Origin of interfacial ferromagnetism between perovskite transition-metal oxides LaNiO$_3$ and LaMnO$_3$

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Heterointerfaces between perovskite transition-metal oxides have attracted much attention because of their novel electronic and/or magnetic properties that are absent in their bulk form. Recently, it has been reported that an unusual spin order occurs in a paramagnetic LaNiO$_3$ (LNO) layer in the heterointerface region with a "ferromagnetic" LaMnO$_3$ (LMO) layer, resulting in the appearance of the exchange bias between the two oxides[1]. For understanding these exotic magnetic properties appearing at the heterointerface between LNO/LMO, it is indispensable to elucidate the relationships between the electronic states at the heterointerface, especially the interfacial charge transfer between Ni and Mn ions, and the interfacial ferromagnetism. In this study, we have investigated the electronic and magnetic states of both the transition-metal ions in the interfacial region by utilizing the elemental selectivity and surface (interface) sensitivity of x-ray absorption spectroscopy (XAS): The changes in valence of both ions caused by the interfacial charge transfer and resultant spatial distributions of the transferred charges are determined by XAS, while the induced magnetization by magnetic circular dichroism (MCD) of XAS.

The XAS measurements on toplayer for for both LNO/LMO and LMO/LNO bilayers demonstrated the occurrence of the electron transfer from LMO layers to LNO layers (Ni^{3+} + Mn^{3+} → Ni^{2+} + Mn^{4+}) in the interface region[2]. Thus, to evaluate the spatial distribution of the interfacial charge transfer, we have measured the thickness dependent XAS spectra of *underlayers* for LNO/LMO and LMO/LNO bilayers as shown in Fig. 1. As can be seen in Fig. 1, the XAS spectra exhibit the small but distinct spectral modulation reflecting the valence changes due to the interfacial electron transfer. Furthermore, a closer look reveals that the spectral change of Mn-$L_{2,3}$ XAS are totally saturated by 1-ML LNO deposition on LMO layer, while that of Ni-$L_{2,3}$ XAS seems to continue up to 3–4 ML deposition of LMO overlayer on LNO. These results demonstrate that the spatial distribution of the interfacial charge transfer is significantly different between the two layers. Judging from the saturation of spectral change in the underlayer, the interfacial region subject to the charge transfer is evaluated to be about 1 ML for the LNO side, while 3–4 ML for the LMO side[2]. Meanwhile, the Ni-$L_{2,3}$ x-ray MCD spectra of LMO/LNO/LMO sandwiched structures revealed that net magnetization was induced only in the Ni^{2+} ions at a 1-ML LNO layer adjacent to the interface owing to the interfacial charge transfer. As for the counterpart Mn ions, the magnetic moment increased from that of LMO bulk within the 3–4 ML LMO layers from the interface where the transferred holes were distributed. Furthermore, both magnetic moments were ferromagnetically coupled to each other. These results suggest that the stabilization of ferromagnetism in LMO layers due to the interfacial charge transfer and the resultant ferromagnetic coupling between Ni and Mn spins are keys to understanding the induced net magnetization in Ni ions at the interface.

Reference

1)　M. Gibert *et al*., Nat. Mater. **11**, 195 (2012).

2)　M. Kitamura *et al*., Appl. Phys. Lett. **108**, 111603 (2016).
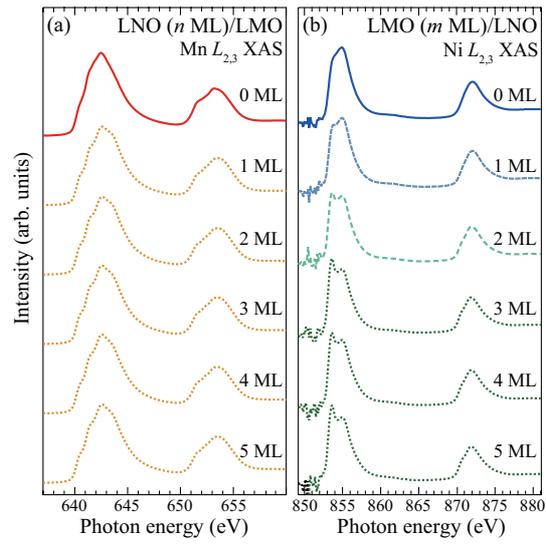
Fig. 1 (a) Mn-$L_{2,3}$ XAS spectra of the LMO *underlayer* of LNO/LMO bilayers (b) Ni-$L_{2,3}$ XAS spectra of the LNO *underlayer* of LMO/LNO bilayers.