Studies of the electroresistive properties of electronic oxides

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Abstract
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Studies of the Electroresistive Properties of Electronic Oxides

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In this paper we report on the non-linear voltage-current relationship of three doped lanthanum manganite compounds ($La_{0.8}Li_{0.2}MnO_3$, $La_{0.66}Sr_{0.33}MnO_3$ and $La_{0.8}Pb_{0.2}MnO_3$) and three manganite cobaltite compounds ($DyMn_{0.5}Co_{0.5}O_3$, $HoMn_{0.5}Co_{0.5}O_3$ and $NdMn_{0.5}Co_{0.5}O_3$). We compare and contrast the magnitude and temperature dependence of the non-linear behaviour, and suggest this behaviour may be largely due to phase separation in the compounds. We also compare the behaviour of single-crystal and polycrystalline bulk samples of lead-doped lanthanum manganite ($La_{0.8}Pb_{0.2}MnO_3$), which provide evidence for grain boundaries playing an important role in the non-linear voltage-current relationships in certain compounds.

1. Introduction

Non-linear current/voltage relationships in oxide materials have become the focus of significant research over the last decades [1]. We report on electric-field induced non-linear conduction, known as electroresistance (ER). This is an interesting and not yet fully explained phenomenon, whereby an electric field applied to a sample can dramatically reduce the resistivity of the sample. ER has a magnetic-field induced analogue, magnetoresistance (MR), however there is significant evidence that these are two separate phenomena [2]. There is evidence that ER can occur intrinsically [3,4] or as a result of grain boundary effects [5] in samples.

Phase separation (PS) has been suggested as a possible model to explain the temperature and field dependence of the phases in manganites [6, 7] whereby ferromagnetic metallic and paramagnetic insulating phases coexist within a sample, and the macroscopic behaviour of the sample is governed by the dominant phase. The results presented in this paper support this model qualitatively.

Our research to date has focused on doping the ferromagnetic-insulating lanthanum manganite compound, $LaMnO_3$, with small concentrations of divalent or tetravalent atoms and observing how these impurities change the fundamental material properties. Measurements have been performed and reported on calcium-doped lanthanum manganite samples ($La_{1-x}Ca_xMnO_3$) with varying doping levels ($x = 0-0.5$) [8]. Likewise measurements on strontium- and lithium-doped lanthanum manganites [9, 10] have shown interesting and unexpected results.

This paper presents data collected on different classes of perovskite-structured manganites and manganite/cobaltites that were examined to characterise their ER, and to determine how varying dopant valences and ionic radii affects ER in each class.

2. Experimental method and sample compositions

We have developed an experimental procedure whereby four-terminal measurements are performed using a Keithley 2400 sourcemeter and customised data acquisition software. The terminals are arranged in a linear array and are formed by applying conductive silver paint between the sample surface and test leads. The sample is mounted with thermal paste on a copper sample mount, which is in turn thermally and mechanically attached to the cold
finger of a helium-based, closed-cycle, cooling system. This apparatus allows the sample temperature to be varied from ~380 K to 8 K. Data for each of these samples were collected while cooling the samples from 295 K to 10 K at a rate of 5 K/min, and then warming to room temperature at 5 K/min.

All sample compounds presented in this paper follow the general perovskite-type structure of the form $ABO_3$. The manganite compounds have Mn situated on the $B$-site (both Mn$^{3+}$ and Mn$^{4+}$ depending on the valence of the $A$-site), with partial-substituted La on the $A$-site, namely La$_{0.8}$Li$_{0.2}$MnO$_3$, La$_{0.67}$Sr$_{0.33}$MnO$_3$ and La$_{0.8}$Pb$_{0.2}$MnO$_3$. The manganite/cobaltite samples comprise of a fixed 1:1 ratio of Mn to Co on the $B$-site, with the $A$-site populated by Ho, Dy or Nd resulting in DyMn$_{0.5}$Co$_{0.5}$O$_3$, HoMn$_{0.5}$Co$_{0.5}$O$_3$ and NdMn$_{0.5}$Co$_{0.5}$O$_3$.

All samples were prepared via conventional solid-state reaction of precursors which were weighed, ground, pelletised, sintered in air at 1100 °C, reground, re-pelletised and sintered in air at 1150 °C, as described in ref. [11].

3. Results

Fig. 1 and Fig. 2 show the temperature and electric field dependence of ER in each sample. The ER is calculated using the formula:

$$ER^* = \frac{R(I_{\text{low}}) - R(I_{\text{high}})}{R(I_{\text{high}})}$$

Where $I_{\text{high}}$ is the maximum current applied to the sample and $I_{\text{low}}$ the current under investigation. The majority of the compounds show a significant (i.e. greater than 0.2 or 20%) level of ER.

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Fig. 1. ER vs. Temperature (K) vs. Current (A) plots for the manganite/cobaltite samples (clockwise from top left) NdMn$_{0.5}$Co$_{0.5}$O$_3$, HoMn$_{0.5}$Co$_{0.5}$O$_3$ and DyMn$_{0.5}$Co$_{0.5}$O$_3$. The vertical axis for each plot is chosen to be the same scale to allow direct comparison between the samples.
3.1 Manganite/Cobaltites

The ER from the three manganite/cobaltite compounds is presented in Fig. 1. All three compounds exhibit ER around room temperature, however this behaviour is not present in any of the manganite/cobaltite samples below 200 K.

The A-site was also substituted in these compounds. When lanthanum is substituted with an atom of approximately the same size, in this case neodymium (NdMn$_{0.5}$Co$_{0.5}$O$_3$), the sample still shows significant ER. However, if the lanthanum is substituted with a smaller atom, dysprosium and holmium in this case (Dy/HoMn$_{0.5}$Co$_{0.5}$O$_3$), only the holmium-doped sample displays ER. The Curie temperature, T$_C$, has been found to be close to 200 K for these manganite/cobaltite samples, and dependent on the radius of the A-site ion [12]. This in turn implies the sample is dominated by the ferromagnetic phase below 200 K. In a phase separation model, the ferromagnetic phase exhibits a linear voltage-current relationship [13] resulting in the absence of ER, as is the case in Fig 2. This is qualitative evidence for phase separation in these compounds.

![Fig. 2. ER vs. Temperature (K) vs. Current (A) plots for the manganite samples (clockwise from top left) La$_{0.8}$Pb$_{0.2}$MnO$_3$, La$_{0.67}$Sr$_{0.33}$MnO$_3$, and La$_{0.8}$Li$_{0.2}$MnO$_3$. The vertical axis for each plot is chosen to be the same scale to allow direct comparison between samples.](image-url)
3.2 Manganites

The strontium-doped sample, La$_{0.67}$Sr$_{0.33}$MnO$_3$, shows significant ER at temperatures below 100 K, but does not show appreciable ER above this temperature.

The lead-doped sample, La$_{0.8}$Pb$_{0.2}$MnO$_3$, is especially interesting as measurements on a single crystal sample of this compound show that ER is not intrinsic in a similarly doped sample [14], implying that the ER exhibited by this sample is not intrinsic to the compound.

The La$_{0.8}$Li$_{0.2}$MnO$_3$ sample shows an ER peak double the magnitude of any other compound presented in this paper. This is important for possible applications of ER in memory in switching devices, as a large ER denotes a considerable difference in sample resistance in high and low electric fields.

The La$_{0.8}$Li$_{0.2}$MnO$_3$ and La$_{0.8}$Pb$_{0.2}$MnO$_3$ samples are of considerable interest, as they exhibit ER below T$_C$, ~ 230 K for both samples [14, 15]. This implies that ER in these samples is not due to phase separation, and is most likely due to grain boundaries or porosity effects in the sample.

4. CONCLUSIONS

We have presented results on the ER of several perovskite-structured compounds. The manganite/cobaltite samples show evidence that ER in those compounds is most likely due to phase separation. In the manganite samples, the results for the Pb- and Li-doped manganite samples are of particular interest, as considerable ER is seen over the entire measured temperature range. This, coupled with data showing single-crystal La$_{0.8}$Pb$_{0.2}$MnO$_3$ does not display ER, implies that ER in polycrystalline samples could be much more strongly dependent on extrinsic factors such as grain size and boundary-layer thickness than on phase separation.

References