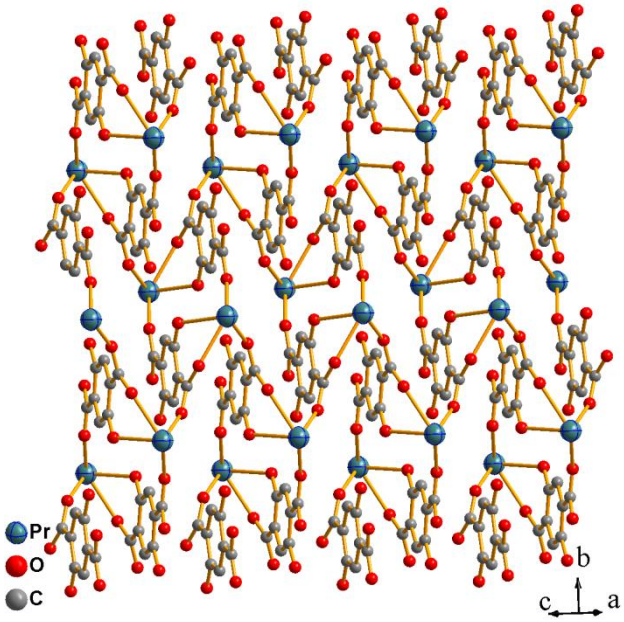



**Fig. S1** 2D layered structure in Pr(**2**).

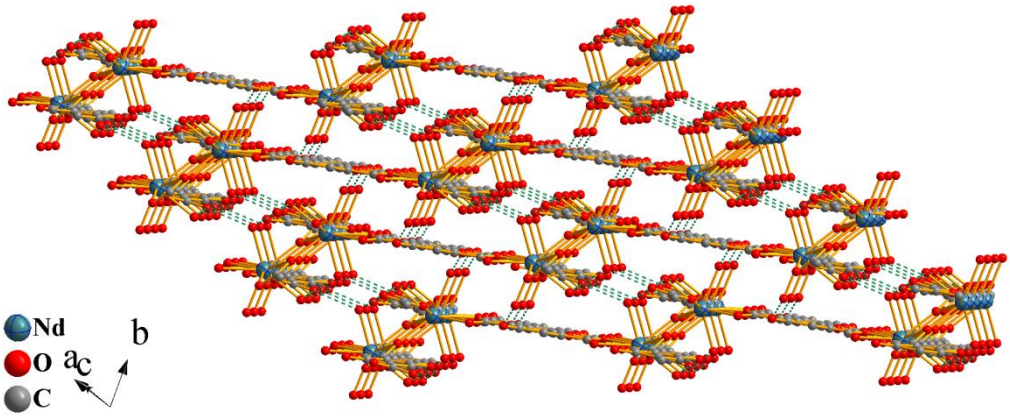



**Fig. S2** The 3D hydrogen bonding network of Nd(**3**). Dotted lines indicate the hydrogen bonds between layers.

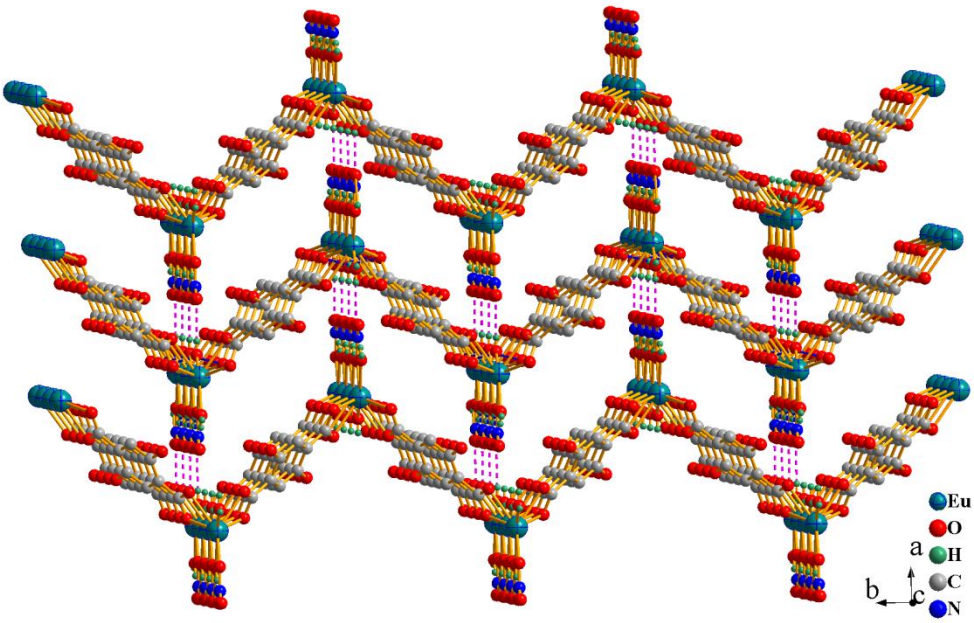



**Fig. S3** The 3D hydrogen bonding framework of Eu(**4**). Dotted lines indicate the hydrogen bonds between layers.

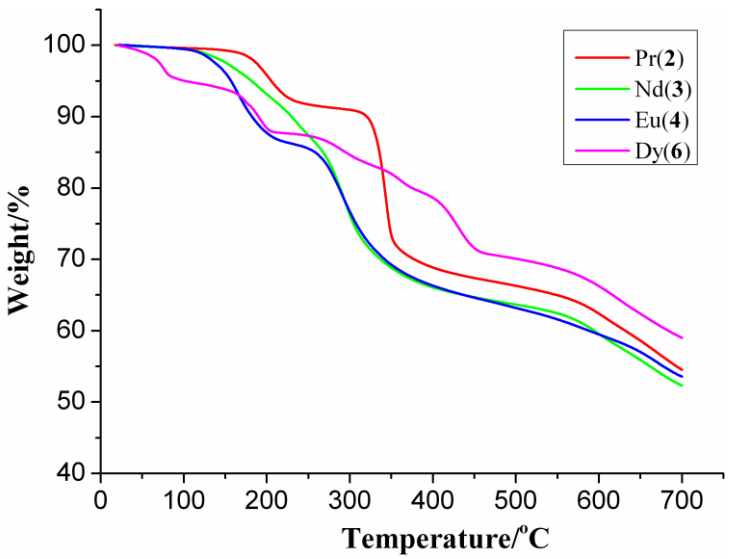
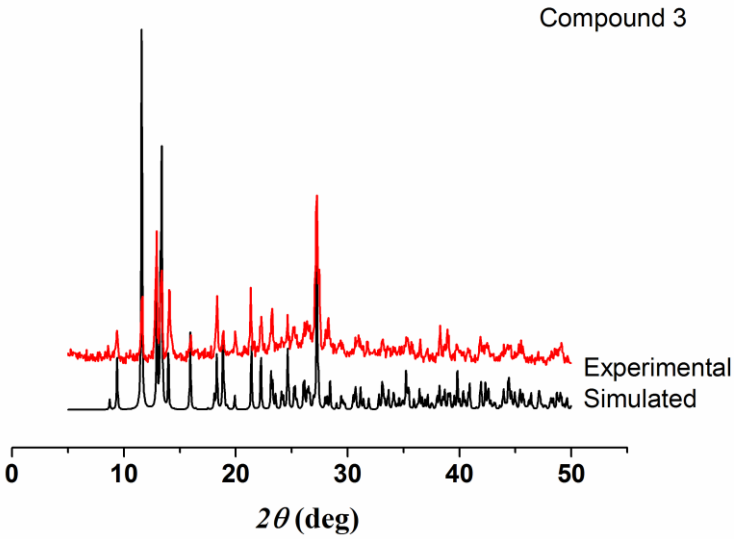
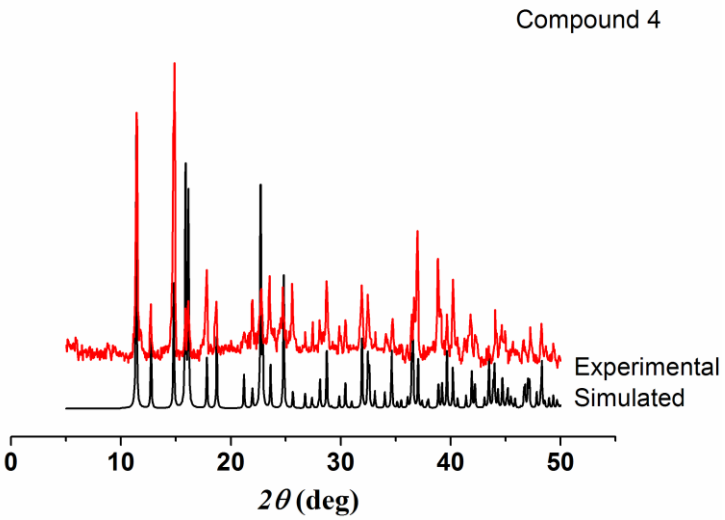
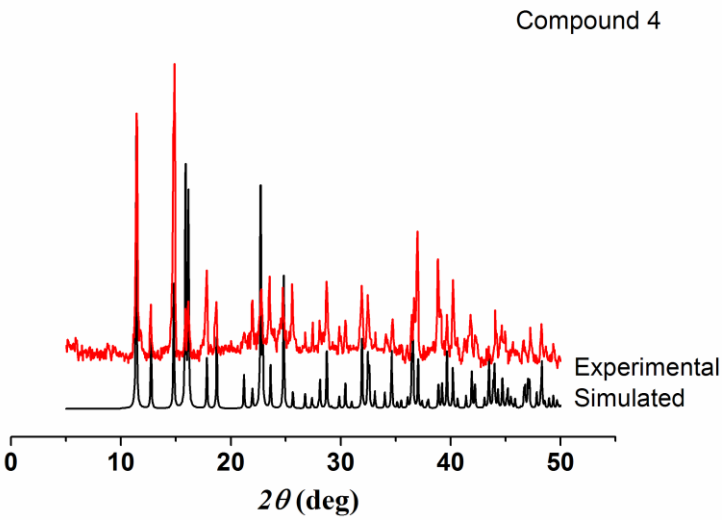



**Fig. S4** TGA curves of compounds **2**–**4** and **6**.



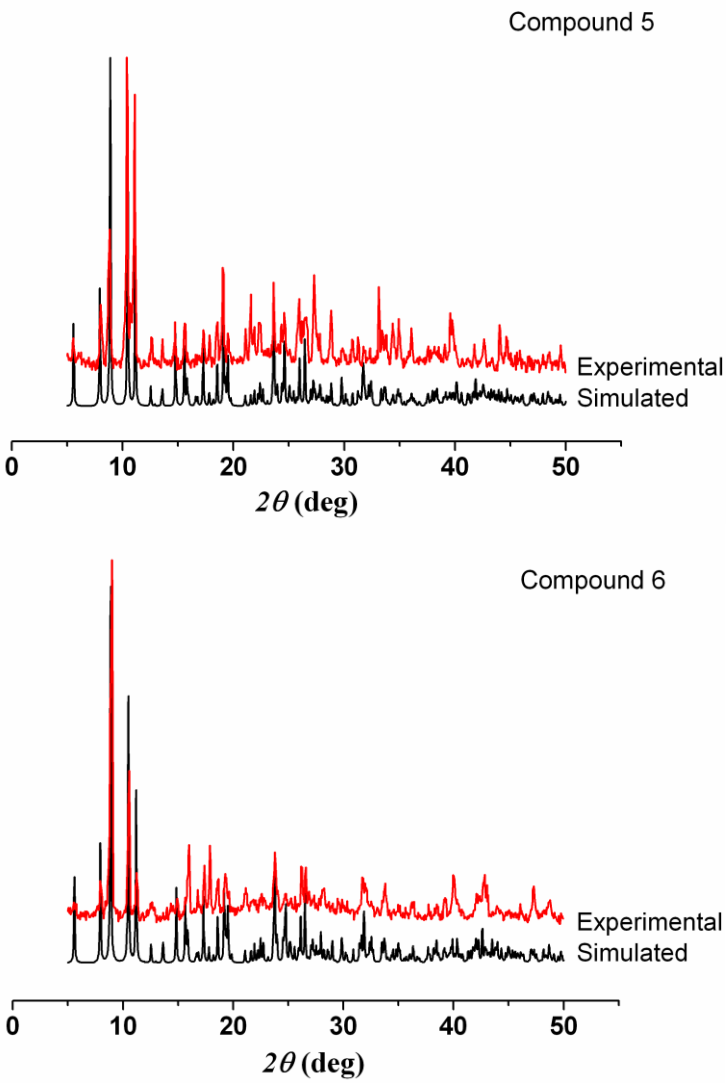



**Fig. S5** Powder X-ray diffraction patterns of compounds **3**–**6**.

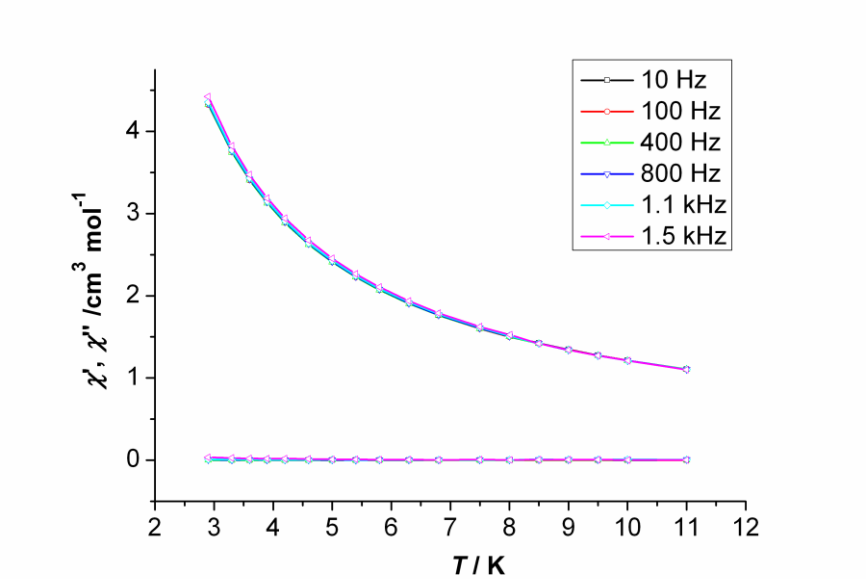



**Fig. S6** Variable-temperature AC magnetic susceptibilities of compound Dy(**6**) in an ac field of 3 Oe with oscillating frequencies.

Table **S1**. Geometrical parameters of hydrogen bonds for **1** − **6**.[a]

| D−H···A | D−H (Å) | H···A (Å) | D···A (Å) | <DHA (º) |
|---|---|---|---|---|
| Compound La(**1**) | | | | |
| O5−H5···O6$^{\#1}$ | 0.82 | 2.00 | 2.714(3) | 145.1 |
| O6−H6···O3 | 0.82 | 1.87 | 2.584(3) | 145.1 |
| O9−H9···O3$^{\#2}$ | 0.82 | 1.78 | 2.588(3) | 170.9 |
| O10−H10A···O1$^{\#3}$ | 0.82 | 2.03 | 2.805(4) | 157.4 |
| O10−H10B···O8$^{\#4}$ | 0.82 | 2.48 | 2.898(4) | 112.7 |
| O11−H11A···O8$^{\#5}$ | 0.82 | 2.00 | 2.795(4) | 163.8 |
| O11−H11B···O2$^{\#6}$ | 0.82 | 2.47 | 3.167(4) | 143.7 |
| Compound Pr(**2**) | | | | |
| O5−H5···O6$^{\#1}$ | 0.82 | 1.98 | 2.702(7) | 146.6 |
| O6−H6···O3 | 0.82 | 1.86 | 2.577(7) | 145.8 |
| O9−H9···O3$^{\#2}$ | 0.82 | 1.75 | 2.566(7) | 171.2 |
| O10−H10A···O1$^{\#3}$ | 0.82 | 1.99 | 2.777(8) | 161.6 |
| O10−H10B···O8$^{\#4}$ | 0.82 | 2.44 | 2.866(8) | 113.5 |
| O11−H11A···O8$^{\#5}$ | 0.82 | 2.01 | 2.804(7) | 162.4 |
| O11−H11B···O2$^{\#6}$ | 0.82 | 2.52 | 3.212(8) | 143.0 |
| Compound Nd(**3**) | | | | |
| O1W−H1WA···O8$^{\#1}$ | 0.82 | 2.11 | 2.908(4) | 162.9 |
| O1W−H1WB···O1$^{\#2}$ | 0.82 | 2.01 | 2.824(4) | 175.6 |
| O5−H5···O2 | 0.82 | 2.27 | 2.948(5) | 139.7 |
| O6−H6···O3 | 0.82 | 1.87 | 2.580(4) | 144.3 |
| O9−H9···O7 | 0.82 | 1.86 | 2.570(4) | 144.0 |
| O10−H10A···O1W$^{\#2}$ | 0.82 | 2.29 | 2.934(4) | 135.5 |
| O10−H10B···O9$^{\#3}$ | 0.82 | 1.92 | 2.738(4) | 172.6 |
| O11−H11A···O4$^{\#4}$ | 0.82 | 1.85 | 2.652(4) | 165.2 |
| O11−H11B···O1W | 0.82 | 2.04 | 2.805(4) | 156.3 |
| O12−H12A···O11$^{\#5}$ | 0.82 | 1.93 | 2.725(4) | 162.7 |
| O12−H12B···O4$^{\#6}$ | 0.82 | 1.90 | 2.712(4) | 171.9 |
| Compound Eu(**4**) | | | | |
| O3−H3···O2 | 0.82 | 1.81 | 2.540(2) | 146.7 |
| O4−H4A···O1W$^{\#1}$ | 0.82 | 2.03 | 2.766(2) | 148.7 |
| O4−H4B···O1W | 0.82 | 2.04 | 2.795(2) | 152.5 |
| O5−H5A···O9$^{\#2}$ | 0.82 | 2.27 | 2.897(4) | 133.7 |
| O5−H5B···O8 | 0.82 | 2.19 | 2.732(4) | 123.8 |
| O6−H6A···O9$^{\#3}$ | 0.82 | 2.21 | 2.940(2) | 148.1 |
| O6−H6B···O1$^{\#4}$ | 0.82 | 2.65 | 2.940(2) | 103.0 |
| O1W−H1WA···O3$^{\#5}$ | 0.82 | 1.94 | 2.758(2) | 171.8 |
| O1W−H1WB···O1$^{\#1}$ | 0.82 | 1.97 | 2.794(2) | 178.9 |
| Compound Gd(**5**) | | | | |
| O5−H5···O1 | 0.82 | 1.92 | 2.631(7) | 144.0 |
| O6−H6···O4 | 0.82 | 1.86 | 2.578(7) | 145.2 |
| O9−H9···O7 | 0.82 | 1.90 | 2.590(7) | 140.9 |
| O12−H12···O11$^{\#1}$ | 0.82 | 1.94 | 2.634(7) | 142.3 |
| O16−H16A···O1W$^{\#2}$ | 0.82 | 2.09 | 2.879(8) | 161.7 |
| O16−H16B···O3W | 0.82 | 1.84 | 2.654(8) | 175.8 |
| O17−H17A···O1W$^{\#3}$ | 0.82 | 2.32 | 3.072(9) | 152.9 |
| O17−H17B···O5$^{\#4}$ | 0.82 | 2.18 | 2.930(7) | 151.9 |
| O18−H18A···O4W | 0.82 | 1.99 | 2.801(10) | 172.7 |
| O18−H18B···O9 | 0.82 | 2.15 | 2.906(7) | 153.7 |
| O2W−H2WA···O4W$^{\#5}$ | 0.82 | 2.47 | 3.065(13) | 130.1 |
| O2W−H2WB···O3 | 0.82 | 2.12 | 2.845(9) | 147.2 |
| O3W−H3WA···O4W$^{\#6}$ | 0.82 | 2.08 | 2.822(9) | 150.1 |
| O3W−H3WB···O9$^{\#4}$ | 0.82 | 2.02 | 2.672(11) | 136.1 |
| O4W−H4WA···O12W$^{\#7}$ | 0.82 | 2.27 | 2.898(10) | 133.6 |
| O4W−H4WB···O1W | 0.82 | 2.50 | 3.090(11) | 130.1 |
| Compound Dy(**6**) | | | | |
| O5−H5···O1 | 0.82 | 1.93 | 2.632(5) | 143.7 |
| O6−H6···O4 | 0.82 | 1.87 | 2.582(5) | 144.3 |
| O9−H9···O7 | 0.82 | 1.90 | 2.582(4) | 140.2 |
| O12−H12···O11$^{\#1}$ | 0.82 | 1.95 | 2.638(5) | 141.5 |
| O16−H16A···O1W$^{\#2}$ | 0.82 | 2.09 | 2.893(6) | 164.7 |
| O16−H16B···O3W | 0.82 | 1.86 | 2.670(6) | 172.3 |
| O17−H17A···O1W$^{\#3}$ | 0.82 | 2.29 | 3.045(6) | 152.3 |
| O17−H17B···O5$^{\#4}$ | 0.82 | 2.20 | 2.933(5) | 149.3 |
| O18−H18A···O4W | 0.82 | 1.97 | 2.783(6) | 171.6 |
| O18−H18B···O9 | 0.82 | 2.13 | 2.911(5) | 160.5 |
| O2W−H2WA···O4W$^{\#5}$ | 0.82 | 2.56 | 3.144(10) | 129.5 |
| O2W−H2WB···O3 | 0.82 | 2.12 | 2.835(6) | 146.1 |

| | | | | |
|---|---|---|---|---|
| O3W−H3WA···O4W[#6] | 0.82 | 2.09 | 2.827(6) | 150.3 |
| O3W−H3WB···O9[#4] | 0.82 | 2.00 | 2.654(8) | 136.5 |
| O4W−H4WA···O12W[#7] | 0.82 | 2.31 | 2.924(7) | 131.9 |
| O4W−H4WB···O1W | 0.82 | 2.53 | 3.104(8) | 127.9 |

[a] Compounds **1** and **2**: #1 − *x* + 1/2, *y* + 1/2, − *z* + 3/2; #2 − *x* + 1/2, − *y* + 3/2, − *z* + 1; #3 *x*, − *y* + 1, *z* −1/2; #4 − *x* + 1, *y*, − *z* + 3/2; #5 − *x* + 1, *y*, − *z* + 5/2; #6 *x*, − *y* + 1, *z* + 1/2. Compound 3: #1 *x* − 1, *y*, *z*; #2 − *x* + 1, − *y*, − *z*; #3 − *x* + 1, − *y*, − *z* + 1; #4 *x* − 1, *y*, *z* + 1; #5 − *x* + 1, − *y* + 1, − *z*; 6# *x*, *y*, *z* + 1. Compound 4: #1 − *x* + 1/2, − *y* + 1, *z* − 1/2; #2 *x*, *y*, *z* + 1; #3 *x* + 1/2, *y*, − *z* − 1/2; #4 *x*, *y*, *z* − 1; #5 − *x* + 1, − *y* + 1, − *z*; 6# − *x* + 1/2, − *y* + 1, *z* − 1/2. Compounds 5 and 6: #1 − *x* + 1, − *y*, − *z* − 1; #2 *x* + 1, *y* − 1, *z*; #3 *x*, *y* − 1, *z*; #4 − *x* + 2, − *y*, − *z*; #5 − *x* + 2, − *y* + 1, − *z*; #6 − *x* + 2, − *y*, − *z* − 1; #7 − *x* + 1, − *y* + 1, − *z* − 1.