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# Plasma Power Effect to the Structure and Morphology of Strontium Stannate Thin Film

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### ABSTRACT

Hybrid p-n heterojunction requires a suitable n-type material to support the charge transfer process in photodiode or solar cell applications. Numerous materials have been researched in terms of wet and dry processes. This paper investigated the effect of varied plasma power levels of 50 W, 80 W and 100 W at room temperature on the structure and morphology of an alkaline-earth-based perovskite oxide thin film. Strontium stannate (SrSnO<sub>3</sub>) thin films are deposited on an indium tin oxide (ITO) substrate using radio frequency (RF) magnetron sputtering at various power levels. The crystallographic orientation of the thin films was examined and it was observed that the (102) plane exhibited dense growth at low power, while the (002) plane showed increased intensity at higher power levels. Additionally, the increase in RF power resulted in larger crystallite and grain sizes, along with modified grain boundaries. Contrarily, surface roughness, resistivity, macrostrain and work adhesion were reduced. The nearly zero W-H linear fitting plot observed at 80 W can serve as a valuable reference for the fabrication of preferred films. These findings provide valuable insights for optimising the growth conditions of alkaline-earth-based perovskite oxide thin films for various applications.

## 1. Introduction

Strontium stannate (SrSnO<sub>3</sub>) is a perovskite-structured semiconducting material with a direct wide band gap ranging from 3.9 eV to 4.2 eV, enabling it to efficiently absorb light in the ultraviolet region [1]. Strontium stannate has an orthorhombic structure and it belongs to Pbnm space group, with pseudo-cubic lattice constants of  $a = 5.709 \text{ \AA}$ ,  $b = 5.703 \text{ \AA}$  and  $c = 8.065 \text{ \AA}$  and it is classified as an n-type material [2]. However, this material suffers from lower mobility as compared with barium stannate (BaSnO<sub>3</sub>) perovskite [3]. Nevertheless, the literature suggests that structural modifications

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on alkaline-earth (Ba, Sr, Ca) stannates have made them promising semiconducting materials for photodetector [4], solar cell [5] and photocatalysis [6] applications.

Strontium stannate has been synthesised using various wet-processes such as sol-gel [7], the hydrothermal method [8], the Pechini method [9], polymeric precursor method [10] and calcination [11]. In contrast, Wakana *et al.*, demonstrated the dry process synthesis of SrSnO<sub>3</sub> on a magnesium oxide (MgO) substrate using the sputtering technique [12]. This technique required a solid target material containing moulded SrSnO<sub>3</sub> powder. Plasma with the energetic inert gas argon collided with the target, resulting in the emission of SrSnO<sub>3</sub> atoms that were deposited on the substrate. The ionised inert gas argon was triggered by RF or DC power applied in the process. The sputtering power significantly influenced the density and distribution of the reactive species in the plasma, which in turn, affected the growth, nucleation and deposition rate of the thin film [13]. By controlling the power, deposition conditions, such as the film thickness, composition and surface roughness, can be optimised. For certain metal materials with high hardness, a significant amount of power, such as DC sputtering or HiPIMS add-ons, is required [14]. On the other hand, the use of RF power is preferred for softer materials, such as SrSnO<sub>3</sub>. Several single or composite materials have been investigated related to RF sputtering power's effects on material properties, including indium thin oxide (ITO) [15], silicon dioxide (SiO<sub>2</sub>) [16], aluminium gallium nitride (AlGaN) [17], zinc oxide (ZnO) [18], aluminium-doped ZnO (AZO) [19] and copper nitride (Cu<sub>3</sub>N) [20].

In this study, SrSnO<sub>3</sub> thin films were synthesised using the RF magnetron sputtering method with RF power ranging from 50 W to 100 W. The effects of different plasma power levels on the structure, surface morphology and electrical properties were examined. This paper investigated the behaviour of thin films in order to obtain a high-quality SrSnO<sub>3</sub> thin film. The results demonstrated that crystal structure characteristics can be controlled by adjusting the RF sputtering power.

## 2. Methodology

### 2.1 Fabrication

A glass-coated Indium Thin Oxide (In<sub>2</sub>O<sub>3</sub>:Sn) substrate, known as ITO of 2 × 2 cm<sup>2</sup> was prepared. The ITO/glass substrate was cleaned by placing it in an ultrasonic bath containing acetone, methanol and de-ionised water. After the cleaning process, the substrate was dried in the oven for 10 minutes at 70° C. This process was important to ensure the substrate's surface is clean from any impurities that may affect its contact quality.

In the experiment, a RF power source with varying outputs of 50 W, 80 W and 100 W at a frequency of 13.65 MHz was utilised for the deposition process. Power was limited to 100 W due to target arcing at high power level. The chamber was maintained at a base pressure of 8.5 × 10<sup>-6</sup> Torr and a working pressure of 5 mTorr. During the process, the substrate was rotated at a speed of 5 rpm per hour per sample, positioned 120 mm away from the target and set at a non-perpendicular height. A mixture of pure inert sputter gas (75% Argon) and reactive gas (25% O<sub>2</sub>) was used, while the target material was a 99.9% pure SrSnO<sub>3</sub>-depleted compound sourced from Plasmaterials Co and measured 3 inches in diameter and 0.125 inches in thickness. A single cathode, set at an angle of 45 degrees to the base and with a bias voltage of 0 V, was connected to the RF power source. To improve the efficiency of the sputtering process for depositing three substrates under different parameters, an isolation gate valve (load lock) was employed to switch the substrates.

The addition of oxygen aimed to improve the deposition of thin film by reducing oxygen vacancies. Inert gas would bombard and collide with the target atoms, freeing them into the plasma. As a result, SrSnO<sub>3</sub> atoms would move kinetically towards the substrate and form a thin film. The sputtering system has numerous components and a chamber and its ambiance is controlled by a

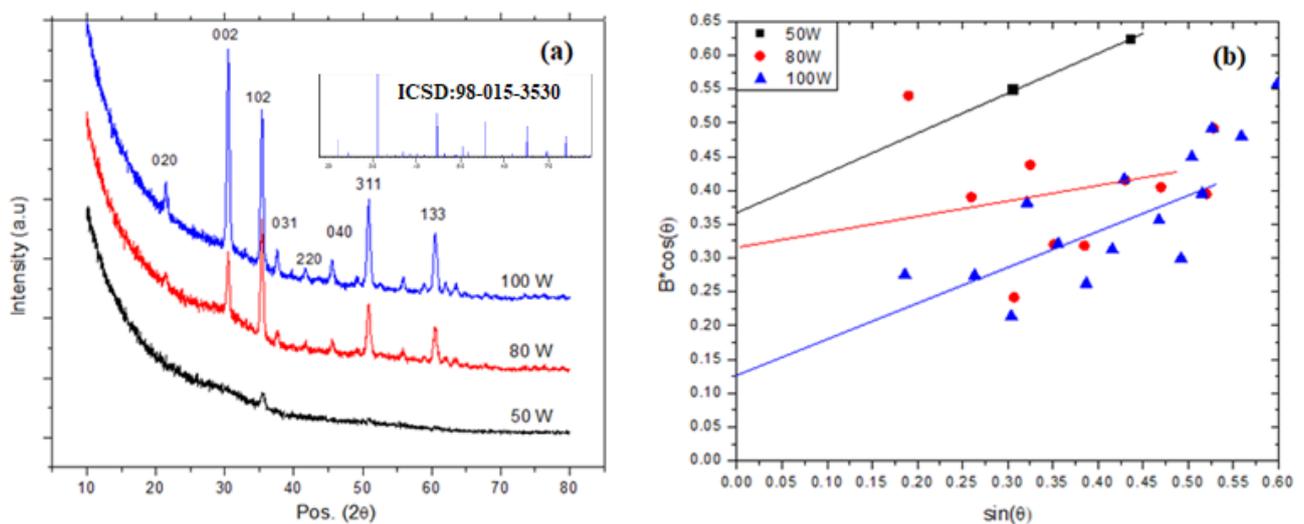
turbo molecular pump (TMP) while being monitored by a vacuum gauge to achieve a high vacuum condition for establishing plasma. This system is fully controlled and monitored by a programmable logic controller (PLC) with a LabVIEW graphical user interface (GUI).

## 2.2 Characterization

The SrSnO<sub>3</sub> films were characterised via X-Ray diffraction (XRD) (PANalytical X'Pert Pro XRD System) by employing Cu-K $\alpha$  radiation at  $\lambda=0.152$  nm. A Hitachi 5100N atomic force microscope (AFM) was used for surface morphology observation, while electrical properties were analysed using a four-point probe to obtain optical characteristics. Surface tension was observed using the VCA Optima XE Contact Angle Goniometer (AST Products Inc).

## 3. Results and Discussion

The XRD patterns for SrSnO<sub>3</sub> thin films deposited at different RF power levels are depicted in Figure 1. At low RF power of 50 W, there was a single peak at  $2\theta = 35.5^\circ$  corresponding to the (102) plane. At higher power levels (80 W and 100 W), increased ion energy and density of the inert gas argon led to deeper penetration into the SrSnO<sub>3</sub> target, resulting in a considerable particle dispersion. The preferred orientations of SrSnO<sub>3</sub> crystal peaks appeared at (020), (002), (102), (031), (220), (040), (311) and (133) planes. It should be noted that at 50 W, the (102) plane showed a higher intensity compared with that of the (002) plane. Generally, in order to achieve a homogeneous substrate surface, a low RF power could be used with a longer deposition. However, in this case, at higher RF power levels, the (002) plane was more dominant than (102) plane in terms of intensity. As shown in Table 1, the intensity of (002) plane at 80 W and 100 W evidenced a progressive growth.



**Fig. 1.** Deposition of SrSnO<sub>3</sub> at 50 W, 80 W and 100 W (a) XRD patterns (b) Williamson-Hall (W-H) plot

Figure 1(b) depicts the data extraction from the XRD patterns shown in Figure 1(a), for obtaining the average strain and average crystallite size. To determine both parameters, a Williamson-Hall plot was used, which is expressed by the following equation:

$$\beta_{total} \cos \theta = C_{\epsilon} \sin \theta + \frac{k\lambda}{D_{W-H}} \quad (1)$$

where  $\beta_{total}$  is the total full width at half maximum (FWHM) of different peaks,  $\lambda$  is the X-ray wavelength (Cu –  $K\alpha = 1.5418 \text{ \AA}$ ),  $k = 0.94$  is the shape factor,  $C_\epsilon$  is the average strain,  $D_{W-H}$  is the crystal size and  $2\theta$  is the peak position. This equation can define the slope ( $m = C_\epsilon$ ) and intercept ( $C = K\lambda / D_{W-H}$ ), as expressed by the linear equation,  $y = mx + C$ . As shown in Figure 1(b), all the positive slope trends indicate that all thin films experienced tensile strain instead of compressive strain. At the low power of 50 W, the thin film exhibited an average crystallite size was 241.5 Å which was the smallest compared with the crystallite sizes of thin films deposited at the higher power levels, which were 275 Å at 80 W and 708 Å at 100 W. This indicates that the low kinetic energy yielded by the low RF sputtering power at 50 W did not have a significant impact on crystallite size. At 50 W, the strain was 0.266% by measuring  $C_\epsilon$ , indicating that low RF power had insufficient kinetic energy to overcome the surface energy barrier of the substrate. As power increased to 80 W, the film experienced a reduction in strain to 0.1%. However, at 100 W, the strain increased to 0.24%, possibly affected by the increasing density of atoms deposited.

Table 1 shows atomic condition information at peak positions  $2\theta = 30.45^\circ$  and  $2\theta = 35.5^\circ$ . The d-spacings of these planes were relatively stable, around 2.92 Å and 2.53 Å for (002) and (102) planes, respectively. When the RF power increased, the atoms had more kinetic energy to overcome the surface energy barrier of the substrate, which promotes larger crystallite size. The microstrain surrounding the crystals also decreased with increased RF power. Generally, an increased crystallite size can improve thermal and mechanical stability. However, larger crystallite sizes can increase brittleness and reduce ductility [21].

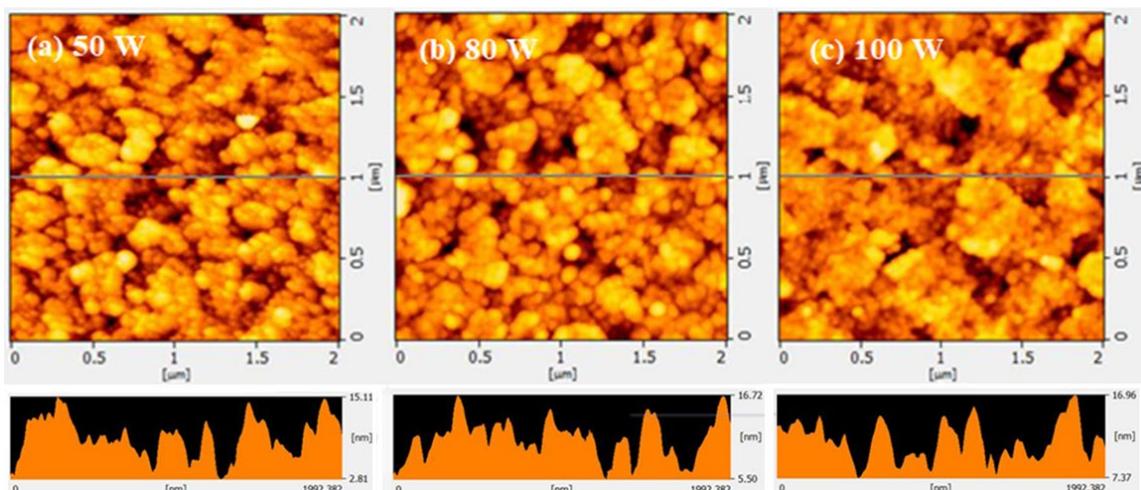
**Table 1**

XRD data of (002) and (102) peaks

| Sputtering power (watt) | $2\theta$ pos. | (hkl) | $\Delta$ Intensity* (a.u) | FWHM  | d-spacing (Å) | Crystallite Size (Å) | Micro strain (%) |
|-------------------------|----------------|-------|---------------------------|-------|---------------|----------------------|------------------|
| 50                      | $30.41^\circ$  | (002) | N/A                       | N/A   | N/A           | N/A                  | N/A              |
|                         | $35.5^\circ$   | (102) | 98                        | 0.59  | 2.52          | 160.18               | 0.78             |
| 80                      | $30.45^\circ$  | (002) | 488                       | 0.413 | 2.92          | 227.30               | 0.64             |
|                         | $35.34^\circ$  | (102) | 983                       | 0.26  | 2.53          | 364.33               | 0.34             |
| 100                     | $30.53^\circ$  | (002) | 1641                      | 0.29  | 2.92          | 322.04               | 0.45             |
|                         | $35.34^\circ$  | (102) | 1270                      | 0.23  | 2.53          | 413.26               | 0.30             |

\*: intensity peak subtracted by baseline

Figure 2 below depicts the surface images scanned via AFM.



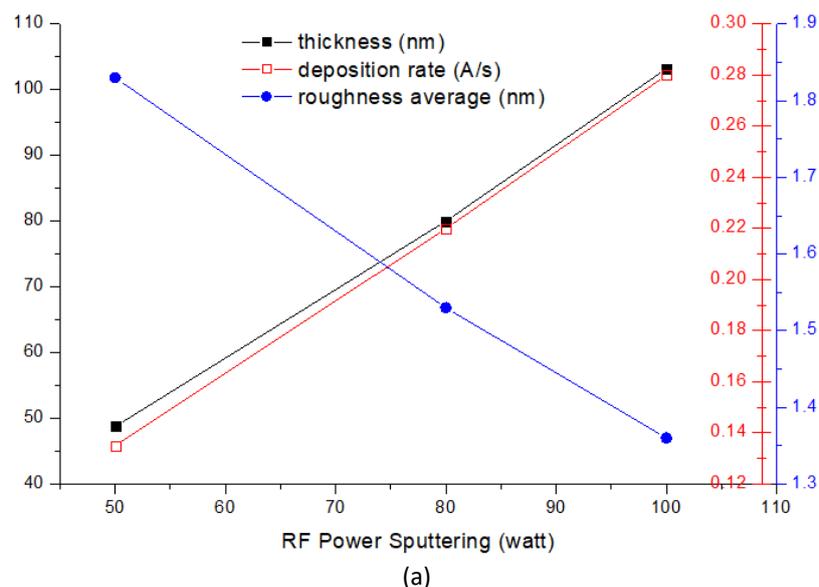
**Fig. 2.** Atomic force microscopy (AFM) topography images (a) 50 W (b) 80 W (c) 100W

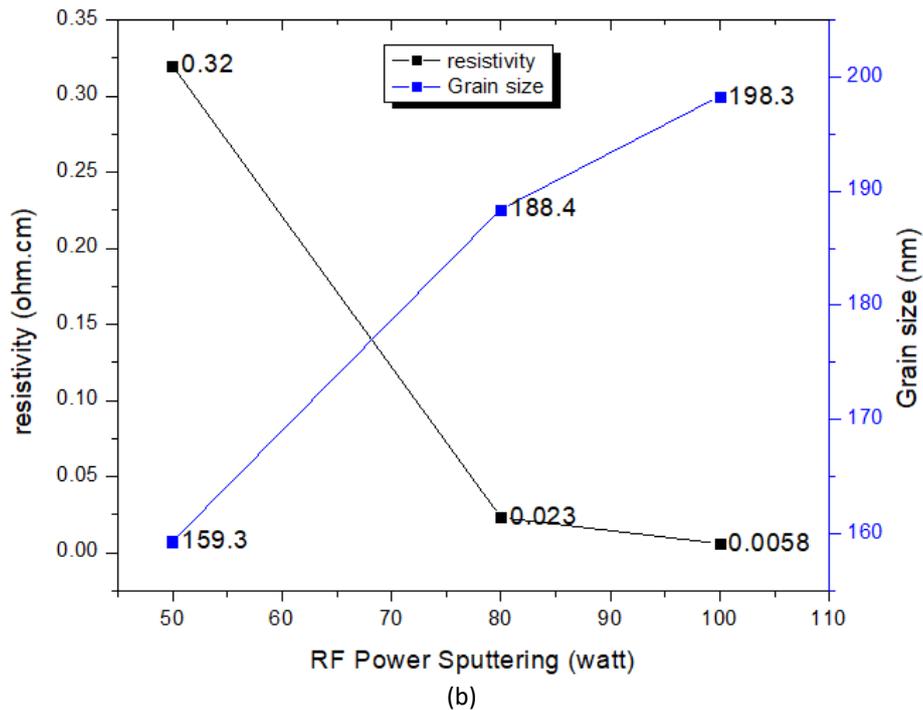
Surface characteristics were observed via AFM using a touching cantilever to scan a  $4 \mu\text{m}^2$  area. Figure 2(a) to Figure(c) depicts the surface of the films to reveal the atomic arrangement on the substrate. It should be noted that surface roughness depends on the applied RF power in the deposition process. A larger crystallite size and decreased roughness in accumulation can affect the grain size, providing the benefits of higher film conductivity or lower resistivity, as shown in Figure 3(b) [22]. High-energy ions, were generated by RF power, can also increase the surface mobility of atoms on a substrate's surface, resulting in more efficient migration and rearrangement of atoms, promoting larger and more ordered crystal structures. Vopsaroiu *et al.*, [23] revealed that RF sputtering power can control grain size for a chromium (Cr) target, where a higher power yielded larger grains. In AFM, the roughness root mean square (RMS) can be defined as  $S_q$  and is expressed as:

$$S_q = \sqrt{\frac{1}{n} \sum_{i=1}^n y_i^2} \quad (2)$$

where  $n$  is the maximum number of points scanned and  $y_i$  is the surface height at a certain point. Roughness trended lower as RF sputtering power increased, as seen in Figure 3(a). The thin films' roughness was affected by the boundaries between grains. This indicates that the higher the number of grains, the more boundaries there were, which in turn increased the scattering. There are plenty of factors that contribute to surface roughness, such as surface texture, topography and the presence of defects or impurities. Thus, grain boundary is not the only factor that contributes to surface roughness.

From Figure 3(a), it can be noted that thickness increased from 50 to 100 nm and the deposition rate increased from 0.14 to 0.28 Å/s as RF power increased. Physically, increased power can emit more atoms from a target due to a higher kinetic energy. The excessive amounts of atoms bombard the substrate immediately in the plasma's atmosphere. This behaviour has also been revealed in several materials, such as ITO, ZnO and  $\text{Cu}_3\text{N}$ , due to the effect of RF sputtering power. Figure 3(b) shows RF sputtering power against resistivity and grain size. As shown in Figure 3(b), the grain sizes of films are 159.3 nm, 188.4 nm and 198.3 nm for 50 W, 80 W and 100 W, respectively. At higher RF power levels, the grain size increased while resistivity decreased, in the thin films.





**Fig. 3.** (a) Plot of thickness, deposition rate and average roughness average versus RF sputtering power (b) resistivity and grain comparison

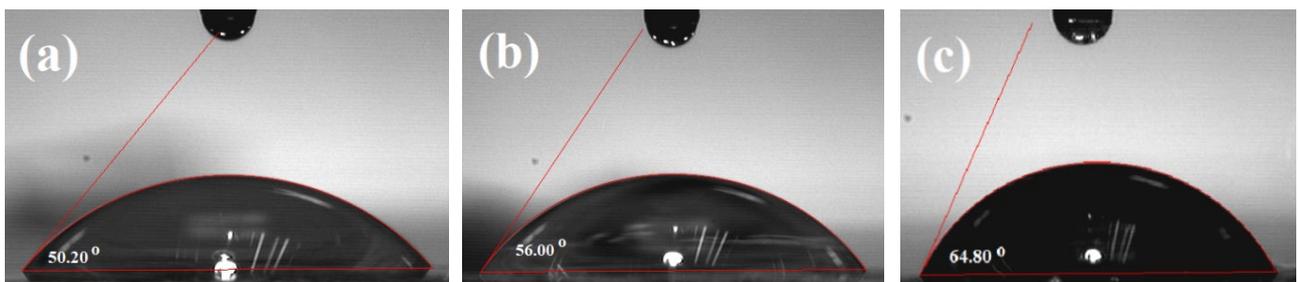
Figure 4 shows the hydrophobicity of surface characteristics measured using a goniometer. When a droplet of liquid is placed on a flat surface, the balance of the three-phase interface is expressed by Young's equation, as follows:

$$\gamma_s = \gamma_{sl} + \gamma_l \times \cos \theta \quad (3)$$

where  $\gamma_l$  is the surface tension of the liquid,  $\gamma_{sl}$  is the interfacial tension,  $\gamma_s$  is the surface energy of the solid and  $\theta$  is the contact angle between the liquid-air interface and the surface [24]. The thin films showed contact angles of  $50.2^\circ$ ,  $56.0^\circ$  and  $64.8^\circ$  when deposited at 50 W, 80 W and 100 W power levels, respectively. This indicates that all deposited films were hydrophilic. Work of adhesion or surface energy, can be determined via contact angle using the following equation:

$$W_a = g_{LV}(1 + \cos \theta) \quad (4)$$

where  $W_a$  is the work of adhesion,  $g_{LV}$  is the surface tension of water in the air (73 dyne/cm) and  $\theta$  is the contact angle. The surface energy values of the thin film deposited at 50 W, 80 W and 100 W were 119.72 dyne/cm, 113.82 dyne/cm and 104.08 dyne/cm, respectively.



**Fig. 4.** Contact angle of films at (a)50 W (b) 80 W (c) 100 W

Many researchers have reported a direct proportional relationship between surface roughness and contact angle, signifying that an increase in roughness leads to an increase in contact angle [25]. This correlation is supported by the Wenzel model, which concludes that surface roughness amplifies wettability on the original surface. However, as depicted in Figure 4, the increase in RF power weakened the work of adhesion of SrSnO<sub>3</sub> films to the substrate. This behaviour depended on the characteristics of the material.

#### 4. Conclusions

This paper presents a successful demonstration of SrSnO<sub>3</sub> thin film deposition using RF magnetron sputtering. The effect of varying RF power levels was investigated on the structure, morphology and electrical behaviour of the SrSnO<sub>3</sub> thin film. At a low RF power level of 50 W, the (102) plane exhibited a more substantial growth (98 a.u.) compared with those of other planes, indicating a lower surface barrier energy for its formation. However, the (002) plane grew progressively at the medium power (488 a.u.) and high power (1641 a.u.) due to the higher kinetic energy on the surface, whereas the (102) plane weakened at 100 W of RF power. The increase in RF sputtering power influenced crystal size (241.5 Å to 708 Å), which was related to grain size and grain boundary. Contrarily, it caused decreased surface roughness, resistivity and macrostrain. Based on Williamson-Hall analysis, it was furthermore found that the SrSnO<sub>3</sub> film exhibited tensile strain, as indicated by gradient values. The increasing RF sputtering power resulted in weakened work of adhesion or surface energy of the thin films (119.72 dyne/cm to 104.08 dyne/cm), which indicated that, at increased power, less energy was required to separate the thin film from the substrate. Based on this study, the increase in sputtering power can enhance the ion density in a plasma, which in turn leads to higher ion bombardment energy on a substrate. Thus, this promotes the nucleation of crystals and the growth of larger crystals. The nearly zero W-H linear fitting at 80 W can be optionally used as a reference to fabricate preferred films.

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