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Strain-mediated electric-field control of photoinduced demagnetization in \( \text{La}_{0.8}\text{Ca}_{0.2}\text{MnO}_3 \) thin films

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La\(_{0.8}\text{Ca}_{0.2}\text{MnO}_3 \) (LCMO) thin films have been epitaxially grown on ferroelectric \( 0.67\text{Pb(Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-0.33\text{PbTiO}_3 \) (PMN-PT) substrates. The substrate-induced strain effects on the transport and photoinduced demagnetization in LCMO films were investigated. The photoinduced resistances (PRs) of LCMO systematically changed versus temperature before and after ferroelectric-poling on PMN-PT, indicating that photoexcited extra carriers in LCMO may suppress the neighboring spin correlation due to the photoassisted hopping of anti-Jahn–Teller polarons. Moreover, a significant modulation on PR by electric fields applied across PMN-PT was observed. In situ x-ray diffraction indicates that the observed variations result from substrate-induced strain due to the ferroelectric polarization or converse piezoelectric effect.


The doped manganites are strongly correlated electron system with charge, orbital, and lattice degrees of freedom; therefore, rich physical phenomena were observed, such as colossal magnetoresistance (CMR),1 charge/orbital ordering (CO),2 and electrical phase separation.3 At the same time, the manganites located on the phase boundary are sensitive to the external field including magnetic field, electrical field, current, and light.4–9 Light irradiation, being one of the external perturbations, offers a convenient way to modify the concentration of carriers and induce the changes in the manganites. Among all the photoinduced effects, photoinduced resistance change and demagnetization in the doped manganites are fascinating phenomena which had been extensively studied.6–9 Both of them think the injection of a large number of photoexcited \( e_g \) carriers, which are expected to significantly affect the magnetic interaction and to modify the \( t_{2g} \)-spin ordering, changes the antiferromagnetic/ferrimagnetic phase balance in the films, favoring the insulating antiferromagnetic state.

Recently, a number of studies had been focused on how substrate-induced lattice strain, which could change the strength of the double-exchange interaction and Jahn–Teller (JT) electron-lattice coupling via modifying Mn–O bond lengths and/or Mn–O–Mn bond angles, will affect the physical properties of manganite thin films.10–15 However, strain effects on the photoinduced effects in the manganites have rarely been reported. In this letter, the substrate-induced strain effects on the transport and the photoinduced demagnetization in LCMO film were carefully studied using an epitaxial \( \text{La}_{0.8}\text{Ca}_{0.2}\text{MnO}_3-0.67\text{Pb(Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-0.33\text{PbTiO}_3 \) (LCMO/PMN-PT) composite. The modulation of resistance and photoinduced resistance (PR) by the induced lattice strain in the PMN-PT substrates was observed.

The commercial ferroelectric (001)-oriented PMN-PT single crystal was chosen as substrates due to its perovskite-type cubic lattice structure and outstanding ferroelectric polarization and convers piezoelectric effects. Therefore, it is a good candidate for investigating the effects of strain-mediated electrical control of physical properties in manganites. In our experiment, the geometry of PMN-PT wafer is 3 mm \( \times \) 3 mm with thickness of 0.5 mm. The LCMO thin film was grown on PMN-PT by pulsed laser deposition using a KrF excimer laser with a wavelength of 248 nm.10 The pulse frequency was 2 Hz, and the energy of laser beam was \( \sim 300 \) mJ. The temperature of the substrate was kept at about 700 °C and the oxygen pressure was maintained at 0.5 mbar throughout the deposition. The film thickness was \( \sim 40 \) nm, controlled by the deposition time. After the deposition, the as grown film was in situ annealed in pure oxygen of 1 atm for 30 min. Then, four silver contact pads were prepared on LCMO film by thermal evaporation. Also, the current leads were connected to the silver pads using a MEI-907 supersonic wire bonder to obtain low Ohmic contacts. The light source used in our experiment is a semiconductor laser with wavelength of 532 nm and power density of 3 mW/cm\(^2\).

The crystallization of the LCMO film was examined by x-ray diffraction (XRD). Figure 1 shows a typical XRD \( \theta \sim 2\theta \) spectrum of the LCMO/PMN-PT structure. Besides the reflection of the substrate and the (00l) diffraction peaks of the LCMO, no other peaks from the impurity phases or randomly oriented gains can be observed, indicating that the film is \( c \)-axis preferentially oriented. We also measured the rocking curve of the (002) peak of LCMO [shown in the inset of Fig. 1(a)]. It presents a very small value (\( \sim 0.3^\circ \)) of full width at half maximum, implying a highly epitaxial and good crystallization of LCMO film.

The resistance of the LCMO films was measured using a standard four-probe method in a closed-cycle cryostat, and the schematic of measuring circuit was shown in the inset of Fig. 1(b). An external dc high-voltage source was used to apply an electric-field \( E \) across the PMN-PT crystal. LCMO was served as top electrode due to its small resistance (\( \sim k\Omega \)) compared with huge resistance of PMN-PT substrate (\( \sim G\Omega \)). Figure 2 shows the temperature dependence of the
resistance for LCMO film with and without light, respectively. When PMN-PT substrate was in the unpolarized state (referred to as $P_r^0$), the resistance of LCMO increases with temperature decreasing and undergoes a paramagnetic (PM) insulating state to ferromagnetic (FM) metallic state transition at $T_p \sim 222$ K, exhibiting the typical electrical characteristics of CMR materials. In order to investigate the strain effect induced by ferroelectric polarization to the LCMO layer, we positively polarized the PMN-PT in situ at room temperature by applying an electrical field $E = +10$ kV/cm, which is much larger than the coercive field of PMN-PT of $\sim 2.8$ kV/cm. Here, the positive direction is defined by the electric dipole moment in the PMN-PT point toward the LCMO. After in situ poling for 30 min, we turned off the electric field, and PMN-PT was in the positively polarized state (referred to as $P_r^+$). As seen in Fig. 2, ferroelectric polarization shifts the $T_p$ slightly to a higher temperature of $\sim 223.7$ K and the resistance decreases significantly in a wide temperature range. The resistance of LCMO was also measured under the same conditions when LCMO was irradiated by light and the photoinduced demagnetization effect was investigated. The inset of Fig. 2 presents the dependence of photoinduced resistance changes (PR = $R_{\text{light}} - R_{\text{dark}}$) when the PMN-PT was in $P_r^0$ and $P_r^+$ states, respectively. The PR in different polarization states showed the same variation. It reached the minimum values of $-56$ $\Omega$ for $P_r^0$ and $-60$ $\Omega$ for $P_r^+$ at $\sim 283$ K, and the maximum values of 296 $\Omega$ for $P_r^0$ and 135 $\Omega$ for $P_r^+$ at $\sim 208$ K, respectively. Note that the PR was negative when LCMO was in PM insulating state, while it turned out to be positive when LCMO underwent insulator-to-metal transition into FM metallic state. It is known that the transport of doped manganites is closely related to the spin system of $e_g$ carriers and localized $t_{2g}$ spin core in Mn ions. At $T > T_p$, the localized $e_g$ carriers and lattice distortion formed small (anti-JT) polarons, which were formed when holes are doped into LCMO, so the transport of LCMO is small polaron hopping conduction in the PM insulating state. With further decreasing temperature, the double-exchange (DE) effect plays a key role in the transport of LCMO. The $e_g$ electrons of Mn$^{3+}$ ions jump more easily between Mn$^{3+}$ and Mn$^{4+}$ ions through O$^{2-}$ ions and the film forms FM metallic conduction, resulting in a reduction of resistance. The photoenergy of 532 nm laser is about 2.34 eV, which is much larger than the band gap of LCMO $\sim 1$ eV. In the insulating state, light can excite more photoinduced carriers and enhanced hopping of small polarons, leading to a decrease of the film resistance. On the contrary, in the metallic state, light could excite spin-down $e_g$ electrons, which destroy the FM coupling between spin-up $e_g$ and $t_{2g}$ electrons in Mn$^{3+}$ ions. As a result, it weakens the DE effect and enhances the resistance of LCMO.

To study the strain effect on the transport and the photoinduced demagnetization in LCMO films, we measured the resistance of LCMO versus temperature with and without light by applying different electric-field $E$ to the LCMO/PMN-PT structure when PMN-PT was in $P_r^+$ state. Figure 3(a) presents the resistance of LCMO systematically decreases with increasing $E$ from 0 to 10 kV/cm when LCMO was kept in the dark. The relative decrease in the resistance ($\Delta R/R$) under different electric fields is shown in the inset of Fig. 3(a). The $\Delta R/R$ shows a linear dependence on $E$ at both high and low temperatures. When $E$ increases from 0 to 10 kV/cm, the resistance decreased by $-5.63\%, 7.74\%, 10.65\%, 18.08\%$, and $31.06\%$ at different temperatures ($270$, $250$, $230$, $T_p \sim 224$, and $210$ K), respectively. When LCMO was irradiated by light, the resistance of LCMO exhibits the same variation as that kept in the dark (the results were not shown here). As shown in the inset of Fig. 3(b), we found that $T_p$ increase linearly with increasing $E$, no matter it is in dark or under light irradiation. Figure 3(b) shows the temperature dependence of PR under different $E$. It was found that PR was strongly modulated by electric-field $E$. The maximum values of PR increase from 135 to 477 $\Omega$ for $E$ increasing from 0 to 10 kV/cm.

We also polarized the PMN-PT negatively (referred to as $P_r^-$) and measured the resistance of LCMO varied with applied field $E$, with and without light. It was found that the resistance of LCMO for $P_r^-$ was almost the same as that for $P_r^+$, and the $T_p$ also coincided with that for $P_r^+$. The same results had been observed in other doped manganites. It was reported that the ferroelectric field effect plays a negligible effect in this situation, since the electronic screening length in the manganite films is much smaller than the thickness of the film. Therefore, ferroelectric polarization induced lattice strain in PMN-PT substrates, which would be transferred to LCMO, is the reason that greatly affects the physical properties of LCMO. To verify this point of view, we measured in situ the XRD curves from 44° to 48° by apply-
reflection peaks systematically shift to the lower 2 of Figs. 4 structure. The inset shows the LCMO film when the different electric-field E is applied to LCMO/PMN-PT measuring temperatures. The resistance of LCMO as a function of applied electric-field E under different fields, respectively. 

The temperature dependence of PR for the LCMO film and PMN-PT increases from 4.019 to c values of 0.115% using the expression \( \Delta \varepsilon_{zz} = -2v/(1-v)\Delta \varepsilon_{xx} \), where \( v = 0.5 \) is the Poisson ratio. The same results were observed by Thiele et al.\textsuperscript{11} and Zheng et al.\textsuperscript{13} It was generally believed that the strain effects on the properties of LCMO are attributed to the reduction of in-plane tensile strain in LCMO, which will influence the JT-type distortion in MnO\(_6\) octahedra and further weaken the electron-lattice coupling. Thus, a higher electric-field E will lead to a much heavy distortion in MnO\(_6\) octahedra, result in a reduction of the separation between upper \( \Gamma_3g \) and lower \( \Gamma_8g \) levels,\textsuperscript{12} and the observed larger PR is understandable. 

In conclusion, LCMO films were epitaxially grown on PMN-PT crystals and the effects of substrate-induced strain on the transport and photoinduced demagnetization properties of LCMO were investigated. It was found that the resistance and PR strongly depend on the electric field applied on the LCMO/PMN-PT structure, which induced the in-plane tensile strain into the LCMO layer. The \textit{in situ} XRD analysis indicates that the variation of resistance and PR result from the induced strain due to the ferroelectric polarization or the converse piezoelectric effect.

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