Growth Parameter Control of Structures and Properties of Perovskite Thin Films

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Abstract:

Application of growth parameters to control thin film structures and properties is an important method for improving film quality for applications in advanced electronic devices. Lattice engineering of thin films is key to improve the crystallinity and quality of films and the devices they comprise. In this study, as a model system we seek to control the growth of a perovskite oxide film, strontium titanate ($SrTiO_3$), through magnetron sputtering. Film growth of $SrTiO_3$ was investigated on lattice-matched lanthanum aluminate-strontium aluminum tantalite ($(LaAlO_3)_{0.3}(Sr_2AlTaO_6)_{0.7}$) substrates, where we focused on the distance between substrates and sputtering target. We find that alteration of the distance leads to good Sr and Ti control, high crystallinity, and smooth surfaces, but oxygen deficiency in the films is still an obstacle to improving the electrical properties.

Introduction:

Stacked thin film heterostructures are important in the development of many devices, such as multilayer ceramic capacitors, batteries, and integrated circuits [1]. Highly crystalline films and interfaces are essential in determining the quality of thin films and improving device performance.

Sample	Temperature (°C)	Sputtering power (W)	Ar flow (sccm)	Ar pressure (mTorr)	Distance (mm)	Deposition time (h)	Thickness (nm)
Sample A	500	40	0.2	20	53	1	165
Sample B	500	40	0.2	20	73	1	315
Sample C	500	40	0.2	20	93	1	22

Table 1: Typical deposition growth parameters for STO thin films.

The most common technique for producing practical stacked thin films is magnetron sputtering, due to the advantages of low-cost deposition and relative simplicity of the system [2]. To obtain high-quality thin films, lattice and orientation matching between substrate and film are important [3]. For instance, selection of substrates with larger or smaller lattice constants than those of the films results in tensile or compressive strain in the films, respectively. Selection of substrates and growth parameter optimization during sputtering deposition are crucial to improve film properties.

Our goal in this project is to alter growth parameters to control perovskite thin film structures and properties. In this study, we used a compact sputtering gun with an adjustable head and controlled the distance between the substrate and sputtering target during deposition to fabricate $SrTiO_3$ (STO) thin films.

Experimental Procedure:

Thin films of STO were grown on lattice-matched $(LaAlO_3)_{0.3}(Sr_2AlTaO_6)_{0.7}$ (100) (LSAT) and lanthanum aluminate, $LaAlO_3$ (LAO) substrates using radio-frequency magnetron sputtering. In-plane lattice constants of STO and LSAT are 0.390 and 0.387 nm, respectively, resulting in $(a_{STO}-a_{LSAT})/a_{LSAT}=0.0103$. Typical growth conditions are summarized in Table 1. These conditions were held constant for each sample, but the sample holder height was changed to 130 mm, 150 mm, or 170 mm, which correspond to a distance ($d_{s\text{-}t}$) between substrate and sputtering target of 53 mm, 73 mm, and 93 mm, respectively. After the sputtering depositions, STO films were characterized via profilometry, atomic force microscopy (AFM), x-ray diffraction (XRD), and x-ray fluorescence (XRF). Gold/titanium electrodes were deposited onto the films using vacuum evaporation to form Ohmic contacts for resistivity and Hall-effect measurements.

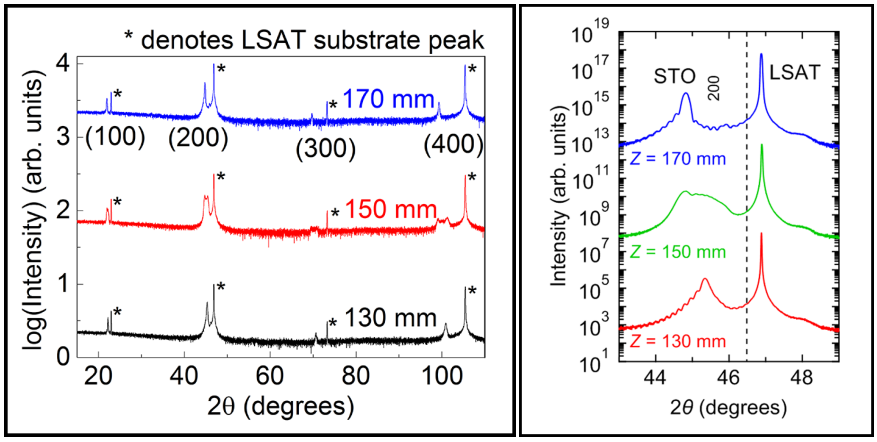

Figure 1, left: XRD wide scan of STO/LSAT samples. Figure 2, right: XRD (200) peak of STO/LSAT samples.

Results and Conclusions:

Figure 1 shows XRD wide scans of the STO/LSAT samples deposited under different $d_{s\text{-}t}$. It is clear the films were *c*-axis oriented and single-phase without impurity peaks. A closer look at the 200 peak of the films shows all three film peaks occurred at lower angles than the bulk STO peak, represented by the dashed line in Figure 2. The lower peak angles indicate the films are compressively strained, as the lattice constant of LSAT is lower than that of STO, which led to the lengthening of the c-axis parameters. Additionally, oscillations visible on the 200 peak are an indication the films are highly crystalline, which was confirmed by rocking curve scans showing peak widths around 0.1°.

The AFM images (Figure 3) of the STO/LSAT samples revealed the thin films were significantly more smooth, with a root-mean-square roughness of 0.177 nm and 0.271 nm for samples A and C, respectively. The faint diagonal lines visible in Figure 3 indicated stepped-and-terraced surface, implying the surface of sample C was very flat. We performed XRF measurements on the STO/LAO samples rather than the STO/LSAT, as LAO does not contain Sr which would convolute the measurement. The XRF analysis of the stoichiometry revealed a Sr deficiency in sample A, but near stoichiometric ratios of Sr:Ti in samples B and C. These results imply that the enlargement of *c*-axis parameters resulted from compressive strain and amount of oxygen rather than cation (non)stoichiometry.

Electric properties collected from Hall-effect measurements showed resistivity (1.1 Ωcm) and carrier concentration ($2.8*10^{19}$ cm^{-3}) in sample C were several orders of magnitude different than the other samples. In fact, sample C looked light gray, indicating an oxygen-deficient film as is usually seen in n-type oxide semiconductors. Although former literatures reported that compressively strained STO films show a ferroelectric transition [4], our STO films, even sample C, could not show these effects.

We were able to conclude that our sputtering method grew highly crystalline STO thin films with smooth surfaces. Cation stoichiometry for Sr and Ti was controlled through the changing of $d_{s\text{-}t}$. Electric properties remained poor due to oxygen deficiency in the STO films.

Future Work:

Oxygen control is necessary in improving the properties of sputtered thin films. In order to achieve this control, we can next use oxygen gas exposure in addition to argon gas during the sputtering deposition procedure. Additionally, we can test the effect of changing the angle of the head of our sputtering gun and tune the angle for property control of the film growth.

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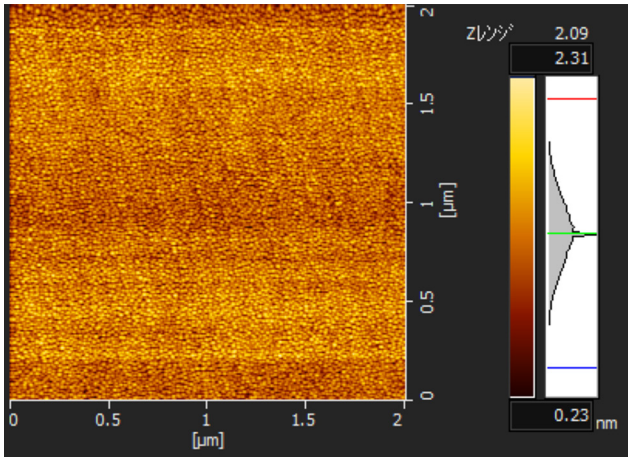

Figure 3: 2000 × 2000 nm 0.98 Hz AFM scan of 170 mm STO/LSAT thin film.