Visible-Light Effects on Nanoscale Conducting Filament in SiO₂/Cu CBRAM

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Abstract- Visible-light-induced resistance switching of the SiO₂/Cu stack was investigated via a conductive atomic force microscope (C-AFM). Results show: (i) the conducting filament (CF) immediately after forming can be disrupted upon illumination, even without an applied voltage; (ii) the CF becomes insensitive to illumination after repeated post- forming voltage sweeps. These findings suggest that the photosensitivity of the CF is suppressed by the formation of a complete Cu filament during the repeated voltage sweeps. Our study shows the prospect of achieving expanded functionality of a single SiO₂/Cu CBRAM device by virtue of the CF's photosensitivity and its modulation by electric-field.

Introduction- Conductive-bridging type resistive switching random access memory (CBRAM) is one of the most promising RRAM candidates for big-data-storage and neuromorphic-computing applications^{[1][2]}. However, for practical application, power consumption and data retention aspects need to be improved. Recently, we have demonstrated visible-light-induced resistance reset (LIR) of the HfO₂ and ZrO₂ resistive memory devices^[3]. However, the detailed mechanism of LIR is still unclear. In this study, we further investigate the effect of light on the nanoscale property of a CF in the SiO₂/Cu stack.

Experimental- A HF-cleaned p-Si(100) was deposited with 30-nm Ti and 30-nm Cu films via DC magnetron sputtering. Subsequently, a 10-nm SiO₂ was formed at 250 °C via parallel-plate plasma-enhanced chemical vapor deposition with SiH₄ and N₂O. For comparison, a non-metal sample of SiO₂(5-nm)/p-Si was prepared (no-Cu sample). Electrical measurement was performed using ultra-high vacuum C-AFM system with the diamond-coated Si probe connected to a Keithley SCS4200 parameter analyzer (Fig.1). The bias voltage was applied to the probe and the substrate was grounded. A commercially available white LED light was used in the light-induced resistance-reset study.

Results and Discussion- By applying a negative-voltage ramp to the C-AFM probe, an abrupt current increase is observed at -10 V for the no-Cu sample (Fig.2(a)) and at -5 V for the Cu sample (Fig.2(b)). Subsequently, a positive-voltage ramp was applied (2^{nd} sweep) . No resistance reset is observed in the former; note that there is no evident drop in the low-resistance state (LRS) current. This is supported by a subsequent negative-voltage ramp (3^{rd} sweep) , in which the current-voltage curve is similar to that of the 1st reverse-sweep. In contrast, a significant current drop is observed in the Cu sample (Fig.2 (b) 2^{nd} sweep), indicating a successful reset. This is confirmed in the 3^{rd} sweep, in which the current is comparable to the pre-forming current. It is believed that various intrinsic defects (e.g., oxygen vacancies (V_o 's) and

dangling bonds) in SiO₂ results in irreversible resistance change in Fig.2(a), while the ionized Cu diffuses into the SiO₂ bulk, leading to the resistive switching observed in Fig.2(b). Fig.3 shows that the LRS current is decreased instantly from $\sim 10^{-12}$ to $\sim 10^{-14}$ A, for both post-forming no-Cu and Cu samples upon illumination at 100-s. Even after the light was turned off at 200-s, the current level remains at $\sim 10^{-14}$ A. Clearly, the samples were restored to the high resistance state (HRS) under light stimulation. It is suggested that light helps reduce the V_0 's in SiO₂ and rupture the CF for both samples, even for the no-Cu sample in which reset is not observed under voltage ramp (Fig.2(a)). For Cu electrode CBRAM, it is reported that Cu precipitates and forms a CF in a solid electrolyte^[4]. Thus, we attribute the light-induced reset in the Cu sample to the reduction of V_0 's which exist in between the Cu filament and the SiO₂/probe interface. Fig.4 depicts the post-LIR set voltage sweep. It is interesting to note that the set voltage of the Cu sample is almost similar to the forming voltage, implying a restoration to HRS by light exposure. While for the no-Cu sample, the set voltage is -6 V, as compared with the forming voltage of -10 V. To improve the stability of CF against light, negative-voltage ramp (0 to -8 V) was repeated several times before illumination. As shown in Fig.5, the no-Cu sample exhibits a significant current drop upon illumination at 100-s. For the Cu sample, on the other hand, the LRS current level is not affected by illumination and remains approximately constant throughout the measurement. However, by applying a positive-voltage ramp after light illumination, field-driven reset is observed as shown in Fig.6. These findings indicate that the photosensitivity of the CF in the Cu sample can be modulated by the repeated voltage sweeps. As illustrated in Fig.7, it is believed that after several voltage sweeps, more Cu ions are drawn into the SiO_2 bulk replacing the V_0 's that formed part of the CF previously. Since the number of the V_0 's is reduced, the CF therefore becomes less reactive to light. However, as the probe is positively biased, some of the Cu ions move back to the electrode and the V_{o} 's near the oxide/probe interface recombine with oxygen ions, thus disrupting the CF. This explains the field-driven reset observed in Fig.6.

Conclusion- Light-induced resistance reset is observed in both SiO_2/p -Si and $SiO_2/Cu/Ti/p$ -Si samples. This phenomenon is attributed to the annihilation of the V_0 's in SiO_2 film under light stimulation. For the latter sample, however, light loses its effect after several repeated voltage sweeps in the LRS, indicating that electric field can modulate the photosensitivity of the CF by drawing more Cu ions into SiO_2 and replacing the V_0 's in the CF. The CF's photosensitivity and its electric-field modulation may enable greater functionality to be realized in a single SiO_2/Cu CBRAM device.

References

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Fig. 1. Schematic illustration of the UHV-CAFM measurement setup and the two test-sample structures (not to scale) in this study. The CAFM probe is connected to a source/monitor unit of a parameter analyzer. The current compliance is limited to 10^{-9} A to avoid film degradation. A white LED lamp (~30 cm from the sample) was directed at the sample through a quartz window of the UHV chamber for the light exposure measurement.



Fig. 2. Current-voltage (*I-V*) curves of (a) SiO₂/p-Si and (b) SiO₂/Cu/Ti/p-Si samples. After the negative-voltage ramp (1st sweep, Black Line) for forming, a positive-voltage ramp (2nd sweep, Red Line) was applied. There is no current drop in the SiO₂/p-Si, indicating no reset. On the other hand, a significant current decrease is observed in the SiO₂/Cu/Ti/p-Si, showing a successful reset. In the following negative-voltage ramp (3rd sweep, Blue Line), the *I-V* curve of the SiO₂/Si overlaps that in the 1st reverse sweep, confirming that the LRS was retained. In contrast, the *I-V* curve of the SiO₂/Cu/Ti/p-Si shows a current jump similar to that of the 1st sweep, showing reforming of the conductive filament.



Fig. 4. The initial and post-light-induced-reset (LIR) current-voltage (*I-V*) curves of (a) SiO_2/p -Si and (b) $SiO_2/Cu/Ti/p$ -Si samples. After LIR, the CF is formed again by electrical field for both samples. The post-LIR set voltage in the SiO_2/Si changes from -10V to -6V possibly due to partial irreparable breakdown. On the contrary, the post-LIR *I-V* curve is almost similar to that in the 1st sweep in the $SiO_2/Cu/Ti/Si$, implying a nearly complete reset by light.



Fig. 3. Current-time (*I-t*) plots of SiO₂/p-Si and SiO₂/Cu/Ti/p-Si samples under light exposure. The currents in both samples show a steep decrease upon illumination at 100-s and remain low at $\sim 10^{-14}$ A after light was turned off at 200-s.

Fig. 5. Current-time (I-t) plots SiO₂/p-Si of and SiO₂/Cu/Ti/p-Si samples under light exposure after both were subjected to repeated post-forming voltage sweeps. LRŠ The current of SiO₂/Cu/Ti/p-Si sample does not drop instantly upon illumination at 100-s, while the LRS current of SiO₂/p-Si sample decreases rapidly at 100-s.



Fig. 6. *I-V* curves of the $SiO_2/Cu/Ti/p-Si$ sample before and after the light exposure measurement in Fig.5. The current before the light exposure measurement (Pink Line) is almost similar to that of the post-forming reverse sweep curve. The field-driven reset is observed in the 1st sweep after the light exposure measurement (Red Line). In the subsequent negative-voltage ramp, the CF is formed again (Blue Line).



Fig. 7. Schematic diagram showing a possible explanation of the Cu-CF stabilization by repeated post-forming voltage sweeps. After the 1st voltage sweep for forming, some oxygen vacancies exist in between the Cu filament and the SiO₂/probe interface as shown in (a). After repeated voltage sweeps, more Cu ions diffuse into the SiO₂ bulk (arrows), replacing the oxygen vacancies as shown in (b). Consequently, the CF becomes less reactive to light.