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Preparation of $LaNiO_3$ thin films with very low room-temperature electrical resistivity by room temperature sputtering and high oxygen-pressure processing

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Abstract

$LaNiO_3$ (LNO) thin films were deposited by radio frequency magnetron sputtering on n-type Si (100) wafers at room temperature (RT). The as-sputtered LNO thin films were amorphous and had very high RT electrical resistivity even after post-annealing at 800 °C. The amorphous as-sputtered LNO films could be transformed to polycrystalline LNO films in rhombohedral phase by heating at 400 °C in an O_2 atmosphere at pressure ranging from 1.5 to 8.0 MPa. Very low RT resistivity of LNO films were obtained by this high oxygen-pressure processing. The lowest value was as low as 1.09×10^{-4} Ω cm by processing at oxygen pressure of 8 MPa. Such preparation of LNO thin films is compatible with the Si-based readout integrated circuits. Highly (100)-oriented perovskite structure of $Pb(Zr_{0.52}Ti_{0.48})O_3$ thin films was formed on this rhombohedral phase LNO, and good ferroelectricity could also be obtained on these HOPP-processed rhombohedral phase LNO films.
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Keywords: Sputtering; High oxygen-pressure processing (HOPP); Amorphous; $LaNiO_{3-\delta}$ thin film

1. Introduction

Recently, metallic oxide with perovskite-type structure have attracted much attention as candidates for electrodes in thin films, which are used for applications in ferroelectric memories, pyroelectric imaging sensors, optical waveguides, and micro-actuators [1–7]. These perovskite-related lattice-matched (with ferroelectric thin films such as $PbTiO_3$, $Pb(Zr,Ti)O_3$ (PZT), and $(Pb,La)(Zr,Ti)O_3$) thin films (such as IrO_2, RuO_2, and $YBa_2Cu_3O_{7-x}$), bearing thus a better crystallographic compatibility with ferroelectric layer at the interface compared to metal electrodes, were considered to be the promising alternatives against fatigue and aging due to oxygen deficiency of ferroelectric thin films [8–10].

$LaNiO_3$ (LNO) is also one of these perovskite related Pauli paramagnetic and metallic oxide [11–14]. The primitive cell of LNO consists of two formula units. One is rhombohedral with lattice parameter $a=0.5461$ nm and the rhombohedral angle 60.41° [11], the other is pseudocubic with $a=0.384$ nm, and it shows a metallic behavior without any doping [15]. LNO thin films have been fabricated by pulsed laser deposition (PLD) [16,17], sputtering [18,19], and metalorganic decomposition [20–25]. Most of LNO films are amorphous when deposited below temperatures of 400–600 °C, but crystallized in cubic [17,26–28] or rhombohedral [29,30] phases when grown or post-processed at higher temperatures. The films deposited by sputtering seem to be an exception, since they are crystalline below 400 °C and even at temperatures as low as 150–250 °C [30,31]. The widely reported room temperature electrical resistivities of LNO films were in the range of $1.5–5\times10^{-4}$ Ω cm for epitaxial films and $5–26\times10^{-4}$ Ω cm for polycrystalline,

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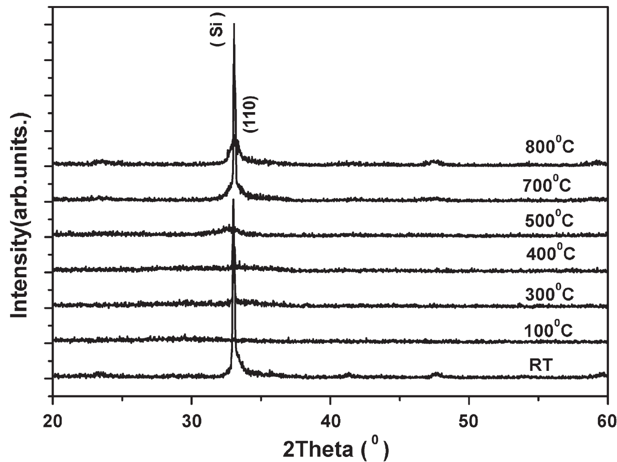

Fig. 1. XRD diffraction patterns of $LaNiO_3$ films sputtered on Si (100) substrates at room temperature and followed by annealing processing (RTA) at various temperatures as indicated in the figure.

respectively. The lowest resistivity reported, 0.5×10^{-4} Ω cm, was found in film deposited on $SrTiO_3$ (100) by PLD [32]. There were no any report about sputtering at substrate temperature of RT. In this work, we report the studies about radio frequency (r.f). magnetron sputtering at RT and subsequent high oxygen-pressure processing (HOPP) for LNO thin films.

In order to test a preparation process compatible with Si-based readout integrated circuit (ROIC), we used the commercial Si (100) wafers (n-type, 5–8 Ω cm) as the substrates, and sputter-deposited and processed LNO films below ROIC's critical temperature 450 °C.

2. Experimental details

The LNO films were obtained by r.f. magnetron sputtering under the deposition parameters of ~1.6 Pa for working pressure, and 40% for oxygen partial pressure with mixed argon and oxygen gas and RT for substrate temperature (neither heating nor cooling). A stoichiometric ceramic target with a diameter of 100 mm was made by sintering calcined powder at 1000 °C for 3 h using a conventional mixing-oxides method, and target to substrate distance is 7 cm. The total r.f. output power was set at 80 W. The as-sputtered LNO films were post-annealed at 100 °C, 300 °C, 400 °C, 500 °C, 700 °C, and 800 °C respectively by rapid thermal annealing (RTA) method for 600 s in dry air atmosphere. The RTA-processed LNO thin films remained amorphous even at annealing temperature up to 800 °C. Afterward the as-sputtered LNO films were loaded into a stainless steel cylindrical pressure chamber, this chamber was put into a purpose-designed cylindrical muffle furnace. The furnace was heated from room temperature to 400 °C while the pressure chamber was filled with oxygen (99.998%). Temperature was maintained at 400 °C for 5 h. Oxygen pressure of 1.5 MPa, 3 MPa, 5 MPa, 7 MPa, and 8 MPa were tested.

To investigate the application of these RT sputter-deposited and high oxygen-pressure processed LNO thin films as bottom electrodes to ferroelectric thin films such as PZT, we deposited $Pb(Zr_{0.52}Ti_{0.48})O_3$ films (~300 nm) on samples that were sputter-deposited at substrate temperature of 200 °C (LNO1) and sputter-deposited at RT and submitted to HOPP (LNO2). The PZT films were sputter-deposited on LNO1 and LNO2 at 580 °C, then Pt top-electrodes were patterned by lithography and direct current sputtering.

The LNO film thickness was measured with a Dektak profilometer. XRD characterization of the LNO thin films was performed using a Rigaku diffractometer (D/Max-ra) with Cu K_α radiation in the θ–2θ scan mode with 2θ ranging from 20° to 60°. The RT electrical resistivities of LNO film were measured by the four-point probe method. Surface morphologies were observed by field-emission scanning electron microscopy (FE-SEM) (Philips xl 30FEG, operating voltage: 10.0 kV) and an atomic force microscopy (AFM) (MultiMode Scanning Probe Microscope, nanoscope IV/controller from Veeco instruments). (working in tapping mode (tip radius: 5–10 nm (nominal), scan rate: 1.4–1.6 Hz). The ferroelectricity of the films was investigated using a Radiant Technology ferroelectric tester used in virtual ground mode.

3. Results and discussions

Fig. 1 shows the XRD patterns of a series of LNO films deposited at RT by r.f. magnetron sputtering and followed post-annealing by RTA at various temperatures. The XRD diffractograms indicated all the LNO films were amorphous from as-sputtered at RT to post-annealed films at various temperatures as indicated in the figure. In spite of the very weak (110) peak of 800 °C post-annealed film, it also indicated the poor crystallinity. The RT resistivities of these LNO films were very high (~5.90×10^{-2} Ω cm), which were performed by a four-probe measurement and these samples were inapplicable to electrodes.

Fig. 2 shows a typical XRD patterns of a series of LNO films, which were initially deposited on Si (100) substrate at RT by r.f. magnetron sputtering and subsequent processing by HOPP at various oxygen pressure as indicated in the figure. The XRD

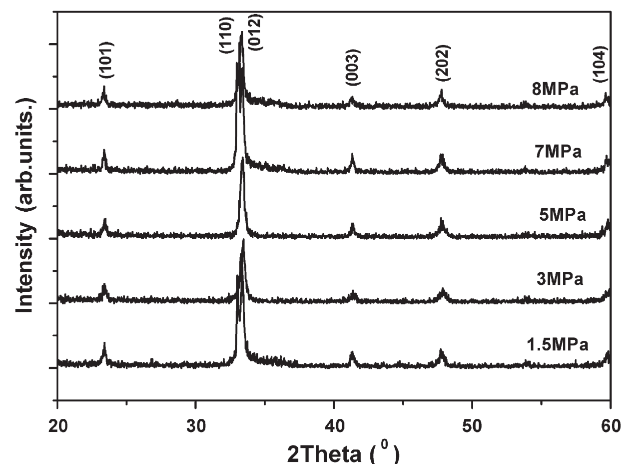

Fig. 2. XRD diffraction patterns of $LaNiO_3$ films sputtered on Si (100) at room temperature and following processed at 400 °C by HOPP at various oxygen pressure as indicated in the figure.

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