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Realization of Synapse Behaviors Based on Memristor and Simulation Study With KMC Method

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ABSTRACT The memristor, emulating biological synapse, is recognized as one key way to overcome the classic von Neumann bottleneck. In this work, by using active metal Cu as electrode, the device of Cu/GeTeO$_x$/TiN exhibited typical resistive switching characteristic based on the electrochemical metallization mechanism (ECM). Moreover, it realized gradual potentiating and depressing conduction under DC and AC modes, which lead to emulating excitation and inhibition of biological synapses. According to these properties, spike-timing-dependent plasticity (STDP) learning rule was reproduced by applying appropriate pulse sequences. Furthermore, the kinetic Monte Carlo method was utilized to analyze and provide further demonstration for the behaviors of gradual change. These indicated that the device had great potential applied in the neuromorphic computing systems and the properties could be commonly realized by ECM mechanism.

INDEX TERMS Electrochemical metallization mechanism, conductive filament, memristor, spike-timing-dependent plasticity, kinetic Monte Carlo.

I. INTRODUCTION

Due to the complicated operations of fetch, decode and execution in the computation, traditional digital computers are confronted with the von Neumann bottleneck [1], indicating that new computing architectures are urgent to be constructed. As we know, biological brains can deal with the complex issues because of its computer in memory [2]. For human, the circuitry of a brain consist of about 10^{12} neurons, which link with each other by 10^{15} synapses that can execute learning and memory functions [3]. Thus, artificial synapses are essential for artificial brain neural network which promote the development of artificial intelligence. Importantly, the character of gradual change is the base of signal transmission between synapses. To realize this performance, the non-volatile memory device such as phase change memory (PCM) [4], magnetoresistive random access memory (MRAM) [5], ferroelectric random access memory (FeRAM) [6] and resistive random access memory (RRAM) [7] have been developing. Among these technologies, RRAM is widely considered as a promising candidate due to favorable CMOS compatibility, high density, good endurance, low power consumption and fast operation speed [8]. When the voltage is applied on the RRAMs, the conductance of it can be changed gradually, which is similar to biological synapses by adapting synaptic weights. These behaviors are closely related to the fluxes of Ca$^+$ and Na$^+$ in biology, which resemble with the migration of active metal ions. Moreover, the simple metal-insulator-metal (MIM) structure of RRAMs correspond to presynaptic terminal, synaptic cleft and postsynaptic terminal of biological synapses [9]. Obviously, the synapse was almost identical with electrical synaptic device in these aspects. Therefore, the RRAMs are expected to simulate biological synapses. In biological systems, spike-timing-dependent plasticity (STDP) rule is essential for learning and memory, while the characteristics of potentiation and depression of synapses are the foundations. These properties are necessary to emulate biological synapses for memristor. And there were some ECM-type memristors had been reported in recent years. For example, the pinched hysteresis loop of Ag/TiO2/FTO device showed similar properties to synaptic weights of biological neurons [10]. Moreover, the biological synaptic behaviors could be emulated by Cu/SiO2/W device through dc and voltage pulse programming [11]. However, there are seldom reports about pure GeTeO$_x$ switching layer as the function layer of memristor. Moreover, the conductive mechanism is worthy to further research in solid electrolyte which can facilitate the immigration of Cu ions.

In this work, we prepared Cu/GeTeOx/TiN devices with sandwich structure for memristor. The type of conductive mechanism was demonstrated, showing that resistive switching of the device was controlled by ECM. By applying voltages on the device, the device exhibited synaptic functions of potentiation and depression through the resistive characteristic of gradual change. Furthermore, the STDP rule had been fully reproduced and the behaviors of gradual change had been further analyzed and demonstrated by kinetic Monte Carlo method. Ultimately, a physical model of conductive filament was established to illustrate the synaptic behaviors of the device.

II. EXPERIMENTS

The simple MIM (Cu/GeTeO$_x$/TiN) device with sandwich structure was prepared [12]. Initially, the TiN/Ti/SiO$_2$/Si substrate was patterned to form the via hole with 0.16μm^2-16μm^2 effective area which was regarded as the device size. Then solid electrolyte GeTeO$_x$ (15nm) thin film was deposited on the substrate by RF magnetron sputtering with the GeTe target (99.99%) using a power of 120W and a working pressure of 4 torr at room temperature for 360s. Moreover, the switching layer was GeTeO$_x$due to the limitations of the manufacturing equipment and conditions. According to the analysis results of X-ray photoelectron spectroscopy (XPS) material analyzer, the mole fraction ratio of O: Te: Ge in the solid electrolyte layer was 11.2%: 30.49%: 58.31% [12]. Subsequently, a 200nm Cu film was deposited on the GeTeO$_x$ layer by dc magnetron sputtering. All electrical performances of the devices were measured by the Agilent B1500A semiconductor parameter analyzer with the voltage applied to Cu electrode and TiN electrode grounded in this work.

III. RESULTS AND DISCUSSION

According to previous research, the RRAM device with electrode of Cu had the potential to be an electrical synapse [13]. To study the electrical properties of the device, an electroforming process with a current compliance of 1mA was imperative before the dc voltage sweeping for initial device. After that, as shown in Fig. 1(a), when the voltage was applied to the device from 0V to 1V, the state of the device changed from high resistance state (HRS) to low resistance state (LRS) with compliance current of 1mA to protect the device from being damaged, which was called "SET" process. Similarly, the state of the device changed from LRS to HRS with negative voltage from 0V to -1V, which was called "RESET" process. Obviously, the current of devices with different effective area (0.16μm^2 to 16μm^2) were coincident at LRS while the current displayed slight difference at HRS, indicating that the performances of device was irrelevant with the size, which might relate to the conduction mechanism of it.

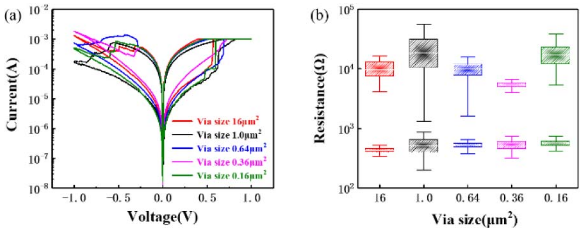

FIGURE 1. **(a) I-V curves of different devices with the effective area from 0.16μm^2 to 16μm^2 (b) The HRS and LRS resistance distribution of various devices.**

The conductive filament and interface barrier potential were the dominated conduction mechanism, although there were multifarious controversies for resistive switching property [14]. To confirm the conduction mechanism of the device, the detail resistance distribution of HRS and LRS (read at 0.2V) were shown in Fig. 1(b). With the device size scaling down, the resistance of LRS had seldom change while the resistance of HRS changed randomly, which implied the area of device had little influences on the resistance. Thus, the state of the device was controlled by the filament [15]. According to the ECM theory, the formation or rupture of the conductive filament formed by Cu atoms from top electrode related to the redox reaction of active metal atoms, which might correspond to the accumulation of Ca$^+$ and Na$^+$ in biology.

For biological synapses, the action potential generated by the accumulation of electric potential which was the result of the action between acetyl choline (ACH) and receptor [16]. This characteristic was similar to resistance change of device gradually with applying voltages. As shown in Fig. 2(a), the voltage from 0V to 0.4V (0.42V, 0.44V and 0.46V) and back to 0V, was applied to the device, causing the current of it increased slightly. Importantly, according to the hardly overlapped current paths, the resistance decreased from the resistance after last sweep, which demonstrated the device could maintain the state of resistance after each sweep. When the voltage from 0V to −0.6V (−0.6V to −0.7V with a step of −0.2V) and back to 0V was applied to the device, the current decreased gradually, which showed similar characteristic for keeping resistance state after each bias

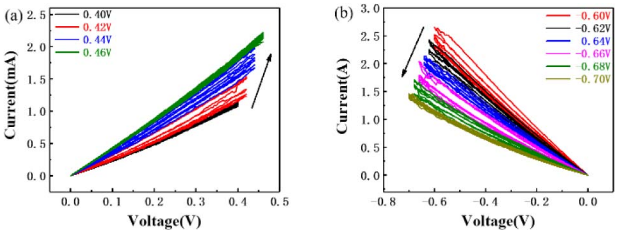

FIGURE 2. **The gradual change of current with (a) Continuous positive sweep and with (b) Continuous negative sweep.**

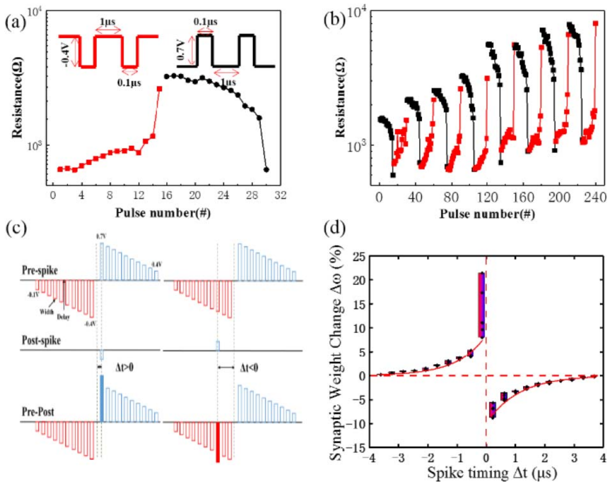

FIGURE 3. **(a) The gradual change of resistance under the pulse sequence; (b) several times repeated processes of (a); (c) the diagrams of the pulses; (d) the STDP fitting curves obtained by the Cu/GeTeO$_X$/TiN device.**

applied. Thus, the homologous accumulation effect of biological synapses behavior could also occur on the device with voltage applied, indicating it had simulated the behaviors of synapse preliminarily.

In order to further study the ability of simulation of synapses behavior, a sequence of pulses was applied to the Cu/GeTeO$_x$/TiN device, which consisted of 15 positive pulses and 15 negative pulses. As shown in Fig. 3(a), when negative pulses (−0.4V, 0.1μs) with short delay time (1μs) were applied continuously, the resistance gradually increased. Importantly, the state could maintain after each pulse, indicating that the resistance change of device was in response to negative stimulations, which illustrated the depression of biological synapses behavior. And then, as the 15 positive pulses (0.7V, 0.1μs) with same delay time were applied, the resistance gradually decreased, showing similar behavior of remain the state after each stimulation, which conformed to the potentiation of biological neural synapses. To further approach the processes of nonlinearity transmission in human brain, the two processes were repeated incessantly several times as shown in Fig. 3(b). The device exhibited continuously gradient characteristic of the device resistance similar to memory function in biology, which verified the device was qualified to the excellent analog characteristic under bipolar and modified pluses. Therefore, the potentiation or depression of biological synapses had been realized by the Cu/GeTeO$_x$/TiN device.

In biology systems, the long-term potentiation (LTP) would occurred when the pre-synaptic stimulation reached earlier than the post-synaptic stimulation ($\Delta t > 0$), which indicated that the connection strength of neurons would be enhanced [17]. Besides that, if the post-synaptic stimulation preceded the presynaptic stimulation ($\Delta t < 0$), the connection strength of neurons would be weaken, which was called the long-term depression (LTD) [18]. Furthermore, the LTP and LTD constituted spike-timing-dependent plasticity (STDP) learning rule which was important Hebbian learning rule.

To realize the emulation of biological synapse STDP learning rules with the Cu/GeTeO$_x$/TiN device, a simulative experiment was implemented. As shown in Fig. 3(c), the shape of the spike was designed with a time-division multiplexing (TDM) method [19]. The pre-spike consisted of 10 negative pulses and 10 positive pulses. For the negative depression pulses, the amplitudes of it were from −0.1V to −0.4V with a step of −0.03V and the pulse width was 0.1μs with 1μs delay time. In addition, the amplitudes of positive potentiation pulses were from 0.4V to 0.7V with a step of 0.03V, the pulse and the delay time were same with negative depression pulses. Moreover, the post-spike was designed as one pulse (0.3V or −0.3V) at positive or negative voltage region. Because the conductance of the device could be adjusted only when sufficient amplitude pulses were obtained by the overlap of pre-spike and post-spike, and the voltage dropped on the memristor was defined as the voltage of pre-spike minus the voltage of post-spike. Similarly, the interval time between the pre-spike arrived and the post-spike reached was defined as $\Delta t = t_{post} - t_{pre}$, and the relative change of synaptic weights was defined as $\Delta\omega = ((I_{after} - I_{before})/I_{before}) \times 100\%$ [20]. Importantly, the characters could be represented by error bas, proving that the STDP-like curves were reproduced in Fig. 3(d). The fitting curves usually accorded with an exponentially decaying function [21]:

$$\Delta\omega = A_{+}\exp(\Delta t/\tau_{+}) \text{ if } \Delta t > 0;$$
$$\Delta\omega = -A_{-}\exp(\Delta t/\tau_{-}) \text{ if } \Delta t < 0;$$

Here, the ranges of pre-to-post synaptic interspike intervals were decided by parameters τ_{+} and τ_{-} when synaptic potentiation or depression occurred. The maximum values of synaptic modification were determined with A_{+} and A_{-} when Δt approached to zero. For the device, when $\Delta t > 0$, the post-spike followed the pre-spike, the LTP would occur and the degree of relative change of weight become more obvious with the Δt decreased gradually. In addition, the LTD would happen when the post-spike preceded the pre-spike ($\Delta t < 0$), and the degree of relative change of weight was more apparent with the Δt increased gradually. Therefore, these results illustrated the agreement between the device and biological signal transmission of neurons,

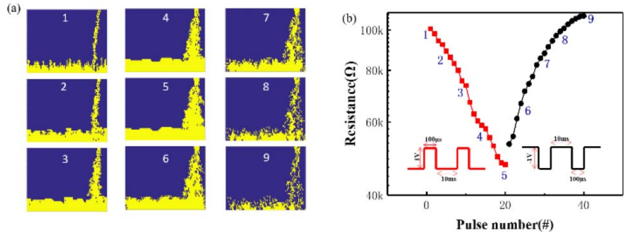

FIGURE 4. Kinetic Monte Carlo simulation: (a) the morphology of conductive filament, (b)the change of resistance by applying continuous pulses.

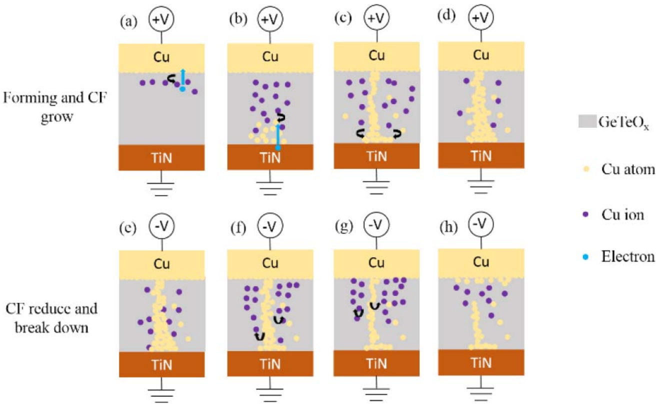

FIGURE 5. Schematic of the resistive switching process in the device (a) initial state (b) the formation and migration of Cu ions (c) the thin CF had formed (d) the CF grew gradually (e)-(f) under negative volatge, the CF began to dissolve and reduce (h) the CF broke down thoroughly.

implying the Cu/GeTeO$_x$/TiN device was one of electrical synaptic device which attained learning and memory function for the application of neuromorphic systems.

In order to understand the physical mechanism of these behaviors, the kinetic Monte Carlo (KMC) simulation was employed to analyze the oxidation, transportation and reduction of ions in two-dimensional matrix [22]. And the probability of transition to happen followed the Boltzmann distribution:

$$P = ve^{\frac{-E_a}{kT}}$$

where v was regarded as attempt frequency in solid dielectric, k was the Boltzmann constant and T was ambient temperature. E$_a$ stood for the migration barrier which meant the activation energy of dynamics. To study the detailed processes of ions dynamics, the time (t) was introduced, following:

$$P(t) = tve^{\frac{-E_a}{kT}}$$

As shown in Fig. 4(a), after necessary forming process, the conductive filament (CF) had formed basically in image 1. By applying 20 continuous positive pulses (1V, 100μs) on Cu electrode, the Cu atoms were oxidized to form Cu ions which would migrate to the bottom electrode, causing the CF grew steadily due to the effect of electric field (images 1-5). When negative pulses were applied, the external Cu atoms of CF began to become ions, which indicated the dissolution and diminution of CF would occur in images 5-9. Moreover, the resistance of device was extracted after every pulse plotted in Fig. 4(b), and the marked numbers were related to the images. Obviously, the resistance of device gradually decreased under positive pulses and then it would increase slowly after applying negative pulses. And the steady growth and gradual reduction of CF could be utilized to explain the resistive change in Fig. 3(a), which was called the character of gradual change. Thus, the migration and redox reaction of Cu ions caused the gradual change of resistance, which was proved by KMC method. Meanwhile, this simulation experiment conducted the establishment of physical model, which was used to explain the typical biological synapses characteristics caused by Cu ions.

According to these results, a simple physical model was established to explain the conduction mechanism of the device. As shown in Fig. 5(a), for the initial device, there were little Cu ions in GeTeO$_x$ solid electrolyte. When sufficient positive voltage was applied to Cu top electrode, an oxidation reaction occurred, causing Cu atoms changing to Cu ions. Then the Cu ions migrated to the direction of bottom electrode through the GeTeO$_x$ solid electrolyte with the effect of external electric field (Fig. 5(b)). Moreover, the Cu atoms regained due to the reduction reaction of Cu ions near TiN surface. Ultimately, the conducting filament (CF) consisted of Cu atoms had formed between top electrode and bottom electrode where the generation and nucleation of Cu atoms happened simultaneously (Fig. 5(c)). After forming process, the device was set to LRS, and the resistance decreased if small positive voltage (DC/Pulse) was applied. This result was caused by the growth of CF (Fig. 5(d)) and it had been demonstrated in the simulation, which was semblable to the potentiation of biological synapse. Similarly, the Cu atoms from CF dissolved with the negative voltage applied in Fig. 5(e), resulting the resistance of device increased gradually. Besides, the constraining force in CF became weaken as the Cu atoms detached gradually, accelerating the dissolution of Cu atoms (Fig. 5(f)-(g)), which was exactly related to the synaptic depression. Thus, the character of gradual change of resistance could completely simulate the basic synaptic behaviors which were the preconditions of complex learning rules. Finally, the CF broke down thoroughly under the effect of incessant small voltage (Fig. 5(h)). Therefore, the redox reaction between Cu atoms and ions, with directional migration of Cu ions under external electric field, generating the gradual change of resistance of Cu/GeTeO$_x$/TiN device, which exactly was unitized to simulate the behaviors of biological synapses in neuromorphic computing systems.

IV. CONCLUSION

In summary, the device based GeTeO$_x$ solid electrolyte was prepared. According to the resistive distribution of device with various areas, it was demonstrated that the resistive switching of the device was controlled by ECM. By applying voltages on the device with different modes, the resistance changed gradually and the state maintained after each change, indicating that the device fully simulated excitation and inhibition of biological synapses. Furthermore, the STDP behavior had been reproduced when the properly complex pulse sequences were applied. And the KMC method was utilized to provide further support for the behaviors of gradual change. Finally, these phenomenons was understood by the model of ECM mechanism, which was implied as a universal approach to realize the synaptic behaviors for neuromorphic computing systems and overcoming the bottleneck of von Neumann.

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