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Investigation of resistive switching in graphite-like carbon thin film for non-volatile memory applications

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ABSTRACT

Graphite-like carbon films were deposited on $Al/SiO_2/Si$ substrates by using Direct Current magnetron sputtering method. Stable and reliable bipolar resistive switching (RS) characteristics were observed in $Cu/a\text{-}C/Al/SiO_2/Si$ multi-layer structures. An ON/OFF ratio of about 3, a retention time of more than 10^5 s, and switching threshold voltages of less than 3 V were achieved. I–V properties in both low resistance state and high resistance state can be well explained by the space charge limited current model. The observed RS behaviors are attributed to the electron trapping and detrapping at deep level defects in a-C films.

1. Introduction

Resistance random access memory (RRAM) based on the resistive switching (RS) effect induced by external electrical stimulus has attracted considerable attention in the past few years for its low power consumption, high operation speed, high scalability and simple structure [1–5]. Although various models have been suggested to be responsible for the RS behavior, such as conducting filament model [6–9], classical trap-controlled space charge limited current (SCLC) model [10–12], Simmons and Verderber (*S*–*V*) model [13,14], and Poole-Frenkel effect [15], the exact switching mechanism has not been fully understood yet. Up to now, the RS effect has been found in a wide variety of materials, including binary metal oxides [5,16,17], perovskite manganites [18,19], chalcogenide materials [20], silicon-based materials [4,9], and organic materials [21]. Comparing to these traditional materials utilized in RRAM, carbon-based films have inspired great interests for their unique advantages including controllable preparation processes, low cost, high stability and compatibility with standard CMOS process.

Stable, rewritable and non-volatile switching behavior has been observed in lithographic graphitic stripe based two-terminal memory cells [22]. Zhuge et al. further found RS effect in graphene oxide (GO) thin films and attributed this phenomenon to the formation and rupture of conducting filaments [7,8]. With different deposition methods and conditions, amorphous carbon (a-C) films contain different volume fraction of the sp^3 bonds and form a great variety of microstructures, which result in various electronic properties [23–25]. Considering the unique advantages mentioned previously, it is interesting and meaningful to investigate the RS properties in amorphous carbon films and fabricate a-C based RRAM. Hydrogenated a-C films derived by linear ion-beam deposition technique showed resistance memory phenomena with device yield 90%, ON/OFF ratio > 100, and retention time $> 10^5$ s [7]. At a later time, Xie et al. obtained RS effect in diamond-like carbon (DLC) films deposited by filtered cathodic vacuum arc (FCVA) and pulsed laser deposition (PLD) [6,26]. Compared to these methods mentioned before, DC magnetron sputtering has advantages of uniformity in large-scale wafers and high device fabrication yield which is more suitable for industrial process. Furthermore, the mechanism for carbon-based RRAM is still controversial. This work would be benefit for understanding the resistive switching mechanism more clearly.

In this paper, the RS characteristics of a-C films grown by DC magnetron sputtering were investigated extensively. Stable bipolar resistive switching phenomena have been observed in the Cu/a-C/Al structure memory cells. The mechanism of the switching phenomenon was also explained by trap-controlled SCLC model in detail.

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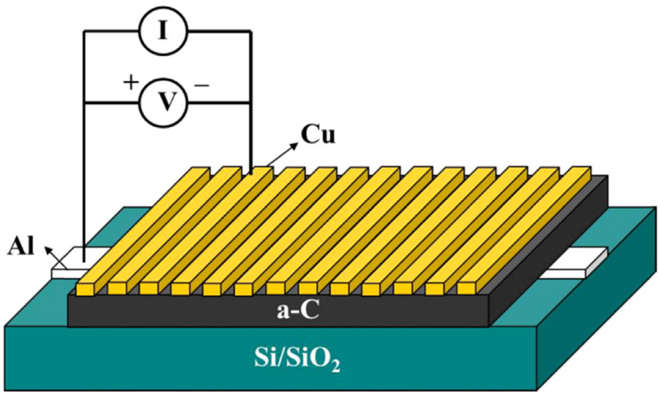

Fig. 1. A schematic diagram of the two-terminal devices and the measurement configuration.

2. Experimental details

Before film deposition, Al bottom electrodes (BE ~ 100 nm) were grown on commercial SiO_2/Si substrates by dual ion-beam sputtering at room temperature. Then amorphous carbon films (about 200 nm in thickness) were deposited on Al-BE/SiO_2/Si substrates at different substrate temperature (200 °C, 300 °C and 400 °C) by DC magnetron sputtering. The target was highly pure graphite (~99.99%) and the base pressure was pumped to less than 10^{-4} Pa. During deposition, the sputtering power was 90 W and the working pressure of argon was kept at 0.4 Pa. In order to measure the RS properties of the films, Cu top electrodes (TE) in a line width of 100 μm were grown by ion-beam sputtering after films deposition, forming a cross-bar pattern structures (see Fig. 1). The morphologies and microstructures of as-deposited films were examined by atomic force microscope (Bruker, Dimension Edge) and Raman spectroscopy (JY, H800UV), respectively. *I*–*V* characteristics were measured in a voltage sweep mode using a Keithley 4200 semiconductor characterization system at room temperature. As schematically shown in Fig. 1, a bias voltage was applied between the bottom (Al) and top (Cu) electrodes with the latter being grounded during the measurement. The resistance in the low resistance state and high resistance state were measured as a function of temperature by using a two probe method with a Keithley 2400 sourcemeter equipped with a liquid nitrogen cooling system.

3. Results and discussions

3.1. Raman spectra and AFM of a-C films

The typical Raman spectrums excited by 514 nm wavelength of a-C films deposited at different temperature are shown in Fig. 2(a). To obtain the positions and intensities of the D and G peaks, we fit the spectra using Double-Gaussian peak model. The ratio of the intensities between D and G peak (I_D/I_G) provides information on sp^2/sp^3 and is widely used to estimate the sp^2 "grain size". The full width at half maximum of G peak ($FWHM_G$) is proportional to the disorder degree of the a-C film. Well-pronounced G peak (graphite peak) centered at ~1550 cm^{-1} and D peak (disordered peak) at ~1380 cm^{-1} can be found in Raman spectrums (see Fig. 2(a)). Fig. 2(b) summarizes the variations of the ratio sp^2/sp^3 and $FWHM_G$ as a function of substrate temperature. From The Raman spectra and fitted results, three obvious features can be observed: with the deposited temperature increasing, (1) G-peak position increases gradually; (2) the ratio of I_D/I_G increases from 0.95 to 1.14; (3) $FWHM_G$ decreases from 143 to 126 cm^{-1}, which means dispersion of the G peak decreases. According to A. C. Ferrari's model for Raman spectroscopy of amorphous carbon [23–25], the as-deposited a-C films contain less than 20% sp^3 bonds (usually called graphite-like carbon). What's more, both the sp^3 fractions in volume decreased and the disorder degree of a-C films decreased with the increasing of the substrate temperature. Fig. 3 is the typical 3D AFM image of a-C films in a scan range of 1 × 1 μm^2. The root mean square of the roughness of the films increases from 0.274 nm to 1.22 nm, when the substrate temperature increases from 200 °C to 400 °C. The increasing of surface roughness may be attributed to the increasing of the sp^2 "grain size". The surface roughness results indicates the film surface is of a highly consistency

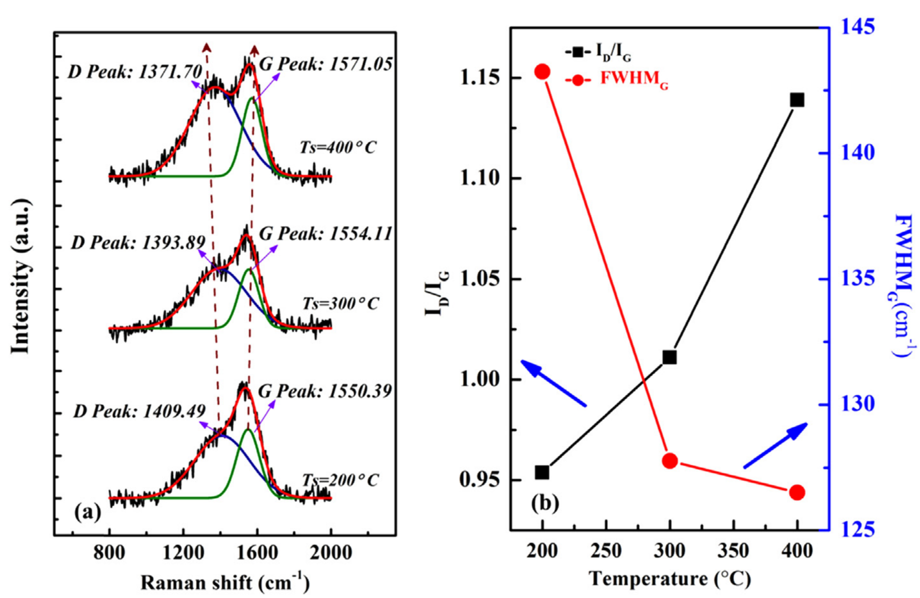

Fig. 2. (a) The typical Raman spectrum of a-C films deposited at various substrate temperature, fitted by Double-Gaussian peak model. (b) Substrate temperature dependence of the full width at half maximum of G peaks ($FWHM_G$) and the ratio of the intensity of D and G peaks (I_D/I_G).