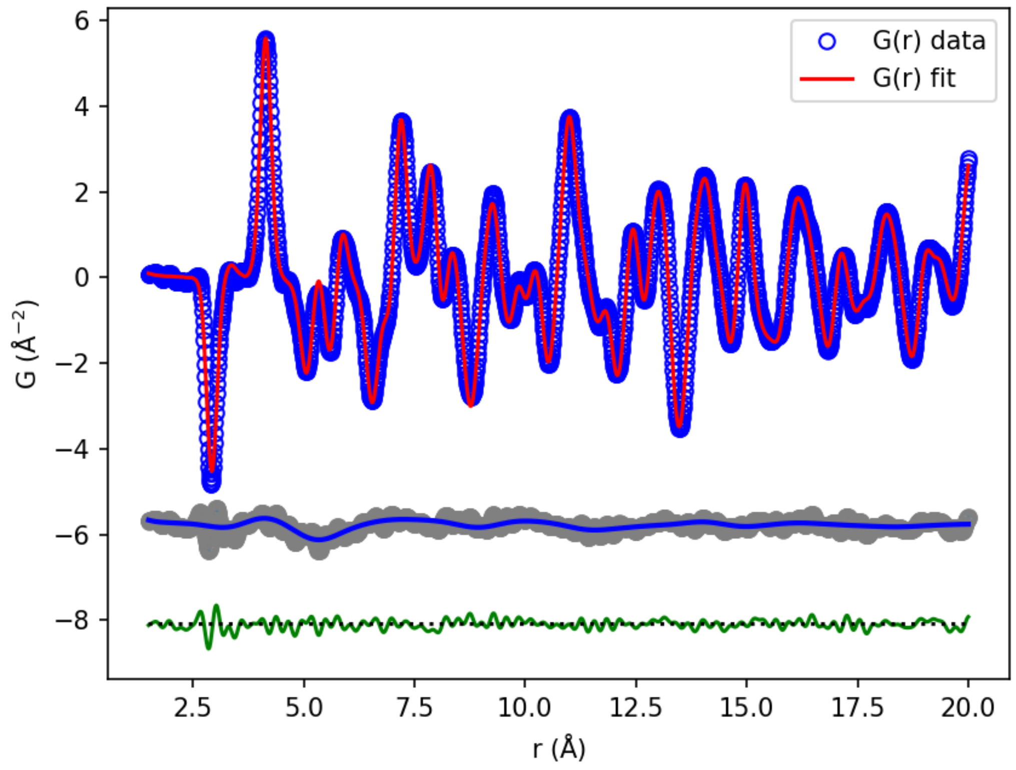
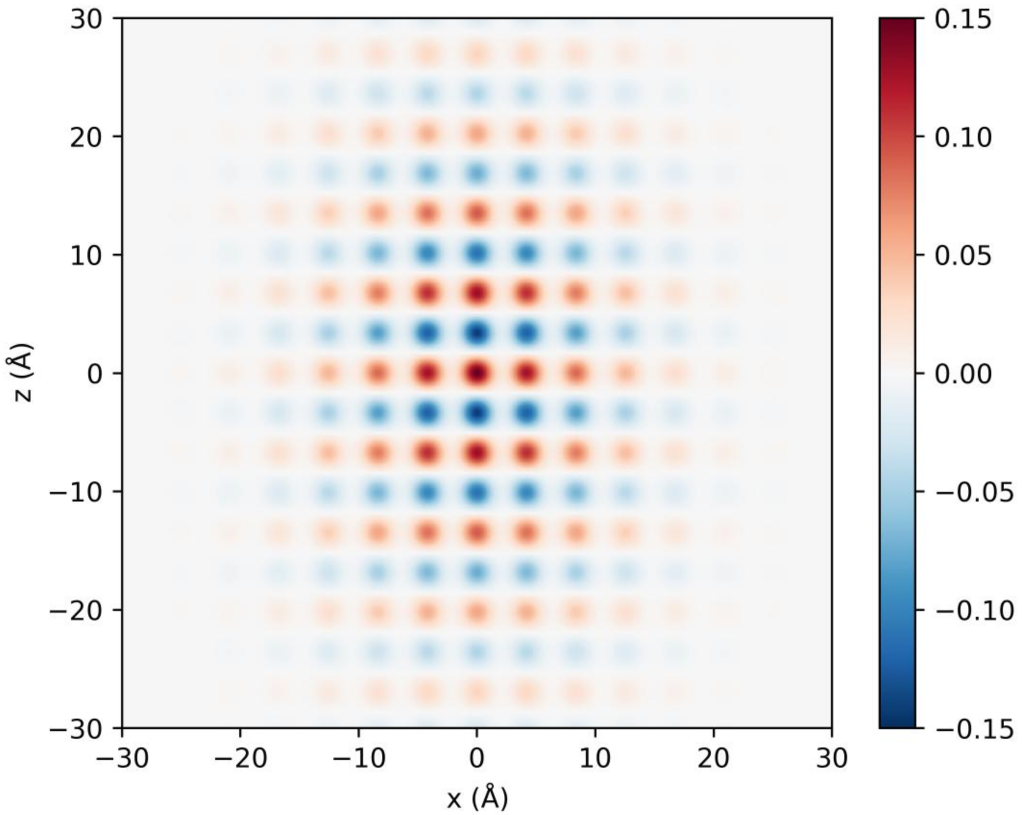
An introduction to magnetic PDF



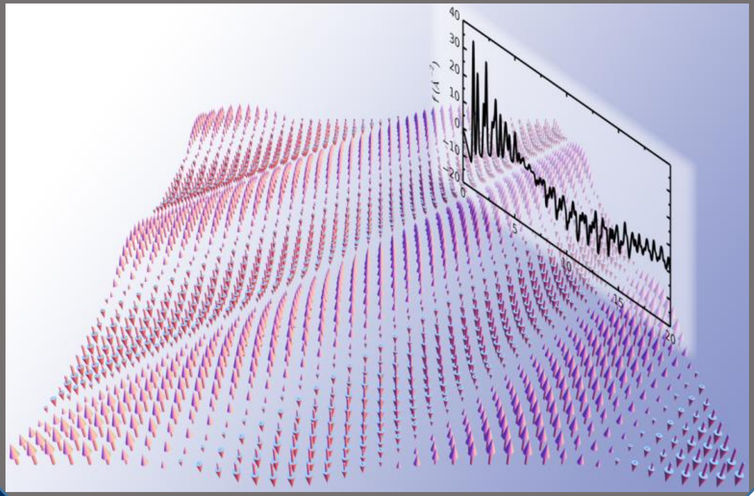


Ben Frandsen, Brigham Young University

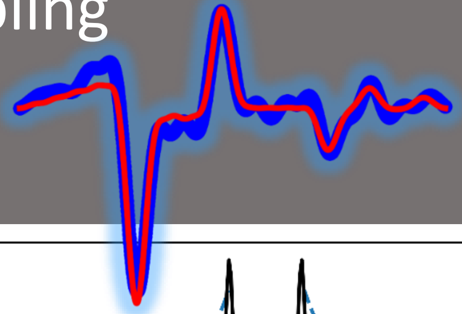
ADD2026, 11-16 January 2026, Grenoble

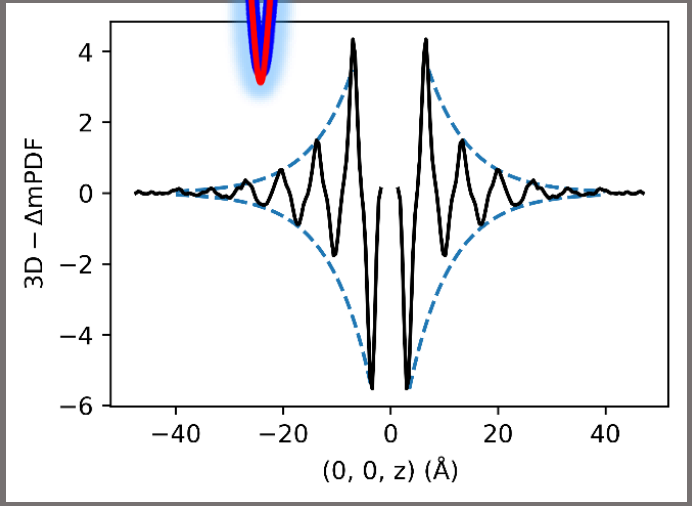
Outline

Brief introduction to magnetic pair distribution function (mPDF) analysis as a means of studying short-range magnetic correlations



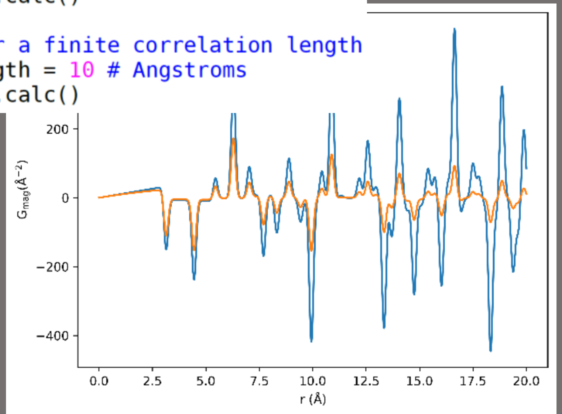
MnTe as a Case Study: Short-range spin correlations, magnetostructural coupling





Overview of diffpy.mpdf python package—come to the tutorial for more information!

```
from diffpy.mpdf import *

# read in the MCIF
mcif = '1.31_MnO.mcif'
mstruc = create_from_mcif(mcif)

# generate spin arrays
mstruc.makeAll()

# calculate the mPDF
mcalc = MPDFcalculator(mstruc)
r1, g1 = mcalc.calc()

# calculate for a finite correlation length
mstruc.corrLength = 10 # Angstroms
r2, g2 = mcalc.calc()
```



Acknowledgements

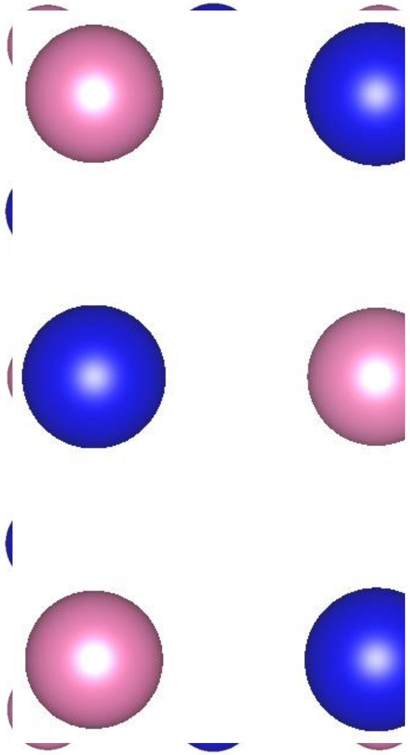
- Original Columbia team: Simon Billinge, Xiaohao Yang
- Facility scientists: Henry Fischer, Jue Liu, Cheng Li, Michelle Everett, Feng Ye, Ovi Garlea
- Other researchers contributing to mPDF: Henry Fischer, Nikolaj Roth, Katsuaki Kodama, Shin-ichi Shamoto, Yuanpeng Zhang, Matt Tucker, Raju Baral, Stuart Calder, Andrew Wildes, and more…
- BYU students: Jacob Christensen, Parker Hamilton, Christiana Zaugg, Emma Zappala, Sabrina Hatt, Edison Carlisle, Eric Stubben
- Funding: U.S. Department of Energy Early Career Award

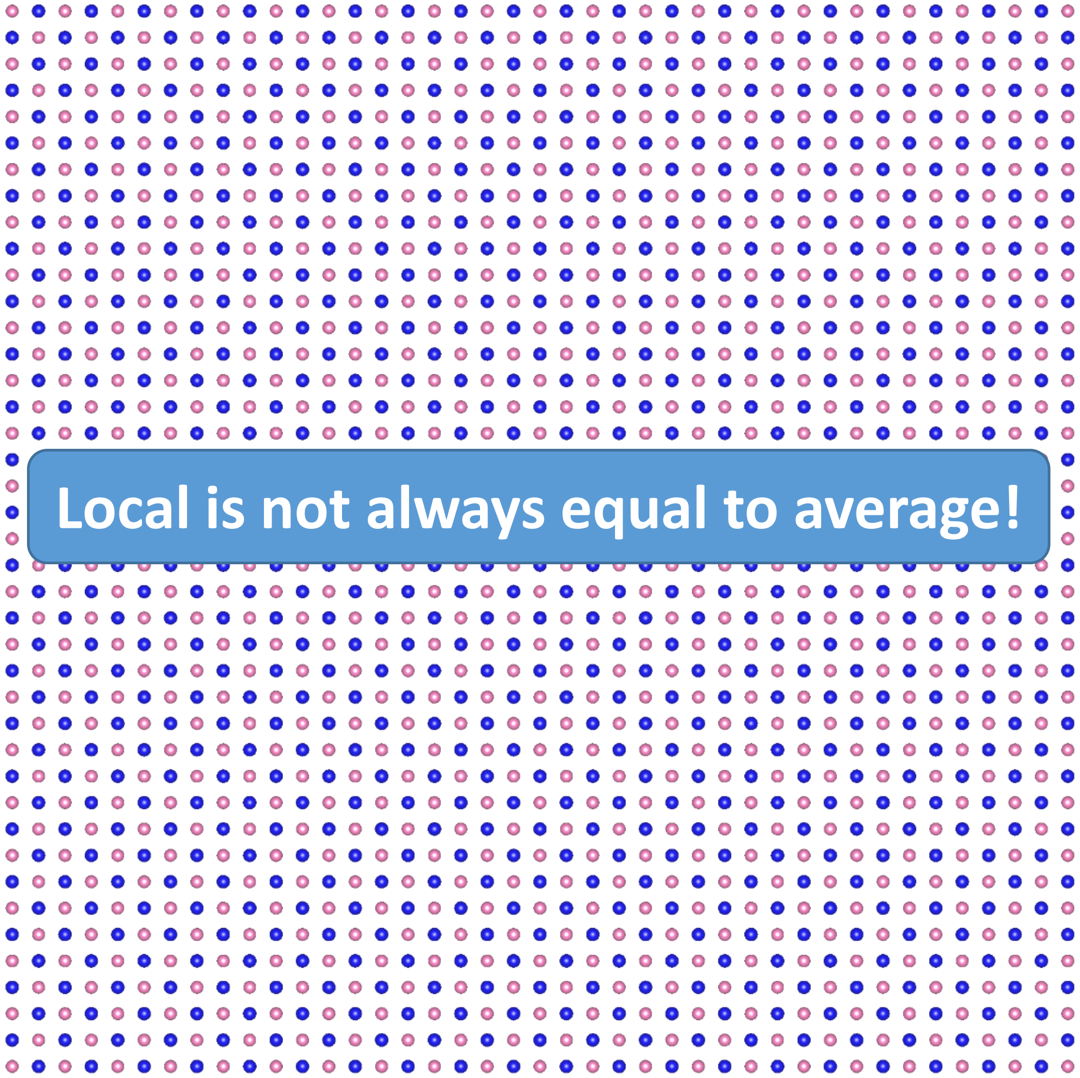
Part I: Introduction to magnetic pair distribution function analysis

Local Structure versus Average Structure

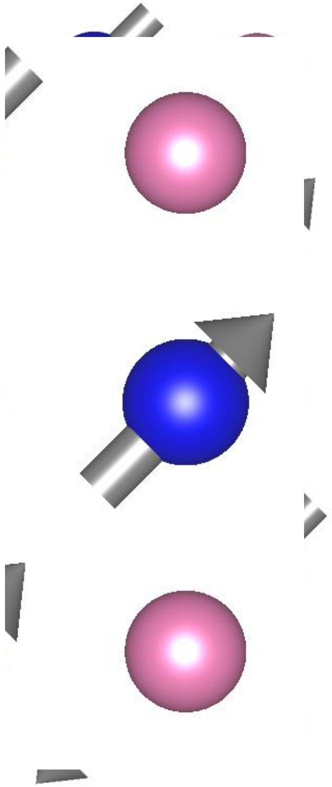
Structure of a material: The arrangement of its constituent parts

Atomic structure





Local is not always equal to average!

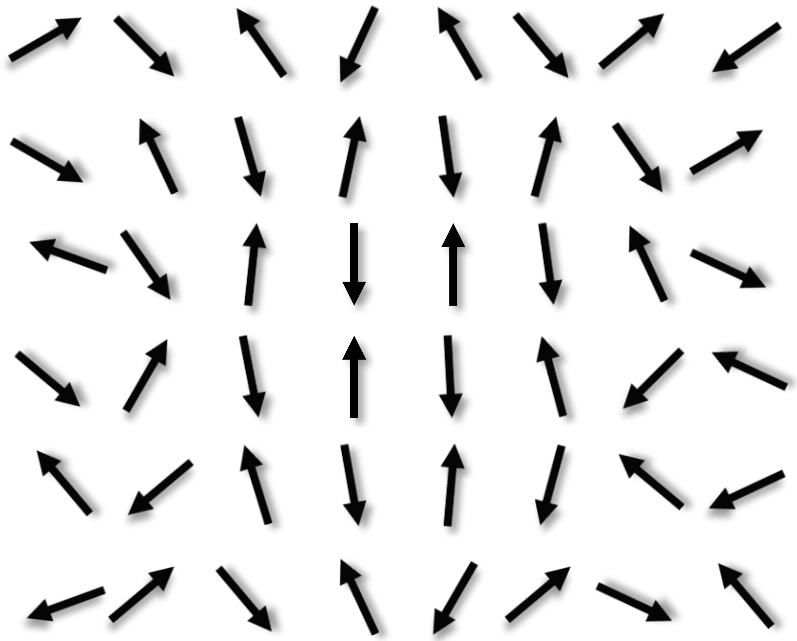


Magnetic structure

Local structure

Average structure

Purpose: Study short-range magnetic order

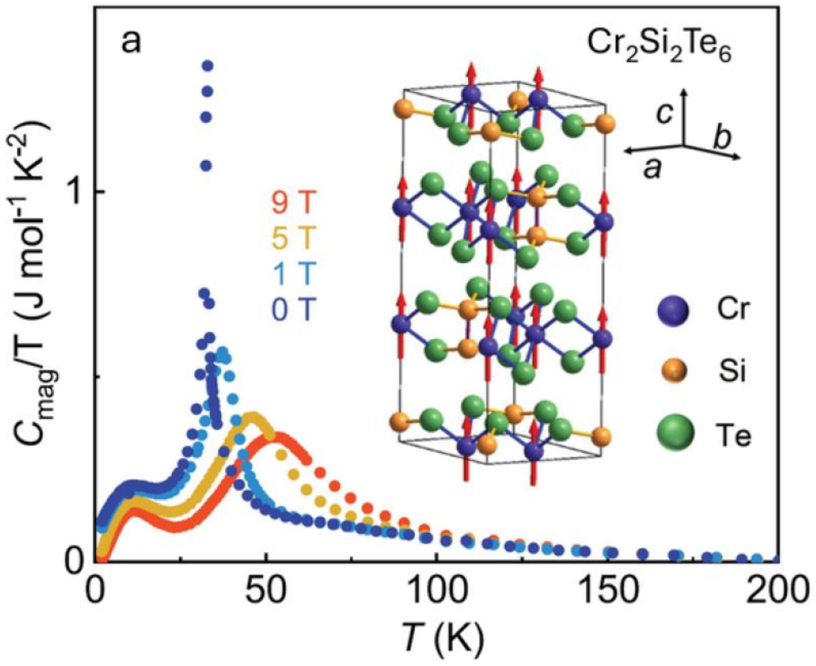


Well-defined correlations only over a finite separation distance, e.g. a few interatomic spacings

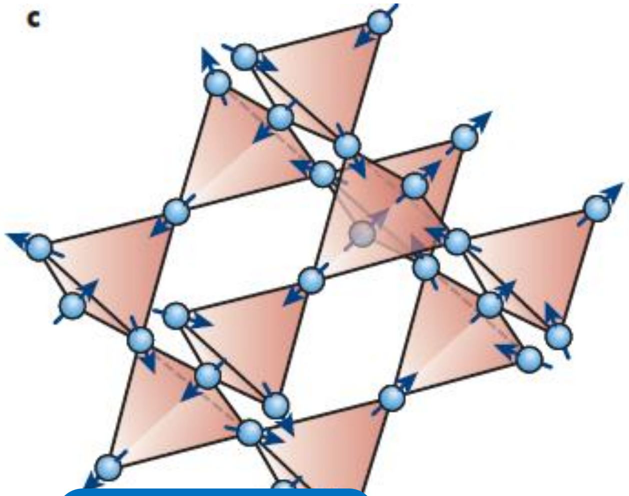
$$\langle S_0 \cdot S_i \rangle(r) \sim e^{-r/\xi}$$

Short-range magnetic order is ubiquitous in modern magnetism

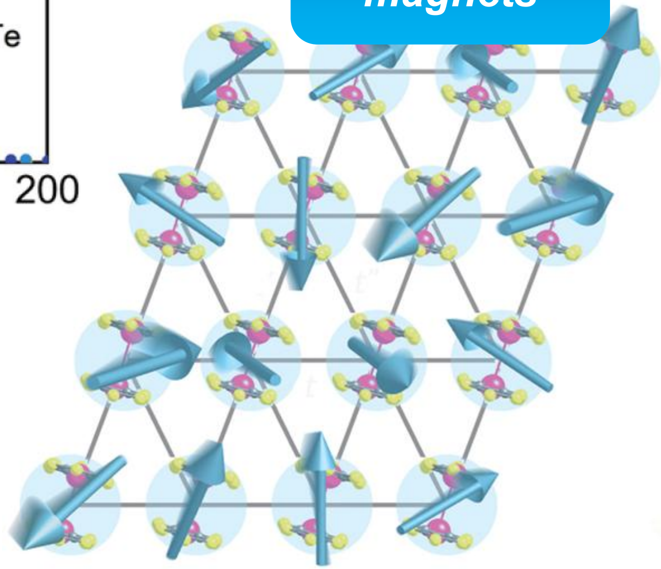
Quantum materials



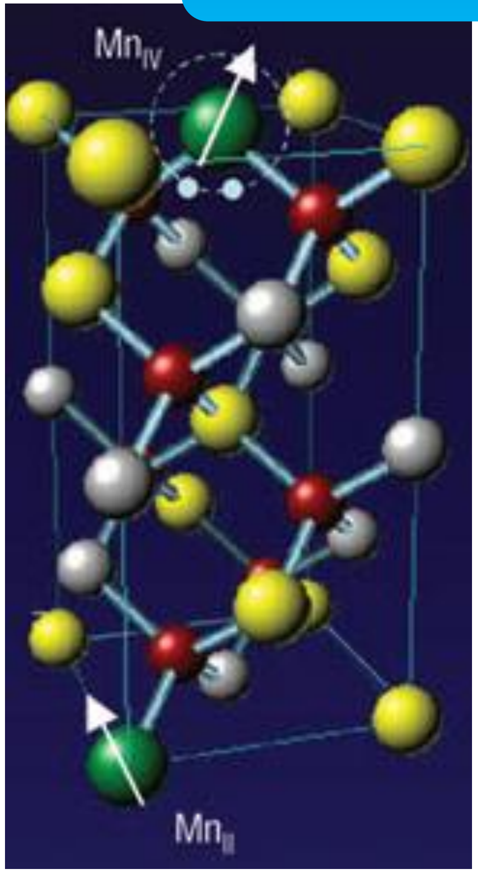
Yang et al, Adv. Funct. Mater. **2023**, 33, 2302191



Frustrated magnets

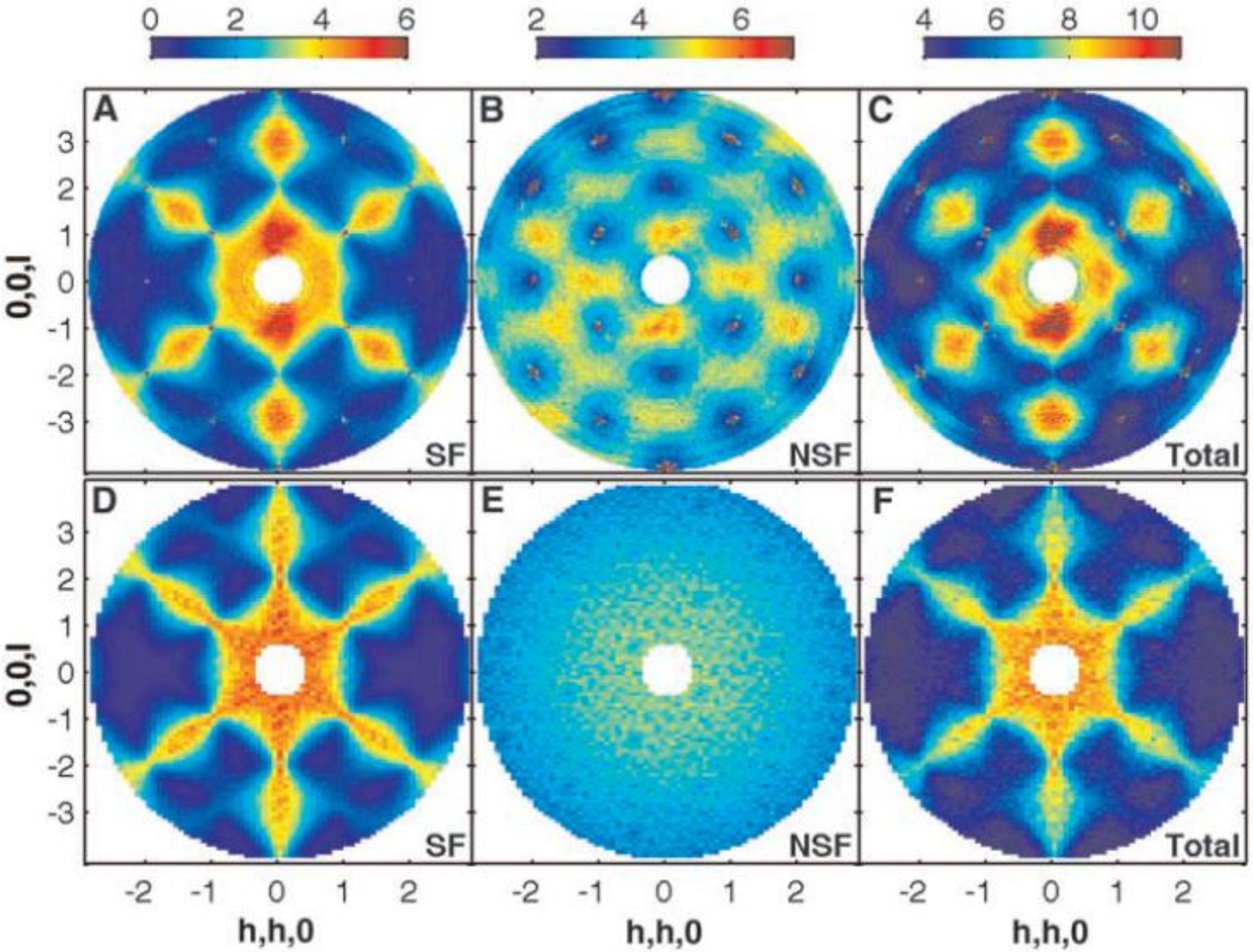


Magnetofunctional materials



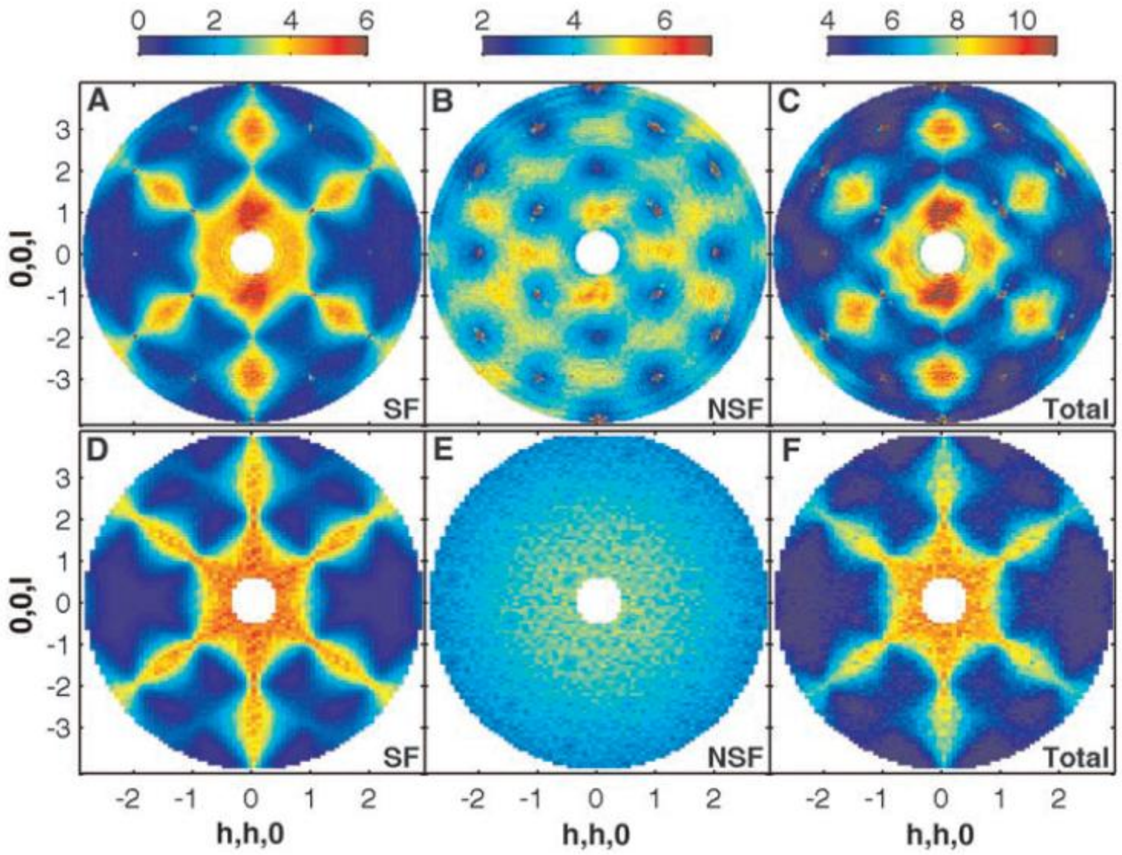
Many more examples!!

Neutron scattering from magnetic short-range order

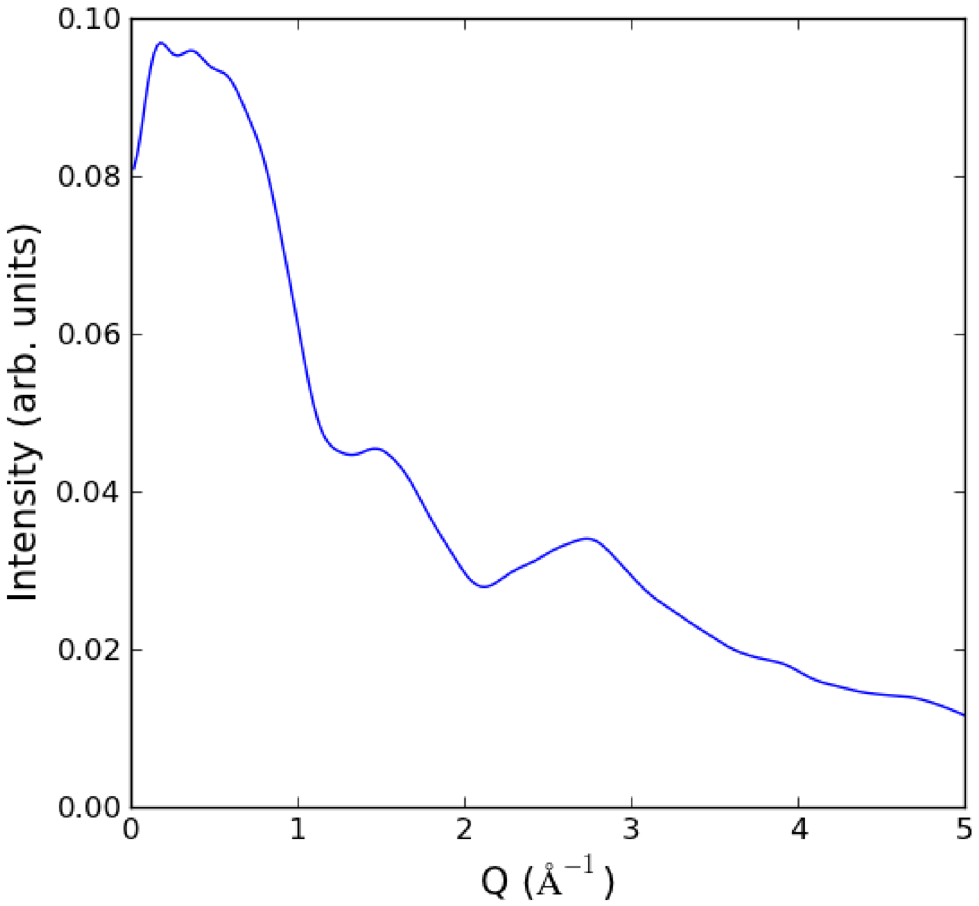


Fennel *et al*, Science **326** 415 (2009)

Neutron scattering from magnetic short-range order



Fennel *et al*, Science **326** 415 (2009)



Simulated powder data from Joe Paddison

Real-space approach: PDF analysis

Scattering from **atoms and nuclei** → Fourier transform → **Atomic PDF**

Scattering from **magnetic moments** → Fourier transform → **Magnetic PDF**

Scattering from **nuclei & magnetic moments** → Fourier transform → **Combined atomic and magnetic PDF**

Magnetic PDF

$$\left.\frac{1}{N}\frac{d\sigma}{d\Omega}\right|_{\mathrm{m}} = p^2\mu_{\mathrm{m}}^2 f_{\mathrm{m}}^2(q)\left\{\frac{2}{3} + \frac{1}{N}\sum_{i\neq j}\left[\hat{A}_{ij}\frac{\sin(qr_{ij})}{qr_{ij}} + \hat{B}_{ij}\left(\frac{\sin(qr_{ij})}{(qr_{ij})^3} - \frac{\cos(qr_{ij})}{(qr_{ij})^2}\right)\right]\right\}$$

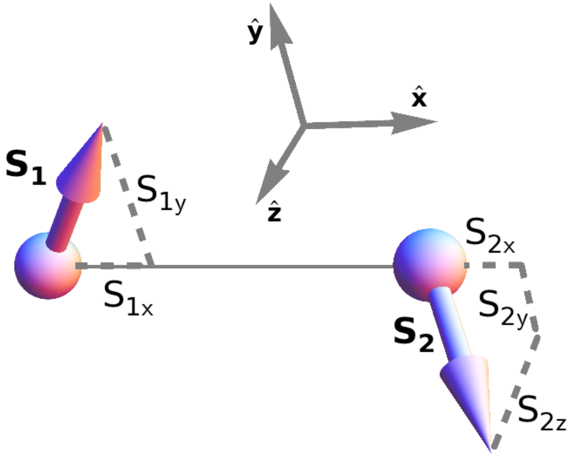
Fourier transform

$$\mathrm{mPDF}(r) \approx \frac{3}{2}\frac{1}{N}\sum_{i\neq j}\left[\frac{\hat{A}_{ij}}{r}\tilde{\delta}(r - r_{ij}) + \hat{B}_{ij}\frac{r}{r_{ij}^3}\tilde{\Theta}(r_{ij} - r)\right] - 4\pi\, r\rho_{\mathrm{m}}|\langle\hat{\boldsymbol{\mu}}_{\mathrm{m}}\rangle|^2$$

Peaks at spin separation distances, overall 1/*r* envelope

Truncated linear term

Sign and strength determined by local spin correlations



Transverse correlations (peak height)

$$A_{ij} = S_i^y S_j^y$$

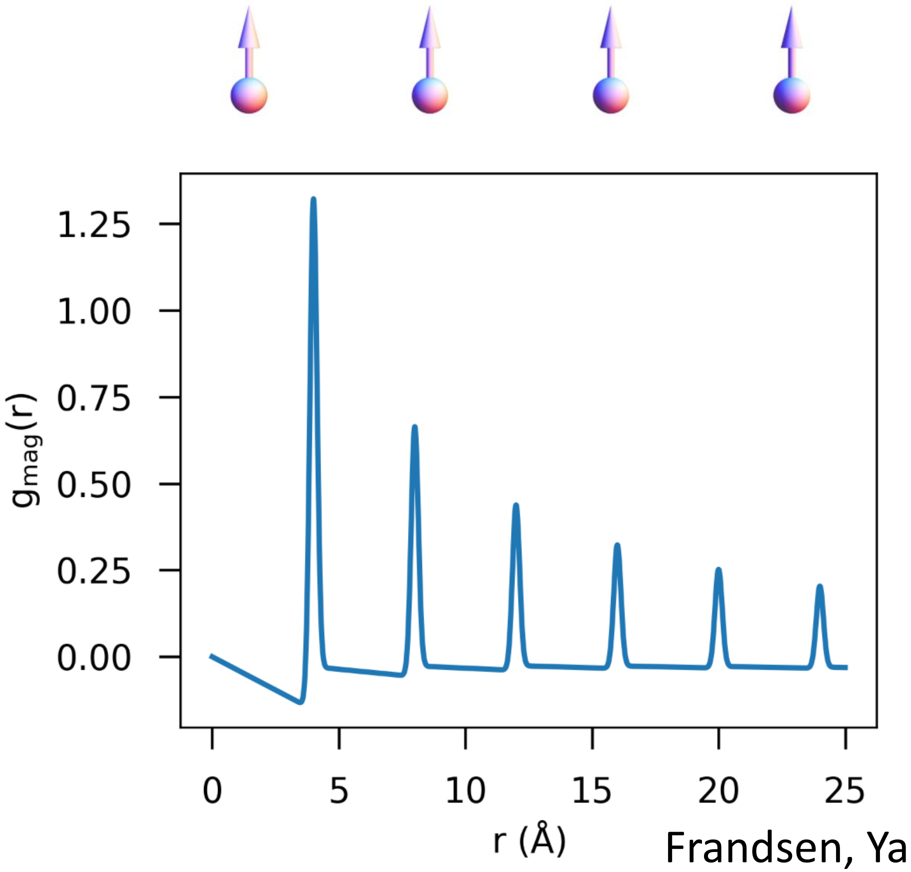
Transverse and longitudinal correlations (baseline)

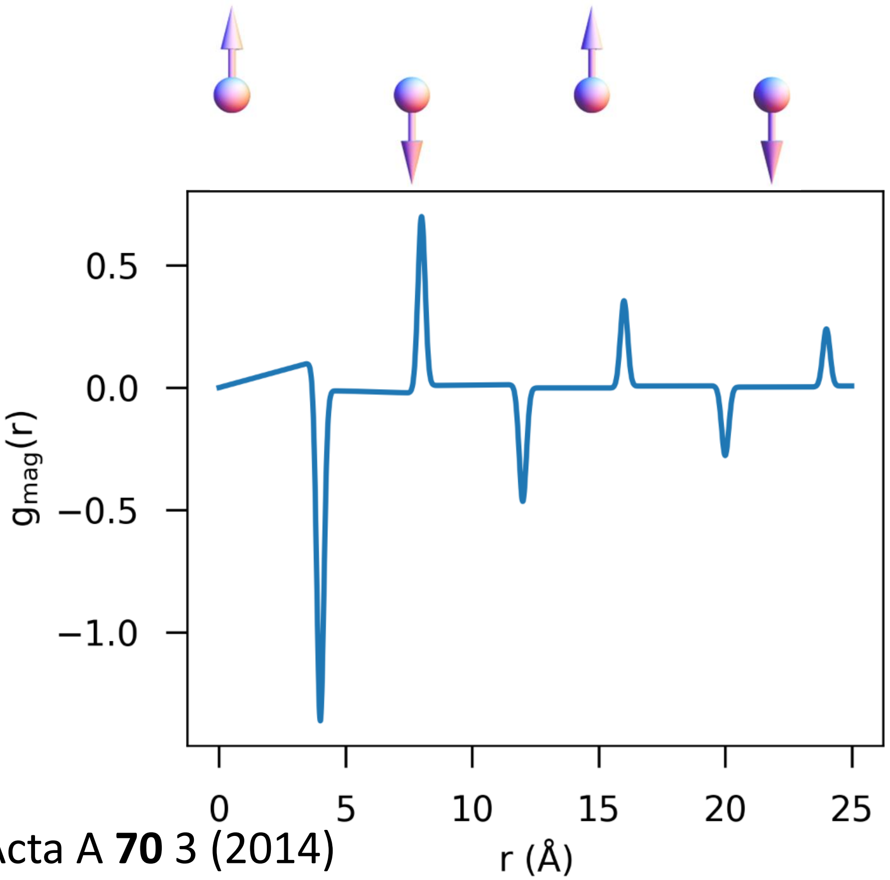
$$B_{ij} = 2S_i^x S_j^x - S_i^y S_j^y$$

Frandsen, Yang, & Billinge, Acta A **70** 3 (2014)
Frandsen and Fischer, Chem Mater **36** 9089 (2024)

Example: One-dimensional chain

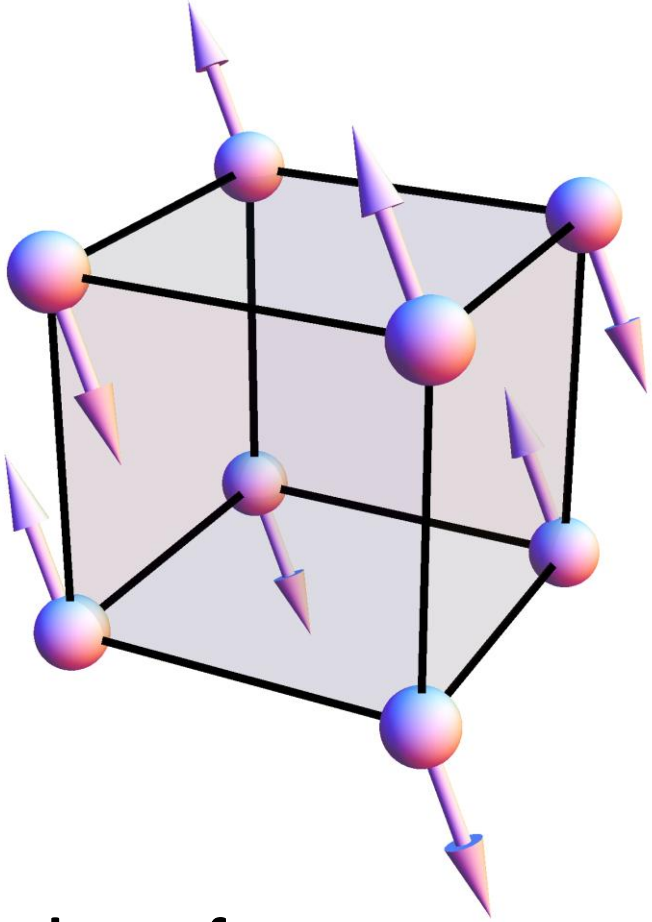
$$\mathrm{mPDF}(r) \approx \frac{3}{2}\frac{1}{N}\sum_{i\neq j}\left[\frac{\hat{A}_{ij}}{r}\tilde{\delta}(r - r_{ij}) + \hat{B}_{ij}\frac{r}{r_{ij}^3}\tilde{\Theta}(r_{ij} - r)\right] - 4\pi\, r\rho_{\mathrm{m}}|\langle\hat{\boldsymbol{\mu}}_{\mathrm{m}}\rangle|^2$$

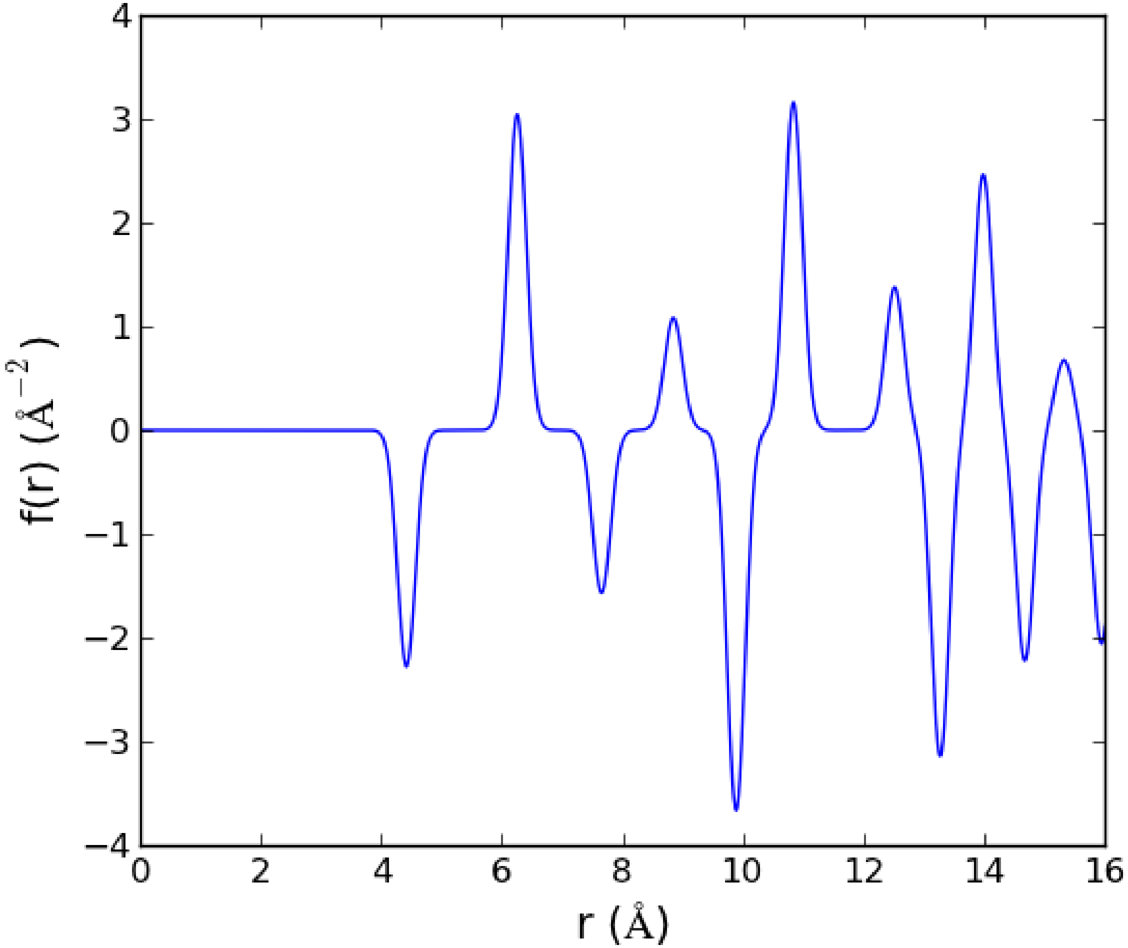




Frandsen, Yang, & Billinge, Acta A **70** 3 (2014)

Example: Simple cubic antiferromagnet

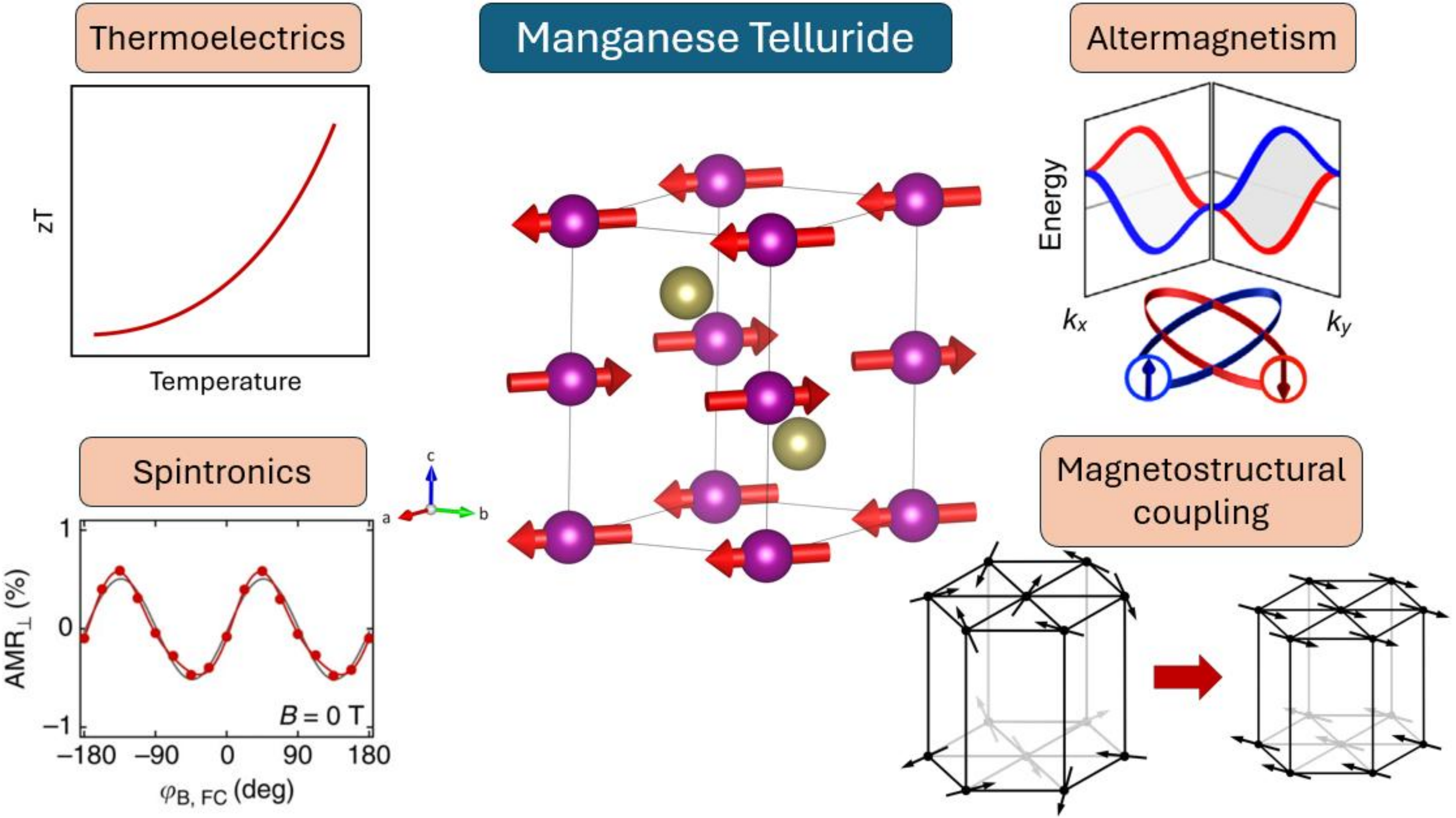




Further references

- *Acta A* **70** 3 (2014)
- *Acta A* **71** 325 (2015)
- *PRL* **116** 197204 (2016)
- *Chem Mater* **36** 9089 (2024)

Part II: MnTe as a Case Study

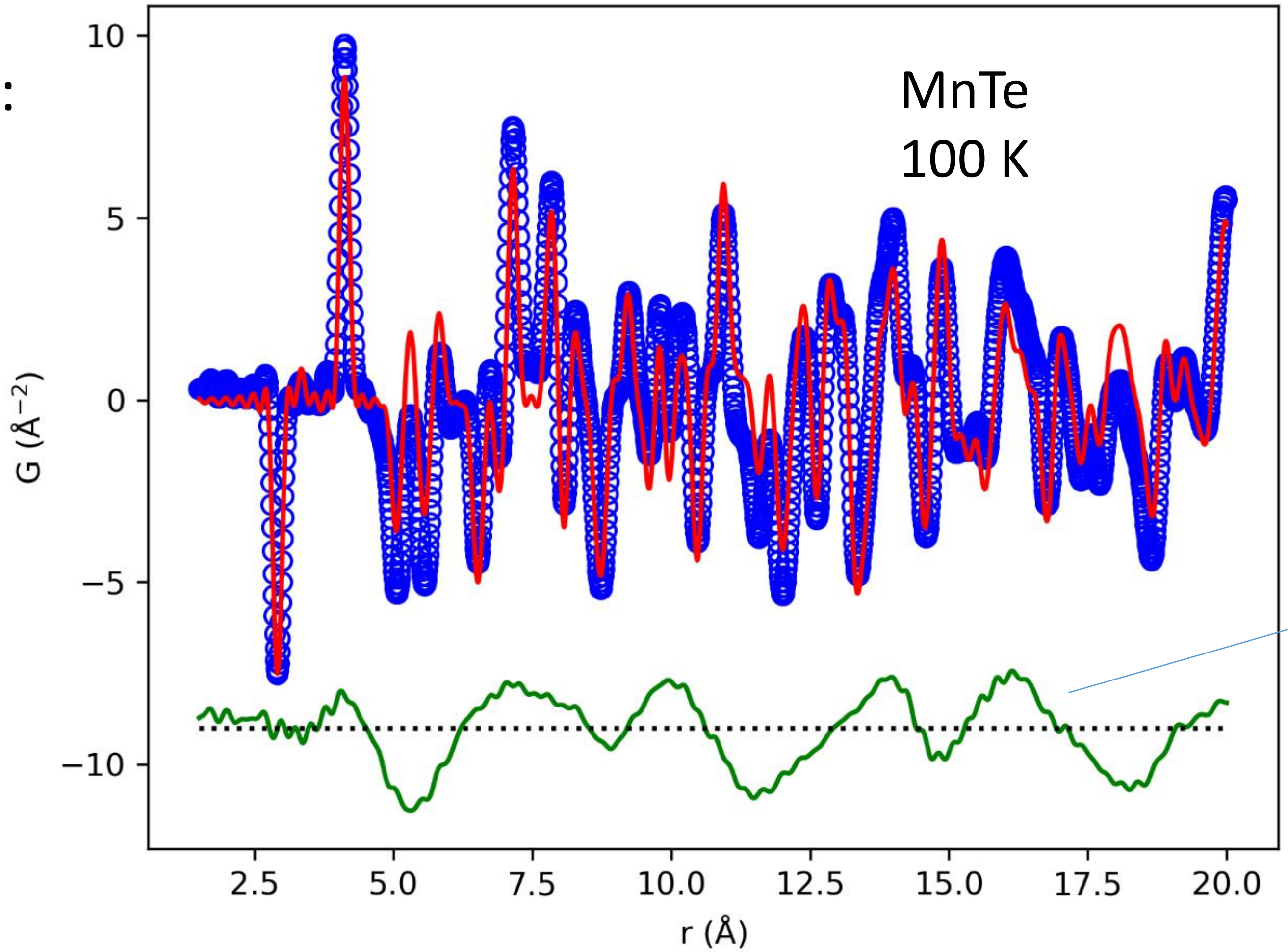


Thermoelectrics
zT
Temperature
Manganese Telluride
Altermagnetism
Energy
k_x
k_y
Spintronics
c
a
b
$AMR_{\perp}$ (%)
1
0
−1
$B = 0$ T
−180
−90
0
90
180
$\varphi_{B,\,FC}$ (deg)
Magnetostructural coupling

Neutron PDF experiment on MnTe

Standard PDFgui fit:

Has something gone horribly wrong??

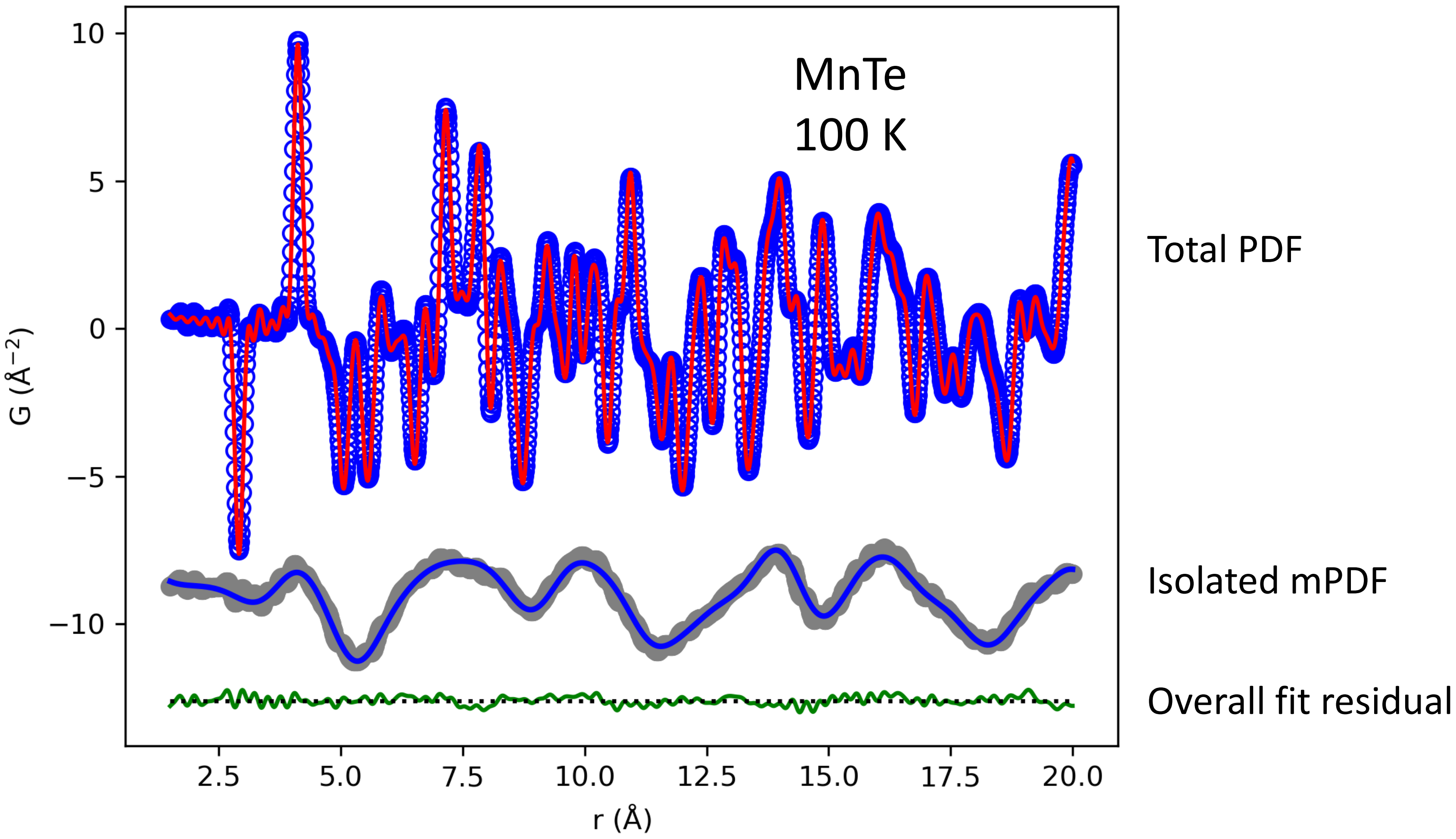


mPDF signal (not included in standard PDFgui fit)

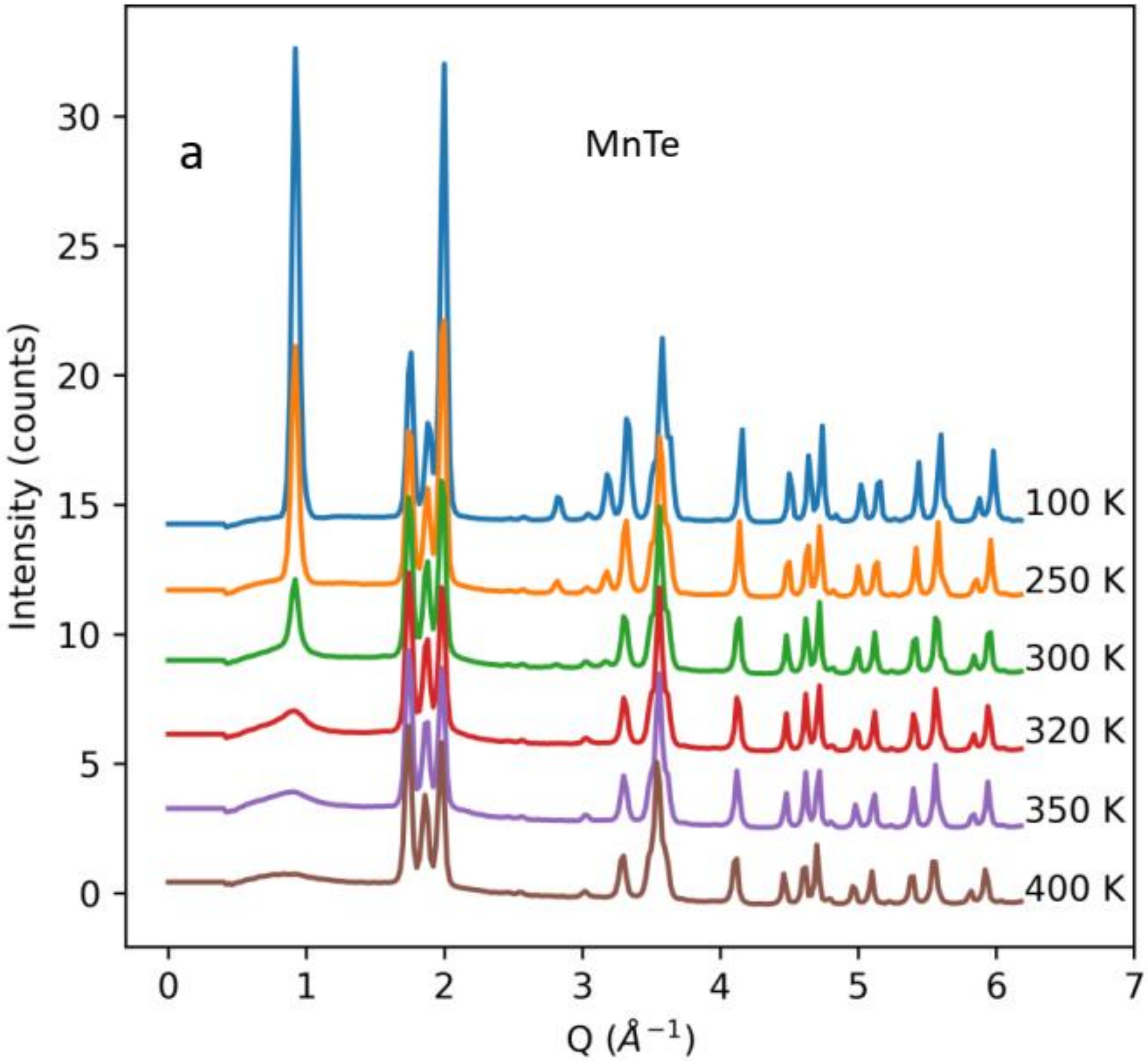
No! We just need to include the magnetic PDF in our model.

Combined atomic and magnetic PDF analysis

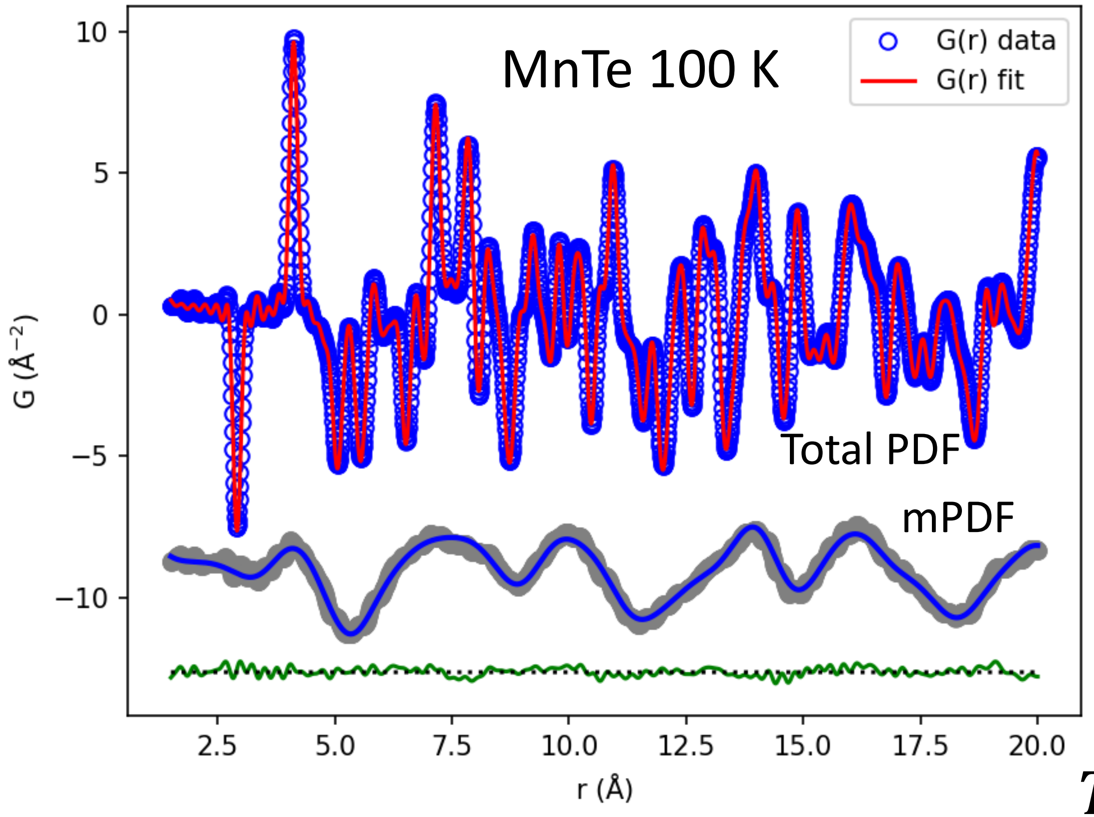
Diffpy.mpdf fit with atomic and magnetic components:



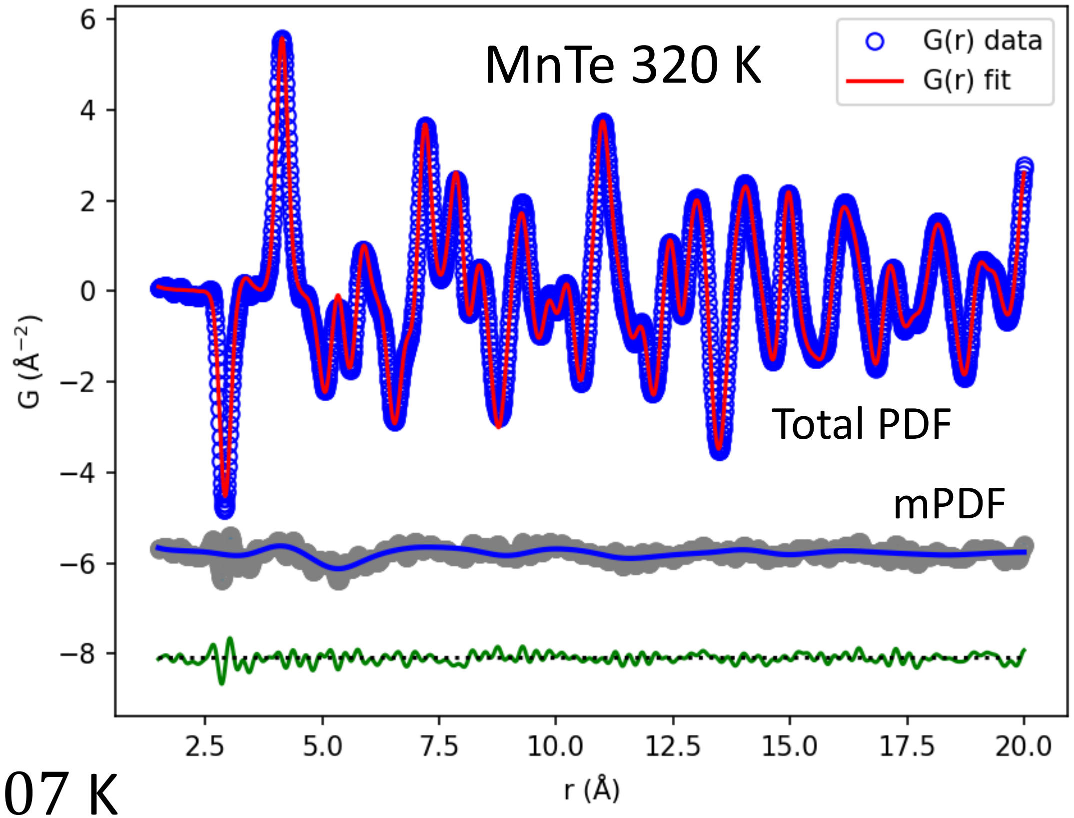
Temperature-dependent scattering signal



Long-range and short-range magnetic order in MnTe

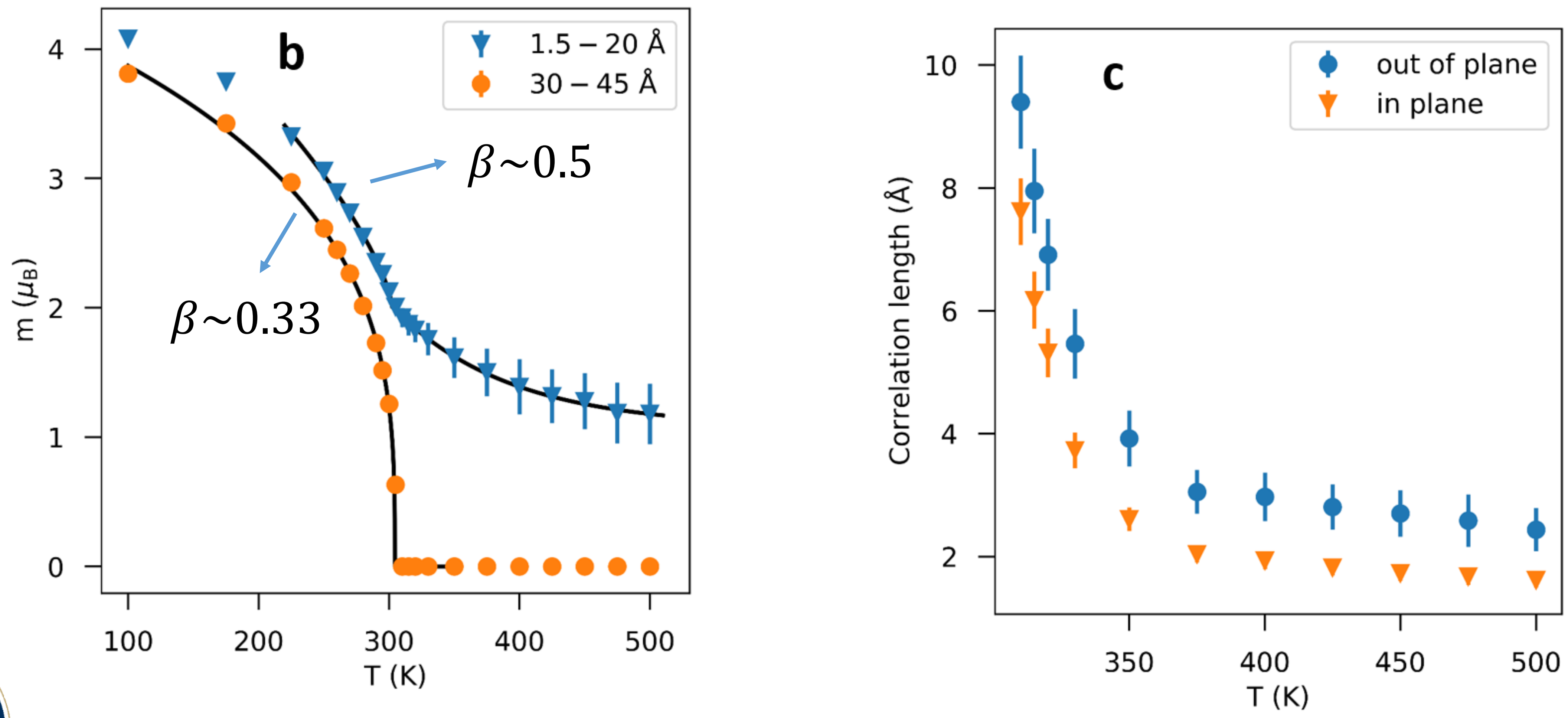


$T_N = 307$ K



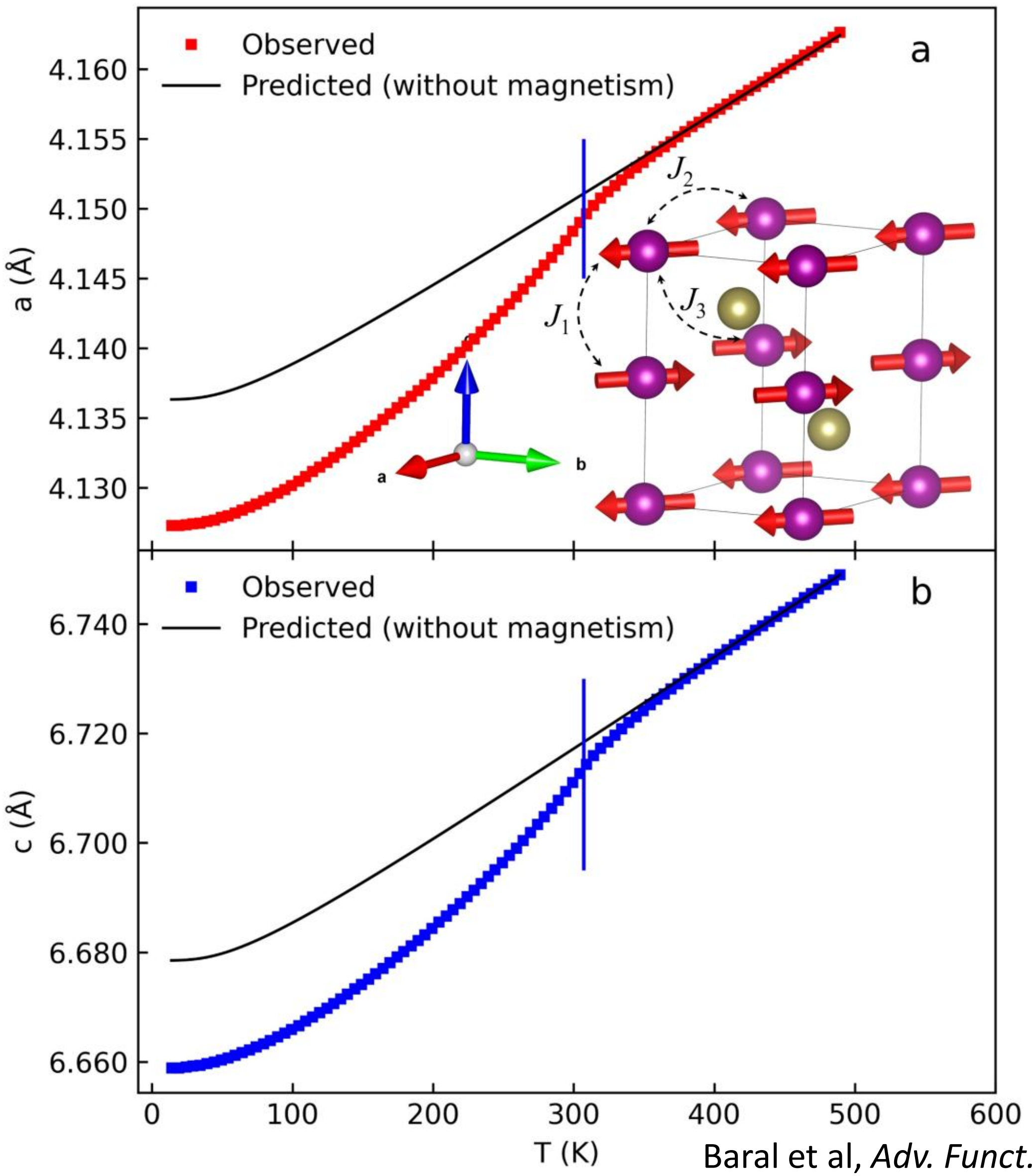
Baral et al, *Matter* **5**, 1853 – 1864 (2022)

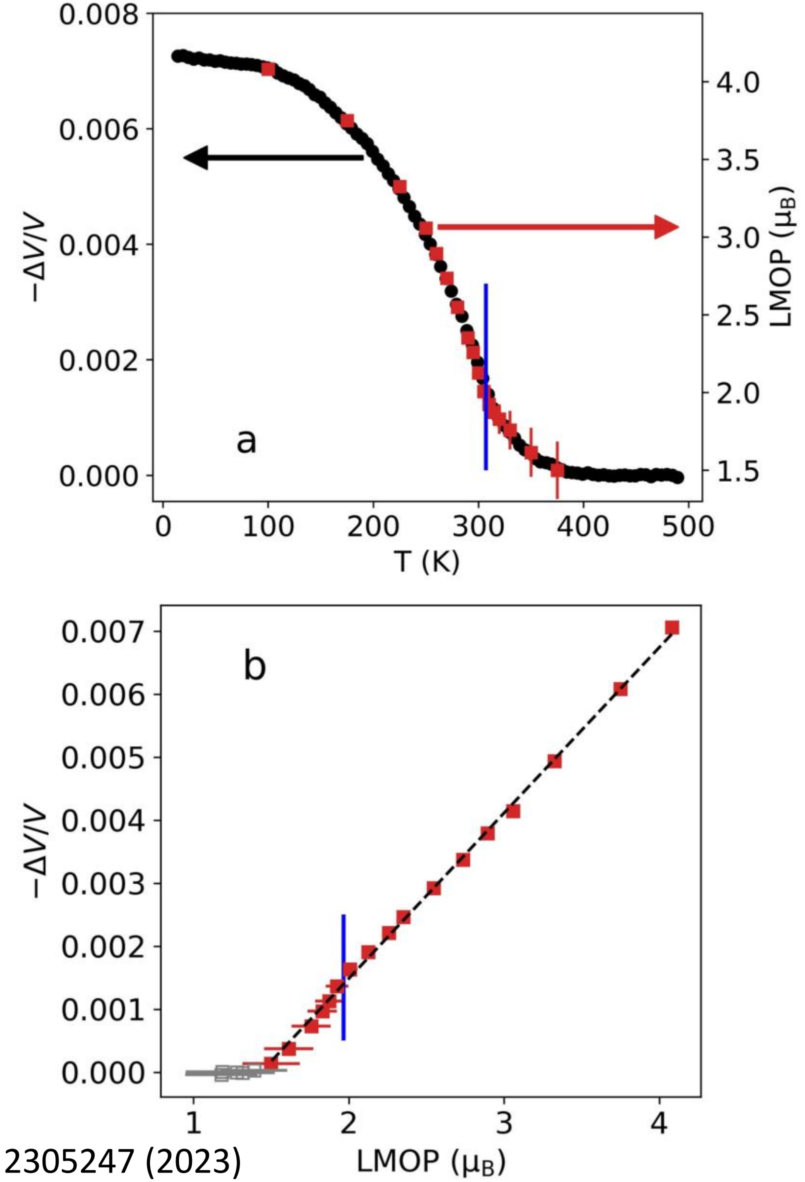
Characterizing the phase transition with mPDF



Baral et al, *Matter* **5**, 1853 – 1864 (2022)

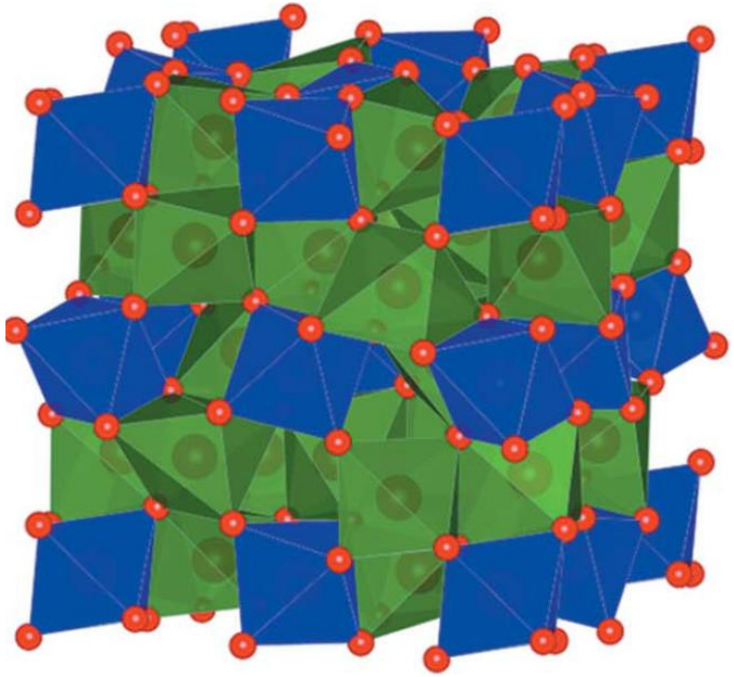
Magnetostructural coupling with combined atomic + magnetic PDF



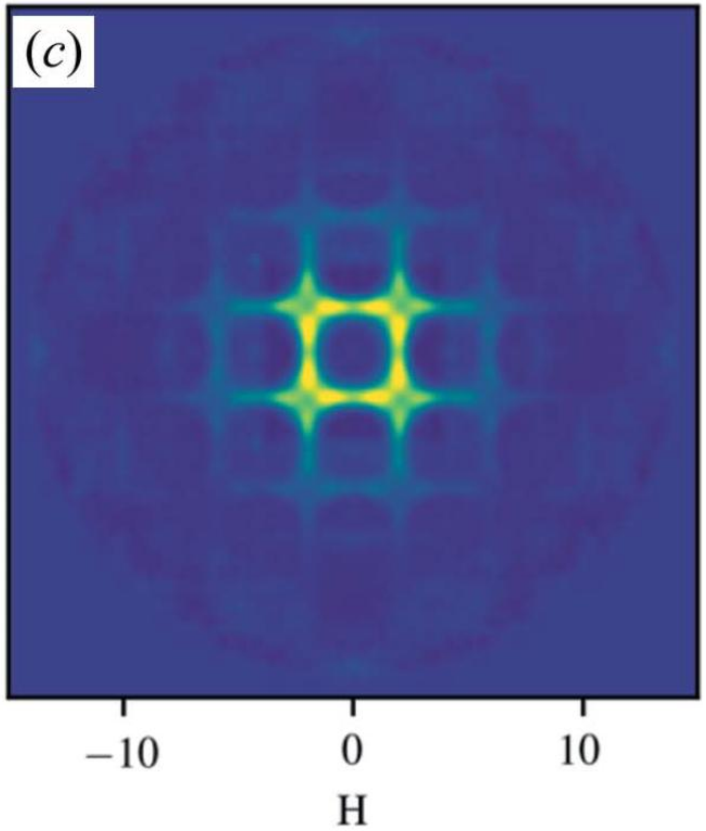


Baral et al, *Adv. Funct. Mater.* **33**, 2305247 (2023)

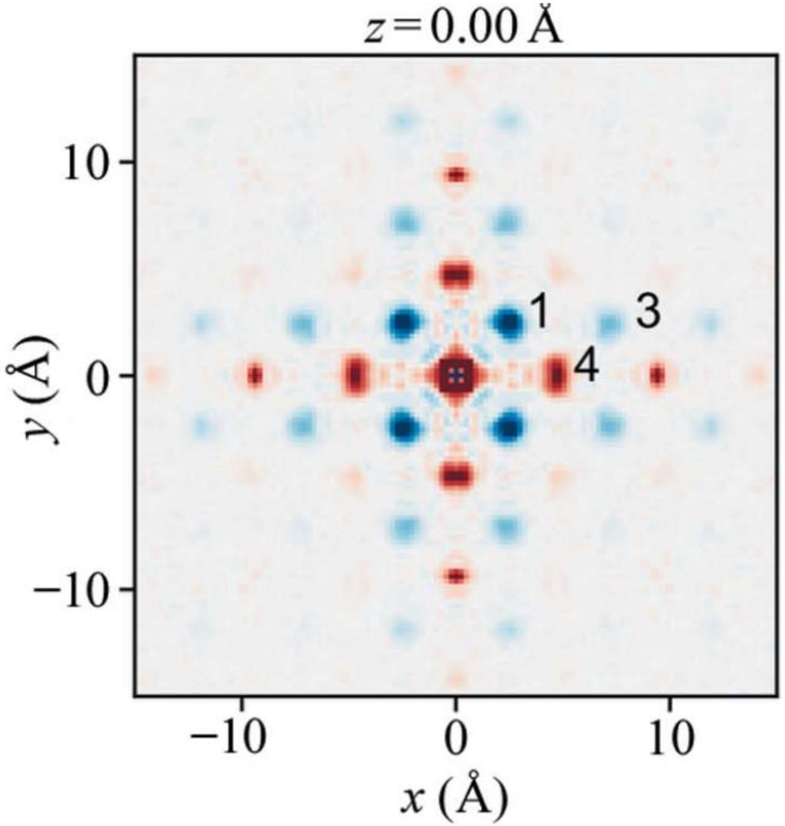
3D-ΔmPDF study of MnTe



Bixbyite, $Fe_{1.1}Mn_{0.9}O_3$



Diffuse magnetic scattering measured from a single crystal on CORELLI at SNS

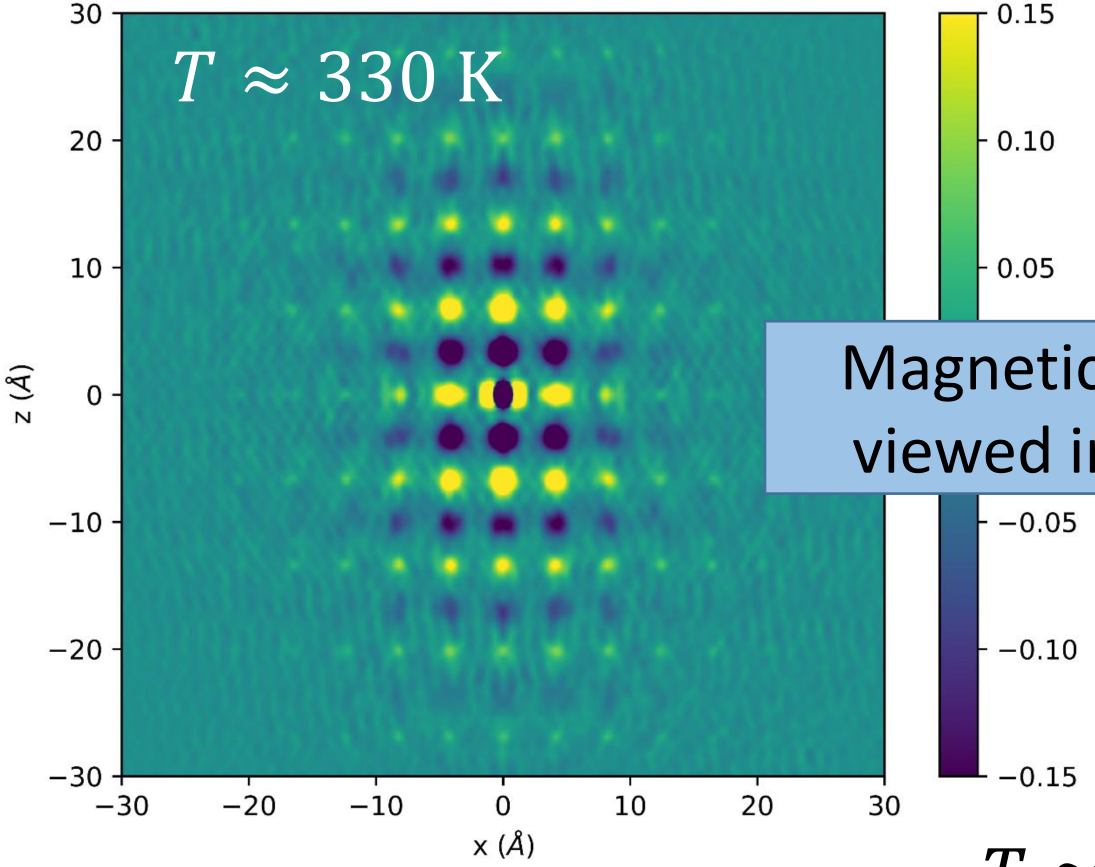


Experimentally obtained 3D-ΔmPDF

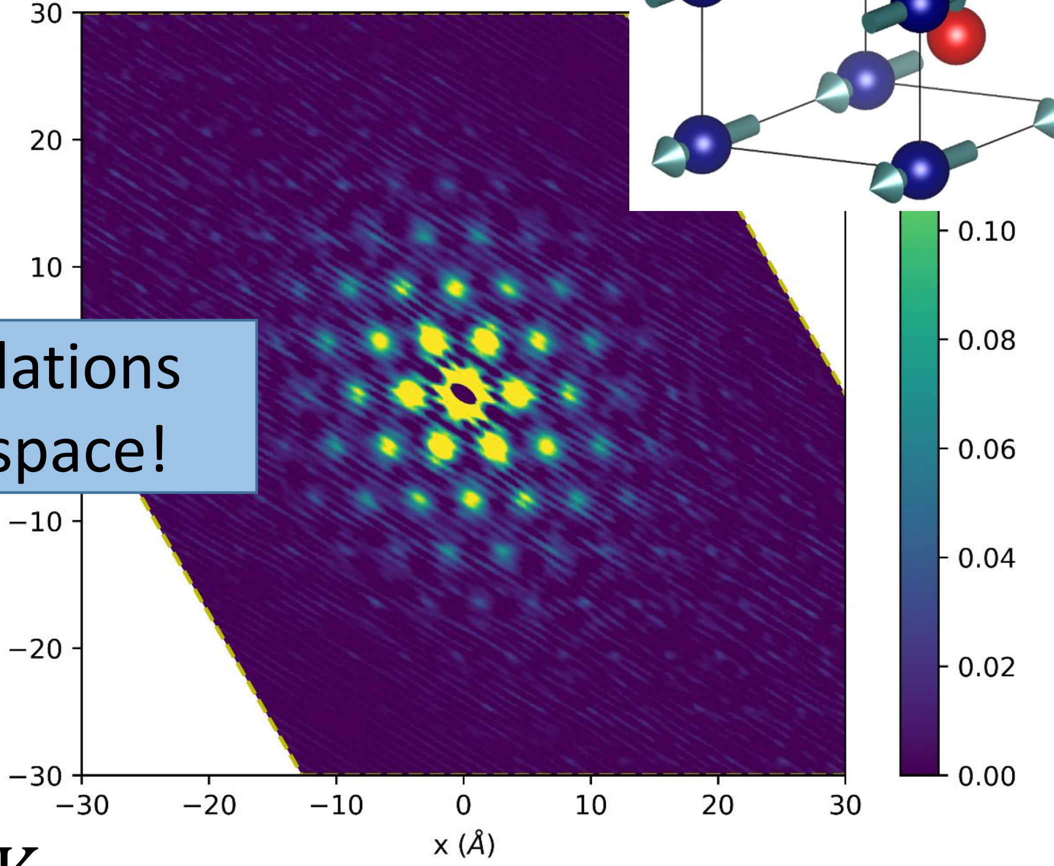
N. Roth et al, *IUCrJ* **5**, 410 (2019)

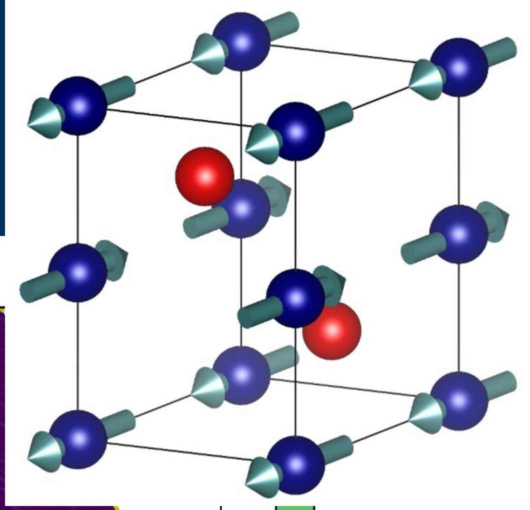
3D-ΔmPDF study of MnTe

Single-crystal measurements at CORELLI



Magnetic correlations viewed in real space!





$T \approx 330$ K

xz plane (*y* = 0)

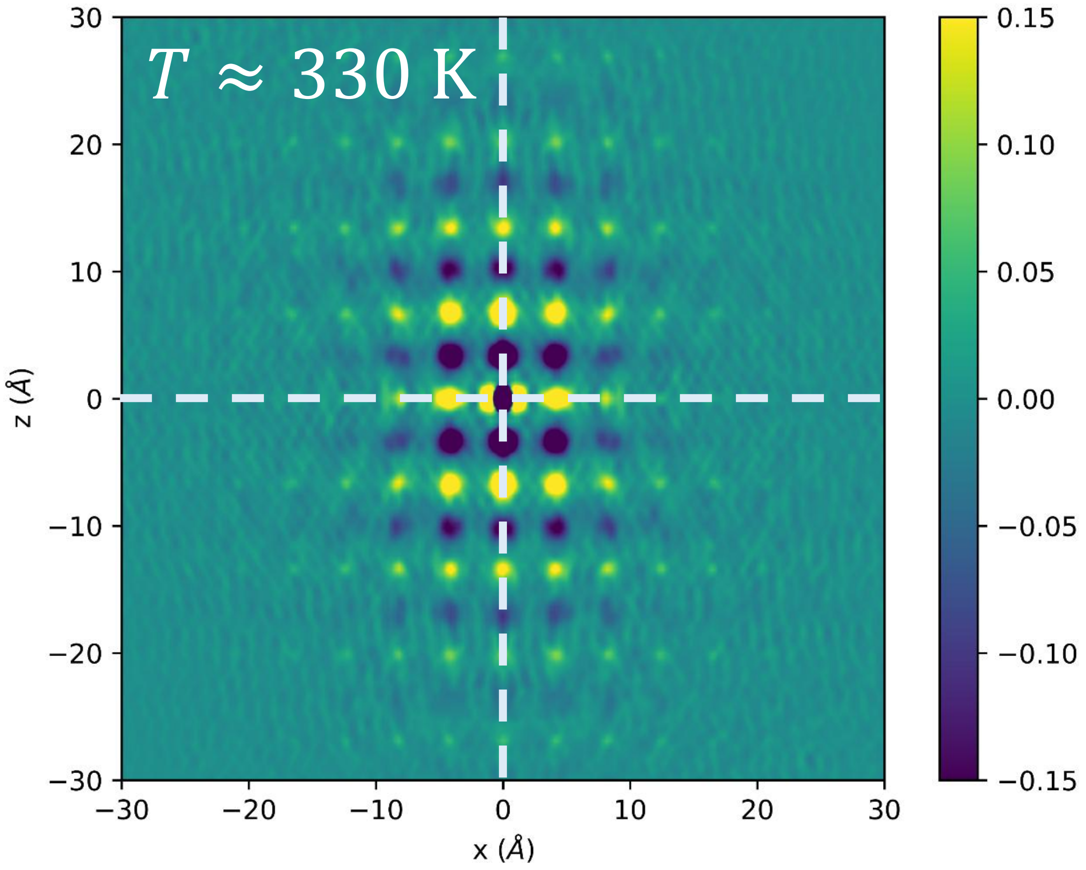
Antiferromagnetic correlations along *z*

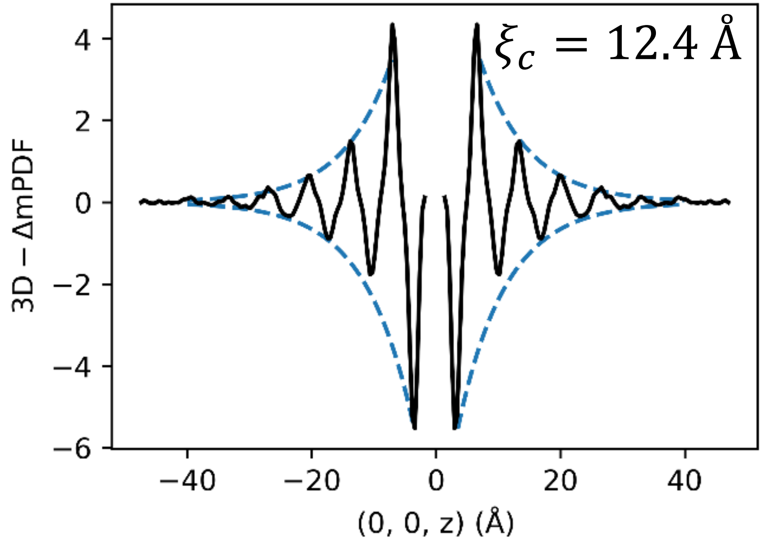
xy plane (*z* = 0)

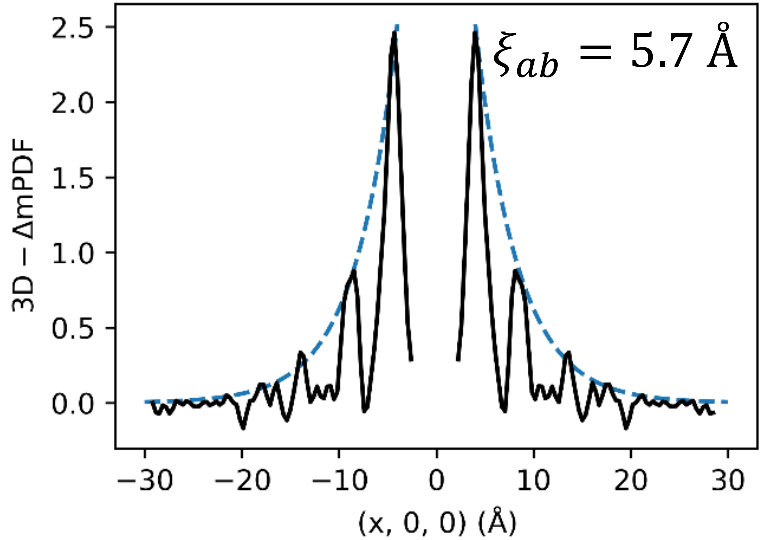
Ferromagnetic within the *xz* plane

Baral et al, *Matter* **5**, 1 – 12 (2022)

Anisotropic correlation length in MnTe



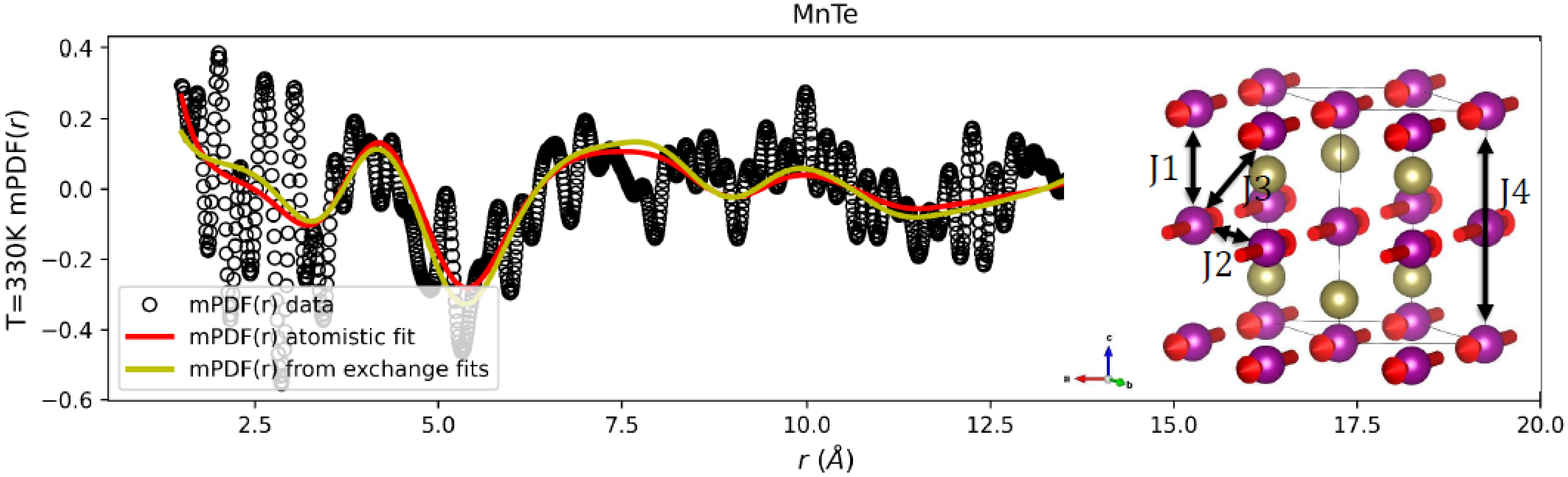




- Correlation length is roughly twice as large along *c* as within the *ab* plane
- Same qualitative conclusion from the energy-integrated results ($\xi_c = 7.7$ Å, $\xi_{ab} = 4.3$ Å)
- New information with practical implications for device design

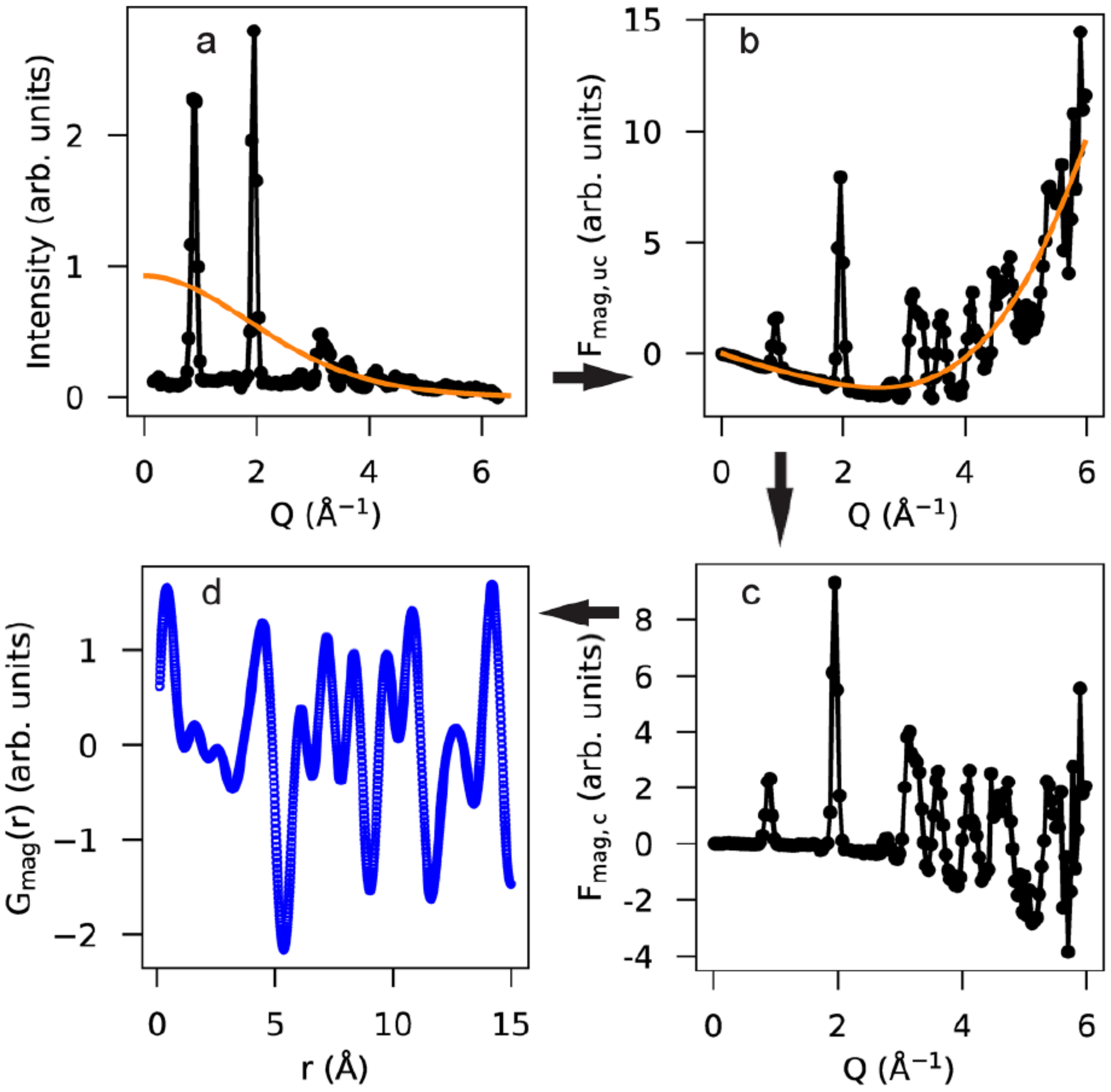
Extracting exchange parameters from mPDF data

- Extract magnetic interactions from mPDF data in paramagnetic state
- Fit to correlation length and locally ordered moment or directly to mPDF data
- Onsager reaction field theory or direct Monte Carlo simulations

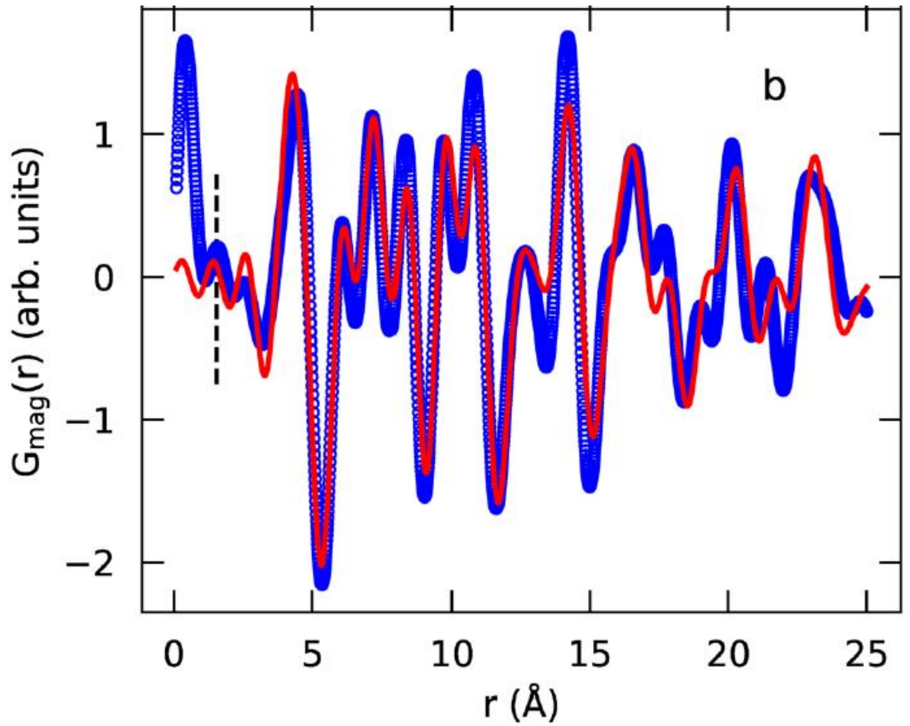


$$\langle S(0)S(r)\rangle \approx \frac{(2\pi)^2}{V_{BZ}\beta}\sum_{q_0}\frac{e^{-iq_0\cdot r}}{\sqrt{-\det C}}\frac{\exp(-\sqrt{r^T D r})}{\sqrt{r^T C^{-1} r}}$$

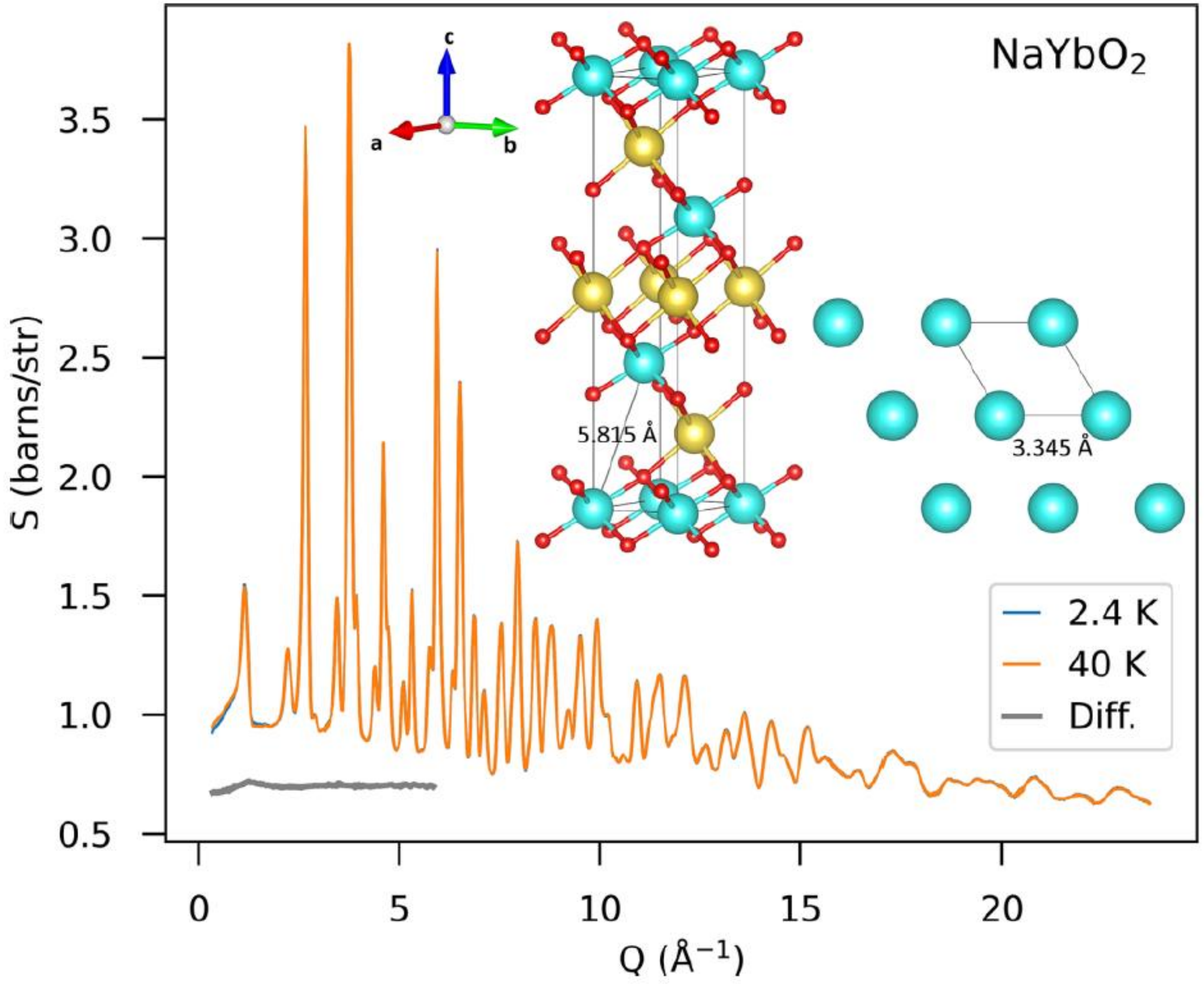
Magnetic PDF with polarized neutrons

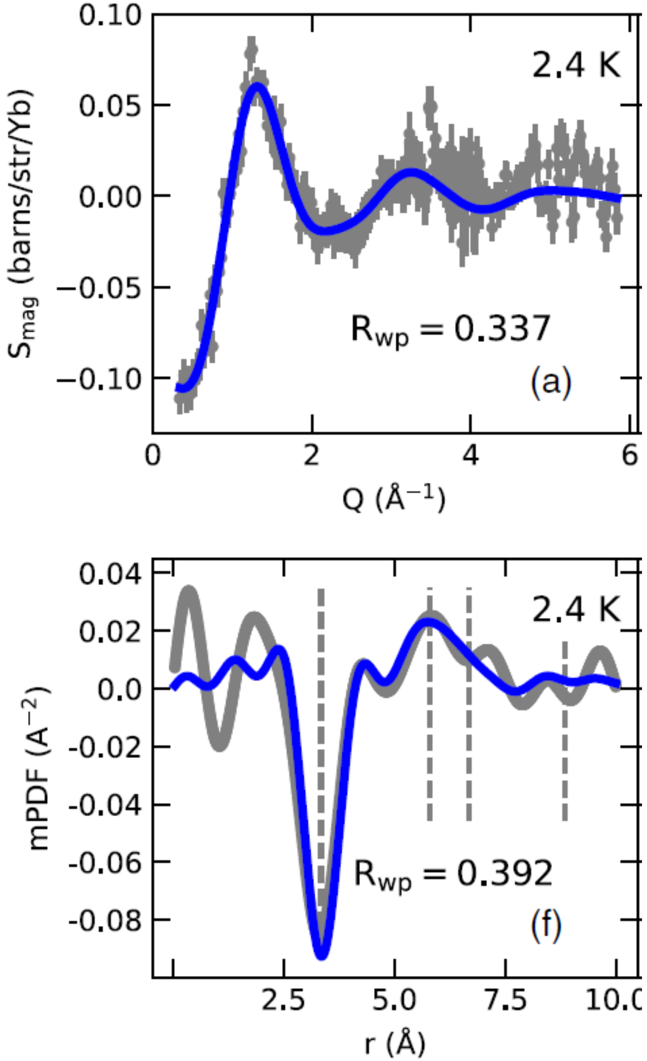


PDFgetX3-style *ad hoc* corrections allow us to obtain high-resolution mPDFs deconvolved from magnetic form factor with minimal human intervention.



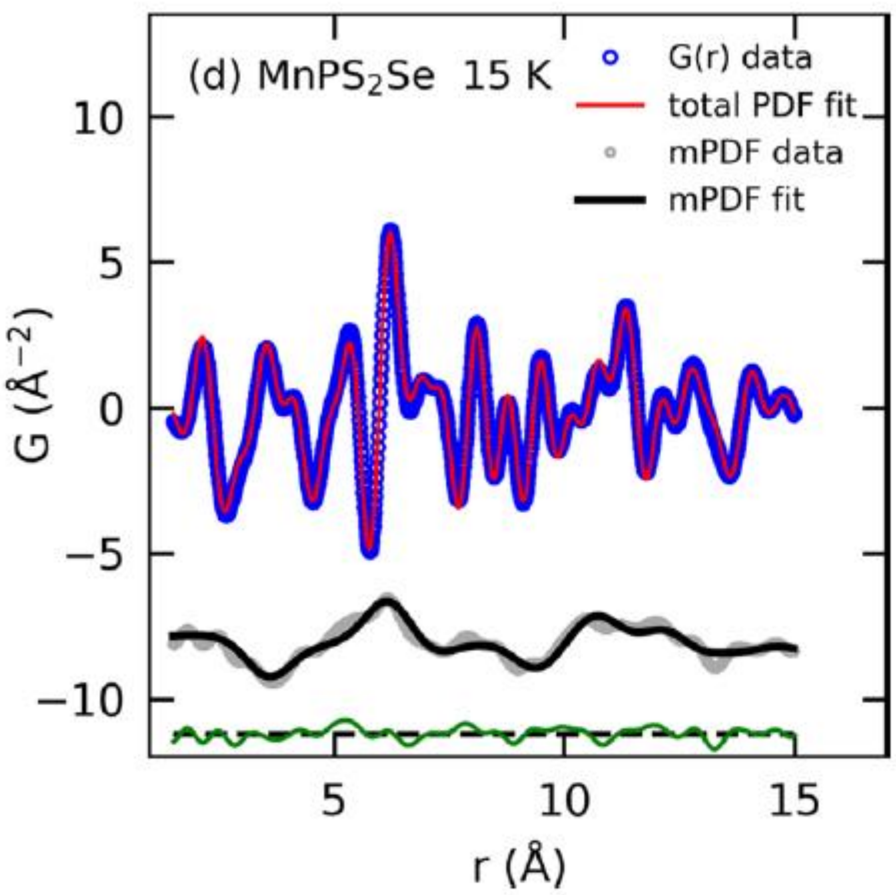
Magnetic PDF on D4 (ILL)



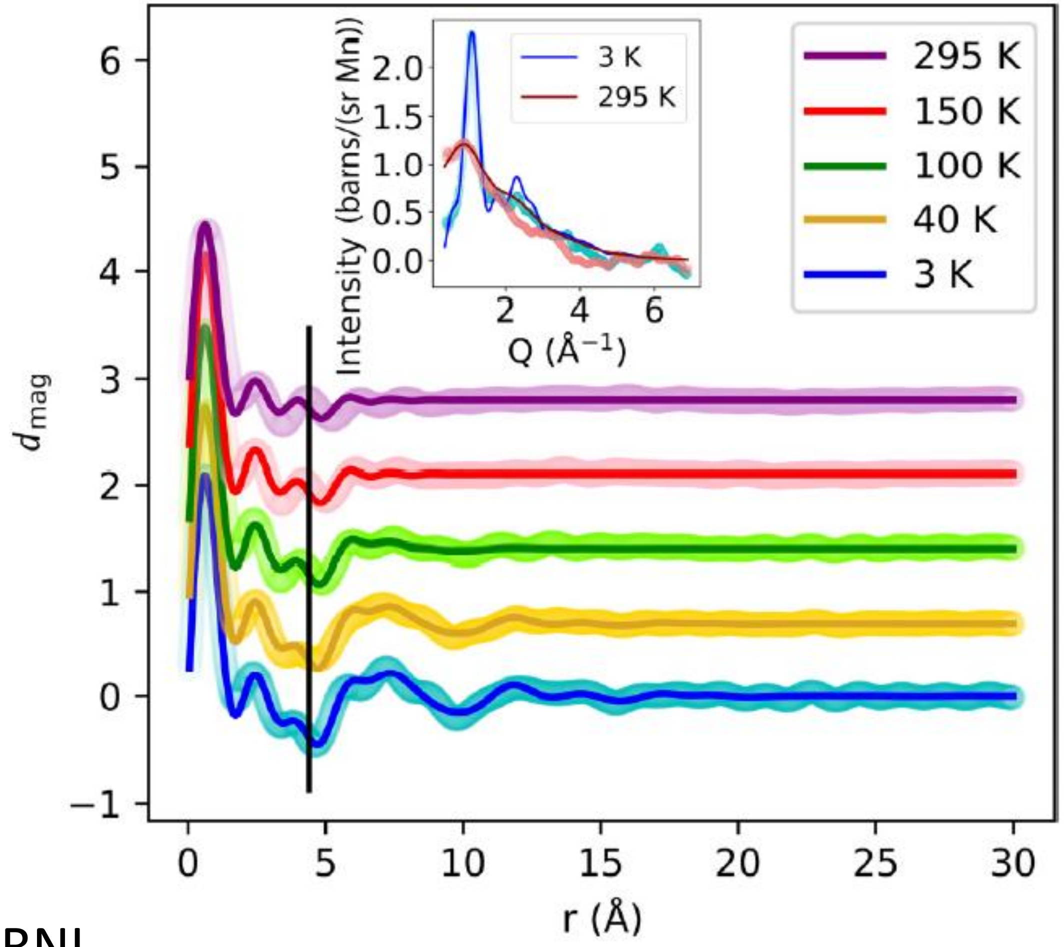


Nuttall et al., *PRB* **108**, L140411 (2023)

Magnetic PDF on conventional powder diffractometers



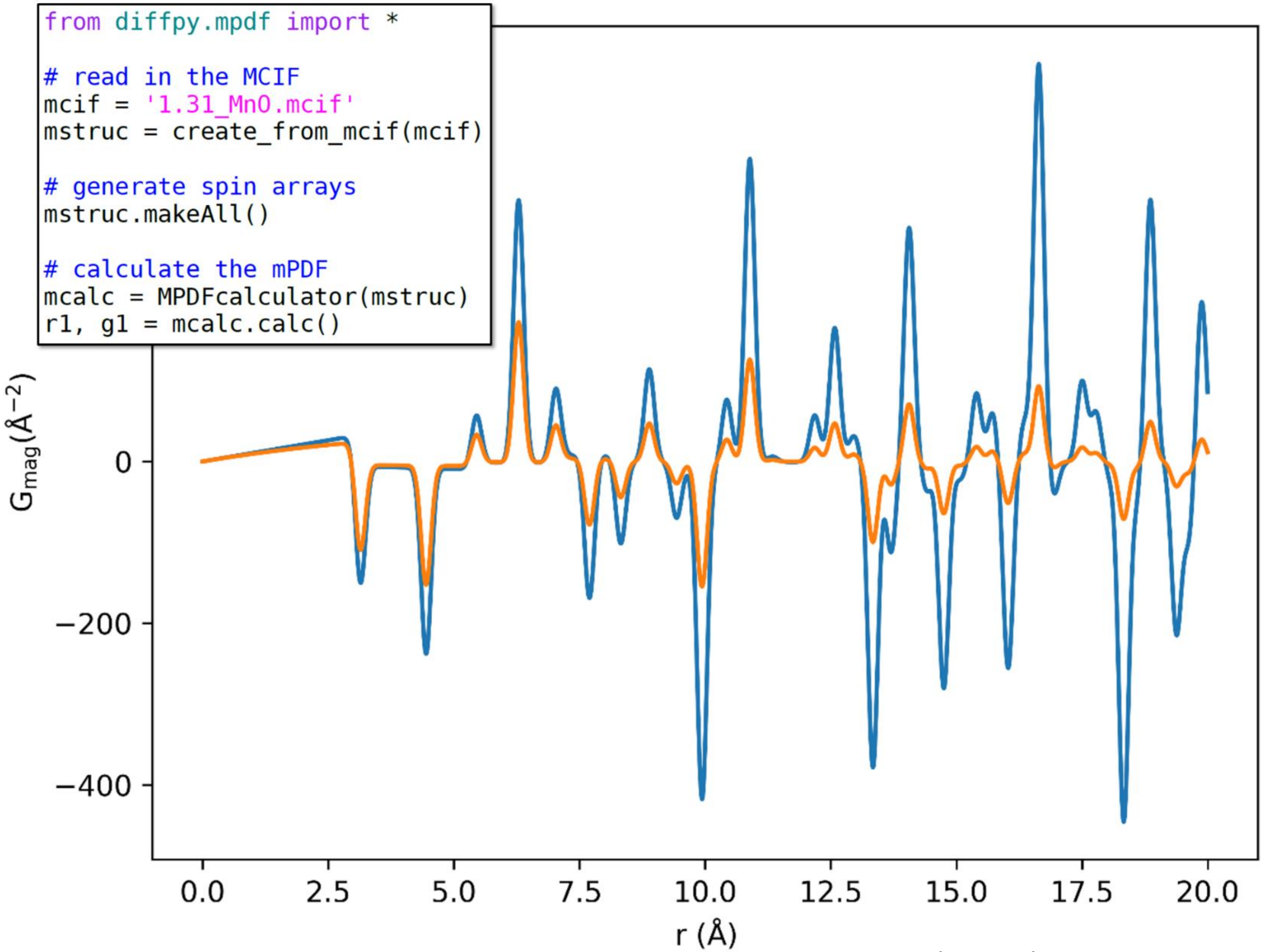
HB-2A, HFIR/ORNL



Baral et al., *PRB* **110**, 014423 (2024)

Hatt et al., *PRB* **112**, 144440 (2025)

`diffpy.mpdf`: Open-source mPDF software



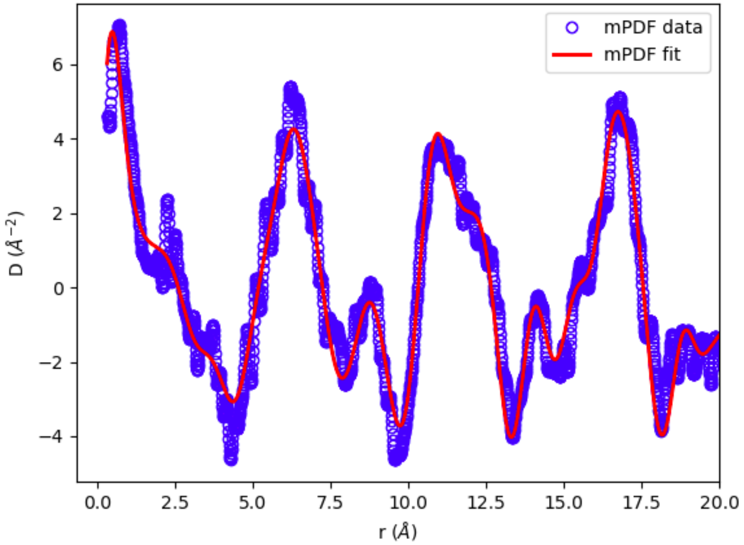
Frandsen et al, *J. Appl. Cryst.*, **55** (2022)

- Python package, integrates with DiffPy suite of PDF/diffraction code
- User-friendly methods for creating models of magnetic structures (e.g. loading in an MCIF file)
- Calculates 1D and 3D mPDF patterns for given magnetic structure
- Perform highly flexible fits
- Free to download and install at https://github.com/FrandsenGroup/diffpy.mpdf
- Check out the tutorial if you can, otherwise I'm very happy to help you get started!

diffpy.mpdf: Open-source mPDF software

Run the refinement

```
In [50]: ### Run the refinement

         # Turn off printout of iteration number.
         recipe.clearFitHooks()

         # Initial structural fit
         print("Refine using scipy's least-squares optimizer:")
         print("  variables:", recipe.names)
         print("  initial values:", recipe.values)

         from scipy.optimize import least_squares

         least_squares(recipe.residual, recipe.values)
         print("  final values:", recipe.valu

         ### Plot the results
         fit = mfit.evaluateEquation("mpdf")
```



Frandsen et al, *J. Appl. Cryst.*, **55** (2022)

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- Calculates 1D and 3D mPDF patterns for given magnetic structure
- Perform highly flexible fits
- Free to download and install at https://github.com/FrandsenGroup/diffpy.mpdf
- Check out the tutorial if you can, otherwise I'm very happy to help you get started!


After all... why not?
Why shouldn't I try magnetic PDF?