

ESI data file

**Table S1.** Coefficients in the empirical law used to evaluate the  $\chi_M T$  product of a magnetic topology shown in Chart 1. This law is expressed as in Eq. (2).

| i | j | $c_{i,j}^{(2)}$ | i | j | $c_{i,j}^{(2)}$ |
|---|---|-----------------|---|---|-----------------|
| 0 | 0 | 0.0             | 0 | 0 | -0.006959       |
| 0 | 1 | 0.0             | 0 | 1 | 0.0             |
| 0 | 2 | 0.0             | 0 | 2 | 0.0             |
| 0 | 3 | 0.0             | 0 | 3 | 0.0             |
| 0 | 4 | 0.0             | 0 | 4 | 0.0             |
| 1 | 0 | 0.009276        | 1 | 0 | 0.1836          |
| 1 | 1 | -0.4629         | 1 | 1 | -0.02137        |
| 1 | 2 | -0.08811        | 1 | 2 | 0.1052          |
| 1 | 3 | 0.0             | 1 | 3 | 0.0             |
| 1 | 4 | 0.0             | 1 | 4 | 0.0             |
| 2 | 0 | 0.1617          | 2 | 0 | -2.1023         |
| 2 | 1 | 1.0244          | 2 | 1 | 0.3751          |
| 2 | 2 | 1.1357          | 2 | 2 | -1.4950         |
| 2 | 3 | 0.0             | 2 | 3 | 0.0             |
| 2 | 4 | 0.0             | 2 | 4 | 0.0             |
| 3 | 0 | -0.8367         | 3 | 0 | 13.6619         |
| 3 | 1 | -7.8174         | 3 | 1 | -2.6369         |
| 3 | 2 | -4.8263         | 3 | 2 | 7.4706          |
| 3 | 3 | 0.0             | 3 | 3 | 0.0             |
| 3 | 4 | 0.2616          | 3 | 4 | 1.1647          |
| 4 | 0 | -1.4800         | 4 | 0 | -55.0615        |
| 4 | 1 | 23.7819         | 4 | 1 | 9.3912          |
| 4 | 2 | 6.0386          | 4 | 2 | -14.7523        |
| 4 | 3 | 0.0             | 4 | 3 | 0.0             |
| 4 | 4 | -0.7027         | 4 | 4 | -1.5596         |
| 5 | 0 | 30.9592         | 5 | 0 | 139.3157        |
| 5 | 1 | -16.5821        | 5 | 1 | -17.2176        |
| 5 | 2 | 4.5398          | 5 | 2 | 8.0546          |
| 5 | 3 | 0.0             | 5 | 3 | 0.0             |
| 5 | 4 | 0.0             | 5 | 4 | 0.0             |
| 6 | 0 | -113.2882       | 6 | 0 | -206.8837       |
| 6 | 1 | -30.2984        | 6 | 1 | 13.2222         |
| 6 | 2 | 0.0             | 6 | 2 | 0.0             |
| 6 | 3 | 0.0             | 6 | 3 | 0.0             |
| 6 | 4 | 0.0             | 6 | 4 | 0.0             |
| 7 | 0 | -139.7014       | 7 | 0 | 0.0             |
| 7 | 1 | 0.0             | 7 | 1 | 0.0             |
| 7 | 2 | 0.0             | 7 | 2 | 0.0             |
| 7 | 3 | 0.0             | 7 | 3 | 0.0             |
| 7 | 4 | 0.0             | 7 | 4 | 0.0             |

**Table S2.** Crystal data and details of the structure determination for compounds **1-3**.

| compound                                | <b>1</b>                                        | <b>2</b>                                       | <b>3</b>                                         |
|-----------------------------------------|-------------------------------------------------|------------------------------------------------|--------------------------------------------------|
| T(K)                                    | 110(2)                                          | 293(2)                                         | 293(2)                                           |
| Empirical formula                       | C <sub>6</sub> H <sub>15</sub> GdO <sub>9</sub> | C <sub>6</sub> H <sub>9</sub> GdO <sub>8</sub> | C <sub>5</sub> H <sub>10</sub> GdO <sub>10</sub> |
| M                                       | 388.43                                          | 366.38                                         | 387.38                                           |
| Crystal System                          | Monoclinic                                      | Triclinic                                      | Orthorhombic                                     |
| Space Group                             | <i>P</i> 2 <sub>1</sub> / <i>c</i>              | <i>P</i> -1                                    | <i>F</i> <i>d d d</i>                            |
| a, Å                                    | 9.0103(6)                                       | 8.4598(3)                                      | 9.6673(3)                                        |
| b, Å                                    | 14.9957(11)                                     | 8.5490(6)                                      | 15.4344(3)                                       |
| c, Å                                    | 9.0466(8)                                       | 8.8472(6)                                      | 27.2399(3)                                       |
| α, (°)                                  | -                                               | 118.662(5)                                     | -                                                |
| β, (°)                                  | 96.139(7)                                       | 113.552(5)                                     | -                                                |
| γ, (°)                                  | -                                               | 90.379(4)                                      | -                                                |
| V, Å <sup>3</sup>                       | 1215.33(16)                                     | 498.80(5)                                      | 4064.44(16)                                      |
| Z                                       | 4                                               | 2                                              | 16                                               |
| Index ranges                            | -11 ≤ h ≤ 11<br>-17 ≤ k ≤ 19<br>-11 ≤ l ≤ 11    | -10 ≤ h ≤ 10<br>-11 ≤ k ≤ 9<br>-11 ≤ l ≤ 9     | -12 ≤ h ≤ 12<br>-18 ≤ k ≤ 20<br>-35 ≤ l ≤ 32     |
| ρ <sub>calc</sub> (Mg m <sup>-3</sup> ) | 2.123                                           | 2.439                                          | 2.532                                            |
| λ (Mo-Kα Å)                             | 0.71073                                         | 0.71073                                        | 0.71073                                          |
| μ (Mo-Kα mm <sup>-1</sup> )             | 5.487                                           | 6.670                                          | 6.570                                            |
| R1, I > 2σ(I) (all)                     | 0.0193 (0.0289)                                 | 0.0476 (0.0526)                                | 0.0238 (0.0363)                                  |
| wR2, I > 2σ(I) (all)                    | 0.0358 (0.0377)                                 | 0.1450 (0.1484)                                | 0.0354 (0.0375)                                  |
| Measured reflections (Rint)             | 8925 (0.0279)                                   | 2476 (0.0150)                                  | 6738 (0.0328)                                    |
| Independent reflections (I > 2σ(I))     | 2770 (2362)                                     | 1907 (1751)                                    | 1154 (944)                                       |
| Crystal size (mm)                       | 0.20 x 0.16 x 0.08                              | 0.16 x 0.16 x 0.06                             | 0.18 x 0.12 x 0.1                                |