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## **Supporting Information**

## Memristive properties and synaptic plasticity in substituted pyridinium iodobismuthates

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Name of the	Thickness	Thickness of	n	k
absorber layer	of	absorber layer		
	Roughness			
4-AmpyBiI3	9.79 nm	202.78 nm	2.13	0.0204
4-MepyBiI3	13.98 nm	150.79 nm	2.33	0.0468
4-DmapyBiI3	20.49 nm	164.06.45 nm	2.1170	0.00068
4-CNpyBiI3	-	169.16	1.5648	0.38812
			$(n_{xy} = 1.6358;$	(k <sub>xy</sub> =0.3363;
			$n_z = 1.4226$ )	$k_z = 0.4917$ )

**Table S1.** Extracted parameters from the fitted experimental data to the introduced model for the fabricated devices from pyridinium-based bismuthates samples as Tauc-Lorentz layer.

Table S2. Effect of the surface area of the electrode on LRS (ON) and HRS (OFF) states.<sup>a</sup>

Entry	Surface area/1 mm <sup>2</sup>		Surface area	$1/9 \text{ mm}^2$	Ratio	Ratio
	Current	Current	Current	Current	ON <sup>b</sup> /ON <sup>c</sup>	OFF <sup>b</sup> /OFF <sup>c</sup>
	(mA)/ON	(mA)/OFF	(mA)/ON	(mA)/OFF		
4-CNpyBiI <sub>3</sub>	2.58±0.1	$1.46\pm0.1$	3.00±0.1	2.03±0.2	1.16	1.39
4-MepyBil <sub>3</sub>	1.92±0.1	$1.42 \pm 0.07$	3.08±0.2	1.84±0.1	1.6	1.29
4-AmpyBiI <sub>3</sub>	2.44±0.1	1.94±0.3	5.53±0.03	3.24±0.4	2.27	1.64

<sup>a</sup>The average amount of 5 different measured devices.

<sup>b</sup>surface area 1mm<sup>2</sup>; <sup>c</sup>surface area 9mm<sup>2</sup>

Table S3	. Rectification	factor for devi	ces with different	t surface area	of electrode. <sup>a</sup>
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Entry	Surface area/1 mm <sup>2</sup>		Ratio	Surface area/9 mm <sup>2</sup>		Ratio
	Current (mA)	Current		Current	Current	
	at +2V	(mA) at		(mA) at	(mA) at	
		-2V		+2V	-2V	
4-CNpyBiI <sub>3</sub>	30.74±1	39.9±0.5	1.3	24.47±1	33.03±0.5	1.35
4-MepyBiI <sub>3</sub>	38.34±0.8	48.02±1	1.25	31.47±0.5	39.05±1	1.24
4-AmpyBiI <sub>3</sub>	78.65±1 <sup>b</sup>	104.24±1.5 °	1.33	75.48±1.5 <sup>b</sup>	83.92±1°	1.11

<sup>a</sup>The average amount of 5 different measured devices.

<sup>b</sup>measured at +4V; <sup>c</sup>measured at -4V



Figure S1. UPS spectra of thin films on ITO glasses a) 4-MetpyBiI<sub>3</sub> b) 4-AmpyBiI<sub>3</sub>



Figure S2. 4-CNpyBiI<sub>3</sub>/ITO glasses with different metal electrodes as top electrode.



Figure S3. Disappearing Ag electrodes after one day from the surface of thin layers of the Bicomplexes on ITO/glasses.





**Figure S4.** Scan rates a) Cu/4-CNpyBiI<sub>3</sub>/ITO b) Cu/4-MepyBiI<sub>3</sub>/ITO c) Cu/4-AmpyBiI<sub>3</sub>/ITO. Voltage ranges d) Cu/4-CNpyBiI<sub>3</sub>/ITO e) Cu/4-MepyBiI<sub>3</sub>/ITO f) Cu/4-AmpyBiI<sub>3</sub>/ITO in different voltages range. Device-to-device reproducibility for 5 different batches of samples is also shown in bottom panel.



Time (s)



**Figure S5.** The Retention measurements of on and off states in 4-CNpyBiI<sub>3</sub> and 4-MepyBiI<sub>3</sub> at  $\pm 2$  V as set and reset voltages. Retention measurements of the devices made of 4-AmpyBiI<sub>3</sub>,  $\pm 4$  V was chosen as the set and rest voltages and the read point was + 0.2 V. The width of all pulse was 0.1 s.



**Figure S6.** Representation of 0D a) 4-CNpyBiI<sub>3</sub> and b) 4-MepyBiI<sub>3</sub> c) 4-DmapyBiI<sub>3</sub> and 1D d) and 4-AmpyBiI<sub>3</sub> of ionic fragments of Bi-I and void shapes.



**Figure S7.** Double logarithmic scales of IV values of 4-CNpyBiI<sub>3</sub>, 4-MepyBiI<sub>3</sub>, 4-DmapyBiI<sub>3</sub> and 4-AmpyBiI<sub>3</sub> in SET process.



**Figure S8.** LTP and LTD for prepared devices with different pulse sequences with different width (0.001 s or 0.01 s) and amplitude 9between 0.4 V -1.6 V).



Figure S9. Illustration of final pulses shape on memristor devices, depends on time difference.



**Figure S10.** STDP of 4-CNpyBiI<sub>3</sub> with RT-pulse (#1 and #4) ; pulse amplitude to ( $\pm$ 1 V) and pulse amplitude to ( $\pm$ 2 V).



**Figure S11.** STDP of 4-MepyBiI<sub>3</sub> in different pulse polarities with  $\pm 1$  V amplitude.



**Figure S12**. STDP of 4-MepyBiI<sub>3</sub> with RT-pulse (#1 and #4); pulse amplitude to ( $\pm$ 1 V) and pulse amplitude to ( $\pm$ 2 V).