Experimental Report

Proposal: **4-03-1697** **Council:** 4/2012

Title: Temperature dependence of the magnetic excitations in Haydeeite, a S=1/2 frustrated kagome magnet

This proposal is a new proposal

Researh Area: Physics

Main proposer: **WILLS Andrew S.**

Experimental Team: BOLDRIN David
FAK Bjorn

Local Contact: OLLIVIER Jacques

Samples: α-Cu$_3$Mg(OD)$_6$Cl$_2$

Instrument	Req. Days	All. Days	From	To
IN6	0	4	28/02/2013	04/03/2013

Abstract:

We propose to study the magnetic excitations of the geometrically frustrated S=1/2 kagome quantum magnet Haydeeite, a system similar to the cuboc2 spin-liquid material Kapellasite, but where the magnetic interactions are expected to lead to a different type of spin-liquid ground state with another type of short-range correlations. The temperature dependence of the low-energy magnetic dynamics will be studied on a powder sample using IN5.

Temperature dependence of the magnetic excitations in haydeeite, a $S = 1/2$ frustrated kagome magnet

The quantum spin liquid (QSL), one of the most elusive states in condensed matter physics, is expected to display exotic ground states and fractionalized excitations [1]. A promising model system for the observation of the QSL state is the $S = 1/2$ kagomé antiferromagnet, which is characterized by quantum spins on a geometrically frustrated lattice of corner-sharing equilateral triangles. Unfortunately, suitable experimental realizations of such systems are scarce: most candidate materials either exhibit distortions from the perfect kagome lattice that partly lifts the frustration or have 3D couplings between the kagome planes.

Haydeeite, α-$Cu_3Mg(OD)_6Cl_2$, is a new $S = 1/2$ kagome magnet [2]. It is an isomagnetic Mg analogue of the Zn-based kapellasite [2,3]. Because of the difference in size of the Mg and Cu ions, the risk of Mg/Cu site disorder is strongly reduced in haydeeite, in strong contrast to the Cu/Zn site disorder that leads to 3D coupling in herbertsmithite [4] and depletion of the two-dimensional magnetic lattice of kapellasite [3]. Despite the similarity of the haydeeite and kapellasite chemical structures, electronic band-structure calculations predict rather different exchange integrals for the two compounds.

Our zero-field cooled and field-cooled magnetic susceptibility measurements on haydeeite show a ferromagnetic-like transition in the magnetic susceptibility at $T = 4.2$ K. An upturn at this temperature in χT vs T confirms the ferromagnetic nature of this transition. However, a subsequent decrease at $T = 3.5$ K indicates an additional antiferromagnetic component. The near-zero value of the Curie-Weiss temperature, $\theta_{\rm cw}$, indicates that these competing terms are essentially compensated and frustrated. Somewhat surprisingly, no magnetic Bragg peaks were observed in neutron powder diffraction data taken on D2B below the transition temperature, although the field dependence of the effective moment at $T = 3$ K indicates a small spontaneous moment of 0.02 μ_B [5]. To further our understanding of the magnetic ground state and the spin dynamics of haydeeite, we proposed an inelastic neutron scattering study of a powder sample using IN5.

The experiment was carried out on IN6 using a fully deuterated powder sample of haydeeite in annular geometry in an orange cryostat. Measurements at a wavelength of $\lambda = 5.1$ Å were performed for two temperatures below the Curie temperature T_C ($T = 1.5$ and 3 K) and for two temperatures above T_C ($T = 6$ and 12 K). In the paramagnetic phase, short-range quasielastic spin fluctuations are observed, whereas the magnetically ordered phase displays ferromagnetic Bragg peaks and a strongly dispersive spin-wave excitation. In order to follow the spin-wave dispersion to higher wave vectors and energies, complementary measurements were performed using $\lambda = 4.14$ Å, but these data are not exploitable due to the strong spurious background scattering from the cryostat. Typical counting times were quite long in order to obtain reasonable statistics, between 7 and 16 hours per temperature and wavelength, and the same time for the empty can. Fortunately, we got one night of test time on IN5 (courtesy: Jacques Ollivier), where for a wavelength of $\lambda = 4.8$ Å we obtained cleaner data with better statistics in less than half the time. Clearly, systems with low spin values, such as haydeeite with $S = 1/2$, should be performed on IN5 (which we had proposed in the original beam time application) rather than on IN6.

Typical results of $S(Q, \omega)$ in the spin-liquid phase are shown on a logarithmic scale in Figure 1. The data above the Curie temperature is currently being analyzed while a spin-wave model is being developed for the data in the ferromagnetically ordered phase.

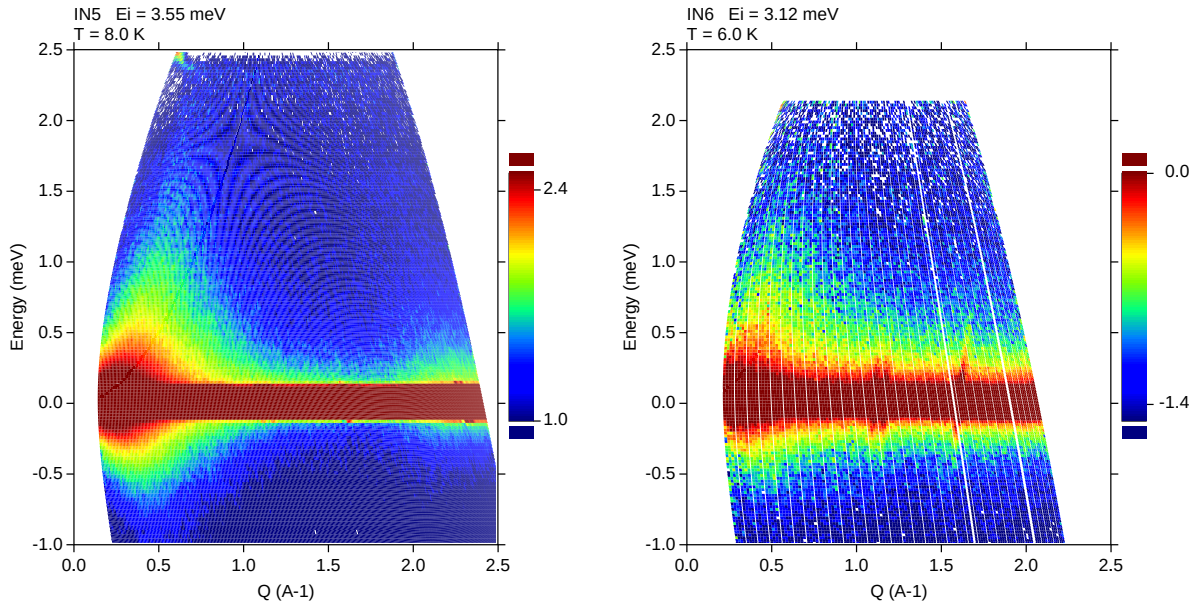

Figure 1: Data above the transition temperature taken on IN5 at $\lambda = 4.8$ Å for 7 hours (left) and on IN6 at $\lambda = 5.12$ Å for 12 hours (right).

References

1. L. Balents, Nature **464** (2010) 199.
2. R. Colman, C. Ritter, and A. S. Wills, Chem. Mater. **20** (2008) 6897.
3. B. Fåk, E. Kermarrec, L. Messio, B. Bernu, C. Lhuillier, F. Bert, P. Mendels, B. Koteswararao, F. Bouquet, J. Ollivier, A. D. Hillier, A. Amato, R. H. Colman, and A. S. Wills, Phys. Rev. Lett. **109** (2012) 037208.
4. T.-H. Han *et al.*, Nature **406** (2012) 406.
5. R. Colman, A. Sinclair, and A. S. Wills, Chem. Mater. **22** (2010) 5774.