5^o

The displacement ellipsoids were drawn at the 50% probability level. The molecule sits on a crystallographic center of symmetry. There are two orientations with occupancies of 0.833(3) for the unprimed atoms and 0.167(3) for the primed atoms. The unprimed atoms are not shown for clarity. The unprimed atoms were refined with isotropic displacement parameters.

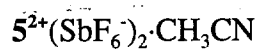
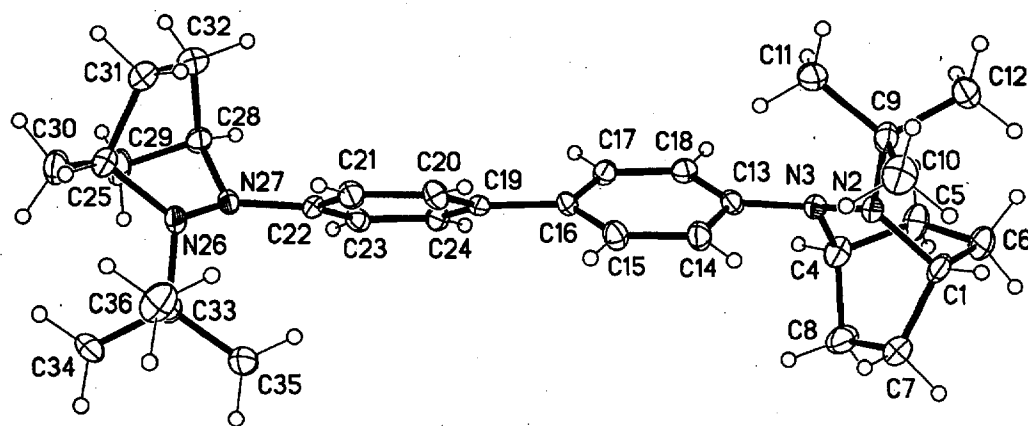
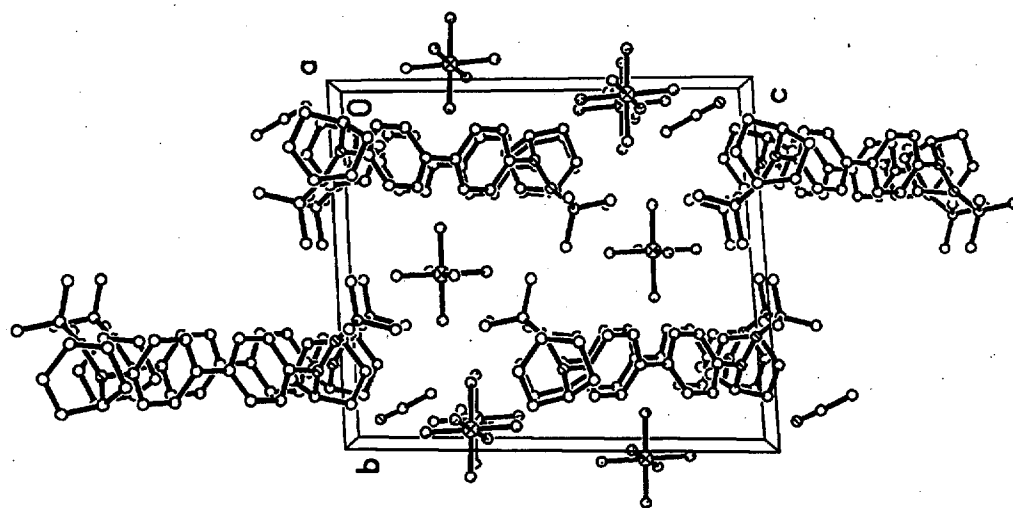
structure ID number	5 ⁰
empirical formula	C ₃₂ H ₄₆ N ₄
temperature, K	133(2)
space group	P2 ₁ /c
Z	2
a, Å	6.3483(2)
b, Å	13.8599(2)
c, Å	15.9484(3)
a, deg	90
b, deg	99.623(2)
g, deg	90
Volume, Å ³	13.83.50(5)
Density, (calcd), Mg/m ³	1.168
F(000)	532
Crystal size, mm	0.50 × 0.35 × 0.03
Reflections collected	5331
Independent reflections ^a	2387(0.0279)
Final R index ^b	0.0670
wR index (all data)	0.1580
Goodness-of-fit on F ²	1.210
Data / restraints / parameters	2386 / 62 (disorder) / 237
Largest dif. Map peaks, eÅ ⁻³	0.410 / -0.241

^a In parenthesis: R_{int}.

^b R₁ [I > 2s(I)]. Full-matrix least-squares refinement on F².

⁵ Atomic coordinates [$\times 10^4$] and equivalent isotropic displacement parameters [$\text{\AA}^2 \times 10^3$] for 1. U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x	y	z	U(eq)
C(1)	4573(5)	2812(2)	2319(2)	33(1)
N(2)	3784(4)	2948(2)	3158(2)	27(1)
N(3)	1624(4)	3309(2)	3014(2)	27(1)
C(4)	741(5)	3367(2)	2103(2)	32(1)
C(5)	840(6)	2351(3)	1753(2)	40(1)
C(6)	3204(10)	2056(3)	1797(3)	41(4)
C(7)	4382(5)	3770(2)	1844(2)	38(1)
C(8)	1993(7)	4022(3)	1599(2)	34(1)
C(9)	4038(5)	2090(2)	3668(2)	25(1)
C(10)	2385(5)	1521(2)	3841(2)	27(1)
C(11)	2775(6)	716(3)	4347(2)	28(1)
C(12)	4813(13)	436(6)	4726(10)	24(2)
C(13)	6452(6)	1019(2)	4538(2)	35(1)
C(14)	6082(6)	1826(2)	4026(2)	35(1)
C(15)	1559(5)	4202(2)	3564(2)	29(1)
C(16)	3148(7)	5001(3)	3477(3)	40(1)
C(17)	1902(13)	3857(3)	4489(2)	41(1)
C(18)	-717(6)	4584(2)	3356(2)	34(1)
C(1')	1197(18)	2318(7)	2174(6)	46(6)
N(2')	1805(13)	2683(5)	3078(5)	22(3)
N(3')	3154(15)	3515(5)	3109(5)	23(3)
C(4')	3639(17)	3758(7)	2262(6)	37(4)
C(5')	4776(19)	2885(11)	1975(10)	56(8)
C(6')	3217(30)	2020(8)	1845(9)	27(18)
C(7')	81(17)	3120(10)	1619(7)	36(4)
C(8')	1682(27)	3953(9)	1595(8)	118(23)
C(9')	2667(19)	1922(6)	3646(7)	23(4)
C(10')	1537(17)	1091(8)	3753(9)	32(4)
C(11')	2426(21)	377(9)	4292(10)	22(5)
C(12')	4543(51)	396(33)	4699(52)	24(13)
C(13')	5614(22)	1255(10)	4610(11)	46(6)
C(14')	4730(19)	1983(9)	4063(10)	33(5)
C(15')	2287(28)	4296(7)	3623(9)	176(26)
C(16')	3339(40)	5286(8)	3629(16)	50(8)
C(17')	2486(59)	3930(15)	4535(9)	61(14)
C(18')	-87(25)	4379(18)	3267(23)	163(25)



The displacement ellipsoids were drawn at the 50% probability level.

structure ID number	$5^{2+}(\text{SbF}_6)_2 \cdot \text{CH}_3\text{CN}$
empirical formula	$\text{C}_{32}\text{H}_{46}\text{N}_4(\text{SbF}_6)_2 \times \text{CH}_3\text{CN}$
temperature, K	133(2)
space group	P1(bar)
Z	2
a, Å	8.0798(2)
b, Å	14.5574(2)
c, Å	16.8708(2)
a, deg	85.297(2)
b, deg	82.283(2)
g, deg	79.876(2)
Volume, Å ³	1932.30(6)
Density, (calcd), Mg/m ³	1.717
F(000)	996
Crystal size, mm	0.42 × 0.18 × 0.12
Reflections collected	10995
Independent reflections ^a	7514(0.0212)
Final R index ^b	0.0502
wR index (all data)	0.1194
Goodness-of-fit on F ²	1.265
Data / restraints / parameters	7513 / 0 / 478
Largest dif. Map peaks, eÅ ⁻³	1.105 / -0.811

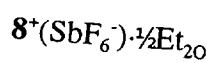
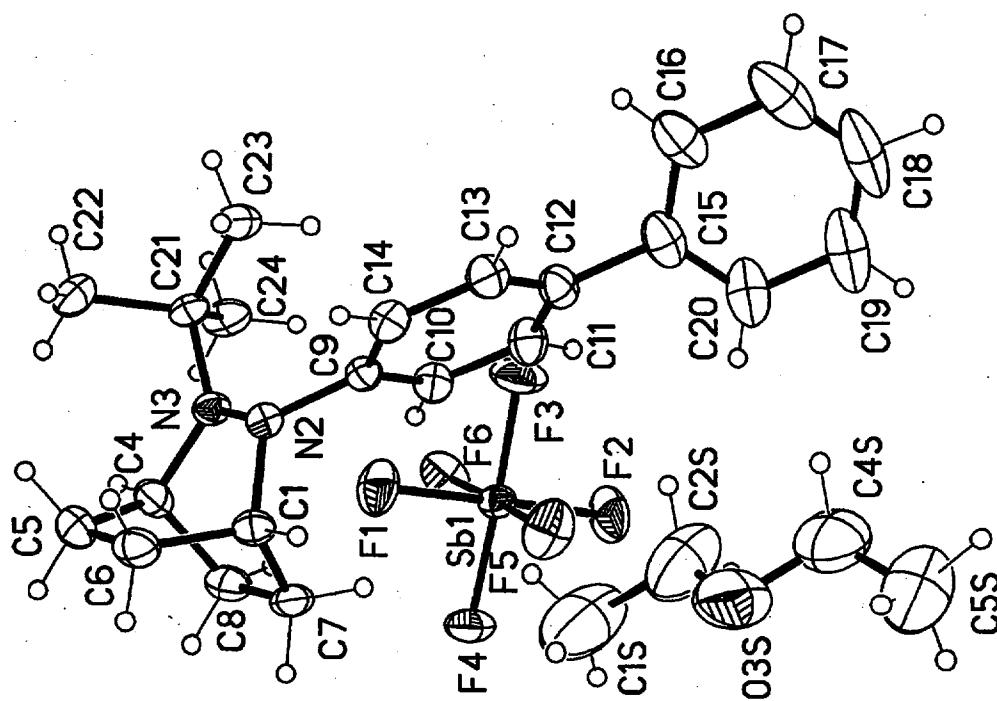
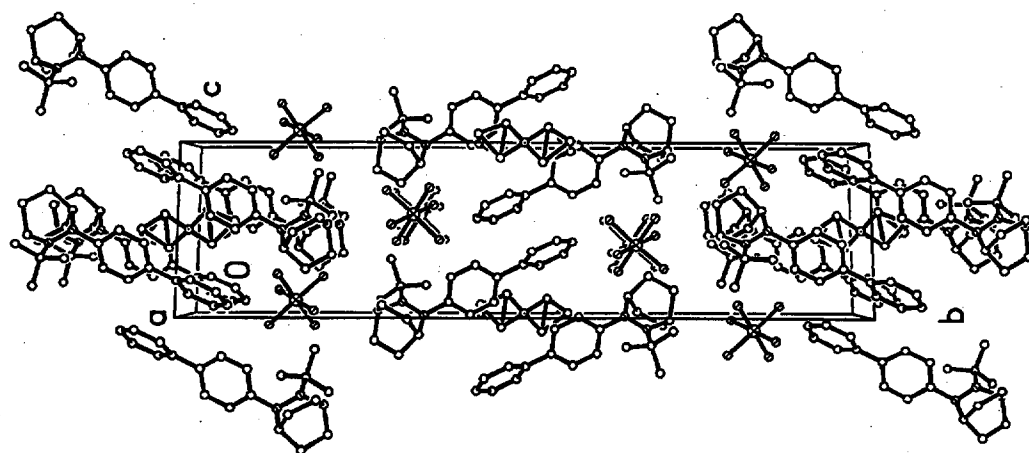
^a In parenthesis: R_{int} .

^b R_1 [$I > 2s(I)$]. Full-matrix least-squares refinement on F^2 .

$5^{2+}(\text{SbF}_6)_2 \cdot \text{CH}_3\text{CN}$ Atomic coordinates [$\times 10^4$] and equivalent isotropic displacement parameters [$\text{\AA}^2 \times 10^3$] for 1. $U(\text{eq})$ is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x	y	z	U(eq)
Sb(1A)	7414(1)	5327(1)	2429(1)	21(1)
F(1A)	9526(5)	5355(3)	2759(3)	47(1)
F(2A)	5314(5)	5259(3)	2105(3)	43(1)
F(3A)	7917(7)	4036(3)	2470(4)	67(2)
F(4A)	6853(9)	6605(3)	2383(5)	105(3)
F(5A)	8342(7)	5369(6)	1365(3)	94(2)
F(6A)	6493(7)	5259(5)	3498(3)	84(2)
Sb(1B)	8705(1)	9496(1)	2935(1)	23(1)
F(1B)	8896(6)	10761(3)	2866(2)	41(1)
F(2B)	8471(6)	8238(3)	2998(3)	47(1)
F(3B)	6389(5)	9809(3)	3282(2)	43(1)
F(4B)	11014(6)	9176(4)	2615(3)	60(1)
F(5B)	8259(6)	9598(3)	1864(2)	39(1)
F(6B)	9148(6)	9392(3)	4009(2)	47(1)
C(1)	-4249(7)	3013(4)	5269(4)	24(1)
N(2)	-2387(6)	2971(3)	5165(3)	19(1)
N(3)	-1538(6)	2187(3)	4818(3)	17(1)
C(4)	-2645(8)	1498(4)	4729(4)	25(1)
C(5)	-3566(8)	1286(5)	5552(4)	31(2)
C(6)	-4762(8)	2188(5)	5825(4)	30(1)
C(7)	-4733(8)	2938(5)	4429(4)	29(1)
C(8)	-3891(8)	1962(5)	4139(4)	33(2)
C(9)	-1553(7)	3534(4)	5673(3)	21(1)
C(10)	-2105(9)	4571(4)	5431(4)	31(1)
C(11)	379(7)	3266(4)	5563(4)	24(1)
C(12)	-2216(8)	3316(5)	6556(4)	28(1)
C(13)	-116(7)	2226(4)	4224(3)	18(1)
C(14)	2(7)	3037(4)	3731(3)	21(1)
C(15)	1342(7)	3039(4)	3127(3)	20(1)
C(16)	2551(7)	2229(4)	2987(3)	19(1)
C(17)	2396(7)	1425(4)	3491(3)	19(1)
C(18)	1088(7)	1419(4)	4112(3)	20(1)
C(19)	3954(7)	2223(4)	2321(3)	17(1)
C(20)	4706(8)	3017(4)	2103(3)	22(1)
C(21)	6039(7)	2991(4)	1495(3)	22(1)
C(22)	6607(7)	2189(4)	1084(3)	18(1)
C(23)	5842(7)	1400(4)	1275(3)	18(1)
C(24)	4535(7)	1423(4)	1897(3)	18(1)
C(25)	10312(7)	2899(4)	-49(4)	24(1)
N(26)	8470(6)	2890(3)	63(3)	19(1)
N(27)	8054(6)	2118(3)	489(3)	16(1)
C(28)	9544(7)	1407(4)	641(4)	23(1)
C(29)	10564(8)	1187(5)	-170(4)	30(1)
C(30)	11286(8)	2060(4)	-526(4)	28(1)
C(31)	10842(8)	2791(5)	798(4)	30(1)
C(32)	10566(9)	1824(5)	1183(4)	35(2)

C(33)	7321(7)	3458(4)	-511(3)	23(1)
C(34)	8095(8)	3208(5)	-1368(3)	29(1)
C(35)	5524(8)	3246(4)	-369(4)	25(1)
C(36)	7315(8)	4498(4)	-405(4)	32(2)
N(1S)	3527(10)	9392(5)	677(4)	52(2)
C(1S)	4084(9)	9069(5)	1237(4)	32(2)
C(2S)	4735(9)	8631(5)	1965(4)	36(2)



The displacement ellipsoids were drawn at the 50% probability level. The diethyl ether molecule is disordered because it is sitting on a crystallographic center of symmetry.

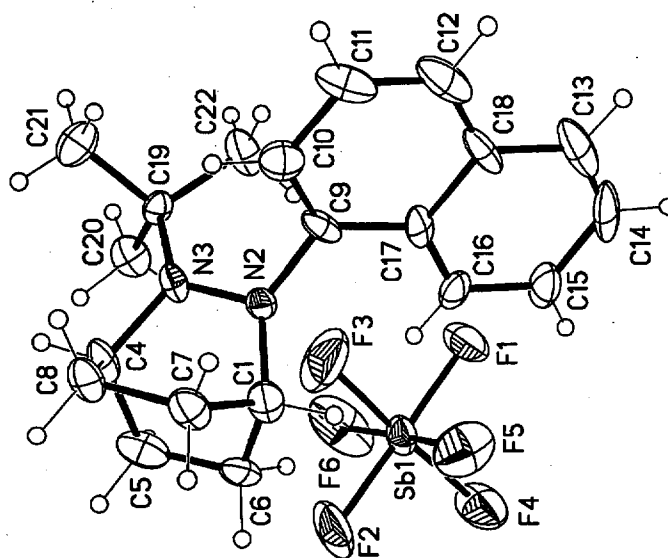
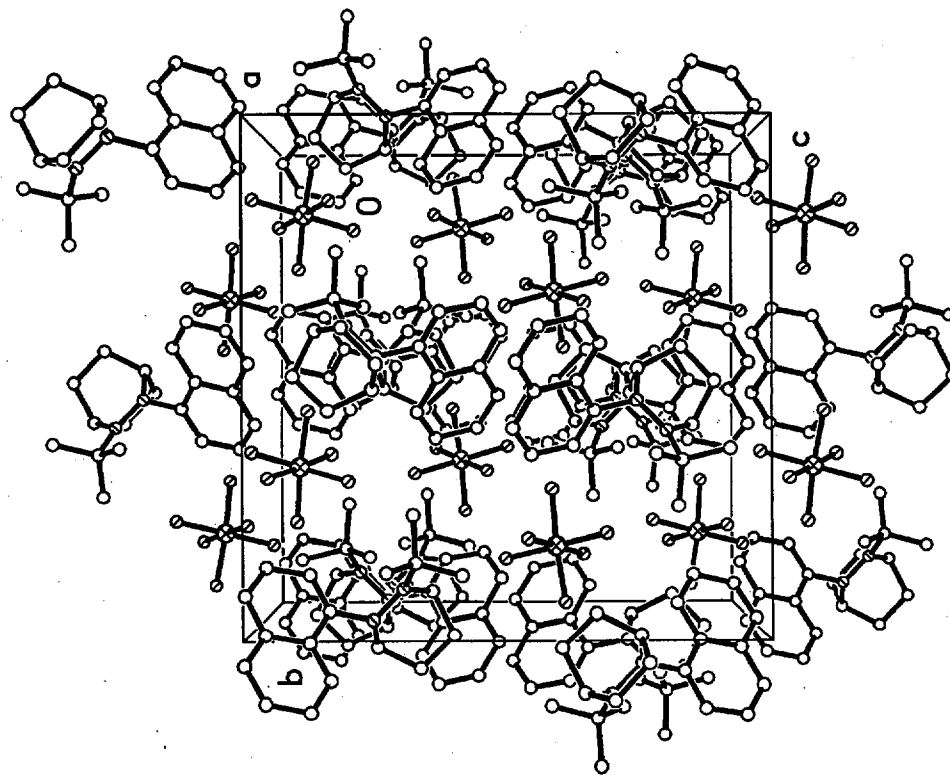
structure ID number	$8^+(\text{SbF}_6^-) \cdot \frac{1}{2}\text{Et}_2\text{O}$
empirical formula	$\text{C}_{22}\text{H}_{28}\text{N}_2\text{SbF}_6 \times 0.5(\text{C}_4\text{H}_{10}\text{O})$
temperature, K	133(2)
space group	$\text{P2}_1/\text{n}$
Z	4
a, Å	8.4725(2)
b, Å	34.6189(5)
c, Å	9.0699(2)
a, deg	90
b, deg	104.438
g, deg	90
Volume, Å ³	2576.26(9)
Density, (calcd), Mg/m ³	1.530
F(000)	1200
Crystal size, mm	0.42 × 0.18 × 0.14
Reflections collected	12308
Independent reflections ^a	5917(0.0244)
Final R index ^b	0.0751
wR index (all data)	0.0362
Goodness-of-fit on F ²	1.082
Data / restraints / parameters	5915/42 (Et ₂ O)/327
Largest dif. Map peaks, eÅ ⁻³	0.594 / -0.863

^a In parenthesis: R_{int} .

^b R_1 [$I > 2s(I)$]. Full-matrix least-squares refinement on F^2 .

$8^+(\text{SbF}_6^-) \cdot \frac{1}{2}\text{Et}_{20}$ Atomic coordinates and equivalent isotropic displacement parameters [\AA^2] for 1. $U(\text{eq})$ is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x	y	z	U(eq)
Sb(1)	0.01857(2)	0.660994(5)	0.41501(2)	0.02580(6)
F(1)	0.1756(2)	0.68697(6)	0.5649(2)	0.0496(5)
F(2)	-0.1379(3)	0.63679(6)	0.2614(2)	0.0546(5)
F(3)	0.1847(2)	0.63332(6)	0.3610(3)	0.0563(6)
F(4)	-0.1426(2)	0.69072(6)	0.4710(2)	0.0514(5)
F(5)	0.0023(3)	0.62305(6)	0.5564(2)	0.0550(5)
F(6)	0.0371(3)	0.70048(5)	0.2775(2)	0.0466(5)
C(1)	0.4011(3)	0.65522(8)	1.1076(3)	0.0262(6)
N(2)	0.5061(3)	0.64898(6)	0.9989(2)	0.0225(4)
N(3)	0.5294(3)	0.68152(6)	0.9243(2)	0.0234(4)
C(4)	0.4511(3)	0.71597(8)	0.9714(3)	0.0281(6)
C(5)	0.5182(4)	0.72172(9)	1.1448(3)	0.0365(7)
C(6)	0.4849(4)	0.68496(9)	1.2264(3)	0.0335(6)
C(7)	0.2364(3)	0.67076(9)	1.0140(3)	0.0306(6)
C(8)	0.2666(3)	0.70836(9)	0.9361(3)	0.0315(6)
C(9)	0.4873(3)	0.61233(8)	0.9214(3)	0.0234(5)
C(10)	0.3809(3)	0.60854(8)	0.7777(3)	0.0278(6)
C(11)	0.3596(4)	0.57247(9)	0.7085(3)	0.0328(6)
C(12)	0.4453(4)	0.54030(8)	0.7789(3)	0.0299(6)
C(13)	0.5516(3)	0.54512(8)	0.9241(3)	0.0295(6)
C(14)	0.5716(3)	0.58049(8)	0.9961(3)	0.0273(6)
C(15)	0.4232(4)	0.50148(9)	0.7040(3)	0.0397(7)
C(16)	0.5549(5)	0.47750(10)	0.7106(4)	0.0498(9)
C(17)	0.5328(7)	0.44087(11)	0.6414(5)	0.0649(13)
C(18)	0.3789(8)	0.42901(12)	0.5673(5)	0.076(2)
C(19)	0.2489(7)	0.45242(12)	0.5589(5)	0.078(2)
C(20)	0.2693(5)	0.48897(11)	0.6258(4)	0.0596(11)
C(21)	0.6607(3)	0.68604(8)	0.8377(3)	0.0261(6)
C(22)	0.7824(3)	0.71670(9)	0.9208(4)	0.0362(7)
C(23)	0.7559(4)	0.64862(9)	0.8335(4)	0.0355(7)
C(24)	0.5755(4)	0.69938(10)	0.6764(3)	0.0359(7)
C(1S)	0.021(2)	0.5673(2)	0.951(2)	0.132(6)
C(2S)	0.0267(13)	0.5268(2)	0.8899(9)	0.098(3)
O(3S)	0.0000	0.5000	1.0000	0.090(2)
C(4S)	-0.0070(11)	0.4649(2)	0.9144(10)	0.084(3)
C(5S)	-0.076(2)	0.4335(2)	0.995(2)	0.100(5)



9+(SbF₆⁻)

The displacement ellipsoids were drawn at the 50% probability level.

structure ID number	9 ⁺ (SbF ₆)
empirical formula	C ₂₀ H ₂₆ N ₂ SbF ₆
temperature, K	133(2)
space group	Pbca
Z	8
a, Å	15.817(2)
b, Å	16.372(2)
c, Å	16.496(2)
a, deg	90
b, deg	90
g, deg	90
Volume, Å ³	4271.8(8)
Density, (calcd), Mg/m ³	1.649
F(000)	2120
Crystal size, mm	0.42 × 0.14 × 0.01
Reflections collected	15954
Independent reflections ^a	3766(0.0814)
Final R index ^b	0.0618
wR index (all data)	0.1517
Goodness-of-fit on F ²	1.018
Data / restraints / parameters	3753 / 0 / 262
Largest dif. Map peaks, eÅ ⁻³	1.167 / -1.310

^a In parenthesis: R_{int}.

^b R₁ [I > 2s(I)]. Full-matrix least-squares refinement on F².

$9^+(\text{SbF}_6^-)$ Atomic coordinates and equivalent isotropic displacement parameters [\AA^2] for 1. $U(\text{eq})$ is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x	y	z	U(eq)
Sb(1)	0.68542 (3)	0.32770 (3)	0.59622 (3)	0.0352 (2)
F(1)	0.7853 (3)	0.3500 (3)	0.6520 (3)	0.054 (2)
F(2)	0.5844 (4)	0.3084 (4)	0.5410 (4)	0.093 (3)
F(3)	0.6530 (5)	0.4369 (4)	0.6137 (5)	0.091 (2)
F(4)	0.7144 (4)	0.2179 (4)	0.5885 (4)	0.076 (2)
F(5)	0.6313 (4)	0.3002 (4)	0.6933 (3)	0.072 (2)
F(6)	0.7329 (4)	0.3554 (5)	0.4981 (3)	0.096 (3)
C(1)	0.4156 (5)	0.4810 (5)	0.7491 (5)	0.031 (2)
N(2)	0.4857 (4)	0.5406 (3)	0.7539 (3)	0.0207 (13)
N(3)	0.4810 (4)	0.5992 (4)	0.6966 (3)	0.0233 (14)
C(4)	0.4069 (5)	0.5894 (5)	0.6433 (4)	0.028 (2)
C(5)	0.4103 (5)	0.5050 (5)	0.6019 (5)	0.037 (2)
C(6)	0.4187 (5)	0.4402 (5)	0.6662 (5)	0.032 (2)
C(7)	0.3317 (5)	0.5263 (5)	0.7598 (5)	0.031 (2)
C(8)	0.3270 (4)	0.5941 (5)	0.6964 (4)	0.030 (2)
C(9)	0.5181 (5)	0.5601 (5)	0.8346 (4)	0.024 (2)
C(10)	0.4879 (5)	0.6270 (5)	0.8734 (5)	0.032 (2)
C(11)	0.5188 (5)	0.6430 (6)	0.9532 (5)	0.041 (2)
C(12)	0.5756 (6)	0.5929 (6)	0.9886 (5)	0.046 (2)
C(13)	0.6671 (5)	0.4694 (7)	0.9828 (5)	0.051 (3)
C(14)	0.6967 (6)	0.4036 (7)	0.9419 (6)	0.056 (3)
C(15)	0.6668 (5)	0.3839 (5)	0.8618 (6)	0.041 (2)
C(16)	0.6087 (5)	0.4357 (5)	0.8274 (5)	0.034 (2)
C(17)	0.5767 (4)	0.5040 (5)	0.8685 (4)	0.026 (2)
C(18)	0.6069 (5)	0.5222 (6)	0.9487 (4)	0.034 (2)
C(19)	0.5544 (4)	0.6530 (4)	0.6713 (4)	0.021 (2)
C(20)	0.5774 (5)	0.6311 (5)	0.5839 (4)	0.032 (2)
C(21)	0.5243 (5)	0.7415 (5)	0.6767 (5)	0.037 (2)
C(22)	0.6321 (5)	0.6395 (5)	0.7235 (5)	0.035 (2)

Experimental

1,4-bis(2-tert-butyl-2,3-diazabicyclo[2.2.2]oct-3-yl)-naphthalene (4). 1,4-dibromonaphthalene was dried in a vacuum oven at ca. 100 °C for 1 hr and then dissolved (143 mg, 0.5 mmole) in ether (6 mL) in an oven-dried 50 mL Schlenk flask. The mixture was cooled to -78 °C and t-BuLi (1.90 M in pentane, 1.05 mL, 2 mmole) was added dropwise. After stirring for 2 hrs, 2-t-Bu-2,3-diazabicyclo[2.2.2]oct-2-enium iodide (294 mg, 1.00 mmole) was added, and the cooling bath was gradually lowered and removed. The mixture was stirred overnight, quenched with water (25 mL) and extracted with toluene (2×30 mL) to give 228 mg of yellow solid upon evaporation. It was dissolved in boiling toluene (ca. 1 mL) and acetonitrile (10 mL) was added carefully in small portions to precipitate **4** as bright-yellow needles (165 mg, 72 %), m.p. 246 °C. The structure was confirmed by x-ray, but was disordered (conformations rotated 180° about the N-Ar bonds were present). The structural parameters are not of useful quality, and are not reported. MS: 460.3570 *m/e*, I = 18.1 %, calcd. for C₃₀H₄₄N₄ 460.3566. ¹H NMR (360 MHz, C₆D₆): δ 8.23 (m., 2H, ArH), 7.91 (s., 1H, ArH), 7.85 (s., 1H, ArH), 7.13 (m., 2H, ArH), 3.11 (m., 1H, NCH), 3.03 (m., 1H, NCH), 2.95 (m., 2H, NCH), 2.05 (m., 4H, CH₂), 1.62 (m., 2H, CH₂), 1.44 (m., 2H, CH₂), 1.18 (m., 4H, CH₂), 0.94 (m., 2H, CH₂), 0.86 (s., 9 H, C(CH₃)₃), 0.84 (s., 9 H, C(CH₃)₃), 0.63 (m., 2H, CH₂). ¹³C{¹H} NMR (90 MHz, C₆D₆): δ 146.46 (NC_{ar}), 129.37 and 129.32 (C_{ar}), 124.98 (C_{ar}H), 124.67 (C_{ar}H, double intensity), 124.45 (C_{ar}H), 119.89 (C_{ar}H), 118.93 (C_{ar}H), 59.13 and 59.05 (C(CH₃)₃), 53.25 and 53.15 (NCH), 46.50 (NCH), 29.61 and 29.59 (CH₂), 28.88 and 28.84 (C(CH₃)₃), 27.02 (CH₂), 23.29 (CH₂), 20.91 and 20.52 (CH₂). CV in CH₃CN with 0.1 M TBAP vs SCE on Au with FeCp*₂ as the internal standard: E°₁ = 0.103 V (ΔE_{1,pp} = 69 mV), E°₂ = 0.373 V (ΔE_{2,pp} = 77 mV), E°_{Fe} = -0.107 V (ΔE_{Fe,pp} = 69 mV).

4,4'-bis(2-tert-butyl-2,3-diazabicyclo[2.2.2]oct-3-yl)biphenyl (5). A 50 mL Schlenk flask was oven-dried and purged with N₂. 4,4'-dibromobiphenyl (352 mg, 1.13 mmole) was added and the flask was purged for another 15 minutes. Ether (7 mL) was added, the solution was cooled to -78°C and t-BuLi (1.74 M in pentane, 2.59 mL, 4.51 mmole) was added dropwise. The mixture was stirred for 20 minutes and 2-t-Bu-2,3-diazabicyclo[2.2.2]oct-2-enium iodide was added. The cooling bath was gradually lowered and removed. In 40 minutes the yellow color disappeared and a white gelatinous suspension formed. After stirring another hour, the mixture was poured into water (20 mL) and extracted with toluene (3 x 25 mL) to give 0.59 g of an off-white solid. It was dissolved in refluxing toluene (3 mL) and allowed to cool to give 348 mg (63%) of white crystals. The toluene solution was diluted with CH₃CN (15 mL) to give an additional 65 mg (12%) **5**, m.p. 258 °C. MS: *m/e* 486.3709 (calculated for C₃₂H₄₆N₄ 486.372246, I=13%). ¹H NMR (500 MHz, C₆D₆): δ 8.01 (d, J = 7.6 Hz, 2H, ArH), 7.62 (d, J = 7.6 Hz, 2H, ArH), 7.54 (d, J = 7.6 Hz, 2H, ArH), 6.96 (d, J = 7.6 Hz, 2H, ArH), 3.53 (br. s., 2H, NCH), 3.16 (br. s., 2H, NCH), 2.21 (qt., J = 10.7, 2.5 Hz, 2H, CH₂), 1.91 (m., 2H, CH₂), 1.72 (m., 4H, CH₂), 1.48 (br. t., J = 12.3 Hz, 2H, CH₂), 1.37 (br. t., J = 12.3 Hz, 2H, CH₂), 1.16 (s., 18H, C(CH₃)₃), 0.95 (m., 2H, CH₂). ¹³C NMR {¹H} (125 MHz, C₆D₆): δ 155.25 (q-C_{ar}), 134.11 (q-C_{ar}), 126.76 (C_{ar}H), 126.56 (C_{ar}H), 120.73 (C_{ar}H), 120.53 (C_{ar}H), 58.57 (C(CH₃)₃), 54.19 (NCH), 46.62 (NCH), 29.05 (C(CH₃)₃), 28.86 (CH₂), 26.65 (CH₂), 22.38 (CH₂), 21.51 (CH₂). CV in CH₃CN with 0.1 M TBAP vs SCE on Au with FeCp*₂ as the internal standard: E°₁ = 0.231 V (ΔE_{1,pp} = 69 mV, assumed), E°₂ = 0.323 V (ΔE_{2,pp} = 69 mV, assumed), E°_{Fe} = -0.113 V (ΔE_{Fe,pp} = 69 mV).

4,4'-diiodo-2,2',6,6'-tetramethyl-biphenyl. 2,2',6,6'-tetramethyl-benzidine^{14c} (0.996 g, 4.14 mmol) was dissolved in a 10% H₂SO₄ solution (0.97 g concentrated H₂SO₄ in 9.2 mL H₂O) and cooled

to 10°C. A NaNO₂ solution (0.631 g, 9.14 mmole in 6.7 mL H₂O) was slowly added dropwise while maintaining a temperature of 8 °C. This solution was then stirred for 30 minutes after which it was poured cold into a stirring solution of KI₃ (prepared by dissolving KI (13.81 g, 83.2 mmol) in H₂O (138.1 mL) with stirring and adding I₂ (21.17 g, 83.2 mmol) and stirring overnight). The reaction mixture was heated to reflux until evolution iodine vapor was observed. The solution was cooled to room temperature and sodium bisulfite was added until gas ceased evolving. The solution was neutral at this point and contained a red solid that was filtered and recrystallized from heptane to yield 719 mg (1.66 mmole, 37.6%) of product, mp = 199-200 °C (Lit. 199-200°C).^{14c} ¹H NMR (300 MHz, CDCl₃): δ 7.486 (s, 4H), 1.835 (s, 12H), consistent with the literature.²³

2,7-bis(2-tert-butyl-2,3-diazabicyclo[2.2.2]oct-3-yl)-9,9-dimethylfluorene (6). 2,7-dibromo-9,9-dimethyl-fluorene²⁴ (170 mg, 0.483 mmole) was placed into an oven dried 50 mL Schlenk flask under nitrogen. Ether (3 mL) was added and the mixture was cooled to -78 °C. t-BuLi (1.74M in pentane, 1.10 mL, 1.91 mmole) was added slowly, giving a raspberry-red solution. After stirring for 75 min, 2-t-Bu-2,3-diazabicyclo[2.2.2]oct-2-ene iodide (294 mg, 1.00 mmole) was added, the mixture stirred for 20 min, and the cooling bath was gradually lowered and then removed. The mixture was stirred overnight and quenched with water (20 mL), extracted with toluene (5 × 30 mL), dried with MgSO₄ and evaporated to give 0.35 g of viscous yellow oil. It was dissolved in toluene and purified by chromatography on neutral alumina to give 111 mg (44%) of a yellow solid. Sublimation at 200 °C, 0.05 mm. Hg gave pink-orange crystals, m.p. 253-254 °C. MS: *m/e* 526.4037, I = 43 %, calcd. for C₃₅H₅₀N₄ 526.40355. ¹H NMR (500 MHz, C₆D₆): δ 8.14 (br. s., 1H, ArH), 8.01 (br. s., 1H, ArH), 7.59 (br. m., 1H, ArH), 7.51 (br. m., 1H, ArH), 7.09 (br. s., 1H, ArH), 6.93 (br. m., 1H, ArH), 3.65 and 3.57 (br. s., 2H, NCH), 3.24 and 3.20 (br. s., 2H, NCH), 2.26 (m., 2H, CH₂), 2.03 (m., 2H, CH₂), 1.77 (m., 4H, CH₂), 1.53 (m., 4H, CH₂), 1.50 - 1.40 (br.s., 6H, ArCH₃), 1.20 (s., 18H, C(CH₃)₃), 1.18 - 1.07 (m., 2H, CH₂), 0.99 (m., 2H, CH₂). ¹³C{¹H} NMR (125 MHz, C₆D₆): δ 155.48 and 155.41 (C_{ar}), 154.48 and 154.31 and 153.97 and 153.81 (C_{ar}), 133.80 and 133.68 and 133.30 and 133.18 (C_{ar}), 119.58 and 119.45 and 119.41 and 119.36 and 119.24 and 119.09 and 118.81 and 118.66 (C_{ar}H), 115.08 and 115.01 and 114.60 and 114.53 and 114.45 (C_{ar}H), 58.76 and 59.66 (C(CH₃)₃), 54.82 and 54.69 (NCH), 46.69 and 46.58 (NCH), 29.12 and 28.99 (CH₂), 29.07 (C(CH₃)₃), 28.03 and 27.72 and 27.66 and 27.33 (CH₃), 26.89 and 26.76 (CH₂), 22.54 (CH₂), 21.44 and 21.36 (CH₂). The quaternary carbon bridging the two aromatic rings was not identified, possibly due to overlap with other signals. CV in CH₃CN with 0.1 M TBAP vs SCE on Au with FeCp*₂ as the internal standard: E₁^o = 0.181 V (ΔE_{1,pp} = 69 mV), E₂^o = 0.300 V (ΔE_{2,pp} = 75 mV), E_{Fe}^o = -0.106 V (ΔE_{Fe,pp} = 68 mV).

2-t-Bu-3-(4-biphenyl)-2,3-diazabicyclo[2.2.2]octane (8). 4-Iodobiphenyl (589 mg, 2.1 mmole) was dissolved in ether (6 mL) in a 50 mL Schlenk flask and cooled to -78 °C, causing precipitation. t-BuLi (1.88 M in pentane, 2.13 mL, 4.0 mmole) was added dropwise, the precipitate dissolved, and the mixture turned bright yellow. The mixture was stirred for 2 hrs and 2-t-Bu-2,3-diazabicyclo[2.2.2]oct-2-enium iodide (588mg, 2.0 mmole) was added. The mixture was stirred overnight, allowed to warm slowly, and was quenched with water (20 mL) and extracted with pentane (4×40 mL) to give upon drying with MgSO₄ and evaporation 600 mg of the product as a pale yellow oil, contaminated with a very small amount of pentane. Sublimation at 90 °C (0.5 mm Hg) afforded **8** as thin colorless plates, m.p. 74-76 °C. MS: *m/e* 320.2252, I = 12 %, calcd. for C₂₂H₂₈N₂ 320.2252. ¹H NMR (360 MHz, CDCl₃): δ

7.87 (d., $J = 7.2$ Hz, 1H, ArH), 7.57 (d., $J = 6.9$ Hz, 2H, ArH), 7.41 (m., 4H, ArH), 7.25 (m., 1H, ArH), 7.00 (d., $J = 6.9$ Hz, 1H, ArH), 3.77 (m., 1H, NCH), 3.38 (m., 1H, NCH), 2.31 (m., 1H, CH₂), 1.92 (m., 2H, CH₂), 1.77 (m., 1H, CH₂), 1.65 (m., 2H, CH₂), 1.41 (m., 1H, CH₂), 1.25 (m., 1H, CH₂), 1.14 (s., 9H, C(CH₃)₃). ¹³C{¹H} NMR (125 MHz, CDCl₃): δ 155.97 (NC_{ar}), 141.27 (C_{ar}), 132.49 (C_{ar}), 128.56 (C_{ar}H), 126.45 (C_{ar}H), 126.13 (C_{ar}H), 120.40 (C_{ar}H), 119.83 (C_{ar}H), 58.49 (C(CH₃)₃), 53.86 (NCH), 46.38 (NCH), 28.86 (C(CH₃)₃), 28.56 (CH₂), 26.33 (CH₂), 22.09 (CH₂), 21.54 (CH₂). CV in CH₃CN with 0.1 M TBAP vs SCE on Au with FeCp*₂ as the internal standard: $E^{\circ}_1 = 0.273$ V ($\Delta E_{1,pp} = 77$ mV), $E^{\circ}_{Fe} = -0.114$ V ($\Delta E_{Fe,pp} = 72$ mV).

2-t-Bu-3-(1-naphthyl)-2,3-diazabicyclo[2.2.2]octane (9). A solution of 1-bromonaphthalene (228 mg, 1.1 mmole) in ether (3 mL) in a 50 mL Schlenk flask was cooled down to -78 °C. t-BuLi (1.90 M in pentane, 1.05 mL, 2 mmole) was added dropwise to generate a white suspension. The mixture was stirred for 2 hrs and 2-t-Bu-2,3-diazabicyclo[2.2.2]oct-2-enium iodide (294 mg, 1.00 mmole) was added. The cooling bath was slowly lowered over 30 min and then removed. The mixture stirred overnight, quenched with water (20 mL) and extracted with pentane (2×30 mL). Recrystallization from aqueous ethanol afforded **9** as bright-yellow needles (206 mg, 70 %), mp 105-106 °C. MS: *m/e* 294.2112, I = 23 %, calcd. for C₂₀H₂₆N₂ 294.2096. ¹H NMR (360 MHz, C₆D₆): δ 8.42 (d., $J = 8.6$ Hz, 1H, ArH), 8.14 (dd., $J = 7.3$ Hz, 1.3 Hz, 1H, ArH), 7.72 (d., $J = 7.7$ Hz, 1H, ArH), 7.40 (m., 3H, ArH), 7.29 (m., 1H, ArH), 3.43 (m., 1H, NCH), 3.21 (m., 1H, NCH), 2.31 (m., 1H, CH₂), 2.17 (m., 1H, CH₂), 1.87 (m., 1H, CH₂), 1.54 (m., 1H, CH₂), 1.43 (m., 2H, CH₂), 1.14 (m., 1H, CH₂), 1.11 (s., 9 H, C(CH₃)₃), 0.83 (m., 1H, CH₂). ¹³C{¹H} NMR (90 MHz, C₆D₆): δ 151.75 (NC_{ar}), 135.56 (C_{ar}), 128.83 (C_{ar}H), 125.91 (C_{ar}H), 125.55 (C_{ar}H), 125.20 (C_{ar}H), 124.12 (C_{ar}H), 122.81 (C_{ar}H), 119.59 (C_{ar}H), 59.14 (C(CH₃)₃), 53.37 (NCH), 46.50 (NCH), 29.23 (CH₂), 28.87 (C(CH₃)₃), 26.85 (CH₂), 23.11 (CH₂), 20.76 (CH₂). One of the quaternary carbons of the naphthalene unit was not observed, presumably due to the overlap with the solvent signal. CV in CH₃CN with 0.1 M TBAP vs SCE on Au with FeCp₂ as the internal standard: $E^{\circ}_1 = 0.228$ V ($\Delta E_{1,pp} = 72$ mV), $E^{\circ}_{Fe} = 0.397$ V ($\Delta E_{Fe,pp} = 75$ mV).

4,4'-bis(2-tert-butyl-2,3-diazabicyclo[2.2.2]oct-3-yl)-2,2',6,6'-tetramethylbiphenyl (10). 4,4'-diiodo-2,2',6,6'-tetramethyl-biphenyl (231 mg, 0.50 mmole) was placed into an oven dried 50 mL Schlenk flask under nitrogen. Ether (3 mL) was added, the mixture cooled to -78 °C, and t-BuLi (1.74M in pentane, 1.15 mL, 2.00 mmole) was added slowly. The mixture was stirred for 75 min and 2-t-Bu-2,3-diazabicyclo[2.2.2]oct-2-enium iodide (294 mg, 1.00 mmole) was added. The mixture was stirred for 20 min and the cooling bath was gradually lowered and then removed. The mixture was stirred overnight and quenched with water (20 mL), extracted with toluene (3 × 20 mL), dried with MgSO₄ and evaporated. The crude product was dissolved in refluxing toluene (3 mL) and CH₃CN (30 mL) was added in small portions over 2 hours, giving colorless crystals of **10** (146 mg, 54%), m.p. 266 °C. MS: *m/e* 542.4320, I = 22 %, calcd. for C₃₆H₅₄N₄ 542.4348. ¹H NMR (500 MHz, C₆D₆): δ 7.85 (s., 2H, ArH), 6.84 (s., 2H, ArH), 3.58 (br. s., 2H, NCH), 3.19 (br. s., 2H, NCH), 2.24 (br. q., $J = 10.8$ Hz, 2H, CH₂), 2.10 and 2.09 and 2.07 and 2.02 (br. s., 12H, ArCH₃), 2.00 - 2.10 (br. m., 2H, CH₂), 1.82 (m., 2H, CH₂), 1.76 (br. t., $J = 12.2$ Hz, 2H, CH₂), 1.51 (br. t., $J = 12.3$ Hz, 2H, CH₂), 1.41 (br. t., $J = 11.1$ Hz, 2H, CH₂), 1.23-1.08 (m., 2H, CH₂), 1.21 (s., 18H, C(CH₃)₃), 0.99 (br. m., 2H, CH₂). ¹³C{¹H} NMR (125 MHz, C₆D₆): δ 155.03 (NC_{ar}), 135.81 and 135.49

(\underline{C}_{ar}), 133.17 (\underline{C}_{ar}), 119.68 and 119.56 ($\underline{C}_{ar}H$), 58.64 ($\underline{C}(\underline{CH}_3)_3$), 54.31 (\underline{NCH}), 46.58(\underline{NCH}), 29.11 (\underline{CH}_2), 29.08 ($\underline{C}(\underline{CH}_3)_3$), 26.82 (\underline{CH}_2), 22.54 (\underline{CH}_2), 21.48 (\underline{CH}_2), 20.64 and 20.57(\underline{CH}_3). CV in CH_3CN with 0.1 M TBAP vs SCE on Au with $FeCp^*_2$ as the internal standard: $E_{Av}^{\circ} = 0.211$ V ($\Delta E_{av,pp} = 125$ mV, $2e^-$ wave), $E_{Fe}^{\circ} = -0.110$ V ($\Delta E_{Fe,pp} = 71$ mV).

The radical cations were prepared by $AgNO_3$ oxidation in CH_2Cl_2 . Typically, 1.9-2.2 mg of $AgNO_3$ was placed in a small test tube, 1.05 equivalents of the bis-hydrazine and 2 mL CH_2Cl_2 was added, and the mixture was vigorously stirred at room temperature ($0^\circ C$ for 6) for 5-10 hours. The resulting solution was centrifuged, filtered and either diluted with CH_2Cl_2 to the desired concentration, or diluted with 2 mL of CH_3CN , evaporated by a nitrogen stream at $0^\circ C$ to 1 mL, diluted with 2 mL of CH_3CN and evaporation - dilution was repeated 3 times to replace CH_2Cl_2 with CH_3CN . This was necessary since direct silver oxidation in CH_3CN is very sluggish. Alternatively, the radical cation solutions were obtained by mixing 1 : 1 neutral and bis(cation).

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