## **Supporting Information**

## A New Family of Ln-radical Chains (Ln = Nd, Sm, Gd, Tb and Dy):

## Synthesis, Structure, and Magnetic Properties

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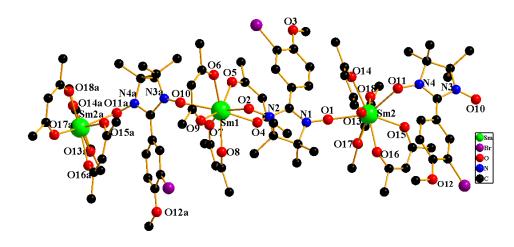


Figure S1. Crystal structure of complex 2. All hydrogen and fluorine atoms are omitted for clarity.

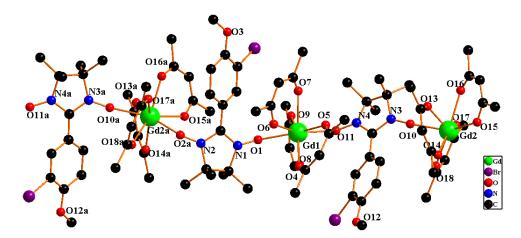


Figure S2. Crystal structure of complex 3. All hydrogen and fluorine atoms are omitted for clarity.

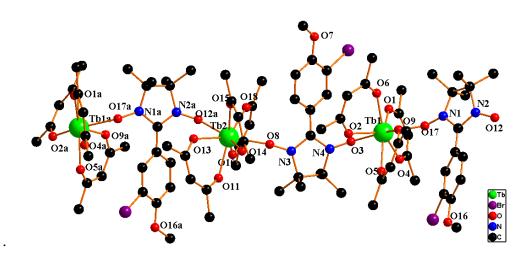


Figure S3. Crystal structure of complex 4. All hydrogen and fluorine atoms are omitted for clarity.

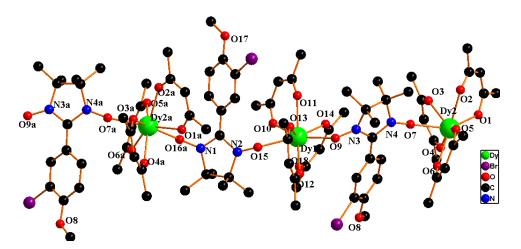


Figure S4. Crystal structure of complex 5. All hydrogen and fluorine atoms are omitted for clarity.

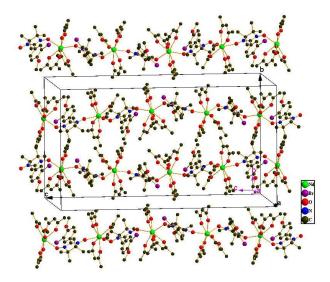


Figure S5. Packing diagram of complex 1. All hydrogen and fluorine atoms are not shown for the sake of clarity.

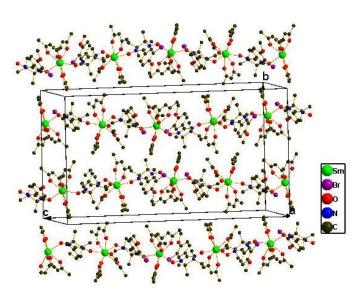


Figure S6. Packing diagram of complex 2. All hydrogen and fluorine atoms are not shown for the sake of clarity.

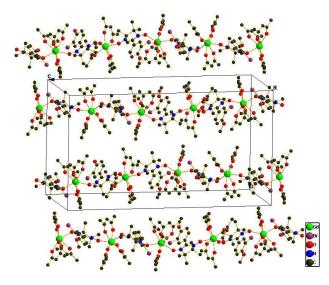


Figure S7. Packing diagram of complex 3. All hydrogen and fluorine atoms are not shown for the sake of clarity.

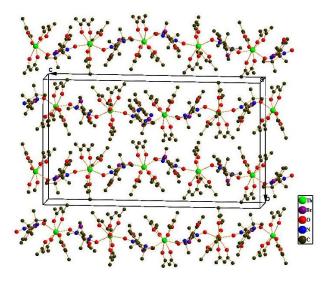


Figure S8. Packing diagram of complex 4. All hydrogen and fluorine atoms are not shown for the sake of clarity.

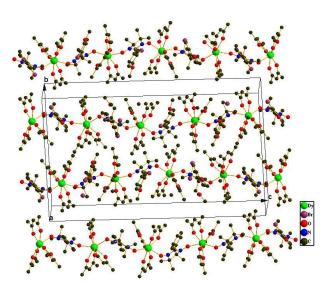


Figure S9. Packing diagram of complex 5. All hydrogen and fluorine atoms are not shown for the sake of clarity.

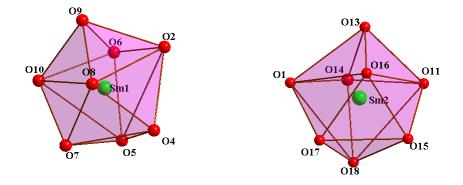


Figure S10. The coordination polyhedron of Sm(III) ion in complex 2.

**Table S1**  $\delta$  (°) and  $\varphi$  (°) values for complex **2.** 

	Sm1		Sm2		TPRS	DD	SAPR
$\delta_1$	O(7)-[O(8)-O(10)]-O(9) <sup>a</sup>	18.8	O(13)-[O(11)-O(16)]-O(15) <sup>a</sup>	18.0	0.0	29.5	0.0
$\delta_2$	O(4)-[O(2)-O(5)]-O(6) <sup>a</sup>	17.8	O(14)-[O(1)-O(18)]-O(17) <sup>a</sup>	29.3	21.8	29.5	0.0
$\delta_3$	O(7)-[O(5)-O(10)]-O(6) <sup>a</sup>	35.1	O(13)-[O(16)-O(1)]-O(17) <sup>a</sup>	30.9	48.2	29.5	52.4
$\delta_4$	O(4)-[O(2)-O(8)]-O(9) <sup>a</sup>	37.3	O(14)-[O(11)-O(18)]-O(15) <sup>a</sup>	36.9	48.2	29.5	52.4
$\varphi_1$	O(7)-O(4)-O(10)-O(2) <sup>b</sup>	8.4	O(13)-O(14)-O(16)-O(18) <sup>b</sup>	6.0	14.1	0	25.4
$\varphi_2$	O(9)-O(6)-O(5)-O(8) <sup>b</sup>	8.8	O(15)-O(17)-O(1)-O(11) <sup>b</sup>	4.4	14.1	0	25.4

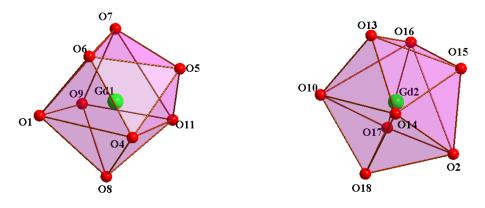


Figure S11. Th	ne coordination	polyhedron of	of Gd(III) ion	in complex <b>3</b> .
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**Table S2**  $\delta$  (°) and  $\varphi$  (°) values for complex **3.** 

	Gd1		Gd2		TPRS	DD	SAPR
$\delta_1$	O(6)-[O(1)-O(7)]-O(9) <sup>a</sup>	19.3	O(13)-[O(10)-O(14)]-O(18) <sup>a</sup>	27.8	0.0	29.5	0.0
$\delta_2$	O(5)-[O(4)-O(11)]-O(8) <sup>a</sup>	18.7	O(15)-[O(2)-O(16)]-O(17) <sup>a</sup>	17.4	21.8	29.5	0.0
$\delta_3$	O(6)-[O(4)-O(1)]-O(8) <sup>a</sup>	35.5	O(13)-[O(16)-O(10)]-O(17) <sup>a</sup>	31.7	48.2	29.5	52.4
$\delta_4$	O(5)-[O(7)-O(11)]-O(9) <sup>a</sup>	37.8	O(15)-[O(2)-O(14)]-O(18) <sup>a</sup>	36.5	48.2	29.5	52.4
$\varphi_1$	O(6)-O(5)-O(1)-O(11) <sup>b</sup>	8.0	O(13)-O(15)-O(10)-O(2) <sup>b</sup>	4.6	14.1	0	25.4
$\varphi_2$	O(9)-O(8)-O(7)-O(4) <sup>b</sup>	9.2	O(18)-O(17)-O(14)-O(16) <sup>b</sup>	6.9	14.1	0	25.4

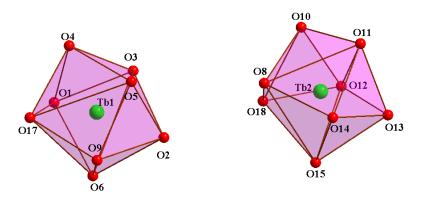


Figure S12. The coordination polyhedron of Tb(III) ion in complex 4.

**Table S3**  $\delta$  (°) and  $\varphi$  (°) values for complex **4.** 

	Tb1		Tb2		TPRS	DD	SAPR
$\delta_1$	O(4)-[O(17)-O(5)]-O(9) <sup>a</sup>	18.2	O(10)-[O(11)-O(12)]-O(13) <sup>a</sup>	18.5	0.0	29.5	0.0
$\delta_2$	O(1)-[O(3)-O(6)]-O(2) <sup>a</sup>	19.1	O(18)-[O(8)-O(15)]-O(14) <sup>a</sup>	28.1	21.8	29.5	0.0
$\delta_3$	O(4)-[O(5)-O(3)]-O(2) <sup>a</sup>	36.7	O(10)-[O(11)-O(8)]-O(14) <sup>a</sup>	31.5	48.2	29.5	52.4
$\delta_4$	O(1)-[O(6)-O(17)]-O(9) <sup>a</sup>	35.0	O(18)-[O(15)-O(12)]-O(13) <sup>a</sup>	36.4	48.2	29.5	52.4
$\varphi_1$	O(4)-O(1)-O(5)-O(6) <sup>b</sup>	9.4	O(10)-O(18)-O(11)-O(15) <sup>b</sup>	6.3	14.1	0	25.4
$\varphi_2$	O(9)-O(2)-O(17)-O(3) <sup>b</sup>	7.1	O(13)-O(14)-O(12)-O(8) <sup>b</sup>	4.1	14.1	0	25.4

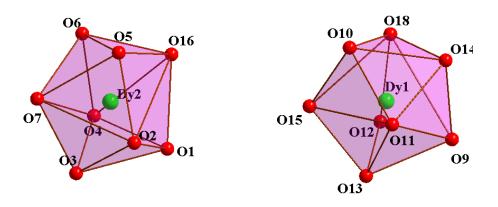
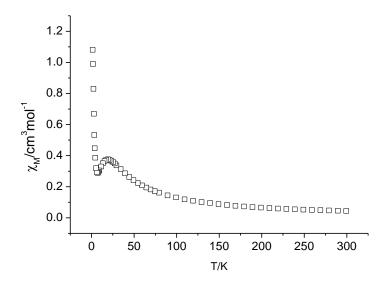


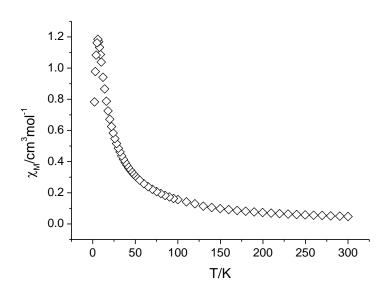
Figure S13. The coordination polyhedron of Dy(III) ion in complex 5.

Table S4	$\delta$ (°) and $\varphi$ (°) values for complex <b>5</b> .
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	Dy1		Dy2		TPRS	DD	SAPR
$\delta_1$	O(13)-[O(15)-O(11)]-O(10) <sup>a</sup>	17.4	O(3)-[O(7)-O(2)]-O(5) <sup>a</sup>	32.3	0.0	29.5	0.0
$\delta_2$	O(12)-[O(18)-O(9)]-O(14) <sup>a</sup>	18.6	O(1)-[O(4)-O(16)]-O(6) <sup>a</sup>	36.2	21.8	29.5	0.0
$\delta_3$	O(13)-[O(11)-O(9)]-O(14) <sup>a</sup>	36.9	O(3)-[O(7)-O(4)]-O(6) <sup>a</sup>	27.6	48.2	29.5	52.4
$\delta_4$	O(12)-[O(15)-O(18)]-O(10) <sup>a</sup>	36.1	O(1)-[O(2)-O(16)]-O(5) <sup>a</sup>	18.7	48.2	29.5	52.4
$\varphi_1$	O(13)-O(12)-O(11)-O(18) <sup>b</sup>	8.0	O(3)-O(1)-O(7)-O(16) <sup>b</sup>	4.6	14.1	0	25.4
$\varphi_2$	O(10)-O(14)-O(15)-O(9) <sup>b</sup>	9.2	O(5)-O(6)-O(2)-O(4) <sup>b</sup>	6.9	14.1	0	25.4



**Figure S14.**  $\chi_M$  versus *T* plot for **4**.



**Figure S15.**  $\chi_M$  versus *T* plot for **5.** 

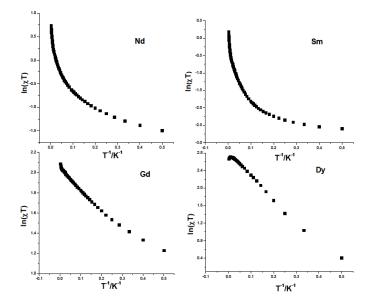


Figure S16.  $ln(\chi T)$  vs. 1/T plot for 1, 2, 3 and 5.

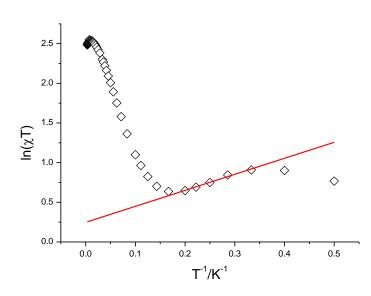


Figure S17.  $\ln(\chi T)$  vs. 1/T plot for 4 and the solid line representing the linear fit.

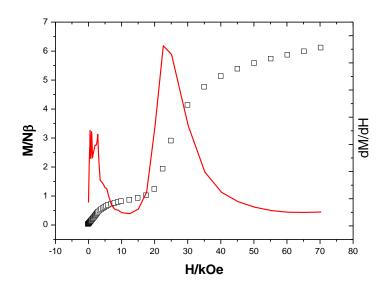


Figure S18. Plots of M vs H and dM/dH vs H for 4 at 2.0K.

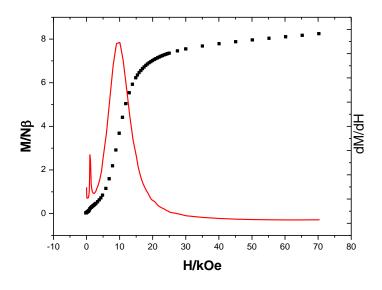


Figure S19. Plots of M vs H and dM/dH vs H for 5 at 2.0K.

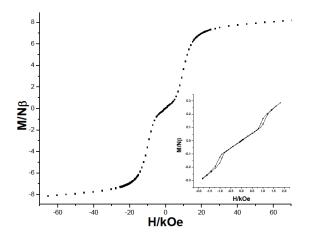


Figure S20. The hysteresis loop at 2.0 K for 5.

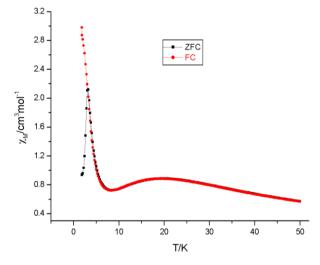


Figure S21 The field-cooled(FC) and Zero-field-cooled(ZFC) susceptibility at 50 Oe for 4

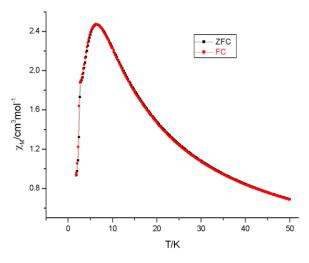


Figure S22 The field-cooled(FC) and Zero-field-cooled(ZFC) susceptibility at 50 Oe for 5

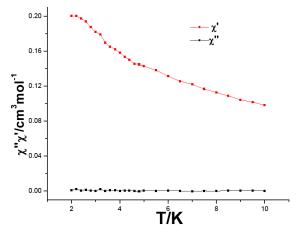


Figure S23. Temperature-dependent ac magnetic susceptibility of 1 in zero dc field with an oscillating of 3.5 Oe.

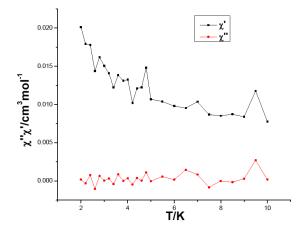


Figure S24. Temperature-dependent ac magnetic susceptibility of 2 in zero dc field with an oscillating of 3.5 Oe.

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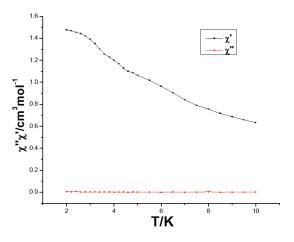


Figure S25. Temperature-dependent ac magnetic susceptibility of 3 in zero dc field with an oscillating of 3.5 Oe.