

Conductance quantization in oxide-based resistive switching devices

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Abstract

Oxide-based resistive switching devices have tremendous potential in next-generation nonvolatile memory and neuromorphic applications. Here, the emergence of quantized conductance is investigated in resistive switching devices based on Ta₂O₅ or HfO₂. By applying sweeping voltages with different current compliances or using consecutive voltage pulses, quantized conductance states including integer and half integer multiples of quantum conductance (G_0) were observed, suggesting well-controlled formation of atomic point contacts. Compared with Pt/Ta/Ta₂O₅/Pt devices, a larger number of quantized conductance states were obtained in the Pt/Ta/HfO₂/Pt devices. Such quantized conductance states are inherently discrete and multilevel, which could be promising for applications as multilevel nonvolatile memory and artificial synapses in hardware neural networks.

Keywords—resistive switching; quantized conductance; multilevel memory; neuromorphic computing.

Introduction

Resistive switching devices or memristors have been extensively studied for next-generation nonvolatile memory or neuromorphic computing applications^[1-4], due to their simple structure, excellent scaling potential, high endurance, high switching speed, low energy consumption and so on^[5,6]. In order to achieve high capacity of data storage and emulate the variable synaptic weights of biological synapses during learning, an analog conductance state will be highly desirable. It has been well understood that the working mechanism of memristors can be attributed to the formation and rupture of nanoscale conducting filaments in the switching layer, as verified by direct *in-situ* observations^[7,8]. When the size of the filament is reduced to extreme cases, for example the thickness of the filament reaches atomic scale, quantum effects will emerge and the device conductance can become quantized^[9,10]. Such quantized conductance states are inherently discrete and multilevel and could thus be promising for memory and neuromorphic applications.

In this work, we report the observation of conductance quantization in technologically relevant Ta₂O₅ or HfO₂ based resistive switching devices. The different quantized conductance states can be reliably obtained and well controlled using voltage sweeps or voltage pulses, which can be ascribed to the gradual formation and rupture of oxygen vacancy based conducting filaments. Exploiting the discrete distribution of device conductance concentrated around fixed quantized values could help mitigate device variations and facilitate the programming or training processes in future

memory/neuromorphic applications.

Experimental

All devices studied in this work were fabricated on SiO₂/Si substrates. First, 5 nm thick Ti and 30 nm thick Pt films were deposited as the adhesion layer and the bottom electrode, respectively. A 10 nm thick dielectric layer of HfO₂ or Ta₂O₅ was then deposited by magnetron sputtering, serving as the switching layer. Finally, 10 nm thick Ta was deposited as the top electrode and capped by 30 nm thick Pt protection layer, where the patterning of the top electrodes was done by photo lithography and lift-off processes. A schematic configuration of the cell structure and its measurement setup are depicted in Fig. 1(a). All the direct-current (DC) current-voltage (I - V) characteristics and pulse measurement results were collected using a semiconductor parameter analyzer (Agilent B1500A) at room temperature. The bottom electrode was always grounded while the voltage was applied to the top electrode throughout the measurements. All the electrical results were obtained from devices with a size of 100×100 μm^2 .

Results and discussion

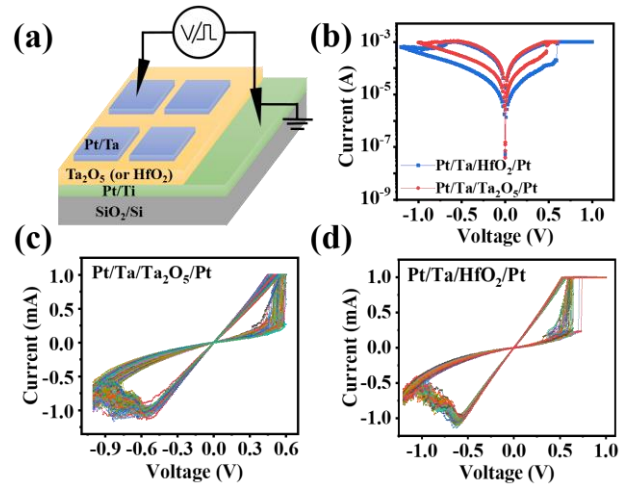


Fig. 1 (a) Schematic illustration of the device structure. (b) Log-scale I - V curves of Pt/Ta/Ta₂O₅/Pt and Pt/Ta/HfO₂/Pt devices. (c) Linear-scale I - V curves in 100 cycles from Pt/Ta/Ta₂O₅/Pt devices. (d) Linear-scale I - V curves in 100 cycles from Pt/Ta/HfO₂/Pt devices.

Figure 1(b) shows the I - V curves of the Pt/Ta/Ta₂O₅/Pt and Pt/Ta/HfO₂/Pt devices, both showing bipolar switching characteristics. The measurements were repeated for 100 cycles to examine the reliability of resistive switching, as shown in Figs. 1(c) and 1(d). The switching effects are highly reliable for both device structures, and it can be found that V_{set} and V_{reset} of the Pt/Ta/Ta₂O₅/Pt device always stay lower than

that of the Pt/Ta/HfO₂/Pt device.

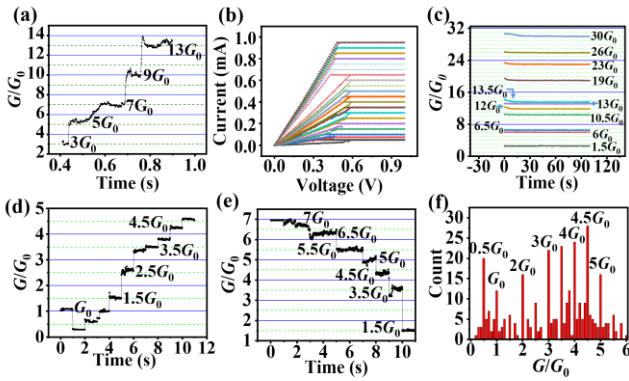


Fig. 2 (a) Conductance quantization observed during set process in Pt/Ta/Ta₂O₅/Pt devices with a low sweeping speed of 1 mV/s. (b) I-V characteristics with different current compliance from 100 μ A to 1 mA. (c) Retention behavior of the quantized conductance states in Pt/Ta/Ta₂O₅/Pt devices at room temperature (d) The evolution of device conductance in Pt/Ta/Ta₂O₅/Pt devices under a series of positive voltage pulses with an amplitude of 1 V and a width of 1 s. (e) The evolution of device conductance in Pt/Ta/Ta₂O₅/Pt devices under a series of negative voltage pulses with an amplitude of -1 V and a width of 1 s. (f) Histogram of the conductance values extracted from the measured data.

Interestingly, when the sweeping rate was reduced to 1 mV/s, discrete conductance levels can be obtained during the set process, as shown in Fig. 2(a). The device conductance G of Pt/Ta/Ta₂O₅/Pt devices was calculated as $G = I/V$ and the results showed that all the accessible conductance states are integer multiples of quantum conductance G_0 , where I is the measured current, V is the applied voltage, and $G_0 = 2e^2/h$ with e as the electron charge and h as Planck's constant. It thus indicates formation of atomic contacts in the Pt/Ta/Ta₂O₅/Pt device^[9-11], which is likely between the oxygen vacancies based filament and the electrode. Such inherently quantized conductance can be utilized to achieve multilevel data storage or high-precision synaptic weight. Indeed, by varying the current compliance from 100 μ A to 1 mA during the set process, different quantized conductance states in the Pt/Ta/Ta₂O₅/Pt cell can be successfully obtained, as demonstrated in Fig. 2(b). Such quantized conductance states are nonvolatile and stable, as verified by the retention results in Fig. 2(c) where the states were probed using 0.1 V read voltage.

Aside from the DC measurements, such quantized conductance states can also be reliably obtained using pulse measurements. A read voltage of 0.1 V was again adopted to monitor the device conductance. Fig. 2(d) shows the evolution of device conductance under a series of positive voltage pulses with an amplitude of 1 V and a width of 1 s. Initially, the conductance was about G_0 , and after an initial dip the conductance constantly increased and temporarily stabilized at different plateaus. Most of the plateaus exhibit conductance levels that are integers or half integers of G_0 . Fig. 2(e) further shows conductance quantization under a series of negative voltage pulses with an amplitude of -1 V and a width of 1 s. The conductance decreases from $7G_0$ to $1.5G_0$, and one can easily find that most of the stable conductance plateaus are once again located at integer or half integer multiples of G_0 , in good agreement with Figs. 2(a) and 2(d). In general, more quantized conductance states can be obtained in the reset

process compared with the set process, which is also consistent with the better analogy behavior in reset as can be seen in Fig. 1. A statistical analysis on the device conductance was further performed on the experimental data obtained from the Pt/Ta/Ta₂O₅/Pt devices, as shown in Fig. 2(f). By analyzing about 50 switching cycles, one can see that conductance peaks are distributed at both integer and half integer multiples of G_0 , with small scatterings around the peaks, once again confirming discrete quantized conductance states in the switching process.

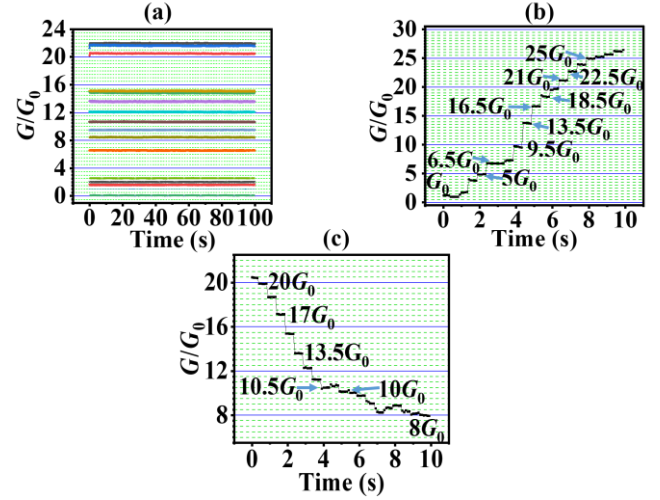


Fig. 3 (a) Retention behavior of the quantized conductance state in Pt/Ta/HfO₂/Pt device at room temperature. (b) The evolution of device conductance in Pt/Ta/HfO₂/Pt devices under a series of positive voltage pulses with an amplitude of 1 V and a width of 1 s. (c) The evolution of device conductance in Pt/Ta/HfO₂/Pt devices under a series of positive voltage pulses with an amplitude of -1 V and a width of 1 s.

Besides, the abovementioned conductance quantization behavior was also observed in HfO₂-based resistive switching devices. As depicted in Fig. 3(a), quantized conductance states can once again be obtained during DC sweeps when different current compliances were applied, which are nonvolatile in nature. Similarly, the discrete conductance states can also be accessed using pulse measurements in both set (Fig. 3(b)) and reset (Fig. 3(c)) processes in Pt/Ta/HfO₂/Pt devices. Compared with Pt/Ta/Ta₂O₅/Pt devices, it seems that a larger number of quantized conductance states can be obtained in the Pt/Ta/HfO₂/Pt devices, making them more promising for multilevel memory and synaptic applications.

The above integer and half integer multiples of G_0 conductance state in oxide based memristive devices can be interpreted by the migration of oxygen vacancies and resultant formation/dissolution of atomic scale conducting filaments^[12], as schematically illustrated in Fig. 4. In Ta₂O₅ and HfO₂ based memory devices, a conducting filament is not formed in high resistance state and thus the initial conductance state is less than G_0 , as illustrated in Fig. 4. When DC or pulsed stimulations are applied to set the device, a conducting filament will be gradually formed in the cell. At a certain stage, a single atomic point contact between the filament and the counter electrode can be established. Since the overall device conductance is dominated by the thinnest part of the filament, such filament will contribute a conductance of G_0 . When the current compliance was increased or more voltage pulses are applied, the filament will continue to grow, leading to the

formation of multiple atomic point contacts at the electrode/oxide interface. This in turn leads to the occurrence of $2G_0$, $3G_0$, $4G_0$, and so on, as depicted in Fig. 4. The half integral multiples of G_0 , however, may be attributed to adsorbed impurities to the conducting filaments that may change the overall constriction configuration^[9]. Also, it is interesting to note that there are missing conductance values in the devices. This can be understood considering the fact that there may be more than one nano-filament formation in parallel^[13]. Since the quantized conductance can be reliably obtained by controlling sweeping voltages with different current compliances or using consecutive voltage pulses, the oxide-based resistive switching devices are capable of multi-bit storage in one cell or achieving tunable weight states that are analogous to biological synapses during learning.

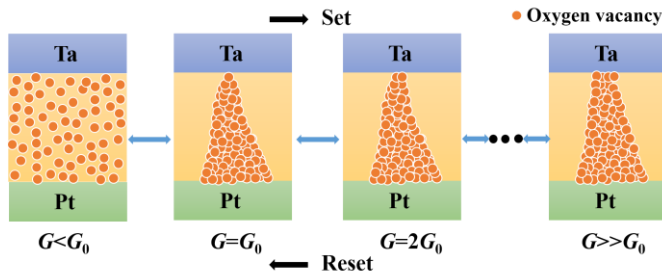


Fig. 4 Schematic illustration of the conductance quantization effect in Ta_2O_5 or HfO_2 based resistive switching devices.

Conclusion

In summary, we reported the observation of quantized conductance in Ta_2O_5 and HfO_2 based resistive switching devices. By applying different current compliances and pulse voltages, the multiple quantized conductance states can be observed, which can be attributed to the atomic point contacts of oxygen-vacancies-composed conducting filament in the Ta_2O_5 and HfO_2 films. The quantized conductance resulting in well separated resistance states would offer the opportunity to achieve multilevel data storage and artificial synapses using oxide-based resistive switching devices.

Acknowledgements

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Abstract

Oxide-based resistive switching devices have tremendous potential in next-generation nonvolatile memory and neuromorphic computing applications. Here, the emergence of quantized conductance is investigated in resistive switching devices based on Ta₂O₅ and HfO₂ by applying sweeping voltages with different current compliances or using consecutive voltage pulses, quantized conductance states were observed, including integer and half integer multiples of quantum conductance (G_0), suggesting well-controlled atomic point contacts. Compared with Pt/TaTa₂O₅/Pt devices, a larger number of quantized conductance states were observed in Pt/TaHfO₂/Pt devices. Such quantized conductance states are inherently discrete and multilevel, which could find applications as multilevel nonvolatile memory and artificial synapses in hardware neural networks.

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In this work, we report the observation of conductance quantization in technologically relevant Ta₂O₅ or HfO₂ based resistive switching devices. Exploiting the discrete distribution of device conductance concentrated around fixed quantized values could help mitigate device variations and facilitate the programming or training processes in future memory/neuromorphic applications.

Resistive Switching Characteristics

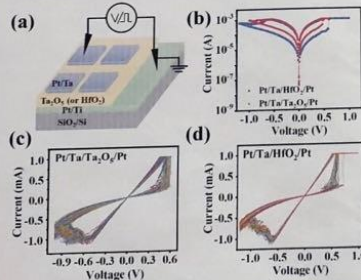


Fig. 1 (a) Schematic illustration of the device structure. (b) Log-scale I-V curves of Pt/TaTa₂O₅/Pt and Pt/TaHfO₂/Pt devices. (c) Linear-scale I-V curves in 100 cycles from Pt/TaTa₂O₅/Pt devices. (d) Linear-scale I-V curves in 100 cycles from Pt/TaHfO₂/Pt devices.

A schematic configuration of the two-terminal memory cell structure and its measurement setup is depicted in Fig. 1(a). Figure 1(b) shows the I-V curves of the Pt/TaTa₂O₅/Pt and Pt/TaHfO₂/Pt devices, both showing bipolar switching characteristics. The measurements were repeated for 100 cycles to examine the reliability of resistive switching, as shown in Figs. 1(c) and 1(d). The switching effects are highly reliable for both device structures, and it can be found that V_{set} and V_{reset} of the Pt/TaTa₂O₅/Pt device always stay lower than that of the Pt/TaHfO₂/Pt device, which indicates the Ta₂O₅-based cells have better retention and lower consumption in future application.

Conductance quantization Characteristics

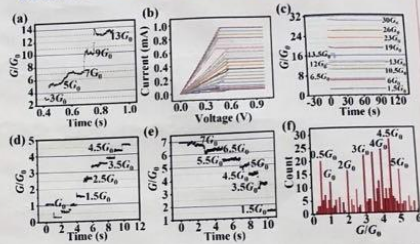


Fig. 2 (a) Conductance quantization observed during set process in Pt/TaTa₂O₅/Pt devices with a low sweeping speed of 1 mV/s. (b) I-V characteristics with different current compliances from 100 μ A to 1 mA. (c) Retention behavior of the quantized conductance states in Pt/TaTa₂O₅/Pt devices at room temperature. (d) The evolution of device conductance in Pt/TaTa₂O₅/Pt devices under a series of positive voltage pulses with an amplitude of 1 V and a width of 1 s. (e) The evolution of device conductance in Pt/TaTa₂O₅/Pt devices under a series of negative voltage pulses with an amplitude of -1 V and a width of 1 s. (f) Histogram of the conductance values extracted from the measured data.

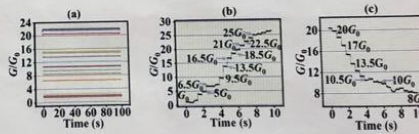


Fig. 3 (a) Retention behavior of the quantized conductance state in Pt/TaHfO₂/Pt device at room temperature. (b) The evolution of device conductance in Pt/TaHfO₂/Pt devices under a series of positive voltage pulses with an amplitude of 1 V and a width of 1 s. (c) The evolution of device conductance in Pt/TaHfO₂/Pt devices under a series of negative voltage pulses with an amplitude of -1 V and a width of 1 s.

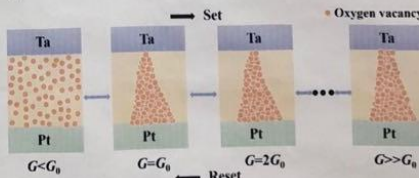


Fig. 4 Schematic illustration of the conductance quantization effect in Ta₂O₅ or HfO₂ based resistive switching devices.

Conclusion

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