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Phys. Rev. Applied **19**, 044077 — Published 25 April 2023

DOI: [10.1103/PhysRevApplied.19.044077](https://doi.org/10.1103/PhysRevApplied.19.044077)

# Light induced decoupling of electronic and magnetic properties in manganites

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The strongly correlated material  $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$  (LSMO) exhibits metal-to-insulator and magnetic transition near room temperature. Although the physical properties of LSMO can be manipulated by strain, chemical doping, temperature, or magnetic field, they often require large external stimuli. To include additional flexibility and tunability, we developed a hybrid optoelectronic heterostructure that uses photocarrier injection from cadmium sulfide (CdS) to an LSMO layer to change its electrical conductivity. LSMO exhibits no significant optical response, however, the CdS/LSMO heterostructures show an enhanced conductivity, with ~ 37 % resistance drop, at the transition temperature under light stimuli. This enhanced conductivity in response to light is comparable to the effect of a 9 T magnetic field in pure LSMO. Surprisingly, the optical and magnetic responses of CdS/LSMO heterostructures are decoupled and exhibit different effects when both stimuli are applied. This unexpected behavior shows that heterostructuring strongly correlated oxides may require a new understanding of the coupling of physical properties across the transitions and provide the means to implement new functionalities.

Keywords: Resistive Switching, Metal-Insulator Transition, Photocarrier injection.

## 1. Introduction

Strongly correlated electron systems display various physical phenomena such as superconductivity, ferroelectricity, ferromagnetism, and antiferromagnetism as a consequence of the intertwining of several degrees of freedom (charge, spin, strain, doping), producing a wide variety of phases and resulting in complex phase diagrams[1-6]. The critical response of these materials to a modification of the parameters makes them promising candidates for the next generation of electronic devices[7-12]. However, many of these variables are determined by intrinsic material properties set during growth and cannot be easily controlled in applications. Additional functionalities can be introduced by post-growth modifications[8, 13] or by interfacing the correlated oxides with other functional materials with very high sensitivity to external stimuli, such as light[12, 14-16].

Manganites are one of the paradigmatic examples of strongly correlated electronic materials, showing a characteristic magnetoresistive behavior[17, 18]. In particular,  $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$  (LSMO) is one of the most studied materials for spintronic applications[19-21] due to its unique ferromagnetic and half-metallic order up to 360 K. The physical properties of LSMO can be tuned by modifying the electrical, magnetic, strain, and lattice degrees of freedom[22-27]. While the magnetic and electronic properties of LSMO are correlated, in practice, it is often inefficient to control one with the other. In addition, spatial and temporal control of temperature with a certain precision often requires complex architectures and is not energy efficient[9],[28]. Although LSMO is not sensitive to light, some oxygen-deficient LSMO thin films, exhibit a persistent increase of the electrical conductivity after light illumination at low temperature[23]. Moreover, LSMO/ $\text{MAPbI}_3$  heterostructures have been change their magnetization with light illumination[22]. These results showed a great potential of hybrid heterostructures enabling optical functionalities with LSMO in technological applications for advanced oxide electronics[12, 14]. This motivates the search for new functionalities by using light to control the electrical and magnetic properties of strongly correlated materials.

Recently, we reported a large modulation of the MIT temperature ( $T_c$ ), for both non-volatile and volatile resistive switching, of Mott-insulators  $\text{VO}_2$ ,  $\text{V}_2\text{O}_3$ [15], and  $\text{V}_3\text{O}_5$ [29] by incorporating them into a heterostructure with photoconductive CdS[30]. Thus, motivated by the expectation of emergent proximity-induced interfacial effects, we heterostructured LSMO with light-sensitive CdS. We found that the resistance in the insulating state drops significantly

when these heterostructures are illuminated with visible light while the transition temperature ( $T_c$ ) remains constant. The volatile change in the resistance upon illumination depends on the LSMO thickness, suggesting that this is an interface effect. On the other hand, contrary to the effect of light while a magnetic field lowers the resistance it also shifts the transition temperature to higher temperatures. We propose a model where the density of charge carriers increases upon light illumination, resulting in enhanced conductivity. Our results show a practical way to control the electronic properties of manganites with light while also suggesting a new way to understand the coupling between electronic and magnetic properties in these strongly correlated systems.

## 2. Results

We measured the resistance versus temperature ( $R(T)$ ) in multiple CdS/LSMO samples with different LSMO thicknesses as shown in the inset of Figure 1c). The results of the hybrid sample with 3.5 nm LSMO thickness are plotted in Figure 1. Figure 1a) shows the  $R(T)$  of the heterostructure when in the dark and illuminated with visible light of different powers. For all measurements, the resistance as a function of temperature is non-hysteretic, as recorded for both warming and cooling branches. In the dark (green curve), the hybrid CdS/LSMO exhibits a metal-to-insulator transition at 280 K. This behavior is expected from pure LSMO[31, 32] and is in good agreement with the measured results of the control sample (pure LSMO thin film) shown in Supplementary Material. Such agreement confirms that the top CdS layer does not modify the LSMO electronic properties. With increasing light power density, the resistance of the insulating ground state decreases while the  $T_c$  (defined as the maximum in the resistivity vs  $T$ ) remains constant (Figure 1a)). The resistance drops more than 37 % at  $T_c$  when the CdS/LSMO is exposed to a white LED light with 731 mW/cm<sup>2</sup> illumination power density. In the absence of CdS, the effect of light on pure LSMO is negligible (Figure S1a), which also indicates that heating effects are not significant.

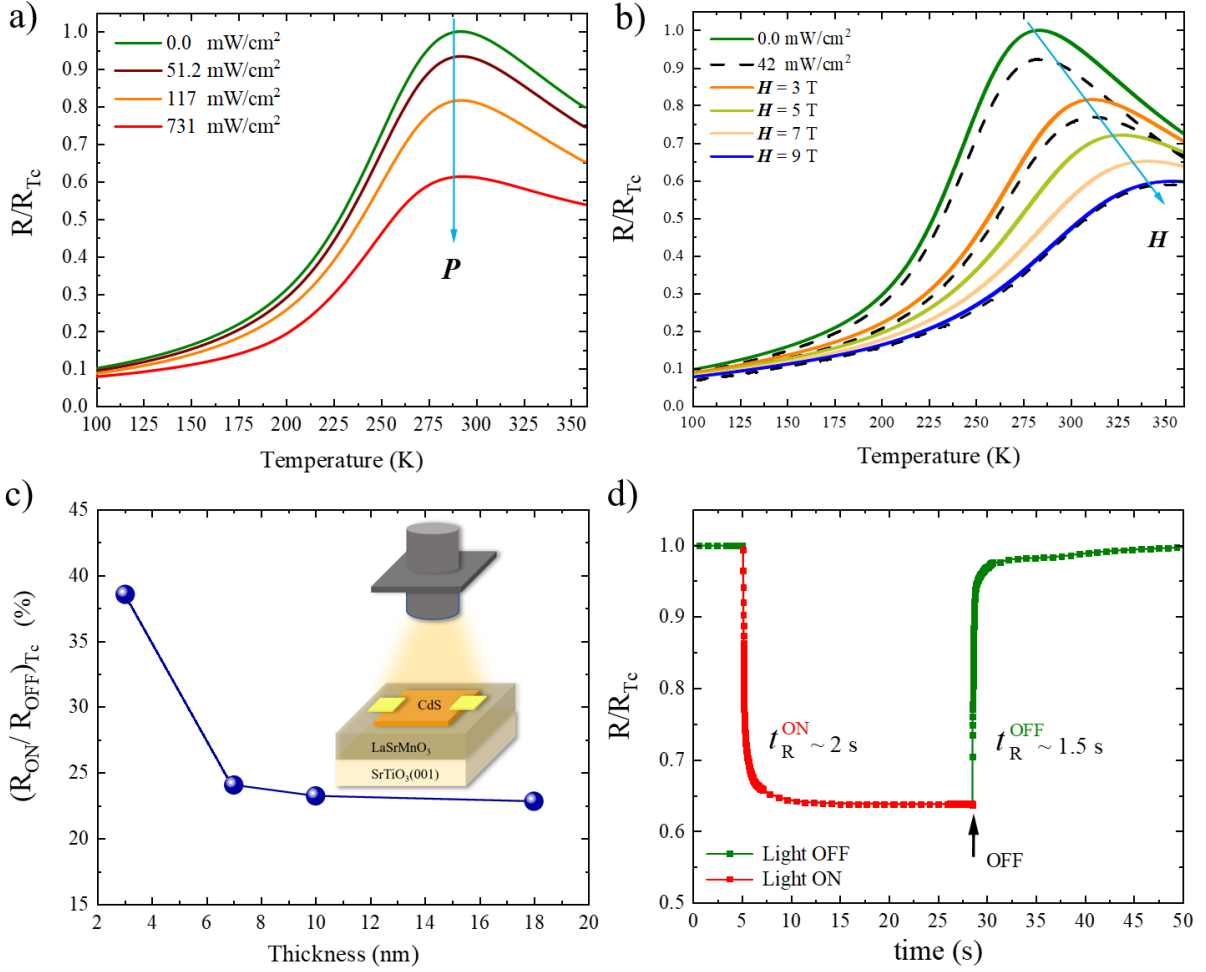


Figure 1: Light-induced modification of the metal-to-insulator transition in CdS/LSMO heterostructures. Resistance vs. Temperature  $R(T)$  measurements for hybrid CdS/LSMO heterostructures, a) varying light power density and b) varying magnetic field. The dashed lines in b) shows the effect of  $42 \text{ mW/cm}^2$  light for selected magnetic fields (0, 3, and 9 Tesla). The solid green curves correspond to resistance vs. temperature without light in all panels. c) Change in the resistance at  $T_c = 280 \text{ K}$ , with illumination for different LSMO thicknesses. d) Relaxation of the resistance (normalized to resistance at  $T_c = 280 \text{ K}$ ).

Figure 1b) shows the resistance versus temperature of the CdS/LSMO heterostructure (with 3.5 nm thick LSMO) when an external magnetic field is applied (3 to 9 T). A larger applied magnetic field results in a more significant resistance drop. For instance, for  $H = 3 \text{ T}$ , the resistance at  $T_c$  decreases by 20 %, shifting the  $T_c$  of 23 K towards higher temperatures. This effect increases as the applied field increases, reaching a 40 % resistance drop at  $T_c$  when a 9 T field is applied. Changes in  $T_c$  to higher temperatures are present for all applied fields. In Figure 1b) all solid lines are acquired in the dark. Interestingly, when the sample is subjected to both magnetic field and light (sequentially), the responses to the two stimuli is additive. The

sample exhibits a  $\sim 20$  K shift in  $T_c$ , similar to the dark, but an additional 5 % resistance drop still appears under illumination (represented by the dashed curves). However, the sample would cease to respond to light if a 9 T field is applied previously. Such behavior is shown in Figure 1b).

The percentage decrease of resistivity with light depends strongly on sample thickness  $\sim 25$  % at 7 nm,  $\sim 24$  % at 10 nm, and 23 % at 18 nm (Figure 1c)). The absolute changes are shown in the Supplementary Material. The decrease in the resistance change under the same illumination with increasing LSMO thickness indicates that this is an interfacial effect, which will be discussed later.

The relaxation time dependence of the CdS/LSMO resistance after light exposure is shown in Figure 1d). The change in the resistance before (green curve) and during illumination (red curve) at 280 K shows a decrease of 40 % in less than 2 s. Once the minimum resistance is reached, the optical light source is turned off, recovering its original resistance in  $\sim 1.5$  s. This shows that the effect of the light in the CdS/LSMO heterostructure resistance is volatile.

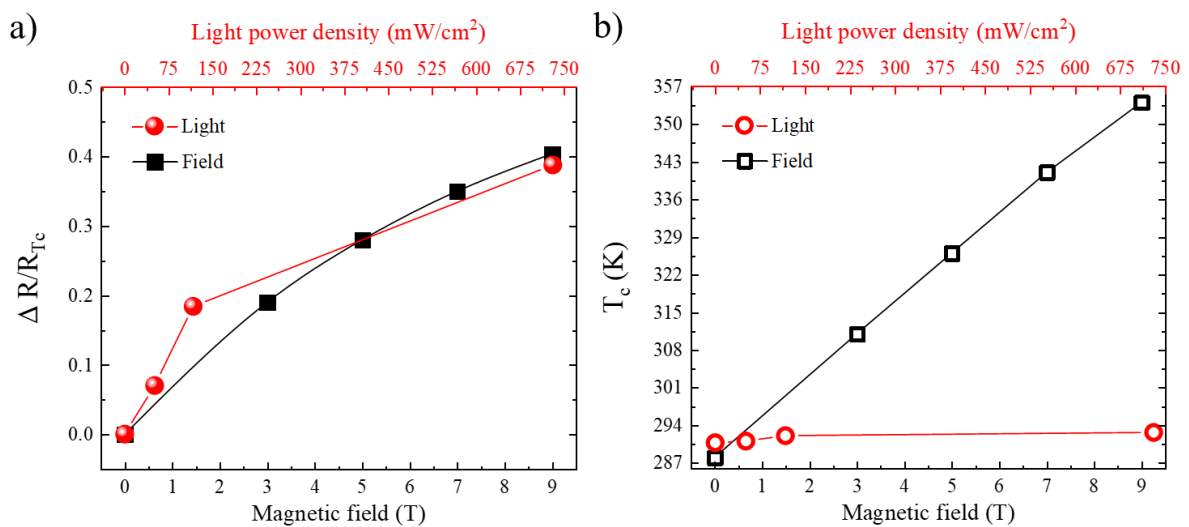


Figure 2: Comparison between two external stimuli in hybrid CdS/LSMO heterostructure. a) Normalized resistance changes  $\Delta R$  at  $T_c$  as a function of applied magnetic field (black squares) and light power density (red circles). b)  $T_c$  as a function of applied magnetic field (black open squares) and power density (red open circles).

Figure 2 compares the effect of the two external stimuli, e.g., light and magnetic field, on the hybrid CdS/LSMO heterostructures. Figure 2a) shows the normalized resistance drop

$\Delta R/R_{T_c}$  at the transition temperature under illumination (solid red circles) and applied magnetic field (black squares). For both external stimuli cases, the resistance decreases with increasing amplitude.  $731 \text{ mW/cm}^2$  light power density decreases the resistance by 40 % at  $T_c$ , nearly the same as applying a very large 9 T external magnetic field.

To further compare the response of electrical properties to the two stimuli independently, Figure 2b) shows the dependence of  $T_c$  on each stimulus. Light illumination had negligible impact on  $T_c$  with a shift of 3 K at the maximum applied power density  $P = 731 \text{ mW/cm}^2$ . In contrast, the transition temperature increases linearly with the applied magnetic field. The  $T_c$  shift can be as large as  $\sim 67 \text{ K}$  when a 9 T external field is applied. This surprising result confirms the different origins of the two mechanisms.

The resistance decrease is expected to be driven by carrier injection from CdS to LSMO film. Similar to our previous report on CdS/VO<sub>x</sub>[29], the light response observed in CdS/LSMO is an interfacial effect between the CdS and LSMO layers. We observed that the response to light becomes smaller with LSMO layer thickness beyond 18 nm. However, in contrast to the present work, in the CdS/VO<sub>x</sub> case, light stimulation changes the resistance and the transition temperature together. This qualitative difference calls for a different physical model to explain the results found for CdS/LSMO.

Furthermore, when we subjected our sample to light while applying an external field (see Fig 1(b)), the effect of light appears in the same way as without a field, namely, an increase of conductivity throughout the whole temperature range without a shift in transition temperature. However, when a larger field is applied, the light response becomes less pronounced. Below, we shall describe a model calculation to understand the cause of this behavior.

### 3. Discussion

Generally, the electronic properties of  $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$  may be described by the Double Exchange Model[33, 34]. Because of the Sr doping, the Mn atoms are in a mixed  $3+/4+$  valence state. Thus, three electrons of the  $3d\ t2g$  band form a “localized” spin  $3/2$  state at each site. On the other hand, there are  $1-x$  electrons per site occupying the conduction  $e_g$  band, therefore, there are nominally  $x$  doped conduction hole carriers. The reason for this is the strong Hund’s rule coupling. Thus, the conduction holes can be either strongly scattered or not by the localized spins  $3/2$ , depending on whether their spins are aligned antiparallel or parallel, respectively.

Hence, when the core 3/2 spins are all aligned into a ferromagnetic state below  $T_c$ , the conduction electrons align to the direction of their magnetic moment  $m_z$ . In contrast, above the transition temperature, the core electrons are in a disordered paramagnetic (PM) state, and the conduction electrons suffer continuous scattering from the unavoidable misalignment of the spin moments. This explains qualitatively the MIT that characterizes the CMR manganites as they cross  $T_c$ .

We have incorporated these qualitative features in a phenomenological model to describe the results in the CdS/LSMO heterostructures. The density of conduction carriers is given by the doping  $x$ , which in the heterostructures is 0.3 per Mn site. Since the photoconductive CdS increases its number of free carriers upon illumination, we assume that part of the photoexcited carriers is transferred into the manganite thin film by a proximity effect. Thus, from the simple Drude model we have,

$$\sigma = \frac{ne^2\tau}{m}, \quad (1)$$

where  $n$  is the carrier density,  $\tau$  is average time between ionic collisions. Further assuming that the additional carrier density from the CdS,  $n_0\Gamma$ , is simply proportional to the illumination power, we have,

$$n = n_0 (1 + \Gamma), \quad (2)$$

and therefore, the resistivity is modulated by the effects of the light as:

$$\rho_\Gamma = \rho_0 / (1 + \Gamma), \quad (3)$$

To take into account the connection between magnetic behavior and resistivity, we adopt a slightly simplified version of the expression introduced by Wang et al[35], which was motivated by the analysis of experimental data in terms of the Double Exchange Model[33, 34]:

$$\rho_0 = ATe^{\frac{\epsilon(1-m_z^2)}{T}}, \quad (4)$$

where  $\epsilon$  is the activation energy,  $T$  is the temperature, and  $A$  is a fitting parameter. The physical content of this formula is clear. At high  $m_z \sim 1$ , deep in the FM phase, the exponent of this formula is small and a first order expansion of the exponential, recovers the semi-empirical



form of the resistivity<sup>31</sup>. On the other hand, at small  $m_z$  the resistivity crosses-over to the activated form of a semiconductor.

The magnetization is governed by a ferromagnetic–paramagnetic transition at  $T_c$ . The effective coupling  $J$  between the local spins of Mn is related to the Hund’s coupling  $J_H$  and the carrier number<sup>31</sup>. Since in LSMO at  $x \sim 0.3$  the  $T_c$  depends weakly on  $x$ , we can take the parameter  $J$  as a constant[36]. Then, for simplicity, we model the temperature- and external field  $h$ -dependent FM transition by means of an Ising model solved in mean-field approximation:

$$m_z = \tanh\left(\frac{h + 4Jm_z}{T}\right), \quad (5)$$

which gives for the critical temperature  $T_c = 4J$  at  $h = 0$ . We may now adopt suitable parameters for this model and explore its predictions.

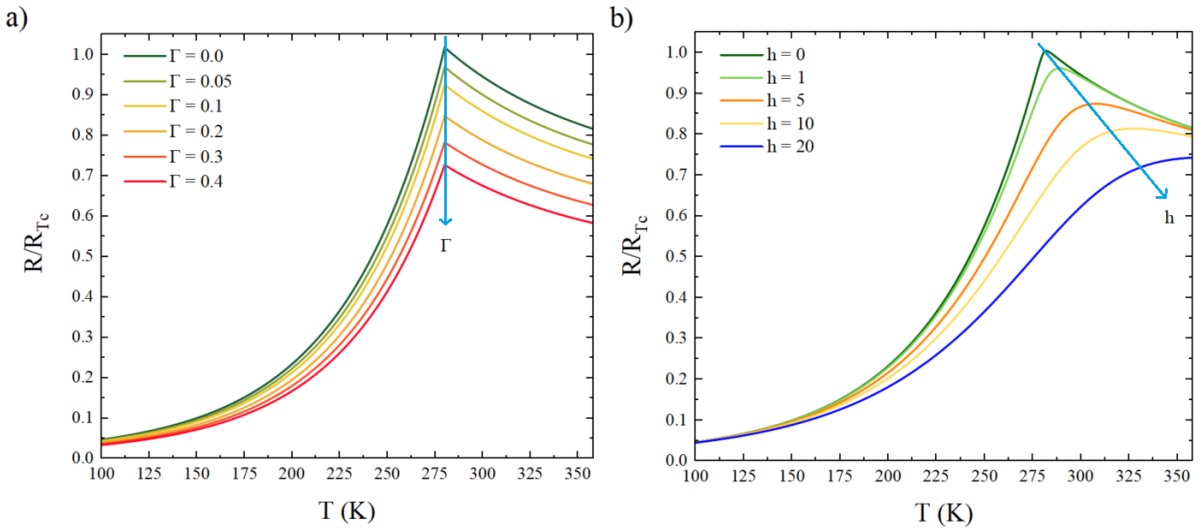


Figure 3: Theoretically computed metal-insulator transition. a) Change in the resistivity of LSMO as a function of temperature for different carrier density  $\Gamma$ , b) and for different applied fields,  $h$ .

Figure 3a) shows the results from our model, which shows a decrease in the resistivity proportional to the applied light power without a shift in  $T_c$ , capturing the qualitative features of the experiment. As a further test of our model, we checked the effects of an external magnetic field  $h$ , as shown in Figure 3b). The magnetic field shifts the  $T_c$  to higher temperatures, also in good qualitative agreement with the experiments. The reason is that the external field provides further stability to the ferromagnetic phase. Thus,  $m_z$  survives at higher temperatures (see

[Supplemental Material Figure S5 for magnetization measurement as a function of thickness](#)).

Consequently, the maximum of the  $\rho(T)$  that defines the MIT is shifted to higher temperatures.

#### 4. Conclusion

In conclusion, we demonstrated that the electronic conductivity of a hybrid optoelectronic heterostructure CdS/LSMO is modified by photocarrier injection from the CdS into the LSMO layer when exposed to light. Although the magnetic and electrical properties in pure LSMO are believed to be correlated, we can independently control the electrical and magnetic properties. While LSMO is known to have no significant optical response, our ultrathin film CdS/LSMO heterostructures show increased light induced conductivity, with ~37 % resistance drop at the transition temperature. The increase in conductivity with light is volatile and equivalent to applying a 9 T magnetic field in this colossal magnetoresistance material. Under a magnetic field, the transition moves to higher temperatures because of the increased stability of the ferromagnetic phase. The changes in the resistance under optical and/or magnetic stimuli are surprisingly independent of each other. Moreover, as the application of large magnetic fields is not readily achievable in most applications, our finding demonstrates a new practical way to control electronic properties with light in complex oxides.

#### 5. Experimental

The ultrathin epitaxial LSMO samples (3.5, 7, 10, 18 nm) were grown on STO  $\langle 100 \rangle$  substrates using a high-pressure (3.2 mbar pure oxygen) and high-temperature (750°C) sputtering. The target-to-substrate distance was set to 1.5 cm, so the highly confined oxygen plasma was tangential to the substrate. 80 nm thick CdS films were grown in a separate deposition with rf magnetron sputtering from a CdS target in a 2-mTorr pure argon atmosphere at 150 °C. For each CdS/LSMO bilayer, two Au (40 nm) electrodes were patterned on the CdS/LSMO heterostructured films.

Morphological characterization measurements were performed in a Veeco Scanning Probe Microscopy (SPM) by analyzing the topographic information in LSMO samples and CdS/LSMO heterostructures. The SPM images can be found in [Supplemental Material](#) at Figure S3 for tapping mode. The structure of the films was analyzed by XRD in a Rigaku SmartLab system at room temperature (See [Supplemental Material](#) at Figure S4 for structural

**characterization**). Transport measurements were carried out on a Montana C2 S50 Cryocooler and TTPX Lakeshore cryogenic probe station, using a Keithley 6221 current source and a Keithley 2182A nanovoltmeter (see [Supplemental Material](#) Figure S1 and S2 **for electrical transport measurements**). A Thorlabs white LED was used for the photoconductivity measurements with a wavelength range from 400 to 700 nm. The magnetic characterizations and resistance measurements with applied field were performed in a Quantum Design DynaCool system equipped with a 9 T superconducting magnet, an optical probe, and a vibrating sample magnetometer.

## **Acknowledgments**

The synthesis of the heterostructures, measurement of transport, magnetic and optical properties were supported by the U.S. Department of Energy's Office of Basic Energy Science, under Grant DE-FG02-87ER45332, the synthesis of LSMO by the Spanish MCI through Grant nos. MAT 2017-87134-C02, PCI 2020-112093 and theory by the French ANR "MoMA" project ANR-19-CE30-0020, Chilean FONDECYT - 1211902 and Basal AFB220001.

## **Author contributions**

HN and IKS conceived the idea. HN designed the experiment. HN, IT, VR and AR-C fabricated the samples. HN, EQ, FA, ACB, TW, performed the transport and magnetic measurements. The X-rays diffraction and SPM measurements were performed by HN, EQ. Simulations and theoretical aspects were performed by LF, SB, FT and MR. HN and IKS wrote the manuscript. All authors participated in the discussion of the results and corrected multiple iterations of the manuscript.

## **Competing interests**

The authors declare no competing interests.

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