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Electrical conduction mechanisms and dielectric relaxation in Al_2O_3 thin films deposited by thermal atomic layer deposition

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ABSTRACT

In the present work, aluminum oxide (Al_2O_3) thin films were deposited on *p*-type silicon substrates by thermal atomic layer deposition technique. The structural properties of as-deposited Al_2O_3 thin films were characterized by grazing-incidence x-ray diffraction and it was determined that the films had an amorphous structure. Electrical transport mechanisms and dielectrical properties of the amorphous Al_2O_3 thin films were then investigated by fabricating and characterizing Al/Al_2O_3/*p*-Si metal–oxide-semiconductor (MOS) capacitors by performing the dc current-voltage and frequency-dependent dielectric measurements. As a function of the applied gate electric fields, the current conduction in low electric fields consists of Schottky emission, but in relatively high electric fields current conduction is dominated by space-charge limited conduction. From the frequency-dependent dielectric measurements, the dielectric loss showed two peaks at the frequencies of 10 kHz and 0.7 MHz which evidence of the dielectric relaxation. The low and high frequency dielectric relaxation behaviors were attributed to space charge polarization and orientation-polarization of dipoles, respectively. It was concluded that defects related localized states in the Al_2O_3 films may contribute to dielectric properties at high frequency.

1. Introduction

Semiconductor technologies have made great strides in producing metal-oxide-semiconductor field effect transistors (MOSFETs), which are the building blocks of the integrated circuits, in low dimensions. However, the downsizing of the design dimensions of the transistors leads to an increased leakage current in transistors due to tunneling that causes the instability and loss of power. Higher leakage current will go through the insulation layer when the thickness of SiO_2 is lower than the order of magnitude of nanometer [1,2]. To minimize the leakage current in MOSFET structure, research on the use of alternative materials called high-k such as Al_2O_3, Y_2O_3, Er_2O_3, TiO_2, HfO_2, GeO_2, ZrO_2, $SrTiO_3$ etc., which have a much higher dielectric constant than traditional silicon dioxide (SiO_2) dielectrics is being done intensively [3–7]. In the family of high-k materials, the amorphous aluminum oxide or alumina (Al_2O_3) has promising properties such as high band gap, high dielectric constant ($\varepsilon_r \sim 9$) which is over twice that of silicon dioxide, high breakdown voltage, and high radiation resistance [8–13]. It is noteworthy that the reported band gap values of the Al_2O_3 thin films are not uniform, and they deviate strongly depending on the method of synthesis, film thickness, and structure of Al_2O_3. It is well known, Al_2O_3 exhibits a number of different metastable or transition phases such as γ, η, δ, θ, and χ phases. From the technological application point of view, amorphous (*am*), gamma (γ), and alpha (α) Al_2O_3 forms are of greatest interest [14].

The experimental band gap of bulk or thick Al_2O_3 was reported as 8.8 eV [15–17]. On the other hand, some experimental band gap values were reported as 8.8 eV, 7–8.7 eV, and 5.1–7.1 eV for α- Al_2O_3, γ- Al_2O_3, and *am*-Al_2O_3, respectively [14,18,19]. This indicates that the Al_2O_3 structure very effective on the band gap value of the films. Jia et al. [20] reported band gap value for *am*-Al_2O_3 as 6.7 eV. Henkel et al. [21] reported band gap value for *am*-Al_2O_3 as 7 eV. Afanes'ev et al. [22] reported band gap energy of *am*-Al_2O_3 as 6.2 eV.

In addition, it is thermodynamic stable on silicon and adheres to the surface well. These properties make it a suitable insulation layer for silicon based metal-oxide-semiconductor (MOS) devices [23–28]. Many techniques have been used to deposit Al_2O_3 thin films such as chemical vapor deposition (CVD) [29], physical vapor deposition (PVD) [30], pulsed laser deposition [31], sputter deposition [32], molecular beam epitaxy (MBE) [33], and atomic layer deposition (ALD) [34–37]. Among them, the ALD technique is of great interest because this technique offers low temperature deposition, perfect uniformity and conformality, and enables accurate control of film thickness at an atomic layer level due to the film growth based on self-limiting surface

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reactions [36].

This paper is focused on the investigating of the electrical transport mechanisms and dielectric behavior of amorphous Al_2O_3 thin films grown at low temperatures by using thermal ALD technique. Investigations of the electrical and dielectrical behavior of the gate oxides is one of the most crucial and important tasks in microelectronic industry.

2. Experimental method

The Al_2O_3 thin films were deposited on *p*-type Si (100) substrates at a temperature of 200 °C using a Savannah thermal ALD system. Trimethylaluminum ($AlMe_3$, TMA) and water (H_2O) sources were used as the aluminum and oxygen precursors, respectively. Before the deposition, the Si substrates were cleaned by acetone ultrasonication and finally by HF acid to remove the native oxide on the surface of substrates and immediately loaded into the ALD reactor. In the ALD deposition chamber, one growth cycle followed steps; a) exposure to the metal precursor (TMA); b) purge; c) exposure to H_2O and d) another purge. Exposure time was used as 0.015 s for trimethylaluminum (TMA) and water (H_2O) precursors and the purge time was adjusted to 10 s. The growth rate was roughly 1.1 Å/cycle. Variable Angle Spectroscopic Ellipsometer (VASE) was used for determining the thicknesses of the films deposited via ALD. Cauchy dispersion function was used for estimating film thicknesses. After deposition of the Al_2O_3 thin films, 80-nm Al layer was then coated back side of the Si wafer using a thermal evaporation system as the bottom electrode and the films were annealed at 450 °C for 2 min under 100 sccm N_2 flow to obtain ohmic form. After, 80-nm Al layer was finally deposited onto the Al_2O_3 thin films with 2 mm diameter circular dots via a metal mask to form the gate electrodes. All processes were done in class 100 and 1000 cleanroom facilities. In order to study electrical and dielectrically properties of the amorphous Al_2O_3 thin films, frequency-dependent (1–800 kHz) capacitance-voltage (*C-V*) and current-voltage (*I-V*) characteristics were performed using a semiconductor parameter analyzer (Keithley 4200-SCS), which is connected to a DC probe station (Cascade Microtech PM-5). The structural properties of Al_2O_3 thin films were studied using grazing incidence X-ray diffraction (GIXRD) system.

3. Results and discussion

3.1. Structural and electrical properties of ALD deposited Al_2O_3 thin films

GIXRD characterization of the as-deposited Al_2O_3 thin films is shown in Fig. 1. The as deposited films (no post-deposition thermal process) show the typical pattern with no obvious diffraction peaks, the as-deposited Al_2O_3 dielectric thin films were amorphous structures.

The conduction current measurements in MOS capacitors using thermal ALD deposited Al_2O_3 thin films as a gate dielectric have been carried out and leakage current density-voltage (*J-V*) characteristics are shown in Fig. 2, in which the current density depends strongly on the applied gate voltage. The plot shows two regions and (≤ 1 V and 1–13.5 V). Above 1 V, the leakage current increased abruptly. This is due to different current conduction mechanisms dominate each of those regions and some of the possible conduction mechanisms were used to simulate and analyze the experimental values.

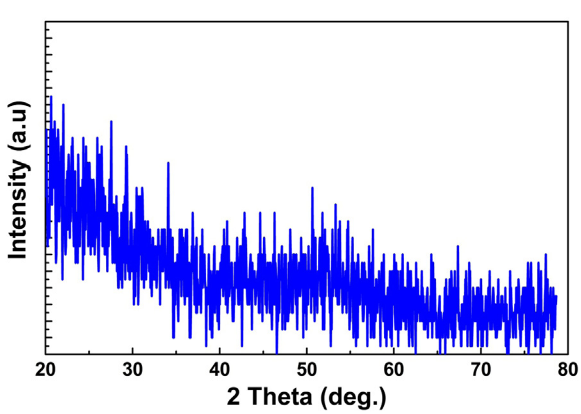

Fig. 1. GIXRD characterization of Al_2O_3 gate dielectric thin film deposited at 200 °C on Si (100).

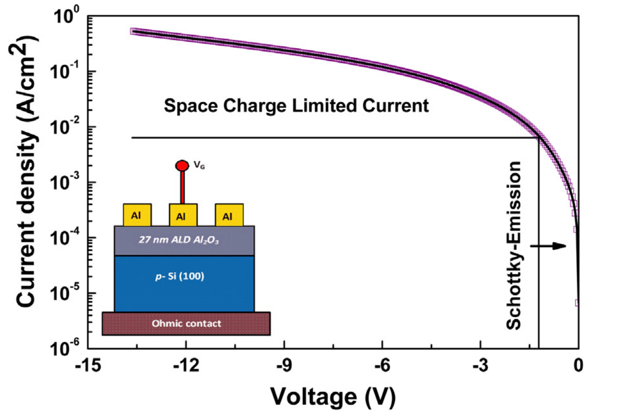

Fig. 2. *J-V* characteristics of Al/Al_2O_3/*p*-Si MOS capacitors under accumulation mode.

Some conduction mechanisms are called electrode-limited conduction mechanisms depend on the electrode-dielectric interface, but, other conduction mechanisms are called bulk-limited conduction mechanism depend on dielectric properties. First, the conduction mechanisms at low applied fields will be considered.

At low electric fields or low gate voltages (≤ 1.1 V), the best fitting were obtained from ln *J* vs. $E^{1/2}$ plot (R^2 = 0.9999). Schottky emission (SE) mechanism is described as [38];

$$J \propto A^* T^2 exp\left[\frac{-q(\phi_B - \sqrt{qE/4\pi\varepsilon_r\varepsilon_o})}{k_B T}\right] \quad (1)$$

where J is the current density, E is the electric field, T is the temperature in Kelvin, A^* is the effective Richardson constant, ϕ_B is the barrier height, ε_r is the dielectric constant of the dielectric films, q is the electronic charge, ε_o is the permittivity of free-space, and k_B is Boltzmann constant. E values were calculated from $(V-V_{FB})/t_{Al_2O_3}$, where V_{FB} is the flat-band voltage and $t_{Al_2O_3}$ corresponds to the thickness of thermal ALD deposited Al_2O_3 film. According to Eq. (1), if the Schottky emission mechanism dominates the carrier transport, the logarithm of the current depends linearly on the square root of the applied voltage as shown in Fig. 3. However, finding a linear correlation between current density and voltage (or applied electric field) does not guarantee that

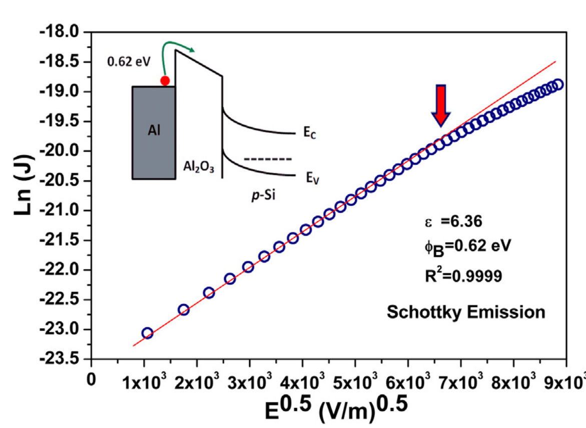

Fig. 3. Schottky emission plot of Al/Al_2O_3/*p*-Si MOS capacitors.