



# Structural, electric transport response and electro-strain - Polarization effect in La and Ni modified bismuth ferrite nanostructures

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## ARTICLE INFO

### Article history:

Received 18 November 2017  
Received in revised form  
8 March 2018  
Accepted 16 March 2018  
Available online 19 March 2018

### Keywords:

Nanostructures  
BFO  
La and Ni doping  
Dielectric constant  
Polarization  
Large strain

## ABSTRACT

In this work, we reported the structural properties, electrical transport response and electro-strain polarization effect of La-Ni bismuth ferrite nanostructures,  $\text{Bi}_{1-x}\text{La}_x\text{Fe}_{1-y}\text{Ni}_y\text{O}_3$  ( $x = 0.0, 0.1$ ;  $y = 0.0, 0.03, 0.05, 0.07$ ) prepared via sol gel technique. X-ray diffraction (XRD) analysis confirmed the rhombohedral crystal structure with space group  $R3c$  of  $\text{BiFeO}_3$ . The Ni ion doping at a Fe - site in  $\text{BiFeO}_3$  escorts to some structural changes and the average crystallite size was found to be in range  $\sim 13$ – $20$  nm. The higher tensile strain was observed in W-H Plot for the prepared samples. The morphological and chemical composition studies of as prepared samples were carried out via using SEM and EDS. Dielectric constant ( $\epsilon$ ) and AC conductivity ( $\sigma_{AC}$ ) were deliberated as a function of frequency in the range 10 Hz to 5 MHz. A significant reduction in the dielectric constant has been observed with increasing Ni doping. An enhancement in the remanent polarization ( $2P_r$ ) value (up to  $1.04 \mu\text{C}/\text{cm}^2$  with applied field 8 kV/cm) and AC conductivity has been observed with La/Ni co-doping. The high Curie temperature ( $T_C$ ) of BFO makes it a suitable candidate for high temperature electromechanical applications. Moreover, an enhanced value of the electric field driven strain as a function of frequency and the applied field is observed which exhibits highly asymmetric S-E hysteresis loops. The peak to peak strain value of  $S_p$  and  $S_N$  is achieved up to 1.019% and 1.582%, respectively, which is the largest value of strain ever obtained in  $\text{BiFeO}_3$  based nano ferrites. The highest value of peak to peak strains ( $S_p$  &  $S_N$ ) and strain memory ( $S_{me}$ ) has been observed for  $\text{Bi}_{0.9}\text{La}_{0.1}\text{Fe}_{0.93}\text{Ni}_{0.07}\text{O}_3$  sample at 8 kV/cm which makes it a suitable candidate for high temperature electromechanical applications (like sensors and actuators etc) in addition to electronic and magnetic applications.

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## 1. Introduction

Over the past decade, the materials which acquire coupled magneto-electric ordering have notable potential applications in multifunctional devices,  $\text{BiFeO}_3$  (BFO) is one of them and this M-E coupling is principally associated with domain switching [1–3]. The perovskite bismuth ferrite (BFO) material has attracted huge interest in scientific research community due their multiferric properties and prospective applications in different fields of science and technology. BFO has a rhombohedrally distorted perovskite structure (space group  $R3c$ ) with outstanding high Curie temperature ( $T_C \approx 1100$  K) and anti-ferromagnetic Neel temperature

( $T_N \approx 675$  K) with a spatially modulated spiral spin structure [4]. In BFO, ferroelectric and magnetic properties arise from distinct sources and occurs independently from each other. In addition, the ferro-electricity arises due to the displacement of  $\text{Bi}^{3+}$  and  $\text{Fe}^{3+}$  ions with respect to [111] direction in the rhombohedral unit cell, while the unpaired electrons in the d shell of  $\text{Fe}^{3+}$  ions originate magnetic exchange interaction through  $\text{O}^{2-}$  which gives G-type anti-ferromagnetic ordering with the existence of very weak ferromagnetism due to Dzyaloshinskii-Moriya [D-M] interaction [5,6]. Such unique characteristics of BFO make it suitable aspirants for room-temperature multiferric materials, which are elegant for applications in magnetic recording media, solar cell, light-emitting diodes, multifunctional devices, spintronics and magnetic memory device [2,7]. The switching mechanism in ferroelastic domains of BFO nano ferrites escorts the orientation of magnetic planes, which contributed to strain [8]. The research on the electric field driven

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strain in BFO nano ferrite is critical to understand the involved mechanism of the switching process in ferroelastic domains. Moreover, in recent, BFO has also been demonstrated as a promising material for the application in actuators, sensors and micro-positioner on account of its appropriate band gap (~2.2 eV), excellent chemical constancy under high temperature condition and intrinsic electric polarization field [9]. The spiral spin period order of 62 nm prevents the net magnetization, thus the Magnetoelectric [M- E] coupling in BFO is improved by reducing the particle size lower than the spiral spin structure of periodicity (62 nm) [1,7,10]. It is extremely complicated to obtain the pure phase of BFO due to a narrow temperature range of phase stabilization, availability of impurities like  $\text{Bi}_2\text{Fe}_4\text{O}_9$ ,  $\text{Bi}_2\text{O}_3$  and  $\text{Bi}_{25}\text{FeO}_{40}$  was dried in oven at 110 °C and lead to the increase in leakage current thus it becomes unacceptable for device application [11,12]. The earlier reports revealed that the electric field driven strain and peak to peak strain value in undoped BFO nano ferrites has comparatively low and less saturated, then some the lead free piezoelectric materials (ex.  $(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3$ ) [8,9,13,14]. The chemical substitution of rare earth metals on A site and transition metals at B site or both, in BFO will enhance the multiferroic properties by reducing the impurity phases and enhance the M-E coupling because of a mismatch in ionic radii between dopant and parent ion [6,15–19]. In addition, the aliovalent doping on BFO sites also making the electric field driven spin disorder to order transition possible at multiferroic triple phase point and electric field driven strain would be typically modified [8,20,21]. The creation of oxygen vacancies by relevant doping and their interface with these dopants elements could have a markable effect of electric field driven strain essence of ferroelectric materials [14]. It is well known that modification on A and B site in BFO nano ferrites by doping reveals high leakage current along with a low breakdown field, then investigation on electric field driven strain behavior is almost scanty [4,8]. Thus, in recent years, there has been mounting curiosity in multiferroic materials  $\text{BiFeO}_3$  and the defective BFO play an important role in low as well as high temperature electro-mechanical applications. This work investigates the doping effect of La and Ni on A and B site of BFO respectively. The selection of La and Ni ions were due to they have very close ionic radii to Bi and Fe ions, which are synthesized via sol-gel technique. This defect engineering improves the ferroelectric, electrical and electric field driven strain of BFO.

Therefore, in the current work, by keeping in view of the above, in order to precisely understand the doping effect of La and Ni on A and B site of BFO, we present an efficient effort to investigate the structural, electrical, ferro-electric and electric field driven strain hysteresis of  $\text{Bi}_{1-x}\text{La}_x\text{Fe}_{1-y}\text{Ni}_y\text{O}_3$  ( $X = 0.0, 0.1$ ;  $Y = 0.0, 0.03, 0.05, 0.07$ ) nano ferrite, which is not reported earlier in the literature.

## 2. Experimental details

### 2.1. Chemicals

For the synthesis of  $\text{Bi}_{1-x}\text{La}_x\text{Fe}_{1-y}\text{Ni}_y\text{O}_3$  ( $X = 0.0, 0.1$ ;  $Y = 0.0, 0.03, 0.05, 0.07$ ) nano ferrite, the precursor materials were used as  $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ ,  $\text{La}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ ,  $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$  and  $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ , Nitric acid,  $\text{NH}_3$  solution and de-ionized  $\text{H}_2\text{O}$  (Loba India). All the chemicals utilized in the current work for the synthesis of BFO based nano ferrite, are of analytical grade and used without supplementary purifications.

### 2.2. Synthesis and characterization

Pure BFO and La-Ni doped  $\text{Bi}_{0.9}\text{La}_{0.1}\text{Fe}_{1-y}\text{Ni}_y\text{O}_3$  (where  $Y = 0.03, 0.05, 0.07$ ) have been synthesized via sol-gel technique. All the precursors were taken in stoichiometric amount and dissolved in

deionized  $\text{H}_2\text{O}$ . 4 mmol of nitric acid ( $\text{HNO}_3$ ) was added to the bismuth nitrate solution to make the solution transparent and the resulting solution was put on the hot plate at 70 °C under continuous magnetic stirring. The ammonia ( $\text{NH}_3$ ) solution used as a precipitating agent, which forms the brownish precipitate. Finally the resulting precipitate was washed several times with deionized  $\text{H}_2\text{O}$  and ethanol. The precipitate was dried in an oven at 110 °C for 12 h and sintered at 650 °C for 4 h to obtain the well crystalline structure of nanoparticles. The sintered powder was grounded well and pressed into pellets of 10 mm in diameter by adding 10% PVA as a binder. These pressed pellets were sintered at 700 °C for 10 min with a heating rate 10 °C per min to remove the PVA as well as other organic compounds, so that these organic compounds do not affect the results and all results measure precisely by using further sintered pellets.

The crystal structure of the synthesized samples was confirmed by X-ray diffraction measurement (Rigaku diffractometer) with Cu ( $K_\alpha$ ) radiation source of wavelength 1.54 Å. The experiment performs in the range between 10° to 80° with scanning step of 2° per minute. The surface morphology and energy dispersive spectra of all the compositions of BFO were studied by scanning electron microscope with Energy Dispersive Spectrometer (EDS). The pellets of all the samples were polished with silver (Ag) paste to perform the electrical and ferroelectric studies by using Potentiostat/Galvanostat (Model No. SP240; Bio Logic) and Automatic PE loop tracer (MARINE India) respectively.

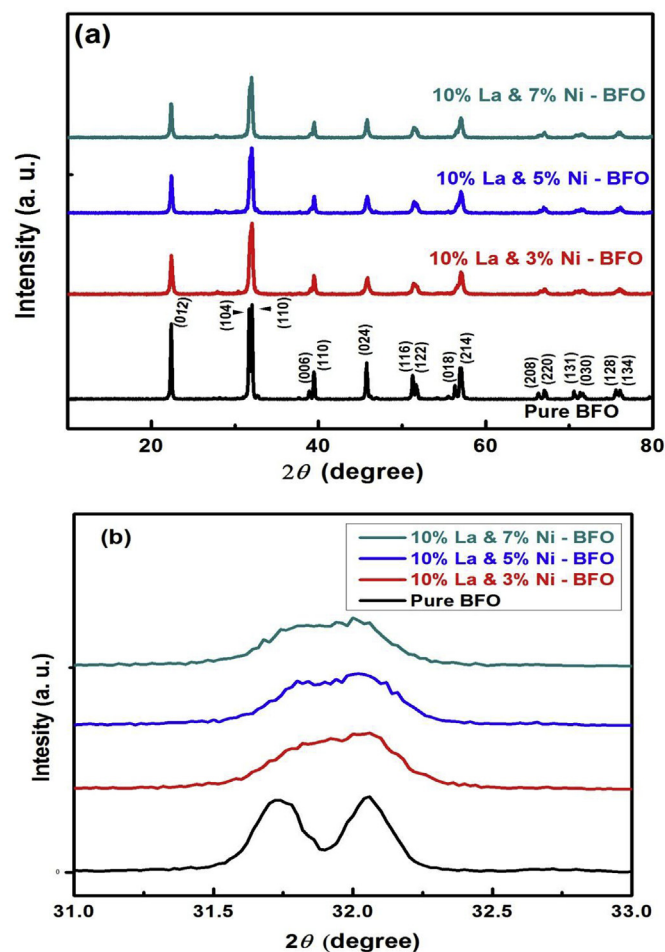


Fig. 1. (a) XRD patterns of  $\text{Bi}_{1-x}\text{La}_x\text{Fe}_{1-y}\text{Ni}_y\text{O}_3$  ( $X = 0.0, 0.1$ ;  $Y = 0.0, 0.03, 0.05, 0.07$ ) nano ferrite samples at room temperature. (b) Magnified view of XRD patterns around 32° of all four BFO nano ferrite samples.

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