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COLOSSAL MAGNETORESISTANCE OF DY DOPED $(La_{1-x}Dy_x)_{0.67}Sr_{0.33}MnO_3$ COMPOUND.

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ABSTRACT

This work studies the correlation between colossal magnetoresistance (CMR), magnetization and microstructural effect in $(La_{1-x}Dy_x)_{0.67}Sr_{0.33}MnO_3$ when x=0.00, 0.10, 0.20 and 0.40. The magnetoresistance which is defined as $MR\% = (Ro - R_H)/R_H \times 100$; was measured by four point probe method over various temperature. All samples exhibit negative colossal magnetoresistance and the magnetoresistance increase proportionally upon substitution of La by Dy, i.e. -12.8%, -21.4% and -26.1% at 1 tesla applied magnetic field while pure LSMO sample MR% is -15.1%. This phenomenon is due to the changed Mn-O-Mn bond angle, caused by the lattice adjusting to the size difference between the La and Dy ions. The XRD patterns reveal that these compounds are orthorhombic distorted perovskite structures and single phase. The AFM images show that the grain sizes reduce when dopant concentrations increase.

INTRODUCTION

The unusual and gigantic negative magnetoresistance (MR) of the mixed valence manganites $(La_{1-x}\ Dy_x)_{0.67}Sr_{0.33}MnO_3$ with perovskite structure have aroused a surge interest because of the expected technical applications in magnetic recording elements. To distinguish the gigantic negative MR from giant magnetoresistance (GMR), it is termed as colossal lmagnetoresistance CMR). It is connected with the unique correlation between magnetism and electrical transportation that occurs from the mixed valence Mn^{3+}/ Mn^{4+} states produced by the $La^{3+}/Dy^{3+}/Sr^{2+}$ ion mixture. This leads to the formation of two noticeable effects while varying the temperature. The first effect is the coexistence of strong ferromagnetism and metallic conductivity at lower temperature that can be explained by double exchange interaction (DE) and followed by the Jahn-Teller distortion or activated behaviour occurs at higher temperature [1,2]. The DE interaction is a phenomenon of electron hopping between oxygen coupled Mn^{3+} and Mn^{4+}ions that attribute the conduction. The charge transportation is favoured when the spin of Mn ions are parallel to each other. The Jahn-Teller effect causes further splitting degeneracy d e_g of Mn^{3+} in MnO_6 octahedra. These properties are strongly influenced by the size of cation (Dy) substituted into La-site. The average ionic radius of Dy, $\langle r_d \rangle$ decreases the Mn-O-Mn bond angle after lattice adjusting; the reduced bond angle suppresses the DE. When the local spins are aligned and ordered in the direction of the

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applied magnetic field, DE is enhanced and the charge carriers scattering is reduced [3]. Thus the absence of DE increases the resistivity and gives rise to larger MR.

EXPERIMENTAL DETAILS

Polycrystalline samples of composition $(La_{1-x}\ Dy_x)0_{.67}Sr_{0.33}MnO_3$ x=0.00, 0.10, 0.20 and 0.40 were synthesized by standard solid state reaction methods. The stoichiometric proportions of La_2O_3, Dy_2O_3, $SrCO_3$ and $MnCO_3$ with nominal purities higher than 99.9% were mixed with dry method, presintering at 900°C for 12 hours, reground and pressed into pellets then reacted in air at 1300°C for 24 hours. The phase purity and crystal structure of the synthesized samples were examined by the X-ray diffractometer (XRD) with Cu Kα radiation. The grain size, existence of grain boundary and particle homogenous is observed by Atomic Force Microscope (AFM). The samples under investigation were characterized by magnetization or hysteresis loops and magnetoresistance by Vibrating Sample Magnetometer (VSM) in room temperature and four point probe method in varying temperature (90K, 100K, 150K, 200K, 250K, 270K and 300k) and magnetic field (0 Tesla to 1 Tesla)

RESULTS AND DISCUSSION

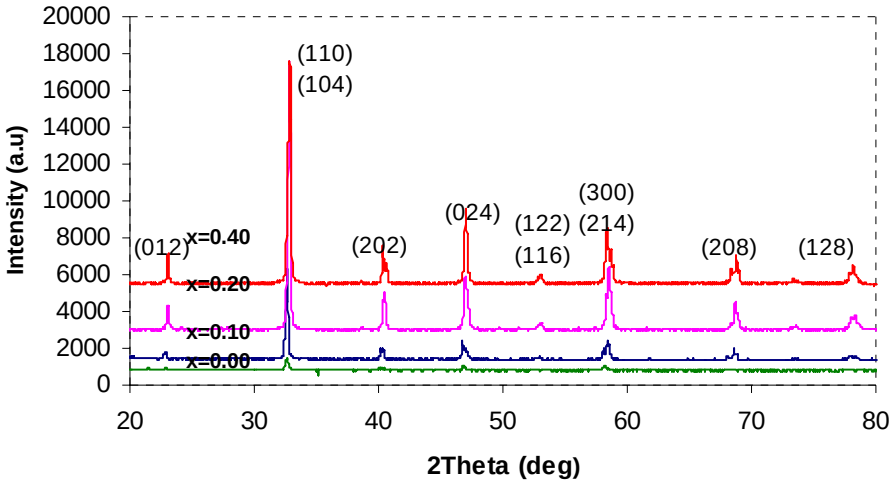

Figure 1: X-ray diffraction (XRD) patterns of $(La_{1-x}Dy_x)Sr_{0.33}MnO_3$ compound with x=0.00,0.10,0.20 and 0.40.

The X-ray diffraction (XRD) patterns of $(La_{1-x}Dy_x)Sr_{0.33}MnO_3$ compound are shown in Figure 1. The patterns reveal that the samples were single phase and samples with Dy substitution remain a similar pattern when refer to ICDD standard [4].The shifting of the peaks indicated the changes of d-spacing due to doping effect. The value of d-spacing is clearly influenced by the difference of La and Dy average ionic radius, $<r_a>$;

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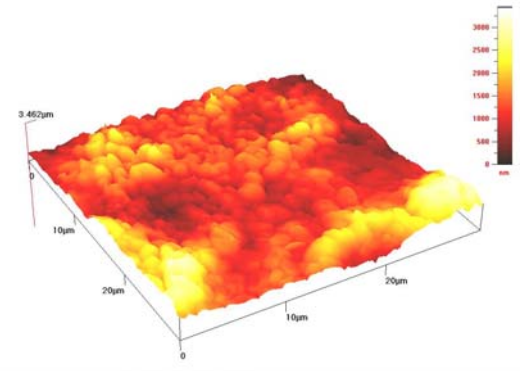
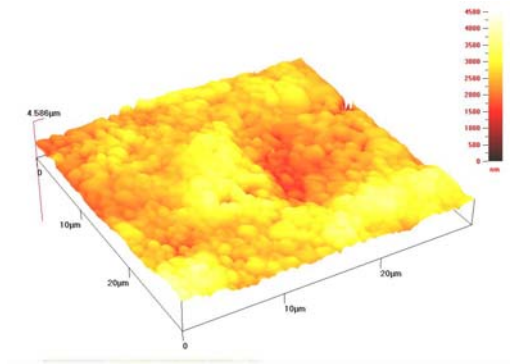
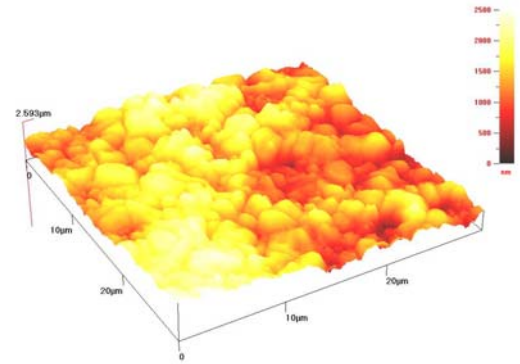
i.e. 1.15Å and 0.99Å respectively [8]. The Partial substitution of Dy at La site might lead to the increased buckling of MnO_6 octahedra resulting in the decrease of bond angle between Mn-O-Mn and structural distortions [5].



X=0.00
Average grain size: 2.24µm



X=0.10
Average grain size: 2.10µm



X=0.20
Average grain size: 1.80µm

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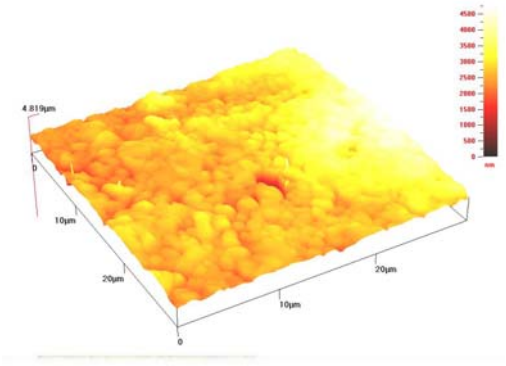

X=0.40
Average grain size: 1.32µm

Figure 2: AFM images for (a) x=0.00, (b) x=0.10, x= 0.20 and (c) x=0.40.

AFM images show that the average grain size decrease as doping concentration increase. The The surface volume ratio is in inverse proportion to the decrease of grain size. Therefore, the increase of grain boundaries can be observed from the Figure 2. The magnetic entity in the grain boundary has been a key issue in magnetoresistive behaviour. The undoubted proof has to further investigate using magnetic force microscope (MFM) for magnetic phase mapping.

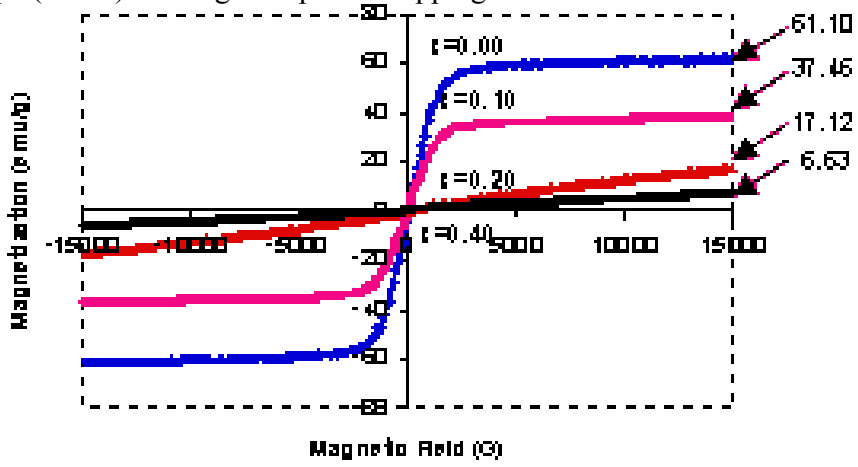

Figure 3: Hysteresis loops of (La1-xDyx)Sr0.33MnO3 compound with x=0.00,0.10,0.20 and 0.40.

The room temperature magnetization or hysteresis loops of the samples at under magnetic field (0G – 15000G) at x=0.00, 0.10, 0.20 and 0.40 is presented in Figure 3. The measured magnetizations for x=0.00, 0.10, 0.20 and 0.40 are 61.1emu/g, 37.46emu/g, 17.12emu/g and 6.63emu/g respectively. The reduced magnetization as substitution concentration increases indicated that the x=0.00 (pure LSMO) and 0.10 samples are in ferromagnetic state, while the rest are in paramagnetic state. This data also suggests that the Curie temperature, T_C reduces to lower temperature as the dopant

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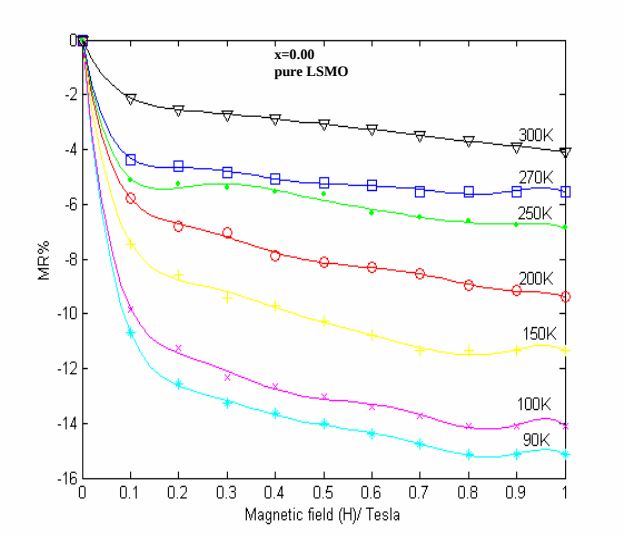
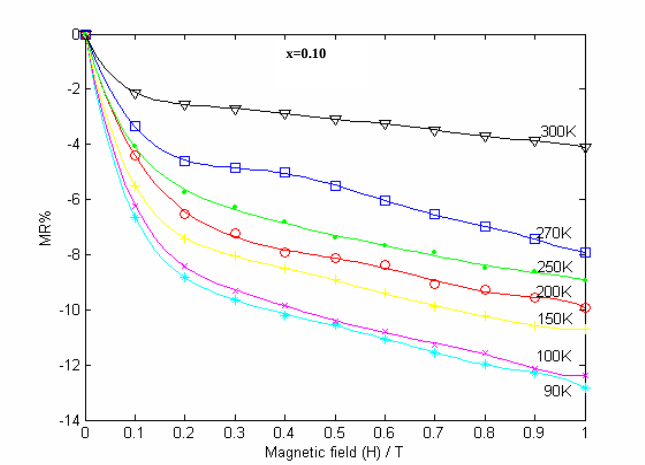
concentration increases. Hence, the strength of double exchange and the ferromagnetic interaction between the neighbouring $Mn^{3+/4+}$ ions weakens. Similar results have been obtained by others [6].



(a)



(b)

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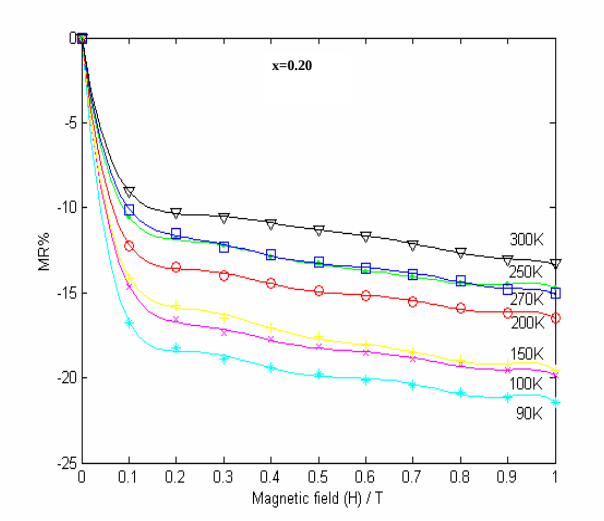
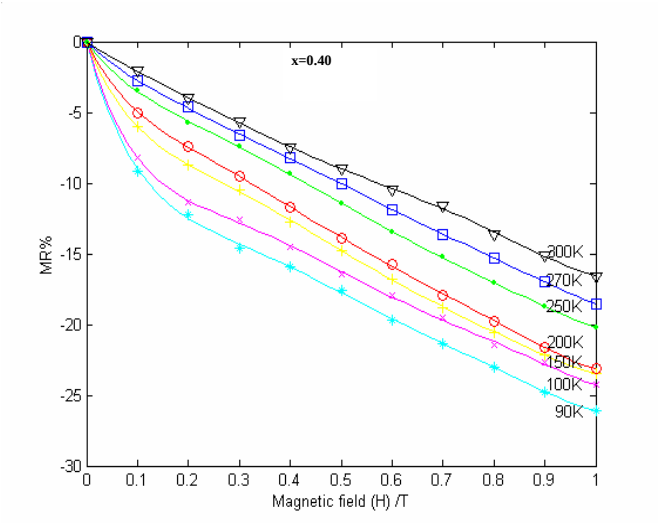


(c)



(d)

Figure 4: MR% versus magnetic field of (La1-xDyx)Sr0.33MnO3 system with (a) x=0.00, (b) x=0.10, (c) x=0.20 and (d) x=0.40.

The magnetoresistive behaviour as a function of temperature at 1 Tesla for x=0.00, 0.10, 0.20 and 0.40 composition is shown in Figure 4. The pure LSMO (x=0.00), x=0.10 and 0.20 samples show two obvious regions of MR% with different gradient where a sudden drop of magnetoresistance in low magnetic filed (0.1 Tesla) and follow by a almost saturation effect in high magnetic field (>0.1 Tesla) throughout the temperature. Its MR% value is -15.1%, -12.8% and – 21.4% at 1 Tesla magnetic field. This initial drop of magnetoresistance is known as low-field magnetoresistance (LFMR). It is due to the alignment of spin in grain boundaries and magnetic domain when low magnetic field is applied. Thus, electrons can transfer more easily across the

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grain boundaries and the resistance reduces. The sudden drop effect of resistance vanishes for the rest samples x=0.40. Its MR% also escalates drastically as the Dy concentrations increase and the MR% value is -26.1%. This behaviour is caused by the existence of amorphous entity in grain boundary and distortion of Mn-O-Mn bond angle. The spins in grain boundary and magnetic domain become hard to align in magnetic field as if they embed in viscous environment. Thus, a higher magnetic field is needed to align all the spins until saturation occur. The distortion of Mn-O-Mn bond angle suppresses the DE mechanism that controls the conduction of charge carriers and consequently gives rise to larger MR%. The observed strong reduction of the MR when increasing temperature in all samples is related to the carrier depolarization that takes place at interfaces which is due to the existence of a non collinear magnetic layer that naturally becomes even softer when increasing the temperature [7].

CONCLUSION

In summary, substitution of La with Dy in $(La_{1-x}Dy_x)Sr_{0.33}MnO_3$ leads to an increase of gain boundary, increased distortion of the Mn-O-Mn bond angle and shifting of the curie temperature. The existence of grain boundary favours the LFMR effect to occur. The change of Mn-O-Mn bond angle weakens the double exchange mechanism. The weaker double exchange mechanism, the more charge scattering occurred and gives rise to large MR.

ACKNOWLEDGEMENT

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