

rf SUSCEPTIBILITY OF $La_{1-x}Sr_xMnO_3$ SINGLE CRYSTALS: MAGNETIC SIGNATURES OF STRUCTURAL CHANGES

P.V. PARIMI, H. SRIKANTH, M. BAILLEUL, and S. SRIDHAR,
Department of Physics, Northeastern University, Boston, MA 02115.

R. SURYANARAYANAN, L. PINSARD and A. REVCOLEVSCHI.
Laboratoire de Chimie des Solides, UA 446, Université Paris-Sud, Orsay 91405, France.

ABSTRACT

A sensitive tunnel diode oscillator (TDO) operating at $4MHz$ is used to probe the dynamic response of $La_{1-x}Sr_xMnO_3$ single crystals for $x = 0.125, 0.175, 0.28$ and 0.33 doping. Systematics of the measured change in reactance (ΔX) as a function of temperature ($30K < T < 320K$) and DC magnetic field ($0 < H < 6kOe$) reveal distinct temperature and field scales associated with the dynamic response of spin. It is notable that these features are far more striking than the corresponding features in static measurements. The results are discussed in the context of structural changes leading to polaron ordering.

INTRODUCTION

The perovskite oxides of the form $Re_{1-x}A_xMnO_3$ (where Re is a rare earth such as La and A is a divalent element such as Sr or Ca) have generated considerable interest in recent times because of the discovery of the colossal magnetoresistance (CMR) effect [1]. The CMR is a direct consequence of an unusual paramagnetic insulator (PMI) to ferromagnetic metal (FMM) transition driven mainly by the double exchange mechanism [2]. However, double exchange alone cannot describe the complete phase diagram of the manganites and it has been pointed out that the interplay of strong electron-phonon coupling and double exchange is required to understand the existence of the high temperature insulating phase, the CMR effect and its sensitivity to magnetic field [3][4].

The deficiency of the double exchange model is the fact that it does not consider spin-lattice or charge-lattice interactions, namely, Jahn-Teller interactions and polarons [5]. Experimental results clearly suggest that lattice contributions are important for a thorough understanding of manganites. Besides MI transition, charge ordering (CO) is one of the characteristic phenomena observed in these materials especially in the low doping regime. CO and stripe correlations of concentrated holes and spins have attracted much attention in recent times, particularly due to their possible role in high T_c superconductivity[6, 7, 8].

A variety of experiments including structural [9], transport [10, 11] and thermal [12] measurements have revealed novel features in the $Re_{1-x}A_xMnO_3$ directly associated with the interplay between structural, electronic and magnetic properties. Most of the experiments on manganites have been *static* and there have been relatively few experiments which probe the *dynamic* response of these systems. Dynamic experiments are likely to provide significant information about the collective response of spin and charge to the oscillating electric and magnetic fields impressed on the materials. In the present work rf dynamic response of $La_{1-x}Sr_xMnO_3$ for concentrations $x=0.125, 0.175, 0.28$ and 0.33 are reported. We focus on the interplay of between holes and lattice distortions to understand the relation between the magnetic and structural properties.

EXPERIMENT

Single crystals of $La_{1-x}Sr_xMnO_3$ were grown using an image furnace technique [13]. Samples used in these measurements had cylindrical disk like shapes with diameter $5mm$ and thickness $2mm$ with polished surfaces and edges. The rf experiments were performed using a tunnel diode oscillator (TDO) which has very high sensitivity in measuring the electro- and magneto-dynamic properties of materials. The crystal is placed inside a copper coil which forms part of an LC -tank circuit driven by a stable tunnel diode oscillator. The inductive coil with the sample is mounted at the end of a rigid co-axial cable can be inserted into a continuous flow Helium cryostat. The temperature of this system can be regulated between $4.2K$ and $320K$ and an electromagnet is used to apply a dc magnetic field up to $6kOe$. The resonant frequency (f_0) is typically in the range of $2 - 4MHz$ depending on the geometric characteristics of the inductive coil and sample dimensions. The quantity that is measured, the change in frequency $\Delta f = f(T, H) - f_0$ as a function of T and H , is proportional

to the change in reactance ΔX . For magnetic metals, from elementary considerations and applying Maxwell's equations, it can be shown that: $\Delta X \propto \sqrt{\chi}$, where χ is the differential susceptibility, dM/dH of the material.

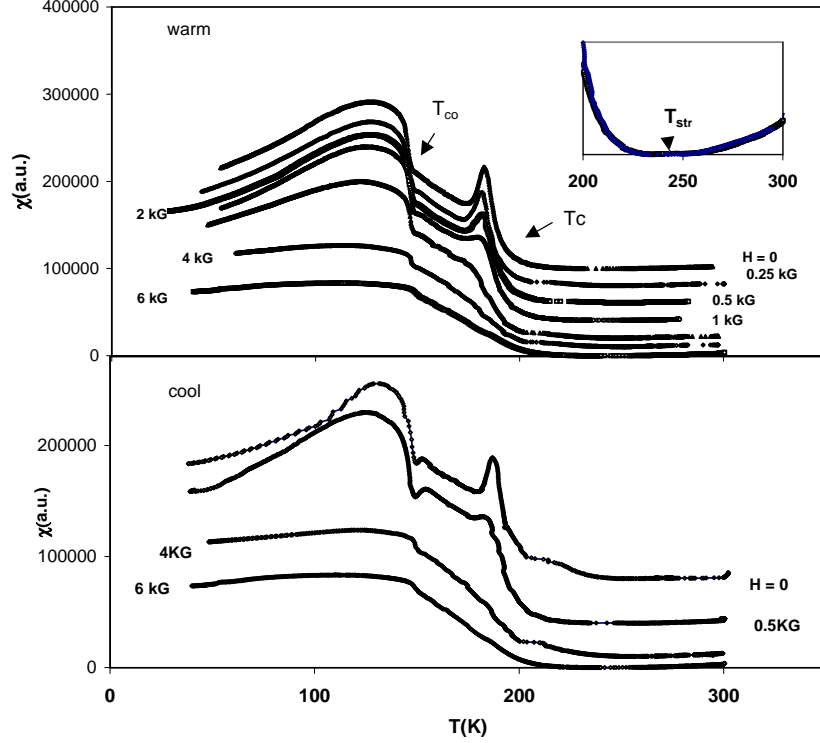


Figure 1: Top panel shows differential susceptibility for $x=0.125$ doping during warming. Bottom panel shows the same during cooling. The first order nature of the CO transition can be seen as a clear difference in the response between warming and cooling. Inset shows a magnified view of the structural transition.

RESULTS

Temperature dependence: $\text{La}_{0.875}\text{Sr}_{0.125}\text{MnO}_3$

The high sensitivity of the rf technique enables us to clearly detect a paramagnetic to ferromagnetic transition at $T_c=180\text{K}$ as well as two additional transitions at $T_s=270\text{K}$ and $T_{co} =150\text{K}$, as shown in Fig. 1. Interestingly, this composition is observed to undergo structural transitions which are manifested in the change of lattice parameters at 150K, 180K and 270K [15]. At T_s the susceptibility shows a dip which is due to a structural phase transition from orthorhombic (pseudo cubic) to a cooperative Jahn-Teller distorted phase at lower temperature. In the presence of a magnetic field two characteristic changes are observed to take place at T_c . First, the peak disappears and secondly, the transition is broadened. In the absence of dc magnetic field the susceptibility is zero above T_c and raises rapidly at T_c . In the presence of dc field the susceptibility is finite at all temperatures and increases with applied field. Therefore, the sharp transition at T_c becomes broadened when field is applied. The hump observed at T_{co} is very clear and strong unlike the CO transition observed in resistivity and magnetization measurements[12]. This fact emphasizes the importance of high frequency measurements to detect CO transitions. The hump at T_{co} is caused by a magnetic transition accompanied by a change in structure[12][15, 9]. We also observed hysteretic behavior in the susceptibility around T_{co} with decreasing and increasing temperature which indicates the first order nature of this transition. As can be seen from the Fig. 1 the rf reactance shows a dip at T_{co} during cooling which is not observed while warming.

The hump associated with T_{co} appears to be a purely ac response of the charge ordering as the dc response[12] does not show any hump. It is worth mentioning that the CO observed in $Nd_{0.45}Ca_{0.55}MnO_3$ at 260K also shows a hump in the ac susceptibility measurement[14]. The reason for the hump only in ac measurement is that in ac measurement the differential susceptibility is measured. The reversible response of the ferromagnetic domains to the rf field just below T_c gives rise to an increase in the differential susceptibility. With further decrease in T the onset of saturation magnetization locks the individual domain and hence the $\chi(T)$ starts decreasing.

The key to understanding the contribution of the structural transitions to the electronic and magnetic properties lies in the Mn-O interionic distance of the octahedra. The interionic distances $m(T)$, $s(T)$, and $l(T)$, which are along a , c and b axes, respectively, are calculated from the representation $m^2 = 0.031(a^2 + b^2 + c^2)$, $s^2 = 0.125c^2 - m^2$ and $l^2 = a^2s^2/(16s^2 - a^2)$. In these calculations the rotation of the octahedra with respect to the axes is neglected. Fig 2 shows the temperature dependence of these parameters. As can be seen from the Fig. $m(T)$ is constant over the entire temperature range, while $s(T)$ and $l(T)$ show clear anomalies at 140K and 270K. These results imply that for temperatures below 140K or above 270K there is no contribution of the rhombic J-T Q2 mode to the formation of crystal lattice. The turning on of the Q2 mode as the sample is warmed above 140K results in structural phase transition from low temperature. The response of a ferromagnet in a magnetic field is also important to describe the first order transitions observed at T_{str} and T_{co} . Below T_{str} the system shows a spontaneous cooperative JT distorted phase[12]. The strong dependence of $\chi(T)$ on magnetic field suggests magnetoelastic coupling for CO besides coulomb repulsion.

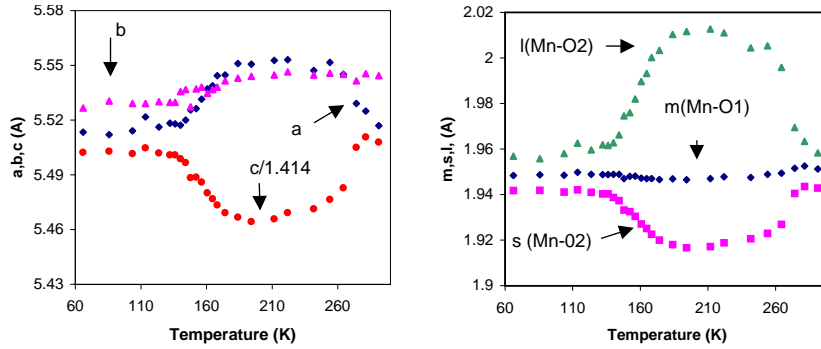


Figure 2: The change in the lattice parameters is shown on the left panel. Right panel shows the Mn-O interionic distances, $m(T)$, $s(T)$ and $l(T)$ which are along a, c and b axes.

An isolated hole in $LaMnO_3$ can be considered a small polaron which is given by a localized hole in the $3d_{x^2-y^2}$ orbital surrounded by inverse Jahn-Teller distortion. The polaron phase is an ordered arrangement of Mn^{3+} and Mn^{4+} ions for which one of the two alternating atomic layers in the (001) plane contains both Mn^{3+} ions, as in pure $LaMnO_3$, while the other layer contains both Mn^{3+} and Mn^{4+} ions, i.e. holes. The local distortion is because the hole site Mn^{4+} is JT inactive whence the electron-phonon energy is lowered by restoring higher symmetry around the hole[16]. In this picture, at high temperatures the $La_{1-x}Sr_xMnO_3$ may be viewed as a polaron liquid which will eventually transform into polaron lattice as the temperature is lowered. We, therefore, identify T_{co} as the onset point for polaron lattice formation, where holes start to freeze on lattice points. From this point of view $La_{1-x}Sr_xMnO_3$ is considered to undergo successive transitions from polaron liquid (insulator) to Fermi liquid (metal) to polaron lattice (insulator).

Field Dependence: $La_{1-x}Sr_xMnO_3$

The field dependence of the differential susceptibility, $\frac{dM}{dh_{ac}}|_{H_{dc}}$, at various temperatures below 300K is shown in Fig. 3. As can be seen from the figure for all the doping levels of Sr the $\chi(H)$ response shows an overall decrease with the increase in magnetic field. The magnetization $M(H)$ shows a monotonic increase with field with an eventual saturation at high fields, for all the compo-

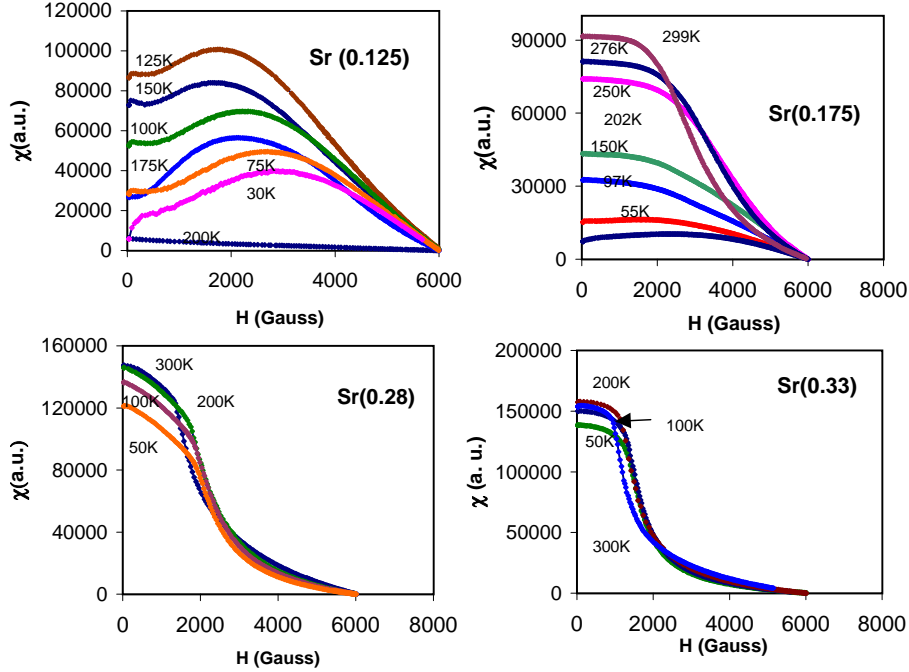


Figure 3: (Left) Field dependence of differential susceptibility for $x=0.125$, 0.175 , 0.28 and 0.33 doping.

sitions studied. Therefore, the decrease in the differential susceptibility is not surprising. There are, however, many subtle changes in the $\chi(H)$ response at low magnetic fields, $H < 2000\text{G}$. In the case of $x=0.125$ composition $\chi(H)$ initially increases, reaches a maximum and starts decreasing forming a peak. For the remaining three compositions the peak is not prominent as can be seen from the figure. The behavior of $\chi(H)$ can be understood by a simple picture of domain response to weak and strong magnetic fields. When a weak field is applied the magnetization process is reversible. In the presence of strong fields the domains are locked and tend to form a single domain. Therefore, the response of the domains to the ac field at low dc fields is greater than that at high dc fields, thus contributing to the initial increase.

CONCLUSIONS

Dynamic rf susceptibility of $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ revealed several magnetic signatures in both temperature and field dependent measurements. These magnetic signatures have direct correlation with structural changes in terms of Mn-O interionic distances of the octahedra, at the corresponding temperatures. Field dependent differential susceptibility is found to decrease monotonically with field with a rich structure at low fields.

ACKNOWLEDGMENT

This work was supported by a US-NSF- 9711910 and NSF-CNRS grant NSF-INT-9726801.

References

- [1] K. Chahara, T. Ohno, M. Kasai and Y. Kozono, Appl. Phys. Lett. 63, 1990 (1993).
- [2] C. Zener, Phys. Rev. 2, 403 (1951).
- [3] A.J.Millis, P.B.Littlewood, and B.I.Shraiman, Phys. Rev. Lett. 74, 5144 (1995)

- [4] H. Roder et al., Phys. Rev. Lett. 76, 1356 (1996).
- [5] J.-S. Zhou, J. B. Goodenough, A. A samitsu, and Y. Tokura, Phys. Rev. Lett. 79, 3234 (1997).
- [6] J. M. Tranquada et al., Nature 375, 561 (1995).
- [7] M. I. Salkola et al., Phys. Rev. Lett. 77, 155 (1996),
- [8] J. Q. Li et al., Phys. Rev. Lett. 82, 2386 (1999).
- [9] H. Kawano, R. Kajimoto, M. Kubota, and H. Yoshizawa, Phys. Rev. B 53, R14709 (1996), H. Kawano, R. Kajimoto, M. Kubota, and H. Yoshizawa, Phys. Rev. B 53, 2202 (1996).
- [10] A. Asamitsu, Y. Moritomo, R. Kumai, Y. Tomioka and Y. Tokura, Phys. Rev. B 54, 1716 (1996),
- [11] A. Anane, et al., J. Mag. Mag. Mater, 165, 377 (1997).
- [12] S. Uhlenbruck, R. Teipen, R. Klinger, B. Buchner, O. Friedt, M. Hucker, H. Kierspel, T. Niemoller, L. Pinsard, A. Revcolevschi, and R. Gross, Phys. Rev. Lett. 82, 185 (1999).
- [13] A. Revcolevschi and D. Dhallene, Adv. Mater., 5, 657 (1993).
- [14] A. A. Mukhin et al., JETP Letters, 68, 356 (1998).
- [15] L. Pinsard et al. J. Alloys Compd. 262-263, 152 (1997).
- [16] Y. Yamada, O. Hino, S. Nohdo, and R. Kanao, Phys. Rev. Lett. 77, 904 (1996).
- [17] A. Anane, C. Dupas, K. Le Lang, J.P. Renard, P. Veillet, A. M. de Leon Guevara, F. Millot, L. Pinsard and A. Revcolevschi, J. Phy: Condensed Matter 7, 7015 (1995).