RESEARCH STATEMENT

Dr Joseph Paddison
Junior Research Fellow, Churchill College
University of Cambridge
CB3 0DS
United Kingdom

Statement of proposed research for the position of Assistant Professor of Physics at Texas Tech University

Background and context

Cool most materials to low enough temperature, and eventually they become solids. Most magnetic materials behave in an analogous way: at low enough temperature, the magnetic moments (spins) condense into an ordered pattern. My research in condensed-matter science has focused on magnets that defy this expectation. Instead, they remain in a "spin liquid" state to the lowest measurable temperatures. One reason for this unconventional behavior is that the lattice geometry prevents all the magnetic interactions from being satisfied at once (**Fig. 1**). This "frustration" can lead to fundamentally-new states of matter [1], and is implicated in important technologies such as high-temperature superconductors [2] and quantum computers [3].

To understand spin liquids, we need accurate models of their spin arrangements at the atomic scale. Unfortunately, however, standard crystallographic methods cannot be applied to spin liquids, because the absence of magnetic order means no sharp magnetic Bragg scattering is observed in neutron-scattering experiments: instead, only broad ("diffuse") scattering is observed. Consequently, solving the spin arrangements in spin liquids is a challenging problem, which has implications for our ability to model disordered systems ranging from amorphous materials to neural networks [4]. My research focuses on using neutron-scattering experiments to understand spin liquid states at the atomic scale. One of my key contributions to this field has been to show that the spin arrangements of spin liquids can be solved using only data from powder samples [5]. To prove this result, I used the atomistic modeling technique of "reverse Monte Carlo" (RMC) refinement [6] to fit spin structures to powder neutron-diffraction data for models of several spin liquids. I then showed that the structures refined to the powder data fully reproduced the three-dimensional single-crystal diffraction patterns of the starting models (**Fig. 1**) [5]. This approach dramatically increased the level of information which can be reliably extracted from neutron-diffraction data for spin liquids. I have since used it to reveal exotic magnetic states in many materials (e.g., [7–9]).

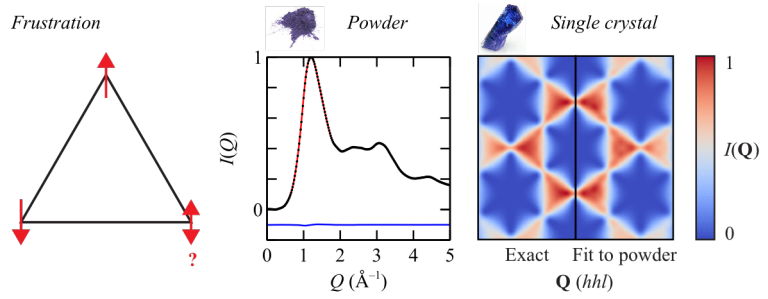


Fig. 1: *Left*: Example of frustration: three vectors on a triangle cannot be mutually opposite. *Right*: I showed that fitting to one-dimensional (powder) neutron-scattering data for a frustrated system allows the three-dimensional (single-crystal) correlations to be reconstructed.

I seek to build on these advances to address three important questions in condensed-matter physics. What is the relationship between structural and magnetic disorder in spin liquids? What are the local magnetic structures of high-temperature superconductors? And how can we understand magnetic interactions in metallic spin liquids?

Research objectives and methodology

My research is interdisciplinary, lying at the interface of condensed-matter physics, crystallography, and materials science. My proposal includes three separate but interlinked themes, involving components of both experiment and modeling. I outline below the motivation, methodology, and measurable objectives for each theme. I also present two case studies of my previous work, to demonstrate the value of the methodology and the expected impact of the results. Some polycrystalline samples will be prepared in-house. I also have existing collaborations with synthetic chemists in the USA (Prof Haidong Zhou, University of Tennessee) and the UK (Dr Siân Dutton, University of Cambridge), and will develop further collaborations as leader of my research group.

1. Interplay of Structural and Magnetic Disorder in Spin Liquids

The conventional route to realizing spin liquids (SLs) in real materials combines two ingredients: antiferromagnetic interactions, and a frustrated lattice built from equilateral triangles. In this field, chemical disorder has been considered an undesirable extraneous effect that suppresses SL behavior. However, recent work has challenged this paradigm, and there is now evidence that chemical disorder may actually play a key role in the magnetic behavior of several prominent SL candidates such as $Pr_2Zr_2O_7$ [10] and $YbMgGaO_4$ [11,12].

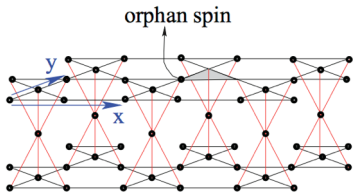


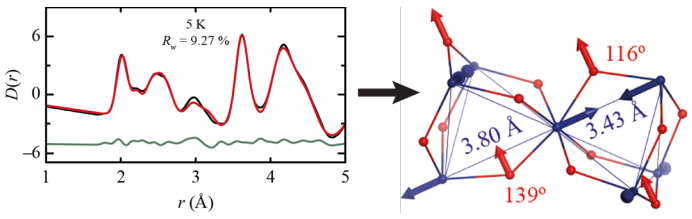
Fig. 2. Magnetic lattice of $SrCr_{9p}Ga_{12-9p}O_{19}$, showing chemical disorder [reproduced from Ref. 14]. I will determine how the chemical disorder is correlated and how this affects the magnetism.

This project aims to address two key questions. First, what is the nature of chemical disorder in candidate SLs? And, second, how does this disorder affect the magnetism? My strategy is to start with the simplest model systems to identify underlying principles. An ideal model system is the canonical frustrated magnet $SrCr_{9p}Ga_{12-9p}O_{19}$, which contains magnetic bilayers of Cr^{3+} ions that form a network of corner-sharing triangles [13]. These layers are diluted by a fraction $x = 1-p$ of nonmagnetic Ga^{3+} ions, introducing a controllable amount of structural disorder (**Fig. 2**). Theoretically, this dilution is proposed to lead to exotic magnetic interactions involving spatially-extended spin textures surrounding "orphan" spins [14]. What is not yet known is how orphan spins are arranged – e.g., if they are entirely random or correlated – and how correlated chemical disorder controls the magnetism.

My study would use a combination of neutron and X-ray diffuse scattering measurements and atomistic modelling to reveal the local structure of $SrCr_{9p}Ga_{12-9p}O_{19}$. I have a track record in gaining access to large-scale facilities through competitive beam-time allocation. Neutron-scattering data collected on powder samples would be analyzed using the pair-distribution function (PDF) approach, in which I am expert (see **case study 1**). X-ray scattering data collected on single-crystal samples would be analyzed using RMC code I have developed for "big data" analysis, which fits atomistic models to $>10^6$ data points [9]. This computational approach enables the structural disorder to be visualized in real space, and – crucially – allows both magnetic and structural disorder to be simultaneously fitted, enabling their coupling between to be determined. I would apply the lessons learned from this study to study the interplay of structural and magnetic disorder in the "relaxor ferromagnet" $La_3Ni_2SbO_9$ [15] and structural analogs of the *quantum* SL candidate $YbMgGaO_4$ [11,12], in collaboration with the laboratory of Prof Martin Mourigal (Georgia Tech, USA). This project would be suitable for a PhD student, providing interdisciplinary training in sample preparation, scattering experiments, PDF analysis, and Monte Carlo modeling.

Case study 1: Orbital Disorder Drives Spin-Glass Formation in $Y_2Mo_2O_7$

The origin of spin-glass behavior in $Y_2Mo_2O_7$ was a longstanding question, because the nature of its structural disorder – considered prerequisite for spin freezing – was not understood. Using fits to PDF data (left), we identified the key local displacements (right) of Mo^{4+} (blue) and O^{2-} (red) as the formation of Mo dimers driven by frustrated orbital interactions of Jahn-Teller active Mo^{4+}. These results demonstrate the importance of local structure for magnetic materials. [Figures reproduced from Thygesen, Paddison *et al.*, *PRL* **118**, 067201 (2017).]



Objectives, milestones, and timeline

- Year 1: Measure X-ray and neutron scattering data of spin-liquid $SrCr_{9p}Ga_{12-9p}O_{19}$
- Year 2: Analyse these data using PDF and Monte Carlo modeling to characterize structural disorder
- Year 3: Identify the interplay between structural and magnetic degrees of freedom
- Years 3-5: Extend to other materials, including $La_3Ni_2SbO_9$ and $YbMgGaO_4$ analogs.

2. Energy-Resolved Local Magnetic Structures of High-Temperature Superconductors

Arguably the most important materials in which magnetic disorder plays a key role are the high-temperature superconductors (SCs). Parent compounds of high-T_c SCs are antiferromagnetic insulators, which become superconducting when doped with small amounts of other elements. While conventional antiferromagnetic order is rapidly suppressed by doping, disordered antiferromagnetic fluctuations remain in the superconducting phase, and it is widely considered that these spin fluctuations are intimately linked to superconductivity [2,16].

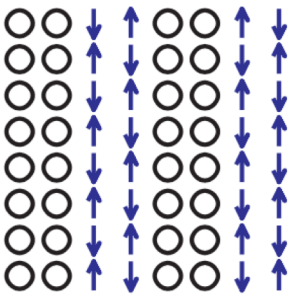


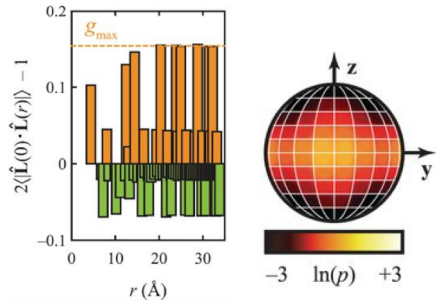
Fig. 3. Pictorial model of fluctuating spin stripes in high-T_c superconductors [reproduced from Tranquada *et al., Nature* **429**, 534 (2004)]. I will determine a data-driven model of the local magnetic structure of these important systems.

Just as for spin liquids, an important technique for studying spin fluctuations in high-T_c SCs is neutron scattering. Again, however, the lack of magnetic order has made data analysis challenging. Experimental studies have often considered magnetic disorder at a relatively simple level; e.g., by assuming that excitations in a "fluctuating spin-stripe" state resemble those for a stripe-ordered phase, modified by a phenomenological peak-broadening function (**Fig. 3**) [16]. Such assumptions are problematic for two reasons: (i) the ordered state must be assumed in advance, which may bias the model; and (ii) the study of frustrated magnets has shown that disordered states can behave in a fundamentally different way to ordered states (see, e.g., [17]).

I propose a new approach to the problem of local magnetic structure of high-T_c SCs, based on inelastic measurements on powder samples coupled with the RMC refinement approach that I have successfully applied to SLs. I have three reasons to reinvestigate these well-studied materials. First, large powder samples allow measurements to be performed rapidly over a range of composition and temperature, allowing key trends to be derived. Second, my approach is quantitative and driven entirely by experimental data: neither the magnetic structure nor the underlying interactions are assumed in advance. And most importantly, in spin liquids – intimately related to high-T_c SCs [18] – I have shown that the direct sensitivity of powder diffuse-scattering data to spin-pair correlations and local magnetic anisotropies can allow sensitivity to non-dipolar order parameters, including the nematic order potentially relevant for high-T_c SCs (see **case study 2**).

To maximize the benefits of this new approach, I will focus on iron pnictide systems such as $LaFeAsO_{1-x}F_x$ that have relatively strong magnetic scattering but cannot be prepared as large crystals. My analysis will enable the spin structures corresponding to correlations of specific energy to be isolated in real space. My ultimate aim is therefore to produce the first atomic-scale model of energy-resolved superconducting spin fluctuations which is entirely data-driven. This approach is previously unexplored in this field: it will not only allow existing theories to be tested, but also has strong potential to reveal new states beyond those currently predicted.

Case study 2: Hidden Order in a Spin Liquid



I discovered that the spin liquid $Gd_3Ga_5O_{12}$ hosts a hidden-order state in which groups of spins show nematic ordering (*left*), while individual spins remain in a fluctuating spin-liquid state. Sensitivity to this state occurs because diffuse scattering measures both spin correlations and spin anisotropy, as shown by the distribution of spin orientations (*right*). These results suggest that my RMC technique is a promising approach to uncover hidden order parameters in systems such as high-T_c superconductors. [Figures reproduced from Paddison *et al., Science* **350**, 179 (2015).]

Objectives, milestones, and timeline

- Year 1: Measure temperature-dependent inelastic neutron scattering data of polycrystalline $LaFeAsO_{1-x}F_x$
- Years 2-3: Develop energy-resolved RMC software and identify energy-resolved spin correlations
- Years 3-5: Apply this approach to other high-T_c systems, such as FeSe.

3. Magnetic Interactions in Unconventional Metals from Neutron Scattering

Magnetic interactions in insulating materials are generally well understood. At the simplest level, for example, the net interaction strength can be determined by fits to the bulk magnetic susceptibility in the paramagnetic regime using the Curie-Weiss law. By comparison, magnetic metals are relatively poorly understood; e.g., mean-field theory predicts that local magnetic moments in metals vanish above their magnetic ordering temperature, in disagreement with experimental evidence even in Fe and Ni [19]. The current state of the art in determining magnetic interactions in metals involves estimating the energy "landscape" by first-principles calculations of different candidate magnetic ground states. The energies of these states are parameterized by an effective spin Hamiltonian containing a Heisenberg-like interaction term that induces correlations between transverse spin components, and a single-site term that allows longitudinal spin fluctuations [20]. This approach has been successful for metals with ordered ferromagnetic ground states, such as Fe and Ni. However, it is challenging to apply to antiferromagnetic metals such as Cr, and metallic spin liquids such as ß-Mn [7] and Sc-doped YMn_2 [21].

Inelastic neutron scattering data contain rich information about magnetic interactions, irrespective of whether magnetic moments are itinerant or localized. In this study, I will develop an experiment-driven approach to determine magnetic interactions in unconventional metallic magnets directly from inelastic neutron-scattering data. My algorithm consists of three stages. First, the total magnetic moment is determined from the total magnetic spectral weight at each temperature, allowing the on-site term to be parameterized. Second, the magnetic correlations are determined using RMC refinement. Third, the magnetic interactions are derived from the first moment of the scattering, which is proportional to the integral of the scattering intensity multiplied by energy transfer. While each step has been shown to be practical, their combination has not been applied to understand magnetic metals.

I will initially study two systems in this project. First, I will perform temperature-dependent measurements of Ni metal in its magnetically-ordered and paramagnetic regimes; this will benchmark my approach since its magnetic interactions have been well-studied by traditional approaches. Second, I will study the canonical metallic spin liquid $Y_{1-x}Sc_xMn_2$, a Laves phase heavy-fermion compound that remains a spin liquid at all measurable temperatures [21]. These results will provide insight into the interaction between itinerant electrons and geometrical frustration. This project would be suitable for a PhD student with interests in computational physics and data analysis.

Objectives, milestones, and timeline

- Year 1: Measure temperature-dependent inelastic neutron scattering data of Ni, and Sc-doped YMn_2
- Years 2-3: Develop and apply analysis software to extract magnetic interactions
- Years 3-5: Identify general implications of these results for frustrated metals.

[1] L. Balents, Nature **464**, 199 (2010).
[2] P. W. Anderson, Science **235**, 1196 (1987).
[3] Broholm, Collin, *Quantum Materials for Energy Relevant Technology* (US Department of Energy, 2016).
[4] E. Schneidman, M. J. Berry II, R. Segev, and W. Bialek, Nature **440**, 1007 (2006).
[5] J. A. M. Paddison and A. L. Goodwin, Phys Rev Lett **108**, 017204 (2012).
[6] R. L. McGreevy and L. Pusztai, Mol Simul **1**, 359 (1988).
[7] J. A. M. Paddison, J. R. Stewart, P. Manuel, P. Courtois, G. J. McIntyre, B. D. Rainford, and A. L. Goodwin, Phys Rev Lett **110**, 267207 (2013).
[8] J. A. M. Paddison, H. S. Ong, J. O. Hamp, P. Mukherjee, X. Bai, M. G. Tucker, N. P. Butch, C. Castelnovo, M. Mourigal, and S. E. Dutton, Nat. Commun. **7**, 13842 EP (2016).
[9] J. A. M. Paddison, M. J. Gutmann, J. R. Stewart, M. G. Tucker, M. T. Dove, D. A. Keen, and A. L. Goodwin, Phys Rev B **97**, 014429 (2018).
[10] O. Benton, Phys Rev Lett **121**, 037203 (2018).
[11] J. A. M. Paddison, M. Daum, Z. Dun, G. Ehlers, Y. Liu, M. B. Stone, H. Zhou, and M. Mourigal, Nat. Phys. **13**, 117 EP (2016).
[12] I. Kimchi, A. Nahum, and T. Senthil, Phys Rev X **8**, 031028 (2018).
[13] C. Broholm, G. Aeppli, G. P. Espinosa, and A. S. Cooper, Phys Rev Lett **65**, 3173 (1990).
[14] A. Sen, K. Damle, and R. Moessner, Phys Rev Lett **106**, 127203 (2011).
[15] P. D. Battle, S. I. Evers, E. C. Hunter, and M. Westwood, Inorg Chem **52**, 6648 (2013).
[16] J. M. Tranquada, H. Woo, T. G. Perring, H. Goka, G. D. Gu, G. Xu, M. Fujita, and K. Yamada, Nature **429**, 534 (2004).
[17] C. Castelnovo, R. Moessner, and S. L. Sondhi, Nature **451**, 42 (2008).
[18] I. Zaliznyak, A. T. Savici, M. Lumsden, A. Tsvelik, R. Hu, and C. Petrovic, Proc. Natl. Acad. Sci. **112**, 10316 (2015).
[19] P. J. Brown, H. Capellmann, J. Déportes, D. Givord, and K. R. A. Ziebeck, J Mag Magn Mater **30**, 243 (1982).
[20] M. Uhl and J. Kübler, Phys Rev Lett **77**, 334 (1996).
[21] R. Ballou, E. Lelièvre-Berna, and B. Fak, Phys Rev Lett **76**, 2125 (1996).
[22] M. Mourigal, M. Enderle, A. Klöpperpieper, J.-S. Caux, A. Stunault, and H. M. Rønnow, Nat. Phys. **9**, 435 (2013).