



Introduction to magnetic diffuse scattering

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ENERGY

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

What is diffuse scattering?

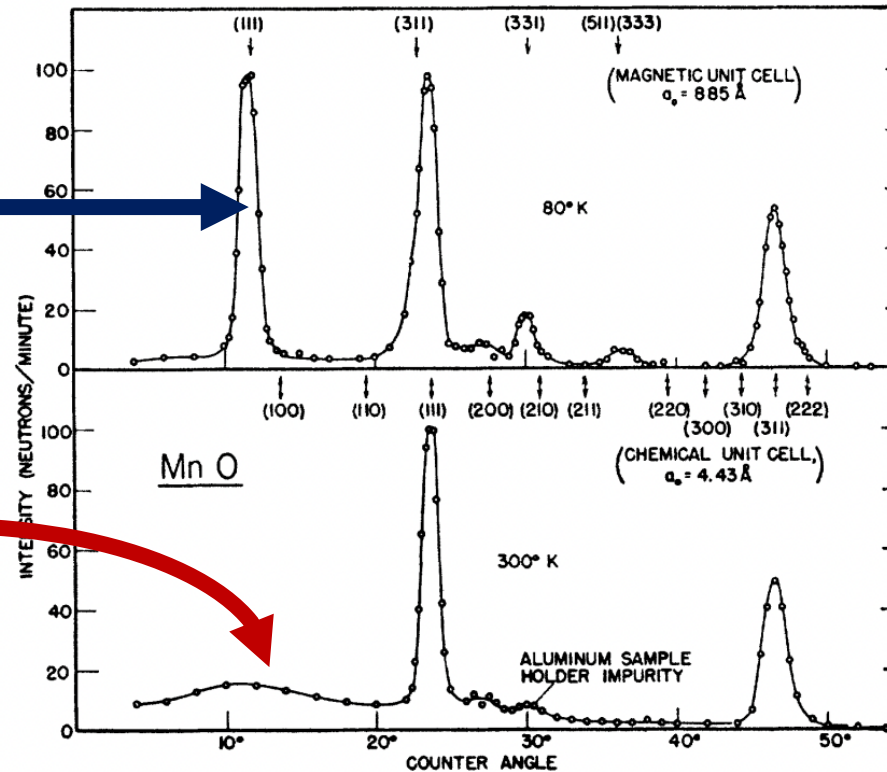
- Broad features beneath and between the sharp (Bragg) peaks
- e.g. magnetic scattering from MnO



Clifford Shull

Magnetic Bragg scattering
→ *long-range magnetic order*

Magnetic diffuse scattering
→ *short-range magnetic order*



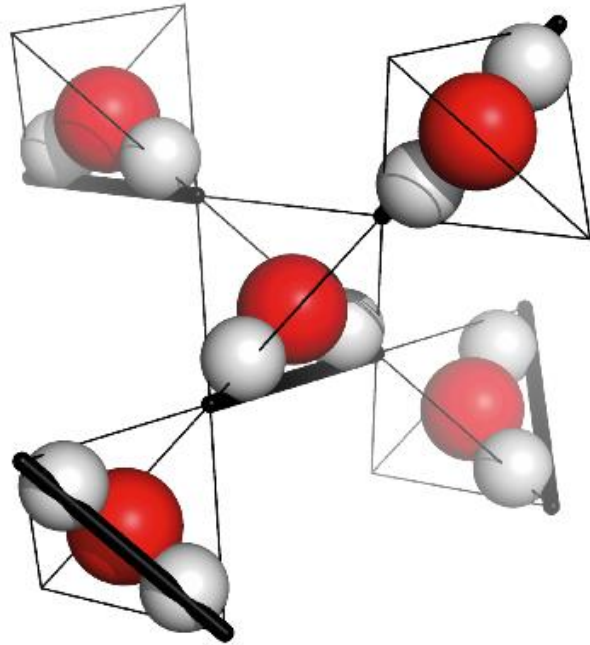
$$T < T_N$$

$$T > T_N$$

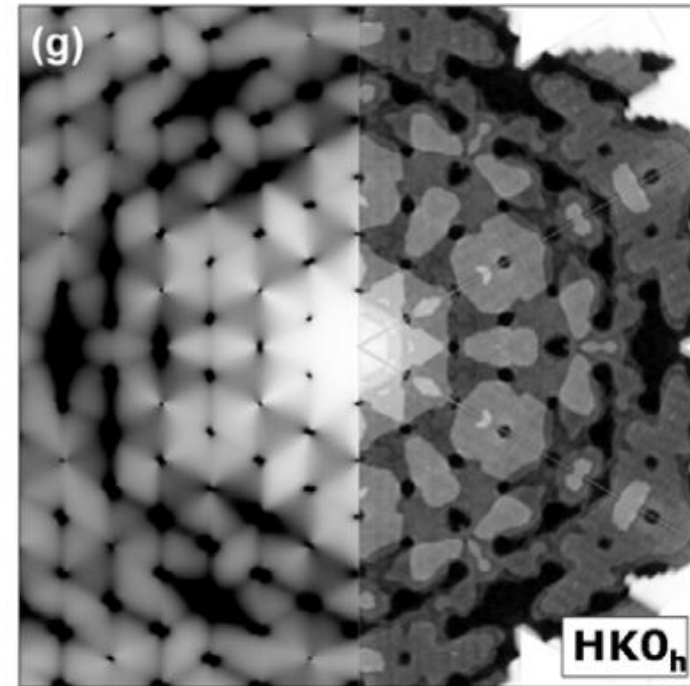
FIG. 1. Neutron diffraction patterns for MnO at room temperature and at 80°K.

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



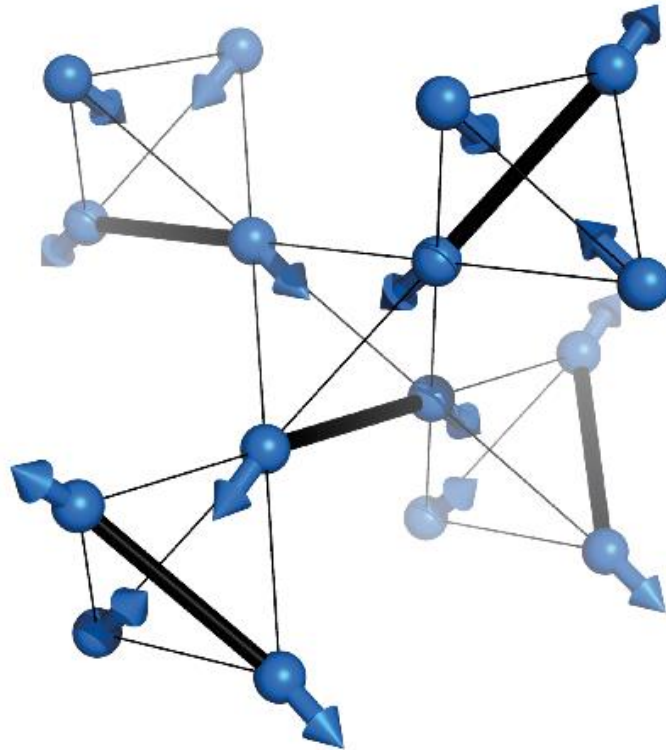
Water ice
Real space, r



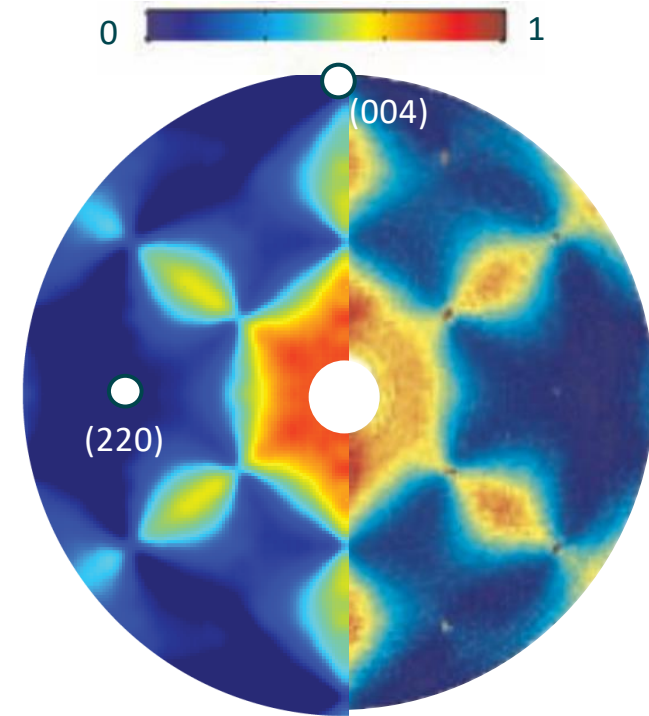
Model **Neutron data***
Reciprocal space, Q

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



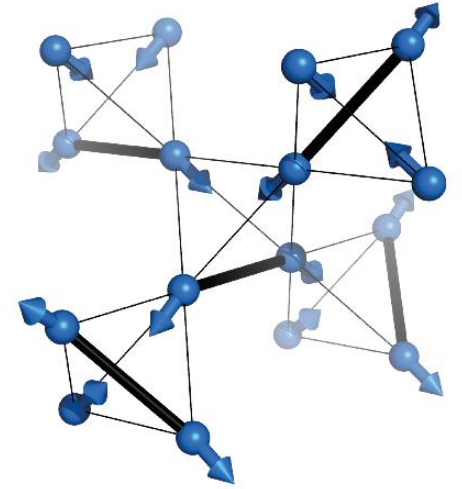
Model

Neutron data*

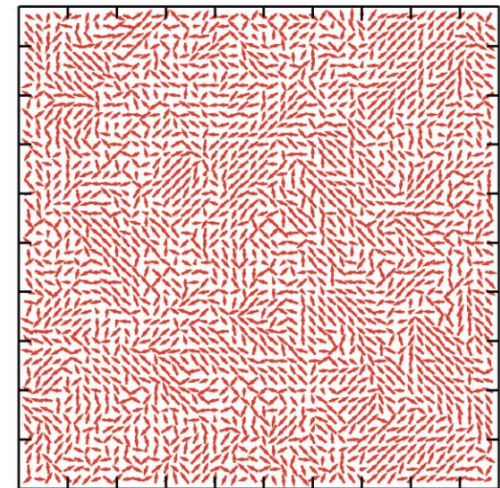
Reciprocal space, Q

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

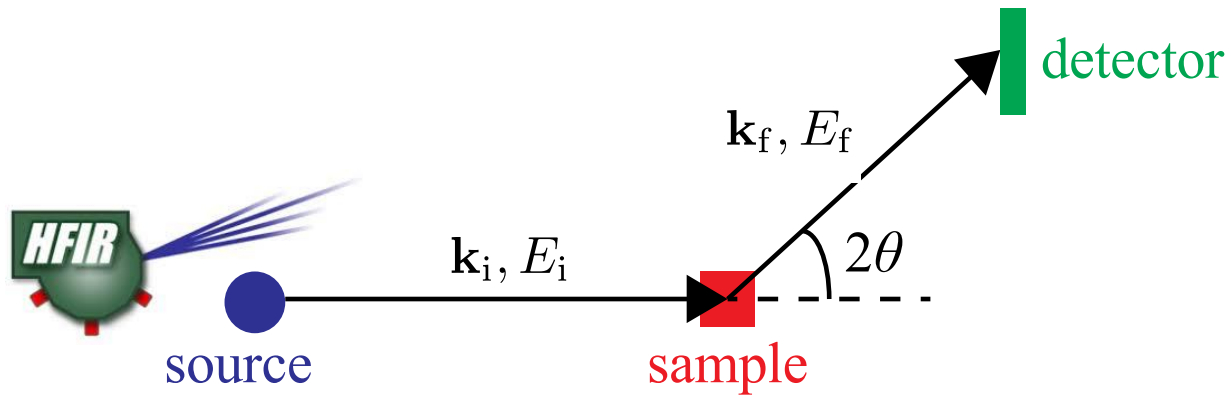


Spin orientations in spin ice



*Dipole orientations in a relaxor ferroelectric**

How do we measure diffuse scattering using neutrons?



Energy transfer $E = E_i - E_f$

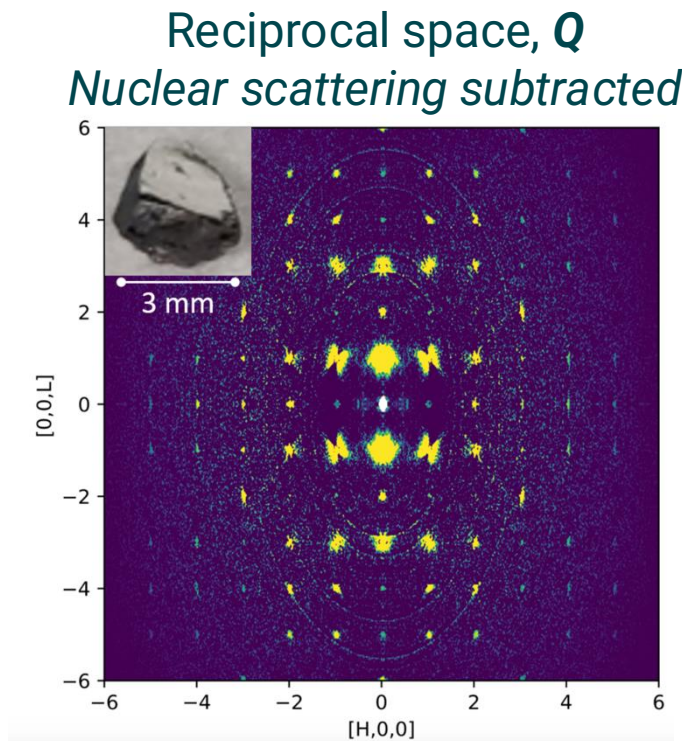
Wavevector transfer $\mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f$

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

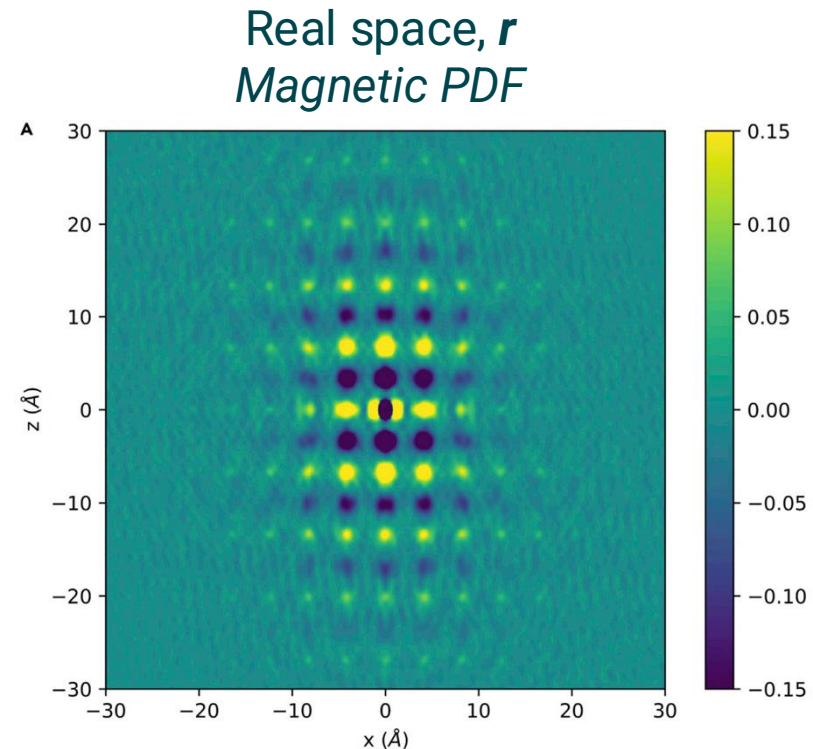
- **Quantity of interest** is E -integrated intensity $I(\mathbf{Q}) = \int_{-\infty}^{\infty} I(\mathbf{Q}, E) dE$
 - Approximated by diffraction measurements if $E \ll E_f$
- **For magnetic crystals:** Measure large volume of \mathbf{Q} (e.g. Corelli @ SNS)
- **For magnetic powders:** Maximise flux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

How can we visualise diffuse scattering data?

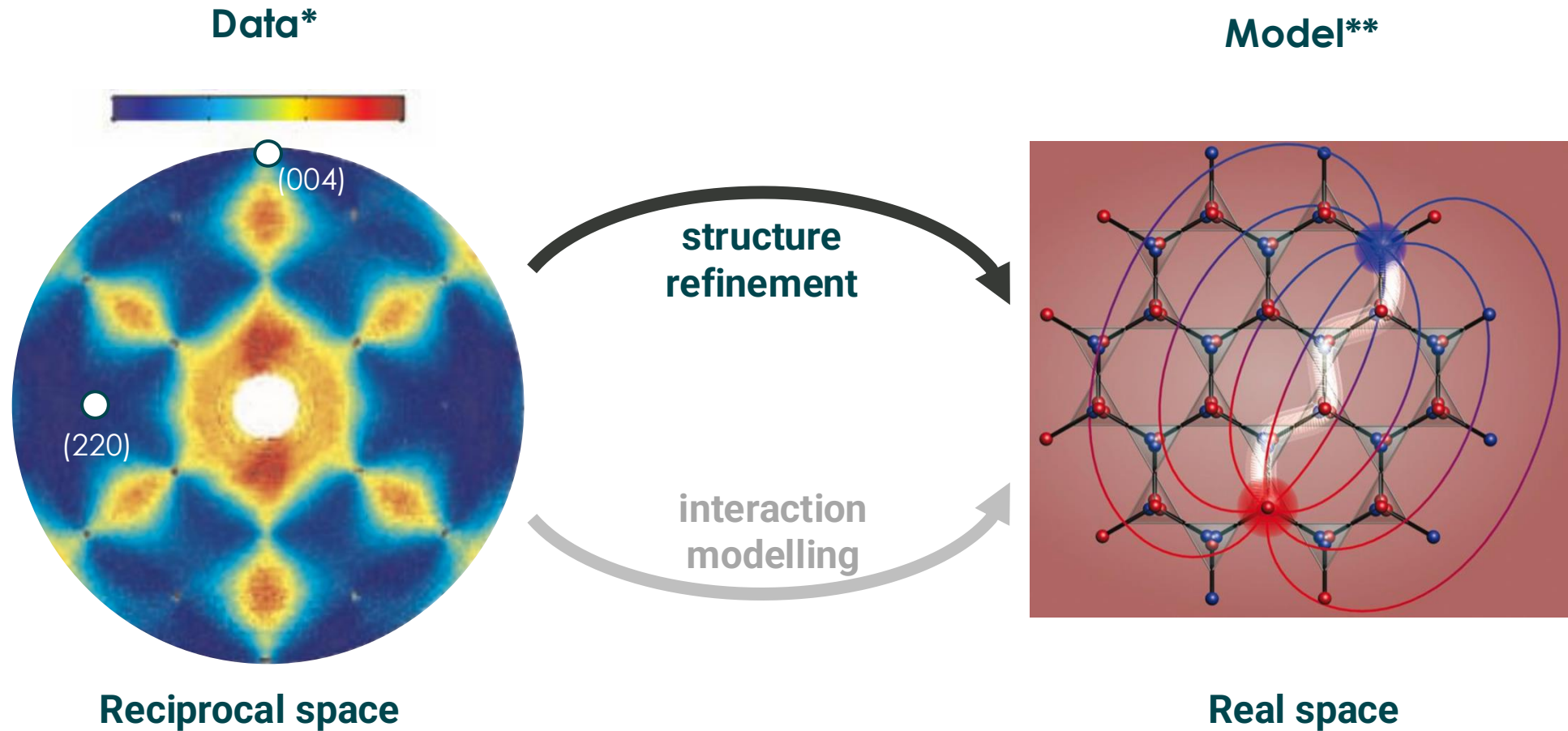
- Fourier transform of magnetic single-crystal data yields *3D magnetic PDF* (spin-pair correlation function)
 - Positive (negative) peaks indicate ferromagnetic (antiferromagnetic) correlations
 - e.g. magnetically-enhanced thermoelectric MnTe at $T > T_N$



Fourier transform
→



How can we analyse diffuse scattering data?

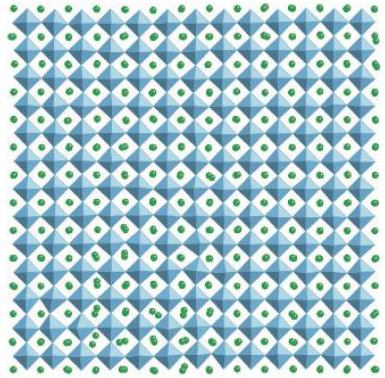


*Fennell *et al.*, *Science* **326**, 415 (2009)

Castelnovo, Moessner & Sondhi, *Nature* **451, 42 (2008)

How can we analyse diffuse scattering data?

Big-box refinement

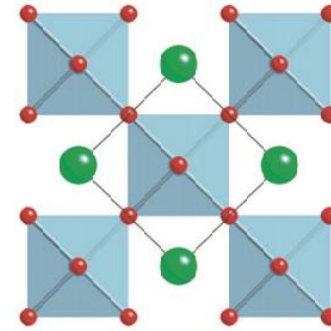


Many unit cells ($\sim 10^4$ atoms) to capture long-range *and* short-range order

e.g. reverse Monte Carlo
→ See also *RMCPProfile*

structure
refinement

Small-box refinement



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

e.g. mPDF analysis
→ Raju Baral’s talk

Reverse Monte Carlo (RMC) method

Create $\sim 10^3$ spins with random orientations and fixed positions

Flip or rotate a randomly-chosen spin

Calculate change in goodness-of-fit to data

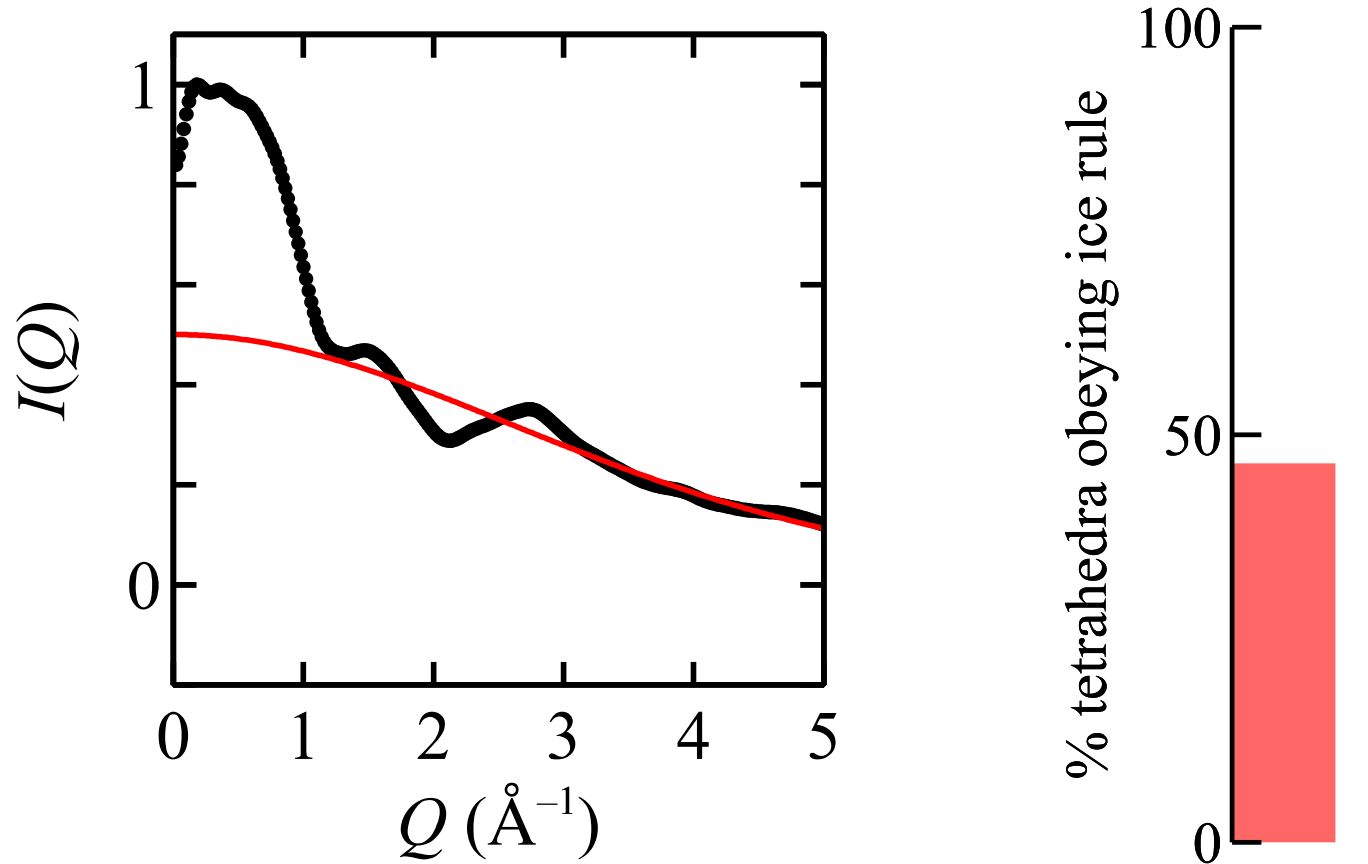
Accept flip if fit improved; otherwise accept flip with some probability

RMCPProfile software: Tucker *et al.*, *JPCM* **19**, 335218 (2007)

Spinvert software: Paddison, Stewart, Goodwin *JPCM* **25**, 454220 (2013). www.joepaddison.com/software

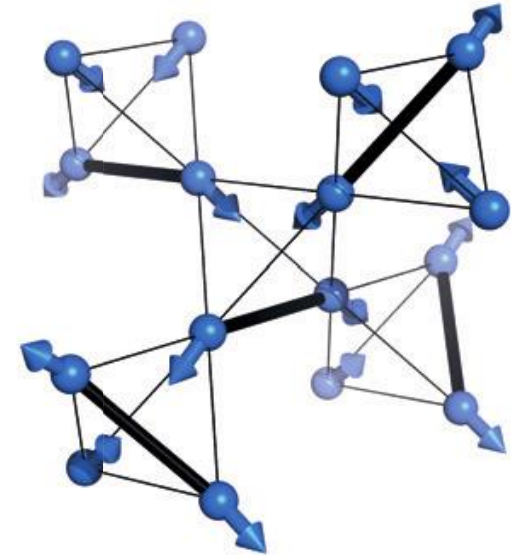
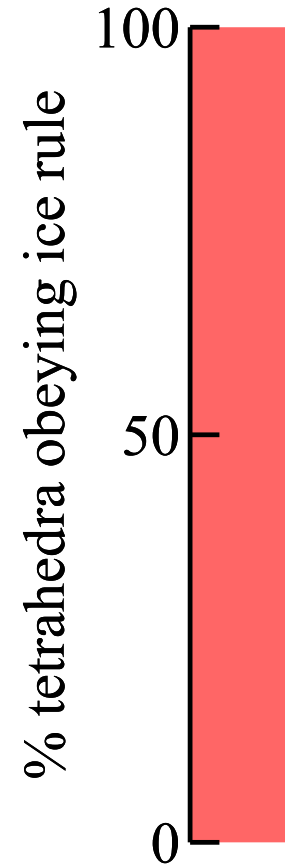
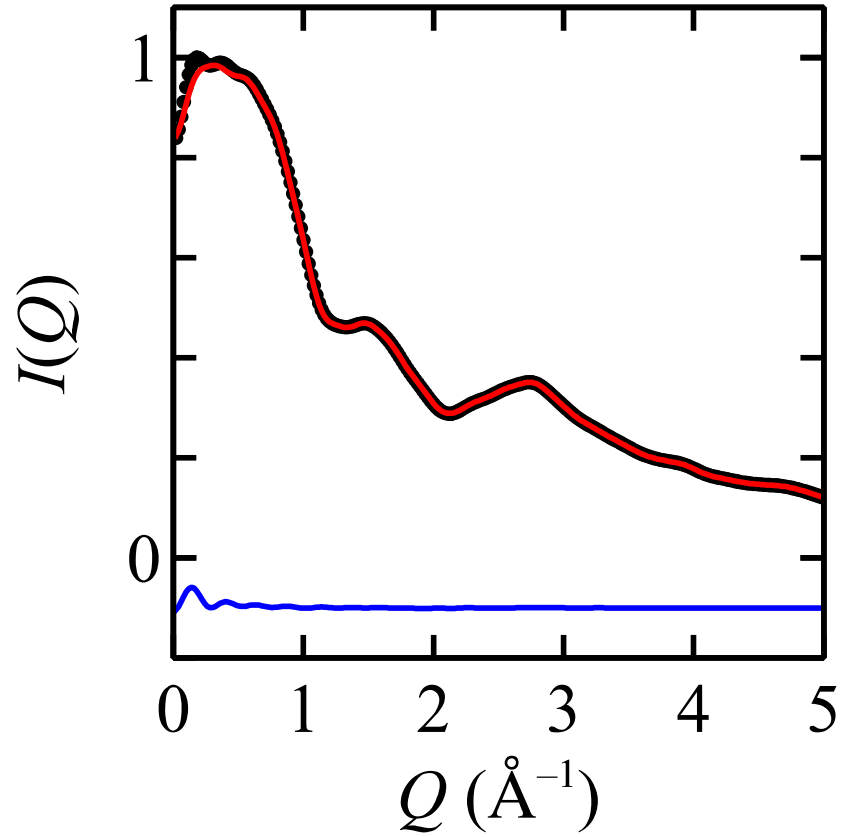
RMC: Proof of principle for spin ice

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

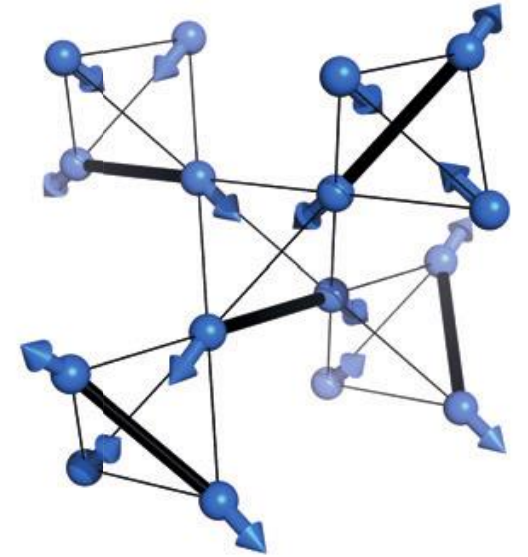
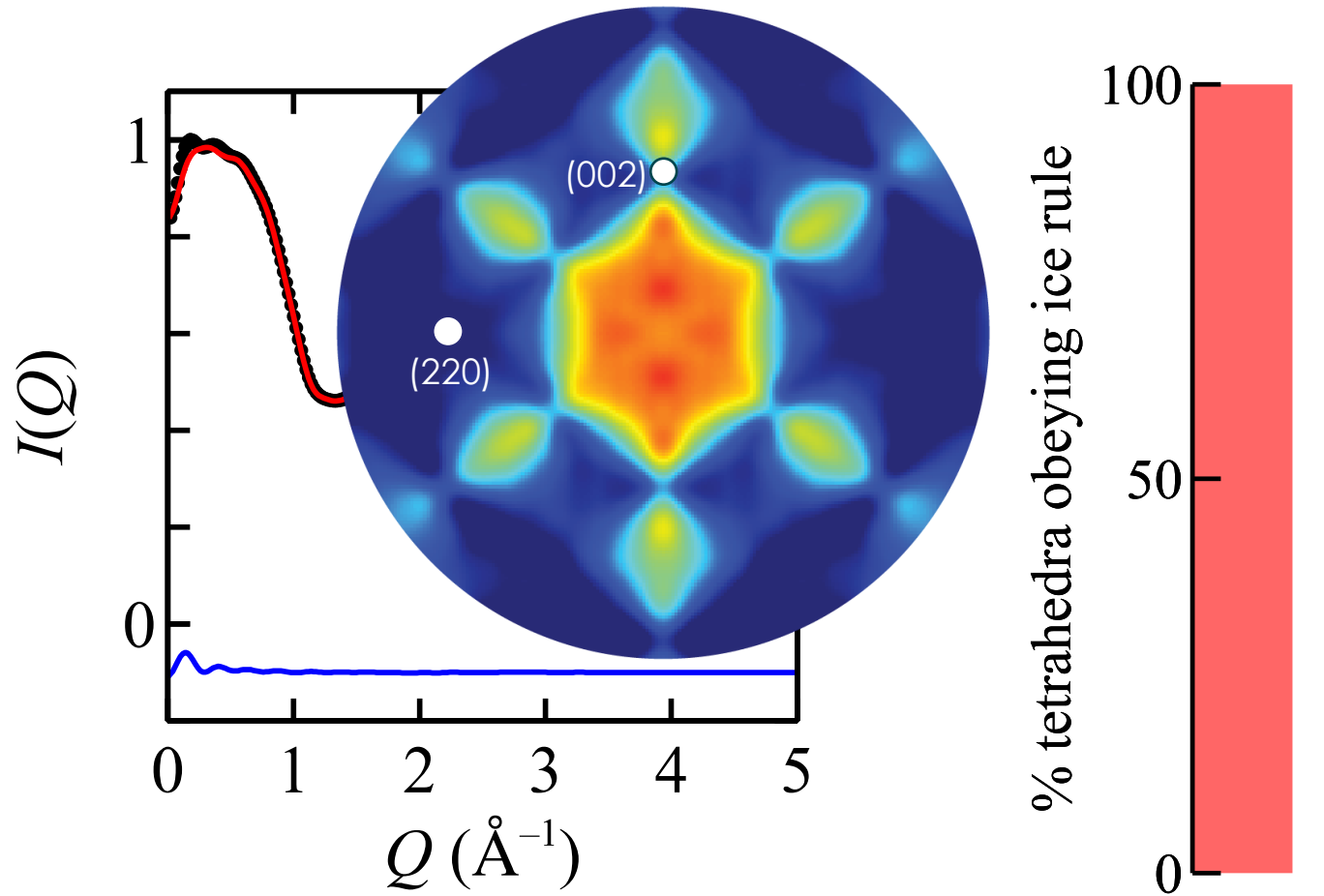


RMC: Proof of principle for spin ice

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

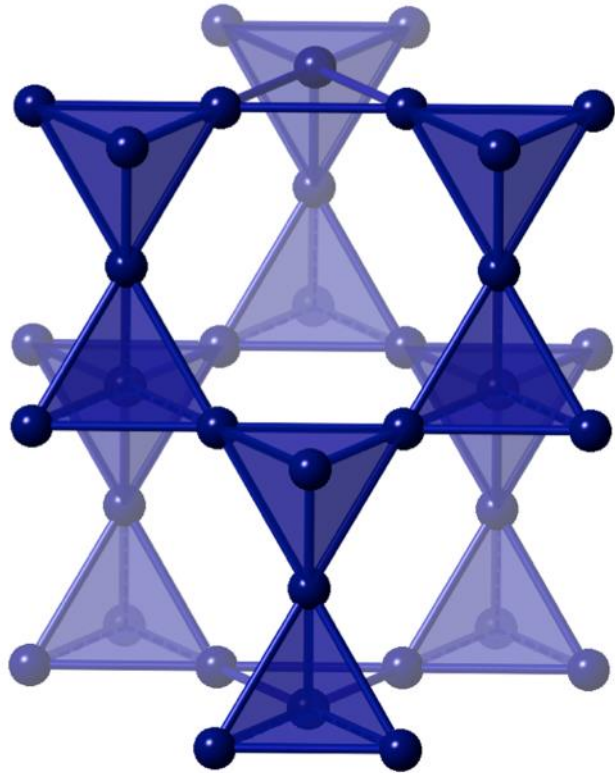


RMC: Proof of principle for spin ice



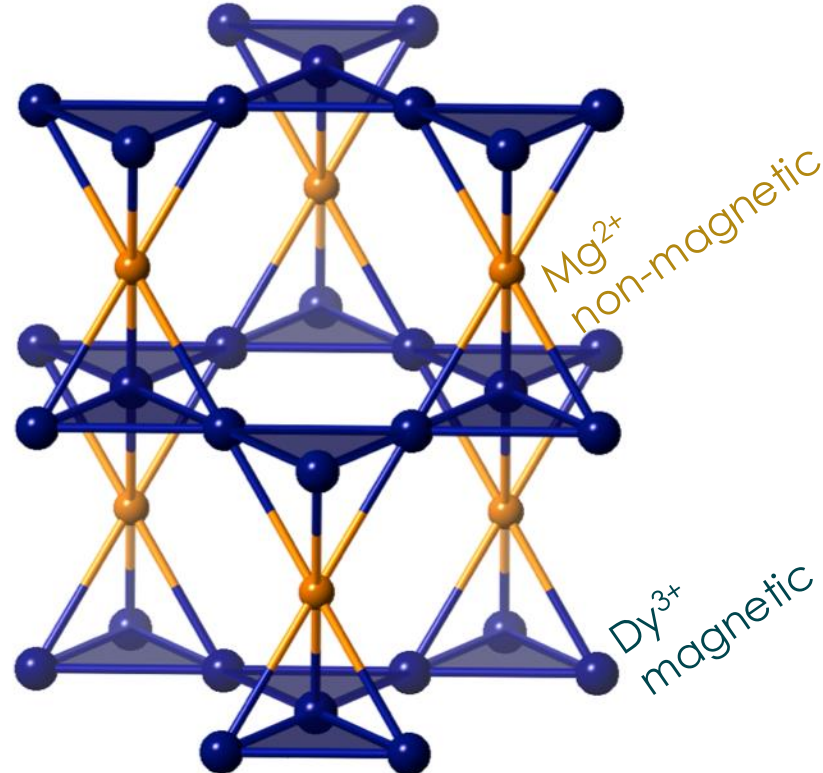
RMC example: Kagome ice $\text{Dy}_3\text{Mg}_2\text{Sb}_3\text{O}_{14}$

Pyrochlore $\text{Dy}_2\text{Ti}_2\text{O}_7$



Space group $Fd\bar{3}m$

Kagome $\text{Dy}_3\text{Mg}_2\text{Sb}_3\text{O}_{14}$



Space group $R\bar{3}m$

**Siân
Dutton**
Cambridge

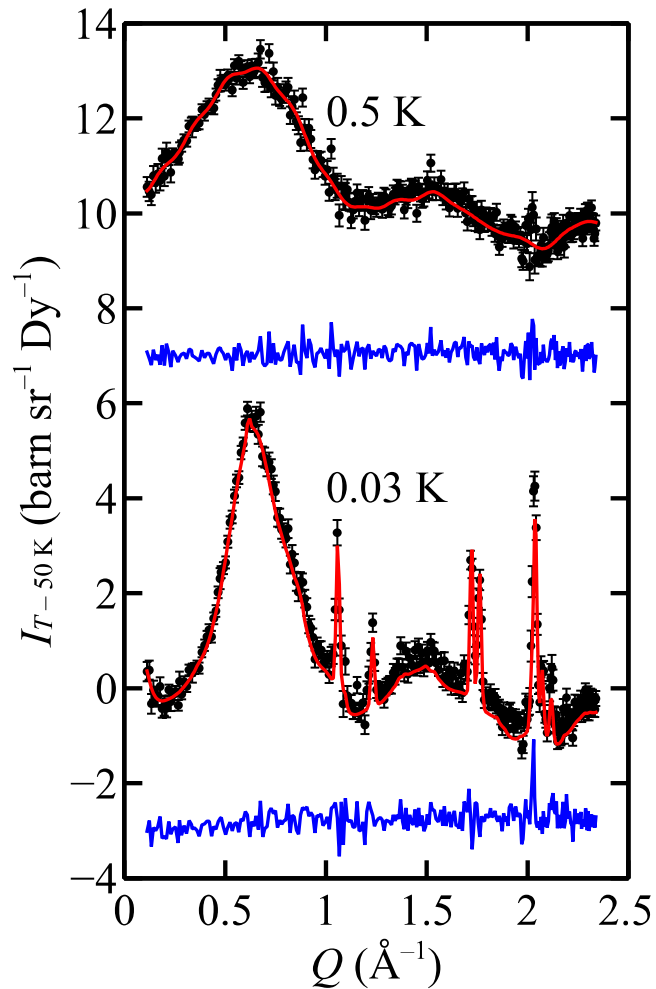
**Martin
Mourigal**
Georgia Tech

**Paromita
Mukherjee**
Cambridge

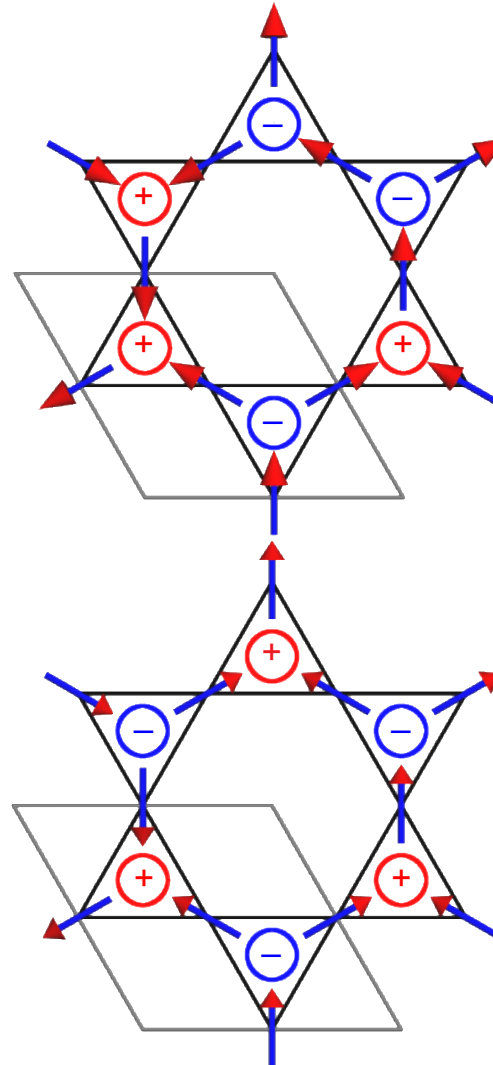
**Xiaojian
Bai**
Georgia Tech
→ LSU

RMC example: Kagome ice $\text{Dy}_3\text{Mg}_2\text{Sb}_3\text{O}_{14}$

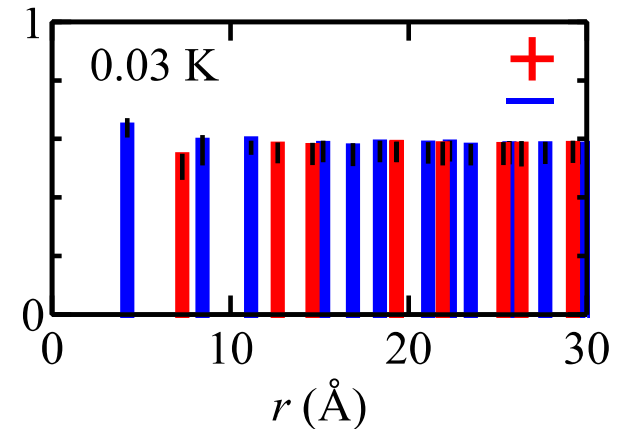
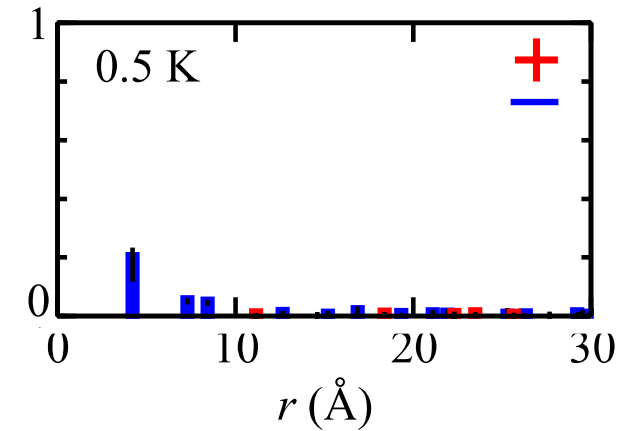
Diffuse scattering



Local magnetic structure

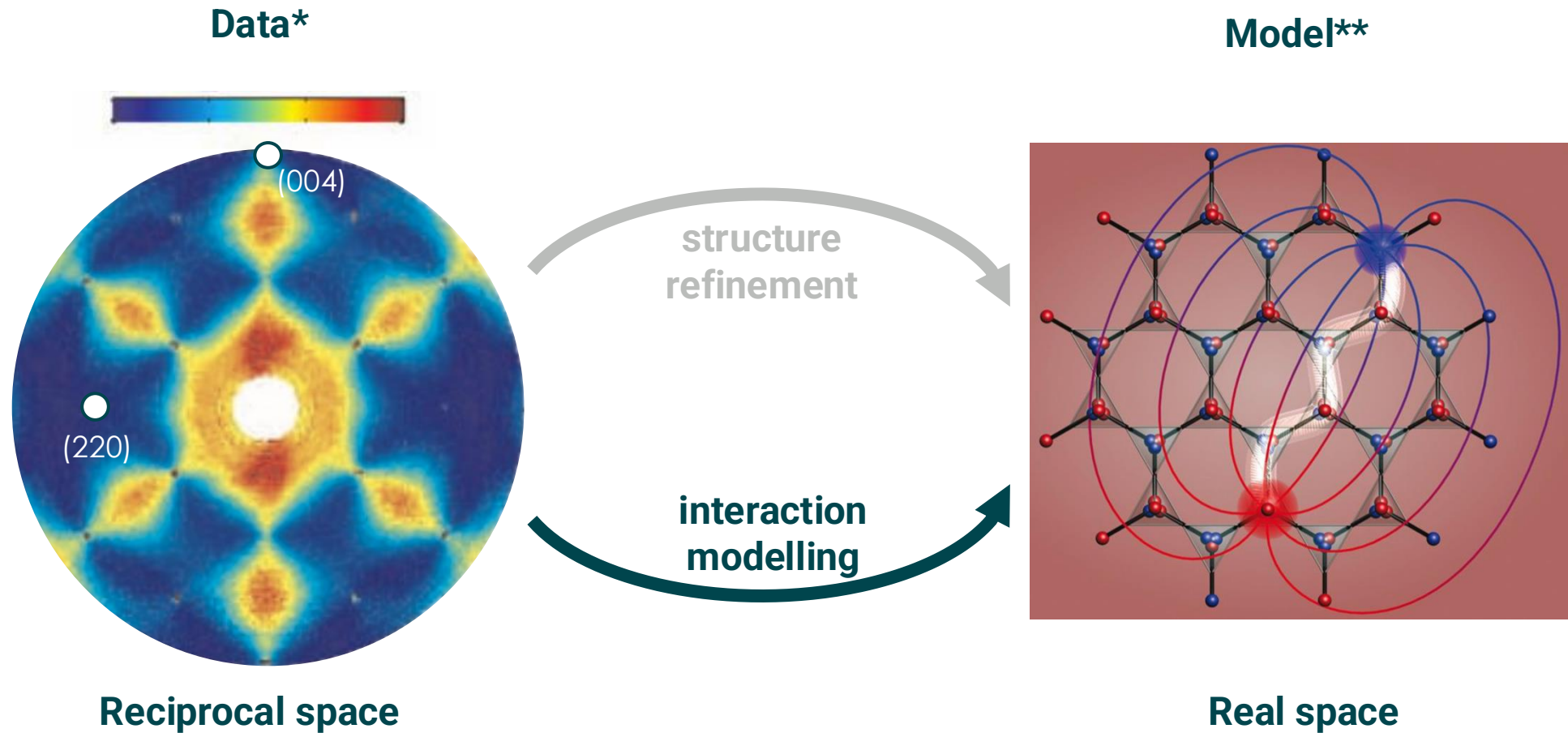


“Emergent charge” correlations



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

How can we analyse diffuse scattering data?



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

Castelnovo, Moessner & Sondhi, *Nature* **451, 42 (2008)

Interaction refinement

Define spin Hamiltonian and guess interaction values

$$H = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

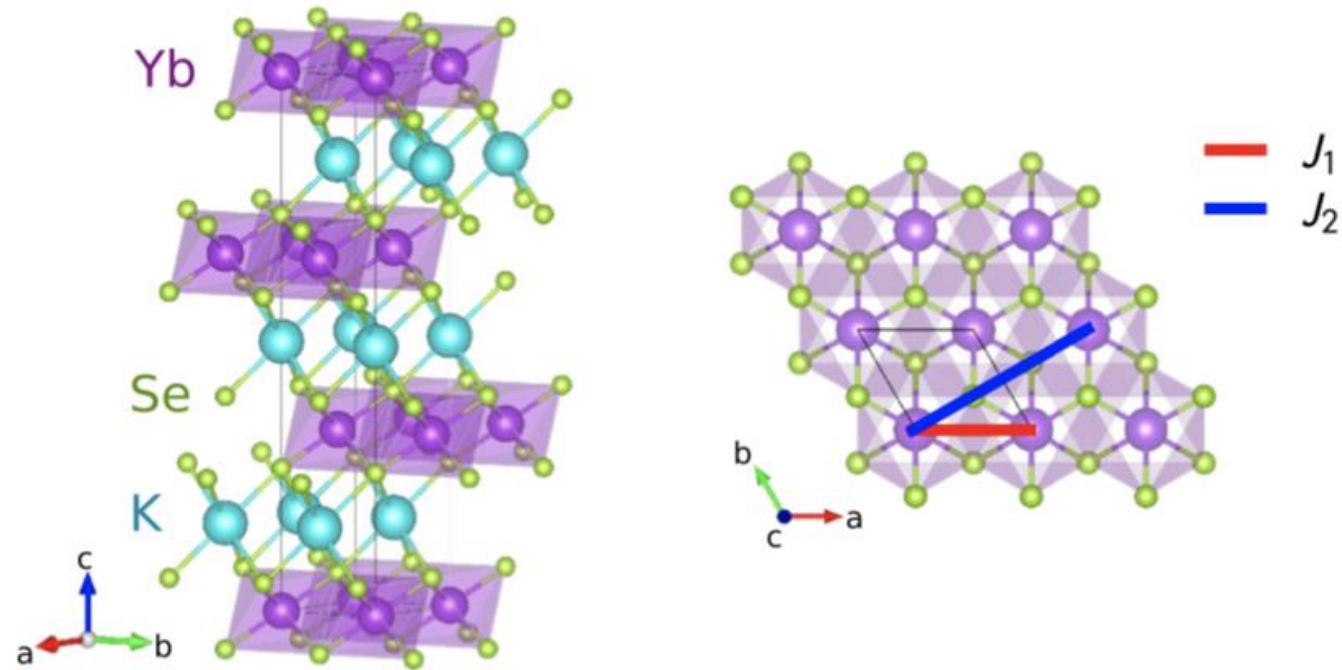
Calculate diffuse scattering *via* field theory

Send goodness-of-fit to least-squares optimiser

Receive new values of interactions from optimiser

Interaction refinement example 1: KYbSe₂

- Triangular lattice of Yb³⁺ with effective spin-1/2



Allen Scheie

ORNL/LANL

Alan Tennant

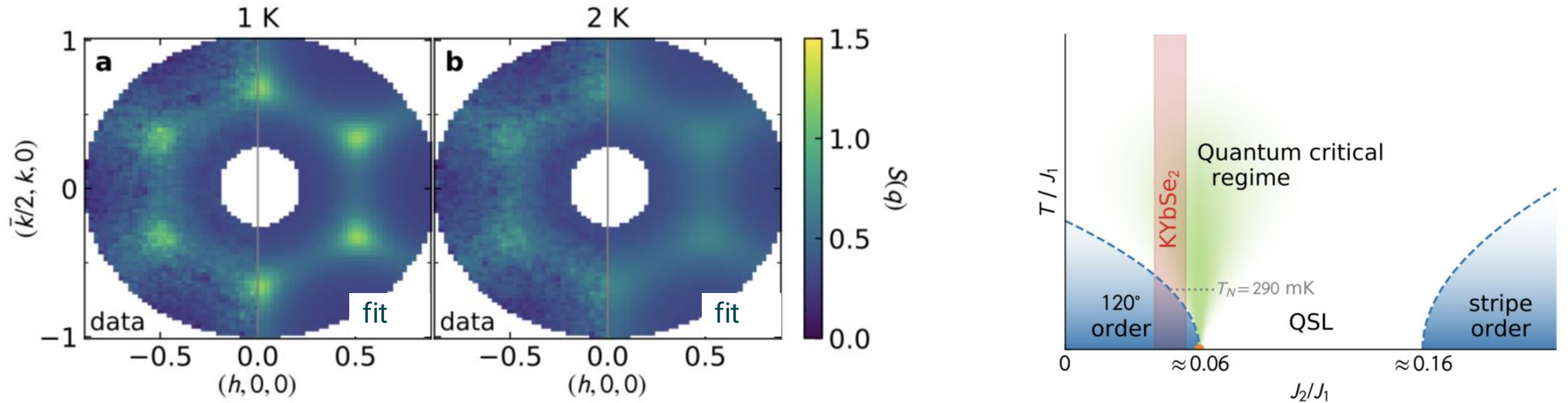
ORNL/UTK

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

Scheie *et al.*, *PRB* **109**, 014425 (2022)

Interaction refinement example 1: KYbSe₂

- Fits show <3% deviation from Heisenberg model

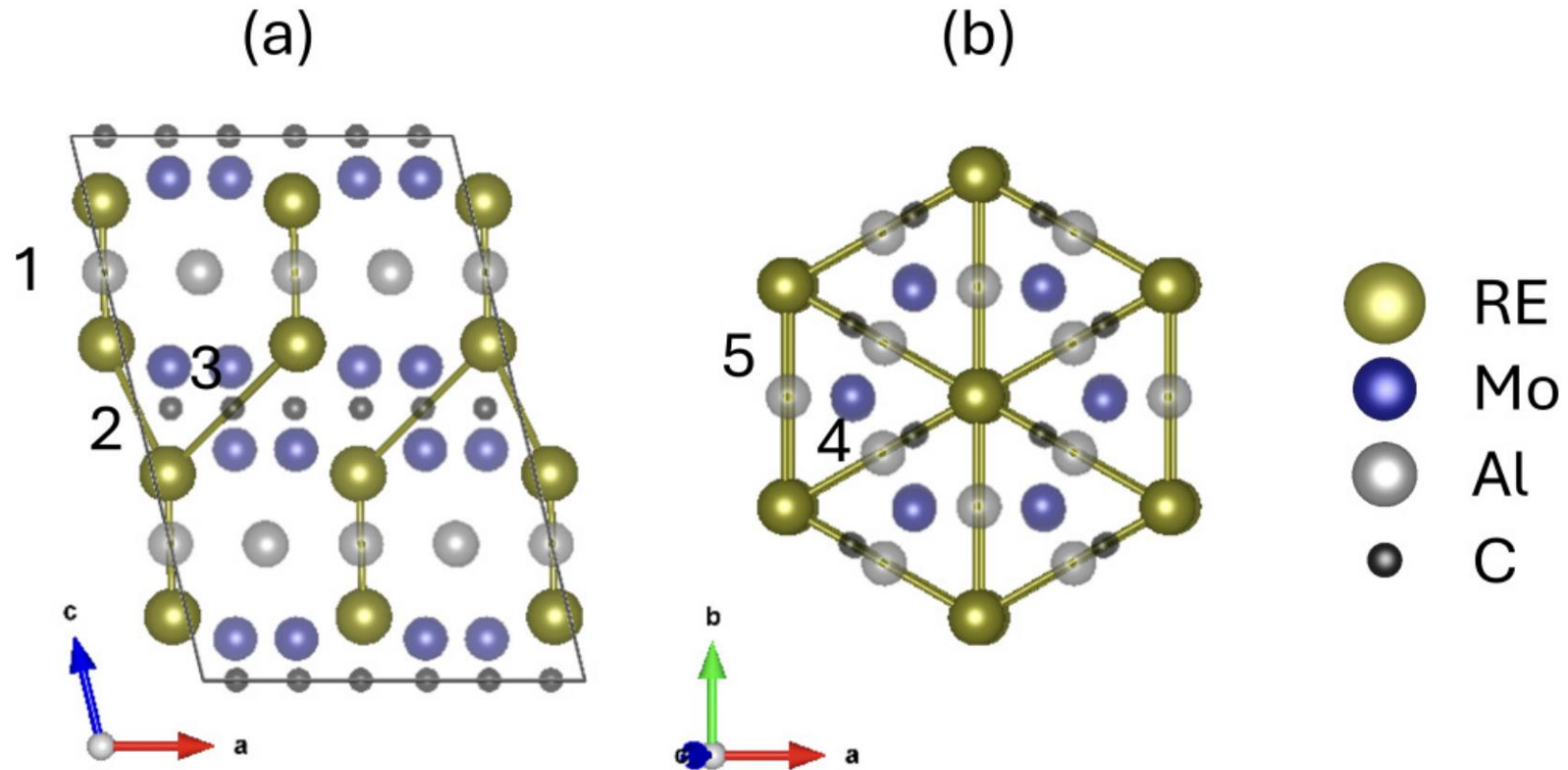


Theoretical technique	J_1 (meV)	J_2/J_1
Onsager reaction field	NA	0.047 ± 0.007
Nonlinear spin waves	0.456 ± 0.013	0.043 ± 0.010
Heat capacity	0.429 ± 0.010	0.037 ± 0.013
Weighted mean:	0.438 ± 0.008	0.044 ± 0.005

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

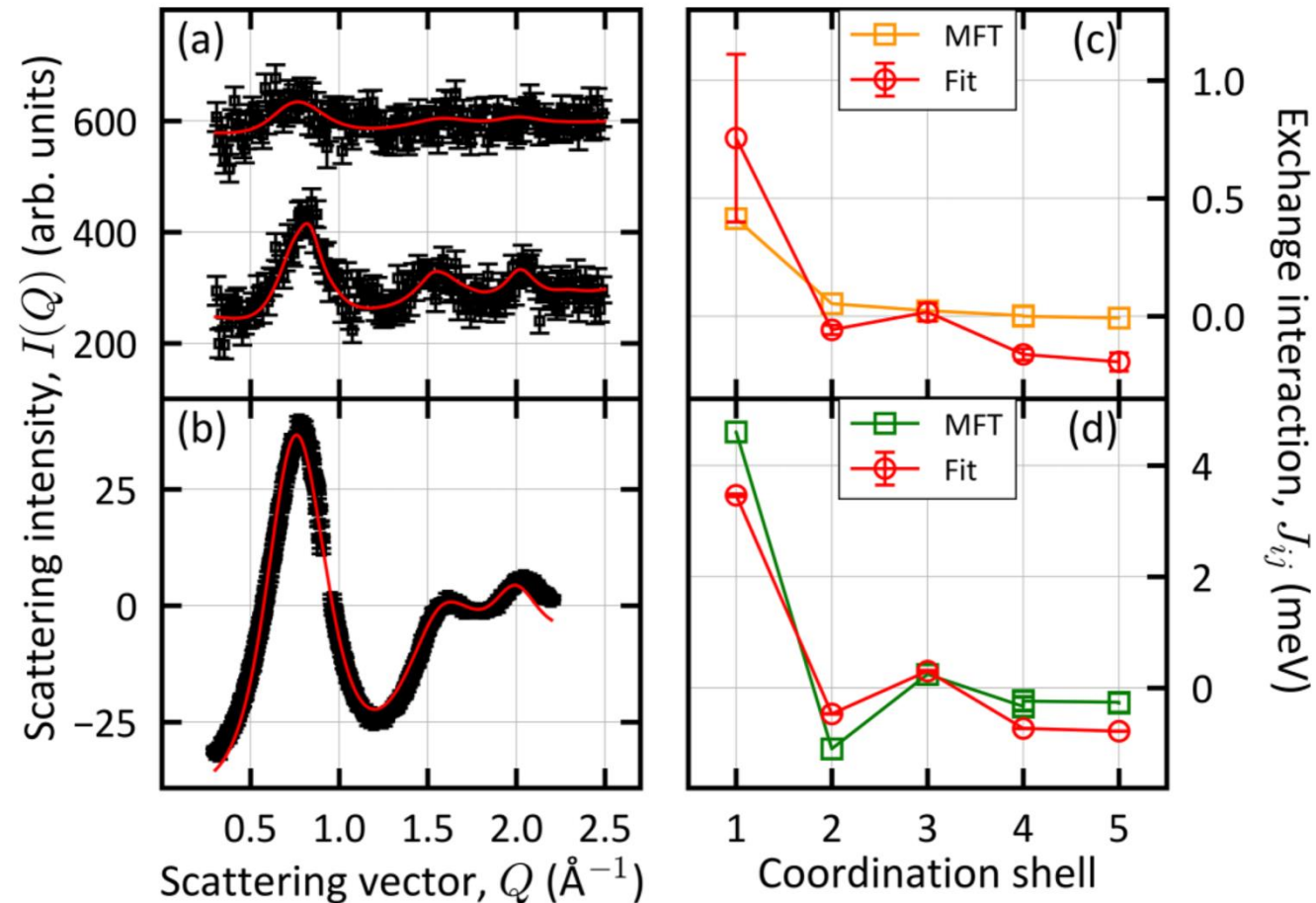
Interaction refinement example 2: $(\text{Mo}_{2/3}\text{RE}_{1/3})_2\text{AlC}$

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



Interaction refinement example 2: $(\text{Mo}_{2/3}\text{Ln}_{1/3})_2\text{AlC}$

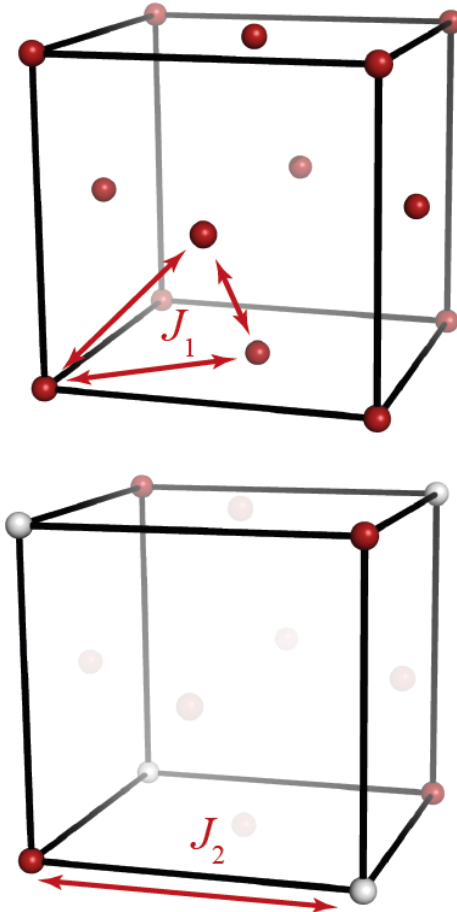
- Fits show good agreement with magnetic force theorem (DFT) simulations



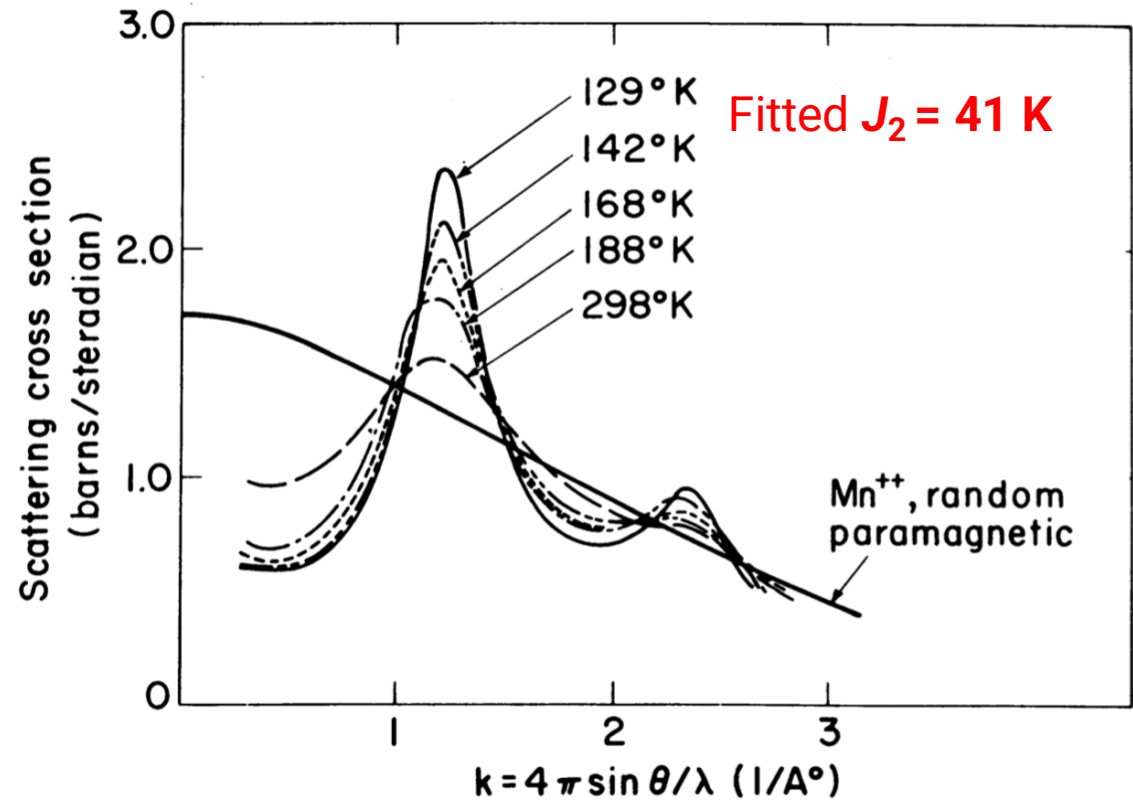
Interaction refinement example 3: MnO

- Face-centred cubic lattice of Mn^{2+}

($T_N = 120 \text{ K}$)



$$H = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$



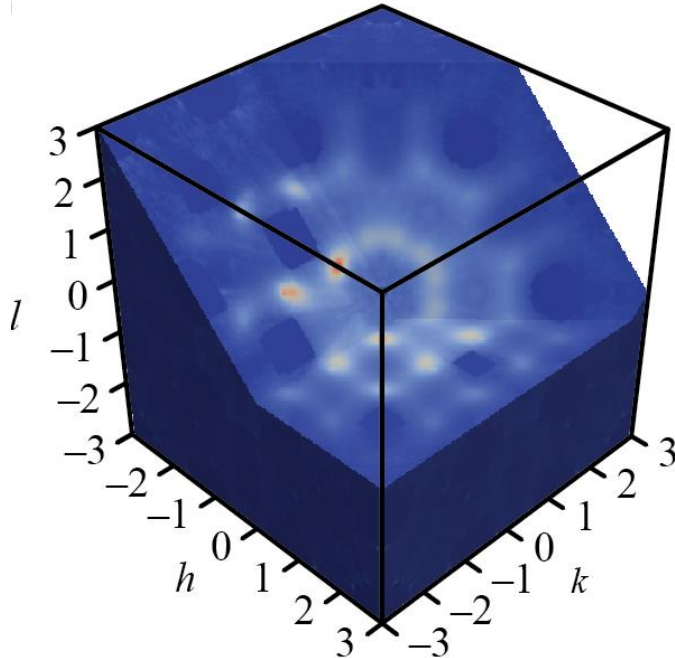
Blech & Averbach, *Physics* 1, 31 (1964)

Interaction refinement example 3: MnO

Andrew Goodwin
Oxford
Matthias Gutmann
ISIS

- Data collected at SXD (ISIS); fit Heisenberg J_1 - J_2

Data
 $T = 160$ K
Provided as list of h,k,l ,intensity,error



$\sim 3 \times 10^6$ data points

CPU time for full refinement: 70 to 200 s

```
MnO_config.txt — Edited
TITLE MnO
CELL 4.4344 4.4344 4.4344 90 90 90
CENTRING F
SITE 0.0 0.0 0.0

SPIN_DIMENSION 3
FORM_FACTOR_J0 0.4220 17.6840 0.5948 6.0050 0.0043 -0.6090 -0.0219

XTAL_SCALE refine
XTAL_FLAT_BACKGROUND refine
XTAL_TEMPERATURE 160.0

BZ_POINTS 32 32 32
```

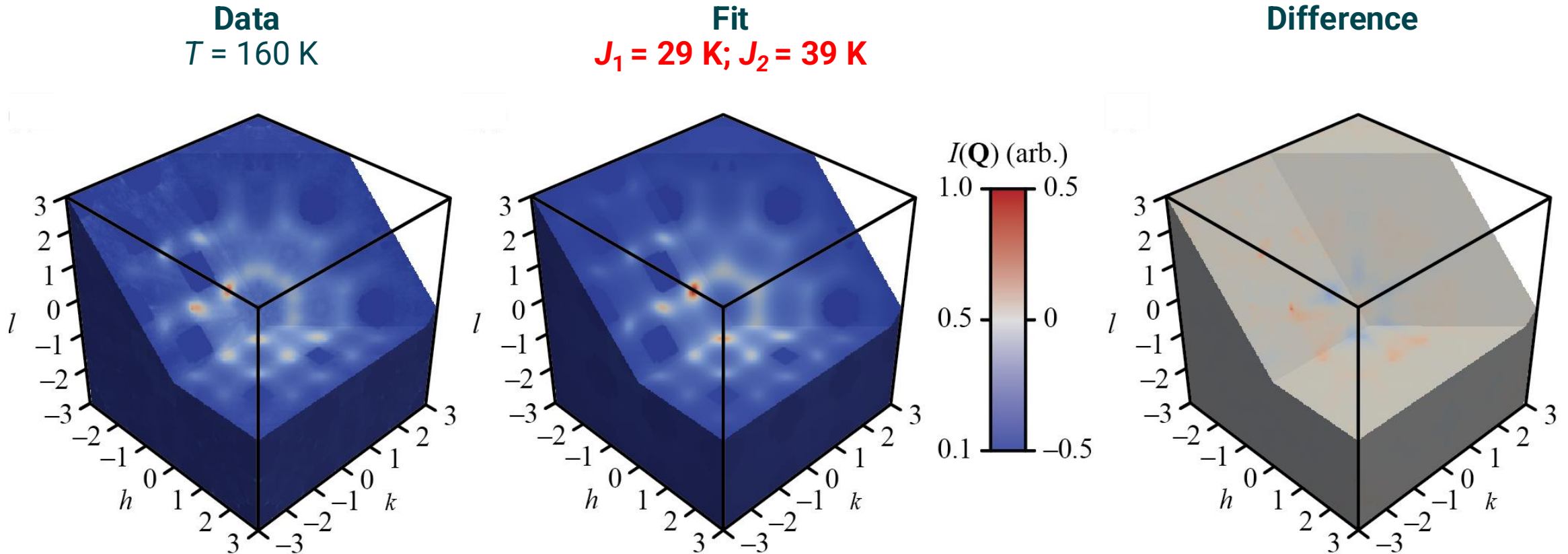
www.ill.eu/sites/ccsl/ffacts



Interaction refinement example 3: MnO

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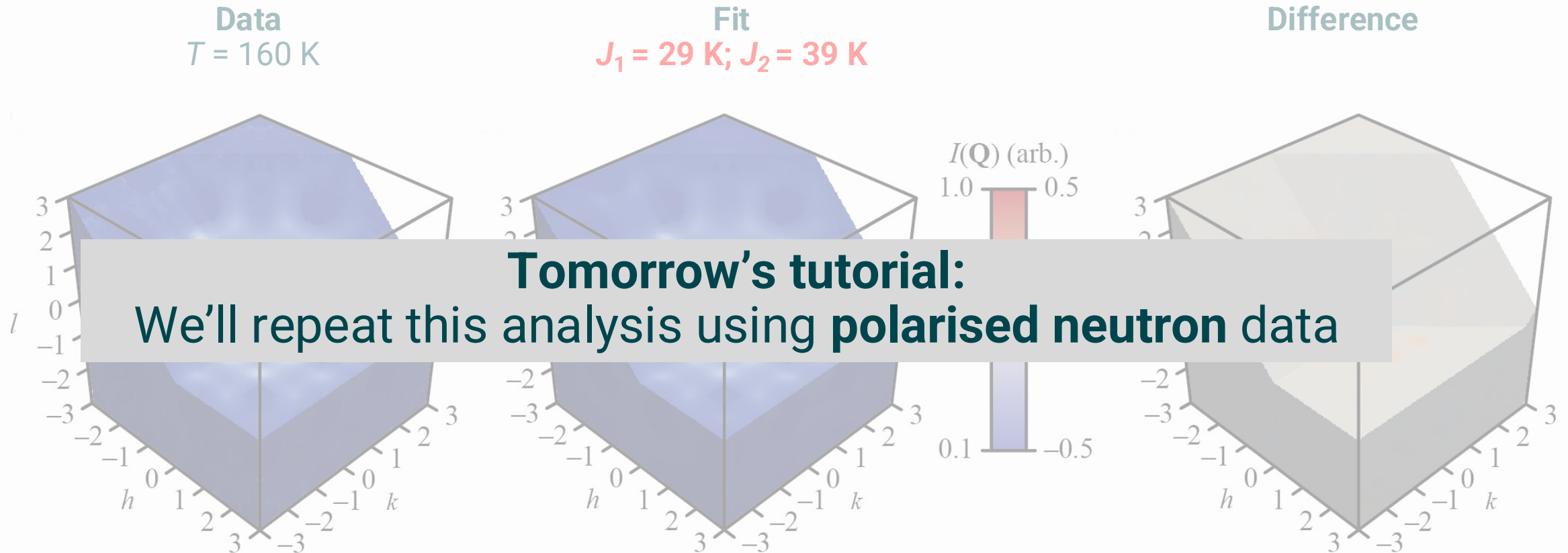
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Interaction refinement example 3: MnO

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CPU time for full refinement: 70 to 200 s

Paddison, *J Phys.: Condensed Matter* **35**, 495802 (2023)

Data: Paddison, Gutmann, Stewart et al., *PRB* **97**, 014429 (2018)



Thanks for listening!

- Diffuse scattering arises from **correlated disorder**
 - Disordered species can be spins, atoms, molecules, or collective objects
- Instruments at SNS and HFIR can measure diffuse scattering accurately
 - Powder and single-crystal data are both information-rich
- We can combine real space, reciprocal space, and interaction space analysis to solve the inverse scattering problem
- Polarised neutrons can isolate magnetic contributions to the signal

- **Don't hesitate to contact instrument scientists or developers!**
- **Email:** paddisonja@ornl.gov

Tutorial: Getting started

- **Option 1: Use your own Windows computer**

- Download the tutorial by scanning QR code
- Or web: www.joepaddison.com/software (top link)
- Do the tutorial! See [instructions.pdf](#)

- **Option 2: Use the analysis cluster**

- Log on to <http://analysis.sns.gov>
- In file manager, go to folder **HFIR/HB2A/shared**
- Copy **2026_magnetic_diffuse_tutorial** to your Documents folder on the cluster
- Do the tutorial! See [instructions.pdf](#)



email: paddisonja@ornl.gov