Oak Ridge National Laboratory | High Flux Isotope Reactor

Magnetic PDF for total scattering analysis

Presenter: Raju Baral

Polarized Neutron Workshop
Date: April 2, 2026

U.S. Department of Energy

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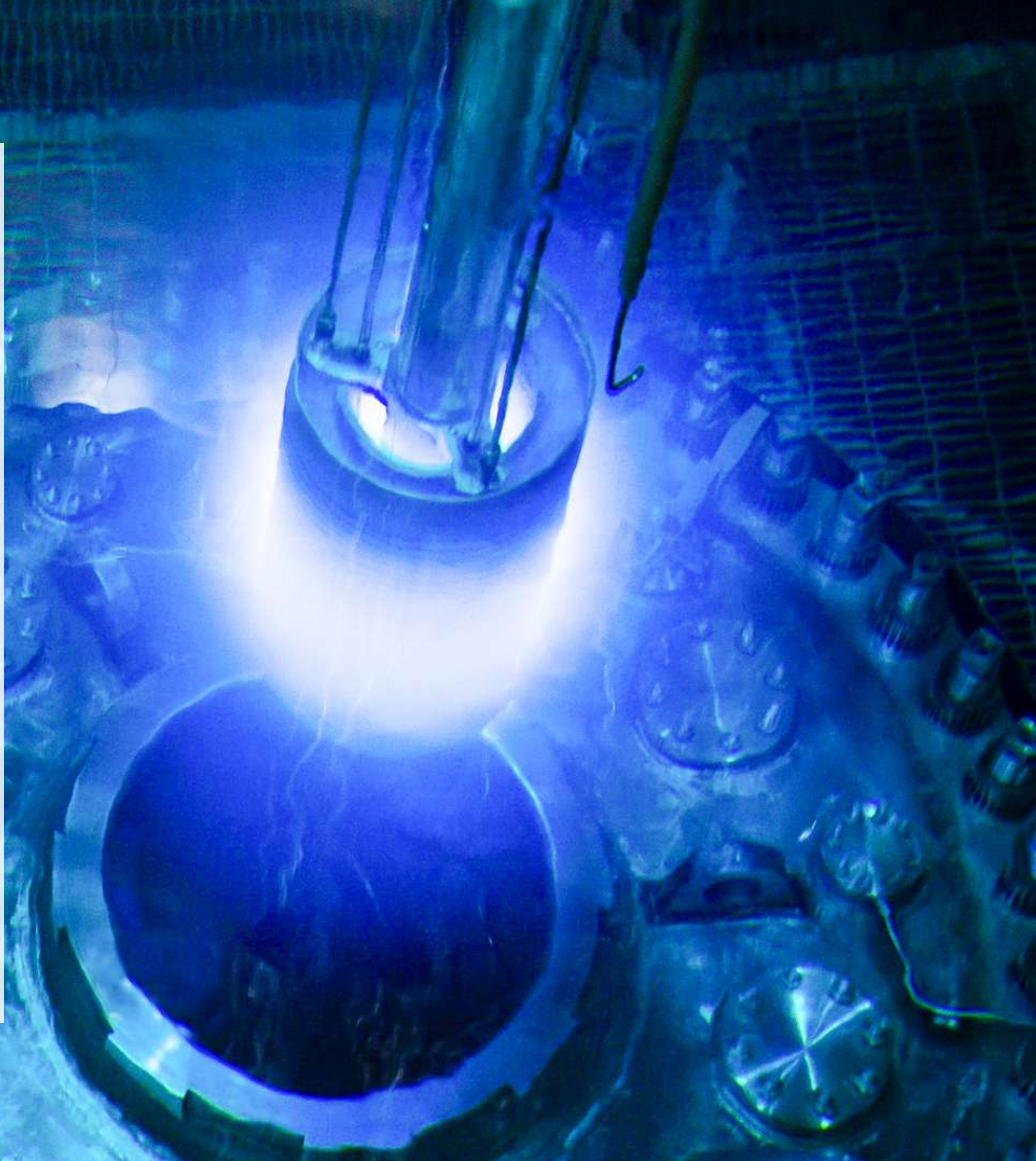


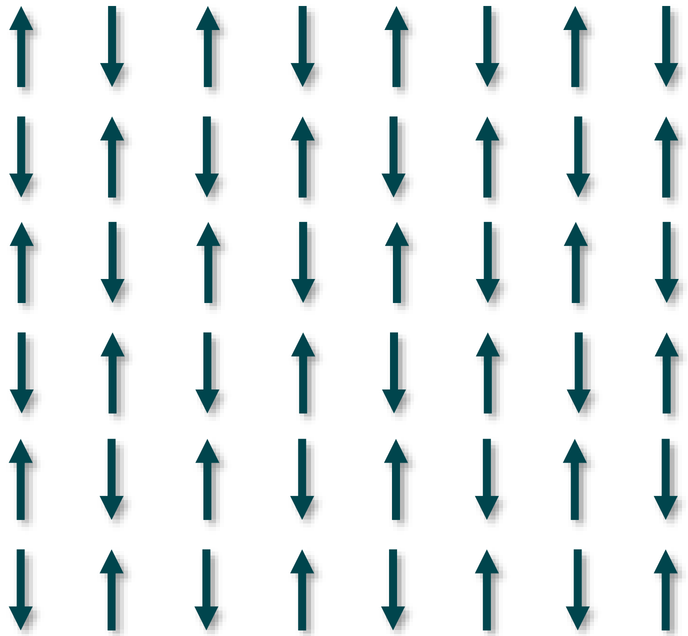
Outline

1. Introduction to magnetic pair distribution function
2. Isolate the magnetic signal and mPDF refinement
3. Case example: $Mn_3Si_2Te_6$

Introduction to magnetic pair distribution function analysis

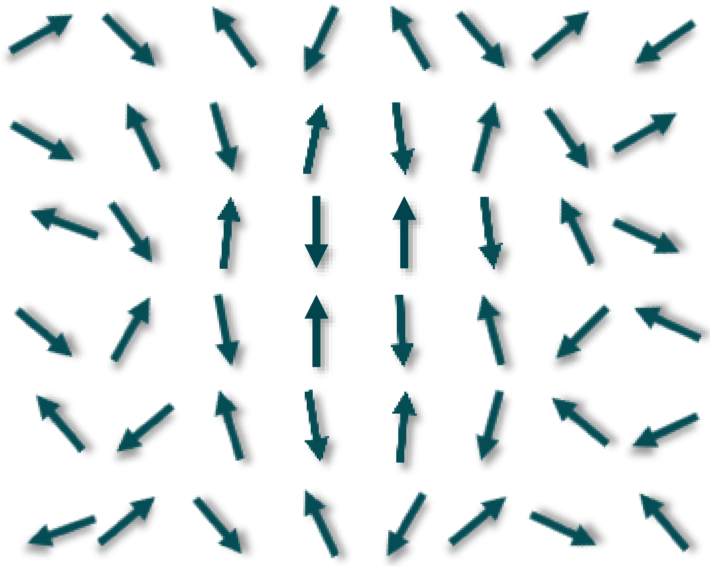
Objectives of mPDF: Study short-range magnetic order in real space

Long Range order



Well-defined correlations over infinite distance

Short Range order

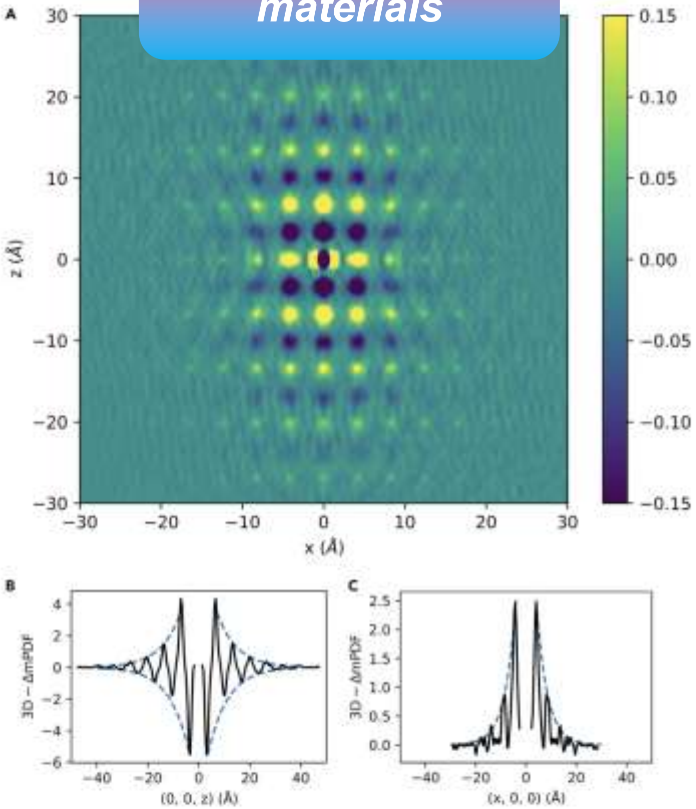


Well-defined correlations only over a finite separation distance

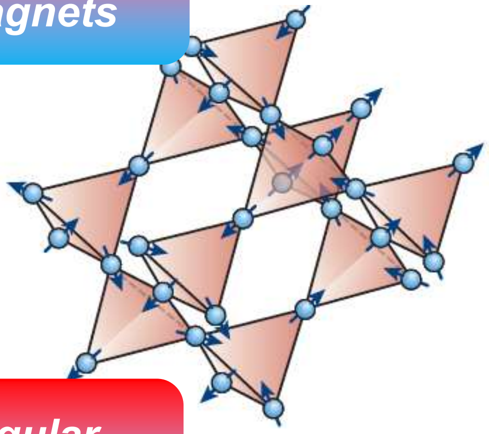
$$\langle S_0 \cdot S_i \rangle (r) \sim e^{-r/\xi}$$

mPDF in modern magnetism

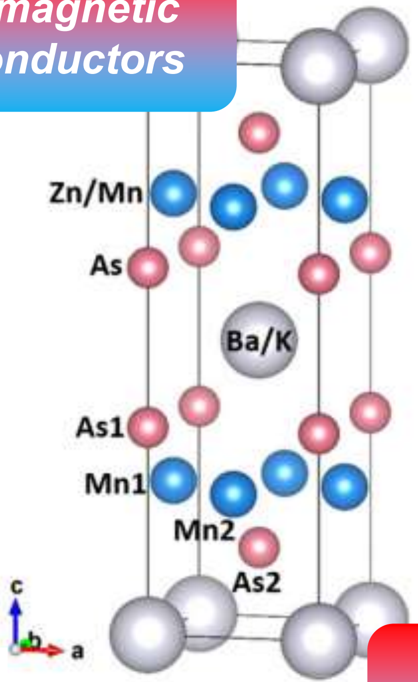
Thermoelectric materials



Frustrated magnets

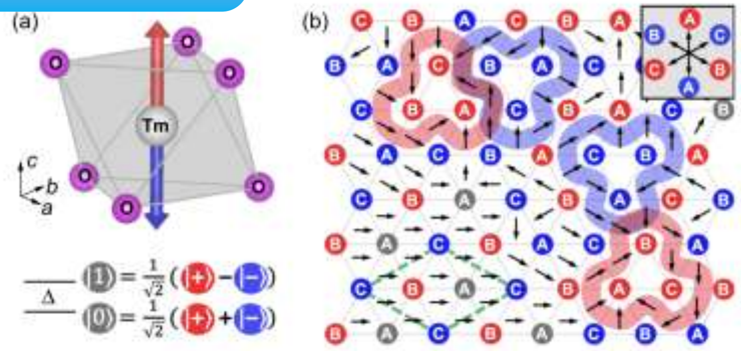


Dilute magnetic semiconductors

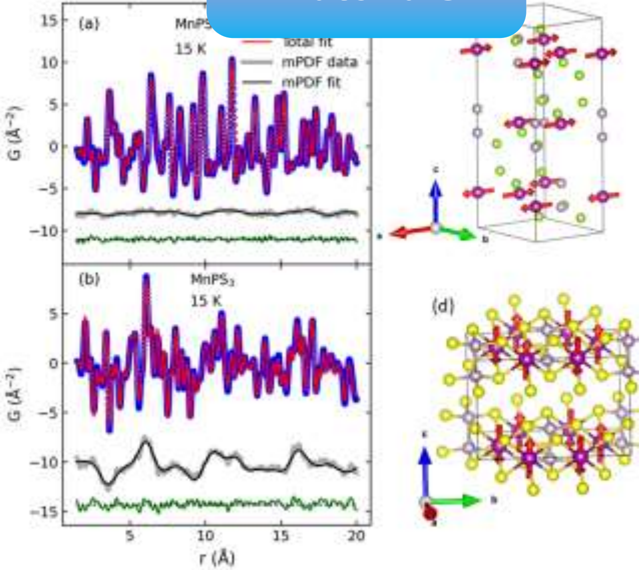


Many more examples!

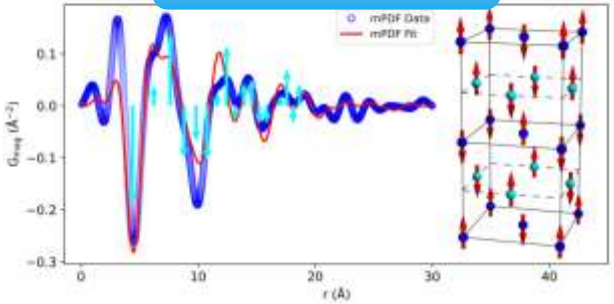
Triangular lattice (V-AV)



2D layered materials



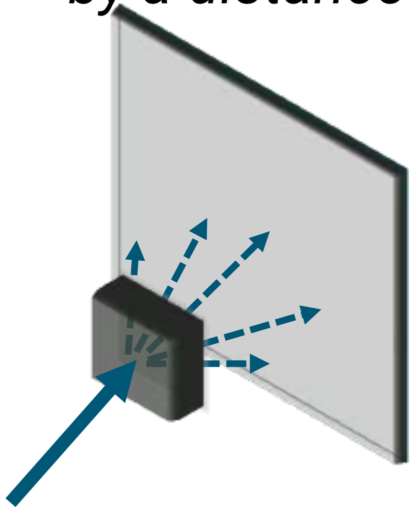
Spin glass

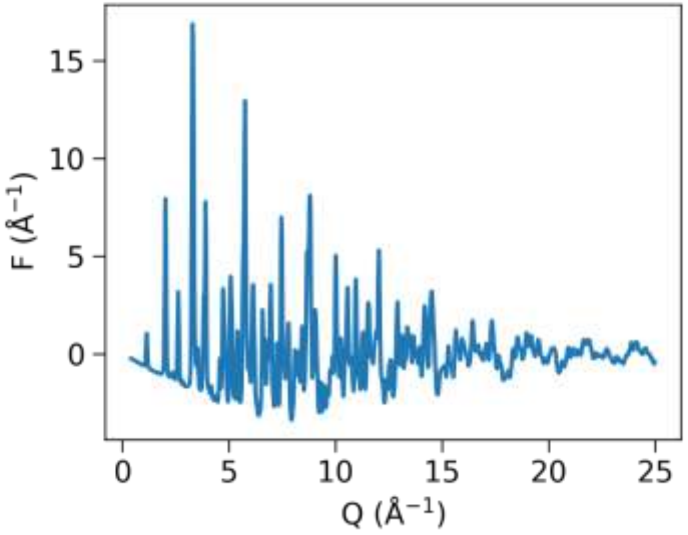


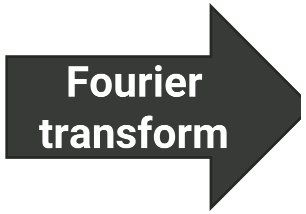
Baral et al., Matter **5**, 1853–1864 (2022)
Hatt et al., PRB B **112**, 144440 (2025)
Frandsen et al., PRB **94**, 094102 (2016)
Dun et al. PRB **103**, 064424 (2021)
Baral et al. PRB **110**, 014423
Nature 464.7286 (2010): 199-208.

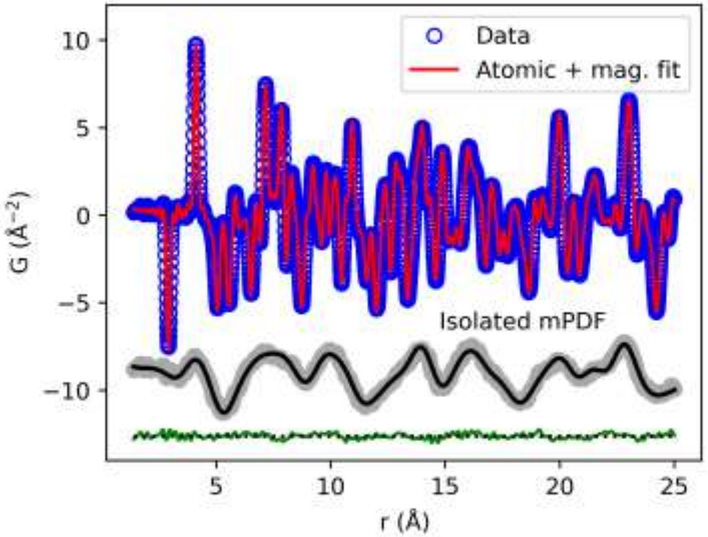
Pair Distribution Function (PDF) Analysis

Utilize the **TOTAL SCATTERING** (Bragg and diffuse) to extract the **AVERAGE AND LOCAL** structure
PDF: Probability of finding pair of atoms separated by a distance









Scattering experiment with powder or crystal sample

Obtain normalized scattering pattern

Obtain PDF and mPDF for analysis and modeling

Momentum Space → **Real Space**

Slide courtesy of B. Frandsen

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Atomic versus magnetic PDF

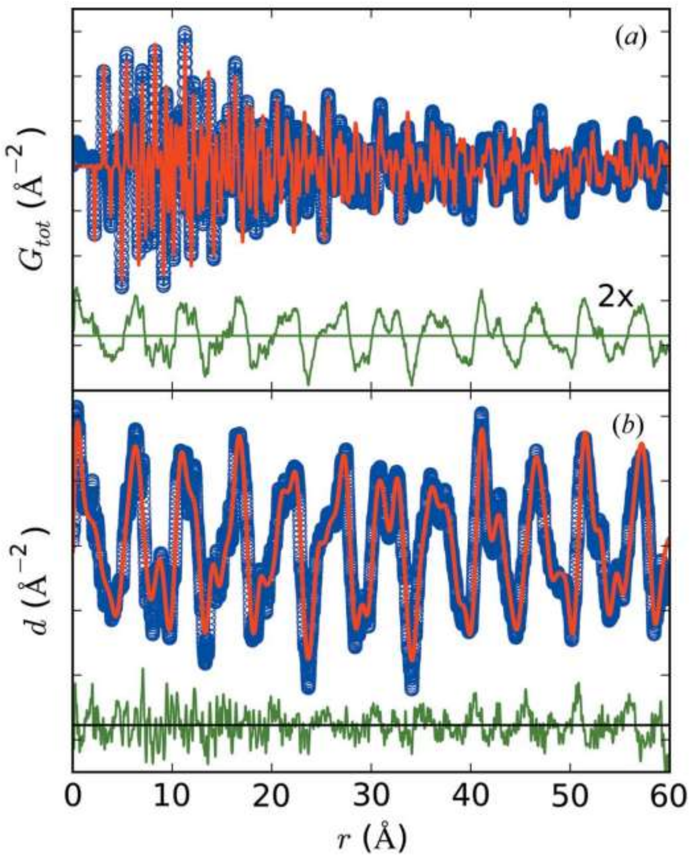
Scattering from **atoms** and **nuclei** → Fourier transform → **Atomic PDF**

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

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

Slide courtesy of B. Frandsen

Magnetic PDF



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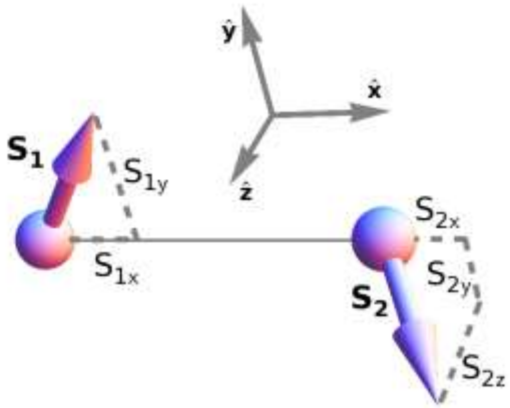
Magnetic structure determination from the magnetic pair distribution function (mPDF): ground state of MnO

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

$$G_{\rm mag}(r) = \frac{2}{\pi}\int_{Q_{\rm min}}^{\infty} Q\left(\frac{({\rm d}\sigma/{\rm d}\Omega)_{\rm mag}}{\frac{2}{3}N_s S(S+1)(\gamma r_0)^2[f(Q)]^2} - 1\right)\sin(Qr){\rm d}Q \quad (1)$$

$$= \frac{3}{2S(S+1)}\left(\frac{1}{N_s}\sum_{i\neq j}\left[\frac{A_{ij}}{r}\delta(r-r_{ij}) + B_{ij}\frac{r}{r_{ij}^3}\Theta(r_{ij}-r)\right] - 4\pi r\rho_0\frac{2}{3}m^2\right). \quad (2)$$



*Frandsen et al. Acta A **70** 3 (2014)*

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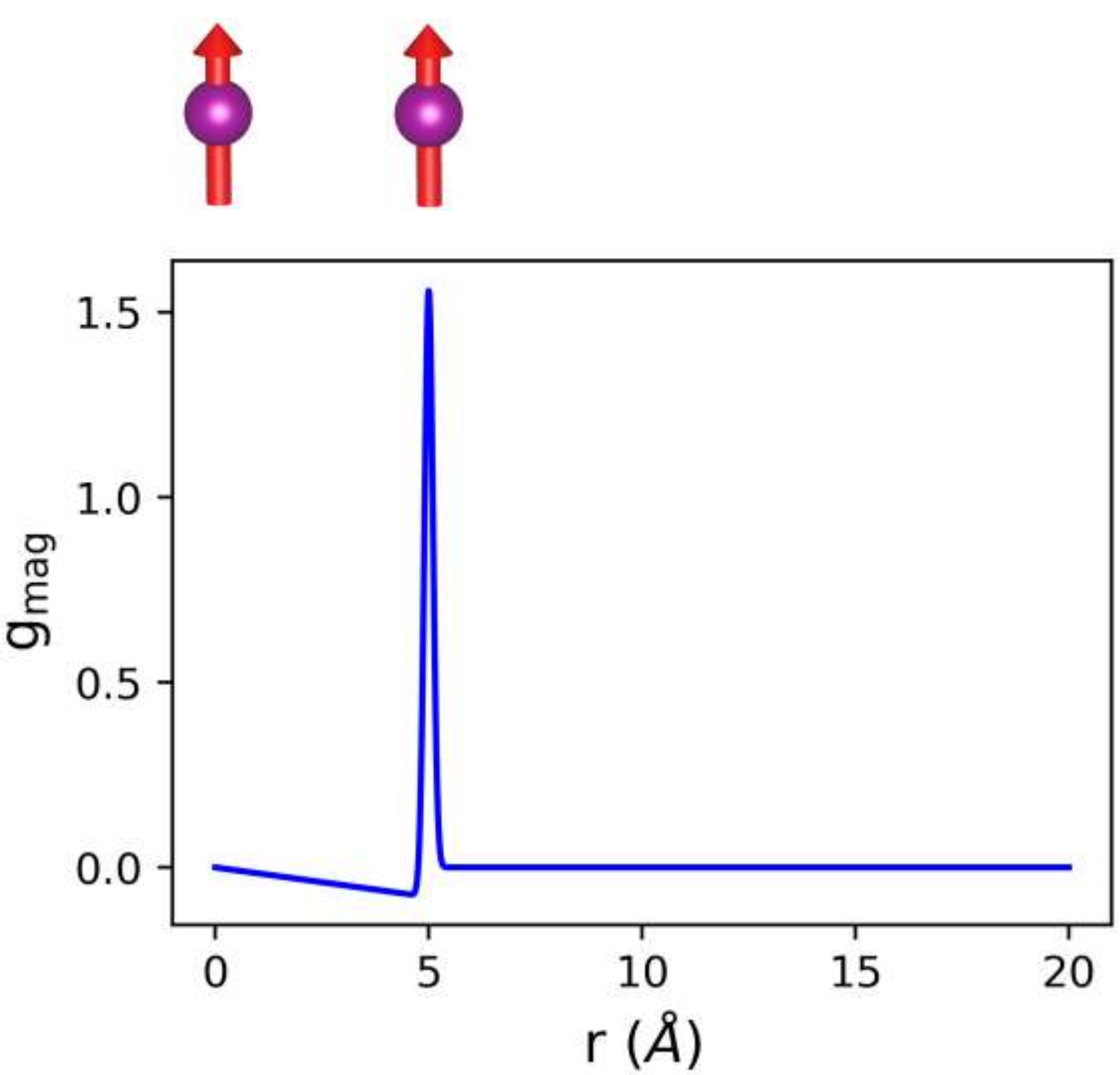
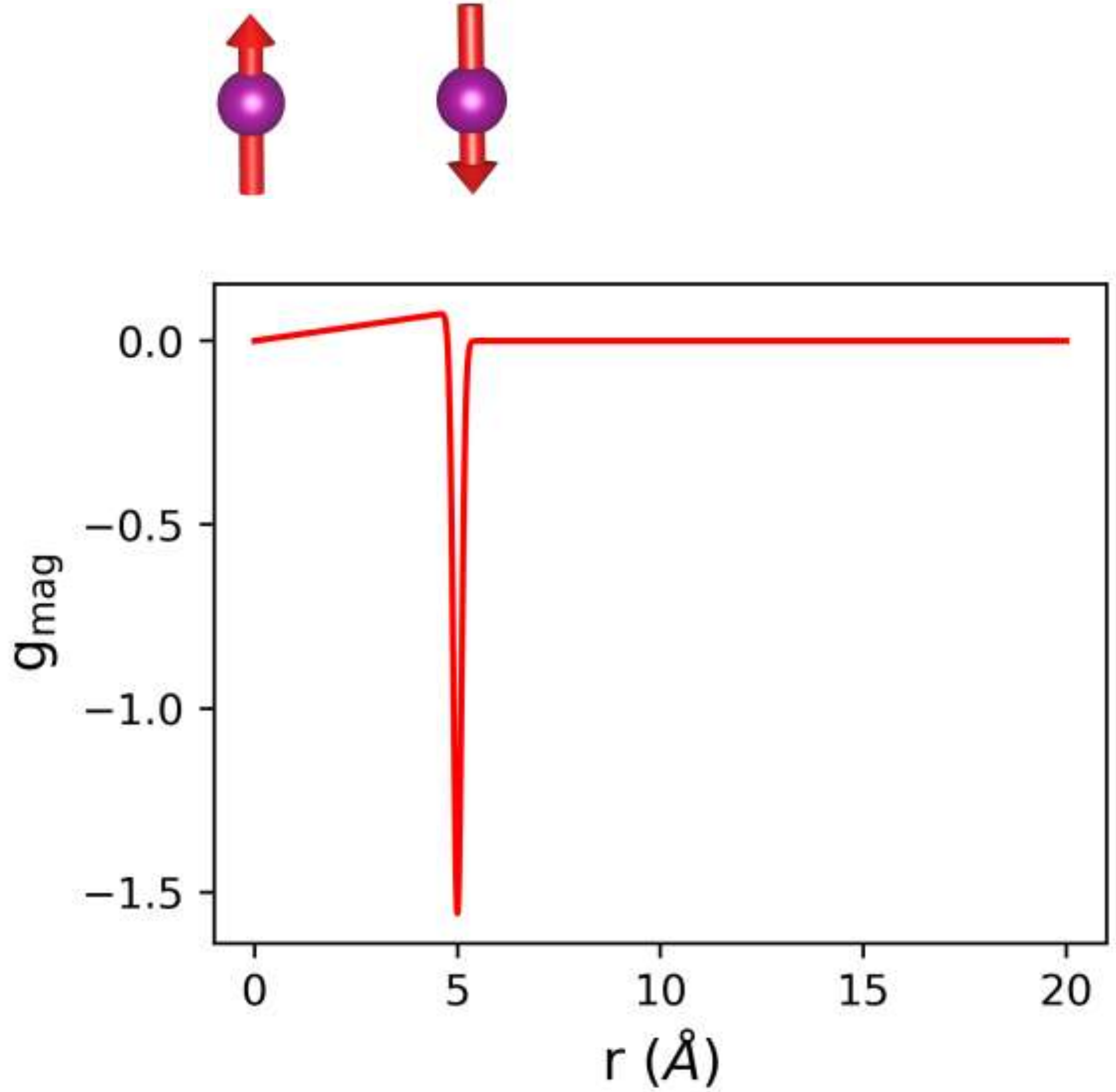
Magnetic PDF pattern of a pair of FM/AF spins





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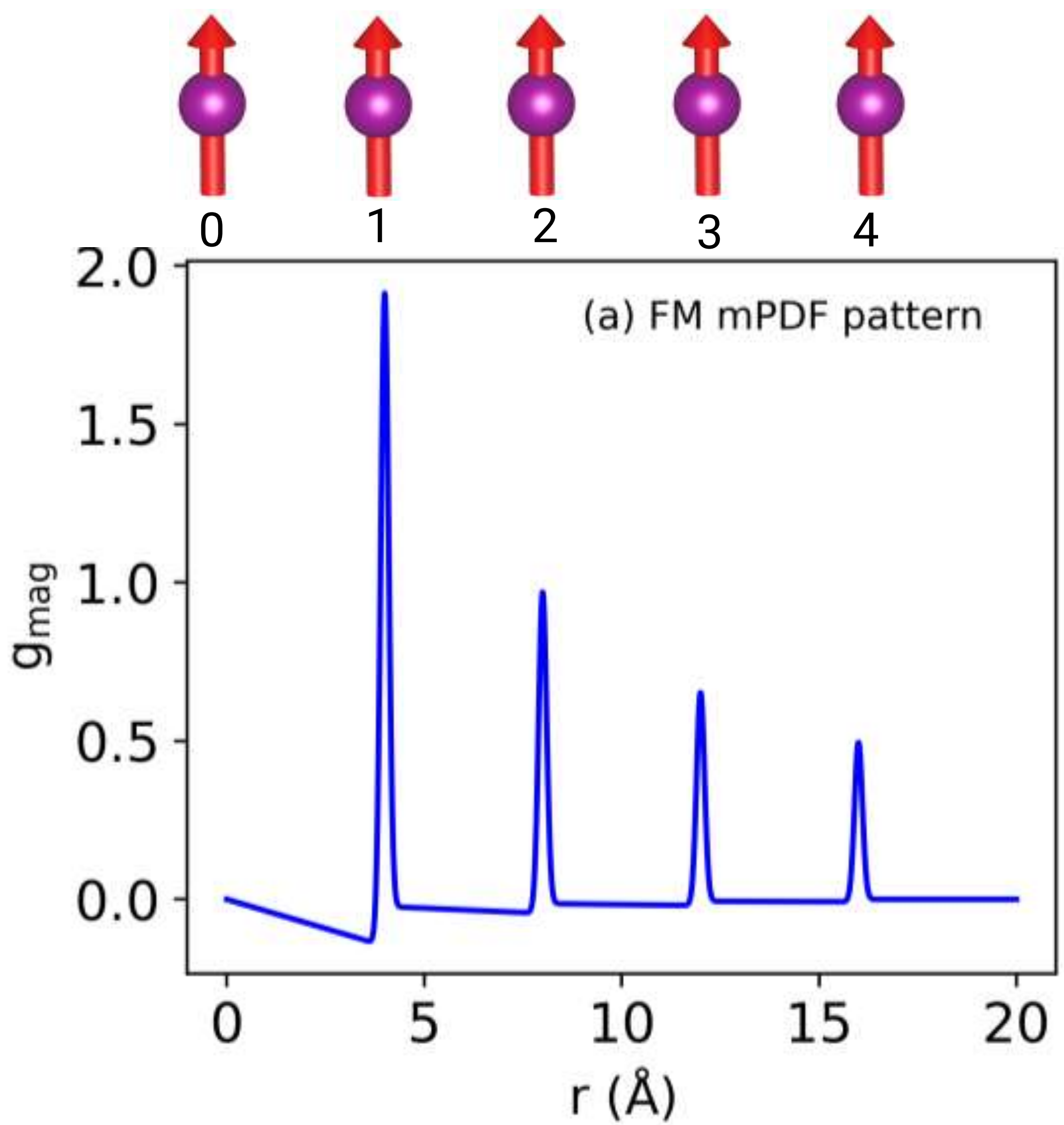
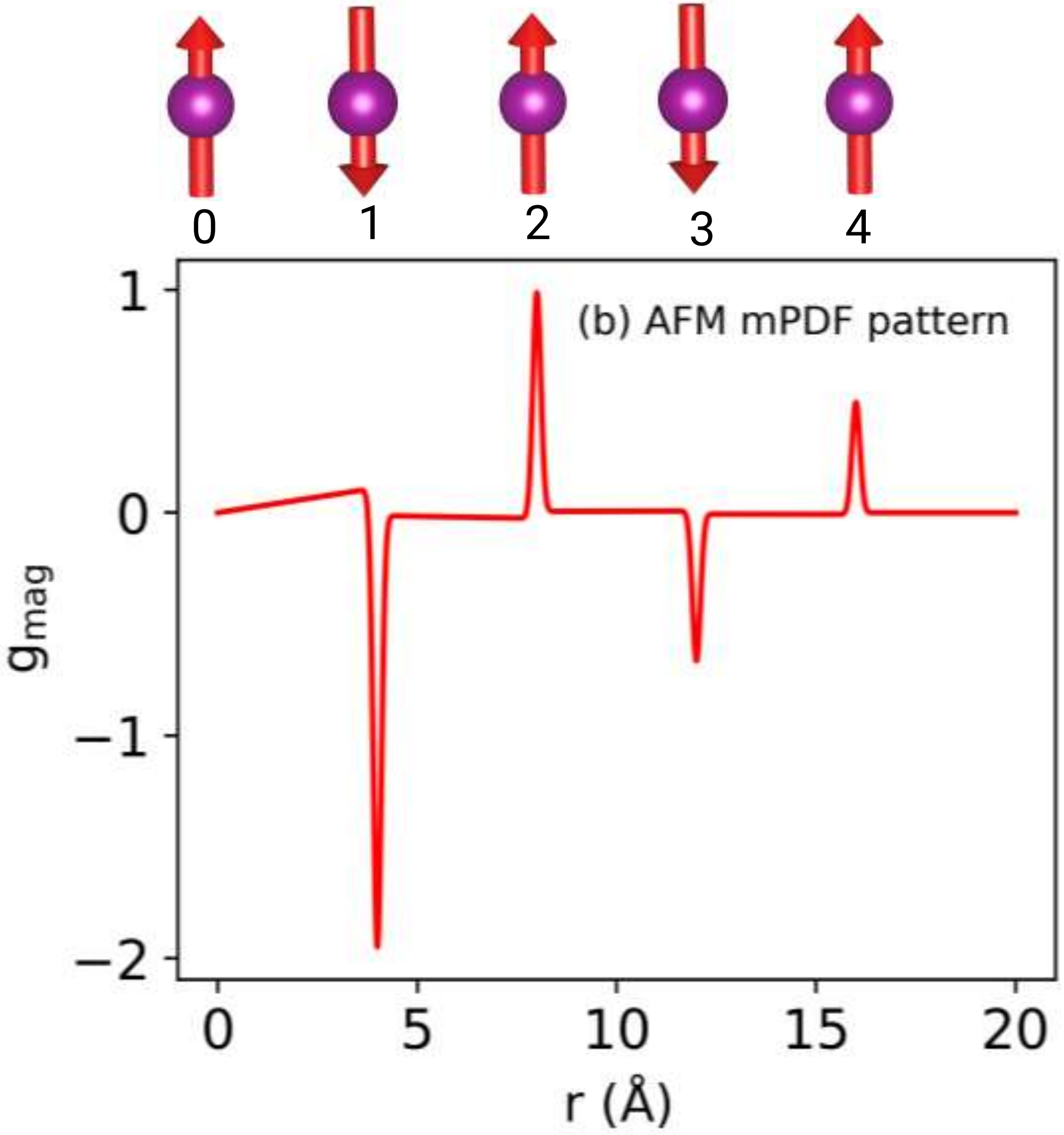
https://github.com/FrandsenGroup/mPDF-tutorial

*Frandsen et al. Acta A **70** 3 (2014)*

Example: One-dimensional chain of spin





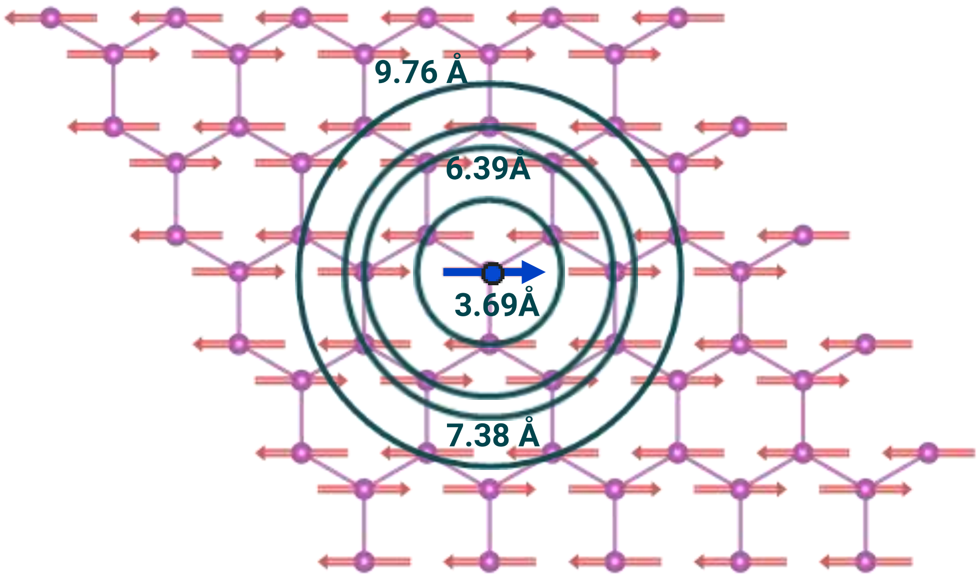
FM spins: Positive peaks
AFM spins: Negative and positive peaks

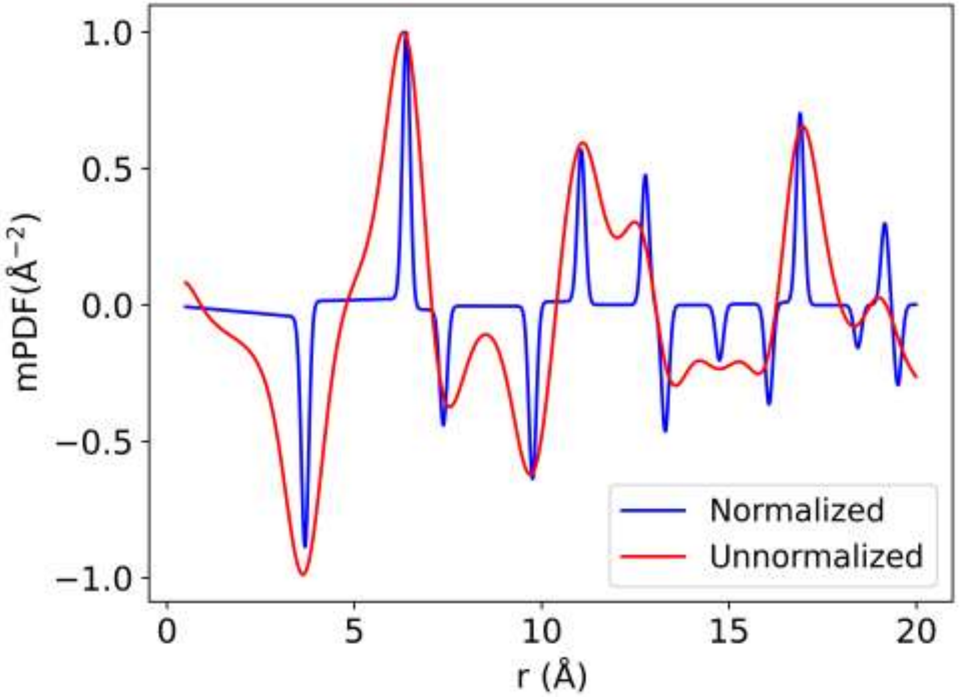
*Frandsen et. al., Acta A **70** 3 (2014)*

Example: Two-dimensional spin arrangement

Magnetic PDF (mPDF) gives the probability of finding a spin at a distance r from a given spin AND gives information on direction of spin

$$f(r) = \frac{1}{N}\frac{3}{2S(S+1)}\sum_{i\neq j}\left(\frac{A_{ij}}{r}\delta(r-r_{ij}) + B_{ij}\frac{r}{r_{ij}^3}\Theta(r_{ij}-r)\right)$$





*Frandsen et al., Acta A **70** 3 (2014)*

mPDF can be used for long or short range magnetic order

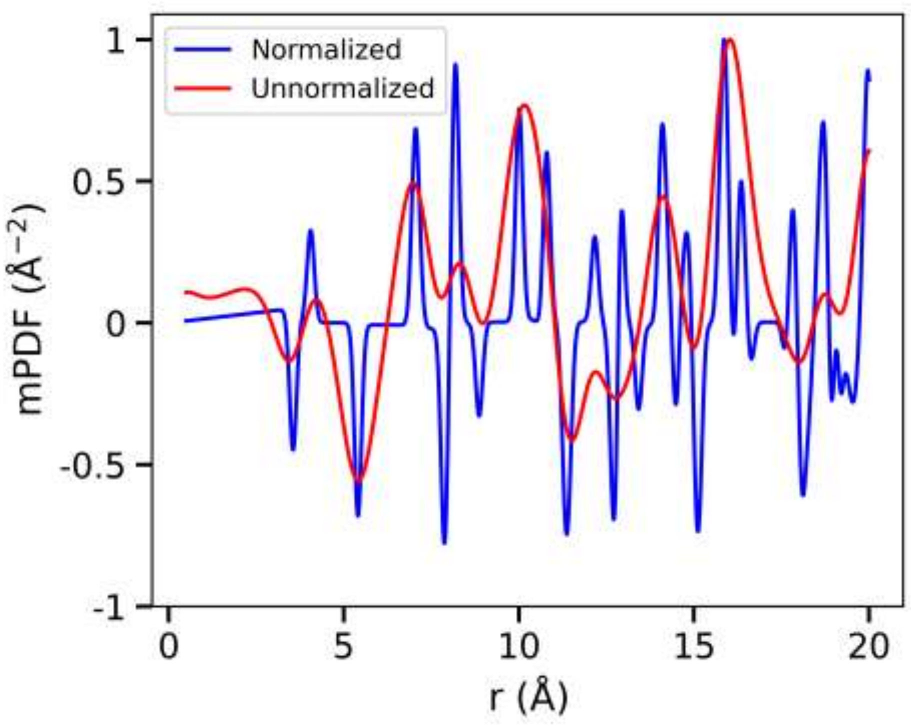
Open-source mPDF software: **diffpy.mpdf**

```python
from diffpy.mpdf import *

## read in the mcif
mcif = 'Mn3Si2Te6.mcif'
mstr = create_from_mcif(mcif, ffparamkey='Mn2')

## generate spin arrays
mstr.makeAll()

## calculate mPDF
mc = MPDFcalculator(mstr,qdamp=0.025,rmin=0.5,rmax=20)
mc.plot(both=True)
```



- ❑ Python package, part of the DiffPy suite of PDF/diffraction code
- ❑ User-friendly methods for creating models of magnetic structures (e.g. loading in an **mcif** file)
- ❑ Calculates 1D, 2D and 3D mPDF patterns for given magnetic structure
- ❑ Free to download and install at https://github.com/FrandsenGroup/diffpy.mpdf
- ❑ mPDF tutorials: *https://github.com/FrandsenGroup/mPDF-tutorial*

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

Neutron Data collection for mPDF

NOMAD/POWGEN at SNS, ORNL

https://neutrons.ornl.gov/nomad

- ❑ Traditional total scattering instrument
- ❑ Large Q_{max}

HB-2A at HFIR, ORNL

https://neutrons.ornl.gov/powder

- ❑ Not a traditional total scattering instrument
- ❑ Q_{max} ~10 A^{-1}
- ❑ Good enough for magnetism
- ❑ Low T and Magnet sample environment to expand mPDF

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1. Introduction to magnetic pair distribution function
2. Isolate the magnetic signal and mPDF refinement
3. Case example: $Mn_3Si_2Te_6$

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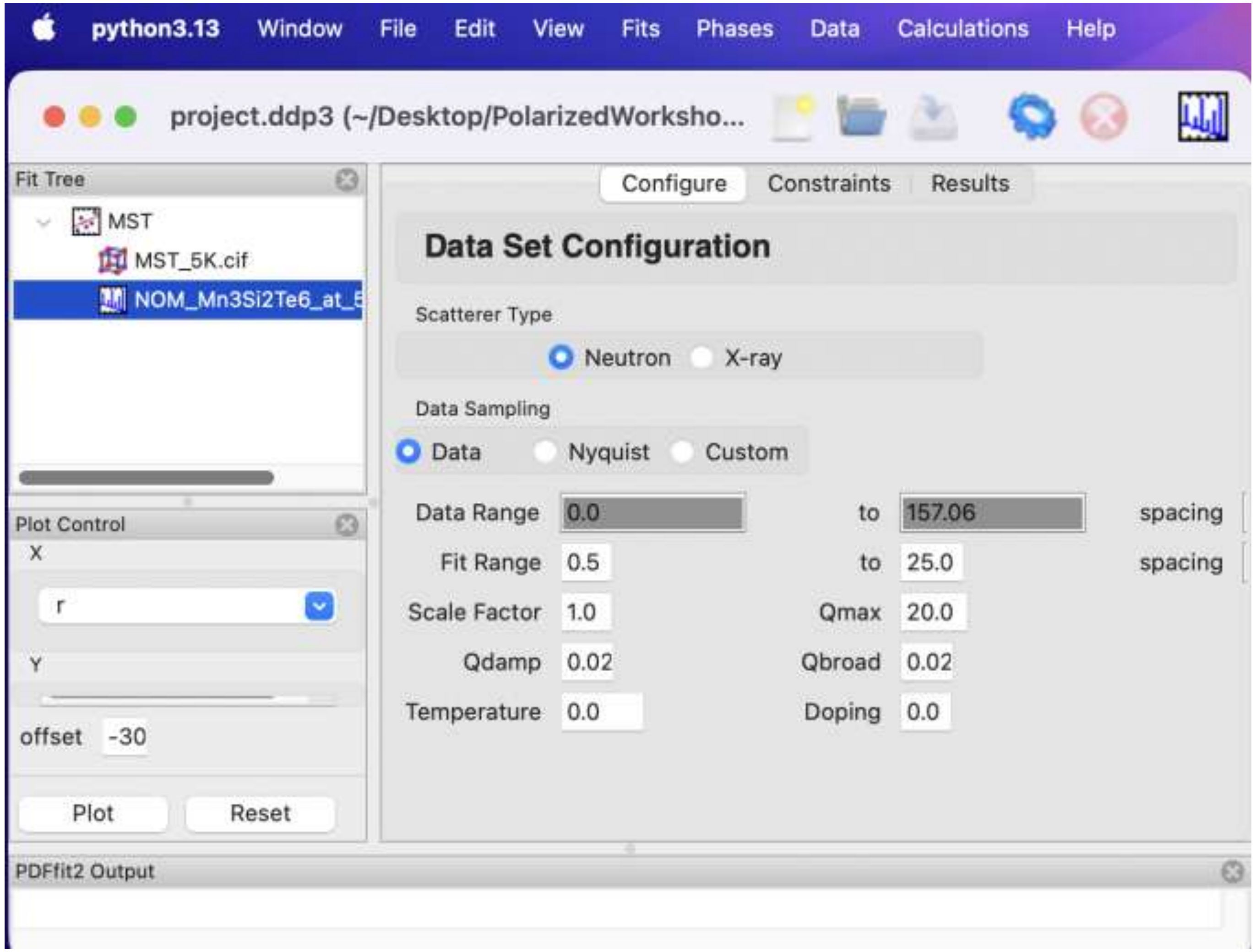
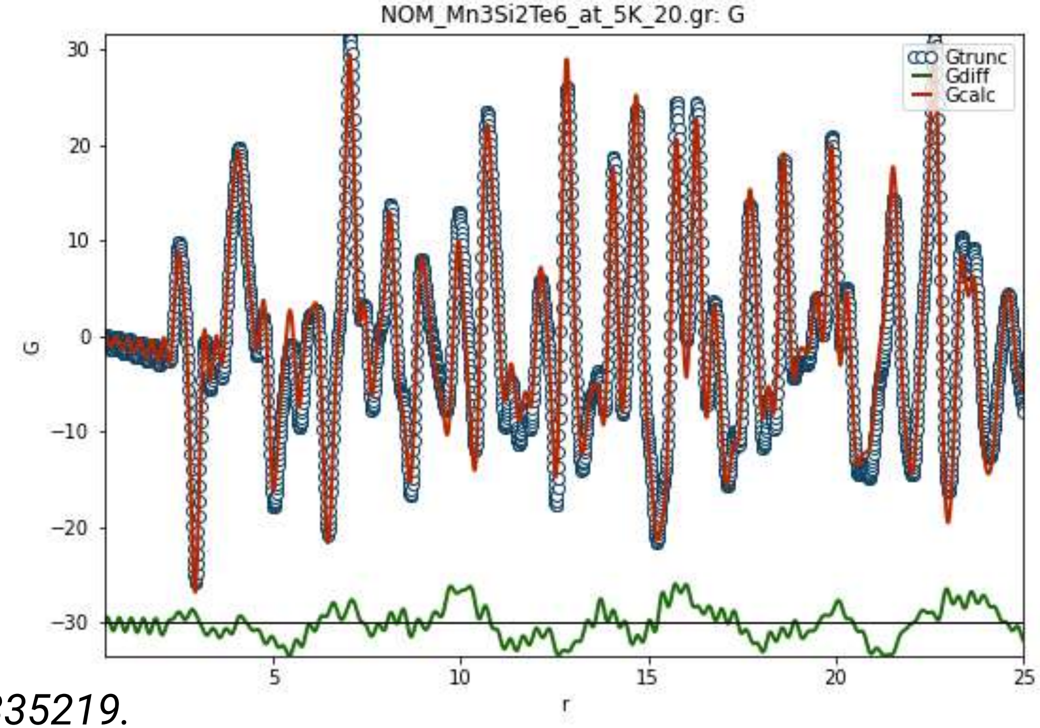
PDFgui Fit for magnetic material



https://github.com/diffpy/diffpy.pdfgui

Perform atomic PDF fit using PDFgui

1. Refine lattice parameter
2. Refine xyz
3. Refine APDs
4. Refine Delta1 or Delta2



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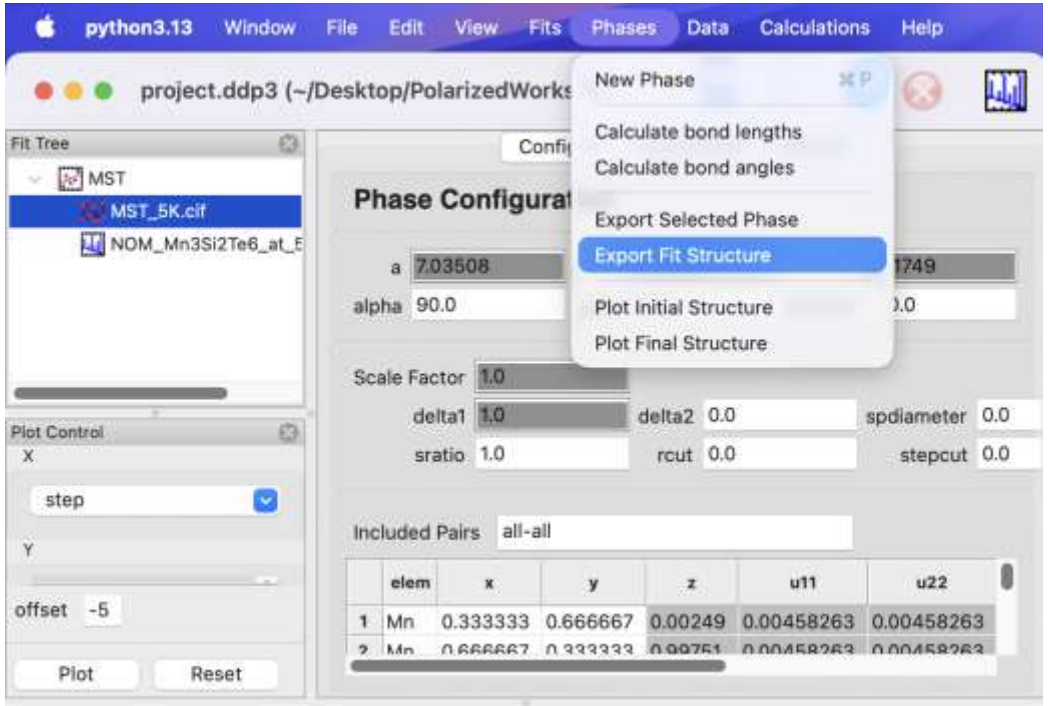
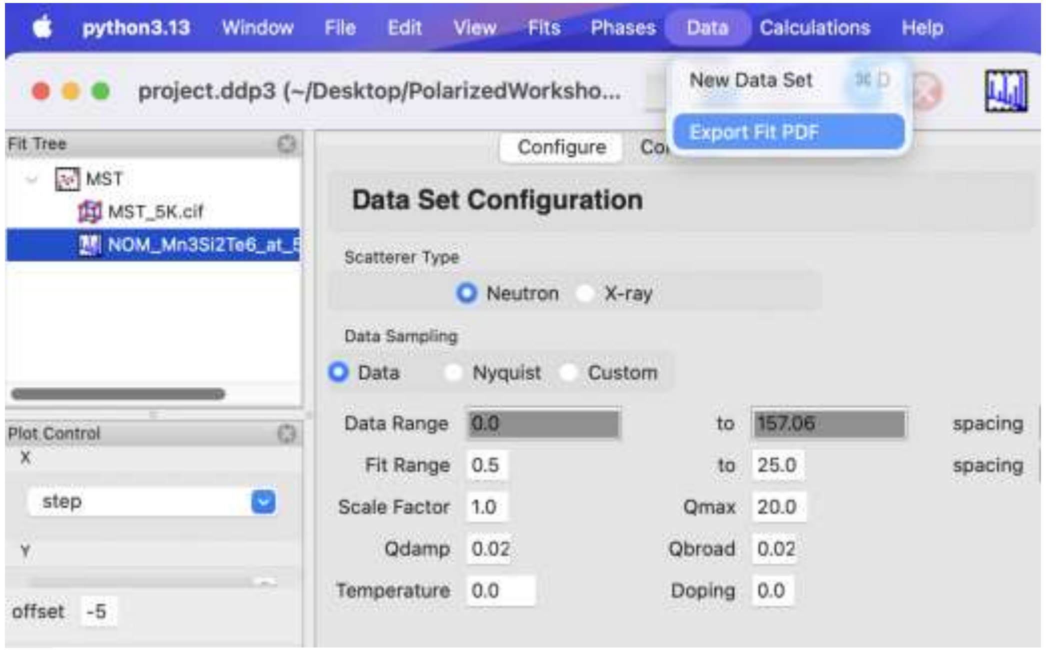
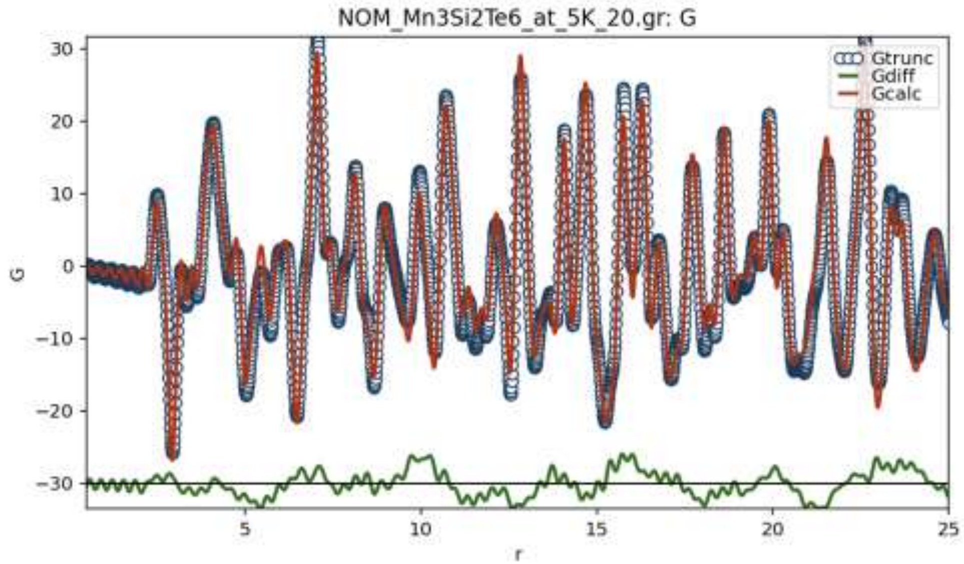
Farrow et al. J. Phys.: Condens. Matter 19 (2007) 335219.

PDFgui Fit for magnetic material







Farrow et al. J. Phys.: Condens. Matter 19 (2007) 335219.

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mPDF refinement using fits exported from PDFgui

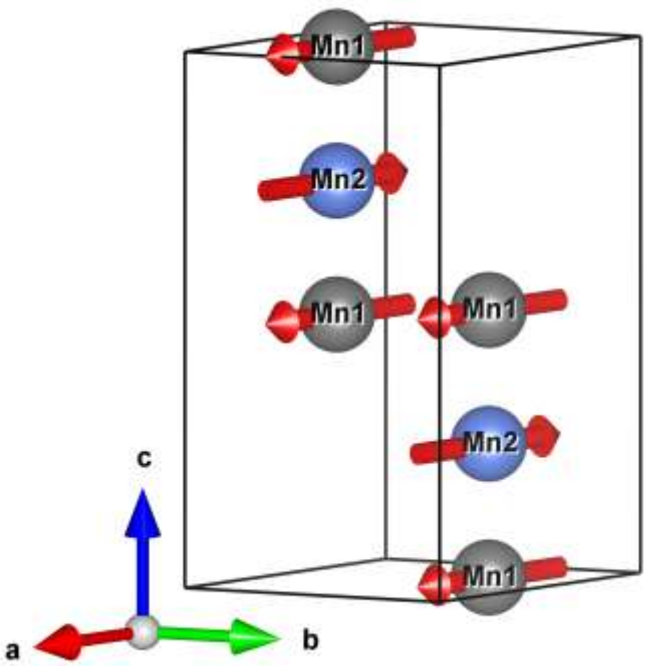
```
import numpy as np
import matplotlib.pyplot as plt
from scipy.optimize import least_squares
from diffpy.mpdf import *
from diffpy.structure import loadStructure
%matplotlib notebook
```

```
### load pdfgui fit
fit_file = 'NOM_Mn3Si2Te6_at_5K_20.fgr'
r, gobs, gcalc, gdiff = read_fgr(fit_file)
gmag = 1.0*gdiff                    ### unnormalized mPDF data
```

```
### Make the magnetic structure
struc = loadStructure('MST_5K.stru')
for i, atom in enumerate(struc):
    print(i, atom)

svec = np.array( [ 1,0,0])
mspec1 = MagSpecies(struc, strucIdxs=[0,1,2,3], ffparamkey='Mn2', rmaxAtoms=55,
                    basisvecs=svec, kvecs=np.array([0,0,0]),label='Mn_1')
mspec2 = MagSpecies(struc, strucIdxs=[4,5], ffparamkey='Mn2', rmaxAtoms=55,
                    basisvecs=-svec, kvecs=np.array([0,0,0]),label='Mn_2')
mstr=MagStructure(rmaxAtoms=55)
mstr.loadSpecies(mspec1)
mstr.loadSpecies(mspec2)
mstr.makeAll()

### Make the calculator
mc = MPDFcalculator(mstr)
mc.rmin = r.min()
mc.rmax = r.max()
mc.qdamp = 0.025
```




```
0 Mn   0.333333 0.666667 0.000357 1.0000
1 Mn   0.666667 0.333333 0.999643 1.0000
2 Mn   0.333333 0.666667 0.499643 1.0000
3 Mn   0.666667 0.333333 0.500357 1.0000
4 Mn   0.333333 0.666667 0.250000 1.0000
5 Mn   0.666667 0.333333 0.750000 1.0000
```

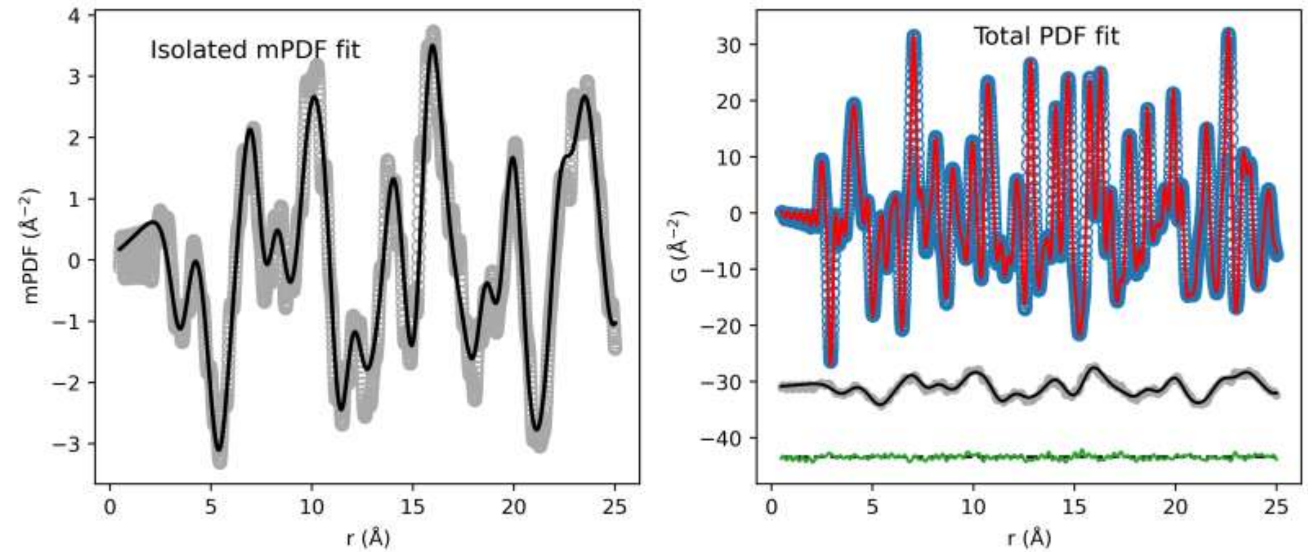
https://github.com/FrandsenGroup/diffpy.mpdf

mPDF refinement using fits exported from PDFgui

```
def mPDF_fit(p,ydata):
    oscale, pscale, xi,th, phi = p
    sx = np.sin(th)*np.cos(phi)
    sy = np.sin(th)*np.sin(phi)
    sz = np.cos(th)
    newSvec = np.array([sx, sy, sz])
    mspec1.basisvecs = newSvec
    mspec2.basisvecs = -newSvec
    mstr.makeSpins()
    mstr.corrLength = xi
    mc.ordScale = oscale
    mc.paraScale = pscale
    return ydata - mc.calc(both=True,linearTermMethod='autoslope')[2] ## ferrimagnet
```

```
p0 = [0.1,0.1,10,np.arccos(np.random.uniform(-1,1)),np.random.uniform(-np.pi,np.pi)]
optimized = least_squares(mPDF_fit,p0,bounds=[[0,0,0,0,-np.pi],[100,100,1000,np.pi,np.pi]], args=(gmag,))
mfit = mc.calc(both=True)[2]
```

```
offset = 1.2 * np.min(gobs)
fig, (ax1, ax2) = plt.subplots(1, 2, figsize=(9, 4))
ax1.plot(r, gmag, 'o', mfc='None', mec='darkgray')
ax1.plot(r, mfit, 'k-', lw=2)
ax1.set_xlabel(r'r ($\mathrm{\AA}$)')
ax1.set_ylabel(r'mPDF($\mathrm{\AA^{-2}}$)')
ax2.plot(r, gobs, 'o', mfc='None')
ax2.plot(r, gcalc + mfit, 'r-')
ax2.plot(r, gmag + offset, 'o',ms=3, mfc='None', mec='darkgray')
ax2.plot(r, mfit + offset, 'k-')
ax2.plot(r, np.zeros_like(r) + 1.4 * offset, 'k', linestyle='--')
ax2.plot(r, gmag - mfit + 1.4 * offset)
ax2.set_ylabel(r'G($\mathrm{\AA^{-2}}$)')
ax2.set_xlabel(r'r ($\mathrm{\AA}$)')
plt.tight_layout()
plt.savefig('fig.png',dpi=400)
plt.show()
```



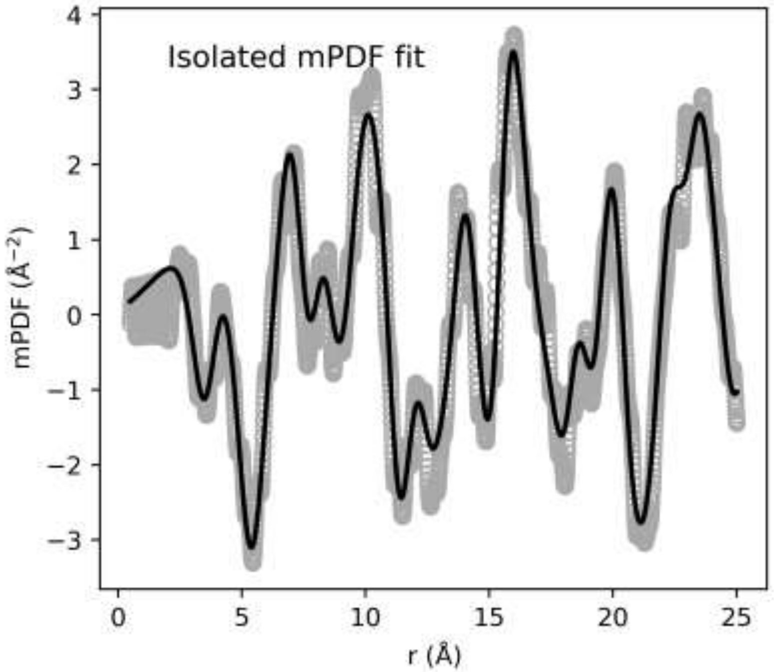
https://github.com/FrandsenGroup/diffpy.mpdf

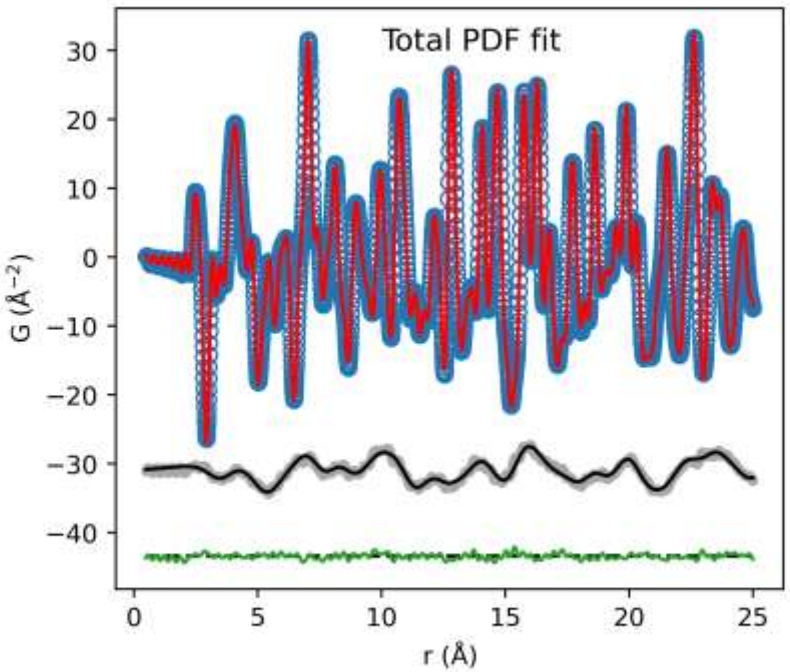
mPDF refinement using fits exported from PDFgui

```
uncertainties = getStdUnc(optimized, gmag)[0]
param_names = ['ordscale', 'pscale', 'xi', 'th', 'phi']
for name, val, unc in zip(param_names, optimized.x, uncertainties):
    print(f"{name} = {val:.4f}")

## calculate local moment
nucScale = 1.542731
unc_ord = uncertainties[0]
unc_xi = uncertainties[2]
unc_nuc = 0.001
m, m_unc = calculate_ordered_moment(mc, nucScale, returnUncertainty=True, inputUnc=[unc_ord, unc_nuc, unc_xi])
print(f'm = {m:.4f} ± {m_unc:.4f} µB')
```

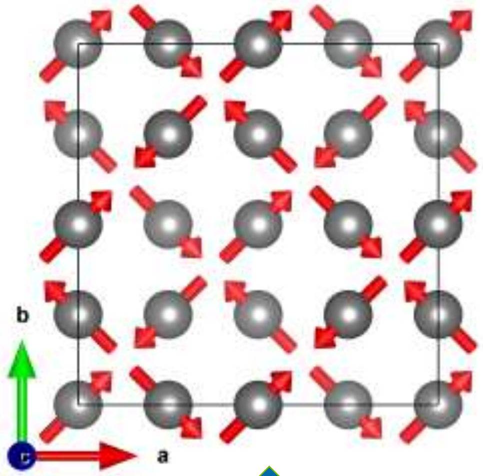
```
ordscale = 0.2558
pscale = 0.0000
xi = 73.7537
th = 1.5676
phi = 2.4210
m = 4.3090 ± 0.0618 µB
```

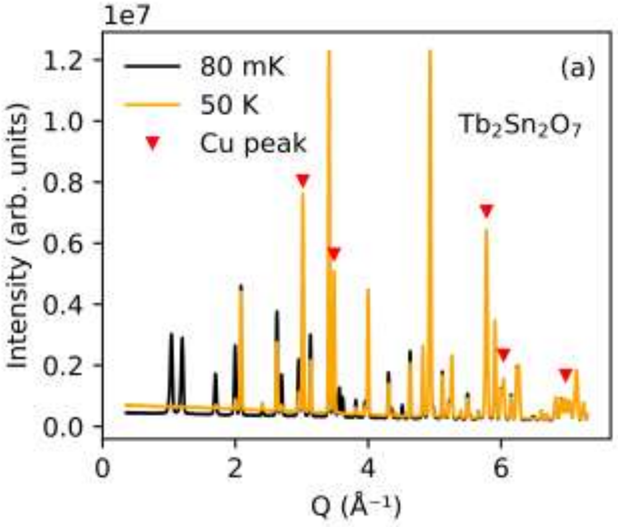


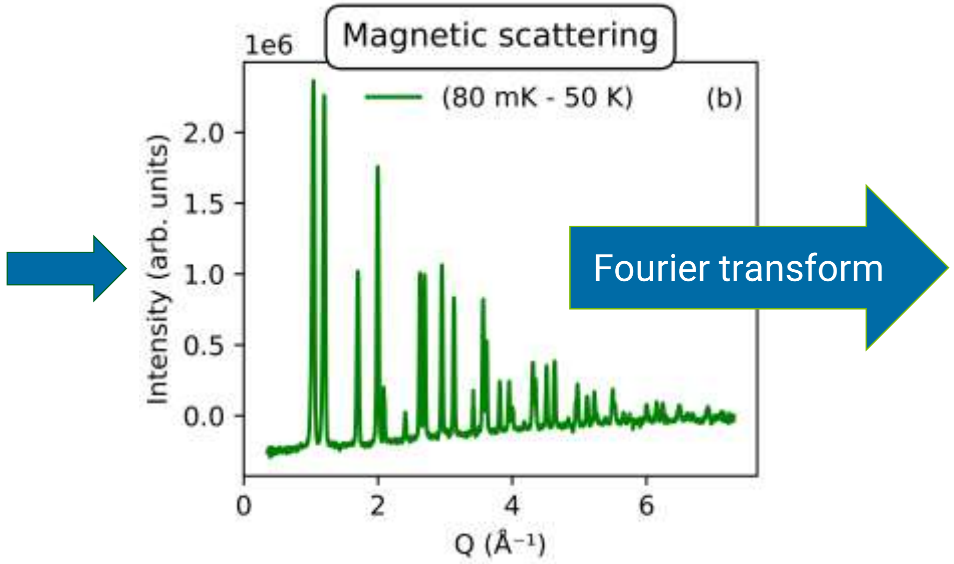


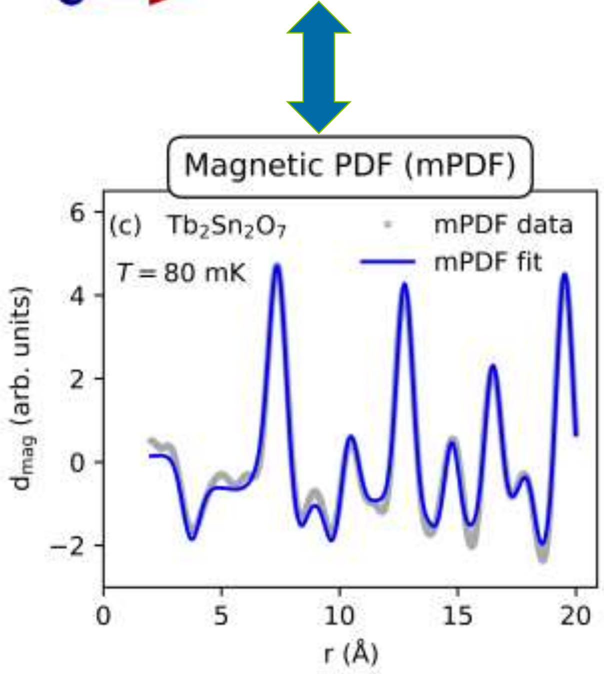
Temperature subtraction

- Measurement at low temperature (e.g. 80 mK)
  -captures both magnetic and nuclear scattering
- Measurement at high temperature (e.g. 50 K)
  - captures nuclear scattering only
- Temperature subtraction (low T minus high T data)
  - magnetic scattering
- Fourier transform the difference
  - convert Q-space data to real space mPDF data









Baral et. al., JMMM 630, 173342 (2025)

Outline

1. Introduction to magnetic pair distribution function

2. Isolate the magnetic signal and mPDF refinement

3. Case example: $Mn_3Si_2Te_6$

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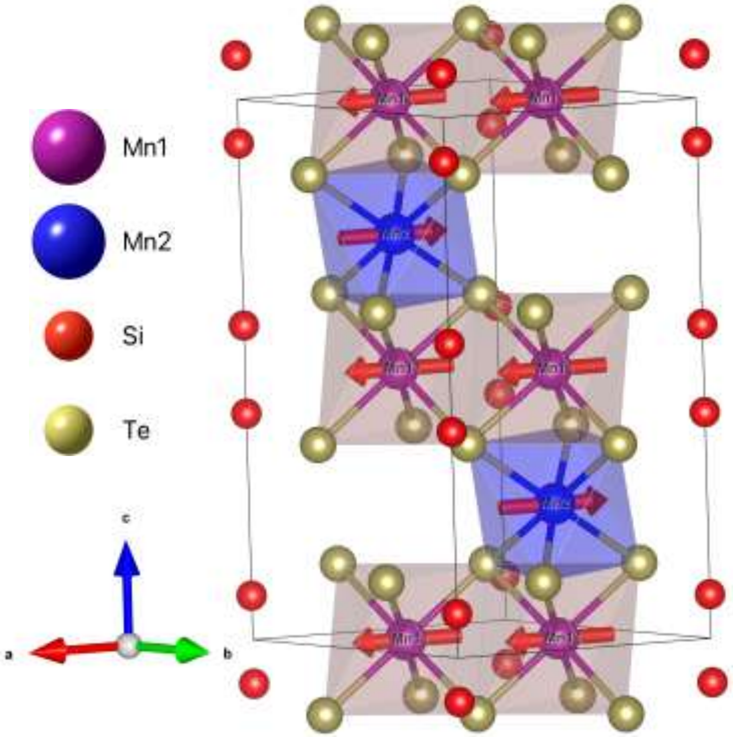
Introduction to $Mn_3Si_2Te_6$

- ❑ It has layered structure with trigonal crystal system(SG 163 P-31c)
- ❑ Two inequivalent sites are Mn1 and Mn2 in a unit cell
- ❑ Semiconducting ferrimagnetic below 78 K
- ❑ S=5/2
- ❑ Magnetic moments lie in a plane



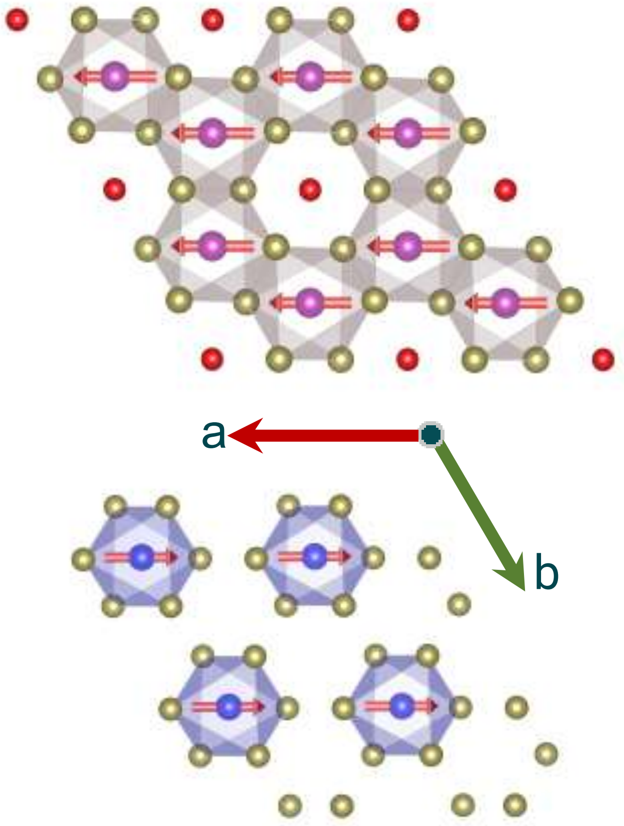
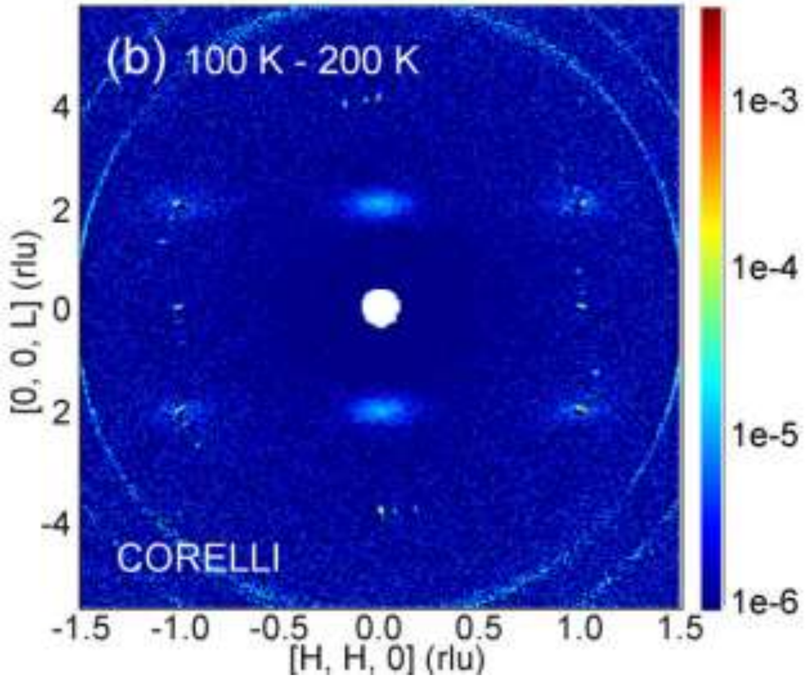


Fig. generated using VESTA
May et al., PRB 95, 174440 (2017)

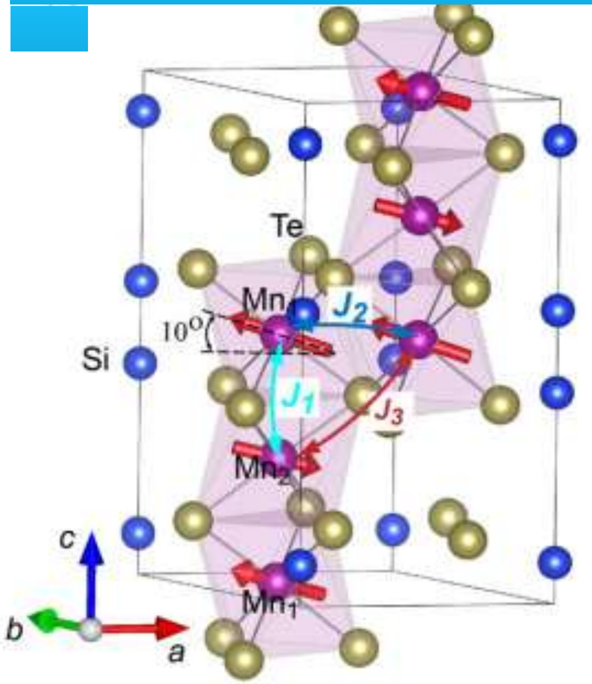
Introduction to $Mn_3Si_2Te_6$

Observation of short-range order, field induced spin canting, and colossal magnetoresistance has provided a recent interest on this material.

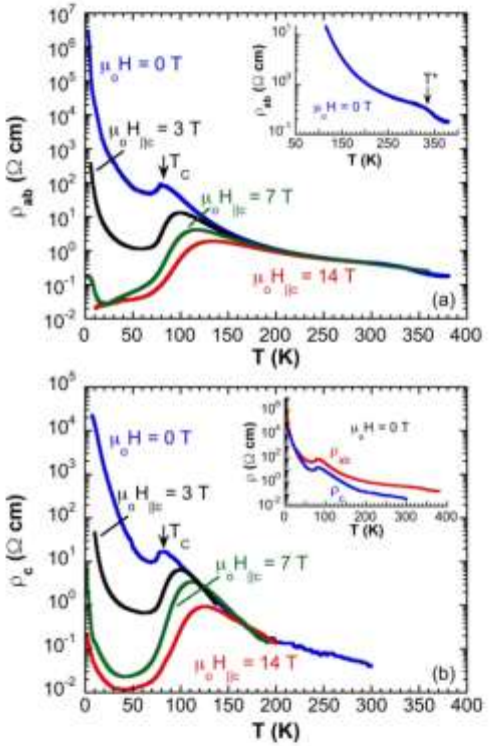
Short-range order



Field induced spin canting



Colossal magnetoresistance



Ye et al., PRB 106, L180402 (2022)

Ni et al., PRB 103, L161105 (2021)

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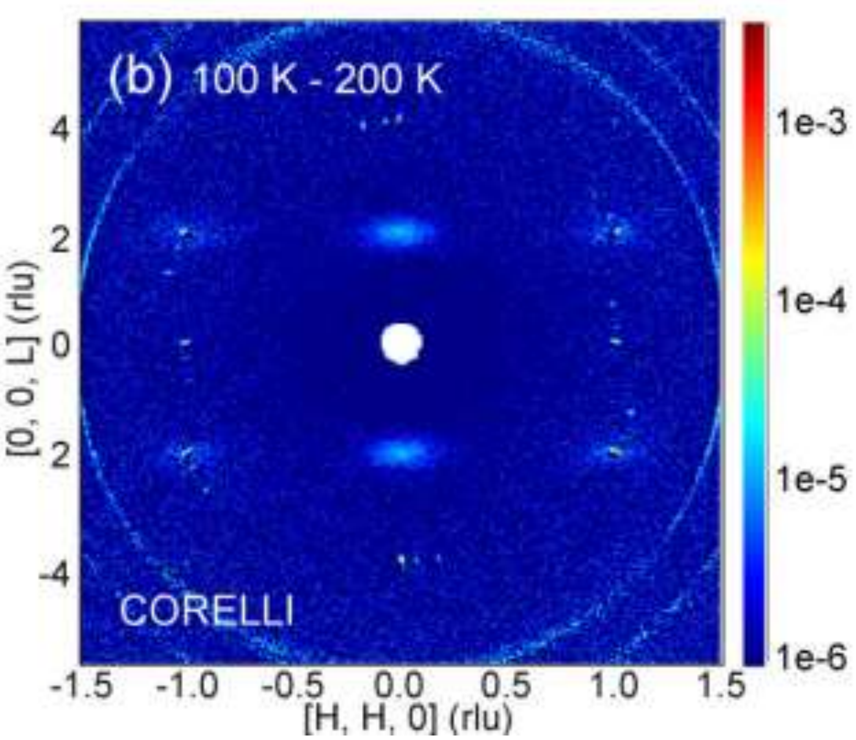
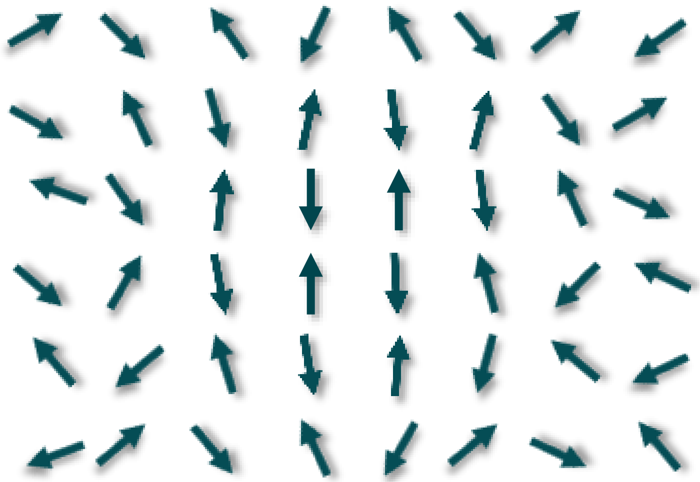
Objectives

- ❑ To study the diffuse scattering or short-range order in real space

- ❑ Conduct magnetic PDF analysis of the neutron total scattering data to probe short-range magnetic correlations at different temperatures

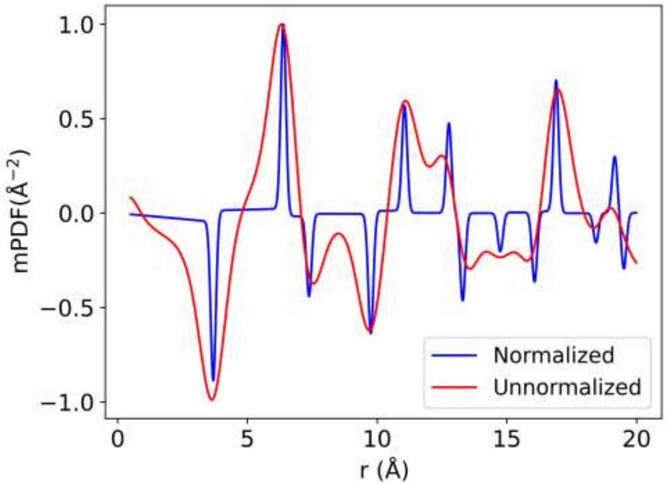




$$\langle S_0 \cdot S_i \rangle (r) \sim e^{-r/\xi}$$

$\xi = exponential\ correlation\ length$

mPDF techique

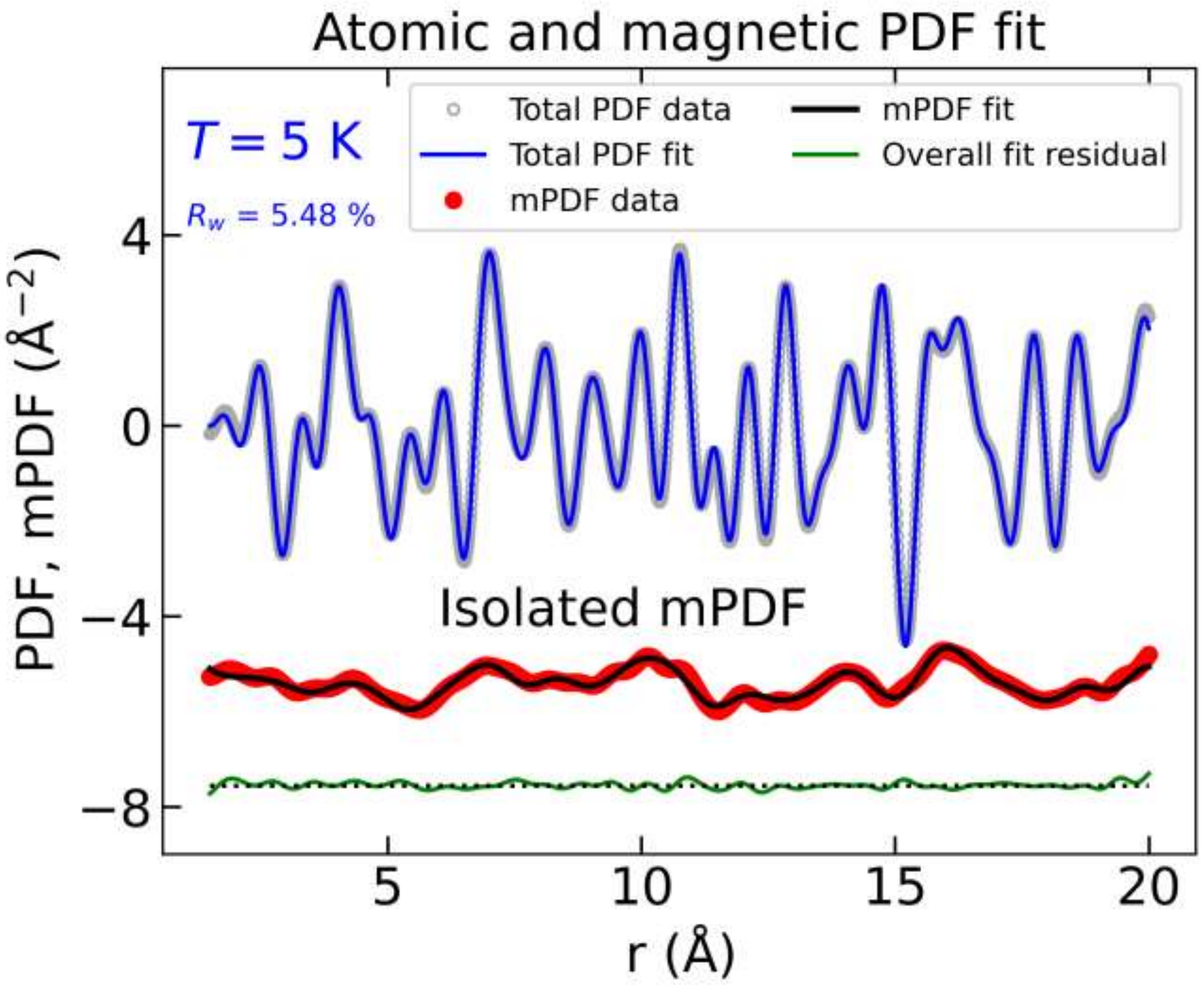


Frandsen et al., J. Appl. Cryst. 55, 1377-1382

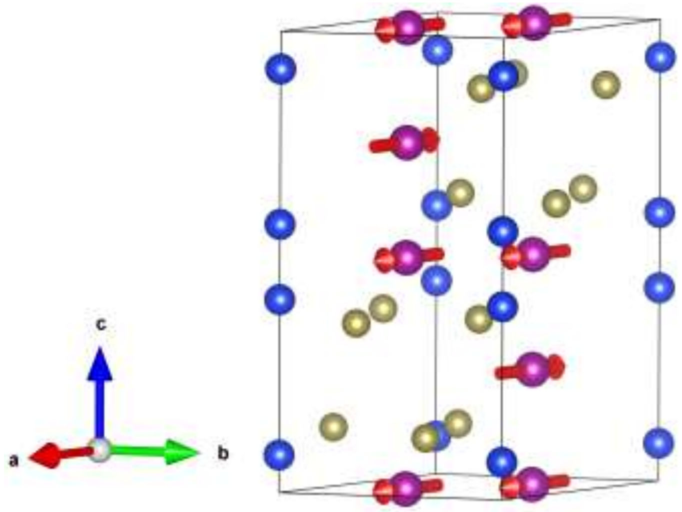
Ye et al., PRB 106, L180402 (2022)

Atomic and magnetic PDF fit at 5 K

Data collected at HB-2A



Baral et al. PRB ***112****, 024405*



Spin-angle = 90º with c-axis

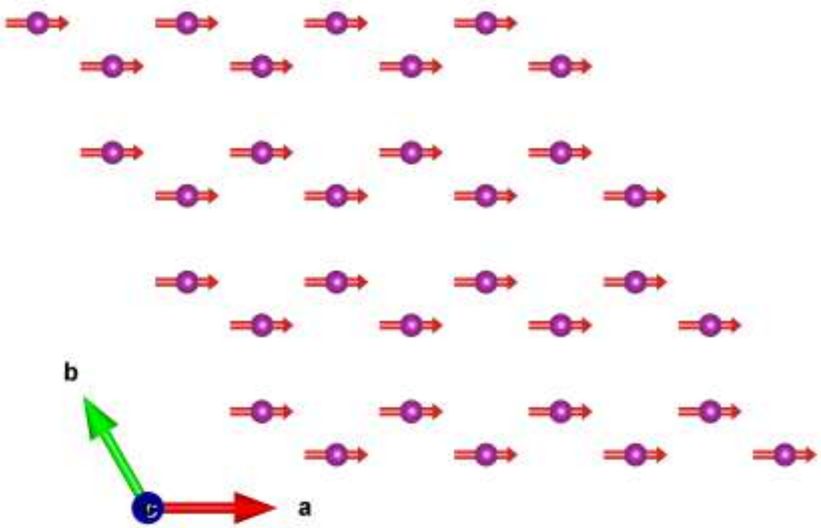
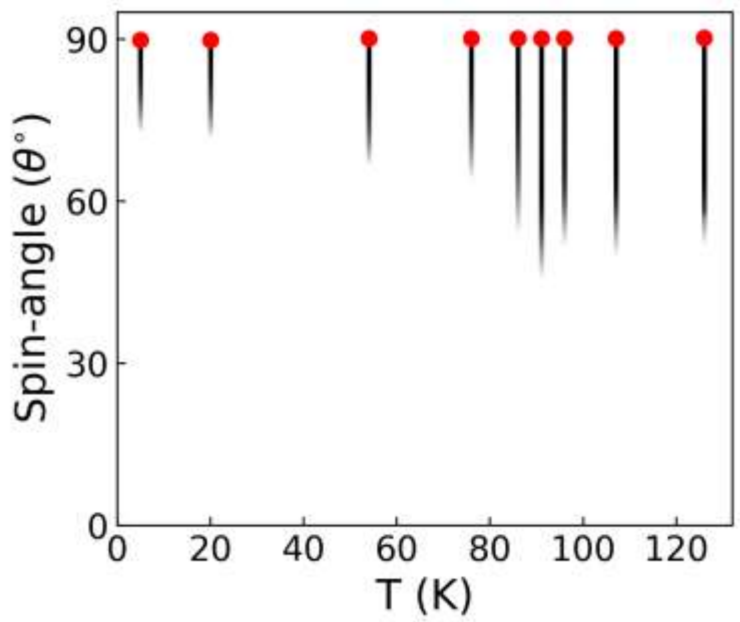


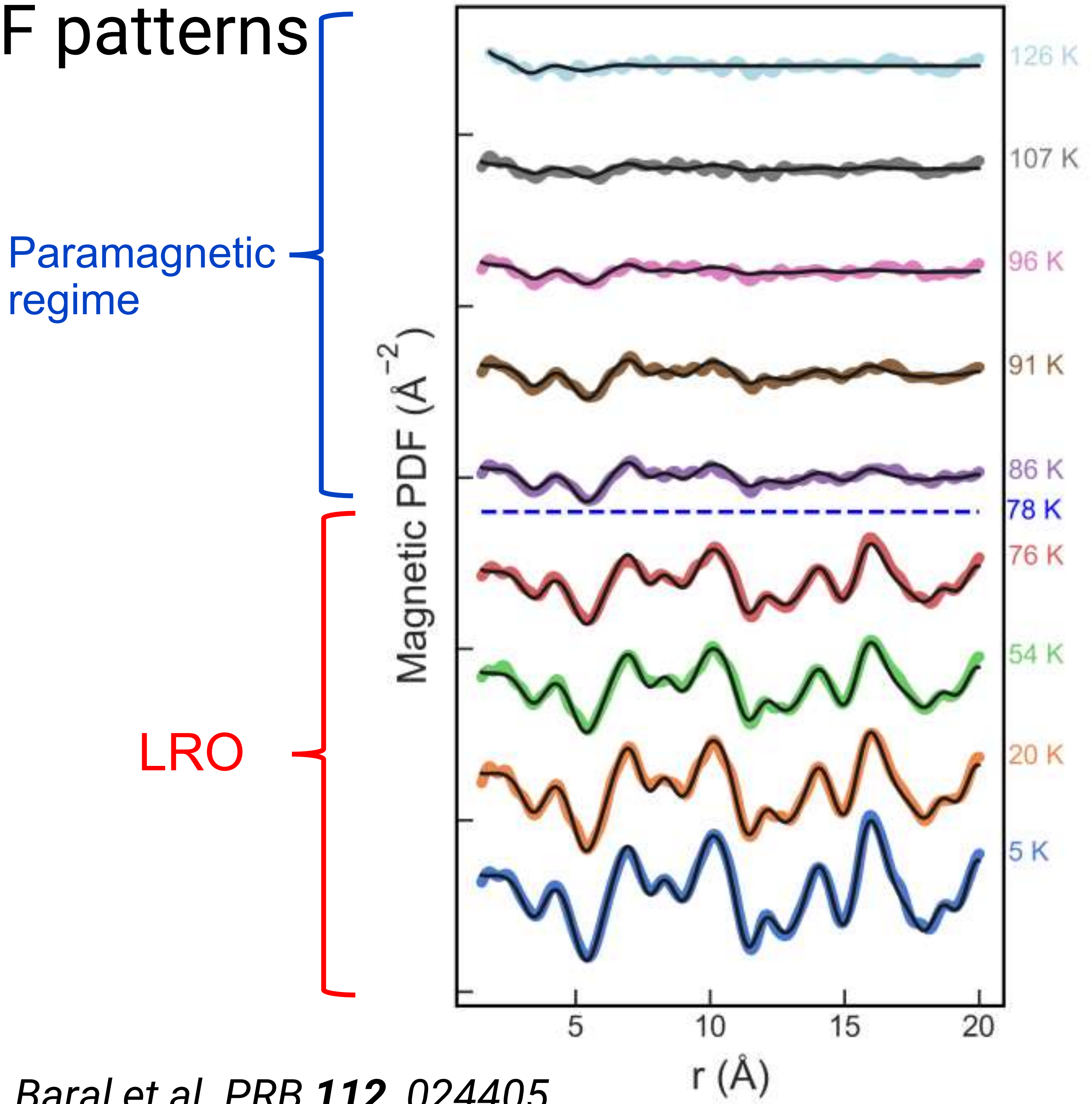
Fig. generated using VESTA

Temperature-dependent mPDF patterns

Data collected at HB-2A

- ❑ Fitting range: 1.5 – 20 Å
- ❑ Transition temperature = 78 K
- ❑ Short-range correlations above 78 K
- ❑ Modeled paramagnetic short-range correlations directly in real space using magnetic PDF





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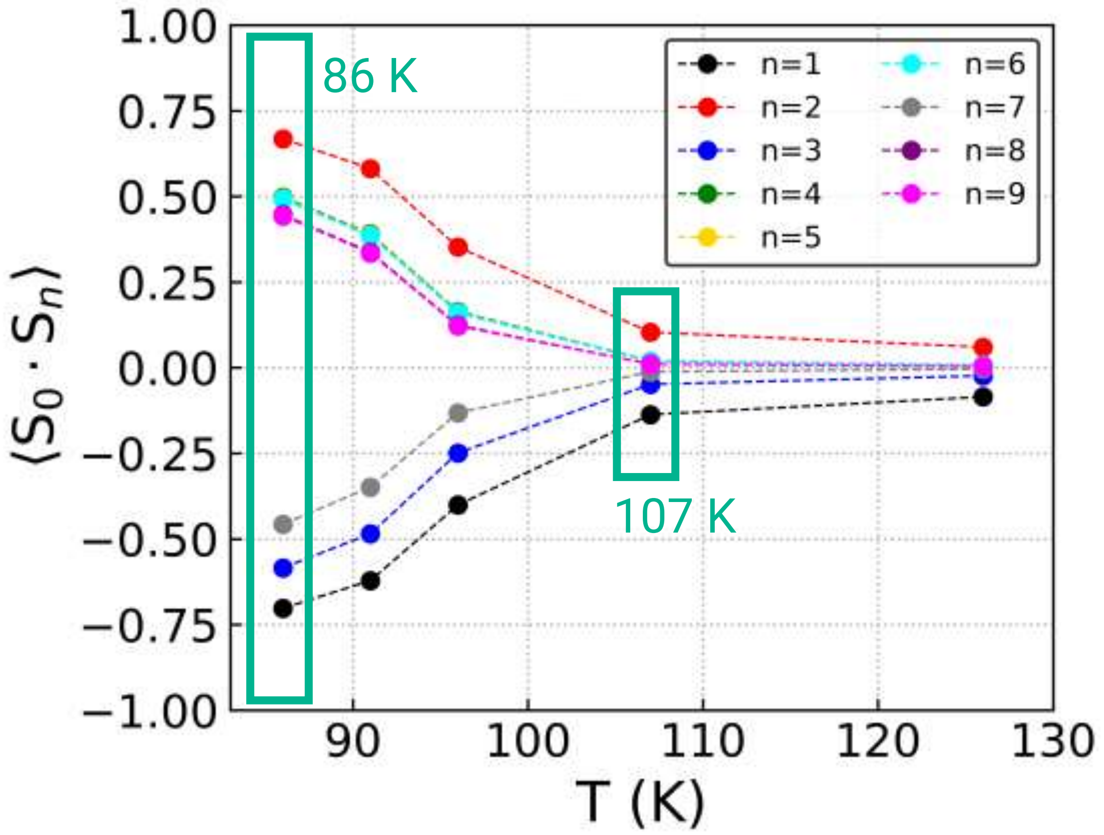
Average spin correlation function



- Negative values indicate net AFM spin directions while positive values indicate net FM spin directions; spins in the calculation have unit length
- At 86 K, spin correlations alternate as AFM (n=1), FM (n=2), and AFM (n=3) for the first three nearest-neighbor shells
- The spin correlation function decreases as temperature rises, consistent with expected paramagnetic behavior
- Even at elevated temperatures (107 K and 126 K), first-, second-, and third-nearest-neighbor correlations remain identifiable as AFM, FM, and AFM

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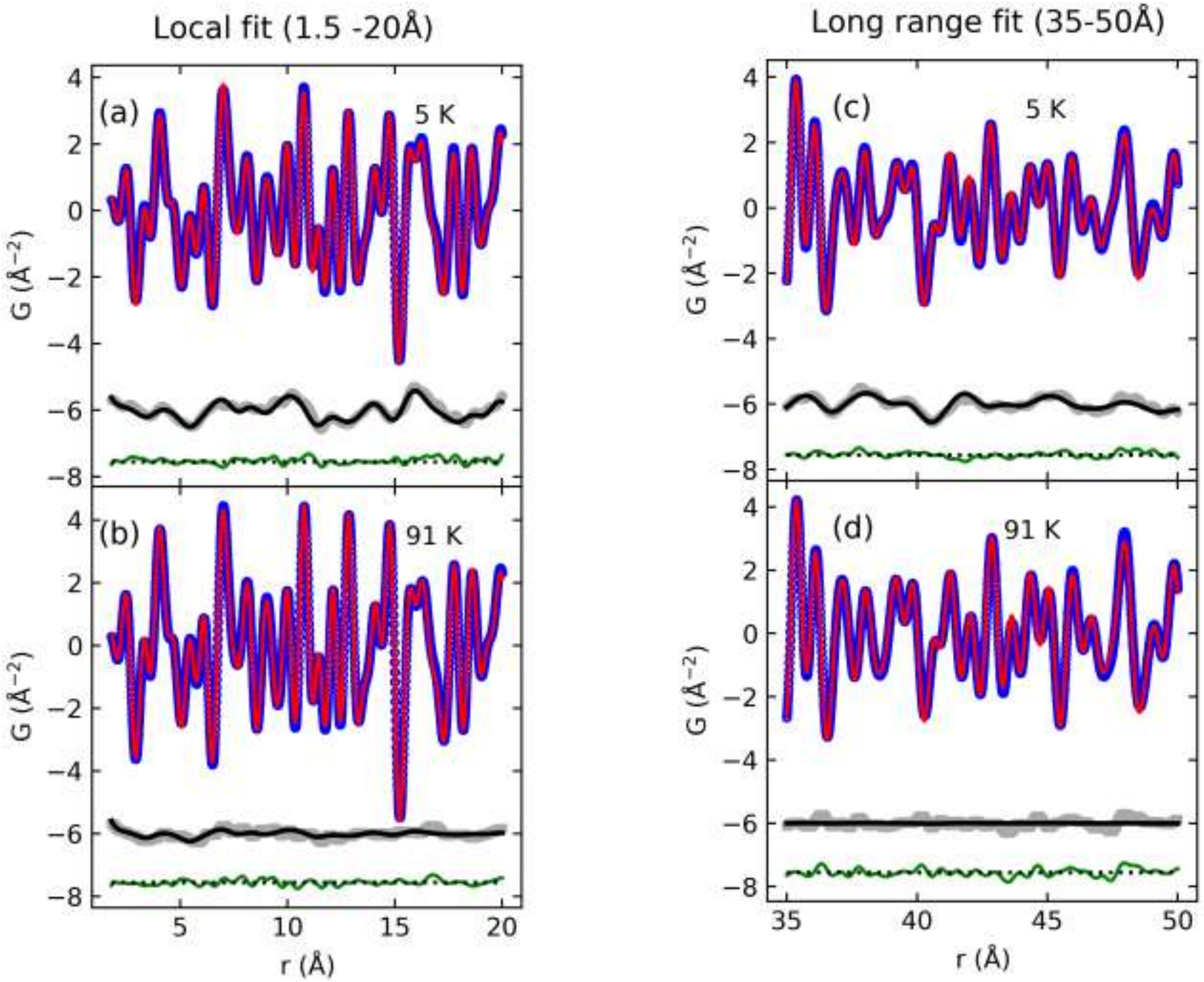
Local vs. Long-range fits

- ❑ Low-r fits or Local fits
- 1.5 – 20 Å

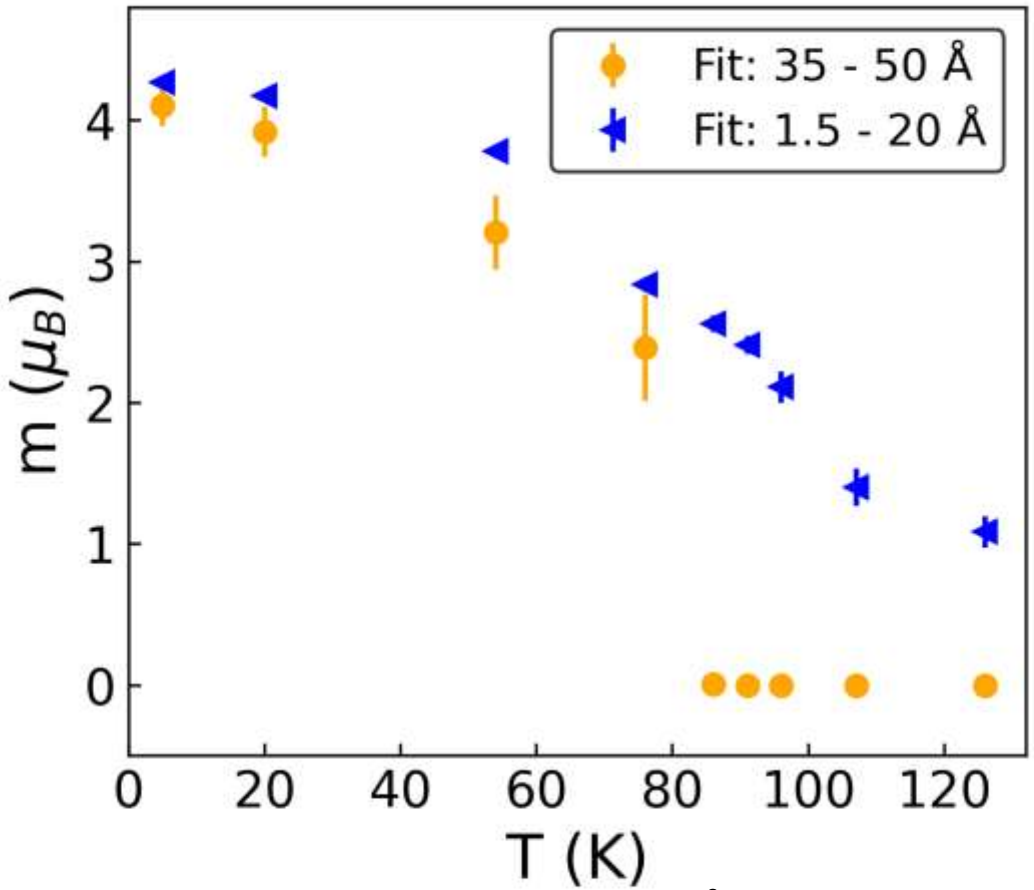
- ❑ High-r fits or Long-range Fits
- 35-50 Å

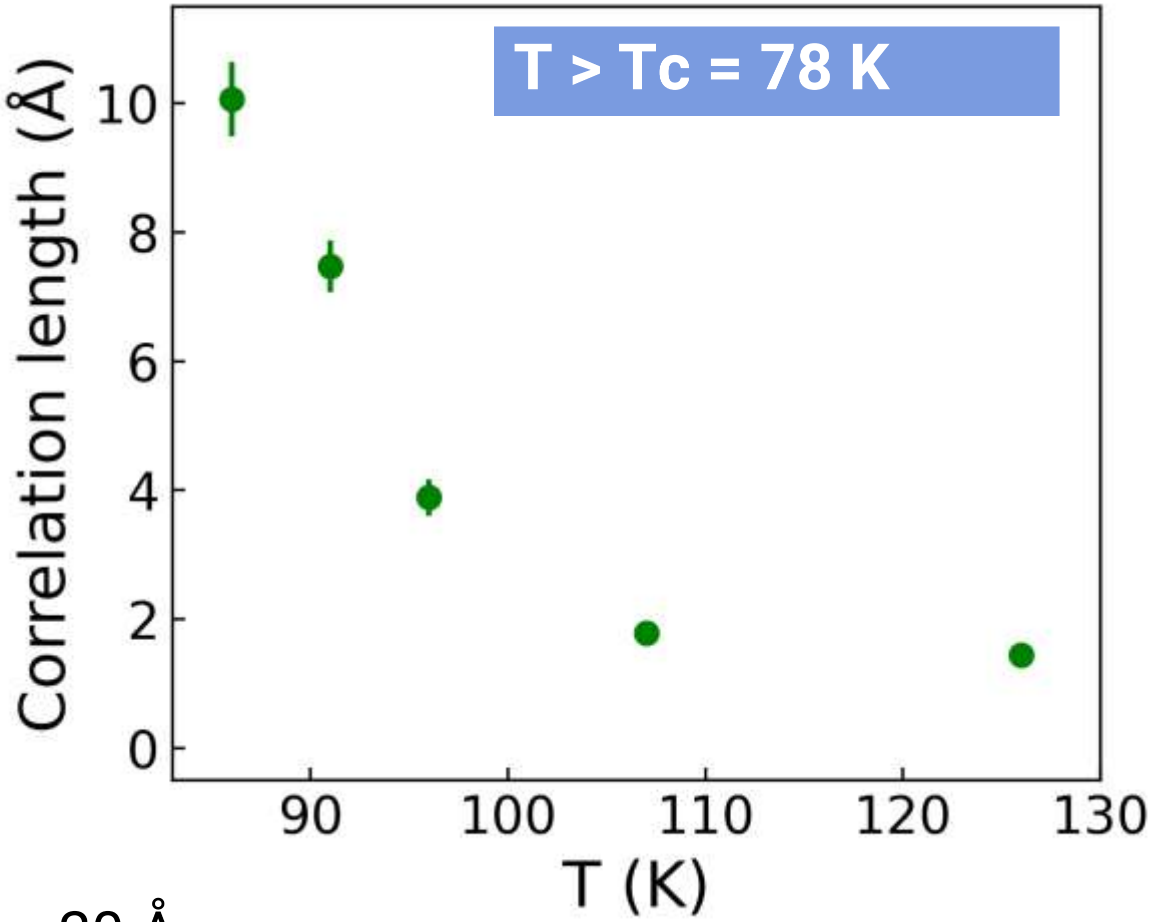
- ❑ Local fits captured the short-range magnetic correlations at paramagnetic regime



*Baral et al. PRB **112**, 024405*

Ordered moment and correlation length

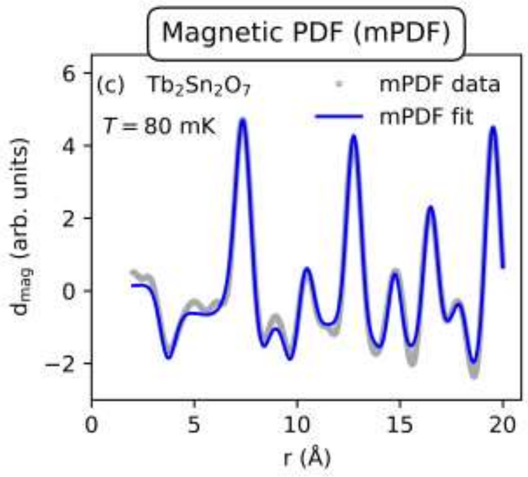


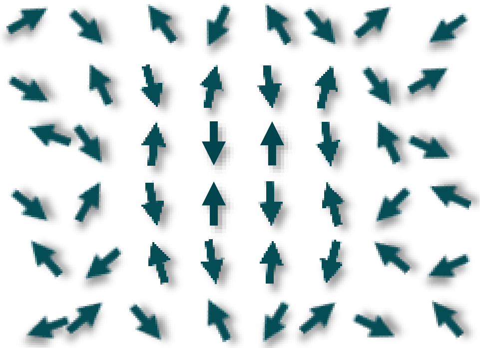


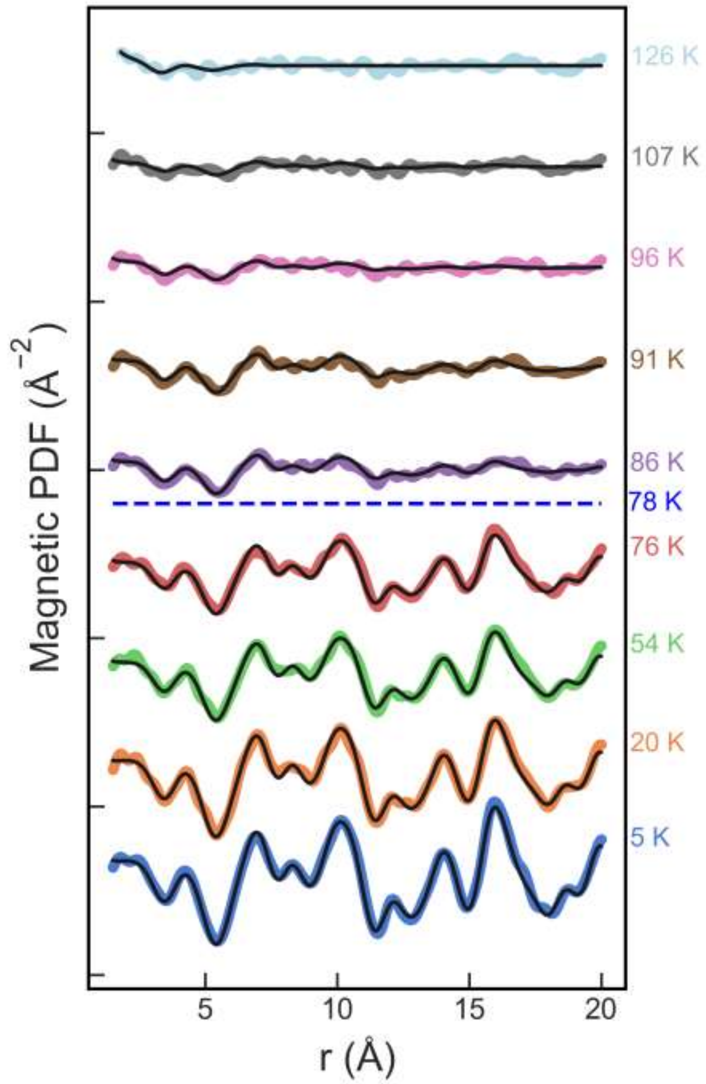
- ❑ High-r fit: 35 – 50 Å and low-r fit: 1.5 – 20 Å
- ❑ Ordered moment decreases with rise in temperature and remains non-zero at 126 K with low-r fit. Significant fraction of moment remains correlated
- ❑ Quantifies the persistence of significant short-range magnetic correlations

Concluding thoughts

- mPDF has been successfully applied to different magnetic materials to model short-range and long-range order
- ***diffpy.mpdf*** software tools available for the mPDF analysis
- mPDF tutorials:
  https://github.com/FrandsenGroup/mPDF-tutorial
- mPDF Installation:
  https://github.com/FrandsenGroup/diffpy.mpdf
- baralr@ornl.gov







Baral et al. PRB ***112****, 024405 (2025)*
Baral et. al., JMMM 630, 173342 (2025)

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