STRUCTURAL, MAGNETIC AND ELECTRICAL PROPERTIES AND COLOSSAL MAGNETORESISTIVE EFFECT OF La0.67Sr0.33MnO3 PEROVSKITES WITH Dy SUBSTITUTION AT La SITE

LEE OON JEW

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MASTER OF SCIENCE
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STRUCTURAL, MAGNETIC AND ELECTRICAL PROPERTIES AND COLOSSAL MAGNETORESISTIVE EFFECT OF La$_{0.67}$Sr$_{0.33}$MnO$_3$ PEROVSKITES WITH Dy SUBSTITUTION AT La SITE

By

LEE OON JEW

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Master of Science

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TABLE OF CONTENTS

DEDICATION ii
ABSTRACT iii
ABSTRAK v
ACKNOWLEDGEMENTS vii
APPROVAL viii
DECLARATION x
LIST OF TABLES xiv
LIST OF FIGURES xv
LIST OF ABBREVIATIONS/NOTATIONS/GLOSSARY OF TERMS xviii

CHAPTER

1 INTRODUCTION
1.1 Colossal Magnetoresistance 1-1
1.2 Application of Colossal Magnetoresistance 1-2
1.3 Motivation 1-4
1.4 Objective of The Thesis 1-5
1.5 Thesis Content 1-6

2 LITERATURE REVIEW
2.1 The Renaissance of Manganites as CMR Compound 2-1
2.2 Manganites 2-2
2.3 LaMnO₃ Parent Compound 2-3
2.4 Mixed Valence Manganites 2-5
  2.4.1 Intrinsic and Extrinsic CMR Characteristic in Mixed Valence Manganites 2-7
2.5 La₁₋ₓSrₓMnO₃ 2-8
  2.5.1 La₀.₃₃Sr₀.₆₇MnO₃ 2-8
2.6 Partial Substituted at La-side 2-10
  2.6.1 The Influence of Partial Substitution at La-site on Structural Distortion 2-11
  2.6.2 The Influence of Partial Substitution at La-site on Tc and TMIT 2-12
  2.6.3 The Influence of Partial Substitution at La-site on MR Effect 2-15
  2.6.4 The Influence of Partial Substitution at La-site on Hysteresis Loop 2-16
2.7 Partial Substitution at Sr-site or Divalent Cation Site 2-16
2.8 Partial Substitution at Mn-site 2-17
2.9 Review on previous work 2-17

3 THEORY OF COLOSSAL MAGNETORESISTANCE
3.1 Fundamentals of Magnetism 3-1
  3.1.1 Diamagnetism, Paramagnetism, Ferromagnetism and Antiferromagnetic 3-2
3.2 Transition from Paramagnetism to Ferromagnetism 3-4
  3.2.1 Curie-Weiss Law 3-4
3.3 Spin Glass 3-5
3.4 Magnetic Interaction in Magnetics 3-5
  3.4.1 Double Exchange 3-6
  3.4.2 Superexchange 3-7
3.5 Jahn-Teller Effect 3-9
3.6 Carriers Self Trapping / Polaron 3-10
3.7 Low Temperature Charge Transport Behaviour 3-12
3.8 High Temperature Charge Transport Behaviour 3-14
3.9 Goldsmith Tolerance Factor 3-16

4 METHODOLOGY
4.1 Sample Preparation (La_{1-x}Dy_x)_{0.67}Sr_{0.33}MnO_3 (LDySMO) 4-1
4.2 Dry Mixing of Starting Powder 4-3
4.3 Calcination 4-3
4.4 Grinding and Pressing Pellets 4-5
4.5 Sintering 4-5
4.6 Pellet Cutting and Storing 4-6
4.7 Sample Characterisation (La_{1-x}Dy_x)_{0.67}Sr_{0.33}MnO_3 (LDySMO) 4-6
4.8 Structures and Phase Identifications 4-7
4.9 Four Point Probe Resistance Measurement 4-8
4.10 Magnetoresistance Measurement in Low Temperature 4-10
4.11 Microstructure Studies 4-12
4.12 Vibrating Sample Magnetometer 4-13
4.13 DC superconducting Quantum Interference Device (SQUID) Magnetometer 4-15
4.14 Accuracy of Measurements 4-15

5 RESULTS AND DISCUSSION
5.1 XRD Pattern and Lattice Parameter of LDySMO System
5.2 Microstructure Properties
5.3 Magnetic Properties of LDySMO Samples
5.4 The Curie Weiss Expression Analysis of LDySMO
5.5 Electrical Transport properties of LDySMO Samples
  5.5.1 Low Temperature regime \( T<T_{MIT} \)
  5.5.2 High Temperature regime \( T>T_{MIT} \)
5.6 Electrical Transport at Low and High Temperature Regime
5.7 Correlation Between \( T_{MIT} \) and \( T_c \)
5.8 Phase Diagram of LDySMO Samples
5.9 Magnetoresistance Properties of LDySMO Samples
5.10 Interplay Between \( \%MR, T_c \) and \( T_{MIT} \)
5.11 Low field magnetoresistance (LFMR) Effect in LDySMO Samples
5.12 Hysteresis Loops of LDySMO Samples
5.13 The Characterization flow of LDySMO Samples

6 CONCLUSIONS AND SUGGESTIONS
6.1 Conclusion
6.2 Recommendation of Future Work

REFERENCES/BIBLIOGRAPHY
APPENDICES
BIODATA OF THE STUDENT
LIST OF PUBLICATIONS
## LIST OF TABLES

<table>
<thead>
<tr>
<th>Table</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.1</td>
<td>Accuracy of measurements</td>
</tr>
<tr>
<td>5.1</td>
<td>The lattice parameter and unit cell volume for LDySMO system</td>
</tr>
<tr>
<td>5.2</td>
<td>The Curie temperature, Néel temperature, paramagnetic Curie temperature and spin glassfrezzing temperature of LDySMO samples</td>
</tr>
<tr>
<td>5.3</td>
<td>The $T_{MIT}$ values of LDySMO samples</td>
</tr>
<tr>
<td>5.4</td>
<td>The best fit parameters ($\rho_2, \rho_0$ and $\rho_{2.5}$) and errors for all the samples</td>
</tr>
<tr>
<td>5.5</td>
<td>The values of $T_{MIT}$, $T_0$ and $N(E_f)$ for $0.10 \leq x \leq 0.90$ samples</td>
</tr>
<tr>
<td>5.6</td>
<td>The $E_a$ values for $0.10 \leq x \leq 0.90$ samples</td>
</tr>
<tr>
<td>5.7</td>
<td>Relationship between $T_c$, $T_{MIT}$ and $T_f$ for samples $0.00&lt;x&lt;0.90$</td>
</tr>
</tbody>
</table>

## LIST OF FIGURES

xiv
2.1 Pervoskite lattice structure
2.2 The A-type magnetic ordering of LaMnO₃
2.3 Phase diagram of La₁₋ₓSrₓMnO
2.4 The overall phase diagram with respect to doping rate x of La₁₋ₓSrₓMnO₃ and temperature, T
3.1 The schematic diagram of spin moment of (a) diamagnetism, (b) paramagnetism, (c) ferromagnetism and (d) antiferromagnetism
3.2 (a) Sketch of the Double Exchange proposed by Zener et al. (b) Sketch of Double exchange presented by Anderson et al and Hasegawa et al.
3.3 Generation of (a) antiferromagnetic and (b) ferromagnetic interactions between the spins of Mn ions mediated by oxygen, depending on the orientation of the Mn orbitals
3.4 Three normal modes of vibration of the oxygen octahedron; (a) Q₁, (b) Q₂ and (c) Q₃
3.5 The intertwining coupling between polaron formation and Jahn-Teller distortion
3.6 The formation of polaron. (a) A conduction electron is shown in a rigid of an ionic crystal. The forces on the ions adjacent to the electron are shown. (b) The displacement of the ions towards the electron due to the strain field
3.7 The lattice deformation due to polaron
4.1 The overall methodological procedure
4.2 Solid state reaction method for preparing samples
4.3 Schematic representation of calcinations stage for preparation of manganite ceramics
4.4 Schematic representation of calcinations stage for preparation of manganite ceramics
4.5 Philips X-ray diffractometer
4.6 Four point probe resistance measurement system

4.7 Magnetoresistance measurement system

4.8 Four-point probe holder in MR measurement system

4.9 (a) The optical lever operates by reflecting a laser beam off the cantilever. The reflected laser beam strikes a position-sensitive photodetector consisting of two side-by-side photodiodes. Data detected will be interpreted by computer

(b) Quesant Q-scope 250 Atomic Force Microscope

4.10 LakeShore Vibrating sample magnetometer

5.1 XRD spectrums for LDySMO system

5.2 The shifting of the most intense line (110) and (104) for LDySMO system

5.3 Average grain size as a function of Dy concentration for LDySMnO system

5.4 (a)-(o) The AFM images show the microstructure of LaDySMO system with doping level $0.00 \leq x \leq 0.90$

5.5 (a) The magnetization as a function of temperature curves, $M$-$T$ and (b) inverse magnetic susceptibility, $1/\chi$ against temperature curve; $1/\chi$-$T$ for the samples with Dy concentration of $x=0.00$, $0.01$, $0.05$, $0.08$, and $0.10$

5.6 (a) The magnetization as a function of temperature curves, $M$-$T$ and for the samples with Dy concentration of $x=0.30$, $0.40$, $0.50$, $0.60$, $0.80$ and $0.90$ (b) The derivative plot of magnetization against temperature curve, $dM/dT$ for the samples with Dy concentration of $x=0.30$, $0.40$, $0.50$ and $0.60$

5.7 The inverse magnetic susceptibility, $1/\chi$ against temperature curve; $1/\chi$-$T$ for the samples with Dy concentration of (a) $x=0.30$, $0.40$, (b) $0.50$, $0.60$, (c) $0.80$ and $0.90$

5.8 The Dy concentration dependent of $T_c$ for LDySMnO samples

5.9 The inverse magnetic susceptibility, $1/\chi$ against temperature curve; $1/\chi$-$T$ for the samples with Dy concentration of (a) $x=0.30$, $0.40$, (b) $0.50$, $0.60$, (c) $0.80$ and $0.90$
5.10 Temperature dependence of resistivity of LDySMO system with (a) x=0.00, 0.01, 0.05 and 0.08 (b) x=0.10, 0.30, 0.40, 0.50, 0.60, 0.80 and 0.90

5.11 The $\frac{dp}{dT}$ versus temperature for LDySMO system (a) x=0.00, 0.01, 0.05 and 0.08 (b) x=0.10, 0.30, 0.40, 0.50, and 0.80 (c) x=0.60 and 0.90

5.12 The Dy concentration dependent of $T_{MIT}$ for LDySMnO samples

5.13 The $\ln \rho$ vs. $T^{1/4}$ plots for (a) x=0.10, (b) x=0.40, (c) x=0.30 and x=0.50, (d) x=0.60 and 0.90; and (e) x=0.80.

5.14 The $\ln \rho/T$ vs. $1/T$ plots of (a) x=0.10, (b) x=0.40, (c) x=0.30 and x=0.50, (d) x=0.60 and 0.90 and (e) x=0.80.

5.15 The phase diagram of LDySMO samples

5.16 The applied magnetic field (0T-1T) and temperature (90K-300K) dependence of MR 3D curve of LDySMO samples with (a) x=0.00 and (b) x=0.60

5.17 MR variation as a function of applied magnetic field for samples with x=0.00, 0.01, 0.02, 0.04, 0.05, 0.06 and 0.08 at temperature 90K, 100K, 150K, 200K, 250K, 270K and 300K

5.18 MR variation as a function of applied magnetic field for samples with x=0.80 and 0.90 at temperature 90K, 100K, 150K, 200K, 250K, 270K and 300K

5.19 The magnetic field dependence of the isothermal magnetization curves, M-H at 300K for the LDySMO samples

5.20 The enlargement of magnetization curve for x=0.00 sample around zero field

5.21 The magnetic field dependence of the isothermal magnetization curves, M-H at 300K for the LDySMO samples with $0.00 \leq x \leq 0.10$

5.22 The magnetic field dependence of the isothermal magnetization curves, M-H at 300K for the LDySMO samples with $0.20 \leq x \leq 0.80$. 

xvii
<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>t_{2g}^{1}e_{g}^{1}</td>
<td>Electron configuration</td>
</tr>
<tr>
<td>&lt;ra&gt;</td>
<td>A-site ionic radius</td>
</tr>
<tr>
<td>2D</td>
<td>2 dimension</td>
</tr>
<tr>
<td>2Theta</td>
<td>Bragg diffraction angle</td>
</tr>
<tr>
<td>3D</td>
<td>3 dimension</td>
</tr>
<tr>
<td>a, b, c</td>
<td>Lattice parameter</td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>ABO$_3$</td>
<td>Pervoskite</td>
</tr>
<tr>
<td>ABS</td>
<td>Antilock Brake System</td>
</tr>
<tr>
<td>AC</td>
<td>Alternating current</td>
</tr>
<tr>
<td>AFM</td>
<td>Atomic Force Microscope</td>
</tr>
<tr>
<td>AMR</td>
<td>Anisotropic magnetoresistance</td>
</tr>
<tr>
<td>CMR</td>
<td>Colossal magnetoresistance</td>
</tr>
<tr>
<td>D</td>
<td>Divalent cation or alkaline earth</td>
</tr>
<tr>
<td>d$_{A-O}$</td>
<td>Distance between the A site</td>
</tr>
<tr>
<td>DC</td>
<td>Direct current</td>
</tr>
<tr>
<td>DE</td>
<td>Double exchange</td>
</tr>
<tr>
<td>d$_{Mn-O}$</td>
<td>Shortest distance of Mn-O</td>
</tr>
<tr>
<td>Dy</td>
<td>Dysprosium</td>
</tr>
<tr>
<td>E$_a$</td>
<td>Activation energy</td>
</tr>
<tr>
<td>EMR</td>
<td>Extraordinary magnetoresistance</td>
</tr>
</tbody>
</table>
GMR Giant magnetoresistance
H Magnetic field
ICDD International Crystal Diffraction Data
La Lanthanum
LFMR Low field magnetoresistance
Ln Trivalent lanthanide cation
M Magnetization
Mn Magnesium
MR Magnetoresistance
MRAM Magnetoresistance Random Access Memory chip
N(E_f) The density of states at the Fermi level
O Oxygen
Q_1, Q_2, Q_3 Normal modes of vibration of the oxygen octahedron
R Resistance
R_0 Resistances in the absence of a magnetic field
R_H Resistances in the presence of a magnetic field
SPH Small polaron hopping
SPM Scanning Probe Microscope
Sr Strontium
T Tesla
t Goldsmith tolerance factor
T_c Curie temperature
T_f Freezing Temperature
<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_{MIT}$</td>
<td>Metal-insulator transition temperature</td>
</tr>
<tr>
<td>TMR</td>
<td>Tunnelling magnetoresistance</td>
</tr>
<tr>
<td>$T_N$</td>
<td>Neel temperature</td>
</tr>
<tr>
<td>VLMR</td>
<td>Very large magnetoresistance</td>
</tr>
<tr>
<td>VRH</td>
<td>Variable range hopping</td>
</tr>
<tr>
<td>VSM</td>
<td>Vibrating Sample Magnetometer</td>
</tr>
<tr>
<td>XRD</td>
<td>X-ray diffraction</td>
</tr>
<tr>
<td>$\alpha, \beta, \gamma$</td>
<td>Phase designations</td>
</tr>
<tr>
<td>$\theta$</td>
<td>Mn-O-Mn bond angle</td>
</tr>
<tr>
<td>$\Theta$</td>
<td>Paramagnetic Curie temperature</td>
</tr>
<tr>
<td>$\mu_0$</td>
<td>Permeability of the free space</td>
</tr>
<tr>
<td>$\rho$</td>
<td>Resistivity</td>
</tr>
<tr>
<td>$\sigma$</td>
<td>Conductivity</td>
</tr>
<tr>
<td>$\chi$</td>
<td>Magnetic susceptibility</td>
</tr>
</tbody>
</table>
To My Family and Friends.........
Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Master of Science

STRUCTURAL, MAGNETIC AND ELECTRICAL PROPERTIES AND COLOSSAL MAGNETORESISTIVE EFFECT OF La$_{0.67}$Sr$_{0.33}$MnO$_3$ PEROVSKITES WITH Dy SUBSTITUTION AT La SITE

By

LEE OON JEW

June 2007

Chairman: Professor Abdul Halim bin Shaari, PhD

Faculty: Science

A thorough study on structural, magnetic and electrical properties and colossal magnetoresistive (CMR) effect in La$_{0.67}$Sr$_{0.33}$MnO$_3$ or generically known as LSMO manganites substituted with dysprosium (Dy) at lanthanum (La) site is the main area of research in this thesis. In the first part of this work, the samples of (La$_{1-x}$Dy$_x$)$_{0.67}$Sr$_{0.33}$MnO$_3$ (LDSMO) with x=0.00-0.90 were synthesized using standard solid state reaction method. The second part involves the characterization of the samples using X-ray Diffractometer (XRD), Vibrating Sample Magnetometer (VSM), four point probe electrical resistivity and magnetoresistance measurement system, and Atomic Force Microscope (AFM). The XRD pattern for all samples reveals a single phase pattern with rhombohedral structure. When Dy concentration increases, the magnetization value at any given temperature decreases markedly. Moreover, the magnetization (temperature dependence of magnetization and isothermally hysteresis loop) show a prominent transition corresponding to Curie temperature, $T_C$, arising from long range ferromagnetic order to a short range nature.
with paramagnetic order (0.00 ≤ x ≤ 0.06), antiferromagnetic (x=0.90) and spin glass state (x=0.80). The electrical resistivity increases with the shifting of metal-insulator transition temperature, $T_{MIT}$ to lower temperature. Low temperature resistivity data signify that the resistance of electrical transport mechanism in the metallic region can be ascribed as electron-electron scattering with minor attribution of magnon-electron scattering. On the other hand, the high temperature resistivity data satisfy the variable range hopping (VRH) model and small polaron hopping (SPH) model. The considerable change in the density of states at Fermi level, $N(E_f)$ and activation energy, $E_a$ which was obtained from fitting the electrical transport data, proved that the mobility of the electron decreases proportionally with Dy concentration. The $N(E_f)$ values were found within the range of $1.53 \times 10^{16}$ eV$^{-1}$ cm$^{-3}$ to $1.45 \times 10^{18}$ eV$^{-1}$ cm$^{-3}$ whereas the $E_a$ values were ranged from $2.96 \times 10^2$ meV to $1.32 \times 10^3$ meV. The $T_C$-$T_{MIT}$ discrepancy is due to the fact that the former is an intrinsic characteristic, the latter depends strongly on the extrinsic factor e.g. grain size (grain boundary). The effect of grain size on the CMR mechanism is analyzed from AFM images. The grain size was found to decrease exponentially from 2.88 $\mu$m (x=0.00) to 1.34 $\mu$m (x=0.90) with the increase of Dy concentration. As the grain size decreases, the ratio of surface over volume increased. Hence, the influence of grain boundaries effect cannot be excluded and eliminated in all samples. Overall, negative CMR in ascending order had been obtained in all samples as decreasing temperature at low magnetic field. This phenomenon is known as Low Field Magnetoresistance (LFMR). Spin dependent scattering, spin polarization and tunneling between neighbouring grains and the magnetically disordered grain boundaries seems to be responsible for the LFMR effect. The highest CMR value with 62.2% is obtained in x=0.60 LDySMO sample at 90K.
SIFAT STRUKTUR, MAGNET DAN ELEKTRIK DAN KESAN MAGNETORINTANGAN RAKSASA PEROVSKIT La$_{0.67}$Sr$_{0.33}$MnO$_3$
DENGAN PENGGANTIAN Dy PADA TAPAK La

Oleh
LEE OON JEW
Jun 2007

Pengerusi: Profesor Abdul Halim bin Shaari, PhD
Fakulti: Sains

Kajian menyeluruh tentang sifat struktur, magnet dan elektrik dan magnetorintangan raksaka terhadap La$_{0.67}$Sr$_{0.33}$MnO$_3$ atau dikenali sebagai manganit LSMO yang digantikan dengan dysprosium (Dy) pada tapak lanthnum (La) adalah kajian utama dalam tesis ini. Dalam bahagian pertama kajian ini, sampel (La$_{1-x}$Dy$_x$)$_{0.67}$Sr$_{0.33}$MnO$_3$ (LDySMO) dengan x=0.00-0.90 telah disintesis melalui kaedah tindak balas keadaan pepejal. Manakala bahagian kedua tesis ini adalah pencirian dan pengukuran sifat sampel menggunakan alat belauan sinar-X (XRD), magnetometer getaran sampel (VSM), kaedah penduga empat titik (pengukuran kerintangan dan magnetorintangan) dan mikroskop daya atom (AFM). Corak belauan sinar-X menunjukkan fasa tunggal dengan kewujudan struktur rombohedral. Apabila kepekatan Dy meningkat, nilai kemagnetan dalam keseluruhan suhu didapati berkurang. Pengukuran kemagnetan (persandaran kemagnetan sampel terhadap suhu dan lengkung histeresis isoterma) menunjukkan peralihan suhu Curie, Tc yang menonjol dan kemunculan dari jatul panjang susunan ferromagnet ke jatul pendek yang berkait rapat dengan susunan
paramagnet \((0.00 \leq x \leq 0.06)\), antiferromagnet \((x=0.90)\) dan keadaan spin kaca \((x=0.80)\). Kerintangan elektrik meningkat dengan peralihan suhu logam-penebat, \(T_{MIT}\) ke suhu rendah. Data kerintangan suhu rendah menandakan mekanisme rintangan dalam bahagian logam ini boleh dikaitkan dengan penyerakan elektron-elektron yang dilengkapi dengan sumbangan minor dari serakan magnon-elektron. Selain itu, data kerintangan suhu tinggi pula memenuhi model Loncatan Julat Bolehubah (VRH) dan model Loncatan polaron kecil (SPH). Ketumpatan keadaan di aras Fermi, \(N(E_f)\) dan tenaga pengaktifan, \(E_a\) yang diperoleh daripada data pemadanan membuktikan kelincahan elektron berkurang secara berkadar langsung dengan kepekatan Dy. Nilai \(N(E_f)\) didapati berjulat antara \(1.53 \times 10^{16} \text{ eV}^{-1}\text{cm}^{-3}\) ke \(1.45 \times 10^{18} \text{ eV}^{-1}\text{cm}^{-3}\) manakala nilai \(E_a\) berubah dari \(2.96 \times 10^2\) meV ke \(1.32 \times 10^3\) meV. Perbezaan antara \(T_c-T_{MIT}\) adalah disebabkan oleh faktor intrinsik dan disusuli dengan faktor ekstrinsik seperti saiz butiran (sempadan butiran). Kesah saiz butiran terhadap mekanisme CMR dianalisis daripada imej AFM. Saiz butiran dalam kesemua sampel didapati menurun secara eksponen dari \(2.88 \mu\text{m} (x=0.00)\) ke \(1.34 \mu\text{m} (x=0.90)\) kerana bahan dopan tidak menyumbang kepada pertumbuhan butiran. Apabila saiz butiran berkurang, nisbah permukaan terhadap isipadu butiran adalah tinggi. Oleh demikain, pengaruh sempadan butiran tidak boleh dikecualikan. Secara keseluruhan, CMR negatif dalam urutan menaik telah dikesan dalam semua sampel ketika suhu menurun pada medan magnet yang rendah. Fenomena ini dikenali sebagai magnetorintangan medan rendah (LFMR). Serakan spin, polarisasi spin dan penerowongan antara butiran bersebelahan melalui sempadan butiran yang tidak terjajar pada arah medan magnet adalah dipertanggungjawab terhadap kesan LFMR. Nilai CMR tertinggi sebanyak 62.2% diperoleh daripada sampel LDySMO \((x=0.60)\) pada suhu 90K.
ACKNOWLEDGEMENTS

I wish to express my deepest gratitude to the chairman of the supervisory committee, Professor Dr. Abdul Halim Shaari, who gave me the platform to pursue my studies, opportunity to explore the colossal magnetoresistance and constant encouragement.

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I certify that an Examination Committee has met on 11\textsuperscript{th} Jun 2007 to conduct the final examination of Lee Oon Jew on her Master of Science thesis entitled “Structural, Magnetic and Electrical Properties and Colossal Magneto resistive effect of Lao.67Sr0.33MnO3 Perovskites with Dy Substitution at La site” in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulations 1981. The Committee recommends that the student be awarded the Master of Science degree.

Members of the Examination Committee are as follows:

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Faculty of Science
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(Chairman)

\textbf{Elias Saion, PhD}
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(Internal Examiner)

\textbf{Hishamuddin Zainuddin, PhD}
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Date:
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Date: 9 August 2007
DECLARATION

I declare that the thesis is my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously, and is not concurrently, submitted for any other degree at Universiti Putra Malaysia or at any other institution.

LEE OON JEW

Date: 2 August 2007
I certify that an Examination Committee has met on 11th June 2007 to conduct the final examination of Lee Oon Jew on her Master of Science thesis entitled “Structural, Magnetic and Electrical Properties and Colossal Magnetoresistive effect of La_{0.67}Sr_{0.33}MnO_{3} Perovskites with Dy Substitution at La site,” in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulations 1981. The Committee recommends that the candidate be awarded the relevant degree. Members of the Examination Committee are as follows:

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xii
CHAPTER 1

INTRODUCTION

1.1 Colossal Magnetoresistance

The continually increasing demand for ultrahigh density magnetic information storage, retrieval and sensing electronic device has driven a significant effort to secure fundamental base for the future manufacturing of devices and sensors based on colossal magnetoresistance (CMR) materials. Thus, there has been intense attention, theoretical and experimental paid to these colossal magnetoresistance materials in the search for a complete understanding of the fundamental science behind the phenomena exhibited by the material.

Magnetoresistance (MR) is a change in the electrical resistance of a material when it is subjected to magnetic field and defined as:

\[
\text{%MR} = \frac{\Delta R}{R_0} = \frac{R_H - R_0}{R_0} \times 100\%
\]

where \(R_H\) and \(R_0\) are the resistances in the presence and absence of a magnetic field respectively. In general, the MR can be classified by their origin and working principle into several types namely, anisotropic magnetoresistance (AMR), giant magnetoresistance (GMR), colossal magnetoresistance (CMR), tunnelling