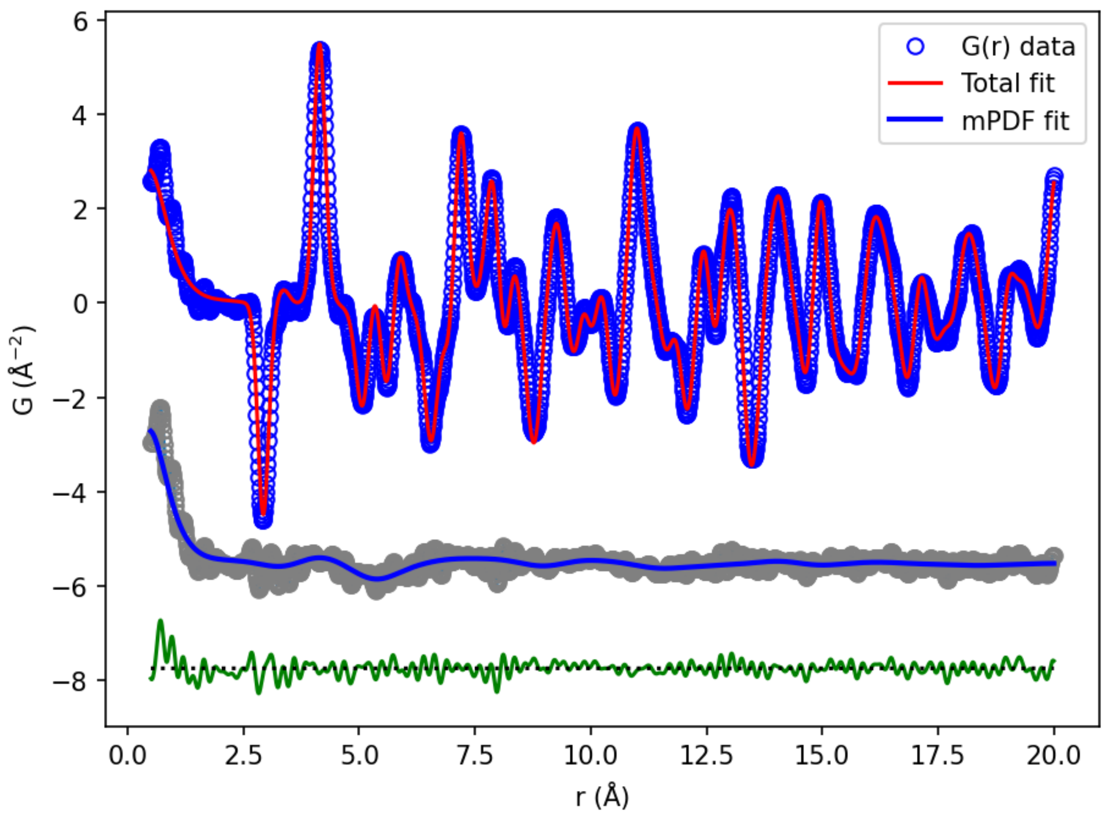
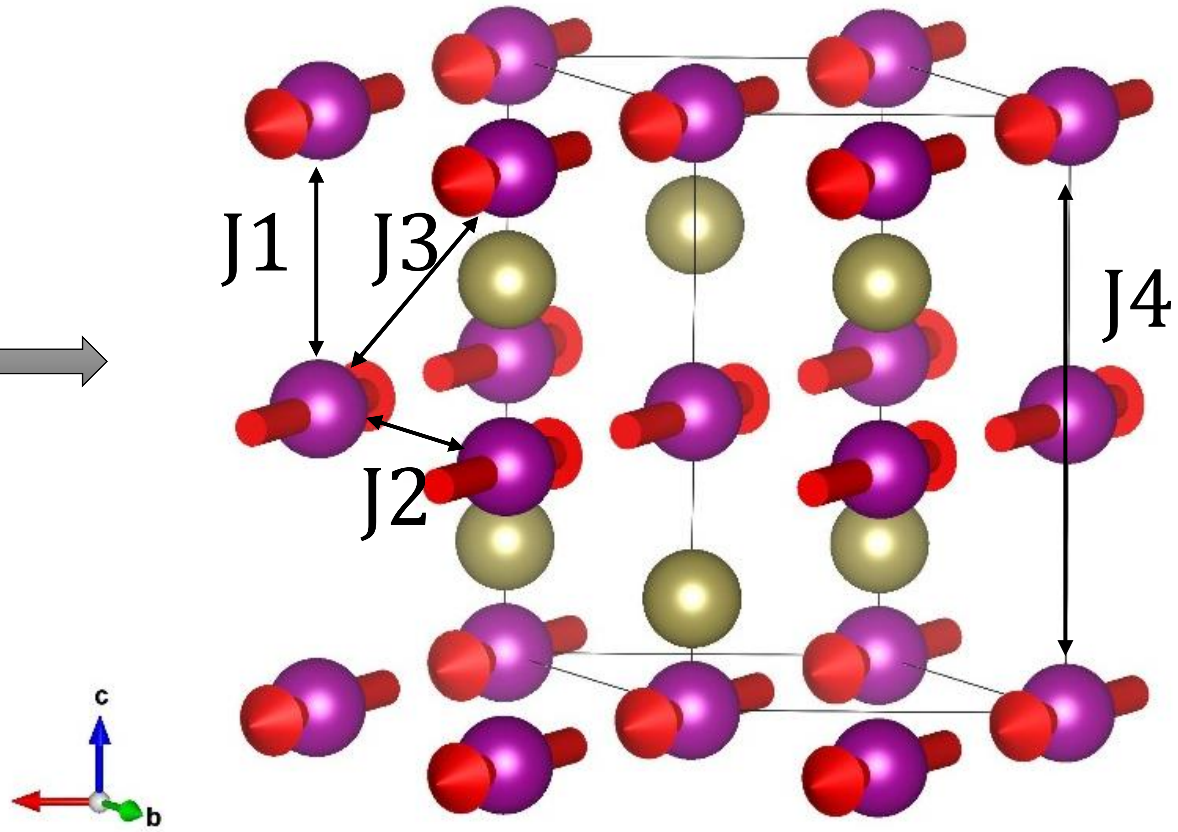
Using mPDF to probe magnetic exchange interactions





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Acknowledgments

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mPDF analysis: Sabrina Hatt, Raju Baral (now at ORNL)

ORNL beamline scientists: Stuart Calder, Michelle Everett, Jue Liu, and Jorg Neuefeind

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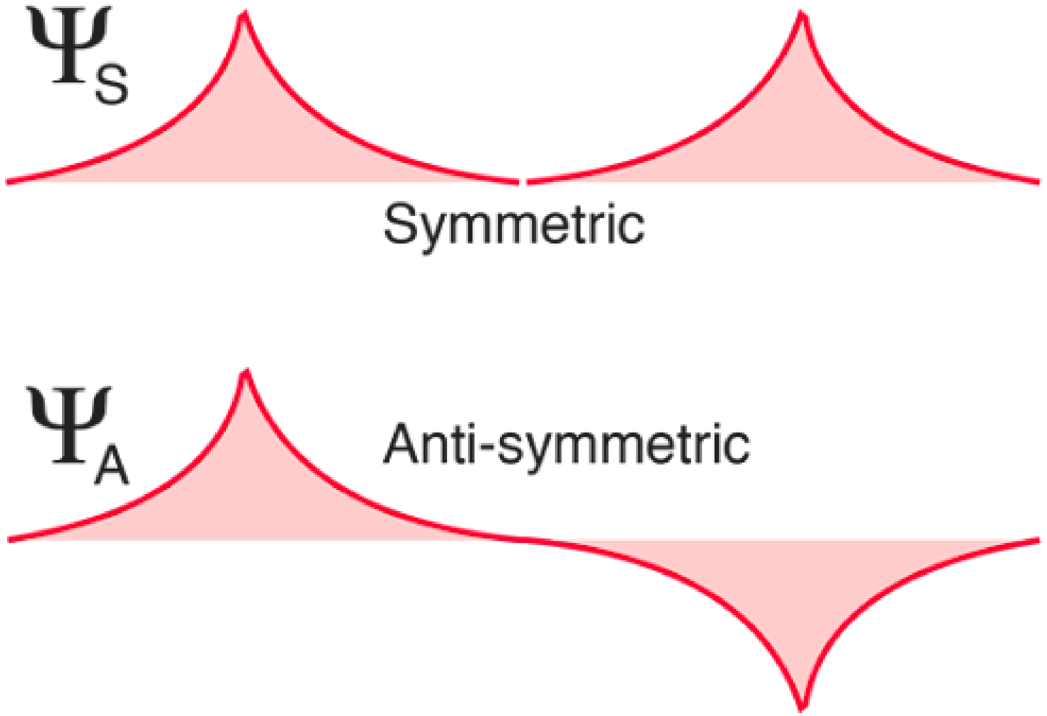
Understanding magnetic properties

- Conventional magnetic diffraction studies focus on:
  - Spin orientation, propagation vectors, ordered moment magnitude, etc
- We can also study interactions between spins
  - Understand magnetic Hamiltonian
  - Directly impacts material properties
- E.g. MnTe: exchange interactions directly influence
  - **Altermagnetism** (influence magnetic order)
  - **Thermoelectric properties** (thermopower influenced by paramagnon drag)
  - **Magnetostructural properties** (magnetism coupled to lattice parameters)

Magnetic Exchange Interaction

Consider a quantum system with two spin ½ electrons bound to two atoms

- **Fermions**: must be in anti-symmetric state
- If system is in Ψ_S, spins must be anti-aligned ($\boldsymbol{S}_1 \cdot \boldsymbol{S}_2 < 0$)
- If system is in Ψ_A, spins must be aligned ($\boldsymbol{S}_1 \cdot \boldsymbol{S}_2 > 0$)



http://hyperphysics.phy-astr.gsu.edu/hbase/molecule/hmol.html

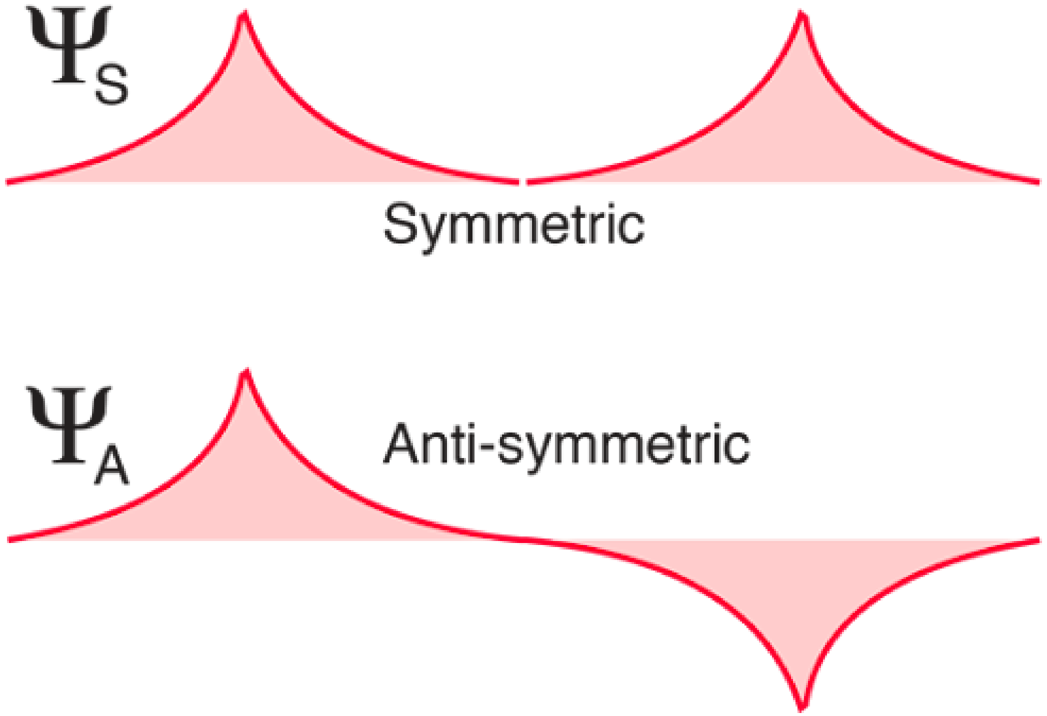
Magnetic Exchange Interaction

2-particle Fermionic wave function and energy:

$$\Psi_{S/A} = \frac{1}{\sqrt{2}}[\psi_a(\boldsymbol{r}_1)\psi_b(\boldsymbol{r}_2) \pm \psi_a(\boldsymbol{r}_2)\psi_b(\boldsymbol{r}_1)]\chi_{S/A}$$

$$E = E_1 + E_2 + C \pm X$$

- E_i is the energy of particle i,
- $C = \frac{e^2}{|\boldsymbol{r}_{12}|}$ is the coulomb energy of the two particles,
- and $\mathrm{X} = \int d\boldsymbol{r}_1 d\boldsymbol{r}_2 \psi_a(\boldsymbol{r}_1)\psi_b(\boldsymbol{r}_2)\frac{e^2}{|\boldsymbol{r}_{12}|}\psi_a^*(\boldsymbol{r}_2)\psi_b^*(\boldsymbol{r}_1)$ is the energy of exchanging the two particles
- + is for symmetric states and – is for antisymmetric states



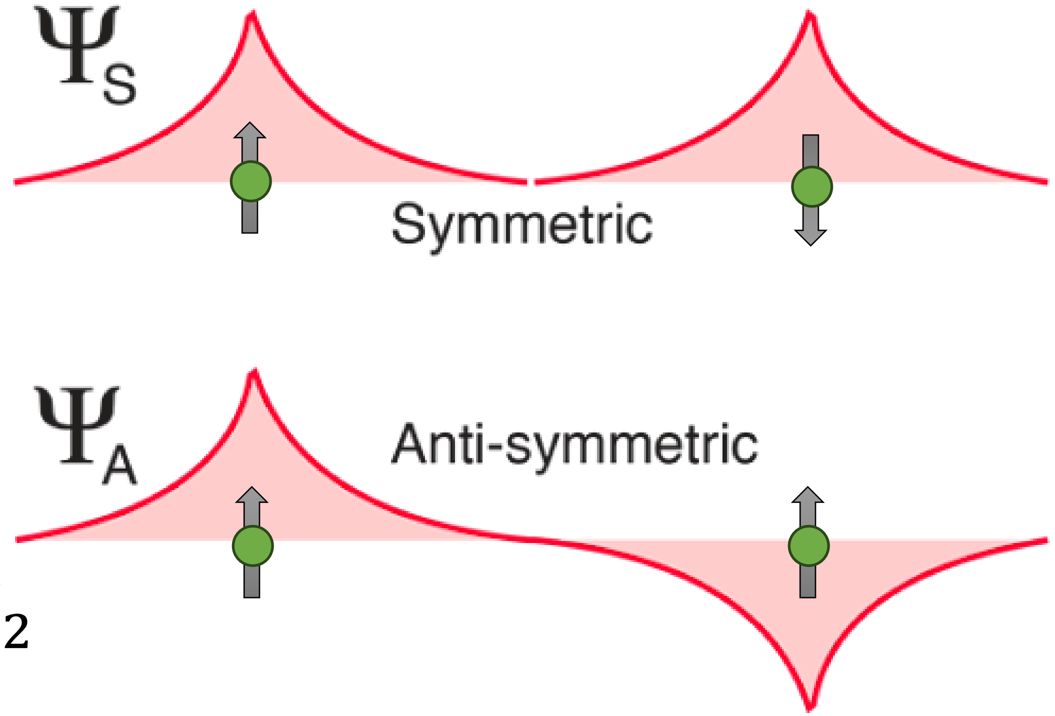
http://hyperphysics.phy-astr.gsu.edu/hbase/molecule/hmol.html

Magnetic Exchange Interaction

Put in terms of spin instead!

- Ψ_S: $\boldsymbol{S}_1 \cdot \boldsymbol{S}_2 < 0$
- Ψ_A: $\boldsymbol{S}_1 \cdot \boldsymbol{S}_2 > 0$
- $\boldsymbol{S}_1 \cdot \boldsymbol{S}_2 = \frac{1}{2}(\boldsymbol{S}_{tot}^2 - \boldsymbol{S}_1^2 - \boldsymbol{S}_2^2)$
  - $= \frac{1}{2}[\mathrm{s}(\mathrm{s}+1) - \frac{3}{4} - \frac{3}{4}]$
  - $= -\frac{3}{4}$ for $s = 0$ (symmetric)
  - $= \frac{1}{4}$ for $s = 1$ (antisymmetric)

$$E = \frac{1}{4}(E_S + 3E_A) - \underbrace{(E_S - E_A)}_{J_{12}\text{: Exchange interaction strength!}}\boldsymbol{S}_1 \cdot \boldsymbol{S}_2$$

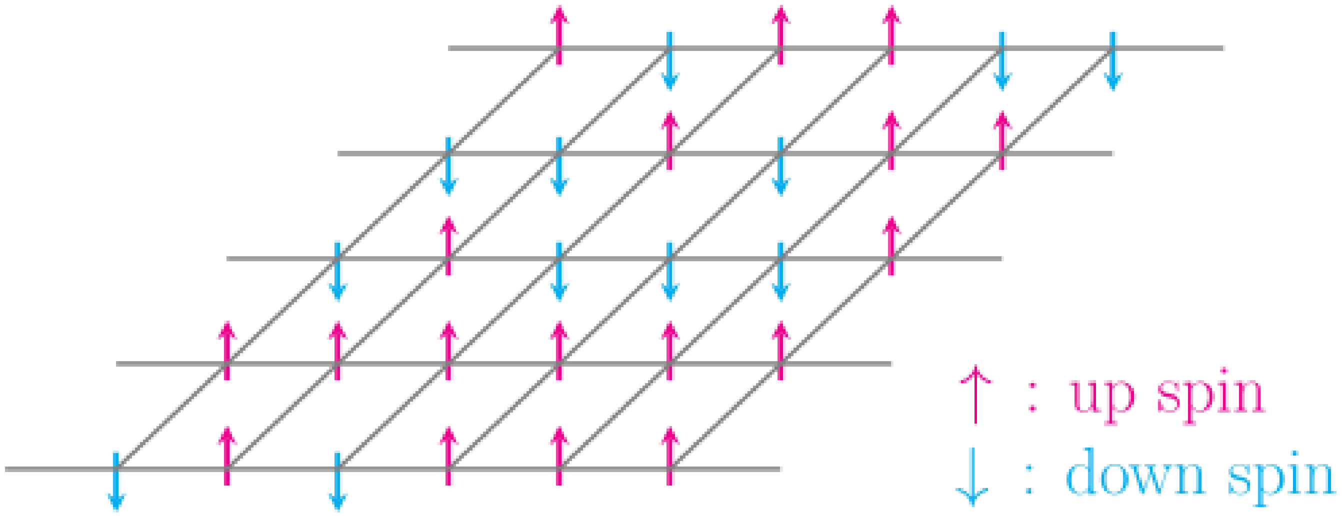


http://hyperphysics.phy-astr.gsu.edu/hbase/molecule/hmol.html

Magnetic Exchange Interaction

Ignore parts independent of spin ("constant" energy)

- $E = -J_{12}\,\boldsymbol{S}_1 \cdot \boldsymbol{S}_2$
- $J_{12} > 0$: **ferromagnetism** favorable
- $J_{12} < 0$: **antiferromagnetism** favorable
- Expand to solid state systems:
  - $E = -\frac{1}{2}\sum_{ij} J_{ij}\boldsymbol{S}_i \cdot \boldsymbol{S}_j$ (Heisenberg)



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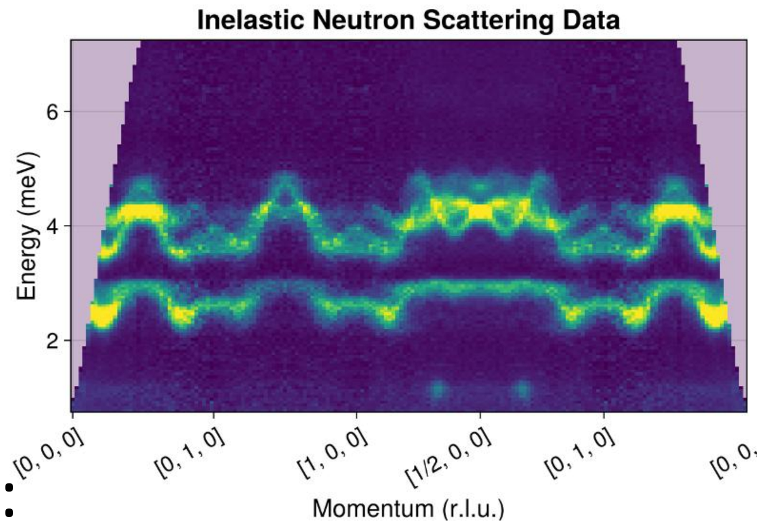
Magnetic Exchange Interactions

How do we find exchange interaction values?

- **Ordered state** (common approach):
  - Inelastic neutron scattering of single-crystal data
  - Fit linear spin-wave model to observed magnon modes
  - Requires ordered state (no spin glasses or spin liquids)
- **Disordered state** (newer approach, e.g. Spinteract*):
  - Diffuse magnetic scattering
  - Models scattering with mean field theory
  - Requires paramagnetic state (above magnetic ordering temp)



Bai et al., Nature Physics, 17 (4), 467–472 (2021)

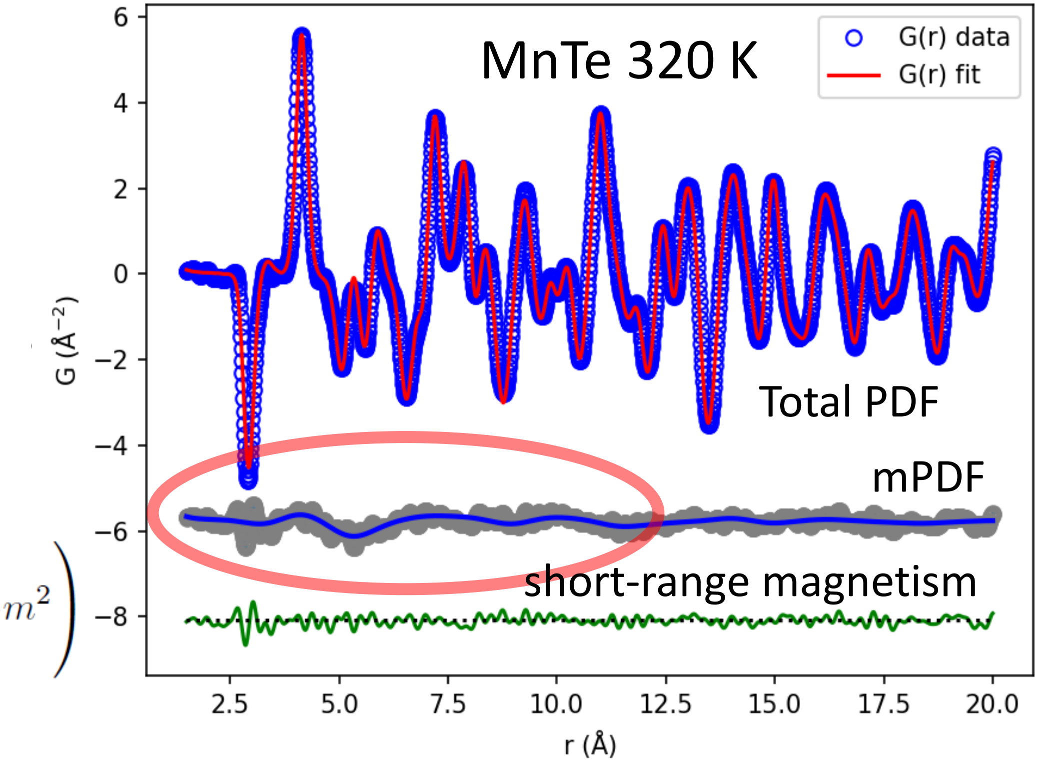
*Joseph A M Paddison 2023 *J. Phys.: Condens. Matter* **35** 495802

Exchange Interactions + mPDF

Finding exchange interactions from diffuse scattering?

- Requires short-range magnetic order!
- **mPDF**: a natural way to study short-range magnetism

$$G_{\mathrm{mag}}(r) = \frac{3}{2\langle g\sqrt{J(J+1)}\rangle^2}\left(\frac{1}{N_s}\sum_{i\neq j} g_i g_j \left[\frac{A_{ij}}{r}\delta(r-r_{ij}) + B_{ij}\frac{r}{r_{ij}^3}\Theta(r_{ij}-r)\right] - 4\pi r\rho_0\frac{2}{3}m^2\right)$$



$T_{\mathrm{N}} = 307$ K

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

Road map

- Math ahead!
- Fun examples and results coming soon

How to find exchange interactions from mPDF?

Start with Hamiltonian

$$-\underbrace{\frac{1}{2}\sum_{ij}\sum_{\boldsymbol{R},\boldsymbol{R}'} J_{ij}(\boldsymbol{R}-\boldsymbol{R}')\boldsymbol{S}_i(\boldsymbol{R})\cdot\boldsymbol{S}_j\,(\boldsymbol{R}')}_{\text{Same as before, but now with dependence on } \boldsymbol{R} \text{ (unit cell position) and } i,j \text{ index spins w/in UC}} - \underbrace{\sum_{i,\boldsymbol{R}} \boldsymbol{H}_i(\boldsymbol{R})\cdot\boldsymbol{S}_i(\boldsymbol{R})}_{\text{Site-dependent magnetic field, will set to 0 later}}$$

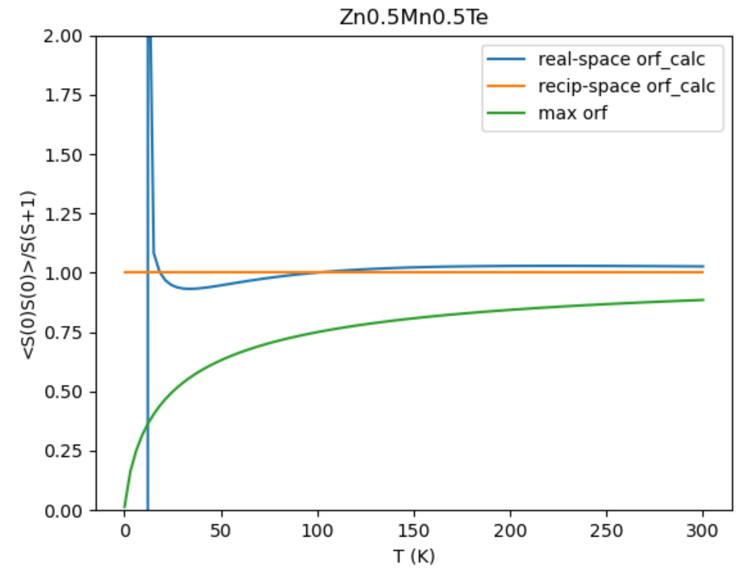
Define mean field on site $\boldsymbol{R}, i$

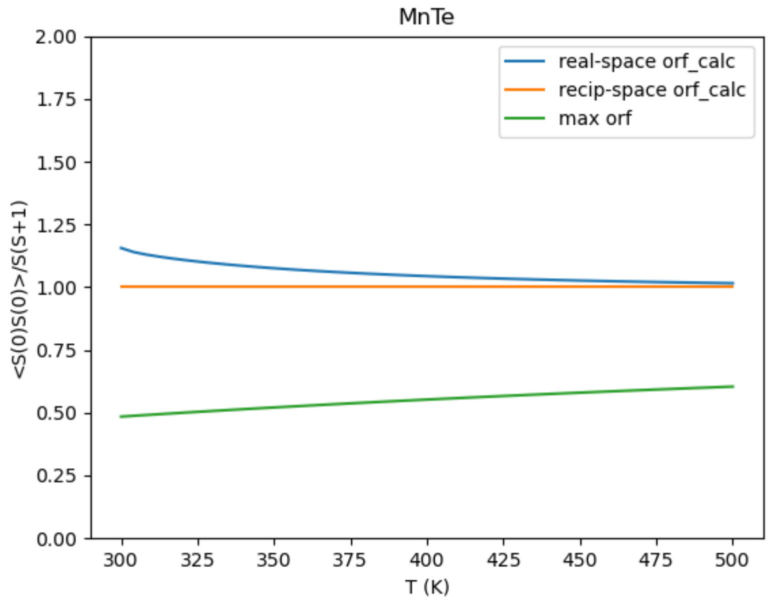
$$\boldsymbol{H}_{mf,i}(\boldsymbol{R}) = \boldsymbol{H}_i(\boldsymbol{R}) - \underbrace{\lambda\langle\boldsymbol{S}_i(\boldsymbol{R})\rangle}_{\text{Onsager Reaction Field}} + \frac{1}{2}\sum_{j,\boldsymbol{R}'} J_{ij}(\boldsymbol{R}-\boldsymbol{R}')\langle\boldsymbol{S}_j(\boldsymbol{R}')\rangle$$

How to find exchange interactions from mPDF?

Onsager Reaction Field (ORF):

- Removes influence of spin on mean field the spin feels
- $\lambda = \sum_{j,\boldsymbol{R}'} J_{ij}(\boldsymbol{R} - \boldsymbol{R}')\langle \boldsymbol{S}_i(\boldsymbol{R}) \cdot \boldsymbol{S}_j(\boldsymbol{R}')\rangle / |\boldsymbol{S}|^2$
- Usually found through reciprocal space sum rule
  - $\sum_q \langle \boldsymbol{S}(\boldsymbol{q}) \cdot \boldsymbol{S}(-\boldsymbol{q})\rangle(\lambda) = N_q S(S+1)$
- However, ORF is defined by real-space correlations
  - Can instead find $\langle \boldsymbol{S}_i(\boldsymbol{R}) \cdot \boldsymbol{S}_j(\boldsymbol{R}')\rangle(\lambda)$ and solve eq. above as transcendental eq.





How to find exchange interactions from mPDF?

Next, find $\boldsymbol{q}$-dependent magnetic susceptibility

- Remember, $\chi_{ij}^{\alpha\beta}(\boldsymbol{q}) = \langle S_i^{\alpha}(\boldsymbol{q})\rangle / H_j^{\beta}(\boldsymbol{q}) = \frac{1}{T}\langle S_i^{\alpha}(\boldsymbol{q}) S_j^{\beta}(-\boldsymbol{q})\rangle$
    - Gives us a correlation function!
- For paramagnetic spin in magnetic field $\langle \boldsymbol{S}_i(\boldsymbol{R})\rangle = \frac{|\boldsymbol{S}|^2}{\underbrace{3T}_{\text{Curie Susceptiblity } \chi_0}} \boldsymbol{H}_i(\boldsymbol{R})$
- Replace magnetic field with mean field used earlier:
    - $\langle \boldsymbol{S}_i(\boldsymbol{R})\rangle = \chi_0[\boldsymbol{H}_i(\boldsymbol{R}) - \lambda\langle \boldsymbol{S}_i(\boldsymbol{R})\rangle + \frac{1}{2}\sum_{j,\boldsymbol{R}'} J_{ij}(\boldsymbol{R} - \boldsymbol{R}')\langle \boldsymbol{S}_j(\boldsymbol{R}')\rangle]$

How to find exchange interactions from mPDF?

Simplify the problem! Fourier transform ($\boldsymbol{R} \rightarrow \boldsymbol{q}$) and diagonalize ($\alpha, \beta, i, j \rightarrow \mu$) then rearrange to solve for $\langle S_\mu(\boldsymbol{q}) \rangle$

$$\langle S_\mu(\boldsymbol{q}) \rangle = \frac{\chi_0}{1 - \chi_0 (J_\mu(\boldsymbol{q}) - \lambda)} H_\mu(\boldsymbol{q})$$

or equivalently,

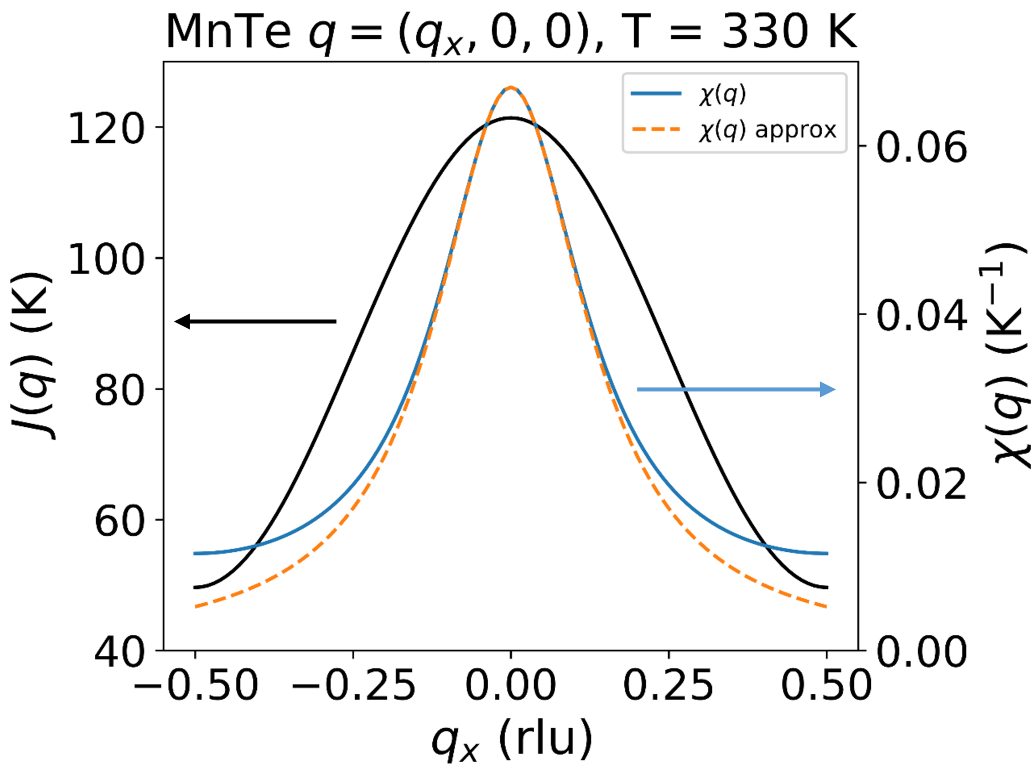
$$\chi_\mu(\boldsymbol{q}) = \frac{\chi_0}{1 - \chi_0 (J_\mu(\boldsymbol{q}) - \lambda)}$$

So, our correlation function becomes

$$\langle S_\mu(\boldsymbol{q}) S_\mu(-\boldsymbol{q}) \rangle = T \frac{\chi_0}{1 - \chi_0 (J_\mu(\boldsymbol{q}) - \lambda)}$$

This is how Spinteract connects exchange interactions and diffuse scattering

How to find exchange interactions from mPDF?



Time to transform back to real space! Doing a general transform is hard, so let's Taylor expand $J_\mu(\boldsymbol{q})$.

- If we expand around a peak in $J_\mu(\boldsymbol{q})$ (call its position $\boldsymbol{q}_0$), then $\chi_\mu(\boldsymbol{q})$ becomes Lorentzian-like
- $J_\mu(\boldsymbol{q}) \approx J_\mu(\boldsymbol{q}_0)$ $+\frac{1}{2}\sum_{\alpha\beta}\partial_{q_\alpha}\partial_{q_\beta}J(\boldsymbol{q}_0)(q_\alpha - q_{0,\alpha})(q_\beta - q_{0,\beta})$

How to find exchange interactions from mPDF?

What do our correlation functions look like in real space?

- 1D: $\langle S_\mu(0)S_\mu(r)\rangle \approx \frac{\pi\sqrt{2}\,T}{L_{BZ}}\sum_{q_0}\frac{e^{-iq_0 r}}{\sqrt{-\partial q^2 J_\mu(q_0)}}\frac{\exp(-r\sqrt{D})}{\sqrt{\frac{1}{\chi_0}+\lambda-J_\mu(q_0)}}$
- 2D: $\langle S_\mu(0)S_\mu(\boldsymbol{r})\rangle \approx \frac{2\sqrt{2\pi}T}{A_{BZ}}\sum_{\boldsymbol{q}_0}\frac{e^{-i\boldsymbol{q}_0\cdot\boldsymbol{r}}}{\sqrt{\det C}}K_0(\sqrt{\boldsymbol{r}^T D\boldsymbol{r}})$
- 3D: $\langle S_\mu(0)S_\mu(\boldsymbol{r})\rangle \approx \frac{(2\pi)^2T}{V_{BZ}}\sum_{\boldsymbol{q}_0}\frac{e^{-i\boldsymbol{q}_0\cdot\boldsymbol{r}}}{\sqrt{-\det C}}\frac{\exp(-\sqrt{\boldsymbol{r}^T D\boldsymbol{r}})}{\sqrt{\boldsymbol{r}^T C^{-1}\boldsymbol{r}}}$

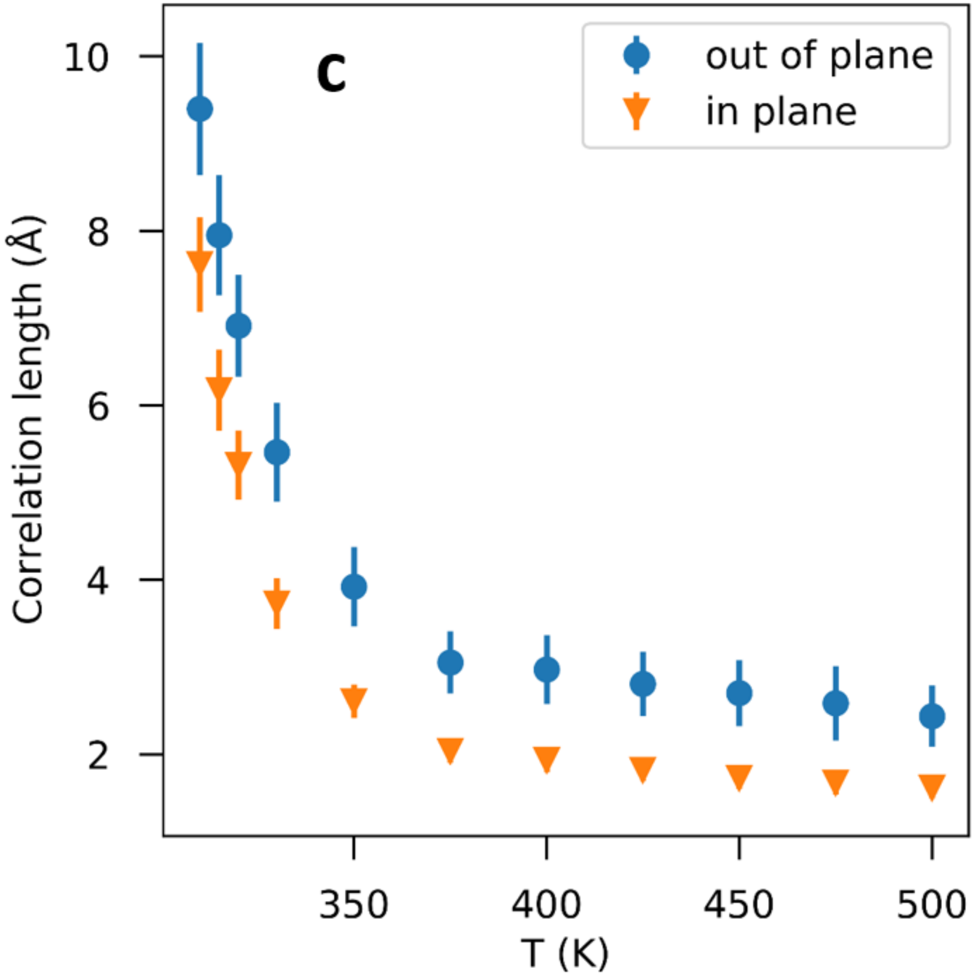
where $C_{\alpha\beta} = \partial_{q_\alpha}\partial_{q_\beta}J(\boldsymbol{q}_0)$ and $[D^{-1}]_{\alpha\beta} = \frac{-\frac{1}{2}\partial_{q_\alpha}\partial_{q_\beta}J_\mu(q_0)}{1/\chi_0+\lambda-J_\mu(q_0)}$

D is called the
damping matrix

How to find exchange interactions from mPDF?

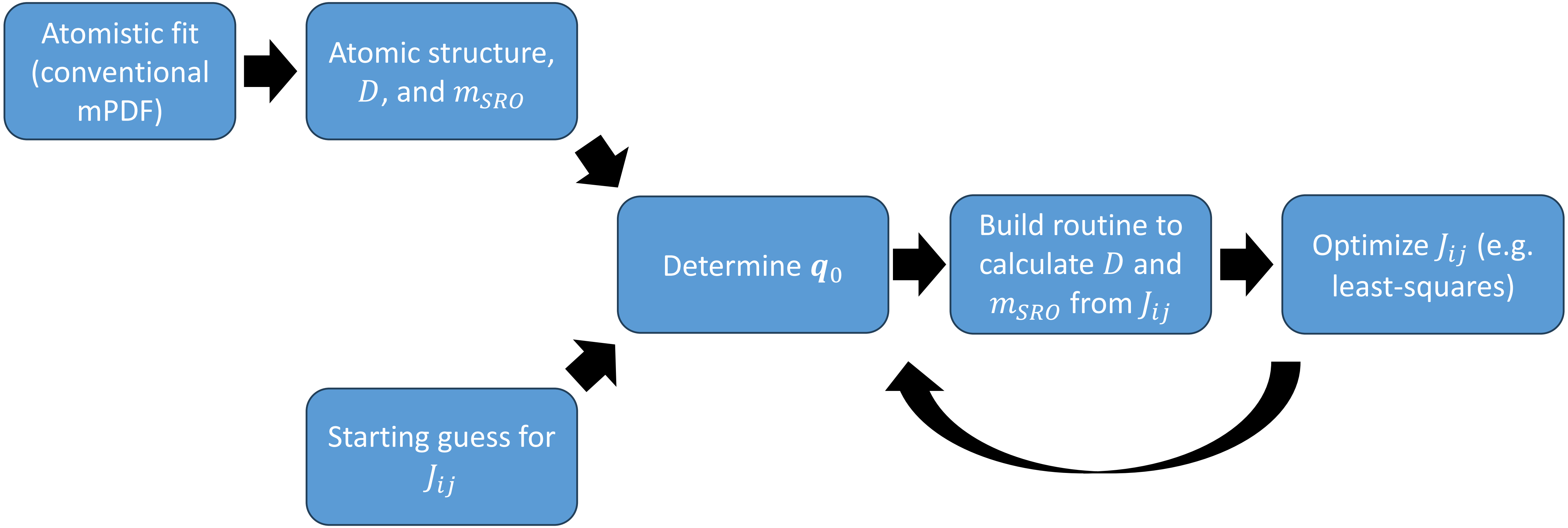
How do we relate these correlation functions to mPDF?

- $\boldsymbol{q}_0$ is the magnetic ordering vector!
- Use eigenvectors $U_{i\mu}(\boldsymbol{q}_0)$ to determine relative spin directions w/in UC
- Compare $m_{SRO} = g\sqrt{|\langle \boldsymbol{S}(0) \cdot \boldsymbol{S}(\boldsymbol{r}_{NN})\rangle|}$ from mPDF and magnetic interactions
- Compare damping matrices from mPDF and magnetic interactions
  - Anisotropic correlation lengths!



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

How to find exchange interactions from mPDF?



Atomistic fit (conventional mPDF)

Atomic structure, D, and m_{SRO}

Starting guess for J_{ij}

Determine $\boldsymbol{q}_0$

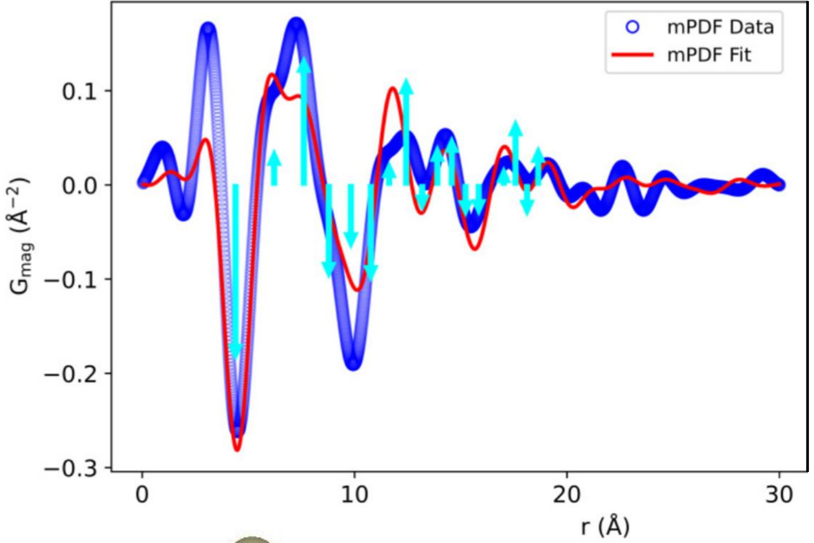
Build routine to calculate D and m_{SRO} from J_{ij}

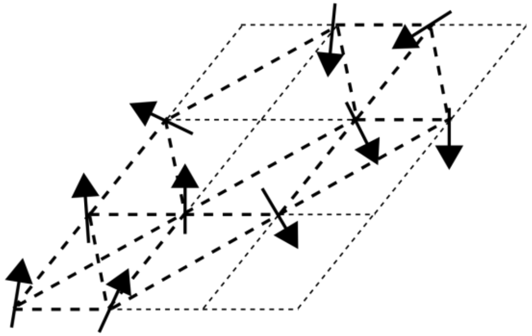
Optimize J_{ij} (e.g. least-squares)

Example 1: $Zn_{0.5}Mn_{0.5}Te$

- Cluster spin glass
  - No long-range order, $T_F \sim 22 - 25$ K
  - mPDF fits find local type III AFM order
- $S = 5/2$ for Mn2+
  - $|\boldsymbol{S}|^2 = S(S+1) = 35/4$
  - $|\boldsymbol{S}|^2 = 35/8$ for $Zn_{0.5}Mn_{0.5}Te$
- $J_1 = -9.5$ K, $J_2 = \frac{1}{5} J_1 = -1.9$ K
  - J. Furdyna and N. Samarth, J. Appl. Phys. 61, 3526 (1987)

S. R. Hatt, et al., Phys. Rev. B 112, 144440 (2025)





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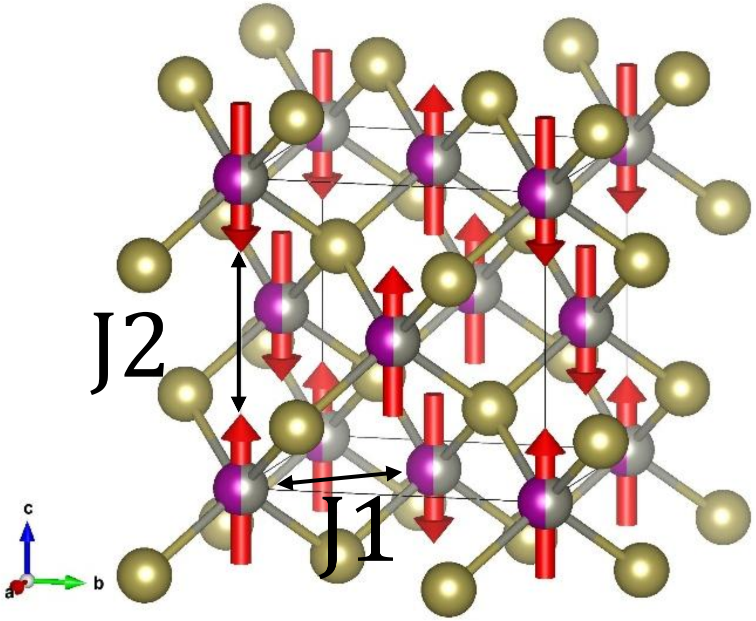
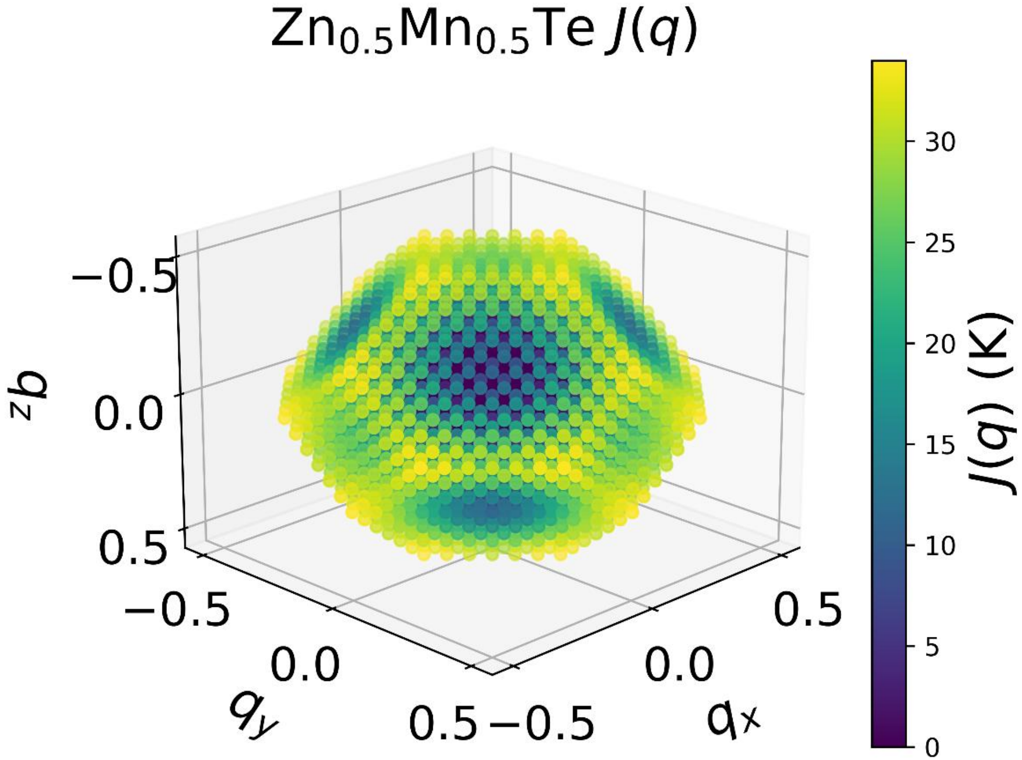


Example 1: $Zn_{0.5}Mn_{0.5}Te$

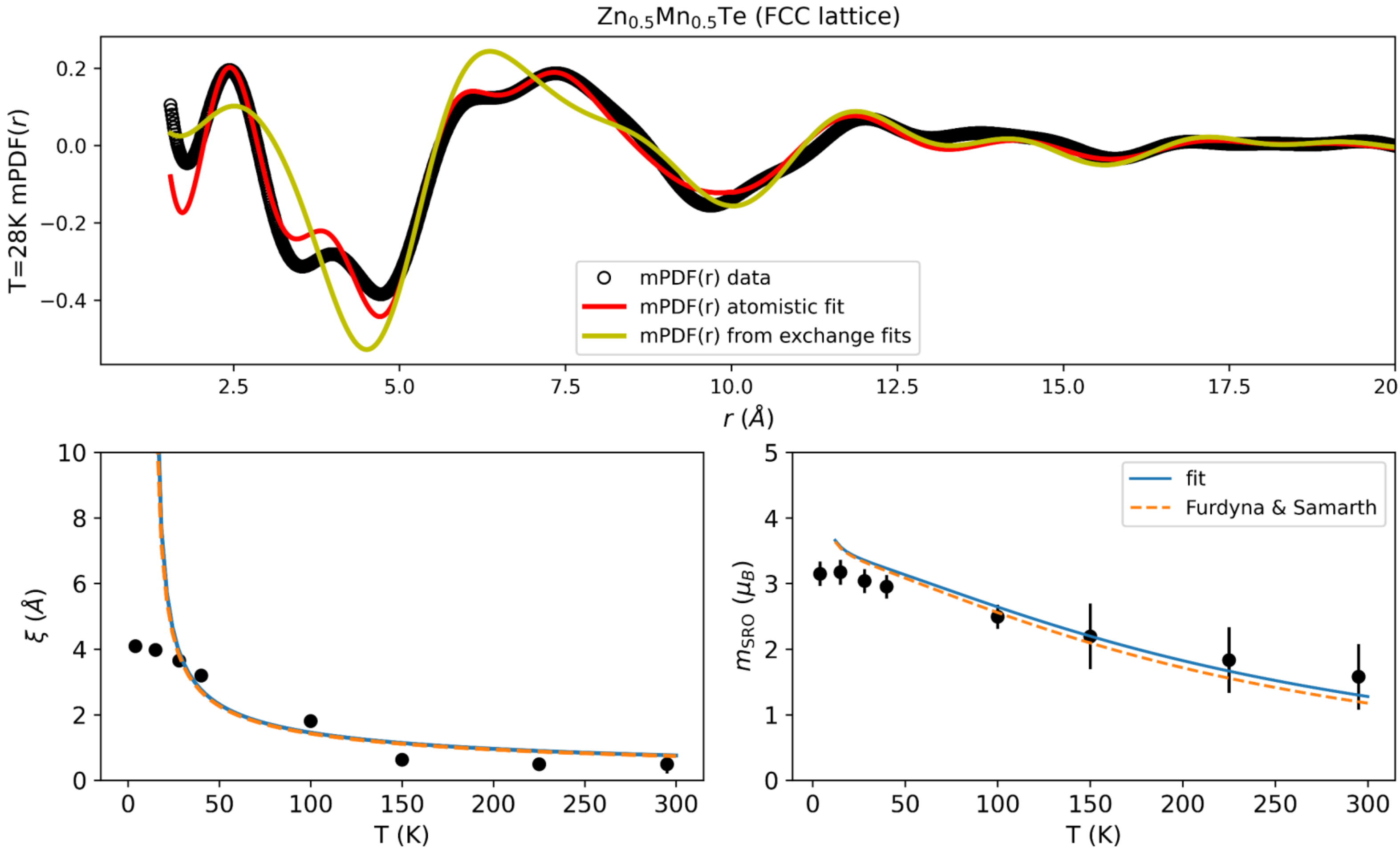


$\boldsymbol{q}_0$ found to be points equivalent to (1,1/2,0) in units of $(2\pi/a)$

- Located on BZ vertices
- Recreates type III AFM order
- 24 vertices, but each point is only fractionally in BZ
  - Only 6 points needed to describe magnetic structure

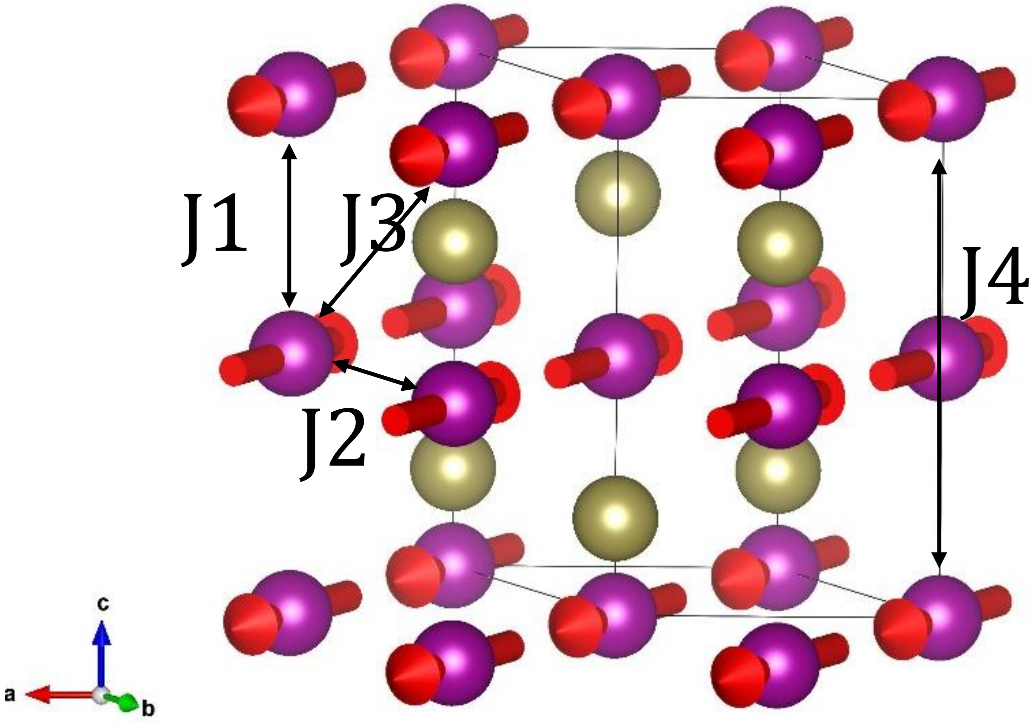
Example 1: $Zn_{0.5}Mn_{0.5}Te$

	J1	J2
Furdyna & Samarth	-9.5 K	-1.9 K
Our fit	-9.8±0.2 K	-1.9±0.1 K



Example 2: MnTe

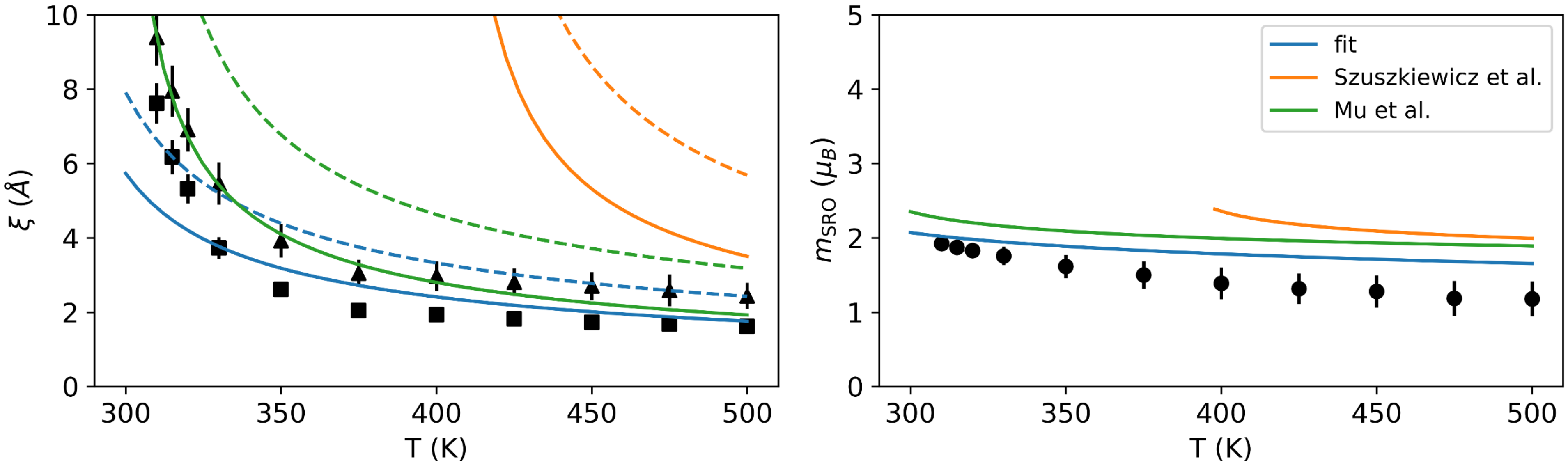
- Altermagnet
  - T_N=307 K
  - Magnetic moments lie within plane
  - FM order in plane, AFM order out of plane
- mPDF analysis done in Baral et al, *Matter* **5**, 1853 – 1864 (2022)
- Exchange interactions are anisotropic
  - J_1 is much stronger than J_2
  - Correlations are stronger out-of-plane than in-plane



Example 2: MnTe

MnTe

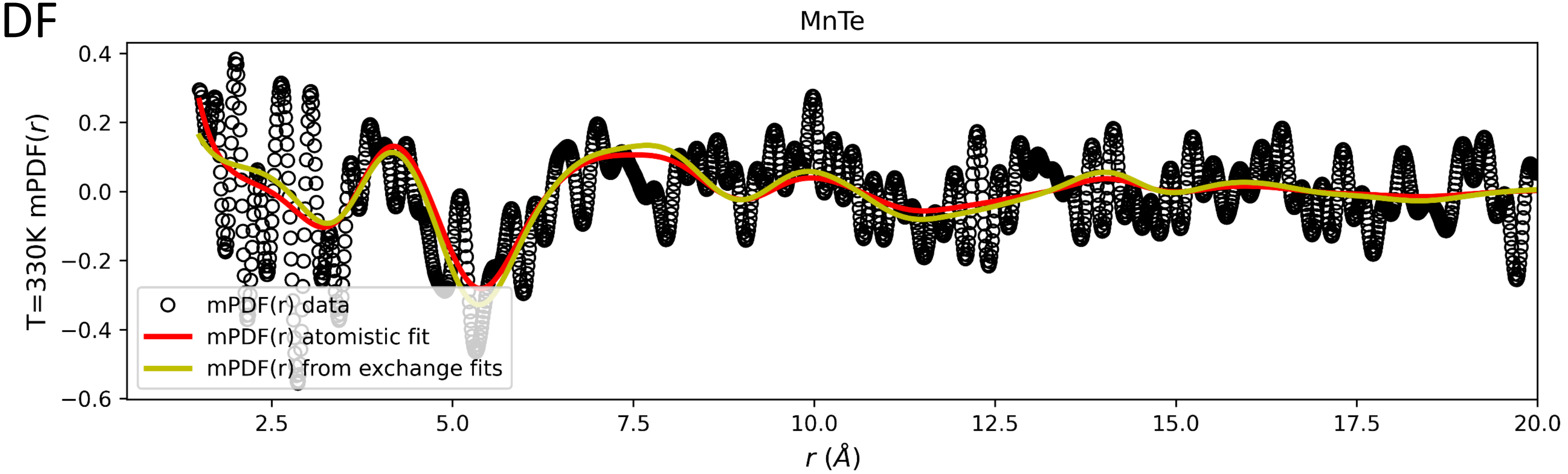
	J1	J2	J3	J4
Mu et al.	-35.6 K	-0.32 K	-4.64 K	-1.86 K
Szuszkiewicz et al.	-43.0 K	1.34 K	-5.74 K	-
Our Fit	-29.6±1.6 K	6.0±1.4 K	-1.2±0.8 K	-



Conclusions and Outlook

- mPDF can tell us more than just the structure!
- Local correlations contain information about local interactions
- Will test method on other materials e.g. 2D frustrated magnets
- More functionality to be developed
  - Anisotropy terms
  - exchange matrices (J_{ij}^{ab})
  - Fit directly to mPDF

Thank you!



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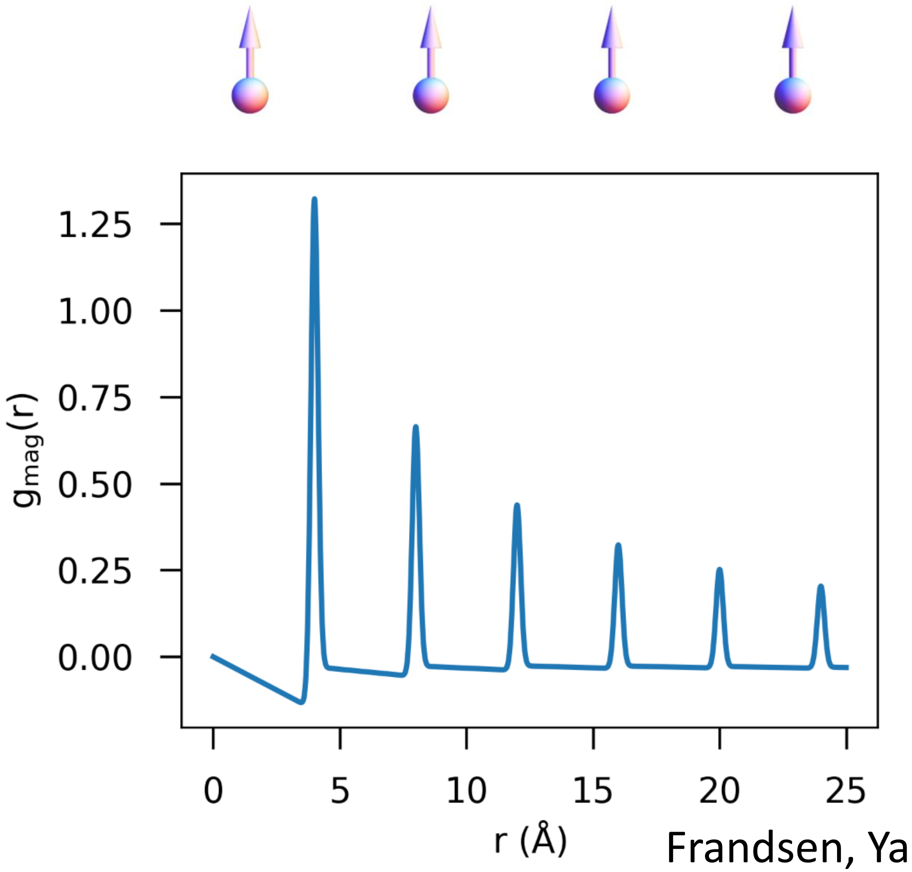
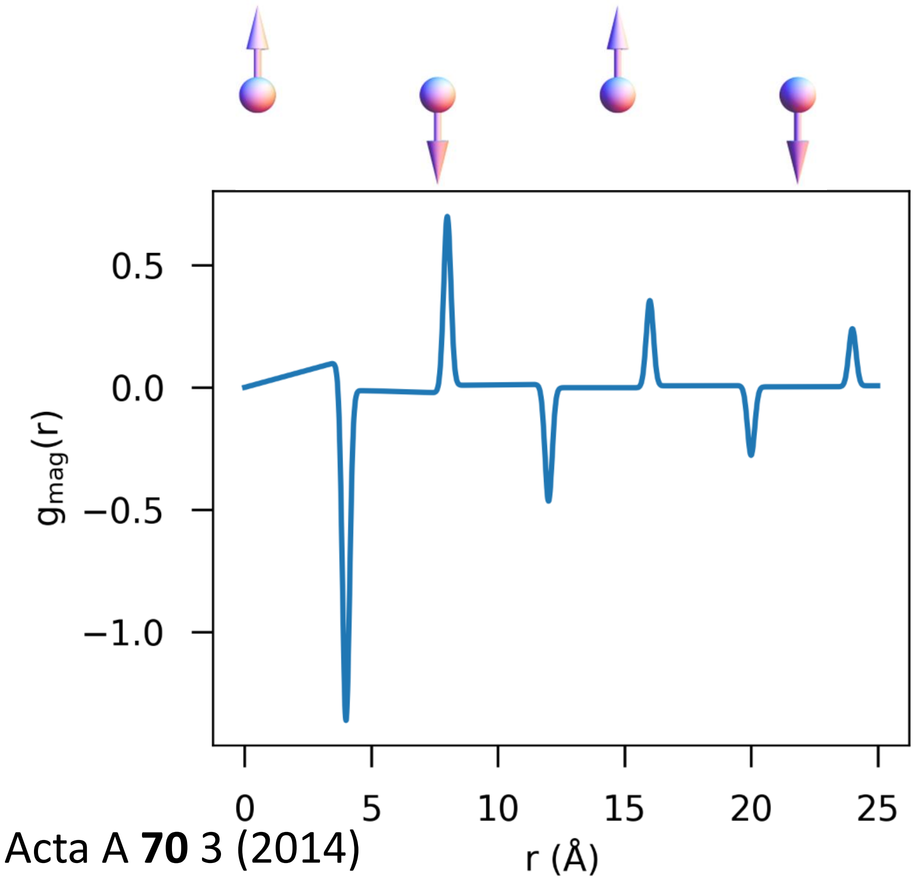
Example: One-dimensional chain

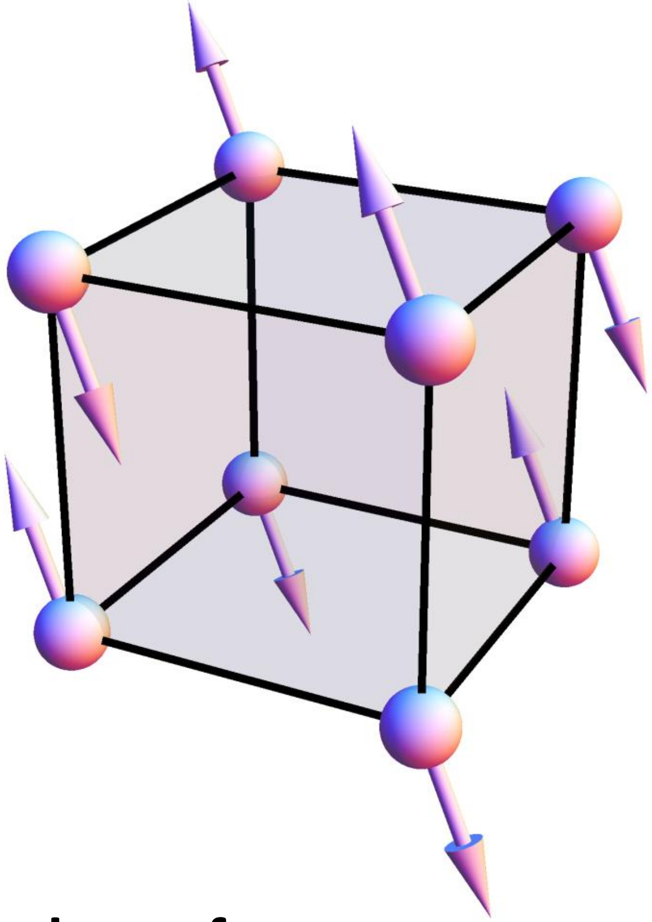
$$G_{\mathrm{mag}}(r) = \frac{3}{2\langle g\sqrt{J(J+1)}\rangle^2}\left(\frac{1}{N_s}\sum_{i\neq j} g_i g_j \left[\frac{A_{ij}}{r}\delta(r-r_{ij}) + B_{ij}\frac{r}{r_{ij}^3}\Theta(r_{ij}-r)\right] - 4\pi r\rho_0\frac{2}{3}m^2\right)$$

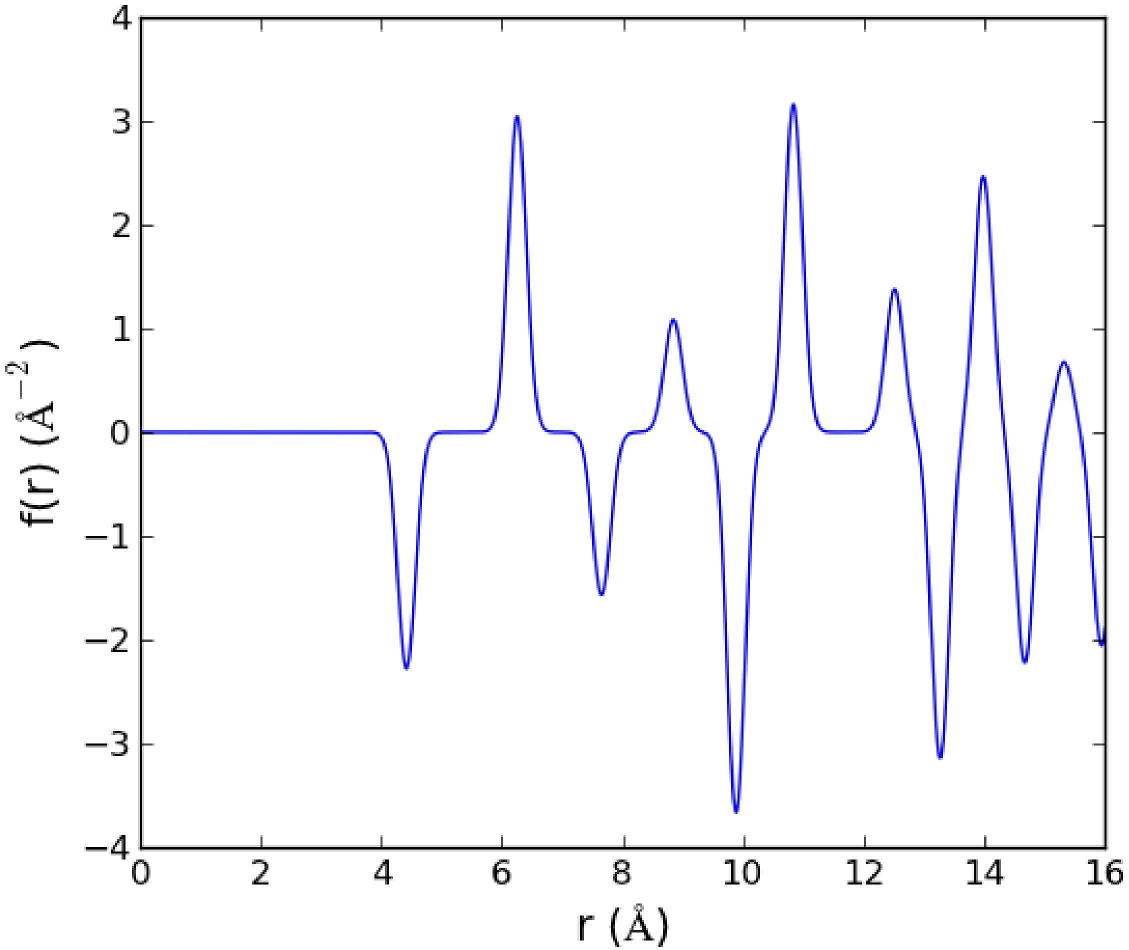




Frandsen, Yang, & Billinge, Acta A **70** 3 (2014)

Example: Simple cubic antiferromagnet





Further references

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- *PRL* **116** 197204 (2016)
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