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From Spin Glass to Spin Liquid Ground States in Pyrochlore Molybdates

LUCY CLARK, University of St Andrews

Magnetic pyrochlores continue to generate intense interest due to the wealth of interesting behaviours that they can display as a result of their highly frustrated nature. Here we will present our study of the molybdate pyrochlore $Lu_2Mo_2O_7$, which contains non-magnetic Lu^{3+} and an antiferromagnetic network of corner-sharing tetrahedra of Mo^{4+} $4d^2$ $S = 1$ ions [1]. Magnetic susceptibility data show that $Lu_2Mo_2O_7$ enters an unconventional spin glass state at $T_f$$\sim$16 K that displays a quadratic dependence of the low temperature magnetic heat capacity, akin to that observed for its well-studied sister compound $Y_2Mo_2O_7$ [2]. This spin glass transition is also clearly marked in our inelastic (CNCS, SNS) and diffuse elastic magnetic (D7, ILL) neutron scattering data. Furthermore, we will show that it is possible to topochemically substitute the oxide, O^{2-}, ions within $Lu_2Mo_2O_7$ for nitride, N^{3-}, to produce an oxynitride molybdate pyrochlore of composition $Lu_2Mo_2O_5N_2$. Magnetic susceptibility measurements confirm that strong antiferromagnetic correlations persist within the oxynitride, which contains Mo^{5+} $4d^1$ $S = \frac{1}{2}$ ions and is thus a prime candidate to host exotic quantum spin liquid behavior. We will discuss how the enhanced quantum spin fluctuations in $Lu_2Mo_2O_5N_2$ appear to suppress the spin freezing transition observed in its parent oxide and instead support the formation of a gapless spin liquid phase that displays a linear dependence of the low temperature magnetic heat capacity [3]. [1] L. Clark *et al.*, J. Solid State Chem. **203**, 199 (2013), [2] H. J. Silverstein *et al.*, Phys. Rev. B **89**, 054433 (2014), [3] L. Clark *et al.*, Phys. Rev. Lett. **113**, 117201 (2014).