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Dependence of magnetic anisotropy of the $La_{0.67}Ca_{0.33}MnO_3$ films on substrate and film thickness

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Abstract

Strain in the $La_{0.67}Ca_{0.33}MnO_3$ films has been tuned by varying substrate and film thickness, and its effects on magnetic anisotropy are studied based on the measurements of isothermal magnetization. Measuring the strain in the films by the out-of-plane lattice parameter (c), we found a strong dependence of the magnetic anisotropy constant (Ku) on strain. Ku decreases linearly from $\sim -1.1\times10^6$ erg/cm^3 for $c=0.763$ nm to 1.2×10^6 erg/cm^3 for $c=0.776$ nm, corresponding to a change from tensile strain to compressive strain. Positive Ku signifies a uniaxial anisotropy with the easy axis perpendicular to the film plane, while negative Ku demonstrates an anisotropy of the easy plane character. Smaller or larger c leads a decrease or increase in Ku, which indicates the presence of other effects in addition to those associated with strain. Three distinctive processes for the magnetization are observed along the hard magnetic axis of the films on (001)$SrTiO_3$, suggesting a possibility of strain relaxation even in ultra-thin films.

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1. Introduction

Manganite thin films have been one of the focuses of scientists in the last decade. Much efforts, both theoretical and experimental, have been devoted to the investigation of the underlying physics associated with their extraordinary magnetoresistive behavior [1–9]. In contrast, work on magnetic properties such as magnetic anisotropy and micromagnetic structure is very limited, while the relevant knowledge is obviously a prerequisite for the design and fabrication of heteroepitaxy devices such as spin valve, tunneling junctions, and etc. Different from the bulk manganites, magnetic anisotropy in thin film could be significant. In addition to the intrinsic magnetocrystalline anisotropy, the epitaxial strain induced by substrate, lattice distortions and defects near interfaces, and geometric anisotropy all will affect the magnetic property of the film. It has been reported that a compressively strained film will exhibit an out-of-plane spin alignment, while a film with a tensile strain favors an in-plane magnetization [10]. The magnetic anisotropy energy (Ku) was further determined by O'Donnel and co-workers for the $La_{0.67}Ca_{0.33}MnO_3$ film on (001) $SrTiO_3$ with a thickness of $t=58$ nm, [11] and they found a Ku of $\sim 1.5\times10^6$ erg/cm^3 at a temperature of $T=$ 5 K. It is obvious that the magnetic anisotropy is a function of film thickness. In addition to the variation of the volume to surface ratio, strain in the film could also be altered when

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t changes. Recently, Steenbeck et al. [12] studied the magnetic anisotropy of the $La_{0.7}Sr_{0.3}MnO_3$ films on $LaAlO_3$ and found that Ku, at a constant temperature $T=100$ K, is $\sim 2\times 10^4$ erg/cm^3 for $t=7$ nm and $\sim 4\times 10^4$ erg/cm^3 for $t=25$ nm. Further increase in film thickness may yield a rapid decrease in Ku for that no magnetic anisotropy was detected when t is below 50 nm. On the contrary, Steren et al. [13] reported a monotonic increase in Ku with the decrease in film thickness for $La_{0.6}Sr_{0.4}MnO_3$.

Disagreements between the results of different groups indicate the necessity of a comprehensive study on the anisotropy property of the manganite films, especially the ultra-thin manganite films, which are of special interest for practical application. We also expect an effect of strain relaxation on magnetic anisotropy, which is less studied before. In the present work, $La_{0.67}Ca_{0.33}MnO_3$ (LCMO) films were chosen as the sample for our study for that their magnetic anisotropy was less invoked before. The LCMO films of different thicknesses were grown on different substrates to realize a continuous variation of the strain from the compressive state to the tensile state, which will allow a thorough study on the correlation between the magnetic anisotropy and the lattice distortions.

2. Experimental

Three series of the $La_{0.67}Ca_{0.33}MnO_3$ films were grown on the (001)$SrTiO_3$ (LCMO/STO), (001)MgO (LCMO/MgO), and (001)$LaAlO_3$ (LCMO/LAO) substrates by pulsed laser deposition. The substrate temperature was kept at 800 °C and the O_2 partial pressure at 140 Pa during the deposition. The film thickness was controlled by the number of laser pulses with the deposition rate being 0.1 Å/pulse. After the deposition, the film was furnace cooled to room temperature in an oxygen atmosphere of $\sim$200 Pa.

Phase purity and crystal structure of the samples were examined by X-ray diffraction using a Rigaku X-ray diffractometer with a rotating anode and Cu K_α radiation. A quantum design magnetometer (SQUID) was used for the magnetic measurement. The resistance was determined by four-probe method.

3. Results and discussions

The films are of high single crystal quality with the (001) axis aligning along the film normal according to the X-ray diffraction analysis. Fig. 1 shows the out-of-plane lattice parameter (c) as a function of film thickness. c is 0.7591 nm for the LCMO/STO with $t=10$ nm, significantly smaller than the bulk value 0.7710 nm. This is a result of tensile strain, which causes a lateral expansion and an out-of-plane shrinkage of the lattice. In contrast, the lattice parameter of LCMO/LAO is 0.7828 nm for $t=10$ nm, manifesting

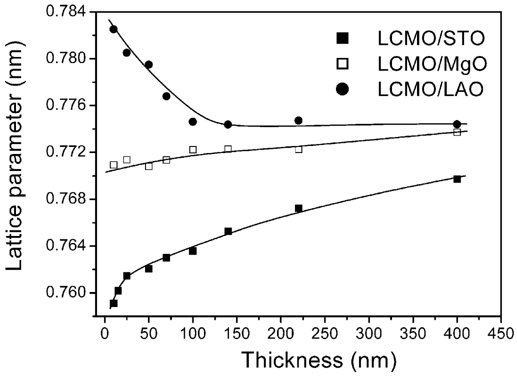

Fig. 1. Out-of-plane lattice parameter of $La_{0.67}Ca_{0.33}MnO_3$ films as functions of film thickness. Lines are guides for the eye.

an in-plane compressive strain. A remarkable observation is the visible turn of the c–t curve at $t=100$ nm for the LCMO/LAO films. It reveals a fact that significant lattice relaxation takes place only in those films thicker than 100 nm. Fascinatingly, the MgO substrate leads to a series of lattice parameters between those of the former two systems. As a result, c increases almost continuously from 0.7591 to 0.7825 nm as the substrate and film thickness vary, and the relative change in lattice parameter is as large as $\sim$3%. This is a quite large variation noting a fact that a reduction of $\sim$2% of the lattice parameter usually yields a completely different behavior of the manganites in the case

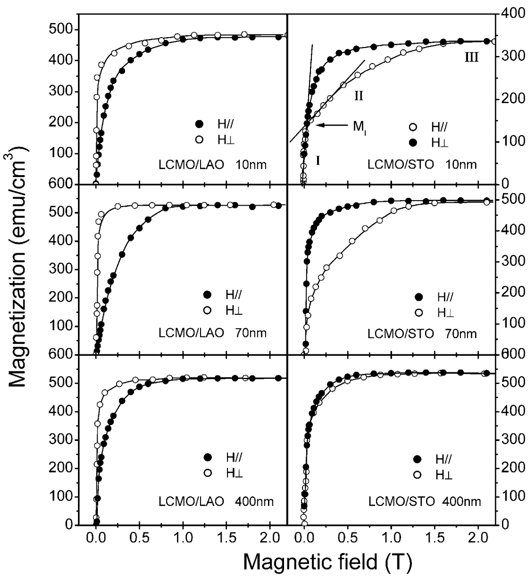

Fig. 2. Selected isothermal magnetization of the $La_{0.67}Ca_{0.33}MnO_3$ films on (001)$LaAlO_3$ (left panel) and (001)$SrTiO_3$ substrates (right panel), measured at $T=5$ K with the magnetic field parallel (H$\|$) or normal (H$\perp$) to the film plane. Arrows in the figure exemplify the determination of M_I. I, II, and III mark the three stages of magnetization.