Experimental report

25/08/2021

Proposal:	**5-32-909**	**Council:**	4/2020

Title: Persistent Magnetic Correlations above TN in the Frustrated FCC Lattices Ba2MnMO6 (M=W, Te)

Research area: Physics

This proposal is a resubmission of 5-32-902

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Experimental team: Lucile MANGIN-THRO
Otto MUSTONEN

Local contacts: Lucile MANGIN-THRO

Samples: Ba2MnWO6
Ba2MnTeO6

Instrument	Requested days	Allocated days	From	To
D7	5	5	14/02/2021	19/02/2021

Abstract:

We have used a combination of magnetic susceptibility, neutron scattering and muon spin relaxation measurements to examine the magnetic properties of the frustrated lattices of the cubic double perovskites Ba2MnMO6 (M=W, Te) These compound maintain Fm-3m symmetry to 2 K, as identified using neutron diffraction data, with Mn/M cations ordered in a rock salt manner across the octahedral sites of the perovskite structure. The S=5/2, L=0 Mn2+ cation introduces no orbital anisotropy into the system and makes them ideal candidates to test ideas of frustration. Muon spin relaxation and inelastic neutron scattering measurements reveal the presence of a short-range correlated magnetic state above TN in both compounds. We will use D7 to measure diffuse magnetic scattering in order to characterize the short-range corelated state, and investigate how it is linked to the magnetically ordered state.

Persistent Magnetic Correlations above T_N in the Frustrated FCC Lattices Ba_2MnMO_6 (M=W, Te)

Proposal no: 5-32-909
Instrument: D7, 5 days of beamtime
Experiment team: Edmund Cussen, Otto Mustonen, Helen Walker, Charlotte Pughe
Local contact: Lucile Mangin-Thro

Ba_2MnTeO_6 and Ba_2MnWO_6 are isostructural Mn^{2+} double perovskites with the cubic space group *Fm*-3*m* [1–4]. The magnetic Mn^{2+} cation forms a fcc lattice, and these compounds are essentially ideal realisations of the S = 5/2 fcc Heisenberg anttiferromagnet model. Despite being diamagnetic, the Te^{6+} $4d^{10}$ and the W^{6+} $5d^0$ cations play a significant role on the magnetic interactions in these compounds. Ba_2MnTeO_6 has a dominant J_1 interaction leading to a Type I antiferromagnetic structure, while J_1 and J_2 are almost equally strong in Ba_2MnWO_6 leading to a Type II antiferromagnetic structure. This difference is due to an orbital hybridization effect on the Te^{6+} and W^{6+}, which allow different extended superexchange pathways in the material [5].

We have previously extensively characterised the ground state properties of Ba_2MnTeO_6 and Ba_2MnWO_6 [3,4]. However, we also observed evidence of a short-range correlated magnetic state above TN in both compounds. In muon experiments, the full paramagnetic fraction is not recovered at T_N. Instead, the paramagnetic fraction increased gradually after T_N suggesting the formation of a short-range correlated state. Hints of this state were also observed in inelastic neutron scattering measurements.

In order to characterise the proposed short-range correlated magnetic states above T_N in Ba_2MnTeO_6 and Ba_2MnWO_6, we performed a polarised neutron experiment on the D7 instrument at ILL. We extracted the diffuse magnetic scattering at a number of temperatures using XYZ polarisation analysis. The data was analysed using Spinvert [6].

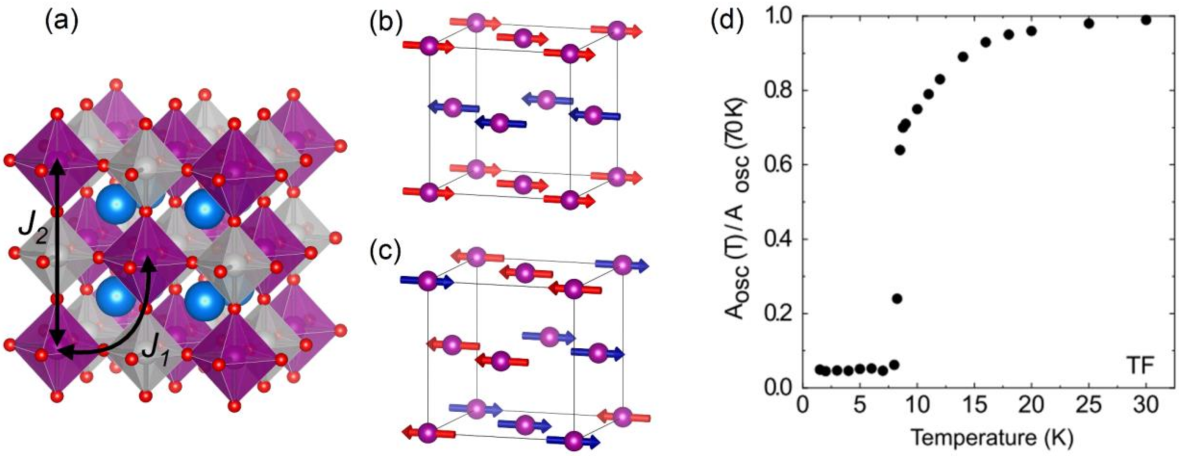

Figure 1. (a) The double perovskite structure of Ba_2MnTeO_6 and Ba_2MnWO_6 and the J_1 and J_2 interactions of the fcc Heisenberg model. (b) The Type I magnetic structure of Ba_2MnTeO_6. (c) The Type II magnetic structure of Ba_2MnWO_6. (d) Paramagnetic fraction of Ba_2MnWO_6 from muon experiments. The full paramagnetic fraction is not recovered at T_N = 8 K, but at gradually at higher temperatures. The panels are from refs. [3,4].

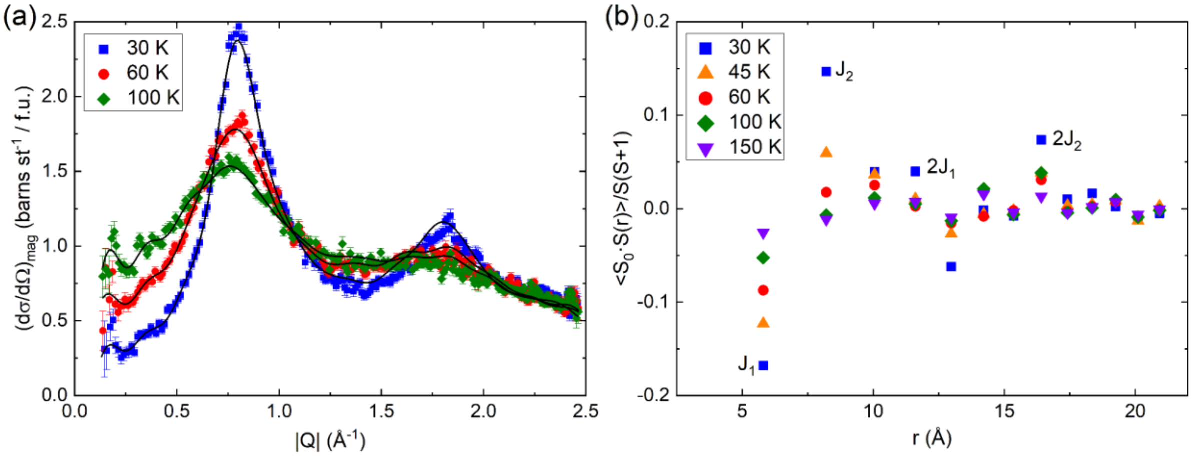

Figure 2. (a) Diffuse magnetic scattering of Ba_2MnTeO_6 at different temperatures and the corresponding Spinvert fits. (b) The spin-spin correlations obtained from Spinvert fits.

The diffuse magnetic scattering of Ba_2MnTeO_6 at a number of temperatures is shown in Figure 2. We observe a broad peak at $|Q|$ ~0.78 Å^{-1}, which is best visible in the data at 30 K. This is 10 K above T_N = 20 K. The main magnetic peak observed in neutron diffraction in the ordered state is at $|Q|$ = 0.78, which suggests that the short-range correlated state above T_N evolves out of the magnetic order below T_N. Another broad peak is observed at $|Q|$ ~1.8 Å^{-1}. This is likely related to two magnetic peaks observed in the ordered state at $|Q|$ = 1.71 and 1.83 Å^{-1}. Significant diffuse magnetic scattering was observed up to 150 K, which is quite high in comparison to T_N = 20 K.

The spin-spin correlations in Ba_2MnTeO_6 extracted from Spinvert fits are shown in Figure 2(b). In the magnetically ordered state, 2/3 of the nearest-neighbor J_1 spins couple antiferromagnetically, while all next-nearest neighbour J_2 spins couple ferromagnetically. Our D7 experiment shows that the short-range order above T_N is closely related to the magnetic order in Ba_2MnTeO_6: J_1 spin-spin correlations are strongly antiferromagnetic, while the J_2 correlations are ferromagnetic. The spin-spin correlations get weaker as temperature is increased, but are correlations persists up to at least 150 K.

The diffuse magnetic scattering for Ba_2MnWO_6 is shown in Figure 3. A broad peak in the magnetic scattering is observed at $|Q|$ ~0.68 Å^{-1}. This is related to the main magnetic Bragg peak observed at $|Q|$ = 0.66 Å^{-1} in the magnetically ordered state. The position of the main diffuse peak in Ba_2MnWO_6 is shifted in comparison to Ba_2MnTeO_6, which is understandable, as the magnetic structures of these two compounds are also different. Diffuse magnetic scattering was observed up to 100 K, which is significantly above T_N = 8 K for Ba_2MnWO_6.

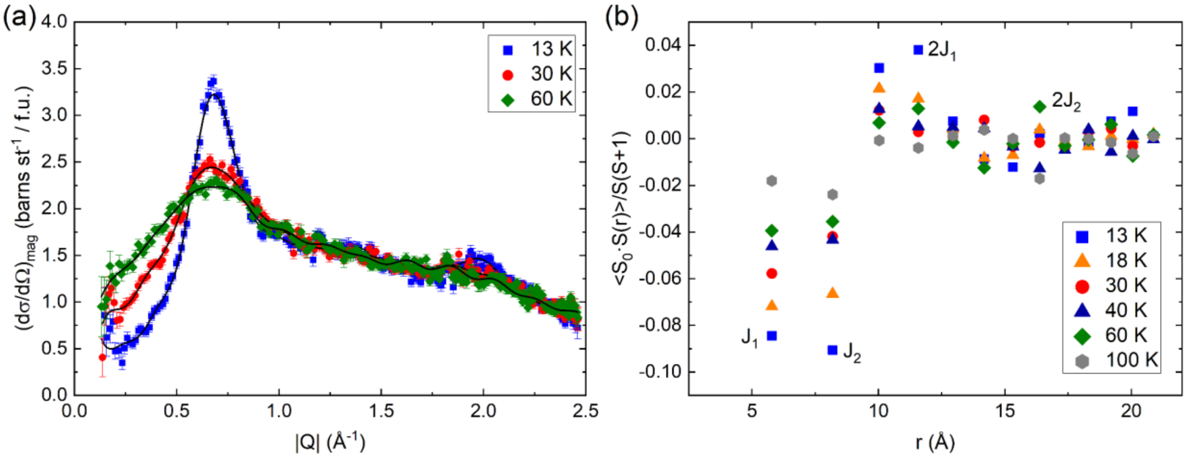

Figure 3. (a) Diffuse magnetic scattering of Ba_2MnWO_6 at different temperatures and the corresponding Spinvert fits. (b) The spin-spin correlations obtained from Spinvert fits.

The spin-spin correlations in Ba_2MnWO_6 extracted from Spinvert fits are shown in Figure 2(b). In the magnetically ordered state, the spin-spin correlation for the nearest-neighbor J_1 spins is 0, as there are an equal number of ferro- and antiferromagnetically coupled spins. In contract, all next-nearest-neighbor J_2 spins couple antiferromagnetically. The spin-spin correlations in the short-range magnetic state in Ba_2MnWO_6 are related to the magnetic order below T_N, although the J_1 correlation is antiferromagnetic instead of 0. The spin-spin correlation for J2 is strong and antiferromagnetic as expected. The antiferromagnetic J_1 spin-spin correlations can be understood in terms of the magnetic exchange constants obtained from inelastic neutron scattering measurements below T_N. This revealed the J_1 interaction almost equal to J_2, which explains the presence of antiferromagnetic J_1 spin-spin correlations above T_N.

Our D7 experiment was a success and we would like to give special thanks to our local contact Lucile Mangin-Thro, who ran the experiment for us due to the COVID-19 pandemic. We were able to show that the short-range magnetic states in Ba_2MnTeO_6 and Ba_2MnWO_6 persist to very high temperatures in comparison to T_N, and that they are related to the magnetic order below T_N. We plan to write a paper based on this experiment in the near future.

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