[MS35- 03] Magnetic Structure Solution of Frustrated Spin Systems

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Frustrated spin systems can exhibit a macroscopic degeneracy of magnetic ground states which suppresses periodic magnetic order [1]. Topical examples include spin ices such as $Ho_2Ti_2O_7$ [2, 3] and quantum spin liquid candidates such as herbertsmithite [4]. While the absolute arrangement of spins differs amongst degenerate states, the states share local spin correlations that distinguish their magnetic structures from classical paramagnets. The importance of understanding these correlations lies primarily in determining the origin of exotic phenomena that emerge from frustrated spin systems, e.g. the evolution of high-temperature superconductivity from spin liquids [5] and the ability of spin ices to support emergent magnetic charges [6].

Spin orientations within ordered magnets can usually be determined using a combination of neutron diffraction measurements and crystallographic analysis of the magnetic Bragg peaks. However, the suppression of periodic magnetic order in frustrated magnets means that no magnetic Bragg peaks are observed. Instead, the magnetic neutron diffraction pattern is a smoothly varying function of three-dimensional reciprocal space. Traditionally, the magnetic correlations of frustrated systems have been studied by calculating the neutron diffraction pattern anticipated from predetermined interaction models and comparing with experimental single-crystal neutron diffraction data (see *e.g.* [7]). While this approach has been unquestionably successful, it has two important limitations: (i) the interactions responsible for local magnetic ordering must be anticipated, and (ii) large single-crystal samples must be available. In my presentation, I will explore the information content of the one-dimensional magnetic powder diffraction pattern $I(Q)$ of frustrated magnets. In particular, I will address how the function $I(Q)$ can be converted robustly into a structural model without any prior knowledge of the underlying magnetic interactions. Our approach is to consider simulated $I(Q)$ data for a number of test cases—frustrated systems whose magnetic structures are relatively well understood. These data are then fitted using the atomistic reverse Monte Carlo (RMC) method [8, 9]. Finally, the quality of the models obtained is assessed by calculating the full single-crystal scattering function I(Q). I will show that the extent of information loss during spherical averaging of the single-crystal magnetic diffraction pattern is surprisingly minimal, and that the full 3D diffraction pattern is recoverable from powder diffraction data for each frustrated system that we explore [10].

I will go on to discuss two real examples where we have used this RMC approach to obtain insight into frustrated materials. First, I will consider the evolution with temperature of ice-rules defects in the newly-synthesized spin ice material, $Ho_2Ge_2O_7$ [11], and show how the powder diffuse scattering pattern contains information on subtle magnetic correlations, which vary between different spin ice materials. Second, I will examine the interplay between geometrical frustration and low dimensionality in the paramagnetic phase of the spin-chain compound $Ca_3Co_2O_6$ [12, 13]. Finally, I will discuss how the RMC data analysis methods—here applied to powder data—can be extended for the analysis of single-crystal data [14].

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