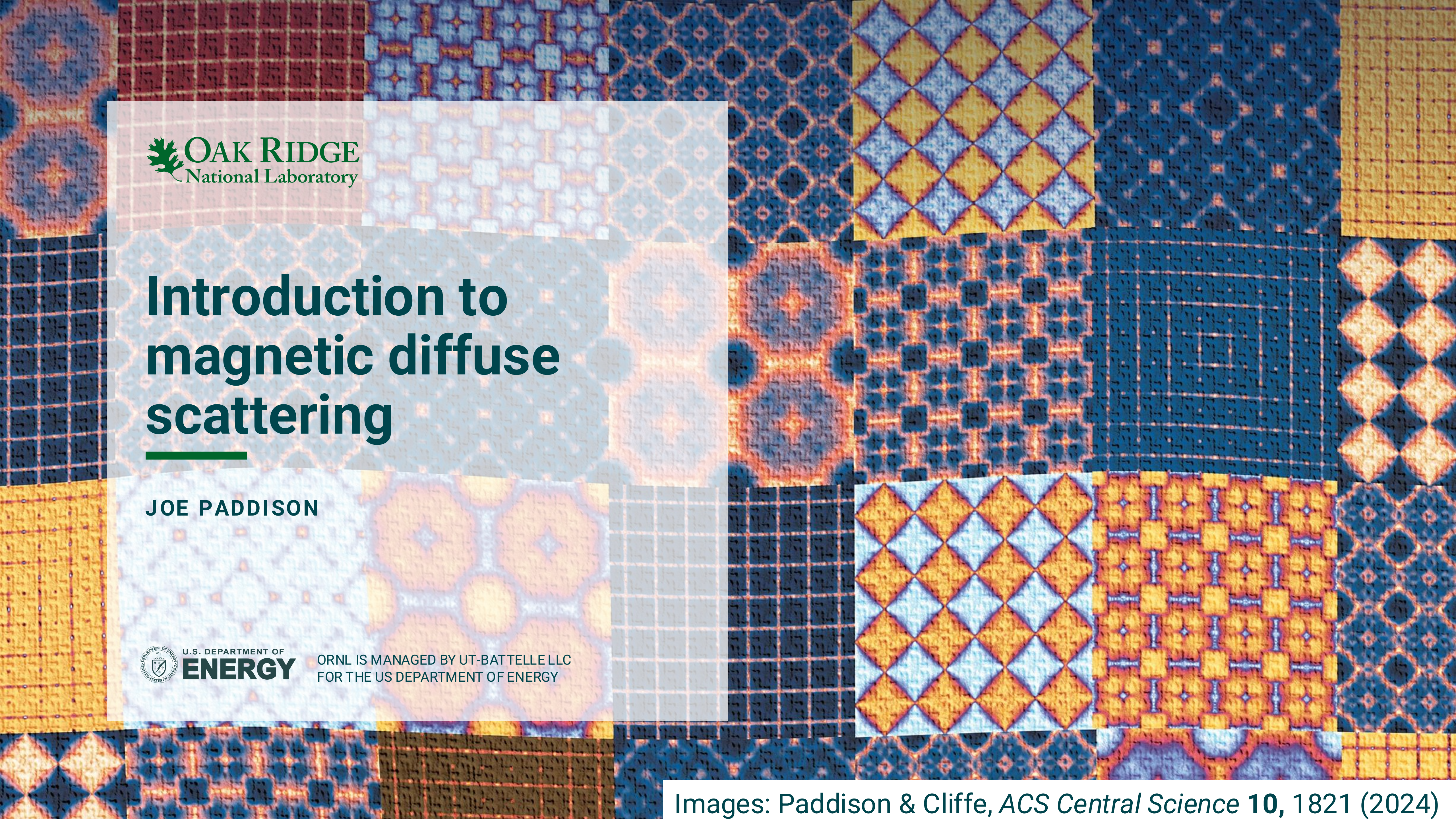

Oak Ridge National Laboratory

Introduction to magnetic diffuse scattering

JOE PADDISON

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ORNL IS MANAGED BY UT-BATTELLE LLC
FOR THE US DEPARTMENT OF ENERGY

Images: Paddison & Cliffe, *ACS Central Science* **10,** 1821 (2024)

What is magnetic diffuse scattering?

- Broad features beneath and between the sharp (Bragg) peaks
- e.g. magnetic scattering from MnO

Clifford Shull

Magnetic Bragg scattering
→ *long-range magnetic order*

Magnetic diffuse scattering
→ *short-range magnetic order*

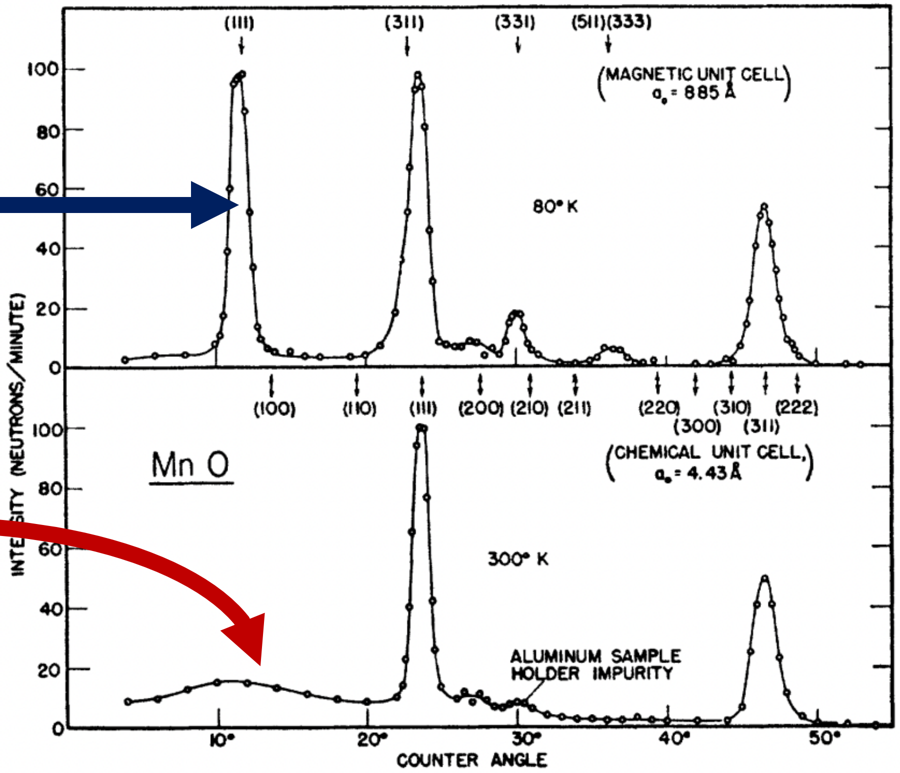


FIG. 1. Neutron diffraction patterns for MnO at room temperature and at 80°K.

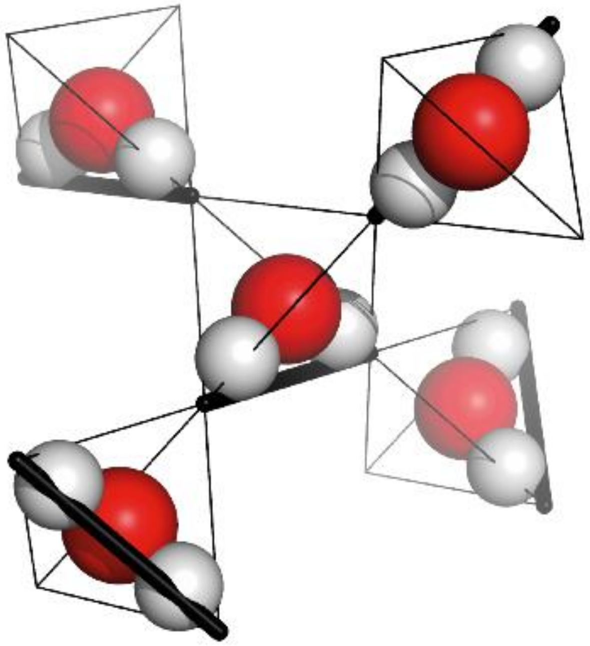
$T < T_N$

$T > T_N$

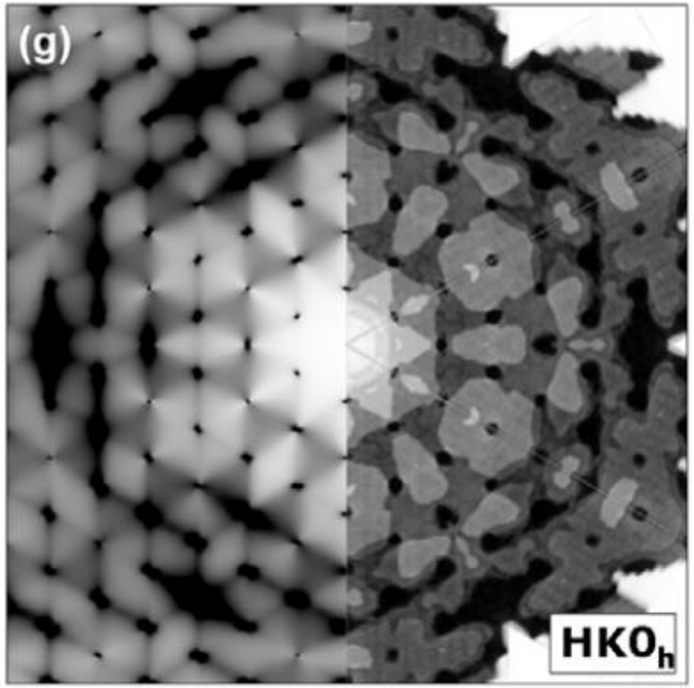
Shull and Smart, *Phys Rev* **76**, 1256 (1949)

What does diffuse scattering measure?

- Deviations from long-range order (either **chemical** or magnetic)
- Correlated disorder → structured diffuse scattering
  - e.g. “ice rules” in water ice



Water ice
*Real space, **r***

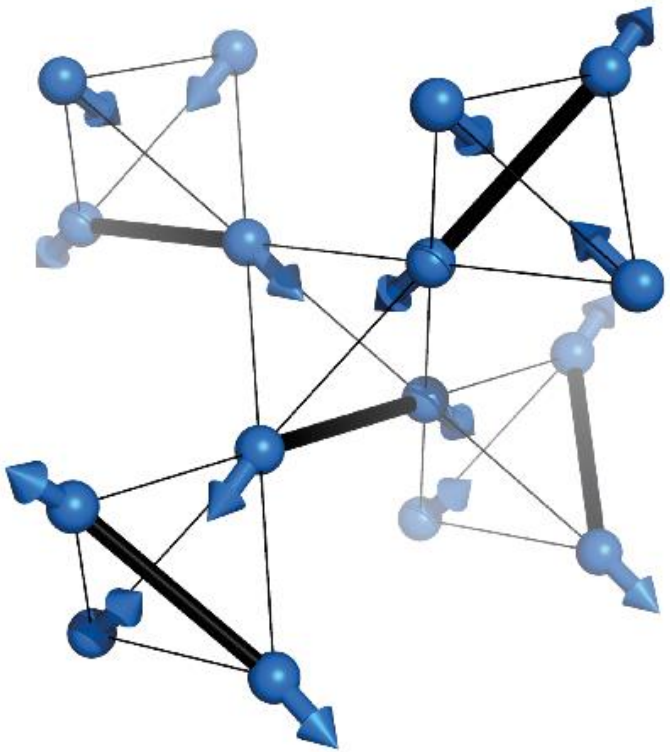


Model **Neutron data***
*Reciprocal space, **Q***

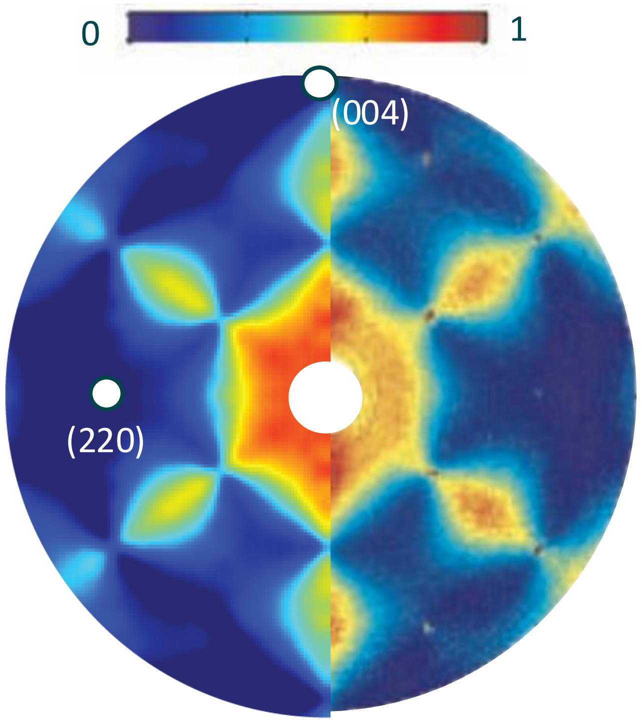
*Wehinger et al., *JPCM* **26** 265401 (2014)

What does diffuse scattering measure?

- Deviations from long-range order (either chemical or **magnetic**)
- Correlated disorder → structured diffuse scattering
  - e.g. “ice rules” in spin ice



Spin ice
*Real space, **r***



Model **Neutron data***
*Reciprocal space, **Q***

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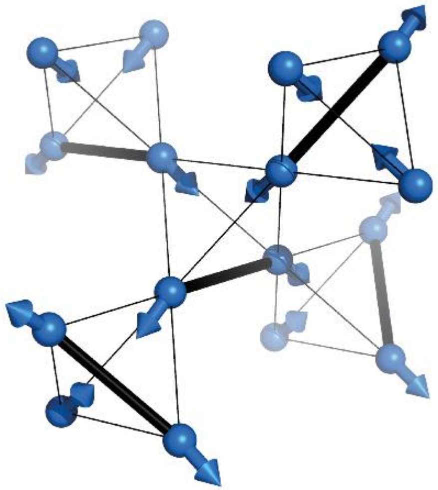
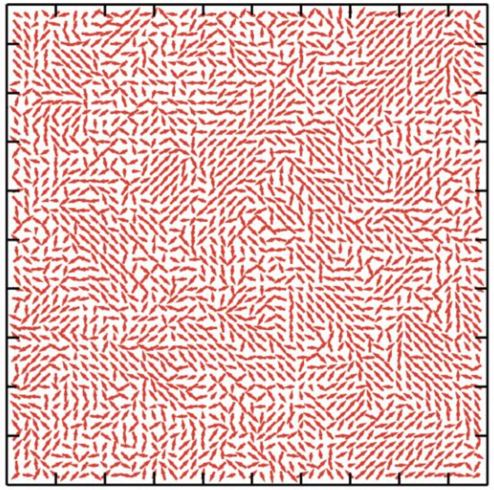
*Fennell et al., *Science* **326**, 415 (2009)

Why do we study diffuse scattering?

- *Exotic (possibly quantum) magnetic states*
  - May not show conventional magnetic order, so best understood *via* diffuse and inelastic scattering
  - e.g. quantum spin liquids, spin ice, "hidden" order
- *Functional materials*
  - Behaviour often driven by local structure distortions
  - e.g. colossal magnetoresistance manganites
- *Insight into interactions*
  - Diffuse scattering provides information about a material's Hamiltonian



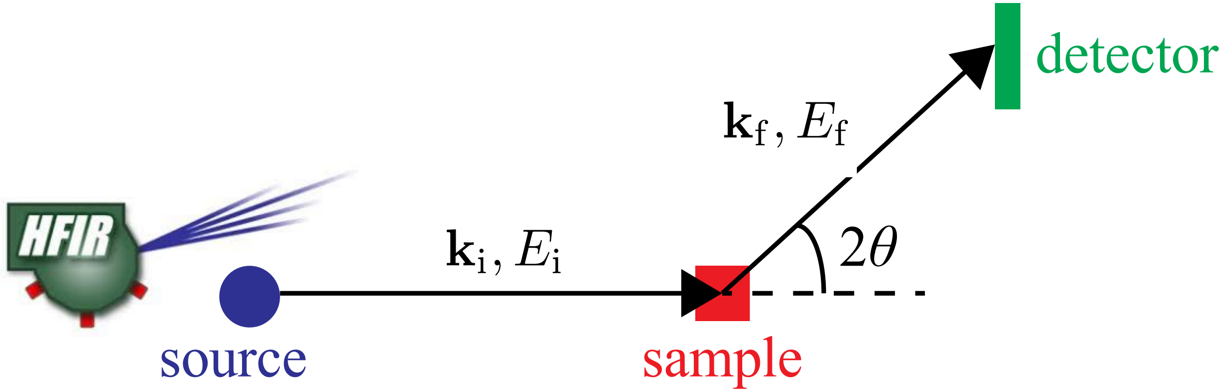
Spin orientations in spin ice



*Dipole orientations in a relaxor ferroelectric**

*Pasciak et al., *PRB* **85**, 224109 (2012)

How do we measure diffuse scattering using neutrons?



Energy transfer $E = E_\mathrm{i} - E_\mathrm{f}$

Wavevector transfer $\mathbf{Q} = \mathbf{k}_\mathrm{i} - \mathbf{k}_\mathrm{f}$

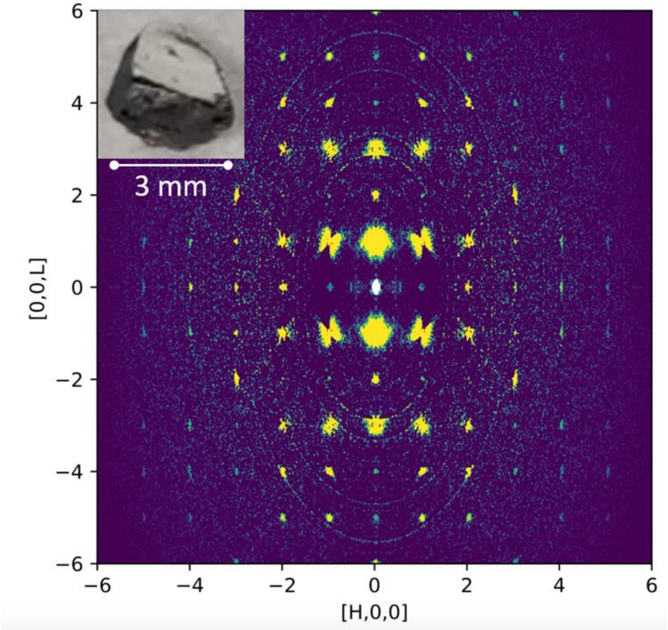
$$Q = |\mathbf{Q}| = \frac{4\pi \sin\theta}{\lambda}$$

- **Quantity of interest** is *E*-integrated intensity $I(\mathbf{Q}) = \int_{-\infty}^{\infty} I(\mathbf{Q}, E)\, \mathrm{d}E$
  - Approximated by diffraction measurements if $E \ll E_\mathrm{f}$
- **For magnetic crystals:** Measure large volume of **Q** (e.g. SXD @ ISIS, Corelli @ SNS)
- **For magnetic powders:** Maximise flux at small *Q* (e.g. D20 @ ILL, POWDER @ HFIR)
- **Polarisation analysis** can effectively isolate magnetic signal
- Otherwise, important to measure and subtract background signal

How can we visualise diffuse scattering data?

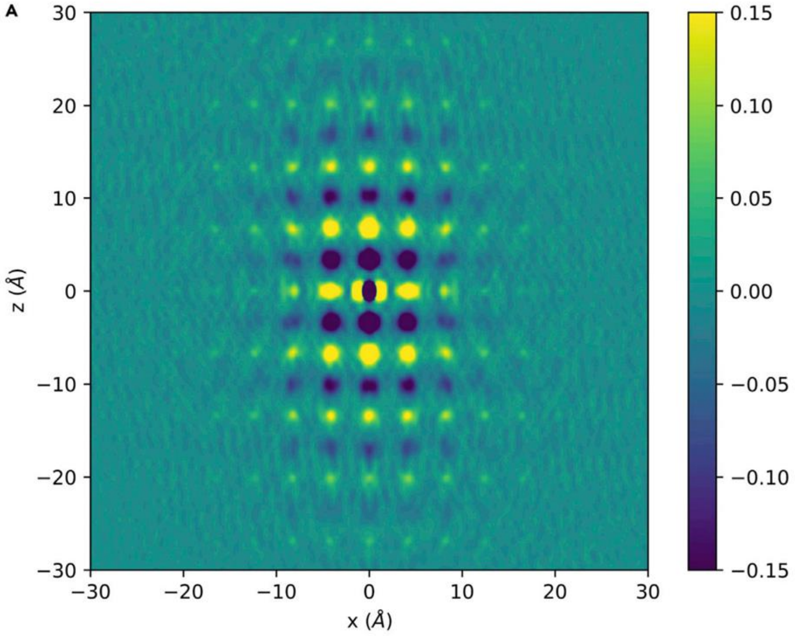
- Fourier transform of magnetic single-crystal data yields *3D magnetic PDF* (spin-pair correlation function)
  - Positive (negative) peaks indicate ferromagnetic (antiferromagnetic) correlations
  - e.g. magnetically-enhanced thermoelectric MnTe at $T > T_N$

Reciprocal space, ***Q***
Nuclear scattering subtracted



Fourier transform →

Real space, ***r***
Magnetic PDF

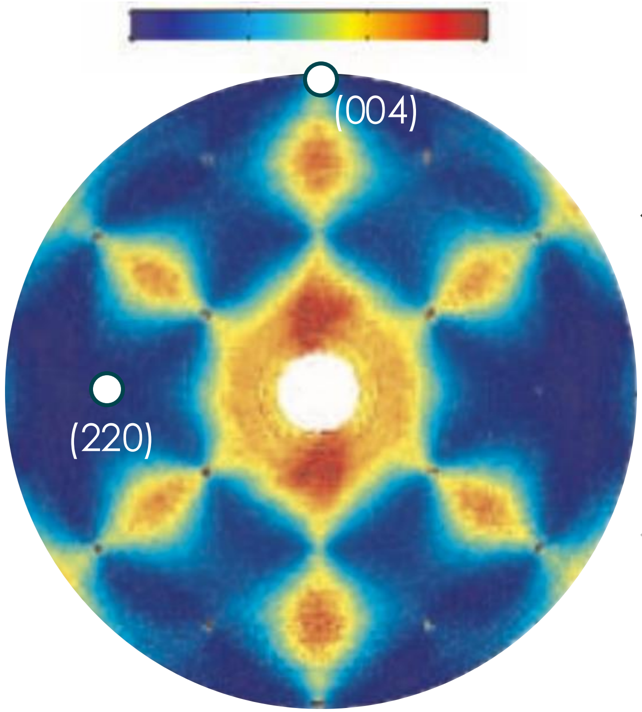


Baral *et al.*, *Matter* **5,** 1853 (2022)

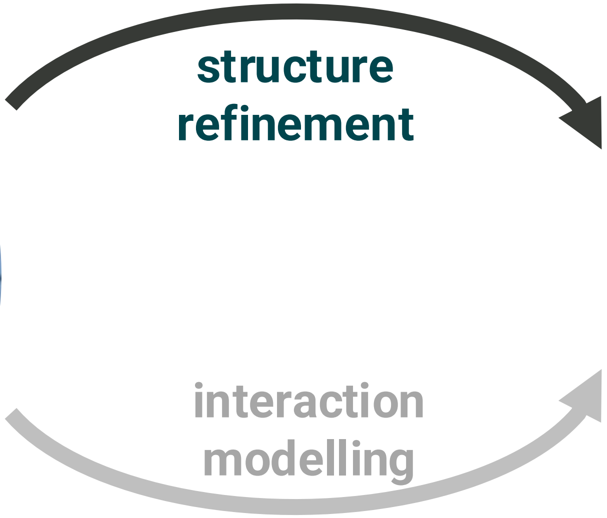
diffpy.mpdf software: Frandsen *et al.*, *J Appl. Cryst.* **5**, 1377 (2022). **https://www.diffpy.org/products/mPDF.html**

How can we analyse diffuse scattering data?

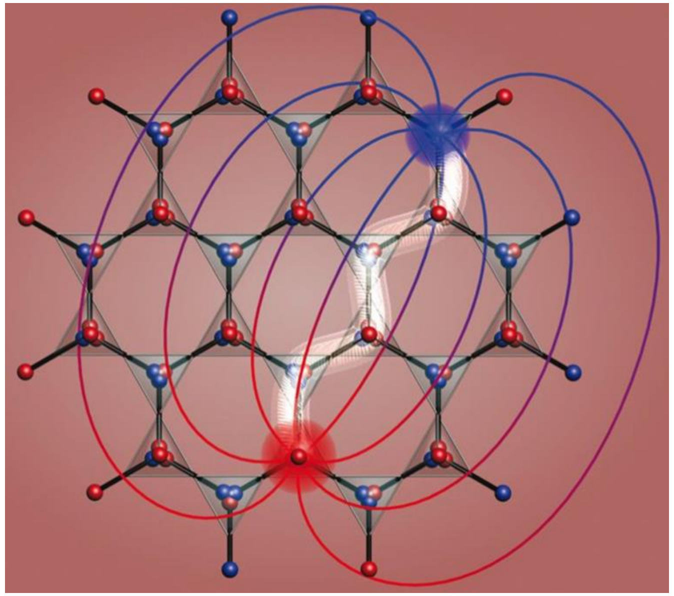
Data*



Reciprocal space



Model**

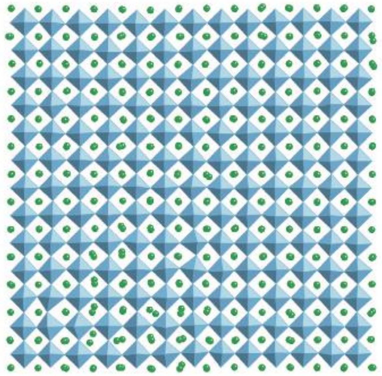


Real space

*Fennell *et al.*, *Science* **326**, 415 (2009)
Castelnovo, Moessner & Sondhi, *Nature* **451, 42 (2008)

How can we analyse diffuse scattering data?

Big-box refinement



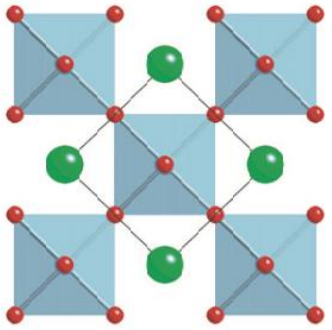
Many unit cells (~10^4 atoms) to capture long-range *and* short-range order

e.g. reverse Monte Carlo

→ *See also RMCProfile*

structure refinement

Small-box refinement



Single unit cell to capture short-range order ("real-space Rietveld")

e.g. mPDF analysis

→ *Ben Frandsen's talk*

Reverse Monte Carlo (RMC) method

Create ~10^3 spins with random orientations and fixed positions

Flip or rotate a randomly-chosen spin

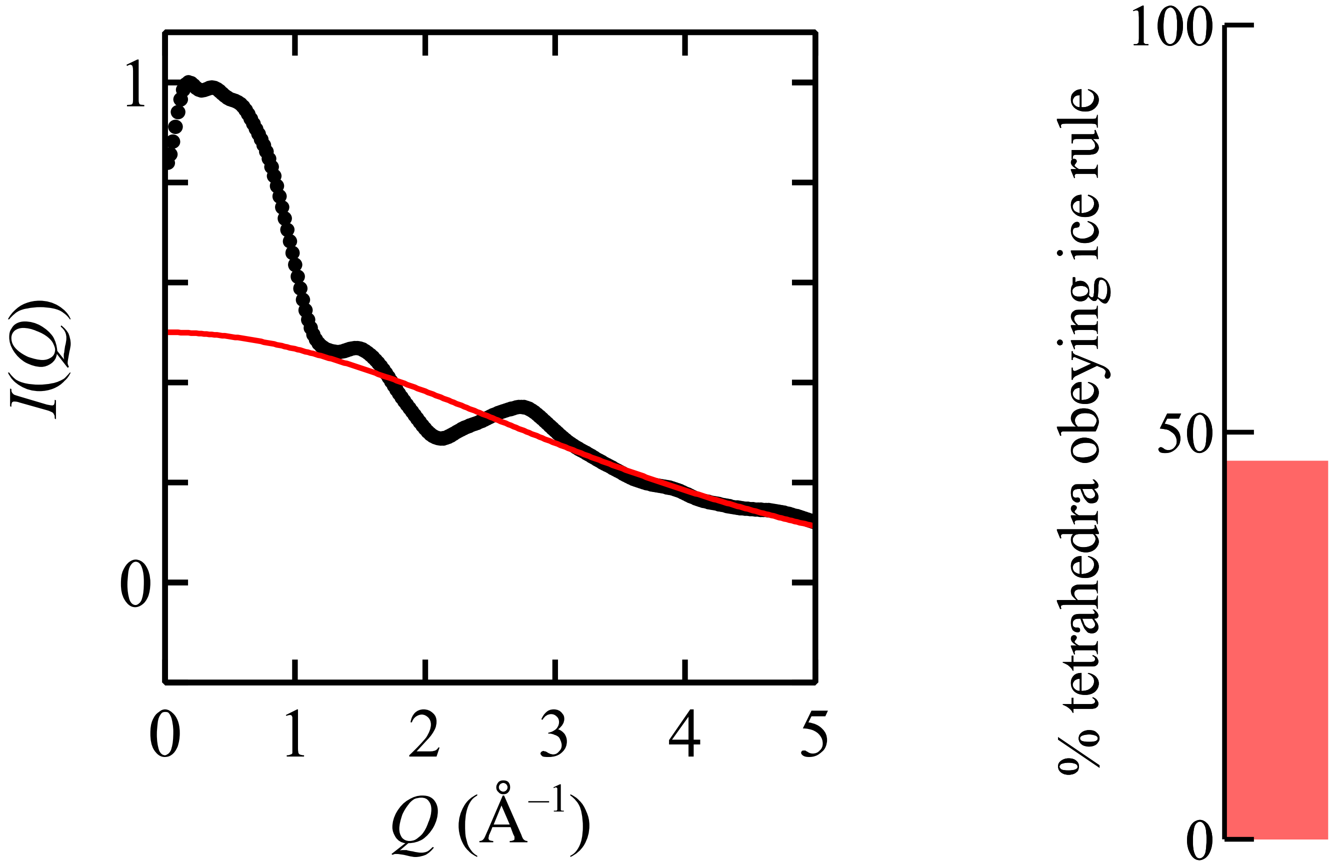
Calculate change in goodness-of-fit to data

Accept flip if fit improved; otherwise accept flip with some probability

RMCProfile software: Tucker *et al.*, *JPCM* **19,** 335218 (2007)
Spinvert software: Paddison, Stewart, Goodwin *JPCM* **25,** 454220 (2013). **www.joepaddison.com/software**

RMC: Proof of principle for spin ice

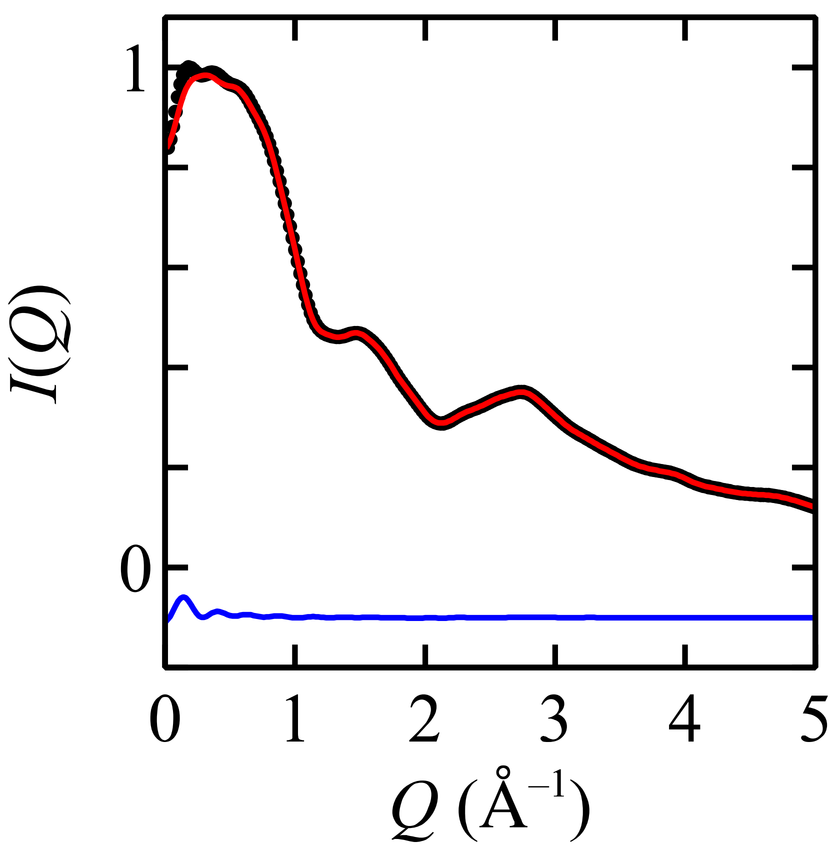
- e.g. fit to virtual “data” for spin ice

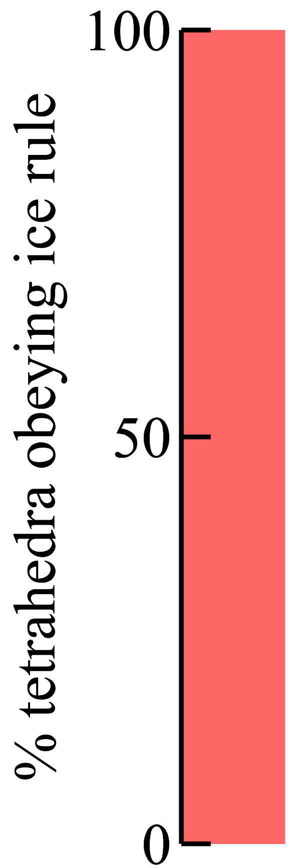


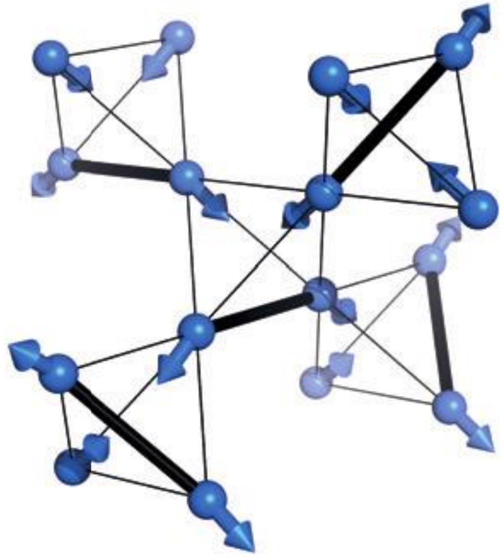
Paddison & Goodwin, *PRL* **108,** 017204 (2012)

RMC: Proof of principle for spin ice

- e.g. fit to virtual “data” for spin ice

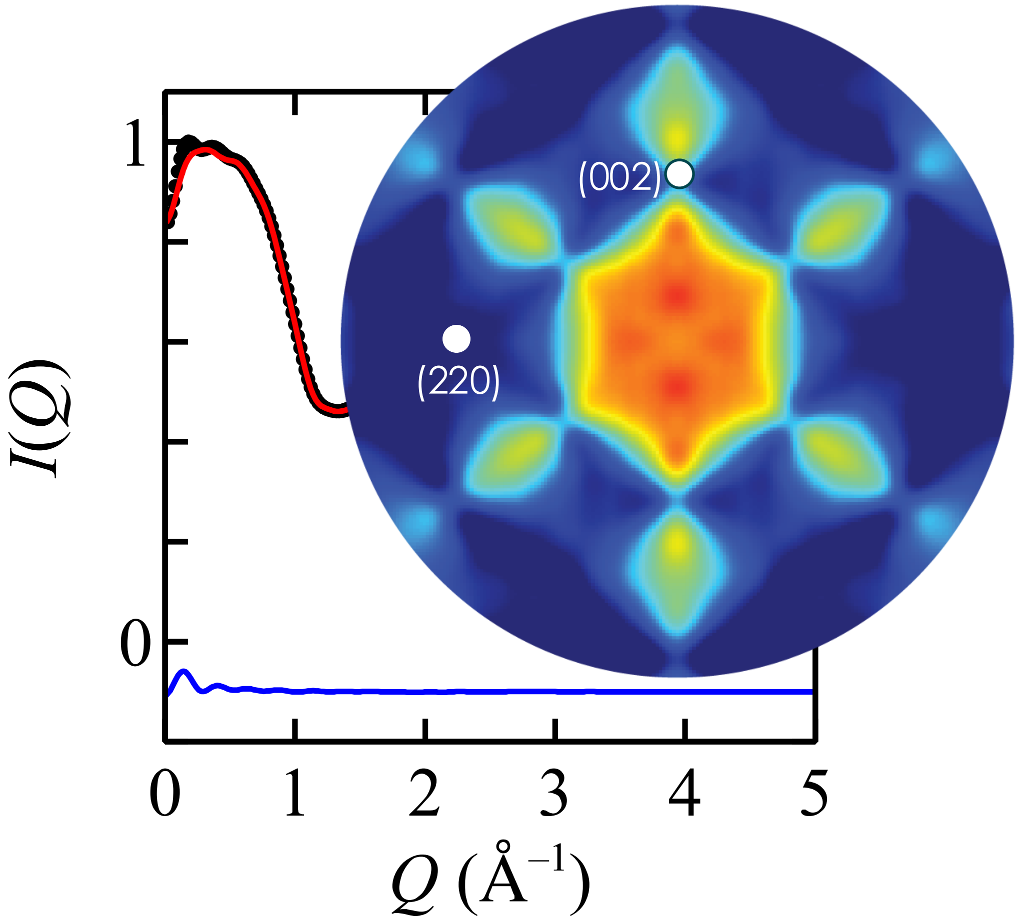


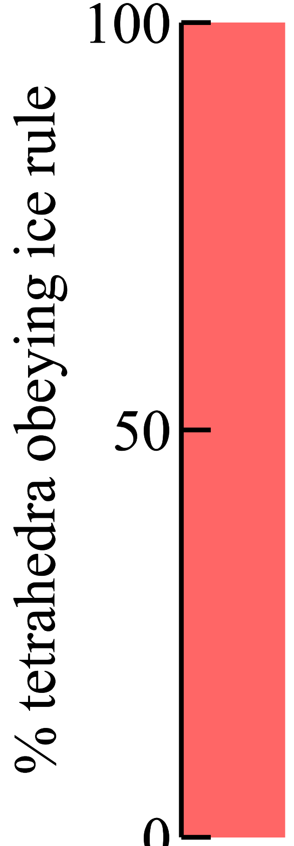


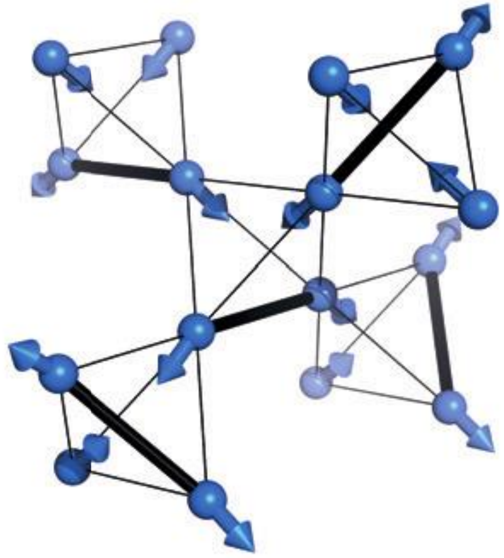


Paddison & Goodwin, *PRL* **108,** 017204 (2012)

RMC: Proof of principle for spin ice

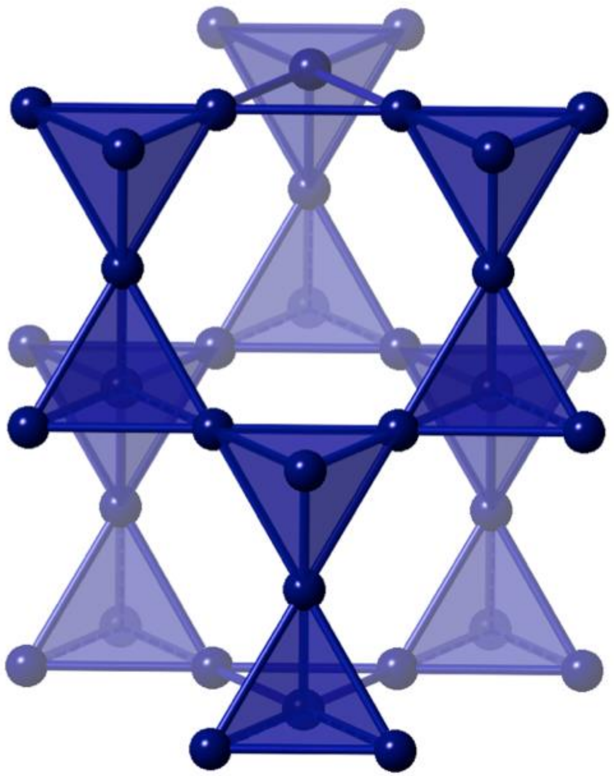


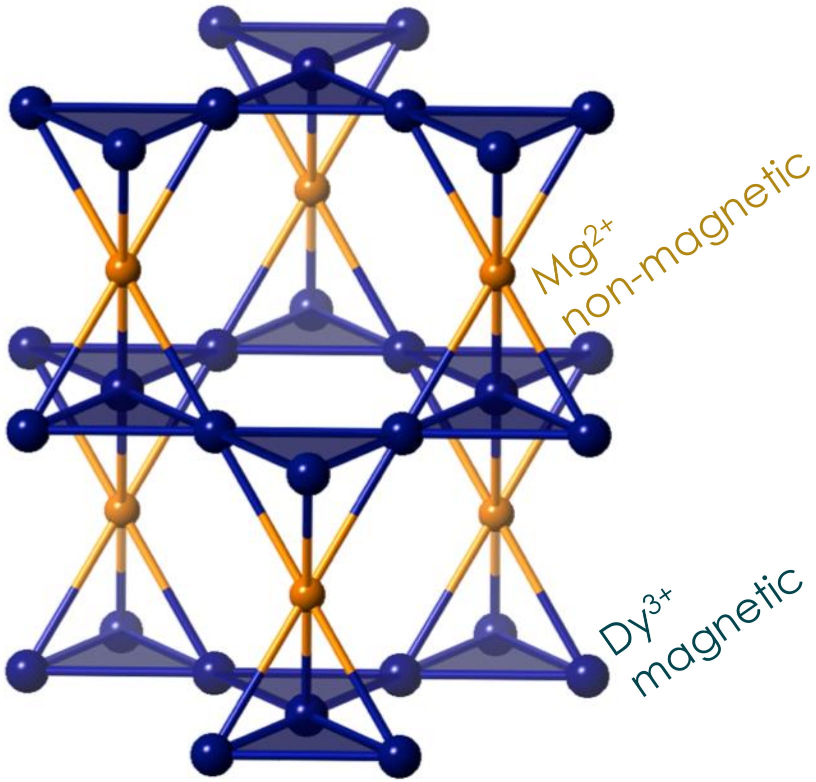




Paddison & Goodwin, *PRL* **108,** 017204 (2012)

RMC example: Kagome ice $Dy_3Mg_2Sb_3O_{14}$





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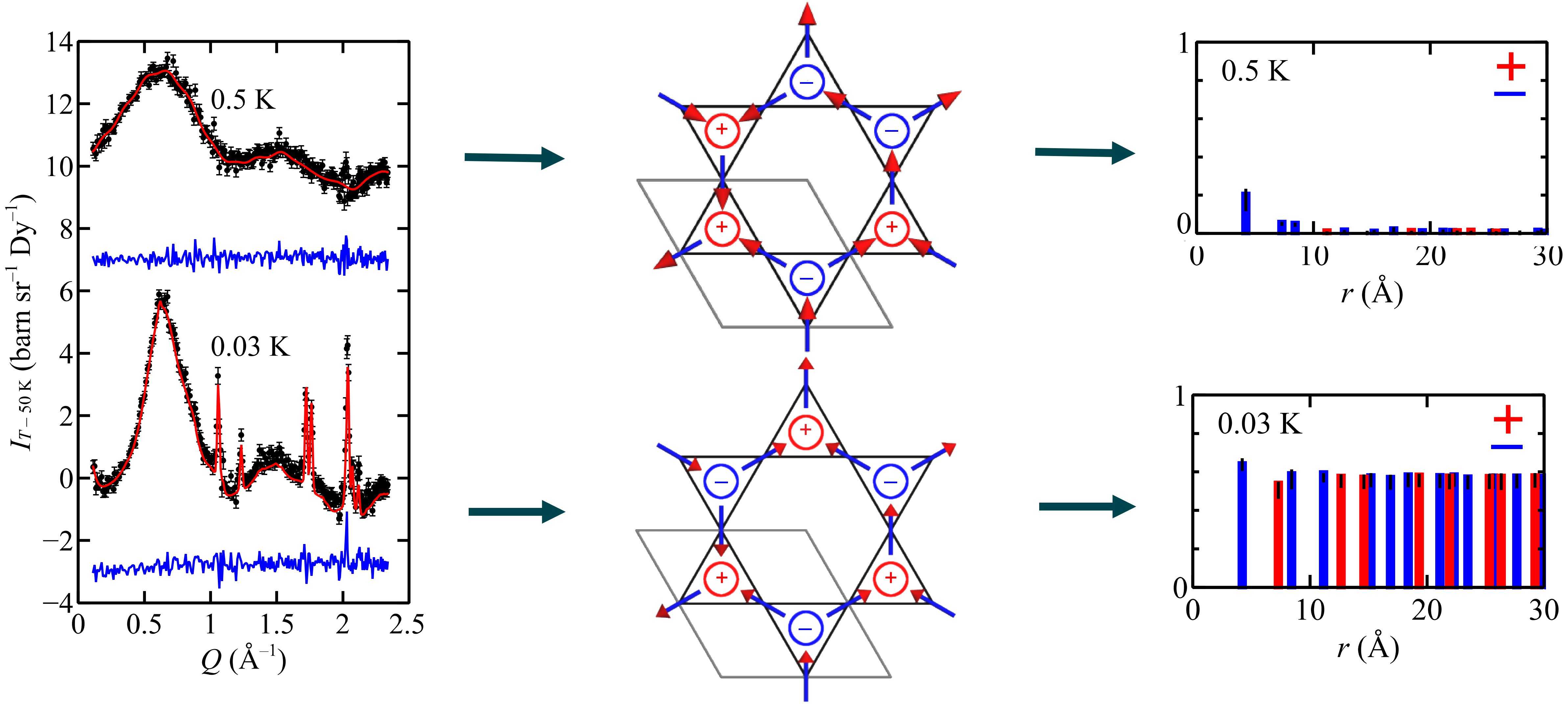
Sanders *et al.*, *J. Mater. Chem. C* **4**, 541–550 (2016)

RMC example: Kagome ice $Dy_3Mg_2Sb_3O_{14}$

Diffuse scattering

Local magnetic structure

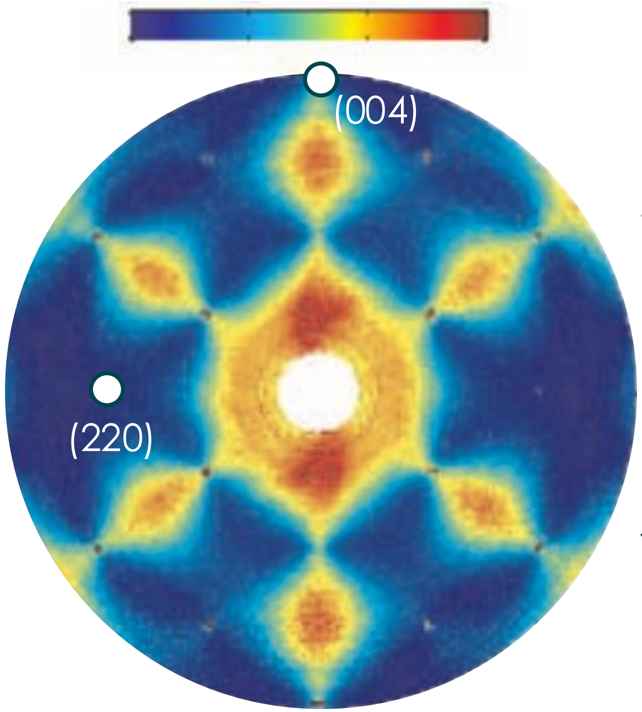
"Emergent charge" correlations



Paddison *et al.*, *Nature Commun.* **7**, 13842 (2016)

How can we analyse diffuse scattering data?

Data*

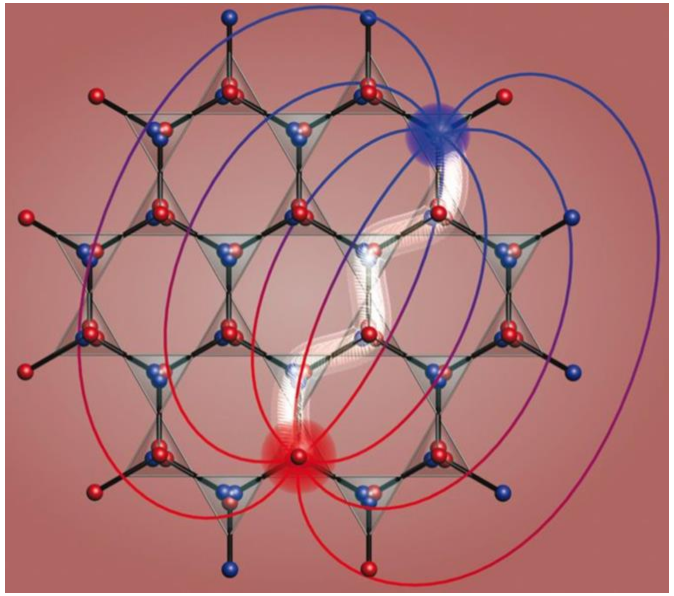


Reciprocal space

structure refinement

interaction modelling

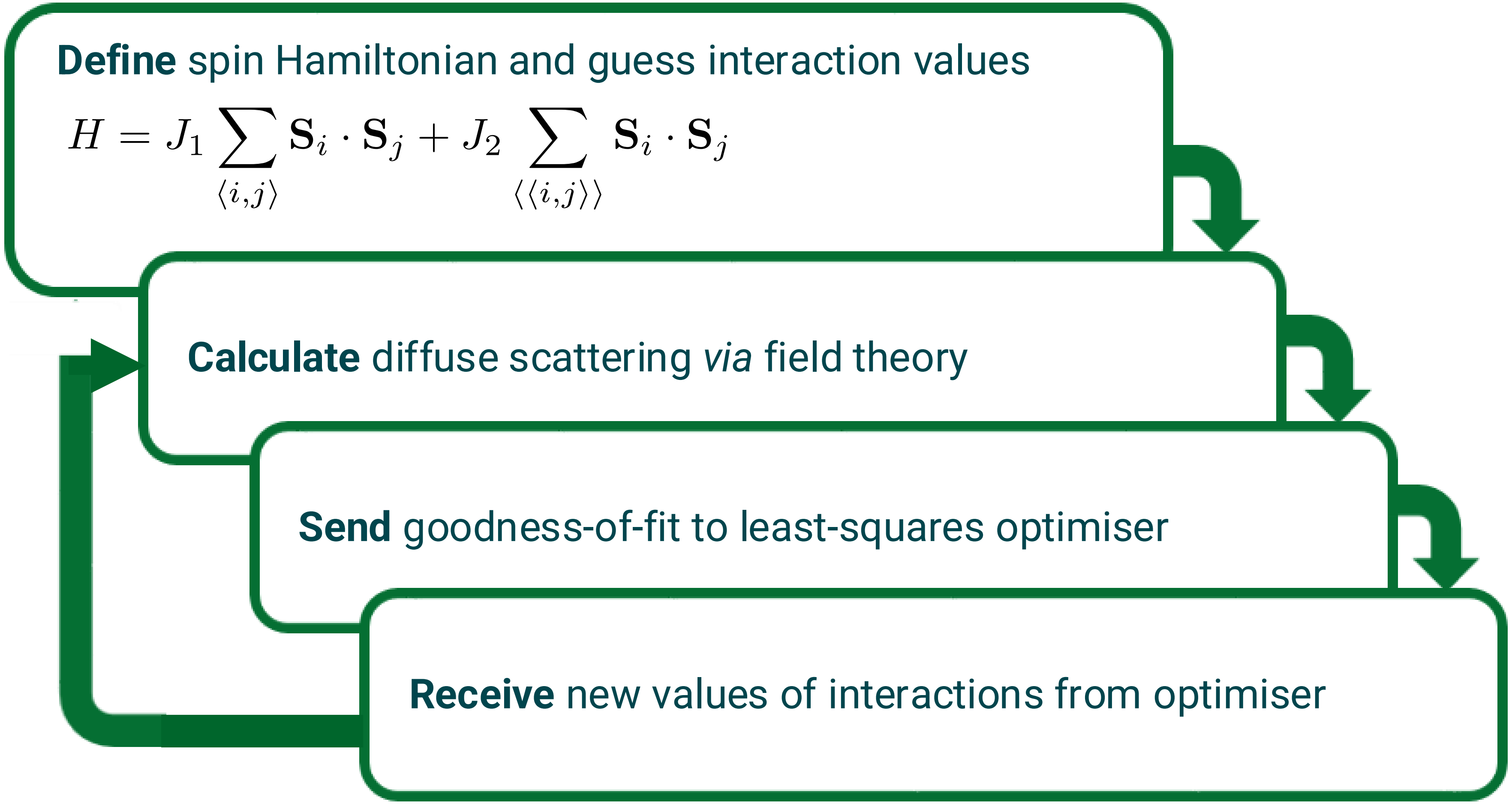
Model**



Real space

*Fennell *et al.*, *Science* **326**, 415 (2009)
Castelnovo, Moessner & Sondhi, *Nature* **451, 42 (2008)

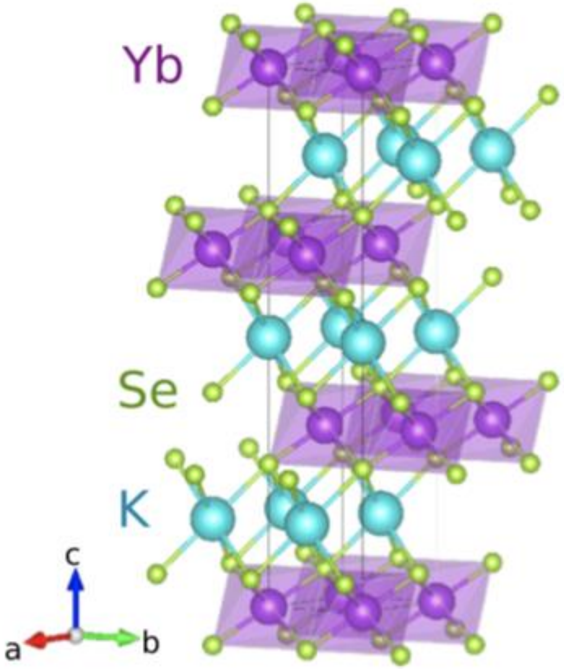
Interaction refinement

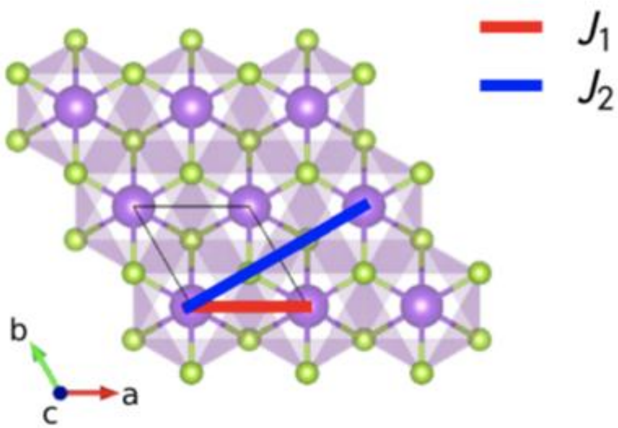


Spinteract software: Paddison, *J Phys Cond Matt* **35,** 495802 (2023). **www.joepaddison.com/software**
Sunny software: Dahlbom *et al.*, *arXiv:2501.13095* (2025). **https://github.com/SunnySuite/Sunny.jl**

Interaction refinement example 1: $KYbSe_2$

- Triangular lattice of Yb^{3+} with effective spin-½





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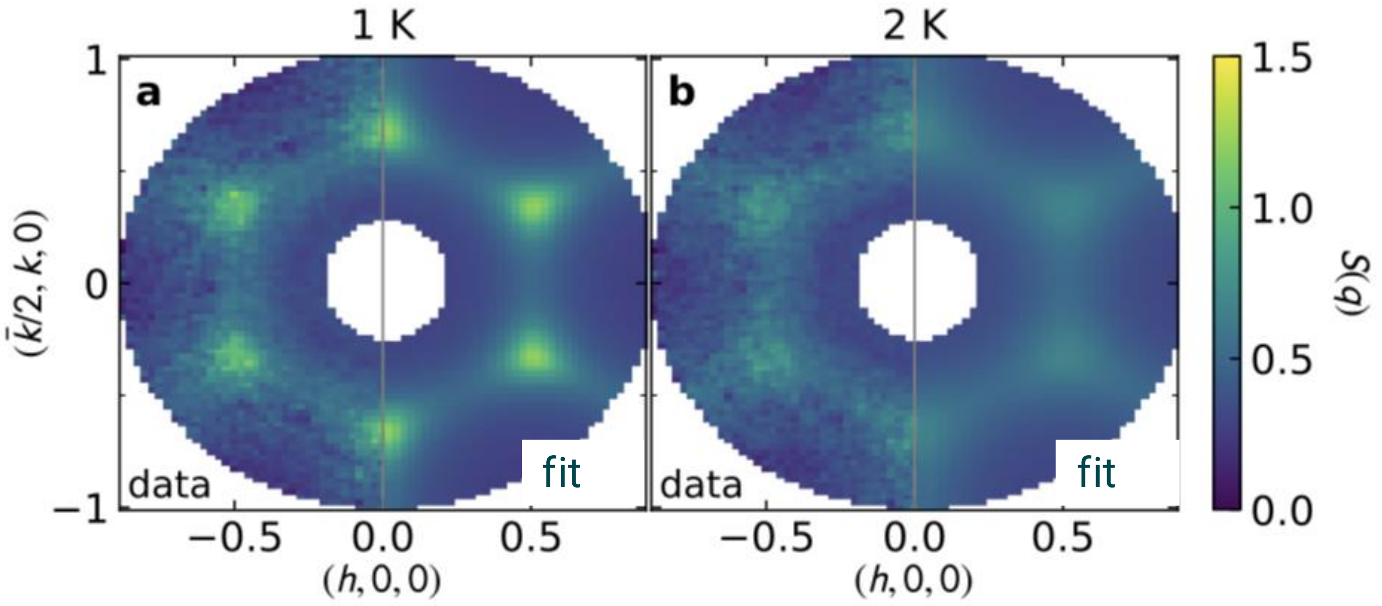
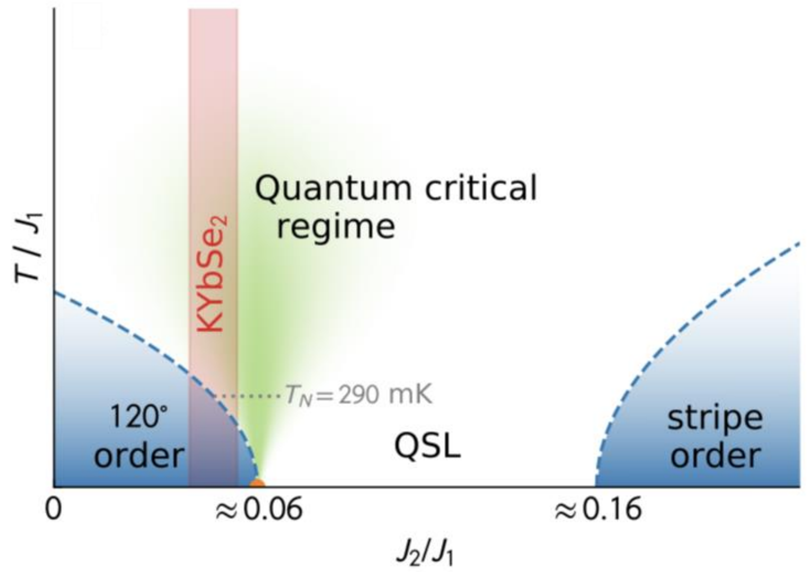
Scheie, Ghioldi, Xing, Paddison *et al.*, *Nat. Phys.* **20**, 74 (2024)
Scheie *et al.*, *PRB* **109**, 014425 (2022)

Interaction refinement example 1: $KYbSe_2$

- Fits show <3% deviation from Heisenberg model



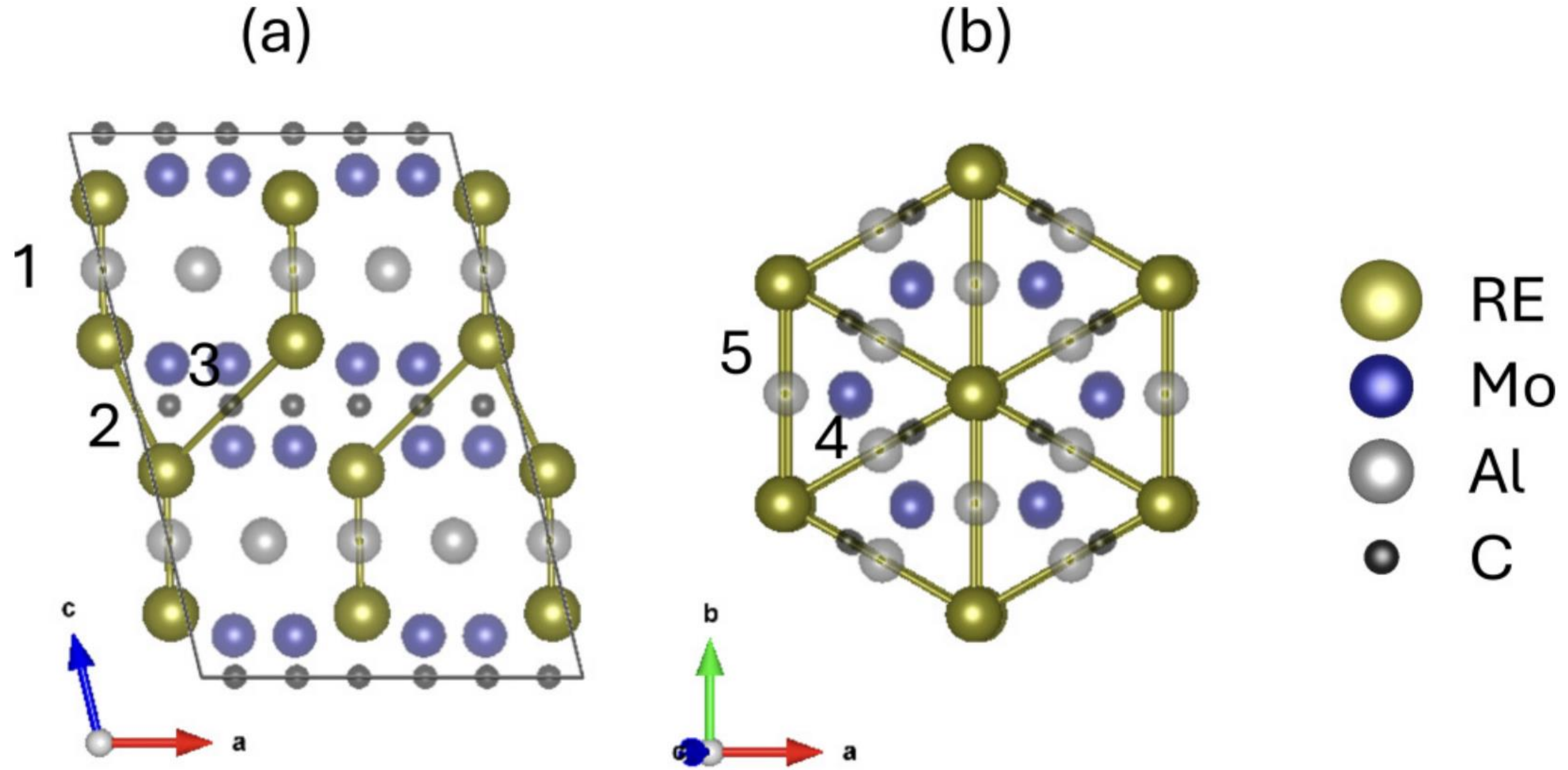


Theoretical technique	J_1 (meV)	J_2/J_1
Onsager reaction field	*NA*	0.047 ± 0.007
Nonlinear spin waves	0.456 ± 0.013	0.043 ± 0.010
Heat capacity	0.429 ± 0.010	0.037 ± 0.013
Weighted mean:	0.438 ± 0.008	0.044 ± 0.005

Scheie, Ghioldi, Xing, Paddison *et al.*, *Nat. Phys.* **20**, 74 (2024)
Scheie *et al.*, *PRB* **109**, 014425 (2022)

Interaction refinement example 2: $(Mo_{2/3}\mathit{RE}_{1/3})_2AlC$

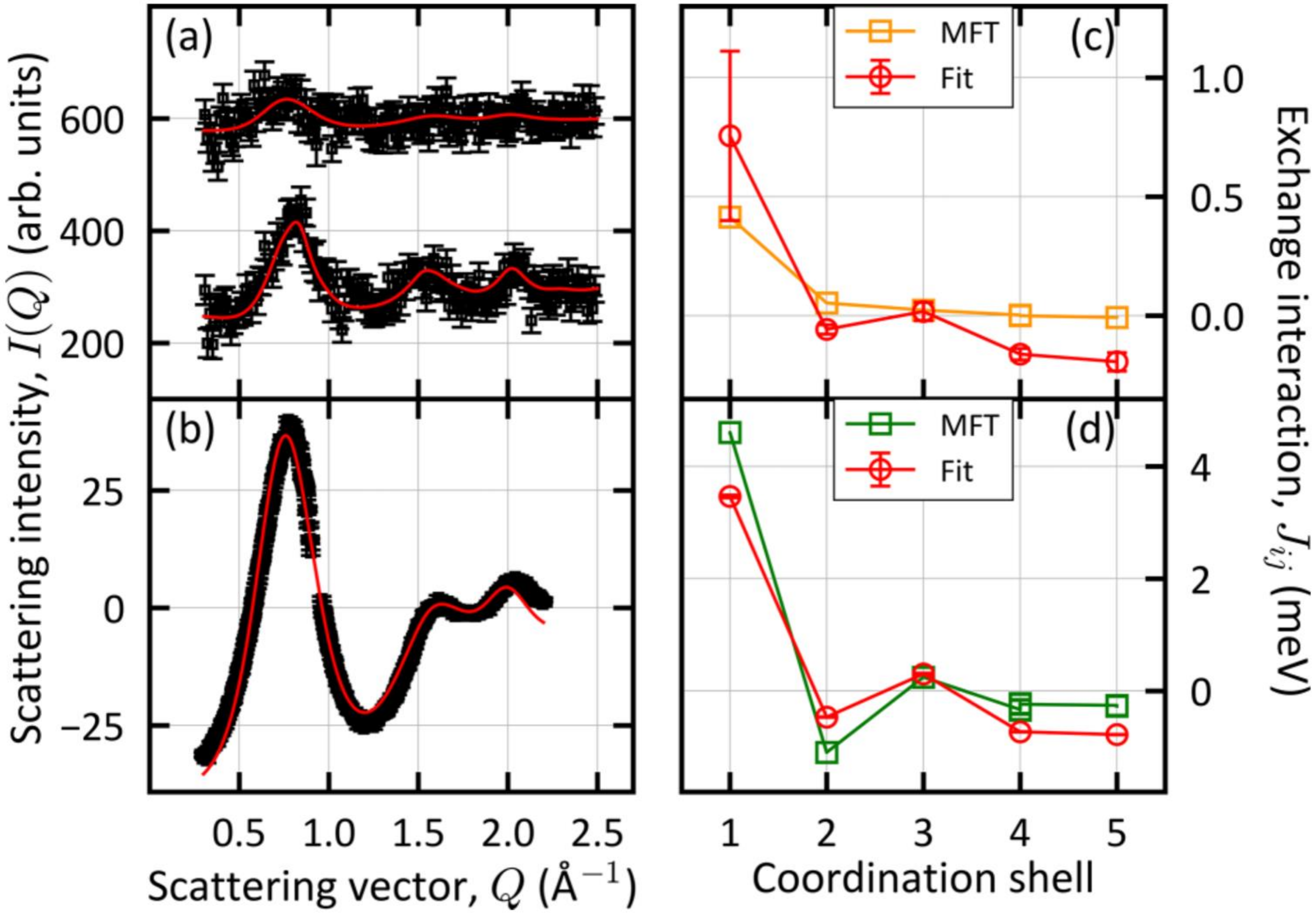
- Quasi-2D magnets with triangular lattice of rare-earth (*RE*) ions



Potashniknov *et al.*, *PRB* **112**, 064407 (2025)

Interaction refinement example 2: $(Mo_{2/3}Ln_{1/3})_2AlC$

- Fits show good agreement with magnetic force theorem (DFT) simulations



Potashniknov *et al.*, *PRB* **112**, 064407 (2025)

Thanks for listening (and hope to see you at the tutorials!)

- Diffuse scattering is a powerful technique to understand quantum materials
  - Powder data can be information rich

- We can combine real space, reciprocal space, and interaction space methods

Tutorial files, also at
www.joepaddison.com/software

Tutorial Zoom link