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Designing carbon conductive filament memristor devices for memory and electronic synapse applications

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Fig. S1. TEM image and film thickness.



Fig. S2. TEM images and defect analysis. (a) Cross-sectional TEM compositional images for GDs. (b) Zoom regions in a) marked by the red frame. (c) Raman spectrum of a graphene edge. The laser excitation was at 514 nm. D peak at 1376 cm⁻¹, G peak at 1581 cm⁻¹, 2D peak at 2701cm⁻¹. (d) -(f) The atomic intensity of graphene layer collected from (b), (d) extracted from the red line, e) extracted from the blue line, (f) extracted from the saffron yellow line.



Fig. S3. Electroforming process for devices. (a) I-V forming curve of GLDs. (b) I-V forming curve of GDs.



Fig. S4. Resistance versus switching power of different GLDs and GDs, Triangles denote R_{ON} and the corresponding

 $P_{RESET,}$ while box denote R_{OFF} and the corresponding $P_{SET_{\circ}}$ Blue: GLDs; Red: GDs.

Device Structure	Filament Type	SET /RESET (V)	SET /RESET Power(W)	Journal
TiN/AlN/Graphene/Pd	С	0.16/-0.32	10-8/10-4	This Work
Ag/SiGe/p-Si	Ag	3.6~4.1/-2	10-7/10-4	Nat. Mater. 2018, 17, 335-340.
Ag/TiO2:Ag/Pt	Ag	0.2/-0.3	10-4/10-3	Adv. Funct. Mater. 2018, 28, 1705320.
Ag/NiO/Pt	Ag	0.5/-0.6	10-5/10-4	ACS Appl. Mater. Interfaces 2018, 10, 24598.
Ag/SiO ₂ /Pt	Ag	0.36~2.53/ -0.42~-1.64	10-5/10-2	ACS Appl. Mater. Interfaces 2017, 9, 34064.
Ag/SiO ₂ /Pt	Ag	4 /-2	10-6/10-2	Adv. Funct. Mater. 2014, 24, 5679.
Ag/amorphous TiO ₂ /Pt	Ag	2.3/ -2.3	10-5/10-4	Adv. Mater. 2017, 29, 1701752
Ag/a-LSMO/Pt	Ag	0.5/-0.25	10-3/100	ACS Appl. Mater. Interfaces 2013, 5, 11258.
Ag/AgInSbTe/Ta	Ag	-0.8/0.4	10-5/10-4	Adv. Electron. Mater. 2015, 1, 1500125.
Cu/AlN/TiN	Cu	1.5/-0.8	10-5/10-4	ACS Appl. Mater. Interfaces 2013, 5, 1793-1799.
Cu/AlN/Pt	Cu	2.25/-0.75	10-5/10-4	ACS Appl. Mater. Interfaces 2013, 5, 1793-1799.
Cu/AlN/Pt	Cu	2.75/-0.8	10-5/10-3	Appl. Phys. Lett. 2010, 97, 083502.

Table S1. Comparison of other devices parameters with Ag and Cu CFs.



Fig. S5. Resistances change before and after electrode cutting. Insets show the cross section of cells with conductive filaments.



Fig. S6. I-V curve and linear fit (a)The typical I–V curve of GDs. Red circle was LRS. (b) Ln *I* vs. Ln *V* plots of the *I-T* characteristics obtained from(a).



Fig. S7. Element cumulative probability for switching region of AlN layer at LRS. Cumulative probability is abbreviated as CP.



Fig. S8. HRTEM images for deformations region. (a) Cross-sectional HRTEM compositional images for deformations region. After electrical operations, showing deformations likely caused by gas bubble formation at AIN/Graphene interface. (b) Enlarged TEM images are taken from the red rectangular region in (a).



Fig. S9. Schematic of Pulse stimulation. (a) -(d) The pulse schemes. (a) The positive and negative pulses correspond to the potentiating and depressing pulses, respectively. (b) Pulse modulation with different intervals, widths, amplitudes. (c) the waveform used for PPF measurement with different interval (Δt). (d) Pulse schemes for the

asymmetric Hebbian learning rule.



Fig. S10. A pair of presynaptic spikes and the EPSC triggered by two spikes with an inter-spike interval time of 10 μ s. A1 and A2 represent the amplitudes of the first and second EPSCs, respectively. The PPF index, defined as the ratio of A2/A1.