Electronic Supplementary Information for

Ordered and Flexible Lanthanide Complex Thin Films Showing Up-conversion and Color-tunable Luminescence

Rui Gao,[a] Minjun Zhao,[a] Yan Guan,[c] Xiaoyu Fang,[a] Xiaohong Li,[a] and Dongpeng Yan,*[,ab]

[a] *State Key Laboratory of Chemical Resource Engineering, Beijing University of Chemical Technology, Beijing 100029, China.*

[b] *Key Laboratory of Theoretical and Computational Photochemistry, Ministry of Education, College of Chemistry, Beijing Normal University, Beijing 100875, China*

[c] *College of Chemistry and Molecular Engineering, Peking University, Beijing, 100871, China.*

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Fig. S1 The FT-IR spectra of a) Eu(DBM)$_3$bath and b) Tb(acac)$_3$Tiron and their corresponding ligands, respectively.

For Fig. S1a, the peak at 1302 cm^{-1} (the C=N stretching vibration of the bath) appeared in Eu(DBM)$_3$bath, while the peaks at 1610 cm^{-1} (C=O stretching vibration), 750 cm^{-1} and 710 cm^{-1} (stretching vibration of benzene) of the DBM can also be observed in Eu(DBM)$_3$bath. For Fig. S1b, the peak at 1566 cm^{-1} can be attributed to the C=O stretching vibration in acac, and the 1244 cm^{-1} (S=O stretching vibration) of the Tiron also appeared in resulting Tb(acac)$_3$Tiron sample.

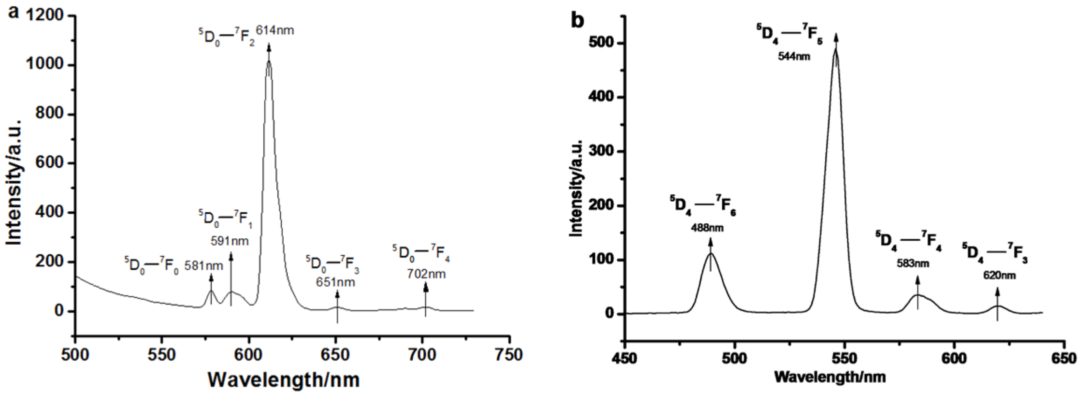
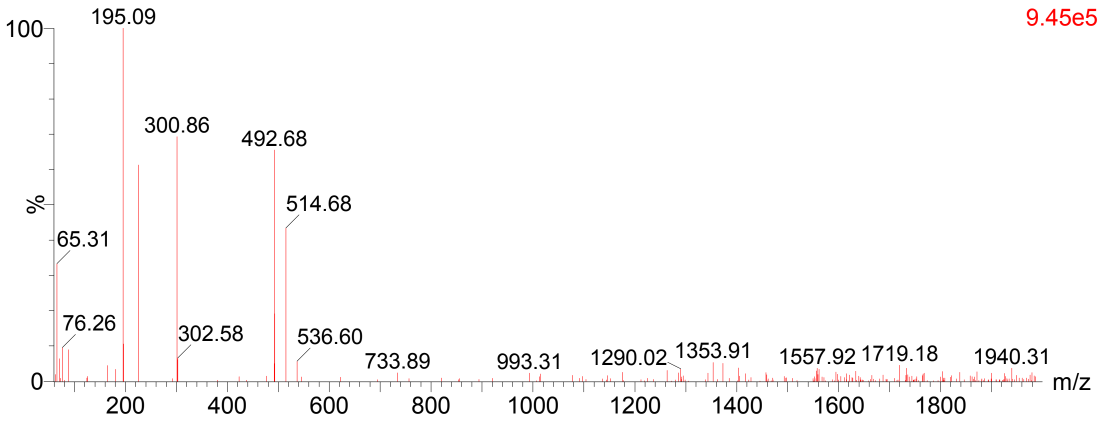


Fig. S2 Emission spectra of LC in solution (10^{-4} mol/L) for a) Eu(DBM)$_3$bath and b) Tb(acac)$_3$Tiron.

Table S1 The comparison of the elemental analysis data between the experimental (Exp.) and theoretical calculated (Cal.) results for two LC systems.

Complexes	C/%	H/%	N/%	S/%
	Exp. (Cal.)	Exp. (Cal.)	Exp. (Cal.)	Exp. (Cal.)
Eu(DBM)$_3$bath	58.65 (58.58)	3.80 (3.65)	1.71 (1.98)	3.39 (4.52)
Tb(acac)$_3$Tiron	34.35 (34.76)	2.57 (3.58)	0.10 (0)	8.91 (8.83)

a)



b)

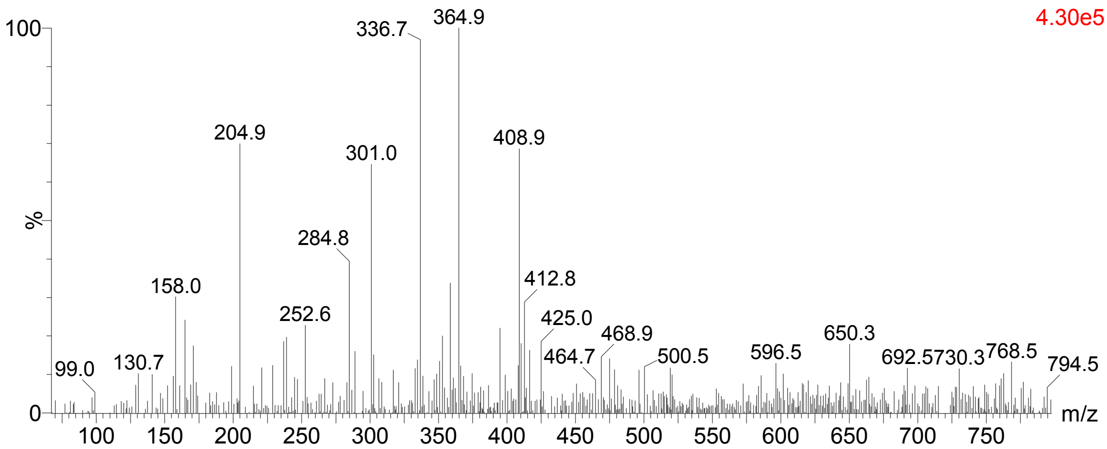


Fig. S3 The MS analysis data of LCs. a) Eu(DBM)$_3$bath and b) Tb(acac)$_3$Tiron

Based on the MS analysis shown in Fig. S3a, it can be observed that the m/z=224.9 matched well with the ligand DBM (M.W.=224). And the m/z=492.68 matched well with the ligand bath (M.W.=490), since there are two N atoms in the bath, which can get two H$^+$. Based on the formula of (m+z)/z, we can deduce that the m/z=300.86 can be assigned to the peak of Eu(DBM)$_2$ ((152+224×2+2)/2)=301). Because the complex is hard to be ionized, the peaks for the supposed compound are still relative lower. Based on the MS analysis shown in Fig. S3b, it can be observed that the m/z=99 matched well with the ligand acac (M.W.=100). Based on the formula of (m+z)/z, we can extrapolate that the peak at 364.9 can be assigned to the Tb(acac)$_3$Tiron ((727+2)/2=364.5). It can be observed that the m/z=130.7 matched well with the Tb(acac) ((159+100+2)/2=130.5). And the m/z=158.0 matched well with the Tb (M.W.=158.9).

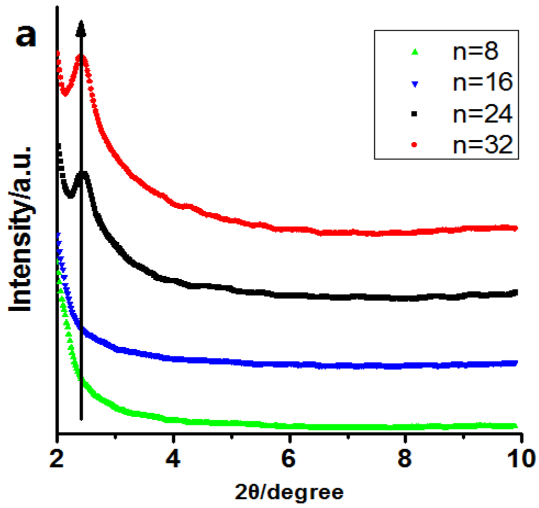
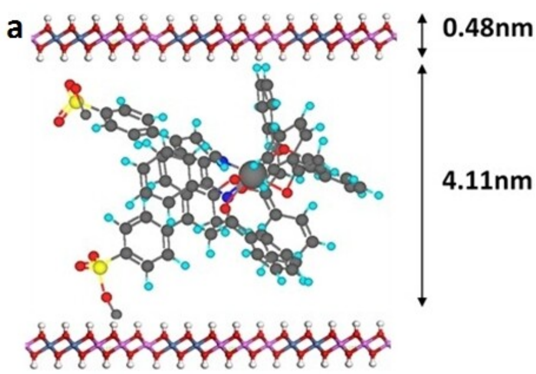


Fig. S4 The XRD profiles for (LC/LDH)$_n$ TFs. a) (Eu(DBM)$_3$bath/LDH)$_n$ (n=8–32) TFs; b) (Tb(acac)$_3$Tiron/LDH)$_n$ (n=8–32) TFs.



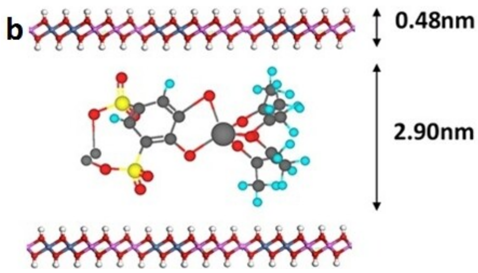


Fig. S5 Structural model of LC/LDHs. a) Eu(DBM)$_3$bath/LDH and b) Tb(acac)$_3$Tiron/LDH

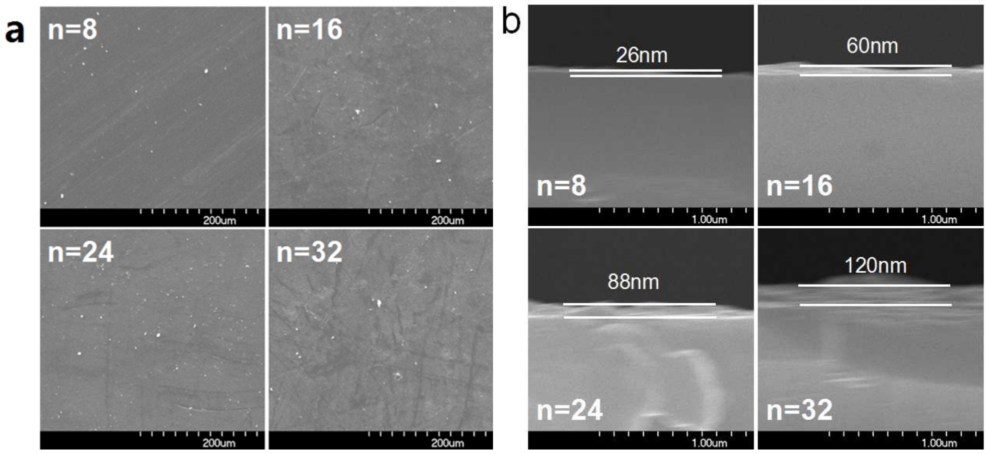


Fig. S6 The top (a) and side (b) view SEM profiles of the (Tb(acac)$_3$Tiron/LDH)$_n$

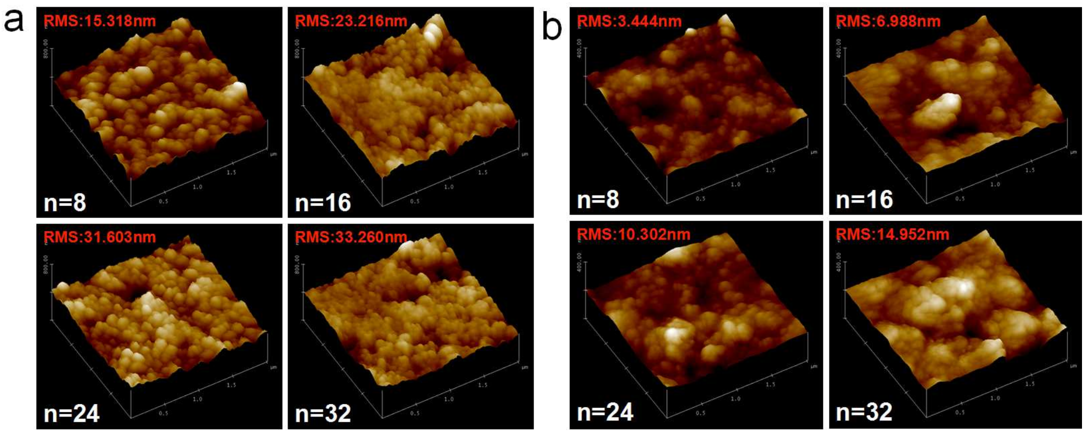


Fig. S7 AFM images of the (LC/LDH)$_n$ TFs for a) (Eu(DBM)$_3$bath/LDH)$_n$ (n=8–32) TFs; b) (Tb(acac)$_3$Tiron/LDH)$_n$ (n=8–32) TFs. Their RMS surface roughness values are also supplied.

Fig. S8 The fluorescence decay profiles of LC/LDH TFs and solutions. a) $(Eu(DBM)_3bath/LDH)_n$ (n=8–32) TFs. b) $(Tb(acac)_3Tiron/LDH)_n$ (n=8–32) TFs

Table S2. Fluorescence decay of the LC/LDH TFs $(Eu(DBM)_3bath/LDH)_n$ (a) and $(Tb(acac)_3Tiron/LDH)_n$ (b)

a

Samples	m	τ_i (ms)	A_i (%)	$\langle\tau\rangle$ (ms)	χ^2
n=8	1	0.3478	100.00		9.747
	2	0.1874	36.81	0.36	1.887
		0.4628	63.19		
n=16	1	0.3754	100.00		9.303
	2	0.2111	38.11	0.39	1.885
		0.4996	61.89		
n=24	1	0.3624	100.00		8.979
	2	0.1881	34.29	0.37	1.665
		0.4718	65.71		
n=32	1	0.3630	100.00		8.353
	2	0.1933	35.12	0.37	1.753
		0.4727	64.88		
Solution	2	0.3102	77.79	0.35	1.773
		0.5324	22.21		
Powder	2	0.18	46.31	0.27	2.46
		0.3627	53.69		

b Samples	m	τ_i (ms)	A_i (%)	<τ> (ms)	χ^2
n=8	1	0.3780	100.00		9.582
	2	0.1326	29.46	0.40	1.846
		0.5143	70.54		
n=16	1	0.4813	100.00		8.368
	2	0.1876	28.41	0.51	1.83
		0.6387	71.59		
n=24	1	0.4101	100.00		10.192
	2	0.1559	32.47	0.44	1.993
		0.5818	67.53		
n=32	1	0.4582	100.00		7.718
	2	0.1798	27.41	0.49	1.876
		0.6018	72.59		
Solution	2	0.2091	74.28	0.28	1.296
		0.5134	25.72		
Powder	2	0.1383	32.04	0.36	1.913
		0.4677	67.96		

m stands for the mono- or double-exponential fitting of the fluorescence decay curve; τ_i is the fluorescence lifetime, for m=1, lifetime is τ_1, and for m=2, two lifetimes are τ_1 and τ_2; A_i stands for the percentage of τ_i . The fitting goodness is indicated by the value of χ^2. In the double-exponential case, $<\tau> = A_1\tau_1 + A_2\tau_2$; $A_1 + A_2 = 1$. The samples are measured at 20 °C.

Table S3. Fluorescence lifetimes (ms) of the film samples of LC/LDH TFs and LC solutions at different temperature.

Samples	**20℃**	**40℃**	**60℃**	**80℃**
$Eu(DBM)_3$bath (solution)	0.3535	0.2304	0.1644	0.1221
$(Eu(DBM)_3bath/LDH)_8$	0.3524	0.2740	0.2218	0.2003
$Tb(acac)_3$Tiron (solution)	0.2825	0.2505	0.2435	0.2292
$(Tb(acac)_3Tiron/LDH)_8$	0.4132	0.3887	0.3476	0.2485

Table S4. Photoluminescence quantum yield of $(LC/LDH)_n$ (n = 12-28) UTFs.

Samples	n=12	n=20	n=28
$Eu(DBM)_3$bath	6.12%	7.40%	7.83%
$Tb(acac)_3$Tiron	8.17%	8.75%	10.72%

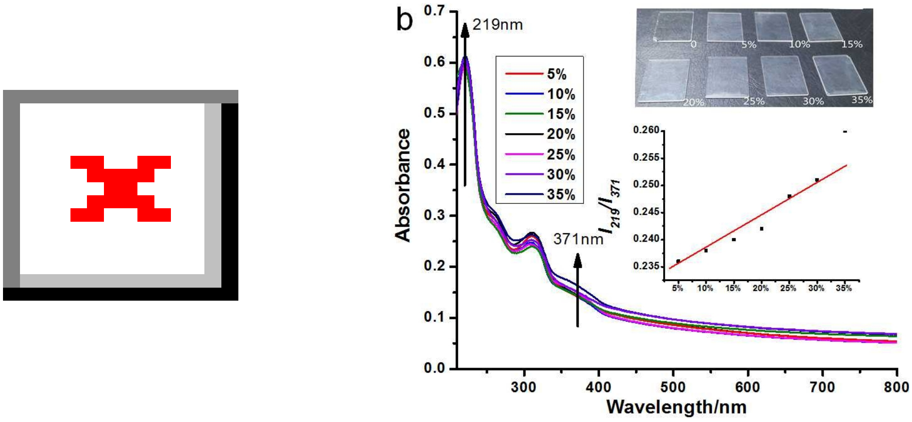


Fig. S9 UV–vis absorption spectra for a) $(Eu(DBM)_3bath/LDH)_{10}/(Tb(acac)_3Tiron/LDH)_n$ (n=0–14) b) $((Eu(DBM)_3bath)_c@Tb(acac)_3Tiron/LDH)_{10}$ (c=0%–35%, which stands for the molar ratio of $Eu(DBM)_3$bath to $Tb(acac)_3$Tiron). Insets show the plot of the absorbance intensity as a function of c, and photographs of TFs with different c when exposed to the daylight.

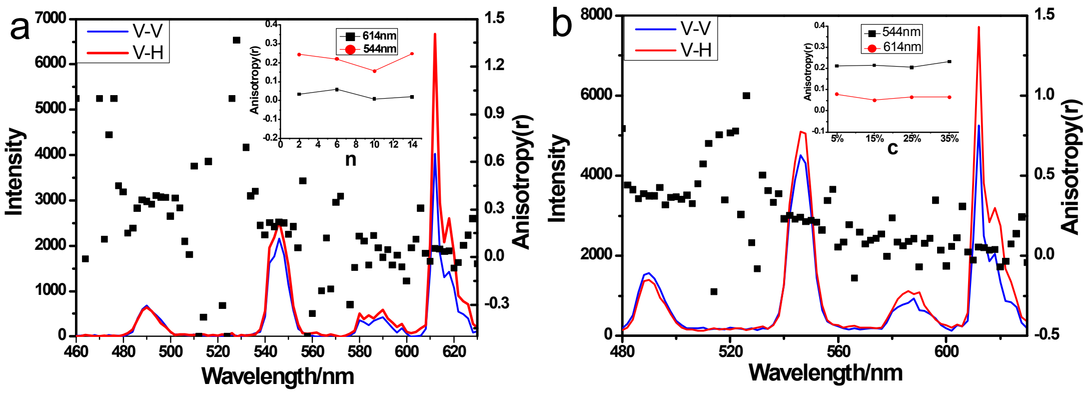


Fig. S10 The polarized fluorescence spectra for the two-color emissive $(LC/LDH)_n$ TFs. a) $(Eu(DBM)_3bath/LDH)_{10}/(Tb(acac)_3Tiron/LDH)_4$ b) $((Eu(DBM)_3bath)_{5\%}@Tb(acac)_3Tiron/LDH)_{10}$ (the insets show the plots of the intensity of the anisotropy as a function of n or c).