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Long- and short-range magnetic order in the spinel $Co_2Ru_{1-x}Mn_xO_4$

G.E. Granroth[a,b,*], D. Mandrus[b], V. Keppens[c], S.E. Nagler[b]

[a] Spallation Neutron Source, Oak Ridge National Laboratory, P.O. Box 2008, MS 6474, Oak Ridge, TN 37831, USA
[b] Condensed Matter Sciences Division, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, TN 37831, USA
[c] Department of Physics, The University of Mississippi, Oxford, MS 38677, USA

Abstract

Neutron scattering measurements were performed on the $x = 0$, 0.5, and 0.7 concentrations of the insulating spinel $Co_2Ru_{1-x}Mn_xO_4$. The experiments show that the $x = 0.7$ system orders as a long-range ferrimagnet, the $x = 0.5$ system has a ground state that is a mix of long-range ferrimagnetic order and short-range order, and the $x = 0$ system only possesses short-range order.
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Magnetic ions on a spinel lattice participate in diverse ground states controlled by several interactions between two sites. Specifically, magnetic ions can occupy interstitial sites with tetrahedral(A) or octahedral(B) coordination. The superexchange through the anion produces strong antiferromagnetic A–B coupling, geometrically frustrated antiferromagnetic B–B coupling, and negligible coupling between the A–A sites. When the A–B interaction is dominant, the magnetic ground state is described by Neél ferrimagnetism [1]. Diverse magnetic ground states are observed in systems where the A–B interaction is reduced by replacing the A site magnetic ions with non-magnetic ions [2,3].

Non-magnetic Co^{3+}, when placed on the B sites, provides a path for antiferromagnetic exchange between neighboring A sites [4]. This results in competing A–B, B–B, and A–A interactions. The system $Co_2Ru_{1-x}Mn_xO_4$ is a spinel that fits the above criterion. The compound Co_2MnO_4 ($x = 1$) has strong A–B interactions and is known to order ferrimagnetically for $T < 180$ K [5]. Recent magnetization studies [6,7] show that as x is reduced the A–B interaction weakens and the nature of the ordered state changes. This paper summarizes neutron scattering experiments that elucidate long- and short-range ordered (SRO) ground states observed for the concentrations $x = 0$, 0.5, and 0.7.

For all of the studied concentrations, the A sites are populated with $S = \frac{3}{2}$ (Co^{2+}), $\frac{1}{2}$ of the B sites are randomly populated by Co^{3+}, and the remaining B sites are populated, according to x, with $S = \frac{1}{2}$ (Ru^{3+}) or $S = 2$ (Mn^{3+}). Materials were prepared as described by Mandrus et al. [6,7] and examined by neutron diffraction using the HB1A and HB1 spectrometers at the HFIR facility, ORNL [8]. Measurements were carried out in two-axis mode using incident neutron energies of 14.7 meV (HB1A) and 13.5 meV (HB1). Powder profiles over the angular range $10° < 2\theta < 110°$ were performed at room temperature and at 10 K. The results show that the $x = 0.7$ system exhibits ferrimagnetic (FIM) order identical to the $x = 1$ compound [5]. Magnetic Bragg peak (MBP) intensity scales as expected for the case when 30% of the magnetic B sites are occupied by $S = \frac{1}{2}$ Ru^{3+}. The MBP intensity for the $x = 0.5$ system is also consistent with FIM order assuming that it is reduced to account for magnetic moment that scatters into a diffuse feature near the (1 1 1) reflection. These measurements

*Corresponding author. Tel.: +1-865-576-0900; fax: +1-865-241-5177.
E-mail address: granrothge@sns.gov (G.E. Granroth).

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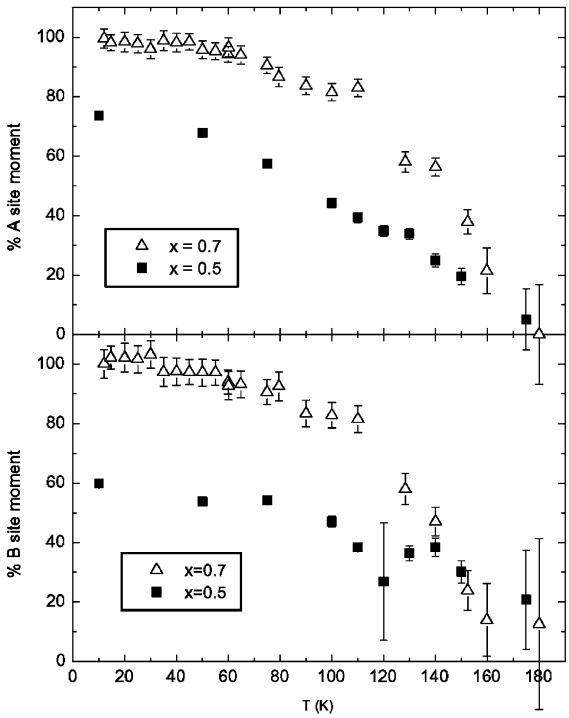

Fig. 1. Percentage of moment on each site participating in long-range FIM order.

also suggest the onset of the long-range state at $T \approx 160$ K for both concentrations. At $T = 10$ K the $x = 0$ system exhibits only the broad diffuse scattering for small 2θ.

The FIM structure leads to significant intensity at the (1 1 1) and (2 2 0) positions, the latter arising strictly from magnetic ions at the A sites. The scattering in the vicinity of each of these peaks was measured as a function of 293 K > T > 10 K. No magnetic scattering was observed for $T \geqslant 180$ K, and intensity changes arising from Debye–Waller and other effects were negligible between 200 and 180 K. We therefore used the scattering at 180 K as a measure of the non-magnetic background. At $T < 180$ K, each scan was fitted to a resolution limited Gaussian peak and a Lorentzian convolved with instrumental resolution centered at the peak position. The 180 K scattering was subtracted from this data. The remnant Gaussian intensities were used to estimate the FIM ordered moment at the A and B sites. The data were normalized to the magnetic intensity expected for full FIM ordering of all the magnetic ions. The result, expressed as a percentage, is plotted in Fig. 1 for the A (upper panel) and B (lower panel) sites. For $x = 0.7$ both sites saturate at low T. For $x = 0.5$ at the lowest measured $T = 10$ K, the A and B site moments are reduced by 25% and 40% of their expected values, respectively. The sum of the intensity in the broad Lorentzian component and the MBPs is consistent with the scattering expected from the total moment. This implies that the residual moment participates in a SRO state where the B site moment fluctuates more strongly. For the $x = 0$ system, only the SRO component is observed.

In summary, $Co_2Ru_{1-x}Mn_xO_4$, where $x = 0$, 0.5, and 0.7, has been investigated by neutron diffraction. The 0.7 system orders as a Neél ferrimagnet. The 0.5 system has a mix of this FIM state and a short-range ordered state. In the $x = 0$ system only a SRO state is observed.

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