Supporting Information for:

The effect of terminal dimethyl and diethyl substituents on the J-aggregate-like molecular arrangement of bisazomethine dye molecules

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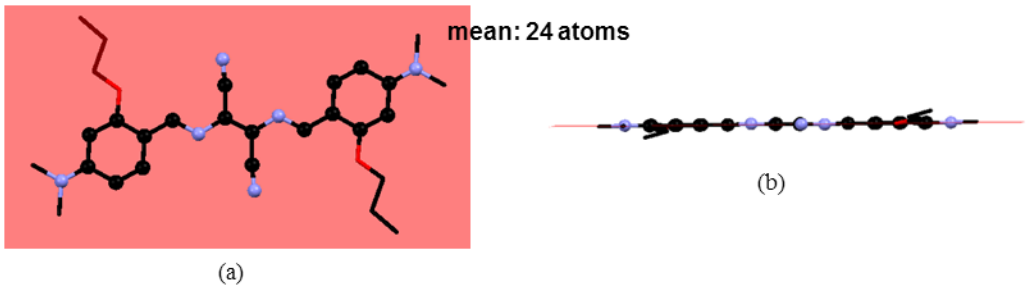

Figure S1 The molecular π-plane drawn by Mercury 3.1: (a) on the top view and (b) on the side view.

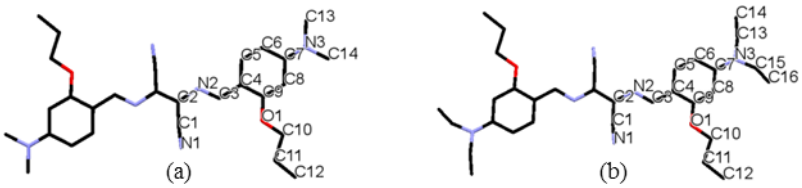

Figure S2 Atomic labels depicted by Mercury 3.1: (a) of **1c** and (b) of **2c**.

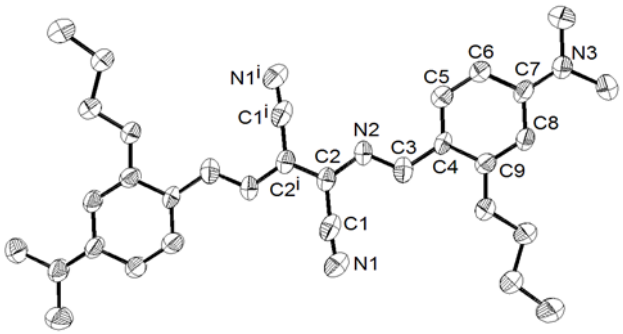

Figure S3 Atomic labels depicted by ORTEP-3 of the component atoms of the π-conjugated system.

Table S1 Deviation of the atoms from the molecular π-plane in the crystalline states of **1a**–**1f** and **2c**–**2f**.

1a	1b	1c	1d	1e	1f	2c	2d	2e	2f
Deviation of the component atoms of the π-conjugationsytem from the π-plane/ Å									
N1 0.029	0.064	0.002	0.008	0.017	0.011	0.016	0.015	0.019	0.026
C1 0.015	0.019	0.011	0.013	0.023	0.005	0.024	0.023	0.018	0.029
C2 0.019	0.001	0.010	0.009	0.004	0.010	0.049	0.038	0.045	0.005
N2 0.009	0.031	0.032	0.012	0.025	0.002	0.120	0.092	0.111	0.048
C3 0.061	0.024	0.014	0.033	0.010	0.048	0.143	0.127	0.149	0.018
C4 0.021	0.003	0.009	0.003	0.018	0.036	0.113	0.110	0.132	0.048
C5 0.101	0.037	0.002	0.005	0.016	0.055	0.178	0.162	0.197	0.032
C6 0.066	0.033	0.002	0.021	0.032	0.043	0.122	0.098	0.120	0.004
C7 0.024	0.000	0.014	0.003	0.004	0.014	0.055	0.058	0.066	0.027
C8 0.059	0.045	0.009	0.006	0.021	0.015	0.093	0.091	0.089	0.002
C9 0.008	0.046	0.017	0.007	0.013	0.013	0.021	0.022	0.033	0.047
N3 0.065	0.024	0.012	0.021	0.000	0.082	0.215	0.175	0.236	0.070
Deviation of the component atoms of the alkoxy substituents from the π-plane/ Å									
O1 0.003	O1 0.102	O1 0.018	O1 0.021	O1 0.007	O1 0.010	O1 0.040	O1 0.056	O1 0.055	O1 0.109
C10 0.470	C10 0.318	C10 0.242	C10 0.098	C10 0.299	C10 0.075	C10 0.110	C10 0.140	C10 0.159	C10 0.081
C11 1.757		C11 0.315	C11 0.011	C11 0.548	C11 0.125	C11 0.288	C11 0.372	C11 0.325	C11 0.213
C12 2.171		C12 0.631	C12 1.099	C12 1.700	C12 0.045	C12 0.298	C12 0.404	C12 0.468	C12 0.894
C13 1.348			C13 0.903	C13 1.969	C13 0.044		C13 0.681	C13 0.698	C13 0.810
C14 0.078				C14 3.088	C14 0.091			C14 0.999	C14 1.859
C15 0.399					C15 0.105				C15 1.789
					C16 0.245				C16 2.823
					C17 0.238				C17 2.738
					C18 0.355				C18 3.696
					C19 0.337				C19 3.609
Deviation of the carbon atoms of the dialkyl substituents on the terminal amino groups from the π-plane/ Å									
C16 0.04	C11 0.06	C13 0.01	C14 0.01	C15 0.06	C20 0.11	C13 0.13	C14 0.18	C15 0.11	C20 0.06
C17 0.04	C12 0.01	C14 0.03	C15 0.02	C16 0.14	C21 0.01	C14 1.01	C15 0.90	C16 1.02	C21 1.25
						C15 0.76	C16 0.81	C17 0.73	C22 0.36
						C16 0.33	C17 0.30	C18 0.36	C23 0.90

Table S2 Intramolecular bond angles and bond distances on the π-conjugated system in the crystalline states of **1a**–**1f** and **2c**–**2f**.

	1a	**1b**	**1c**	**1d**	**1e**	**1f**	Mean deviation in **1a - 1f**	**2c**	**2d**	**2e**	**2f**	Mean deviation in **1a - 2f**
Bond angles/ °												
∠N3C7C6	121.70 (8)	121.10 (3)	122.70 (9)	121.70 (4)	121.32 (18)	121.55 (13)	0.36	122.70 (5)	121.60 (5)	121.19 (18)	121.36 (15)	0.41
∠N3C7C8	119.50 (9)	120.90 (3)	120.50 (7)	121.10 (4)	121.14 (18)	120.34 (13)	0.47	120.70 (5)	121.00 (5)	121.16 (18)	120.71 (17)	0.36
∠C6C7C8	118.80 (8)	118.00 (3)	116.70 (9)	117.20 (3)	117.50 (2)	118.10 (13)	0.58	116.60 (5)	117.40 (5)	117.64 (18)	117.92 (19)	0.51
∠C5C6C7	120.20 (8)	120.00 (3)	121.90 (9)	121.40 (4)	120.67 (17)	120.07 (14)	0.63	121.20 (5)	120.90 (5)	120.10 (19)	119.97 (15)	0.57
∠C4C5C6	122.60 (7)	123.10 (3)	122.00 (8)	122.20 (4)	122.68 (17)	122.58 (14)	0.28	122.20 (5)	123.30 (6)	122.81 (19)	123.17 (17)	0.35
∠C5C4C9	115.70 (7)	116.40 (3)	116.50 (9)	116.90 (3)	116.50 (2)	117.17 (13)	0.34	117.30 (5)	116.30 (5)	117.00 (17)	116.52 (18)	0.37
∠C4C9C8	122.60 (8)	121.70 (3)	122.50 (8)	121.90 (4)	121.44 (16)	121.15 (13)	0.45	120.60 (5)	120.30 (5)	120.78 (17)	121.28 (14)	0.60
∠C7C8C9	120.00 (8)	120.80 (3)	120.30 (8)	120.40 (4)	121.09 (17)	120.89 (13)	0.35	121.90 (5)	121.60 (5)	121.47 (18)	121.12 (17)	0.48
∠C3C4C5	121.50 (7)	122.10 (2)	122.70 (7)	122.10 (3)	122.57 (17)	121.99 (13)	0.32	122.30 (5)	122.90 (5)	122.41 (17)	122.06 (16)	0.31
∠C3C4C9	122.60 (8)	121.50 (3)	120.70 (9)	121.00 (4)	120.82 (16)	120.84 (13)	0.54	120.30 (5)	120.70 (5)	120.59 (17)	121.39 (14)	0.47
∠N2C3C4	121.60 (7)	121.80 (3)	123.10 (8)	122.10 (4)	122.55 (17)	121.65 (14)	0.46	121.20 (5)	121.70 (5)	122.36 (18)	121.86 (14)	0.43
∠C2N2C3	119.70 (7)	120.30 (3)	121.80 (7)	120.00 (4)	119.36 (16)	119.80 (13)	0.59	119.80 (5)	120.00 (5)	119.77 (17)	119.78 (13)	0.41
∠N2C2C2^{i}	118.40 (10)	121.20 (3)	122.10 (12)	120.20 (4)	120.87 (16)	120.68 (17)	0.85	120.00 (5)	119.50 (5)	120.62 (18)	120.58 (13)	0.71
∠N2C2C1	122.20 (6)	121.50 (2)	120.70 (6)	122.00 (3)	121.11 (16)	121.72 (13)	0.44	122.50 (5)	121.30 (5)	120.77 (17)	121.45 (16)	0.46
∠C1C2C2^{i}	119.40 (10)	117.30 (3)	117.00 (12)	117.80 (3)	118.02 (19)	117.59 (16)	0.57	117.50 (6)	119.20 (5)	118.57 (18)	117.94 (17)	0.61
Bond distances/ Å												
N3—C7	1.366 (7)	1.365 (4)	1.336 (9)	1.361 (5)	1.358 (4)	1.364 (19)	0.008	1.365 (8)	1.351 (8)	1.371 (2)	1.365 (3)	0.007
C6—C7	1.400 (8)	1.415 (4)	1.412 (9)	1.407 (5)	1.416 (3)	1.420 (2)	0.005	1.408 (7)	1.396 (8)	1.409 (3)	1.413 (2)	0.006
C5—C6	1.360 (7)	1.366 (5)	1.354 (9)	1.366 (5)	1.362 (4)	1.373 (2)	0.005	1.369 (8)	1.353 (9)	1.367 (3)	1.371 (3)	0.005
C4—C5	1.404 (7)	1.402 (4)	1.400 (9)	1.399 (6)	1.405 (3)	1.406 (2)	0.002	1.385 (7)	1.388 (9)	1.395 (2)	1.398 (2)	0.005
C4—C9	1.416 (8)	1.412 (3)	1.406 (9)	1.406 (5)	1.411 (3)	1.415 (2)	0.003	1.409 (7)	1.425 (8)	1.406 (2)	1.416 (2)	0.005
C8—C9	1.354 (8)	1.379 (5)	1.367 (9)	1.382 (5)	1.381 (4)	1.385 (2)	0.009	1.378 (8)	1.377 (8)	1.377 (2)	1.379 (2)	0.006
C7—C8	1.404 (8)	1.406 (4)	1.419 (9)	1.422 (6)	1.405 (3)	1.412 (2)	0.006	1.403 (7)	1.402 (8)	1.403 (2)	1.408 (2)	0.006
C3—C4	1.429 (8)	1.423 (5)	1.433 (9)	1.425 (5)	1.429 (4)	1.438 (2)	0.004	1.423 (8)	1.439 (8)	1.433 (2)	1.432 (2)	0.005
N2—C3	1.307 (7)	1.294 (3)	1.296 (8)	1.294 (5)	1.295 (3)	1.295 (2)	0.003	1.313 (7)	1.302 (7)	1.297 (2)	1.298 (2)	0.005
N2—C2	1.376 (8)	1.376 (4)	1.366 (9)	1.378 (4)	1.377 (3)	1.381 (19)	0.003	1.381 (8)	1.384 (7)	1.383 (2)	1.378 (2)	0.003
C1—C2	1.435 (9)	1.446 (5)	1.462 (12)	1.440 (6)	1.448 (3)	1.449 (2)	0.006	1.445 (10)	1.445 (10)	1.448 (3)	1.443 (2)	0.005
N1—C1	1.129 (7)	1.133 (5)	1.144 (10)	1.131 (7)	1.133 (3)	1.148 (2)	0.006	1.144 (9)	1.131 (10)	1.139 (3)	1.140 (2)	0.006
C2—C2^{i}	1.377 (12)	1.375 (3)	1.340 (13)	1.380 (5)	1.369 (3)	1.371 (3)	0.010	1.379 (8)	1.353 (8)	1.363 (2)	1.372 (2)	0.010

Table S3 Results of the lattice energy calculations in **1c**–**1f** and **2c**–**2f** focusing on the 3rd–4th and 5th–6th.

		Contribution of molecular pairs to the total lattice energies					
	Total lattice enegy/ kJ mol^{-1}	3rd and 4th contributions/ kJ mol^{-1} (ratio, ADC)		5th and 6th contributions/ kJ mol^{-1} (ratio, ADC)		Enregy gap between 3rd - 4th and 5th - 6th contributions/ kJ mol^{-1} (ratio)	
1c	-508.0	-47.2	(9.3%, 45601 and 65401)	-40.3	(7.9%, 44501 and 66501)	-6.9	(1.4%)
1d	-528.2	-50.2	(9.5%, 44501 and 66501)	-32.6	(6.2%, 34401 and 76601)	-17.6	(3.3%)
1e	-537.3	-49.5	(9.2%, 45401 and 65601)	-41.6	(7.7%, 46501 and 64501)	-7.9	(1.5%)
1f	-706.9	-64.7	(9.2%, 54501 and 56501)	-50.4	(7.1%, 46401 and 64601)	-14.3	(2.0%)
2c[a]	-574.4	-86.5	(15.1%, 44501 and 66501)	-15.0	(2.6%, 43501 and 67501)	-71.5	(12.4%)
2d	-592.9	-85.9	(14.5%, 45401 and 65601)	-17.6	(3.0%, 35401 and 75601)	-68.3	(11.5%)
2e[a]	-596.4	-88.6	(14.9%, 45601 and 65401)	-21.4	(3.6%, 45701 and 65301)	-67.2	(11.3%)
2f[a]	-724.1	-62.6	(8.6%, 46401 and 64601)	-37.6	(5.2%, 44501 and 66501)	-25	(3.5%)

[a] The results of the lattice energy calculations of **2c**, **2e** and **2f** have been reported previously.[30]