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Abnormal bipolar resistive switching behavior in carbon-iron composite films with different thicknesses

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ABSTRACT

Carbon-iron composite films with different thicknesses (50, 100 and 150 nm) were deposited by DC magnetron sputtering method using a composite target (Fe: 4 at%, C: 96 at%). The resistive switching behaviors of Pt/Al/*a*-C:Fe/Au/Ti structures with different *a*-C:Fe film thicknesses are investigated. Abnormal bipolar resistive switching characteristics were observed in all Pt/Al/*a*-C:Fe/Au/Ti memory cells. Considering the endurance and retention properties, the RRAM cell with 100 nm thick *a*-C:Fe film as the dielectric layer showed the best performance with on/off-resistance ratio ~10, and retention time $>10^4$ s. Based on the results of the X-ray photoelectron spectroscopy depth profile, the resistive switching behavior is attributed to mutative Al^{3+} fraction at the Al/*a*-C:Fe interface and the migration of Au ions between two electrodes.

1. Introduction

With flash memory reaching its scaling down limits, resistive random access memory (RRAM) has attracted considerable attention in the past few years due to its low power consumption, high write/read speed, simple structure, and high scalability characteristics [1–3]. Generally, with respect to the electrical polarity required for resistive switching, normal resistive switching (RS) behaviors can be classified into unipolar resistive switching (URS) and bipolar resistive switching (BRS). URS and BRS have been found in a wide variety of materials, including metal oxides [4–9], organic compounds [10–12], chalcogenide materials [13], and some kind of carbon-based films [14–17]. However, abnormal bipolar-like resistive switching (ARS) phenomena was recently reported in ZnO thin films [18] and Nb:$SrTiO_3$ (NSTO) [19], in which the RS took place at zero bias voltage.

Compared with other dielectric materials utilized in RRAM, carbon-based materials have many advantages including scaling down endurance, low cost, high stability and compatibility with standard CMOS process [2]. Fu et al. proposed a RRAM device based on nanoscale diamond-like carbon films, exhibiting URS behavior with on/off-resistance ratio $R_{on/off} > 300$ [14]. Zhuge et al. prepared metal/*a*-C:H/Pt sandwiched structures with different top electrodes (TE) and BRS characteristics with a retention time of more than 10^5 s were achieved in the Cu/*a*-C:H/Pt cell [15]. Amongst the various carbon-based materials, nano-composite carbon-based thin films, consisting of nano-particles of ferromagnetic metals (Fe, Co, Ni) embedded in amorphous carbon films, are of potential use in high-density magnetic recording media due to their peculiar magnetic and magneto-transport properties like enhanced coercivity and giant magneto-resistance [20]. However, little investigation has been done on nano-composite carbon-based materials. Thus, attempts to fabricate ferromagnetic element-amorphous carbon (*a*-C) composite based memory cells and understanding of their RS mechanism are crucial for RRAM applications discussed above.

In this study, RS characteristics of carbon-iron composite (*a*-C:Fe) films with different thicknesses ($t = 50$, 100 and 150 nm) were investigated systematically. Repeatable and reproducible switching phenomena were observed in these Pt/Al/*a*-C:Fe/Au/Ti memory cells and the possible RS mechanism is discussed.

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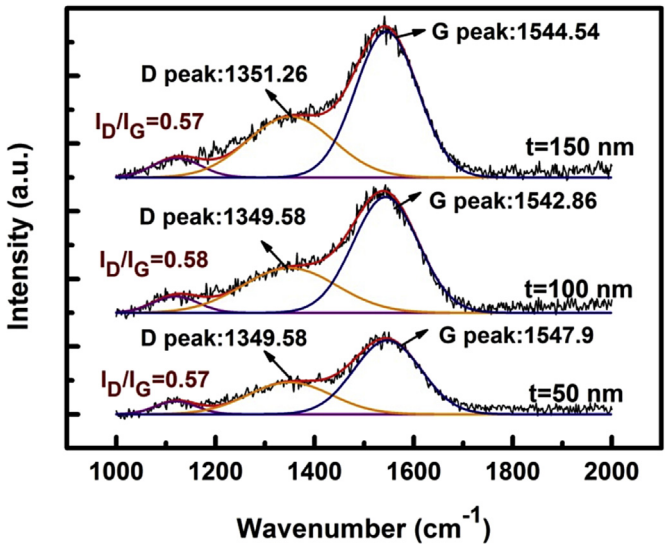

Fig. 1. Raman spectra of *a*-C:Fe films with different thicknesses (t = 50, 100, 150 nm).

2. Experimental

To prepare the samples, a 5-nm-thick Ti adhesion layer and a 50-nm-thick Au bottom electrode were deposited on glass substrates by dual ion-beam sputtering. Subsequently, *a*-C:Fe thin films with different thicknesses (t = 50, 100 and 150 nm) were grown on the Au/Ti/glass substrates at room temperature by DC magnetron sputtering. A commercial composite sputtering target (Fe:4 at%, C:96 at%) was employed. Prior to deposition, the base pressure in the chamber was pumped to less than 1×10^{-3} Pa. During the deposition, the sputtering pressure of argon and the DC power were kept at 0.4 Pa and 100 W, respectively. After *a*-C:Fe film growth, top electrodes Al/Pt (~70 nm/~5 nm) were deposited by dual ion-beam sputtering, forming a cross-bar structure with the bottom electrodes (see Fig. 2(a)).

Raman scattering measurements (JY, HR800UV) were employed to study the structure of *a*-C:Fe films with the wavelength of 514.5 nm excitation. The cross-sectional morphology of the multilayered structure with t = 100 nm was examined by scanning electron microscopy (SEM, S-4800, Hitachi). Current-voltage (I–V) characteristics of the cells were measured in a voltage sweep by Keithley 2400 semiconductor characterization system at room temperature. The chemical states of elements near the surface and after sputter etching were analyzed by X-ray photoelectron spectroscopy (XPS, Thermo ESCALAB 250Xi) with focused monochromatic Al Kα radiation. Depth profiles of O 1*s* and Al 2*s* were acquired from Pt/Al/*a*-C:Fe/Au/Ti multilayer structure with a pass energy of 30 eV at 0.1 eV step size and Ar^+ beam energies of 1 kV rastered over 2 mm × 2 mm. All the XPS spectra reported here were calibrated against the C 1*s* peak (284.8 eV) of adventitious carbon. A 200 μm x-ray beam size was used for all acquisitions.

3. Results and discussions

Fig. 1 shows the Raman spectra of *a*-C:Fe films with different thicknesses (t = 50, 100, 150 nm). Raman bands were plotted by fitting the measured spectra and decomposing the fitted curves into Gaussian components. Two characteristic peaks related to amorphous carbon appeared, which are designated as D peak at around 1350 cm^{-1} and G peak at around 1545 cm^{-1}. It is well known that the peak positions and the ratio of the intensities between D and G peaks (I_D/I_G) provide information on sp^2/sp^3 and the sp^2 cluster size in the films. As shown in Fig. 1, no significant change in both peak position and I_D/I_G ratio is noticed, indicating that the thickness has little impact on the structure of *a*-C:Fe films in our study.

The schematic diagram and the cross-sectional SEM image of the Pt/Al/*a*-C:Fe/Au/Ti/glass multilayered structure are shown in Fig. 2(a). During the measurement, a bias voltage was applied between the bottom and the top electrode with the latter being grounded.

Repeatable abnormal bipolar resistive switching behaviors were observed in all memory cells with different *t*. Taking memory cells

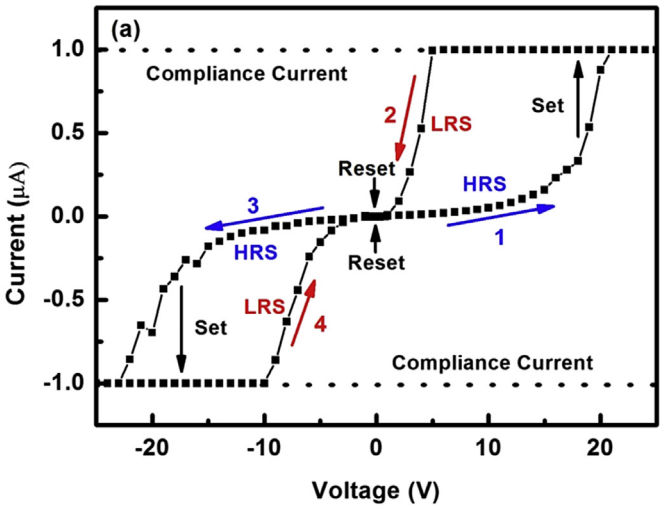

Fig. 3. *I-V* characteristics of Pt/Al/*a*-C:Fe (100 nm)/Au/Ti memory cell in linear scale.

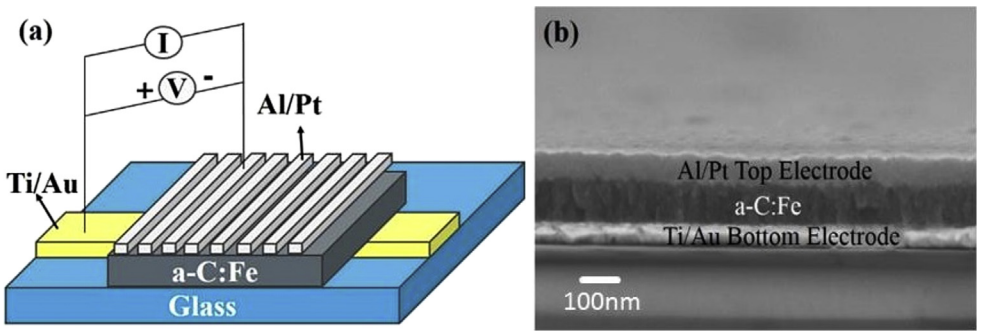

Fig. 2. (a) Schematic diagram of the Pt/Al/*a*-C:Fe/Au/Ti/Glass multilayered structure. (b) Cross-sectional SEM image of a typical multilayered structure with *a*-C:Fe film thickness t = 100 nm.