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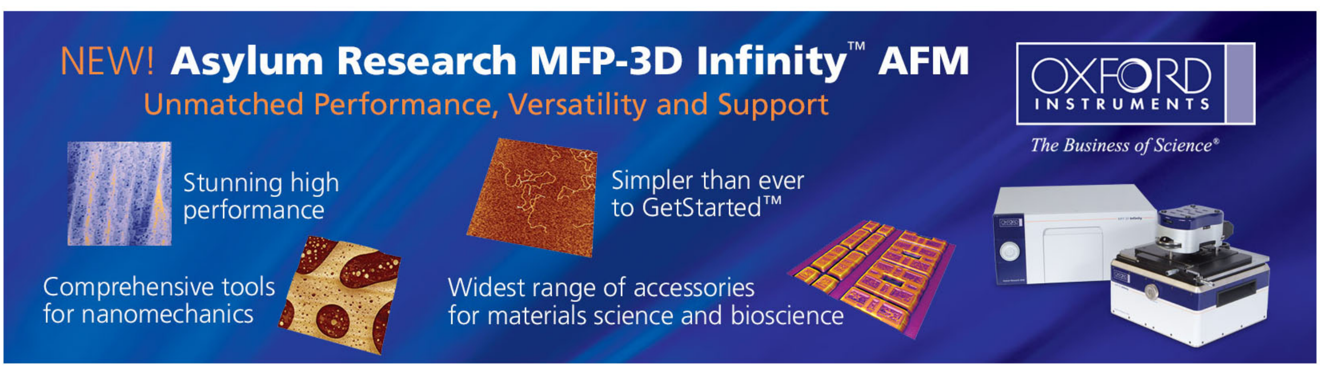

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APPLIED PHYSICS LETTERS **102**, 203507 (2013)

The effect of high/low permittivity in bilayer HfO_2/BN resistance random access memory

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This letter investigated the electrical characteristics of resistance random access memory (RRAM) with HfO_2/BN bilayer structures. By adopting the high/low permittivity structure, we obtained the excellent device characteristics such as uniform distribution of switching voltage and more stable resistance switching properties of RRAM. The current conduction mechanism of low resistance state in the HfO_2/BN device was transferred to space-charge-limited current conduction from Ohmic conduction owing to space electric effect concentrated by the high/low permittivity bilayer structures. The electric field in the bilayer can be verified by COMSOL simulation software. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4807577]

With intensive demands for communication in digital age, consumer electronic products are broadly integrated with display,[1–3] memory circuits, and logic circuits. Next-generation nonvolatile memory (NVM) is needed to develop for powerful portable electronic products, as conventional charge storage-based memories[4–6] suffered the technical and physical limitation issues. Resistance random access memory (RRAM) is one of promising candidates for next generation NVMs, due to its simple cell structure, scalability, high operating speed, and non-destructive read out.[7–13]

Various materials have been reported owning resistive switching behaviors. And HfO_2 is one of the most thoroughly investigated materials in RRAM area. However, switching uniformity and operating stability has always been the problem to be solved in order to achieve satisfactory performance.[14,15] In our previous research, single layer RRAM devices of different materials have been investigated. Even though some merits are gained by tuning the material type and the thickness of active layer or modified by $SCCO_2$ restoration,[16] switching uniformity is still one problem.[17,18] Previously, we have applied stacking layer technology by switching sputtering gas ambient to achieve $SiGeO_x$/SiGeON structure,[19] but the result is not very satisfactory as well. In this work, the HfO_2/BN bilayer high/low permittivity structure was proposed to improve the uniformity of device parameters, which was inspired by the electric field concentrating ability of low permittivity material. The electric field simulation by COMSOL was applied to confirm the electrical field distribution. And conduction mechanism analysis was conducted to explain the reason why the resistive switching behaviors were improved.

After the standard cleaning of the TiN/Ti/SiO_2/Si substrate, 6-nm-thick BN film combined with 12-nm-thick HfO_2 film is deposited on the TiN/Ti/SiO_2/Si substrate by RF magnetron sputtering sequentially. Finally, Pt top electrode with a thickness of 200 nm was deposited on HfO_2 film to form Pt/HfO_2/BN/TiN structure RRAM devices. Besides the devices with Pt/HfO_2/TiN sandwich structure, using same HfO_2 and Pt sputtering conditions, were made as control samples. The entire electrical measurements of devices with the Pt electrode of 4 μm diameter were performed using Agilent B1500 semiconductor parameter analyzer.

Figure 1(a) shows the resistive switching properties of the memory devices with single HfO_2 layer and HfO_2/BN bilayer structures, respectively. The repeatable resistive switching behaviors between high-resistance state (HRS) and low-resistance state (LRS) were obtained for both structures after the forming process. The voltage sweep bias was applied on TiN electrode with the grounded Pt electrode as shown in Fig. 1(b). The distributions of the switch voltage and resistance states were counted with continuous I–V sweep measurement of 100 cycles, as shown in Figs. 1(c) and 1(d). The set voltage distribution of HfO_2/BN device concentrates from 0.8 V to 1 V, which is more concentrative than that of HfO_2 device (Fig. 1(c)). This improvement can be also observed with the comparison of the results in references.[14,15] And it is quite normal for single metal oxide layer RRAM device working unstable, owing to the drastic formation and rupture of conduction filament.[20] From Fig. 1

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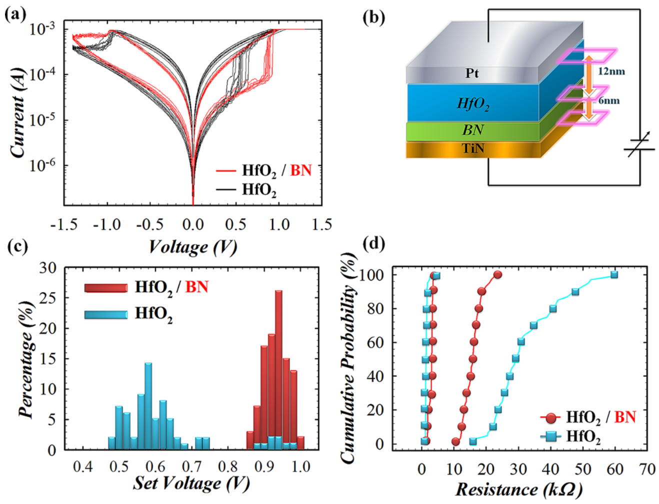

FIG. 1. (a) Comparison of resistive switching characteristics between single HfO_2 layer and HfO_2/BN bilayer in dc voltage switching. (b) Schematic structure of HfO_2/BN bilayer device. Distributions of (c) the switch voltage during 100 resistance switching cycles and (d) the resistance states of HRS and LRS between single HfO_2 layer and HfO_2/BN bilayer.

we can find that resistance state distribution shows the stable resistive switching behaviors that can be obtained in HfO_2/BN devices as compared to that in HfO_2 devices. This indicates that the excellent device characteristics such as uniform distribution of switching voltage and more stable resistive switching properties of RRAM can be achieved by using the high/low permittivity stacking structure.

Fourier transform infrared (FTIR) spectroscopy was used to investigate the chemical bonding of the HfO_2 and BN films in this study. Figure 2(a) shows the Hf-O polycrystalline bonding was found in the HfO_2 film at 770 cm^{-1} from the FTIR spectrum. The monoclinic phases of Hf-O bonds were also discovered at 594 cm^{-1} and 512 cm^{-1}.[21] Additionally the spectrum of FTIR in Fig. 2(b) indicates the B-O-B stretch mode bonding around the absorption peak at 1186 cm^{-1}. The peaks at 1430 cm^{-1} and 3206 cm^{-1} belong to the hexagonal B-N and N-H stretching bonds, respectively.[22] According to the FTIR spectrums, high/low permittivity stacking structure constructed with HfO_2/BN layer is confirmed.

To investigate the interesting phenomena, we analyzed the current conduction mechanisms of the bilayer HfO_2/BN

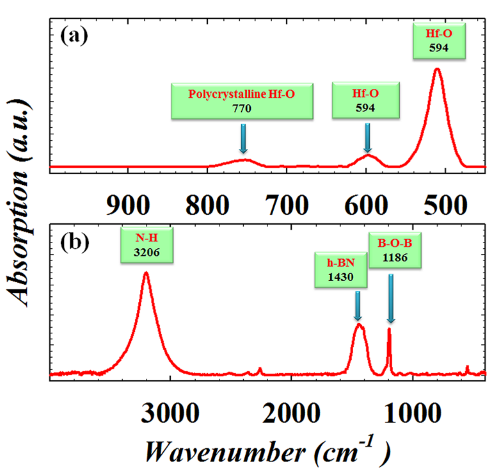

FIG. 2. FTIR spectra of (a) HfO_2 film and (b) BN film measured in the middle infrared region.

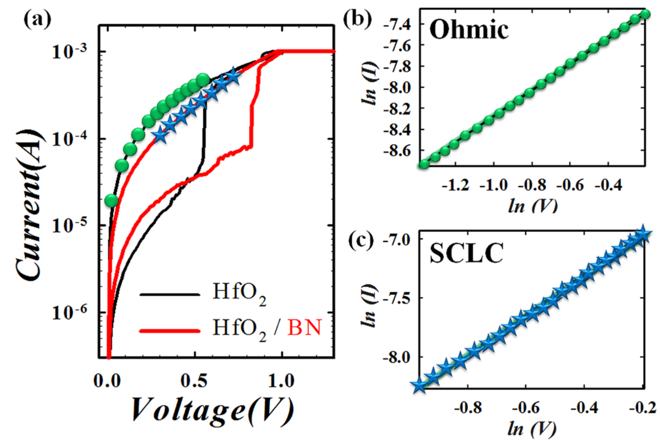

FIG. 3. (a) Comparison of electrical characteristics of memory devices with typical I-V curves. (b) A plot of ln(I) vs ln(V) in LRS of HfO_2 device. (c) A plot of ln(I) vs ln(V) in LRS of HfO_2/BN device.

and the single HfO_2 devices as shown in Fig. 3(a). The I-V curve fitting exhibits that the current conduction in the LRS of HfO_2 device is dominated by the Ohmic conduction (Fig. 3(b)). However the current conduction in the LRS of bilayer HfO_2/BN device is complied with space-charge-limited current (SCLC) conduction mechanism (Fig. 3(c)). Based on the material and electrical analysis, the continuous filament is formed after the forming process in single layer device, leading to the conductive filament connects between two electrodes. Therefore, the electrical current is dominated by Ohmic conduction in LRS of the HfO_2 device. On contrast, the conductive filament formed in HfO_2/BN bilayer, especially in the BN layer, is not thick as that in single layer device. The concentrated electric field leads to the restriction of conduction carriers, owing to the inserted BN layer with relative low permittivity between HfO_2 and TiN electrode. And that is also the reason why we can obtain SCLC conduction mechanism, as limited cross section of conduction filament confines the carrier conduction path.

To evaluate the effect of concentrated electric field in the bilayer HfO_2/BN on resistive switching characteristics of RRAM device in this work, COMSOL simulation was employed. Figure 4 shows the conduction model of LRS and the distribution of electric field in HRS of the HfO_2 and HfO_2/BN devices, respectively. A more concentrated electric

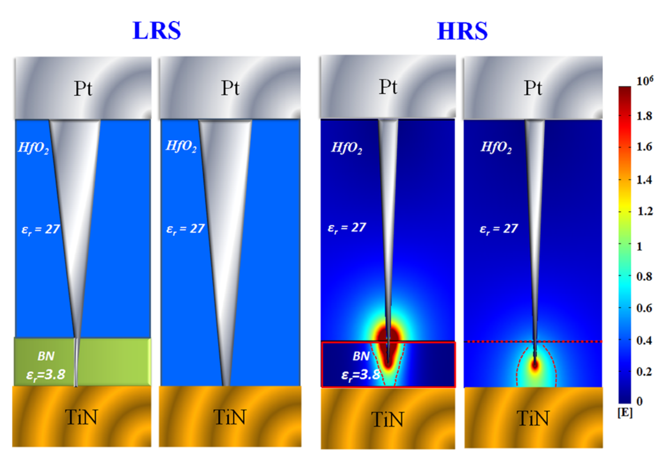

FIG. 4. Electric field simulation in HRS and the schematic model in LRS for Pt/HfO_2/TiN and Pt/HfO_2/BN/TiN memory devices.

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field occurs obviously at the tip of the conductive filament in bilayer HfO_2/BN device. Consequently, a fine and slim conductive filament will form due to the space electric field concentration in the high/low permittivity bilayer structure, leading to the SCLC current conduction in LRS. Owing to the restriction of conduction path, the resistive switching uniformity of the HfO_2/BN devices is improved compared with single HfO_2 layer devices, whose carrier conduction is highly influenced by drastic filament formation and rupture, while the stochastic process can be alleviated by inserting the low permittivity BN layer.[14,15,20]

In conclusion, the high/low permittivity effect of RRAM with bilayer HfO_2/BN structure was investigated for NVM applications. This device exhibits good reproducibility and switching uniformity. According to the analysis of current fitting and COMSOL simulation, the current conduction in LRS is dominated by SCLC conduction due to the electric field concentration in the high/low permittivity bilayer stacking structure. The results implicates that the resistive switching performance of RRAM device can be improved by adopting the high/low permittivity structure for next generation NVM applications.

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