Substitutional disorder in the $Pr_2Zr_2O_7$ family of spin-liquid candidate materials

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Thomas J Hicken[1,2], P Meadows[2], T Northam[2], DJ Voneshen[3,2], D Prabhakaran[4], LE Chern[5], SE Dutton[5], C Castelnovo[5], JP Goff[2]

PSI Center for Neutron and Muon Sciences

[1]Center for Neutron and Muon Sciences, Paul Scherrer Institute, Villigen PSI, Switzerland
[1]Department of Physics, Royal Holloway, University of London, Egham TW20 0EX, United Kingdom
[2]ISIS Pulsed Neutron and Muon Source, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot, OX11 0QX, United Kingdom
[3]Department of Physics, Clarendon Laboratory, University of Oxford, Parks Road, Oxford, OX1 3PU, United Kingdom
[4]TCM Group, Cavendish Laboratory, University of Cambridge, Cambridge, CB3 0HE, United Kingdom

1. Pyrochlore magnets host exciting magnetism

Pyrochlores have the chemical formula $A_2B_2O_7$. The magnetism is driven by the structure: **corner-sharing tetrahedra** of Ising-like moments.

The interaction between moments is **frustrated**, leading to exotic magnetic states with short-range correlations, but no long-range order. We name these states **quantum spin-liquids**!

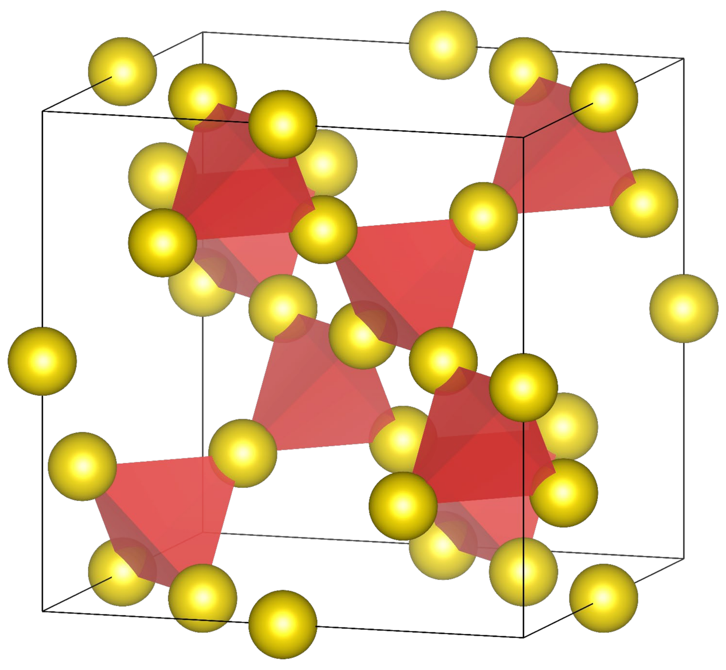

***Fig. 1**: Pyrochlores host magnetic moments that often point either in or out of the corner-sharing tetrahedra*

2. Disorder in $Pr_2Zr_2O_7$ gives rise to quantum fluctuations

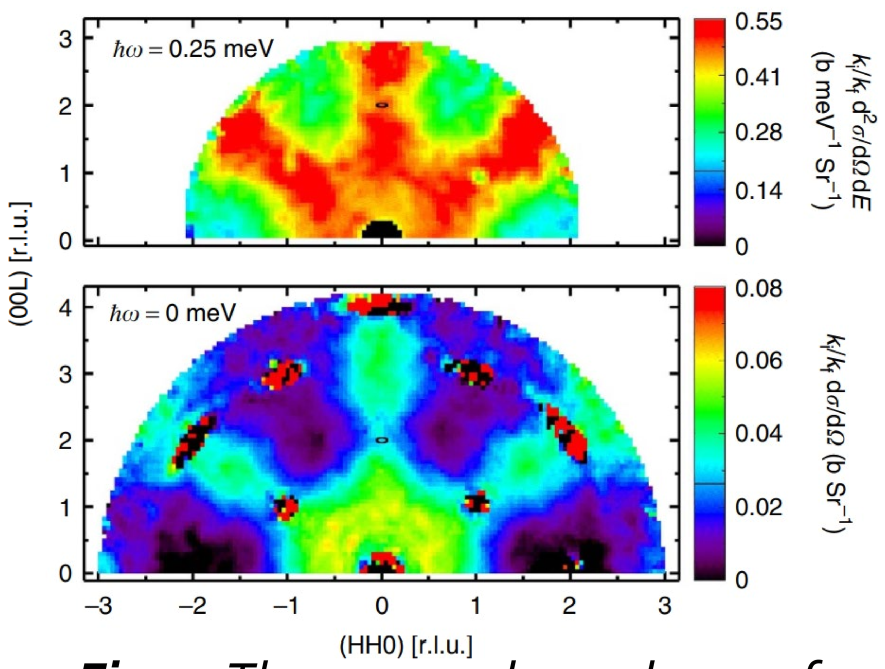

***Fig. 2**: The energy dependence of inelastic neutron scattering shows how quantum fluctuations affect the spin-ice correlations. From Ref. [1].*

$Pr_2Zr_2O_7$ is reported to be a **spin ice**, with 2 spins pointing in and 2 pointing out of each tetrahedra.

Intrinsic disorder is believed to give rise to **quantum fluctuations**, changing the Ising-like magnetic moments.

We want to understand how this works!

3. We introduce more disorder in Pr_2ScNbO_7

To understand the disorder, we add more! By replacing Zr^{4+} with Sc^{3+} and Nb^{5+}, we **introduce strains** without charge imbalance or vacancies.

One might expect disorder to relieve the frustration and lead to long-range magnetic order, but theory suggests that for certain systems it may **stabilise quantum spin liquid states**.

We want to place our systems on this phase diagram. We first have to understand what happens to the structure.

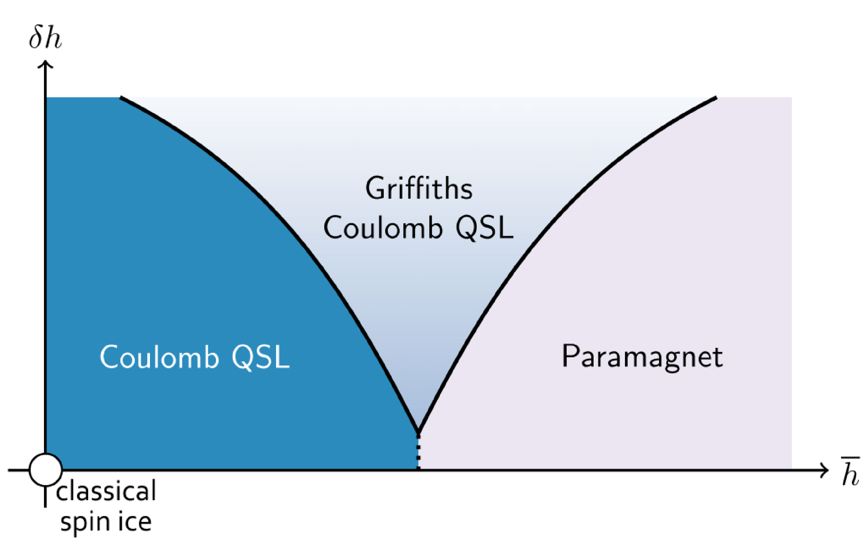

***Fig. 3**: By changing the disorder (h) in pyrochlores with non-Kramers magnetic ions, theory predicts quantum spin-liquid states are stabilised. From Ref. [2].*

4. Density functional theory explains disorder-sensitive diffuse neutron scattering

The structure can be found using density functional theory. We find there are **short-range correlations** for the Sc-Nb locations: they form a **charge ice** and prefer to be in chains of alternating Sc and Nb.

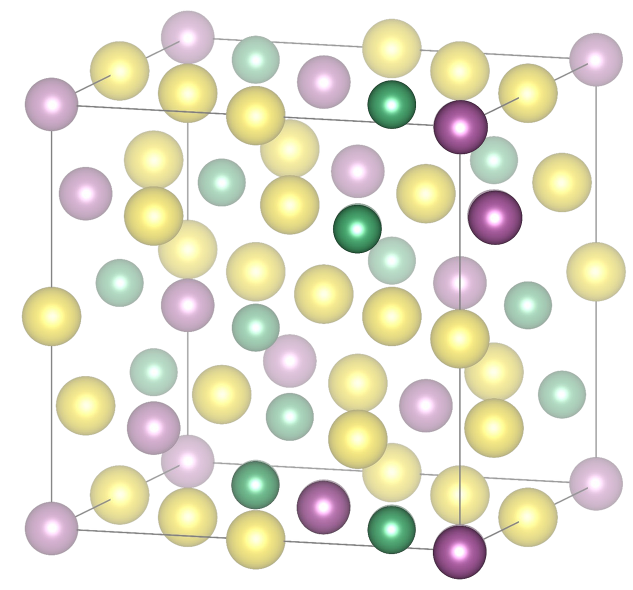

***Fig. 4**: The lowest energy configuration of Pr_2ScNbO_7, a charge ice with Sc-Nb chains*

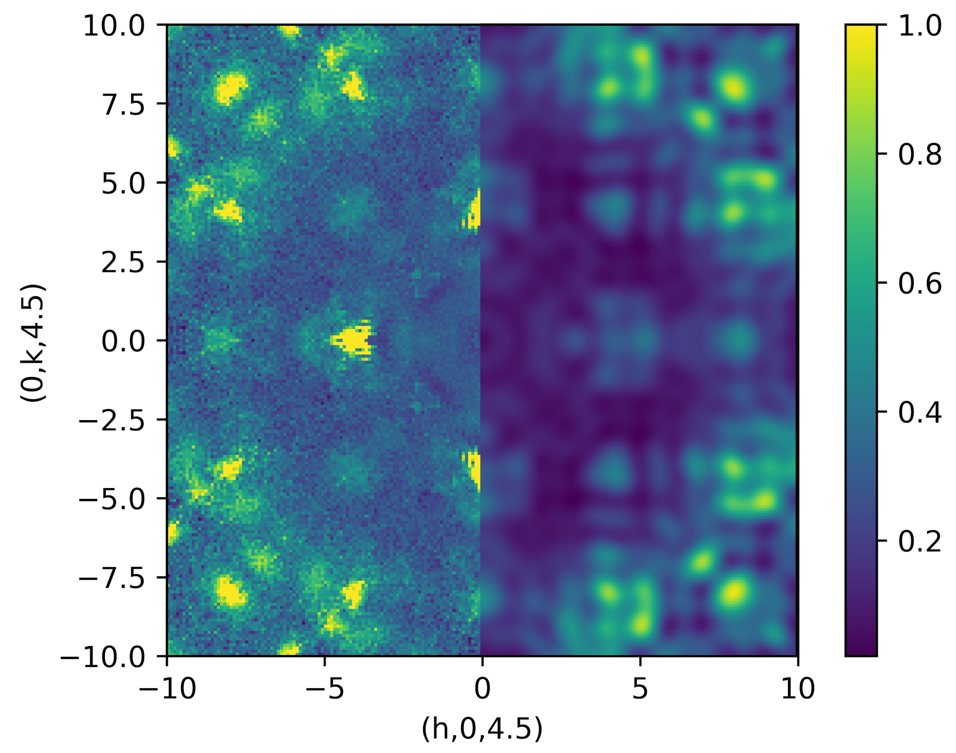

***Fig. 5**: Diffuse neutron scattering, measured experimentally on SXD at ISIS (left) and theoretically predicted using SCARF at STFC (right)*

We can calculate the diffuse neutron scattering from this first-principles prediction. This **matches experimental** measurements very well, demonstrating **we understand the disorder**.

5. We find unexpected changes to the magnetism

We predict that Pr_2ScNbO_7 has **two inequivalent Pr sites**. Only one of these sites is magnetic!

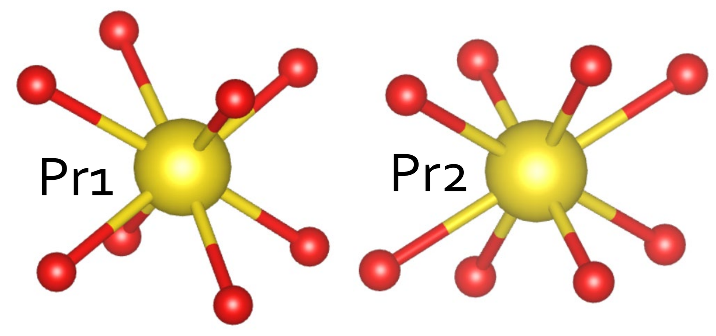

***Fig. 6**: The two inequivalent Pr sites in Pr_2ScNbO_7*

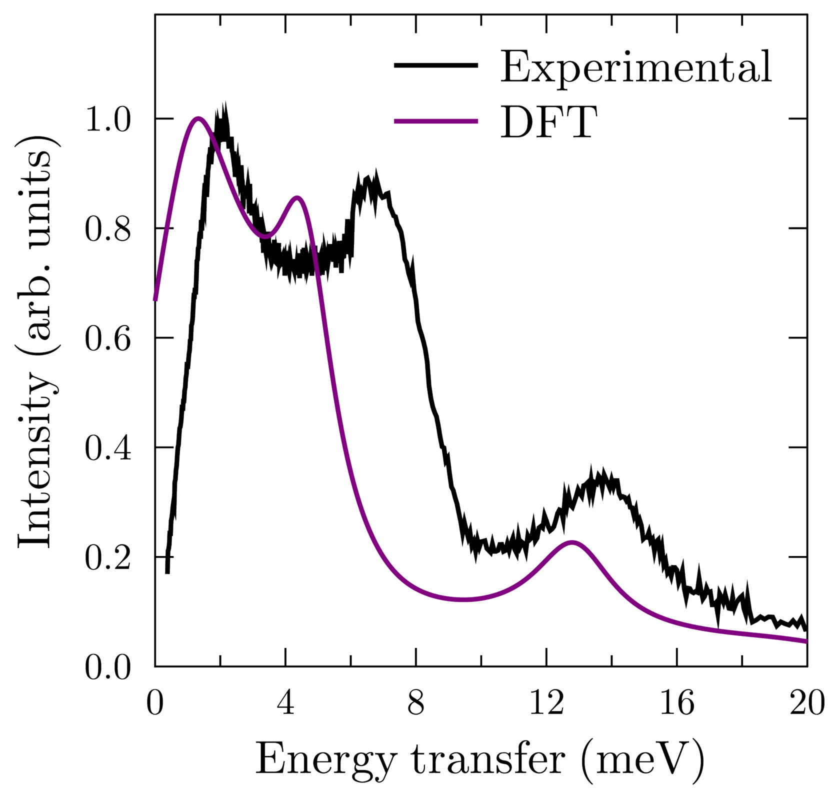

***Fig. 7**: The density functional theory calculation well explains the features in the experimental inelastic neutron scattering from LET at ISIS*

Our model predicts all the experimentally found **inelastic neutron scattering** features. This means that the calculations **explain the single-ion magnetism**.

Now we can keep going to **predict and measure the stabilised magnetic states** in this material and others in the series!

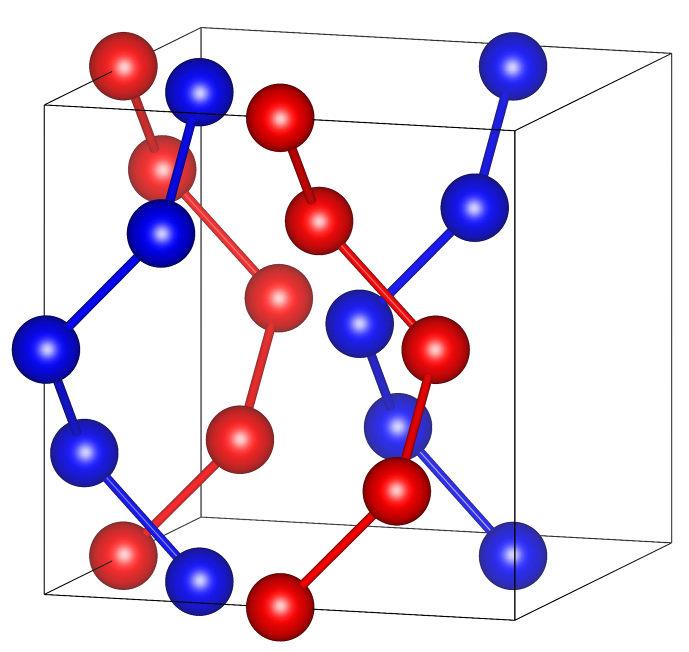

***Fig. 8**: The two Pr sites form a complex pattern in Pr_2ScNbO_7*

[1] K Kimura *et al.*, Nat. Comm. **4**, 1934 (2013)
[2] L Savary *et al.*, Phys. Rev. Lett. **118**, 087203 (2017)

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