Structure, Transport and Magnetic properties in $La_xSr_{1-x}Co_xRu_{1-x}O_3$

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Abstract

The structure, transport and magnetic properties of the $La_xSr_{1-x}Co_xRu_{1-x}O_3$ perovskites have been investigated. The magnetization measurements were performed in the temperature range of 5K to 300K. At 50 Oe the Ru rich samples exhibit a temperature induced magnetization reversal which disappears in larger magnetic fields. Magnetization (emu/mole) decreases with increasing x and the compound with x = 0.75 exhibits antiferromagnetic ordering. Resistivity exhibits insulating behavior in the entire temperature range. While for x = 0.75, the resistivity can be fitted to Mott's variable-range hopping behavior, the other compounds show a Coulomb glass behavior typically seen in phase separated compounds.

INTRODUCTION

Perovskite $SrRuO_3$ is a Stoner ferromagnet despite its relatively low (bad metallic) conductivity [1]. The Stoner criterion is satisfied because of high density of states at the Fermi level that critically enhances the electron magnetic susceptibility. Metals that possess high-density of states at the Fermi levels can form giant localized magnetic moments around magnetic impurities that polarize the neighbouring itinerant electrons. The first evidence for this phenomenon in oxides was found in $SrRuO_3$ substituted with $LaFeO_3$, in which Fe^{3+} cations induce an enhanced saturation moment in the host $SrRuO_3$ matrix [2]. By aligning the spin-polarized electron atmospheres around Fe^{3+} under a field, a large negative magnetoresistance (MR) emerges at low temperatures in $Sr_{1-x}La_xRu_{1-x}Fe_xO_3$ semiconductor [2]. The $LaCoO_3$ substituted $SrRuO_3$, despite complications of charge transfer and B-site ordering also exhibits large saturation magnetization and negative magnetoresistance [3]. In order to understand this phenomena of charge transfer and B-site ordering we have carried out detailed investigations on structural, magnetic and transport properties of $La_xSr_{1-x}Co_xRu_{1-x}O_3$ for $0.25 \leq x \leq 0.75$ and the preliminary findings are reported here.

EXPERIMENTAL

Polycrystalline samples of $La_xSr_{1-x}(Co_xRu_{1-x})O_3$ (x = 0.25, 0.4, 0.5, 0.6 and 0.75) were synthesized by solid state reaction method. All samples were deemed to be phase pure, as X-ray diffraction (XRD) data collected on a Rigaku X-ray diffractometer in the 2θ range of 18° to 80° using CuK_α radiation showed no impurity reflections [7]. Magnetization measurements were carried out using a Quantum Design SQUID magnetometer at fields of 50Oe and 1000Oe in the temperature range 5K to 300K. The temperature (80 to 300K) dependence of the electrical resistivity was measured using a standard four probe set up.

RESULTS AND DISCUSSION

The XRD patterns of the two compounds are presented in Figure1. All the samples crystallize in $P2_1/n$ monoclinic structure with steady decrease in cell volume as $LaCoO_3$ is added to $SrRuO_3$ to form solid solution.

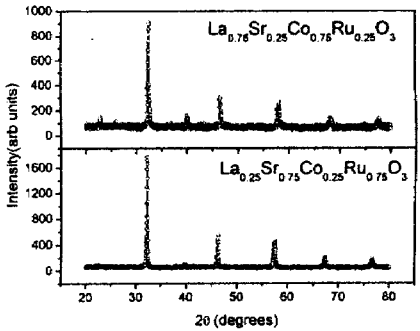

Figure 1: XRD patterns of $La_xSr_{1-x}Co_xRu_{1-x}O_3$ for x = 0.25 and 0.75

Magnetization measurements at 50Oe and 1000Oe during the zero field cooled (ZFC) and field cooled (FC) cycles are presented in Figures 2 and 3. It can be seen that magnetization decreases with increasing x indicating decrease in ferromagnetic moment due to decrease in Ru content. For all the samples except x = 0.75, in a magnetic field of 50Oe, during ZFC cycle, M(T) exhibits a cusp at T_p = 162K and then decreases to zero at T_{comp} =

158 K where magnetic contributions of all sublattices cancel each other. Below T_{comp}, M(T) is negative, indicating that the net magnetic moment is opposite to the external magnetic field. During the FC cycle, magnetization remains positive due to field induced moment reversal.

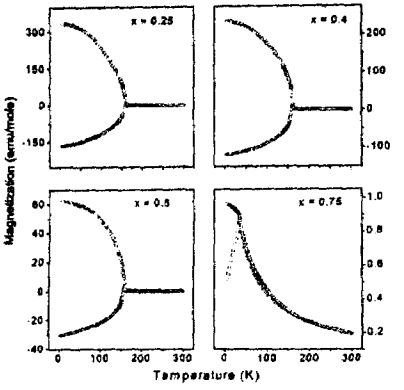

Figure 2: Magnetization versus temperature at 50 Oe in $La_xSr_{1-x}Co_xRu_{1-x}O_3$.

It may be noted that $SrRuO_3$ orders ferromagnetically at about 160K. On the other hand, in a large magnetic field of 1000Oe, the magnetization is positive at all temperatures (see Fig. 3) and its magnitude decreases with increasing La and Co content. The magnetic irreversibility between the ZFC and FC curves indicates complex magnetic behavior. However, this also reduces with increasing Co and La content until x = 0.75 which shows antiferromagnetic behavior with T_N ~ 30K. It is known that $SrRuO_3$ is a metallic ferromagnet while $LaCoO_3$ exhibits spin state transitions accompanied by a transformation from para to diamagnetism. It may be noted that a well ordered sample with x = 0.5 exhibits long range antiferromagnetism [4]. This has been attributed to presence of only two magnetic sublattices, Co^{2+} and Ru^{5+} that couple antiferromagnetically with each other. It is also known that when Co and Ru are neighbours, energetically Co^{2+} and Ru^{5+} states are favoured. When $LaCoO_3$ is doped into $SrRuO_3$, the complex magnetic order exhibited by these solid solution can be understood to be due to a competition between ferromagnetic Ru^{4+}-O-Ru^{4+} networks and the Co^{2+}-O-Ru^{5+} antiferromagnetic networks. This explains the magnetic compensation and decreasing magnitude of magnetization with increasing Co content and the antiferromagnetic ordering in x = 0.75 sample.

The resistivity as a function of temperature for x= 0.25 and x = 0.75 is plotted in fig. 4. The samples exhibit insulating behavior in the entire measured range. With doping of Co for Ru, the resistivity increases significantly. This is due to disruption of metallic Ru^{4+}-O-Ru^{4+} networks by interjection of Co ions. It is also interesting to note that the behavior of resistivity changes from that of a Coulomb glass ($T^{1/2}$) to variable range hopping (VRH) type ($T^{1/4}$) with increase in Co concentration. Usually $T^{1/2}$ behaviour is seen in compounds having electronic phase separation, eg. manganites, etc. A crossover from $T^{1/2}$ behavior to that of VRH type augers well with observed magnetic properties.

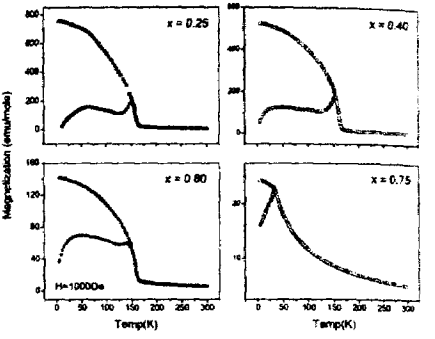

Figure 3: Magnetization versus temperature at 1000 Oe in $La_xSr_{1-x}Co_xRu_{1-x}O_3$.

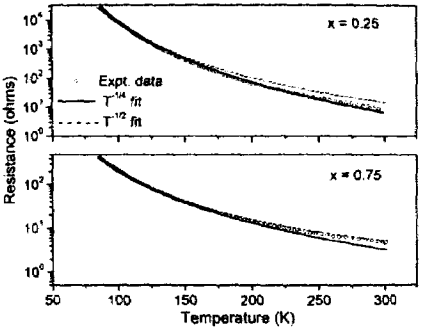

Figure 4: Resistivity versus temperature for $La_xSr_{1-x}Co_xRu_{1-x}O_3$.

CONCLUSIONS

In summary, the structural, transport and magnetic properties in $La_xSr_{1-x}Co_xRu_{1-x}O_6$ are presented. A crossover from magnetic compensated ground state to antiferromagnetic ground state is seen with increase in Co and La concentration. Resistivity also shows a change in behavior from $T^{-1/2}$ to $T^{-1/4}$. This is due to replacement of ferromagnetic Ru^{4+}-O-Ru^{4+} networks by antiferromagnetic Co^{2+}-O-Ru^{5+} networks.

REFERENCES

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