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Enhanced tunability of transparent epitaxial $Ba_{0.5}Sr_{0.5}TiO_3/Ga_2O_3/GaN$ structures fabricated by pulsed laser deposition

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ABSTRACT

We fabricated all-epitaxial $Ba_{0.5}Sr_{0.5}TiO_3$ (BST) (111)/β-Ga_2O_3 (−201) thin films on GaN (002)/Al_2O_3 (001) substrates by pulsed laser deposition. BST films with different textures were obtained by depositing β-Ga_2O_3 buffer layers on GaN substrates under varied deposition conditions. The results for dielectric constant, tunability, and dielectric loss of the BST/Ga_2O_3/GaN and BST/GaN layers at an applied voltage of 20 V, were 225 and 199, 20% and 14%, and 0.006 and 0.018, respectively. The epitaxial growth of the BST film and enhancement of the dielectric properties of the BST/Ga_2O_3/GaN heterostructure were achieved in the presence of an epitaxial Ga_2O_3 buffer layer.

1. Introduction

Ferroelectric thin films have been widely investigated from the perspective of their application in integrated dynamic random access memory cells, decoupling capacitors, and microwave tunable devices such as electrically tunable filters, voltage-controlled oscillators, and phase shifters [1–3]. One of the most promising ferroelectric materials for use in tunable microwave devices due to advantages such as high dielectric constant, large optical refractivity, good dielectric tunability, and low dielectric loss is $(Ba_{1-x}Sr_x)TiO_3$ (BST). Previous studies have been carried out on the fabrication of polycrystalline BST films with low strain and preferential orientation in the (111) direction, as well as attempts to optimize the composition, thickness, and grain size of these films [4,5]. It was reported that BST films grown on MgO and $LaAlO_3$ single-crystal substrates had good crystallinity, small lattice mismatch, and high dielectric constant and tunability. However, the methods used for mounting components when fabricating hybrid microwave integrated circuits are undesirably complex when using these substrates [6]. Furthermore, BST films grown directly on semiconductors have low tunability due to the formation of undesired interfacial layers between the ferroelectric film and semiconductor. Unfortunately, most ferroelectric materials become thermodynamically unstable when they come in direct contact with silicon. Therefore, a suitable semiconductor substrate and a buffer layer with adequate chemical and thermal endurance are essential for realizing a stable interface, as well as for suppressing chemical reaction between the ferroelectric thin film and the semiconductor substrate. In particular, the buffer layer plays an important role for inducing epitaxial growth of the film and minimizing lattice mismatch between the film and substrate [7]. In recent years, GaN-based devices have been widely used in electronic and optoelectronic devices such as blue-light-emitting diodes and laser diodes because GaN possesses excellent intrinsic properties including high-temperature stability, chemical stability, wide band-gap, and good mobility [8]. Several studies have reported on epitaxial and other properties of ferroelectric films on GaN [9–12]. Lee et al. demonstrated that a transparent epitaxial β-Ga_2O_3/GaN heterostructure, applied by pulsed laser deposition (PLD), provided adequate results related to crystal quality, dielectric properties, environmental (structural and chemical) stability, and produced a sharp film–substrate interface [13]. Since Ga_2O_3 is a native oxide of GaN, it is a promising material for consideration in the fabrication of contaminant free oxide/GaN interfaces. Therefore, we used a Ga_2O_3 buffer layer and a GaN semiconductor substrate to fabricate a BST film, and the resulting film had a stable structure and sharp film–substrate interface.

In this paper, we report the epitaxial growth of fully epitaxial BST/Ga_2O_3/GaN heterostructures fabricated by the PLD method. We investigated the structural and electrical properties of the resulting BST films, with and without Ga_2O_3 buffer layers on GaN substrates, with

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emphasis placed on the relationship between the Ga_2O_3 buffer layer and the physical properties of the resultant BST films.

2. Experimental procedures

The GaN layer on an Al_2O_3 substrate acted as a semiconductor. A bottom electrode was prepared by metal-organic chemical vapor deposition (MOCVD). In order to increase the concentration and mobility of the carriers, we artificially doped the GaN layer with Si during the MOCVD process. The thickness of the Si-doped GaN epilayer was 2.5 μm. The $Ba_{0.5}Sr_{0.5}TiO_3$ (BST) thin films and β-Ga_2O_3 buffer layers were deposited on n-GaN/Al_2O_3 substrates by PLD. A KrF laser (248 nm; Lambda Physik, COMPex 205 with raster function) providing beams with energy densities of 3.0 mJ/cm^2 and 1.6 mJ/cm^2 were used for fabrication of the BST thin film and β-Ga_2O_3 layer, respectively. The substrate temperature was fixed at 800 °C. The oxygen flow rates were 5 sccm and 15 sccm for the β-Ga_2O_3 buffer layers and 25 sccm for the BST thin films, respectively. The working pressures used during deposition of the Ga_2O_3 buffer layers and BST films were 0.73 Pa and 26.7 Pa, respectively. The vertical distance between the target and the substrate was fixed at 5 cm. The structures, orientations, and the epitaxial relationships of the resulting BST/β-Ga_2O_3/GaN thin films were characterized by X-ray diffraction (XRD; X'Pert Pro MPD, Phillips) experiments performed using monochromatized Cu–$K_{\alpha 1}$ radiation for the φ-scan. The surface morphology and roughness of the BST thin films were characterized by scanning electron microscopy (SEM; Hitachi S-4200) at 15 kV and atomic force microscopy (AFM; DI, Nanoscope IV) using non-contact mode. A silicon cantilever of force constant ~48 N/m and resistivity ~0.01–0.02 Ω·cm was used for the AFM studies. The optical properties of the films, including the visible and UV spectral regions, were measured by UV–vis spectrometer (Cary 5000, Varian). Resultant effects of the various oxygen flow rates used in application of the β-Ga_2O_3 buffer layer were evaluated for their effect on the dielectric properties of the metal-oxide-semiconductor structure. For this purpose, capacitance–frequency (*C*–f) and capacitance–voltage (*C*–V) measurements were performed using a high-precision impedance analyzer (HP4294A).

3. Results and discussion

3.1. Structural properties of resulting BST thin films

Fig. 1(a) reveals the XRD patterns of BST films grown on GaN and Ga_2O_3/GaN substrates, respectively. The Ga_2O_3 buffer layers deposited at an oxygen flow rate of 5 sccm (henceforth represented as Ga_2O_3-5 sccm), underwent crystallization to form monoclinic β-Ga_2O_3 phases. No secondary phases were formed in this case. In contrast to the polycrystalline BST films grown directly on GaN and Ga_2O_3-15 sccm/GaN substrates, the BST film deposited on Ga_2O_3-5 sccm/GaN demonstrated preferential orientation in the (*hhh*) direction. We hypothesize that the orientation of the BST thin film is dependent on the structural characteristics of the Ga_2O_3 buffer layer. The lattice misfit was calculated as $(d_{film} - d_{sub})/d_{sub}$, where d_{film} and d_{sub} are in-plane lattice parameters of the film and substrate, respectively [14]. For the misfit calculation, in-plane lattices of β-Ga_2O_3 (010), GaN (100), and BST (−110) were chosen and d-values were 0.3039 nm, 0.3186 nm, and 0.2791 nm, respectively [8,11]. The lattice misfits of β-Ga_2O_3 (−201) on GaN (002), BST (111) on β-Ga_2O_3 (−201), and BST (111) on GaN (002) films were approximately −4.61%, −8.16%, and −12.40%, respectively. The strains of the BST films were tensile, not compressive, because in-plane lattice of BST is smaller than that of substrate (Ga_2O_3/GaN or GaN). These results imply that improved structural alignment is more likely to be achievable for BST films on Ga_2O_3/GaN rather than for BST/GaN films.

Fig. 1(b) provides the BST/Ga_2O_3-5 sccm/GaN film φ-scan results for the (110), (111), and (103) reflections corresponding to BST, β-Ga_2O_3, and GaN, respectively. The scans for BST, β-Ga_2O_3, and GaN were obtained at ψ = 35.26° (2θ = 32.042°), ψ = 80.59° (2θ = 35.184°), and ψ = 32.04° (2θ = 63.435°), respectively. The six-fold symmetry peaks found in the φ-scan of the (111) reflection indicate the specific, in-plane epitaxial growth (twinned epitaxy) of the Ga_2O_3 (−201) film on the GaN (002) substrate. By performing XRD analysis, the epitaxy relations were confirmed to be BST (111)‖Ga_2O_3 (−201)‖GaN (002) and BST (−110)‖Ga_2O_3 (010)‖GaN (100).

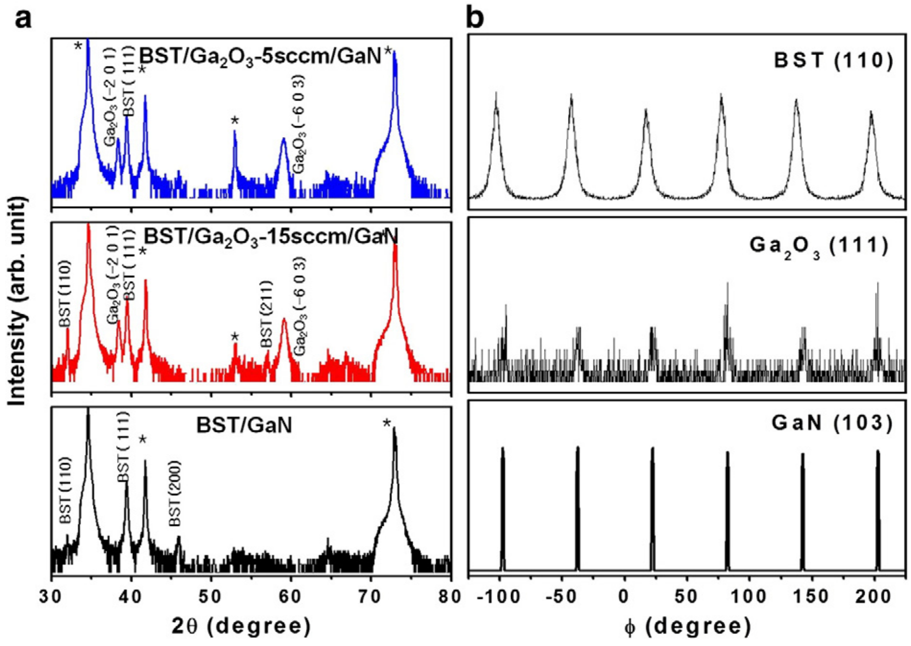

Fig. 1. (a) X-ray diffraction patterns of $Ba_{0.5}Sr_{0.5}TiO_3$ films with and without a Ga_2O_3 layer, and (b) ϕ-scans of $Ba_{0.5}Sr_{0.5}TiO_3$/Ga_2O_3-5 sccm/GaN structures. (*GaN/Al_2O_3 substrate).