

## Supporting Information

### **Emulating Frog's behavior: Humidity-Driven Paired-Pulse Facilitation and Metaplasticity realised in a 1D Supramolecular Nanofibre based neuromorphic device**

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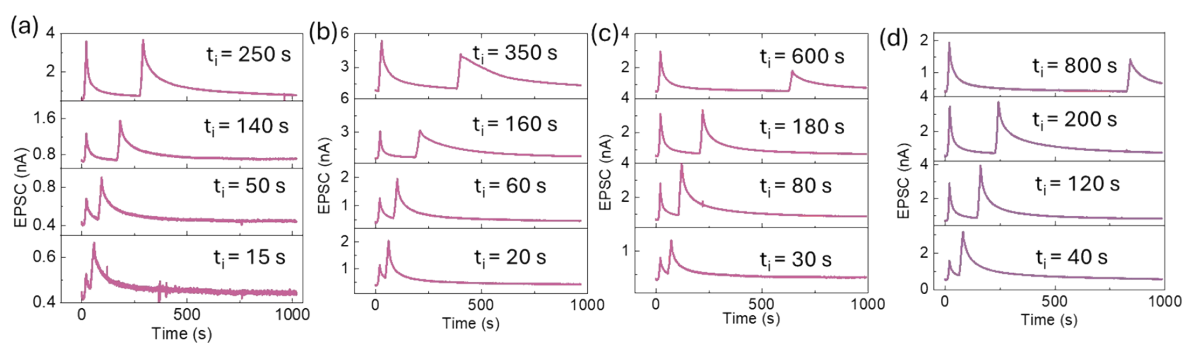
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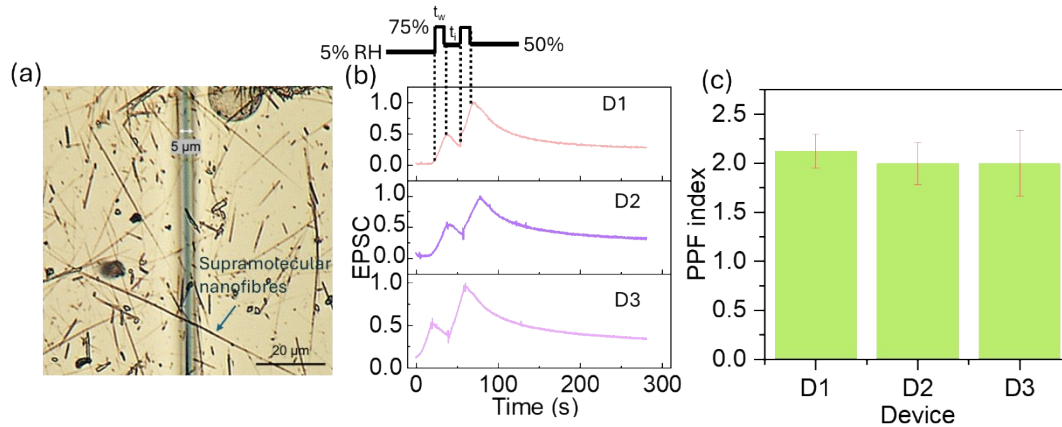
Device geometry	Lateral or Vertical (L/V)	Organic or Inorganic (O/I)	Dimensionality (0/1/2/3D)	Stimuli Electrical-E Optical-O Humidity-H Pressure-P	Tunability of PPF	Multi stimuli operation	Metaplasticity	Ref
graphene/h BN/CsPbBr <sub>3</sub> QDs	L	I	0D-2D heterostructure	O, E	×	×	×	1
Au/Gelatin-PEDOT: PSS/Al	V	O	2D	E, H	×	✓	×	2
Ag/CsPbBr <sub>3</sub> /PVA/FTO	V	I	2D	E, P	×	×	×	3
Au/CuPc/Au	L	O	2D	O, E	×	×	×	4
Au/ZnO/Au	L	I	2D	O	×	×	×	5
Au/P3HT/ITO	V	O	2D	O	×	×	×	6
ITO/(R/S-MBA) <sub>2</sub> PbI <sub>4</sub> /PMMA/Al	V	Hybrid	2D	O, E	✓	×	×	7
Ag/CdPS <sub>3</sub> /ITO	V	I	2D	E	×	×	×	8
Au/C8-BTBT: P3HT/Au	L	O	2D	O, E	×	×	×	9
This work	L	O	1D	O, H	✓	✓	✓	

**Table S1- Comparison of two terminal neuromorphic devices and their potential to exhibit tunable PPF, multi stimuli operation, and metaplasticity**

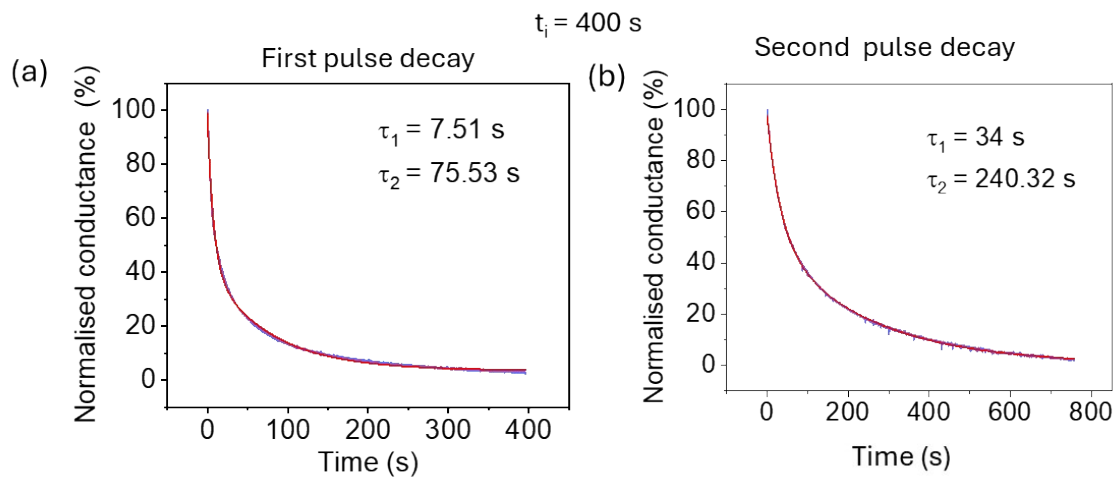


**Figure S1.** (a)-(d) Response of the nanofibres with two 75% RH pulses ( $t_w = 15$  s) where  $t_i$  was varied from 15 s to 800 s and monitored at 40% RH.

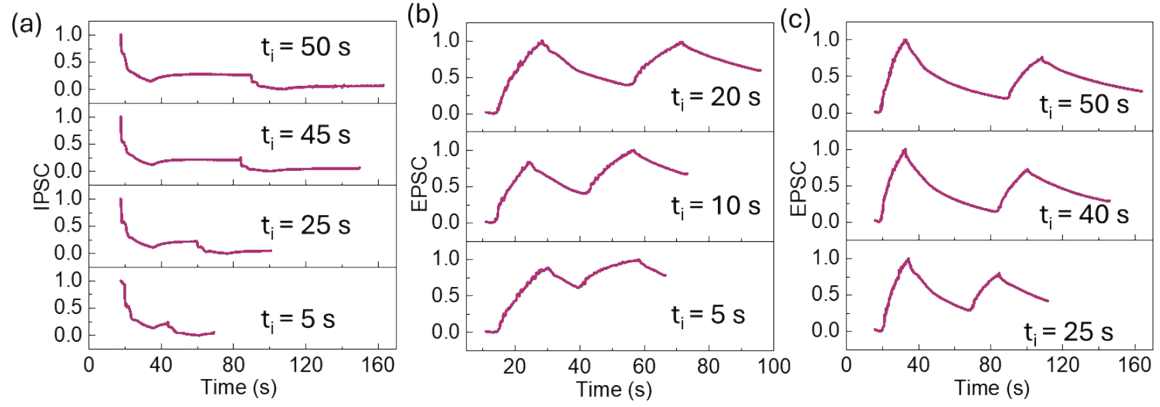
Figure S1 shows the remaining representative response curves for different  $t_i$ , utilised for PPF calculation in Figure 2c. Considering  $t_i = 20$  s (Figure S1b), current increases from 0.4 nA ( $A_0$ ) to 1.1 nA with the application of the first RH pulse (75%) and finally increases to 2 nA with the second RH pulse, showing facilitation with a PPF index of 2.2.



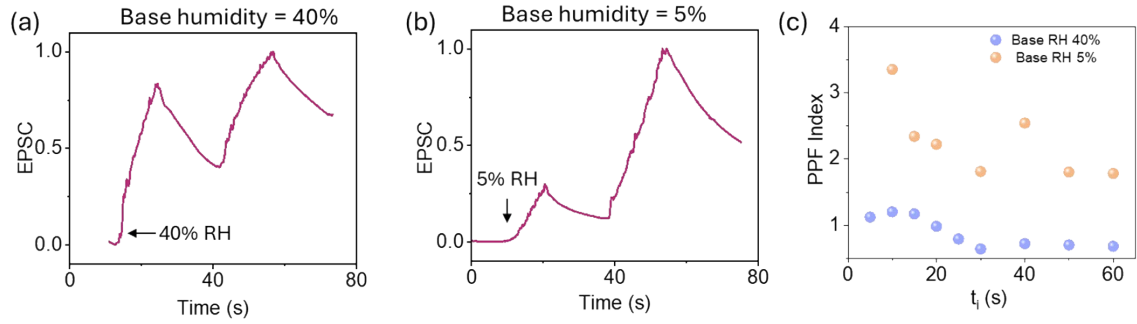
**Figure S2. Device-to-device variability.** (a) Optical image of the nanofibre device with nanofibres spread across a 5 µm gap Ti electrodes. (b) Response to two 75% RH pulses with  $t_w = t_i = 15$  s for 3 different devices. (c) Average PPF index achieved for 3 devices, with error bars standing for 5 measurements each.



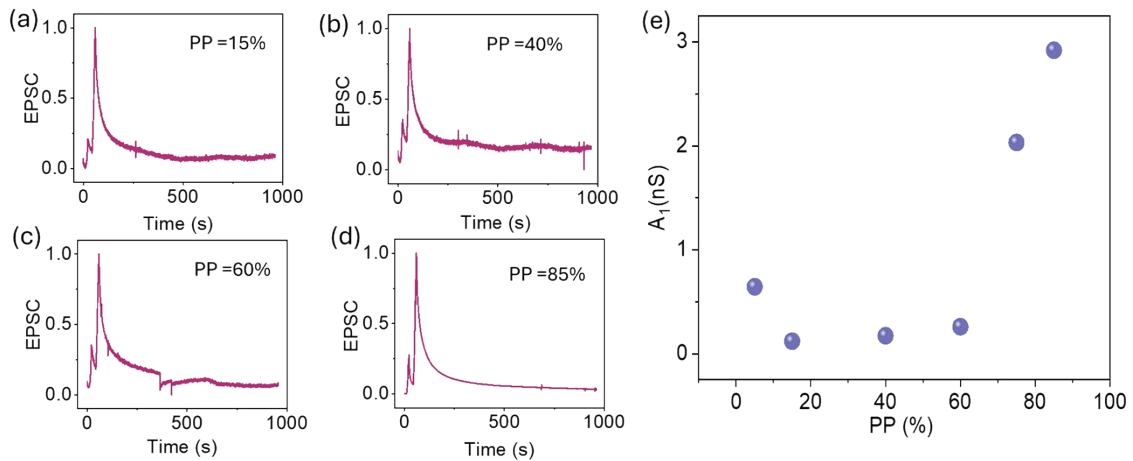
**Figure S3.** (a-b) Double exponentially fitted decay curves post first pulse and second pulse, considering percentage decay till 3% of maximum EPSC with  $\tau_1$  and  $\tau_2$  mentioned in the inset. The decay post first and second pulse has been fitted with a double exponential function. The percentage of decay during  $t_i$  (after the first pulse) is considered, and the same decay percentage has been utilized during the second pulse decay fitting. This has been done at all the  $t_i$ 's.



**Figure S4.** (a) Response curves for different  $t_i$  for showing PPD with 5% RH pulses ( $t_w = 15$  s) with other parameters as described in Figure 3a. (b-c) Response curves for different  $t_i$  for showing PPF with 75% RH pulses ( $t_w = 15$  s) as described in Figure 3c, and  $t_i$  is monitored at 40% RH.

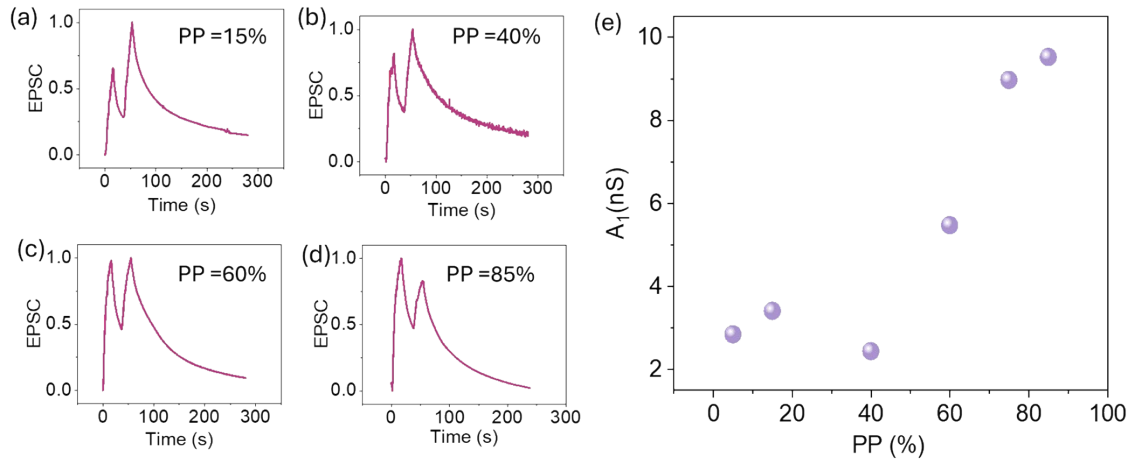


**Figure S5.** Comparison of base humidity-dependent facilitation in the device with base (a) 40% RH, (b) 5% RH,  $t_w = 15$  s,  $t_i = 15$  s (c) Variation in PPF index with two different base RH.



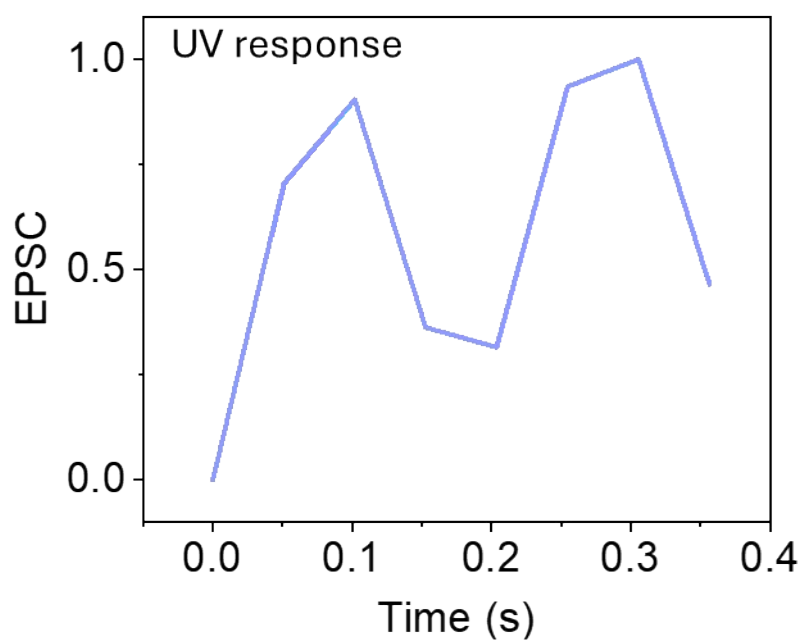
**Figure S6.** Priming protocol discussed in Figure 4d. Priming pulse RH was varied from (a) 15% to (d) 85% with PP of 5 minutes, and post-PP delay is 5 minutes, and responses were recorded with 75% RH (TP) with  $t_w=t_i= 15$  s and  $t_i$  monitored at 40% RH. (e)  $A_1$  plotted with increasing PP.

The remaining response curves (Figure S6a–d) corresponding to the PPF index in Figure 4d indicate a similar  $A_2/A_1$  ratio across all priming conditions.  $A_1$  maintains a similar value till 60% PP RH proves that 5 minutes of post-PP delay is sufficient to diminish the effect of Priming (Figure S6e).

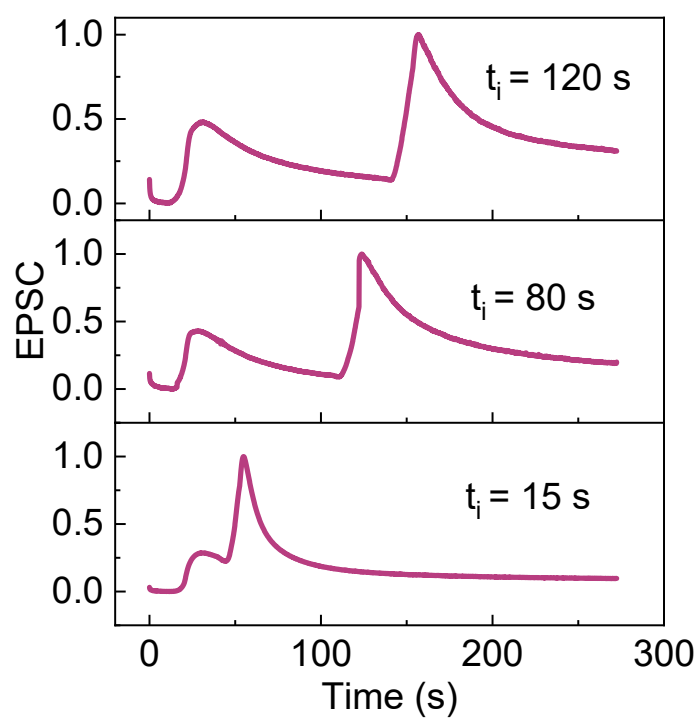


**Figure S7.** Priming protocol discussed in Figure 4g. Priming pulse RH was varied from (a) 15% to (d) 85% with  $t_w$  of 2 minutes, and post PP delay is 20 s, and responses were recorded with 75% RH (TP) with  $t_w=t_i=15$  s and  $t_i$  monitored at 40% RH. (e)  $A_1$  variation with varying PP.

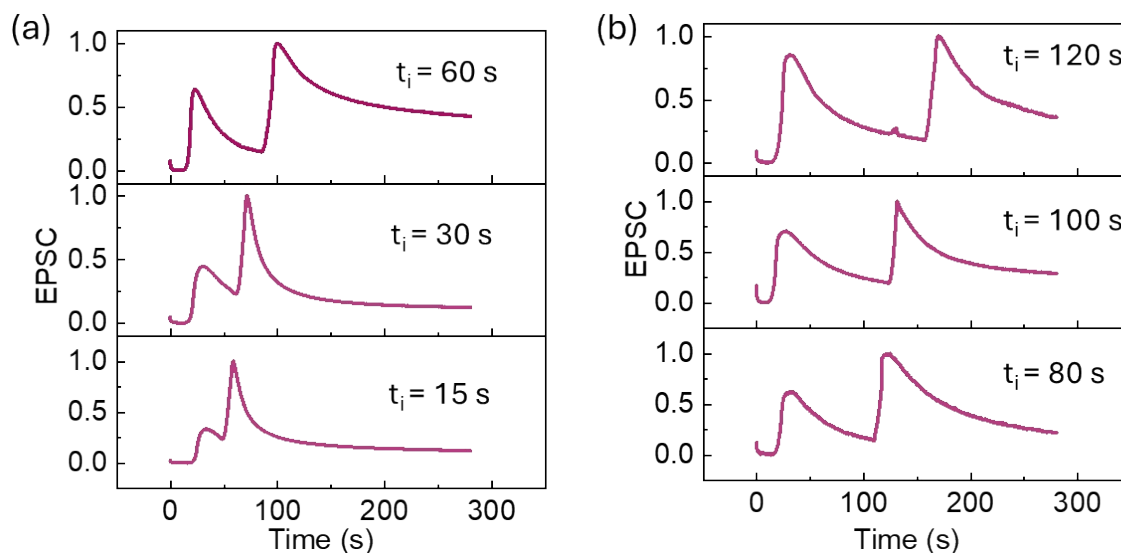
The remaining representative response curves for the index shown in Figure 4g explain that with increasing PP,  $A_2$  decreases with respect to  $A_1$  (Figure S7a-d).  $A_1$  shows an increasing trend with increasing PP (Figure S7e), proving the influence of PP, with 20 s post-PP delay on the nanofibres.



**Figure S8.** Response with UV light of wavelength 365 nm ( $t_i = t_w = 100$  ms) and power 20.5 mW.



**Figure S9.** Remaining response curves for PPF index shown in Figure 5c (with light), where 75% RH pulses with  $t_w$  of 10 s were applied with continuous light illumination.  $t_i$  was varied from 15 to 120 s, monitored at 40% RH.



**Figure S10.** Response curves for the PPF index, shown in Figure 5c, without light show a lower PPF index as compared to that with light.

## References

- 1 C. Han, X. Han, J. Han, M. He, S. Peng, C. Zhang, X. Liu, J. Gou and J. Wang, *Adv Funct Mater*, 2022, 32, 2113053.
- 2 R. Sadhukhan, S. P. Verma, S. Mondal, A. Das, R. Banerjee, A. Mandal, M. Banerjee and D. K. Goswami, *Small*, 2024, 20, 2307439.
- 3 D. Chen, X. Zhi, Y. Xia, S. Li, B. Xi, C. Zhao and X. Wang, *Small*, 2023, 19, 2301196.
- 4 A. B. Mishra and R. Thamankar, *Nanoscale*, 2024, 16, 18597–18608.
- 5 W. Xiao, L. Shan, H. Zhang, Y. Fu, Y. Zhao, D. Yang, C. Jiao, G. Sun, Q. Wang and D. He, *Nanoscale*, 2021, 13, 2502–2510.
- 6 P. Zhao, R. Ji, J. Lao, W. Xu, C. Jiang, C. Luo, H. Lin, H. Peng and C.-G. Duan, *Org Electron*, 2022, 100, 106390.
- 7 S. Dan, S. Paramanik and A. J. Pal, *ACS Nano*, 2024, 18, 14457–14468.
- 8 Z. Peng, Z. Cheng, S. Ke, Y. Xiao, Z. Ye, Z. Wang, T. Shi, C. Ye, X. Wen, P. K. Chu, X.-F. Yu and J. Wang, *Adv Funct Mater*, 2023, 33, 2211269.
- 9 Y. Ni, L. Yang, J. Feng, J. Liu, L. Sun and W. Xu, *SmartMat*, 2023, 4, e1154.

