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Morphology and magneto-transport properties of electron doped $La_{0.85}Te_{0.15}MnO₃$ thin film deposited on LaAlO₃ substrate

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We report the structural, electronic transport and X-ray photoemission spectroscopic study of 100 nm thin film of La_{0.85}Te_{0.15}MnO₃ grown on (001) LaAlO₃ single crystal substrate by pulsed laser deposition. XRD results confirm that the film has good crystalline quality, single phase, and has a c-axis orientation. The atomic force microscopic (AFM) results showed that the film consists of grains with an average diameter of 60 nm. The resistivity measurement showed double insulator-metal transitions in absence and as well as in presence of the magnetic field. The resistivity peaks are ascribed to the intrinsic contribution of LTMO film and the tunnelling of spin-polarized electrons at grain boundaries. X-ray photoemission spectroscopy measurements suggest that Te ions are in the Te^{4+} state, while the Mn ions are forced to stay in the Mn^{2+} and Mn^{3+} valence state.

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1. Introduction

Manganite thin films have attracted a great deal of attention as these materials are able to vary their resistivity by orders of magnitude in presence of magnetic [\[1\]](#page--1-0) or electric fields [\[2\]](#page--1-0). This property of colossal magneto-resistance (CMR) has made them promising candidate to be used as magnetic field sensors, as elements in magnetic random access memories and spintronic devices [\[1](#page--1-0)–4]. In addition to the observed colossal magnetoresistance (CMR), their large temperature coefficient of resistivity (TCR), calculated as TCR = $(1/R)[dR/dT]$, in the vicinity of insulatormetal transition temperature, T_c , makes the manganite materials eminently suitable for infrared detector applications [5–[8\]](#page--1-0).

These properties have triggered research interest towards the development and optimization of films by various techniques. Owing to the strong interplay between magnetic, transport, and structural properties, different experimental techniques must be employed in order to study the properties of manganites. In particular, improved experimental microscopic data are necessary in order to appreciate the role of distortions induced by the substrate in thin manganite films. Beside technical problems

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<http://dx.doi.org/10.1016/j.materresbull.2014.05.022> 0025-5408/ \circ 2014 Elsevier Ltd. All rights reserved. related to film growth, some issues have been shown to be relevant in connection with thin manganite films, namely, epitaxial strain-induced effects and surface/interface effects. Positive or negative value of lattice strain (δ') indicates a tensile strain or compressive strain in the film. These strains have vital importance in manipulating the electrical transport, magnetotransport and magnetic properties of the films. Several substrates have been shown to be suitable for the epitaxial growth of manganite films, the most commonly used are $SrTiO₃$ (STO) (0 0 1), LaAlO₃ (LAO) (0 0 1), and NdGaO₃ (NGO) (110). These substrates have a perovskite structure (or a distorted perovskite structure in the case of $NdGaO₃$), which matches well with the structure of the manganites. STO is cubic with $a = 3.905 \text{ Å}$, LAO is nearly cubic with $a = 3.79$ Å, and NGO is orthorhombic with pseudocubic lattice parameters $\sqrt{(a^2 + b^2)/2} = 3.862 \text{ Å}$ and c/ 2 = 3.854Å. Such substrates can induce different strain conditions in the films [9–[11\]](#page--1-0).

Pulsed laser deposition (PLD) has been extensively used to produce good-quality oriented/epitaxial thin films of desired thickness. The physical properties of thin films can be tailored by controlling the growth conditions viz. oxygen pressure during deposition, energy of the laser beam at the target, substrate temperature etc.

In literature, we have not found studies on electron doped thin films of Te doped LaMnO₃ on LAO substrate. However, there are few

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successful reports of the same system grown on MgO and $SrTiO₃$ as substrates [\[12,13\]](#page--1-0). In the present work, we have synthesized the electron doped thin films of $La_{0.85}Te_{0.15}MnO₃$ on LAO substrate by pulsed laser deposition technique and investigated its structural, morphological and magneto-transport properties.

2. Experimental

The standard solid state reaction method was employed to prepare the (target) bulk polycrystalline sample of $La_{0.85}Te_{0.15}MnO₃$. Highly pure powders of MnO₂, TeO₂, and La₂O₃ were mixed thoroughly in stoichiometric ratio and preheated at 1050 °C for 20 h. After first heat treatment, these samples were ground again and sintered at 1130 $\,^{\circ}$ C, with two intermediate grindings. Finally, the mixture was pressed into pellets followed by sintering in air at 1130 °C and slowly cooled at the rate of 2 °C/min down to room temperature. Powder XRD measurement was used to confirm the single phase crystal structure of the bulk target.

The PLD (Lamda Physic, Germany model complex, 201, λ = 248 nm) of the La_{0.85}Te_{0.15}MnO₃ (LTMO) film was carried out using an excimer laser charged with KrF (wavelength of 248 nm and repetition rate of 10 Hz). The sintered bulk sample of $La_{0.85}Te_{0.15}MnO₃$ (LTMO) was used as a target for the synthesis of the thin film with thickness of 100 nm on LaAlO₃ [LAO (001)] substrate. The laser beam, with energy of 200 mJ per pulse, was focused onto the target in a vacuum chamber. The substrate was placed at a distance of about 50 mm from the target on a holder at elevated temperature. The substrate temperature was kept at \sim 750 °C in 350 mTorr oxygen pressure. In order to improve the oxygen stoichiometry, the film is (in situ) annealed at 750° C for 30 min in excess of oxygen pressure. The structure of the film was characterized using high resolution X-ray diffractometer (HRXRD-Bruker D8). The thickness of the film is measured with the help of an XP1 telystep profilometer. Surface growth has been studied by atomic force microscope (AFM), using contact mode Nanoscope Edigital (NSE) instrument. The resistivity of the film was measured using standard four-probe method. The core level X-ray photoemission spectra were recorded using VSW make spectrometer (Al-K_{α} radiation) with a resolution of 0.9 eV and energy 1486.6 eV. The spectra were analysed using XPSPEAK41 software.

3. Results and discussions

3.1. Crystal structure and morphology

Fig. 1 shows the normal XRD pattern in the 2θ range of 20–80 $^{\circ}$ for LTMO film grown on the LaAlO₃ (0 0 1) ($c = 3.7915$ Å) substrate. Peaks corresponding to only the (0 01) (with respect to the pseudocubic unit cell) family of planes of LTMO film are seen at the lower 2θ of the substrate peaks, confirming that the films have good crystalline quality, single phase and c-axis orientation. We have not observed any peak except the (0 0 l) family of peaks of the film and substrate is observed. Further, the epitaxial nature of the film is confirmed by taking a ϕ -scan measurement along the (110) plane of LTMO. The epitaxial nature of the films requires a very high crystalline nature of the films. Fig. 1(a and b) shows the ϕ -scans of LTMO (110) and LAO (220) crystal planes. Each of the LTMO (110) and LAO (220) profiles show four diffraction peaks separated by 90°. The four-fold symmetry confirms the "cube on cube" relative to the LAO substrates. The out of plan lattice constant obtained from (002) peak position of LTMO is 3.942 Å. The epitaxial growth process always brings in a lattice mismatch between the film and the substrate. The lattice mismatch between the substrate and the film leads to the lattice strain which can be calculated using formula, $\delta''(\%) = [(d_{\rm substrate} - d_{\rm films})/d_{\rm substrate}] \times 100$ $[10,11]$. The lattice strain of about -3.97% is found, indicates that the strain is compressive and inplane lattice is compressed, while the out of plane axis will be expanded. The full width at half maxima (FWHM) of the (002) peak is found to be 0.17 $^{\circ}$. The thickness of the film measured with the help of XP1 telystep profilometer is found to be 100 nm.The surface morphology of the LTMO thin film is examined by atomic force microscopy (AFM) for an area of 5 mm² as shown in [Fig.](#page--1-0) 2. As evident from the 2D and 3D images (of size $1 \times 1 \mu m^2$), that the La_{0.85}Te_{0.15}MnO₃ film appears to consist of grains with an average diameter of 60 nm.The surface roughness $(R_{p-\nu})$ of the film is 2.68 nm. The root-mean square roughness $(R_{\rm rms})$ of the film is 0.2 nm revealing very smooth surface. These results indicate that smooth and continuous LTMO thin films can be grown on LAO by PLD technique.

Fig. 1. X-ray diffraction pattern of $La_{0.85}Te_{0.15}MnO_3$ thin film. The symbol '*' denotes reflections from the substrate. (a) ϕ -Scan of LTMO (110) and (b) LAO (220) planes.

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