

Supporting Information

Epitaxial Ternary Nitride Thin Films Prepared by a Chemical Solution Method

Hongmei Luo,^{,†} Haiyan Wang,[‡] Zhenxing Bi,[‡] David M. Feldmann,[†] Yongqiang Wang,[§] Anthony K. Burrell,[†] T. Mark McCleskey,[†] Eve Bauer,[†] Marilyn E. Hawley,[§] and Q. X. Jia^{*,†}*

[†] Materials Physics and Applications Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545

[‡] Department of Electrical and Computer Engineering, Texas A&M University, College Station, Texas 77843

[§] Materials Science and Technology Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545

1. Sample Preparation

The precursor for the growth of SrTiN₂ (STN) films was a mixture of aqueous solutions of Sr and Ti bound to polymers. Both polyethyleneimine (PEI) and ethylenediaminetetraacetic acid (EDTA) were purchased from BASF Corporation of Clifton, NJ and were used without further purification. The two solutions were separately purified by repeated Amicon filtration that retains the high molecular weight polymer with bound metal atoms, while allows any low molecular weight (<10,000 g/mol) species to pass through. Metal analysis was conducted using a Varian Liberty 220 inductively coupled plasma–atomic emission spectrometer (ICP-AES), following the standard SW846 EPA (Environmental Protection Agency) Method 6010 procedure.

For the Sr solution, 1 g of EDTA and 1 g of PEI were dissolved in 30 mL of water, followed by addition of 1 g of Sr(NO₃)₂. For the Ti solution, small amounts of the titanium solution (2.5 g titanium tetrachloride was added slowly to a mixture of 2.5 g of 30% peroxide in 30 mL of water) were added to a solution containing 1 g PEI, 1g EDTA and 30 mL water until precipitation occurred (the pH was maintained at 7.5). After filtration, the final Sr and Ti concentration were 157 and 408 mM, respectively. The precursor solutions with the desired stoichiometric molar ratio of Sr/Ti = 1 from Sr and Ti solutions were spin-coated on (001) LaAlO₃ (LAO) substrates at 2000 rpm for 30 s. The films were annealed in ammonia gas at 1000 °C for 1 h (or 900 °C for 3 h). Films with a thickness in the range of 30-40 nm were obtained from one spin-coat. Thicker films could be deposited by multiple spin-coats.

2. Sample Characterization

X-ray diffraction (XRD) was used to characterize the crystallographic orientation of the films. The surface morphology and surface roughness of the films were analyzed by scanning electron microscopy (SEM) and atomic force microscopy (AFM). The microstructure of the films was analyzed by transmission electron microscopy (TEM). Rutherford backscattering spectrometry (RBS) with 2 MeV $^4\text{He}^+$ beam was used to measure atomic ratio between metals in the film. The backscattering silicon detector was located at 167 degree from the beam direction. The analysis did not attempt to measure N amount. Nuclear resonances at 3.045 MeV and 4.28 MeV $^4\text{He}^+$ beam energies were used to measure oxygen and carbon contaminations in the film, respectively. The optical properties of the films were examined by ultraviolet-visible (UV-Vis) transmittance and absorption at room temperature. The electrical resistivity (ρ) was measured from 5-300 K using a standard four-probe technique by a Quantum Design Physical Properties Measurement System (PPMS).

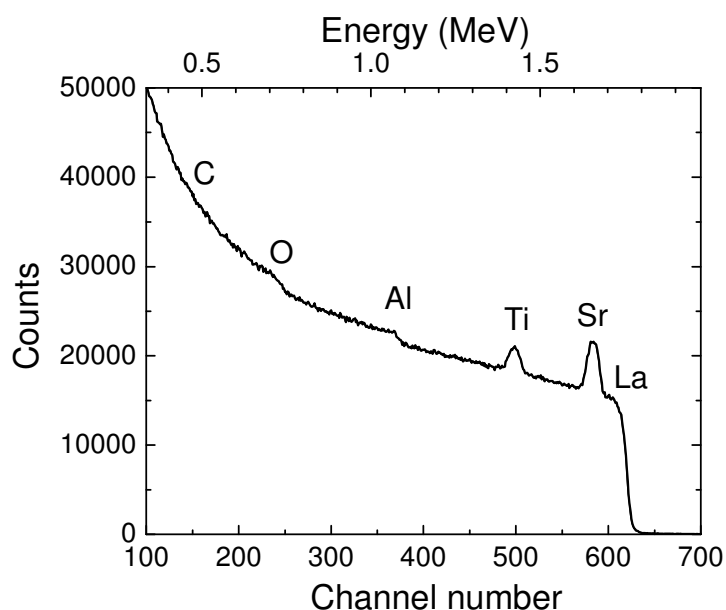


Figure S1. RBS spectrum of a STN film on LAO. This spectrum was taken with 2 MeV $^4\text{He}^+$ when the film was tilted 60 degree from the beam direction to enhance the film signals on top of the substrate. A little amount of carbon and oxygen (should be less than 10 %) were detected in the film.

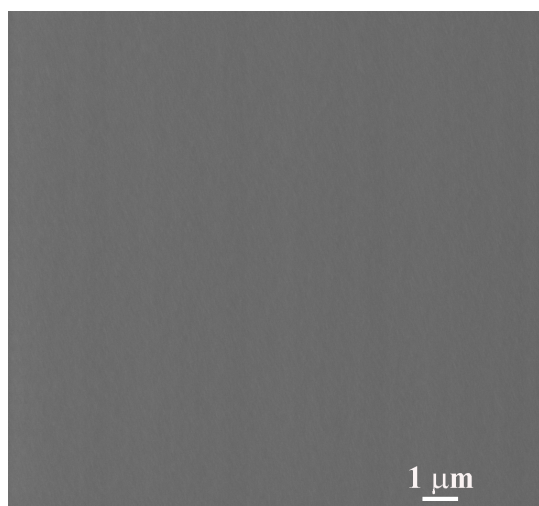


Figure S2. SEM image of a STN film on LAO.

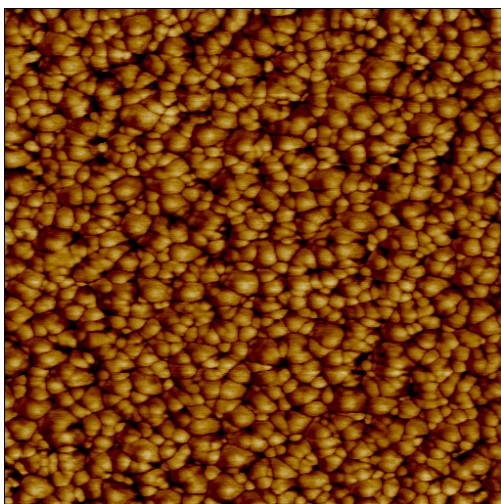


Figure S3. AFM phase image of a STN film on LAO ($1\ \mu\text{m} \times 1\ \mu\text{m}$).

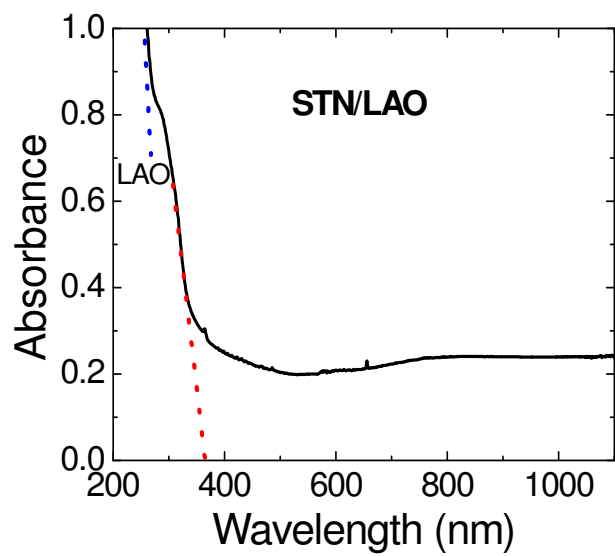


Figure S4. Absorption spectrum of a STN film on LAO.