

Supplementary Information

Inkjet-printed h-BN memristors for hardware security

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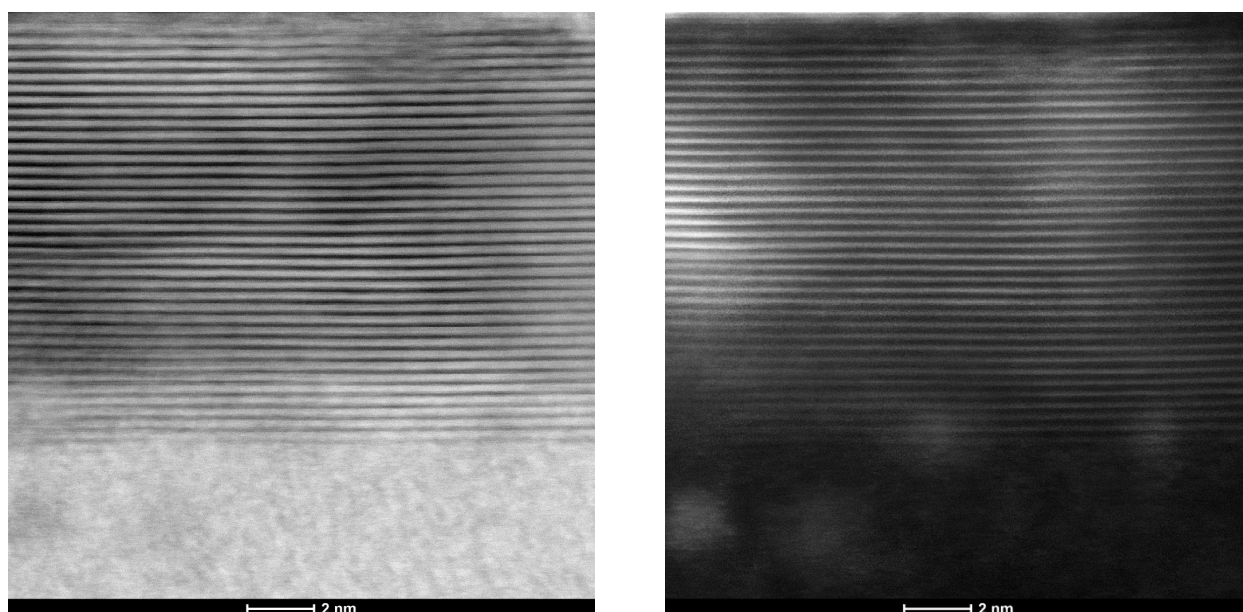
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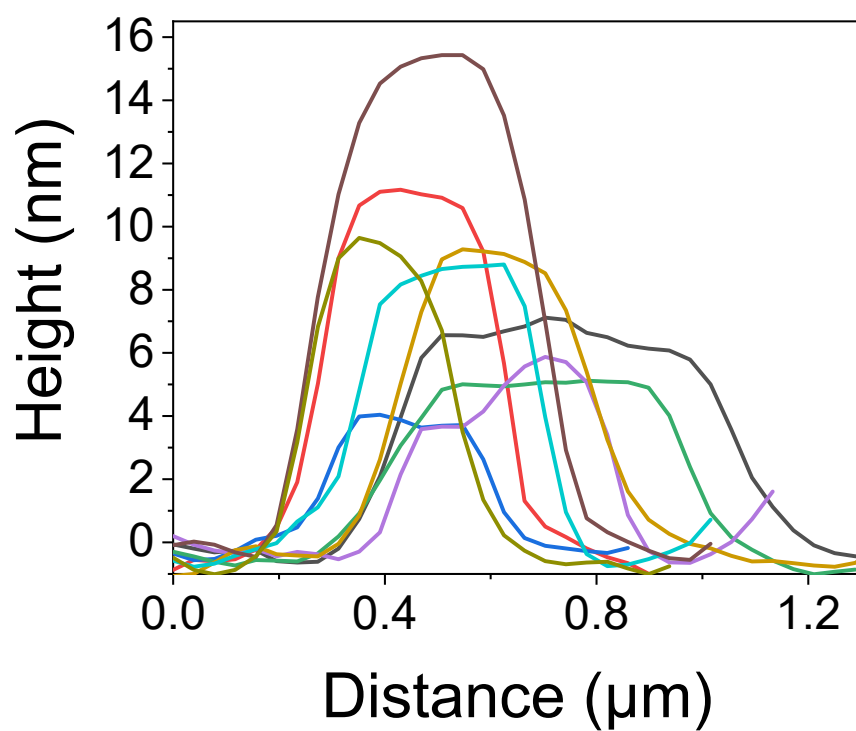
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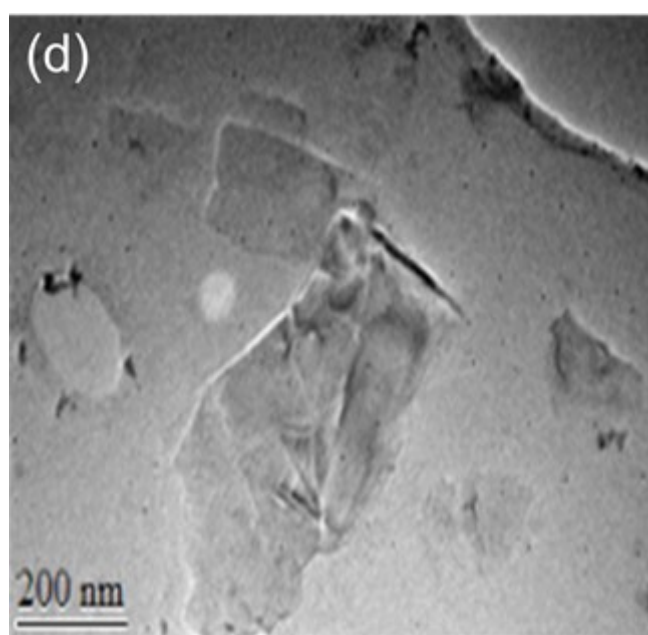
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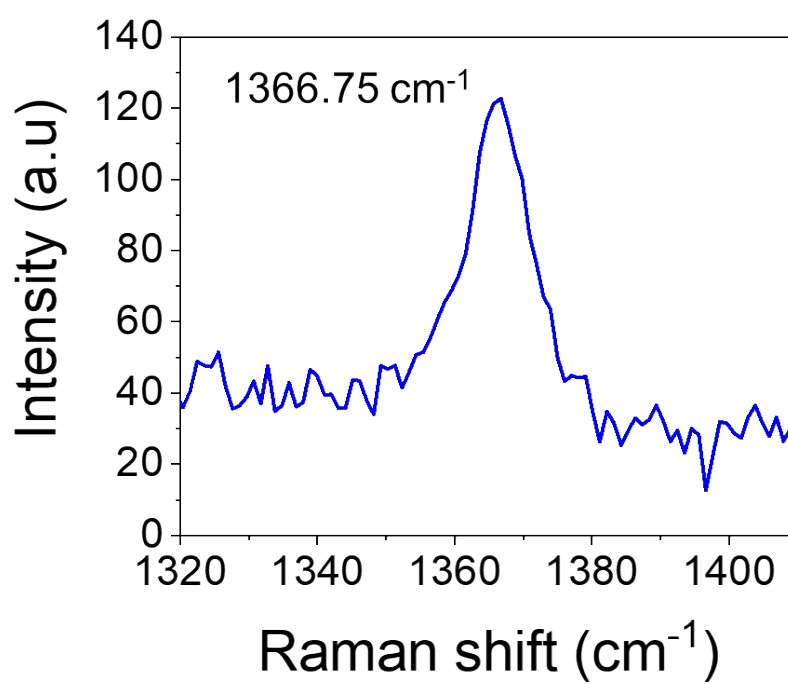
Supplementary Figure 1. The cross-sectional scanning TEM (STEM) images of the same h-BN nanoflake in bright field (BF) mode and high-angle annular dark field (HAADF) mode. In BF image (left), the black lines represent h-BN. In HAADF mode, the white lines represent h-BN.



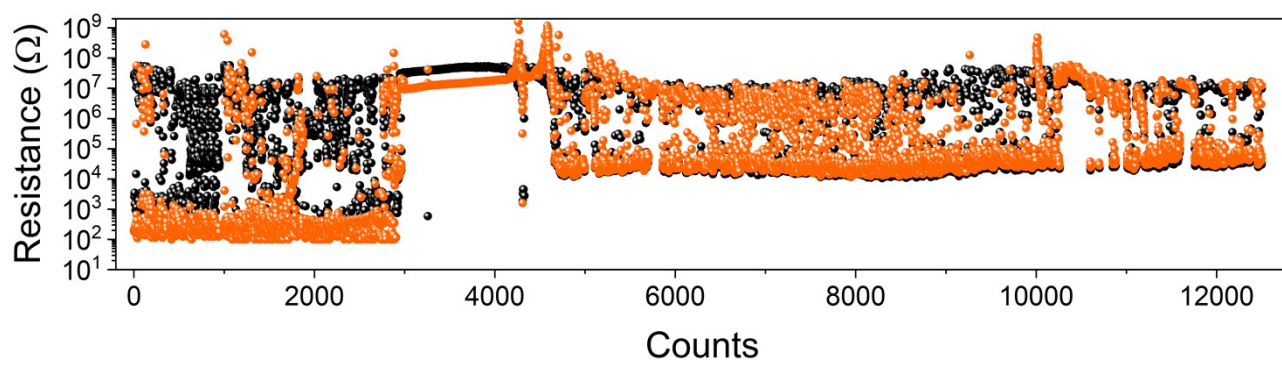
Supplementary Figure 2. Statistical analysis of the thickness and size of the h-BN nanoflakes by AFM profiles from Figure 1c.



Supplementary Figure 3. Top view TEM image of h-BN nanoflakes.



Supplementary Figure 4. Raman spectra of the inkjet-printed h-BN thin film.



Supplementary Figure 5. Endurance test of Ag/h-BN/Ag memristor showing self-recovery.

Supplementary Table 1: LPE BP based memristor

| Ref | TE/RS medium/BE | Print method | Endurance | data points presented |
|-----|-----------------------|------------------------|-----------|-----------------------|
| 1 | Al/BP/ITO | Drop-casting | - | - |
| 2 | Al/PMMA/BPQDs/PMMA/Al | Sonication exfoliation | 100 | 21 |
| 3 | Al/BP:PS/Al | Centrifuge | - | - |

Supplementary Table 2: LPE GO based memristor

| Ref | TE/RS medium/BE | Print method | Endurance | data points presented |
|-----|----------------------------------|-----------------------|-----------------|-----------------------|
| 4 | Cu/GO/Pt | Vacuum filtration | 100 | 100 |
| 5 | Al/GO-PVK/ITO | Toluene-cast | 10 | 9 |
| 6 | Al/G-O film/Al | Spin-casting | 100 | 21 |
| 7 | Al/GO/ITO | Spin coating | 100 | 50 |
| 8 | Al/TPAPAM-GO/ITO | Spin or blade coating | 5 | - |
| 9 | Al/GO/ITO | Spin coating | 100 | 50 |
| 10 | Al/RGO/ITO | Drop casting | hundreds | 8 |
| 11 | Cu/GO/Pt | Vacuum filtration | - | - |
| 12 | Pt/rGO-th/Pt | Vacuum filtration | 350 | 350 |
| 13 | Al/FPA-rGO/ITO | Spin coating | 1000 | - |
| 14 | Ag/GO/Ag | Spin coating | - | - |
| 15 | Al/GO/ITO | Spin coating | 100 | 100 |
| 16 | Al/GO/p-Si | Spin coating | 100 | 50 |
| | Al/GO/p-Ge | | 100 | 50 |
| 17 | Pt/GO/ITO | Spin coating | 100 | 21 |
| 18 | Al/rGO/Al | Spin coating | ~100 | 50 |
| 19 | Ag/HfO _x /LSG | Laser-scribing | 100 | - |
| 20 | Ag/GO/ITO | Spin coating | - | - |
| 21 | Al/CuO/GO/CuO/Al | Spray-coating | - | - |
| 22 | Au/GO/ITO | Spin coating | 100 | 9 |
| 23 | ITO/RGO/ITO | Dip coating | 10 ⁵ | - |
| 24 | Al/GO/Al | Spin coating | - | - |
| 25 | Al/Au NPs-RGO-PVA/ITO | Spin casting | 50 | - |
| 26 | Ag/GO/ITO | Inkjet printing | - | - |
| 27 | ITO/GO/ITO/PES | Spin coating | - | - |
| 28 | Al/Au NP inserted GO/ITO | Spin coating | 10 ⁴ | - |
| 29 | Ag/GO/rGO/SiO ₂ /p-Si | Spin coating | 100 | - |
| 30 | Au/GO/Au | Drip | - | - |
| 31 | Au/GO/Al | Spin coating | 200 | - |
| 32 | Ag/GO/rGO/Ag | Spin coating | 100 | - |
| 33 | Ag/ZnO-rGO/FTO | Dip coating | - | - |
| 34 | Metal/GO/Metal | Spin coating | 250 | 15 |
| 35 | Pt/Ti/GO/Pt/Ti | Spin coating | 10 ⁴ | - |
| 36 | PEDOT:PSS/GO/PEDOT:PSS | Spin coating | 10 ³ | - |
| 37 | Pt/GO/ZnONR/ZnO/ITO | Drop-casting | 10 ³ | - |
| 38 | Au/GO/Au | Dripping | - | - |
| 39 | Pt/brGO/Pt | Spin coating | - | - |
| 40 | Yarns/RGO/Yarns | Spin coating | 100 | - |
| 41 | Ag/N-GO QDs/Pt | Drop-casting | 50 | - |
| 42 | Ag/RGO/ITO | Drop-casting | - | - |
| 43 | Al/HGO/ITO | Spin coating | 100 | - |
| 44 | Al/GO-TiO ₂ /ITO | Spin coating | 60 | - |

| | | | | |
|----|------------------------|--------------|--------|----|
| 45 | Pt/ZnO-G/Pt | Spin coating | - | - |
| 46 | Ni/PMMA:GO/ITO | Spin coating | 300 | - |
| 47 | Paint/GO/PET | Spin coating | 100 | 15 |
| 48 | Al/GOAu/ITO | Spin coating | 100 | - |
| 49 | LSMO-rGO | - | 10^3 | - |
| 50 | Al/rGO-ZnO HC/PMMA/ITO | Spin coating | 10^3 | - |
| 51 | Al/GO/Al/PES | Spin coating | 100 | - |
| 52 | Al/GO/Au/GO/ITO | Spin coating | - | 20 |

Supplementary Table 3: LPE MoS₂ based memristor

| Ref | TE/RS medium/BE | Print method | Endurance | data points presented |
|-----|---|-------------------------|-----------|-----------------------|
| 55 | Al/MoS ₂ -PVP/(rGO) | Spin coating | - | - |
| 56 | rGO/MoS ₂ /ITO | Spin coating | - | - |
| 57 | Ag/MoO _x /MoS ₂ /Ag | Inkjet, screen printing | 10^4 | 14 |
| 58 | Al/MoS ₂ -MoO _x /Al | Spin coating | 100 | 11 |
| 59 | Au/PMMA/MoS ₂ | Spin coating | 650 | - |
| | QDs/PMMA/FTO | | | |
| 60 | Al/MoS ₂ -UCNPs/ITO | Spin coating | 200 | 40 |
| 61 | Ag/Gr-MoS ₂ /Cu | Hydrothermal process | 500 | - |
| 62 | Ag/MoS ₂ /Ag | aerosol-jet printing | 120 | 25 |
| 63 | Gr/MoS ₂ QDNS/Gr | Spin coating | 50 | - |
| 64 | Pt/MoS ₂ /Ti | Spin coating | 10^7 | 22 |

Reference 64 claims a high endurance 10^7 while only 22 data points are shown, which is not reliable. The correct characterization method of endurance measurement should be one data point per cycle, as explained in M. Lanza et al, Standards for the Characterization of Endurance in Resistive Switching Devices. ACS Nano 2021, 15, 11, 17214–17231.

Supplementary Table 4: LPE h-BN based memristor

| Ref | TE/RS medium/BE | Print method | Endurance | data points presented |
|------|------------------|-----------------|-----------|-----------------------|
| 53 | Ag/h-BN-PVOH/ITO | EHDA printing | 1500 | 33 |
| 54 | Ag/ZnO/BNNSs/Pt | Spin coating | - | - |
| This | Ag/h-BN/Pt | Inkjet printing | 600,000 | 600,000 |
| work | Ag/h-BN/Ag | Inkjet printing | 11,000 | 11,000 |

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