Electronic Supplementary Information

Utilizing Bis(imino)dihydroacridanide Pincer Ligands in p-Block Chemistry: Synthesis and Catalysis of an Antimony Monocation Salt

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I. General information

All experiments were carried out under an inert gas atmosphere by using Schlenk-type glassware or in a N_2-filled glove box. Toluene, n-hexane, Et_2O and THF were purified by distillation over sodium or $LiAlH_4$, while n-pentane was dried with sodium/potassium alloy prior to use, and stored with molecular sieves. NMR spectra were recorded on Bruker Advance 400 (^{1}H: 400 MHz) or 600 (^{1}H: 600 MHz, ^{13}C: 151 MHz, ^{11}B NMR: 193 MHz, ^{19}F NMR: 565 MHz) spectrometer at 298 K. Data are presented as follows: chemical shift, multiplicity (s = singlet, d = doublet, t = triplet, dd = doublet of doublets, q = quartet, m = multiplet, br = broad), integration, coupling constant in hertz (Hz). High resolution mass spectrometry (HRMS) was performed with a Thermo Fisher Scientific Q-Exactive MS System. UV-vis spectra were recorded by Perkin-Elmer model Lambda 365 UV-vis spectrophotometer. Crystal data were collected on a Bruker D8 Venture diffractometer with graphite monochromated Cu Kα radiation (λ =1.54178) and an Excillum METALJET diffractometer using Ga Kα radiation (λ = 1.34139). Data reduction, scaling and absorption corrections were performed using SAINT (Bruker, V8.38A, 2013). The structure was solved with the XT structure solution program using the Intrinsic Phasing solution method and by using Olex2 as the graphical interface. The frames were integrated with the Bruker SAINT software package using a narrow-frame algorithm. Data were corrected for absorption effects using the empirical multiscan method (SADABS). The model was refined with the ShelXL program using Least Squares minimization. All non-hydrogen atoms were refined anisotropically. Hydrogen atoms were included in structure factor calculations. All hydrogen atoms were assigned to idealized geometric positions. Commercial reagents were purchased from Energy Chemical, J&K, or TCI Chemical Co. and used as received. Compounds 2,7-*di-tert*-butyl-9,9-dimethyl-9,10-dihydroacridine[S1] and 4,5-dibromo-2,7-*di-tert*-butyl-9,9-dimethyl-9,10-dihydroacridine[S2] were prepared according to the procedure described in the literature.

Sensitivity of the compounds: When exposed to air, compound **2** readily hydrolyzed to form compound **1** (Figure S25). Compound **3**, upon exposure to ambient air, partially converted into compound **1** (Figure S26). In contrast, compound **4**[$B(Ar_F)_4$] exhibited stability in the presence of air (Figure S28). On subjecting **5**[$B(Ar_F)_4$] to air, a minor fraction converted to compound **1**, while another unidentified compound was formed (Figure S29). Additionally, when water was introduced to the C_6D_6 solution containing compound **3**, it led to the formation of compound **1** (Figure S27).

II. Experimental Section

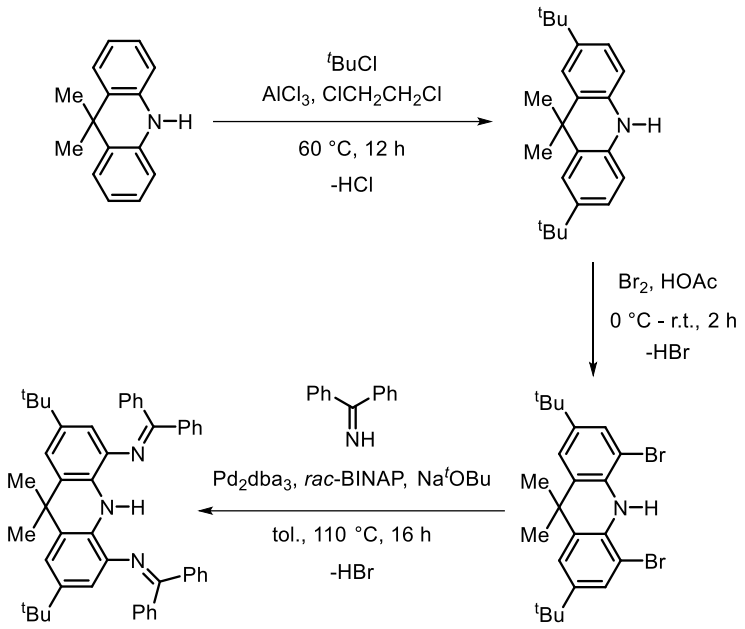
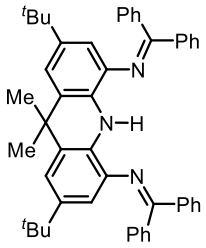
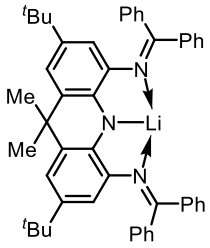


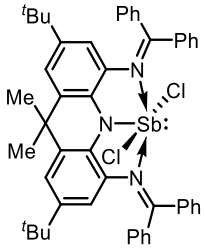
Figure S1. Synthetic route towards **1**.



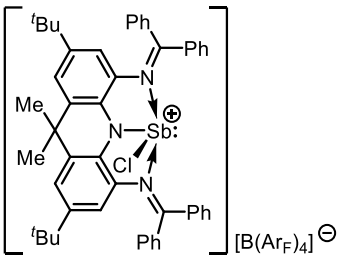
Preparation of **1**:[S3] The catalyst mixture, comprising Pd_2dba_3 (0.78g, 0.85 mmol), *rac*-BINAP (1.70 mmol, 1.06g), and NaO^tBu (5.30g, 55.15 mmol), was heated to 110℃ in toluene. A solution of 2,7-di-tert-butyl-9,9-dimethyl-9,10-dihydroacridine (10.12g, 21.21mmol) and benzophenone imine (9.25 mL, 55.15 mmol) in toluene was then added to this mixture, followed by refluxing for 16 hours. Afterward, the reaction mixture was cooled to room temperature and extracted with Et_2O. The combined organic layers were then dried using Na_2SO_4, filtered using gravity, and the filtrate was concentrated under reduced pressure to yield a dark orange oil. This crude product was purified through column chromatography using CH_2Cl_2. After concentration under reduced pressure, it was filtered with Et_2O, producing an orange solid (13.24 g, 92%). Orange crystals of compound **1**, suitable for X-ray analysis, were obtained by cooling a saturated Et_2O solution to -15℃. ^{1}H NMR (600 MHz, C_6D_6) δ 8.67 (s, 1H), 7.92 (d, *J* = 7.0 Hz, 4H), 7.30 (d, *J* = 2.0 Hz, 2H), 7.13 (t, *J* = 7.4 Hz, 2H), 7.02 (dd, *J* = 7.6, 1.9 Hz, 4H), 6.91 - 6.87(m, 10H), 6.45 (d, *J* = 2.0 Hz, 2H), 1.73 (s, 6H), 1.15 (s, 18H). ^{13}C{^{1}H} NMR (151 MHz, C_6D_6): δ 167.74, 141.55, 140.19, 137.73, 135.26, 132.60, 130.39, 129.58, 129.30, 129.27, 128.50, 128.41, 118.15, 116.50, 37.60, 34.41, 31.69, 30.23. HMRS(ESI) $[M+H]^+$ $[C_{49}H_{50}N_3]^+$: calcd. 680.39993, found 680.39740 m/z.



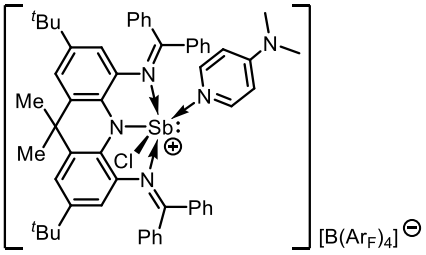
Preparation of **2**: $TMSCH_2Li$ (2.68 mL, 0.55M in hexane, 1.47 mmol) was added to a toluene solution of compound **1** (1.00 g, 1.47 mmol) at room temperature. The mixture turned into a dark blue solution and was stirred overnight. After the solvent was removed under vacuum, the resultant solid was filtered through Celite. The crude product was then dissolved in toluene to prepare a concentrated solution. By layering hexane onto this toluene solution at -30℃, a dark blue solid was recrystallized, yielding 808 mg (80% yield). Blue crystals of compound **2**, suitable for X-ray analysis, were obtained by cooling a saturated Et_2O solution at room temperature. ^{1}H NMR (600 MHz, C_6D_6) δ 7.45 (s, 2H), 7.18 (d, *J* = 7.7 Hz, 4H), 7.09 (d, *J* = 7.7 Hz, 4H), 6.99 – 6.96 (m, 8H), 6.82 (t, *J* = 7.6 Hz, 4H), 6.52 (s, 2H), 1.95 (s, 6H), 1.23 (s, 18H). ^{13}C{^{1}H} NMR (151 MHz, C_6D_6) δ 164.07, 146.96, 142.71, 138.58, 136.59, 135.13, 131.71, 130.31, 129.61, 128.98, 120.96, 117.54, 37.86, 34.10, 31.85. HMRS(ESI) $[M\text{-}Li]^-$ $[C_{49}H_{48}N_3]^-$: calcd. 678.38537, found 678.38525 m/z.



Preparation of **3**: A toluene solution containing $SbCl_3$ (135.6 mg, 0.60 mmol) was added to another toluene solution of compound **2** (411.2 mg, 0.60 mmol) at room temperature. This produced a violet solution that was stirred for 3 hours before being filtered through Celite. The solvent was then removed under vacuum. The remaining residue was washed with hexane and subsequently dried under vacuum, yielding a violet solid (418 mg, 80% yield). Violet crystals of compound **3**, suitable for X-ray analysis, were obtained by layering hexane over the toluene solution at room temperature. ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.66 (d, *J* = 7.6 Hz, 4H), 7.54 (d, *J* = 6.8 Hz, 4H), 7.49 – 7.39 (m, 8H), 7.31 (t, *J* = 7.6 Hz, 4H), 7.18 (s, 2H), 6.43 (d, *J* = 2.0 Hz, 2H), 1.66 (s, 6H), 0.93 (s, 18H). ^{13}C{^{1}H} NMR (151 MHz, $CDCl_3$) δ 173.75, 141.08, 138.83, 136.90, 136.20, 133.09, 132.51, 131.82, 131.18, 130.09, 129.55, 129.10, 128.46, 123.98, 117.86, 36.70, 34.56, 34.02, 31.02. HMRS(ESI) $[M]^+$ $[C_{49}H_{48}N_3Cl_2Sb]^+$: calcd. 869.22580, found 869.22516 m/z. UV-vis (toluene): λ_{max} = 328 nm, 586 nm.



Preparation of **4**[B(Ar_F)$_4$]: To a mixture of compound **3** (173.8 mg, 0.20 mmol) and NaB(Ar_F)$_4$ (Ar_F = 3,5-$(CF_3)_2C_6H_3$) (177.2 mg, 0.20 mmol), toluene was added at room temperature. The mixture was stirred for 2 hours at room temperature and then filtered through Celite. After removing the solvent under vacuum, the residue was washed with hexane and dried under vacuum, yielding a reddish solid (280 mg, 82% yield). Light orange crystals of **4**[B(Ar_F)$_4$], suitable for X-ray analysis, were obtained from a toluene/pentane solution at -30℃. ^{1}H NMR (600 MHz, C_6D_6) δ 8.37 (s, 8H), 7.72 (br, 2H), 7.64 (s, 4H), 7.38 (d, *J* = 2.0 Hz, 2H), 7.17 (s, 2H), 7.11 (t, *J* = 7.5 Hz, 2H), 7.06 – 6.89 (m, 8H) 6.79 (d, *J* = 7.9 Hz, 4H), 6.66 (br, 2H), 6.59 (d, *J* = 1.9 Hz, 2H), 1.63 (s, 3H), 1.40 (s, 3H), 0.97 (s, 18H). ^{13}C{^{1}H} NMR (151 MHz, C_6D_6) 176.51, 162.81 (q, *J* = 49.9 Hz), 144.79, 138.50, 135.96, 135.44, 134.49, 134.25, 134.18, 133.32, 132.36, 131.69, 130.18, 129.98, 129.77, 129.57, 129.33, 128.92, 128.69, 126.18, 124.38, 124.02, 122.57, 118.07, 117.79, 36.90, 35.43, 34.28, 30.80, 26.06. ^{11}B NMR (193 MHz, C_6D_6) δ -5.92. ^{19}F NMR (565 MHz, C_6D_6) δ -62.05. HMRS(ESI) [M]$^+$ [$C_{49}H_{48}ClN_3Sb$]$^+$: calcd. 834.25695, found 834.25714 m/z. UV-vis (toluene): λ_{max} = 332 nm, 520 nm.



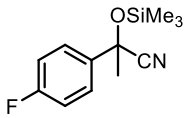
Preparation of **5**[B(Ar_F)$_4$]: To a mixture of **4**[B(Ar_F)$_4$] (20.0 mg, 0.0118 mmol) and 4-dimethylaminopyridine (DMAP) (1.4 mg, 0.0118 mmol), toluene was added at room temperature. The solution was stirred for 1 hour and then filtered through Celite. The solvent was subsequently removed under vacuum. The remaining residue was washed with pentane and dried under vacuum, yielding a purple solid (18.6 mg, 86% yield). Violet crystals of **5**[B(Ar_F)$_4$], suitable for X-ray analysis, were obtained from a toluene/pentane solution at room temperature. ^{1}H NMR (600 MHz, Toluene-d_8) δ 8.26 (s, 8H), 7.61 (s, 4H), 7.40 (s, 2H), 7.29 (d, *J* = 5.9 Hz, 2H), 7.20-6.80 (m, 20H), 6.52 (d, *J* = 1.9 Hz, 2H), 5.82 (d, *J* = 6.3 Hz, 2H), 2.08 (s, 6H), 1.72 (s, 6H), 0.95 (s, 18H). ^{13}C{^{1}H} NMR (151 MHz, Toluene-d_8) δ 175.24, 162.66 (q, *J* = 49.8 Hz), 155.70, 144.72, 138.72, 136.27, 135.41, 134.04, 132.98, 132.13, 131.52, 130.62, 130.02, 126.09, 124.28, 122.48, 118.13, 117.96, 107.02, 38.02, 36.95, 34.17, 30.83. ^{19}F NMR (565 MHz, Toluene-d_8) δ -62.24. ^{11}B NMR (193 MHz, Toluene-d8) δ -6.02.

HMRS(ESI) $[M\text{-}DMAP]^+$ $[C_{49}H_{48}ClN_3Sb]^+$: calcd. 834.25695, found 834.25507 m/z. UV-vis (toluene): λ_{max} = 558 nm.

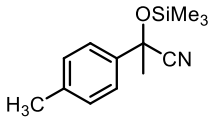
Gutmann-Beckett tests for **4**[$B(Ar_F)_4$]: Inside a nitrogen-filled glovebox**,** the mixture of **4**[$B(Ar_F)_4$] (0.01 mmol, 17.0 mg) and triethylphosphine oxide (0.01 mmol, 1.3 mg) in a sealed J-Young NMR tube was added 0.5 mL CD_2Cl_2. The resulted solution was analyzed by ^{31}P NMR spectroscopy.

Cyanosilylation of Ketone: Metod A: TMSCN (0.13 mmol, 12.9 mg) was added to the C_6D_6 solution (0.6 mL) of ketone **6** (0.10 mmol), **4**[$B(Ar_F)_4$] (2 mol%, 3.4 mg) and 1,3,5-trimethoxybenzene (0.10 mmol, 16.8 mg) in a J-Young NMR tube at room temperature. The resulting solution was stirred for 9 hours. The yield was calculated from the ratio of reactant and product from the crude ^1H NMR spectra by using 1,3,5-trimethoxybenzene as an internal standard.

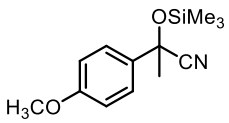
Method B: TMSCN (0.26 mmol, 25.8 mg) was added to the toluene solution (1.2 mL) of ketone **6** (0.20 mmol) and **4**[$B(Ar_F)_4$] (4 mol%, 6.8 mg) in a vial at room temperature. The resulting solution was stirred for 12 hours, then brought outside of the glove box and the solvent was removed with ra otary evaporator. The residue was chromatographed by silica column to give the desired product **7**.



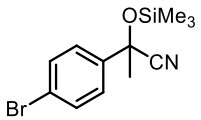
7a: 9.5 mg, yellow liquid. ^1H NMR (400 MHz, $CDCl_3$) δ 7.52 (dd, *J* = 7.3, 5.1 Hz, 2H), 7.08 (t, *J* = 7.7 Hz, 2H), 1.84 (s, 3H), 0.18 (s, 9H).



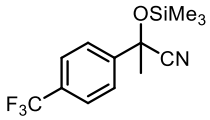
7b: 28.6 mg, yellow liquid. ^1H NMR (600 MHz, $CDCl_3$) δ 7.43 (d, *J* = 8.2 Hz, 2H), 7.20 (d, *J* = 7.9 Hz, 2H), 2.37 (s, 3H), 1.85 (s, 3H), 0.17 (s, 9H).



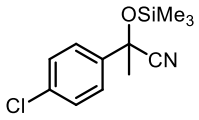
7c: 35.0 mg, yellow liquid. ^1H NMR (600 MHz, $CDCl_3$) δ 7.46 (d, *J* = 8.9 Hz, 2H), 6.91 (d, *J* = 8.8 Hz, 2H), 3.82 (s, 3H), 1.85 (s, 3H), 0.16 (s, 9H).



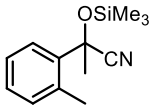
7d: 21.3 mg, yellow liquid. ^1H NMR (600 MHz, $CDCl_3$) δ 7.53 (d, *J* = 8.4 Hz, 2H), 7.42 (d, *J* = 8.4 Hz, 2H), 1.83 (s, 3H), 0.19 (s, 9H).



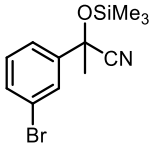
7f: 11.6 mg, yellow liquid.^{1}H NMR (600 MHz, $CDCl_3$) δ 7.67 (s, 4H), 1.86 (s, 3H), 0.22 (s, 9H).



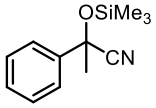
7g: 14.5 mg, yellow liquid. ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.48 (d, *J* = 8.6 Hz, 2H), 7.37 (d, *J* = 8.5 Hz, 2H), 1.83 (s, 3H), 0.19 (s, 9H).



7h: 12.7 mg, yellow liquid. ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.58 (d, *J* = 8.3 Hz, 1H), 7.26 (d, *J* = 16.1 Hz, 1H), 7.21 (t, *J* = 8.3 Hz, 2H), 2.58 (s, 3H), 1.97 (s, 3H), 0.19 (s, 9H).



7i: 22.3 mg, yellow liquid. ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.68 (s, 1H), 7.48 (t, *J* = 6.3 Hz, 2H), 7.28 (d, *J* = 8.0 Hz, 1H), 1.84 (s, 3H), 0.21 (s, 9H).



7j: 15.8 mg, yellow liquid. ^{1}H NMR (600 MHz, $CDCl_3$) δ 7.55 (d, *J* = 8.2 Hz, 2H), 7.40 (t, *J* = 7.4 Hz, 2H), 1.86 (s, 3H), 0.18 (s, 9H).

III. UV-vis spectra

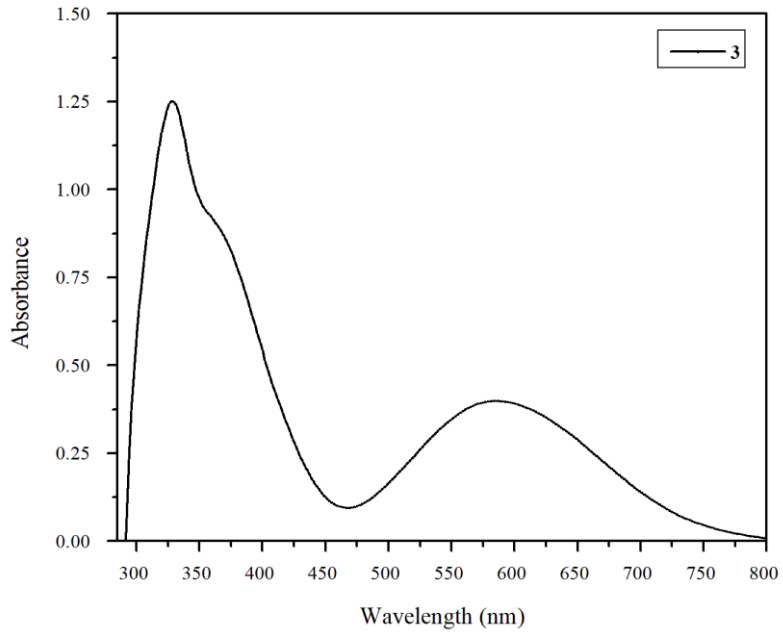


Figure S2. UV-vis spectrum of **3** in toluene (5.75×10^{-5} mol/L).

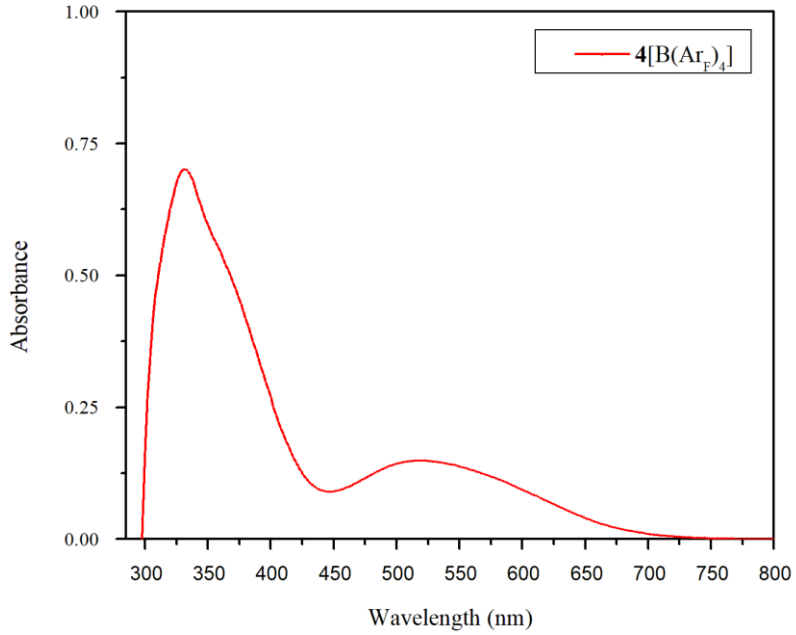


Figure S3. UV-vis spectrum of **4**[$B(Ar_F)_4$] in toluene (2.94×10^{-5} mol/L).

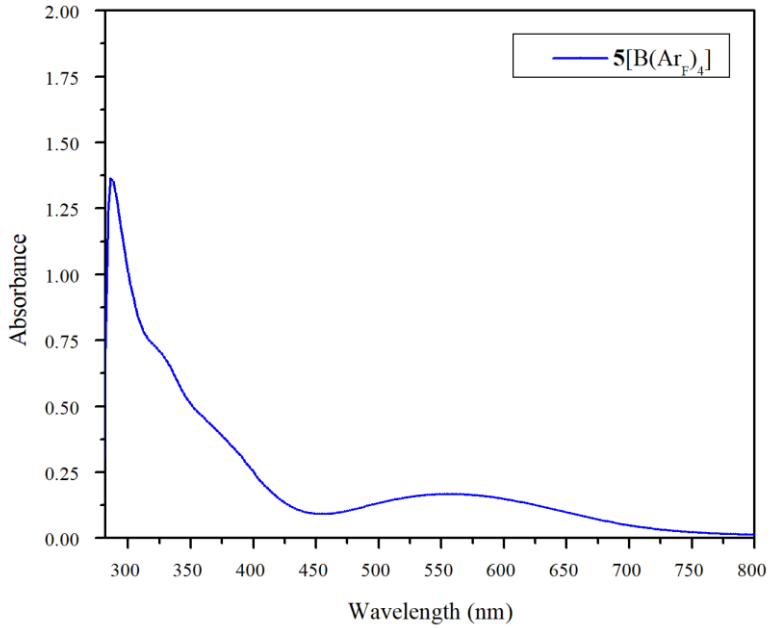


Figure S4. UV-vis spectrum of **5**[B(Ar$_F$)$_4$] in toluene (2.74×10^{-5} mol/L).

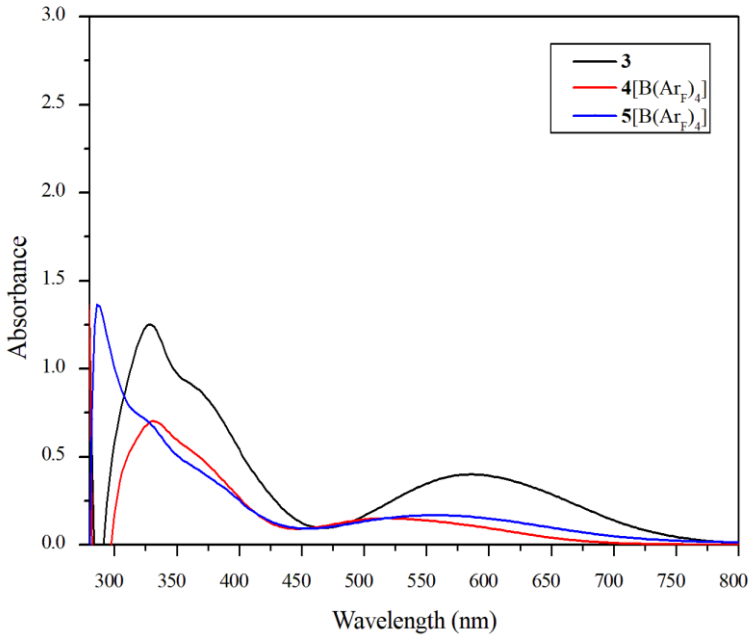


Figure S5. UV-vis spectrum of **3**, **4**[B(Ar$_F$)$_4$], **5**[B(Ar$_F$)$_4$] in toluene.

IV. X-ray crystallographic data

Table S1 Crystal data and structure refinement for **1**.

Identification code	2220792
Empirical formula	$C_{49}H_{49}N_3$
Formula weight	679.91
Temperature/K	100.0
Crystal system	monoclinic
Space group	$P2_1/n$
a/Å	13.6632(6)
b/Å	18.3235(8)
c/Å	15.7420(6)
α/°	90
β/°	104.596(2)
γ/°	90
Volume/Å^3	3813.9(3)
Z	4
ρ_{calc}g/cm^3	1.184
μ/mm^{-1}	0.520
F(000)	1456.0
Crystal size/mm^3	0.3 × 0.3 × 0.3
Radiation	CuKα (λ = 1.54178)
2Θ range for data collection/°	7.546 to 136.752
Index ranges	$-16 \leq h \leq 16$, $-22 \leq k \leq 22$, $-18 \leq l \leq 14$
Reflections collected	39746
Independent reflections	6734 [R_{int} = 0.0356, R_{sigma} = 0.0245]
Data/restraints/parameters	6734/66/507
Goodness-of-fit on F^2	1.083
Final R indexes [I>=2σ (I)]	R_1 = 0.0390, wR_2 = 0.0971
Final R indexes [all data]	R_1 = 0.0436, wR_2 = 0.1000
Largest diff. peak/hole / e Å^{-3}	0.55/-0.55

Table S2 Crystal data and structure refinement for **2**.

Identification code	2220794
Empirical formula	$C_{53}H_{58}LiN_3O$
Formula weight	759.96
Temperature/K	100.0
Crystal system	triclinic
Space group	P-1
a/Å	12.0383(5)
b/Å	13.7852(6)

c/Å	14.1852(6)
α/°	87.949(2)
β/°	69.745(2)
γ/°	81.798(2)
Volume/Å^3	2185.64(16)
Z	2
ρ_{calc}g/cm^3	1.155
μ/mm^{-1}	0.515
F(000)	816.0
Crystal size/mm^3	0.3 × 0.3 × 0.2
Radiation	CuKα (λ = 1.54178)
2Θ range for data collection/°	6.478 to 145.022
Index ranges	-14 ≤ h ≤ 14, -17 ≤ k ≤ 16, -17 ≤ l ≤ 17
Reflections collected	29440
Independent reflections	8351 [R_{int} = 0.0561, R_{sigma} = 0.0488]
Data/restraints/parameters	8351/0/533
Goodness-of-fit on F^2	0.973
Final R indexes [I>=2σ (I)]	R_1 = 0.0536, wR_2 = 0.1352
Final R indexes [all data]	R_1 = 0.0573, wR_2 = 0.1379
Largest diff. peak/hole / e Å^{-3}	0.33/-0.28

Table S3 Crystal data and structure refinement for **3**.

Identification code	2220791
Empirical formula	$C_{56}H_{56}Cl_2N_3Sb$
Formula weight	963.68
Temperature/K	293.15
Crystal system	triclinic
Space group	P-1
a/Å	12.9492(14)
b/Å	15.3029(19)
c/Å	15.575(2)
α/°	98.084(9)
β/°	114.097(6)
γ/°	114.290(6)
Volume/Å^3	2384.9(5)
Z	2
ρ_{calc}g/cm^3	1.342
μ/mm^{-1}	5.918
F(000)	996.0
Crystal size/mm^3	0.2 × 0.15 × 0.1

Radiation	CuKα (λ = 1.54178)
2Θ range for data collection/°	6.694 to 145.412
Index ranges	$-15 \leq h \leq 15$, $-18 \leq k \leq 18$, $-19 \leq l \leq 19$
Reflections collected	55621
Independent reflections	9260 [R_{int} = 0.0365, R_{sigma} = 0.0228]
Data/restraints/parameters	9260/36/598
Goodness-of-fit on F^2	1.035
Final R indexes [I>=2σ (I)]	R_1 = 0.0221, wR_2 = 0.0508
Final R indexes [all data]	R_1 = 0.0239, wR_2 = 0.0515
Largest diff. peak/hole / e $Å^{-3}$	0.67/-0.30

Table S4 Crystal data and structure refinement for **4**[$B(Ar_F)_4$].

Identification code	2220796
Empirical formula	$C_{86}H_{72}BClF_{24}N_3Sb$
Formula weight	1771.47
Temperature/K	100.0(2)
Crystal system	triclinic
Space group	P-1
a/Å	12.0357(8)
b/Å	15.0197(11)
c/Å	24.7036(17)
α/°	91.171(5)
β/°	101.962(4)
γ/°	112.763(4)
Volume/$Å^3$	4003.3(5)
Z	2
ρ_{calc}g/cm^3	1.470
μ/mm^{-1}	3.948
F(000)	1796.0
Crystal size/mm^3	0.2 × 0.1 × 0.1
Radiation	CuKα (λ = 1.54178)
2Θ range for data collection/°	6.422 to 134.998
Index ranges	$-13 \leq h \leq 14$, $-17 \leq k \leq 17$, $-29 \leq l \leq 29$
Reflections collected	37201
Independent reflections	14260 [R_{int} = 0.1029, R_{sigma} = 0.1307]
Data/restraints/parameters	14260/0/1049
Goodness-of-fit on F^2	1.096
Final R indexes [I>=2σ (I)]	R_1 = 0.1107, wR_2 = 0.2576
Final R indexes [all data]	R_1 = 0.1442, wR_2 = 0.2729
Largest diff. peak/hole / e $Å^{-3}$	1.97/-1.51

Table S5 Crystal data and structure refinement for **5**[$B(Ar_F)_4$].

Identification code	2220795
Empirical formula	$C_{88}H_{70}BClF_{24}N_5Sb$
Formula weight	1821.50
Temperature/K	100.0
Crystal system	triclinic
Space group	P-1
a/Å	16.8928(7)
b/Å	17.0063(8)
c/Å	19.8764(9)
α/°	84.139(2)
β/°	74.036(2)
γ/°	66.218(2)
Volume/$Å^3$	5023.5(4)
Z	2
$\rho_{calc}g/cm^3$	1.204
μ/mm^{-1}	2.126
F(000)	1844.0
Crystal size/mm^3	0.3 × 0.2 × 0.15
Radiation	GaKα (λ = 1.34139)
2Θ range for data collection/°	4.022 to 108.31
Index ranges	$-20 \leq h \leq 20$, $-20 \leq k \leq 20$, $-23 \leq l \leq 23$
Reflections collected	107781
Independent reflections	18455 [R_{int} = 0.0669, R_{sigma} = 0.0550]
Data/restraints/parameters	18455/816/1118
Goodness-of-fit on F^2	1.101
Final R indexes [I>=2σ (I)]	R_1 = 0.0902, wR_2 = 0.2432
Final R indexes [all data]	R_1 = 0.1134, wR_2 = 0.2596
Largest diff. peak/hole / e $Å^{-3}$	1.84/-1.75

V. Computational details

Geometry optimizations were performed using the Gaussian 16 package[S4] with the hybrid B3LYP density functional augmented with the D3(BJ) version of Grimme's empirical dispersion correction[S5]. The Def2-SVP basis set was employed for all the atoms. Frequency calculations at the same level of theory were performed to identify the number of imaginary frequencies (zero for local minimum and one for transition states) and provide the thermal corrections of Gibbs free energy. The corrections of Gibbs free energy from frequency calculations were added to the electronic energies to obtain the Gibbs free energy, respectively. The Transition states were submitted to intrinsic reaction coordinate (IRC) calculations to determine two corresponding minima. Orbital decomposition analysis was performed using Multiwfn using the wave functions from the optimized structure. [S6] TD-DFT calculations were carried out at the PBE38/def2-SVP level of theory.

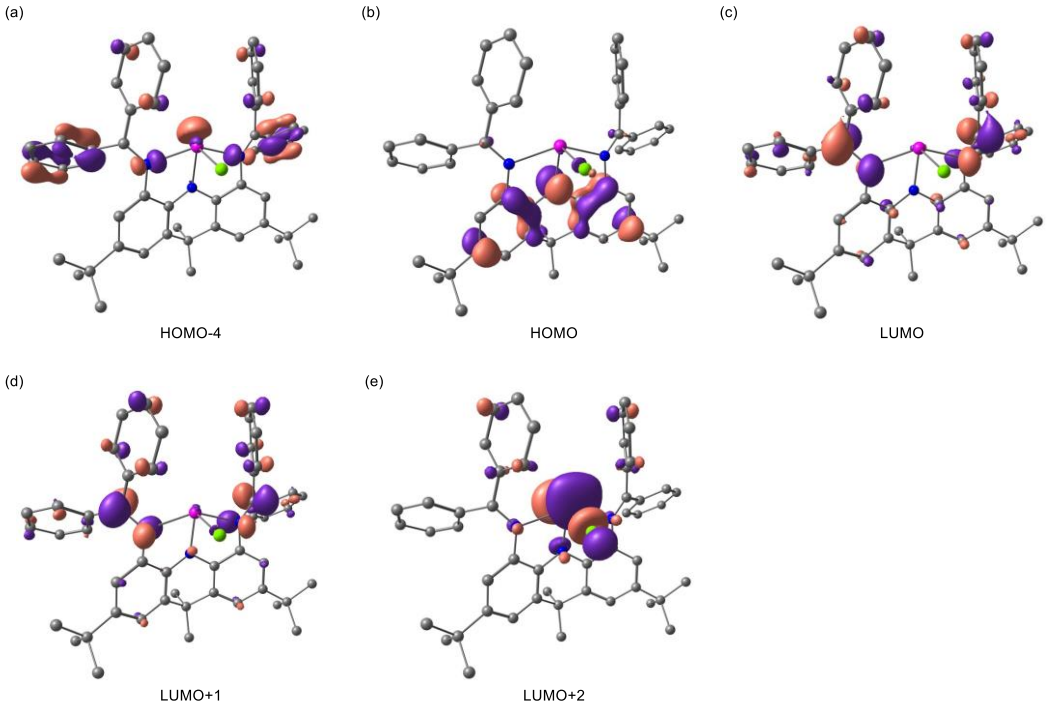


Figure S6. Frontier molecular orbitals of **4**[B(Ar_F)$_4$].

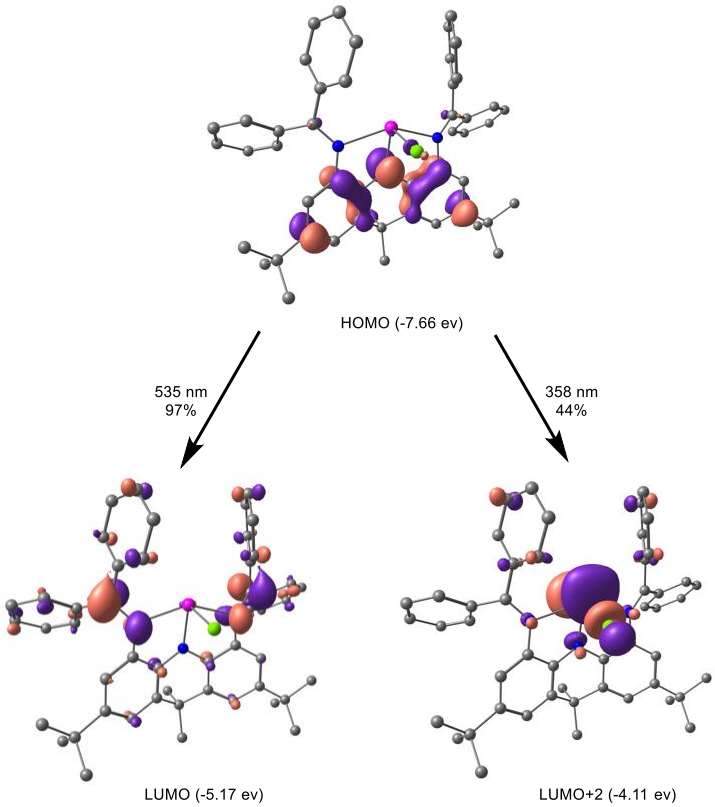


Figure S7. Major transition orbitals of **4** for transition at 358 nm and 535 nm from TD-DFT calculations.

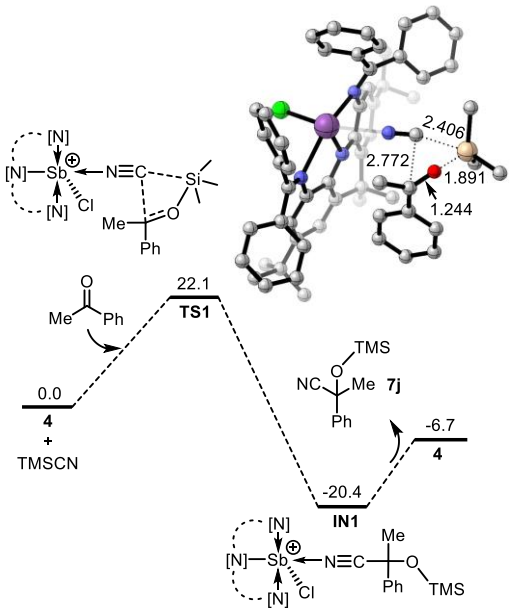


Figure S8. Free energy profile for the catalytic cyanosilylation of acetophenone (Energies are given in kcal/mol). The bis(imino)dihydroacridanide ligand is omitted for clarity. In the 3D structure of **TS1**, bond lengths are given in angstrom.

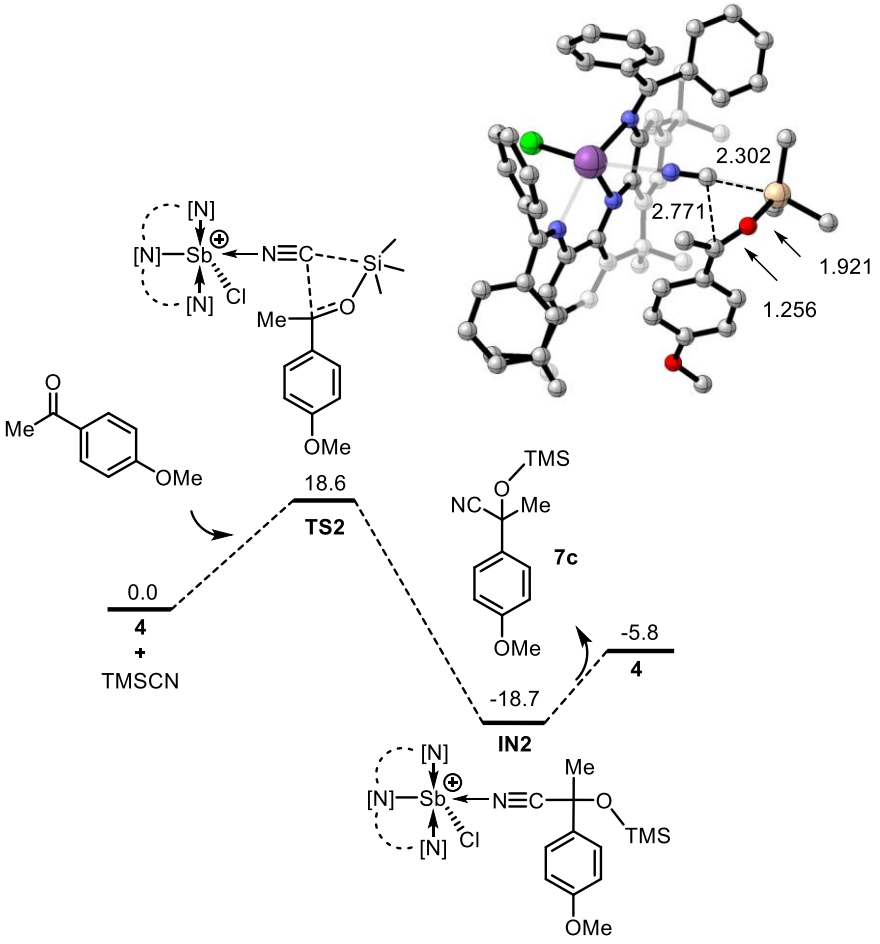


Figure S9. Free energy profile for the catalytic cyanosilylation of 4'-methoxyacetophenone (Energies are given in kcal/mol). The bis(imino)dihydroacridanide ligand is omitted for clarity. In the 3D structure of **TS2**, bond lengths are given in angstrom.

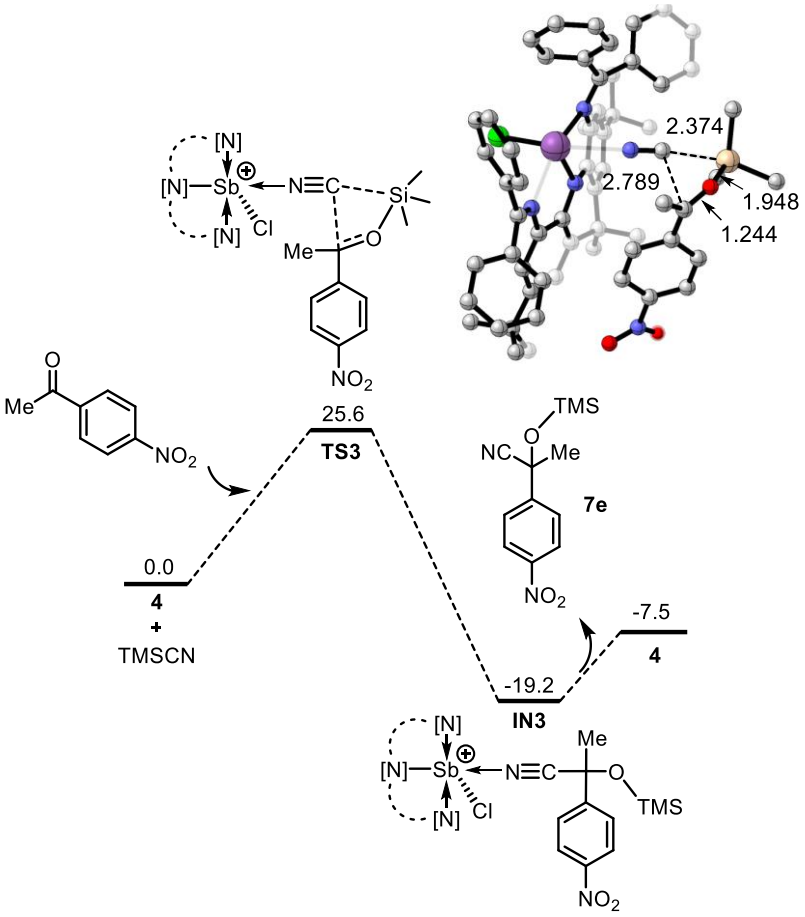


Figure S10. Free energy profile for the catalytic cyanosilylation of 4-nitroacetophenone (Energies are given in kcal/mol). The bis(imino)dihydroacridanide ligand is omitted for clarity. In the 3D structure of **TS3**, bond lengths are given in angstrom.

Optimized structure of **4** (B3LYP-D3BJ)

Sb	0.00004400	1.24843900	-0.25130300
Cl	0.00001700	0.86750100	-2.65102200
N	0.00000500	-0.79635600	0.23679700
N	-2.24586300	0.62750000	-0.18027500
N	2.24593600	0.62740700	-0.18027400
C	1.19820400	-1.49621600	0.07730200
C	-3.15367100	1.50458300	0.17015600
C	1.24655100	-2.90606000	0.13475300
C	-1.19823800	-1.49616100	0.07737200
C	4.39598400	1.15652200	0.89668300
C	-0.00010600	-5.12161900	0.13912000
H	-0.00015100	-5.19629900	-0.95828600
H	-0.87750900	-5.65699700	0.52603300
H	0.87730300	-5.65703900	0.52596400
C	2.38218200	-0.77150200	-0.17264600
C	2.46138700	-3.53338900	-0.13272000

H	2.50260200	-4.62030300	-0.09458200
C	0.00000900	-3.63070000	2.15904900
H	0.89521200	-4.13794000	2.55066800
H	-0.89519000	-4.13789400	2.55073800
H	0.00005000	-2.59631100	2.53426000
C	2.89326800	2.92573100	-0.13234800
C	3.22944400	3.92661400	0.80019600
H	3.72130000	3.65046300	1.73434400
C	-3.64334900	-2.82922800	-0.44805300
C	-0.00005300	-3.66032300	0.60596600
C	2.27125200	3.29294700	-1.34330000
H	2.06040000	2.53365900	-2.09750200
C	-1.95998600	4.62976800	-1.59589600
H	-1.48813300	4.90387900	-2.54169400
C	4.94114400	-3.59766900	-0.73160600
C	-2.27051200	3.29298600	-1.34314600
H	-2.05934600	2.53364100	-2.09720400
C	3.58171100	-1.44024200	-0.46035900
H	4.46024100	-0.84969100	-0.70012400
C	-2.46155200	-3.53326800	-0.13250600
H	-2.50282400	-4.62017900	-0.09434000
C	-4.39602800	1.15670900	0.89634700
C	-3.58179100	-1.44006800	-0.46009500
H	-4.46030700	-0.84946600	-0.69979200
C	-2.38219100	-0.77139400	-0.17253300
C	5.53128800	-0.04099800	2.67438700
H	5.49512800	-0.71488900	3.53282700
C	5.62125400	1.72118600	0.50396100
H	5.65170100	2.41221100	-0.34068300
C	1.96086500	4.62975200	-1.59610400
H	1.48940300	4.90392600	-2.54207800
C	2.90633900	5.25788800	0.54493100
H	3.15230900	6.02585600	1.28123900
C	3.15376400	1.50443500	0.17026900
C	-1.24665800	-2.90600000	0.13485100
C	3.64319000	-2.82940500	-0.44836900
C	-2.89302800	2.92585700	-0.13242500
C	6.79782600	1.38005600	1.17347300
H	7.74958900	1.80647100	0.85020900
C	4.35953200	0.27560000	1.99360700
H	3.40975000	-0.15837200	2.30979400
C	-4.94137000	-3.59743200	-0.73114800
C	2.26858500	5.61158000	-0.65029500
H	2.02522000	6.65774900	-0.84861100

C	-3.22952200	3.92679300	0.79994700
H	-3.72175200	3.65070100	1.73391600
C	5.31805100	-4.41957600	0.51960500
H	5.47141700	-3.76070700	1.38874900
H	6.25074800	-4.97841700	0.34412000
H	4.53771000	-5.14956400	0.78221500
C	-2.90627500	5.25804300	0.54474000
H	-3.15250600	6.02605600	1.28091400
C	6.75463200	0.49969700	2.25760900
H	7.67416800	0.23857100	2.78591400
C	-2.26804900	5.61165400	-0.65025800
H	-2.02457100	6.65780500	-0.84853100
C	-5.62116100	1.72167700	0.50363400
C	6.10689200	-2.65056600	-1.05583000
H	5.90557300	-2.04715300	-1.95448300
H	7.01799100	-3.23610800	-1.24935700
H	6.32380000	-1.96648600	-0.22051800
C	-4.35984300	0.27546000	1.99301400
H	-3.41016800	-0.15877600	2.30916400
C	4.72653500	-4.54385900	-1.93083200
H	3.92799100	-5.27590900	-1.73786600
H	5.64878300	-5.10657600	-2.14468500
H	4.45324900	-3.97729000	-2.83442800
C	-6.79786100	1.38056100	1.17292600
H	-7.74951800	1.80722500	0.84967600
C	-6.10710700	-2.65027800	-1.05526900
H	-7.01824900	-3.23578200	-1.24870900
H	-5.90584400	-2.04687800	-1.95394300
H	-6.32390900	-1.96618600	-0.21994000
C	-5.31819000	-4.41930200	0.52011300
H	-5.47143700	-3.76041400	1.38926300
H	-4.53785800	-5.14932400	0.78265400
H	-6.25093200	-4.97810100	0.34473000
C	-5.53172700	-0.04113000	2.67357900
H	-5.49577600	-0.71527500	3.53182900
C	-6.75493200	0.49989700	2.25682500
H	-7.67457000	0.23877900	2.78495800
C	-4.72692900	-4.54365000	-1.93038100
H	-5.64922600	-5.10632400	-2.14413300
H	-3.92840200	-5.27573700	-1.73748400
H	-4.45370600	-3.97710800	-2.83401300
H	-5.65139700	2.41294100	-0.34082300

Optimized structure of **TS1** (B3LYP-D3BJ)

Sb	-0.03354600	-1.19596200	-1.27236800
Cl	0.17804200	-0.67312900	-3.66952000
N	0.06384300	0.86917800	-0.80282100
N	2.31310200	-0.63904000	-1.02735300
N	-2.21959700	-0.45798900	-1.39207400
C	-1.08996900	1.61223100	-1.03321600
C	3.18531600	-1.58961200	-0.84234500
C	-1.10034900	3.02072700	-0.94058300
C	1.29020200	1.50776700	-0.79388700
C	-4.43618400	-0.88069400	-0.40211700
C	0.22072500	5.17971400	-0.71573000
H	0.36487900	5.28734700	-1.80079900
H	1.05747600	5.67140600	-0.20125100
H	-0.68122700	5.73543600	-0.42664100
C	-2.28145100	0.94425100	-1.38831500
C	-2.26179700	3.70330700	-1.29499600
H	-2.26653600	4.78969800	-1.22749300
C	-0.09498700	3.62972000	1.23219100
H	-1.02510700	4.14626200	1.51501500
H	0.74728700	4.10555900	1.75595100
H	-0.16497000	2.58611300	1.57125400
C	-2.94021900	-2.72152100	-1.31162900
C	-3.29244700	-3.67341800	-0.33688200
H	-3.77960400	-3.34559200	0.58266300
C	3.82431200	2.78989700	-0.93718100
C	0.10982500	3.70535600	-0.30529700
C	-2.30213400	-3.14463100	-2.49498300
H	-2.05255900	-2.41448800	-3.26749700
C	1.72329000	-4.60616900	-2.62289800
H	1.24875100	-4.84184100	-3.57753700
C	-4.67631800	3.87797000	-2.10315100
C	2.15012200	-3.30031400	-2.37230100
H	2.00925800	-2.52586900	-3.12789500
C	-3.42460700	1.66423800	-1.76320200
H	-4.30087800	1.10839900	-2.08327800
C	2.63177300	3.51463100	-0.72824000
H	2.69716400	4.59554900	-0.61907600
C	4.57772500	-1.43905500	-0.33507500
C	3.72413000	1.40730700	-1.04131000
H	4.61158900	0.81409800	-1.22396600
C	2.48460200	0.75147200	-0.93711400
C	-5.59354300	0.38859000	1.31092200
H	-5.56610000	1.09732200	2.14155600
C	-5.65718700	-1.45953300	-0.78597100

H	-5.67733300	-2.18769900	-1.59913100
C	-2.00783200	-4.49520000	-2.68677300
H	-1.52537400	-4.81601800	-3.61213300
C	-2.98177300	-5.01817600	-0.53007400
H	-3.24013200	-5.75009700	0.23818100
C	-3.17794000	-1.28356500	-1.06960100
C	1.37807400	2.91626900	-0.65265700
C	-3.43675100	3.05443300	-1.72935200
C	2.77165500	-2.98374700	-1.15108100
C	-6.84191900	-1.09316400	-0.14415300
H	-7.78955000	-1.53375500	-0.46062500
C	-4.41396100	0.04606100	0.65688500
H	-3.46603800	0.48275900	0.96694100
C	5.16908000	3.52606800	-1.00502200
C	-2.33948100	-5.43125400	-1.70341900
H	-2.10654800	-6.48781600	-1.85383600
C	2.99284800	-4.00207300	-0.20581400
H	3.50455100	-3.76616200	0.72972300
C	-5.13549400	4.67893400	-0.86586900
H	-5.38858800	4.00172800	-0.03484400
H	-6.02880600	5.27738900	-1.10523000
H	-4.35602300	5.37090300	-0.51286500
C	2.54561500	-5.29924600	-0.45371600
H	2.70148400	-6.07999500	0.29394200
C	-6.81208700	-0.17060600	0.90483700
H	-7.73795800	0.11023000	1.41170100
C	1.90733200	-5.60316100	-1.66185500
H	1.56729700	-6.62245500	-1.85763200
C	5.62294000	-2.03873000	-1.05835200
H	5.39778900	-2.58380800	-1.97709400
C	-5.84051400	2.98416100	-2.55757700
H	-5.57795900	2.39452000	-3.44945100
H	-6.70880500	3.60803800	-2.81754900
H	-6.15734600	2.28930700	-1.76416400
C	4.87645900	-0.76166200	0.85896900
H	4.08169400	-0.29548300	1.43471200
C	-4.32428900	4.84807300	-3.24946500
H	-3.52094800	5.54428200	-2.96514600
H	-5.20395100	5.45027300	-3.52665600
H	-3.99109500	4.29623800	-4.14196900
C	6.94335600	-1.92491900	-0.61919100
H	7.74869000	-2.37728400	-1.20159700
C	6.33507600	2.56346700	-1.27910900
H	7.27983600	3.12589200	-1.32075300

H	6.21645200	2.04426700	-2.24253400
H	6.43611600	1.80329700	-0.48934000
C	5.42151000	4.22183400	0.35020800
H	5.46085200	3.48333600	1.16732000
H	4.63197000	4.95078800	0.58873900
H	6.38064400	4.76325100	0.33448100
C	6.19193200	-0.67301300	1.30969400
H	6.40842000	-0.15067200	2.24409800
C	7.23099100	-1.24422600	0.56640700
H	8.26269600	-1.16315700	0.91525900
C	5.12758000	4.58068900	-2.12960900
H	6.08721700	5.11807000	-2.18755300
H	4.33799400	5.32874900	-1.96332000
H	4.94110000	4.10567800	-3.10503700
Si	-2.17238800	-0.63071300	4.44437100
C	-2.09684800	-0.64580700	6.34471300
H	-1.91861000	-1.65618400	6.74669800
H	-1.32788300	0.02365900	6.76434000
H	-3.06981400	-0.29989900	6.73449600
C	-2.38975300	1.19069200	4.00799900
H	-1.42555200	1.66567000	3.77350700
H	-3.02295900	1.29115500	3.11406800
H	-2.84688200	1.74080300	4.84583300
C	-3.53113500	-1.86080900	4.03408300
H	-3.09145100	-2.86274500	3.90310200
H	-4.26587800	-1.91934400	4.85260900
H	-4.04191600	-1.59125500	3.09798300
C	-1.51302700	-0.89237900	2.22014600
N	-0.96235700	-1.14128500	1.22065000
C	1.60435300	2.24881600	4.68522200
C	1.01917900	0.98722300	4.78081600
C	1.20976500	0.04727000	3.75095500
C	1.95353900	0.40223300	2.61019200
C	2.50929400	1.67268400	2.50540100
C	2.35099300	2.58826400	3.55279600
H	1.48014000	2.96765200	5.49736500
H	0.44926800	0.70894000	5.66762300
H	2.04353100	-0.30315100	1.78572600
H	3.05575000	1.95787700	1.60589500
H	2.80547200	3.57848400	3.47674500
C	0.66418100	-1.30514300	3.89182700
O	-0.44373000	-1.51155600	4.43282400
C	1.40173700	-2.47827800	3.34371400
H	2.49157800	-2.36231600	3.39198100

H	1.06902600	-3.39865100	3.84011100
H	1.09781900	-2.54653000	2.28109100

Optimized structure of **IN1** (B3LYP-D3BJ)

Sb	-0.04142600	-2.02329000	-0.75323500
Cl	0.03038900	-2.68873000	-3.09746900
N	0.01883100	0.01556900	-1.27711800
N	2.23357600	-1.39407800	-0.74176600
N	-2.26785000	-1.32487900	-0.83397400
C	-1.16950000	0.66015800	-1.58214700
C	3.03959800	-2.04700300	0.05701500
C	-1.20096000	1.95214400	-2.14589000
C	1.23521200	0.59961400	-1.58953200
C	-4.27225800	-1.37805000	0.59883200
C	0.07035400	2.91867900	-4.06979500
H	0.03420300	1.95611000	-4.60050100
H	0.97163100	3.45812400	-4.39678800
H	-0.80535800	3.51129300	-4.37290700
C	-2.38933900	-0.00734500	-1.30672900
C	-2.44456700	2.54016300	-2.37439500
H	-2.46632000	3.54431400	-2.79851300
C	0.13482700	4.05857700	-1.81172900
H	-0.73969100	4.67444700	-2.06631200
H	1.03130200	4.62604500	-2.10096000
H	0.15192100	3.92704900	-0.72081700
C	-2.88107700	-3.43640300	0.08860200
C	-3.10065400	-4.03036900	1.34669000
H	-3.49797300	-3.42686700	2.16431100
C	3.79486100	1.69340900	-2.15768000
C	0.08339200	2.69346900	-2.53789800
C	-2.37651200	-4.22326700	-0.96695800
H	-2.24928400	-3.78545700	-1.95791900
C	1.89701700	-5.60642800	-0.51077800
H	1.52124200	-6.22112400	-1.33121100
C	-4.99412400	2.62968500	-2.36578200
C	2.25927600	-4.28135800	-0.75362700
H	2.17965700	-3.87152900	-1.76159500
C	-3.61664800	0.60957300	-1.58868500
H	-4.53105400	0.05608000	-1.40600800
C	2.60306800	2.38973100	-2.43612600
H	2.67379900	3.37991500	-2.88707900
C	4.16827600	-1.42584000	0.78702000
C	3.67790000	0.42769500	-1.58979100
H	4.56200100	-0.16503100	-1.37855100

C	2.42149600	-0.11221200	-1.28277000
C	-5.15059500	0.33390500	2.07210300
H	-4.98849900	1.20683100	2.70730800
C	-5.55739100	-1.93032500	0.48065600
H	-5.70995000	-2.81847000	-0.13612000
C	-2.06142700	-5.56564400	-0.75388900
H	-1.67520200	-6.16566100	-1.58040900
C	-2.77561800	-5.36936200	1.55395200
H	-2.92898400	-5.81833700	2.53754800
C	-3.12267500	-1.99371800	-0.10425000
C	1.33062700	1.87894300	-2.17480800
C	-3.66901700	1.90141200	-2.10360500
C	2.74995700	-3.47526300	0.29418800
C	-6.63899800	-1.32924400	1.12797700
H	-7.64060200	-1.74946000	1.01591500
C	-4.07267500	-0.24813700	1.41026100
H	-3.06872700	0.15864500	1.52158000
C	5.15434700	2.34529900	-2.44429000
C	-2.25124800	-6.13754400	0.50727100
H	-2.00361100	-7.18826300	0.67403500
C	2.90577200	-4.03500200	1.57722000
H	3.29197100	-3.41882800	2.39077500
C	-5.04285800	3.90118100	-1.49150000
H	-4.98775600	3.64288500	-0.42261000
H	-5.98183700	4.45043700	-1.66421300
H	-4.21034900	4.58491300	-1.71686900
C	2.53414500	-5.35706200	1.81411800
H	2.63856600	-5.77822600	2.81624600
C	-6.43792400	-0.19617600	1.92110000
H	-7.28354900	0.26995100	2.43145200
C	2.02483600	-6.14306400	0.77354000
H	1.73924800	-7.18000700	0.96393500
C	5.41636400	-2.06673600	0.85597000
H	5.56138900	-3.02518400	0.35354000
C	-6.20661400	1.75448900	-2.01180100
H	-6.24270800	0.83989900	-2.62356000
H	-7.13578600	2.31316500	-2.19932300
H	-6.20377200	1.45953300	-0.95095500
C	3.98426900	-0.19785000	1.44641800
H	3.02080100	0.30555600	1.39844200
C	-5.08560200	3.01980800	-3.85512800
H	-4.26432000	3.68773400	-4.15576600
H	-6.03199100	3.54581600	-4.05712700
H	-5.04571700	2.12674500	-4.49776900

C	6.47338800	-1.46791500	1.54462000
H	7.44712300	-1.96096300	1.57837300
C	6.32096000	1.39722600	-2.12605400
H	7.27655300	1.89710500	-2.34415300
H	6.27960400	0.48165400	-2.73601000
H	6.33471700	1.10512200	-1.06447700
C	5.29479600	3.60041700	-1.55545700
H	5.25453700	3.32757500	-0.48925400
H	4.49509200	4.33134900	-1.75083200
H	6.25752000	4.10101500	-1.74461300
C	5.03272700	0.38489100	2.15217400
H	4.87063400	1.33495700	2.66476400
C	6.28365200	-0.24403300	2.19299500
H	7.11038500	0.21852900	2.73672200
C	5.23883900	2.74807600	-3.93040100
H	6.21225400	3.21582600	-4.14618000
H	4.45648500	3.47072500	-4.20683300
H	5.12965400	1.86755900	-4.58213900
Si	-1.76669300	3.41082400	2.94730600
C	-0.62072000	4.89090600	3.05131200
H	-0.25577300	5.04157600	4.07917900
H	0.25201300	4.79984400	2.38839400
H	-1.17709400	5.79672600	2.75653800
C	-2.17019300	3.01617000	1.15922200
H	-1.31003400	2.61262300	0.60889800
H	-2.99404000	2.29573700	1.06034000
H	-2.48060500	3.93573900	0.63756200
C	-3.32031700	3.70076600	3.94784200
H	-3.07601800	3.99017300	4.98208700
H	-3.92834800	4.50576800	3.50394800
H	-3.93661400	2.78907000	3.99317900
C	-0.26753100	0.22934000	2.47796300
N	-0.55301100	-0.61209800	1.73640100
C	3.02819500	3.70783500	3.35396000
C	2.02729100	2.86257400	3.83590100
C	1.22469300	2.13255900	2.94809000
C	1.44558700	2.26488600	1.57071600
C	2.45016300	3.10598400	1.08656600
C	3.24307600	3.83143900	1.97767500
H	3.64128300	4.27473000	4.05801700
H	1.86613300	2.78924500	4.91154700
H	0.83961700	1.69945700	0.86193100
H	2.61626000	3.17809800	0.01258600
H	4.02832500	4.48979100	1.59990900

C	0.06361900	1.28375800	3.48263200
O	-1.09440500	2.04157700	3.72360600
C	0.40698800	0.54201400	4.78505100
H	0.56660400	1.27739400	5.58344000
H	-0.44102400	-0.09258200	5.07532300
H	1.30908300	-0.07501200	4.66761100

Optimized structure of **TS2** (B3LYP-D3BJ)

Sb	0.27397000	1.65874000	-0.88576200
Cl	0.13997800	1.83274000	-3.34264400
N	-0.01266800	-0.43209200	-1.01654100
N	-2.12335900	1.28074600	-0.89505900
N	2.39453000	0.79964800	-1.18868300
C	1.08647500	-1.18265000	-1.42135600
C	-2.92886600	2.23072400	-0.51625100
C	0.97944900	-2.55941200	-1.71486400
C	-1.28695100	-0.93951200	-1.20519800
C	4.63271900	0.75940200	-0.15842800
C	-0.51812100	-4.57017300	-2.11641600
H	-0.62365900	-4.36275600	-3.19132400
H	-1.41382900	-5.10664300	-1.77531300
H	0.32292800	-5.26223900	-1.97560600
C	2.34022400	-0.55020400	-1.56955200
C	2.09245900	-3.21990500	-2.22974400
H	2.00687800	-4.28040600	-2.45943100
C	-0.16442500	-3.65039200	0.18925500
H	0.71480900	-4.29460300	0.34362900
H	-1.06326400	-4.18465500	0.52750200
H	-0.04293500	-2.74951200	0.80700600
C	3.29669700	2.89055500	-0.50078600
C	3.70909200	3.50218200	0.69756300
H	4.14917900	2.89220300	1.48794500
C	-3.90765600	-1.92196600	-1.71186300
C	-0.30598600	-3.28083000	-1.31118200
C	2.72156900	3.67714700	-1.51880100
H	2.42662500	3.21289700	-2.46190000
C	-1.15138300	5.47779600	-1.35822700
H	-0.59052000	5.91237800	-2.18807400
C	4.50964000	-3.37905200	-3.03433700
C	-1.66687100	4.18571700	-1.48066700
H	-1.51086000	3.62148300	-2.40115000
C	3.43071900	-1.23917000	-2.11952800
H	4.35846900	-0.69650600	-2.27229700
C	-2.78817800	-2.77136800	-1.68864000

H	-2.93641000	-3.83314400	-1.85963400
C	-4.36726700	2.08166500	-0.15221400
C	-3.69123700	-0.56831200	-1.45644600
H	-4.52927800	0.11872200	-1.48482500
C	-2.41176700	-0.06711500	-1.17109000
C	5.67755400	-1.01862400	1.11878700
H	5.58866100	-1.91990300	1.72929200
C	5.89847400	1.31741600	-0.40219000
H	5.98046500	2.23468500	-0.98853700
C	2.54806000	5.04886700	-1.32874000
H	2.11472500	5.65347100	-2.12783400
C	3.51735800	4.86968900	0.88515600
H	3.82071500	5.33580800	1.82502900
C	3.41305000	1.42700700	-0.66655800
C	-1.49226100	-2.31807500	-1.44486600
C	3.32913100	-2.58251800	-2.46295500
C	-2.40167800	3.61695700	-0.42500600
C	7.04777500	0.69250500	0.08626500
H	8.02970900	1.12180900	-0.12302200
C	4.53178100	-0.41355500	0.61168600
H	3.54923000	-0.83509600	0.81604800
C	-5.33697000	-2.44594400	-1.91888300
C	2.93726200	5.64481300	-0.12613400
H	2.79806500	6.71803100	0.02226100
C	-2.64492800	4.37753800	0.73365200
H	-3.24419300	3.95208400	1.54138000
C	4.86544500	-4.51653500	-2.05313100
H	5.14736200	-4.10900700	-1.06943100
H	5.71421000	-5.10443100	-2.43719300
H	4.02210700	-5.20758600	-1.90375400
C	-2.10935100	5.65861100	0.85767300
H	-2.28386600	6.23436900	1.76919300
C	6.93949800	-0.47455600	0.84707700
H	7.83832000	-0.95856500	1.23549900
C	-1.35907700	6.21052300	-0.18732000
H	-0.95054500	7.21899100	-0.09198700
C	-5.31843200	2.84145400	-0.85274200
H	-4.99083300	3.51225100	-1.64958600
C	5.75480600	-2.49877200	-3.22319600
H	5.56745400	-1.67288900	-3.92665800
H	6.57820900	-3.10236000	-3.63365200
H	6.10136800	-2.06949500	-2.27018400
C	-4.79333500	1.24452900	0.89043800
H	-4.06861200	0.65635800	1.44762100

C	4.11644500	-3.97543100	-4.40131600
H	3.25211600	-4.65166100	-4.32014200
H	4.95339200	-4.55420000	-4.82305700
H	3.85501300	-3.17965400	-5.11579800
C	-6.67603400	2.72684300	-0.54702200
H	-7.40920300	3.30628500	-1.11199800
C	-6.05917200	-2.42019900	-0.55313000
H	-7.08870700	-2.80113100	-0.65053100
H	-6.11417700	-1.39642500	-0.15260300
H	-5.52707700	-3.04523400	0.18106100
C	-5.34654900	-3.88745200	-2.45200900
H	-4.90797700	-4.59549600	-1.73233800
H	-4.79479800	-3.97363900	-3.40083400
H	-6.38254800	-4.21016200	-2.63438400
C	-6.14670200	1.15263300	1.21219600
H	-6.46461600	0.50272100	2.03015400
C	-7.09314400	1.88344100	0.48627200
H	-8.15410100	1.80109200	0.73159200
C	-6.09742500	-1.55351500	-2.91941800
H	-7.11322200	-1.94620000	-3.08160700
H	-5.58286000	-1.52203700	-3.89220000
H	-6.20357600	-0.52018700	-2.55683300
Si	2.21515600	-0.93968300	4.28902600
C	2.13889800	-1.65404000	6.05496100
H	2.05058700	-0.86472100	6.81886200
H	1.30717300	-2.36194800	6.21029100
H	3.07251400	-2.20800700	6.25403200
C	2.29416500	-2.48989100	3.21147200
H	1.29085700	-2.79212900	2.87614600
H	2.88833900	-2.30690000	2.30425000
H	2.73244600	-3.33064300	3.77245200
C	3.68768000	0.22722100	4.35299300
H	3.34204500	1.23097200	4.64930600
H	4.42048700	-0.11017700	5.10291600
H	4.17909400	0.31564600	3.37342500
C	1.61406400	0.16592900	2.36136200
N	1.14274500	0.80895200	1.50771700
C	-1.82243900	-3.30381600	3.46066400
C	-1.10068900	-2.24905300	4.00605000
C	-1.23710200	-0.94037100	3.50323700
C	-2.09855100	-0.72395100	2.40304000
C	-2.79036400	-1.76871100	1.82743200
C	-2.67253100	-3.06944500	2.36252900
H	-1.71954300	-4.29961200	3.88880700

H	-0.45358400	-2.42870000	4.86487600
H	-2.16689200	0.27122100	1.96537600
H	-3.41899000	-1.62504700	0.94997800
C	-0.52734700	0.16392200	4.12021200
O	0.58761000	0.00421800	4.67804500
C	-1.07069500	1.55319400	4.06799300
H	-2.16673200	1.59205900	4.08121100
H	-0.64187800	2.14879900	4.88401200
H	-0.70506200	1.98493200	3.11688900
O	-3.39010900	-4.01766700	1.74759500
C	-3.33634000	-5.36122500	2.20601700
H	-3.68417400	-5.44058400	3.24888000
H	-4.00752900	-5.93302100	1.55448600
H	-2.31451300	-5.76712400	2.12491800

Optimized structure of **IN2** (B3LYP-D3BJ)

Sb	0.15197400	2.03516400	-0.54039900
Cl	0.03048400	2.68254700	-2.89207800
N	-0.08741700	0.01057000	-1.03475100
N	-2.18261000	1.64166200	-0.55602700
N	2.30526300	1.25135300	-0.87444900
C	1.03520400	-0.68517700	-1.46966100
C	-2.96179000	2.42750300	0.13988400
C	0.94523300	-1.98397900	-2.01370000
C	-1.35771800	-0.46014000	-1.34727700
C	4.58867300	1.31542000	0.08214100
C	-0.52284200	-3.32938600	-3.54755400
H	-0.47935100	-2.55253100	-4.32473200
H	-1.46804700	-3.87777900	-3.65944200
H	0.28439000	-4.05051300	-3.73474200
C	2.30465600	-0.05714800	-1.38852900
C	2.12224000	-2.61402700	-2.41490700
H	2.05357800	-3.63043000	-2.80108900
C	-0.47217700	-3.82244100	-1.07039500
H	0.33676000	-4.55392000	-1.22056000
H	-1.43556300	-4.35103700	-1.12592000
H	-0.37805500	-3.40244700	-0.06122300
C	3.07637700	3.33330800	0.00370700
C	3.41402600	3.82531600	1.27830300
H	3.86043300	3.15262900	2.01255900
C	-3.99527900	-1.32159700	-1.96401700
C	-0.40141800	-2.70494400	-2.13915000
C	2.51186900	4.20414200	-0.94774100

H	2.28496900	3.84005300	-1.95030600
C	-1.41347000	5.84725300	-0.35906600
H	-0.94005100	6.42122400	-1.15797500
C	4.63386000	-2.76165600	-2.84384300
C	-1.88579700	4.56096700	-0.62379200
H	-1.79286400	4.14302100	-1.62665000
C	3.45054100	-0.70213000	-1.87712300
H	4.39325400	-0.16792300	-1.86571900
C	-2.87412800	-2.13433500	-2.18640100
H	-3.02532200	-3.13290000	-2.58692700
C	-4.26063900	2.02643000	0.73618500
C	-3.76982400	-0.05291200	-1.42637400
H	-4.60666600	0.62006300	-1.26166600
C	-2.47986800	0.37388300	-1.08727500
C	5.90537400	-0.41772800	1.14387100
H	5.95923300	-1.34656100	1.71198100
C	5.77159100	1.98711500	-0.26986400
H	5.71373900	2.93047400	-0.81639400
C	2.26153900	5.53691700	-0.61484300
H	1.83411800	6.20721700	-1.36321000
C	3.14410600	5.15191000	1.60844300
H	3.38623500	5.52217800	2.60686200
C	3.28590500	1.90142200	-0.30872800
C	-1.56462800	-1.73447600	-1.90575100
C	3.38793500	-1.99969400	-2.37221400
C	-2.51060100	3.81282300	0.39314200
C	7.01427500	1.43931600	0.05322700
H	7.92947200	1.95659800	-0.24185700
C	4.66320100	0.11358800	0.80668400
H	3.74626200	-0.39633300	1.09676800
C	-5.43246000	-1.79009200	-2.23561600
C	2.56600600	6.00907500	0.66406600
H	2.36507800	7.05054100	0.92488400
C	-2.68694200	4.39175400	1.66492500
H	-3.18749100	3.82356900	2.45093400
C	4.84923500	-3.97696300	-1.91669500
H	5.02601500	-3.64885300	-0.88021300
H	5.72422300	-4.56096200	-2.24328600
H	3.97820900	-4.64964000	-1.91450700
C	-2.19811500	5.67017200	1.92606000
H	-2.31834200	6.10386800	2.92103400
C	7.08239100	0.23490500	0.75726300
H	8.05328800	-0.19396800	1.01463700
C	-1.55770400	6.39885000	0.91643500

H	-1.18454700	7.40431900	1.12344600
C	-5.40318100	2.81283100	0.51495900
H	-5.33108900	3.70881400	-0.10494300
C	5.89441100	-1.88434700	-2.79167700
H	5.80460100	-1.00291600	-3.44519000
H	6.76471000	-2.46335200	-3.13523300
H	6.11039400	-1.53573500	-1.77017100
C	-4.35310600	0.88826900	1.55324100
H	-3.46852600	0.28557600	1.74573400
C	4.43210000	-3.24491900	-4.29452500
H	3.57284600	-3.92593700	-4.38687100
H	5.32383600	-3.78806900	-4.64484900
H	4.26157100	-2.39347000	-4.97130100
C	-6.62944000	2.43723400	1.06708900
H	-7.51853100	3.04128300	0.87456700
C	-6.20406400	-1.81904600	-0.89792600
H	-7.24363100	-2.14452800	-1.06339400
H	-6.23607500	-0.82732800	-0.42297700
H	-5.73129800	-2.52053900	-0.19344800
C	-5.46819300	-3.20176900	-2.84003300
H	-5.02221900	-3.94140700	-2.15760700
H	-4.93708000	-3.25013900	-3.80319300
H	-6.51122800	-3.50192200	-3.02078200
C	-5.57257300	0.53055800	2.12283300
H	-5.63121700	-0.35422900	2.75997400
C	-6.71624200	1.29679000	1.87068300
H	-7.67475000	1.00795300	2.30722300
C	-6.11897000	-0.81452900	-3.21239500
H	-7.15114700	-1.14013200	-3.41649000
H	-5.57786100	-0.76910300	-4.17021500
H	-6.16934400	0.20632300	-2.80399000
Si	2.62166300	-3.04720200	3.17028200
C	2.61579000	-4.81764400	3.77727400
H	2.57247000	-4.85367100	4.87747800
H	1.75595300	-5.37978300	3.38034000
H	3.53322900	-5.33956200	3.45886800
C	2.43089100	-2.98456000	1.30457300
H	2.25439900	-1.97721300	0.90199400
H	3.34005600	-3.37623100	0.82051900
H	1.58936400	-3.61511700	0.98447700
C	4.14733400	-2.15474000	3.79152100
H	4.17056900	-2.18993000	4.89256200
H	5.07043000	-2.63132700	3.42370100
H	4.15807700	-1.09624600	3.49043000

C	0.80444200	-0.36780300	2.62641800
N	1.19181300	0.49643400	1.95909700
C	-2.00081600	-4.31724900	2.32363000
C	-0.97807200	-3.60366700	2.95914300
C	-0.79305200	-2.24106600	2.71962600
C	-1.66827000	-1.59232500	1.83829800
C	-2.70011100	-2.28011900	1.21642600
C	-2.86656400	-3.65541100	1.44134000
H	-2.10656300	-5.38289200	2.52337500
H	-0.31671800	-4.11852800	3.65603900
H	-1.52557000	-0.53370900	1.61530200
H	-3.37232900	-1.78260500	0.52179800
C	0.27435500	-1.46297900	3.49805000
O	1.30988300	-2.28205200	3.94599600
C	-0.35420200	-0.79825100	4.73901800
H	-0.74782000	-1.59675900	5.38251700
H	0.41158900	-0.23495700	5.29091700
H	-1.17744500	-0.12790200	4.45466400
O	-3.86414900	-4.25231100	0.74581500
C	-4.10011700	-5.63487700	0.92668000
H	-4.36364300	-5.86767700	1.97275100
H	-4.94536000	-5.89336800	0.27662000
H	-3.22209100	-6.23642700	0.63389200

Optimized structure of **TS3** (B3LYP-D3BJ)

Sb	0.32695100	1.72461100	-0.82215100
Cl	0.20384000	1.91603400	-3.27498000
N	-0.00027100	-0.36047300	-0.96005300
N	-2.06818600	1.40347200	-0.82660500
N	2.42858000	0.82317000	-1.14633000
C	1.07471000	-1.12280500	-1.40846000
C	-2.85575500	2.36226300	-0.42761900
C	0.92934600	-2.48703200	-1.74094200
C	-1.28788200	-0.83630300	-1.15325600
C	4.69354500	0.71463400	-0.18504600
C	-0.61253000	-4.47334700	-2.12011000
H	-0.74037100	-4.25807800	-3.19106100
H	-1.50853900	-4.99473700	-1.75733500
H	0.21875800	-5.18147200	-2.00281400
C	2.33843800	-0.51373200	-1.56531400
C	2.01261500	-3.15403300	-2.30762000
H	1.89599600	-4.20406600	-2.56964600
C	-0.16548500	-3.58403600	0.17703800

H	0.74278600	-4.19510000	0.29161100
H	-1.01963200	-4.17371100	0.53630400
H	-0.05511000	-2.68939800	0.80612900
C	3.39294600	2.88069700	-0.43683000
C	3.85358100	3.45752200	0.76108200
H	4.30222800	2.82131500	1.52561100
C	-3.92472000	-1.73363600	-1.72953200
C	-0.35690600	-3.19387100	-1.31276700
C	2.81083700	3.70234200	-1.42263700
H	2.48278400	3.26601000	-2.36821100
C	-1.05113800	5.57748000	-1.32478800
H	-0.48475600	5.99126500	-2.16160500
C	4.40287300	-3.33752200	-3.18396000
C	-1.57512700	4.28693300	-1.42260000
H	-1.42309200	3.70528900	-2.33268200
C	3.39944900	-1.20824600	-2.16456700
H	4.33550800	-0.68191100	-2.32341300
C	-2.83214900	-2.61867300	-1.68880600
H	-3.00887400	-3.67116500	-1.88904200
C	-4.27909800	2.21197800	-0.01419500
C	-3.67583800	-0.39360000	-1.43490900
H	-4.49087400	0.32106700	-1.48101100
C	-2.38833500	0.06490400	-1.11485200
C	5.74062600	-1.12754100	0.99652700
H	5.65216500	-2.04795300	1.57789000
C	5.96257500	1.25341500	-0.45349300
H	6.04572100	2.18698500	-1.01329400
C	2.67547700	5.07345700	-1.19980000
H	2.23521900	5.70491500	-1.97391900
C	3.70131400	4.82511000	0.98153000
H	4.04321000	5.26446600	1.92107700
C	3.47230900	1.41891900	-0.63749600
C	-1.52705000	-2.20620400	-1.41459600
C	3.25731900	-2.53595900	-2.55186200
C	-2.31916300	3.74442800	-0.35898900
C	7.11326600	0.58796700	-0.02519500
H	8.09701200	1.00272200	-0.25429300
C	4.59277600	-0.48042500	0.54992000
H	3.60790300	-0.88715300	0.77215800
C	-5.34316600	-2.17832600	-2.11793400
C	3.11208100	5.63480300	0.00316000
H	3.00357900	6.70773300	0.17691900
C	-2.56310900	4.53165900	0.78217900
H	-3.16918800	4.12775400	1.59573000

C	4.75442400	-4.52150500	-2.25777000
H	5.07108100	-4.16052900	-1.26641900
H	5.57848300	-5.11349900	-2.68656400
H	3.89852100	-5.19801600	-2.11394800
C	-2.02058100	5.81173700	0.88130700
H	-2.19711900	6.40849100	1.77882000
C	7.00469600	-0.60182800	0.69944800
H	7.90505900	-1.11850000	1.03911400
C	-1.26071900	6.33640400	-0.17091600
H	-0.84726900	7.34454900	-0.09517500
C	-5.25825000	2.98854000	-0.65487500
H	-4.96448700	3.67906000	-1.44801700
C	5.66468100	-2.48082600	-3.37075300
H	5.48108000	-1.62390300	-4.03711300
H	6.46217000	-3.08770500	-3.82508800
H	6.04550200	-2.09752300	-2.41114800
C	-4.66226800	1.34739700	1.02260500
H	-3.91086400	0.75324100	1.53599000
C	3.95997300	-3.86923800	-4.56241800
H	3.08077200	-4.52638600	-4.48494300
H	4.77076400	-4.45149100	-5.02808000
H	3.70138100	-3.03950600	-5.23825200
C	-6.60174200	2.86413200	-0.29505800
H	-7.35830100	3.45661700	-0.81372300
C	-6.35885100	-1.67143200	-1.07297300
H	-7.37380400	-2.00390500	-1.34134400
H	-6.38013100	-0.57328000	-1.01385900
H	-6.12218000	-2.06432000	-0.07361500
C	-5.45940200	-3.70848400	-2.20567500
H	-5.20349300	-4.18776600	-1.24939500
H	-4.81345800	-4.12453700	-2.99406400
H	-6.49453000	-3.98600100	-2.45510000
C	-6.00025700	1.24266800	1.39739000
H	-6.28539800	0.56716500	2.20655700
C	-6.97546700	1.99202200	0.73091600
H	-8.02562900	1.89928900	1.01551900
C	-5.68198200	-1.57239700	-3.49693500
H	-6.69497600	-1.87168300	-3.80987500
H	-4.97057000	-1.91562700	-4.26393100
H	-5.64734700	-0.47205800	-3.47402700
Si	2.38764800	-1.22506200	4.19064200
C	2.32616700	-2.05255000	5.90061400
H	2.28223300	-1.31784800	6.72033500
H	1.47975500	-2.74837300	6.02598700

H	3.24760400	-2.64505800	6.03410300
C	2.35308800	-2.67890800	2.99625200
H	1.31925600	-2.97767300	2.76872000
H	2.83010000	-2.43138700	2.03813500
H	2.85892700	-3.54679300	3.44930800
C	3.86860400	-0.08006700	4.27433300
H	3.55174200	0.89303200	4.68267900
H	4.63497300	-0.49384900	4.94894800
H	4.30563100	0.09434200	3.28165000
C	1.68479400	0.08798700	2.34067300
N	1.14559300	0.81732700	1.60297100
C	-1.82554500	-3.34877300	3.37358900
C	-1.04716000	-2.37154300	3.98902700
C	-1.11790200	-1.03640900	3.55144100
C	-1.95397100	-0.69158400	2.47465500
C	-2.73325100	-1.66004400	1.85285400
C	-2.66215700	-2.96791900	2.32720800
H	-1.79368600	-4.39134500	3.68873000
H	-0.40650600	-2.63421100	4.83095700
H	-1.95768400	0.33007900	2.09728500
H	-3.37643300	-1.42831200	1.00509300
C	-0.34334000	0.00145000	4.25455700
O	0.79304200	-0.23127800	4.70636400
C	-0.88832300	1.37895300	4.39570100
H	-1.97992000	1.40483100	4.50206200
H	-0.38733900	1.90095800	5.22059600
H	-0.60736400	1.89402900	3.45571600
N	-3.48808900	-3.99746000	1.66517600
O	-4.49407200	-3.61475800	1.09383000
O	-3.10178700	-5.15077400	1.73115500

Optimized structure of **IN3** (B3LYP-D3BJ)

Sb	0.50175400	2.09156800	-0.59919000
Cl	0.46498800	2.84141200	-2.91772700
N	0.03602900	0.12206900	-1.17135800
N	-1.85690000	1.93907400	-0.59487600
N	2.55540300	1.02636300	-0.85598900
C	1.07505100	-0.70854200	-1.57156700
C	-2.53881000	2.72193000	0.20106900
C	0.84095000	-1.96177800	-2.17552900
C	-1.27318000	-0.18013700	-1.53423400
C	4.74983900	0.78311100	0.26773600
C	-0.64298700	-2.76326200	-4.02675200

H	-0.43313300	-1.86113400	-4.61968000
H	-1.63758300	-3.13756400	-4.30782100
H	0.08866000	-3.53542900	-4.30399600
C	2.41111600	-0.27207000	-1.37552100
C	1.94224900	-2.75704200	-2.48975800
H	1.75633700	-3.73843200	-2.92639400
C	-0.88666700	-3.72873000	-1.70276500
H	-0.18764100	-4.53794800	-1.96142000
H	-1.90860600	-4.08350400	-1.90132900
H	-0.80228200	-3.53713000	-0.62465500
C	3.54756400	2.99330800	0.05972000
C	3.89805000	3.46421200	1.33920800
H	4.22996800	2.75590000	2.10013500
C	-3.98576700	-0.67570000	-2.22582100
C	-0.57411600	-2.44766400	-2.51297600
C	3.13226600	3.91068900	-0.92499000
H	2.90201900	3.56009100	-1.93153900
C	-0.67076200	5.99419200	-0.13568400
H	-0.14878200	6.56030200	-0.90961200
C	4.43141100	-3.29365700	-2.65862200
C	-1.27896300	4.78178500	-0.46190000
H	-1.24586300	4.41549400	-1.48895000
C	3.48728300	-1.08056100	-1.77127700
H	4.49507300	-0.69785400	-1.66211600
C	-2.96555700	-1.59703200	-2.50493100
H	-3.22765800	-2.53464400	-2.98894800
C	-3.82426500	2.35031200	0.83888800
C	-3.61946600	0.50854500	-1.58135700
H	-4.37197800	1.26288100	-1.36604400
C	-2.29365700	0.75363400	-1.21166700
C	5.71918700	-1.08296100	1.47117300
H	5.59612600	-1.98729200	2.06813900
C	6.04066300	1.25504300	-0.02347800
H	6.16122900	2.17298500	-0.60223200
C	3.03500300	5.26977700	-0.62072200
H	2.71969200	5.97293800	-1.39434500
C	3.78440500	4.82013800	1.63948600
H	4.03596000	5.17659700	2.64060000
C	3.57626700	1.54184200	-0.22467900
C	-1.62010500	-1.37823300	-2.18199200
C	3.27592700	-2.34917500	-2.30123400
C	-1.96339700	4.04323400	0.52473500
C	7.16298100	0.54152300	0.40126500
H	8.16277900	0.90359300	0.15317600

C	4.59596700	-0.38562400	1.03311400
H	3.59544400	-0.73866500	1.27863200
C	-5.45886200	-0.91784200	-2.58513000
C	3.34990600	5.72427900	0.66234000
H	3.27061900	6.78725100	0.90085000
C	-2.05924600	4.56255200	1.83034300
H	-2.60002300	4.00243600	2.59504200
C	4.35430900	-4.52981400	-1.73665400
H	4.46097200	-4.23436100	-0.68080100
H	5.16110400	-5.24065700	-1.97551900
H	3.39642500	-5.06103800	-1.84527300
C	-1.43972200	5.76891100	2.15123100
H	-1.50082800	6.15550300	3.17069700
C	7.00352000	-0.62914900	1.14620400
H	7.88004500	-1.18692900	1.48286200
C	-0.74094600	6.48414200	1.17136700
H	-0.26281700	7.43247000	1.42649600
C	-4.90692200	3.24478200	0.82442100
H	-4.80107500	4.21456900	0.33409400
C	5.80012400	-2.62460400	-2.45750800
H	5.91797500	-1.73727600	-3.09847100
H	6.60142900	-3.33118200	-2.72065000
H	5.95839500	-2.31813300	-1.41215900
C	-3.96179200	1.11014700	1.48561800
H	-3.12305700	0.41744500	1.50994100
C	4.31260700	-3.73548500	-4.13133200
H	3.37330000	-4.27407900	-4.32673700
H	5.14136800	-4.41045100	-4.39657700
H	4.34960300	-2.86616300	-4.80584000
C	-6.12106600	2.88431500	1.41308700
H	-6.96575400	3.57550300	1.38049600
C	-6.30753500	-0.85906100	-1.29668200
H	-7.37051100	-1.02227000	-1.53444200
H	-6.22478800	0.11516100	-0.79167800
H	-5.99207500	-1.64121100	-0.59047800
C	-5.67138700	-2.28967100	-3.24302300
H	-5.37659800	-3.11032300	-2.57304400
H	-5.11029100	-2.38195800	-4.18595000
H	-6.73721000	-2.42249300	-3.48175200
C	-5.16570300	0.76374300	2.09148600
H	-5.26058800	-0.20066600	2.59499800
C	-6.25274900	1.64521600	2.04586000
H	-7.20179000	1.36655600	2.50882800
C	-5.92514800	0.18006300	-3.56376700

H	-6.98092800	0.02515200	-3.83630100
H	-5.32698800	0.16429300	-4.48805400
H	-5.83941000	1.18548100	-3.12349500
Si	1.86753300	-3.41784100	3.06142500
C	1.11787900	-5.07722000	3.49760500
H	1.03493700	-5.19779600	4.58966400
H	0.11681000	-5.20391700	3.05842200
H	1.75698800	-5.89221600	3.11904300
C	1.75034800	-3.11933600	1.21277800
H	0.71880400	-3.24089400	0.85367300
H	2.09625900	-2.12779000	0.89052600
H	2.36665800	-3.86437800	0.68401000
C	3.61560000	-3.28214400	3.71184400
H	3.64878200	-3.53663100	4.78313800
H	4.29508600	-3.96815900	3.18040700
H	3.99499300	-2.25484400	3.60238700
C	0.65878100	-0.28597200	2.59696400
N	1.15288400	0.50475000	1.91075600
C	-2.80937100	-3.74112000	2.89648400
C	-1.74354900	-3.06485000	3.48390700
C	-1.12782900	-1.99102100	2.82072000
C	-1.59381600	-1.60009400	1.56235100
C	-2.66647500	-2.26122900	0.96422200
C	-3.25150200	-3.32656300	1.63863900
H	-3.29783600	-4.58261400	3.38678900
H	-1.37070000	-3.38608600	4.45724700
H	-1.10274100	-0.79300800	1.02065400
H	-3.03702600	-1.97261400	-0.01549800
C	0.04695500	-1.29173000	3.51684600
O	1.02929200	-2.19664700	3.92705800
C	-0.44182400	-0.52492400	4.75987100
H	-0.88373900	-1.24269500	5.46369400
H	0.41178800	-0.03355100	5.24668000
H	-1.19791800	0.22554600	4.48814400
N	-4.35590700	-4.05721100	0.98490400
O	-4.61881700	-3.75378200	-0.17068500
O	-4.92623900	-4.91409800	1.63290000

VI. NMR Spectra

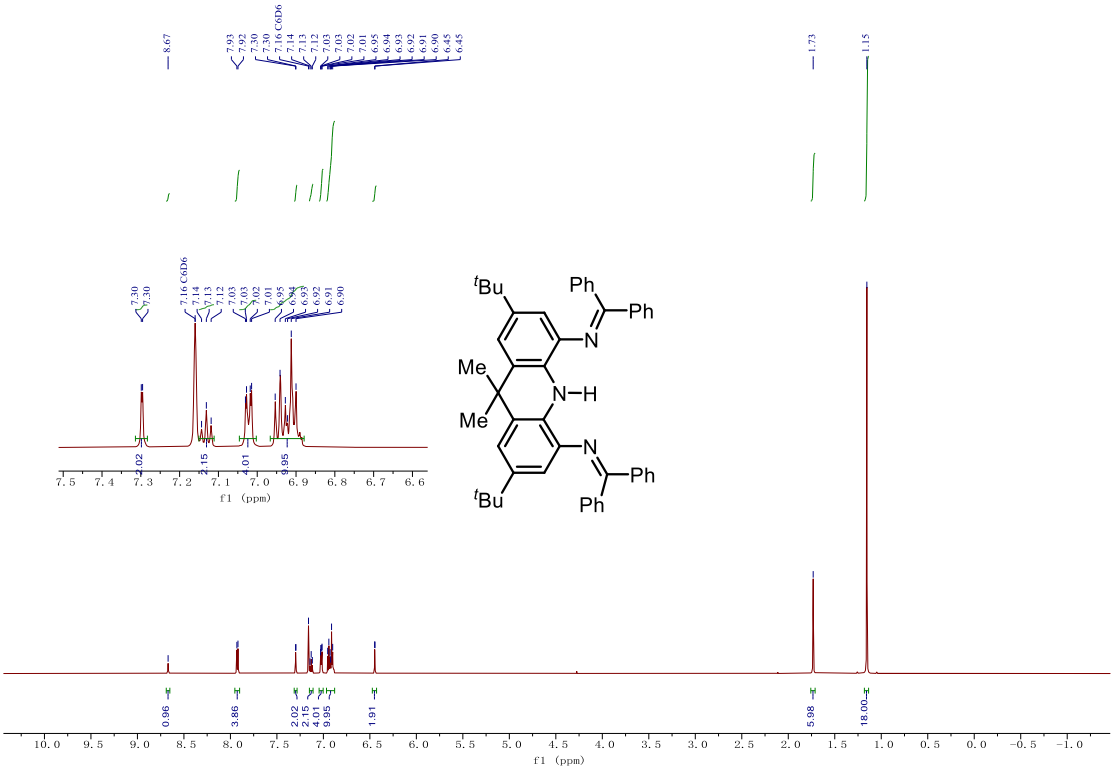


Figure S11. ^{1}H NMR spectrum of **1**.

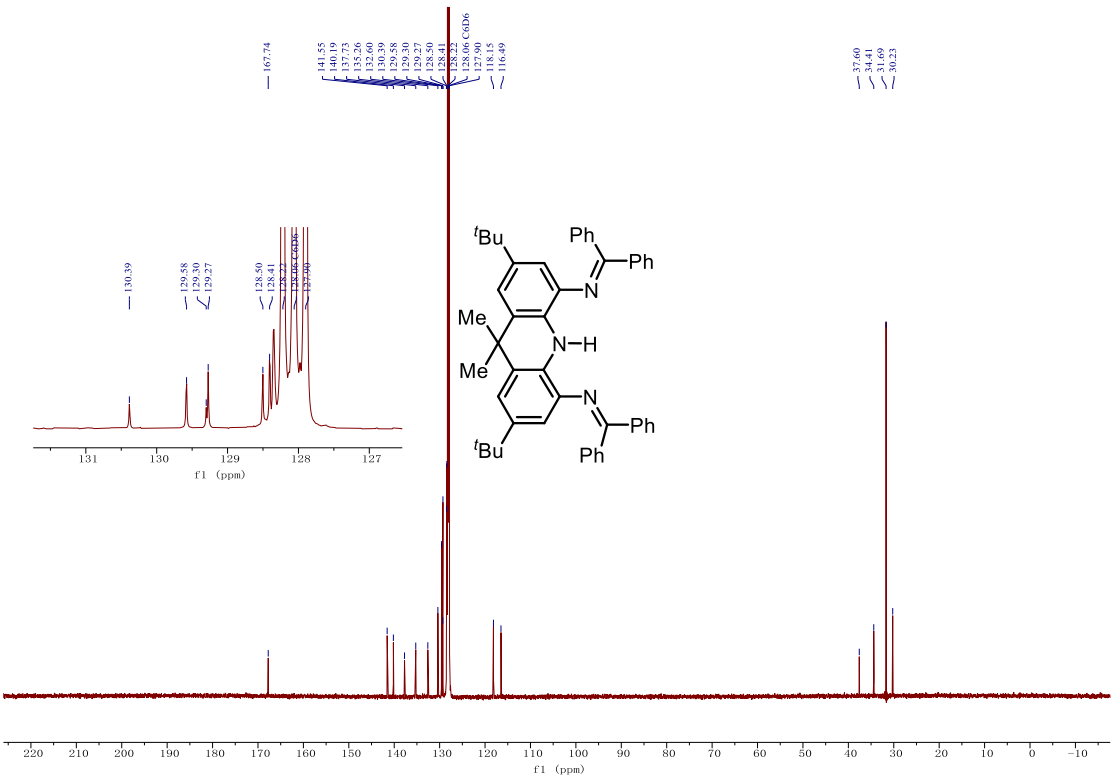


Figure S12. ^{13}C NMR spectrum of **1**.

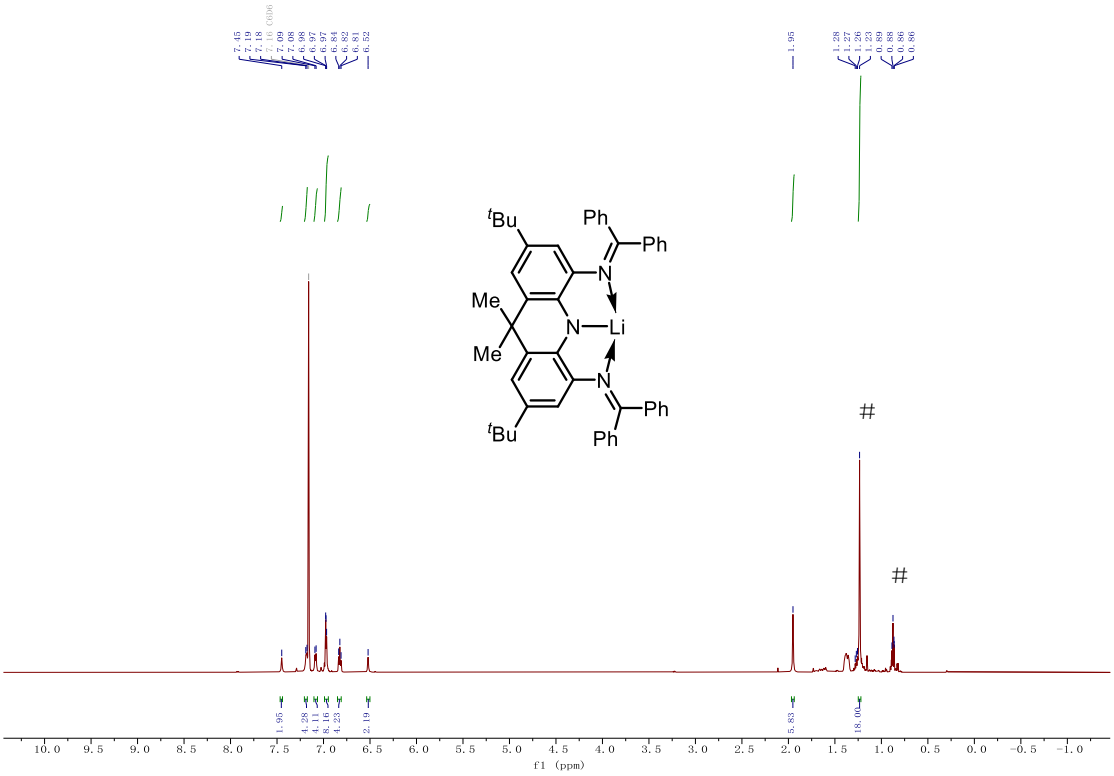


Figure S13. ^{1}H NMR spectrum of **2** (# indicate compound pentane).

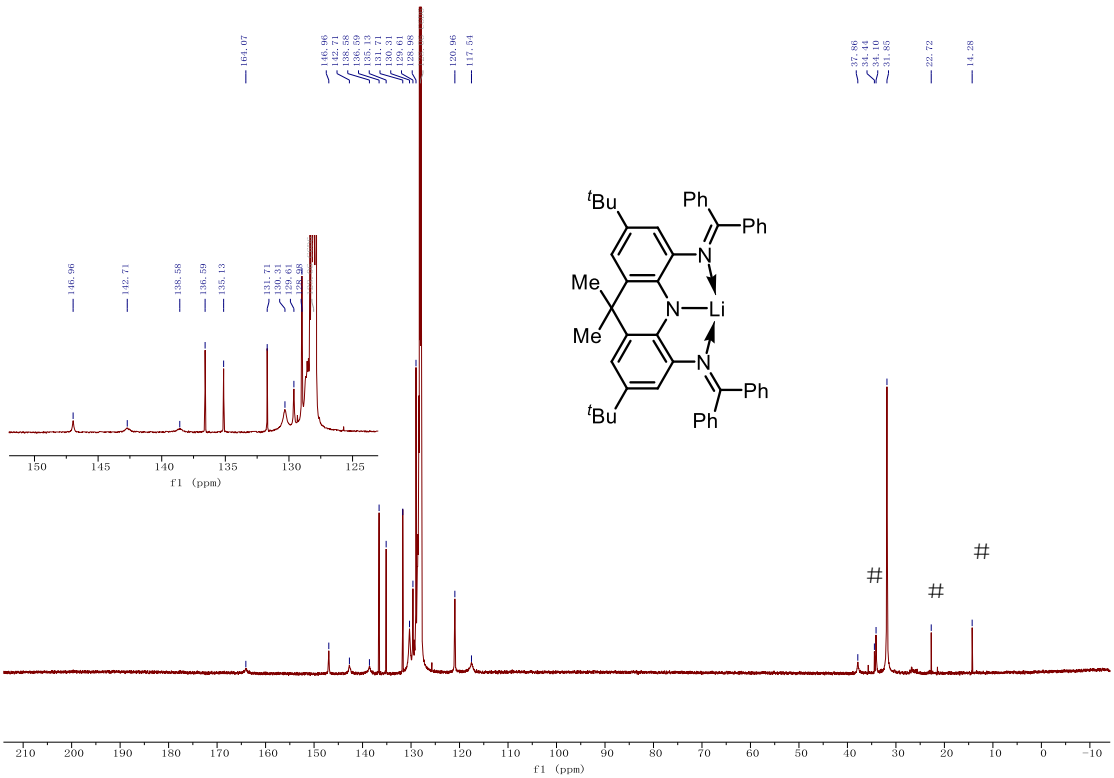


Figure S14. ^{13}C NMR spectrum of **2** (# indicate compound pentane).

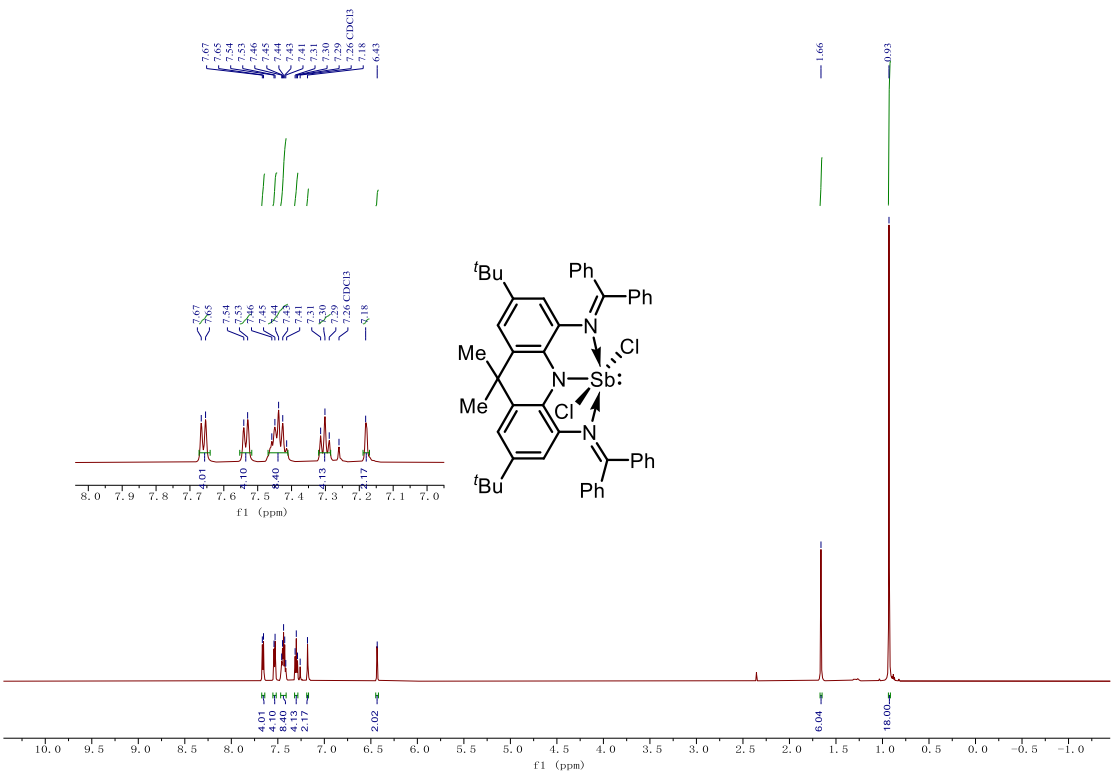


Figure S15. ^{1}H NMR spectrum of **3**.

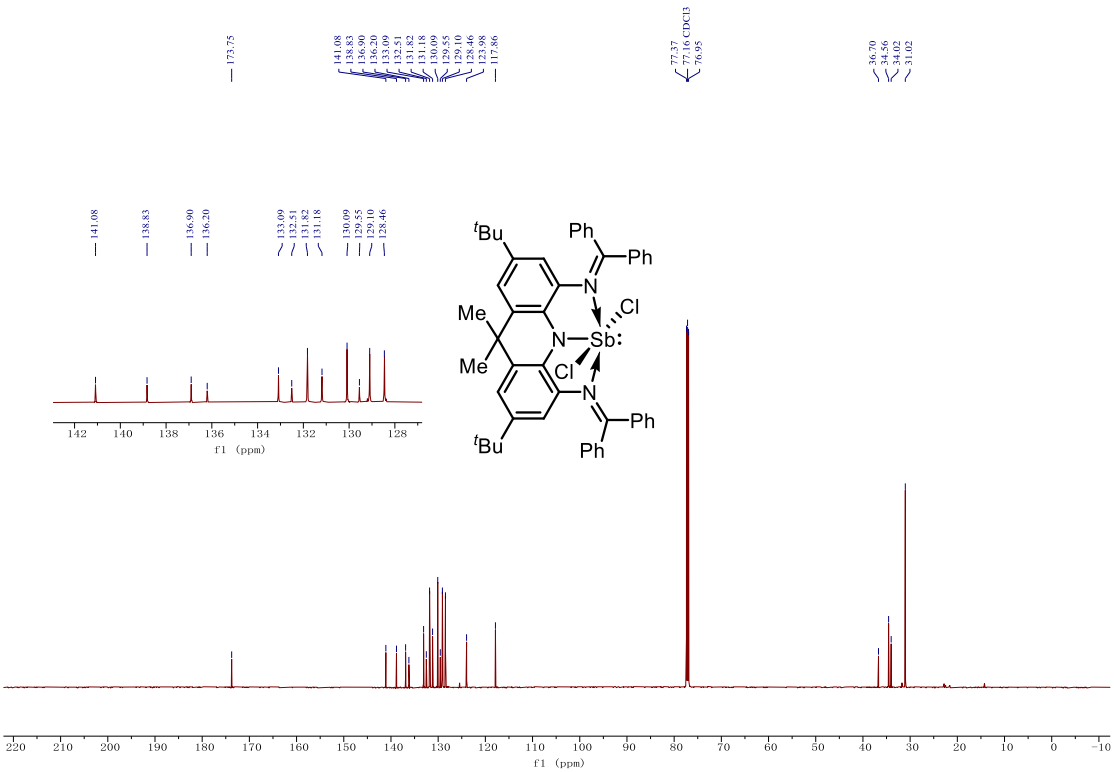


Figure S16. ^{13}C NMR spectrum of **3**.

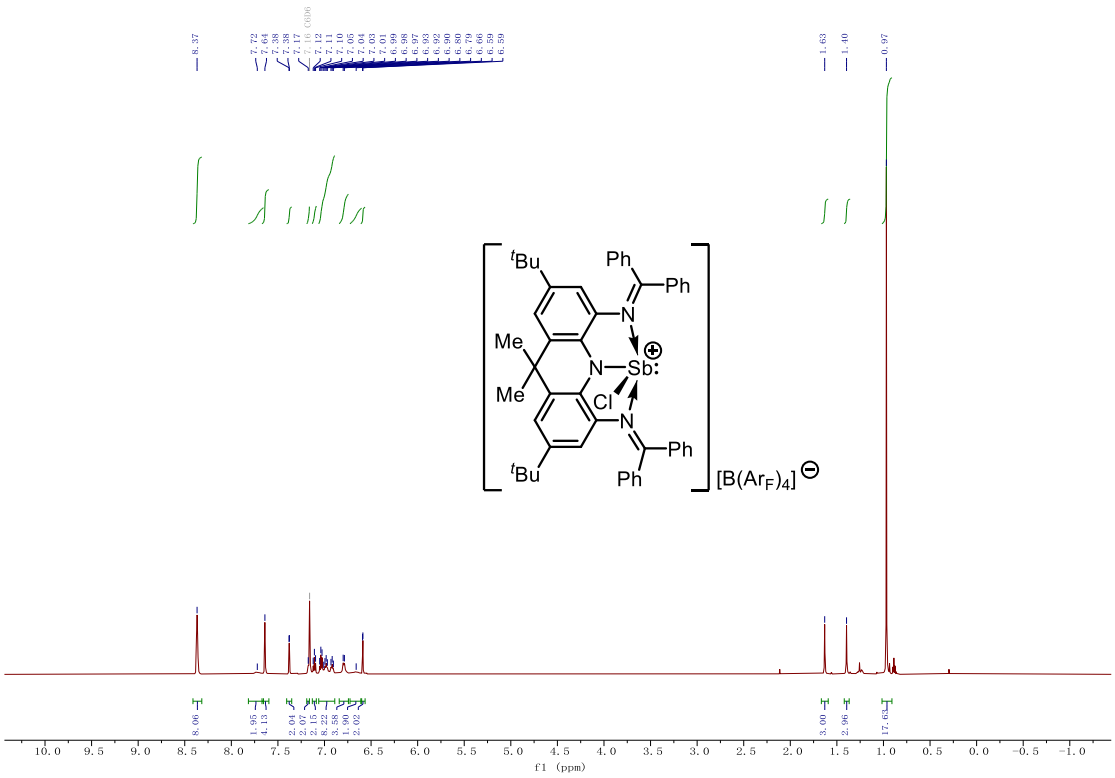


Figure S17. ^{1}H NMR spectrum of **4**[B(Ar$_F$)$_4$].

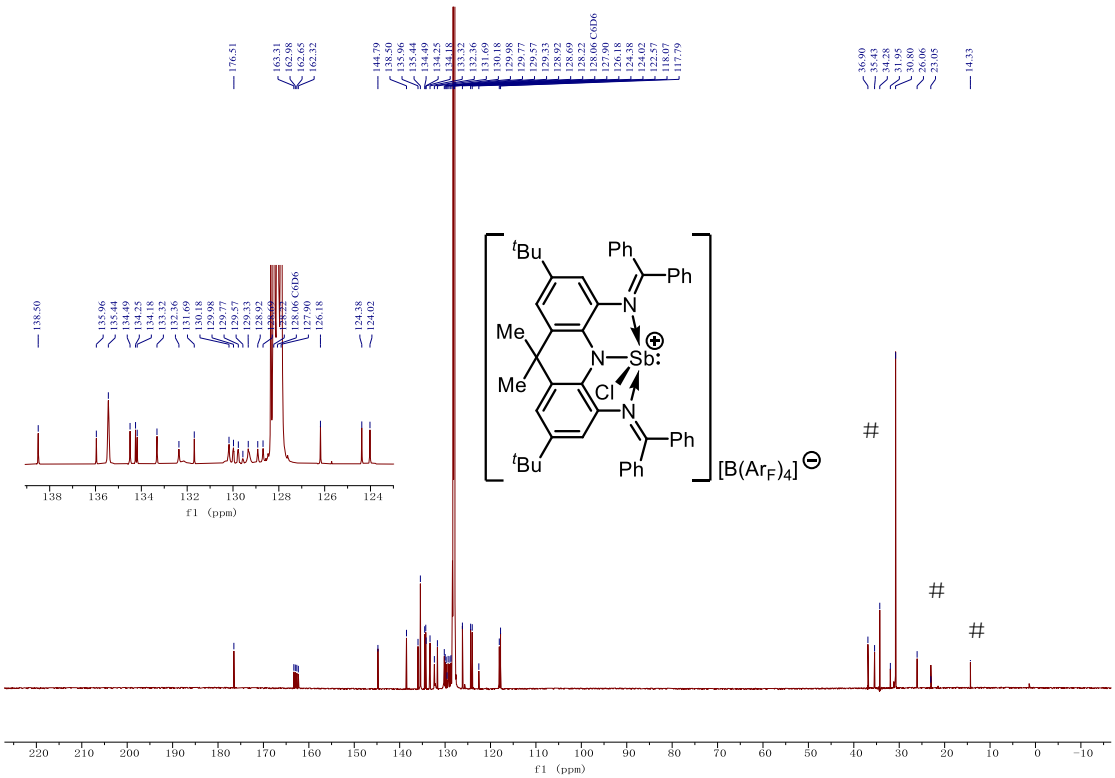


Figure S18. ^{13}C NMR spectrum of **4**[B(Ar$_F$)$_4$] (# indicates Hexane).

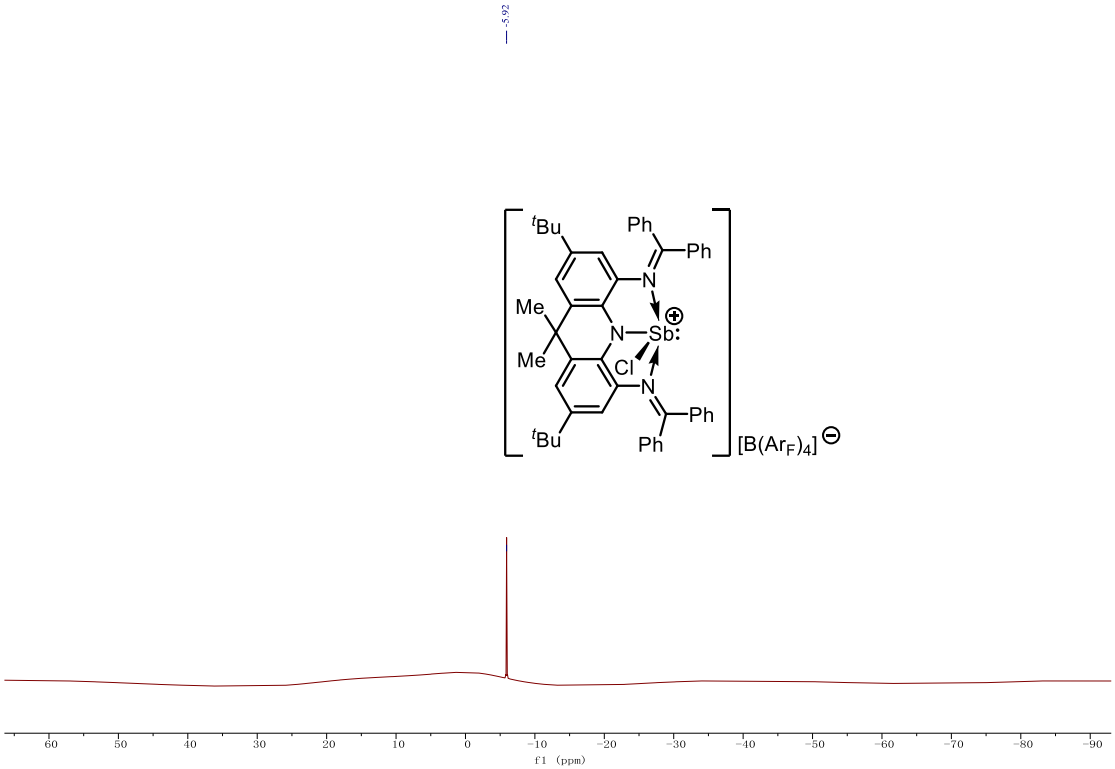


Figure S19. ^{11}B NMR spectrum of **4**[B(Ar$_F$)$_4$].

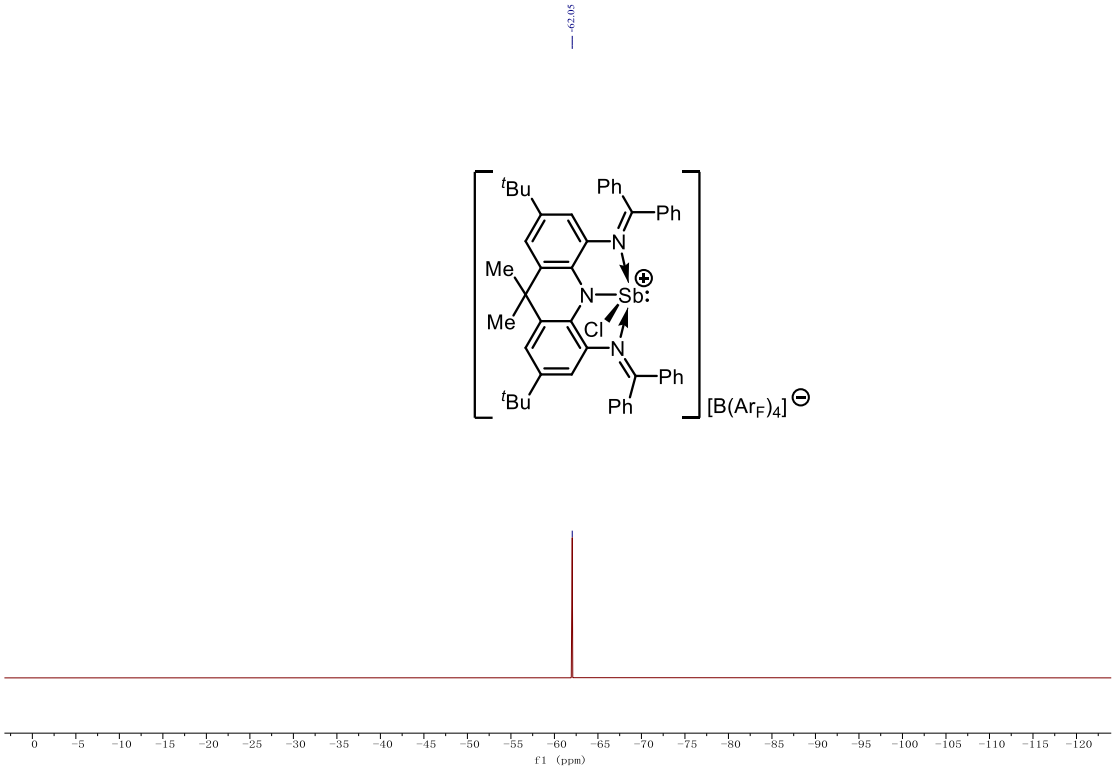


Figure S20. ^{19}F NMR spectrum of **4**[B(Ar$_F$)$_4$].

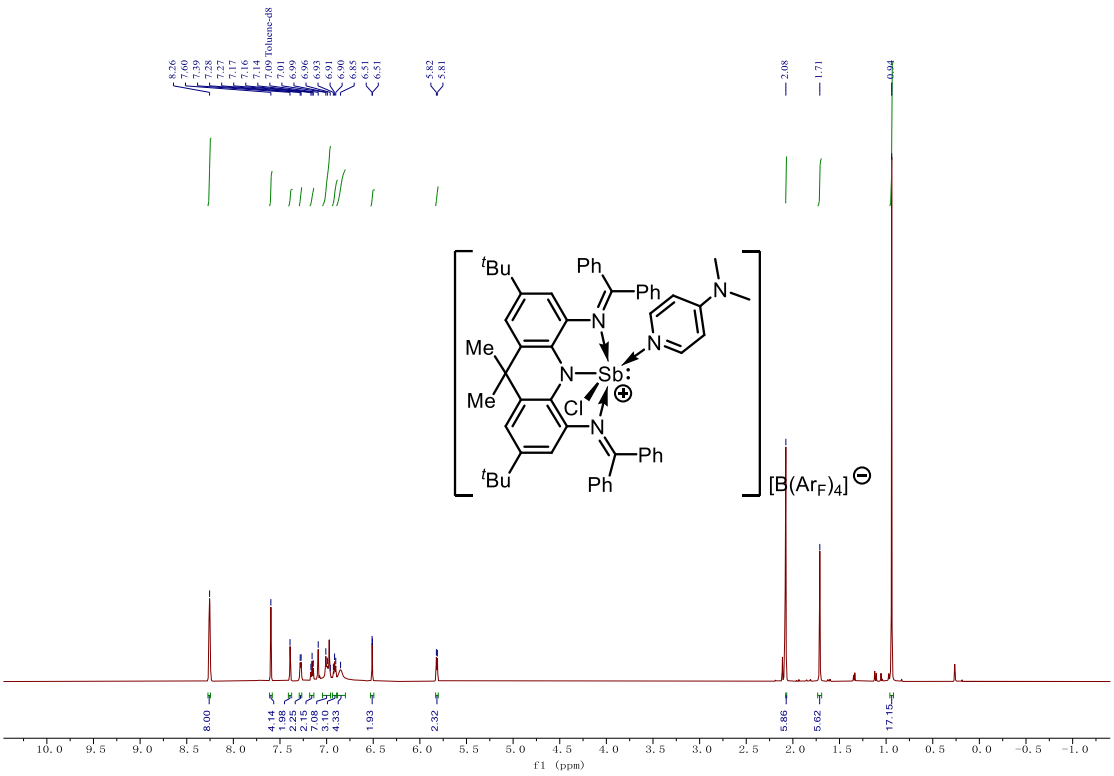


Figure S21. ^{1}H NMR spectrum of **5**[B(Ar$_F$)$_4$].

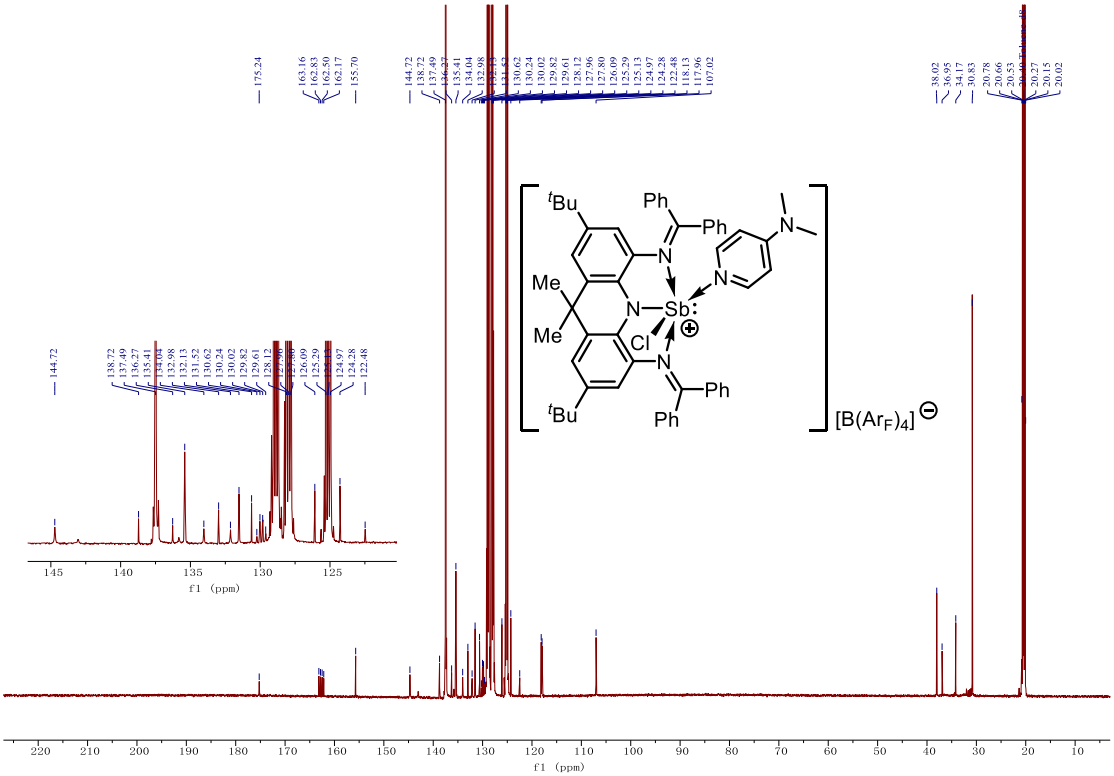


Figure S22. ^{13}C NMR spectrum of **5**[B(Ar$_F$)$_4$].

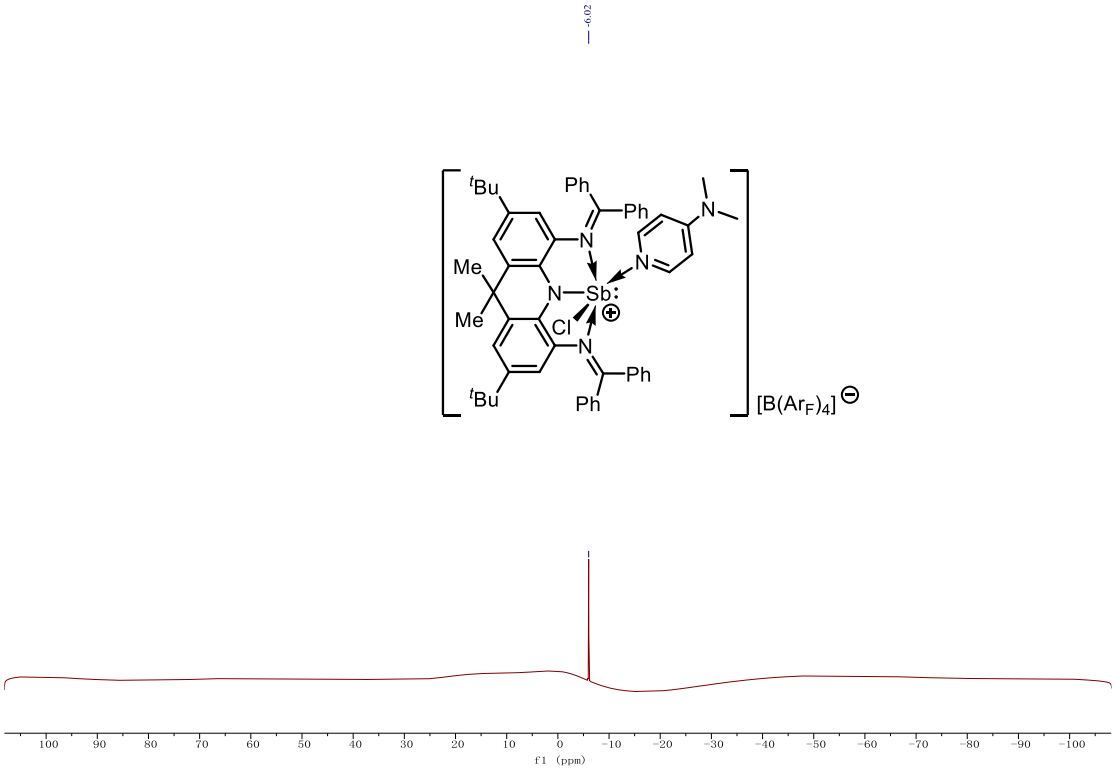


Figure S23. ^{11}B NMR spectrum of **5**[B(Ar$_F$)$_4$].

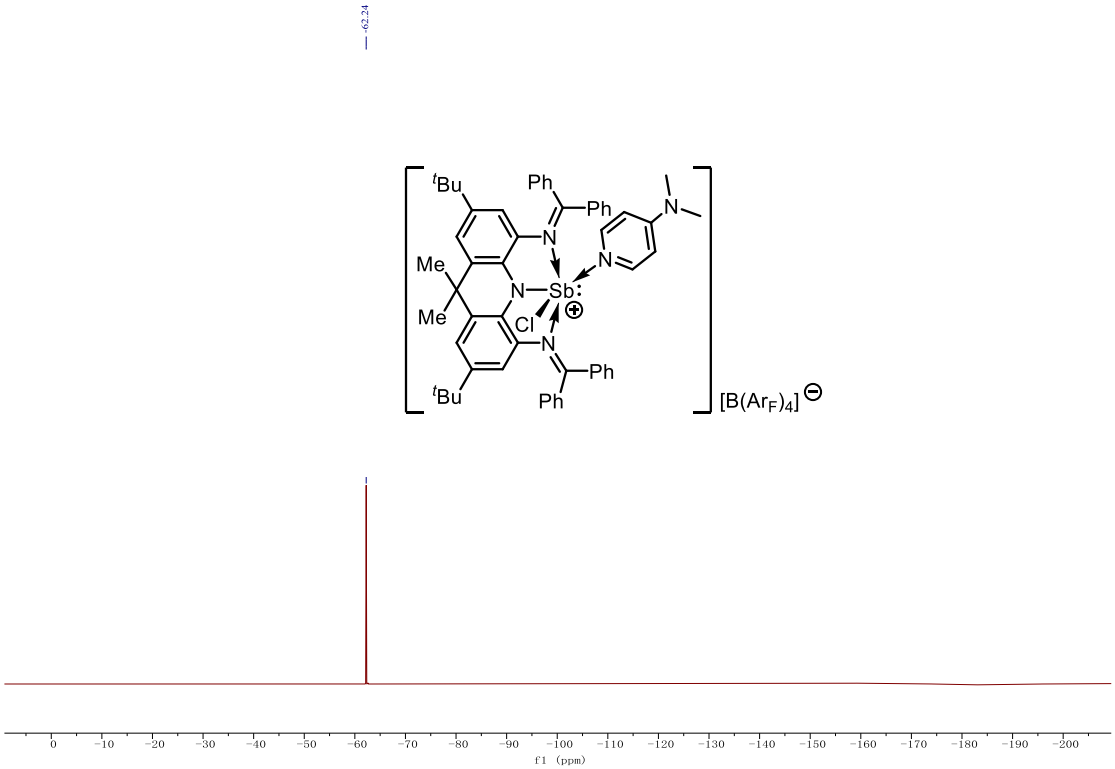


Figure S24. ^{19}F NMR spectrum of **5**[B(Ar$_F$)$_4$].

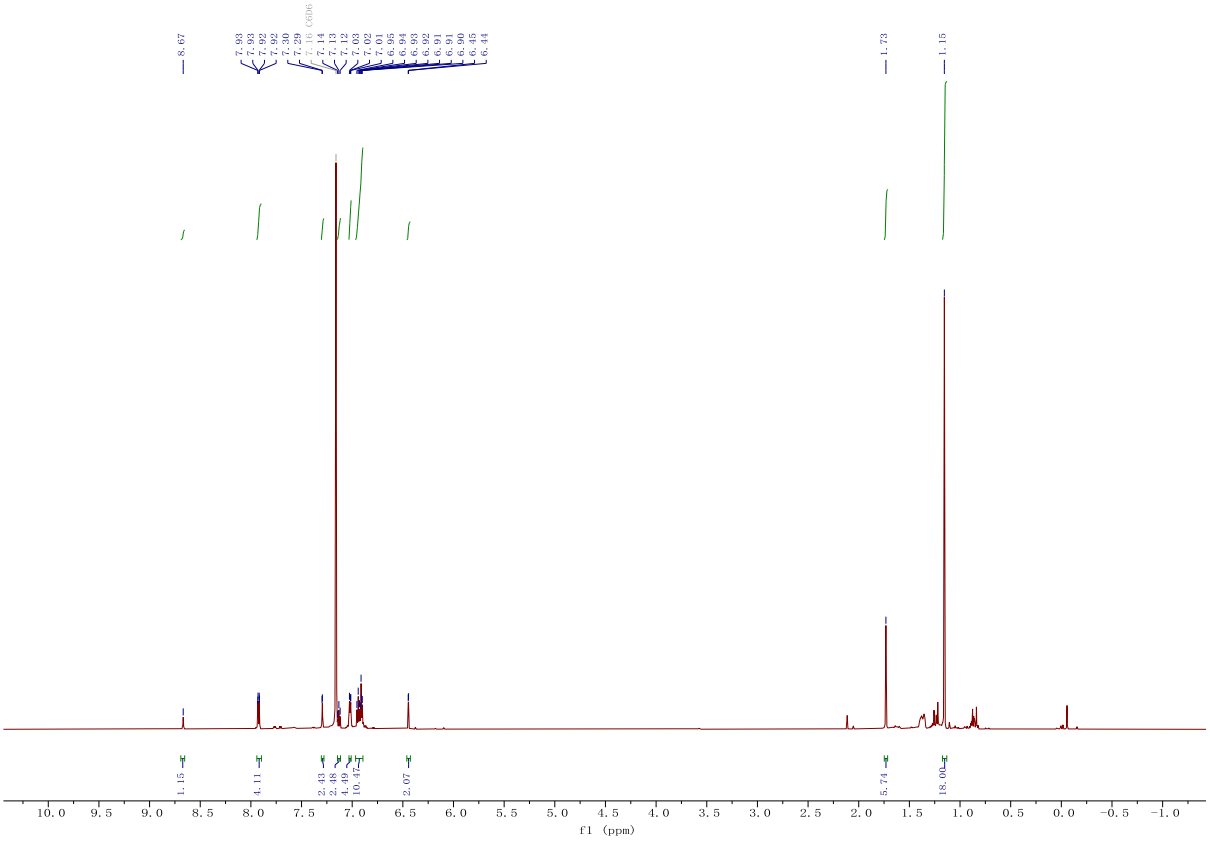


Figure S25. ^{1}H NMR spectrum of **2** in air.

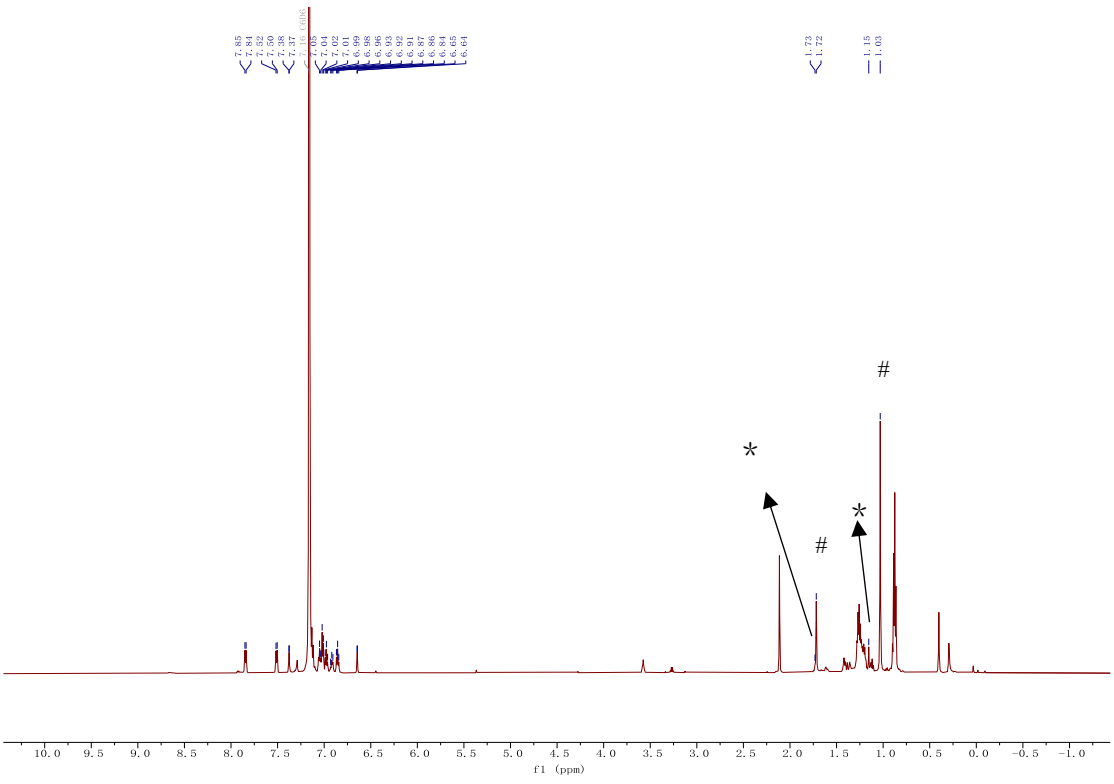


Figure S26. ^{1}H NMR spectrum of **3** in air (#indicate compound **3,** * indicate compound **1**).

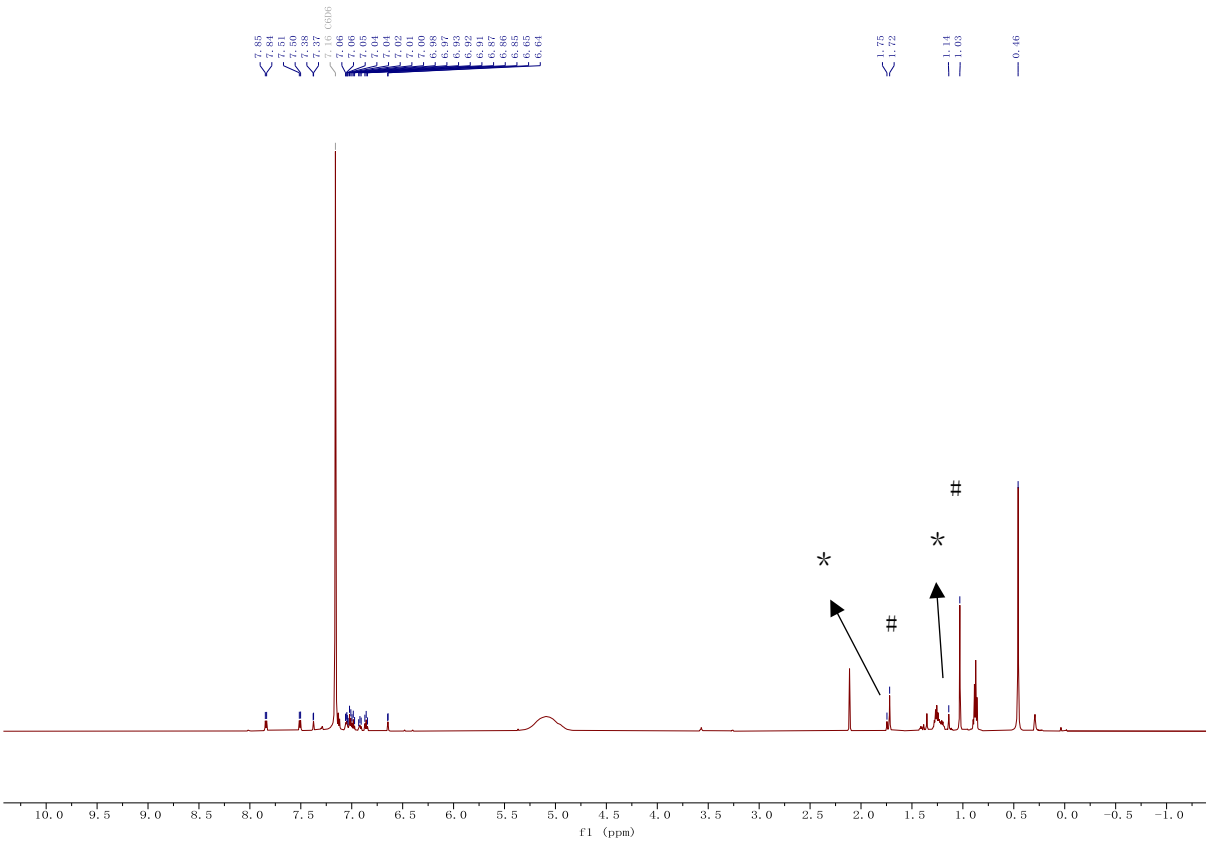


Figure S27. ^{1}H NMR spectrum of **3** with water (#indicate compound **3,** * indicate compound **1**).

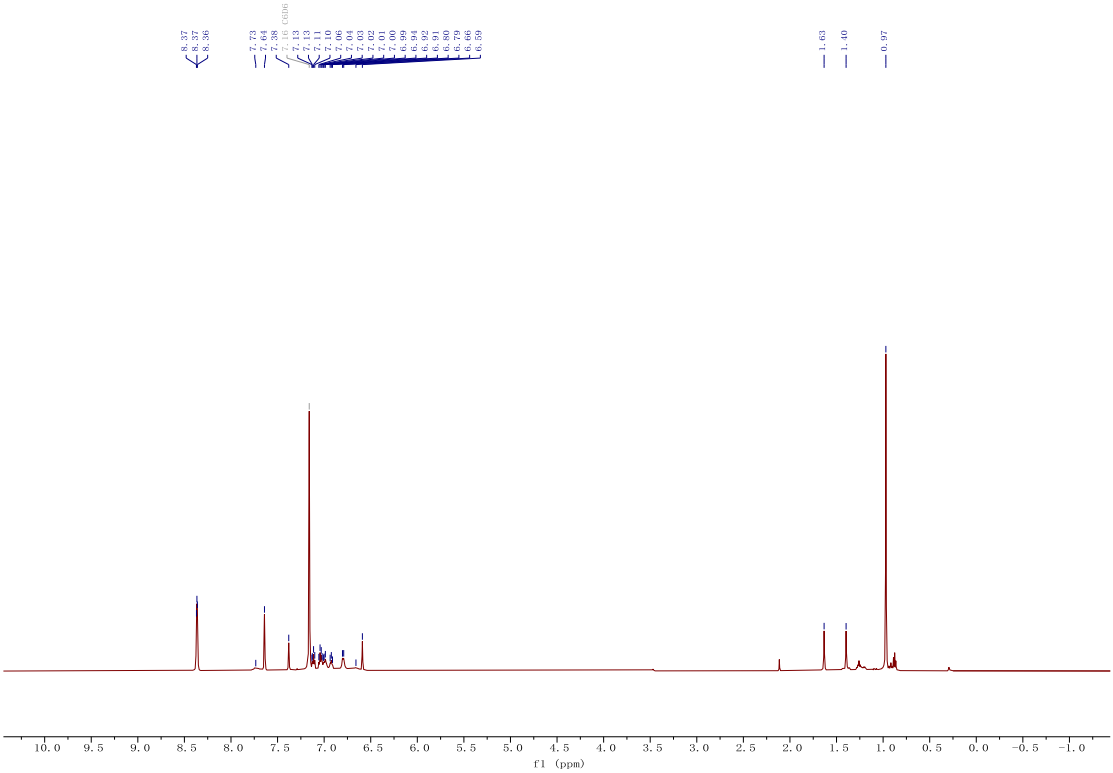


Figure S28. ^{1}H NMR spectrum of **4**[B(Ar$_F$)$_4$] in air.

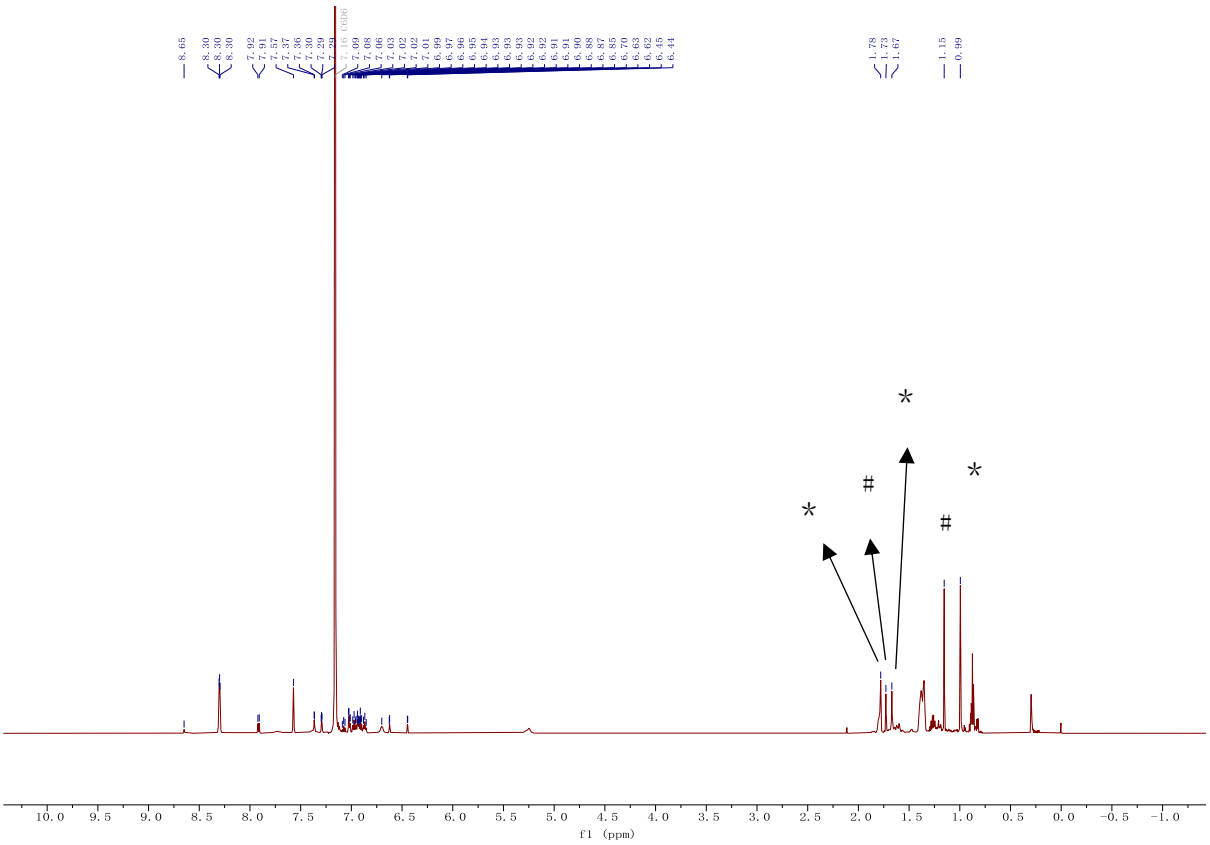


Figure S29. ^{1}H NMR spectrum of **5**[B(Ar$_F$)$_4$] in air (#indicate compound **1**, * indicate new compound).

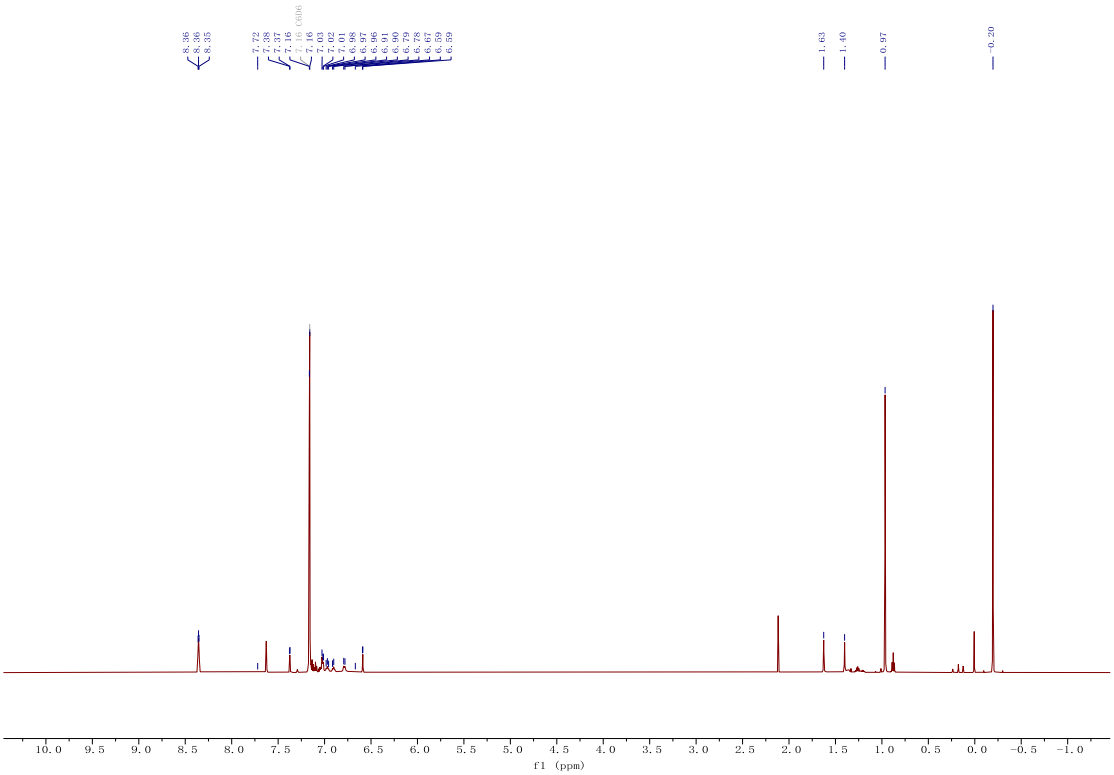


Figure S30. ^{1}H NMR spectrum of the reaction **4**[B(Ar$_F$)$_4$] and TMSCN in C_6D_6

NMR spectra of Lewis acidity measurement

Figure S31. ^{31}P NMR spectrum of Et_3PO in CD_2Cl_2.

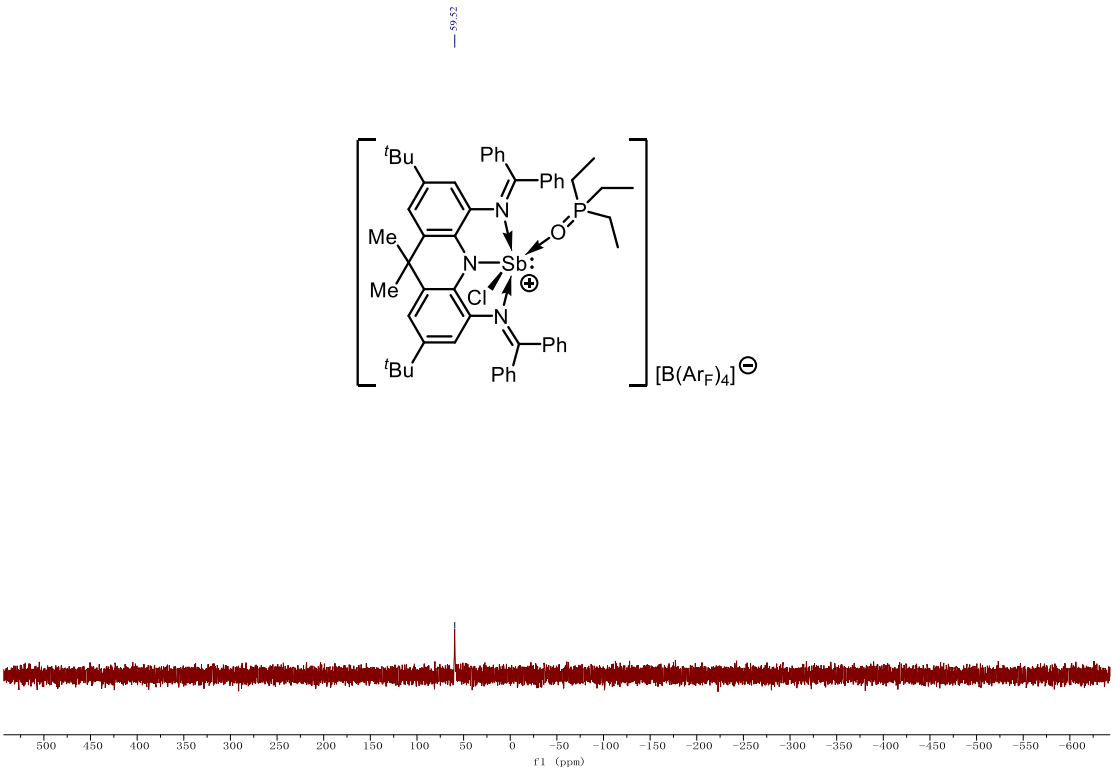


Figure S32. ^{31}P NMR spectrum of the reaction **4**[$B(Ar_F)_4$] and Et_3PO in CD_2Cl_2.

NMR spectra for ketone cayanosilyation products：

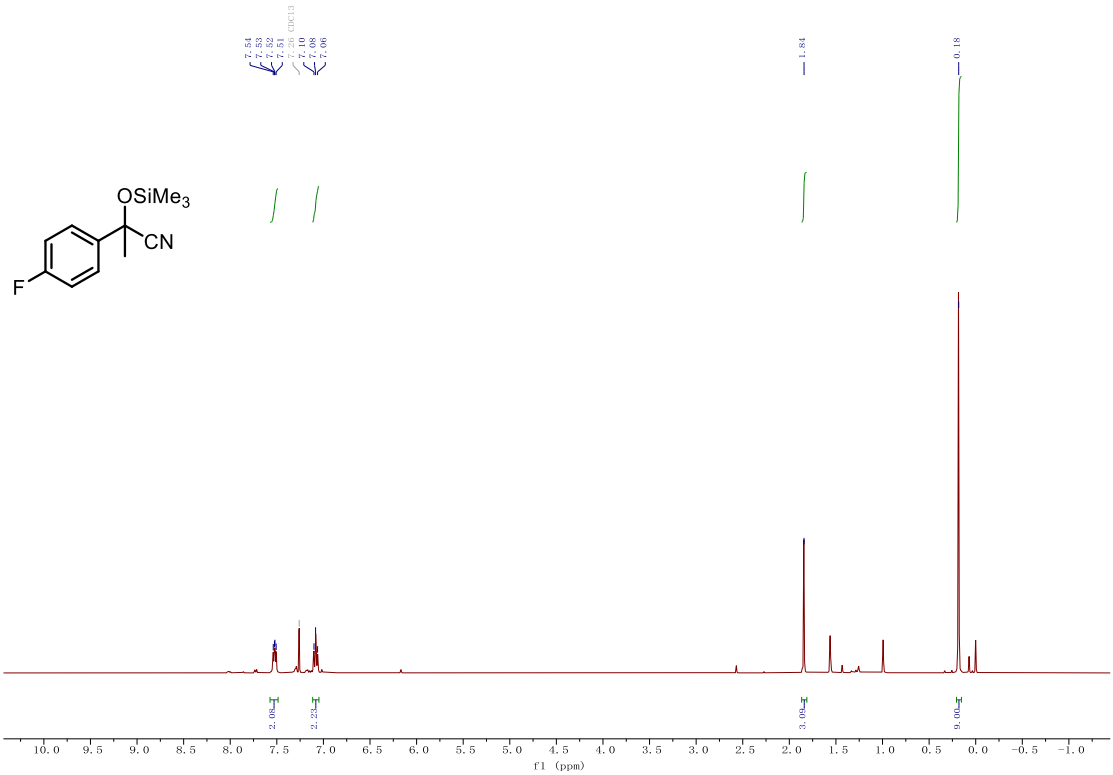


Figure S33. ^{1}H NMR spectrum of **7a**.

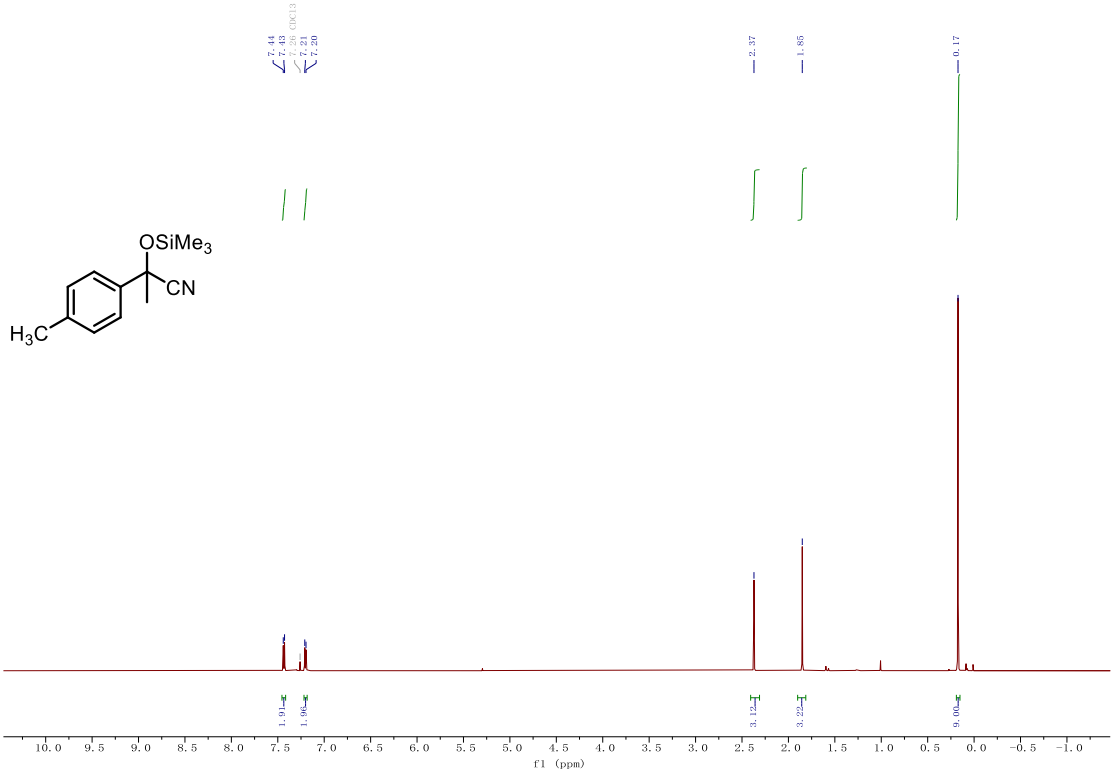


Figure S34. ^{1}H NMR spectrum of **7b**.

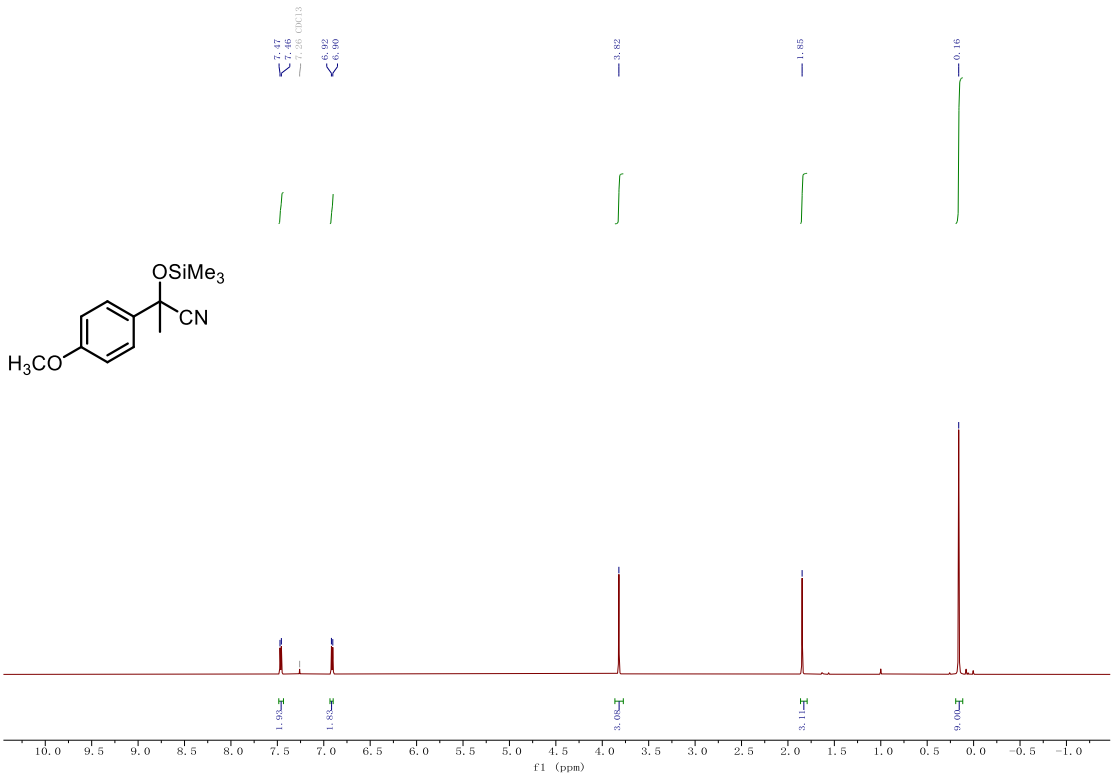


Figure S35. ^{1}H NMR spectrum of **7c**.

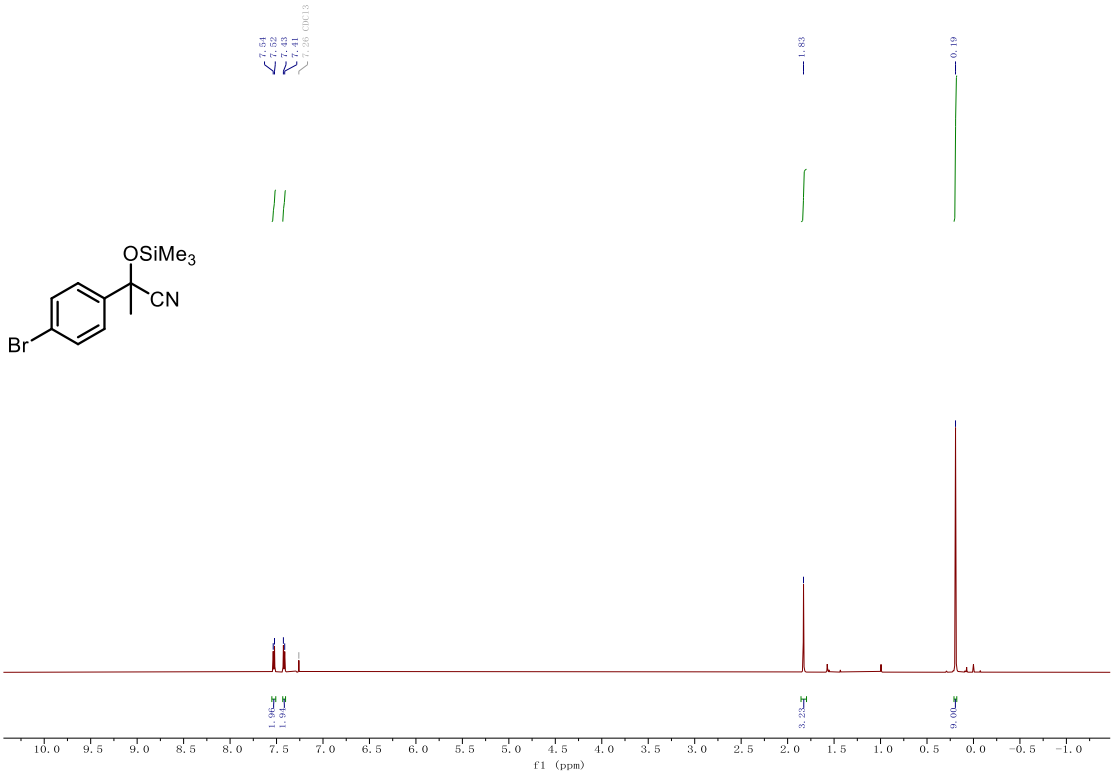


Figure S36. ^{1}H NMR spectrum of **7d**.

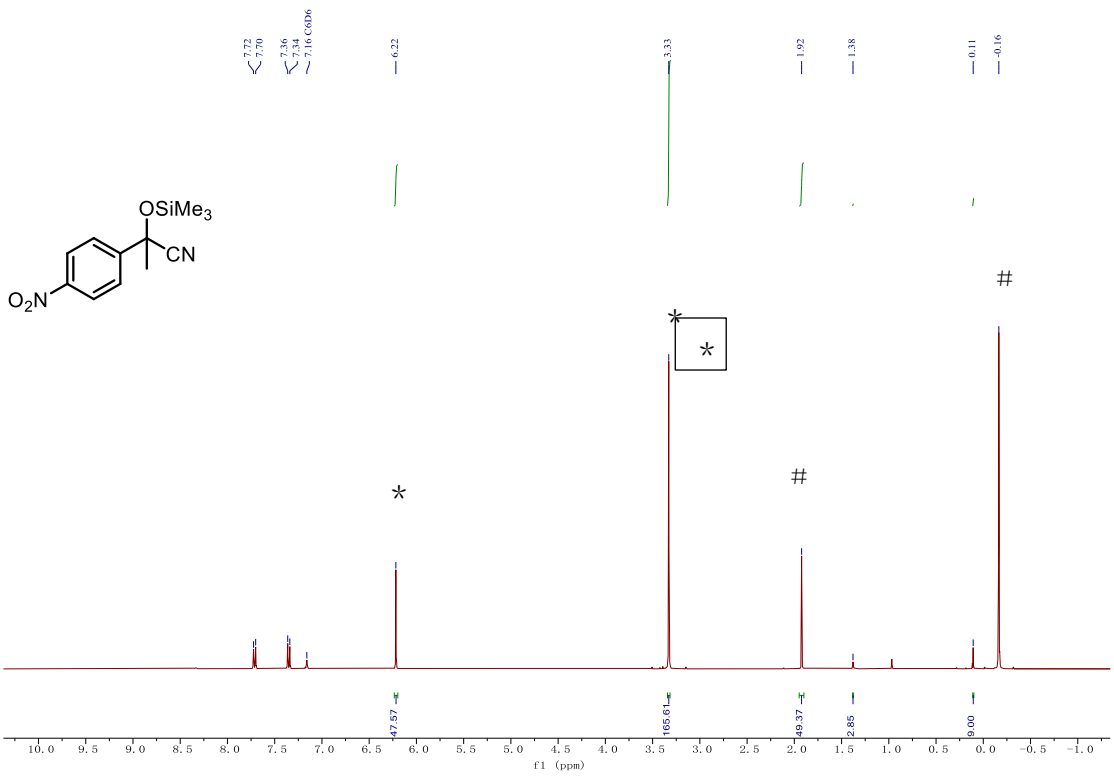


Figure S37. ^{1}H NMR spectrum of **7e** (* indicates internal standard-1,3,5-trimethoxybenzene, # indicates unreacted reactant).

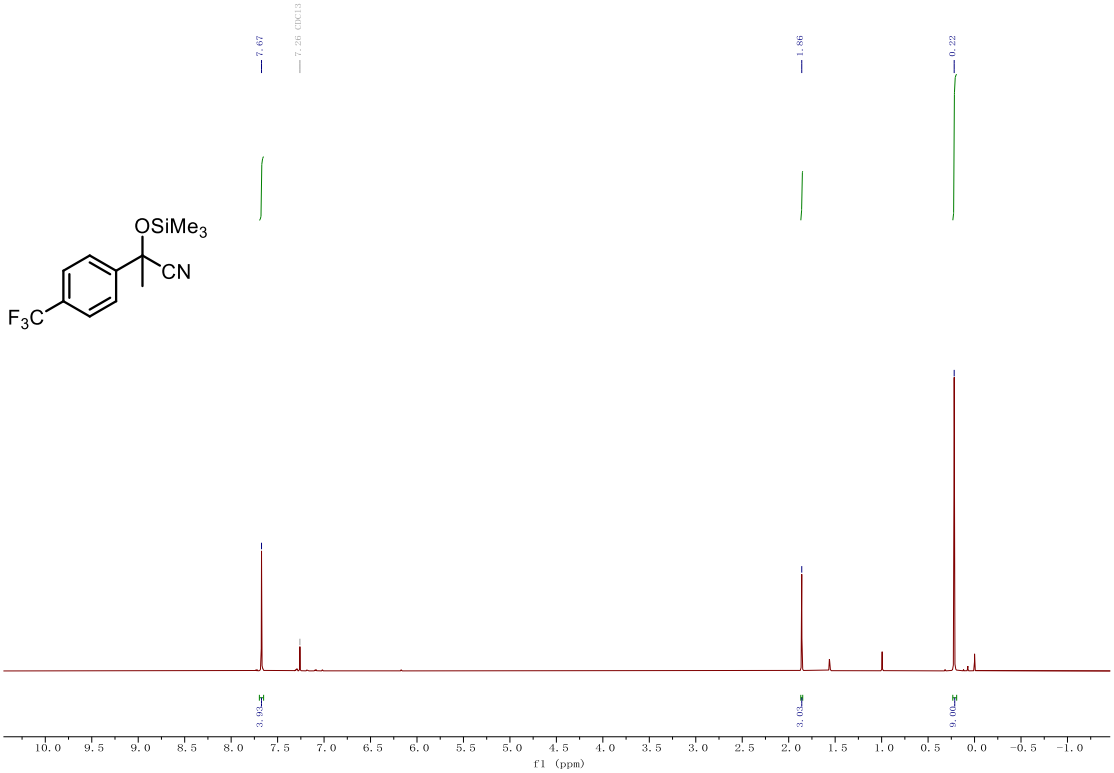


Figure S38. ^{1}H NMR spectrum of **7f**.

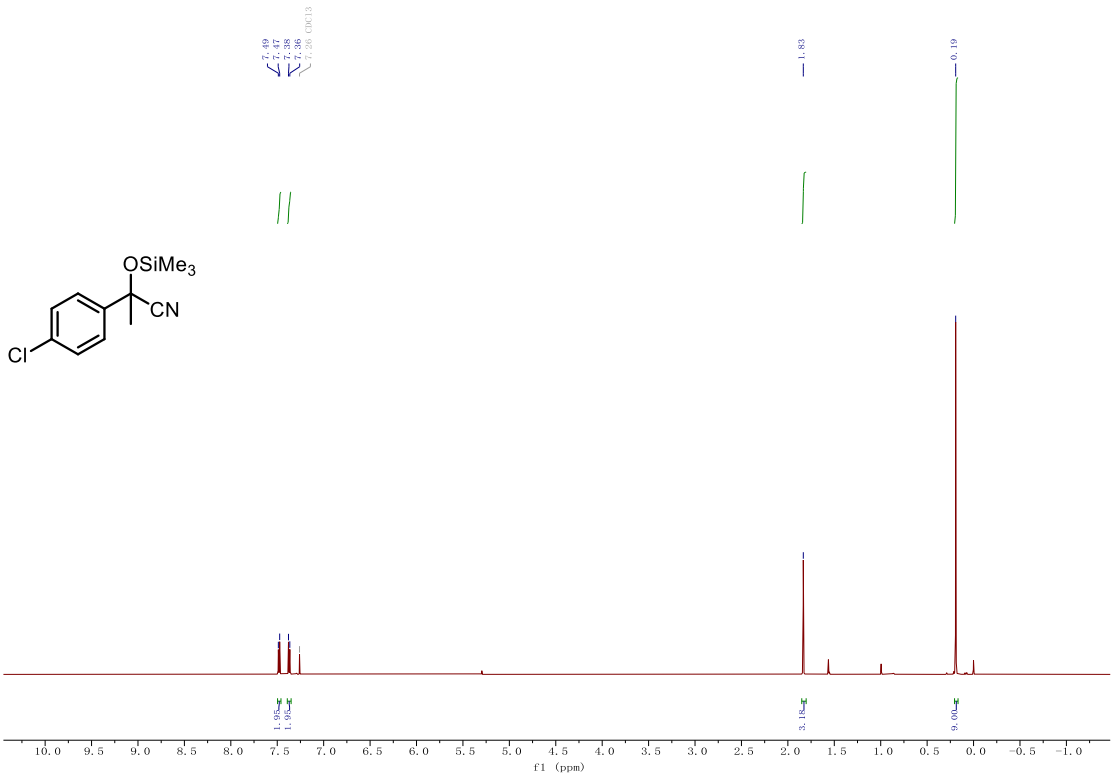


Figure S39. ^{1}H NMR spectrum of **7g**.

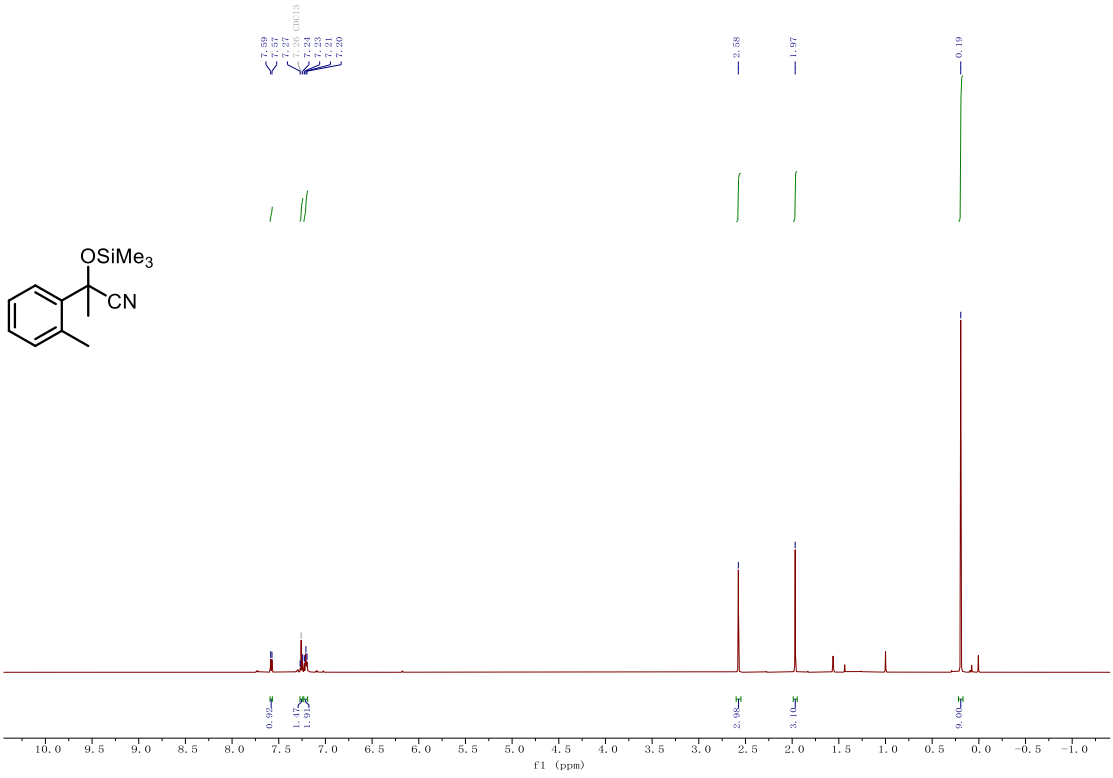


Figure S40. ^{1}H NMR spectrum of **7h**.

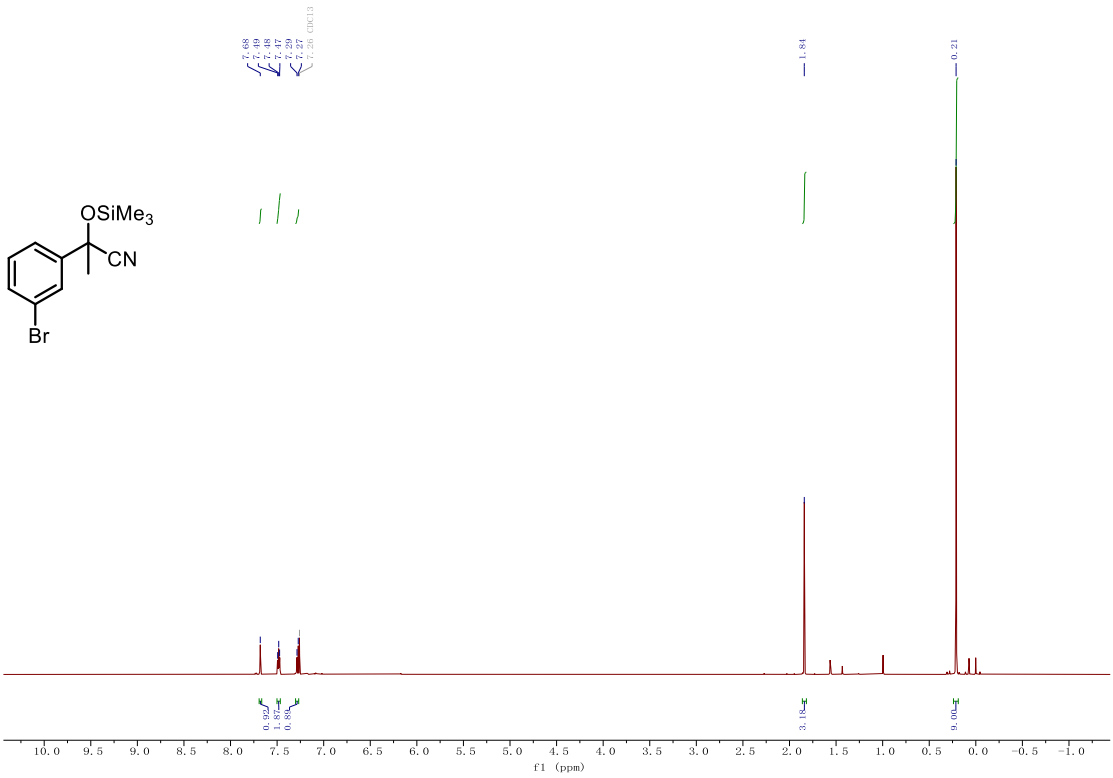


Figure S41. ^{1}H NMR spectrum of **7i**.

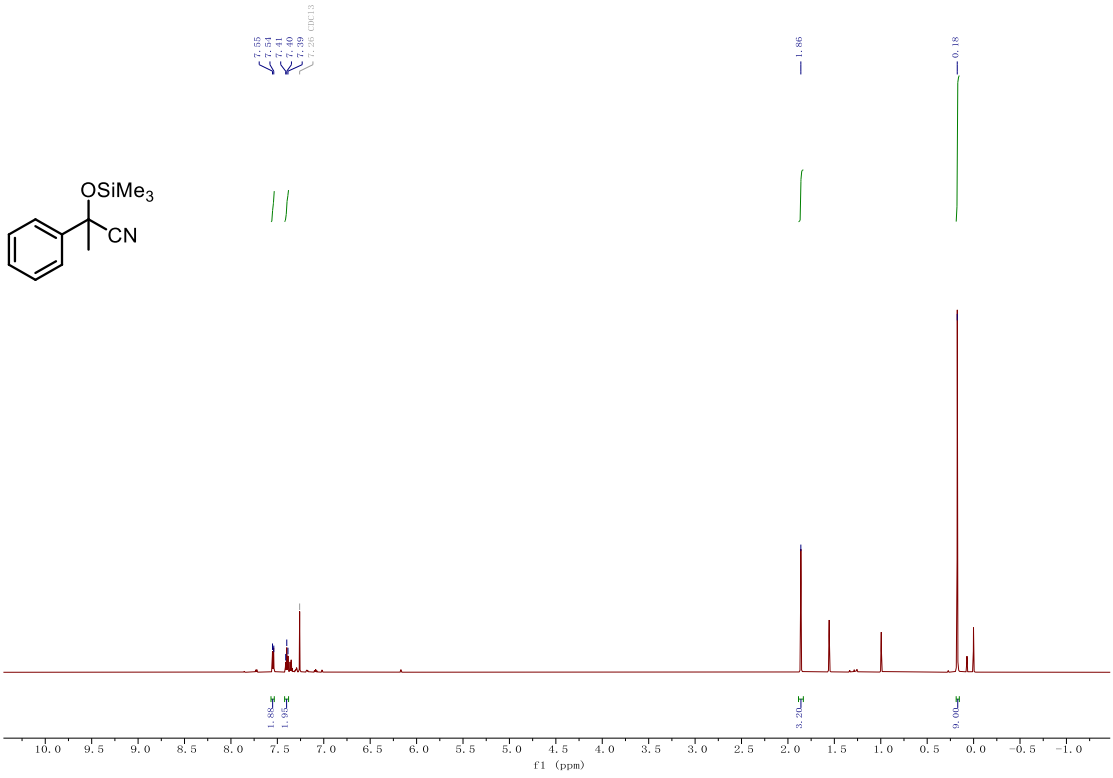


Figure S42. ^{1}H NMR spectrum of **7j**.

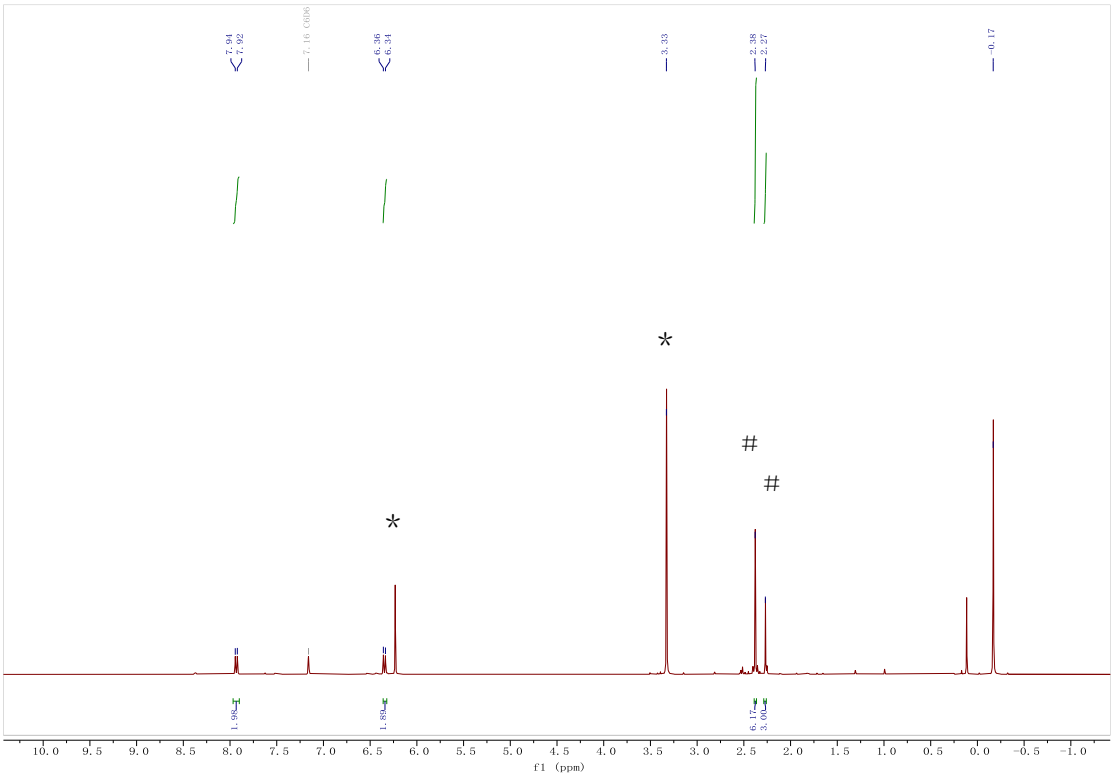


Figure S43. ^{1}H NMR spectrum of the reaction 4-$(CH_3)_2N$-$PhCOCH_3$ and TMSCN in C_6D_6 (* indicates internal standard-1,3,5-trimethoxybenzene, # indicates unreacted reactant).

VII. Reference

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