Electronic Supplementary Information for

Chameleonic electrochemical metallization cells: dual-layer solid electrolyte-inducing various switching behaviours

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1. Rutherford backscattering spectrometry analysis on PGSC and PSGC ECM stacks

Rutherford backscattering spectrometry (RBS) analysis revealed the role of the upper SiO_x (SO) layer (10 nm) in the Pt/$GeSe_x$/SiO_x/Cu stack (PGSC for short) in blocking Cu diffusion into the lower $GeSe_x$ (GS) layer. The Pt/$GeSe_x$/Cu stack (PGC for short) includes a large number of Cu atoms/ions in the GS layer as idenified by the Cu shoulder around channel 700 in Figure S1a. Dissimilar to the PGC stack, the RBS spectrum of the PGSC in Figure S1b exhibits remarkable suppression of Cu diffusion into the solid electrolyte layers underneath, which leads to the reduction in the channel width for the Cu signal. Note that the thicknesses of the Cu layers in both stacks were the same (50 nm).

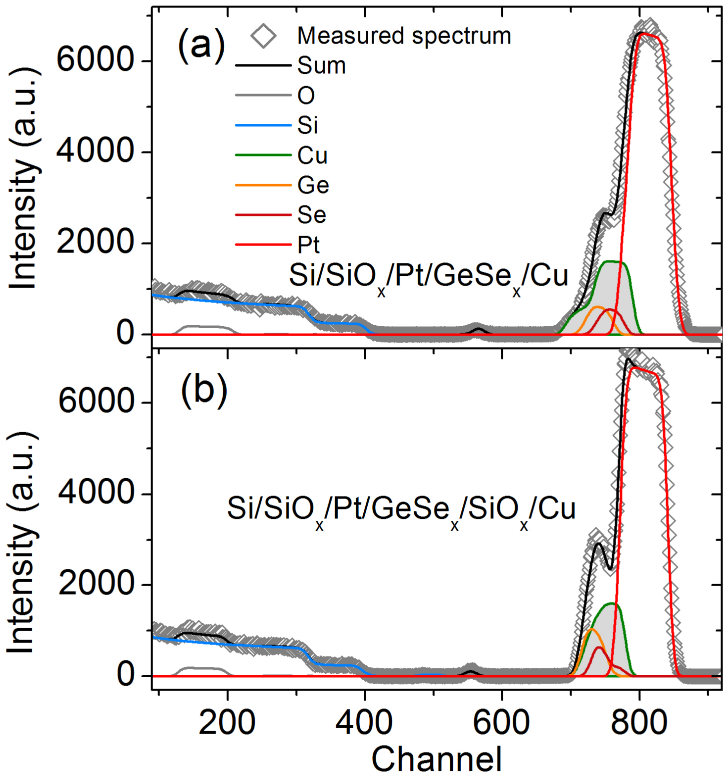

Figure S1. RBS spectra for (a) PGC and (b) PGSC stacks.

2. SO thickness-dependence of complex switching behaviour

The ECM cell with a thicker SO layer (≥10 nm) does not show counter-eightwise bipolar switching behavior; instead, threshold switching with compliance current and complex switching without compliance current are elicited. A typical *I-V* hysteretic loop for each case is plotted in Figure S2. In general, the thicker the SO layer, the less obvious threshold switching behaviour is observed as seen in Figure S2. Note that no electroforming was applied to all ECM cells irrespective of thickness of the SO layer.

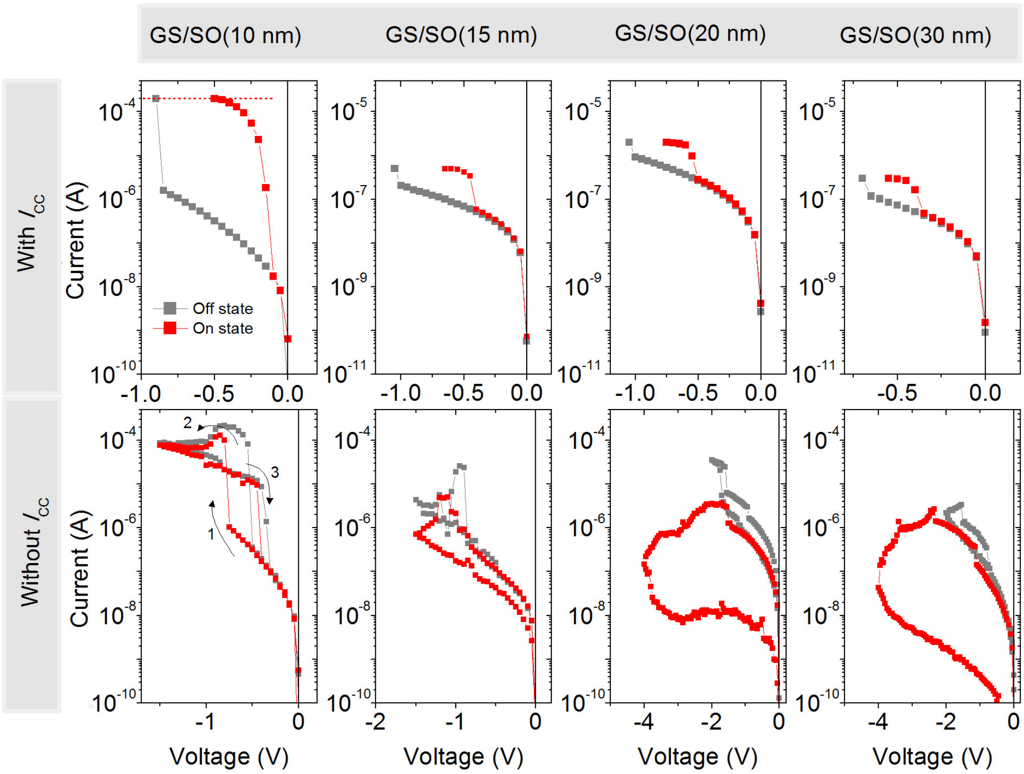

Figure S2. I-V hysteresis change upon the thickness of the upper SO layer (10, 15, 20, and 30 nm)—measured (upper panel) with and (lower panel) without compliance current (I_{CC}).