Technical Aspects and Practices in Using Neutron Total Scattering for Local Ordering Studies in Magnetic Systems

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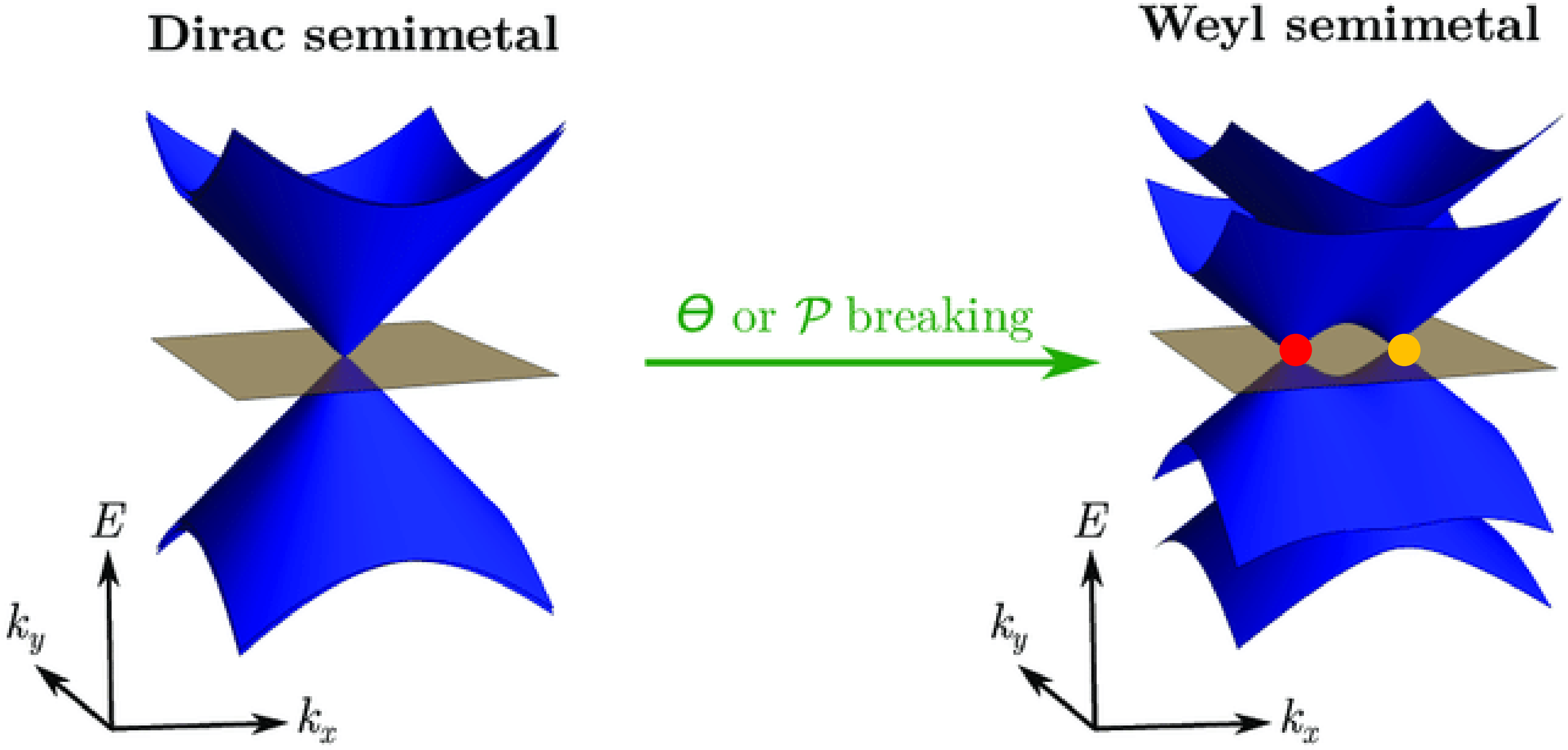
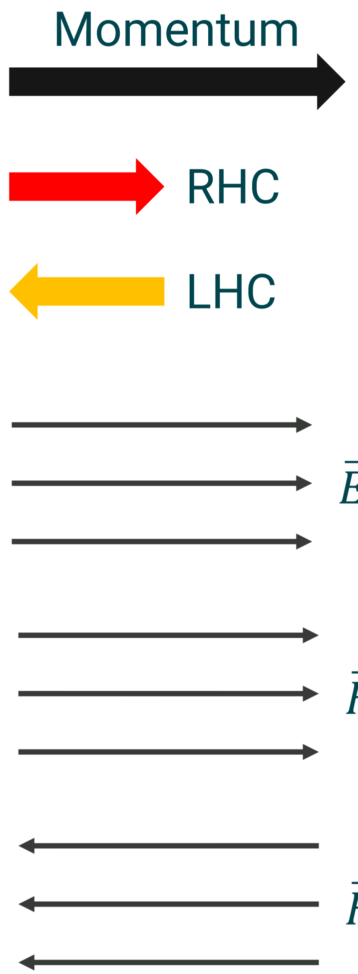
ADD 2026 Grenoble

Outline

- Spin Lattice Coupling Cases
  - Hidden local symmetry breaking in Kagome-lattice $Co_3Sn_2S_2$
  - Local distortion driven magnetic phase switching in pyrochlore $Yb_2(Ti_{1-x}Sn_x)_2O_7$
- Local Magnetic Ordering with Total Scattering
  - Technical Aspects
  - Local Spin Cluster in $Sn_xFe_{4-x}N$ Spin Glass

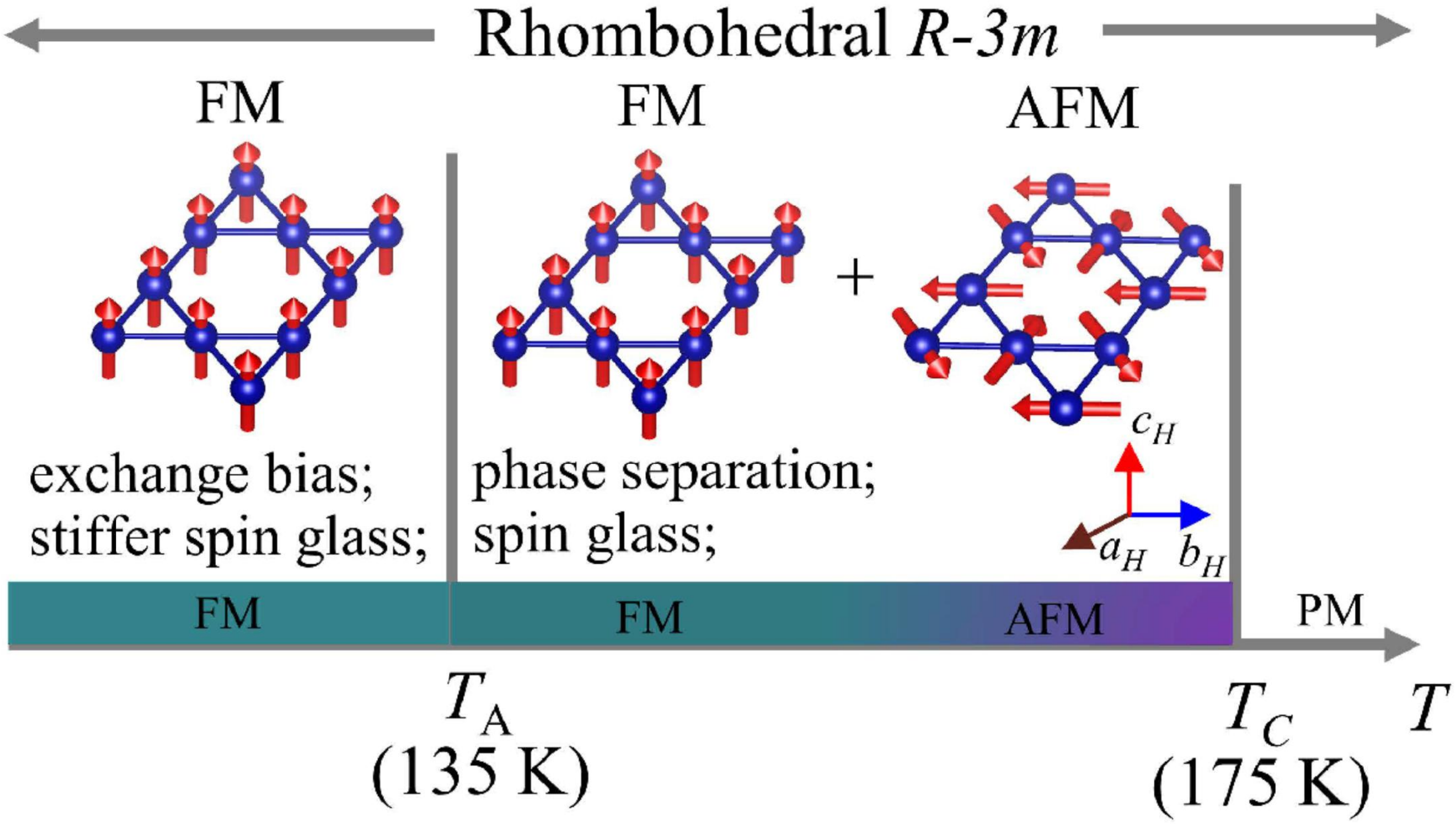
Hidden local symmetry breaking in Kagome-lattice $Co_3Sn_2S_2$





A. Lau, Symmetry-enriched topological states of matter in insulators and semimetals

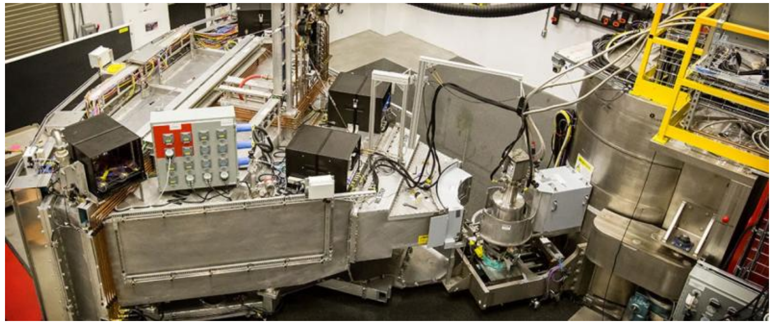
Magnetic Transition Pathway for $Co_3Sn_2S_2$



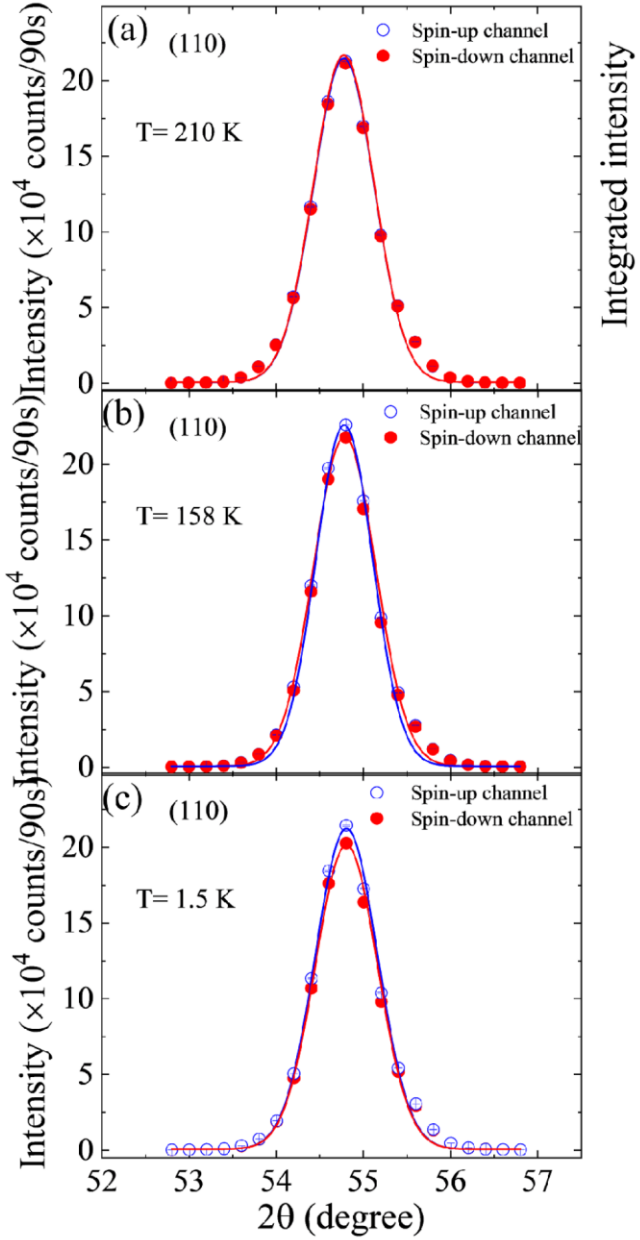
Q. Zhang, **Y. P. Zhang**, *et al*. *J. Am. Chem. Soc.* 2022, **144**, 31, 14339−14350

Polarized Neutron Data

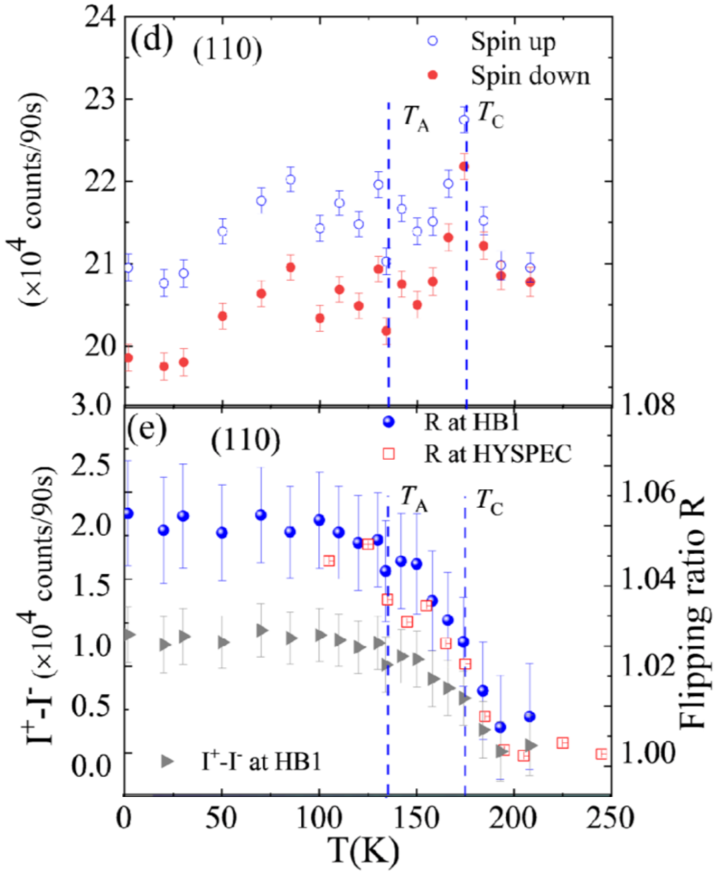
HB1 @ HFIR, ORNL



HYSPEC @ SNS, ORNL



Integrated Int.



Flipping Difference (Left)
&
Flipping Ratio (Right)

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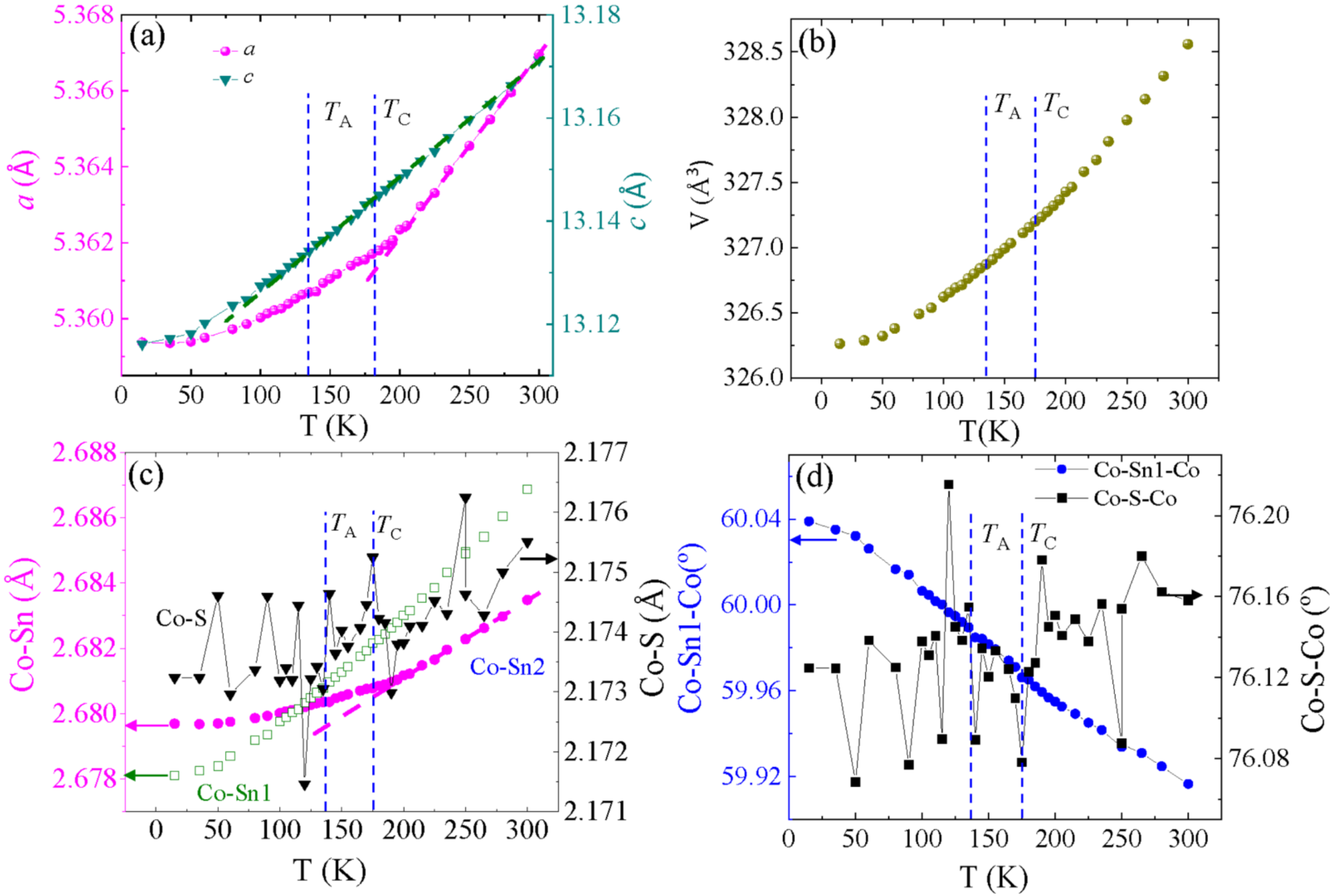
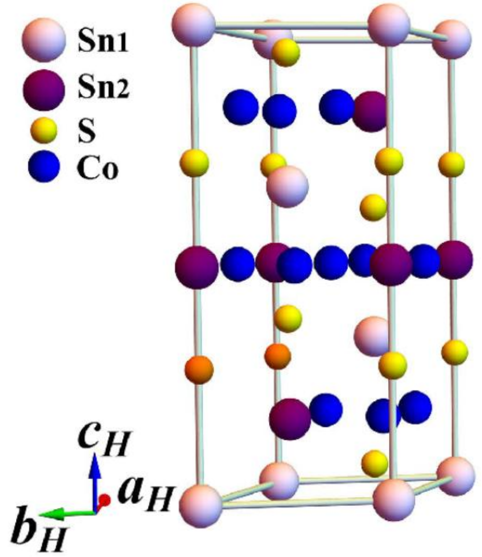
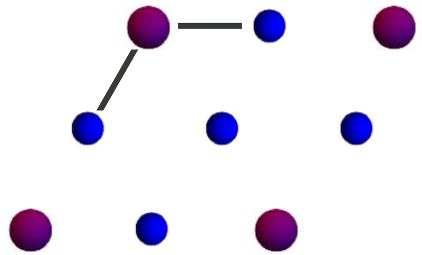
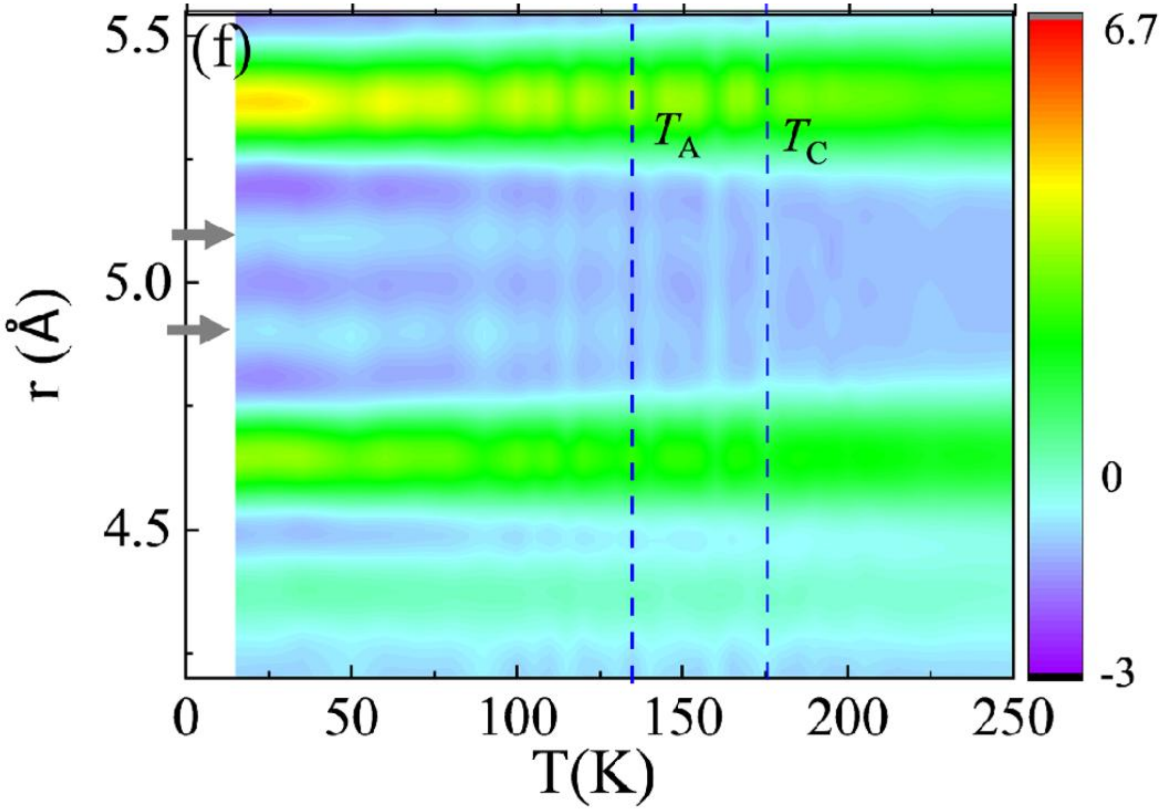
Average Structure





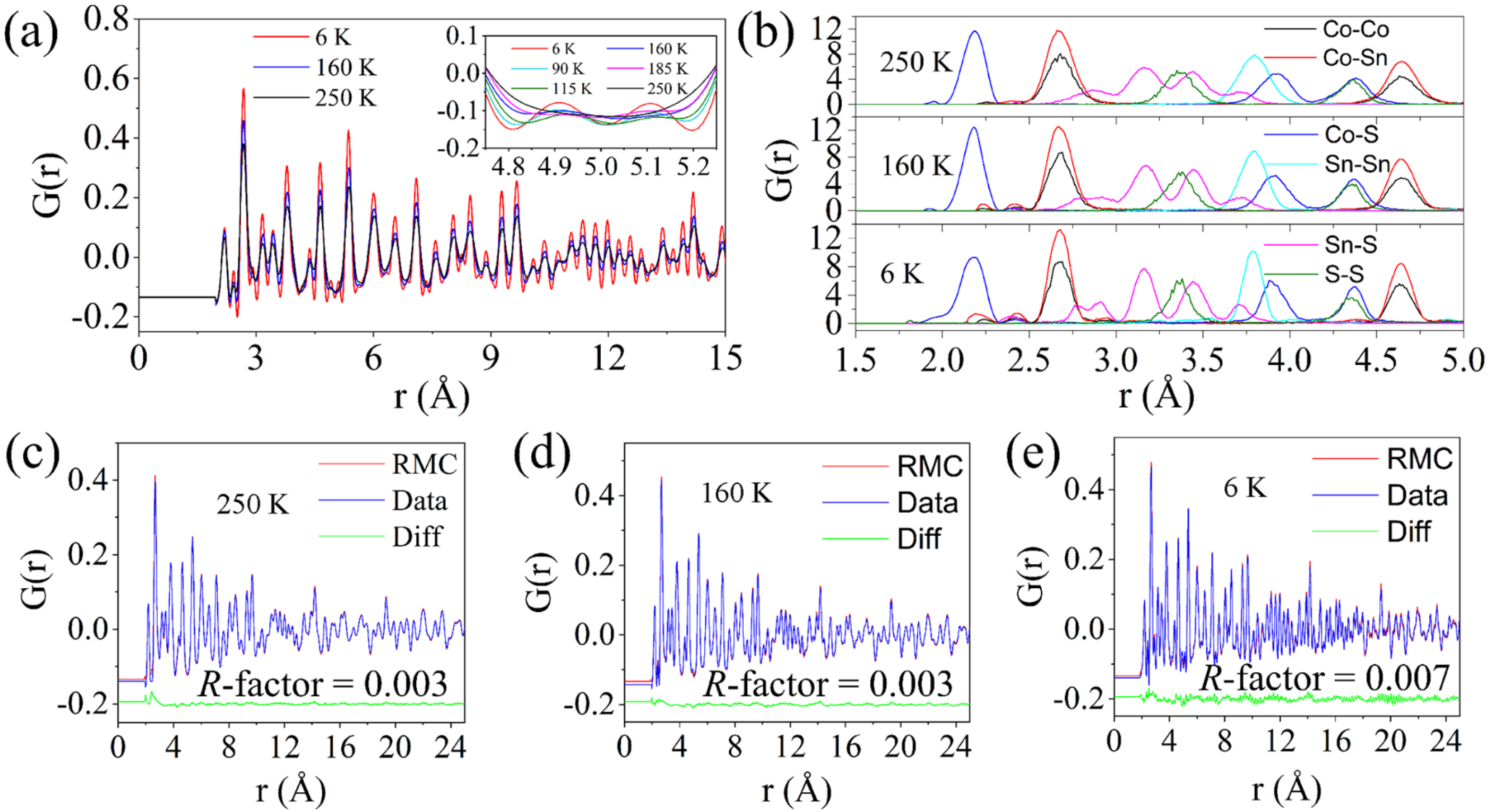


Local Structure Data



POWGEN @ SNS, ORNL

Local Structure Modeling



Metropolis RMC

$$\frac{e^{-\beta E_i}}{\mathcal{Z}}, \beta = \frac{1}{k_B T}$$

$$P_i = \frac{e^{-\beta E_i}}{\mathcal{Z}}$$

$$P_j = \frac{e^{-\beta E_j}}{\mathcal{Z}}$$

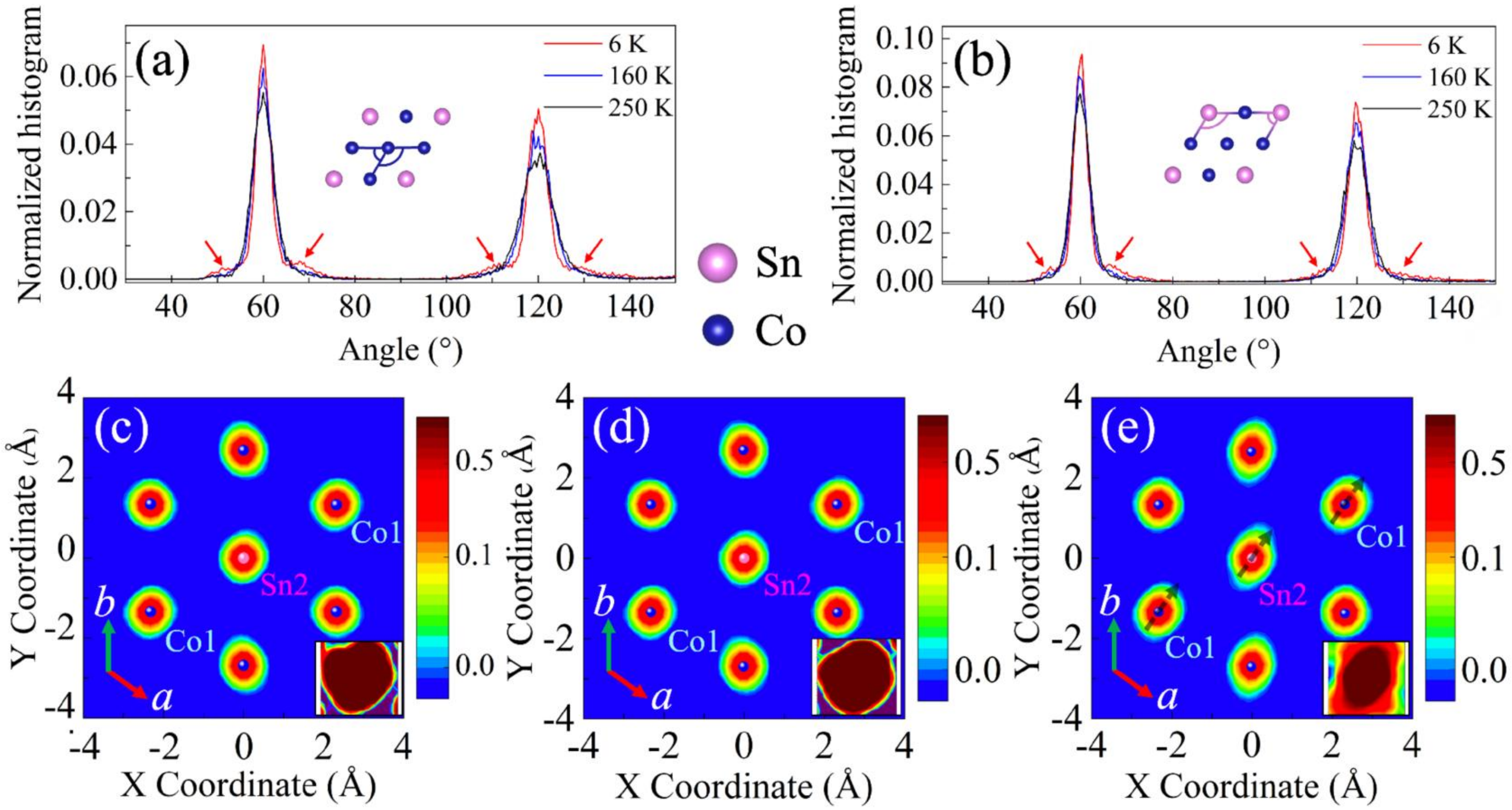
$$\frac{P_j}{P_i} = e^{\beta(E_i - E_j)}$$

$$E \Rightarrow \chi^2$$

Local Structure Modeling

Bond Angle Distribution

Kernel Density Estimation (KDE)

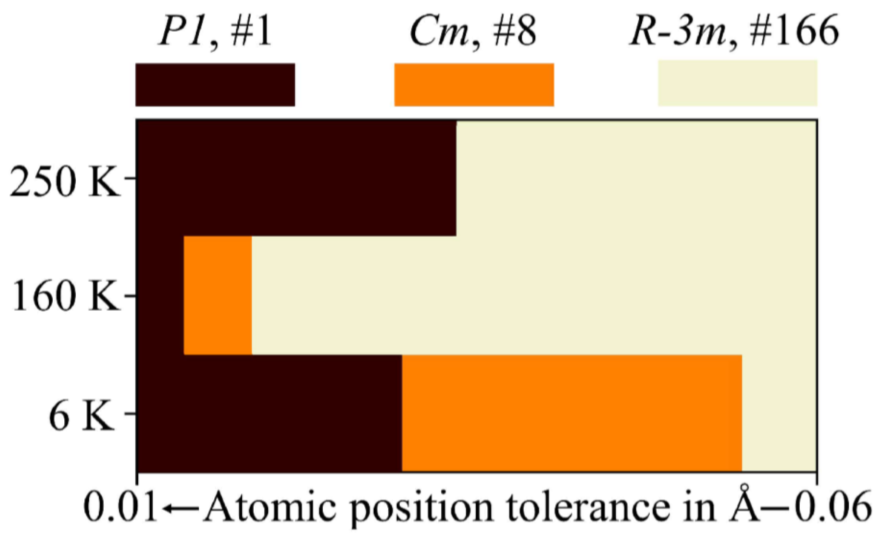


250 K

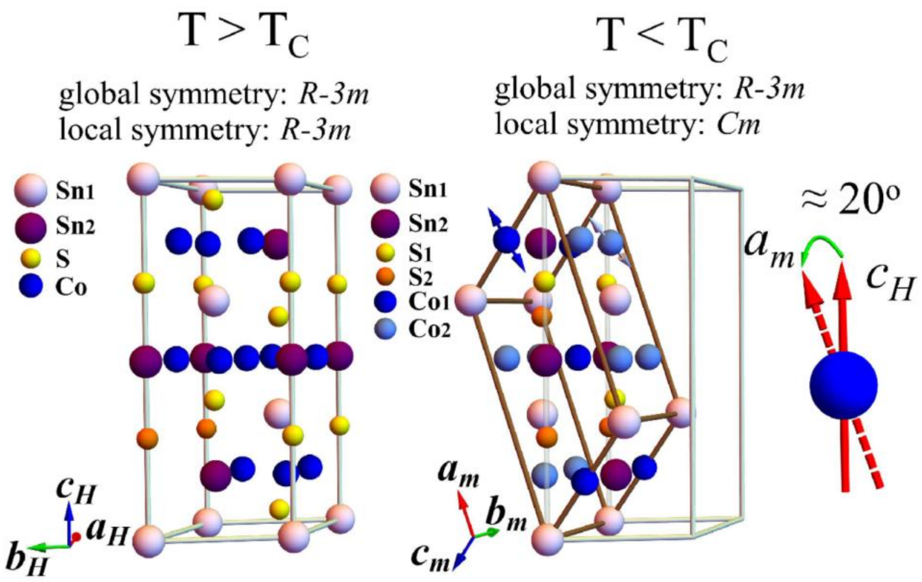
160 K

6 K

Local Structure Modeling



Supercell ⇒ unit cell ⇒ *findsym*



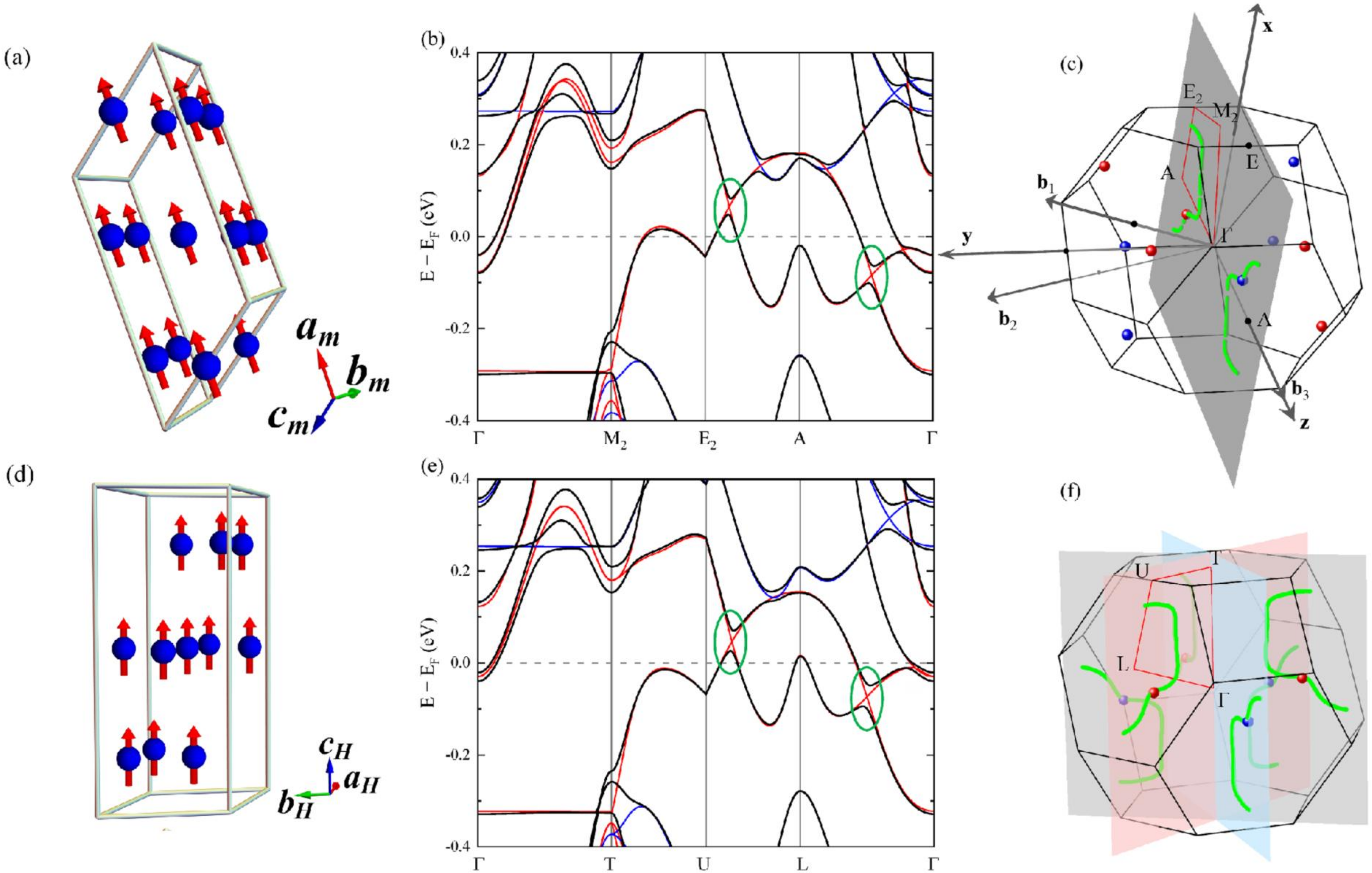
Identified local symmetry above (left) and below (right) T_C

H. T. Stokes, D. M. Hatch, and B. J. Campbell, FINDSYM, ISOTROPY Software Suite, iso.byu.edu.
H. T. Stokes and D. M. Hatch, J. Appl. Cryst. 38, 237-238 (2005).

Impact of Local Symmetry Breaking on Magnetic Ordering & Weyl Points

$< T_C$

$> T_C$

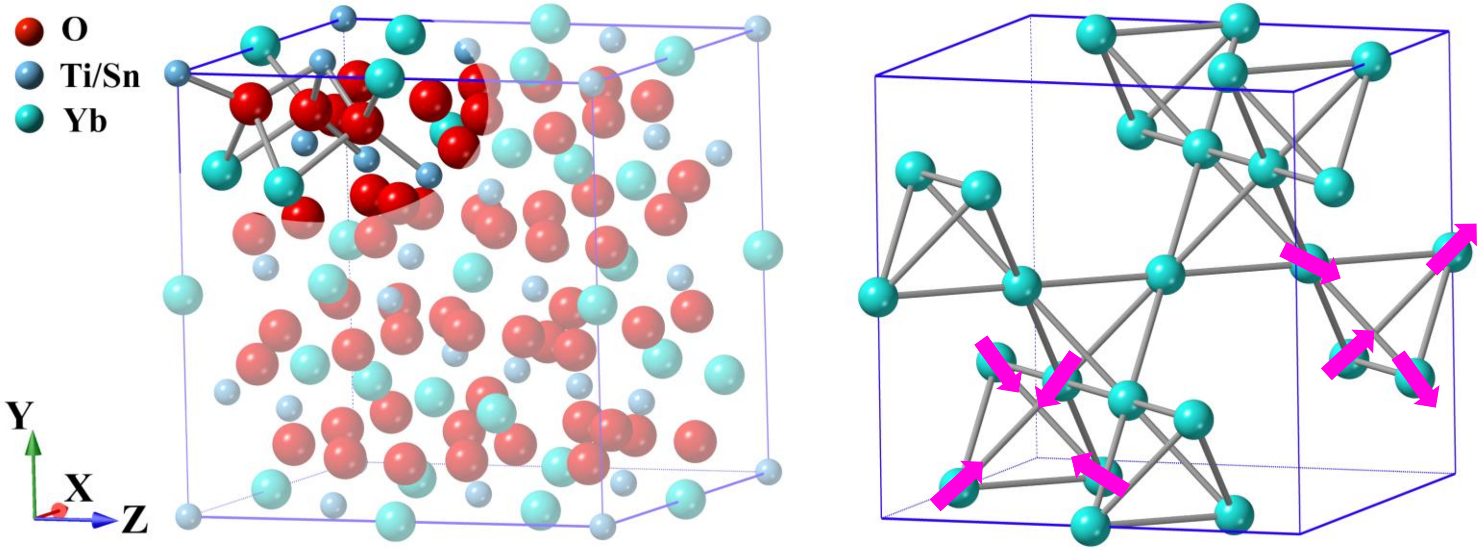


Q. Zhang, **Y. P. Zhang**, *et al. J. Am. Chem. Soc.* 2022, **144**, 31, 14339−14350

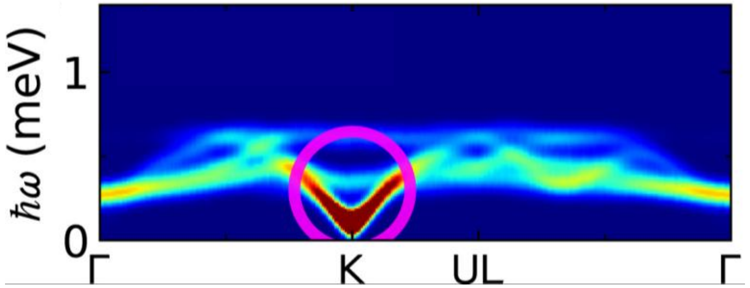
Take-Away

- ❖ Magnetic transition induces LSB
- ❖ LSB further induces FM ordering instability
- ❖ FM instability then possibly accounts for the emergence of the spin glass behavior
- ❖ LSB has impacts upon the Weyl points due to losing mirror planes
- ❖ Further Weyl properties studies need to take the LSB into account

Magnetic Phase Switching in Quantum Spin Liquid Pyrochlore $Yb_2(Ti_{1-x}Sn_x)_2O_7$



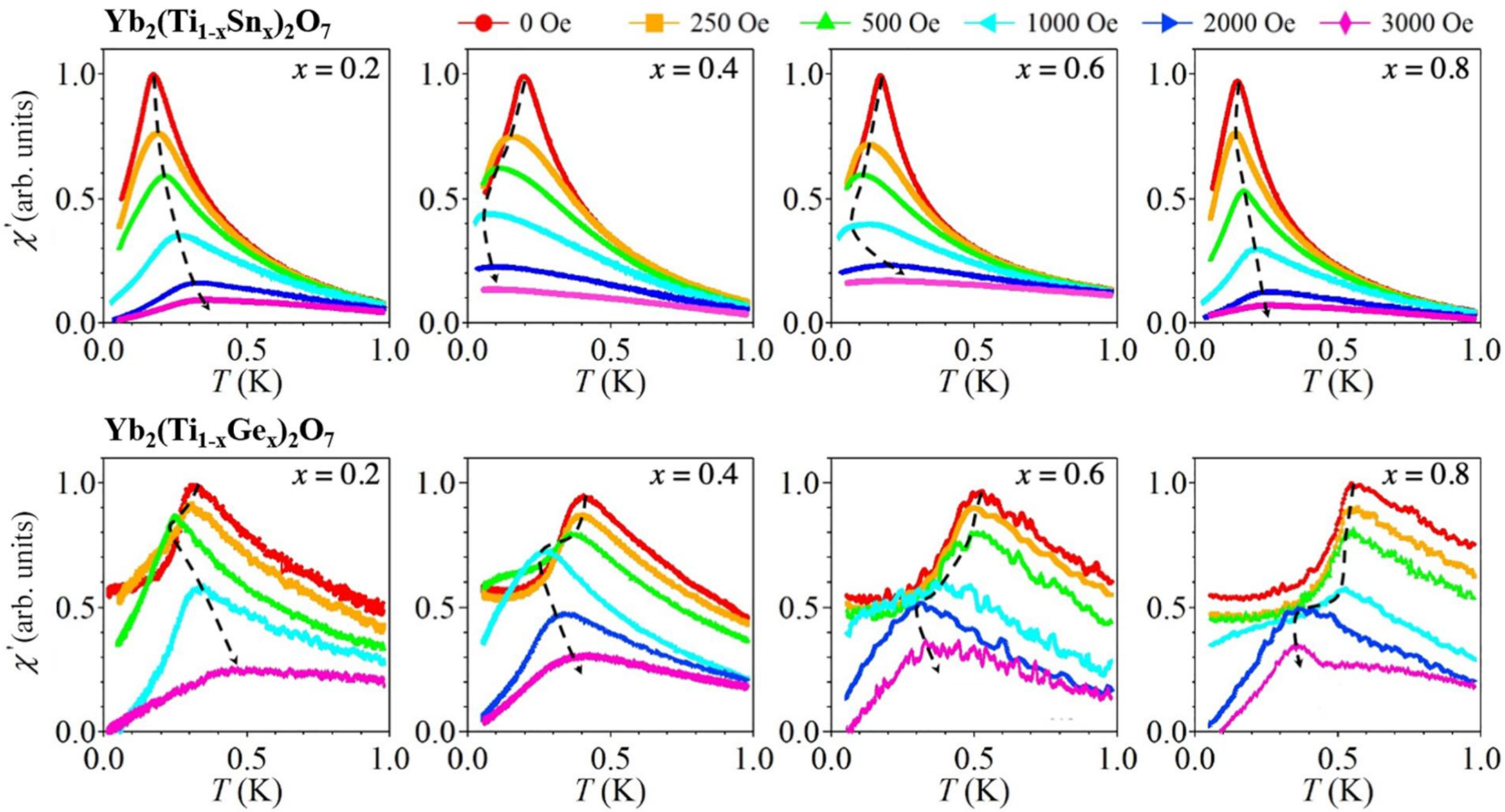
- Error-proof quantum computation
- HT superconductivity



In 2020, A. Scheie, et al. showed the continuum excitation as originated from FM+AFM competition rather than QSL

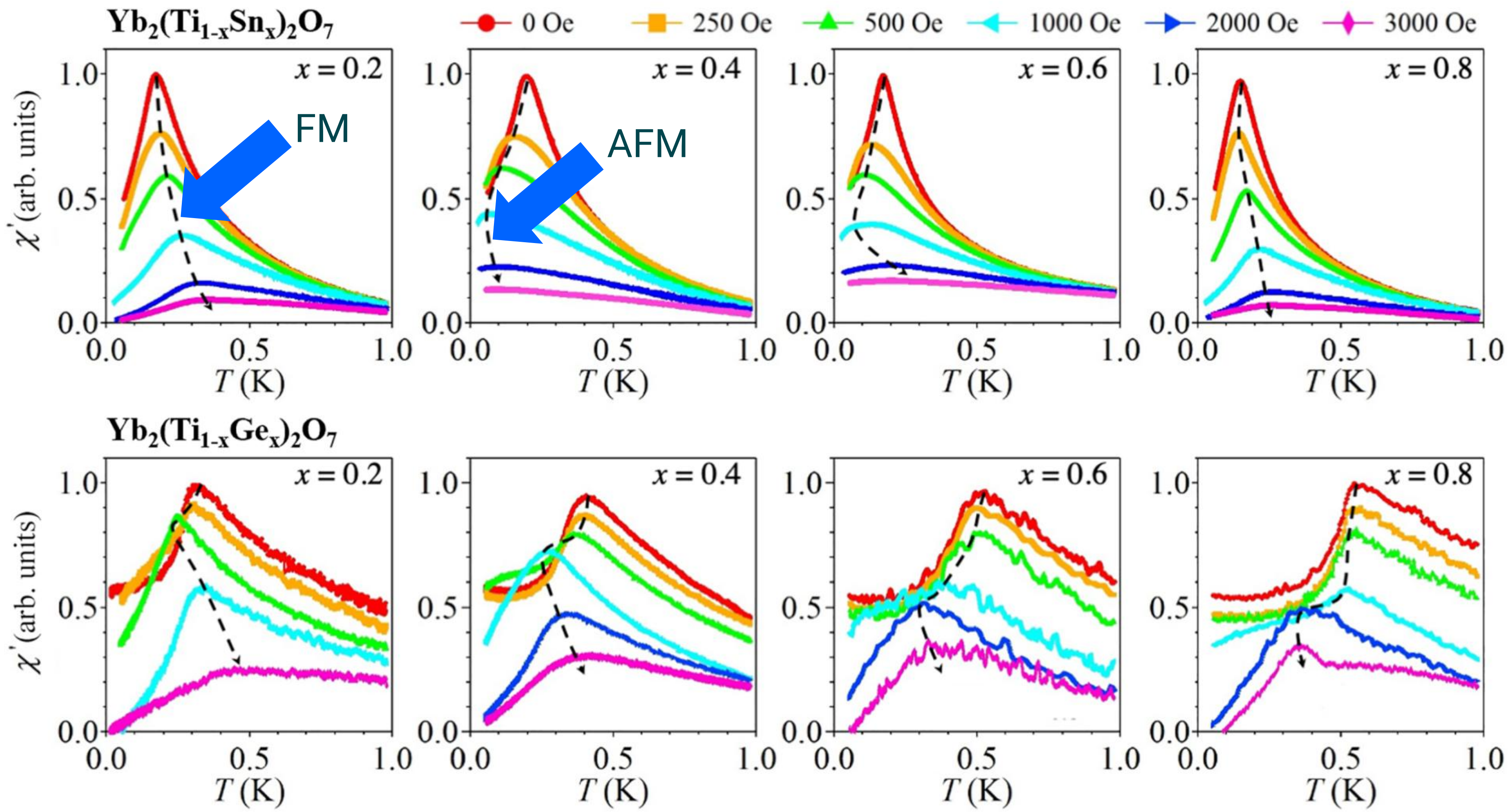
Y. P. Zhang, *et al. Phys. Rev. B*. 109, 144407 (2024)
A. Scheie, *et al*. PNAS. 117 (44) 27245-27254, 2020.

AC Susceptibility



Z. L. Dun, *et al.* Phys. Rev. B 89, 064401, 2014

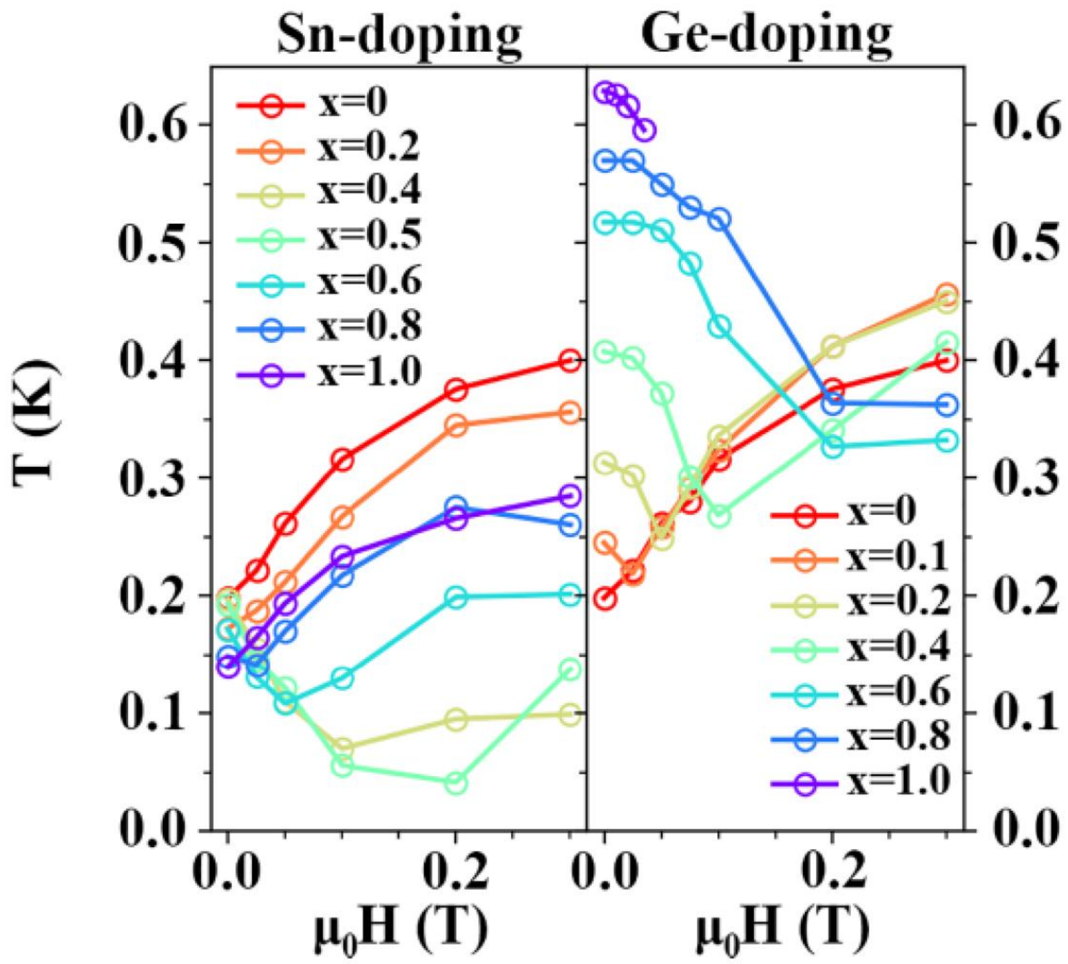
AC Susceptibility



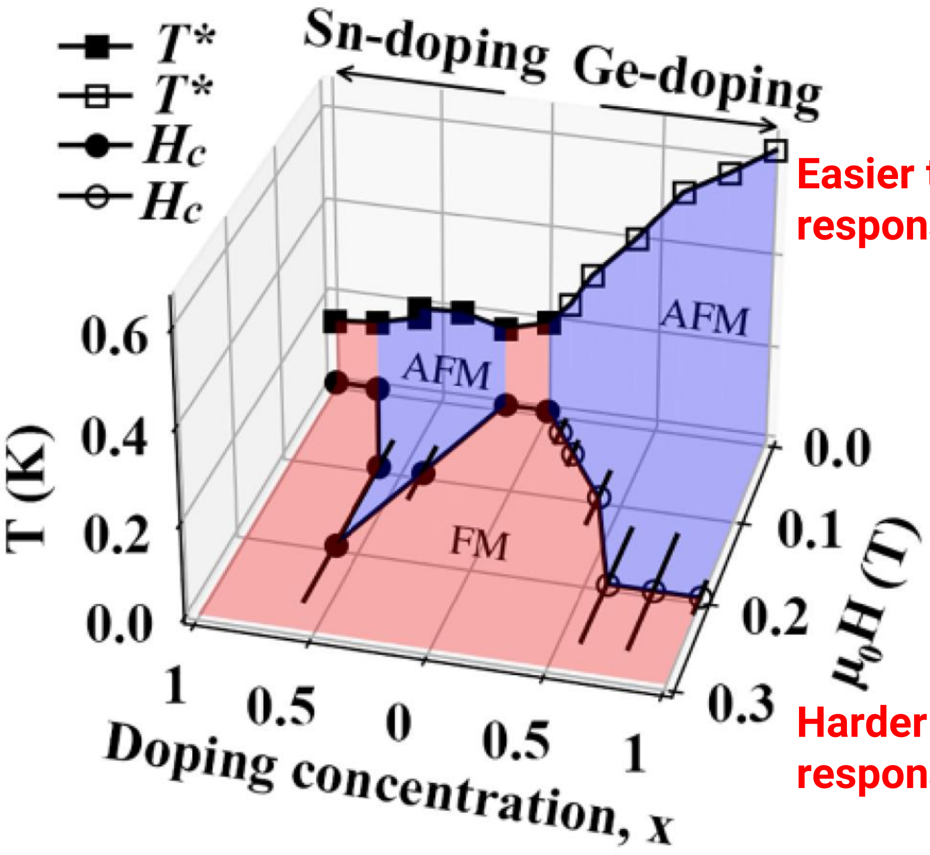
Z. L. Dun, *et al.* Phys. Rev. B 89, 064401, 2014

Magnetic Phase Switching

Transition temperature versus DC field



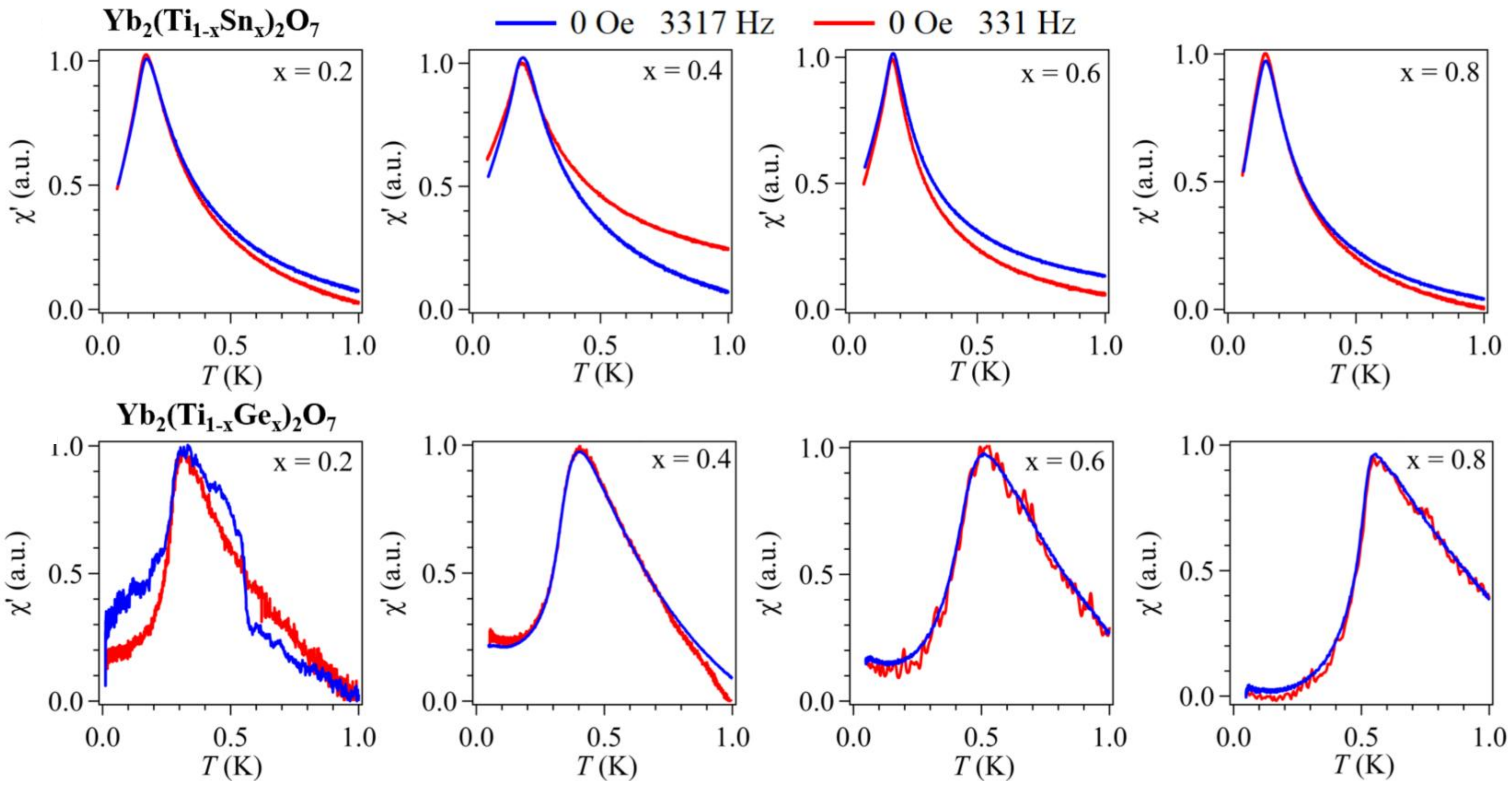
Magnetic phase diagram

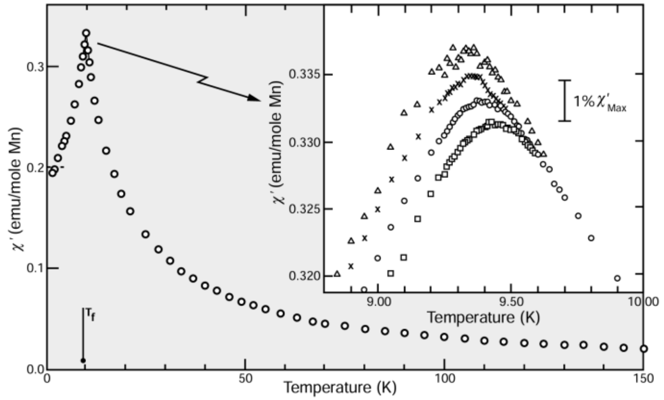


Easier to be less responsive

Harder to be responsive

Magnetic Phase Switching





Spin glass behavior in CuMn

No spin glass behavior in our sample

Z. L. Dun, *et al.* Phys. Rev. B 89, 064401, 2014
D. Martien, Introduction to AC Susceptibility

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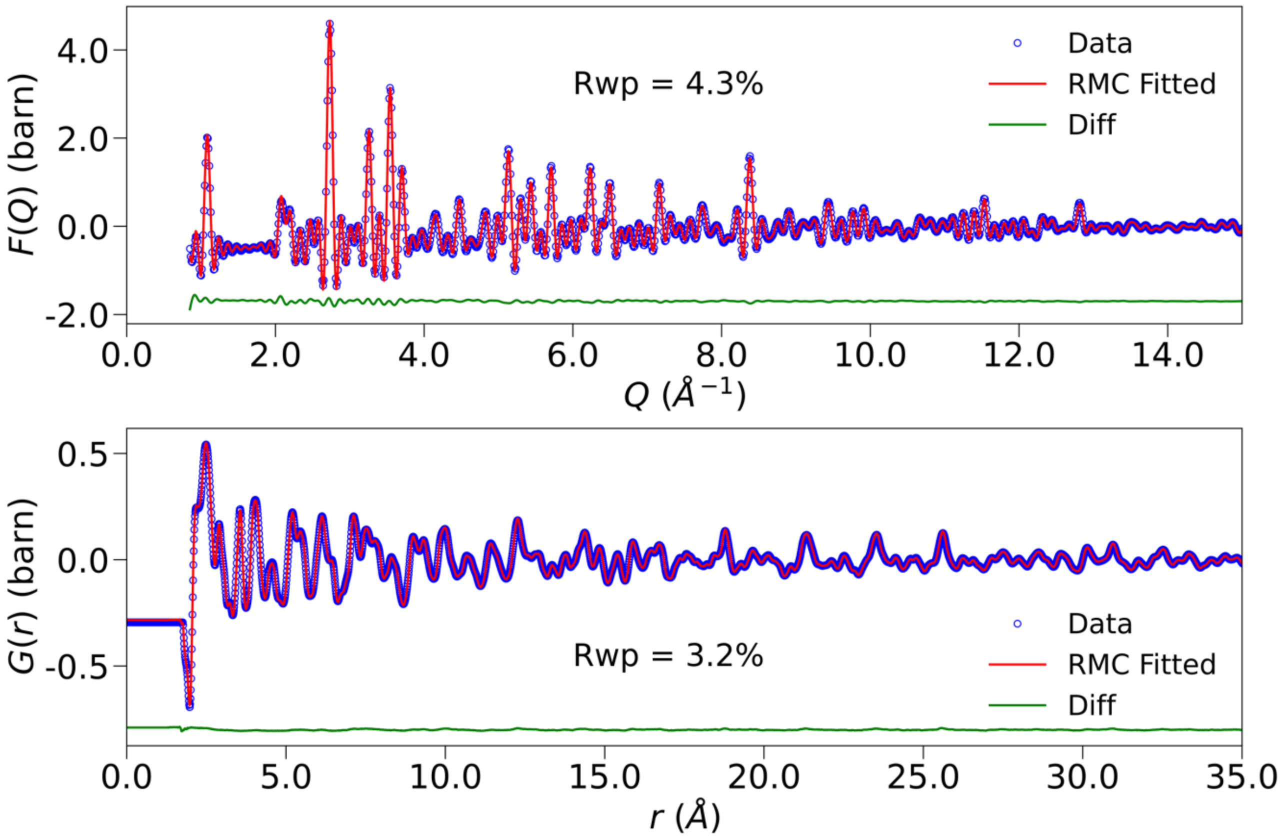
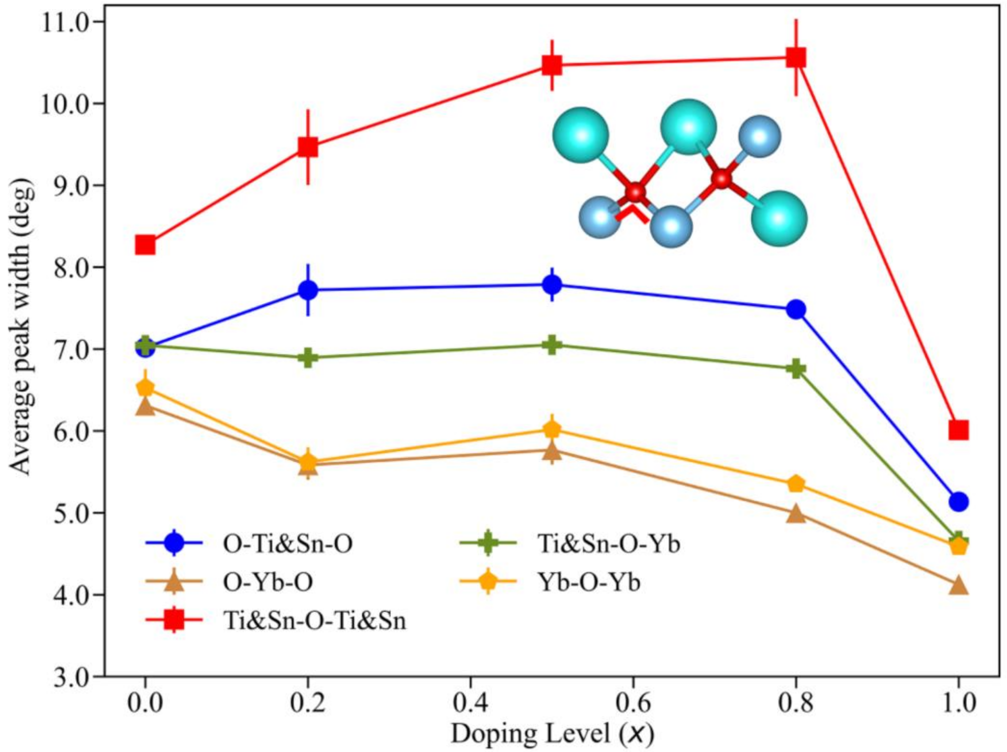
Magnetic Phase Switching in Quantum Spin Liquid Pyrochlore $Yb_2(Ti_{1-x}Sn_x)_2O_7$



Reasonable total scattering fitting in real & reciprocal space



Local variations in Ti-O-Ti chain coincides with the FM-AFM switching

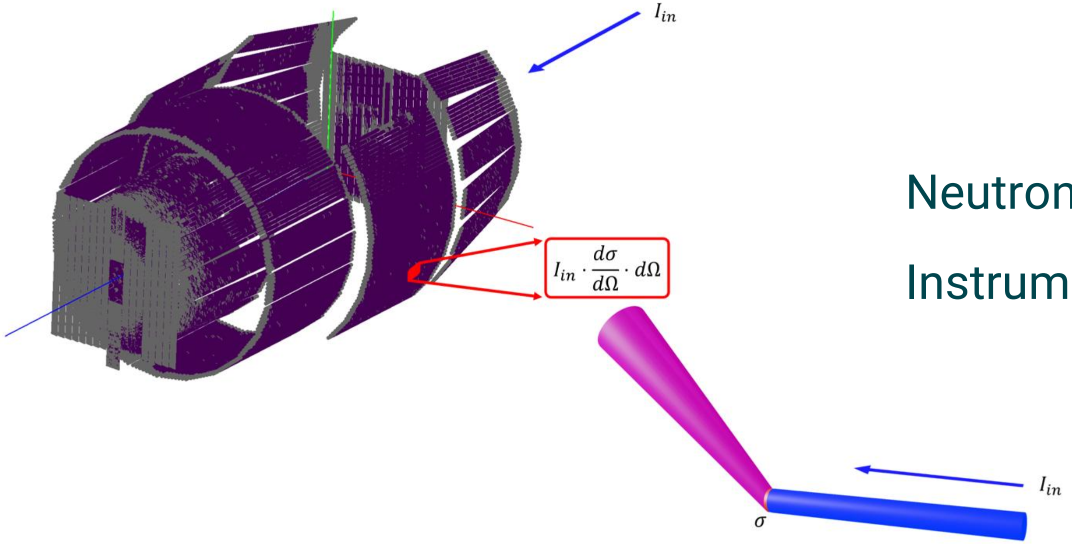
Y. P. Zhang, *et al. Phys. Rev. B.* 109, 144407 (2024)

Take-Away

- ❖ Chemical pressure induced FM-AFM switching
- ❖ Phenomenal link with strong local variation in Ti-O-Ti chain
- ❖ Local structure should be considered for future QSL candidate system studies

Local Magnetic Ordering with Total Scattering – with RMC (here, RMCProfile)

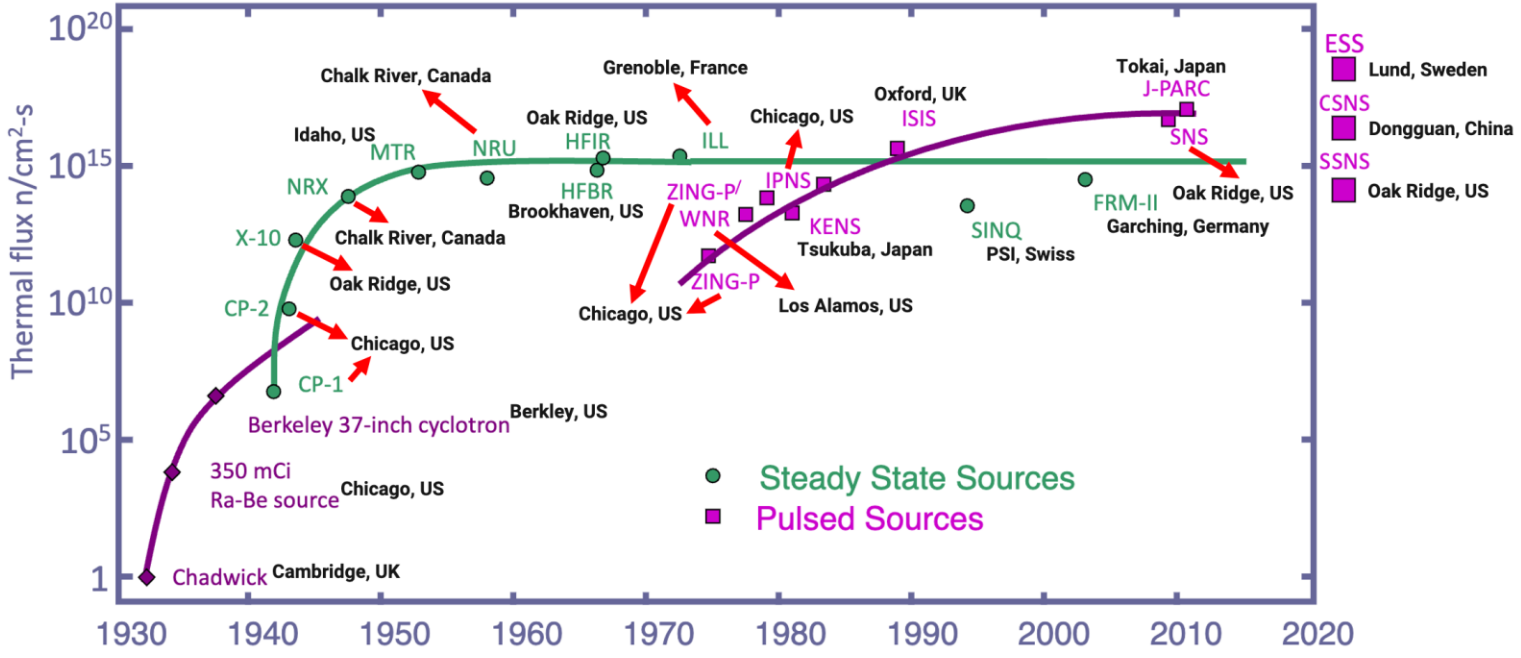
Local Magnetic Ordering with Total Scattering – with RMC (here, RMCProfile)



Neutron total scattering

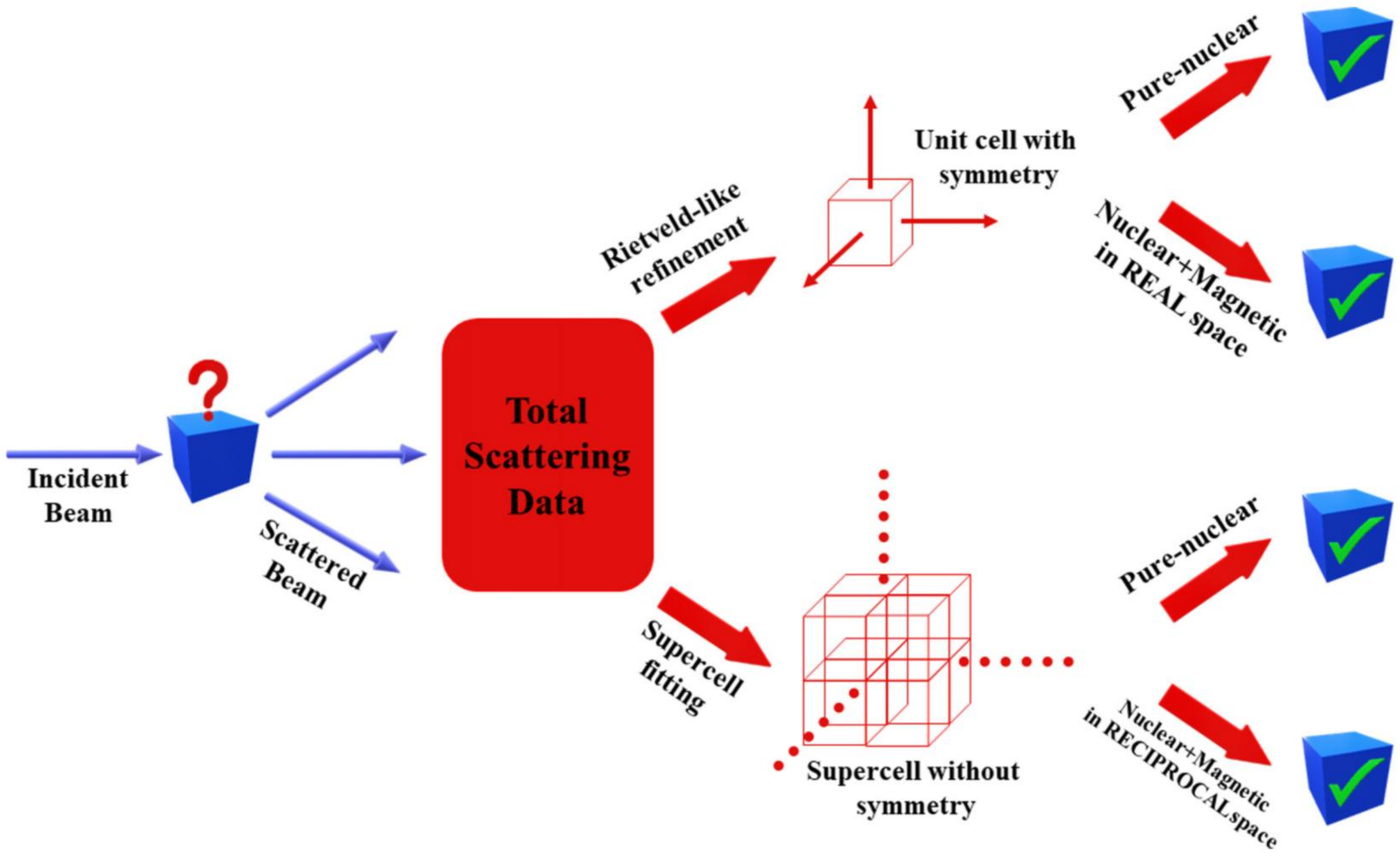
Instrument shown ⇒ NOMAD @ SNS, ORNL

Neutron Instruments History



https://powder.ornl.gov/general_aspects/nsources.html

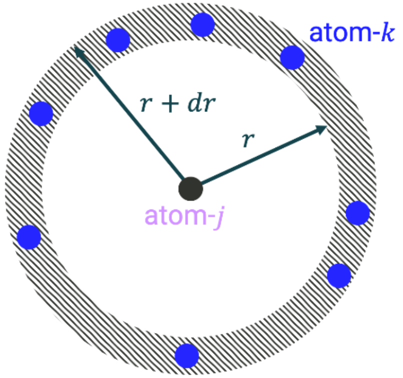
Local Magnetic Ordering with Total Scattering



Y. P. Zhang, *et al. Phys. Rev. B.* 100, 014419 (2019)

Real & Reciprocal Space Calculation

$$g_{jk}(r) = \frac{n_{jk}(r)}{4\pi r^2 dr \rho_k}$$



$$I(\vec{Q}) = \left| \sum_{0}^{N} \exp[i\vec{Q} \cdot \vec{r}_n] \right|^2$$

$\Downarrow$ Isotropic Averaging

$$I(Q) = \frac{1}{N_S} \sum_{j' \neq k'} b_{j'} b_{k'} \frac{\sin(Q r_{j'k'})}{Q r_{j'k'}} + \langle b^2 \rangle$$

Debye Approach

$$\Downarrow$$

$$I(Q) = \rho_0 \sum_{j \neq k} c_j c_k b_j b_k 4\pi \int_0^\infty r^2 [g_{jk}(r) - 1] \frac{\sin(Qr)}{Qr} dr + \langle b^2 \rangle$$

$$\Downarrow$$

$$G(r) = \sum_{j,k} c_j c_k b_j b_k [g_{jk}(r) - 1]$$

$$\Downarrow$$

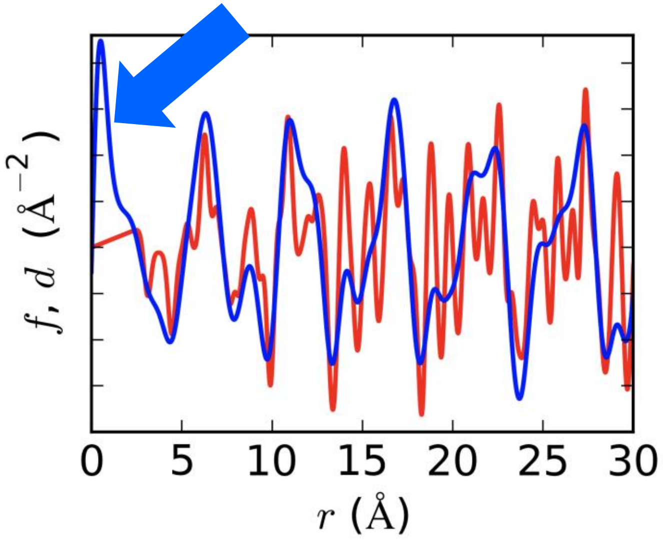
$$I(Q) = F(Q) + \langle b^2 \rangle = \rho_0 \int_0^\infty 4\pi r^2 G(r) \frac{\sin(Qr)}{Qr} dr + \langle b^2 \rangle$$

Real-space Approach

The Self-Scattering Term

Reciprocal space [1]:

$$S_{mag}(Q) = \boxed{\frac{2}{3}c_M[\frac{e^2\gamma}{2m_ec^2}gJf(Q)]^2} + 4\pi\rho c_M[\frac{e^2\gamma}{2m_ec^2}f(Q)]^2 \times \int r^2\{A(r)\frac{\sin Qr}{Qr} + B(r)[\frac{\sin Qr}{(Qr)^3} - \frac{\cos Qr}{(Qr)^2}]\}dr$$



Real space [2, 3]:

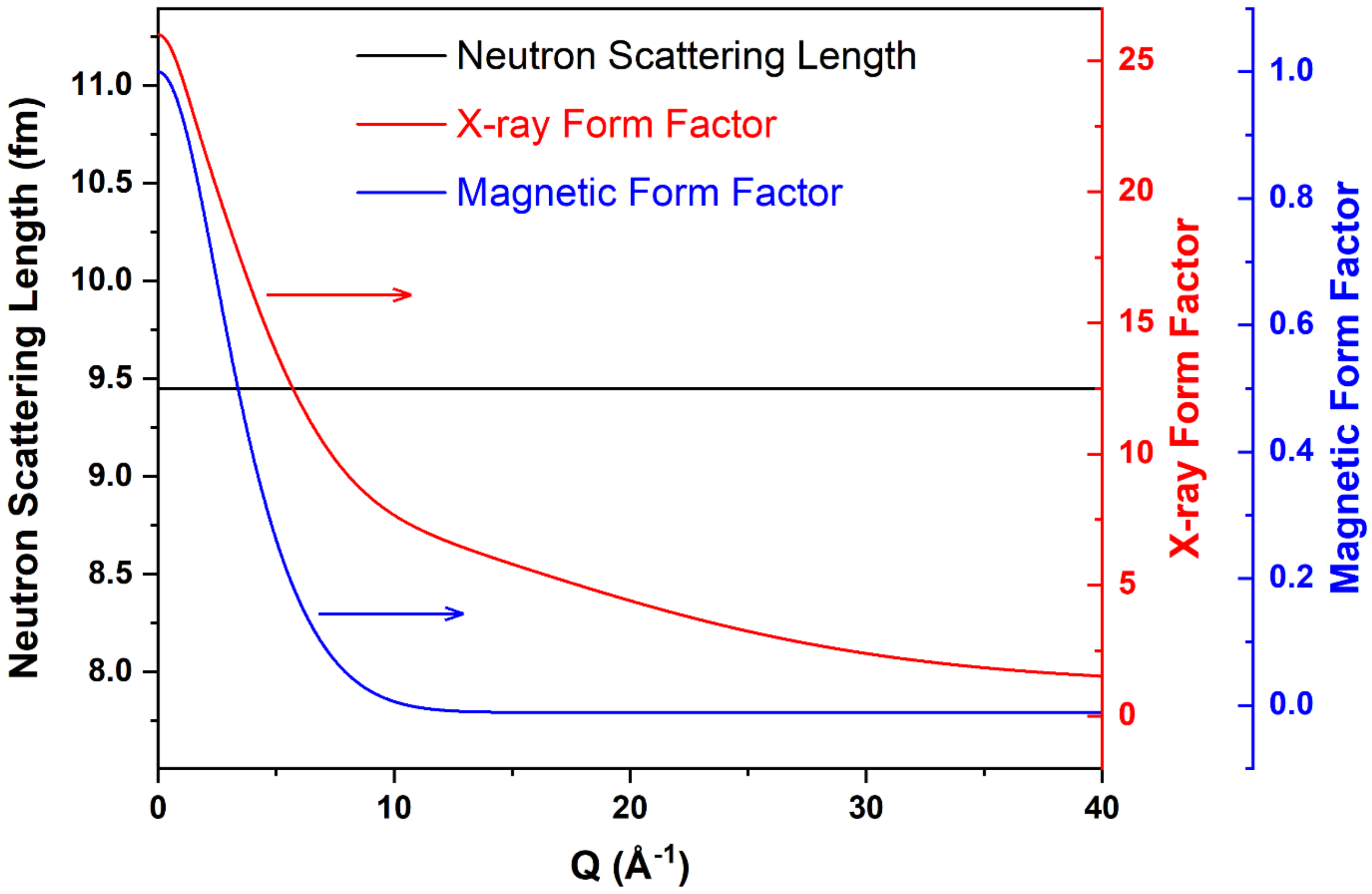
$$d(r) = \boxed{C_2 \times \frac{dS}{dr}} + C_1 \times f(r) \otimes S(r)$$

Spin self-scattering term, which appears as a peak at very low r.

$$= C_2 \times \frac{dS}{dr} + C_1 \times \frac{1}{N}\frac{3}{2S(S+1)}\sum_{i\neq j}\{\frac{A_{ij}}{r} + B_{ij}\frac{r}{r_{ij}^3}[1-\Theta(r-r_{ij})]\} \otimes \{\mathrm{F}[f_m(Q)] \otimes \mathrm{F}[f_m(Q)]\}$$

[1] Refer to RMCProfile manual section-2.10.2.
[2] B. A. Frandsen, et al., Acta Cryst. (2014). **A70**, 3-11.
[3] B. A. Frandsen, et al., Phys. Rev. Lett. (2016), **116**, 197204.
[4] B. A. Frandsen, et al. Acta Cryst. (2015). A71, 325-334

Dealing with the Form Factor



$$\mathcal{F}\,[\text{nucleus, electron cloud, unpaired electrons}]$$

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Dealing with the Form Factor

$$f_{ij}(Q) = f_i(Q) f_j(Q) \Big/ \Big[\sum_{i=1}^{n} c_i f_i(Q) \Big]^2$$

$$F^X(Q) = \Big[\frac{1}{N} \frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} - \sum_{i=1}^{n} c_i f_i(Q)^2 \Big] \Big/ \Big[\sum_{i=1}^{n} c_i f_i(Q) \Big]^2 \quad \Leftrightarrow \quad F^X(Q) = \rho_0 \sum_{i=1}^{n} c_i c_j f_{ij}(Q) \underline{\int_0^\infty 4\pi r^2 [g_{ij}(r) - 1] \frac{\sin Qr}{Qr} \, \mathrm{d}r}$$

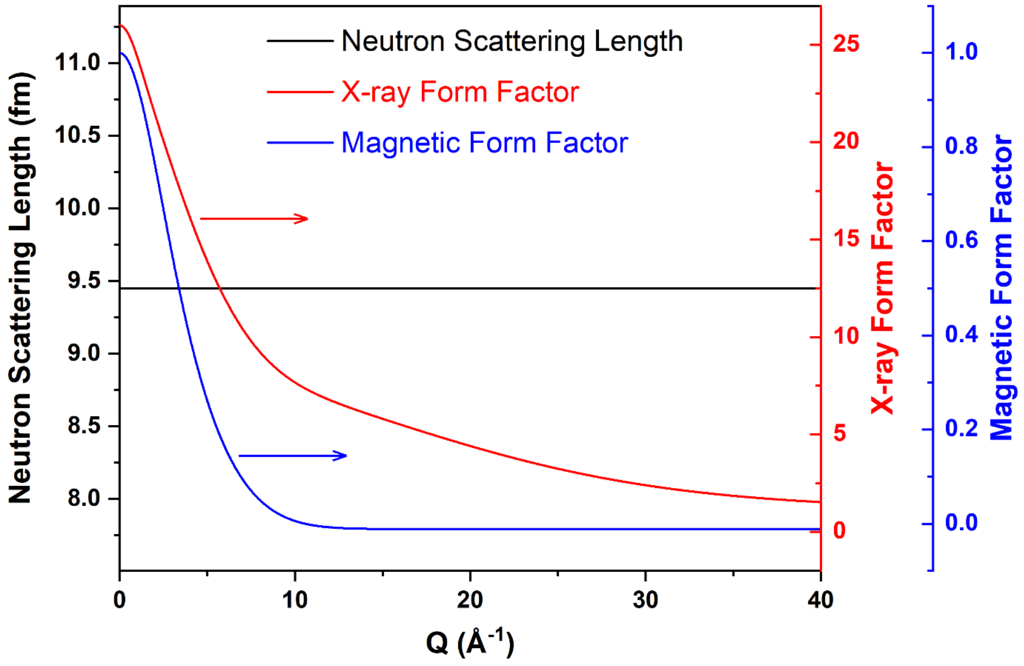
$\searrow \qquad \swarrow$

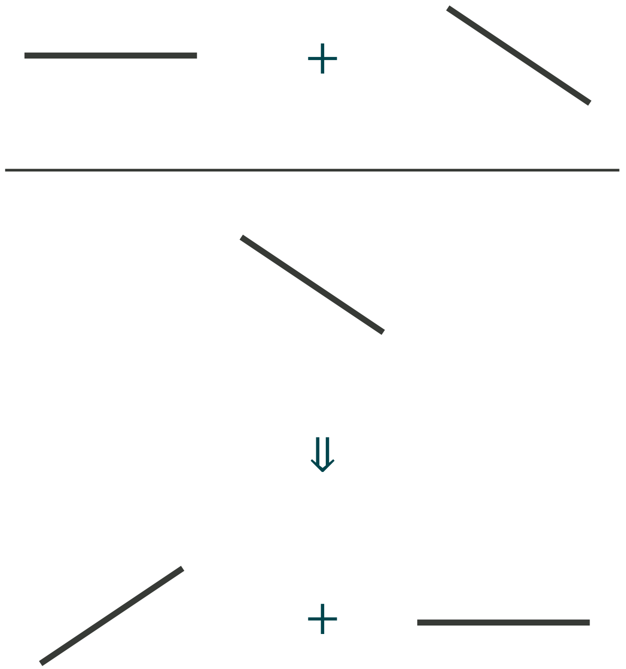
$$G^X(r) = \frac{1}{(2\pi)^3 \rho_0} \int_0^\infty 4\pi Q^2 F^X(Q) \frac{\sin Qr}{Qr} \, \mathrm{d}Q$$

This is how we deal with X-ray calculation

OAK RIDGE National Laboratory D. Keen, *J. Appl. Cryst.* (2001). 34, 172-177.

Dealing with the Form Factor





Meaning, if we cannot separate nuc and mag contribution, it does not make sense to do the normalization over the magnetic form factor

- Option-1: Stay in Q-space ⇒ ***currently what we do***
- Option-2: $\mathcal{F}\left[\left[F_{nuc}(Q) + S_{mag}(Q) - S_{mag}^{self}(Q)\right]/\langle b \rangle^2\right]$ ⇒ ***planned***

A Little Trick at This Moment – Combine mPDF with RMCProfile

Nuc & Mag co-refinement

$\Downarrow$

Subtract mag part

$\Downarrow$

Clean nuc PDF

Nuc + Mag $F(Q)$

$\Searrow$

$\Swarrow$

Nuc-only model

Nuc + Mag model

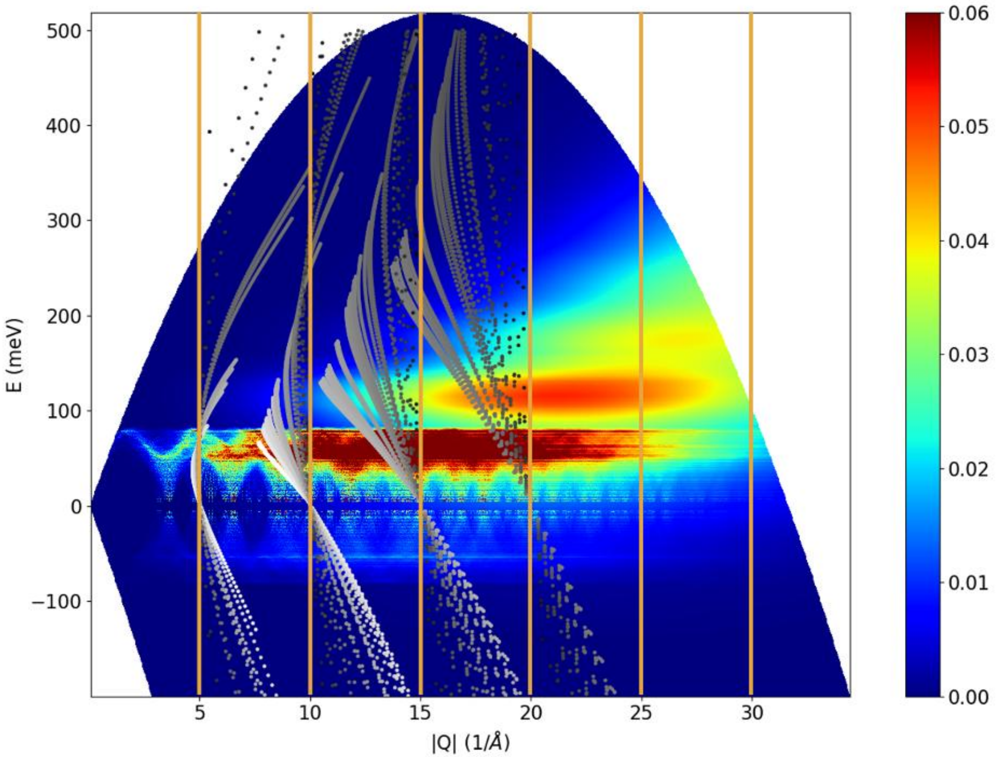
$\Searrow$

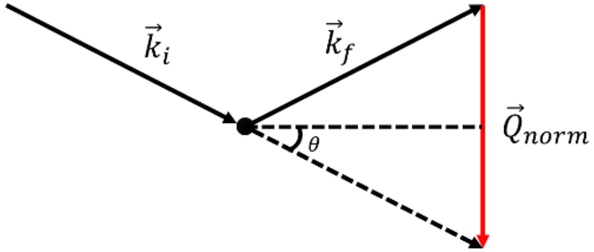
$\Swarrow$

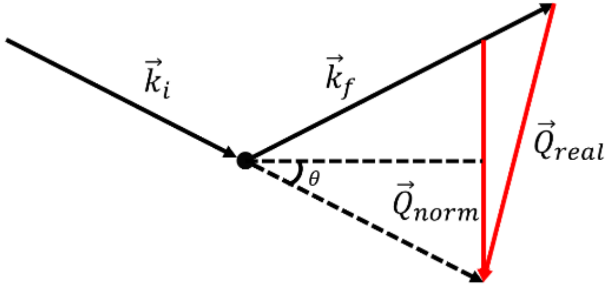
RMCProfile

Inelastic Scattering Effect

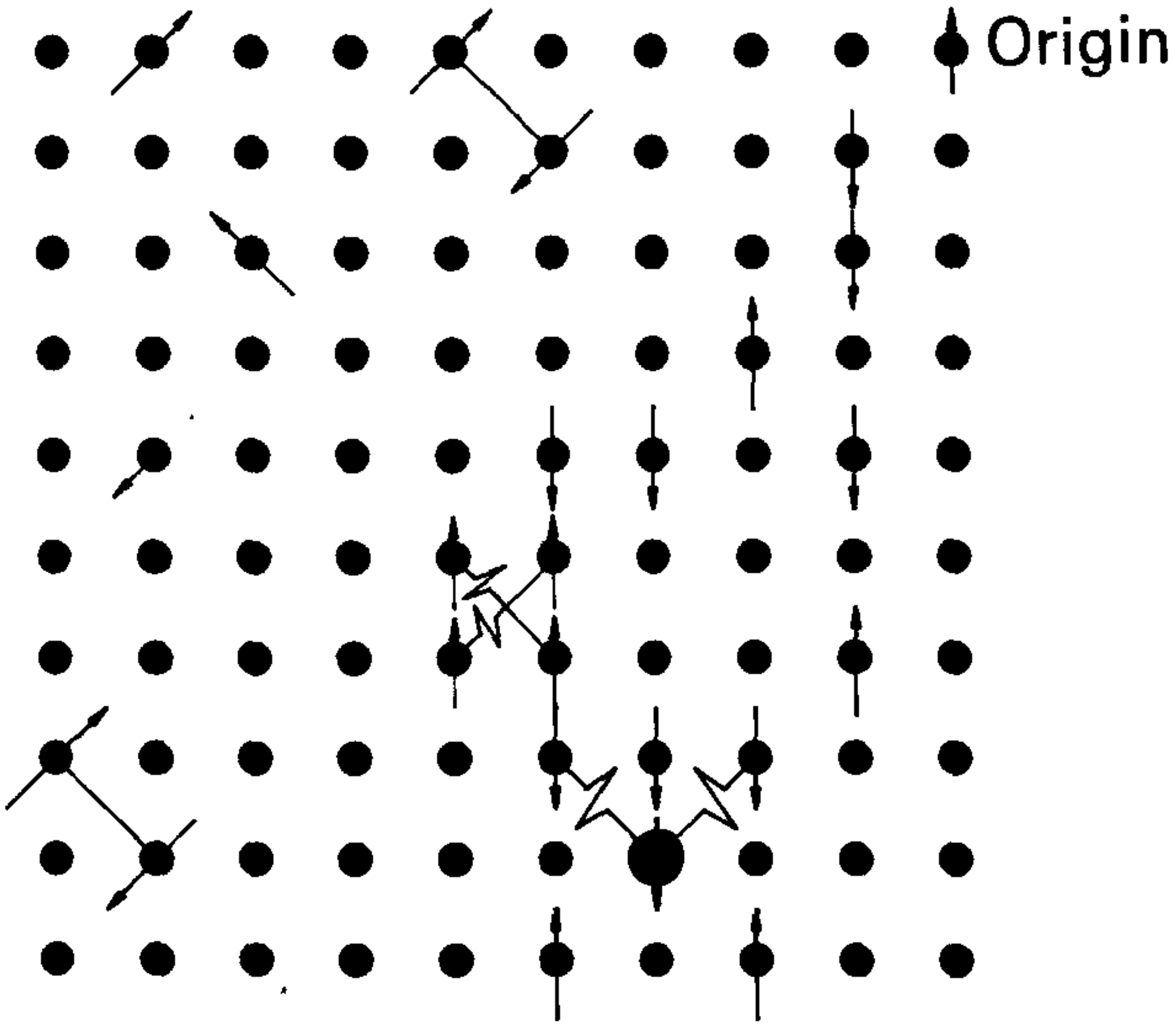
- Mainly in the low-Q region
- Mainly with light elements





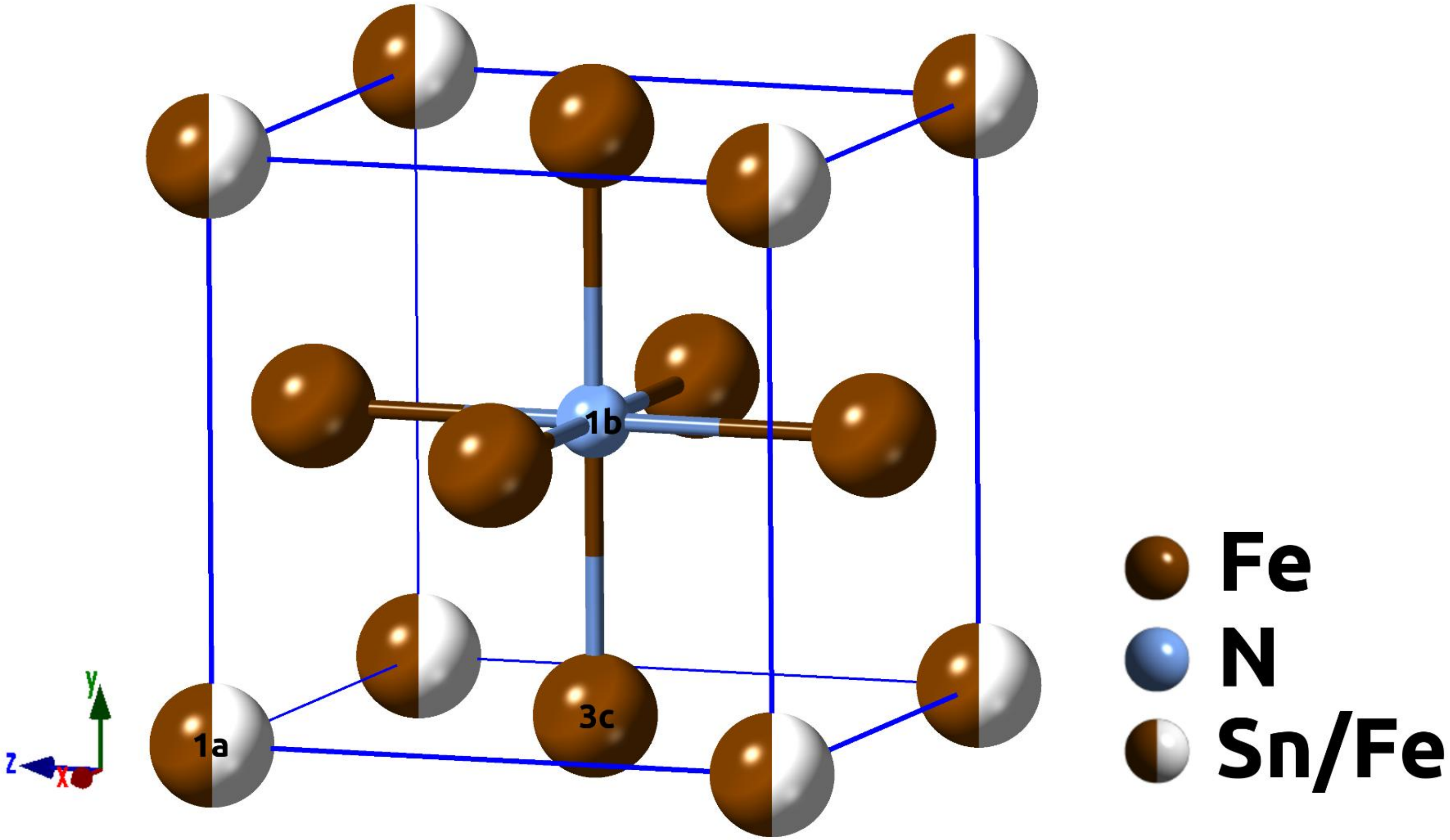


Local Magnetic Clustering in Spin Glass $Sn_xFe_{4-x}N$



J. A. Mydosh, *Spin glass: an experimental introduction,*
Taylor & Francis, London · Washington, DC, 1993.

$Sn_xFe_{4-x}N$ – The Underlying Structure



Substitution happens on the 1a Wyckoff site only

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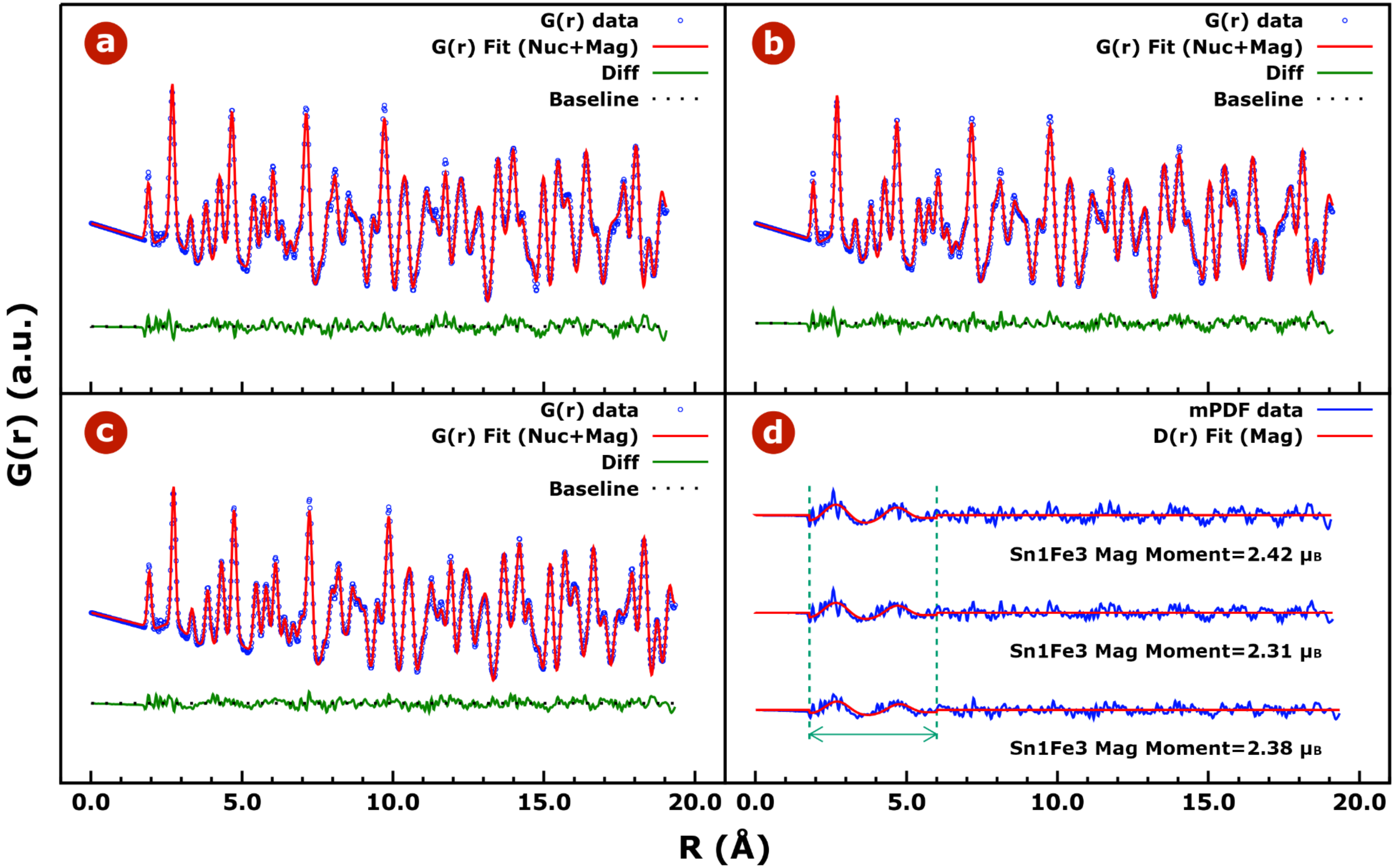
[1] T. Scholz, *et al. AIP Adv.*, **6**, 055107, 2016.
[2] Y. P. Zhang, *et al*. 100, 014419 (2019).

mPDF Fitting in Real Space

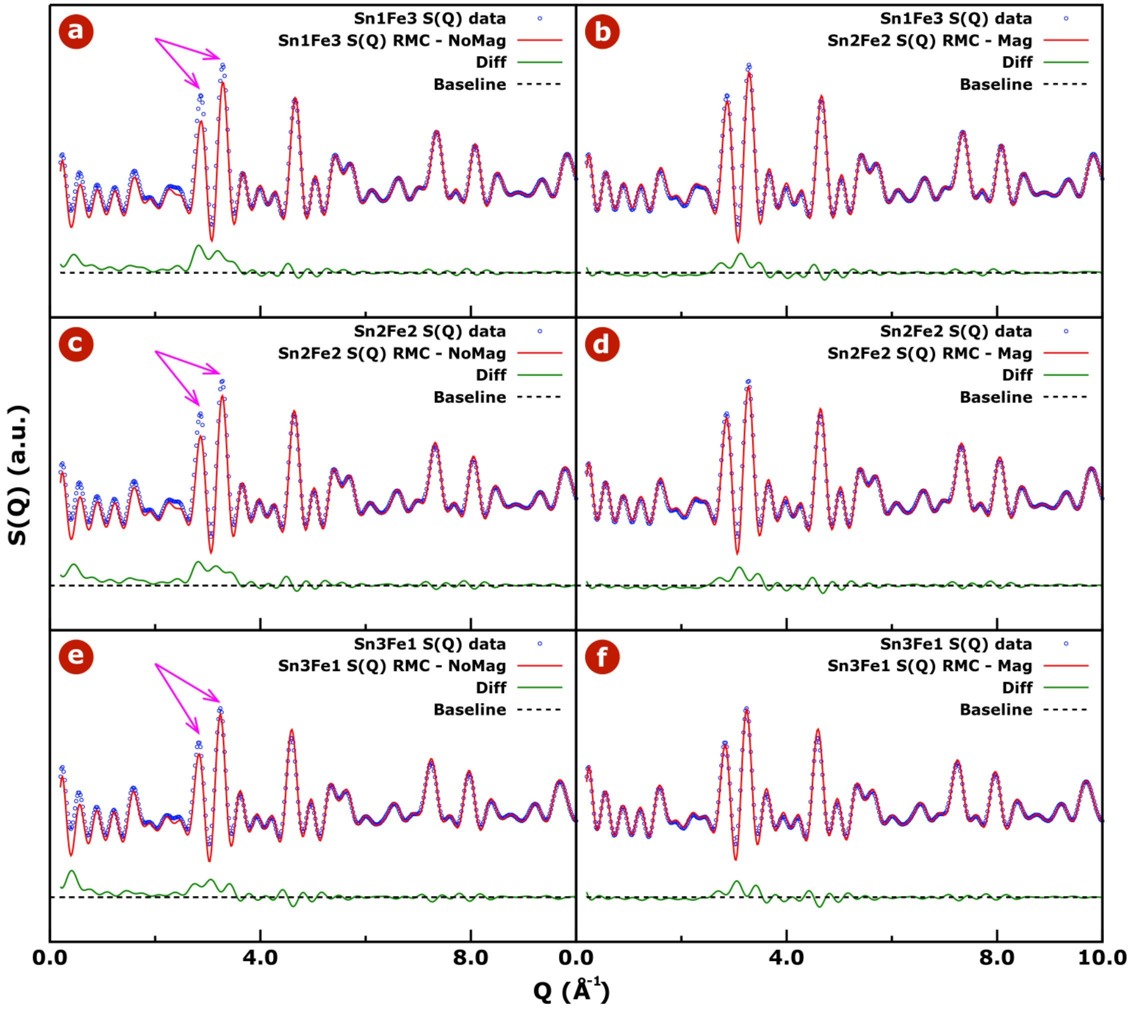


Nucleus & Magnetic co-refinement with diffpy.mpdf

$$S_{fit} = \sqrt{\frac{gB\langle b\rangle^2}{An_s}}S_{norm}$$

[1] P. Juhas, et al., *Acta Cryst.*, **A71**, 562-568, 2015.
[2] B. Frandsen, et al., *Acta Cryst.*, **A70**, 3-11, 2014.

mPDF Fitting in Real Space



RMCProfile fitting in *Q*-space with (left) and without (right) magnetic contribution.

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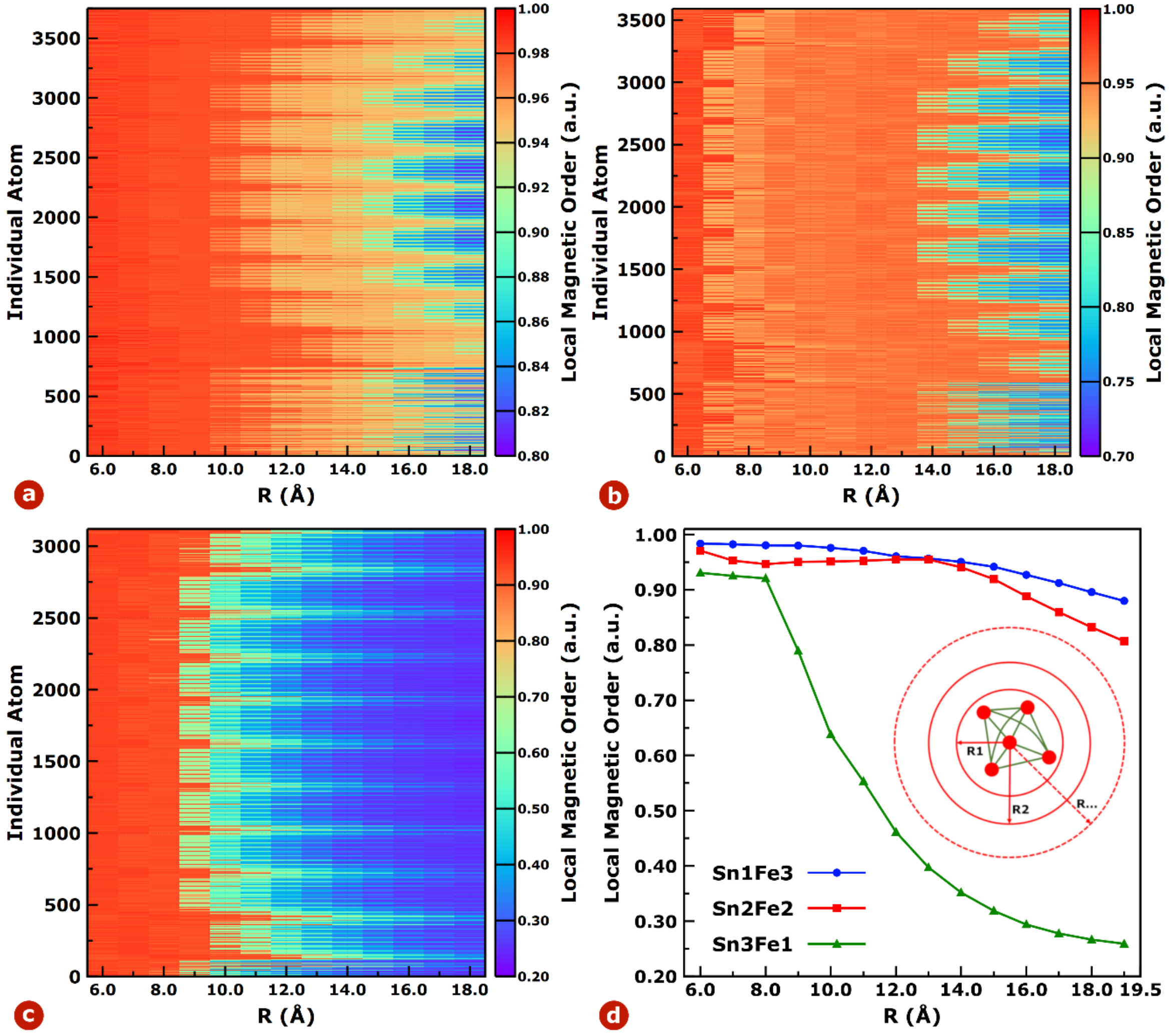
Local Correlation in Spin Glass

$g = \langle S_i S_j \rangle_{i \neq j}$ - Local magnetic order



Local correlation analysis.

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Masaaki Matsuda
Matt Tucker
Michael A. McGuire
Qiang Zhang
Richard Dronskowski
Satoshi Okamoto
Tanja Scholz
Vasile Ovidiu Garlea
Yongqiang Cheng
Zhiling Dun


Thank you!
Questions?