

Interfacial effects on the electrical properties of multiferroic BiFeO₃/Pt/Si thin film heterostructures

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Received 14 February 2005; received in revised form 2 June 2005; accepted 7 June 2005

Available online 11 July 2005

Abstract

Polycrystalline BiFeO₃ thin films of various thickness were fabricated on (111)Pt/Ti/SiO₂/Si substrates via chemical solution deposition. The electrical properties were investigated using impedance and leakage current measurements. X-ray photoelectron spectroscopy (XPS) combined with Ar ion milling (depth profiling) was used to investigate elemental distribution near the electrode–film interface. It is shown that the dielectric constant depends on film thickness due to the presence of an interfacial film–electrode layer evidenced by XPS investigation. Direct current conductivity is found to be governed by Schottky and/or Poole-Frenkel mechanisms.

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PACS: 68.35.Fx; 77.84.-s

Keywords: Bismuth ferrite; Dielectric properties; Leakage currents; Interfaces

1. Introduction

The structural and electrophysical properties of bismuth ferrite (BiFeO₃) have been investigated during the last decades due to simultaneous presence of ferroelectricity and antiferromagnetic order with weak ferromagnetism in this material [1–5]. Bismuth ferrite belongs to perovskite class of complex oxides with rhombohedrally distorted cell that can be described by the polar *R3c* space group. Apart from the fact that the properties and magnetic structure of BiFeO₃ are of great interest from fundamental point of view [6,7], the possibility to create new electronic devices is also discussed [8]. In the last years, particular attention has been paid to BiFeO₃ thin films [9–13] since they allow direct integration of the material into up-to-date semiconductor technology. Successful fabrication of epitaxial [9] and polycrystalline [10–13] thin films has been reported.

A distinctive feature of ferroelectric thin films is the dependence of dielectric properties on film thickness. A number of experimental results published show that the dielectric constant of the films decreases with film thickness. For example, Paek et al. [14] reported some degradation of dielectric and leakage properties of BST ((Ba,Sr)TiO₃, barium strontium titanate) thin films of thickness ranging from 50 to 300 nm deposited on platinum electrode. They reported high resolution transmission electron microscopy investigations of a very thin layer (of thickness between 9.5 and 11 nm) with crystallinity different from that of the bulk film. This layer is claimed to have a significantly lower dielectric constant, and to contain a high concentration of defects (which, in turn, serve as an effective trap centre for mobile charges and domain walls). Such a layer has been made responsible for the thickness dependence of the dielectric properties, especially for very thin films (thinner than 200 nm). It is thought that the formation of dead layers occurs rather at film–bottom electrode interface than film–top electrode interface since the former is exposed to high temperature during film fabrication.

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Leakage current properties constitute another critical aspect limiting the applicability of ferroelectric thin films in electronic devices. A profuse literature has been devoted to the analysis of leakage currents in ferroelectric thin films. The basic conduction mechanisms in dielectric films together with an exhaustive literature review can be found in textbooks [15,16]. Following Scott [15], different mechanisms including Schottky currents and quantum mechanical Fowler-Nordheim currents (surface-limited), as well as Poole-Frenkel and Space-Charge-Limited Currents (bulk-limited) can all be present in the same temperature and/or voltage ranges [15].

Analysis of the literature shows that dielectric and leakage current properties of ferroelectric thin films were mainly investigated on PZT (*lead zirconate titanate*), BST and SrTiO_3 , and were not addressed for BiFeO_3 . In view of growing practical importance of multiferroic materials (especially in the form of thin films), attempt is made in this work to systematically study the dielectric and leakage current properties of chemical solution deposited BiFeO_3 thin films, and to understand the mechanisms of electrical conductivity. Additionally, the results of XPS analysis (X-ray photoelectron spectroscopy) together with depth profiling are presented and discussed with emphasis on interfacial composition and interface formation between Pt bottom electrode and BiFeO_3 film.

2. Experimental details

Bi-acetate and Fe-acetylacetonate were weighted according to $\text{Bi}_{1.02}\text{FeO}_3$ stoichiometry and dissolved in 1:2 mixture of deionized water and acetic acid. The final concentration of the precursor solution was 0.25 mol/l. Thin films were spin-coated at 5000 rpm for 30 s on commercial (111)Pt/Ti/SiO₂/Si substrates. Each layer was dried on a hot plate at 140 °C–150 °C and subsequently at 210 °C–235 °C for 5 min. In order to prevent cracking and peeling, the films were intermittently annealed in a pre-heated tube furnace in oxygen flow or in air at 650 °C for 10 min after each third layer deposition. Films consisting of 6, 9, 12 and 15 coatings were deposited. The samples were finally annealed at 650 °C for 1 h in oxygen or air.

The microstructures were observed using AFM (atomic force microscope, non-contact mode). Additionally, a one-coating specimen processed under the same conditions as above was prepared for XPS analysis. XPS characterization was conducted on Omicron Full Lab System. Ar ion sputtering (3 keV) served for depth profiling of the elemental composition. In order to suppress the radiation from the sample near edge, the peripheral part of the surface area was covered with a tantalum foil. The sputter rate was obtained from profilometric measurements of the Ar ion etched step. Systematic shifts of the XPS spectra (due to surface charging effect) were corrected using the 4f lines of Pt.

For electrical characterization, Pt and Au were sputtered on the films considered for electrical measurements (6, 9, 12 and 15 coatings) through a shadow mask one after another to get round electrodes of 0.6 mm diameter. Post top electrode deposition annealing was made at 400–450 °C for 15 min in air. The frequency-dependent conductivity and dielectric properties of the films were measured at room temperature at alternating driving signal amplitude of 25 mV using a computer-controlled Agilent 4192A impedance analyzer. Time–voltage- and temperature-dependent leakage currents were measured using a high-precision electrometer (Keithley 6517A). Current density–voltage (J – V) characteristics were acquired in the step mode with a delay time from 30 s to 200 s. The signal was applied to the bottom electrode in all cases. The temperature was measured with a thermocouple placed on the sample surface. Heating was performed using computer controlled heating stage which allows temperature control with an accuracy of 0.2 K.

3. Results and discussions

3.1. Microstructure and phase content

XRD (*X-ray diffraction*) and AFM analysis revealed that the films crystallized into the perovskite phase with (100) preferentially oriented grains (pseudocubic). The ellipsometric film thickness was 100 nm, 175 nm, 270 nm and 365 nm for the specimens consisting of 6, 9, 12 and 15 coatings, respectively. Microstructural aspects of chemical-solution-derived BiFeO_3 thin films annealed in different atmospheres are discussed in more details in our earlier paper [13]. In the present paper an AFM micrograph of the 365 nm thick film specimen is presented exemplary (Fig. 1). Uniform, dense and crack-free surface morphology is observed for all specimens investigated. Very similar morphologies were observed for the thinner films, though they were found to be smoother with a finer grain size.

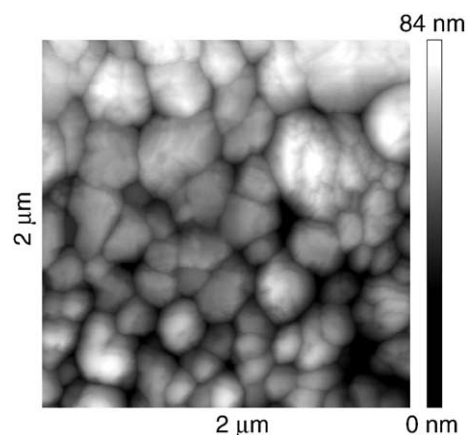


Fig. 1. AFM micrograph of BiFeO_3 thin film consisting of 15 coatings and annealed in flowing oxygen.

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