Materials Letters 228 (2018) 13–16

Contents lists available at ScienceDirect

Materials Letters

journal homepage: www.elsevier.com/locate/mlblue

Featured Letter

Resistive switching and ferroelectric properties in $BiFeO_3$ superlattice films

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ARTICLE INFO

Article history:
Received 14 April 2018
Received in revised form 16 May 2018
Accepted 28 May 2018
Available online 29 May 2018

Keywords:
Superlattice
Thin films
Interface
Ferroelectric properties
Resistance switching behaviors

ABSTRACT

$Bi_{0.89}Ho_{0.08}Sr_{0.03}Fe_{0.97-a}Mn_{0.03}Ni_aO_3/Bi_{0.89}Ho_{0.08}Sr_{0.03}Fe_{0.97-b}Mn_{0.03}Ni_bO_3$ (Ni_a/Ni_b) superlattice films were prepared via a CSD method. The influences of interface effects on ferroelectric properties and resistance switching behaviors of the superlattice films were investigated. There was a weak interface effect in the superlattice films, which was caused by the misfit dislocation generated at the interfaces between Ni_a and Ni_b. The results indicate that the superlattice films show superior ferroelectric properties and good resistance switching behaviors, which is caused by the interface effect. Those superlattice films may have potential applications in multifunctional devices due to their excellent electric properties.

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1. Introduction

In recent years, multiferroic $BiFeO_3$ (BFO) has gained considerable attention, due to its underlying fascinating science and promising industrial applications, such as ferroelectric random access memories and resistive random access memories [1,2]. Compared with the single-phase BFO, ion doping is an effective method to improve the electrical properties of thin films, which makes it useful for the preparation of multifunctional devices [3,4]. However, doping can lead to the increase of the defects which may cause the pinning effect, and the doped thin films show obvious resistance switching (RS) behaviors with the increase of defect density and leakage current [5,6]. To further improve the electrical properties of thin films, the interface engineering is also a common method [7,8]. According to previous studies [9], $Bi_{0.89}Ho_{0.08}Sr_{0.03}Fe_{0.97-x}Mn_{0.03}Ni_xO_3$ (Ni_x, $x = 0.01 \sim 0.04$, $a,b \in x$, $a \neq b$) thin films have the similar crystal structure with different space groups, which leads to the lattice strain and the interface effect in Ni_a/Ni_b superlattice films. Therefore, the enhanced electrical properties are expected from the multi-element co-doping BFO superlattice films.

In this paper, the multi-element co-doping BFO superlattice films were prepared using the chemical solution deposition (CSD) method. The influences of the interface effect on ferroelectric properties and resistance switching behaviors of the Ni_a/Ni_b superlattice films were investigated.

2. Experimental procedure

Ni_a/Ni_b superlattice films were prepared on the FTO (fluorine doped tin oxide)/glass substrates by the CSD method. The appropriate proportions of $Bi(NO_3)_3{\cdot}5H_2O$, $Ho(NO_3)_3{\cdot}6H_2O$, $Sr(NO_3)_2$, $Fe(NO_3)_3{\cdot}9H_2O$, $C_4H_6MnO_4{\cdot}4H_2O$ and $C_4H_6NiO_4{\cdot}4H_2O$ were dissolved in the mixed solution of acetic anhydride and 2-methoxyethanol. Than the solutions were stirred continuously for 2 h at room temperature, stable precursor solutions of Ni_x were obtained. The Ni_a precursor solution was initially spin-coated on FTO glass and annealed at 550 °C. Then, the Ni_b precursor solution was spin-coated on the Ni_a layer and subsequently annealed at 550 °C. These procedures were repeated for 14 times to obtain the desired thickness of Ni_a/Ni_b superlattice films. The Ni_x films were prepared by the same processes.

The structures of the films were studied by X-ray diffraction (XRD, Rigaku, D/MAX-2200). The leakage current densities and the hysteresis loops of the BFO films were tested by an Agilent B2901A instrument and a Radiant Multiferroic system. Capacitance–voltage (*C–V*) measurements were carried out with an Agilent E4980A Concise LCR meter.

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https://doi.org/10.1016/j.matlet.2018.05.125
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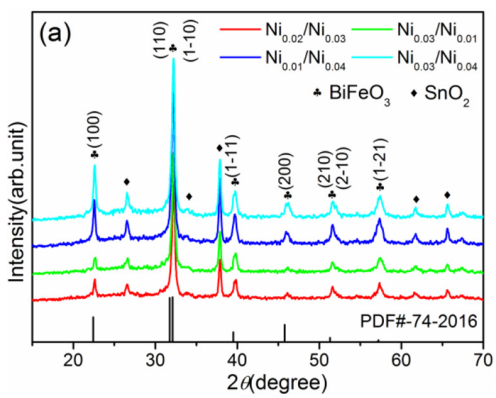
3. Results and discussion

Fig. 1(a) shows the XRD patterns of Ni_a/Ni_b superlattice films. The XRD diffraction peaks of all the films are in conformity with the Standard Card of PDF (JCPDS No. 74-2016). All the films have (1 1 0) preferred orientation. Fig. 1(b) shows the Rietveld-refined space group structural and lattice parameters of Ni_x films [9]. The Ni_a/Ni_b superlattice film is synthesized by two components, Ni_a and Ni_b, which are similar in structures as shown in Fig. 1(b). The lattice mismatchs (f) of the $Ni_{0.02}/Ni_{0.03}$, $Ni_{0.03}/Ni_{0.01}$, $Ni_{0.01}/Ni_{0.04}$ and $Ni_{0.03}/Ni_{0.04}$ superlattice films are −0.46%, 0.20%, 0.38% and 0.58%, respectively according to Eq. (1) [10]. This indicates that there is a tensile stress at the vertical interfaces in $Ni_{0.02}/Ni_{0.03}$ superlattice film, and in other Ni_a/Ni_b superlattice films there is a compressive stress. Therefore, the misfit dislocation is generated at the interfaces, resulting in strain relaxation in the superlattice films. While there is a strain in the interior of Ni_x films, but without a misfit dislocation ($f = 0$), which cannot relax the strain.

$$f = \frac{2(a_{Ni_a} - a_{Ni_b})}{a_{Ni_a} + a_{Ni_b}} \tag{1}$$

where f is a lattice mismatch, a_{Nia} and a_{Nib} are the lattice parameters of R3c:H space group in Ni_a and Ni_b films.

Fig. 2 shows the ferroelectric properties of Ni_a/Ni_b superlattice films. In each diagram, the ferroelectric hysteresis loops (P–E) and instantaneous current curves (I–E) of $Ni_{0.01}$, $Ni_{0.02}$, $Ni_{0.03}$, and $Ni_{0.04}$ films are represented by light color lines as control groups. In an applied electric field of 700 kV/cm, the remnant polarizations P_r ($-P_r$) of Ni_a/Ni_b superlattice films are 122 (−1 1 8), 100 (−8 4), 70 (−5 7) and 82 (−7 2) μC/cm^2, respectively. The coercive fields E_c ($-E_c$) of Ni_a/Ni_b superlattice films are 294 (−2 4 8), 288 (−2 4 5), 321 (−2 5 4) and 314 (−2 4 9) kV/cm, respectively. The switching currents I_S ($-I_S$) of Ni_a/Ni_b superlattice films are 1.0 (−1.0), 0.8 (−0.8), 0.4 (−0.5) and 0.6 (−0.7) mA, respectively. The ferroelectric leakage currents (I_L) of Ni_a/Ni_b superlattice films are less than 0.25 mA. Compared with Ni_x films with obvious pinning effects caused



(b)

Samples	Space group	Lattice parameters (Å)
$Ni_{0.01}$	R3c:H(30.9%)	a = 5.6000
		c = 13.6071
	R3m:R(69.1%)	a = 3.9574
$Ni_{0.02}$	R3c:H(48.5%)	a = 5.5852
		c = 13.6694
	R3m:R(51.5%)	a = 3.9522
$Ni_{0.03}$	R3c:H(65.0%)	a = 5.6112
		c = 14.2709
	R3m:R(35.0%)	a = 3.9550
$Ni_{0.04}$	R3c:H(62.5%)	a = 5.5790
		c = 13.6422
	R3m:R(37.5%)	a = 3.9539

Fig. 1. (a) XRD patterns of Ni_a/Ni_b superlattice films; (b) Details of the Rietveld-refined structural parameters of Ni_x films.

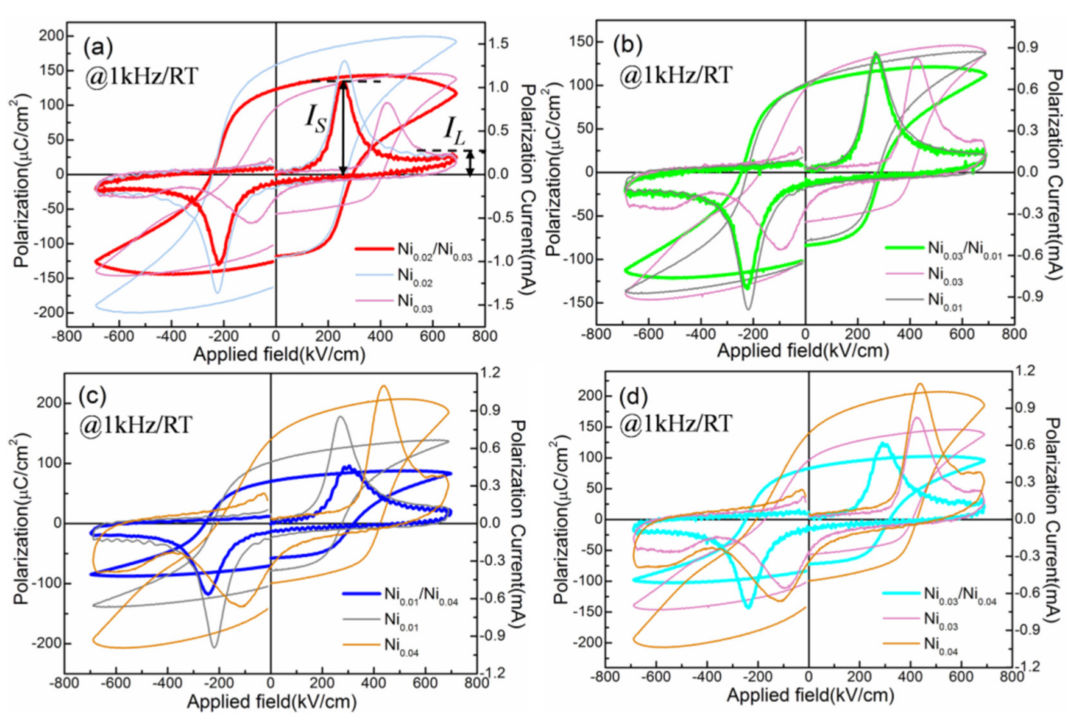


Fig. 2. Ferroelectric hysteresis loops and polarization current curves of (a) $Ni_{0.02}/Ni_{0.03}$, $Ni_{0.02}$ and $Ni_{0.03}$; (b) $Ni_{0.03}/Ni_{0.01}$, $Ni_{0.03}$ and $Ni_{0.01}$; (c) $Ni_{0.01}/Ni_{0.04}$, $Ni_{0.01}$ and $Ni_{0.04}$; (d) $Ni_{0.03}/Ni_{0.04}$, $Ni_{0.03}$ and $Ni_{0.04}$ films.

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