

Supplementary Information

A journey in lanthanide coordination chemistry: from evaporable dimers to magnetic materials and luminescent devices

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Table S1. Main geometric parameters of the dimers

Dimer	Dimensionality	Site symmetry and CShM	Dy-Dy distance (Å)	Remark	Ref
DyPyNO	0D	D_{4d} , 0.757	4.07(2)	Double-bridged dimer	1
Dy ₂ PyNO ₃	0D	Dy ₁ , C _{4v} , 0.927 Dy ₂ , C _{4v} , 0.605	3.86(1)	Triple-bridge dimer	2
[(Dy ₂ PyNO ₃)(biPyNO)] _∞	3D	Dy ₁ , C _{4v} , 0.828 Dy ₂ , C _{4v} , 0.682	3.94(1) 3.93(2)	3D assembly of triple-bridged dimers	2
DyttaPyNO	0D	D_{2d} , 0.403	4.15 (6)	Double-bridged dimer	3
161DyttaPyNO	0D	Idem	Idem-	Isostructural to DyttaPyNO	3
164DyttaPyNO	0D	Idem	Idem	Isostructural to DyttaPyNO	3
YDyttaPyNO	0D	Idem	Idem	Isostructural to DyttaPyNO	3
DyPyNOS1	0D	D_{4d} , 0.618	4.02(8)	Double-bridged dimer	4
DyPyNOS2	0D	D_{4d} , 0.345	4.05(2)	Double-bridged dimer	4
DyPyNO-NO ₂	0D	D_{4d} , 0.551	4.07(4)	Double-bridged dimer	5
DyPyNO-NO ₂ -film	Thick film	-	-	330 nm thick film on Teflon substrate	5

Table S2. Main magnetic figures of merit of the dimers

Dimer	g_z	Dy-Dy interaction (cm^{-1})*	H_{dc} (Oe)	τ_0 (s)	U_{eff} (K)	C ($\text{s}^{-1}\cdot\text{K}^{-1}$), n , and τ_{tun} (s) in $\tau^{-1} = CT^n + \tau_{tun}^{-1}$	α_{2K}	τ_{2K} (s)	Relaxing fraction ($1-(\chi_s/\chi_T)_{2K}$)	Ref
DyPyNO	18.69 exp 19.53 theo	$J_{exp} = -2.46$ (0.03) $J_{theo} = J_{dip} + J_{exch}$ $J_{theo} = -2.44 - 0.25 = -2.69$ $J_{crossing} = -3.36 \text{ cm}^{-1}$	0	1.87 (2.2) $\times 10^{-12}$	192 (1)	$C = 0.037$ (6), $n = 4.79$ (4), $\tau_{tun} = 0.55$ (1)	0.05	0.38	0.99	1
Dy ₂ PyNO ₃	n.a.	n.a.	0 1900	n.a. 3.18 (0.22) $\times 10^{-6}$	n.a. 24.4 (1)	n.a. n.a.	0.22 0.45	1.95×10^{-4} 0.01	0.57 0.94	2
[(Dy ₂ PyNO ₃)(biPyNO)] _∞	n.a.	n.a.	1900	1.25×10^{-5}	10.3	n.a.	0.51	1.11×10^{-3}	0.85	2
DyttaPyNO	19.59	$J_{exp} = 2.92$ $J_{theo} = J_{dip} + J_{exch}$ $= -2.36 - 0.50 = -2.86$	0	n.a.	n.a.	$C = 2.25$ (12) $\times 10^{-3}$ $n = 5.42$ (19) $\tau_{tun} = 2.2$ (1)	0.21	1.13	0.99	3
¹⁶¹ DyttaPyNO	**	**	0	n.a.	n.a.	$C = 0.00199$ (78) $n = 5.38$ (17) $\tau_{tun} = 1.8$ (1)	0.20	1.42	0.98	3
¹⁶⁴ DyttaPyNO	**	**	0	n.a.	n.a.	$C = 0.00219$ (11) $n = 5.34$ (18) $\tau_{tun} = 3.0$ (1)	0.20	2.04	0.99	3
YDyttaPyNO	n.a.	n.a.	0	1.81 (2.8) 10^{-7}	117(7)	$C = 0.149$ (1) $n = 3.27$ (1) $\tau_{tun} = 0.08$ (0.05)	0.32	0.09	0.97	3
DyPyNOS1	19.50	$J_{theo} = J_{dip} + J_{exch}$ $= -2.525 - 0.125 = -2.65$	0	n.a.	n.a.	$C = 0.200$ (249) $n = 4.05$ (54) $\tau_{tun} = 0.08$ (1)	0.24	0.061	0.91	4
DyPyNOS2	19.52	$J_{theo} = J_{dip} + J_{exch}$ $= -2.49 - 0.5 = -2.99$	0	n.a.	n.a.	$C = 0.279$ (148) $n = 4.12$ (29) $\tau_{tun} = 0.07$ (1)	0.28	0.047	0.97	4

DyPyNO₂	19.56	J _{exp} = 2.9	0	3.1 (1.5) 10 ⁻¹⁰	187 (3)	C=0.037(3) n=3.93(5)	0.42	1.54	0.96	⁵
DyPyNO₂-film	-	-		6.6 (2.2) 10 ⁻¹¹	166 (4)	C=0.104(6) n=4.25(4)**	0.24 **	0.128**	0.95	⁵

Fitted values may differ from the originally reported ones because up-to-date homogenous fitting procedure is applied.

*In the S=1/2 ground state approximation. ** isotopic substitution does not influence g values and magnetic coupling. *** Values at 4K.

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