

Electronic Supplementary Information

**Syntheses, Structures, and Magnetic Properties of Homodinuclear Lanthanide Complexes Based on
Dinucleating Schiff Base Ligands**

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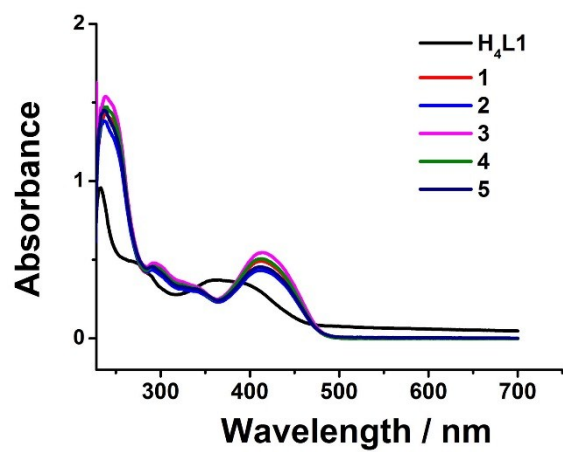


Fig. S1 UV-Vis absorption spectra for H_4L1 and complexes **1–5** in CH_2Cl_2 (5×10^{-5} M).

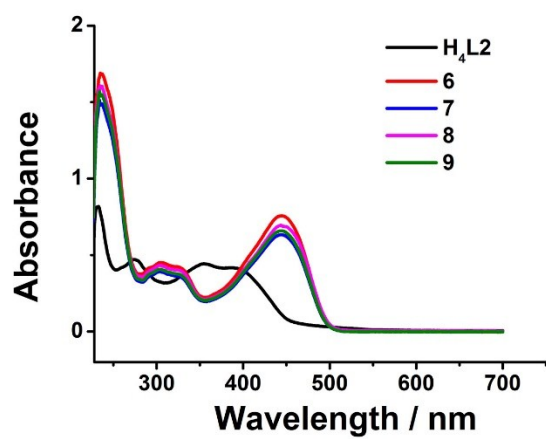


Fig. S2 UV-Vis absorption spectra for H_4L2 and complexes **6–9** in CH_2Cl_2 (5×10^{-5} M).

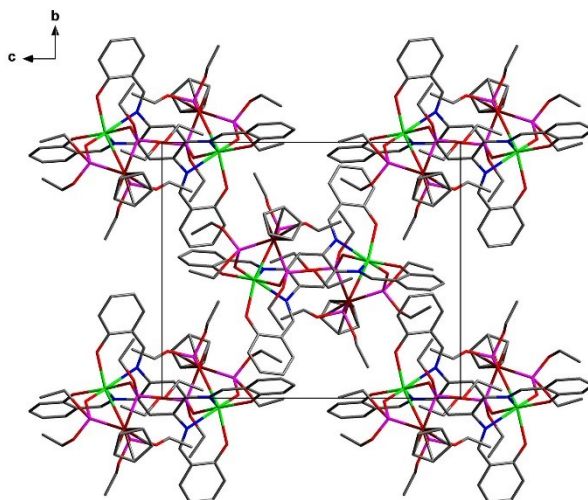


Fig. S3 Crystal packing of complexes **1–5** with hydrogen atoms omitted for clarity. (Ln (Ln = Dy for **1**, Tb for **2**, Ho for **3**, Gd for **4**, and Y for **5**), green; Co, brown; N, blue; P, purple; O, red; and C grey.)

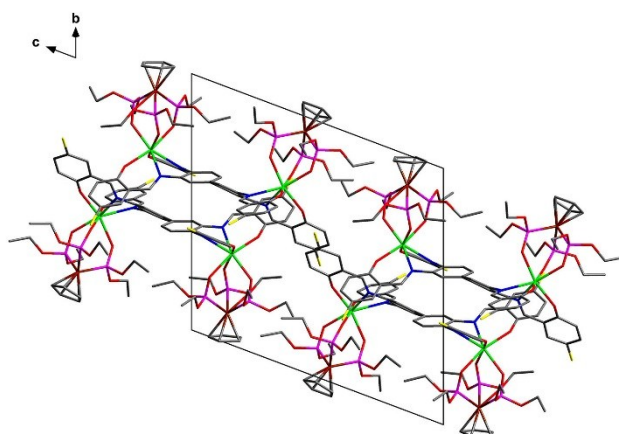


Fig. S4 Crystal packing of complexes **6–9** with hydrogen atoms omitted for clarity. (Ln (Ln = Dy for **6**, Tb for **7**, Ho for **8**, and Gd for **9**), green; Co, brown; Cl, yellow; N, blue; P, purple; O, red; and C grey.)

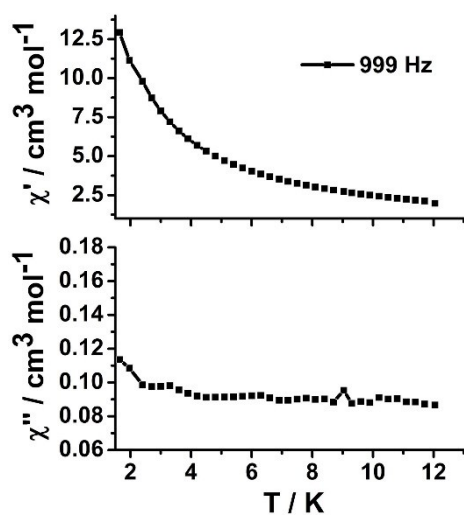


Fig. S5 Temperature-dependent in-phase (χ') and out-of-phase (χ'') ac susceptibilities for complex **1** under zero dc field in the frequency of 999 Hz. The solid lines are guides only.

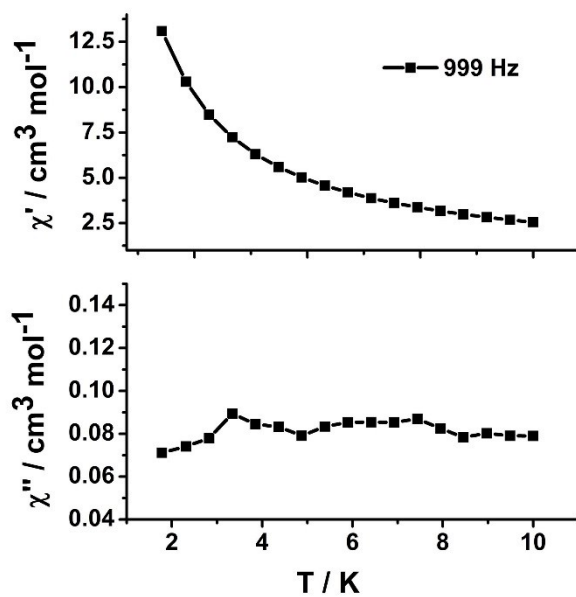


Fig. S6 Temperature-dependent in-phase (χ') and out-of-phase (χ'') ac susceptibilities for complex **6** under zero dc field in the frequency of 999 Hz. The solid lines are guides only.

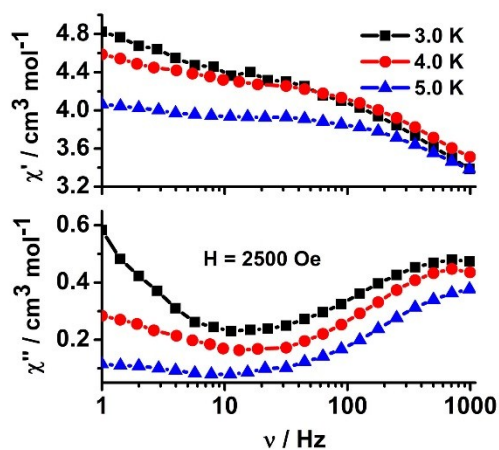


Fig. S7 Frequency-dependent in-phase (χ') and out-of-phase (χ'') ac susceptibilities from 3.0 K to 5.0 K for complex **1** under 2500 Oe dc field. The solid lines are guides only.

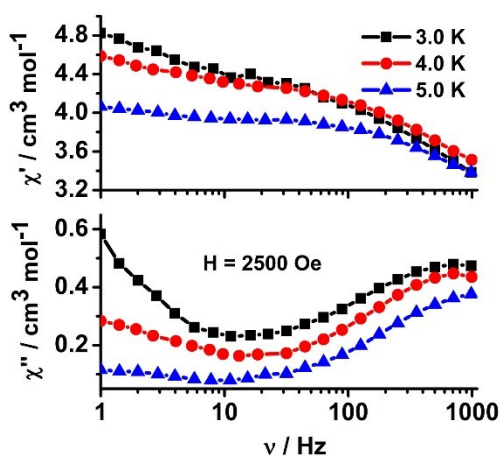


Fig. S8 Frequency-dependent in-phase (χ') and out-of-phase (χ'') ac susceptibilities from 3.0 K to 5.0 K for complex **6** under 2500 Oe dc field. The solid lines are guides only.

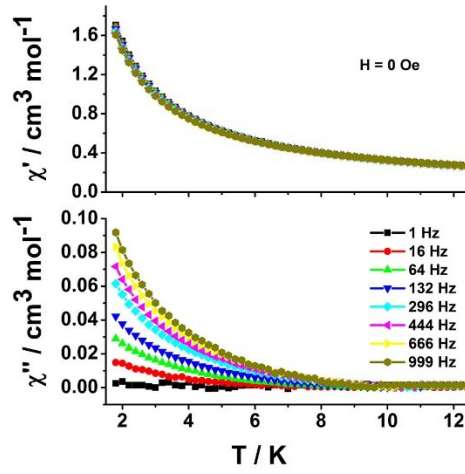


Fig. S9 Temperature-dependent in-phase (χ') and out-of-phase (χ'') ac susceptibilities for diluted-1 under zero dc field in the frequencies of 1–999 Hz. The solid lines are guides only.

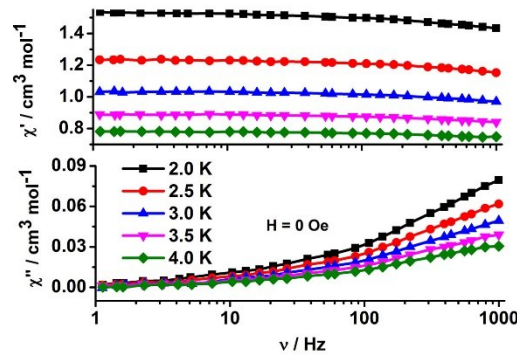


Fig. S10 Frequency-dependent in-phase (χ') and out-of-phase (χ'') ac susceptibilities from 2.0 K to 4.0 K for diluted-1 under zero dc field. The solid lines are guides only.

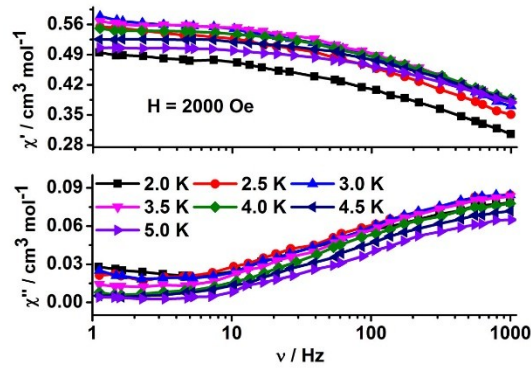


Fig. S11 Frequency-dependent in-phase (χ') and out-of-phase (χ'') ac susceptibilities from 2.0 K to 5.0 K for diluted-1 under 2000 Oe dc field. The solid lines are guides only.

Table S1 Selected bond lengths (Å) and angles (°) for complexes **1–9**.

	1	2	3	4	5
	Bond Distances (Å)				
Ln1–O3	2.293(3)	2.303(3)	2.308(3)	2.347(3)	2.275(2)
Ln1–O6	2.288(3)	2.326(3)	2.275(3)	2.315(3)	2.264(2)
Ln1–O9	2.315(3)	2.300(3)	2.274(3)	2.321(3)	2.303(2)
Ln1–O10	2.231(3)	2.207(3)	2.216(3)	2.218(3)	2.218(2)
Ln1–O11	2.193(3)	2.238(3)	2.187(3)	2.256(3)	2.191(2)
Ln1–N1	2.461(3)	2.486(3)	2.441(3)	2.506(3)	2.449(2)
Ln1–N2	2.478(3)	2.472(3)	2.465(3)	2.491(3)	2.483(2)
	Bond Angles (°)				
O3–Ln1–O6	82.45(10)	78.61(11)	78.70(11)	76.37(10)	81.99(8)
O6–Ln1–O9	76.82(10)	76.62(12)	82.38(11)	81.80(10)	77.38(7)
O3–Ln1–O9	78.58(10)	81.70(11)	76.90(11)	78.03(10)	78.46(8)
O10–Ln1–O11	90.69(10)	91.82(12)	90.35(11)	91.83(10)	90.26(8)
O10–Ln1–N1	73.75(10)	72.67(11)	73.99(11)	72.44(10)	73.46(8)
O11–Ln1–N1	123.33(10)	112.96(11)	123.60(11)	112.46(11)	122.56(8)
O10–Ln1–N2	113.15(10)	123.24(12)	113.53(11)	122.48(11)	113.34(8)
O11–Ln1–N2	73.15(10)	73.38(11)	73.38(11)	72.91(10)	72.55(8)
N1–Ln1–N2	64.88(10)	64.68(11)	65.19(12)	64.27(11)	65.35(8)
	6	7	8	9	
	Bond Distances (Å)				
Ln1–O3	2.311(4)	2.311(4)	2.270(4)	2.322(5)	
Ln1–O6	2.245(4)	2.227(4)	2.315(4)	2.246(5)	
Ln1–O9	2.301(4)	2.306(4)	2.210(4)	2.311(4)	
Ln1–O10	2.197(4)	2.201(4)	2.157(4)	2.210(4)	
Ln1–O11	2.235(4)	2.222(4)	2.217(4)	2.227(5)	
Ln1–N1	2.483(5)	2.486(5)	2.507(5)	2.491(6)	
Ln1–N2	2.498(5)	2.498(5)	2.481(5)	2.494(6)	
Ln2–O14	2.336(4)	2.325(4)	2.266(5)	2.325(5)	
Ln2–O17	2.366(5)	2.372(5)	2.272(5)	2.379(6)	
Ln2–O20	2.251(4)	2.246(4)	2.257(4)	2.256(5)	
Ln2–O21	2.234(4)	2.238(5)	2.143(4)	2.245(5)	
Ln2–O22	2.201(4)	2.188(4)	2.198(4)	2.186(5)	
Ln2–N3	2.527(5)	2.520(5)	2.497(5)	2.530(6)	
Ln2–N4	2.487(5)	2.485(5)	2.446(5)	2.476(5)	
	Bond Angles (°)				
O3–Ln1–O6	77.46(15)	77.33(15)	82.23(16)	76.35(18)	
O6–Ln1–O9	78.91(16)	78.59(16)	81.56(15)	78.74(17)	
O3–Ln1–O9	81.70(16)	81.79(15)	77.14(16)	81.41(16)	
O10–Ln1–O11	94.43(14)	94.61(14)	92.54(16)	94.20(17)	
O10–Ln1–N1	72.24(15)	71.89(15)	72.02(16)	72.57(17)	
O11–Ln1–N1	120.55(16)	120.37(16)	116.68(16)	119.61(18)	
O10–Ln1–N2	115.72(15)	115.68(16)	120.16(16)	115.66(17)	
O11–Ln1–N2	72.32(15)	72.26(15)	74.75(15)	71.70(18)	
N1–Ln1–N2	63.93(16)	63.96(16)	63.78(16)	63.56(18)	

O14–Ln2–O17	80.75(18)	80.96(17)	74.02(18)	80.3(2)
O17–Ln2–O20	73.42(18)	72.63(16)	81.45(16)	73.8(2)
O14–Ln2–O20	84.78(16)	84.42(15)	80.37(16)	84.15(18)
O21–Ln2–O22	90.94(16)	91.12(16)	90.52(16)	91.23(18)
O21–Ln2–N3	71.68(15)	71.42(16)	74.66(16)	71.59(19)
O22–Ln2–N3	116.64(16)	115.90(16)	118.88(15)	116.21(17)
O21–Ln2–N4	117.30(15)	116.99(15)	116.80(16)	117.05(17)
O22–Ln2–N4	74.60(15)	74.29(15)	73.50(16)	74.08(17)
N3–Ln2–N4	62.92(15)	62.50(16)	62.94(18)	62.72(18)

Table S2 Summary of structural parameters for complexes **1–5**.

Structural parameter (Å)	1	2	3	4	5
Average Ln–N bond distance	2.470	2.479	2.453	2.498	2.466
Average Ln–O bond distance	2.264	2.275	2.252	2.291	2.250
Ln to O3O6O9 plane center distance	1.555(8)	1.568(9)	1.545(9)	1.585(6)	1.543(3)
Ln to O1O011N1N2 plane center distance	1.199(8)	1.205(8)	1.186(9)	1.225(5)	1.202(3)
Centers distance for above two planes	2.725(6)	2.744(7)	2.704(7)	2.778(3)	2.721(5)
Shortest intermolecular Ln ³⁺ ...Ln ³⁺ distance	10.738(8)	10.760(6)	10.723(7)	10.891(6)	11.038(5)
Intramolecular Ln ³⁺ ...Ln ³⁺ distance	8.525(7)	8.538(5)	8.493(6)	8.583(5)	8.492(6)

Table S3 Summary of structural parameters for complexes **6–9**.

Structural parameter (Å)	6	7	8	9
Average Ln–N bond distance	2.499	2.497	2.483	2.498
Average Ln–O bond distance	2.267	2.263	2.231	2.271
Ln1 to O3O6O9 plane center distance	1.544(6)	1.543(6)	1.511(7)	1.558(3)
Ln1 to O1O011N1N2 plane center distance	1.205(6)	1.206(7)	1.198(7)	1.215(3)
Centers distance for above two planes	2.730(7)	2.731(7)	2.690(8)	2.755(3)
Ln2 to O14O17O20 plane center distance	1.560(7)	1.564(7)	1.544(8)	1.566(4)
Ln2 to O21O22N3N4 plane center distance	1.233(7)	1.240(8)	1.199(8)	1.237(4)
Centers distance for above two planes	2.757(6)	2.767(7)	2.708(7)	2.769(3)
Shortest intermolecular Ln ³⁺ ...Ln ³⁺ distance	7.886(6)	7.861(6)	7.790(7)	7.866(6)
Intramolecular Ln ³⁺ ...Ln ³⁺ distance	12.164(7)	12.094(7)	12.102(8)	12.080(7)

Table S4 Parameters obtained by continuous shape measure (CShM) method for the study of seven-coordinated Ln(III) coordination sphere for complexes **1–5**. (The *S* values indicate the proximity to the selected ideal polyhedron, *S* = 0 corresponds to the non-distorted polyhedron).

	1-<i>S</i>_{Dy1}	2-<i>S</i>_{Tb1}	3-<i>S</i>_{Ho1}	4-<i>S</i>_{Gd1}	5-<i>S</i>_{Y1}
Capped octahedron (COC-7)	1.65	1.72	1.61	1.73	1.59
Capped trigonal prism (CTPR-7)	2.05	2.05	2.05	1.96	1.89
Pentagonal bipyramid (PBPY-7)	3.87	3.86	3.91	3.91	4.09

Table S5 Parameters obtained by continuous shape measure (CShM) method for the study of seven-coordinated Ln(III) coordination sphere for complexes **6–9**.

	6-<i>S</i>_{Dy1}	6-<i>S</i>_{Dy2}	7-<i>S</i>_{Tb1}	7-<i>S</i>_{Tb2}	8-<i>S</i>_{Ho1}	8-<i>S</i>_{Ho2}	9-<i>S</i>_{Gd1}	9-<i>S</i>_{Gd2}
Capped octahedron (COC-7)	1.93	2.07	1.94	2.21	1.73	1.55	1.89	2.05
Capped trigonal prism (CTPR-7)	0.82	0.86	0.85	0.89	0.78	1.09	0.88	0.87
Pentagonal bipyramid (PBPY-7)	5.14	5.46	5.21	5.48	4.93	5.52	5.13	5.48

Table S6 Relaxation fitting parameters based on the extended Debye model^a from 3.0 to 5.0 K for complex **1**.

T / K	χ_s / cm^3	$\chi_T / \text{cm}^3 \text{ mol}^{-1}$	$\ln(\tau_1 / \text{s})$	α_1	$\ln(\tau_2 / \text{s})$	α_2	θ
3.0	1.512	6.663	-1.357	0.32	-9.116	0.45	0.54
4.0	2.252	5.335	-0.899	0.41	-8.513	0.36	0.37
5.0	2.393	4.393	-0.299	0.13	-8.555	0.33	0.22

$$^a \chi_{total}(\omega) = \chi_s + (\chi_T - \chi_s) \times \left[\frac{\beta}{1 + (i\omega\tau_1)^{1-\alpha_1}} + \frac{1 - \beta}{1 + (i\omega\tau_2)^{1-\alpha_2}} \right]$$

Where χ_s is the adiabatic susceptibility, χ_T is the isothermal susceptibility, ω ($=2\pi f$) is the angular frequency, τ_1 (low-frequency part) and τ_2 (high-frequency part) represent the magnetization relaxation times, θ is the weight of the relaxation process.

Table S7 Relaxation fitting parameters based on the extended Debye model from 3.0 to 5.0 K for complex **6**.

T / K	χ_s / cm^3	$\chi_T / \text{cm}^3 \text{ mol}^{-1}$	$\ln(\tau_1 / \text{s})$	α_1	$\ln(\tau_2 / \text{s})$	α_2	θ
3.0	2.656	8.790	0.857	0.37	-8.320	0.37	0.72
4.0	2.879	5.589	-0.563	0.38	-8.486	0.28	0.49
5.0	2.792	4.280	-1.485	0.22	-8.720	0.27	0.22

Table S8 Relaxation fitting parameters based on the generalized Debye model from 3.5 to 5.0 K for diluted-**1**.

T / K	χ_s / cm^3	$\chi_T / \text{cm}^3 \text{ mol}^{-1}$	$\ln(\tau / \text{s})$	α
3.5	0.199	0.573	-8.597	0.48
4.0	0.233	0.554	-8.513	0.44
4.5	0.257	0.531	-8.451	0.40
5.0	0.258	0.507	-8.615	0.40