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Slow magnetic relaxation in mononuclear complexes of Tb, Dy, Ho and Er with the pentadentate (N_3O_2) Schiff-base dapsc ligand.

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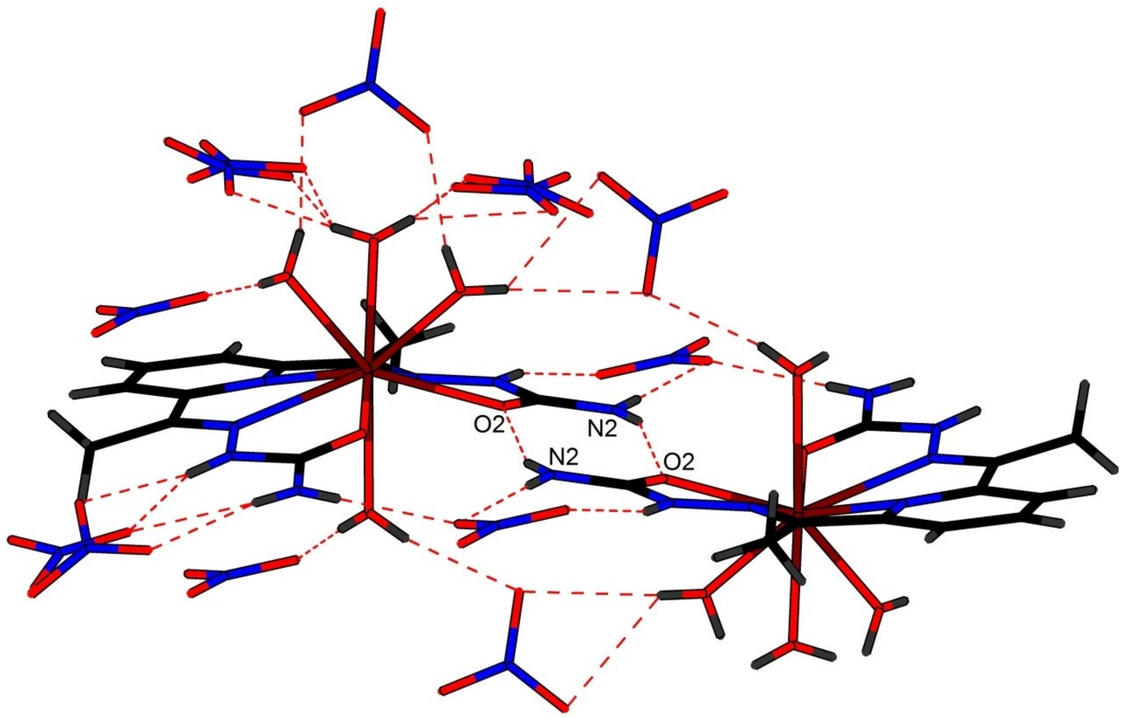


Figure S1. Hydrogen bonding in **1-Tb**.

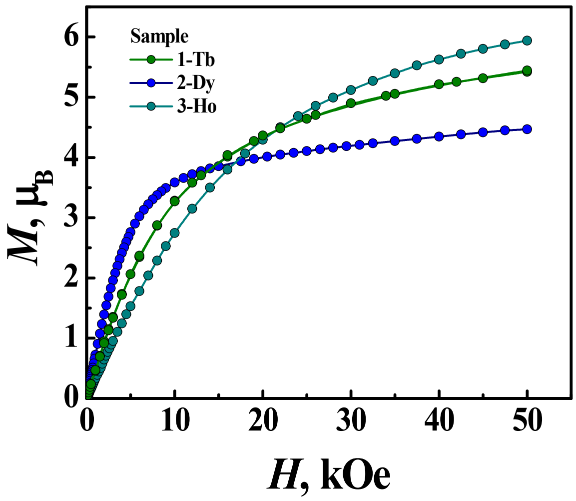


Figure S2. *M* vs *H* for **1-Tb**, **2-Dy** and **3-Ho** complexes.

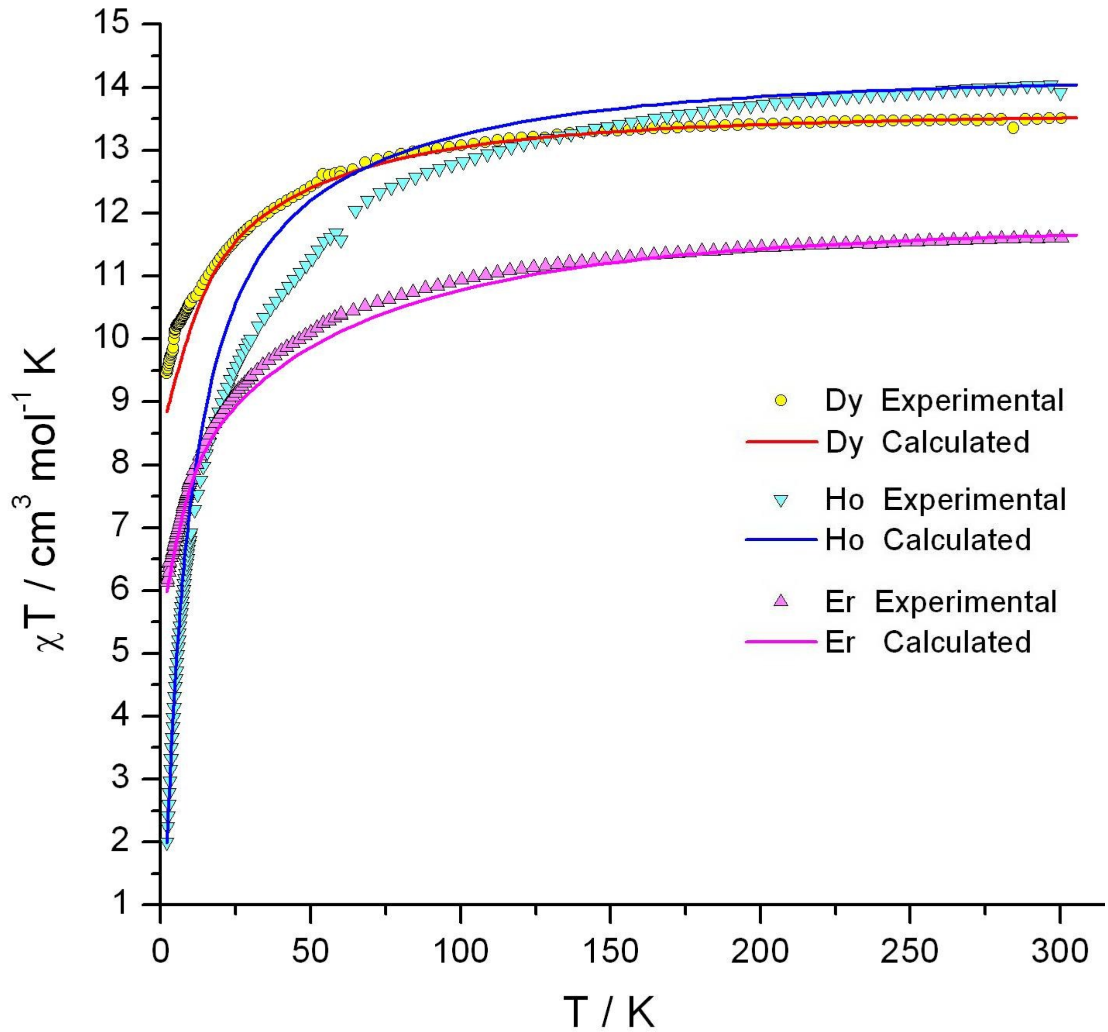
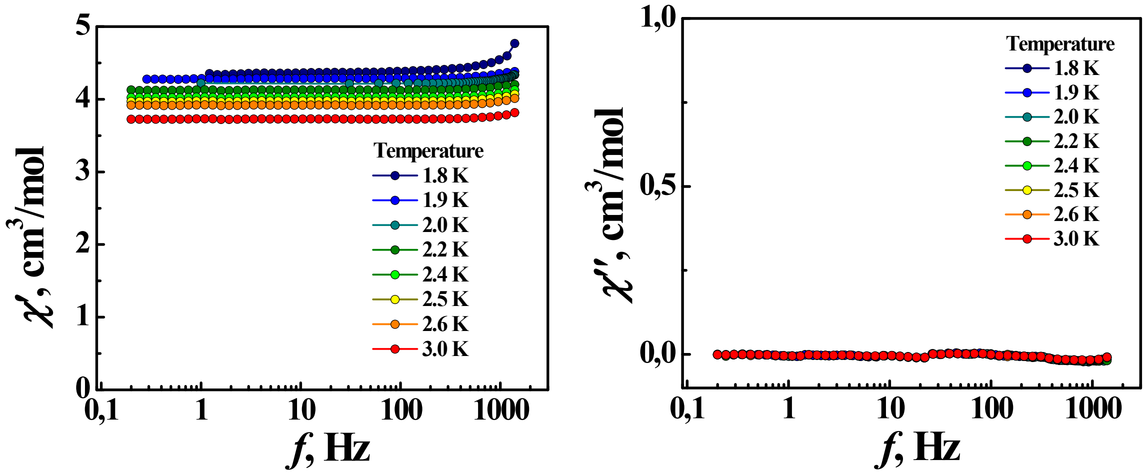
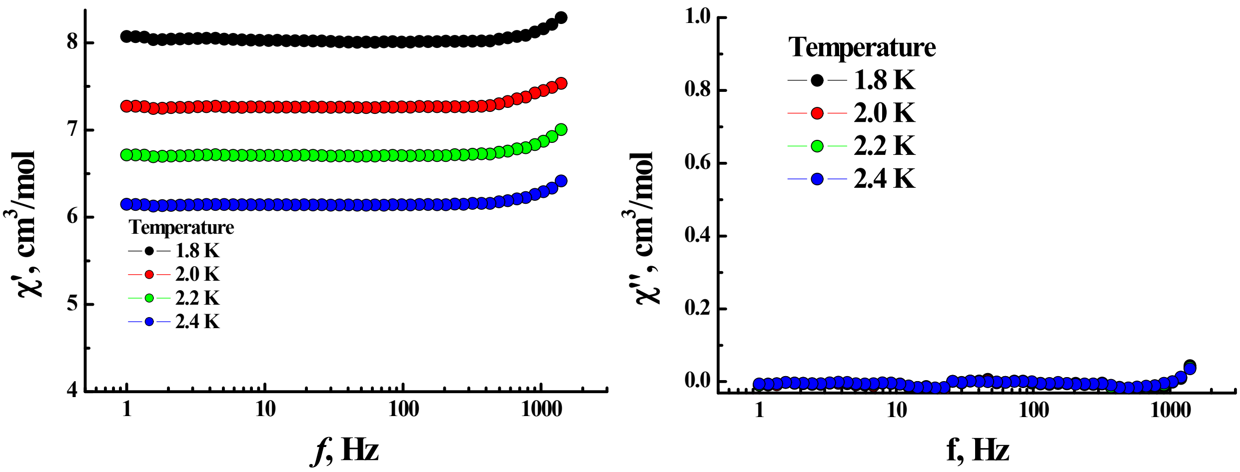


Figure S3. Experimental and calculated temperature variation of the $\chi_{mol}T$ product for the $[Ln(H_2dapsc)(H_2O)_4](NO_3)_3$ complexes **2-4** obtained with the initial set of intrinsic CF parameters b_2 = 1000, b_4 = 420, b_6 = 270 cm^{-1} (at R_0 = 2.45 Å), for O and N ligand atoms.



(a) (b)

Figure S4. The frequency dependencies of in-phase χ' (a) and out-of-phase χ″ (b) ac magnetic susceptibility for **1-Tb** at temperatures of 1.8-3.0 K in zero dc field.



(a) (b)

Figure S5. The frequency dependencies of in-phase χ' (a) and out-of-phase χ″ (b) ac magnetic susceptibility for **2-Dy** at temperatures of 1.8-2.4 K in zero dc field.

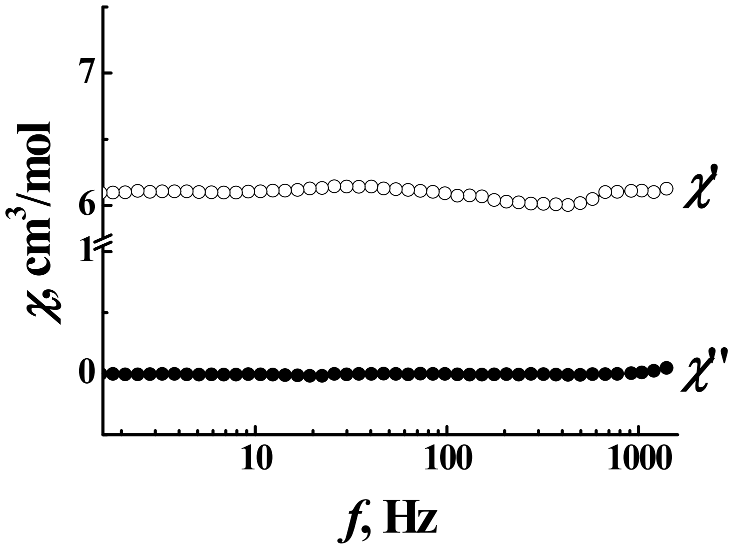


Figure S6. The frequency dependencies of in-phase χ' and out-of-phase χ″ ac magnetic susceptibility for **4-Er** at temperatures of 1.8 K in zero dc field.

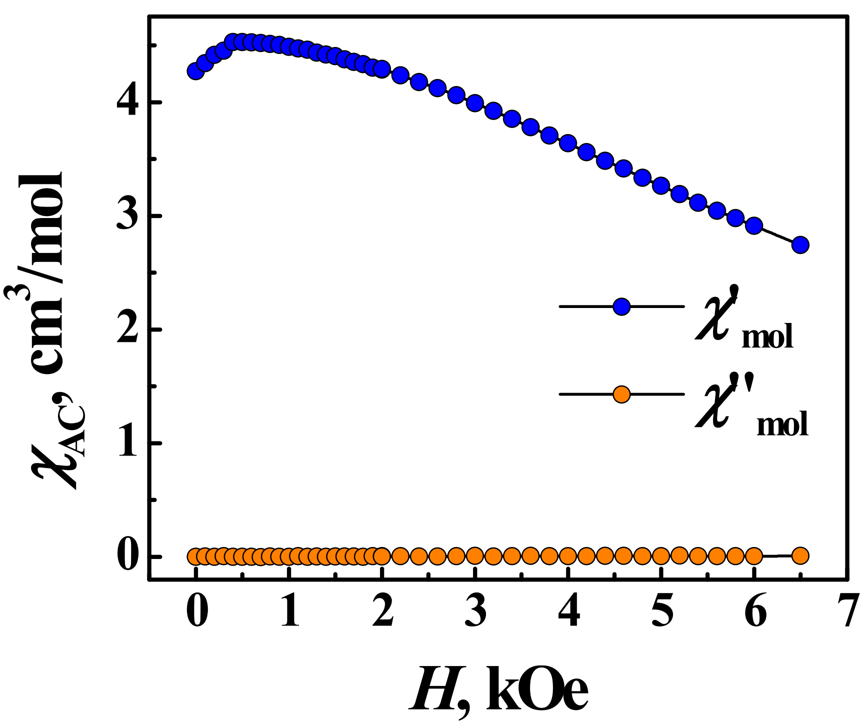


Figure S7. *Dc* field dependencies of ac magnetic susceptibility for **1-Tb** at T = 1.8 K and frequency (f) of 100 Hz (ac field amplitude of 4 Oe).

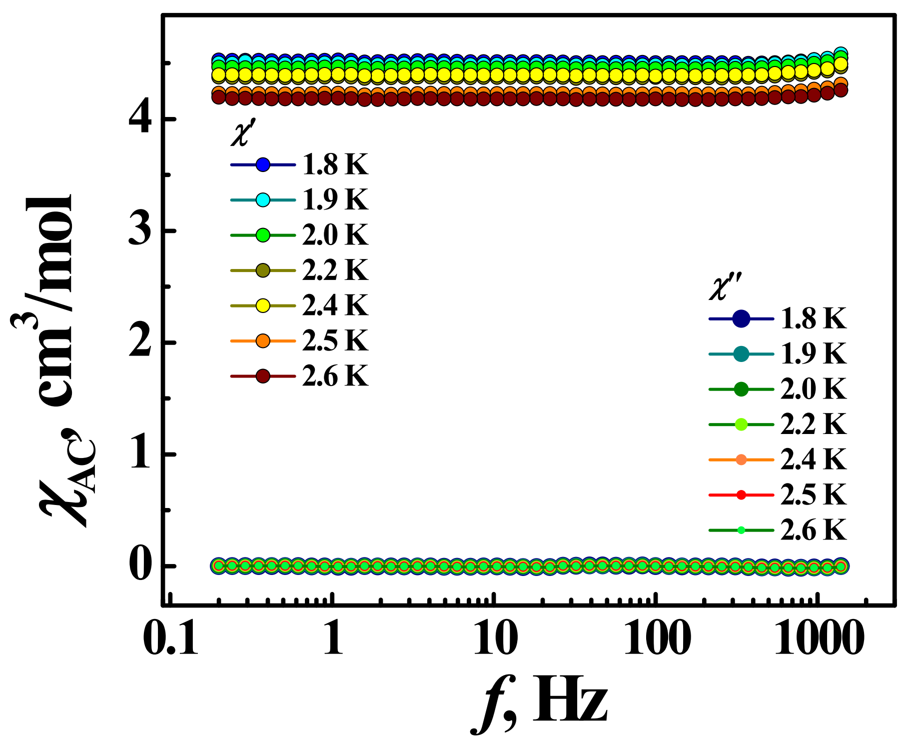


Figure S8. The frequency dependencies of in-phase χ' and out-of-phase χ″ ac magnetic susceptibility for **1-Tb** at temperatures of 1.8-2.6 K in dc field of 1000 Oe (ac field amplitude is 4 Oe).

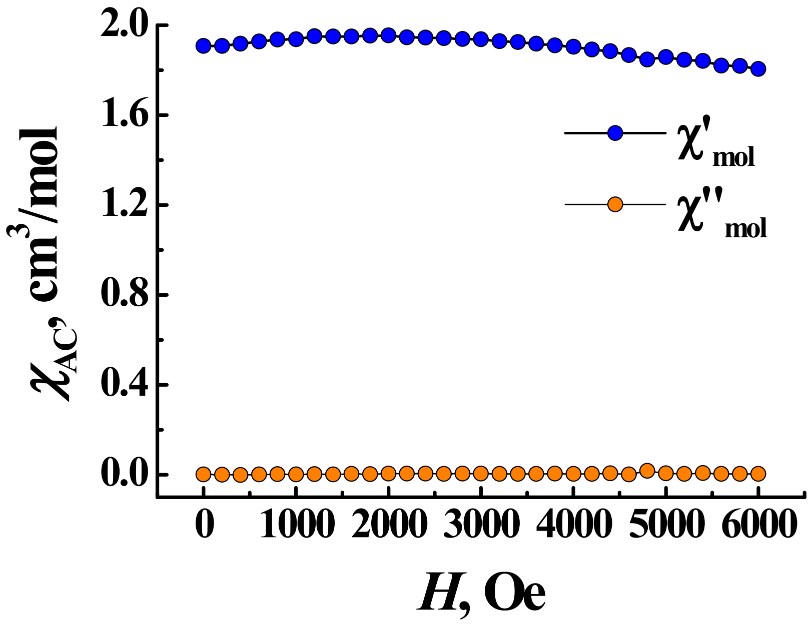


Figure S9. Dc field dependencies of ac magnetic susceptibility for **3-Ho** at T = 1.8 K and frequency (*f*) of 100 Hz (ac field amplitude of 4 Oe).

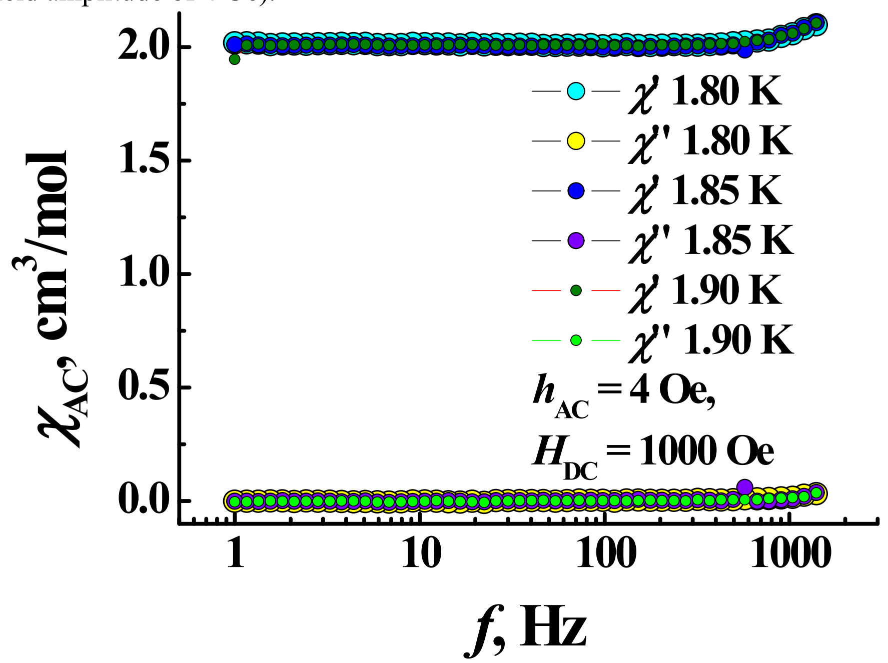


Figure S10. The frequency dependencies of in-phase χ' and out-of-phase χ″ ac magnetic susceptibility for **3-Ho** at temperatures of 1.8-1.9 K in dc field of 1000 Oe (ac field amplitude is 4 Oe).

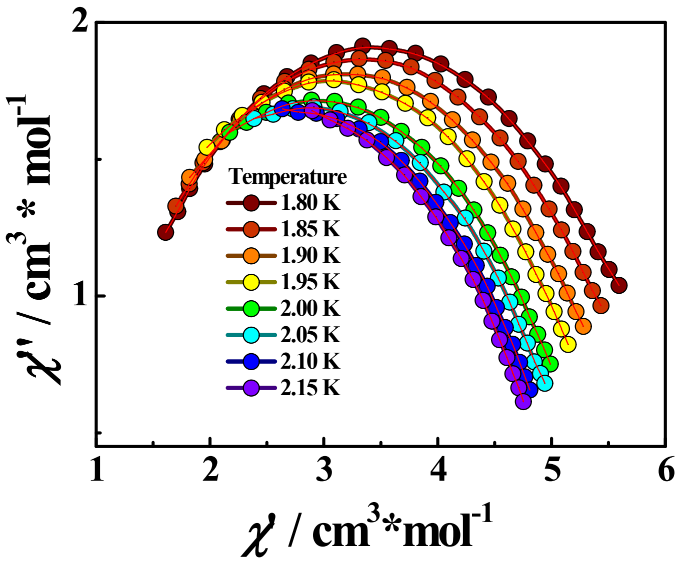


Figure S11. High frequency part of Cole-Cole plots under 1000 Oe for **2-Dy**. Solid lines are the fits by the generalized Debye model with the best-fit values of the parameters listed in Table S4.

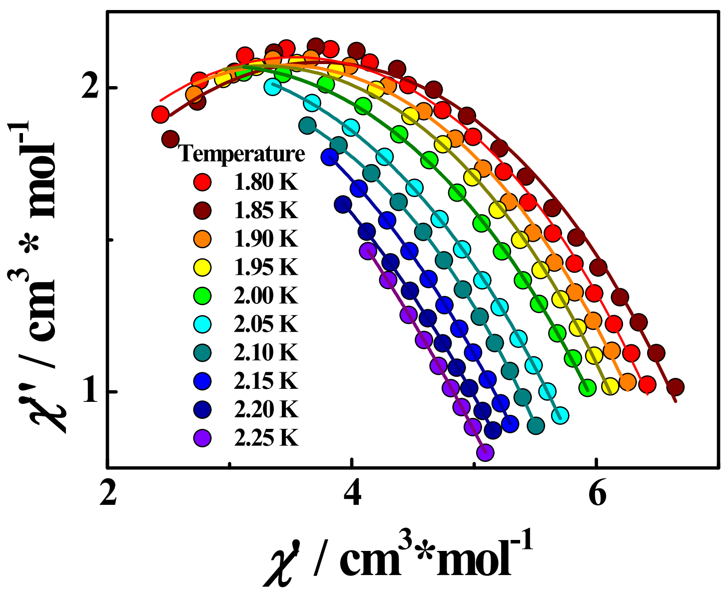


Figure S12. High frequency part of Cole-Cole plots under 1000 Oe for **4-Er**. Solid lines are the fits by the generalized Debye model with the best-fit values of the parameters listed in Table S4.

Table S1. Calculated crystal-field parameters B_{kq} (in cm^{-1}) for **1-Tb**, **2-Dy**, **3-Ho** and **4-Er** complexes. Real (Re) and imaginary (Im) parts of complex B_{kq} parameters are indicated[(a)]

		1-Tb		**2-Dy**		**3-Ho**		**4-Er**	
k	q	ReB_{kq}	ImB_{kq}	ReB_{kq}	ImB_{kq}	ReB_{kq}	ImB_{kq}	ReB_{kq}	ImB_{kq}
2	0	360	0	77	0	175	0	120	0
2	1	1	-26	67	-33	-29	76	-75	56
2	2	-100	107	232	-15	122	-15	216	-36
4	0	302	0	320	0	352	0	354	0
4	1	121	12	-58	104	-41	178	-58	108
4	2	-16	78	61	-51	71	-35	69	-56
4	3	-7	601	299	544	275	447	328	573
4	4	-9	356	-372	65	-307	59	-405	67
6	0	172	0	184	0	244	0	203	0
6	1	149	17	-66	146	-94	99	-78	163
6	2	72	180	58	-196	49	-279	64	-220
6	3	131	156	203	76	266	158	228	99
6	4	4	-90	93	-6	92	14	99	-9
6	5	69	297	-24	-344	-17	-581	-20	-401
6	6	-248	-241	-339	92	-580	124	-342	105
S [(b)], cm^{-1}		468		466		512		501	

[(a)] The B_{kq} CF parameters are obtained from the superposition-model CF calculations with the follows sets of intrinsic b_k parameters, (1) for **2**-Dy and **4**-Er: $b_2 = 1000$, $b_4 = 420$, $b_6 = 270$ cm^{-1} (at $R_0 = 2.45$ Å), for O and N ligand atoms, (2) for **1**-Tb: $b_2 = 700$, $b_4 = 420$, $b_6 = 270$ cm^{-1} (at $R_0 = 2.45$ Å) for O, and $b_2 = 400$, $b_4 = 420$, $b_6 = 270$ cm^{-1} (at $R_0 = 2.45$ Å) for N; some fine extra correction was applied for the k=2 B_{kq} parameters, (3) for **3**-Ho: $b_2 = 600$, $b_4 = 380$, $b_6 = 330$ cm^{-1} (at $R_0 = 2.45$ Å) for O, and $b_2 = 400$, $b_4 = 390$, $b_6 = 340$ cm^{-1} (at $R_0 = 2.55$ Å) for N. The t^k parameters are all fixes at $t^2 = 5$, $t^4 = 8$ and $t^6 = 11$.
[(b)] S criterion estimates the overall strength of the crystal-field potential of Ln^{3+} ions in terms of B_{kq} parameters, $S = \left[\frac{1}{3} \sum_{k=2,4,6} \left(\frac{1}{2k+1} \right) \left(B_{k0}^2 + 2 \sum_{k=2,4,6} (\mathrm{Re}\, B_{kq}^2 + \mathrm{Im}\, B_{kq}^2) \right) \right]^{1/2}$, ref. [1S]

Table S2. Real (Re) and imaginary (Im) parts of complex expansion coefficients $C(M_J)$ of the $|^6H_{15/2}, M_J\rangle$ composition of wave functions of the ground Kramers doublet $\varphi^{\pm}$ of Dy^{3+} ion in **2-Dy** complex

	φ^+		φ^-	
M_J	Re	Im	Re	Im
–15/2	-0.110	-0.108	0.015	0.015
–13/2	0.167	0.019	0.202	0.004
–11/2	-0.209	-0.054	-0.095	0.252
–9/2	0.345	-0.147	0.055	-0.021
–7/2	-0.071	0.095	-0.063	0.348
–5/2	0.133	-0.203	-0.196	0.124
–3/2	0.309	0.186	0.013	0.147
–1/2	-0.090	-0.127	-0.457	0.076
1/2	0.273	0.374	-0.153	0.028
3/2	0.112	-0.096	-0.351	-0.083
5/2	0.054	0.225	-0.047	0.238
7/2	0.198	-0.293	-0.015	0.117
9/2	-0.024	-0.054	0.144	0.346
11/2	0.108	-0.246	0.188	0.107
13/2	-0.148	-0.138	0.133	0.103
15/2	0.022	0	0.154	0

Table S3. Real (Re) and imaginary (Im) parts of complex expansion coefficients $C(M_J)$ of the $|^4H_{15/2}, M_J>$ composition of wave functions of the ground Kramers doublet $\varphi^{\pm}$ of Er^{3+} ion in **4-Er** complex

	φ^+		φ^-	
M_J	Re	Im	Re	Im
–15/2	0.054	-0.156	-0.005	0.016
–13/2	-0.120	-0.396	0.075	0.199
–11/2	0.001	0.219	0.192	0.149
–9/2	0.091	0.239	-0.075	-0.159
–7/2	-0.155	0.204	-0.058	-0.201
–5/2	0.152	-0.071	-0.025	-0.157
–3/2	0.369	-0.053	-0.197	0.113
–1/2	-0.253	0.056	-0.311	0.185
1/2	-0.277	0.233	0.136	-0.221
3/2	0.172	-0.149	0.172	-0.331
5/2	0.140	0.076	-0.117	0.120
7/2	-0.171	-0.121	-0.243	0.079
9/2	0.125	0.123	0.195	0.165
11/2	0.078	0.230	-0.206	-0.073
13/2	-0.163	-0.137	-0.335	-0.243
15/2	0.017	0	0.166	0

Table S4. Parameters for **2-Dy** and **4-Er** resulting from the analysis of *ac* magnetic susceptibility by generalized Debye model where χ_S is the adiabatic susceptibility, χ_T is the isothermal susceptibility and α is the dispersion coefficient showing the distribution of the relaxation time.

2-Dy				**4-Er**			
T, K	χ_S, cm^3/mol	χ_T, cm^3/mol	α	T, K	χ_S, cm^3/mol	χ_T, cm^3/mol	α
1.8	0.75	6.25	0.22	1.8	0	7.45	0.23
1.85	0.725	5.925	0.21	1.85	0	7.23	0.24
1.9	0.675	5.75	0.21	1.9	0	7.08	0.25
1.95	0.55	5.575	0.213	1.95	0	6.95	0.24
2	0.4475	5.4075	0.23	2	0	6.85	0.26
2.05	0.205	5.355	0.24	2.05	0	6.53	0.25
2.1	0.1025	5.2025	0.25	2.1	0	6.25	0.26
2.15	0.05	5.05	0.28	2.15	0	6.15	0.24
				2.2	0	6.09	0.25
				2.25	0	6	0.26

References

(1S) N.C. Chang, J.B. Gruber, R.P. Leavitt, C.A. Morrison, *J. Chem. Phys.,* 1982, **78**, 3877 − 3889.