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Structures, spin glass and spin states in perovskite

GdMn_xCo_{1-x}O₃ (x≤0.5)

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Introduction

In magnetic systems frozen spins in random alignment causes the spin glass state. The spin glass state has been observed in spinels, amorphous materials, and mostly in dilutes magnetic alloys. Spin glass could arise from paramagnetic state in disordered system or from paramagnetic to ferromagnetic state. In latter case, the spin glass co-exists with ferromagnetism. Recently, spin glass states have also been widely observed in a number of ReMO₃ perovskite (Re=rare-earth, M=3d transition metal) compounds. In particular, REMn_{0.5}Co_{0.5}O₃ perovskites exhibited very interesting paramagnetic, meta-magnetism and ferromagnetism depending on the size of RE. The determination of valences of Co and Mn and their spin states are important issues in clarifying the magnetism in the REMn_{0.5}Co_{0.5}O₃ system. Very recently, a typical spin glass behavior has been observed in perovskite GdCo_{0.5}Mn_{0.5}O₃ with very sharp transition width of 1K at 115 K and also observed in YbMn_{0.5}Co_{0.5}O₃ and LaMn_{0.5}Co_{0.5}O₃ compounds. In this paper, we presented our studies on the structures, ferromagnetism, spin glass and spin states of Co and Mn in GdCo_{1-x}Mn_xO₃ (x=0.2, 0.3, 0.4, 0.5). We observed spin glass state and two peak features in dc and ac susceptibility. The valences of Co or Mn changed from 2+ or 4+ to 3+ when Co amount increases and possible spin state transformation of Co and Mn was also suggested.

Experimental

Polycrystalline ceramic samples of GdCo_{1-x}Mn_xO₃ (x=0.2, 0.3, 0.4, 0.5) were prepared by solid stat reaction method. The samples structures were characterized radiating X-ray diffraction and the structures are refined using Rietveld refinement program. Results showed that all the samples are single phase and crystallized as orthorhombic structures and the lattice parameters gradually increase with x.

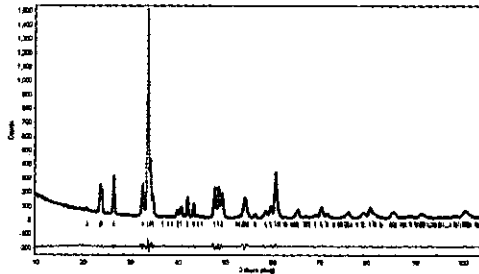


Figure 1: Rietveld Refinement of GdMn_{0.4}Co_{0.6}O₃

Zero-field cooled (ZFC) and field cooled (FC) magnetization and ac susceptibility were measured using commercial PPMS in difference magnetic fields and frequencies over a wide temperature range.

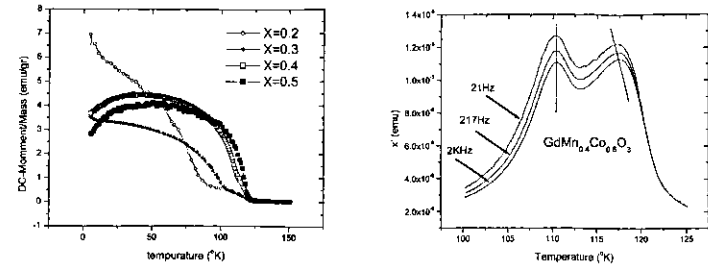


Figure 2: Inverse susceptibility vs. temperature for GdMn_{0.5}Co_{0.5}O₃

straight line is Currie-Wise fitting

All samples show ferromagnetic transition at temperature around 122K. For samples with x>0.5, we found that there is a secondary transition which decrease with x in addition to the first transition at 122K. The secondary transition is also present in ac susceptibility measurement. The first peak shifts to high temperature with frequency indicating a spin glass state, but the second one is independent of frequency.

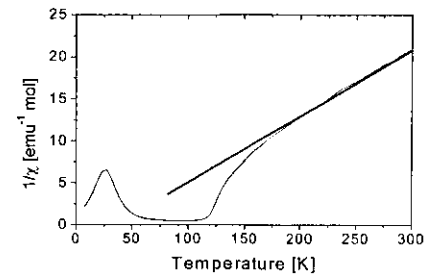


Figure 3: DC-moment of different doping levels of GdMn_{0.5}Co_{0.5}O₃

By fitting to Currie-Wise Law from $1/\chi$ vs. temperature, we found that μ_{eff} obtained gradually decreased from 10 to 8 μ_B/mol as Co increasing from 0.2 to 0.8. By assuming appropriate combination of different valences and spin states of Co and Mn, we found that the valences of Co and Mn take 2+ and 4+ with low spin for Co²⁺ and intermediate spin for Mn⁴⁺ when x=0.5. As x increase, they changed from 2+ and 4+ to 3+ and possible spin state transformation was also suggested.