Supplementary Material (ESI) for Dalton Transactions
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The solution structure of homotrimetallic lanthanide helicates investigated with novel model-free multi-centre paramagnetic NMR methods.

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Supporting information

Table S1. Selected Structural Parameters $C_{ikl} = R_{ik} \cdot \left(\frac{G_i^1 - G_l^1 R_{il}}{G_k^1 R_{ik} - G_l^1 R_{il}} \right)$ and $D_{ikl} = -R_{il} \cdot \left(\frac{G_i^1 - G_k^1 R_{ik}}{G_k^1 R_{ik} - G_l^1 R_{il}} \right)$ and Intercepts $B_{ikl} = F_i - F_k C_{ikl} - F_l D_{ikl}$

Obtained from Least-square Planes for $\left(\frac{d_{ij}^{\text{para}}}{\langle S_z \rangle_j}, \frac{d_{kj}^{\text{para}}}{\langle S_z \rangle_j}, \frac{d_{lj}^{\text{para}}}{\langle S_z \rangle_j} \right)$ According to Eqn (12) for the Aromatic Protons H1-H11 in $[Ln_3(L1)_3]^{9+}$

(Ln = Ce-Yb, CD_3CN, 298 K).[a]

H*i*,*k*,*l*	$B_{ikl}^{\text{solution}}$	$C_{ikl}^{\text{solution}}$	$D_{ikl}^{\text{solution}}$	AF_i [b]	AF_k [b]	AF_l [b]	AF_{ikl} [b]	C_{ikl}^{solid} [c]	D_{ikl}^{solid} [c]	$C_{ikl}^{\text{gas-phase}}$ [d]	$D_{ikl}^{\text{gas-phase}}$ [d]
H1,2,3	-1.20	-5.54	6.69	0.054	0.095	0.005	0.036	-287.42	384.28	-19.47	25.24
H1,2,4	0.22	0.85	1.12	0.088	0.071	0.051	0.041	0.00	1.54	-0.16	0.76
H1,2,5	0.72	2.03	-0.19	0.194	0.002	0.047	0.066	-1.26	-0.38	-0.86	-0.14
H1,2,6	1.27	3.22	-0.17	0.218	0.001	0.033	0.073	-0.52	-0.30	-0.51	-0.14
H1,2,7	0.52	1.18	1.45	0.066	0.058	0.027	0.031	0.04	1.62	-0.21	0.39
H1,2,8	-0.99	1.05	-4.45	0.023	0.147	0.011	0.050	-2.47	-13.49	-1.32	-8.05
H1,2,9	1.27	3.12	-2.16	0.153	0.114	0.024	0.064	-0.43	-4.24	1.69	-4.08
H1,2,10	1.61	5.27	-6.32	0.101	0.345	0.009	0.120	0.63	-4.53	0.20	-2.83
H1,2,11	0.37	1.01	-3.75	0.021	0.153	0.010	0.052	-0.55	-3.53	-0.26	-2.39
H1,3,4	0.03	0.89	0.97	0.157	0.071	0.059	0.061	0.00	1.54	-0.21	0.77
H1,3,5	0.21	1.85	-0.13	0.245	0.001	0.044	0.083	-1.70	-0.38	-1.16	-0.15
H1,3,6	0.37	2.49	-0.11	0.266	0.001	0.036	0.089	-0.69	-0.30	-0.69	-0.15
H1,3,7	0.22	1.18	1.19	0.117	0.058	0.033	0.045	0.06	1.62	-0.28	0.40
H1,3,8	-1.00	1.14	-3.67	0.040	0.135	0.012	0.047	-3.33	-13.61	-1.84	-8.64
H1,3,9	0.39	2.49	-1.33	0.219	0.086	0.030	0.079	-0.57	-4.24	2.02	-3.75

H1,3,10	0.26	3.30	-3.14	0.149	0.275	0.015	0.104	0.84	-4.52	0.26	-2.80
H1,3,11	0.12	0.98	-3.22	0.037	0.161	0.013	0.055	-0.74	-3.53	-0.34	-2.42
H1,4,5	-0.14	2.12	0.17	0.091	0.001	0.034	0.032	1.53	0.00	0.94	0.03
H1,4,6	-0.16	1.59	0.07	0.091	0.001	0.046	0.034	1.53	0.00	1.11	0.07
H1,4,7	-0.89	6.36	-6.77	0.055	0.069	0.007	0.029	1.58	-0.05	3.23	-1.28
H1,4,8	473.17	407.99	1621.69	0.015	0.155	0.000	0.052	1.54	-0.01	0.87	1.12
H1,4,9	-0.18	1.63	1.00	0.063	0.069	0.031	0.033	1.53	-0.01	0.69	-0.36
H1,4,10	-0.04	1.36	1.31	0.061	0.278	0.036	0.096	1.54	0.01	0.42	-1.26
H1,4,11	-1.58	20.87	65.91	0.014	0.142	0.001	0.048	1.53	-0.01	2.08	4.14
H1,5,6	-0.25	-0.53	0.31	0.010	0.005	0.092	0.031	0.26	-0.50	0.22	-0.36
H1,5,7	0.31	0.36	4.14	0.001	0.086	0.014	0.029	-0.01	1.57	0.05	0.52
H1,5,8	-4.27	0.42	-13.97	0.000	0.136	0.003	0.045	-0.78	14.11	-0.41	14.80
H1,5,9	-0.28	-0.49	3.38	0.002	0.154	0.021	0.052	0.19	-6.39	-0.10	-1.37
H1,5,10	0.12	-0.29	3.30	0.001	0.373	0.019	0.125	-0.13	-3.02	-0.03	-2.29
H1,5,11	0.08	0.47	-12.53	0.000	0.100	0.002	0.033	0.30	-6.27	0.06	-3.40
H1,6,7	0.08	0.13	2.47	0.001	0.090	0.025	0.031	-0.02	1.50	0.10	0.67
H1,6,8	-2.45	0.13	-7.66	0.000	0.144	0.006	0.048	-0.38	3.57	-0.23	5.13
H1,6,9	-0.97	-5.07	60.62	0.002	0.212	0.002	0.071	1.40	-24.19	-0.11	-0.95
H1,6,10	0.57	-0.38	7.29	0.001	0.425	0.010	0.142	-0.16	-2.04	-0.04	-2.03
H1,6,11	-0.07	0.15	-6.75	0.000	0.112	0.004	0.037	-4.58	50.72	0.14	-4.73
H1,7,8	13.12	14.53	39.39	0.018	0.104	0.001	0.035	1.60	-0.23	0.47	1.54
H1,7,9	0.06	2.48	1.59	0.032	0.031	0.009	0.015	1.48	-0.39	0.35	-0.45

H1,7,10	0.21	1.87	1.85	0.030	0.117	0.011	0.040	1.74	0.33	0.19	-1.45
H1,7,11	4.01	66.55	166.02	0.011	0.072	0.000	0.024	1.51	-0.25	2.31	11.58
H1,8,9	-2.58	-7.96	1.82	0.111	0.018	0.004	0.038	2.81	-5.12	-4.52	-1.79
H1,8,10	-1.70	-5.82	2.11	0.104	0.070	0.006	0.042	-2.75	-3.61	-1.06	-2.46
H1,8,11	6.16	19.82	-20.69	0.069	0.091	0.001	0.038	3.89	-4.54	1.93	-2.96
H1,9,10	0.68	-4.89	7.60	0.038	0.156	0.004	0.053	-2.53	-1.82	0.55	-3.21
H1,9,11	-0.09	1.59	-6.36	0.013	0.091	0.004	0.031	-18.52	11.90	-0.53	-2.07
H1,10,11	0.10	1.82	-4.78	0.054	0.095	0.005	0.036	-2.11	-1.88	-1.58	-1.05
H2,3,4	-0.21	1.11	-0.20	0.071	0.005	0.044	0.028	1.34	-0.01	1.31	-0.04
H2,3,5	-0.24	0.96	0.03	0.090	0.000	0.065	0.037	1.34	0.00	1.36	0.01
H2,3,6	-0.27	0.86	0.02	0.094	0.000	0.075	0.040	1.34	0.00	1.33	0.01
H2,3,7	-0.25	1.01	-0.22	0.081	0.009	0.055	0.033	1.34	-0.01	1.31	-0.02
H2,3,8	-0.04	1.00	0.65	0.088	0.060	0.060	0.041	1.35	0.05	1.39	0.44
H2,3,9	-0.27	0.83	0.26	0.088	0.020	0.073	0.039	1.34	0.01	1.19	0.19
H2,3,10	-0.25	0.67	0.51	0.086	0.129	0.088	0.060	1.33	0.02	1.28	0.14
H2,3,11	-0.24	1.03	0.59	0.076	0.057	0.051	0.036	1.34	0.01	1.31	0.12
H2,4,5	-0.42	1.16	0.20	0.093	0.003	0.133	0.054	-1.22	-0.30	-1.09	-0.21
H2,4,6	-0.44	0.49	0.08	0.063	0.001	0.212	0.074	-2.97	-0.58	-2.15	-0.41
H2,4,7	-0.75	2.45	-3.14	0.039	0.059	0.026	0.025	36.86	-38.96	-15.27	7.93
H2,4,8	3.16	3.10	12.16	0.015	0.149	0.008	0.050	-0.62	-5.46	-0.66	-6.94
H2,4,9	-0.47	0.55	1.06	0.036	0.123	0.108	0.056	-3.59	-9.91	0.41	2.20
H2,4,10	-0.31	0.26	1.43	0.014	0.355	0.087	0.122	2.44	7.19	2.09	7.79

H2,4,11	-0.63	3.37	11.09	0.013	0.132	0.006	0.044	-2.77	-6.37	-8.14	-25.55
H2,5,6	-0.47	-0.15	0.14	0.004	0.003	0.244	0.081	-0.51	0.40	-0.42	0.42
H2,5,7	0.02	0.59	4.40	0.002	0.124	0.040	0.044	-0.29	-1.24	-0.22	-0.61
H2,5,8	-4.17	0.55	-12.59	0.000	0.146	0.008	0.049	0.31	-11.18	0.31	-17.29
H2,5,9	-0.50	-0.17	1.86	0.001	0.148	0.074	0.055	-0.45	5.06	-0.06	1.60
H2,5,10	-0.28	-0.06	1.81	0.000	0.350	0.068	0.119	-0.20	2.39	-0.14	2.68
H2,5,11	-0.26	0.55	-10.55	0.000	0.123	0.006	0.041	-0.53	4.97	-0.24	3.97
H2,6,7	-0.36	0.10	0.85	0.002	0.115	0.191	0.074	-0.53	-2.90	-0.47	-1.30
H2,6,8	-1.64	0.15	-3.93	0.000	0.190	0.032	0.064	0.15	-6.91	0.18	-9.97
H2,6,9	-0.49	-0.44	5.49	0.001	0.201	0.034	0.068	-3.29	46.84	-0.06	1.85
H2,6,10	-0.21	-0.07	2.38	0.000	0.361	0.053	0.122	-0.26	3.95	-0.20	3.95
H2,6,11	-0.42	0.16	-3.36	0.000	0.203	0.031	0.068	8.29	-98.19	-0.55	9.20
H2,7,8	-19.13	-18.70	-56.57	0.016	0.106	0.001	0.036	-0.65	-5.37	-0.36	-7.25
H2,7,9	-0.39	0.99	1.48	0.054	0.122	0.077	0.052	-3.46	-9.03	0.21	2.14
H2,7,10	-0.25	0.39	1.66	0.020	0.341	0.072	0.116	2.76	7.70	0.96	6.85
H2,7,11	-1.32	-15.33	-39.21	0.010	0.071	0.001	0.024	-2.73	-5.93	-9.06	-54.72
H2,8,9	-1.45	-3.23	1.56	0.132	0.045	0.027	0.047	-6.60	2.07	-2.67	1.35
H2,8,10	-0.67	-1.31	1.69	0.100	0.240	0.050	0.088	-4.35	1.46	-5.28	1.86
H2,8,11	52.78	170.39	-145.34	0.079	0.084	0.000	0.038	-7.04	1.84	-7.55	2.24
H2,9,10	-0.12	-1.17	3.05	0.035	0.239	0.028	0.081	-4.01	4.28	2.73	-1.90
H2,9,11	-0.44	1.54	-2.69	0.051	0.159	0.031	0.056	33.52	-27.92	2.09	-1.22
H2,10,11	-0.27	1.66	-1.07	0.266	0.116	0.057	0.099	3.83	-2.98	6.20	-5.22

H3,4,5	-0.19	1.17	0.17	0.064	0.002	0.132	0.049	-0.91	-0.22	-0.81	-0.16
H3,4,6	-0.21	0.66	0.07	0.056	0.001	0.207	0.072	-2.22	-0.43	-1.62	-0.31
H3,4,7	-0.43	2.03	-2.37	0.034	0.047	0.040	0.024	27.57	-29.14	-11.65	6.07
H3,4,8	1.74	1.85	6.50	0.015	0.136	0.020	0.046	-0.46	-4.08	-0.47	-5.31
H3,4,9	-0.23	0.66	0.92	0.033	0.081	0.119	0.049	-2.68	-7.41	0.34	1.68
H3,4,10	-0.09	0.40	1.22	0.018	0.254	0.107	0.092	1.83	5.38	1.63	5.96
H3,4,11	-0.32	2.47	7.51	0.013	0.121	0.012	0.041	-2.07	-4.77	-6.20	-19.55
H3,5,6	-0.24	-0.21	0.16	0.005	0.003	0.311	0.104	-0.38	0.30	-0.32	0.31
H3,5,7	0.15	0.41	3.32	0.002	0.112	0.069	0.044	-0.22	-0.93	-0.17	-0.45
H3,5,8	-4.52	0.61	-14.54	0.000	0.147	0.010	0.049	0.23	-8.32	0.22	-12.75
H3,5,9	-0.27	-0.20	1.92	0.001	0.145	0.102	0.059	-0.34	3.77	-0.05	1.18
H3,5,10	-0.05	-0.09	1.81	0.001	0.301	0.086	0.104	-0.15	1.78	-0.11	1.97
H3,5,11	-0.04	0.45	-9.16	0.000	0.115	0.010	0.038	-0.40	3.70	-0.18	2.93
H3,6,7	-0.11	0.10	1.06	0.001	0.110	0.210	0.079	-0.40	-2.17	-0.36	-0.98
H3,6,8	-1.48	0.14	-4.18	0.000	0.180	0.041	0.062	0.11	-5.16	0.13	-7.49
H3,6,9	-0.31	-0.81	9.73	0.001	0.208	0.029	0.070	-2.46	34.97	-0.05	1.39
H3,6,10	0.06	-0.10	2.67	0.000	0.344	0.066	0.117	-0.19	2.95	-0.16	2.97
H3,6,11	-0.18	0.13	-3.37	0.000	0.165	0.037	0.057	6.19	-73.31	-0.42	6.91
H3,7,8	14.69	15.08	44.72	0.017	0.106	0.002	0.036	-0.48	-4.02	-0.26	-5.53
H3,7,9	-0.13	1.14	1.37	0.054	0.097	0.096	0.049	-2.59	-6.76	0.17	1.63
H3,7,10	-0.01	0.59	1.54	0.025	0.258	0.086	0.091	2.07	5.76	0.74	5.23
H3,7,11	-1.57	-23.37	-60.28	0.010	0.072	0.001	0.024	-2.04	-4.43	-6.89	-41.75

H3,8,9	-1.38	-3.78	1.52	0.133	0.038	0.034	0.047	-4.93	1.54	-2.24	0.97
H3,8,10	-0.61	-1.85	1.59	0.104	0.166	0.054	0.068	-3.26	1.08	-4.12	1.34
H3,8,11	6.73	22.03	-19.44	0.076	0.084	0.003	0.038	-5.25	1.36	-5.75	1.61
H3,9,10	0.13	-1.49	3.13	0.029	0.162	0.027	0.056	-3.00	3.20	2.13	-1.59
H3,9,11	-0.19	1.36	-2.92	0.036	0.137	0.036	0.049	25.02	-20.86	1.59	-1.03
H3,10,11	-0.04	1.48	-1.51	0.162	0.111	0.056	0.068	2.86	-2.23	4.72	-4.07
H4,5,6	-0.06	-0.34	0.16	0.005	0.002	0.080	0.027	0.17	-0.33	0.20	-0.38
H4,5,7	0.22	0.10	2.02	0.001	0.077	0.032	0.028	-0.01	1.02	0.01	0.56
H4,5,8	-2.78	0.24	-9.35	0.000	0.143	0.006	0.048	-0.51	9.20	-0.47	15.80
H4,5,9	-0.08	-0.33	1.84	0.003	0.170	0.052	0.059	0.13	-4.17	-0.14	-1.46
H4,5,10	0.13	-0.22	1.62	0.002	0.337	0.044	0.113	-0.08	-1.97	-0.07	-2.45
H4,5,11	0.12	0.22	-7.39	0.000	0.109	0.005	0.037	0.19	-4.09	0.03	-3.63
H4,6,7	0.15	0.03	1.56	0.000	0.073	0.040	0.028	-0.01	0.98	0.03	0.60
H4,6,8	-1.51	0.05	-5.05	0.000	0.148	0.012	0.050	-0.25	2.33	-0.27	4.63
H4,6,9	0.51	2.13	-28.07	0.001	0.212	0.004	0.071	0.92	-15.79	-0.16	-0.86
H4,6,10	0.49	-0.30	4.90	0.001	0.436	0.019	0.146	-0.11	-1.33	-0.10	-1.83
H4,6,11	0.05	0.06	-4.41	0.000	0.118	0.008	0.040	-2.99	33.09	0.07	-4.27
H4,7,8	2.17	3.30	6.09	0.023	0.089	0.006	0.031	1.04	-0.15	0.54	0.48
H4,7,9	0.15	1.65	0.56	0.062	0.032	0.032	0.025	0.96	-0.25	0.51	-0.14
H4,7,10	0.21	1.45	0.76	0.046	0.097	0.027	0.037	1.13	0.21	0.46	-0.45
H4,7,11	1.25	19.46	46.36	0.011	0.071	0.000	0.024	0.98	-0.16	1.11	3.58
H4,8,9	-1.70	-5.59	0.85	0.136	0.015	0.010	0.046	1.84	-3.33	-6.52	-2.06

H4,8,10	-1.28	-4.57	1.01	0.129	0.053	0.011	0.047	-1.78	-2.35	-2.53	-2.84
H4,8,11	1.35	4.10	-6.77	0.046	0.096	0.004	0.035	2.54	-2.96	0.93	-3.41
H4,9,10	0.51	-3.53	4.54	0.025	0.085	0.004	0.030	-1.64	-1.19	1.31	-4.63
H4,9,11	0.05	0.57	-4.23	0.009	0.119	0.009	0.040	-12.08	7.77	-0.26	-2.99
H4,10,11	0.12	0.72	-3.69	0.032	0.109	0.009	0.038	-1.38	-1.22	-0.76	-2.50
H5,6,7	-0.62	0.37	-4.54	0.001	0.083	0.003	0.028	1.83	5.70	2.11	3.09
H5,6,8	3.82	0.38	13.42	0.000	0.128	0.001	0.043	0.48	13.57	0.57	23.62
H5,6,9	-1.49	-4.89	71.45	0.001	0.212	0.000	0.071	7.26	-92.03	1.14	-4.37
H5,6,10	-1.74	1.38	-16.03	0.001	0.468	0.001	0.156	1.29	-7.75	1.46	-9.36
H5,6,11	-0.33	0.33	12.27	0.000	0.098	0.000	0.033	-15.50	192.93	2.30	-21.79
H5,7,8	-54.44	-61.09	-162.31	0.018	0.105	0.000	0.035	-2.05	18.46	-1.15	32.40
H5,7,9	-0.70	-5.85	2.51	0.119	0.076	0.003	0.047	7.63	31.05	-3.66	-9.56
H5,7,10	-0.40	-6.66	3.66	0.108	0.237	0.002	0.087	-13.77	-26.46	-7.01	-30.61
H5,7,11	1.85	33.82	104.90	0.008	0.071	0.000	0.024	5.10	20.38	37.72	244.51
H5,8,9	6.57	21.99	0.35	0.152	0.002	0.000	0.051	14.55	6.59	47.20	4.36
H5,8,10	6.66	22.03	0.95	0.152	0.012	0.000	0.051	21.70	4.64	38.75	6.00
H5,8,11	-5.99	-18.48	34.51	0.042	0.099	0.000	0.036	13.16	5.85	31.45	7.22
H5,9,10	-2.14	17.66	-17.96	0.043	0.117	0.000	0.042	19.98	-9.43	-20.03	33.53
H5,9,11	-0.31	1.73	16.14	0.008	0.130	0.000	0.044	-62.69	61.52	-8.71	21.64
H5,10,11	-0.10	2.12	17.80	0.023	0.129	0.000	0.044	-7.15	14.87	-25.81	38.30
H6,7,8	-185.55	-193.01	-559.53	0.017	0.106	0.000	0.036	-4.24	10.09	-2.01	15.36
H6,7,9	-0.12	2.58	16.06	0.022	0.202	0.001	0.068	1.05	16.96	-3.20	-4.53

H6,7,10	1.27	-3.98	17.58	0.028	0.499	0.001	0.167	-10.64	-14.46	-4.78	-14.51
H6,7,11	6.73	104.95	287.17	0.010	0.071	0.000	0.024	-0.33	11.13	16.42	115.92
H6,8,9	1628.72	5061.73	-2383.09	0.193	0.064	0.000	0.068	2.00	13.59	41.23	7.63
H6,8,10	6.91	19.90	7.78	0.207	0.150	0.001	0.085	16.77	9.58	26.47	10.49
H6,8,11	-34.63	-112.04	115.14	0.069	0.089	0.000	0.038	-0.85	12.07	13.69	12.63
H6,9,10	-1.99	19.97	-20.21	0.081	0.217	0.000	0.077	15.44	-1.30	-13.68	29.29
H6,9,11	0.38	-10.59	40.39	0.040	0.272	0.000	0.092	4.04	8.48	-3.79	18.90
H6,10,11	0.82	8.75	15.94	0.172	0.211	0.001	0.091	0.46	11.49	-11.24	26.15
H7,8,9	-1.04	-3.13	0.06	0.107	0.001	0.016	0.036	1.91	-3.21	-12.89	-3.80
H7,8,10	-1.02	-3.09	0.11	0.106	0.007	0.016	0.036	-1.58	-2.26	-5.53	-5.22
H7,8,11	-0.14	-0.27	-2.32	0.006	0.070	0.011	0.024	2.58	-2.85	0.83	-6.29
H7,9,10	0.25	-2.59	3.02	0.043	0.133	0.011	0.047	-1.45	-1.24	2.86	-9.15
H7,9,11	-0.06	-0.01	-2.54	0.000	0.072	0.011	0.024	-12.29	8.07	-0.23	-5.91
H7,10,11	-0.06	-0.01	-2.55	0.000	0.073	0.011	0.024	-1.40	-1.08	-0.68	-5.47
H8,9,10	-0.41	0.86	-1.00	0.040	0.123	0.066	0.048	0.92	-0.65	-0.52	0.71
H8,9,11	-0.31	-0.01	0.87	0.001	0.085	0.078	0.038	-4.76	4.23	-0.28	0.46
H8,10,11	-0.31	0.03	0.84	0.004	0.083	0.079	0.038	-0.54	0.69	-0.82	0.99
H9,10,11	0.12	1.14	1.00	0.104	0.062	0.035	0.042	0.11	0.74	2.96	-1.91

[a] The errors on $B_{ikl}^{\text{solution}}$, $C_{ikl}^{\text{solution}}$ and $D_{ikl}^{\text{solution}}$ are typically between 10-20%. [b] $AF_{\alpha} = \sqrt{\frac{\sum_j \left(\delta_{\alpha j}^{\text{para obs}} / \langle S_z \rangle_j - \delta_{\alpha j}^{\text{para calc}} / \langle S_z \rangle_j\right)^2}{\sum_j \left(\delta_{\alpha j}^{\text{para obs}} / \langle S_z \rangle_j\right)^2}}$ ($\alpha = i, k, l$) and $AF_{ikl} = 1/3\sqrt{AF_i^2 + AF_k^2 + AF_l^2}$ (see text) [c] Calculated from the crystal structure of $[Eu_3(L1)_3](CF_3SO_3)_9(CH_3CN)_9(H_2O)_2$ [16] after avering to D_3 symmetry. [d] Calculated from the D_3-symmetrical optimized structure for $[Eu_3(L1)_3]^{9+}$ in the gas phase.

Table S2 Geometrical Factors G_l^1 and $G_l^2 + G_l^3$ for the Aromatic Protons H1-H11 Calculated in the Structure of $[Eu_3(L1)_3]^{9+}$ Optimized in the Gas Phase ($G_l^{m\,\text{gas-phase}}$) and in Solution ($G_l^{m\,\text{solution}}$) with Eqns (22) and (23) for $[Ln_3(L1)_3]^{9+}$ (CD_3CN, 298 K)

Proton	$G_l^{1\,\text{gas-phase}}$	$G_l^{2\,\text{gas-phase}} + G_l^{3\,\text{gas-phase}}$	$G_l^{1\,\text{solution}}$	$G_l^{2\,\text{solution}} + G_l^{3\,\text{solution}}$
H1	-1.21E-03	2.48E-03	7.77E-03	-8.75E-04
H2	3.98E-03	-1.70E-03	2.38E-03	-1.06E-03
H3	3.02E-03	-1.21E-03	[a]	[a]
H4	-7.41E-04	2.90E-03	5.39E-03	-1.50E-03
H5	-1.53E-02	-7.11E-03	-2.02E-02	-8.23E-03
H6	-5.88E-03	-1.12E-02	-1.09E-02	-1.17E-02
H7	-9.24E-04	5.36E-03	3.28E-03	5.04E-04
H8	-5.04E-04	-2.94E-05	-1.03E-03	-7.60E-05
H9	1.95E-03	-1.31E-03	-5.37E-04	-8.05E-04
H10	7.10E-04	-9.96E-04	[a]	[a]
H11	8.01E-05	-8.57E-04	-1.30E-03	7.24E-05

[a] The G_l^1 and $G_l^2 + G_l^3$ for H3 and H10 in the structure of $[Eu_3(L1)_3]^{9+}$ optimized in the gas phase have been taken as references for calculating the related geometrical factors in solution for the remaining aromatic protons.

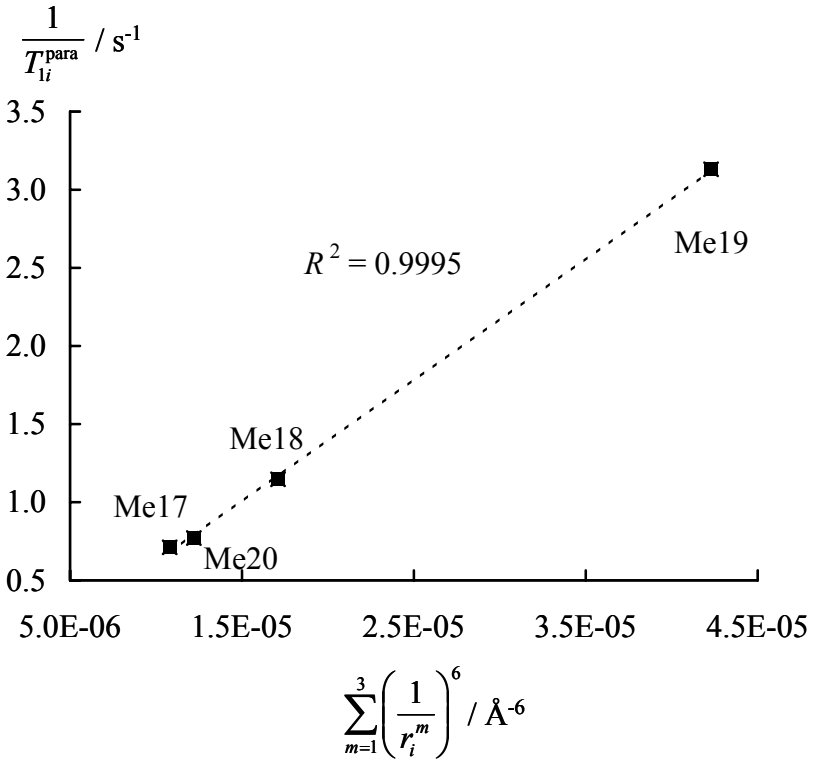

Figure S1 Plot of $1/T_{1i}^{\text{para}}$ *vs* $\sum_{m=1}^{3}\frac{1}{\left(r_i^m\right)^6}$ according to eqn (19) for the methyl groups Me17-Me20 in $[Ce_3(L1)_3]^{9+}$.

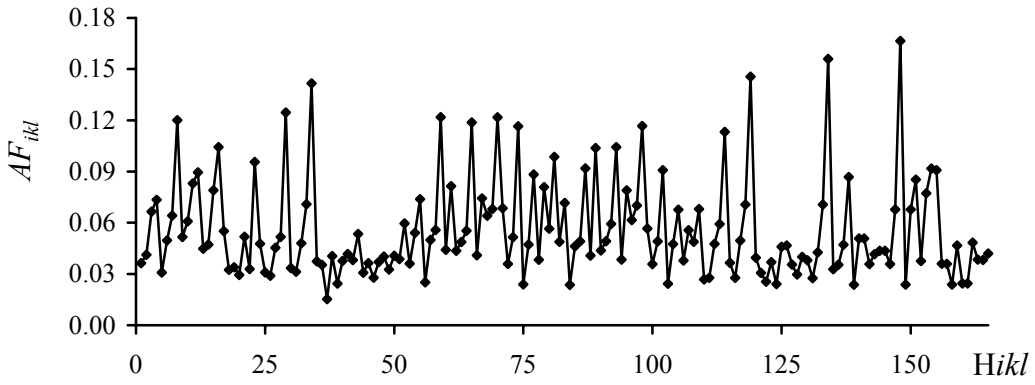

Figure S2

Figure S2 Plots of $AF_{ikl} = 1/3\sqrt{AF_i^2 + AF_k^2 + AF_l^2}$ for the 165 H*ikl* triplets in $[Ln_3(L1)_3]^{9+}$(Ln = Ce-Yb).

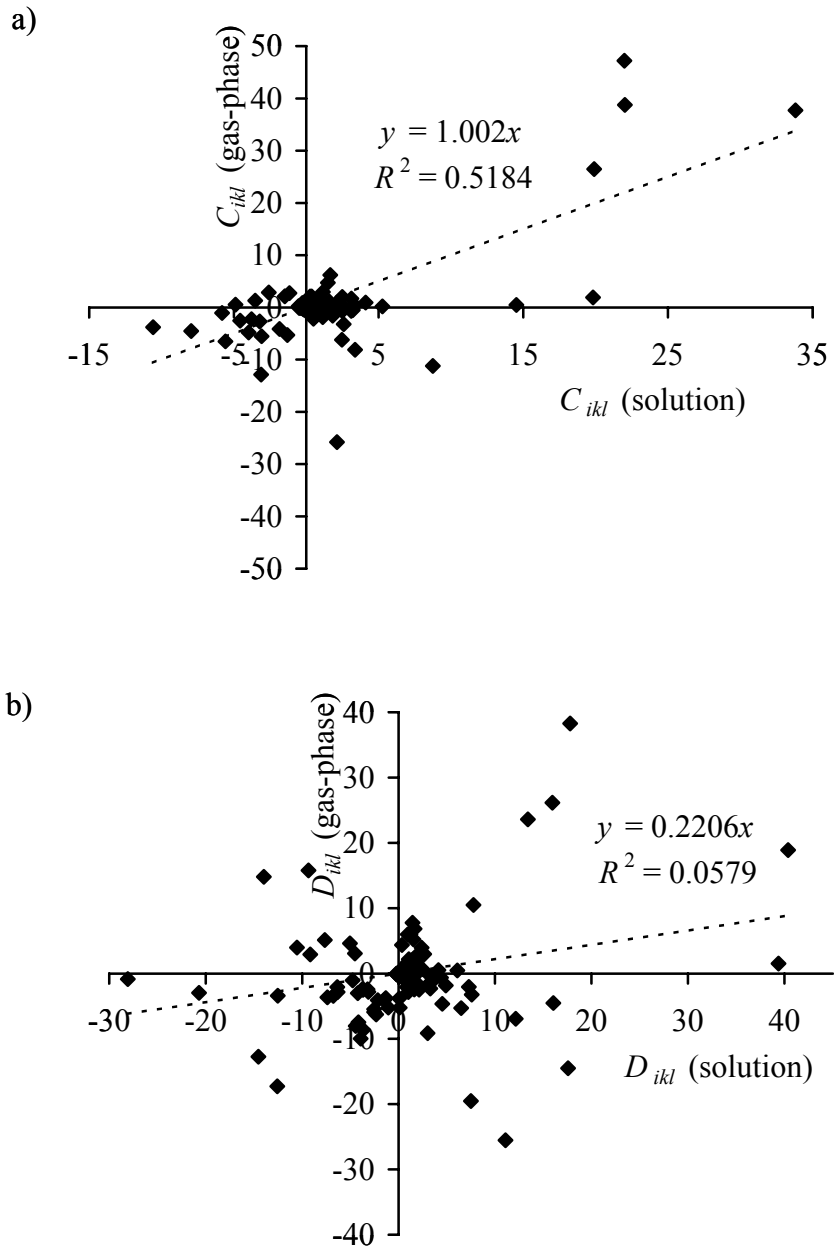

Figure S3 Plots of a) $C_{ikl}^{\text{gas phase}}$ *vs* $C_{ikl}^{\text{solution}}$, and b) $D_{ikl}^{\text{gas phase}}$ *vs* $D_{ikl}^{\text{solution}}$ for the 125 contributing H*ikl* triplets in $[Ln_3(L1)_3]^{9+}$.

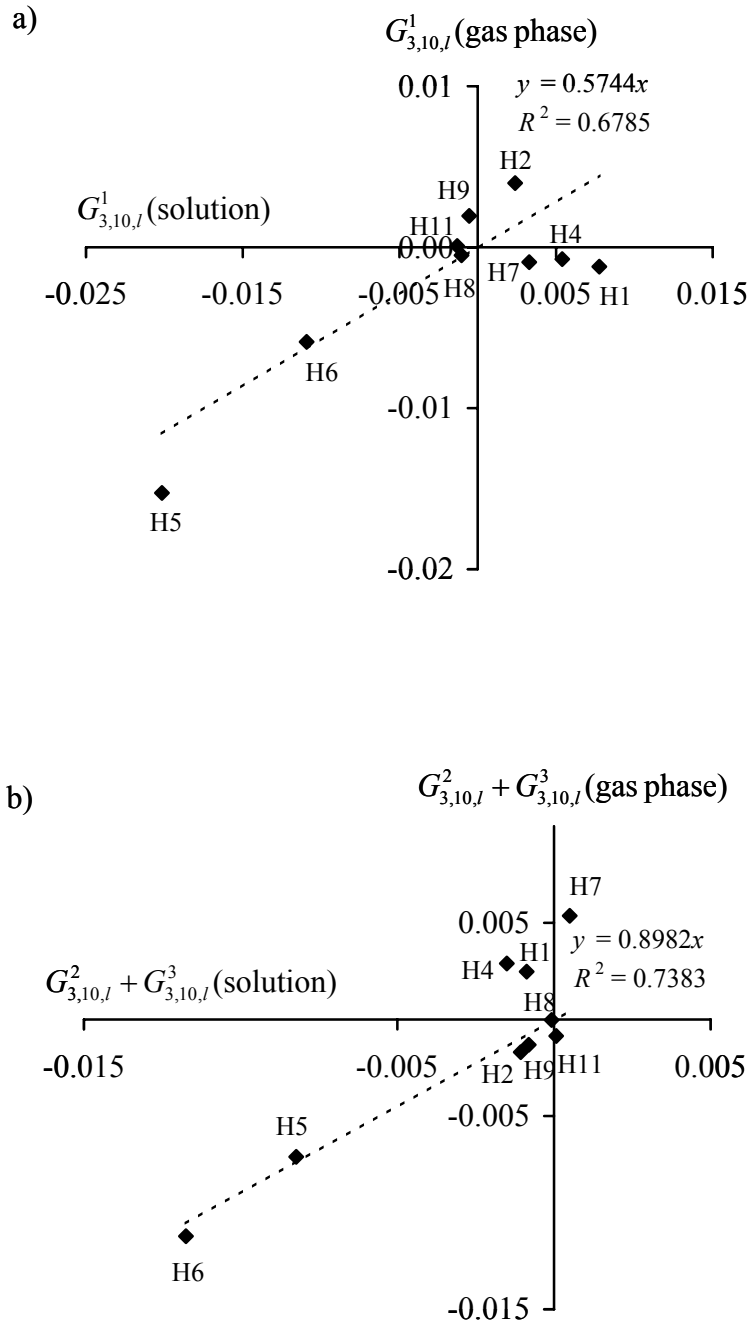

Figure S3

Figure S4 Plots of a) $G_l^{1\,\text{gas phase}}$ *vs* $G_l^{1\,\text{solution}}$, and b) $G_l^{2\,\text{gas phase}} + G_l^{3\,\text{gas phase}}$ *vs* $G_l^{2\,\text{solution}} + G_l^{3\,\text{solution}}$ for H1, H2, H4, H5, H6, H7, H8, H9, H11 (H3 and H10 are taken as references, see text).