Supporting Information

The application of single-crystal-to-single-crystal transformation towards adjustable SMM properties

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The Synthesis of 1-2.

A water solution (10mL) of $DyCl_3$ (1mmol), phen(1mmol), HL(3mmol), and Na_2CO_3(1.5mmol) in the ratio 1:1:3:1.5 was sealed in a Teflon reactor, and heated at 210˚C for 3 day, and then cooled to room temperature at 3˚C/h. Subsequently, yellowish block crystals were obtained in 90% yield based on Dy. Element analysis (%) for **1**: calc: C 62.39, H 3.61, N 3.23; found: C 62.36, H 3.60, N 3.22. Compound **2** is obtained by calcination of **1** under vacuum for 24h. Element analysis (%) for **2**: calc: C 63.12, H 3.41, N 3.27; found: C 62.15, H 3.39, N 3.28.

General Physical Measurements. Magnetic susceptibility measurements were conducted with a Quantum Design SQUID magnetometer (MPMS XL-7) in the temperature of 2.7 to 300 K at dc=1000 Oe. AC measurements were performed at various frequencies ranging from 1 to 1500 Hz with an ac field amplitude of 3 Oe. Polycrystalline samples embedded in liquid paraffin were measured. Experimental data were corrected for the sample holder and liquid paraffin and for the diamagnetic contribution calculated from Pascal constants.

Crystallography. Single crystals with suitable dimensions were mounted on glass fibers using Nujol. All measurements were made on a Bruker SMART CCD area detector with a graphite monochromated Mo KR radiation (λ =0.71070Å). The structures were solved by direct methods *via* SHELXL-97. All non-hydrogen atoms in the structure were refined anisotropically. Hydrogen atoms were introduced as fixed contributors, except that on guest water molecule, where the hydrogen atoms is found firstly then fixed by O-H=0.85±0.01,H-H=1.25±0.01. These data have been deposited as CIFs at the Cambridge Data Centre as supplementary publication nos. CCDC: 845006/**1**, 845007/rehydrated phase, 845008/**2**, 845009/re-dehydrated phase. Copies of the data can be obtained free of charge by application to CCDC, 12 Union Road, Cambridge CB21EZ, UK (fax: (+44)1223-336033; email: deposit@ccdc.cam. ac.uk).

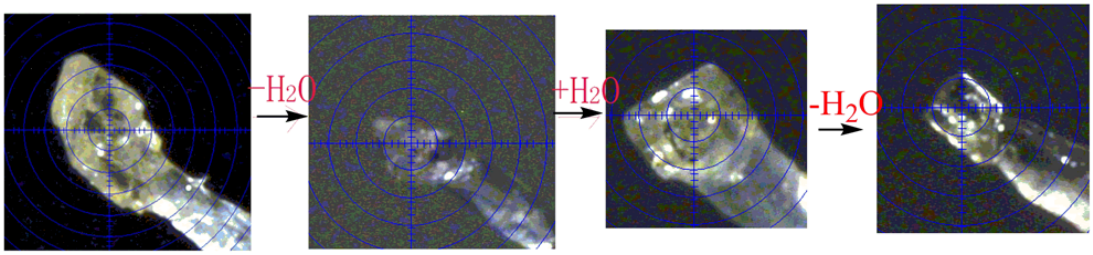

Figure S1. Photograph of SCSC between hydrated/dehydrated phases. The single crystals used here are not the same, but is randomly selected from corresponding bulk samples, respectively.

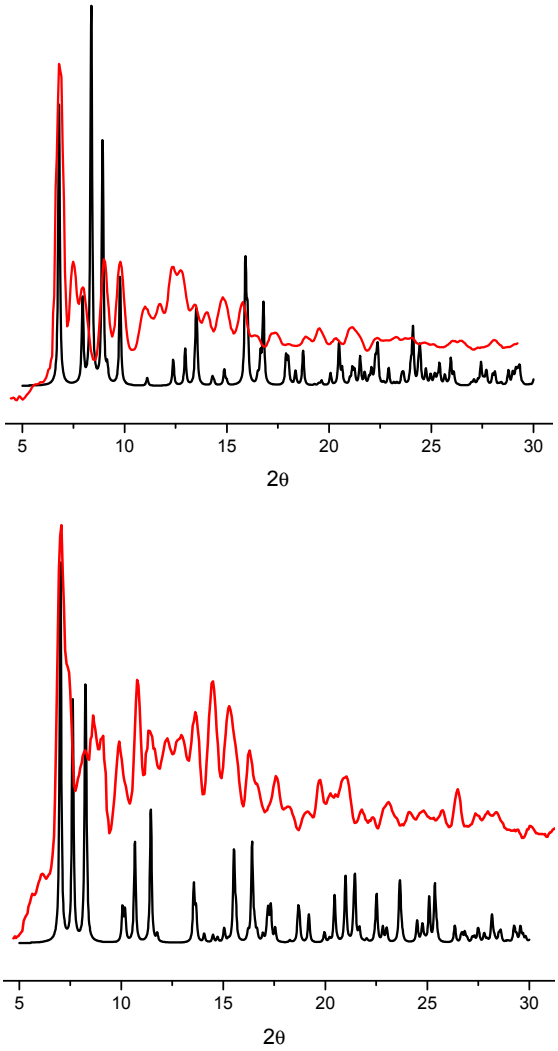

Figure S2. The experimental XRD pattern of bulk samples/red and the calculated XRD pattern of single crystal X-ray diffraction data/black for **1**-**2**, respectively.

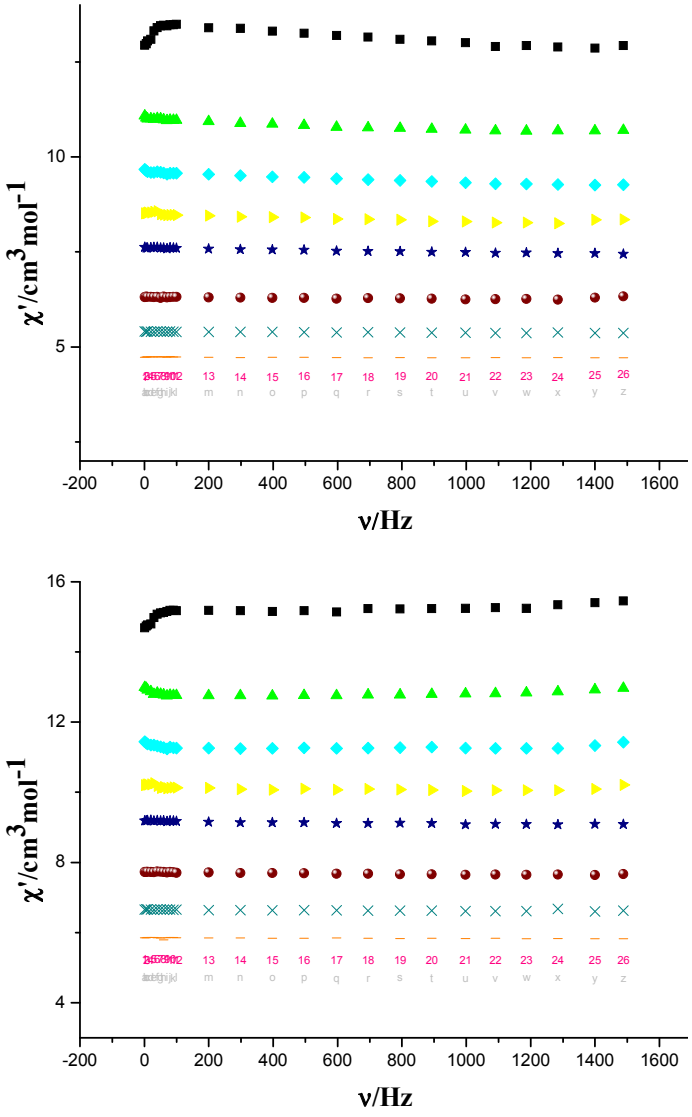

Figure S3. The χ' versus ν plots (dc/0Oe, ac/3Oe) of **1**/above, **2**/below.

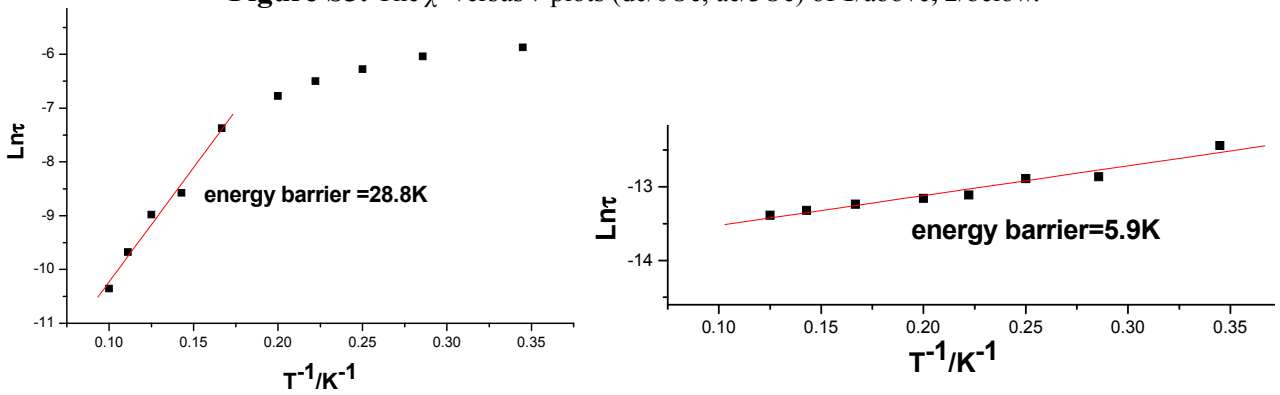

Figure S4. The lnτ vs. T^{-1} plot and the fitting by Arrhenius law marked in red: **1**/left, **2**/right.

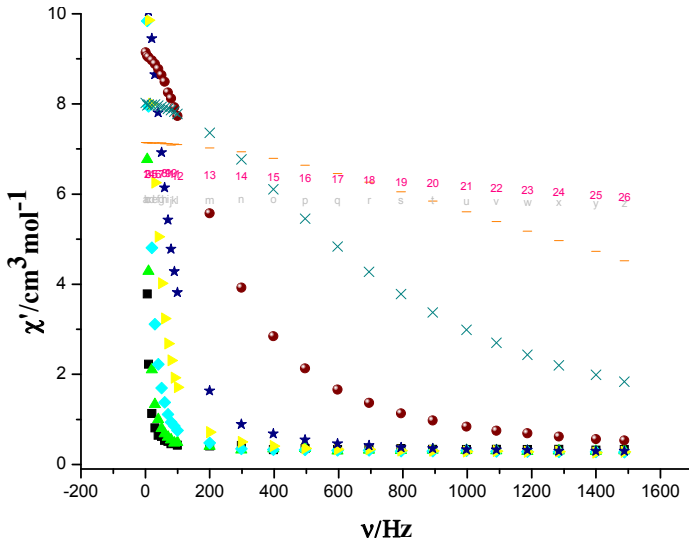

Figure S5. The χ' versus ν plots (dc/2000 Oe, ac/3Oe) of **2**.

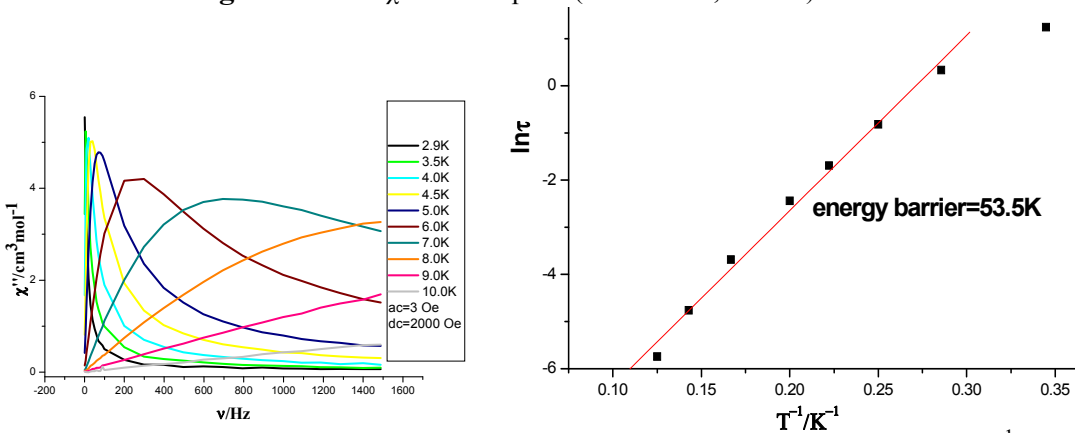

Figure S6. Left: The χ'' versus ν plots (dc/2000 Oe, ac/3Oe) of **2**; Right: The lnτ vs. T^{-1} plot and the fitting by Arrhenius law marked in red.