

## Magnetoresistance in Oxide Compounds: Structural and Electronic transport Properties

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### Abstract

In the recent years, renewed interest in the studies on manganite ceramics and their thin films has been accelerated due to the various potential applications of these materials in bolometers, magnetic field sensors and computer read heads. The discovery of CMR in manganites and its relation to various structural and electronic transport properties rejuvenated research interest in mixed oxide compounds. Since manganites appear promising candidates, both from basic research and applications point of view, a major fraction of materials scientists have contributed to the better understanding of these materials. This article is devoted to the understanding of various physical properties of complex manganite systems and some efforts to evaluate the application potentiality of these compounds.

**Keywords-** Oxides, Double Exchange, Jahn-Teller, Anisotropic Magnetoresistance, Tunneling Magnetoresistance, Giant Magnetoresistance, Colossal Magnetoresistance

### I. INTRODUCTION

Perovskite based mixed oxide and superconducting materials are most attractive research materials in the field of material science and condensed matter physics, in the last decade [1 - 3]. The research on these materials was initially stimulated by the new discovery in the field of high-temperature superconductivity in the late eighties which provided the platform for reconsideration about the various use of mixed oxides for diversity of applications with involving many interesting properties exhibited by them. Amongst numerous perovskite mixed oxide compounds studied and also known as the mixed valence perovskite manganites,  $A_{1-x}B_xMnO_3$  (where A= rare earth, B= alkaline earth) provide a unique opportunity to study the structure-property correlation due to the relationship among charge carriers, charge transport magnetic coupling as well as ordering and temperature dependent structural transition and distortion.

Perovskite type manganites are interesting compounds having exciting structural and manganite properties from both the basic research as well as from technological point of view. As the technology pursued, extensive studies have been made on material synthesis, structural and physical characterization and device fabrication at nano and microscale on these materials [4 - 8].

The mixed valent perovskite based manganese oxides possess rich, complex and interesting physics, chemistry and structural magnetism related to the importance of electron-lattice and electron-electron interactions. The structural, magnetic, electronics and surface properties of these compounds are intricately related and tailored by the synthesis parameters [9 - 13]. In addition, the Colossal Magneto Resistance (CMR) effect exhibited by manganites and the occurrence of ferromagnetic metallic phases with a fully spin-polarized conduction band are hopeful for probable applications. Various technologically important applications like memory devices and magnetic read head from manganites require their high quality epitaxial thin films having well-controlled & tailored physical and mechanical properties. Development

in the epitaxial and amorphous growth of thin films opens a way to all oxide or oxide-metal devices. Magnetoresistance studies on perovskite based oxides done in our previous communications [1-15]. There are so many reports on structural and transport studies of functional oxides by our group [16-23]. Apart from manganite, many other functional oxide materials at nanoscale, synthesized and studied for the various applications point of view [21 - 23].

### II. COLOSSAL MAGNETORESISTANCE

Magnetoresistance (MR) is the modification in electrical resistance of material or thin film on application of external magnetic field. It is defined as

$$MR(\%) = \frac{\rho_{wf} - \rho_f}{\rho_{wf}} \times 100$$

where  $\rho_{wf}$  and  $\rho_f$  are the resistivities at a given temperature in the presence and absence of the magnetic field (H), respectively. MR may be negative or positive depending upon fall or rise in the resistivity, respectively, on the application of external applied magnetic field. The positive MR% is commonly referred as ordinary or normal magneto resistance (OMR). Naturally, the cause of such MR is attributed to the effect of the Lorentz force (due to applied field) on the itinerant electrons causing extra deflection, which increases resistance in material. Such type of MR is dependent on the angle between current direction and orientation of magnetization as well grain structure of material.

The reason for very high negative MR (~50%) in artificially grown multilayers of antiferromagnetically coupled metallic layers of Fe/Cr and Cu/Co by Baibich et al. resulted in boosting of the research on synthesizing new materials with higher negative MR having applications in magnetic read heads, bolometer, etc [24, 25]. High magnetoresistance was reported by J. H. Markna et al. in the LPSMO based hetero structure. In that, scattering due to additional insulating  $Al_2O_3$  nano particles were shown as the main source for it and could be controlled by application of

external magnetic field, resulting in to high magnetoresistance [2].

Depending on the nature and origin of the MR exhibited by different types of materials and thin film, it can be classified as follows:

### 2.1. Anisotropic Magnetoresistance (AMR)

In this type of magnetoresistance, change in electrical resistance in substance is related to the change in the magnetization of ferromagnetic film layer. The AMR can be expressed as

$$AMR = \frac{R_p - R_{pp}}{R_p}$$

Where,  $R_p$  and  $R_{pp}$  are electrical resistances with the magnetic field applied to parallel and perpendicular schematic to the sample, respectively. In the case of AMR, the electrical resistance is anisotropic and be governed by orientation of the magnetization with respect to the current. The origin of AMR effect lies in spin-orbit coupling and the splitting of d-bands in ferromagnetic metals. C. Wang et al. reported the point-contact measurements of AMR (Anisotropic Magneto Resistance in a single crystal antiferromagnetic material and shown the negative magneto transport properties in above material at nanoscale [26].

### 2.2. Giant Magnetoresistance (GMR)

Large magnetoresistance, mentioned to as giant magneto resistance (GMR), was first observed, on the application of external magnetic field to atomically engineered ferromagnetic super lattice structure. This form of magnetoresistance was observed in metal multilayers such as Fe/Cr, Co/Cu [24 - 27]. The magnetic field enhances the conduction of electrons resulting in MR  $\sim$  50%. Owing to large amount of MR, name giant magnetoresistance was given to emphasize the strength of MR in these materials. In the case of GMR, electrical resistance is dependent on the direction of the applied magnetization because electrons have different scattering prospects in the presence of parallel and anti- parallel magnetization directions. The GMR is usually defined as

$$GMR = \frac{R_p - R_{AP}}{R_{AP}}$$

where,  $R_{AP}$  and  $R_p$  are resistances of the stack of layers with anti-parallel (AP) and parallel (P) state of the magnetizations, respectively.

### 2.3. Tunneling Magnetoresistance (TMR)

Tunneling magnetoresistance phenomenon is observed in the tunneling junction of the two ferromagnetic layers which are separated by an insulating barrier. Similar to other MR effect, in the TMR, considerable change in resistance is observed when the relative orientation of the two magnetic film layers change from anti-parallel to parallel. The relative change in the resistance, TMR, is quantified by the equation

$$TMR = \frac{I_p - I_{AP}}{I_{AP}}$$

where,  $I_p$  is the current when both the ferromagnetic layers have parallel magnetizations while  $I_{AP}$  is the current when

one ferromagnetic layer has a parallel magnetization and another layer has anti-parallel magnetization.

MJTS (Magnetically engineered magnetic tunnel junctions) was fabricated and show promise as non-volatile storage cells with high-performance Magnetic Random Access Memories (MRAM) [28]. At room temperature, such devices show the modest performance  $\sim$ 70% based on phenomenon of tunneling magnetoresistance (TMR). With Single crystal substrate, theoretically, it also showed the possibility of higher TMR values [28].

### 2.4. Colossal Magnetoresistance (CMR)

Recently, MR as large as  $\sim$ 100% was observed in manganites. This was an extraordinarily large magneto resistance as compared to formerly reported GMR. Therefore, to highlight the strength of MR in these materials, a new term called "Colossal Magneto Resistance – CMR" was given and studied [1]. In the perovskite based materials, origin of MR in manganites is quite different than the origin of other forms of MR discussed above. The CMR is an intrinsic property related to crystal structure and has its origin in the spin disorder of conduction electrons, which can be suppressed by the application of the magnetic field resulting in large magneto resistance [1, 29-31]. The CMR can be determined from the ratio of a change in the electrical resistivity under the magnetic field, which can calculated as,

$$CMR = \frac{\rho_0 - \rho_H}{\rho_0} \times 100$$

## III. STRUCTURAL AND ELECTRONIC TRANSPROT PROPERTIES

### 3.1.1 Structural Properties

Structure of the  $A_{1-x}B_xMnO_3$  oxides is close to that of the cubic perovskite (Figure 1). The large sized A-trivalent ions and B-divalent ions with 12 - fold oxygen crystallographic co-ordination. The smaller Mn ions in the  $Mn^{+3} - Mn^{+4}$  mixed valent states are located at the centre of an oxygen octahedron i.e. at B-site with 6-fold crystallographic coordination. For the stoichiometric perovskite manganite based oxide, the proportions of Mn ions in +3 & +4 valence states are  $1-x$  and  $x$ , respectively.

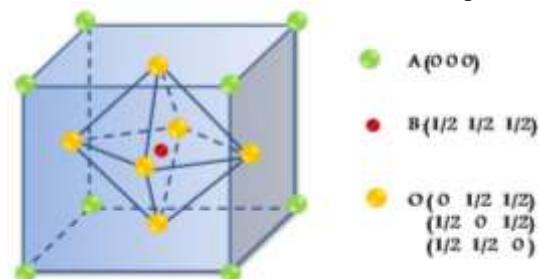


Figure 1: Schematic view of the cubic perovskite structure, Crystallographic structure of  $LaMnO_3$ .

The structure of the manganite is governed by the tolerance factor  $t = (r_A + r_O)/\sqrt{2} (r_B + r_O)$ . Manganite based perovskite structures are more stable in the range  $0.89 < t < 1.02$ ,  $t = 1$ , corresponding to the perfect cubic closely packed crystal structure [1]. Generally, it varies significantly from 1 and manganites have, at least, at low temperature, a lower rhombohedral crystallographic symmetry or orthorhombic structure. This is illustrated by the

orthorhombic structure of  $\text{LaMnO}_3$  (Figure 1), which is the parental structure of most of the magnetoresistive manganite materials studied for potential application and devices by partial substitution of La by Ca and Sr. For an isolated 3d ion in the manganite material, five degenerate orbital states are available to the 3d electrons with  $l = 2$ . In a single crystal, the degeneracy is partly lifted by crystal field. Five d-orbitals split by a cubic crystal field into three  $t_{2g}$  orbitals and two  $e_g$  orbitals. For the  $\text{MnO}_6$  octahedron, the splitting between the lowest  $t_{2g}$  level and the highest  $e_g$  level is  $\Delta \sim 1.5$  eV (Figure 2). For the  $\text{Mn}^{3+}$  and  $\text{Mn}^{4+}$  ions, the interatomic correlations ensure parallel alignment of electron spins having corresponding exchange energy of about 2.5 eV larger than the crystal field splitting  $\Delta$ .  $\text{Mn}^{3+}$  is  $3d^4$ ,  $t_{2g3}\uparrow e_g\uparrow$  with  $S = 2$  whereas  $\text{Mn}^{4+}$  is  $3d^3$ ,  $t_{2g3}\uparrow$  with  $S = 3/2$ . Their respective magnetic moments are  $4\mu_B$  and  $3\mu_B$ , give very small orbital contribution in the structural modifications.

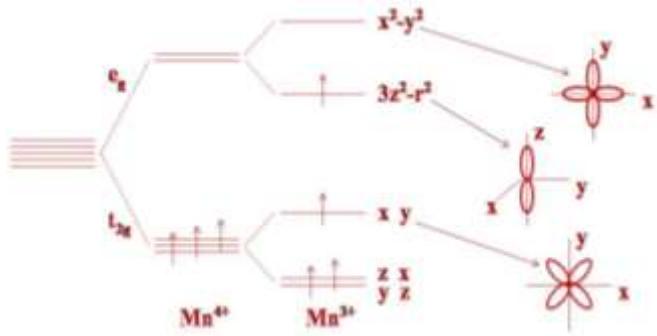


Figure 2: Energy levels and orbitals of  $\text{Mn}^{4+}$  and  $\text{Mn}^{3+}$  in a crystal field of octahedral symmetry and with axial elongation.

In a crystal field of symmetry lower than cubic, the degeneracy of the  $e_g$  and  $t_{2g}$  levels is lifted, as shown in the figure 2 for an axial elongation of oxygen octahedron in the structure. Although, the energy of  $\text{Mn}^{4+}$  remains unaffected by above mention distortion, the energy level of  $\text{Mn}^{3+}$  is lowered. Due to that,  $\text{Mn}^{3+}$  has a noticeable tendency to distort its surrounding octahedral structural environment in contrast to  $\text{Mn}^{4+}$ . This Jahn – Teller distortion is rather effective in the lightly doped manganites, i.e., with a large concentration,  $1-x$ , of  $\text{Mn}^{3+}$  ions; the Jahn-Teller distortions are not independent from one  $\text{Mn}^{3+}$  site to another (cooperative Jahn-Teller effect). It was also reported that, the structure of  $\text{LaMnO}_3$  (Figure 1) in which the  $\text{MnO}_6$  octahedra is strongly elongated within various crystallographic plane in a regular way resulting in to a doubling of the unit cell. On increasing the  $\text{Mn}^{4+}$  content in material, the Jahn-Teller distortions are decrease and the equilibrium of the  $3z^2-r^2$   $e_g$  orbital becomes less effective [1]. Nevertheless, in a large number of manganites, the  $e_g$  orbitals of two types,  $3z^2-r^2$  and  $x^2-y^2$  are not occupied by the  $e_g$  electrons of  $\text{Mn}^{3+}$  at random and an orbital order is achieved.

### 3.1.2 Double Exchange

An unusual and exciting case is that of  $\text{Mn}^{3+}-\text{O}-\text{Mn}^{4+}$  for which Mn ions can exchange their valence by a simultaneous hop of the  $e_g$  electron of  $\text{Mn}^{3+}$  on  $\text{O}$ -orbital and from  $\text{O}$ -orbital to the empty  $e_g$  orbital of  $\text{Mn}^{4+}$ . This mechanism of DE initially proposed by Zener ensures a

strong ferromagnetic-type interaction [32]. As shown by Anderson and Hasegawa, the probability of the  $e_g$  electron transfer from  $\text{Mn}^{3+}$  to neighboring  $\text{Mn}^{4+}$  is to  $\text{Cos}(\theta/2)$ ,  $\theta$  being the angle between Mn spins, in the case of strong Hunds coupling (Figure 3) [33]. The process of electron transfer lifts the degeneracy of the configurations  $\text{Mn}^{+3} - \text{O} - \text{Mn}^{+4}$  and  $\text{Mn}^{+4} - \text{O} - \text{Mn}^{+3}$  leading to two energy levels  $E_l=0 \pm \text{to Cos}(\theta/2)$ . The energy gain of the parallel spin configurations to  $\theta = 0$ , which maximizes  $t$ , with respect to the anti-parallel one  $\theta = \pi$ , reveals the ferromagnetic character of DE interaction. However in DE, angular dependence of  $\text{Cos}(\theta/2)$  is quite different from  $\text{Cos}(\theta)$  of usual exchange interaction. This different angular dependence in conjunction with the competition between double exchange through ferromagnetism and super-exchange anti-ferromagnetism is at the origin of the complex magnetic phase diagram of manganites against the A-site doping level,  $x$ . In particular, canted antiferromagnetic phases at small  $x$  were predicted early on by P. G. de Gennes [34].

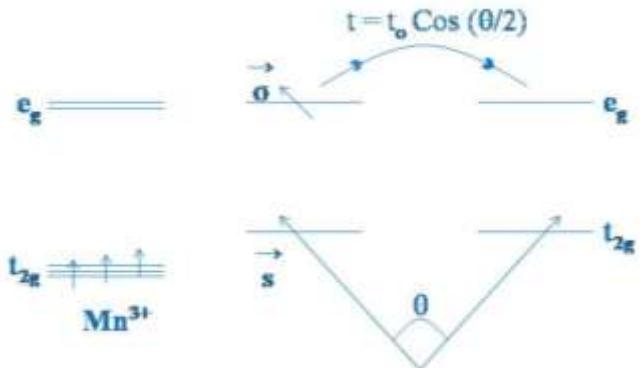


Figure 3: Schematic view of double exchange (DE) mechanism.

### 3.2.1 Electronic Transport

The simple manganites, such as  $\text{La}_{1-x}\text{M}_x\text{MnO}_3$  ( $\text{M} = \text{Ca}, \text{Sr}, \text{Ba}$ ) with  $x = 1/3$  with strong DE exhibits a transition from high temperature paramagnetic (PM) semi-conducting or insulating (I) phase to a low temperature FM phase.

### 3.2.2 Paramagnetic Region

In the P phase, the electrical resistivity generally exhibits strong temperature dependence. Different  $\rho(T)$  laws have been used to fit experimental data, the most popular ones being: (i) simple thermal activation law  $\rho = \rho_\infty \exp(E_0/k_B T)$  with a typical gap value of  $\sim 0.1$  eV; (ii) hopping of adiabatic polarons  $\rho \sim T \exp(E_0/k_B T)$ ; (iii) Mott variable range-hopping (VRH),  $\rho = \rho_\infty \exp[(T_0/T)^{1/4}]$ . Every law has some physical origins, which are respectively (i) the existence of a pseudo gap at the Fermi level in the P phase; (ii) the local lattice distortion accompanying the moving charge carrier (Jahn-Teller polarons); (iii) the localization of the charge carriers by the magnetic disorder [35, 36]. In a narrow range of temperature, it is practically impossible to discriminate between these different  $\rho(T)$  laws.

### 3.2.3 Ferromagnetic Region

At low temperature, below the Curie temperature,  $T_C$ , the spontaneous alignment of Mn spins allows the

delocalization of the  $e_g$  electrons, leading to a low resistive FM phase with  $\rho \approx \rho_0 + aT_2$  for  $T \leq T_C$ . This alignment of Mn spins can be induced for  $T \geq T_C$ , or reinforced for  $T \leq T_C$ , by applying an external magnetic field. The maximum effect is obtained close to  $T_C$  since the initial magnetic susceptibility diverges as  $T \rightarrow T_C$ . Thus, these manganites have a rather huge negative magnetoresistance, the so – called CMR, which peaks at about  $T_C$ . This phenomenon is observed in large number of manganites. In general, since the resistivity of the P phase strongly increases on decreasing T, whereas that of the FM phases decreases, the CMR becomes larger as  $T_C$  becomes smaller at least for a given doping level.

### 3.2.4 Effects of doping level and A-site cation size

Two main parameters determine physical properties of perovskite-type manganites: the doping level,  $x = Mn^{4+}/(Mn^{3+} + Mn^{4+})$ , and the average size of the cation A,  $\langle r_A \rangle$ . A third relevant parameter is the degree of disorder at site A, defined by  $\sigma^2 = \sum x_i r_i^2 - \langle r_A \rangle^2$ . A huge number of experimental studies have been performed to quantify these effects [30]. In general, ferromagnetic DE is maximum around  $x=1/3$  and for  $\langle r_A \rangle \approx 1.24 \text{ \AA}$ . The reduction of  $\langle r_A \rangle$  from this optimum value leads to a rising distortion of crystallographic structure. The resulting decrease of the Mn – O - Mn angle from  $180^\circ$  to a smaller value weakens the ferromagnetic DE and increases the tendency to localize the charge carriers [33, 34].

## III. CONCLUSION

In the research area in materials science and condensed matter physics, perovskite based manganites are the attractive class of materials with rich scientific aspect and potential applications. The magnetoresistance in the above manganite materials is truly fascinating and shows its use in many potential applications. In the present communication, different types of magnetoresistance in functional oxides are discussed in the vicinity of structural and electronic transport.

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