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Investigation of electronic phase segregation in $La_{0.75}Ca_{0.15}Sr_{0.10}MnO_3$ manganite

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Abstract. The effect of electronic phase segregation in a broad metal-Insulator transition (MIT) observed in $La_{0.75}Ca_{0.25-x}Sr_xMnO_3$ (x=0.1) composition is investigated using heat capacity, magnetization, electrical resistivity and magnetoresistance measurements. The negative magnetoresistance of 65% in an applied magnetic field of 12T and 15% in 1T with a broad working range of 18K around 300K which is beneficial for room temperature colossal magnetoresistance (CMR) applications. The broad transition in temperature dependent zero field resistivity measurement is analyzed in the light of percolation model indicates the abundance of insulating/metallic clusters in metallic/insulating region. A significant difference between the metallic fraction around the MIT and the ferromagnetic phases observed around the Curie temperature demonstrates the interplay between volume of itinerant and polaronic electronic phases.

1. Introduction

Manganese perovskites are extensively investigated for the past few decades for their colossal magnetoresistance (CMR) property exhibited in a sprawling range of metal to insulator transition temperatures (T_P) with a concomitant magnetic phase transition indicated by Curie temperature (T_C) The colossal magnetoresistance (CMR) observed in manganites around the Curie temperature T_C are attributed to the interplay between spin, lattice, charge and orbital degrees of freedom.[1].CMR property shows a strong dependence on the nature and order of magnetic/electronic phase transitions and it is reported to be controlled by the electronic phase segregation of the system and its relative volume change at T_P [2]. The manganite $La_{0.75}Ca_{0.25}MnO_3$ has a significant colossal magnetoresistance around its curie temperature, and the order of transition in $La_{0.75}Ca_{0.25}MnO_3$ changes from first to second order upon the application of a magnetic field.[3]. In this study an unusually broad phase transition was observed in $La_{0.75}Ca_{0.25-x}Sr_xMnO_3$ (x= 0.10) composition. This composition is investigated using Magnetic, magnetoresistance, heat capacity measurements and analyzed in the light of percolation model is reported in this work.

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2. Experimental details

Polycrystalline powder samples of $La_{0.75}Ca_{0.25-x}Sr_xMnO_3$ (x=0.10) were prepared by the conventional solid-state reaction method. Stoichiometric amounts of La_2O_3, $CaCO_3$, $SrCO_3$ and MnO_2 were mixed well and calcined initially at 1200^O C for 24hrs and then cooled to room temperature. The second calcinations took place at 1250^OC for 24 hrs and cooled back to room temperature. After this second calcination two batches of samples were extracted as LCSM1250 and LCSM1400 whose processing details are reported elsewhere [4].The phase purity and crystal structure of the powder samples were checked by X-ray powder diffraction (XRD) using Cu K_α radiation in a Bruker D8 Advance powder diffractometer, and reported in our earlier work [4]. In brief, both LCSM1250 and LCSM1400 are orthorhombic structured (*pnma* space group), $La_{0.75}Ca_{0.15}Sr_{0.10}MnO_3$ composition, but processed in different sintering temperatures 1250^OC (12hrs) and 1400^OC (24hrs) respectively. The magnetic measurements were taken with fields up to 1T using a Lake Shore 7407 vibrating sample magnetometer. Our heat capacity measurements were made with a custom made differential scanning calorimeter (DSC) [5] and the magnetoresistance measurements were made using Quantum design PPMS upto a magnetic field of 12T and down to a temperature of 2K

3. Results and Analysis

The temperature dependent magnetization in Zerofield cooled (ZFC) and Field cooled (FC) conditions for the sample LCSM1250 and LCSM1400 in an applied magnetic field of 0.01T is shown in figure 1. The maximum change in slope observed in the magnetization *vs* temperature plot at 0.01T is used to determine the Curie temperature (Tc). The Tc value of LCSM1400 is 276 K indicated by sharp dM/dT curve in figure 1 agree very well with the earlier studies of Guo *et al* [6]. A prominent splitting between the ZFC and FC behavior is observed in LCSM1400 upto a Curie temperature of 276K after which it merges into a single curve. In the case of LCSM1250 the splitting is visible upto 295K which

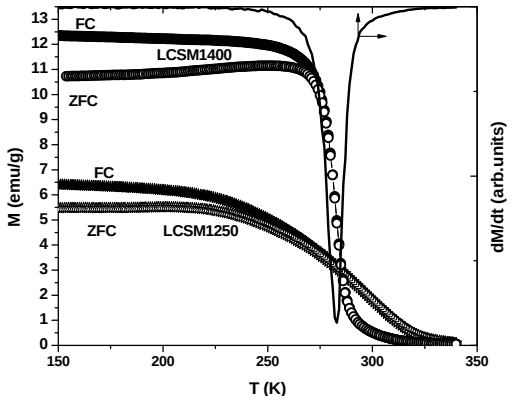

Figure 1. M vs T measurements in LCSM 1400 and LCSM1250 with right axis showing T_C for LCSM1400

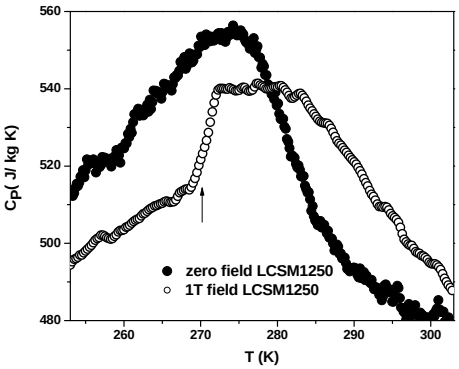

Figure 2. C_P vs T measurements in zero and 1T with arrow pointing the slope change below the curie temperature in LCSM1250 (measured using DSC)

is above the Curie temperature of 287 K. The broad magnetic transition observed in LCSM1250 is very much interesting for room temperature CMR applications and hence the LCSM1250 is further investigated using heat capacity and magnetoresistance measurements. The heat capacity of sample LCSM1250 in zero magnetic fields and in an applied field of 1T is shown in figure 2. The zero field heat capacity of LCSM1250 shows a peak at 275K which indicates the position of the Curie temperature (T_C). There is a significant difference between the T_C observed in magnetization measurements and zero field heat capacity measurement. The spread of transition temperatures in different measurements occurring in LCSM type perovskite manganites has already been well documented by Bahl et al [7]. On application of magnetic field of 1T the transition gets broadened

with an additional slope change below the magnetic transition temperature as indicated by an arrow in figure 2.

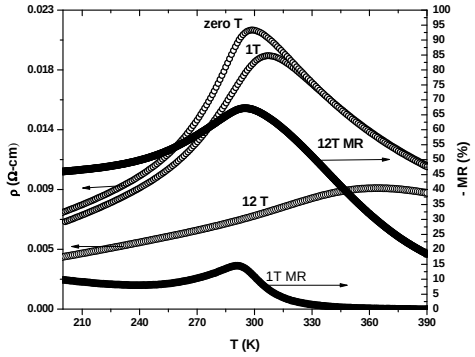

Figure 3. Temperature dependent electrical resistivity with right axis giving the percentage of magnetoresistance in 1T and 12T

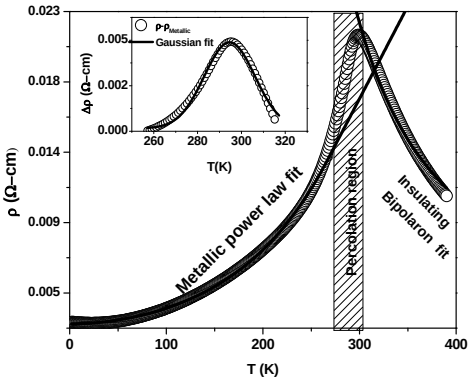

Figure 4. Zero field ρ vs T plot with power law fit for low temperature and bipolaron fit for high temperature, Inset shows MIT with Gaussian fitting

Temperature dependence of electrical resistivity in an applied magnetic field of 0T, 1T and 12T is shown in figure 3. A metal to Insulator transition temperature (T_P) occurring at 299K in the absence of magnetic field and shifts to a higher temperature of 307 K in 1T.The temperature further increases to 350K in an applied magnetic field of 12T. The magnitude of resistance decreases in an applied magnetic field and this percentage of negative magnetoresistance is 15 and 65% in the magnetic field of 1T and 12T respectively. The magnetoresistance is almost constant with a variation of less than 2% for a temperature range of 18K around the MR_{peak} temperature. In the case of LCSM1250 relatively constant magnetoresistance in the range of 286-304K indicates that it is a promising candidate for room temperature CMR applications.

4. Discussions

The nature and electronic properties of this broad MIT transition in LCSM1250 is further investigated by using the temperature dependence of zerofield resistivity as shown in figure 4. The low temperature metallic region below the MIT transition and upto 275 K is fitted using metallic power law with $\rho_m(T)= \rho_0+ \rho_2T^2+ \rho_5T^5$ where ρ_0 is the residual resistivity, ρ_2 and ρ_5 are electron-electron and electron-phonon scattering coefficients respectively. The higher temperature insulating region above the MIT from 305-390K could be fitted using Bipolaron hopping model using $\rho(T)= AT[\exp(\Delta/2k_BT)]$ where A is resistivity constant, Δ is the bipolaronic binding energy and k_B is the Bolzmen constant. The bipolaronic binding energy comes out to be 206 meV which is higher than the Bipolaron binding energy reported in disordered thinfilms with a broad transition.[8].The temperature region of 276-305K around the MIT where itinerant electron model and bipolaron hopping model fails is considered to be the percolation region as indicated by shaded region in figure 4. On extrapolating and subtracting the metallic power law fit,the remaining MIT transition peak is fitted with Gaussian as shown in inset of figure 4 by keeping the width of Gaussian (Γ) as a free parameter. The obtained value of Γ is found to be 27 K.The metallic fractions in the whole temperature range can be estimated from the equation $\gamma(T)=1/2(\text{erfc}([T-T_P]/\Gamma))$ as shown in figure 5. According to the double exchange mechanism proposed for the manganites, MIT as obtained from electrical resistivity measurements usually coincides with the ferromagnetic to paramagnetic transitions obtained from magnetic measurements[8]. A comparison with the metallic fraction obtained in percolation model with that of the ferromagnetic fraction

obtained from M vs T measurements (figure 5) shows a mismatch around MIT. $La_{0.75}Ca_{0.25-x}Sr_xMnO_3$ (x=0.1) composition exist in the close proximity of structural transition from orthorhombic to rhombohedral structure as shown in inset of figure5 . Especially LCSM1250 crystallizing in orthorhombic structure with Jhan-Teller distortion posses inhomogeneous strain and enhanced magnetoelastic coupling [4] when compared to LCSM1400 which dictates the volume of itinerant/ polaronic phases and hence the electronic phase segregation leading to the broadening of MIT.

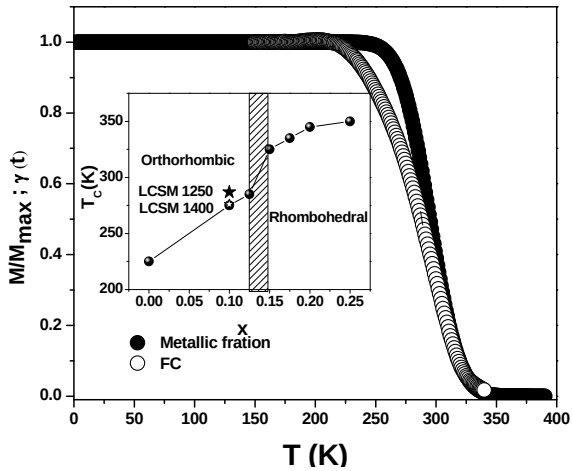

Figure 5. Comparison of metallic fraction with magnetization measurements;(inset) phase diagram of LCSM reproduced from Guo *et* al [6].

5. Conclusion

The broad metal to Insulator transition observed in $La_{0.75}Ca_{0.15}Sr_{0.10}MnO_3$ (LCSM1250) is a promising candidate for room temperature CMR applications. The MIT is analyzed in the light of percolation model. The metallic fraction obtained using electrical resistivity measurements are compared with ferromagnetic phase estimated from magnetic measurements which indicates that the broadening of phase transition can be attributed to the electronic phase segregation (itinerant and polaronic) in and around the MIT.

Acknowledgements

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