

Correlation of Magnetoresistance and Thermoelectric Power in $\text{La}_{1-x}\text{Li}_x\text{MnO}_y$ Compounds

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ABSTRACT

The temperature dependences of the thermoelectric power, TEP, (S) and magnetoresistance (MR) effect of $\text{La}_{1-x}\text{Li}_x\text{MnO}_y$ ($x = 0.05, 0.1, 0.15, 0.2$ & 0.25 at %) fixed valence doped compounds were studied between 80K and 320K. X-ray powder diffraction (XRD) showed that the samples are single phase. We found the correlation of structures to the magnetoresistance (MR) and the thermoelectric power (S) which we assigned to change the specific branches of the magnon/phonon spectra with x . We also observed a splitting of the Curie and the metal insulating temperature $T_C > T_{ms}$.

Keywords: Thermoelectric Power, Magnetoresistance, the Curie Temperature

1. Introduction

Recently, the rediscovery of colossal magnetoresistance (CMR) in perovskite manganese oxides $\text{R}_{1-x}\text{A}_x\text{MnO}_3$ (where R is a trivalent rare-earth element and A is a divalent element such as Ca, Sr, Ba, or Pb) has generated a considerable interest because of their various electronic, magnetic and structural properties and potential applications [1-4]. It has long been thought that the magnetic structure and electronic transport properties of $\text{R}_{1-x}\text{A}_x\text{MnO}_3$ are correlated via the double-exchange (DE) mechanism [5], *i.e.* the hopping of e_g electrons between Mn^{3+} and Mn^{4+} ions mediated by oxygen anions. However, recent studies have proved that the strong Jahn–Teller type electron–phonon coupling [6], orbital degree of freedom [7,8], cation size mismatch [9-11], oxygen non-stoichiometry [12-14], etc. can strongly influence the behavior of these types of systems under the external conditions such as temperature, magnetic field, electric fields etc. In case of doping the compound $\text{La}^{+3}_{1-x}\text{A}^{+1}_x\text{MnO}_3$ by monovalent alkali (A) atoms like Li^{1+} , Na^{1+} , Rb^{1+} because of larger valence difference between La^{+3} and alkali-metal ions, fewer alkali-metal ions are required to obtain a specific carrier concentration compared to divalent doping and a consequent increase in conductivity is achieved with less inhomogeneity [15-19].

In general, the thermal variation of the electrical resistivity for both divalent and monovalent lanthanum manganites are known to be dominated by polaronic transport for $T > T_C$ [15,20], while below T_C , electron–electron and electron–magnon interactions are usually believed to dominate the conductivity [21-23]. Seebeck coefficient has also been subject of several studies [15,21,24-26], particularly in divalent-doped manganites. In the high temperature ($T > T_C$), most of the reports on thermoelectric power suggested a small polaron hopping [27,28], while, below T_C in the ferromagnetic region obtained from the TEP is governed by the coexistence of both phonon drag and magnon drag effects [26].

2. Experimental

The polycrystalline $\text{La}_{1-x}\text{Li}_x\text{MnO}_y$ samples ($x = 0.05, 0.1, 0.15, 0.2$ & 0.25 at %) have been prepared by standard solid state reaction method. Stoichiometric amounts of La_2O_3 , Li_2CO_3 and MnCO_3 powders (all having 99.99% purity) were thoroughly mixed and ground. After grinding, the powders were pressed into pellets with a pressure of 2 tones cm^{-2} and calcined at 1273K for 15 h in air. Followed by cooling to room temperature, they were re-ground and again pressed into pellets with a pressure of 7 tones cm^{-2} and subsequently annealed at 1373K for 10 h. The samples were examined by X-ray powder diffraction

analysis which indicated the presence of single phase with perovskite-type structure. The XRD analysis was performed using Bruker (Axs-D8Advance) diffractometer at room temperature with CuK_α radiation ($\lambda = 1.5406 \text{ \AA}$). Resistivity measurements were performed in a variable temperature liquid nitrogen cryostat. The electrical resistivity was measured by using the standard four-probe technique using an air drying conducting silver paste. The sample temperature was monitored by calibrated Pt-100 thermocouple in the range 80–320 K. The temperature accuracy was 0.5 K. A constant current in the range of 10 mA–100 mA was supplied by the current source [Delta G/electronic 0-20, 0-100 mA] and voltage across the sample was measured with a digital voltmeter.

The magnetoresistance (MR) ratio is defined by $\text{MR} = \delta\rho/\rho_o = (\rho_H - \rho_o)/\rho_o$, where ρ_H and ρ_o are resistivities with and without vertical applied magnetic field (0.5 T) respectively using [GMW magnet system model 347-70 G1 electromagnet 0.6 T]. The thermoelectric power measurements were carried out using the sample two-heater method with copper electrodes [29-31]. The difference in temperature between the two opposite surfaces of the sample was adjusted to be equal to 3K during the entire measurement. The temperature accuracy of the measurements is described above. The thermoelectric voltage was measured by a digital voltmeter with sensitivity 1 mV as described in Reference [29-31].

3. Results and Discussion

Figure 1 shows powder X-ray diffraction patterns of the polycrystalline $\text{La}_{1-x}\text{Li}_x\text{MnO}_y$ samples ($x = 0.05, 0.1, 0.15, 0.2$ & 0.25 at %). The XRD patterns of samples reveals that the prepared samples have a single-phase rhombohedral structure with a space group $R\bar{3}c$. A remarkable fact from the comparison of these patterns is that the systematic substitution of La by Li does not produce relevant effect on them. In general, all the peaks of five samples satisfy the La-Li-Mn-O phase. The structural parameters are refined by the standard Rietveld technique [32]. The lattice volume, lattice distortion and the bond lengths (Mn-O) decrease with increasing Li substitution [33]. It is well known that one of the possible origins of the lattice distortion of perovskite-based structures is the deformation of the MnO_6 octahedra originating from Jahn-Teller (JT) effect that is inherent to the high-spin ($S = 2$) Mn^{3+} ions with double degeneracy of the e_g orbital. Obviously, this kind of distortion is directly related to the concentration of Mn^{3+} ions.

Another possible origin of the lattice distortion is the average ionic of the A-site element, which is governed by the tolerance factor $t = (r_A + r_o) / \sqrt{r_B + r_o}$, where r_A, r_B and r_o radius of A cation, B cation and oxygen element. As t is close to 1, the cubic perovskite structure is real-

ized. As r_A decreases, so does t , and the lattice structure transforms to the rhombohedral ($0.96 < t < 1$) and then to the orthorhombic structure ($t < 0.96$), in which the bending of B-O-B bond increase and the bond angle deviates from 180° . For LaLiMnO_3 samples, we think that the room temperature structural transformation originates mainly from the variation of the tolerance factor t induced by the partial substitution of smaller Li^{1+} ($r = 0.76 \text{ \AA}$) ions for large La^{3+} ions ($r = 1.032 \text{ \AA}$). The reverse lattice distortion from orthorhombic to rhombohedral symmetry due to larger Sr^{3+} ions partially substituting for La^{3+} ions has been observed $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ compounds [34,35].

The resistivity versus temperature shows a semiconducting behavior above the metal-semiconductor transition temperature (T_{ms}) for all cases as shown in **Figure 2**. An applied magnetic field of 0.5 T suppresses the resistivity, giving a large negative magnetoresistance. It is observed that above T_{ms} the resistivity is much dependent of T and it changes from fraction of $\Omega\text{-cm}$ to 500 $\Omega\text{-cm}$ for samples rich with Li content. Therefore, it is observed that the values of MR are values close along the range temperature. In other side, the widening of transition peak narrow with increasing Li content [36].

The negative magnetoresistance MR% defined as: $\text{MR} = (\rho_H - \rho_o) / \rho_o$ was calculated and plotted versus ambient temperature as in **Figure 3**. In general, we observed that the value of MR increase with decreasing Li content. For $x = 0.15$ sample, a MR ratio as high as fifty percentage is obtained around T_c in magnetic field of 0.5 T.

The magneto-resistance MR of ceramic $\text{La}_{1-x}\text{Li}_x\text{MnO}_y$ can be viewed as a superposition of a continuously decreasing function which is connected with spin-dependent transfer at grain boundaries and an intrinsic MR-peak

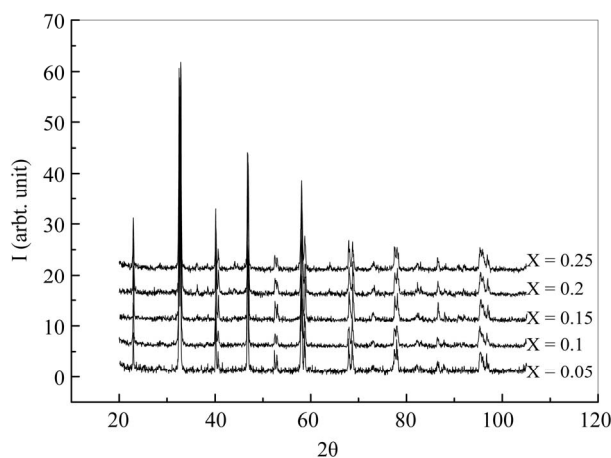


Figure 1. X-ray diffraction patterns of $\text{La}_{1-x}\text{Li}_x\text{MnO}_y$ ($x = 0.05, 0.1, 0.15, 0.2$ & 0.25 at %). All patterns of samples reveals that the samples have a single-phase rhombohedral structure with a space group $R\bar{3}c$.

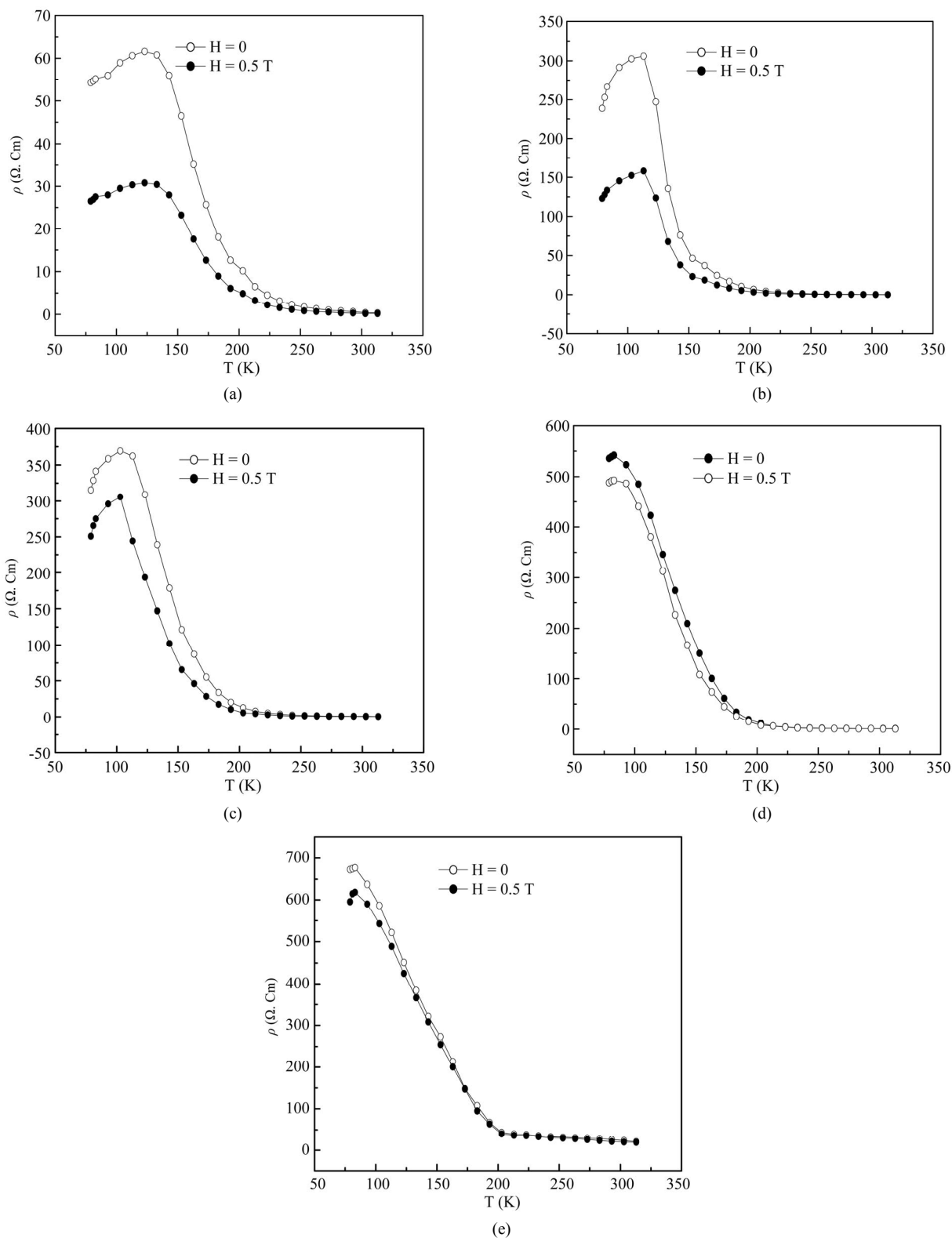


Figure 2. The variation of resistivity in zero magnetic field and $H = 0.5$ T with temperature, T (K), for Li-content of $\text{La}_{1-x}\text{Li}_x\text{MnO}_y$ = where a - $x = 0.05$, b - $x = 0.1$, c - $x = 0.15$, d - $x = 0.2$ and e - $x = 0.25$ at %. The resistivity shows a semiconducting behaviour above the metal–semiconductor transition temperature (T_m) for all cases. The semiconductor–metal transition (T_m), moves to a lower temperatures with the increment of the Li concentration.

at T_c [37]. The superposition was observed for all the five composition (i-MR) as in **Figure 3**. That means that there are additional structures in the i-MR. This superposition corresponds to the first peak of $S(T)$ in **Figures 3,4**. In other side we observe that while the values of $S(T)$ increase with decreasing Li-content the value of MR decrease. As shown in **Figure 4** there are two transitions for each composition. The first peak has a value of T_c corresponds to that value obtained from the magnetoresistance $\delta\rho/\rho_o(T)$ curve [38] can be identified with the (highest) peak of the (bulk quantity) i-MR (at T_c). The second peak of $S(T)$ corresponds to the m-s transition which determined of ρ_o as shown in from **Figure 2**. This observation shows that there are a good correlation between MR and TEP.

However, both the value of Seebeck coefficient (S) and the value of resistivity (ρ) increase with increasing doping of Lithium as in **Figures 2,4**. We expect that when the Li-content increase not only the La-content decreases but also the charge carrier density does [38]. Therefore, the La/Li mixture ions play a prominent role in controlling the resistivity.

However, the Li-doping can also have beneficial effects in regard to the magnetoresistance: 1) we find that MR increase with decreasing Li content and 2) the low Li-substitution tends to broaden the magnetoresistive peak around T_c (**Figure 2**); both effects are helpful if one thinks about the robust magnetoresistive sensors. In an effort to understand this behavior we found an unexpected correlation of structures in MR and the thermoelectric power TEP results; which gives us an important clue to the origin of this broaden magnetoresistive and the fine structure which agrees with it.

The values of metal-semiconductor transition temperature (T_{m-s}) and the ferromagnetic transition temperature (T_c) tabulated in **Table 1**. Generally the value of both T_c and T_{ms} which are determined from Seebeck or resistivity measurements decreases with increasing Li content. In addition, the values of $T_c(S)$ are higher than $T_c(MR)$ and also the values of $T_{ms}(S)$ are equal or higher than $T_{ms}(\rho)$. We believe that the lithium alters the $\text{Mn}^{4+}/\text{Mn}^{3+}$ ratio which is one of the factors which determine the transport and magnetic properties of samples. The decrease in T_{ms} with increasing of Li-content can be interpreted due to an increasing strength of the Mn–O–Mn bond with decreasing the average radius average $\langle r_A \rangle$ of ions of A-sites because of the partial substitution of smaller Li^{1+} ions for larger La^{3+} ions. This substitution causes a narrowing of the bandwidth, thus, decreasing of, e_g electrons which, in turn, results in a weakening of the double exchange interaction magnetism [9].

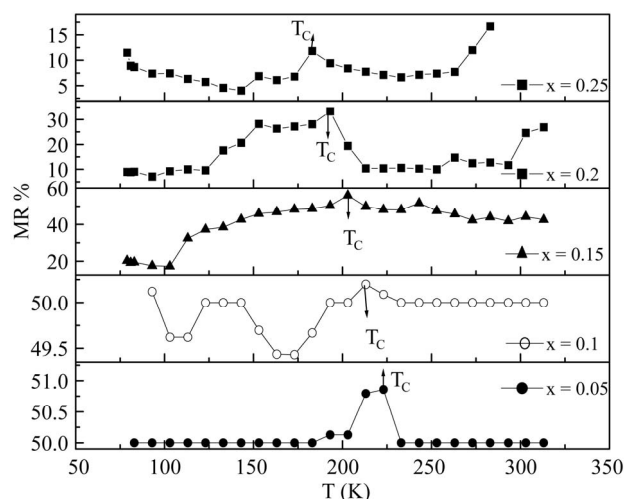


Figure 3. The variation of magnetoresistance with temperature, $T(K)$, for Li-content of $\text{La}_{1-x}\text{Li}_x\text{MnO}_y$. The value of MR increase with decreasing Li content. The MR ratio of the $\text{La}_{1-x}\text{Li}_x\text{MnO}_y$ reaches to 55% under a low field ($H = 0.5$ Tesla) for low doping Li-content.

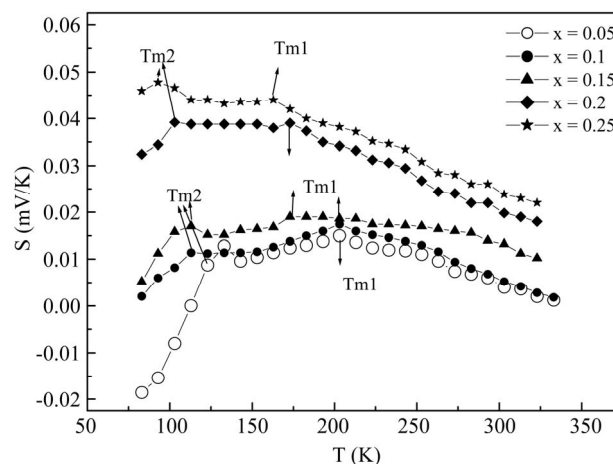


Figure 4. The variation of Seebeck coefficient with temperature, $T(K)$, for Li-content of $\text{La}_{1-x}\text{Li}_x\text{MnO}_y$. The values of $S(T)$ increase with increasing Li concentration. T_{m1} corresponds to the Curie temperature but T_{m2} corresponds to m-s transition temperature.

Table 1. Curie and M-S transition temperature as function of doping, x .

	$T_c(M)(K)$	$T_c(S)$	$T_c(MR)$	$T_{ms}(S)$	$T_{ms}(\rho)$
$x = 0.05$	-	203	223	123	123
$x = 0.1$	215	203	213	113	113
$x = 0.15$	211	173	203	113	103
$x = 0.2$	150	173	193	103	83
$x = 0.25$		163	183	93	83

4. Conclusions

The thermoelectric power and magnetoresistance of monovalent alkali metal (Li) substituted LaMnO_3 polycrystalline pellets prepared by solid-state reaction procedure has been studied between 80 and 320K. X-ray diffraction patterns showed a single-phase rhombohedral structure of all samples. The M-S transition temperature decreases with increasing Li content. This is due to the partial substitution of smaller Li^{1+} ions for larger La^{3+} ions or due to Zener bond blocking. The MR ratio of the $\text{La}_{1-x}\text{Li}_x\text{MnO}_y$ is about 55% under a low field ($H = 0.5$ Tesla) for low doping Li-content. This result opens new opportunities to improve performance of colossal magnetoresistive devices. The $S(T)$ curves show that Seebeck coefficient is positive sign. In addition, there are two transition the first we point it correspond to T_c which determine from MR measurements, and the second peak correspond to $T_{ms}(\rho)$. The two corresponds of transition temperature confirm that there are a correlation of structures in MR and the thermoelectric power TEP; which gives us an important clue as to the origin of this broaden magneto resistive and the fine structure which goes with it.

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