COLOSSAL MAGNETORESISTANCE EFFECT OF LANTHANUM STRONTIUM MANGANESE OXIDE CERAMICS DOPED WITH TIN, BISMUTH AND INDIUM ON MANGANESE SITES

By

RAMADAN EBRAHIM ALI SHAIBOUB

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

April 2004

DEDICATION

To my parents, for their care, patience and teaching

To my family and Dear wife

For her support, understanding and concern…

My Sons, brothers and sisters

Fellow friends, course mates and Universiti Putra Malaysia as a whole

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Master of Science

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Chairman: Professor Abdul Halim Shaari, Ph.D.

Faculty: Science and Environmental Studies

Since the discovery of colossal magnetoresistance (CMR) effect in the perovskite type of manganites, much research works have been carried out to improve the CMR value by introducing different type of dopant in the system. In this work the effect of rare earth elements Tin (Sn), Bismuth (Bi) and Indium (In) substitution in the $La_{0.67}Sr_{0.33}MnO_3$ system on Mn site had been investigated. The (LSMSnO), (LSMBiO) and (LSMInO), with x=0.00 to x=0.40 ceramic samples were prepared by solid-state reaction technique. The temperature dependence of the magnetic susceptibility, the electrical transport, XRD spectrum of all samples, microstructure and magnetoresistance measurements in the three systems have been studied systematically. With increasing Sn, Bi and In concentration, Tc shifted toward lower temperature, but the lattice constants remain almost unchanged. For higher doping, the spin glass-like state is observed and the system behaves like insulator. These results strongly suggest that the dopant Sn, Bi and In ions weaken the double

exchange interaction. The X-ray diffraction patterns indicate that $La_{0.67}Sr_{0.33}MnO_3$ compounds are in single phase with rhombohedra structure. From the ac magnetic susceptibility χ measurements, transitions from paramagnetic (PM) to ferromagnetic (FM) state are observed, indicating the loss of ferromagnetical ordering and consequently weaken the double exchange (DE) mechanism. When the competition between ferromagnetism and antiferromagnetism is strong, spin glass behavior is usually found with freezing temperature for all systems. From the resistance measurements, the metal-to-insulator (M-I) transition occurs at a T_{mi}. The (M-I) transitions temperature, T_{mi} shifts towards lower temperature as the Sn, Bi and In content increases and the resistance increases accordingly. The grain size for the dopant concentration of Tin, Bismuth and Indium doped with LSMO system, shows that the grain size essentially decreases with the increase of the dopant amount but remains almost constant size at $x>0.12$, which indicates that the dissolving amount of the dopant in LSMO may be less when $x<0.12$. MR values of the three systems are very temperature dependent. The samples exhibit the maximum CMR value at certain temperature. For undoped sample, the maximum MR value observed is 11 %. Among these three doped systems, the highest value of CMR with dopant is observed for LSMBiO system with $x=0.06$ with the value of 45 % at 250 K. The colossal magnetoresistive effect of the three systems, appear between 100 K and 250 K.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia
sebagai memenuhi keperluan ijazah Master Sains

KESAN MAGNETORINTANGAN RAKSAKSA BAGI SERAMIK LANTHANUM STRONTIUM MANGANIS OKSIDA DIDOP DENGAN TIMAH, BISMUTH DAN INDIUM PADA TAPAK MANGANIS

Oleh

RAMADAN EBRAHIM ALI SHAIBOUB

April 2004

Pengerusi: Profesor Abdul Halim Shaari, Ph.D.

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Sejak penemuan kesan magnetorintangan Raksaksa (MRR) dalam bahan manganit jenis perovskite, banyak kajian telah dijalankan untuk memperbaiki nilai MRR dengan menggunakan jenis bahan campuran yang berlainan pada sistem. Dalam kajian ini, kesan dari unsur nadir-bumi seperti penggantian Timah (Sn), Bismuth (Bi) dan Indium (In) dalam sistem $La_{0.67}Sr_{0.33}MnO_3$ pada tapak Manganis telah dikaji. Sampel seramik dari jenis LSMSnO, LSMBiO dan LSMInO dengan x=0.00 sehingga x=0.40 disediakan melalui teknik tindakbalas keadaan pepejal. Persandaran terhadap oleh suhu kerentanan magnet, pengaliran elektrik, spektrum XRD untuk semua sampel mikrostruktur dan pengukuran magnetorintangan dalam ketiga-tiga sistem telah dikaji secara sistematik. Dengan penambahan kandungan Sn, Bi dan In, Tc beralih ke suhu lebih rendah, tetapi pemalar kekisi hampir tidak berubah. Untuk pendopan yang lebih tinggi, keadaan menyerupai spin-kaca telah ditemui dan sistem berkelakuan seperti penebat. Hasil ini mengukuhkan cadangan bahawa bahan dopan

Sn, Bi dan In ion melemahkan interaksi pertukaran ganda dua. Corak belauan sinaran-x menunjukkan bahawa bahan $La_{0.67}Sr_{0.33}MnO_3$ adalah dalam keadaan satu fasa dengan struktur rhombohedron. Dari pengukuran kerentanan magnet AC,χ, menunjukkan satu peralihan dari keadaan paramagnet (PM) ke feromagnet (FM) kelihatan, yag menunjukkan kehilangan tertib feromagnet dan seterusnya melemahkan mekanisma pertukaran ganda dua (DE). Apabila pertandingan antara feromagnet dengan antiferomagnet adalah kuat, kelakuan spin kaca akan dijumpai dengan suhu beku untuk semua sistem. Dari ukuran rintangan, peralihan logam ke penebat (M-I) wujud pada T_{mi}. Suhu peralihan (M-I), T_{mi} beralih ke suhu lebih rendah bila kandungan Sn, Bi dan In bertambah dan rintangan turut bertambah. Saiz butiran untuk kandungan dopan bagi timah, Bismuth dan Indium untuk LSMO, menurun apabila amaun dopan bertambah tetapi hampir tidak berubah pada $x>0.12$, ini menunjukkan amoun resapan dopan dalam LSMO mungkin kurang apabila $x<0.12$. Nilai MR untuk ketiga-tiga sistem adalah sangat bergantung kepada suhu. Sampel menunjukkan nilai MRR maksimum dalam suhu tertentu. Untuk sampel tulen, nilai maksimum MRR yang didapati adalah 11%. Antara ketiga-tiga sistem ini, nilai MRR yang paling tinggi adalah untuk sistem LSMBiO dengan $x=0.06$ iaitu 45% pada 250K. Magnetorintangan Raksaksa untuk ketiga-tiga sistem, wujud antara 100K dan 250K.

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I certify that an Examination Committee met on 09 / 4 / 2004 to conduct the final examination of Ramadan Ebrahim Ali Shaiboub on his Master of Science thesis entitled "Colossal Magnetoresistance Effect of Lanthanum Strontium Manganese Oxide Ceramics Doped with Tin, Bismuth and Indium on Manganese Sites" in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulation 1981. The Committee recommends that the candidate be awarded the relevant degree. Members of the Examination Committee are as follows:

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This thesis submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Master of Science. The members of the Supervisory Committee are as follows:

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DECLARATION

I hereby declare that the thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at UPM or other institutions.

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RAMADAN EBRAHIM ALI SHAIBOUB

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