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In-situ X-ray diffraction studies and magneto-optic Kerr effect on RF sputtered thin films of $BaTiO_3$ and Co, Nb co-doped $BaTiO_3$

R. Siddheswaran[a,*], Pavol Šutta[a], Petr Novák[a], Marie Netrvalová[a], Aleš Hendrych[b], Ondřej Životský[b,c]

[a]New Technologies Research Centre, University of West Bohemia, Plzeň 30614, Czech Republic
[b]Institute of Physics, VŠB-Technical University of Ostrava, 17. Listopadu 15, Poruba, 708 33 Ostrava, Czech Republic
[c]RMTVC, VŠB-Technical University of Ostrava, 17. Listopadu 15, Poruba, 708 33 Ostrava, Czech Republic

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Abstract

The effect of annealing on structural and magnetic properties of the RF sputtered $BaTiO_3$ and Co, Nb co-doped $BaTiO_3$ thin films on Si (001) substrates were studied. The structure of the as-deposited (amorphous) films was changed into cubic perovskite phase during the in-situ X-ray diffraction from room temperature to 900 °C. The enhancement of crystalline quality with respect to the increase of annealing temperature was observed by the in-situ XRD. The magnetic properties of the films before and after annealing were studied by the measurement of Magneto-Optic Kerr Effect (MOKE). The pure $BaTiO_3$ revealed a paramagnetic behavior, whereas the Co and Nb co-doped $BaTiO_3$ films exhibited room temperature ferromagnetism. The increase in ferromagnetic response was observed in the Co and Nb co-doped $BaTiO_3$ films annealed at 900 °C rather than the as-deposited film.
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Keywords: $BaTiO_3$; Thin film; RF sputtering; In-situ XRD; Magnetic properties

1. Introduction

Multiferroics are considered as one of the prominent classes of multifunctional materials possess two or more ferroic properties which generally refer to the coexistence of ferroelectric and ferromagnetic properties [1]. During the past two decades, the multiferroic materials have gained a tremendous potential to revolutionize the future technologies of data storage, multiple state memory element, sensors, actuators, resonators and capacitors [2]. Moreover, the multiferroic materials have received much attention due to their potential applications based on the magneto-electric coupling effect [3]. Barium titanate ($BaTiO_3$) is a ferroelectric and highly insulating material with perovskite structure has increased much interest due to its many potential applications such as, high dielectric constant capacitors, dynamic random access memories, piezoelectric generator and optical waveguide devices [4]. As well, it has high dielectric constant, low dielectric loss, large electro-optic coefficients, chemical stability and low toxicity [5]. The addition of dopants like rare earth ions, transition metal ions or other impurities makes the $BaTiO_3$ as a promising multifunctional material for various applications [6]. The ferromagnetic property in a ferroelectric material can be usually obtained by the incorporation of transition metal impurities such as Fe, Ni, Co, etc. into the ferroelectric host material [7,8]. Furthermore, the $BaTiO_3$ can be changed into semiconductor material by some kind of doping which alters the structure, grain and grain boundaries in order to use it for optoelectronic applications [9]. Among the doping in $BaTiO_3$, the incorporation of niobium promotes the dielectric properties [10]. The doping of niobium in $BaTiO_3$ is used to make a multilayer ceramic capacitor (MLCC) [11]. In addition to this, the incorporation of an appropriate amount of Co makes the $BaTiO_3$ as a ferromagnetic material at room temperature.

*Corresponding author. Tel.: +420 37763 4771; fax: +420 37763 4702
E-mail addresses: rsiddhes@yahoo.com, rajendra@ntc.zcu.cz (R. Siddheswaran).

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Hence, the combined nature of ferroelectricity and ferromagnetism in $BaTiO_3$ develops a multiferroic behavior. The present work describes the RF sputtering deposition of pure $BaTiO_3$ and Co and Nb co-doped $BaTiO_3$ thin films, studies on structural phase changes during the in-situ X-ray diffraction and analysis of magnetic properties of the films by magneto-optic Kerr measurements at room temperature.

2. Experimental

2.1. RF sputtering deposition

Thin films of pure $BaTiO_3$ and Co, Nb co-doped $BaTiO_3$ were deposited on silicon (001) substrates by RF sputtering (with RF frequency of 13.56 MHz) using the BOC Edwards TF 600 coating system. The deposition of the films was carried out in argon atmosphere from a $BaTiO_3$ ceramic target (purity 99.99%). The Co and Nb doping was attained by the pieces of respective metal pellets kept over the erosion area of the $BaTiO_3$ target. The doping concentration was controlled by the number of metal pieces. Before the deposition process, the substrates were cleaned ultrasonically in acetone and then etched by RF discharge in the chamber. Afterwards, the deposition chamber was evacuated to a base pressure of 10^{-4} Pa. The depositions were started at the following conditions: the constant discharge power of 400 W, argon pressure of 0.6 Pa, argon flow rate of 5 sccm and substrate temperature of 350 °C. The as-deposited films were amorphous state, and then annealed at 900 °C to acquire crystalline structure. The films thickness range from 600–700 nm was measured by profilometer. The incorporation of Co and Nb content was found to be 6.5 and 3.5 at%, respectively. This elemental analysis was carried out by energy dispersive X-ray spectroscopy (EDS) using SEM (JEOL JSM-7600F) equipped with EDS detector (X-Max, Oxford Instruments).

2.2. X-ray diffraction analysis

The structural characterizations (phase analysis, crystallite size, crystallographic orientation and micro-strains) of the films were investigated by in-situ X-ray diffraction (XRD) analysis. The XRD patterns were carried out by using an X-ray diffractometer X′Pert Pro equipped with an ultra fast linear semiconductor detector PIXcel and a point detector. The CuK_α radiation ($\lambda = 0.154$ nm) was used as an X-rays source. The XRD patterns were recorded from 15 to 60° angles.

For this experiment, the position, height, integrated intensity (area) and full widths at half maximum (FWHM) are the main four parameters that characterize the diffraction lines. In this experiment, two different goniometer arrangements were used: the first one is characterized by symmetric (Bragg–Brentano) ϑ –ϑ geometry (Fig. 1), where the X-rays angle of incidence to the sample surface is equal to the angle of reflectance. It means that the lattice planes (*hkl*) where diffraction takes place are parallel to the sample surface. This condition was used during the whole measurement. The second one is characterized by an asymmetric ω–2ϑ (Seemann–Bohlin) geometry (Fig. 2).

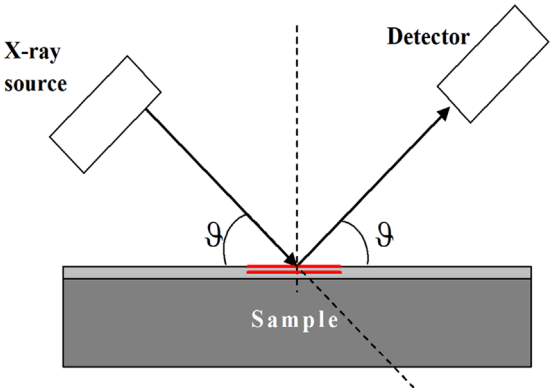

Fig. 1. X-rays reflection by Bragg–Brentano (ϑ –ϑ) geometry.

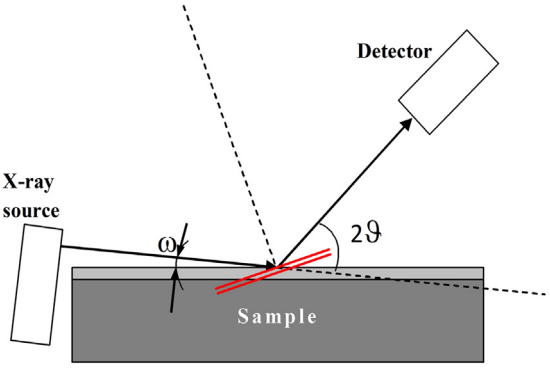

Fig. 2. X-rays reflection by Seemann–Bohlin (ω–2ϑ) geometry.

In ω–2ϑ geometry, the angle (ω) of X-ray incidence to the sample surface is small and fixed (usually much less than 10°), whereas the detector moves in 2ϑ scales. In this case, the lattice planes are inclined from the sample surface about the angle ϑ_{hkl}–ω. The advantage of this geometry in comparison with the ϑ–ϑ geometry is that the X-ray beam do not penetrate abundant into the sample (if the investigated film is sufficient in thickness, we do not observe diffraction lines from the substrate material) and furthermore, the irradiated area of the sample surface is constant during the measurement due to the fixed angle of incidence.

In our experiment, the asymmetric geometry was used for the analysis of as-deposited films in the amorphous state, whereas the symmetric geometry was used for the in-situ experiment. The annealing was performed in a high temperature chamber (HTK 1200 AP) with vacuum pressure of 10^{-3} Pa. According to our previous experiences with similar materials, we decided to apply the heating temperature only up to 900 °C. The heating procedure contains several steps in the record of XRD patterns: (1) Recorded at 25 °C before heating (at room temperature), (2) at the second step, it was recorded at 250 °C, (3) the third step was at 700 °C, (4) then, recorded at 900 °C and finally, (5) recorded at 25 °C after cooling down. Usually, the second step (at 250 °C) is considered to be a procedure in order to clean the chamber from the residual gases. The diffraction patterns recorded during the in-situ experiment

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