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Naphthalene Amidine Imide Dyes by Transamination of Naphthalene Bisimides

Heinz Langhals* and Harald Jaschke[a]

Abstract: Derivatives of naphthalene-1,4,5,8-tetracarboxylic acid with amidine structures have been prepared. The light absorption of the bisimide derivatives in the UV region is shifted to the visible for the amidine imides, which also fluoresce with a large Stokes shift. It has been shown how the bisimide–lactam rearrangement can be extended to amidine structures.

Keywords: amidines · dyes/pigments · fluorescence spectroscopy · heterocycles · naphthalene · UV/Vis spectroscopy

Introduction

Naphthalene-1,8:4,5-tetracarboxylic bisimides (**1**)[1–4] are preparatively easily accessible from the bisanhydride **2**[5] (Scheme 1) and are formally the lower analogues of the well-known perylene dyes.[6,7] However, the light absorption of the bisimides **1** is so hypsochromically shifted that they are colourless; bisimides **1** are only weakly fluorescent (a florescence quantum yield of 3.5% for **1a** in dichloromethane versus quinine sulfate[8]; see also refs. [9–11]).

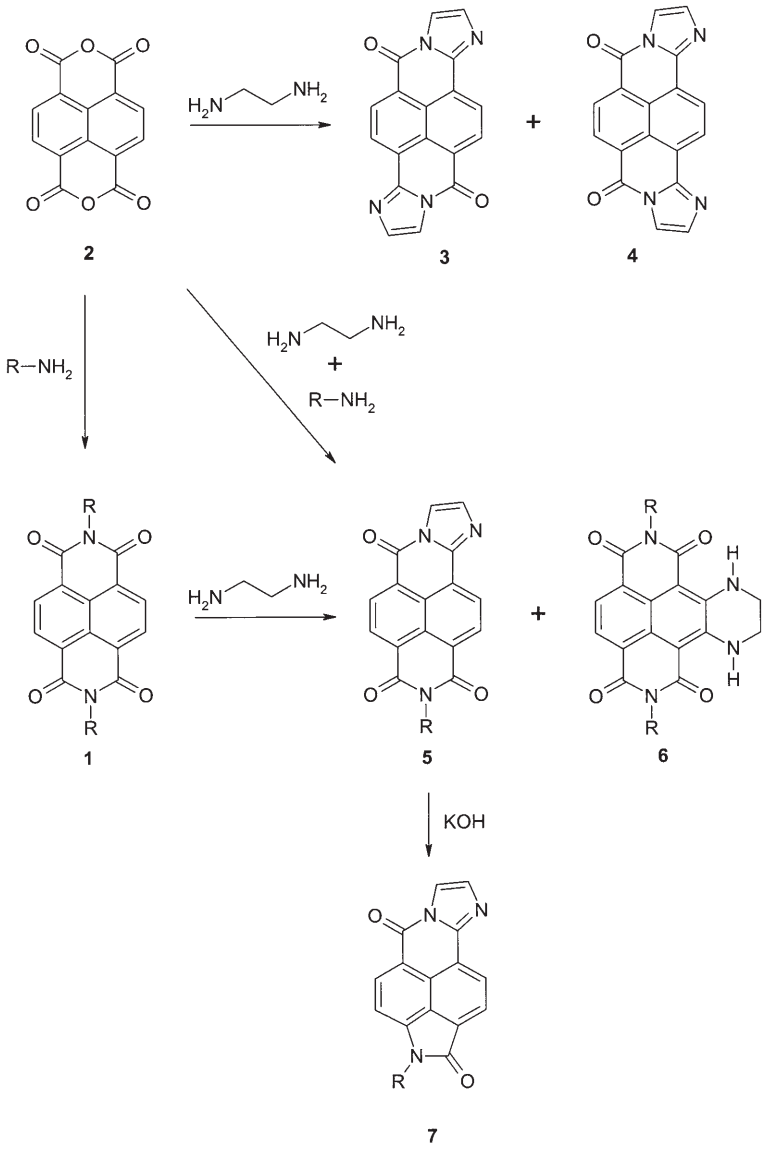
It is of interest to produce a bathochromic shift of the absorption of **1** to increase its applications by exploiting its absorptivity in the visible region. Such a shift has been obtained by ring-contraction of the imide groups[12,13] or by substitution of the aromatic core with donor groups in the 2,6- and 2,7- positions.[14–21] Alternatively, one of the carbonyl groups could be replaced by an imino group; such an arrangement causes a pronounced bathochromic shift in the perylene dyes.[22–24] Anhydride imides are suitable starting materials for the preparation of iminoimidic perylene dyes, however, the synthesis of the naphthalene anhydride imide analogues has proved difficult.

Results and Discussion

The condensation of ethylenediamine with the bisanhydride **2** led to the isomeric bisamidines **3** and **4**, characterised by two imidazole moieties, instead of the formation of **1** with two imino groups in the molecular framework (see Scheme 1). Ethylenediamine seems to act as an oxidant, as has already been found in a similar reaction with perylene dyes;[25] isomers **3** and **4** are the lower debenzocondensed homologues of Indanthrene Scarlet and the two pure isomers Indanthrene Brilliant Red (Vat Orange 7, C.I. 71105) and Indanthrene Bordaux (Vat Orange 15, C.I. 71100), respectively. Isomers **3** and **4** are sufficiently soluble to be used in homogeneous solution and can be separated by column chromatography. We simultaneously condensed the bis-anhydride **2** with a primary amine ($R-NH_2$) and ethylenediamine to prepare **5** in which one of the carbonyl groups of **1** has been exchanged for an imino group; compound **5** was separated from the reaction products by column chromatography (Scheme 1).

The co-condensation of the two amines is an acceptable procedure with aliphatic amines, however, it is not practicable for the less reactive aromatic amines because the competing condensation to **3** and **4** becomes dominant. Thus, an alternative route is required for the preparation of naphthalene iminoimides **5**. Perylene-3,4:9,10-tetracarboxylic bisimides are the larger analogues of **1**. They are very stable with respect to attack by primary amines[26] and so they are not suitable for exchange reactions and even their hydrolysis[27] requires severe reaction conditions. Remarkably, the lower naphthalene bisimide analogues (**1**) proved to be much more labile towards the exchange reactions of the R–N units. Thus, we first prepared naphthalene bisimides **1** by the condensation of **2** with aromatic amines. The structure of **1s** was established by X-ray crystallography; the orientation of the *tert*-butyl groups in **1s** was unambiguously determined, although the final refinement of the structure analysis was unsuccessful, see Figure 1.

[a] Prof. Dr. H. Langhals, Dr. H. Jaschke
Department of Chemistry, LMU University of Munich
Butenandtstrasse 13, 81377 Munich (Germany)
Fax: (+49) 2180-77640
E-mail: Langhals@lrz.uni-muenchen.de



Scheme 1. Synthesis of naphthalene amidine derivatives. **1**, **5**–**7**: **a**: R = $CH(CH_3)C_2H_5$; **b**: R = $CH(CH_3)C_3H_7$; **c**: R = $CH(C_6H_{13})_2$; **d**: *cyclo*-C_6H_{11}; **e**: CH_2CH_2OH; **f**: $CH_2CHOHCH_3$; **g**: $CH_2C(CH_3)(C_3H_7)_2$; **h**: $CH_2C(CH_3)_2C_5H_{11}$; **i**: $CH_2C(C_6H_{13})_3$; **j**: CH_2(1-C_2H_5-*cyclo*-C_6H_{10}); **k**: CH_2(1-C_3H_7-*cyclo*-C_6H_{10}); **l**: $CH_2C(C_2H_5)_2C_6H_5$; **m**: $CH_2C(C_4H_9)_2C_6H_5$; **n**: 2,3-$(CH_3)C_6H_3$; **o**: 2,5-$(CH_3)C_6H_3$; **p**: 5-*t*Bu-2-$CH_3C_6H_3$; **q**: 2-*t*Bu-C_6H_4; **r**: 4-*t*Bu-C_6H_4; **s**: 2,5-$(t\text{Bu})_2$-C_6H_3.

Naphthalene iminoimides **5** with aromatic substituents R were successfully synthesised by the reaction of **1** with ethylenediamine; compare also the reaction of the even more reactive alkylated phthalimide imides (five-membered ring imides) with ethylenediamine in Gabriel's synthesis.[28] Surprisingly, not only the amidines **5** were obtained, but also the byproducts **6** in which two of the hydrogen atoms of the core were replaced by an ethylenediamine bridge; naphthalene derivatives with donor groups in the 2- and 3-positions are unknown. The formation of **6** also indicates a formal oxidation of **1** and corresponds to the reaction that leads to the formation of **3** and **4** from **2**.

The amidines **5** contain one complete carboxylic imide substructure. On the opposite side of the naphthalene core one remaining carbonyl group is conjugated to the amidine structure. Therefore, we tested whether the novel carboxylic bisimide–amidine rearrangement[12] could also be carried out with **6** in spite of its structural differences. The rearrangement proceeded as a vinylogous benzylic acid rearrangement, which has also been verified by high-level quantum mechanical calculations.[29] Thus, the involvement of the two conjugating carbonyl groups is expected to be preferred determining the regioselectivity of the reaction. Indeed, the treatment of **5r** with alcoholic alkali gave the amidine lactam **7r**. The regioselectivity of the ring contraction was determined by X-ray crystallography; the orientation of the carbonyl groups could be unambiguously determined, although the final refinement of the structure analysis of **7r** was unsuccessful. The formation of **7r** indicates that the novel vinylogous benzylic acid rearrangement is not limited to tetracarboxylic bisimides, but is a more general reaction.

The bisimdes **1** form as colourless crystals, although some derivatives are reported to exhibit a rose-to-red colour. This is a consequence of the formation of the corresponding (difficult-to-remove) perylene bisimides as byproducts, which have strong absorptivities of some $10^5\,L\,mol^{-1}\,cm^{-1}$. These are preferentially formed if the condensation of amine with **2** is carried out in DMF[30] and suppressed if carried out in acetic acid.[31] Dissolved **1** absorbs in the UV region and the structure of the spectra are similar to those of the perylene dyes (Figure 2). A Gaussian analysis (Table 1) of the spectrum of **1a** (R value = 1.7%) indicates a vibronic pattern similar to those of the perylene dyes.[32] The fluorescence of **1** is very weak.[10] According to Adachi et al., two overlapping electronic levels are responsible for the fluorescence deactivation.[33]

The replacement of two carbonyl groups in **1** by imino groups causes a bathochromic shift of the absorption into the visible region (Figure 3). The absorption of the red *cis* isomer **4** (*syn*) is more bathochromically shifted than the

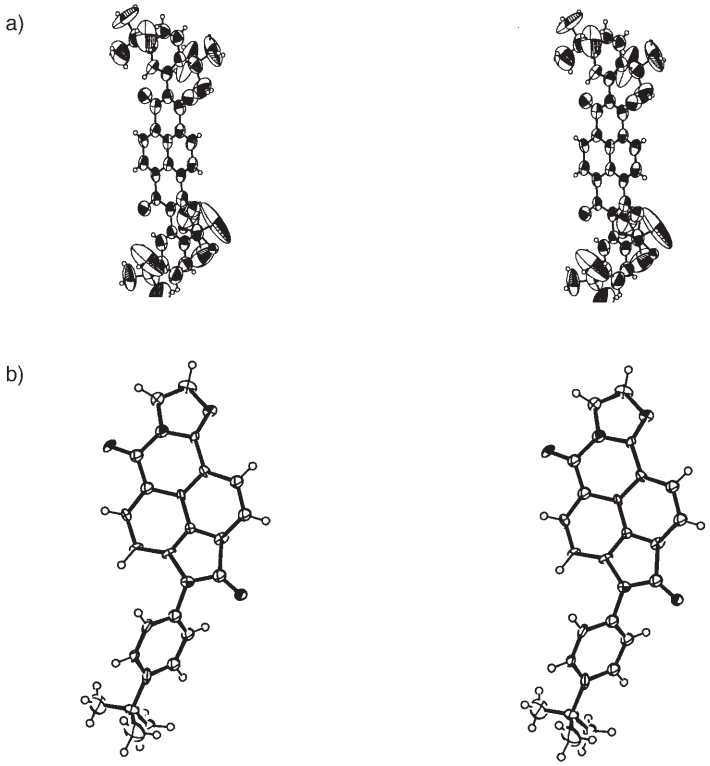


Figure 1. Stereo plots of the X-ray crystal structures of a) **1s** and b) **7r**.

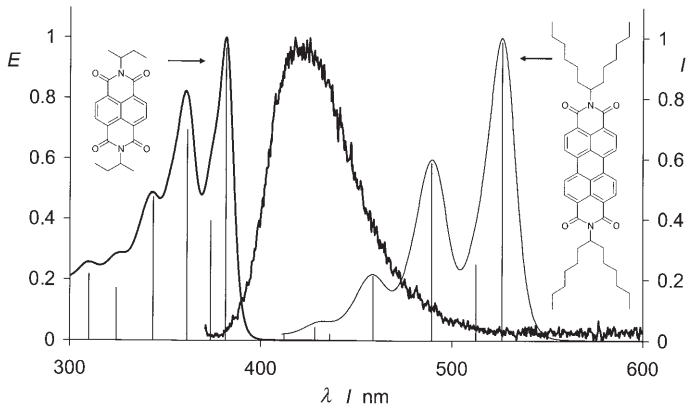


Figure 2. UV/Vis absorption spectra of **1a** (thick line, left) in chloroform compared with the perylene dyes (thin line, right). The vertical bars indicate the positions and intensities of the vibration bands according to Gaussian analysis. Thick line in the middle: fluorescence spectrum of **1a**.

Table 1. Gaussian analysis of the UV/Vis spectrum of **1a**.

Nr.	λ_{max} [nm]	$2\sigma^2$ [10^4 cm^{-2}]	E_{max}
1	381.6	0.147	0.964
2	373.7	0.096	0.394
3	361.4	0.427	0.695
4	343.7	1.37	0.472
5	324.3	0.572	0.172
6	310.1	1.309	0.218
7	296.5	0.579	0.063
8	290.0	1.559	0.066
9	275.1	17.87	0.074

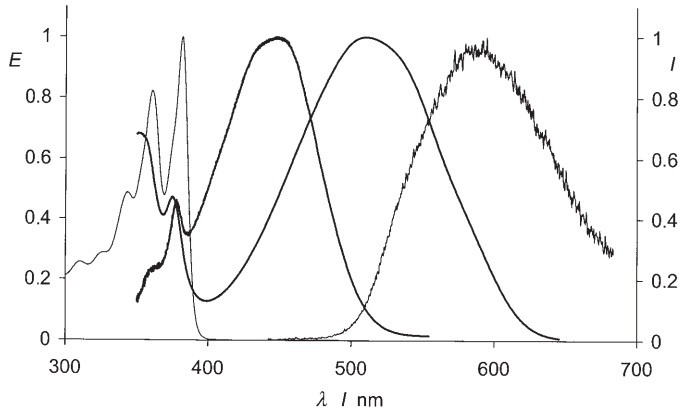


Figure 3. UV/Vis absorption spectra of naphthalene derivatives in chloroform. From left to right: **1a**, **3** and **4** and the fluorescence spectrum of **3**.

yellow *trans* isomer **3** (*anti*). The different absorption spectra for these isomers correspond to the different colours of the benzo analogues of **3** and **4**, respectively, the dyes Vat Orange 7 (C.I. 71105) and Vat Orange 15 (C.I. 71100).[30,34–36] Compound **3** fluoresces with a quantum yield of 40% and the Stokes shift is remarkable, whereas no fluorescence was detected for the more bathochromically absorbing **4**. This is similar to the well-known different fluorescence properties of the isomeric bimanes,[37–39] although in this case the *syn* isomer is fluorescent and the *anti* isomer is not.

The replacement of a single carbonyl group of **1** by an imino group is sufficient to account for most of the bathochromic shift of **3** and so **5** also absorbs in the visible region (Figure 4). Compound **4** is fluorescent with a quantum yield of 20% and its Stokes shift is large. The substituent R in **5** is useful for linking with substrates and so fluorescent labelling is possible. The absorption of the byproducts **6** is shifted far into the visible. The UV/Vis spectrum of **6** is strongly bathochromically shifted (500 nm) compared with **1** and struc-

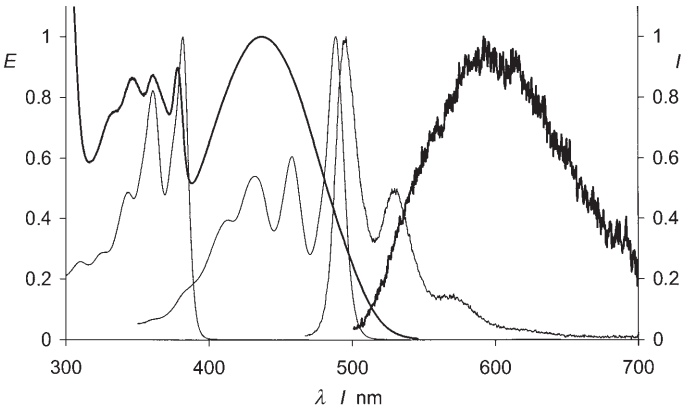


Figure 4. UV/Vis absorption spectra of naphthalene derivatives in chloroform. Absorption and fluorescence spectra of **5c** (thick lines) and **6a** (thin lines in the centre). The absorption spectrum of **1a** (thin line left) is shown for comparison.

tured in contrast to the other naphthalene derivatives **3**–**5**. The fluorescence of **5** (quantum yield of 6%) is also structured and the Stokes shift is small compared with the other naphthalene derivatives. The spectra of **5** indicate that the novel substitution pattern causes a bathochromic shift similar to that of the well-known donor disubstitution at the 2- and 6-positions.

The ring contraction that occurs in the formation of **7** from **5** causes a hypsochromic shift of the absorption: $\lambda_{max}=$ 424 nm (Figure 5). The materials are moderately fluorescent ($\Phi=12\%$) and the Stokes shift remains high.

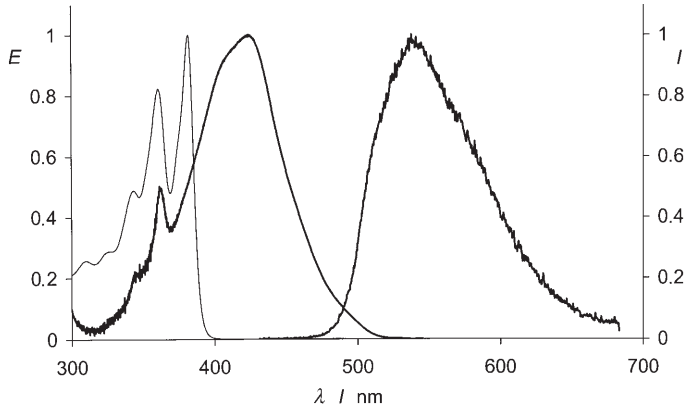


Figure 5. UV/Vis absorption and fluorescence spectra of **7a** in chloroform (thick lines). The absorption spectrum of **1a** (thin line left) is shown for comparison.

Experimental Section

2,7-Bis(1-methylpropyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (1a): Naphthalenetetracarboxylic bisanhydride (**2**, 2.75 g, 10.0 mmol) was dispersed in *N*,*N*-dimethylformamide (150 mL). 2-Aminobutane (2.24 g, 30.0 mmol) was added dropwise with stirring and the mixture was heated at 170 °C for 24 h, cooled to room temperature, poured into cooled 2 N HCl (400 mL, 0 °C) and allowed to stand for 1 h. The fine, beige precipitate was collected by vacuum filtration through a D4 glass filter, dried (8 h, 110 °C), treated with a saturated sodium carbonate solution (400 mL), collected by vacuum filtration, washed with distilled water, dried again (8 h, 110 °C), dissolved three times in ethanol (100 mL), filtered and evaporated to remove oily byproducts (yield 3.82 g). The material exhibits a weak rose coloration as a consequence of traces of *N*,*N*′-bis(1-methylpropyl)perylene-3,4:9,10-tetracaboxylic bisimide as the only detectable byproduct. These were removed by column chromatography (silica gel, chloroform/acetic acid 39:1); chromatographic separation with toluene gave similar results. Analytically pure material was obtained by a subsequent extractive crystallisation[40] from ethanol. Yield 3.82 g (99%), small, faint yellowish needles, m.p. 192 °C. R_f ($CHC1_3$) = 0.4. R_f (silica gel, $CHC1_3$/acetic acid 10:1) = 0.8. IR (KBr): $\tilde{\nu}=$2967 (s), 2935 (m), 2877 (m), 1703 (s), 1664 (s), 1582 (s), 1454 (s), 1397 (s), 1380 (m), 1331 (s), 1279 (m), 1250 (s), 1216 (m), 1172 (w), 1080 (m), 784 (m), 771 (s), 735 (m), 590 (w) cm^{-1}. ^1H NMR (400 MHz, $CDC1_3$): $\delta=$0.89 (t, 6H, $2CH_3$), 1.56 (d, 6H, $2CH_3$), 1.93 (m, 2H), 2.18 (m, 2H), 5.17 (sextet, 2H, 2CH), 8.70 (s, 4H, aromatic H) ppm. ^{13}C NMR ($CDC1_3$): $\delta=$11.41, 17.96, 26.40, 51.90, 126.66, 126.79, 130.83, 163.33 ppm. UV/Vis ($CHC1_3$): λ_{max} (ε) = 381 (47910), 360 (39040), 343 (22930), 242 (50560 $dm^3mol^{-1}cm^{-1}$) nm. MS (70 eV): *m*/*z* (%): 378 (62) [M^+], 349 (32) [$M^+-C_2H_5$], 323 (100) [$M^+-C_4H_7$], 293 (13), 267 (24), 249 (29), 221 (6), 193 (3). Elemental analysis calcd (%) for $C_{22}H_{22}N_2O_4$ (378.4): C 69.83, H 5.86, N 7.40; found: C 69.97, H 5.93, N 7.07.

2,7-Bis(2-methyl-2-propylpentyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (1g): Naphthalenetetracarboxylic bisanhydride (**2**, 2.75 g, 10.0 mmol), 2-methyl-2-propylpentylamine (4.38 g, 30.6 mmol) and *N*,*N*-dimethylformamide (150 mL) were refluxed for 24 h, cooled to 0 °C, treated dropwise with 2 N HCl (400 mL, 0 °C, fine, beige precipitate), stirred for 1 h, collected by vacuum filtration (D4 glass filter, 4.90 g, 92%, colourless material) and purified by column chromatography (silica gel, chloroform/acetic acid 39:1). Yield 4.7 g (88%), colourless powder, m.p. 225 °C. R_f (silica gel, $CHCl_3$) = 0.29. IR (KBr): $\tilde{\nu}=$3450 (w), 3090 (w), 2960 (s), 2932 (s), 2871 (m), 1708 (s), 1668 (s), 1651 (m), 1582 (s), 1467 (m), 1454 (s), 1426 (w), 1380 (s), 1329 (s), 1263 (w), 1246 (s), 1224 (w), 1164 (m), 1115 (w), 1077 (w), 1070 (w), 1029 (w), 988 (m), 778 (w), 717 (w) cm^{-1}. ^1H NMR (300 MHz, $CDC1_3$): $\delta=$8.72 (s, 4H, naphthalene), 4.17 (s, 4H, 2N-CH_2), 1.41–1.19 (m, 16H, $8CH_2$), 0.88 (t, 12H, $4CH_3$), 0.85 (s, 6H, $2CH_3$) ppm. ^{13}C NMR ($CDC1_3$): $\delta=$163.74, 131.02, 126.69, 126.56, 47.90, 40.68, 38.98, 24.03, 16.89, 15.02 ppm. UV/Vis ($CHC1_3$): λ_{max} (ε) = 381 (27660), 360 (23170), 342 (14650), 327 (9690 $dm^3mol^{-1}cm^{-1}$) nm. MS (70 eV): *m*/*z* (%): 518 (46) [M^+], 475 (10), 406 (100), 294 (26), 280 (11), 249 (9), 112 (12), 71 (25), 57 (23), 55 (10). Elemental analysis calcd (%) for $C_{32}H_{42}N_2O_4$ (518.7): C 74.10, H 8.16, N 5.40; found: C 74.16, H 8.04, N 5.58.

2,7-Bis(2,2-dimethylheptyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (1h): Naphthalenetetracarboxylic bisanhydride (**2**, 2.75 g, 10.0 mmol), 2,2-dimethylheptylamine (4.38 g, 30.0 mmol) and acetic acid (60 mL) were refluxed for 2.5 h, cooled, treated with 2 N HCl (400 mL), stirred for 1 h (fine, beige precipitate), collected by vacuum filtration (D4 glass filter), dried in air (110 °C, 8 h, 2.4 g, 45%), dissolved in hot chloroform (removing 150 mg of an insoluble byproduct), treated dropwise with ethanol until crystallisation, left to stand for 16 h and collected by vacuum filtration (further material can be obtained by evaporation of the mother liquor) and purified by column chromatography (silica gel, chloroform). Yield 1.90 g (36%), colourless powder, m.p. 165 °C. R_f (silica gel, $CHCl_3$) = 0.59. R_f (silica gel, $CHCl_3$/acetic acid 9.75:0.25) = 0.61. IR (KBr): $\tilde{\nu}=$3456 (m), 2957 (s), 2933 (s), 2872 (m), 2860 (m), 1705 (s), 1666 (s), 1582 (s), 1467 (m), 1453 (s), 1425 (w), 1392 (m), 1373 (s), 1330 (s), 1275 (w), 1246 (s), 1221 (m), 1167 (m), 1114 (w), 1097 (w), 1075 (w), 1023 (w), 980 (w), 880 (w), 875 (w), 807 (w), 775 (s), 770 (m), 716 (m), 645 (w), 564 (w) cm^{-1}. ^1H NMR (300 MHz, $CDC1_3$): $\delta=$8.74 (s, 4H, aromatic), 4.17 (s, 4H, 2N-CH_2), 1.37 (m, 16H, $8CH_2$), 0.94 (s, 12H, $4CH_3$), 0.90 (t, 6H, $2CH_3$) ppm. ^{13}C NMR ($CDC1_3$): $\delta=$163.6, 131.05, 126.68, 126.57, 49.26, 41.56, 36.69, 32.83, 26.03, 23.70, 22.70, 14.12 ppm. UV/Vis ($CHC1_3$): λ_{max} (ε) = 381 (17160), 361 (16940), 344 (12100), 242 (23280), 327 (7180 $dm^3mol^{-1}cm^{-1}$) nm. MS (70 eV): *m*/*z* (%) 518 (99.9) [M^+], 503 (3.4) [M^+-CH_3], 447 (4.4) [$M^+-C_5H_{11}$], 406 (100) [$M^+-C_8H_{16}$], 393 (5.7) [$M^+-C_9H_{19}$], 294 (22.3), 280 (6.0), 113 (5.4), 112 (5.8), 71 (17.5) [$C_5H_{11}^+$], 69 (6.0), 57 (21.9), 43 (7.9). Elemental analysis calcd (%) for $C_{32}H_{42}N_2O_4$ (518.7): C 74.10, H 8.16, N 5.40; found: C 74.26, H 8.36, N 5.30.

2,7-Bis(2,2-dihexyloctyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (1i): Naphthalenetetracarboxylic bisanhydride (**2**, 2.75 g, 10.0 mmol), 2,2-dihexyloctylamine (9.11 g, 30.0 mmol) and *N*,*N*-dimethylformamide (150 mL) were refluxed for 24 h, cooled for 16 h, treated at 0 °C with 2 N HCl (400 mL), collected by vacuum filtration, treated with boiling 10% K_2CO_3 (400 mL, melting the reaction product), cooled to 0 °C, collected by vacuum filtration (D4 glass filter), dried in vacuo with calcium chloride (8 h), dissolved in chloroform to remove an insoluble byproduct and purified by column chromatography (silica gel, chloroform). Yield 6.4 g (76%), colourless powder, m.p. 77 °C. R_f (silica gel, $CHCl_3$) = 0.95. R_f (silica gel, $CHCl_3$/acetic acid 9.75:0.25) = 0.90. IR (KBr): $\tilde{\nu}=$3449 (m), 2955 (s), 2930 (s), 2857 (s), 2362 (w), 2340 (w), 1707 (s), 1664 (s), 1634 (m), 1581 (s), 1575 (m), 1467 (m), 1455 (s), 1378 (m), 1327 (s), 1244 (s), 1221 (w), 1185 (w), 1105 (w), 1075 (w), 985 (w), 887 (w), 774 (m), 717 (w) cm^{-1}. ^1H NMR (300 MHz, $CDC1_3$): $\delta=$0.80–0.90 (t, 18H, $6CH_3$), 1.22–1.35 (m, 52H, $26CH_2$), 4.11 (s, 1H, N-CH), 4.13 (s, 1H, N-CH), 4.20 (s, 2H, N-CH_2), 8.75 (m, 4H, naphthalene) ppm. UV/Vis ($CHC1_3$): λ_{max} (ε) = 381 (29150), 361 (24440), 342 (14810), 327 (9550), 242 (31670 $dm^3mol^{-1}cm^{-1}$) nm. MS (70 eV): *m*/*z* (%) 826 (9.6) [M^+], 745

(18), 743 (29) [$M^+-C_6H_{12}$], 742 (18) [$M^+-C_6H_{13}$], 741 (22) [$M^+-C_6H_{14}$], 659 (9), 657 (11), 548 (11), 547 (24), 475 (11), 463 (40), 268 (63), 249 (25), 197 (13), 154 (50), 136 (52), 89 (57), 77 (100), 69 (64), 57 (60), 55 (91), 42 (81), 41 (84.0). Elemental analysis calcd (%) for $C_{54}H_{86}N_2O_4$ (827.3): C 78.40, H 10.48, N 3.39; found: C 77.79, H 10.18, N 3.61.

2,7-Bis(2-ethylcyclohexylmethyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (1j): Naphthalenetetracarboxylic bisanhydride (**2**, 2.75 g, 10.0 mmol), 2-ethyl-1-aminomethylcyclohexane (4.33 g, 30.0 mmol) and acetic acid (30 mL) were allowed to react as described above for **1i** and purified by column chromatography (silica gel, chloroform/acetic acid 39:1). Yield 550 mg (10.5%), m.p. 256°C. R_f (silica gel, $CHCl_3$) = 0.5. R_f (silica gel, $CHCl_3$/acetic acid 10:1) = 0.87. IR (KBr): $\tilde{\nu}$ = 2929 (m), 2860 (w), 1710 (s), 1668 (s), 1636 (w), 1582 (m), 1454 (m), 1424 (w), 1374 (m), 1351 (w), 1326 (s), 1298 (w), 1280 (w), 1248 (s), 1221 (w), 1171 (w), 989 (w), 772 (m), 716 (w) cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.74 (s, 4H, aromatic), 4.19 (s, 4H, 2N-CH_2), 1.56–1.36 (m, 16H, 8CH_2), 1.25–1.10 (m, 8H, 4CH_2), 0.96 (t, 6H, 2CH_3) ppm. ^{13}C NMR ($CDCl_3$): δ = 163.87, 131.05, 126.68, 126.56, 47.99, 38.78, 33.49, 26.29, 25.86, 21.54, 7.70 ppm. UV/Vis ($CHCl_3$): λ_{max} (ε) = 382 (22580), 361 (19090), 343 (11960), 326 (7780), 241 (24380 $dm^3mol^{-1}cm^{-1}$) nm. MS (70 eV): *m/z* (%) 514 (73) [M^+], 485 (4.3) [$M^+-C_2H_3$], 404 (100) [$M^+-C_8H_{14}$], 391 (8), 294 (31), 281 (11), 280 (10), 263 (5), 249 (6), 111 (36), 110 (22), 81 (17), 69 (39), 55 (12). Elemental analysis calcd (%) for $C_{32}H_{38}N_2O_4$ (514.7): C 74.68, H 7.44, N 5.44; found: C 74.82, H 7.32, N 5.59.

2,7-Bis(2-propylcyclohexylmethyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (1k): Naphthalenetetracarboxylic bisanhydride (**2**, 2.75 g, 10.0 mmol), 2-propyl-1-aminomethylcyclohexane (4.75 g, 30.0 mmol) and acetic acid (30 mL) were allowed to react as described above for **1i** and purified by column chromatography (silica gel, chloroform) and recrystallisation from chloroform/ethanol. Yield 0.78 g (15%), colourless, small leaflets, m.p. 227°C. R_f ($CHCl_3$) = 0.80. IR (KBr): $\tilde{\nu}$ = 3441 (m), 2931 (s), 2867 (s), 1710 (s), 1667 (s), 1634 (w), 1582 (s), 1454 (s), 1435 (m), 1423 (w), 1378 (m), 1326 (s), 1287 (w), 1245 (s), 1221 (m), 1171 (m), 1155 (w), 1142 (w), 1118 (w), 1095 (w), 1065 (w), 1016 (w), 993 (w), 772 (s) cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.72 (s, 4H, naphthalene), 4.40 (s, 4H, 2N-CH_2), 1.50–1.23 (m, 20H, 10CH_2), 1.24–1.10 (m, 8H, 4CH_2), 0.91 (t, 6H, 2CH_3) ppm. ^{13}C NMR ($CDCl_3$): δ = 163.84, 131.04, 126.70, 126.57, 48.43, 38.97, 36.39, 33.99, 25.86, 21.59, 16.47, 15.13 ppm. MS (70 eV): *m/z* (%) 542 (83) [M^+], 499 (3) [$M^+-C_3H_7$], 418 (100) [$M^+-C_9H_{16}$], 405 (6) [$M^+-C_{10}H_{17}$], 294 (20) [418–C_9H_{16}]. Elemental analysis calcd (%) for $C_{34}H_{42}N_2O_4$ (542.7): C 75.24, H 7.80, N 5.20; found: C 74.78, H 7.52, N 5.22.

2,7-Bis(2-ethyl-2-phenylbutyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (1l): Naphthalenetetracarboxylic bisanhydride (**2**, 2.75 g, 10.0 mmol), 2-ethyl-2-phenylbutylamine (4.20 g, 20.0 mmol) and acetic acid (60 mL) were allowed to react as described above for **1i**, and finally extracted with ethanol. Yield 80 mg (1.3%), m.p. 220°C. R_f (silica gel, $CHCl_3$) = 0.38. R_f (silica gel, $CHCl_3$/acetic acid 9.75:0.25) = 0.57. IR (KBr): $\tilde{\nu}$ = 3447 (m), 2968 (m), 2950 (w), 2882 (w), 1709 (s), 1669 (s), 1651 (m), 1582 (m), 1499 (w), 1454 (m), 1372 (w), 1326 (s), 1247 (s), 1166 (w), 1100 (w), 1083 (w), 1025 (w), 1004 (w), 770 (w), 759 (m), 701 (m) cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.44 (s, 4H, naphthalene), 7.26–7.19 and 7.11–7.00 (2×m, 10H, 2phenyl), 4.37 (s, 4H, 2N-CH_2), 2.13–1.85 (m, 20H, 4C_2H_5) ppm. UV/Vis ($CHCl_3$): λ_{max} (ε) = 381 (19800), 361 (19660), 345 (13740), 326 (82230), 240 (27990 $dm^3mol^{-1}cm^{-1}$) nm. MS (70 eV): *m/z* (%) 586 (19) [M^+], 557 (3) [M^+–ethyl], 440 (13) [$M^+-Ph(C_2H_5)C{=}CH\text{-}CH_3$], 427 (10), 160 (48), 147 (100), 146 (26), 105 (87), 91 (76). Elemental analysis calcd (%) for $C_{38}H_{38}N_2O_4$ (586.7): C 77.79, H 6.53, N 4.77; found: C 77.27, H 6.30, N 4.71.

2,7-Bis(2-butyl-2-phenylhexyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (1m): Naphthalenetetracarboxylic bisanhydride (**2**, 2.75 g, 10.0 mmol), 2-butyl-2-phenylhexylamine (7.14 g, 30.0 mmol) and *N,N*-dimethylformamide (150 mL) were allowed to react as described above for **1a**. Yield 6.0 g (84%), fine, colourless needles, m.p. 162°C. R_f (silica gel, $CHCl_3$) = 0.68. R_f (silica gel, $CHCl_3$/acetic acid 39:1) = 0.66. IR (KBr): $\tilde{\nu}$ = 2957 (s), 2935 (m), 2871 (m), 1710 (s), 1671 (s), 1602 (w), 1582 (m), 1499 (w), 1454 (m), 1420 (w), 1378 (m), 1326 (s), 1247 (s), 1222 (w), 1161 (w), 1009 (w), 763 (m), 698 (m), 582 (w) cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.44 (s, 4H, naphthalene), 7.25–7.17 and 7.09–7.00 (2m, 10H, 2phenyl), 4.37 (s, 4H, 2N-CH_2), 1.84, 1.40–1.31 and 1.20–1.116 (3m, 24H, 12CH_2), 0.91 (t, 12H, 4CH_3) ppm. ^{13}C NMR ($CDCl_3$): δ = 163.03, 144.35, 130.58, 127.72, 127.05, 126.41, 126.15, 125.86, 48.48, 45.70, 34.75, 26.04, 23.57, 14.14 ppm. UV/Vis ($CHCl_3$): λ_{max} (ε) = 381 (19690), 361 (19470), 343 (13760), 241 (31090), 327 (7180 $dm^3mol^{-1}cm^{-1}$) nm. MS (70 eV): *m/z* (%) 698 (3) [M^+], 496 (6) [M^+–202], 483 (3), 280 (3), 249 (3), 216 (45), 203 (100), 147 (43), 133 (32), 119 (28), 118 (13), 117 (14), 115 (6). Elemental analysis calcd (%) for $C_{46}H_{54}N_2O_4$ (698.9): C 79.05, H 7.79, N 4.01; found: C 78.76, H 7.75, N 4.07.

2,7-Bis(2,3-dimethylphenyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (1n): Naphthalenetetracarboxylic bisanhydride (**2**, 1 g, 4 mmol), 2,3-dimethylaniline (1.37 g, 10.0 mmol) and acetic acid (20 mL) were allowed to react as described above for **1i**. Yield 1.10 g (61%), fine, yellow needles, m.p. >350°C. R_f (silica gel, $CHCl_3$) = 0.25. R_f (silica gel, $CHCl_3$/acetic acid 39:1) = 0.57. IR (KBr): $\tilde{\nu}$ = 2980 (w), 2924 (w), 2861 (w), 1719 (s), 1684 (s), 1585 (s), 1476 (m), 1455 (m), 1350 (s), 1255 (s), 1220 (m), 1205 (s), 1161 (w), 1125 (w), 1096 (w), 1030 (w), 988 (m), 904 (w), 885 (w), 830 (w), 792 (m), 778 (s), 750 (s), 715 (m), 611 (w) cm^{-1}. ^1H NMR (80 MHz, $CDCl_3$): δ = 9.84 ppm (s, 4H, naphthalene), 8.25 (m, 6H, 2phenyl), 2.41 (s, 6H, 2CH_3), 2.10 (s, 6H, 2CH_3) ppm. UV/Vis ($CHCl_3$): λ_{max} (ε) = 380 (26560), 360 (23940), 342 (14780), 327 (7430), 242 (30300) nm. MS (70 eV): *m/z* (%) 474 (87) [M^+], 459 (38) [M^+-CH_3], 457 (100), 441 (6), 425 (7). Elemental analysis calcd (%) for $C_{30}H_{22}N_2O_4$ (474.5): C 75.94, H 4.67, N 5.90; found: C 75.61, H 4.63, N 5.76.

2,7-Bis(2,5-dimethylphenyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (1o): Naphthalenetetracarboxylic bisanhydride (**2**, 2.75 g, 10.3 mmol), 2,5-dimethylaniline (4.97 g, 41.0 mmol) and acetic acid (100 mL) were allowed to react as described above for **1i** and purified by extractive recrystallisation[40] from ethanol. Yield 2.54 g (52%), m.p. >350°C. R_f (silica gel, $CHCl_3$/acetone 15:1) = 0.49. IR (KBr): $\tilde{\nu}$ = 3435 (m, br), 2924 (m), 1715 (s), 1679 (s), 1582 (m), 1509 (m), 1448 (m), 1344 (s), 1251 (s), 1199 (m), 1148 (w), 1120 (w), 982 (w), 930 (w), 886 (w), 818 (w), 772 (m), 754 (m) cm^{-1}. ^1H NMR (80 MHz, $CDCl_3$): δ = 8.86 (s, 4H, naphthalene), 7.33 (d, J = 7.8 Hz, 2H, phenyl), 7.20 (d, J = 7.6 Hz, 2H, phenyl), 7.04 (s, 2H, phenyl), 2.40 (s, 6H, 2CH_3), 2.15 (s, 6H, 2CH_3) ppm. ^{13}C NMR ($CDCl_3$): δ = 166.6 (4C=O), 144.3 (2C-phenyl), 137.0, 134.3, 132.6, 132.5, 130.9, 130.8, 130.0, 128.8, 128.7, 125.3, 123.4, 123.1 (10C-phenyl and 10C-naphthalene), 20.9 (2CH_3), 17.1 (2CH_3) ppm. UV/Vis ($CHCl_3$): λ_{max} (ε) = 380.1 (26050), 359.2 (23620), 341.9 (14540), 274.6 (7040), 269.8 (6600 $dm^3mol^{-1}cm^{-1}$) nm. MS (70 eV): *m/z* (%) 475 (24) [M^++1], 474 (80) [M^+], 459 (27) [M^+-CH_3], 458 (29), 457 (100), 441 (4), 439 (6), 429 (4), 425 (7), 413 (3), 310 (2), 308 (2), 237 (3) [M^{2+}]. Elemental analysis calcd (%) for $C_{30}H_{22}N_2O_4$ (474.5): C 75.94, H 4.67, N 5.90; found: C 75.61, H 4.63, N 5.76.

2,7-Bis(5-*tert*-butyl-2-methylphenyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (1p): Naphthalenetetracarboxylic bisanhydride (**2**, 2.75 g, 10.0 mmol), 5-*tert*-butyl-2-methylaniline (5.00 g, 30.0 mmol) and acetic acid (20 mL) were allowed to react as described above for **1i**. Yield 3.5 g (61%), fine, yellow needles, m.p. 318°C. R_f (silica gel, $CHCl_3$) = 0.46. R_f (silica gel, $CHCl_3$/acetic acid 39:1) = 0.57. IR (KBr): $\tilde{\nu}$ = 2960 (m), 2920 (m), 2870 (m), 1725 (s), 1680 (s), 1585 (s), 1520 (m), 1450 (m), 1420 (w), 1379 (m), 1350 (s), 1251 (s), 1215 (m), 1200 (s), 1178 (m), 1135 (w), 1125 (w), 1035 (w), 985 (m), 925 (w), 885 (m), 835 (m), 824 (m), 770 (s), 755 (s), 714 (w), 680 (w), 650 (m), 623 (w) cm^{-1}. ^1H NMR (80 MHz, $CDCl_3$): δ = 9.83 (s, 4H, naphthalene), 8.29 (m, 6H, 2phenyl), 2.18 (s, 6H, 2CH_3), 1.38 (s, 18H, 2$C(CH_3)_3$) ppm. UV/Vis ($CHCl_3$): λ_{max} (ε) = 380 (27130), 360 (24410), 343 (14920), 326 (7460), 240 (36720 $dm^3mol^{-1}cm^{-1}$) nm. MS (70 eV): *m/z* (%) 558 (31) [M^+], 543 (100) [M^+-CH_3], 264 (12). Elemental analysis calcd (%) for $C_{36}H_{34}N_2O_4$ (558.7): C 77.40, H 6.13, N 5.01; found: C 77.69, H 6.11, N 5.20.

2,7-Bis(2-*tert*-butylphenyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (1q): Naphthalenetetracarboxylic bisanhydride (**2**, 2.75 g, 10.0 mmol), 2-*tert*-butylaniline (4.57 g, 30.0 mmol) and acetic acid (20 mL) were allowed to react as described for **1i**. Yield 4.88 g (90%), colourless powder of a 1:1 mixture of the *cis* and *trans* atropic isomers. The two isomers were separated by column chromatography (silica gel, $CHCl_3$).

Isomer **I** (*trans*, *anti*[41]). Yield 1.4 g (26%), colourless powder, m.p. 332–338°C. R_f (silica gel, $CHCl_3$) = 0.30. R_f (silica gel, $CHCl_3$/acetic acid 39:1) = 0.54. IR (KBr): $\tilde{\nu}$ = 3424 (m), 2965 (m), 1912 (m), 2880 (m), 1714 (s), 1676 (s), 1581 (s), 1490 (m), 1447 (m), 1365 (m), 1344 (s), 1249 (s), 1216 (m), 1200 (m), 1148 (w), 1121 (w), 1085 (w), 1054 (w), 982 (m), 880 (w), 866 (m), 830 (w), 802 (w), 772 (s), 754 (s), 716 (s), 686 (w), 667 (w), 629 (m) cm^{-1}. ^1H NMR (600 MHz, $CDCl_3$): δ = 8.84 (s, 4H, naphthalene), 7.69/7.67 (d, 2H, phenyl), 7.46 (t, 2H, phenyl), 7.36 (t, 2H, phenyl), 7.02/7.01 (d, 2H, phenyl), 1.29 (s, 18H, $6CH_3$) ppm. ^{13}C NMR ($CDCl_3$): δ = 163.74, 147.05, 132.48, 131.48, 130.74, 129.52, 129.44, 127.47, 127.38, 127.12, 31.73 ppm. UV/Vis ($CHCl_3$): λ_{max} (ε) = 381 (28080), 360 (24290), 343 (14740), 326 (7580), 242 (26140 $dm^3 mol^{-1} cm^{-1}$) nm. MS (70 eV): *m/z* (%) 515 (4) [M^+], 473 (100) [$M^+ - C(CH_3)_3$]. Elemental analysis calcd (%) for $C_{34}H_{30}N_2O_4$ (530.6): C 76.96, H 5.70, N 5.28; found: C 77.34, H 5.86, N 4.98.

Isomer **II** (*cis*, *syn*[40]). Yield 1.28 g (23.5%), colourless, fine needles, m.p. 333°C. R_f (silica gel, $CHCl_3$) = 0.11. R_f (silica gel, $CHCl_3$/acetic acid 9.75:0.25) = 0.54. IR (KBr): $\tilde{\nu}$ = 3424 (w), 2965 (m), 2912 (m), 2880 (w), 1714 (s), 1676 (s), 1581 (s), 1490 (m), 1442 (m), 1365 (m), 1344 (s), 1249 (s), 1216 (m), 1200 (m), 1147 (w), 1121 (w), 1089 (w), 1054 (w), 982 (m), 880 (w), 866 (m), 832 (w), 802 (w), 722 (s), 754 (m), 716 (m), 682 (w), 670 (w), 629 (m) cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.84 (s, 4H, naphthalene), 7.70/7.68 (d, 2H, phenyl), 7.48 (t, 2H, phenyl), 7.37 (t, 2H, phenyl), 7.06/7.04 (d, 2H, phenyl), 1.31 (s, 18H, $6CH_3$) ppm. ^{13}C NMR ($CDCl_3$): δ = 163.74, 147.11, 132.56, 131.49, 130.79, 129.56, 129.40, 127.51, 127.31, 127.10, 31.79 ppm. UV/Vis ($CHCl_3$): $\lambda_{max.}$(ε) = 381 (28220), 360 (24690), 342 (14880), 326 (7500), 241 (36560 $dm^3 mol^{-1} cm^{-1}$) nm. MS (70 eV): *m/z* (%) 515 (4) [M^+], 473 (100) [$M^+ - C(CH_3)_3$]. Elemental analysis calcd (%) for $C_{34}H_{30}N_2O_4$ (530.6): C 76.96, H 5.70, N 5.28; found: C 76.55, H 5.70, N 5.31.

2,7-Bis(4-*tert*-butylphenyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (1r): Naphthalenetetracarboxylic bisanhydride (**2**, 2.75 g, 10.0 mmol), 4-*tert*-butylaniline (4.75 g, 30.0 mmol) and acetic acid (20 mL) were allowed to react as described above for **1i**, then dissolved in chloroform and precipitated with ethanol. Yield 2.72 g (50%), yellow, fine crystals, m.p. > 350°C. R_f (silica gel, $CHCl_3$) = 0.32. R_f (silica gel, $CHCl_3$/acetic acid 9.75:0.25) = 0.57. IR (KBr): $\tilde{\nu}$ = 2963 (m), 2906 (w), 2870 (w), 1718 (s), 1676 (s), 1653 (w), 1582 (m), 1515 (m), 1506 (w), 1447 (m), 1365 (m), 1347 (s), 1247 (s), 1198 (m), 1105 (m), 982 (m), 862 (m), 835 (m), 769 (s), 755 (m), 716 (m), 699 (w), 561 (m), 465 (w) cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ = 1.40 (s, 18H, $6CH_3$), 7.27 (dd, ^3J = 8.40 Hz, $^4J \approx 0.5$ Hz, AA′BB′ type, 4H, 4CH), 7.60 (dd, ^3J = 8.40 Hz, $^4J \approx 0.5$ Hz, AA′BB′ type, 4H, 4CH), 8.84 (s, 4H, 4CH) ppm. ^{13}C NMR ($CDCl_3$): δ = 31.35, 34.83, 126.58, 127.04, 127.15, 127.77, 131.38, 131.71, 152.05, 163.03 ppm. UV/Vis ($CHCl_3$): λ_{max} (ε) = 380 (28070), 360 (25270), 343 (15750), 325 (7240), 241 (34130 $dm^3 mol^{-1} cm^{-1}$) nm. MS (70 eV): *m/z* (%) 531 (55) [M^+], 515 (32) [$M^+ - CH_3$], 475 (28) [$M^+ - C(CH_3)_3$]. Elemental analysis calcd (%) for $C_{34}H_{30}N_2O_4$ (530.6): C 76.96, H 5.84, N 7.71; found: C 77.25, H 5.98, N 7.79.

2,7-Bis(2,5-di-*tert*-butylphenyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (1s): Naphthalenetetracarboxylic bisanhydride (**2**, 2.75 g, 10.0 mmol), 2,5-di-*tert*-butylaniline (4.17 g, 20.0 mmol) and acetic acid (60 mL) were allowed to react as described above for **1i**. Yield 3.43 g (52%), fine, pale yellow needles, m.p. >250°C. R_f (silica gel, $CHCl_3$) = 0.73. R_f (silica gel, $CHCl_3$/acetic acid 39:1) = 0.69. IR (KBr): $\tilde{\nu}$ = 2963 (m), 2870 (w), 1716 (s), 1680 (s), 1616 (w), 1583 (m), 1502 (w), 1447 (w), 1394 (w), 1364 (m), 1343 (s), 1251 (s), 1200 (m), 981 (w), 828 (m), 772 (m), 644 (w) cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ = 0.89 (t, 6H, $2CH_3$), 1.56 (d, 6H, $2CH_3$), 1.93 (m, 2H), 2.18 (m, 2H), 5.17 (sextet, 2H, 2CH), 8.70 (s, 4H, aromatic) ppm. ^{13}C NMR ($CDCl_3$): δ = 11.41, 17.96, 26.40, 51.90, 126.66, 126.79, 130.83, 163.33 ppm. UV/Vis ($CHCl_3$): λ_{max} (ε) = 381 (24980), 360 (22910), 343 (14890), 328 (8400), 242 (40110 $dm^3 mol^{-1} cm^{-1}$) nm. MS (70 eV): *m/z* (%) 642 (1) [M^+], 627 (7) [$M^+ - CH_3$], 585 (100) [$M^+ - C(CH_3)_3$], 528). Elemental analysis calcd (%) for $C_{42}H_{46}N_2O_4$ (642.8): C 78.47, H 7.21, N 4.36; found: C 77.48, H 7.28, N 4.32.

Benzo[*lmn*]bisimidazo[2,1-*b*:2′,1′-*i*]phenanthroline-6,12-dione (3) and benzo[*lmn*]bisimidazo[2,1-*b*:2′,1′-*j*]phenanthroline-3,6-dione (4): Naphthalene-1,8:4,5-tetracarboxylic bisanhydride (**2**, 500 mg, 1.9 mmol) and 1,2-diaminoethane (560 mg, 9.3 mmol) in quinoline (5 mL) were heated at 180°C for 2 h, cooled to room temperature, treated with ethanol (10 mL) and poured into 2N HCl (50 mL). This mixture was stirred for 1 h, allowed to stand for 1 h, collected by vacuum filtration, dried in air at 100°C, extracted with chloroform and purified by column chromatography (silica gel, chloroform/acetic acid 10:1). Dione **3** was obtained as the first yellow and yellow fluorescent fraction and dione **4** as the second nonfluorescent red one. Both fractions were separately further purified by column chromatography (silica gel, $CHCl_3$/acetone 5:1) and by extractive recrystallisation[40] from ethanol.

Dione **3**: Yield 98 mg (17%), m.p. >350°C. R_f (silica gel, $CHCl_3$/acetone 5:1) = 0.24. IR (KBr): $\tilde{\nu}$ = 3168 (m), 2925 (m), 2855 (m), 1712 (s, br), 1583 (m), 1551 (s), 1503 (s), 1470 (m), 1417 (s), 1393 (s), 1360 (s), 1338 (m), 1277 (s), 1245 (s), 1229 (m), 1110 (w), 1083 (w), 1067 (m), 1059 (s), 978 (m), 910 (w), 867 (m), 800 (w), 765 (s), 749 (m), 730 (m) cm^{-1}. ^1H NMR ($CDCl_3$): δ = 8.90 (d, J = 7.7 Hz, 2H, naphthalene), 8.78 (d, J = 7.7 Hz, 2H, naphthalene), 7.95 (d, J = 1.4 Hz, 2H, amidine), 7.45 (d, J = 1.7 Hz, 2H, amidine) ppm. UV ($CHCl_3$): λ_{max} (ε) = 448.1 (19050), 377.8 (8610), 364.9 (sh) (4700 $dm^3 mol^{-1} cm^{-1}$) nm. Fluorescence ($CHCl_3$): λ_{max} = 544 nm. Fluorescence quantum yield ($CHCl_3$, E = 0.0364 cm^{-1}, λ_{excit} = 430 nm, reference: perylene-3,4,9,10-tetracarboxylic tetramethyl ester with Φ = 1.0[42]) = 0.16. MS (70 eV): *m/z* (%) 313 (17) [M^++1], 312 (100) [M^+], 285 (7) [M^+−HCN], 230 (4), 202 (3), 188 (3), 176 (3), 156 (6) [M^{2+}]. Elemental analysis calcd (%) for $C_{18}H_8N_4O_2$ (312.3): C 69.23, H 2.56, N 17.95; found: C 69.42, H 2.72, N 17.90.

Dione **4**: Yield 47 mg (8%), m.p. >350°C. R_f (silica gel, $CHCl_3$/acetone 5:1) = 0.06. R_f (silica gel, $CHCl_3$/acetic acid 10:1) = 0.28. IR (KBr): $\tilde{\nu}$ = 3162 (w), 2925 (w), 2855 (w), 1712 (s), 1583 (w), 1549 (m), 1488 (w), 1469 (w), 1421 (m), 1389 (m), 1360 (m), 1285 (s), 1279 (s), 1233 (w), 1216 (m), 1185 (w), 1152 (w), 1076 (w), 893 (w), 862 (w), 804 (w), 760 (m), 749 (m), 744 (m), 709 (w) cm^{-1}. ^1H NMR ($CDCl_3$): δ = 8.95 (s, 2H, naphthalene), 8.73 (s, 2H, naphthalene), 7.90 (d, J = 1.5 Hz, 2H, amidine), 7.41 (d, J = 1.6 Hz, 2H, amidine) ppm. UV ($CHCl_3$): λ_{max} (ε) = 510.7 (8340), 374.9 (3930), 351.9 (5690 $dm^3 mol^{-1} cm^{-1}$) nm. UV (H_2SO_4): λ_{max} = 451.9, 400.1, 382.8 nm. Fluorescence (H_2SO_4): λ_{max} = 536.7 nm. MS (70 eV): *m/z* (%) 313 (17) [M^++1], 312 (100) [M^+], 285 (6) [M^+−HCN], 230 (6), 202 (3), 188 (3), 176 (2), 156 (6) [M^{2+}]. Elemental analysis calcd (%) for $C_{18}H_8N_4O_2$ (312.3): C 69.23, H 2.56, N 17.95; found: C 68.90, H 2.65, N 17.71.

2-(1-Methylpropyl)-benzo[*lmn*]imidazolo[1,2-*j*][3,8]phenanthrolin-1,3,6-trione (5a): Method 1: Naphthalene-1,8:4,5-tetracarboxylic bisanhydride (**2**, 1.4 g, 5.2 mmol), 1,2-diaminoethane (470 mg, 7.8 mmol) and 2-aminobutane (570 mg, 7.8 mmol) in quinoline (30 mL) were heated at 180°C for 2 h, cooled to room temperature, and poured into a mixture of ethanol (50 mL) and 2N HCl (100 mL). The yellowish-orange precipitate was allowed to stand for 1 h and collected by vacuum filtration through a D4 glass filter, dried in air (12 h, 100°C), extracted with chloroform and the chloroform phase purified by column chromatography (silica gel, $CHCl_3$/acetone 5:1). The reaction product was obtained as the first yellow band. Analytically pure material was prepared by further column chromatography (silica gel, $CHCl_3$/acetone 15:1) and extractive recrystallisation[40] from petroleum ether. Yield 90 mg (5%). Method 2: Naphthalene-1,8:4,5-tetracarboxylic bisanhydride (**2**, 1.4 g, 5.2 mmol), 1,2-diaminoethane (470 mg, 7.8 mmol) and 2-aminobutane (570 mg, 7.8 mmol) in *N*,*N*′-dimethylformamide (70 mL) were refluxed for 2 h. The solvent was removed in vacuo and the residue purified as described in method 1. Yield 380 mg (21%) of **4a**, m.p. 265°C. R_f (silica gel, $CHCl_3$/acetone 5:1) = 0.75. IR (KBr): $\tilde{\nu}$ = 3435 (m, br), 2969 (w), 2947 (w), 2875 (w), 1706 (s), 1667 (s), 1618 (w), 1580 (w), 1553 (w), 1510 (w), 1412 (m), 1380 (w), 1342 (s), 1282 (s), 1245 (m), 1150 (w), 1076 (w), 974 (w), 880 (w), 767 (m), 751 (m) cm^{-1}. ^1H NMR ($CDCl_3$): δ = 8.88 (d, J = 7.6 Hz, 1H, naphthalene), 8.75 (d, J = 7.6 Hz, 1H, naphthalene), 8.72 (s, 2H, naphthalene), 7.93 (d, J = 1.6 Hz, 1H, amidine), 7.42 (d, J = 1.6 Hz, 1H, amidine), 5.18 (m_c, 1H, CH), 2.17 (m_c, 1H, CH_2), 1.97 (m_c, 1H, CH_2), 1.57 (d, J = 7.0 Hz, 3H, CH_3), 0.91 (t, J = 7.5 Hz, 3H, CH_3) ppm. ^{13}C NMR ($CDCl_3$): δ = 163.4 (C=O), 163.2 (C=O), 158.3 (C=O), 144.5 (C=N), 133.0 (C-amidine), 132.0, 131.9, 130.5, 128.8, 127.5, 126.3, 125.4, 125.3, 124.6, 124.1

(10−C-naphthalene), 116.3 (C-amidine), 51.9 (CH), 26.5 (CH_2), 18.0 (CH_3), 11.5 (CH_3) ppm. UV ($CHCl_3$): λ_{max} (ε) = 436.1 (8010), 377.0 (7310), 359.0 (6700 $dm^3 mol^{-1} cm^{-1}$) nm. Fluorescence ($CHCl_3$): λ_{max} = 546 nm. Solid-state fluorescence: λ_{max} = 571 nm. MS (70 eV): *m/z* (%) 346 (4) [M^++1], 345 (72) [M^+], 316 (25) [$M^+-C_2H_5$], 289 (100) [$M^+-C_4H_8$], 272 (23), 244 (14), 218 (6), 190 (5). Elemental analysis calcd (%) for $C_{20}H_{15}N_3O_3$ (345.4): C 69.56, H 4.38, N 12.17; found: C 69.25, H 4.43, N 11.87.

2-(1-Methylbutyl)benzo[*lmn*]imidazolo[1,2-*j*][3,8]phenanthroline-1,3,6-trione (5b): Naphthalene-1,8:4,5-tetracarboxylic bisanhydride (**2**, 1.4 g, 5.2 mmol), 1,2-diaminoethane (470 mg, 7.8 mmol) and 2-aminopentane (680 mg, 7.8 mmol) in *N*,*N*′-dimethylformamide (70 mL) were allowed to react (4 h) and the product purified as described above for **5a**, method 2. Yield 210 mg (11%), m.p. 204 °C. R_f (silica gel, $CHCl_3$/acetone 5:1) = 0.58. IR (KBr): $\tilde{\nu}$ = 3432 (m, br), 2960 (m), 2931 (m), 2859 (w), 1706 (s), 1666 (s), 1616 (w), 1580 (m), 1552 (w), 1508 (w), 1439 (w), 1410 (m), 1385 (w), 1344 (s), 1277 (s), 1241 (s), 1088 (w), 880 (w), 766 (m), 752 (m) cm^{-1}. ^{1}H NMR ($CDCl_3$): δ = 8.90 (d, *J* = 7.6 Hz, 1H, naphthalene), 8.79 (d, *J* = 7.6 Hz, 1H, naphthalene), 8.75 (s, 2H, naphthalene), 7.95 (d, *J* = 1.6 Hz, 1H, amidine), 7.45 (d, *J* = 1.6 Hz, 1H, amidine), 4.20 (t, *J* = 7.6 Hz, 2H, CH_2), 1.75 (m_c, 2H, CH_2), 1.41 (m_c, 4H, CH_2), 0.93 (t, *J* = 7.1 Hz, 3H, CH_3) ppm. ^{13}C NMR ($CDCl_3$): δ = 162.9 (C=O), 162.7 (C=O), 158.2 (C=O), 144.5 (C=N), 133.1 (C-amidine), 132.0, 131.9, 130.5, 128.4, 127.5, 126.5, 125.6, 124.7, 123.8 (10C-naphthalene), 116.3 (C-amidine), 40.9 (CH_2), 29.2 (CH_2), 27.8 (CH_2), 22.4 (CH_2), 13.9 (CH_3) ppm. UV ($CHCl_3$): λ_{max} (ε) = 436.9 (7230), 377.3 (6490), 359.5 (6060 $dm^3 mol^{-1} cm^{-1}$) nm. Fluorescence ($CHCl_3$): λ_{max} = 541 nm. Solid-state fluorescence: λ_{max} = 573 nm. MS (70 eV): *m/z* (%) 360 (24) [M^++1], 359 (100) [M^+], 342 (6) [M^+−OH], 303 (23) [$M^+-C_4H_8$], 290 (25), 289 (60) [$M^+-C_5H_{10}$], 272 (12), 244 (8). Elemental analysis calcd (%) for $C_{21}H_{17}N_3O_3$ (359.4): C 70.18, H 4.77, N 11.69; found: C 70.15, H 4.87, N 11.53.

2-(1-Hexylheptyl)benzo[*lmn*]imidazolo[1,2-*j*][3,8]phenanthroline-1,3,6-trione (5c): Naphthalene-1,8:4,5-tetracarboxylic bisanhydride (**2**, 1.4 g, 5.2 mmol), 1,2-diaminoethane (470 mg, 7.8 mmol) and 7-aminotridecane (1.56 g, 7.8 mmol) in *N*,*N*′-dimethylformamide (70 mL) were allowed to react (4 h) and the product purified as described above for **5a**, method 2. Yield 390 mg (16%), thin needles from chloroform or long needles from DMF, m.p. 108 °C. R_f (silica gel, $CHCl_3$/acetone 5:1) = 0.85. IR (KBr): $\tilde{\nu}$ = 3427 (m, br), 2956 (m), 2928 (s), 2857 (m), 1707 (s), 1665 (s), 1616 (w), 1578 (w), 1552 (w), 1509 (w), 1458 (w), 1433 (w), 1410 (m), 1379 (w), 1342 (s), 1280 (s), 1244 (s), 1179 (w), 1104 (w), 1068 (w), 973 (w), 875 (w), 768 (m), 748 (m) cm^{-1}. ^{1}H NMR ($CDCl_3$): δ = 8.88 (d, *J* = 7.6 Hz, 1H, naphthalene), 8.73 (br s, 3H, naphthalene), 7.93 (d, *J* = 1.6 Hz, 1H, amidine), 7.43 (d, *J* = 1.6 Hz, 1H, amidine), 5.12 (m_c, 1H, CH), 2.20 (m_c, 2H, α-CH_2), 1.83 (m_c, 2H, α-CH_2), 1.24 (m_c, 16H, 8CH_2), 0.80 (t, *J* = 6.7 Hz, 6H, 2CH_3) ppm. ^{13}C NMR ($CDCl_3$): δ = 158.3 (3C=O), 144.5 (C=N), 133.0 (C-amidine), 132.0, 130.8, 127.6, 126.3, 125.4, 125.3, 124.6 (10C-naphthalene), 116.3 (C-amidine), 55.2 (CH), 32.3, 31.7, 29.7, 29.6, 29.3, 29.1, 29.0, 26.9, 22.7, 22.5 (10CH_2), 14.1 (CH_3), 14.0 (CH_3) ppm. UV ($CHCl_3$): λ_{max} (ε) = 436.4 (9960), 377.8 (8750), 360.0 (8500 $dm^3 mol^{-1} cm^{-1}$) nm. Fluorescence ($CHCl_3$): λ_{max} = 551 nm. Solid-state fluorescence: λ_{max} = 571 nm. Fluorescence quantum yield ($CHCl_3$, E = 0.0193 cm^{-1}, λ_{excit} = 375 nm, reference: perylene-3,4,9,10-tetracarboxylic tetramethyl ester with Φ = 1.0[41]) = 0.06. MS (70 eV): *m/z* (%) 472 (28) [M^++1], 471 (88) [M^+], 386 (5) [$M^+-C_6H_{13}$], 302 (10) [$M^+-C_{12}H_{25}$], 290 (100) [$M^+-C_{13}H_{25}$], 272 (17), 244 (6), 218 (3). Elemental analysis calcd (%) for $C_{29}H_{33}N_3O_3$ (471.6): C 73.86, H 7.05, N 8.91; found: C 74.10, H 6.96, N 8.90.

2-Cyclohexylbenzo[*lmn*]imidazolo[1,2-*j*][3,8]phenanthroline-1,3,6-trione (5d): Naphthalene-1,8:4,5-tetracarboxylic bisanhydride (**2**, 1.4 g, 5.2 mmol), 1,2-diaminoethane (470 mg, 7.8 mmol) and aminocyclohexane (870 mg, 7.8 mmol) in DMF (70 mL) were allowed to react (4 h) and the product purified as described above for **5a**, method 2. Yield 225 mg (12%), fine needles from chloroform, m.p. >350 °C. R_f (silica gel, $CHCl_3$/acetone 5:1) = 0.56. IR (KBr): $\tilde{\nu}$ = 3430 (m, br), 3155 (w), 2928 (m), 2854 (w), 1705 (s), 1667 (s), 1618 (w), 1580 (w), 1552 (w), 1509 (w), 1454 (w), 1438 (w), 1411 (m), 1341 (s), 1281 (s), 1242 (m), 1188 (w), 1108 (w), 1068 (w), 976 (w), 882 (w), 768 (m), 751 (m) cm^{-1}. ^{1}H NMR ($CDCl_3$): δ = 8.87 (d, *J* = 7.7 Hz, 1H, naphthalene), 8.74 (d, *J* = 7.7 Hz, 1H, naphthalene), 8.72 (s, 2H, naphthalene), 7.92 (d, *J* = 1.7 Hz, 1H, amidine), 7.42 (d, *J* = 1.5 Hz, 1H, amidine), 5.01 (m_c, 1H, CH), 2.52 (m_c, 2H, CH_2), 1.89 (m_c, 2H, CH_2), 1.74 (m_c, 2H, CH_2), 1.43 (m_c, 2H, CH_2), 1.33 (m_c, 2H, CH_2) ppm. ^{13}C NMR ($CDCl_3$): δ = 163.3 (C=O), 163.1 (C=O), 158.3 (C=O), 144.5 (C=N), 133.0 (C-amidine), 132.0, 131.9, 130.4, 129.0, 127.5, 126.2, 125.3, 125.2, 124.5, 124.3, (10C-naphthalene), 116.2 (C-amidine), 54.4 (CH), 29.1 (2CH_2), 26.5 (2CH_2), 25.3 (CH_2) ppm. UV ($CHCl_3$): λ_{max} (ε) = 436.1 (9670), 377.1 (8470), 360.4 (8470), 346.8 (8240), 330.2 (sh) (7260), 299.2 (14050), 288.6 (13510), 263.6 (15180 $dm^3 mol^{-1} cm^{-1}$) nm. Fluorescence ($CHCl_3$): λ_{max} = 552 nm. Solid-state fluorescence: λ_{max} = 571 nm. MS (70 eV): *m/z* (%) 372 (14) [M^++1], 371 (60) [M^+], 290 (100) [$M^+-C_6H_9$], 289 (87) [$M^+-C_6H_{10}$], 272 (11), 245 (6), 244 (6), 218 (4). Elemental analysis calcd (%) for $C_{22}H_{17}N_3O_3$ (371.4): C 71.15, H 4.61, N 11.31; found: C 70.79, H 4.52, N 11.30.

2-(2-Hydroxyethyl)benzo[*lmn*]imidazolo[1,2-*j*][3,8]phenanthroline-1,3,6-trione (5e): Naphthalene-1,8:4,5-tetracarboxylic bisanhydride (**2**, 1.4 g, 5.2 mmol), 1,2-diaminoethane (470 mg, 7.8 mmol) and 2-aminoethanol (480 mg, 7.8 mmol) in DMF (70 mL) were allowed to react (4 h) and the product purified as described above for **5a**, method 2 (silica gel, $CHCl_3$/ethanol 10:1 and silica gel, $CHCl_3$/acetone 5:1 to remove two yellow foreruns). Yield 280 mg (16%), m.p. 302 °C. R_f (silica gel, $CHCl_3$/ethanol 10:1) = 0.52. IR (KBr): $\tilde{\nu}$ = 3428 (s, br), 2926 (w), 1706 (s), 1665 (s), 1616 (w), 1580 (m), 1553 (w), 1508 (w), 1480 (w), 1438 (w), 1410 (m), 1344 (m), 1284 (s), 1241 (m), 1175 (w), 1054 (w), 971 (w), 874 (w), 766 (m), 754 (w) cm^{-1}. ^{1}H NMR ($CDCl_3$): δ = 8.85 (d, *J* = 7.7 Hz, 1H, naphthalene), 8.75 (d, *J* = 7.6 Hz, 1H, naphthalene), 8.71 (d, *J* = 7.6 Hz, 2H, naphthalene), 7.89 (d, *J* = 1.4 Hz, 1H, amidine), 7.39 (d, *J* = 1.4 Hz, 1H, amidine), 4.42 (t, *J* = 5.3 Hz, 2H, CH_2), 3.95 (t, *J* = 5.0 Hz, 2H, CH_2) ppm. ^{13}C NMR ($CDCl_3$): δ = 163.6 (C=O), 142.3 (C=N), 140.3, 136.9, 135.8, 133.2, (C-amidine), 132.3, 132.0, 130.8, 128.0, 127.6, 126.8, 125.3, 123.4, (10C-naphthalene), 116.4 (C-amidine), 61.4 (CH_2), 43.0 (CH_2) ppm. UV ($CHCl_3$): λ_{max} (ε) = 439.0 (8010), 377.5 (7390), 360.1 (6850), 347.1 (6660), 331.0 (sh) (5420), 299.4 (11900), 289.7 (11670), 263.0 (12810 $dm^3 mol^{-1} cm^{-1}$) nm. Fluorescence ($CHCl_3$): λ_{max} = 555 nm. Solid-state fluorescence: λ_{max} = 575 nm. MS (70 eV): *m/z* (%) 334 (9) [M^++1], 333 (35) [M^+], 304 (5), 303 (14) [M^+-CH_2O], 302 (25) [M^+-CH_2OH], 290 (100), 289 (27) [$M^+-C_2H_4OH$], 272 (22), 244 (10), 218 (7), 191 (4). Elemental analysis calcd (%) for $C_{18}H_{11}N_3O_4$ (333.3): C 64.87, H 3.33, N 12.61; found: C 64.57, H 3.24, N 12.55.

2-(2-Hydroxypropyl)benzo[*lmn*]imidazolo[1,2-*j*][3,8]phenanthroline-1,3,6-trione (5f): Naphthalene-1,8:4,5-tetracarboxylic bisanhydride (**2**, 1.4 g, 5.2 mmol), 1,2-diaminoethane (470 mg, 7.8 mmol) and 1-aminopropan-2-ol (590 mg, 7.8 mmol) in DMF (70 mL) were allowed to react (4 h) and the product purified as described above for **5a**, method 2 (silica gel, $CHCl_3$/ethanol 5:1 and silica gel, $CHCl_3$/ethanol 10:1) and extractive[40] recrystallisation from methanol. Yield 570 mg (32%), m.p. 278 °C. R_f (silica gel, $CHCl_3$/ethanol 10:1) = 0.14. IR (KBr): $\tilde{\nu}$ = 3436 (s, br), 2929 (w), 1707 (s), 1666 (s), 1611 (m), 1581 (w), 1553 (w), 1509 (w), 1438 (w), 1411 (m), 1386 (w), 1351 (m), 1286 (s), 1243 (m), 1067 (w), 981 (w), 767 (m), 754 (w) cm^{-1}. ^{1}H NMR ($CDCl_3$): δ = 8.90 (d, *J* = 7.7 Hz, 1H, naphthalene), 8.79 (d, *J* = 7.7 Hz, 1H, naphthalene), 8.75 (s, 2H, naphthalene), 7.93 (d, *J* = 1.5 Hz, 1H, amidine), 7.44 (d, *J* = 1.6 Hz, 1H, amidine), 4.36 (m_c, 1H, CH), 4.28 (d, *J* = 3.5 Hz, 2H, CH_2), 1.35 (d, *J* = 6.2 Hz, 3H, CH_3) ppm. UV ($CHCl_3$): λ_{max} (ε) = 439.6 (7350), 377.5 (6540), 360.1 (6000 $dm^3 mol^{-1} cm^{-1}$) nm. Fluorescence ($CHCl_3$): λ_{max} = 556 nm. Solid-state fluorescence: λ_{max} = 575 nm. MS (70 eV): *m/z* (%) 348 (4) [M^++1], 347 (16) [M^+], 304 (21) [M^++1−C_2H_4O], 303 (100) [$M^+-C_2H_4O$], 290 (49) [$M^+-C_3H_5O$], 275 (15), 272 (11), 259 (8), 246 (8), 219 (9). Elemental analysis calcd (%) for $C_{19}H_{13}N_3O_4$ (347.3): C 65.70, H 3.77, N 12.10; found: C 65.50, H 3.69, N 12.10.

2-(4-*tert*-Butylphenyl)benzo[*lmn*]imidazolo[1,2-*j*][3,8]phenanthroline-1,3,6-trione (5r) and *N*,*N*′-bis(4-*tert*-butylphenyl)-1,2,3,4-tetrahydronaphtho[2,3-*b*]pyrazine-5,6:9,10-bis(dicarboximide) (6r): 2,7-Bis(4-*tert*-butylphenyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (**1r**, 530 mg, 1.0 mmol) was refluxed in DMF (10 mL, bath 160 °C) and treated with 1,2-diaminoethane (1.2 g, 20 mmol). The mixture was cooled,

poured into a mixture of water (40 mL) and 2 N HCl (20 mL), stirred for 1 h, allowed to stand for 16 h, collected by vacuum filtration (D4 glass filter), dried in air (100 °C) and purified by column chromatography (silica gel, $CHCl_3$/acetone 5:1). The first yellow, strongly fluorescent fraction of **6r** was collected and purified by column chromatography (silica gel, chloroform) and the second fraction of **5r** was collected and also purified by column chromatography (silica gel, $CHCl_3$/acetone 15:1).

6r: Yield 62 mg (11 %), m.p. >350 °C. R_f (silica gel, $CHCl_3$/acetone 5:1) = 0.88. IR (KBr): $\tilde{\nu}$ = 3436 (m, br), 3248 (w), 2961 (m), 2927 (m), 2869 (w), 1690 (s), 1636 (s), 1601 (m), 1549 (s), 1496 (m), 1438 (w), 1394 (m), 1364 (w), 1340 (m), 1284 (w), 1210 (m), 1040 (w), 864 (w), 834 (w), 783 (w), 717 (w) cm^{-1}. ^1H NMR ($CDCl_3$): δ = 10.58 (s, 2H, 2NH), 8.42 (s, 2H, naphthalene), 7.59 (d, J = 8.7 Hz, 4H, phenyl), 7.25 (d, J = 9.1 Hz, 4H, phenyl), 3.73 (s, 4H, $2CH_2$), 1.39 (s, 18H, $2C(CH_3)_3$) ppm. ^{13}C NMR ($CDCl_3$): δ = 167.0 (2C=O), 163.8 (2C=O), 151.6 (2C-phenyl), 144.3 (2C-naphthalene), 132.5 (2C-naphthalene), 127.8, 126.5, 125.3 (2C-naphthalene), 123.2 (2C-phenyl), 38.2 ($2CH_2$), 34.8 ($2C(CH_3)_3$), 31.4 ($6CH_3$) ppm. UV ($CHCl_3$): λ_{max} (ε) = 488.9 (32680), 458.0 (19790), 432.5 (17670), 413.0 (12930 $dm^3 mol^{-1} cm^{-1}$) nm. Fluorescence ($CHCl_3$): λ_{max} (I_{rel}) = 500 (1), 526 (0.65) nm. MS (70 eV): m/z (%) 587 (42) [M^++1], 586 (100) [M^+], 571 (21) [M^+-CH_3], 437 (15) [M^+-CH_3-$C_{10}H_{14}$], 377 (3), 278 (6). Elemental analysis calcd (%) for $C_{36}H_{34}N_4O_4$ (586.7): C 73.70, H 5.84, N 9.55; found: C 73.63, H 5.88, N 9.15.

5r: Yield 45 mg (11 %), m.p. >350 °C. R_f (silica gel, $CHCl_3$/acetone 5:1) = 0.65. IR (KBr): $\tilde{\nu}$ = 3435 (m, br), 2924 (s), 2852 (m), 1712 (s), 1664 (s), 1582 (w), 1552 (w), 1506 (w), 1464 (w), 1407 (m), 1382 (w), 1352 (m), 1281 (m), 1241 (m), 1195 (m), 1130 (w), 1066 (w), 976 (w), 884 (w), 832 (w), 766 (m), 748 (m), 720 (w) cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.92 (d, J = 7.6 Hz, 1H, naphthalene), 8.82 (d, J = 7.9 Hz, 1H, naphthalene), 8.78 (s, 2H, naphthalene), 7.95 (d, J = 1.7 Hz, 1H, amidine), 7.57 (d, J = 7.5 Hz, 2H, phenyl), 7.45 (d, J = 1.5 Hz, 1H, amidine), 7.24 (d, J = 8.3 Hz, 2H, phenyl), 1.38 (s, 9H, $3CH_3$) ppm. ^{13}C NMR ($CDCl_3$): δ = 163.2 (C=O), 163.0 (C=O), 158.2 (C=O), 151.9 (C-phenyl), 144.5 (C=N), 133.2 (C-amidine), 132.4, 132.1, 131.8, 130.9, 128.5, 127.9, 127.8, 126.8, 126.5, 125.9, 125.4, 124.8, 123.9, (10C-naphthalene and 5C-phenyl), 116.4 (C-amidine), 34.8 ($C(CH_3)_3$), 31.3 ($3CH_3$) ppm. UV/Vis ($CHC1_3$): λ_{max} (ε) = 437.1 (8620), 375.4 (7680), 357.7 (7220 $dm^3 mol^{-1} cm^{-1}$) nm. Fluorescence ($CHC1_3$): λ_{max} = 553 nm. Solid-state fluorescence: λ_{max} = 573 nm. MS (70 eV): m/z (%) 422 (13) [M^++1], 421 (43) [M^+], 407 (29) [M^++1−CH_3], 406 (100) [M^+-CH_3], 388 (4), 272 (24), 244 (10), 203 (6), 189 (13). Elemental analysis calcd (%) for $C_{26}H_{19}N_3O_3$ (421.5): 74.10, H 4.54, N 9.97; found: 74.18, H 4.50, N 9.94.

2-(2,5-Di-*tert*-butylphenyl)benzo[*lmn*]imidazolo[1,2-*j*]-[3,8]phenanthroline-1,3,6-trione (5s) and *N,N'*-bis(2,5-di-*tert*-butylphenyl)-1,2,3,4-tetrahydronaphtho[2,3-*b*]pyrazine-5,6:9,10-bis(dicarboximide) (6s): 2,7-Bis(2,5-di-*tert*-butylphenyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (**1s,** 480 mg, 1.0 mmol), diaminoethane (1.2 g, 20 mmol) and *N,N'*-dimethylformamide (10 mL) were allowed to react as described above for 2-(4-*tert*-butylphenyl)benzo[*lmn*]imidazolo[1,2-*j*][3,8]phenanthroline-1,3,6-trione (**5r**).

6s: Yield 22 mg (3 %), m.p. >350 °C. R_f (silica gel, $CHCl_3$) = 0.24. IR (KBr): $\tilde{\nu}$ = 3435 (m, br), 3248 (w), 2963 (m), 2870 (m), 1690 (s), 1636 (s), 1602 (m), 1580 (w), 1547 (s), 1497 (m), 1437 (w), 1396 (m) , 1363 (w), 1318 (w), 1253 (m), 1126 (w), 1042 (w), 828 (w), 791 (w), 765 (w), 750 (w) cm^{-1}. ^1H NMR (300 MHz, $CDC1_3$): δ = 10.61 (s, 2H, 2NH), 8.42 (d, J = 3.1 Hz, 2H, naphthalene), 7.60 (d, J = 0.9 Hz, 1H, phenyl), 7.57 (d, J = 0.9 Hz, 1H, phenyl), 7.46 (dd, J^3 = 2.5 Hz, J^4 = 1.3 Hz, 1H, phenyl), 7.44 (dd, J^3 = 2.5 Hz, J^4 = 1.3 Hz, 1H, phenyl), 6.99 (d, J = 2.2 Hz, 1H, phenyl), 6.93 (d, J = 2.2 Hz, 1H, phenyl), 3.72 (br s, 4H, $2CH_2$), 1.32 (s, 9H, C-$(CH_3)_3$), 1.30 (s, 9H, $C(CH_3)_3$), 1.27 (s, 9H, $C(CH_3)_3$), 1.26 (s, 9H, C-$(CH_3)_3$) ppm. ^{13}C NMR ($CDC1_3$): δ = 167.7 (2C=O), 164.5 (2C=O), 150.1 (2C-phenyl), 144.3 (2C-phenyl), 143.7 (2C-phenyl), 132.7 (2C-phenyl), 128.2 (2C-phenyl), 127.6 (2C-phenyl), 126.2, 125.3, 123.4, 123.3, 123.2, (10C-naphthalene), 38.2 ($2CH_2$), 35.6 ($2C(CH_3)_3$), 34.2 ($2C(CH_3)_3$), 31.8 ($3CH_3$), 31.7 ($3CH_3$), 31.3 ($3CH_3$), 31.2 ($3CH_3$) ppm. UV/Vis ($CHC1_3$): λ_{max} (ε) = 488.3 (32580), 457.7 (19400) 432.1 (17090), 412.6 (12390 $dm^3 mol^{-1} cm^{-1}$) nm. Fluorescence ($CHCl_3$): λ_{max} (I_{rel}) = 500 (1), 526 (0.65) nm. Fluorescence quantum yield ($CHCl_3$, E = 0.0191 cm^{-1}, λ_{excit} = 455 nm, reference: perylene-3,4,9,10-tetracarboxylic tetramethyl ester with Φ = 1.0[41]) = 0.42. MS (70 eV): m/z (%) 699 (14) [M^++1], 698 (29) [M^+], 656 (6), 642 (44), 641 (100) [$M^+-C(CH_3)_3$], 585 (13) [$M^+-2\times C(CH_3)_3$], 571 (3), 434 (1), 313 (2). Elemental analysis calcd (%) for $C_{44}H_{50}N_4O_4$ (698.9): C 75.54, H 7.21, N 8.02; found: C 75.01, H 7.15, N 7.85.

5s: Yield 18 mg (4 %), m.p. >350 °C. R_f (silica gel, $CHCl_3$/acetone 5:1) = 0.68. IR (KBr): $\tilde{\nu}$ = 3430 (m, br), 2962 (m), 2868 (w), 1715 (s), 1676 (s), 1582 (w), 1552 (w), 1507 (w), 1481 (w), 1406 (m), 1345 (s), 1280 (s), 1245 (m), 1191 (w), 1067 (w), 977 (w), 824 (w), 768 (m), 749 (m), 720 (w) cm^{-1}. ^1H NMR (400 MHz, $CDC1_3$): δ = 8.94 (d, J = 7.7 Hz, 1H, naphthalene), 8.84 (d, J = 7.5 Hz, 1H, naphthalene), 8.80 (s, 2H, naphthalene), 7.96 (d, J = 1.7 Hz, 1H, amidine), 7.58 (d, J = 8.6 Hz, 2H, phenyl), 7.47 (d, J = 8.6 Hz, 1H, phenyl), 7.46 (d, J = 1.4 Hz, 1H, amidine), 7.00 (d, J = 2.2 Hz, 1H, phenyl), 1.31 (s, 9H, $3CH_3$), 1.27 (s, 9H, $3CH_3$) ppm. ^{13}C NMR ($CDC1_3$): δ = 163.9 (C=O), 163.8 (C=O), 158.3 (C=O), 150.3 (C-Phenyl), 144.5 (C=N), 143.8, 133.2 (C-amidine), 132.5, 132.1, 132.0, 131.0, 128.9, 128.6, 127.9, 127.6, 126.8, 126.6, 126.0, 125.4, 124.9, 124.0, (10C-naphthalene and 5C-phenyl), 116.4 (C-amidine), 35.6 ($C(CH_3)_3$), 34.3 ($C-(CH_3)_3$), 31.7 ($3CH_3$), 31.2 ($3CH_3$) ppm. UV/Vis ($CHC1_3$): λ_{max} (ε) = 437.6 (8010), 376.5 (8890), 359.0 (8290 $dm^3 mol^{-1} cm^{-1}$) nm. Fluorescence ($CHC1_3$): λ_{max} = 553 nm. Solid-state fluorescence: λ_{max} = 573 nm. MS (70 eV): m/z (%) 477 (2) [M^+], 462 (7) [M^+-CH_3], 421 (25) [M^++1−$C(CH_3)_3$], 420 (100) [$M^+-C(CH_3)_3$], 404 (8), 388 (4), 272 (2), 190 (3). HRMS (70 eV) for $C_{30}H_{27}N_3O_3$: calcd 477.2053; found: 477.2066.

2-(2,5-Dimethylphenyl)benzo[*lmn*]imidazolo[1,2-*j*][3,8]phenanthroline-1,3,6-trione (5o) and *N,N'*-bis(2,5-dimethylphenyl)-1,2,3,4-tetrahydronaphtho[2,3-*b*]pyrazine-5,6:9,10-bis(dicarboximide) (6o): 2,7-Bis(2,3-dimethylphenyl)benzo[*lmn*][3,8]phenanthroline-1,3,6,8-tetrone (**1o**, 470 mg, 1.0 mmol), diaminoethane (1.2 g, 20 mmol) and *N,N'*-dimethylformamide (10 mL) were allowed to react as described above for 2-(4-*tert*-butylphenyl)benzo[*lmn*]imidazolo[1,2-*j*][3,8]phenanthroline-1,3,6-trione (**5r**).

6o: Yield 37 mg (7 %), m.p. >350 °C. R_f (silica gel, $CHCl_3$/acetone 15:1) = 0.23. IR (KBr): $\tilde{\nu}$ = 3436 (m, br), 3245 (w), 2923 (w), 2867 (w), 1689 (s), 1635 (s), 1601 (m), 1548 (s), 1496 (s), 1461 (w), 1438 (w), 1394 (m) , 1363 (w), 1339 (m), 1282 (m), 1258 (w), 1215 (m), 1164 (w), 1045 (w), 811 (w), 786 (m) cm^{-1}. ^1H NMR (300 MHz, $CDC1_3$): δ = 10.57 (s, 2H, 2NH), 8.41 (s, 2H, naphthalene), 7.29 (d, J = 7.8 Hz, 2H, phenyl), 7.20 (d, J = 7.8 Hz, 2H, phenyl), 7.02 (d, J = 3.5 Hz, 2H, phenyl), 3.71 (s, 4H, $2CH_2$), 2.37 (s, 6H, $2CH_3$), 2.12 (s, 6H, $2CH_3$) ppm. ^{13}C NMR ($CDCl_3$): δ = 166.6 (2C=O), 163.3 (2C=O), 144.3 (2C-phenyl), 137.0, 134.3, 132.6, 132.5, 130.9, 130.8, 130.0, 128.8, 128.7, 125.3, 123.4, 123.1 (10C-phenyl and 8C-naphthalene), 98.1 (2C-naphthalene), 38.2 ($2CH_2$), 20.9 ($2CH_3$), 17.1 ($2CH_3$) ppm. UV/Vis ($CHC1_3$): λ_{max} (ε) = 488.8 (31170), 458.0 (19780), 431.5 (17640), 413.3 (12560), 383.3 (sh) (7110 $dm^3 mol^{-1} cm^{-1}$) nm. Fluorescence ($CHC1_3$): λ_{max} (I_{rel}) = 498 nm (1), 527 (0.52), 567 (sh) (0.1) nm. MS (70 eV): m/z (%) 531 (36) [M^++1], 530 (100) [M^+], 514 (25), 513 (71), 409 (4), 338 (4), 265 (6), 105 (11). Elemental analysis calcd (%) for $C_{32}H_{26}N_4O_4$ (530.6): C 72.30, H 4.94, N 10.54; found: C 71.97, H 5.15, N 10.40.

5o: Yield 28 mg (7 %), m.p. >350 °C. R_f (silica gel, $CHCl_3$/acetone 5:1) = 0.49. IR (KBr): $\tilde{\nu}$ = 3431 (m, br), 2924 (m), 2855 (w), 1715 (s), 1677 (s), 1581 (m), 1552 (w), 1508 (m), 1438 (m), 1407 (w), 1380 (w), 1343 (s), 1319 (w), 1282 (m), 1247 (s), 1194 (w), 1146 (w), 978 (w), 812 (w), 767 (m), 756 (m) cm^{-1}. ^1H NMR (400 MHz, $CDC1_3$): δ = 8.94 (d, J = 7.5 Hz, 1H, naphthalene), 8.83 (d, J = 7.5 Hz, 1H, naphthalene), 8.79 (s, 2H, naphthalene), 7.96 (d, J = 1.5 Hz, 1H, amidine), 7.46 (d, J = 1.5 Hz, 1H, amidine), 7.29 (d, J = 7.9 Hz, 1H, phenyl), 7.23 (d, J = 7.9 Hz, 1H, phenyl), 7.02 (d, J = 2.5 Hz, 1H, phenyl), 2.37 (s, 3H, CH_3), 2.15 (s, 3H, CH_3) ppm. ^{13}C NMR ($CDC1_3$): δ = 162.7 (C=O), 162.5 (C=O), 158.2 (C=O), 144.5 (C=N), 137.0, 133.6 (C-amidine), 133.2, 132.5, 132.4, 132.1, 131.0, 130.9, 130.3, 128.8, 128.4, 128.0, 126.8, 126.0, 125.4, 124.8, 123.7, (10C-naphthalene and 6C-phenyl), 116.4 (C-amidine), 20.9 (CH_3), 17.2 (CH_3). UV/Vis ($CHC1_3$): λ_{max} (ε) = 437.1 (8250), 376.4 (9210), 358.1 (8570), 437.6 (8010), 376.5 (8890), 359.0 (8290 $dm^3 mol^{-1} cm^{-1}$) nm. Fluorescence ($CHC1_3$): λ_{max} = 555 nm. Solid-state fluorescence: λ_{max} = 571 nm. MS (70 eV): m/z (%) 394 (13) [M^++1], 393 (51) [M^+], 378 (25) [M^+

+1−CH_3], 377 (27) [M^+−CH_3], 376 (100) [M^+−OH], 361 (16) [M^+−OH−CH_3], 348 (7), 333 (5), 282 (4), 244 (4), 222 (4). Elemental analysis calcd (%) for $C_{24}H_{15}N_3O_3$ (393.4): C 73.19, H 3.84, N 10.67; found: C 72.15, H 3.98, N 10.15.

***N,N′*-Bis(1-methylpropyl)-1,2,3,4-tetrahydronaphtho[2,3-*b*]pyrazine-5,6:9,10-bis(dicarboximide) (6a)**: 2,7-Bis(2,3-dimethylphenyl)benzo[*lmn*]-[3,8]phenanthroline-1,3,6,8-tetrone (**1a**, 430 mg, 1.0 mmol), diaminoethane (1.2 g, 20 mmol) and *N,N′*-dimethylformamide (10 mL) were allowed to react as described above for 2-(4-*tert*-butylphenyl)benzo-[*lmn*]imidazolo[1,2-*j*][3,8]phenanthroline-1,3,6-trione (**5r**). The second, strongly yellow fluorescent fraction was collected and purified by two further column-chromatographic separations (silical gel, $CHCl_3$/acetone 15:1 and silica gel, $CHCl_3$). Yield 4 mg (1%), m.p. 217°C. R_f (silica gel, $CHCl_3$/acetone 15:1) = 0.33. IR (KBr): $\tilde{\nu}$ = 3435 (m, br), 3248 (w), 2963 (m), 2870 (m), 1690 (s), 1636 (s), 1602 (m), 1580 (w), 1547 (s), 1497 (m), 1437 (w), 1396 (m), 1363 (w), 1318 (w), 1253 (m), 1126 (w), 1042 (w), 828 (w), 791 (w), 765 (w), 750 (w) cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ = 10.63 (s, 2H, 2NH), 8.30 (s, 2H, naphthalene), 5.22 (m_c, 2H, 2CH), 3.76 (s, 4H, $2CH_2$), 2.19 (m_c, 2H, $2CH_2$), 1.93 (m_c, 2H, $2CH_2$), 1.55 (d, J = 6.9 Hz, 6H, $2CH_3$), 0.87 (d, J = 7.5 Hz, 6H, $2CH_3$) ppm. ^{13}C NMR ($CDCl_3$): δ = 167.3 (2C=O), 164.5 (2C=O), 144.0, 124.8, 122.6, 98.2 (10C-naphthalene), 51.0 (2CH), 38.3 ($2CH_2$), 26.5 ($2CH_2$), 18.0 (CH_3), 11.5 (CH_3) ppm. UV/Vis ($CHCl_3$): λ_{max} (E_{rel}) = 488.9 (1), 458.0 (0.63), 432.5 (0.55), 413.0 (0.41) nm. Fluorescence ($CHCl_3$): λ_{max} (I_{rel}) = 500 (1), 526 (0.63) nm. MS (70 eV): m/z (%) 435 (27) [M^++1], 434 (100) [M^+], 418 (12), 404 (18), 395 (7), 322 (92) [M^+−C_8H_{16}], 320 (61), 274 (11), 249 (8). HRMS (70 eV) for $C_{24}H_{26}N_4O_4$: calcd 434.1954; found: 434.1924.

7a: 2-(1-Methylpropyl)benzo[*lmn*]imidazolo[1,2-*j*][3,8]phenanthroline-1,3,6-trione (**5a**, 60 mg, 0.17 mmol) was dispersed in a mixture of methanol (3 mL) and DMSO (3 mL), treated with 85% solid KOH (490 mg, 7.5 mmol), refluxed for 3 h (colour change to red), cooled, poured into conc. HCl, stirred for 1 h, left to stand for 16 h, collected by vacuum filtration (D4 glass filter, orange solid) and purified by column chromatography (silica gel, $CHCl_3$/acetone 15:1). Yield 6 mg (12%), m.p. 248°C. R_f (silica gel, $CHCl_3$/acetone 15:1) = 0.48. IR (KBr): $\tilde{\nu}$ = 3431 (m, br), 2971 (w), 2934 (w), 2878 (w), 1706 (s), 1657 (m), 1623 (w), 1585 (w), 1556 (w), 1495 (s), 1469 (w), 1446 (w), 1418 (m), 1390 (w), 1352 (m), 1270 (m), 1179 (w), 1065 (w), 1029 (w), 953 (w), 755 (m) cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.47 (d, J = 7.6 Hz, 1H, naphthalene), 8.46 (d, J = 7.5 Hz, 1H, naphthalene), 8.10 (d, J = 7.3 Hz, 1H, naphthalene), 7.87 (d, J = 1.5 Hz, 1H, amidine), 7.31 (d, J = 1.5 Hz, 1H, amidine), 7.12 (d, J = 7.6 Hz, 1H, naphthalene), 4.52 (m_c, 1H, CH), 2.01 (m_c, 1H, CH_2), 1.88 (m_c, 1H, CH_2), 1.55 (d, J = 6.9 Hz, 3H, CH_3), 0.93 (t, J = 7.4 Hz, 3H, CH_3) ppm. ^{13}C NMR ($CDCl_3$): δ = 167.8 (C=O), 158.6 (C=O), 146.1, 144.9 (C=N), 136.0 (C-amidine), 131.9, 126.0, 125.4, 125.3, 107.1 (10C-naphthaliene), 116.8 (C-amidine), 50.3 (CH), 27.5 (CH_2), 18.8 (CH_3), 11.2 (CH_3) ppm. UV/Vis ($CHCl_3$): λ_{max} (ε) = 424.1 (15910), 408.5 (sh) (14710), 362.4 (7830), 346.8 (sh) (3370), 255.6 (32840 $dm^3 mol^{-1} cm^{-1}$) nm. Fluorescence ($CHCl_3$): λ_{max} = 536 nm. Solid-state fluorescence: λ_{max} = 570 nm. Fluorescence quantum yield ($CHCl_3$, E = 0.0198 cm^{-1}, λ_{excit} = 405 nm, reference: perylene-3,4,9,10-tetracarboxylic tetramethyl ester with Φ = 1.0[41]) = 0.12. MS (70 eV): m/z (%) 318 (11) [M^++1], 317 (48) [M^+], 302 (6) [M^+−CH_3], 288 (100) [M^+−C_2H_5], 261 (15) [M^+−C_4H_8], 219 (4), 191 (3). HRMS (70 eV) for $C_{19}H_{15}N_3O_2$: calcd 317.1164; found: 317.1167.

7r: 2-(4-*tert*-Butylphenyl)benzo[*lmn*]imidazolo[1,2-*j*][3,8]phenanthroline-1,3,6-trione (**5r**, 50 mg, 0.12 mmol) and 85% KOH (350 mg, 5.3 mmol) were allowed to react as was described for **7a** and purified by extractive[39] recrystallisation from ethanol. Yield 35 mg (74%), m.p. >350°C. R_f (silica gel, $CHCl_3$/acetone 15:1) = 0.18. IR (KBr): $\tilde{\nu}$ = 3431 (m, br), 2961 (m), 2925 (m), 2854 (w), 1732 (m), 1709 (s), 1656 (m), 1628 (w), 1518 (m), 1495 (s), 1468 (w), 1447 (w), 1415 (m), 1365 (m), 1271 (m), 1248 (w), 1235 (w), 1175 (w), 1031 (w), 967 (w), 912 (w), 834 (w), 754 (m) cm^{-1}. ^1H NMR (400 MHz, $CDCl_3$): δ = 8.52 (d, J = 7.4 Hz, 1H, naphthalene), 8.48 (d, J = 7.7 Hz, 1H, naphthalene), 8.21 (d, J = 7.4 Hz, 1H, naphthalene), 7.89 (d, J = 1.7 Hz, 1H, amidine), 7.58 (d, J = 8.8 Hz, 2H, phenyl), 7.45 (d, J = 8.8 Hz, 2H, phenyl), 7.33 (d, J = 1.4 Hz, 1H, amidine), 7.11 (d, J = 7.7 Hz, 1H, naphthalene), 1.38 (s, 9H, $3CH_3$) ppm. ^{13}C NMR ($CDCl_3$): δ = 167.0 (C=O), 158.5 (C=O), 151.4 (C-phenyl), 144.8 (C=N), 136.1 (C-amidine), 132.0, 131.3, 126.7, 126.6, 125.8, 125.7, 125.5, 125.3, 124.8, 123.4, 106.9 (10C-naphthalene and 5C-phenyl), 116.6 (C-amidine), 34.8 ($C(CH_3)_3$), 31.3 ($3CH_3$) ppm. UV/Vis ($CHCl_3$): λ_{max} (ε) = 428.7 (sh) (12800), 413.3 (13870), 364.8 (8650), 258.0 (32660 $dm^3 mol^{-1} cm^{-1}$) nm. Fluorescence ($CHCl_3$): λ_{max} = 544 nm. Solid-state fluorescence: λ_{max} = 575 nm. MS (70 eV): m/z (%) 394 (19) [M^++1], 393 (68) [M^+], 379 (23) [M^++1−CH_3], 378 (100) [M^+−CH_3], 360 (3), 350 (7) [M^+−CH_3−CO], 338 (4), 337 (4), 189 (5), 175 (8). HRMS (70 eV) for $C_{25}H_{19}N_3O_2$: calcd 393.1464; found: 393.1471. Elemental analysis calcd (%) for $C_{25}H_{19}N_3O_2$ (393.4): C 76.25, H 4.87, N 10.68; found: C 74.54, H 4.73, N 10.35.

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