

## Competing electronic orders in a composition-spread (Pr<sub>0.6</sub>Ca<sub>0.4</sub>)<sub>1-y</sub>(La<sub>0.6</sub>Sr<sub>0.4</sub>)<sub>y</sub>MnO<sub>3</sub> thin film

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### Introduction

The state-of-the-art thin-film technology enables us to realize the composition dependent phase transition from insulator to metal. In the vicinity of the critical area between insulator and metal in a composition spread manganite film, we have found a new insulator state, which is not observed in the bulk materials.

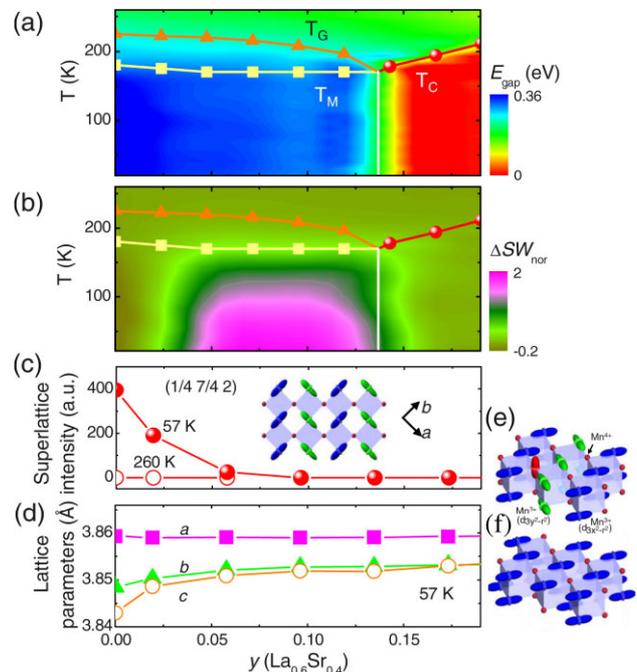
### Experimental results and Discussion

The composition-spread film of (Pr<sub>0.6</sub>Ca<sub>0.4</sub>)<sub>1-y</sub>(La<sub>0.6</sub>Sr<sub>0.4</sub>)<sub>y</sub>MnO<sub>3</sub> on (LaAlO<sub>3</sub>)<sub>0.3</sub>(SrAl<sub>0.5</sub>Ta<sub>0.5</sub>O<sub>3</sub>)<sub>0.7</sub> (011) substrate. We can control the electronic state from insulator to metal by changing the  $y$  (content of La<sub>0.6</sub>Sr<sub>0.4</sub>) composition, as shown in Fig. 1 (a). The well-defined phase boundary between insulator and metal around  $y=0.14$  was established by infrared spectroscopy. In the insulator region, a difference in the spectral weight of the infrared spectra between polarizations is clearly observed along [1 0 0] and [0 1 -1] in the pseudo-cubic setting. Especially, for  $0.04 < y < 0.14$ , the compound shows strong optical anisotropy (See Fig. 1 (b)). To understand the insulating state and the origin of the optical anisotropy, we performed synchrotron x-ray diffraction experiment to investigate the structural properties.

Fig. 1 (c) shows the composition dependence of a superlattice reflection of (1/4 7/4 2) at 57 K, which has been observed for Pr<sub>0.5</sub>Ca<sub>0.5</sub>MnO<sub>3</sub> thin film and related to the stripe-type charge and orbital order as shown in Fig. 1 (e) [1]. The composition dependence of the lattice constant was also measured (Fig. 1 (d)). In the vicinity of  $y=0$ , the lattice constants  $a$ ,  $b$ ,  $c$  satisfy the relation  $a > b > c$ . This is also consistent with that of Pr<sub>0.5</sub>Ca<sub>0.5</sub>MnO<sub>3</sub> thin film [1]. However, for  $0.04 < y < 0.14$ , the superlattice reflections (1/4 7/4 2) and other superlattice and forbidden reflection accompanied by the phase transition are not observed. Further, the difference between the lattice constant  $b$  and  $c$  becomes smaller. From these experimental results, we concluded that the orbital directional order as shown in Fig. 1 (f) maybe take place at low temperature.

The experimental results of x-ray diffraction and infrared spectroscopy studies evidently indicated the

presence of a hidden orbital ordered state between stripe-type charge and orbital ordering insulator and the metallic phases. Detailed information is found in ref. [2].



**Fig. 1:** (a) The phase diagram of (Pr<sub>0.6</sub>Ca<sub>0.4</sub>)<sub>1-y</sub>(La<sub>0.6</sub>Sr<sub>0.4</sub>)<sub>y</sub>MnO<sub>3</sub> determined by the charge gap from the infrared spectrum. Blue and red regions indicate the insulator and metallic phases, respectively. (b) The phase diagram of a difference in the spectral weight. (c, d) The composition dependences of the intensity of (1/4 7/4 2) superlattice reflection (c) and the lattice constants (d). (e, f) Schematic views of the charge and orbital states for  $y < 0.04$  (e) and for  $0.04 < y < 0.14$  (f).

### References

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[2] J. S. Lee et al., Phys. Rev. B **82**, 052406 (2010).

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