Supporting Information

Single-molecule magnet behavior in octanuclear dysprosium(III) aggregate inherited from helical triangular Dy_3 SMM-building blocks

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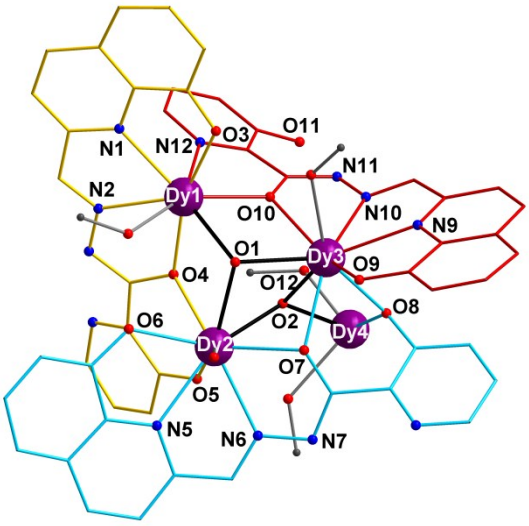

Fig. S1 Asymmetric unit of complex **1**.

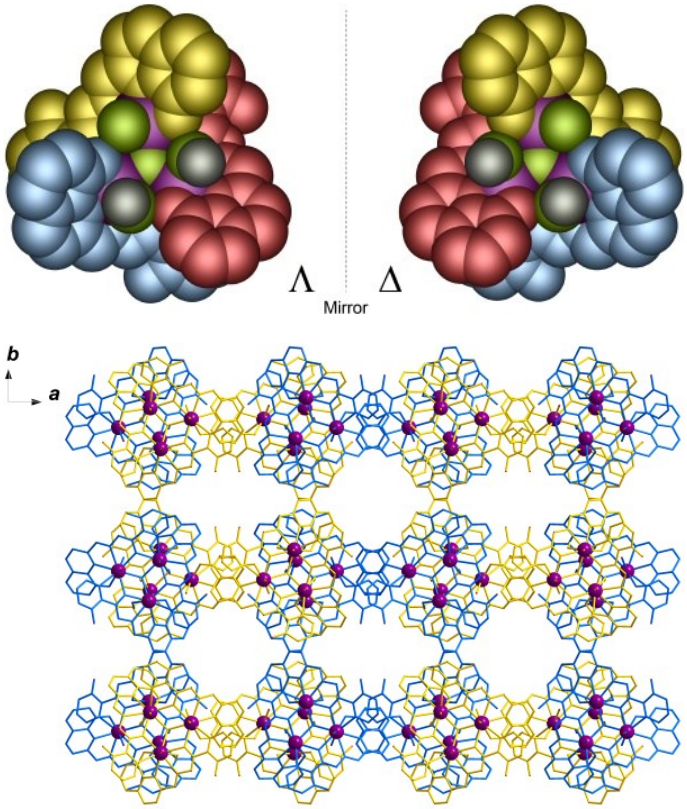

Fig. S2 Top: Space-filling representations of the left- (Λ) and right-hand (Δ) configurations of the circular-helical Dy_3 triangles present in complex **2a**. Bottom: packing arrangement of the molecules along *c* axis showing the different stereoisomers. Blue and yellow molecules correspond to the left- and right-hand configurations, respectively.

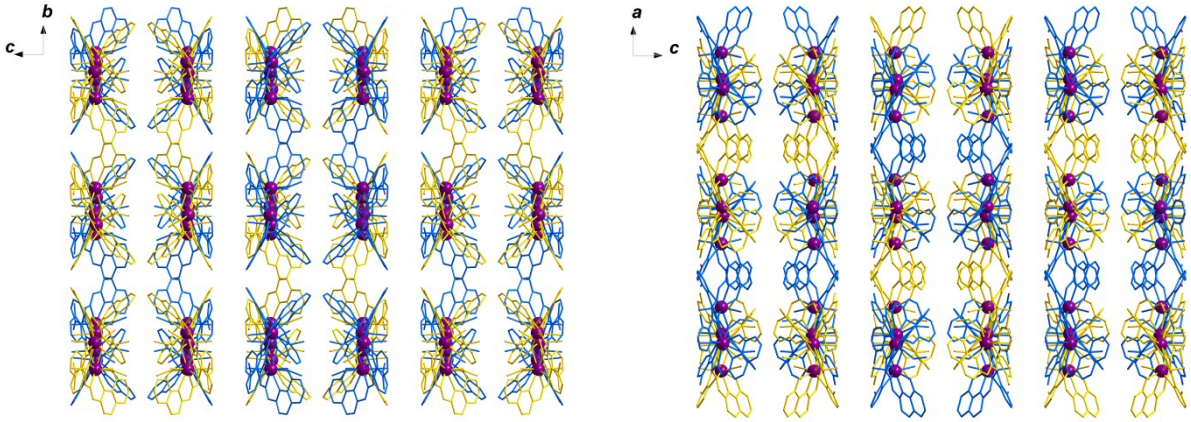

Fig. S3 Packing arrangement of the molecules along *a* and *b* axis showing the different stereoisomers for complex **2a**. Blue and yellow molecules correspond to the left- and right-hand configurations, respectively.

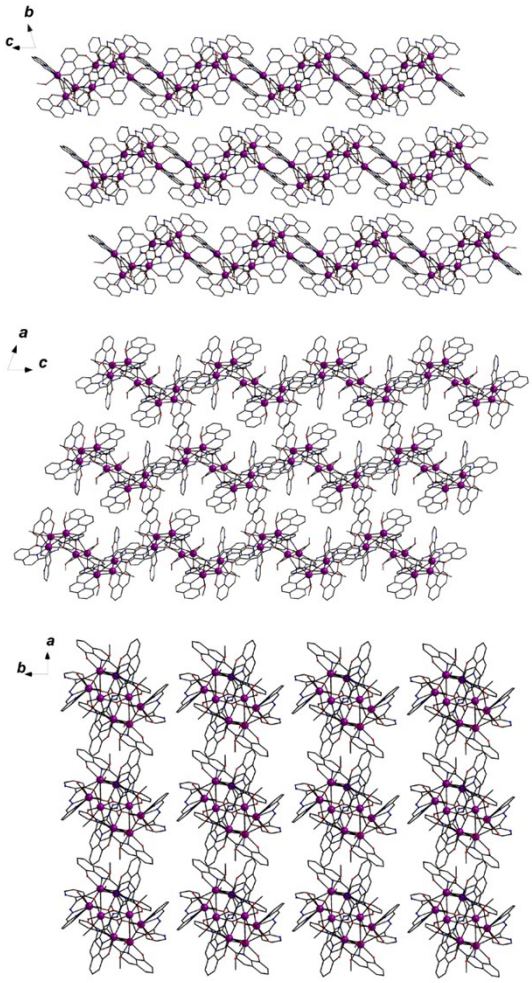

Fig. S4 Packing arrangement along the crystallographic *a*, *b*, and *c*-axis for complex **1**.

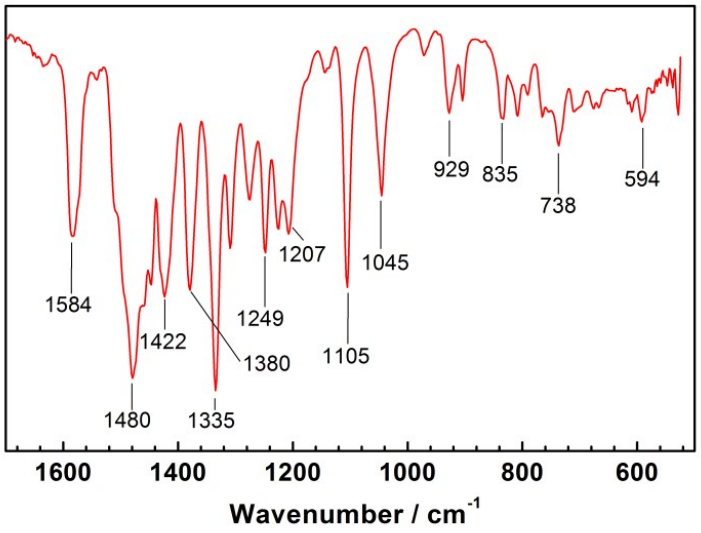

Fig. S5 Infrared spectrum of **1**.

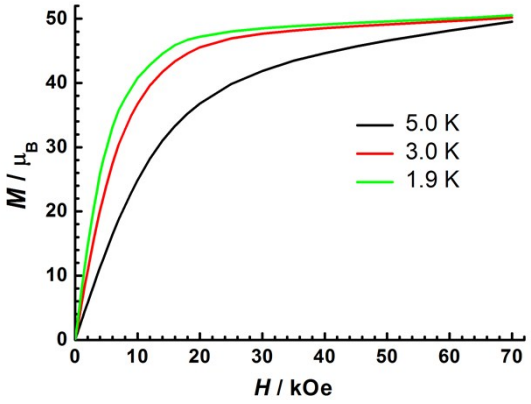

Fig. S6 Plots of the magnetization *M* versus *H* in the field range 0-70 kOe and the temperature range of 1.9-5.0 K for **1**.

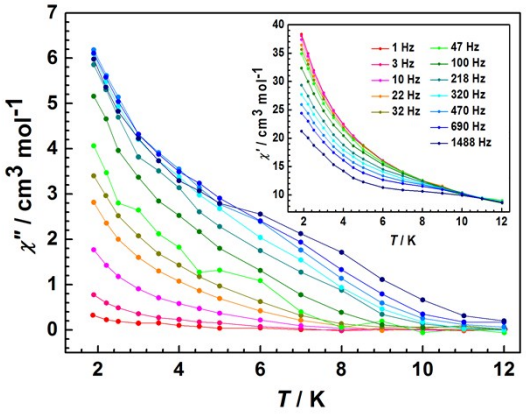

Fig. S7 Temperature dependence of the out-of-phase ac susceptibility under zero dc field for **1**. Inset: corresponding temperature dependence of the in-phase ac susceptibility.

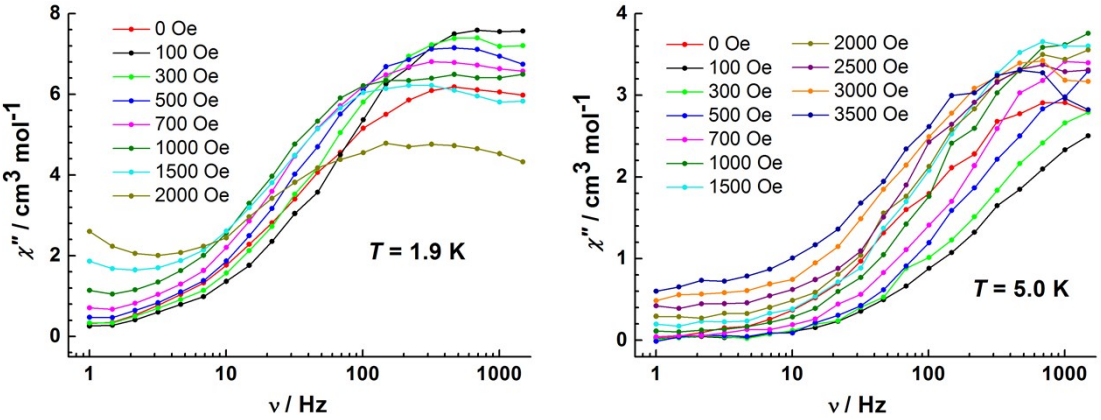

Fig. S8 Out-of-phase ac susceptibility (χ'') collected on **1** at 1.9 K (left) and 5.0 K (right) under the indicated dc field.

Table S1. Lanthanide(III) single-molecule magnets with high nuclearity (Ln_n, n > 5).

compound[a]	structural feature	U_{eff} / K	ref
$[Dy_6(\mu_3\text{-}OH)_4(L^1)_4(L^2)_2(H_2O)_9Cl]Cl_5\cdot 15H_2O$	two Dy_3 triangles in a vertex-to-vertex fashion	200	1
$[Dy_6(\mu_3\text{-}OH)_4(ovn)_4(avn)_2(NO_3)_4(H_2O)_4](NO_3)_2\cdot(H_2O)\cdot 3(CH_3)_2CO$	two Dy_3 triangles in a vertex-to-vertex fashion	9.6	2
$[Dy_6(L^3)_4(\mu_4\text{-}O)(NO_3)_4(CH_3OH)]\cdot CH_3OH$	two Dy_3 triangles in a edge-to-edge fashion	33.9, 40.7	3
$[Dy_6(L^4)_4(\mu_3\text{-}OH)_4(CH_3OH)_2(NO_3)_2]\cdot 6CH_3CN$	two Dy_3 triangles in a edge-to-edge fashion	3.2	4
$[\{Dy_6(L^5)_2(L^5H)_2\}(\mu_3\text{-}OH)_4][MeOH]_2[H_2O]_6[Cl]_4\cdot 8H_2O\cdot 4CH_3OH$	two Dy_3 triangles in a edge-to-edge fashion	46.2 (5 kOe dc field)	5
$[Dy_6(L^6)_2(\mu_3\text{-}OH)_4(\mu_2\text{-}OH)_2(SCN)_8(H_2O)_4]\cdot 6CH_3CN\cdot 2CH_3OH\cdot H_2O$	two Dy_3 triangles in a planar Dy_3+Dy_3 fashion	116	6
$[Dy_6(L^6)_2(\mu_3\text{-}OH)_4(\mu_2\text{-}OH)_2(NO_3)_6(H_2O)_6]\cdot 2NO_3\cdot 10H_2O$		181	
$[Dy_6(\mu_3\text{-}OH)_3(\mu_3\text{-}CO_3)(\mu\text{-}OMe)(HL^7)_6(MeOH)_4(H_2O)_2]_3\cdot 3MeOH\cdot 2H_2O$	vertex- and edge-sharing Dy_3 triangles	37.9	7
$[Dy_6(\mu_3\text{-}CO_3)_2(L^8)_5(HL^8)(MeOH)_2]\cdot 4H_2O\cdot 5MeOH\cdot EtOH$	Trigonal Prism	56	8
$[Dy_6(ovph)_4(Hpvph)_2Cl_4(H_2O)_2(CO_3)_2]\cdot CH_3OH\cdot H_2O\cdot CH_3CN$	Trigonal Prism	76	9
$[Dy_6(apovh)_4(Hapovh)_4(CO_3)_2(SCN)_2]\cdot 6CH_3CN\cdot 8CH_3OH\cdot 2H_2O$	Dy_3 molecular cluster pair	29	10
$[Dy_6(L^9)_6(L^{11})_6(OCH_3)_6\cdot 2CH_3OH$	Dy_6 wheels	12.2	11
$[Dy_6(L^{10})_6(L^{11})_6(OCH_3)_6\cdot 2CH_3OH$		11.5	
$[Dy_7(OH)_6(thmeH_2)_5(thmeH)(tpa)_6(MeCN)_2](NO_3)_2$	centred-hexagonal Dy_7 core	140	12
$[Dy_8(\mu_3\text{-}CO_3)_4(L^8)_8(H_2O)_8]\cdot 10MeOH\cdot 2H_2O$	quadruple-CO_3^{2-} bridged Dy_8	74.2	13
$Dy_8(\mu_3\text{-}OH)_4(L_{12})_4(DEA)_4Cl_4$	butterfly-shaped Dy_8	49.3 (5 kOe)	14
$Dy_8(\mu_3\text{-}OH)_4(L_{13})_6(DMF)_4(H_2O)_8$	Dy_8 core in a staircase-type arrangement	36.5 (5 kOe)	
$[Dy_7(EDDC)(opch)_4(O_3PC_{10}H_7)_3(OAc)_5(MeOH)_2]\cdot 4MeOH$	cyclic Dy_7	74, 177	15
$[Dy_{14}(EDDC)_4(opch)_4(O_3PC_{10}H_7)_{10}(OAc)_6(H_2O)_4]\cdot xH_2O$	dimer of cyclic Dy_7	20.7	
$[Dy_{10}(L^{14}H)_{10}(\kappa^2\text{-}Piv)_{10}]\cdot 9CHCl_3\cdot 4CH_3CN$	Dy_{10} wheels	16.1	16

[a] *Abbreviations*: HL^1, o-vanillin; H_2L^2, 2-hydrox-ymethyl-6-methoxyphenol; ovnH, o-vanillin; $avnH_2$, aldol-vanillin; H_3L^3, 2,6-bis((2-hydroxyethylimino)methyl)-4-methylphenol; H_3L^4, 1,3-bis(salicylideneamino)-2-propanol; L^5H_3, *N′*-(2-hydroxy-3-methoxybenzylidene)-6-(hydroxymethyl)picolinohydrazide); H_2L^6, 6,6'-((1*E*,1'*E*)-(2,2'-(pyrimidine-4,6-diyl)bis(hydrazin-2-yl-1-ylidene))bis(methanylylidene))bis(2-methoxyphenol); H_2L^7, (*E*)-*N′*-(2-hydroxybenzylidene)pyrazine-2-carbohydrazide; H_2L^8, (*E*)-*N′*-(2-hyborxy-3-methoxybenzylidene) pyrazine-2-carbohydrazide; H_2ovph, o-vanillin picolinoylhydrazone; H_2apovh, (*N′*-(amino(pyridin-2-yl) methylene)-o-vanilloylhydrazine); L^9H, pivalic acid; L^{10}H, 3,5-dinitrobenzoic acid; L^{11}H, 2,6-dimethoxyphenol; $thmeH_3$, tris(hydroxymethyl)ethane; tpaH, triphenylacetic acid; H_2EDDC, (*N′,N″,E,N′,N″,E*)-*N′*,*N″*- (ethane-1,2-diylidene)dipyrazine-2-carbohydrazide; H_2opch (*E*)-*N′*-(2-hydroxy-3-methoxybenzylidene) pyrazine-2-carbohydrazide; H_2L^{12}, 3-(pyridin-2-yl)- *N′*-((pyridin-2-yl)methylene)-1*H*-pyrazole-5-carbohydrazide; H_3L^{13}, *N′*-(2-hydroxybenzylidene)-3-(pyr-idin-2-yl)-1H-pyrazole-5-carbohydrazide; $L^{14}H_3$, (E)-2-((2-hydroxy-ethylimino)methyl)-6-(hydroxymethyl)-4-methylphenol.

Table S2. Selected bond distances (Å) and angles (deg) in complex **1**.

Dy(1)-O(1)	2.384(5)	Dy(3)-O(1)	2.378(5)
Dy(1)-O(3)	2.278(6)	Dy(3)-O(2)	2.405(5)
Dy(1)-O(4)	2.342(5)	Dy(3)-O(7)	2.446(5)
Dy(1)-O(10)	2.326(5)	Dy(3)-O(8)	2.391(5)

Dy(1)-O(13)	2.409(5)	Dy(3)-O(9)	2.327(6)
Dy(1)-N(1)	2.420(7)	Dy(3)-O(10)	2.483(5)
Dy(1)-N(2)	2.465(6)	Dy(3)-O(15)	2.448(6)
Dy(1)-N(12)	2.537(7)	Dy(3)-N(9)	2.511(6)
Dy(2)-O(1)	2.383(5)	Dy(3)-N(10)	2.531(6)
Dy(2)-O(2)	2.521(5)	Dy(4)-O(2)	2.352(5)
Dy(2)-O(4)	2.570(5)	Dy(4)-O(8)	2.341(6)
Dy(2)-O(5)	2.285(6)	Dy(4)-O(12)	2.271(5)
Dy(2)-O(6)	2.336(5)	Dy(4)-O(16)	2.376(5)
Dy(2)-O(7)	2.402(5)	Dy(4)-O(11)#1	2.228(5)
Dy(2)-O(14)	2.411(6)	Dy(4)-O(12)#1	2.275(5)
Dy(2)-N(5)	2.491(7)	Dy(4)-N(11)#1	2.607(7)
Dy(2)-N(6)	2.514(7)	Dy(4)-Dy(4)#1	3.6928(8)
Dy(1)-Dy(2)	3.8832(5)	Dy(2)-Dy(3)	3.6379(5)
Dy(1)-Dy(3)	3.9632(5)	Dy(3)-Dy(4)	3.7913(5)
O(1)-Dy(1)-O(3)	92.5(2)	O(1)-Dy(3)-O(2)	73.29(17)
O(1)-Dy(1)-O(4)	74.28(18)	O(1)-Dy(3)-O(7)	72.27(17)
O(1)-Dy(1)-O(10)	69.07(17)	O(1)-Dy(3)-O(8)	135.60(18)
O(3)-Dy(1)-O(4)	164.40(19)	O(1)-Dy(3)-O(10)	66.60(17)
O(3)-Dy(1)-O(10)	81.81(19)	O(2)-Dy(3)-O(7)	63.82(18)
O(4)-Dy(1)-O(10)	85.68(18)	O(2)-Dy(3)-O(8)	69.29(17)
O(1)-Dy(2)-O(2)	71.14(17)	O(2)-Dy(3)-O(10)	68.83(17)
O(1)-Dy(2)-O(4)	70.24(18)	O(7)-Dy(3)-O(8)	70.33(17)
O(1)-Dy(2)-O(7)	72.96(18)	O(7)-Dy(3)-O(10)	123.86(17)
O(2)-Dy(2)-O(4)	68.65(16)	O(8)-Dy(3)-O(10)	118.23(18)
O(2)-Dy(2)-O(5)	75.86(18)	Dy(1)-O(1)-Dy(2)	109.1(2)
O(2)-Dy(2)-O(7)	62.72(18)	Dy(1)-O(4)-Dy(2)	104.4(2)
O(4)-Dy(2)-O(7)	125.95(17)	Dy(2)-O(1)-Dy(3)	99.64(18)
Dy(2)-Dy(1)-Dy(3)	55.233(9)	Dy(2)-O(2)-Dy(3)	95.17(18)
Dy(1)-Dy(2)-Dy(3)	63.501(10)	Dy(2)-O(7)-Dy(3)	97.23(18)
Dy(2)-Dy(3)-Dy(4)	75.593(11)	Dy(1)-O(1)-Dy(3)	112.7(2)
Dy(1)-Dy(3)-Dy(2)	61.267(10)	Dy(1)-O(10)-Dy(3)	111.0(2)
Dy(1)-Dy(3)-Dy(4)	102.341(12)	Dy(2)-O(2)-Dy(4)	138.3(2)
Dy(4)#1-Dy(4)-Dy(3)	93.435(14)	Dy(3)-O(2)-Dy(4)	105.69(19)
Dy(4)-O(12)-Dy(4)#1	108.7(2)	Dy(3)-O(8)-Dy(4)	106.5(2)

Symmetry codes: #1 -x+2,-y+2,-z

Table S3. Results of continuous shape measures analysis for the Dy^{III} coordination spheres.

Dy1 TDD-8 (D_{2d})	Dy2 MFF-9 (C_s)	Dy3 MFF-9 (C_s)	Dy4 CTPR-7 (C_{2v})
2.034	1.194	1.488	1.140

TDD-8: Triangular dodecahedron

MFF-9: Muffin

CTPR-7: Capped trigonal prism

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