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The Effect of Epitaxial Strain on Phase Stability in Double-Perovskite La_2NiMnO_6 Films

Sheng-Qiang Wu[1], Sheng Cheng[2], Xiao-Wei Jin[2], Lu Lu[2], Ming Liu[2], Shao-Dong Cheng[1,2] and Shao-Bo Mi[1]

[1.] State Key Laboratory for Mechanical Behavior of Materials, Xi'an Jiaotong University, Xi'an, China
[2.] School of Microelectronics, Xi'an Jiaotong University, Xi'an, China

Double-perovskite multiferroic La_2NiMnO_6 (LNMO) received considerable attention in recent decades due to its abundant magnetoresistance, magnetocapacitance and dielectric properties. Particularly, the near room-temperature magnetic properties have opened up its potential applications in ferromagnetic semiconductor for spintronics and magnetoelectronics [1]. It was reported that for the epitaxial films the synthetic conditions such as growth temperature and oxygen pressure could essentially affect crystal structure and physical properties of the films [2]. In addition, the epitaxial strain of thin films affects the microstructural and physical properties of the films [3]. In comparison with the extensive studies on the synthesis and properties of bulk LNMO materials, the epitaxial strain induced structure evolution remains unclear in the double-perovskite LNMO films.

In this work, LNMO thin films have been prepared on $KTaO_3$ (KTO), $DyScO_3$ (DSO), $SrTiO_3$ (STO) and $LaAlO_3$ (LAO) substrates by the pulsed laser deposition. A KrF excimer laser (wavelength: 248 nm) was applied for ablation of a sintered LNMO target. The same film deposition conditions were held for all different substrates, including the substrate temperature (900 °C), oxygen pressure (250 mTorr), target-substrate distance (10 cm), laser fluence (2 J/cm^2) and repetition frequency (5 Hz). Cross-sectional transmission/scanning transmission electron microscopy (TEM/STEM) specimens were prepared by focused ion beam (FIB) lift-out technique using an FEI Helios600i FIB/SEM system. Bright-field (BF) TEM images and selected area electron diffraction (SAED) patterns were recorded on a JEM 2100 microscope. HAADF imaging was performed on a JEOL ARM200F microscope equipped with an aberration corrector for a probe-forming system, operated at 200 kV. By applying the advanced electron microscopy techniques, we have systematically investigated phase stability of LNMO films affected by the epitaxial strain imposed by different perovskite-type substrates.

Figures 1(a) and 1(b) are the low-magnification BF-TEM images of LNMO films grown on (001) KTO and $(001)_p$ LAO substrates, respectively, showing the overview of the heterostructure. In addition, LNMO films on LAO substrates exhibit a bilayer structure (referred as $LNMO_L$ and $LNMO_H$ respectively). The interface is indicated by horizontal arrows. A typical SAED pattern of the LNMO/KTO heterostructure is displayed in Figure 1(c), viewed along the [010] zone axis. In Figure 1(c), a vertical white arrow shows the splitting of diffraction spots between LNMO and KTO, indicating that the relaxation of misfit strain occurs between LNMO film and KTO substrate. In addition, weak diffraction spots from LNMO films but different oriented domains are presented, as denoted by a blue and a red arrow. In fact, these weak reflections also appear in the SAED patterns of LNMO film prepared on the STO and DSO substrates. The appearance of the weak reflections in the diffraction patterns indicates that LNMO films have a monoclinic or an orthorhombic structure. In contrast to the coherent growth of LNMO films on (001) STO substrates [4], the interface of LNMO/DSO and LNMO/KTO is semi-coherent and misfit dislocations form at the interfaces. Figure 1(d) shows the SAED pattern of the LNMO/LAO heterostructure, viewed along the $[010]_P$ zone axis. The splitting of diffraction spots can be discerned for the high-index reflections. The inset shows the reflection around 402_{LAO} as denoted by a white rectangle in Figure 2d. No weak diffraction spots from $LNMO_L$ and $LNMO_H$ are observed, which indicates that both LNMO layers have a rhombohedral structure.

In addition, the rhombohedral LNMO films were obtained on $(La_{0.289}Sr_{0.712})(Al_{0.633}Ta_{0.356})O_3$ (LAST) and $LaSrAlO_4$ (LSAO) substrates [4]. To understand the effect of the film-substrate misfit strain on the phase structure of LNMO films, the misfit strain is calculated by using the equation of $\Delta f=(a_f-a_0)/a_0*100\%$, where a_0 and a_f are the lattice parameter of substrate and pseudo-cubic LNMO, respectively. The strain-induced phase transition in LNMO films is displayed in Figure 2. It is found that monoclinic LNMO films form under the tensile strain and rhombohedral LNMO films under the compressive strain. Our results demonstrate that phase stability can be tuned by using strain engineering in double-perovskite La_2NiMnO_6 films [5].

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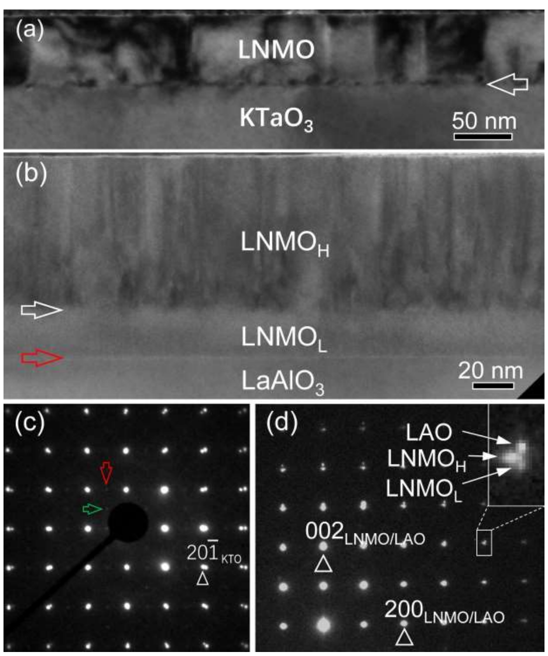

Figure 1. Low-magnification BF-TEM image of LNMO on KTO (a) and LAO (b) substrates. SAED pattern of the LNMO/KTO (c) and LNMO/LAO (d) systems, viewed along the $[010]_P$ zone axis.

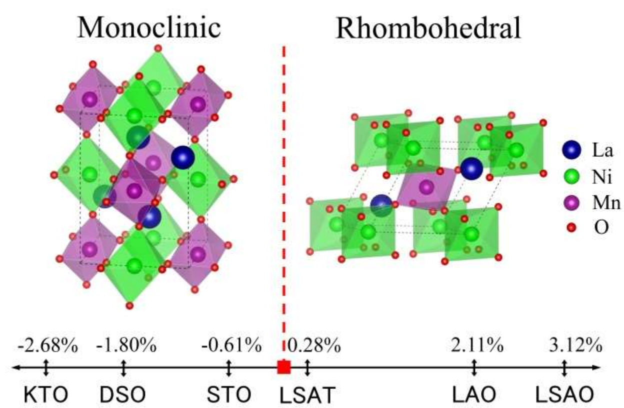

Figure 2. Epitaxial-strain-induced phase transition in LNMO films. The in-plane lattice mismatch values between LNMO and a variety of substrates are given. For monoclinic phase, the value is the minimal in-plane lattice misfit.

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