Experimental report

16/09/2024

Proposal: 5-31-2938 **Council:** 10/2022

Title: Determining the nuclear and magnetic structures of multi-component manganese antiperovskite nitride alloys

Research area: Materials

This proposal is a new proposal

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Samples: Mn3(Cu,Ga,Ni,Zn)N
Mn3(Cu,Ga,Sn,Zn)N

Instrument	Requested days	Allocated days	From	To
D2B	2	1	05/06/2023	06/06/2023
D20	2	0		

Abstract:

Materials with large caloric effects offer the opportunity to replace current cooling technologies that require the compression of hazardous gases. Whilst magnetocaloric materials driven using magnetic fields are currently the most developed for applications, there is growing interest in barocaloric materials driven using hydrostatic pressure. Our recent studies on the Mn antiperovskites Mn3(A,B)N have revealed giant barocaloric effects with minimal thermal hysteresis at first-order paramagnetic to antiferromagnetic transitions. Alloying on the (A,B) site facilitates the tuning of the transition for room temperature applications whilst maintaining the giant barocaloric response. Inspired by the field of high-entropy alloys, we have now begun to investigate multi-component systems with >3 elements on a single crystallographic site. For Mn3(Cu,Ga,Ni,Zn)N and Mn3(Cu,Ga,Sn,Zn)N we find the magnetic transition remains strongly 1st-order yet the hysteresis is again negligible. Here we propose to use D20 to determine the magnetic structure of these compounds as a function of temperature. Ultimately, the results will guide future exploration of these flexible and functional material family

1 PRINCIPAL INVESTIGATOR
David Boldrin (PI), Connor Inglis University of Glasgow

2 EXPERIMENT DETAILS	
Proposal No: **5-31-2938**	
Title: **Determining the nuclear and magnetic structures of multi-component manganese antiperovskite nitride alloys** (DOI: 10.5291/ILL-DATA.5-31-2938)	
Instrument: **D20**	
Dates scheduled: 05/06/2023	No. Days allocated: 1
Date of experimental report: 15/09/2023	

3 EXPERIMENT OBJECTIVES

The experimental aim of this neutron experiment was to understanding if the magnetic structure of doped Mn_3AN anti-perovskites adopt the non-collinear antiferromagnetic structure present in many of the ternary $Mn_3(A,B)N$ compounds, and to investigate whether the transition involves a change of structural symmetry.

4 EXPERIMENT REPORT

The experiment was performed remotely due to last minute complications with our travel arrangements from the UK. The beamline scientists kindly performed the experiment on our behalf. Neutron diffraction was performed using lambda = 2.41A on the quaternary anti-perovskite sample ($Mn_3Cu_{0.25}Zn_{0.25}In_{0.25}Sn_{0.25}N$) between 0 ˚ and 150 ˚ 2θ at temperatures 10, 30, 50, 100, 200, 250 and 314 K. Since $Mn_3Cu_{0.25}Zn_{0.25}In_{0.25}Sn_{0.25}N$ transitions at 301 K, magnetic peaks appear in the diffraction data from 250 K down to 10 K.

Rietveld refinement of the data was performed using GSAS-II, with the nuclear peaks fitted using CIF files from previous refinements of X-ray diffraction data of this sample. Magnetic peaks in the T = 10K dataset were refined using three different magnetic structures R-3, R-3m and R-3m' as these are the structures commonly adopted by these manganese nitride anti-perovskites. From comparing the refinement of all three magnetic structures, the R-3 structure gave the lowest chi-squared and R-values across the entire temperature range. This suggests that $Mn_3Cu_{0.25}Zn_{0.25}In_{0.25}Sn_{0.25}N$ exhibits a stable non-coplanar magnetic structure as opposed to the non-collinear structures commonly seen in these materials. The volume change across the transition was also established from our temperature dependent data. No significant changes in the magnetic structure were observed upon heating, although analysis is still ongoing to fully confirm this.

The literature suggests that the valence electrons (s- and p-electrons) of the A-site metals in Mn_3AN may influence how the magnetic structure of the anti-perovskite develops, however this is still being investigated. Further analysis of this material will likely involve inelastic neutron scattering experiments (possibly using PANTHER) to probe how the doping of the anti-perovskite A-site influences the phonon behaviour compared to undoped anti-perovskite samples. The results will be a key part of the thesis of Connor Inglis and will be combined with a number of other characterisation techniques in a publication exploring the link between A-site chemistry and magnetic properties in this material family.

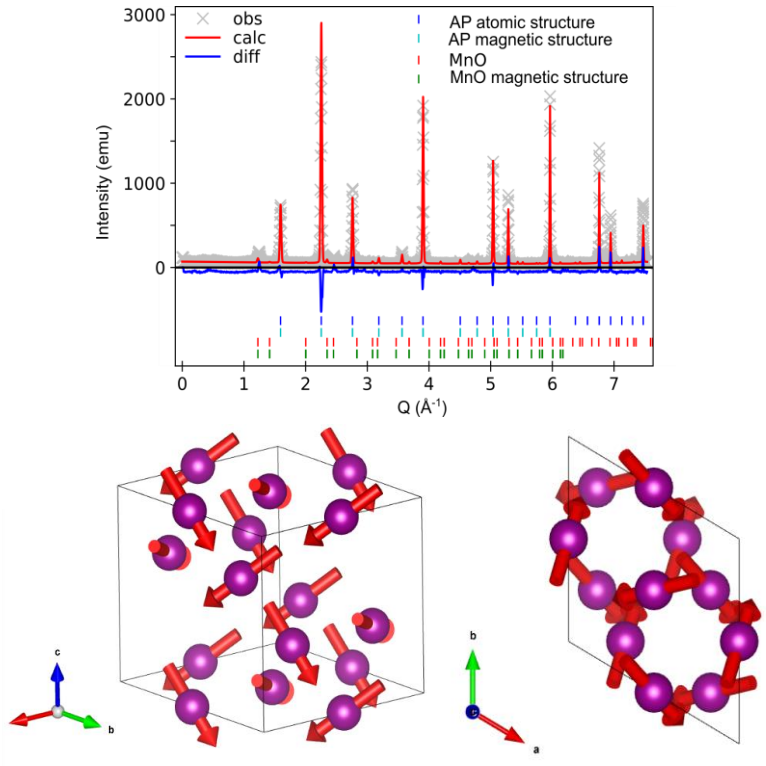

Figure 1: *Neutron diffraction data taken from D20 of $Mn_3Cu_{0.25}Zn_{0.25}In_{0.25}Sn_{0.25}N$ at 10 K with 3D representation of the refined magnetic structure showing a non-coplanar magnetic structure*