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journal homepage: [www.elsevier.com/locate/jmmm](http://www.elsevier.com/locate/jmmm)Correlation between electrical and magnetic properties of polycrystalline  $\text{La}_{0.5}\text{Ca}_{0.5}\text{Mn}_{0.98}\text{Bi}_{0.02}\text{O}_3$ A. Krichene<sup>a,\*</sup>, M. Bourouina<sup>a</sup>, D. Venkateshwarlu<sup>b</sup>, P.S. Solanki<sup>c</sup>, S. Rayaprol<sup>d</sup>, V. Ganesan<sup>b</sup>, W. Boujelben<sup>a</sup>, D.G. Kuberkar<sup>c</sup><sup>a</sup> Laboratoire de Physique des Matériaux, Faculté des Sciences de Sfax, Université de Sfax, B. P. 1171, 3000 Sfax, Tunisia<sup>b</sup> UGC-DAE Consortium for Scientific Research, University Campus, Khandwa Road, Indore 452017, India<sup>c</sup> Department of Physics, Saurashtra University, Rajkot 360005, India<sup>d</sup> UGC-DAE Consortium for Scientific Research, Mumbai Centre, B.A.R.C. Campus, Mumbai 400085, India

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## ABSTRACT

We have reported in this work the study of correlation between electrical transport and magnetic properties of  $\text{La}_{0.5}\text{Ca}_{0.5}\text{Mn}_{0.98}\text{Bi}_{0.02}\text{O}_3$  polycrystalline sample prepared by solid state method. Structural analysis reveals that presently studied compound crystallizes in the orthorhombic structure with  $Pnma$  space group. Temperature dependence of magnetization indicates that our studied compound undergoes a paramagnetic–ferromagnetic transition at Curie temperature  $T_C=237$  K. Magnetotransport analysis was successfully carried out using percolation model in the temperature range 40–300 K for magnetic field values up to 14 T. Strong correlation between electrical and magnetic properties was observed along with the absence of charge ordering inside the structure of our sample.

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## 1. Introduction

Perovskite manganites characterized by general formula  $\text{Re}_{1-x}\text{Ae}_x\text{MnO}_3$  (where Re is a lanthanide and Ae is an alkaline earth) were widely studied in the last decades. Although these compounds are easy to elaborate, they present a diversity of fascinating phenomena due to the highly correlated electrons, which implies the presence of some correlations between their different physical properties such as electrical and magnetic ones [1–7]. Theoretically, half doped manganites are antiferromagnetic insulators in nature at low temperatures with a competing ferromagnetic state in the vicinity having different energy criterion [8]. It is widely accepted that mixed valence manganites are intrinsically disordered systems having small scale phases present, such as ferromagnetic metallic (FMM), paramagnetic insulating (PMI), antiferromagnetic insulating (AFMI), charge ordered insulating (COI), etc, coexisting in one phase resulting in the inhomogeneous electronic and magnetic behavior, which is strongly dependent on the temperature and applied magnetic field [9,10]. Delicate interplay between different sources of energy, generated from different ground states of manganites such as FMM, PMI, AFMI, COI, etc, is the cause for unique transport of charge carriers (itinerant  $e_g$  electrons) and their coupling with lattice, spin, orbital

and charge resulting in the phase coexistence and phase separation scenario in manganites leading to a wide range of striking physical phenomena. Generally, two major consequences are expected from the phase separated/phase coexisted manganites: (i) electronic phase separation between the phases with different densities that lead to nanometer scale equal-density phases and (ii) disorder induced phase separation with percolative characteristics between equal-density phases, driven by disorder near first-order metal–insulator phase transitions [9].

Amongst all the charge ordered manganites,  $\text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$  is the most studied compound in the last years. It shows paramagnetic (PM) to ferromagnetic (FM) phase transition at  $T_C\sim 220$  K followed by a transformation to charge ordered antiferromagnetic (CO-AFM) phase at  $T_{CO}\sim 150$  K [11]. Effect of B-site substitution in manganites has been less studied due to the fact that B-site substitution directly affects the ferromagnetic ground state of manganites and alters the Zener double exchange mechanism [12,13]. This effect produces an important disorder in manganese network which influences the electrical and magnetic properties of manganites [12,13]. Substitution of diamagnetic Bismuth ion at La-site in  $\text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$  alters strongly its physical properties and CO gets stabilized effectively [14,15]. The effect of Bi doping at B-site was investigated in several perovskites [16–24].

In a previous work, we have studied the effect of B-site bismuth doping effect on the structural, magnetic and electrical properties of  $\text{La}_{0.5}\text{Ca}_{0.5}\text{Mn}_{1-x}\text{Bi}_x\text{O}_3$  ( $x=0, 0.02$  and  $0.05$ ) polycrystalline samples [17]. We have reported that the CO state has been found in the

\* Corresponding author.

E-mail address: [akramkri@hotmail.fr](mailto:akramkri@hotmail.fr) (A. Krichene).

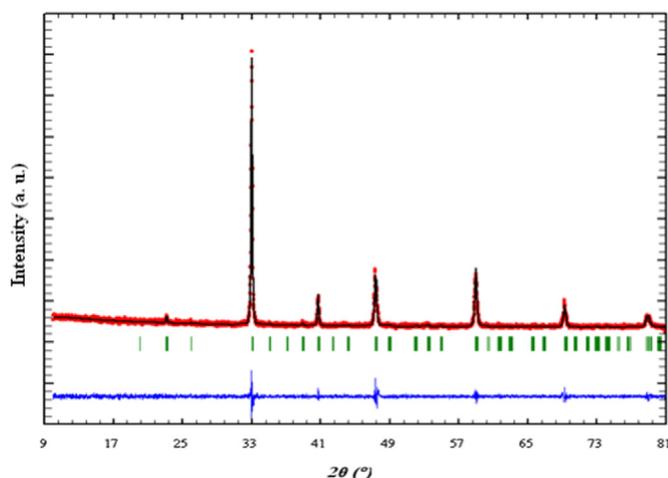
parent compound ( $x=0$ ) which was first annihilated with 2% Bi doping, but then, it was established again for the sample with  $x=0.05$ . Besides, we did not find a suitable model to describe the evolution of resistivity in the whole temperature range (especially around the temperature of electrical transition  $T_\rho$ ) for all the applied magnetic field values varying from 0 to 14 T. As per the best of our knowledge, there is no model which explains the magneto-transport mechanism for a phase-separated compound like the case of our samples with  $x=0$  and 0.05. For this fact, we have only treated, in this work, the sample with  $x=0.02$  in order to understand the magnetotransport process in the whole temperature range 40–300 K for all the magnetic field values up to 14 T and to confirm the absence of CO state inside its structure; the obtained results were discussed.

## 2. Experimental techniques

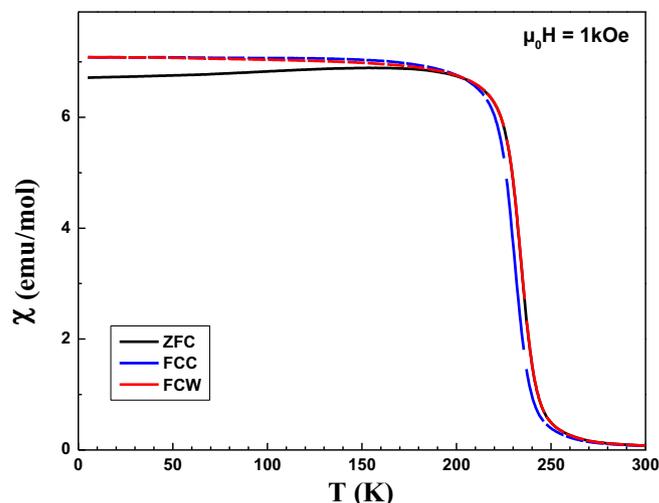
Polycrystalline  $\text{La}_{0.5}\text{Ca}_{0.5}\text{Mn}_{0.98}\text{Bi}_{0.02}\text{O}_3$  was prepared using the standard solid state reaction at high temperature. High purity  $\text{La}_2\text{O}_3$ ,  $\text{CaCO}_3$ ,  $\text{MnO}_2$  and  $\text{Bi}_2\text{O}_3$  were mixed in stoichiometric proportions and ground for 1 h in an agate mortar. The mixture was then fired for 18 h at 1073 K. The obtained powders underwent then several cycles of grinding, pelletizing and heating for 18 h in 1273 K. And finally, the pellets were sintered at 1473 K for 48 h. Structural study was carried out by X-ray powder diffraction followed by Rietveld refinement. The magnetic measurement in the temperature range 5–300 K under an applied magnetic field of 1 kOe was performed by a vibrating sample magnetometer. PPMS facility, based on standard four probe method, was used to get electrical measurements in the temperature range 2–300 K under an applied magnetic field values varying from 0 to 14 T.

## 3. Results and discussions

The refinement of X-ray powder diffraction pattern for our sample is shown in Fig. 1. It is clear that structure of the sample is pure and single phase. The refinement results indicate that our sample belongs to the orthorhombic structure with  $Pnma$  space group (No. 62). The cell parameters were found to be  $a=5.423$  Å,  $b=7.685$  Å and  $c=5.414$  Å. In order to study the magnetic behavior of our elaborated specimen, we have represented in Fig. 2 the evolution of magnetic susceptibility as a function of temperature



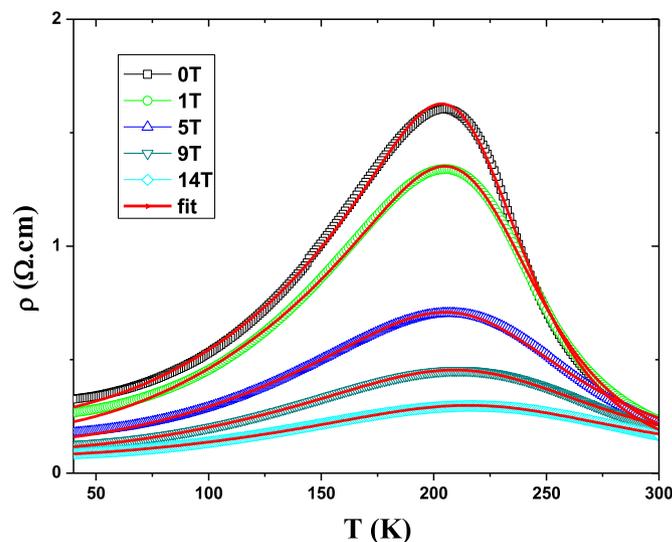
**Fig. 1.** Refinement for the sample  $\text{La}_{0.5}\text{Ca}_{0.5}\text{Mn}_{0.98}\text{Bi}_{0.02}\text{O}_3$ : experimental data in red, calculated data in black, difference between them in blue and Bragg positions in green. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)



**Fig. 2.** Temperature dependence of zero field cooled (ZFC), field cooled cooling (FCC) and field cooled warming (FCW) magnetic susceptibility for  $\text{La}_{0.5}\text{Ca}_{0.5}\text{Mn}_{0.98}\text{Bi}_{0.02}\text{O}_3$  sample.

under an applied magnetic field of 1 kOe. It can be seen that our sample exhibits a unique magnetic transition from paramagnetic to ferromagnetic state with decreasing temperature at  $T_C=237$  K. This fact indicates that the charge ordering state in the pristine compound  $\text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$  [25] gets suppressed by only 2% of non magnetic quenched disorder (i.e. Bi). In addition, ZFC (zero field cooled) magnetization is lower than FCC (field cooled cooling) one for low temperature values suggesting the presence of a spin glass like state. The presence of such state can be explained by the effect of both quenched disorder and spin frustrations. The magnetic frustration inside the structure is a direct result of the competition between ferromagnetic double exchange mechanism and anti-ferromagnetic super exchange one.

We have performed resistivity measurements as a function of temperature for several values of applied magnetic field, the results are represented in Fig. 3. It is clear that our sample exhibits a metal–insulator transition with increasing temperature around  $T_\rho=205$  K, this value is close to  $T_C$  which suggests the presence of a correlation between electrical and magnetic properties. The



**Fig. 3.** Temperature dependence of resistivity for several values of applied magnetic field for  $\text{La}_{0.5}\text{Ca}_{0.5}\text{Mn}_{0.98}\text{Bi}_{0.02}\text{O}_3$  sample. The red curves represent the fitting using relation (8). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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