Geometric frustration on the trillium lattice in a magnetic metal–organic framework

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Geometrically-frustrated magnets are of fundamental interest because their macroscopic ground state degeneracies give rise to a number of exotic effects [1]. Despite the large number of possible nets satisfying this local constraint, much of the field has focused on a few structure types common amongst ceramic materials *e.g.,* pyrochlore, kagome *etc*. The study of frustration on more exotic lattices may therefore allow for the discovery of novel magnetic phases and their corresponding physics [2].

In the dense metal-organic framework $Na[Mn(HCOO)_3]$, Mn^{2+} ions (S = 5/2) occupy the nodes of a 'trillium' net. We show that this material exhibits a variety of behaviour characteristic of geometric frustration: the Néel transition is suppressed well below the characteristic magnetic interaction strength [Figure 1c]; neutron scattering indicates that short-range magnetic order persists far above the Néel temperature [Figure 1d]; and the magnetic susceptibility exhibits a pseudo-plateau at 1/3-saturation magnetisation. We demonstrate that a simple nearest-neighbour Heisenberg antiferromagnet model accounts quantitatively for each observation, and hence $Na[Mn(HCOO)_3]$ is the first experimental realisation of this model on the trillium net. We demonstrate how both link geometric frustration within the classical spin liquid regime to a strong magnetocaloric response at low fields.

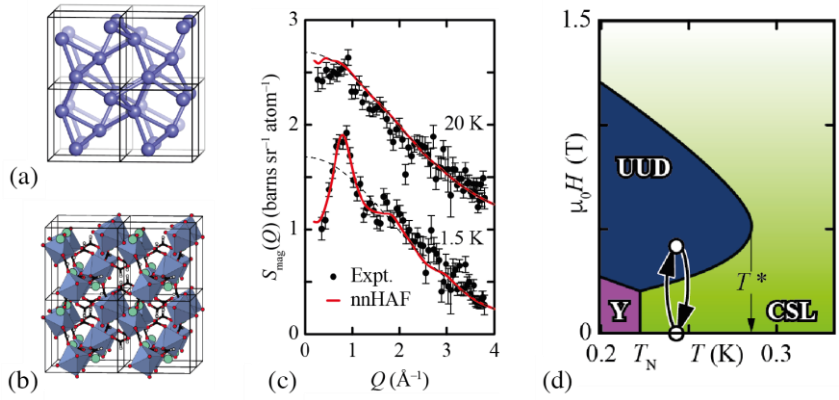

Figure 1. (a) The trillium lattice, shown here as a 2x2x2 supercell of the corresponding cubic unit-cell [3]. (b) Representation of the crystal structure of Na[Mn(HCOO)3]; Na, C, H and O atoms shown as green, black, white and red spheres, respectively, and Mn coordination environments shown as filled polyhedra [4]. (c) The magnetic diffuse neutron scattering functions for the same sample, measured and simulated at 20 and 1.5 K; The 20K values have been shifted vertically by 1 unit for clarity. (d) Schematic phase diagram, showing the low-field / low-temperature ('Y') phase stabilised for $T < T_N$, and the UUD and CSL phases that persist to higher temperatures. Within the regime $T_N < T < T$, it is possible to cycle between disordered and ordered states under the application and removal of a low magnetic field (white circles).

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