Supplementary Information for

Embedded nanoparticle dynamics and their influence on switching behaviour of resistive memory devices

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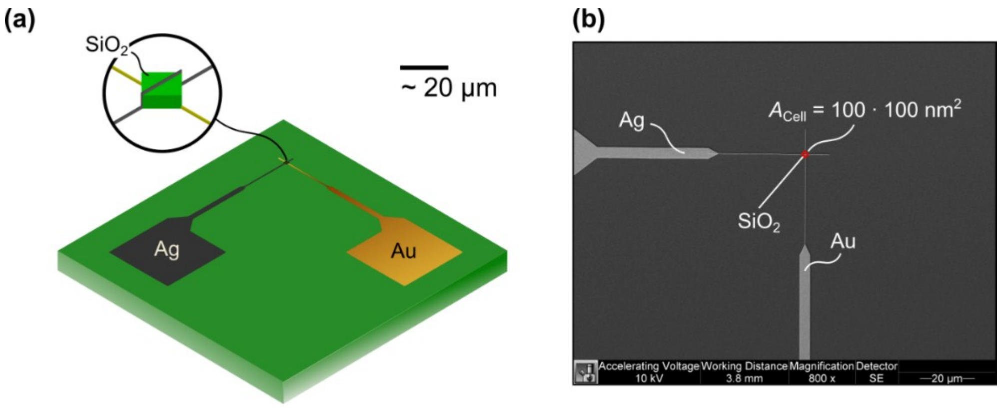
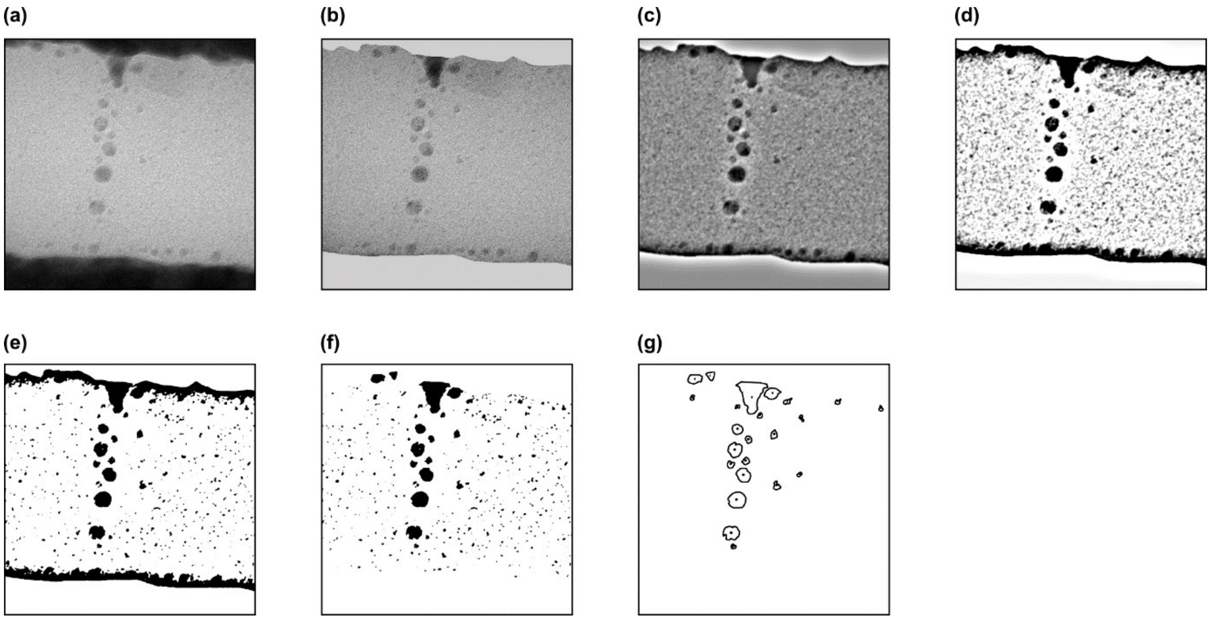
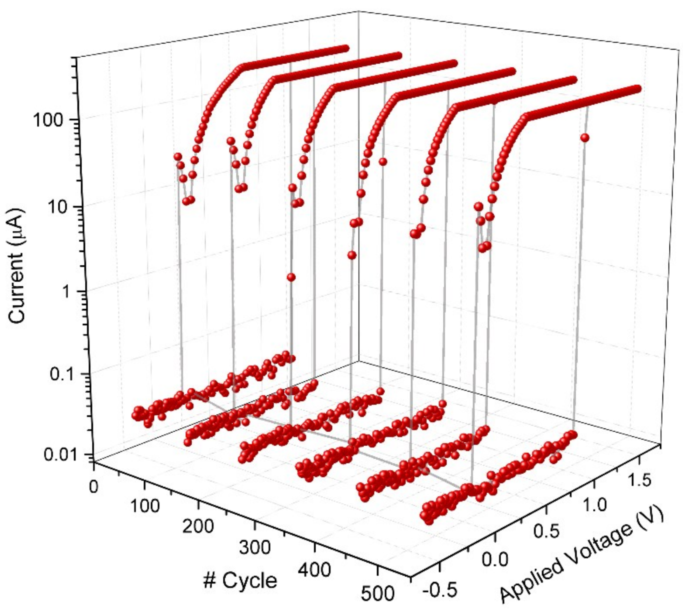


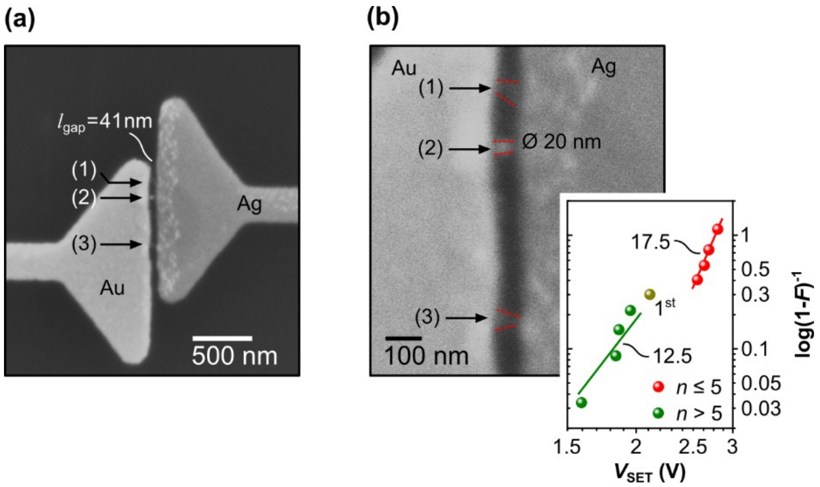
Supplementary Figure S1: Vertical CBRAM cell geometries. (a) Schematic of a vertical Au/SiO_2/Ag device (not to scale). The SiO_2 thickness is 7 nm. The Ag electrode is additionally covered by a Au layer to prevent oxidation (not shown here for simplicity) (b) SEM picture of a vertical device with $A_{Cell} = 100 \cdot 100$ nm^2 overlapping electrode area (switching area).



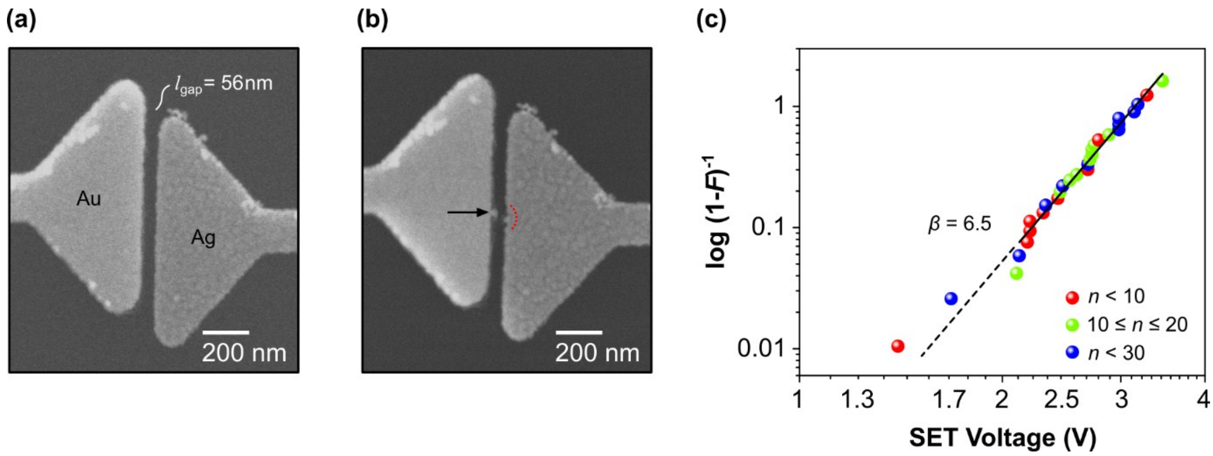
Supplementary Figure S2: Nanoparticle identification using the software ImageJ. (a) Original TEM image. (b) Manual contrast optimization and removal of electrode areas. (c) Fast Fourier transform (FFT) bandpass filter (we found best results for: Filter for small structures 3 … 7 px and for large structures 30 … 70 px, tolerance 5 %, automatic saturation). (d) Re-optimized contrast and manual Gamma correction. (f) 1-Bit threshold filter (manually adjusted), removal of electrode artefacts and particles which were visible in the pristine device. (g) Identification of particles using the "Analyze Particles" functionality of ImageJ. A pixel threshold is used to identify only particles with a diameter larger than ≈ 1 nm.[1] We assume spherical particles for simplification (although some particles differ from spheres). The next nearest neighbour distance is calculated by an ImageJ plugin by Y. Mao.



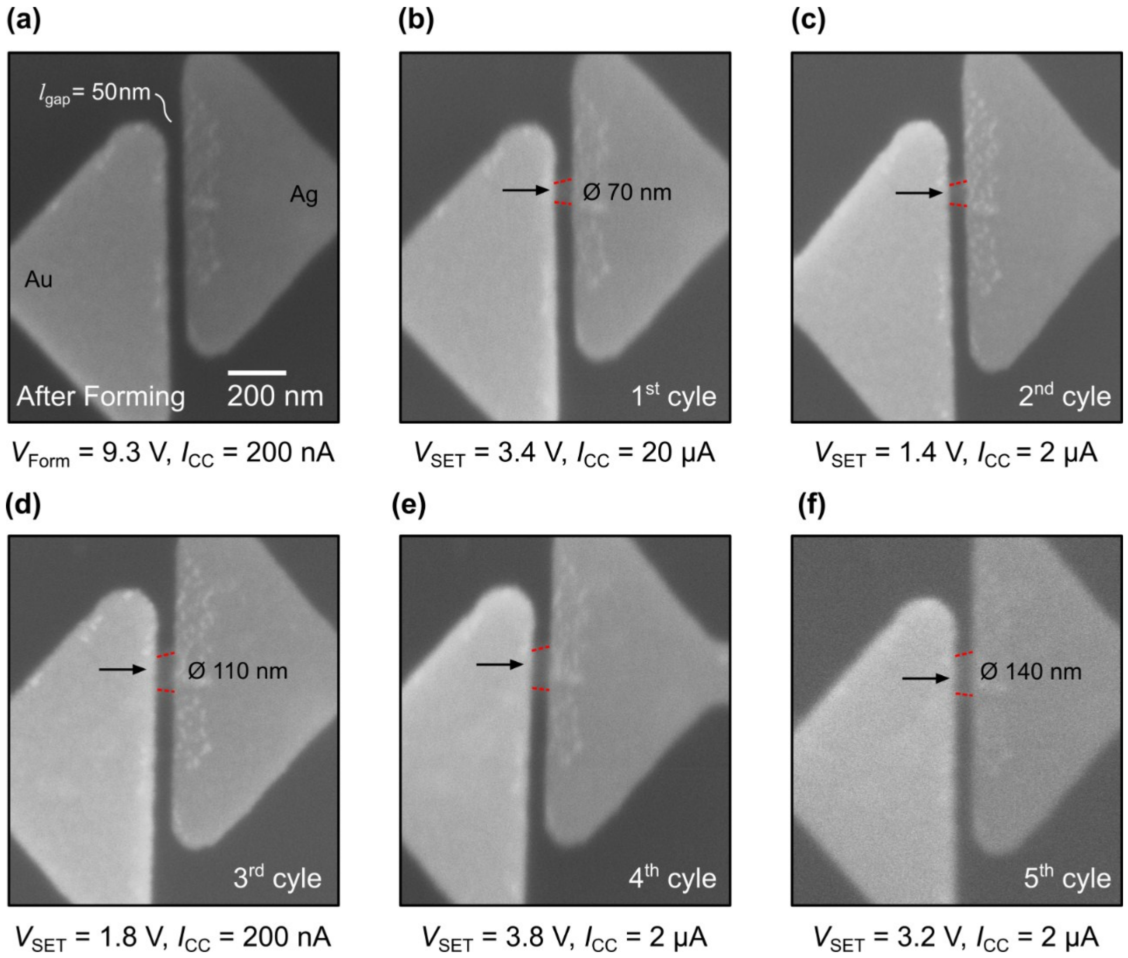
Supplementary Figure S3: Resistive switching of a vertical Au/SiO_2/Ag crossbar device shown in Supplementary Figure S1. Here, a current compliance of I_{CC} = 200 µA is used. For readability only every 100th cycle is shown.



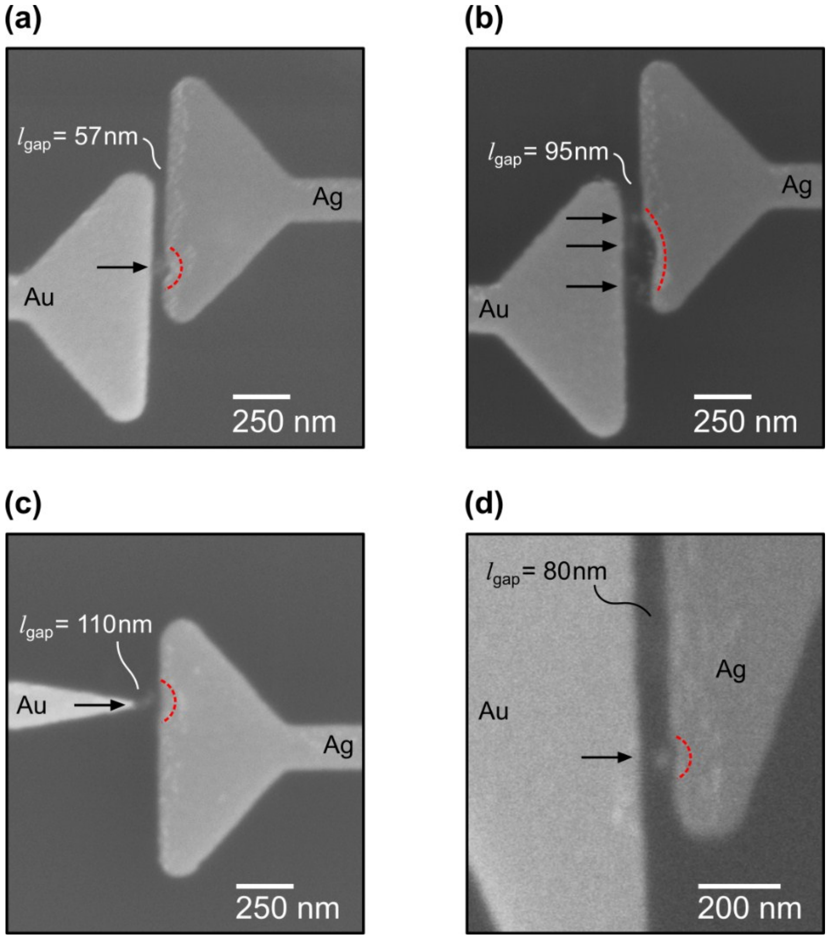
Supplementary Figure S4: Resistive switching of a lateral Au/PMMA/Ag device. (a) SEM picture after 10 switching cycles with up to three different filaments. (b) Zoom into the gap region of (a). The most dominating filament (by contrast) has a diameter of about 20 nm. The Weibull statistics with at least two different slopes is shown in the inset.



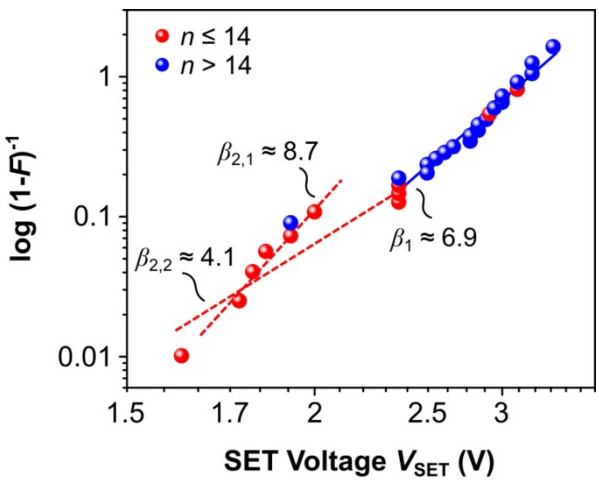
Supplementary Figure S5: Resistive switching of a lateral Au/SiO_2/Ag device. SEM picture of the device before switching (a) and after 30 cycles and a subsequent switch off the device (b). (c) The Weibull statistics can be fitted to a single slope of β = 6.5 for all cycles n.



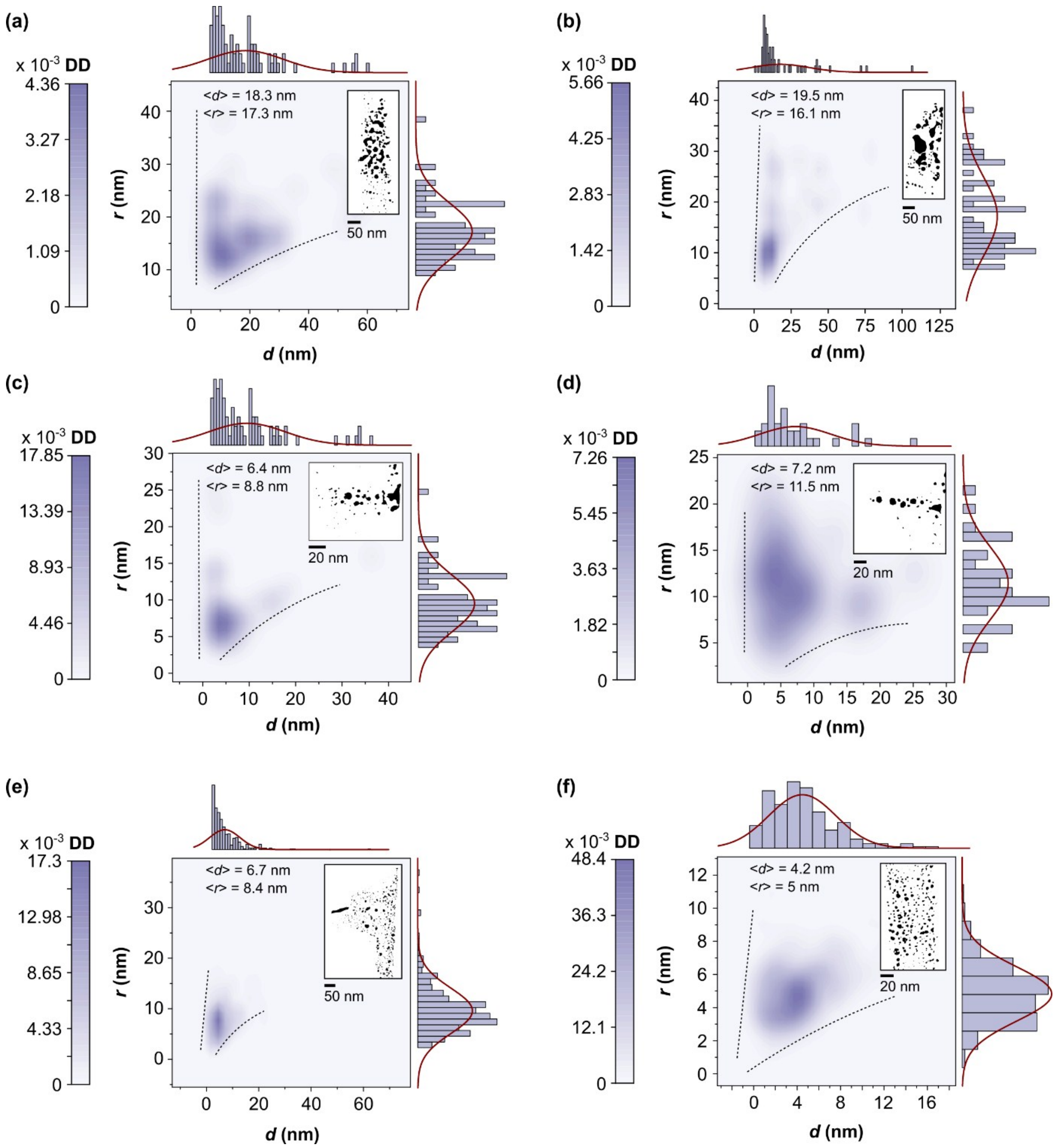
Supplementary Figure S6: Formation of a conductive filament in a Ag/PMMA/Au lateral device (a) – (f). The filament diameter is increasing from about 70 nm to 140 nm upon switching regardless of the current compliance used.



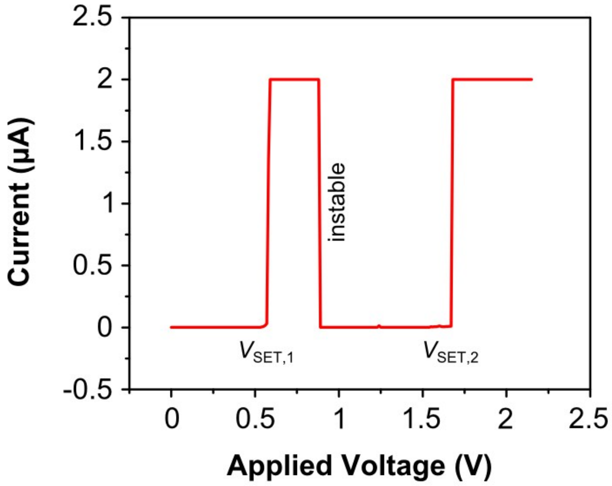
Supplementary Figure S7: Examples of consumption of the active electrode material during resistive switching (a) – (d) of lateral Ag/PMMA/Au devices.



Supplementary Figure S8: Weibull statistics of the SET voltage of the Ag/SiO_2/Pt based device from Figure 3 (with n = 30 cycles). Two slopes can be identified β_1 and $\beta_{2,x}$ (with x = 1, 2). However, definition of the exact slope for cycles $n \leq 14$ is difficult and two potential slopes $\beta_{2,1}$ and $\beta_{2,2}$ are shown here.



Supplementary Figure S9: Data density (DD) plots of the particle diameter *d* and next nearest neighbour distance *r* for several Ag/SiO_2/Au lateral devices patterned on Si_3N_4 windows. Average values for <*d*> and <*r*> (and their standard deviation, not shown here) are calculated from a Gaussian distribution fit of the histograms. The insets depict the black/white TEM images for particle identification. (a), (b) Data density plot for the TEM images shown in Figure 3c (after cycle 30) and 3g (after cycle 31), respectively. (c), (d) Data density plot for the TEM images shown in Figure 3d (after cycle 30) and 3h (after cycle 31), respectively. (e) DD plot of a device with large gap distance l_{gap} = 255 nm. The device could not be switched to a low resistive ON state but nevertheless particles are present in the gap after voltage stress (up to 20 V). (f) Another lateral device with a large area of filament clusters.



Supplementary Figure S10: Instable resistive switching of a lateral Ag/SiO_2/Au device. The device is switched ON to the LRS at $V_{SET,1} \approx 0.6$ V. While still applying a positive voltage to adjust the current to reach the current compliance $I_{CC} = 2$ µA the device becomes instable and switches OFF at ≈ 0.9 V. At $V_{SET,2} \approx 1.7$ V the device is again switched ON.

References

(1) Medasani, B.; Park, Y. H.; Vasiliev, I. Theoretical Study of the Surface Energy, Stress, and Lattice Contraction of Silver Nanoparticles. *Phys. Rev. B* **2007**, *75*, 235436.