Defect-induced short-range magnetic order in $Co_2(OD)_3Cl$

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In recent years we found a new geometric frustration system $Cu_2(OH)_3Cl$ and $Co_2(OH)_3Cl$ with a 3-dimensional network of corner-sharing tetrahedrons of the Cu^{2+} and Co^{2+} spins, respectively 1-3. We found coexisting antiferromagnetic order and spin fluctuation down to T=0 in ideal quantum spin material $Cu_2(OH)_3Cl$ 2, and a kagome-ice-like partial ferromagnetic order in $Co_2(OH)_3Cl$ 3, which raise great interest on this new material system. The latter compound can be compared with the rare-earth pyrochlore compounds $Ho_2Ti_2O_7$, $Dy_2Ti_2O_7$ and $Ho_2Sn_2O_7$, i.e., the " spin ice " compounds. In these materials the spin alignment and the macroscopic quantum degeneracy are shown to be a magnetic equivalence of the ice rule in water ice. The spin ice has received intense attention because of the dynamics associated with the macroscopic degeneracy are experimentally accessible by many magnetic probes, and because the geometrical frustration of competing interactions is an essential element of important problems in fields extending to protein folding and neural networks. The experimentally observed degeneracy and zero-point residual entropy for water ice and spin ice apparently contradict a fundamental principle of thermodynamics (the 3rd law), which demands a single ordered ground state and hence zero entropy. As to the spin ice, a first-order phase transition to a true ground state is theoretically predicted, but has not been observed to date, despite rigorous efforts down to the experimentally attainable temperature of 10 mK. Whether the spin ice keeps the macroscopic quantum degeneracy down to 0 K remains an open and controversial issue.

Because $Co_2(OH)_3Cl$ has a comparatively very higher transition temperature of T_C = 10.5 K, we explored the possibility of a low-temperature order with impurity doping. For this purpose neutron diffraction experiment was carried out at T1-3 (Hermes) using powder sample of $(Co_{0.9}Al_{0.1})_2(OD)_3Cl$.

Figure 1 shows the neutron diffraction patterns for $(Co_{0.9}Al_{0.1})_2(OD)_3Cl$ at typical temperatures. The results clearly show that a short-range spin correlation develops below around 4 K. AC and DC susceptibility measurements suggest that the remaining spin degree of freedom freeze completely below 4 K with a drastically increasing relaxation time.

The above-described result can be considered as an interesting analogy to the low-temperature anomaly in doped water ice, especially in the light of its relevance to the problem of residual entropy in ice-water or spin-ice transition. We hope it contributes to a complete understanding of the true ground state of geometrically frustrated systems.

References

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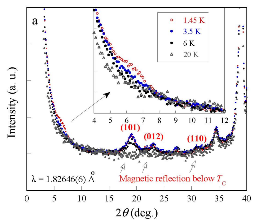

Fig. 1. Neutron diffraction result for $(Co_{0.9}Al_{0.1})_2(OD)_3Cl$.