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APPLIED PHYSICS LETTERS **103**, 043503 (2013)

Dynamic switching mechanism of conduction/set process in Cu/a-Si/Si memristive device

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The conduction/set processes of resistive switching have been systemically investigated for Cu/a-Si/Si electrochemical memristive devices. Experimental results indicate that the set process was driven by two different mechanisms, depending on the programming pulse amplitude: a purely electrical dielectric breakdown and a thermally assisted dielectric breakdown. For the latter process, we observe that the set time decreased exponentially with the increase in the programming pulse amplitude, whereas the former process shows amplitude independence. Through the temperature-dependent set transition characteristics, we argue that the filament growth in set process could be dominated by cation transport in the dielectric film. The thermal activation energy of Cu hopping in a-Si is extracted to be 0.16 eV. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4816327]

Electrochemical memristive (ECM) devices (also known as conductive bridge memory) have attracted extensive attention due to promising applications in digital memories, programmable logic, and bio-inspired computing.[1,2] Application of electrical bias across two terminals in such device leads to the formation of a conducting filament composed of metallic ions from the active electrode such as Ag and Cu. For example, for Cu/a-Si/Si memristive devices the switching to the ON state (set process) involves electro-oxidation of the Cu electrode and drift of Cu ions towards Si electrode in a-Si film with subsequent reduction. The resulting filament can be dissolved (reset process) by applying reverse bias polarity. Compared to other ECM devices, e.g., to those based on chalcogenide materials, Cu/a-Si/Si devices have superior properties, e.g., CMOS processing compatibility.[3–7] In addition, a-Si memristive device can be switched continuously, i.e., in analog manner, by applying electrical bias (current or voltage pulses) with gradually increasing amplitude and/or duration, which is promising for analog computing circuits application.[8–11]

Despite its excellent performance, several reliability-related scientific and technical issues need to be elucidated before practical applications. One of the issues is about the wide set voltage distribution,[12–16] resulting in serious failure during writing information. The wide set voltage distribution is closely related to the random formation of the conducting filaments, and multi-conduction paths could be formed by forming/set process. Then, parts of these paths are ruptured and formed by applying voltages, leading to reset and set switching, respectively. In general, tree-like structures conduction paths randomly appear and distribute irregularly in the switching materials due to a random soft breakdown.[17–21] Recovery of these paths can occur for repeated set/reset cycles due to the random ionic migration, which spoils the repeatability of the devices. Some methods have been reported to improve this problem, such as impurity doping,[13–15] interface engineering,[22] S-type negative differential resistance integration,[16] and nanoparticle incorporation.[12] Therefore, a complete understanding of the set process is required for both scientific interest and numerous practical applications. In this paper, we study on the kinetics of conduction/set process, i.e., from high resistance state (HRS) to low resistance state (LRS), of Cu/a-Si/Si ECM devices under different pulse amplitude. Temperature-dependent measurement in HRS, LRS, and set transition are investigated to get an insight into the dynamics switching mechanism as well.

The Cu/a-Si/Si device studied here consists of a highly doped p-type Si substrate with the resistivity of 0.001 Ω cm, an a-Si layer, and a Cu layer acting as the bottom electrode, the switching layer, and the top electrode, respectively, as schematically shown in the inset of Fig. 1(a). The blanket a-Si films with the thickness of 75 nm were deposited by using a low-pressure chemical vapor deposition technique at room temperature. The top Cu electrodes with a diameter of 5 μm were fabricated by using magnetron sputtering technique and a subsequent lift-off process. More details of the device fabrication process were described in Ref. 23. The current-voltage (I-V) characteristics were carried out with an Agilent 4155C semiconductor parameter analyzer. The pulse waveform was applied to the device by using a Yokogawa FG300 synthesized function generator, while the output was measured with a Yokogawa DL7100 digital oscilloscope. The temperature dependence measurement was measured by Cryogenic Lakeshore Probe Station and cooled down with liquid nitrogen.

Figure 1(a) shows typical bipolar resistance switching characteristics of Cu/a-Si/Si device. The as-fabricated device has a very high resistance, 10^9 Ω, and the irreversible forming

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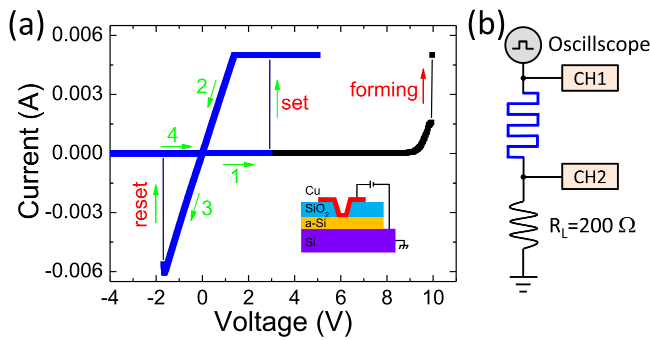

FIG. 1. (a) A typical I-V switching characteristics of Cu/a-Si/Si device. After the forming sweep ($0 \rightarrow 10$ V), a full voltage sweeping cycle follows this sequence: $0\,V \rightarrow +5\,V \rightarrow 0\,V \rightarrow -4\,V \rightarrow 0\,V$. The current is limited to a compliance of 5 mA to prevent the permanent breakdown during forming/set process. The inset is the schematic structure of Cu/a-Si/Si device. (b) The schematic pulse measurement setup. Load resistor (R_L) is equal to 200 Ω.

process was required to activate the pristine device, triggering an abrupt jump from high resistance state to low resistance state with a current compliance of 5 mA to avoid permanent dielectric breakdown. After the first cycle, the voltage was applied in a sweeping sequence of $1 \rightarrow 2 \rightarrow 3 \rightarrow 4$. As the voltage was swept from 0 to 5 V with a 5 mA current compliance, an abrupt increase in current was observed at +2.9 V. The resistance switches to LRS from HRS, called as "set" process. While sweeping the voltage from 0 to −1.7 V, the resistance switched to HRS again, called as "reset" process. Our Cu/a-Si/Si device exhibits a typical bipolar nature, i.e., it can only be written with a positive bias and erased with a negative one, which indicates that the resistive switching effect is likely due to electrochemical redox reactions.[24]

To further clarify the conduction and switching mechanisms of the Cu/a-Si/Si device, the *I-V* curve was redrawn in logarithmic plot, and the linear fitting results were shown in Figure 2(a). The LRS exhibits a linearly Ohmic behavior with a slope of 1.03, which corresponds to the formation of conducting filaments in a-Si films during set process.[25] The conduction mechanism in HRS, however, is entirely different. It consists of three portions: the Ohmic region ($I \propto V$), the Child's law region ($I \propto V^2$), and the steep current increase region, which can be well explained by trap-controlled space charge limited conduction (SCLC).[26]

Figures 2(b) and 2(c) exhibit the temperature dependence of resistance in LRS and HRS, respectively, and the measurements were carried out in vacuum. The resistance of LRS increases linearly with increasing the temperature, which is a typical metallic characteristics and can be written as $R(T) = R_0[1 + \alpha(T - T_0)]$, where R_0 is the resistance at temperature T_0, α is the temperature coefficient. For the $5 \times 5\,\mu m^2$ device, $\alpha = 2.99 \times 10^{-3}\,K^{-1}$, which is good agreement with temperature coefficient of Cu/ZrO_2:Cu/Pt memristive device,[27] as shown in Fig. 2(b). This means that there is a metallic conducting Cu filament formed in the a-Si films. The measured α is smaller than the value of high-purity Cu ($\sim 3.9 \times 10^{-3}\,K^{-1}$).[28] This discrepancy is due to inevitable defects in the nano Cu filaments, since the presence of defects can reduce the value α significantly by shortening the mean free path of electrons.[29] The resistance of HRS [see in Fig. 2(c)], however, shows a decreasing trend as increasing the temperature, which could be ascribed to a typical semiconducting behavior. The temperature dependence of current in a semiconductor is given by

$$I = I_0 \exp(-\phi_t/kT), \quad (1)$$

where k is the Boltzmann constant, ϕ_t is the thermal activation energy, and T is the absolute temperature. ϕ_t can be extracted to be 0.15 eV from the slope of the Arrhenius plot, as shown in the inset of Fig. 2(c).

A pulse waveform voltage with gradually incremental amplitude (V_p) was employed to reveal the influence of the set voltage on the dynamics mechanism of the set process. The measurement setup is schematically shown in Fig. 1(b). There is a sudden increase of current, when time reaches the set time T_{SET}, as shown in Fig. 3(a). Figure 3(b) shows a $\log T_{SET}$ versus V_p plot of the experimental data measured on Cu/a-Si/Si device. As shown with the blue diamond, T_{SET} decreases from 0.8 s to 48 ns at 300 K by increasing voltage from 3 V to 10 V. Two clear exponential relationships between T_{SET} and V_p were observed with cross-point at $V_p^{cp} \approx 7$ V. The T_{SET} for $V_p < V_p^{cp}$ decreases much faster than those for $V_p > V_p^{cp}$. It is worth to note that these results were reported in other memristive devices.[30,31] The temperature dependences of two exponential relationships between V_p and T_{SET} also show opposite behaviors as displayed by red circles in Fig. 3(b). As temperature changes from 300 K to 100 K, T_{SET} increases in $V_p < V_p^{cp}$, but there is no obvious difference in $V_p > V_p^{cp}$. As we know, dielectric breakdown is governed by two mechanisms: purely electrical dielectric breakdown and thermally assisted dielectric breakdown.[32] The former comes from electronic process, such as single-electron collision ionization, space charge production

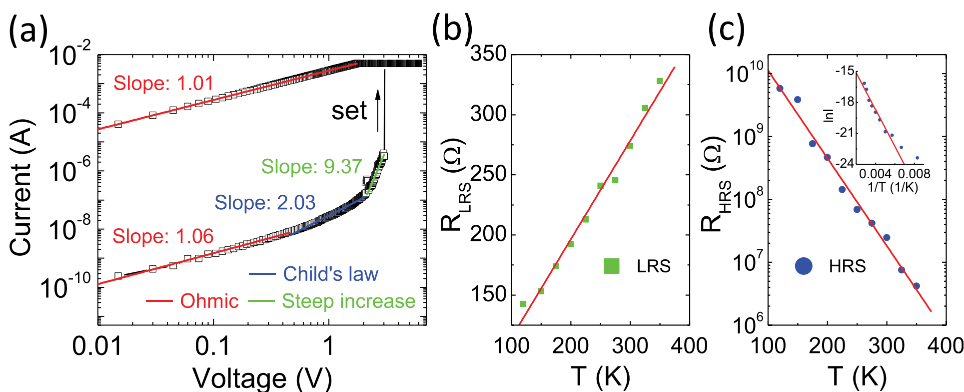

FIG. 2. (a) The linear fitting for the I-V curve of Cu/a-Si/Si device in log-log scale in set process and the corresponding slopes for each part. Temperature dependent resistance in (b) low resistance state and (c) high resistance state. Inset is Arrhenius plot for the current–temperature curve measured in the high resistance state.

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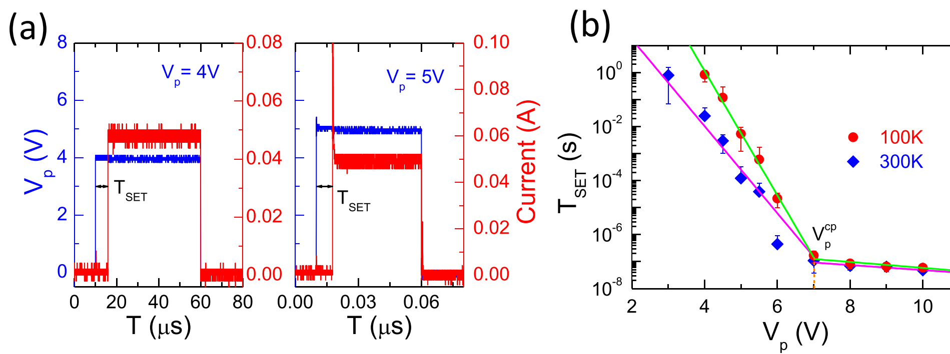

FIG. 3. (a) The switching dynamics of set processes of the Cu/a-Si/Si device triggered by square pulses with gradually incremental amplitude. (b) Set time as a function of square pulse amplitude (V_p) at 300 °C and 100 °C.

from collision ionization. In the latter, the Joule heating of the pre-breakdown current plays a key role, which is highly related to ion migration inside the film. For Cu/a-Si/Si device, Cu^{z-} ions act as free ions for the set process in the thermally assisted breakdown ($V_p < V_p^{cp}$ regimes). On the other hand, in $V_p > V_p^{cp}$ regimes, the purely electrical dielectric breakdown could induce the set process.

The filament growth dynamics in set process was further investigated by examining the wait time (i.e., T_{SET}) before the sudden resistance switching as a function of temperature. Figure 4(a) shows the time dependent set process as a function of temperature under constant amplitude of square pulse. The device was originally programmed in the OFF state. The sudden jump to the ON state corresponds to the formation of the Cu filament through thermally activated diffusion of the Cu ions toward the bottom electrode. The thermally activation energy E_a can be extracted to be 0.16 eV from the Arrhenius type plot of the wait time t versus 1/T based on Eq. (2), as shown in Fig. 4(b)

$$t = t_0 \exp(E_a/kT), \qquad (2)$$

where t is wait time and t_0 is a characteristic dwell time. As we know, the filaments growth of set process involve three steps: (1) anodic dissolution of Cu, (2) transport of the Cu^{z+} across the a-Si film, (3) reduction of Cu^{z+} and crystallization of conducting pathway. All these steps may be the rate-limiting process. In principle any of these steps, if they are the rate-limiting one, could give rise to an exponential relationship (i.e., ln $t - 1/T$). Schindler *et al.*[33] indicated that crystallization of Cu on the cathode is the rate-limiting process in Cu/SiO_2/Pt device. Yang *et al.*,[20] however, showed that the rate-limiting process is dominated by the cation transport process in Ag/a-Si/Pt device but is dominated by crystallization of Ag onto Pt in Ag/SiO_2/Pt devices, suggesting a strong dielectric dependence on the rate limiting process. Here, the ln t vs. $1/T$ is linear, and the extracted activation energy, E_a, is close to the value of the diffusion barrier of Cu in Si.[34] This implies the hypothesis of field-assisted ion transport inside the dielectric film and supports that the kinetics of the set process in Cu/a-Si/Si device is dominated by cation transport process.[20]

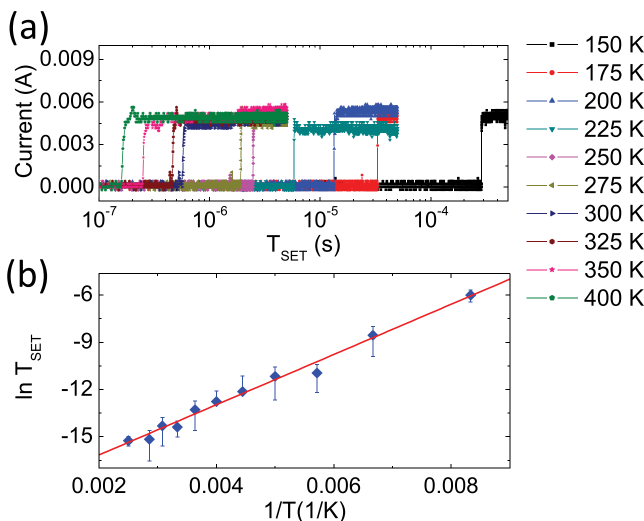

FIG. 4. (a) The switching dynamics of the Cu/a-Si/Si device set by square pulses with fixed amplitude (5 V) at temperature ranging from 150 K to 400 K. (b) Arrhenius type plot of the set time as a function of temperature (1/T). Thermal activation energy is extracted at 0.16 eV.

In summary, we systemically investigated the dynamics mechanism of the conduction/set processes for Cu/a-Si/Si electrochemical memristive devices, which is classified a purely electrical dielectric breakdown and a thermally assisted dielectric breakdown. The thermally activated Cu ions migration in a-Si might play a key role in the set process for the thermally assisted dielectric breakdown. The thermal activation energy of Cu hopping in a-Si is extracted to be 0.16 eV by Arrhenius equation.

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