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Thickness dependent metal-insulator transition in ferromagnetic $La_{0.6}Sr_{0.4}MnO_3$ thin films studied by x-ray magnetic circular dichroism

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Introduction

Perovskite-type transition-metal oxides (TMO) have been widely investigated due to their variety of physical properties. Recently, the physical properties of TMO ultrathin films have been intensively studied. A number of studies have shown that metal-insulator transitions (MITs) are widely observed in TMO thin films as a function of film thickness [1-5]. $La_{0.6}Sr_{0.4}MnO_3$ (LSMO) is a ferromagnetic metal which shows half metallicity. It has been found that the metallicity and ferromagnetism are lost below a critical thickness around 6-8 ML [1, 2]. We have performed x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD) measurements of the LSMO thin films grown on $SrTiO_3$ (STO) substrates to clarify the microscopic electronic and magnetic states of the LSMO ultrathin films across the thickness-dependent MIT.

Experimental condition

LSMO thin films with thicknesses ranging from 2 to 15 monolayer (ML) were grown on STO substrates by the laser molecular beam epitaxy. The films were capped with 1 ML of $La_{0.6}Sr_{0.4}TiO_3$ (LSTO) and 2 ML of STO. The LSTO layer was deposited to avoid excess hole doping into the topmost MnO_2 plane of LSMO [1]. After deposition, the samples were annealed in an O_2 atmosphere of 1 atm for 45 minutes to remove oxygen deficiencies. This procedure was repeated several hours before the measurements. Detailed sample growth conditions and characterizations are described in Ref. 1.

The XAS and XMCD spectra were taken at BL-16A2 at Photon Factory (PF) and BL23SU at Spring-8 in the total electron yield (TEY) mode. The magnetic field was applied perpendicular to the sample surface.

Results and discussion

Figure 1 shows the Mn 2*p*-3*d* XAS and XMCD spectra of LSMO films with various thicknesses. As shown in Fig. 1(b), the XMCD intensity decreases as the film thickness decreases, reflecting the loss of magnetization observed below the critical thickness [1, 2]. In the XAS spectra [Fig. 1(a)], with decreasing LSMO thickness, the intensities of shoulder structures a and d increase and peaks c and e are shifted towards lower photon energies. The reference spectra in Refs. 6 and 7 show that structures a and d originate from the Mn^{3+} and that peaks

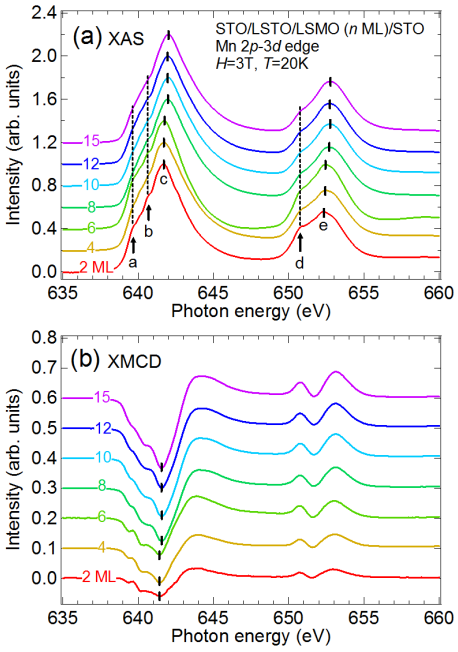

Fig. 1: Mn 2*p*-3*d* XAS (a) and XMCD (b) spectra for various LSMO thicknesses.

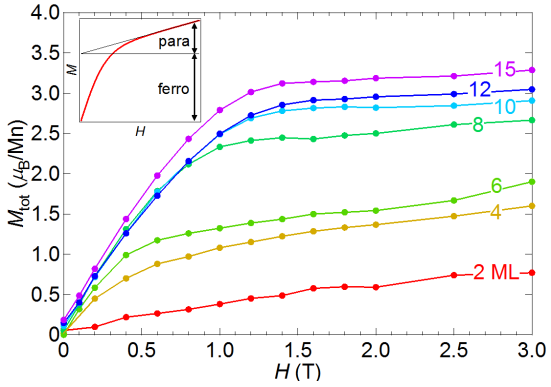

Fig. 2: Magnetic-field (H) dependence of the total magnetic moment (M_{tot}) of Mn estimated from the XMCD sum rules [8, 9]. M_{tot} is defined as the sum of the spin (M_{spin}) and orbital (M_{orb}) magnetic moments. Inset shows the definition of the ferromagnetic and paramagnetic components.

c and e are located at lower photon energies for Mn^{3+} than Mn^{4+}. The present results indicate that the valence of the Mn ions is shifted from Mn^{4+} to Mn^{3+} as the LSMO thickness is decreased. Similar peak shifts are also observed in the XMCD spectra in Fig. 1(b).

Using XMCD sum rules [8, 9], we have estimated the spin and orbital magnetic moments of Mn ions. The orbital magnetic moment M_{orb} was found to be negligibly small compared to the spin magnetic moment M_{spin} ($M_{orb}/M_{spin} \sim 0.01$). In Fig. 2 we show the magnetic field (H) dependence of the total magnetic moment M_{tot}. We have decomposed the M_{tot}-H curves into two components: the ferromagnetic component which saturates around $H \sim 1.5$ T and the paramagnetic component which increases linearly with H up to higher magnetic fields. We have estimated the saturation magnetization and the paramagnetic susceptibility, defined as the intercept and the slope of the M_{tot}-H curves at higher magnetic fields, respectively, and plotted them in Fig. 3 as functions of the LSMO thickness. It can be clearly seen that the ferromagnetic component gradually decreases and the paramagnetic component increases.

In order to test the validity of the 'dead layer' model [1], we have simulated the expected intensity of the ferromagnetic component for a model depicted in the inset of Fig. 3(a). In this model, one assumes that at the LSMO/STO interfaces there are layers with thickness $d \sim$ 4ML (half of the critical thickness) where ferromagnetism is lost. The result of the simulation is shown in Fig. 3(a). The observed ferromagnetic component is quite larger than the simulation. Even if we varied the thicknesses of the top and bottom dead layers independently, we could not reproduce the experimental ferromagnetic moment. This suggests that the loss of ferromagnetism in ultrathin LSMO films cannot be explained by the dead layer model. Alternatively the ferromagnetism may be rather uniformly lost in the entire film.

We have also simulated the paramagnetic susceptibility assuming that all the Mn ions which do not participate in

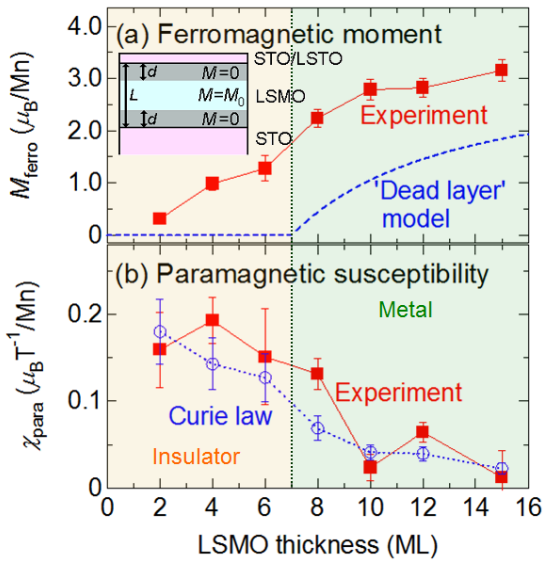

Fig. 3: Thickness dependence of the magnetic properties of the LSMO thin films. (a) Ferromagnetic moment of the experiment (red solid curve) and the simulation (blue dotted curve). Inset shows the 'dead layer' model used for the simulation. (b) Paramagnetic susceptibility estimated from the experiment (red solid curve) and the simulation (blue dotted curve).

the ferromagnetism are in the paramagnetic state of Curie-type. The result of the simulation is shown in Fig. 3(b). The experimental susceptibility is comparable to the simulation at 2 ML and above 10 ML, or slightly larger between 4 ML and 8 ML. This indicates that the magnetic state of the Mn ions can be explained as a mixture of the ferromagnetic insulating (FM-I) phase and the Curie-Weiss-type paramagnetic insulating phase with small Curie temperature (that is, there is weak ferromagnetic spin correlation between the paramagnetic Mn ions). According to the bulk phase diagram of $La_{1-x}Sr_xMnO_3$ [10], a FM-I phase appears around $0.1<x<0.15$. The emergence of the FM-I phase may be related to the valence shift towards Mn^{3+} inferred from the XAS spectra.

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