Experimental report

09/02/2018

Proposal: **4-05-644** **Council:** 4/2016

Title: Magnetic excitations in the S=1/2 kagome antiferromagnet and quantum spin liquid candidate `Mg-herbertsmithite', $MgCu_3(OD)_6Cl_2$

Research area: Physics

This proposal is a continuation of 4-05-629

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Samples: $MgCu_2(OD)_6Cl_2$

Instrument	Requested days	Allocated days	From	To
IN4	6	6	16/12/2016	22/12/2016

Abstract:

We propose to study the magnetic excitations in the quantum spin liquid candidate `Mg-herbertsmithite', which forms a geometrically perfect and highly frustrated two-dimensional kagome lattice of S=1/2 quantum spins with dominant antiferromagnetic nearest-neighbor interactions. No long-range order is observed down to T=20 mK and recent theoretical work predicts a rich phase diagram with different types of gapless, chiral, and U(1) quantum spin liquids that await experimental confirmation. The measurements aim at determining the high-energy part of the magnetic excitations, which will complement the low-energy measurements already performed on IN5, and hopefully lead to a full picture of this fascinating S=1/2 kagome physics.

Magnetic excitations in the $S = 1/2$ kagome antiferromagnet and quantum spin liquid candidate 'Mg-herbertsmithite', γ-$MgCu_3(OD)_6Cl_2$

The search for quantum spin liquids (QSLs) is a cornerstone of condensed matter physics. These are new and exotic states of matter that in two dimensions were proposed by Anderson to underpin the transition to unconventional superconductivity of the high-T_C cuprates [1]. Formed by geometric frustration of a magnetic lattice and the quantum fluctuations that are maximal for $S = 1/2$ spins, QSLs do not break symmetry as conventional magnets do, but are instead characterised by dynamic short-range correlations and fractionalised excitations [2,3]. The $S = 1/2$ kagome antiferromagnet (KAFM) is considered the most promising model system for the realisation of 2D QSLs.

Despite the importance of such systems, good experimental examples are rare. Much effort has been focused on the paratacamite family of minerals, $Cu_3(Cu,Zn)(OH)_6Cl_2$, and in particular the $S = 1/2$ KAFM herbertsmithite, γ-$Cu_3Zn(OH)_6Cl_2$ [3–5]. Since its discovery, several factors such as Cu/Zn inter-site mixing and Dzyaloshinski-Moriya (DM) antisymmetric exchange have been found to occur. Inter-site mixing within herbertsmithite couples the kagome planes, destroys the 2-dimensionality of the system and introduces an additional term into the Hamiltonian that acts to relieve the frustration. Extracting the intrinsic physics of the $S = 1/2$ KAFM is therefore complicated, although recent experiments have found several hallmarks of a quantum spin liquid ground state [3].

Interest in paratacamite based materials as model $S = 1/2$ KAFMs has grown further with the discoveries of kapellasite, α-$Cu_3Zn(OH)_6Cl_2$, a polymorph of herbertsmithite, and its magnesium analogue haydeeite, α-$Cu_3Mg(OH)_6Cl_2$ [6,7]. In these materials, the kagome lattice is constructed by regular doping of a triangular lattice of Cu^{2+} with Mg^{2+} or Zn^{2+} ions, as opposed to the 3-dimensional parent pyrochlore lattice in herbertsmithite. As such, any antisite disorder between the divalent cation sites in both haydeeite and kapellasite cannot introduce 3-dimensionality into the magnetic system. In haydeeite, the ground-state and the excitations measured recently by INS experiments are in agreement with theoretical predictions of a classical multi-exchange model on the $S = 1/2$ kagome lattice [8,9]. In the closely related paratacamite member kapellasite, INS revealed unique QSL behaviour with dynamic short-range gapless excitations resembling the non-coplanar cuboc2 regular state [2,9], but which in view of further theoretical work can be understood in terms of the chiral gapless spin liquid state CSL-A [10], which has similar short-range correlations.

Here, we focused on another member of the paratacamite family, 'Mg-herbertsmithite', γ-$MgCu_3(OH)_6Cl_2$, which has not yet attracted much attention. This material is isoelectronic and isostructural with herbertsmithite (Figure 1a) and possesses a similar degree of magnetic frustration. Despite strong mean-field antiferromagnetic interactions, $\theta_W = -284\,K$, μSR experiments do not detect any long-range magnetic order down to $T = 20\,mK$, giving a record frustration index $f = \theta_W/T_N > 10000$, whilst the low-T spin dynamics are consistent with a QSL ground-state (Figure 1b) [7,11].

The experiment was carried out on IN4 using 3.6 g of fully deuterated powder sample of γ-$MgCu_3(OH)_6Cl_2$ in slab geometry in an orange cryostat. Measurements were performed at temperatures of $T = 1.5$, 9, 50, and 125 K using wavelengths of 1.6, 2.2, and 3.2 Å. Typical counting times were 6 to 24 hours per run. The magnetic signal seen one year earlier on IN5 on the same sample was not observed on IN4, see Figure 2. It appears as if the sample, despite having been stored in a sealed container between the two experiments, has deteriorated. This highlights the difficulties with the current proposal system to study materials that are sensitive to external aggressions or have limited stability in time.

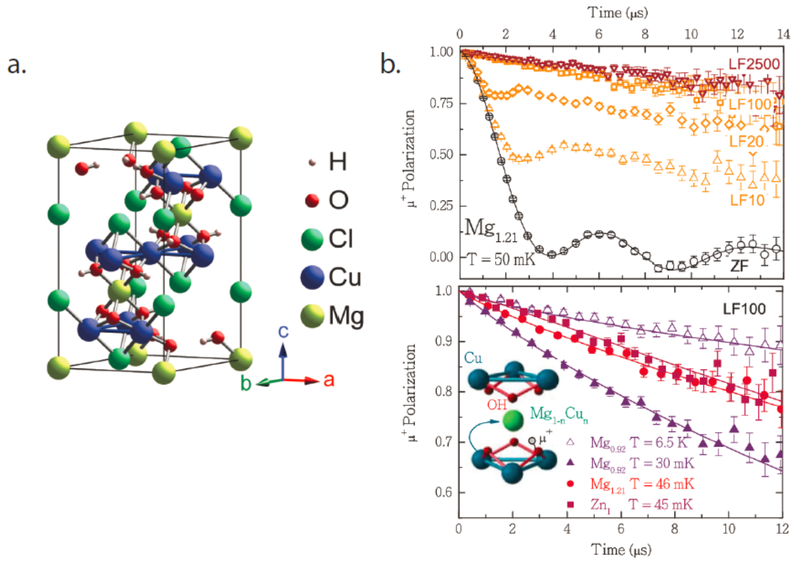

Figure 1: (a) Crystal structure of 'Mg-herbertsmithite'. (b) Muon polarisation.

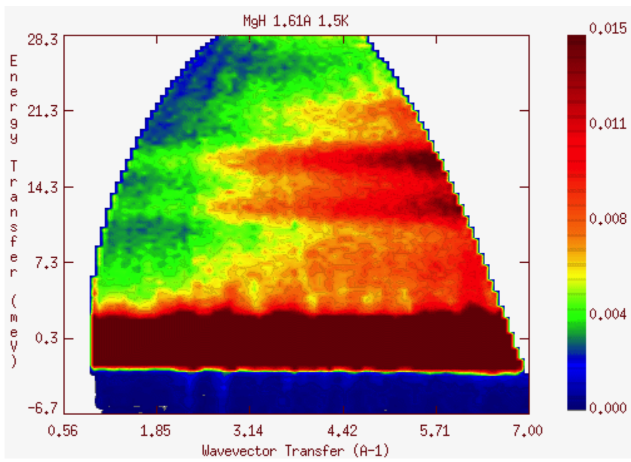

Figure 2: $S(Q, E)$ of 'Mg-herbertsmithite' measured on IN4 at $T = 1.5$ K with an incoming wavelength of $\lambda = 1.6$ Å.

References

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