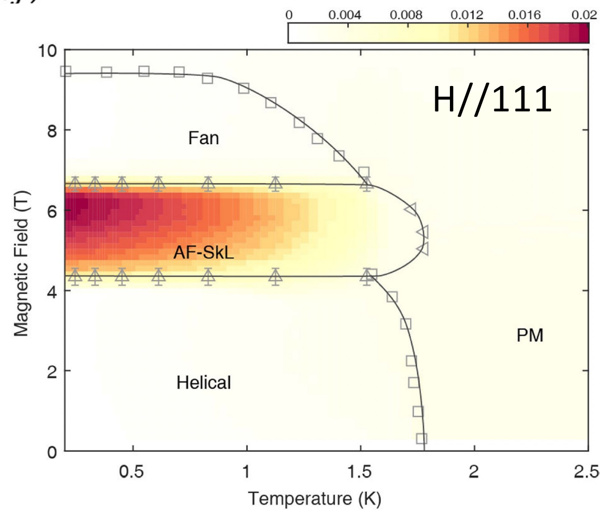

*$J_1$= -0.31, $J_2$= 0,46, $J_3$= 0.087 K*

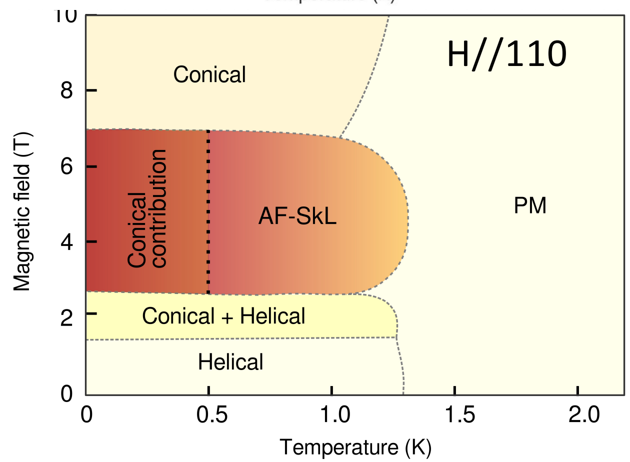
*$J_{||}$= -0.01K, $A_4$= 0.0016 K*

*Total scalar spin chirality*

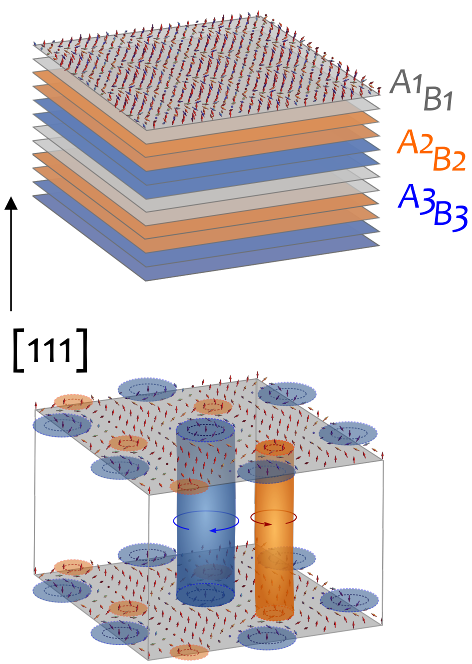
$$\chi_{tot} = \langle\frac{1}{N}\sum_n \chi_n\rangle$$
$$\chi_n = \mathbf{S}_i\cdot(\mathbf{S}_j\times\mathbf{S}_k)$$

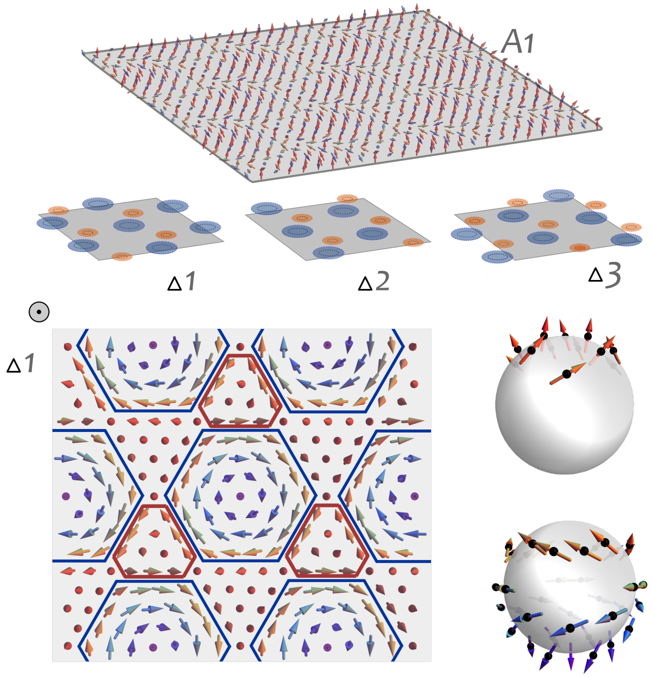
*fractional AF skyrmion lattice*

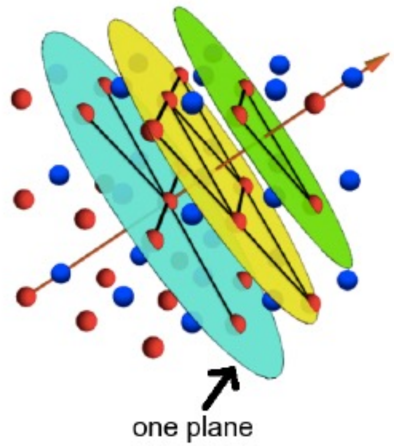


# AF spinel MnSc2S4: AF SkL description




*A and B – two fcc sublattices (t= 1/4 1/4 1/4)*

*A1, A2, A3 – triangular layers (t= 1/2 1/2 0)*

*△1, △2, △3 – sublayer with AF SkL*

$$\mathbf{S_r} = \mathbf{S_r}^{\perp} + \mathbf{S_r}^{[111]} \qquad \mathbf{S_r}^{\perp} = \frac{1}{n_s} A^{\perp} \sum_{i=1}^{3} sin(\mathbf{Q}_i \cdot \mathbf{r} + \theta_i)\mathbf{e}_i$$

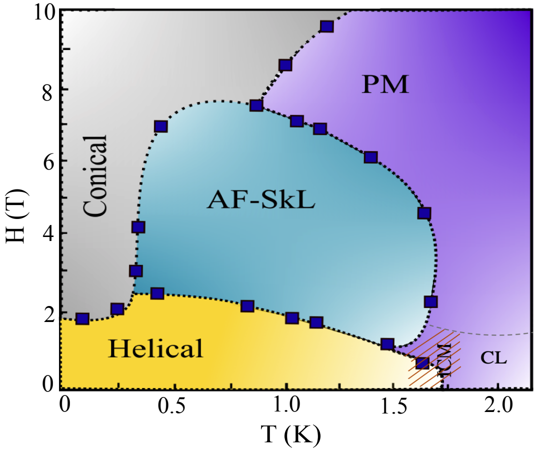
$$\mathbf{S_r}^{[111]} = \frac{1}{n_s}[M^{[111]} + A^{[111]} \sum_{i=1}^{3} cos(\mathbf{Q}_i \cdot \mathbf{r} + \phi_i)]\mathbf{e}^{[111]}$$

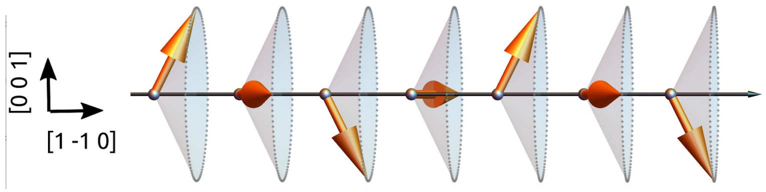
*distance between cores ~ 55 A*

*Bloch type*

# AF spinel MnSc2S4: locking of AF SkL axis

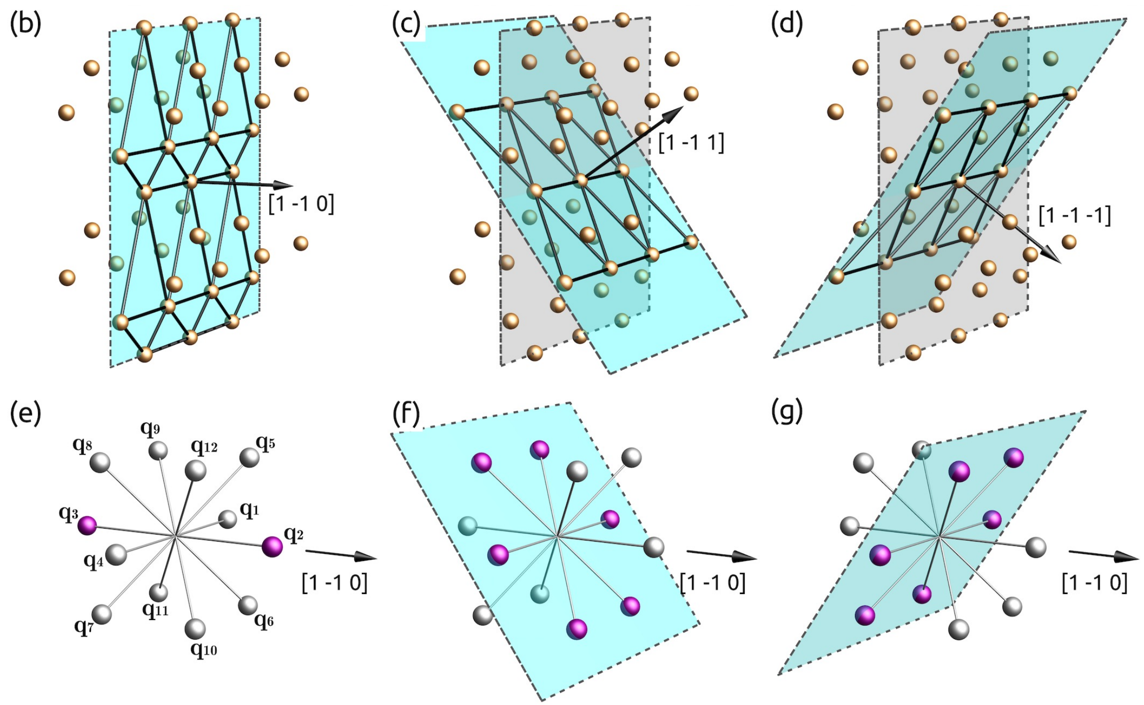
H // 110




*Conical phase*

*Two domains of AF SkL phase*



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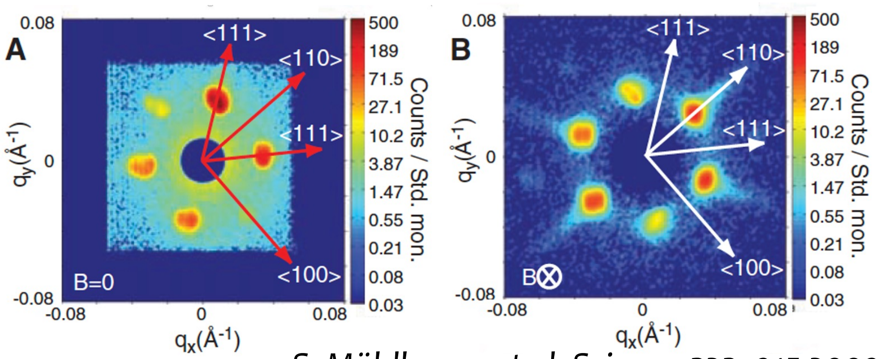
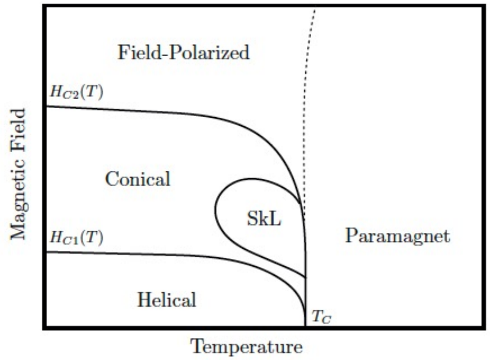
# MnSc2S4 and other SkL



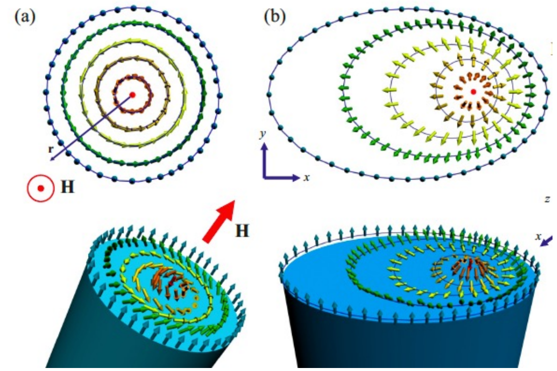

S. Mühlbauer et al. Science 323, 915 2009
SANS experiment

*MnSi, $Cu_2OSeO_3$ B20-family*
*soft chiral ferromagnets*
*magnetic helix ~190 A, skyrmions of the Bloch type*
*$P2_13$ non-centrosymmetric*
*FM interaction > DM interaction > anisotropy*




$MnSc_2S_4$
*dominant AF coupling*
centrosymmetric, as $Gd_2PdSi_3$
tiny anisotropy has strong effect
fractional Sk, $d_{cores}$ ~ 55 A

$GaV_4X_8$ (X=S, Se) F-43m – R3m $T_s$ = 42 K four polar domains
orientation confinement of SkL to the anisotropy axis
skyrmions of the Neel type distort and core displaces
SkL region is larger then in the B20 family




I. Kezsmarki et al. Nature Physics 14, 1116 2015
A. O. Leonov, I. Kezsmarki, PhysRevB. 96, 214413 2017

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