

Electronic Supplementary Information for:

Molecular Precursor-Directed Growth of Nanostructured SnS₂ for Memristive and Neuromorphic Electronics

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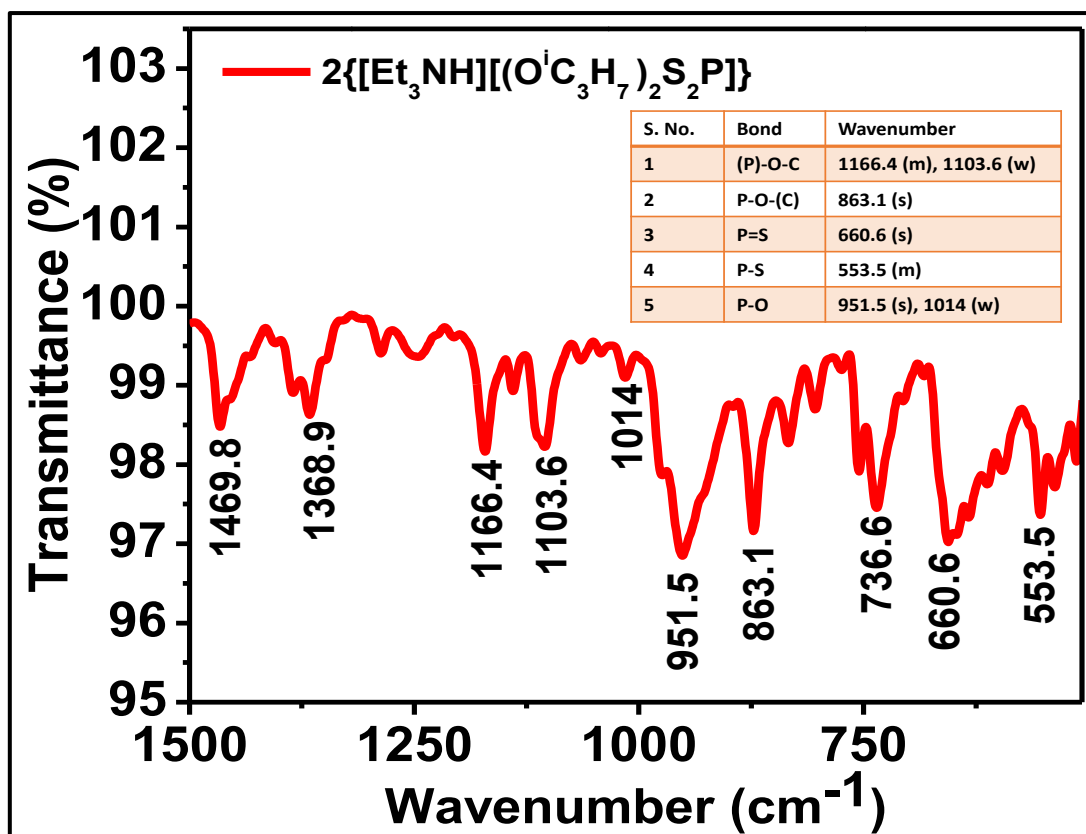


Figure S1. FT-IR spectrum of the [Et₃NH][(OⁱC₃H₇)₂S₂P] using KBr discs

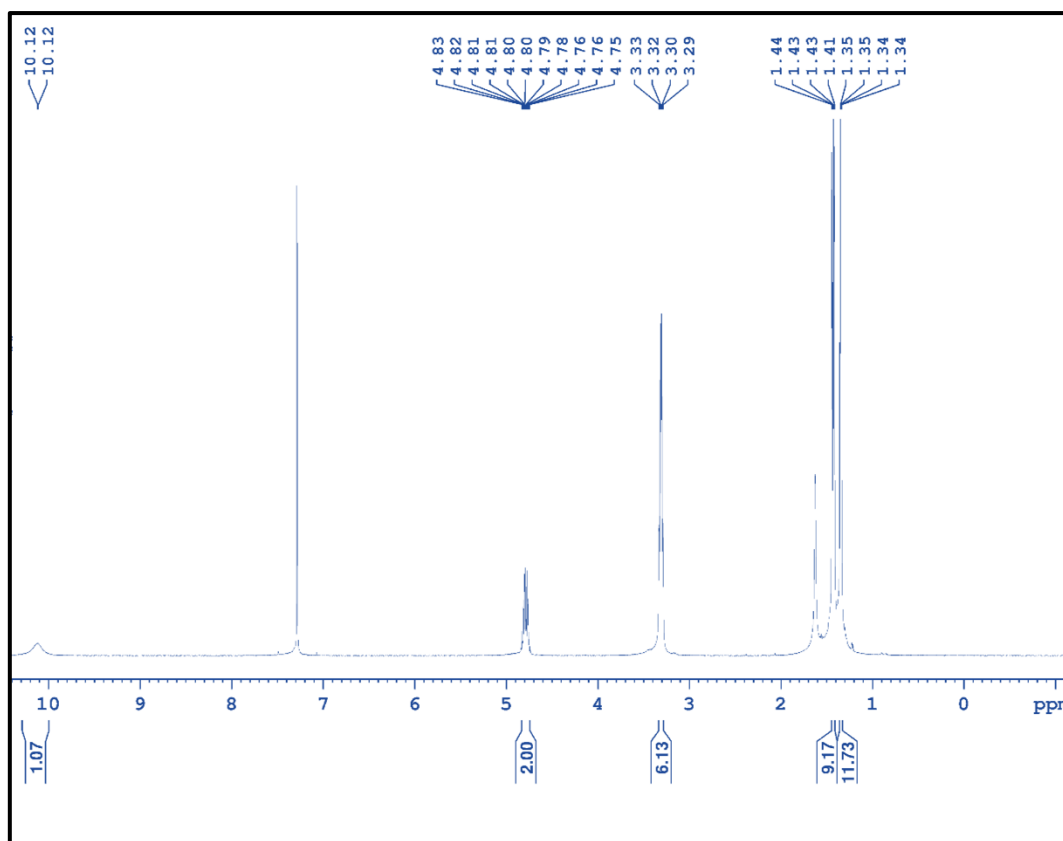


Figure S2. Room temperature $\{^1\text{H}\}$ NMR spectrum of $[\text{Et}_3\text{NH}][(\text{O}^i\text{C}_3\text{H}_7)_2\text{S}_2\text{P}]$ (in CDCl_3).

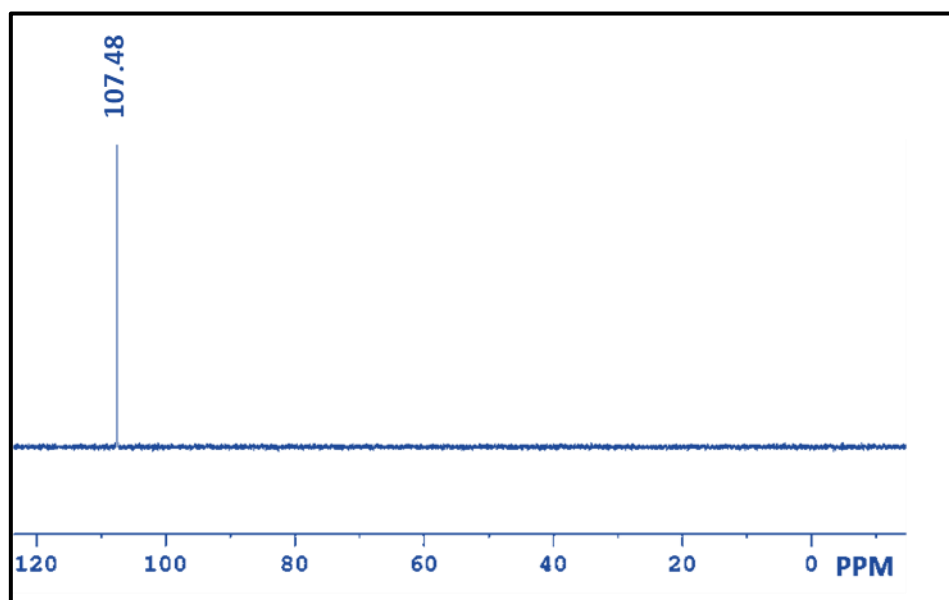


Figure S3. Room temperature $^{31}\text{P}\{^1\text{H}\}$ NMR spectrum of $[\text{Et}_3\text{NH}][(\text{O}^i\text{C}_3\text{H}_7)_2\text{S}_2\text{P}]$ (in CDCl_3).

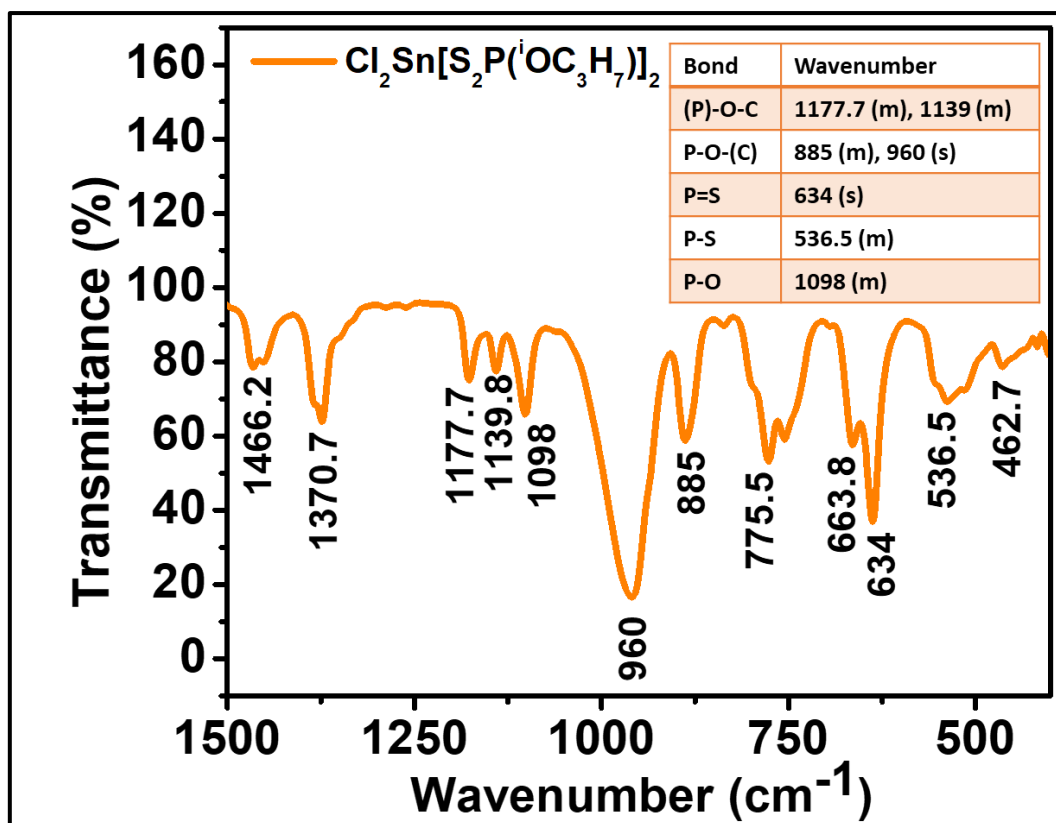


Figure S4. FT-IR spectrum of the $\text{Cl}_2\text{Sn}[\text{S}_2\text{P}(\text{O}^i\text{C}_3\text{H}_7)]_2$ complex using KBr discs

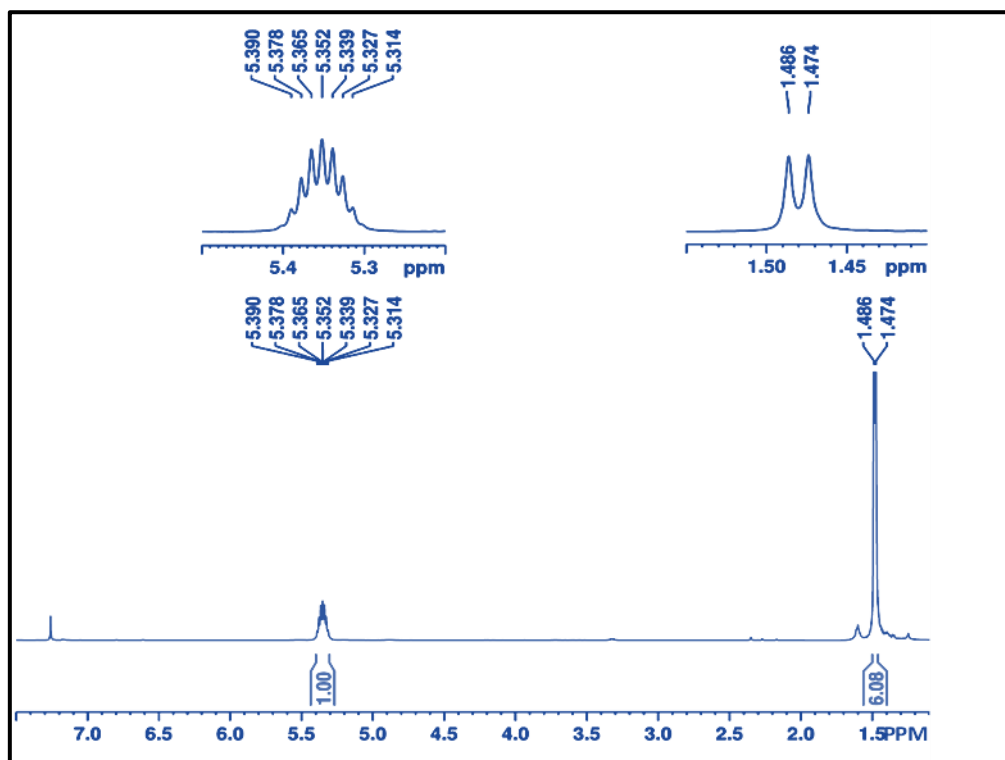


Figure S5. Room temperature $\{^1\text{H}\}$ NMR spectrum of $\text{Cl}_2\text{Sn}[\text{S}_2\text{P}(\text{O}^i\text{C}_3\text{H}_7)]_2$ complex (in CDCl_3).

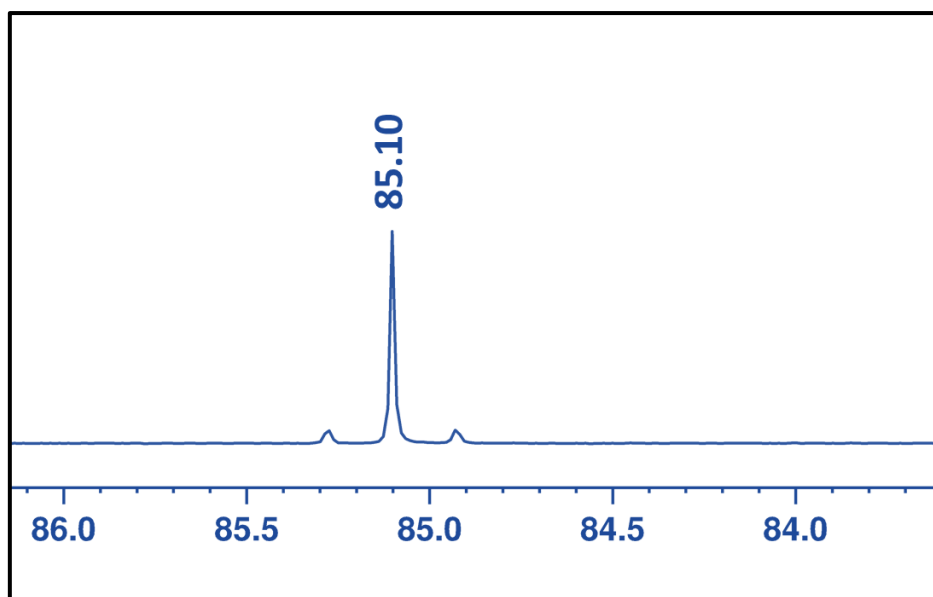


Figure S6. Room temperature $^{31}\text{P}\{^1\text{H}\}$ NMR spectrum of $\text{Cl}_2\text{Sn}[\text{S}_2\text{P}(\text{O}^i\text{C}_3\text{H}_7)]_2$ complex (in CDCl_3).

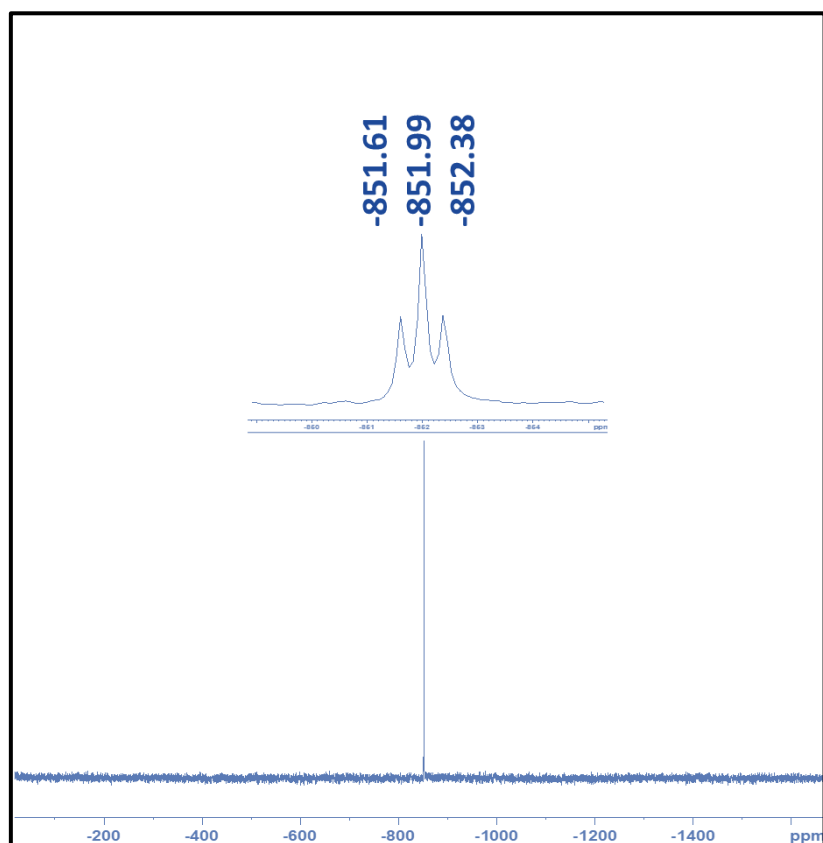


Figure S7. Room temperature $^{119}\text{Sn}\{^1\text{H}\}$ NMR spectrum of $\text{Cl}_2\text{Sn}[\text{S}_2\text{P}(\text{O}^i\text{C}_3\text{H}_7)]_2$ complex (in CDCl_3).

Table S1. Crystal data and structure refinement for [Cl₂Sn[S₂P(OⁱC₃H₇)₂]₂].

| Parameter | Value |
|---|---|
| Empirical formula | C ₁₂ H ₂₈ Cl ₂ O ₄ P ₂ S ₄ Sn |
| Formula weight | 616.11 |
| Temperature (K) | 156.0 |
| Crystal system | Orthorhombic |
| Space group | C2221 |
| a (Å) | 9.9605(7) |
| b (Å) | 18.5085(14) |
| c (Å) | 28.902(2) |
| α (°) | 90 |
| β (°) | 90 |
| γ (°) | 90 |
| Volume (Å ³) | 5328.1(7) |
| Z | 8 |
| Density (ρ _{calc}) (g/cm ³) | 1.536 |
| μ (mm ⁻¹) | 1.606 |
| F(000) | 2480.0 |
| Crystal size (mm ³) | 0.23 × 0.221 × 0.221 |
| Radiation | MoKα (λ = 0.71073 Å) |
| 2θ range for data collection (°) | 4.622 to 51.36 |
| Index ranges | -11 ≤ h ≤ 12, -22 ≤ k ≤ 21, -35 ≤ l ≤ 33 |
| Reflections collected | 39408 |
| Independent reflections | 5069 [R _{int} = 0.0360, Rσ = 0.0231] |
| Data/restraints/parameters | 5069 / 160 / 223 |
| Goodness-of-fit on F ² | 1.202 |
| Final R indexes [I ≥ 2σ(I)] | R ₁ = 0.0387, wR ₂ = 0.0944 |
| Final R indexes [all data] | R ₁ = 0.0401, wR ₂ = 0.0952 |
| Largest diff. peak/hole (e Å ⁻³) | 0.81 / -0.66 |
| Flack parameter | 0.063(6) |

Table S2: Bond lengths (Å) for [Cl₂Sn[S₂P(OⁱC₃H₇)₂]₂

| Atom | Atom | Length (Å) | Atom | Atom | Length (Å) |
|------|------|------------|------|------|------------|
| Sn1 | S2 | 2.5993(15) | P005 | O1 | 1.556(4) |
| Sn1 | S21 | 2.5994(15) | P006 | S3 | 2.022(3) |
| Sn1 | S1 | 2.5138(16) | P006 | O4 | 1.561(5) |
| Sn1 | S11 | 2.5138(16) | P006 | O3 | 1.555(5) |
| Sn1 | Cl1 | 2.3894(19) | O2 | C4 | 1.475(7) |
| Sn1 | Cl11 | 2.3894(19) | O1 | C1 | 1.483(7) |
| Sn2 | S4 | 2.5983(16) | O4 | C10 | 1.472(11) |
| Sn2 | S42 | 2.5983(16) | O3 | C7 | 1.461(9) |
| Sn2 | S32 | 2.526(2) | C7 | C8 | 1.484(17) |
| Sn2 | S3 | 2.526(2) | C7 | C9 | 1.515(14) |
| Sn2 | Cl2 | 2.373(2) | C1 | C2 | 1.521(12) |
| Sn2 | Cl22 | 2.373(2) | C1 | C3 | 1.482(12) |
| S2 | P005 | 2.004(2) | C4 | C5 | 1.496(15) |
| S4 | P006 | 2.003(2) | C4 | C6 | 1.491(15) |
| P005 | S1 | 2.022(2) | C12 | C10 | 1.483(14) |
| P005 | O2 | 1.555(5) | C11 | C10 | 1.459(13) |

Table S3: Bond angles (°) for [Cl₂Sn[S₂P(OⁱC₃H₇)]₂

| Atom | Atom | Atom | Angle/° | Atom | Atom | Atom | Angle/° |
|------------------|------|------------------|------------|------|------|------|------------|
| S2 | Sn1 | S2 ¹ | 88.14(7) | P006 | S4 | Sn2 | 86.52(7) |
| S1 ¹ | Sn1 | S2 ¹ | 78.65(6) | S2 | P005 | S1 | 107.23(9) |
| S1 | Sn1 | S2 ¹ | 92.72(6) | O2 | P005 | S2 | 113.1(2) |
| S1 ¹ | Sn1 | S2 | 92.72(6) | O2 | P005 | S1 | 112.2(2) |
| S1 | Sn1 | S2 | 78.65(6) | O2 | P005 | O1 | 98.4(2) |
| S1 | Sn1 | S1 ¹ | 168.06(9) | O1 | P005 | S2 | 114.0(2) |
| Cl1 | Sn1 | S2 | 90.39(7) | O1 | P005 | S1 | 111.9(2) |
| Cl1 | Sn1 | S2 ¹ | 168.59(6) | S4 | P006 | S3 | 107.06(11) |
| Cl1 ¹ | Sn1 | S2 ¹ | 90.39(7) | O4 | P006 | S4 | 113.0(2) |
| Cl1 ¹ | Sn1 | S2 | 168.58(6) | O4 | P006 | S3 | 112.4(2) |
| Cl1 | Sn1 | S1 ¹ | 90.12(7) | O3 | P006 | S4 | 112.8(2) |
| Cl1 | Sn1 | S1 | 98.09(7) | O3 | P006 | S3 | 112.8(3) |
| Cl1 ¹ | Sn1 | S1 ¹ | 98.09(7) | O3 | P006 | O4 | 98.8(3) |
| Cl1 ¹ | Sn1 | S1 | 90.12(7) | P005 | S1 | Sn1 | 88.02(7) |
| Cl1 | Sn1 | Cl1 ¹ | 93.23(12) | P006 | S3 | Sn2 | 88.08(8) |
| S4 | Sn2 | S4 ² | 89.15(7) | C4 | O2 | P005 | 121.8(4) |
| S3 ² | Sn2 | S4 ² | 78.33(6) | C1 | O1 | P005 | 121.5(4) |
| S3 ² | Sn2 | S4 | 90.92(6) | C10 | O4 | P006 | 122.0(6) |
| S3 | Sn2 | S4 | 78.33(6) | C7 | O3 | P006 | 122.9(5) |
| S3 | Sn2 | S4 ² | 90.92(6) | O3 | C7 | C8 | 107.5(9) |
| S3 | Sn2 | S3 ² | 164.97(10) | O3 | C7 | C9 | 106.1(8) |
| Cl2 ² | Sn2 | S4 | 169.76(9) | C8 | C7 | C9 | 112.4(10) |
| Cl2 | Sn2 | S4 | 90.28(7) | O1 | C1 | C2 | 106.5(7) |
| Cl2 ² | Sn2 | S4 ² | 90.28(7) | C3 | C1 | O1 | 107.6(6) |
| Cl2 | Sn2 | S4 ² | 169.76(9) | C3 | C1 | C2 | 113.0(7) |
| Cl2 | Sn2 | S3 | 98.98(9) | O2 | C4 | C5 | 106.5(7) |
| Cl2 ² | Sn2 | S3 | 91.46(10) | O2 | C4 | C6 | 107.9(7) |
| Cl2 ² | Sn2 | S3 ² | 98.98(9) | C6 | C4 | C5 | 112.4(8) |
| Cl2 | Sn2 | S3 ² | 91.46(10) | O4 | C10 | C12 | 107.0(9) |
| Cl2 | Sn2 | Cl2 ² | 92.09(13) | C11 | C10 | O4 | 108.2(9) |
| P005 | S2 | Sn1 | 86.05(7) | C11 | C10 | C12 | 114.2(10) |

Table S4. Comparative study of SSPs utilized for the deposition of SnS₂ thin films.

| Precursor | Synthesis Method and Deposition Condition | Need for Additional Sulphur Source | Sulphur Deficient Tin Sulphide | Device Integration | Ref. |
|---|---|------------------------------------|--------------------------------|-------------------------|-----------|
| ⁿ Bu ₂ Sn(SnBu) ₂ | AACVD + Annealing (440–460 °C) | No | Yes | Na-ion Battery | 1 |
| SnCl ₄ { ⁿ BuS(CH ₂) ₃ S ^m Bu} | LPCVD (650 °C) | No | No | Not explored | 2 |
| ⁿ Bu ₂ Sn(SnBu) ₂ | LPCVD (440–530 °C) | No | No | Thermoelectric | 3 |
| Sn(SCH ₂ CF ₃) ₄ | APCVD (≥300 °C) | Yes | No | Not explored | 4 |
| Sn(S ₂ CNEt ₂) ₄ | CVD (250–350 °C) | Yes | No | Not explored | 5 |
| R _n Sn[S ₂ CN(C ₄ H ₉)CH ₃] _{4-n} | APCVD+ Annealing | Yes | No | Not explored | 6 |
| This work (DTP-based SSP) | One-step solvothermal (200 °C) | Not required | No | Memristor+ Neuromorphic | This work |

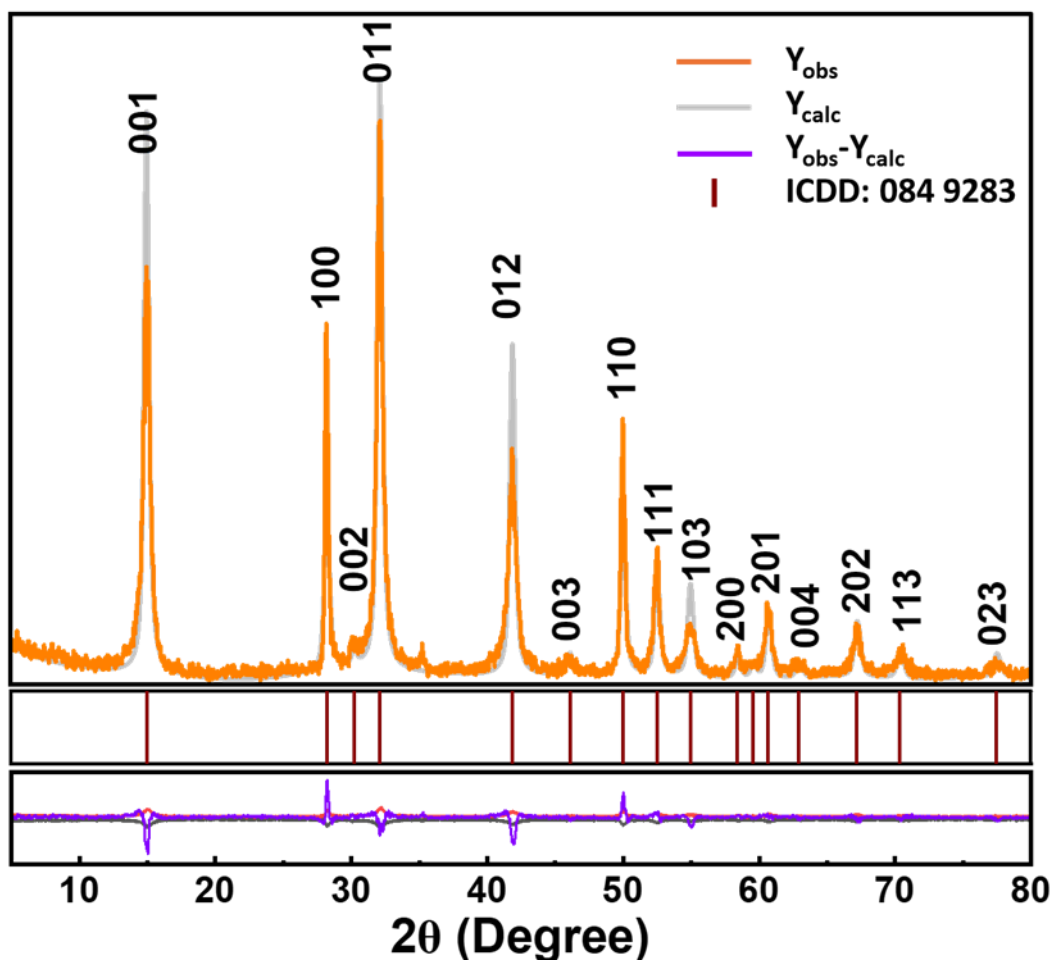


Figure S8: Rietveld refinement pattern of nanostructured SnS₂ thin film

Table S5: Microstrain (ϵ) values estimated from XRD peak broadening of SnS₂ thin film.

| Peak (2θ) | FWHM (deg) | Microstrain (ϵ) |
|--------------------|------------|----------------------------|
| 15.02 | 0.40422 | 0.0134 |
| 28.27 | 0.28947 | 0.0050 |
| 32.13 | 0.37345 | 0.0056 |
| 41.89 | 0.50812 | 0.0058 |
| 50.01 | 0.21850 | 0.0020 |

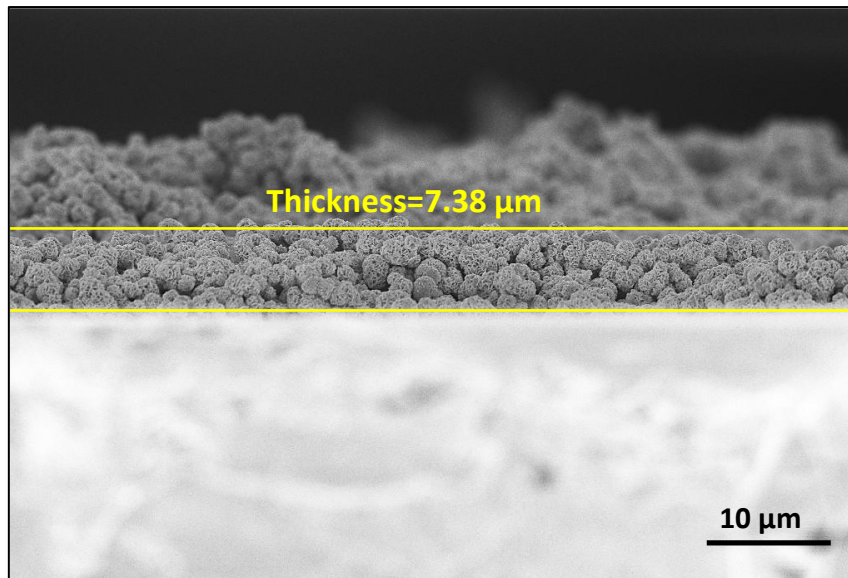


Figure S9. Cross-sectional micrograph of the SnS₂ nanostructured thin film, highlighting the film thickness

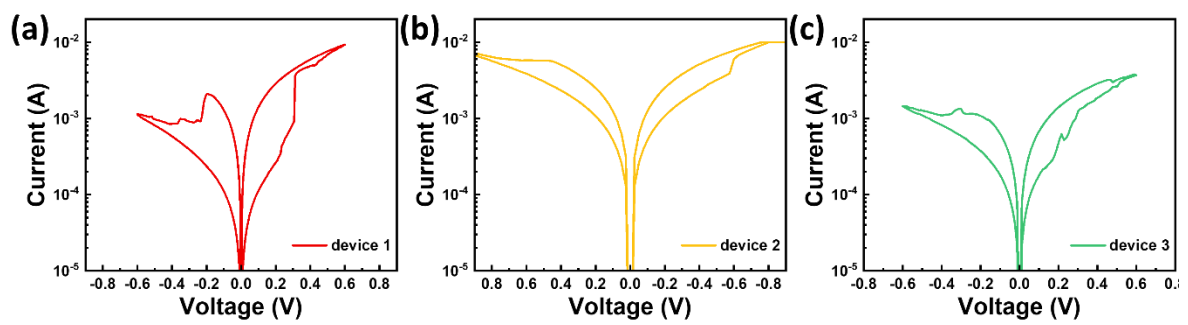


Figure S10 DC I–V characteristics of three representative Ag/SnS₂/ITO memristor devices, showing bipolar resistive switching behaviour with electroforming-free operation.

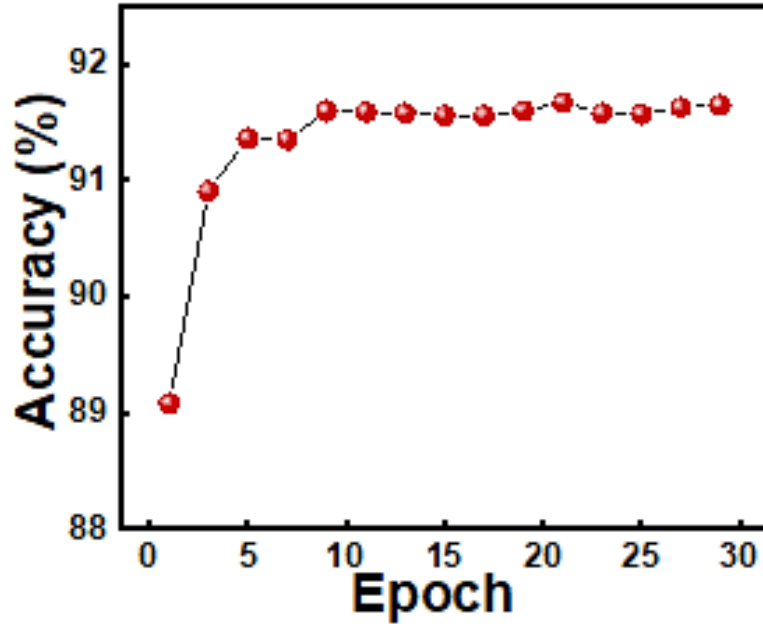


Figure S11. Validation accuracy as a function of training epoch.

Table S6. Comparison of the resistive switching performance of the Ag/SnS₂/ITO memristor in this work against other SnS₂-based memristors.

| Device | Steps in Synthesis | Temperature | Switching Voltage | Electro-forming | Synaptic Switching | MNIST Accuracy | Ref. |
|--|--------------------|-------------|-------------------|-----------------|--------------------|----------------|-----------|
| SnO _x /SnS ₂ Heterostructure | Multistep | 250 °C | ~1 V | Yes | Yes | 90% | 7 |
| SnS ₂ Quantum Dots | Multistep | 160–200 °C | 0.7-1 V | Yes | No | Not explored | 8 |
| SnS ₂ -rGO Composite | Multistep | 180–200 °C | 2 V | Yes | Yes | Not explored | 9 |
| Ca ²⁺ -doped SnS ₂ | Multistep | 180–220 °C | 2-4 V | Yes | Yes | 90% | 10 |
| Ag/SnS ₂ /ITO (This work) | Multistep | ~200 °C | 0.6 V | No | Yes | 92% | This work |

References:

- 1 Z. Zhu, G. Hyett, G. Reid, F. Robinson, G. Cibin and A. L. Hector, *Small Struct.*, 2023, **4**, 2200396.
- 2 C. Gurnani, S. L. Hawken, A. L. Hector, R. Huang, M. Jura, W. Levason, J. Perkins, G. Reid and G. B. G. Stenning, *Dalton Trans.*, 2018, **47**, 2628–2637.
- 3 F. Robinson, P. J. Curran, C. H. De Groot, D. Hardie, A. L. Hector, K. Holloway, R. Huang, D. Newbrook and G. Reid, *Mater. Adv.*, 2021, **2**, 4814–4823.
- 4 G. Barone, T. G. Hibbert, M. F. Mahon, K. C. Molloy, L. S. Price, I. P. Parkin, A. M. E. Hardy and M. N. Field, *J. Mater. Chem.*, 2001, **11**, 464–468.
- 5 G. Barone, T. Chaplin, T. G. Hibbert, A. T. Kana, M. F. Mahon, K. C. Molloy, I. D. Worsley, I. P. Parkin and L. S. Price, *J. Chem. Soc. Dalt. Trans.*, 2002, 1085–1092.
- 6 A. T. Kana, T. G. Hibbert, M. F. Mahon, K. C. Molloy, I. P. Parkin and L. S. Price, *Polyhedron*, 2001, **20**, 2989–2995.
- 7 Y. Wu, S. Li, Y. Ji, Z. Weng, H. Xing, L. Arauz, T. Hu, J. Hong, K. W. Ang and S. Liu, *Sci. China Mater.*, 2025, **68**, 581–589.
- 8 H. An, Y. Li, Y. Ren, Y. Wan, W. Wang, Z. Sun, J. Zhong and Z. Peng, *Nanoscale*, 2024, **16**, 12142–12148.
- 9 E. K. Jang, Y. Park and J. S. Lee, *Nanoscale*, 2019, **11**, 15382–15388.
- 10 J. Feng, J. Fan, Z. Zhang, Y. Gao, S. Xue, G. Cai and J. S. Zhao, *Adv. Funct. Mater.*, 2024, **34**, 2401228.