Supporting Information

Condition-Controlled Selective Synthesis of Spiroisoquinolinones or Spiroisoindolinones *via* CHA−Initiated Spiroannulation, Ring Degradation and Ring Contraction

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Table of Contents

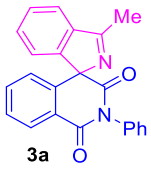
I. General experimental information

Commercial reagents were used without further purification. 3-Aryl-5-isoxazolones **1**[1], diazo homophthalimides **2**[2] were prepared based on literature procedures. Melting points were recorded with a micro melting point apparatus and uncorrected. The ^{1}H NMR spectra were recorded at 400 MHz or 600 MHz. The ^{13}C NMR spectra were recorded at 100 MHz or 150 MHz. The ^{19}F NMR spectra were recorded at 376 MHz or 565 MHz. Chemical shifts were expressed in parts per million (δ), and were reported as s (singlet), d (doublet), t (triplet), dd (doublet of doublets), q (quartet), m (multiplet), etc. The coupling constants *J* were given in Hz. High resolution mass spectra (HRMS) were obtained *via* ESI-TOF mode by using a BRUKER compact mass spectrometer. All reactions were monitored by thin layer chromatography (TLC) using silica gel plates (silica gel 60 F254 0.25 mm), and components were visualized by observation under UV light (254 and 365 nm).

II. Experimental procedures and spectroscopic data

1. Typical procedure for the synthesis of 3a and spectroscopic data of 3a-3ee

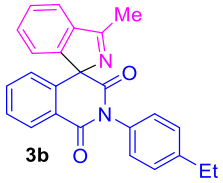
To a reaction tube equipped with a stir bar were charged with 3-phenylisoxazol-5(4*H*)-one (**1a**, 32.3 mg, 0.2 mmol), NaOAc (32.8 mg, 0.4 mmol), $[RhCp^*Cl_2]_2$ (6.2 mg, 0.01 mmol), 4-diazo-2-phenylisoquinoline-1,3(2*H*,4*H*)-dione (**2a**, 63.2 mg, 0.24 mmol) and DCE (2 mL). The tube was then sealed, and the mixture was stirred at 80 °C under argon for 1 h. Upon completion, it was cooled to room temperature, filtered through a pad of celite and concentrated under reduced pressure. The residue was purified by silica gel column chromatography using petroleum ether/ethyl acetate (3:1) as eluent to afford **3a**. Other products **3b-3ee** were obtained in a similar manner.



3a

3-Methyl-2'-phenyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione[3] (3a)

Eluent: petroleum ether/ethyl acetate (3:1). White solid (50.7 mg, 72%), mp 160.2-161.1 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.38-8.36 (m, 1H), 7.61 (d, *J* = 7.6 Hz, 1H), 7.54-7.38 (m, 7H), 7.34 (d, *J* = 7.6 Hz, 1H), 7.22-7.20 (m, 2H), 6.77-6.74 (m, 1H), 2.69 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 176.4, 168.5, 164.6, 153.2, 139.9, 136.4, 135.1, 134.4, 130.1, 129.7, 129.2, 129.0, 128.7, 128.4, 125.45, 125.40, 122.3, 121.7, 82.6, 16.9. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{23}H_{17}N_2O_2$ 353.1285; Found 353.1290.

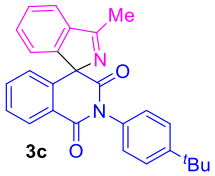


3b

2'-(4-Ethylphenyl)-3-methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3b)

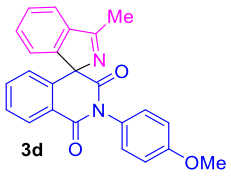
Eluent: petroleum ether/ethyl acetate (3:1). White solid (54.0 mg, 71%), mp 211.6-212.0 °C. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.37-8.35 (m, 1H), 7.61 (d, *J* = 7.6 Hz, 1H), 7.51-7.46 (m, 3H), 7.40 (td, J_1 = 7.6 Hz, J_2 = 1.2

Hz, 1H), 7.33 (d, *J* = 7.2 Hz, 1H), 7.27 (d, *J* = 10.8 Hz, 2H), 7.11 (d, *J* = 8.0 Hz, 2H), 6.76-6.74 (m, 1H), 2.70-2.65 (m, 5H), 1.24 (t, *J* = 7.6 Hz, 3H). $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$): δ 176.3, 168.5, 164.7, 153.3, 144.8, 139.9, 136.3, 134.3, 132.5, 130.1, 129.7, 129.2, 128.9, 128.7, 128.1, 125.5, 125.4, 122.3, 121.7, 82.6, 28.6, 16.9, 15.3. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{25}H_{21}N_2O_2$ 381.1598; Found 381.1607.



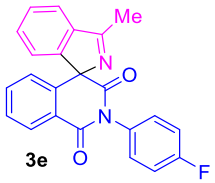
2'-(4-*(tert*-Butyl)phenyl)-3-methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione[3] (3c)

Eluent: petroleum ether/ethyl acetate (3:1). White solid (51.5 mg, 63%), mp 216.7-217.4 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.36 (dd, J_1 = 7.6 Hz, J_2 = 1.6 Hz, 1H), 7.60 (d, *J* = 7.6 Hz, 1H), 7.51-7.45 (m, 5H), 7.40 (t, *J* = 7.2 Hz, 1H), 7.33 (d, *J* = 7.2 Hz, 1H), 7.12 (d, *J* = 8.0 Hz, 2H), 6.77-6.75 (m, 1H), 2.68 (s, 3H), 1.31 (s, 9H). $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$): δ 176.3, 168.6, 164.7, 153.2, 151.5, 139.9, 136.3, 134.3, 132.3, 130.1, 129.7, 129.2, 129.0, 127.7, 126.3, 125.5, 125.4, 122.2, 121.7, 82.6, 34.7, 31.3, 16.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{27}H_{25}N_2O_2$ 409.1911; Found 409.1910



2'-(4-Methoxyphenyl)-3-methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione[3] (3d)

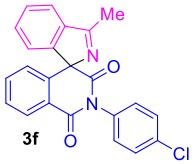
Eluent: petroleum ether/ethyl acetate (3:1). White solid (59.7 mg, 78%), mp 104.9-105.6 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.37-8.35 (m, 1H), 7.61 (d, *J* = 7.6 Hz, 1H), 7.50-7.46 (m, 3H), 7.40 (td, J_1 = 7.2 Hz, J_2 = 0.4 Hz, 1H), 7.32 (d, *J* = 7.6 Hz, 1H), 7.11 (d, *J* = 8.8 Hz, 2H), 6.96 (d, *J* = 9.2 Hz, 2H), 6.75-6.73 (m, 1H), 3.80 (s, 3H), 2.68 (s, 3H). $^{13}C\{^{1}H\}$ NMR (100 MHz, $CDCl_3$): δ 176.3, 168.6, 164.8, 159.6, 153.3, 139.9, 136.3, 134.3, 130.1, 129.7, 129.3, 129.2, 129.0, 127.6 125.5, 125.4, 122.3, 121.7, 114.6, 82.6, 55.5, 16.9. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{24}H_{19}N_2O_3$ 383.1390; Found 383.1399.



3e

2'-(4-Fluorophenyl)-3-methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione[3] (3e)

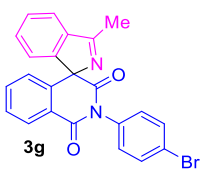
Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (58.5 mg, 79%), mp 217.8-218.5 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.37-8.34 (m, 1H), 7.62 (d, J = 7.6 Hz, 1H), 7.52-7.46 (m, 3H), 7.41 (td, J_1 = 7.2 Hz, J_2 = 0.8 Hz, 1H), 7.31 (d, J = 7.6 Hz, 1H), 7.20-7.10 (m, 4H), 6.75-6.73 (m. 1H), 2.68 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 176.4, 168.6, 164.6, 162.5 (d, $^{1}J_{C\text{-}F}$ = 245.8 Hz), 153.1, 140.0, 136.3, 134.5, 130.8 (d, $^{4}J_{C\text{-}F}$ = 2.6 Hz), 130.21 (d, $^{3}J_{C\text{-}F}$ = 8.3 Hz), 130.18, 129.7, 129.3, 129.1, 125.5, 125.3, 122.4, 121.7, 116.3 (d, $^{2}J_{C\text{-}F}$ = 23.4 Hz), 82.6, 16.9. ^{19}F NMR (376 MHz, $CDCl_3$): δ -112.84 – -112.91 (m). HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{23}H_{16}FN_2O_2$ 371.1190; Found 371.1193.



3f

2'-(4-Chlorophenyl)-3-methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione[3] (3f)

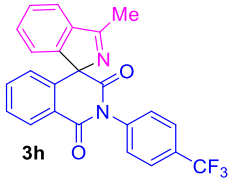
Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (54.2 mg, 70%), mp 277.2-278.1 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.37-8.34 (m, 1H), 7.62 (d, J = 7.2 Hz, 1H), 7.52-7.47 (m, 3H), 7.43-7.40 (m, 3H), 7.31 (d, J = 7.2 Hz, 1H), 7.15 (d, J = 8.8 Hz, 2H), 6.75-6.73 (m, 1H), 2.68 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 176.4, 168.4, 164.4, 153.0, 140.0, 136.3, 134.7, 134.5, 133.5, 130.2, 129.8, 129.7, 129.5, 129.3 129.1, 125.5, 125.3, 122.4, 121.7, 82.6, 16.9. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{23}H_{16}ClN_2O_2$ 387.0895; Found 387.0893.



3g

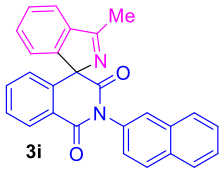
2'-(4-Bromophenyl)-3-methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione[3] (3g)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (52.6 mg, 61%), mp 191.2-192.0 ºC. ^{1}H NMR (600 MHz, $CDCl_3$): δ 8.35 (dd, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H), 7.62 (d, J = 7.8 Hz, 1H), 7.57 (d, J = 8.4 Hz, 2H), 7.53-7.47 (m, 3H), 7.41 (td, J_1 = 7.2 Hz, J_2 = 0.6 Hz, 1H), 7.31 (d, J = 7.8 Hz, 1H), 7.09 (d, J = 8.4 Hz, 2H), 6.74 (dd, J_1 = 7.8 Hz, J_2 = 1.2 Hz, 1H), 2.68 (s, 3H). ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 176.4, 168.3, 164.4, 153.0, 140.0, 136.3, 134.6, 134.1, 132.5, 130.2, 129.7, 129.4, 129.1, 125.5, 125.2, 122.8, 122.4, 121.7, 82.6, 16.9. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{23}H_{16}BrN_2O_2$ 431.0390; Found 431.0392.



3-Methyl-2'-(4-(trifluoromethyl)phenyl)-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione[3] (3h)

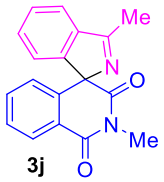
Eluent: petroleum ether/ethyl acetate (3:1). White solid (50.4 mg, 60%), mp 191.2-192.7 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.37-8.35 (m, 1H), 7.72 (d, J = 8.4 Hz, 2H), 7.63 (d, J = 7.6 Hz, 1H), 7.53-7.48 (m, 3H), 7.43 (td, J_1 = 7.2 Hz, J_2 = 0.8 Hz, 1H), 7.34 (t, J_1 = 7.2 Hz, 3H), 6.77-6.74 (m. 1H), 2.68 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 176.5, 168.4, 164.3, 152.9, 140.0, 138.3, 136.3, 134.7, 130.9 (q, $^{2}J_{C-F}$ = 32.9 Hz), 130.3, 129.8, 129.4, 129.2, 129.1, 126.4 (q, $^{3}J_{C-F}$ = 3.5 Hz), 125.5, 125.1, 122.4, 121.7, 82.6, 16.9. ^{19}F NMR (376 MHz, $CDCl_3$): δ -62.68 (s). HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{24}H_{16}F_3N_2O_2$ 421.1158; Found 421.1160.



3-Methyl-2'-(naphthalen-2-yl)-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione[3] (3i)

Eluent: petroleum ether/ethyl acetate (3:1). White solid (41.1 mg, 51%), mp 247.8-248.2 ºC. ^{1}H NMR (600 MHz, $CDCl_3$): δ 8.40 (dd, J_1 = 7.8 Hz, J_2 = 1.2 Hz, 1H), 7.92 (d, J = 9.0 Hz, 1H), 7.87 (d, J = 8.4 Hz, 1H), 7.82 (d, J = 7.8 Hz, 1H), 7.73 (s, 1H), 7.62 (d, J = 7.8 Hz, 1H), 7.53-7.48 (m, 5H), 7.44 (t, J = 6.6 Hz, 1H), 7.40 (d, J = 7.8 Hz, 1H), 7.30-7.29 (m, 1H), 6.78-6.77 (m, 1H), 2.69 (s, 3H). ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 176.4,

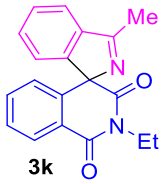
168.6, 164.8, 153.2, 140.0, 136.4, 134.4, 133.5, 133.1, 132.5, 130.2, 129.8, 129.3, 129.2, 129.0, 128.2, 127.8, 127.5, 126.8, 126.4, 125.9, 125.51, 125.46, 122.3, 121.8, 82.7, 16.9. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{27}H_{19}N_2O_2$ 403.1441; Found 403.1442.



3j

2',3-Dimethyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3j)

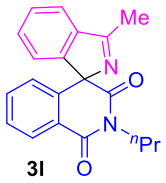
Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (30.8 mg, 53%), mp 135.7-136.2 ºC. ^{1}H NMR (600 MHz, $CDCl_3$): δ 8.34 (dd, J_1 = 7.8 Hz, J_2 = 1.2 Hz, 1H), 7.62 (d, J = 7.2 Hz, 1H), 7.49-7.46 (m, 2H), 7.41 (td, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H), 7.36 (td, J_1 = 7.2 Hz, J_2 = 0.6 Hz, 1H), 7.15 (d, J = 7.8 Hz, 1H), 6.60 (dd, J_1 = 7.8 Hz, J_2 = 0.6 Hz, 1H), 3.41 (s, 3H), 2.68 (s, 3H). ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 175.9, 168.8, 164.6, 153.5, 140.2, 136.1, 134.0, 130.1, 129.3, 129.1, 128.9, 125.5, 125.3, 122.1, 121.9, 82.1, 27.8, 16.9. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{18}H_{15}N_2O_2$ 291.1128; Found 291.1123.



3k

2'-Ethyl-3-methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3k)

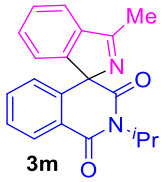
Eluent: petroleum ether/ethyl acetate (3:1). Purple solid (47.5 mg, 78%). ^{1}H NMR (600 MHz, $CDCl_3$): δ 8.33 (dd, J_1 = 7.8 Hz, J_2 = 1.2 Hz, 1H), 7.61 (d, J = 7.2 Hz, 1H), 7.48 (t, J = 7.2 Hz, 2H), 7.41 (td, J_1 = 7.8 Hz, J_2 = 1.2 Hz, 1H), 7.35 (t, J = 7.2 Hz, 1H), 7.13 (d, J = 7.8 Hz, 1H), 6.63 (d, J = 8.4 Hz, 1H), 4.14-4.01 (m, 2H), 2.68 (s, 3H), 1.22 (t, J = 7.2 Hz, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 176.1, 168.3, 164.1, 153.5, 139.9, 136.1, 134.0, 130.0, 129.3, 129.1, 128.8, 125.32, 125.30, 122.1, 121.6, 82.0, 36.3, 16.9, 13.1. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{19}H_{17}N_2O_2$ 305.1285; Found 305.1289.



3l

3-Methyl-2'-propyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3l)

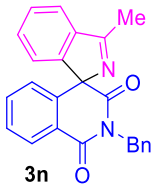
Eluent: petroleum ether/ethyl acetate (3:1). White solid (37.6 mg, 59%), mp 102.5-103.1 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.33 (dd, J_1 = 8.0 Hz, J_2 = 1.2 Hz, 1H), 7.61 (d, J = 7.6 Hz, 1H), 7.49-7.45 (m, 2H), 7.41 (td, J_1 = 7.6 Hz, J_2 = 1.2 Hz, 1H), 7.35 (td, J_1 = 7.6 Hz, J_2 = 0.8 Hz, 1H), 7.13 (d, J = 7.6 Hz, 1H), 6.64-6.62 (m, 1H), 4.04-3.91 (m, 2H), 2.68 (s, 3H), 1.69-1,61 (m, 2H), 0.92 (t, J = 7.6 Hz, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 176.0, 168.5, 164.4, 153.6, 139.9, 136.1, 133.9, 130.0, 129.3, 129.1, 128.8, 125.3, 122.1, 121.6, 82.0, 42.7, 21.3, 16.8, 11.4. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{20}H_{19}N_2O_2$ 319.1441; Found 319.1443.



3m

2'-Isopropyl-3-methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3m)

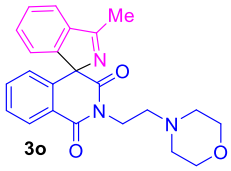
Eluent: petroleum ether/ethyl acetate (3:1). White solid (34.4 mg, 54%), mp 175.6-176.5 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.31 (dd, J_1 = 7.6 Hz, J_2 = 1.2 Hz, 1H), 7.60 (d, J = 7.6 Hz, 1H), 7.48-7.44 (m, 2H), 7.40 (td, J_1 = 7.2 Hz, J_2 = 1.6 Hz, 1H), 7.34 (td, J_1 = 7.2 Hz, J_2 = 0.8 Hz, 1H), 7.15 (d, J = 7.6 Hz, 1H), 6.66 (dd, J_1 = 7.6 Hz, J_2 = 0.8 Hz, 1H), 5.22-5.15 (m, 1H), 2.70 (s, 3H), 1.49 (d, J = 7.2 Hz, 3H), 1.39 (d, J = 6.8 Hz, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 176.1, 168.8, 164.7, 153.3, 139.6, 136.0, 133.8, 129.9, 129.4, 129.1, 128.7, 125.7, 125.0, 122.2, 121.5, 82.5, 46.3, 20.0, 19.1, 16.9. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{20}H_{19}N_2O_2$ 319.1441; Found 319.1443.



3n

2'-Benzyl-3-methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione[3] (3n)

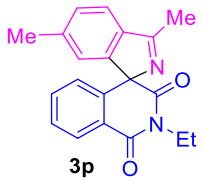
Eluent: petroleum ether/ethyl acetate (3:1). White solid (51.3 mg, 70%), mp 150.2-151.1 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.32 (dd, J_1 = 7.6 Hz, J_2 = 1.2 Hz, 1H), 7.59 (d, J = 7.6 Hz, 1H), 7.47-7.37 (m, 5H), 7.29-7.18 (m, 4H), 7.02 (d, J = 7.6 Hz, 1H), 6.63 (dd, J_1 = 7.6 Hz, J_2 = 0.8 Hz, 1H), 5.22 (d, J = 13.6 Hz, 1H), 5.16 (d, J = 14.0 Hz, 1H), 2.67 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 176.2, 168.6, 164.3, 153.3, 139.9, 136.8, 136.1, 134.1, 130.0, 129.5, 129.1, 129.0, 128.9, 128.4, 127.6, 125.3, 125.2, 122.2, 121.8, 82.2, 44.4, 16.9. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{24}H_{19}N_2O_2$ 367.1441; Found 367.1443.



3o

3-Methyl-2'-(2-morpholinoethyl)-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3o)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (40.5 mg, 52%), mp 102.9-103.2 ºC. ^{1}H NMR (600 MHz, $CDCl_3$): δ 8.32 (dd, J_1 = 7.8 Hz, J_2 = 1.2 Hz, 1H), 7.61 (d, J = 7.2 Hz, 1H), 7.49-7.45 (m, 2H), 7.42 (td, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H), 7.33 (td, J_1 = 7.8 Hz, J_2 = 1.2 Hz, 1H), 7.22 (d, J = 7.8 Hz, 1H), 6.65 (d, J = 7.8 Hz, 1H), 4.22-4.14 (m, 2H), 3.61-3.3.55 (m, 4H), 2.70 (s, 3H), 2.63-2.54 (m, 2H), 2.48 (br s, 4H). ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 176.3, 168.5, 164.4, 153.6, 139.7, 136.1, 134.0, 129.8, 129.3, 129.1, 128.8, 125.2, 125.1, 122.1, 121.9, 82.0, 67.0, 55.9, 53.7, 37.6, 16.9. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{23}H_{24}N_3O_3$ 390.1812; Found 390.1804.

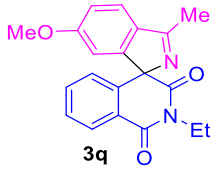


3p

2'-Ethyl-3,6-dimethyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3p)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (46.5 mg, 73%), mp 136.5-137.2 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.33 (dd, J_1 = 7.6 Hz, J_2 = 1.6 Hz, 1H), 7.49-7.44 (m, 2H), 7.41 (td, J_1 = 7.6 Hz, J_2 = 1.6 Hz,

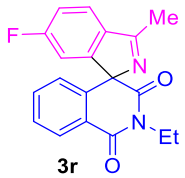
1H), 7.26 (d, J = 8.8 Hz, 1H), 6.92 (s, 1H), 6.64 (dd, J_1 = 7.6 Hz, J_2 = 0.8 Hz, 1H), 4.15-4.00 (m, 2H), 2.65 (s, 3H), 2.33 (s, 3H), 1.23 (t, J = 6.8 Hz, 3H). $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ 176.0, 168.5, 164.2, 154.0, 140.8, 137.6, 136.4, 134.0, 130.0, 129.2, 128.7, 125.4, 125.3, 122.3, 121.8, 81.6, 36.3, 21.7, 16.9, 13.2. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{20}H_{19}N_2O_2$ 319.1441; Found 319.1443.



3q

2'-Ethyl-6-methoxy-3-methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3q)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (49.5 mg, 74%), mp 126.3-127.0 ºC. ^1H NMR (400 MHz, $CDCl_3$): δ 8.32 (dd, J_1 = 7.6 Hz, J_2 = 1.6 Hz, 1H), 7.50 (d, J = 8.4 Hz, 1H), 7.47 (td, J_1 = 7.6 Hz, J_2 = 1.2 Hz, 1H), 7.42 (td, J_1 = 7.6 Hz, J_2 = 1.6 Hz, 1H), 6.97 (dd, J_1 = 8.4 Hz, J_2 = 1.6 Hz, 1H), 6.66-6.62 (m, 2H), 4.15-4.01 (m, 2H), 3.74 (s, 3H), 2.62 (s, 3H), 1.23 (t, J = 7.2 Hz, 3H). $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ 175.4, 168.4, 164.1, 161.7, 155.9, 136.4, 134.0, 133.1, 129.2, 128.8, 125.5, 125.3, 123.1, 115.0, 107.5, 81.4, 55.7, 36.3, 16.8, 13.2. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{20}H_{19}N_2O_3$ 335.1390; Found 335.1395.

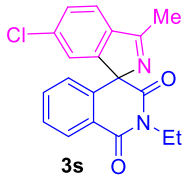


3r

2'-Ethyl-6-fluoro-3-methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3r)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (45.8 mg, 71%), mp 113.8-114.6 ºC. ^1H NMR (400 MHz, $CDCl_3$): δ 8.34 (dd, J_1 = 7.6 Hz, J_2 = 1.2 Hz, 1H), 7.57 (dd, J_1 = 8.4 Hz, J_2 = 4.4 Hz, 1H), 7.50 (td, J_1 = 7.6 Hz, J_2 = 1.2 Hz, 1H), 7.44 (td, J_1 = 7.6 Hz, J_2 = 1.2 Hz, 1H), 7.17 (td, J_1 = 8.4 Hz, J_2 = 2.0 Hz, 1H), 6.84 (dd, J_1 = 7.6 Hz, J_2 = 2.4 Hz, 1H), 6.62 (dd, J_1 = 7.6 Hz, J_2 = 0.8 Hz, 1H), 4.14-4.00 (m, 2H), 2.65 (s, 3H), 1.23 (t, J = 6.8 Hz, 3H). $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ 174.9, 167.8, 164.0 (d, $^1J_{C\text{-}F}$ = 251.4 Hz), 163.8, 156.0 (d, $^3J_{C\text{-}F}$ = 8.0 Hz), 136.3 (d, $^4J_{C\text{-}F}$ = 1.9 Hz), 135.4, 134.1, 129.5, 129.1, 125.4, 125.3, 123.5 (d, $^3J_{C\text{-}F}$ = 9.8 Hz),

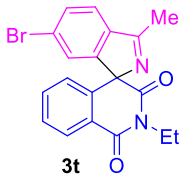
116.6 (d, $^{2}J_{C\text{-}F}$ = 24.1 Hz), 109.9 (d, $^{2}J_{C\text{-}F}$ = 25.0 Hz), 81.7 (d, $^{4}J_{C\text{-}F}$ = 2.4 Hz), 36.5, 16.8, 13.2. ^{19}F NMR (376 MHz, $CDCl_3$): δ -109.23 (td, J_1 = 8.6 Hz, J_2 = 5.3 Hz). HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{19}H_{16}FN_2O_2$ 323.1190; Found 323.1189.



3s

6-Chloro-2'-ethyl-3-methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3s)

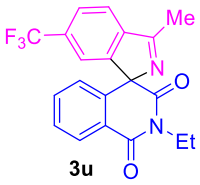
Eluent: petroleum ether/ethyl acetate (3:1). White solid (43.4 mg, 64%), mp 131.9-132.8 ºC. ^{1}H NMR (600 MHz, $CDCl_3$): δ 8.34 (dd, J_1 = 7.8 Hz, J_2 = 1.2 Hz, 1H), 7.53 (d, J = 7.8 Hz, 1H), 7.50 (td, J_1 = 7.2 Hz, J_2 = 0.6 Hz, 1H), 7.46-7.44 (m, 2H), 7.12 (d, J = 1.8 Hz, 1H), 6.61 (d, J = 7.8 Hz, 1H), 4.13-4.02 (m, 2H), 2.65 (s, 3H), 1.24 (t, J = 7.2 Hz, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 175.0, 167.7, 163.8 155.2, 138.7 136.8, 135.3, 134.1, 129.6, 129.5, 129.1, 125.4, 125.3, 122.9, 122.5, 81.8, 36.5, 16.8, 13.2. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{19}H_{16}ClN_2O_2$ 339.0895; Found 339.0898.



3t

6-Bromo-2'-ethyl-3-methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3t)

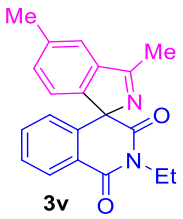
Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (46.0 mg, 60%), mp 118.7-119.1 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.34 (dd, J_1 = 7.6 Hz, J_2 = 0.8 Hz, 1H), 7.61 (dd, J_1 = 7.6 Hz, J_2 = 1.2 Hz, 1H), 7.50 (td, J_1 = 7.6 Hz, J_2 = 1.2 Hz, 1H), 7.48-7.42 (m, 2H), 7.28 (d, J = 1.2 Hz, 1H), 6.62 (dd, J_1 = 7.6 Hz, J_2 = 0.8 Hz, 1H), 4.15-4.00 (m, 2H), 2.65 (s, 3H), 1.24 (t, J = 6.8 Hz, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 175.1, 167.6, 163.8, 155.4, 139.1 135.2, 134.1, 132.4, 129.5, 129.1, 125.41, 125.35, 125.3, 125.2, 123.2, 81.8, 36.5, 16.8, 13.2. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{19}H_{16}BrN_2O_2$ 383.0390; Found 383.0384.



3u

2'-Ethyl-3-methyl-6-(trifluoromethyl)-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3u)

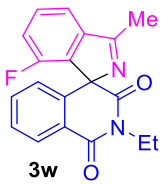
Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (45.4 mg, 61%), mp 127.9-128.6 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.37 (d, J = 7.6 Hz, 1H), 7.78 (d, J = 8.0 Hz, 1H), 7.73 (d, J = 8.0 Hz, 1H), 7.52 (t, J = 7.2 Hz, 1H), 7.45 (td, J_1 = 7.2 Hz, J_2 = 0.8 Hz, 1H), 7.38 (s, 1H), 6.57 (dd, J_1 = 7.6 Hz, J_2 = 0.4 Hz, 1H), 4.15-4.02 (m, 2H), 2.70 (s, 3H), 1.23 (t, J = 7.2 Hz, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 174.8, 167.4, 163.7, 154.0, 143.1, 134.7, 134.2, 132.1 (q, $^{2}J_{C\text{-}F}$ = 31.5 Hz), 129.6, 129.3, 126.8 (q, $^{3}J_{C\text{-}F}$ = 3.6 Hz), 125.43, 125.37, 123.6 (q, $^{1}J_{C\text{-}F}$ = 272.0 Hz), 122.5, 118.8 (q, $^{3}J_{C\text{-}F}$ = 3.3 Hz), 82.3, 36.5, 16.9, 13.1. ^{19}F NMR (376 MHz, $CDCl_3$): δ -62.08 (s). HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{20}H_{16}F_3N_2O_2$ 373.1158; Found 373.1157.



3v

2'-Ethyl-3,5-dimethyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3v)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (48.4 mg, 76%), mp 137.6-138.3 ºC. ^{1}H NMR (600 MHz, $CDCl_3$): δ 8.32 (dd, J_1 = 7.8 Hz, J_2 = 1.2 Hz, 1H), 7.46 (td, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H), 7.42-7.39 (m, 2H), 7.16 (d, J = 7.2 Hz, 1H), 7.00 (d, J = 7.8 Hz, 1H), 6.64 (d, J = 7.8 Hz, 1H), 4.12-4.01 (m, 2H), 2.67 (s, 3H), 2.44 (s, 3H), 1.21 (t, J = 7.2 Hz, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 176.1, 168.5, 164.2 150.9, 140.2, 139.3, 136.4, 133.9, 130.9, 129.2, 128.7, 125.3, 122.6, 121.3, 81.6, 36.3, 21.4, 16.8, 13.1. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{20}H_{19}N_2O_2$ 319.1441; Found 319.1446.



3w

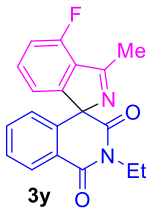
2'-Ethyl-7-fluoro-3-methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3w)

Eluent: petroleum ether/ethyl acetate (3:1). Purple solid (43.8 mg, 68%), mp 110.2-110.9 ºC. ^{1}H NMR (600 MHz, $CDCl_3$): δ 8.33 (dd, J_1 = 7.8 Hz, J_2 = 0.6 Hz, 1H), 7.51-7.48 (m, 2H), 7.44-7.41 (m, 2H), 7.04 (t, J = 7.8 Hz, 1H), 6.60 (d, J = 7.8 Hz, 1H), 4.10 (q, J = 7.2 Hz, 2H), 2.64 (s, 3H), 1.24 (t, J = 7.2 Hz, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 174.5, 167.2, 163.8, 157.0 (d, $^{1}J_{C\text{-}F}$ = 249.8 Hz), 143.9 (d, $^{3}J_{C\text{-}F}$ = 4.0 Hz), 138.8 (d, $^{2}J_{C\text{-}F}$ = 16.4 Hz), 134.2, 133.8, 132.0 (d, $^{3}J_{C\text{-}F}$ = 6.3 Hz), 129.4, 129.1, 125.7, 125.1, 117.9 (d, $^{4}J_{C\text{-}F}$ = 3.8 Hz), 117.3 (d, $^{2}J_{C\text{-}F}$ = 19.6 Hz), 80.5 (d, $^{3}J_{C\text{-}F}$ = 3.6 Hz), 36.4, 16.9, 13.0. ^{19}F NMR (565 MHz, $CDCl_3$): δ -118.39 (dd, J_1 = 8.5 Hz, J_2 = 4.5 Hz). HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{19}H_{16}FN_2O_2$ 323.1190; Found 323.1188.

3x

2'-Ethyl-3,4-dimethyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3x)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (31.8 mg, 50%), mp 137.7-138.2 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.32 (dd, J_1 = 7.6 Hz, J_2 = 1.2 Hz, 1H), 7.46 (td, J_1 = 7.6 Hz, J_2 = 1.6 Hz, 1H), 7.41 (td, J_1 = 7.6 Hz, J_2 = 1.6 Hz, 1H), 7.20-7.18 (m, 2H), 6.94-6.91 (m, 1H), 6.67-6.65 (m, 1H), 4.12-3.98 (m, 2H), 2.80 (s, 3H), 2.69 (s, 3H), 1.21 (t, J = 7.2 Hz, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 176.7, 168.4, 164.2, 154.5, 137.7, 136.5, 134.2, 133.9, 131.6, 130.1, 129.2, 128.7, 125.4, 125.2, 119.5, 81.1, 36.3, 21.4, 19.7, 13.1. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{20}H_{19}N_2O_2$ 319.1441; Found 319.1443.



3y

2'-Ethyl-4-fluoro-3-methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3y)

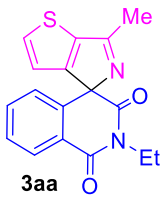
Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (36.1 mg, 56%), mp 133.0-133.9 ºC. ^{1}H NMR (600

MHz, $CDCl_3$): δ 8.33 (dd, J_1 = 7.2 Hz, J_2 = 0.6 Hz, 1H), 7.50-7.48 (m, 1H), 7.45 (td, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H), 7.34-7.31 (m, 1H), 7.10 (t, J = 9.0 Hz, 1H), 6.90 (d, J = 7.8 Hz, 1H), 6.67 (d, J = 7.2 Hz, 1H), 4.12-4.01 (m, 2H), 2.78 (s, 3H), 1.22 (t, J = 6.6 Hz, 3H). $^{13}C\{^1H\}$ NMR (150 MHz, $CDCl_3$): δ 173.3 (d, $^3J_{C-F}$ = 3.9 Hz), 167.7, 163.9, 157.0 (d, $^1J_{C-F}$ = 254.9 Hz), 156.7 (d, $^3J_{C-F}$ = 3.9 Hz), 135.5, 134.1, 132.6 (d, $^3J_{C-F}$ = 6.2 Hz), 129.4, 129.0, 127.1 (d, $^2J_{C-F}$ = 16.8 Hz), 125.3, 125.2, 117.8 (d, $^4J_{C-F}$ = 4.8 Hz), 116.3 (d, $^2J_{C-F}$ = 20.4 Hz), 82.5, 36.4, 19.7 (d, $^4J_{C-F}$ = 1.7 Hz), 13.1. ^{19}F NMR (565 MHz, $CDCl_3$): δ -117.23 – -117.24 (m). HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{19}H_{16}FN_2O_2$ 323.1190; Found 323.1196.

3z

4-Chloro-2'-ethyl-3-methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3z)

Eluent: petroleum ether/ethyl acetate (3:1). White solid (42.7 mg, 63%), mp 135.3-136.1 ºC. ^1H NMR (600 MHz, $CDCl_3$): δ 8.33 (dd, J = 7.8 Hz, 1H), 7.49 (t, J = 7.2 Hz 1H), 7.44 (t, J = 7.2 Hz, 1H), 7.39 (d, J = 7.8 Hz, 1H), 7.26 (t, J = 7.8 Hz, 1H), 7.00 (d, J = 7.2 Hz, 1H), 6.66 (d, J_1 = 7.8 Hz, 1H), 4.10-4.00 (m, 2H), 2.87 (s, 3H), 1.22 (t, J = 7.2 Hz, 3H). $^{13}C\{^1H\}$ NMR (150 MHz, $CDCl_3$): δ 175.1, 167.7, 163.9, 156.2, 136.5, 135.5, 134.1, 131.4, 130.6, 129.4, 129.2, 129.1, 125.4, 125.3, 120.3, 81.6, 36.4, 21.0, 13.1. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{19}H_{16}ClN_2O_2$ 339.0895; Found 339.0894.

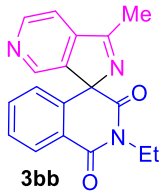


3aa

2-Ethyl-6'-methyl-1*H*-spiro[isoquinoline-4,4'-thieno[2,3-c]pyrrole]-1,3(2*H*)-dione (3aa)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (27.9 mg, 45%), mp 108.2-109.0 ºC. ^1H NMR (400 MHz, $CDCl_3$): δ 8.31-8.29 (m, 1H), 7.55 (d, J = 4.8 Hz, 1H), 7.50-7.42 (m, 2H), 6.81 (d, J = 4.8 Hz, 1H),

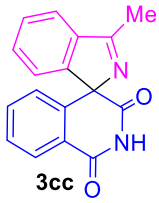
6.72-6.70 (m, 1H), 4.07 (q, J = 6.8 Hz, 2H), 2.57 (s, 3H), 1.23 (t, J = 7.2 Hz, 3H). $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ 169.5, 167.3, 163.9, 163.7, 144.9, 135.0, 134.9, 134.0, 129.4, 129.0, 125.5, 125.2, 120.2, 78.7, 36.5, 18.2, 13.2. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{17}H_{15}N_2O_2S$ 311.0849; Found 311.0851.



3bb

2-Ethyl-1'-methyl-1*H*-spiro[isoquinoline-4,3'-pyrrolo[3,4-c]pyridine]-1,3(2*H*)-dione (3bb)

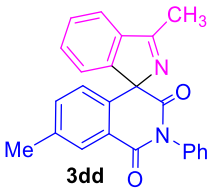
Eluent: petroleum ether/ethyl acetate (3:1). Yellow syrup (26.9 mg, 44%).^1H NMR (400 MHz, $CDCl_3$): δ 8.78 (d, J = 5.2 Hz, 1H), 8.49 (s, 1H), 8.37 (dd, J_1 = 7.6 Hz, J_2 = 1.2 Hz, 1H), 7.54-7.50 (m, 2H), 7.44 (td, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H), 6.56 (dd, J_1 = 7.2 Hz, J_2 = 0.4 Hz, 1H), 4.15-4.01 (m, 2H), 2.69 (s, 3H), 1.23 (t, J = 6.8 Hz, 3H). $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ 174.4, 167.2, 163.6, 150.4, 147.8, 147.2, 143.5, 134.3, 134.1, 129.7, 129.4, 125.4, 125.2, 116.1, 82.2, 36.6, 16.8, 13.2. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{18}H_{16}N_3O_2$ 306.1237; Found 306.1238.



3cc

3-Methyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3cc)

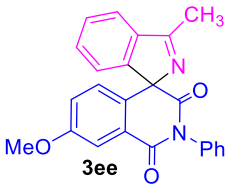
Eluent: petroleum ether/ethyl acetate (3:1). White solid (41.4 mg, 75%), mp 216.9-217.1 ºC. ^1H NMR (400 MHz, DMSO-d_6): δ 11.92 (s, 1H), 8.20 (d, J = 7.6 Hz 1H), 7.78 (t, J = 7.6 Hz, 1H), 7.57-7.49 (m, 3H), 7.41 (t, J = 7.2 Hz, 1H), 7.33 (d, J = 7.2 Hz, 1H), 6.56 (d, J_1 = 7.6 Hz, 1H), 2.59 (s, 3H). $^{13}C\{^1H\}$ NMR (100 MHz, DMSO-d_6): δ 175.6, 169.2, 164.9, 153.7, 140.6, 137.8, 134.7, 130.4, 129.4, 129.3, 128.5, 126.2, 125.4, 122.8, 122.4, 82.1, 16.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{17}H_{13}N_2O_2$ 277.0792; Found 277.0796.



3dd

3,7'-Dimethyl-2'-phenyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3dd)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow syrup (55.7 mg, 76%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.17 (s, 1H), 7.60 (d, *J* = 7.2 Hz 1H), 7.50-7.44 (m, 3H), 7.42-7.37 (m, 2H), 7.33-7.28 (m, 2H), 7.21-7.19 (m, 2H), 6.64 (d, *J* = 8.0 Hz, 1H), 2.67 (s, 3H), 2.43 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 176.1, 168.6, 164.7, 153.3, 139.9, 139.1, 135.4, 135.2, 133.4, 130.1, 129.8, 129.2, 129.1, 128.7, 128.4, 125.4, 125.2, 122.2, 121.7, 82.5, 21.2, 16.9. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{24}H_{19}N_2O_2$ 367.1441; Found 367.1442.



3ee

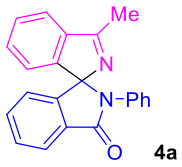
7'-Methoxy-3-methyl-2'-phenyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (3ee)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (55.1 mg, 72%), mp 183.3-183.6 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.82 (d, *J* = 2.8 Hz, 1H), 7.60 (d, *J* = 7.6 Hz 1H), 7.50-7.38 (m, 5H), 7.33 (d, *J* = 7.6 Hz, 1H), 7.22-7.20 (m, 2H), 7.03 (dd, J_1 = 8.8 Hz, J_2 = 2.8 Hz, 1H), 6.64 (d, *J* = 8.4 Hz, 1H), 3.87 (s, 3H), 2.66 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 176.0, 168.7, 164.5, 160.1, 153.3, 140.0, 135.2, 130.1, 129.22, 129.16, 128.7, 128.4, 127.0, 126.6, 122.5, 122.2, 121.7, 112.0, 82.4, 55.7, 16.9. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{24}H_{19}N_2O_3$ 383.1390; Found 383.1392.

2. Typical procedure for the synthesis of 4a and spectroscopic data of 4a-4ee

To a reaction tube equipped with a stir bar were added 3-phenylisoxazol-5(4*H*)-one (**1a**, 32.3 mg, 0.2 mmol), KOAc (39.3 mg, 0.4 mmol), $[RhCp^*Cl_2]_2$ (6.2 mg, 0.01 mmol), 4-diazo-2-phenylisoquinoline-1,3(2*H*,4*H*)-dione (**2a**, 63.2 mg, 0.24 mmol) and DCE (2 mL). The tube was then sealed, and the mixture was stirred at 80 °C under air for 12 h. Upon completion, it was cooled to room temperature, filtered through a pad of celite

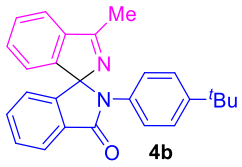
and concentrated under reduced pressure. The residue was purified by silica gel column chromatography using petroleum ether/ethyl acetate (3:1) as eluent to afford **4a**. Other products **4b-4ee** were obtained in a similar manner.



4a

3-Methyl-2'-phenylspiro[isoindole-1,1'-isoindolin]-3'-one (4a)

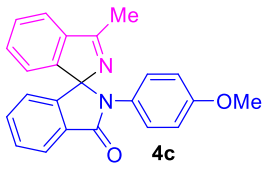
Eluent: petroleum ether/ethyl acetate (3:1). White solid (49.9 mg, 77%), mp 213.7-214.0 ºC. ^{1}H NMR (600 MHz, $CDCl_3$): δ 8.01 (d, J = 7.2 Hz, 1H), 7.51 (td, J_1 = 7.2 Hz, J_2 = 0.6 Hz, 1H), 7.47 (d, J = 7.2 Hz, 1H), 7.44 (td, J_1 = 7.2 Hz, J_2 = 0.6 Hz, 1H), 7.40 (td, J_1 = 7.2 Hz, J_2 = 0.6 Hz, 1H), 7.34 (td, J_1 = 7.2 Hz, J_2 = 0.6 Hz, 1H), 7.22-7.18 (m, 3H), 7.16-7.12 (m, 3H), 6.75 (d, J = 7.8 Hz, 1H), 2.52 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 174.2, 168.5, 149.0, 142.9, 139.9, 135.7, 132.7, 132.1, 130.1, 129.5, 129.4, 128.7, 127.3, 127.2, 124.6, 122.7, 121.5, 121.4, 95.1, 16.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{22}H_{17}N_2O$ 325.1335; Found 325.1329.



4b

2'-(4-(*tert*-Butyl)phenyl)-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4b)

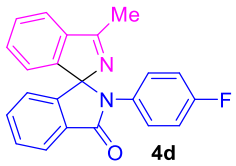
Eluent: petroleum ether/ethyl acetate (3:1). White solid (43.4 mg, 57%), mp 161.6-162.3 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.00 (d, J = 7.2 Hz, 1H), 7.51-7.48 (m, 2H), 7.42 (t, J = 7.2 Hz, 2H), 7.35 (td, J_1 = 7.2 Hz, J_2 = 0.8 Hz, 1H), 7.23-7.17 (m, 3H), 7.07-7.04 (m, 2H), 6.72 (d, J = 7.6 Hz, 1H), 2.53 (s, 3H), 1.22 (s, 9H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 174.0, 168.5, 149.8, 149.2, 142.9, 139.9, 132.9, 132.5, 132.2, 130.1, 129.4, 129.3, 126.4, 125.7, 124.5, 122.7, 121.5, 121.3, 95.1, 34.5, 31.3, 16.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{26}H_{25}N_2O$ 381.1961; Found 381.1966.



4c

2'-(4-Methoxyphenyl)-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4c)

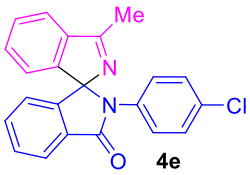
Eluent: petroleum ether/ethyl acetate (3:1). White solid (41.1 mg, 58%), mp 140.8-141.4 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.00 (d, J = 7.6 Hz, 1H), 7.50 (t, J = 7.2 Hz, 1H), 7.46-7.39 (m, 3H), 7.35 (td, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H), 7.22 (d, J = 7.2 Hz, 1H), 7.06 (d, J = 9.2 Hz, 2H), 6.76 (d, J = 7.6 Hz, 1H), 6.72 (d, J = 8.8 Hz, 2H), 3.71 (s, 3H), 2.50 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 174.2, 168.7, 158.6, 148.9, 142.8, 140.0, 132.5, 132.3, 130.1, 129.5, 129.4, 129.1, 128.0, 124.5, 122.7, 121.5, 121.4, 114.0, 95.1, 55.3, 16.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{23}H_{19}N_2O_2$ 355.1441; Found 355.1447.



4d

2'-(4-Fluorophenyl)-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4d)

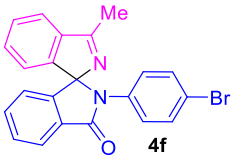
Eluent: petroleum ether/ethyl acetate (3:1). White solid (41.1 mg, 60%), mp 142.7-143.6 ºC. ^{1}H NMR (600 MHz, $CDCl_3$): δ 8.01 (d, J = 7.8 Hz, 1H), 7.52 (td, J_1 = 7.2 Hz, J_2 = 0.6 Hz, 1H), 7.48 (d, J = 7.8 Hz, 1H), 7.46-7.41 (m, 2H), 7.36 (td, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H), 7.21 (d, J = 7.2 Hz, 1H), 7.14-7.11 (m, 2H), 6.90-6.86 (m, 2H), 6.77 (d, J = 7.8 Hz, 1H), 2.52 (s, 3H). ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 174.4, 168.5, 161.5 (d, $^{1}J_{C\text{-}F}$ = 245.0 Hz), 148.7, 142.7, 139.9, 132.8, 131.9, 131.4 (d, $^{4}J_{C\text{-}F}$ = 3.8 Hz), 130.2, 129.7, 129.5 (d, $^{3}J_{C\text{-}F}$ = 8.6 Hz), 129.4, 124.6, 122.7, 121.6, 121.5, 115.7 (d, $^{2}J_{C\text{-}F}$ = 21.8 Hz), 95.0, 16.8. ^{19}F NMR (565 MHz, $CDCl_3$): δ -114.19 – -114.24 (m). HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{22}H_{16}FN_2O$ 343.1241; Found 343.1239.



4e

2'-(4-Chlorophenyl)-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4e)

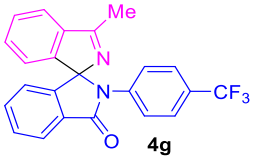
Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (58.1 mg, 81%), mp 124.5-124.9 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.01-7.99 (m, 1H), 7.53-7.49 (m, 2H), 7.47-7.41 (m, 2H), 7.35 (td, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H), 7.19 (d, J = 7.6 Hz, 1H), 7.18-7.14 (m, 2H), 7.12-7.08 (m, 2H), 6.75 (d, J = 7.6 Hz, 1H), 2.53 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 174.5, 168.4, 148.7, 142.8, 139.9, 134.3, 132.9, 132.8, 131.8, 130.3, 129.7, 129.5, 129.0, 128.4, 124.6, 122.6, 121.7, 121.4, 95.0, 16.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{22}H_{16}ClN_2O$ 359.0946; Found 359.0945.



4f

2'-(4-Bromophenyl)-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4f)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (58.1 mg, 72%), mp 142.8-143.5 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.99 (d, J = 7.6 Hz, 1H), 7.52-7.49 (m, 2H), 7.46-7.41 (m, 2H), 7.32 (t, J = 7.2 Hz, 1H), 7.33-7.29 (m, 2H), 7.19 (d, J = 7.6 Hz, 1H), 7.06-7.02 (m, 2H), 6.75 (d, J = 7.6 Hz, 1H), 2.54 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 174.5, 168.4, 148.7, 142.8, 139.9, 134.9, 132.9, 132.0, 131.8, 130.3, 129.8, 129.5, 128.6, 124.6, 122.6, 121.7, 121.4, 120.9, 95.0, 16.9. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{22}H_{16}BrN_2O$ 403.0441; Found 403.0440.

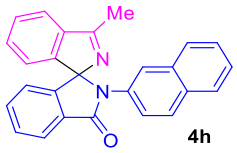


4g

3-Methyl-2'-(4-(trifluoromethyl)phenyl)spiro[isoindole-1,1'-isoindolin]-3'-one (4g)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (48.7 mg, 62%), mp 145.6-146.1 ºC. ^{1}H NMR (400

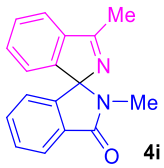
MHz, $CDCl_3$): δ 8.02 (d, *J* = 7.2 Hz, 1H), 7.55-7.51 (m, 2H), 7.48-7.44 (m, 4H), 7.37 (t, *J* = 7.2 Hz, 1H), 7.30 (d, *J* = 8.4 Hz, 2H), 7.20 (d, *J* = 7.6 Hz, 1H), 6.74 (d, *J* = 7.6 Hz, 1H), 2.56 (s, 3H). $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ 174.6, 168.4, 148.9, 142.8, 139.8, 139.5, 133.1, 131.5, 130.5, 129.8, 129.6, 128.6 (q, $^2J_{C\text{-}F}$ = 32.5 Hz), 126.6 (q, $^1J_{C\text{-}F}$ = 269.9 Hz), 126.1, 125.9 (q, $^3J_{C\text{-}F}$ = 3.9 Hz), 124.7, 122.5, 121.8, 121.3, 95.1, 16.9. ^{19}F NMR (376 MHz, $CDCl_3$): δ -62.57 (s). HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{23}H_{16}F_3N_2O$ 393.1209; Found 393.1208.



4h

3-Methyl-2'-(naphthalen-2-yl)spiro[isoindole-1,1'-isoindolin]-3'-one (4h)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (48.7 mg, 65%), mp 136.4-137.2 ºC. ^1H NMR (400 MHz, $CDCl_3$): δ 8.04 (d, *J* = 7.2 Hz, 1H), 7.73-7.61 (m, 4H), 7.53 (td, J_1 = 7.6 Hz, J_2 = 0.4 Hz, 1H), 7.48-7.47 (m, 8H), 6.78 (d, *J* = 7.6 Hz, 1H), 2.51 (s, 3H). $^{13}C\{^1H\}$ NMR (150 MHz, $CDCl_3$): δ 174.3, 168.7, 149.0, 143.0, 140.0, 133.35, 133.32, 132.7, 132.3, 132.1, 130.2, 129.6, 129.5, 128.5, 128.1, 127.5, 126.1, 126.0, 125.8, 125.6, 124.6, 122.7, 121.6, 121.4, 95.4, 16.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{26}H_{19}N_2O$ 375.1492; Found 375.1488.

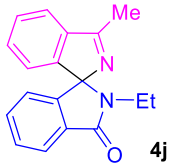


4i

2',3-Dimethylspiro[isoindole-1,1'-isoindolin]-3'-one (4i)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow syrup (21.5 mg, 41%). ^1H NMR (600 MHz, $CDCl_3$): δ 7.91 (d, *J* = 7.8 Hz, 1H), 7.61 (d, *J* = 7.8 Hz, 1H), 7.51 (t, *J* = 7.2 Hz, 1H), 7.47 (t, *J* = 7.2 Hz, 1H), 7.38 (t, *J* = 7.2 Hz, 2H), 7.06 (d, *J* = 7.2 Hz, 1H), 6.75 (d, *J* = 7.2 Hz, 1H), 2.63 (s, 3H), 2.62 (s, 3H). $^{13}C\{^1H\}$ NMR (150 MHz, $CDCl_3$): δ 174.6, 168.6, 148.2, 142.5, 139.9, 132.6, 132.0, 130.3, 129.8, 129.3, 123.9, 122.6, 121.6, 121.3, 94.0,

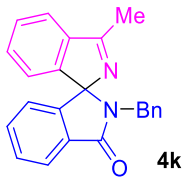
24.6, 16.9. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{17}H_{15}N_2O$ 263.1179; Found 263.1178.



4j

2'-Ethyl-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4j)

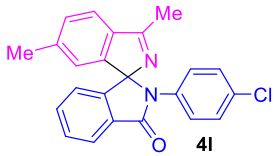
Eluent: petroleum ether/ethyl acetate (3:1). Yellow syrup (21.0 mg, 38%). ^{1}H NMR (600 MHz, $CDCl_3$): δ 7.91 (d, *J* = 7.2 Hz, 1H), 7.61 (d, *J* = 7.8 Hz, 1H), 7.50 (t, *J* = 7.2 Hz, 1H), 7.45 (t, *J* = 7.2 Hz, 1H), 7.39-7.36 (m, 2H), 7.11 (d, *J* = 7.2 Hz, 1H), 6.70 (d, *J* = 7.8 Hz, 1H), 3.20-3.23 (m, 2H), 2.61 (s, 3H), 1.01 (t, *J* = 7.2 Hz, 3H). ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 174.0, 168.8, 148.9, 142.7, 139.9, 132.7, 132.0, 130.1, 129.7, 129.2, 123.8, 122.9, 121.5, 121.2, 94.3, 35.0, 16.9, 14.6. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{18}H_{17}N_2O$ 277.1335; Found 277.1334.



4k

2'-Benzyl-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4k)

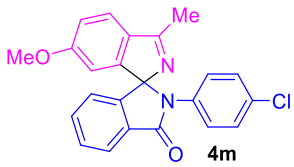
Eluent: petroleum ether/ethyl acetate (3:1). Yellow syrup (29.8 mg, 44%). ^{1}H NMR (600 MHz, $CDCl_3$): δ 7.96 (d, *J* = 7.2 Hz, 1H), 7.51 (d, *J* = 7.2 Hz, 1H), 7.47 (t, *J* = 7.2 Hz, 1H), 7.39-7.36 (m, 2H), 7.10 (t, *J* = 7.2 Hz, 1H), 7.07-7.04 (m, 3H), 6.95-6.93 (m, 2H), 6.72 (d, *J* = 7.8 Hz, 1H), 6.69 (d, *J* = 7.8 Hz, 1H), 4.44 (d, *J* = 15.0 Hz, 1H), 4.18 (d, *J* = 15.0 Hz, 1H), 2.48 (s, 3H). ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 174.2, 168.9, 148.3, 142.9, 139.8, 137.7, 132.3, 132.2, 129.8, 129.33, 129.27, 128.5, 127.9, 126.9, 124.1, 123.2, 121.3, 121.2, 93.9, 43.6, 16.7. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{23}H_{19}N_2O$ 339.1492; Found 339.1496.



4l

2'-(4-Chlorophenyl)-3,6-dimethylspiro[isoindole-1,1'-isoindolin]-3'-one (4l)

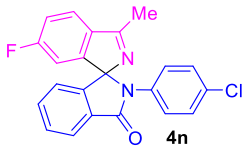
Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (46.2 mg, 62%), mp 189.7-190.3 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.00 (d, J = 7.2 Hz, 1H), 7.51 (td, J_1 = 7.2 Hz, J_2 = 0.8 Hz, 1H), 7.45 (td, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H), 7.37 (d, J = 8.0 Hz, 1H), 7.22 (d, J = 7.6 Hz, 1H), 7.19-7.15 (m, 2H), 7.12-7.09 (m, 2H), 6.98 (s, 1H), 6.75 (d, J = 7.6 Hz, 1H), 2.50 (s, 3H), 2.33 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 174.4, 168.4, 149.3, 143.1, 141.1, 137.7, 134.5, 132.8, 132.6, 131.7, 130.5, 129.4, 128.9, 128.1, 124.5, 123.2, 121.42, 121.38, 94.7, 21.7, 16.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{23}H_{18}ClN_2O$ 373.1102; Found 373.1099.



4m

2'-(4-Chlorophenyl)-6-methoxy-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4m)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow syrup (57.5 mg, 74%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.80-7.98 (m, 1H), 7.51 (td, J_1 = 7.2 Hz, J_2 = 0.8 Hz, 1H), 7.45 (td, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H), 7.39 (d, J = 8.4 Hz, 1H), 7.19-7.16 (m, 2H), 7.14-7.11 (m, 2H), 6.92 (dd, J_1 = 8.4 Hz, J_2 = 2.0 Hz, 1H), 6.78 (d, J = 7.6 Hz, 1H), 6.67 (d, J = 2.0 Hz, 1H), 3.74 (s, 3H), 2.48 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 174.1, 168.4, 162.0, 151.5, 143.2, 134.5, 133.0, 132.9, 132.6, 131.6, 129.5, 129.0, 128.0, 124.5, 122.7, 121.5, 115.6, 108.1, 94.4, 55.8, 16.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{23}H_{18}ClN_2O_2$ 389.1051; Found 389.1052.

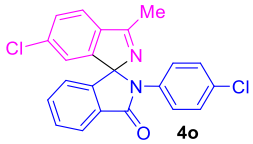


4n

2'-(4-Chlorophenyl)-6-fluoro-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4n)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (53.5 mg, 71%), mp 106.7-107.1 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.00 (d, J = 7.2 Hz, 1H), 7.53 (td, J_1 = 7.2 Hz, J_2 = 0.8 Hz, 1H), 7.49-7.43 (m, 2H), 7.21-7.18 (m, 2H), 7.14-7.10 (m, 3H), 6.89 (dd, J_1 = 7.2 Hz, J_2 = 2.0 Hz, 1H), 6.77 (d, J = 7.6 Hz, 1H), 2.52 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 173.6, 168.3, 164.4 (d, $^{1}J_{C\text{-}F}$ = 252.3 Hz), 151.8 (d, $^{3}J_{C\text{-}F}$ = 9.1 Hz), 142.2,

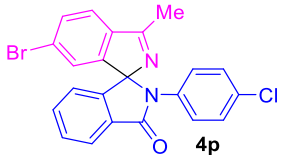
135.9 (d, $^{4}J_{C\text{-}F}$ = 1.8 Hz), 134.1, 133.1, 133.0, 131.6, 129.8, 129.1, 128.4, 124.8, 123.1 (d, $^{3}J_{C\text{-}F}$ = 8.9 Hz), 121.4, 117.2 (d, $^{2}J_{C\text{-}F}$ = 23.0 Hz), 110.6 (d, $^{2}J_{C\text{-}F}$ = 24.5 Hz), 94.4 (d, $^{4}J_{C\text{-}F}$ = 2.3 Hz), 16.8. ^{19}F NMR (376 MHz, $CDCl_3$): δ -108.79 (td, J_1 = 8.3 Hz, J_2 = 5.3 Hz). HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{22}H_{15}ClFN_2O$ 377.0851; Found 377.0847.



4o

6-Chloro-2'-(4-chlorophenyl)-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4o)

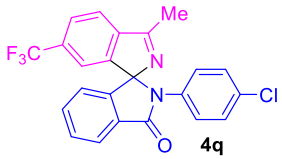
Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (62.9 mg, 80%), mp 138.4-139.2 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.00 (d, J = 7.6 Hz, 1H), 7.54 (td, J_1 = 7.6 Hz, J_2 = 0.8 Hz, 1H), 7.48 (td, J_1 = 7.6 Hz, J_2 = 0.8 Hz, 1H), 7.41-7.40 (m, 2H), 7.22-7.17 (m, 3H), 7.12-7.08 (m, 2H), 6.76 (d, J = 7.2 Hz, 1H), 2.51 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 173.5, 168.2, 150.8, 142.0, 138.3, 137.2, 134.0, 133.1, 133.0, 131.6, 130.2, 129.8, 129.2, 128.5, 124.8, 123.2, 122.5, 121.4, 94.6, 16.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{22}H_{15}Cl_2N_2O$ 393.0556; Found 393.0554.



4p

6-Bromo-2'-(4-chlorophenyl)-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4p)

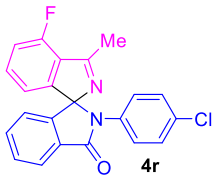
Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (74.4 mg, 85%), mp 165.6-166.2 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.01 (d, J = 7.2 Hz, 1H), 7.58-7.52 (m, 2H), 7.48 (td, J_1 = 7.6 Hz, J_2 = 1.2 Hz, 1H), 7.35-7.33 (m, 2H), 7.22-7.19 (m, 2H), 7.12-7.08 (m, 2H), 6.76 (d, J = 7.2 Hz, 1H), 2.51 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 173.6, 168.2, 151.0, 142.0, 138.7, 134.0, 133.11, 133.08, 133.0, 131.6, 129.8, 129.2, 128.5, 126.1, 125.6, 124.8, 122.8, 121.4, 94.7, 16.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{22}H_{15}BrClN_2O$ 437.0051; Found 437.0047.



4q

2'-(4-Chlorophenyl)-3-methyl-6-(trifluoromethyl)spiro[isoindole-1,1'-isoindolin]-3'-one (4q)

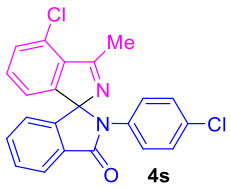
Eluent: petroleum ether/ethyl acetate (3:1). Yellow syrup (64.0 mg, 75%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.02 (d, *J* = 7.6 Hz, 1H), 7.72 (d, *J* = 8.0 Hz, 1H), 7.60 (d, *J* = 8.0 Hz, 1H), 7.58-7.54 (m, 1H), 7.49 (td, J_1 = 7.2 Hz, J_2 = 0.8 Hz, 1H), 7.44 (s, 1H), 7.20-7.17 (m, 2H), 7.09-7.05 (m, 2H), 6.76 (d, *J* = 7.6 Hz, 1H), 2.57 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 173.5, 168.3, 149.7, 142.5, 141.5, 133.8, 133.4, 133.2, 132.4 (q, $^{2}J_{C-F}$ = 32.2 Hz), 131.7, 130.0, 129.2, 128.7, 127.5 (q, $^{3}J_{C-F}$ = 3.7 Hz), 124.9, 122.1, 121.4, 120.9 (q, $^{1}J_{C-F}$ = 268.8 Hz), 119.5 (q, $^{3}J_{C-F}$ = 3.3 Hz), 95.0, 16.9. ^{19}F NMR (376 MHz, $CDCl_3$): δ -62.18 (s). HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{23}H_{15}ClF_3N_2O$ 427.0820; Found 427.0815.



4r

2'-(4-Chlorophenyl)-4-fluoro-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4r)

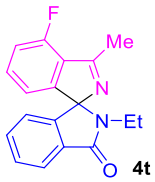
Eluent: petroleum ether/ethyl acetate (3:1). Yellow syrup (58.0 mg, 77%). ^{1}H NMR (600 MHz, $CDCl_3$): δ 8.00 (d, *J* = 7.8 Hz, 1H), 7.53 (t, *J* = 7.2 Hz, 1H), 7.47 (t, *J* = 7.2 Hz, 1H), 7.34-7.31 (m, 1H), 7.19 (d, *J* = 7.8 Hz, 2H), 7.11 (d, *J* = 7.8 Hz, 2H), 7.06 (t, *J* = 8.4 Hz, 1H), 6.97 (d, *J* = 7.2 Hz, 1H), 6.81 (d, *J* = 7.8 Hz, 1H), 2.64 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 171.5 (d, $^{3}J_{C-F}$ = 4.3 Hz), 168.2, 156.9 (d, $^{1}J_{C-F}$ = 254.8 Hz), 152.4 (d, $^{3}J_{C-F}$ = 2.6 Hz), 142.2, 134.1, 133.1, 133.0, 132.9 (d, $^{3}J_{C-F}$ = 6.0 Hz), 131.6, 129.7, 129.1, 128.5, 126.7 (d, $^{2}J_{C-F}$ = 15.9 Hz), 124.7, 121.4, 118.7 (d, $^{4}J_{C-F}$ = 3.6 Hz), 117.0 (d, $^{2}J_{C-F}$ = 20.2 Hz), 95.4, 19.7 (d, $^{4}J_{C-F}$ = 2.0 Hz). ^{19}F NMR (565 MHz, $CDCl_3$): δ -117.51 – -117.52 (m). HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{22}H_{15}ClFN_2O$ 377.0851; Found 377.0849.



4s

4-Chloro-2'-(4-chlorophenyl)-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4s)

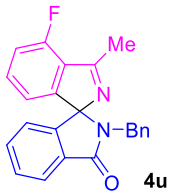
Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (39.3 mg, 50%), mp 160.2-161.1 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.00 (d, J = 7.6 Hz, 1H), 7.53 (td, J_1 = 7.2 Hz, J_2 = 0.8 Hz, 1H), 7.48 (td, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H), 7.35 (d, J = 7.2 Hz, 1H), 7.29-7.25 (m, 1H), 7.22-7.19 (m, 2H), 7.12-7.08 (m, 2H), 7.07 (d, J = 7.6 Hz, 1H), 6.80 (d, J = 7.6 Hz, 1H), 2.73 (s, 3H). ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 173.3, 168.3, 151.9, 142.2, 136.3, 134.0, 133.2, 133.0, 131.6, 131.3, 129.7, 129.2, 128.6, 124.7, 121.4, 121.2, 94.5, 20.9. HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{22}H_{15}Cl_2N_2O$ 393.0556; Found 393.0559.



4t

2'-Ethyl-4-fluoro-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4t)

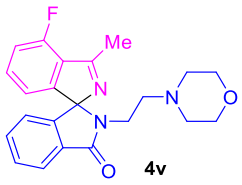
Eluent: petroleum ether/ethyl acetate (3:1). Yellow syrup (31.2 mg, 53%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.89 (d, J = 7.6 Hz, 1H), 7.45 (td, J_1 = 7.2 Hz, J_2 = 0.8 Hz, 1H), 7.38 (td, J_1 = 7.6 Hz, J_2 = 0.8 Hz, 1H), 7.35-7.31 (m, 1H), 7.12 (t, J = 8.4 Hz, 1H), 6.87 (d, J = 7.6 Hz, 1H), 6.73 (d, J = 7.2 Hz, 1H), 3.24-3.01 (m, 2H), 2.70 (d, J = 1.2 Hz, 3H), 1.01 (t, J = 7.2 Hz, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 171.1 (d, $^{3}J_{C\text{-}F}$ = 4.5 Hz), 168.6, 157.0 (d, $^{1}J_{C\text{-}F}$ = 255.4 Hz), 152.7 (d, $^{3}J_{C\text{-}F}$ = 2.4 Hz), 142.2, 132.7 (d, $^{3}J_{C\text{-}F}$ = 6.8 Hz), 132.5, 132.1, 129.4, 126.7 (d, $^{2}J_{C\text{-}F}$ = 15.9 Hz), 123.9, 121.1, 119.0 (d, $^{4}J_{C\text{-}F}$ = 3.7 Hz), 116.9 (d, $^{2}J_{C\text{-}F}$ = 21.0 Hz), 94.7, 35.0, 19.8 (d, $^{4}J_{C\text{-}F}$ = 1.9 Hz), 14.6. ^{19}F NMR (376 MHz, $CDCl_3$): δ -118.06 (dd, J_1 = 8.6 Hz, J_2 = 4.1 Hz). HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{18}H_{16}FN_2O$ 295.1241; Found 295.1246.



4u

2'-Benzyl-4-fluoro-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4u)

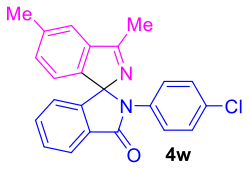
Eluent: petroleum ether/ethyl acetate (3:1). Yellow syrup (29.2 mg, 41%). ^{1}H NMR (600 MHz, $CDCl_3$): δ 7.96 (d, J = 7.8 Hz, 1H), 7.49 (t, J = 7.2 Hz, 1H), 7.40 (td, J_1 = 7.2 Hz, J_2 = 0.6 Hz, 1H), 7.08-7.06 (m, 3H), 7.05-7.03 (m, 1H), 7.01-6.97 (m, 3H), 6.73 (d, J = 7.8 Hz, 1H), 6.47 (d, J = 7.2 Hz, 1H), 4.53 (d, J = 15.0 Hz, 1H), 4.18 (d, J = 15.0 Hz, 1H), 2.59 (s, 3H). ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 171.3 (d, $^{3}J_{C\text{-}F}$ = 3.6 Hz), 168.7, 156.8 (d, $^{1}J_{C\text{-}F}$ = 254.7 Hz), 152.1 (d, $^{3}J_{C\text{-}F}$ = 2.4 Hz), 142.4, 137.5, 132.4, 132.2 (d, $^{3}J_{C\text{-}F}$ = 7.4 Hz), 132.1, 129.5, 128.5, 128.0, 127.1, 126.5 (d, $^{2}J_{C\text{-}F}$ = 15.6 Hz), 124.2, 121.3, 119.3 (d, $^{4}J_{C\text{-}F}$ = 3.3 Hz), 116.4 (d, $^{2}J_{C\text{-}F}$ = 20.7 Hz), 94.3, 43.7, 19.6 (d, $^{4}J_{C\text{-}F}$ = 2.1 Hz). ^{19}F NMR (565 MHz, $CDCl_3$): δ -118.63 (dd, J_1 = 8.5 Hz, J_2 = 4.0 Hz). HRMS (ESI) m/z: $[M+H]^+$ Calcd for $C_{23}H_{18}FN_2O$ 357.1398; Found 357.1397.



4v

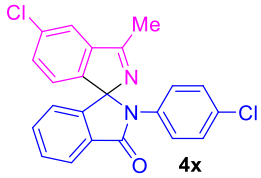
4-Fluoro-3-methyl-2'-(2-morpholinoethyl)spiro[isoindole-1,1'-isoindolin]-3'-one (4v)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow syrup (34.1 mg, 45%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.90 (d, J = 7.6 Hz, 1H), 7.48 (t, J = 7.2 Hz, 1H), 7.41 (t, J = 7.6 Hz, 1H), 7.37-7.32 (m, 1H), 7.14 (t, J = 8.4 Hz, 1H), 6.90 (d, J = 7.2 Hz, 1H), 6.75 (d, J = 7.6 Hz, 1H), 3.63-3.61 (m, 4H), 3.35-3.13 (m, 2H), 2.71 (s, 3H), 2.40-2.30 (m, 6H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 171.3, 168.8, 157.0 (d, $^{1}J_{C\text{-}F}$ = 255.0 Hz), 152.6, 142.2, 132.6 (d, $^{3}J_{C\text{-}F}$ = 6.3 Hz), 132.3, 132.2, 129.5, 126.6 (d, $^{2}J_{C\text{-}F}$ = 16.4 Hz), 124.0, 121 2, 119.1 (d, $^{4}J_{C\text{-}F}$ = 3.3 Hz), 117.0 (d, $^{2}J_{C\text{-}F}$ = 20.1 Hz), 94.6, 66.9, 56.8, 53.5, 37.2, 19.8 (d, $^{4}J_{C\text{-}F}$ = 2.3 Hz). ^{19}F NMR (565 MHz, $CDCl_3$): δ -117.86 (dd, J_1 = 8.3 Hz, J_2 = 4.5 Hz). HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{22}H_{23}FN_3O_2$ 380.1769; Found 380.1767.



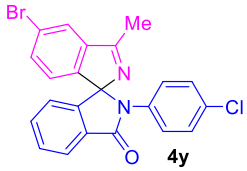
2'-(4-Chlorophenyl)-3,5-dimethylspiro[isoindole-1,1'-isoindolin]-3'-one (4w)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (50.0 mg, 67%), mp 169.7-170.3 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.99 (d, J = 7.2 Hz, 1H), 7.52-7.48 (m, 1H), 7.44 (td, J_1 = 7.2 Hz, J_2 = 0.8 Hz, 1H), 7.29 (s, 1H), 7.18-7.15 (m, 3H), 7.12-7.08 (m, 2H), 7.06 (d, J = 7.6 Hz, 1H), 6.75 (d, J = 7.6 Hz, 1H), 2.50 (s, 3H), 2.42 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 174.5, 168.4, 145.8, 143.1, 140.3, 140.0, 134.4, 132.8, 132.7, 131.8, 131.2, 129.4, 129.0, 128.4, 124.5, 122.27, 122.26, 121.4, 94.8, 21.5, 16.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{23}H_{18}ClN_2O$ 373.1102; Found 373.1100.



5-Chloro-2'-(4-chlorophenyl)-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4x)

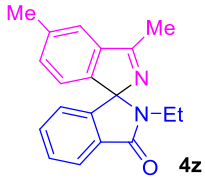
Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (39.3 mg, 50%), mp 203.1-204.0 ºC. ^{1}H NMR (600 MHz, $CDCl_3$): δ 8.01 (d, J = 7.2 Hz, 1H), 7.53 (td, J_1 = 7.2 Hz, J_2 = 0.6 Hz, 1H), 7.48-7.46 (m, 2H), 7.33 (dd, J_1 = 8.4 Hz, J_2 = 1.8 Hz, 1H), 7.20-7.18 (m, 2H), 7.12 (d, J = 8.4 Hz, 1H), 7.10-7.08 (m, 2H), 6.76 (d, J = 7.2 Hz, 1H), 2.52 (s, 3H). ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 173.3, 168.2, 147.1, 142.1, 141.4, 136.0, 134.0, 133.2, 133.0, 131.7, 130.3, 129.8, 129.1, 128.5, 124.8, 123.6, 122.1, 121.4, 94.8, 16.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{22}H_{15}Cl_2N_2O$ 393.0556; Found 393.0559.



5-Bromo-2'-(4-chlorophenyl)-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4y)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (68.3 mg, 78%), mp 203.6-204.1 ºC. ^{1}H NMR (600

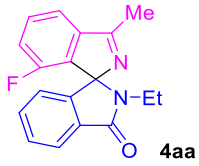
MHz, $CDCl_3$): δ 8.00 (d, *J* = 7.2 Hz, 1H), 7.62 (s, 1H), 7.53 (t, *J* = 7.2 Hz, 1H), 7.48-7.45 (m, 2H), 7.19 (d, *J* = 8.4 Hz, 2H), 7.09-7.06 (m, 3H), 6.75 (d, *J* = 7.8 Hz, 1H), 2.51 (s, 3H). $^{13}C\{^1H\}$ NMR (150 MHz, $CDCl_3$): δ 173.1, 168.2, 147.7, 142.0, 141.7, 134.1, 133.15, 133.12, 133.0, 131.7, 129.7, 129.1, 128.5, 125.0, 124.8, 123.9, 123.8, 121.3, 94.9, 16.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{22}H_{15}BrClN_2O$ 437.0056; Found 437.0050.



4z

2'-Ethyl-3,5-dimethylspiro[isoindole-1,1'-isoindolin]-3'-one (4z)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow syrup (24.4 mg, 42%). ^1H NMR (400 MHz, $CDCl_3$): δ 7.90 (d, *J* = 7.6 Hz, 1H), 7.46-7.42 (m, 1H), 7.40 (s, 1H), 7.36 (td, J_1 = 7.6 Hz, J_2 = 0.8 Hz, 1H), 7.18 (d, *J* = 7.6 Hz, 1H), 6.99 (d, *J* = 7.6 Hz, 1H), 6.70 (d, *J* = 7.6 Hz, 1H), 3.18-3.03 (m, 2H), 2.59 (s, 3H), 2.47 (s, 3H), 1.01 (t, *J* = 7.2 Hz, 3H). $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ 174.1, 168.7, 145.8, 143.0, 140.4, 139.9, 132.7, 131.9, 131.0, 129.1, 123.7, 122.6, 122.1, 121.1, 94.0, 34.9, 21.5, 16.8, 14.6. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{19}H_{19}N_2O$ 291.1492; Found 291.1496.

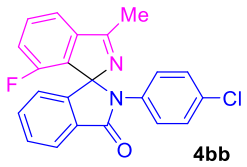


4aa

2'-Ethyl-7-fluoro-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4aa)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow syrup (32.4 mg, 55%). ^1H NMR (400 MHz, $CDCl_3$): δ 7.92 (d, *J* = 7.6 Hz, 1H), 7.55-7.50 (m, 1H), 7.48 (td, J_1 = 6.8 Hz, J_2 = 0.8 Hz, 1H), 7.42-7.38 (m, 2H), 7.02 (t, *J* = 8.0 Hz, 1H), 6.76 (d, *J* = 7.6 Hz, 1H), 3.27-3.09 (m, 2H), 2.61 (s, 3H), 1.02 (t, *J* = 7.2 Hz, 3H). $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$): δ 173.0, 168.4, 157.6 (d, $^1J_{C\text{-}F}$ = 253.1 Hz), 143.0 (d, $^3J_{C\text{-}F}$ = 3.7 Hz), 141.0, 133.9 (d, $^2J_{C\text{-}F}$ = 15.9 Hz), 132.9, 132.7 (d, $^3J_{C\text{-}F}$ = 6.6 Hz), 132.0, 129.5, 124.0, 120.8, 117.7 (d, $^2J_{C\text{-}F}$ = 19.9 Hz), 117.5 (d,

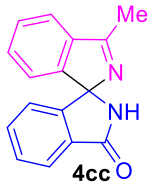
$^{4}J_{C\text{-}F}$ = 4.5 Hz), 92.9 (d, $^{3}J_{C\text{-}F}$ = 2.1 Hz), 34.9, 16.9, 14.0. ^{19}F NMR (376 MHz, $CDCl_3$): δ -118.29 (dd, J_1 = 7.9 Hz, J_2 = 3.8 Hz). HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{18}H_{16}FN_2O$ 295.1241; Found 295.1242.



4bb

2'-(4-Chlorophenyl)-7-fluoro-3-methylspiro[isoindole-1,1'-isoindolin]-3'-one (4bb)

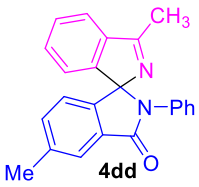
Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (64.1 mg, 85%), mp 210.2-210.9 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 8.02 (d, J = 7.2 Hz, 1H), 7.54 (td, J_1 = 7.2 Hz, J_2 = 0.4 Hz, 1H), 7.48 (td, J_1 = 7.6 Hz, J_2 = 0.8 Hz, 1H), 7.42 (td, J_1 = 8.0 Hz, J_2 = 4.8 Hz, 1H), 7.27-7.25 (m, 1H), 7.22-7.17 (m, 4H), 6.98 (t, J = 8.4 Hz, 1H), 6.82 (d, J = 7.6 Hz, 1H), 2.54 (s, 3H). ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 173.4, 168.0, 157.3 (d, $^{1}J_{C\text{-}F}$ = 252.8 Hz), 142.9 (d, $^{3}J_{C\text{-}F}$ = 3.3 Hz), 141.0, 134.1, 133.5 (d, $^{2}J_{C\text{-}F}$ = 16.1 Hz), 133.4, 132.82, 132.81 (d, $^{3}J_{C\text{-}F}$ = 6.5 Hz), 131.9, 129.8, 129.2, 128.8, 124.8, 121.2, 117.7 (d, $^{2}J_{C\text{-}F}$ = 22.1 Hz), 117.6, 94.0 (d, $^{3}J_{C\text{-}F}$ = 3.9 Hz), 16.9. ^{19}F NMR (376 MHz, $CDCl_3$): δ -118.93 (dd, J_1 = 7.9 Hz, J_2 = 3.8 Hz). HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{22}H_{15}ClFN_2O$ 377.0851; Found 377.0850.



4cc

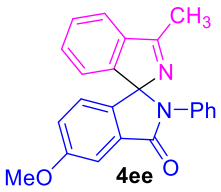
3-Methylspiro[isoindole-1,1'-isoindolin]-3'-one (4cc)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow solid (17.9 mg, 36%), mp 229.5-230.1 ºC. ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.90 (d, J = 7.2 Hz, 1H), 7.57 (d, J = 7.6 Hz 1H), 7.49 (t, J = 7.6 Hz, 2H), 7.44-7.38 (m, 2H), 7.21 (d, J = 7.6 Hz, 1H), 6.75 (d, J = 7.6 Hz, 1H), 6.30 (s, 1H), 2.57 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 173.9, 170.7, 149.3, 144.5, 139.4, 132.7, 131.6, 130.3, 129.7, 129.4, 124.3, 122.6, 121.6, 121.5, 90.6, 16.9. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{16}H_{13}N_2O$ 249.1022; Found 249.1027.



3,5'-Dimethyl-2'-phenylspiro[isoindole-1,1'-isoindolin]-3'-one (4dd)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow syrup (50.1 mg, 74%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.81 (s, 1H), 7.45 (d, *J* = 7.2 Hz 1H), 7.39 (t, *J* = 7.2 Hz 1H), 7.33 (t, *J* = 7.2 Hz 1H), 7.25-7.10 (m, 7H), 6.64 (d, *J* = 8.0 Hz, 1H), 2.50 (s, 3H), 2.44 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 173.9, 168.6, 149.2, 140.0, 139.9, 139.6, 135.9, 133.6, 132.2, 130.1, 129.4, 128.7, 127.2, 127.1, 124.7, 122.6, 121.5, 121.1, 95.1, 21.4, 16.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{23}H_{19}N_2O$ 339.1492; Found 339.1495.



5'-Methoxy-3-methyl-2'-phenylspiro[isoindole-1,1'-isoindolin]-3'-one (4ee)

Eluent: petroleum ether/ethyl acetate (3:1). Yellow syrup (39.7 mg, 56%). ^{1}H NMR (400 MHz, $CDCl_3$): δ 7.49 (d, *J* = 2.0 Hz, 1H), 7.44 (d, *J* = 6.8 Hz 1H), 7.39 (t, *J* = 7.2 Hz, 1H), 7.34 (t, *J* = 7.2 Hz, 1H), 7.22-7.11 (m, 6H), 6.99 (dd, J_1 = 8.4 Hz, J_1 = 2.4 Hz, 1H), 6.65 (d d, *J* = 8.4 Hz, 1H), 3.88 (s, 3H), 2.50 (s, 3H). ^{13}C{^{1}H} NMR (100 MHz, $CDCl_3$): δ 173.9, 168.4, 161.0, 149.1, 139.8, 135.8, 134.7, 133.6, 130.1, 129.4, 128.7, 127.2, 122.6, 122.4, 121.5, 121.0, 107.3, 94.9, 55.9, 16.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{23}H_{19}N_2O_2$ 355.1441; Found 355.1443.

3. Gram-Scale Synthesis of 3k

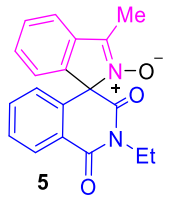
To a reaction tube equipped with a stir bar were charged with **1a** (0.81 g, 5.0 mmol), NaOAc (0.82 g, 10.0 mmol), $[RhCp^*Cl_2]_2$ (77.3 mg, 0.125 mmol), **2k** (1.29 g, 6.0 mmol) and DCE (20 mL). The tube was then sealed, and the mixture was stirred at 80 °C under argon for 6 h. Upon completion, it was cooled to room temperature, filtered through a pad of celite and concentrated under reduced pressure. The residue was purified

by silica gel column chromatography using petroleum ether/ethyl acetate (3:1) as eluent to afford **3k** (1.142 g, 75%).

4. Structural elaborations of 3k

4.1 Synthesis of 5 [3]

To a solution of **3k** (60.9 mg, 0.2 mmol) in CH_2Cl_2 (2 mL) was added *m*-chloroperoxybenzoic acid (69.0 mg, 0.4 mmol) at 0 °C. The mixture was stirred at room temperature for 24 h. Upon completion, it was quenched with saturated Na_2CO_3 aqueous solution and extracted with dichloromethane (3 × 10 mL). The combined organic layers were washed with brine, dried over anhydrous Na_2SO_4, filtered and concentrated under reduced pressure. The residue was purified by column chromatography on silica gel using petroleum ether/ethyl acetate (3:1) as eluent to afford **5** (28.8 mg, 45%).



5

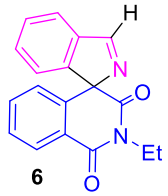
2'-Ethyl-3-methyl-1',3'-dioxo-2',3'-dihydro-1'*H*-spiro[isoindole-1,4'-isoquinoline] 2-oxide (5)

White solid. mp 180.2-181.0 ºC. ^{1}H NMR (600 MHz, $CDCl_3$): δ 8.35 (dd, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H), 7.89-7.54 (m, 2H), 7.46-7.45 (m, 2H), 7.31-7.28 (m, 1H), 6.94 (d, J = 7.8 Hz, 1H), 6.90-6.89 (m, 1H), 4.15-4.03 (m, 2H), 2.53 (s, 3H), 1.24 (t, J = 7.2 Hz, 3H). ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 165.0, 163.2, 145.7, 139.3, 136.3, 134.7, 133.0, 130.12, 130.05, 129.5, 128.9, 126.5, 125.4, 121.2, 120.2, 83.9, 36.7, 13.1, 9.8. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{19}H_{17}N_2O_3$ 321.1234; Found 321.1229.

4.2 Synthesis of 6[4]

To a solution of **3k** (60.9 mg, 0.2 mmol) in THF (2 mL) was added SeO_2 (33.3 mg, 0.3 mmol). The mixture was refluxed for 3 h. Upon completion, it was cooled to room temperature, diluted with water and extracted with ethyl acetate (3 × 10 mL). The organic layer was dried over anhydrous Na_2SO_4 and concentrated under

reduced pressure. The residue was purified by column chromatography on silica gel with petroleum ether/ethyl acetate (3:1) as eluent to give **6** (23.2 mg, 40%).



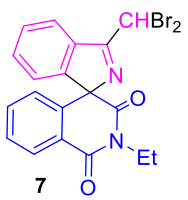
6

2'-Ethyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (6)

Yellow syrup. ^{1}H NMR (600 MHz, $CDCl_3$): δ 8.91 (s, 1H), 8.35 (dd, J_1 = 7.8 Hz, J_2 = 1.2 Hz, 1H), 7.70 (d, J = 7.8 Hz, 1H), 7.51-7.48 (m, 1H), 7.46 (td, J_1 = 7.2 Hz, J_2 = 0.6 Hz, 1H), 7.43 (td, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H), 7.36 (td, J_1 = 7.2 Hz, J_2 = 0.6 Hz, 1H), 7.16 (dd, J_1 = 7.2 Hz, J_2 = 0.6 Hz, 1H), 6.64 (dd, J_1 = 7.8 Hz, J_2 = 0.6 Hz, 1H), 4.15-4.02 (m, 2H), 1.23 (t, J = 7.2 Hz, 3H). ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 168.4, 167.4, 164.1, 153.3, 139.2, 135.1, 134.1, 130.3, 129.4, 129.3, 129.0, 125.3, 125.1, 123.2, 121.7, 83.8, 36.4, 13.1. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{18}H_{15}N_2O_2$ 291.1128; Found 291.1130.

4.3 Synthesis of 7[5]

To a reaction tube equipped with a stir bar were charged with **3k** (60.9 mg, 0.2 mmol), *N*-bromosuccinimide (37.4 mg, 0.21 mmol), benzoyl peroxide (4.8 mg, 0.02 mmol) and CCl_4 (2 mL). The tube was then sealed and the resulting mixture was stirred at 50 °C under air for 0.5 h. Upon completion, it was cooled to room temperature, diluted with water and extracted with dichloromethane (3 × 10 mL). The combined organic layers were washed with aqueous solution of $NaHCO_3$, aqueous solution of $Na_2S_2O_3$ and diluted HCl (0.1 M), dried over anhydrous Na_2SO_4, filtered and concentrated under reduced pressure. The residue was purified by column chromatography on silica gel using petroleum ether/ethyl acetate (10:1) as eluent to afford **7** (55.5 mg, 60%).



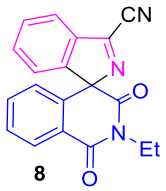
7

3-(Dibromomethyl)-2'-ethyl-1'*H*-spiro[isoindole-1,4'-isoquinoline]-1',3'(2'*H*)-dione (7)

Yellow syrup. ^{1}H NMR (600 MHz, $CDCl_3$): δ 8.35 (d, J = 7.8 Hz, 1H), 8.14 (d, J = 7.8 Hz, 1H), 7.54-7.50 (m, 2H), 7.45 (td, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H), 7.39 (t, J = 7.2 Hz, 1H), 7.14 (d, J = 7.8 Hz, 1H), 6.81 (s, 1H), 6.63 (d, J = 7.2 Hz, 1H), 4.14-4.01 (m, 2H), 1.22 (t, J = 6.6 Hz, 3H). ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 174.0, 166.9, 163.8, 154.7, 134.8, 134.6, 134.3, 130.7, 129.6, 129.2, 129.0, 125.15, 125.07, 124.6, 121.9, 81.1, 36.5, 33.7, 13.1. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{19}H_{15}Br_2N_2O_2$ 460.9495; Found 460.9499.

4.4 Synthesis of 8[6]

To a reaction tube equipped with a stir bar were added with **3k** (60.9 mg, 0.2 mmol), azidotrimethylsilane (46.1 mg, 0.4 mmol), sodium bromide (41.2 mg, 0.4 mmol), iodobenzene diacetate (193.3 mg, 0.6 mmol,) and DMSO (2 mL). The tube was then sealed, and the mixture was stirred at room temperature under air for 12 h. Upon completion, it was diluted with water and extracted with ethyl acetate (3 × 10 mL). The combined organic layers were washed with brine, dried over anhydrous Na_2SO_4, filtered and concentrated under reduced pressure. The residue was purified by column chromatography on silica gel with petroleum ether/ethyl acetate (3:1) as eluent to afford **8** (35.9 mg, 57%)**.**



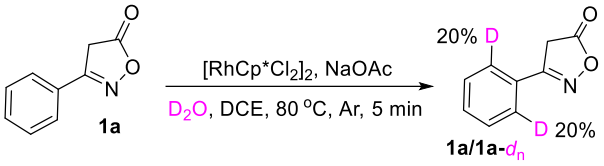
8

2'-Ethyl-1',3'-dioxo-2',3'-dihydro-1'*H*-spiro[isoindole-1,4'-isoquinoline]-3-carbonitrile (8)

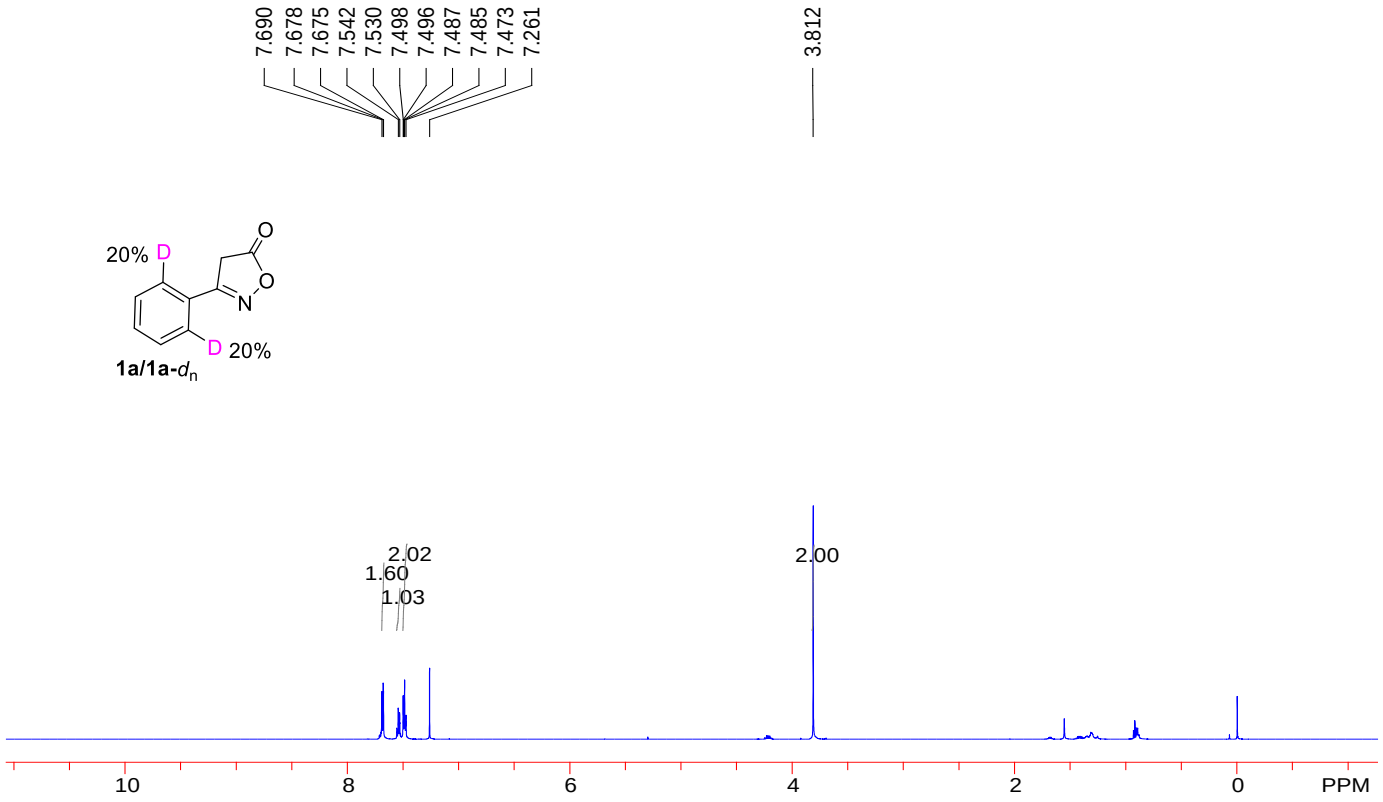
Yellow solid. mp 159.2-159.7 ºC. ^{1}H NMR (600 MHz, $CDCl_3$): δ 8.38 (dd, J_1 = 7.8 Hz, J_2 = 1.2 Hz, 1H), 7.85 (d, J = 7.8 Hz, 1H), 7.60 (t, J = 7.2 Hz, 1H), 7.58-7.55 (m, 1H), 7.51 (td, J_1 = 7.2 Hz, J_2 = 0.6 Hz, 1H), 7.48 (td, J_1 = 7.2 Hz, J_2 = 1.2 Hz, 1H), 7.25 (d, J = 7.8 Hz, 1H), 6.55 (d, J = 7.8 Hz, 1H), 4.14-4.01 (m, 2H), 1.23 (t, J = 7.2 Hz, 3H). ^{13}C{^{1}H} NMR (150 MHz, $CDCl_3$): δ 165.3, 163.4, 153.3, 153.0, 136.8, 134.4, 132.8, 132.0, 130.1, 129.91, 129.85, 125.4, 125.3, 122.50, 122.45, 112.1, 84.6, 36.7, 13.1. HRMS (ESI-TOF) m/z: $[M+H]^+$ Calcd for $C_{19}H_{14}N_3O_2$ 316.1081; Found 316.1085.

III. Mechanism studies

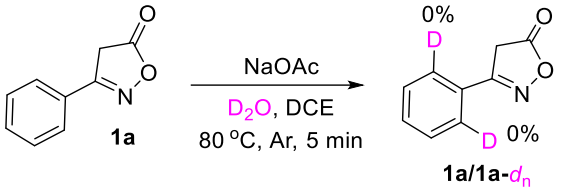
1. Studies on the reversibility of C−H bond activation



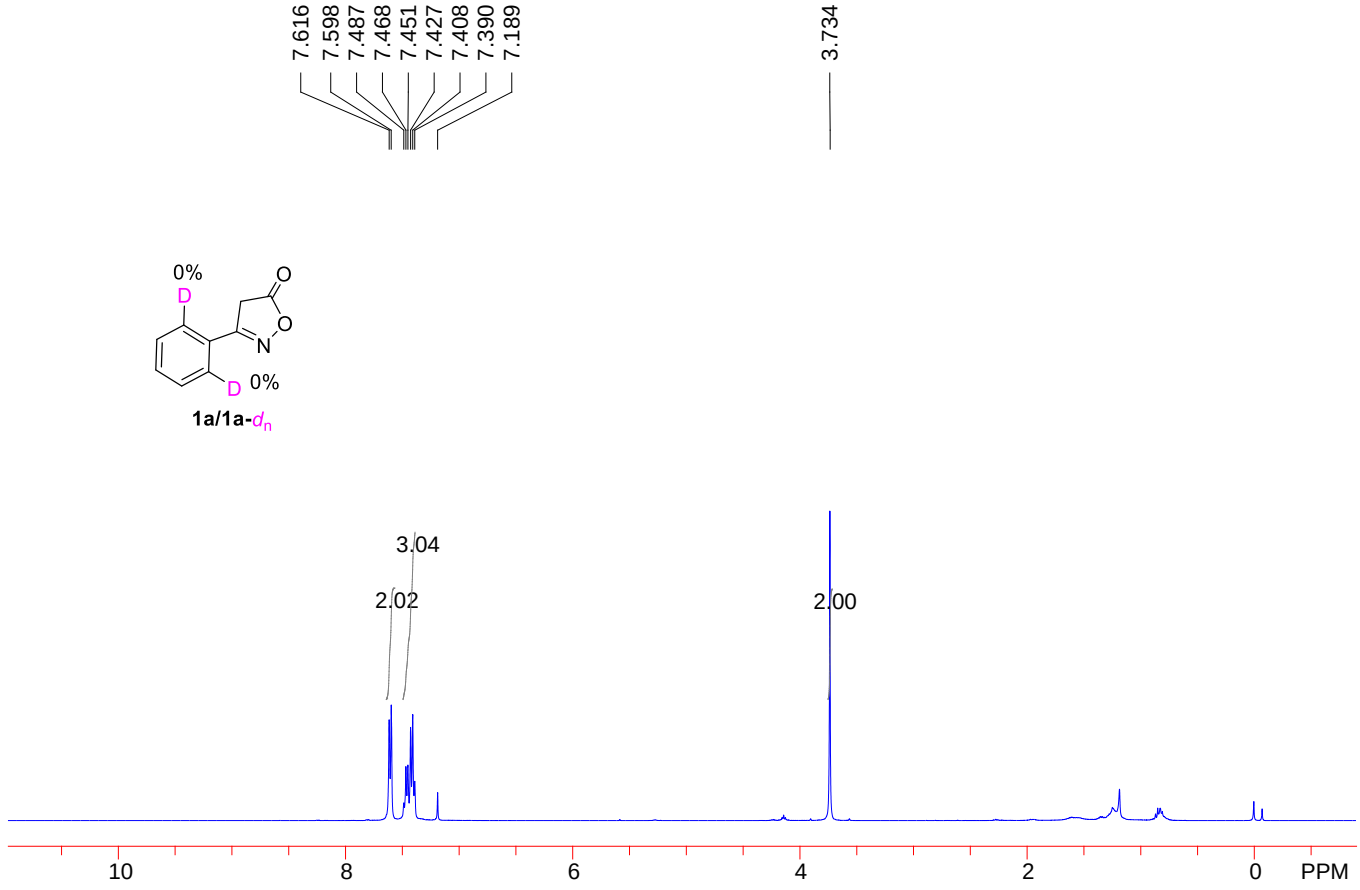
To a reaction tube equipped with a stir bar were charged with **1a** (32.3 mg, 0.2 mmol), NaOAc (32.8 mg, 0.4 mmol), $[RhCp^*Cl_2]_2$ (6.2 mg, 0.01 mmol), D_2O (36 μL, 2.0 mmol) and DCE (2 mL). The tube was then sealed, and the resulting mixture was stirred at 80 °C under argon for 5 min. Afterwards, it was cooled to room temperature, filtered through a pad of celite and concentrated under reduced pressure. The residue was purified by silica gel column chromatography using petroleum ether/ethyl acetate (10:1) as eluent to give a mixture of **1a** and **1a**-d_n. Upon analyzing the ^{1}H NMR spectrum of the mixture, the deuteration percentage on the *ortho*-positions of the phenyl ring of **1a** was determined to be 20%.



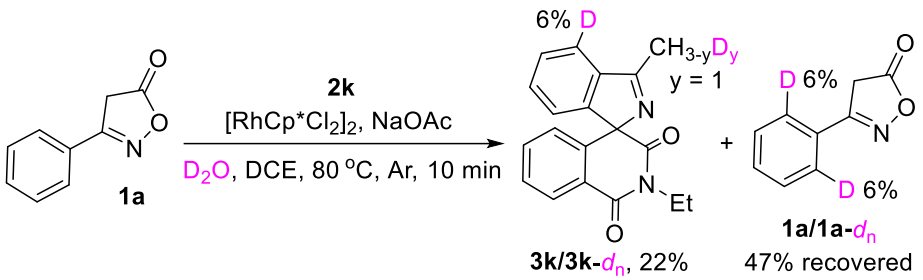
The ^{1}H NMR spectrum of the products obtained from the H/D exchange experiment (I)



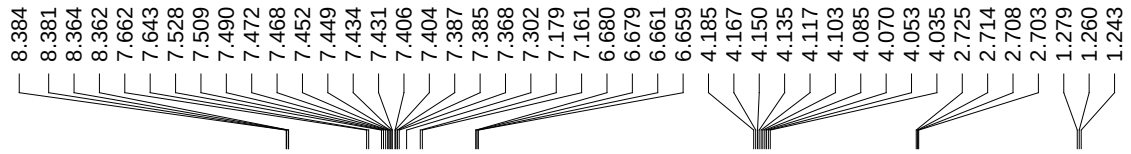
To a reaction tube equipped with a stir bar were charged with **1a** (32.3 mg, 0.2 mmol), NaOAc (32.8 mg, 0.4 mmol), D_2O (36 μL, 2.0 mmol) and DCE (2 mL). The tube was then sealed, and the resulting mixture was stirred at 80 °C under argon for 5 min. Afterwards, it was cooled to room temperature, filtered through a pad of celite and concentrated under reduced pressure. The residue was purified by silica gel column chromatography using petroleum ether/ethyl acetate (10:1) as eluent to give a mixture of **1a** and **1a**-d_n. Upon analyzing the ^{1}H NMR spectrum of the mixture, there is no deuteration was observed with **1a**.

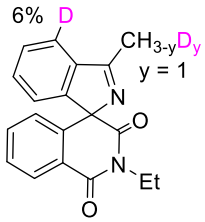


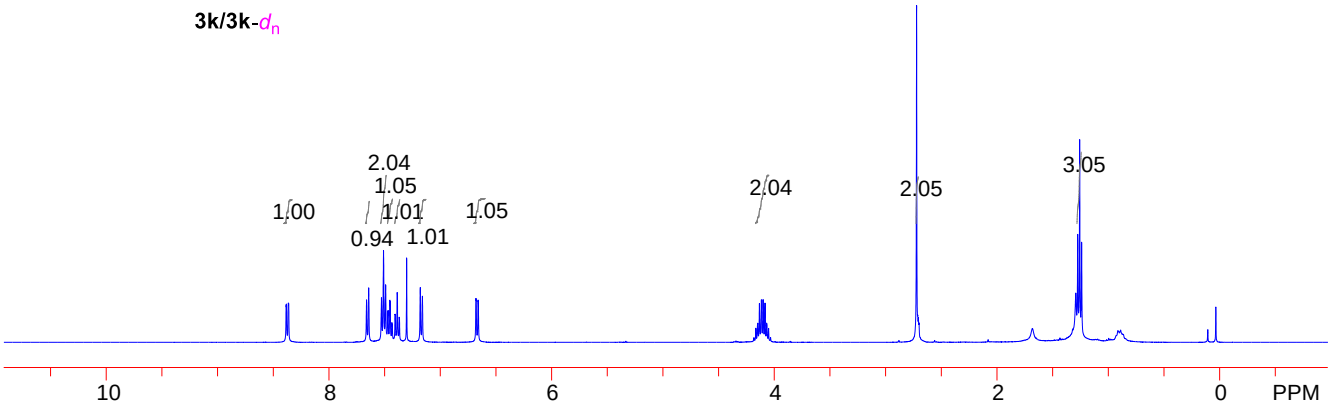
The ^{1}H NMR spectrum of the products obtained from the H/D exchange experiment (I')

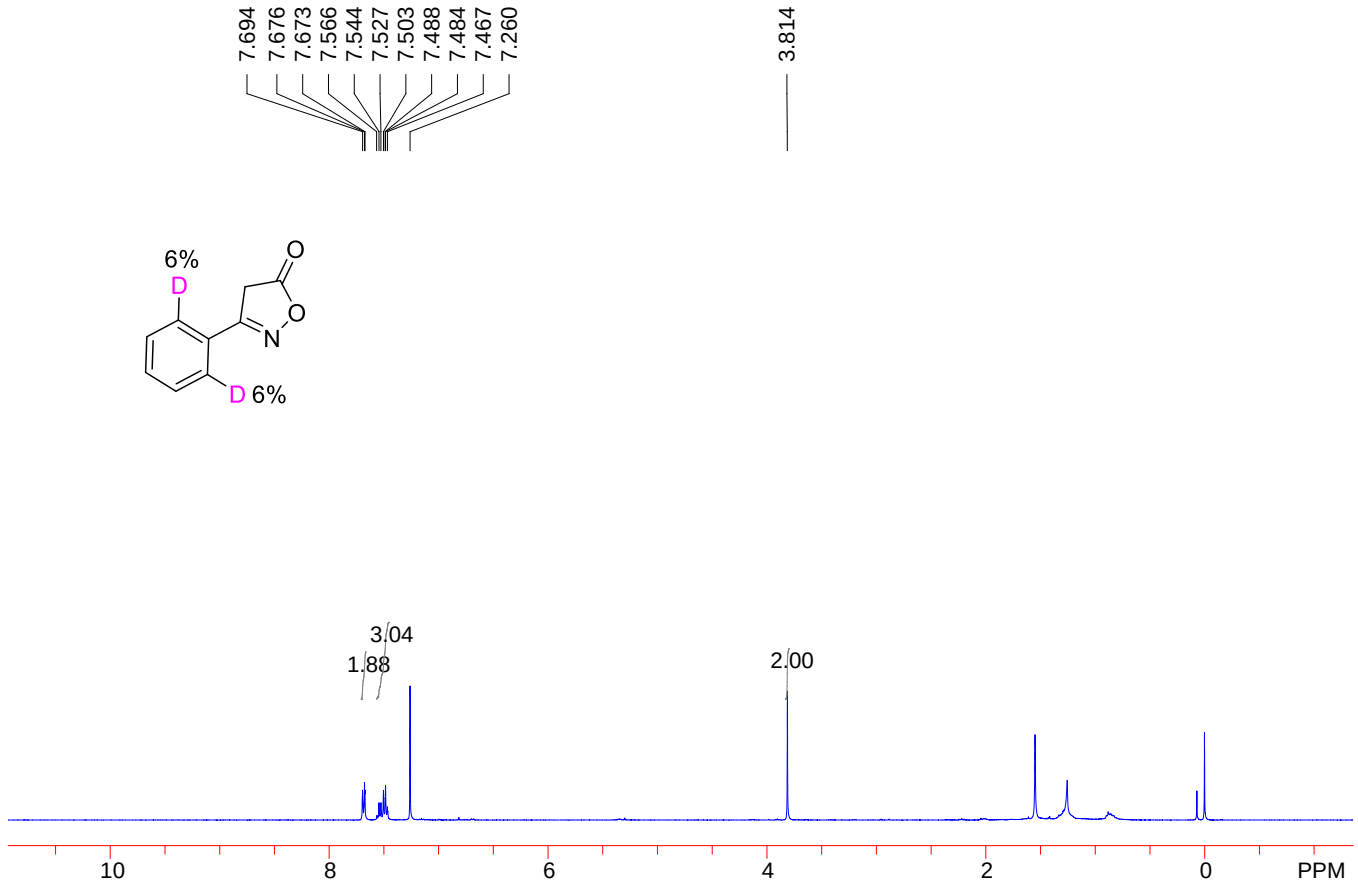


To a reaction tube equipped with a stir bar were added **1a** (32.3 mg, 0.2 mmol), NaOAc (32.8 mg, 0.4 mmol), $[RhCp^*Cl_2]_2$ (6.2 mg, 0.01 mmol), **2k** (51.7 mg, 0.24 mmol), D_2O (36 μL, 2.0 mmol) and DCE (2 mL). The tube was then sealed and the mixture was stirred at 80 °C under argon for 10 min. Afterwards, it was cooled to room temperature, filtered through a pad of celite and concentrated under reduced pressure. The residue was purified by column chromatography on silica gel using petroleum ether/ethyl acetate (3:1) as eluent to give a mixture of **3k** and **3k**-d_n in 22% yield. Upon analyzing the ^{1}H NMR spectrum of the mixture, it is confirmed that there is 6% H/D exchange at the unreacted *ortho*-site of the phenyl moiety of product **3k**. Meanwhile, **1a** was recovered in 47% yield with 6% deuteration H/D exchange at the *ortho*-positions of the phenyl ring.



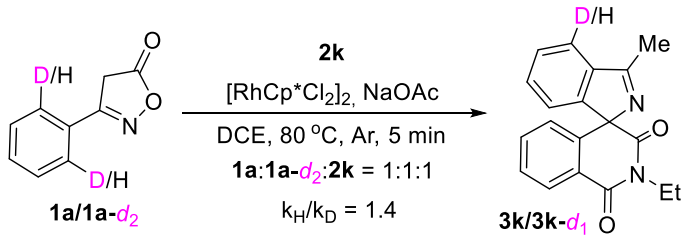




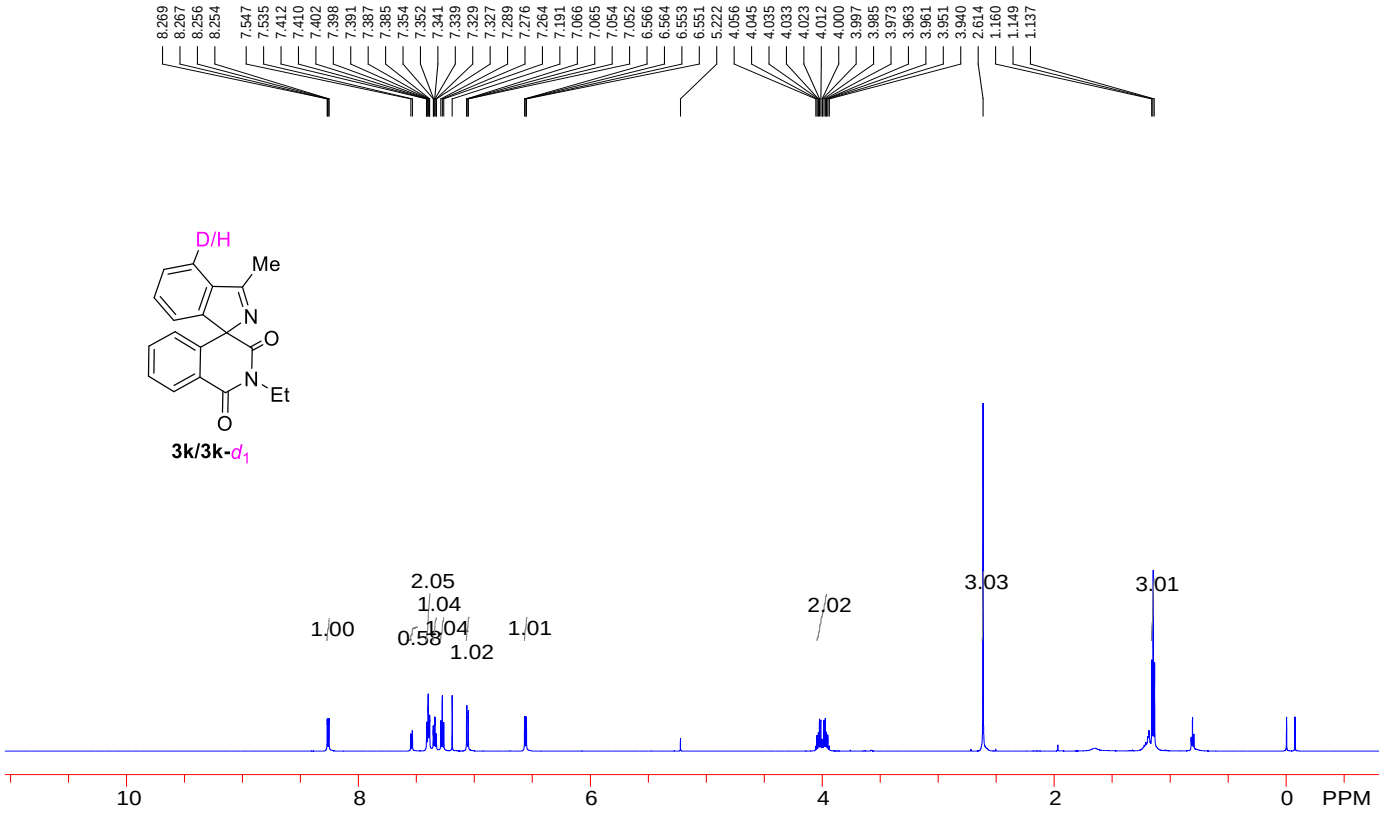


The ^{1}H NMR spectrum of the products obtained from the H/D exchange experiment (II)

2. Kinetic isotope effect study

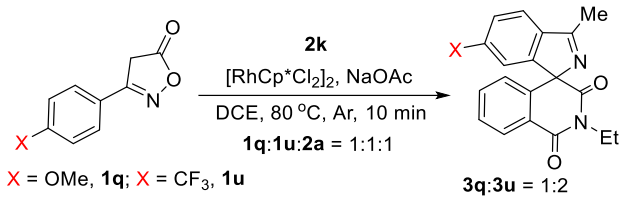


To a reaction tube equipped with a stir bar were added **1a**-d_2 (32.6 mg, 0.2 mmol), **1a** (32.3 mg, 0.2 mmol), NaOAc (32.8 mg, 0.4 mmol), $[RhCp^*Cl_2]_2$ (6.2 mg, 0.01 mmol), **2k** (51.7 mg, 0.2 mmol) and DCE (2 mL). The tube was then sealed, and the mixture was stirred at 80 °C under argon for 5 min. Afterwards, it was cooled to room temperature, filtered through a pad of celite and concentrated under reduced pressure. The residue was purified by silica gel column chromatography using petroleum ether/ethyl acetate (3:1) as eluent to give a mixture of **3k** and **3k**-d_1. Upon analyzing the ^{1}H NMR spectrum of the product, the ratio of **3k** to **3k**-d_1 was determined to be 0.58:0.42. Accordingly, the intermolecular KIE value (k_H/k_D) was calculated to be 1.4.

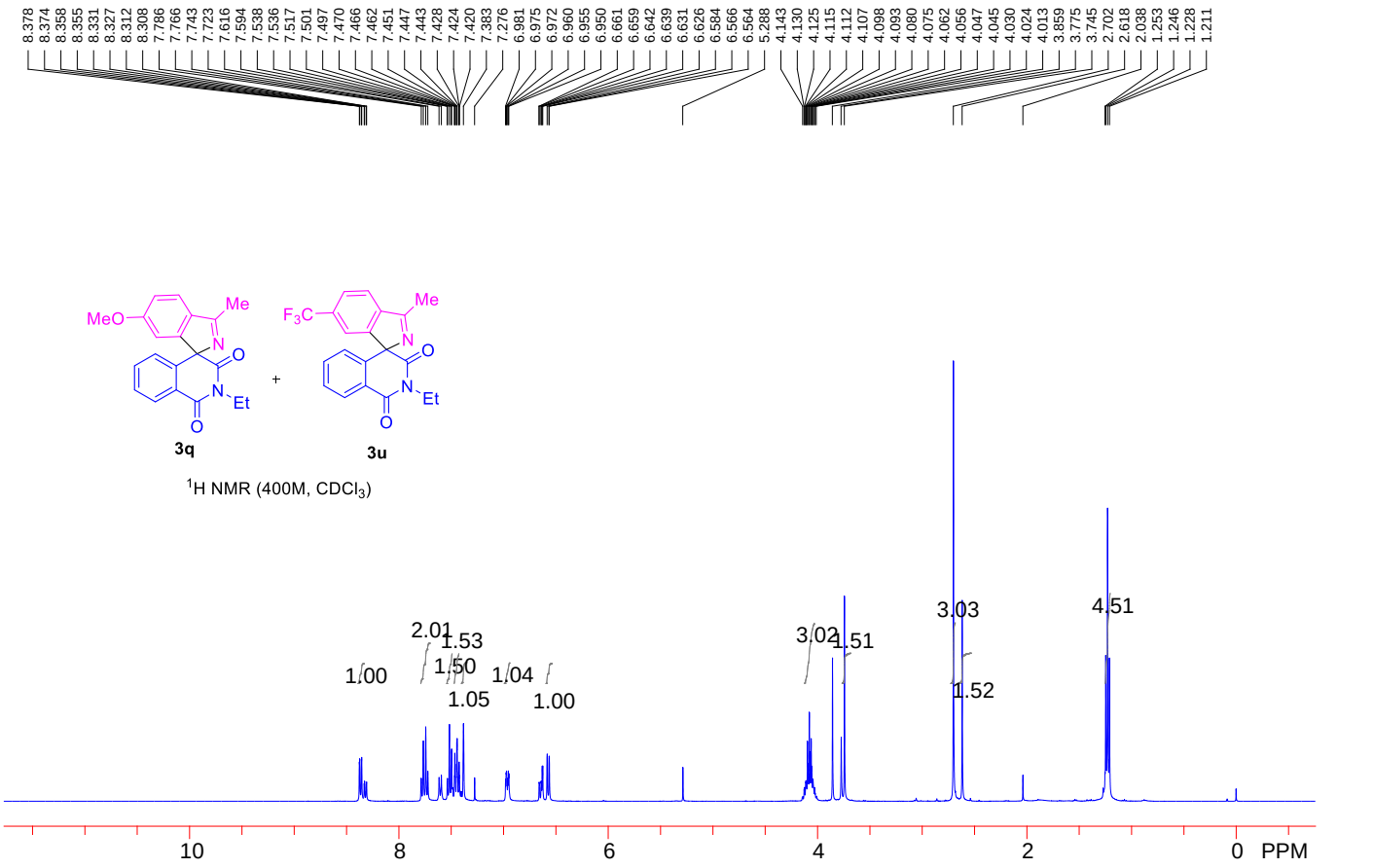


The ^{1}H NMR spectrum of products obtained from the intermolecular KIE experiment (III)

3. Competing experiments between 1q and 1u

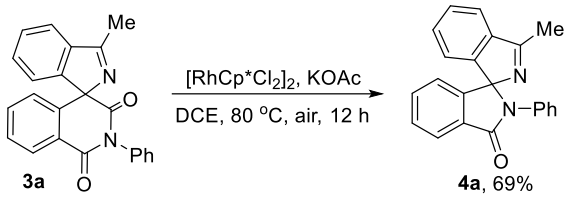


To a reaction tube equipped with a stir bar were added **1q** (38.2 mg, 0.2 mmol), **1u** (45.8 mg, 0.2 mmol), NaOAc (32.8 mg, 0.4 mmol), $[RhCp^*Cl_2]_2$ (6.2 mg, 0.01 mmol), **2k** (51.7 mg, 0.24 mmol) and DCE (2 mL). The tube was then sealed, and the mixture was stirred at 80 °C under argon for 10 min. Afterwards, it was cooled to room temperature, filtered through a pad of celite and concentrated under reduced pressure. The residue was purified by silica gel column chromatography using petroleum ether/ethyl acetate (3:1) as eluent to afford a mixture of **3q** and **3u**. Upon analyzing the ^{1}H NMR spectrum of the mixture, the ratio of **3q** to **3u** was determined to be about 1:2.

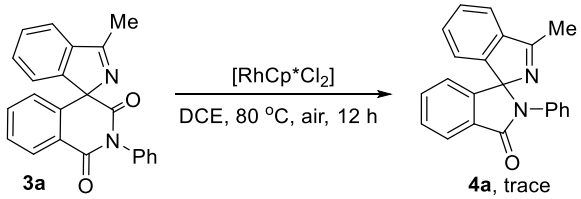


The ^{1}H NMR spectrum of the mixture of **3q** and **3u** obtained from the competing experiments

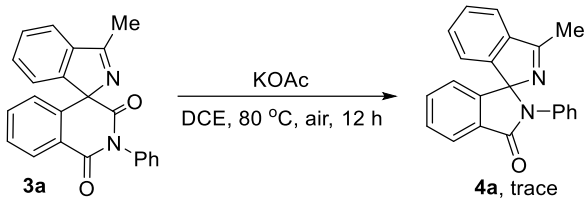
4. Control Experiments to Elucidate the Mechanism



To a reaction tube equipped with a stir bar were added **3a** (70.5 mg, 0.2 mmol), KOAc (39.3 mg, 0.4 mmol), $[RhCp^*Cl_2]_2$ (6.2 mg, 0.01 mmol) and DCE (2 mL). The tube was then sealed and the mixture was stirred at 80 °C under air for 12 h. Afterwards, it was cooled to room temperature, filtered through a pad of celite and concentrated under reduced pressure. The residue was purified by column chromatography on silica gel using petroleum ether/ethyl acetate (3:1) as eluent to afford **4a** (44.8 mg, 69%).

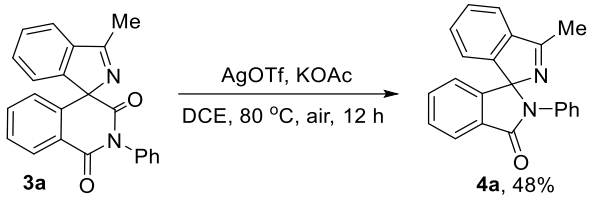


To a reaction tube equipped with a stir bar were added **3a** (70.5 mg, 0.2 mmol), $[RhCp^*Cl_2]_2$ (6.2 mg, 0.01 mmol) and DCE (2 mL). The tube was then sealed and the mixture was stirred at 80 °C under air for 12 h. Afterwards, it was cooled to room temperature, filtered through a pad of celite and concentrated under reduced pressure. The residue was purified by column chromatography on silica gel using petroleum ether/ethyl acetate (3:1) as eluent to afford product **4a** in a trace amount.

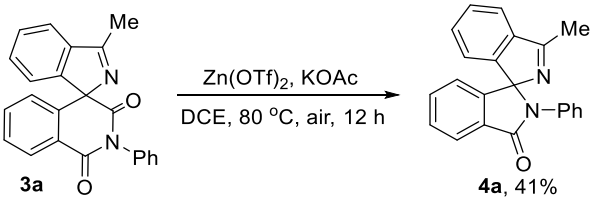


To a reaction tube equipped with a stir bar were added **3a** (70.5 mg, 0.2 mmol), KOAc (39.3 mg, 0.4 mmol) and DCE (2 mL). The tube was then sealed and the mixture was stirred at 80 °C under air for 12 h. Afterwards, it was cooled to room temperature, filtered through a pad of celite and concentrated under reduced pressure. The residue was purified by column chromatography on silica gel using petroleum ether/ethyl acetate (3:1) as

eluent to afford product **4a** in a trace amount.

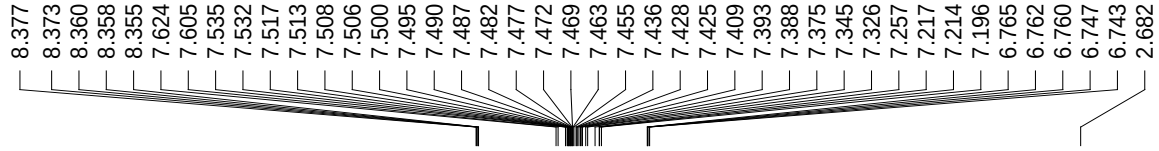


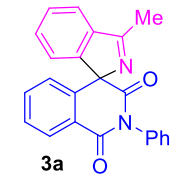
To a reaction tube equipped with a stir bar were added **3a** (70.5 mg, 0.2 mmol), AgOTf (51.4 mg, 0.2 mmol), KOAc (39.3 mg, 0.4 mmol) and DCE (2 mL). The tube was then sealed and the mixture was stirred at 80 °C under air for 12 h. Afterwards, it was cooled to room temperature, filtered through a pad of celite and concentrated under reduced pressure. The residue was purified by column chromatography on silica gel using petroleum ether/ethyl acetate (3:1) as eluent to afford **4a** (31.1 mg, 48%).



To a reaction tube equipped with a stir bar were added **3a** (70.5 mg, 0.2 mmol), $Zn(OTf)_2$ (72.7 mg, 0.2 mmol), KOAc (39.3 mg, 0.4 mmol) and DCE (2 mL). The tube was then sealed and the mixture was stirred at 80 °C under air for 12 h. Afterwards, it was cooled to room temperature, filtered through a pad of celite and concentrated under reduced pressure. The residue was purified by column chromatography on silica gel using petroleum ether/ethyl acetate (3:1) as eluent to afford **4a** (26.6 mg, 41%).

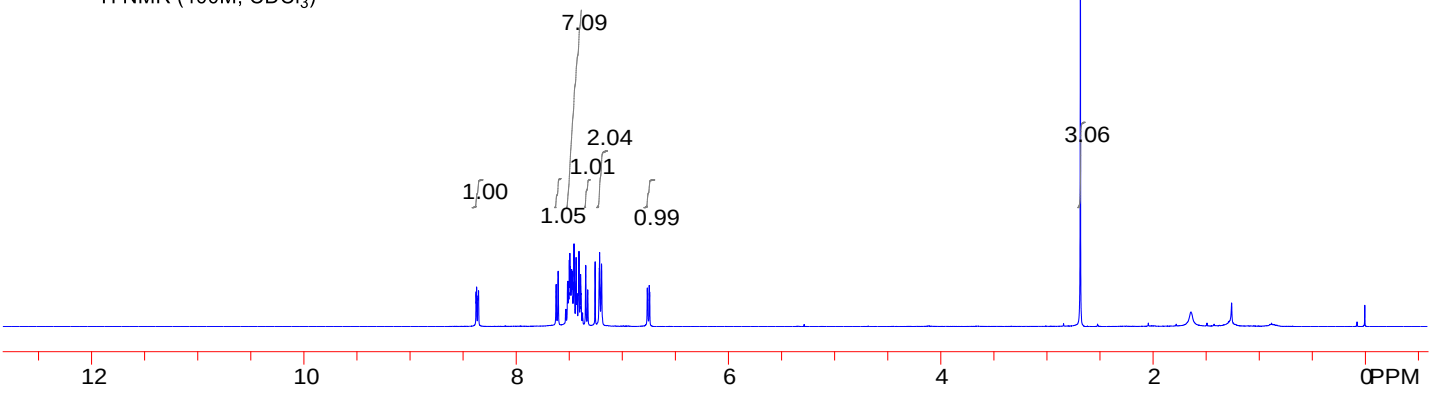
IV. NMR spectra of 3a-3ee

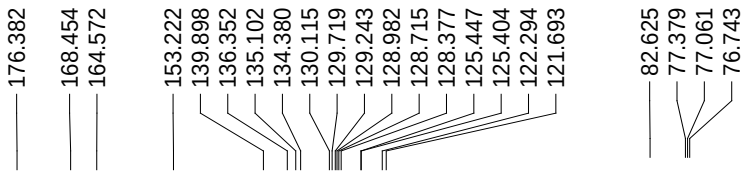


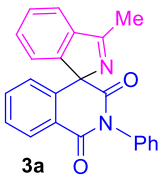


3a

^{1}H NMR (400M, $CDCl_3$)

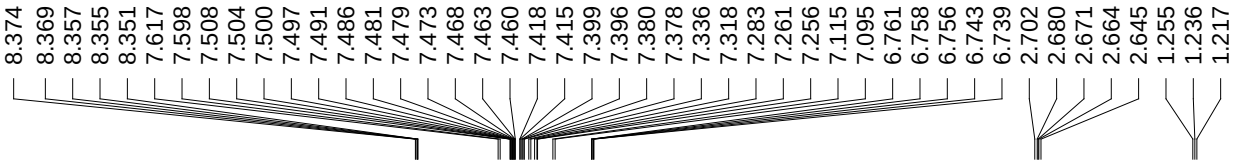


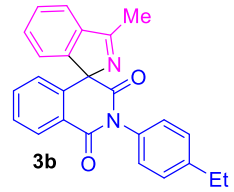




3a

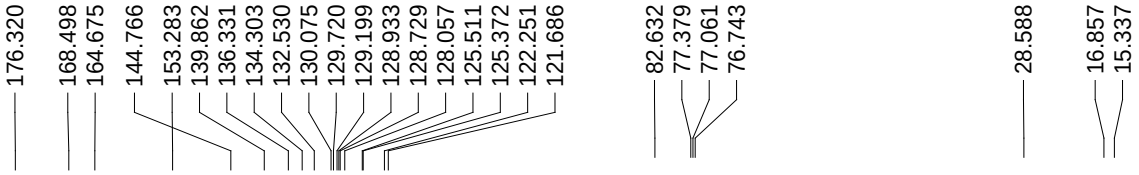
^{13}C{^{1}H} NMR (100M, $CDCl_3$)

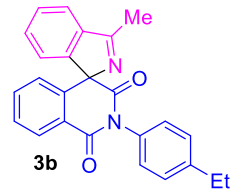




3b

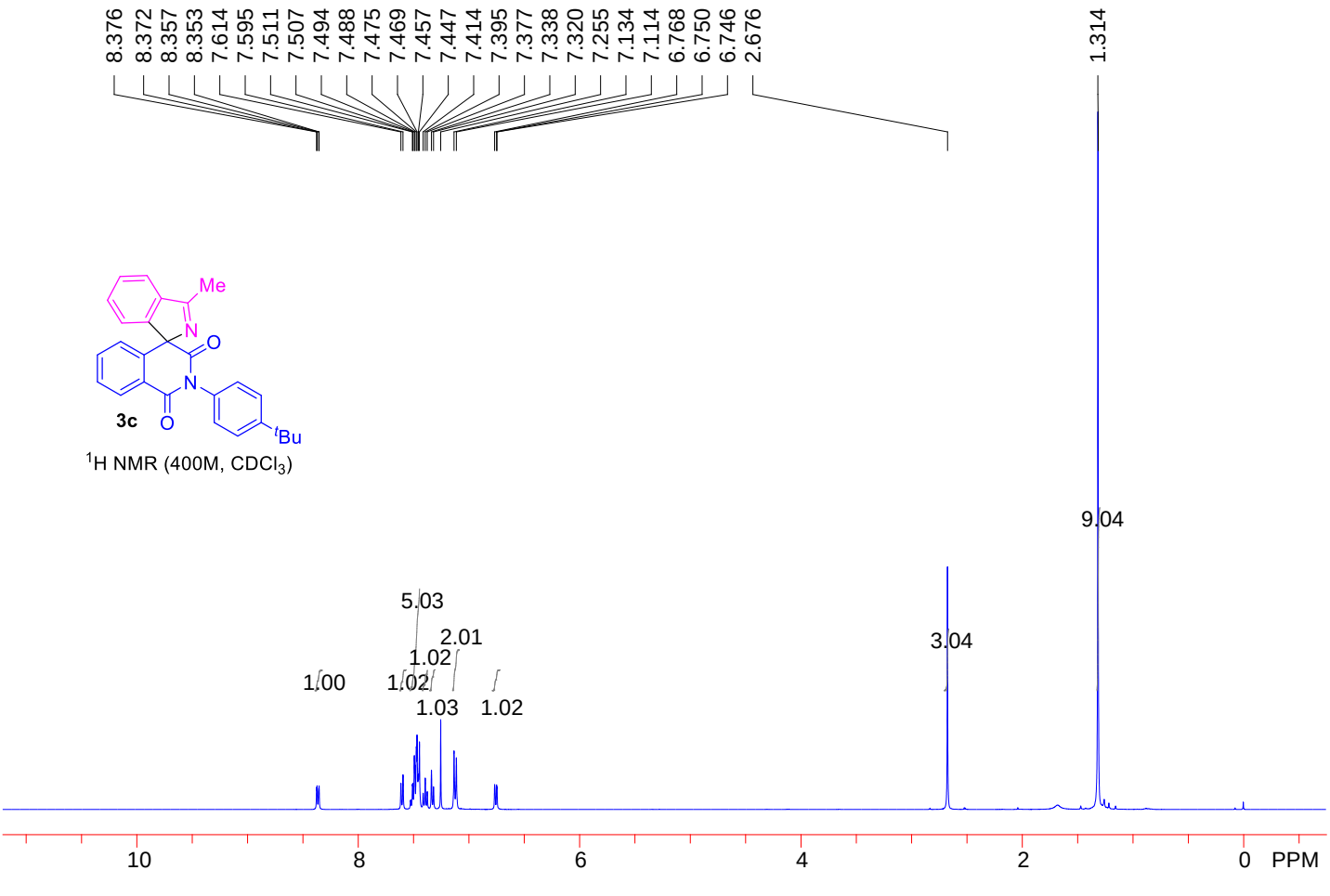
^{1}H NMR (400M, $CDCl_3$)



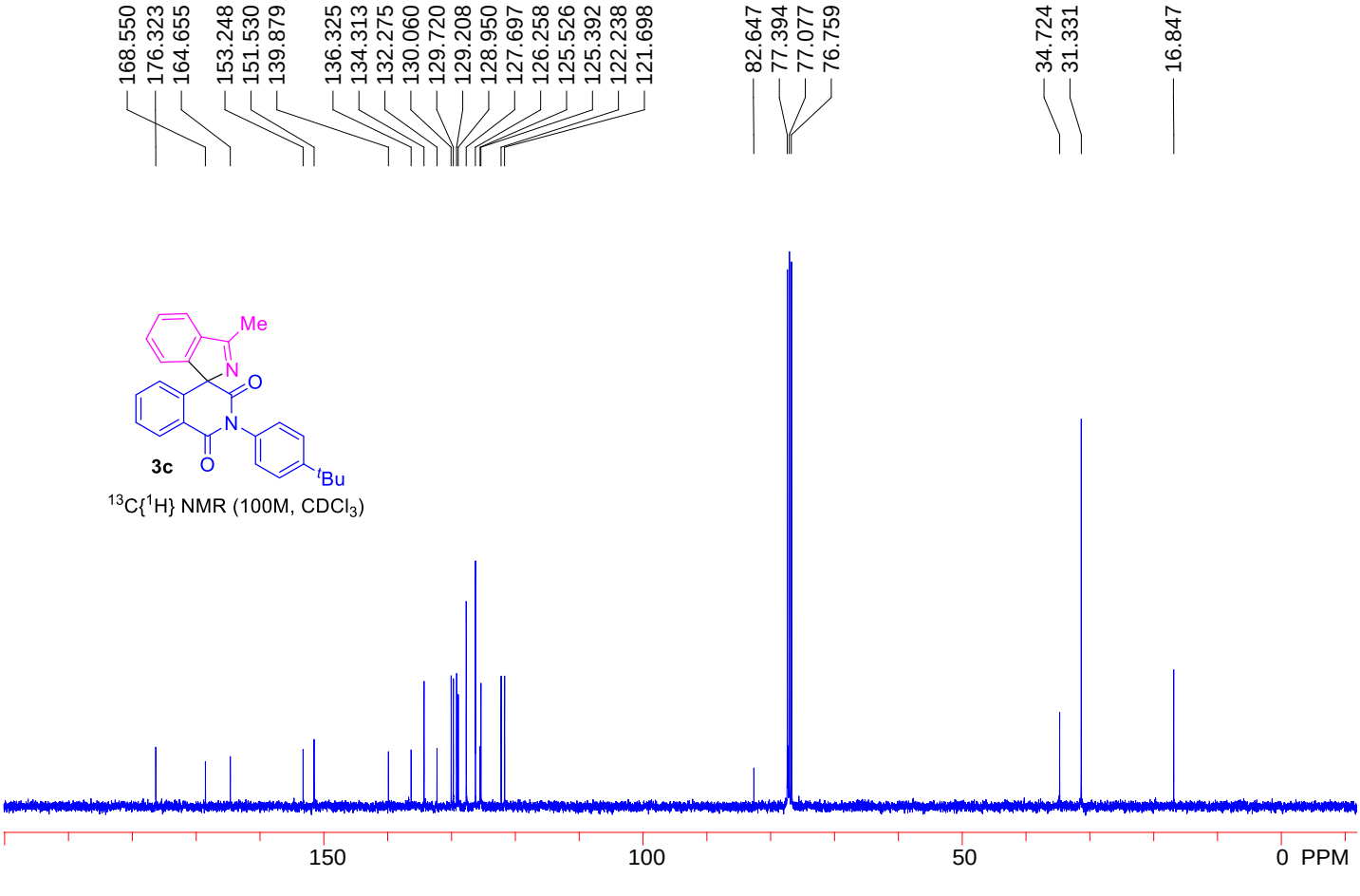


3b

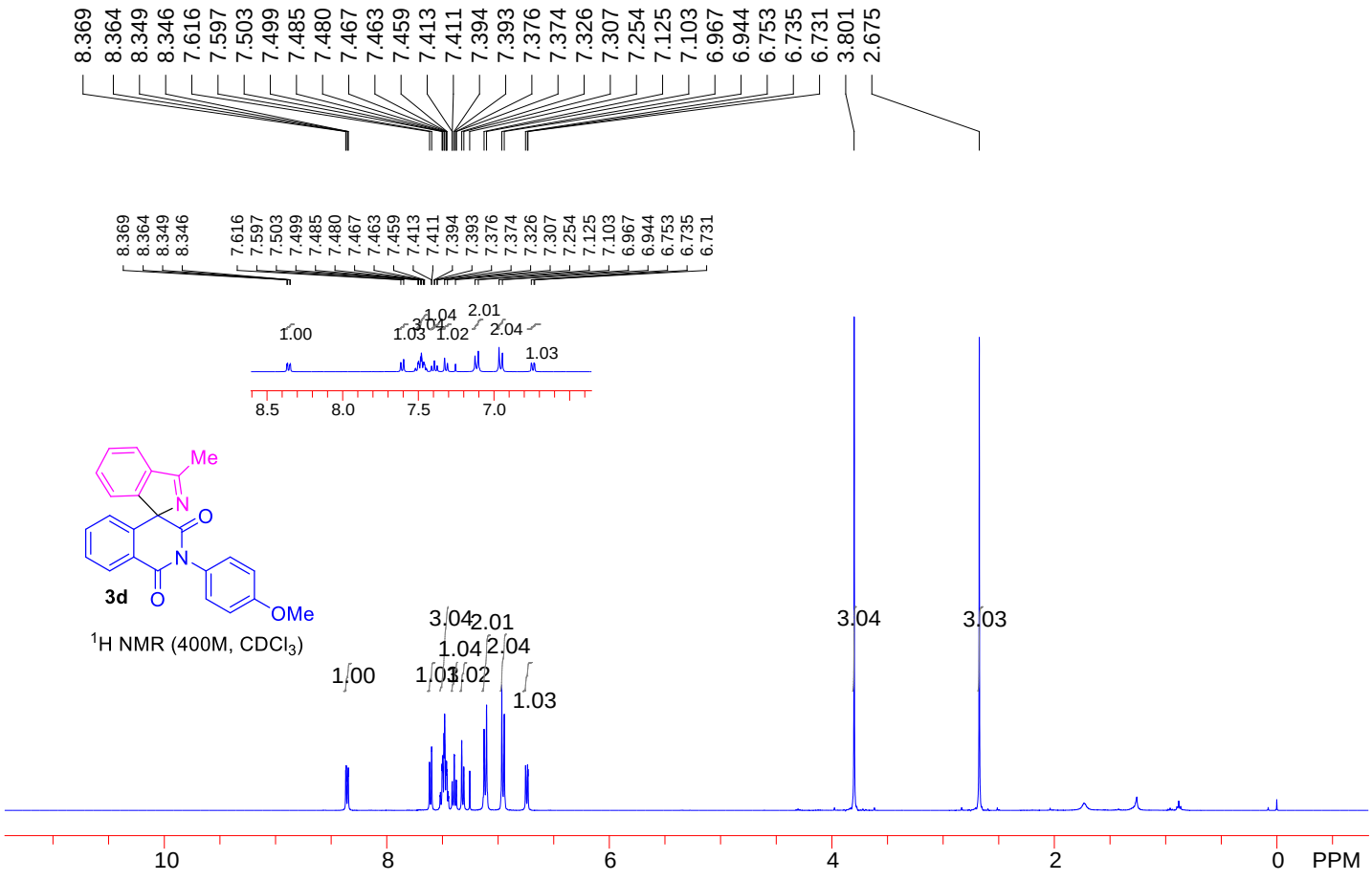
$^{13}C\{^{1}H\}$ NMR (100M, $CDCl_3$)

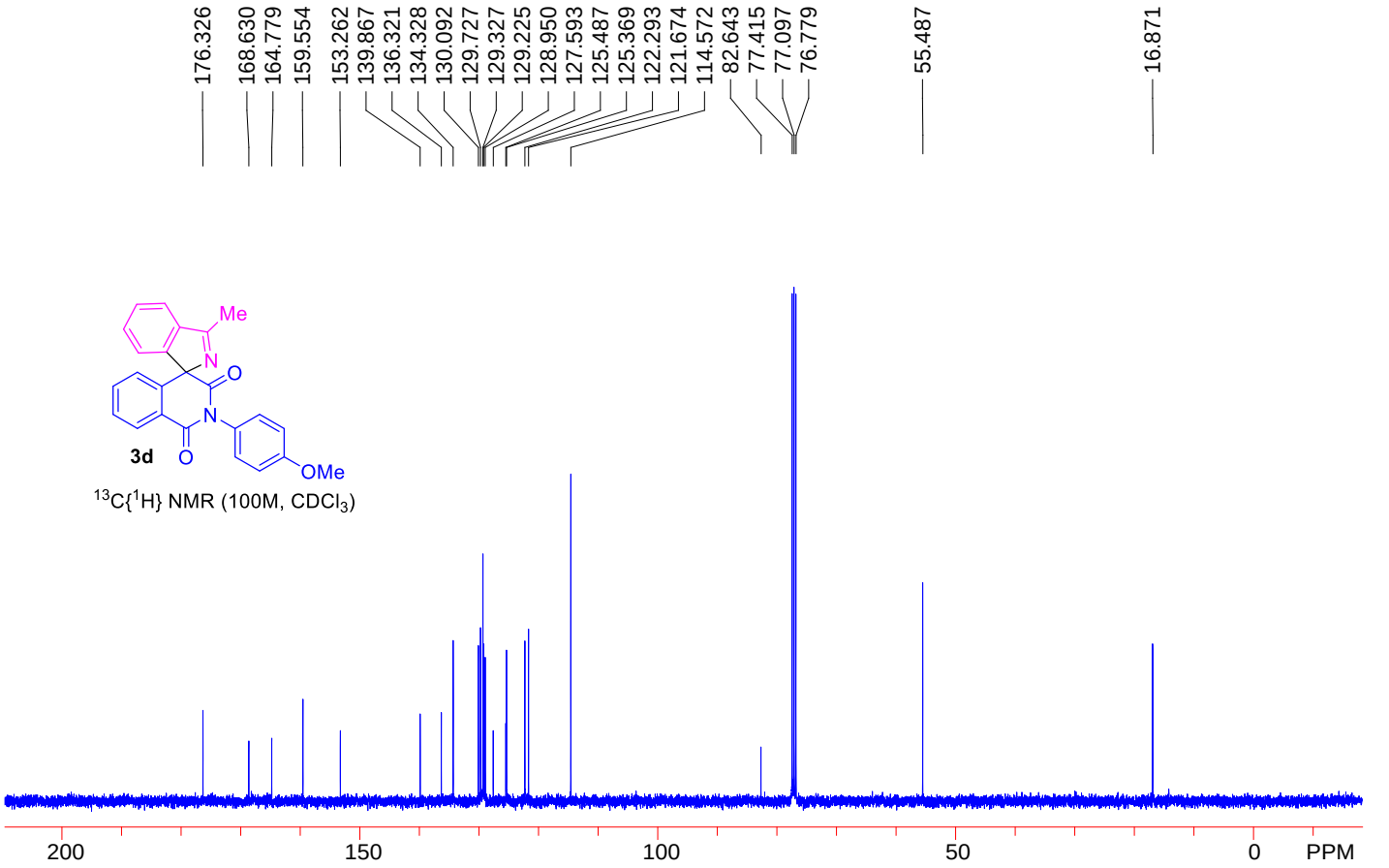


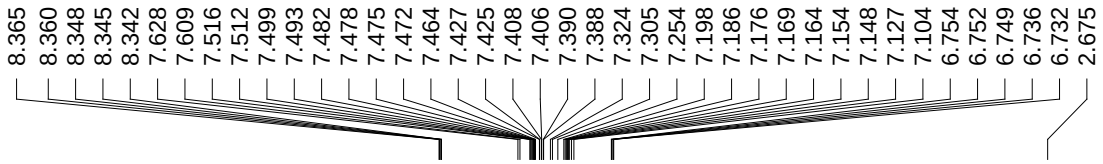
^{1}H NMR (400M, $CDCl_3$)

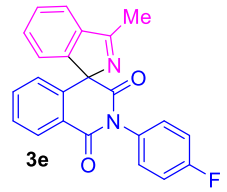


$^{13}C\{^{1}H\}$ NMR (100M, $CDCl_3$)


8.369 8.364 8.349 8.346 7.616 7.597 7.503 7.499 7.485 7.480 7.467 7.463 7.459 7.413 7.411 7.394 7.393 7.376 7.374 7.326 7.307 7.254 7.125 7.103 6.967 6.944 6.753 6.735 6.731 3.801 2.675
1.00 1.03 3.04 1.02 1.04 2.01 2.04 1.03
8.5 8.0 7.5 7.0
Me
N
O
N
O
3d
OMe
^{1}H NMR (400M, CDCl$_3$)
3.04 2.01 1.04 2.04 1.00 1.03 1.02 1.03
3.04 3.03
10 8 6 4 2 0 PPM

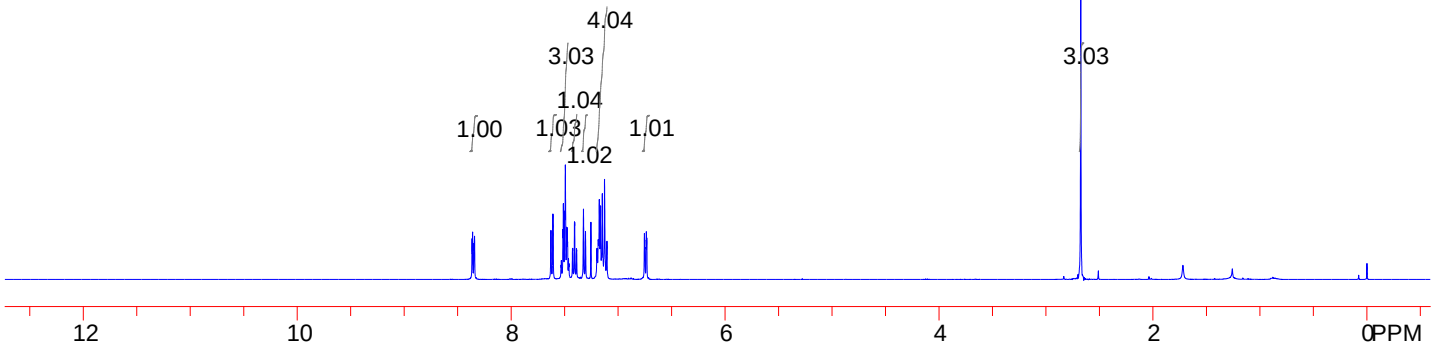

176.326 168.630 164.779 159.554 153.262 139.867 136.321 134.328 130.092 129.727 129.327 129.225 128.950 127.593 125.487 125.369 122.293 121.674 114.572 82.643 77.415 77.097 76.779 55.487 16.871
Me
N
O
N
O
3d
OMe
^{13}C{^{1}H} NMR (100M, CDCl$_3$)
200 150 100 50 0 PPM

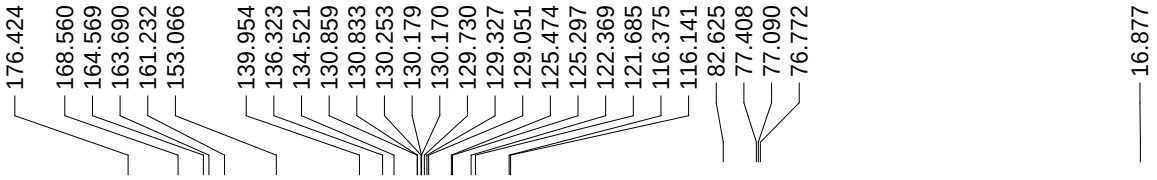


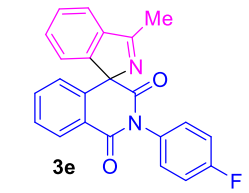


3e

^{1}H NMR (400M, $CDCl_3$)

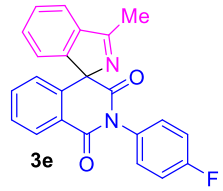






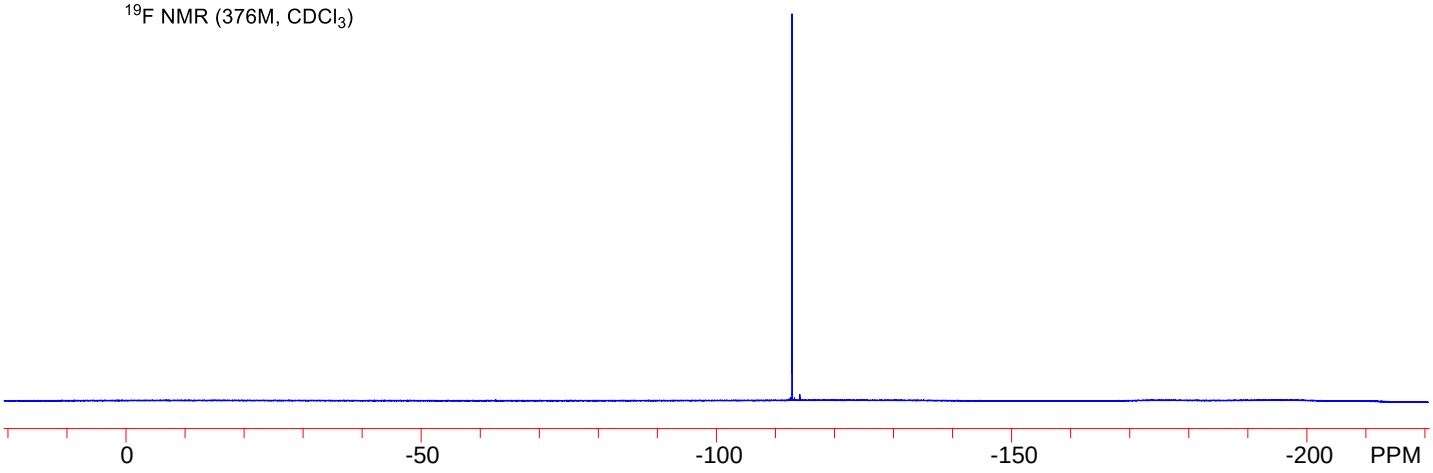
3e

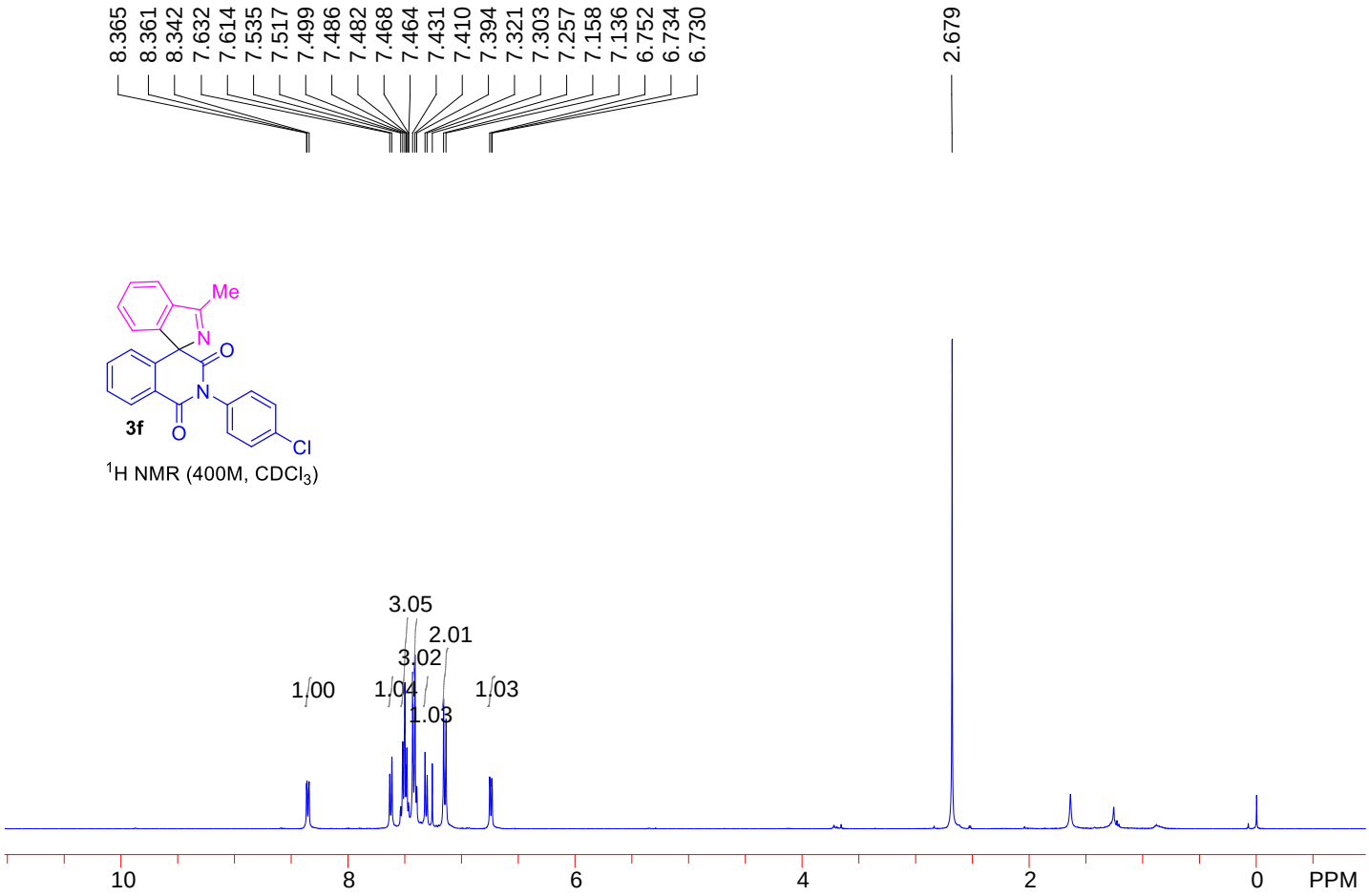
^{13}C{^{1}H} NMR (100M, $CDCl_3$)



3e

^{19}F NMR (376M, $CDCl_3$)

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8.342
7.632
7.614
7.535
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7.499
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7.482
7.468
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7.410
7.394
7.321
7.303
7.257
7.158
7.136
6.752
6.734
6.730
2.679
Me
N
O
N
O
O
Cl
3f
1H NMR (400M, CDCl3)
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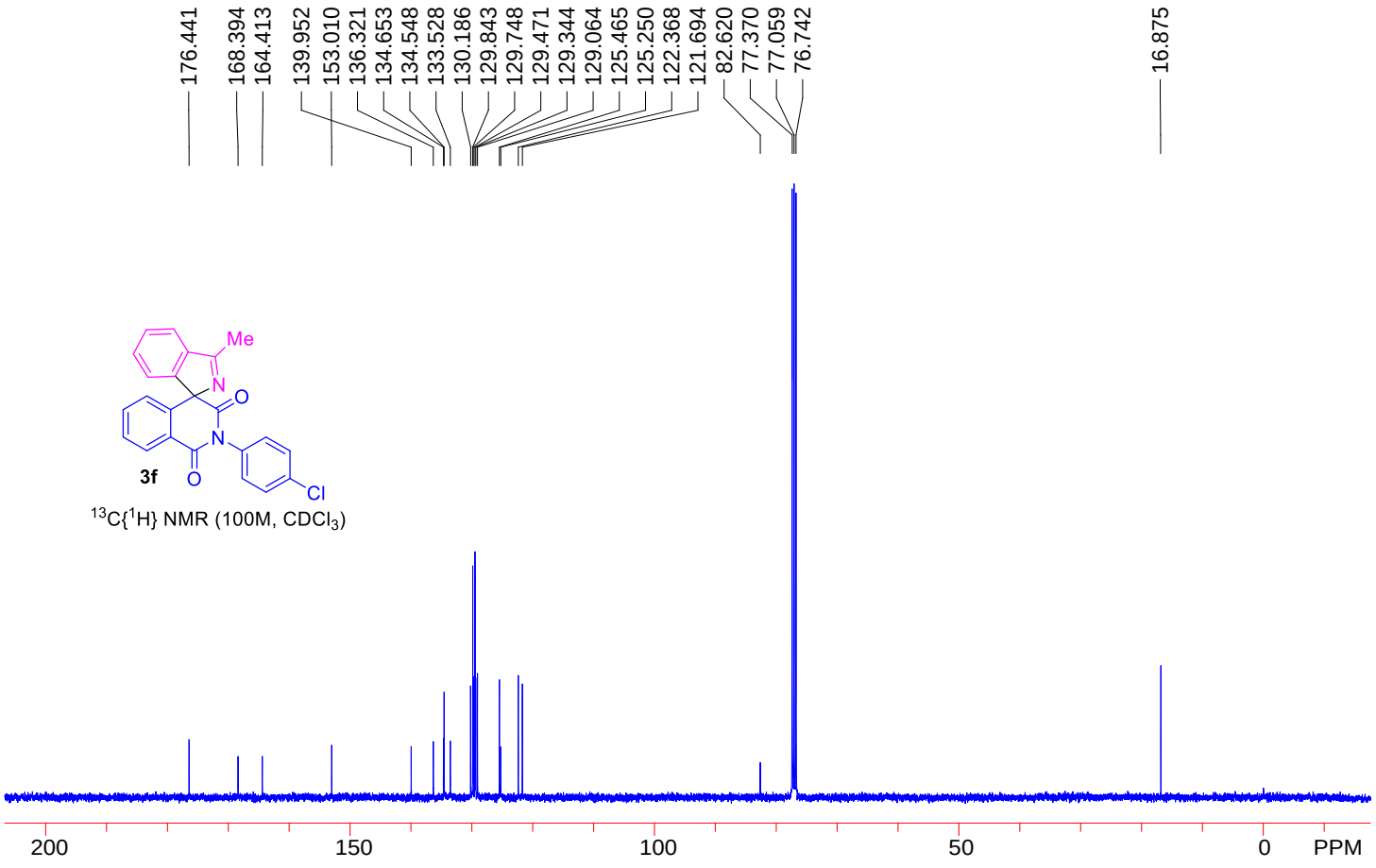
2.01
3.02
1.00
1.04
1.03
1.03
10
8
6
4
2
0
PPM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139.952
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129.064
125.465
125.250
122.368
121.694
82.620
77.370
77.059
76.742
16.875
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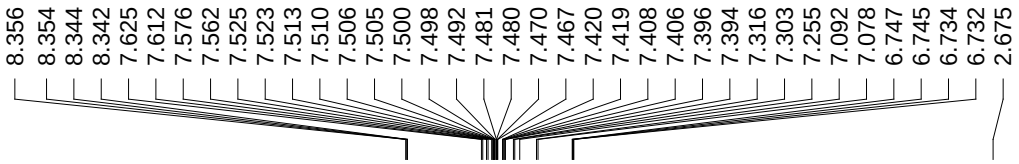
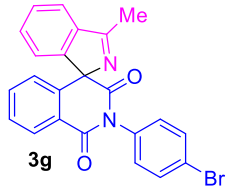
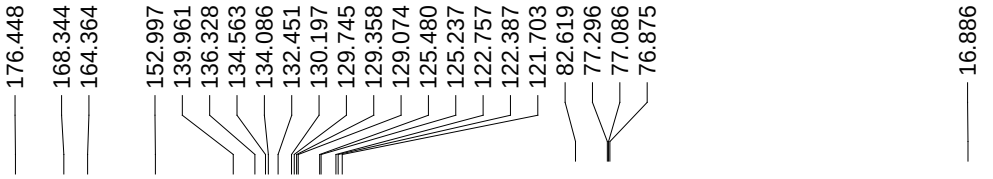
N
O
N
O
O
Cl
3f
13C{1H} NMR (100M, CDCl3)
200
150
100
50
0
PPM

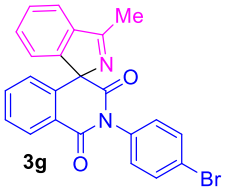




3g

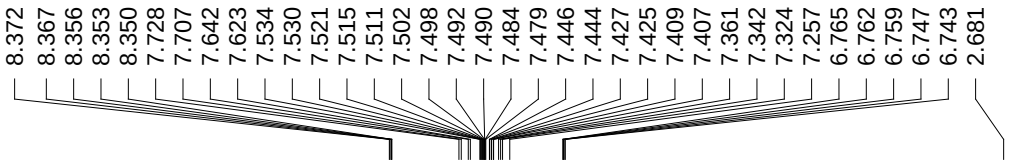
^{1}H NMR (600M, $CDCl_3$)

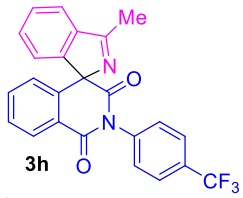




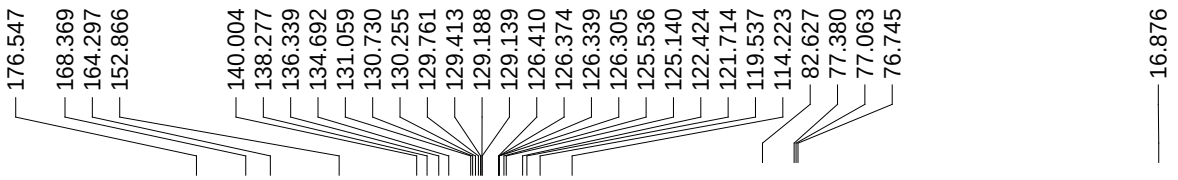
3g

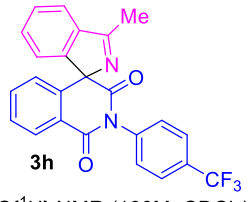
^{13}C{^{1}H} NMR (150M, $CDCl_3$)



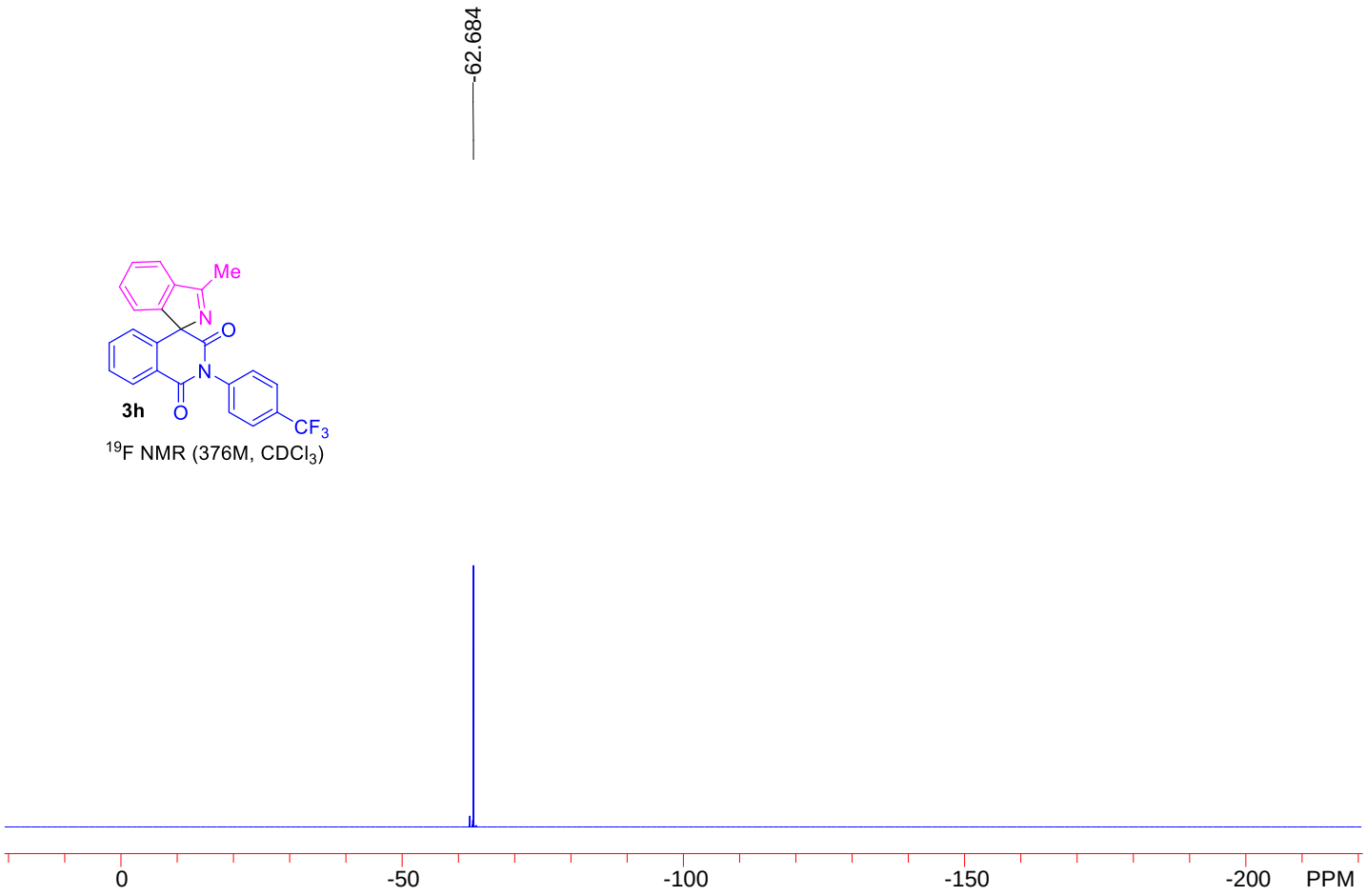


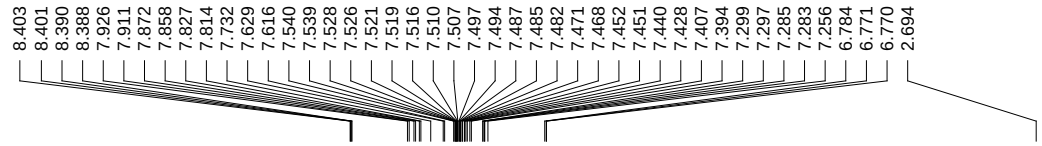
^{1}H NMR (400M, $CDCl_3$)

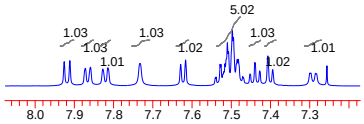


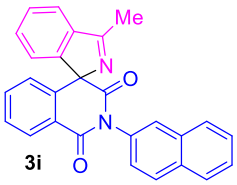


^{13}C{^{1}H} NMR (100M, $CDCl_3$)

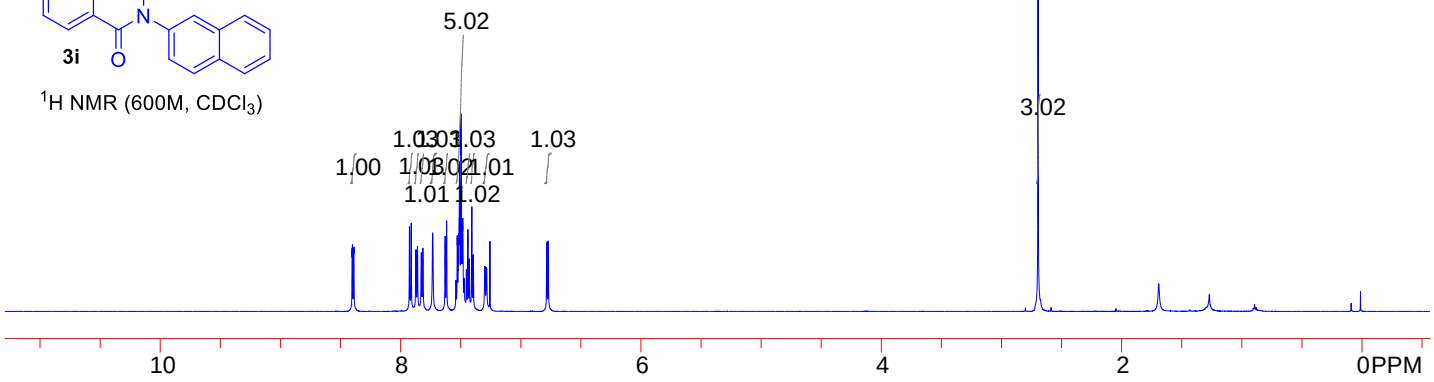

-62.684
Me
N
O
N
O
CF3
3h
19F NMR (376M, CDCl3)
0
-50
-100
-150
-200
PPM

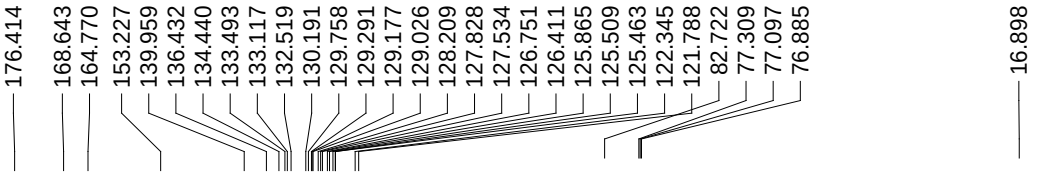


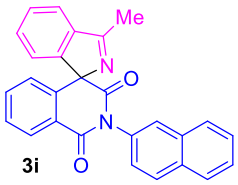




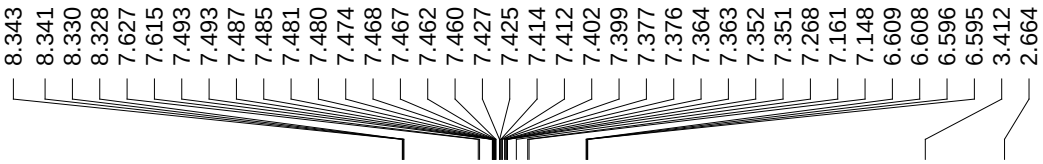
^{1}H NMR (600M, $CDCl_3$)

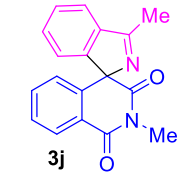






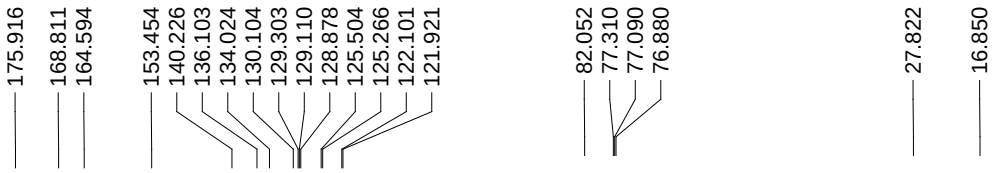
^{13}C{^{1}H} NMR (150M, $CDCl_3$)

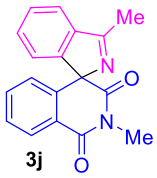




3j

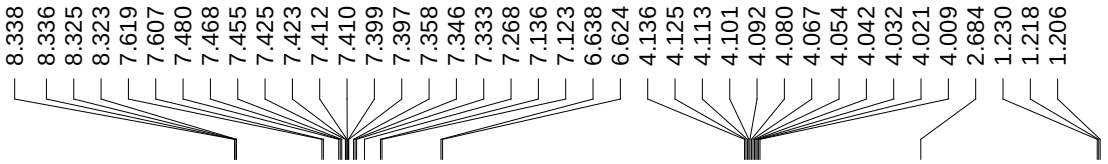
^{1}H NMR (600M, $CDCl_3$)

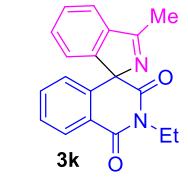




3j

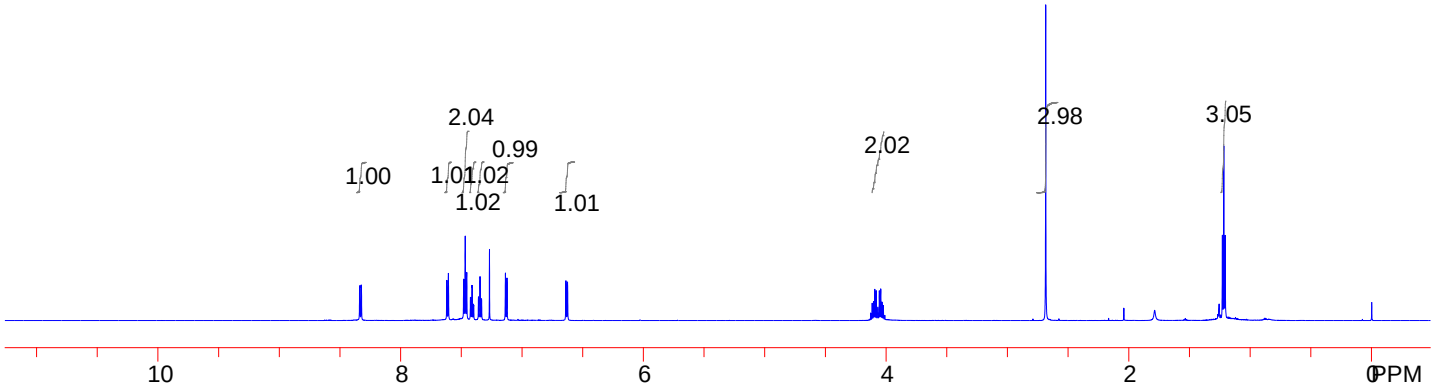
^{13}C{^{1}H} NMR (150M, $CDCl_3$)

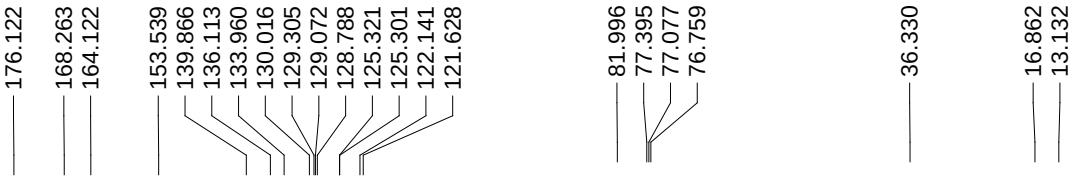


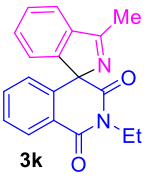


3k

^{1}H NMR (600M, $CDCl_3$)

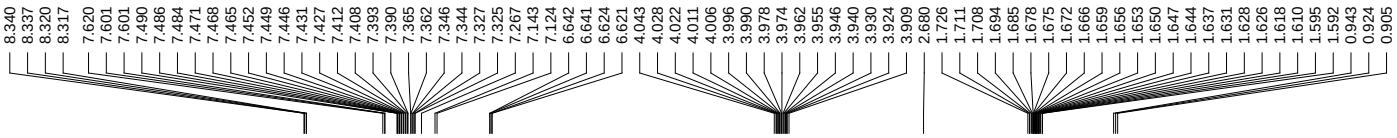


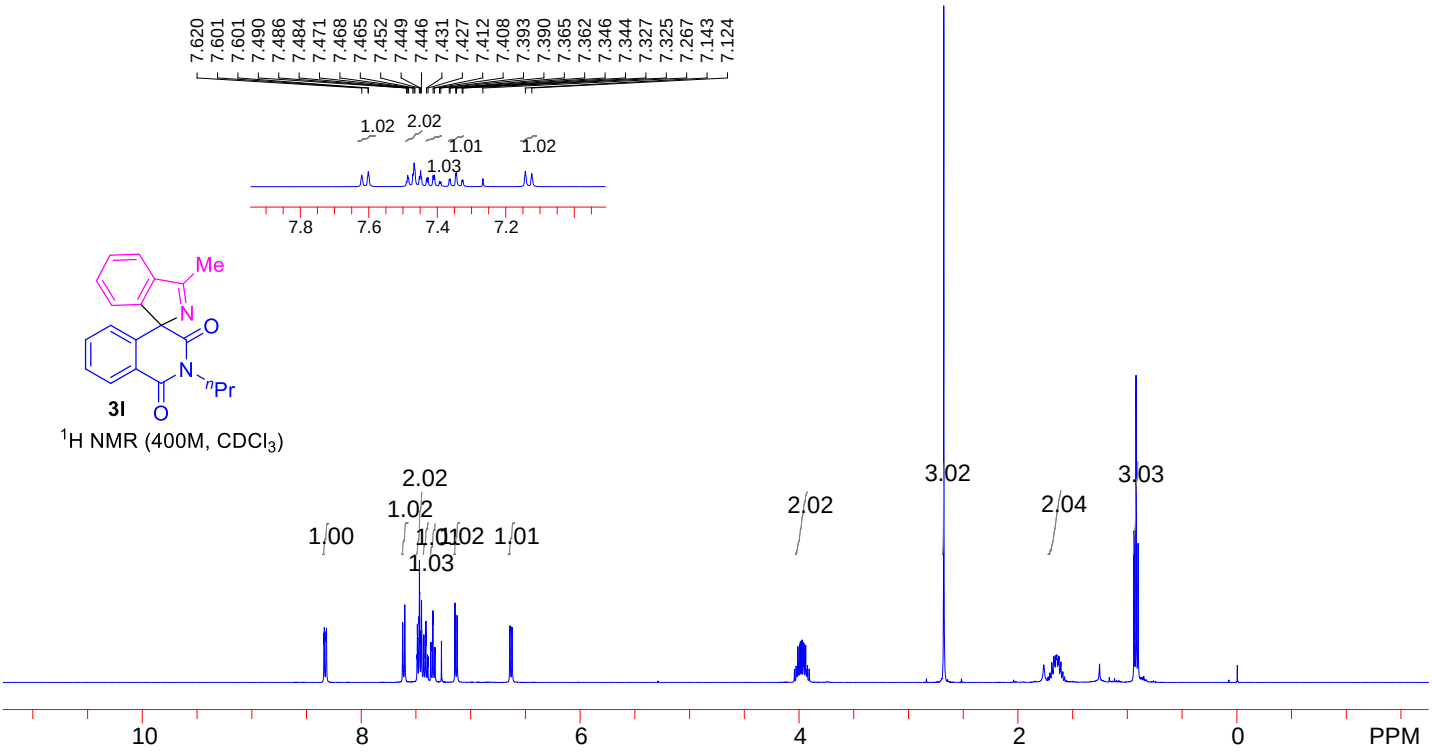




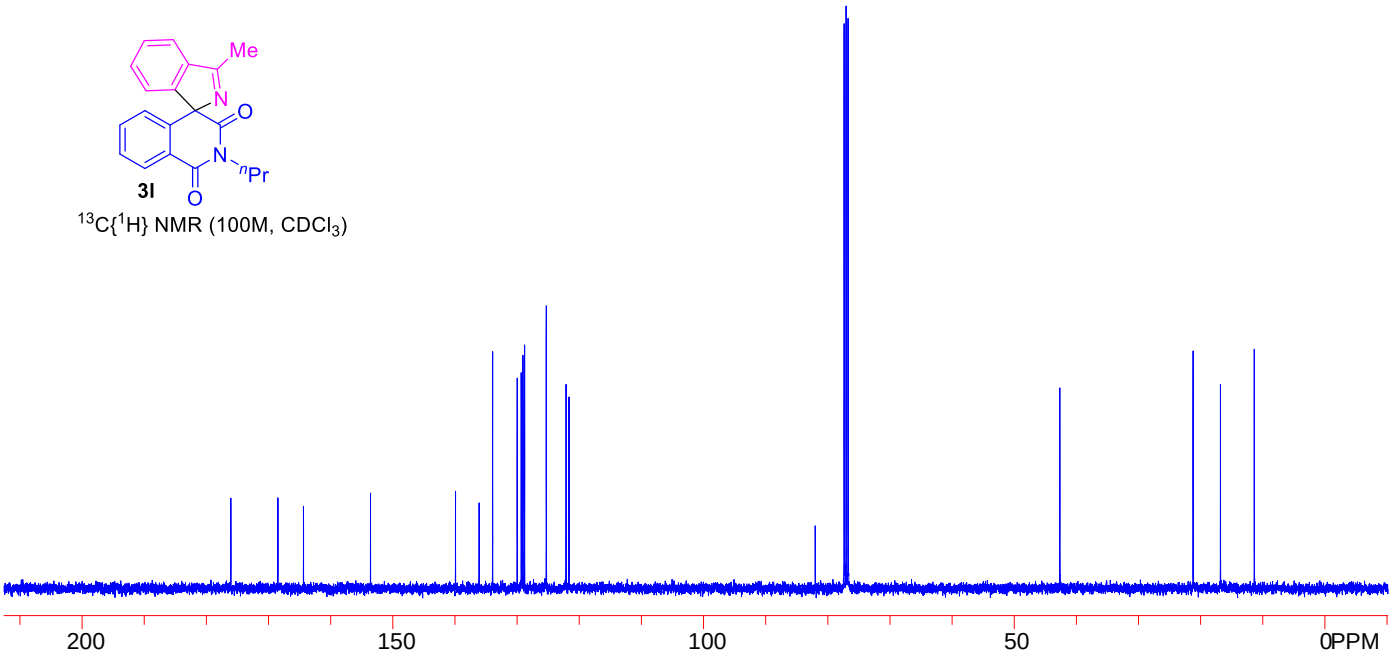
3k

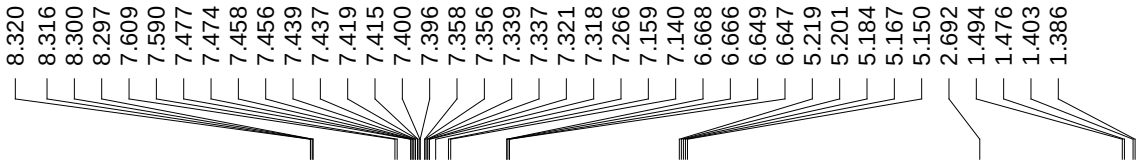
^{13}C{^{1}H} NMR (100M, $CDCl_3$)

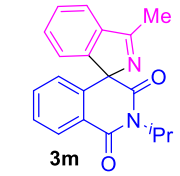

8.340 8.337 8.320 8.317 7.620 7.601 7.601 7.490 7.486 7.484 7.471 7.468 7.465 7.452 7.449 7.446 7.431 7.427 7.412 7.408 7.393 7.390 7.365 7.362 7.346 7.344 7.327 7.325 7.267 7.143 7.124 6.642 6.641 6.624 6.621 4.043 4.028 4.022 4.011 4.006 3.996 3.990 3.978 3.974 3.962 3.955 3.946 3.940 3.930 3.924 3.909 2.680 1.726 1.711 1.708 1.694 1.685 1.678 1.675 1.672 1.666 1.659 1.656 1.653 1.650 1.647 1.644 1.637 1.631 1.628 1.626 1.618 1.610 1.595 1.592 0.943 0.924 0.905


7.620 7.601 7.601 7.490 7.486 7.484 7.471 7.468 7.465 7.452 7.449 7.446 7.431 7.427 7.412 7.408 7.393 7.390 7.365 7.362 7.346 7.344 7.327 7.325 7.267 7.143 7.124
1.02 2.02 1.01 1.02 1.03
7.8 7.6 7.4 7.2
Me N O N nPr O
3l
1H NMR (400M, CDCl3)
2.02 1.02 1.00 1.01 1.02 1.01 1.03 2.02 3.02 2.04 3.03
10 8 6 4 2 0 PPM


176.048 168.490 164.370 153.589 139.910 136.110 133.947 129.992 129.342 129.063 128.797 125.304 122.126 121.649 82.040 77.396 77.078 76.760 42.685 21.251 16.844 11.406

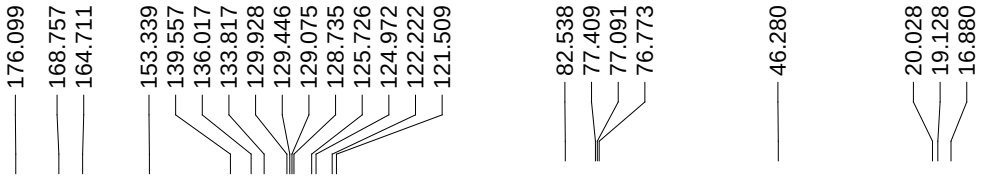

Me N O N nPr O
3l
13C{1H} NMR (100M, CDCl3)
200 150 100 50 0PPM

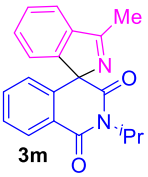




3m

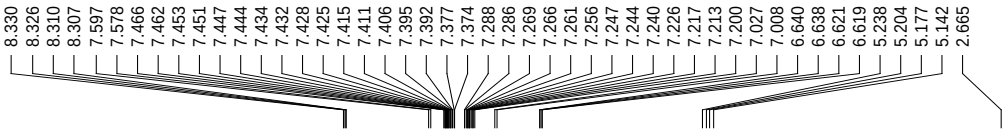
^{1}H NMR (400M, $CDCl_3$)

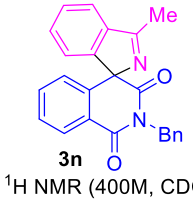




3m

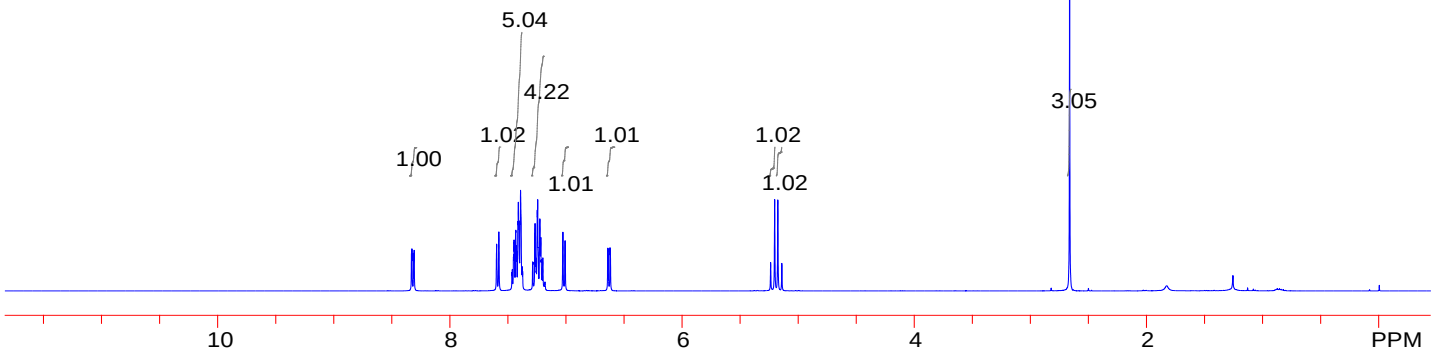
^{13}C{^{1}H} NMR (100M, $CDCl_3$)

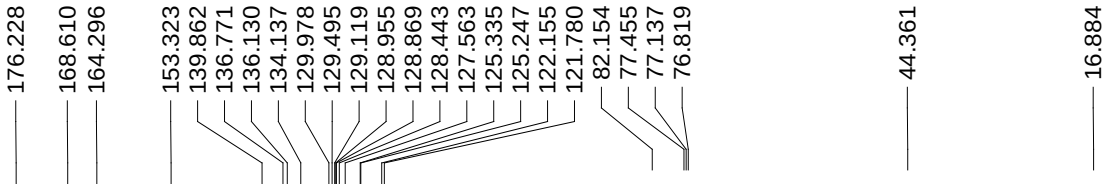


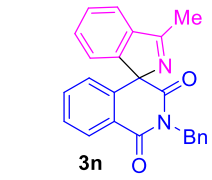


3n

^{1}H NMR (400M, $CDCl_3$)

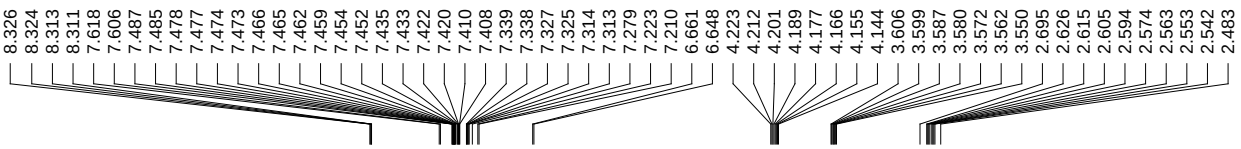


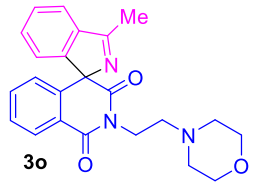




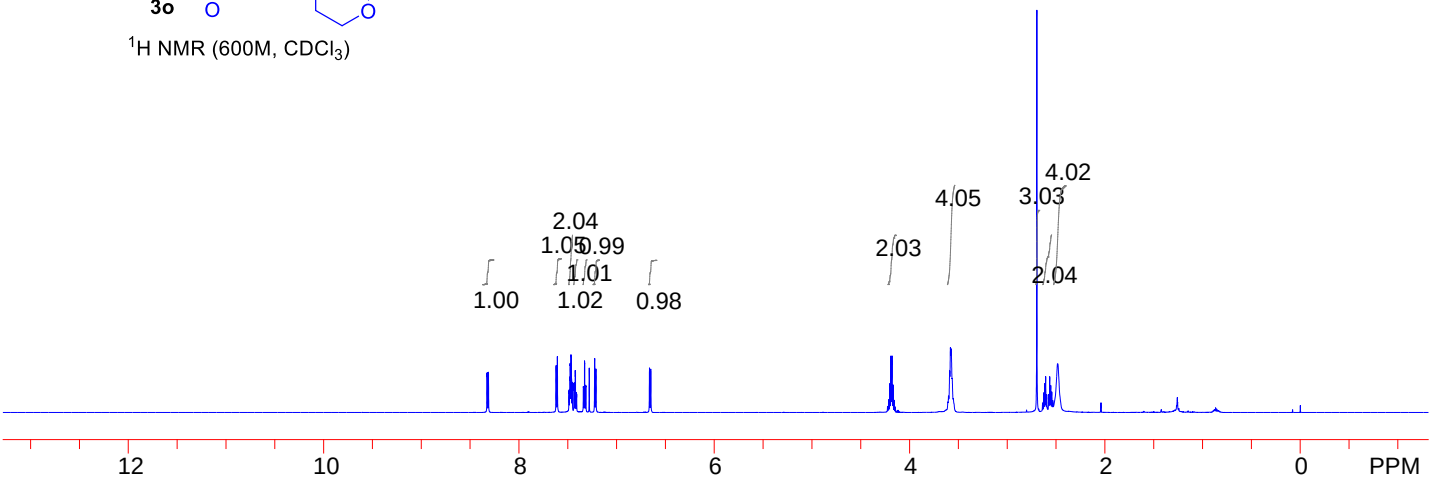
3n

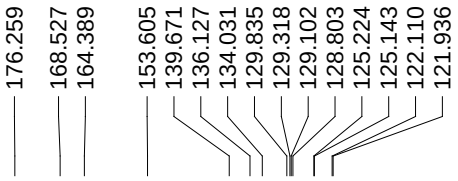
^{13}C{^{1}H} NMR (100M, $CDCl_3$)

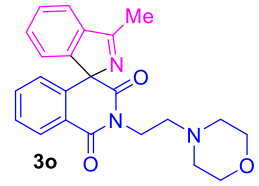




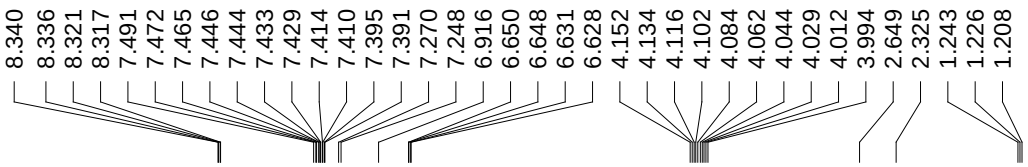
^{1}H NMR (600M, $CDCl_3$)

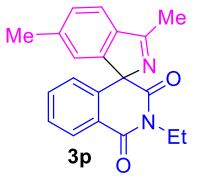






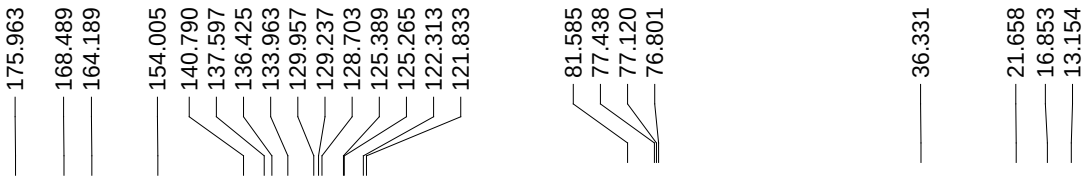
$^{13}C\{^{1}H\}$ NMR (150M, $CDCl_3$)

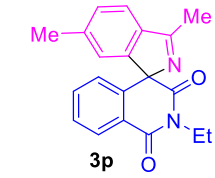




3p

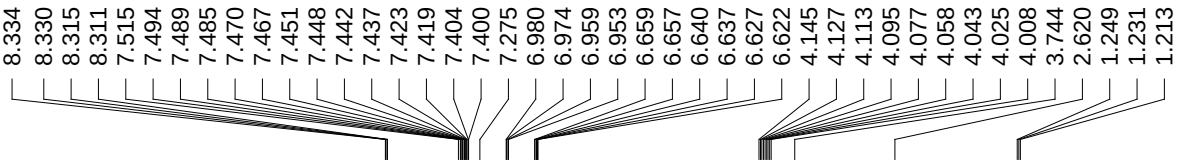
^{1}H NMR (400M, $CDCl_3$)

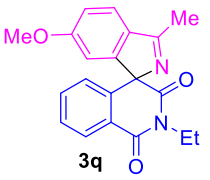




3p

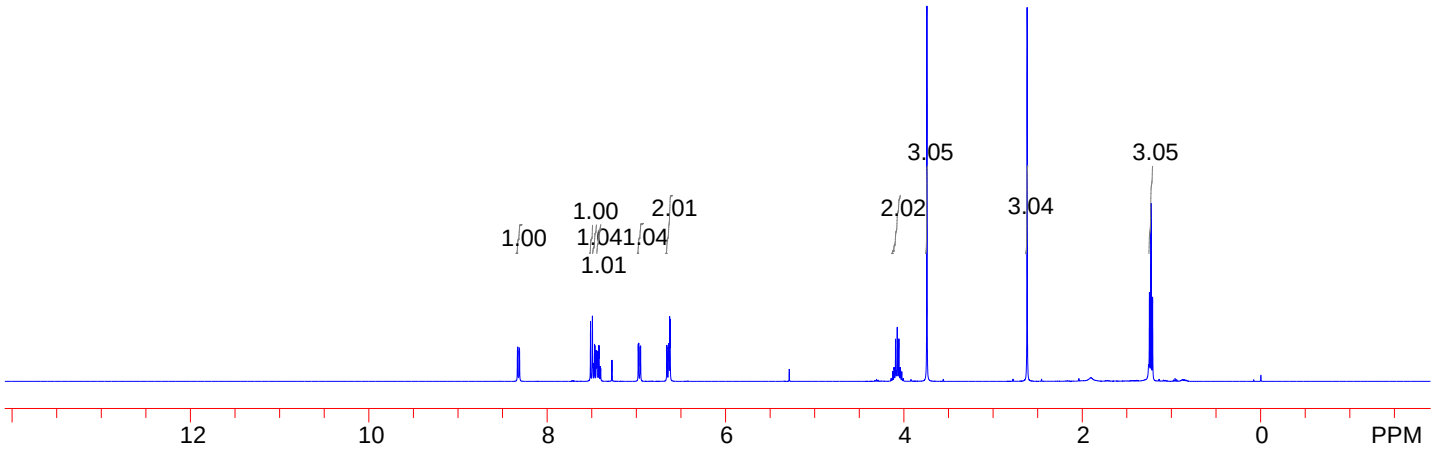
$^{13}C\{^{1}H\}$ NMR (100M, $CDCl_3$)

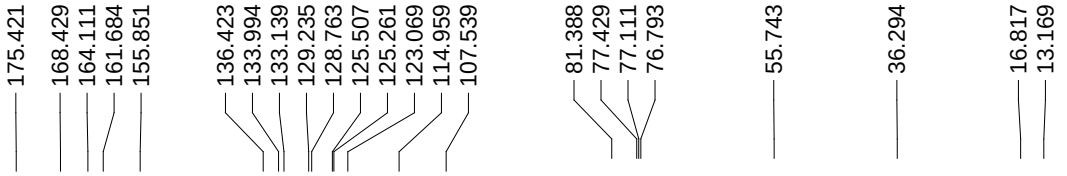


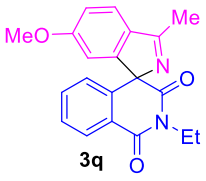


3q

^{1}H NMR (400M, $CDCl_3$)

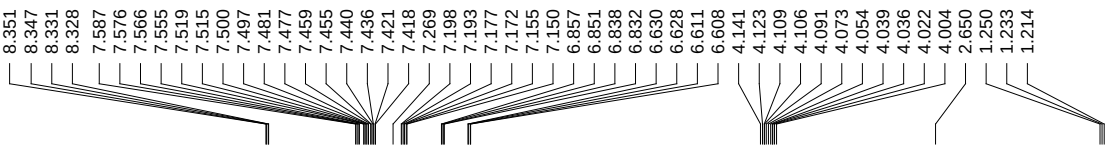


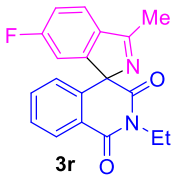




3q

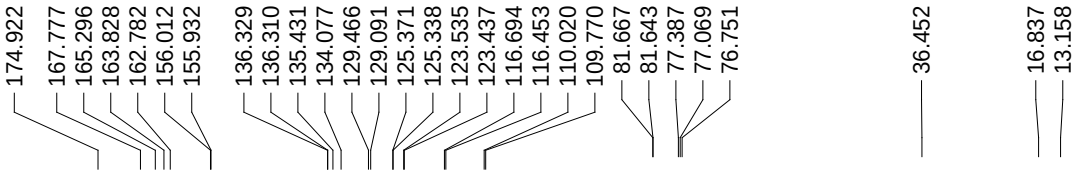
$^{13}C\{^{1}H\}$ NMR (100M, $CDCl_3$)

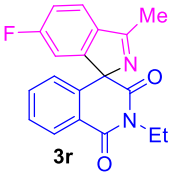




3r

^{1}H NMR (400M, $CDCl_3$)

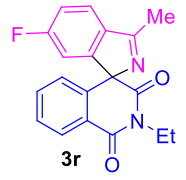




3r

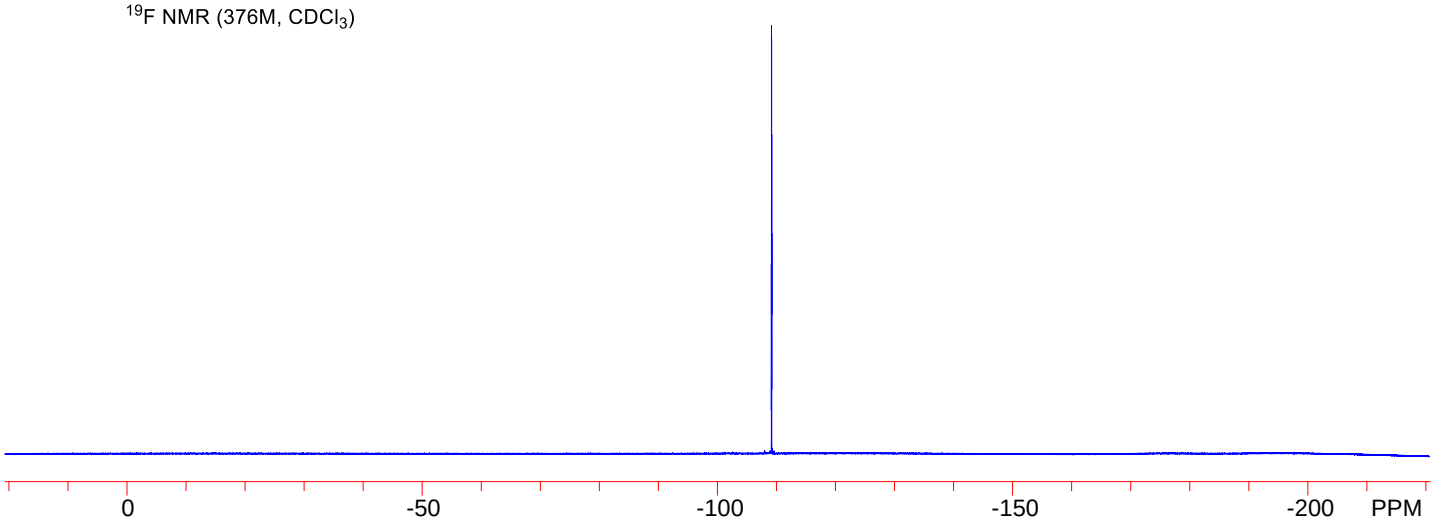
$^{13}C\{^{1}H\}$ NMR (100M, $CDCl_3$)

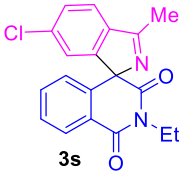
109.202
109.216
109.225
109.240
109.245
109.262



3r

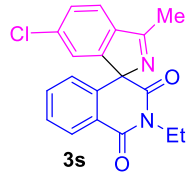
^{19}F NMR (376M, $CDCl_3$)





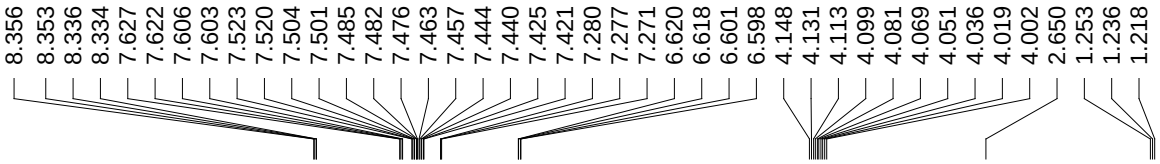
3s

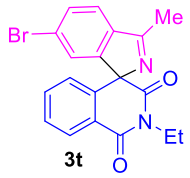
^{1}H NMR (600M, $CDCl_3$)



3s

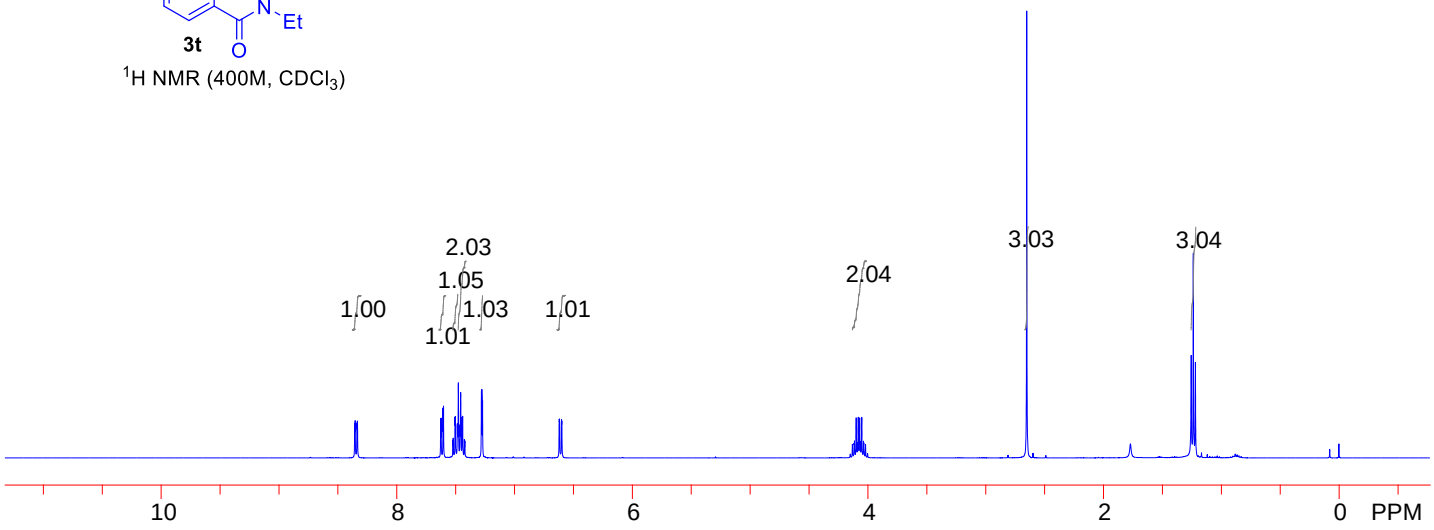
$^{13}C\{^{1}H\}$ NMR (100M, $CDCl_3$)

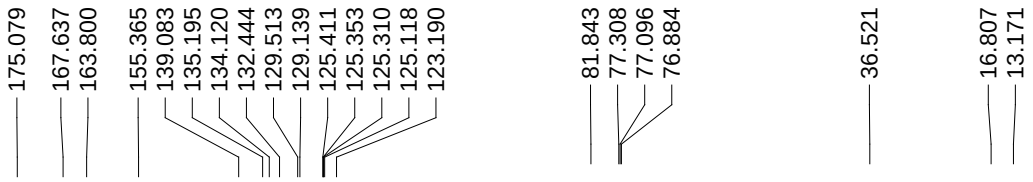


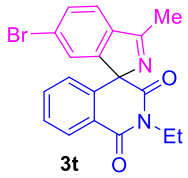


3t

^{1}H NMR (400M, $CDCl_3$)

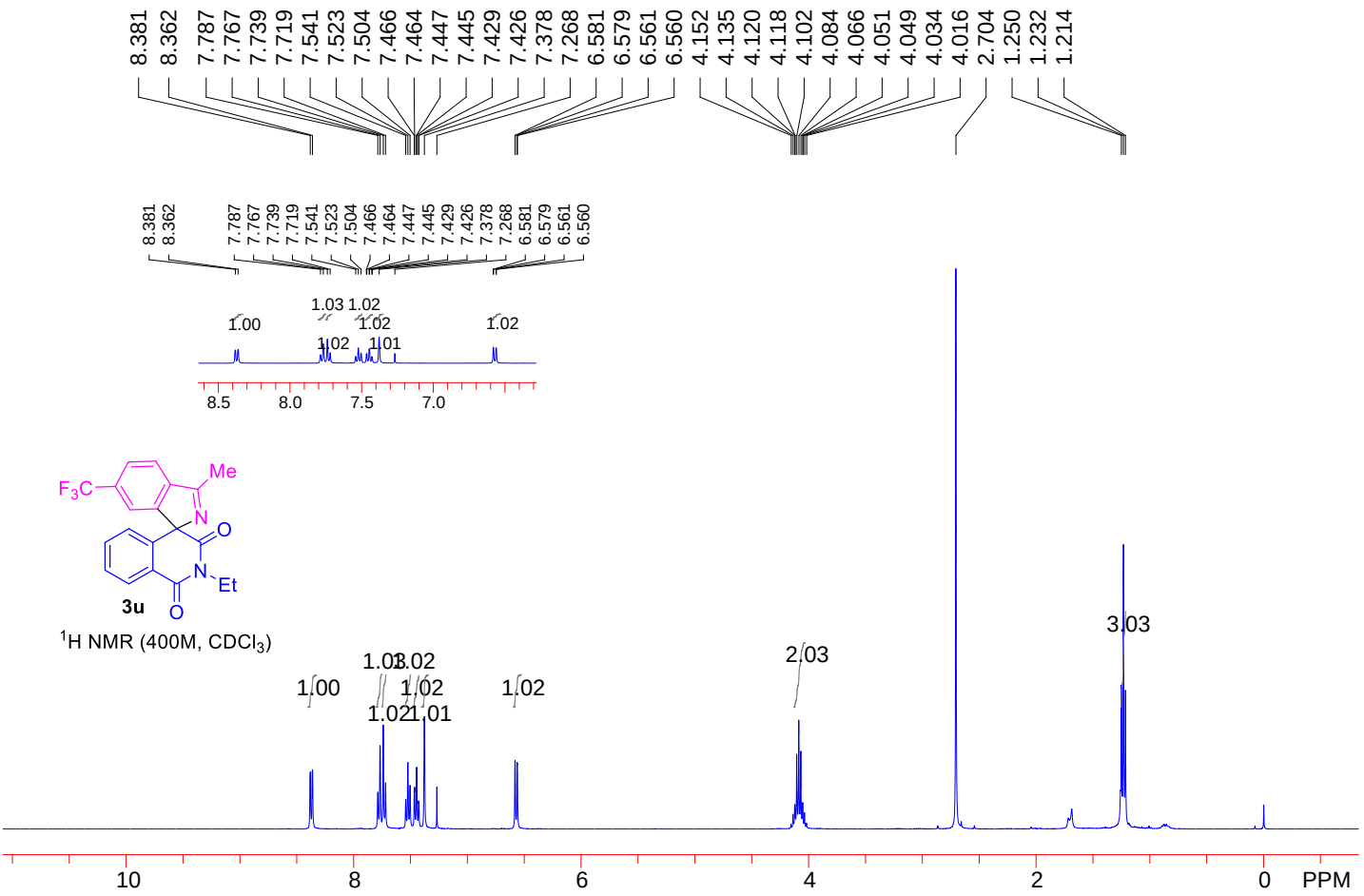






3t

$^{13}C\{^{1}H\}$ NMR (100M, $CDCl_3$)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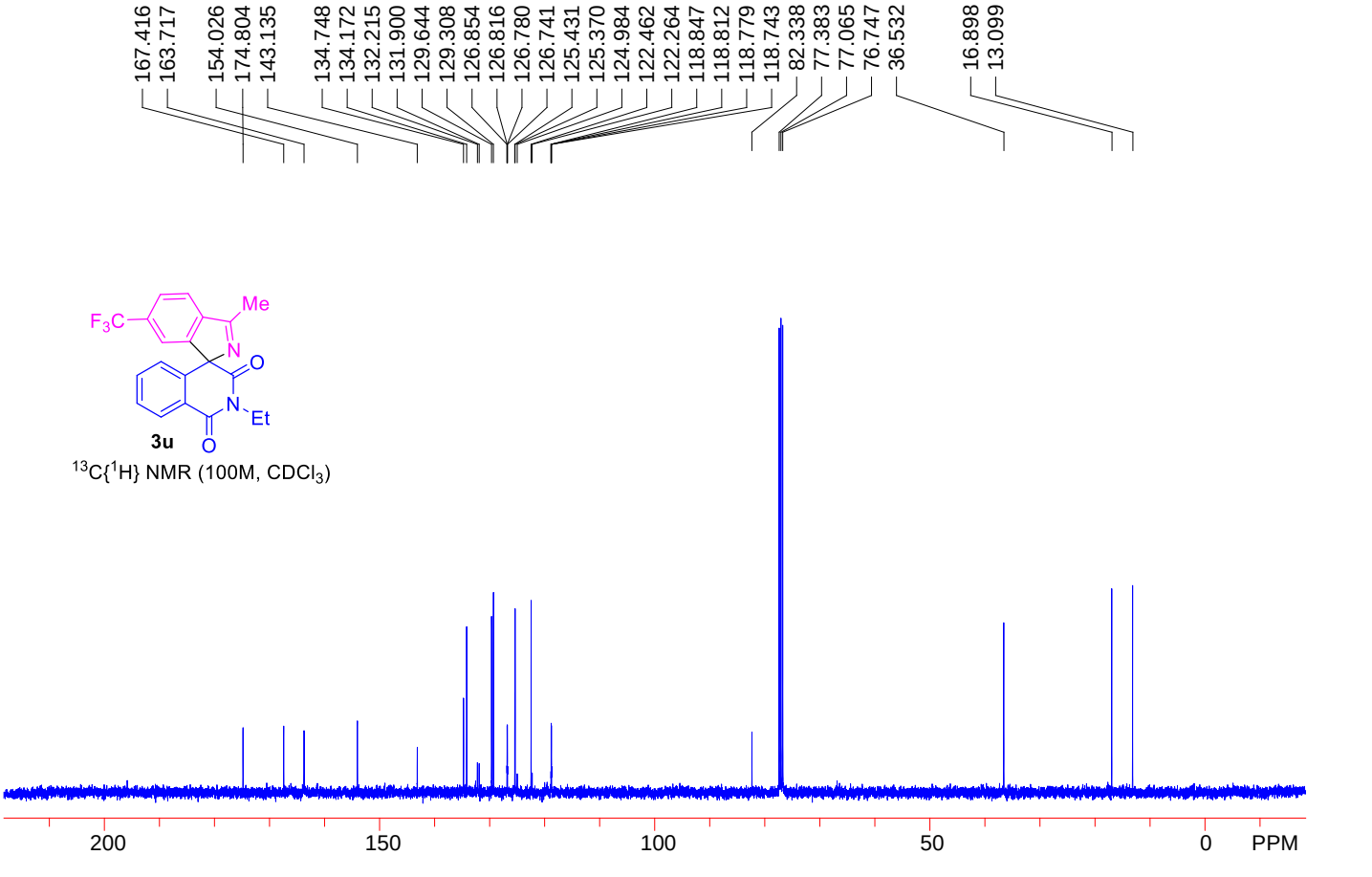
7.464
7.447
7.445
7.429
7.426
7.378
7.268
6.581
6.579
6.561
6.560
4.152
4.135
4.120
4.118
4.102
4.084
4.066
4.051
4.049
4.034
4.016
2.704
1.250
1.232
1.214
1.00
1.03
1.02
1.02
1.02
1.02
1.01
1.02
8.5
8.0
7.5
7.0
Me
F3C
N
O
N
Et
O
3u
1H NMR (400M, CDCl3)
1.00
1.03
1.02
1.02
1.02
1.02
1.01
1.02
2.03
3.03
10
8
6
4
2
0
PPM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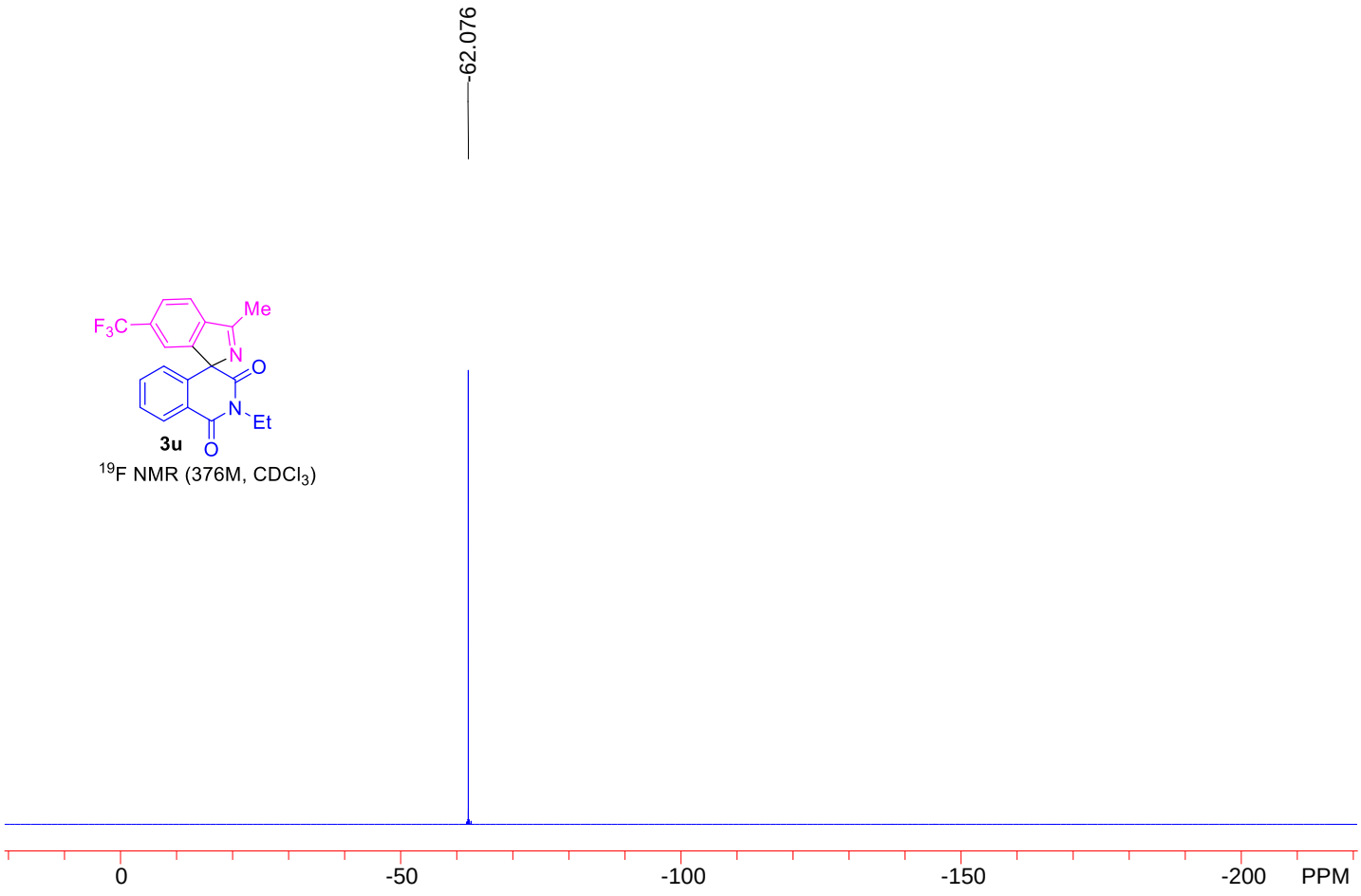
126.780
126.741
125.431
125.370
124.984
122.462
122.264
118.847
118.812
118.779
118.743
82.338
77.383
77.065
76.747
36.532
16.898
13.099
Me
F3C
N
O
N
Et
O
3u
13C{1H} NMR (100M, CDCl3)
200
150
100
50
0
PPM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N
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Et
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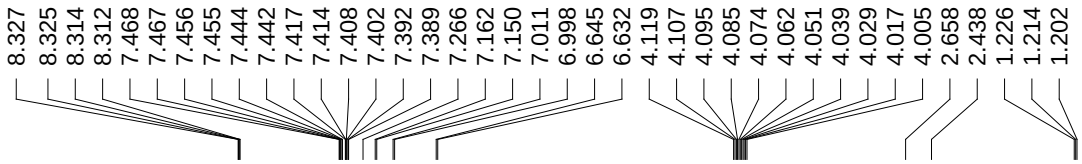
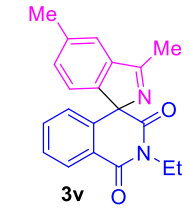
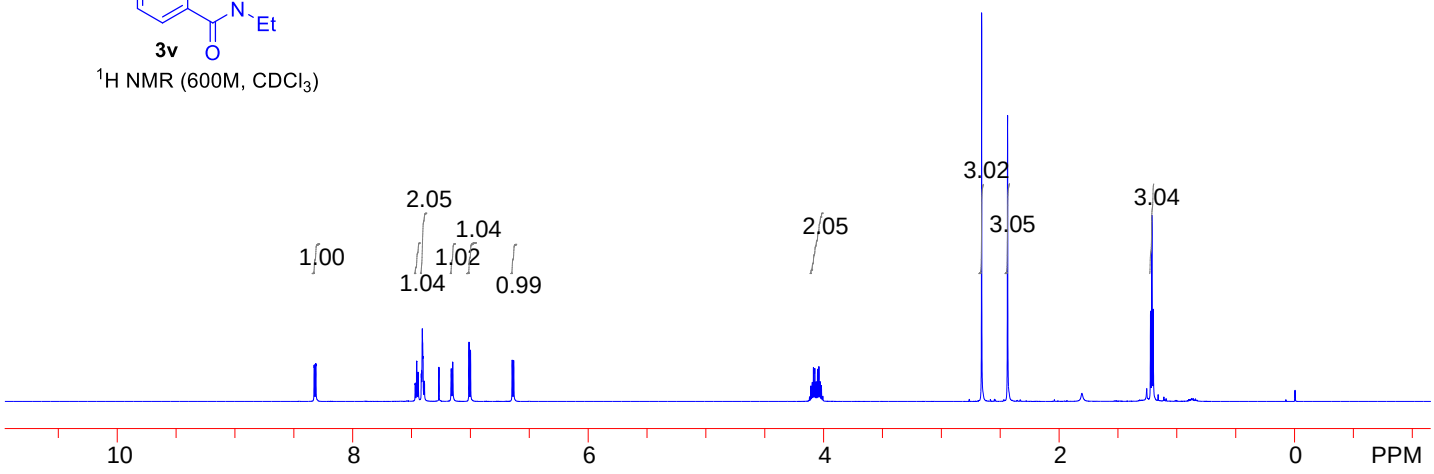
19F NMR (376M, CDCl3)
0
-50
-100
-150
-200
PPM

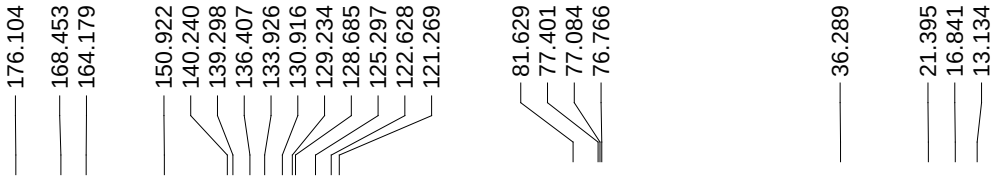


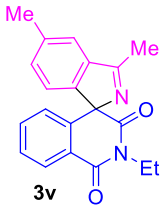


3v

^{1}H NMR (600M, $CDCl_3$)

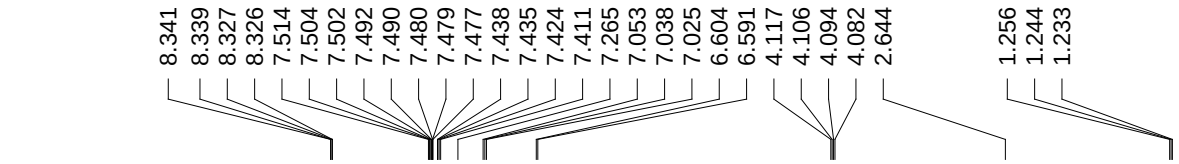


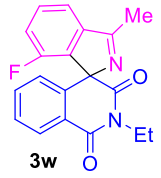




3v

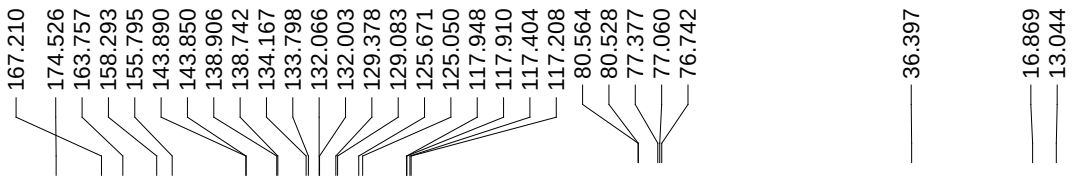
^{13}C{^{1}H} NMR (100M, $CDCl_3$)

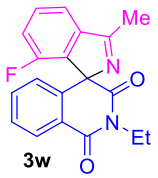




3w

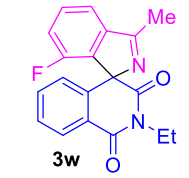
^{1}H NMR (600M, $CDCl_3$)





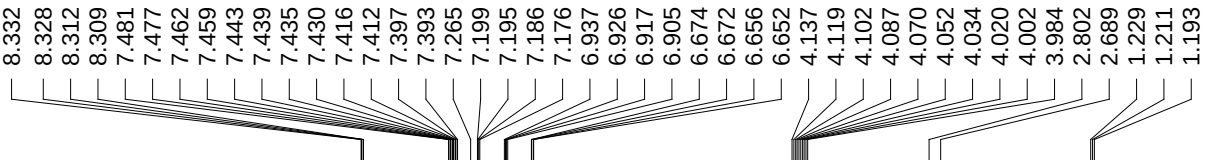
3w

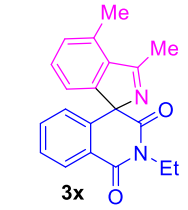
^{13}C{^{1}H} NMR (100M, $CDCl_3$)



3w

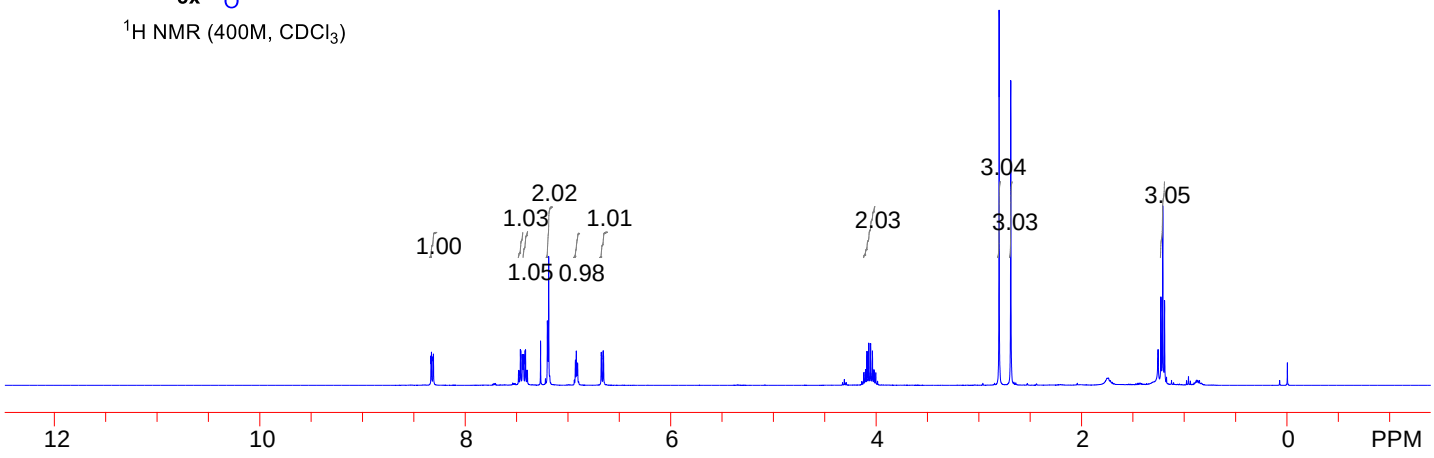
^{19}F NMR (565M, $CDCl_3$)

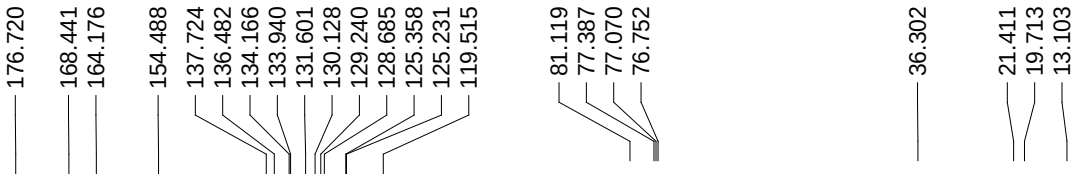


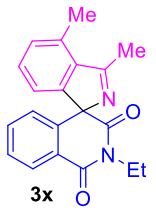


3x

^{1}H NMR (400M, $CDCl_3$)

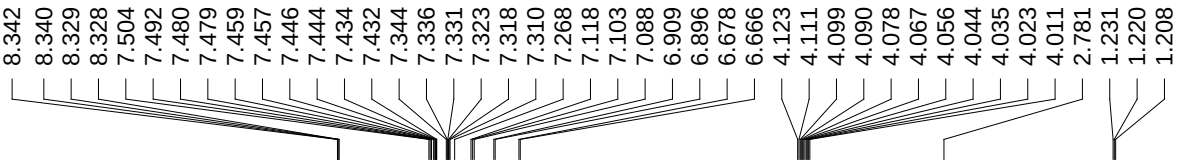


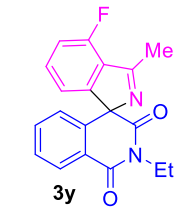




3x

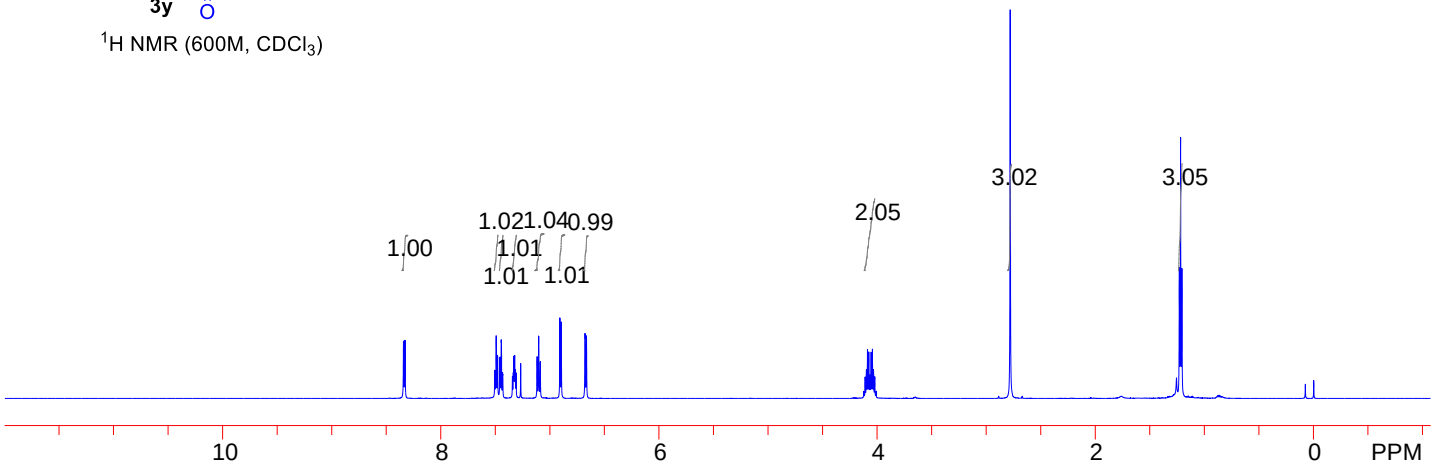
^{13}C{^{1}H} NMR (100M, $CDCl_3$)

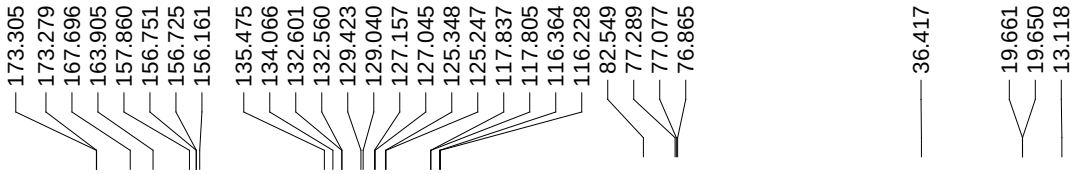


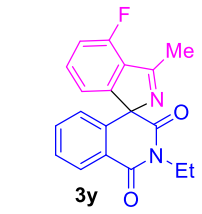


3y

^{1}H NMR (600M, $CDCl_3$)

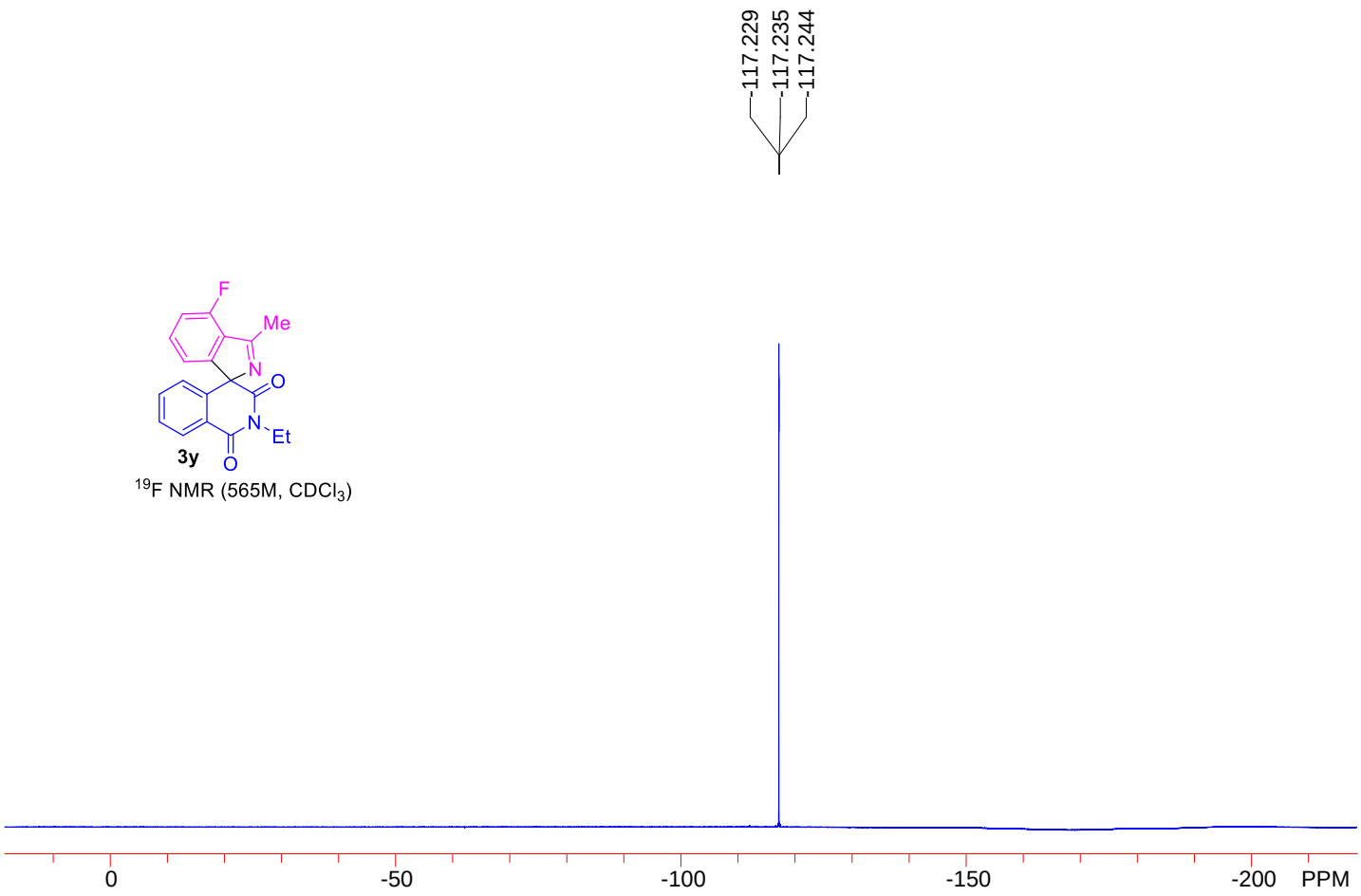






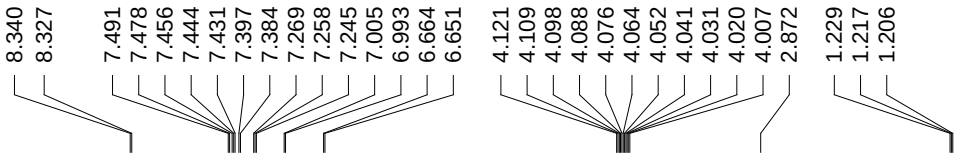
3y

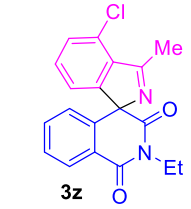
^{13}C{^{1}H} NMR (150M, $CDCl_3$)



3y

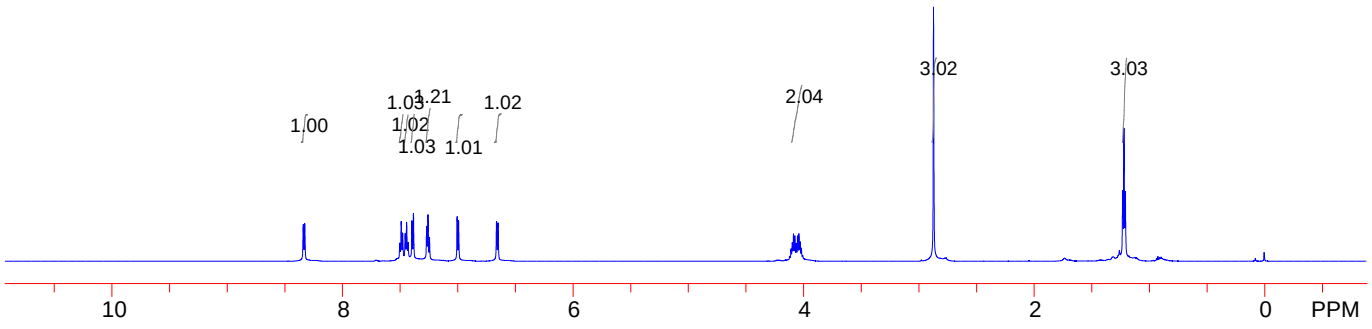
^{19}F NMR (565M, $CDCl_3$)

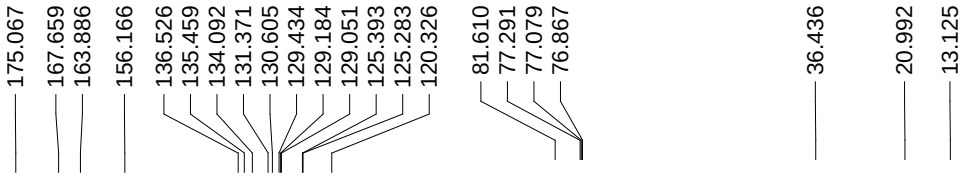


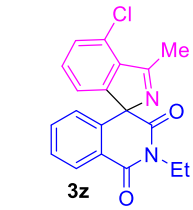


3z

^{1}H NMR (400M, $CDCl_3$)

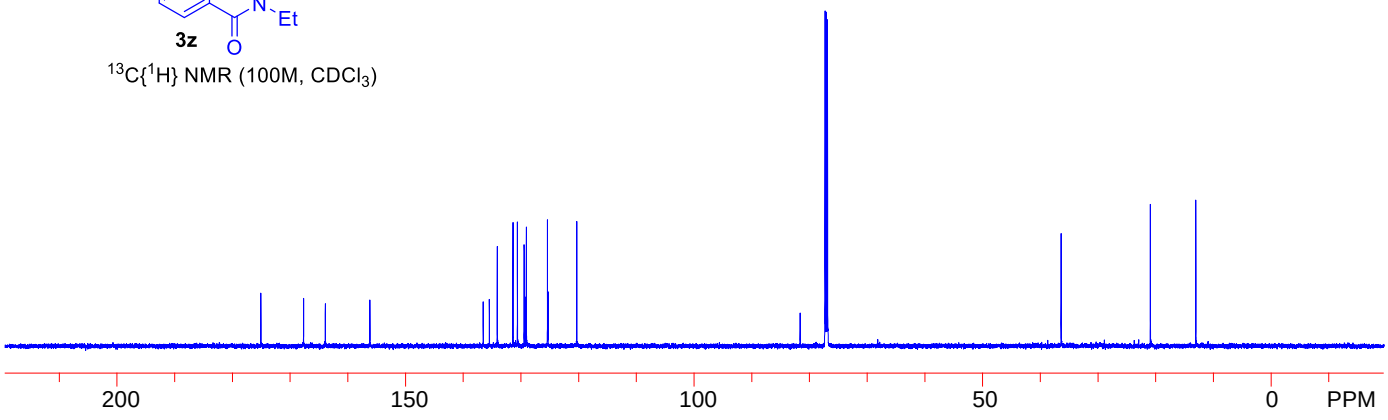


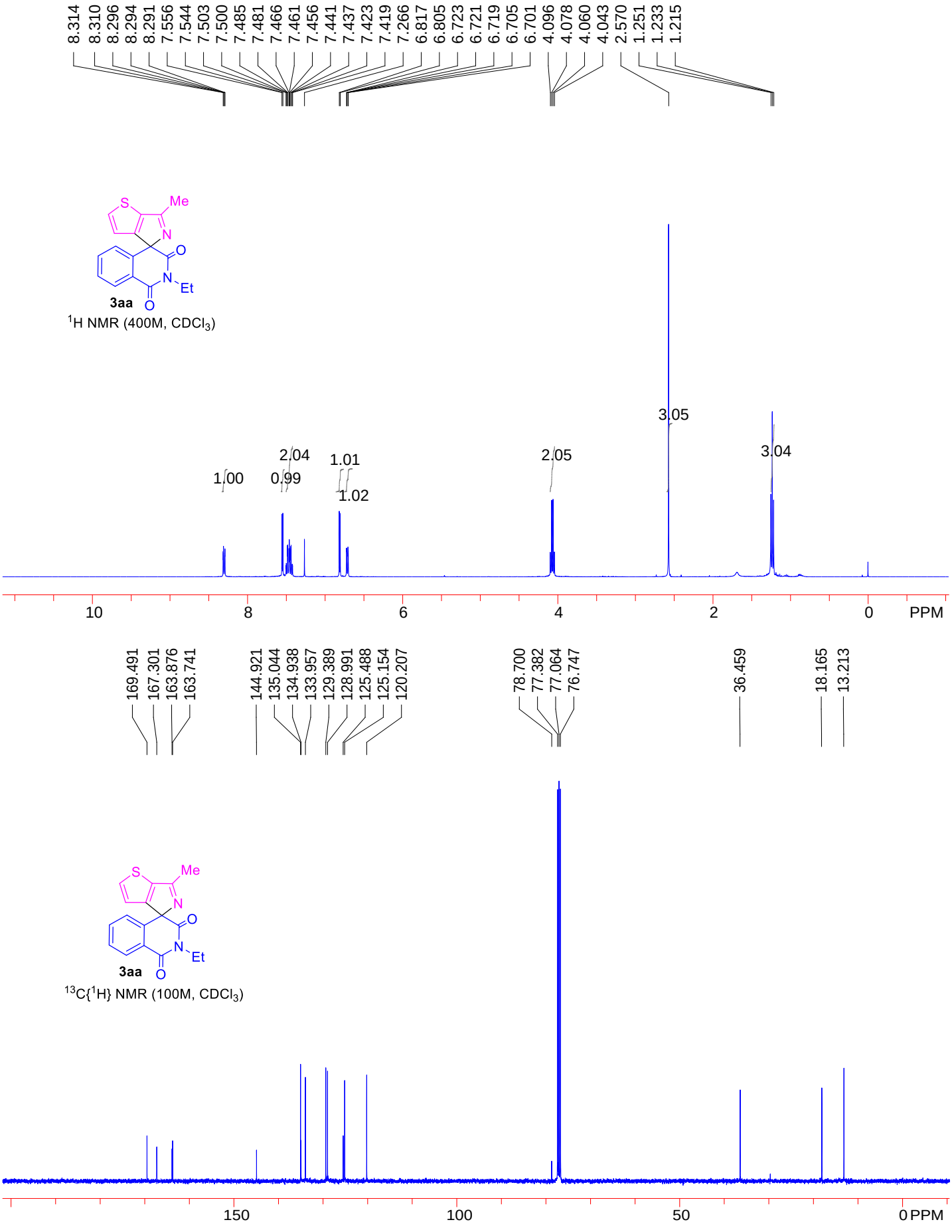




3z

$^{13}C\{^{1}H\}$ NMR (100M, $CDCl_3$)

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6.705
6.701
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4.078
4.060
4.043
2.570
1.251
1.233
1.215
Me
S
N
O
N
Et
O
3aa
1H NMR (400M, CDCl3)
1.00
2.04
0.99
1.01
1.02
2.05
3.05
3.04
10
8
6
4
2
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PPM
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120.207
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77.382
77.064
76.747
36.459
18.165
13.213
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N
O
N
Et
O
3aa
13C{1H} NMR (100M, CDCl3)
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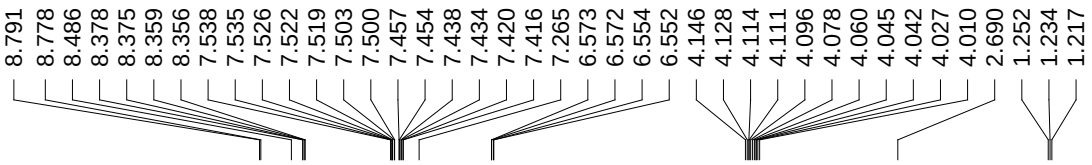
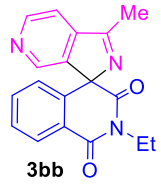
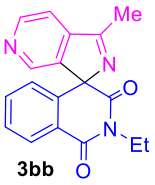
0 PPM





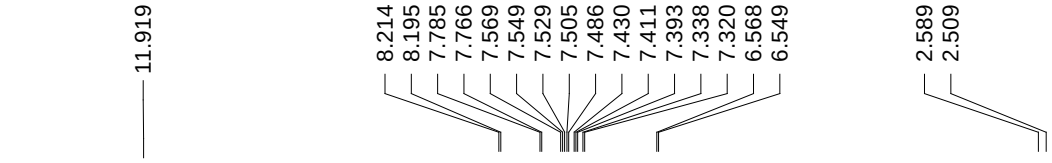
3bb

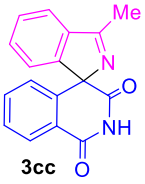
^{1}H NMR (400M, $CDCl_3$)



3bb

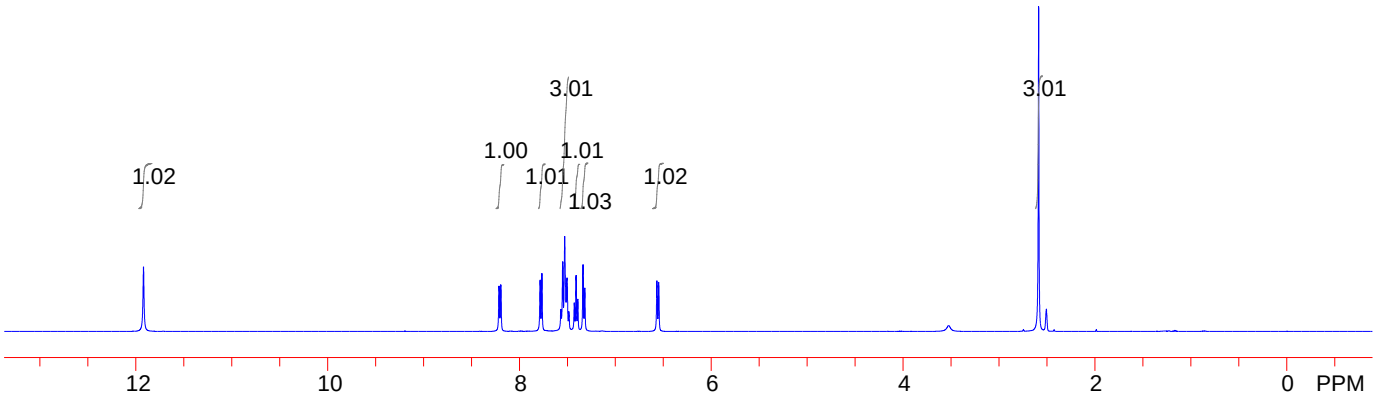
^{13}C{^{1}H} NMR (100M, $CDCl_3$)

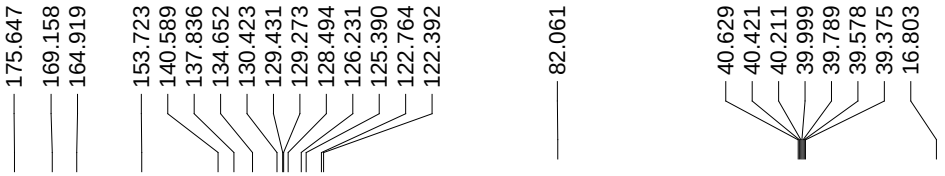


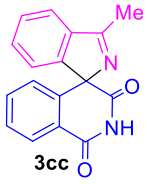


3cc

^{1}H NMR (400M, DMSO-d_6)

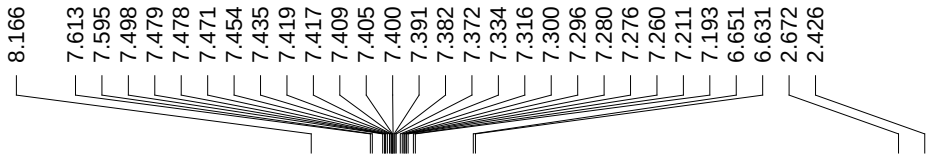


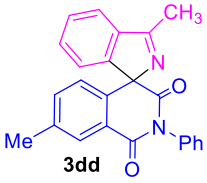




3cc

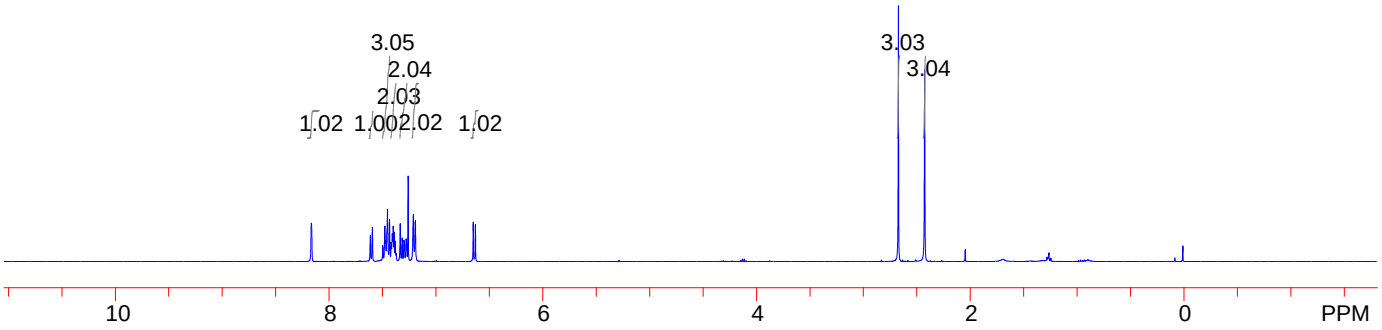
^{13}C{^{1}H} NMR (100M, DMSO-d_6)

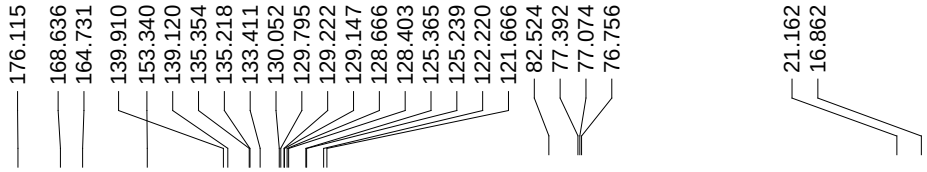


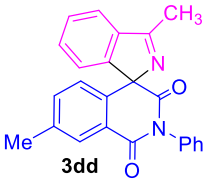


3dd

^{1}H NMR (400M, $CDCl_3$)

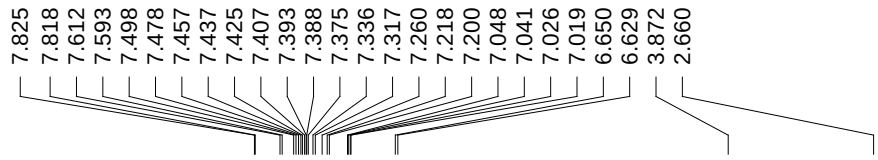


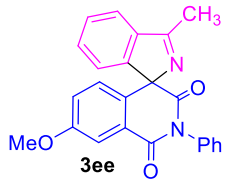




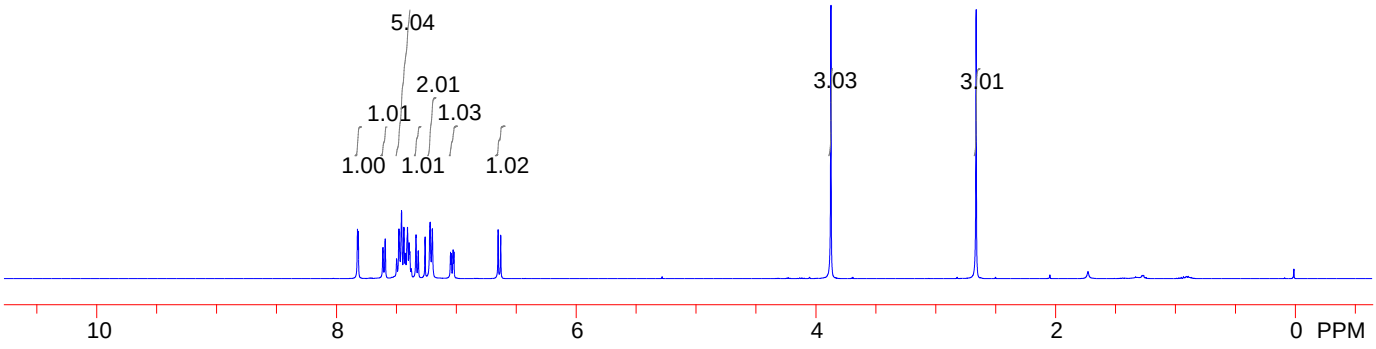
3dd

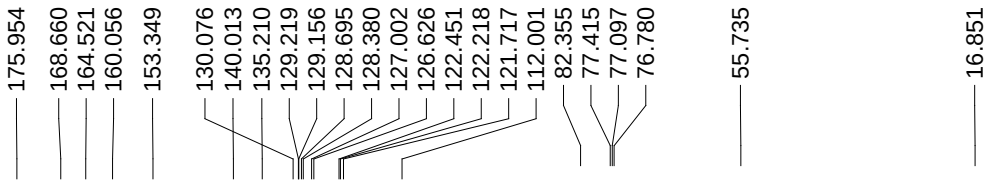
^{13}C{^{1}H} NMR (100M, $CDCl_3$)

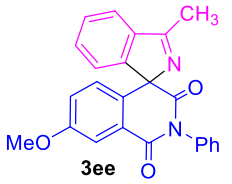




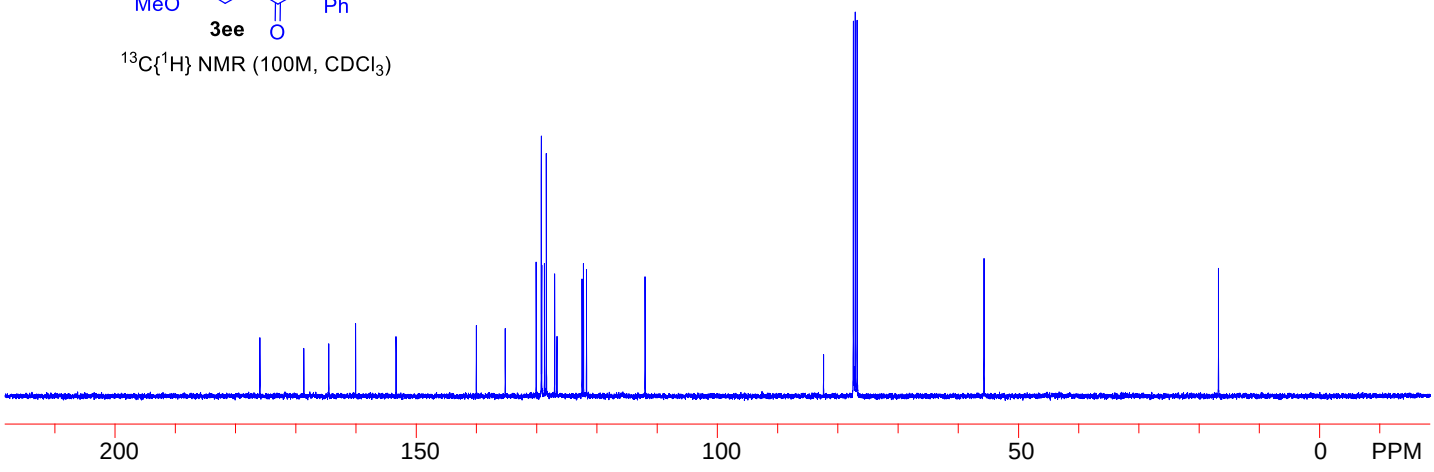
^{1}H NMR (400M, $CDCl_3$)



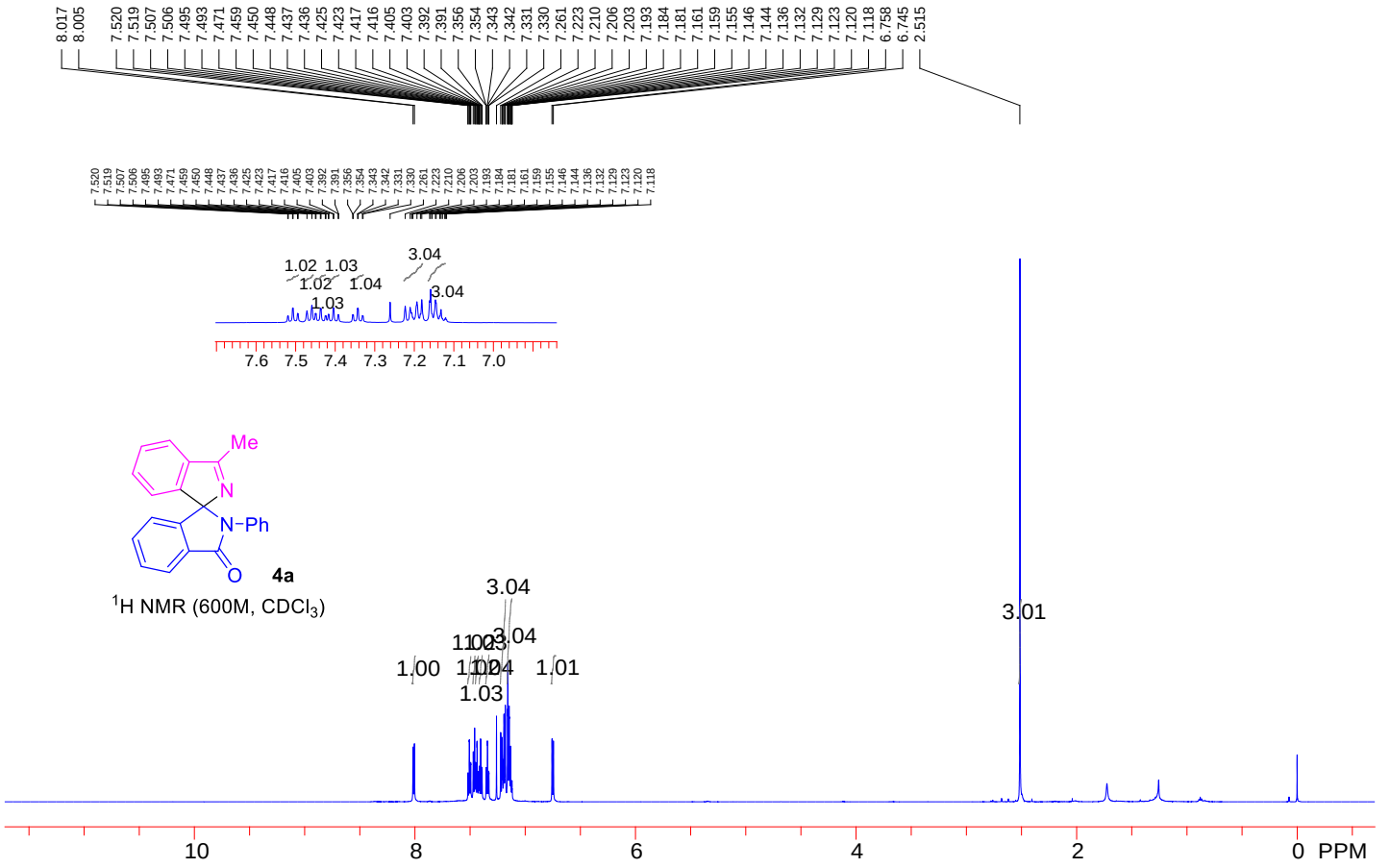


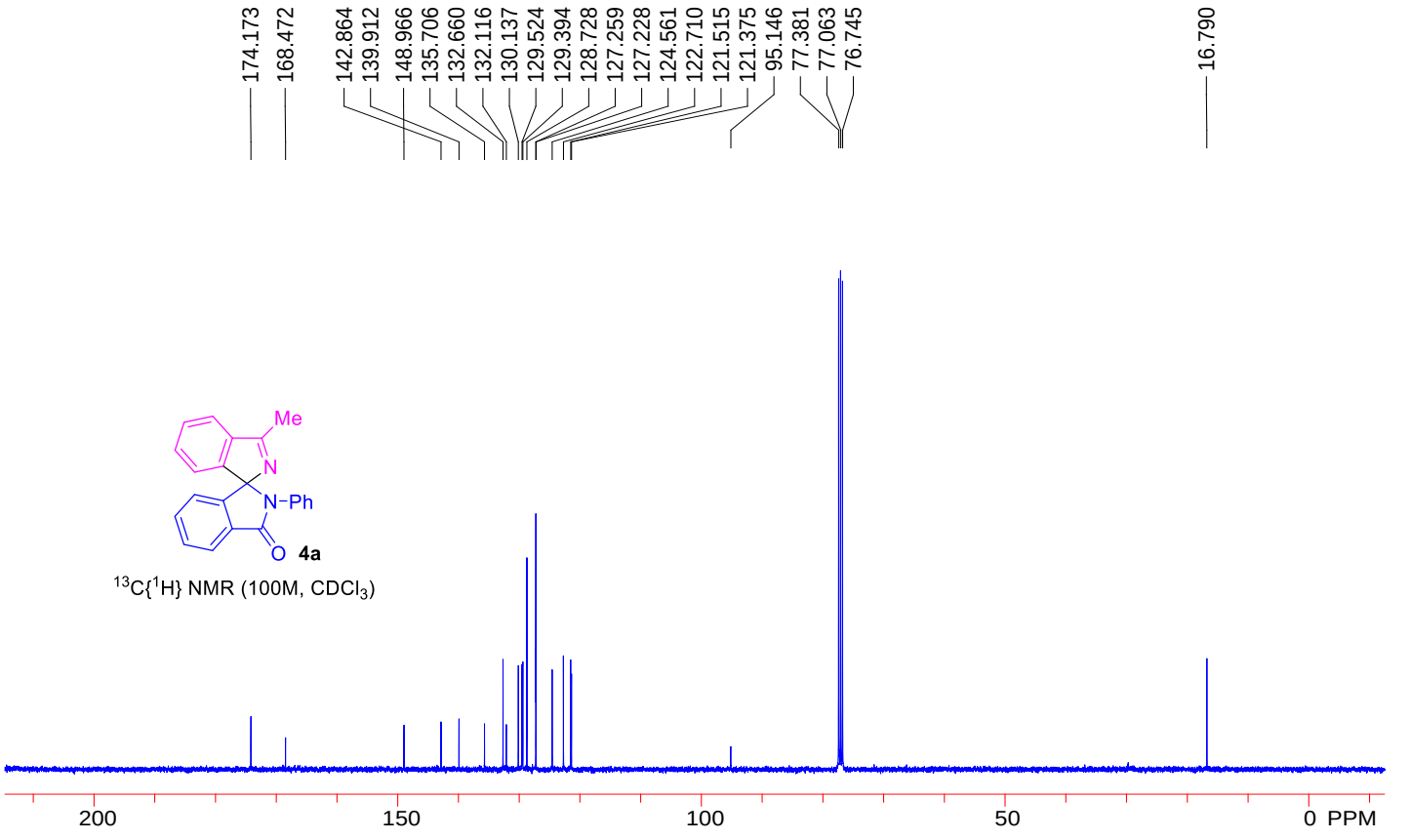


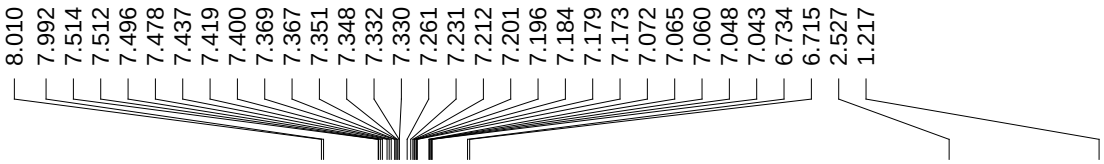
$^{13}C\{^{1}H\}$ NMR (100M, $CDCl_3$)

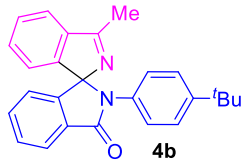


V. NMR spectra of 4a-4ee



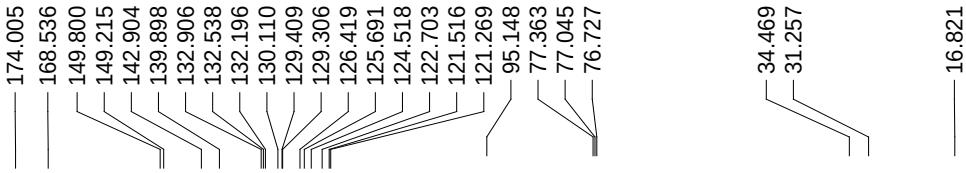


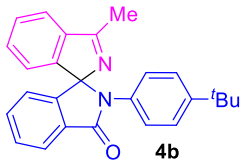




4b

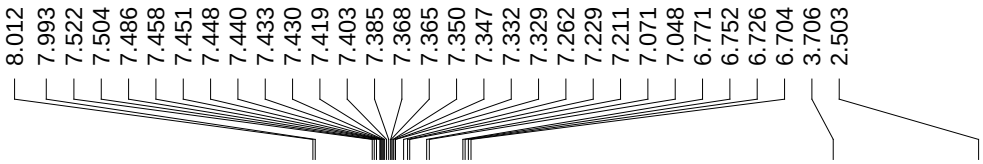
^{1}H NMR (400M, $CDCl_3$)

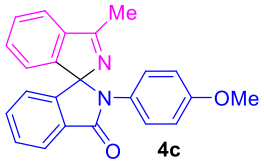




4b

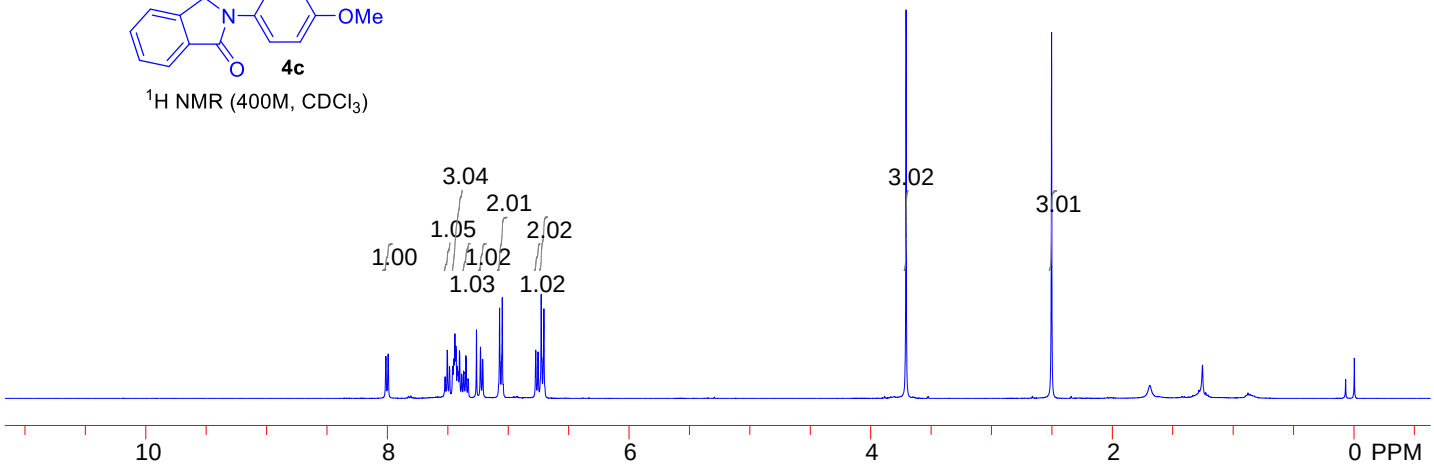
$^{13}C\{^{1}H\}$ NMR (100M, $CDCl_3$)

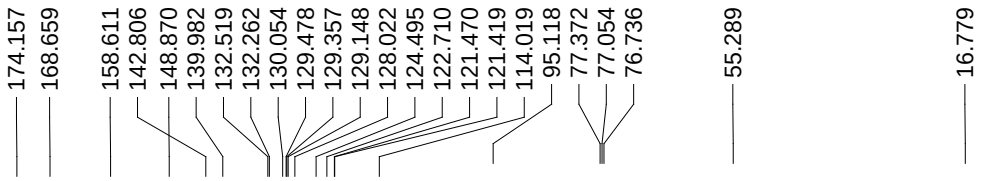


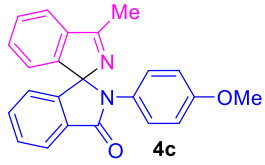


4c

^{1}H NMR (400M, $CDCl_3$)

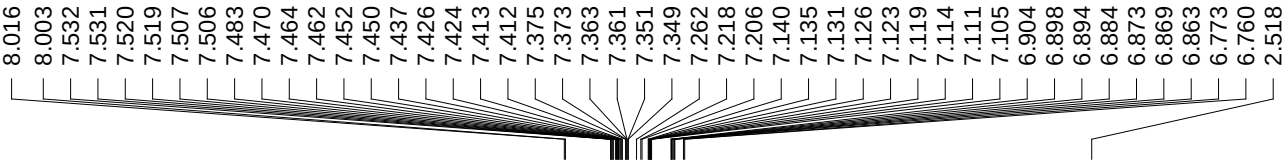


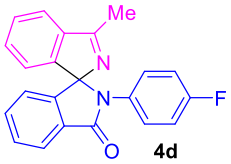




4c

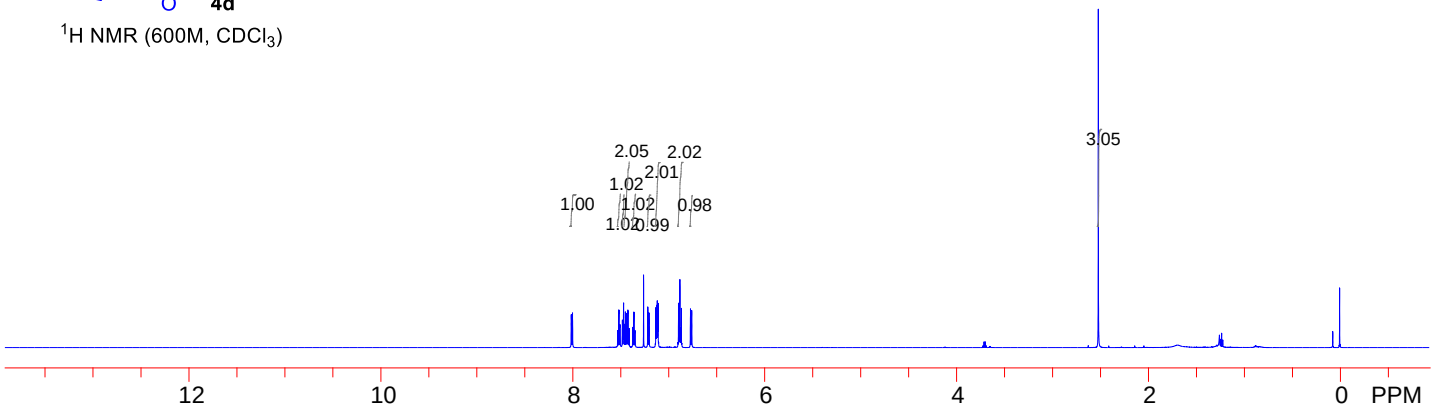
^{13}C{^{1}H} NMR (100M, $CDCl_3$)

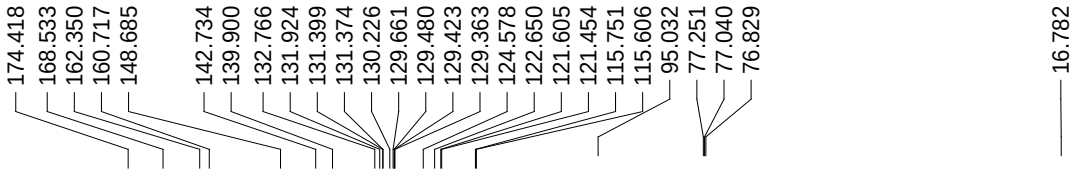


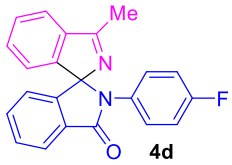


4d

^{1}H NMR (600M, $CDCl_3$)

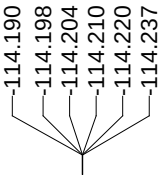


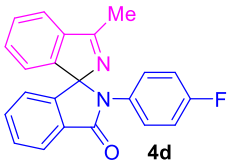




4d

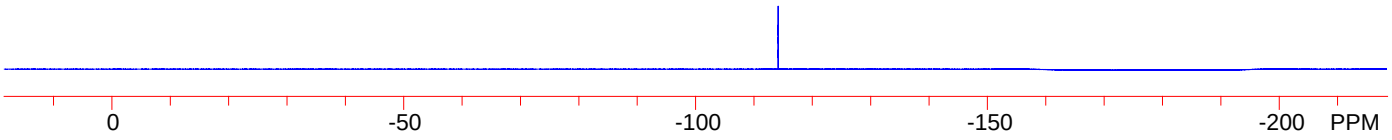
^{13}C{^{1}H} NMR (150M, $CDCl_3$)

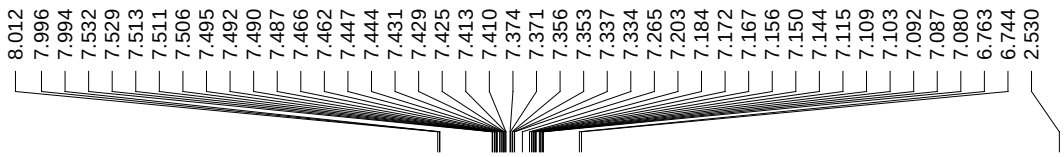


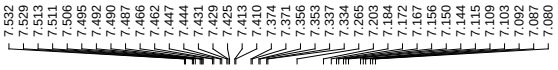


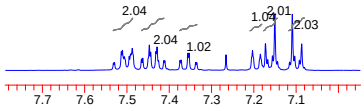
4d

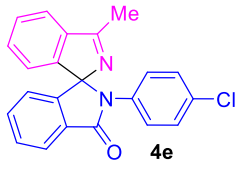
^{19}F NMR (565M, $CDCl_3$)





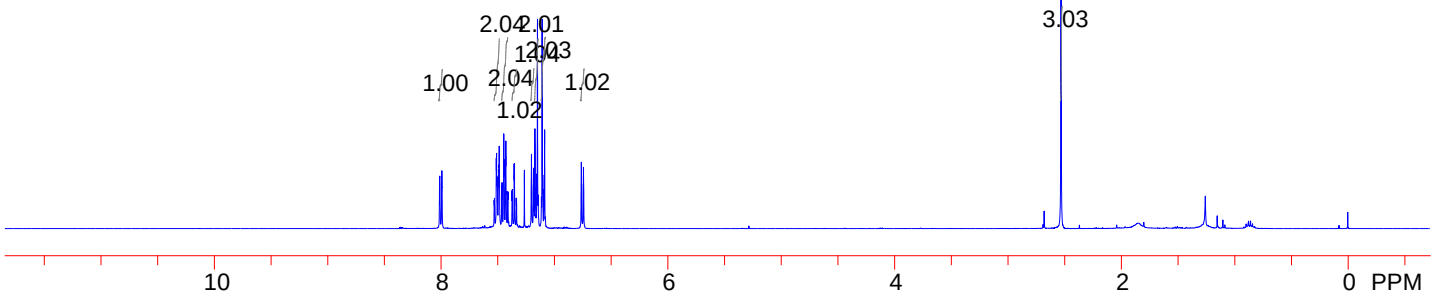


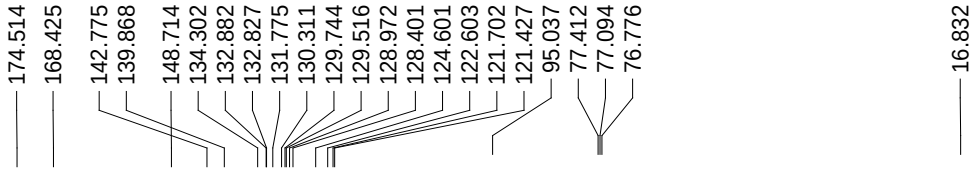


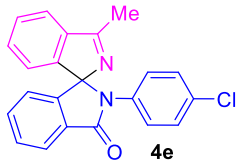


4e

^{1}H NMR (400M, $CDCl_3$)

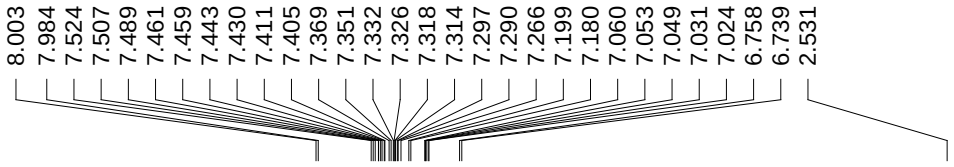


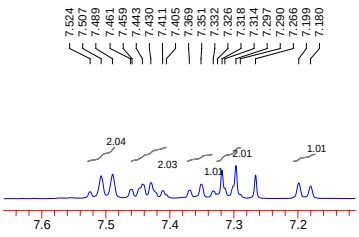


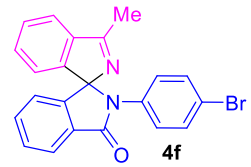


4e

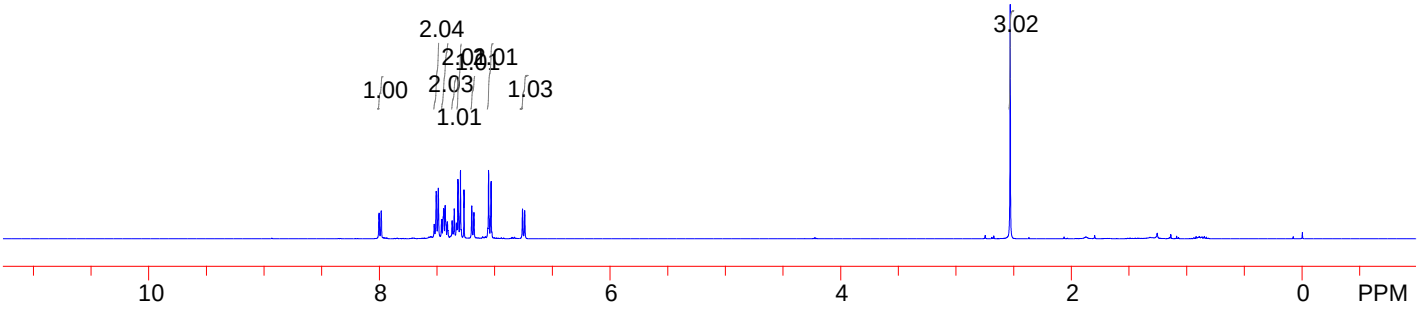
^{13}C{^{1}H} NMR (100M, $CDCl_3$)

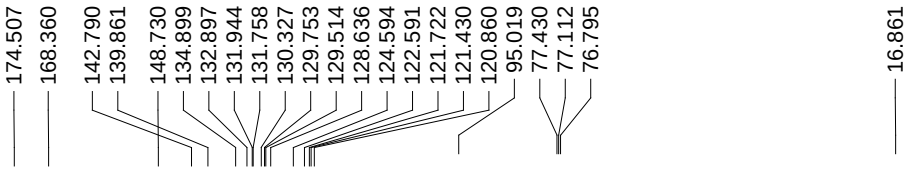


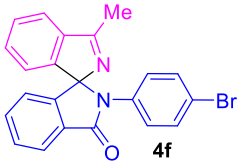




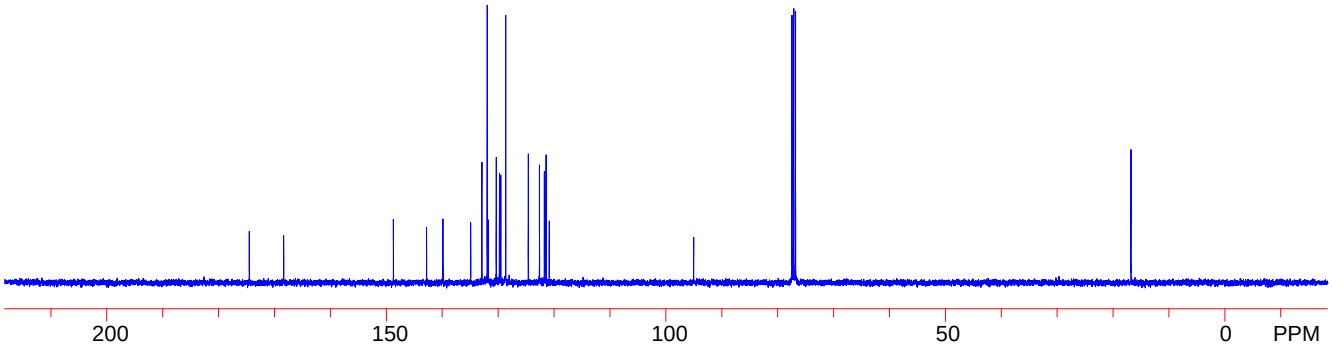
^{1}H NMR (600M, $CDCl_3$)

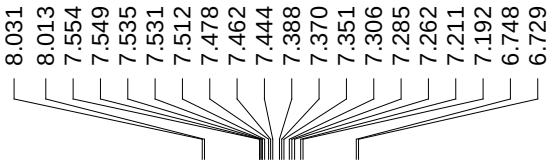


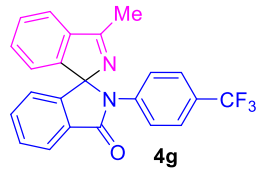




$^{13}C\{^{1}H\}$ NMR (150M, $CDCl_3$)

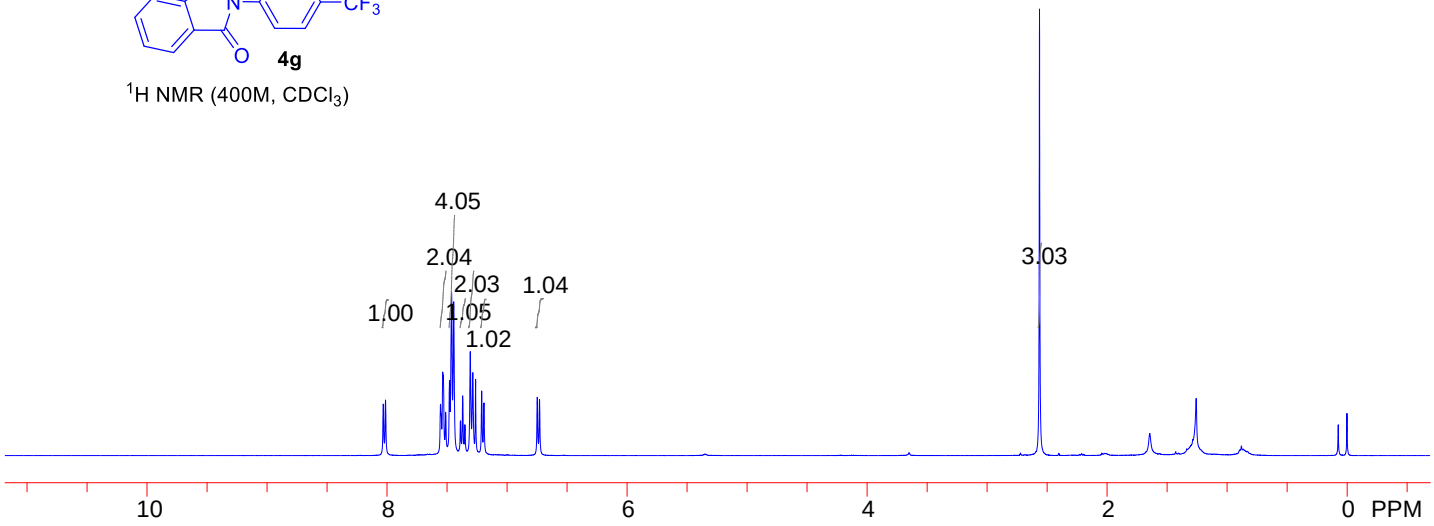


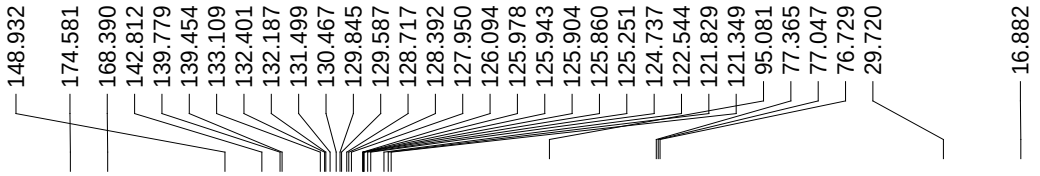


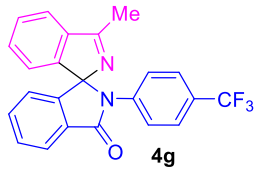


4g

^{1}H NMR (400M, $CDCl_3$)

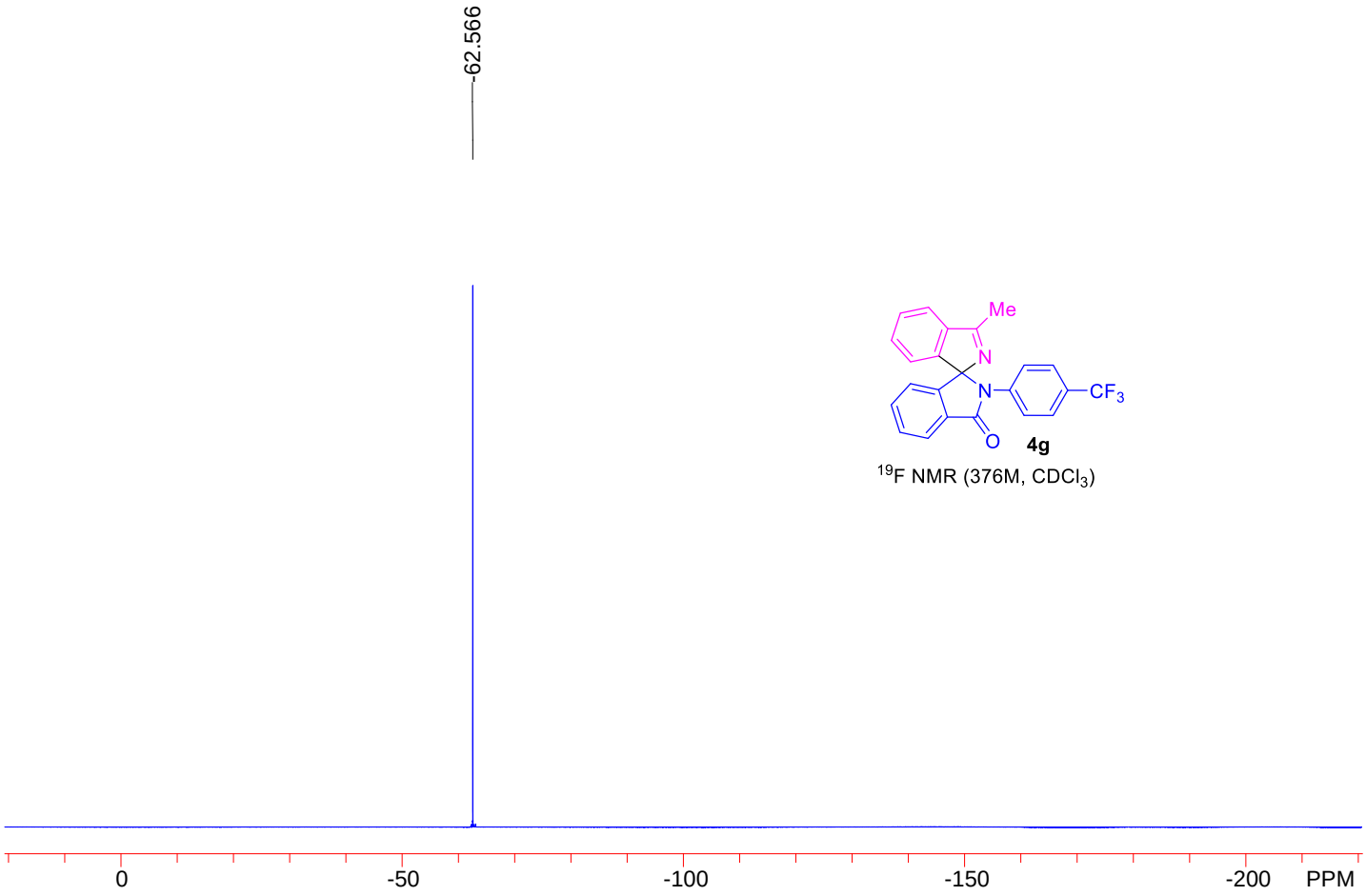






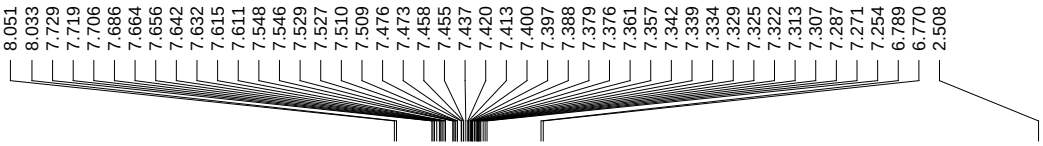
4g

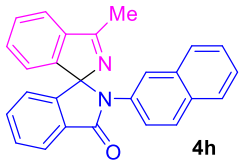
^{13}C{^{1}H} NMR (100M, $CDCl_3$)



4g

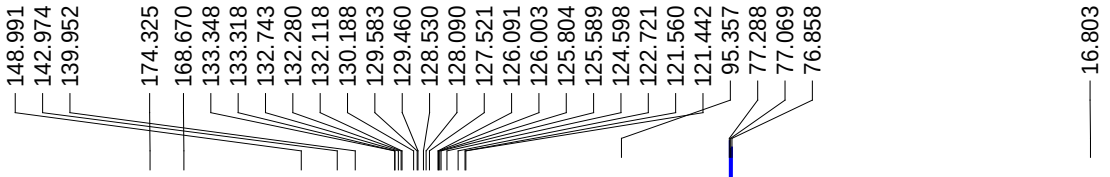
^{19}F NMR (376M, $CDCl_3$)

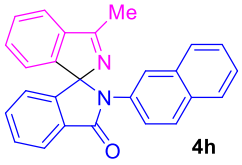




4h

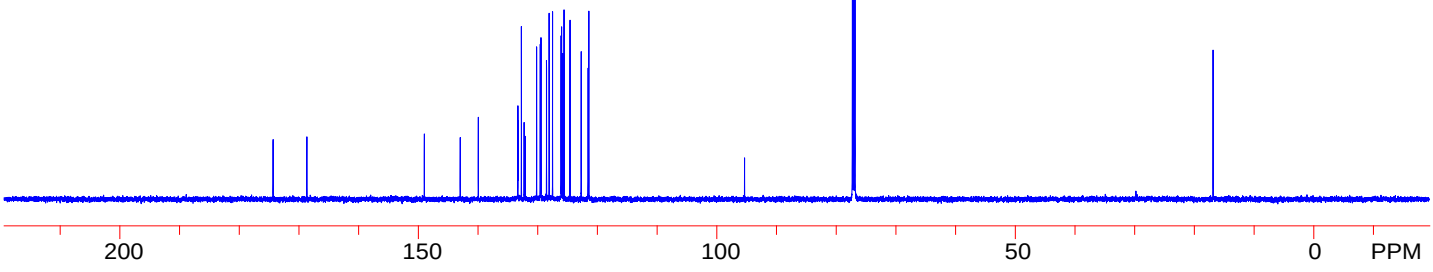
^{1}H NMR (400M, $CDCl_3$)

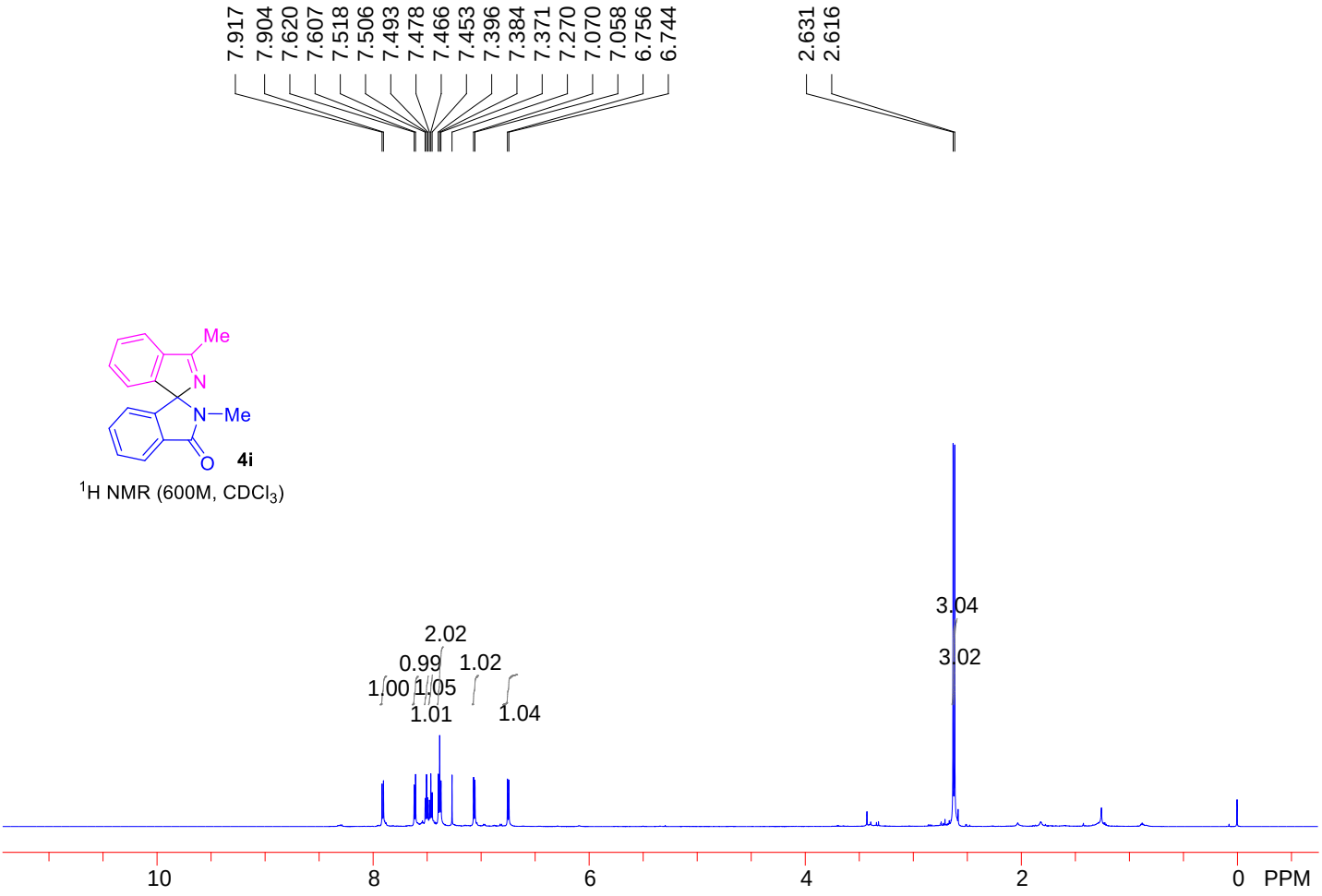




4h

^{13}C{^{1}H} NMR (150M, $CDCl_3$)

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7.270
7.070
7.058
6.756
6.744
2.631
2.616
Me
N
N—Me
O
4i
1H NMR (600M, CDCl3)
2.02
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1.00
1.05
1.01
1.04
3.04
3.02
10
8
6
4
2
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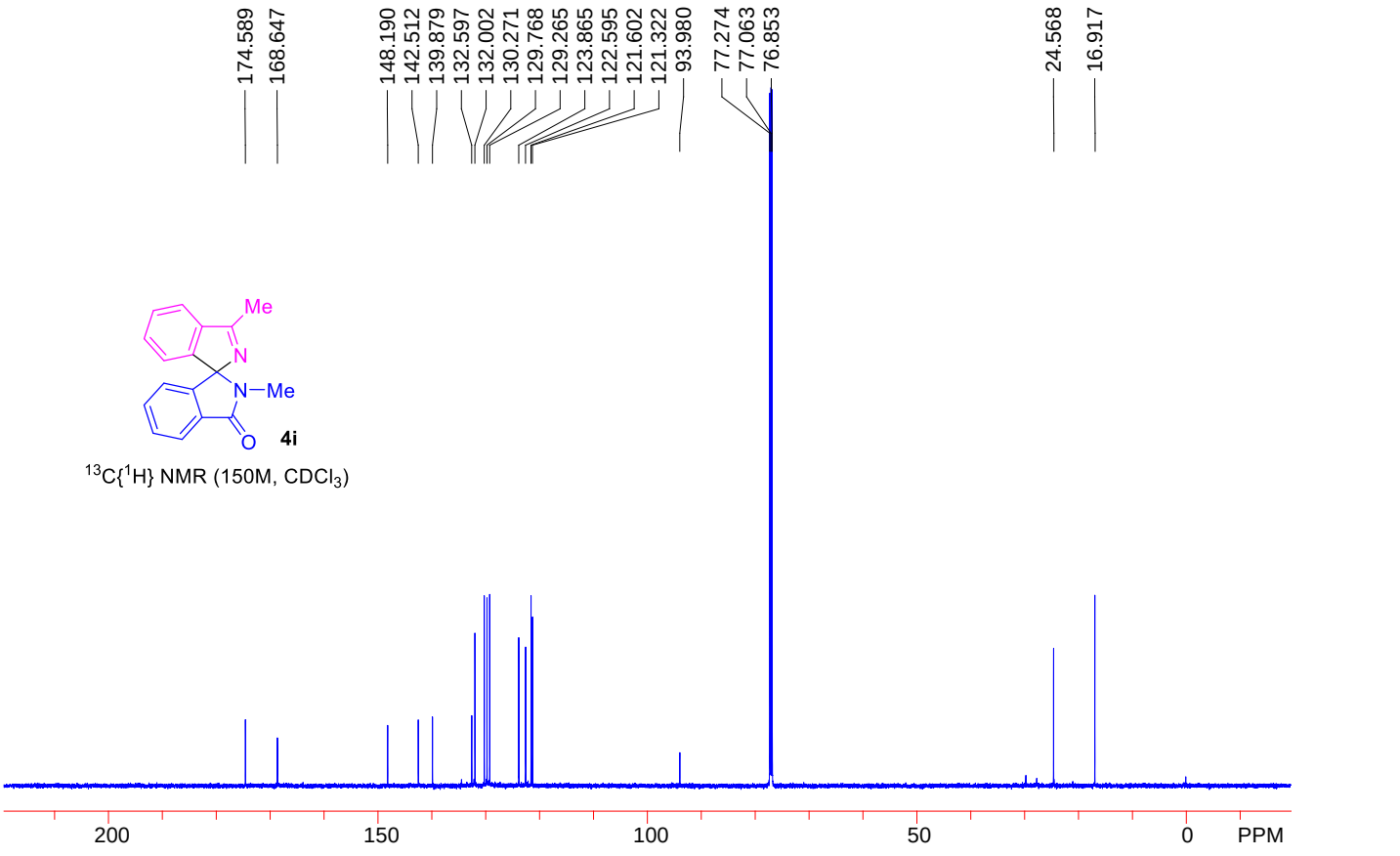
PPM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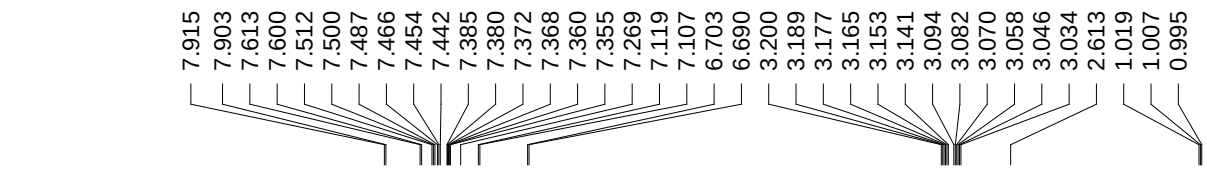
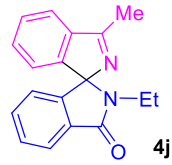
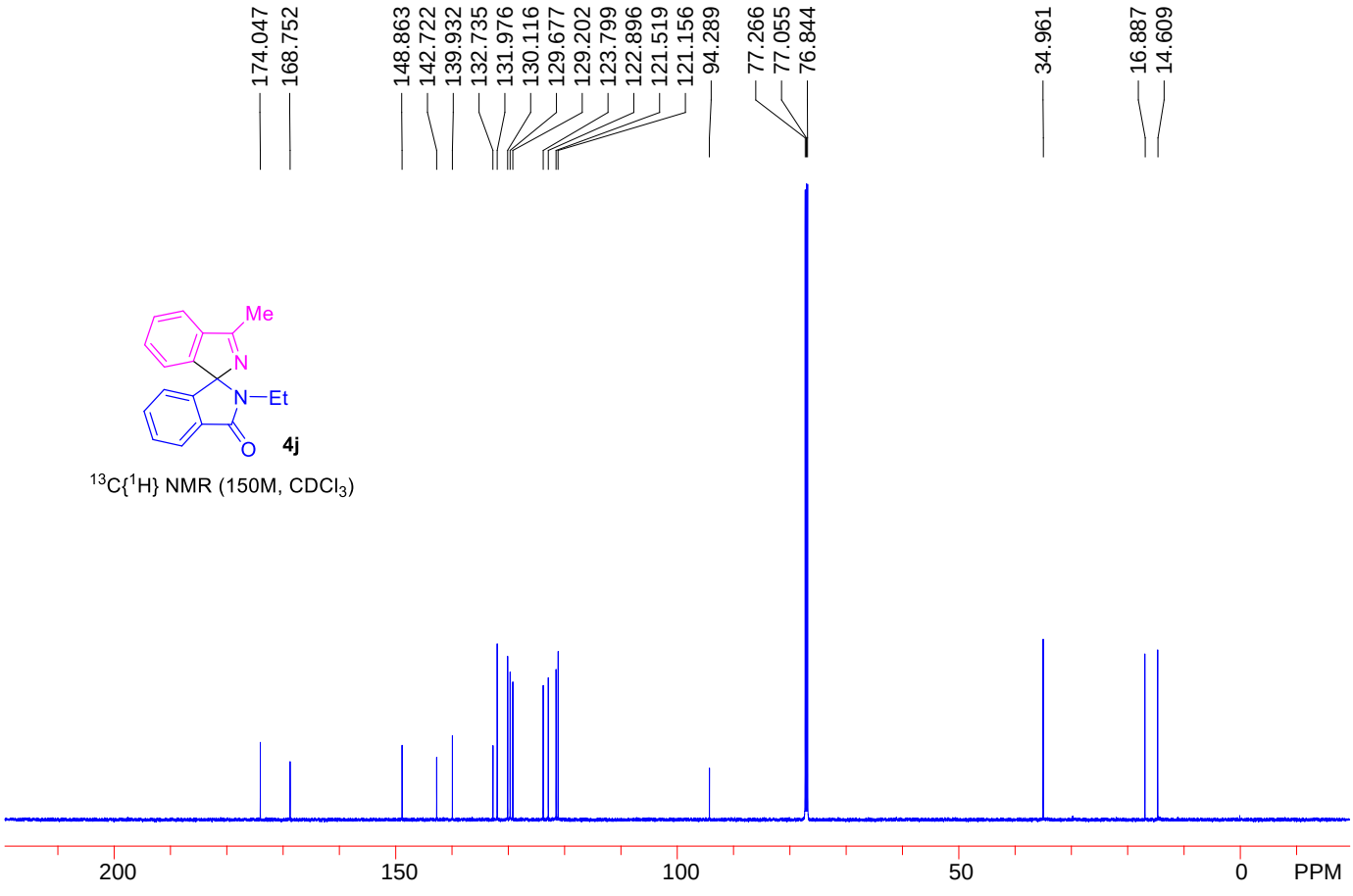
122.595
121.602
121.322
93.980
77.274
77.063
76.853
24.568
16.917
Me
N
N—Me
O
4i
13C{1H} NMR (150M, CDCl3)
200
150
100
50
0
PPM





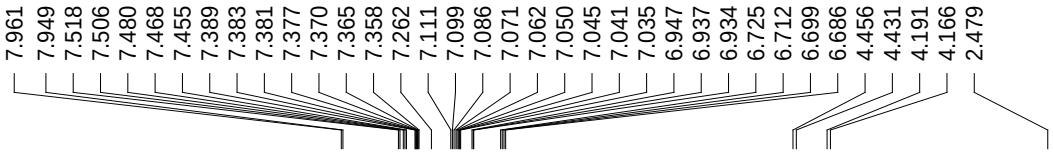
4j

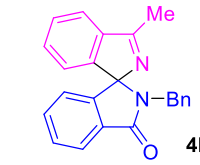
^{1}H NMR (600M, $CDCl_3$)



4j

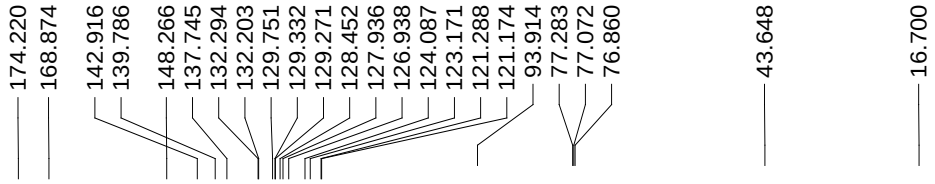
$^{13}C\{^{1}H\}$ NMR (150M, $CDCl_3$)

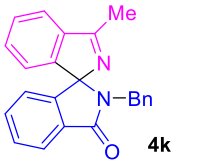




4k

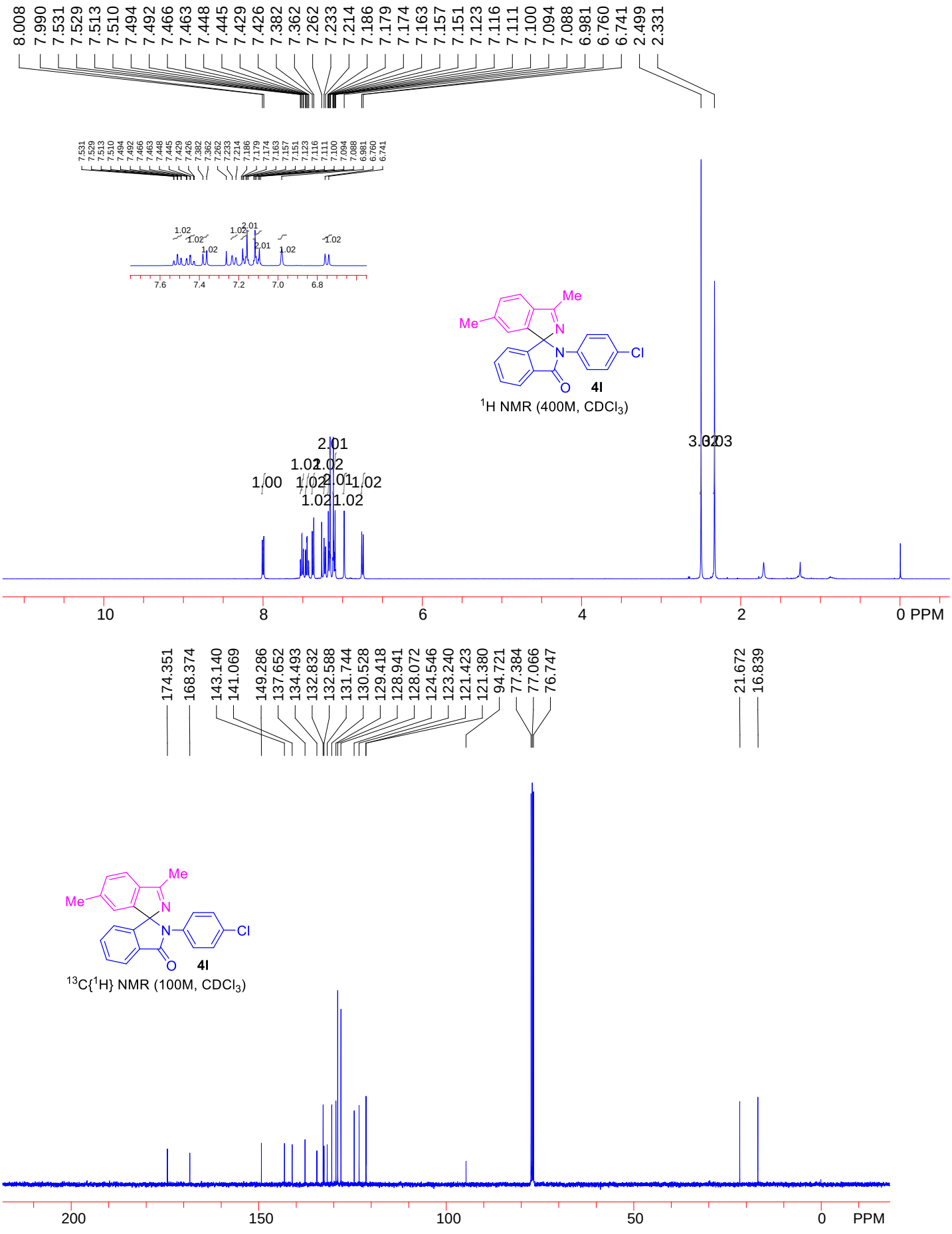
^{1}H NMR (600M, $CDCl_3$)

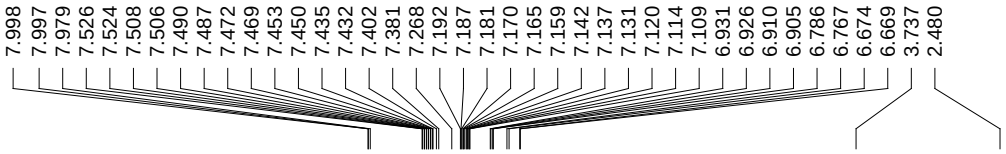


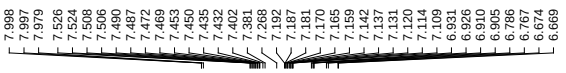


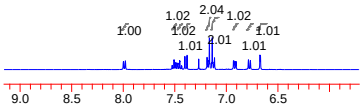
4k

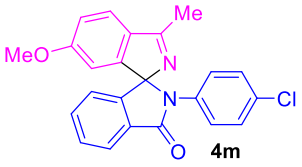
$^{13}C\{^{1}H\}$ NMR (150M, $CDCl_3$)


8.008
7.990
7.531
7.529
7.513
7.510
7.494
7.492
7.466
7.463
7.448
7.445
7.429
7.426
7.382
7.362
7.262
7.233
7.214
7.186
7.179
7.174
7.163
7.157
7.151
7.123
7.116
7.111
7.100
7.094
7.088
6.981
6.760
6.741
2.499
2.331
Me
Me
N
N
Cl
O
4l
1H NMR (400M, CDCl3)
2.01
1.02
1.00
3.03
10
8
6
4
2
0 PPM
174.351
168.374
143.140
141.069
149.286
137.652
134.493
132.832
132.588
131.744
130.528
129.418
128.941
128.072
124.546
123.240
121.423
121.380
94.721
77.384
77.066
76.747
21.672
16.839
13C{1H} NMR (100M, CDCl3)
200
150
100
50
0 PPM

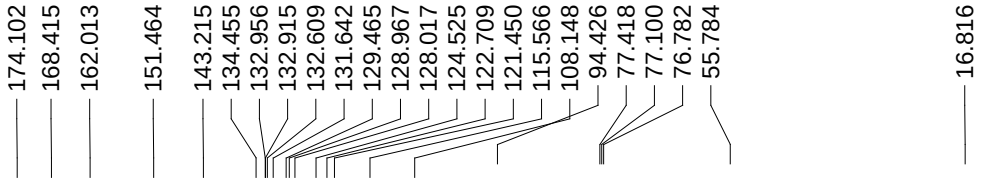


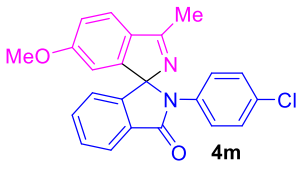




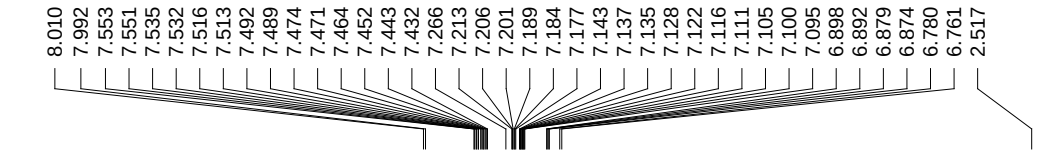


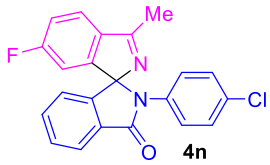
^{1}H NMR (400M, $CDCl_3$)





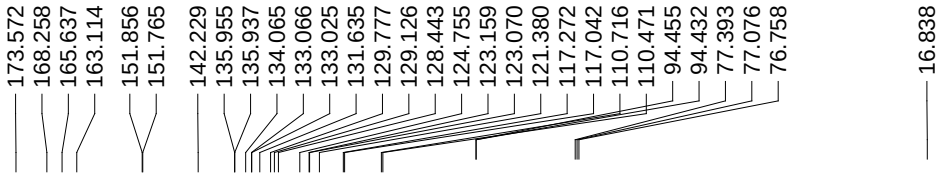
$^{13}C\{^1H\}$ NMR (100M, $CDCl_3$)

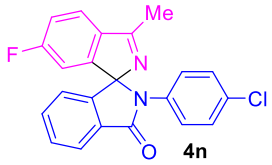




4n

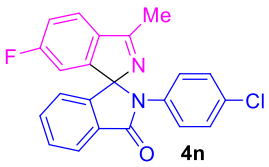
^{1}H NMR (400M, $CDCl_3$)





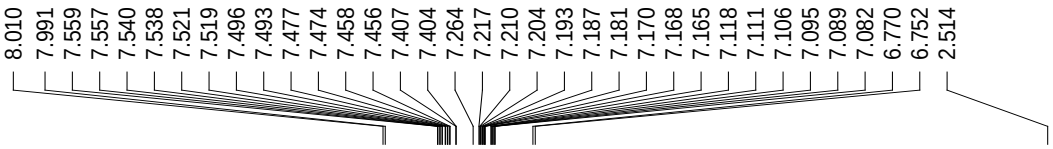
4n

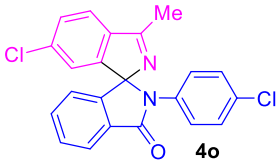
^{13}C{^{1}H} NMR (100M, $CDCl_3$)



4n

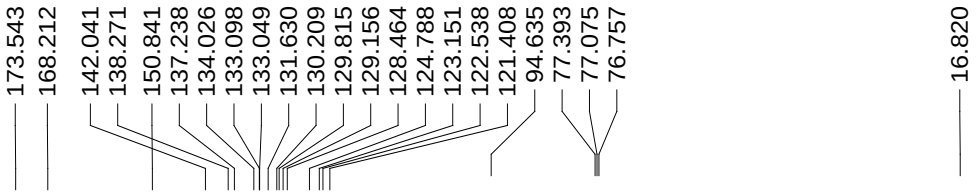
^{19}F NMR (376M, $CDCl_3$)

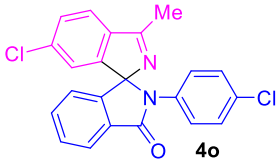




4o

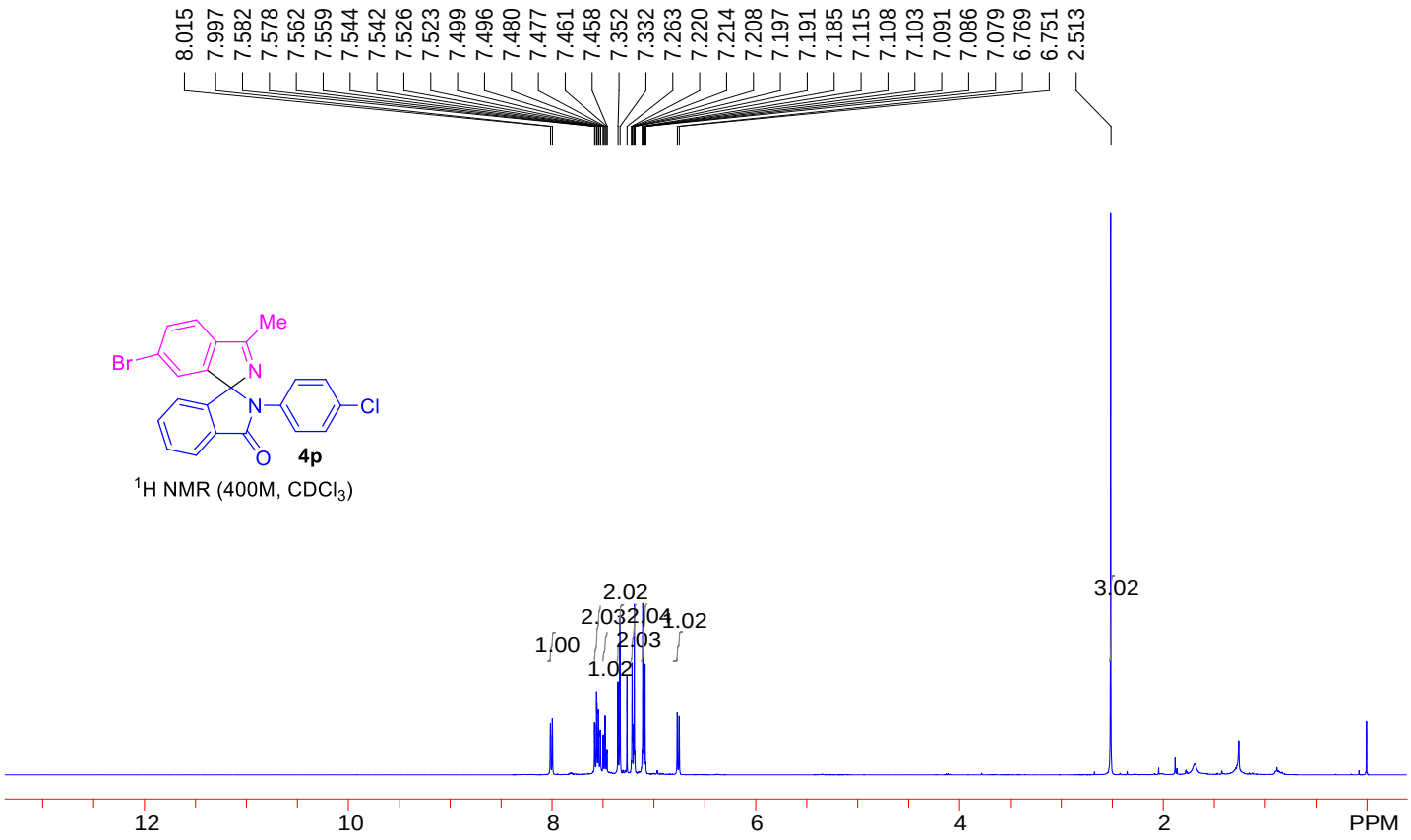
^{1}H NMR (400M, $CDCl_3$)

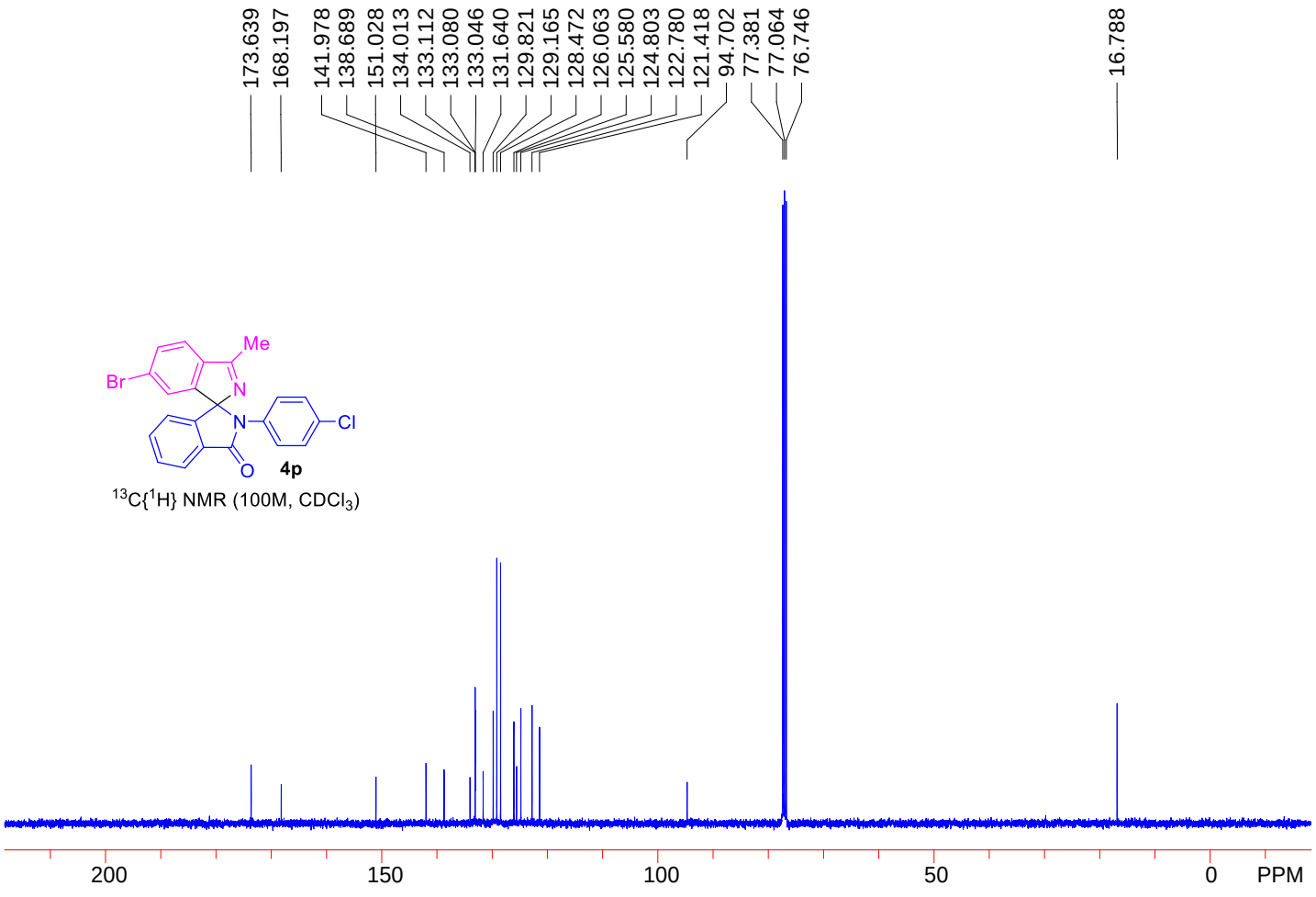


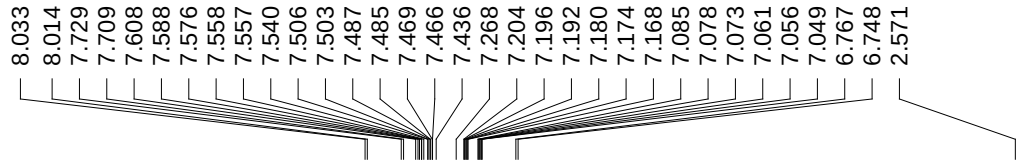


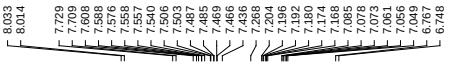
4o

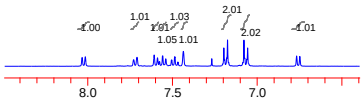
$^{13}C\{^{1}H\}$ NMR (100M, $CDCl_3$)

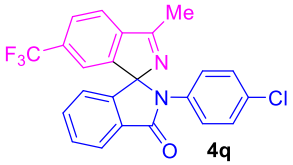

8.015
7.997
7.582
7.578
7.562
7.559
7.544
7.542
7.526
7.523
7.499
7.496
7.480
7.477
7.461
7.458
7.352
7.332
7.263
7.220
7.214
7.208
7.197
7.191
7.185
7.115
7.108
7.103
7.091
7.086
7.079
6.769
6.751
2.513
Me
Br
N
N
Cl
O
4p
^{1}H NMR (400M, CDCl$_3$)
2.02
2.03
2.04
1.02
1.00
2.03
1.02
3.02
12
10
8
6
4
2
PPM


173.639
168.197
141.978
138.689
151.028
134.013
133.112
133.080
133.046
131.640
129.821
129.165
128.472
126.063
125.580
124.803
122.780
121.418
94.702
77.381
77.064
76.746
16.788
Me
Br
N
N
Cl
O
4p
^{13}C{^{1}H} NMR (100M, CDCl$_3$)
200
150
100
50
0
PPM

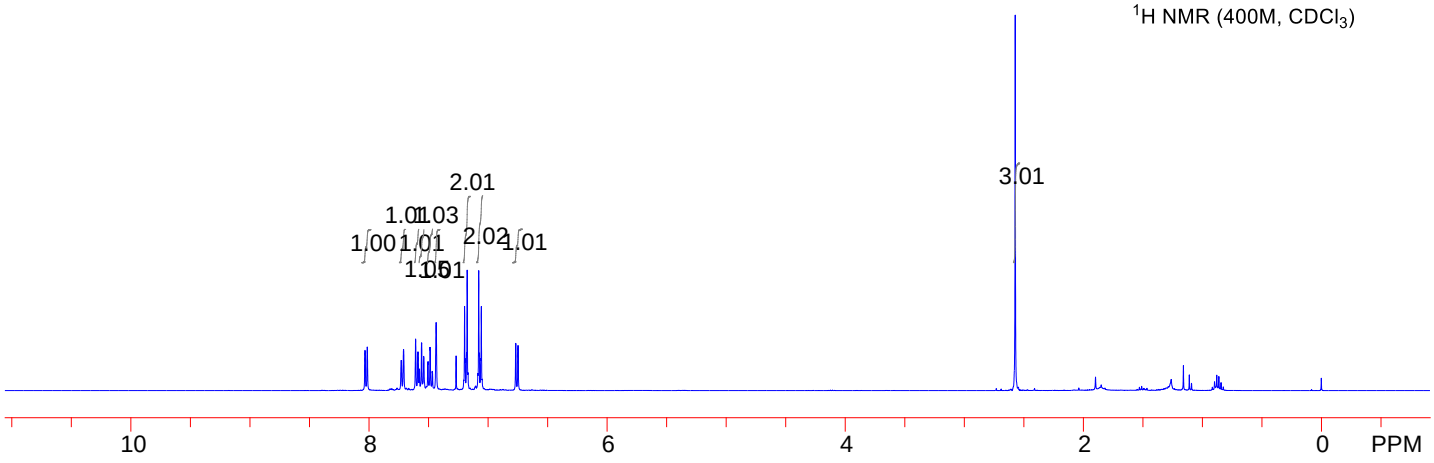


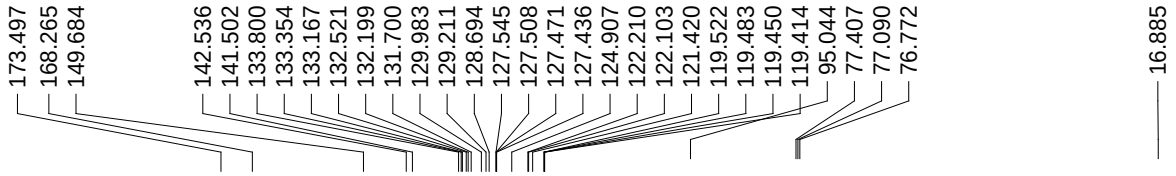


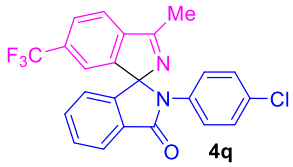




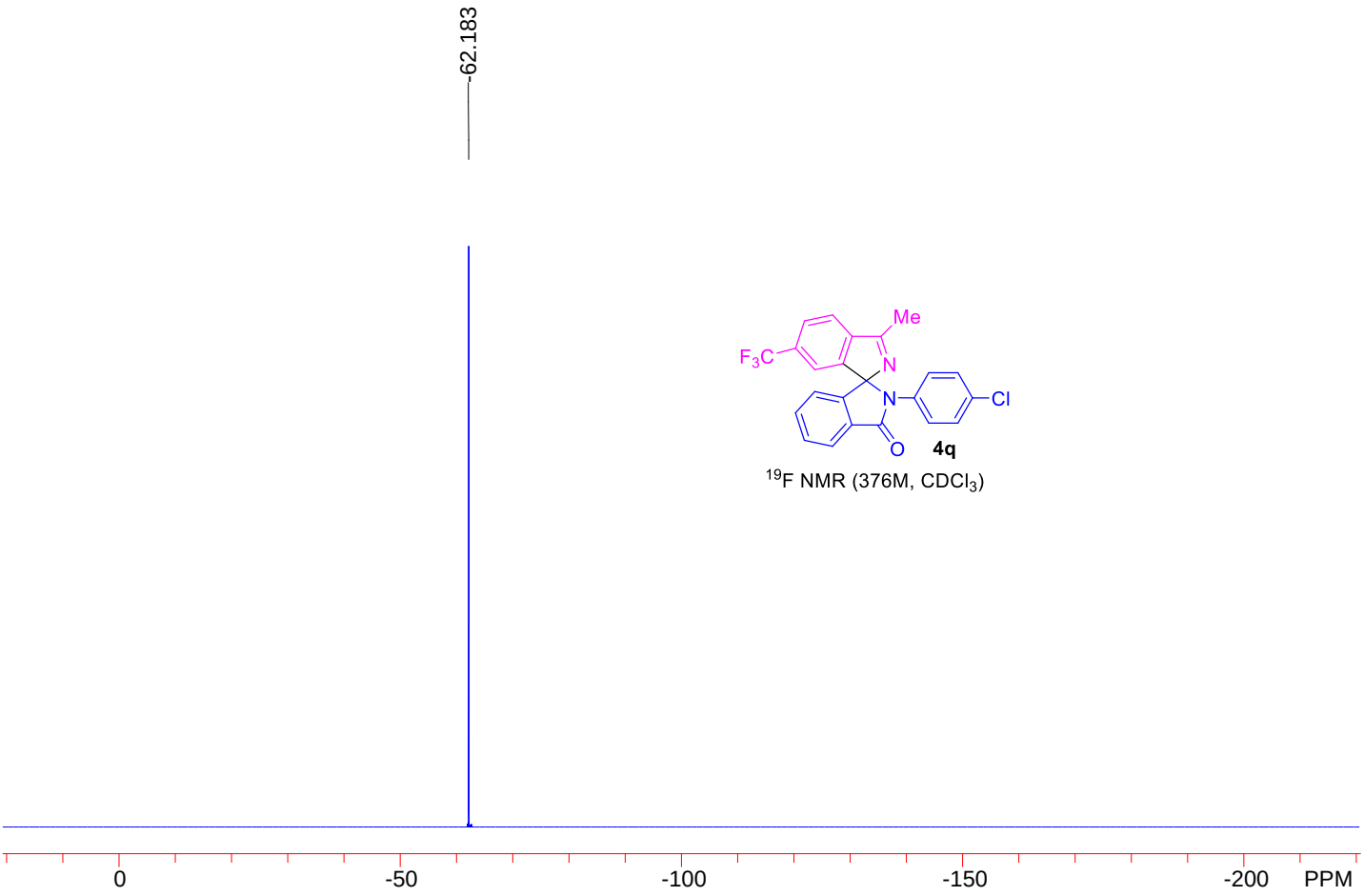
^{1}H NMR (400M, $CDCl_3$)

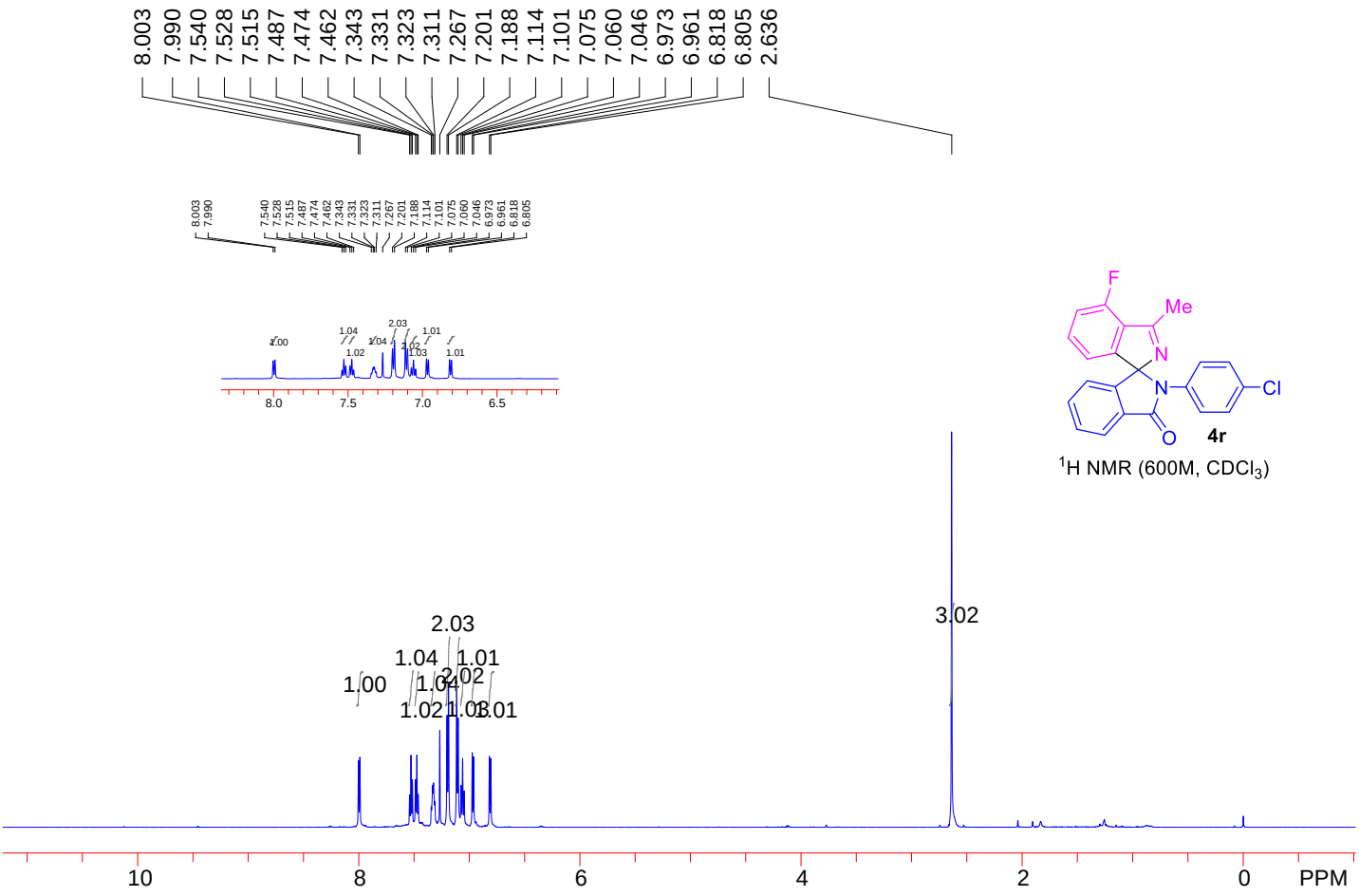


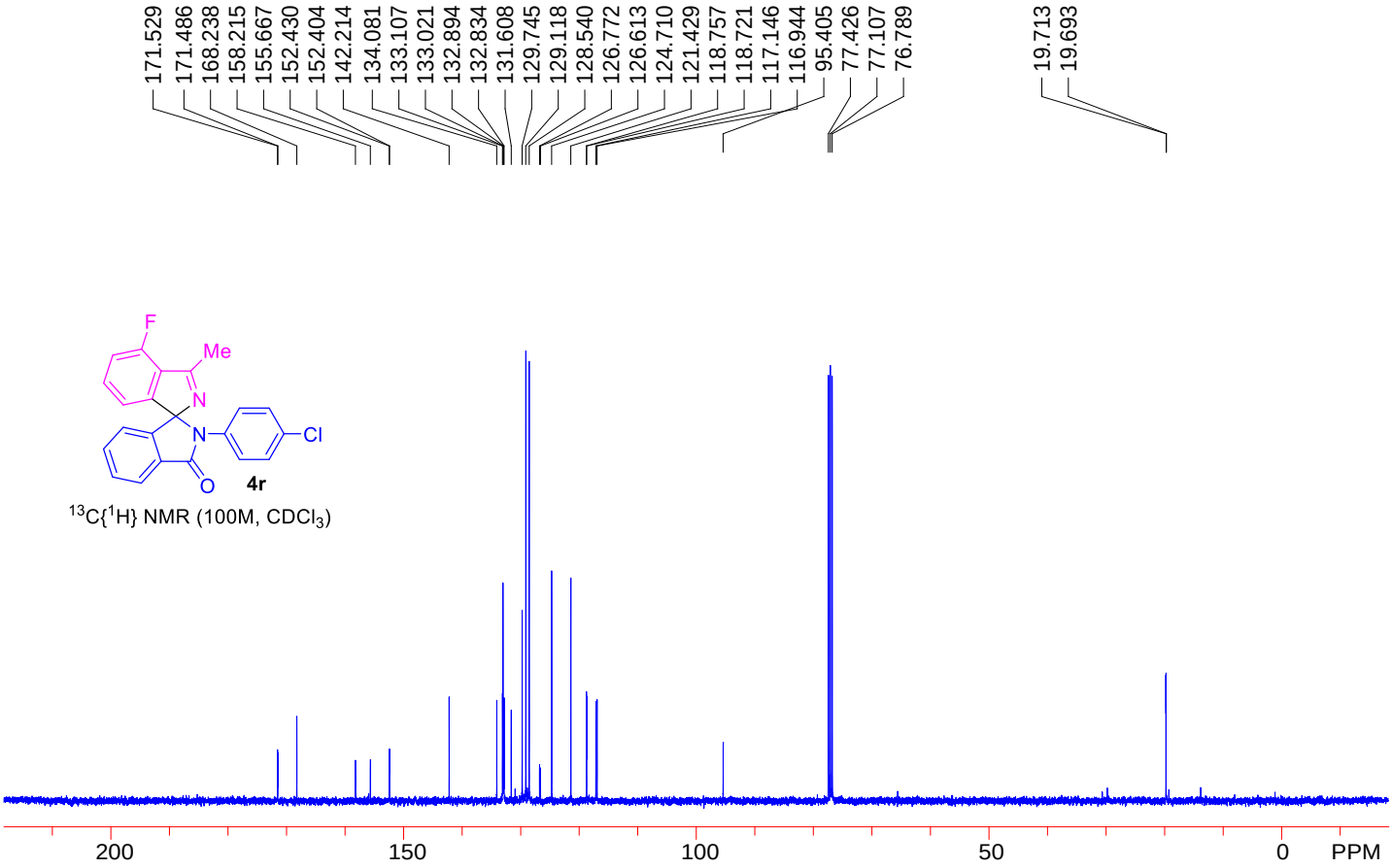


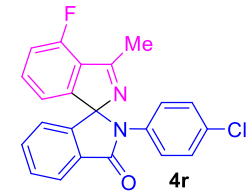


^{13}C{^{1}H} NMR (100M, $CDCl_3$)


-62.183
Me
F3C
N
N
Cl
O
4q
19F NMR (376M, CDCl3)
0
-50
-100
-150
-200
PPM

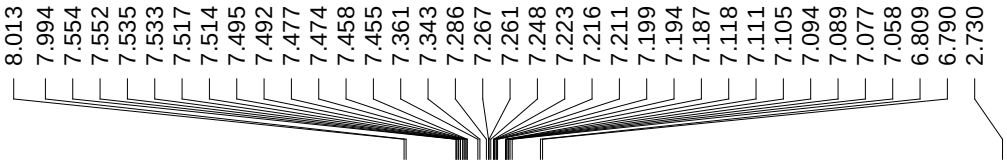

8.003
7.990
7.540
7.528
7.515
7.487
7.474
7.462
7.343
7.331
7.323
7.311
7.267
7.201
7.188
7.114
7.101
7.075
7.060
7.046
6.973
6.961
6.818
6.805
2.636
F
Me
N
N
Cl
O
4r
1H NMR (600M, CDCl3)
1.00
1.04
1.02
2.03
2.02
1.03
1.01
3.02
10
8
6
4
2
0
PPM

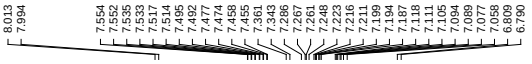

171.529
171.486
168.238
158.215
155.667
152.430
152.404
142.214
134.081
133.107
133.021
132.894
132.834
131.608
129.745
129.118
128.540
126.772
126.613
124.710
121.429
118.757
118.721
117.146
116.944
95.405
77.426
77.107
76.789
19.713
19.693
F
Me
N
N
Cl
O
4r
13C{1H} NMR (100M, CDCl3)
200
150
100
50
0
PPM

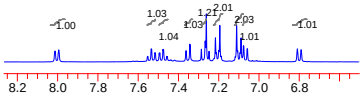


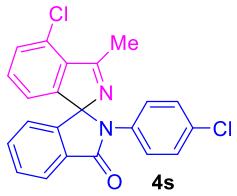
4r

^{19}F NMR (565M, $CDCl_3$)

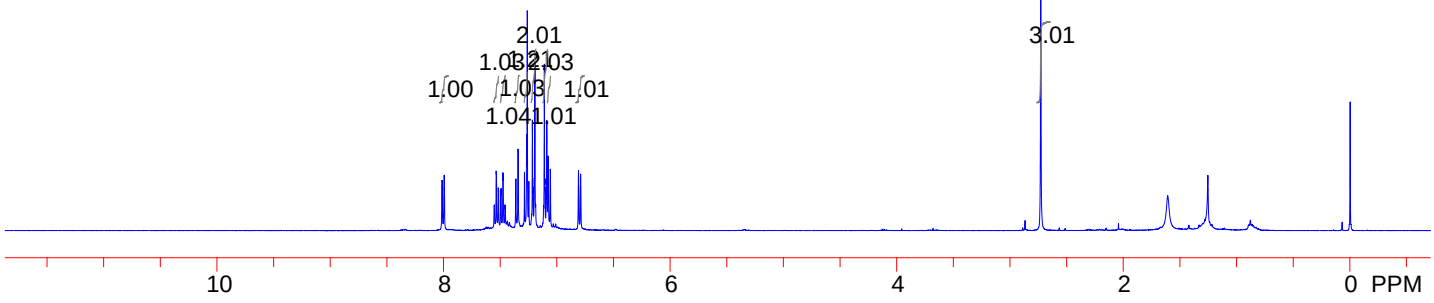


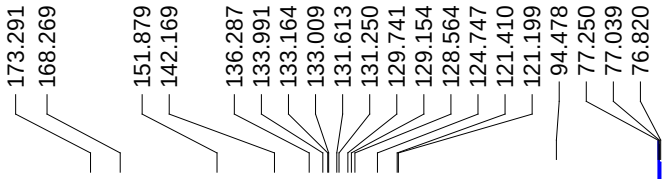


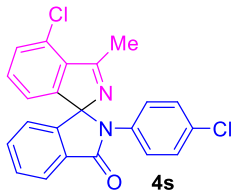




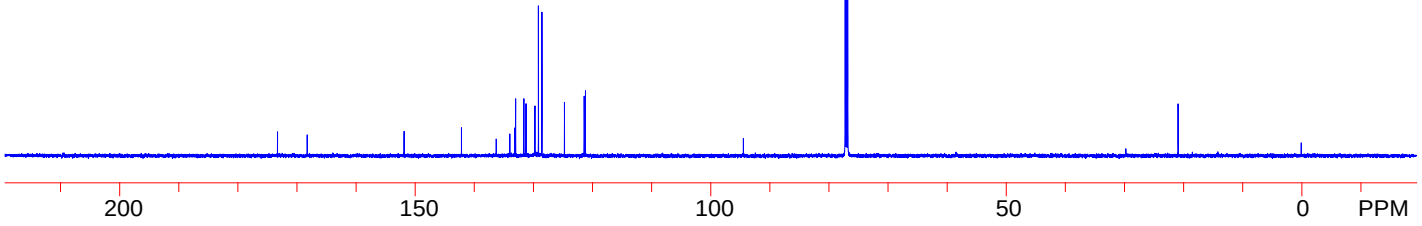
^{1}H NMR (400M, $CDCl_3$)

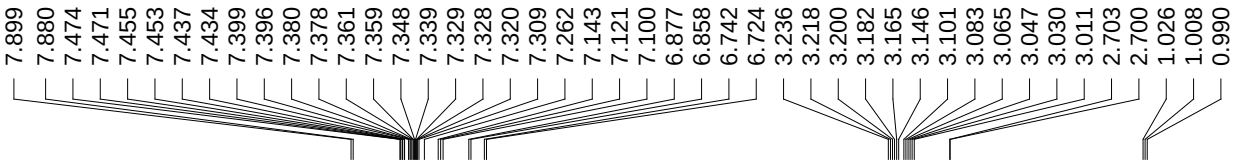


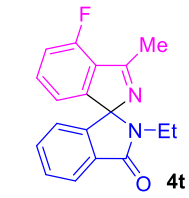




^{13}C{^{1}H} NMR (150M, $CDCl_3$)

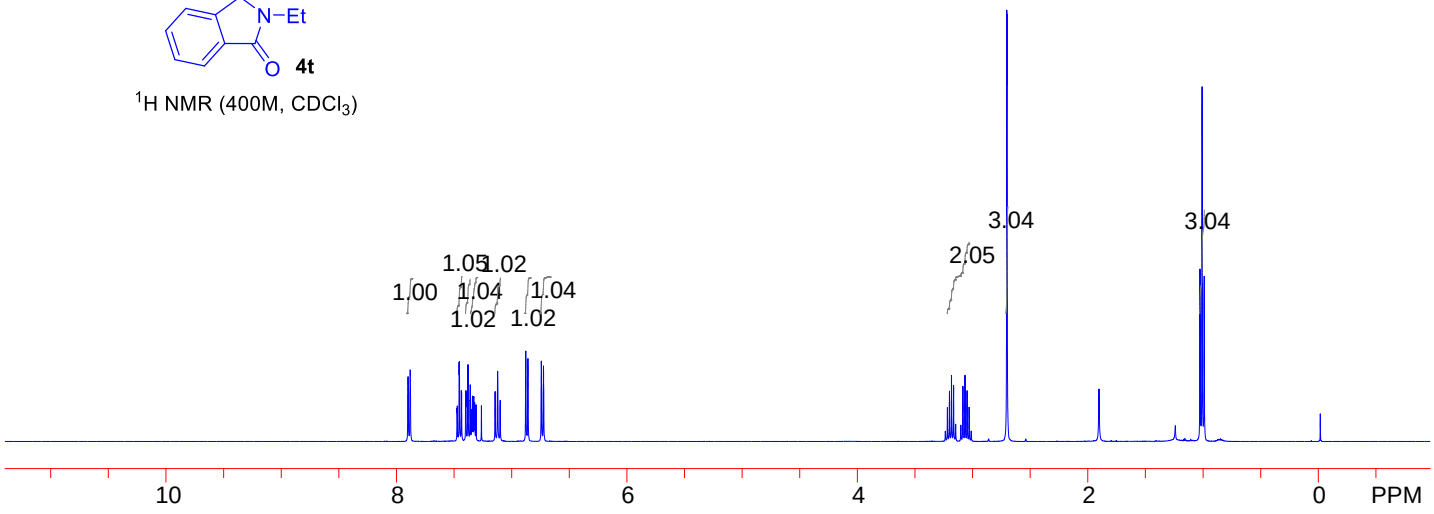


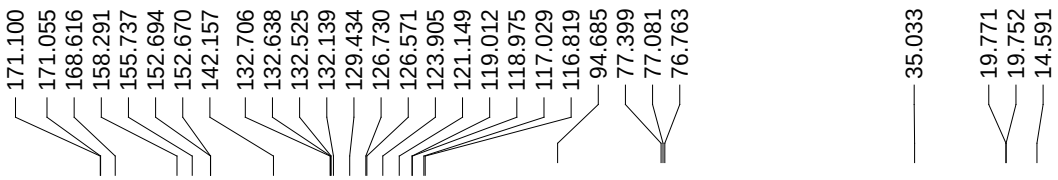


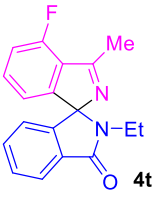


4t

^{1}H NMR (400M, $CDCl_3$)

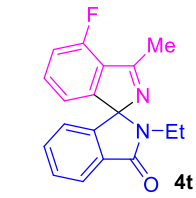






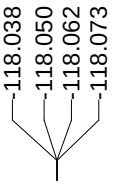
4t

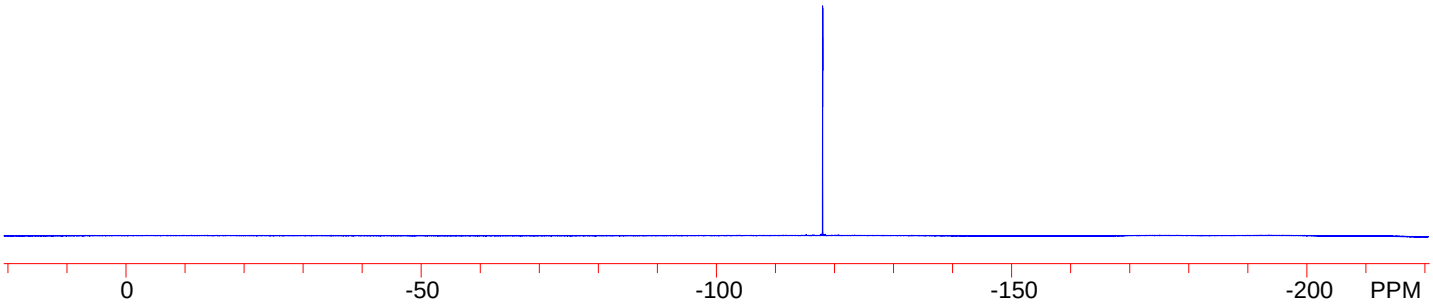
^{13}C{^{1}H} NMR (100M, $CDCl_3$)

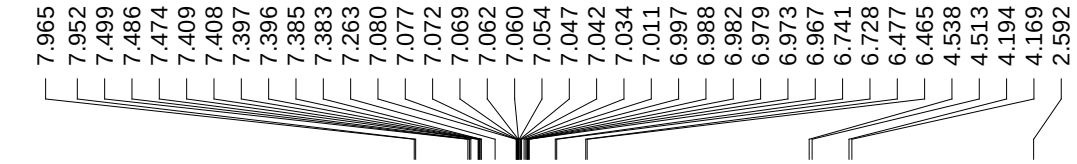


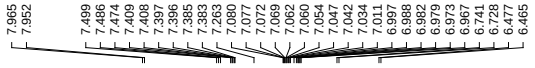
4t

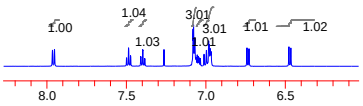
^{19}F NMR (376M, $CDCl_3$)

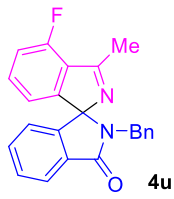






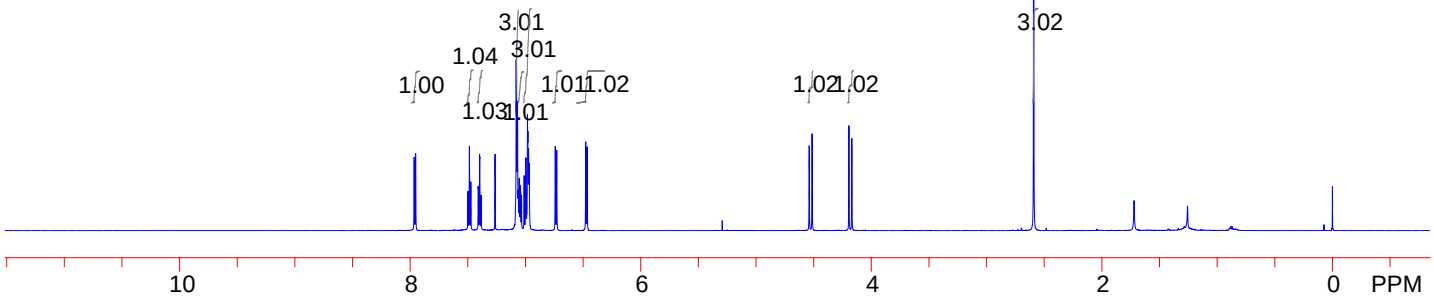


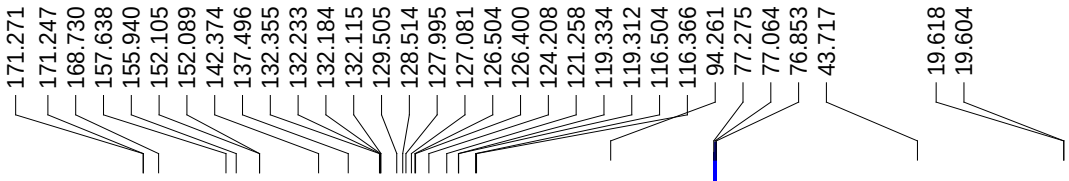


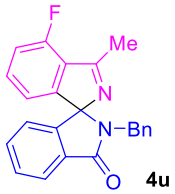


4u

^{1}H NMR (600M, $CDCl_3$)

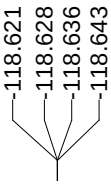


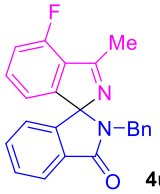




4u

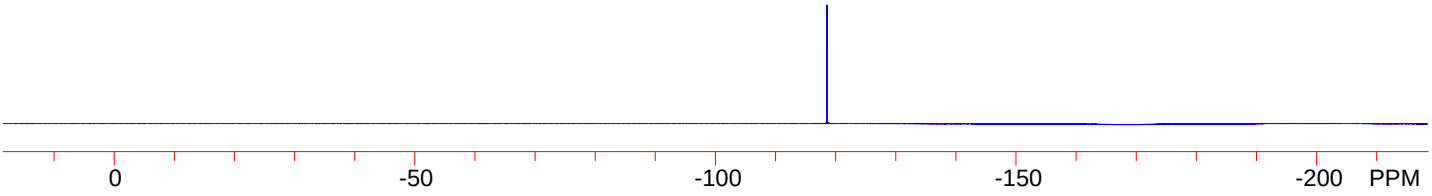
$^{13}C\{^{1}H\}$ NMR (150M, $CDCl_3$)

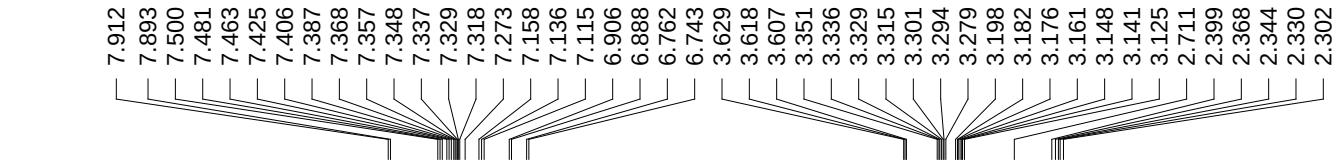


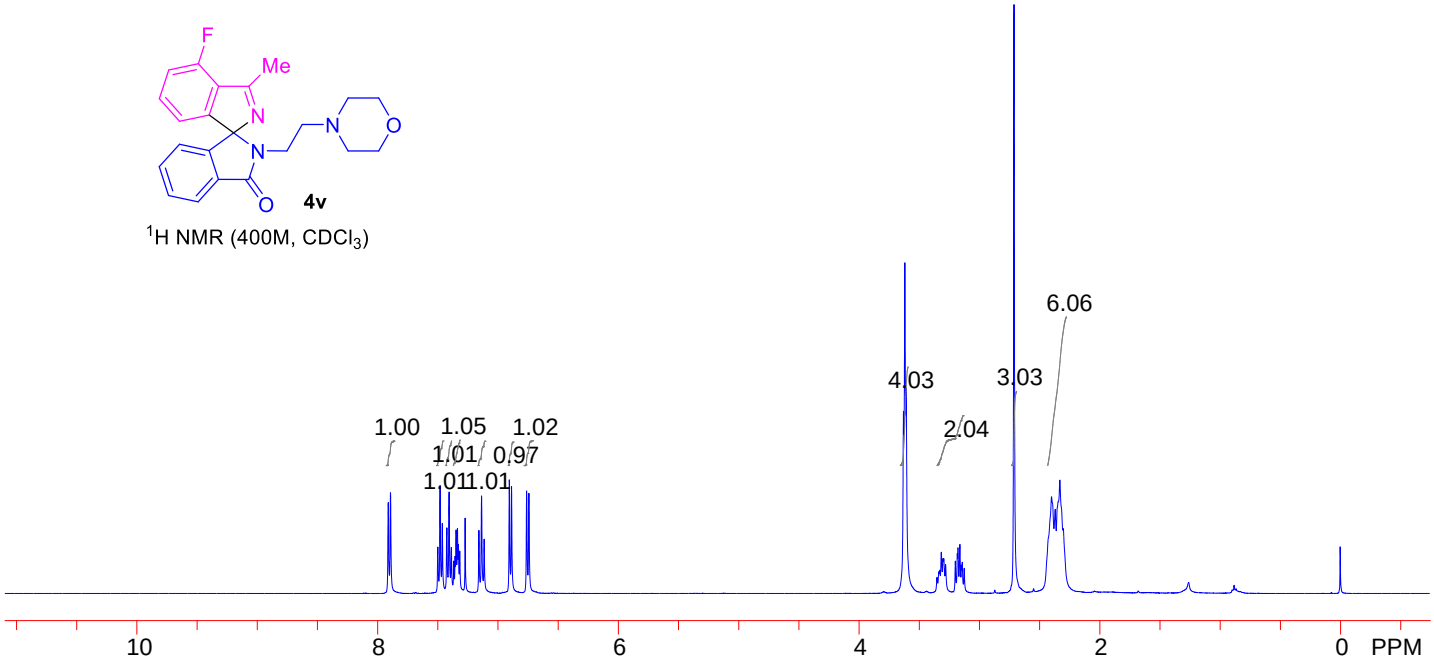


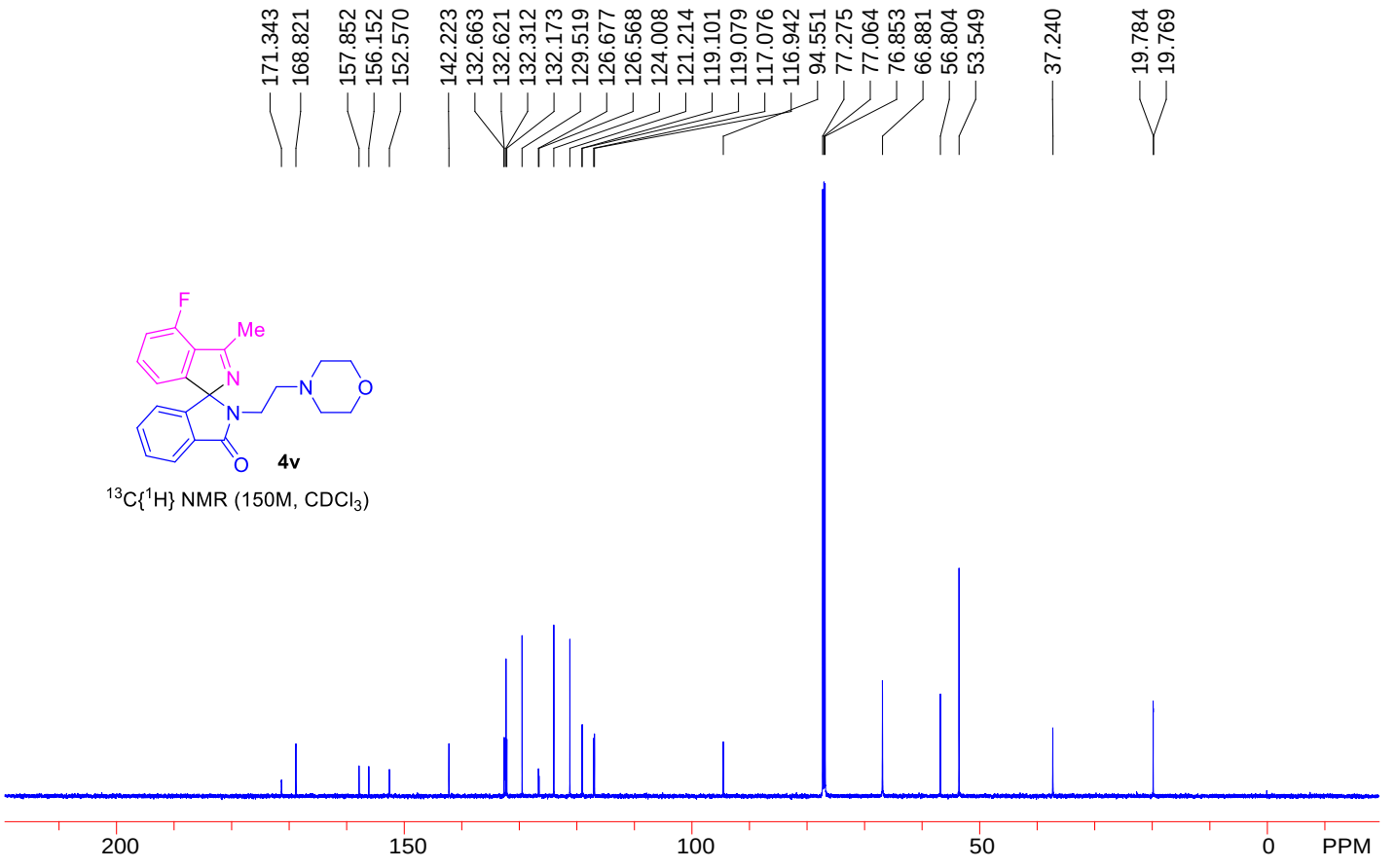
4u

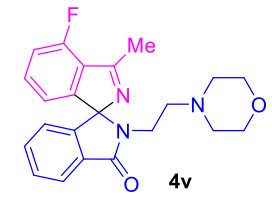
^{19}F NMR (565M, $CDCl_3$)




7.912
7.893
7.500
7.481
7.463
7.425
7.406
7.387
7.368
7.357
7.348
7.337
7.329
7.318
7.273
7.158
7.136
7.115
6.906
6.888
6.762
6.743
3.629
3.618
3.607
3.351
3.336
3.329
3.315
3.301
3.294
3.279
3.198
3.182
3.176
3.161
3.148
3.141
3.125
2.711
2.399
2.368
2.344
2.330
2.302

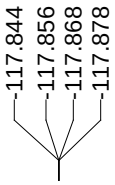

F
Me
N
N
N
O
O
4v
1H NMR (400M, CDCl3)
1.00
1.05
1.02
1.01
0.97
1.01
1.01
4.03
2.04
3.03
6.06
10
8
6
4
2
0
PPM

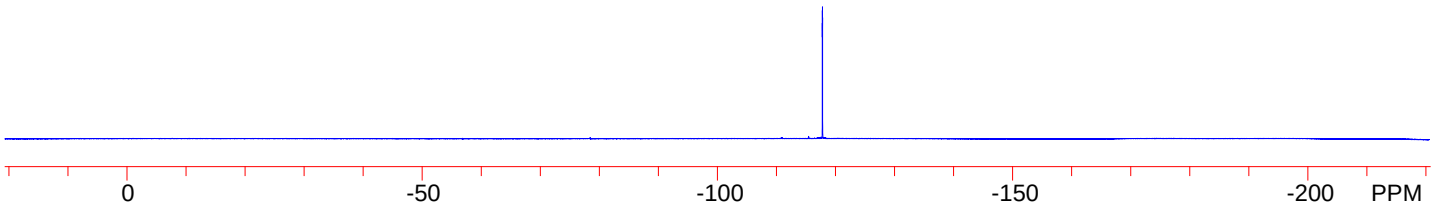

171.343
168.821
157.852
156.152
152.570
142.223
132.663
132.621
132.312
132.173
129.519
126.677
126.568
124.008
121.214
119.101
119.079
117.076
116.942
94.551
77.275
77.064
76.853
66.881
56.804
53.549
37.240
19.784
19.769
F
Me
N
N
N
O
O
4v
13C{1H} NMR (150M, CDCl3)
200
150
100
50
0
PPM

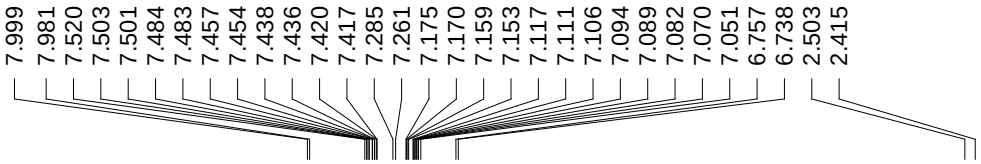


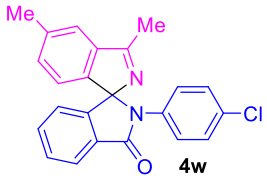
4v

^{19}F NMR (376M, $CDCl_3$)



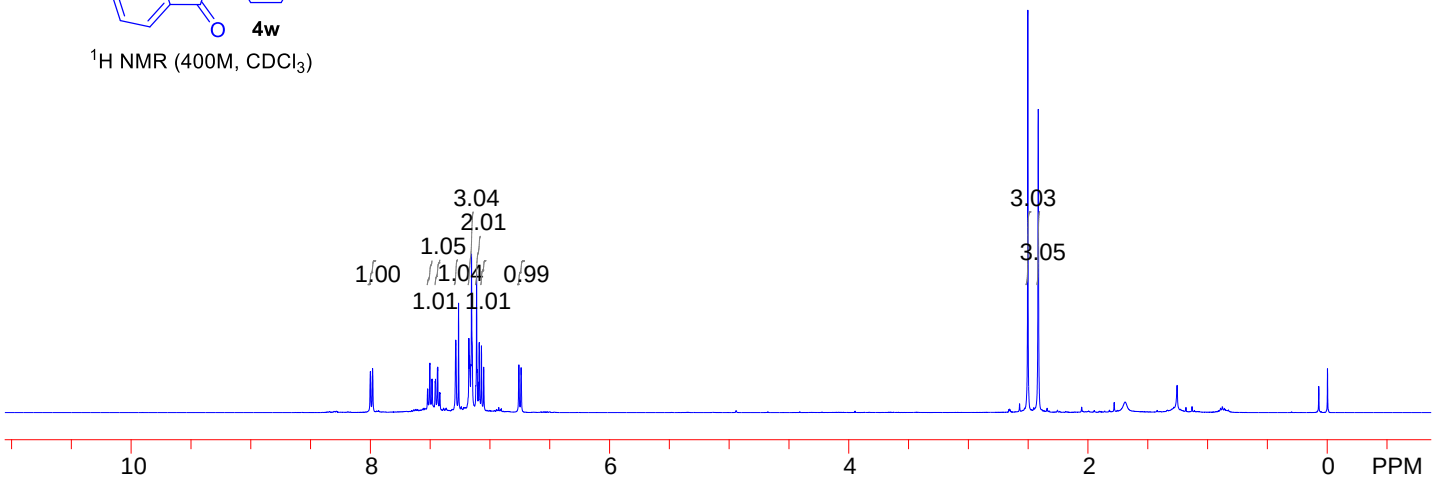


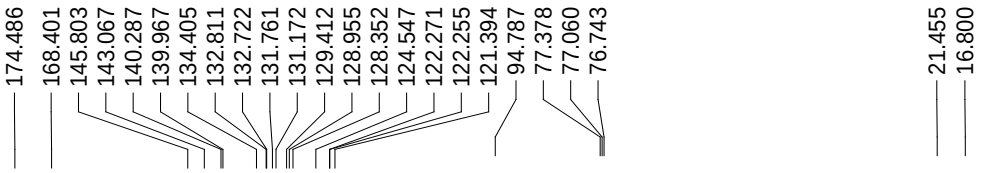


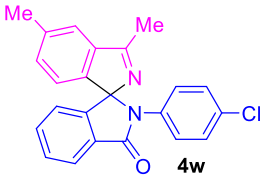


4w

^{1}H NMR (400M, $CDCl_3$)

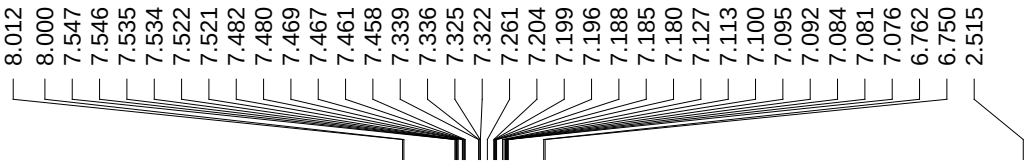


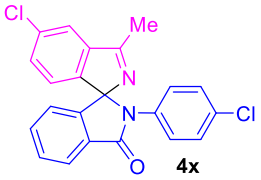




4w

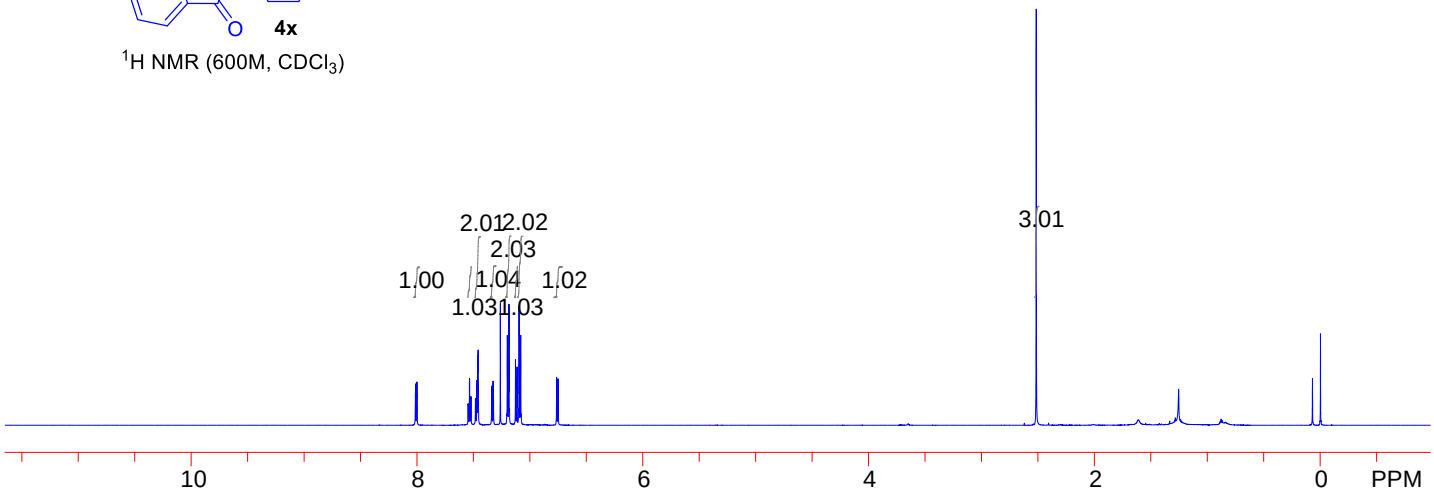
$^{13}C\{^{1}H\}$ NMR (100M, $CDCl_3$)

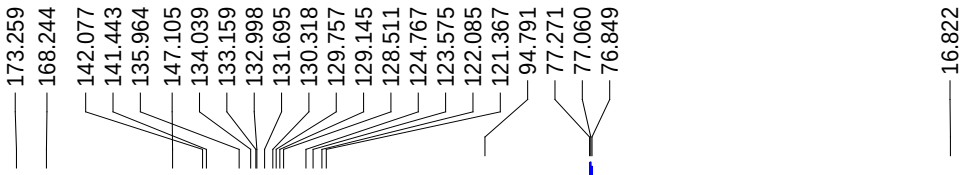


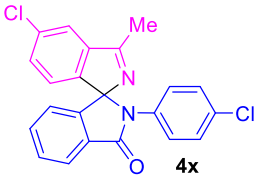


4x

^{1}H NMR (600M, $CDCl_3$)

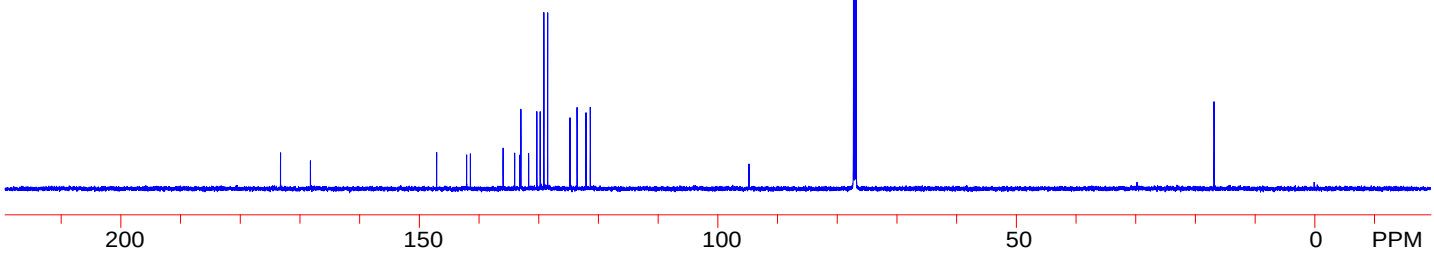


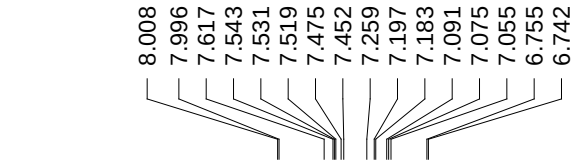


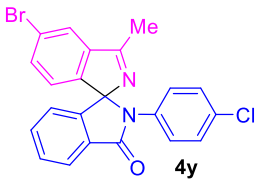


4x

$^{13}C\{^{1}H\}$ NMR (150M, $CDCl_3$)

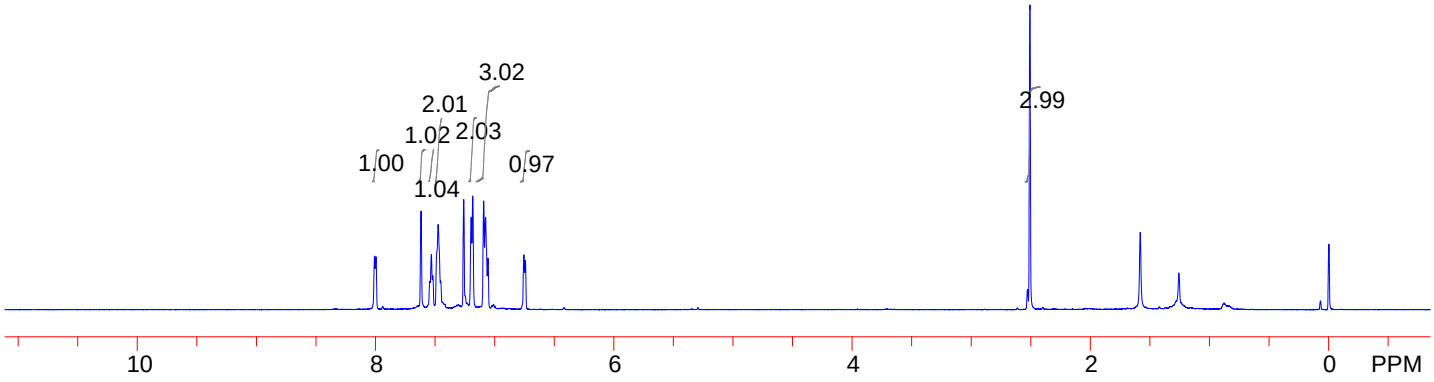


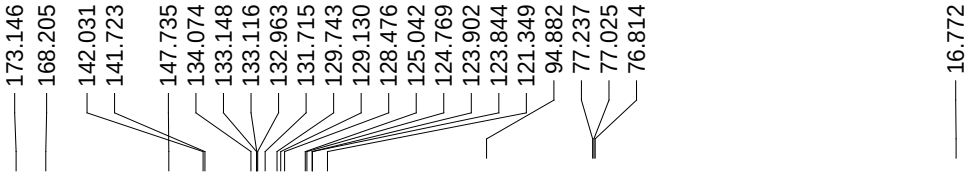


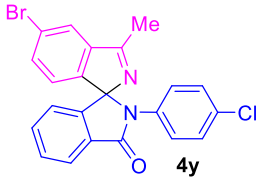


4y

^{1}H NMR (600M, $CDCl_3$)

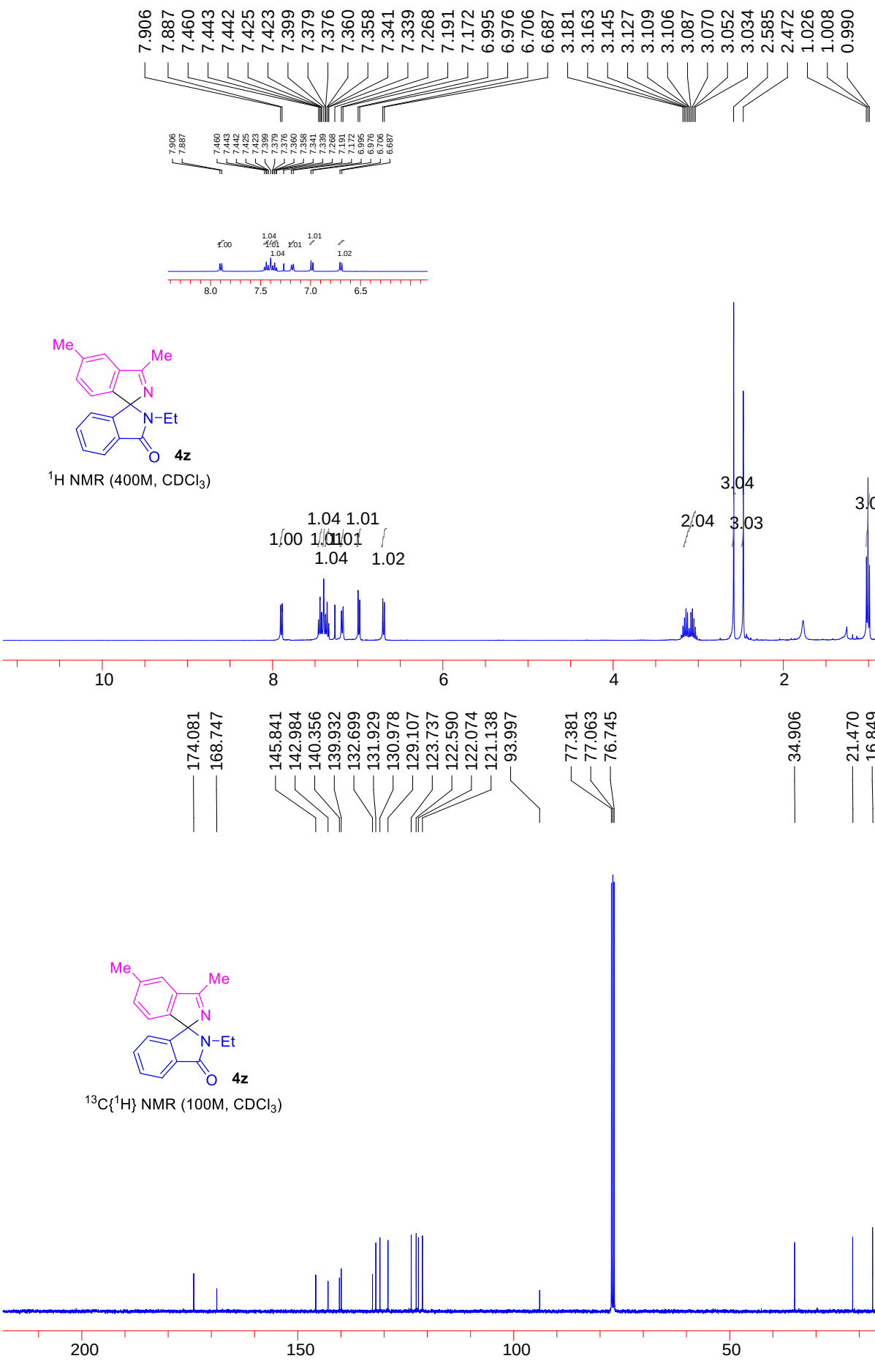


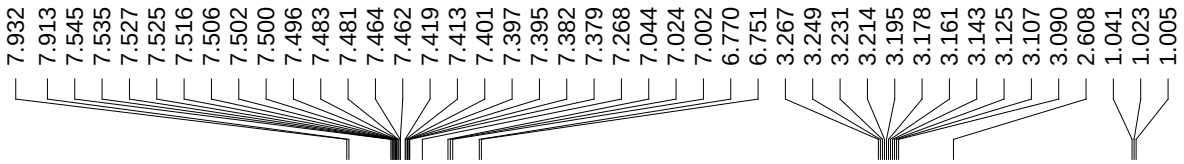


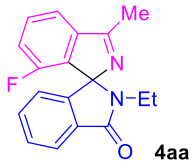


4y

^{13}C{^{1}H} NMR (150M, $CDCl_3$)

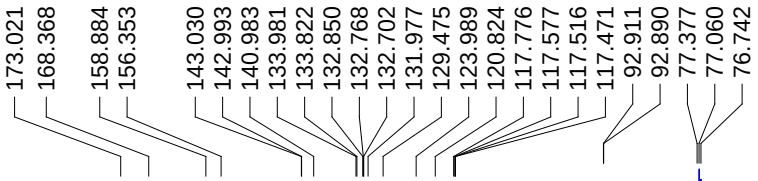

7.906
7.887
7.460
7.443
7.442
7.425
7.423
7.399
7.379
7.376
7.360
7.358
7.341
7.339
7.268
7.191
7.172
6.995
6.976
6.706
6.687
3.181
3.163
3.145
3.127
3.109
3.106
3.087
3.070
3.052
3.034
2.585
2.472
1.026
1.008
0.990
Me
Me
N
N–Et
O
4z
^{1}H NMR (400M, CDCl$_3$)
1.00
1.04
1.01
1.02
2.04
3.03
3.04
3.04
10
8
6
4
2
0
PPM
174.081
168.747
145.841
142.984
140.356
139.932
132.699
131.929
130.978
129.107
123.737
122.590
122.074
121.138
93.997
77.381
77.063
76.745
34.906
21.470
16.849
14.612
Me
Me
N
N–Et
O
4z
^{13}C{^{1}H} NMR (100M, CDCl$_3$)
200
150
100
50
0
PPM

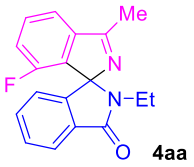




4aa

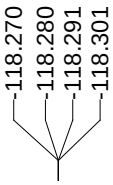
^{1}H NMR (400M, $CDCl_3$)

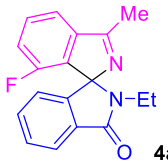




4aa

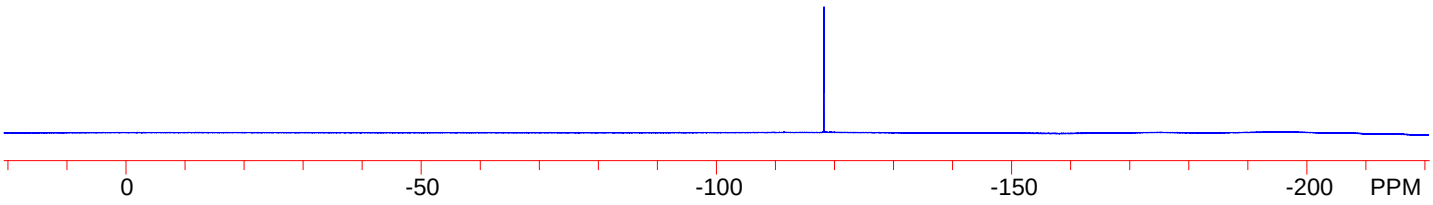
^{13}C{^{1}H} NMR (100M, $CDCl_3$)

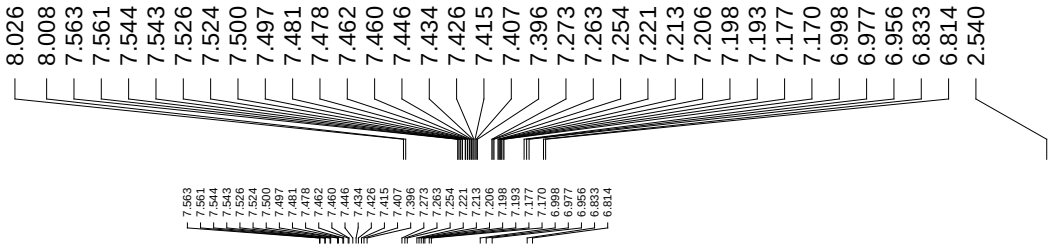


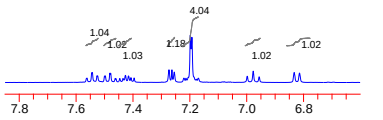


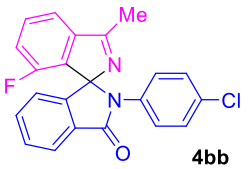
4aa

^{19}F NMR (376M, $CDCl_3$)



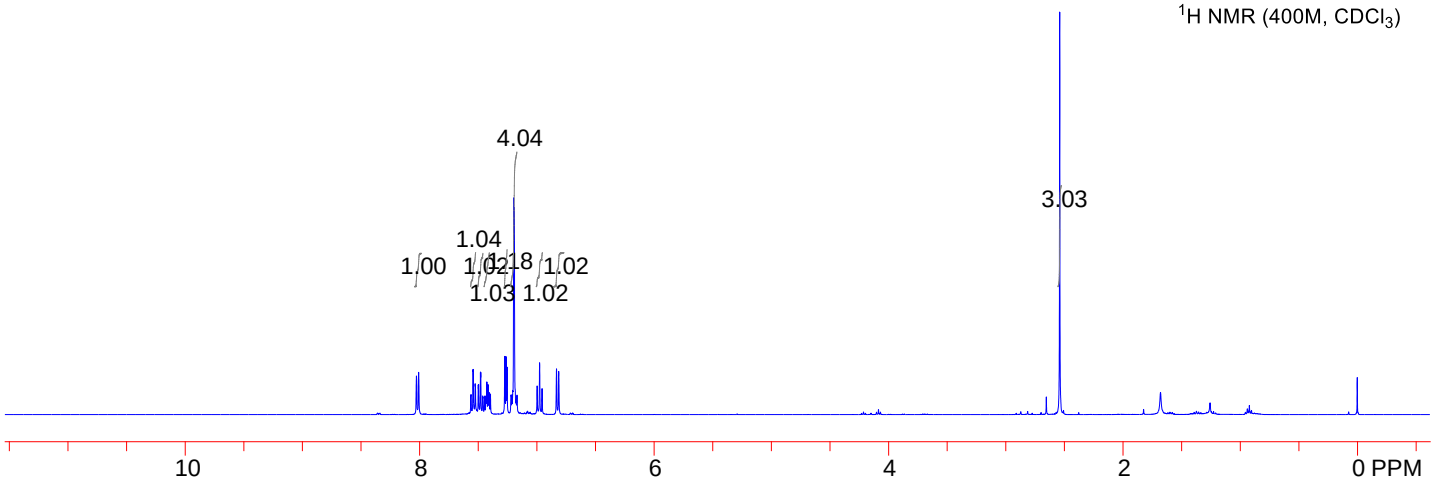


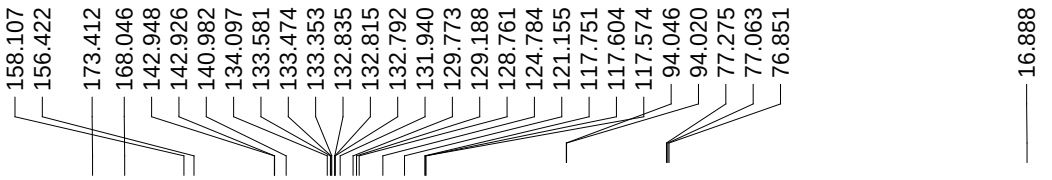


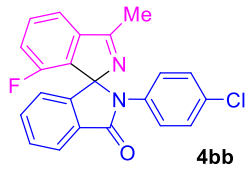


4bb

^{1}H NMR (400M, $CDCl_3$)

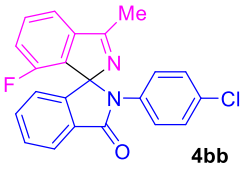






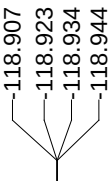
4bb

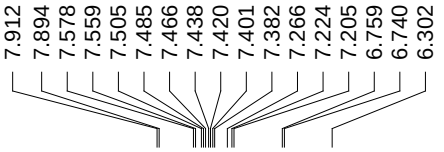
^{13}C{^{1}H} NMR (150M, $CDCl_3$)

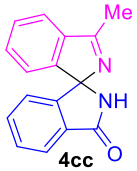


4bb

^{19}F NMR (376M, $CDCl_3$)

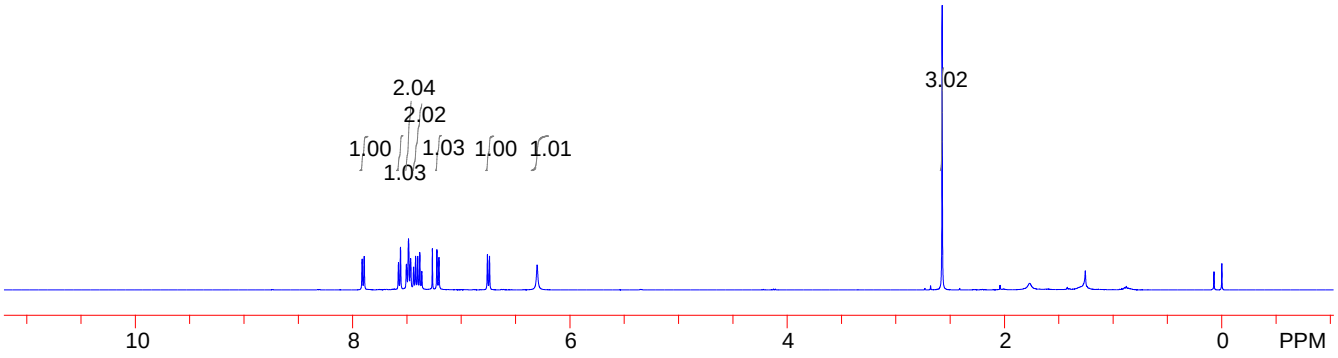


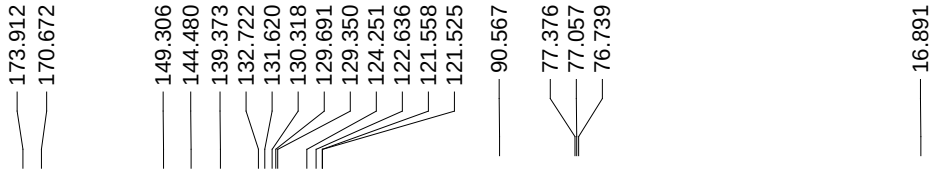


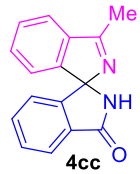


4cc

^{1}H NMR (400M, $CDCl_3$)

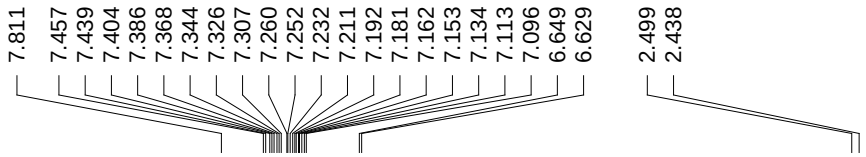


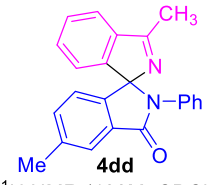




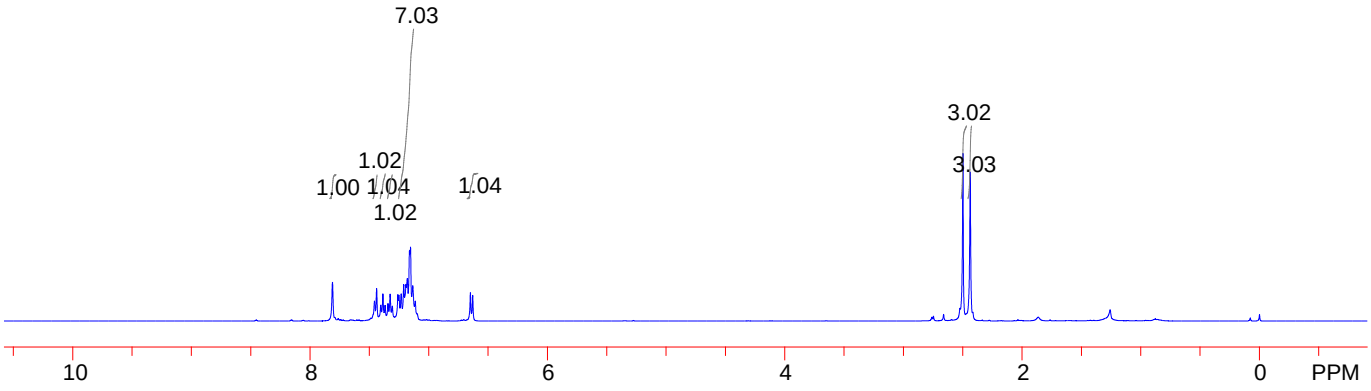
4cc

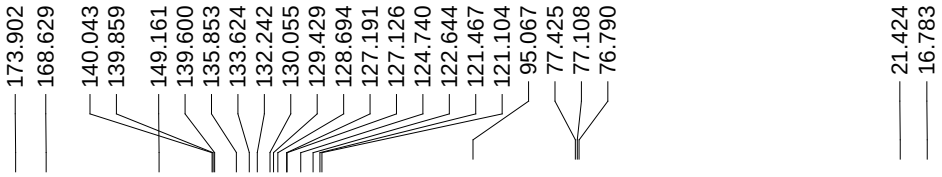
^{13}C{^{1}H} NMR (100M, $CDCl_3$)

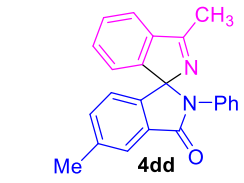




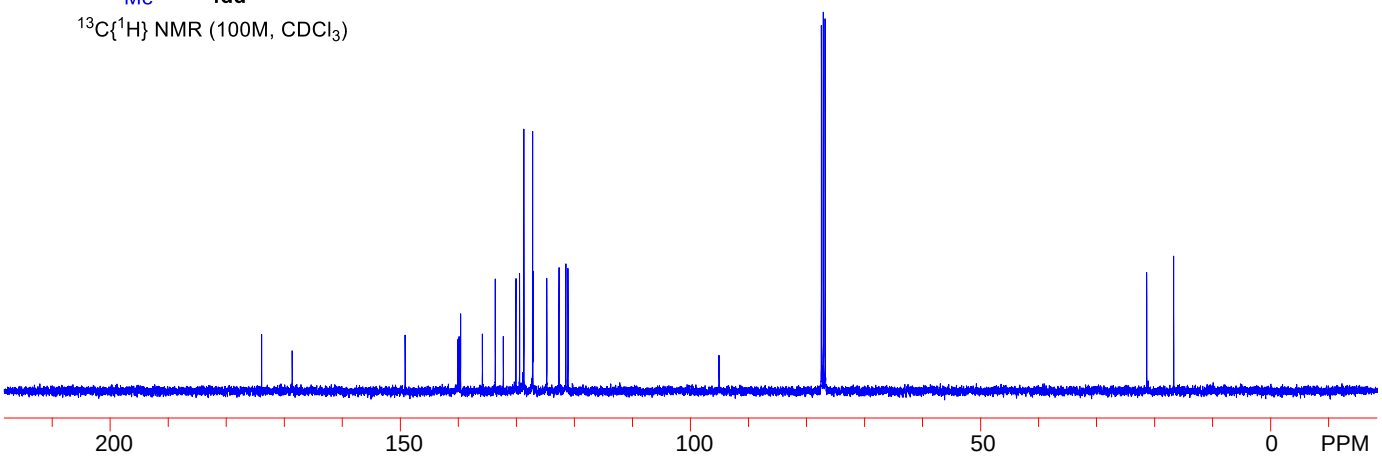
^{1}H NMR (400M, $CDCl_3$)

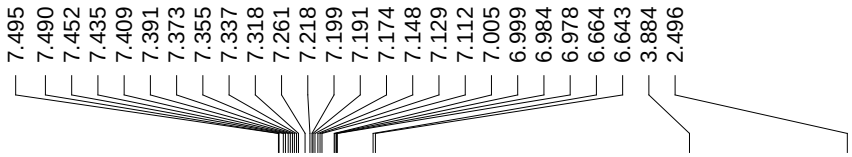


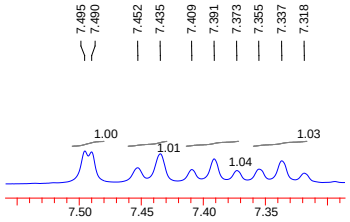


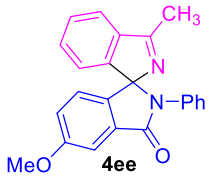


$^{13}C\{^{1}H\}$ NMR (100M, $CDCl_3$)

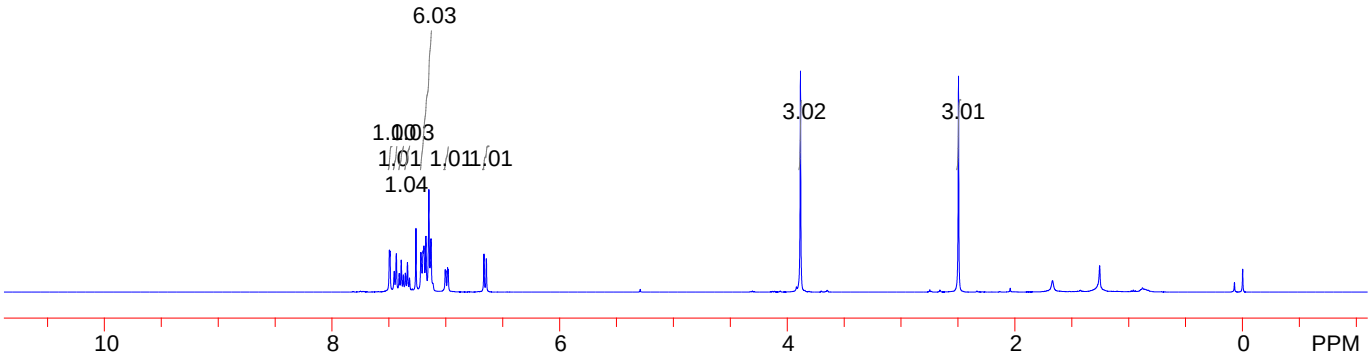


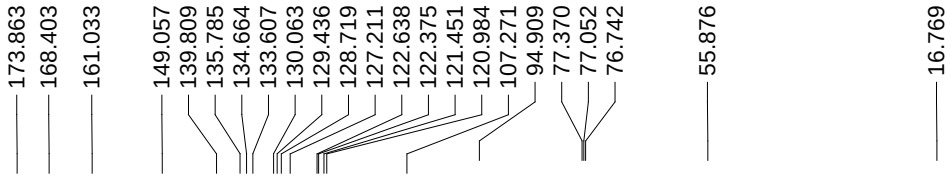


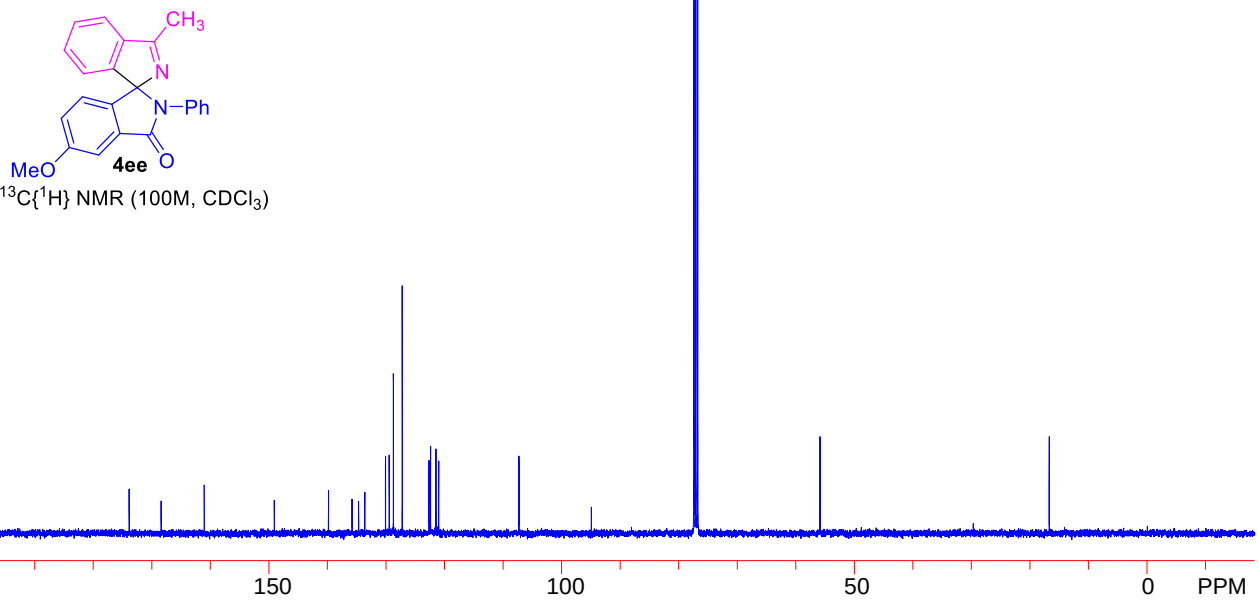




^{1}H NMR (400M, $CDCl_3$)

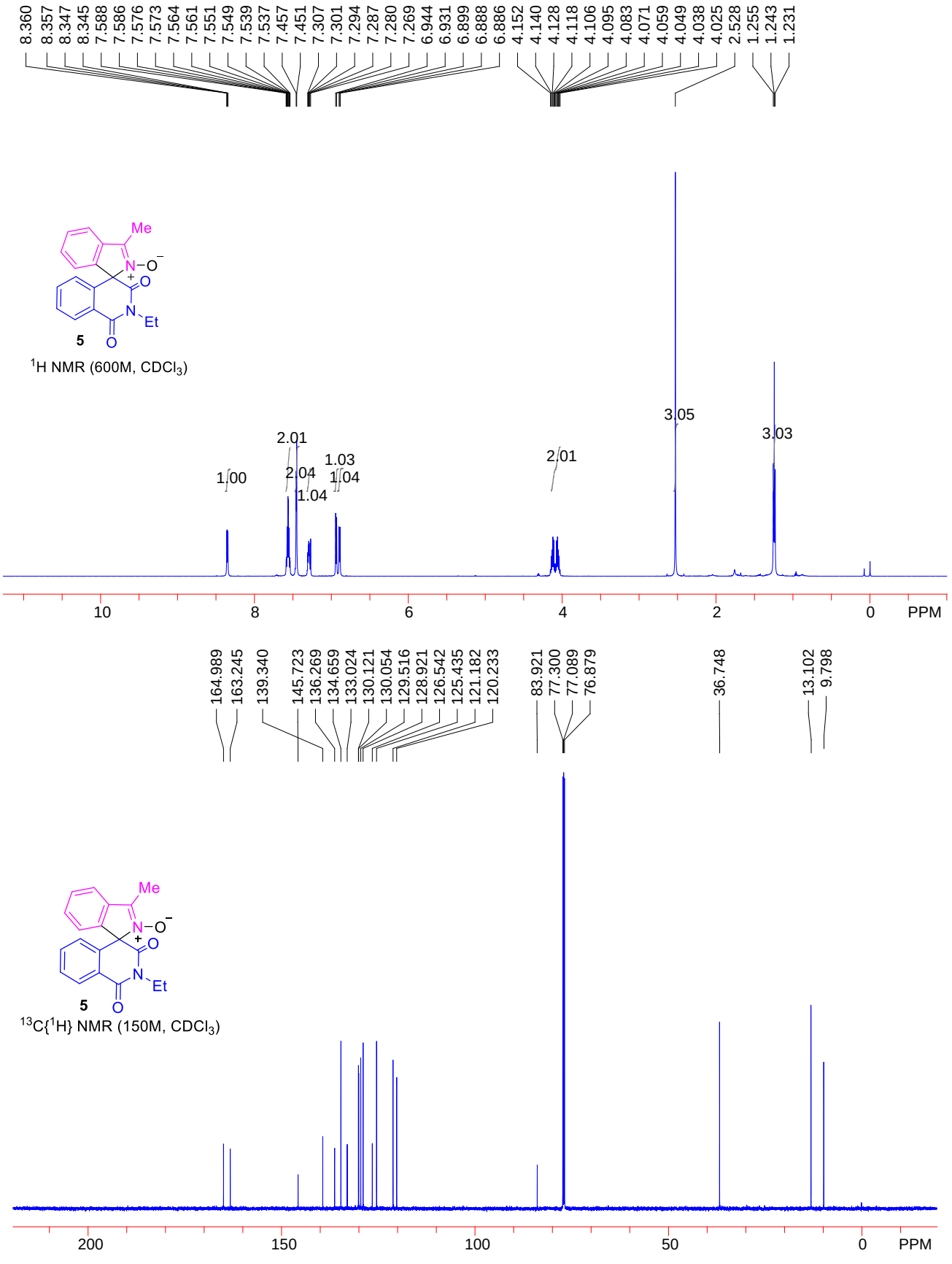


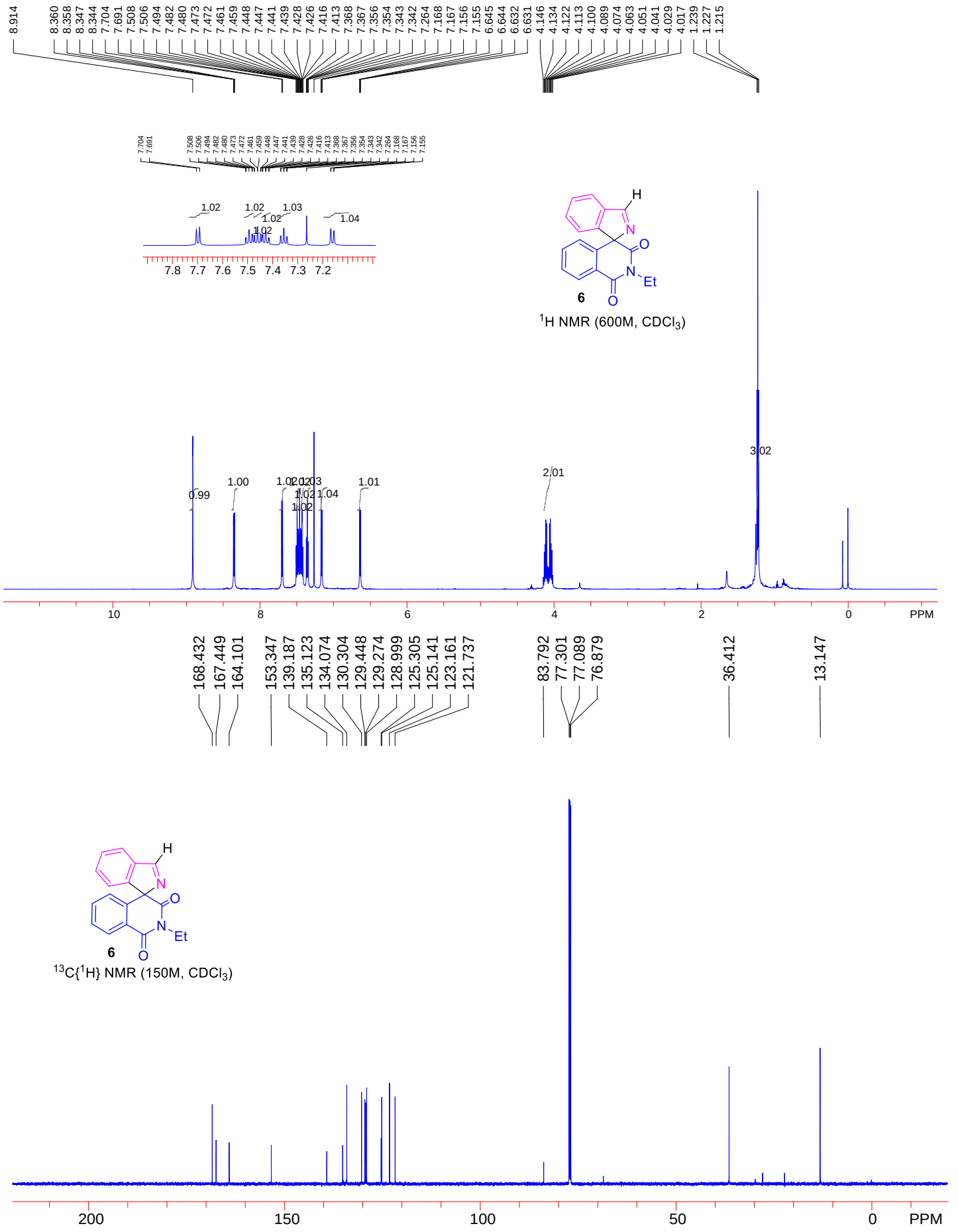


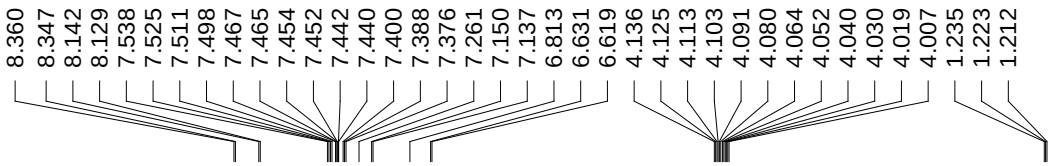


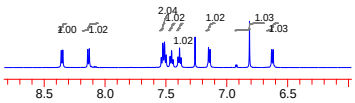
^{13}C{^{1}H} NMR (100M, $CDCl_3$)

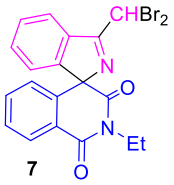
VI. NMR spectra of 5-8




8.914 8.360 8.358 8.347 8.344 7.704 7.691 7.508 7.506 7.494 7.482 7.480 7.473 7.472 7.461 7.459 7.448 7.447 7.441 7.439 7.428 7.426 7.416 7.413 7.368 7.367 7.356 7.354 7.343 7.342 7.264 7.168 7.167 7.156 7.155 6.645 6.644 6.632 6.631 4.146 4.134 4.122 4.113 4.100 4.089 4.074 4.063 4.051 4.041 4.029 4.017 1.239 1.227 1.215
1.02 1.02 1.03 1.02 1.02 1.04
7.8 7.7 7.6 7.5 7.4 7.3 7.2
H N O N Et O
6
1H NMR (600M, CDCl3)
0.99 1.00 1.02 1.02 1.03 1.02 1.02 1.04 1.01 2.01 3.02
10 8 6 4 2 0 PPM
168.432 167.449 164.101 153.347 139.187 135.123 134.074 130.304 129.448 129.274 128.999 125.305 125.141 123.161 121.737 83.792 77.301 77.089 76.879 36.412 13.147
H N O N Et O
6
13C{1H} NMR (150M, CDCl3)
200 150 100 50 0 PPM

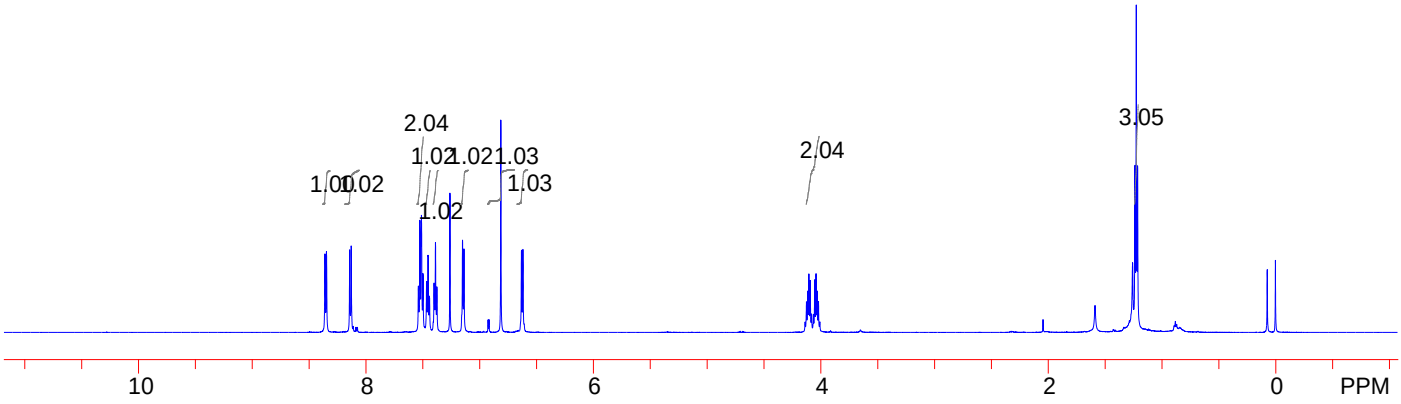


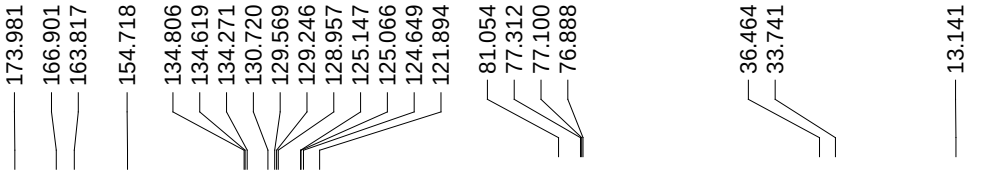


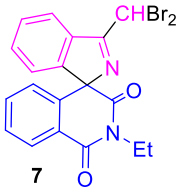


7

^{1}H NMR (600M, $CDCl_3$)

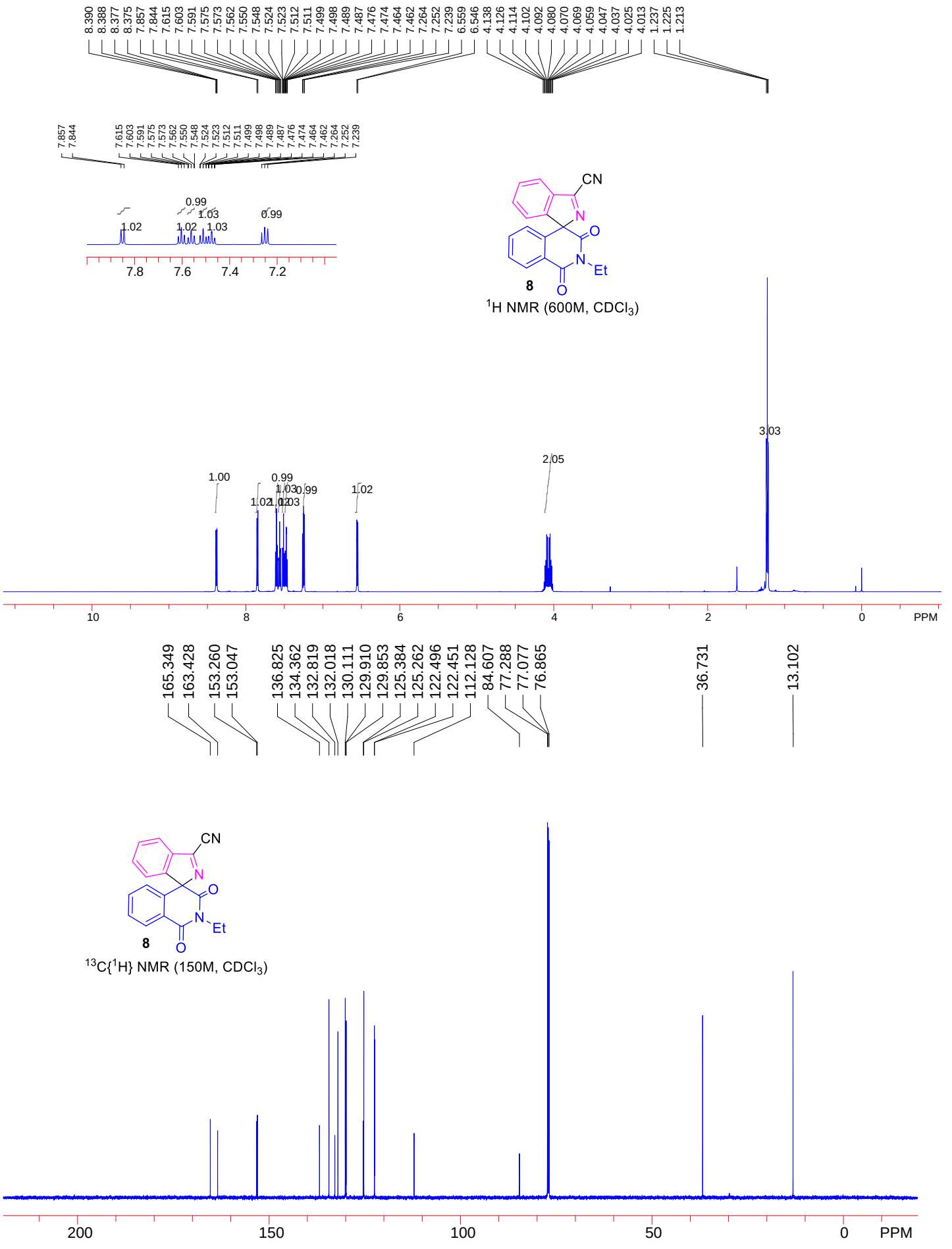






7

$^{13}C\{^{1}H\}$ NMR (150M, $CDCl_3$)


8.390 8.388 8.377 8.375 7.857 7.844 7.615 7.603 7.591 7.575 7.573 7.562 7.550 7.548 7.524 7.523 7.512 7.511 7.499 7.498 7.489 7.487 7.476 7.474 7.464 7.462 7.264 7.252 7.239 6.559 6.546 4.138 4.126 4.114 4.102 4.092 4.080 4.070 4.069 4.059 4.047 4.037 4.025 4.013 1.237 1.225 1.213
0.99 1.03 1.02 1.02 1.03 0.99
7.8 7.6 7.4 7.2
CN N O N Et O
8
1H NMR (600M, CDCl3)
3.03 2.05 1.00 0.99 1.03 1.02 0.99 1.02 1.02 1.02 1.03
10 8 6 4 2 0 PPM
165.349 163.428 153.260 153.047 136.825 134.362 132.819 132.018 130.111 129.910 129.853 125.384 125.262 122.496 122.451 112.128 84.607 77.288 77.077 76.865 36.731 13.102
CN N O N Et O
8
13C{1H} NMR (150M, CDCl3)
200 150 100 50 0 PPM

VII. X-ray crystal structure and data of 3a

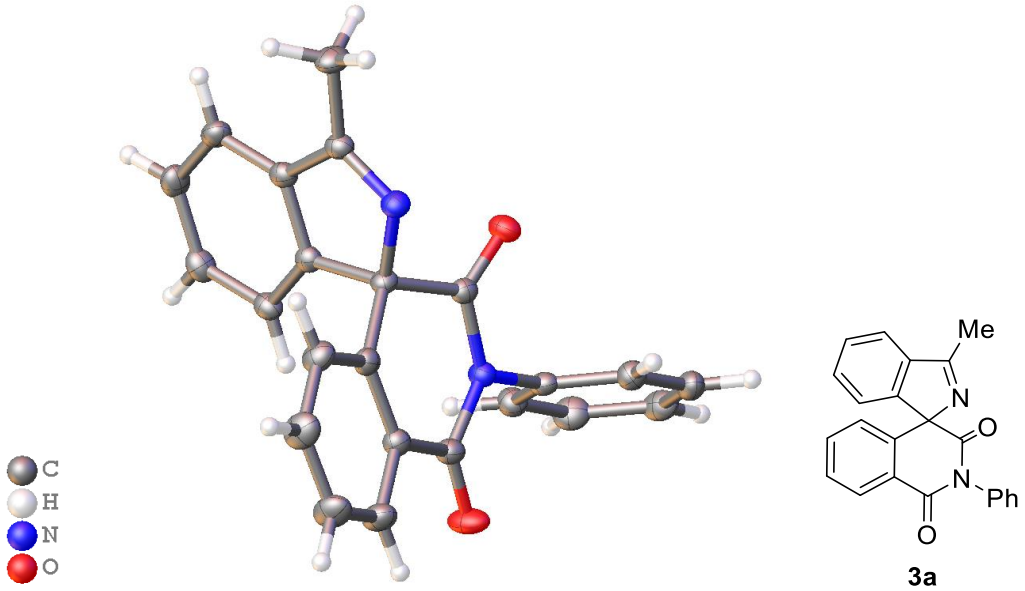


Figure S1. X-ray crystal structure of **3a** with 50% ellipsoid probability

X-ray structure determination. Single crystals suitable for X-ray diffraction were obtained by slow evaporation of the solvent from an ethyl acetate/dichloromethane (4:1) solution of **3a**. Crystal data collection and refinement parameters of **3a** are summarized in Table S1. Intensity data were collected at 150 K on a SuperNova Dual diffractometer using mirror-monochromated Cu Kα radiation, λ = 1.54184 Å. The data were corrected for decay, Lorentz, and polarization effects as well as absorption and beam corrections based on the multi-scan technique. Using Olex2, the structure was solved with the SHELXS structure solution program using Direct Methods and refined with the SHELXL refinement package using Least Squares minimisation. Nonhydrogen atoms were refined with anisotropic displacement parameters. The H-atoms were either located or calculated and subsequently treated with a riding model.

Table S1 Crystallographic data and structure refinement results of **3a**

Empirical formula	$C_{23}H_{16}N_2O_2$
Formula weight	352.38
Temp, K	149.99(10)
Crystal system	monoclinic
Space group	$P2_1/c$
a, Å	8.1641(2)
b, Å	11.1647(2)
c, Å	19.4105(3)
α (°)	90
β (°)	100.089(2)
γ (°)	90
Volume, Å^3	1741.90(6)
Z	4
ρ_{calc}, g cm^{-3}	1.344
λ, Å	1.54184
μ, mm^{-1}	0.696
No. of data collected	7171
No. of unique data	3310
R_{int}	0.0178
Goodness-of-fit on F^2	1.063
R_1, wR_2 (I> =2σ(I))	0.0367, 0.0902
R_1, wR_2 (all data)	0.0409, 0.0935

VIII. X-ray crystal structure and data of 4j

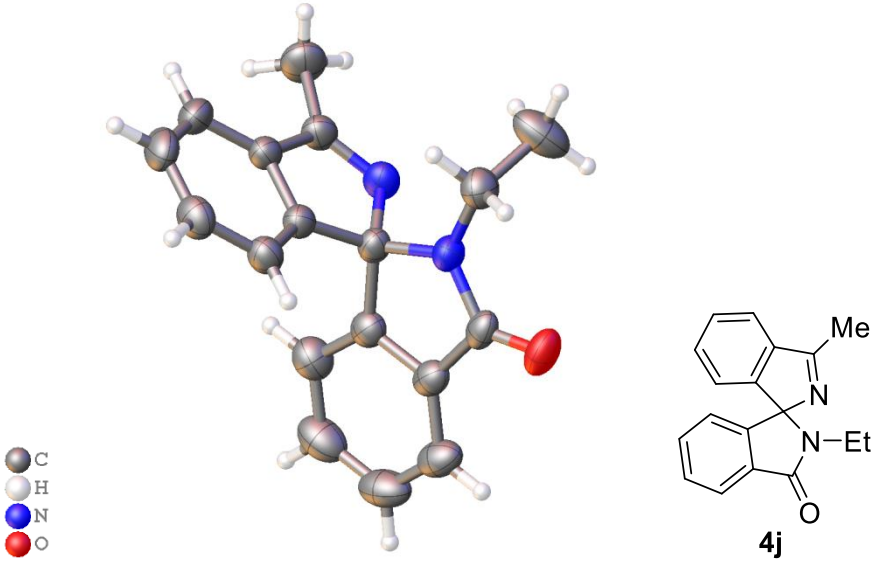


Figure S2. X-ray crystal structure of **4j** with 50% ellipsoid probability

X-ray structure determination. Single crystals suitable for X-ray diffraction were obtained by slow evaporation of the solvent from an ethyl acetate/diethyl ether (1:1) solution of **4j**. Crystal data collection and refinement parameters of **4j** are summarized in Table S2. Intensity data were collected at 293 K on a SuperNova Dual diffractometer using mirror-monochromated Cu Kα radiation, λ = 1.54184 Å. The data were corrected for decay, Lorentz, and polarization effects as well as absorption and beam corrections based on the multi-scan technique. Using Olex2, the structure was solved with the SHELXS structure solution program using Direct Methods and refined with the SHELXL refinement package using Least Squares minimisation. Nonhydrogen atoms were refined with anisotropic displacement parameters. The H-atoms were either located or calculated and subsequently treated with a riding model.

Table S2 Crystallographic data and structure refinement results of **4j**

Empirical formula	$C_{18}H_{16}N_2O$
Formula weight	276.33
Temp, K	293(2)
Crystal system	monoclinic
Space group	$P2_1/n$
a, Å	8.9460(2)
b, Å	14.0474(3)
c, Å	11.9068(3)
α (°)	90
β (°)	103.636(2)
γ (°)	90
Volume, Å^3	1454.13(6)
Z	4
ρ_{calc}, g cm^{-3}	1.262
λ, Å	1.54184
μ, mm^{-1}	0.627
No. of data collected	6325
No. of unique data	2757
R_{int}	0.0190
Goodness-of-fit on F^2	1.061
R_1, wR_2 (I> =2σ(I))	0.0441, 0.1137
R_1, wR_2 (all data)	0.0497, 0.1179

IX. References

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