Table S1 Unit cell parameters for the La(III) complexes in crystalline configuration before (Initial) and after (Opt.) unit cell optimization by DFT/PBE

No	Complex		*a*, Å	*b*, Å	*c*, Å	*α*, °	*β*, °	*γ*, °
1	$[La(CPDK_{8\text{-}1})_3]_2$Bpm	Initial	47.58	28.92	19.35	90.00	90.00	120.00
	1 molecule	Opt.	36.83	14.60	13.24	74.10	122.76	142.90
	3 molecules	MD	22.23	26.48	23.72	63.45	124.12	100.31
	4 molecules	MD	26.68	26.48	23.72	63.45	83.25	66.61
2	$[La(CPDK_{8\text{-}1})_3]_2$Bptz	Initial	46.41	18.54	30.51	90.00	90.00	120.00
		Opt.	26.15	10.27	16.56	92.41	90.57	122.15
3	$[La(CPDK_{8\text{-}1})_3]_2$BibzIm	Initial	50.07	18.37	30.81	90.00	90.00	120.00
		Opt.	26.56	9.79	16.89	88.21	90.34	119.85
4	$[La(CPDK_{8\text{-}1})_3]_2$Dpop	Initial	46.60	18.32	31.78	90.00	90.00	120.00
		Opt.	25.50	10.17	17.75	88.12	91.67	122.05
5	$[La(CPDK_{8\text{-}1})_3]_2$Tpp	Initial	51.26	16.91	39.55	90.00	90.00	120.00
		Opt.	27.31	9.19	20.33	89.41	88.97	121.64

Table S2 The calculated vertical energies of the lowest triplet (T_1) and singlet (S_1) excited states, intramolecular energy transfer (W_{ET}) and back transfer (W_{BT}) rates

No	Complex	S_1, eV	T_1, eV	Channels	W_{ET}, s^{-1}	W_{BT}, s^{-1}
1	$[Eu(CPDK_{8\text{-}1})_3]_2$Bpm	3.869	2.650	$T_1 \rightarrow {}^5D_1$	$6.02 \cdot 10^{6}$	$4.29 \cdot 10$
				$T_1 \rightarrow {}^5D_4$	$7.46 \cdot 10^{-3}$	$3.58 \cdot 10^{10}$
2	$[Tb(CPDK_{8\text{-}1})_3]_2$Bpm	3.785	2.646	$T_1 \rightarrow {}^5D_1$	$5.18 \cdot 10^{6}$	$5.52 \cdot 10$
				$T_1 \rightarrow {}^5D_4$	$4.55 \cdot 10^{-3}$	$3.26 \cdot 10^{10}$
3	$[Eu(CPDK_{8\text{-}1})_3]_2$Bptz	2.706	2.129	$T_1 \rightarrow {}^5D_1$	$2.24 \cdot 10^{8}$	$2.54 \cdot 10^{10}$
				$T_1 \rightarrow {}^5D_4$	$5.43 \cdot 10^{-3}$	$4.13 \cdot 10^{17}$
				$S_1 \rightarrow {}^5D_4$	$6.15 \cdot 10^{5}$	$1.10 \cdot 10^{4}$
4	$[Tb(CPDK_{8\text{-}1})_3]_2$Bptz	2.709	2.132	$T_1 \rightarrow {}^5D_1$	$2.02 \cdot 10^{8}$	$2.28 \cdot 10^{10}$
				$T_1 \rightarrow {}^5D_4$	$4.99 \cdot 10^{-3}$	$3.79 \cdot 10^{17}$
				$S_1 \rightarrow {}^5D_4$	$5.66 \cdot 10^{5}$	$1.14 \cdot 10^{4}$
5	$[Eu(CPDK_{8\text{-}1})_3]_2$BibzIm	3.486	2.553	$T_1 \rightarrow {}^5D_1$	$2.45 \cdot 10^{9}$	$1.31 \cdot 10^{6}$
6	$[Tb(CPDK_{8\text{-}1})_3]_2$BibzIm	3.414	2.539	$T_1 \rightarrow {}^5D_1$	$2.35 \cdot 10^{9}$	$2.19 \cdot 10^{6}$
7	$[Eu(CPDK_{8\text{-}1})_3]_2$Dpop	3.166	1.930	$T_1 \rightarrow {}^5D_1$	$2.49 \cdot 10^{7}$	$2.34 \cdot 10^{9}$
				$T_1 \rightarrow {}^5D_4$	$5.41 \cdot 10^{-4}$	$3.42 \cdot 10^{16}$
				$S_1 \rightarrow {}^5D_4$	$1.11 \cdot 10^{5}$	$6.75 \cdot 10^{4}$
8	$[Tb(CPDK_{8\text{-}1})_3]_2$Dpop	3.175	1.940	$T_1 \rightarrow {}^5D_1$	$2.13 \cdot 10^{7}$	$3.22 \cdot 10^{9}$
				$T_1 \rightarrow {}^5D_4$	$3.83 \cdot 10^{-4}$	$3.89 \cdot 10^{16}$
				$S_1 \rightarrow {}^5D_4$	$8.63 \cdot 10^{4}$	$1.99 \cdot 10^{4}$
9	$[Eu(CPDK_{8\text{-}1})_3]_2$Tpp	3.217	1.975	$T_1 \rightarrow {}^5D_1$	$2.34 \cdot 10^{7}$	$8.54 \cdot 10^{10}$
10	$[Tb(CPDK_{8\text{-}1})_3]_2$Tpp	3.213	1.968	$T_1 \rightarrow {}^5D_1$	$1.83 \cdot 10^{7}$	$7.43 \cdot 10^{10}$
11	$Gd(H_2O)_3Cl_3$Bpm	4.060	2.899	-	-	-
12	$Gd(H_2O)_3Cl_3$Bptz	3.818	2.454	-	-	-
13	$Gd(H_2O)_3Cl_3$BibzIm	3.213	1.980	-	-	-
14	$Gd(H_2O)_3Cl_3$Dpop	3.119	2.021	-	-	-
15	$Gd(H_2O)_3Cl_3$Tpp	3.255	2.275	-	-	-
16	$Gd(CPDK_{8\text{-}1})_3(H_2O)_2$	4.022	2.500	-	-	-

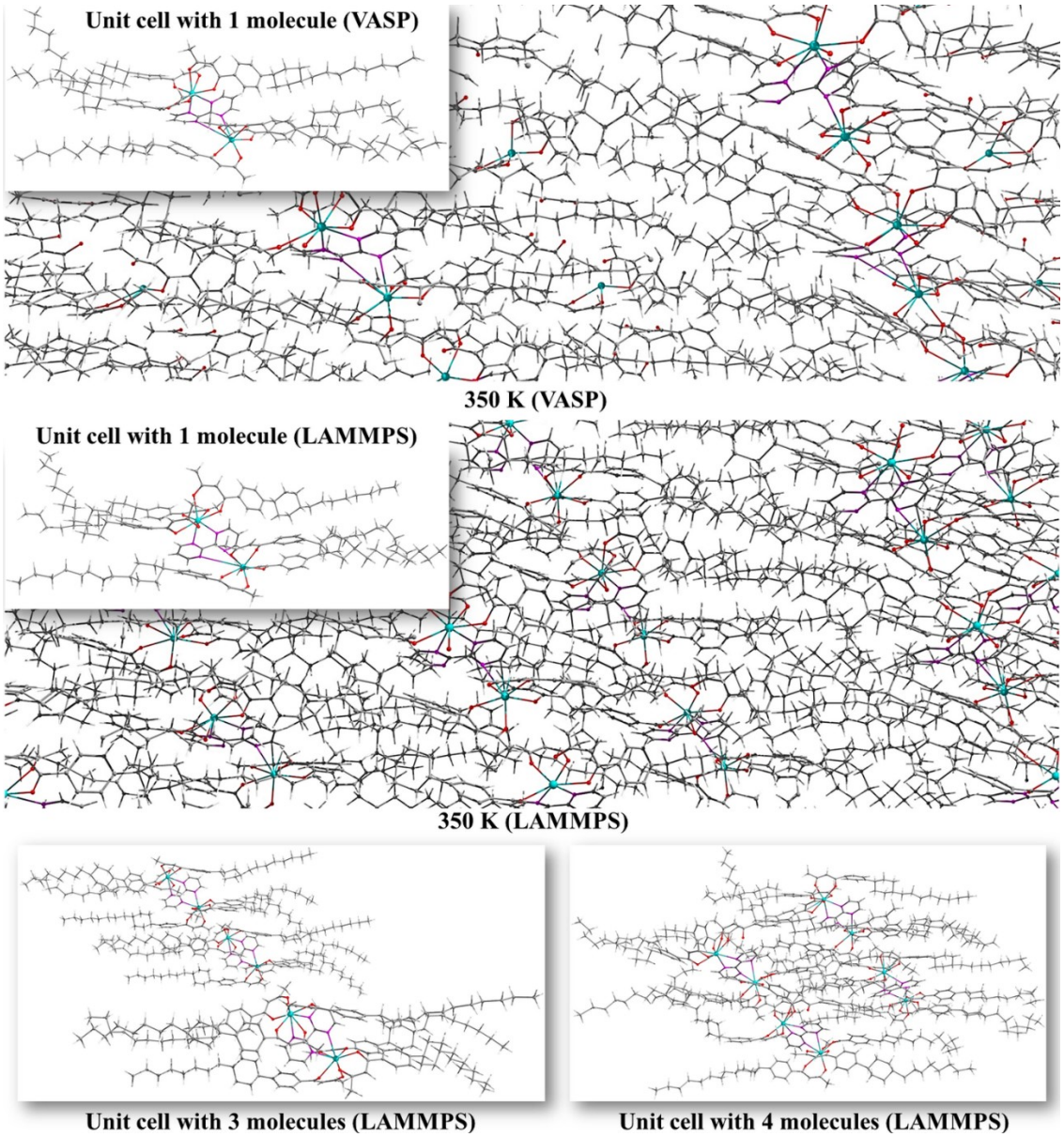

Fig. S1 Optimized geometries of crystalline configurations of the $[La(CPDK_{8\text{-}1})_3]_2Bpm$ complex and some snapshots from the corresponding MD simulations (view from (001)).

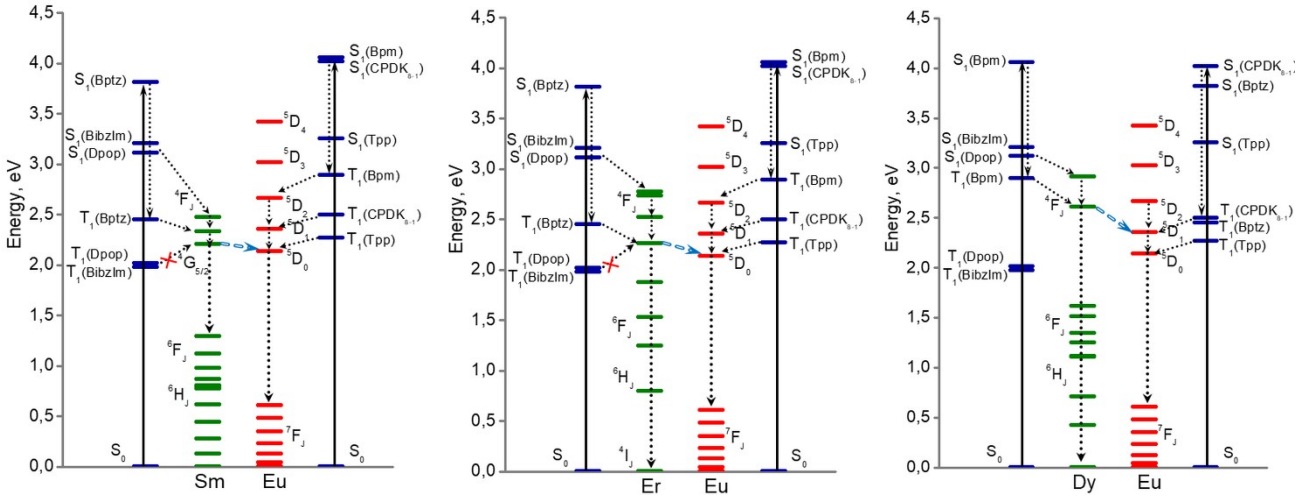

Fig. S2 The energy level diagram for the binuclear Ln complexes used in the analysis of channels of emission sensitization.