Application of Direct Stress Using a Multilayered Actuator to finetune Transport and Magnetic Properties

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Introduction

Manganite thin films are manganese oxides films derived of the form $AMnO_3$ which can be either metallic or insulating at room temperature depending on the composition. Manganite thin films have been intensely studied for the past several years for their changes in transport[1-3], electronic[2,4], and magnetic[1-4] properties with respect to temperature as well as the presence of intriguing magnetic properties[1-5]. These properties include colossal magnetoresistance (CMR), the metal-to-insulator transition (MIT), and electronic and magnetic phase separation. These properties are dependent on the mixed-valence of Mn, the perovskite structure of manganites, pictured in Figure 1, as well as the multiphase coexistence of the two competing phases; the ferromagnetic metallic phase (FMM) and the charge-ordered insulating (COI) antiferromagnetic phase[2,3].

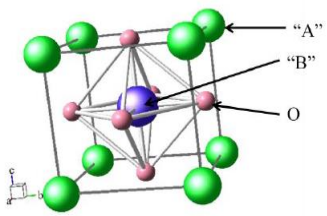

Figure 1 Model of the Mixed-valence Perovskite Structure of Manganites. The sites are labelled "A," "B," and "O" where "A" represents La, Pr, and Ca, "B" represents Mn and "O" represents O.

In manganites, the pseudocubic FMM phase is favored at lower temperatures for materials with greater average A-site cation radii. However, in hole doped manganites, the COI phase becomes more favorable as the A-site is substituted with smaller ions and the free energy of the COI phase becomes comparable to that of the FMM phase. Such is the case for the manganite $(La_{1-y}Pr_y)_{1-x}Ca_xMnO_3$ (LPCMO), a Pr doped manganite[2]. The presence of Pr introduces quenched disorder due to the ions La and Pr being of different sizes as well as long-range strain interactions. The result is micrometer scale phase separation of the FMM and COI phases and a decrease in the MIT temperature. This causes the first order transition to occur at lower temperatures than usual[12].

First order transitions are transformations in which the first derivative of the Gibbs free energy is discontinuous at the equilibrium transformation temperature[6]. An illustration of this is shown in Figure 2(a). Pictured are three curves of Resistivity vs temperature for $(La_{1-y}Pr_y)_{1-x}Ca_xMnO_3$ (x=0.33, y=0.4, 0.5, 0.6) thin films on Neodymium Gadolinium Oxide (NGO) substrates. At various temperatures for all three films there is a rise in resistivity of several orders of magnitude in the warming of the films and a similar fall in resistivity for the cooling run. This is the MIT and is characteristic of manganites. The MIT in manganites characterizes a change in various transport properties of the material as well as changes in the crystal structure and electronic properties[3,5].

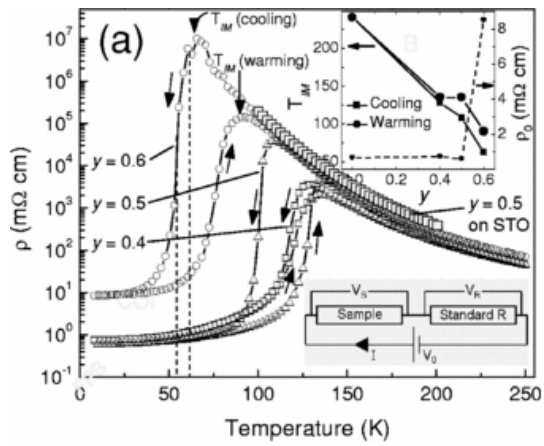

Figure 2 Resistivity vs Temperature for LPCMO ((x=0.33, y=0.4, 0.5,0.6) measured by Dhakal et al[2]. The curves exhibit a hystersis loops and accurately show the MIT that occurs in manganites due to the FMM and COI competing phases.

The MIT confirms a change in the transport properties of manganites and application of an electric field, magnetic field, stress, or a combination of these factors can lead to the change in its magnetic properties[3,5]. The application of stress on LPCMO tunes the volume fraction of the FMM and COI phases in LPCMO via strain and corresponds to a change in the MIT[3,5]. There have been previous investigations of the transport properties as a function of substrate induced strain to investigate the role of long-range strain interactions and quenched disorder[1,2]. However, lattice mismatch due to the differences in unit cells and varying growth morphologies of the substrate and the film cause non-uniform strain[1]. Results obtained are indicative of epitaxial strain and

substrate choice and not a good means of testing strain via direct stress. To obtain a better result, a three-point bending measurement was adapted to apply direct stress and induce strain. Tosado et al.[1] obtained large changes in resistance with applied stress, showing that transport properties can be altered using applied direct stress[1]. Although, significant strain was obtained (~10^{-4}), such measurements are difficult to perform due to the brittle nature of oxide substrates. Singh et al.[3] investigated transport properties as a function of applied elastic stress using a four-point bending method. Although a more direct stress was obtained, this is a mechanical method and is not suitable for device fabrication or real-world application. An electronic (voltage) dependent applied stress would be more suitable, but has yet to be studied. Here, we use a multilayered actuator (MLA) composed of Lead Zirconate Titanate (PZT) to apply a direct stress to the film to investigate the effect of strain on an $(La_{1-y}Pr_y)_{1-x}Ca_xMnO_3$ (x=0.33, y=0.5) thin film. We obtained a strain of ~10^{-6}. The study shows that even such a small strain is sufficient to induce a shift in the MIT and perhaps alter magnetic properties and that electronic applications to apply stress are promising for future experiments and devices.

Methods

An LPCMO film (x=0.33, y=0.5) was grown on a 1 cm by 1 cm (110) NGO substrate using pulsed laser deposition. Indium contacts were placed on gold pads which were placed perpendicular to the length of the sample as shown in Figure 3(a). Figure 2 inset shows the schematic for the electrical transport measurements. Since, the film can achieve resistances as high as 1GΩ[2], a two-probe measurement was sufficient for our purposes[2]. For the electrical transport measurements, 1V was applied across the sample and a 99KOhm load resistance was used. The measurements were taken inside of a

helium dewar in which the temperature inside the dewar was varied from 50K to 150K. Manganites are sensitive to warming and cooling rates[3] so the sample was warmed and cool at a rate of 2K/min and a temperature accuracy of 0.2K. Direct stress was applied on the film using a PICMA Chip miniature MLA PL033 provided by PI ceramic. The MLA was composed of PZT and a surrounding insulating layer. The MLA was used for

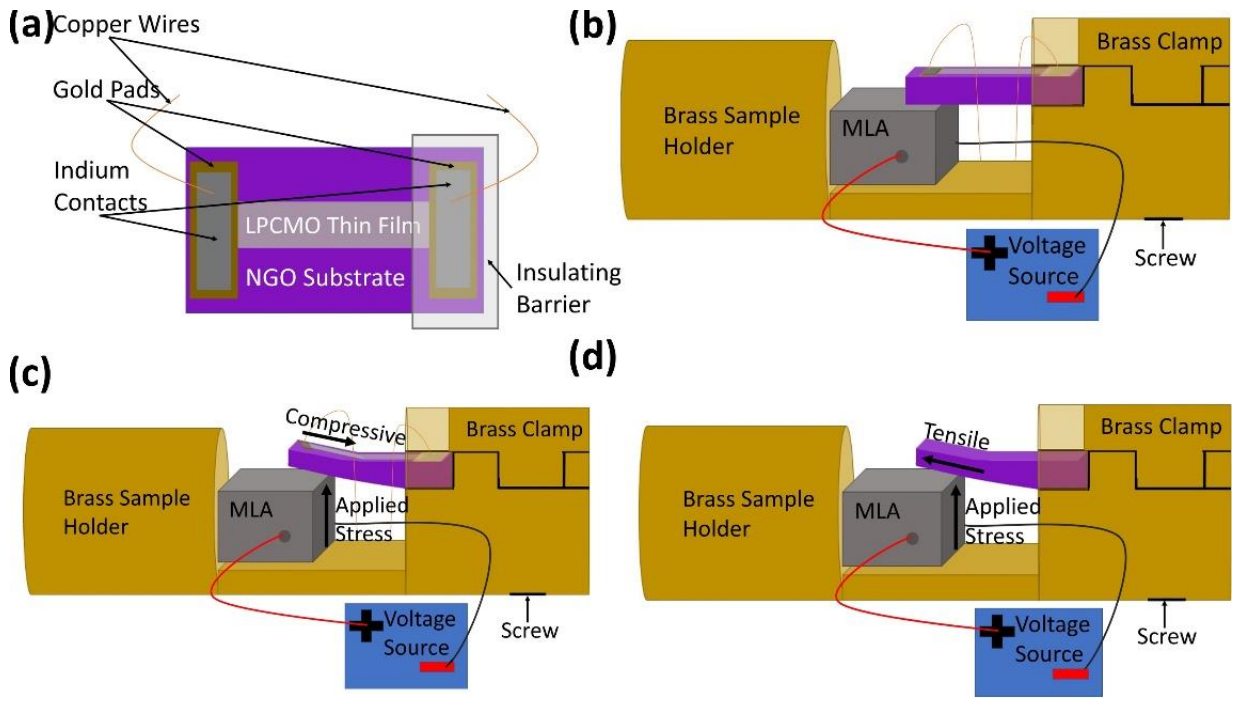

Figure 3 (a) Schematic of the sample. (b) Illustration of what the sample prior to voltage being applied to the MLA. The sample is flatly placed on the MLA. Measurements taken from this arrangement are "0" strain measurements. (c) The sample is facing up and voltage has been applied to the MLA, causing for the MLA to displace upwards. The result is compressive strain on the sample, detailed by the black arrow (d) The sample is facing up and voltage has been applied to the MLA, causing for the MLA to displace upwards. The result is tensile strain on the sample, detailed by the black arrow

its micrometer displacements and position resolution, functionality at cryogenic temperatures, and nonferromagnetic properties. In addition, 60V, 70V, and 80V were applied across the MLA which correspond to different applied stresses. Due to operation at cryogenic temperatures, we were not able to achieve the full displacement of the MLA at the corresponding voltages. We estimate that only about 20-40% of the displacement that would be achieved at room temperature was achieved during our measurements for

the 74 K to 140 K range. The sample and the MLA were secured in a brass sample holder. An adhesive was used to secure the MLA to the floor of the sample holder while the film was secured on the end furthest from the MLA using a clamp and screw as shown in figure 3(b). It was ensured that the MLA was in direct contact with the sample as the maximum displacement of the film was approximately 2μm. In Figure 3(a), an insulating layer of material was placed over one contact to avoid a short circuit due to contacts between the indium contact and the brass sample holder. Measurements were taken with the contacts facing upward, figure 3(c) and downward, figure 3(d) to apply both compressive and tensile stress, respectively.

To confirm the functionality of the method as well as estimate the maximum displacement of the MLA, an optical experiment was conducted. In Figure 4, the optical experiment is shown. Figure 4(a) depicts a laser being reflected off the LPCMO film onto an opaque screen. Figure 4(b) depicts that same laser reflected off the same

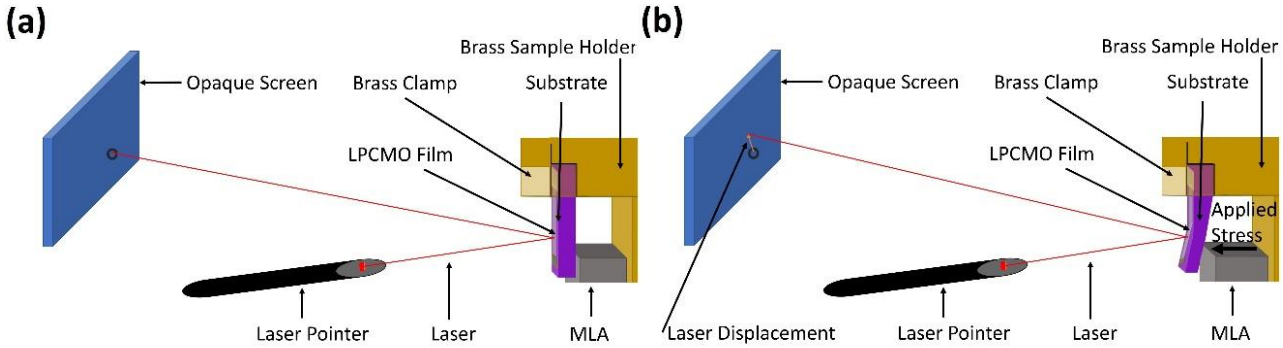

Figure 4 Schematic of the optical experiment conducted to test the MLA. a) Exhibits the position of the reflected laser before voltage is applied to the MLA. A black circle is drawn on the opaque screen to detail the reflected laser's position. b) The laser has changed position and an orange arrow is drawn to show the displacement of the reflected laser

LPCMO film, however this time 100V was applied to the MLA. If the method works, the film will bend and change the reflected angle which will result in a displacement of the laser's position on the opaque screen. We found the displacement of the laser spot to be approximately 1mm. We could estimate the film displacement since we knew the

distance between the film and the screen and determined the displacement of the MLA to be ≈ 2μm. We found the displacement of the laser spot to be approximately 1 mm. Clamps were used to prevent the movement of the sample holder and laser pointer.

Results

Figure 5 shows the resistance vs. temperature measurements for the LPCMO film in compressive strain, tensile strain, and no strain. A log scale of the resistance is used to highlight the MIT as well as the change in metal to insulator transition. The plots show sharp transitions in the resistance of the film, exhibiting the MIT in the warming and cooling of the sample. For the compressive strain measurements, the transition temperature during warming with no strain was 124 K and 102 K during cooling, resulting in a $\Delta T \sim 22$K. For the tensile strain measurements, the transition temperature during warming with no strain was 122 K and 102 K during cooling, resulting in a $\Delta T \sim$ 20K. Figure 5(a) displays the resistance vs temperature measurements for 0V, 60V, and 70V applied to the MLA when the sample was in tensile strain. We found no significant change in the MIT temperature for tensile strain at these voltages. Similarly, Figure 5(b) displays the resistance vs temperature measurements for 0V, 60V, and 70V applied to the MLA when the sample was in compressive strain. We found no significant change in the MIT temperature for compressive strain at these voltages. For 80V applied to the MLA, there was a small, but significant, change in the MIT temperature for both tensile and compressive strain during the warming run. In Figure 5(c), there is a 2K shift in the transition temperature in the warming run as the MIT temperature rises to 124 K from the previous value of 122 K when the sample is in tensile strain. Likewise, in Figure 5(d) there is a 2K increase in the warming run as the MIT temperature rises from 124K to

126K for compressive strain. However, on the cooling run, the transition temperature does not change for both compressive and tensile strain when 80V is applied to the MLA.

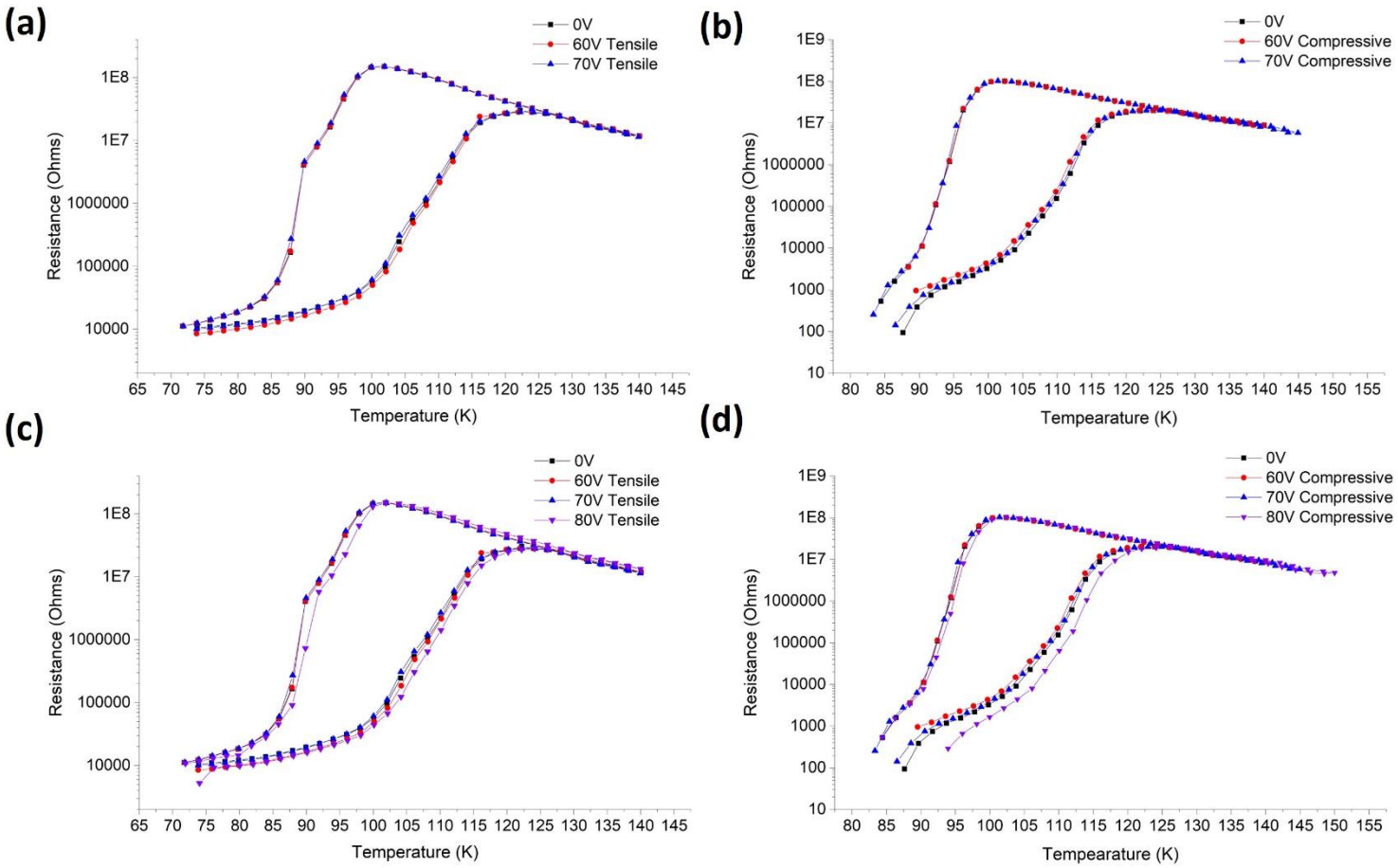

Figure 5 Resistance vs Temperature measurements for LPCMO (x=0.33, y=0.5). a) Resistance vs temperature measurements for 0V, 60V, and 70V applied to the LPCMO film when in tensile strain. b) Resistance vs temperature measurements for 0V, 60V, and 70V applied to the LPCMO film when in compressive strain. c) Resistance vs temperature measurements for 0V, 60V, 70V, and 80V applied to the LPCMO film when in tensile strain. d) Resistance vs temperature measurements for 0V, 60V, 70V, and 80V applied to the LPCMO film when in compressive strain.

We calculated the strain of the film at 80V applied to the MLA using by dividing the change in the length of the film by the original length. Using $\varepsilon = {\Delta L}/{L_\circ} = {xt}/{L_\circ^2}$ [1], we obtained a strain of 10^{-6}. "$L_\circ$" is the original length of the film, "x" was the displacement of the MLA, and "t" was the thickness of the film and substrate.

Discussion

A voltage dependent method of applying direct stress to alter and finetune transport and magnetic properties of manganites has not yet been studied. Using an MLA, we increased the MIT temperature by 2K in the warming run. In doing so, we have shown that minimal strain is required to induce change in the transport properties of manganites.

We induced a 2K increase in the MIT in the warming run when applying 80V to the MLA for compressive strain. This agrees with previous measurements done by Tosado et. al[1] and Singh et. al.[3]. However, we obtained no shift in the MIT temperature on the cooling run. This result does not agree with previous studies as Singh et. al.[3] and Tosado et. al[1] found that their shifts in the warming run equaled the shifts in the cooling run when both compressive and tensile stress was applied, resulting in no change in ΔT. This can be explained by the negative coefficient of thermal expansion for the MLA. As temperature decreases, the MLA contracts, resulting in non-uniform stress during the entirety of the experiment.

In addition, we induced a 2K shift in the MIT in the warming run when applying 80V to the MLA for tensile strain. This result does not agree with Tosado et. al[1] and Singh et al[3]. We suspect that other stresses could have been induced due to the design of the sample holder. Future work will improve upon the sample holder design.

We obtained strain of ~ 10^{-6} in the material, two orders of magnitude lower than previous studies done by Tosado et al[1]. This marks one of the advantages of using an MLA. In doing so, we found that minimal strain is required to induced a shift in MIT temperature.

In summary, the presence of the FMM and COI competing phases produces changes in the transport properties of $(La_{1-y}Pr_y)_{1-x}Ca_xMnO_3$ when the film is subjected to applied direct stress. The use of an MLA is a promising alternative to mechanical methods as we induced a shift in the MIT temperature in the warming run for compressive and tensile strain. However, no difference in the direction of the shift in the MIT was obtained. Our results will be instrumental in future methods in fine-tuning transport and magnetic properties of $(La_{1-y}Pr_y)_{1-x}Ca_xMnO_3$ through electronic dependent methods.

Acknowledgments

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