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Optical description of HfO_2/Al/HfO_2 multilayer thin film devices

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ABSTRACT

A three-layer system of dielectric/metal/dielectric (D/M/D) has been prepared on Marienfeld commercial glass substrates with Metal = Al, and Dielectric = HfO_2 for energy efficient windows applications. Subsequently, HfO_2/Al/HfO_2 multilayers have been deposited with 10 nm each HfO_2 layer and 5 nm thick Al layer using electron beam evaporation. The microstructural characteristics of D/M/D thin films have been investigated using X-ray diffraction (XRD) and atomic force microscopy (AFM). Present results indicate the formation of HfO_2 weak polycrystals embedded in the disordered lattice. AFM data reveals quite a smooth surface involving a structure of slightly elongated grains with almost Gaussian size distribution with mean grain size in the range from 7 to 23 nm. Regarding optical properties, maximum transmittance of the D/M/D structure is noticed to occur in the UV-region, whereas reflectance rises to ~60% in the visible to near infrared (NIR)-regions. To optimize the performance of these D/M/D devices, computer calculations have been performed by varying either the thickness of both HfO_2 layers and/or thickness of metallic Al layer. A satisfactory agreement between theoretical and experimental spectra is noticed. Such D/M/D structures can be useful in heat mirror applications involving energy efficient windows etc.

1. Introduction

Thin film technology is widely used in numerous industrial applications such as optical coatings, hardness coatings and opto-electronic devices. One of the most interesting thin film coating applications is an optical thin film coated on the glass window, which allows the transmission of visible light and the reflection of infrared radiations. This consequently helps to save the electrical energy used in buildings [1]. The rapid progress in solid-state electronic and optical devices would not have been possible without the development of new thin film deposition processes, improved film characteristics and superior film qualities [2]. Now-a-days thin films with different structures such as transparent conducting oxide (TCO) in the form of single/multi-layered films are being used in a variety of optoelectronic devices, namely flat displays [3], energy efficient windows [4], thin film transistors [5], gas sensors [6], light emitting diodes [7] and for photovoltaic applications [8]. Such transparent conductive thin films not only show good electrical performance but also exhibit very high transparency in the visible and high reflectance in the infrared (IR) regions [9]. In 1974, Fan et al. [10] reported a dielectric/metal/dielectric (D/M/D) multilayer system by embedding a metal mirror layer between two dielectric layers, which can suppress reflection from the metal layer in the visible region, and shows a selective high transparent effect. Since then, such D/M/D multilayer films on glass substrates have been a focus of research on selective transparent oxides [11,12] for various applications including energy-efficient windows [12,13]. Transparent heat mirror films (HMFs) are special thin coatings that transmit visible light but reflect infrared radiations. Earlier investigations have been focused on D/M/D structures such as indium tin oxide (ITO)/Ag/ITO, zinc oxide (ZnO)/Ag/ZnO, ZnO/Al/ZnO, indium zinc oxide (IZO)/Ag/IZO, zinc sulfide (ZnS)/Ag/ZnS and Zn/Al multilayered films for their improved electrical and optical properties [14–19]. Optical constants and thickness of metal as well as oxide layers play critical roles in the performance of a heat mirror [20]. Moreover, degree of crystallinity, nonstoichiometry, impurities and defects in oxide film also influence the optical properties.

A lot of research has been performed on hafnium oxide (HfO_2) during the last few years because of its high dielectric constant and refractive index, bulk modulus, melting point and chemical stability [21,22]. Moreover, it is considered as one of the most important oxide thin film materials for interference multilayer coatings down to 250 nm [23,24] and a good candidate for transparent conductive and heat mirror applications. That's why, HfO_2 has been chosen as

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the D/M/D multilayer structure for the transparent conductive applications during this research work. In addition, because of high reflectance over a broad light spectrum, environmental stability, light weight and cost effective surface finishing, aluminum thin films are widely used for reflective optics, electronic and opto-electronic device applications, and dielectric protective coatings [25,26]. HfO_2-based heat mirror ($HfO_2/Ag/HfO_2$) fabricated by Al-Kuhaili [27] showed average transmittance of 72.4% with average reflectance of 67.0% in the NIR region. Moreover, $HfO_2/M/HfO_2$ heat mirrors (with M = Hf, Mo, Al) fabricated using magnetron sputtering [28,29] illustrated high solar absorptance of 90.5–92.5% but decrease in absorptance was noted on annealing up to 500 °C. The $HfO_2/Al/HfO_2$ heat mirrors were found to be thermally stable in air up to 350 °C. A systematic study of HfO_2-based heat mirrors have not yet been made. That's why, this work was performed under three main objectives: First, the deposition of hafnium oxide thin films using conventional electron beam evaporation on glass substrates. Second, the optical characterization and applications of hafnium oxide thin films in the three-layer system [$HfO_2/Al/HfO_2$] to produce heat mirrors and/or transparent conducting films. Third, optimization of the optical performance of these heat mirrors. Present research work reports experimentally observed data on the structural and optical properties of nanostructured $HfO_2/Al/HfO_2$ thin films deposited by electron beam evaporation. These films were characterized using X-ray diffraction (XRD), atomic force microscopy (AFM) and spectrophotometry. Present results indicate the formation of nano-polycrystalline HfO_2 along with metallic Hf grains in the disordered lattice. Maximum transmittance of the D/M/D structure occurs in the UV-region which reduces to 20% in the NIR-region (optimized to 5–10%) whereas reflectance shows opposite trend i.e. it is low in the UV-region and rises to ~65% in the visible to NIR-regions (optimized to 80–85%) resulting in high refractive index in these regions.

2. Experimental

To reduce strong reflection from the metal layer, dielectric layer of high refractive index n is required. Since hafnium oxide has high value of n (1.45–1.75) [30] and is therefore a potential candidate for its use as an antireflection layer. In addition, it can be directly evaporated without the aid of a protecting atmosphere, which lowers production costs of the film. The metal layer is required mainly for high transmittance in the visible and high reflection in the IR regions and for good conductivity. Au, Ag and Al all show low absorptivity in the visible region and good electrical conductivity. Among these Al was selected as the metal layer in the present D/M/D structure. Marienfeld glass micro-slides (made of soda lime glass of 3. hydrolytic class) were used as the substrate to deposit the multilayered D/M/D structures. Before deposition, surface of each substrate was cleaned with standard chemical and ultrasonic methods using acetone, isopropyl alcohol, Piranha-solution (H_2SO_4:H_2O_2 (30%) with ratio 3:1) in ultrasonic bath successively. These glass substrates were finally dried using N_2/Ar environment.

Thin films were deposited by low energy thermal evaporating technique using BOS Edward AUTO500 electron beam evaporation system on optically plane Marienfeld (commercial) glass substrates having dimensions approx. 75 × 25 mm^2 and thickness ~1 mm. Substrates were kept at temperatures of 30 °C (A2), 100 °C (A5) and 150 °C (A8) during deposition. Metal films of ~5 nm thickness were deposited from 99.98% pure aluminum wire (Φ = 1.5 mm) and sandwiched between two dielectric layers (each ~ 10 nm thick as shown in Fig. 1) coated by e-beam evaporation of granular hafnium oxide (99.95% pure) using graphite crucible to form a D/M/D structure under vacuum (better than 10^{-5} mbar). Film thickness and deposition rates were monitored by a calibrated quartz crystal monitor (Edward FTM7). The actual film thickness was measured by spectroscopic ellipsometry showing a difference of ±1 nm among the two measurements. Surface topography of thin multi-layer D/M/D films was investigated with Agilent 5100 atomic force microscope in tapping mode. Crystal structure of these D/M/D films was investigated through x-ray diffraction (XRD) patterns recorded at room temperature using PANalytical's X'Pert PRO diffractometer equipped with Cu K_α radiations in the 2θ range 20–80°. The optical transmittance and reflectance of thin films in the wavelength range 250–2500 nm were measured using Perkin–Elmer Lambda 9 type UV/Vis/NIR dual beam spectrophotometer. The sheet resistance of the top layer in D/M/D structure was measured using van der Pauw technique.

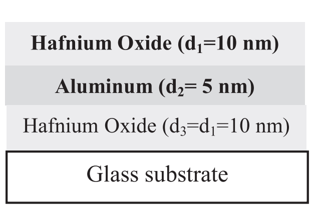

Fig. 1. Schematic three layers structure of D/M/D thin films on glass substrate.

3. Results and discussion

3.1. X-ray diffraction

The HfO_2 based D/M/D thin films deposited on clean Marienfeld glass substrates at various substrate temperatures were found physically stable and had very good adhesion to the substrates. These films were examined for their crystalline structure by recording XRD patterns at room temperature as shown in Fig. 2. Broader peaks observed in the 2θ range ~25–37° along with low intensity diffraction peaks indicate the amorphous nature of the D/M/D structure involving some polycrystalline phases. In accordance with standard JCPDS cards, three types of diffraction peaks have been identified corresponding to HfO_2 monoclinic structure [space group P21/c [31] and lattice parameters; a = 5.12 Å, b = 5.18 Å and c = 5.25 Å, Ref.# 06-0318], α-Hf (hexagonal, Ref.# 38-1478) phase, and Al (cubic, Ref.# 04-0787) phase. Presence of the metallic-Hf grains signifies that the crystal structure is multiphase and

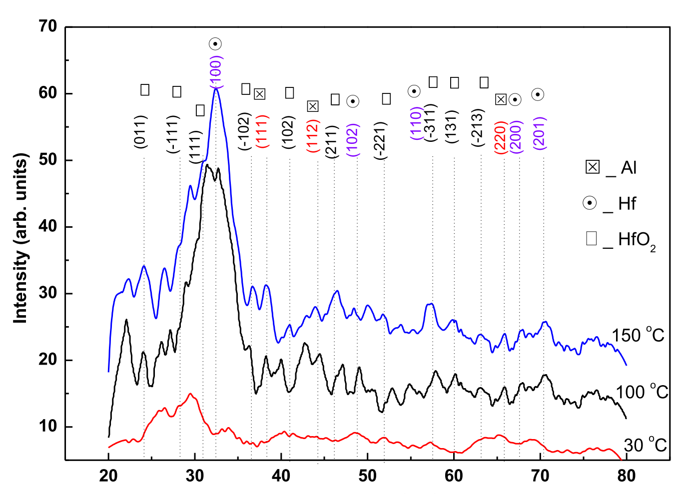

Fig. 2. X-ray diffraction patterns of $HfO_2/Al/HfO_2$ multilayers deposited at various substrate temperatures.