Large photoinduced conductivity reduction in thin films of metallic ferromagnetic manganites

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(Received 25 April 2011; accepted 2 November 2011; published online 29 November 2011)

This paper reports on a study of photoinduced resistivity changes in thin films of ferromagnetic metallic (FMM) manganites. We have observed a significant increase of resistance in $La_{0.7}Ba_{0.3}MnO_3$ thin film under continuous wave argon ion laser illumination presumably associated with photoinduced demagnetization. Strong dependence of resistance on thermal/illumination history is consistent with the coexistence of two phases: ferromagnetic metallic phase and photoinduced less conductive phase. © *2011 American Institute of Physics*. [doi:10.1063/1.3665039]

Doped rare-earth manganites have a very rich phase diagram due to strong correlation of different degrees of freedom.[1–3] Moreover, these materials can cross phase boundaries under application of different external fields and can support phase separation. Illumination with x-ray or visible light can destroy the charge ordering (CO) in manganites leading to insulator-to-metal transition.[4,5] The origin of the photoinduced transitions in manganites is not well understood yet and remains a subject of continuous research[6–10] not only due to interesting fundamental physics but also because of potential applications in photonic and opto-electronic devices. The melting of the charge ordering by visible light was studied in some detail in manganites of different compositions.[4–6,9–12] There are fewer reports on effects of light on manganites with ferromagnetic metallic (FMM) ground state. Increase of absorption in FMM manganite in pump-probe experiments[13,14] was explained by significant change in spin alignment due to the photoinjection of e_g carriers (photoinduced demagnetization).

In this paper, we report the effects of continuous wave visible light illumination on manganites with FMM ground state. We observed a significant photoinduced increase of resistance in the FMM state in a wide temperature region consistent with photoinduced demagnetization and studied thermal/illumination history dependence of this effect and its dynamics.

Thin films of $La_{0.7}Ba_{0.3}MnO_3$ and $La_{0.7}Sr_{0.3}MnO_3$ were grown by the pulsed laser deposition (PLD) technique. The film thicknesses were around 40 nm. $La_{0.7}Ba_{0.3}MnO_3$ and $La_{0.7}Sr_{0.3}MnO_3$ were grown on (110) oriented $NdGaO_3$ (NGO), (100) oriented $LaAlO_3$ (LAO), and (100) oriented $SrTiO_3$ (STO) substrates. Deposition conditions are described elsewhere.[12] X-ray diffraction patterns indicate that the films are epitaxial and single phase. The 0.1° width of rocking curves shows good crystallinity of these films.

Resistivity was measured by a standard four-probe technique (Fig. 2, inset). The distance between voltage contacts was approximately 0.3 mm. A continuous wave argon ion laser (150 mW in the multi-line mode) and He-Ne laser (24 mW) were used for sample illumination. The space between voltage contacts was completely covered with a laser beam (Fig. 2, inset) of approximate diameter 3 mm.

Temperature dependence of resistance of $La_{0.7}Ba_{0.3}MnO_3$ thin film grown on NGO is shown in Fig. 1. The temperature of metal-insulator transition for this sample is around room temperature. Therefore, the sample is in FMM state in the experimental temperature range. When the sample is cooled down to low temperature and then illuminated with full power of argon-ion laser light, the resistance increased drastically. The resistance taken under warming with illumination (Fig. 1, thick black curve) is significantly higher than resistance without illumination in the whole temperature range up to room temperature, where light induced changes become insignificant.

We interpret these changes as destruction of ferromagnetic (FM) ordering by light, since conductivity and spin alignment of Mn ions in manganites are mutually interdependent through the double-exchange mechanism. Previously, photoinduced increase of the resistivity under illumination was observed in the narrow temperature range in the hysteresis region near the COI-FMM transition in $Pr_{0.55}(Ca_{0.75}Sr_{0.25})_{0.45}MnO_3$,[7] where the free energies of COI and FMM states are similar. In our case, a significant increase of resistance due to destruction (or partial destruction) of the FM ordering occurs over a wide temperature range, where the $La_{0.7}Ba_{0.3}MnO_3$ is in FMM state and the phase boundary with insulating phase is far away. Absorption and transmission data from the pump-probe experiments also indicated a reduction of FM correlation between t_{2g} spins in the FM state under illumination.[13,14]

When the sample is cooled under illumination (Fig. 1, dashed line), the resistivity differs insignificantly from the $R(T)$ taken without illumination (Fig. 1, gray line) for temperatures above ~160 K. At around 160 K, as the sample is cooled under illumination, the resistivity increases abruptly and has significantly larger resistance, than resistance measured without illumination. In contrast, $R(T)$ curve taken under illumination but with different thermal/illumination history (warming with illumination after cooling without illumination), Fig. 1 solid black curve, has significantly larger resistance values. Such drastic dependence of the conductivity on history can be explained as follows. Illumination creates regions of magnetically disordered photoinduced resistive

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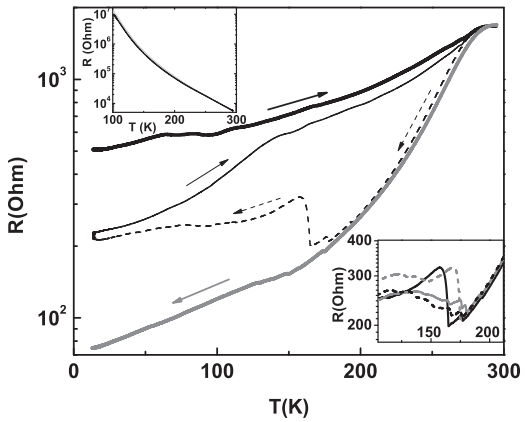

FIG. 1. Temperature dependence of the resistance of the $La_{0.7}Ba_{0.3}MnO_3$ thin film on NGO: thick solid gray line—cooling without illumination; thick solid black line—warming with illumination after cooling without illumination; thin dashed line—cooling with illumination; thin solid line—warming with illumination after cooling with illumination. Cooling and warming is shown by arrows for each curve. Upper inset shows temperature dependence of the resistance of the $Bi_{0.75}Sr_{0.25}MnO_3$ thin film: gray line—cooling without illumination; black line—warming with illumination. Lower inset shows different runs of resistance taken on cooling with illumination.

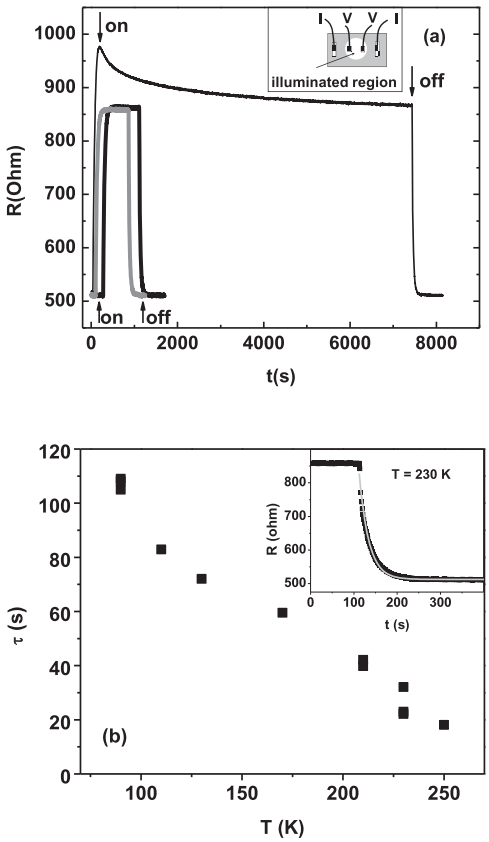

FIG. 2. (a) Time dependence of the resistance of the $La_{0.7}Ba_{0.3}MnO_3$ thin film when the sample illumination was switched on and off at $T = 230$ K: thin black line—initial illumination at this temperature; thick gray and black lines—subsequent runs at this temperature. The inset schematically shows experimental current and voltage contact configuration and illuminated region. (b) Temperature dependence of the time constants of the process of transition to original state after illumination. The data were taken on warming. The inset shows a fit (gray line) of the time dependence of the resistance after the laser illumination was switched off to the exponential decay $\Delta R \propto \exp(-t/\tau)$ at $T = 230$ K.

(PIR) phase. As the sample is cooled under illumination from room temperature, isolated regions of this PIR phase start to appear in the FMM matrix, which does not affect the overall resistivity, since there is percolation through the FMM phase. Although optical conductivity of FMM manganites increases with decrease of temperature at low frequencies, at frequencies of Ar ion laser light (around 2.5 eV), optical conductivity decreases as temperature decreases.[15,16] Thus, the material becomes more transparent for visible light with decrease in temperature. This promotes growth of the PIR phase, which blocks some of the percolative paths in the FMM matrix leading to a jump-like transition to higher resistance, when the sample is cooled under illumination. Such jump-like changes of resistance, which occurs at around 160 K, when sample is cooled down under illumination, are not exactly reproducible from run to run (Fig. 1, lower inset). This indicates the percolative nature of this process, which is also found in insulator-to-metal transition in other manganites.[17–19]

When the sample is illuminated at low temperature after cooling down without illumination, the resistivity increases by almost one order of magnitude and is significantly larger than resistivity after cooling under illumination. This difference could be explained by different energy balance of FMM and PIR phase at higher and lower temperatures. Let us consider a model with two local energy minima, where at higher temperature under illumination, the FMM phase occupies a deeper local energy minimum than the PIR phase and vice versa at low temperatures. Then, when the system is cooled under illumination, it is energetically preferable to remain in the conducting state, even though at low temperature, the FMM state under illumination does not occupy the deepest energy minimum. However, when the system is illuminated at low temperature, it is energetically preferable to occupy the deepest energy minimum corresponding to the PIR phase. Thin films of $La_{0.7}Sr_{0.3}MnO_3$ grown on LAO and NGO exhibit qualitatively similar behavior.

It is important to address issues of heating by laser light in our experiment. As the sample is cooled under illumination, its resistivity is almost identical to the resistivity taken while cooling without illumination down to approximately 160 K (Fig. 1). If we were to assume that the increase of the resistivity under illumination for this path is due to heating only, the change of resistance at 200 K would correspond to heating by 2 K, which cannot explain the large photoinduced increase of resistivity in this temperature region observed on warming with illumination. This is a very straightforward proof of the fact that in our experimental setup, the heating due to laser illumination is small in this temperature range. Moreover, a strongly hysteretic and path dependent R(T) taken under illumination (dashed, thin, and thick solid black curves in Fig. 1 differ significantly, while the same laser power was delivered) proves that observed photoinduced effects are not artifacts of heating, but is an intrinsic property of this material. At lower temperatures, the heating effects due to illumination could be larger. However, as discussed above, this does not affect our main experimental result.

To confirm absence of significant heating in our experimental arrangement, R(T) of $Bi_{0.75}Sr_{0.25}MnO_3$ manganite thin film was measured with and without illumination (Fig. 1, upper inset). If we assume that change in resistance is due to heating only, than the heating does not exceed 3 K in the temperature range above 100 K.

Dynamics of the photoinduced resistance changes of the $La_{0.7}Ba_{0.3}MnO_3$ thin film when the sample illumination was switched on and off is shown in Fig. 2(a). When the sample is illuminated first time, the resistance increases significantly during first 200 s and then slowly decreases and levels off. Such peak feature in the time dependence of resistance after illumination is switched on is present only during first illumination at a given temperature. Subsequent runs do not exhibit peaks after the illumination is switched on.

The difference in the increase of resistance during the first run and subsequent runs (Fig. 2(a)) could be due to the percolative nature of conductance under illumination. As illumination induces spin misalignment and the PIR phase starts to grow rapidly under illumination, the fraction of FMM phase decreases. If the conductivity of these two phases differs appreciably, most of current will flow through the FMM phase, which will lead to the increase of the current density through the reducing fraction of the FMM phase. It was found previously that in phase separated manganites, current and/or electric field stabilizes the conducting phase.[5,20–22] This tendency could lead to a formation and widening of conducting channels through a less conducting matrix and/or reduction of the volume of PIR phase, and subsequently to a gradual increase of conductivity. As a result, the resistivity under illumination slowly decreases to a value corresponding to equilibrium between FMM and PIR phase at this temperature and power of illumination. If these conducting channels remain open during subsequent runs, the resistivity rises under illumination to the equilibrium value without "overshoot."

When the illumination is switched off, the resistance decreases exponentially to its original value with time constant τ (Fig. 2(b), inset). Thus, the PIR phase coexists with the ground state metallic phase on a timescale on the order of one minute, even when the illumination is switched off. The time constant τ decreases with increase of temperature (Fig. 2(b)). This behavior is consistent with existence of a local energy minimum corresponding to a less conducting phase separated by an energy barrier from a deeper energy minimum corresponding to the ground state metallic phase. After illumination is switched off, the system is in a local minima and transitioning through activation process to the ground state. Therefore, the increase of temperature leads to shorter time constants.

The magnitude of the photoinduced changes increase with the intensity of illumination (Fig. 3). The magnitude of the photoinduced changes does not exhibit appreciable wavelengths dependence in the visible range (Fig. 3, inset). Slightly smaller effect for 633 nm may arise from the slightly smaller power density of the illumination of He-Ne laser due to the size of the laser beam.

To summarize, we have observed a significant photoinduced increase of resistance in ferromagnetic metallic state of $La_{0.7}Ba_{0.3}MnO_3$ thin films consistent with photoinduced demagnetization. These photoinduced effects strongly depend on thermal/illumination history and are associated with the energy balance between FMM and PIR phases at different temperatures. Abrupt step-like changes of resistance under illumination indicate percolative nature of photo-induced transition. The lifetime of the photoinduced phase decreases with the increase of temperature showing qualitative agreement with activation process of relaxation to the metallic ground state.

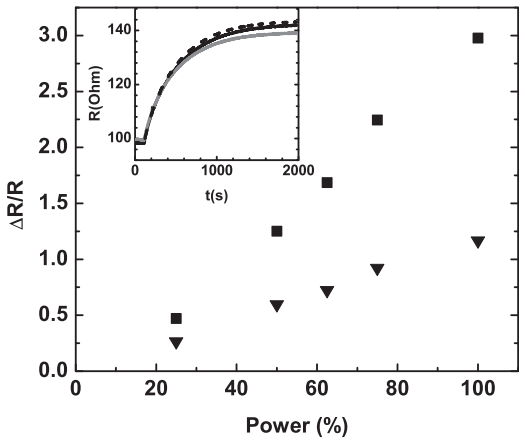

FIG. 3. Dependence of the relative photoinduced change of the resistance of the $La_{0.7}Ba_{0.3}MnO_3$ thin film on the power of illumination at $T = 110$ K (squares) and $T = 230$ K (triangles). The inset shows a time dependence of resistance after illumination was switched on for 633 nm (gray line), 514 nm (dashed line), and 488 nm (black line) wavelengths of 24 mW laser output power at $T = 150$ K.

We acknowledge I. I. Smolyaninov and J. Simpson for helpful discussions and J. Klupt for experimental help. This work is supported by the NSF Grant No. DMR-0348939.

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