## **Supporting Information for**

## Weak exchange coupling effects leading to fast magnetic relaxations in a trinuclear dysprosium single-molecule magnet

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Table S1: Crystallographic data and refinement for 1Dy<sub>3</sub> and 2Dy. (Note: The disordered toluene was squeezed using PLATON program, details are provided in the corresponding cif document)

	1Dy <sub>3</sub>	2Dy
Mr	2072.93	653.67
formula	$C_{86}H_{109}B_3Dy_3N_{27}O_2$	$C_{23}H_{25}BDyN_{12}$
cryst syst	monoclinic	orthorhombic
space group	$P2_{1}/c$	Pnma
<i>a</i> , Å	14.6588(5)	25.1963(7)
b, Å	22.3712(7)	10.9647(4)
<i>c</i> , Å	28.6408(8)	11.6734(3)
<i>V</i> , Å <sup>3</sup>	9211.4(5)	3225.03(18)
$\alpha$ , deg	90	90
$\beta$ , deg	101.265(3)	90
γ, deg	90	90
Ζ	4	4
<i>Т</i> , К	180(2)	180(2)
$\mu$ , mm <sup>-1</sup>	2.470	2.348
λ, Å	0.71073	0.71073
Cryst size, mm <sup>3</sup>	0.24 * 0.12 * 0.09	0.35 * 0.23 * 0.17
GOF	1.114	1.052
R <sub>int</sub>	0.0780	0.0443
$R_1$ , $wR_2[I > 2\sigma(I)]$	0.0591, 0.0986	0.0274, 0.0389
$R_1$ , w $R_2$ [all data]	0.1383, 0.1601	0.0649, 0.0724

KDe		$1Dy_3(1)$			$1\mathrm{Dy}_3(2)$			$1\mathrm{Dy}_3(3)$			
	E		E			E					
1	0.0			0.0			0.0				
2	186.6			194.0			197.8				
3	225.3				237.7		234.9				
4	262.5			279.0			280.2				
5	307.8			314.5			324.4				
6	360.7			348.8			374.6				
7	375.5			399.0			397.4				
8	479.0			543.6			493.1				
KDs	$g_x$	$g_y$	$g_z$	$g_x$	$g_y$	$g_z$	$g_x$	$g_y$	$g_z$		
1	0.005	0.029	19.715	0.0006	0.018	19.730	0.001	0.0200	19.735		
2	1.583	2.968	14.582	0.810	1.713	15.537	1.460	2.779	14.345		
3	1.582	5.573	12.166	1.758	5.029	12.158	1.167	5.065	11.991		
4	1.004	4.521	8.328	1.703	2.465	8.650	2.458	4.651	8.851		
5	2.174	3.760	13.964	10.833	7.070	2.899	1.913	3.412	14.328		
6	0.488	3.883	13.440	0.224	0.414	18.868	0.768	1.772	15.208		
7	0.320	2.731	15.884	0.488	0.680	16.083	0.193	0.862	18.315		
8	0.047	0.079	19.286	0.031	0.055	19.396	0.048	0.080	19.337		
KDs		2Dy									
		Ε									
1	0.0										
2	187.6										
3	212.0										
4	225.7										
5	264.1										
6	299.8										
7	391.2										
8	628.8										
KDs	$g_x$	$g_y$	$g_z$								
1	0.005	0.033	19.757								
2	1.264	2.810	15.235								
3	10.648	7.263	3.395								
4	1.122	1.458	14.952								
5	2.455	4.852	9.333								
6	1.316	1.547	17.559								
7	0.425	0.514	16.336								
8	0.015	0.024	19.514								

**Table S2**: Kramers doublets (KDs) energy spectrum (cm<sup>-1</sup>), *g*-tensor of ground and excited KDs for each Dv ion in  $1Dv_3$  and  $2Dv_3$ .

Figure S1: (a) Cole-Cole plots fitting for the determination of the temperature dependence of  $\tau$  for 2Dy in the absence of dc field; (b) Cole-Cole plots fitting for the determination of the temperature dependence of  $\tau$  for 2Dy' in the absence of dc field







Figure S3: (a) Cole-Cole plots fitting for the determination of the temperature dependence of  $\tau$  for 2Dy under 1 kOe dc field; (b) Cole-Cole plots fitting for the determination of the temperature dependence of  $\tau$  for 2Dy' under 1 kOe dc field



**Figure S4**. Out-of-phase signals  $(\chi_m)$  versus frequency (v) plots for **1Dy**<sub>3</sub> in the absence of dc field from 1 Hz to 1000 Hz. Cole-Cole plots fitting for the determination of the temperature dependence of  $\tau$  for **1Dy**<sub>3</sub> in the absence of dc field.



Figure S5. Temperature dependence ac susceptibility under 3 kOe dc field for 1Dy<sub>3</sub>.







Figure S7. Frequency dependence ac susceptibility in the absence of dc field for magnetically diluted  $1Dy_3$ .



**Figure S8**. Frequency dependence ac susceptibility under 1 kOe dc field for magnetically diluted **1Dy**<sub>3</sub>.



**Figure S9**. Cole-Cole plots fitting for the determination of the temperature dependence of  $\tau$  for diluted **1Dy**<sub>3</sub> in the absence of dc field (a) and under 1 kOe dc field.



**Figure S10**. Relaxation times ( $\tau$ ) versus  $T^{-1}$  plots for diluted **1Dy**<sub>3</sub>.





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**Figure S12**. (a) The angular-dependent susceptibility plots and simultaneous fitting under 1 kOe dc field; (b) fitted *g* value along main magnetic axes using linear fitting function: y = ax corresponding to the equation:  $g = \sqrt{\frac{32\chi_m T}{3}}$ .

