

## Electronic Supplementary Information For

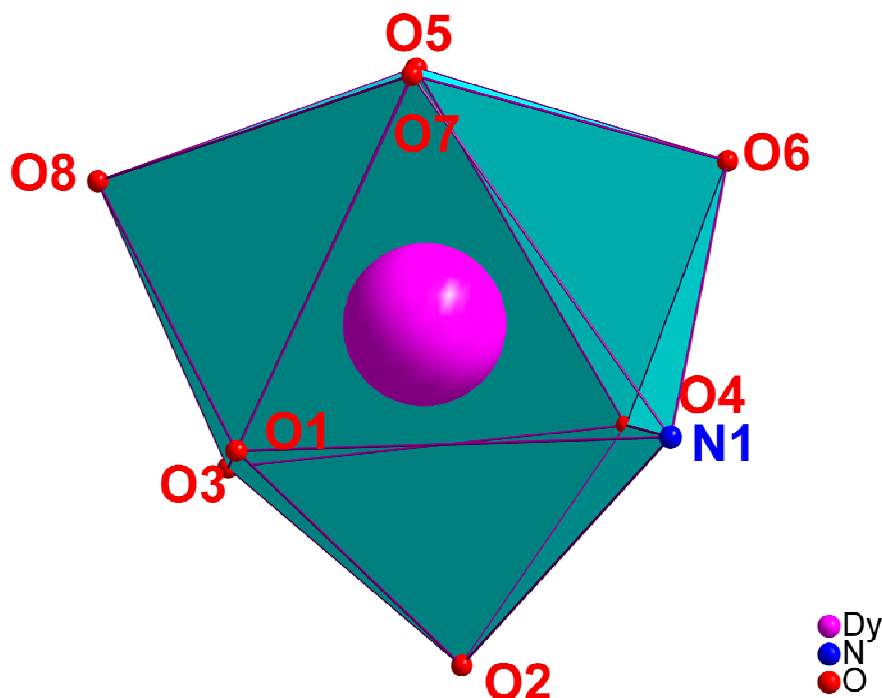
### Pyrazine-Bridged Dy<sub>2</sub> Single-Molecule Magnet with a Large Anisotropic Barrier

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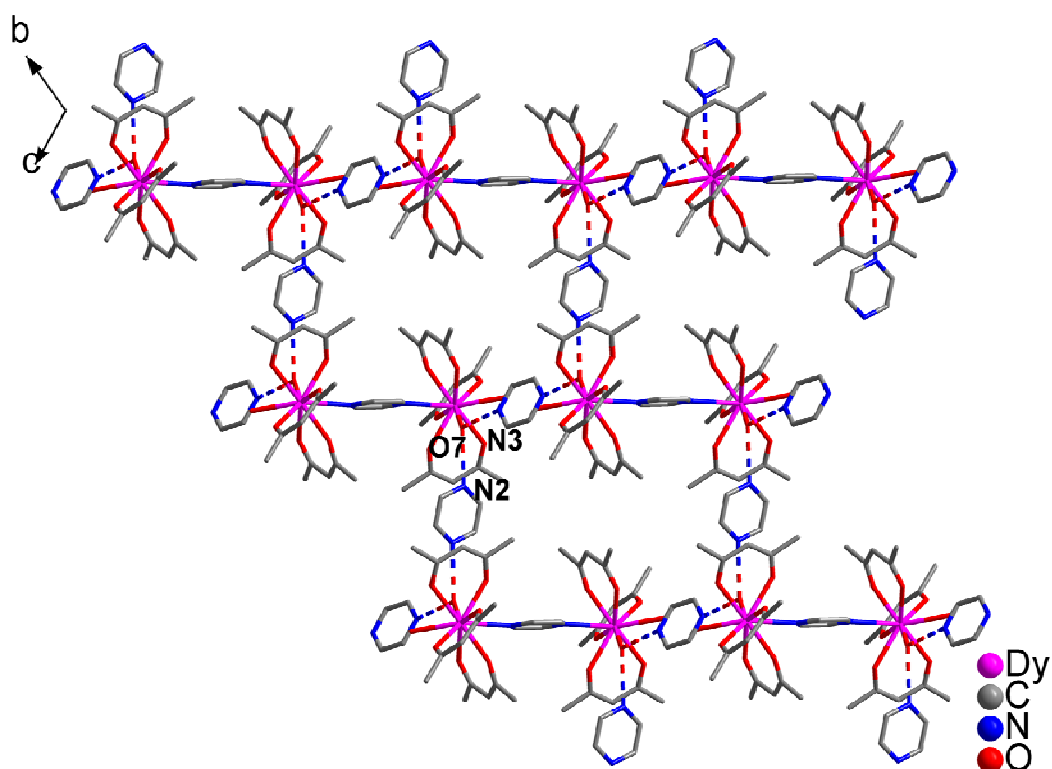
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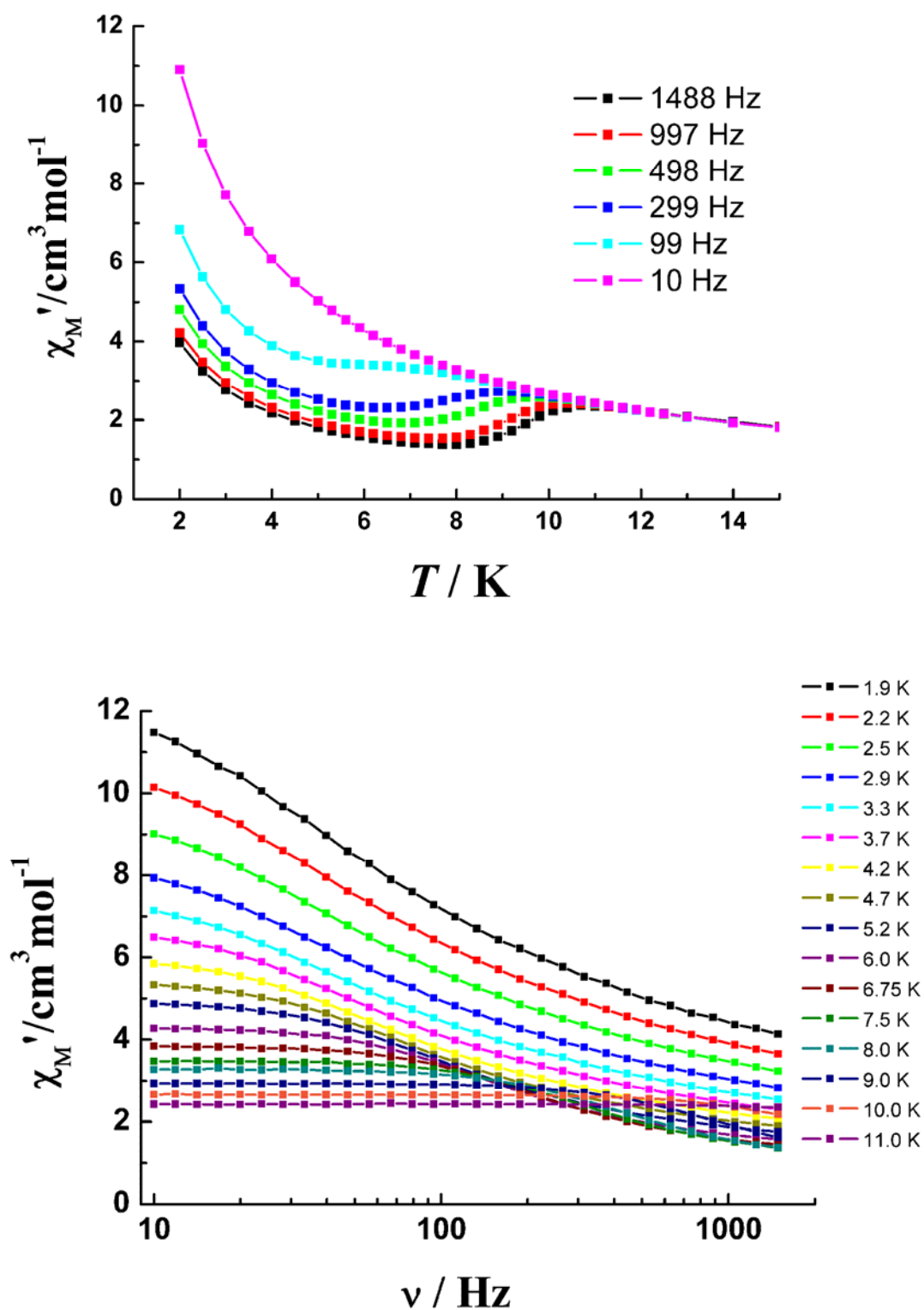
**Synthesis of Dy<sub>2</sub>(hfac)<sub>6</sub>(H<sub>2</sub>O)<sub>4</sub>pz·2pz (1):** Dy(hfac)<sub>3</sub>·2H<sub>2</sub>O (0.1 mmol) was dissolved in boiling dry *n*-heptane (20 mL). After stirring for 2 hours, pyrazine (0.1 mmol) in CH<sub>2</sub>Cl<sub>2</sub> (5 mL) was added and refluxed for 30 minutes. Then the solution was cooled to room temperature, filtrated and the filtrate was stored in a refrigerator at 4°C for a week to give pale-yellow crystals, which are suitable for X-ray analysis. Yield: 0.0095g, 5.1%. Elemental Analysis for C<sub>42</sub>H<sub>26</sub>Dy<sub>2</sub>F<sub>36</sub>N<sub>6</sub>O<sub>16</sub>: calcd: C 26.84, H 1.39, N 4.47%; found: C 26.69, H 1.28, N 4.56%. IR spectra of complex **1** (KBr cm<sup>-1</sup>): 3677.46m, 3618.61m, 3483.55m, 1652.19 vs, 1614.04w 1563.86s, 1538.16s, 1495.37vs, 1261.62s, 1225.06s, 1144.77s, 1103.79w, 1044.33w, 804.66m, 742.80w, 663.12s, 588.79.



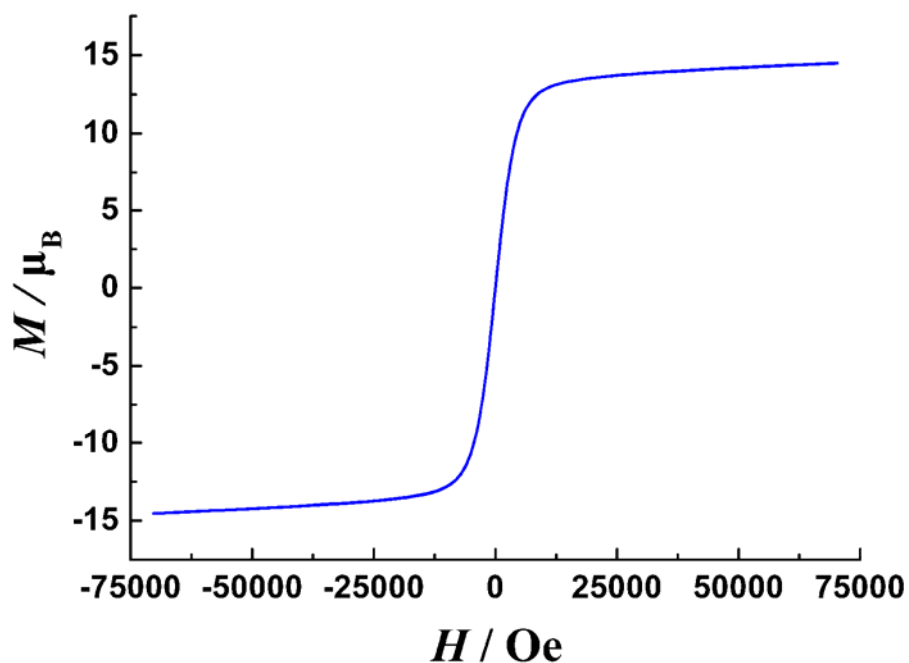
**Fig. S1.** The three capped trigonal prism environment of the nine- coordinated Dy<sup>III</sup> ions.



**Fig. S2** Packing diagram of compound **1**, showing the 2D plan linked by the hydrogen bonding and the shortest intermolecular Dy...Dy distance between dinuclear [Dy<sub>2</sub>] units.



**Fig. S3** Temperature (top) and frequency (bottom) dependence of the in-phase ac susceptibility for complex **1** measured under zero static field.



**Fig. S4**  $M$  vs.  $H$  data of **1** at 1.9 K emphasizing the absence of significant magnetic hysteresis loop at 1.9 K

Magnetic measurements were performed in the temperature range 1.9–300 K, using a Quantum Design MPMS-XL SQUID magnetometer equipped with a 7 T magnet. The diamagnetic corrections for the compounds were estimated using Pascal's constants, and magnetic data were corrected for diamagnetic contributions of the sample holder.