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Full Length Article

Metal-insulator transition in epitaxial $NdNiO_3$ thin film: A structural, electrical and optical study

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ABSTRACT

$NdNiO_3$ thin film has been prepared by pulsed laser deposition on $LaAlO_3$ (001) single crystalline substrate. Temperature-dependent resistivity measurement shows a sharp metal-insulator transition in such thin film. The phase transition temperature can be tuned from 90 K to 121 K by changing the thickness of thin film. The structure evolution during phase transition is studied by Raman spectroscopy. Optical conductivity reveals that the variation carrier density in the process of phase transition. The results of structural, electrical and optical studies provide useful insights to understand the mechanism of metal-insulator transition of $NdNiO_3$ thin film.

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1. Introduction

Materials with tunable metal–insulator transition (MIT) attract growing interest in various applications, such as sensors, electronic switches, thermochromic coatings, non-volatile memory, etc [1–4]. Among several MIT materials, correlated oxides such as rare earth nickelates ($RNiO_3$, R = Rare Earth) that undergo temperature-induced first order MIT, where the MIT temperature (T_{MI}) increases with decreasing cation radius (increasing R atomic number) [3], has been intensively studied in recent years. As more scientific and technological importance create increasing interests in the research of $RNiO_3$, it is difficult to be prepared this material in bulk form because it requires very high temperatures and high oxygen pressure in synthesis process [5]. Thin film deposition provides an opportunity to stabilize the metastable phases and enables a near single crystal films under epitaxial stabilization. Besides, certain strain states can be achieved in thin films by choosing the substrate of compatible lattice parameters [6–8], which may induce the desired compressive or tensile strain in the films [9,10]. Furthermore, the stain can also be manipulated by conveniently adjusting thin film thickness[11]. $RNiO_3$ thin film is, therefore, an ideal candidate to investigate the behavior of MIT when subjected to confinement, lattice mismatch and strain. It has been reported that different strain states can change the phase transition properties of $RNiO_3$ thin film. For example, the transport properties of $NdNiO_3$(NNO) thin films are altered by the changing of lattice mismatch of the film with substrates [12,13]. Keimer's et al. have carried out a series of depth resolved Raman scattering measurements to investigate the lattice dynamics of ultra-thin films of $LaNiO_3$ [14]. Catalan et al. found the thickness dependence of the MIT of NNO film on $LaAlO_3$(LAO) substrate [15,16].

In this paper, a group of NNO thin films with different thicknesses were prepared on $LaAlO_3$ substrate by pulsed laser deposition (PLD) method. The structural, electrical and optical properties are studied by combining X-ray diffraction, resistivity, Raman and infrared spectroscopy. The results provide insights into understanding the temperature and thickness effects on the metal-insulator transition of NNO thin film.

2. Experiment

To prepare the target material for PLD deposition [17], the precursor compounds, Nd_2O_3 (99.99%) and NiO (99.99%) were weighed in the correct proportions and mixed together. The mixture was firstly wet ground in ethanol and then dry ground. Both grindings were for 20 min using an alumina mortar and pestle. The ground powders were pressed into pellets and were annealed in two different steps [18]. The first annealing step was at 1000 °C for 12 h and the second step was at 1200 °C for 24 h. Between each annealing step, the pellets were re-ground and re-pressed as described above. A KrF-Excimer Laser (LPX200, Lambda Physik) with a wavelength of 248 nm and an energy density of 2 J/cm^2 was used for the thin film

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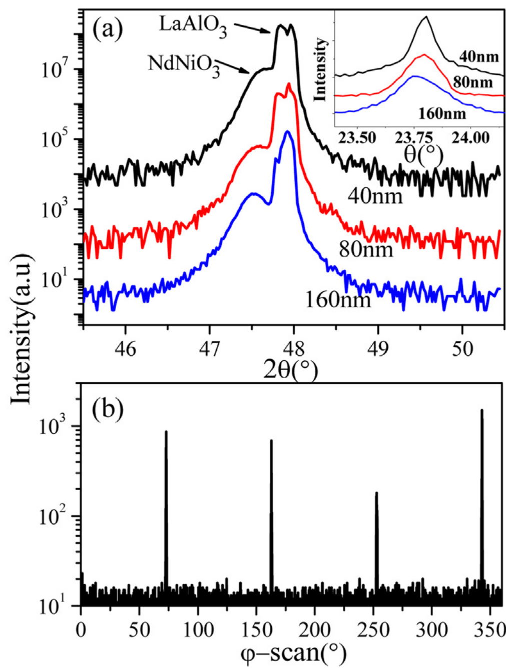

Fig. 1. (a) (002) diffraction peak and rocking curve(insert) of NNO thin film deposited on LAO substrate, (b) The φ-scan curves of the NNO film at 80 nm thickness.

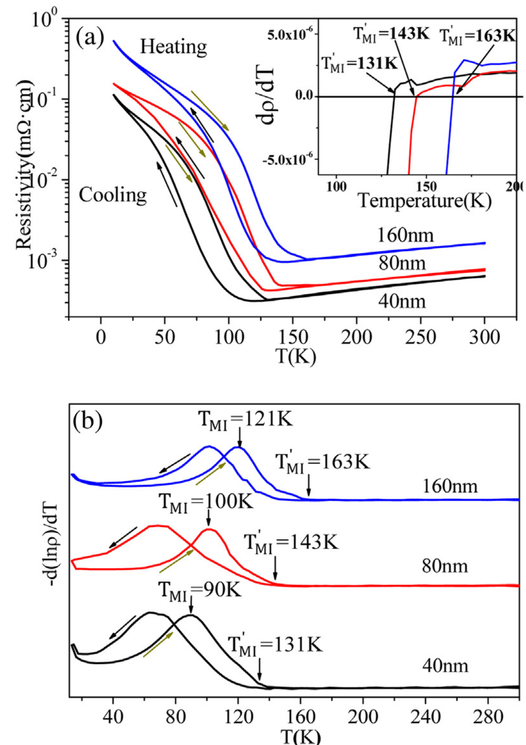

Fig. 2. Measured resistivity ρ (a) and −d(lnρ)/dT as a function of temperature (b) for NNO films at different thickness. Inset: dρ/dT as a function of increasing temperature for NNO films at different thickness.

deposition. The different thickness of NNO films was achieved by alternating the deposition time. The chamber was turbo-molecular pumped to a vacuum of 2.4×10^{-4} Pa, then backfilled with O_2 to a pressure of 3 mbar. During the growth, the substrate was kept at a temperature of 710 °C. After deposition, the pressure of O_2 was increased to 300 mbar and the thin film was kept at 710 °C for 15 min before being cooling to room temperature at a rate of 20 °C/min. The above optimized conditions ensure us to obtain the best stabilized the NNO thin film samples.

The structural properties of the different thickness samples were characterized by X-ray diffraction (XRD). The thin film thickness was determined to be 40 nm, 80 nm and 160 nm by measuring the cross-section of the thin film using scanning electron microscope (SEM), corresponding to the deposition time of 2.5 min, 5 min and 10 min, respectively. The temperature-dependent resistivity of the samples was measured by the four-probe technique using a Physical Property Measurement System (PPMS). The temperature-dependent Raman spectra were recorded with an inVia-Reflex spectrometer with triple monochromatoron. The excitation source was 532 nm line of an Ar^+ laser. The temperature-dependent infrared reflectivity was measured using a Bruker IFS 66 v FTIR spectrometer on an infrared beamline (BL01B) at the National Synchrotron Radiation Laboratory(NSRL), China. The sample was mounted on the cold finger of a continuous flow cryostat, and the measurement temperatures from the room temperature to 78 K, which included the metal-insulator transition region.

3. Result and discussion

Fig. 1(a) shows the XRD patterns of (002) diffraction plane for NNO/LAO thin films at three kinds of thickness. For the bulk NNO and LAO, both (002) diffraction peaks appear at 2θ∼48° [19,20]. For the thin film, the (002) peaks of NNO film and LAO substrate do not overlap, indicateing a small lattice mismatch between the film and the substrate [21,22]. This mismatch causes an in-plane compressive strain (∼0.75%) in thin film and results in the NNO diffraction peak appears at a lower 2θangle. The peak shifts to lower angle when the film thickness is increased, indicating an increase of out-of-plane lattice constant. The rocking curves are shown in the insert of Fig. 1(a). It is clear that the thicker film has broader peak suggesting higher mosaicity. This is consistent with strain relaxation causing increasing defects as the symmetry of parts of the film returns to bulk-like O—Ni—O bond angles. The in-plane epitaxial relationship between the NNO film and the LAO substrate was further examined through XRD φ-scans of the NNO(101) planes as shown in Fig. 1(b). Four sharp peaks are observed in φ-scan results,which confirms that the NNO film has four-fold symmetry corresponding to a cube-on-cube growth direction, suggesting the epitaxial nature of the NNO film on the LAO substrate [23].

In order to evaluate the phase transition property of the NNO thin film, temperature-dependent resistivity for different thickness NNO thin films was measured and shown in Fig. 2. It is clear that a sharp insulator-metal transition occurs from a high resistivity insulate state to a metallic state with a hysteresis behavior across the phase transition boundary. To quantitatively compare the difference of these samples, the resistivity-temperature curves were further analyzed, and the following parameters such as $T_{MI}T'_{MI}$,ransition sharpness, transition hysteresis, and quality of the metallic state were obtained and listed in Table 1. The definition of these parameters adopts the same convention, as shown in Ref [24], where T_{MI} corresponds to the temperature associated with the −d(lnρ)/dT maximum on heating, which is shown in Fig. 2(b). T'_{MI}s the temperature at which the sign of dρ/dT changed, which is shown in the insert of Fig. 2(a). Transition sharpness is defined as the magnitude of the peak height of −d(lnρ)/dT on heating. The quality of the metallic state is quantified by normalized resistivity slope at high temperature (e.g.-d(lnρ)/dT at 300 K). Transition hysteresis represents the T_{MI} difference between heating and cooling processes. According to the results, the T_{MI} increases from 90 K to 121 K and the hysteresis between heating and cooling curves changes from 7 K to 19 K when the thin film thickness changes

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