Neutron Scattering Study of Frustration and Magnetic Order in Spinels

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The ‘A-site spinels’ are materials with magnetic cations constrained to lie on the diamond sublattice of the spinel structure. These systems have been of increasing interest, as novel theoretical and experimental results have emphasized the central role of frustration arising from competing interactions. In particular, the sub-family of diamond lattice antiferromagnets with spin-only degrees-of-freedom is predicted to exhibit novel ‘spiral-spin-liquid’ and order-by-disorder physics (Bergman et al., Nature Physics (2007)). Real systems MnSc2S4 (Krimmel et al., PRB (2006)) and CoAl2O4 (Krimmel et al., Physica B (2006)) have been studied with powder neutron diffraction, and the results are argued to be consistent with this theory. However, as has been expressed on several occasions, studies on single-crystals are needed for a definitive answer. Here I will discuss neutron scattering results on single-crystalline CoAl2O4, which have given a much more complete picture of magnetic correlations in this material. Both elastic and inelastic measurements have been made using triple-axis and cold chopper spectrometry. With decreasing temperature, we observe intense diffuse scattering centred about locations in reciprocal space associated with collinear antiferromagnetism. At T* 6.5K, we further observe an unexpected change in the diffuse scattering lineshape, coupled with the emergence of well-defined spin-wave excitations. This temperature has been associated with an anomalous spin glass transition in the past, but we argue instead that the available data implies a first-order phase transition to an ordered state, possibly via the order-by-disorder mechanism. The ground state is degenerate, and kinetically frozen walls separating different domains give rise to the broadened scattering at magnetic wavevectors. This scenario may be present in many other frustrated systems. If time permits, I will also talk about new results on crystals of related systems MnAl2O4 and FeAl2O4.

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