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Colossal piezoresistance in phase separated manganites

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Online at stacks.iop.org/JPhysCM/21/192203**Abstract**

We have measured the strain dependent transport properties of phase separated manganite thin films. We subjected $(\text{La}_{1-y}\text{Pr}_y)_{1-x}\text{Ca}_x\text{MnO}_3$ thin films grown on $\text{NdGaO}_3(110)$ substrates to direct external mechanical stress using a three-point beam bending method. The resultant change in resistance reveals a colossal piezoresistance (CPR) in manganites. Our experiments reveal that phase separation is a necessary but not sufficient condition for CPR. The maximum CPR is observed only when the phase boundaries are free to move in the fluid-like phase separated state. Our results show that both long-range strain interactions and quenched disorder play an important role in micrometer scale phase separation in manganites, albeit in different temperature ranges.

(Some figures in this article are in colour only in the electronic version)

The transport and magnetic properties of hole-doped manganites are determined by the competition between a pseudotetragonal charge-ordered insulating phase (COI) and a pseudocubic ferromagnetic metallic (FMM) phase [1]. In compounds such as $(\text{La}_{1-y}\text{Pr}_y)_{1-x}\text{Ca}_x\text{MnO}_3$ this competition leads to micrometer scale multiphase coexistence or phase separation [2]. The two leading scenarios which are possibly responsible for micrometer scale phase separation are the presence of quenched disorder in the vicinity of a first-order transition [3] and long-range strain interactions [4, 5]. In the first scenario the phase separation is static because the phase boundaries are pinned by the disorder sites [3]. In such a phase separated state the effect of strain at constant temperature should essentially be the sum of the strain effects on the pure FMM and COI phases since the phase boundaries are not free to move [4, 6]. In the long-range strain interaction scenario the difference in structure between the FMM and COI phases introduces the surface energy between the two phases i.e. the phase boundaries separate regions of different structures [4]. Such a situation commonly arises in first-order phase transitions between two solid phases with different structures and leads to martensitic phases [7]. The propagation

of the phase boundaries is stopped by the long-range strain interactions in the material [4]. These two scenarios are related because both require similar free energies for the competing FMM and COI phases. In one model the phase boundaries are pinned by the disorder whereas in the other, the change in structure at the phase boundaries can be considered similar to disorder. However, a clear distinction between the two models is the fact that while the quenched disorder is localized in space, the phase boundaries due to long-range strain interactions are free to move on the application of external mechanical stresses. Hence, the effect of strain on the transport properties of manganites should determine the actual mechanism of micrometer scale phase separation [4].

The effect of strain on manganites has been studied extensively by measuring the properties of manganite thin films grown on substrates with different amounts of lattice mismatch and also the thickness dependent properties of the thin films [8, 9]. Some unique experiments have also used a piezoelectric buffer layer grown between the substrate and the manganite enabling one to change the strain on the film using an electric field [6]. However, all such techniques may lead to non-uniform strain due to variation in the growth morphologies depending on the amount of lattice mismatch with the substrate and film thickness [8]. To get a quantitative measurement of

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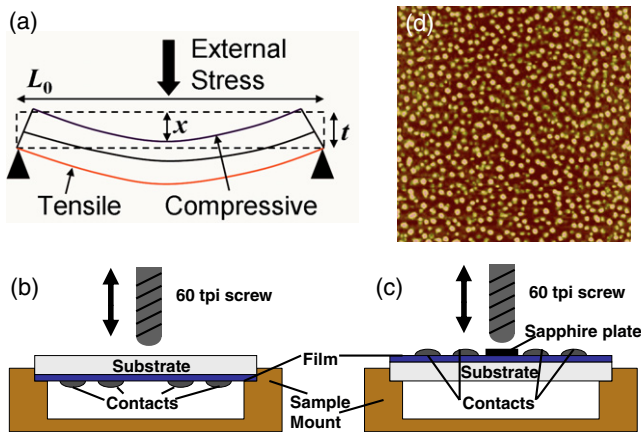


Figure 1. (a) Three-point beam bending technique for applying compressive and tensile strain to a thin film. The dashed box is the undistorted beam. (b) and (c) Schematic diagrams for the application tensile and compressive strain, respectively and four-probe resistance measurements. (d) A $5 \mu\text{m} \times 5 \mu\text{m}$ AFM image of a 30 nm LP5CMO thin film.

the strain effect, strain dependent measurements need to be performed on a single, well-characterized thin film. We present here a direct measurement of the strain effect using a three-point beam bending technique on manganite thin films. While such experiments have been performed on several materials including semiconductor nanowires and ferroelectrics [10], it is difficult to perform a three-point beam bending measurement on manganite thin films due to the relatively brittle oxide substrates. However, we have measured a significant strain effect in phase separated manganites even at low strains of $\sim 10^{-4}$.

We have grown thin films of $(\text{La}_{1-y}\text{Pr}_y)_{0.67}\text{Ca}_{0.33}\text{MnO}_3$ ($y = 0.5$) using pulsed laser deposition (PLD) [11]. The films were grown in an oxygen atmosphere of 420 mTorr on $\text{NdGaO}_3(110)$ (NGO) substrates kept at 820°C . All the films described in this letter are 30 nm thick and were grown at a rate of about 0.05 nm s^{-1} . The target was ablated at a laser energy density of $1.0 \pm 0.2 \text{ J cm}^{-2}$. These growth conditions were optimized to obtain an insulator-to-metal transition temperature while cooling, T_{IMC} , close to that observed in bulk compounds of similar composition and the minimum transition width at T_{IMC} . Such an optimization is crucial for measuring the effect of strain on $(\text{La}_{1-y}\text{Pr}_y)_{0.67}\text{Ca}_{0.33}\text{MnO}_3$, since the properties of thin films of this compound vary markedly depending on the growth conditions. Standard θ - 2θ x-ray diffraction data show that the films are epitaxial and of a single chemical phase.

Direct external stress was applied to the thin films using a three-point beam bending technique. In such measurements the substrate and thin film system is treated like a classical beam which is supported at the edges and the external stress is applied to the center of the beam as shown in figure 1(a). For this arrangement, the strain at the surface of the beam $\epsilon = \Delta L/L_0 \approx xt/L_0^2$, where ΔL is the change in length at the surface of the beam due to the small displacement x of the beam center. L_0 and t are the original length and thickness of the beam, respectively. The stress is applied using a 60 tpi screw which can be turned from outside the

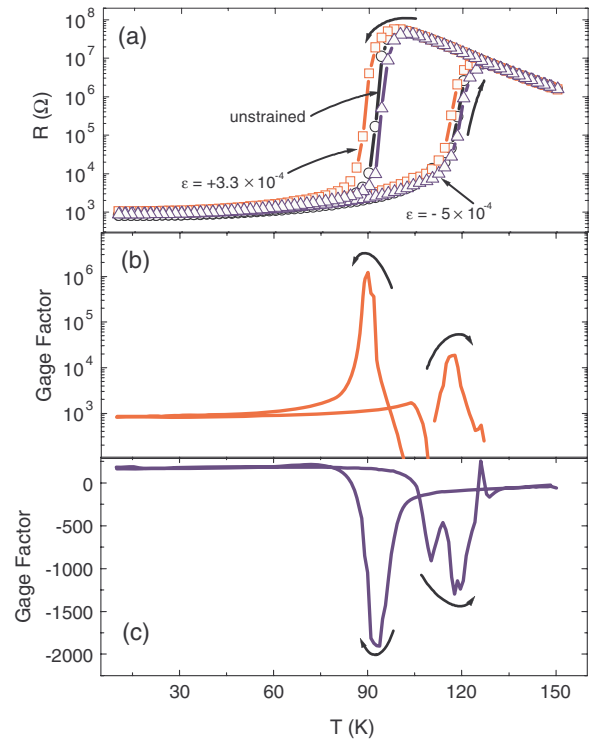


Figure 2. (a) Effect of tensile (positive) and compressive (negative) strain on the resistance versus temperature characteristics of an LP5CMO thin film. (b) and (c) Temperature dependence of the gage factor under tensile and compressive strain, respectively.

helium dewar using a worm gear, which allows us to move the screw with a precision of about $1 \mu\text{m}$. Both compressive and tensile strain can be applied to the thin film by using the geometries shown in figures 1(b) and (c). In the ideal case the resultant strain in this arrangement is uniaxial. A $5 \mu\text{m} \times 5 \mu\text{m}$ atomic force microscope (AFM) image of a 30 nm $(\text{La}_{0.5}\text{Pr}_{0.5})_{0.67}\text{Ca}_{0.33}\text{MnO}_3$ (LP5CMO) thin film is shown in figure 1(d). The roughness of the film is about 1.8 nm. We have observed optimal resistance versus temperature (R - T) characteristics for such growth morphology.

Figure 2(a) shows the R - T characteristics of an LP5CMO thin film. The external stress was applied at 20 K and then the R - T curves were recorded while warming and then cooling the sample. T_{IMC} shifts to a lower temperature on the application of tensile strain and to a higher temperature under compressive strain. We define $\Delta R/R_0$ as the piezoresistance and compute the quantity $(\Delta R/R_0)/(\Delta L/L_0)$ which is the standard definition of the gage factor for strain gages. The gage factor versus the T curve for tensile strain is shown in figure 2(b) and for compressive strain in figure 2(c) and it shows that the change in resistance is maximum near the transition region. Magnetic force microscopy imaging has shown that the different phases present in an LP5CMO thin film display a fluid-like behavior near the transition region [12]. Transport measurements have also picked up the presence of a fluid phase separated (FPS) state near the transition temperature [11]. Hence, we believe that the magnitude of piezoresistance is related to the presence of this fluid phase separation. Figures 2(b) and (c) also show that the piezoresistance is negligible at higher temperatures which

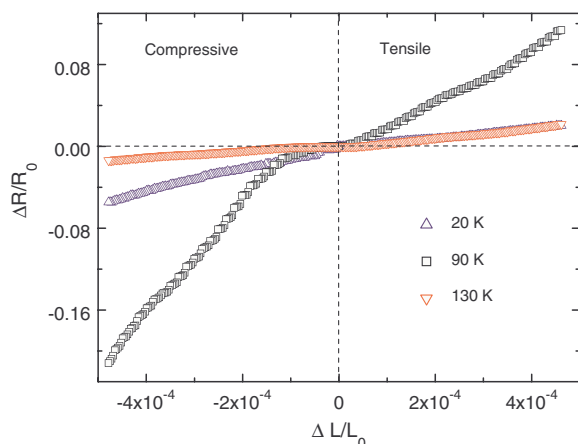


Figure 3. Variation of resistance as a function of strain at three different temperatures.

shows that phase coexistence is essential for piezoresistance in manganites. However, even at low temperatures the piezoresistance drops by about 3 orders of magnitude, although the material is still phase separated (figure 2(b)). Hence, phase separation is a necessary but not sufficient condition for colossal piezoresistance (CPR). Using transport and magnetization measurements it has been shown that at low temperatures $(La_{1-y}Pr_y)_{1-x}Ca_xMnO_3$ freezes into a glassy, static phase separated (SPS) state due to the quenched disorder present in the sample [1, 11, 13]. Thus the suppression of piezoresistance at low temperatures suggests that the fluid-like behavior of the different phases is essential for CPR. In addition, since the SPS state is formed at low temperatures, the gage factor is lower for the warming cycle compared to the cooling cycle for both tensile and compressive strain.

To further confirm the temperature dependence of piezoresistance in manganites we measured the effect of strain at certain fixed temperatures. Figure 3 shows the $\Delta R/R_0$ versus ϵ (strain) curves of the thin film at three different temperatures in the cooling cycle. At 130 K the film is predominantly in the paramagnetic insulating (PMI) phase and hence displays a small but measurable piezoresistance. At 90 K the film is in the FPS state [11, 14] and the piezoresistance reaches its maximum value. At 20 K the film reaches the SPS state [11] and the piezoresistance drops back to values similar to that observed at 130 K. The behavior of the curves at different temperatures confirms the correlation of fluid phase separation with larger values of piezoresistance. However, the value of the piezoresistance even at 90 K never reaches the values observed in figure 2 where the temperature was changed at a constant strain. This difference suggests that a static strain applied at high temperatures suppresses the formation of a particular phase whereas if that phase has already been nucleated, the application of strain at constant temperature is less effective in removing it. Another remarkable fact about these curves is that they are reversible unlike the effect of an electric field previously observed in $(La_{0.4}Pr_{0.6})_{0.67}Ca_{0.33}MnO_3$ [11]. The reversibility suggests that the motion of the phase boundaries in the FPS state is reversible, at least for the amount of strain applied in this

experiment. Such reversible motion of the phase boundaries suggests that they are not pinned by the quenched disorder present in the material. It is possible that higher amounts of strain would drive the phase boundaries into an irreversible region but we were unable to reach such strain values without breaking the sample. We are currently measuring samples in which disorder is introduced by Ar-ion bombardment of the thin films.

From both figures 2 and 3 we conclude that the sign of the piezoresistance depends on the sign of the strain i.e. the piezoresistance is an odd function of strain. This behavior is expected due to the lattice mismatch between the NGO substrate and LP5CMO. The in-plane lattice constant of the NGO (110) plane is about 3.86 Å (with a small in-plane anisotropy of about 0.01 Å) while the cubic FMM phase of LPCMO has a lattice parameter of 3.84 Å [15]. Hence, compressive strain reduces the in-plane lattice parameter of NGO and brings it closer to the lattice parameter of the FMM phase and vice versa. Therefore compressive strain leads to an enhancement of the FMM phase and a reduction of the sample resistance while tensile strain has the opposite effect.

In summary, the application of direct external stress and the resultant strain has a profound effect on the transport properties of phase separated manganites due to the difference in the structures of the FMM and COI phases. The effect of strain described above elucidates the underlying mechanism of phase separation in manganites. In the low temperature SPS state the gage factor has a smaller value compared to its value in the intermediate temperature FPS state. Hence, we conclude that while in the SPS state quenched disorder plays a major role in the formation of a phase separated state, in the FPS state long-range strain interactions play a dominant role.

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References

- [1] Sharma P A, Kim S B, Koo T Y, Guha S and Cheong S-W 2005 *Phys. Rev. B* **71** 224416
- [2] Uehara M, Mori S, Chen C H and Cheong S-W 1999 *Nature* **399** 560
- [3] Dagotto E 2005 *Science* **309** 257
- [4] Ahn K H, Lookman T and Bishop A R 2004 *Nature* **428** 401
- [5] Sartbaeva A, Wells S A, Thorpe M F, Boin E S and Billinge S J 2006 *Phys. Rev. Lett.* **97** 065501
- [6] Thiele C, Dörr K, Schultz L, Beyreuther E and Lin W-M 2005 *Appl. Phys. Lett.* **87** 162512
- [7] Podzorov V, Kim B G, Kiryukhin V, Gershenson M E and Cheong S-W 2001 *Phys. Rev. B* **64** 140406(R)
- [8] Biswas A, Rajeswari M, Srivastava R C, Li Y H, Venkatesan T, Greene R L and Millis A J 2000 *Phys. Rev. B* **61** 9665
- [9] Petit M, Rajeswari M, Biswas A, Greene R L, Venkatesan T and Martínez-Miranda L J 2005 *J. Appl. Phys.* **97** 093512
- [10] Zubko P, Catalan G, Buckley A, Welche P R L and Scott J F 2007 *Phys. Rev. Lett.* **99** 167601
- [11] Dhakal T, Tosado J and Biswas A 2007 *Phys. Rev. B* **75** 092404
- [12] Zhang L, Israel C, Biswas A, Greene R L and de Lozanne A 2002 *Science* **298** 805
- [13] Ghivelder L and Parisi F 2005 *Phys. Rev. B* **71** 184425
- [14] Milward G C, Caldern M J and Littlewood P B 2005 *Nature* **433** 607
- [15] Gillaspie D, Ma J X, Zhai H-Y and Ward T Z 2006 *J. Appl. Phys.* **99** 08S901