Experimental report

10/08/2022

Proposal: **5-31-2829** **Council:** 10/2020

Title: Investigating Spin Order in a New Perovskite with Multiple Cation Order

Research area: Materials

This proposal is a new proposal

Main proposer: **Graham MCNALLY**

Experimental team: Clemens RITTER

Local contacts: Clemens RITTER

Samples: CaMnFeMoO6

Instrument	Requested days	Allocated days	From	To
D20	2	2	21/05/2021	23/05/2021

Abstract:

A new perovskite of formula CaMnFeMoO6, with columnar A-site and rock-salt B-site cation order, has been synthesised using high-pressure high-temperature techniques. From magnetisation measurements, magnetic order is evidenced with TC of 290 K. However, magnetisation has a maximum at 200 K and subsequently decreases with lowering temperature, suggesting that this new molybdate has a significantly different A-site magnetic structure than is seen in the related CaMnFeReO6. An additional low temperature magnetic transition is observed in ZFC magnetisation measurements, which is possibly a signature of a spin reorientation, as observed in Mn2FeReO6 and Mn2(Fe0.8Mo0.2)MoO6. Two days of beamtime on D20 are requested to determine the spin structures of a 100 mg sample of CaMnFeMoO6 as a function of temperature between 2 and 350 K. These results will be essential to understand the magnetic properties of this material.

Experimental Report

A sample of $CaMnFeMoO_6$ (178 mg), comprised of a mixture of samples prepared from high-pressure high-temperature synthesis, was measured in zero field at several temperatures on D20 at the ILL. A temperature dependant study of the magnetic structure of $CaMnFeMoO_6$ has allowed us to determine the magnetic structure of this ordered perovskite and disentangle the rather complex evolution of magnetic order arising from the four different independent magnetic sites in the structure.

Relating this analysis of individual site moments from neutron data to our bulk magnetisation data now allows for a more comprehensive understanding of the properties of this material. We find that Fe/Mo moments order ferrimagnetically at ~290 K, and a subsequent order of Mn^{2+} moments arises at ~140 K. Square planar Mn and tetrahedral Mn moments are ferromagnetically ordered, but both are antiferromagnetically coupled to Fe^{3+}, resulting in a gradual reduction of the total magnetic moment of the system as Mn^{2+} moments slowly saturate and become comparable to, and then exceed, the Fe^{3+} magnitude. This confirms that complex evolution of collinear moments is responsible for the unusual temperature evolution of magnetic properties, rather than spin reorientation(s) being the source of this.

We find that there is a relatively strong degree of cation order across the five different cation sites. However, unlike many double perovskites with 8-coordinate Mn A-sites, where no Mn is found to mix onto the B-sites, we find that in this tetragonal “double-double” perovskite structure there is now some measureable Mn/Fe (A/B) site mixing which is possible due to the 4-coordinate A-sites. This non-negligible degree of site mixing leads to a reduced saturated magnetic moment of the Fe^{3+} B-site due to disorder, and has implications for the evolution of the magnetic structure with temperature.