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Supporting Information

Crystal Structure, Magnetic and Thermal Properties of Basic Copper Formates with Two-dimensional Triangular Lattice Magnetic Networks

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1. Photograph of products



Figure S1. Photograph of the products obtained by hydrolysis of the solution at 75 $^{\circ}$ C. The greenish plates are 1. The dark red crystals are Cu₂O.

2. Infrared spectrum of 1



Figure S2. Infrared spectrum of 1.

3. Crystal Structure of 2



Figure S3. Crystal structure of **2**. Spheres correspond to Cu (blue), O (red), C (dark grey), and H (light grey). (a) Stacking of the copper hydroxide layers and the organic anion layers. (b) Projection of the atomic arrangement of the copper hydroxide layer along the c axis. (c) Schematic representation of the copper hydroxide layer of **2**. The bold lines are the axial bonds. Coloured O's correspond to the oxygen atom of formate anions. (d) Molecular alignment of formate ions in **1**. Spheres correspond to O (red, magenta), C (dark grey), and H (light grey). The light gray formate ions are coordinating to the neighboring copper hydroxide layer.

4. Crystal Structure of 3



Figure S4. Crystal structure of **3**. Spheres correspond to Cu (blue), O (red), C (dark grey), and H (light grey). (a) Stacking of the copper hydroxide layers. (b) Projection of the atomic arrangement of the copper hydroxide layer along the c axis. (c) Schematic representation of the copper hydroxide layer. The bold lines are the axial bonds. Broken lines are carboxylate groups of formate ions bridging copper ions.

5. Magnetic interaction pathways in 1



Figure S5. Five kinds of magnetic interaction pathways in the copper hydroxide layer of **1**.