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Magnetotransport Study of Electron Doping in Sr_2FeMoO_6

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Abstract. We report the results from magnetotransport measurements on polycrystalline $Sr_{2-x}La_xFeMoO_6$ samples at magnetic fields of up to 8 T. We observe a normal as well as an anomalous Hall effect. We find that there is a small decrease in the normal Hall coefficient for La doped samples.

Introduction

There has been recent interest in compounds that display a degree of electronic spin polarization because they can lead to new spin transport electronic ("spintronic") devices [1,2] as well as highly sensitive magnetic sensors [1-3]. Sr_2FeMoO_6 (SFMO) is one compound that is predicted to display electronic spin polarization [2] and it also has a Curie temperature that is above room temperature as well as a colossal magnetoresistance [2,4-6]. Band structure calculations predict 100% electronic spin polarization at low temperatures where the Fermi level is within a spin-down band comprising an admixture of $Mo(t_{2g})$ and $Fe(t_{2g})$ orbitals while the up-spin Fe^{3+} $3d^5$ electrons are localized within $Fe(t_{2g})$ and (e_g) orbitals [2]. However, the underlying interactions that lead to half metallicity are not fully understood [7-10]. It is of interest to study the effect of electronic doping on the magnetic and electronic properties of these compounds. For this reason we have performed magnetotransport measurements on electron doped Sr_2FeMoO_6 and report our results in this paper. These measurements were made on $Sr_{2-x}La_xFeMoO_6$ where replacing the divalent Sr ions with trivalent La is expected to result in electron doping [11].

Experimental Details

A series of polycrystalline $Sr_{2-x}La_xFeMoO_6$ (x = 0, 0.2 and 0.4) samples were prepared by a solid-state reaction method from stoichiometric mixes of $Sr(NO_3)_2$, Fe_2O_3, MoO_3, and La_2O_3. The powder was denitrated at 700 °C in air. After pressing the powder at 20 MPa, the pellets were reacted in air at 1200 °C for 8 h. The samples were then ground, pressed into pellets, and then sintered at 1150 °C in an atmosphere of 5% H_2 – 95% N_2 for 3 hours. The last step in the synthesis process was repeated until all the minority phases were removed and homogeneous samples were obtained. The phase composition and purity were determined from X-Ray Diffraction (XRD) measurements using Co K_α radiation. The magnetization measurements were performed using the Vibrating Sample Magnetometer (VSM) option of the Physical Property Measurement System (PPMS 9 T, Quantum Design). The electrical resistances were measured in a four-terminal configuration using the resistivity option of the PPMS. Five-wire Hall effect measurements were done using the AC transport option of the PPMS. In this method, an additional voltage lead is attached in parallel to one of the Hall voltage leads. A 100 Ω potentiometer between these two leads was used to cancel the voltage offset due to the sample resistance.

Results and Analysis

XRD measurements show that all of the $Sr_{2-x}La_xFeMoO_6$ samples are single phase (not shown). Magnetization data under an applied magnetic field of up to 8 T at 50 K are plotted in Fig.1. There is a systematic decrease in the high field magnetization with increasing La-concentration, which can be primarily attributed to Fe-Mo antisite disorder (ASD) that can be estimated from the saturation moment per formulae unit, m_s, according to the equation [12],

$$m_s = 4.0-9*ASD. \quad (1)$$

The Sr_2FeMoO_6, $Sr_{1.8}La_{0.2}FeMoO_6$, and $Sr_{1.6}La_{0.4}FeMoO_6$ samples were found to have ASD values of 15, 25, and 32%, respectively.

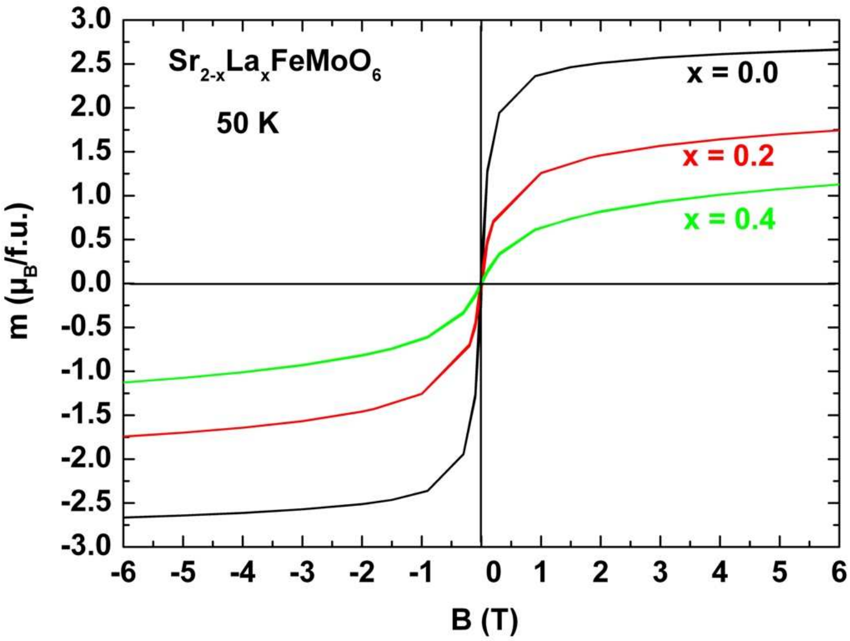

Fig. 1. (Color online) The moment per formulae unit at 50K for the $Sr_{2-x}La_xFeMoO_6$ samples.

The zero-field resistivities as well as the resistivities in a field of 8 T were measured from 300 K down to 5 K. All the samples exhibit an initial decrease in the resistivity with decreasing temperature and then an increase in the resistivity at low temperatures. The resistivity ranges from 1 and 7 mΩcm without any systematic variation with La concentration. The magnetoresistance (MR), which is the change in the resistivity in an applied magnetic field, can be written as,

$$MR = (\rho(H,T)-\rho(0,T))/\rho(0,T), \quad (2)$$

where ρ(H,T) and ρ(0,T) are the electrical resistivities at temperature T, with and without the applied magnetic field H, respectively.

It can be clearly seen in Fig. 2 that all the samples show magnetoresistance where the magnitude increases with decreasing temperature. This large magnetoresistance is attributed to spin dependent tunnelling across grain boundaries [2, 7, 13]. The decrease in the magnetoresistance with increasing La concentration indicates a reduction in the spin polarized current. This is likely to be due to an

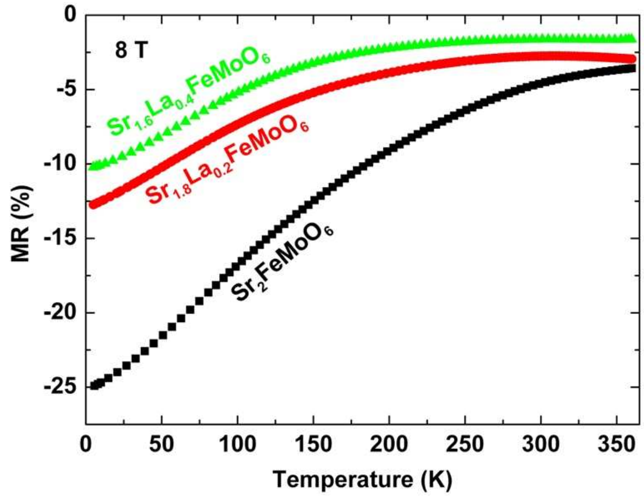

Fig. 2. (Color Online) Magnetoresistance as a function of temperature measured at 8 T for $Sr_{2-x}La_xFeMoO_6$.

increase in disorder near the grain boundaries resulting in spin scattering and not a result of a change in the carrier concentration.

The measured Hall voltages for pure and electron (La) doped SFMO initially increases with increasing magnetic field and then decreases after magnetic moment saturation as shown in Fig. 3. The Hall resistivity for ferromagnetic materials is given by,

$$\rho_{Hall} = R_H B + R_A \mu_0 M, \quad (3)$$

where M is the magnetization, B is the applied magnetic field, μ_0 is the permeability of free space, and R_H and R_A are the normal and anomalous Hall coefficients respectively. The negative slope of the normal Hall coefficient beyond magnetic saturation suggests that the carriers are electron-like.

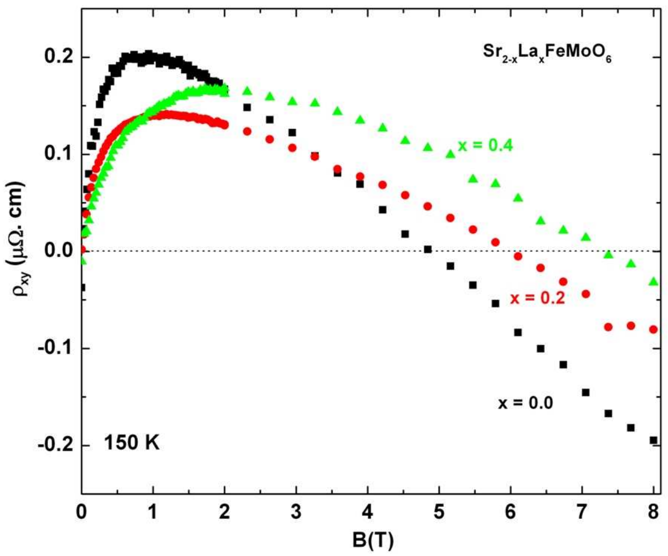

Fig. 3. The magnetic field dependence of the Hall resistivity at 150 K for $Sr_{2-x}La_xFeMoO_6$.

There is a small decrease in the normal Hall coefficient for La doped samples that can be attributed to an increase in the carrier concentration. The negative normal Hall coefficient and the positive anomalous Hall coefficient is in agreement with earlier studies on SFMO single crystals [14, 15] and epitaxial thin films [16].

Conclusions

We have investigated the magnetotransport properties of polycrystalline samples of pure and La-doped SFMO. The magnitude of the magnetoresistance is less after electron doping, which is likely to be due to more antisite disorder in the grain boundary regions. The Hall effect studies show that the Hall resistivity has contributions from normal and anomalous components for all La concentrations. The normal Hall coefficient is observed to decrease in magnitude after partial substitution of Sr by La.

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