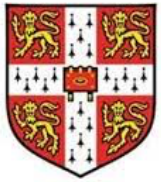


UNIVERSITY OF CAMBRIDGE

Spinvert: Magnetic structure refinement for correlated paramagnets

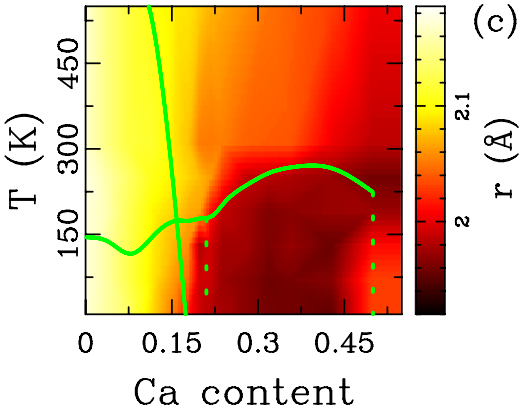
Joe Paddison
University of Cambidge

ADD2019 Grenoble
March 2019

Why magnetic disorder?

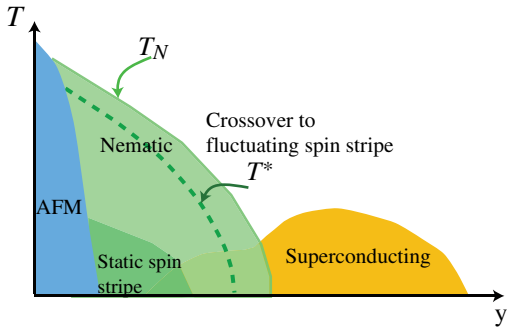
- ❑ Relevant to important magnetic materials
- ❑ Can generate new & exotic magnetic states

Colossal magnetoresistance



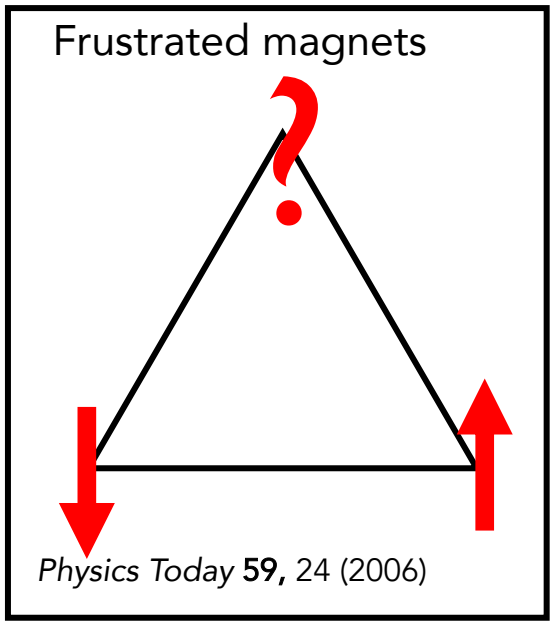
PRL **98**, 137203 (2007)

High-T_c superconductors



Rev. Mod. Phys. **75**, 1201 (2003)

Frustrated magnets

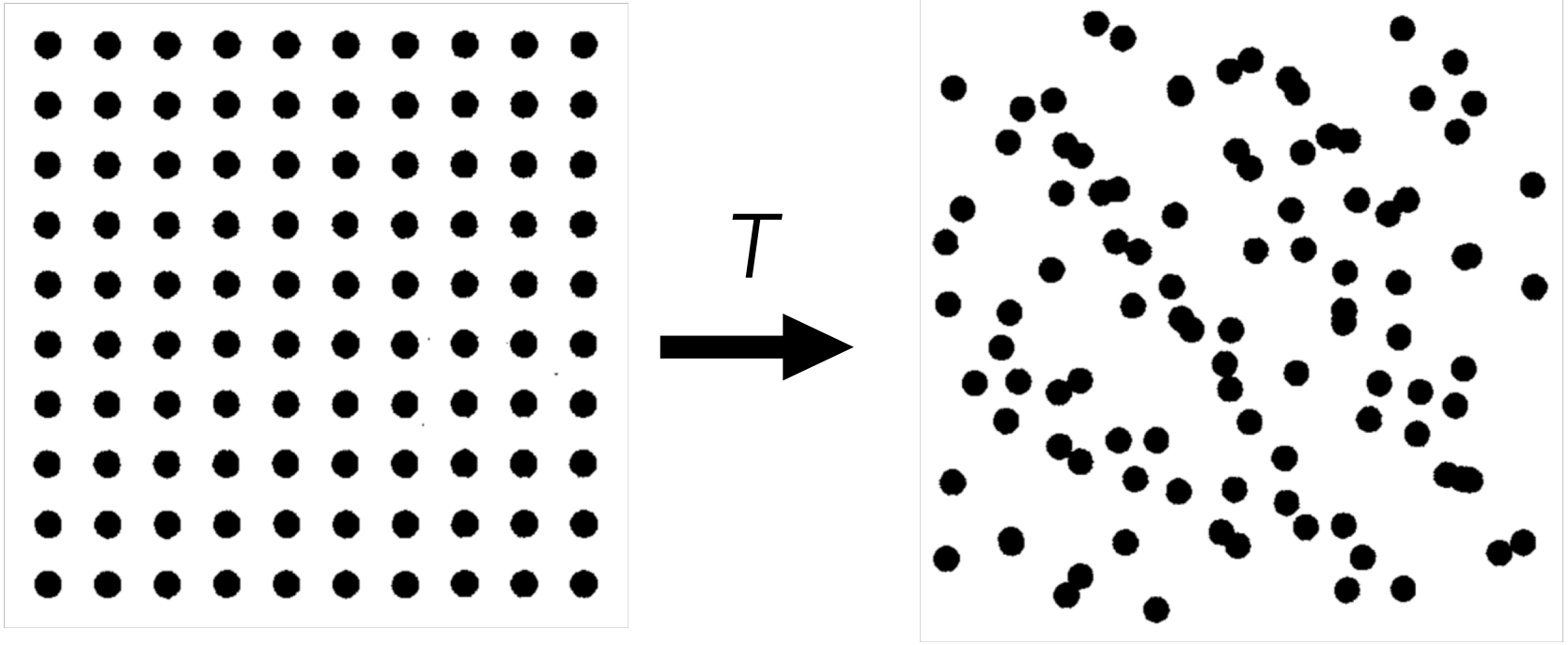


Physics Today **59,** 24 (2006)

Plan

- Introduction
- Magnetic neutron scattering
- Reverse Monte Carlo & Spinvert
- Powder case study: $Gd_3Ga_5O_{12}$
- Single-crystal refinements

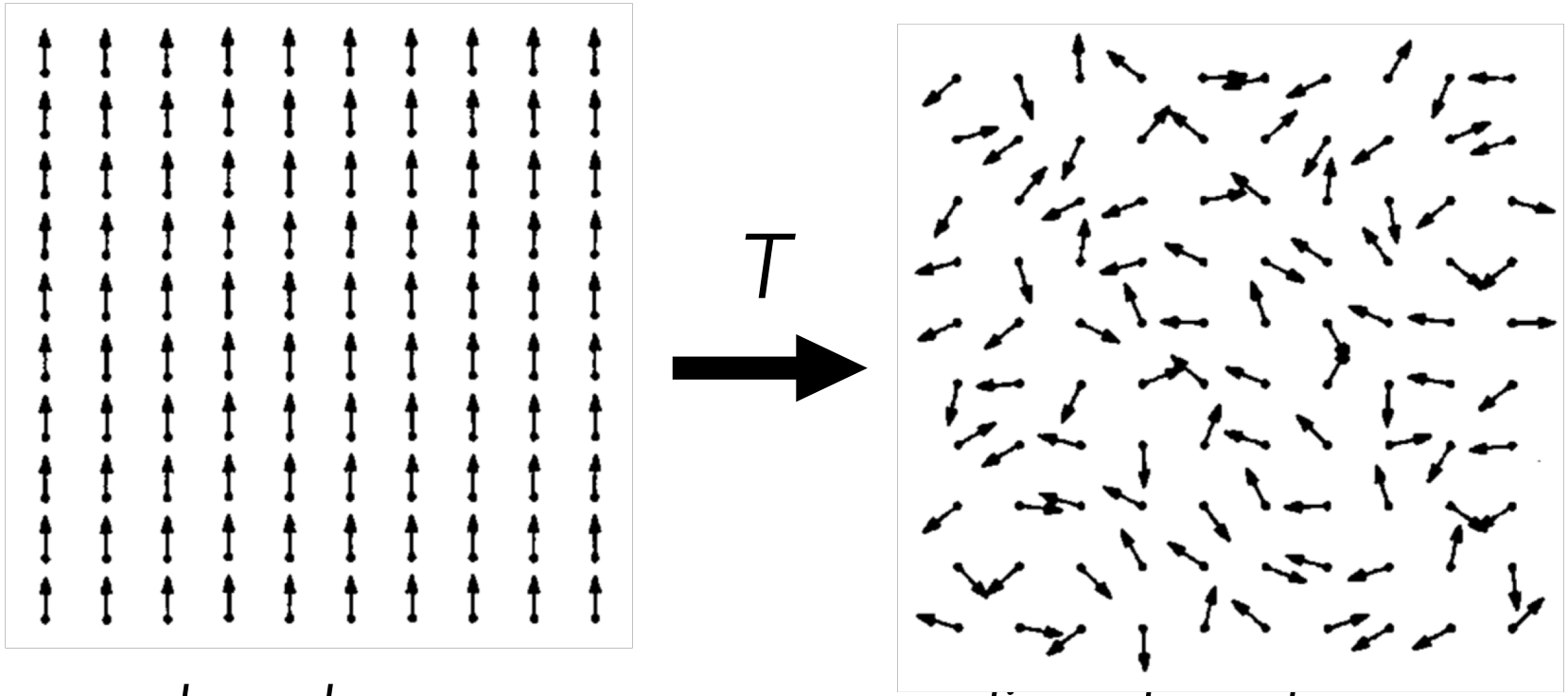
Disorder ≠ randomness



ordered

disordered

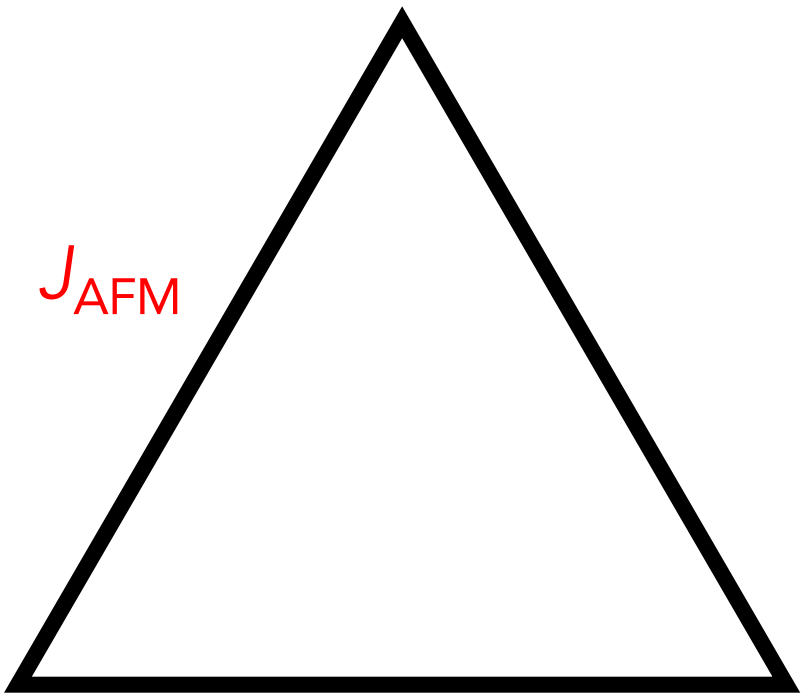
Disorder ≠ randomness



ordered

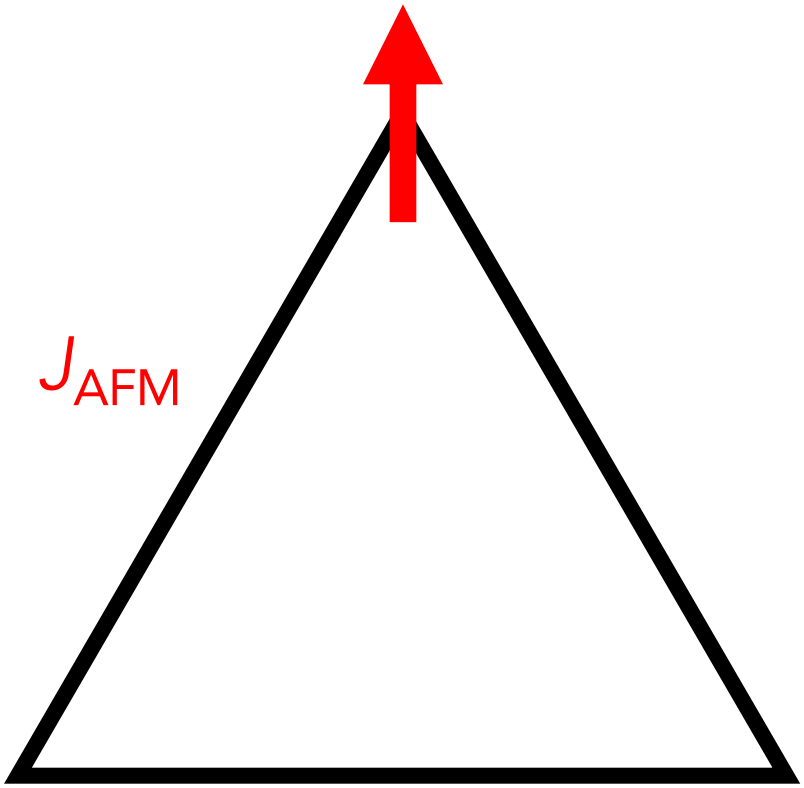
disordered

Magnetic frustration



Wannier, *Phys Rev* **79**, 357 (1950)

Magnetic frustration



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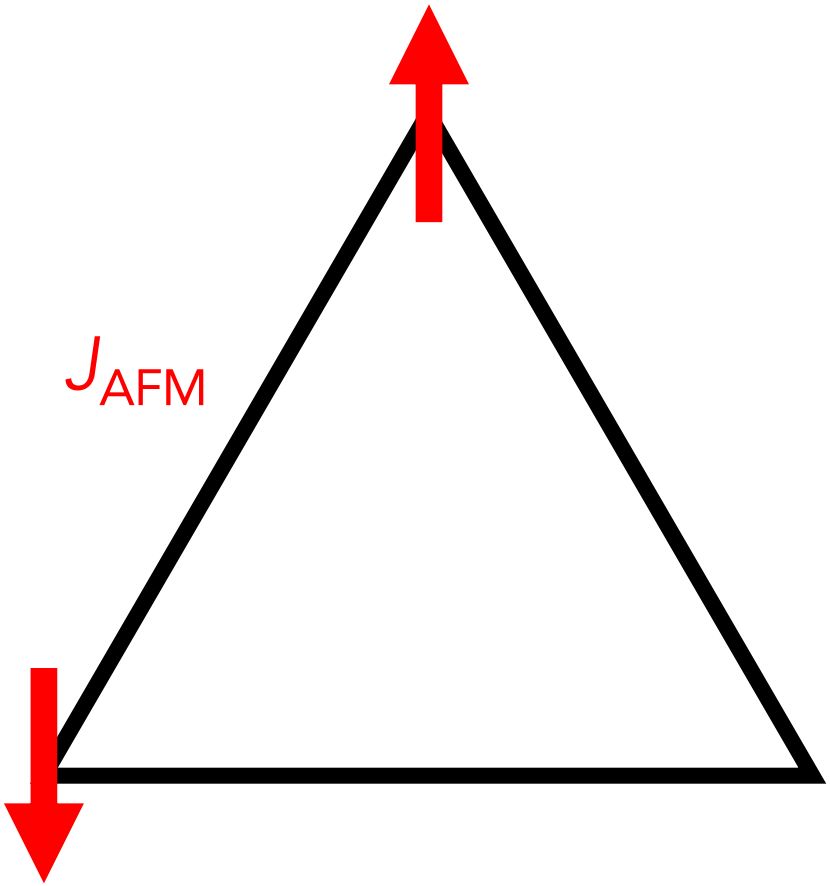

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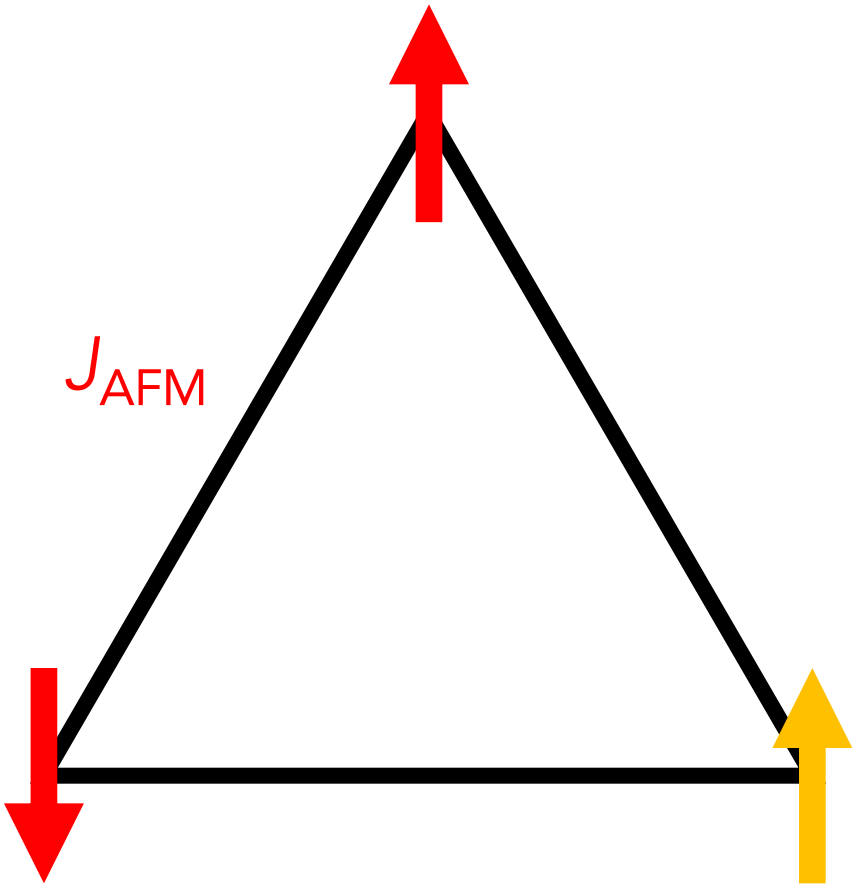
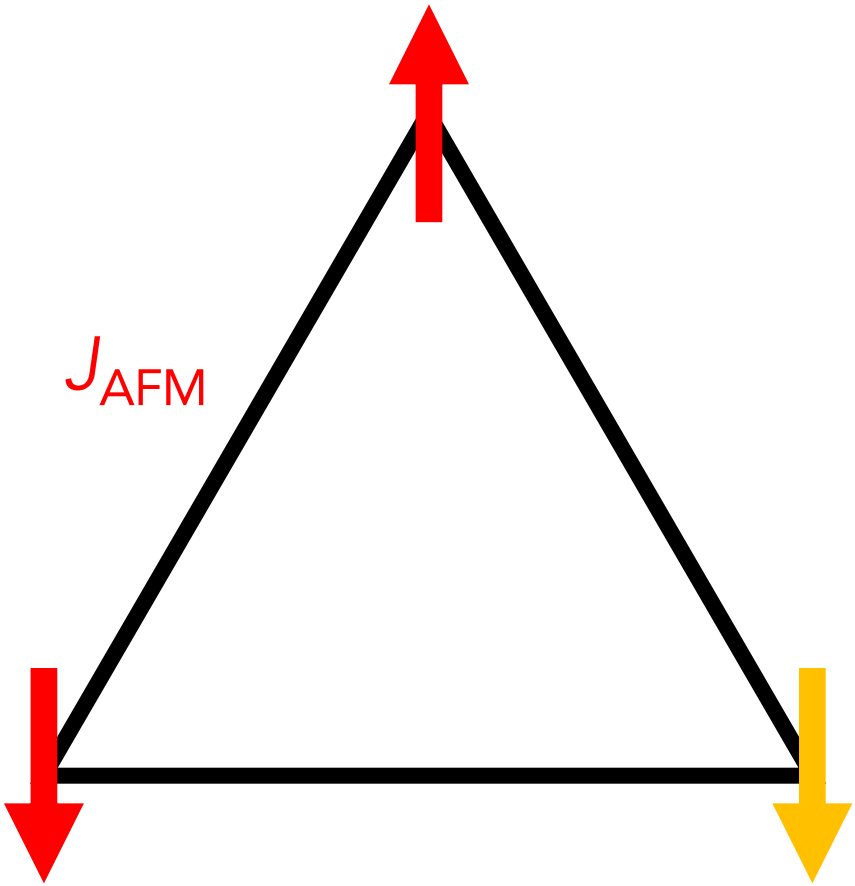


(geometrical) frustration

Wannier, *Phys Rev* **79**, 357 (1950)

Magnetic frustration



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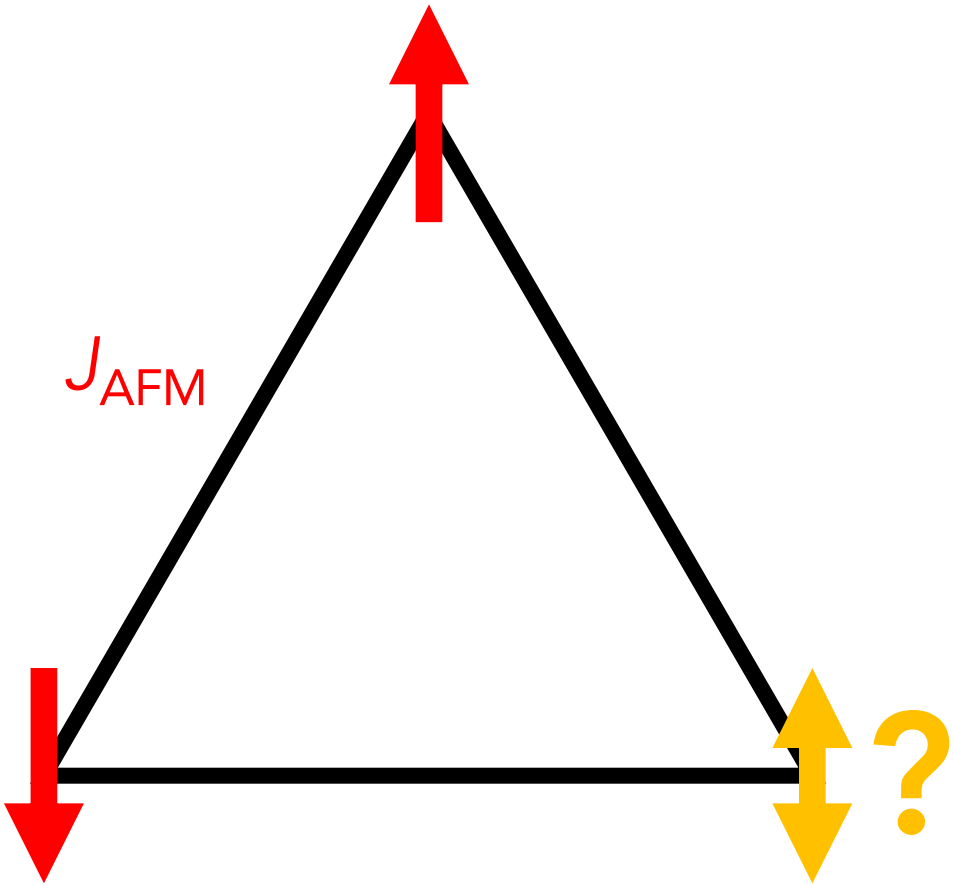
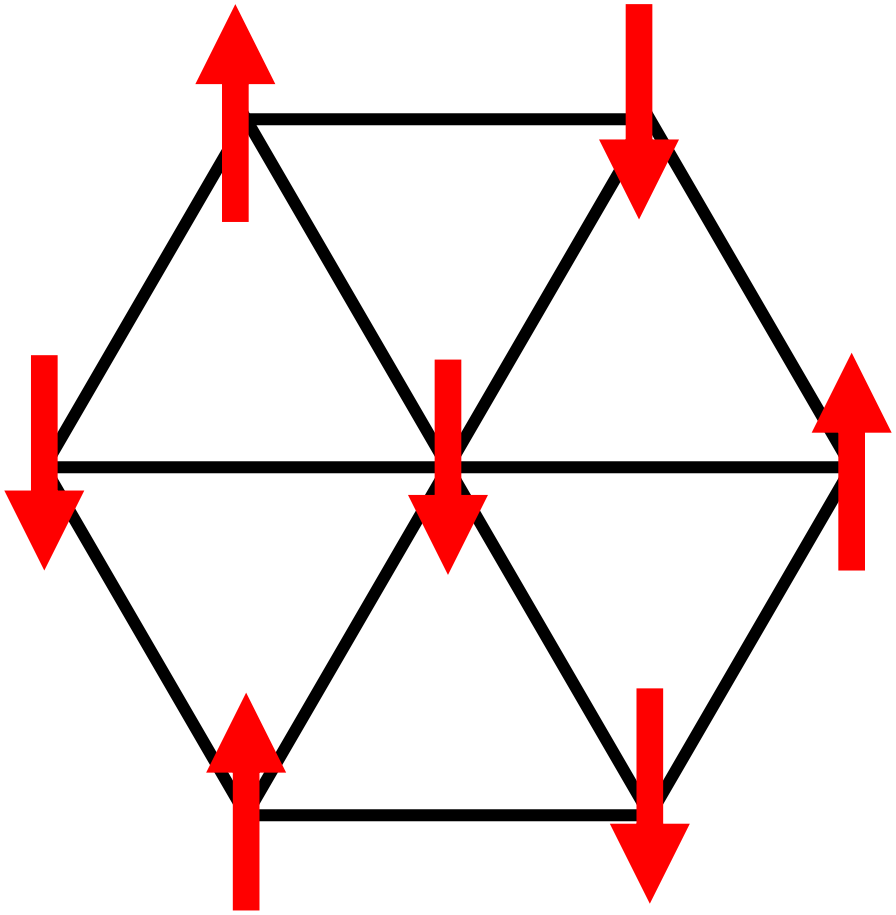


frustration → degeneracy

Wannier, *Phys Rev* **79**, 357 (1950)

Magnetic frustration



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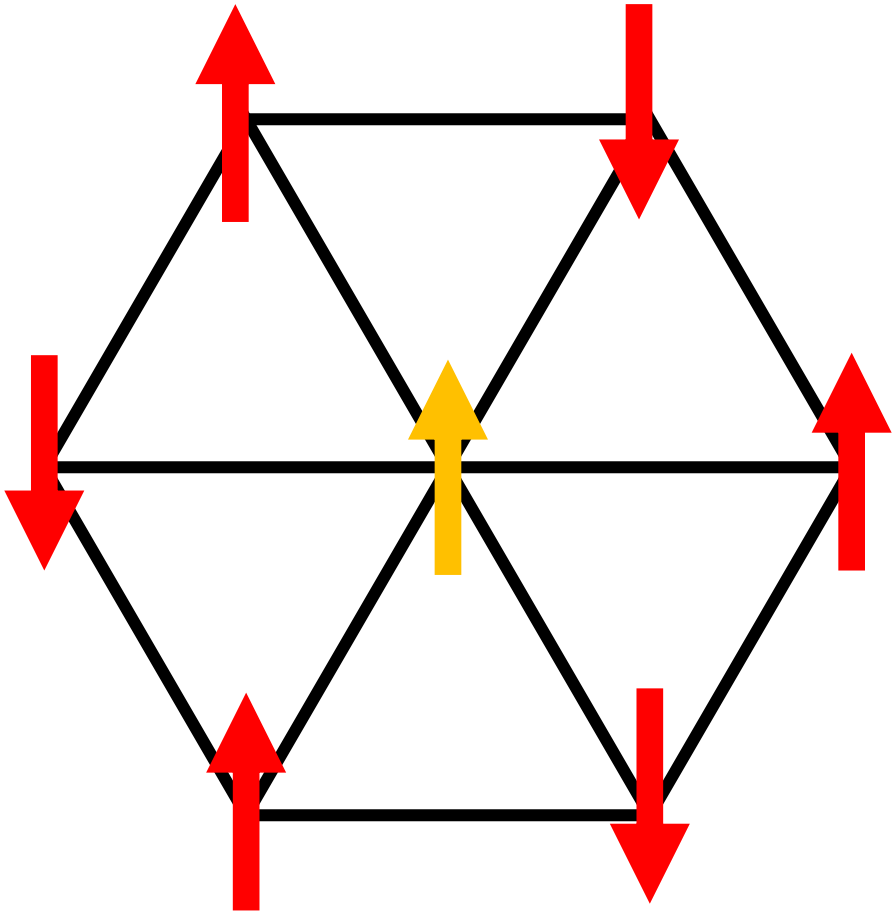

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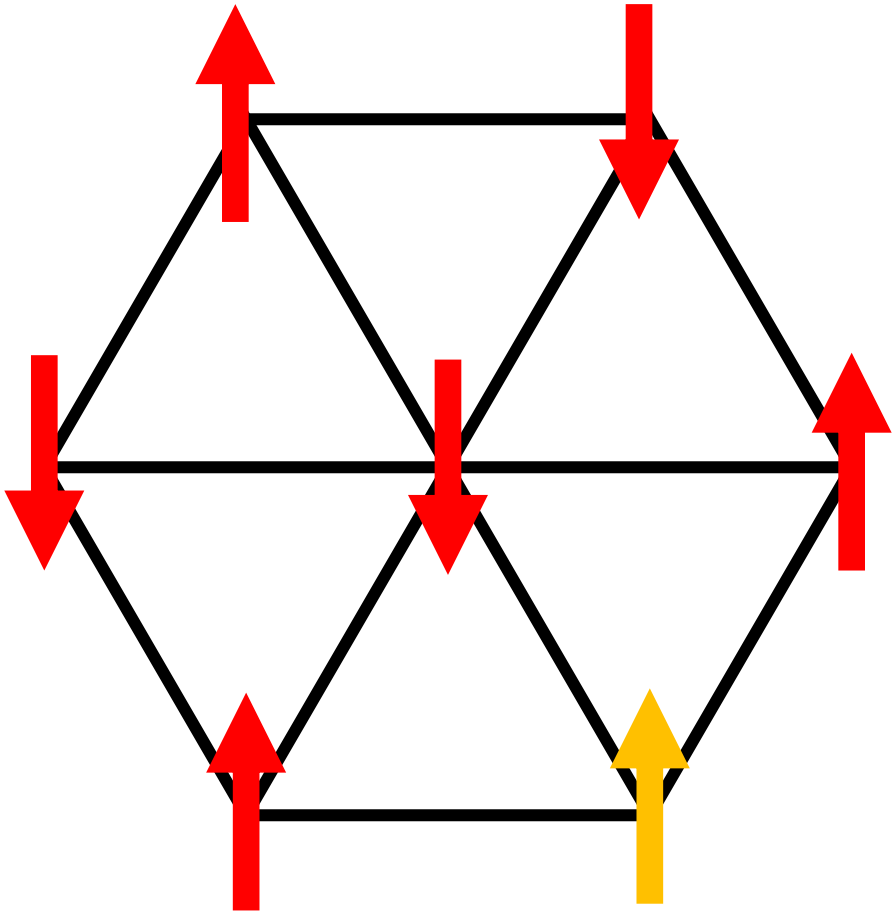

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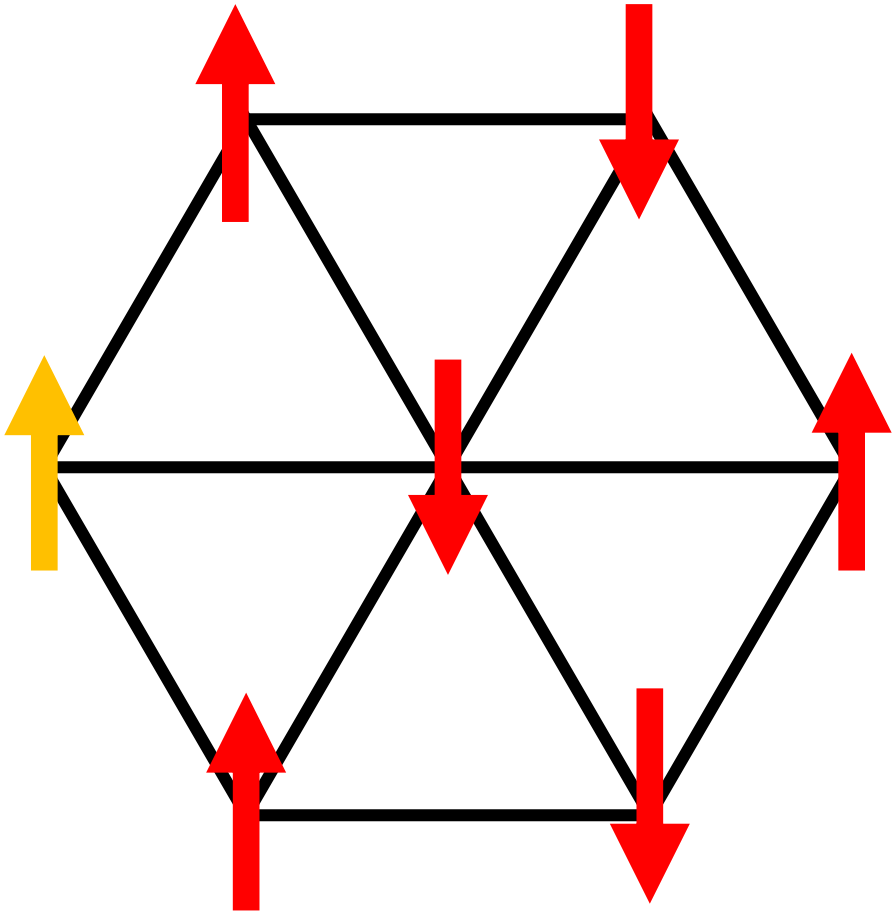

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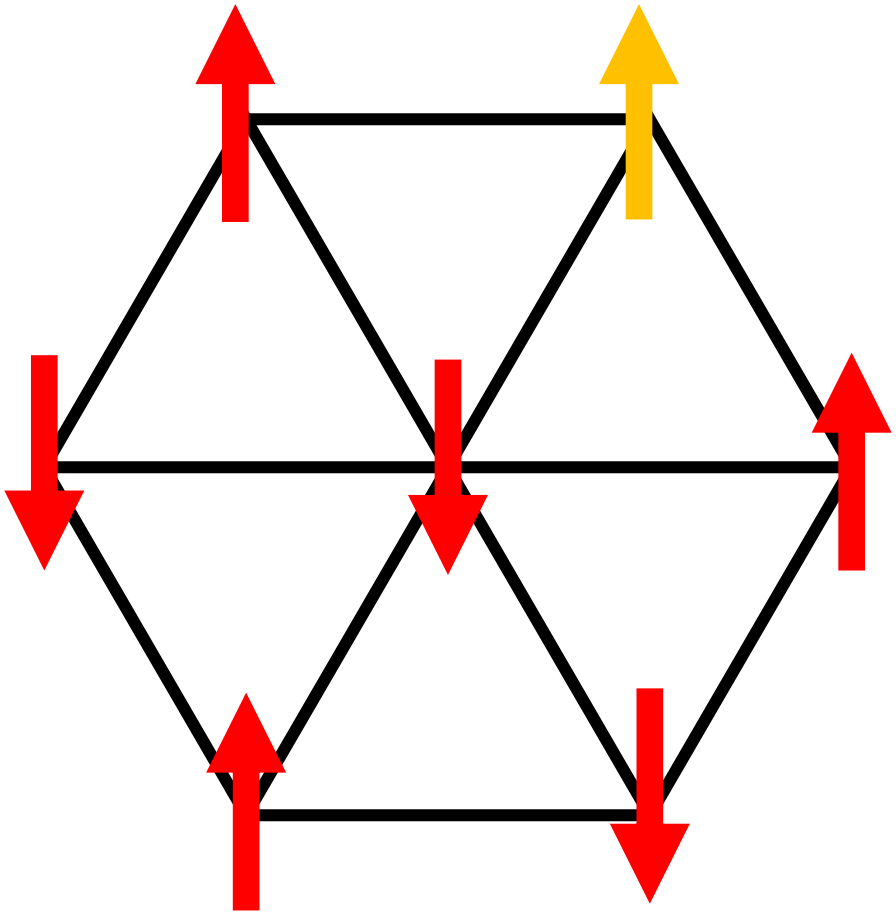

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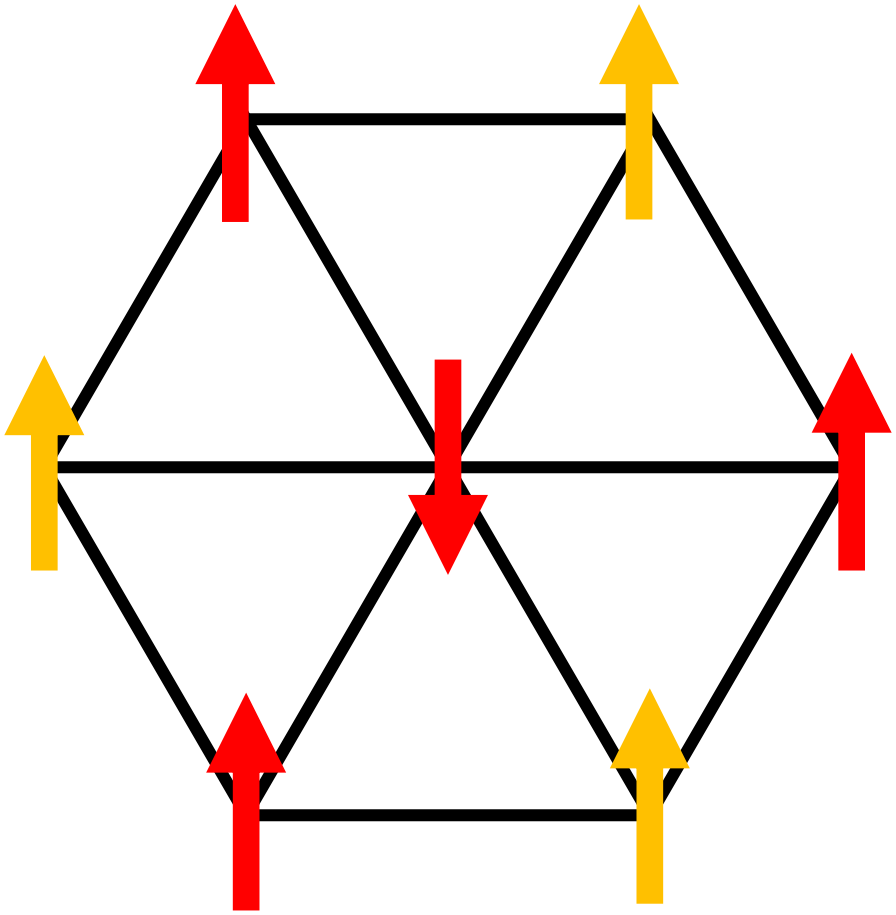

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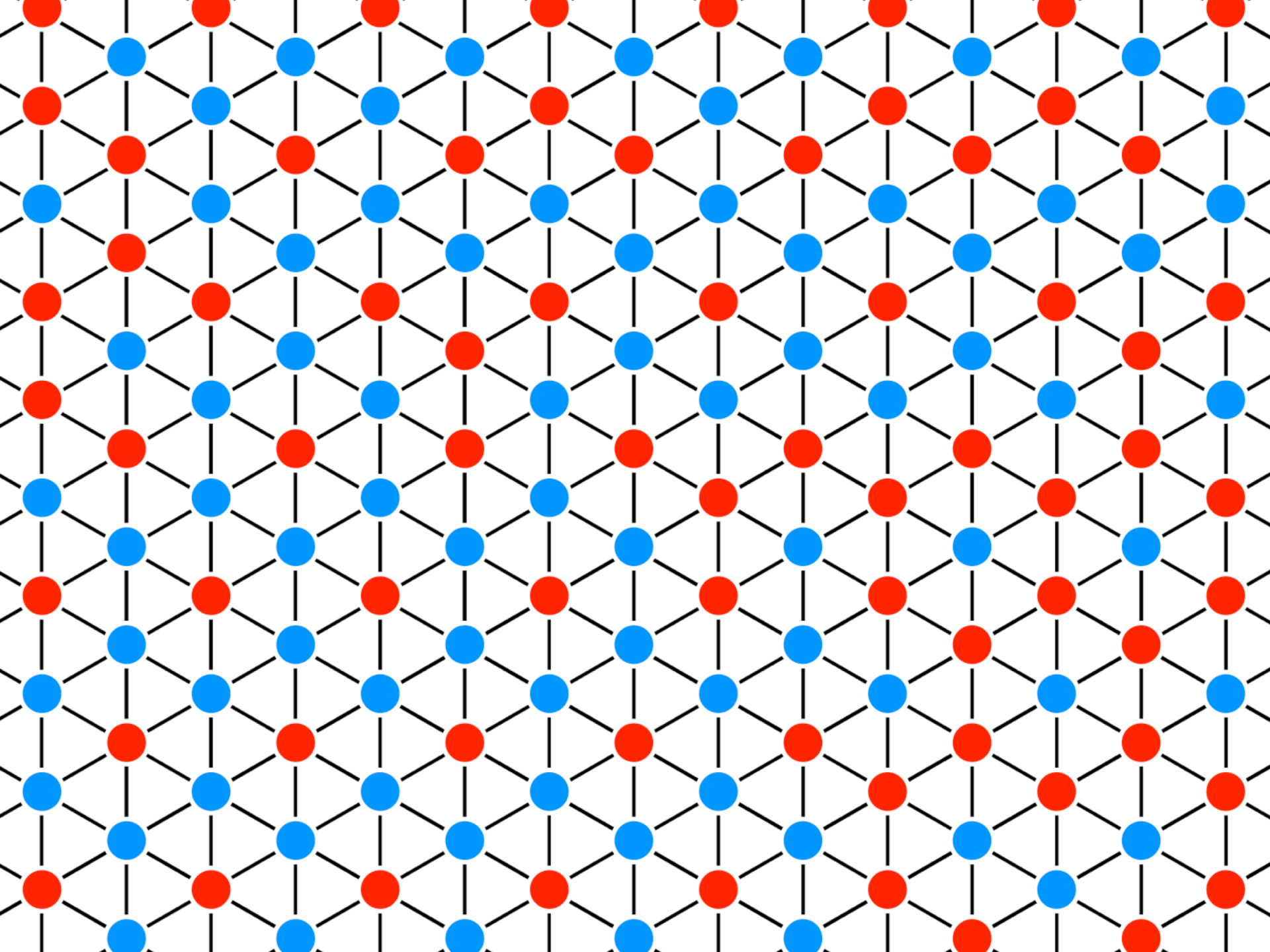
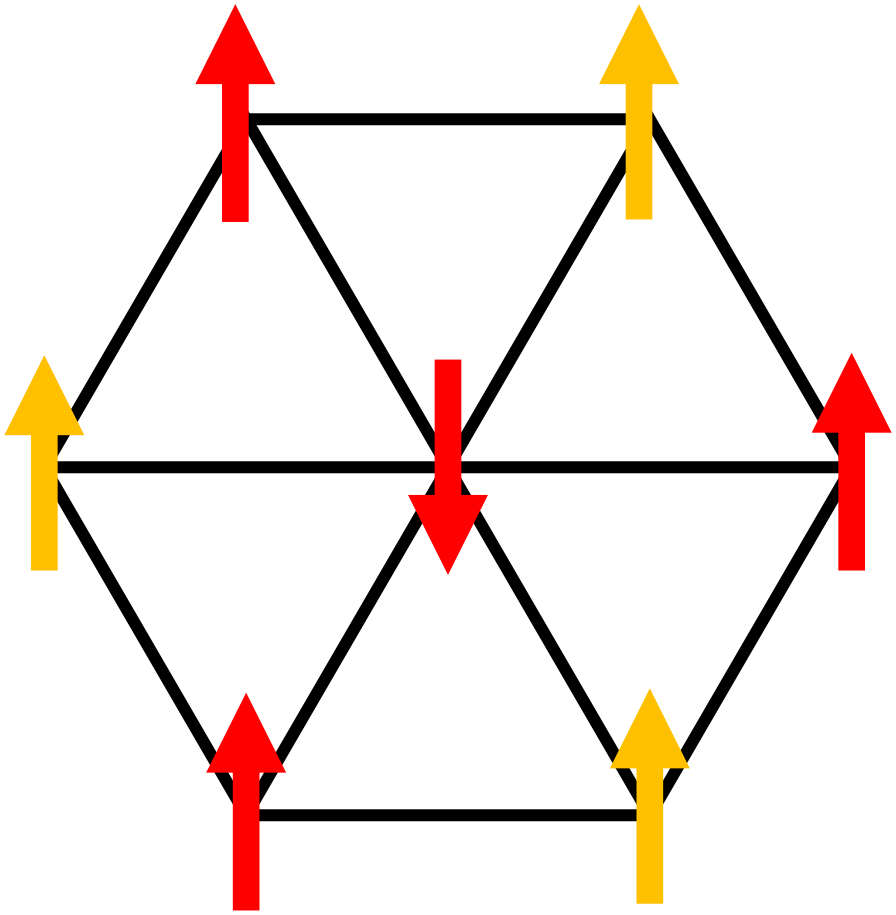


Magnetic frustration

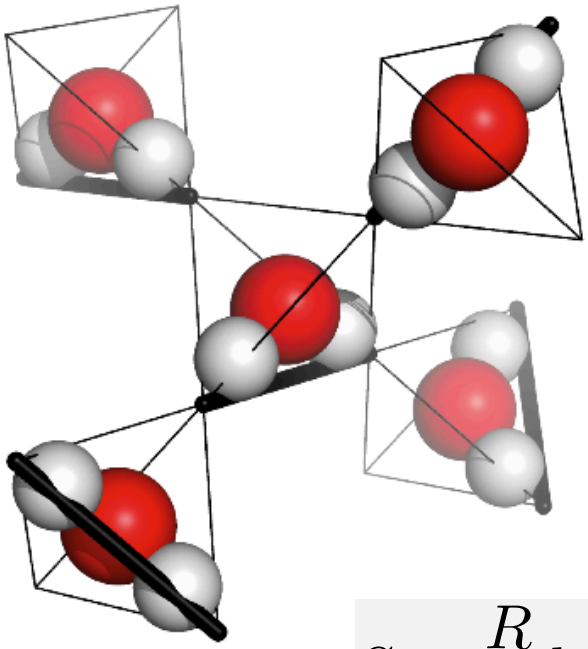


frustration → *degeneracy* → **correlated** *disorder*

Wannier, *Phys Rev* **79**, 357 (1950)

"Ice rules"

(cubic) water ice



$$S = \frac{R}{2} \ln\left(\frac{3}{2}\right)$$

frustration → *degeneracy* → **correlated** *disorder*

Pauling, *J. Am. Chem. Soc* **57**, 2680 (1935)

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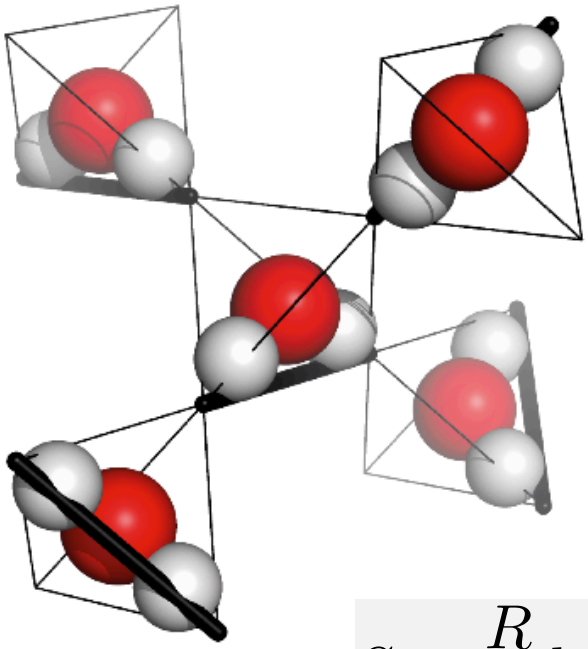
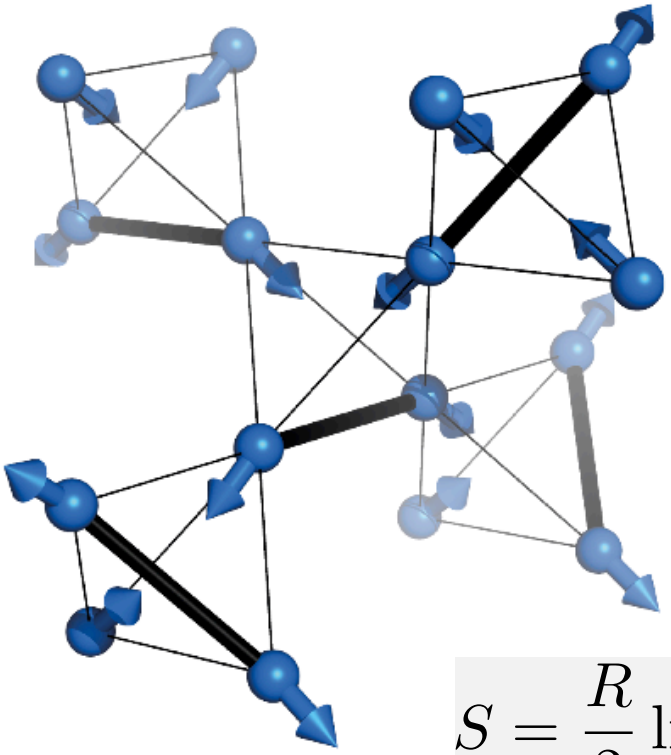


$$S = \frac{R}{2} \ln\left(\frac{3}{2}\right)$$

spin ice



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Bramwell & Harris, *PRL* **79**, 2554 (1997)
Pauling, *J. Am. Chem. Soc* **57**, 2680 (1935)

Plan

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- Powder case study: $Gd_3Ga_5O_{12}$
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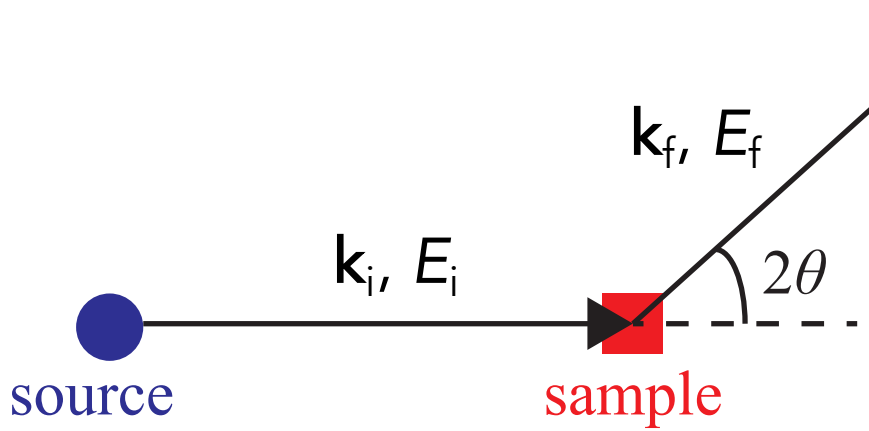
Total scattering



Energy transfer

$E = E_i - E_f$

Wavevector transfer

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

- **Total scattering:** integral over energy transfer

$$I(\mathbf{Q}) = \int_{-\infty}^{\infty} I(\mathbf{Q}, E)\,\mathrm{d}E$$

- Measures **instantaneous** correlations
- **Quasi-static approximation:** $\int \mathrm{d}E \approx \int \mathrm{d}E_f$ if $E << E_i$

diffraction (E_f not analysed)

- **Total scattering = Bragg + diffuse**

Nuclear neutron scattering – Equations

- Assume elemental system (for simplicity)

  - **Single crystal**

$$I_{\mathrm{n}}(\mathbf{Q}) = b^2 \left(1 + \frac{1}{N} \sum_{i,j \neq i} \langle \exp(\mathrm{i}\mathbf{Q} \cdot \mathbf{r}_{ij}) \rangle \right)$$

b = coherent scattering length
N = number of atoms
$\mathbf{r}_{ij}$ = vector separation
$\langle\ \rangle$ = thermal average

Squires, *Introduction to Thermal Neutron Scattering*, CUP (1978)
Debye, *Ann. Phys. (Berlin)*, **351**, 809 (1915)

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spherical average

- **Powder**

$$I_{\mathrm{n}}(Q) = b^2 \left(1 + \frac{1}{N} \sum_{i,j \neq i} \left\langle \frac{\sin Q r_{ij}}{Q r_{ij}} \right\rangle \right)$$

single-atom scattering (ideal gas)

scattering from atom-pair correlations

b = coherent scattering length
N = number of atoms
$\mathbf{r}_{ij}$ = vector separation
$\langle\ \rangle$ = thermal average

Debye formula
r_{ij} = radial separation

Squires, *Introduction to Thermal Neutron Scattering*, CUP (1978)
Debye, *Ann. Phys. (Berlin)*, **351**, 809 (1915)

Magnetic neutron scattering – Equations

- Assume magnetic ions of a single type, with fixed locations

  - **Single crystal**

$$I_{\mathrm{m}}(\mathbf{Q}) = C\,[gf(Q)]^2 \left[\frac{2}{3}S(S+1) + \frac{1}{N}\sum_{i,j\neq i}\left\langle \mathbf{S}_i^{\perp}\cdot\mathbf{S}_j^{\perp}\right\rangle \exp(\mathrm{i}\mathbf{Q}\cdot\mathbf{r}_{ij})\right]$$

C = 0.07265 barn
$f(Q)$ = form factor

$\mathbf{S}^{\perp} = \mathbf{S} - (\mathbf{S}\cdot\hat{\mathbf{Q}})\hat{\mathbf{Q}}$ component of spin **S** perpendicular to **Q**

Lovesey, *Theory of Neutron Scattering from Condensed Matter*, OUP (1987)

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➢ **Powder**

$$I_{\mathrm{m}}(Q) = C\,[gf(Q)]^2 \left\{\frac{2}{3}S(S+1) + \frac{1}{N}\sum_{i,j\neq i}\left[\langle A_{ij}\rangle \frac{\sin Qr_{ij}}{Qr_{ij}} + \langle B_{ij}\rangle\left(\frac{\sin Qr_{ij}}{(Qr_{ij})^3} - \frac{\cos Qr_{ij}}{(Qr_{ij})^2}\right)\right]\right\}$$

single-spin scattering (ideal paramagnet)

scattering from spin-pair correlations

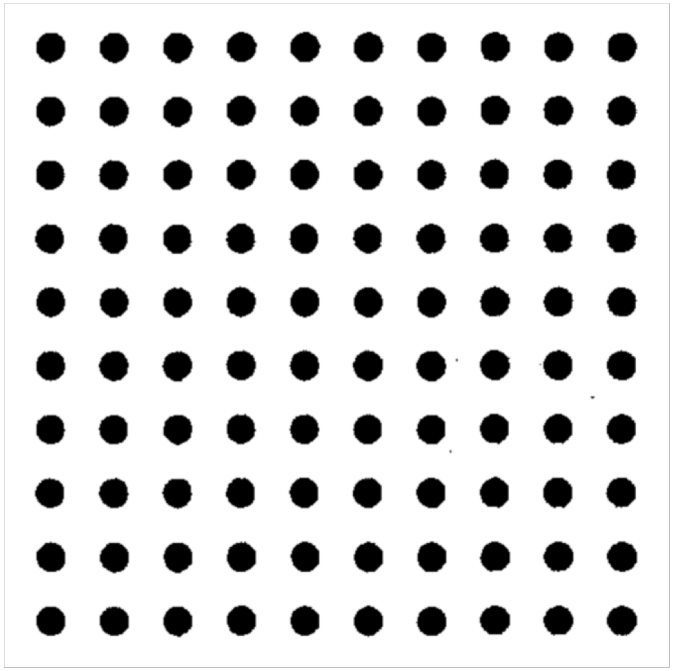
$$A_{ij} = \mathbf{S}_i\cdot\mathbf{S}_j - (\mathbf{S}_i\cdot\hat{\mathbf{r}}_{ij})(\mathbf{S}_j\cdot\hat{\mathbf{r}}_{ij})$$
$$B_{ij} = 3(\mathbf{S}_i\cdot\hat{\mathbf{r}}_{ij})(\mathbf{S}_j\cdot\hat{\mathbf{r}}_{ij}) - \mathbf{S}_i\cdot\mathbf{S}_j$$

spin correlation coefficients

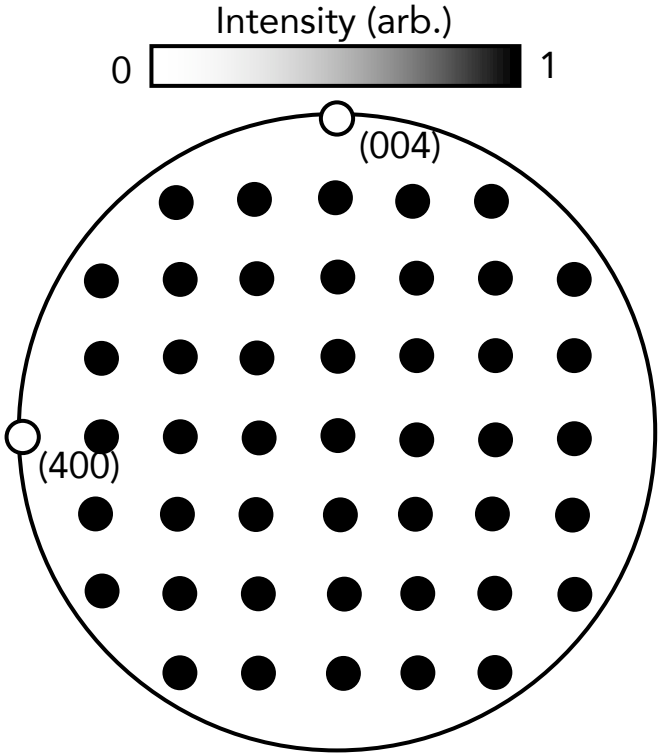
Lovesey, *Theory of Neutron Scattering from Condensed Matter*, OUP (1987)
Blech & Averbach, *Physics* **1**, 31 (1964)

Neutron scattering – Order *vs* disorder

- Long-range order → Bragg scattering



Real space, **r**



Reciprocal space, **Q**

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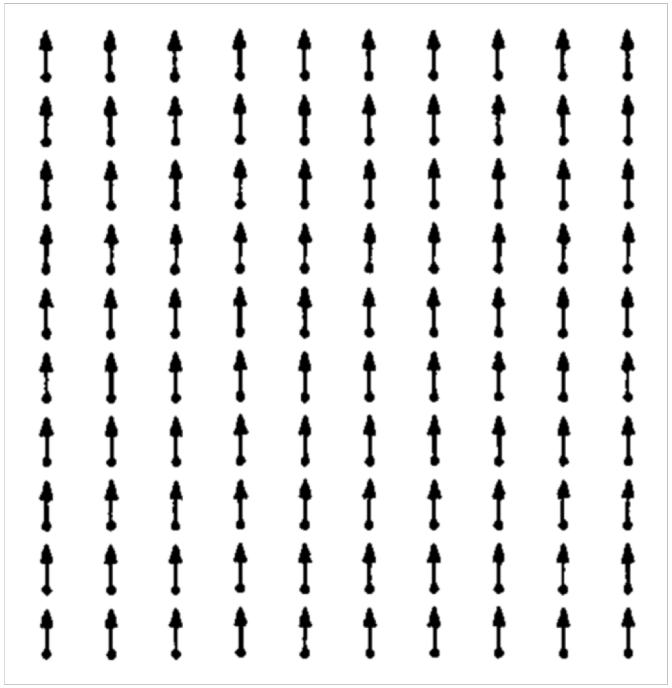

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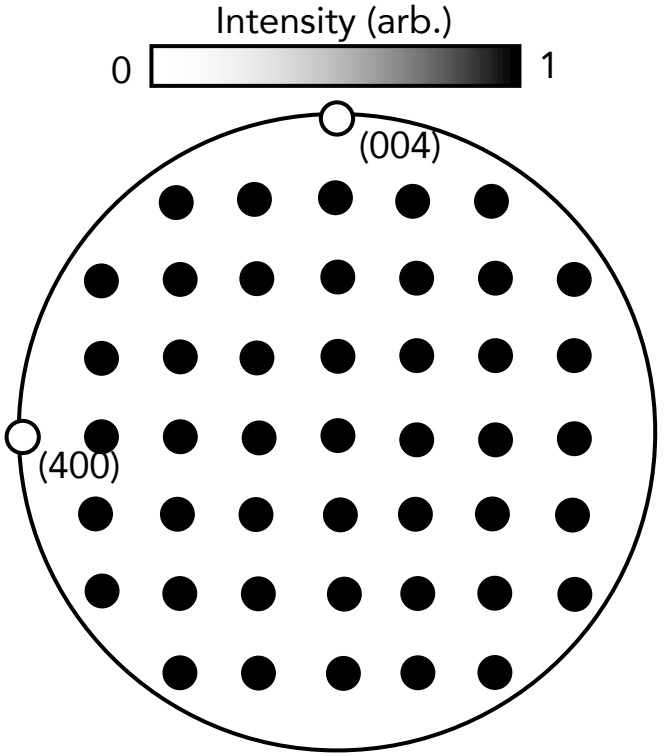
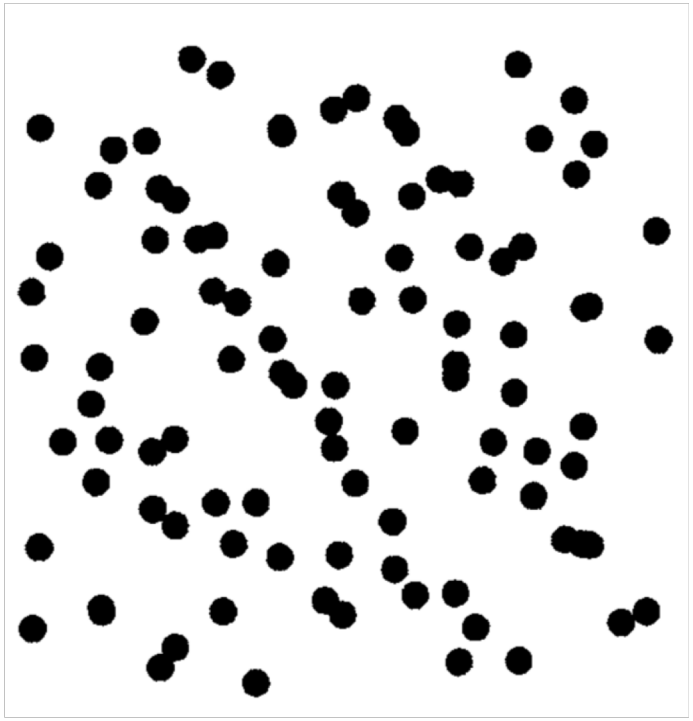


Reciprocal space, **Q**

Neutron scattering – Order *vs* disorder

- Random disorder → flat scattering



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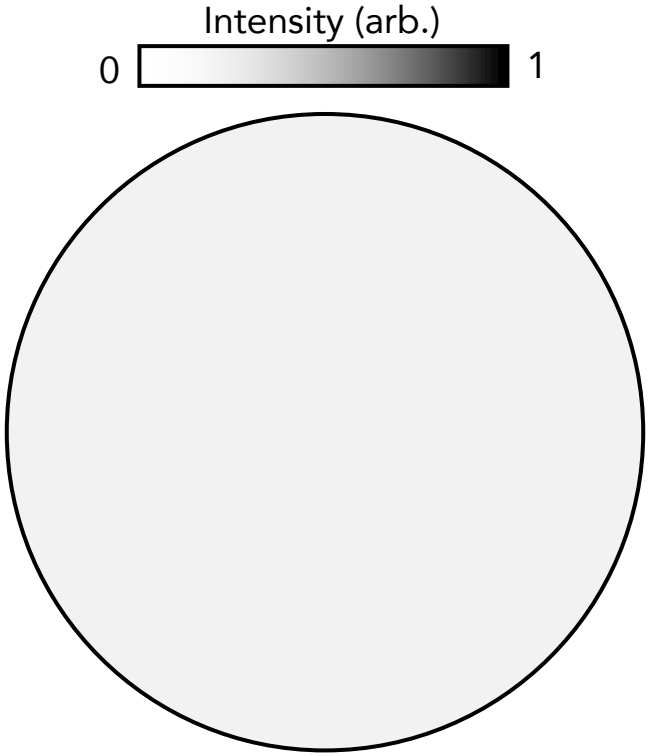

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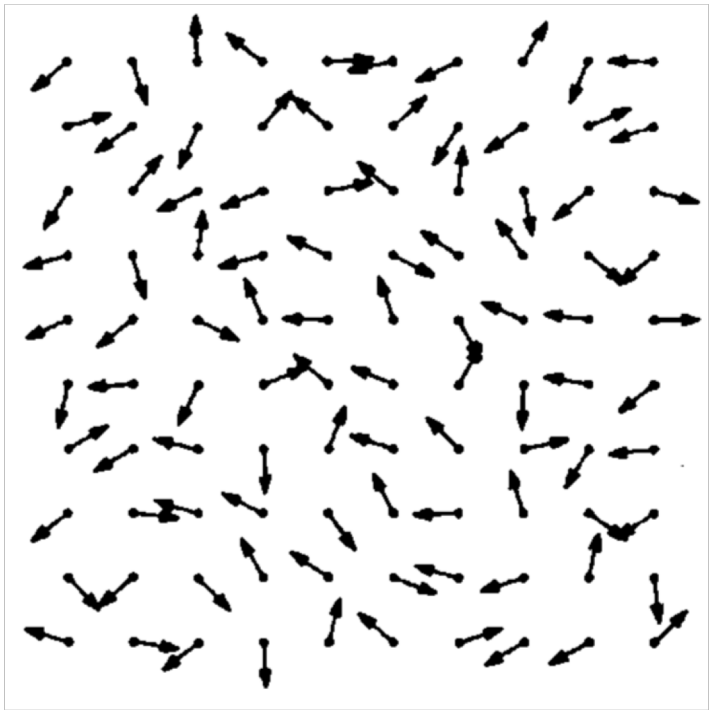

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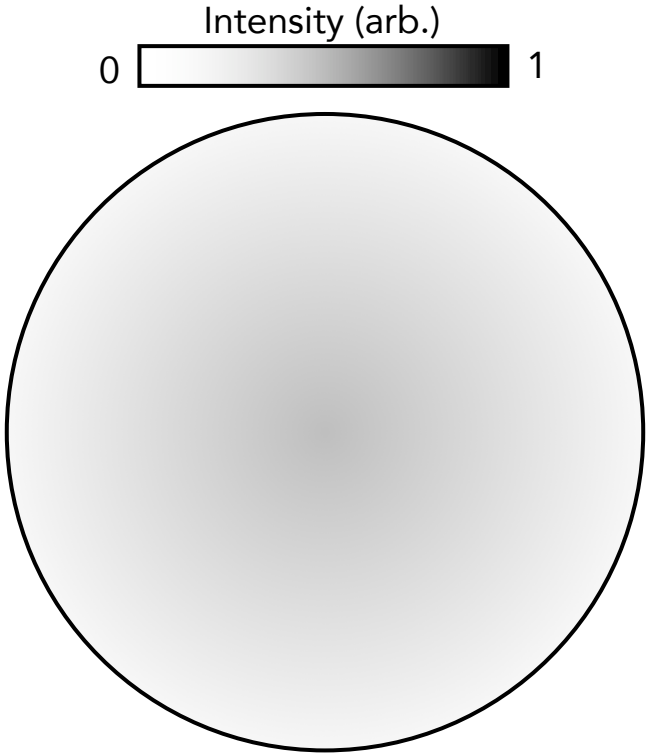
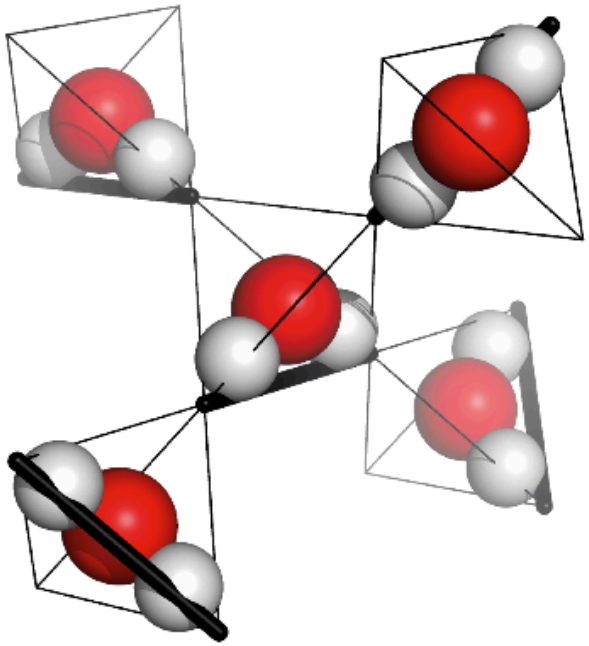
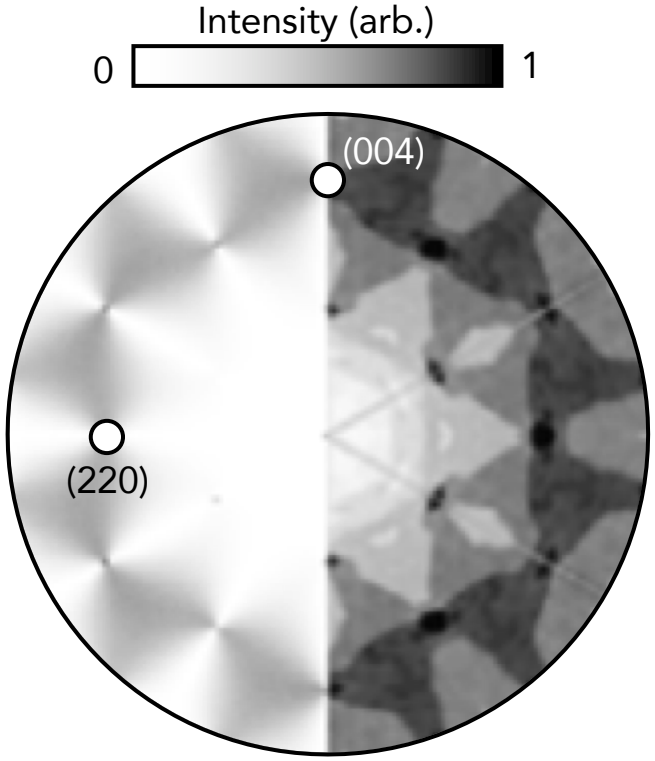


Reciprocal space, **Q**

Neutron scattering – Order *vs* disorder

- Correlated disorder → structured diffuse scattering





Real space, **r**

Reciprocal space, **Q**

Neutron scattering

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

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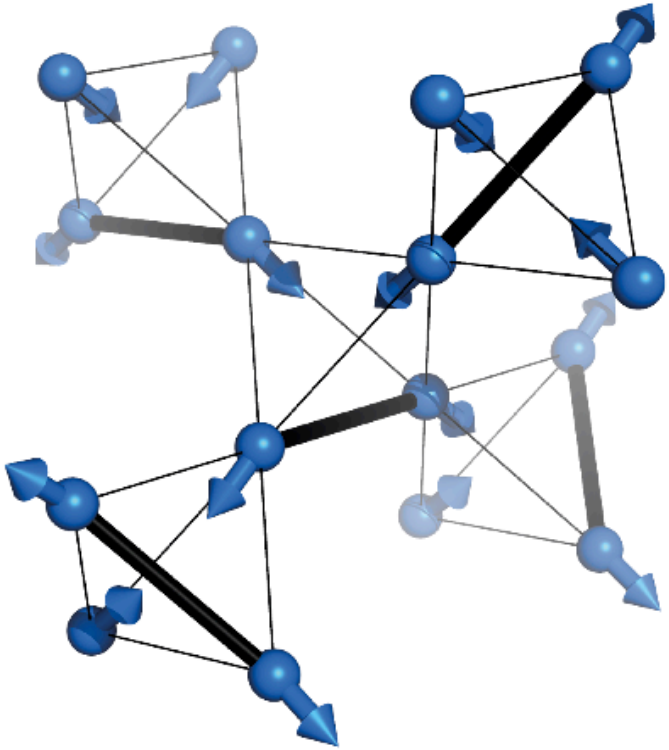

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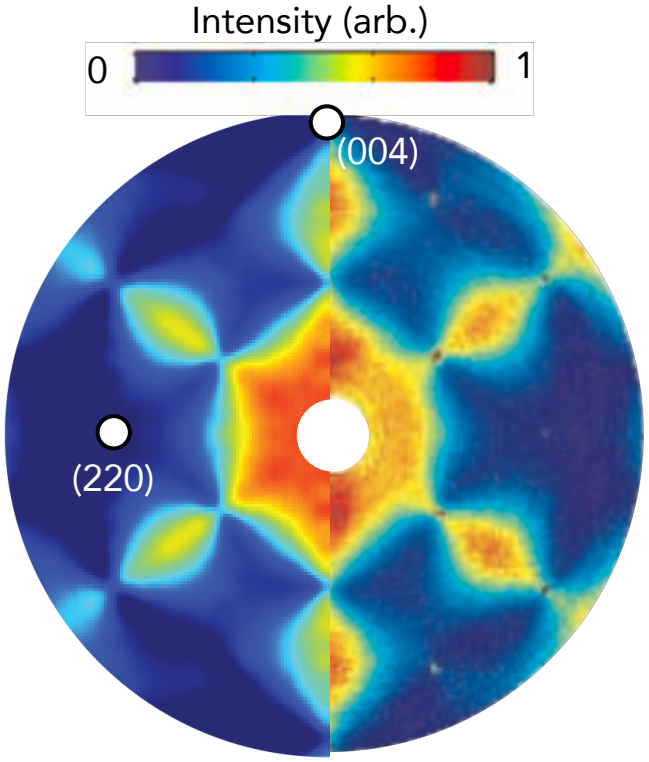


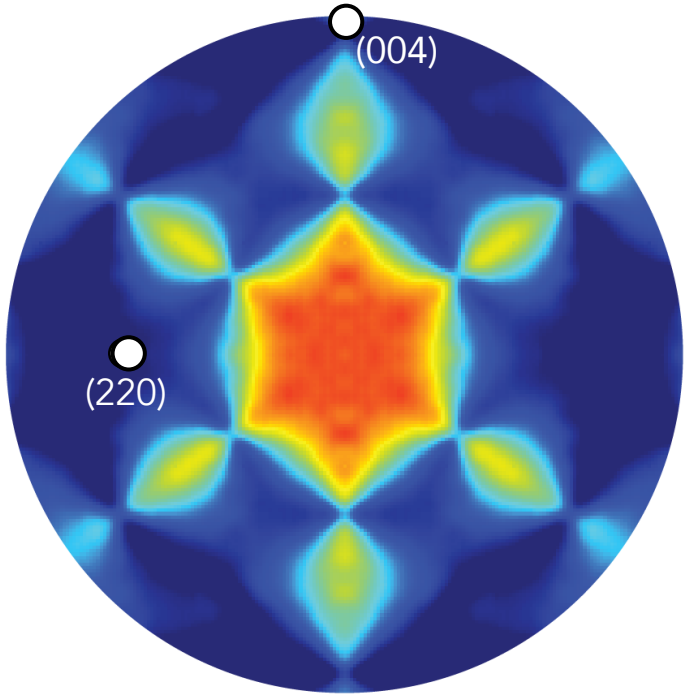
Reciprocal space, **Q**

Spin-polarised neutron scattering

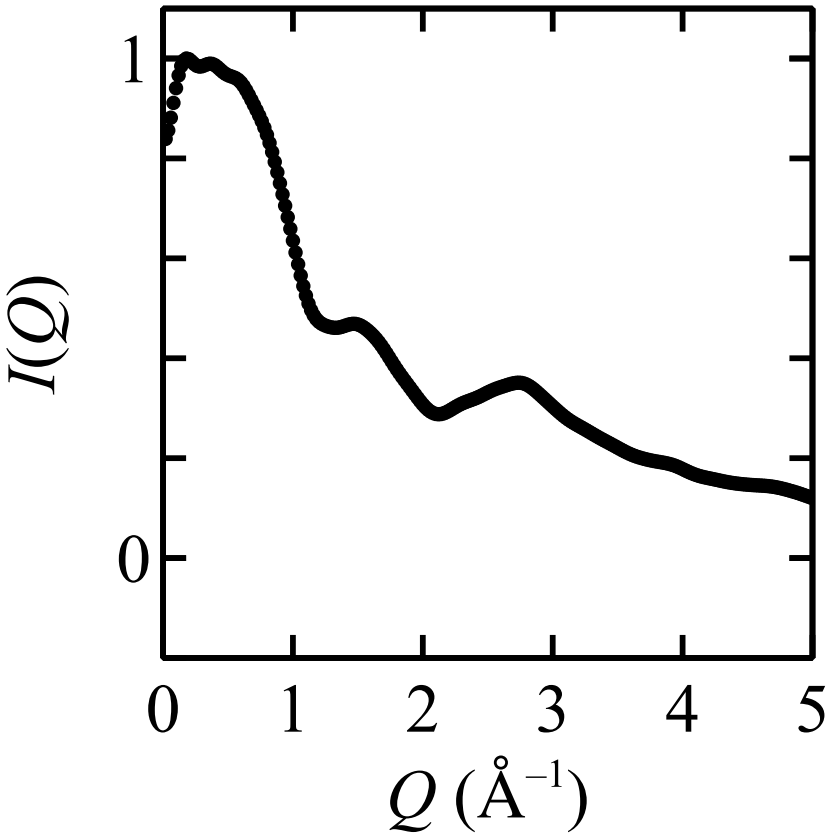
Fennell *et al.*, *Science* **326,** 415 (2009)

Neutron scattering – Powder *vs* crystal

single crystal



powder



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

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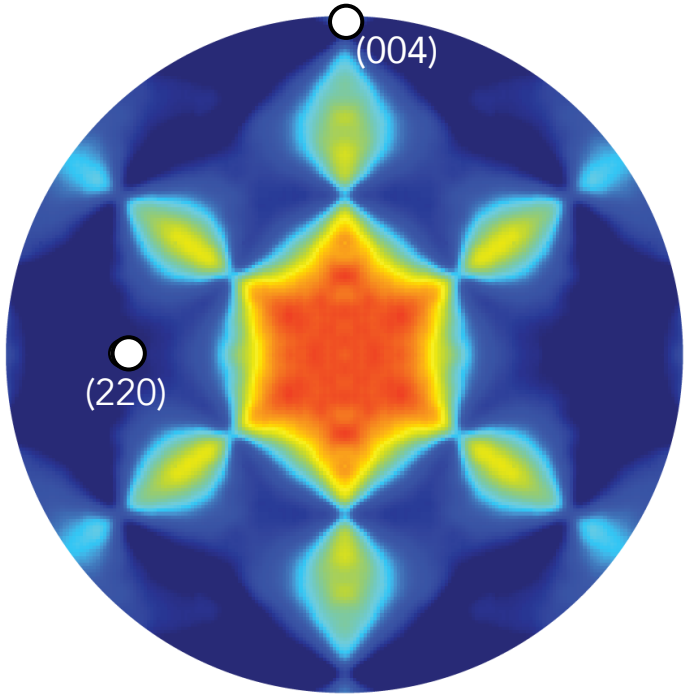
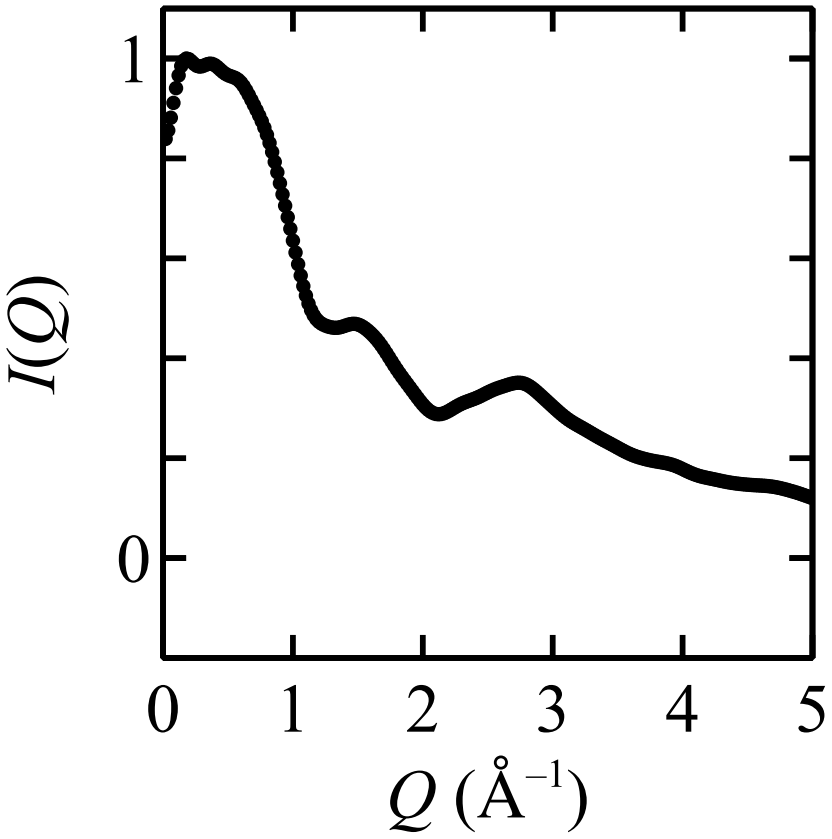


?

powder



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

Plan

- Introduction
- Magnetic neutron scattering
- Reverse Monte Carlo & Spinvert
- Powder case study: $Gd_3Ga_5O_{12}$
- Single-crystal refinements

Reverse Monte Carlo (RMC) refinement

❑ **Goodness-of-fit metric:**

$$\chi^2 = W \sum_Q \left[\frac{I_{\mathrm{calc}}(Q) - I_{\mathrm{expt}}(Q)}{\sigma(Q)} \right]^2$$

1. Big box: ~10^4 spin vectors, initially randomised
2. **Rotate** a spin at random

Accept move if $\Delta\chi^2 \leq 0$

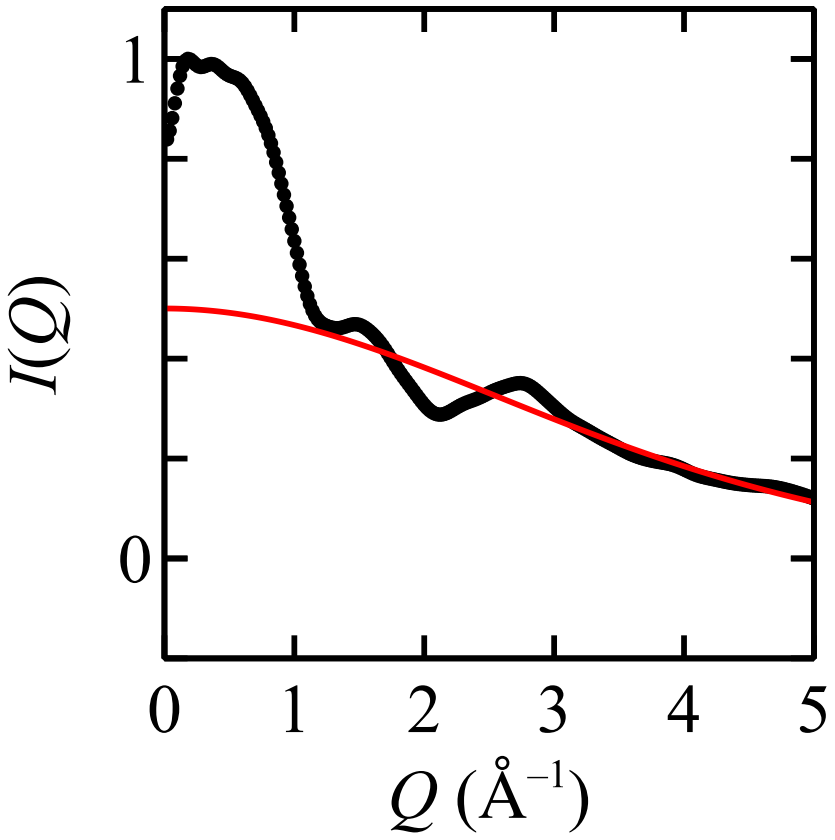
Otherwise, accept with probability $\exp(-\Delta\chi^2/2)$

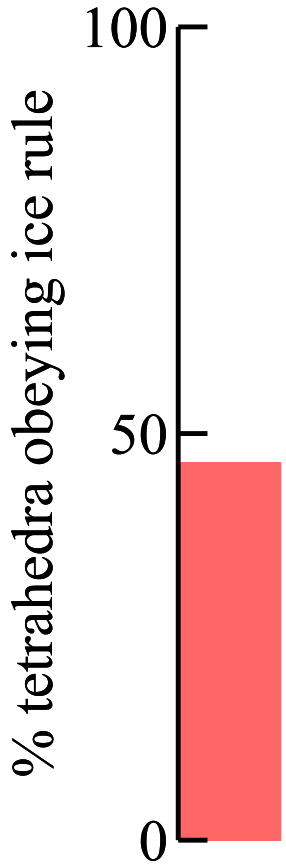
3. **Repeat step 2** until fit achieved
4. **Calculate** diffuse scattering, correlation functions, etc..

McGreevy, *JPCM* **13**, R877 (2001)
Keen & McGreevy, *JPCM* **3**, 7383 (1991)

Spin ice – Local structure solution

- Reverse Monte Carlo refinement from random initial configurations



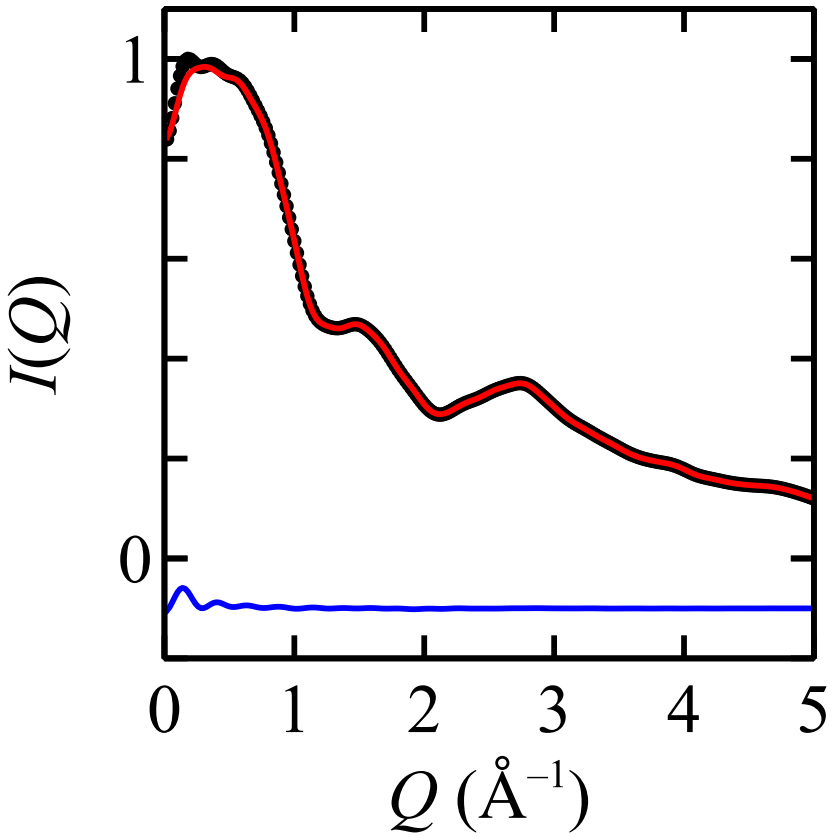


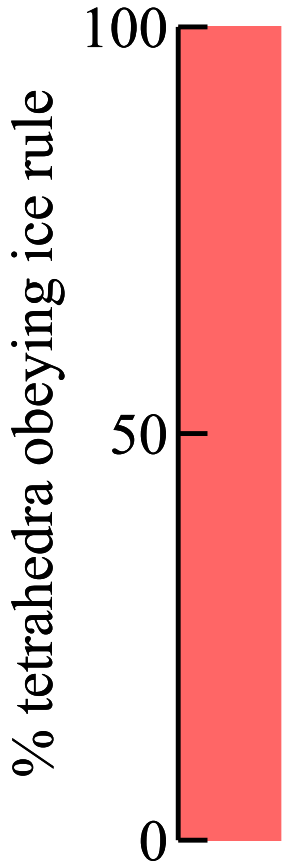
Andrew Goodwin
Oxford

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

Spin ice – Local structure solution

- Reverse Monte Carlo refinement from random initial configurations



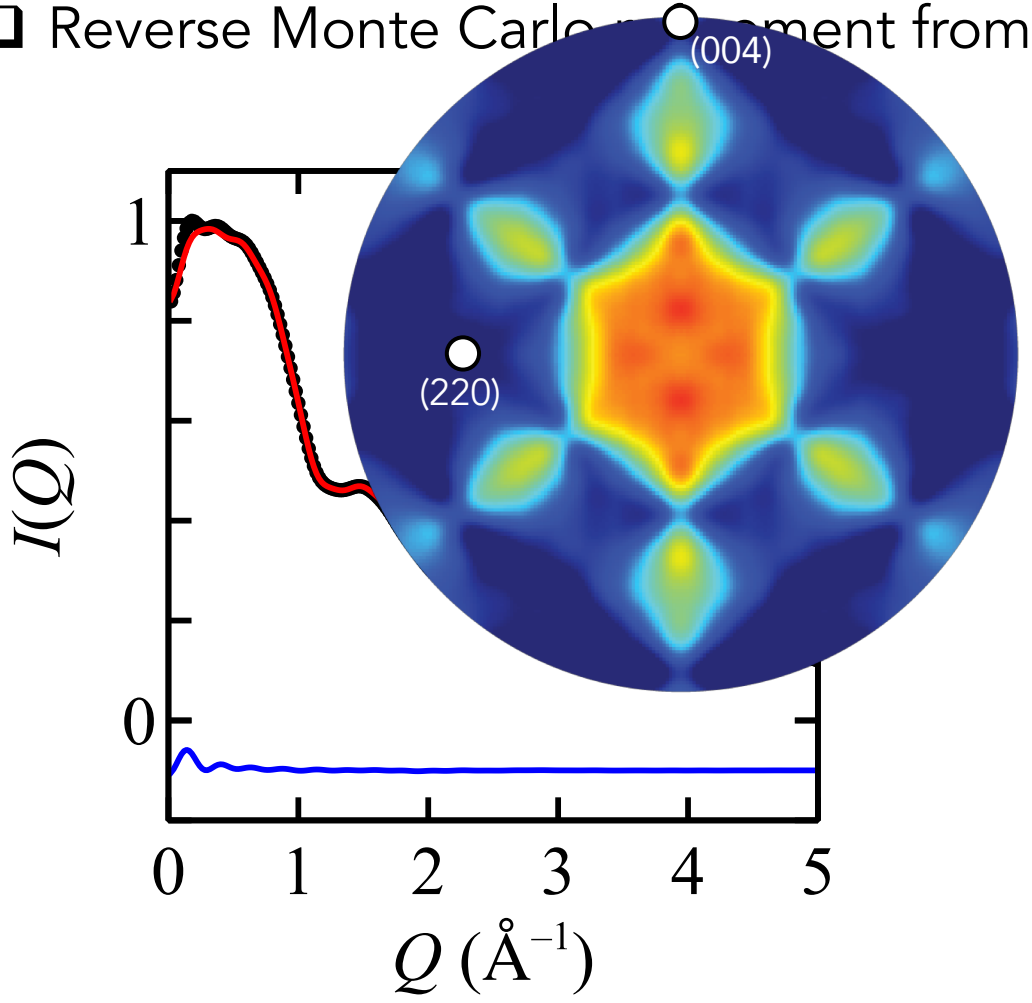


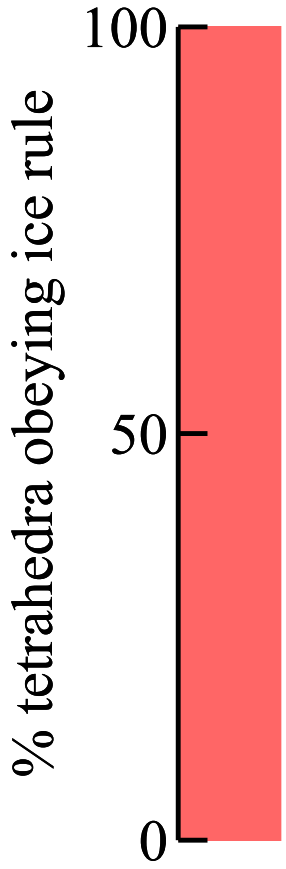
Andrew Goodwin
Oxford

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

Spin ice – Local structure solution

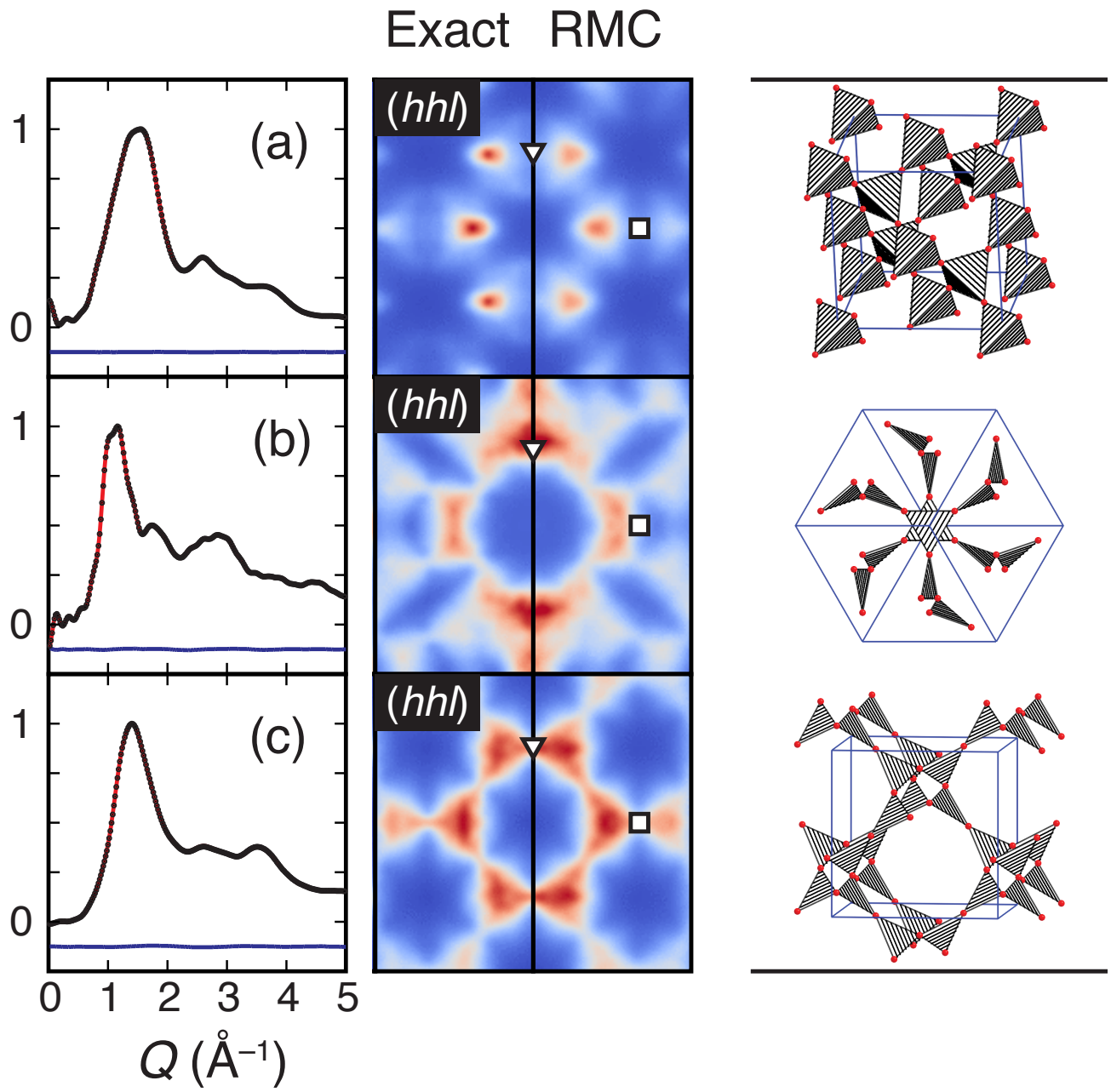
- Reverse Monte Carlo refinement from random initial configurations





Andrew Goodwin
Oxford

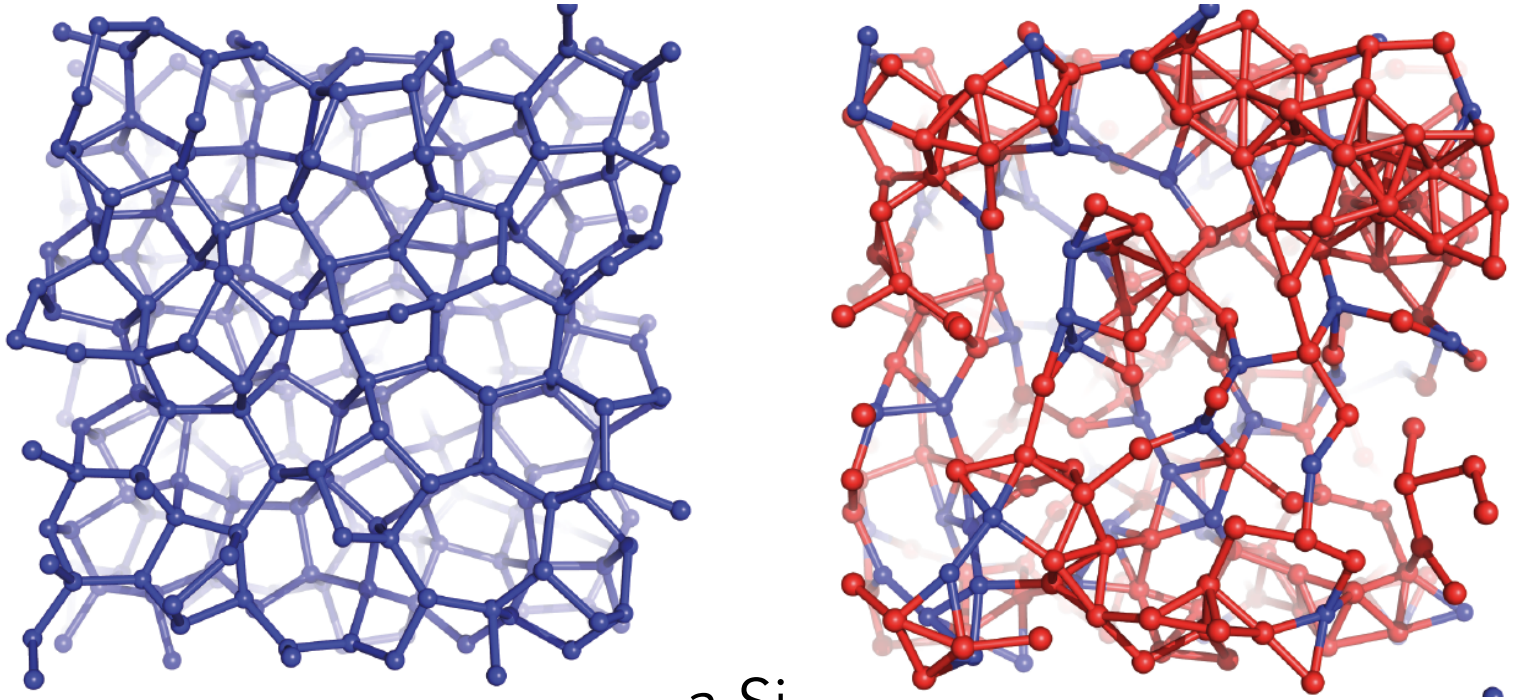
Paddison & Goodwin, *PRL* **108**, 017204 (2012)



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

A note of caution

- RMC produces most disordered configuration consistent with **input data** and **constraints** (crystal structure, spin length)
- Spin liquids **more strongly constrained** than actual liquids & amorphous systems



a-Si

Cliffe & Goodwin, *Phys Stat Sol B* **250**, 949 (2013)

Spinvert – What does it do?

IOP PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **25** (2013) 454220 (15pp) doi:10.1088/0953-8984/25/45/454220

SPINVERT: a program for refinement of paramagnetic diffuse scattering data

Joseph A M Paddison[1,2], J Ross Stewart[2] and Andrew L Goodwin[1]

- Refinement of *atomistic model* to *magnetic diffuse scattering* data
- "Model independent" – no spin Hamiltonian
- **Design principles:**
  - Easy to use
  - Fast to run
  - Straightforward to customise

Paddison, Stewart & Goodwin, *J. Phys.: Condens. Matter* **25,** 454220 (2013)

Spinvert – What doesn't it do?

IOP PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **25** (2013) 454220 (15pp) doi:10.1088/0953-8984/25/45/454220

SPINVERT: a program for refinement of paramagnetic diffuse scattering data

Joseph A M Paddison[1,2], J Ross Stewart[2] and Andrew L Goodwin[1]

- Magnetic diffuse scattering *only*
- No positional disorder, no magneto-elastic coupling
- Single type of magnetic ion

Paddison, Stewart & Goodwin, *J. Phys.: Condens. Matter* **25,** 454220 (2013)

Spinvert – How to get it?

IOP PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **25** (2013) 454220 (15pp) doi:10.1088/0953-8984/25/45/454220

SPINVERT: a program for refinement of paramagnetic diffuse scattering data

Joseph A M Paddison[1,2], J Ross Stewart[2] and Andrew L Goodwin[1]

paddisongroup.wordpress.com/software

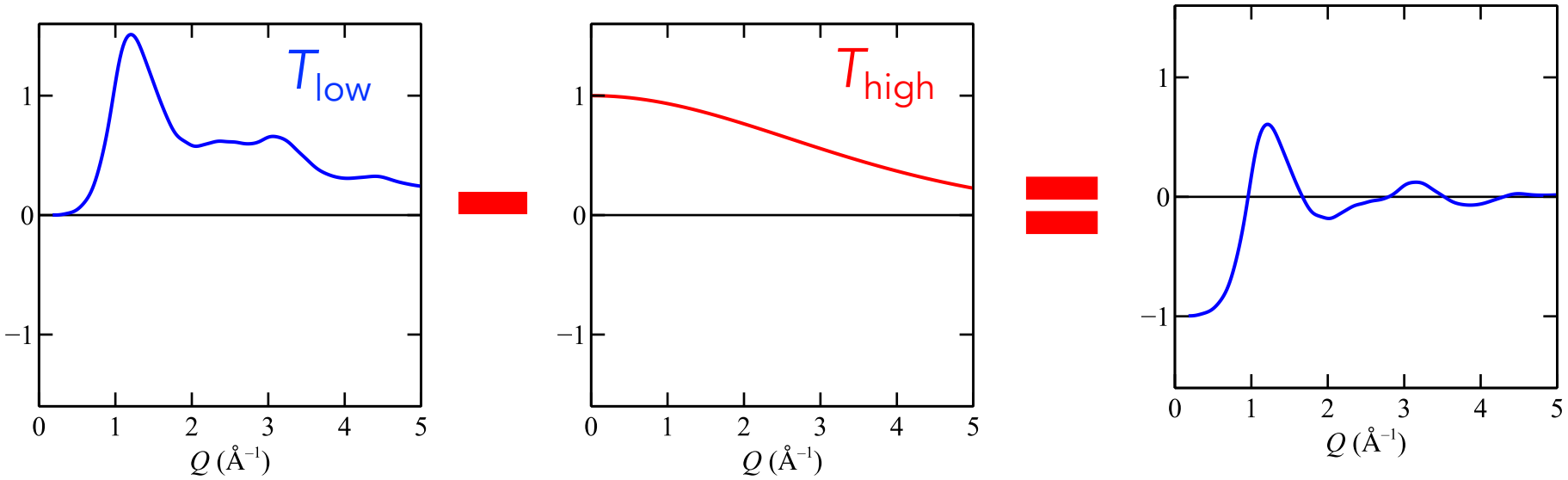
- Program, instructions, examples

- Distributed as Fortran90 source code
  - Needs to be compiled on your system
  - Download compiler from gcc.gnu.org/wiki/GFortranBinaries
  - gfortran spinvert.f90 –o spinvert –O3

Paddison, Stewart & Goodwin, *J. Phys.: Condens. Matter* **25,** 454220 (2013)

Spinvert – How to get suitable data?

- **Key challenge:** isolating magnetic diffuse scattering
- **Three main methods:**
  1. Polarised neutrons → e.g. D7 at ILL
  2. "Total scattering" → Matt Tucker & Helen Playford's talk
  3. Temperature subtractions → **special requirements for Spinvert:**



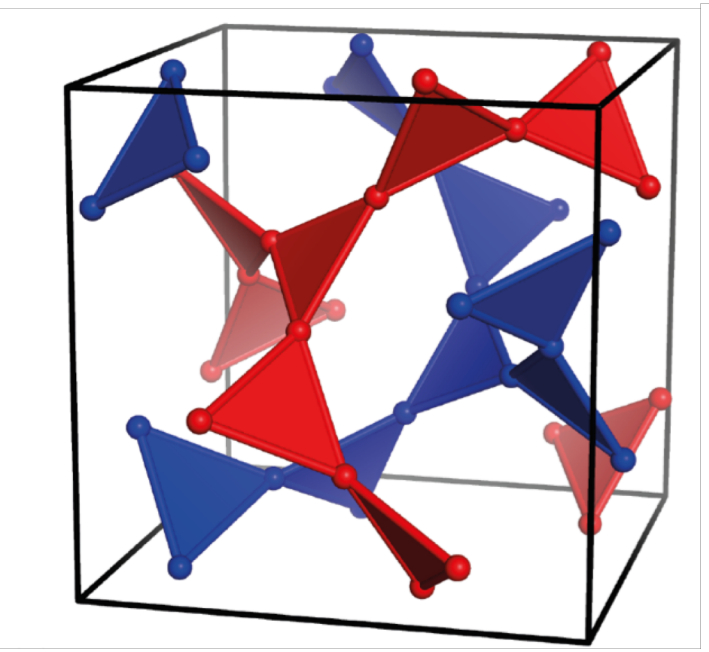
- place data on absolute intensity scale (barn sr^{-1} spin^{-1})
- measure magnetic moment length

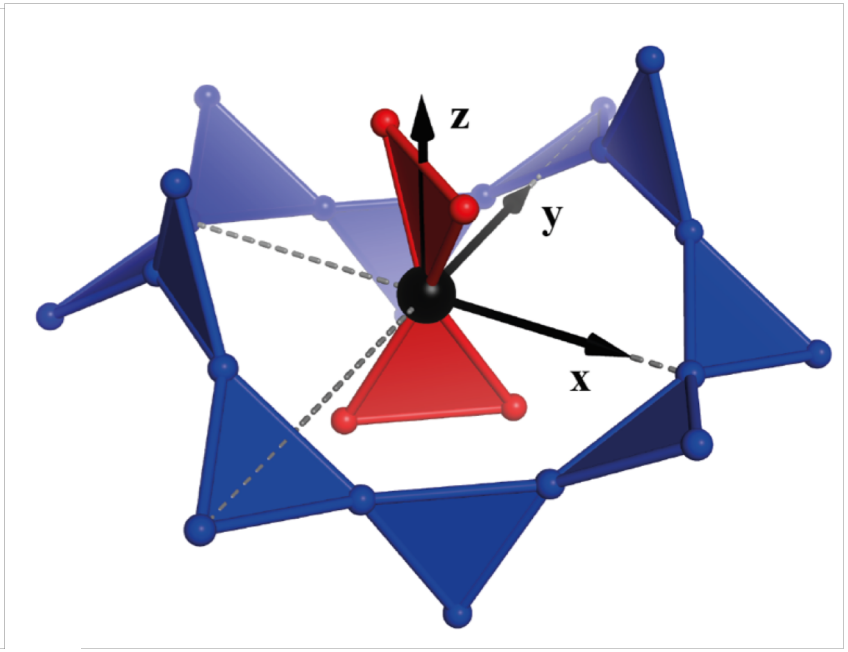
Plan

- Introduction
- Magnetic neutron scattering
- Reverse Monte Carlo & Spinvert
- Powder case study: $Gd_3Ga_5O_{12}$
- Single-crystal refinements

$Gd_3Ga_5O_{12}$ – Structure

- Interpenetrating networks of corner-sharing triangles in 3D
- Antiferromagnetic interactions of strength ~2 K





Pascale Deen
(ESS/Lund)

Henrik Jacobsen
(Copenhagen)

Kinney & Wolf, *J. Appl. Phys.* **50**, 2115 (1979)
Yavors'kii, Enjalran & Gingras, *PRL* **97**, 267203 (2006)

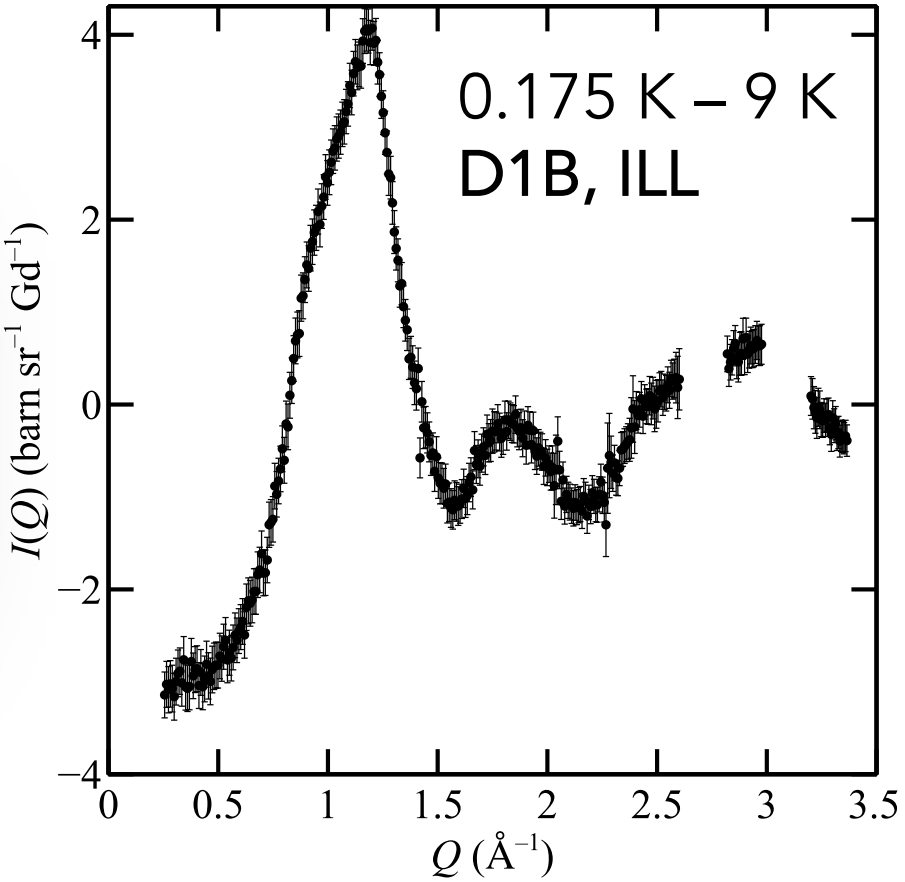
$Gd_3Ga_5O_{12}$ – Experimental data

- Input file: "*name*_data.txt"

ggg_data.txt

```
0.25577 -3.14174 0.24815
0.26446 -3.02750 0.24897
0.27315 -3.08490 0.25056
0.28184 -3.02204 0.25138
0.29053 -3.07314 0.25281
```

Q ($Å^{-1}$) | Intensity ($bn\ sr^{-1}\ spin^{-1}$) | Error



Petrenko, Ritter, Yethiraj & Paul, *PRL* **80**, 4570 (1998)

$Gd_3Ga_5O_{12}$ – Config file

ggg_config.txt — Edited

```
TITLE ggg

CELL 12.349 12.349 12.349 90.0 90.0 90.0

SITE 0.1250  0.0000  0.2500
SITE 0.6250  0.5000  0.7500
SITE 0.2500  0.1250  0.0000
SITE 0.7500  0.6250  0.5000
SITE 0.0000  0.2500  0.1250
SITE 0.5000  0.7500  0.6250
SITE 0.6250  0.0000  0.2500
SITE 0.1250  0.5000  0.7500
SITE 0.7500  0.1250  0.5000
SITE 0.2500  0.6250  0.0000
SITE 0.5000  0.2500  0.3750
SITE 0.0000  0.7500  0.8750
SITE 0.3750  0.5000  0.2500
SITE 0.8750  0.0000  0.7500
SITE 0.5000  0.7500  0.1250
SITE 0.0000  0.2500  0.6250
SITE 0.8750  0.5000  0.2500
SITE 0.3750  0.0000  0.7500
SITE 0.0000  0.7500  0.3750
SITE 0.5000  0.2500  0.8750
SITE 0.2500  0.3750  0.5000
SITE 0.7500  0.8750  0.0000
SITE 0.2500  0.8750  0.5000
SITE 0.7500  0.3750  0.0000

FORM_FACTOR_J0 0.0186 25.3867 0.2895 11.1421 0.7135 3.7520 -0.0217
SPIN_DIMENSION 3

TEMP_SUBTRACT
SCALE 63.0

WEIGHT 500.0
BOX 6 6 6
MOVES 300
```

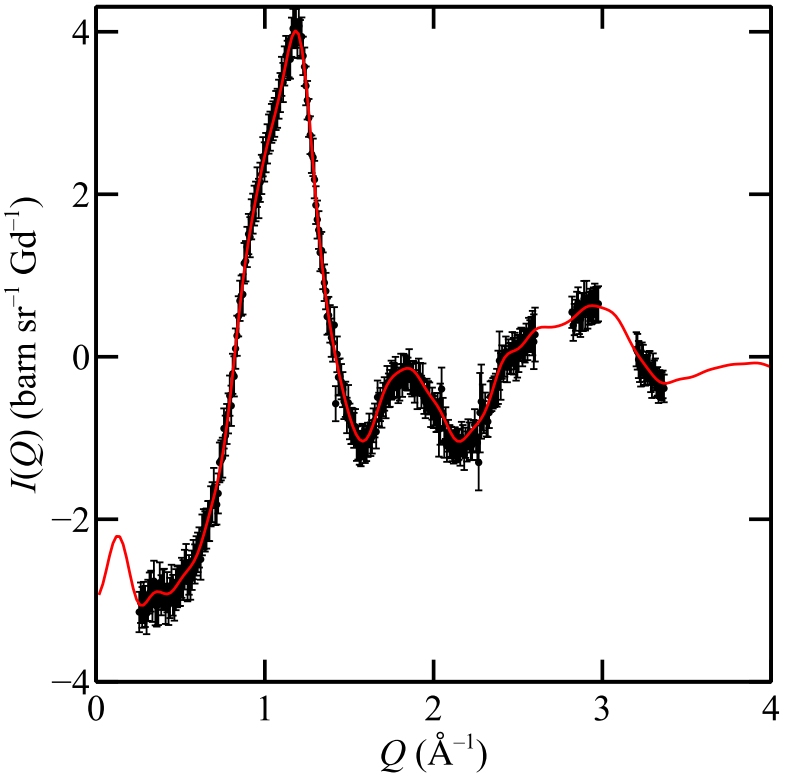
- *"name*_config.txt"
- List of **keywords** followed by **values**
- Keywords can be in any order

$$\text{Scale} = g^2 S(S+1)$$

$$\chi^2 = W \sum_Q \left[\frac{I_{\text{calc}}(Q) - I_{\text{expt}}(Q)}{\sigma(Q)} \right]^2$$

$Gd_3Ga_5O_{12}$ – Fit

- Output file: "*name*_fit_01.txt"
- Calculate powder pattern over extended *Q*-range to check results "make sense"



$Gd_3Ga_5O_{12}$ – Spins file

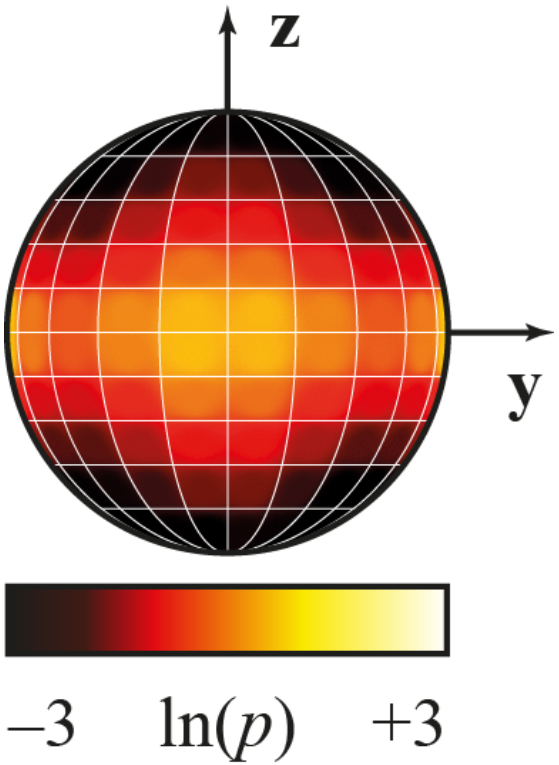
- Output file: "*name*_spins_01.txt"
- Spin orientations and metadata

ggg_spins_01.txt

```
TITLE ggg
CELL    12.349000000000000        12.349000000000000        12.349000000000000        90.0000000        90.0000000        90.0000000
SITE   0.12500000000000000        0.0000000000000000       0.25000000000000000
SITE   0.62500000000000000       0.50000000000000000       0.75000000000000000
SITE   0.25000000000000000       0.12500000000000000        0.0000000000000000
SITE   0.75000000000000000       0.62500000000000000       0.50000000000000000
SITE    0.0000000000000000       0.25000000000000000       0.12500000000000000
SITE   0.50000000000000000       0.75000000000000000       0.62500000000000000
SITE   0.62500000000000000        0.0000000000000000       0.25000000000000000
SITE   0.12500000000000000       0.50000000000000000       0.75000000000000000
SITE   0.75000000000000000       0.12500000000000000       0.50000000000000000
SITE   0.25000000000000000       0.62500000000000000        0.0000000000000000
SITE   0.50000000000000000       0.25000000000000000       0.37500000000000000
SITE    0.0000000000000000       0.75000000000000000       0.87500000000000000
SITE   0.37500000000000000       0.50000000000000000       0.25000000000000000
SITE   0.87500000000000000        0.0000000000000000       0.75000000000000000
SITE   0.50000000000000000       0.75000000000000000       0.12500000000000000
SITE    0.0000000000000000       0.25000000000000000       0.62500000000000000
SITE   0.87500000000000000       0.50000000000000000       0.25000000000000000
SITE   0.37500000000000000        0.0000000000000000       0.75000000000000000
SITE    0.0000000000000000       0.75000000000000000       0.37500000000000000
SITE   0.50000000000000000       0.25000000000000000       0.87500000000000000
SITE   0.25000000000000000       0.37500000000000000       0.50000000000000000
SITE   0.75000000000000000       0.87500000000000000        0.0000000000000000
SITE   0.25000000000000000       0.87500000000000000       0.50000000000000000
SITE   0.75000000000000000       0.37500000000000000        0.0000000000000000
BOX           6           6           6
FORM_FACTOR_J0    1.8599999999999998E-002   25.386700000000001       0.28949999999999998        11.142099999999999       0.71350000000000002
3.7519999999999998       -2.1700000000000001E-002
PROPOSED_MOVES       886464
ACCEPTED_MOVES       398957
WEIGHT    500.00000000000000
TEMP_SUBTRACT
CHI_SQUARED    69.073821040774575
R_FACTOR    6.4884619747326413
SCALE    63.000000000000000
FLAT_BACKGROUND    0.0000000000000000
LINEAR_BACKGROUND    0.0000000000000000
SPIN           1           0           0           0  0.82264674345196132      -0.56685015688850382       -4.3968569721580822E-002
SPIN           2           0           0           0  0.95521777682432363       -8.8001464444131025E-002  0.28251502808609136
SPIN           3           0           0           0 -0.73250525848391879       0.18794069290076534      -0.65430447212699649
SPIN           4           0           0           0 -0.52705374016138040       0.84962067581191258        1.8948937036386431E-002
```

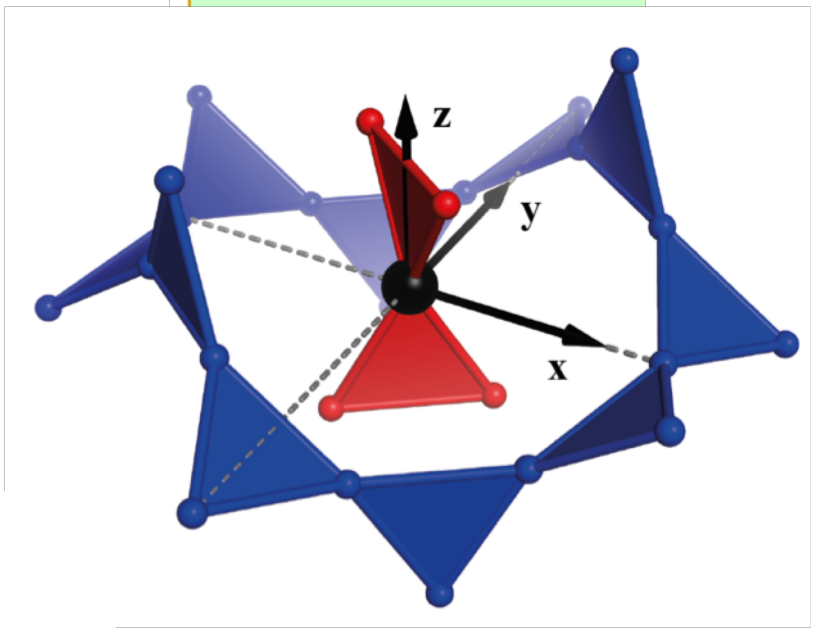
$Gd_3Ga_5O_{12}$ – Analysing the results
Single spins

- "Spinplot" program



Gd^{3+} site symmetry from International Tables:

24	*c*	2.22



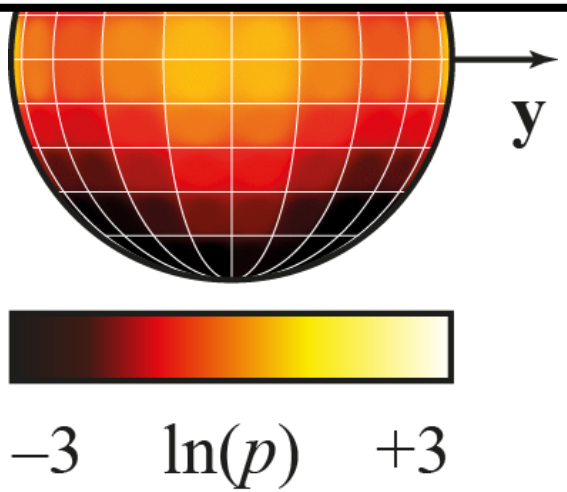
$Gd_3Ga_5O_{12}$ – Analysing the results
Single spins

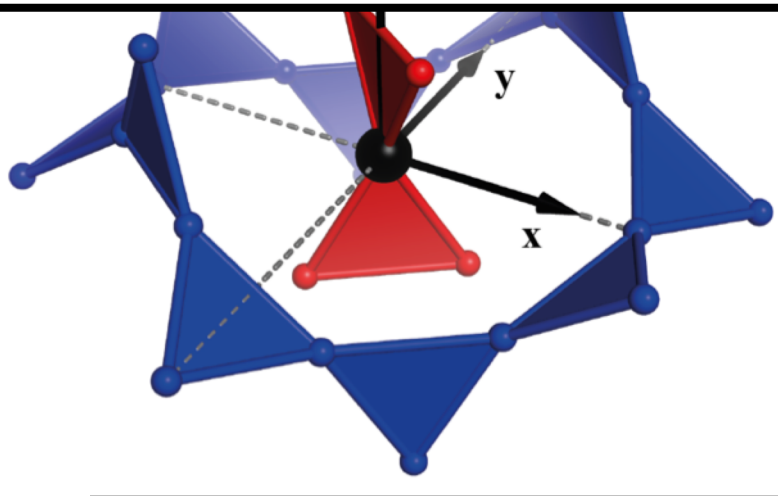
VOLUME 92, NUMBER 16 PHYSICAL REVIEW LETTERS week ending 23 APRIL 2004

Planar Spin Fluctuations with a Quadratic Thermal Dependence Rate in Spin Liquid $Gd_3Ga_5O_{12}$

P. Bonville and J. A. Hodges
C.E.A., Centre d'Etudes de Saclay, Service de Physique de l'Etat Condensé, 91191 Gif-sur-Yvette, France

J. P. Sanchez and P. Vulliet
C.E.A., Centre d'Etudes de Grenoble, Service de Physique Statistique, Magnétisme et Supraconductivité, 38054 Grenoble, France
(Received 10 November 2003; published 23 April 2004)

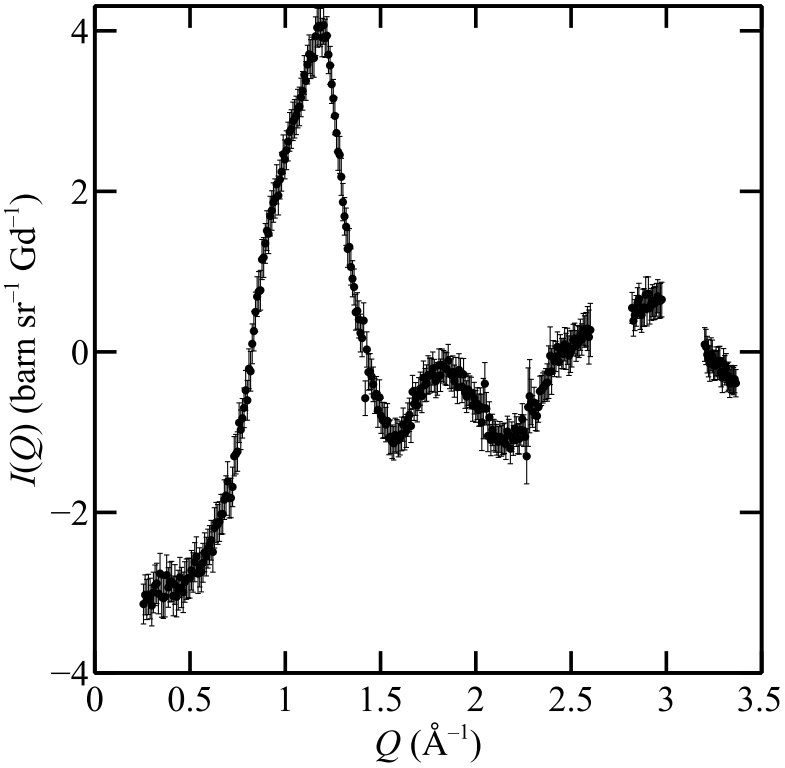




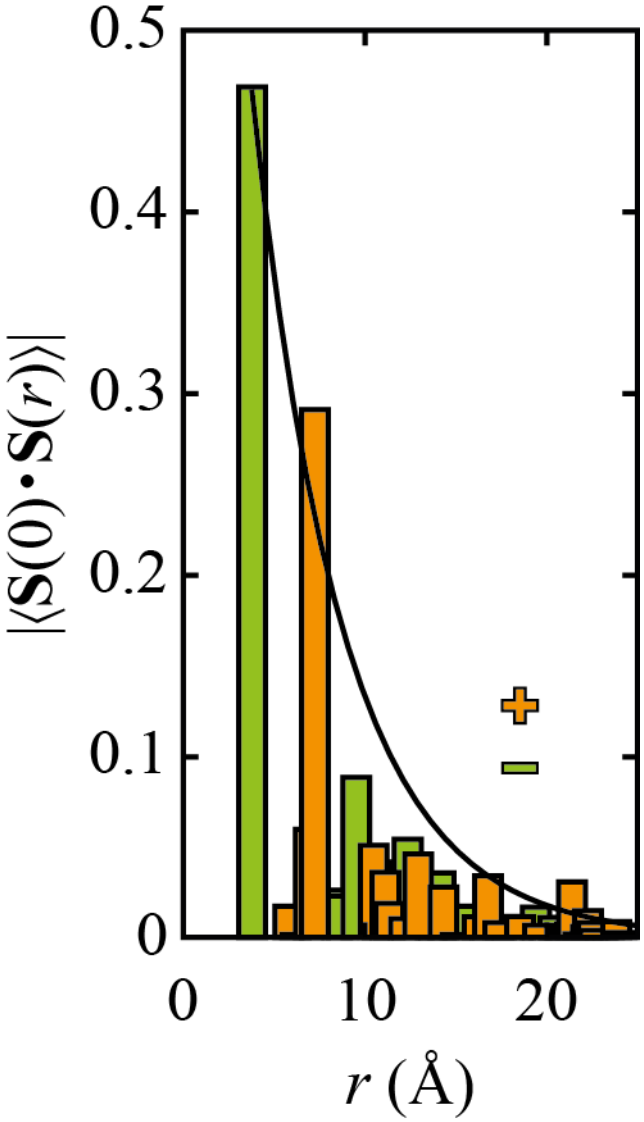
Bonville, Hodges, Sanchez & Vulliet, *PRL* **92**, 167202 (2004)

$Gd_3Ga_5O_{12}$ – Analysing the results
Spin correlations

- "Spincorrel" program



"Fourier transform"

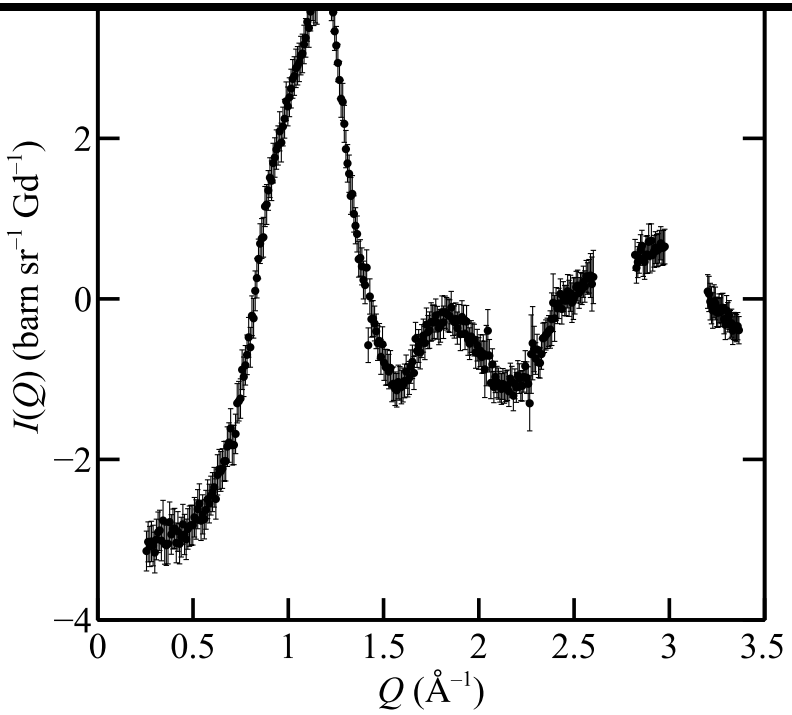


$Gd_3Ga_5O_{12}$ – Analysing the results
Spin correlations

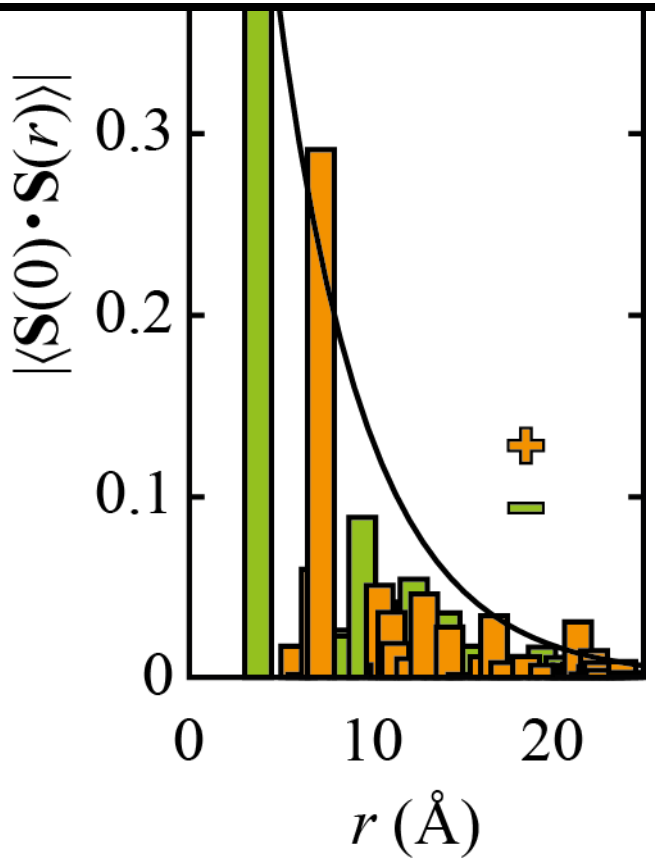
Acta Crystallographica Section A
Foundations and Advances
ISSN 2053-2733

Magnetic pair distribution function analysis of local magnetic correlations

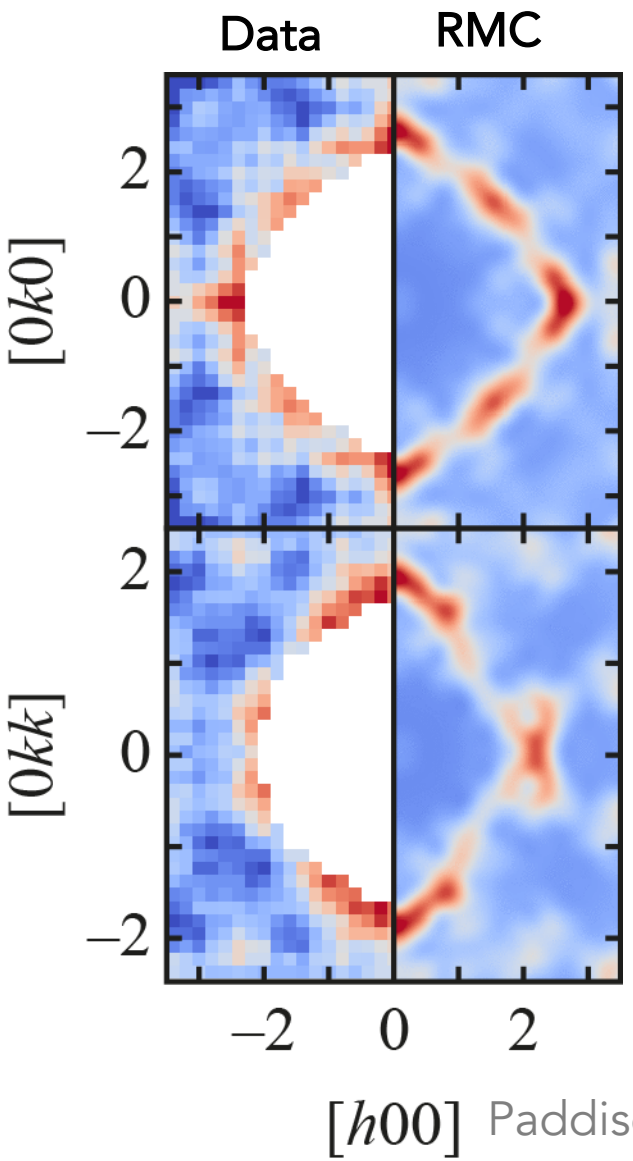
Benjamin A. Frandsen,[a] Xiaohao Yang[b] and Simon J. L. Billinge[b,c]*



"Fourier transform"



$Gd_3Ga_5O_{12}$ – Analysing the results
Single-crystal scattering



- "**Scatty**" program
- Average single-crystal calculation over >10 boxes
- Input file: scatty_config.txt

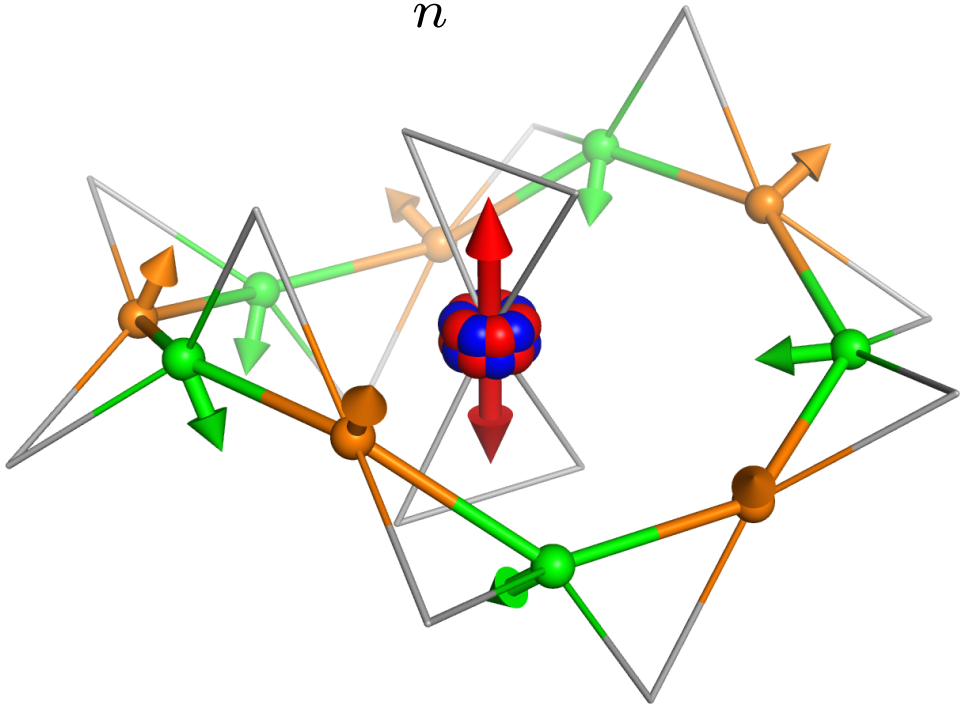
```
NAME hhl
X_AXIS 3 3 0 50
Y_AXIS 0 0 4 50
Z_AXIS 0 0 0 1

SYMMETRY m-3m
RADIATION N
```

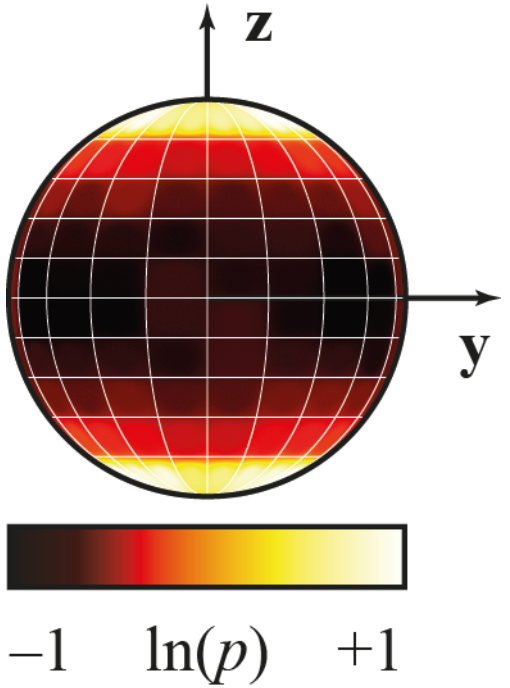
- Data at T = 0.175 K (D9, ILL)

Paddison, *Acta Cryst A* **75**, 14-24 (2019)

$Gd_3Ga_5O_{12}$ – Going further
Collective effects

$$\mathbf{L}(\mathbf{r}) = \frac{1}{10}\sum_n \cos(n\pi)\mathbf{S}_n(\mathbf{r})$$

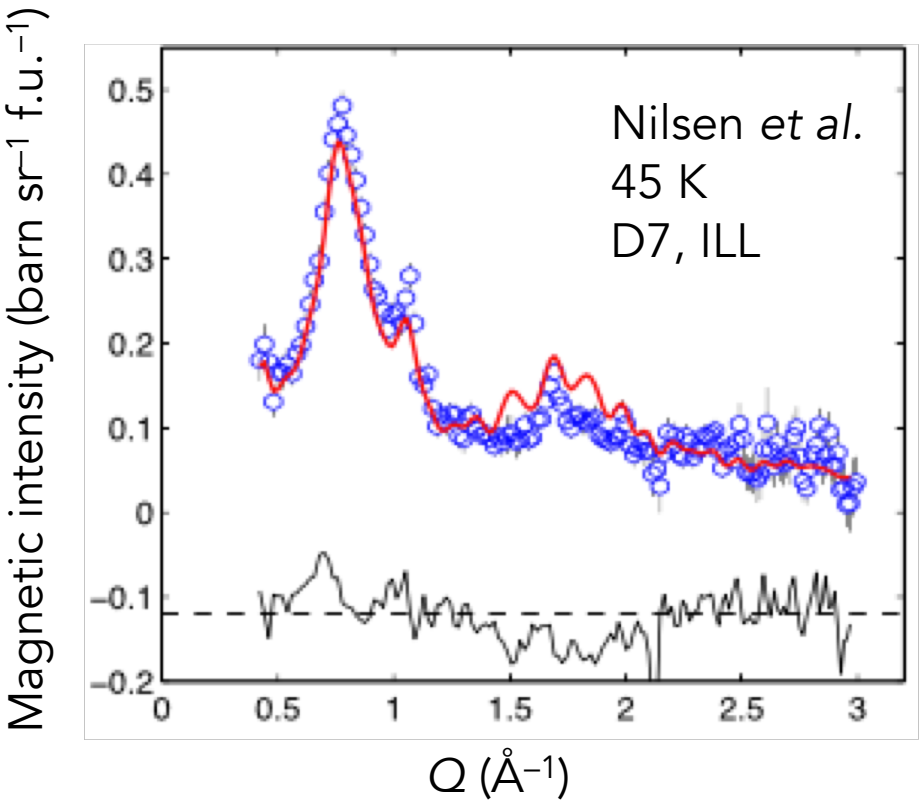


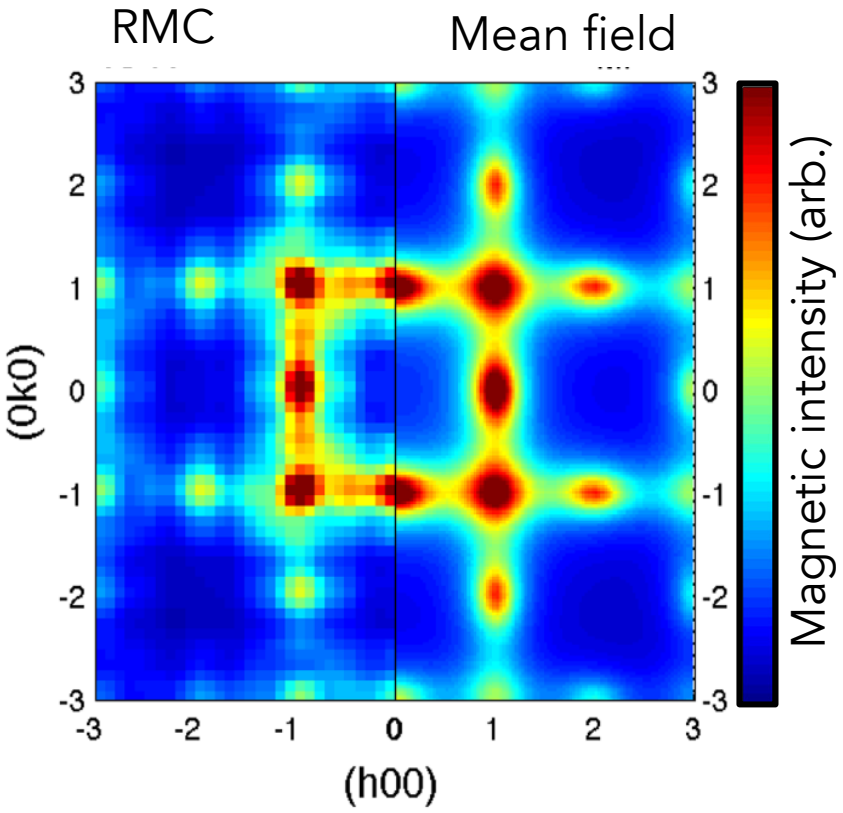


Paddison et al., *Science* **350**, 179 (2015)

Ba_2YRuO_6 – Another example

- Double perovskite structure (FCC magnetic lattice)
- Compare RMC results with J_1-J_2 spin Hamiltonian





Nilsen, Thompson, Ehlers, Marjerrison & Greedan, *PRB* **91**, 054415 (2015)

Plan

- Introduction
- Magnetic neutron scattering
- Reverse Monte Carlo & Spinvert
- Powder case study: $Gd_3Ga_5O_{12}$
- Single-crystal refinements

Spinvert-3D – Single-crystal refinements

- "Spinvert-3D" for 3D refinements is available
- Beta version – please let me know any bugs!

PHYSICAL REVIEW B **97**, 014429 (2018)

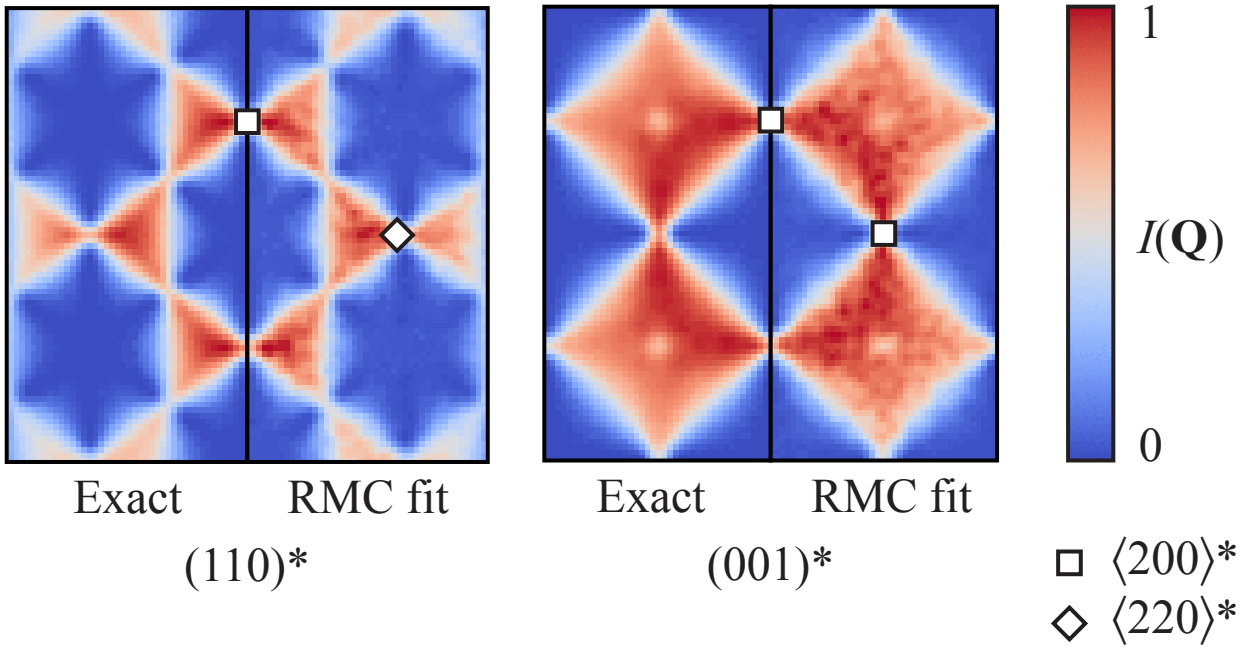
Magnetic structure of paramagnetic MnO

Joseph A. M. Paddison,[1,2,3,4] Matthias J. Gutmann,[2] J. Ross Stewart,[2] Matthew G. Tucker,[2,5,6] Martin T. Dove,[7] David A. Keen,[2] and Andrew L. Goodwin[1,*]

Paddison *et al.*, *PRB* **97**, 014429 (2018)

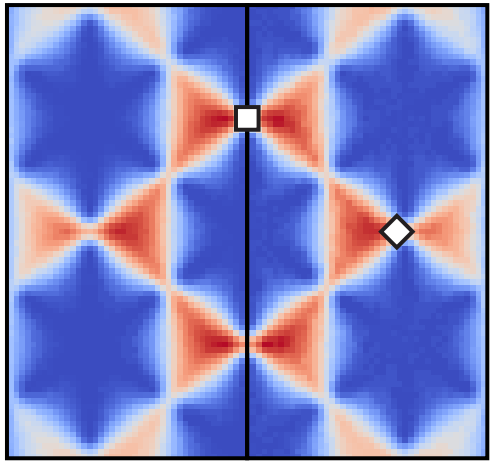
Spinvert-3D – Proof of principle

- Fit to simulated single-crystal data for a model frustrated magnet
  (Heisenberg spins on pyrochlore lattice with AF interactions)
- **First**, fit to **volume** of reciprocal space (~500,000 data points)



Spinvert-3D – Proof of principle

- Fit to simulated single-crystal data for a model frustrated magnet
  (Heisenberg spins on pyrochlore lattice with AF interactions)
- **Second**, fit to **only (110)* plane** of reciprocal space

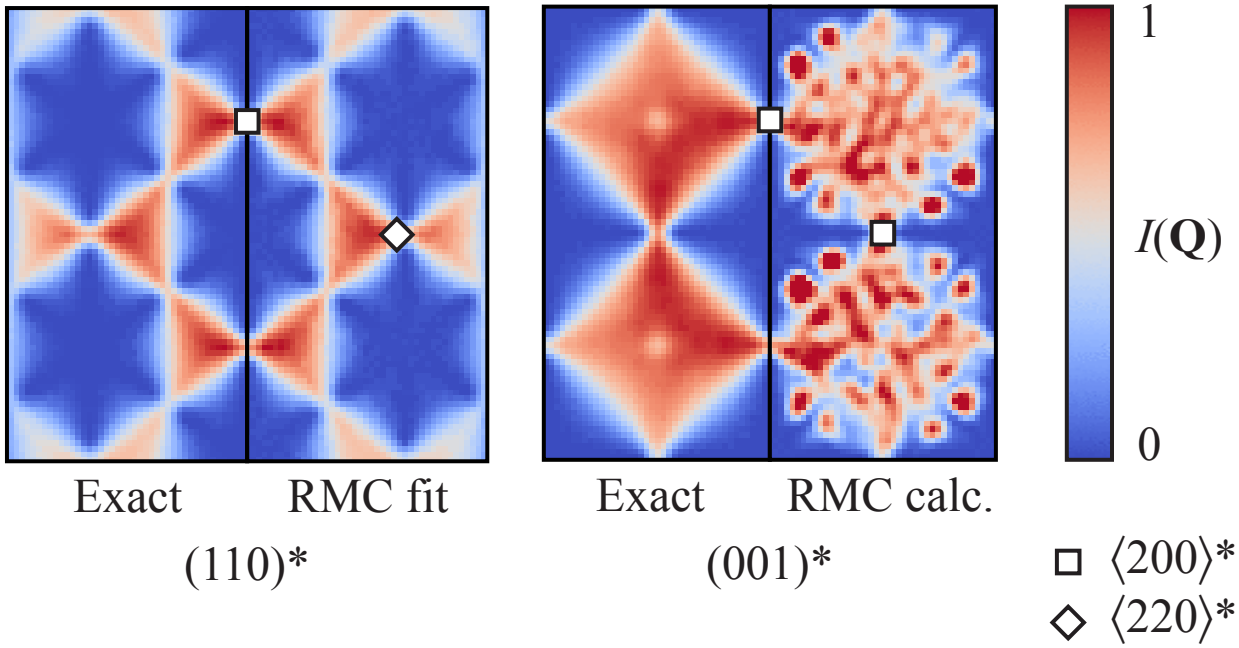


Exact RMC fit

(110)*

Spinvert-3D – Proof of principle

- Fit to simulated single-crystal data for a model frustrated magnet
  (Heisenberg spins on pyrochlore lattice with AF interactions)
- **Second**, fit to **only (110)* plane** of reciprocal space



- **Conclusion:** Spinvert-3D works best with a large volume of data!

Spinvert-3D – Data file

- Input file: "*name*_xtal_data.vtk"
- Visualization Toolkit (.vtk) format
  - Readable in 3D display software, e.g. **ParaView**

py_afm_xtal_data_01.vtk

```
# vtk DataFile Version 2.0
TITLE diffuse magnetic S(Q)
ASCII
DATASET STRUCTURED_POINTS
DIMENSIONS    81    81    81
ORIGIN -4.000000 -4.000000 -4.000000
SPACING  0.100000  0.100000  0.100000
POINT_DATA    531441
SCALARS py_afm float
LOOKUP_TABLE default
   0.0000000000000000
```

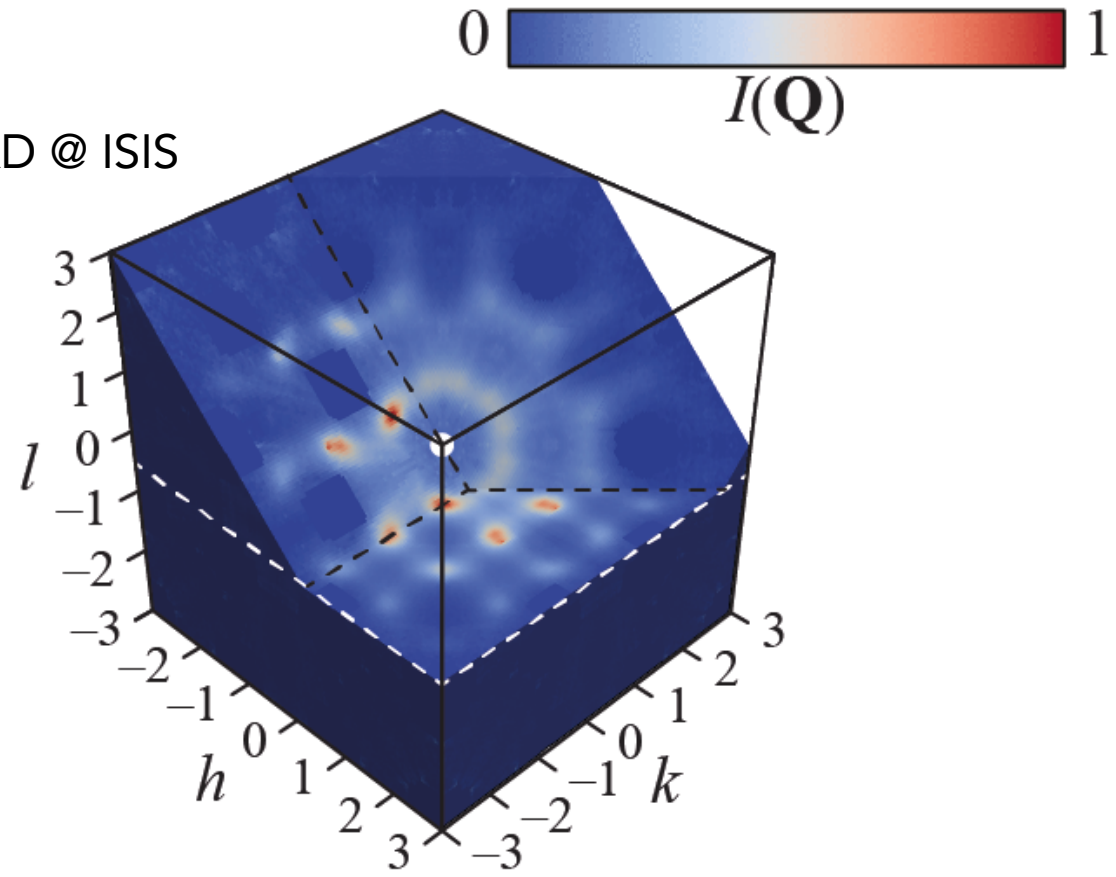
Number of data points along each axis

Origin in reciprocal-lattice (*hkl*) units

Spacing in reciprocal-lattice (*hkl*) units

Spinvert-3D – Paramagnetic MnO

- Rock-salt structure, antiferromagnetic T_N = 118 K

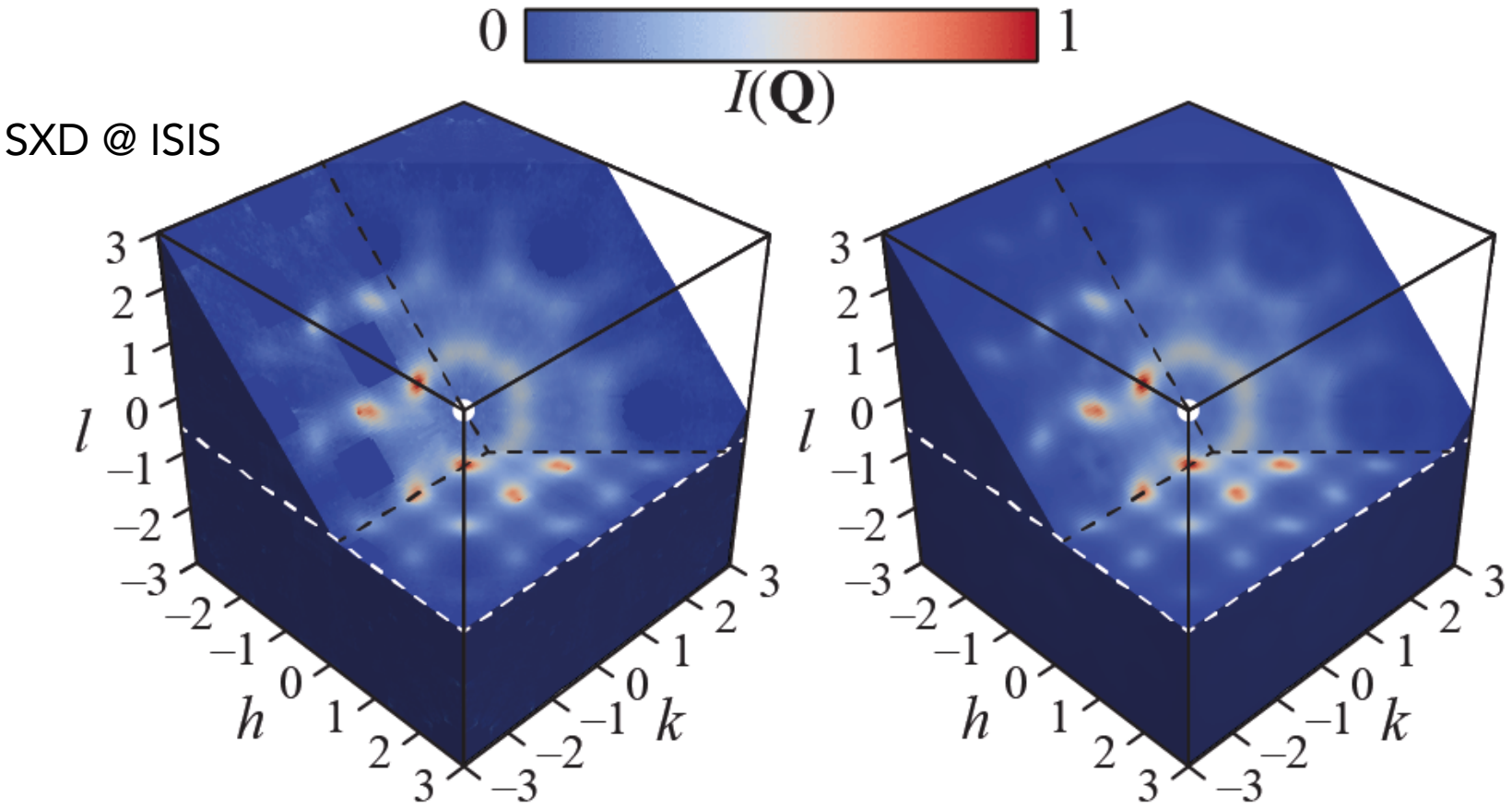


MnO, T = 160 K (> T_N) data

Paddison *et al.*, *PRB* **97**, 014429 (2018)

Spinvert-3D – Paramagnetic MnO

- Exploit information content of 3D scattering data



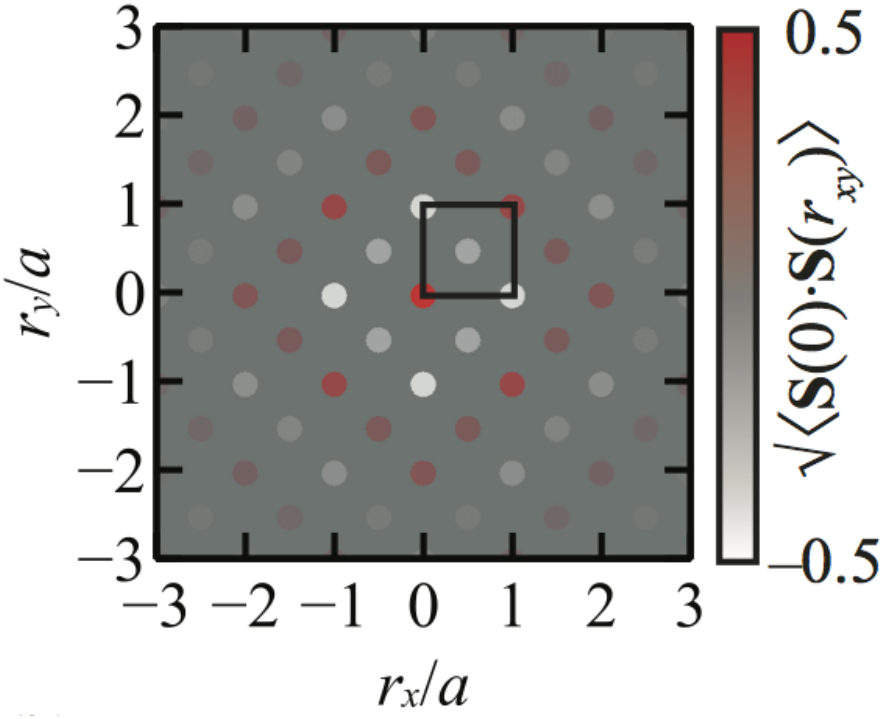
MnO, T = 160 K (> T_N) data *RMC fit*

Paddison *et al.*, *PRB* **97**, 014429 (2018)

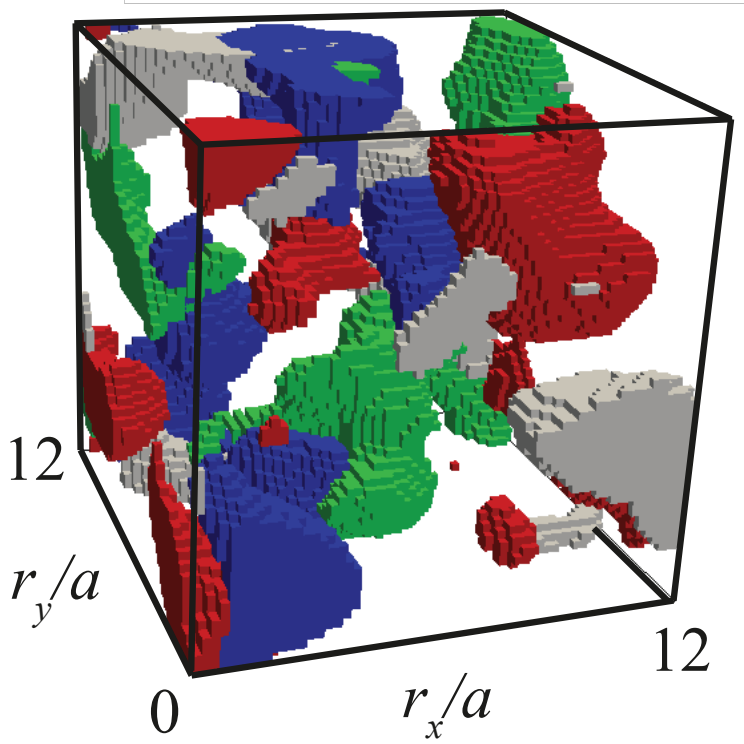
Spinvert-3D – Paramagnetic MnO

- Extract 3D model of domain structure in paramagnetic phase

$$S_{\mathbf{k}}(\mathbf{r}) = \left| \sum_i w(|\mathbf{r} - \mathbf{r}_i|)\, \mathbf{S}_i \exp\left(\mathrm{i} \frac{2\pi}{a} \mathbf{k} \cdot \mathbf{r}_i \right) \right|^2$$



spin correlations

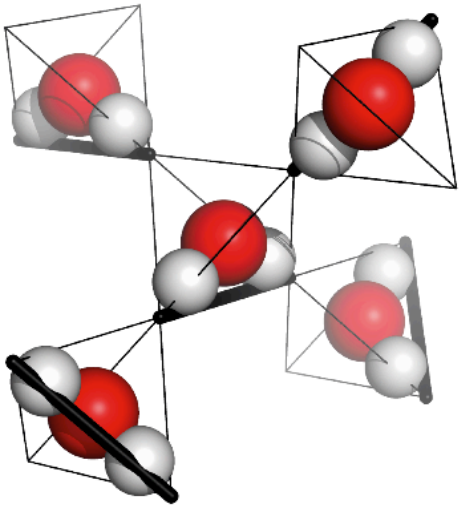


paramagnetic domains

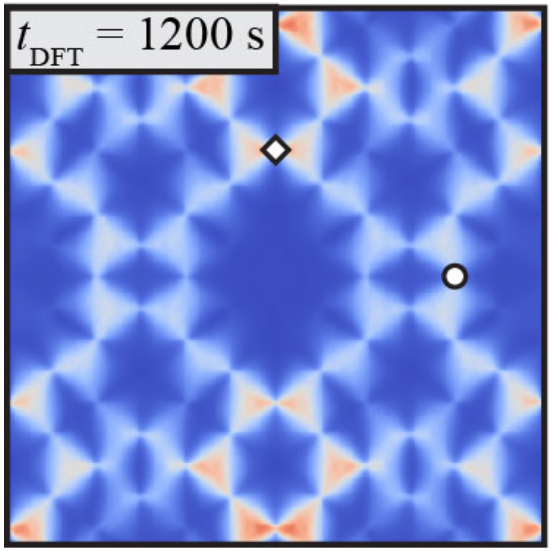
Paddison *et al.*, *PRB* **97**, 014429 (2018)
See also mPDF: Frandsen *et al.*, *Phys. Rev. Lett.* **116**, 197204 (2016)

Ultrafast calculation of 3D diffuse scattering

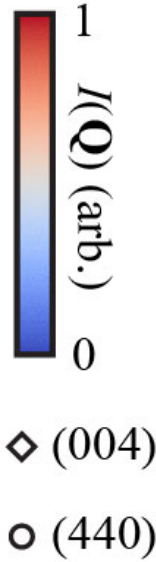
- 3D diffuse-scattering patterns traditionally **slow** to calculate
  - Many atoms/spins, many wavevectors
  - Real-space processing ("sub-boxes") employed to reduce noise
  - Fast Fourier transform considered unsuitable



water ice



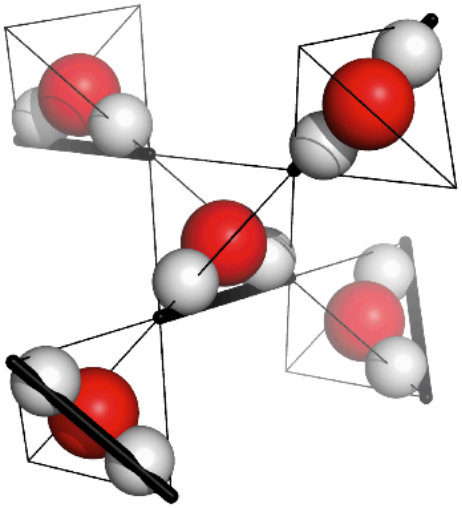
(110) plane*
DFT + sub-boxes



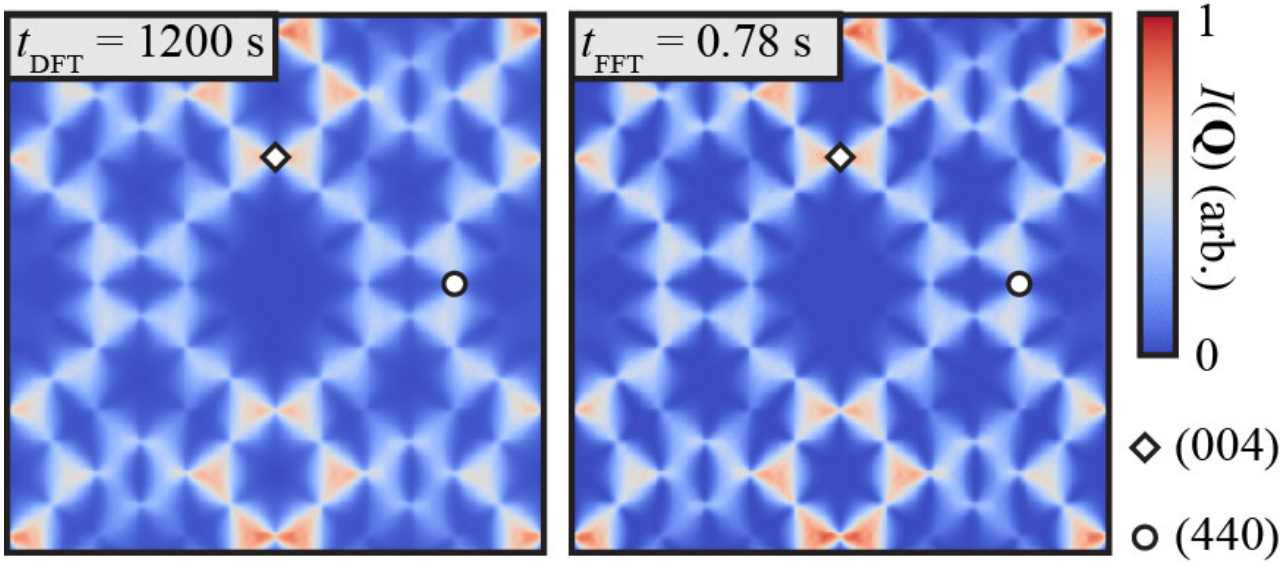
Butler & Welberry, *J Appl Cryst* **25**, 391 (1992)

Ultrafast calculation of 3D diffuse scattering

- Accelerate 3D diffuse-scattering calculations using two results
  - Average structure always periodic → FFT **can** be used
  - Nyquist theorem → complete scattering pattern can be reconstructed from supercell Bragg intensities



water ice



(110) plane*
DFT + sub-boxes

(110) plane*
FFT + filtering

Paddison, *Acta Cryst A* **75**, 14-24 (2019)

Ultrafast calculation of 3D diffuse scattering

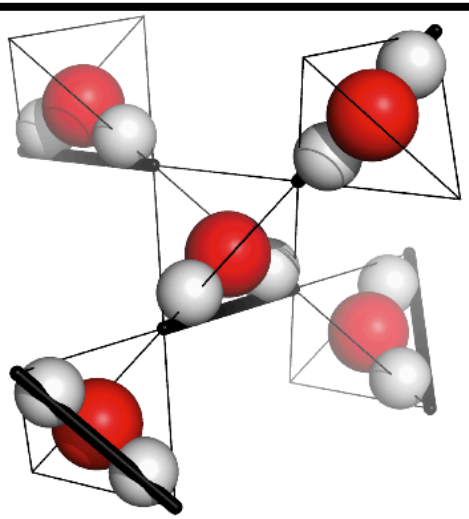
Acta Cryst A FOUNDATIONS ADVANCES

ISSN 2053-2733

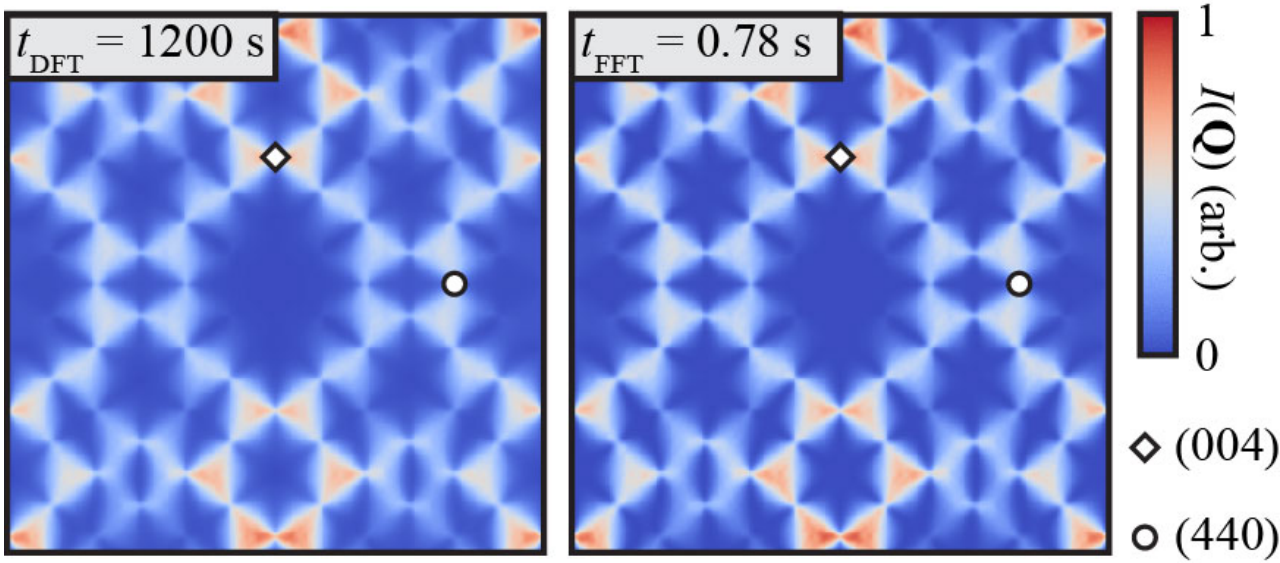
Ultrafast calculation of diffuse scattering from atomistic models

Joseph A. M. Paddison*

paddisongroup.wordpress.com/software



water ice



(110) plane*
DFT + sub-boxes

(110) plane*
FFT + filtering

Paddison, *Acta Cryst A* **75**, 14-24 (2019)
Roth *et al*, *IUCrJ* **5**, 410 (2018)

Thanks for listening!

Andrew Goodwin, University of Oxford, UK
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Henrik Jacobsen, University of Copenhagen, Denmark & ESS, Sweden
Maria Teresa Fernández-Díaz, ILL, France
Oleg Petrenko, University of Warwick, UK
Matthias Gutmann, STFC-ISIS, UK
Dave Keen, STFC-ISIS, UK
Matt Tucker, Oak Ridge National Lab, USA

UNIVERSITY OF
CAMBRIDGE

Useful info

paddisongroup.wordpress.com/software

email: jamp3@cam.ac.uk or paddison@gatech.edu

Software references:

Spinvert: Paddison, Stewart, and Goodwin, *JPCM* **25,** 454220 (2013)
Scatty: Paddison, *Acta Cryst A* **75**, 14-24 (2019)

Examples of use:

Paddison *et al.*, *Nature Commun.* **7**, 13842 (2016)
Paddison *et al.*, *Science* **350,** 179 (2015)
Nilsen *et al.*, *Phys. Rev. B* **91**, 054415 (2015)
Saines *et al.*, *Materials Horizons* **2**, 528 (2015)
Paddison *et al.*, *Phys. Rev. B* **90**, 014411 (2014)

Bonus – Absolute powder data normalization

Three steps

1) Rietveld (Fullprof) refinement to nuclear profile to determine refined scale factor, FullprofScale
2) AbsScale = FullprofScale×MagicNumber
3) NormalizedData = RawData/AbsScale

The small print: Assumes absorption, preferred orientation, and other special intensity corrections **not** included in Fullprof model. Also, site occupancy Occ = (occupancy of special position)/(occupancy of general position).

Rodríguez-Carvajal, *Physica B* **192**, 55 (1993)

Bonus – Fullprof "magic numbers"

(to normalize intensity in units barn sr^{-1} $spin^{-1}$)

Constant-wavelength diffraction

$$\text{MagicNumber} = \frac{2\pi^2 NV}{45\lambda^3}$$

where *N* is number of spins per unit cell, *V* is unit-cell volume in A^3, and λ is neutron wavelength in A.

Time-of-flight diffraction

$$\text{MagicNumber} = \frac{4\pi NV \sin\theta}{\texttt{DIFC}}$$

where **θ** is scattering angle of detector and `DIFC` (sometimes called `dtt1`) is instrument parameter relating to TOF in *μs* to *d*-spacing in A.