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Structural, electrical and optical properties of GZO/HfO_2/GZO transparent MIM capacitors

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Abstract

Metal–insulator–metal (MIM) transparent capacitors were prepared by pulsed laser deposition (PLD) on glass substrates. The effect of the thickness of the dielectric layer and oxygen pressure on structural, electrical, and optical properties of these capacitors was investigated. Experimental results show that film thickness and oxygen pressure have no effect on the structural properties. It is also found that the optical properties of the HfO_2 thin films depend strongly on both the film thickness and oxygen pressure. The electrical properties of transparent capacitors were investigated at various thickness of the dielectric layer. The capacitor shows an overall high performance, such as a high dielectric constant of 28 and a low leakage current of 2.03×10^{-6} A/cm^2 at ± 5 V. Transmittance above 70% was observed in visible region.
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1. Introduction

Transparent electronic circuits are expected to serve as the basis for new optoelectronic devices [1]. A key device for realizing transparent circuits is a transparent thin-film transistor (TTFT). The use of high dielectric constant (k) materials for the gate insulator has several potential advantages such as a compensation for the relatively low mobility in the semiconductor, a reduction of the operating voltage by increasing the gate capacitance, and an increment of the charge density at lower gate potentials [3]. Nomura et al. have reported that the use of a gate insulator with a high dielectric constant (amorphous HfO_2) results in an overall improvement of the TTFT performance [2].

Several metal oxides could be the candidates due to their high transparency and a high dielectric constant. The examples are titanium oxide (TiO_2), tantalum oxide (Ta_2O_5), zirconium oxide (ZrO_2) and hafnium oxide (HfO_2). Among these materials, both ZrO_2 and HfO_2 are good candidates once they are stable up to 900 °C [4], their bands offsets and barrier heights are suitable [5], and they have a wide band gap (higher than 5 eV) and a high k of about 20–25 [6]. The improved thermal stability and better interfacial properties after thermal annealing when compared with ZrO_2 justify the option for HfO_2 [7].

To improve the TTFT performance, extremely reliable and high-quality HfO_2 thin films are

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desired. Several thin-film growth techniques such as atomic layer deposition [8], evaporation with ion-assisted deposition [9], sol gel [10], sputtering [11], and metal–organic molecular beam epitaxy [12] have been employed to fabricate good-quality HfO_2 thin films. The properties of HfO_2 thin films have been reported to be strongly dependent on the fabrication method, and an understanding of process–structure–property correlation is of a great importance to understand and exploit these films for devices. Pulsed laser deposition (PLD) is increasingly being used to prepare a wide variety of materials in thin-film form, since PLD has been established as a simple, reliable, and fast technique that offers a great experimental versatility [13]. More importantly, the PLD technique is well known for the quality of the layers grown at relatively lower substrate temperatures than other thin-film deposition methods [13].

In order to realize TTFT devices, research on several materials is needed: transparent semiconducting oxides used as the FET active channel materials, transparent conducting oxides as the source, drain and gate metals, and transparent dielectric materials as gate dielectrics. There have been few reports on transparent dielectric materials for TTFT applications.

In this study, we investigated the structural, electrical, and optical properties of HfO_2 film deposited on Ga-doped ZnO (GZO) transparent electrodes using the PLD method to confirm the feasibility of their application in TTFTs. The effects of the film thickness and oxygen pressure on the properties of HfO_2 thin films grown at room temperature were analyzed and compared.

2. Experimental

The HfO_2 films were deposited at room temperature by PLD. The target used in this study was a sintered HfO_2 pellet. It was placed on the target holder that was constantly rotated by an external motor in order to prevent the formation of surface craters. Glass substrates were used for HfO_2 thin films at various oxygen pressures varying from vacuum to 100 mTorr and at various film thicknesses ranging from 100 to 145 nm. At first, GZO thin films were deposited at 500 °C by sputtering on glass as bottom electrodes. Following that, high-*k* HfO_2 dielectric films were deposited in oxygen ambient of 100 mTorr. Finally, GZO films were deposited at room temperature as top electrodes by PLD. The resistivity of bottom and top GZO layers is about 2.4×10^{-4} and $3.4 \times 10^{-4}\,\Omega\,\mathrm{cm}$, respectively. In our previous study we found that post-deposition annealing and rapid thermal annealing (RTA) cause the increment the leakage current and transmittance without the increment of the dielectric constant. It is because the structure of HfO_2 changes from amorphous to crystalline.

The crystal structures of HfO_2 thin films were investigated by X-ray diffraction (XRD) where a Ni-filtered Cu $K\alpha$ ($\lambda = 1.54056$ Å) source was used. The surface morphology was measured by using an Au-to-Probe CP atomic force microscope (AFM) in the contact mode. The film thickness was measured by scanning electron microscopy (SEM). The optical transmission measurements were performed using a UV-near IR grating spectrometer. For electrical measurements, the leakage current was measured using an HP4155 parameter analyzer, and the capacitance was characterized using an HP4284 precision LCR meter at 1 MHz.

3. Results and discussion

HfO_2 is a material forming several polymorphs. Pure HfO_2 tends to appear in the monoclinic phase at room temperature and atmospheric pressure. Orthorhombic and tetragonal phases can be formed at high pressures and/or high temperatures [14,15]; these phases are generally thought to be metastable if present at room temperature and atmospheric pressure. Fig. 1 shows the XRD patterns of HfO_2 films deposited at room temperature with different oxygen pressures and film thickness. As shown in Fig. 1, all films are amorphous without any crystallization peak independently of the oxygen pressures and film thickness. The broad peaks observed around 25° correspond to the glass substrate and can be attributed to the low substrate temperature. With increasing the oxygen pressure, there are no obvious effects on the crystallinity of the HfO_2 films.

Amorphous HfO_2 films may have better dielectric properties and higher laser damage threshold than crystalline films [16]. Moon et al. suggested that HfO_2 films transformed from amorphous to polycrystalline form as the film thickness increases [17]. Hang et al. suggested that the HfO_2 films transform from the amorphous to the polycrystalline form when the substrate temperature was higher than 300 °C [18].

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