Supporting Information

Reversible Shuttle Action upon Dehydration and Rehydration Processes in Cationic Coordinatively-Bonded (4,4) Square-Grid Nets Threaded by Supramolecular Bonded Anions, $\{[Cu^{II}(4,4'\text{-bpy})_2(H_2O)][Cu^{II}(2\text{-py}SO_3)_3](NO_3)\}\cdot H_2O$

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Table S1. Hydrogen bonds for **1**, **2** and **1′**.

	D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
1	O(1*w*)-H(1*w*A)...O(2*w*)	0.84	1.86	2.678(5)	166.2
	O(1*w*)-H(1*w*B)...O(11*e*)	0.83	1.95	2.784(5)	174.2
	O(2*w*)-H(2*w*B)...O(1*a*)	0.85	2.02	2.833(5)	159.8
	O(2*w*)-H(2*w*B)...O(4*a*)	0.85	2.04	2.875(6)	167.0

	O(2*w*)-H(2*w*B)...S(2*a*)	0.85	2.96	3.666(5)	141.5
2	O(1*w*)-H(1*w*B)...O(11*e*)	0.84	1.97	2.81(3)	176.8
	O(1*w*)-H(1*w*A)...O(2*w*)	0.83	1.83	2.651(4)	167.4
	O(1*w*)-H(1*w*B)...O(11*e*)	0.83	1.93	2.761(4)	174.5
1′	O(2*w*)-H(2*w*B)...O(1*a*)	0.85	2.01	2.821(4)	161.6
	O(2*w*)-H(2*w*B)...O(4*a*)	0.85	2.00	2.835(5)	167.7
	O(2*w*)-H(2*w*B)...S(2*a*)	0.85	2.91	3.613(4)	141.3

Symmetry Codes: a) x, -y+3/2, -z +1/2; e) -x, y+1/2, -z+1/2.

Conductivity measurement

Instrument: Keithley Source metre 2400.

Use voltage: 2 V.

Method: The electrical conductivity measurements at room temperature on single crystals of **1** were performed by contacting gold lines with electrically conductive polymer gel, the surfaces of the crystal were covered with polymer gel to form electrodes.

Result: By curiosity we thought of measuring the dc-electrical conductivity and therefore prepared a single crystal of **1** with contact gold electrodes glued with electrically conductive polymer gel (Figure S1) for measurement at room temperature. To our surprise the conductivity was measureable with $\sigma_{\parallel}$ corresponding to the *c* direction at 1.32×10^{-6} S/m and $\sigma_{\perp}$ at 4.30×10^{-7} S/m. These values are not outstanding compared to those reported for organic-inorganic hybrid electrical materials,[1] but they are comparable to the first example in which I_2 molecules arranged in a chain within the cavities of a π-donor-type framework ($\sigma_{\parallel}$ values in the order of 10^{-6} - 10^{-8} S/m).[2] The measured anisotropy factor ($\sigma_{\parallel}, \sigma_{\perp}$) of 3 provides evidence for a preferred conductivity along the complex anions stacking chains.

1. (a) Bilgin, A.; Yagci, C.; Yildiz, U.; Ozkazanc, E.; Tarcan, E. *Polyhedron* **2009**, *28*, 2268. (b) Xu, Z. T. *Coord. Chem. Rev.* **2006**, *250*, 2745. (c) Cui, H.-B.; Zhou, B.; Long, L.-S.; Okano, Y.; Kobayashi, H.; Kobayashi, A. *Angew. Chem., Int. Ed.* **2008**, 47, 3376. (d) Mitsumi, M.; Ueda, H.; Furukawa, K.; Ozawa,Y.; Toriumi, K.; Kurmoo, M. *J. Am. Chem. Soc.* **2008**, *130*, 14102. (e) Zeng, M.-H.; Wang, Q.-X.; Tan, Y.-X.; Hu, S.; Zhao, H.-X.; Long, L.-S.; Kurmoo, M. *J. Am. Chem. Soc.* **2010**, *132*, 2561.
2. Hertzsch, T.; Budde, F.; Weber, E.; Hulliger, J. *Angew. Chem., Int. Ed.* **2002**, *41*, 2282.

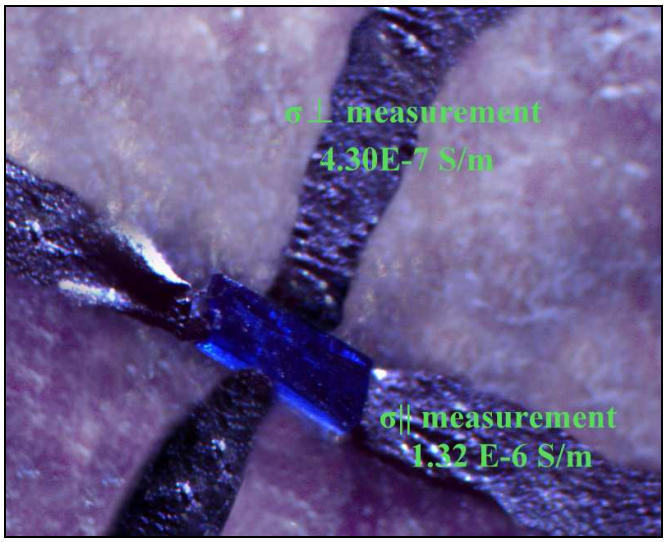

Figure S1. Tipcal image for the conductivity measurement of a sigle crystal **1**.