Supporting Information

Spiro-[indene-1,2'-indolin] Scaffolds *via* Rh (III) Catalyzed C-H Activation/[3+2]Spiroannulation

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Table of Contents

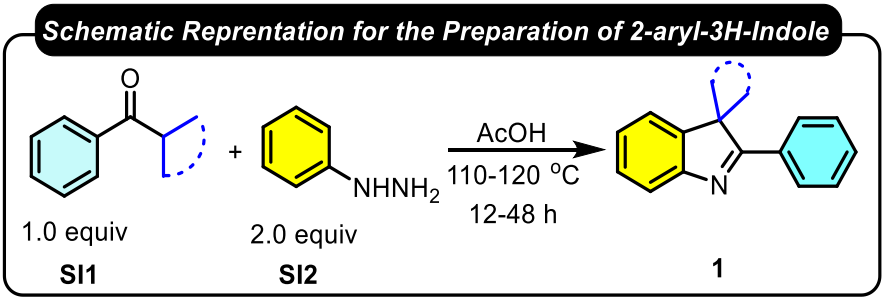
General Information:

All reactions were carried out in oven-dried Schlenk tubes under an argon atmosphere unless otherwise mentioned. TLC analysis was performed on silica gel TLC plates. Column chromatography was done using 230−400 mesh silica gel by applying pressure through an air pump. ^{1}H and ^{13}C, ^{19}F NMR spectra were recorded on a 400 MHz spectrometer and are reported as chemical shifts (δ) in parts per million (ppm), and multiplicities are abbreviated as s = singlet, d = doublet, t = triplet, q = quartet, m = multiplet, comp = complex, app = apparent. Internal standards or residual solvent signals were used as references. HRMS (m/z) was recorded using ESI (Q-Tof, positive ion) mode, EI, and LTQ-Orbitrap. Melting points were determined in a capillary melting point apparatus and were uncorrected. Optical rotation of compounds measured from Anton Paar made MCP 100 polarimeter, and HPLC purity data for compounds **4a**-**4e** were recorded using a Shim-pack GIST, C18-5μm column and Phenomenex™ Luna 5 μm C18(2) 100 Å, LC Column 250 x 4.6 mm column in MeCN/H_2O solvent system. Single-crystal X-ray diffraction data were collected on a Bruker D8 Venture with microfocus optics using Cu Kα radiation. The CIF files were submitted to CCDC (2534747-2534750, 2552131, and 2551983) and can be obtained at https://summary.ccdc.cam.ac.uk/structure-summary-form. 2-aryl-3*H*-indole[1] and Ynones/Ynoates[2] were prepared following literature methods. Single crystals of the cyclized compounds were obtained by taking 5-10 mg of the sample in 5 mL vials using a bi-solvent system applying the solvent diffusion technique. The choice of solvent systems and method for crystal growth for each compound (**3a, 3h, 3l**, **3m**, **5m**, and **5n**) are provided in the characterization data section.

General Procedure for the Preparation of Starting Materials:

(a) Preparation of Directing Group:

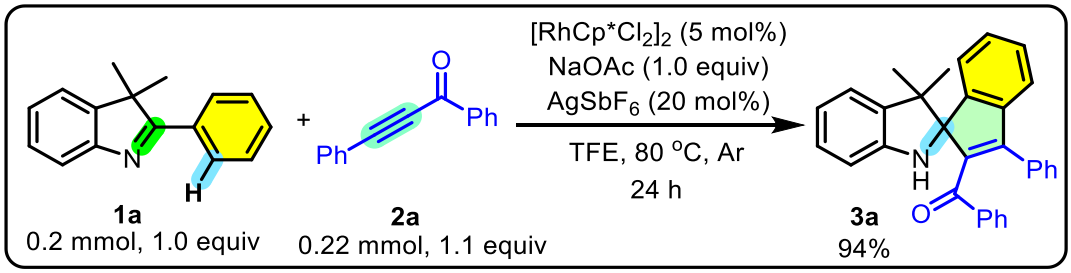
The corresponding heterocyclic 2-phenyl-3*H*-indole (**1**) was synthesized from various substituted carbonyl compounds **SI1 with** hydrazine hydrochloride **SI2** by following a known literature procedure.[1]



To a solution of hydrazine hydrochloride (2.0 equiv) in acetic acid was added corresponding carbnyl compounds (1.0 equiv) under an argon atmosphere. And then the reaction mixture was stirred at 100 °C in an oil bath for 12 h to 48 h (monitored by TLC). The solvent was removed under vacuum, and the residue was then quenched by saturated $NaHCO_3$ solution and extracted with dichloromethane three times. The organic layer was dried over Na_2SO_4 and evaporated under vacuum. Finally, the residue was purified by column chromatography (PE/EA 2-5%) to afford the respective products **1**.

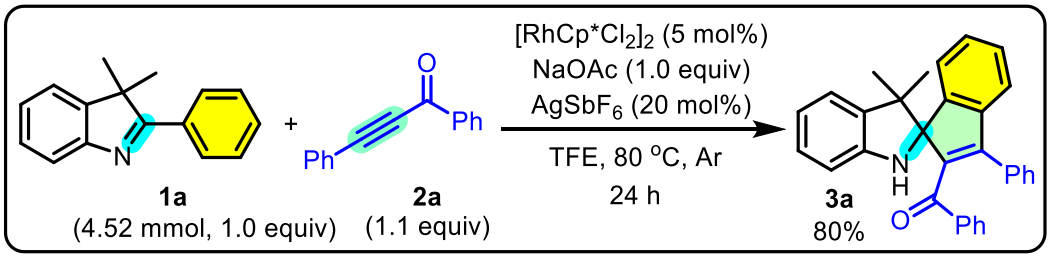
General Procedure for Spiro-Annulation Reaction:

(0.2 mmol scale):



In an oven-dried 10 mL Schlenk tube, 3,3-dimethyl-2-phenyl-3H-indole (**1a**) (44.2 mg, 0.2 mmol, 1.0 equiv), 1,3-diphenylprop-2-yn-1-one (**2a**) (45.4 mg, 0.22 mmol, 1.1 equiv), $[Cp^*RhCl_2]_2$ (6.2 mg, 0.01 mmol, 0.05 equiv), and NaOAc (16.4 mg, 0.2 mmol, 1.0 equiv) and, $AgSbF_6$ (13.7 mg, 0.04 mmol) were taken; then 2 mL (0.1 M) of TFE was added under an Argon atmosphere. Next, the tube was closed with a teflon-lined cap, and gas-degassing using Ar was done for three times before it was kept on stirring in a preheated oil bath at 80 °C. After 24 hours, the reaction was stopped and cooled to room temperature. The reaction mixture was filtered through a short pad of celite and concentrated under a vacuum. The crude reaction mixture was directly purified by column chromatography on silica gel using pet ether/ethyl acetate (19:1) as eluent to obtain (3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (**3a**) (80.4 mg, 94% yellow solid).

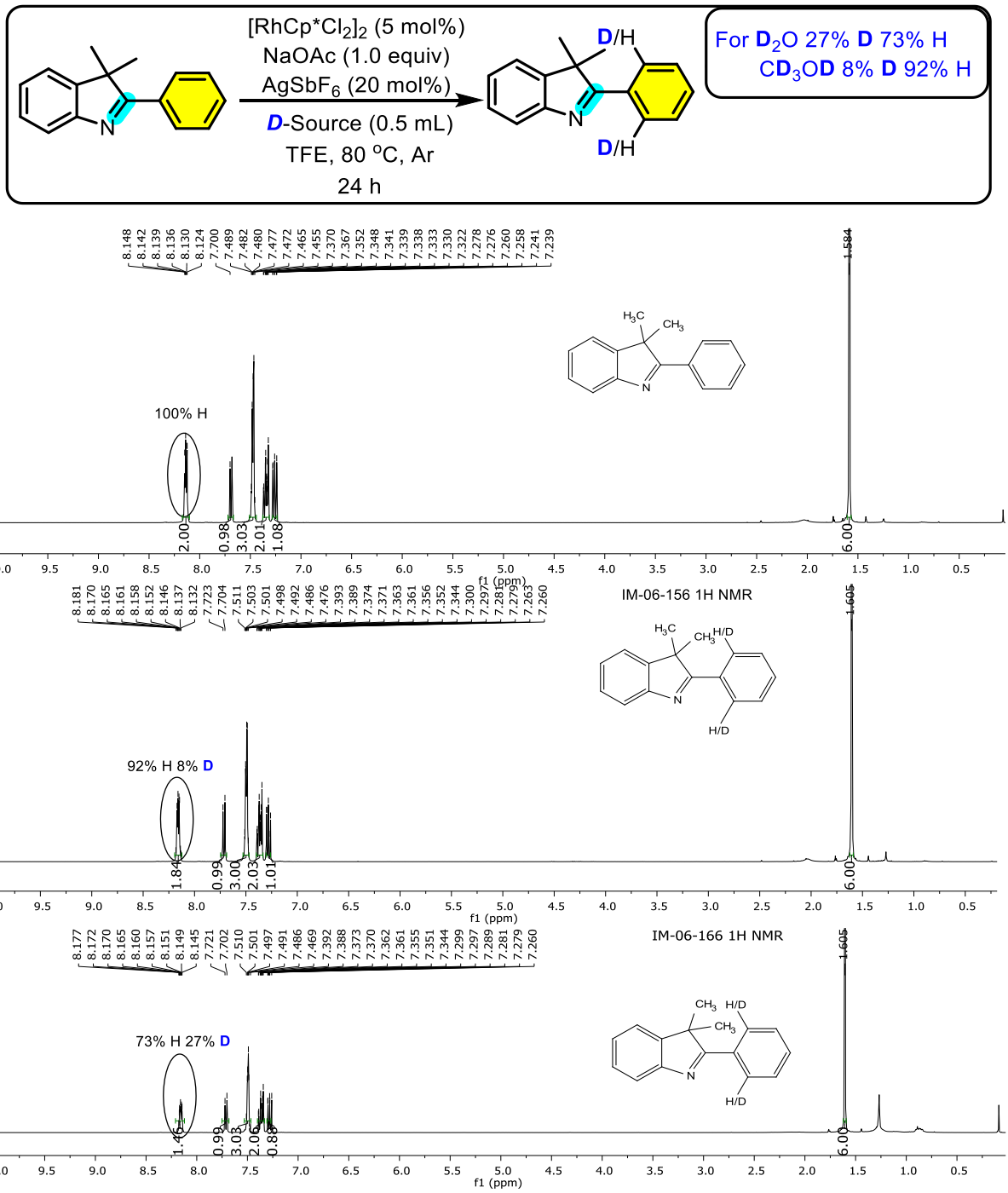
(Gram scale):



In an oven-dried 25 mL Schlenk tube, 3,3-dimethyl-2-phenyl-3*H*-indole (**1a**) (1 g, 4.52 mmol, 1.0 equiv), 1,3-diphenylprop-2-yn-1-one (**2a**) (1.02 g, 4.97 mmol, 1.1 equiv), $[Cp^*RhCl_2]_2$ (140 mg, 0.23 mmol, 0.05 equiv), and NaOAc (371 mg, 4.52 mmol, 1.0 equiv) and, $AgSbF_6$ (311 mg, 0.9 mmol) were taken; then 20 mL of TFE was added under an Argon atmosphere. Next, the tube was closed with a teflon-lined cap, and gas-degassing using Ar was done three times before it was kept on stirring in a preheated oil bath at 80 °C. After 24 hours, the reaction was stopped and cooled to room temperature. The reaction mixture was filtered through a short pad of celite and concentrated under a vacuum. The crude reaction mixture was directly purified by column chromatography on silica gel using pet ether/ethyl acetate (19:1) as eluent to obtain (3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (**3a**) (1.54 g, 80% yellow solid).

3. Mechanistic Experiments:

Procedure for H/D Exchange Experiment:

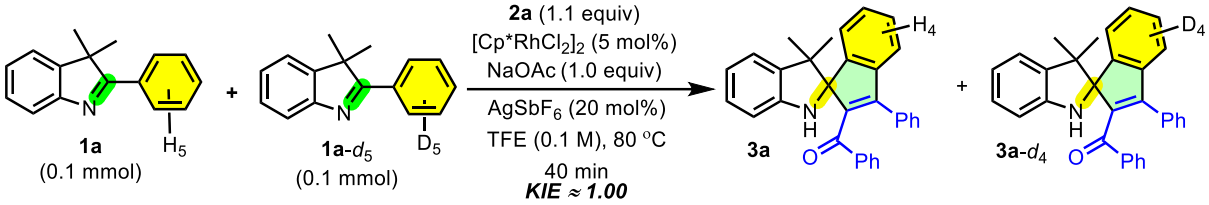


Two oven-dried 10 mL Schlenk tubes were taken, 3,3-dimethyl-2-phenyl-3*H*-indole (**1a**) (44.2 mg, 0.2 mmol, 1.0 equiv), $[Cp^*RhCl_2]_2$ (6.2 mg, 0.01 mmol, 0.05 equiv), and NaOAc (16.4 mg, 0.2 mmol, 1.0 equiv) and $AgSbF_6$ (13.7 mg, 0.04 mmol) were added to both of them; then an excess amount of D-sources (CD_3OD and D_2O) were added in two different reaction mixture, respectively. Next, 2 mL (0.1 M) of TFE was added under an Argon atmosphere into both reaction mixtures. Subsequently, the tubes were closed with a teflon-lined cap, and gas-degassing using Ar was done for three times before they were kept on stirring in a preheated oil bath at 80 °C. After 24 hours, the reactions were stopped and cooled to room temperature. The reaction mixtures were filtered through a short pad of celite and concentrated under a vacuum. The desired product (**1a**-d_n) was purified through column

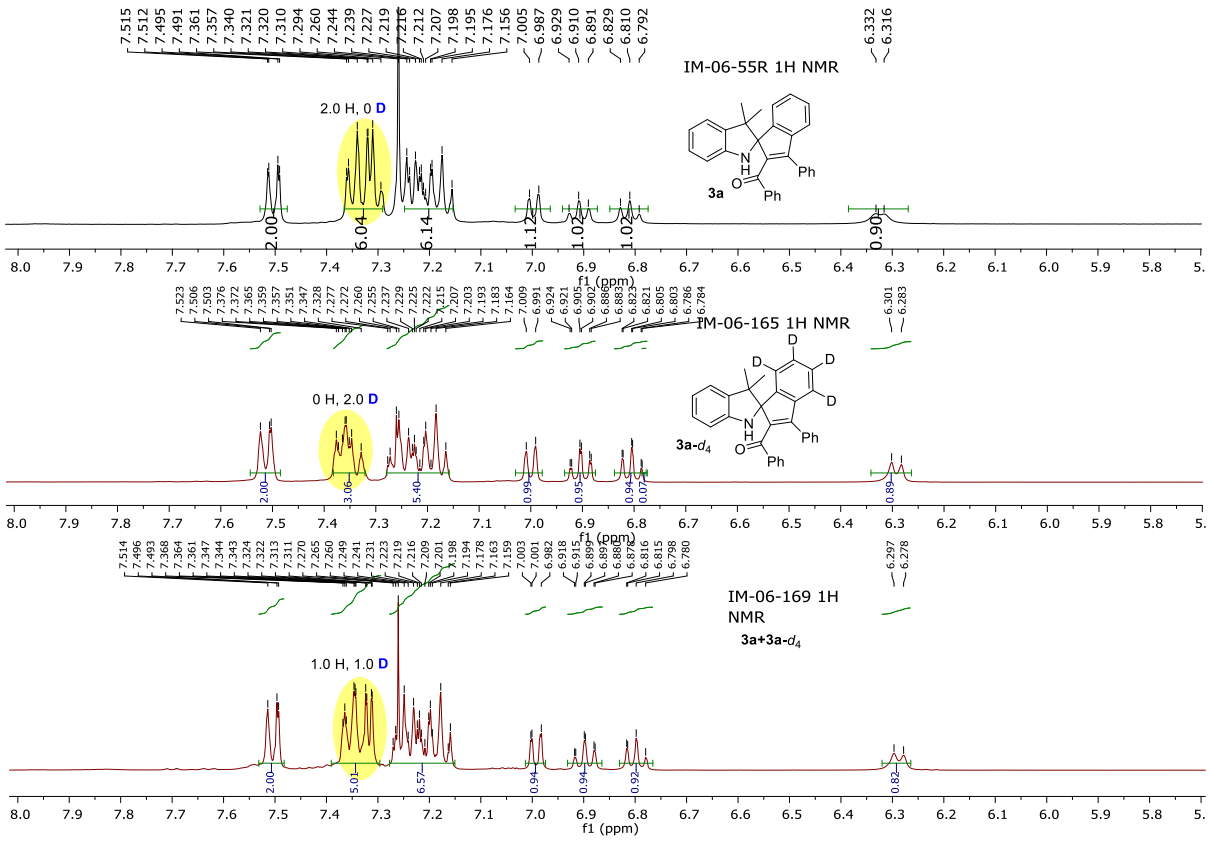
chromatography on silica gel using pet ether/ethyl acetate (49:1) as eluent. The deuterium incorporation was determined by ^{1}H NMR spectroscopy for both cases separately as 8% and 27%. The observed difference in deuterium incorporation (27% with D_2O vs 8% with CD_3OD) likely arises from the combined effects of the different acidities (pKa) of the deuterium sources and the distinct solvation properties of the reaction medium.

Competitive and Parallel Experiment

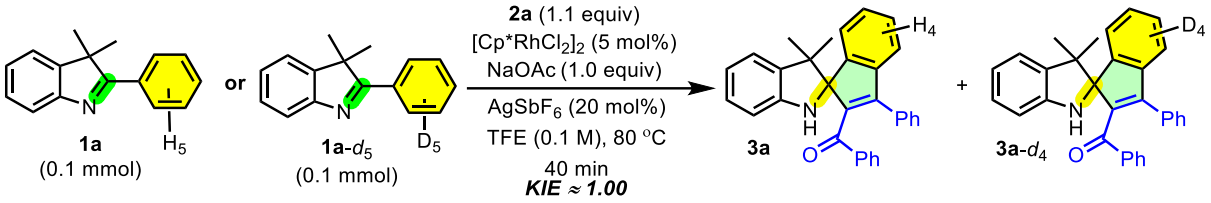
(i) Competitive Experiment between 3,3-dimethyl-2-phenyl-3*H*-indole (1a) and 3,3-dimethyl-2-(phenyl-d_5)-3*H*-indole (1a-d_5):



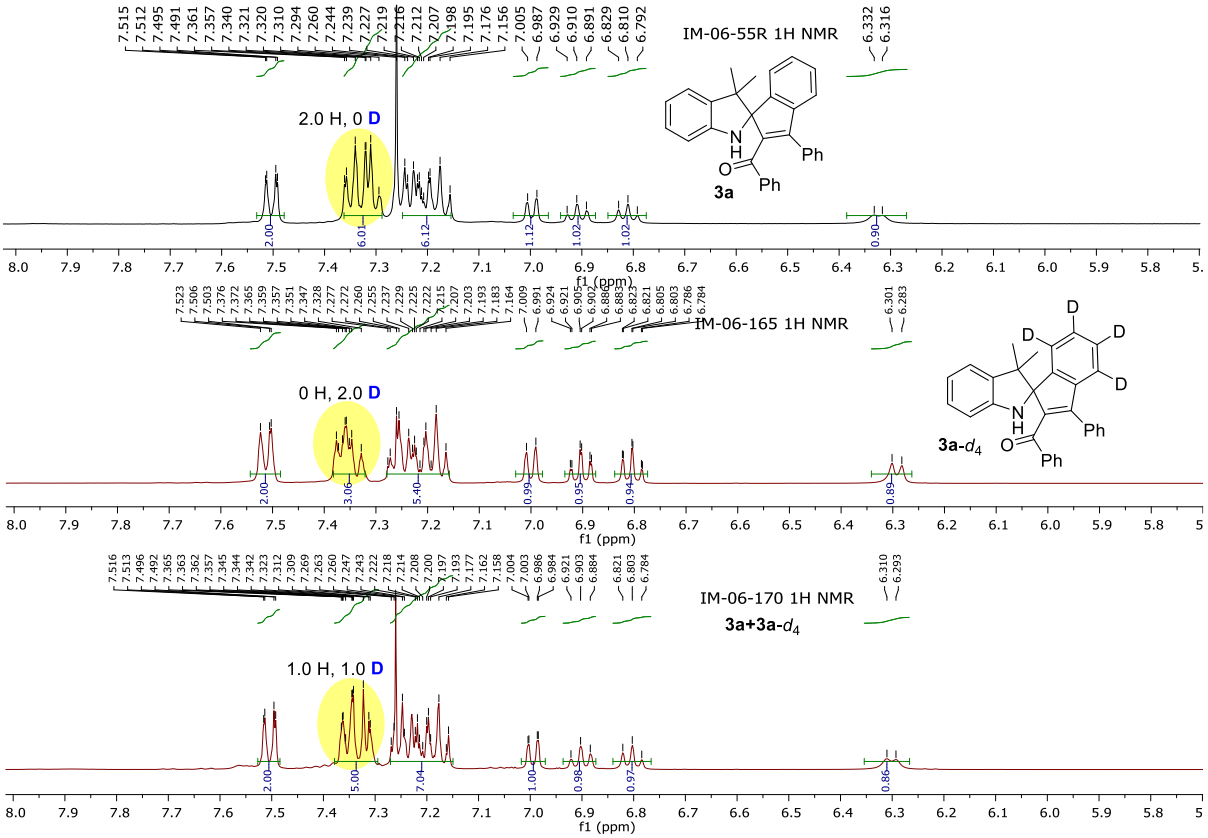
In an oven-dried 10 mL Schlenk tube, 3,3-dimethyl-2-phenyl-3*H*-indole (**1a**) (22.1 mg, 0.1 mmol, 1.0 equiv), 3,3-dimethyl-2-(phenyl-d_5)-3*H*-indole (**1a**-d_5) (22.6 mg, 0.1 mmol, 1.0 equiv), 1,3-diphenylprop-2-yn-1-one (**2a**) (45.4 mg, 0.22 mmol, 1.1 equiv), $[Cp^*RhCl_2]_2$ (6.2 mg, 0.01 mmol, 0.05 equiv), and NaOAc (16.4 mg, 0.2 mmol, 1.0 equiv) and $AgSbF_6$ (13.7 mg, 0.04 mmol) were taken, and 2.0 mL of TFE (0.1 M) was added under Ar-atmosphere. Then it was the tube was closed with a teflon-lined cap and gas-degassing was done before it was stirred in a preheated oil bath at 80 °C. After 40 min, the reaction was stopped and quenched in an ice bath. The reaction mixtures were filtered through a short pad of celite and concentrated under vacuo. The crude reaction mixtures were directly purified through column chromatography on silica gel using pet ether/ethyl acetate (19:1) as eluent. The ratio of **3a** and **3a**-d_4 was determined by ^{1}H NMR spectroscopy. The primary kinetic isotopic effect was **1.0/1.0** ≈ **1.00**.



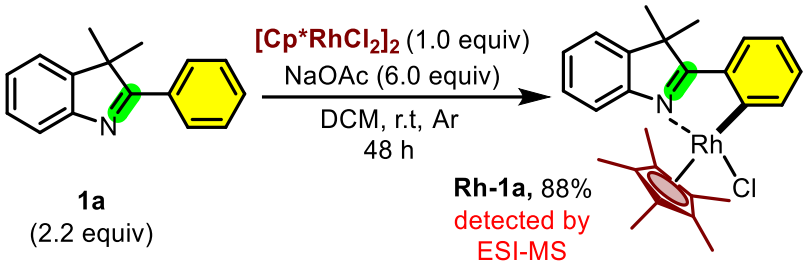
(ii) Parallel experiment between 1a and 1a-d_5:



Two separate oven dried 10 mL Schlenk tubes were charged with 3,3-dimethyl-2-phenyl-3*H*-indole (**1a**) (22.1 mg, 0.1 mmol, 1.0 equiv), 3,3-dimethyl-2-(phenyl-d_5)-3*H*-indole (**1a**-d_5) (22.6 mg, 0.1 mmol, 1.0 equiv), 1,3-diphenylprop-2-yn-1-one (**2a**) (27.8 mg, 0.11 mmol, 1.1 equiv), $[Cp^*RhCl_2]_2$ (3.1 mg, 0.05 equiv), and NaOAc (8.2 mg, 0.1 mmol, 1.0 equiv) and $AgSbF_6$ (13.7 mg, 0.02 mmol) were taken, and 1.0 mL of TFE (0.1 M) was added to both of them under Ar-atmosphere. Then it was the tube was closed with a teflon-lined cap and gas-degassing was done before it was stirred in a preheated oil bath at 80 °C. After 40 min the reaction was stopped and quenched in an ice bath. The reaction mixture was filtered through a short pad of celite and concentrated under vacuo. The crude reaction mixture was directly purified through column chromatography on silica gel using pet ether/ethyl acetate (19:1) as eluent. The ratio of **3a** and **3a**-d_4 was determined by ^{1}H NMR spectroscopy. The primary kinetic isotopic effect was found as $\boldsymbol{k_H/k_D}$ = **1.0/1.0** ≈ **1.00**.

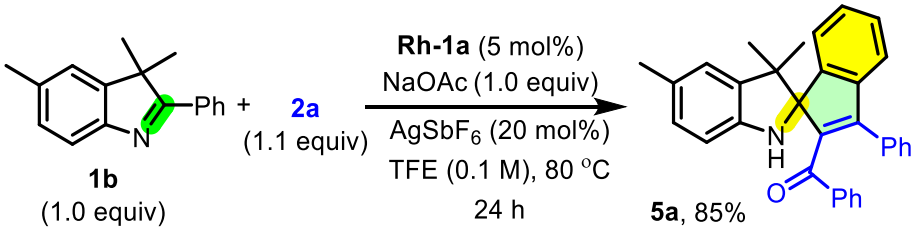


General Procedure for the Preparation of the Rhodacycle Complex:



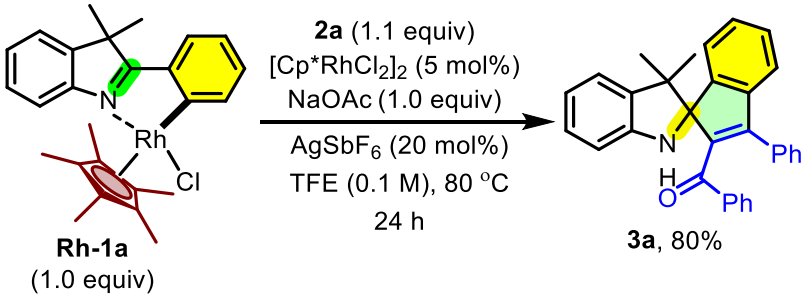
Following an analogous literature procedure,[3] a mixture of $[Cp^*RhCl_2]_2$ (50.0 mg, 0.08 mmol, 1.0 equiv), 3,3-dimethyl-2-phenyl-3*H*-indole (**1a**) (38.9 mg, 0.176 mmol, 2.2 equiv), and NaOAc (39.4 mg, 0.48 mmol, 6.0 equiv) in 10.0 mL of dry DCM was allowed to stir under argon atmosphere at r.t (approximately 36 °C) for 48 h. After completion of the reaction, the reaction mixture was passed through a short pad of celite and concentrated under vacuo, followed by washing with hexane and finally vacuum-dried to give the Rhodacycle compound **Rh-1a** as a reddish-brown solid (40 mg, 88% yield). ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.99-7.96 (m, 1H), 7.72 – 7.63 (comp, 2H), 7.43 – 7.34 (comp, 2H), 7.31-7.26 (m, 1H), 7.25-7.22 (m, 1H), 7.12-7.07 (m, 1H), 1.66 (s, 15H), 1.56 (s, 6H). 149.4, 146.3, 139.8, 139.5, 130.5, 127.8, 127.2, 126.0, 122.4, 121.2, 119.0, 96.3, 25.0, 9.8, 8.3; HRMS (ESI, m/z) calcd for $C_{26}H_{29}NRh$ $[M - Cl]^+$ 458.1355, found 458.1362.

Procedure for the Treatment of the Rhodacycle Complex Rh-1a as Catalyst:



In an oven-dried 10 mL Schlenk tube, 3,3,5-trimethyl-2-phenyl-3*H*-indole (**1b**) (47 mg, 0.2 mmol, 1.0 equiv), 1,3-diphenylprop-2-yn-1-one (**2a**) (45.4 mg, 0.22 mmol, 1.1 equiv), **Rh-1a** (10 mg, 0.01 mmol, 0.05 equiv), and NaOAc (16.4 mg, 0.2 mmol, 1.0 equiv) and, $AgSbF_6$ (13.7 mg, 0.04 mmol) were taken; then 20 mL of TFE was added under an Argon atmosphere. Next, the tube was closed with a teflon-lined cap, and gas-degassing using Ar was done for three times before it was kept on stirring in a preheated oil bath at 80 °C. After 24 hours, the reaction was stopped and cooled to room temperature. The reaction mixture was filtered through a short pad of celite and concentrated under a vacuum. The crude reaction mixture was directly purified by column chromatography on silica gel using pet ether/ethyl acetate (19:1) as eluent to obtain phenyl(3',3',5'-trimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)methanone (**5a**) (75 mg, 85% yellow solid).

Procedure for the Treatment of the Rhodacycle Complex Rh-1a as a Substrate:



In an oven-dried 10 mL Schlenk tube, **Rh-1a** (100 mg, 0.2 mmol, 1.0 equiv), 1,3-diphenylprop-2-yn-1-one (**2a**) (45.4 mg, 0.22 mmol, 1.1 equiv), $[Cp^*RhCl_2]_2$ (6.2 mg, 0.01 mmol, 0.05 equiv), and NaOAc (16.4 mg, 0.2 mmol, 1.0 equiv) and $AgSbF_6$ (13.7 mg, 0.04 mmol) were taken, and 2.0 mL of TFE (0.1

M) was added under Ar-atmosphere. Next, the tube was closed with a teflon-lined cap, and gas-degassing using Ar was done for three times before it was kept on stirring in a preheated oil bath at 80 °C. After 24 hours, the reaction was stopped and cooled to room temperature. The reaction mixture was filtered through a short pad of celite and concentrated under a vacuum. The crude reaction mixture was directly purified by column chromatography on silica gel using pet ether/ethyl acetate (19:1) as eluent to obtain (3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (**3a**) (68.4 mg, 80% yellow solid).

References:

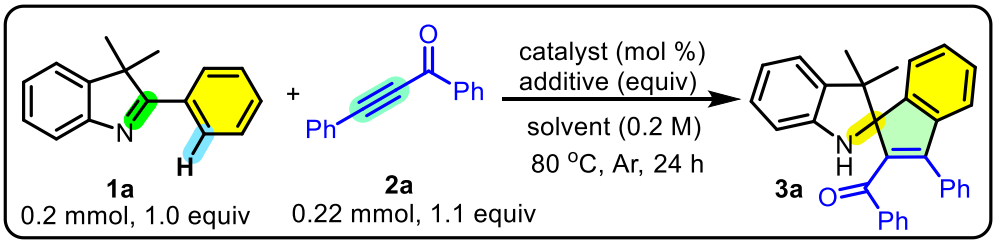
1. J. Gao, K. Luo, X. Wei, H. Wang, H. Liu and Y. Zhou, *Org. Lett*., 2023, **25**, 3341-3346.

2. K. Naskar, S. Karmakar, I. Mondal, W. Sarkar, S. Roy, A. Roy and I. Deb, *Chem. Commun*., 2023, **59**, 7751-7754.

3. K. Naskar, I. Mondal, S. Das, N. S. Bliss, M. Saha, S. Dhar and I. Deb, *Org. Lett*., 2025, **27**, 9619-9624.

4. W. Sarkar, K. Naskar, S. Roy, I. Mondal, S. Karmakar, A. Mishra and I. Deb, *J. Org. Chem*., 2022, **87**, 9988-10002.

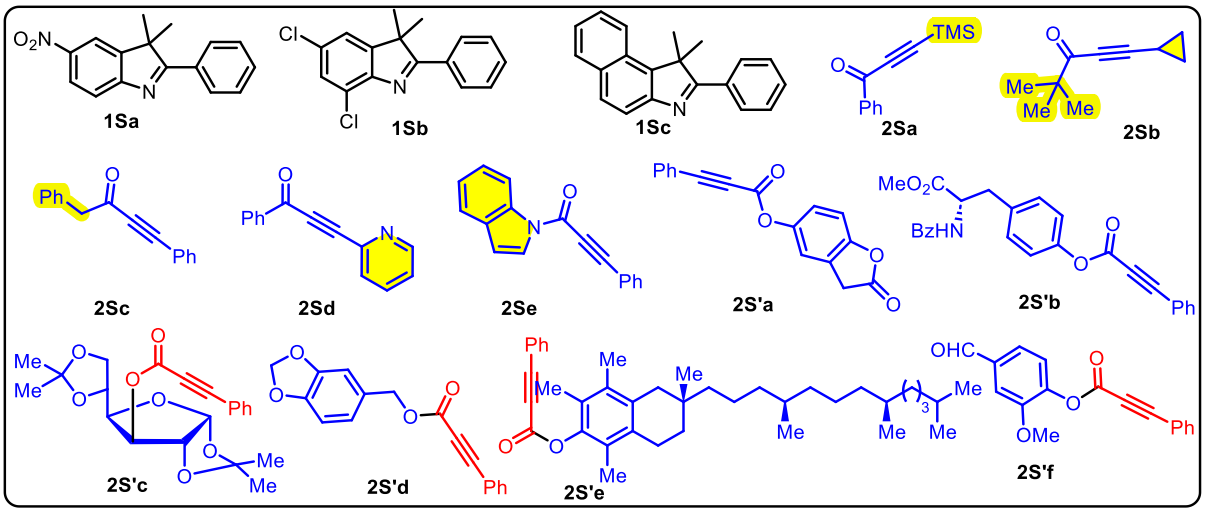
Table S1. Additional Optimization of Reaction Conditions[a]



Entry	$[Cp^*RhCl_2]_2$ (mol %)	Additives (equiv)	Temp (°C)	Time (h)	Yield (%) of 3a[b]
1	3	NaOAc (1.0 equiv)	80	24	68
2	5	NaOAc (1.0 equiv)	60	24	70
3	5	KOAc (1.0 equiv)	80	24	80
4	5	NaOPiv (1.0 equiv)	80	24	70
5	5	$Zn(OAc)_2$ (1.0 equiv)	80	24	88
6	5	PivOH (1.0 equiv)	80	24	60
7	5	$Cu(OAc)_2$ (1.0 equiv)	80	24	75
8	5	NaOAc (1.0 equiv) + $AgNTf_2$ (20)	80	24	85

[a]Reaction conditions: **1a** (1.0 equiv, 0.2 mmol), **2a** (1.1 equiv), $[Cp^*RhCl_2]_2$ (mol %), NaOAc (equiv), $AgSbF_6$ (mol%), 2.0 mL TFE (0.1 M), under Ar. [b]Isolated yield. TFE = 2,2,2-trifluoroethanol.

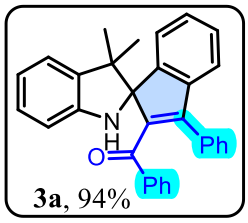
Table S2. Unsuccessful Substrates



Nitro, di-chloro-substituted indolin and a naphthalene fused DG did not work under the current reaction conditions; in all cases the starting materials were unreacted. For the TMS and cyclopropane containing substrates, several new spots formed, however could not be characterized from complex mixtures. Benzyl, pyridine and indole containing ynones were left unreacted and failed to furnish the desired products. For the other natural product derived ynoates (2s'a-2s'f), no desired spiro-cyclized products were afforded; in some cases, complex mixtures found which could not be characterized. For all the substrates, we investigated the reactivity using various additives and further optimized the reaction conditions (changing time and temperature parameters); however, none of these attempts were successful.

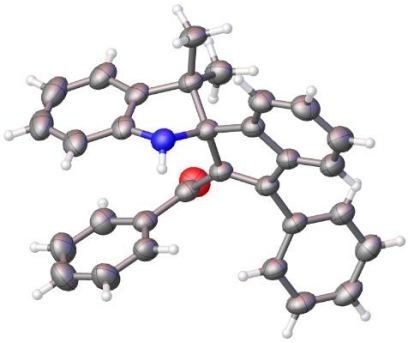
The characterization data for all the products is given below

(3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (3a):



Yield 94% (80.3 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 154–56 °C; Crystallization was done from hot ethanol; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.52–7.49 (comp, 2H), 7.38–7.28 (comp, 6H), 7.25–7.13 (comp, 6H), 7.01–6.97 (m, 1H), 6.92–6.89 (m, 1H), 6.82–6.78 (m, 1H), 6.33–6.31 (m, 1H), 1.34 (s, 3H), 1.26 (s, 3H); **^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$)** δ 196.2, 148.2, 146.6, 146.5, 144.2, 141.7, 139.6, 137.9, 133.2, 132.4, 128.8 (X2), 128.4, 128.3 (X2), 127.9, 127.4, 127.0, 124.4, 121.9, 121.8, 119.8, 111.1, 86.7, 49.4, 26.1, 24.8; **HRMS** (ESI, m/z) calcd for $C_{31}H_{26}NO$ $[M + H]^+$ 428.2014, found 428.2003.

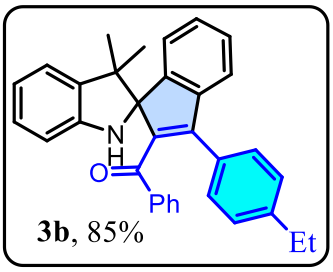
Figure S1. X-ray crystal structure of **3a** (ellipsoid contour at 50% probability level)



Crystal data and structure refinement for 3a (CCDC 2534747)

Identification code	IBD_IM_06_168_0m_a	ρ_{calc}g/cm^3	1.256
Empirical formula	$C_{31}H_{25}NO$	μ/mm^{-1}	0.580
Formula weight	427.52	F(000)	904.0
Temperature/K	297.00	Crystal size/mm^3	0.32 × 0.23 × 0.19
Crystal system	monoclinic	Radiation	Cu Kα (λ = 1.54178)
Space group	$P2_1/n$	2Θ range for data collection/°	8.872 to 136.522
a/Å	10.3386(16)	Index ranges	-12 ≤ h ≤ 12, -22 ≤ k ≤ 22, -14 ≤ l ≤ 14
b/Å	18.480(3)	Reflections collected	43667
c/Å	11.8838(17)	Independent reflections	4073 [R_{int} = 0.0526, R_{sigma} = 0.0263]
α/°	90	Data/restraints/parameters	4073/0/301
β/°	95.167(5)	Goodness-of-fit on F^2	1.086
γ/°	90	Final R indexes [I>=2σ (I)]	R_1 = 0.0554, wR_2 = 0.1465
Volume/Å^3	2261.3(6)	Final R indexes [all data]	R_1 = 0.0563, wR_2 = 0.1475
Z	4	Largest diff. peak/hole / e Å^{-3}	0.23/-0.22

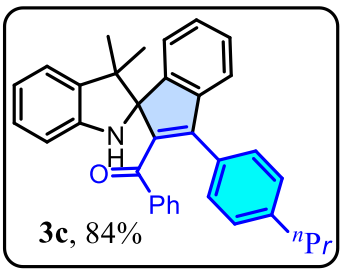
(3-(4-ethylphenyl)-3',3'-dimethylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (3b):



3b, 85%

Yield 85% (77.3 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 163-165 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.52–7.47 (comp, 2H), 7.37–7.30 (comp, 4H), 7.30–7.25 (comp, 2H), 7.24–7.21 (m, 1H), 7.19–7.15 (comp, 2H), 7.07 (app d, *J* = 8.0 Hz, 2H), 6.99 (app d, *J* = 7.2 Hz, 1H), 6.89 (app td, *J* = 7.6, 1.0 Hz, 1H), 6.82–6.77 (m, 1H), 6.26 (app d, *J* = 7.6, 1H), 4.49 (br s, 1 N-H), 2.56 (q, *J* = 7.6 Hz, 2H), 1.35 (s, 3H), 1.26 (s, 3H), 1.15 (t, *J* = 7.6 Hz, 3H); **^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$)** δ 196.4, 148.3, 146.5, 144.4, 143.9, 141.9, 139.7, 138.1, 132.3, 130.4, 128.8, 128.3 (X2), 127.9, 127.8, 127.4, 126.9, 124.4, 122.0, 121.8, 119.8, 111.1, 86.6, 49.4, 28.6, 26.2, 24.7, 15.3; **HRMS** (ESI, m/z) calcd for $C_{33}H_{30}NO$ $[M + H]^+$ 456.2317, found 456.2314.

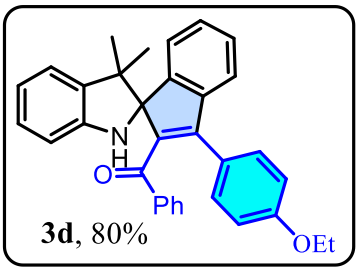
(3',3'-dimethyl-3-(4-propylphenyl)spiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (3c):



3c, 84%

Yield 84% (79 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 160-162 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.54–7.49 (comp, 2H), 7.37–7.28 (comp, 4H), 7.28–7.24 (comp, 2H), 7.23–7.14 (comp, 3H), 7.06-7.03 (comp, 2H), 7.02–6.98 (m, 1H), 6.92 (app td, *J* = 7.6, 1.3 Hz, 1H), 6.80 (app td, *J* = 7.4, 0.9 Hz, 1H), 6.32 (app d, *J* = 7.6 Hz, 1H), 4.55 (br s, 1NH), 2.50 (t, *J* = 7.6 Hz, 2H), 1.65–1.48 (m, 2H), 1.35 (s, 3H), 1.26 (s, 3H), 0.85 (t, *J* = 7.3 Hz, 3H) **^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$)** δ 196.4, 148.3, 147.0, 146.9, 143.5, 142.9, 141.7, 139.8, 138.0, 132.3, 130.5, 128.9, 128.7, 128.4, 128.3, 127.8, 127.4, 127.0, 124.2, 122.0, 121.8, 119.8, 111.2, 86.5, 49.5, 37.7, 26.0, 25.1, 24.2, 13.6; **HRMS** (ESI, m/z) calcd for $C_{34}H_{31}NO$ $[M]^+$ 469.2406, found 469.2411.

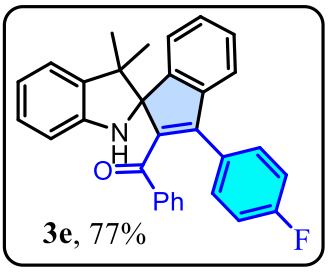
(3-(4-ethoxyphenyl)-3',3'-dimethylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (3d):



3d, 80%

Yield 80% (75.5 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 176-178 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.56–7.49 (comp, 2H), 7.39–7.25 (comp, 6H), 7.24–7.15 (comp, 3H), 7.03–6.97 (m, 1H), 6.90 (app td, *J* = 7.6, 1.2 Hz, 1H), 6.83–6.73 (comp, 3H), 6.29 (app d, *J* = 7.6, 1H), 4.47 (br s, 1N-H), 3.95 (q, *J* = 7.0 Hz, 2H), 1.41–1.31 (m, 6H), 1.26 (s, 3H); **^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$)** δ 196.5, 158.9, 148.3, 146.8, 146.2, 143.3, 141.9, 139.7, 138.0, 132.4, 130.1, 128.9, 128.3, 127.9, 127.4, 126.9, 125.4, 124.3, 121.9, 121.8, 119.8, 114.3, 111.1, 86.5, 63.3, 49.4, 26.0, 24.9, 14.7; **HRMS** (ESI, m/z) calcd for $C_{33}H_{30}NO_2$ $[M + H]^+$ 472.2277, found 472.2274.

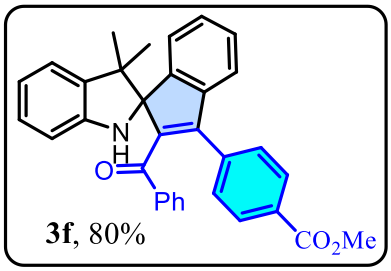
(3-(4-fluorophenyl)-3',3'-dimethylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (3e):



3e, 77%

Yield 77% (68.5 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 165-167 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.51–7.45 (comp, 2H), 7.41–7.31 (comp, 5H), 7.30–7.25 (m, 1H and $CDCl_3$ peak), 7.24–7.17 (comp, 3H), 7.02–6.92 (m, 3H), 6.88 (app td, *J* = 7.6, 1.3 Hz, 1H), 6.82–6.77 (m, 1H), 6.24 (app d, *J* = 7.6 Hz, 1H), 4.38 (br s, 1 N-H), 1.35 (s, 3H), 1.27 (s, 3H); **^{13}C{^{1}H} NMR (101 MHz, $CDCl_3$)** δ 196.1, 163.8, 161.3, 148.1, 146.3, 145.54 (d, *J* = 45.4 Hz), 141.6, 139.5, 137.9, 132.6, 130.65 (d, *J* = 8.2 Hz)., 129.27–129.11 (m), 128.7, 128.5, 128.0, 127.4, 127.2, 124.5, 121.8, 121.7, 119.9, 115.5, 115.3, 111.1, 86.8, 49.4, 26.2, 24.50; **^{19}F NMR** (376 MHz, $CDCl_3$) -112.5; **HRMS** (ESI, m/z) calcd for $C_{31}H_{25}NOF$ $[M + H]^+$ 446.1920, found 446.1906.

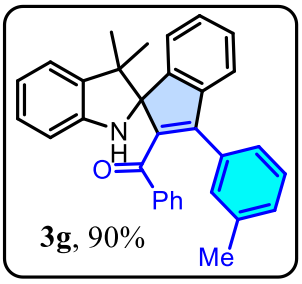
methyl 4-(2-benzoyl-3',3'-dimethylspiro[indene-1,2'-indolin]-3-yl)benzoate (3f):



3f, 80%

Yield 80% (71.2 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 181-183 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.97–7.90 (comp, 2H), 7.49-7.42 (comp, 4H), 7.40–7.31 (comp, 3H), 7.29–7.26 (comp, 2H), 7.21–7.17 (comp, 2H), 7.02–6.96 (m, 1H), 6.87 (app td, *J* = 7.6, 1.3 Hz, 1H), 6.81–6.75 (m, 1H), 6.21 (d, *J* = 7.6 Hz, 1H), 4.46 (br s, 1N-H), 3.87 (s, 3H), 1.36 (s, 3H), 1.26 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 195.8, 166.5, 148.0, 146.1, 145.9, 145.1, 141.3, 139.4, 137.9, 137.8, 132.7, 129.8, 129.6, 128.8, 128.7, 128.6, 128.1, 127.5, 127.2, 124.7, 121.8, 121.7, 119.9, 111.0, 87.0, 52.1, 49.5, 26.4, 24.2; **HRMS** (ESI, m/z) calcd for $C_{33}H_{28}NO_3$ $[M + H]^+$ 486.2069, found 486.2060.

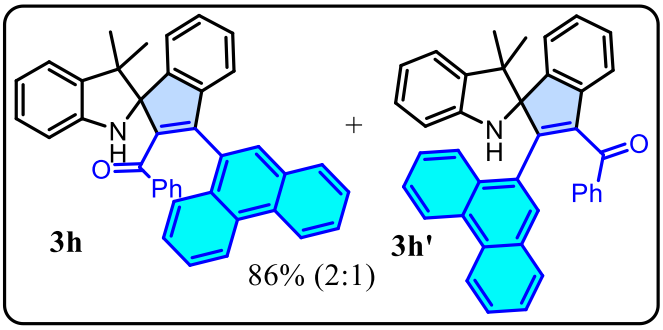
(3',3'-dimethyl-3-(m-tolyl)spiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (3g):



3g, 90%

Yield 90% (79.4 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 74-76 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.54–7.47 (comp, 2H), 7.37–7.28 (comp, 4H), 7.24–7.10 (comp, 6H), 7.03–6.96 (comp, 2H), 6.90 (app td, *J* = 7.6, 1.2 Hz, 1H), 6.83–6.76 (m, 1H), 6.30 (app d, *J* = 7.7 Hz, 1H), 4.49 (br s, 1N-H), 2.24 (s, 3H), 1.35 (s, 3H), 1.26 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 196.3, 148.3, 146.9, 146.8, 143.8, 141.7, 139.7, 138.0, 137.9, 133.1, 132.8, 129.6, 129.1, 128.8, 128.3, 128.2, 127.8, 127.4, 127.0, 125.8, 124.3, 122.0, 121.8, 119.8, 111.2, 86.6, 49.5, 26.0, 25.0, 21.3; **HRMS** (ESI, m/z) calcd for $C_{32}H_{28}NO$ $[M + H]^+$ 442.2171, found 442.2187.

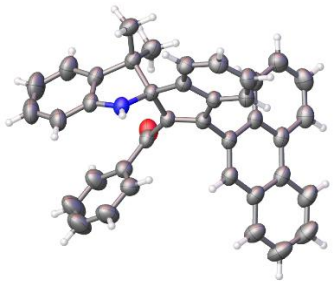
(3',3'-dimethyl-3-(phenanthren-9-yl)spiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (3h) and (3',3'-dimethyl-2-(phenanthren-9-yl)spiro[indene-1,2'-indolin]-3-yl)(phenyl)methanone (3h´):



Combined Yield 86% (91 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 173-175 °C; Crystallization was done from hot ethanol; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 8.71–8.63 (m, 1.08H), 8.58 (comp, 2H), 8.07 (d, *J* = 7.9 Hz, 1H), 7.93–7.88 (m, 0.48H), 7.88–7.83 (m, 0.90H), 7.72–7.66 (m, 1.21H), 7.67–7.54 (comp, 7H), 7.53–7.43 (comp, 3.17H), 7.36–7.31 (m, 1H), 7.29–7.19 (comp, 3.69H), 7.18–7.09 (comp, 2.57H), 7.09–6.99 (comp, 4.44H), 6.93 (t, *J* = 7.8 Hz, 1H), 6.90–

6.80 (comp, 5.34H), 6.66–6.63 (m, 1H), 6.30–6.20 (m, 0.30H), 4.99 (br s, 0.71 NH), 4.67 (br s, 0.19 NH), 1.54 (s, 1.42H), 1.43 (s, 3.14H), 1.42 (s, 3H), 1.37 (s, 1.36H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 196.2, 196.0, 148.9, 148.7, 148.3, 148.0, 146.0, 145.5, 144.9, 142.8, 142.2, 139.9, 139.8, 138.1, 137.8, 131.1, 131.0, 130.5, 130.32, 130.28, 130.21, 130.18, 129.4, 129.2, 128.9, 128.6, 128.5, 128.4, 128.3, 127.7, 127.6, 127.51, 127.46, 127.4, 127.2, 127.1, 127.0, 126.8, 126.7, 126.63, 126.57, 126.5, 126.4, 124.5, 123.6, 123.1, 123.0, 122.8, 122.6, 122.3, 121.9, 121.9, 120.1, 119.9, 111.8, 111.3, 86.9, 86.7, 49.8, 27.5, 26.6, 24.7, 24.3; **HRMS** (ESI, m/z) calcd for $C_{39}H_{30}NO$ $[M + H]^+$ 528.2329 found 528.2327.

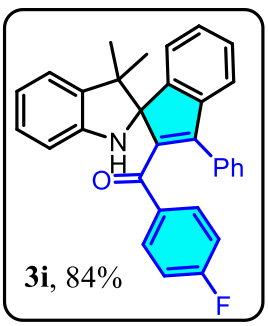
Figure S2. X-ray crystal structure of **3h** (ellipsoid contour at 50% probability level)



Crystal data and structure refinement for 3h (CCDC 2552131)

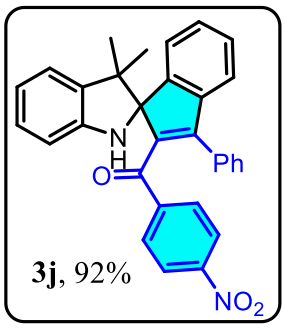
Identification code	IM_06_113N_0m_a	ρ_{calc}g/cm^3	1.195
Empirical formula	$C_{39}H_{29}NO$	μ/mm^{-1}	0.546
Formula weight	527.671	F(000)	557.7
Temperature/K	294.00	Crystal size/mm^3	0.16 × 0.14 × 0.09
Crystal system	triclinic	Radiation	Cu Kα (λ = 1.54178)
Space group	P-1	2Θ range for data collection/°	6.52 to 133.3
a/Å	10.3817(2)	Index ranges	-12 ≤ h ≤ 12, -13 ≤ k ≤ 12, -17 ≤ l ≤ 17
b/Å	11.3734(2)	Reflections collected	31540
c/Å	14.3921(3)	Independent reflections	5182 [R_{int} = 0.0492, R_{sigma} = 0.0331]
α/°	92.141(1)	Data/restraints/parameters	5182/0/373
β/°	106.885(1)	Goodness-of-fit on F^2	1.055
γ/°	113.637(1)	Final R indexes [I>=2σ (I)]	R_1 = 0.0515, wR_2 = 0.1413
Volume/Å^3	1466.56(5)	Final R indexes [all data]	R_1 = 0.0555, wR_2 = 0.1462
Z	2	Largest diff. peak/hole / e Å^{-3}	0.47/-0.37

(3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(4-fluorophenyl)methanone (3i):



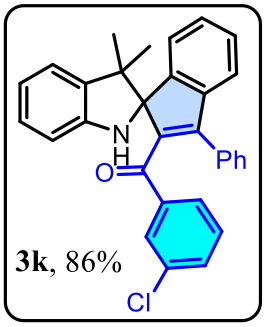
Yield 84% (75 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 176-178 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.58–7.51 (comp, 2H), 7.37–7.30 (comp, 5H), 7.30–7.20 (m, 4H), 7.04–6.98 (m, 1H), 6.94 (td, *J* = 7.6, 1.3 Hz, 1H), 6.89–6.78 (comp, 3H), 6.37 (d, *J* = 7.7 Hz, 1H), 4.47 (br s, 1NH), 1.34 (s, 3H), 1.27 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 194.5, 166.5, 163.9, 148.3, 146.77 (d, *J* = 4.5 Hz), 143.7, 141.5, 139.6, 134.30 (d, *J* = 2.6 Hz), 133.1, 131.51–131.27 (m), 128.7, 128.5, 128.4 (X2), 127.4, 127.2, 124.3, 121.91 (d, *J* = 12.2 Hz), 119.8, 115.1, 114.9, 111.0, 86.7, 49.4, 25.9, 25.0; **^{19}F NMR (376 MHz, $CDCl_3$)** δ -105.7; **HRMS** (ESI, m/z) calcd for $C_{31}H_{25}FNO$ $[M + H]^+$ 446.1906 found 446.1920.

(3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(4-nitrophenyl)methanone (3j):



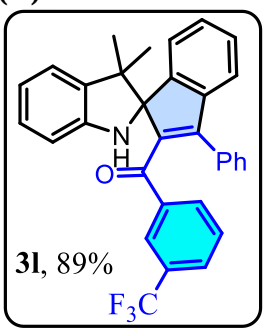
Yield 92% (87 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 197-199 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 8.04–7.97 (comp, 2H), 7.66–7.59 (comp, 2H), 7.36–7.29 (comp, 5H), 7.28–7.21 (comp, 4H), 7.04–6.98 (m, 1H), 6.97–6.90(m, 1H), 6.86–6.77 (m, 1H), 6,37 (d, *J* = 7.6 Hz, 1H), 4.45 (br s, 1 NH), 1.31 (s, 3H), 1.26 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 194.3, 149.5, 148.9, 148.2, 146.8, 143.0, 142.6, 141.1, 139.3, 132.8, 129.6, 128.9, 128.8, 128.64, 128.57, 127.8, 127.7, 124.4, 123.1, 122.4, 122.0, 119.9, 110.9, 86.8, 49.6, 26.1, 25.2; **HRMS** (ESI, m/z) calcd for $C_{31}H_{25}N_2O_3$ $[M + H]^+$ 473.1865 found 473.1853.

(3-chlorophenyl)(3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)methanone (3k):



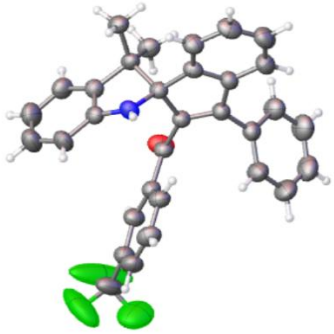
Yield 86% (80 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 163-165 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.44–l7.37 (comp, 2H), 7.36–7.21 (comp, 10H), 7.11 (t, *J* = 7.8 Hz, 1H), 7.02–6.98 (m, 1H), 6.94 (td, *J* = 7.6, 1.2 Hz, 1H), 6.81 (td, *J* = 7.4, 0.8 Hz, 1H), 6.45–6.23 (m, 1H), 4.45 (br s, 1 NH), 1.33 (s, 3H), 1.27 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 194.7, 148.3, 147.7, 146.6, 143.5, 141.4, 139.6, 139.4, 134.2, 133.0, 132.2, 129.2, 128.8, 128.7, 128.6, 128.5, 128.4, 127.5, 127.4, 126.8, 124.4, 122.1, 121.9, 119.9, 111.0, 86.8, 49.5, 26.1, 24.9; **HRMS** (ESI, m/z) calcd for $C_{31}H_{25}NOCl$ $[M + H]^+$ 462.1625 found 462.1628

(3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(3-(trifluoromethyl)phenyl)methanone (3l):



Yield 89% (88 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 148-150 °C; Crystallization was done from hot ethanol; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.75–7.67 (comp, 2H), 7.57 (d, *J* = 7.8 Hz, 1H), 7.36–7.29 (comp, 6H), 7.28–7.17 (comp, 4H), 7.00 (d, *J* = 7.3 Hz, 1H), 6.93 (td, *J* = 7.6, 1.0 Hz, 1H), 6.83-6.78 (m, 1H), 6.35 (d, *J* = 7.7 Hz, 4H), 4.46 (br s, 1 NH), 1.32 (s, 3H), 1.27 (s, 3H).; **^{13}C NMR (101 MHz, $CDCl_3$)** δ 194.7, 148.4 (d, *J* = 11.8 Hz), 146.8, 143.1, 141.3, 139.4, 138.5, 132.9, 131.6, 130.45 (d, *J* = 32.8 Hz), 128.8 (X2), 128.6, 128.52, 128.47 (X2), 127.61 (d, *J* = 7.1 Hz), 125.84 (d, *J* = 3.7 Hz), 124.9, 124.4, 123.52 (d, *J* = 271.6 Hz), 122.2, 121.9, 119.8, 110.9, 86.8, 49.5, 26.0, 25.2; **^{19}F NMR (376 MHz, $CDCl_3$)** δ -62.8; **HRMS** (ESI, m/z) calcd for $C_{32}H_{25}F_3NO$ $[M + H]^+$ 496.1888 found 496.1879.

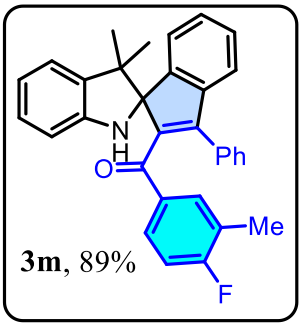
Figure S3. X-ray crystal structure of **3l** (ellipsoid contour at 50% probability level)



Crystal data and structure refinement for 3l (CCDC 2534748)

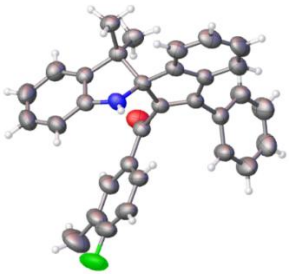
Identification code	IM_06_108ree_0ma_a	ρ_{calc}g/cm^3	1.305
Empirical formula	$C_{32}H_{24}F_3NO$	μ/mm^{-1}	0.771
Formula weight	495.52	F(000)	1032.0
Temperature/K	299.00	Crystal size/mm^3	0.12 × 0.09 × 0.05
Crystal system	orthorhombic	Radiation	Cu Kα (λ = 1.54178)
Space group	$Pca2_1$	2Θ range for data collection/°	7.28 to 133.636
a/Å	24.2871(14)	Index ranges	-28 ≤ h ≤ 28, -7 ≤ k ≤ 7, -19 ≤ l ≤ 19
b/Å	6.3963(4)	Reflections collected	55048
c/Å	16.2325(10)	Independent reflections	4450 [R_{int} = 0.0710, R_{sigma} = 0.0372]
α/°	90	Data/restraints/parameters	4450/1/336
β/°	90	Goodness-of-fit on F^2	1.050
γ/°	90	Final R indexes [I>=2σ (I)]	R_1 = 0.0607, wR_2 = 0.1626
Volume/Å^3	2521.7(3)	Final R indexes [all data]	R_1 = 0.0618, wR_2 = 0.1641
Z	4	Largest diff. peak/hole / e Å^{-3}	0.58/-0.39

(3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(4-fluoro-3-methylphenyl)methanone (3m):



Yield 89% (82 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 168-170 °C; Crystallization was done from hot ethanol; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.40–7.30 (comp, 7H), 7.30–7.20 (comp, 4H), 7.02–6.98 (m, 1H), 6.94 (td, *J* = 7.6, 1.3 Hz), 6.83–6.76 (comp, 2H), 6.37 (d, *J* = 7.7 Hz), 4.49 (br s, 1H), 2.14 (d, *J* = 1.6 Hz, 3H), 1.33 (s, 3H), 1.27 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 194.8, 165.1, 162.6, 148.4, 146.8 (d, *J* = 4.9 Hz), 143.8, 141.5, 139.6, 133.97 (d, *J* = 2.9 Hz), 133.2, 132.57 (d, *J* = 6.4 Hz), 128.8, 128.7, 128.4, 128.4, 128.3, 127.3, 127.2, 124.70 (d, *J* = 17.9 Hz), 124.3, 121.9 (d, *J* = 12.7 Hz), 119.8, 114.52 (d, J = 23.1 Hz), 111.1, 86.7, 49.5, 26.0, 25.1, 14.27 (d, *J* = 3.1 Hz); **^{19}F NMR (376 MHz, $CDCl_3$)** δ -109.9; **HRMS** (ESI, m/z) calcd for $C_{32}H_{27}FNO$ $[M + H]^+$ 460.2077 found 460.2070.

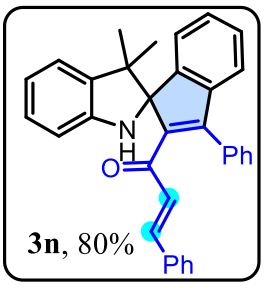
Figure S4. X-ray crystal structure of **3m** (ellipsoid contour at 50% probability level)



Crystal data and structure refinement for 3m (CCDC 2534749)

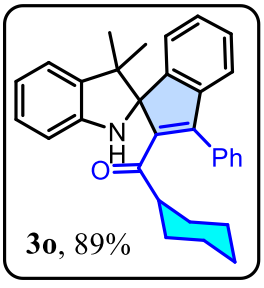
Identification code	ORTHO_a	ρ_{calc}g/cm^3	1.235
Empirical formula	$C_{32}H_{26}FNO$	μ/mm^{-1}	0.626
Formula weight	459.54	F(000)	968.0
Temperature/K	300.00	Crystal size/mm^3	0.15 × 0.1 × 0.06
Crystal system	orthorhombic	Radiation	Cu Kα (λ = 1.54178)
Space group	$Pca2_1$	2Θ range for data collection/°	7.436 to 136.518
a/Å	23.7750(6)	Index ranges	-28 ≤ h ≤ 28, -7 ≤ k ≤ 7, -19 ≤ l ≤ 19
b/Å	6.4009(2)	Reflections collected	51697
c/Å	16.2406(5)	Independent reflections	4519 [R_{int} = 0.0490, R_{sigma} = 0.0257]
α/°	90	Data/restraints/parameters	4519/1/320
β/°	90	Goodness-of-fit on F^2	1.045
γ/°	90	Final R indexes [I>=2σ (I)]	R_1 = 0.0402, wR_2 = 0.1089
Volume/Å^3	2471.52(13)	Final R indexes [all data]	R_1 = 0.0408, wR_2 = 0.1096
Z	4	Largest diff. peak/hole / e Å^{-3}	0.28/-0.17

(*E*)-1-(3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)-3-phenylprop-2-en-1-one (3n):



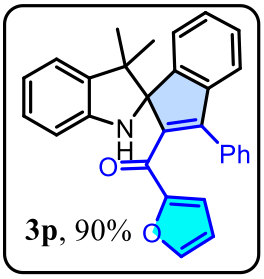
Yield 80% (73 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 103-105 °C; **^{1}H NMR (400 MHz, CDCl$_3$)** δ 7.48–7.35 (comp, 6H), 7.32–7.22 (comp, 6H), 7.20–7.12 (m, 5H), 7.08–7.03 (m, 1H), 6.89 (t, *J* = 7.0 Hz, 2H), 6.61 (d, *J* = 15.8 Hz, 1H), 1.33 (s, 3H), 1.15 (s, 3H); **^{13}C NMR (101 MHz, CDCl$_3$)** δ 192.2, 141.8, 141.2, 134.8, 133.8, 130.1, 128.9 (X2), 128.8, 128.70 (X3), 128.66 (X3), 128.3, 128.1(X3), 127.8, 127.6, 127.0, 123.7, 122.4, 122.1, 85.5, 49.8, 28.0, 25.3; **HRMS** (ESI, m/z) calcd for $C_{33}H_{28}NO$ $[M + H]^+$ 454.2171 found 454.2161.

cyclohexyl(3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)methanone (3o):



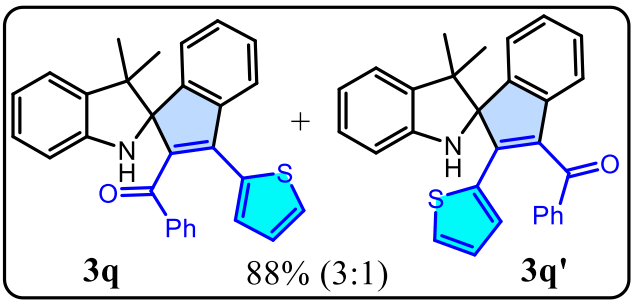
Yield 89% (77 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 113-115 °C; **^{1}H NMR (400 MHz, CDCl$_3$)** δ 7.46–7.35 (comp, 5H), 7.25-7.21 (m, 1H), 7.16–7.11 (m, 3H), 7.10–7.04 (comp, 2H), 6.90–6.83 (m, 1H), 6.77 (d, *J* = 7.7 Hz, 1H), 4.35 (br s, 1 NH), 2.83 (tt, *J* = 11.4, 3.0 Hz, 1H), 1.64–1.47 (m, 6H), 1.35 (s, 3H), 1.29–1.14 (m, 2H), 1.12 (s, 3H), 1.06–0.95 (m, 3H), 0.93–0.81 (m, 1H).**^{13}C NMR (101 MHz, CDCl$_3$)** δ 207.0, 148.8, 147.6, 147.3, 142.0, 139.7, 133.9, 128.9 (X2), 128.31, 128.26 (X2), 127.5, 127.1, 123.5, 122.2, 122.1, 119.8, 110.6, 85.2, 50.6, 49.6, 29.6, 27.2, 26.6, 26.1, 25.7, 25.4, 25.3; **HRMS** (ESI, m/z) calcd for $C_{31}H_{32}NO$ $[M + H]^+$ 434.2484 found 434.2472.

(3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(furan-2-yl)methanone (3p):



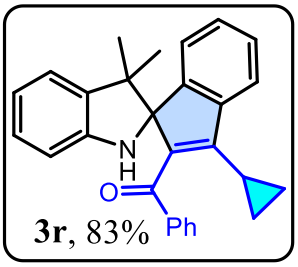
Yield 90% (76 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 145-147 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.37–7.25 (m, 10H), 7.25–7.18 (m, 1H), 7.02–6.94 (m, 2H), 6.80 (td, *J* = 7.4, 0.9 Hz, 1H), 6.77–6.74 (m, 1H), 6.55 (d, *J* = 7.7 Hz, 1H), 6.26 (dd, *J* = 3.6, 1.7 Hz, 1H), 4.62 (br s, 1 NH), 1.35 (s, 3H), 1.26 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 183.0, 153.2, 148.7, 147.2, 146.9, 145.8, 143.5, 141.7, 139.4, 133.3, 128.5, 128.4 (X2), 128.3, 127.4, 127.2, 124.3, 122.0, 121.8, 119.9, 117.9, 112.1, 110.5, 86.9, 49.3, 25.9, 24.9; **HRMS** (ESI, m/z) calcd for $C_{29}H_{24}NO_2$ $[M + H]^+$ 418.1807 found 418.1801.

(3',3'-dimethyl-3-(thiophen-2-yl)spiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (3q), (3',3'-dimethyl-2-(thiophen-2-yl)spiro[indene-1,2'-indolin]-3-yl)(phenyl)methanone (3q´):



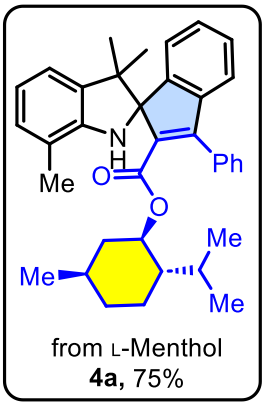
Combined Yield 88% inseparable regioisomers (**3r**:**3r´** = 3:1) (76 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 133-135 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.91–7.86 (m, 0.75H), 7.57-7.54 (m, 0.46H), 7.50–7.48 (comp, 2.32H), 7.48–7.43 (m, 1.22H), 7.42–7.32 (m, 5H), 7.25–7.18 (m, 4.15H), 7.18–7.10 (comp, 1.77H), 7.08–6.96 (comp, 3H), 6.90–6.76 (comp, 2.82H), 6.20 (d, *J* = 7.6 Hz, 1H), 4.32 (br s, 1 NH), 1.34 (s, 3H), 1.27 (s, 3H), 1.11 (s, 1H), 1.08 (s, 1H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 196.4, 196.1, 149.1, 148.1, 147.9, 146.2, 144.9, 144.5, 141.6, 140.7, 140.54, 140.47, 139.50, 139.1, 138.0, 136.3, 136.2, 133.5, 133.3, 132.6, 129.3, 128.6, 128.52, 128.49, 128.4, 128.3, 128.0, 127.8, 127.6, 127.4, 127.0, 126.0, 125.8, 125.4, 124.9, 124.6, 124.4, 122.9, 122.3, 121.8, 121.8, 120.9, 119.8, 119.4, 110.9, 109.9, 86.7, 84.8, 49.7, 49.4, 28.9, 26.4, 24.4, 24.2; **HRMS** (ESI, m/z) calcd for $C_{29}H_{24}NOS$ $[M + H]^+$ 434.1579 found 434.1570.

(3-cyclopropyl-3',3'-dimethylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (3r):



Yield 83% (66 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 108-110 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.72-7.68 (comp, 2H), 7.53 (tt, *J* = 7.0, 1.3 Hz, 1H), 7.47–7.44 (m, 1H), 7.40-7.34 (comp, 2H), 7.33 (td, *J* = 7.4, 1.5 Hz, 1H), 7.23–7.14 (comp, 2H), 7.01–6.93 (m, 1H), 6.91–6.84 (m, 1H), 6.78 (t, ϑ = 7.3 Hz, 1H), 6.25 (d, *J* = 6.9 Hz, 1H), 4.38 (br s, 1 NH), 1.68–1.56 (m, 1H), 1.26 (s, 3H), 1.18 (s, 3H), 0.84-0.77 (m, 1H), 0.74–0.66 (m, 1H), 0.64–0.52 (m, 2H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 197.1, 146.4, 142.1, 139.8, 139.1, 132.7, 128.7 (X3), 128.4, 128.2 (X3), 127.4, 126.8, 124.0, 121.7, 121.2, 119.9, 86.2, 49.0, 25.8, 24.9, 9.8, 6.1, 6.0; **HRMS** (ESI, m/z) calcd for $C_{28}H_{26}NO$ $[M + H]^+$ 392.2014 found 392.2009.

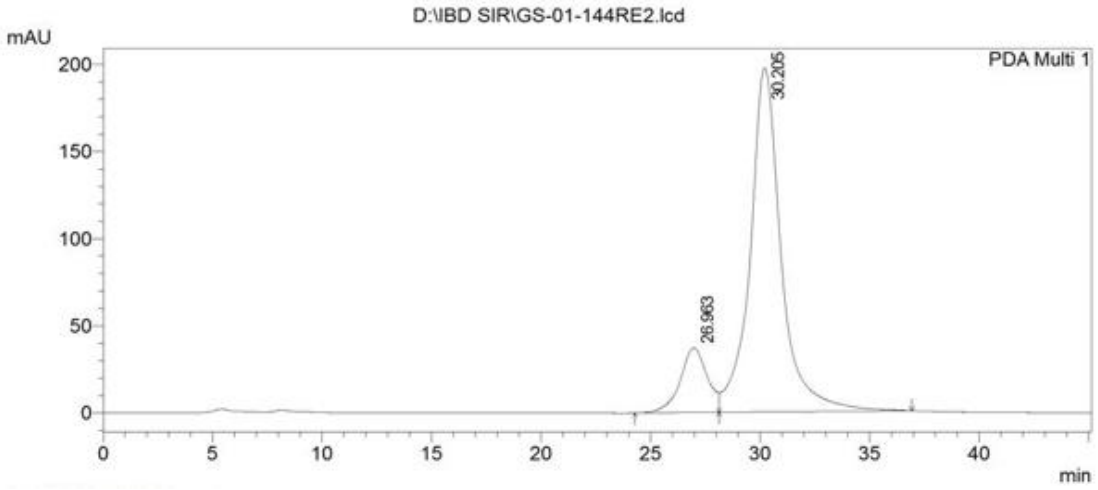
(1R,2S,5R)-2-isopropyl-5-methylcyclohexyl 3',3',7'-trimethyl-3-phenylspiro[indene-1,2'-indoline]-2-carboxylate (4a)



Yield 70% (72 mg); diastereomeric ratio (dr = 74:26); Yellow gel; **eluent composition** petroleum ether/ethyl acetate = 19:1; Data for major isomer; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.47-7.43 (m, 1H), 7.42-7.32 (comp, 5H), 7.27-7.18 (comp, 2H), 7.10-7.05 (m, 1H), 6.91 (d, J = 7.3 Hz, 1H), 6.86 (d, J = 7.3 Hz, 1H), 6.75-6.70 (m, 1H), 4.32 (td, J = 10.8, 4.2 Hz, 1H), 2.23 (s, 3H), 1.55-1.42 (m, 4H), 1.34 (s, 3H), 1.23 (s, 3H), 1.21-1.15 (m, 1H), 1.01-0.91 (m, 2H), 0.88-0.79 (m, 2H), 0.76 (d, J = 6.4 Hz, 3H), 0.72-0.66 (m, 2H), 0.51 (d, J = 7.0 Hz, 3H), 0.35 (d, J = 6.8 Hz, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 166.8, 156.6, 145.9, 142.7, 138.2, 133.8, 128.56, 128.61, 128.5, 128.4 (x3), 128.3, 128.2, 128.0, 126.6, 125.1, 121.7, 119.7, 85.5, 74.9, 49.3, 46.3, 39.7, 34.1, 31.2, 28.7, 24.5, 23.4, 22.5, 22.0, 20.9, 16.8, 15.6.; **HRMS** (ESI, m/z) calcd for $C_{36}H_{42}NO_2$ $[M + H]^+$ 520.3203 found 520.3203; ; $[\alpha]_D^{25}$ = (-) 54.900 (c = 1, EtOH).

HPLC purity data for 4a: (Phenomenex™ Luna 5 µm C18(2) 100 Å, LC Column 250 x 4.6 mm, 90% MeCN/H_20, 0.45 mL/min)

<Chromatogram>

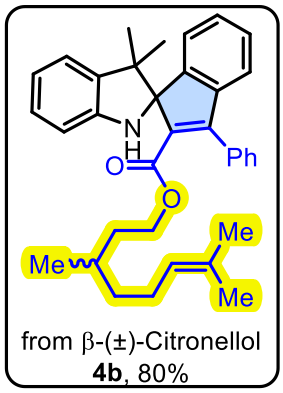


PeakTable

PDA Ch1 254nm 4nm

Peak#	Ret. Time	Area	Height	Area %	Height %
1	26.963	3205785	37092	14.115	15.815
2	30.205	19505353	197447	85.885	84.185
Total		22711138	234539	100.000	100.000

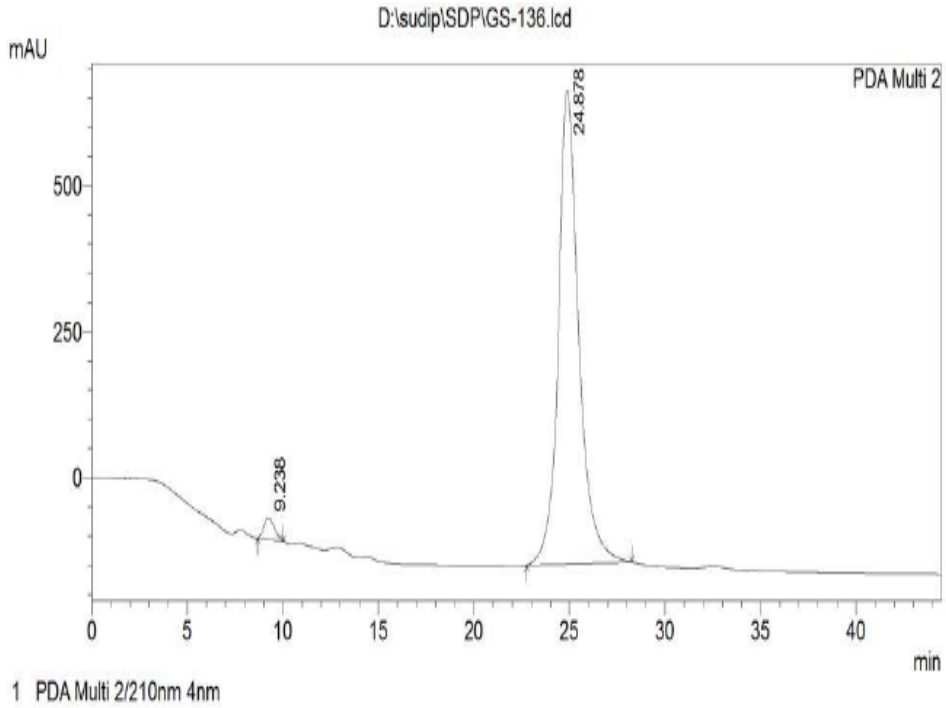
3,7-dimethyloct-6-en-1-yl 3',3'-dimethyl-3-phenylspiro[indene-1,2'-indoline]-2-carboxylate (4b)



Yield 80% (81 mg) inseparable diastereomers (dr = 1:1); Yellow gel; **eluent composition** petroleum ether/ethyl acetate = 19:1; **^{1}H NMR (400 MHz, $CDCl_3$)** Data for major isomer δ 7.46 – 7.34 (comp, 6H), 7.30 – 7.16 (comp, 4H), 7.9-7.07 (m, 1H), 7.04 (app d, *J* = 6.9 Hz, 1H), 6.87 – 6.78 (comp, 2H), 5.08 – 5.02 (m, 1H), 3.88 – 3.76 (m, 1H), 3.71 – 3.61 (m, 1H), 1.95-178 (m, 2H), 1.68 (s, 3H), 1.59 (s, 3H), 1.36 (s, 3H), 1.33-1.13 (m, 3H and d, *J* = 2.0, 3H), 1.10-0.96 (m, 2H), 0.73 (dd, *J* = 6.4, 1.3 Hz, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** 166.9, 149.1, 146.4, 141.9, 139.1, 133.6, 131.2, 128.29 (x3), 128.27 (x3), 127.5, 127.0, 124.6, 124.7, 122.0 (x2), 119.6, 111.1, 84.9, 63.1, 49.2, 36.9, 34.9, 29.2, 27.5, 25.7, 25.3, 24.1, 19.2, 17.6; **HRMS** (ESI, m/z) calcd for $C_{35}H_{40}NO_2$ $[M + H]^+$ 506.3047 found 506.3056; $[\alpha]_D^{25}$ = (-) 0.100 (c = 1, EtOH).

HPLC purity data for 4b: (Phenomenex™ Luna 5 µm C18(2) 100 Å, LC Column 250 x 4.6 mm, 90% MeCN/H_20, 0.45 mL/min)

<Chromatogram>



1 PDA Multi 2/210nm 4nm

PeakTable

PDA Ch2 210nm 4nm

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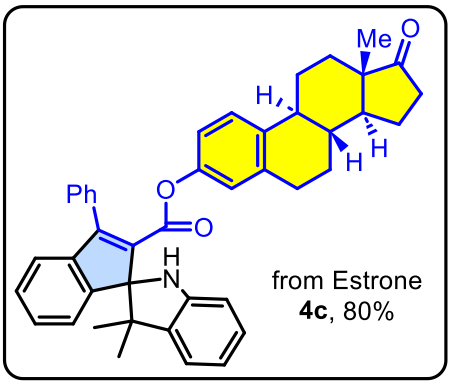
1	9.238	1413826	37732	2.276	4.451
2	24.878	60718111	809972	97.724	95.549
Total		62131937	847705	100.000	100.000

(8R,9S,13S,14S)-13-methyl-17-oxo-7,8,9,11,12,13,14,15,16,17-decahydro-6*H*-cyclopenta[a]phenanthren-3-yl 3',3'-dimethyl-3-phenylspiro[indene-1,2'-indoline]-2-carboxylate (4c)

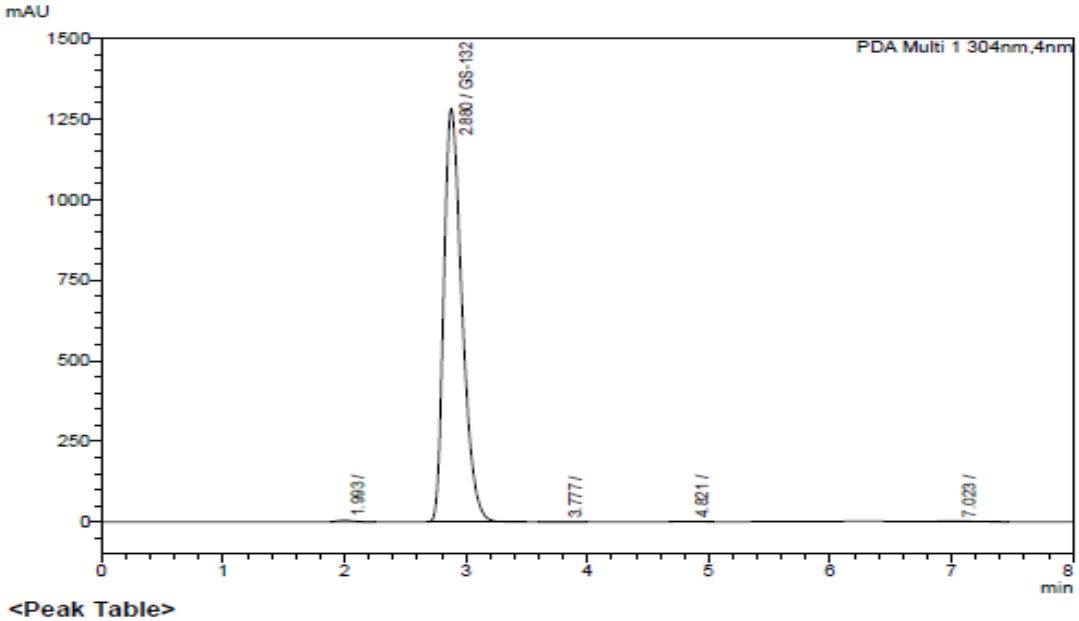


Yield 80% (99 mg) inseparable diastereomers (dr = 77:23); Yellow gel; **eluent composition** petroleum ether/ethyl acetate = 19:1; Data for major isomer **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.54 – 7.39 (comp, 5H), 7.36 – 7.23 (comp, 4H), 7.15-7.06 (comp, 3H), 6.97-6.87 (m, 1H), 6.89 (app t, *J* = 7.4 Hz, 1H), 6.82 (d, *J* = 7.8 Hz, 1H), 6.24 (app td, *J* = 8.5, 2.4 Hz, 1H), 6.09 (dd, *J* = 6.3, 2.2 Hz, 1H), 2.80 – 2.73 (m, 2H), 2.57 – 2.44 (m, 1H), 2.40 – 2.29 (m, 1H), 2.27 – 2.00 (m, 3H), 1.98-1.91 (m, 2H), 1.68 – 1.43 (m, 4H), 1.47 (s, 3H), 1.46-1.42 (m, 1H), 1.41 – 1.34 (m, 1H), 1.30 (s, 3H), 0.87 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** Data for major isomer δ 220.8, 165.6, 149.1, 148.0, 146.0, 141.8, 138.9, 137.6, 137.1, 133.2, 128.5, 128.5, 128.4 (x3), 127.88, 127.86, 127.2, 126.0, 125.0, 122.3, 122.2, 121.4, 119.7, 118.4, 111.3, 85.5, 50.4, 49.3, 47.9, 44.1, 37.9, 35.8, 31.5, 29.3, 28.1, 26.3, 25.7, 23.4, 21.5, 13.8; **HRMS** (ESI, m/z) calcd for $C_{43}H_{42}NO_3$ $[M + H]^+$ 620.3153 found 620.3165;

$[\alpha]_D^{25}$ = (-) 0.400 (c = 1, EtOH).

HPLC purity data for 4c: (Shimadzu, Shim-pack GIST, C18-5µm, 4.6x2500 mm; 90% MeCN/H_20, 2.5 mL/min)

<Chromatogram>



<Peak Table>

PDA Ch1 304nm

Peak#	Ret. Time	Area	Height	Name	Area%
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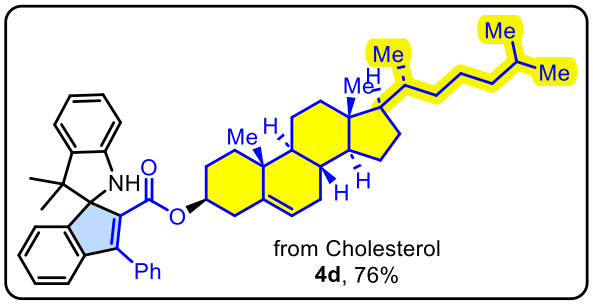
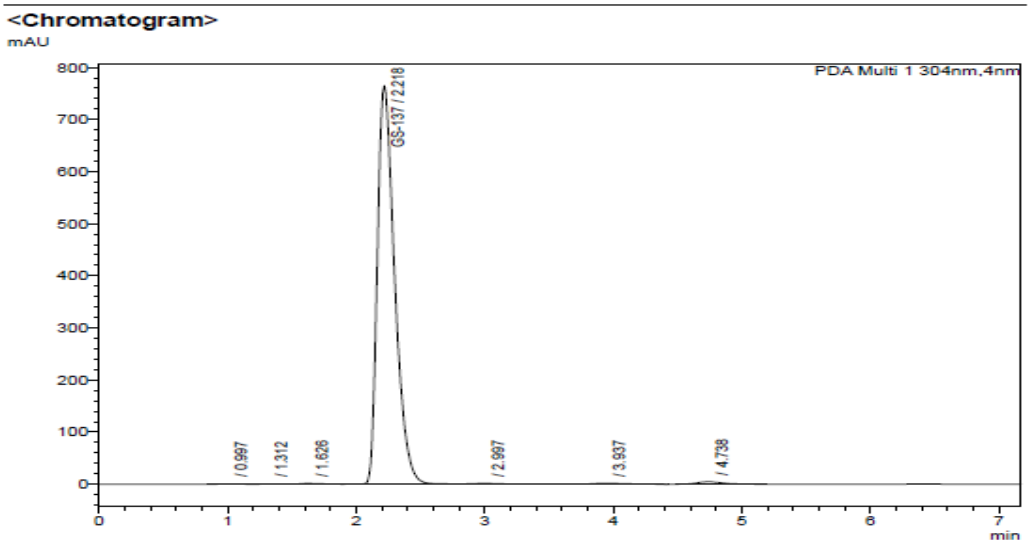
2	2.880	13330008	1283026	GS-132	99.336
3	3.777	7428	613		0.055
4	4.821	10487	912		0.078
5	7.023	26011	1629		0.194
Total		13419143	1291351		100.000

(3S,8S,9S,10R,13R,14S,17R)-10,13-dimethyl-17-((R)-6-methylheptan-2-yl)-2,3,4,7,8,9,10,11,12,13,14,15,16,17-tetradecahydro-1*H*-cyclopenta[a]phenanthren-3-yl 3',3'-dimethyl-3-phenylspiro[indene-1,2'-indoline]-2-carboxylate (4d)



Yield 76% (112 mg) inseparable diastereomers (dr = 1:1); Yellow gel; **eluent composition** petroleum ether/ethyl acetate = 19:1; Data for inseparable diastereomers **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.46 – 7.33 (comp, 12H), 7.28 – 7.09 (comp, 5H), 7.04 (app d, *J* = 7.6 Hz, 2H), 6.88 – 6.81 (comp, 3H), 5.21-5,24 (m, 1H), 5.21 – 5.17 (m, 1H), 4.39-4.29 (m, 2H), 1.99-192 (m, 5H), 1.91 – 1.85 (m, 1H), 1.85 – 1.76 (m, 3H), 1.73-1.62 (m, 4H), 1.57-1.46 (m, 8H), 1.46 – 1.39 (m, 5H), 1.37 (d, *J* = 1.9 Hz, 6H), 1.35 – 1.31 (m, 3H), 1.26 (s, , 3H), 1.21 (s, 6H), 1.17-1.07 (m, 10H), 1.06 – 0.94 (m, 11H), 0.91 (s, 3H), 0.89 (s, 3H), 0.88 (s, 6H), 0.87 (d, *J* = 1.7 Hz, 6H), 0.85 (d, *J* = 1.7 Hz, 6H), 0.65 (s, 6H); Data for major isomer **^{13}C NMR (101 MHz, $CDCl_3$)** 166.1, 146.8, 142.0, 139.7, 139.4, 133.8, 128.5 (x2), 128.4, 128.3 (x2), 128.2 (x2), 127.7, 127.2, 124.8, 122.4, 122.22, 122.19, 119.9, 111.5, 84.7, 74.4, 56.7, 50.1, 49.4, 42.4, 39.8, 39.8, 37.5, 37.1, 36.9, 36.6, 36.3, 35.9, 31.9, 28.3, 28.1, 27.9, 27.1, 27.2, 24.6, 24.3, 23.9, 22.8, 22.6, 21.0, 19.2, 18.7, 11.8; **HRMS** (ESI, m/z) calcd for $C_{52}H_{66}NO_2$ $[M + H]^+$ 736.5094 found 736.5093; $[\alpha]_D^{25}$ = (-) 2.700 (c = 1, EtOH).

HPLC purity data for 4d: (Shimadzu, Shim-pack GIST, C18-5µm, 4.6x2500 mm; 90% MeCN/H_20, 2.5 mL/min)

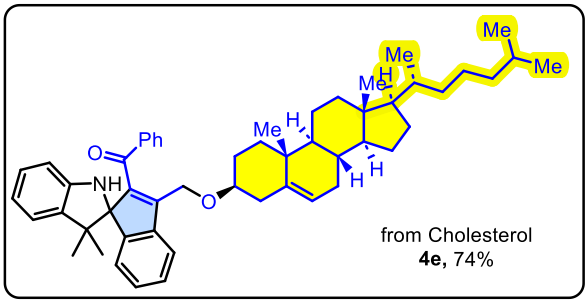


<Peak Table>

PDA Ch1 304nm

Peak#	Ret. Time	Area	Height	Name	Area%
1	0.997	2016	191		0.028
2	1.312	1313	94		0.018
3	1.626	11867	1195		0.164
4	2.218	7126116	764735	GS-137	98.468
5	2.997	16768	1317		0.232
6	3.937	21620	1535		0.299
7	4.738	57320	4487		0.792
Total		7237020	773554		100.000

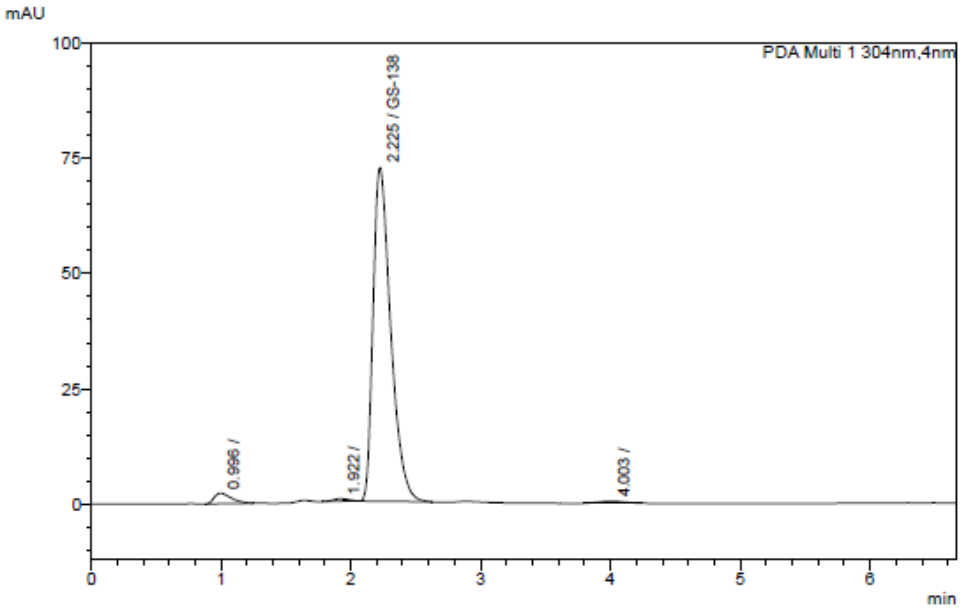
(3-((((3S,8S,9S,10R,13R,14S,17R)-10,13-dimethyl-17-((R)-6-methylheptan-2-yl)-2,3,4,7,8,9,10,11,12,13,14,15,16,17-tetradecahydro-1*H*-cyclopenta[a]phenanthren-3-yl)oxy)methyl)-3',3'-dimethylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (4e)



Yield 74% (111 mg) inseparable diastereomers (dr = 1:1); Yellow gel; **eluent composition** petroleum ether/ethyl acetate = 19:1; **^{1}H NMR (400 MHz, $CDCl_3$)** data for inseparable mixture δ 7.92 – 7.86 (m, 1H), 7.67 – 7.47 (comp, 10H), 7.41-7.28 (comp, 9H), 7.22-7.18 (comp, 2H), 6.94 (app d, *J* = 6.7 Hz, 4H), 6.83 – 6.71 (comp, 4H), 6.13 – 6.02 (comp, 2H), 5.32 – 5.24 (m, 1H), 5.21-5.18 (m, 1H), 4.37 (s, 2H), 4.37 (s, 2H), 3.17-3.07 (comp, 2H), 2.27 – 2.17 (m, 2H), 2.14 – 2.03-1.90 (m, 4H), 1.87-1.74(m, 6H), 1.60 – 1.28 (m, 27H), 1.25 (d, *J* = 1.6 Hz, 6H), 1.12 (s, 6H), 1.17-0.94 (m, 20H), 0.93 (s, 6H), 0.92 (s, 3H), 0.90 (s, 3H), 0.87 (d, *J* = 1.6 Hz, 6H), 0.86 (d, *J* = 1.6 Hz, 6H), 0.66 (s, 6H); **^{13}C NMR (101 MHz, $CDCl_3$)** data for major isomer 196.4, 147.9, 146.5, 142.9, 141.4, 140.6, 138.9, 132.8, 129.7, 129.0, 128.6, 128.47, 128.46, 128.2, 127.4, 126.8, 124.4, 121.81, 121.75, 119.7, 110.7, 87.0, 78.9, 62.2, 56.7, 56.1, 50.1, 48.9, 42.3, 39.8, 39.5, 38.7, 38.6, 37.1, 36.8, 36.2, 36.8, 31.9, 31.8, 28.2, 28.1, 28.0, 27.9, 26.7, 24.3, 23.8, 22.8, 22.9, 21.0, 19.3, 18.7, 11.8; **HRMS** (ESI, m/z) calcd for $C_{53}H_{68}NO_2$ $[M + H]^+$ 750.5250 found 750.5231. $[\alpha]_D^{25}$ = (-) 16.300 (c = 1, EtOH).

HPLC purity data for 4e: (Shimadzu, Shim-pack GIST, C18-5µm, 4.6x2500 mm; 90% MeCN/H_20, 2.5 mL/min)

<Chromatogram>

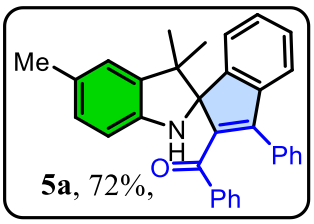


<Peak Table>

PDA Ch1 304nm

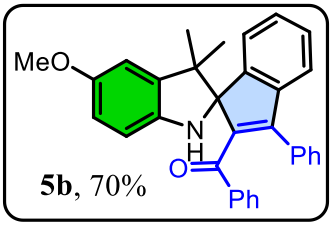
Peak#	Ret. Time	Area	Height	Name	Area%
1	0.996	20704	2286		2.894
2	1.922	4377	551		0.612
3	2.225	684338	72371	GS-138	95.648
4	4.003	6060	420		0.847
Total		715479	75628		100.000

phenyl(3',3',5'-trimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)methanone (5a):



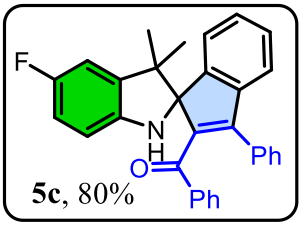
Yield 72% (64 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 168-170 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.53–7.50 (comp, 2H), 7.36–7.33 (comp, 2H), 7.32–7.29 (m, 2H), 7.28 (s, 1H), 7.26–7.15 (m, 7H), 6.81 (s, 1H), 6.73 (d, *J* = 7.8 Hz, 1H), 6.24 (d, *J* = 7.8 Hz, 1H), 2.29 (s, 3H), 1.32 (s, 3H), 1.24 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 196.4, 147.0, 141.6, 140.2, 137.9, 133.3, 132.4, 128.9 (X2), 128.8 (X2), 128.3 (X4), 127.9 (X2), 127.8, 127.1, 124.3, 122.6, 121.9, 111.5, 86.8, 49.5, 25.7, 25.3, 21.0; **HRMS** (ESI, m/z) calcd for $C_{32}H_{28}NO$ $[M + H]^+$ 442.2171 found 442.2164.

(5'-methoxy-3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (5b):



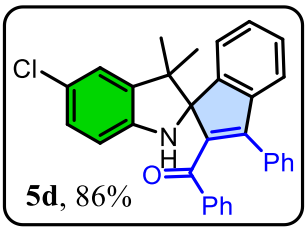
Yield 70% (64 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 163-165 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.55–7.51 (comp, 2H), 7.36–7.30 (comp, 5H), 7.28–7.15 (m, 7H), 6.63 (d, *J* = 2.6 Hz, 1H), 6.50 (dd, *J* = 8.4, 2.5 Hz, 1H), 6.29 (d, *J* = 6.1 Hz, 1H), 3.78 (s, 3H), 1.31 (s, 3H), 1.24 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 196.4, 154.7, 147.2, 147.0, 143.4, 142.0, 141.7, 141.6, 137.9, 133.2, 132.5, 128.8 (X2), 128.3 (X2), 127.9, 127.2, 124.2, 122.0, 112.6, 112.2, 108.8, 87.0, 55.8, 49.8, 25.5, 25.3; **HRMS** (ESI, m/z) calcd for $C_{32}H_{28}NO_2$ $[M + H]^+$ 458.2120 found 458.2119.

(5'-fluoro-3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (5c):



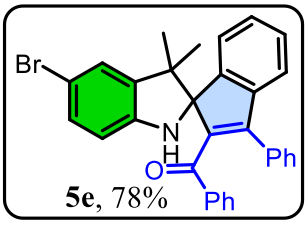
Yield 80% (72 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 136-138 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.54–7.50 (comp, 2H), 7.38–7.31 (comp, 5H), 7.28–7.17 (comp, 6H), 6.73 (dd, *J* = 8.2, 2.6 Hz, 1H), 6.64–6.57 (m, 1H), 6.21 (dd, *J* = 8.4, 4.3 Hz, 1H), 4.57 (br s, 1 NH), 1.34 (s, 3H), 1.26 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 196.3, 159.3, 156.9, 147.0, 146.3, 143.80 (d, *J* = 45.2 Hz), 141.80 (d, *J* = 7.2 Hz), 141.7, 137.8, 133.1, 132.6, 128.84, 128.80, 128.5, 128.4, 128.3, 128.0, 127.2, 124.4, 122.1, 113.48 (d, *J* = 23.4 Hz), 111.79 (d, *J* = 7.9 Hz), 109.55 (d, *J* = 23.9 Hz), 87.1, 49.7, 25.9, 24.7; **^{19}F NMR (376 MHz, $CDCl_3$)** δ -124.2; **HRMS** (ESI, m/z) calcd for $C_{31}H_{25}NOF$ $[M + H]^+$ 446.1920 found 446.1928.

(5'-chloro-3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (5d):



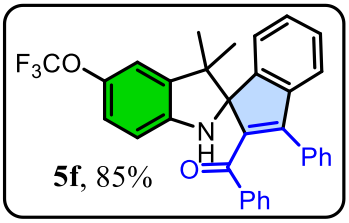
Yield 86% (80 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 174-176 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.56–7.52 (comp, 2H), 7.38–7.29 (comp, 6H), 7.28–7.15 (comp, 6H), 6.95 (d, *J* = 2.1 Hz, 1H), 6.88 (dd, *J* = 8.2, 2.1 Hz, 1H), 6.24 (d, *J* = 8.2 Hz, 1H), 1.33 (s, 3H), 1.25 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 196.1, 147.1, 146.9, 146.3, 143.4, 141.5, 137.7, 133.0, 132.6, 128.9, 128.8, 128.6, 128.4, 128.3 (X2), 128.0 (X2), 127.2, 124.6, 124.3, 122.3, 122.1, 111.8, 49.7, 25.9, 25.0; **HRMS** (ESI, m/z) calcd for $C_{31}H_{25}NOCl$ $[M + H]^+$ 462.1625 found 462.1631.

(5'-bromo-3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (5e):



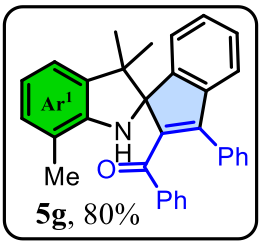
Yield 78% (79 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 176-178 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.57–7.50 (comp, 2H), 7.38–7.29 (comp, 6H), 7.28–7.15 (comp, 6H), 6.95 (d, *J* = 2.0 Hz, 1H), 6.88 (dd, *J* = 8.2, 2.1 Hz, 1H), 6.24 (d, *J* = 8.2 Hz, 1H), 4.56 (br s, 1 NH), 1.33 (s, 3H), 1.25 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 196.1, 147.1, 146.9, 146.3, 143.3, 141.6, 137.8, 133.0, 132.6, 128.9, 128.8, 128.6, 128.4, 128.4 (X2), 128.0 (X2), 127.2, 124.6, 124.3, 122.3, 122.1, 111.9, 86.8, 49.7, 25.9, 25.0; **HRMS** (ESI, m/z) calcd for $C_{31}H_{25}NOBr$ $[M + H]^+$ 506.1120 found 506.1104.

(3',3'-dimethyl-3-phenyl-5'-(trifluoromethoxy)spiro[indene-1,2'-indolin]-2yl)(phenyl)methanone (5f):



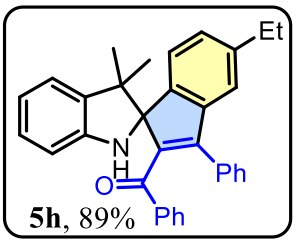
Yield 85% (87 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 154-156 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.50-7.46 (comp, 2H), 7.40–7.32 (comp, 6H), 7.29–7.16 (comp, 6H), 6.86–6.82 (m, 1H), 6.76–6.70 (m, 1H), 6.16 (d, *J* = 8.4, 1H) 4.49 (s, 1 NH), 1.35 (s, 3H), 1.28 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 196.1, 147.0, 146.6, 145.5, 144.0, 142.7, 141.8, 140.9, 133.0, 132.6, 128.8, 128.70, 128.67, 128.44, 128.38, 128.0, 127.1, 124.6, 122.1, 121.93–119.39 (m), 120.5, 119.4, 115.7, 110.7, 87.1, 49.5, 26.5, 24.0; **^{19}F NMR (376 MHz, $CDCl_3$)** δ -58.2; **HRMS** (ESI, m/z) calcd for $C_{31}H_{25}NOF_3$ $[M + H]^+$ 512.1837 found 512.1832.

phenyl(3',3',7'-trimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)methanone (5g):



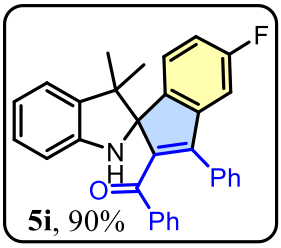
Yield 80% (71 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 184-186 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.51–7.48 (m, 1H), 7.46–7.40 (comp, 4H), 7.38–7.33 (comp, 3H), 7.32–7.19 (comp, 6H), 6.87 (d, *J* = 7.0 Hz, 1H), 6.79–6.68 (m, 2H), 4.23 (br s, 1 NH), 1.56 (s, 3H), 1.42 (s, 3H), 1.31 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 196.3, 146.3, 145.8, 145.6, 144.5, 142.3, 139.2, 138.3, 133.1, 132.5, 128.8, 128.6, 128.44, 128.36, 128.2, 128.1, 128.0, 126.7, 125.0, 121.7, 121.0, 120.2, 119.4, 87.0, 49.5, 27.3, 23.0, 15.8; **HRMS** (ESI, m/z) calcd for $C_{32}H_{28}NO$ $[M + H]^+$ 442.2171 found 442.2181.

(5-ethyl-3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (5h):



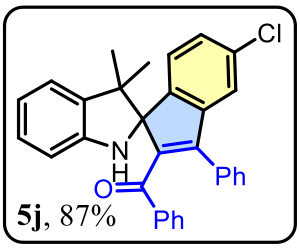
Yield 89% (81 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 155-157 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.53–7.48 (comp, 2H), 7.41–7.31 (comp, 3H), 7.30–7.21 (comp, 4H), 7.21–7.14 (comp, 3H), 7.08 (d, *J* = 7.7 Hz, 0H), 7.00 (d, *J* = 7.2 Hz, 0H), 6.89 (t, *J* = 7.5 Hz, 0H), 6.79 (t, J = 7.3 Hz, 0H), 6.32 – 6.22 (m, 17H), 4.49 (br s, 1 NH), 2.66 (q, *J* = 7.5 Hz, 2H), 1.37 (s, 3H), 1.29 (s, 3H), 1.26–1.21 (m, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 196.2, 148.2, 146.5, 144.8, 144.6, 143.7, 142.0, 139.7, 138.1, 133.3, 132.4, 128.84, 128.78, 128.3, 128.2, 127.9, 127.3, 126.6, 124.3, 121.8, 121.5, 119.8, 111.0, 86.5, 49.3, 28.9, 26.3, 24.5, 15.7; **HRMS** (ESI, m/z) calcd for $C_{33}H_{30}NO$ $[M + H]^+$ 456.2327 found 456.2314.

(5-fluoro-3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (5i):



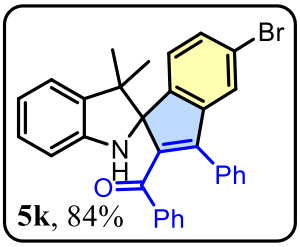
Yield 90% (80 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 164-166 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.52–7.46 (comp, 2H), 7.39–7.32 (comp, 3H), 7.31–7.16 (comp, 6H), 7.06–6.97 (comp, 2H), 6.95–6.85 (comp, 2H), 6.79 (td, *J* = 7.4, 0.9 Hz, 1H), 6.24 (d, *J* = 7.6 Hz, 1H), 4.48 (br s, 1 NH), 1.35 (s, 3H), 1.27 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 195.7, 164.6, 162.2, 147.9, 146.3, 145.31–145.02 (m), 144.01 (d, *J* = 8.5 Hz), 141.6 (d, *J* = 2.4 Hz), 139.4, 137.7, 132.6, 128.73, 128.66, 128.6, 128.5, 128.0, 127.5, 125.49 (d, *J* = 8.6 Hz), 121.9, 120.0, 113.41 (d, *J* = 22.6 Hz), 111.2, 109.26 (d, *J* = 23.9 Hz), 86.2, 49.4, 26.4, 24.3; **^{19}F NMR (376 MHz, $CDCl_3$)** δ -113.4; **HRMS** (ESI, m/z) calcd for $C_{31}H_{25}NOF$ $[M + H]^+$ 446.1920 found 446.1908.

(5-chloro-3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (5j):



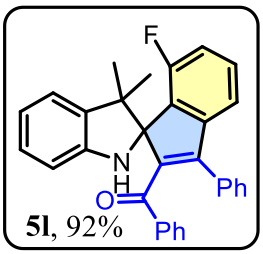
Yield 87% (81 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 172-174 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.50-7.46 (comp, 2H), 7.39–7.31 (comp, 3H), 7.30–7.22 (comp, 5H), 7.22–7.17 (comp, 3H), 7.01–6.97 (m, 1H), 6.90 (td, *J* = 7.6, 1.3 Hz, 1H), 6.81 (td, *J* = 7.4, 0.9 Hz, 1H), 4.63 (br s, 1 NH), 1.34 (s, 3H), 1.26 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 195.7, 147.7, 145.6, 145.4, 144.6, 143.6, 139.4, 137.6, 134.4, 132.7, 132.5, 128.8, 128.7, 128.6, 128.5, 128.0, 127.6, 126.9, 125.4, 122.1, 121.9, 120.2, 111.3, 86.3, 49.5, 26.2, 24.6; **HRMS** (ESI, m/z) calcd for $C_{31}H_{25}ClNO$ $[M + H]^+$ 462.1625 found 462.1620.

(5-bromo-3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (5k):



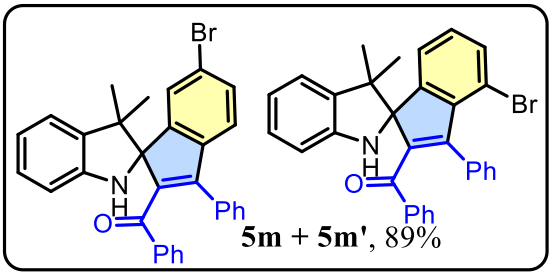
Yield 84% (85 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 165-167 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.50–7.45 (comp, 2H), 7.43 (d, *J* = 1.7 Hz, 1H), 7.39–7.30 (comp, 4H), 7.29–7.22 (comp, 3H), 7.21–7.16 (comp, 3H), 6.99 (d, *J* = 7.3 Hz, 1H), 6.94–6.88 (m, 1H), 6.81 (t, *J* = 7.3 Hz, 1H), 6.30 (d, *J* = 7.4 Hz, 1H), 4.76 (br s, 1 NH), 1.34 (s, 3H), 1.25 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 195.7, 145.1, 143.9, 139.4, 137.6, 132.7, 132.5, 129.8, 128.8, 128.7, 128.6 (X2), 128.5 (X2), 128.0 (X2), 127.6, 125.8, 125.0, 122.4, 121.9, 120.3, 111.5, 86.4, 49.5, 26.1, 24.7; **HRMS** (ESI, m/z) calcd for $C_{31}H_{25}NOBr$ $[M + H]^+$ 506.1120 found 506.1125.

(7-fluoro-3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (5l):



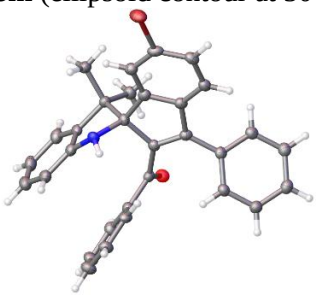
Yield 92% (92 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 149-151 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.52–7.47 (comp, 2H), 7.38–7.32 (m, 3H), 7.31–7.16 (comp, 7H), 7.11 (d, *J* = 7.4 Hz, 1H), 7.01–6.93 (m, 2H), 6.90 (td, *J* = 7.6, 1.3 Hz, 1H), 6.79 (td, *J* = 7.4, 0.9 Hz, 1H), 6.28 (d, *J* = 7.7 Hz, 1H), 4.74 (br s, 1 NH), 1.39 (s, 3H), 1.35 (d, *J* = 2.9 Hz, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 195.6, 160.1, 157.5, 147.5, 145.69-145.53 (m), 145.45-145.29 (m), 145.0 (d, *J* = 5.9 Hz), 140.0, 137.7, 132.8, 132.6, 131.24 (d, *J* = 15.5 Hz), 130.25 (d, *J* = 7.7 Hz), 128.85 (d, *J* = 2.6 Hz), 128.5, 128.4, 127.9, 127.4, 121.4, 119.9, 118.09 (d, *J* = 2.6 Hz), 115.79 (d, *J* = 23.7 Hz), 111.3, 87.78 (d, *J* = 3.0 Hz), 50.7, 26.88 (d, *J* = 5.1 Hz), 25.5; ; **^{19}F NMR (376 MHz, $CDCl_3$)** δ -109.1; **HRMS** (ESI, m/z) calcd for $C_{31}H_{25}NOF$ $[M + H]^+$ 446.1920 found 446.1917.

(6-bromo-3',3'-dimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)(phenyl)methanone (5m):



Combined Yield 89% (90 mg) regioisomeric ratio (**5m**:**5m'**=94:6) ; Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 150-152 °C; Crystallization was done from hot ethanol; Data for major isomer **^{1}H NMR (400 MHz, CDCl$_3$)** 1H NMR (400 MHz, Chloroform-d) δ 7.48-7.43 (comp, 3H), 7.44–7.37 (m, 1H), 7.37–7.29 (comp, 3H), 7.27-7.24 (comp, 2H), 7.23–7.14 (comp, 4H), 7.00 – 6.96 (m, 1H), 6.89 (td, *J* = 7.6, 1.3 Hz, 1H), 6.82-6.76 (m, 1H), 6.26 (d, *J* = 7.6 Hz 1H), 4.62 (br s, 1 NH), 1.34 (s, 3H), 1.27 (s, 3H); **^{13}C NMR (101 MHz, CDCl$_3$)** δ 195.7, 148.5, 147.6, 145.7, 144.4, 140.8, 139.3, 137.7, 134.4, 132.7, 132.6, 131.5, 128.7, 128.7, 128.5, 128.4, 128.0, 127.9, 127.74, 127.66, 123.2, 121.9, 121.4, 120.3, 111.4, 86.5, 49.7, 26.3, 24.6; **HRMS** (ESI, m/z) calcd for $C_{31}H_{25}NOBr$ $[M + H]^+$ 506.1120 found 506.1116.

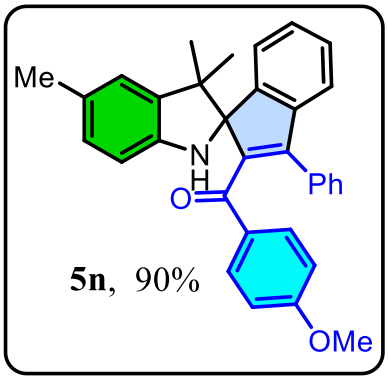
Figure S5. X-ray crystal structure of **5m** (ellipsoid contour at 50% probability level)



Crystal data and structure refinement for 5m (CCDC 2251983)

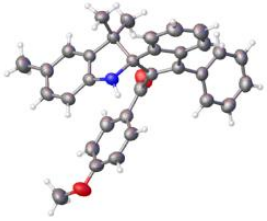
Identification code	GS01_129NEW_0m_a	ρ_{calc}g/cm^3	1.428
Empirical formula	$C_{31}H_{24}BrNO$	μ/mm^{-1}	2.562
Formula weight	506.446	F(000)	1040.1
Temperature/K	100.00	Crystal size/mm^3	0.12 × 0.11 × 0.09
Crystal system	monoclinic	Radiation	Cu Kα (λ = 1.54178)
Space group	$P2_1/n$	2Θ range for data collection/°	8.88 to 133.82
a/Å	10.6361(19)	Index ranges	-12 ≤ h ≤ 12, -22 ≤ k ≤ 22, -13 ≤ l ≤ 13
b/Å	18.871(3)	Reflections collected	36092
c/Å	11.745(2)	Independent reflections	4130 [R_{int} = 0.0771, R_{sigma} = 0.0443]
α/°	90	Data/restraints/parameters	4130/0/310
β/°	91.944(7)	Goodness-of-fit on F^2	1.044
γ/°	90	Final R indexes [I>=2σ (I)]	R_1 = 0.0575, wR_2 = 0.1702
Volume/Å^3	2355.9(7)	Final R indexes [all data]	R_1 = 0.0607, wR_2 = 0.1741
Z	4	Largest diff. peak/hole / e Å^{-3}	1.11/-1.04

(4-methoxyphenyl)(3',3',5'-trimethyl-3-phenylspiro[indene-1,2'-indolin]-2-yl)methanone (5n):



Yield 90% (85 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 154-156 °C; Crystallization was done from hot ethanol; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.57–7.50 (comp, 2H), 7.37–7.33 (comp, 2H), 7.30–7.23 (comp, 2H), 7.27–7.15 (comp, 5H), 6.82 (s, 1H), 6.77 (d, *J* = 7.8 Hz, 1H), 6.69–6.63 (comp, 2H), 6.36 (d, *J* = 7.5 Hz, 1H), 4.63 (br s, 1 NH), 3.77 (s, 3H), 2.30 (s, 3H), 1.31 (s, 3H), 1.24 (s, 3H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 194.9, 163.0, 147.3, 141.6, 140.4, 133.4, 131.3 (X2), 130.8, 128.8 (X2), 128.3 (X2), 128.24, 128.20, 127.7, 127.0, 124.1, 122.6, 121.8, 113.2 (X2), 111.7, 86.7, 55.3, 49.5, 25.7, 25.4, 21.0; **HRMS** (ESI, m/z) calcd for $C_{33}H_{30}NO_2$ $[M + H]^+$ 472.2277 found 472.2271.

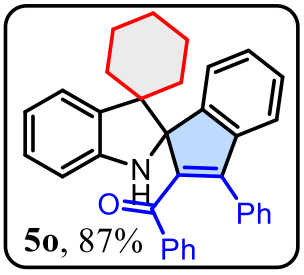
Figure S6. X-ray crystal structure of **5n** (ellipsoid contour at 50% probability level)



Crystal data and structure refinement for 5n (CCDC 2534750)

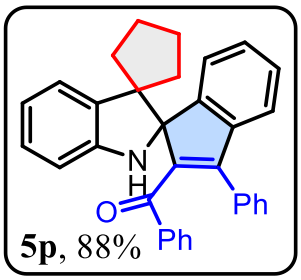
Identification code	IM_06_49_FINAL_a	ρ_{calc}g/cm^3	1.246
Empirical formula	$C_{33}H_{29}NO_2$	μ/mm^{-1}	0.599
Formula weight	471.57	F(000)	500.0
Temperature/K	299.00	Crystal size/mm^3	0.2 × 0.13 × 0.09
Crystal system	triclinic	Radiation	Cu Kα (λ = 1.54178)
Space group	P-1	2Θ range for data collection/°	8.498 to 136.658
a/Å	10.5299(4)	Index ranges	-12 ≤ h ≤ 12, -14 ≤ k ≤ 14, -14 ≤ l ≤ 13
b/Å	11.7051(5)	Reflections collected	45352
c/Å	12.2331(6)	Independent reflections	4576 [R_{int} = 0.0422, R_{sigma} = 0.0273]
α/°	110.007(2)	Data/restraints/parameters	4576/0/330
β/°	108.936(2)	Goodness-of-fit on F^2	1.062
γ/°	101.341(2)	Final R indexes [I>=2σ (I)]	R_1 = 0.0436, wR_2 = 0.1159
Volume/Å^3	1257.25(10)	Final R indexes [all data]	R_1 = 0.0468, wR_2 = 0.1172
Z	2	Largest diff. peak/hole / e Å^{-3}	0.24/-0.17

phenyl(3''-phenyldispiro[cyclohexane-1,3'-indoline-2',1''-inden]-2''-yl)methanone (5o):



Yield 87% (82 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 176-178 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.51–7.45 (comp, 2H), 7.40 (d, *J* = 7.3 Hz, 1H), 7.38–7.29 (comp, 6H), 7.28–7.21 (comp, 4H), 7.21–7.16 (comp, 2H), 6.89 – 6.83 (m, 1H), 6.79–6.74 (m, 1H), 6.19 (d, *J* = 7.4 Hz, 1H), 4.52 (br s, 1 NH), 2.49–2.45 (m, 1H), 1.94-1.90 (m, 1H), 1.78–1.49 (m, 6H), 1.37–1.23 (m, 2H), 1.09–0.99 (m, 1H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 196.0, 146.1, 142.2, 138.2, 138.0, 133.1, 132.4, 128.8, 128.7, 128.4, 128.3 (X2), 127.9(X2), 127.3, 126.8, 125.7, 125.3, 121.9, 119.1, 111.2, 88.1, 52.8, 33.7, 32.6, 25.7, 22.8, 22.6; **HRMS** (ESI, m/z) calcd for $C_{34}H_{30}NO$ $[M + H]^+$ 468.2327 found 468.2313.

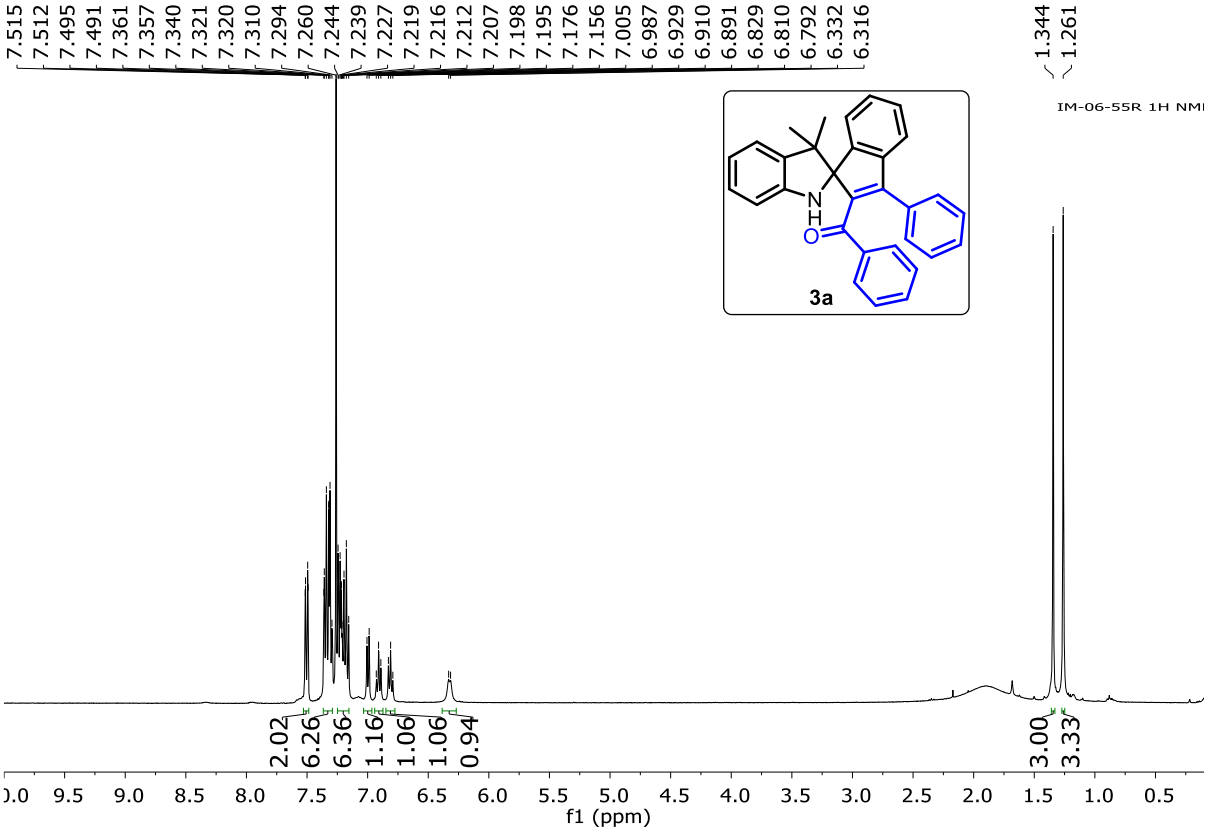
phenyl(3''-phenyldispiro[cyclopentane-1,3'-indoline-2',1''-inden]-2''-yl)methanone (5p):



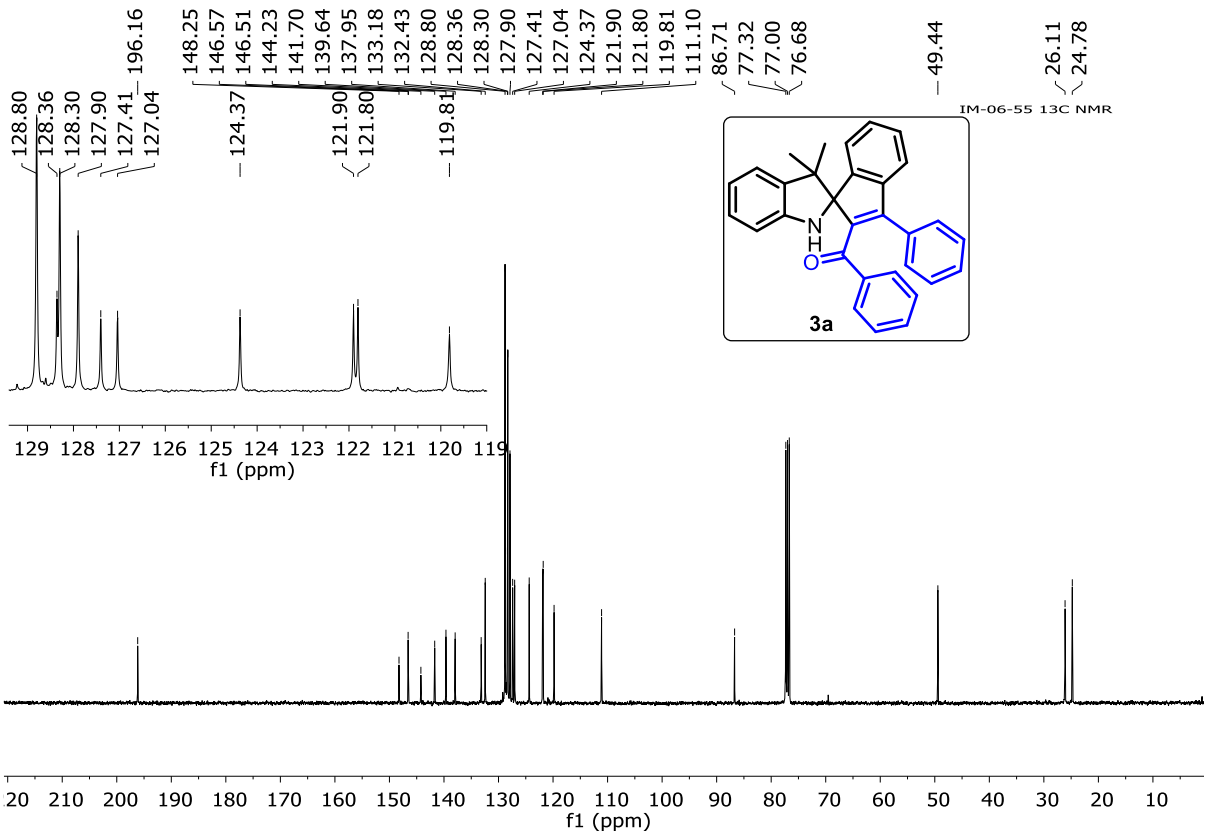
Yield 88% (80 mg); Yellow solid; **eluent composition** petroleum ether/ethyl acetate = 19:1; **mp** 172-174 °C; **^{1}H NMR (400 MHz, $CDCl_3$)** δ 7.50 (d, *J* = 7.3 Hz, 1H), 7.44–7.32 (comp, 7H), 7.32–7.22 (comp, 4H), 7.21–7.14 (comp, 2H), 6.98–6.93 (m, 1H), 6.86 – 6.78 (m, 1H), 6.76 – 6.70 (m, 1H), 6.13 (d, *J* = 7.6 Hz, 1H), 4.43 (br s, 1 NH), 2.19–2.13 (m, 1H), 2.02–1.91 (m, 1H), 1.83-1.63 (m, 4H), 1.62–1.42 (m, 2H); **^{13}C NMR (101 MHz, $CDCl_3$)** δ 195.9, 148.2, 145.8, 145.7, 145.3, 142.3, 139.6, 138.3, 133.1, 132.3, 128.7, 128.5, 128.4, 128.26, 128.34, 127.9, 127.4, 126.9, 125.2, 122.7, 121.8, 119.5, 110.6, 86.5, 59.6, 37.7, 33.5, 24.1, 23.6; **HRMS** (ESI, m/z) calcd for $C_{33}H_{28}NO$ $[M + H]^+$ 454.2171 found 454.2159.

^{1}H, ^{13}C and ^{19}F NMR spectra of spiro-products:

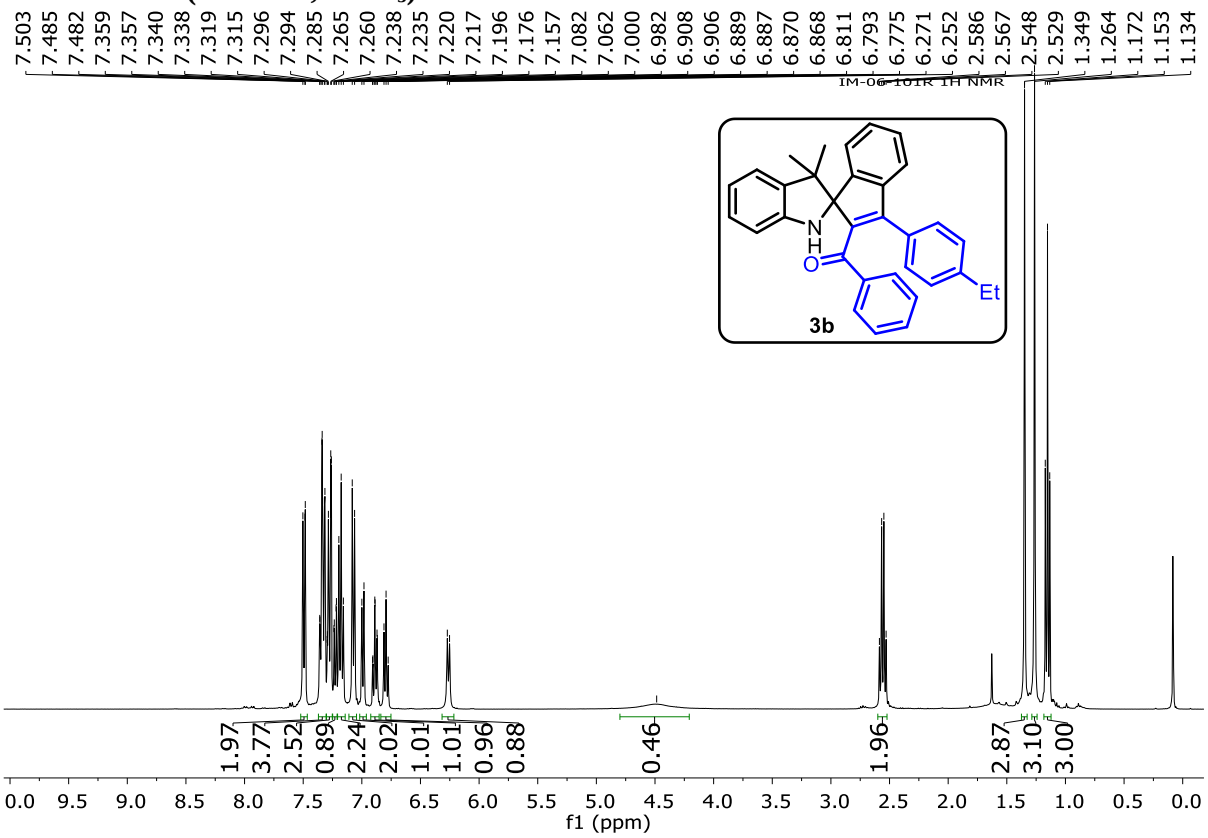
^{1}H NMR of 3a (400 MHz, $CDCl_3$):



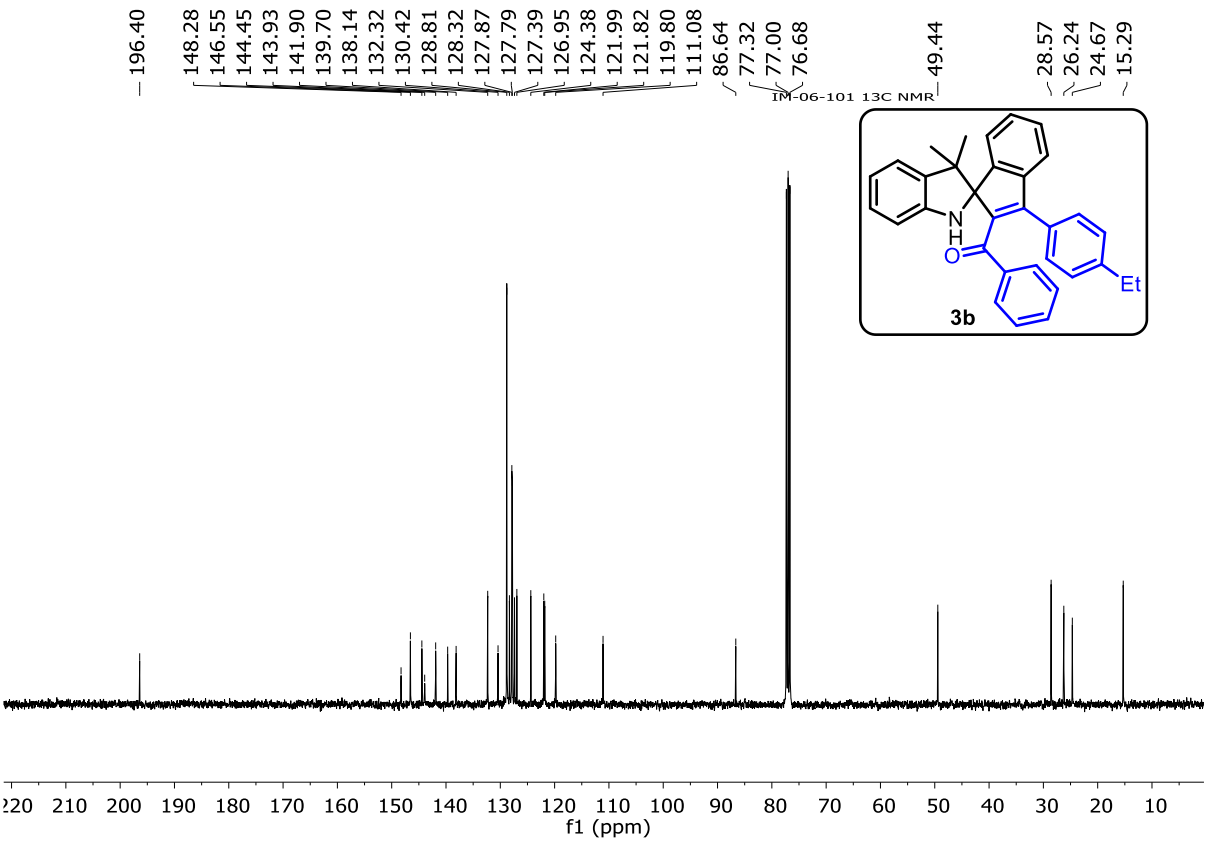
^{13}C{^{1}H} NMR of 3a (100 MHz, $CDCl_3$):



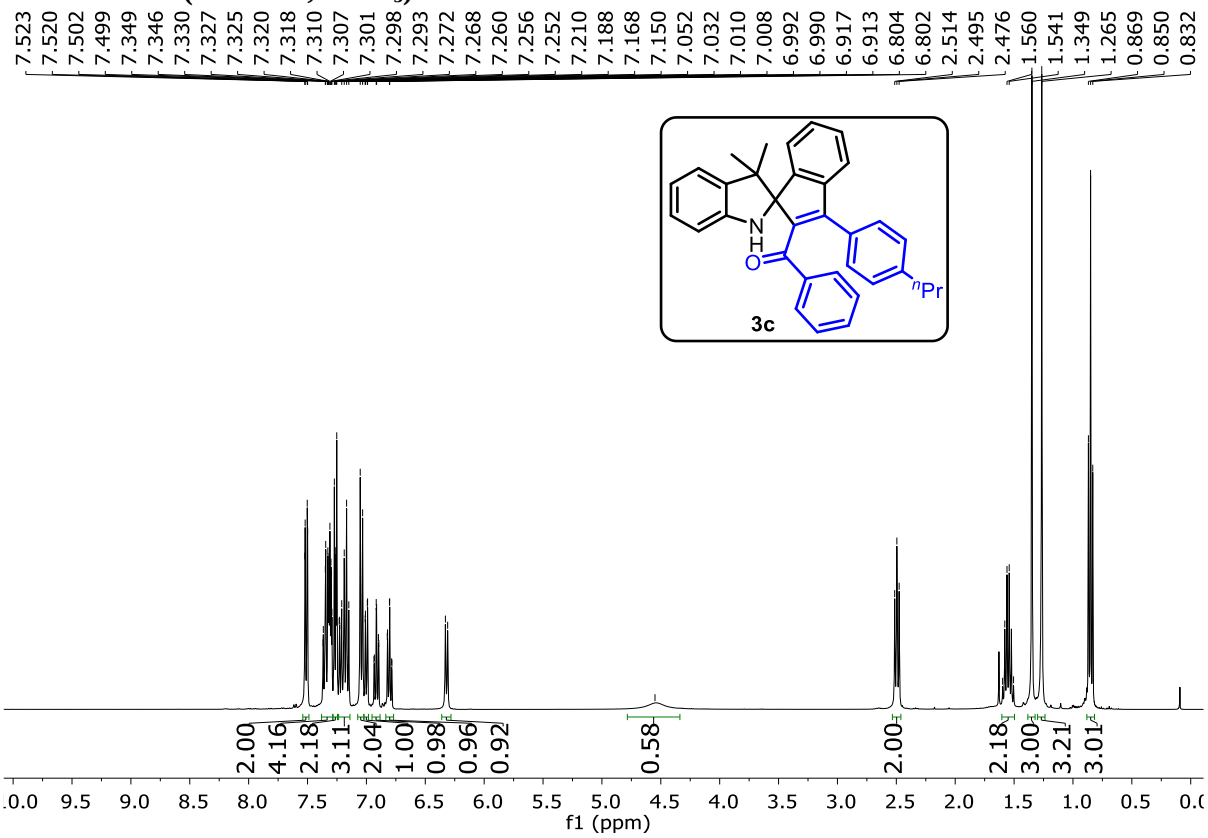
^{1}H NMR of 3b (400 MHz, $CDCl_3$):



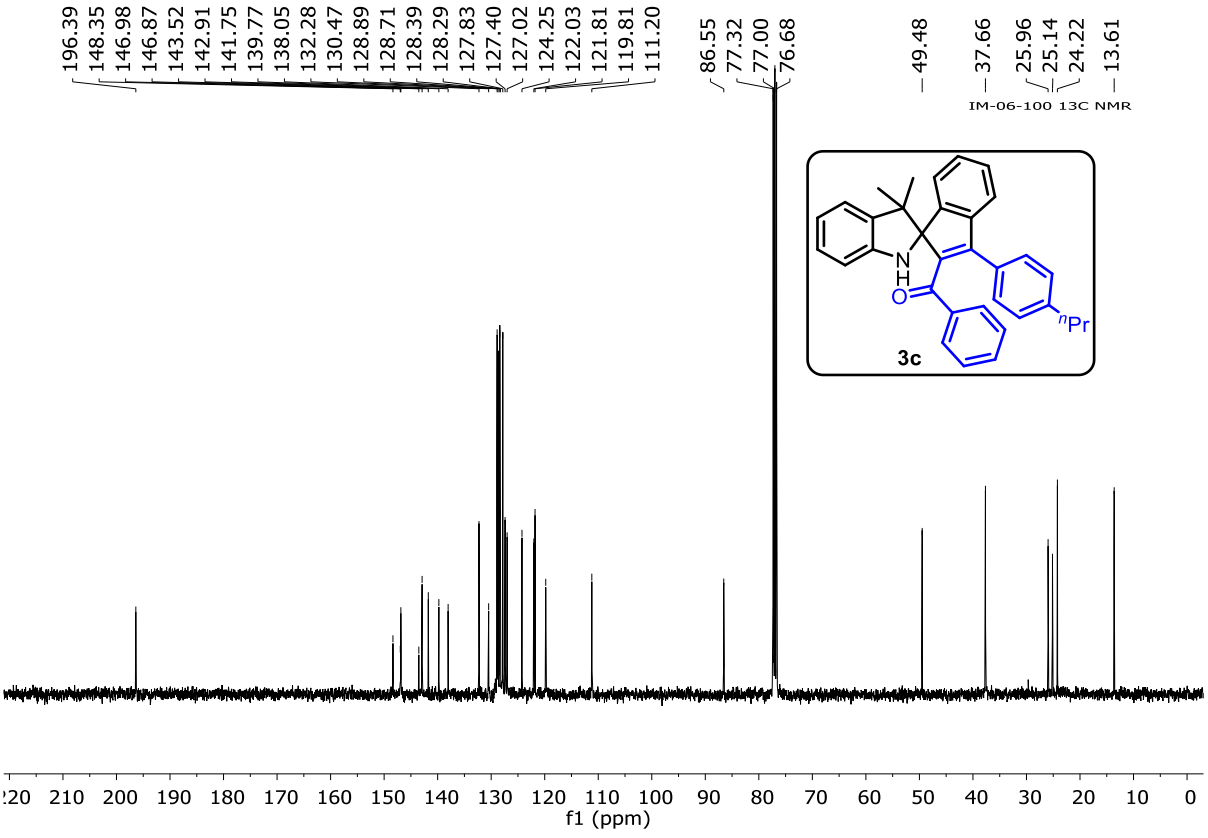
^{13}C{^{1}H} NMR of 3b (101 MHz, $CDCl_3$):



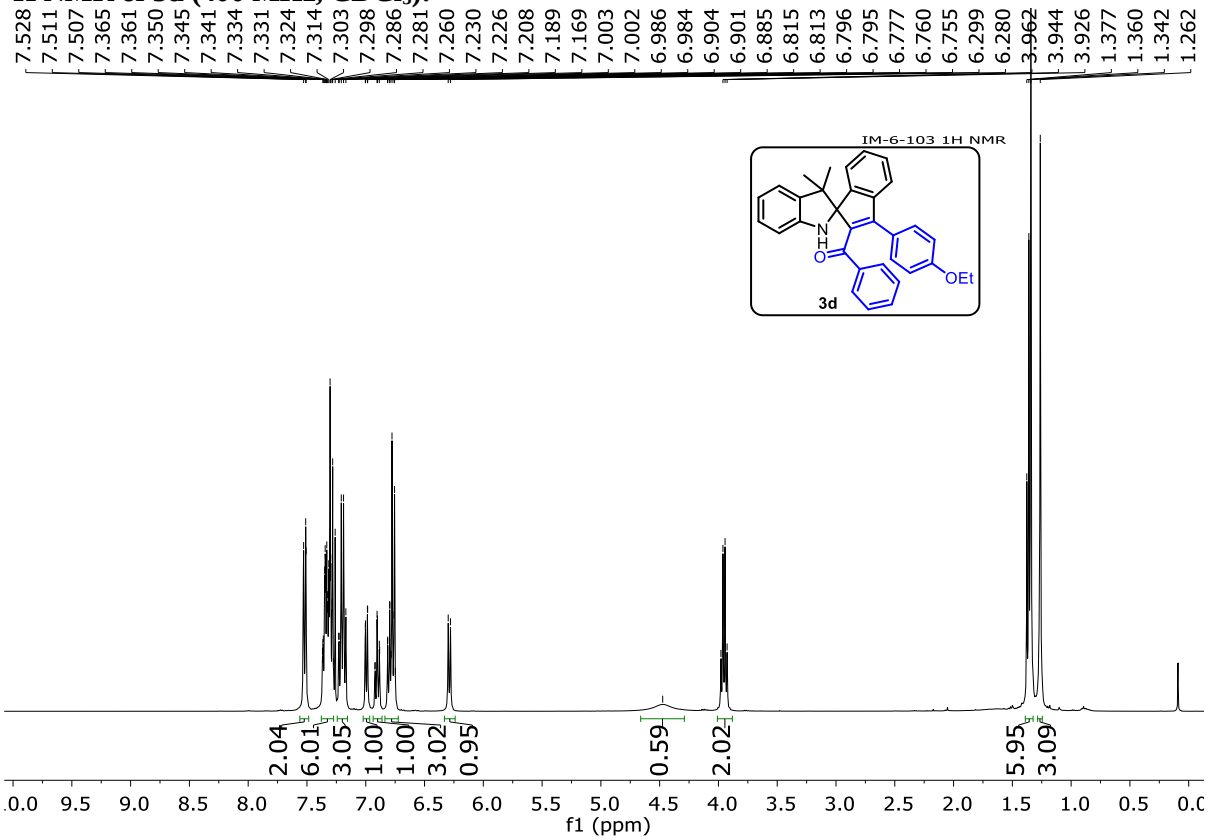
^{1}H NMR of 3c (400 MHz, $CDCl_3$):



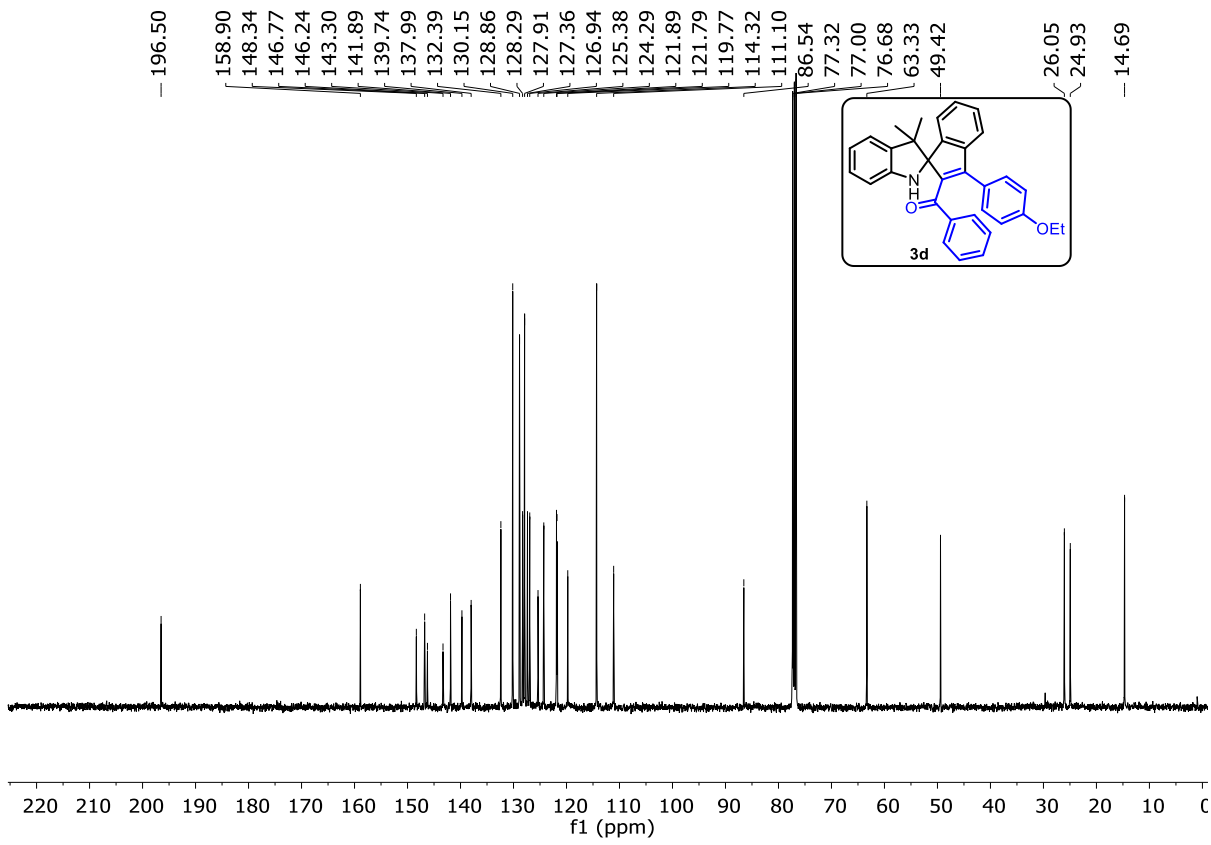
^{13}C{^{1}H} NMR of 3c (101 MHz, $CDCl_3$):



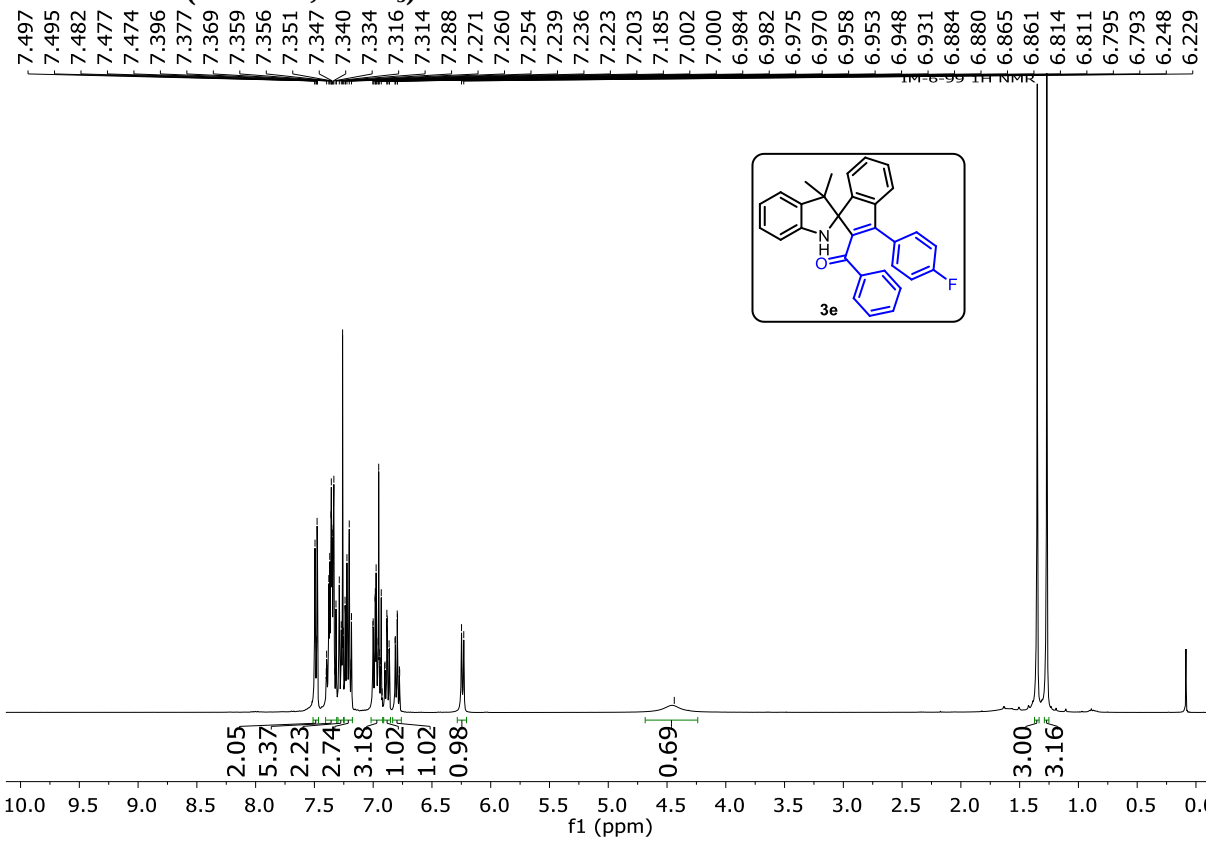
^{1}H NMR of 3d (400 MHz, $CDCl_3$):



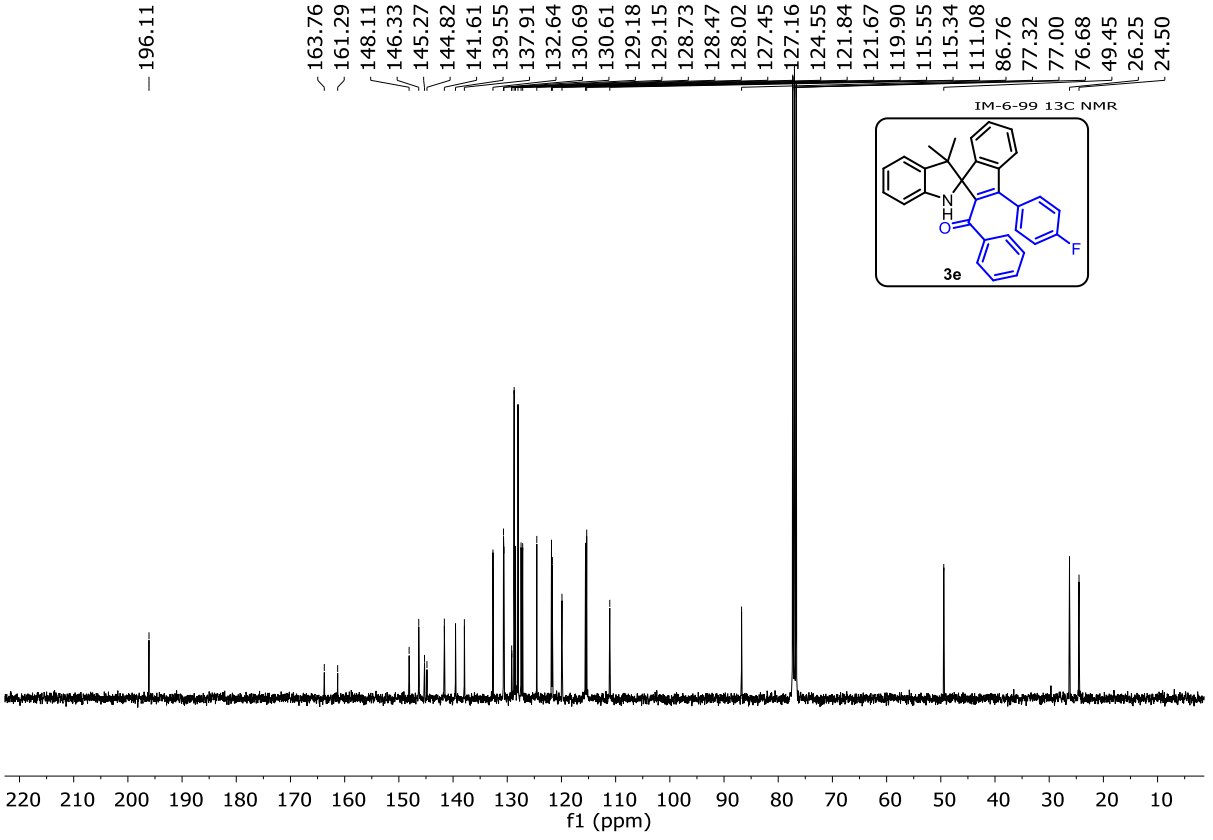
^{13}C{^{1}H} NMR of 3d (101 MHz, $CDCl_3$):



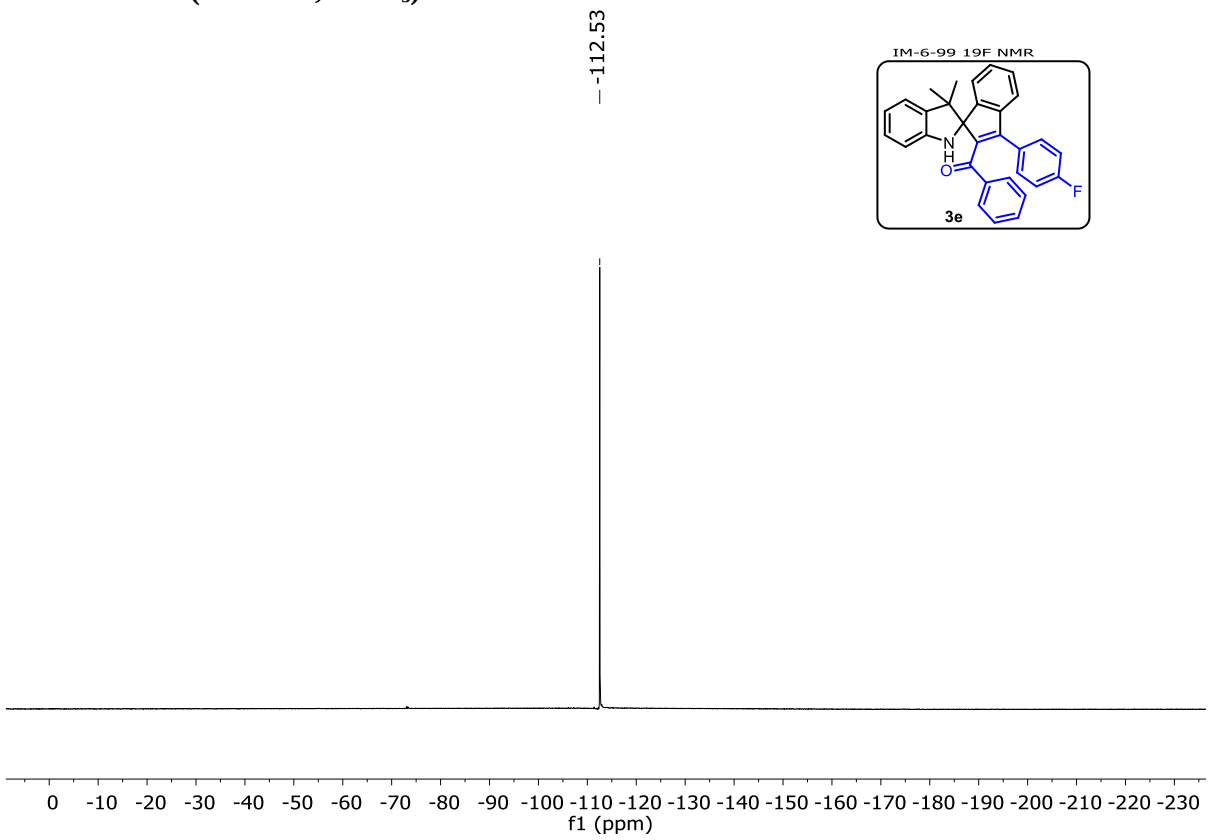
^{1}H NMR of 3e (400 MHz, $CDCl_3$):



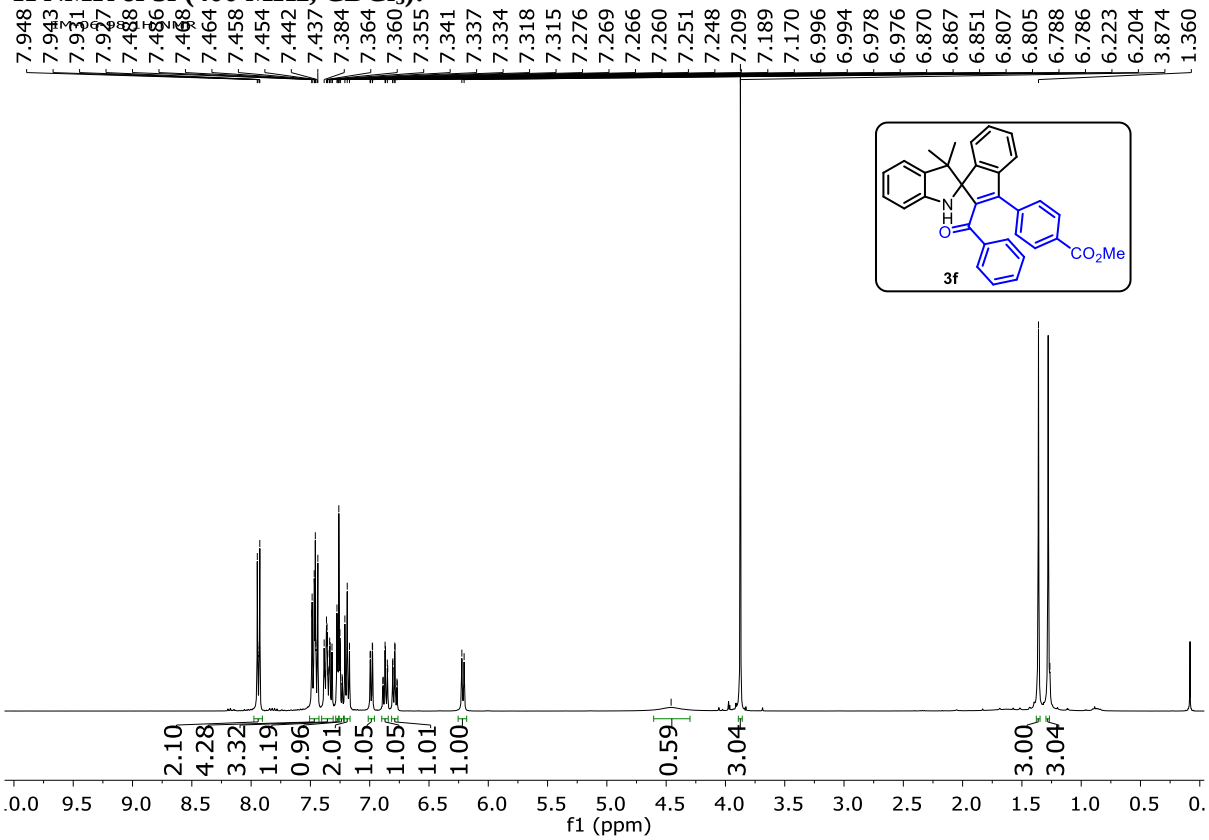
^{13}C{^{1}H} NMR of 3e (101 MHz, $CDCl_3$):



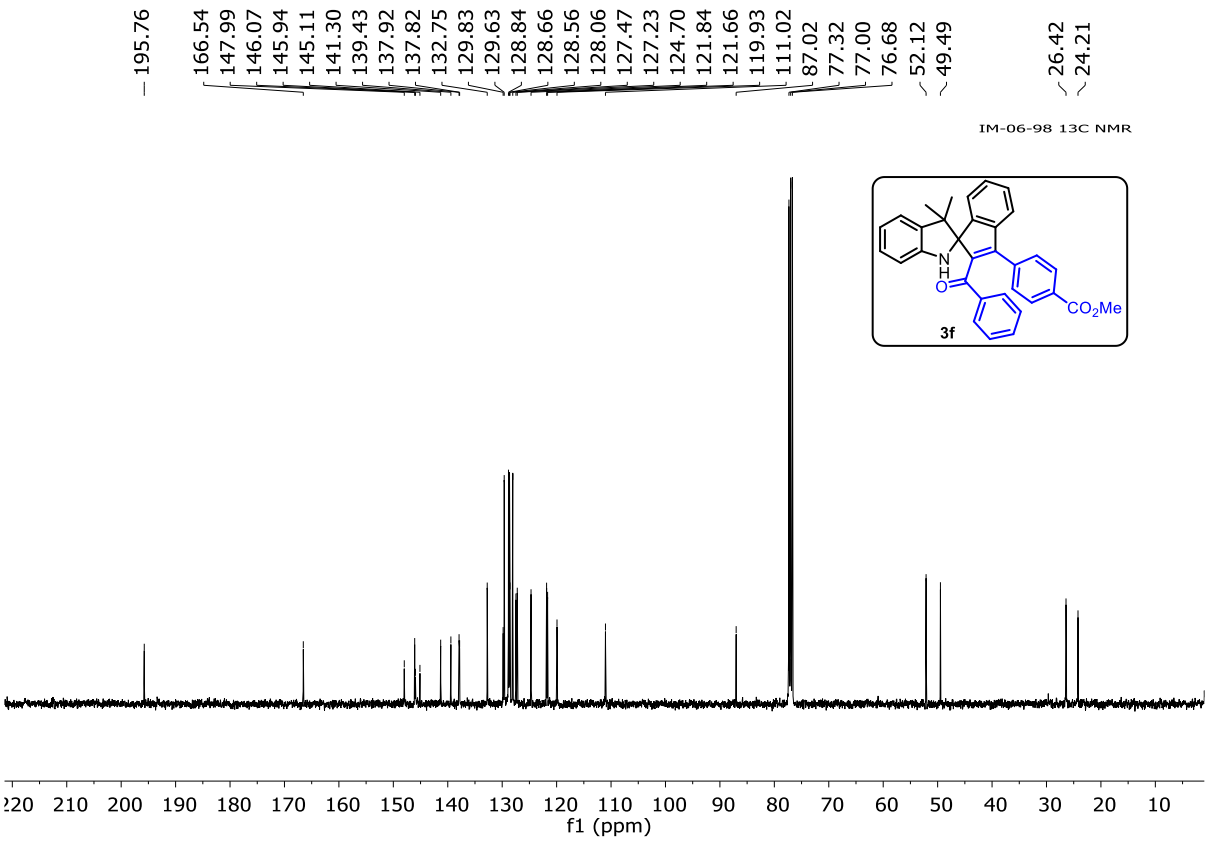
^{19}F NMR of 3e (376 MHz, $CDCl_3$):



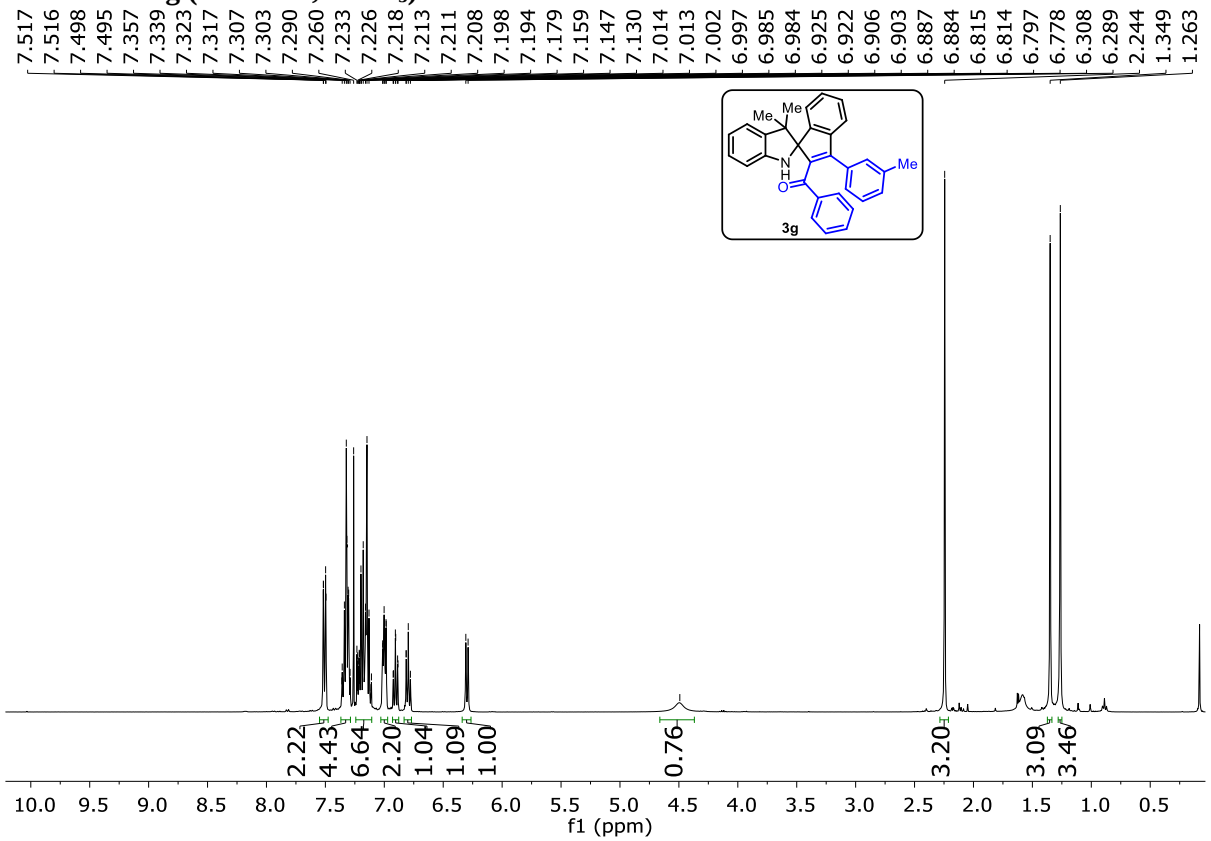
^{1}H NMR of 3f (400 MHz, $CDCl_3$):



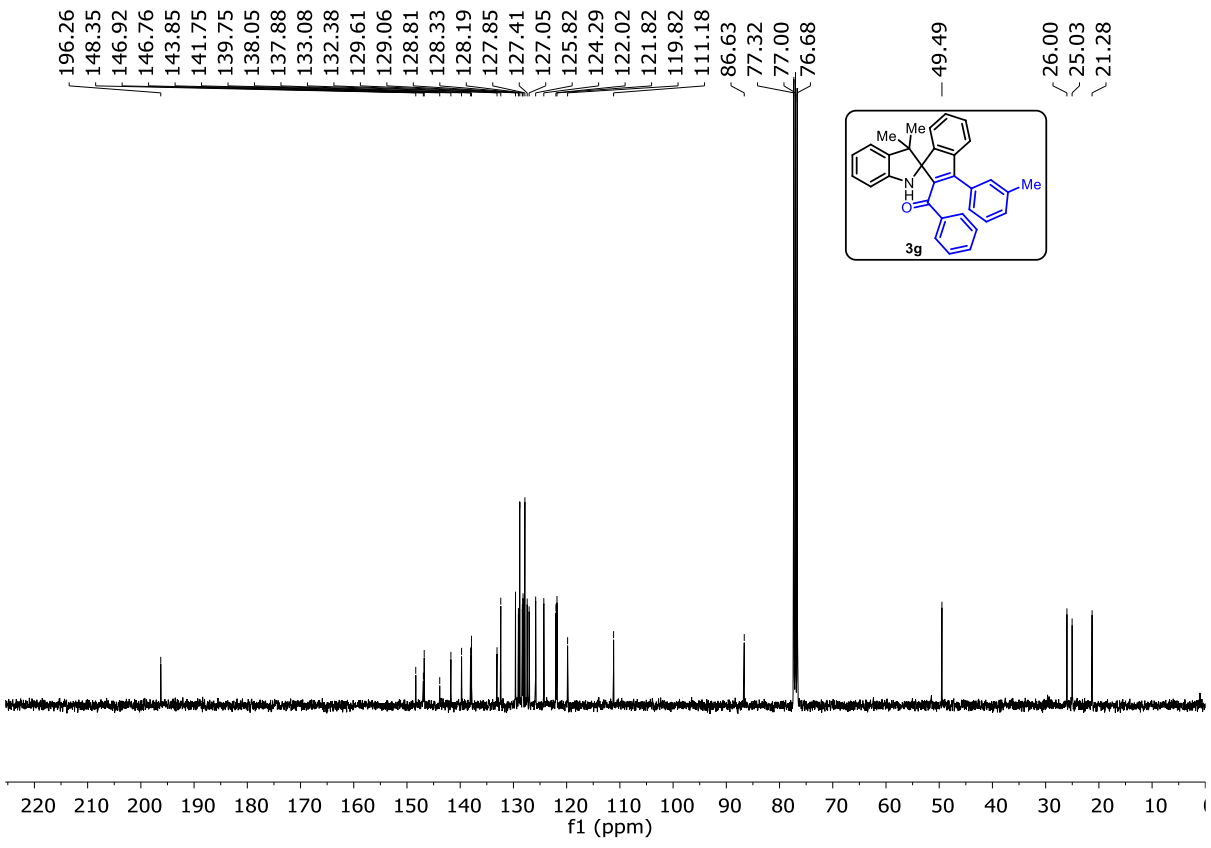
^{13}C{^{1}H} NMR of 3f (101 MHz, $CDCl_3$):



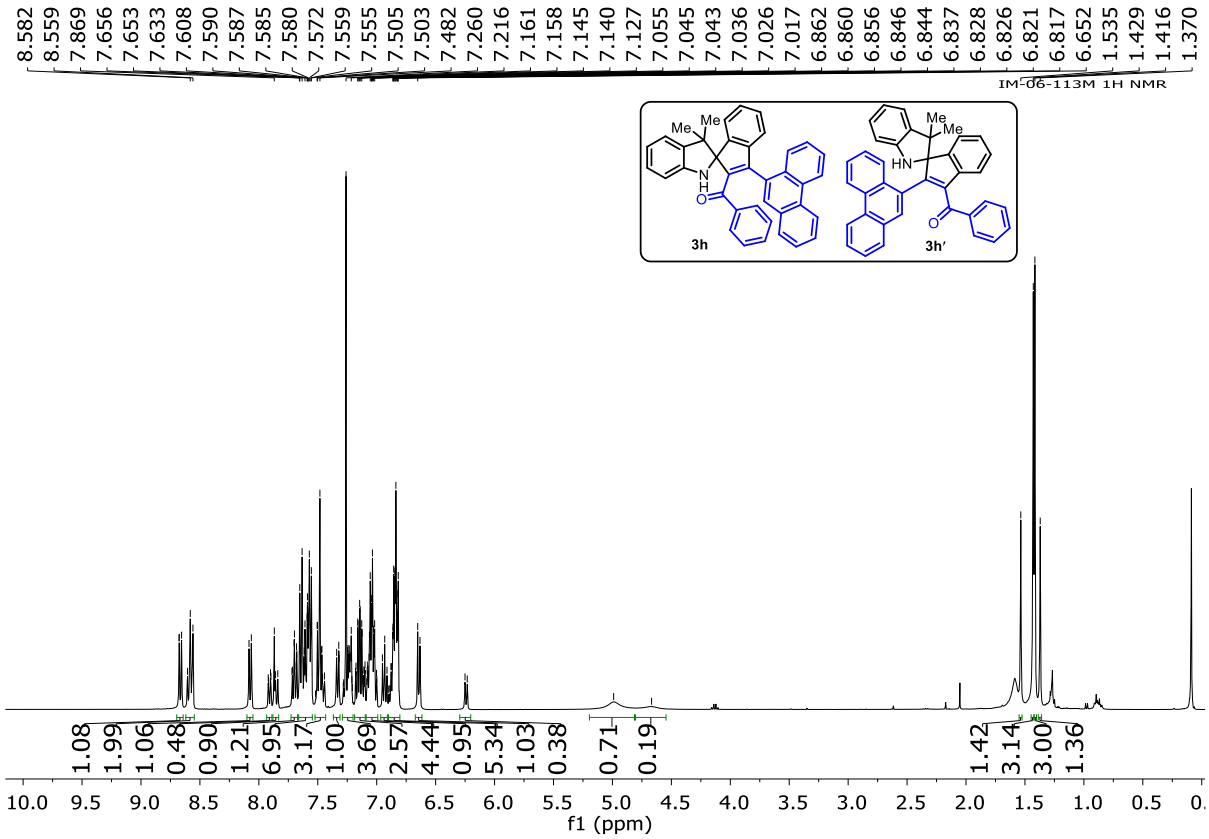
^{1}H NMR of 3g (400 MHz, $CDCl_3$):



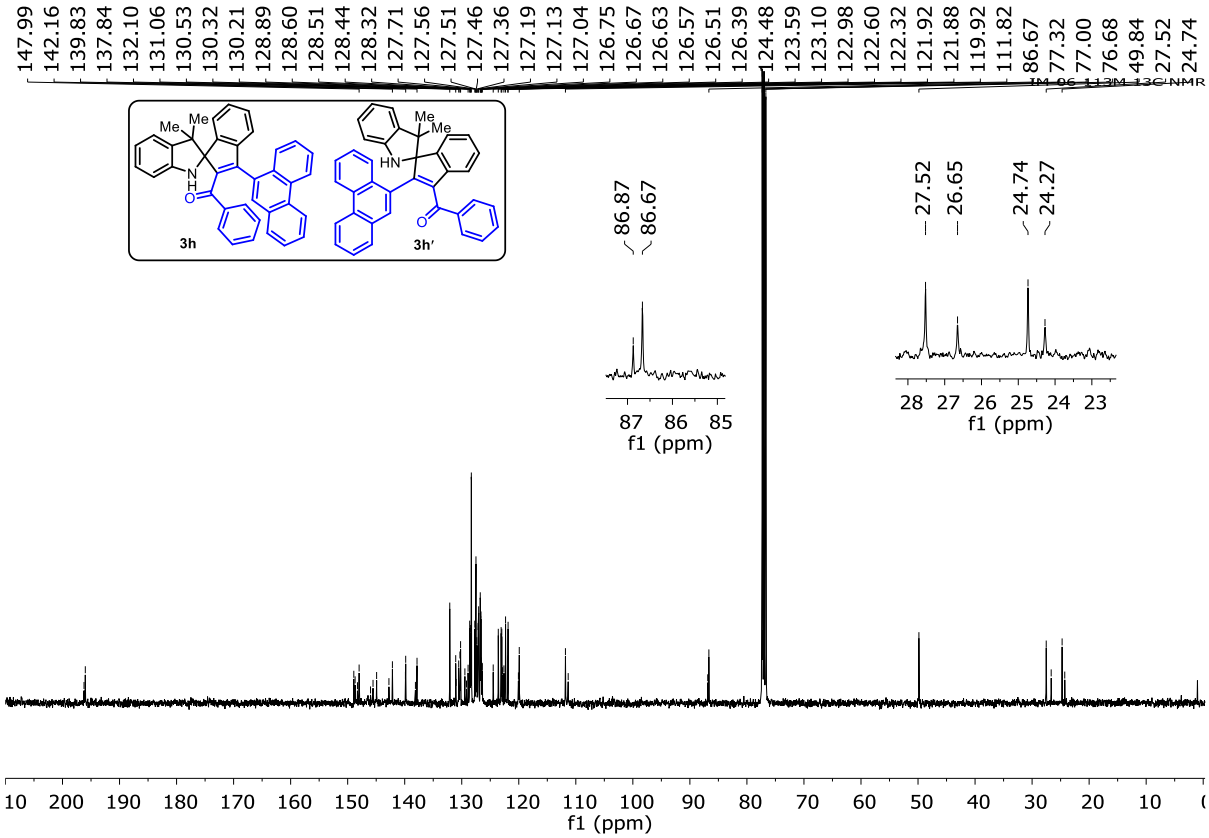
^{13}C{^{1}H} NMR of 3g (101 MHz, $CDCl_3$):



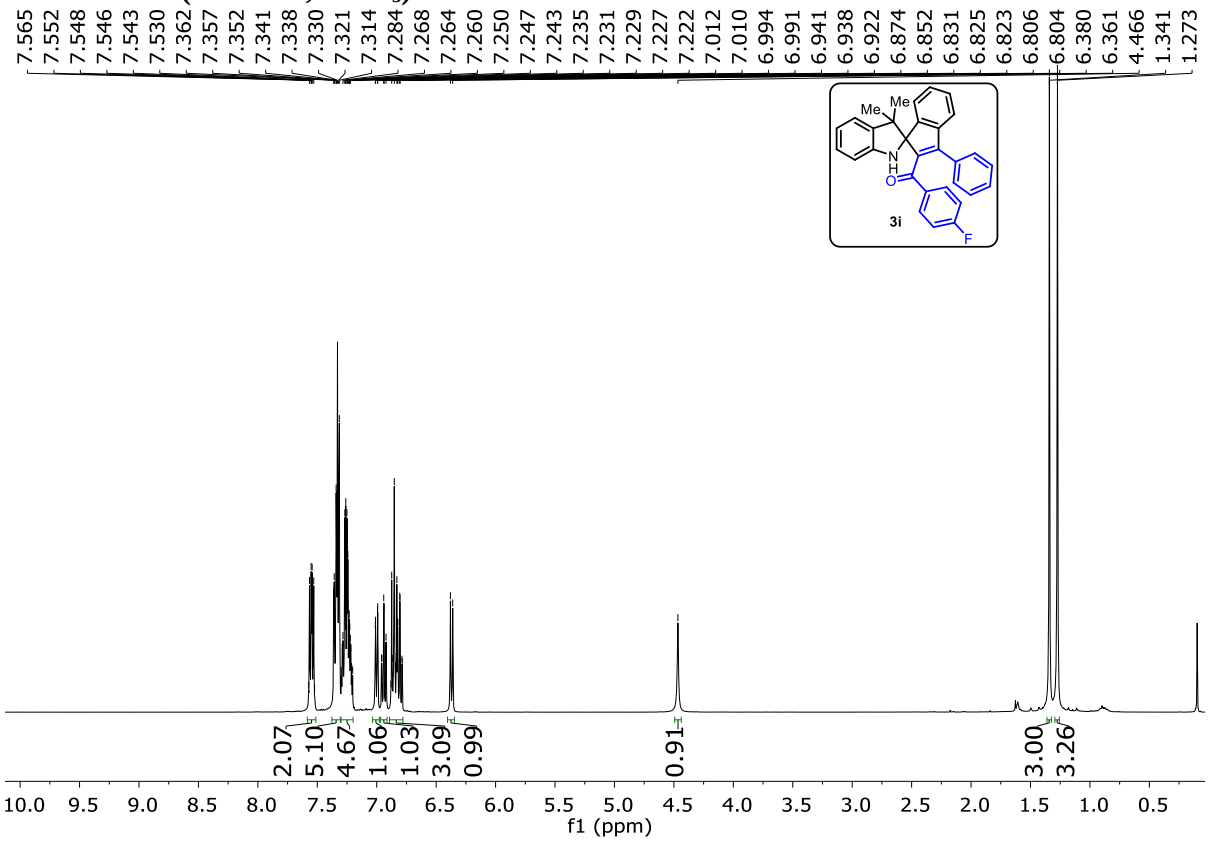
^{1}H NMR of 3h and 3h' (400 MHz, $CDCl_3$):



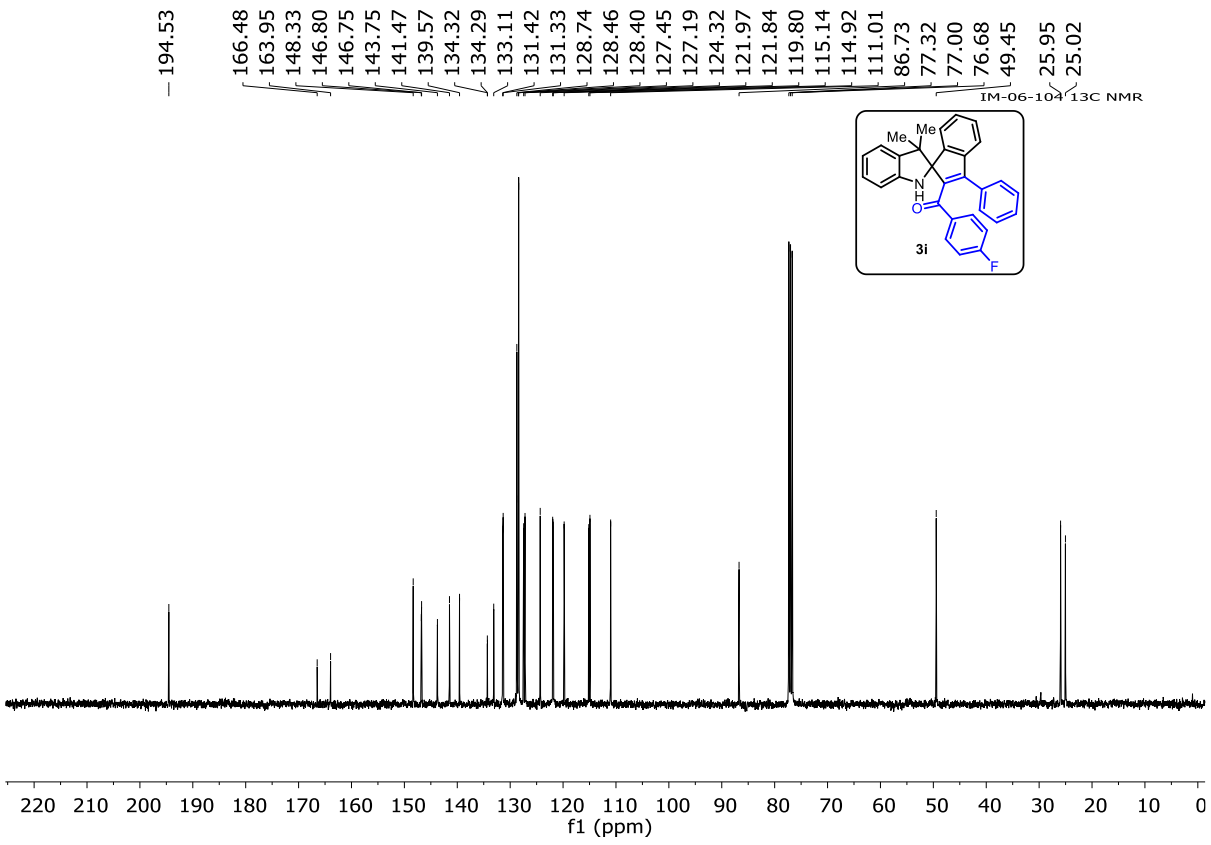
^{13}C{^{1}H} NMR of 3h and 3h' (101 MHz, $CDCl_3$):



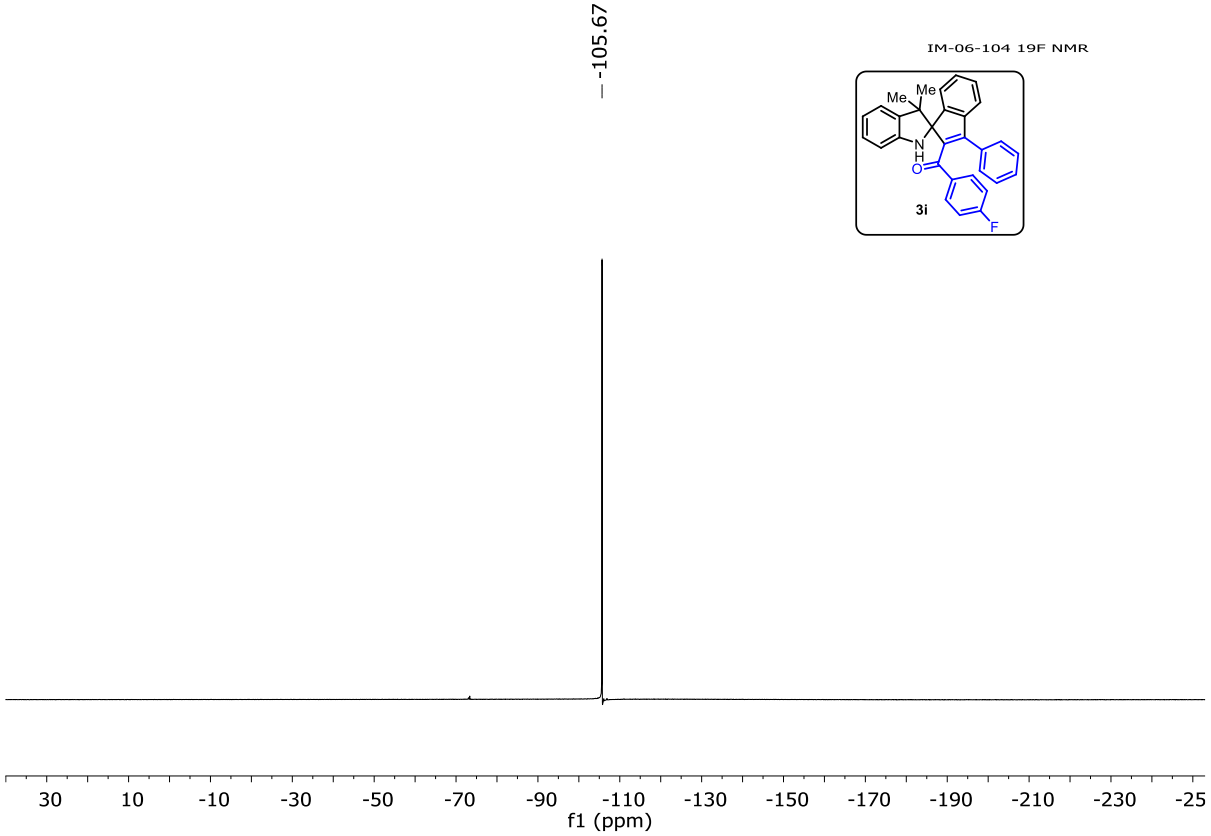
^{1}H NMR of 3i (400 MHz, $CDCl_3$):



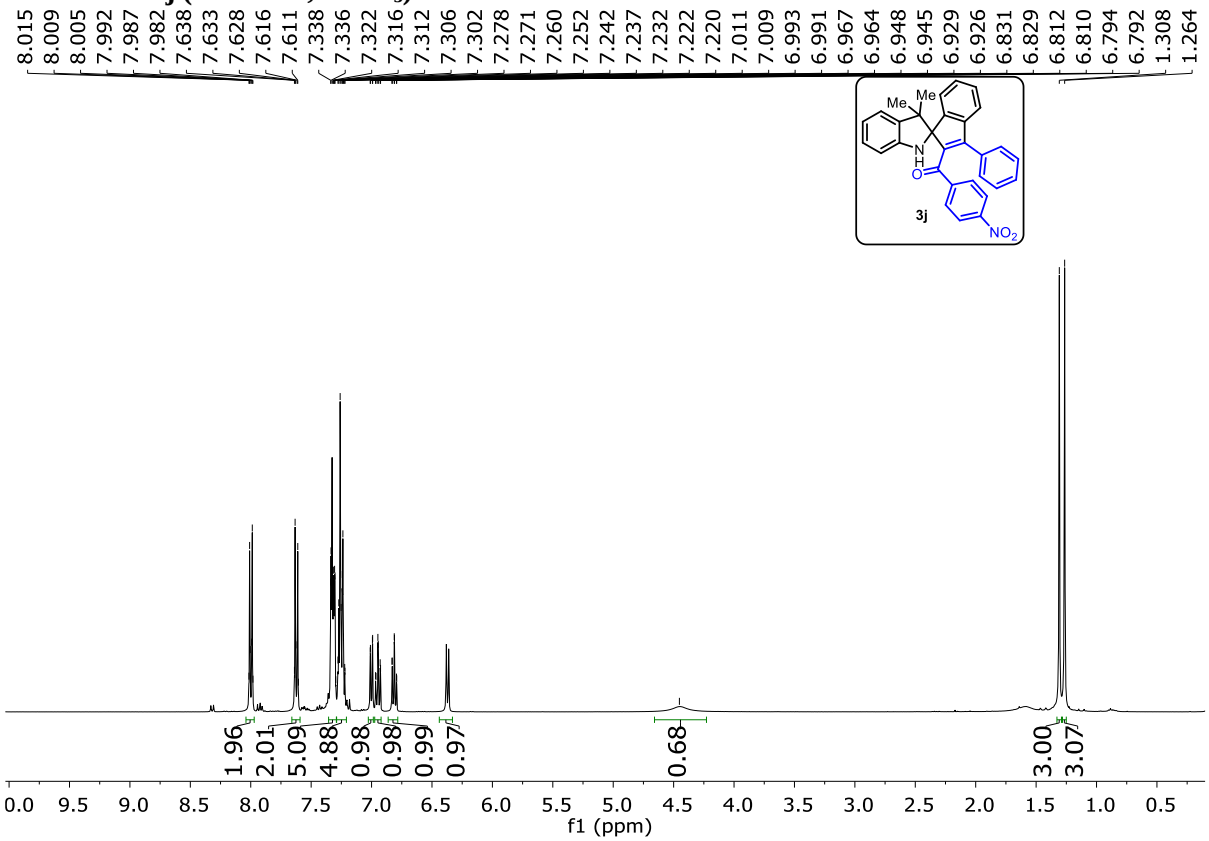
^{13}C{^{1}H} NMR of 3i (101 MHz, $CDCl_3$):



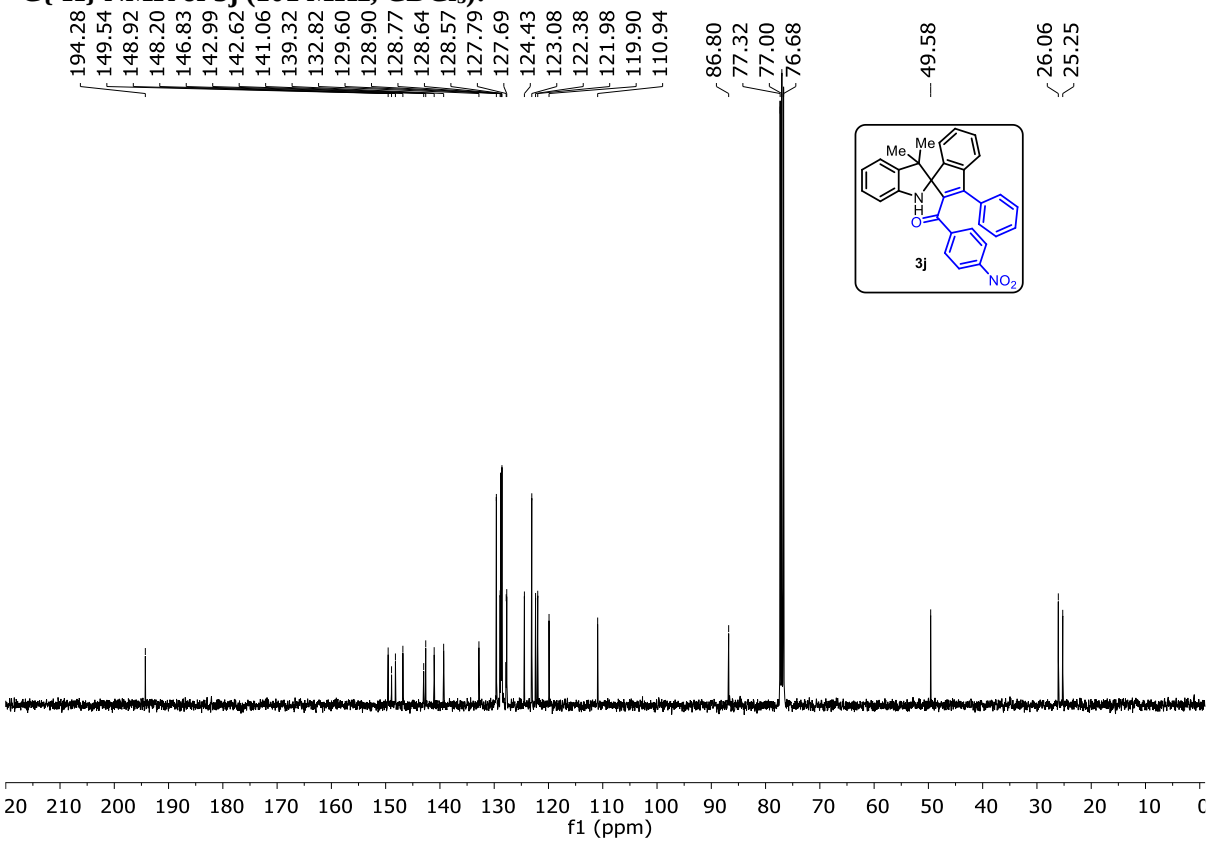
^{19}F NMR of 3i (376 MHz, $CDCl_3$):



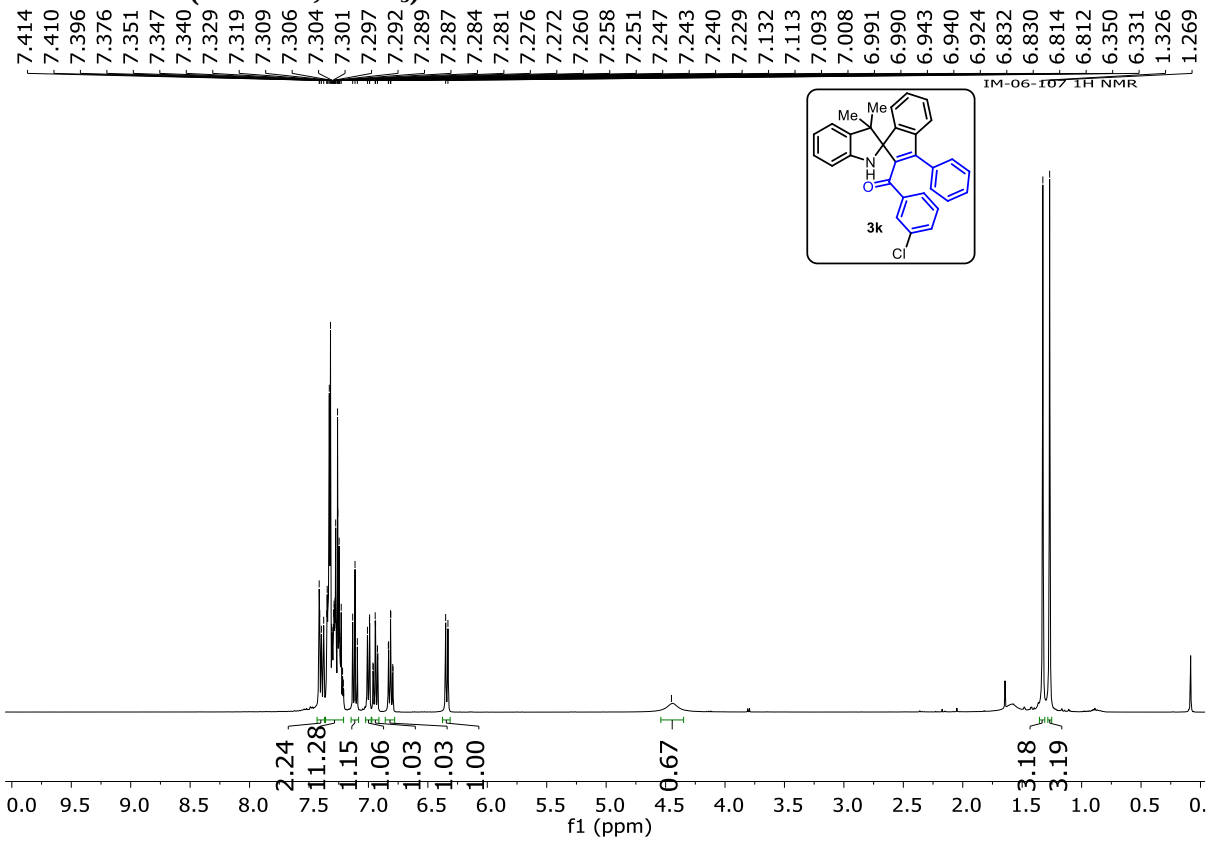
^{1}H NMR of 3j (400 MHz, $CDCl_3$):



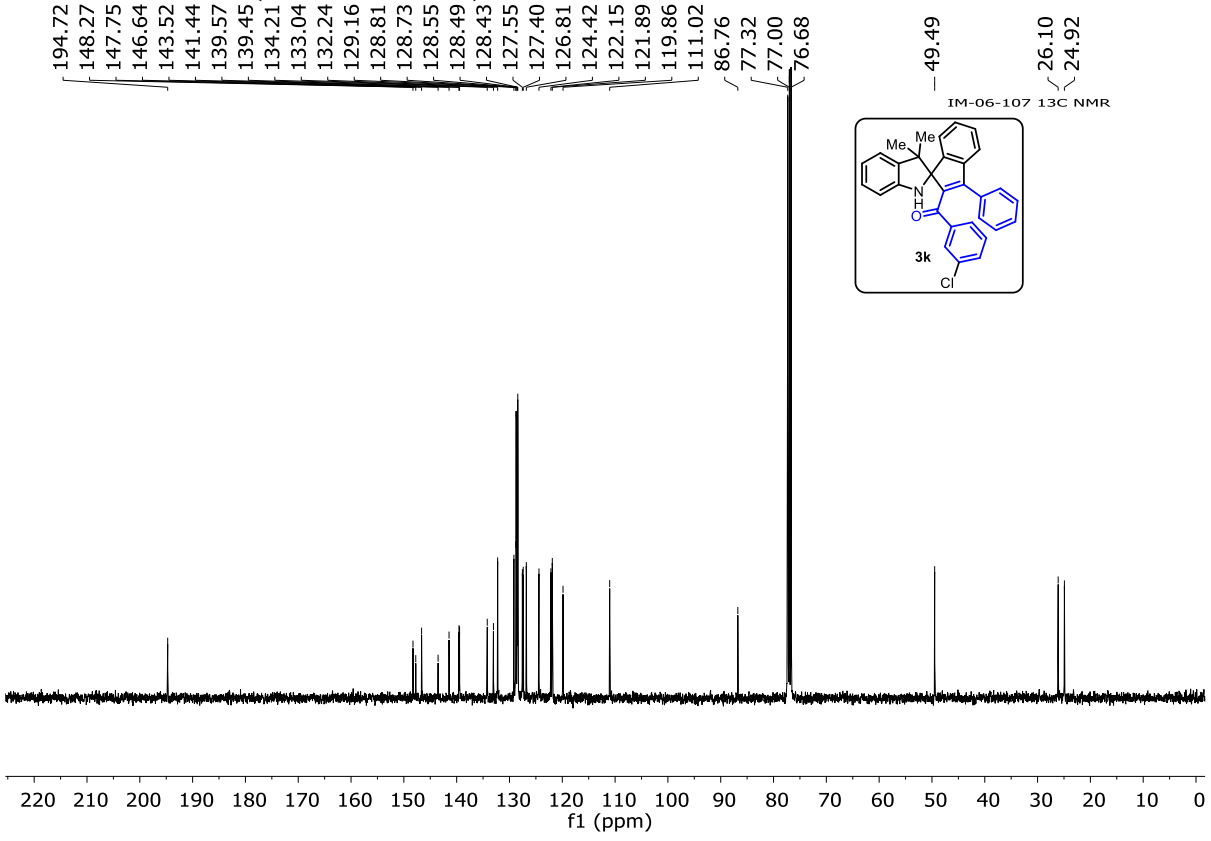
^{13}C{^{1}H} NMR of 3j (101 MHz, $CDCl_3$):



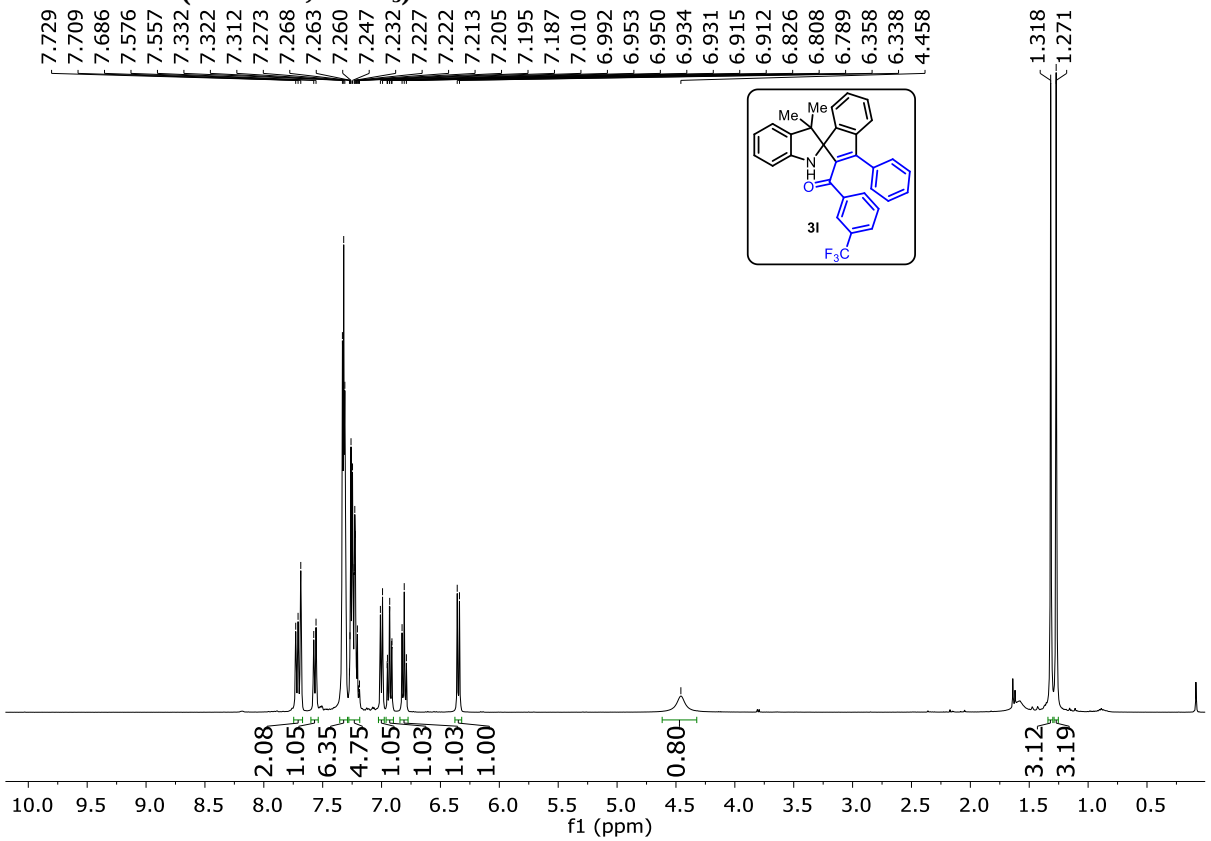
^{1}H NMR of 3k (400 MHz, $CDCl_3$):



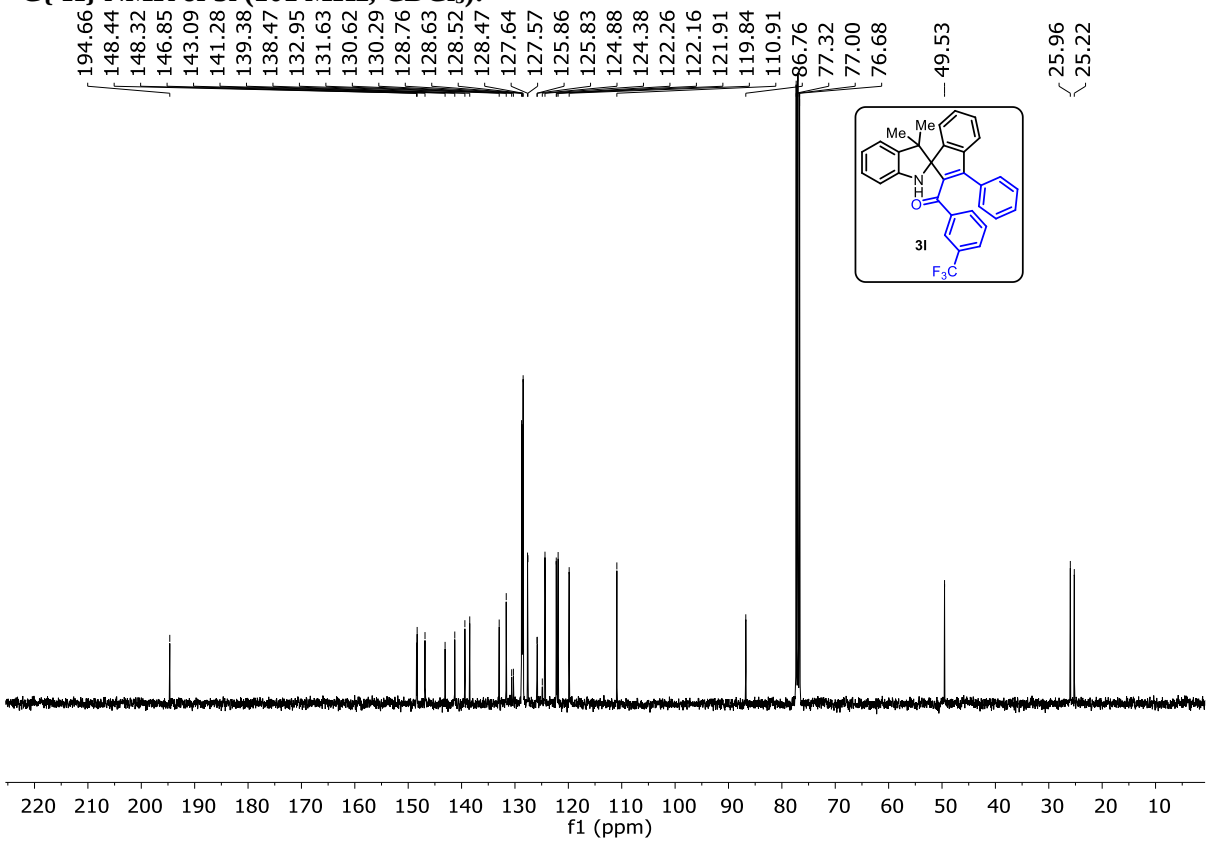
^{13}C{^{1}H} NMR of 3k (101 MHz, $CDCl_3$):



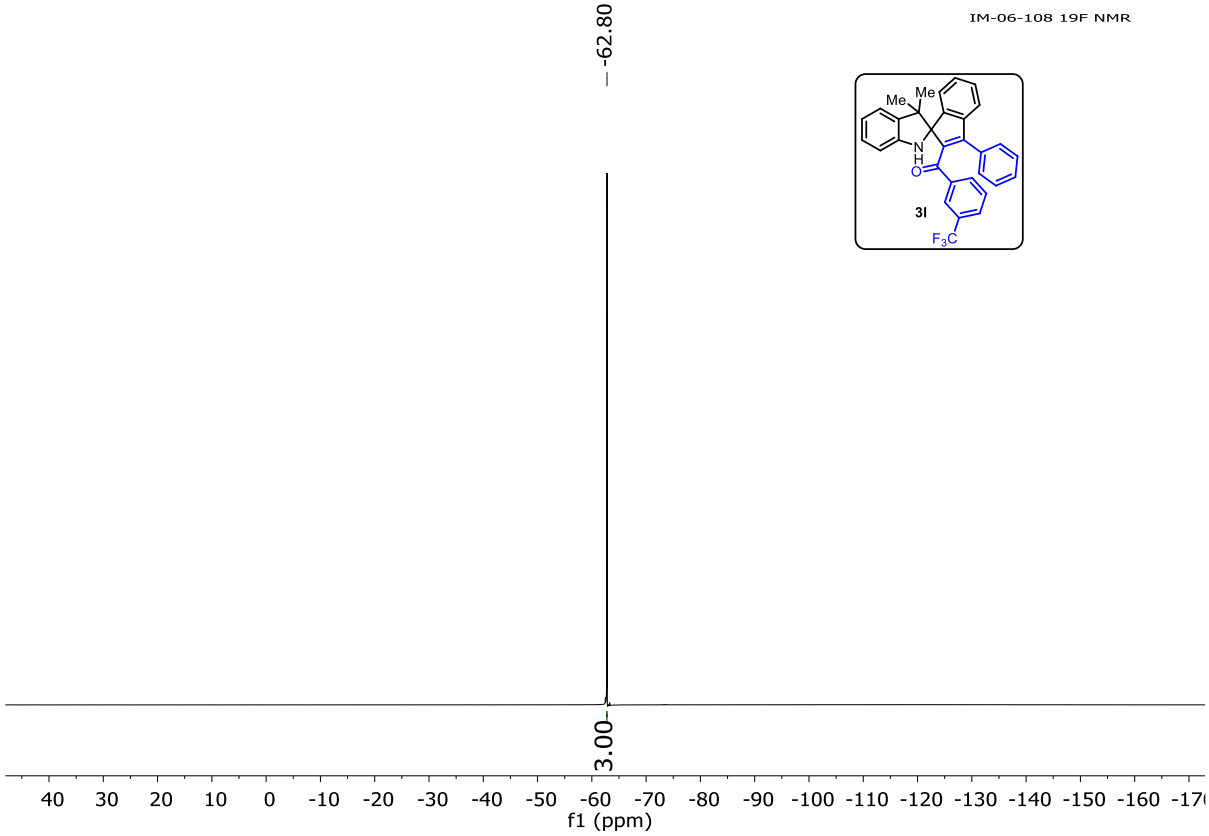
^{1}H NMR of 3l (400 MHz, $CDCl_3$):



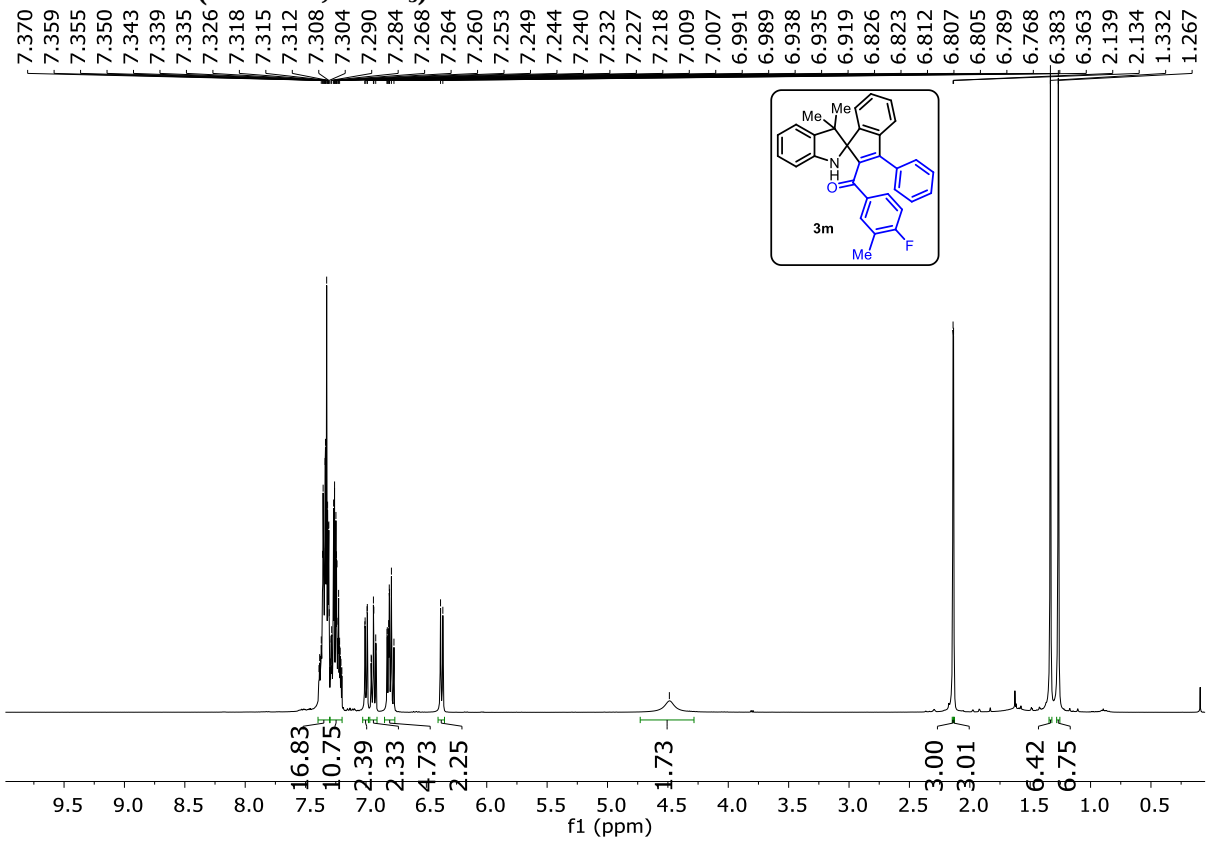
^{13}C{^{1}H} NMR of 3l (101 MHz, $CDCl_3$):



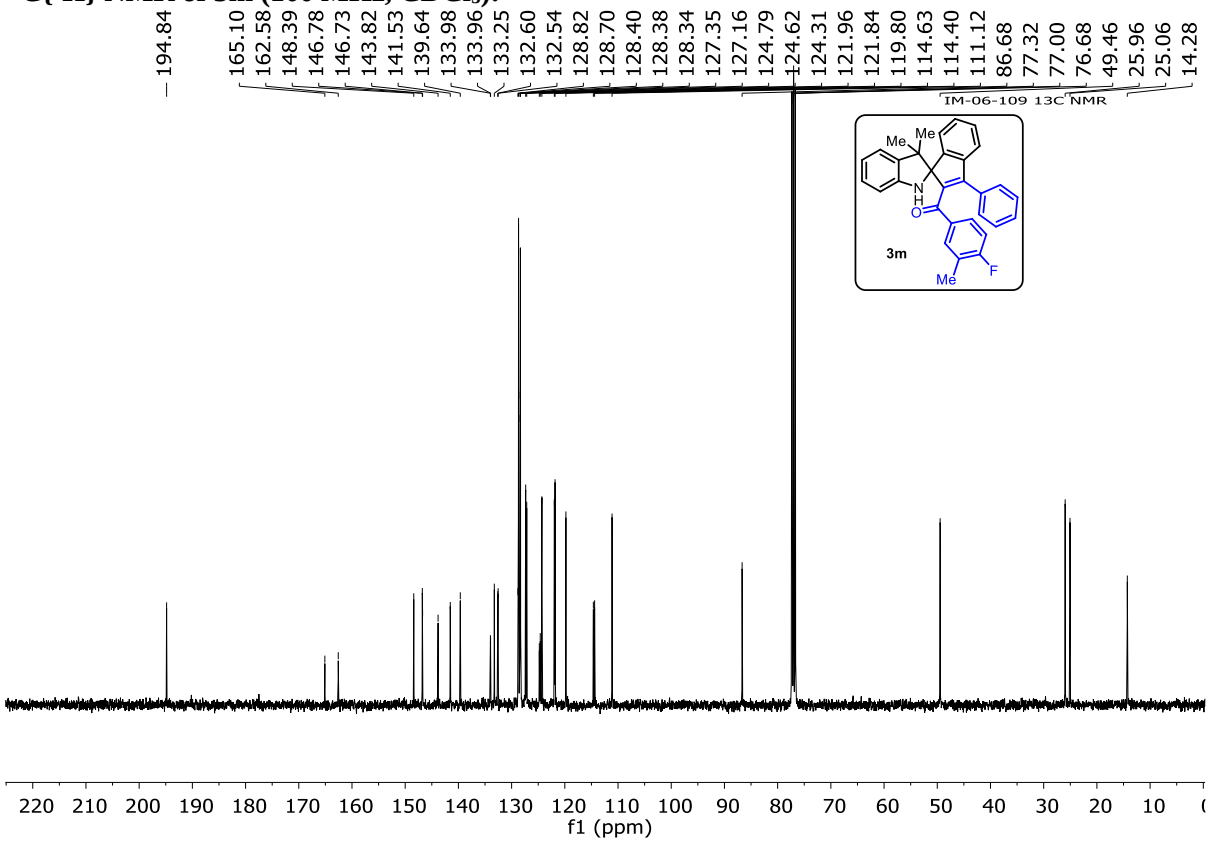
^{19}F NMR of 3l (376 MHz, $CDCl_3$):



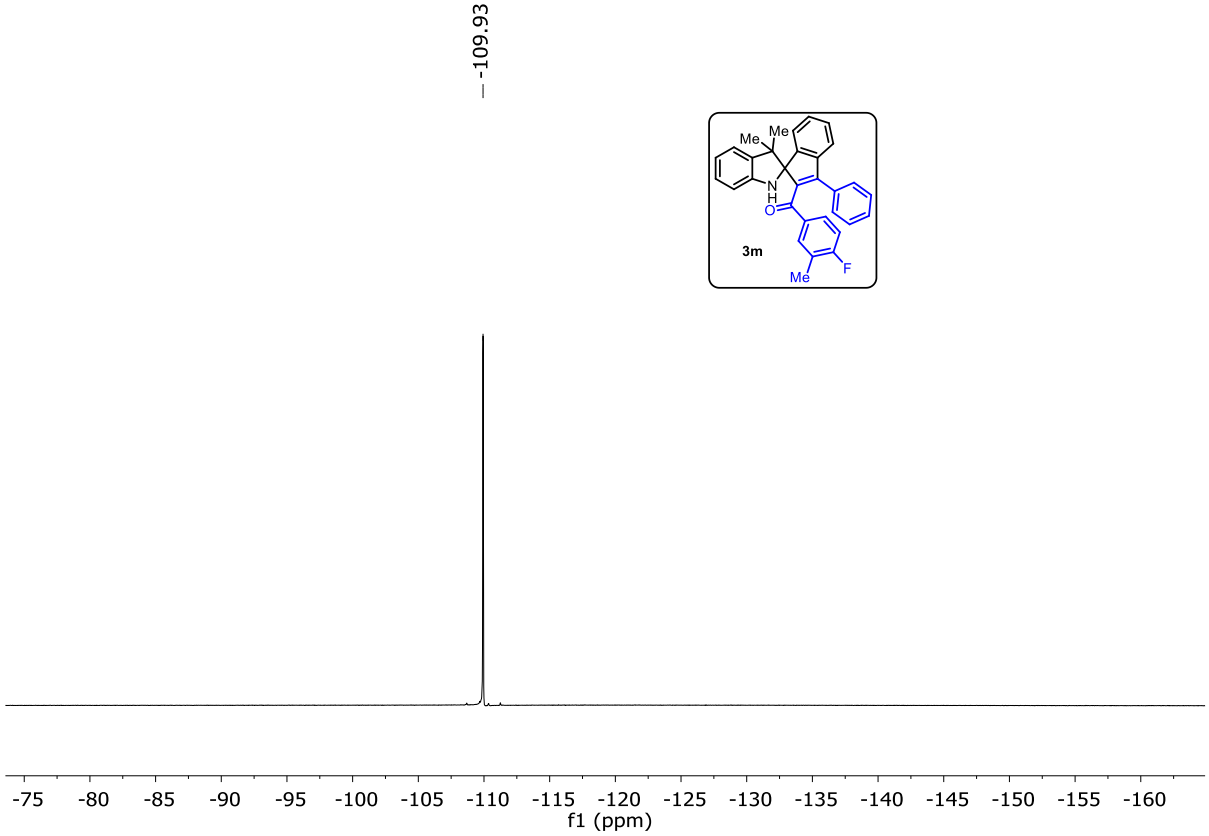
^{1}H NMR of 3m (400 MHz, $CDCl_3$):



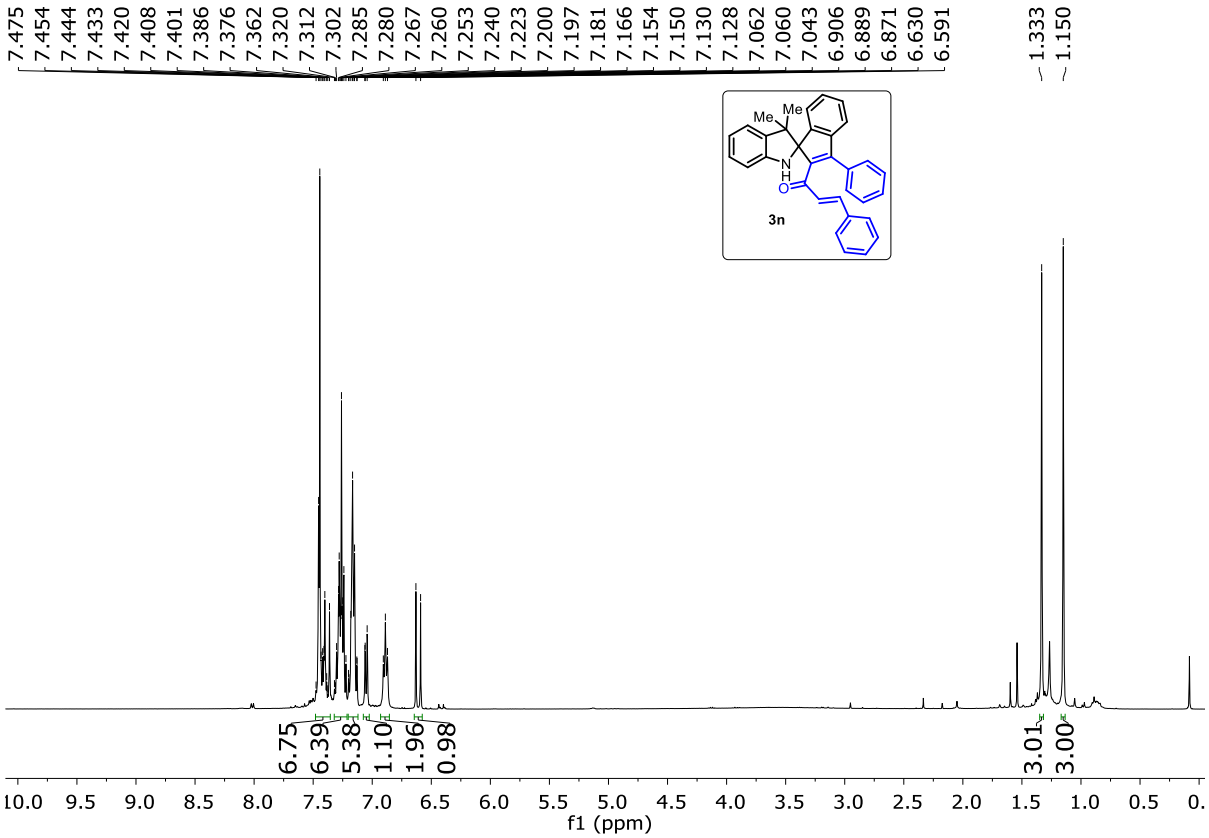
^{13}C{^{1}H} NMR of 3m (100 MHz, $CDCl_3$):



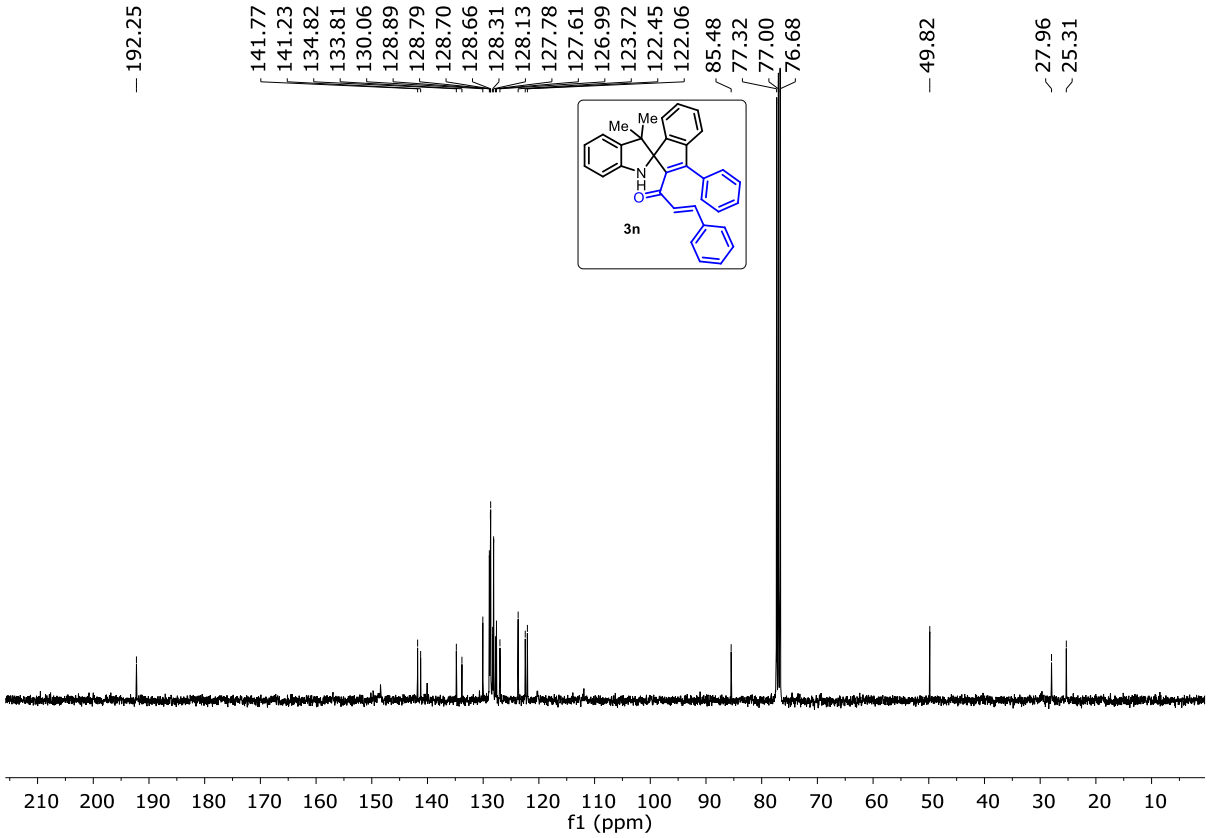
^{19}F NMR of 3m (376 MHz, $CDCl_3$):



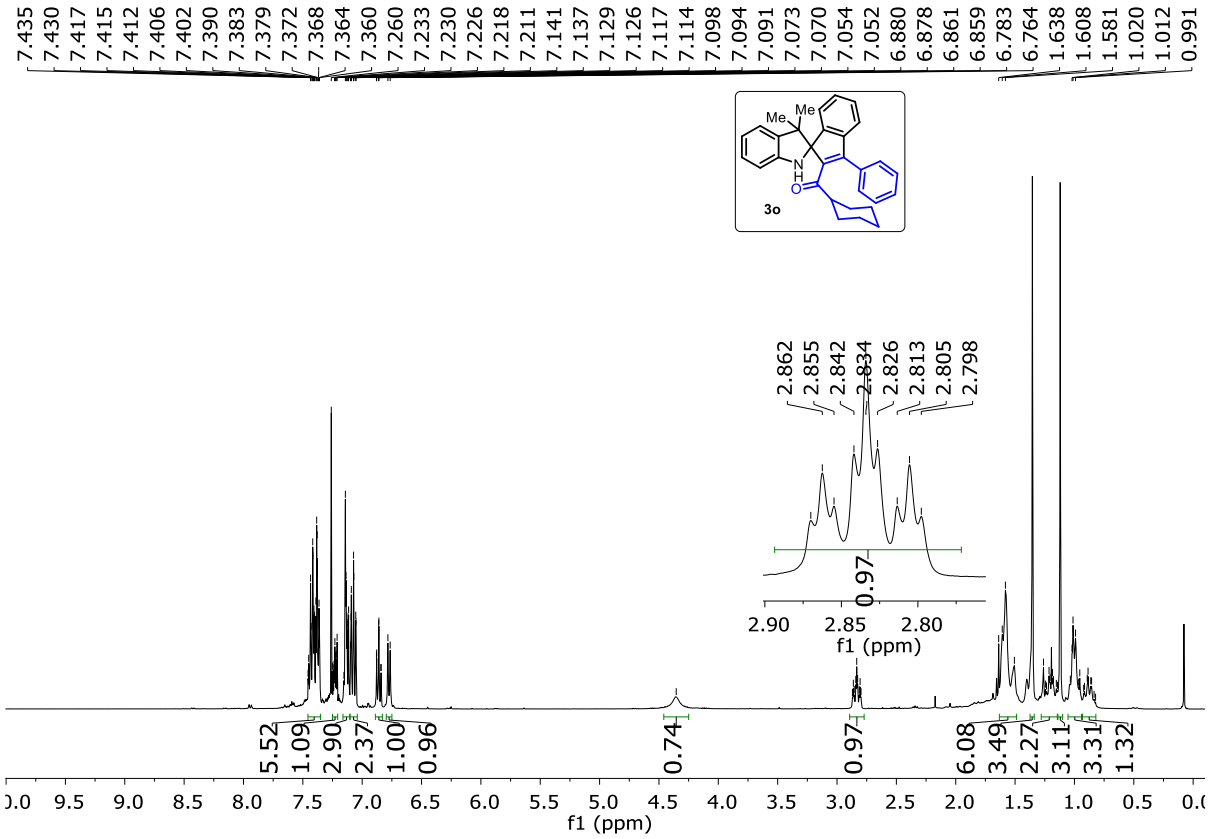
^{1}H NMR of 3n (400 MHz, $CDCl_3$):



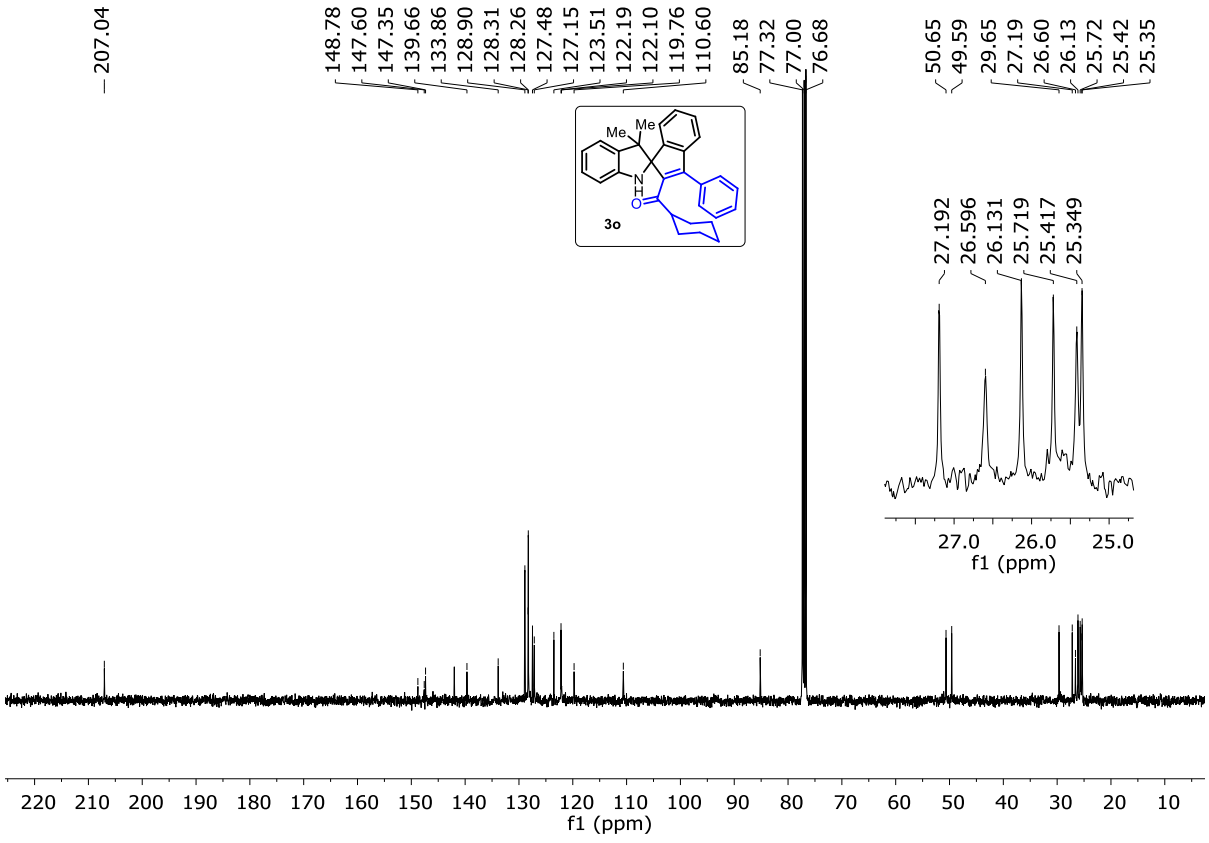
^{13}C{^{1}H} NMR of 3n (101 MHz, $CDCl_3$):



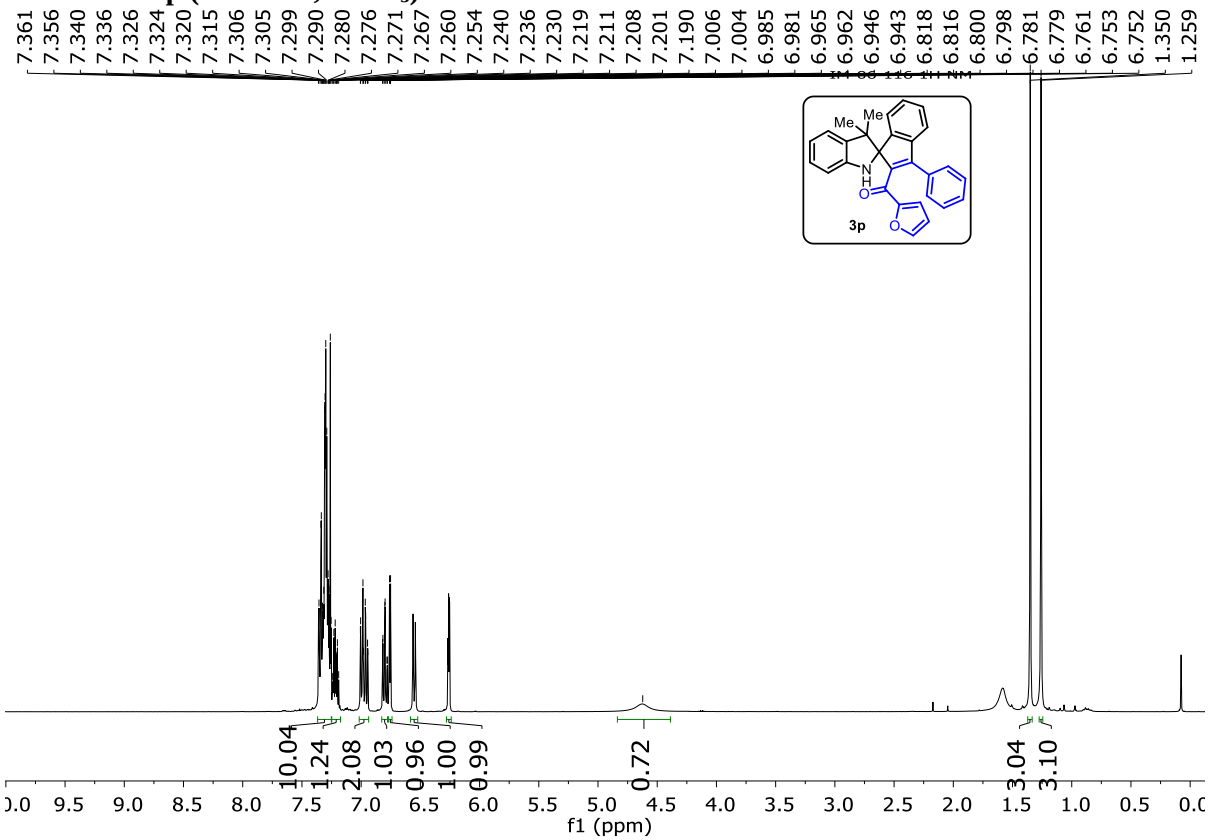
^{1}H NMR of 3o (400 MHz, $CDCl_3$):



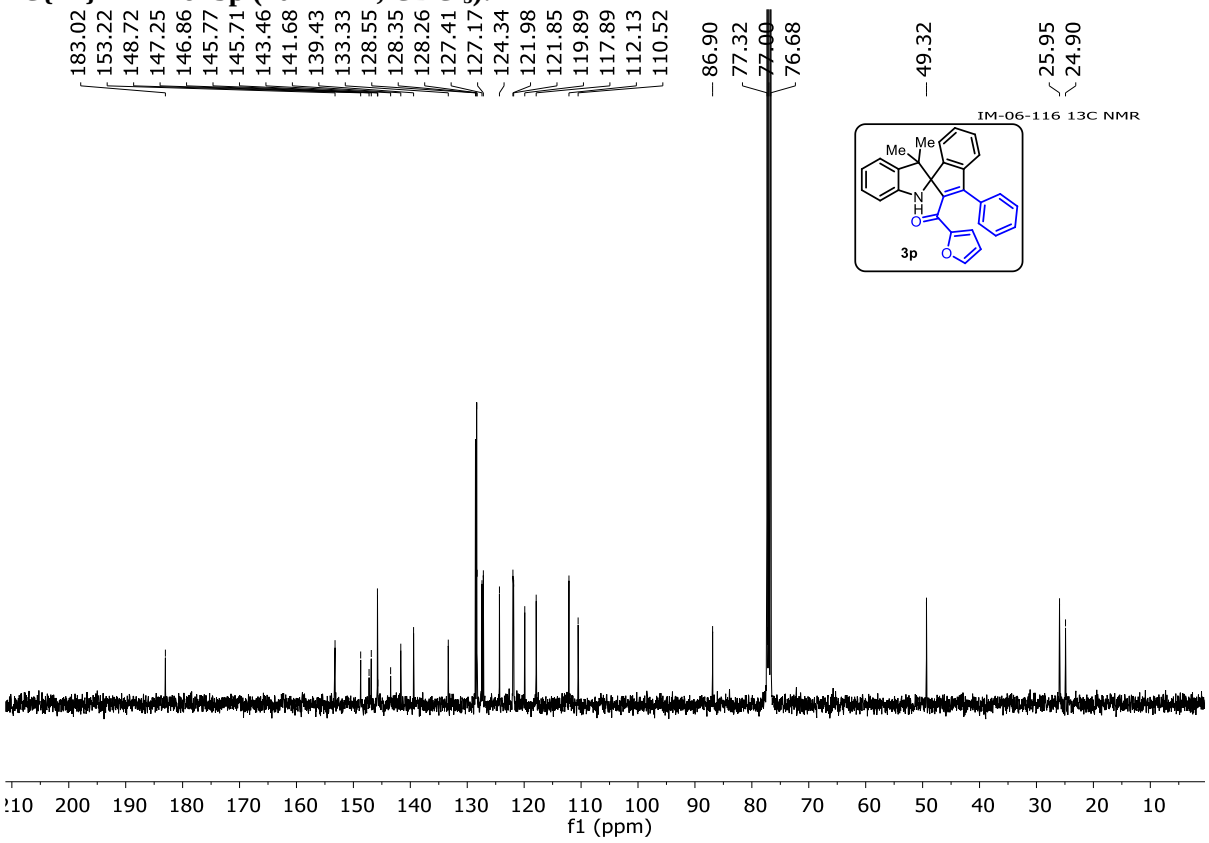
^{13}C{^{1}H} NMR of 3o (101 MHz, $CDCl_3$):



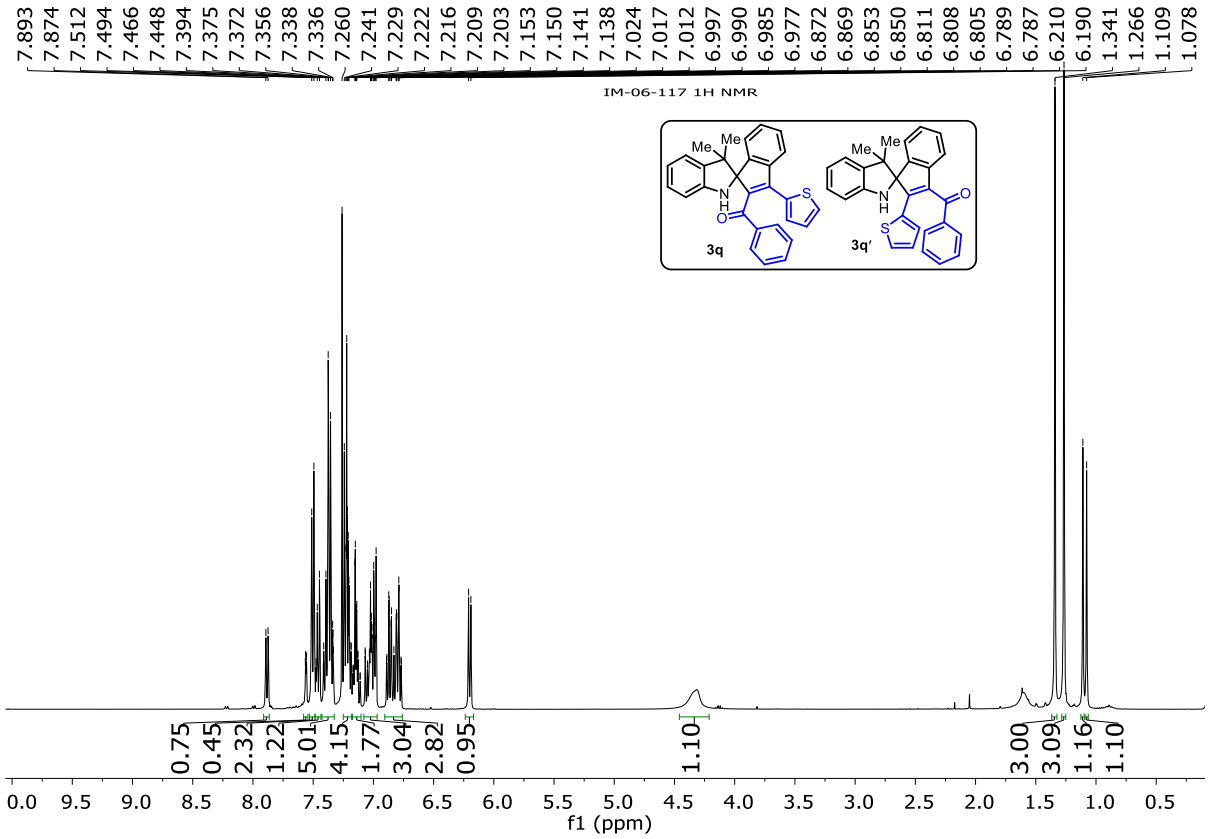
^{1}H NMR of 3p (400 MHz, $CDCl_3$):



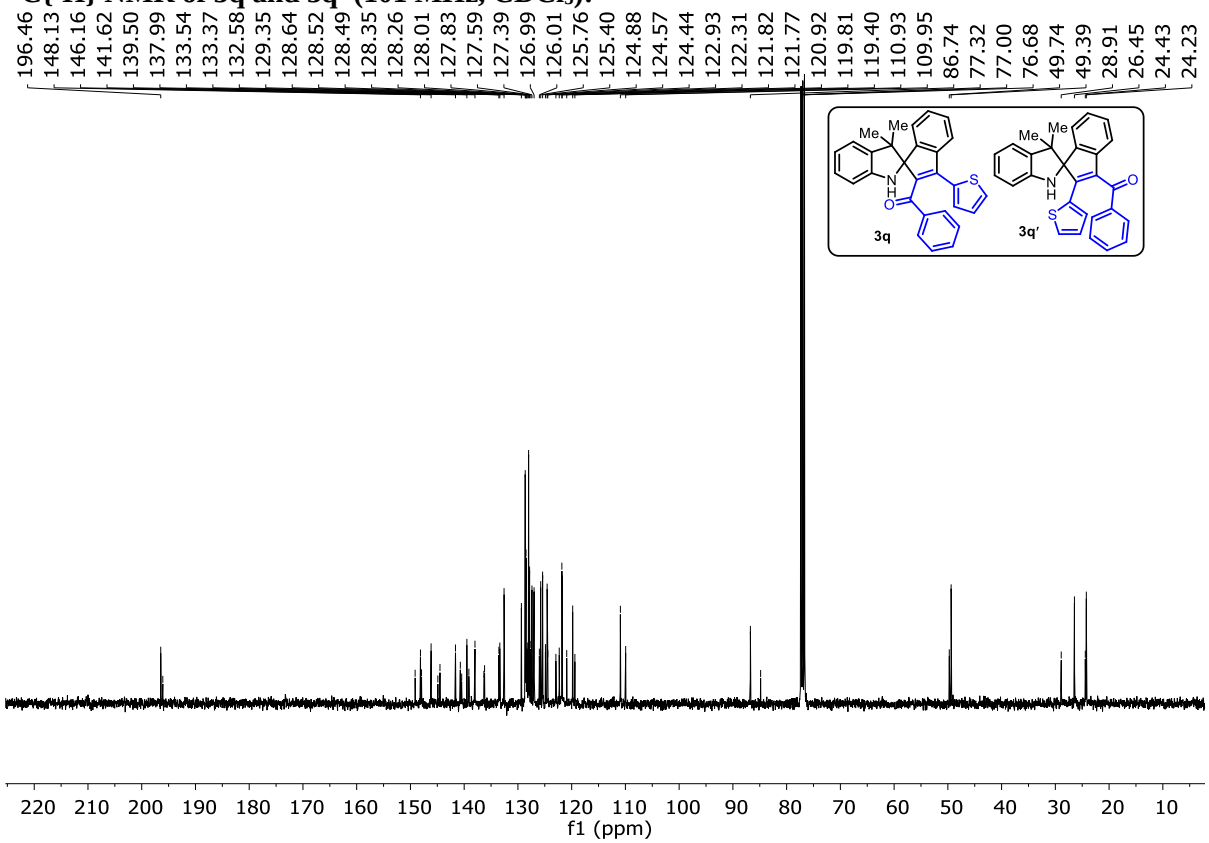
^{13}C{^{1}H} NMR of 3p (101 MHz, $CDCl_3$):



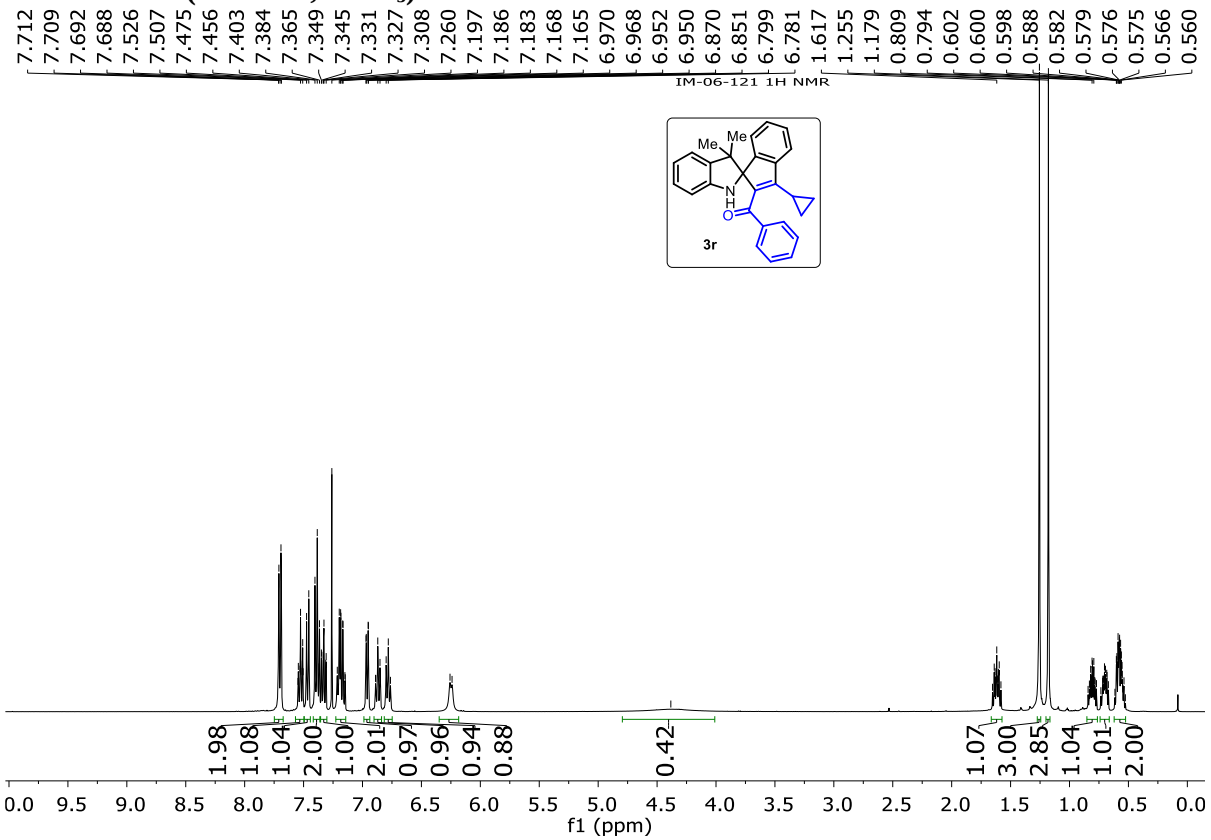
^{1}H NMR of 3q and 3q' (400 MHz, $CDCl_3$):



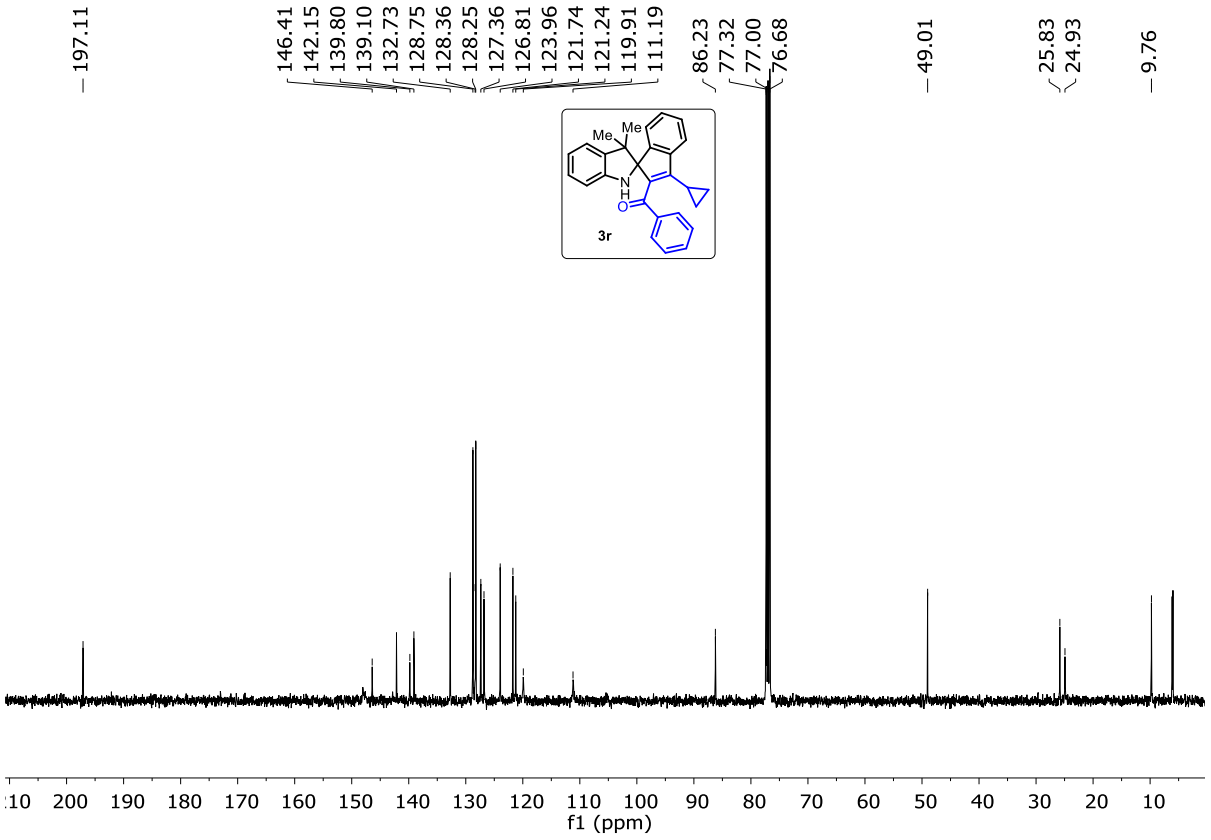
^{13}C{^{1}H} NMR of 3q and 3q' (101 MHz, $CDCl_3$):



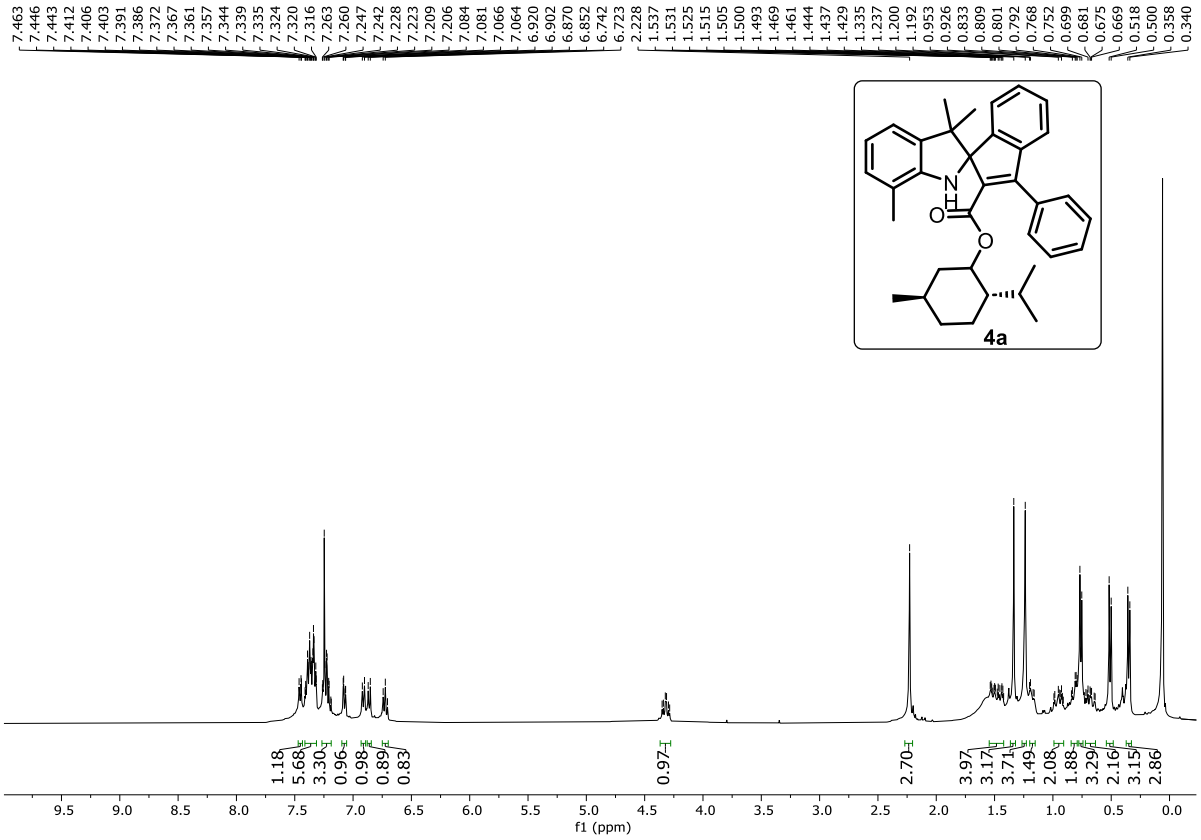
^{1}H NMR of 3r (400 MHz, $CDCl_3$):



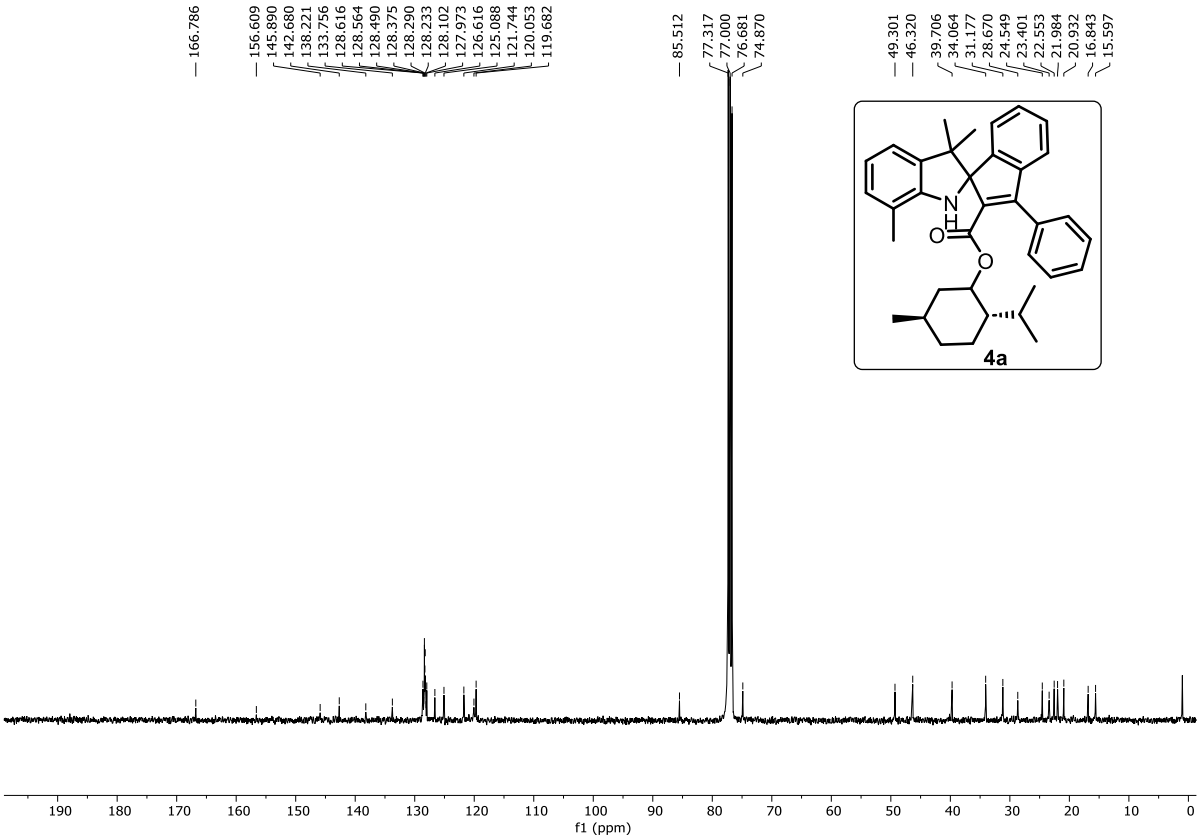
^{13}C{^{1}H} NMR of 3r (101 MHz, $CDCl_3$):



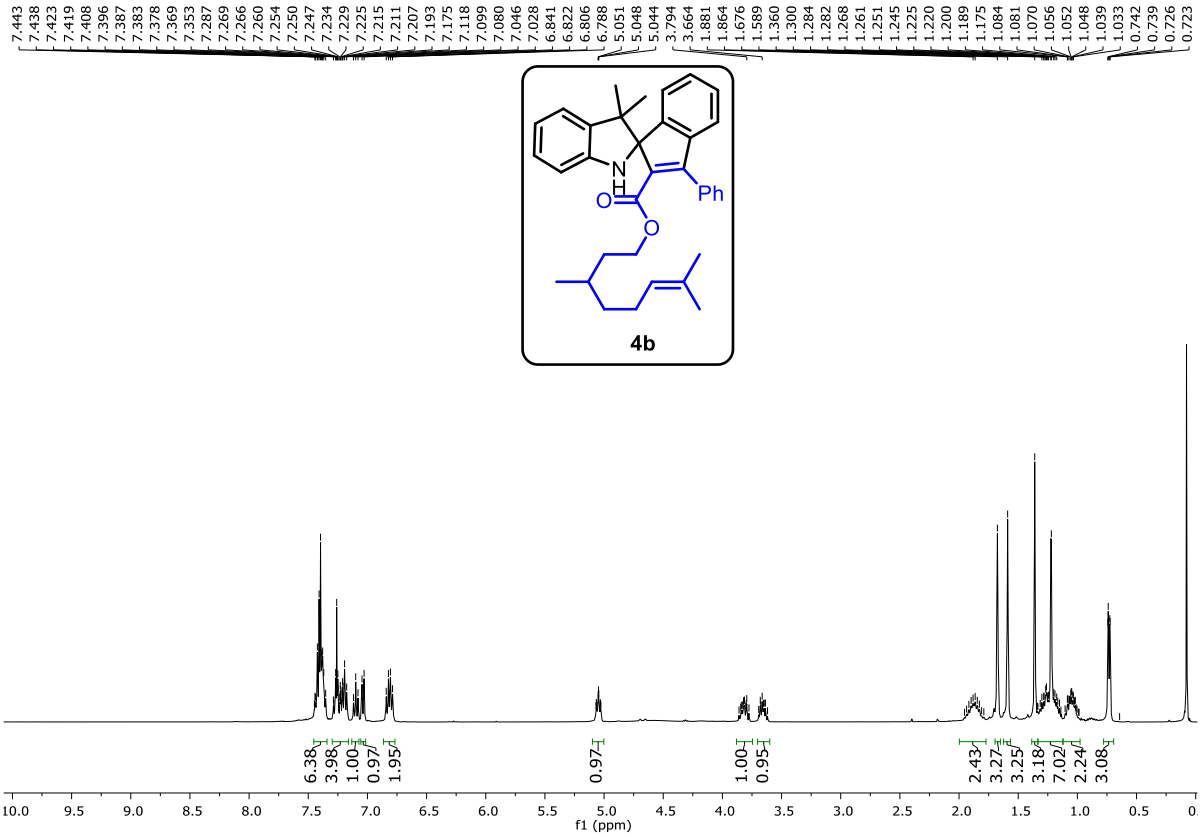
^{1}H NMR of 4a (400 MHz, $CDCl_3$):



^{13}C{^{1}H} NMR of 4a (101 MHz, $CDCl_3$):



^{1}H NMR of 4b (400 MHz, $CDCl_3$):



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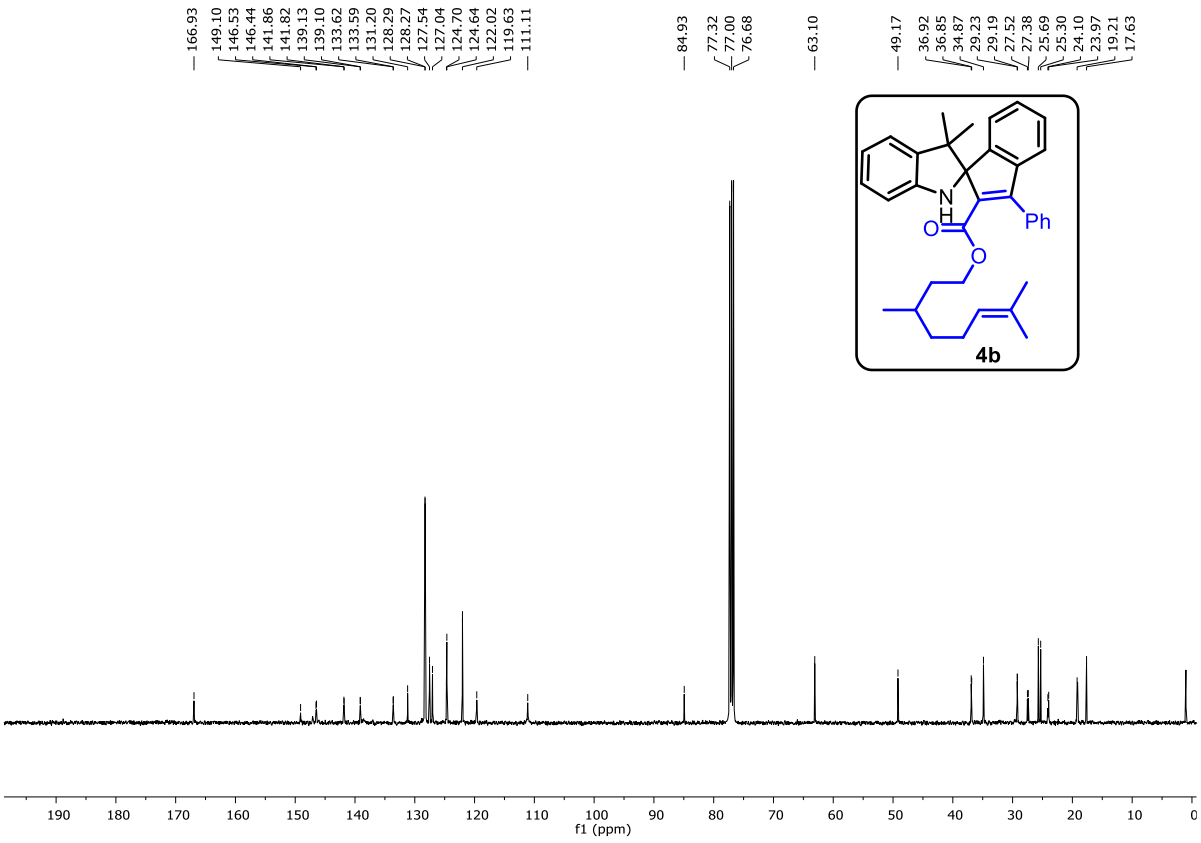
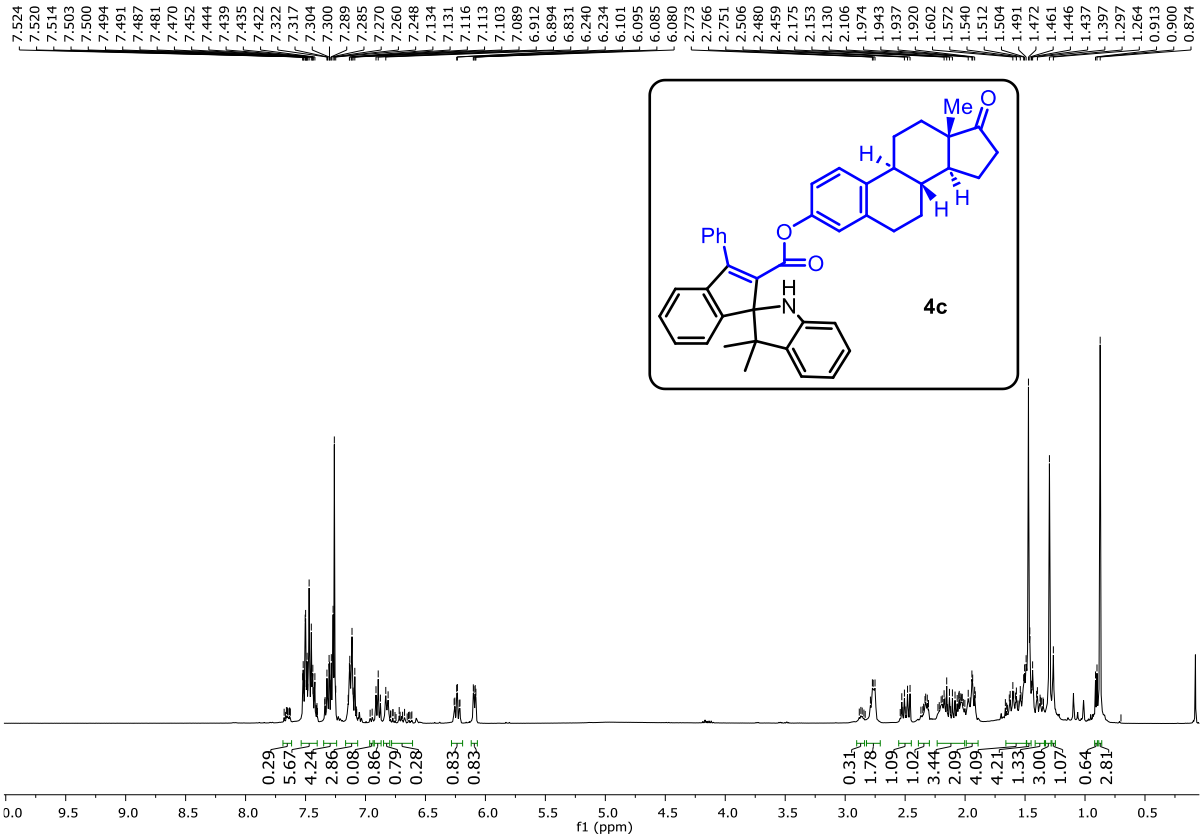
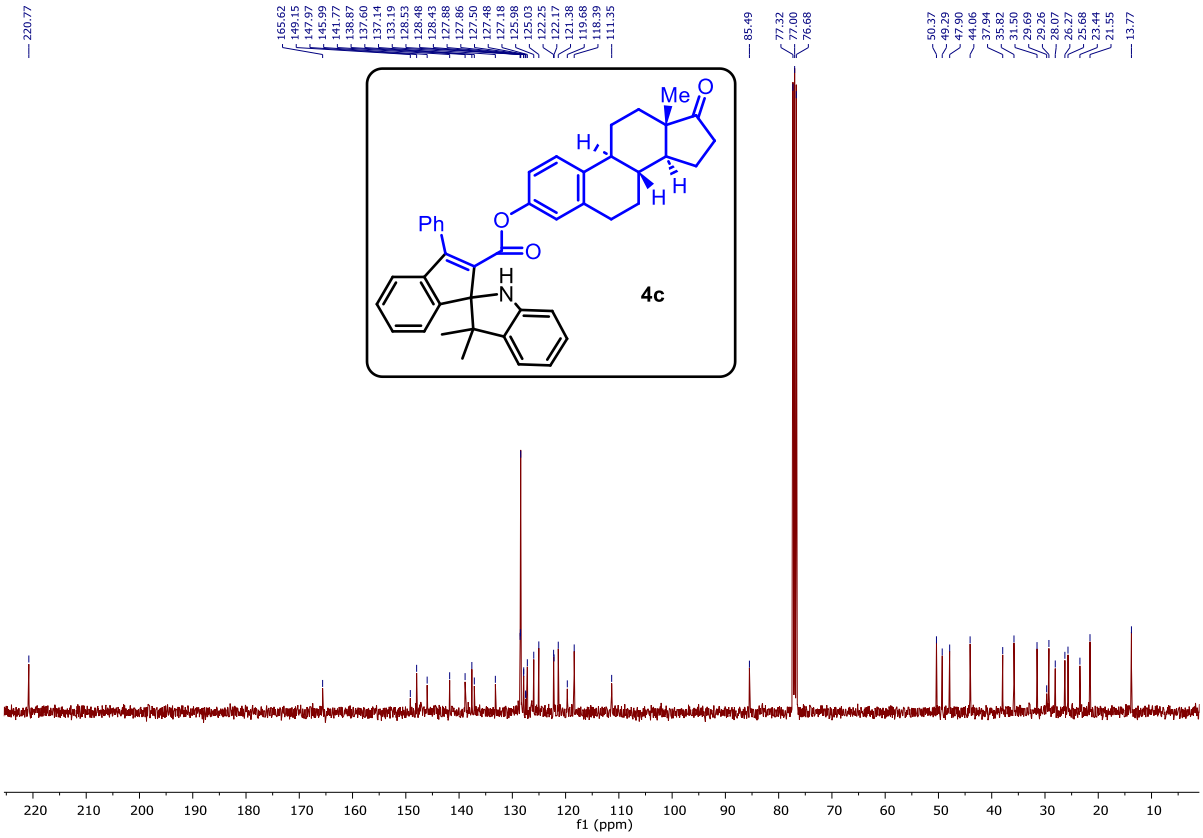


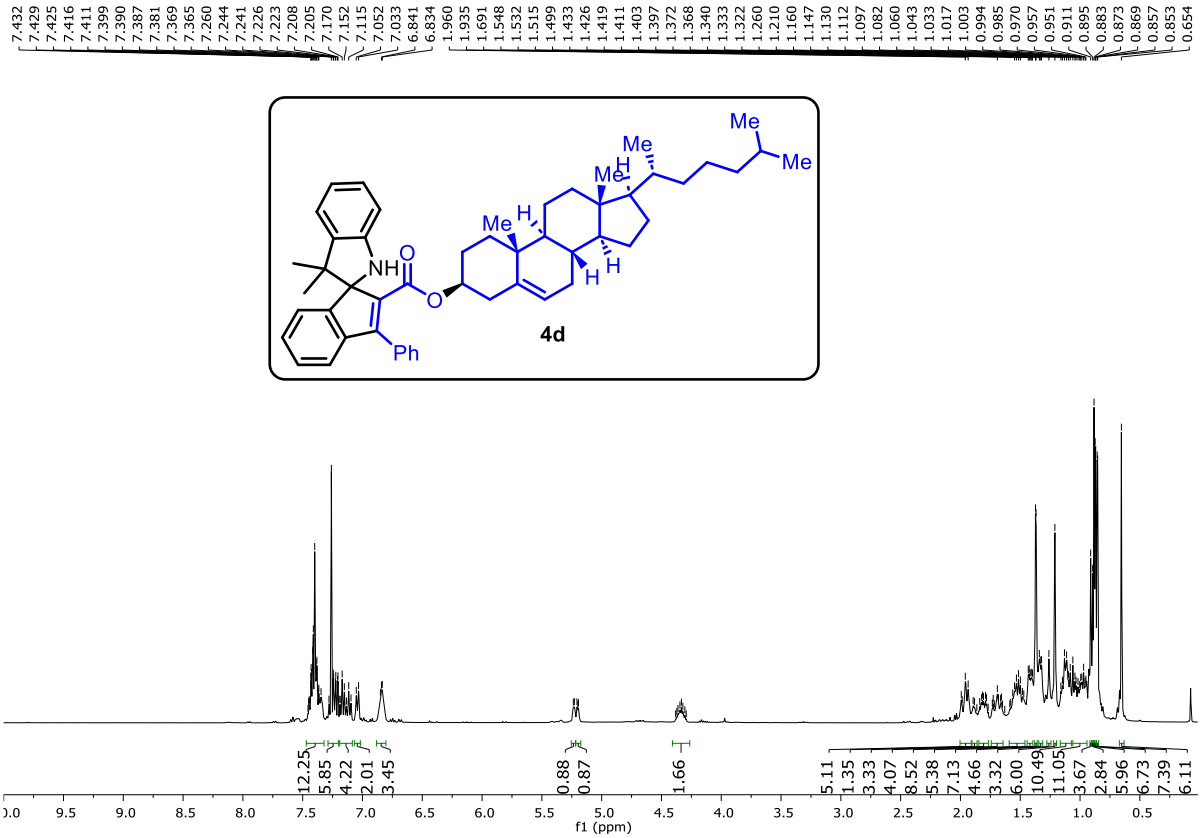
^{1}H NMR of 4c (400 MHz, $CDCl_3$):



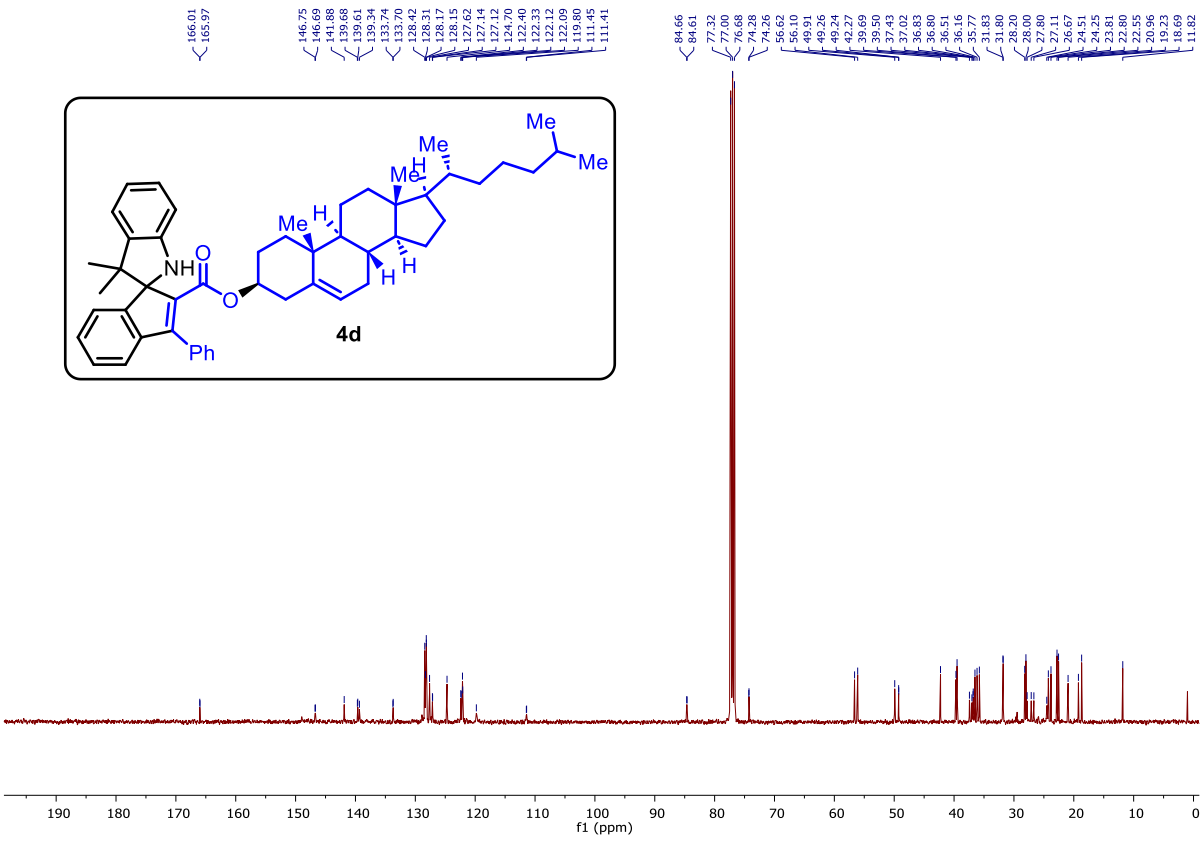
^{13}C{^{1}H} NMR of 4c (101 MHz, $CDCl_3$):



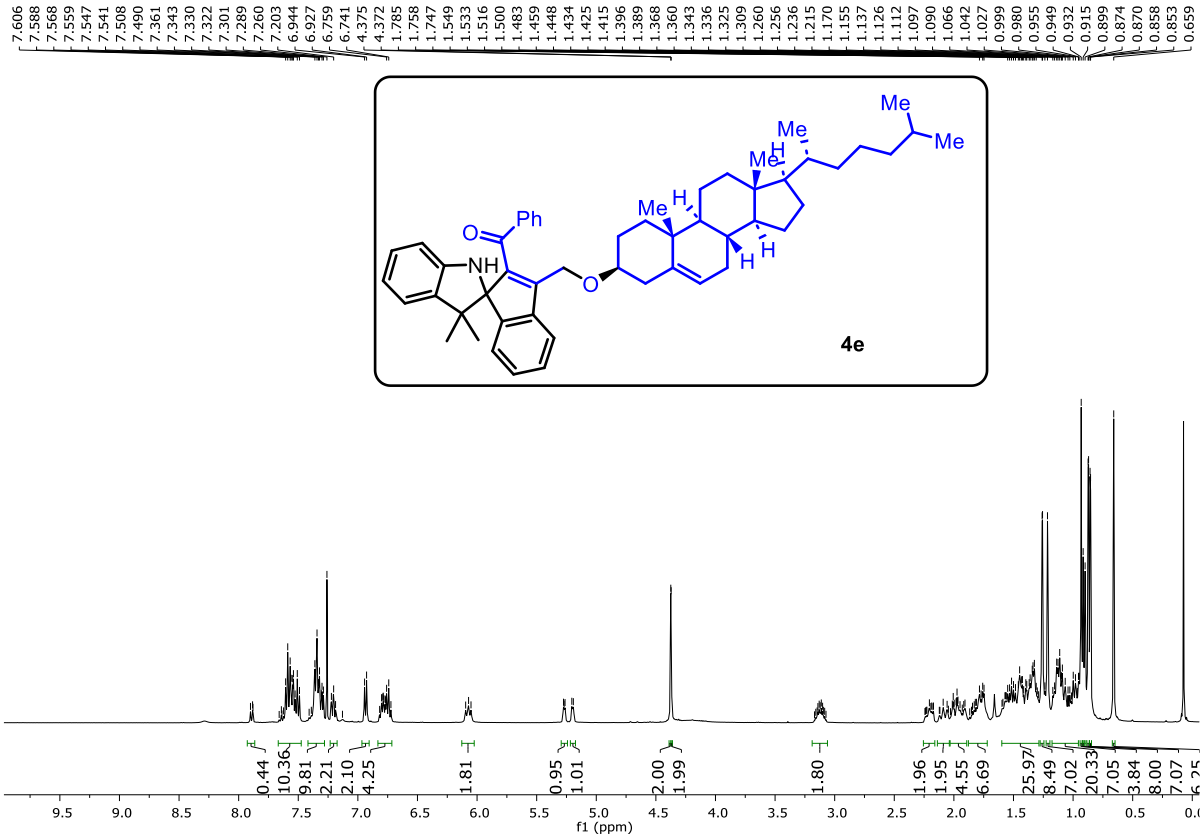
^{1}H NMR of 4d (400 MHz, $CDCl_3$):



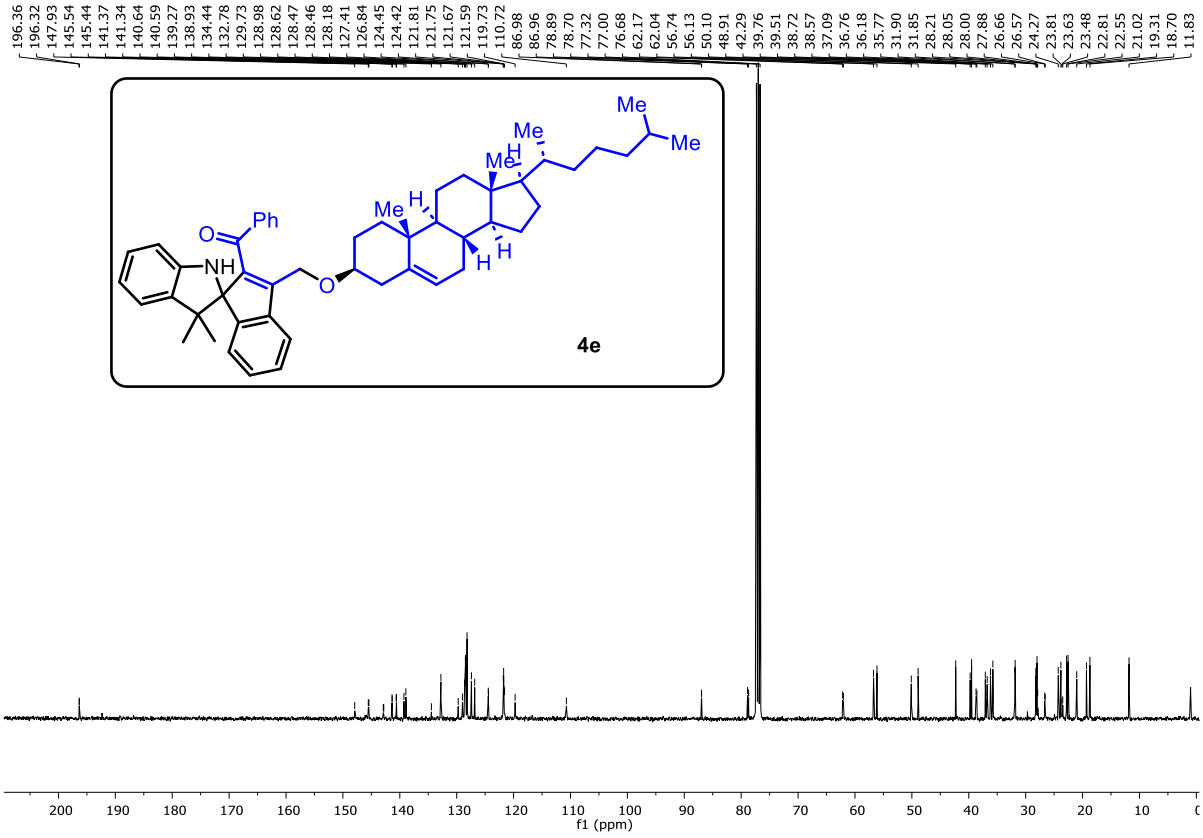
^{13}C{^{1}H} NMR of 4d (101 MHz, $CDCl_3$):



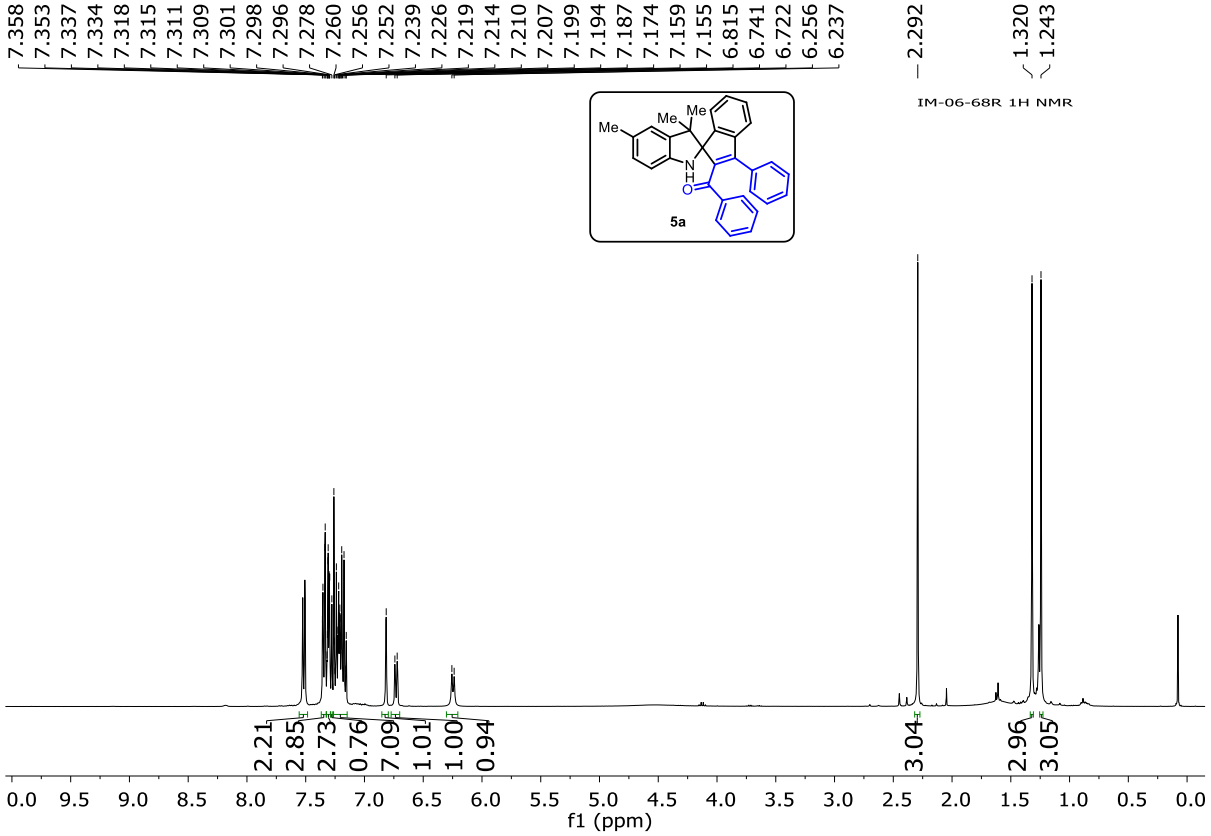
^{1}H NMR of 4e (400 MHz, $CDCl_3$):



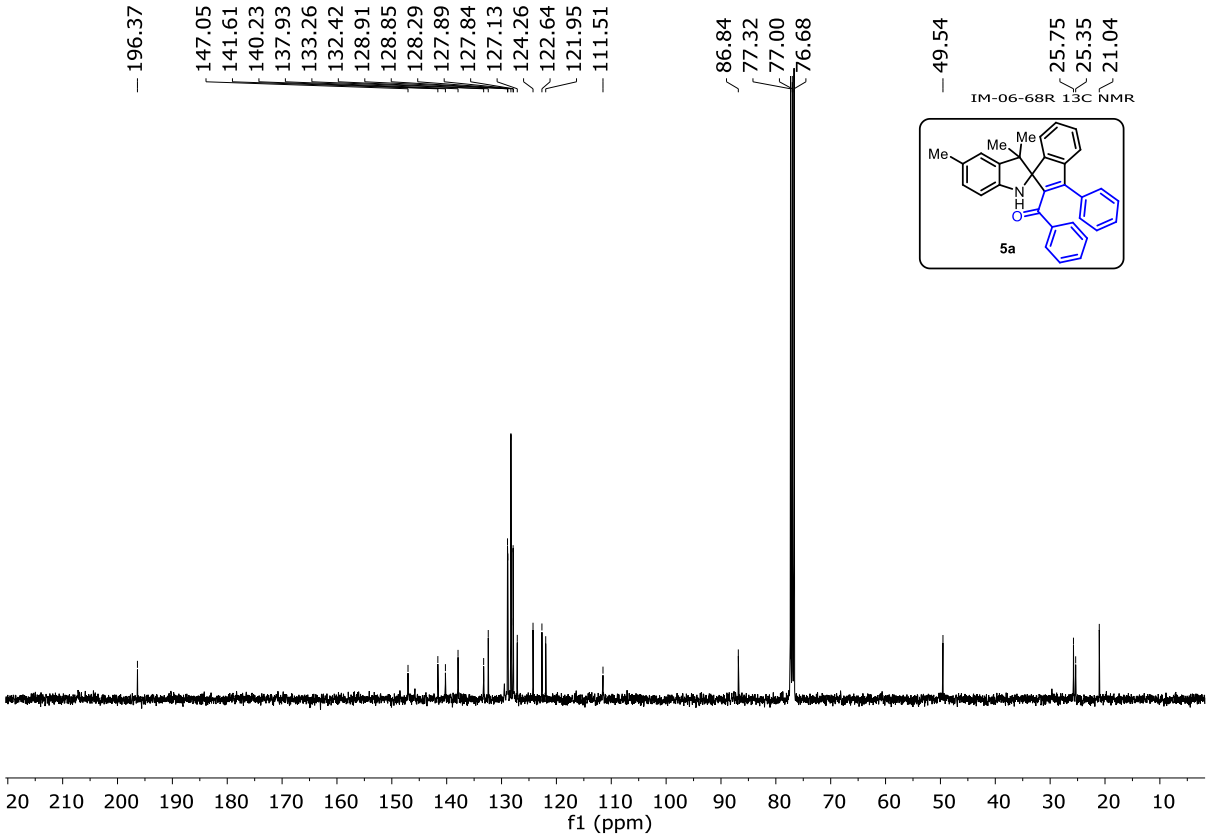
^{13}C{^{1}H} NMR of 4e (101 MHz, $CDCl_3$):



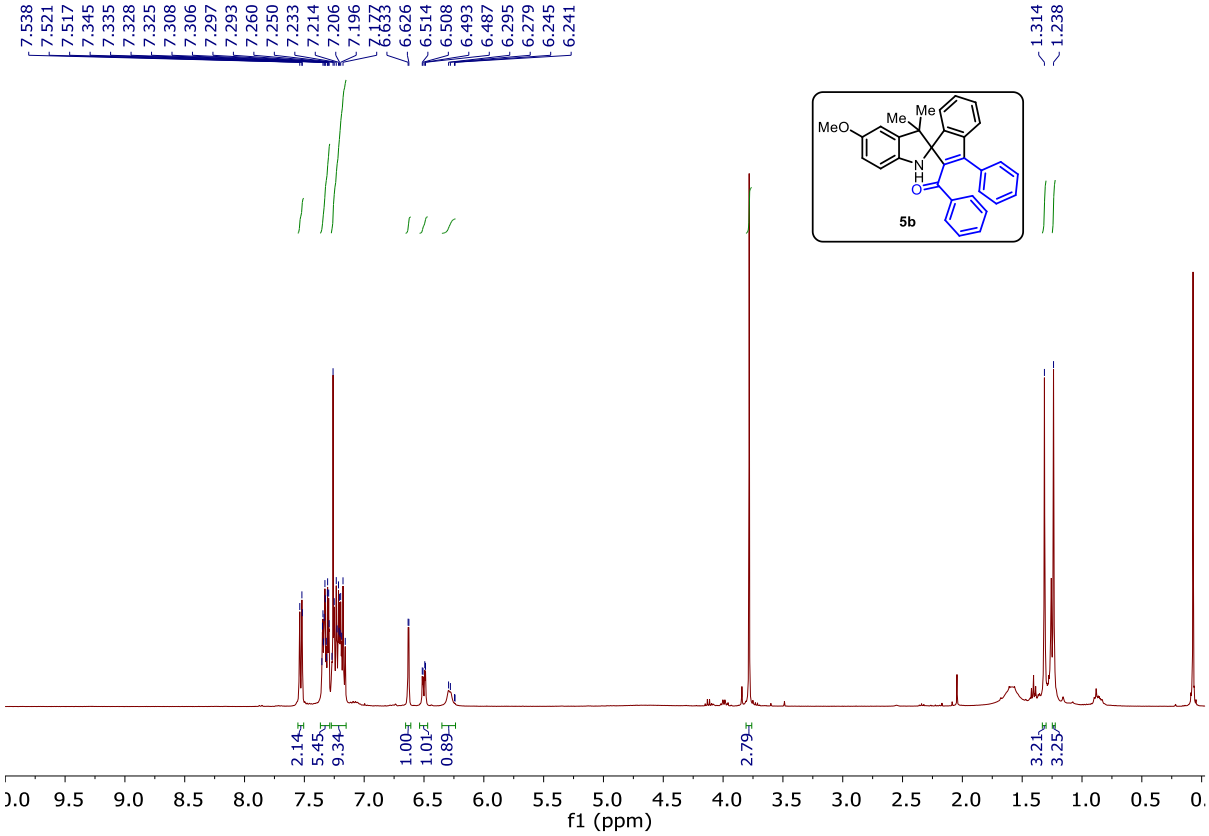
^{1}H NMR of 5a (400 MHz, $CDCl_3$):



^{13}C{^{1}H} NMR of 5a (101 MHz, $CDCl_3$):



^{1}H NMR of 5b (400 MHz, $CDCl_3$):



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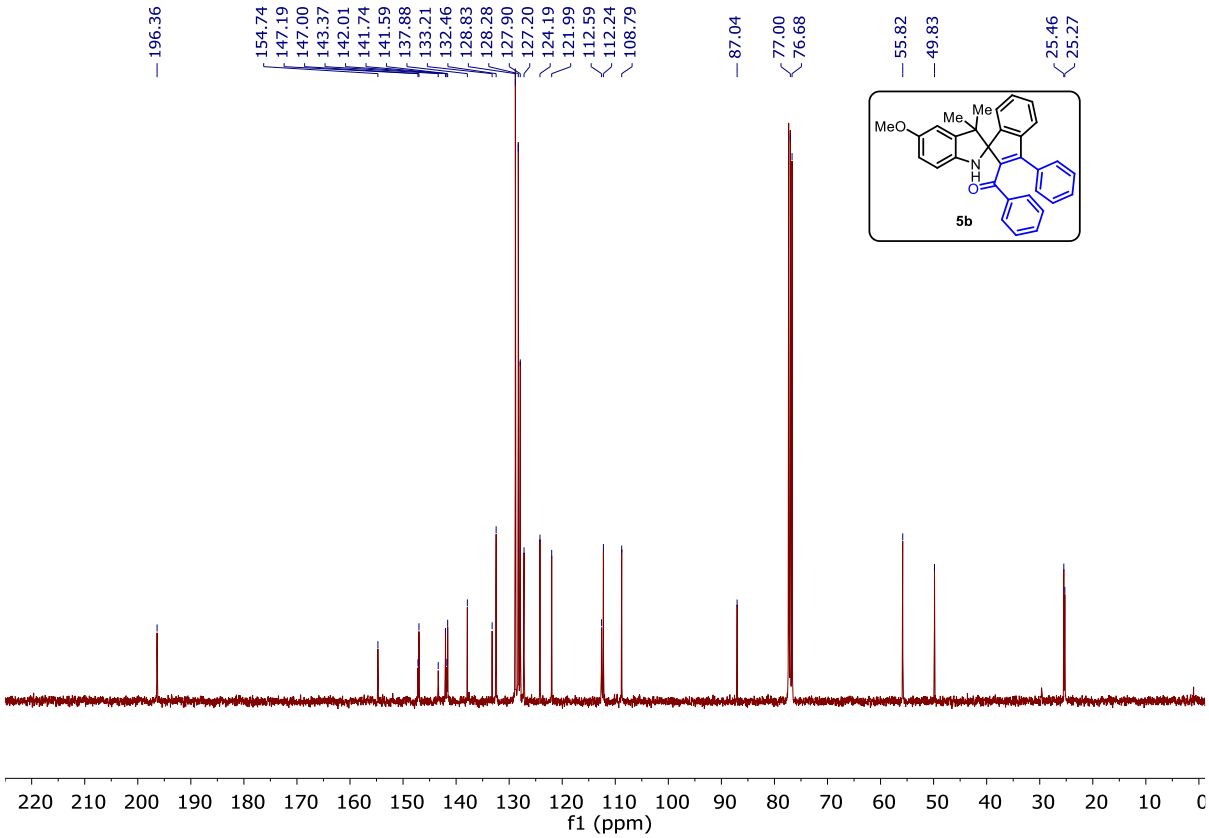
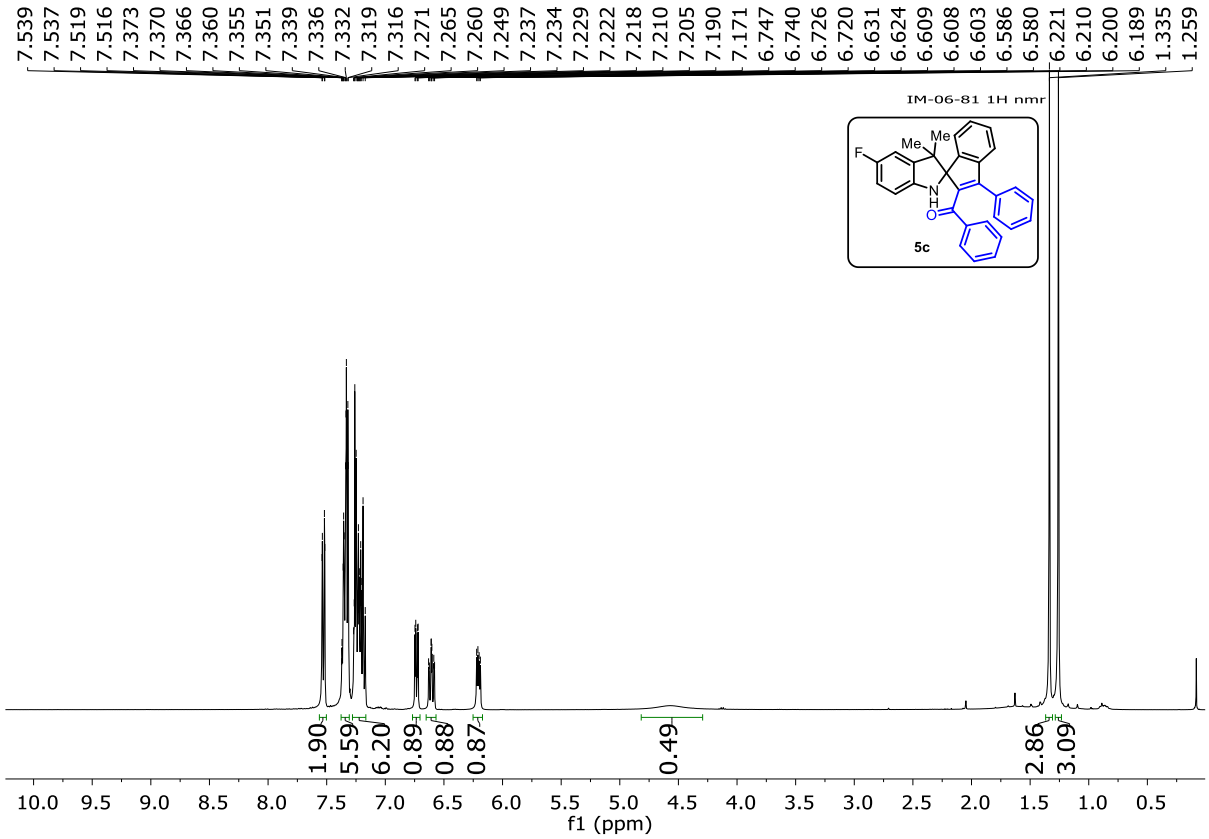
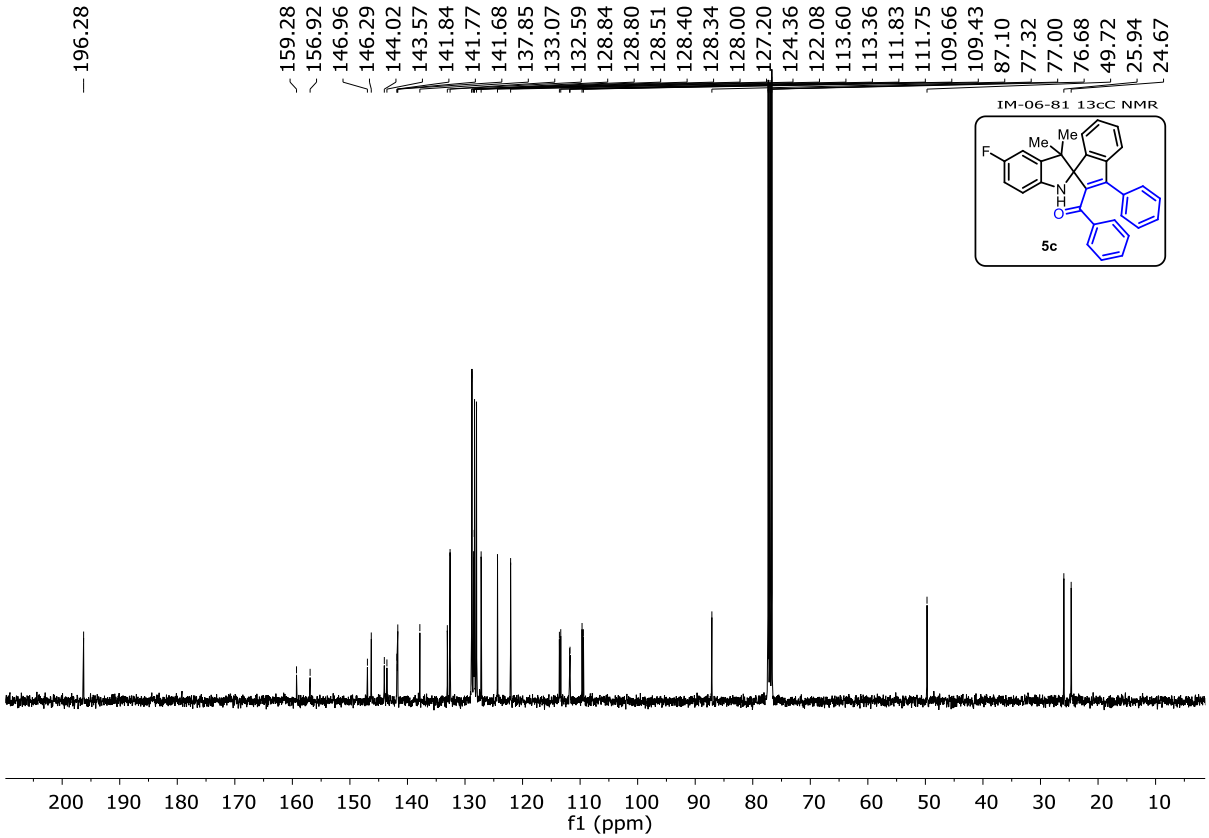


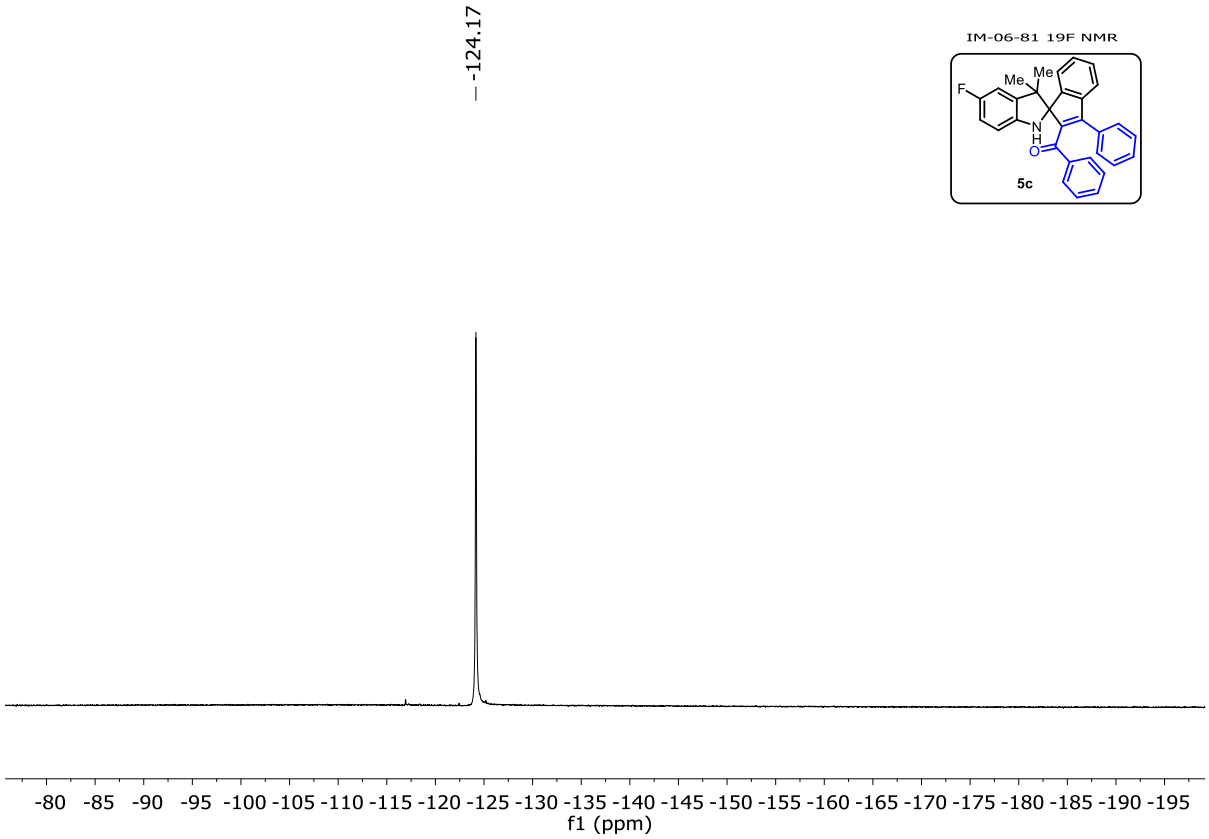
^{1}H NMR of 5c (400 MHz, $CDCl_3$):



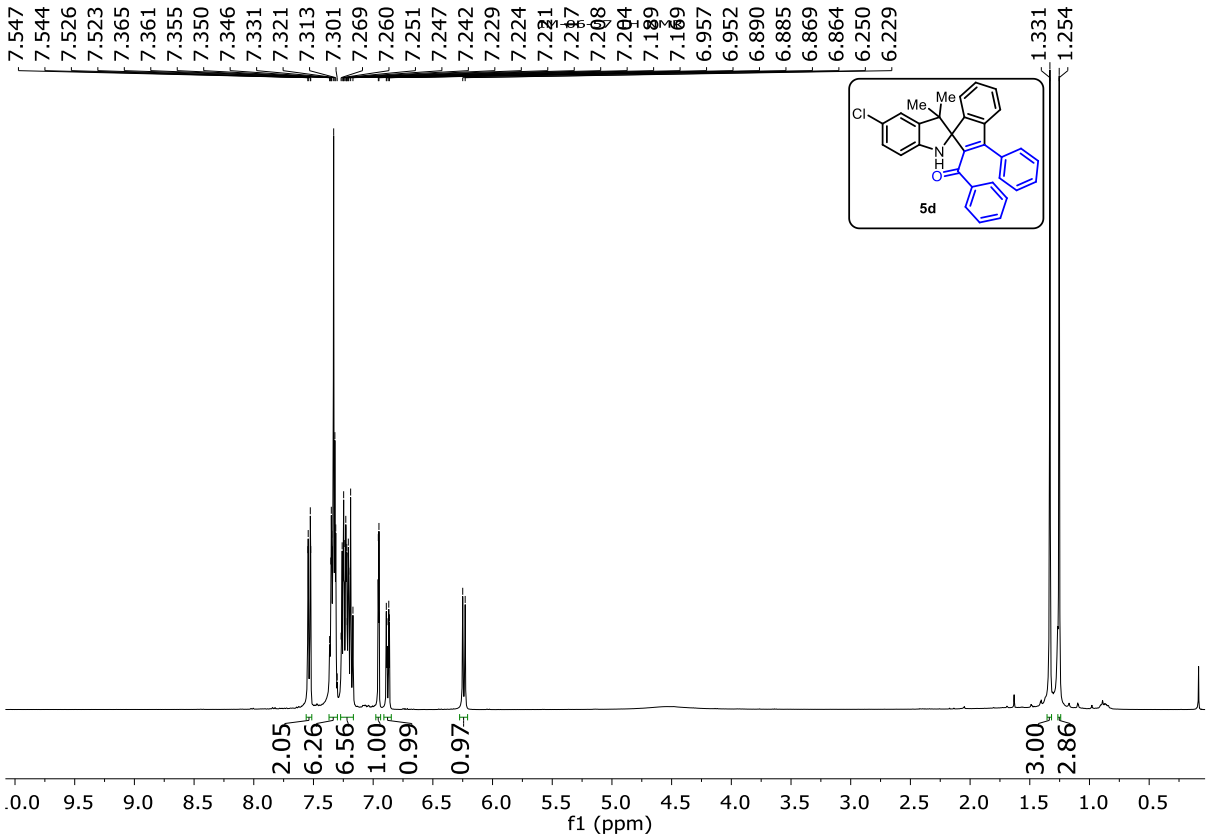
^{13}C{^{1}H} NMR of 5c (101 MHz, $CDCl_3$):



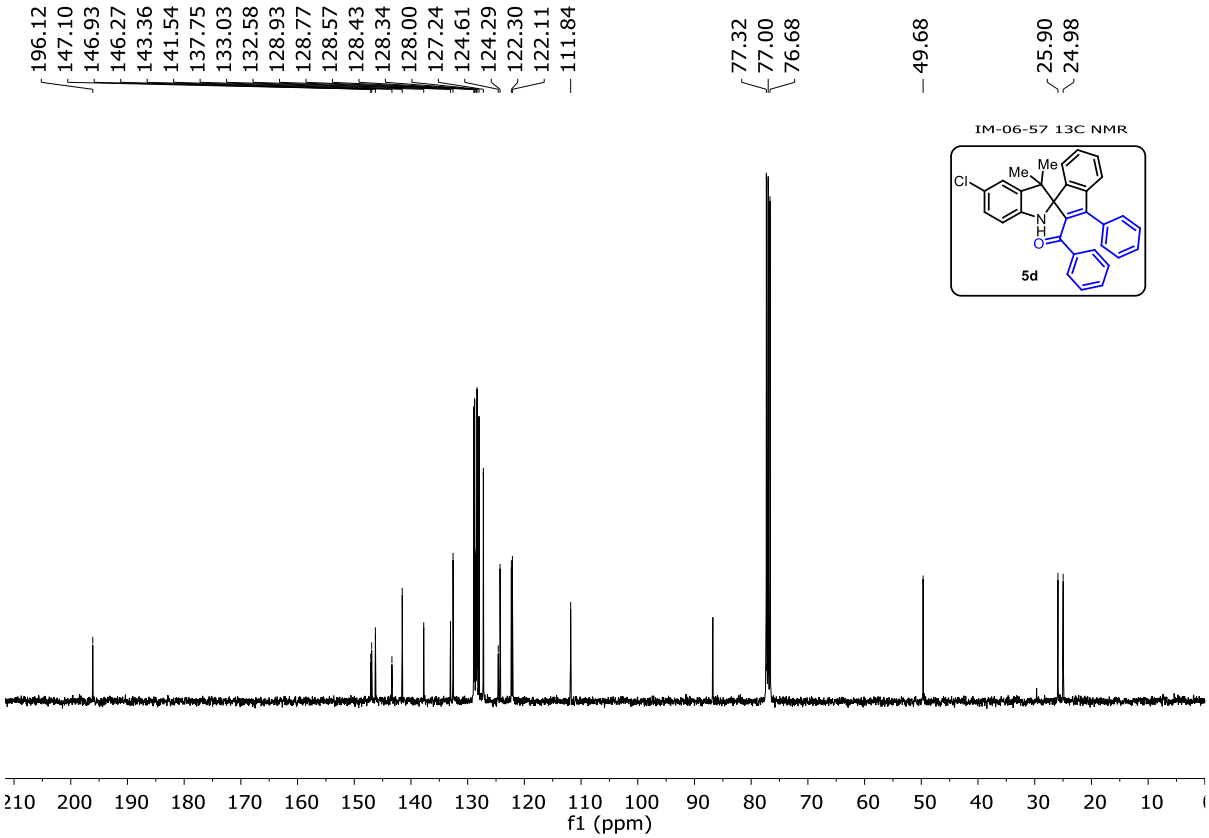
^{19}F NMR of 5c (376 MHz, $CDCl_3$):



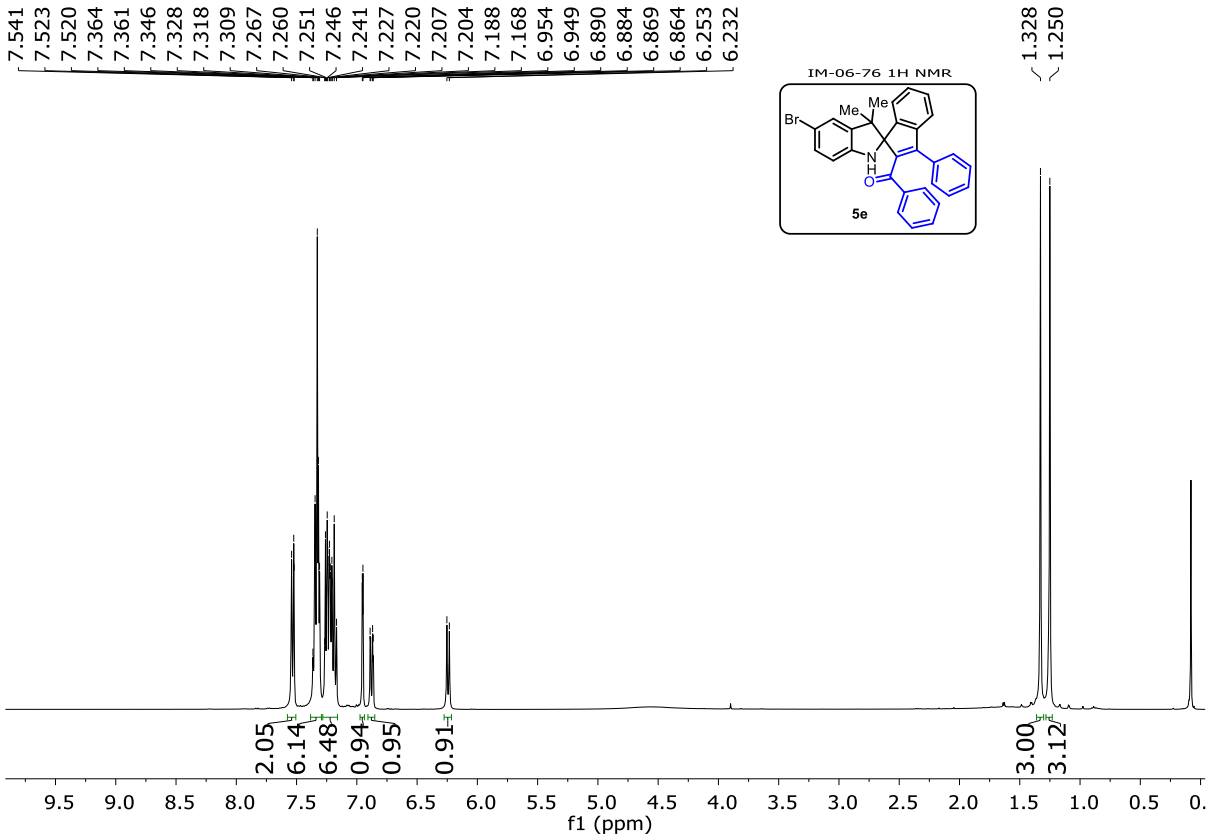
^{1}H NMR of 5d (400 MHz, $CDCl_3$):



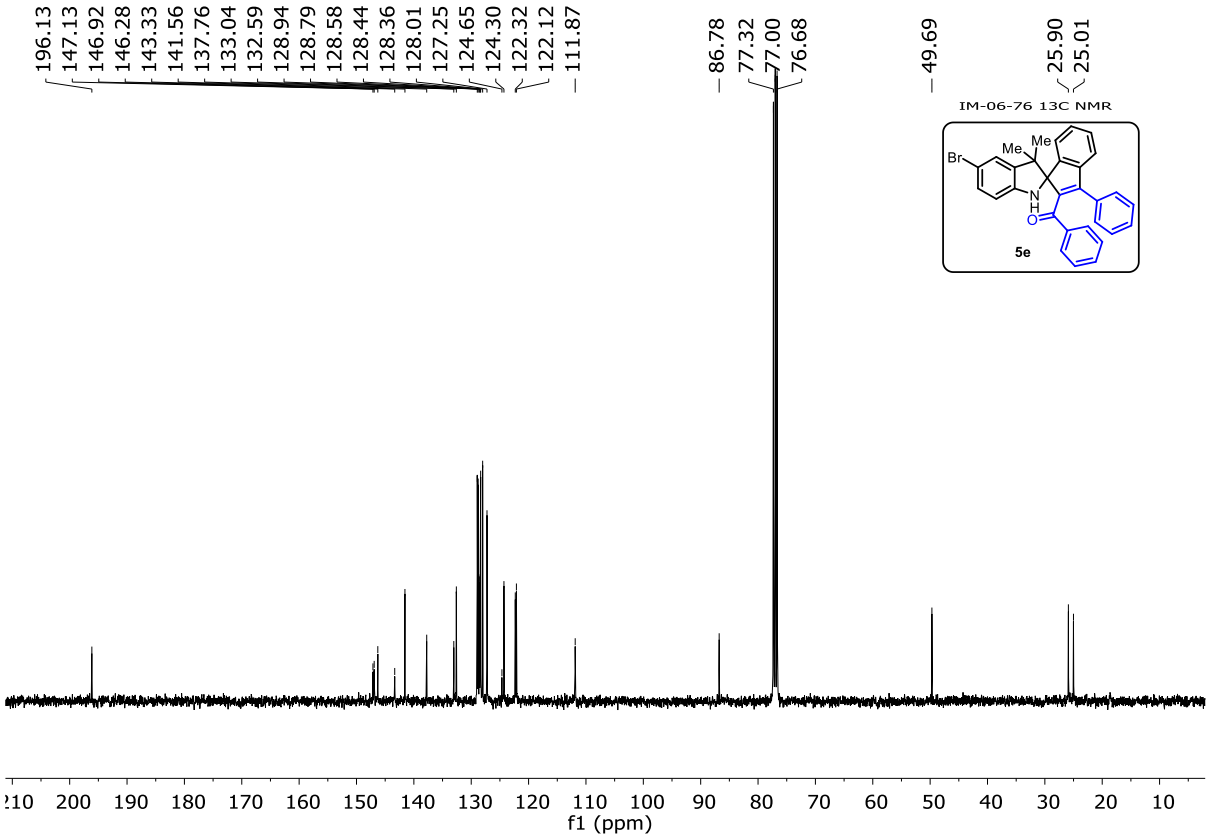
^{13}C{^{1}H} NMR of 5d (101 MHz, $CDCl_3$):



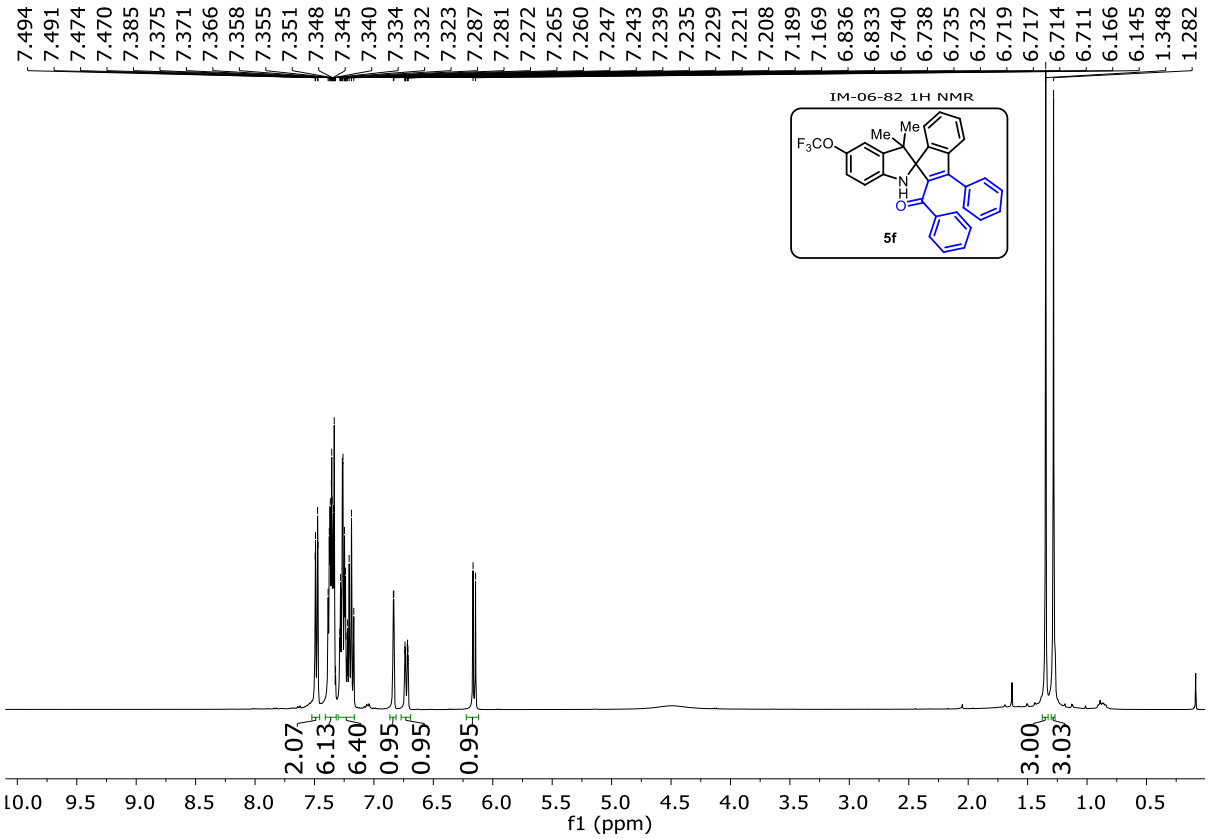
^{1}H NMR of 5e (400 MHz, $CDCl_3$):



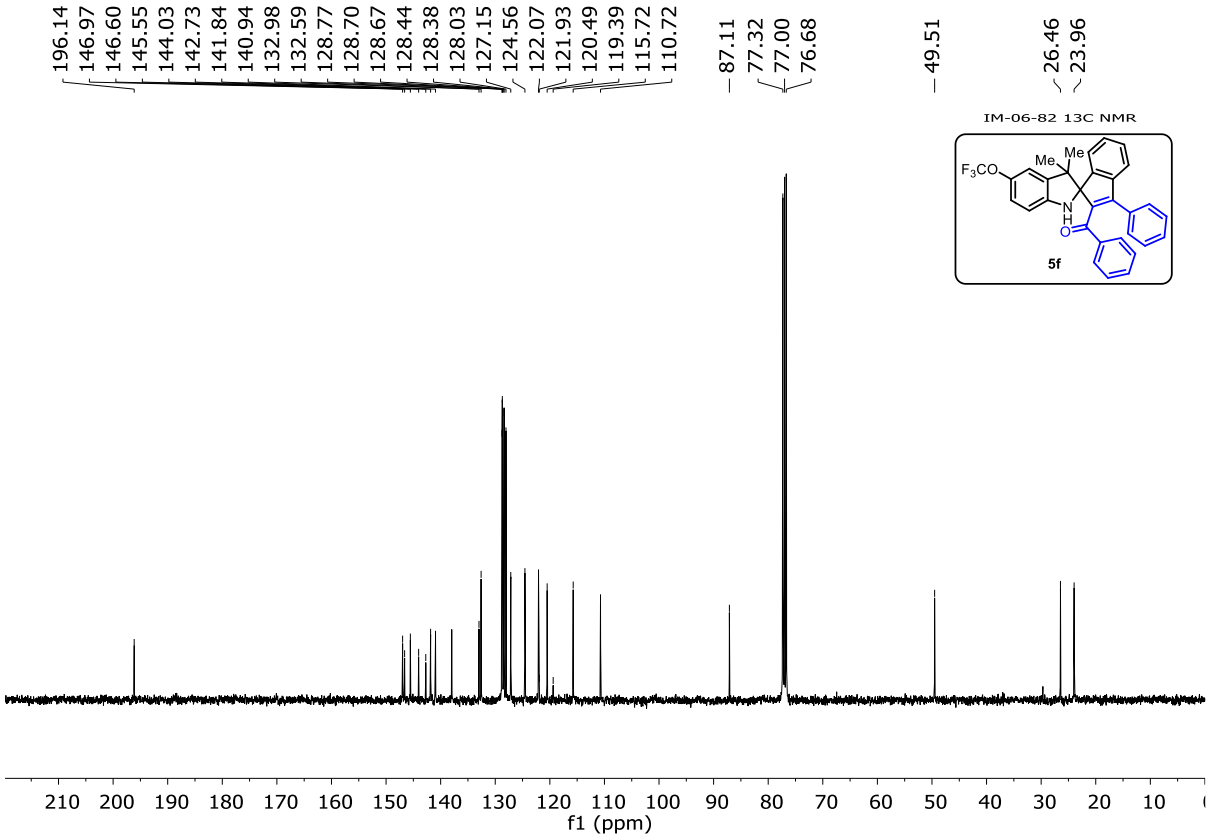
^{13}C{^{1}H} NMR of 5e (101 MHz, $CDCl_3$):



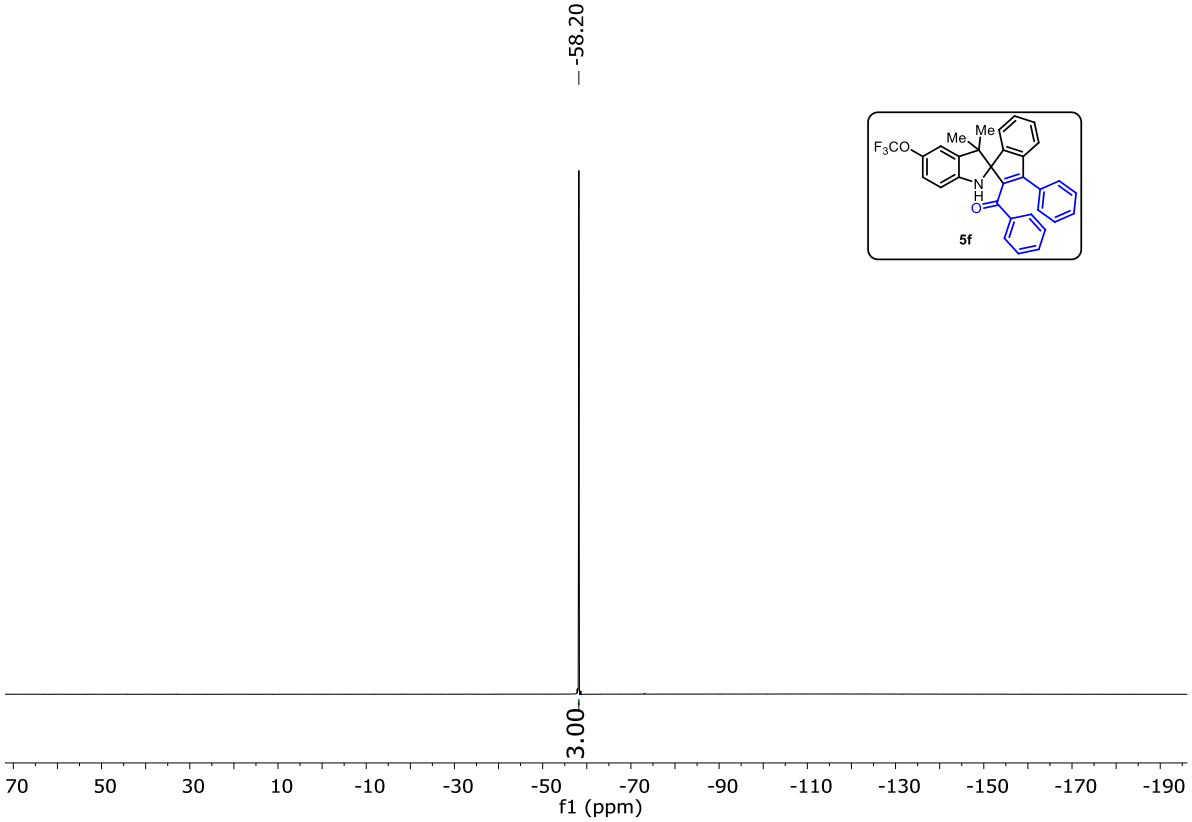
^{1}H NMR of 5f (400 MHz, $CDCl_3$):



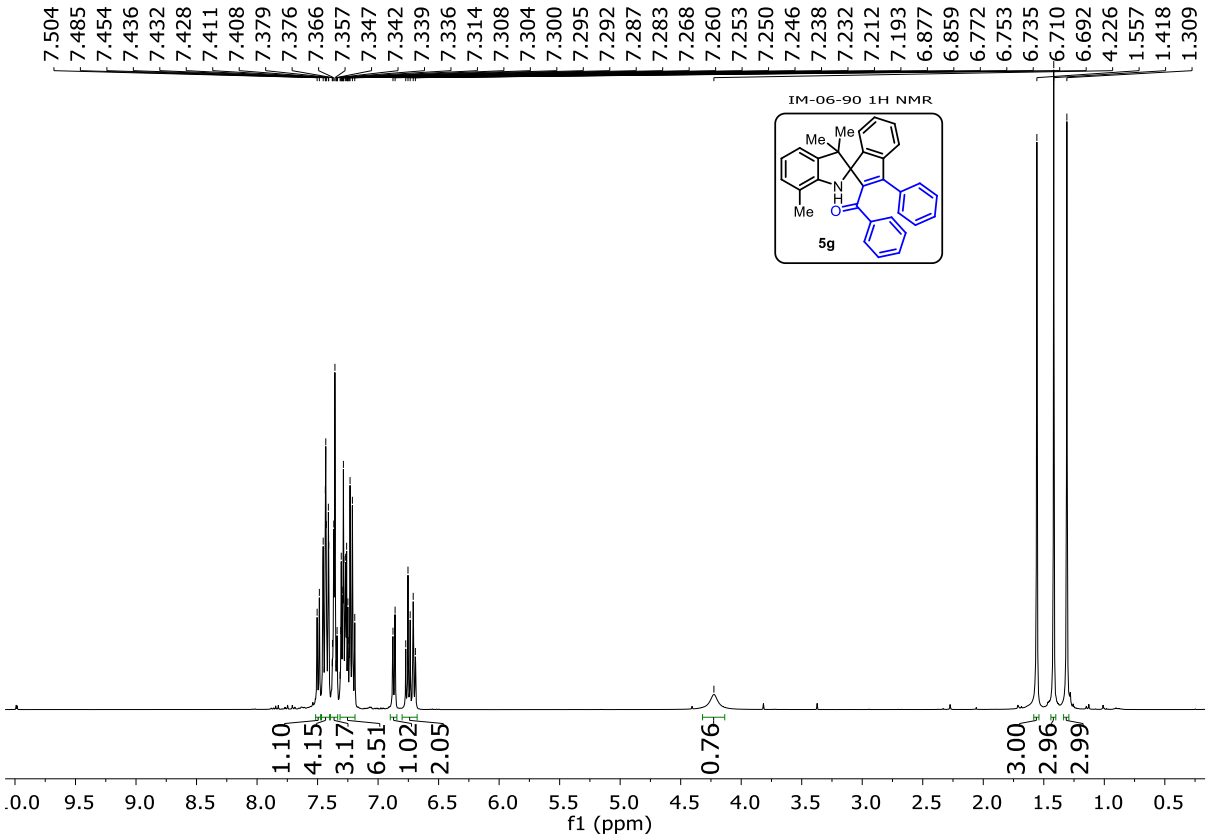
^{13}C{^{1}H} NMR of 5f (101 MHz, $CDCl_3$):



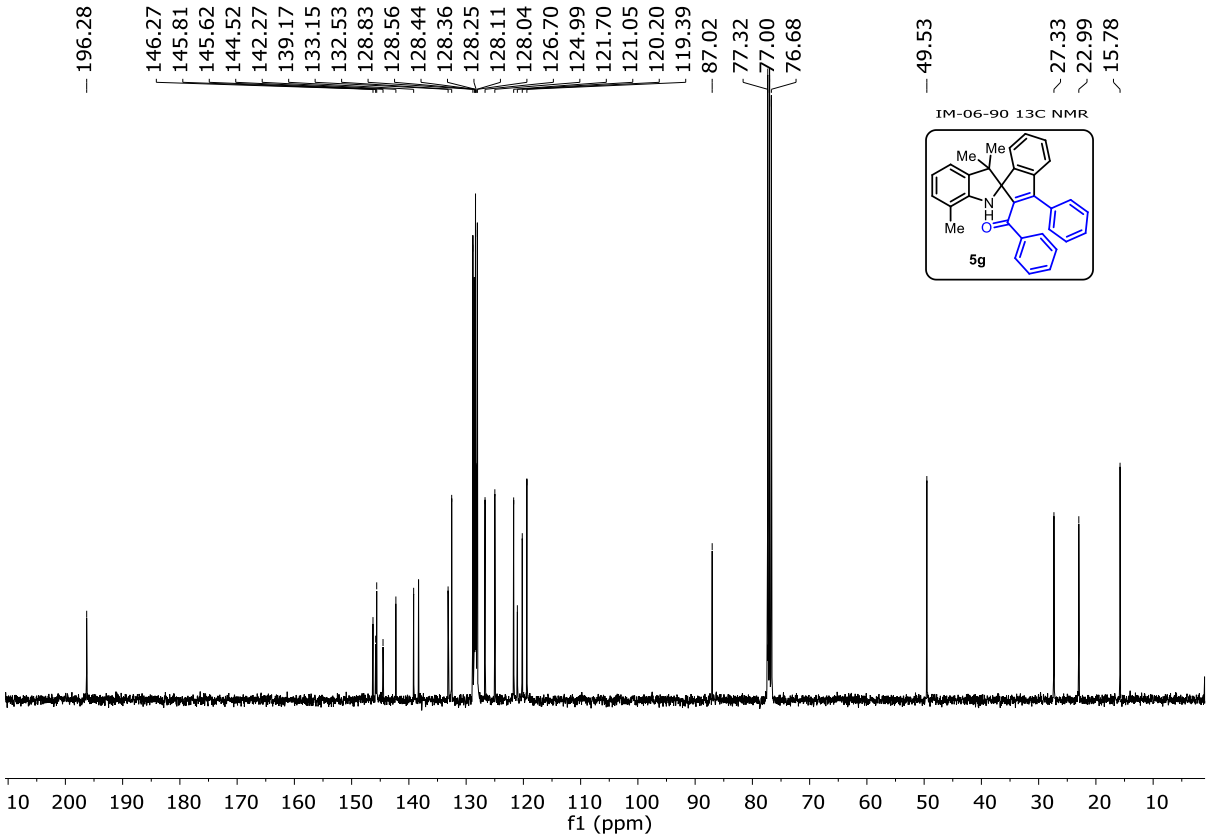
^{19}F NMR of 5f (376 MHz, $CDCl_3$):



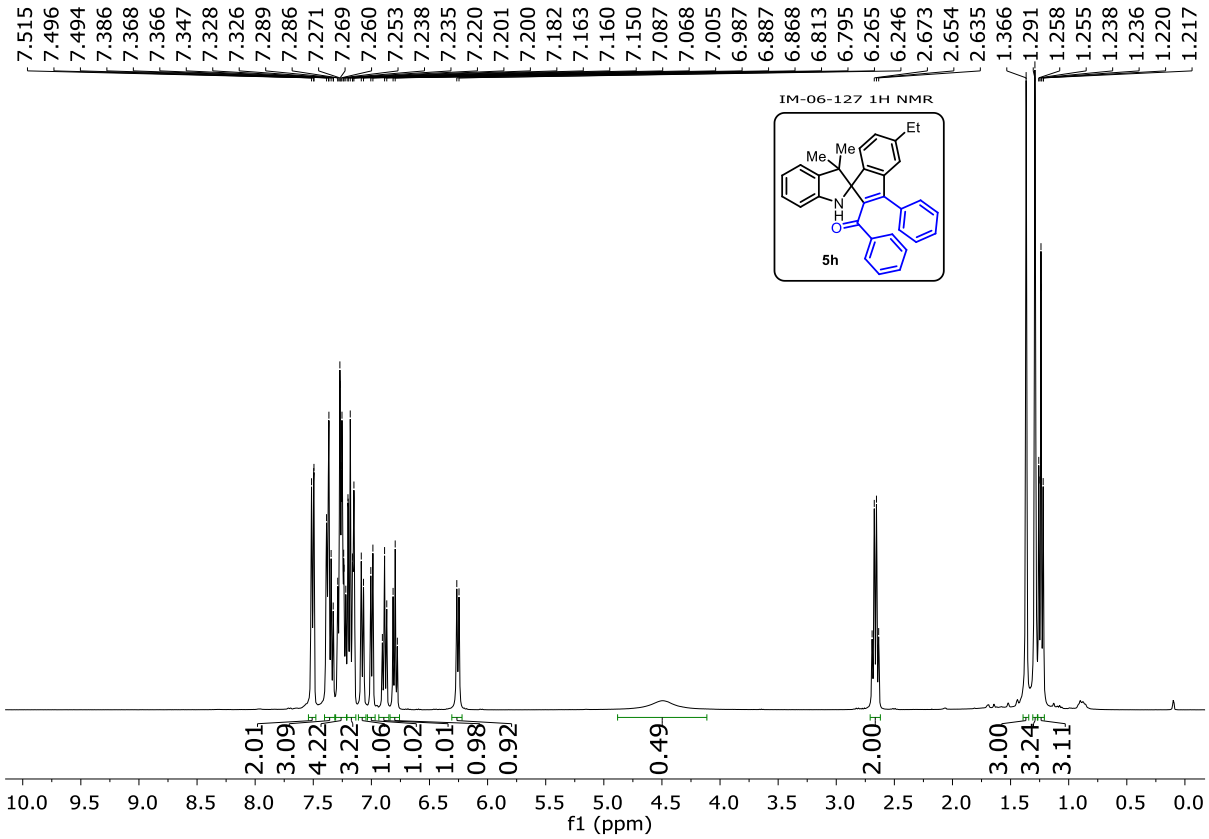
^{1}H NMR of 5g (400 MHz, $CDCl_3$):



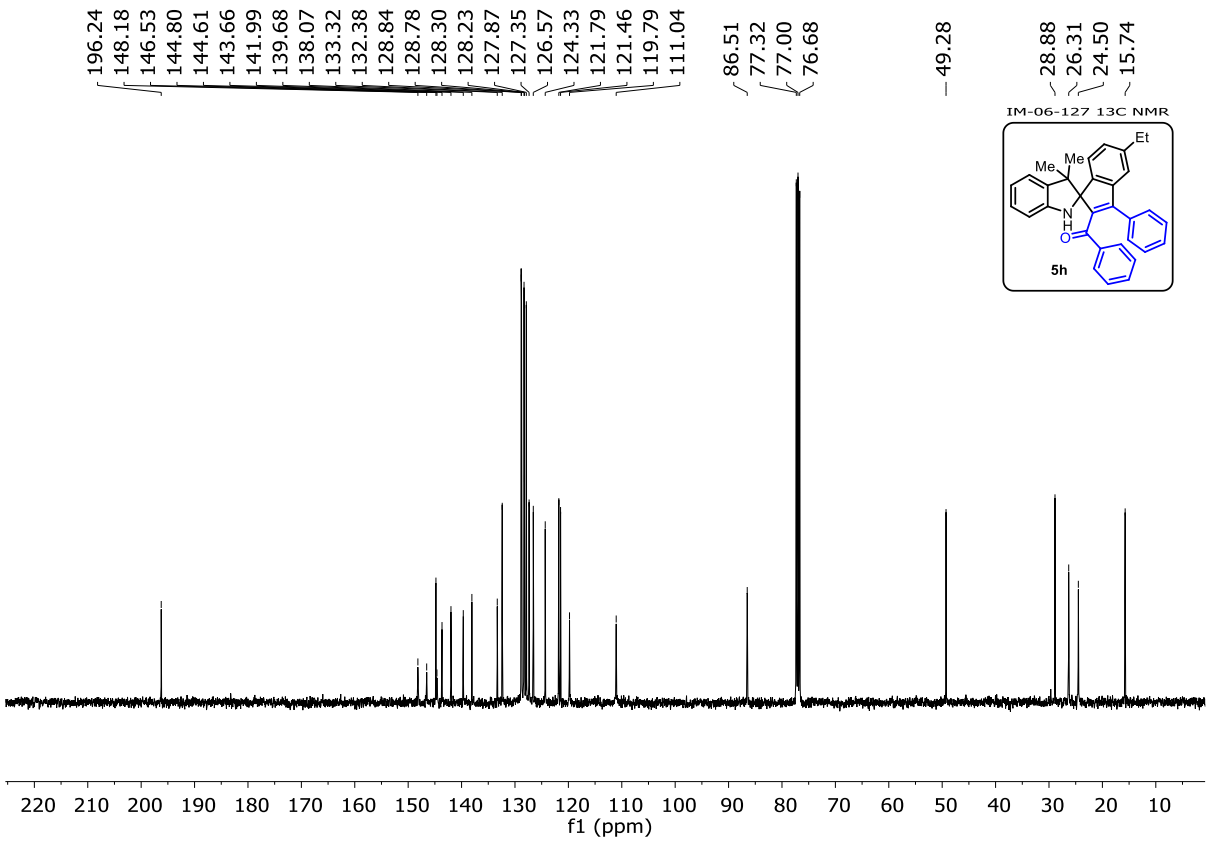
^{13}C{^{1}H} NMR of 5g (101 MHz, $CDCl_3$):



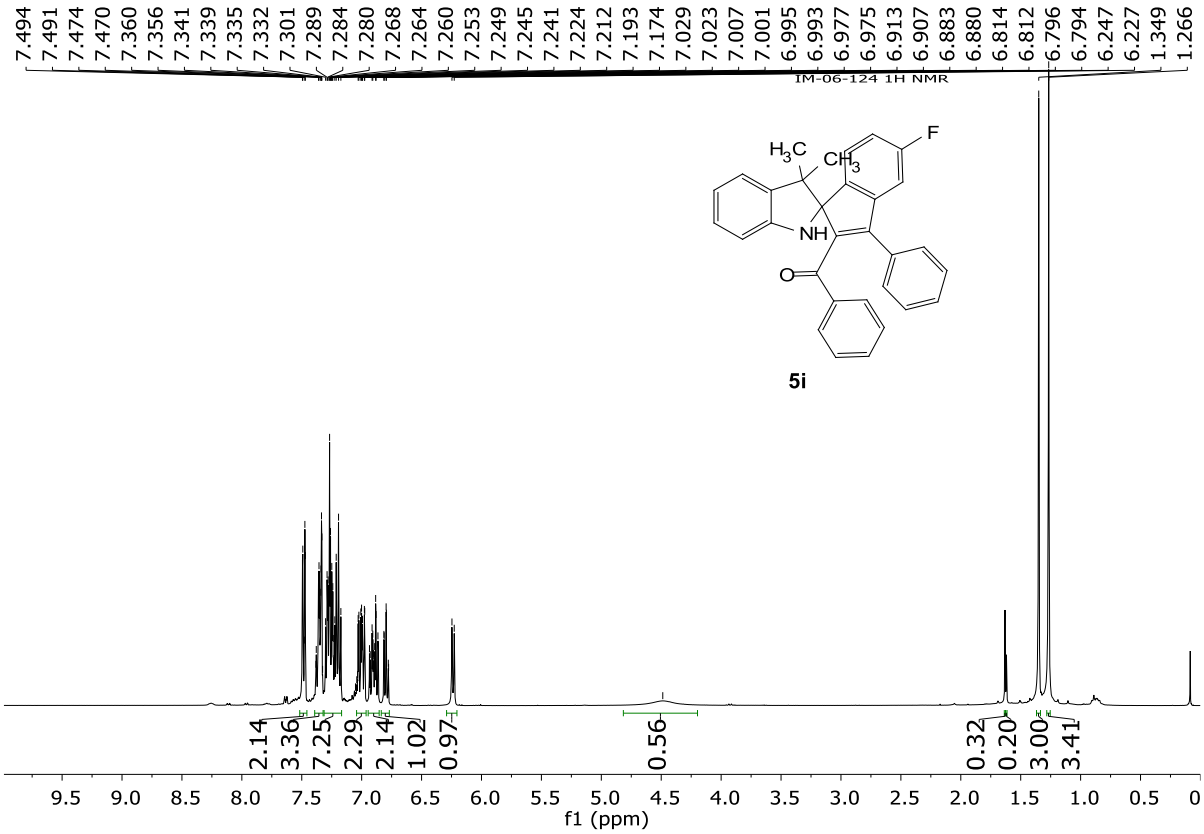
^{1}H NMR of 5h (400 MHz, $CDCl_3$):



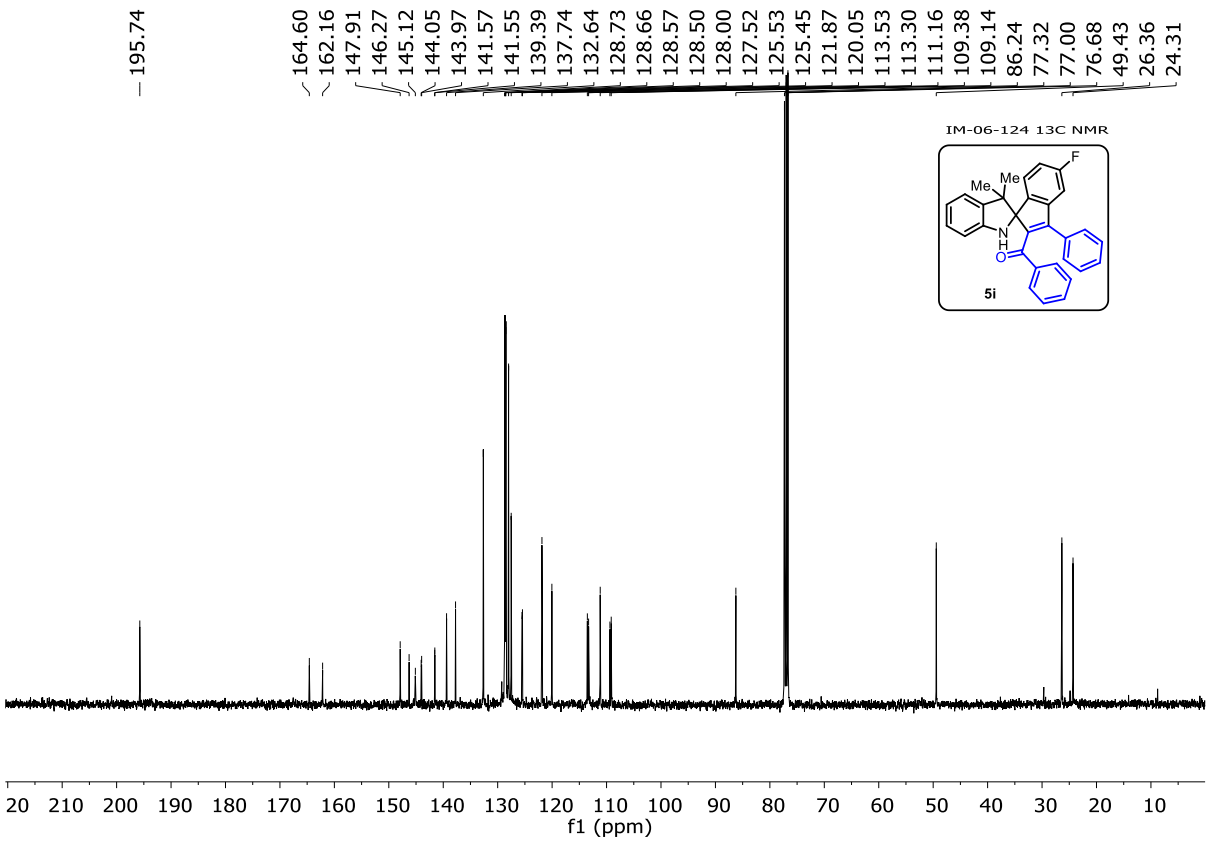
^{13}C{^{1}H} NMR of 5h (101 MHz, $CDCl_3$):



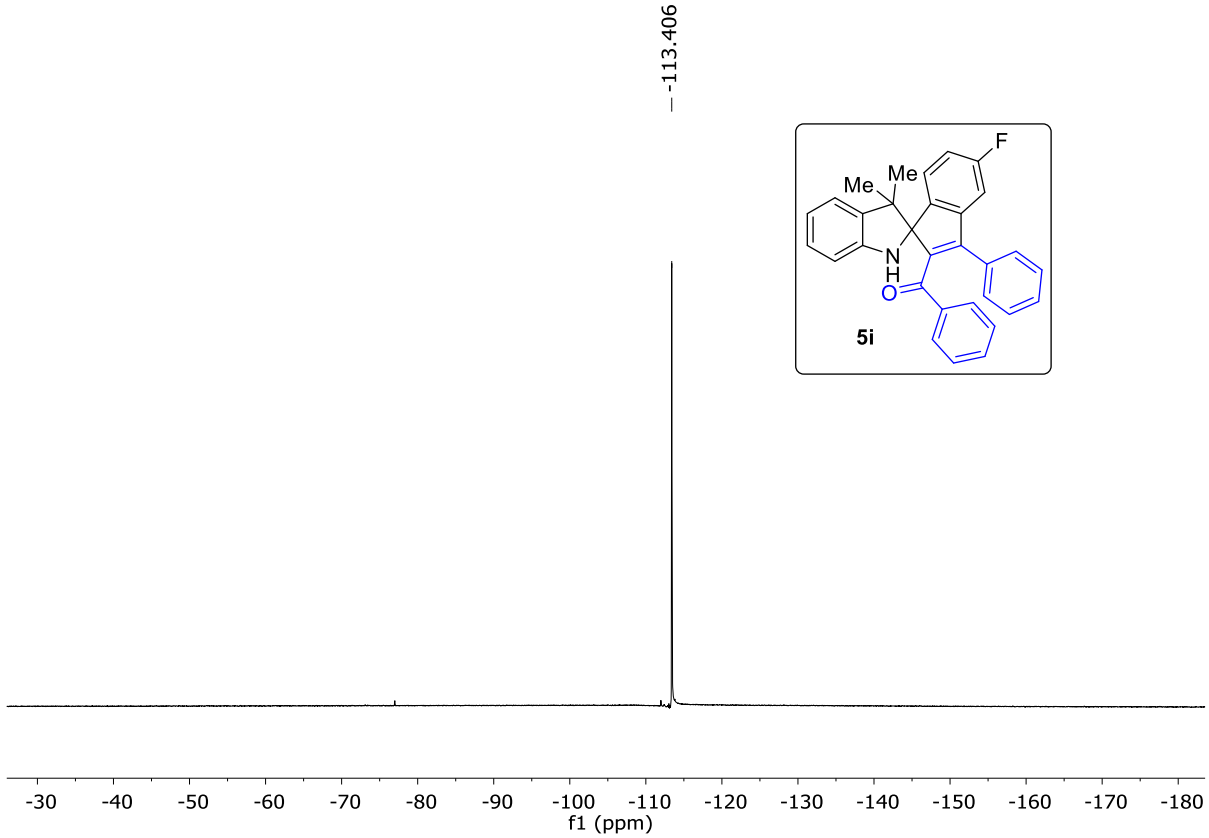
^{1}H NMR of 5i (400 MHz, $CDCl_3$):



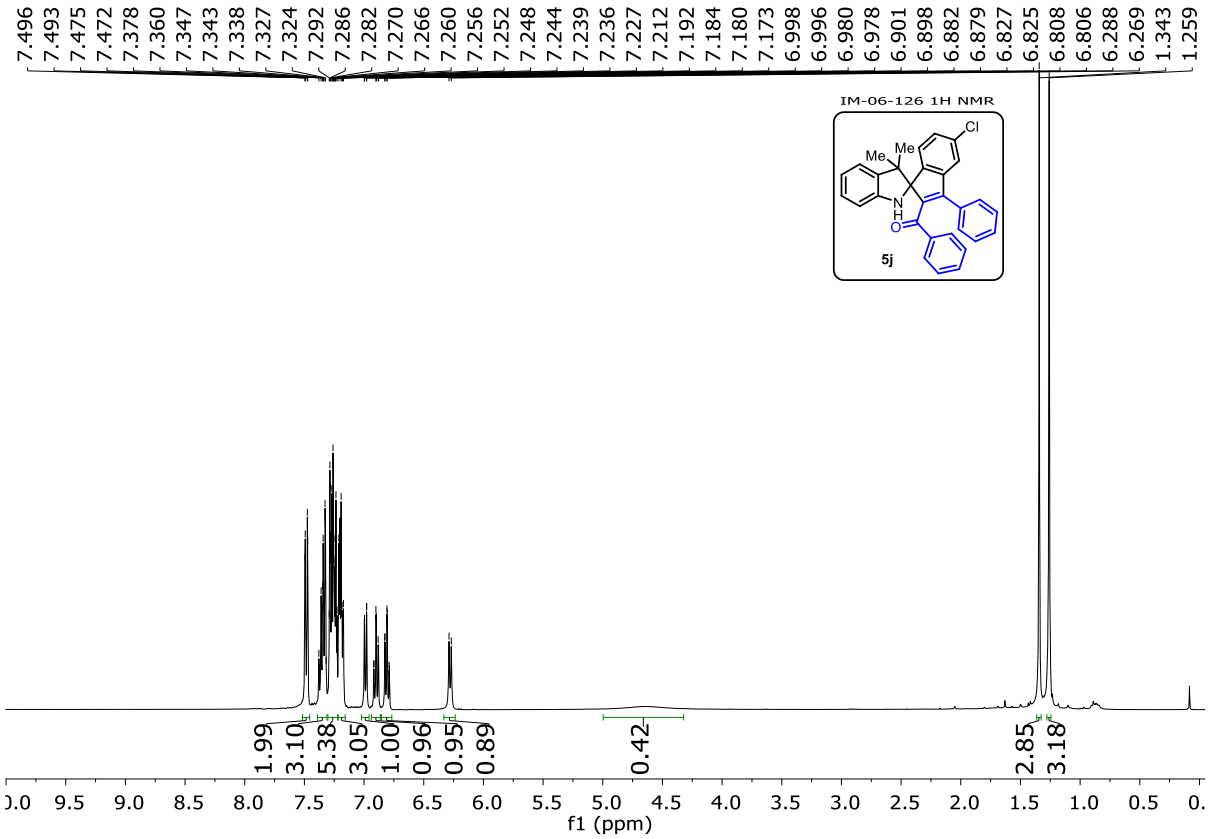
^{13}C{^{1}H} NMR of 5i (101 MHz, $CDCl_3$):



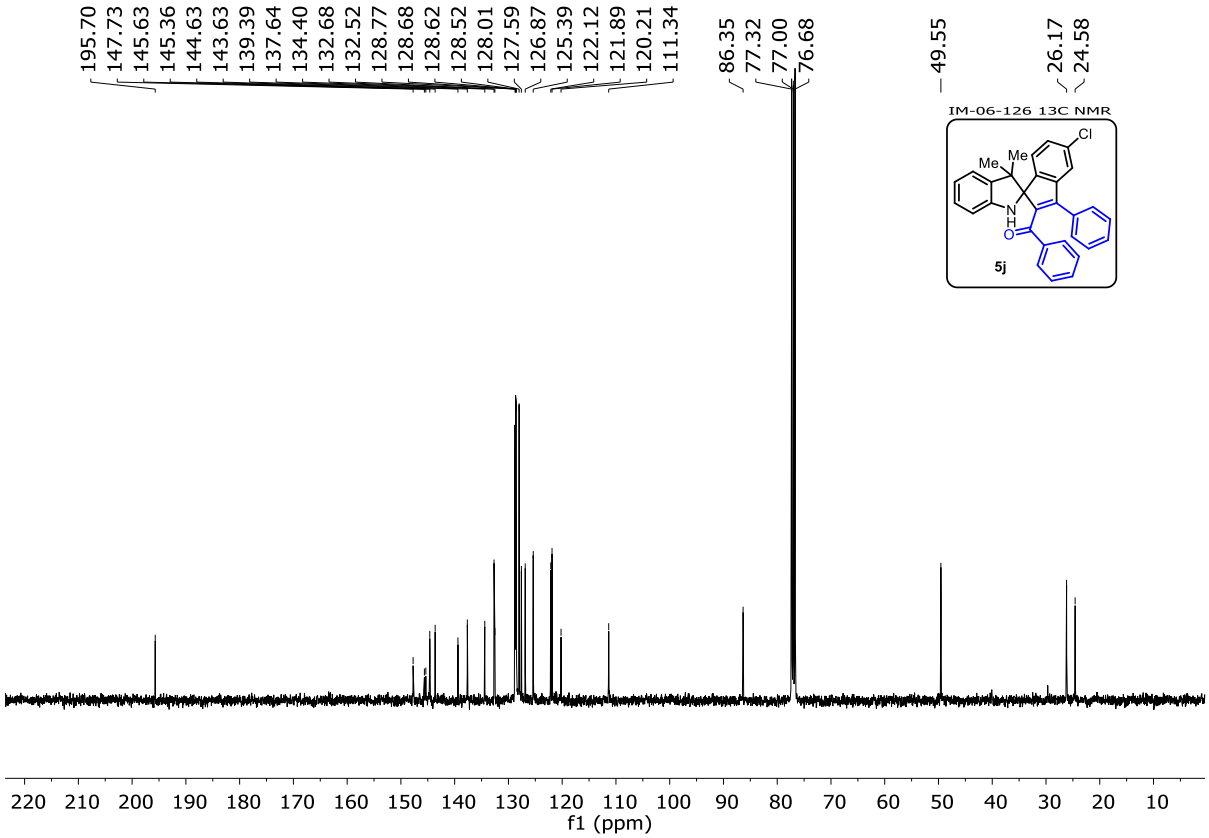
^{19}F NMR of 5i (376 MHz, $CDCl_3$):



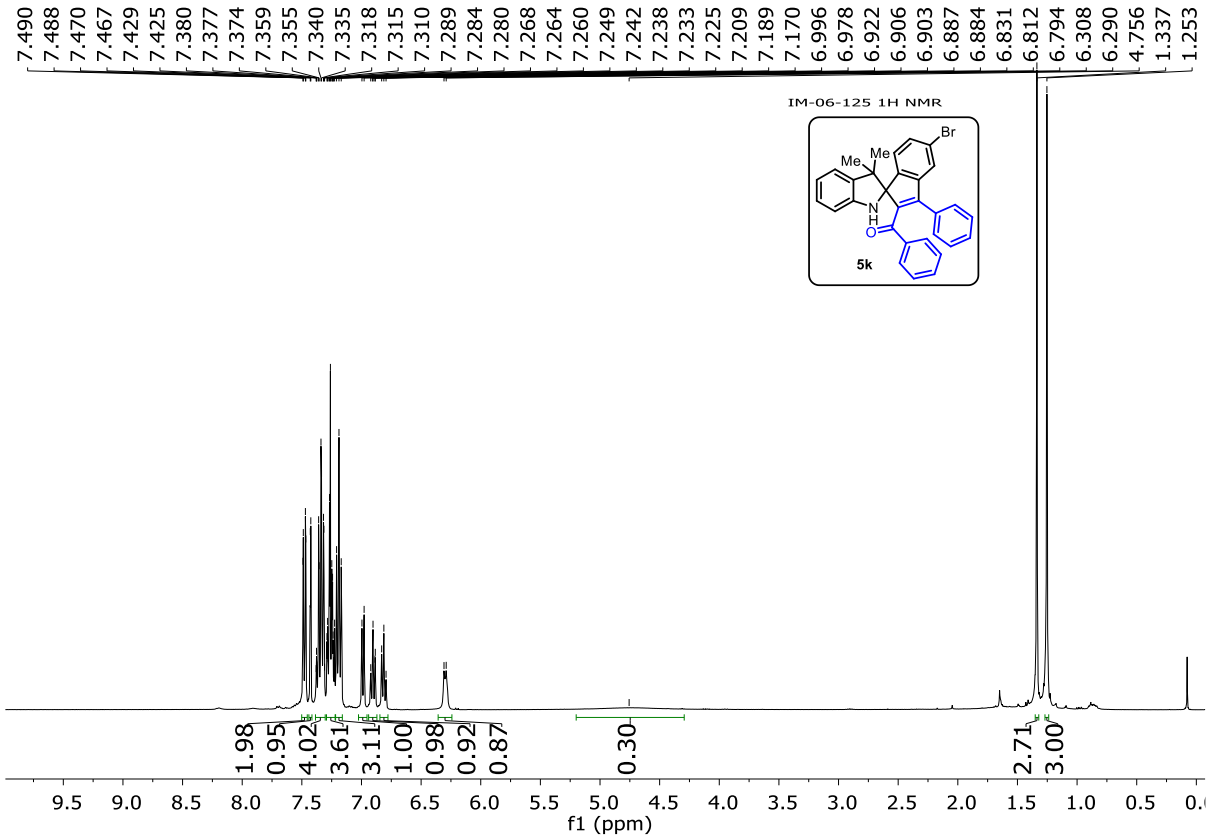
^{1}H NMR of 5j (400 MHz, $CDCl_3$):



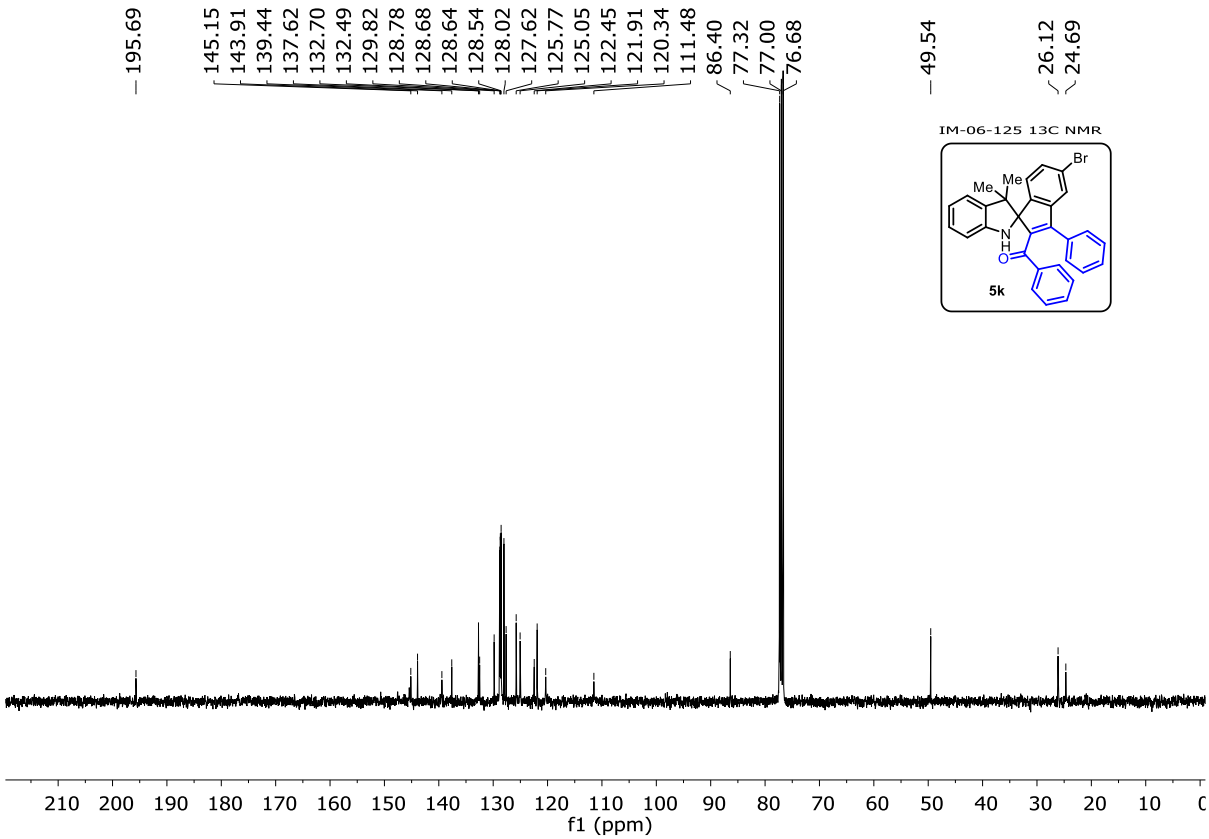
^{13}C{^{1}H} NMR of 5j (101 MHz, $CDCl_3$):



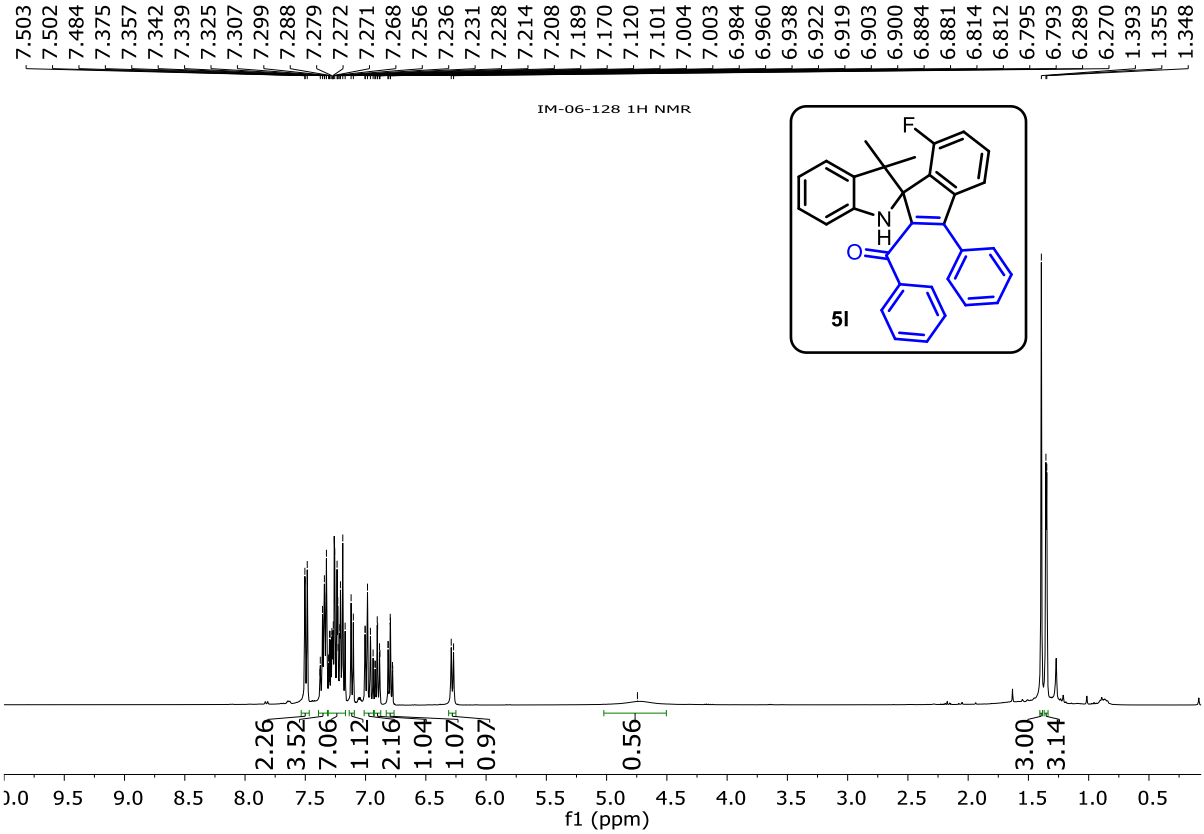
^{1}H NMR of 5k (400 MHz, $CDCl_3$):



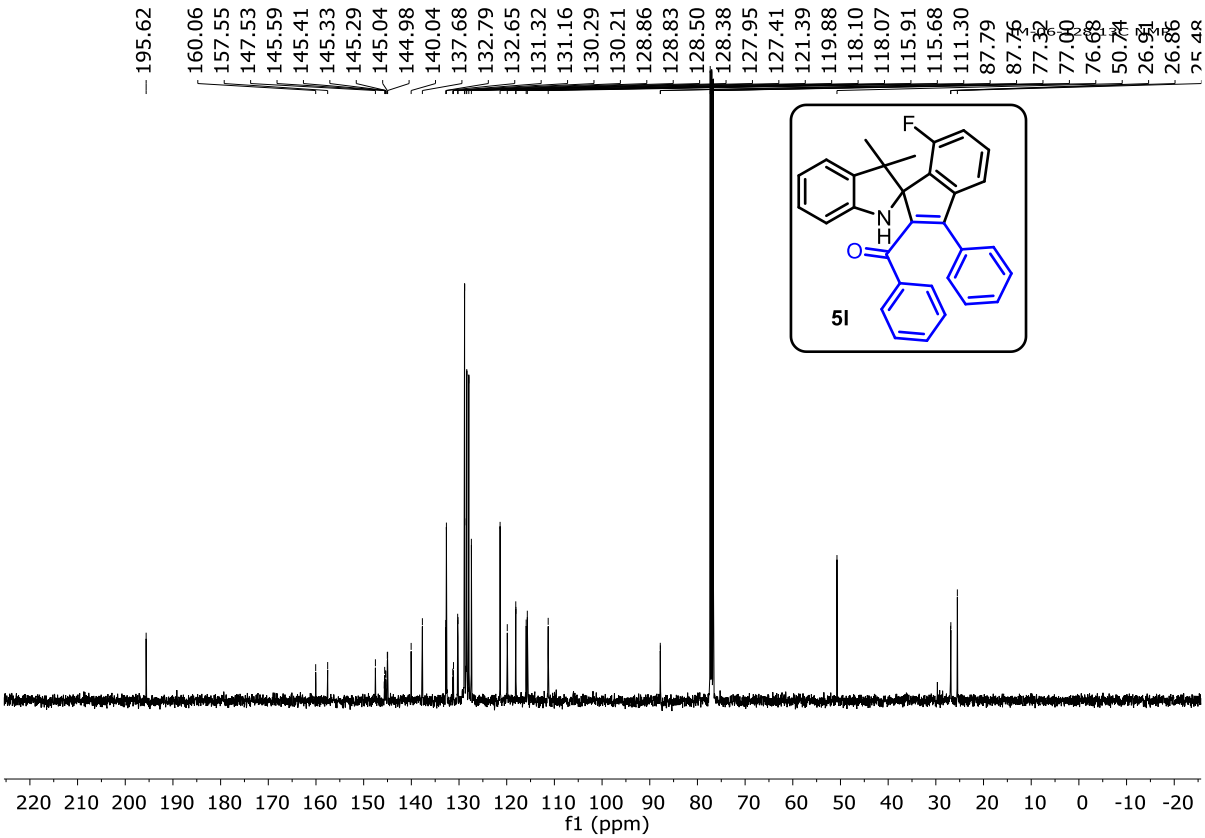
^{13}C{^{1}H} NMR of 5k (101 MHz, $CDCl_3$):



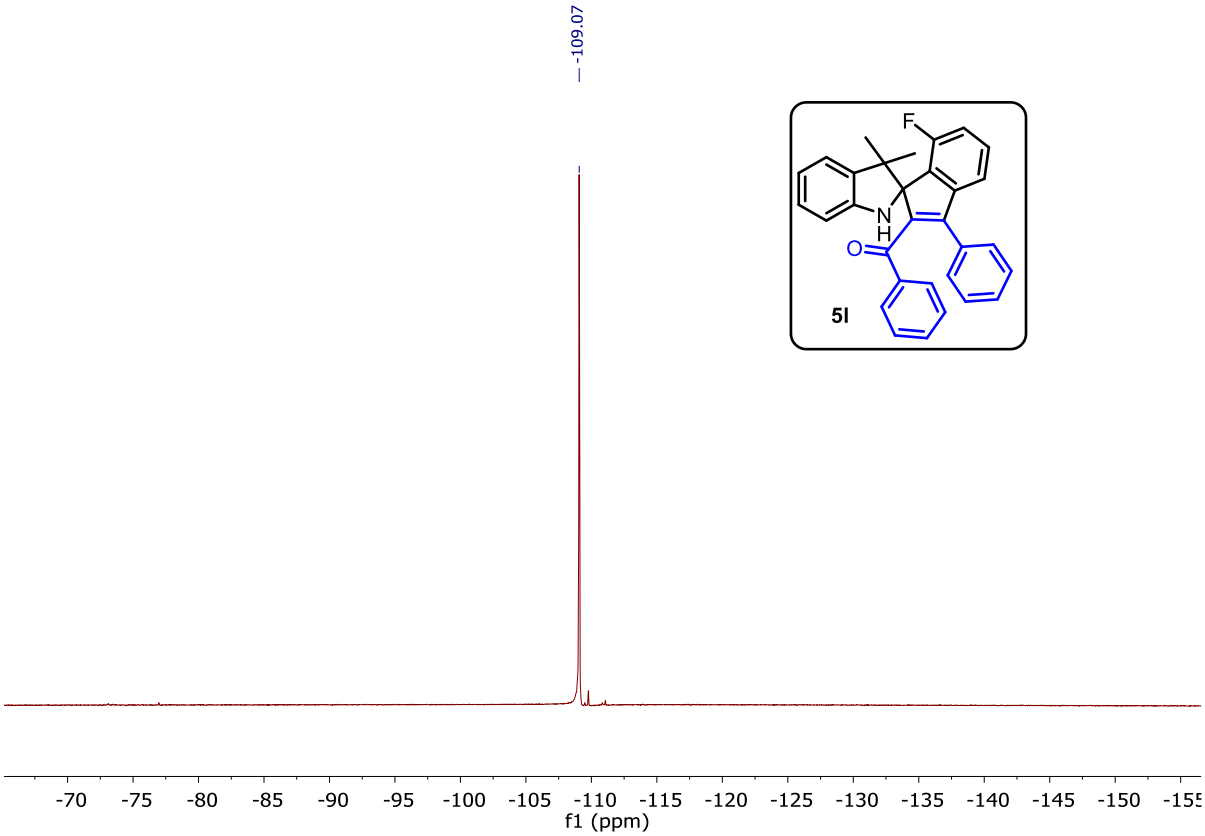
^{1}H NMR of 5l (400 MHz, $CDCl_3$):



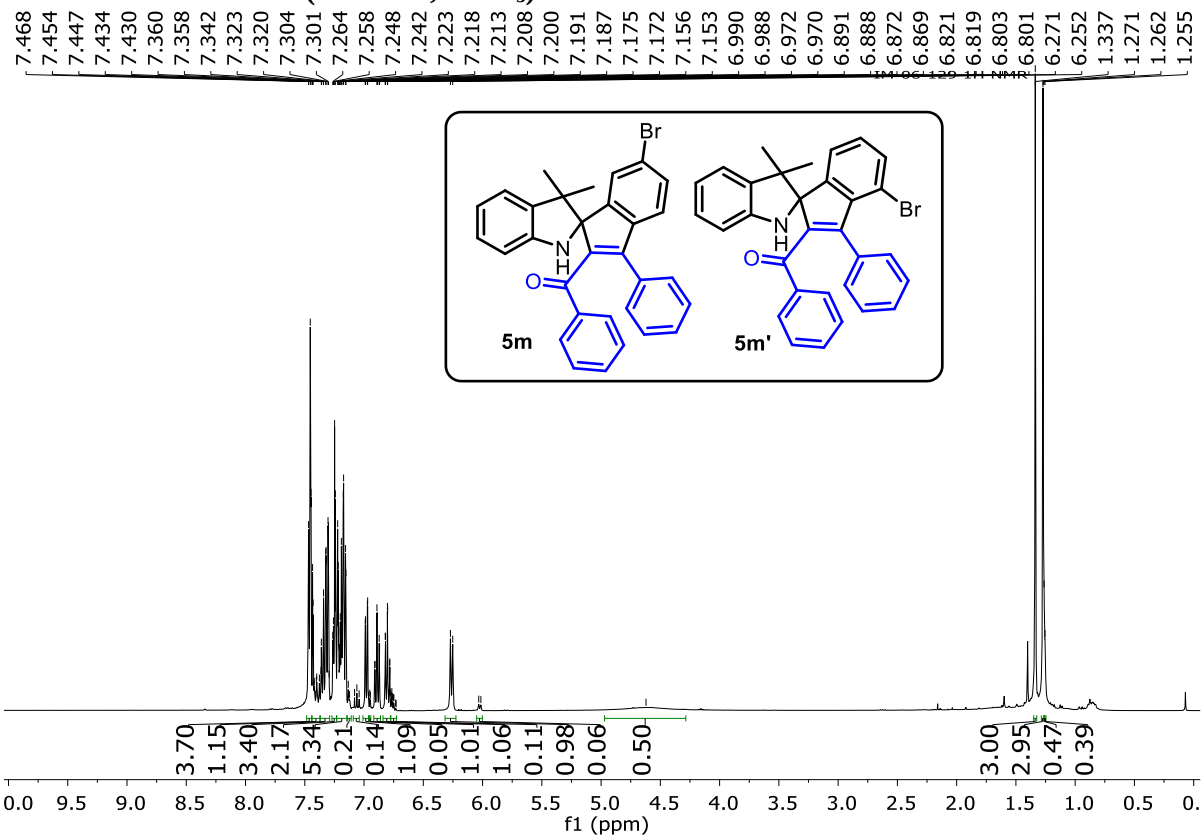
^{13}C{^{1}H} NMR of 5l (101 MHz, $CDCl_3$):



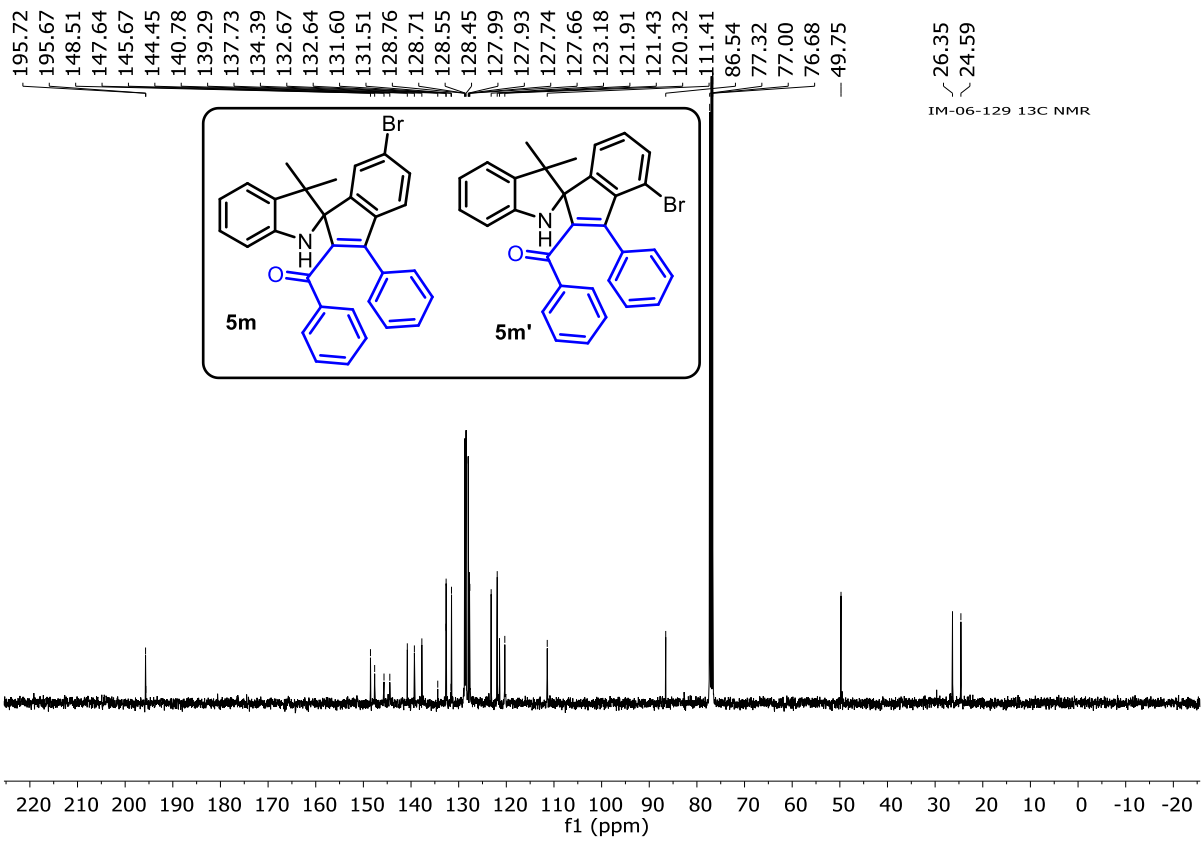
^{19}F NMR of 5l (376 MHz, $CDCl_3$):



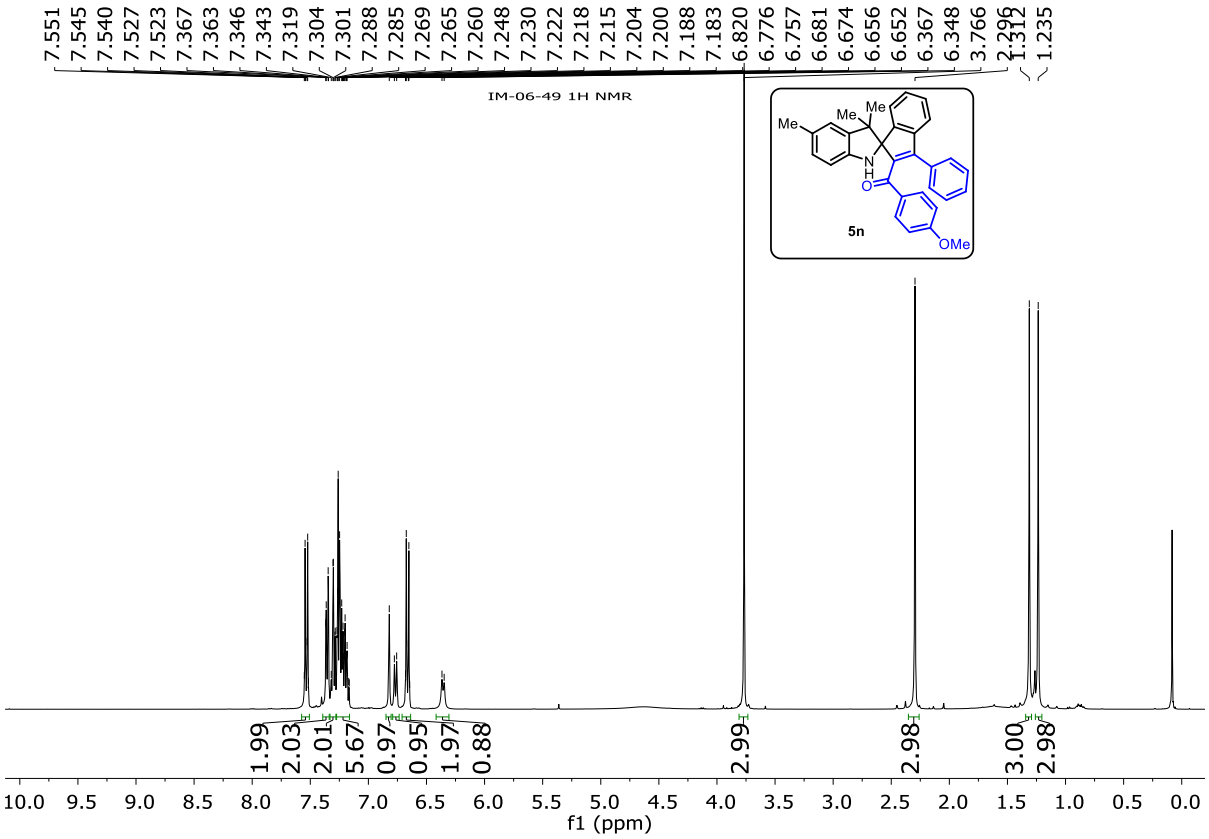
^{1}H NMR of 5m and 5m´ (400 MHz, $CDCl_3$):



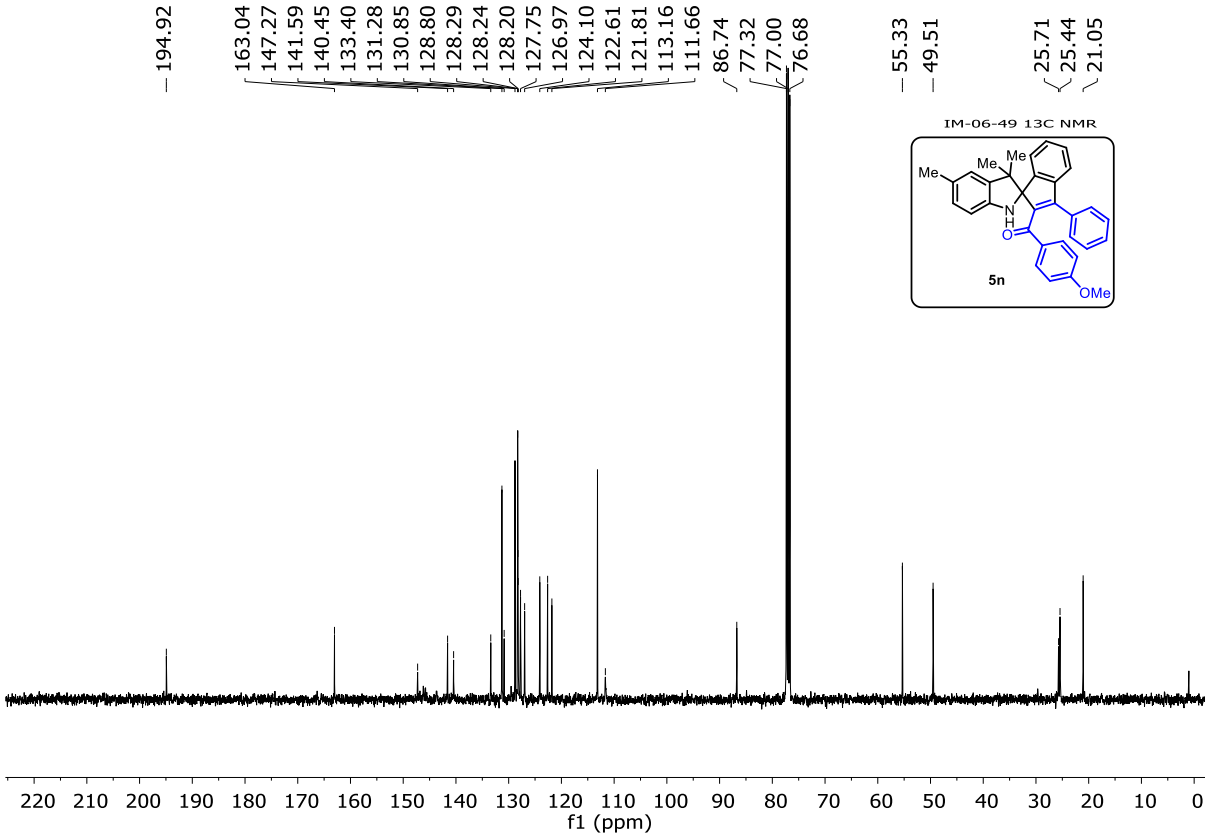
^{13}C{^{1}H} NMR of 5m and 5m´ (101 MHz, $CDCl_3$):



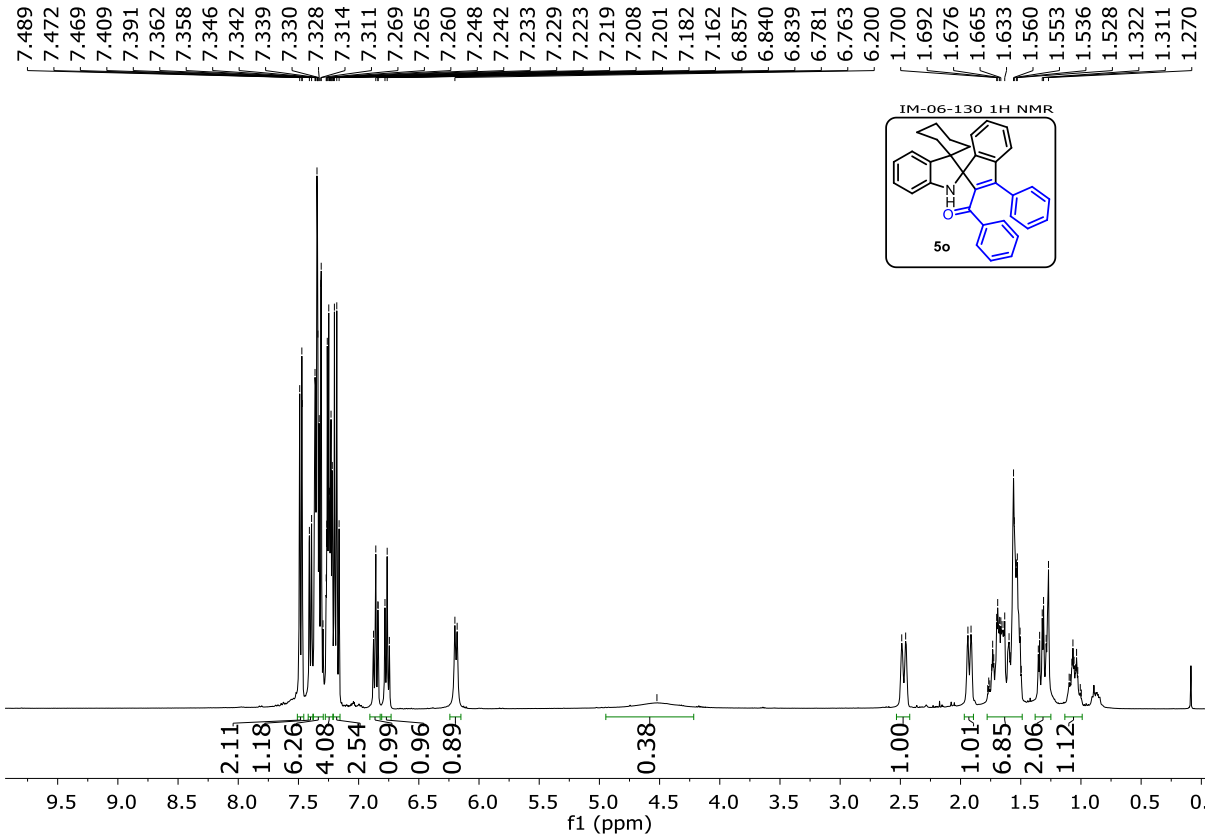
^{1}H NMR of 5n (400 MHz, $CDCl_3$):



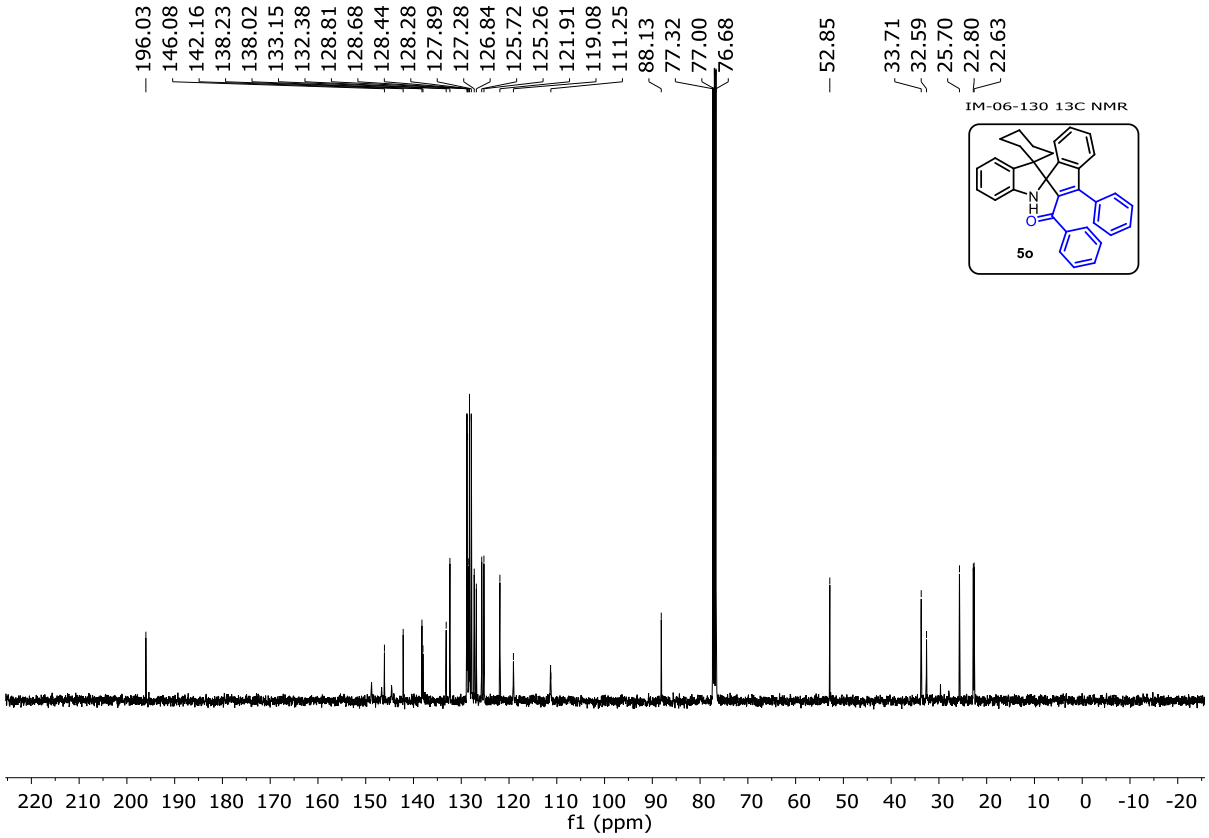
^{13}C{^{1}H} NMR of 5n (101 MHz, $CDCl_3$):



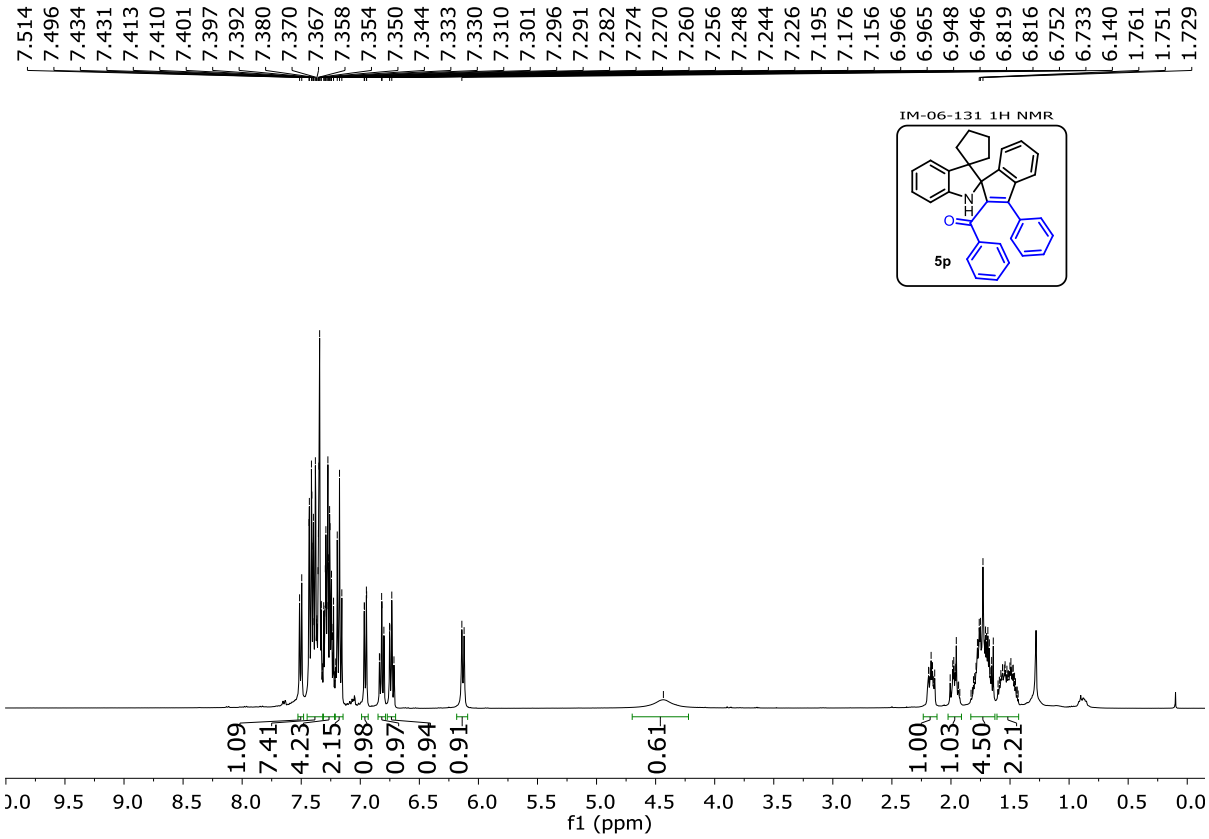
^{1}H NMR of 5o (400 MHz, $CDCl_3$):



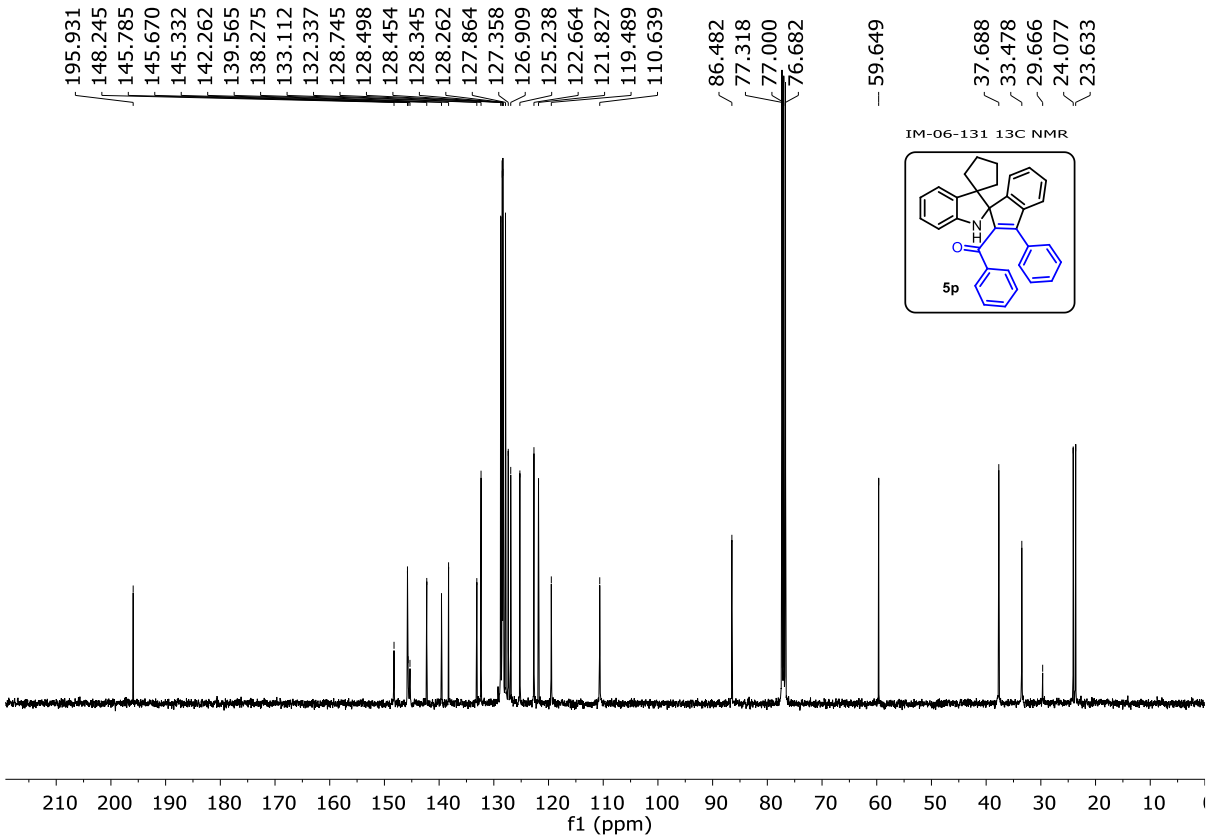
^{13}C{^{1}H} NMR of 5o (101 MHz, $CDCl_3$):



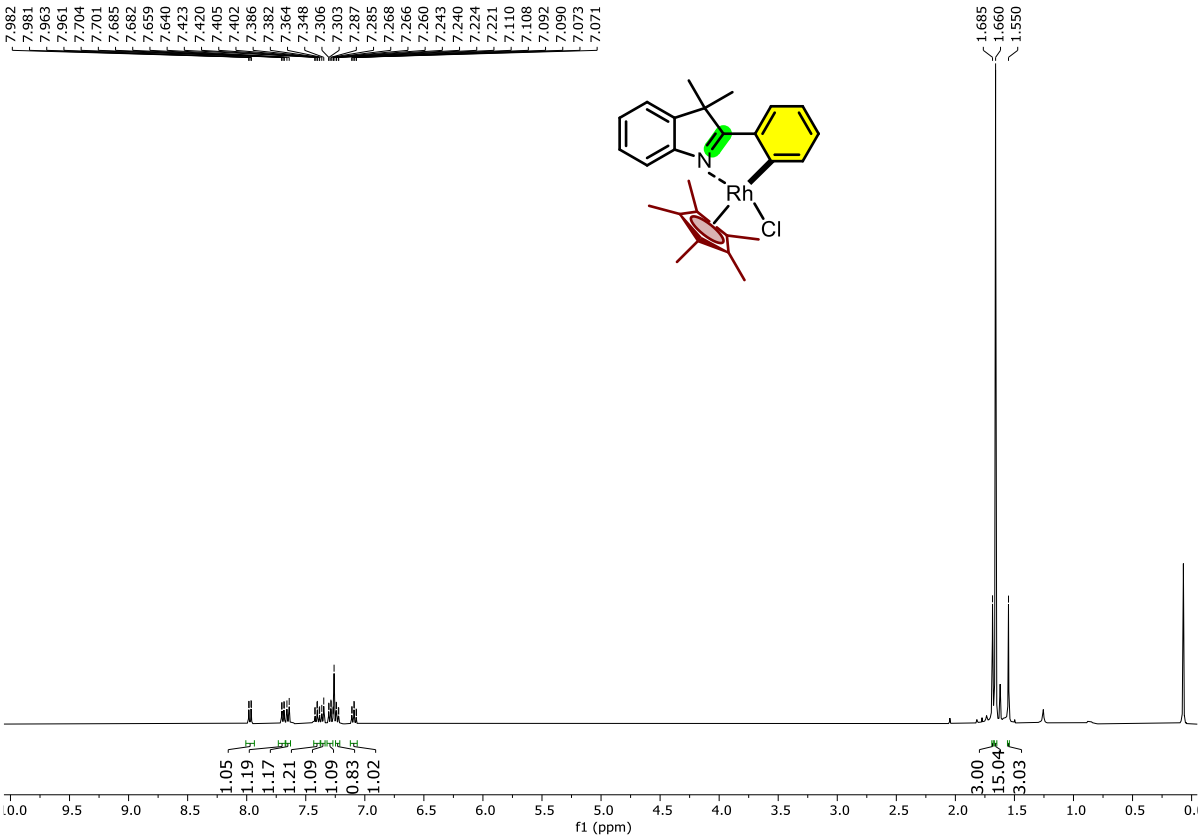
^{1}H NMR of 5p (400 MHz, $CDCl_3$):



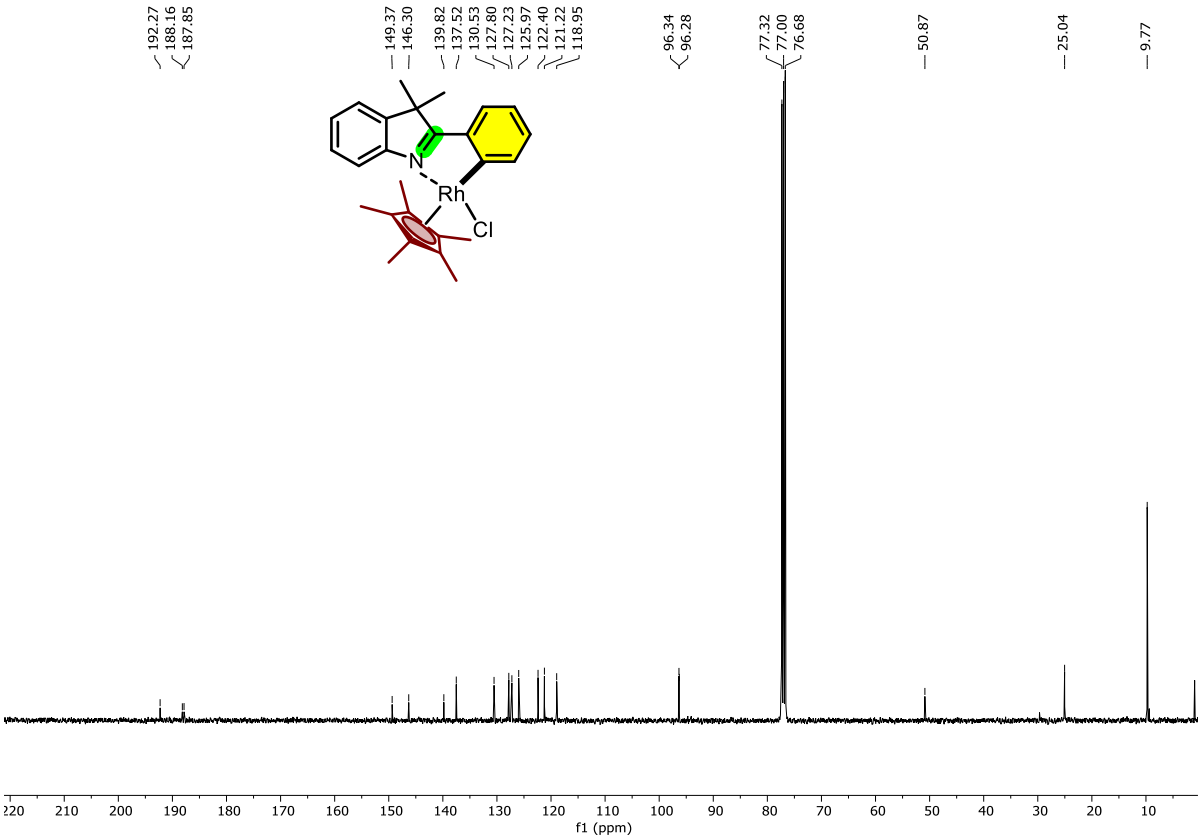
^{13}C{^{1}H} NMR of 5p (101 MHz, $CDCl_3$):



^{1}H NMR of Rh-1a (400 MHz, $CDCl_3$):



^{13}C{^{1}H} NMR of Rh-1a (101 MHz, $CDCl_3$):



****End****