Thin Solid Films 540 (2013) 194–201

Contents lists available at SciVerse ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Uniaxial stress influence on electrical conductivity of thin epitaxial lanthanum-strontium manganite films

V. Stankevič [a,b,]*, Č. Šimkevičius [a,b], S. Balevičius [a,b], N. Žurauskienė [a,b], P. Cimmperman [a], A. Abrutis [c], V. Plaušinaitienė [a,c]

[a] *Center for Physical Sciences and Technology, Semiconductor Physics Institute, A.Gostauto 11, Vilnius, Lithuania*
[b] *Vilnius Gediminas Technical University, Sauletekio 11, Vilnius, Lithuania*
[c] *Vilnius University, Dept. of General and Inorganic Chemistry, Naugarduko 24, Vilnius, Lithuania*

ARTICLE INFO

Article history:
Received 25 June 2012
Received in revised form 3 May 2013
Accepted 31 May 2013
Available online 14 June 2013

Keywords:
Colossal magnetoresistance
Thin films
Strain effects

ABSTRACT

This is a study of the influence of external uniaxial mechanical strains on the transport properties of thin epitaxial $La_{0.83}Sr_{0.17}MnO_3$ (LSMO) films. Our measurements were carried out using standard isosceles triangle-shaped cantilever. Films which were tensed in-plane or compressed or were subjected to both tension and compression strains were grown onto $SrTiO_3$ (STO), $LaAlO_3$ (LAO) and (001) $NdGaO_3$ (NGO) substrates, respectively. It was found that for thin films (less than 100 nm), the uniaxial compression of such films which were initially tensed in-plane (grown onto STO substrates) produces a decrease of their resistance, whereas the compression of initially compressed films (on LAO substrates) produces an increase of the films' resistance. The same results were obtained for LSMO films grown onto (001) NGO substrates when they were compressed along the [010] and [100] directions, respectively. For thicker films (more than 100 nm), the resistance behavior after uniaxial compression was found to be identical to that produced by hydrostatic compression, namely, the resistance decreases irrespective of the substrate. These experiments also reveal an increase of resistance and a shift of metal–insulator transition temperature T_m to lower temperatures corresponding to a decrease of the film thickness. The occurrence of this effect is also independent of the kind of substrate used. Thus it was concluded that the influence of film thickness on its resistance as well as on the behavior of such films while under external uniaxial compression cannot be explained fully by only the presence of residual stress in these films. A possible reason is that the inhomogeneous distribution of the mechanical stresses in the films can lead to the appearance of two conductivity phases, each having a different mechanism. The results which were obtained when these films were subjected to hydrostatic compression were also explained by this model.

1. Introduction

The discovery of colossal magnetoresistance (CMR) in epitaxial perovskite manganite thin films has led to an increasing interest in the use of these materials both for fundamental science and for other applications. Since the main applications of perovskite manganites are in the design of integrated devices, the properties of thin films are of great importance. The experimental and theoretical studies performed thus far have shown that the biaxial strain due to the lattice mismatch between the substrate and the thin film has a strong effect on the changes of the structural, magnetic and transport properties of these thin manganite films. In particular, variations of their resistivity (ρ), their Curie temperature (T_C) and their metal–insulator transition temperature (T_m) were obtained when the thicknesses of these films were decreased [1–3]. Thus far, these changes have been attributed in most cases to substrate-induced strains and the disorder present in these thin films. It has been assumed that the compressive strains usually reduce the resistivity of these manganite films and increase their T_C and T_m [4,5]. This lattice strain effect has been interpreted using the double-exchange model. Thus it has been predicted that compressive strain could induce an increase of the T_C by increasing the electron transfer due to the compression of the Mn–O bond lengths. Millis et al. [6] proposed an analytical model to describe the effects of the biaxial strain (ε_{xx} and ε_{yy}) on the transport properties of CMR manganites. According to this model, the T_C depends on two parameters: a) the bulk compression $\varepsilon_B = 1/3(\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz})$, and b) the biaxial distortion $\varepsilon^* = 1/2(\varepsilon_{xx} - \varepsilon_{yy})$. This model indicates the very high sensitivity of the material to such strains: a 1% biaxial strain can cause a 10% shift of the T_C. Such behavior can be explained by a decrease of the electron transfer due to the stretching of the Mn–O bonds. This predicted dependence in general was confirmed experimentally using films which were grown on different substrates [3–5,7]. However, recent detailed studies

---

* Corresponding author at: Center for Physical Sciences and Technology, Semiconductor Physics Institute, A.Gostauto 11, Vilnius, Lithuania.
E-mail address: wstan@pfi.lt (V. Stankevič).

0040-6090/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.tsf.2013.05.127

have shown that many of these properties in thin films cannot be explained as due only to the presence of mechanical stresses. During deposition of the thin films, a lattice mismatch between the film and the substrate can cause not only biaxial mechanical stresses, but also distortions of the crystallographic structure [8,9] and chemical composition [10]. Thus was introduced the concept of the coexistence of two (or more) phases in these thin manganite films, which have different conductivity mechanisms (unstressed ferromagnetic (FM) metallic and stressed charge-ordered (CO) insulating). This phase separation is due to structural inhomogeneity caused by non-uniform distribution of the strains within these thin films [11–13]. According to this conception, it is assumed that the shift of the metal–insulator transition temperature (T_m) in their resistivity *vs.* temperature dependence can be explained by changing the quantity of the FM or CO insulating phase fractions [14]. One possible reason for the increase of this paramagnetic phase in stressed films is that this may be due to an increase of the oxygen vacancies [15], which decreases the lattice parameters and in this way compensates for the lattice mismatch between film and substrate [16]. Moreover, the existence of a so-called “dead layer” at the interface between the film and the substrate [17] as well as the presence of a layer which is formed due to the diffusion of components from the substrate into thin film has to be taken into consideration [18]. As a result, these may cause an increase in the resistivity of the film and a shift of the T_m towards lower temperatures.

In order to investigate the influence of mechanical strain on the electrical properties of manganite films, hydrostatic pressure was applied [19,20]. Also, inner pressure was produced by changing the average sizes of the divalent alkaline-earth ions (Sr, Ca, etc.). It is known that hydrostatic pressure changes the T_C and the resistivity in thin films in a manner that is similar to that of epitaxial strain in thin films. However, as these methods induce only average stresses in thin epitaxial films (hydrostatic pressure can be treated as a uniaxial pressure applied in three perpendicular directions), thus they cannot give a clear enough answer as to the exact influence of biaxial strains on the resistivity behavior and the magnetoresistance phenomenon in manganites.

There have been only a few reported investigations in which the influence of the uniaxial external strain on the electrical and magnetic properties of manganite films has been analyzed [21–25]. The gauge factor G of polycrystalline $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) and $La_{0.67}Ca_{0.33}MnO_3$ (LCMO) thin films deposited on oxidized Si has been measured at different temperatures [21]. The measured value of G, defined as the ratio of the relative resistance change to the strain ε, was found to be about 50 for LCMO films and 70 for LSMO films at temperatures of maximum resistance. It decreased to values of about 10 at room temperature. Very high gauge factor values (about 10^5–10^6) were also obtained for epitaxial $(La_{1-y}Pr_y)_{1-x}Ca_xMnO_3$ films at temperatures of maximum resistance [23]. It has also been proposed that strains could be generated in manganite films by using piezoelectric substrates and applying electric fields to them [22,24]. The influence of these substrate-induced strains was investigated in $La_{0.75}Ca_{0.25}MnO_3$ epitaxial thin films [24] and $La_{0.7}(Ca,Sr)_{0.3}MnO_3$ polycrystalline films [22] grown onto piezoelectric substrates. Strain-induced resistance changes in $La_{0.7}Sr_{0.3}MnO_3$ film grown onto (001) $NdGaO_3$ substrates were also investigated previously by our group [25]. It was found that the shift of the metal-insulator transition temperature and the resulting increased resistivity in thin film with respect to thicker films cannot be explained only by the strains induced by lattice mismatches. Therefore, the investigation of the influence of these uniaxial external strains on the properties of epitaxial manganite thin films remained of continuing interest.

In this paper, we present a comprehensive study of the influence of uniaxial external mechanical strains on the transport properties of thin epitaxial $La_{0.83}.Sr_{0.17}MnO_3$ films grown onto $SrTiO_3$ (STO), $LaAlO_3$ (LAO) and (001) $NdGaO_3$ (NGO) substrates.

2. Experimental details

A series of thin $La_{0.83}Sr_{0.17}MnO_3$ films were grown onto (001) $NdGaO_3$, (001) $SrTiO_3$ and (001) $LaAlO_3$ substrates using a vertical hot wall Injection Chemical Vapour Deposition reactor. Details of the epitaxial growth of these films are described in [26]. All the samples were prepared under the same conditions, while their thicknesses were varied from 8 nm to 140 nm. The metal–insulator transition temperature T_m of these films was determined in a range between 240 K and 340 K, respectively. The structural characterization of these films and the orientation of their substrates were determined using an X-ray diffractometer. {100} and {010} $NdGaO_3$ reflections were used to identify the orientations of the substrates. The analysis of the surface morphology of these films was conducted using an atomic force microscope.

It is known that the LAO substrate has a pseudo-cubic crystallographic structure with lattice constant $a = 0.3788$ nm, the STO substrate has a cubic structure with $a = 0.3905$ nm, and NGO substrate has an orthorhombic structure with $a = 0.5426$ nm, $b = 0.5499$ nm and $c = 0.7706$ nm. On the other hand, bulk LSMO has a pseudocubic perovskite structure with lattice parameter $a_p = 0.387$ nm [27]. As a result, the coherent LSMO films deposited onto STO substrates are tensile, while films deposited on LAO substrates are compressed. In-plane lattice sketches with the pseudo-cubic lattice constants for bulk crystals of LSMO (dashed line) and three different substrates (continuous line) are illustrated in Fig. 1. The LSMO films grown onto (001) NGO substrates having orthorhombic structures with different a and b parameters in the (001) plane were of particular interest. In their case, the crystallographic structure of LSMO can be represented as an orthorhombic structure with lattice parameters $a \approx b = 0.547$ nm, $c = 0.771$ nm [27–29] rotated at a 45° angle with respect to the cubic lattice. For this reason, thin LSMO films grown onto (001) $NdGaO_3$ substrates have different lattice mismatch signs for different crystallographic directions in the film plane [30]. Thus it can be assumed that these films are compressed in the a [100] direction and are tensile in the b [010] direction. Therefore, films coherently grown onto NGO substrates having (001) crystallographic orientation are anisotropically strained. In our case, the relative lattice mismatch between the film and NGO substrate is calculated according to the equation $\varepsilon = (a_{sub} - a_{bulk})/a_{bulk}$, where a_{sub} and a_{bulk} are the lattice parameters of the substrate material and the bulk manganite, respectively. Thus they are equal to −0.8% in direction a [100] and 0.53% in direction b [010]. The lattice

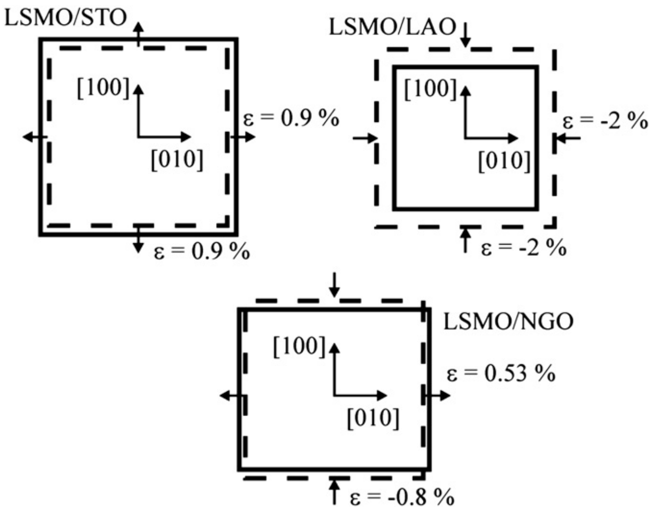

Fig. 1. In-plane lattice sketches with pseudocubic lattice constants for bulk LSMO crystals (dashed line) and for different substrates (continuous line).