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In Situ STEM of Ag and Cu Conducting Bridge Formation through Al_2O_3 in Nanoscale Resistive Memory Devices

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Non-volatile resistive memory, specifically conducting-bridge RAM or CBRAM, is a potential successor to flash memory. CBRAM requires less power than flash memory, can switch on and off faster, and can withstand a larger number of on/off cycles [1]. In principle CBRAM devices are also expected to be scalable to only a few nanometers. In CBRAM, the memory element switches to an "on" state when a conducting bridge forms through the insulating layer separating two conducting electrodes. Despite recent intense interest in CBRAM, the specifics of the formation and breaking of this bridge is not well understood. We have fabricated horizontally-aligned CBRAM devices specifically designed for high-resolution S/TEM imaging of conducting filaments *in situ.*

$Pt/Al_2O_3/(Cu$ or $Ag)$ CBRAM elements are fabricated on electron-transparent Si_3N_4 membranes. The metal electrodes are offset laterally by a few nanometers, as seen in Figure 1a, allowing an unobstructed view of the region between electrodes where filaments form. Depositing the Al_2O_3 via ALD ensures that the conducting bridge forms through the insulating layer and not along layer interfaces. An additional capping layer of Al_2O_3, also deposited by ALD, prevents surface migration of the active metal. Cu and Ag are chosen because they are known to diffuse in several bi-metal oxides, including Al_2O_3, under a voltage bias [2]. Both perform similarly in our devices, with Ag requiring a slightly smaller bias to form a filament.

Figure 1b plots the absolute value of current vs. voltage collected during a switching cycle of an Ag device. The device is simultaneously imaged with dark-field STEM at 300 kV. To accurately correlate events in STEM videos with transport data, a buffered signal of the source voltage is connected to a detector input for our microscope. The signal becomes digitized and an image displaying the voltage value at each pixel is generated concurrently with the dark-field STEM image. Figure 1c is a STEM image of the $Pt/Al_2O_3/Ag$ device from which the data in figure 1b was measured, and figures 1d-f are difference images corresponding to regions 1-3 in the plot, respectively. The difference images are generated by subtracting the image in 1c from frames of a STEM video acquired while biasing the device. A threshold filter was used to highlight regions of large intensity increase between frames.

In the plot, the device is initially in a many-GΩ high resistance state (HRS). As the voltage is increased, patches of increased intensity form on the Pt electrode and comparably-sized regions of intensity decrease appear on the Ag electrode, as seen in 1d. The re-deposition of Ag onto the Pt prior to switching indicates that Ag filaments are growing from the Pt electrode back towards the Ag electrode, consistent with recent theoretical work [3]. In this device a "partial switch" occurs at region 2, in which a very narrow filament, with a resistance of ~100 kΩ, forms a weak bridge (left side of 1e). After several seconds a wider Ag filament forms (right side of 1f) and the device switches to a low resistance state (LRS) at region 3, where resistance drops to less than 1kΩ. The electrical leads that join the CBRAM element to the voltage source have ~1kΩ of impedance, so the LRS resistance of 1kΩ serves as an upper limit. The voltage is reversed until the filament breaks and the device returns to the HRS, which occurs at region 4 in 1b (difference image not shown).

The design of these devices makes high-resolution imaging of the entire active region of a CBRAM device possible. High temporal resolution of events recorded *in situ* can be obtained by correlation of STEM and transport data pixel-by-pixel. With this novel approach to CBRAM imaging we hope to further clarify the dynamics of conducting bridge formation and breaking, and to explore the limits of minimum memory element size.

References:
[1] J. Yang, et al, *Nature Nanotechnology* **8** (2013), 13-24
[2] S. Rahaman, et al, *Nanoscale Research Letters 7* (2012) 345
[3] I. Valov, et al, *Nature Communications* **4** (2013) 1771
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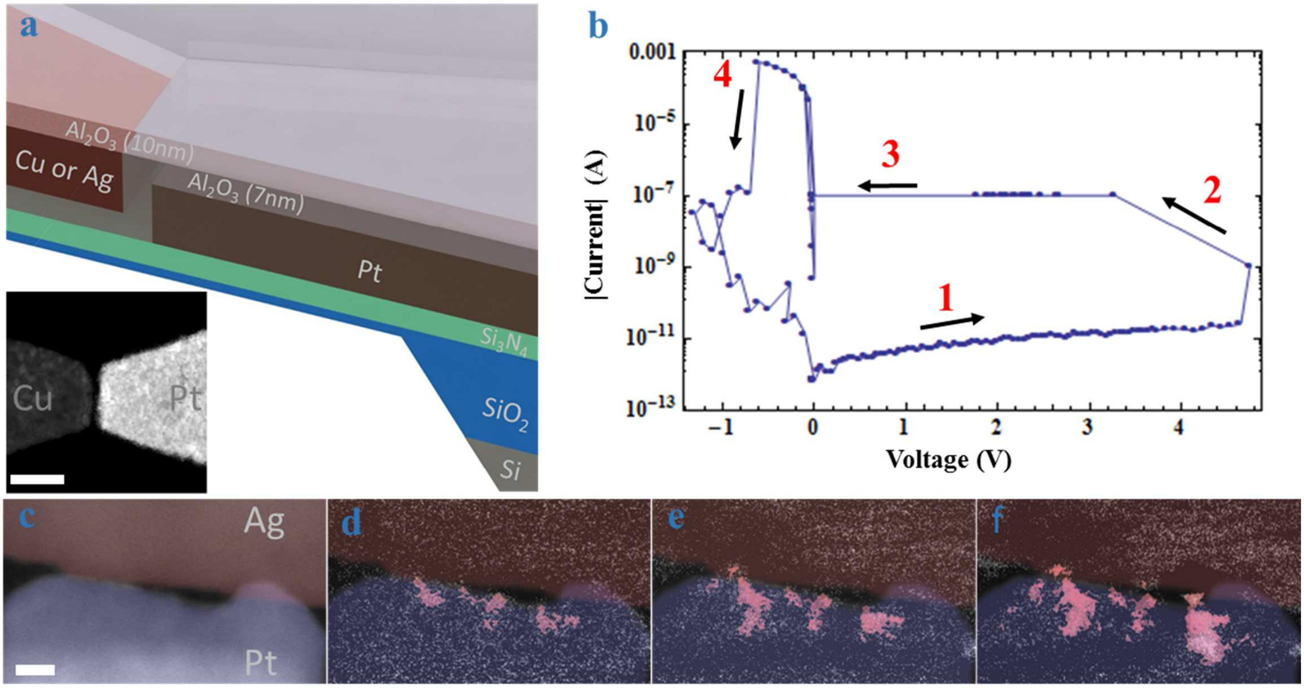

Figure 1. (a) Schematic of a Pt/Al_2O_3/Cu or Ag CBRAM device on a TEM biasing chip. Inset: A dark-field STEM image of a Cu device with a 50 nm scale bar. (b) Plot of |current| vs. voltage for a single switching cycle for a Pt/Al_2O_3/Ag device, collected simultaneously with the images used to produce (c)-(f). (c) A dark-field STEM image of the same Pt/Al_2O_3/Ag device before biasing, with Ag in red and Pt in blue. The scale bar is 10nm. (d)-(f) Difference images showing changes in intensity relative to the initial image in (c). (d)-(f) show changes in the device at regions 1-3 on the plot in (b), respectively, with 10s elapsing between each of the 3 images. The initial state of the electrodes is overlaid in red and blue, and the light red corresponds to a large increase in intensity after image subtraction.