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Short Communication:

The role of strain and polar discontinuity in magnetism in $\text{LaMnO}_3/\text{SrTiO}_3/\text{LaAlO}_3$ heterostructures

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Creating novel interfacial effects in quantum-matter-based heterostructures is at the heart of condensed matter physics and materials science.^[1] Utilizing the subtle interplay among charge, spin, orbit and lattice degrees of freedom, these interfacial effects can act as additional knobs to control the future oxide electronics. Among these interplays, two particularly interesting effects, the inevitable strain effect induced by unequal lattice constants and the ubiquitous charge redistribution effect at interfaces with a polar discontinuity. These two effects were also considered as important factors in determining the magnetic properties of polar LaMnO_3 (LMO) in its thin film forms.^[2] Hence, the role of strain and polar discontinuity in determining the oxide interfacial magnetism is of great importance. However, to study their interaction is intricate, because SrTiO_3 (STO) is the only non-polar material among the various kinds of commercially available perovskite-type substrates^[3]. The modulation of strain by changing substrates from non-polar STO to other polar substrates, such as LaAlO_3

(LAO) could also alter the polarity across the interface. Here, we designed LMO/STO/LAO heterostructures to investigate the role of strain and polar discontinuity effects in magnetic properties of LMO. To be specific, we inserted STO as a buffer layer with different thicknesses into LMO/LAO, which could not only regain the polar discontinuity at the interface of LMO/STO, but also retain the strain imposed by the LAO substrate. By varying the thickness of the STO buffer layer (*i.e.*, 6 and 12 uc), the interaction of the two kinds of polarity-discontinuous interfaces (LMO/STO and STO/LAO) was also discussed. Furthermore, two reference samples of 12 uc LMO on STO and LAO substrates were also investigated.

Fig. 1. Ferromagnetism in LMO/STO/LAO heterostructures. (a) Reflection high energy electron diffraction (RHEED) oscillations obtained during the growth of 12 uc LMO/12 uc STO/LAO. The

RHEED pattern is shown in the inset. (b) Reciprocal space mapping (RSM) of 12 uc LMO/12 uc STO/LAO from LAO(103) direction. (c) Schematic view of the interaction of strain and polar discontinuity in LMO/STO/LAO heterostructures. (d) M - T curves of LMO/STO/LAO heterostructures with varying STO thickness and a reference sample of LMO/STO. The cooling field was 1 T, and measurement field was 0.1 T. (e) M - H loops of the same samples at 10 K.

All the samples were prepared by pulsed laser deposition (PLD) equipped with *in-situ* RHEED. **Fig. 1a** shows typical RHEED oscillations of sample 12 uc LMO/12 uc STO/LAO. The thickness of LMO and STO was precisely monitored in a unit-cell precision by counting the period of RHEED oscillations. The corresponding RHEED pattern obtained after the growth (see the inset of **Fig. 1a**) indicates an atomically smooth surface and high sample quality. As shown in **Fig. 1b**, the Q_x values in RSM of the thin films and substrate are identical, which demonstrates that the thin films were coherently grown on LAO substrate^[4, 5]. It would be a direct inference that the strain was not relaxed by buffering 12 uc or less STO in LMO/LAO. The detailed growth conditions and other characterizations of the high quality of these epitaxial thin films can be found in our previous reports.^[2, 6]

Fig. 1c shows a design principle of restoration of the polar discontinuity in strained heterostructure by buffering STO of appropriate thickness in LMO/LAO. It should be noted that the polar discontinuity will drive the charge redistribution, where the charge can leak across the interface^[7], or spread within one constituent^[2, 8] determined by the band structures of the heterostructure. This spreading length is called the charge

screening length. For STO, the screening length L_{TF} was estimated to be about 4.2 nm (~ 11 uc)^[9] based on the Thomas-Fermi model. In our case, this implies that the interaction of polar discontinuities at STO/LAO and LMO/STO interfaces is negligible when the buffering STO exceeds this length. On the other hand, within this length, the buffering STO cannot effectively screen the electrostatic effect of STO/LAO interface. As a consequence, the two interfaces with polarity discontinuity (*i.e.*, LMO/STO and STO/LAO) cannot be regarded as separate factors. In this *short communication*, we chose two thicknesses of STO buffer layer across this screening length, *i.e.* 6 and 12 uc, to investigate the role of these two kinds of polar discontinuity interfaces while maintaining the same strain states. **Fig. 1d,f** shows the M - T curves and M - H loops of LMO/STO/LAO heterostructures with varying STO thickness and the reference sample of LMO/STO, measured by superconducting quantum interference device (SQUID). In line with previous report^[2], the 12 LMO grown on STO substrate showed an unambiguous ferromagnetic signal. However, LMO with the same thickness grown on LAO substrate remained non-ferromagnetic. As mentioned already, the concurrent changes of polarity and strain when swapping the substrates of STO and LAO make it difficult in distinguish their roles in the ferromagnetism of LMO. . It is worth noting that, the 12 uc STO-buffered 12 uc LMO grown on LAO substrate (12 uc LMO/12 uc STO/LAO) shows the similar magnetic property (both M - T and M - H) compared to unbuffered 12 uc LMO grown on STO substrate (12 uc LMO/STO). Since these two heterostructures, *i.e.* STO-buffered and unbuffered LMO grown on LAO substrates, shared similar polar discontinuity at the interface between LMO and STO but have

dramatically different strain conditions, it is more solid that polar discontinuity plays a dominating role in inducing ferromagnetism in LMO. Besides, the 6 uc STO-buffered sample (12 uc LMO/12 uc STO/LAO) shows an advent of ferromagnetism in LMO, which was much weaker than that of the 12 uc STO-buffered one. This weaker ferromagnetism can be explained by the charge screening model since the 6 uc STO is not thick enough to screen the polar nature of LAO. The polar discontinuity effect at LMO/STO interface is weaker, and consequently the ferromagnetism of LMO is suppressed. The charge transfer in the two-dimensional electron gas system of LAO/STO equivalently alters the LAO from polar to non-polar. Therefore, based on this analogy, LAO substrate can also induce polarity in the thin non-polar STO films grown on top.

In summary, the ferromagnetism was created by restoring the polar discontinuity in STO-buffered LMO grown on LAO substrate. The unchanged strain states imposed by LAO substrate disentangled the roles of polar discontinuity and strain, and identified the role of polar discontinuity in ferromagnetism. Moreover, the STO thickness-dependent screening effect was also studied, which exerted great influence upon the polar discontinuity and ferromagnetism.

Conflict of interest

The authors declare that they have no conflict of interest.

Acknowledgments

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