

LSMO Films with Increased Temperature of MI Transition

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Epitaxial $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ thin films with a significant increased temperature of metal-insulator transition (~ 450 K) are prepared on single crystal MgO (001) substrates using different deposition techniques - a dc magnetron sputtering or a pulsed laser deposition. The crystalline perfection of the films is characterized by X-ray diffraction technique (rocking curve measurements and reciprocal space maps). As a consequence of different methods of the film preparation we show various types of the LSMO crystal structure. Our results indicate that all the LSMO layers grown on the MgO substrate with a lattice misfit of about 8% are relaxed.

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1. Introduction

Since the discovery of colossal magnetoresistance (CMR) effect in perovskite manganites the magnetic and transport properties of these materials and their relations to microstructure have attracted much interest. Among these materials $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ (LSMO) has been intensively studied because of its high Curie temperature (T_C) (higher than room temperature), making this material suitable for uncooled bolometric detectors, sensors, etc. At this temperature the manganites undergo a ferromagnetic-paramagnetic transition and simultaneously, in most cases, a metal-insulator transition - when they reach the resistivity maximum at a temperature T_{MI} .

Beside preferred substrates with a perfect matching to the LSMO, significant results on MgO substrates or MgO buffer layers have also been obtained with a $T_C = 365$ K [1], $T_{MI} = 416$ K [2] and a $T_{MI} = 382$ K [3], respectively.

LSMO has a perovskite structures with a rhombohedral distortion. In the pseudocubic description the lattice parameter and the unit cell angle are 0.3876 nm and 90.26° , respectively. The MgO substrate has the rock-salt cubic structure with a lattice parameter of 0.421 nm. The lattice mismatch between the LSMO film and the MgO substrate is 8%.

2. Experimental details

The LSMO films were deposited using two different deposition techniques, on axis dc magnetron sputtering or pulsed laser deposition (PLD) onto a one-side polished MgO (001) substrate. In case of sputtering the deposition was performed in an Ar(80%)+O₂(20%) atmosphere at a total pressure of 65 Pa. During the deposition, the substrate was heated to 800 °C. In order to increase the oxygen content, the films were subsequently in-situ annealed

in O₂ (10^4 Pa) at 800 °C for an hour. In case of PLD a KrF excimer laser operating at 248 nm with a pulse width of 20 ns, a repetition rate of 10 Hz and an energy density of 2 W/cm² was used to grow the LSMO films. A temperature of substrate holder of 850 °C and oxygen pressure of 53 Pa were set during the deposition. After the deposition the films were cooled down 20 °C/min in O₂ (4×10^4 Pa).

The thickness of the films was about 50 nm, the growth rate of the sputtered and the laser ablated LSMO were 1.2 nm/min and 6.5 nm/min, respectively.

The temperature dependence of the resistance was measured using a standard four-probe technique in a temperature range of 4-480 K.

XRD analyses (θ - 2θ scans, rocking curves, reciprocal space maps) were carried out using Bruker D8 DISCOVER diffractometer equipped with X-ray tube with rotating Cu anode operating at 12 kW. All measurements were performed in parallel beam geometry with medium resolution set-up with parabolic Goebel mirror in the primary beam but without monochromator.

3. Results and discussion

The resistance vs. temperature ($R - T$) dependences of the LSMO films prepared by both dc magnetron sputtering and PLD methods exhibit usually a broader resistance peak with maximal values in temperature range above 400 K (Fig. 1). The temperature of the maximal resistance, denoted as T_{MI} , (temperature of metal-insulator transition, Fig. 1, inset) is significantly higher than the LSMO bulk value (370 K) and corresponds to the onset temperature of the ferromagnetic transition in the LSMO films. The increased values of the T_{MI} and temperature of the ferromagnetic transition are ascribed to an increased strain in the films [2, 4].

XRD measurements carried out in $\theta - 2\theta$ configuration indicate that both types of the LSMO films exhibit only 00 l orientation. The degree of the preferred orientation of the LSMO films, perpendicular to the MgO substrate surface, was deduced from the full width at half maximum (FWHM) of the rocking curve. The FWHM value

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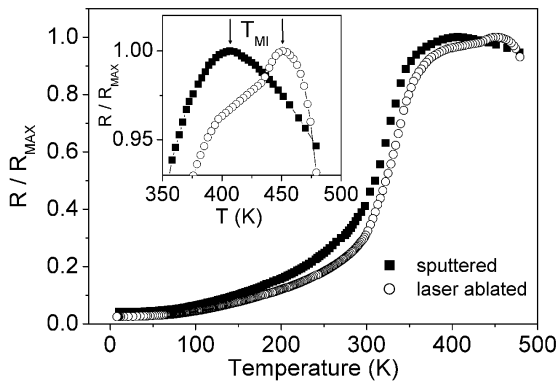


Fig. 1. R-T dependences of two LSMO films with enhanced values of T_{MI} prepared by different methods. The top of the resistance peak for both films is shown in the inset.

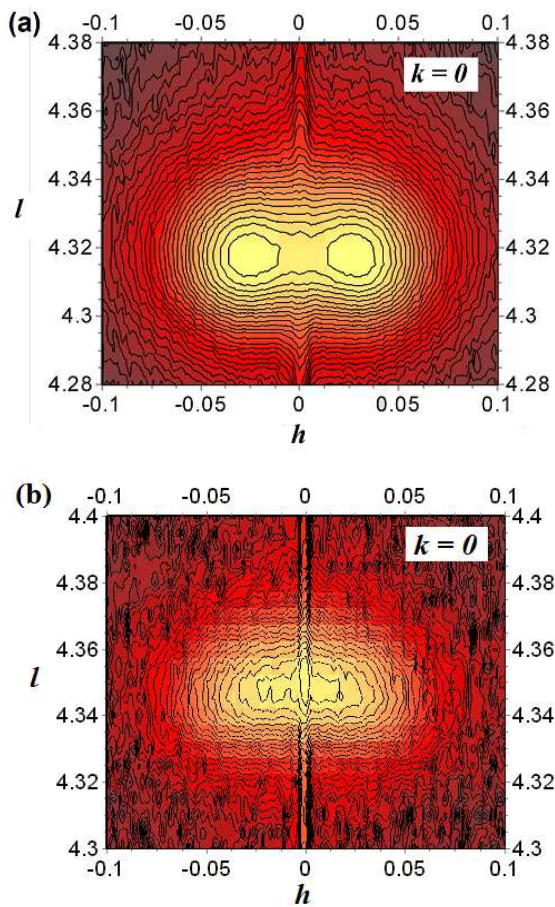


Fig. 2. XRD analyses (reciprocal space maps around 004_C diffraction) showing the difference in the growth of the sputtered (a) and laser deposited (b) LSMO films. measured on the 004_C LSMO peak reached systematically $1-1.3^\circ$. However, we have revealed different shapes of the rocking curves of the LSMO - in case of the sputtered LSMO film two distinct maxima of the rocking

curve, while in case of the laser ablated LSMO film a simple shape of the rocking curve with one maximum was observed. To explain the origin of the rocking curve splitting, the reciprocal space maps were recorded around the symmetric 004_C and asymmetric 204_C Bragg diffractions (Fig. 2a and 2b). The LSMO films prepared by sputtering exhibited splitted rocking curves in the direction parallel to the sample surface, indicating a distorted orthorhombic LSMO unit cell with $[110]$ out-of-plane orientation and $[1-10]$ and $[001]$ in-plane orientations. The out-of plane lattice parameter a_C^\perp of the sputtered LSMO, found by X-ray diffraction in symmetric configuration, is 0.3899 nm. The in-plane parameter a_C^\parallel , deduced from the asymmetric diffraction, reaches a value of 0.3865 nm. On the other hand, no splitting of the rocking curves was observed in case of laser ablated films. These films possess a pseudocubic structure with a_C^\perp parameter of 0.3871 nm and a_C^\parallel parameter of 0.3844 nm.

XRD analyses have revealed that both types of the LSMO films grown on the MgO are relaxed with respect to the substrate. The LSMO lattice parameters are different from the LSMO bulk value, indicating a presence of some strains in the LSMO films. These strains are created during the film growth and are brought into by the adjusting of the deposition conditions of the films.

4. Conclusions

We prepared epitaxial LSMO films using different deposition techniques. According to XRD measurements the crystal structures of the sputtered and the laser ablated LSMO films are orthorhombic and pseudocubic, respectively. The unit cell parameters calculated from the positions of the diffraction maxima in the reciprocal space maps indicate an existence of compressive stress in the LSMO layers. Regardless of the deposition method, our LSMO films exhibited significantly higher T_{MI} values than the bulk value.

Acknowledgments

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