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Determination of elastic constants of transition metal oxide based thin films using surface brillouin scattering

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Abstract

Bismuth ferrite ($BiFeO_3$) is a transition metal oxide that exhibits both antiferromagnetic and ferroelectric orderings and is termed a magnetoelectric multiferroic. These functional properties make it crucial for applications in various nanoelectronic devices and sensors. However, the integration of $BiFeO_3$ in devices requires the scaling down of bulk $BiFeO_3$ to nano dimensional length scales in thin film format. For this purpose, the elements of the elastic constant tensor of $BiFeO_3$ thin films are requisite, especially in multilayered or single layer-on-substrate device configurations. It is thus essential that mechanical properties of $BiFeO_3$ thin films be established due to their size and growth mode dependence.

Therefore, the study aims to determine the propagation of the surface acoustic waves and the elastic constants of $BiFeO_3$ (BFO) thin films in order to tailor the mechanical properties for device applications. In this approach the effect of morphology and microstructure on the elastic constants has been investigated.

To this end, BFO thin films have been grown following the predictions of the Structural Zone Model (SZM) using unbalanced RF Magnetron Sputtering. The RF sputter power (ad-atom energy) 30 -75 W, the negative substrate bias voltage $0-100$ V and the substrate temperature (ad-atom mobility) 100-600° C have been varied to establish the role of ad-atom energy, induced stress and ad-atom mobility on the film properties.

Atomic force microscope (AFM), scanning electron microscopy (FESEM), X-ray reflectivity (XRR), Rutherford backscattering (RBS), grazing incidence X-ray diffraction (GIXRD) and surface Brillouin scattering (SBS) techniques have been used for thin film characterization. The combined use of these techniques provide a powerful tool to correlate the microstructure and morphology of BFO thin films. Furthermore, the mechanisms of stress evolution in BFO thin films through Ar^+ implantation is demonstrated using an atomistic understanding.

Atomic force microscope (AFM) and field emission scanning electron microscopy (FESEM) studies on the surface topography (roughness) and the cross-sectional view of the microstructure of the films' growth have shown a surface roughness in the range 0.17- 4.17 nm and a compact granular structure with columnar growth mode. These growth modes identified by FESEM have been found to be compatible to the zone 1 and zone 2 of the SZM for the deposition conditions of interest. X-ray reflectivity (XRR) provided a measure of the mass density ($7.71 - 8.25 \pm 0.05$ gcm^{-3}) and deposition rate in the range $4.1 - 11.1$ nms^{-1} for all the films. Rutherford backscattering (RBS) measurements of the film grown at the deposition conditions of interest yielded non-stoichiometric composition. In addition, measurements

of these films under a negative substrate bias have confirmed the incorporation of Ar in the films. The amount of Ar was evaluated to be $0.1 - 5.5$ at. %. A study of the crystalline structure of the BFO thin films by grazing incidence X-ray diffraction (GIXRD) have shown amorphous and nanocrystalline phases in the films. The formation of these phases is strongly dependent on the ad-atom energy and surface diffusivity, which are determined by the sputter power and the substrate temperature, respectively.

For the surface Brillouin scattering measurements, a backscattering geometry was used to illuminate and collect the inelastically scattered light. The spectra were excited by the 514.5 nm line of an Innova Coherent Ar^+ laser operated in a single axial mode. The backscattered light was guided by a series of optics to be analysed by a Sandercock $(3 + 3)$ pass tandem Fabry–Pérot interferometer. The measured inelastically scattered light is frequency shifted by the vibrational modes propagating in the film-substrate system. These modes have been identified as the true surface waves (SAWs) comprising of the Rayleigh surface acoustic wave and the film guided modes or Sezawa waves (SW). This observation presents the scenario of a slow on fast system. The phase velocities of the SAWs that propagate in every direction of the film-substrate system are obtained from the measured frequency shift as a function of $k_{\parallel}h$ (where $k_{\parallel}$ is a probing wave vector parallel to the surface of the film and h is the film thickness), which provide the experimental dispersion curves. Using the method of least squares minimization the elastic constants have been extracted through the simulation of the experimental dispersion curves. Fitting the experimental with the calculated dispersion curves using elastic constants of the BFO films as free parameters yielded the effective elastic constants of the films. Theoretical modelling based on the surface Green's functions was used to simulate and compare Brillouin spectra with the experimental spectra for selected thin films. The elastic constants obtained seem to have some relationships with the grain size that can be correlated with the stress developed in the films. The Ar^+incorporation has confirmed stress evolution in BFO thin films due to changes in the elastic constants based on the film growth conditions. The results have shown a decrease in elastic constants and corresponding Bulk modulus as a signature of compressive stress. Moreover, we report an increase in the shear modulus of the biased BFO films by a factor greater than 2, $(21\ GPa < (G = C_{44}) < 49\ GPa)$. This increase is attributed to the localized deformation in the film due to Ar^+ incorporation. The incorporation of Ar^+ also decreases the bulk modulus by 57 %.