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Study of Multilevel High Resistance States in HfO_x-Based Resistive Switching Random Access Memory by Impedance Spectroscopy

H. K. Li, T. P. Chen, S. G. Hu, P. Liu, Y. Liu, P. S. Lee, X. P. Wang, H. Y. Li, and G. Q. Lo

Abstract—Multilevel high resistance states are achieved in TiN/HfO_x/Pt resistive switching random access memory device by controlling the reset stop voltage. Impedance spectroscopy is used to study the multilevel high resistance states. It is shown that the high resistance states can be described with an equivalent circuit consisting of the major components R_s , R , and C corresponding to the series resistance of the TiON interfacial layer, the equivalent parallel resistance and capacitance of the leakage gap between the TiON layer and the residual conductive filament, respectively. These components show a strong dependence on the stop voltage, which can be explained in the framework of oxygen vacancy model and conductive filament concept. On the other hand, R is observed to decrease with DC bias, which can be attributed to the barrier lowering effect of the Coulombic trap well in the Poole-Frenkel emission model.

Index Terms—Impedance spectroscopy, multilevel high resistance states, resistive switching random access memory (RRAM).

I. INTRODUCTION

Resistive switching random access memory (RRAM) based on metal oxides like Al₂O₃, ZnO, NiO_x, TiO₂, and Cu_xO is promising in the application of next-generation non-volatile memory due to its simple structure, low power consumption,

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high read/write speed, and good reliability [1]-[7]. Recently, multilevel resistance states in RRAM devices have been demonstrated to increase the storage density of the RRAM device [8], [9]. Until now, most of the research works were focused on the performance improvement and reliability of the multibit storage, there are relatively few studies on the mechanism for the multilevel resistance states of RRAM device. In this work, impedance spectroscopy is employed to investigate multilevel high resistance states in TiN/HfO_x/Pt RRAM structure. Impedance spectroscopy is a powerful tool to examine the conduction properties of dielectric thin films, making it suitable for resistive switching property study [10], [11]. For the TiN/HfO_x/Pt RRAM structure used in this work, the multilevel high resistance states may be attributed to the different rupture degrees of the conductive filament, which is hard to be detected by the microscopic techniques such as transmission electron microscopy and scanning probe microscopy. However, through the analysis of the impedance measurement, we have been able to obtain information about the redox reaction relating to the change of TiON interfacial layer and rupture of the conductive filament for different high resistance states.

II. EXPERIMENT

The TiN/HfO_x/Pt RRAM structure was fabricated with the following sequence. Firstly, a 500 nm SiO₂ film was deposited on an 8-inch *p*-type Si wafer by plasma-enhanced chemical vapor deposition to prevent the leakage during the switching operation of the RRAM device. Then, 50 nm Pt/ 20 nm Ti layer was deposited by electron-beam evaporation to form the bottom electrode on the SiO₂ film. Subsequently, a 10 nm HfO_x layer was deposited by atomic layer deposition on the bottom electrode. Finally, a 100 nm TiN layer was deposited onto the HfO_x layer using DC sputtering and patterned to form the top electrode with the area of about 4 μm². A Keithley 4200 semiconductor characterization system was used to characterize the switching properties of the device. Impedance measurement of different high resistance states was carried out with an Agilent E4980A Precision LCR Meter in the frequency range of 20 Hz to 2 MHz with a 30 mV AC signal.

III. RESULTS AND DISCUSSIONS

Fig. 1 shows the typical bipolar resistive switching behavior

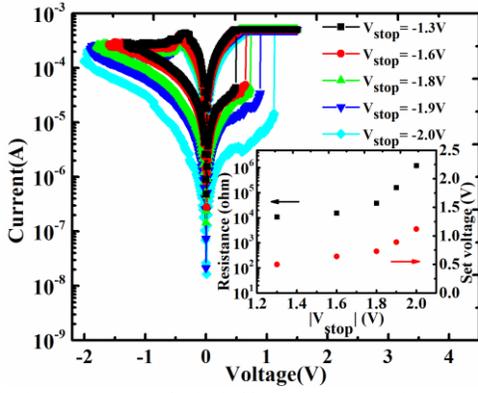


Fig. 1. Bipolar I - V curves of TiN/HfO_x/Pt RRAM device. Multilevel high resistance states can be achieved with different V_{stop} . The inset shows the values of the high resistance states created with different V_{stop} (read at 0.1 V) and the corresponding V_{set} .

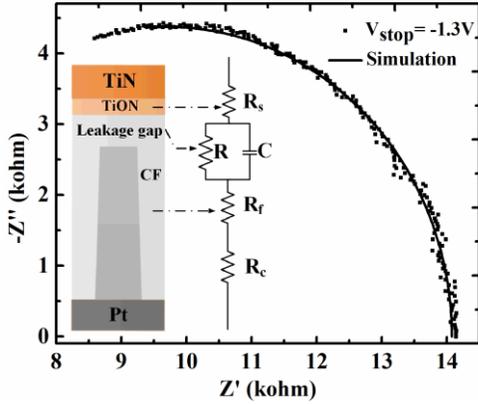


Fig. 2. Complex impedance spectra of the high resistance state obtained with the V_{stop} of -1.3V. The curve is the best fitting based on (2). The inset shows the schematic illustration of the conductive filament and the equivalent circuit for high resistance state.

of the TiN/HfO_x/Pt RRAM structure under different reset stop voltages (V_{stop}) ranging from -1.3 V to -2.0 V. Multilevel high resistance states can be achieved by varying V_{stop} . The inset of Fig. 1 shows the values of the high resistance states created with different V_{stop} (read at 0.1 V) and the corresponding set voltages (V_{set}). As observed in the inset of Fig. 1, the resistance of the device increases with $|V_{stop}|$. Based on the conductive filament model, when a positive voltage is applied to the TiN top electrode, the oxygen atoms inside the HfO_x layer will be knocked out of the lattice and become oxygen ions to react with the TiN electrode to form TiON [12]. The formation of TiON could result from the replacement of nitrogen by oxygen or defects that favor the incorporation of oxygen in the cation sub-lattice [13]. When the generated oxygen vacancies form a strong enough conductive filament to connect the top and bottom electrodes, the device will be set to a low resistance state. For the reset process, a negative voltage is applied to the top electrode, and the oxygen ions will move back to eliminate the oxygen vacancies, breaking the conductive filament; as a consequence, a leakage gap in the oxide is formed between the top electrode and the residual filament, as schematically illustrated in the inset of Fig. 2. When V_{stop} is of a larger magnitude, more oxygen ions will move back to the HfO_x layer to eliminate the oxygen vacancies, which will widen the gap

between the top electrode and the residual conductive filament, and thus the resistance value will increase due to the gap widening [14]. In this case, the V_{set} during the following set process also increases with the magnitude of V_{stop} , as demonstrated in the inset of Fig. 1. It is because that a larger V_{set} is needed to rebuild the conductive filament for a wider leakage gap.

The above arguments could be verified with the impedance measurement. The complex impedance measurement is conducted after the reset process by applying a 30 mV small AC signal in the frequency range of 20 Hz to 2 MHz without DC bias. The scatter plot in Fig. 2 shows the complex impedance spectra of the high resistance state after reset operation with the V_{stop} of -1.3 V. The approximately semicircle shape of the Nyquist plots ($-Z''$ VS Z') indicates that the high resistance state can be modelled as a parallel connection of a resistor and a capacitor in series with some resistors, as shown in the inset of Fig. 2. The RRAM devices with other sizes ($5 \times 5 \mu\text{m}^2$ and $8 \times 8 \mu\text{m}^2$) are also tested, and similar semicircle shapes of the Nyquist plots are observed (not shown here). This indicates that device size in the range has no influence on the equivalent circuit. The impedance of the equivalent circuit can be described with

$$Z = R_s + R_f + R_c + \frac{R}{1 + \omega^2 R^2 C^2} - j \frac{\omega R^2 C}{1 + \omega^2 R^2 C^2} \quad (1)$$

where Z is the complex impedance, ω is the angular frequency, R_s , R_f and R_c are the equivalent series resistance for the TiON interfacial layer, residual conductive filament and measurement connections, respectively, and R and C are the equivalent parallel resistance and capacitance of the leakage gap, respectively. As discussed above, the redox reaction between the TiN electrode and the oxygen ions during the set process results in a TiON interfacial layer, which is more resistive than the pure TiN electrode. Though the series resistance should be the sum of the resistance of the TiON interfacial layer, residual filaments and measurement connections, the last two items can be neglected because of their much lower values [15]. Therefore, (1) can be rewritten as

$$Z = Z' + jZ'' = R_s + \frac{R}{1 + \omega^2 R^2 C^2} + j \left(-\frac{\omega R^2 C}{1 + \omega^2 R^2 C^2} \right) \quad (2)$$

where Z' and Z'' are the real part and imaginary part of the complex impedance, respectively. The fitting with (2) to the measurement data is shown in Fig. 2. An excellent agreement between the fitting and measurement is achieved, which indicates that the equivalent circuit shown in the inset of Fig. 2 can well describe the electrical characteristic of the RRAM structure. The fitting yields the values of 5336 Ω , 8741 Ω and 9.81 pF for R_s , R and C , respectively. With the obtained R_s value, one may estimate the resistivity ρ of the TiON interfacial layer with $\rho = (A/t_{TiON})R_s$, where t_{TiON} is the thickness of the interfacial layer, and the device area $A = 4 \mu\text{m}^2$. Under the assumption that t_{TiON} is 5 nm [16], [17], the resistivity is estimated to be $\sim 400 \Omega \cdot \text{cm}$, which is within the reported resistivity range of TiON films [18].

To examine the behavior of different high resistance states, we conducted the complex impedance measurement after each

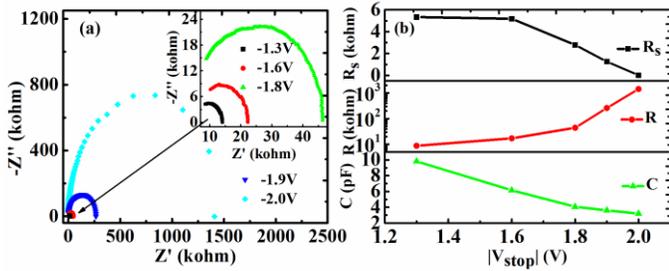


Fig. 3. (a) Complex impedance spectra of different high resistance states obtained with different $|V_{stop}|$. The inset in (a) shows the enlarged complex impedance spectra with the V_{stop} of -1.3 V, -1.6 V and -1.8 V. (b) The values of R_s , R and C as a function of $|V_{stop}|$.

reset process at different V_{stop} , and the result is shown in Fig. 3(a). The Nyquist plots of all V_{stop} can be well fitted with (2), which indicates that all the high resistance states can be modelled with the equivalent circuit shown in the inset of Fig. 2. The values of the components in the equivalent circuit are shown in Fig. 3(b). The R_s value has an obvious decreasing trend when the V_{stop} changes from -1.3 V to -2.0 V. Since TiON is more resistive than pure TiN, the decrease of the R_s means more TiON is reduced to TiN for a larger $|V_{stop}|$. The parallel RC element represents the leakage gap between the top electrode and the residual conductive filament, and the modulation of the gap width is responsible for the changes in the R and C values. As shown in Fig. 3(b), the R value increases with the increase of $|V_{stop}|$, which means the width of the leakage gap gradually increases. It is reasonable to argue that the recombination of the oxygen vacancies with the oxygen ions supplied from the TiON layer or even the surrounding HfO_x layer is enhanced as the V_{stop} changes from -1.3 V to -2.0 V. It is worth noting that R_s and R have an opposite evolution tendency, while the change of the DC resistance, shown in the inset of Fig. 1, is consistent with the evolution of R because the R value is much larger than the R_s value. The capacitance of the RC element should be inversely proportional to the width of the leakage gap between the top electrode and the residual filament, being similar to the situation of a parallel plate capacitor; thus the decrease in the C value with $|V_{stop}|$ suggests that a larger $|V_{stop}|$ results in a wider leakage gap. Therefore, both dependence of R and C on $|V_{stop}|$ suggest that the leakage gap width increases with $|V_{stop}|$, which explains the increase of the DC resistance with $|V_{stop}|$ as shown in the inset of Fig. 1.

C can be described with $C = \epsilon_r \epsilon_0 A / t_{ox}$, where ϵ_0 is the permittivity in vacuum, and ϵ_r and t_{ox} are the dielectric constant and thickness of the leakage gap, respectively; and R can be assumed to be $R = R_0 \exp(\alpha t_{ox})$, where R_0 and α are two constants as it has been suggested by Monte Carlo simulation that R has an exponential dependence on t_{ox} [19], [20]. Under the above assumptions, one may find the simple relationship between R and C as follows

$$\ln R = \ln R_0 + \frac{\epsilon_o \epsilon_r \alpha A}{C} \quad (3)$$

The linear relationship between $\ln(R)$ and $1/C$ predicted by (3) roughly agrees with the experimental result, as shown in Fig. 4.

To examine the influence of DC bias on the complex impedance measurement, a positive voltage ranging from 0 V

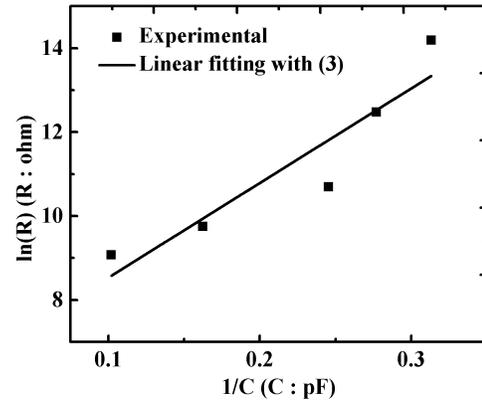


Fig. 4. $\ln(R)$ versus $1/C$.

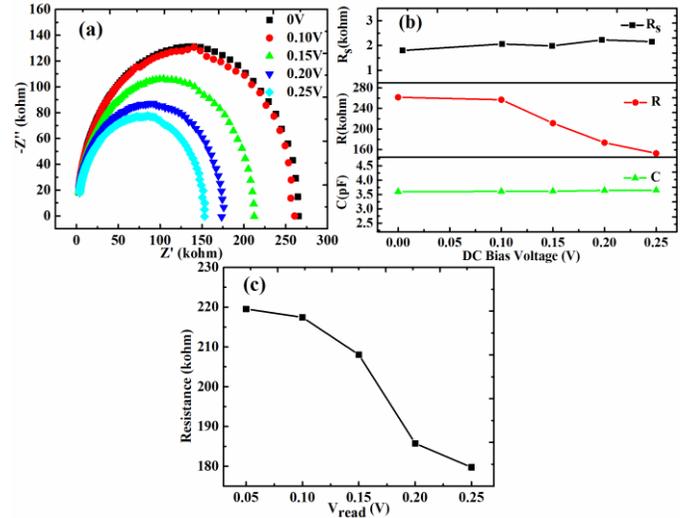


Fig. 5. (a) Complex impedance spectra of the high resistance state under different positive DC biases. (b) The values of R_s , R and C as a function of the DC bias. (c) The resistance value of the high resistance state as a function of V_{read} .

to 0.25 V was superimposed to the AC signal during the impedance measurement, and the complex impedance spectra of the high resistance state under different positive DC biases are shown in Fig. 5(a). The semicircle shape of all the Nyquist plots indicates that the DC bias has little influence on the equivalent circuit of the high resistance state. The values of R_s , R and C obtained from the fitting as a function of the DC bias are shown in Fig. 5(b). Only the R has an obvious decreasing trend with the increase of DC bias. The little change of both R_s and C indicates that the redox reaction at the TiN/ HfO_x interface is not significant under the influence of a small positive DC bias and the leakage gap should change little or remain unchanged. Therefore the change of the R value cannot origin from the leakage gap modulation effect. Previous studies on the current transport in HfO_x -based RRAM device show that Poole-Frenkel emission model can well describe the conduction of the high resistance state; and the oxygen vacancies inside the HfO_x layer could act as the Coulombic traps in the Poole-Frenkel emission model [21], [22]. When a bias is applied to the switching layer, one side of the barrier height of the Coulombic traps will be reduced, the probability for the electrons escaping from the trap well by thermal

emission increases, thus the R value will decrease with increasing DC bias [23]. The decrease of R with DC bias is indeed observed in Fig. 5(b). This is also consistent with the experimental result shown in Fig. 5(c) that the DC resistance of the high resistance state decreases with the read voltage (V_{read}).

IV. CONCLUSION

In conclusion, impedance spectroscopy is a useful technique to study multilevel high resistance states in HfO_x -based RRAM device. The analysis of complex impedance suggests that the redox reaction at the TiN/HfO_x interface and the modulation of the leakage gap should be responsible for the changes in the parameters of the equivalent circuit of the RRAM device. The leakage gap widening effect is shown to be the main reason for the higher resistance value associated with a larger $|V_{stop}|$. Both R_s and C show little change with DC bias; however, R decreases with the DC bias, which can be attributed to the barrier lowering effect of the Coulombic trap well in the Poole-Frenkel emission model.

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