# Experimental report

28/02/2022

**Proposal:** 5-32-906 **Council:** 4/2020

**Title:** Nanoscale magnetic inhomogeneitiesin the triple A-site columnar ordered quadruple perovskites RMn3O6 (R= Y or Dy)

**Research area:** Physics

**This proposal is a new proposal**


**Main proposer:** **Roger JOHNSON**

**Experimental team:** Nina-Juliane STEINKE

**Local contacts:** Nina-Juliane STEINKE
Robert CUBITT


**Samples:** Y0.89Mn3.11O6
Y0.92Mn3.08O6
Dy1.03Mn2.97O6

| Instrument | Requested days | Allocated days | From | To |
|---|---|---|---|---|
| D33 | 4 | 3 | 03/02/2021 | 06/02/2021 |


## Abstract:

The YMn3O6 and DyMn3O6 quadruple perovskite manganese oxides have a rich and varied magnetic phase diagram. In Y0.89Mn3.11O6, Y0.92Mn3.08O6 and Dy1.03Mn2.97O6 we observe the experimental signatures of a Griffiths phase from DC magnetometry measurements, which we believe to originate from spin doping, analogous to the emergence of a Griffiths phase in the simple perovskite oxides from charge doping. Following neutron diffraction measurements we have determined that YMn3O6 adopts a long range ordered ferrimagnetic structure, which succeeds the Griffiths phase and persists down to the lowest measured temperature. In comparison no long range order has been observed in Dy1.03Mn2.97O6, and instead broad diffuse scattering is observed that is indicative of short range order. Furthermore the appearance of short range order is observed from ~30 K in both YMn3O6 samples. We propose a SANS experiment to directly observe and determine the nature of the Griffiths phase and the low temperature short range clustered phase. This experiment will be crucial in building the magnetic phase diagram of these systems, and would be a novel observation of a Griffiths phase induced by spin doping.

# Nanoscale magnetic inhomogeneities in the triple A-site columnar ordered quadruple perovskite $RMn_3O_6$ (R = Y or Dy)

A. M. Vibhakar,[1] D. D. Khalyavin,[2] P. Manuel,[2] N. J. Steinke,[3] A. A. Belik,[4] and R. D. Johnson[5]

[1]*Diamond Light Source Ltd, Harwell Science and Innovation Campus, Didcot, Oxfordshire, OX11 0DE, United Kingdom*

[2]*ISIS facility, Rutherford Appleton Laboratory-STFC, Chilton, Didcot, OX11 0QX, United Kingdom*

[3]*Institut Laue-Langevin, 71 avenue des Martyrs, F-38042 Grenoble, France*

[4]*Research Center for Functional Materials, National Institute for Materials Science (NIMS), Namiki 1-1, Tsukuba, Ibaraki 305-0044, Japan*

[5]*Department of Physics and Astronomy, University College London, Gower Street, London, WC1E 6BT, United Kingdom*

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Small angle neutron scattering (SANS) measurements were performed to probe whether any short range ferromagnetic order is present between 111 K and $T_C$ in the $[Y_{2-x}Mn_x]MnMnMn_4O_{12}$ columnar ordered quadruple perovskite manganites $x = 0.21$ and $x = 0.16$. SANS measurements were performed on D33 at the Institute Laue Langevin (ILL). Each of the samples listed above were pressed into a flat disc like pellet, and placed in an aluminium sample mount. Data was collected in a single detector setting for both samples ( $0.003\text{Å}^{-1} \leqslant Q \leqslant 0.1\text{Å}^{-1}$ $\Delta Q = 0.0005$ $\text{Å}^{-1}$). Data on $Y_{1.84}Mn_{0.16}MnMnMn_4O_{12}$ was collected on cooling at 130 K, between 90 K and 60 K in 10 K steps and between 60 K and 1.5 K in 15 K steps. Data on $Y_{1.79}Mn_{0.21}MnMn_4O_{12}$ was collected on cooling at 130 K, 90 K and between 80 K and 5 K in 5 K steps.

The transmitted intensity from the full beam and the scattered intensity from an empty sample cell was used to normalise each SANS pattern, so that any contributions from the transmitted beam and aluminium sample holder were subtracted away. For the temperature dependent data collected under zero applied field, each pattern was also normalised by the SANS pattern collected at high temperature, to subtract away the nuclear contribution to the scattered intensity. The scattering length density is calculated by determining the average neutron scattering lengths in a unit cell, while the magnetic scattering length density is the average magnetic moment in a unit cell, which is zero for antiferromagnetic order, and hence SANS is only sensitive to the presence of ferromagnetism.

Fig. S1 shows the variation in the magnetic intensity summed over the collected Q range, and plotted as a function of temperature for both samples. No statistically significant change to the magnetic intensity was observed between $T_C$ and 111 K of either sample, indicating the absence of any ferro/ferri magnetic clusters in this temperature region.

Magnetic SANS intensity begins to appear below $T_c$ in both samples, Fig. S1. To fit this intensity, a number of

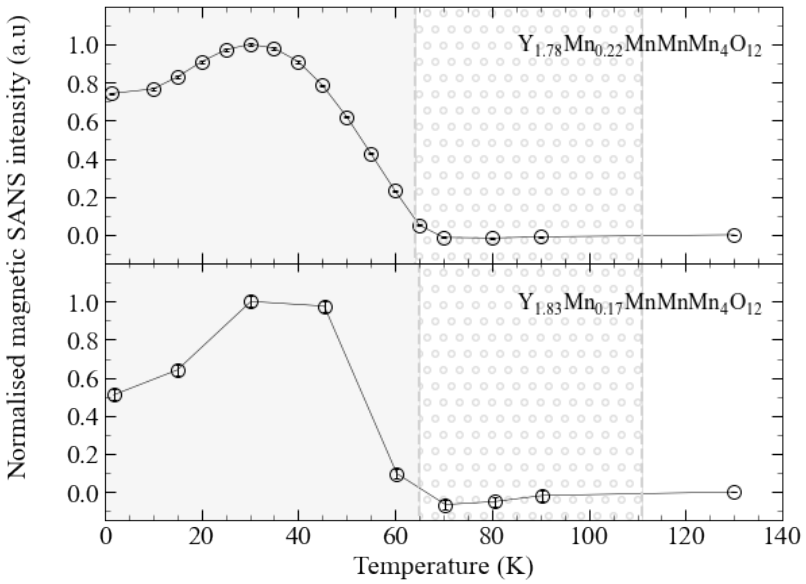


FIG. S1. Temperature dependence of the normalised magnetic SANS intensity of the Q = 0.0028 Å$^{-1}$ peak as a function of temperature for the (a) $Y_{1.79}Mn_{0.21}MnMn_4O_{12}$ and (b) $Y_{1.84}Mn_{0.16}MnMnMn_4O_{12}$ samples.

standard expressions for the form factor were tested, and a Lorentzian squared, centred about Q = 0, gave the best fit to the data. However there was a high degree of correlation between the fit parameters (the amplitude and half-width half maximum), and it is therefore likely that the SANS measurements had only captured the tail of the peak, and as a consequence the amplitude and half width half maximum could not be decorrelated. The variation in the peak intensity as a function of temperature, shown in Fig. S1, reproduces the observed variation in intensity of the Bragg peaks measured from neutron powder diffraction measurements, and thus this intensity was identified as scattering from the (0, 0, 0) reflection of the FIM phase.

Field dependent SANS measurements were also performed on both samples. In the presence of any ferro/ferri magnetic clusters, one would expect the field to couple to the net magnetisation of the cluster and cause a change to the observed magnetic SANS intensity, as has been observed in other systems. No statistically significant change to the SANS intensity was observed between $T_C$ and 111 K as a function of magnetic field, as shown in Fig. S2(b), conclusively ruling out the presence of any ferromagnetic order. Upon application of a magnetic field, the (0, 0, 0) reflection appears to sharpen at 4 K, Fig. S2(a), which may imply that the magnetic field suppress the AFM clusters and favours the FIM phase.

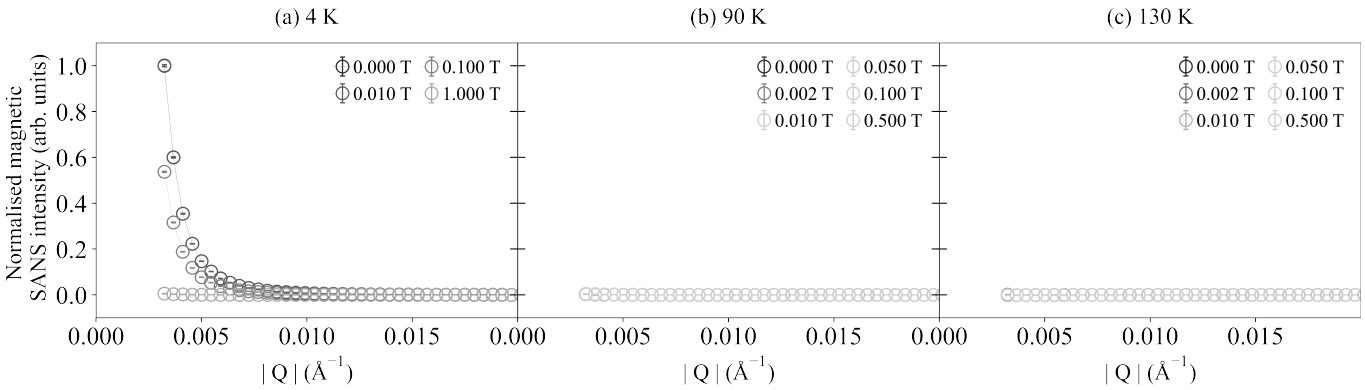


FIG. S2. Normalised magnetic SANS intensity under different applied magnetic fields at (a) 4 K (b) 90 K and (c) 130 K for $Y_{1.79}Mn_{0.21}MnMn_4O_{12}$ .