Nonbolometric photoresponse in $(La,Pr)_{0.67}Ca_{0.33}MnO_3$ thin films

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We have studied light-induced resistance changes (photoresponse) in the colossal magnetoresistive manganite material $(La,Pr)_{0.67}Ca_{0.33}MnO_3$. The metal-insulator transition in this material is understood to be driven by the percolation of the metallic channel in an inhomogeneous matrix of insulating and metallic components. Our experiments reveal a nonthermal (nonbolometric) component of the light-induced resistance change, in addition to the expected resistance change related to heating (bolometric effect). This nonthermal component is seen only in the metal-insulator transition region. Our results suggest that this component may be associated with the light-induced resistance decrease in the insulating regions through an electronic mechanism. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2168687]

Recent investigations of colossal magnetoresistive (CMR) manganite materials have revealed the key role of intrinsic phase separation and the resulting multiphase coexistence in the physics of several manganite systems.[1–5] One such material that shows the remarkable effects of such multiphase coexistence is $(La_{1-y}Pr_y)_{1-x}Ca_xMnO_3$ for a range of cationic compositions.[2,3] Multiphase coexistence in this material is understood as arising from the local strains induced by the ionic size differences between the La and Pr ions that occupy similar sites in the perovskite unit cell. Due to such strains, regions that are insulating and charge ordered are found to coexist with ferromagnetic metallic regions close to the ferromagnetic transition temperature. As the temperature is lowered, the metallic regions grow at the expense of the insulating material and a very steep drop in resistance is observed when the metallic regions form a percolative path for charge transport.

CMR manganites have been explored for potential bolometric infrared detector applications,[7–9] this functionality being associated with the pronounced resistivity changes with temperature accompanying the insulator-metal transition. In this context, percolative transitions offer the advantage of the very steep temperature dependence of resistivity in the percolative region (see Fig. 1), although the presence of hysteresis is expected to be a hurdle in actual practical applications. Another issue relevant to the bolometric applications is the possible occurrence of resistance changes accompanying nonthermal effects of radiation. Such nonthermal effects, in general, arise from radiation-induced electronic changes in the material—examples being photoconductivity in semiconductors and the radiation response of Josephson weak links in superconductors.[10] In the present context of mixed phase manganite systems, this issue is particularly relevant, given the fact the charge ordered state is known to be susceptible to radiation over a wide range of wavelengths.[11–13] Nonthermal effects, when present, are also valuable in terms of understanding the physics of the metal-insulator transitions. We have therefore studied the radiation-induced resistance changes in the mixed phase manganite system $(LaPr)_{0.67}Ca_{0.33}MnO_3$ (LPCMO).

The study presented here was done on epitaxial thin films grown on (100)-oriented $LaAlO_3$ substrates by Pulsed Laser Deposition (PLD).[7] The deposition conditions were optimized to obtain the metal-insulator transition close to the expected value based on the reported studies in bulk.[6] X-ray diffraction studies indicated the films to be single phase and in-plane aligned. Typical rocking curves show a full width at

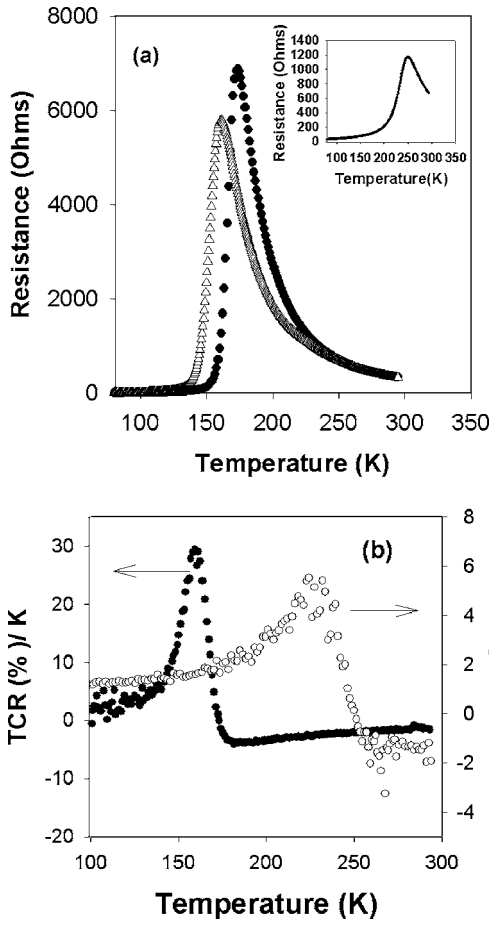

FIG. 1. A comparison of the insulator-metal transition in LCMO and LPCMO-1. (a) Resistance versus temperature data of LPCMO-1 obtained during cooling (●) and warming (△), showing the thermal hysteresis. Inset shows resistance versus temperature data of LCMO. (b) Temperature-coefficient of resistance of LCMO (○) and LPCMO-1 (●).

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half-maximum (FWHM) of ~0.2°, indicating good crystallinity. Electrical resistance was measured by the standard four-probe technique. Photoresponse measurements were done in an optical cryostat using phase-sensitive detection using a lock-in amplifier. The samples were irradiated by square pulses of visible light from a Laser diode (632 nm), generated by passing the light through a mechanical chopper. The reference signal for the lock-in detection was obtained from the chopper to facilitate phase-sensitive detection. Typical modulation frequency in our experiments was ~1 KHz.

A brief discussion of the photoresponse mechanism is in order in the context of the results that follow. In a simple bolometric response, the modulated energy input (ΔQ) from the incident light causes temperature modulations (ΔT) in the sample, which in turn cause resistance modulations (ΔR). In the presence of the dc bias current (I_B), these resistance modulations manifest as voltage modulations (bolometric photoresponse), given by

$$(\Delta V)_{\text{bolometric}} = I_B(dR/dT)\Delta T. \quad (1)$$

Thus the temperature dependence of the bolometric response may be expected to closely mimic that of dR/dT, barring any strong temperature dependence of the temperature modulation ΔT. This temperature modulation is governed by the thermal properties of the sample and the various thermal links involved in the heat transfer process.

It is important to note that in an actual experiment the observed photoresponse could have sources other than the simple bolometric mechanism outlined above. Light-induced electronic changes, if present, could cause such a "nonbolometric" response due to concomitant resistance changes that are decoupled from the thermal effect. In such cases, the observed photoresponse is expected to be a superposition of the two independent terms as

$$(\Delta V)_{\text{total}} = (\Delta V)_{\text{bolometric}} + (\Delta V)_{\text{nonbolometric}}. \quad (2)$$

Since the temperature dependence of the bolometric term is expected to be similar to that of dR/dT (assuming no strong temperature dependence of thermal properties), it follows that the measured photoresponse whose temperature dependence deviates significantly from that of dR/dT is indicative of the presence of nonbolometric mechanisms.

Figure 1 shows a typical resistivity versus temperature data of films with the composition $La_{0.37}Pr_{0.33}Ca_{0.33}MnO_3$ (LPCMO-1) as compared to its counterpart without Pr, which is $La_{0.67}Ca_{0.33}MnO_3$ (LCMO). The data in Fig. 1(a) clearly shows the hysteresis and steep resistivity changes associated with the percolative transition resulting from the Pr-induced phase coexistence. The comparison of the temperature coefficient of resistance (TCR) of the two samples [Fig. 1(b)] underscores the same point.

In Fig. 2 we show the photoresponse in LCMO as compared with dR/dT. This data, in agreement with previously published results,[14] clearly conforms to the presence of a purely bolometric photoresponse in this material. Substituting the measured photoresponse and dR/dT and the known bias current value in Eq. (1), we obtain an estimate of the temperature modulation $\Delta T \sim 2$ mK.

We now present the photoresponse and dR/dT data of the Pr-substituted film LPCMO-1 in Fig. 3(a). In marked contrast to the LCMO data, it is clear that there is a striking deviation of the temperature dependence of the photoresponse from that of the dR/dT in the middle of the resistivity

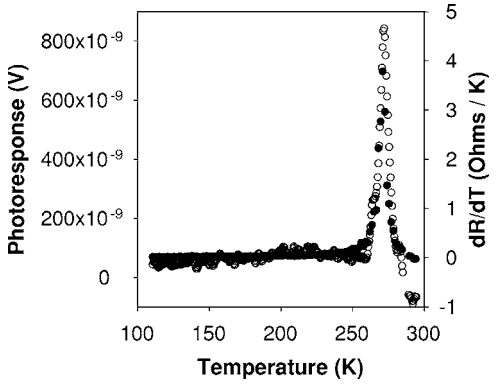

FIG. 2. Photoresponse in LCMO (●) as compared to dR/dT (○).

transition region. The most noteworthy feature here is the fact that there is a temperature range (165–180 K) where the sign of the measured photoresponse is opposite to what is expected for the bolometric response. In this temperature range where dR/dT has a positive sign, the bolometric effect should cause the sample resistance to increase due to the light-induced heating. However, the measured photoresponse is negative, i.e., the resistance decreases in the presence of light. This sign anomaly clearly indicates the presence of a photoresponse mechanism that competes with the bolometric response, as represented by the second term in Eq. (2). The fact that this additional term is negative implies a light-induced decrease in the resistance. A question may be raised at this point as to whether the observed "nonbolometric" characteristics could be related to possible anomalies in the thermal properties of LPCMO near the transition region.

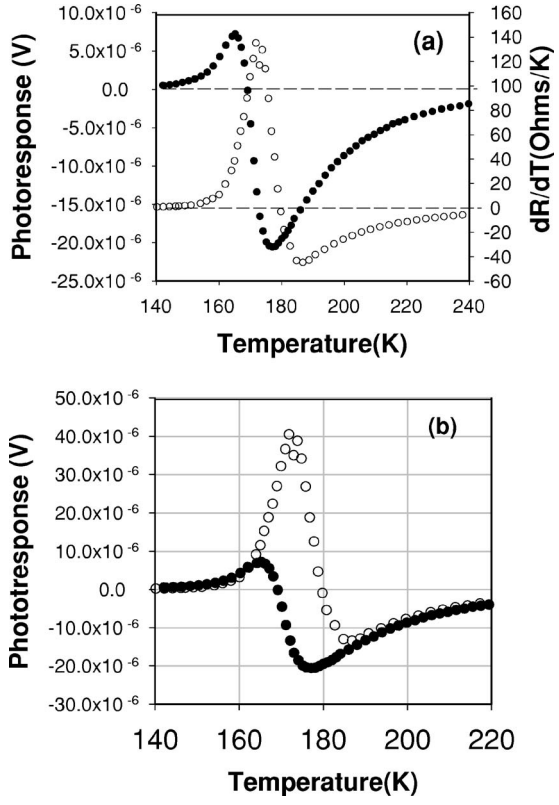

FIG. 3. Photoresponse in LPCMO-1. (a) Measured photoresponse (●) as compared to dR/dT (○). (b) Measured photoresponse (●) as compared to the expected bolometric photoresponse (○).

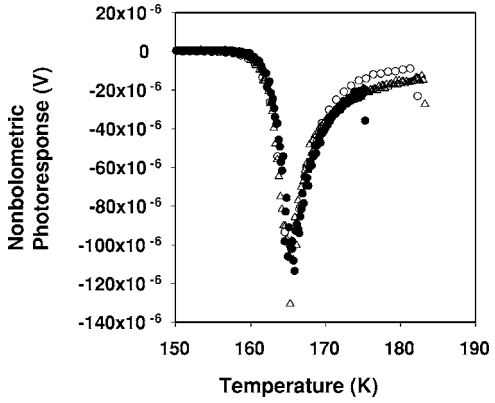

FIG. 4. Temperature dependence of the nonbolometric component of the photoresponse of LPCMO-1 measured under various experimental conditions: heating rate 0.3 K/min (●); heating rate 0.5 K/min (△); temperature stabilized (○).

Such a scenario cannot cause the sample resistance to decrease in the presence of light.

We have applied the simple model of Eq. (2) to isolate the nonbolometric component in order to see its temperature dependence. While the actual temperature modulation is not known, we assume it to be close to the ΔT calculated using the bolometric response of LCMO. Under our experimental conditions, ΔT would be determined by the thermal conductance between the substrate and the cryostat heat sink, together with the heat capacity of the substrate (the film thermal mass being very small). Both these quantities can be reasonably assumed to be the same for LCMO and LPCMO samples. In Fig. 3(b) we show the expected bolometric photoresponse in LPCMO-1 calculated using the ΔT thus obtained. Subtracting this bolometric component from the observed total photoresponse gives us the nonbolometric component (Fig. 4). We have studied the dependence of the observed photoresponse on the dynamics of the heating and cooling procedures during the experiment. As the data in Fig. 4 shows, the measured photoresponse does not depend on the heating and/or cooling rates or on whether the data is acquired during a temperature sweep or when held at a fixed temperature.

In considering the origin of the observed nonbolometric photoresponse in samples with a higher Pr content, we have investigated its possible role of resistance hysteresis. As can be seen from the resistivity data in Fig. 1, hysteresis extends over a broad temperature region (140 K–220 K) while the nonbolometric response we observe is limited to a narrower temperature range (165–180 K) close to the percolative transition. Also, our observation that the nonbolometric behavior is independent of the experiment dynamics (see Fig. 4) is not consistent with hysteresis being its cause. We believe that these observations, together with the fact that the actual temperature excursions induced by the modulated radiation is very small ($\sim$mK), rule out hysteresis as the cause of the nonbolometric behavior.

We hypothesize that the observed nonbolometric photoresponse may be attributed to light-induced resistance changes mediated by electronic mechanisms. As mentioned earlier, Pr doping is known to introduce the coexistence of charge ordered insulating and ferromagnetic metallic regions within the material. It is known that charge ordering in several manganite systems, including ($Pr_{1-x}Ca_xMnO_3$), can be destroyed by radiation over a broad wavelength range,[11–13] thus promoting metallicity. However, if the observed nonthermal effect was caused by the light-induced destruction of charge ordering, one would expect the light-induced resistance decrease to persist at lower temperatures, contrary to our observation that it is confined to the percolative transition region. In this context, it is interesting to note that previous work[6] has suggested that a second insulating phase (which is not charge ordered) may be responsible for the percolative transition. This suggestion is based on the fact that x-ray scattering studies do not show a reduction in the charge ordered component in proportion to the growth of the metallic component as temperature is lowered in the percolation temperature range. While the nature of the second insulating phase is not known, it is hypothesized that it could be insulating 'bottlenecks' in the metallic matrix, which turn metallic as the temperature is lowered. Such insulating bottlenecks have been suggested to be associated with the "martensitic strain" present at the boundaries of charge ordered and metallic regions that drive part of the metallic region to an insulating state. The nonbolometric photoresponse we observe may arise if light causes electronic changes in these insulating regions, lowering their resistance.

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