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"Synthesis, structure and optical properties of two novel polar dysprosium metal-organic frameworks: $[(CH_3)_2NH_2][Dy(HCOO)_4]$ and $[N_2H_5][Dy(HCOO)_4]$"

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Table S1. Crystal data, data collection and refinement results for DMFeMg at 298, 190, and 100 K. For both structures: $Z = 4$. Experiments were carried outwith Mo $K\alpha$ radiation. H atoms were treated by a mixture of independent and constrained refinement.

	HYD-Dy	**DMA-Dy**
Crystal data		
Chemical formula	$C_4H_9DyN_2O_8$	$C_6H_{12}DyNO_8$
M_r	375.63	388.67
Crystal system, space group	Orthorhombic, $Pca2_1$	Orthorhombic, $Pna2_1$
Temperature (K)	298	297
a, b, c (Å)	18.3359 (5), 6.60183 (17), 7.6049 (2)	8.7383 (1), 18.1604 (2), 6.7072 (1)
V (Å^3)	920.57 (4)	1064.37 (2)
μ (mm^{-1})	8.15	7.05
Crystal size (mm)	0.18 × 0.15 × 0.14	0.2 × 0.16 × 0.15
Data collection		
T_{min}, T_{max}	0.316, 1.000	0.566, 1.000
No. of measured, independent and observed [$I > 2\sigma(I)$] reflections	7972, 1715, 1689	22091, 2322, 2239
R_{int}	0.025	0.016
$(\sin\theta/\lambda)_{max}$ (Å^{-1})	0.610	0.658
Refinement		
$R[F^2 > 2\sigma(F^2)]$, $wR(F^2)$, S	0.015, 0.038, 1.12	0.012, 0.027, 1.11
No. of reflections	1715	2322
No. of parameters	143	145
No. of restraints	6	1
$\Delta\rho_{max}$, $\Delta\rho_{min}$ (e Å^{-3})	0.51, -0.53	0.33, -0.57
Absolute structure	Flack x determined using 745 quotients [(I+)-(I-)]/[(I+)+(I-)] (Parsons, Flack and Wagner, ActaCryst. B69 (2013) 249-259).	Flack x determined using 923 quotients [(I+)-(I-)]/[(I+)+(I-)] (Parsons, Flack and Wagner, ActaCryst. B69 (2013)
Absolute structure parameter	-0.032 (11)	-0.013(7)

Table S2. Selected distances and angles in DMA-Dy and HYD-Dy.

HYD-Dy			
Dy1—O5	2.291 (4)	O3—C1	1.270 (7)
Dy1—O8^{i}	2.336 (4)	O4—C2	1.254 (7)
Dy1—O2	2.367 (4)	O5—C3	1.227 (7)
Dy1—O3	2.383 (4)	O6—C2^{iii}	1.241 (6)
Dy1—O7	2.398 (4)	O7—C1^{iv}	1.229 (8)
Dy1—O1	2.398 (4)	O8—C4	1.228 (6)
Dy1—O6	2.406 (4)	C1—O7^{vi}	1.229 (8)
Dy1—O4	2.430 (3)	C2—O6^{vii}	1.241 (6)
O1—C3^{ii}	1.222 (7)	C3—O1^{viii}	1.222 (7)
O2—C4	1.264 (7)	N1—N2	1.425 (7)
O-Dy-O	71.0(2)- 147.5(2)		
O-C-O	124.3 (9)- 128.4 (5)		
DMA-Dy			
Dy1—O2	2.325 (2)	O2—C3	1.235 (4)
Dy1—O7	2.331 (2)	O3—C2	1.228 (4)
Dy1—O4	2.353 (2)	O4—C1^{xi}	1.240 (3)
Dy1—O6^{vii}	2.380 (2)	O5—C3^{xiv}	1.226 (4)
Dy1—O3	2.400 (2)	O6—C2	1.212 (4)
Dy1—O1^{ix}	2.400 (5)	O7—C1	1.241 (3)
Dy1—O8	2.433 (5)	O8—C4	1.230 (12)
Dy1—O5	2.464 (2)	N1—C5	1.445 (5)
O1—C4	1.255 (1)	O1—C6	1.452 (5)
O-Dy-O	68.43 (14)- 148.6 (1)		
O-C-O	124.3 (9)- 128.4 (5)		

(i) -*x*+1, -*y*, *z*-1/2; (ii) -*x*+3/2, *y*, *z*+1/2; (iii) -*x*+1, -*y*+1, *z*+1/2; (iv) *x*, *y*-1, *z*; (vi) *x*, *y*+1, *z*; (vii) -*x*+1, -*y*+1, *z*-1/2; (viii) -*x*+3/2, *y*, *z*-1/2; (ix) *x*, *y*, *z*+1; (xi) *x*-1/2, -*y*+3/2, *z*; (xii) *x*+1/2, -*y*+3/2, *z*; (xiii) -*x*+2, -*y*+1, *z*-1/2; (xiv) -*x*+2, -*y*+1, *z*+1/2.

Table S3. Selected hydrogen-bond parameters.

D—H···*A*	*D*—H (Å)	H···*A* (Å)	*D*···*A* (Å)	*D*—H···*A* (°)
HYD-Dy				
C3—H3···O7^{i}	0.93	2.57	3.101 (7)	116.4
N1—H1A···O1^{ii}	0.89	2.26	2.852 (7)	124.1
N1—H1A···O3^{ii}	0.89	2.29	3.132 (6)	158.1
N1—H1B···O1	0.89	2.57	3.265 (9)	135.3
N1—H1B···O2	0.89	2.13	2.919 (7)	148.0
N1—H1C···O4^{iii}	0.89	1.99	2.816 (7)	152.8
N1—H1C···O5^{iii}	0.89	2.56	3.210 (7)	130.4
N2—H2A···O6^{iv}	0.910 (3)	2.29 (2)	3.103 (8)	148 (3)
N2—H2B···O3	0.910 (3)	2.411 (8)	3.228 (7)	149.5 (7)
N2—H2B···O7^{v}	0.910 (3)	2.342 (9)	3.189 (7)	154.9 (12)
DMA-Dy				
N1—H1A···O4^{vi}	0.90	2.16	2.939 (4)	145.0
N1—H1A···O1^{vii}	0.90	2.32	3.076 (5)	141.5
N1—H1B···O5	0.90	2.03	2.893 (3)	160.6
N1—H1B···O7	0.90	2.41	3.000 (4)	123.2

Symmetry code(s): (i) -*x*+3/2, *y*, *z*-1/2; (ii) -*x*+3/2, *y*, *z*+1/2; (iii) *x*, *y*, *z*+1; (iv) -*x*+1, -*y*+1, *z*+1/2; (v) *x*, *y*+1, *z*; (vi) *x*+1/2, -*y*+3/2, *z*; (vii) *x*+1/2, -*y*+3/2, *z*+1.

Table S4. Factor group analysis for DMA-Dy. The number of nonequivalent $HCOO^-$ ions is four and, therefore, the total number of vibrational modes for this ion is four time larger than presented by the correlation diagram.

ion	vibration	Free ion symmetry	Site symmetry	Factor group symmetry
		C_{2v}	**C_1**	**C_{2v}**
$HCOO^-$	ν_1, ν_2 or ν_3	A_1	A	$A_1 + A_2 + B_1 + B_2$
	ν_4, ν_5 or ν_6	B_1	A	$A_1 + A_2 + B_1 + B_2$
	T’	$A_1 + B_1 + B_2$	3A	$3A_1 + 3A_2 + 3B_1 + 3B_2$
	L	$A_2 + B_1 + B_2$	3A	$3A_1 + 3A_2 + 3B_1 + 3B_2$
		C_{2v}	**C_1**	**C_{2v}**
DMA^+	$\nu_s(NH_2)$	A_1	A	$A_1 + A_2 + B_1 + B_2$
	$\nu_{as}(NH_2)$	B_2	A	$A_1 + A_2 + B_1 + B_2$
	$\delta(NH_2)$	A_1	A	$A_1 + A_2 + B_1 + B_2$
	$\rho(NH_2)$	B_2	A	$A_1 + A_2 + B_1 + B_2$
	$\omega(NH_2)$	B_1	A	$A_1 + A_2 + B_1 + B_{21}$
	$\tau(NH_2)$	A_2	A	$A_1 + A_2 + B_1 + B_2$
	$\nu_s(CNC)$	A_1	A	$A_1 + A_2 + B_1 + B_2$
	$\nu_{as}(CNC)$	B_1	A	$A_1 + A_2 + B_1 + B_2$
	$\delta(CNC)$	A_1	A	$A_1 + A_2 + B_1 + B_2$
	$\nu_s(CH_3)$	$A_1 + B_1$	2A	$2A_1 + 2A_2 + 2B_1 + 2B_2$
	$\nu_{as}(CH_3)$	$A_1 + B_1 + B_2 + A_2$	4A	$4A_1 + 4A_2 + 4B_1 + 4B_2$
	$\delta_s(CH_3)$	$A_1 + B_1$	2A	$2A_1 + 2A_2 + 2B_1 + 2B_{21}$
	$\delta_{as}(CH_3)$	$A_1 + B_1 + B_2 + A_2$	4A	$4A_1 + 4A_2 + 4B_1 + 4B_2$
	$\rho(CH_3)$	$A_1 + B_1 + B_2 + A_2$	4A	$4A_1 + 4A_2 + 4B_1 + 4B_2$
	$\tau(CH_3)$	$A_2 + B_2$	2A	$2A_1 + 2A_2 + 2B_1 + 2B_2$
	T’	$A_1 + B_1 + B_2$	3A	$3A_1 + 3A_2 + 3B_1 + 3B_2$
	L	$A_2 + B_1 + B_2$	3A	$3A_1 + 3A_2 + 3B_1 + 3B_2$
Dy(III)			**C_1**	**C_{2v}**
			3A	$3A_1 + 3A_2 + 3B_1 + 3B_2$

Table S5. Factor group analysis for HYD-Dy. The number of nonequivalent $HCOO^-$ ions is four and, therefore, the total number of vibrational modes for this ion is four time larger than presented by the correlation diagram.

Ion	Vibration	Free ion symmetry	Site symmetry	Factor group symmetry
		C_{2v}	**C_1**	**C_{2v}**
$HCOO^-$	ν_1, ν_2 or ν_3	A_1	A	$A_1 + A_2 + B_1 + B_2$
	ν_4, ν_5 or ν_6	B_1	A	$A_1 + A_2 + B_1 + B_2$
	T'	$A_1 + B_1 + B_2$	3A	$3A_1 + 3A_2 + 3B_1 + 3B_2$
	L	$A_2 + B_1 + B_2$	3A	$3A_1 + 3A_2 + 3B_1 + 3B_2$
		C_1	**C_1**	**C_{2v}**
hyd^+	$\nu_s(NH_2)$	A	A	$A_1 + A_2 + B_1 + B_2$
	$\nu_{as}(NH_2)$	A	A	$A_1 + A_2 + B_1 + B_2$
	$\delta(NH_2)$	A	A	$A_1 + A_2 + B_1 + B_2$
	$\rho(NH_2)$	A	A	$A_1 + A_2 + B_1 + B_2$
	$\omega(NH_2)$	A	A	$A_1 + A_2 + B_1 + B_2$
	$\nu_s(NH_3)$	A	A	$A_1 + A_2 + B_1 + B_2$
	$\nu_{as}(NH_3)$	A	2A	$2A_1 + 2A_2 + 2B_1 + 2B_2$
	$\delta_s(NH_3)$	A	A	$A_1 + A_2 + B_1 + B_2$
	$\delta_{as}(NH_3)$	A	2A	$2A_1 + 2A_2 + 2B_1 + 2B_2$
	$\rho(NH_3)$	A	2A	$2A_1 + 2A_2 + 2B_1 + 2B_2$
	τ	A	A	$A_1 + A_2 + B_1 + B_2$
	ν(NN)	A	A	$A_1 + A_2 + B_1 + B_2$
	T'	3A	3A	$3A_1 + 3A_2 + 3B_1 + 3B_2$
	L	3A	3A	$3A_1 + 3A_2 + 3B_1 + 3B_2$
			C_1	**C_{2v}**
Dy(III)			3A	$3A_1 + 3A_2 + 3B_1 + 3B_2$

Table S6. IR and Raman frequencies (in cm^{-1}) of DMA-Dy and suggested assignments.[a]

IR	Raman	Assignment
3101 w,b	3093 w	$\nu(NH_2)$
	3045 w	$\nu_{as}(CH_3)$
3029 w	3033 w	$\nu_{as}(CH_3)$
	2978 m	$\nu_s(CH_3)$
2969 w	2963 w	$\nu_s(CH_3)$
2927 w	2929 w	$\nu_s(CH_3)$
2878 w	2881 w	$\nu_1(HCOO)$
2862 w	2860 w	$\nu(NH_2)$
2845 w	2847 w	$\nu(NH_2)$
2822 w	2811 vw	$\nu(NH_2)$
	2751 vw	$2\nu_2(HCOO)$
2721 vw	2722 vw	$2\nu_2(HCOO)$
1675 w	1688 vw	$\delta(NH_2)$
1607 vs		$\nu_4(HCOO)$
1592 vs	1576 w	$\nu_4(HCOO)$
1480 w	1485 w	$\delta_{as}(CH_3)$
1467 vw	1467 w	$\delta_s(CH_3)$
1436w		$\omega(NH_2)$
1397 m	1398 sh	$\nu_5(HCOO)$
1386 sh	1387 vs	$\nu_5(HCOO)$
1379 m	1376 s	$\nu_5(HCOO)$
1358 m	1360 m	$\nu_2(HCOO)$
1344 m	1342 w	$\nu_2(HCOO)$
1247 vw		$\rho(CH_3)$
	1076 w	$\nu_6(HCOO)$
1027 w	1032 w	$\nu_{as}(CNC)$
905 vw	906 w	$\nu_s(CNC)$
851 w		$\rho(NH_2)$
801 sh		$\nu_3(HCOO)$

797 sh	797 w	ν_3(HCOO)
792 m		ν_3(HCOO)
786 sh	787 w	ν_3(HCOO)
780 sh	780 w	ν_3(HCOO)
294 w		T'(HCOO)
265 m	257 w	T'(HCOO)
192 m	198 w	L(HCOO)
178 w	187 w	L(HCOO)
	155 w	L(HCOO)
	115 w	L(HCOO)

[a]Key: vs, very strong; s, strong; m, medium; w, weak; vw, very weak;

Table S7. IR and Raman frequencies (in cm^{-1}) of HYD-Dy and suggested assignments.[a]

IR	Raman	Assignment
3328 vw		$\nu(NH_2)$ and $\nu(NH_3)$
3303 w	3301 w	$\nu(NH_2)$ and $\nu(NH_3)$
3206 w	3212 w	$\nu(NH_2)$ and $\nu(NH_3)$
3158 w		$\nu(NH_2)$ and $\nu(NH_3)$
3093 vw		$\nu(NH_2)$ and $\nu(NH_3)$
3020 w		$\nu(NH_2)$ and $\nu(NH_3)$
2965 w	2954 w	$\nu(NH_2)$ and $\nu(NH_3)$
2904 vw		$\nu(NH_2)$ and $\nu(NH_3)$
2874 w	2877 m	$\nu_1(HCOO)$
2841 vw		$\nu(NH_3)$
2735 w	2726 w	$2\nu_2(HCOO)$
2660 w		$2\nu_2(HCOO)$
2600 vw		$2\nu_2(HCOO)$
1646 m	1649 w	$\delta(NH_2)$
	1619 w	$\delta_{as}(NH_3)$
1590 vs	1584 w	$\nu_4(HCOO)$
1525 m	1528 w	$\delta_s(NH_3)$
1395 m	1399 vw	$\nu_5(HCOO)$
1388 sh	1388 vs	$\nu_5(HCOO)$
1381 sh		$\nu_5(HCOO)$
1378 m		$\nu_5(HCOO)$
1371 m	1374 w	$\nu_5(HCOO)$
1364 m	1367 m	$\nu_2(HCOO)$
1340 m	1342 s	$\nu_2(HCOO)$
	1222 w	$\tau(NH_2)$
1143 w		$\tau(NH_2)$
1115 w	1119 w	$\tau(NH_2)$
1090 vw	1100 vw	$\rho(NH_3)$
1081 vw	1083 w	$\rho(NH_3)$

1067 vw	1067 w	ν_6(HCOO)
1062 vw		ν_6(HCOO)
953 w	955 m	ν(NN)
942 w		ν(NN)
799 m	801 w	ν_3(HCOO)
795 sh	795 w	ν_3(HCOO)
791 sh		ν_3(HCOO)
786 w	786 w	ν_3(HCOO)
781 w	781 m	ν_3(HCOO)
770 w	773 w	$\omega(NH_2)$
765 w		$\omega(NH_2)$
	287 sh	T'(HCOO)
275 m	265 sh	T'(HCOO)
	241 w	L(HCOO)
201 m	195 sh	L(HCOO)
	171 m	L(HCOO)
149 vw	149 sh	L(HCOO)
	132 vw	L(HCOO)
	112 w	L(HCOO)
79 vw		T'(Dy)

[a]Key: vs, very strong; s, strong; m, medium; w, weak; vw, very weak;

Table S8. Comparison of the optical data (emission wavenumbers (λ_{em}) and lifetimes (τ)), crystal structures and the shortes distances between lanthaninde ions (d) for other dysprosium MOFs and coordination polymers as well as related lanthanide formates templated by protonated amines.

Dy(III)-based MOF	Space group	d_{Dy-Dy} (Å)	λ_{em} (nm)	τ (μs)	Ref.
Dy$_2$(1,4-bdc)$_3$	$P\bar{1}$			0.8±0.01	S1
Dy$_2$(atpa)$_3$(dmf)$_2$(H$_2$O)$_2$	$P\bar{1}$	4.3044(8)	–[a]		S2
[**Dy$_4$**(bpt)$_4$(dmf)$_2$(H$_2$O)$_8$]·(dmf)$_5$ ·(H$_2$O)$_3$	$P\bar{1}$		480, 573, 662[b]		S3
[**Dy$_2$**(*N*-bdc)$_3$(dmf)$_4$]·2dmf	$P\bar{1}$	4.0867(4)			S4
Dy(dcbpy)(dmf)$_2$(NO$_3$)	$P\bar{1}$	5.1707(5)			S5
[**Dy$_3$**(bptc)$_3$(H$_2$O)$_2$]·3H$_3$O·4H$_2$O· 3dmf	$P\bar{1}$	4.0091(4)	484, 576, 664, 752[c]	2.117±0.014 (τ_1), 12.65±0.151 (τ_2)	S6
Dy$_2$(1,4-bdc)$_3$(H$_2$O)$_4$	Pm		481, 575[d]	1.5±0.02	S1
[**Dy**(pta)(H$_2$O)$_3$]·4H$_2$O	$P2_1/c$		476, 569[e]		S7
Dy(1,4-bdc)(HCOO)	$P2_1/c$				37
[**Dy$_2$**(pda$_3$)(H$_2$O)]·2H$_2$O	$P2_1/c$				S8
[**Dy**(cmdcp)(H$_2$O)$_3$](NO$_3$)·2H$_2$O	$P2_1/n$	4.6484(3)			S9
Dy$_2$(hfipbb)$_3$	$Pnan$		~575[f]		S10
Dy(H$_2$cmp)(H$_2$O)	$Pbca$	5.5925(43)			S11
Dy$_4$(μ_4-H$_2$O)(tatb)$_{8/3}$(SO$_4$)$_2$·3H$_2$O ·10C$_2$H$_6$SO	$Im\bar{1}m$	6.392	485, 579, 667[g]		S12
Heterometallic Dy(III)-based MOF	Space group	d_{Dy-Dy} (Å)	λ_{em} (nm)	τ (μs)	Ref.
[**DyCd**(imdc)(SO$_4$)(H$_2$O)$_3$] ·0.5H$_2$O	$P2_1/c$	4.0034(3)	478,573, 657[h]	1130	59
[**DyAg**(pda)$_2$(H$_2$O)$_3$]·2.5H$_2$O	$P2_1/n$	7.364(1)	483, 573[i]		S13
K$_5$[**Dy$_5$**(idc)$_4$(ox)$_4$]·20H$_2$O	$P\bar{4}2_1c$		479, 573[j]		S14
KDy(ox)$_2$(H$_2$O)$_4$	$I4_1/amd$	6.172(2)	485, 576[k]	7.984 (τ_1), 1.558 (τ_2)	S14
KDy(ox)$_2$	$I4_1/amd$		~485, ~576[k]	10.258 (τ_1), 2.063 (τ_2)	S15
Dy$_2$Cu(μ_6-O)$_{1/6}$(μ_3-OH)$_{2/3}$(μ_3-OH)$_2$(hpzc)$_2$(H$_2$O)$_2$·2H$_2$O	$R\bar{3}$	3.6924(8)			S16

Amine templated Ln(III)-based MOF	Space group	d_{Ln-Ln} (Å)	λ_{em} (nm)	τ (μs)	Ref.
Sm$(HCOO)_3 \cdot (HCOONH_2)_2$	$C2$	6.655(2)	558, 595, 642[l]		42
(NH_2CHNH_2)**Sm**$(HCOO)_4$	$C222_1$	6.7668(1)			
Eu$(HCOO)_3 \cdot (HCOONH_2)_2$	$C2$	6.6405(9)	589, 614, 648, 697[m]		42
(NH_2CHNH_2)**Eu**$(HCOO)_4$	$C222_1$		594, 618, 700[n] (590, 615, 686, 692)[o]	1700 (2128±2)	40(41)
Tb$(HCOO)_3 \cdot (HCOONH_2)_2$	$C2$	6.6080(6)	488, 542, 586, 620, 643, 668, 678[p]		42
(NH_2CHNH_2)**Tb**$(HCOO)_4$	$C222_1$		492, 544, 590, 624[n] (491, 544, 585, 621)[o]	2000 (2132±2)	40(41)
Dy$(HCOO)_3 \cdot (HCOONH_2)_2$	$C2$				42
(NH_2CHNH_2)**Dy**$(HCOO)_4$	$C222_1$		490, 574[n]	2.1	40(41)
(NH_4)**Er**$(HCOO)_4$	$P2_1$		255, 364, 379, 407, 451, 488, 522, 545, 653[l]		38
(CH_3NH_3)**Er**$(HCOO)_4$	$P2_1$				
$[tmenH_2]$**Er**$[(HCOO)_4]_2$	$C2/c$	6.6299(1)			39
$(CH_3CH_2NH_3)$**Er**$(HCOO)_4$	$P2_1$		255, 364, 379, 407, 451, 488, 522, 545, 653[l]		38
$(HOCH_2CH_2NH_3)$**Er**$(HCOO)_4$	$P2_1$				
$[C(NH_2)_3]$**Er**$(HCOO)_4$	$P2_12_12_1$				
$(C_3H_5N_2)$**Er**$(HCOO)_4$	$P2_12_12_1$				
(NH_2CHNH_2)**Er**$(HCOO)_4$	$C222_1$	6.5577(5)			38(41)
$[dmenH_2]$**Er**$[(HCOO)_4]_2$	$Pna2_1$				39
(NH_2CHNH_2)**Yb**$(HCOO)_4$	$C222_1$				41

This work	Space group	d_{Dy-Dy} (Å)	λ_{em} (nm)	τ (μs)
HYD-Dy	$Pca2_1$	5.975(1)	480, 574	96
DMA-Dy	$Pna2_1$	6.6444(1)		57

Key: 1,4-bdc (1,4-benzenedicarboxylate), atpa (2-aminoterephtalate), dmf (*N*,*N*-dimethylformamide), bpt (biphenyl-3,4',5-tricarboxylate), *N*-bdc (2-amino-1,4-benzenedicarboxylate), dcbpy (2,2'-bipyridine-4,4'-dicarboxylate), bptc (3,3',5,5'-biphenyltetracarboxylate), pta (2,4,6-pyridinetricarboxylate), pda (1,4-phenylenediacetate), cmdcp (zwitterionic *N*-carboxymethylo-(3,5-dicarboxylate)-pyridine), hfipbb (4,4'-(hexafluoroisopropylidene)bisbenzoate),

H_2cmp ((*N*-carboxymethylo)iminodi(methylphosphonate), H_3imdc (4,5-imidazoledicarboxylic acid), tatb (4,4',4''-*s*-triazine-2,4,6-triyl-tribenzoate), hpzc (3-hydoxypyrazine-2-carboxylate), pda (pyridine-2,6-dicarboxylate), idc (imidazole-3,5-dicarboxylate), ox (oxalate), dmen (protonated N,N'-dimethylethylenediamine), tmen (protonated N,N,N',N'-tetramethylethylenediamine). Excitation wavenumber (nm): [a]300 (–)-no characteristic Dy^{III} bands observed due to LMCT emission band, [b]320, [c]345, [d]312, [e]305, [f]364, [g]325, [h]312, [i]293, [j]307, [k]365, [l]not given, [m]394, [n]395, [o]350, [p]370.

Table S9. Electric-dipole oscillator strengths and Judd-Ofelt parameters Ω_λ for Dy(III) in HYD-Dy crystalline host.

Transition from $^6H_{15/2}$	Energy		Oscillator strength (10^{-6})		Residual
	(cm^{-1})	(nm)	P_{exp}	P_{calc}	$\|\Delta P\|$ (10^{-6})
$^6H_{11/2}$	5811	1721	2.7083	2.7198	0.01
$^6F_{9/2}$+ $^6H_{7/2}$	9019	1109	4.3474	4.4556	0.11
$^6H_{5/2}$	10204	980	0.1910	0.0100	0.18
$^6F_{7/2}$	10984	910	3.2965	3.5198	0,22
$^6F_{5/2}$	12379	808	2.4415	1.6266	0,81
$^6F_{3/2}$	13203	757	0.5222	0.3066	0.22
$^4F_{9/2}$	21115	474	0.5825	0.2724	0.31
$^4I_{15/2}$	22232	450	0.8297	0.9269	0.09
$^4G_{11/2}$	23397	427	0.4531	0.1449	0.31
$^4F_{7/2}$+$^4I_{13/2}$+$^4M_{21/2}$+$^4K_{17/2}$	25800	388	4.2875	3.9676	0.32
$^4M_{19/2}$+(^4P,^4D)$_{3/2}$+$^6P_{5/2}$	27352	366	2.2321	2.5241	0.29
$^4I_{11/2}$	27871	359	0.3143	0.0849	0.23
$^6P_{7/2}$	28458	351	5.8907	5.4283	0.47
(^4M,^4I)$_{15/2}$+(^4F,^4D)$_{5/2}$+$^4I_{9/2}$	29551	338	0.8083	0.4574	0.35

$\delta_{RMS} = 3.76x10^{-7}$

Ω_λ (10^{-20} cm^2):	$\Omega_2 = 25.204$	$\Omega_4 = 3.926$	$\Omega_6 = 4.196$

Table S10. Predicted spontaneous emission probabilities (A_{rad}), branching ratios (β) and lifetimes (τ_{rad}) for radiative transitions of Dy(III) in HYD-Dy.

Transition		λ (nm)	Wavenumber (cm^{-1})	A_{rad} (s^{-1})	β	τ_{rad} (μs)
$^4F_{9/2} \rightarrow$	$^6H_{15/2}$	474	21115	291.66	0.127	435
	$^6H_{13/2}$	592	16879	1619.29	0.706	
	$^6H_{11/2}$	653	15304	201.66	0.088	
	$^6H_{9/2}$+$^6F_{11/2}$	741	13498	97.47	0.042	
	$^6F_{9/2}$+ $^6H_{7/2}$	827	12096	46.88	0.020	
	$^6H_{5/2}$	917	10911	5.60	0.002	
	$^6F_{7/2}$	987	10131	8.10	0.004	
	$^6F_{5/2}$	1145	8736	23.67	0.010	
	$^6F_{3/2}$	1264	7912	0.14	0.000	

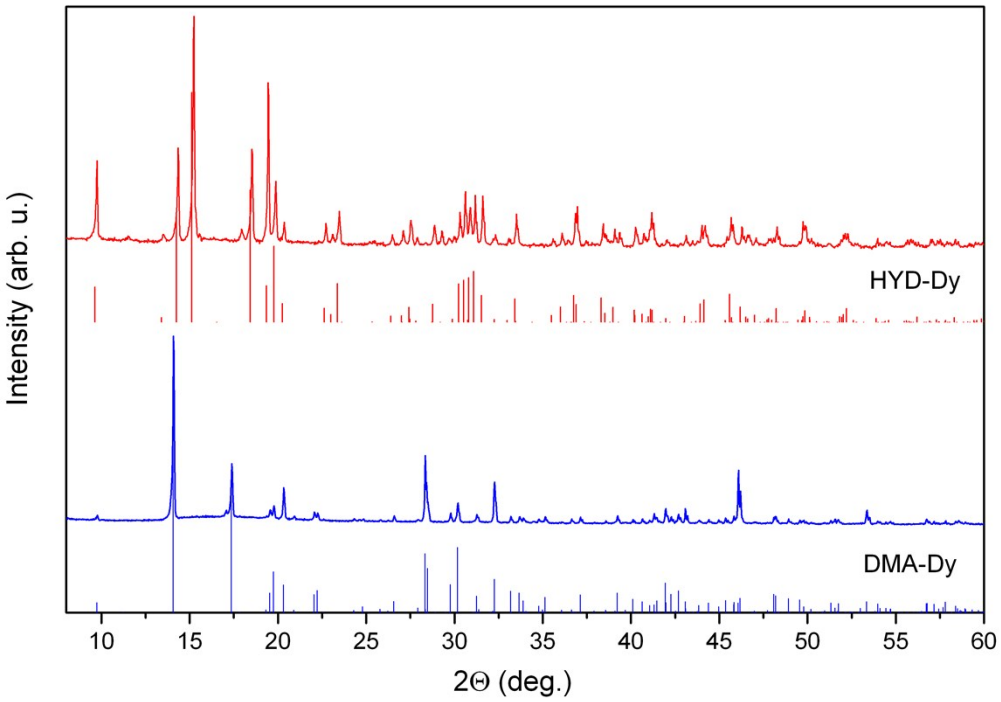

Figure S1. Powder XRD patterns for the as-prepared bulk samples of DMA-Dy and HYD-Dy, with the calculated ones based on the single crystal structures at 293 K.

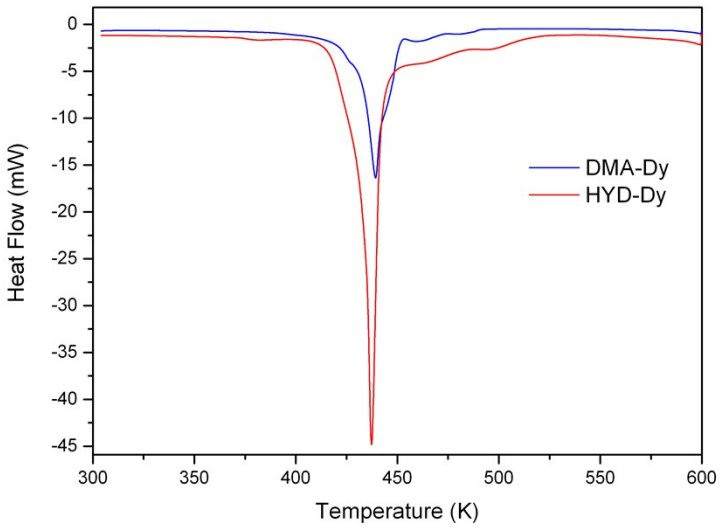

Figure S2. The DSC traces for DMA-Dy (blue line) and HYD-Dy (red line).

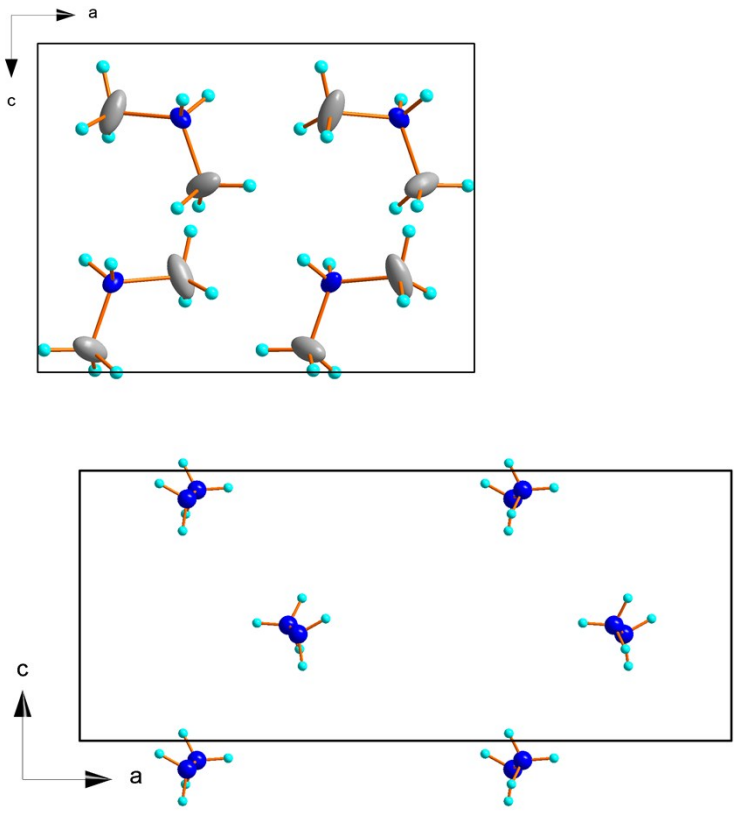

Figure S3. Arrangement of template cations in the strystal structures of DMA-Dy and HYD-Dy. The *c* axis is the polar direction.

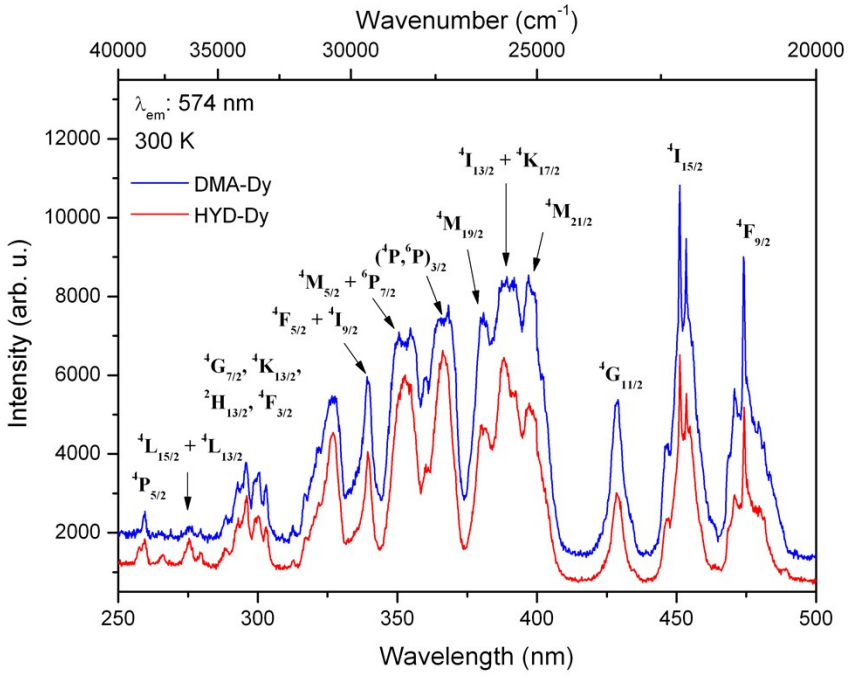

Figure S4. Excitation spectrum of HYD-Dy and DMA-Dy.

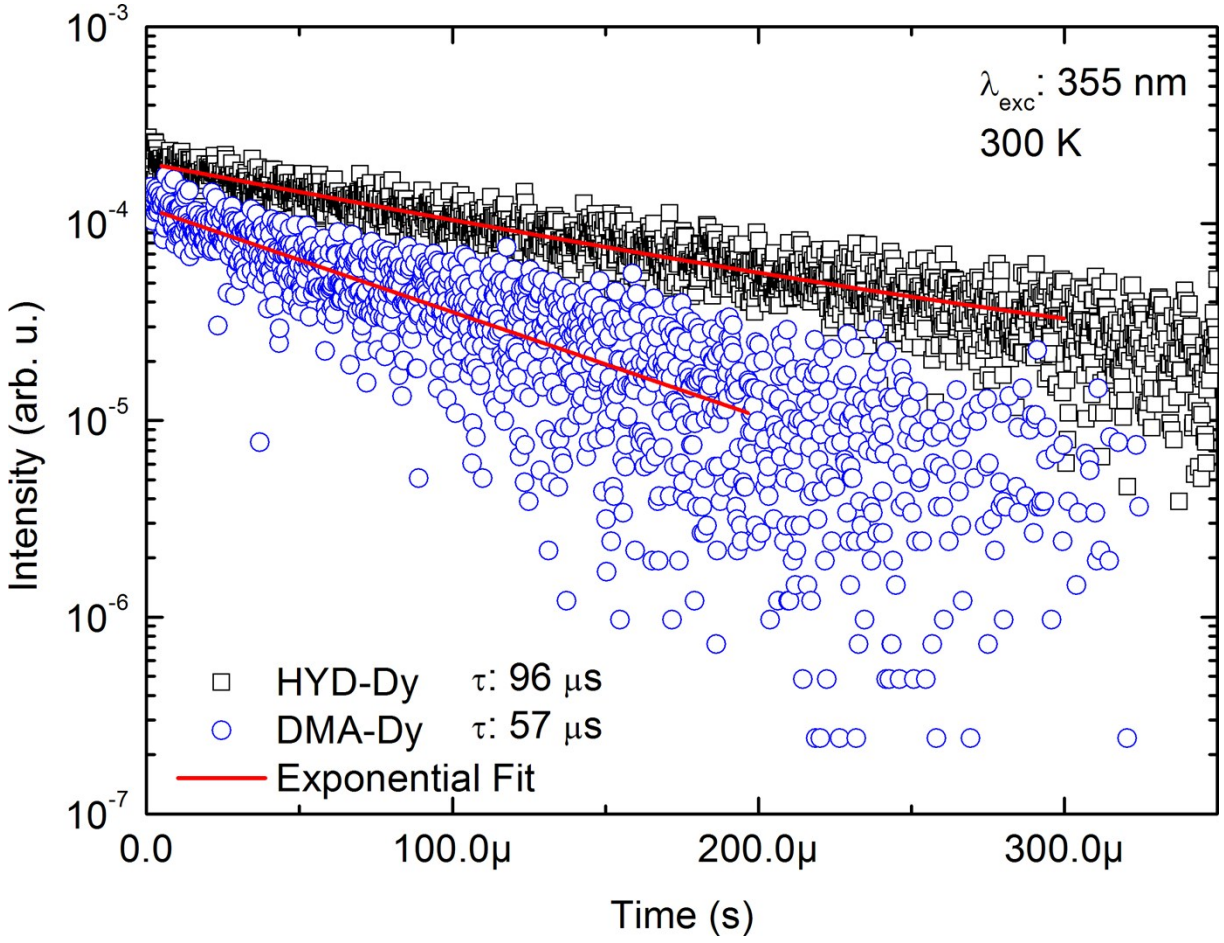

Figure S5. Emission decay curves registered for DMA-Dy and HYD-Dy under 355 nm excitation. Solid lines (red) represent exponential fits.

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