

## Electronic Supplementary Information

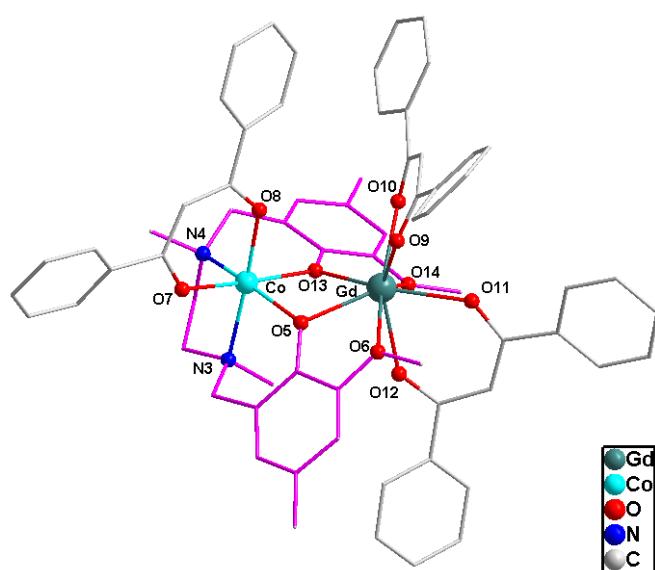
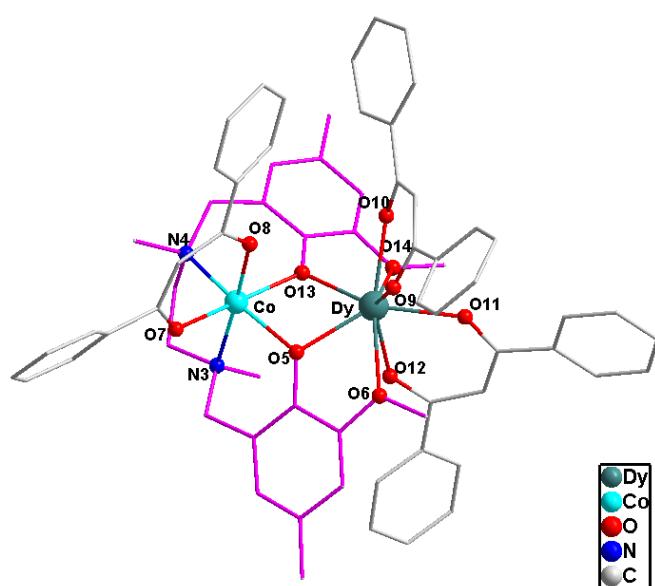
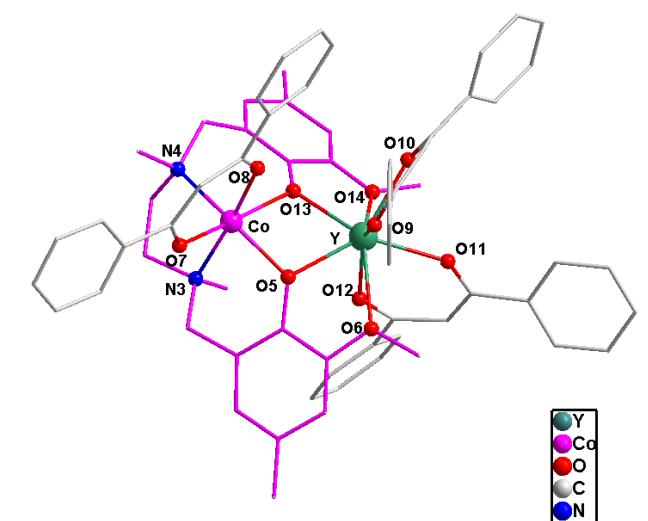
### Heterodinuclear M<sup>II</sup>-Ln<sup>III</sup> single molecule magnets constructed from exchange-coupled single ion magnets

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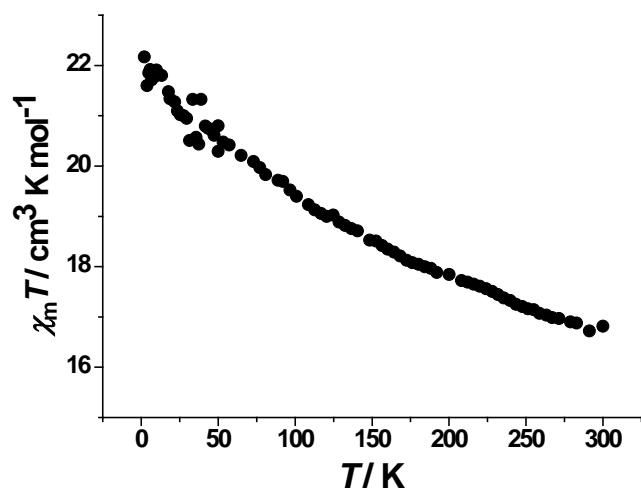
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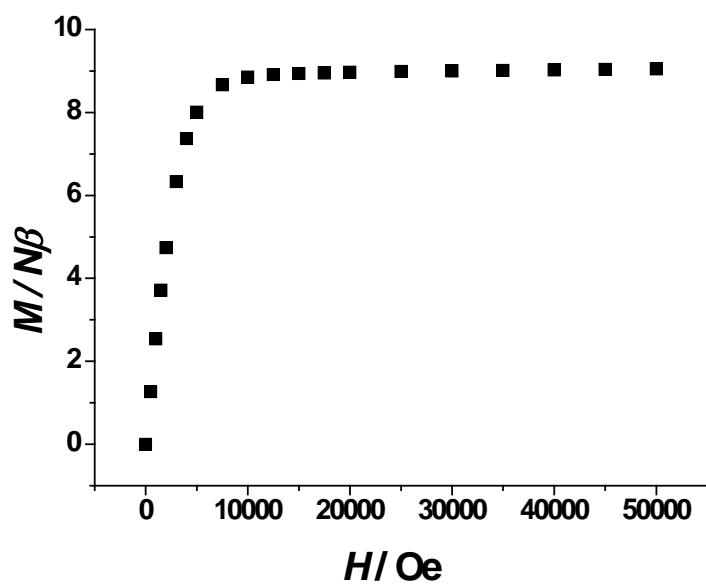
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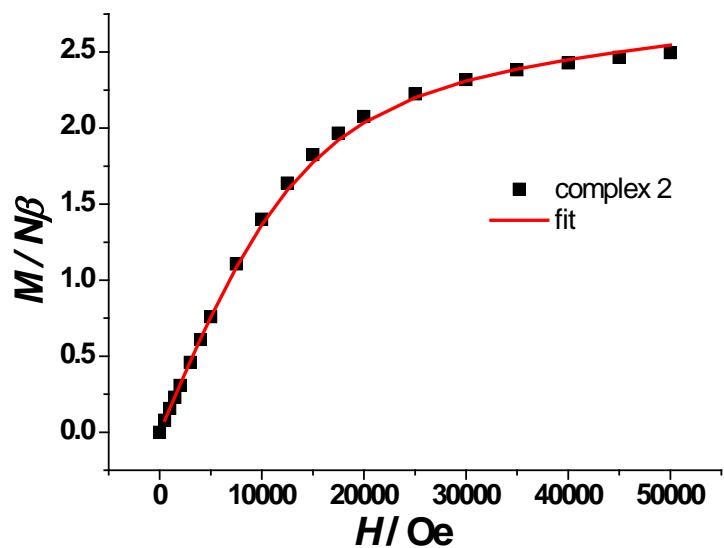
**Fig. S1.** Crystal structure of complexes 2-4.



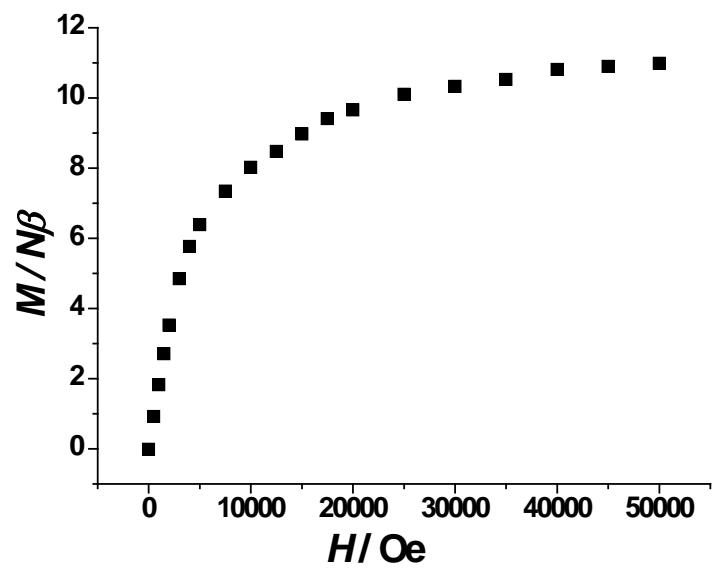
**Fig. S2.** Temperature dependence of  $\chi_m T$  per  $\text{Zn}^{\text{II}}\text{Dy}^{\text{III}}$  unit for a diluted sample  $\text{ZnDy}_{0.0885}\text{Y}_{0.9115}$  ( $\text{Dy:Y} = 1:10.3$  determined by ICP analysis) of complex **1**.



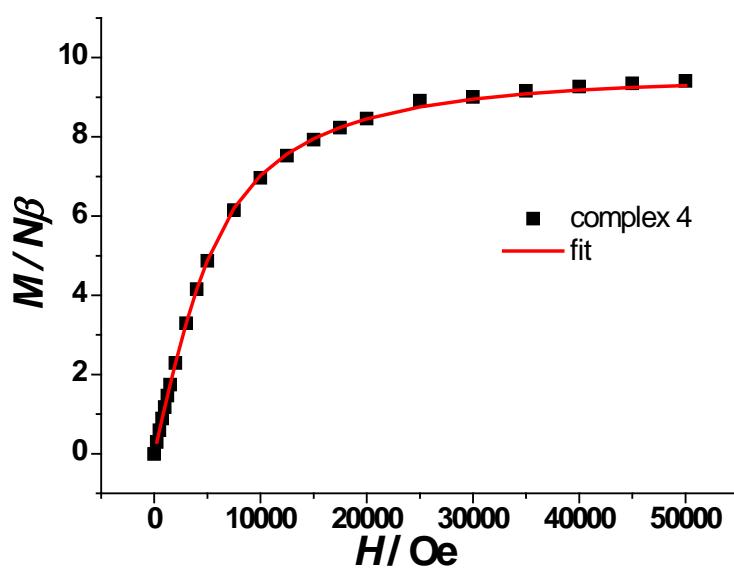
**Fig. S3.** Field dependence of magnetization for complex **1** at 2.0 K.



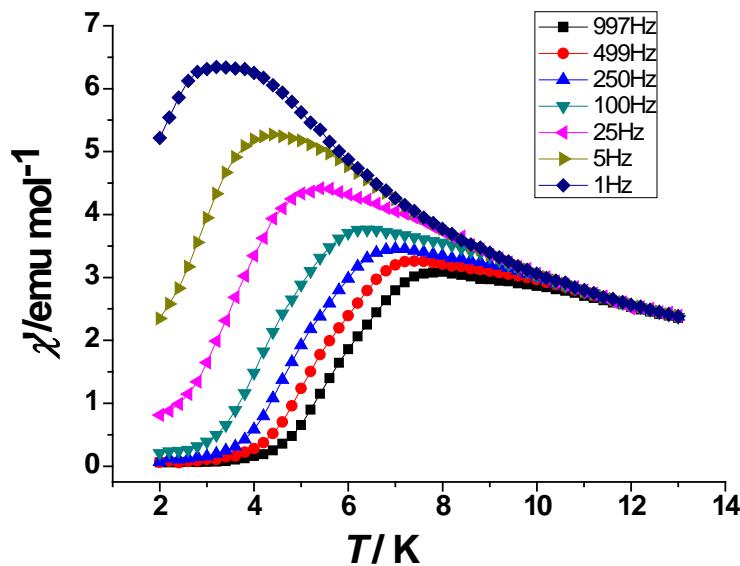
**Fig. S4.** Field dependence of magnetization for complex **2** at 2.0 K. The red line is the best fitting result with  $g = 2.39$ ,  $D = 10.3 \text{ cm}^{-1}$  and  $E = 4 \times 10^{-4} \text{ cm}^{-1}$ .



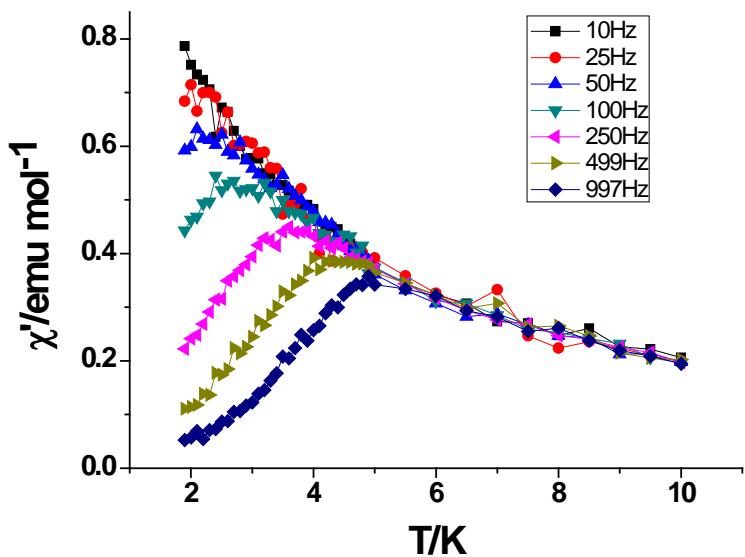
**Fig. S5.** Field dependence of magnetization for complex **3** at 2.0 K.



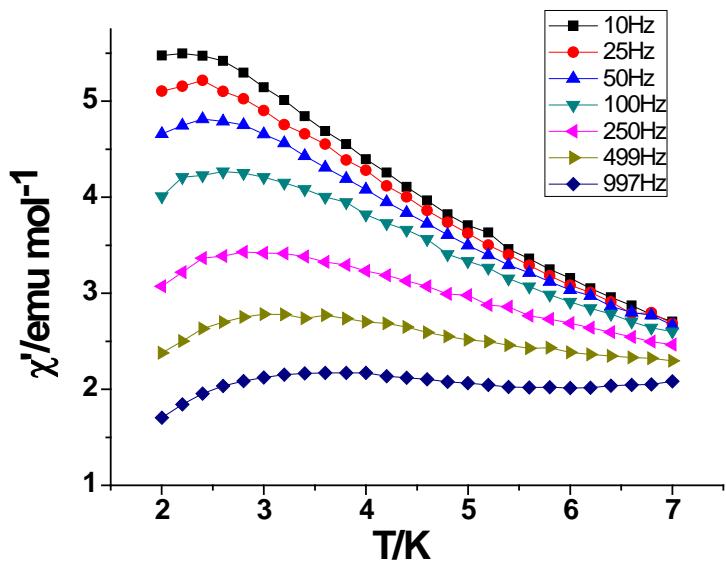
**Fig. S6.** Field dependence of magnetization for complex **4** at 2.0 K. The red line is the best fitting result with  $g = 1.89$  and  $D = 0.17 \text{ cm}^{-1}$ .



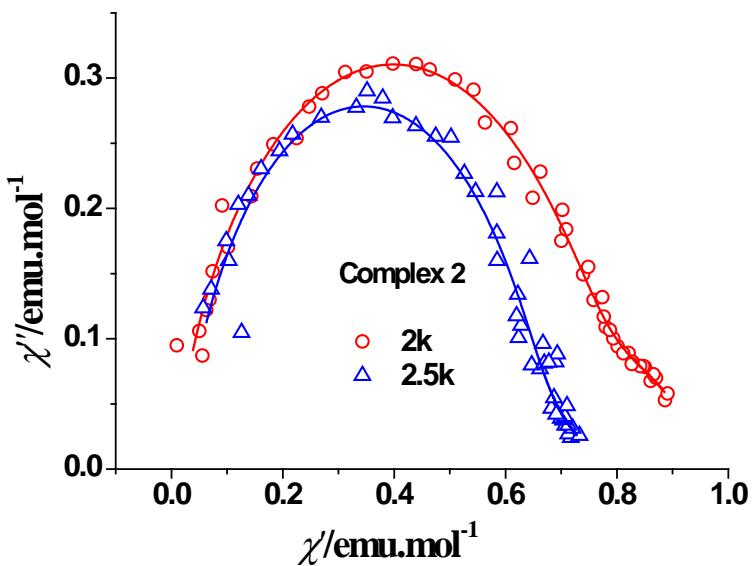
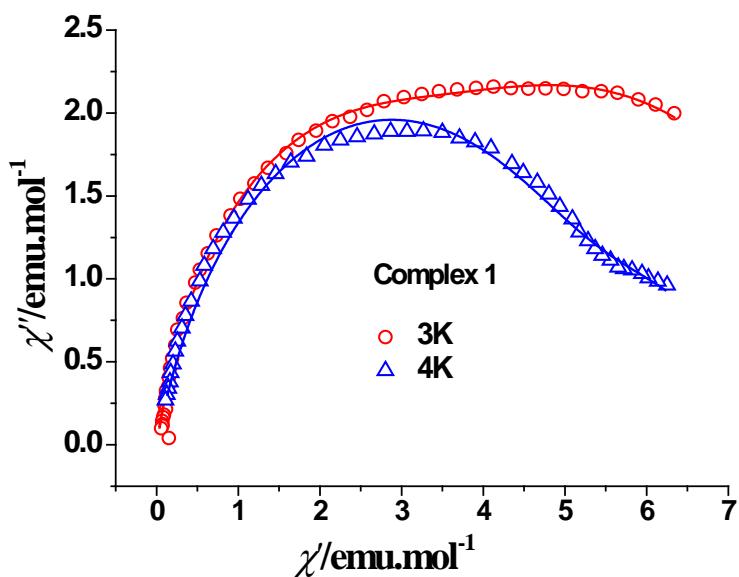
**Fig. S7.** Temperature dependence of in-phase magnetic susceptibility of complex **1** under 2 kOe dc field.



**Fig. S8.** Temperature dependence of in-phase magnetic susceptibility of complex **2** under 2 kOe dc field.



**Fig. S9.** Temperature dependence of in-phase magnetic susceptibility of complex **3** under 2 kOe dc field.



**Fig. S10.** Cole-Cole plots in 2 kOe applied dc field for complexes **1** (top) and **2** (bottom). The solid lines represent the best fit by using the parameters in Tables S1 and S2, respectively.

**Table S1.** Relaxation Fitting Parameters from Least-Squares Fitting of  $\chi(\omega)$  data for Complex **1**

$T$ (K)	$\chi_s$	$\chi_t$	$\beta$	$\alpha_1$	$\tau_1$	$\alpha_2$	$\tau_2$
3	8.05E-17	8.82	0.30	0.11	0.048	0.31	0.64
4	6.92E-18	7.53	0.33	0.36	0.91	0.22	0.024

**Table S2.** Relaxation Fitting Parameters from Least-Squares Fitting of  $\chi(\omega)$  data for Complex **2**

$T$ (K)	$\chi_s$	$\chi_t$	$\beta$	$\alpha_1$	$\tau_1$	$\alpha_2$	$\tau_2$
2	0.0049	0.95	0.79	0.14	0.0072	0.38	0.40
2.5	2.32E-16	0.77	0.71	0.059	0.0041	0.62	0.034

The corresponding expression of molar magnetic susceptibility for complex **4** is given in Eq. (S1).

$$\hat{H} = -2J_{\text{CoGd}} \hat{S}_{\text{Gd}} \hat{S}_{\text{Co}}$$

$$\chi_m = \frac{N\beta^2}{3k(T-\theta)} \cdot \frac{330g_{(5)}^2 \exp(24x) + 180g_{(4)}^2 \exp(14x) + 84g_{(3)}^2 \exp(6x) + 30g_{(2)}^2}{11\exp(24x) + 9\exp(14x) + 7\exp(6x) + 5} \quad (\text{S1})$$

$$x = J_{\text{CoGd}} / kT,$$

$$g_{(5)} = \frac{7}{10}g_{\text{Gd}} + \frac{3}{10}g_{\text{Co}},$$

$$g_{(4)} = \frac{8}{10}g_{\text{Gd}} + \frac{2}{10}g_{\text{Co}},$$

$$g_{(3)} = g_{\text{Gd}},$$

$$g_{(2)} = \frac{3}{2}g_{\text{Gd}} - \frac{1}{2}g_{\text{Co}}$$