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Conductive filament structure in HfO_2 resistive switching memory devices

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ABSTRACT

We study the filament structure in 50 nm × 50 nm Resistive Random Access Memory (ReRAM) cells in the forming/set state with a Hf/HfO_2/TiN metal–insulator–metal stack by scanning transmission electron microscopy in cross section view. We reveal the filament morphology and, by the measurement of filament size and electrical resistance, evaluate the average resistivity of the filament material. The combination of the various data indicates the nanostructure of the conductive filament.

1. Introduction

As the current non-volatile FLASH memory technology is approaching the scaling limits, several different memory concepts have been proposed and have attracted extensive attention, such as phase change memories, Magneto-Resistive Random Access Memories (MRAM) and resistive switching memories (ReRAM). All these approaches are based on the possibility to change the resistance of a material by applying electric pulses and, in particular, ReRAM devices are considered to be very promising since they exhibit low power consumption, high switching speed, exceptional endurance and device fabrication simplicity, with potential for highest density arrays, as required for 3-D integrated circuit architecture [1–3]. A ReRAM device is based on a metal–insulator–metal (MIM) stack, in which a thin dielectric layer is sandwiched between two electrodes, and it is possible to form a conductive filament (CF) in the dielectric, connecting top and bottom metal electrodes. The filament formation may occur without involving the cation migration from an active electrode, but rather by local redox processes [4]. There are many materials showing this kind of resistive switching, such as TaO_2, HfO, NiO, CuO, TiO_2, and ZnO. In particular, Hf oxide is one the most promising materials, since it is already largely employed in CMOS technology.

The presence of a conductive filament inside a MIM largely modify its resistance. Formation of the filament is proposed to be caused by oxygen ion diffusion (induced by the applied electric field) out of a certain limited dielectric region and by subsequent field-temperature-driven migration of the released ions through the surrounding dielectric film [1,4,5]. Repeatable resistance switching is then described as involving as major limiting step the motion of these oxygen ions under an electric field bias back to the conductive filament, oxidizing a portion of the filament ('reset' to high resistance state, HRS), and their subsequent release and migration away from the filament ('set' to low resistance state, LRS) [4]. The physical dimensions of the filament and its physical structure throughout the hafnia dielectric is still subject of debate. Indeed, generally, the observation of a conductive filament (CF) in a real device is very difficult for several reasons. First, the CF is embedded between many surrounding layers, second, the filament size is expected to be only a few nanometers, third, the nucleation and growth of a conductive filament is a stochastic process, therefore the exact location of such a narrow filament is unpredictable. In the case of HfO_2, the observation is further complicated because Hf is a heavy element with a large number of electrons (atomic number $Z = 72$). This means that by employing, as an example, a powerful technique such as Transmission Electron Microscopy (TEM), the electron beam is strongly diffused and back-reflected, obtaining poor images in transmission. These difficulties have prevented the observation of the CF in real devices based on HfO_2. Also, for all these reasons, previous *in-situ* TEM observations were done on specially-designed structures, different from those used in fabricated memory devices, and with different dielectric layers, typically characterized by lower atomic number Z, such as NiO, ZnO or SiO_2 [6–8]. However, the information on the physical mechanisms determining the filament formation, as well as its

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composition and size are crucial for a better understanding of the memory operation and for the device optimization and stability. Moreover, since the filament size is expected to be of the order of a few nanometers, the correlation between the size and the electrical properties, which could be strongly modified due to the confinement, can give more insight on the scaling properties of these memories.

In this work we have investigated a crossbar memory with size $50 \times 50\ nm^2$ with HfO_2 dielectric and have employed the Scanning Transmission Electron Microscopy (STEM) to study the structure of the filaments in the low resistance state, after the forming process was completed. The measured filament size has been then correlated to the electrical resistance, obtaining insights on the filament resistivity, composition and nanostructure.

2. Experimental methods

The studied ReRAM cells are crossbar devices, 50 nm × 50 nm of size, with a TiN bottom electrode, a 5 nm HfO_2 dielectric, deposited by atomic layer deposition (ALD), and a metallic Hf top electrode with thickness 8 nm. On top of the metallic Hf, a thick TiN metal layer is deposited and patterned. The memory is integrated with a transistor, which allows to limit the current during the device operation. Filament forming is achieved by applying forward biased (with the Hf top layer set as positive electrode) current–voltage (*I–V*) sweeps under well-defined current compliance conditions, in the 100 μA–1 mA range. For the 1 mA current compliance case a HP4155 semiconductor parameter analyser was used, while for all the lower compliance levels the configuration with the transistor in series was employed.

To overcome the common problem in the TEM analyses of resistive switching devices, associated with difficulties to find the location of the filament within the entire volume of the memory cell, we have employed devices of a very small size, where the entire device volume is observable, allowing to collect data from the filament region. In such small area devices, to prepare cross-section specimens for TEM with the required accuracy, that is cross-sections of the entire device area of $50 \times 50\ nm^2$, we have used a technique based on the Focused Ion Beam (FIB) sample thinning. The TEM and STEM analyses were performed in a JEOL 2010 electron microscope with a 200 kV accelerating voltage. In the STEM mode under the analysis condition, the minimum probe size was estimated to be about 0.5 nm. The microscope is also equipped with electron energy loss spectroscopy (EELS) imaging.

To study the morphology of the formed conductive filaments, both STEM in dark field configuration and EELS at low energy have been employed. 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. The information on the CF size, shape and composition has been extracted by fitting EELS spectra in the low energy region. The filament size measured by dark-field STEM imaging in any filament section has been determined as the local full-width at half-maximum (FWHM) of the number of counts. The error/resolution of both the dark field and EELS STEM measurements, and therefore the minimum observable filament size, is of the order of 1 nm.

3. Results

Fig. 1(a) shows a TEM micrograph in bright field, acquired in a fresh device. The region of interest is very dark because of the thickness of the specimen and of the high electron beam diffusion and back-scattering. Fig. 1(b) shows the MIM device by STEM in dark field configuration, and at higher magnification. The image has been acquired using an annular detector, in order to enhance the mass contrast. The metallic Hf top electrode (brighter region) and the HfO_2 layer below are clearly distinguished, given the large difference in local charge density (Z-contrast). On top of Hf a greyish region is also visible, corresponding to an additional Hf oxide layer present at the edges of the device.

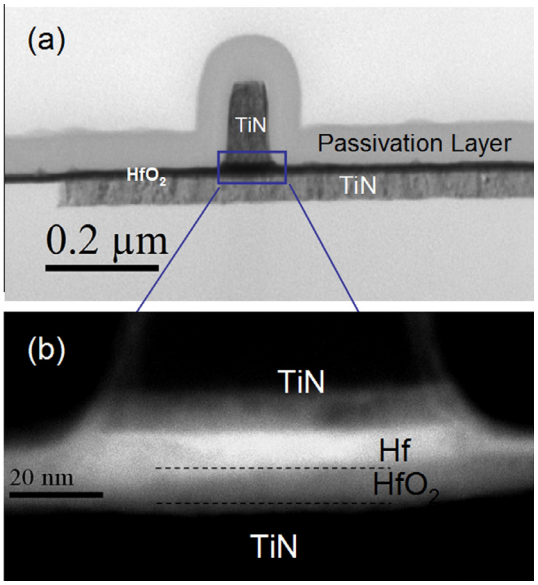

Fig. 1. (a) TEM bright field image of a crossbar device. (b) Details of the ReRAM device, acquired by STEM in dark field.

The pristine cell has a very high resistance, thermally activated with activation energy of about 0.1 eV [9]. The filament forming is achieved by applying *I–V* sweeps. Fig. 2(a) shows the *I–V* during forming, with a compliance of 500 μA. The filament is formed typically at +1.5–+2.5 V and after forming the resistance is of the order of a few kΩ. In such a condition, the devices show a metallic behavior, when conductance linearly decreases with temperature [4,9].

Switching to a high resistance state is obtained by applying a negative bias, as shown in Fig. 2(b). In the HRS, the conductance is low and increases with temperature, as typical of semiconductors.

In the LRS the filament conductance can be written as $N \times G_Q$, where G_Q is the quantum conductance [10]

$$G_Q = \frac{2e^2}{h} \tag{1}$$

equal to about 1/13 $k\Omega^{-1}$, and the factor N is about 2–10 in the LRS.

The conductance of the filament is strongly dependent on the forming current compliance, as shown in Fig. 3, where the conductance is plotted in units of G_Q. It is evident that, even in the LRS, the conductance is at the level of few G_Q units. This datum points to a physical size of the involved filament. In fact, according to Landauer [11], for a 1D metallic wire subjected to a small voltage drop V, the flowing current I is:

$$I/V = G = N \times G_Q \tag{2}$$

where N is the number of 1D sub-bands involved in the charge transport. For $N = 1$, only the first sub-band is involved and this suggests that the filament cross-section diameter, by assuming a simple cylindrical geometry and symmetry, is equal to $\lambda_F/2$, where λ_F is the conduction electron wavelength. Since in a metal λ_F is of the order of 1 nm [12], one should expect that the size of the metallic wire in the ReRAM cell is of that magnitude order, i.e., ≈1 nm. In similar stacks, the filaments described as Hf-rich in HfO_x-based