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Ultraviolet radiation detection by barium titanate thin films grown by sol–gel hydrothermal method☆

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ABSTRACT

Ultraviolet (UV) photodetector using ferroelectric barium titanate, $BaTiO_3$ thin film has been prepared successfully. $BaTiO_3$ (BTO) this film is deposited by sol–gel hydrothermal (SG-HT) method. The deposited BTO films were found to be polycrystalline having a band gap of about 3.51 eV. The photoconductive gain ($K = I_{on}/I_{off}$) of bare BTO based photodetector was found to be 8.36×10^2 for UV radiation ($\lambda = 365$ nm and intensity = 24 μW/cm^2). The modifier, tungsten (W) in the form of both thin overlayer and uniformly distributed circular dotted structures (600 μm diameter) were integrated with the surface of BTO thin film by rf-magnetron sputtering technique to improve the photoresponse characteristics. The photoconductive gain was enhanced to about two orders of magnitude (1.84×10^4) after integration of W modifier in the form of circular dots. The significant enhancement in photoresponse for W(dots)/BTO photodetector is related to the twin effect of (1) reduction in dark current (I_{off}) due to formation of schottky junction between the oxide (BTO) and metal (W), and (2) enhancement in photocurrent (I_{on}) due to high absorption of UV radiation on the detector surface.

1. Introduction

Ferroelectric materials have potential applications, for high dielectric-constant capacitors, ferroelectric memories, sensor, transducers, etc. Ferroelectric thin films were also important for tunable microwave devices, acoustic and pressure sensors, surface acoustic wave devices, MEMS and actuator applications [1]. Few reports are also available on realization of UV photodetectors using ferroelectric thin films by combining their unique optical and piezoelectric properties [2–9]. In the last two decades, there has been increasing demand for reliable photodetectors for detection of UV radiation due to its harmful effects [10]. The development of UV photodetectors is important because of its potential applications in the various fields of science and technology, including space science, aircraft science, defense and other industries [11]. Lead zirconate titanate (PZT) having very high piezoelectric coefficient has been widely used for the realization of UV photodetectors [2,3]. However, there is urgent need for identification of suitable green materials, which must be lead free, for these applications.

Barium titanate, $BaTiO_3$ is a well known lead free ferroelectric material with high dielectric constant and promising ferroelectric properties which is being exploited in a number of applications like gas sensors, capacitors, memory devices, etc. [12,13]. However, the material properties of $BaTiO_3$ (BTO) need to be tailored for photonic device applications [8] including photo-detectors. Thus, BTO after desired modifications in structural property has also been used for the photovoltaic applications [14]. A number of physical and chemical deposition techniques have been utilized for the growth of BTO thin films, including pulsed laser deposition (PLD), magnetron sputtering, thermal evaporation, chemical vapor deposition, co-precipitation, sol–gel, chemical bath deposition, etc. [13,15–18]. Amongst them, Sol-gel-hydrothermal (SG-HT) technique is a promising chemical method for low cost, low temperature fabrication of BTO thin films with large surface area and hence may be useful for sensing applications [11]. Although, there are few reports on the fabrication of BTO thin film using sol–gel hydrothermal method, but have not been explored for the sensing applications [18,19]. Further it is reported that incorporation of various metals in different forms including dopants, overlayer and dispersal of nanoparticles may be useful in improving the photoconducting properties of compound semiconductors [20,21] and hence results in development of UV photodetector with improved response due to formation of schottky junction [22,23]. However no such study has been made towards the influence of suitable

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metal modifier on the photoresponse of UV photodetector based on multicomponent ferroelectric materials especially BTO thin film. Amongst different metals including Pt, Au, Al, Cu, Sn, W, etc., W has comparatively lower work function (4.55 eV) in comparison to that of BTO (5.6 eV), which may result in the formation of schottky junction. Thus, in the present work, UV photodetector has been prepared using BTO thin film deposited by low-temperature sol–gel hydrothermal method. BTO thin film is also integrated with W modifier in the form of both a thin (20 nm) overlayer and uniformly distributed circular dots (600 μm dia) W(dots)/BTO using rf magnetron sputtering technique. The photoresponse characteristics of both W(cont.)/BTO and W(dots)/BTO photodetectors have been studied towards UV radiations and compared with the bare BTO thin film based photodetectors.

2. Experimental details

The sol–gel hydrothermal (SG-HT) technique consists of two steps: (1) preparation of BTO gel film by a conventional sol–gel process as a seeding layer and (2) hydrothermal treatment of the as-deposited seeding layer of BTO [19]. The starting materials for sol–gel process were barium acetate and titanium(IV)isopropoxide. Glacial acetic acid was used a solvent, whereas 2-methoxyethanol, and ethylene glycol were taken as polymerizing agents. Barium acetate was first dissolved into heated glacial acetic acid followed by addition of titanium isopropoxide under constant stirring. Mixture was added with ethylene glycol to form a final BTO complex solution. Subsequently, the final solution was diluted with equivolume amounts of glacial acetic acid and 2-methoxyethanol. The concentration of solution was about 0.2 M. For the fabrication of UV photodetector, BTO thin films were deposited on the silicon (Si) substrate having platinum (Pt) interdigital electrodes (IDEs) patterned on it. Fig. 1 shows the schematic of UV photodetector using BTO thin film as the functional layer. For the patterning of IDEs, 80 nm thin layer of Pt was deposited on SiO_2 passivated Si substrate by rf sputtering technique using a 3 inch diameter Pt (99.99% pure) target in 100% Ar gas ambient by applying an rf power of 100 W. To improve the adhesion of Pt layer, a buffer layer of titanium (10 nm) was sputtered prior to Pt deposition on the passivated Si substrate in situ under similar deposition condition. Pt IDEs were patterned on Si substrate (IDEs/Si) using the conventional photolithographic technique. The prepared BTO complex solution was spin coated over the IDE/Si substrates as the seeding layer protecting the contact pads of IDEs for electrical connections. After each coating, the film was pyrolyzed in air at 400 °C for 5 min. The thickness of seeding layer of BTO was maintained

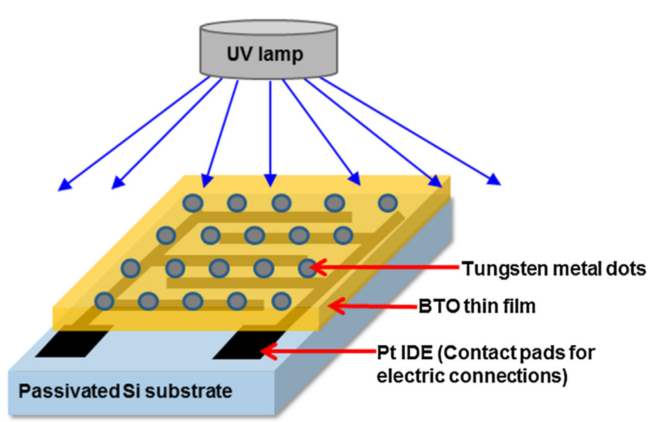

Fig. 1. Schematic of UV photodetector based on BTO thin film integrated with uniformly distributed circular dots (600 μm diameter and 20 nm thickness) of W modifier.

Table 1
Deposition parameters for tungsten (W) modifier.

Parameters	Deposited conditions
Target (T)	Tungsten (99.95% pure) (diameter: 2″)
Substrate (S)	Pt IDE/Si, fused quartz
Substrate temperature	No heating
Growth pressure	10 mT
T-S distance	6 cm
Ar:O_2	100:0
RF Power	40 W

to be 150 nm. A suitable concentration of $Ba(OH)_2$ and $(C_4H_9O)_4Ti$ aqueous solutions was added to a Teflon vessel, and the seeding layer coated sample was placed at the centre of the vessel. Subsequently the vessel was kept into a sealed autoclave at 150 °C for 12 h to perform the hydrothermal treatment of the seeding BTO layer. After hydrothermal treatment, the sample was rinsed several times with de-ionized water, absolute ethanol, and glacial acetic acid to clean the surface of the BTO thin film and then dried at 90 °C for 2 h. Tungsten (W) modifier was integrated with the prepared BTO thin film in the form of thin overlayer and uniformly distributed circular dots using rf-magnetron sputtering technique. The optimized deposition parameters for W are summarized in Table 1. W circular dots (600 μm diameter) were deposited using a shadow mask having uniformly distributed pores of 600 μm diameter. The thickness of both W overlayer and circular dots was kept fixed at 20 nm. The photodetectors based on bare BTO, W(cont)/BTO and W(dots)/BTO samples are referred to as PD-1, PD-2 and PD-3 respectively.

The structural and optical properties of the BTO thin film was studied after depositing on fused quartz substrate under identical growth condition. The crystallinity and surface morphology of thin films were investigated by X-ray diffraction (XRD) (Bruker D8 Discover) and scanning electron microscope (SEM) [Tescan, Mira 3] respectively. The optical properties of prepared samples were studied using UV–visible spectrophotometer (Perkin Elmer, lambda 35) over the wavelength range 300–1100 nm. Thickness and surface roughness measurements of thin films were carried out using surface profiler (Veeco Dektak 150). Steady-state photoresponse characteristics of UV photodetectors were measured at a fixed bias of 5 V in metal–semiconductor–metal (MSM) configuration under the illumination of UV radiation (λ = 365 nm, intensity = 24 μW/cm^2). Intensity of the UV radiations was measured with the help of an optical power metre (Newport, Model 1830C). The transient response of photodetectors was recorded at room temperature using a semiconductor characterization system (Keithley; 4200 SCS). The photoconductive gain of UV photodetector is defined as $K = I_{on}/I_{off}$, where I_{on} is the photocurrent measured under illumination of UV radiations and I_{off} is the corresponding dark current measured without any illumination.

3. Results and discussion

3.1. Structural studies

Fig. 2 shows the high resolution X-ray diffraction (XRD) pattern of the BTO thin film prepared by sol–gel hydrothermal method. All reflections in Fig. 2 are corresponding to the tetragonal structure of BTO indicating the growth of polycrystalline and single phase BTO thin film without the formation of any other secondary phases. The average crystallite size of the BTO thin films as calculated from the full width at half maxima of the dominant (1 1 0) peak is about 33 nm. No additional peak is observed in the XRD pattern for samples having W modifier, and may be attributed to the presence of very small amount of W (20 nm) over the surface of BTO thin film.