Electrically induced resistive switching in metal-insulator-metal structures is a subject of increasing scientific interest, because it is one of the alternatives that satisfies current requirements for universal non-volatile memories. However, the origin of the switching mechanism is still controversial. There have been numerous attempts to identify the origin of resistance changes in various resistive random access memory (ReRAM) materials and thereby understand the switching mechanism associated with the behaviors of oxygen vacancies [1] and the metal ions [2]. Here, we introduce the observation and identification of conducting paths in the solid electrolyte-based and oxide-based resistive switching devices under different switching conditions using a unique in-situ probing technique inside TEM in conjunction with the high spatial resolution of EELS and EDS.

To understand switching behaviors in a solid electrolyte memory composed of Cu-doped GeTe sandwiched between a Cu BE and a TE, we performed in-situ TEM observations at various voltages and measured the corresponding I–V characteristics. Figure 1a shows an in-situ I–V scan. Starting from the high resistance state, we applied a negative voltage up to –0.8 V, and then applied a positive voltage of + 0.4 V. Cross-sectional Z-contrast STEM images were obtained after each voltage application (Figs. 1b–e). After applying –0.8 V, the multiple filaments become strengthened (Fig. 1d). Subsequent application of a positive voltage annihilates the filaments (Fig. 1e) [3]. We also constructed the oxide-based ReRAM device using a cross-sectional sample of the Pt/SiO$_2$/Ta$_2$O$_{x}$–TaO$_{2–x}$/TaO$_{5–x}$/Pt structure to enable real-time observation of the voltage-induced structural changes in the conduction paths (Figs. 2a, b). The switching behavior of the ReRAM device inside the TEM was confirmed by in-situ measurements of the I–V characteristics at various voltages. This device sample exhibited reversible bipolar resistance switching behavior between the LRS and HRS by DC I–V sweeps (Fig. 2c). Comparison of Z-contrast STEM images taken at the same location under LRS (Fig. 2d) and HRS (Fig. 2f) unambiguously exhibited the nanoscale filament formation in the SiO$_2$ layer; it was ~ 1.5-2 nm in width and 0.6-1.5 nm in length (Fig. 2e) [4].

These in-situ studies provide crucial information for understanding the dynamics of filament formation and annihilation processes. We expect that the in-situ experimental technique can further expand its applications in a variety of nonvolatile memory system.

References
Fig. 1: In-situ observations of voltage-induced changes in microstructure. a, In-situ I-V scan. b-e, Cross-sectional Z-contrast STEM images obtained after voltage applications of 0, -0.4, -0.8 and +0.4 V, respectively.

Fig. 2: In-situ observation of the conducting paths. a, Schematic of the in-situ experimental setup. b, Cross-sectional STEM image when the Pt-Ir tip approached the top Pt electrode. c, A series of I-V measurements. d-f, In-situ STEM images in LRS (d) and HRS (f). Magnified images of nanoscale filaments (e) obtained from the red boxed areas (A and B) in d.