

Supplementary Materials

Gas-Dependent Reversible Structural and Magnetic Transformation between Two Ladder Compounds

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1. Experimental setup and schematic diagram

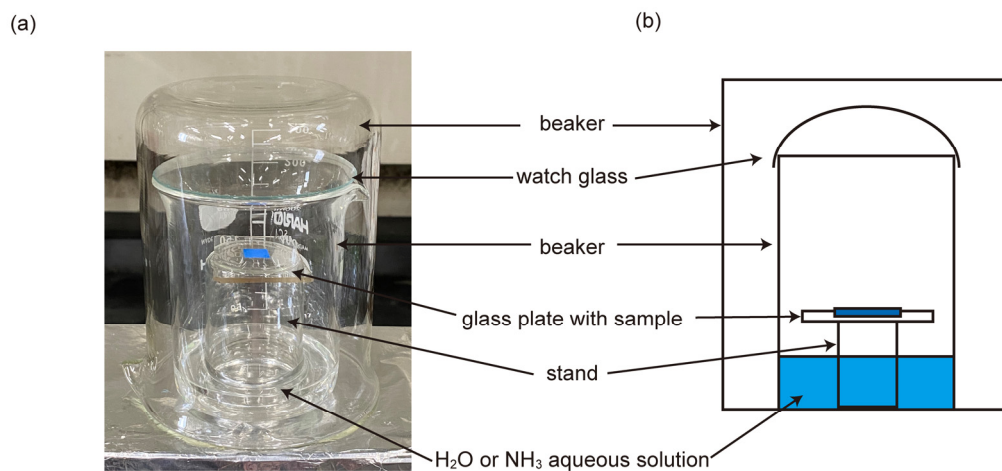


Figure S1. (a) Photo and (b) schematic diagram of the experimental setup for preparing the gaseous vapors.

2. Temperature-dependent magnetization of **1**

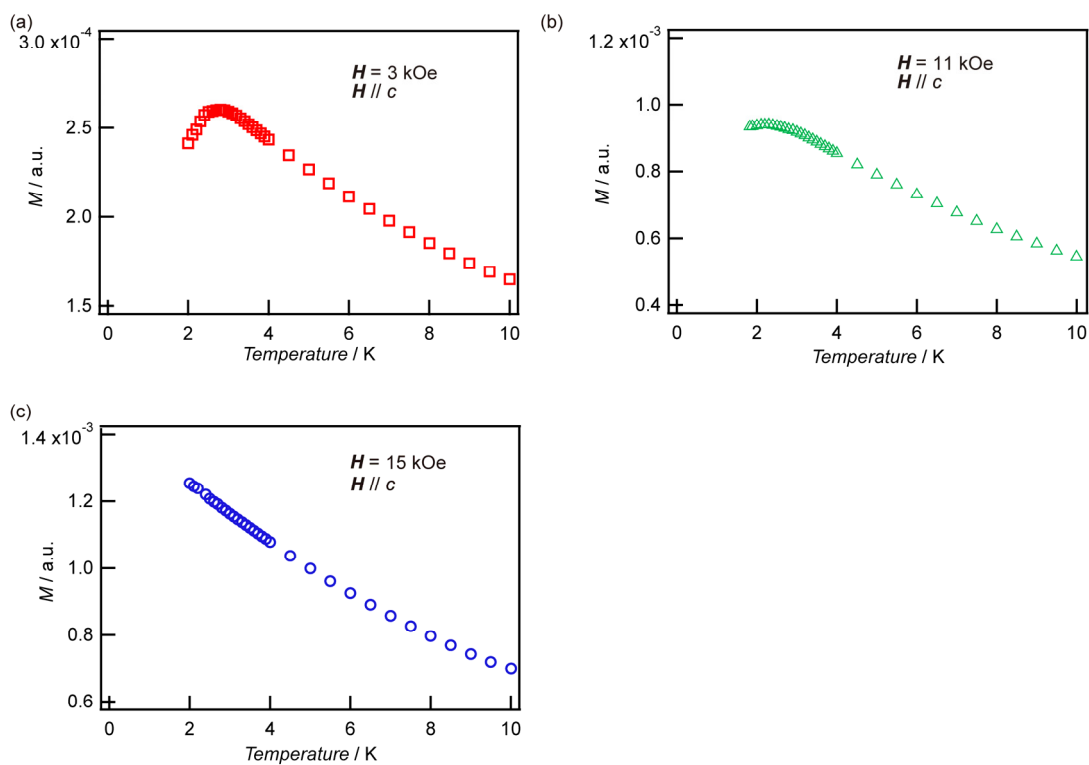


Figure S2. Temperature-dependent magnetization of **1** in fields of (a) 3 kOe, (b) 11 kOe, and (c) 15 kOe applied parallel to the c -axis.

3. Temperature-dependent heat capacity of **1** under variable DC magnetic fields

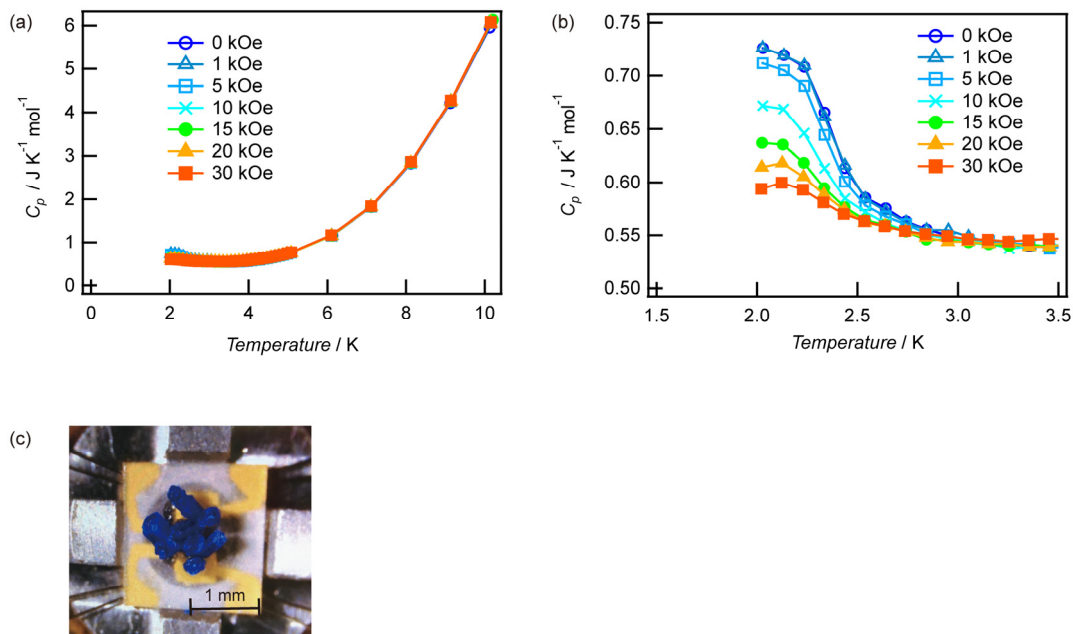


Figure S3. (a) Temperature-dependent heat capacity of **1** under magnetic fields of 0-30 kOe applied parallel to the *c*-axis. (b) An enlarged section of (a) showing data from 1.5 to 3.5 K. (c) Sample setup for the measurements. The solid lines are guides for the eye.

4. Temperature-dependent magnetic susceptibility of **1** and **1** exposed to air

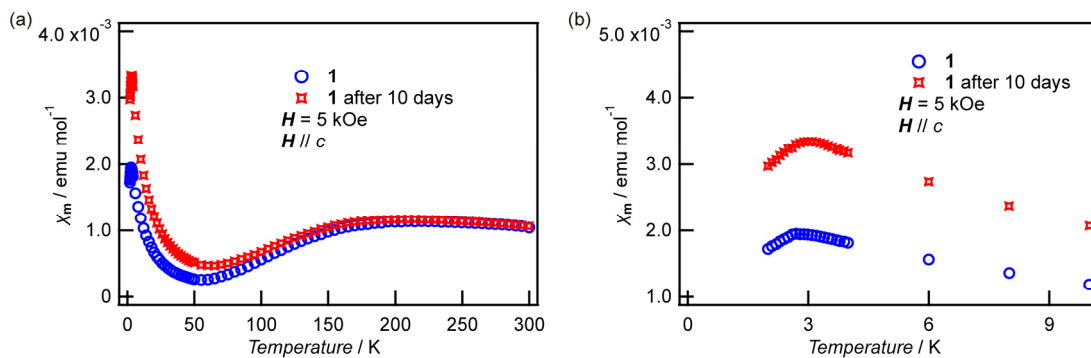


Figure S4. (a) Temperature-dependent molar magnetic susceptibility of **1** (blue) and **1** exposed to air for 10 days (red). (b) An enlarged section of the data in (a) in the low-temperature range.

5. TG and GC-MS analysis

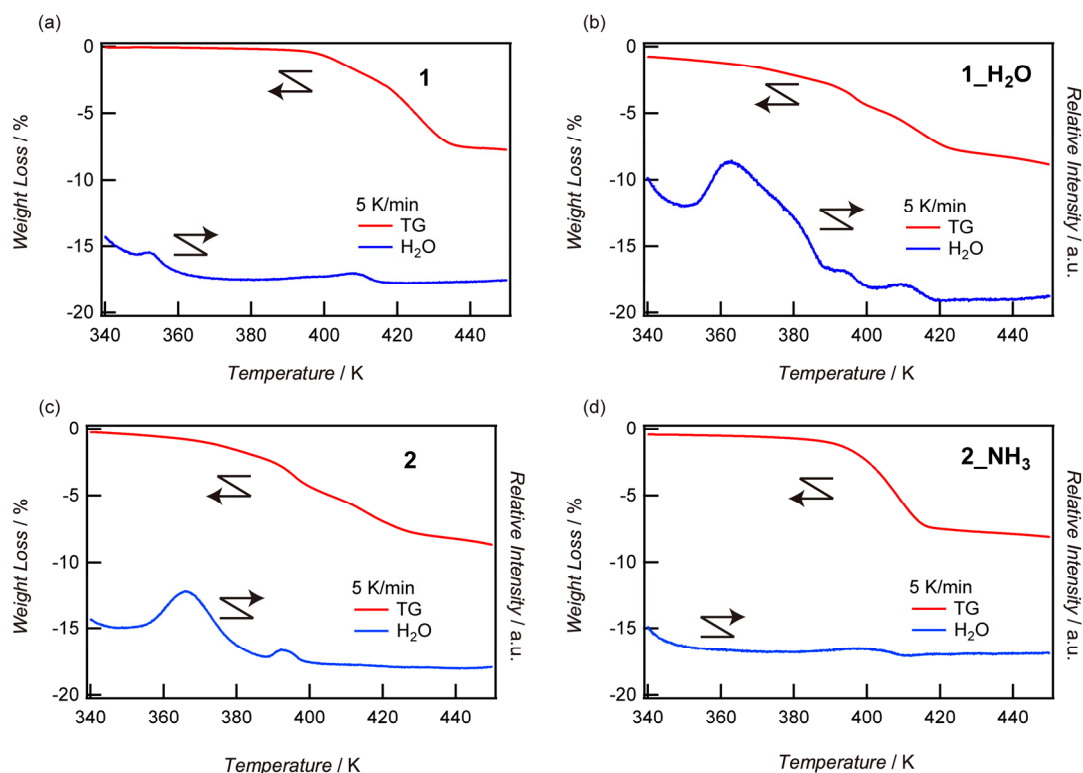


Figure S5. TG and GC-MS plots of (a) **1**, (b) **1_H₂O** (**1** exposed to H₂O vapor for 4.5 h), (c) **2**, and (d) **2_NH₃** (**2** exposed to NH₃ vapor for 2 h). Red and blue lines show weight loss and MS intensity corresponding to H₂O ($m/z = 18$), respectively.

6. IR spectra

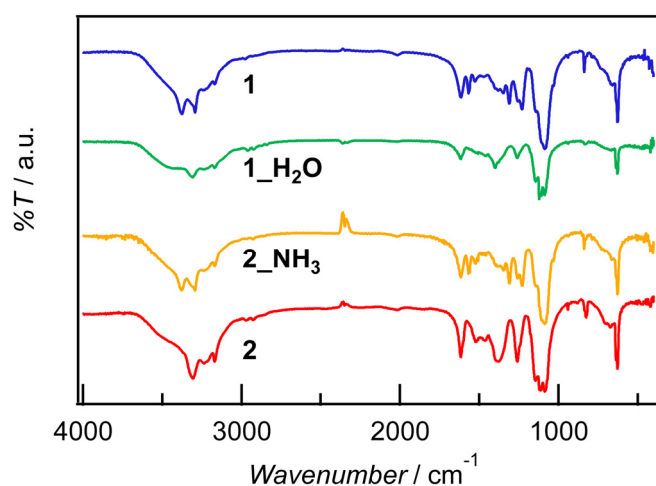


Figure S6. IR spectra of **1** (blue), **1_H₂O** (**1** exposed to H₂O vapor for 4.5 h; green), **2_NH₃** (**2** exposed to NH₃ vapor for 2 h; yellow), and **2** (red).

7. PXRD patterns of 1' and 2'

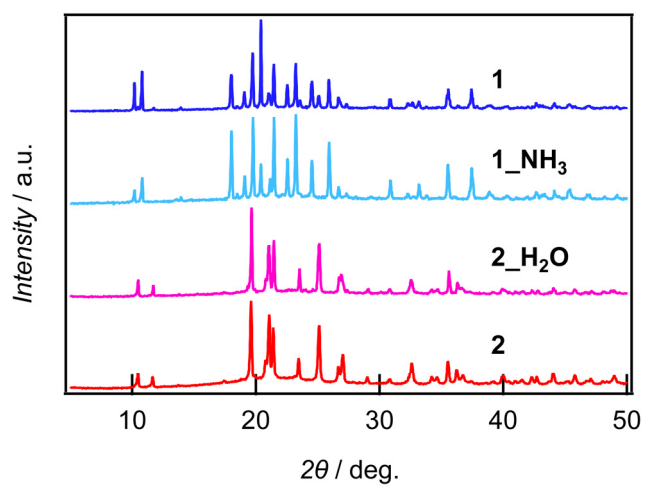


Figure S7. PXRD patterns of **1** (blue), **1'** (**1** exposed to NH₃ vapor for 2 h; light-blue), **2'** (**2** exposed to H₂O vapor for 4.5 h; pink), and **2** (red).