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Bipolar resistive switching properties of AlN films deposited by plasma-enhanced atomic layer deposition

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ABSTRACT

AlN thin films deposited by plasma-enhanced atomic layer deposition (PEALD) have been used to investigate the resistive switching (RS) behavior. The bipolar RS properties were observed in the Cu/PEALD-AlN/Pt devices, which are induced upon the formation/disruption of Cu conducting filaments, as confirmed by the temperature dependent resistances relationships at different resistance states. The resistance ratio of the high and low resistance states (HRS/LRS) is 10^2–10^5. The dominant conduction mechanisms at HRS and LRS are trap-controlled space charge limited current and Ohmic behavior, respectively. This study demonstrated that the PEALD-AlN films have a great potential for the applications in high-density resistance random access memory.

1. Introduction

Bipolar resistive switching (BRS) property is an interesting phenomenon, which has attracted great attention as the next generation nonvolatile resistance random access memory (ReRAM) [1–3]. The BRS behavior originated from the repeated resistance change between the high resistance state (HRS) and low resistance state (LRS), has been observed from various material systems, including solid electrolytes, perovskites, organics, binary transition metal oxides and nitrides [2,4–11].

Owing to its high thermal conductivity, good insulating properties and high dielectric constant, aluminum nitride (AlN) is an important wide band gap semiconductor and is ideally suitable for applications in microelectronics [11–14]. Recent researches have shown some attractive RS properties from AlN-based structures such as Cu/AlN/Pt and Pt/AlN/Pt devices. In general, the AlN films were deposited by physical vapor-phase deposition (PVD) method, especially the magnetron sputtering [11,13]. However, there has been limited activity in using chemical vapor-phase deposited (CVD) AlN films to make the AlN-based RS devices. Due to its precise thickness control, large area uniformity and low process temperature, atomic layer deposition (ALD), a special CVD technique, has been established as an excellent method for semiconductor processes, especially when the films are extremely thin in nanometers range. Furthermore, ALD growth process has a perfect step-coverage, which is particularly suitable for three-dimensional (3D) memories which are constructed in the cross-bar architecture [15,16]. Thus, it is of great interest to investigate whether AlN thin films grown by ALD can be used for ReRAM application.

In this paper, we report the BRS behaviors in AlN films deposited by a plasma-enhanced ALD (PEALD) method and discuss the possible physical mechanism of the BRS behaviors.

2. Experimental

2.1. Material preparation

AlN films were prepared on Si(1 0 0) and Pt(1 1 1)/Ti/SiO_2/Si substrates using a showerhead-type ALD system (Kurt J. Lesker Co. ALD-150LX), which has a remote plasma generation configuration. Trimethylaluminum (TMA, 99.999%) was used as the metal precursor for the deposition. The nitrogen source (N-source) reactant was hydrogen nitrogen mixture plasma, which was generated remotely in the upper part of the reaction chamber within a separate generation chamber. The generated radicals were flowed into the deposition chamber. The carrier gas and purging medium were 99.999% purity argon. During the growth process, the pressure in the chamber was ~1 torr, and the substrate temperature was held at 332 °C. The flow rates of the carrier gas and the N-source gases (N_2:H_2 = 1:1, 99.999%) were 20 and 60 sccm, respectively. A typical plasma enhanced ALD growth sequence was composed of 0.1 s of metal precursor exposure, 5 s of Ar purging, 5 s of N-source

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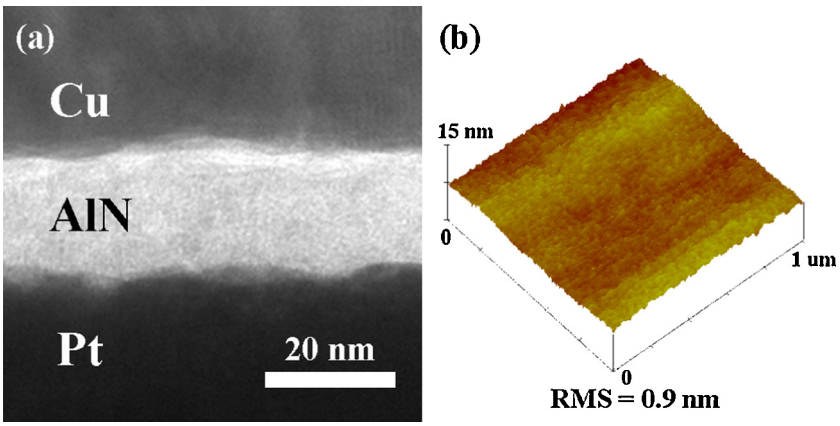

Fig. 1. (a) The cross-sectional TEM image of a Cu/PEALD-AlN/Pt structure. (b) AFM image and RMS data of the PEALD-AlN films.

reactant exposure with an RF power of 600 W, and 5 s of Ar purging. The films were deposited for 250 ALD cycles.

2.2. Device fabrication and characterization

The thickness of all the films was measured by both ellipsometer and transmission electron microscope (TEM). The microstructures of all the films were characterized by a 200 kV field emission TEM (Tecnai F20). The TEM specimens were prepared using FIB etching technique to reveal the cross section of the Cu/AlN/Pt structure. Topographic images of the films were obtained using an atomic force microscope (AFM, Veeco) in the tapping mode at a nominal load of ~2 nN, and the scanning rate for a 1 × 1 μm image was 1.0 Hz.

The chemical state of each element in the films was analyzed by x-ray photoelectron spectroscopy (XPS, Escalab 250Xi) with Al K_α radiation of 1486.6 eV. The samples were etched by Ar^+ bombardment for ~3 nm to avoid interference of surface-absorbed oxygen and surface-oxidized nitride layer. Data collection was consisted of a survey scan ranging from 0 to 1300 eV followed by multiplex scans on Al2p and N1s peaks. The scan step for all peaks was 0.05 eV/step and 60 ms/step.

For electrical measurement, the Cu top electrodes (100 nm) with a diameter of 200 μm were deposited by thermal evaporation using a metal shadow mask to form the Cu/AlN/Pt sandwich device structure. The current-voltage (*I–V*) characteristics of the devices were measured using a semiconductor parameter analyzer (Agilent B1500A) in the voltage sweeping mode at room temperature in air.

3. Results and discussions

Using the deposition parameters listed above, the film growth process is self-limiting. As shown in Figure S1, the AlN film was polycrystalline with a wurtzite structure. The growth rates for the AlN films were 0.08 nm/cycle, confirmed by an ellipsometer. Fig. 1(a) shows the cross-sectional TEM image of a Cu/AlN/Pt structure. The thickness of the AlN films was experimentally determined to be ~ 20 nm, consistent with the design. Fig. 1(b) shows the surface morphologies of the AlN films measured by AFM. The root-mean-square (RMS) surface roughness of the films was ~ 0.9 nm, implying a smooth flat surface.

Combined with the surface chemistry of ALD process [15], the main possible reaction process can be written as follows,

$$Al(CH_3)_{3(ads)} + N_{(g)} + 3H_{(g)} \rightarrow AlN_{(ads)} + 3CH_{4(g)}$$

XPS was employed to analyze the chemical composition of the AlN films. The semiquantitative analysis of approximate composition (N/O in at%) for AlN films, which was tested by XPS, was 80.3:19.7. These results demonstrated the existence of oxygen elements in the films. Fig. 2 shows the XPS spectra and their Gaussian fittings of the Al2p, N1s and O1s peaks for the AlN films. These peaks were fitted with two to three subpeaks, representing their distinct chemical states. The details of the chemical states for each subpeak are summarized in Table 1.

As shown in Fig. 2(a), the Al2p #1 and #2 subpeaks at ~74 and 75 eV correspond to the Al—N and Al—N—O bond, respectively [17,18]. Al—N—O is generally referred to as aluminum oxynitride

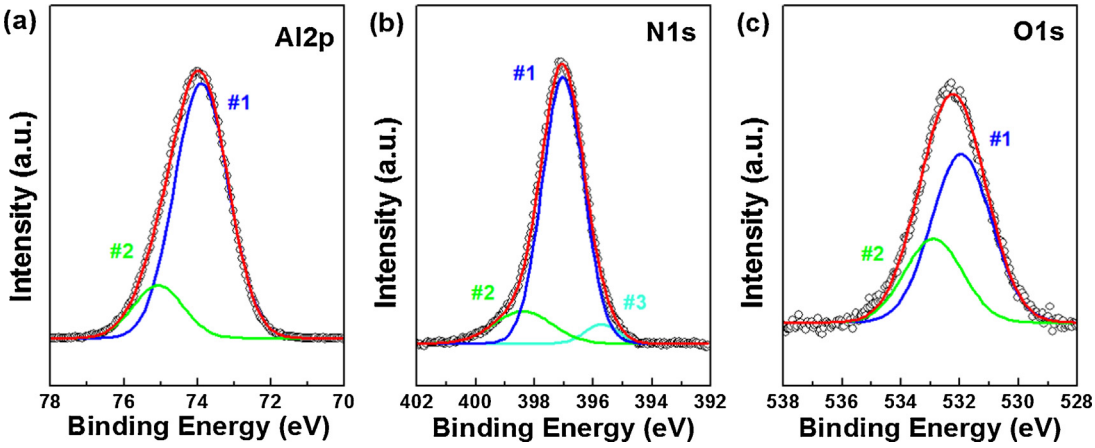

Fig. 2. XPS spectra and their Gaussian fittings of the Al2p (a), N1s (b) and O1s (c) peaks for the PEALD-AlN films, respectively. The colored lines are the fittings.