Microelectronic Engineering 109 (2013) 75–78

Contents lists available at SciVerse ScienceDirect

Microelectronic Engineering

journal homepage: www.elsevier.com/locate/mee

Microscopy study of the conductive filament in HfO_2 resistive switching memory devices

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ARTICLE INFO

Article history:
Available online 3 April 2013

Keywords:
Resistive switching
Conductive filament
Electron energy loss spectroscopy

ABSTRACT

A detailed physical analysis of the conductive filament electrically formed in HfO_2-based resistive switching memory devices with both Hf and Ti metal oxygen exchange layers is presented. The filament, observed by applying transmission electron microscopy (TEM), scanning TEM (STEM), and electron energy loss spectroscopy (EELS) techniques to 50×50 nm^2 cells, is a cone-shaped metal-rich region in the HfO_2 dielectric of the resistive switching device.

1. Introduction

Resistive switching (RS) random access memory (RRAM) based on the resistance change of transition metal oxides, such as HfO_2, has attracted significant interest due to its low power operation, switching speed, high endurance and dense integration. The conductive filament formation mechanism in RRAMs is not yet fully understood, although it has been shown that the RS properties strongly depend upon the metal electrodes. Proposed models [1–6] (primarily based on electrical characterization) agree that the switching phenomenon is due to formation and rupture of a conductive filament. However, the filament physical properties remain a controversial issue, in part due to a complexity associated with locating a filament in the device, and preparing a sample for TEM study without affecting the filament composition. Direct microscopic observation of the conductive filament in the memory cell is thus critically important to support RS models. In this study, we have employed scanning transmission electron microscopy (STEM) and electron energy loss spectroscopy (EELS) to observe electrical stress-formed conductive filaments in the HfO_2-based crossbar devices with Hf or Ti top metal gettering or oxygen exchange layers (OEL), and TiN electrodes.

2. Experimental procedure

Crossbar devices with size of 50 nm × 50 nm have been manufactured using either Hf or Ti metal layer over the HfO_2 dielectric film as an oxygen exchange layer for oxygen gettering. TiN bottom electrodes have been employed in both type of devices. Thickness of HfO_2 layer is 5 nm.

Current–voltage (I–V) characteristics for both fresh and formed devices have been measured as a function of temperature in the −50 °C to 100 °C range using an Agilent HP4156B parameter analyser. The conductive filament formation (the forming operation) has been achieved by DC I–V voltage sweep at room temperature.

The structural characterization of the conductive filament by the Transmission Electron Microscopy (TEM) technique has been performed using a JEOL JEM2010 equipped with the electron energy loss spectroscopy (EELS) and with scanning TEM (STEM) imaging capabilities. To study the morphology of the formed conductive filaments, two different techniques have been employed: STEM in dark field configuration and EELS at low energy. The former is sensitive to the local average atomic number while the latter is strictly related to the plasmon losses, determined by the local chemical composition and phase. Resulting micrographs have a lateral resolution of about 1 nm, mainly determined by the STEM electron beam size.

3. Results

Fig. 1(a) shows the I–V characteristics measured from −50 °C to 100 °C, for a fresh (prior to forming) 50 nm × 50 nm crossbar device. The fresh cell exhibits low conductivity in the entire temperature range, with its conductance (dI/dV) exponentially increasing as a function of temperature (see Fig. 1(b)). Extracted activation energy for the conductance is 0.11 eV.

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http://dx.doi.org/10.1016/j.mee.2013.03.145

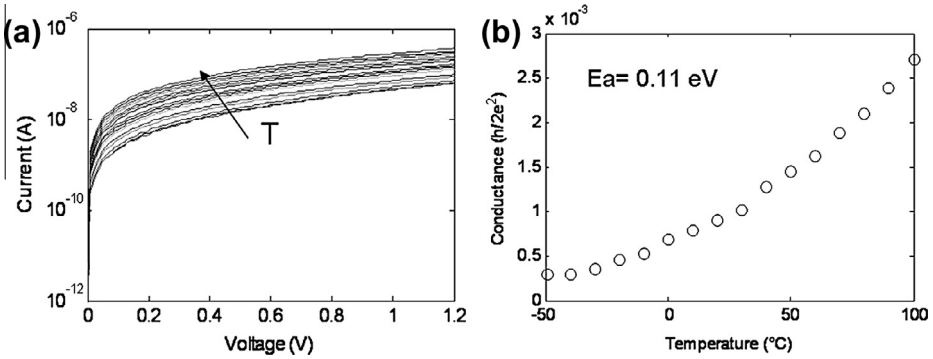

Fig. 1. (a) The current–voltage measurements performed on a fresh device in the temperature range of −50 °C to 100 °C. (b) Conductance versus temperature dependency extracted from the data in (a).

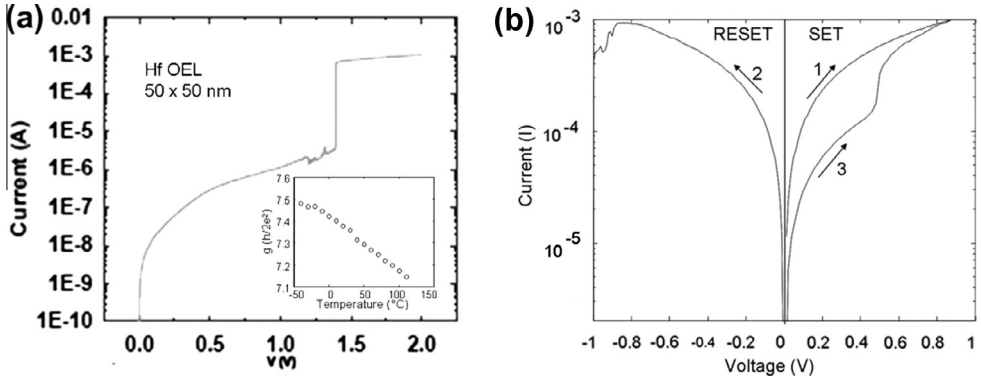

Fig. 2. (a) The current–voltage characteristic measured during the forming operation. In the inset: Conductance versus temperature measured after the filament was formed. (b) Current–voltage characteristics after filament formation. The device is initially in the SET state (line 1). It can be reset by applying negative voltage (line 2) and then set again by positive voltage (line 3).

Fig. 2(a) shows *I–V* sweep performed under the current compliance limit of 1 mA at room temperature, during which the formation of the conductive filament occurred at around 1.4 V. After the filament formation, the device exhibits much higher conductance, which linearly decreases with temperature as shown in the inset of Fig. 2(a), thus suggesting that the conductive filament is of metallic nature [6]. Fig. 2(b) shows typical operation of a formed filament as resistive switch memory. After filament formation the device is in the high conductive "SET" state (line 1). By applying negative voltage it can be reset (line 2) to a low conductive state, and then set again by positive voltage (line 3).

To study the morphology and microstructure of the conductive filament formed under these conditions, a number of crossbar devices after the forming have been analysed by TEM. To avoid a filament oxidation, which may occur when the sample cross-section is done through the filament region, the entire device volume was included in the TEM specimen, which was about 60 nm thick, as prepared by using a Focused Ion Beam tool.

Fig. 3 reports a STEM dark field micrograph showing a bright conically shaped region in the HfO_2 oxide. The contrast in this image is determined by the local electronic density, i.e. by the local atomic number *Z* per unit volume. Therefore, a brighter area in this image corresponds to a region in the insulating layer with higher average *Z*. This observation can be explained by considering that a metallic Hf-rich filament was formed in HfO_2.

The atomic density of the pure metallic Hf is 4.5×10^{22} atoms/cm^3 while the Hf atomic density in HfO_2 is 9.27×10^{21} atoms/cm^3. Therefore, in the 60 nm thick sample the number of Hf atoms in pure Hf and in HfO_2 is 2.7×10^{17} atoms/cm^2 and 5.56×10^{16} atoms/cm^2, respectively. If a pure metallic Hf filament with the width and thickness of 5 nm is formed in the HfO_2 film, then the local Hf atoms concentration should increase by 30%. Therefore, given a large difference of the Hf atoms density in the metallic and dielectric phases, even a few nm of metallic Hf within the Hf oxide volume can be clearly seen by the *Z* contrast. This explains well the image in Fig. 3, which indicates the presence of a metallic Hf-rich filament in HfO_2.

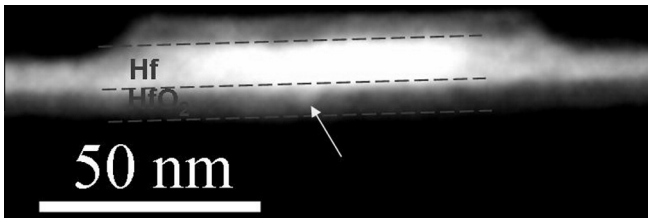

Fig. 3. STEM dark field image of a crossbar device.

Complementary to a *Z*-contrast dark field STEM, EELS-STEM imaging has also been performed. The EELS spectra were collected point-by-point (<1 nm diameter electron beam spot with the pixels of 1 nm × 1 nm) throughout the entire image area. Electron energy loss at low energy is mainly dominated by the plasmon losses, sensitive to the local chemical composition. Fig. 4 shows EELS spectra collected in the region containing either Hf, or HfO_2, or TiN. Each spectrum can be fitted by a sum of Gaussian functions, whose peak position, width and height are characteristic of a specific material. Due to a higher scattering factor of the atoms with higher atomic number, like Hf, the signal intensity acquired in the regions containing pure metallic Hf is correspondingly much lower.