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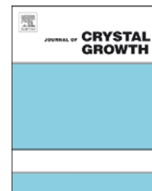

Orientation control of $LaNiO_3$ thin films by RF magnetron sputtering with different oxygen partial pressure

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Highly (1 1 0)- and (1 0 0)-oriented $LaNiO_3$ (LNO) thin films were successfully grown on Si (1 0 0) substrate using radio frequency (RF) magnetron sputtering at room temperature (RT). Effects of oxygen partial pressures on the orientation, film composition, surface morphology, and electrical properties of the films were investigated. The nearly complete (1 0 0) orientation was first achieved with oxygen partial pressure beyond 15% in the sputtering gas. The preferred (1 0 0) orientation of growing films is determined by uniform distribution of Ni^{3+} and La/Ni ratio in the films caused by oxygen during sputtering, as well as the lowest surface energy of the films in the crystalline process. LNO films with controlled orientation have low resistivity of $7.0\times10^{-6}\,\Omega\,m$ which is a good basis for integrating ferroelectric capacitors.

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1. Introduction

Perovskite-oxides have good physical characteristics in ferroelectricity, superconductivity, and giant magnetoresistance [1–3]. Recently, it is noted that conductive oxides with perovskite-type structure such as $YBa_2Cu_3O_{7-\sigma}$, $La_{0.5}Sr_{0.5}CoO_3$, and IrO_2 have excellent potential as electrodes on ferroelectric substrates for the great improvement on the fatigue and aging of ferroelectric capacitors [4–6]. Among them, pseudo-cubic perovskite lanthanum nickel oxide, $LaNiO_3$ (LNO), with a lattice parameter of 3.84 Å, is a very promising candidate, due to its good metallic conductivity and simple composition. Moreover, the use of LNO bottom electrodes has the advantage of decreasing the leakage current and polarization fatigue of the $Pb(Zr,Ti)O_3$ ferroelectric films [7].

Considering its effect on the properties of the subsequent functional oxides deposited on it, the preparation of LNO films on Si (1 0 0) having a good conductivity and controlled orientation is highly desirable. LNO films with controlled crystalline orientation served not only as a promising bottom electrode material in various ferroelectric related devices [8–10], but also as a seed layer for the growth of highly textured ferroelectric thin films [11–13]. Therefore, the investigation on the orientation control of LNO films is an intriguing subject of great importance. The orientation control of LNO films on the Si substrate has been carried out by a variety of chemical and physical approaches. Usually, the chemical solution deposition methods can prepare both (1 0 0)- and (1 1 0)-oriented LNO films by modifying the concentration or solvent of the precursor solution [14,15] or by changing the heating rates of the temperature range of 200–400 °C [16]. Pulsed laser deposition (PLD) methods can obtain (1 0 0)- and (1 1 0)-oriented and polycrystalline LNO films using an oriented MgO film as a buffer layer [17], while the radio frequency (RF) magnetron sputtering methods often result in (1 0 0)-oriented LNO films by heating substrate from 150 to 600 °C [18–20] and (1 1 0) oriented LNO films by cold deposition [21]. However, the (1 0 0)-textured nature of the LNO film was gradually destroyed as the substrate temperature increased. It arises from the formation of two addition La-rich phases, i.e., La_2NiO_4 and $La_4Ni_3O_{10}$, which was induced by the loss of Ni during the course of high-temperature deposition [18]. Therefore, it is of great significance to control the growth of preferred texture of LNO thin film at room temperature (RT) without decomposition. However, there is still lack of reports about the LNO films with preferential (1 0 0) orientation sputtered at RT. The purpose of this work is to investigate the possibility of orientation control growth of LNO

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films and clarify the changes in composition, crystal structure, and resistivity of the films with oxygen partial pressures. The orientation mechanism is also discussed in detail.

2. Experiment

LNO thin films were deposited on Si (1 0 0) substrates by RF magnetron sputtering at RT. A 3-in.-diameter target for LNO thin films was made by using a conventional mixing-oxides method, which included calcining La_2O_3 and Ni_2O_3 powders (both of 99.99% purity). The sintering was carried out at 1180 °C for 2 h. The composition of the target was set to La/Ni = 1:1.1 to get stoichiometric composition in the resultant LNO thin films, which was determined on the basis of results of La/Ni ratio (minimum of 1.1) in the films using an LNO target of stoichiometric composition [21]. The sputtering parameters are listed in Table 1.

The crystalline phases developed in the films were examined by X-ray diffraction (XRD) (JOEL Rigaku D/Max) measurements and the compositions of the films were analyzed using inductively coupled argon plasma atomic emission spectrometry (ICP-AES). The thicknesses of the films were measured by field emission scanning electron microscope (SEM) (JEOL JSM-6700F). Atomic force microscopy (AFM) was used to characterize the surface images and to measure the roughness of the films. The RT electrical resistivity of LNO films were measured by a four-point probe (Model: Keithley 5805 Kelvin Probes) and a Keithley 2001 multimeter.

3. Results and discussion

Fig. 1 shows the XRD patterns of LNO thin films deposited at RT with (a) $O_2/(Ar+O_2) = 0$ and (b) $O_2/(Ar+O_2) = 20\%$ followed by RTA at temperatures from 450 to 700 °C. The perovskite LNO films with preferential (1 1 0) orientation were obtained for the case of the sputtering gas with pure argon at above the annealing temperature of 550 °C, and the perovskite LNO films with highly (1 0 0) orientation were deposited at above the temperature of 450 °C if 20% of oxygen partial pressure was used for sputtering. On the other hand, the LNO films exhibited relatively good crystallinity if annealed at 700 °C, independent of the oxygen.

Relations of lattice parameter and full-width at half-maximum (FWHM) of (1 1 0) and (1 0 0) peak with annealing temperature are shown in Fig. 2. (a) $O_2/(Ar+O_2) = 0$ and (b) $O_2/(Ar+O_2) = 20\%$. A decrease of lattice parameter with annealing temperature was observed for both cases. Note that the values were lower than the LNO bulk lattice parameter (3.84 Å) for LNO films deposited with pure argon, indicating tensile stress in the films. A decrease of FWHM with annealing temperature from 450 to 700 °C was also observed, which could be due to an increase in grain size and decrease of defects. This conformed LNO crystallization improvement with annealing temperature is shown in Fig. 1.

Table 1
Summary of deposition and annealing parameters.

Parameters	Range
Background (Pa)	5×10^{-4}
Working pressure (Pa)	2.2
Substrate temperature (°C)	RT
Power density (W/cm^2)	1.8
Sputtering time (min)	60
Atmosphere $O_2/(Ar+O_2)$	0; 5%; 10%; 15%; 20%
Annealing temperature (°C)	450–700 °C/10 min in O_2

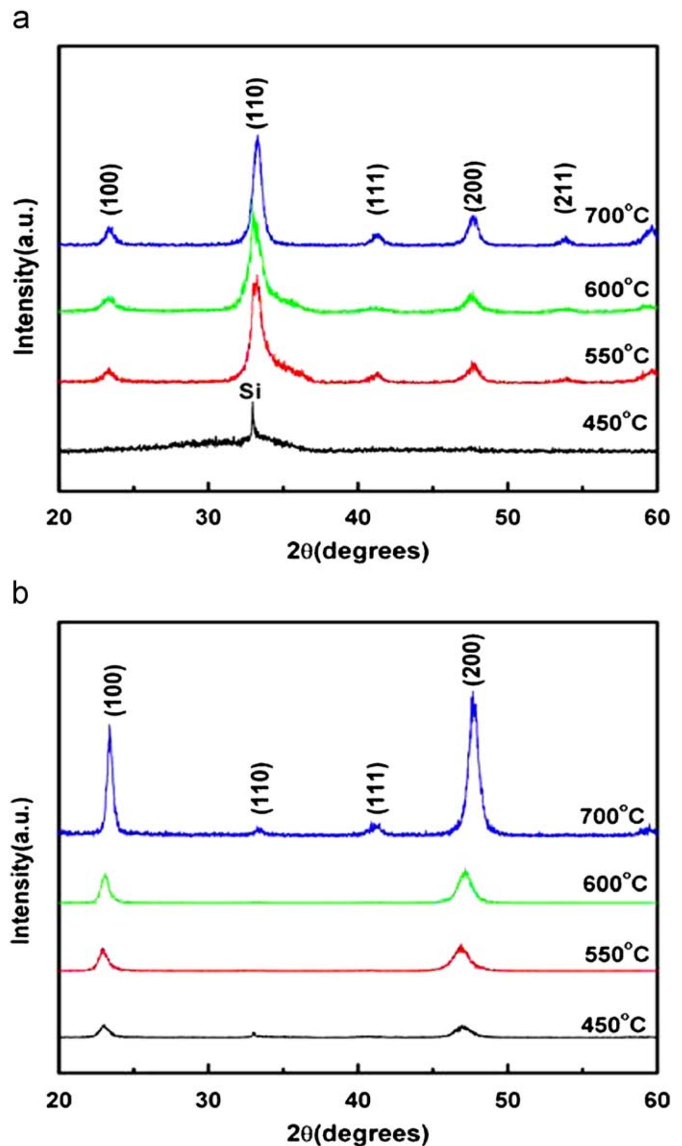

Fig. 1. XRD patterns of LNO thin films deposited at RT with (a) $O_2/(Ar+O_2) = 0$ and (b) $O_2/(Ar+O_2) = 20\%$ followed by RTA at temperatures from 450 to 700 °C.

The influence of oxygen partial pressure on orientation was then investigated. Fig. 3 shows the XRD patterns of a series of LNO films deposited at various oxygen partial pressures (total pressure is 2.2 Pa), annealed at 700 °C. It was interesting to find that the oxygen partial pressures have a considerable influence on the orientation of LNO films. It can be seen that LNO films with higher oxygen partial pressure (higher than 15%) have a preferred (1 0 0) orientation, while the LNO films with lower oxygen partial pressure (lower than 10%) have a preferred (1 1 0) orientation. In the case of oxygen partial pressure of 5 and 10%, the intensity of (1 1 0) was weak due to the thicknesses of films, only 90 and 100 nm, respectively. Wang et al. [11] have reported that tensile stresses arise within the film caused by the substrate if the film was produced with a thickness that was smaller than a critical value (120 nm). It was believed that the thickness changing was mainly due to the differences of oxygen partial pressures.

The (1 0 0)-orientation parameter, α_{100} was calculated from the relative heights of the (1 1 0) and (1 0 0) reflection, i.e.,

$$\alpha(1\,0\,0) = \frac{I^f(1\,0\,0)/I^f(1\,1\,0)}{I^p(1\,0\,0)/I^p(1\,1\,0)} \qquad (1)$$

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