Journal of Magnetism and Magnetic Materials 332 (2013) 67–70

Contents lists available at SciVerse ScienceDirect

Journal of Magnetism and Magnetic Materials

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

Structural and magnetic properties of $CoTiO_3$ thin films on $SrTiO_3$ (001)

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ARTICLE INFO

Article history:
Received 25 April 2012
Received in revised form
16 November 2012
Available online 13 December 2012

Keywords:
Cobalt titanate
Pulsed laser deposition
Antiferromagnetism

ABSTRACT

$CoTiO_3$ (CTO) thin films were grown on single crystal $SrTiO_3$ (STO) (001) substrates by pulsed laser deposition, with thicknesses ranging between 2.6 and 25 nm. Two-dimensional growth is observed up to a thickness of 2 unit cells, but then the film partially relaxes with CTO [100] oriented parallel to STO [110] and CTO [001] oriented parallel to the STO [001]. Magnetic characterization of our films revealed an enhancement of the antiferromagnetic Néel temperature (to ~53 K) when compared to the bulk (38 K). This is attributed to a tensile *c*-axis strain as evidenced from x-ray diffraction measurements.
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1. Introduction

Thin films of functional oxides are of great interest for many possible novel device applications [1]. A popular group of materials is based on the general formula ABO_3, where A and B are typically transition metal ions. These oxides usually crystallize in a perovskite structure, e.g. $SrTiO_3$ (STO), $LaAlO_3$ (LAO), or an ilmenite structure, e.g. $FeTiO_3$, $NiTiO_3$.

$CoTiO_3$ (CTO) belongs to the latter group with $a=0.5066$ nm and $c=1.3918$ nm (hexagonal crystal axes). It behaves as an antiferromagnetic insulator with a bulk Néel temperature of approximately 38 K [2]. It has mostly been investigated as a potential high-k gate dielectric to replace SiO_2 in CMOS [3]. The fabrication procedure is commonly a direct oxidation of sputtered metal layers [3] or a sol–gel procedure for thicker films [4]. However so far, pulsed laser deposition (PLD), despite being a widespread technique for the deposition of oxides, has not been applied for the growth of CTO thin films.

In this work, we investigate pulsed laser deposition growth of thin films of CTO on single crystal STO (001) and investigate the growth, the morphology and the corresponding functional properties of the thin films.

2. Material and methods

$CoTiO_3$ films were grown on single-crystal $SrTiO_3$ (001) substrates (Crystal GmbH) by pulsed laser deposition using a KrF laser ($\lambda=248$ nm). The repetition rate was set to 10 Hz at a target to substrate distance of 80 mm and laser fluence of 1.5 J/cm^2. The substrate heater temperature was varied between 600 and 850 °C. The oxygen partial pressure during deposition and cooling down was kept constant at 10^{-2} mbar. The CTO target was prepared by solid state reaction from a stoichiometric mixture of Co_3O_4 (Alfa Aesar, 99.9985%) and TiO_2 (Alfa Aesar, 99.99%) powders. The powders were milled, mixed, pressed and sintered at 1100 °C (9 h).The resulting phase of the target, as evidenced by X-ray diffraction (XRD), was mainly $CoTiO_3$.

Film growth was monitored by in-situ reflection high-energy electron diffraction (RHEED). XRD data were collected on a Bruker D8 theta/theta 4-circle diffractometer with Cu Kα radiation and a graded mirror. Film thicknesses were determined by x-ray reflectometry on the same instrument. Lattice parameters were calculated from profile fitting of four (00l) x-ray diffraction peaks, using the substrate peaks as an internal standard to correct for the height displacement error.

Surface topography was measured with a Veeco AFM Multimode Nanoscope III in tapping mode and the images processed with the WSxM software [5].The magnetic susceptibility was measured using a Cryogenics vibrating sample magnetometer with variable temperature control. Samples were cooled in the absence of a magnetic field ('zero field cooled') and subsequently measured in an applied magnetic field in order to pick up the antiferromagnetic transition.

3. Results and discussion

The deposition temperature we employ for the layer-by-layer growth of LAO and STO perovskites is 850 °C [6–8]. An AFM image of a CTO film grown at this temperature is shown in Fig. 1. The in-plane cubic directions of the STO are parallel to the edges of the

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http://dx.doi.org/10.1016/j.jmmm.2012.12.017

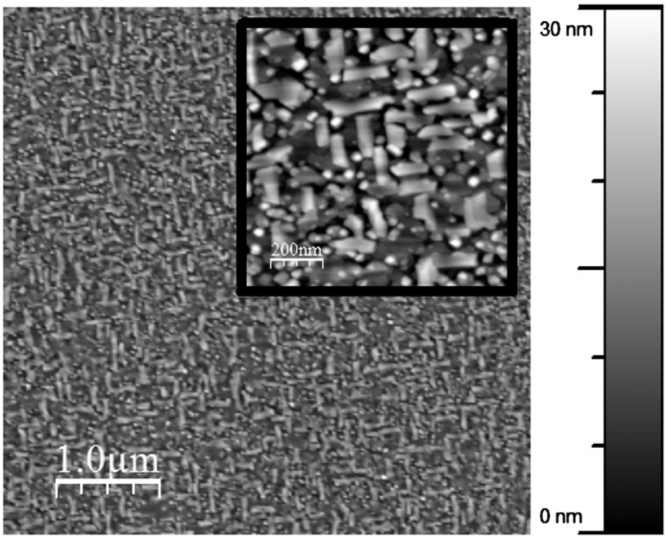

Fig. 1. AFM image of the surface of a CTO film grown at 850 °C. The inset shows a detailed view.

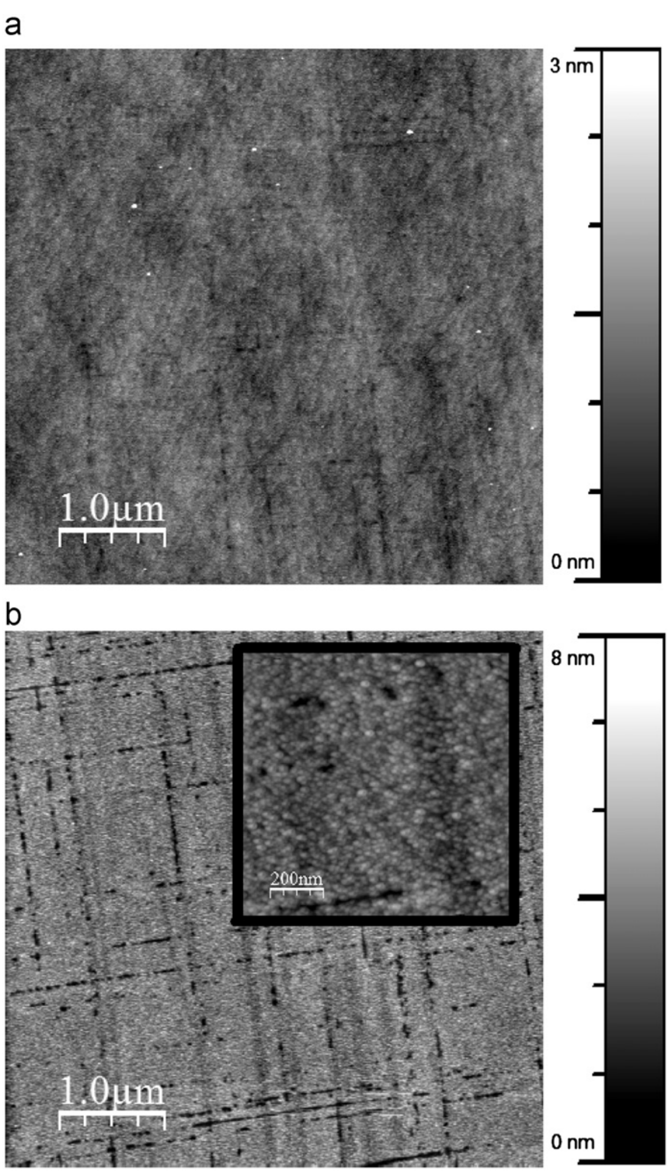

Fig. 2. AFM images of a CTO film grown at 600 °C with a thickness of (a) 2.6 nm and (b) 4.9 nm.

image. The CTO exhibits a clear island growth whereby two perpendicular orientations appear to be favorable.

Upon decreasing the deposition temperature to 700 °C or 600 °C, the CTO initially grows in a layer-by-layer mode up to two unit cells (2.6 nm thickness), after which islands nucleate. Fig. 2 shows an AFM image of a 2.6 nm film (with surface steps clearly visible) and a 5 nm film for comparison. The island size is equal to or smaller than the AFM tip (20–30 nm) so cannot be determined by this method. The RHEED specular spot intensity oscillation during the growth of 2 initial unit cells of CTO at 700 °C is displayed in Fig. 3. The diffraction pattern recorded along STO [100] is also shown before and after the deposition. Streaks indicating a smooth surface are clearly visible.

CTO films grown at 700 °C were studied by XRD to reveal the crystallographic relationship with the STO (Fig. 4). For a 5 nm film, no diffraction peaks other than the substrate stand out from the background. Thicker films (9.2 and 24.5 nm) exhibit dominant peaks corresponding to the (003) and (006) planes, as well as

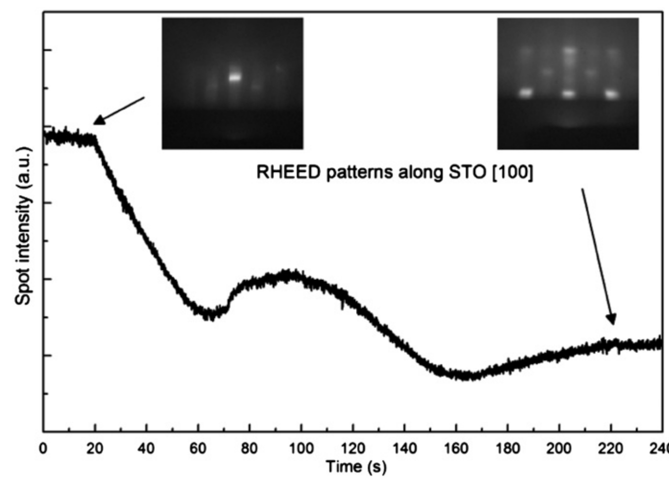

Fig. 3. RHEED specular spot intensity versus time, with the diffraction pattern before and after the deposition of 2 unit cells of CTO on STO, recorded along the STO [100] at 700 °C.

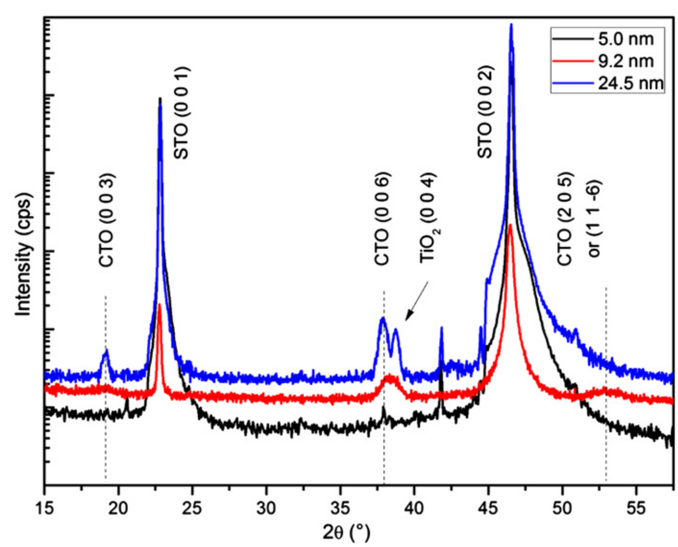

Fig. 4. XRD pattern of a 5, 9.2 and 24.5 nm thick CTO film grown at 700 °C. STO (00*l*) substrate peaks are indicated. Peaks corresponding to CTO (003) (around 18°) and (006) (around 38.7°) are visible in the higher thicknesses. There is a spurious CTO orientation near 53° in the intermediate thickness (possibly an interface contribution) and a TiO_2 (004) peak at 37.8° in the 24.5 nm film.

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