Electronic Supplementary Material (ESI) for ChemComm.

Supporting Information

Lewis Acid Catalyzed Annulation of Spirocyclic Donor-Acceptor Cyclopropanes with *exo*-Heterocyclic Olefins: Access to Highly Functionalized bis-Spirocyclopentane Oxindole Frameworks

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1. General Experimental.

Unless otherwise indicated, all reactions were conducted with oven or flame-dried glassware and maintaining an inert (under nitrogen or argon) atmosphere. Solvents were dried according to standard procedures. All the reagents/catalysts were purchased commercially and used without any further purification. Reactions were monitored by TLC, using Merck silica gel 60 F 254 plates. The plates were visualized under UV light (254 nm) or by using 10% ethanolic phosphomolybdic acid (PMA) or 1% aqueous $KMnO_4$ or iodine. Flash column chromatography was performed using silica gel (230-400 mesh). ^{1}H, ^{13}C and ^{19}F NMR spectra were recorded on Avance III, Bruker 400 MHz 100 MHz and 376 MHz spectrometers respectively using $CDCl_3$ or DMSO-d_6. ^{1}H NMR chemicals shift are expressed in ppm (δ) relative to δ = 7.26 for $CDCl_3$ and δ = 2.50 for DMSO-d_6. ^{13}C NMR chemical shift are expressed in ppm (δ) relative to δ = 77.16 for $CDCl_3$ and δ = 39.51 for DMSO-d_6 resonance. Coupling constants (*J*) are expressed in Hz. The following abbreviations were used to describe multiplicities of the signals: s = singlet, d = doublet, t = triplet, q = quartet, dq= doublet of quartets, m = multiplet, b = broad. FT-IR experiments were performed on PerkinElmer Spectrum Version 10.03.08. HRMS and Electron Spray Ionization (ESI) (m/z) spectra were recorded on Agilent Technologies 6530 Accurate- Mass Q-TOF LC/MS. The melting points (Mps) were determined using a STUART SMP30 melting point apparatus and are uncorrected.

2. (a) Preparation of starting material: *Spirocyclopropyl oxindoles*

Following Cyclopropyl spirooxindoles (**Figure S1**) were used in this study and were prepared according to the literature procedures as indicated in the parenthesis for each compound. Compounds **1d** was prepared using similar method as described in **Scheme S1**.

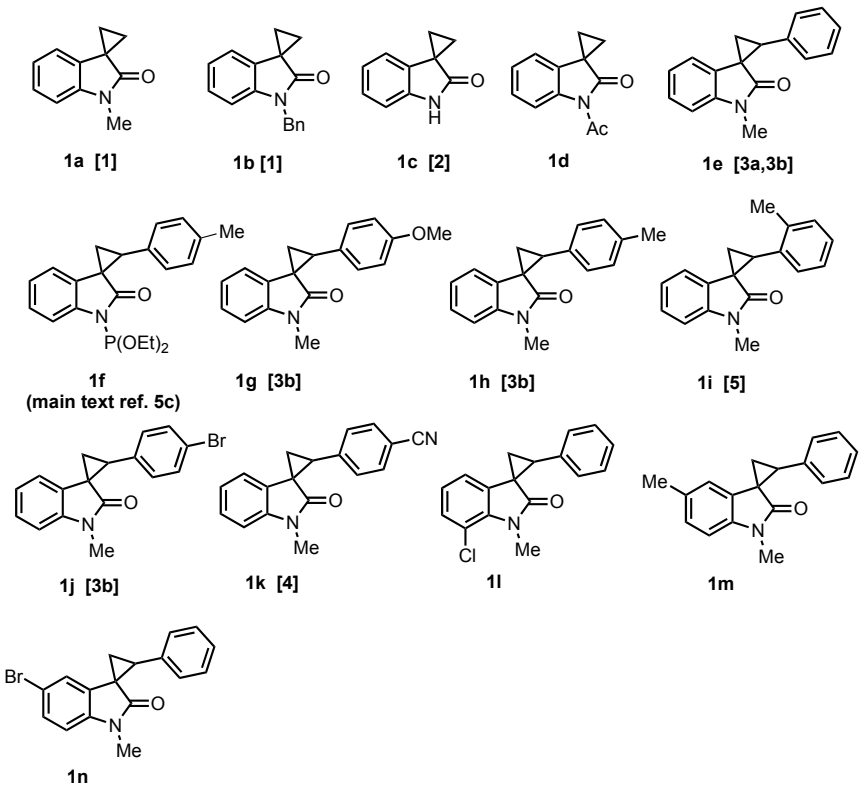
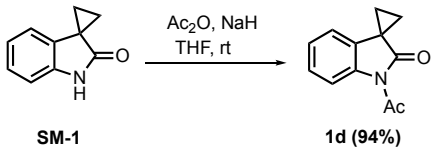
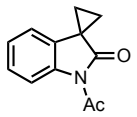


Figure S1. List of spirocyclopropyl oxindoles used in the study.



Scheme S1. Synthetic steps for the preparation of spirocyclopropyl oxindoles **1d**.

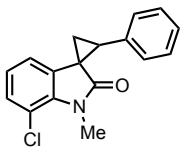
1'-acetylspiro[cyclopropane-1,3'-indolin]-2'-one (1d):



1d

Compound **SM-1** is a known compound and prepared according to the literature procedure [(2)]. To a solution of compound **SM-1** (0.03 g, 0.19 mmol, 1.0 equiv) in dry THF (2 mL) was added sodium hydride (0.007 g, 0.28 mmol, 1.5 equiv) followed by acetic anhydride (0.023 g, 0.23 mmol, 1.2 equiv) at 0 °C. Then the reaction mixture was stirred at room temperature for 3h. Upon completion of the reaction (TLC controlled), water was added to the reaction mixture (2 mL), extracted in EtOAc (10 mL) and dried over anhydrous Na_2SO_4. Solvents were removed under reduced pressure and the crude product was purified by flash silica gel column chromatography (using 1:9 EtOAc: Hexanes as eluent) to obtain compound **1d** as colorless oil in 94% (0.035 g) yield. R_f 0.6 (1:9 EtOAc: Hexanes); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.26 (d, *J* = 8.1 Hz, 1H), 7.28-7.21 (m, 1H), 7.14 (t, *J* = 7.5 Hz, 1H), 6.80 (d, *J* = 7.4 Hz, 1H), 2.67 (s, 3H), 1.81-1.80 (m, 2H), 1.22 (brs, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 178.0, 171.1, 139.8, 129.9, 127.4, 125.1, 118.0, 116.7, 27.9, 26.8, 21.8; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{12}H_{11}NNaO_2$ 224.0687, mass found 224.0679.

7'-chloro-1'-methyl-2-phenylspiro[cyclopropane-1,3'-indolin]-2'-one (1l):

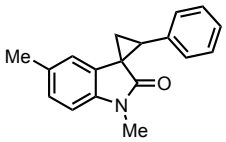


1l

Compound **1l** was prepared following the literature method that describes the synthesis of similar compounds[3,4] and was isolated as mixture of diastereomers in 70% yield (d.r = 2:1). R_f 0.4 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.27-7.20 (m, 5.5H), 7.16-7.11 (2.5H), 7.01 (d, *J* = 8.2 Hz, 1H), 6.92 (t, *J* = 7.7 Hz, 0.5H), 6.78 (d, *J* = 7.4 Hz, 0.5H), 6.52 (t, *J* = 7.8 Hz, 1H), 5.76 (d, *J* = 7.5 Hz, 1H), 3.66 (s, 3H), 3.49 (s, 1.5H), 3.33 (t, *J* = 8.6 Hz, 1H), 3.10 (t, *J* = 8.9 Hz, 0.5H), 2.40 (dd, *J* = 8.6, 5.0 Hz, 0.5H), 2.19 (dd, *J* = 9.1, 4.5 Hz, 1H), 2.02 (dd, *J* = 9.1, 5.1 Hz, 0.5H), 1.95 (dd, *J* = 8.0, 4.6 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 176.8, 174.0, 139.4,

139.4, 134.8, 134.0, 133.7, 130.4, 130.1, 129.4, 129.2 129.0, 128.6, 128.1, 127.7, 127.6, 122.7, 122.2, 119.2, 116.5, 115.5, 115.4, 39.3, 37.1, 33.9, 33.2, 30.2, 29.8, 23.4 (2); HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{17}H_{14}ClNNaO$ 306.0662, mass found 306.0646.

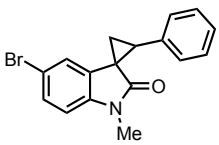
1',5'-dimethyl-2-phenylspiro[cyclopropane-1,3'-indolin]-2'-one (1m):



1m

Compound **1m** was prepared following the similar procedure as compound **1l**, and isolated in 79% yield as a mixture of diastereomers (d.r = 2.5:1). R_f 0.5 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.35-7.27 (m, 4.3H), 7.24-7.20 (m, 2H), 7.14-7.04 (m, 1.3H), 5.99 (s, 1H), 3.35-3.28 (m, 1H), 3.26 (s, 3H), 3.33-3.13 (m, 0.3H), 2.39 (dd, *J* = 8.6, 5.1 Hz, 0.3H), 2.17 (dd, *J* = 9.1, 4.6 Hz, 1H), 2.03 (dd, *J* = 9.0, 5.0 Hz, 0.3H), 1.97 (dd, *J* = 7.9, 4.7 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 175.9, 173.2, 142.9, 142.5, 134.6, 133.9, 133.0, 129.9, 129.8, 129.6, 129.4, 129.3, 128.6, 128.0, 127.8, 127.5, 123.9, 121.5, 114.6, 114.3, 109.2, 109.1, 38.7, 36.4, 33.9, 33.4, 26.8, 26.6, 22.9, 22.8; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{18}H_{17}NNaO$ 286.1208, mass found 286.1195.

5'-bromo-1'-methyl-2-phenylspiro[cyclopropane-1,3'-indolin]-2'-one (1n):



1n

Compound **1n** was prepared following the similar procedure as compound **1l**, and isolated in 77% yield as a mixture of diastereomers (d.r = 3:1). R_f 0.5 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.35-7.27 (m, 4.3H), 7.24-7.20 (m, 2H), 7.14-7.04 (m, 1.3H), 5.99 (s, 1H), 3.35-3.28 (m, 1H), 3.26 (s, 3H), 3.33-3.13 (m, 0.3H), 2.39 (dd, *J* = 8.6, 5.1 Hz, 0.3H), 2.17 (dd, *J* = 9.1, 4.6 Hz, 1H), 2.03 (dd, *J* = 9.0, 5.0 Hz, 0.3H), 1.97 (dd, *J* = 7.9, 4.7 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 175.9, 173.2, 142.9, 142.5, 134.6, 133.9, 133.0, 129.9, 129.8, 129.6, 129.4, 129.3,

128.6, 128.0, 127.8, 127.5, 123.9, 121.5, 114.6, 114.3, 109.2, 109.1, 38.7, 36.4, 33.9, 33.4, 26.8, 26.6, 22.9, 22.8; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{17}H_{14}BrNNaO$ 350.0156, mass found 350.0139.

(b) 2,3-dioxopyrrolidine derivatives:

The following 2,3-dioxopyrrolidine derivatives (**2a**-**2m**) were used in the study (Figure S2). Spirooxindoles **2a-2e**, **2g**-**2k** and **2m** are known compounds and were prepared according to literature methods (references are given in the parenthesis). Compounds **2f** and **2l** were synthesized using similar method.

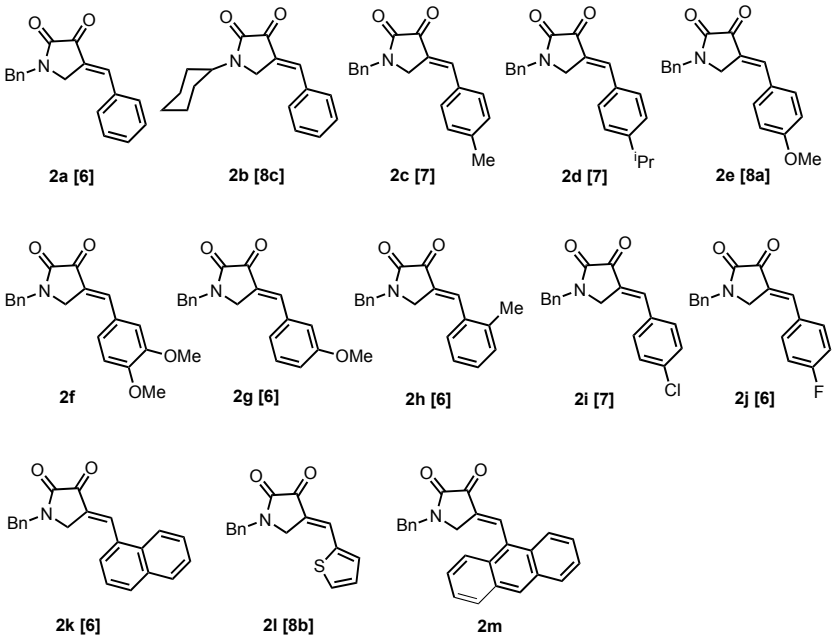
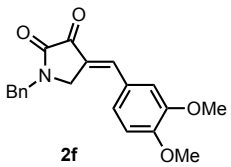


Figure S2. List of 2,3-dioxopyrrolidine derivatives used in the study.

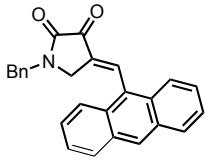
(E)-1-benzyl-4-(3,4-dimethoxybenzylidene)pyrrolidine-2,3-dione (2f):



Compound **2f** was prepared following the literature method that describes the synthesis of similar compounds[7,8] and was isolated in 59% yield as light yellow solid. M.p. 190-192 °C; R_f 0.3 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.54 (s, 1H), 7.29-7.25 (m, 5H), 6.96 (d, J = 8.4 Hz, 1H), 6.84-6.81 (m, 2H), 4.71 (s, 2H),

4.28 (s, 2H), 3.83 (s, 3H), 3.78 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 186.1, 161.1, 152.3, 149.4, 138.5, 134.8, 129.2, 128.6, 128.5, 126.5, 125.6, 122.8, 114.3, 111.6, 56.2, 56.1, 48.1, 46.5; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{20}H_{19}NNaO_4$ 360.1212, mass found 360.1195.

(E)-4-(anthracen-9-ylmethylene)-1-benzylpyrrolidine-2,3-dione (2m):



2m

Compound **2m** was prepared following the literature method that describes the synthesis of similar compounds[7,8] and was isolated in 55% yield as light yellow solid M.p. 210-212 °C; R_f 0.3 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, DMSO-d_6) δ 7.58 (s, 1H), 7.34 (s, 1H), 7.05-6.91 (m, 4H), 6.46-6.44 (m, 4H), 6.14-6.10 (m, 5H), 3.46 (s, 2H); ^{13}C NMR (100 MHz, DMSO-d_6) δ 186.0, 160.8, 135.4, 133.6, 132.8, 130.9, 129.2, 128.9, 128.8, 128.6, 128.2, 127.8, 127.4, 127.1, 126.0, 125.4, 47.0, 45.8; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{26}H_{19}NNaO_2$ 400.1313, mass found 400.1327.

(c) Alkylidene pyrazolones derivatives:

The following substituted alkylidene pyrazolones (**4a**-**4l**) were used in the study (Figure S3) and synthesized according to literature methods (references are given in the parenthesis).

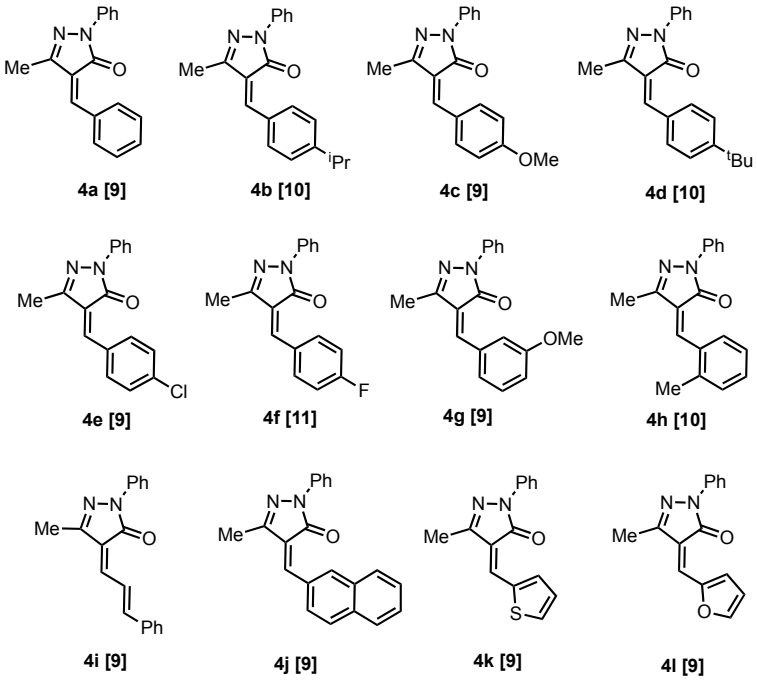
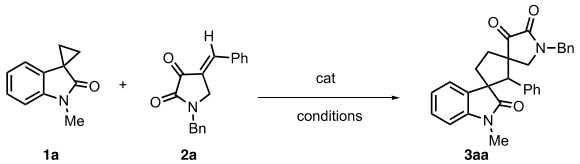


Figure S3. List of substituted alkylidene pyrazolones used in the study.

3. Optimization studies.



Entry	**1a** [equiv]	**2a** [equiv]	Cat.	[*mol%*]	Solvent	Conc[M]	Temp	Yield[%]	d.r
1	1.5	1.0	$Sc(OTf)_3$	20	DCM	0.2	50	0	n.a
2	1.5	1.0	$In(OTf)_3$	20	DCM	0.2	50	0	n.a
3	1.5	1.0	$Mg(OTf)_2$	20	THF	0.2	85	0	n.a
4	1.5	1.0	$MgBr_2$	20	THF	0.2	85	55	20:1
5	**1.5**	**1.0**	**MgI_2**	**20**	**THF**	**0.2**	**85**	**96**	**8:1:1**
6	1.0	1.0	MgI_2	20	THF	0.2	85	55	n.d
7	1.5	1.0	MgI_2	10	THF	0.2	85	58	12:3:2:1
8	1.5	1.0	MgI_2	20	1,4-dioxane	0.2	85	50	7:1:1
9	1.5	1.0	MgI_2	20	THF	0.1	85	46	8:1:1
10	1.5	1.0	MgI_2	20	THF	0.2	rt	0	n.a

d.r= diastereomeric ratio, DCM = dichloromethane, THF = tetrahydrofuran, n.a= not applicable, n.d = not determined, rt= room temperature.

Note: Following control experiments were performed in the study; (a) when only **1a** was subjected to the reaction conditions used in entry 5, no significant side reaction or decomposition of **1a** was observed by TLC and NMR analysis. (b) when compound **2a** was subjected to the optimized reaction conditions used in entry 5, the substrate was decomposed to intractable product mixture over the period of 8 hours. No dimer mass was detected by MS analysis. This mixture was formed at the TLC base line in 4:1 EtOAc-hexane and formation of the same was observed during substrate scope investigation.
For entries 1-4, No trace of desired product was detected. **1a** was found completely unreacted in the reaction mixture and was recovered, however **2a** was completely decomposed in these cases; For entry 7, reaction was run for 24 h; For entry 10, both the starting materials were recovered from the reaction mixture.

The following olefins were initially screened for the reaction with **1a**.

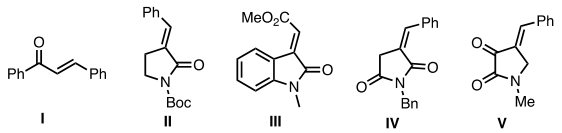
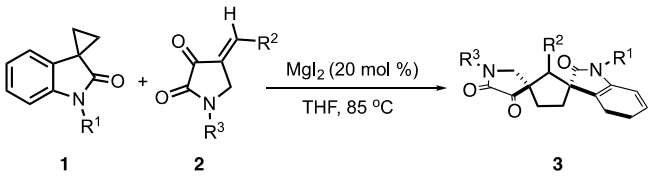


Figure S4. List of olefins screened in the study.

4. General procedure for the synthesis of dispiro-2,3-dioxopyrrolidine[cyclopentane]oxindole (3).



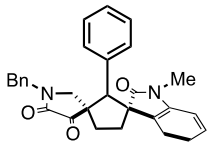
1 2 3

In a 10 ml sealed tube, oxindole-activated spiro-DAC (**1**; 1.5 equiv) and 2,3-dioxopyrrolidine derivative (**2**; 1.0 equiv) were dissolved in dry THF (0.2 M) under argon. MgI_2 (20 mol%) was then added to this reaction mixture and it was heated to 85°C. Progress of the reaction was monitored by thin layer chromatography (TLC) until disappearance of the starting materials (ca. 12-15 h) was observed.

The reaction was then diluted with EtOAc (2 mL), washed with water (2 mL) and brine (2 mL) and the organic layer was dried over anhydrous Na_2SO_4. Solvents were removed under reduced pressure and the crude product was purified by flash silica gel column chromatography to obtain the bis-spirocyclopentane oxindole derivatives (**3**).

4. Characterization of dispiro-2,3-dioxopyrrolidine[cyclopentane]oxindole derivatives (3).

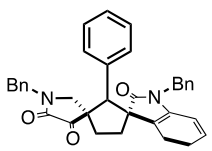
1''-benzyl-1-methyl-2'-phenyldispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (3aa):



3aa

Following the general procedure, reaction between oxindole-activated spiro-DAC **1a** (0.047 g, 0.27, 1.5 equiv) and 2,3-dioxopyrrolidine derivative **2a** (0.05 g, 0.18 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **3aa**, which was purified by silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title compound as colorless oil in 68% (0.055 g, major diastereomer) yield. R_f 0.4 (2:3 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.32 (d, *J* = 7.4 Hz, 1H), 7.22-7.15 (m, 4H), 7.08-6.95 (m, 6H), 6.78-6.71 (m, 3H), 4.50 (d, *J* = 14.6 Hz, 1H), 4.38 (d, *J* = 14.6 Hz, 1H), 4.19 (s, 1H), 3.90 (d, *J* = 12.2 Hz, 1H), 3.65 (d, *J* = 12.2 Hz, 1H), 3.18 (s, 3H), 2.57- 2.49 (m, 1H), 2.-2.29 (m, 2H), 2.20-2.14 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 205.33, 179.46, 158.99, 143.12, 134.51, 134.22, 130.25, 128.94, 128.77, 128.50, 128.28, 128.05, 127.95, 127.92, 123.29, 122.19, 108.23, 62.23, 58.77, 53.94, 48.42, 37.95, 37.57, 26.23; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{29}H_{27}N_2O_3$ 451.2022, mass found 451.2011.

1,1''-dibenzyl-2'-phenyldispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (3ba):

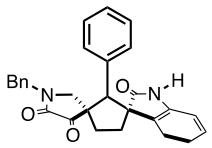


3ba

Following the general procedure, reaction between oxindole-activated spiro-DAC **1b** (0.067 g, 0.27 mmol, 1.5 equiv) and 2,3-dioxopyrrolidine derivative **2a** (0.05 g, 0.18 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **3ba,** which was purified by silica gel column chromatography (2:3 EtOAc:Hexane as eluent) to give

the title compound as colorless oil in 65% (0.054 g, major diastereomer) yield. Note: This compound can be isolated in 62 % yield when the reaction is performed with 1g of **2a**. R_f 0.3 (2:3 EtOAc:Hex); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.36 (d, *J* = 7.2 Hz, 1H), 7.25-7.22 (m, 6H), 7.15-7.06 (m, 4H), 7.02-6.94 (m, 5H), 6.65 (d, *J* = 7.7 Hz, 1H), 6.82-6.81 (m, 2H), 5.01-4.97 (m, 1H), 4.78 (d, *J* = 15.7 Hz, 1H), 4.55 (d, *J* = 14.6 Hz, 1H), 4.37 (d, *J* = 14.6 Hz, 1H), 4.23 (s, 1H), 3.96 (d, *J* = 12.2 Hz, 1H), 3.68 (d, *J* = 12.2 Hz, 1H), 2.61-2.53 (m, 1H), 2.47-2.33 (m, 2H), 2.26-2.21 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 205.1, 179.3, 158.9, 142.4, 135.4, 134.4, 134.1, 130.4, 128.9, 128.8, 128.7, 128.6, 128.4, 128.0, 127.9 (2), 127.7, 127.3, 123.3, 122.3, 109.1, 61.9, 58.9, 53.9, 53.8, 48.3, 43.7, 37.9, 37.6; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{35}H_{31}N_2O_3$ 527.2335, mass found 527.2314.

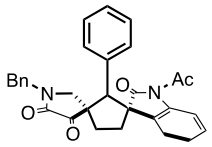
1''-benzyl-2'-phenyldispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (3ca):



3ca

Following the general procedure, reaction between oxindole-activated spiro-DAC **1c** (0.043 g, 0.27 mmol, 1.5 equiv) and 2,3-dioxopyrrolidine derivative **2a** (0.05 g, 0.18 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **3ca**, which was purified by silica gel column chromatography (1:1 EtOAc:Hexane as eluent) to give the title compound as colorless oil in 38% (0.03 g, major diastereomer) yield. R_f 0.03 (2:3 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.81 (s, 1H), 7.31 (d, *J* = 7.5 Hz, 1H), 7.22-7.11 (m, 3H), 7.14-7.05 (m, 2H), 7.01-6.99 (m, 3H), 6.95 (d, *J* = 6.3 Hz, 2H), 6.88 (d, *J* = 7.6 Hz, 2H), 6.75 (d, *J* = 7.7 Hz, 1H), 4.51- 4.40 (m, 2H), 4.20 (s, 1H), 3.88 (d, *J* = 12.2 Hz, 1H), 3.61 (d, *J* = 12.2 Hz, 1H), 2.56-2.40 (m, 2H), 2.38-2.21 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 205.4, 181.3, 159.1, 140.1, 134.5, 134.2, 130.8, 129.0, 128.9, 128.5, 128.3, 128.1, 128.0, 123.2, 122.7, 109.7, 62.1, 59.0, 53.9, 53.8, 48.1, 38.0, 37.5; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{28}H_{24}N_2NaO_3$ 459.1685, mass found 459.1665.

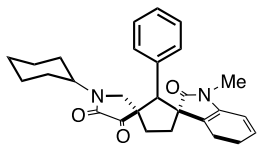
1-acetyl-1''-benzyl-2',4'-diphenyldispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (3da):



3da

Following the general procedure, reaction between oxindole-activated spiro-DAC **1d** (0.054 g, 0.27 mmol, 1.5 equiv) and 2,3-dioxopyrrolidine derivative **2a** (0.05 g, 0.18 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **3da**, which was purified by silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title compound as colorless oil in 36% (0.031 g, major diastereomer) yield. R_f 0.6 (2:3 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.08 (d, *J* = 7.8 Hz, 1H), 7.44-7.42 (m, 1H), 7.28-7.27 (m, 2H), 7.25-7.19 (m, 3H), 7.11 (t, *J* = 7.2 Hz, 1H), 7.06-7.02 (m, 4H), 6.71 (d, *J* = 7.5 Hz, 2H), 4.59 (d, *J* = 14.6 Hz, 1H), 4.47 (d, *J* = 14.6 Hz, 1H), 4.21 (s, 1H), 3.87 (d, *J* = 12.0 Hz, 1H), 3.63 (d, *J* = 12.0 Hz, 1H), 2.92 (d, *J* = 29.0 Hz, 1H), 2.61 (s, 3H), 2.51 – 2.44 (m, 2H), 2.38-2.26 (m, 2H) ^{13}C NMR (100 MHz, $CDCl_3$) δ 204.4, 180.8, 179.6, 158.9, 139.5, 134.5, 137.8, 129.7, 129.1, 129.1, 129.0, 128.3 (2), 128.2, 128.1, 126.0, 122.0, 116.6, 62.9, 59.6, 53.8, 53.6, 48.5, 38.6, 38.2, 26.9; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{30}H_{27}N_2O_4$ 479.1971, mass found 479.1964.

1''-cyclohexyl-1-methyl-2'-phenyldispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (3ab):

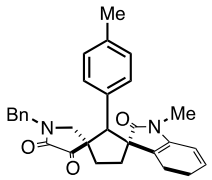


3ab

Following the general procedure, reaction between oxindole-activated spiro-DAC **1a** (0.049 g, 0.29 mmol, 1.5 equiv) and 2,3-dioxopyrrolidine derivative **2b** (0.05 g, 0.19 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **3ab**, which purified by silica gel column chromatography (3:7 EtOAc:Hexane as eluent) to give the title compound as colorless oil in 65% (0.053 g) yield. R_f 0.4 (1:1 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.31 (d, *J* = 7.4 Hz, 1H), 7.19 (t, *J* = 7.7 Hz, 1H), 7.07-7.06 (m, 3H), 7.02 (t, *J* = 7.5 Hz, 1H), 6.83-6.81 (m, 2H), 6.75 (d, *J*

= 7.8 Hz, 1H), 4.13 (s, 1H), 3.99-3.91 (m, 2H), 3.63 (d, *J* = 12.2 Hz, 1H), 3.25 (s, 3H), 2.62-2.54 (m, 1H), 2.48-2.43 (m, 2H), 2.23-2.18 (m, 1H), 1.79-1.76 (m, 1H), 1.69-1.59 (m, 3H), 1.47-1.40 (m, 2H), 1.07-0.91 (m, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 206.4, 179.6, 158.6, 143.1, 134.3, 130.2, 128.8, 128.5, 128.3, 127.9, 123.3, 120.2, 108.3, 62.8, 58.8, 53.8, 52.4, 50.1, 37.9, 37.0, 29.6, 29.0, 26.3, 25.3, 25.2, 25.2; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{28}H_{31}N_2O_3$ 443.2335, mass found 443.2325.

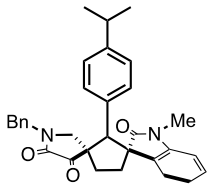
1''-benzyl-1-methyl-2'-(p-tolyl)dispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (3ac):



3ac

Following the general procedure, reaction between oxindole-activated spiro-DAC **1a** (0.044 g, 0.26 mmol, 1.5 equiv) and 2,3-dioxopyrrolidine derivative **2c** (0.05 g, 0.17 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **3ac**, which was purified by silica gel column chromatography (3:7 EtOAc:Hexane as eluent) to give the title compound as colorless oil in 57% (0.045 g) yield: R_f 0.5 (1:1 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.31 (d, *J* = 7.4 Hz, 1H), 7.23-7.15 (m, 4H), 7.03-6.96 (m, 3H), 6.79 (d, *J* = 7.9 Hz, 2H), 6.72 (d, *J* = 7.7 Hz, 1H), 6.65 (d, *J* = 8.0 Hz, 2H), 4.49-4.41 (m 2H), 4.15 (s, 1H), 3.92 (d, *J* = 12.2 Hz, 1H), 3.64 (d, *J* = 12.2 Hz, 1H), 3.18 (s, 3H), 2.56-2.48 (m, 1H), 2.45-2.29 (m, 2H), 2.19-2.13 (m, 4H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 205.6, 179.6, 159.0, 143.1, 137.5, 134.6, 131.0, 130.4, 129.5, 128.8, 128.4, 128.1 (2), 127.8, 123.3, 122.2, 108.2, 62.1, 58.8, 54.0, 53.9, 48.4, 37.6, 37.5, 26.2, 21.1; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{30}H_{29}N_2O_3$ 465.2178, mass found 465.2165.

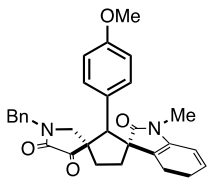
1''-benzyl-2'-(4-isopropylphenyl)-1-methyldispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (3ad):



3ad

Following the general procedure, reaction between oxindole-activated spiro-DAC **1a** (0.042 g, 0.24 mmol, 1.5 equiv) and 2,3-dioxopyrrolidine derivative **2d** (0.05 g, 0.16 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **3ad**, which was purified by silica gel column chromatography (1:3 EtOAc:Hexane as eluent) to give the title compound as colorless oil in 61% (0.047 g) yield. R_f 0.4 (1:1 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.33 (d, *J* = 7.4 Hz, 1H), 7.24-7.17 (m, 4H), 7.05-7.01 (m, 3H), 6.87 (d, *J* = 8.0 Hz, 2H), 6.74 (d, *J* = 7.8 Hz, 1H), 6.68 (d, *J* = 8.0 Hz, 2H), 4.60 (d, *J* = 14.5 Hz, 1H), 4.26 (d, *J* = 14.5 Hz, 1H), 4.19 (s, 1H), 3.87 (d, *J* = 12.1 Hz, 1H), 3.63 (d, *J* = 12.1 Hz, 1H), 3.19 (s, 3H), 2.76-2.69 (m, 1H), 2.53-2.38 (m, 3H), 2.19-2.13 (m, 1H), 1.11 (d, *J* = 6.8 Hz, 6H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 205.22, 179.56, 159.12, 148.21, 143.17, 134.60, 131.55, 130.61, 128.91, 128.42, 128.22, 128.14, 128.06, 126.82, 123.30, 122.26, 108.23, 61.56, 58.72, 54.28, 53.93, 48.49, 37.72, 37.56, 33.52, 26.25, 23.74; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{32}H_{33}N_2O_3$ 493.2491, mass found 493.2482.

1''-benzyl-2'-(4-methoxyphenyl)-1-methyldispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (3ae):

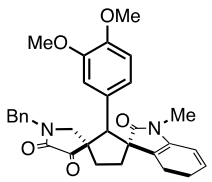


3ae

Following the general procedure, reaction between oxindole-activated spiro-DAC **1a** (0.042 g, 0.24 mmol, 1.5 equiv) and 2,3-dioxopyrrolidine derivative **2e** (0.05 g, 0.16 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **3ae,** which was purified by silica gel column chromatography (3:7 EtOAc:Hexane as eluent) to give the title compound as colorless oil in 66% (0.052 g) yield. R_f 0.5 (1:1

EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.31 (d, *J* = 7.4 Hz, 1H), 7.22-7.15 (m, 4H), 7.02 (t, *J* = 7.5 Hz, 1H), 6.96-6.94 (m, 2H), 6.71 (d, *J* = 8.9 Hz, 3H), 6.50 (d, *J* = 8.6 Hz, 2H), 4.51-4.42 (m, 2H), 4.11 (s, 1H), 3.95 (d, *J* = 12.2 Hz, 1H), 3.66-3.62 (m, 4H), 3.18 (s, 3H), 2.56-2.48 (m, 1H), 2.45-2.29 (m, 2H), 2.19-2.13 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 205.7, 179.6, 159.0 (2), 143.2, 134.6, 130.4, 129.5, 128.9, 128.5, 128.1, 127.9, 126.0, 123.3, 122.2, 114.1, 108.2, 62.1, 58.9, 55.1, 53.9, 48.4, 37.5, 37.4, 26.2; HRMS m/z $[M+Na]^+$ calculated for $C_{30}H_{28}N_2NaO_4$ 503.1947, mass found 503.1935.

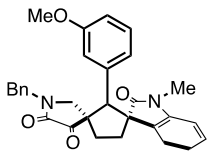
1''-benzyl-2'-(3,4-dimethoxyphenyl)-1-methyldispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (3af):



3af

Following the general procedure, reaction between oxindole-activated spiro-DAC **1a** (0.039 g, 0.23 mmol, 1.5 equiv) and 2,3-dioxopyrrolidine derivative **2f** (0.05 g, 0.15 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **3af,** which was purified by silica gel column chromatography (1:13 EtOAc:Hexane as eluent) to give the title compound as colorless oil in 54% (0.041 g) yield. R_f 0.35 (1:1 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.31 (d, *J* = 7.4 Hz, 1H), 7.23-7.27 (m, 4H), 7.03 (t, *J* = 7.5 Hz, 1H), 6.92 (d, *J* = 7.1 Hz, 2H), 6.73 (d, *J* = 7.7 Hz, 1H), 6.46 (d, *J* = 8.3 Hz, 1H), 6.31-6.27 (m, 2H), 4.62 (d, *J* = 14.5 Hz, 1H), 4.35 (d, *J* = 14.5 Hz, 1H), 4.10 (s, 1H), 3.96 (d, *J* = 12.2 Hz, 1H), 3.74 (s, 3H), 3.63 (d, *J* = 12.2 Hz, 1H), 3.54 (s, 3H), 3.19 (s, 3H), 2.58-2.30 (m, 3H), 2.21-2.15 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 206.03, 179.67, 159.06, 148.65, 148.46, 143.10, 134.49, 130.36, 128.83, 128.54, 128.06, 128.04, 126.39, 123.40, 122.25, 120.84, 111.01, 110.88, 108.23, 62.56, 58.87, 55.68, 55.62, 54.05, 53.79, 48.49, 37.62, 37.42, 26.22; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{31}H_{31}N_2O_5$ 511.2233, mass found 511.2216.

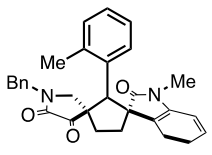
1''-benzyl-2'-(3-methoxyphenyl)-1-methyldispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (3ag):



3ag

Following the general procedure, reaction between oxindole-activated spiro-DAC **1a** (0.042 g, 0.24 mmol, 1.5 equiv) and 2,3-dioxopyrrolidine derivative **2g** (0.05 g, 0.16 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **3ag,** which was purified by silica gel column chromatography (7:13 EtOAc:Hexane as eluent) to give the title compound as colorless oil in 58% (0.045 g) yield. R_f 0.4 (1:1 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.31 (d, *J* = 7.4 Hz, 1H), 7.22-7.19 (m, 4H), 7.04-6.93 (m, 3H), 6.91 (t, *J* = 8.0 Hz, 1H), 6.74 (d, *J* = 7.7 Hz, 1H), 6.58 (d, *J* = 6.7 Hz, 1H), 6.35 (d, *J* = 7.7 Hz, 1H), 6.28 (s, 1H), 4.47 (brs, 2H), 4.17 (s, 1H), 3.90 (d, *J* = 12.2 Hz, 1H), 3.64 (d, *J* = 12.2 Hz, 1H), 3.54 (s, 3H), 3.19 (s, 3H), 2.56-2.28 (m, 3H), 2.19-2.14 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 205.5, 179.5, 159.6, 159.0, 143.1, 135.7, 134.6, 130.2, 129.8, 128.9, 128.5, 128.1, 128.0, 123.3, 122.2, 120.4, 113.9, 113.4, 108.3, 62.2, 58.6, 55.0, 54.1, 53.8, 48.5, 37.7, 37.6, 26.2; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{30}H_{29}N_2O_4$ 481.2127, mass found 481.2115.

1''-benzyl-1-methyl-2'-(o-tolyl)dispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (3ah):

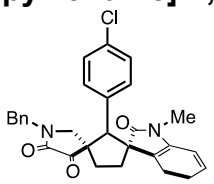


3ah

Following the general procedure, reaction between oxindole-activated spiro-DAC **1a** (0.044 g, 0.26 mmol, 1.5 equiv) and 2,3-dioxopyrrolidine derivative **2h** (0.05 g, 0.17 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **3ah,** which was purified by silica gel column chromatography (3:7 EtOAc:Hexane as eluent) to give the title compound as colorless oil in 53% (0.042 g) yield. R_f 0.5 (1:1 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.40 (d, *J* = 7.4 Hz, 1H), 7.25 (brs, 3H), 7.18-7.13 (m, 2H), 7.04-6.85 (m, 6H), 6.63 (d, *J* = 7.7 Hz, 1H), 4.67 (d, *J* = 14.6

Hz, 1H), 4.57 (s, 1H), 4.20 (d, *J* = 14.6 Hz, 1H), 4.01 (d, *J* = 11.8 Hz, 1H), 3.66 (d, *J* = 11.8 Hz, 1H), 3.11 (s, 3H), 2.70-2.62 (m, 1H), 2.47-2.33 (m, 2H), 2.17-2.14 (m, 1H), 2.09 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 205.01, 179.83, 158.94, 143.19, 137.72, 134.56, 132.58, 131.22, 130.30, 129.04, 128.51, 128.20, 128.17, 128.10, 127.67, 125.72, 122.99, 122.09, 108.19, 60.59, 50.56, 54.02, 53.67, 48.45, 38.00, 36.63, 26.16, 20.54; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{30}H_{28}N_2NaO_3$ 487.1998, mass found 487.1988.

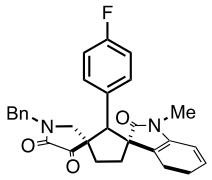
1''-benzyl-2'-(4-chlorophenyl)-1-methyldispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (3ai):



3ai

Following the general procedure, reaction between oxindole-activated spiro-DAC **1a** (0.042 g, 0.24 mmol, 1.5 equiv) and 2,3-dioxopyrrolidine derivative **2i** (0.05 g, 0.16 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **3ai,** which was purified by silica gel column chromatography (3:7 EtOAc:Hexane as eluent) to give the title compound as colorless oil in 62% (0.048 g) yield. R_f 0.45 (1:1 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.66-7.52 (m, 6H), 7.38 (t, *J* = 7.5 Hz, 1H), 7.31-7.28 (m, 3H), 7.08-7.03 (m, 3H), 4.98 (d, *J* = 14.5 Hz, 1H), 4.70 (d, *J* = 14.5 Hz, 1H), 4.45 (s, 1H), 4.22 (d, *J* = 12.3 Hz, 1H), 4.01 (d, *J* = 12.3 Hz, 1H), 3.52 (s, 3H), 2.94-2.86 (m, 1H), 2.81-2.64 (m, 2H), 2.56-2.50 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 205.3, 179.2, 158.8, 143.1, 134.4, 133.9, 132.6, 129.9, 129.7, 129.0, 128.7, 128.1 (2), 123.4, 122.2, 108.4, 61.7, 58.9, 53.7, 53.6, 48.5, 37.7, 37.5, 26.3; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{29}H_{26}ClN_2O_3$ 485.1632, mass found 485.1619.

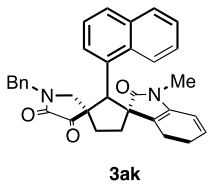
1''-benzyl-2'-(4-fluorophenyl)-1-methyldispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (3aj):



3aj

Following the general procedure, reaction between oxindole-activated spiro-DAC **1a** (0.044 g, 0.26 mmol, 1.5 equiv) and 2,3-dioxopyrrolidine derivative **2j** (0.05 g, 0.17 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **3aj**, which was purified by silica gel column chromatography (3:7 EtOAc:Hexane as eluent) to give the title compound as colorless oil in 54% (0.043 g) yield. R_f 0.4 (1:1 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.33 (d, *J* = 7.4 Hz, 1H), 7.28-7.19 (m, 4H), 7.05 (t, *J* = 7.5 Hz, 1H), 7.00-6.99 (m, 2H), 6.78-6.73 (m, 3H), 6.66 (t, *J* = 8.5 Hz, 2H), 4.61 (d, *J* = 14.5 Hz, 1H), 4.40 (d, *J* = 14.5 Hz, 1H), 4.14 (s, 1H), 3.91 (d, *J* = 12.2 Hz, 1H), 3.68 (d, *J* = 12.2 Hz, 1H), 3.20 (s, 3H), 2.61-2.53 (m, 1H), 2.48-2.31 (m, 2H), 2.23-2.17 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 205.3, 179.3, 162.2 (d, *J* = 247.5 Hz), 158.9, 143.1, 134.5, 130.1, 130.1, 130.0, 129.9 (2), 129.0, 128.7, 128.2, 128.1, 122.8 (d, *J* = 122.9 Hz), 115.8 (d, *J* = 21.3 Hz), 108.3, 61.7, 60.0, 53.8, 53.7, 48.5, 37.6, 37.4, 26.3; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{29}H_{26}FN_2O_3$ 469.1927, mass found 469.1916.

1''-benzyl-1-methyl-2'-(naphthalen-1-yl)dispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (3ak):

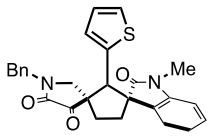


3ak

Following the general procedure, reaction between oxindole-activated spiro-DAC **1a** (0.039 g, 0.23 mmol, 1.5 equiv) and 2,3-dioxopyrrolidine derivative **2k** (0.05 g, 0.15 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **3ak,** which was purified by silica gel column chromatography (3:7 EtOAc:Hexane as eluent) to give the title compound as a mixture of two diastereomers in 55% (0.046 g) yield. R_f 0.45 (1:1 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.92 (d, *J* = 8.8 Hz, 1H), 7.83 (d,

J = 8.5 Hz, 0.2H), 7.75-7.68 (m, 1.2H), 7.57-7.47 (m, 2.2H), 7.41-7.34 (m, 2.6H), 7.29-7.27 (m, 1H), 7.22-7.04 (m, 6H), 6.96-6.87 (m, 2H), 6.76 (d, *J* = 7.5 Hz, 2H), 6.69 (d, *J* = 7.8 Hz, 0.4H), 6.64 (d, *J* = 7.8 Hz, 1H), 6.36 (d, *J* = 8.1 Hz, 0.2H), 5.15 (s, 1H), 4.92 (s, 0.2H), 4.50 (d, *J* = 15.1 Hz, 0.2H), 4.39-4.33 (m, 1.2H), 4.18 (d, *J* = 14.6 Hz, 1H), 3.84 (d, *J* = 11.9 Hz, 1H), 3.62 (d, *J* = 11.9 Hz, 1H), 3.51 (d, *J* = 11.0 Hz, 0.2H), 3.41-3.38 (m, 0.2H), 3.20 (s, 3H), 3.00-2.95 (m, 0.2H), 2.90 (s, 0.5H), 2.82-2.74 (m, 1H), 2.55-2.36 (m, 2.6H), 2.21-2.26 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 205.7, 205.2, 181.7, 179.9, 159.5, 158.7, 143.2, 143.0, 134.4, 133.9, 133.8, 133.7, 132.9, 132.2, 131.7, 130.4, 130.2, 129.8, 129.0 (2), 128.9, 128.8 (2), 128.6 (2), 128.4, 128.0, 127.9, 128.8 (2), 127.6, 127.3, 126.5, 126.1, 126.0, 125.7, 125.6, 124.7, 124.5, 123.2, 122.9, 122.8, 122.6, 122.0, 108.2, 107.6, 60.4, 59.8, 58.3, 55.4, 54.5, 54.2, 53.4, 51.7, 48.2, 48.1, 37.9, 37.7, 37.2, 36.9, 26.4, 26.3; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for C33H28N2NaO3 523.1988, mass found 523.1981.

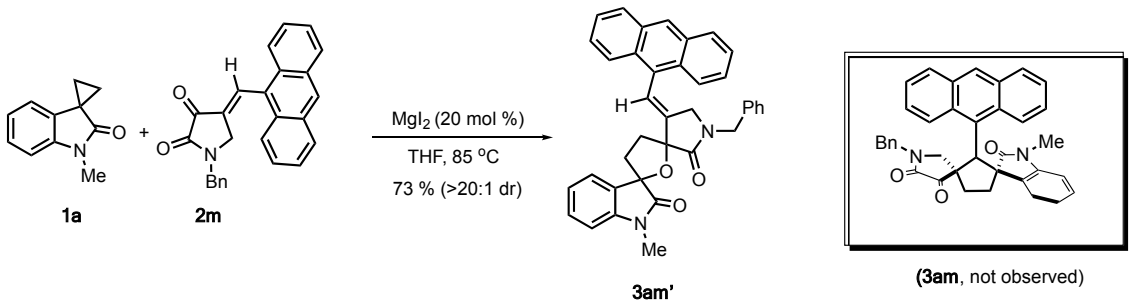
1''-benzyl-1-methyl-2'-(thiophen-2-yl)dispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (3al):



3al

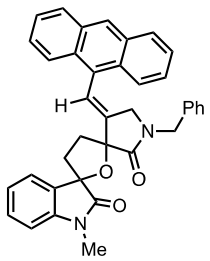
Following the general procedure, reaction between oxindole-activated spiro-DAC **1a** (0.047 g, 0.27 mmol, 1.5 equiv) and 2,3-dioxopyrrolidine derivative **2m** (0.05 g, 0.18 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **3al,** which was purified by silica gel column chromatography (3:7 EtOAc:Hexane as eluent) to give the title compound as colorless oil in 56% (0.045 g) yield. R_f 0.6 (1:1 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.33 (d, *J* = 7.4 Hz, 1H), 7.27 (brs, 3H), 7.22 (t, *J* = 7.7 Hz, 1H), 7.09-7.04 (m, 3H), 6.93 (d, *J* = 5.1 Hz, 1H), 6.74 (d, *J* = 7.8 Hz, 1H), 6.66-6.64 (m, 1H), 6.47-6.46 (m, 1H), 4.62 (d, *J* = 14.5 Hz, 1H), 4.45 (d, *J* = 14.5 Hz, 1H), 4.40 (s, 1H), 4.13 (d, *J* = 12.4 Hz, 1H), 3.66 (d, *J* = 12.4 Hz, 1H), 3.17 (s, 3H), 2.46-2.42 (m, 2H), 2.35-2.21 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 204.70, 178.92, 158.94, 143.44, 135.66, 134.65, 130.43, 129.00, 128.74, 128.30, 128.07, 126.97, 126.73, 125.17, 123.38, 122.31, 108.31, 59.27, 57.87, 54.43, 53.72, 48.59, 37.63, 36.83, 26.36; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{27}H_{25}N_2O_3S$ 457.1586, mass found 457.1574.

5. C-3 carbonyl annulation of 9-anthacene substituted pyrrolidine-2,3-dione derivative (3m) with DAC (1a).



Scheme S2: Observed C-3 carbonyl annulation of 9-anthacene substituted pyrrolidine-2,3-dione derivative **2m** with DAC **1a**.

2'-(anthracen-9-yl)-1''-benzyl-1-methyldispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (3am'):

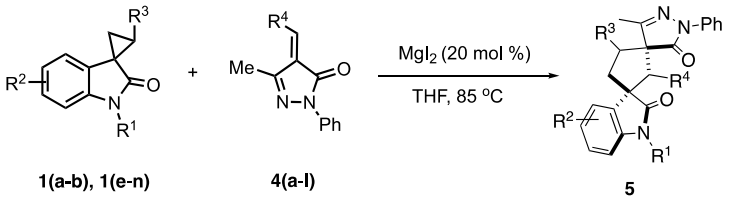


3am'

Following the general procedure, reaction between oxindole-activated spiro-DAC **1a** (0.035 g, 0.20mmol, 1.5 equiv) and 2,3-dioxopyrrolidine derivative **2m** (0.05 g, 0.13 mmol, 1.0 equiv) afforded bis-spiroheterocyclic derivative **3am',** which was purified by silica gel column chromatography (3:7 EtOAc:Hexane as eluent) to give the title compound as colorless oil in 73% (0.052 g) yield as single diastereomer. R_f 0.5 (1:1 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.26 (s, 1H), 7.90-7.84 (m, 3H), 7.58 (d, *J* = 7.4 Hz, 1H), 7.47-7.38 (m, 2H), 7.35-7.29 (m, 2H), 7.20-7.12 (m, 6H), 7.06-7.02 (m, 3H), 6.21 (d, *J* = 8.8 Hz, 1H), 5.18-5.13 (m, 1H), 4.66 (q, *J* = 8.1 Hz, 1H), 4.50 (d, *J* = 15.0 Hz, 1H), 4.31 (d, *J* = 15.0 Hz, 1H), 3.58 (dd, *J* = 21.2, 9.9 Hz, 1H), 3.25 (s, 3H), 2.96 (s, 2H), 2.43-2.48 (m, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 173.7, 171.8, 143.3, 135.4, 134.7, 131.2, 131.1, 131.0, 129.4, 129.2, 128.9, 128.6 (2), 128.5, 128.4 (2), 127.4, 126.8, 126.1, 125.8, 125.5, 125.4, 125.3 (2), 125.1, 125.0,

123.4, 108.4, 89.0, 68.2, 59.8, 47.0, 46.8, 36.1, 26.6; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{37}H_{30}N_2NaO_3$ 573.2154, mass found 573.2139.

6. General procedure for the synthesis of dispiropyrazolone[cyclopentane]oxindole (5).

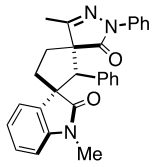


Oxindole-activated spiro-DAC (**1**; 1.5 equiv) and alkylidene pyrazolone (**4**; 1.0 equiv) were taken in dry THF (0.2 M) under argon and was added MgI_2 (20 mol%). The reaction mixture was heated to 85°C. Progress of the reaction was monitored by thin layer chromatography (TLC) until disappearance of the starting materials (ca. 12-15 h) was observed.

The reaction was then diluted with EtOAc (2 mL), washed with water (2 mL) and brine (2 mL) and the organic layer was dried over anhydrous Na_2SO_4. Solvents were removed under reduced pressure and the crude product was purified by flash silica gel column chromatography to obtain the bis-spirocyclopentane oxindole derivatives (**5**).

7. Characterization of dispiropyrazolone[cyclopentane]oxindole derivatives (5).

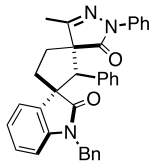
1,3''-dimethyl-1'',2'-diphenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5aa):



5aa

Following the general procedure, reaction between oxindole-activated spiro-DAC **1a** (0.049 g, 0.29 mmol, 1.5 equiv) and alkylidene pyrazolone **4a** (0.05 g, 0.19 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5aa**, which was purified by silica gel column chromatography (3:7 EtOAc:Hexane as eluent) to give the title compound as single diastereomer in 82% (0.068 g) yield. R_f 0.3 (2:3 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.91 (d, *J* = 8.4 Hz, 2H), 7.55 (d, *J* = 7.3 Hz, 1H), 7.51-7.47 (m, 2H), 7.43 (t, *J* = 7.7 Hz, 1H), 7.33-7.29 (m, 4H), 7.27-7.25 (m, 3H), 6.86 (d, *J* = 7.7 Hz, 1H), 4.10 (s, 1H), 3.44-3.36 (m, 1H), 3.27 (s, 3H), 3.14-3.07 (m, 1H), 2.58 (s, 3H), 2.56-2.48 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.3, 173.7, 161.0, 143.6, 138.1, 133.3, 132.7, 129.4, 128.6, 128.5, 128.1 (2), 124.6, 122.6, 121.3, 118.9, 108.0, 64.2, 64.1, 59.1, 37.5, 34.2, 26.2, 13.8; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{28}H_{25}N_3NaO_2$ 458.1844, mass found 458.1827.

1-benzyl-3''-methyl-1'',2'-diphenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5ba):

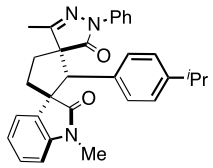


5ba

Following the general procedure, reaction between oxindole-activated spiro-DAC **1b** (0.056 g, 0.23 mmol, 1.5 equiv) and alkylidene pyrazolone **4a** (0.04 g, 0.15 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5ba**, which was purified by silica gel column chromatography (1:4 EtOAc:Hex as eluent) to give the title compound as single diastereomer in 85% (0.067 g) yield. R_f 0.3 (3:7

EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.80 (d, *J* = 7.7 Hz, 2H), 7.40-7.38 (m, 1H), 7.31 (t, *J* = 8.0 Hz, 2H), 7.20-7.18 (m, 2H), 7.14-7.04 (m, 9H), 6.72 (d, *J* = 6.9 Hz, 2H), 6.49-6.47 (m, 1H), 5.13 (d, *J* = 15.7 Hz, 1H), 4.40 (d, *J* = 15.7 Hz, 1H), 3.96 (s, 1H), 3.25-3.18 (m, 1H), 3.01-2.95 (m, 1H), 2.45-2.38 (m, 1H), 2.36-2.29 (m, 4H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.1, 173.8, 160.6, 142.6, 138.1, 135.7, 134.0, 133.3, 129.8, 128.7, 128.6, 128.4 (2), 128.2, 127.2, 127.0, 124.7, 122.7, 121.4, 118.8, 109.0, 64.6, 64.4, 59.1, 53.7, 37.7, 34.6, 13.7; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{34}H_{30}N_3O_2$ 512.2338, mass found 512.2318.

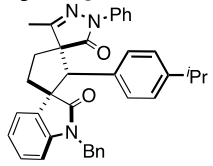
2'-(4-isopropylphenyl)-1,3''-dimethyl-1''-phenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5ab):



5ab

Following the general procedure, reaction between oxindole-activated spiro-DAC **1a** (0.042 g, 0.24 mmol, 1.5 equiv) and alkylidene pyrazolone **4b** (0.05 g, 0.16 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5ab**, which was purified by silica gel column chromatography (3:7 EtOAc:Hexane as eluent) to give the title compound as single diastereomer in 70% (0.055 g, colorless oil) yield. R_f 0.5 (3:7 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.63 (d, *J* = 8.2 Hz, 2H), 7.33 (d, *J* = 7.3 Hz, 1H), 7.28-7.19 (m, 3H), 7.09-7.04 (m, 2H), 7.00 (d, *J* = 8.1 Hz, 2H), 6.88 (d, *J* = 8.1 Hz, 2H), 6.66 (d, *J* = 7.7 Hz, 1H), 3.89 (s, 1H), 3.18-3.13 (m, 1H), 3.06 (s, 3H), 2.91-2.84 (m, 1H), 2.71-2.64 (m, 1H), 2.36 (s, 3H), 2.34-2.24 (m, 2H), 1.04 (d, *J* = 6.9 Hz, 6H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.4, 173.7, 161.0, 148.4, 143.6, 138.0, 132.9, 130.6, 129.1, 128.5, 128.4, 126.4, 124.6, 122.5, 121.3, 119.1, 107.9, 64.3, 63.7, 58.8, 37.7, 33.8, 33.6, 26.1, 23.8, 23.7, 13.7; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{31}H_{31}N_3NaO_2$ 500.2314, mass found 500.2304.

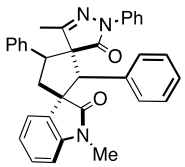
1-benzyl-2'-(4-isopropylphenyl)-3''-methyl-1''-phenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5bb):



5bb

Following the general procedure, reaction between oxindole-activated spiro-DAC **1b** (0.048 g, 0.20 mmol, 1.5 equiv) and alkylidene pyrazolone **4b** (0.04 g, 0.13 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5bb**, which was purified by silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title compound as single diastereomer in 97% (0.07 g, colorless oil) yield. R_f 0.3 (3:7 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.72 (d, *J* = 8.3 Hz, 2H), 7.38 (d, *J* = 7.4 Hz, 1H), 7.30 (t, *J* = 7.8 Hz, 2H), 7.16-7.06 (m, 8H), 6.91 (d, *J* = 8.0 Hz, 2H), 6.85 (d, *J* = 7.1 Hz, 2H), 6.48 (d, *J* = 7.1 Hz, 1H), 5.16 (d, *J* = 15.7 Hz, 1H), 4.45 (d, *J* = 15.8 Hz, 1H), 3.96 (s, 1H), 3.17-3.11 (m, 1H), 3.04-2.98 (m, 1H), 2.77-2.70 (m, 1H), 2.42-2.27 (m, 5H), 1.10 (d, *J* = 6.9 Hz, 6H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.2, 173.8, 160.7, 148.5, 142.7, 138.1, 135.8, 133.9, 130.6, 129.6, 128.6 (2), 128.3, 127.2 (2), 126.4, 124.8, 122.7, 121.4, 119.0, 109.1, 64.5, 64.0, 58.9, 43.8, 38.1, 34.2, 33.6, 23.9, 23.7, 13.7; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{37}H_{36}N_3O_2$ 554.2808, mass found 554.2797.

1,3''-dimethyl-1'',2',4'-triphenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5ea):



5ea

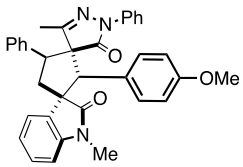
Following the general procedure, reaction between oxindole-activated spiro-DAC **1e** (0.071 g, 0.29 mmol, 1.5 equiv) and alkylidene pyrazolone **4a** (0.05 g, 0.19 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5ea** as mixture of diastereomer (d.r =1.5:1) in 98% (0.1 g) combined yield. This diastereomeric mixture was subjected to further chromatographic separation (1:6 EtOAc:Hexane as eluent) to obtain each of the isomer separately for the purpose of X-ray single crystal analysis. This operation afforded the major diastereomer in 44% (0.043 g) yield and

minor diastereomer was isolated in 28% (0.027 g) yield. Note: Compound **5ea** can be isolated in 92 % combined yield when the reaction is performed with 1g of **4a.**

Major diastereomer: R_f 0.24 (1:5 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.59 (t, *J* = 7.3 Hz, 3H), 7.40 (d, *J* = 7.3 Hz, 2H), 7.35-7.28 (m, 4H), 7.24-7.19 (m, 3H), 7.11-7.07 (m, 6H), 6.73 (d, *J* = 7.7 Hz, 1H), 4.35 (t, *J* = 13.3 Hz, 1H), 4.18-4.12 (m, 2H), 2.95 (s, 3H), 2.37-2.34 (m, 4H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 176.4, 170.9, 158.2, 143.3, 137.8, 137.0, 135.1, 133.8, 129.2, 128.6, 128.5 (2), 128.4, 128.1 (2), 128.0, 124.9, 122.8, 121.4, 119.4, 107.9, 69.5, 64.3, 57.3, 52.6, 40.3, 26.3, 13.7; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{34}H_{29}N_3NaO_2$ 534.2157, mass found 534.2147.

Minor diastereomer: R_f 0.22 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.60 (d, *J* = 8.3 Hz, 2H), 7.41 (d, *J* = 7.4 Hz, 1H), 7.30-7.28 (m, 3H), 7.25-7.19 (m, 4H), 7.16-7.06 (m, 7H), 6.69 (d, *J* = 7.7 Hz, 1H), 5.65 (dd, *J* = 13.7, 5.9 Hz, 1H), 4.06 (s, 1H), 3.19 (s, 3H), 2.89 (t, *J* = 13.2 Hz, 1H), 2.68 (dd, *J* = 12.7, 6.0 Hz, 1H), 2.07 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.6, 173.7, 160.6, 143.7, 138.3, 138.0, 133.2, 129.7, 128.8 (2), 128.6, 128.1 (2), 127.3, 126.5, 124.8, 122.6, 121.4, 119.1, 108.1, 68.7, 62.9, 58.1, 50.0, 40.7, 26.3, 15.6; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{34}H_{29}N_3NaO_2$ 534.2157, mass found 534.2150.

2'-(4-methoxyphenyl)-1,3''-dimethyl-1'',4'-diphenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5ec):

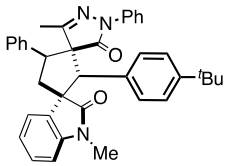


5ec

Following the general procedure, reaction between oxindole-activated spiro-DAC **1e** (0.052 g, 0.21 mmol, 1.5 equiv) and alkylidene pyrazolone **4c** (0.04 g, 0.14 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5ec**, which was purified by silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title compound as mixture of diastereomer (d.r =2:1) in 91% (0.067 g, colorless oil) combined yield. R_f 0.2 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.64 (d, *J* = 8.1 Hz, 1H), 7.59-7.56 (m, 3H), 7.38-7.36 (m, 2.6H), 7.31-7.27 (m, 2H), 7.25-7.23 (m, 5.6H), 7.20-7.04 (m, 9H), 6.71-6.65 (m, 1.6H), 6.58 (t, *J* = 8.3 Hz, 3H), 5.61 (dd,

J = 13.7, 5.9 Hz, 0.6H), 4.31 (t, *J* = 13.3 Hz, 1H), 4.13-4.08 (m, 2H), 3.99 (s, 0.6H), 3.63 (s, 3H), 3.60 (s, 1.8H), 3.15 (s, 1.7H), 2.94 (s, 3H), 2.85 (t, *J* = 13.2 Hz, 0.6H), 2.65 (dd, *J* = 12.7, 6.0 Hz, 0.6H), 2.34-2.30 (m, 4H), 2.04 (s, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.6, 176.5, 173.8, 171.0, 160.7, 159.3, 158.3, 143.7, 143.3, 138.3, 138.0, 137.8, 137.0, 135.3, 131.3, 130.7, 130.5, 128.7, 128.6, 128.5 (2), 128.4, 128.0, 127.9, 127.2, 126.4, 125.7, 125.2, 124.8, 124.7, 122.8, 122.5, 121.4, 121.3, 119.3, 119.0, 113.7, 113.4, 108.1, 107.8, 69.7, 68.7, 63.8, 62.3, 58.3, 57.3, 55.1, 55.0, 52.4, 49.9, 40.4, 40.3, 26.3, 26.2, 15.6, 13.7; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{35}H_{31}N_3NaO_3$ 564.2263, mass found 564.2241.

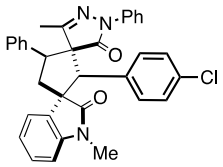
2'-(4-(tert-butyl)phenyl)-1,3''-dimethyl-1'',4'-diphenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5ed):



5ed

Following the general procedure, reaction between oxindole-activated spiro-DAC **1e** (0.049 g, 0.20 mmol, 1.5 equiv) and alkylidene pyrazolone **4d** (0.04 g, 0.13 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5ed**, which was purified by silica gel column chromatography (1:4 EtOAc:Hex as eluent) to give the title compound as mixture of diastereomer (d.r = 1.2:1) in 98% (0.07 g) combined yield. R_f 0.25 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.76 (t, *J* = 7.9 Hz, 2H), 7.63 (d, *J* = 8.2 Hz, 2H), 7.59-7.56 (m, 3H), 7.53-7.36 (m, 14H), 7.33-7.30 (m, 5H), 7.19-7.17 (m, 5H), 6.92 (d, *J* = 7.7 Hz, 2H), 5.78 (dd, *J* = 13.7, 5.8 Hz, 0.8H), 4.51 (t, *J* = 13.3 Hz, 1H), 4.35-4.26 (m, 3.7H), 3.42 (s, 3H), 3.13 (s, 2.4H), 3.05 (t, *J* = 13.2 Hz, 1H), 2.85 (dd, *J* = 12.6, 6.0 Hz, 1H), 2.58 -2.49 (m, 3.4H), 2.24 (s, 3H), 1.36 (s, 7H), 1.31 (s, 9H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.7, 176.5, 173.8, 171.0, 160.6, 158.4, 150.8, 150.7, 143.9, 143.3, 138.5, 137.8, 137.2,135.2, 130.8 (2), 130.3, 129.0, 128.8, 128.7 (2), 128.5, 128.4, 128.1, 128.0, 127.2, 126.5, 125.3, 125.0, 124.8, 122.8, 122.6, 121.4 (2), 119.4, 108.2, 107.9, 69.5, 67.0, 63.8, 63.2, 57.5, 57.2, 52.6, 49.1, 41.3, 40.4, 34.4 (2), 31.2 (2), 26.4, 26.2, 15.6, 13.8; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{38}H_{38}N_3O_2$ 568.2964, mass found 568.2956.

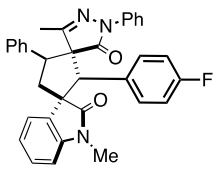
2'-(4-chlorophenyl)-1,3''-dimethyl-1'',4'-diphenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5ee):



5ee

Following the general procedure, reaction between oxindole-activated spiro-DAC **1e** (0.049 g, 0.20 mmol, 1.5 equiv) and alkylidene pyrazolone **4e** (0.04 g, 0.13 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5ee**, which was purified by silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title compound as mixture of diastereomer (d.r = 2:1) in 88% (0.065 g) combined yield. R_f 0.2 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.63 (d, *J* = 8.2 Hz, 1H), 7.58-7.54 (m, 3H), 7.39-7.36 (m, 3H), 7.33 – 7.27 (m, 3H), 7.25-7.22 (m, 5H), 7.18-7.10 (m, 5H), 7.06-7.06 (m, 6H), 6.74-6.68 (m, 2H), 5.61 (dd, *J* = 13.7, 5.9 Hz, 0.6H), 4.31 (t, *J* = 13.3 Hz, 1H), 4.14-4.11 (m, 2H), 3.99 (s, 0.6H), 3.15 (s, 1.7H), 2.96 (s, 3H), 2.87 (t, *J* = 13.3 Hz, 0.6H), 2.66 (dd, *J* = 12.7, 6.0 Hz, 0.6H), 2.36-2.31 (m, 4H), 2.05 (s, 1.7H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.3, 176.2, 173.5, 170.7, 160.5, 158.0, 143.6, 143.2, 137.9, 137.6 (2), 134.9, 134.2 (2), 132.3, 131.7, 131.3, 130.8, 130.2, 129.0, 128.8, 128.7, 128.6 (4), 128.2, 128.1, 127.9, 127.4, 126.4, 125.0, 124.9, 123.0, 122.7, 121.4, 121.3, 119.3, 119.0, 108.3, 108.0, 69.6, 68.5, 63.4, 62.0, 58.3, 57.2, 52.5, 50.3, 40.4, 26.3 (2), 15.7, 13.7; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{34}H_{28}ClN_3NaO_2$ 568.1768, mass found 568.1747.

2'-(4-fluorophenyl)-1,3''-dimethyl-1'',4'-diphenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5ef):

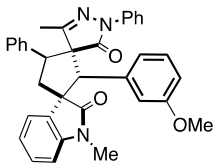


5ef

Following the general procedure, reaction between oxindole-activated spiro-DAC **1e** (0.052 g, 0.21 mmol, 1.5 equiv) and alkylidene pyrazolone **4f** (0.04 g, 0.14 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5ef**, which was purified by

silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title compound as mixture of diastereomer (d.r = 1.5:1) in 98% (0.074 g) combined yield. R_f 0.2 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.64-7.54 (m, 4.4H), 7.39-7.36 (m, 3H), 7.33-7.29 (m, 3H), 7.24-7.05 (m, 14H), 6.78-6.67 (m, 5.21H), 5.62 (dd, *J* = 13.7, 5.9 Hz, 0.7H), 4.32 (t, *J* = 13.3 Hz, 1H), 4.14-4.09 (m, 2.1H), 4.00 (s, 0.7H), 3.15 (s, 2H), 2.95 (s, 3H), 2.87 (t, *J* = 13.2 Hz, 1H), 2.66 (dd, *J* = 12.7, 6.0 Hz, 1H), 2.36-2.32 (m, 4H), 2.05 (s, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.4, 176.3, 162.60 (d, *J* = 247.3 Hz), 173.7, 170.8, 160.6, 158.1, 143.6, 143.2, 138.1, 137.9, 137.7, 136.7, 135.0, 131.7 (2), 131.2 (2), 130.3, 129.5 (2), 128.9, 128.8, 128.7, 128.6 (2), 128.1, 127.9, 127.3, 126.4, 125.0, 124.9, 123.0, 122.7, 121.4, 121.3, 119.3, 119.0, 115.4, 115.2, 11.51, 114.9, 108.2, 108.0, 69.7, 68.6, 63.5, 62.1, 58.4, 57.2, 52.4, 50.1, 40.4, 26.3 (2), 15.6, 13.7; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{34}H_{28}FN_3NaO_2$ 522.2063, mass found 522.2051.

2'-(3-methoxyphenyl)-1,3''-dimethyl-1'',4'-diphenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5eg):

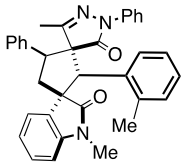


5eg

Following the general procedure, reaction between oxindole-activated spiro-DAC **1e** (0.052 g, 0.21 mmol, 1.5 equiv) and alkylidene pyrazolone **4g** (0.04 g, 0.14 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5eg**, which was purified by silica gel column chromatography (1:3 EtOAc:Hexane as eluent) to give the title compound as mixture of diastereomer (d.r = 2:1) in 76% (0.056 g) combined yield. R_f 0.2 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.68.7.59 (m, 4H), 7.41 (d, *J* = 7.5 Hz, 3H), 7.35-7.28 (m, 6H), 7.23-7.20 (m, 3H), 7.17-7.07 (m, 4H), 6.99-6.89 (m, 4H), 6.76-6.56 (m, 5H), 5.64 (dd, *J* = 13.8, 5.8 Hz, 0.6H), 4.35 (t, *J* = 13.3 Hz, 1H), 4.17-4.11 (m, 2H), 4.05 (s, 0.6H), 3.71 (s, 1.8H), 3.58 (s, 3H), 3.20 (s, 1.8H), 2.99 (s, 3H), 2.89 (t, *J* = 13.3 Hz, 1H), 2.67 (dd, *J* = 12.7, 5.9 Hz, 0.6H), 2.38-2.34 (m, 4H), 2.09 (s, 1.8H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.6, 176.4, 173.7, 171.1, 160.6, 159.3, 159.1, 158.3, 143.7, 143.3, 138.1, 137.8, 137.0, 135.3, 135.1, 134.7, 130.5,

129.1, 128.9, 128.8 (2), 128.7, 128.6, 128.5 (2), 128.1, 128.0, 127.3, 126.5, 124.8, 124.7, 122.8, 122.6, 122.0, 121.4 (2), 121.3, 119.1, 119.0, 114.7 (2), 114.4, 113.9, 108.2, 107.9, 69.6, 68.6, 64.1 ,62.6, 58.1, 57.2, 55.2, 55.1, 52.5, 50.2, 40.7, 40.5, 26.3, 26.2, 15.7, 13.7; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{35}H_{32}N_3O_3$ 542.2444, mass found 542.2435.

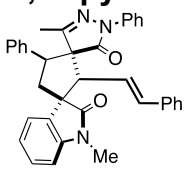
1,3''-dimethyl-1'',4'-diphenyl-2'-(m-tolyl)dispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5eh):



5eh

Following the general procedure, reaction between oxindole-activated spiro-DAC **1e** (0.052 g, 0.21 mmol, 1.5 equiv) and alkylidene pyrazolone **4h** (0.04 g, 0.14 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5eh**, which was purified by silica gel column chromatography (1:4 EtOAc:Hex as eluent) to give the title compound as mixture of diastereomer (d.r = 2:1) in 85% (0.065 g) combined yield. R_f 0.2 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.98 (d, *J* = 8.0 Hz, 0.5H), 7.91 (d, *J* = 7.7 Hz, 1H), 7.62 (d, *J* = 7.3 Hz, 1H), 7.55 (t, *J* = 7.9 Hz, 3H), 7.45 (d, *J* = 7.3 Hz, 0.5H), 7.39 (d, *J* = 7.6 Hz, 2H), 7.31-7.28 (m, 2H), 7.24-7.10 (m, 8H), 7.09-7.04 (m, 2H), 7.01-6.90 (m, 3H), 6.86 (d, *J* = 7.2 Hz, 1H), 6.76 (d, *J* = 7.5 Hz, 0.5H), 6.68-6.62 (m, 1.5H), 5.69 (dd, *J* = 13.7, 5.8 Hz, 0.5H), 4.62 (s, 0.5H), 4.60 (s, 1H), 4.42 (t, *J* = 13.3 Hz, 1H), 4.17 (dd, *J* = 14.0, 5.9 Hz, 1H), 3.13 (s, 1.5H), 2.92 (d, *J* = 13.2 Hz, 0.5H), 2.86 (s, 3H), 2.67 (dd, *J* = 12.6, 5.9 Hz, 1H), 2.41-2.35 (m, 4H), 2.07 (s, 1.5H), 2.03 (s, 1.5H), 1.65 (s, 3H) ^{13}C NMR (100 MHz, $CDCl_3$) δ 178.1, 176.5, 173.8, 171.4, 160.8, 158.3, 143.7, 143.4, 138.4, 137.9, 137.8, 136.5, 136.3, 135.9, 135.2, 132.1, 131.8, 130.7, 130.5, 130.0, 129.9, 128.9, 128.7, 128.6, 128.5 (3), 128.0 (2), 127.7, 127.6, 127.2, 126.5, 126.4, 125.8, 124.9, 124.8, 122.8, 122.3, 121.6, 121.5, 119.4, 119.2, 108.2, 107.9, 70.1, 68.1, 58.9, 58.4, 57.3, 56.2, 52.8, 49.9, 40.5, 39.9, 26.3, 26.2, 20.2, 19.8, 15.9, 14.0; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{35}H_{32}N_3O_2$ 526.2495, mass found 526.2487.

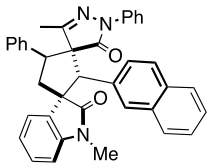
(E)-1,3''-dimethyl-1'',4'-diphenyl-2'-styryldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5ei):



5ei

Following the general procedure, reaction between oxindole-activated spiro-DAC **1e** (0.052 g, 0.21 mmol, 1.5 equiv) and alkylidene pyrazolone **4i** (0.04 g, 0.14 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5ei**, which was purified by silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title compound as mixture of diastereomer (d.r = 2:1) in 97% (0.073 g) combined yield. R_f 0.25 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.88 (d, *J* = 8.3 Hz, 2H), 7.61 (d, *J* = 8.3 Hz, 1H), 7.54-7.50 (m, 0.6H), 7.43-7.30 (m, 6.6H), 7.24 – 7.03 (m, 18H), 6.82-6.77 (m, 1.6H), 6.65-6.53 (m, 1.6H), 6.11 (d, *J* = 15.7 Hz, 0.6H), 5.93 (d, *J* = 15.6 Hz, 1H), 5.45 (dd, *J* = 13.8, 5.7 Hz, 1H), 4.23-4.10 (m, 1H), 3.62 (d, *J* = 10.1 Hz, 0.6H), 3.53 (d, *J* = 9.8 Hz, 1H), 3.20 (s, 3H), 3.17 (s, 1.7H), 2.90 (t, *J* = 13.4 Hz, 1H), 2.66-2.59 (m, 1H), 2.46 (s, 1.7H), 2.26 (dd, *J* = 10.5, 3.5 Hz, 0.6H), 2.12 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.13, 176.18, 173.46, 170.99, 160.76, 158.05, 144.11, 143.37, 138.16, 137.65, 137.61, 136.95, 136.22, 136.11, 135.32, 134.00, 133.82, 130.81, 128.86, 128.77, 128.71, 128.60, 128.50, 128.43, 128.42, 128.00, 127.82, 127.75 (2), 127.35, 126.92, 126.81, 126.40, 124.81, 124.00, 123.31, 122.87, 122.76, 121.77, 121.59, 119.25, 118.93, 108.27, 108.16, 69.81, 68.86, 60.11, 59.13, 58.14, 56.55, 52.21, 51.00, 41.32, 40.35, 26.51, 26.24, 15.99, 13.77; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{36}H_{32}N_3O_2$ 538.2495, mass found 538.2485.

1,3''-dimethyl-2'-(naphthalen-2-yl)-1'',4'-diphenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5ej):

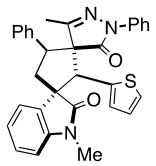


5ej

Following the general procedure, reaction between oxindole-activated spiro-DAC **1e** (0.049 g, 0.20 mmol, 1.5 equiv) and alkylidene pyrazolone **4j** (0.04 g, 0.13 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5ej**, which was purified by

silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title compound as mixture of diastereomer (d.r = 1.5:1) in 93% (0.067 g) combined yield. R_f 0.15 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.67-7.61 (m, 3.3H), 7.59-7.49 (m, 7.3H), 7.45-7.40 (m, 5H), 7.36-7.27 (m, 6.6H), 7.25-7.15 (m, 8.4H), 7.12 – 6.99 (m, 2.3H), 6.70 (d, *J* = 7.7 Hz, 1H), 6.60 (d, *J* = 7.6 Hz, 0.6H), 5.70 (dd, *J* = 13.7, 5.8 Hz, 0.6H), 4.43 – 4.35 (m, 2H), 4.21 – 4.16 (m, 1.6 H), 3.15 (s, 2H), 2.96-2.89 (m, 3.6 H), 2.70 (dd, *J* = 12.6, 6.0 Hz, 1H), 2.40 – 2.35 (m, 4H), 2.12 (s, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.59, 176.41, 173.73,170.99, 160.62, 158.20, 143.64, 143.30, 138.13, 137.91, 137.75, 136.99, 135.08, 132.95, 132.84, 131.62, 130.88, 130.46, 129.31, 128.80,128.76, 128.57, 128.53, 128.32, 128.10, 128.08, 127.99, 127.67, 127.45, 127.37, 127.30,126.95,126.49, 126.09, 126.07, 125.88, 125.79, 124.89, 124.68, 122.87, 122.60, 121.42, 121.37, 119.37, 118.94, 108.19, 107.98, 69.52, 68.64, 64.36, 62.97, 58.29, 57.20, 52.55, 50.39, 40.64, 26.30, 26.23, 15.76, 13.76; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{38}H_{31}N_3NaO_2$ 584.2314, mass found 584.2297.

1,3''-dimethyl-1'',4'-diphenyl-2'-(thiophen-2-yl)dispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5ek):

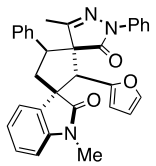


5ek

Following the general procedure, reaction between oxindole-activated spiro-DAC **1e** (0.056 g, 0.23 mmol, 1.5 equiv) and alkylidene pyrazolone **4k** (0.04 g, 0.15 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5ek**, which was purified by silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title compound as mixture of diastereomer (d.r = 1.5:1) in 97% (0.075 g) combined yield. R_f 0.2 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.71 (d, *J* = 8.4 Hz, 1H), 7.63 (d, *J* = 8.4 Hz, 2H), 7.55 (d, *J* = 7.3 Hz, 1H), 7.39 (d, *J* = 7.5 Hz, 3H), 7.35-7.27 (m, 5H), 7.24-7.16 (m, 5H), 7.14– 6.92 (m, 7H), 6.78-6.73 (m, 3H), 5.59 (dd, *J* = 13.8, 5.9 Hz, 0.7H), 4.42 (s, 1H), 4.35 (s, 0.7H), 4.34 – 4.24 (m, 1H), 4.10 (dd, *J* = 14.1, 5.9 Hz, 1H), 3.20 (s, 2H), 3.02 (s, 3H), 2.87 (t, *J* = 13.3 Hz, 0.7H), 2.66 (dd, *J* = 12.8, 6.0 Hz, 0.7H), 2.38 (s, 3H), 2.33 (dd, *J* = 12.6, 5.8 Hz, 1H), 2.06 (s, 2H); ^{13}C

NMR (100 MHz, $CDCl_3$) δ 177.00, 176.02, 173.20, 170.049, 160.35, 157.84, 143.93, 143.35, 138.02, 137.86, 137.76, 136.57, 134.95,134.63, 134.38, 130.26, 129.00, 128.74, 128.67, 128.61, 128.58, 128.52, 128.11, 127.86, 127.55, 127.53, 127.37, 127.31, 127.15, 126.41,124.82, 124.76, 124.59, 122.89, 122.67, 121.39, 121.34, 119.17, 118.98, 108.19, 108.00, 69.87, 68.49, 58.40, 58.29, 57.23, 56.82, 52.26, 49.81, 40.33, 40.24, 26.33, 26.30, 15.58, 13.55; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{32}H_{27}N_3NaO_2S$ 540.1722, mass found 540.1711.

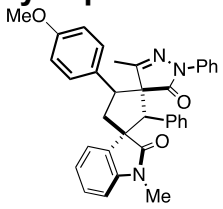
2'-(furan-2-yl)-1,3''-dimethyl-1'',4'-diphenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5el):



5el

Following the general procedure, reaction between oxindole-activated spiro-DAC **1e** (0.060 g, 0.24 mmol, 1.5 equiv) and alkylidene pyrazolone **4l** (0.04 g, 0.16 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5el**, which was purified by silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title compound as mixture of diastereomer (d.r = 1:1) in 95% (0.075 g) combined yield. R_f 0.2 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.77 (d, *J* = 8.1 Hz, 2H), 7.62 (d, *J* = 8.1 Hz, 2H), 7.54 (d, *J* = 7.3 Hz, 1H), 7.40-7.27 (m, 8H), 7.24-6.99 (m, 15H), 6.82 (d, *J* = 7.7 Hz, 2H), 6.22-6.16 (m, 2H), 6.11 (brs, 2H), 5.46 (dd, *J* = 13.9, 5.7 Hz, 1H), 4.37 (s, 1H), 4.31 (s, 1H), 4.24-4.18 (m, 1H), 4.09 (dd, *J* = 14.1, 5.4 Hz, 1H), 3.27 (s, 3H), 3.13 (s, 3H), 2.86 (t, *J* = 13.3 Hz, 1H), 2.61 (dd, *J* = 12.8, 5.8 Hz, 1H), 2.41 (s, 3H), 2.24 (dd, *J* = 12.1, 5.4 Hz, 1H), 2.08 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 176.83, 175.95, 172.95, 170.36, 160.41, 158.40, 148.49, 148.39, 153.85, 143.22, 141.65, 141.32, 138.21, 137.74, 137.48, 136.89, 134.74, 130.31, 129.07, 128.73, 128.71, 128.57, 128.55, 128.15, 127.83, 127.36, 126.39, 124.80, 124.72, 122.80, 122.74, 121.61, 121.40, 119.12, 118.95, 111.09, 110.73, 108.54, 108.38, 108.22, 108.10, 68.17, 66.97, 56.10, 55.63, 55.55, 54.39, 52.26, 50.11, 41.04, 40.82, 26.45, 26.34, 15.72, 13.54; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{32}H_{28}N_3O_3$ 502.2131, mass found 502.2127.

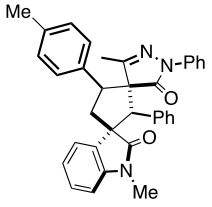
4'-(4-methoxyphenyl)-1,3''-dimethyl-1'',2'-diphenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5ga):



5ga

Following the general procedure, reaction between oxindole-activated spiro-DAC **1g** (0.063 g, 0.23 mmol, 1.5 equiv) and alkylidene pyrazolone **4a** (0.04 g, 0.15mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5ga**, which was purified by silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title compound as mixture of diastereomer (d.r = 1.3:1) in 85% (0.07 g) combined yield. R_f 0.2 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.62-7.57 (m, 4H), 7.53-7.45 (m, 1H), 7.40-7.36 (m, 1H), 7.32-7.28 (m, 4H), 7.25-7.16 (m, 6H), 7.12-7.04 (m, 10H), 6.99-6.92 (m, 1H), 6.79-6.77 (m, 4H), 6.71-6.65 (m, 2H), 5.56 (dd, *J* = 13.8, 5.8 Hz, 0.7H), 4.27 (t, *J* = 13.3 Hz, 1H), 4.14 (s, 1H), 4.08 (dd, *J* = 14.1, 5.9 Hz, 1H), 4.05 (s, 1H), 3.75 (s, 2H), 3.69 (s, 3H), 3.68 (brs, 0.7H), 3.29 (t, *J* = 11.5 Hz, 1H), 3.16 (s, 2H), 2.92 (s, 3H), 2.84 (t, *J* = 13.2 Hz, 0.7H), 2.62 (dd, *J* = 12.6, 5.9 Hz, 0.7H), 2.33 (s, 3H), 2.31-2.28 (m, 1H), 2.09 (s, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.6, 176.4, 173.7, 171.1, 160.8, 159.2, 158.6, 158.3, 143.7, 143.2, 138.0, 137.8, 137.0, 133.9, 133.3, 130.6, 130.1, 129.7, 129.1 (2), 128.8 (2), 128.6, 128.5, 128.4 (2), 128.1, 128.0, 127.5, 127.1, 124.8, 124.7, 122.8, 122.7, 121.4 (2), 119.3, 119.0, 114.1, 113.9, 108.1, 107.9, 69.5, 68.7, 64.2, 62.7, 58.1, 57.2, 55.2, 52.1, 49.6, 41.0, 40.6, 26.2 (2), 15.7, 13.7; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{35}H_{31}N_3NaO_3$ 564.2263, mass found 564.2252.

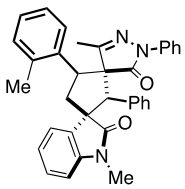
1,3''-dimethyl-1'',2'-diphenyl-4'-(p-tolyl)dispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5ha):



5ha

Following the general procedure, reaction between oxindole-activated spiro-DAC **1h** (0.063 g, 0.24 mmol, 1.5 equiv) and alkylidene pyrazolone **4a** (0.05 g, 0.19 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5ha**, which was purified by silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title compound as mixture of diastereomer (d.r = 1.6:1) in 65% (0.065 g) combined yield. R_f 0.3 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.61-7.57 (m, 5H), 7.39 (d, *J* = 7.3 Hz, 1H), 7.32-7.28 (m, 3H), 7.25-7.16 (m, 6H), 7.11-7.00 (m, 17H), 6.71-6.65 (m, 2H), 5.57 (dd, *J* = 13.7, 5.8 Hz, 0.7H), 4.30 (t, *J* = 13.3 Hz, 1H), 4.14-4.06 (m, 2H), 4.04 (s, 0.7H), 3.16 (s, 2H), 2.92 (s, 3H), 2.84 (t, *J* = 13.3 Hz, 1H), 2.63 (dd, *J* = 12.6, 5.9 Hz, 0.7H), 2.33-2.30 (m, 4H), 2.29 (s, 2H), 2.23 (s, 3H), 2.07 (s, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.6, 176.5, 173.8, 171.1, 160.7, 158.3, 143.7, 143.3, 138.0, 137.9, 137.7, 137.0, 136.8, 135.1, 133.9, 133.3, 133.1, 130.6, 129.7, 129.4, 129.3, 129.2, 128.8, 128.6, 128.5, 128.4 (2), 128.1 (2), 127.9, 126.4, 124.8, 124.7, 122.8, 122.6, 121.4, 119.3, 119.1, 108.1, 107.9, 69.5, 68.7, 64.4, 62.8, 58.1, 57.3, 52.4, 49.8, 40.8, 40.6, 26.3, 26.2, 21.2 (2), 15.7, 13.7; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{35}H_{31}N_3NaO_2$ 548.2314, mass found 548.2290.

1,3''-dimethyl-1'',2'-diphenyl-4'-(o-tolyl)dispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5ia):

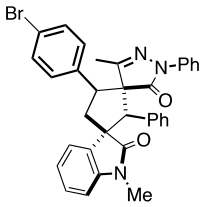


5ia

Following the general procedure, reaction between oxindole-activated spiro-DAC **1i** (0.068 g, 0.26 mmol, 1.5 equiv) and alkylidene pyrazolone **4a** (0.045 g, 0.17 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5ia**, which was purified by silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title compound as mixture of diastereomer (d.r = 1.5:1) in 44% (0.04 g) combined yield. R_f 0.25 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.84-7.82 (m, 1H), 7.68 (d, *J* = 8.3 Hz, 2H), 7.60-7.49 (m, 3H), 7.42 (d, *J* = 7.4 Hz, 1H), 7.33-7.28 (m, 3H), 7.23-7.17 (m, 5H), 7.14-7.02 (m, 16H), 6.72-6.67 (m, 2H), 5.69 (dd, *J* = 13.7, 5.9 Hz, 0.7H), 4.41 (dd, *J* = 13.9, 5.7 Hz, 1H), 4.22-4.14 (m, 2H), 4.12 (s, 0.7H), 3.22 (s, 2H), 3.08 (t, *J* = 13.2 Hz, 0.7H), 2.93 (s, 3H), 2.5.-2.49 (m, 4H), 2.29-2.27 (m, 1H), 2.25-

2.24 (m, 5H), 2.21 (s, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.7, 176.3, 173.1, 171.5, 160.0, 158.5, 143.7, 143.3, 138.4, 138.0, 137.9, 137.1, 135.9, 135.6, 133.8 (2), 133.0, 131.7, 130.7, 130.6, 129.6, 129.4, 128.8, 128.6 (2), 128.5, 128.4, 128.2, 128.1 (2), 127.8, 127.4, 126.7, 126.6, 125.3, 124.9, 124.6, 122.9, 122.6, 121.3, 121.1, 119.3, 118.7, 108.2, 108.0, 68.9, 68.2, 64.3, 63.1, 57.4, 57.2, 48.7, 48.0, 42.7, 42.5, 26.4, 26.3, 20.5, 20.2, 16.4, 14.5; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{35}H_{31}N_3NaO_2$ 548.2314, mass found 548.2295.

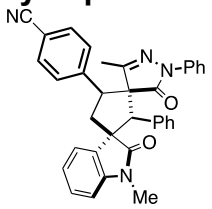
4'-(4-bromophenyl)-1,3''-dimethyl-1'',2'-diphenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5ja):



5ja

Following the general procedure, reaction between oxindole-activated spiro-DAC **1j** (0.094 g, 0.29 mmol, 1.5 equiv) and alkylidene pyrazolone **4a** (0.05 g, 0.19 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5ja**, which was purified by silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title compound as mixture of diastereomer (d.r = 7:1) in 58% (0.065 g) combined yield. R_f 0.4 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.59-7.55 (m, 3H), 7.38 (d, *J* = 8.3 Hz, 2H), 7.32-7.28 (m, 2H), 7.24-7.15 (m, 5H), 7.12-6.99 (m, 7H), 6.71 (d, *J* = 7.7 Hz, 1H), 6.67 (d, *J* = 7.8 Hz, 0.2H), 5.56 (dd, *J* = 13.7, 5.9 Hz, 0.2H), 4.26 (t, *J* = 13.3 Hz, 1H), 4.13 (s, 1H), 4.07– 4.04 (m, 1H), 4.03 (s, 0.2H), 3.16 (s, 0.5H), 2.92 (s, 3H), 2.79 (dd, *J* = 26.4, 13.2 Hz, 0.2H), 2.62 (dd, *J* = 12.6, 5.9 Hz, 0.2H), 2.34-2.29 (m, 4H), 2.07 (s, 0.5H) ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.5, 176.3, 173.5, 170.8, 160.2, 158.0, 143.7, 143.3, 137.8, 137.7, 137.6, 136.7, 134.3, 133.5, 133.0, 131.9, 131.7, 130.3, 129.7 (2), 129.2, 128.9, 128.7, 128.6 (2), 128.4, 128.2 (3), 125.1, 124.9, 122.9, 122.7, 122.1, 121.4, 121.2, 119.3, 119.0, 108.2, 108.0, 69.3, 68.5, 64.5, 62.8, 58.0, 57.2, 51.8, 49.4, 40.6, 40.5, 29.8, 26.3, 15.7; HRMS (ESI-TOF) m/z: $[(M+2)+Na]^+$ calculated for $C_{34}H_{28}BrN_3NaO_2$ 612.1263, mass found 614.1235.

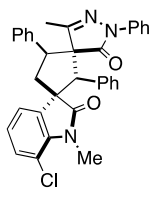
4-(1,3''-dimethyl-2,5''-dioxo-1'',2'-diphenyl-1'',5''-dihydrodispiro[indoline-3,1'-cyclopentane-3',4''-pyrazol]-4'-yl)benzonitrile (5ka):



5ka

Following the general procedure, reaction between oxindole-activated spiro-DAC **1k** (0.080 g, 0.29 mmol, 1.5 equiv) and alkylidene pyrazolone **4a** (0.05 g, 0.19 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5ka**, which was purified by silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title compound as mixture of diastereomer (d.r = 3:1) in 30% (0.031 g) combined yield. R_f 0.25 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.76-7.71 (m, 7H), 7.66-7.64 (m, 3H), 7.56-7.36 (m, 9H), 7.31-7.27 (m, 3H), 7.25 (brs, 1H), 6.90 (d, *J* = 7.8 Hz, 1H), 6.88-6.86 (m, 0.3H), 5.86 (dd, *J* = 13.4, 5.9 Hz, 0.3H), 4.48 (t, *J* = 13.2 Hz, 1H), 4.33-4.28 (m, 2H), 4.20 (s, 0.3H), 3.35 (s, 1H), 3.11 (s, 3H), 3.04-3.98 (s, 0.3H), 2.85 (dd, *J* = 12.5, 5.8 Hz, 0.3H), 2.56-2.51 (m, 4H), 2.23 (s, 1H) ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.4, 176.2, 173.3, 170.6, 159.7, 157.7, 144.0, 143.7, 143.3, 140.8, 137.6, 137.4, 136.4, 133.2, 132.6, 132.4, 130.3, 129.7, 129.2, 129.1, 129.0, 128.9, 128.8, 128.7, 128.6, 128.5, 128.4, 128.2, 127.4, 125.3, 125.1, 123.0, 122.8, 121.4, 121.3, 119.1, 119.0, 118.6, 112.0, 111.4, 108.4, 108.1, 69.4, 68.4, 64.6, 62.8, 60.5, 57.9, 57.2, 51.9, 49.7, 40.3, 40.1, 26.3, 21.2, 15.7, 14.3, 13.7; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{35}H_{28}N_4NaO_2$ 559.2110, mass found 559.2136.

7-chloro-1,3''-dimethyl-1'',2',4'-triphenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5la):

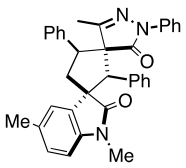


5la

Following the general procedure, reaction between oxindole-activated spiro-DAC **1l** (0.085 g, 0.29 mmol, 1.5 equiv) and alkylidene pyrazolone **4a** (0.052 g, 0.19 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5la**, which was purified by silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title

compound as mixture of diastereomer (d.r = 2.5:1) in 82% (0.085 g) combined yield. R_f 0.3 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.59-7.56 (m, 3H), 7.48 (d, *J* = 7.3 Hz, 1H), 7.37 (d, *J* = 7.6 Hz, 2H), 7.28 (d, *J* = 8.2 Hz, 2H), 7.24-7.15 (m, 7H), 7.13-7.01 (m, 11H), 5.61 (dd, *J* = 13.7, 5.9 Hz, 0.4H), 4.33 (t, *J* = 13.4 Hz, 1H), 4.12-4.04 (m, 2H), 3.98 (s, 0.4H), 3.55 (s, 1H), 3.28 (s, 3H), 2.83 (t, *J* = 13.2 Hz, 0.4H), 2.65 (dd, *J* = 12.7, 6.0 Hz, 0.4H), 2.36 – 2.31 (m, 4H), 2.04 (s, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.7, 176.6, 173.5, 170.8, 160.3, 157.9, 139.7, 139.6,139.3, 138.0, 137.9, 137.7, 134.9, 133.4, 133.3, 132.8, 131.1, 130.8, 129.6, 129.1, 128.8, 128.6 (2), 128.5 (2), 128.4, 128.3, 128.2, 128.1, 127.9, 127.3, 126.4, 124.9, 124.8, 123.5, 123.3, 120.2, 119.3, 119.1, 115.5, 115.3, 69.4, 68.6, 64.7, 63.1, 57.8, 57.1, 52.5, 49.8, 41.0, 40.6, 29.5, 15.6, 13.7; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{34}H_{29}ClN_3O_2$ 546.1948, mass found 546.1941.

1,3'',5-trimethyl-1'',2',4'-triphenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5ma):



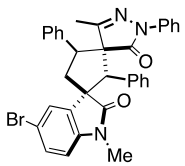
5ma

Following the general procedure, reaction between oxindole-activated spiro-DAC **1m** (0.038 g, 0.14 mmol, 1.5 equiv) and alkylidene pyrazolone **4a** (0.025 g, 0.095 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5ma** as mixture of diastereomer (d.r = 2:1), which was purified by silica gel column chromatography (1:5 EtOAc:Hexane as eluent) to isolate each diastereomer separately. Major diastreomer: 40% yield (0.020 g); Minor diastereomer: 20% (0.010 g).

***Major diastereoisomer*: R_f** 0.25 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.57 (d, *J* = 8.3 Hz, 2H), 7.28-7.22 (m, 6H), 7.19-7.12 (m, 5H), 7.09-7.01 (m, 5H), 6.57 (d, *J* = 7.9 Hz, 1H), 5.61 (dd, *J* = 13.7, 5.9 Hz, 1H), 4.03 (s, 1H), 3.16 (s, 3H), 2.84 (t, *J* = 13.2 Hz, 1H), 2.64 (dd, *J* = 12.6, 6.0 Hz, 1H), 2.39 (s, 3H), 2.07 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.5, 173.6, 160.5, 141.3, 138.2, 137.9, 133.3, 132.0, 130.5, 129.5, 129.0, 128.6, 128.5, 128.0 (2), 127.1, 126.4, 124.7, 122.0, 119.0, 107.8, 68.6, 62.5, 57.9, 49.7, 40.8, 26.2, 21.3, 15.6; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{35}H_{31}N_3NaO_2$ 548.2314, mass found 548.2335.

Minor diastereoisomer: R_f 0.23 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.73 (d, *J* = 8.2 Hz, 2H), 7.56-7.52 (m, 3H), 7.44-7.34 (m, 6H), 7.28-7.22 (m, 7H), 6.78 (d, *J* = 7.8 Hz, 1H), 4.48 (t, *J* = 13.3 Hz, 1H), 4.31-4.26 (m, 2H), 3.08 (s, 3H), 2.64 (s, 3H), 2.51 (s, 3H), 2.49 (dd, *J* = 12.6, 5.9 Hz, 1H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 176.5, 171.0, 158.3, 141.0, 137.9, 137.1, 135.2, 134.0, 132.3, 129.2, 128.7, 128.6 (2), 128.4, 128.1 (2), 128.0, 124.9, 122.2, 119.4, 107.7, 69.5, 64.3, 57.4, 52.6, 40.4, 26.3, 21.6, 13.8; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{35}H_{31}N_3NaO_2$ 548.2314, mass found 548.2332.

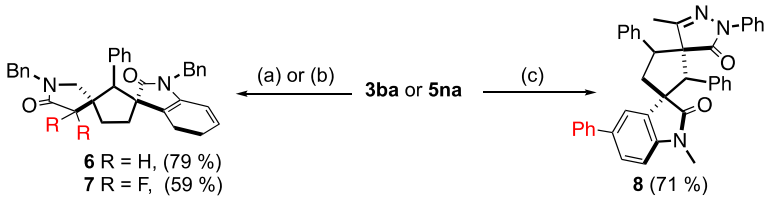
5-bromo-1,3''-dimethyl-1'',2',4'-triphenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (5na):



5na

Following the general procedure, reaction between oxindole-activated spiro-DAC **1n** (0.074 g, 0.23 mmol, 1.5 equiv) and alkylidene pyrazolone **4a** (0.04 g, 0.15 mmol, 1.0 equiv) afforded bis-spirocyclopentane oxindole derivative **5na**, which was purified by silica gel column chromatography (1:4 EtOAc:Hexane as eluent) to give the title compound as mixture of diastereomer (d.r = 2.4:1) in 63% (0.056 g) combined yield. R_f 0.25 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.64 (brs, 1H), 7.58-7.54 (m, 3H), 7.45-7.42 (m, 1H), 7.38-7.33 (m, 3H), 7.27 (brs, 1H), 7.23-7.17 (m, 7H), 7.13-7.04 (m, 9H), 6.59 (d, *J* = 8.2 Hz, 1H), 6.58 (d, *J* = 8.2 Hz, 0.4H), 5.59 (dd, *J* = 13.6, 5.8 Hz, 0.4H), 4.32 (t, *J* = 13.4 Hz, 1H), 4.15-4.02 (m, 2H), 3.96 (s, 0.4H), 3.14 (s, 1.2H), 2.89 (s, 3H), 2.87-2.80 (m, 0.4H), 2.65 (dd, *J* = 12.7, 5.9 Hz, 0.4H), 2.34-2.31 (m, 4H), 2.04 (s, 1.2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 177.0, 175.9, 173.6, 170.8, 160.3, 158.0, 142.8, 142.4, 138.9, 137.9, 137.7, 134.8, 133.4, 132.8, 132.7, 131.7, 131.4,129.9, 129.6, 129.2, 129.1, 128.8, 128.7, 128.6 (2), 128.5, 128.3, 128.2 (2), 128.0, 127.4, 126.5, 125.0, 124.9, 124.7 (2), 119.4, 119.1, 115.1 (2), 109.6, 109.4, 69.4, 68.6, 64.4, 62.8, 58.1, 57.5, 52.6, 49.9, 40.7, 40.1, 26.4 (2), 15.7, 13.8; HRMS (ESI-TOF) m/z: $[(M+2)+Na]^+$ calculated for $C_{34}H_{28}BrN_3NaO_2$ 614.1242, mass found 614.1234.

8. Follow-up Chemistry:

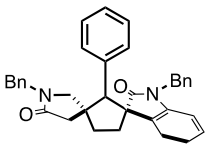


6 R = H, (79 %)
7 R = F, (59 %)

8 (71 %)

Scheme 3 (*from main text*): Follow up chemistry. Reaction conditions: (a) $NH_2NH_2.H_2O$ (30 equiv.), KOH, EtOH-H_2O, reflux, 6h; (b) DAST (3 equiv.), DCM, 40 ^oC, 12h; (c) $PhB(OH)_2$ (1.5 equiv.), $Pd(PPh_3)_4$ (5 mol %), K_3PO_4 (2.5 equiv.), toluene-H_2O, 80 ^oC, 12h.

1,1''-dibenzyl-2'-phenyldispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,4'',5''-trione (6):

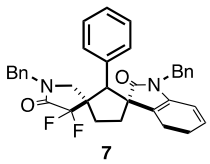


6

To a solution of compound **3ba** (0.045 g, 0.085 mmol, 1.0 equiv) in EtOH:H_2O (1 mL, 4:1) was added hydrazine hydrate (0.082 g, 25.66 mmol, 30 equiv) and potassium hydroxide (0.007 g, 0.12 mmol, 1.5 equiv). The reaction mixture was stirred at 100 °C temperature for 6 h. Upon completion of the reaction, the solvent was removed in vacuo and the crude product was purified by flash silica gel column chromatography (using 1:4 EtOAc:Hexane as eluent) to obtain compound **6** as colorless oil in 79% (0.034g) yield. R_f 0.4 (3:7 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.41-7.35 (m, 5H), 7.30-7.24 (m, 5H), 7.19-7.11 (m, 6H), 6.94 (d, *J* = 6.9 Hz, 1H), 6.56-6.54 (m, 1H), 5.08 (d, *J* = 15.9 Hz, 1H), 4.74 (d, *J* = 14.8 Hz, 1H), 4.63 (d, *J* = 15.9 Hz, 1H), 4.02 (dd, *J* = 22.7, 12.8 Hz, 2H), 3.60 (d, *J* = 10.7 Hz, 1H), 3.54 (s, 1H), 2.65-2.64 (m, 2H), 2.60-2.55 (m, 1H), 2.47-2.39 (m, 1H), 2.27-2.20 (m, 2H).

^{13}C NMR (100 MHz, $CDCl_3$) δ 179.9, 173.0, 142.4, 136.6, 135.4, 135.1, 133.8, 130.1, 128.8, 128.7, 128.6, 128.1, 128.0, 127.7, 127.5, 127.4, 126.9, 123.0, 121.8, 109.0, 64.9, 59.2, 55.2, 48.0, 16.6, 44.6, 43.7, 40.7, 36.5; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{35}H_{33}N_2O_2$ 513.2542, mass found 513.2522.

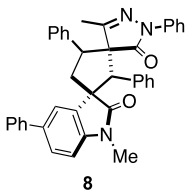
1,1''-dibenzyl-4',4'-difluoro-2'-phenyldispiro[indoline-3,1'-cyclopentane-3',3''-pyrrolidine]-2,5''-dione (7):



7

To a solution of compound **3ba** (0.065 g, 0.12 mmol, 1.0 equiv) in dry DCM (4 mL) was added DAST (0.06 g, 0.36 mmol, 3.0 equiv) at 0 °C. The reaction mixture was then stirred at 40 °C for 12 h. Upon completion of the reaction (TLC controlled), the crude was purified by flash silica gel column chromatography (using 1:3 EtOAc:Hexane as eluent) to obtain compound **7** as colorless oil in 59% (0.040 g) yield. R_f 0.4 (3:7 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.53 (d, *J* = 7.1 Hz, 1H), 7.38-7.29 m, 3H), 7.23 (d, *J* = 7.4 Hz, 3H), 7.15-7.05 (m, 6H), 6.91 (d, *J* = 7.7 Hz, 2H), 6.46 (d, *J* = 7.6 Hz, 2H), 6.39 (d, *J* = 7.7 Hz, 1H), 5.81-5.70 (m, 1H), 4.88 (d, *J* = 16.0 Hz, 1H), 4.61 (d, *J* = 14.9 Hz, 1H), 4.51 (d, *J* = 14.9 Hz, 1H), 4.31 (d, *J* = 16.0 Hz, 1H), 3.62-3.49 (m, 2H), 2.69-2.62 (m, 1H), 2.55-2.47 (m, 1H), 2.17-2.03 (m, 2H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 175.43 (d, *J* = 11.6 Hz), 163.22 (d, *J* = 30.5 Hz), 148.24 (d, *J* = 271.1 Hz), 143.4, 136.5, 134.87 (t, *J* = 10.6 Hz), 129.3, 129.0, 129.0, 128.7, 128.4, 128.1, 128.0, 127.4, 127.2, 127.1, 126.6, 126.3, 125.46 (d, *J* = 2.5 Hz), 123.0, 109.7, 97.33 (d, *J* = 182.9 Hz), 58.63 (d, *J* = 24.0 Hz), 47.60 (d, *J* = 4.9 Hz), 46.8, 43.5, 32.6, 30.5, 20.2; ^{19}F NMR (376 MHz, $CDCl_3$) δ -144.1, -187.3; HRMS (ESI-TOF) m/z: $[M+H]^+$ calculated for $C_{35}H_{31}F_2N_2O_2$ 549.2354, mass found 549.2329.

1,3''-dimethyl-1'',2',4',5-tetraphenyldispiro[indoline-3,1'-cyclopentane-3',4''-pyrazole]-2,5''(1''H)-dione (8):

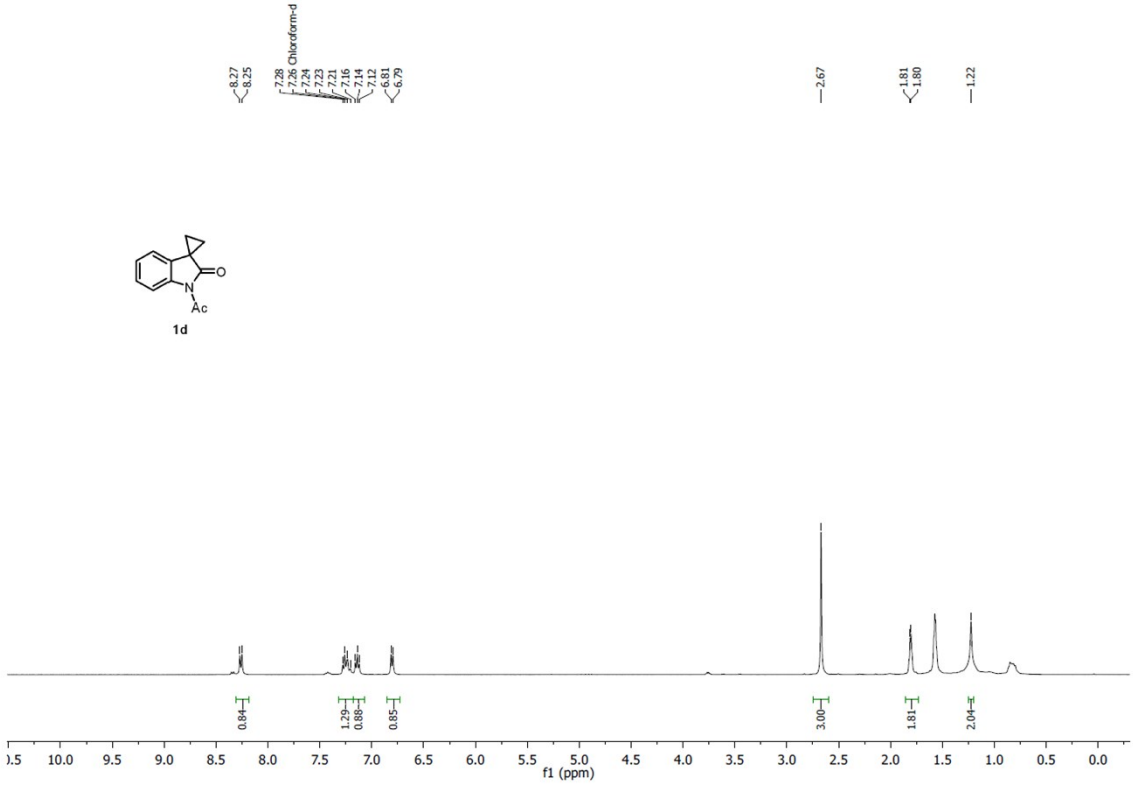


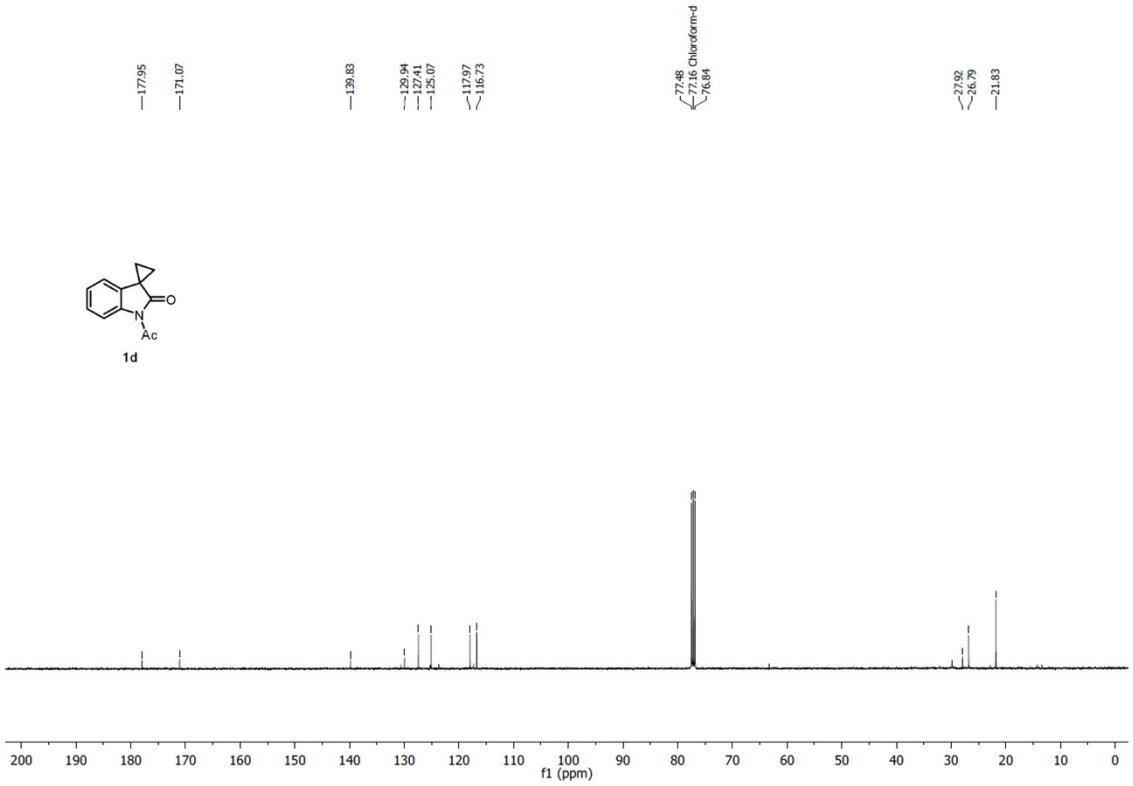
8

To a flame dried Schlenk tube were successively added compound **5na**-major diastereomer (0.025 g, 0.04 mmol, 1.0 equiv), phenylboronic acid (0.007g, 0.06 mmol, 1.5 equiv), $Pd(PPh_3)_4$ (0.002 mg, 0.002 mmol, 0.05 equiv), K_3PO_4 (0.022 g, 0.10 mmol, 2.5 equiv) in a mixture of toluene (0.6 mL) and water (0.2 mL) as solvent.

The reaction mixture was first degassed *via* purging argon (ca. 5 minutes) and then heated at room 80 °C for 12 h. The reaction mixture was passed through celite and the filtrate was concentrated under vacuo. The crude product was purified by flash silica gel column chromatography (using 1:4 EtOAc:Hexane as eluent) to obtain compound **8** as colorless oil in 71% (0.017g) yield as single diastereomer. R_f 0.3 (1:4 EtOAc:Hexane); ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.75 (brs, 1H), 7.64 (d, *J* = 7.7 Hz, 1H), 7.57-7.50 (m, 5H), 7.42-7.38 (m, 3H), 7.28-7.17 (m, 5H), 7.12-7.06 (m, 6H), 6.78 (d, *J* = 8.0 Hz, 1H), 4.40-4.43 (m, 1H), 4.20-4.12 (m, 2H), 2.96 (s, 3H), 2.41 (dd, *J* = 12.7, 5.9 Hz, 1H), 2.34 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) δ 176.5, 170.9, 158.2, 142.8, 141.3, 137.8, 137.6, 136.4, 135.0, 133.8, 129.2, 129.1, 128.6 (2), 128.5, 128.2, 128.1, 128.0, 127.5, 127.3, 127.1, 124.9, 120.3, 119.4, 108.1, 69.5, 64.5, 57.5, 52.6, 40.2, 26.4, 13.9; HRMS (ESI-TOF) m/z: $[M+Na]^+$ calculated for $C_{40}H_{33}N_3NaO_2$ 610.2470, mass found 610.2448.

9. NMR Spectra of New Compounds



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Chloroform-d
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7.25
7.23
7.21
7.20
7.16
7.13
7.11
7.02
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6.92
6.90
6.79
6.77
6.54
6.52
6.50
5.77
5.75
3.66
3.49
3.35
3.33
3.31
3.12
3.10
3.07
2.41
2.40
2.39
2.38
2.21
2.19
2.18
2.17
2.04
2.03
2.02
2.01
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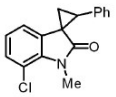
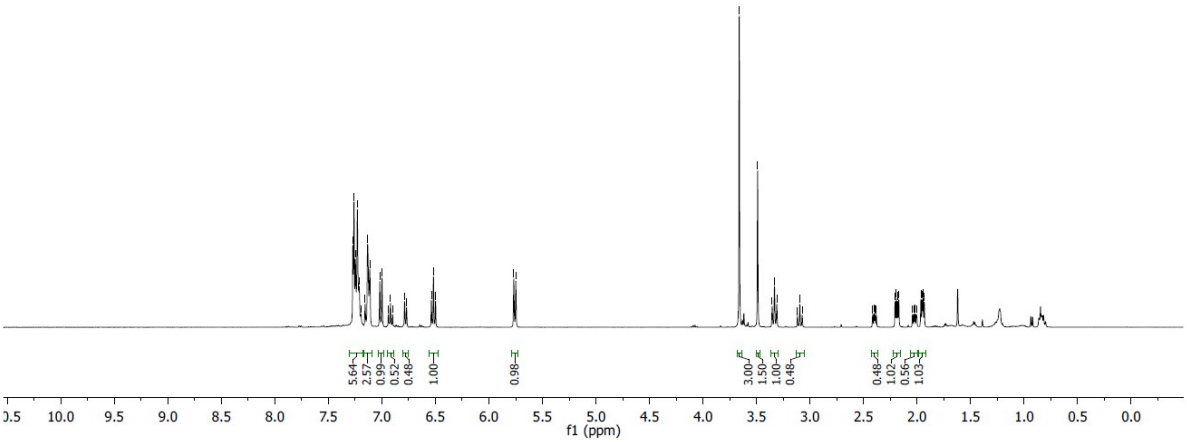
1.95
1.94
1.93


Ph
O
N
Me
Cl

1l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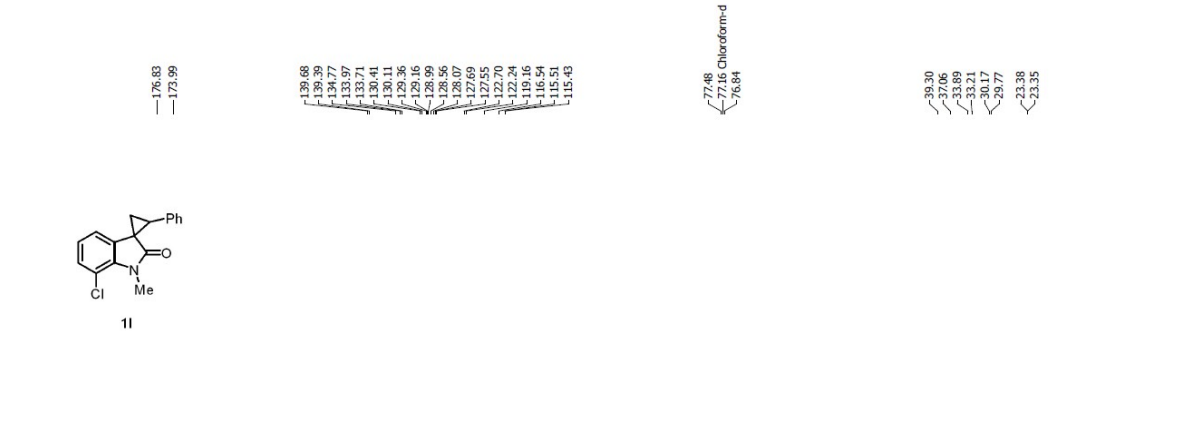
f1 (ppm)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128.07
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127.55
122.70
122.24
119.16
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115.51
115.43
77.48
77.16
Chloroform-d
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37.06
33.89
33.21
30.17
29.77
23.38
23.35
f1 (ppm)

Ph
O
N
Me
Cl

1l


7.32
7.30
Chloroform-d
7.26
7.24
7.22
7.18
7.16
7.07
7.05
6.94
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6.78
6.76
6.74
6.72
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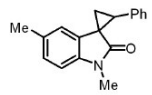
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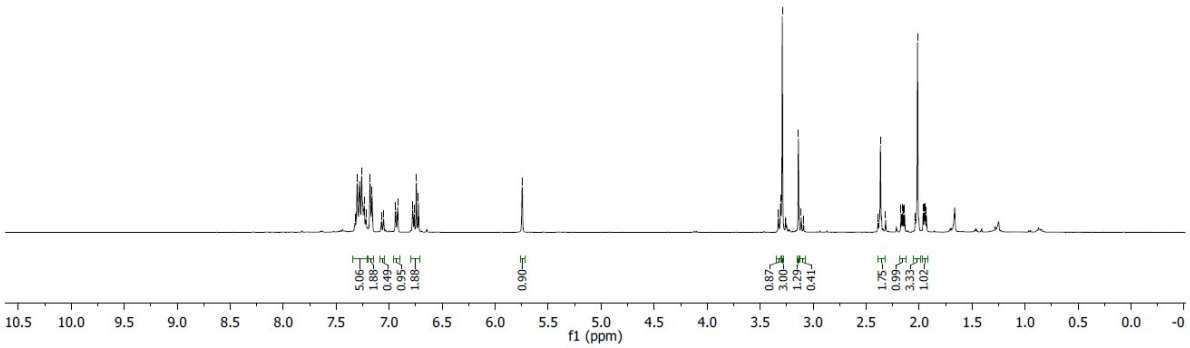
N
Me

1m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0.87
3.00
1.29
0.41
1.75
0.99
3.33
1.02
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
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1.0
0.5
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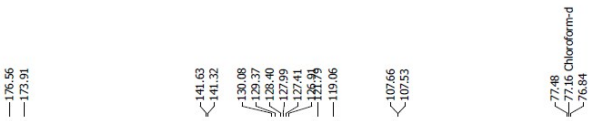
-0
f1 (ppm)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119.06
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107.53
77.48
77.16
Chloroform-d
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38.11
35.77
33.98
33.35
26.77
26.59
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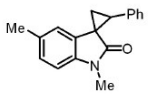
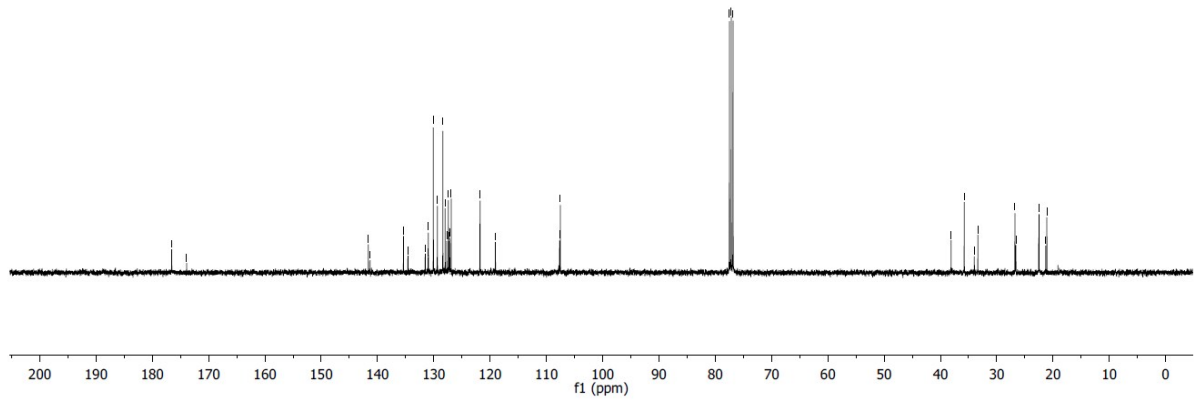
22.49
21.31
21.09


Me
Ph
O
N
Me

1m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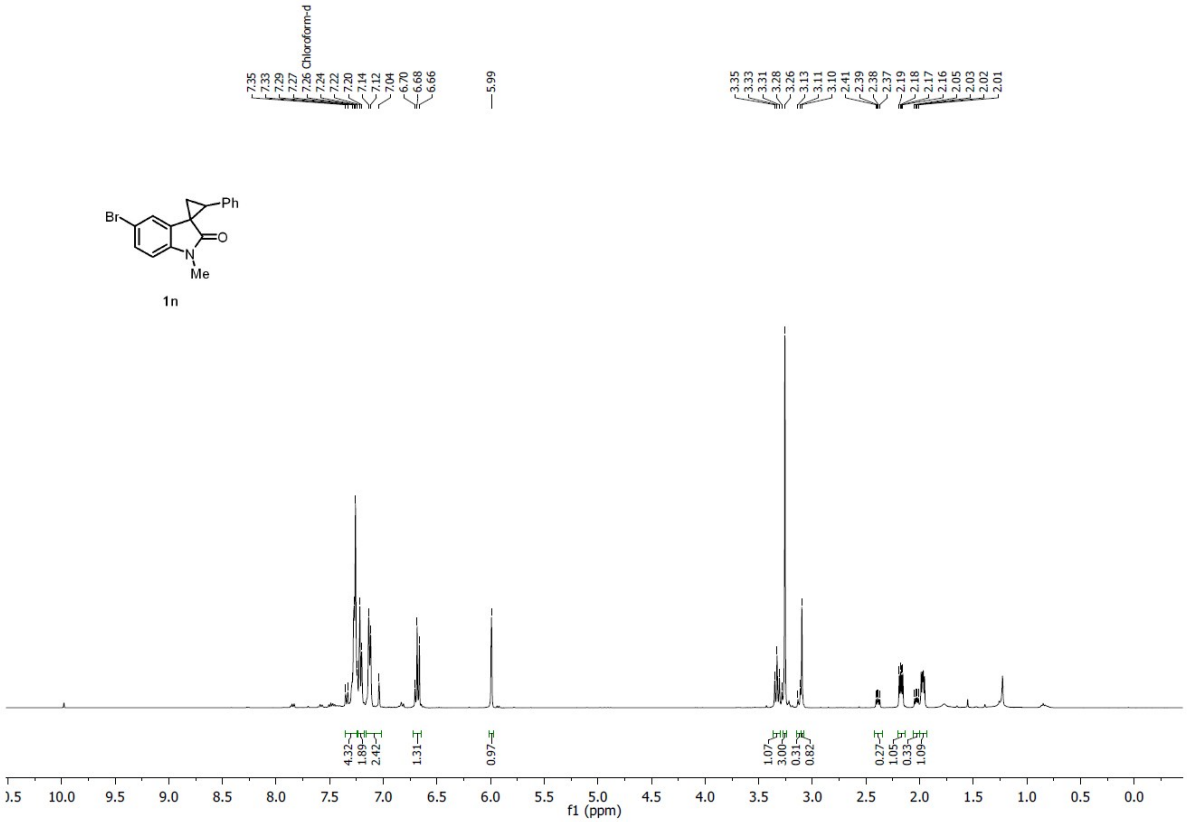
f1 (ppm)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7.27
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Ph
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N
Me
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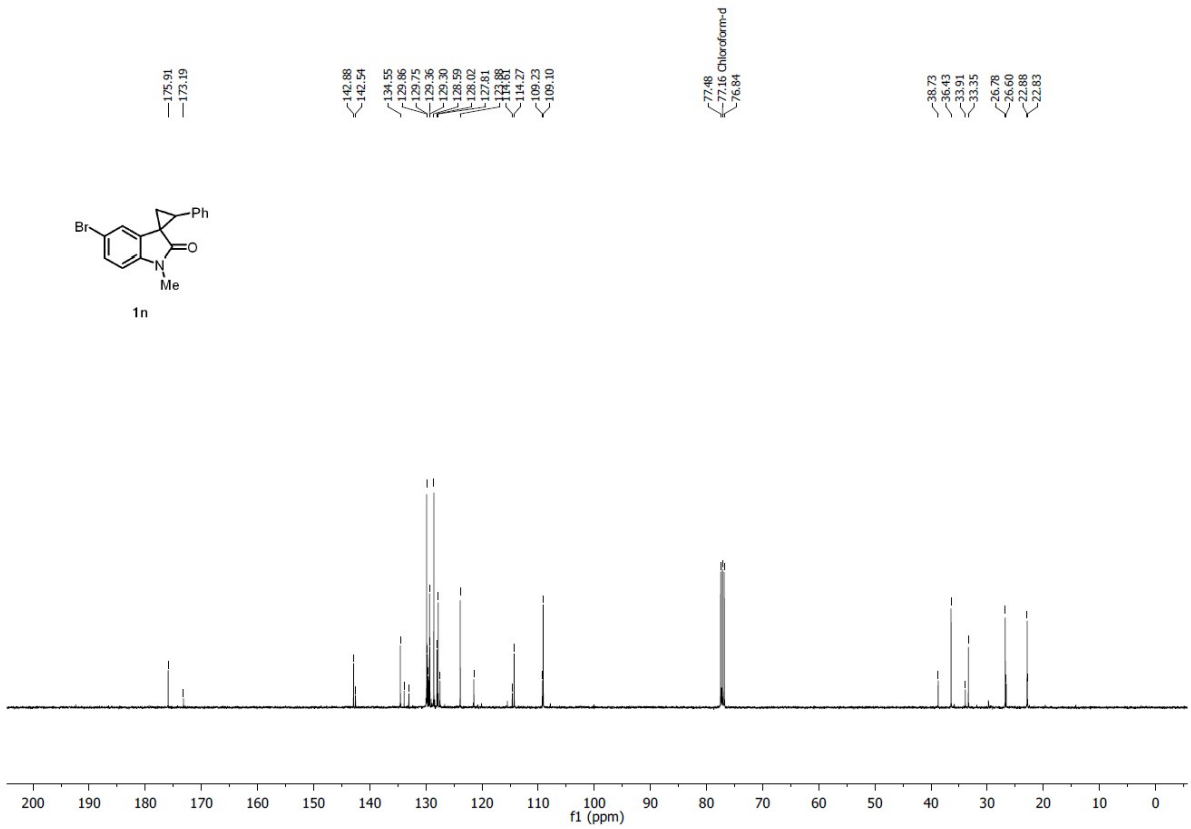
f1 (ppm)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128.59
128.02
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109.23
109.10
77.48
77.16
Chloroform-d
76.84
38.73
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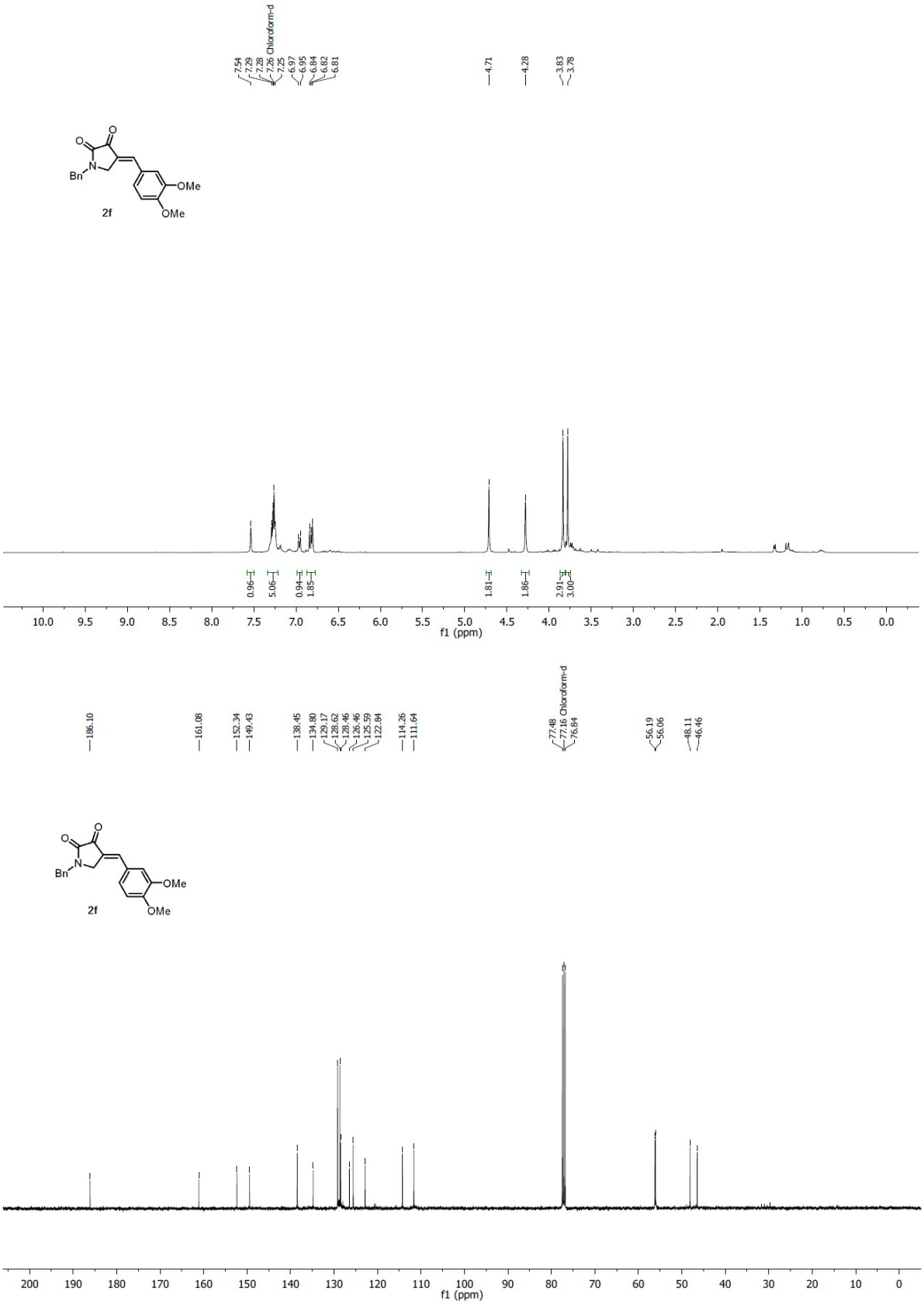
26.78
26.60
22.88
22.83
Br
Ph
O
N
Me
1n
f1 (ppm)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7.26 Chloroform-d
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6.81
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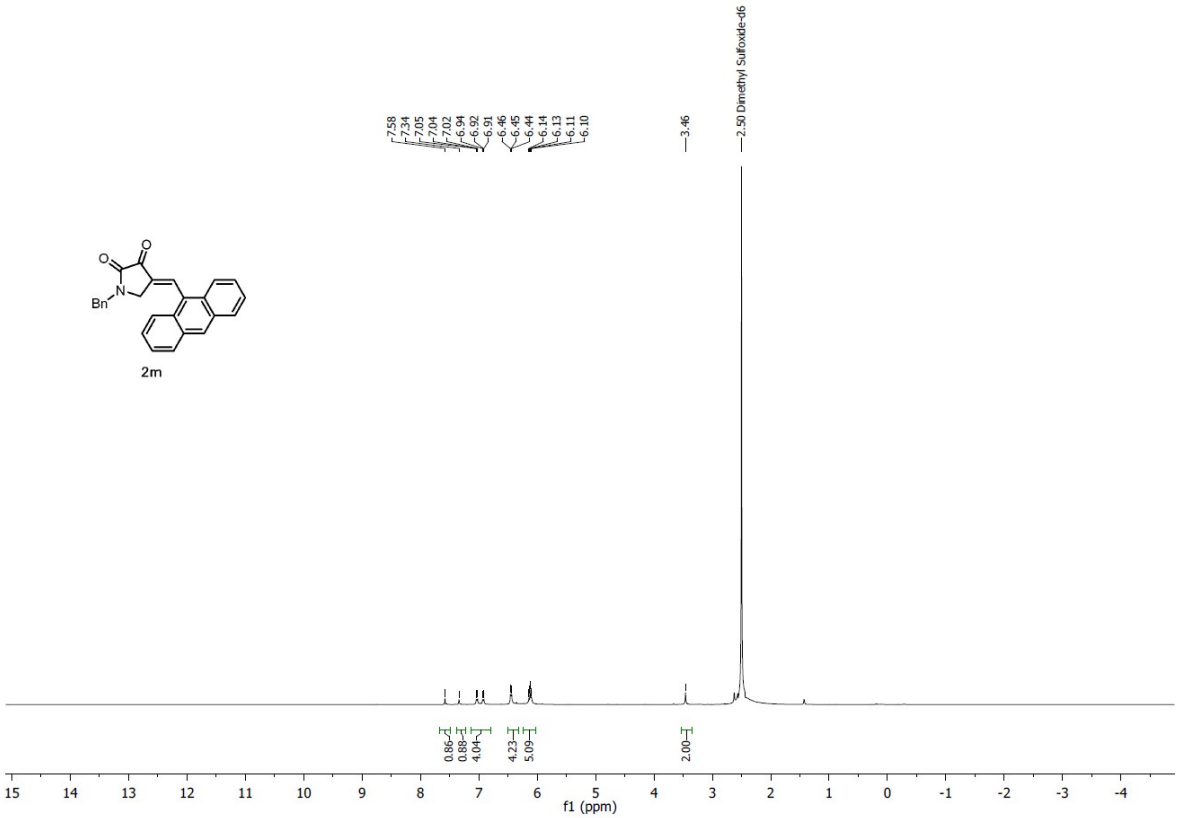
3.83
3.78
O
O
N
Bn
OMe
OMe
2f
0.96
5.06
0.94
1.85
1.81
1.86
2.91
3.00
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)
186.10
161.08
152.34
149.43
138.45
134.80
129.17
128.62
128.46
126.46
125.59
122.84
114.26
111.64
77.48
77.16 Chloroform-d
76.84
56.19
56.06
48.11
46.46
O
O
N
Bn
OMe
OMe
2f
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)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7.34
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7.04
7.02
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6.92
6.91
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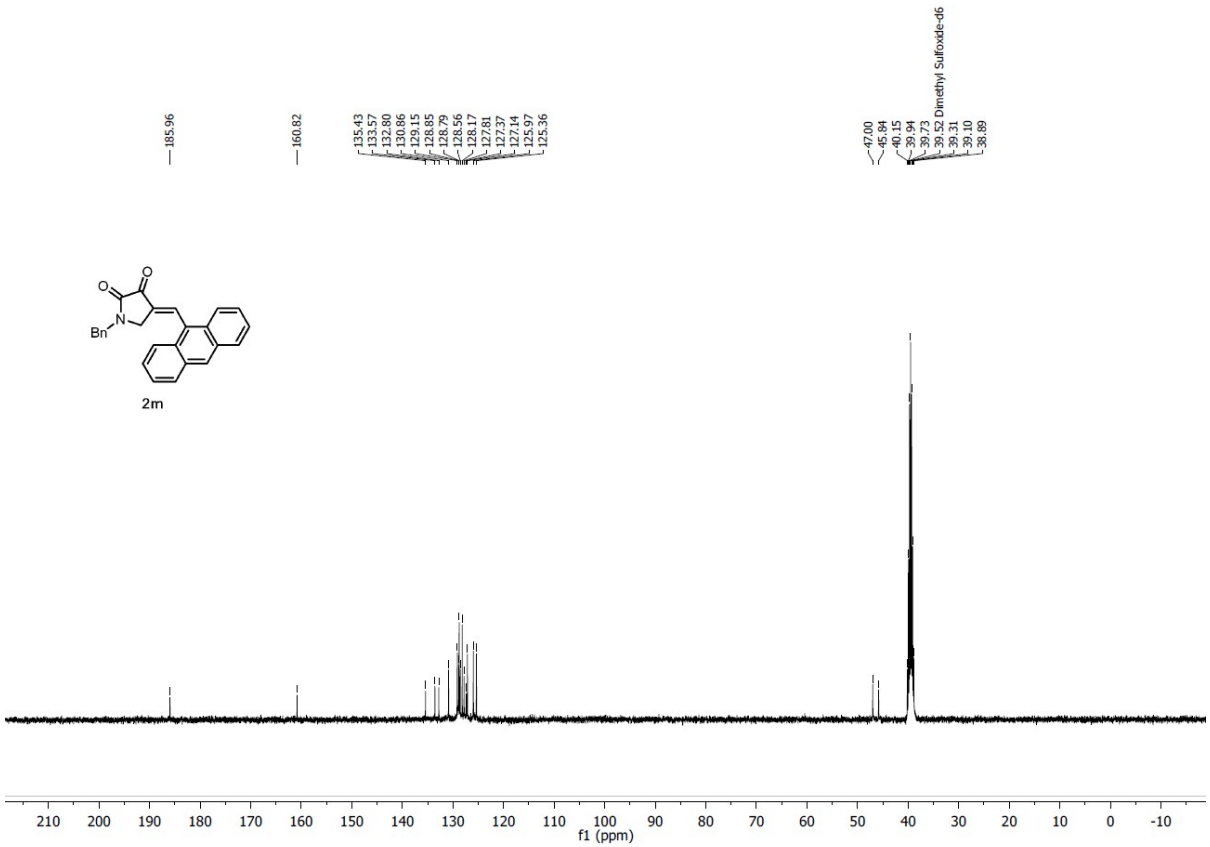
6.45
6.44
6.14
6.13
6.11
6.10
3.46
2.50 Dimethyl Sulfoxide-d6
Bn
2m
0.86
0.88
4.04
4.23
5.09
2.00
f1 (ppm)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127.37
127.14
125.97
125.36
47.00
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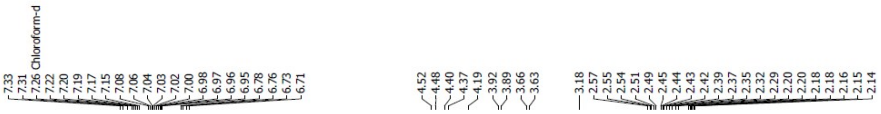
39.73
39.52 Dimethyl Sulfoxide-d6
39.31
39.10
38.89
Bn
2m
f1 (ppm)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7.04
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7.02
7.00
6.98
6.97
6.96
6.95
6.78
6.76
6.73
6.71
4.52
4.48
4.40
4.37
4.19
3.92
3.89
3.66
3.63
3.18
2.57
2.55
2.54
2.51
2.49
2.45
2.44
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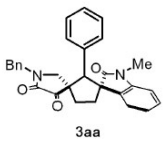
2.14


Bn
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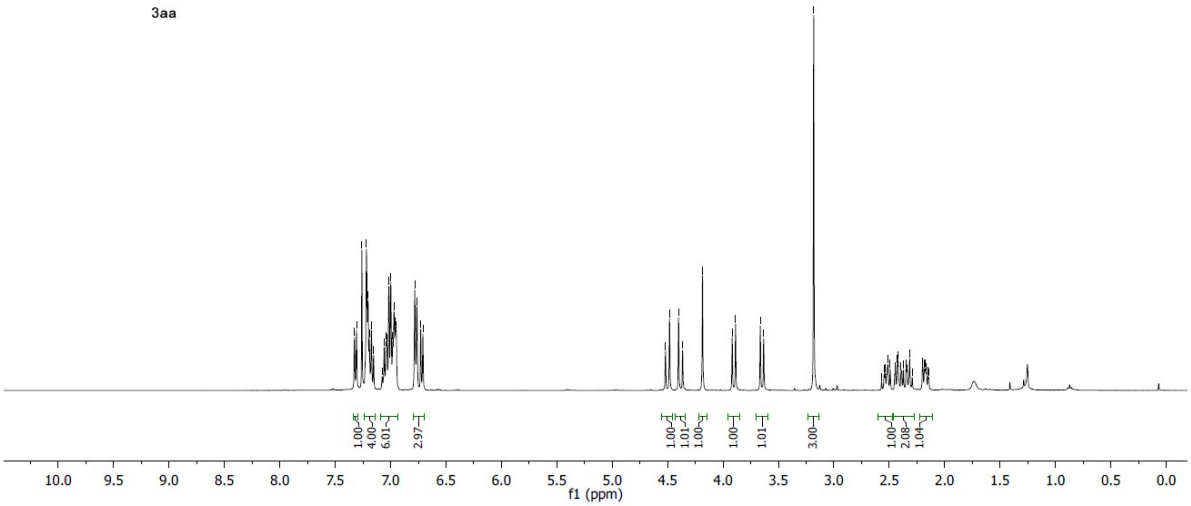
O
O
O
N
Me

3aa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1.01
1.00
1.00
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3.00
1.00
2.08
1.04
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
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4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
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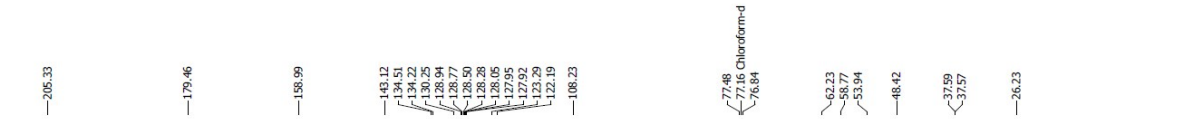
0.0
f1 (ppm)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128.50
128.28
128.05
127.95
127.92
123.29
122.19
108.23
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77.16 Chloroform-d
76.84
62.23
58.77
53.94
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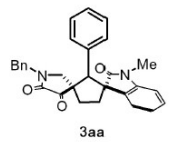
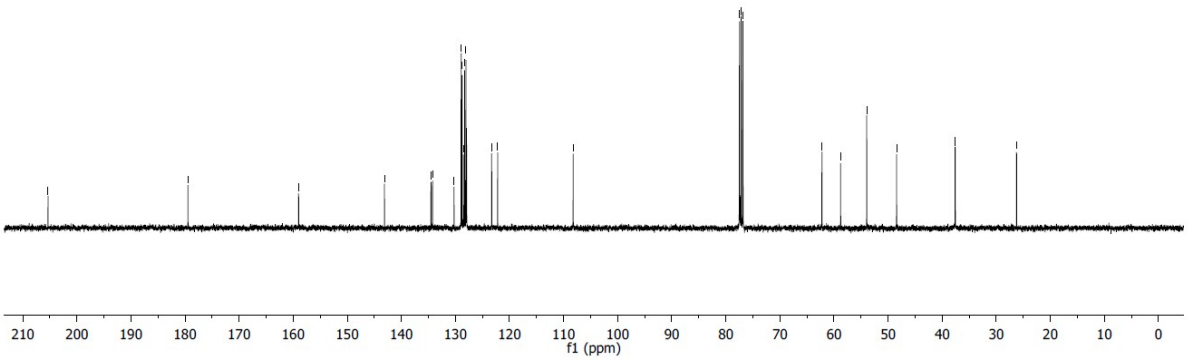
37.59
37.57
26.23


Bn
N
O
O
O
N
Me

3aa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180
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120
110
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90
80
70
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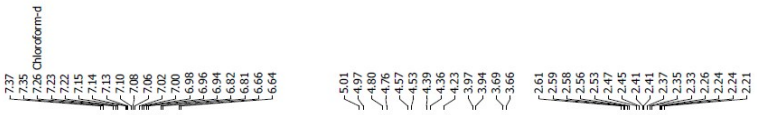
50
40
30
20
10
0
f1 (ppm)


7.37
7.35
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7.23
7.22
7.15
7.14
7.13
7.10
7.08
7.06
7.02
7.00
6.98
6.96
6.94
6.82
6.81
6.66
6.64
5.01
4.97
4.80
4.76
4.57
4.53
4.39
4.36
4.23
3.97
3.94
3.69
3.66
2.61
2.59
2.58
2.56
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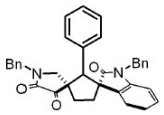
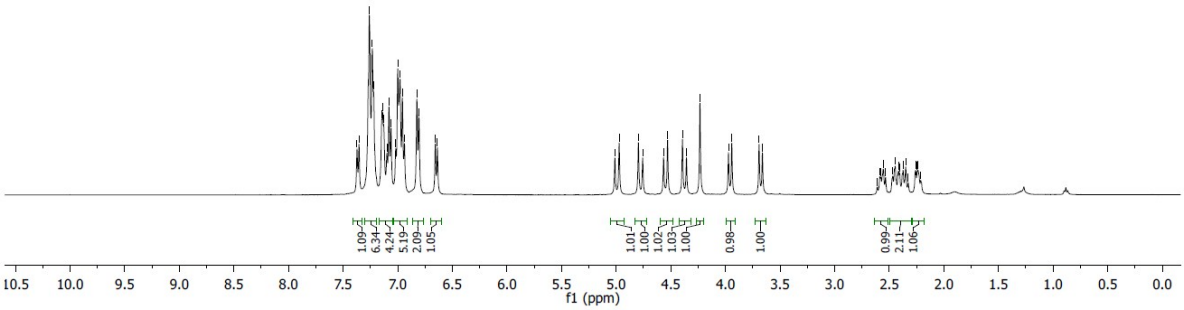
2.33
2.26
2.24
2.24
2.21


Bn
N
O
O
O
N
Bn

3ba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5.19
2.09
1.05
1.01
1.00
1.02
1.03
1.00
0.98
1.00
0.99
2.11
1.06
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


205.05
179.32
158.91
142.35
128.89
128.81
128.72
128.56
128.01
127.92
127.90
127.69
127.27
123.25
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76.84
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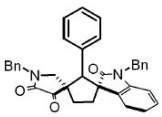
58.92
53.94
53.77
48.33
43.69
37.94
37.59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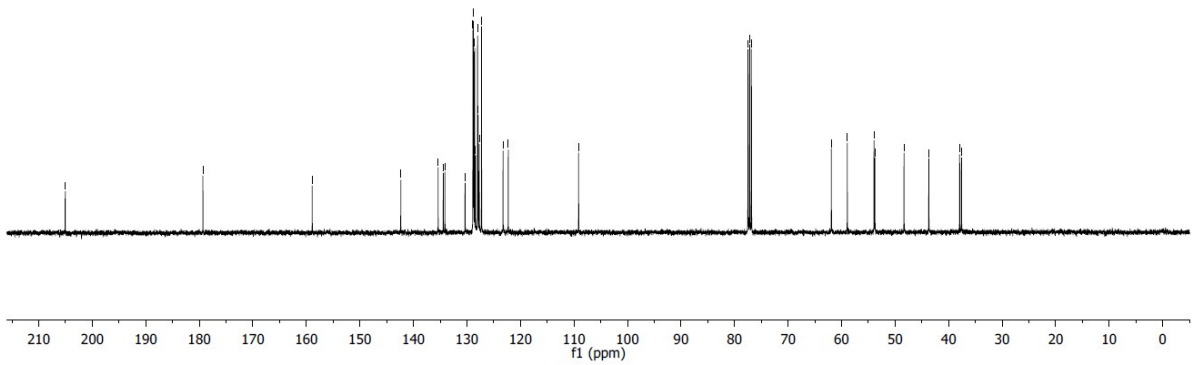
N
O
O
O
N
Bn

3ba


210
200
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180
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150
140
130
120
110
100
90
80
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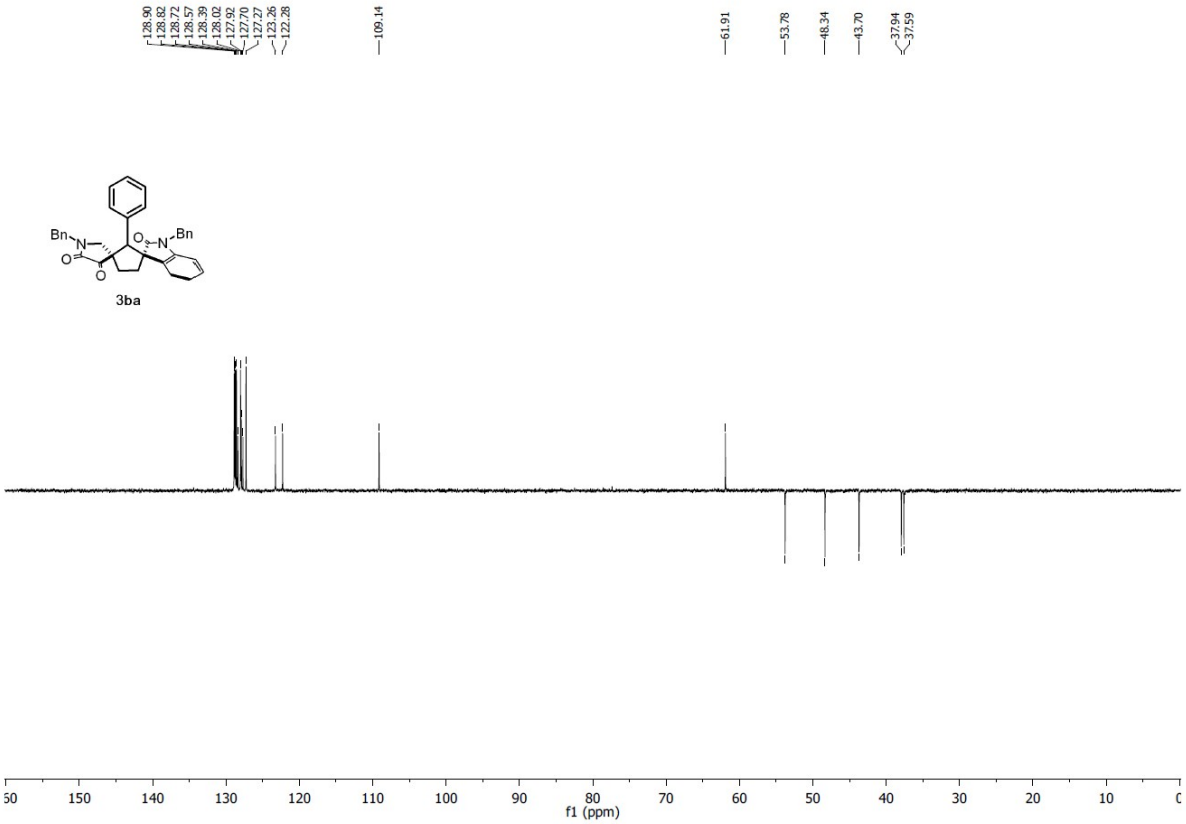
50
40
30
20
10
0
f1 (ppm)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128.82
128.72
128.57
128.39
128.02
127.92
127.70
127.27
123.26
122.28
109.14
61.91
53.78
48.34
43.70
37.94
37.59
Bn
N
O
O
O
N
Bn
3ba
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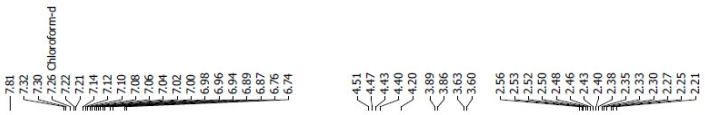
140
130
120
110
100
90
80
70
60
50
40
30
20
10
f1 (ppm)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7.30
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7.12
7.10
7.08
7.06
7.04
7.02
7.00
6.98
6.96
6.94
6.89
6.87
6.76
6.74
4.51
4.47
4.43
4.40
4.20
3.89
3.86
3.63
3.60
2.56
2.53
2.52
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2.48
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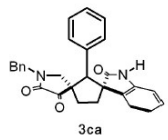
2.38
2.35
2.33
2.30
2.27
2.25
2.21


Bn
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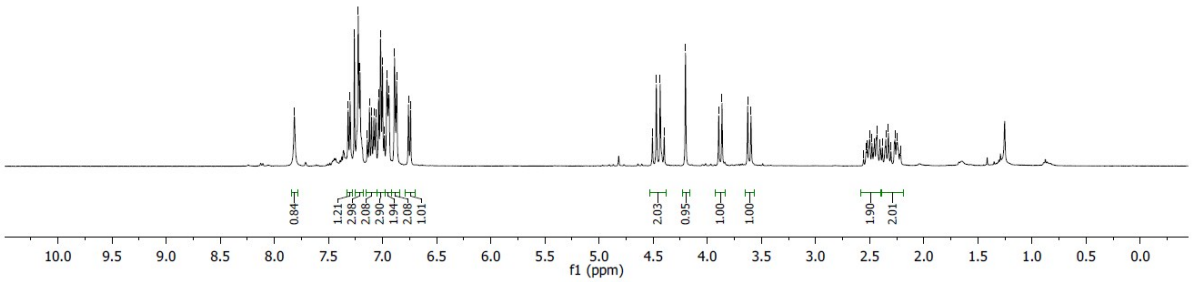
O
O
N
H

3ca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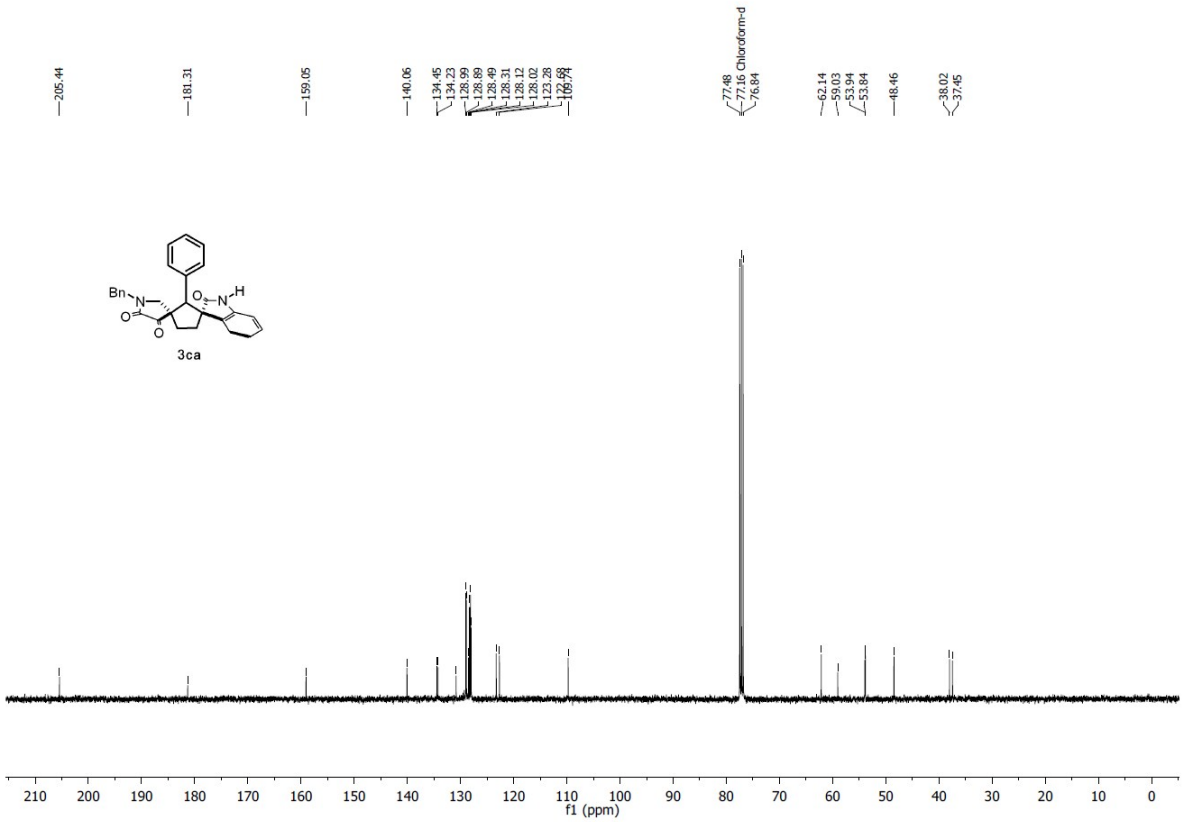
2.03
0.95
1.00
1.00
1.90
2.01
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)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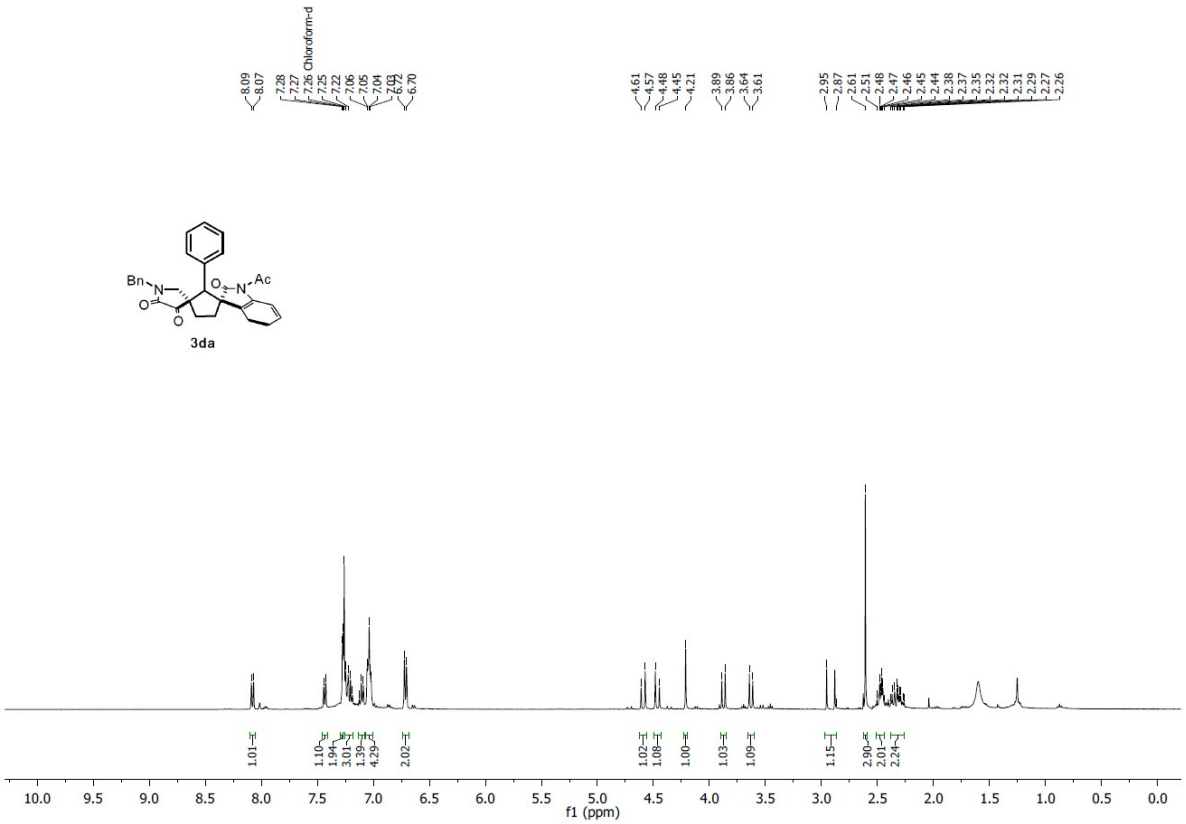
134.23
128.99
128.89
128.49
128.31
128.12
128.02
123.28
77.48
77.16 Chloroform-d
76.84
62.14
59.03
53.94
53.84
48.46
38.02
37.45
210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

Bn
N
O
O
O
N
H

3ca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Chloroform-d
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7.22
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7.05
7.04
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4.57
4.48
4.45
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3.89
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3.64
3.61
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2.87
2.61
2.51
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2.47
2.46
2.45
2.44
2.38
2.37
2.35
2.32
2.32
2.31
2.29
2.27
2.26
Bn
N
O
O
O
N
Ac
3da
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1.10
1.94
3.01
1.39
4.29
2.02
1.02
1.08
1.00
1.03
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1.15
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8.0
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6.0
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2.5
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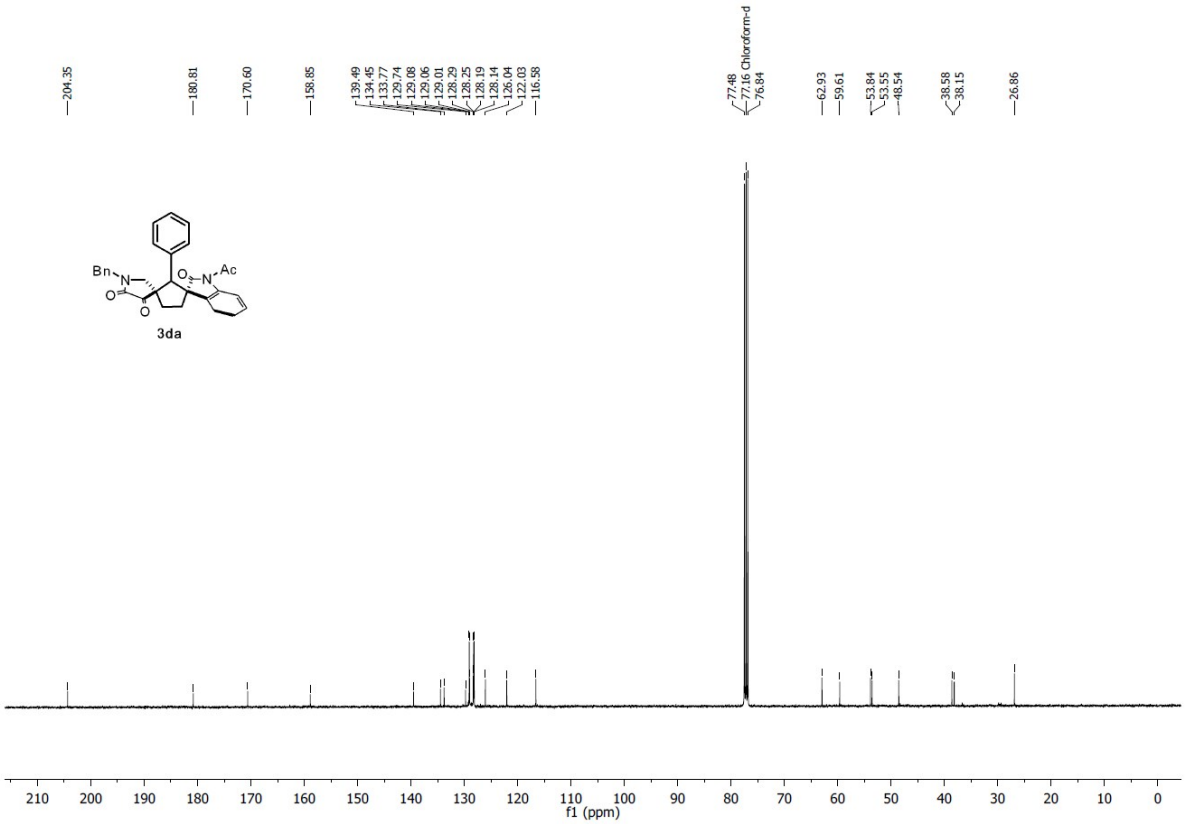
0.0
f1 (ppm)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128.19
128.14
126.04
122.03
116.58
77.48
77.16
Chloroform-d
76.84
62.93
59.61
53.84
53.55
48.54
38.58
38.15
26.86
Bn
N
O
O
O
N
Ac
3da
210
200
190
180
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150
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130
120
110
100
90
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70
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50
40
30
20
10
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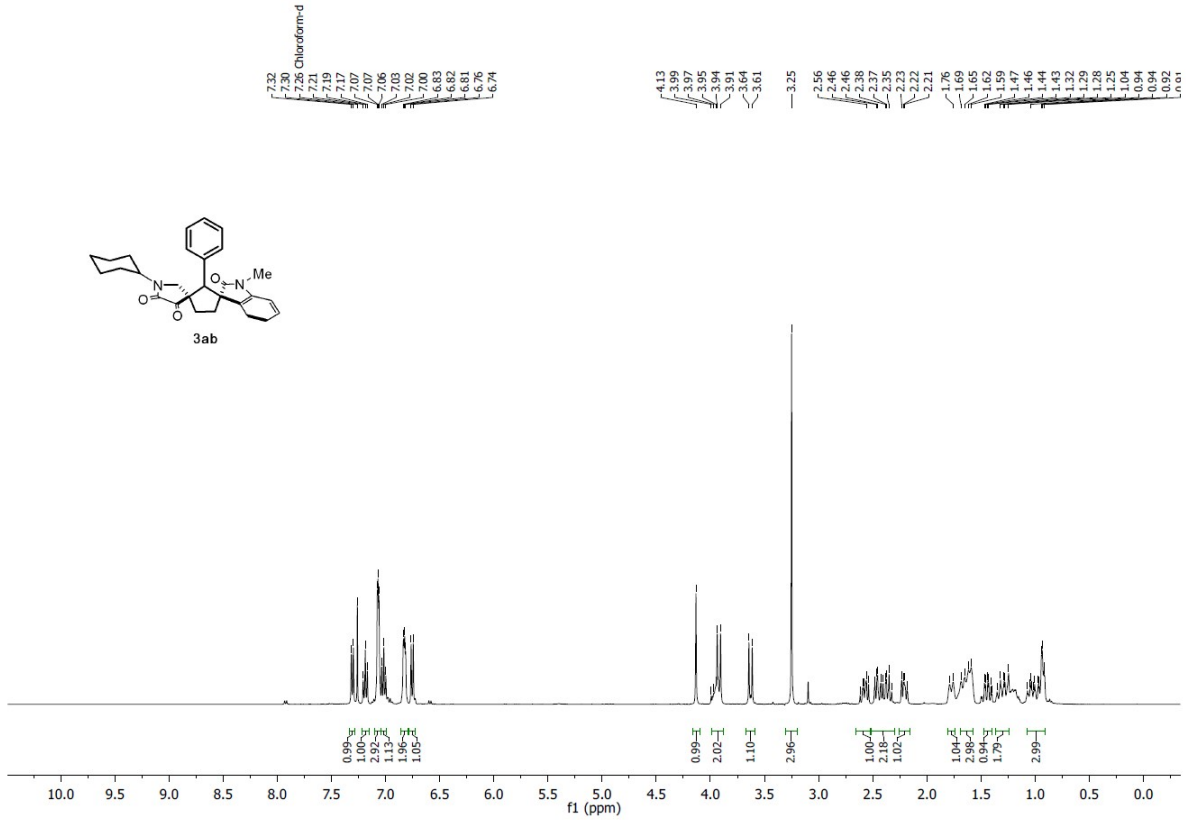
f1 (ppm)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7.06
7.03
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7.00
6.83
6.82
6.81
6.76
6.74
4.13
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3.97
3.95
3.94
3.91
3.64
3.61
3.25
2.56
2.46
2.46
2.38
2.37
2.35
2.23
2.22
2.21
1.76
1.69
1.65
1.62
1.59
1.47
1.46
1.44
1.43
1.32
1.29
1.28
1.25
1.04
0.94
0.94
0.92
Me
O
N
O
O
3ab
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1.13
1.96
1.05
0.99
2.02
1.10
2.96
1.00
2.18
1.02
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2.98
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1.79
2.99
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9.0
8.5
8.0
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6.5
6.0
5.5
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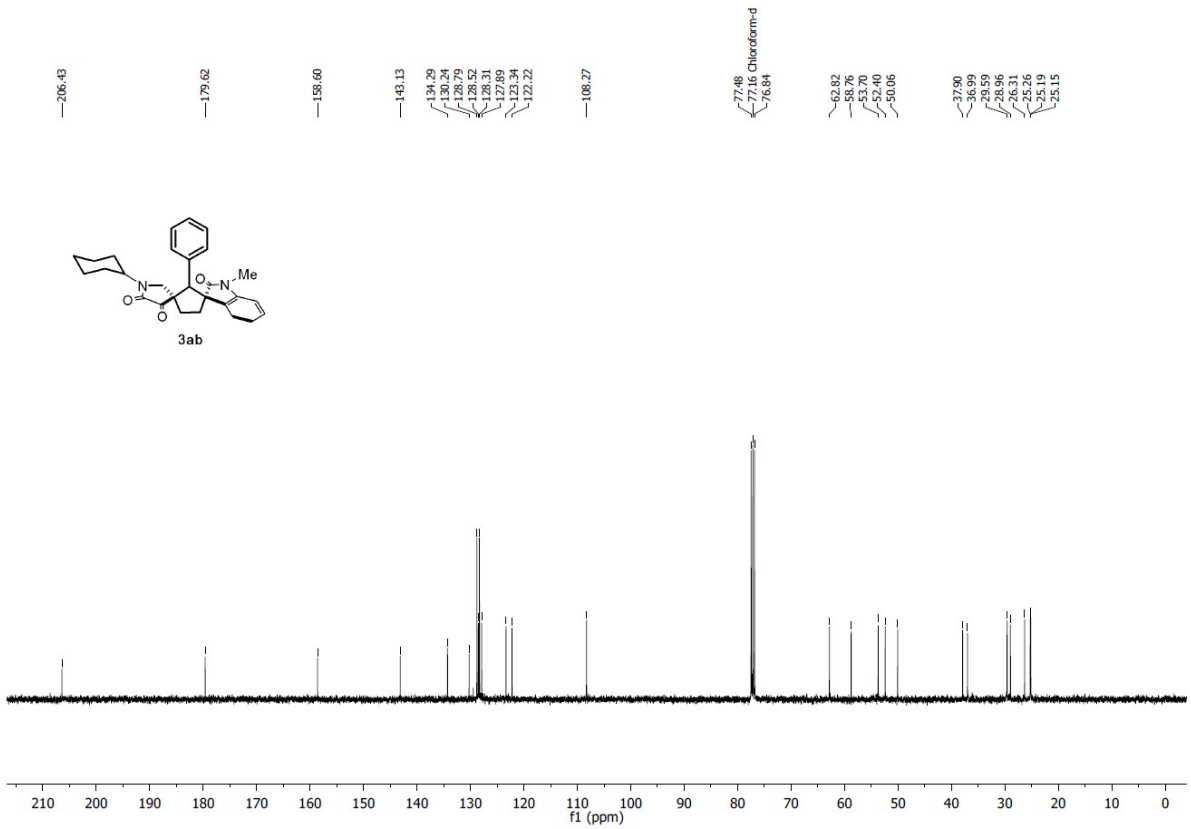
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)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127.89
123.34
122.22
108.27
77.48
77.16
Chloroform-d
76.84
62.82
58.76
53.70
52.40
50.06
37.90
36.99
29.59
28.96
26.31
25.26
25.19
25.15
Me
O
N
O
O
3ab
210
200
190
180
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120
110
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90
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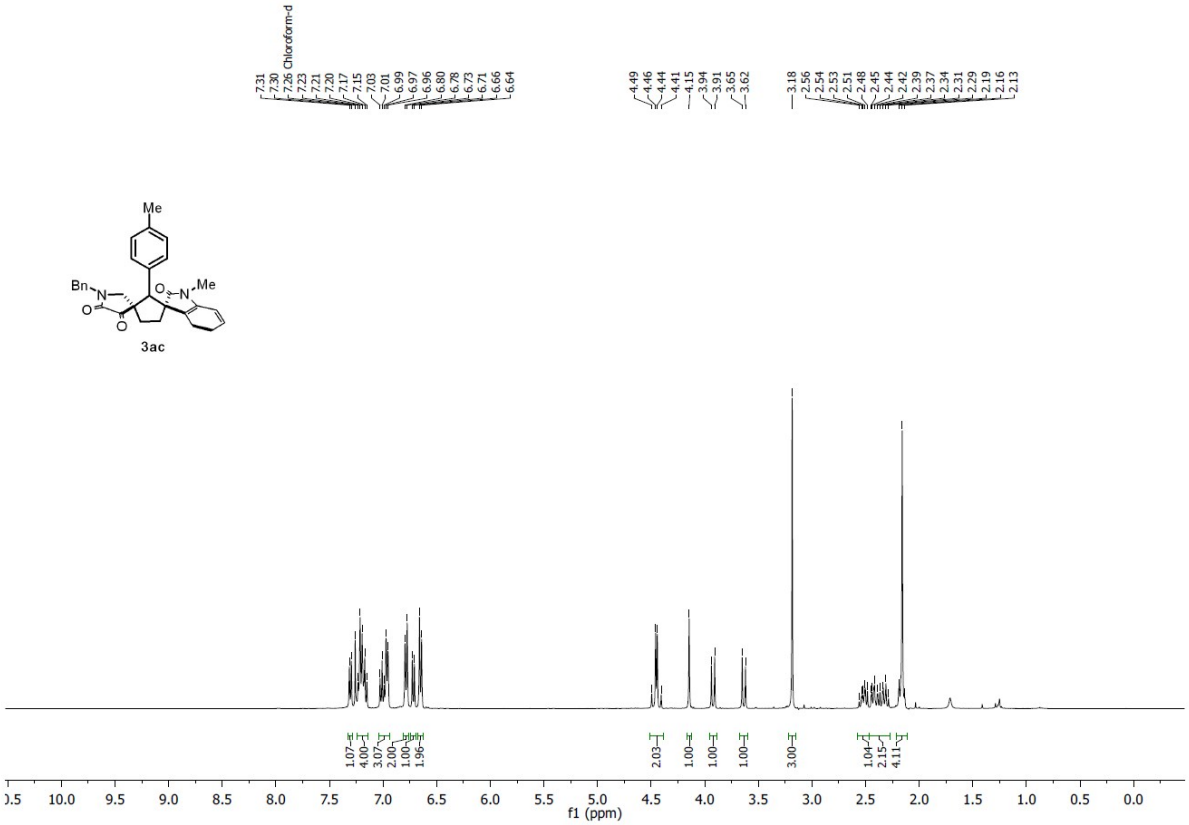
f1 (ppm)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Chloroform-d
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6.78
6.73
6.71
6.66
6.64
4.49
4.46
4.44
4.41
4.15
3.94
3.91
3.65
3.62
3.18
2.56
2.54
2.53
2.51
2.48
2.45
2.44
2.42
2.39
2.37
2.34
2.31
2.29
2.19
2.16
2.13
Me
Bn
N
O
N
Me
O
O
3ac
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4.00
3.07
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1.00
1.00
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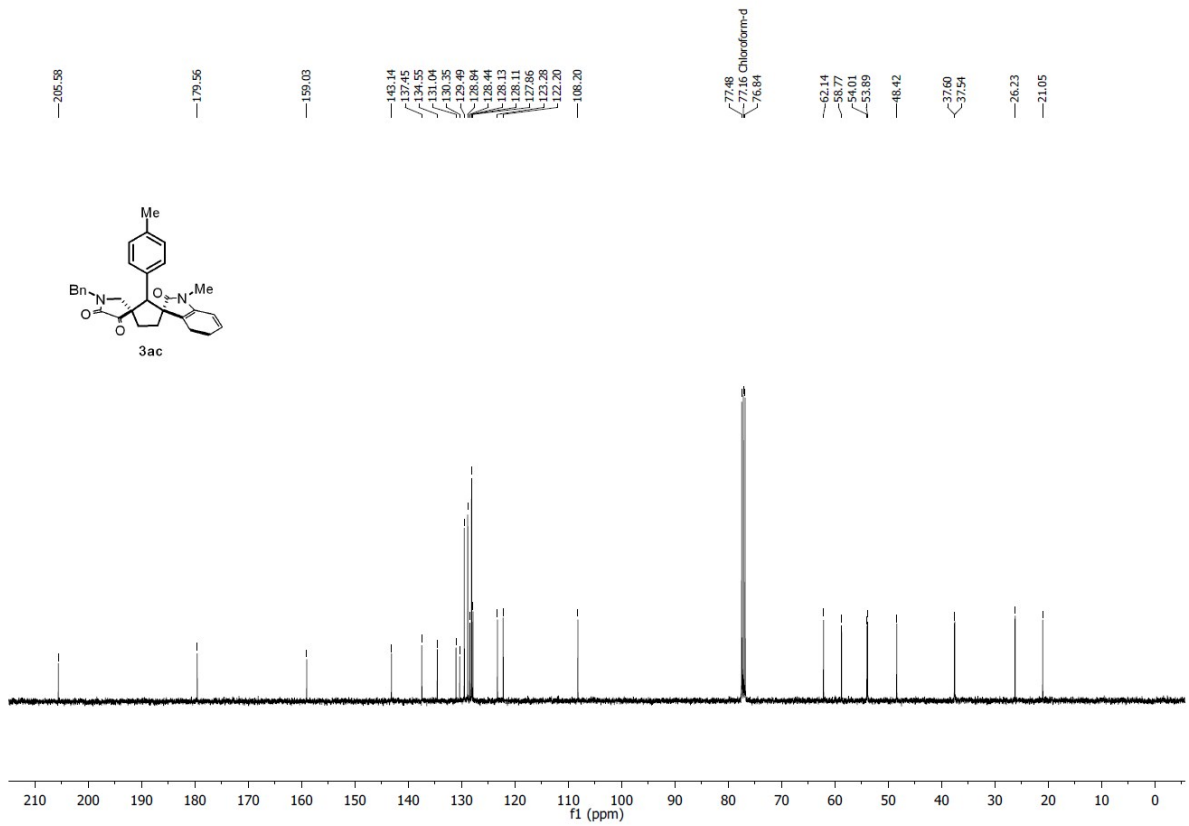
1.04
2.15
4.11
f1 (ppm)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128.44
128.13
128.11
127.86
123.38
122.20
108.20
77.48
77.16
Chloroform-d
76.84
62.14
58.77
54.01
53.89
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26.23
21.05
Me
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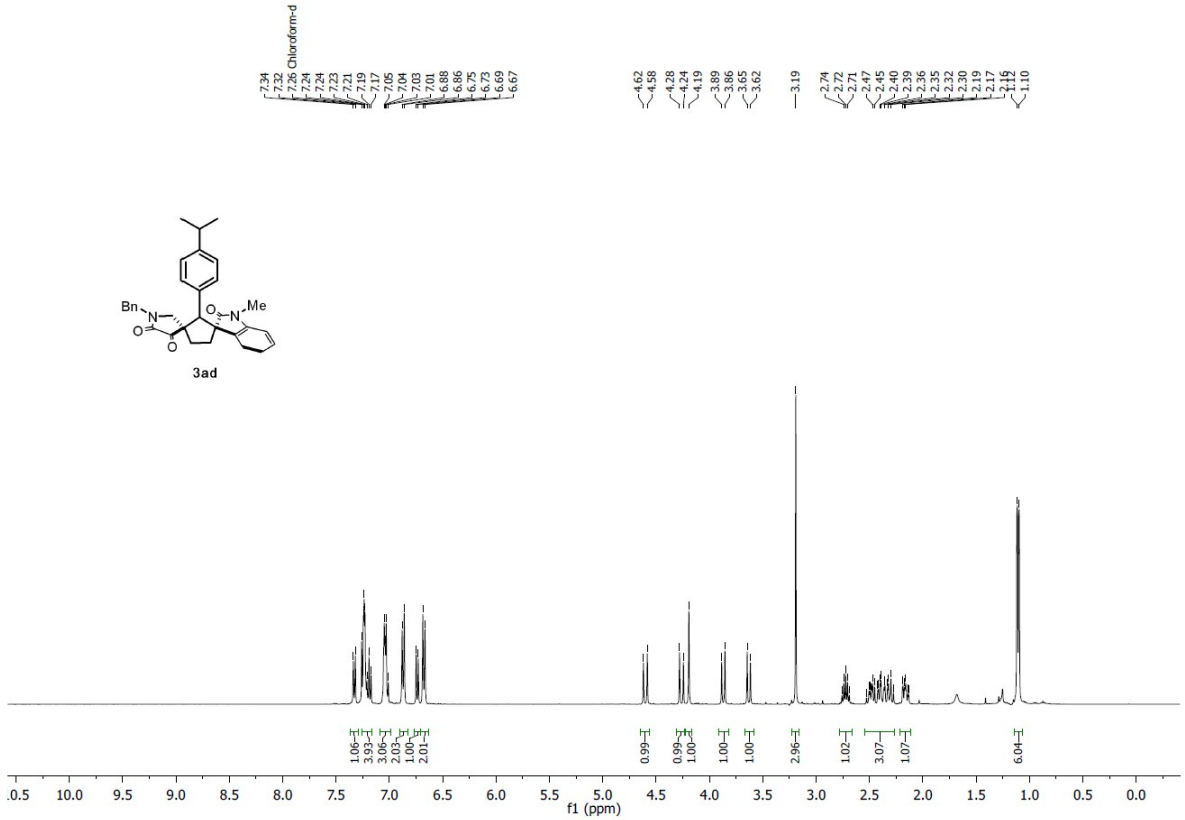
N
Me
O
O
3ac
f1 (ppm)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7.32
Chloroform-d
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7.03
7.01
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3.89
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3.62
3.19
2.74
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2.47
2.45
2.40
2.39
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2.35
2.32
2.20
2.19
2.17
2.16
1.12
1.10
Bn
N
O
O
O
N
Me
3ad
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3.93
3.06
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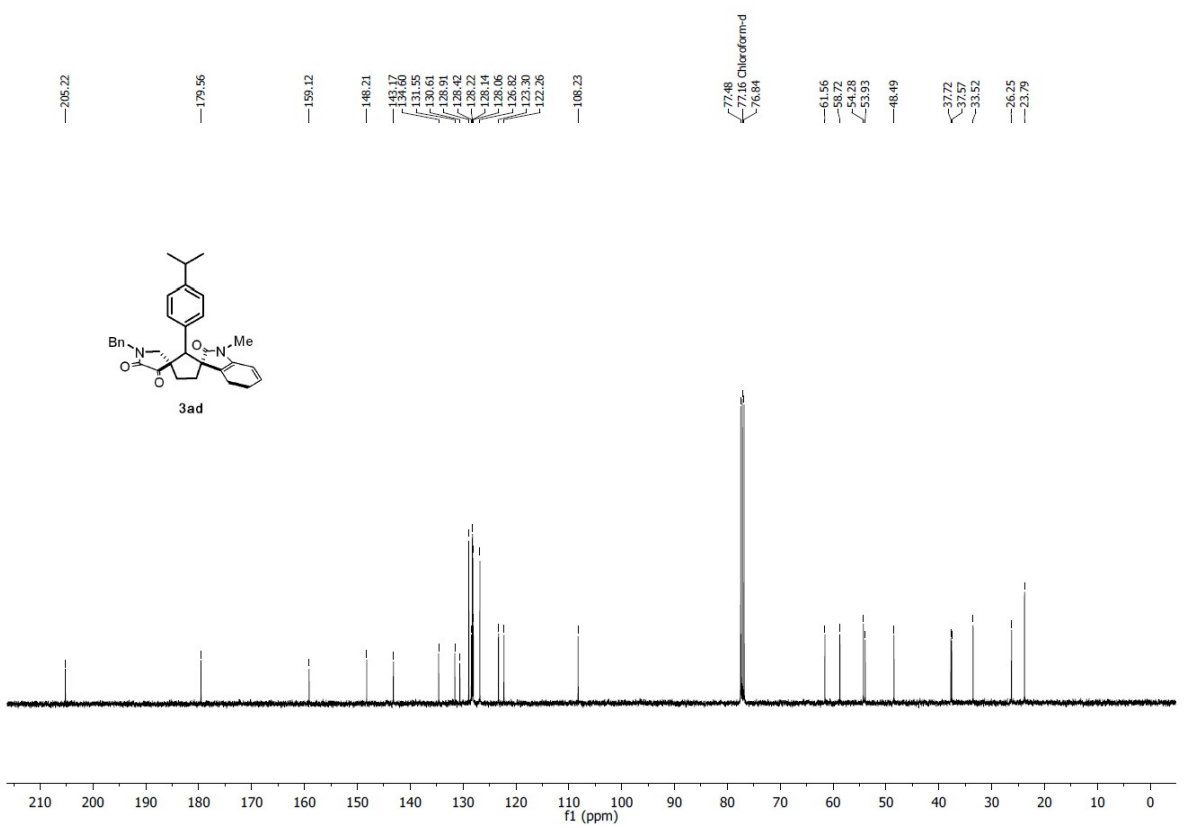
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122.26
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77.16
Chloroform-d
76.84
61.56
58.72
54.28
53.93
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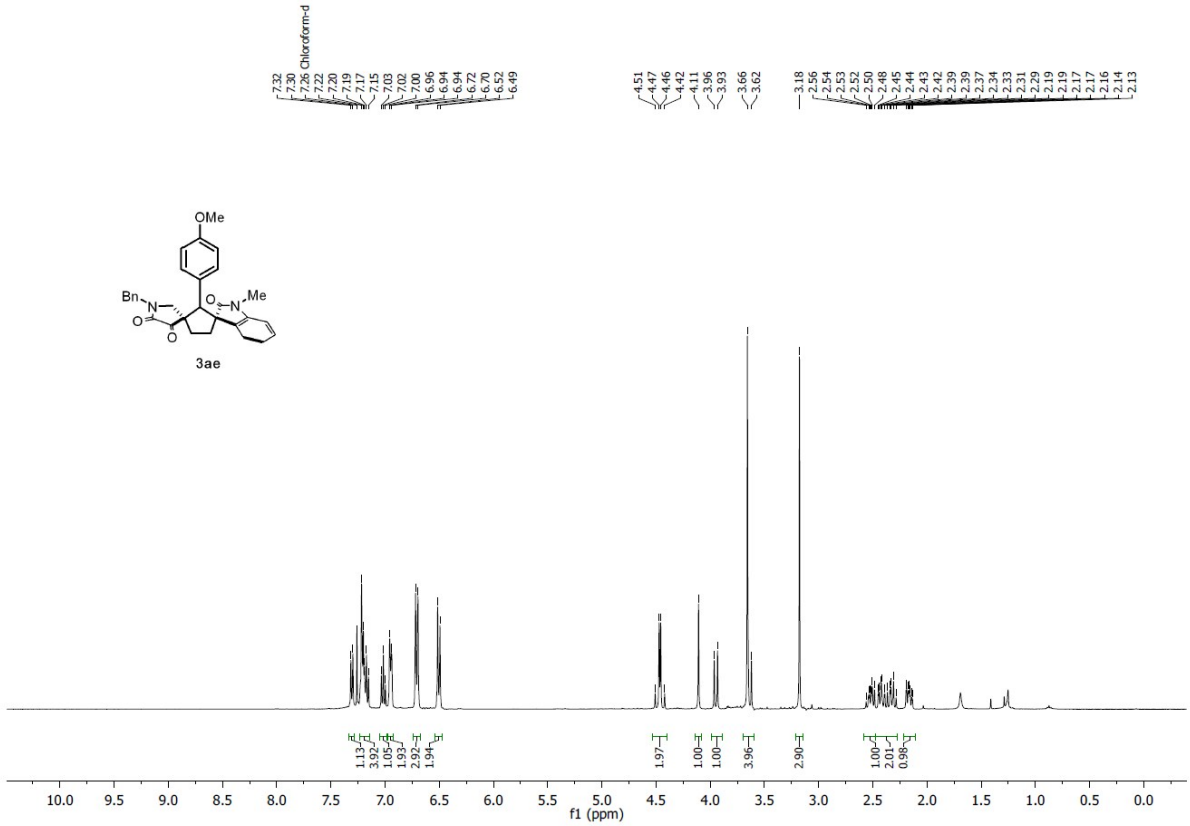
O
O
N
Me
3ad
f1 (ppm)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6.94
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4.47
4.46
4.42
4.11
3.96
3.93
3.66
3.62
3.18
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2.53
2.52
2.50
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2.44
2.43
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2.39
2.37
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2.33
2.31
2.29
2.19
2.19
2.17
2.17
2.16
2.14
2.13
OMe
Bn
N
O
O
O
N
Me
3ae
1.13
3.92
1.05
1.93
2.92
1.94
1.97
1.00
1.00
3.96
2.90
1.00
2.01
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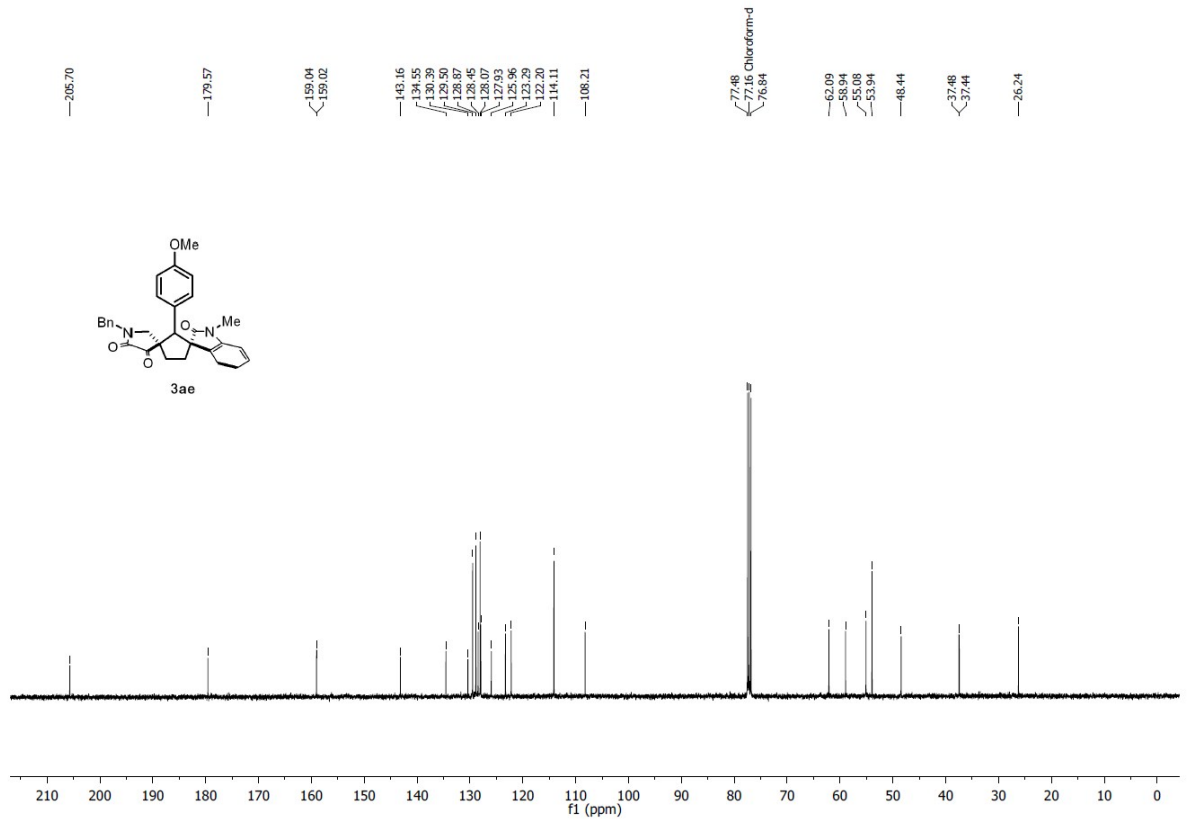
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77.48
77.16
Chloroform-d
76.84
62.09
58.94
55.08
53.94
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37.48
37.44
26.24
OMe
Bn
N
O
O
O
N
Me
3ae
210
200
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180
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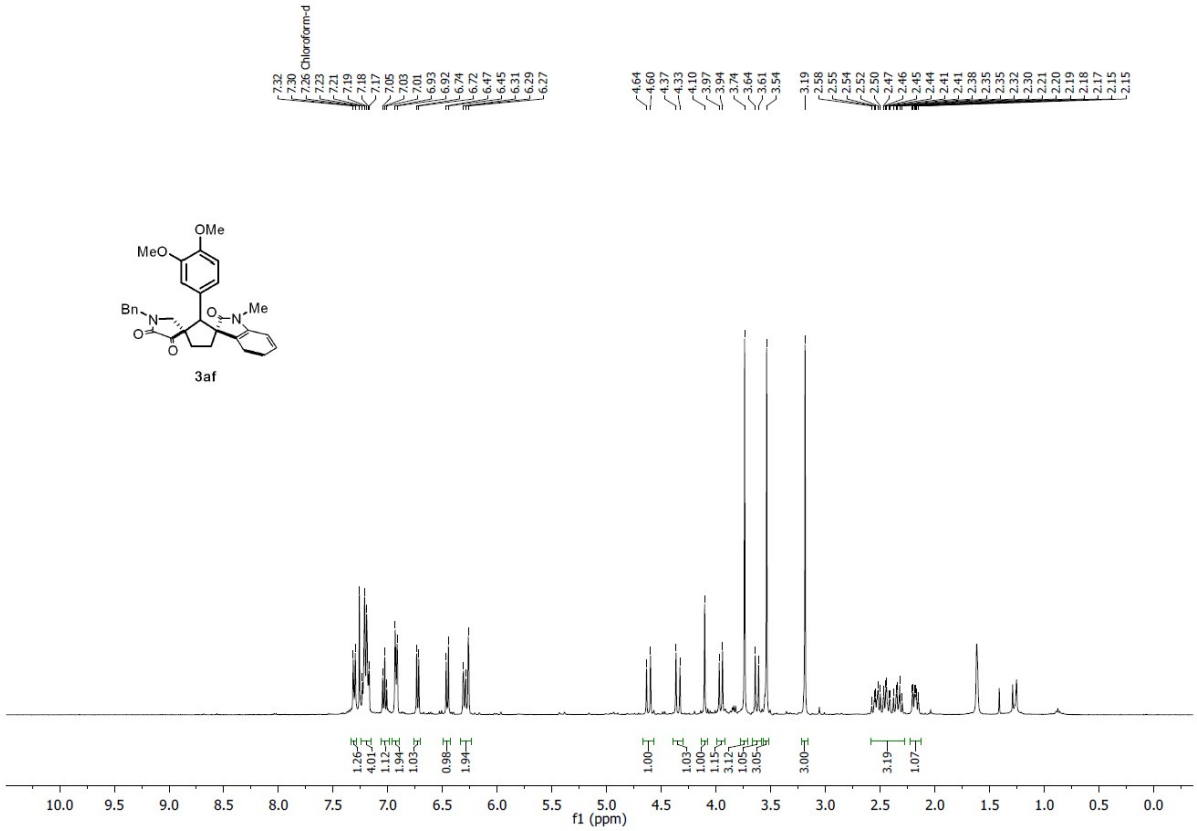
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6.92
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6.47
6.45
6.31
6.29
6.27
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4.60
4.37
4.33
4.10
3.97
3.94
3.74
3.64
3.61
3.54
3.19
2.58
2.55
2.54
2.52
2.50
2.47
2.46
2.45
2.44
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2.41
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2.19
2.18
2.17
2.15
2.15
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MeO
Bn
N
O
N
Me
O
O
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1.00
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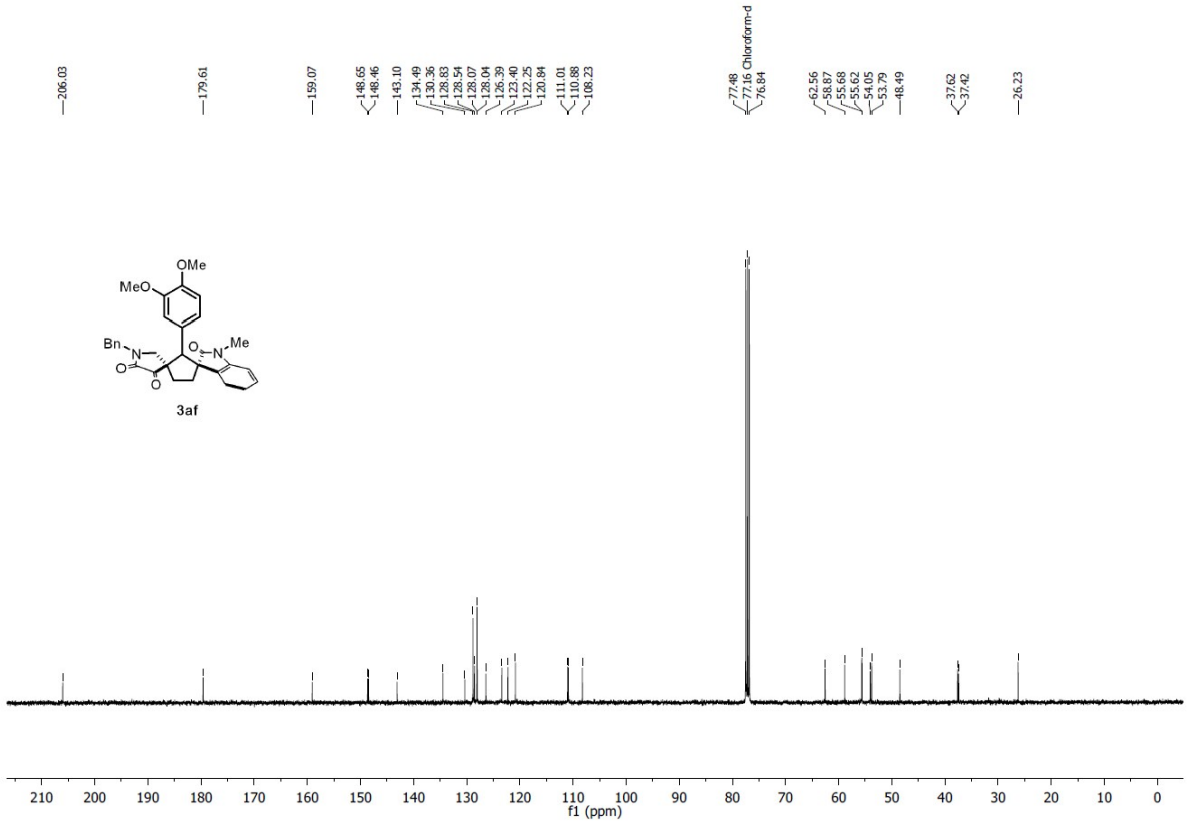
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77.48
77.16
Chloroform-d
76.84
62.56
58.87
55.68
55.62
54.05
53.79
48.49
37.62
37.42
26.23
OMe
MeO
Bn
N
O
N
Me
O
O
3af
210
200
190
180
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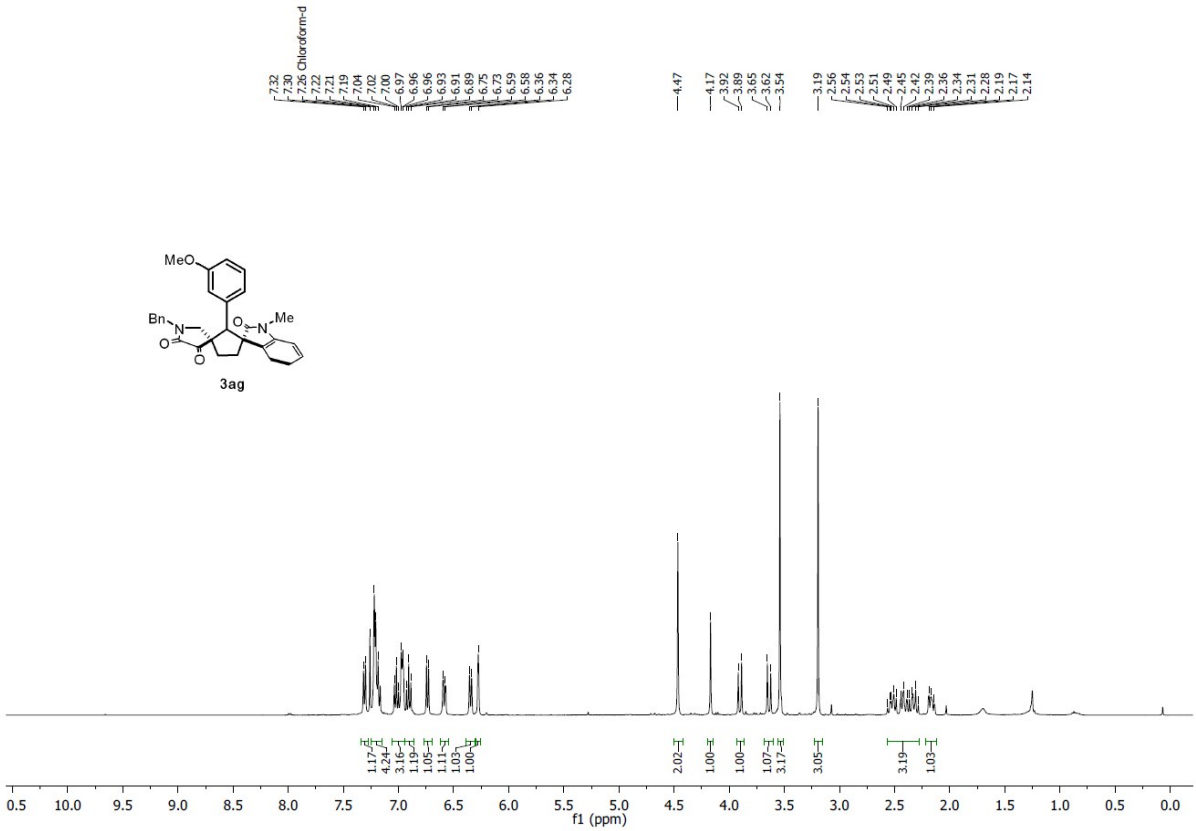
f1 (ppm)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6.36
6.34
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4.17
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3.62
3.54
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2.49
2.45
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2.34
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2.19
2.17
2.14
MeO
Bn
N
O
O
N
Me
O
3ag
1.17
4.24
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1.05
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1.03
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2.02
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1.00
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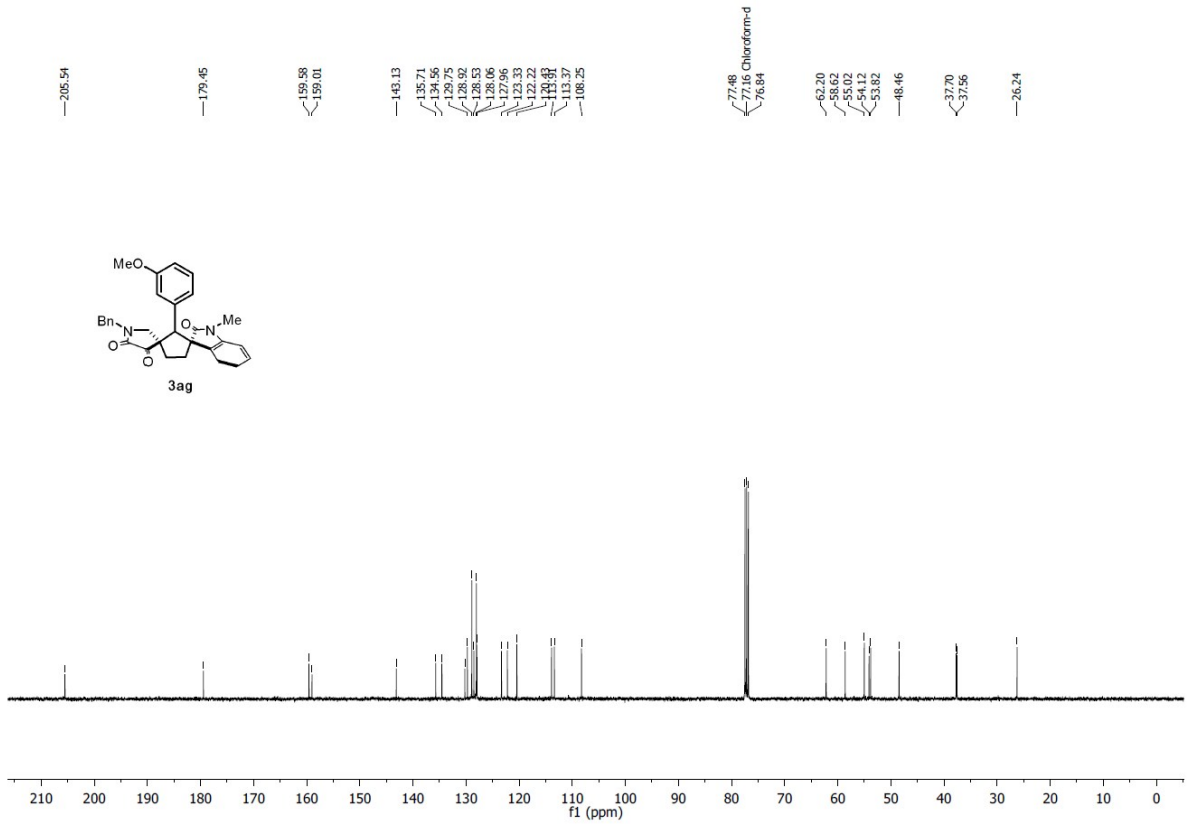
0.0
f1 (ppm)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122.22
113.37
108.25
77.48
77.16
Chloroform-d
76.84
62.20
58.62
55.02
54.12
53.82
48.46
37.70
37.56
26.24
MeO
Bn
N
O
O
N
Me
O
3ag
210
200
190
180
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120
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100
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80
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50
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10
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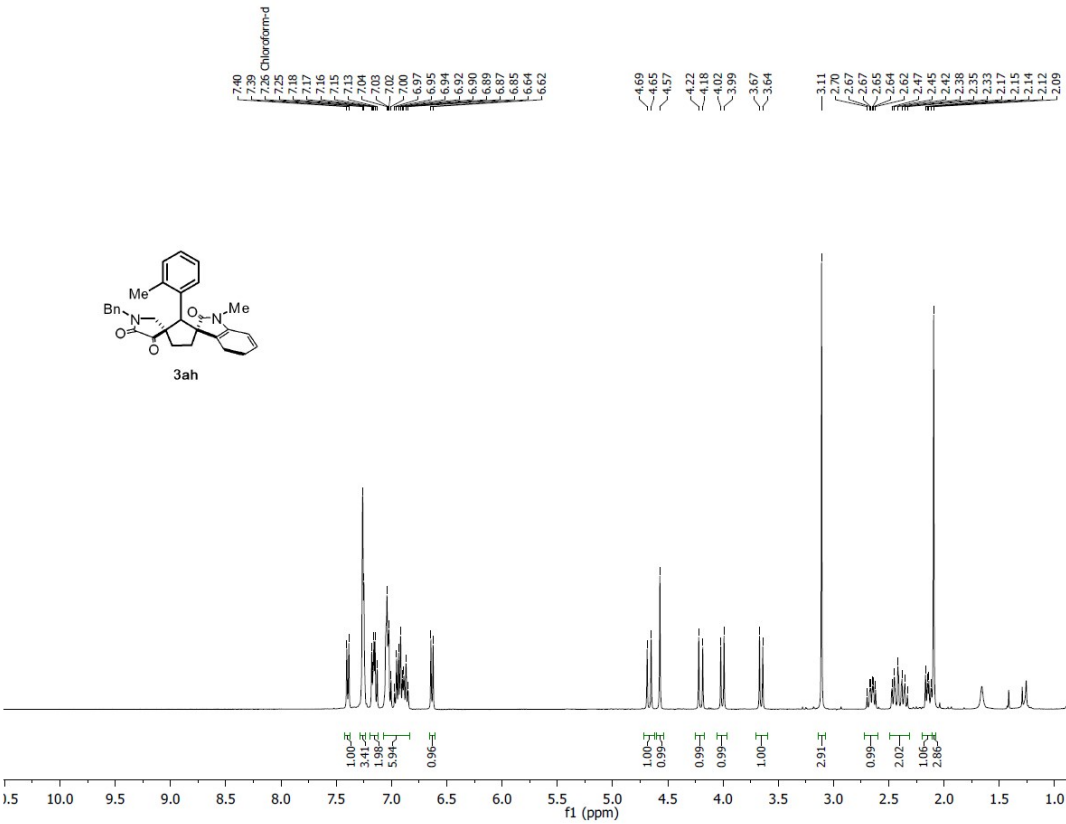
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7.02
7.00
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6.90
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6.87
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2.17
2.15
2.14
2.12
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O
N
Me
O
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3.41
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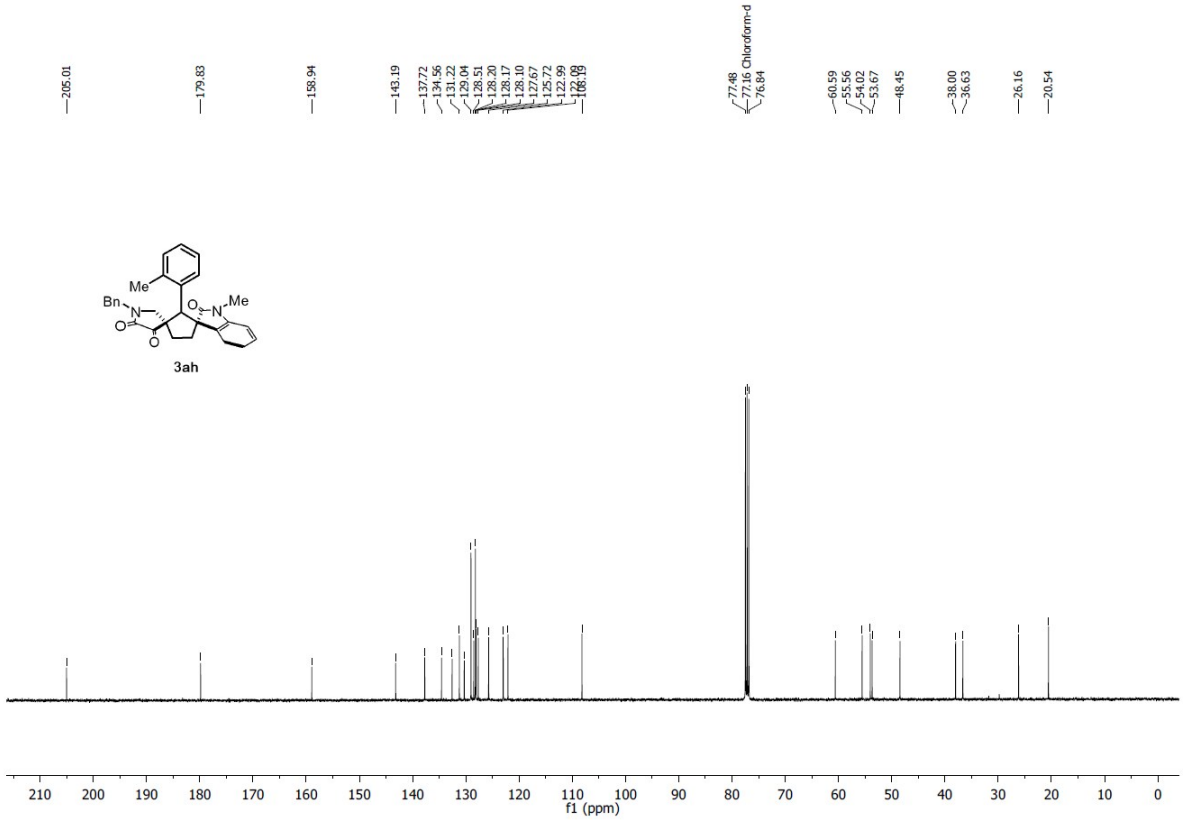
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122.99
122.09
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77.48
77.16
Chloroform-d
76.84
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55.56
54.02
53.67
48.45
38.00
36.63
26.16
20.54
Me
Bn
N
O
O
N
Me
O
3ah
f1 (ppm)


7.66
7.64
7.62
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7.60
7.58
7.56
7.54
7.52
7.39
7.38
7.36
7.31
7.29
7.28
7.26 Chloroform-d
7.08
7.06
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7.03
5.00
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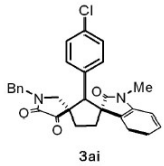
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Me
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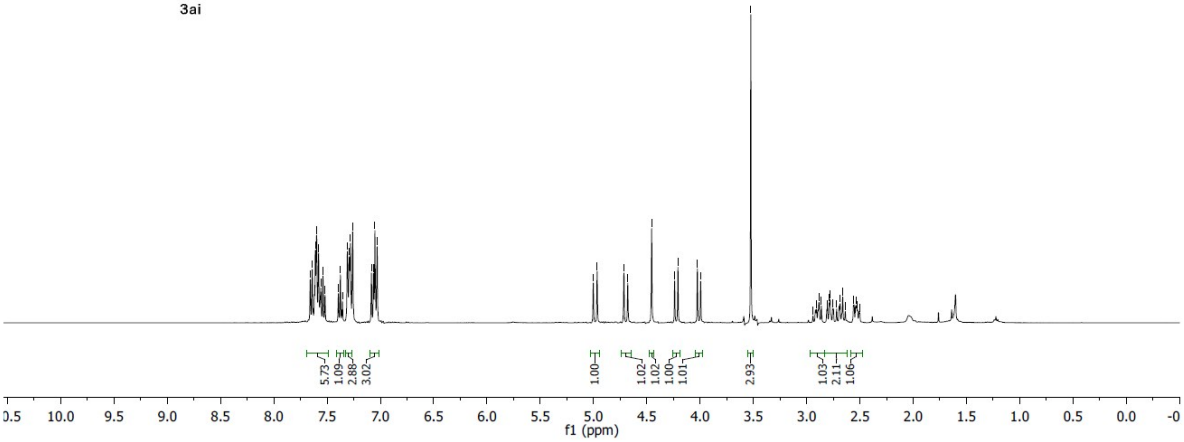
O

3ai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1.06
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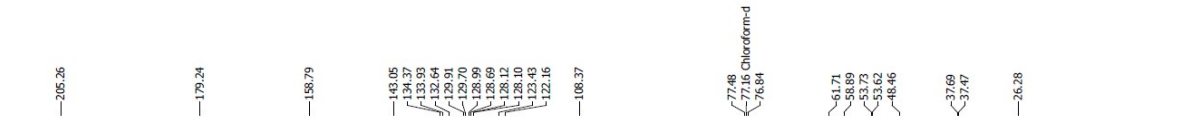
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128.10
123.43
122.16
108.37
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77.16 Chloroform-d
76.84
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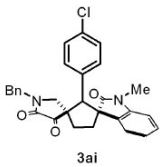
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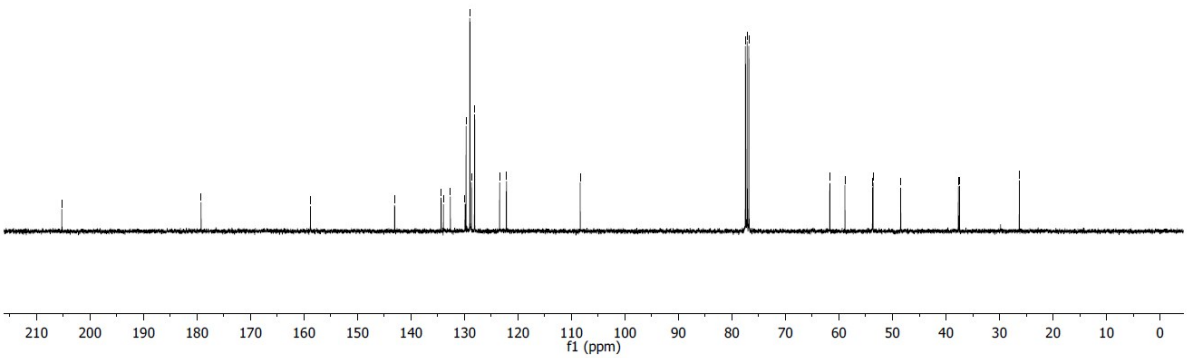
Me
O
O

3ai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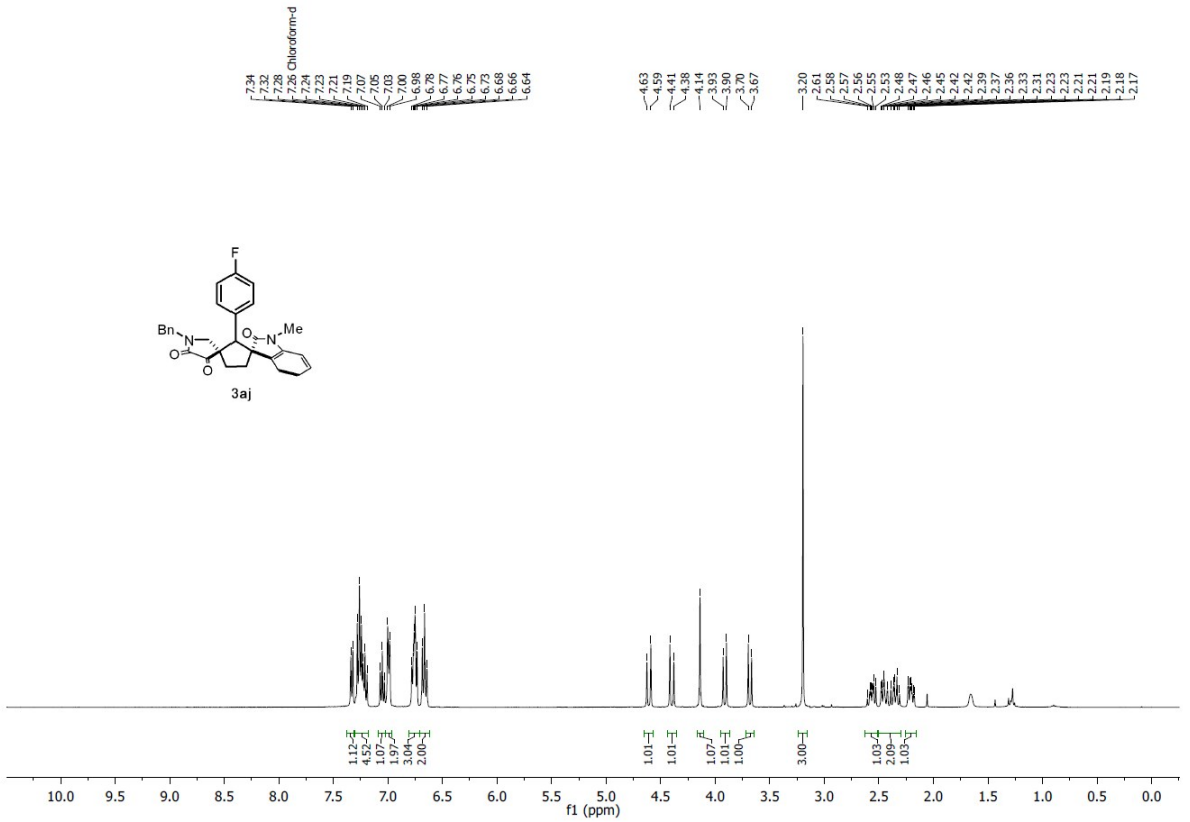
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Chloroform-d
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6.66
6.64
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2.23
2.23
2.21
2.21
2.19
2.18
2.17
F
Bn
N
O
O
O
N
Me
3aj
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1.07
1.01
1.00
3.00
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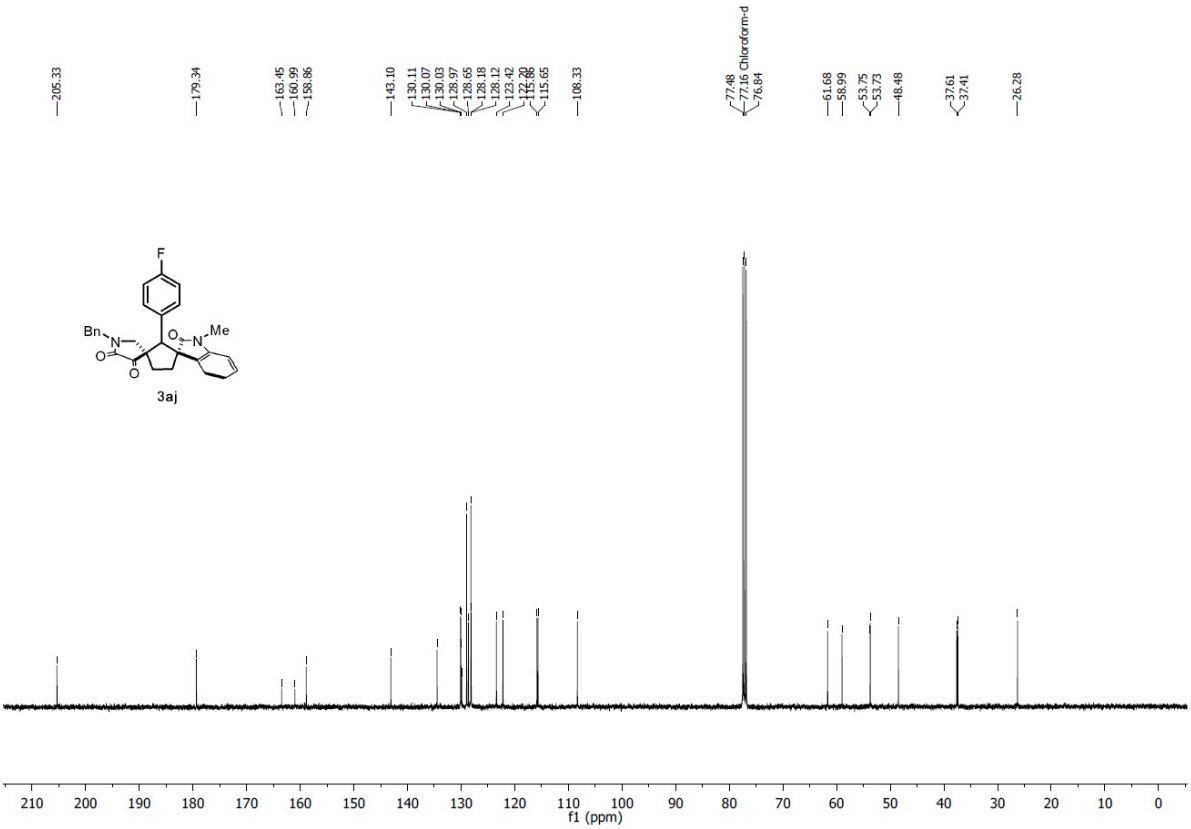
f1 (ppm)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128.12
123.42
115.88
115.65
108.33
77.48
77.16
Chloroform-d
76.84
61.68
58.99
53.75
53.73
48.48
37.61
37.41
26.28
F
Bn
N
O
O
O
N
Me
3aj
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200
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180
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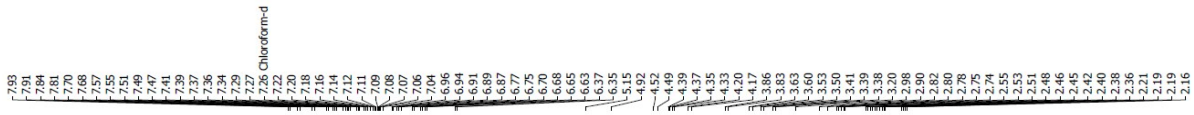
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7.37
7.36
7.34
7.29
7.27
7.26 Chloroform-d
7.22
7.20
7.18
7.16
7.14
7.12
7.11
7.09
7.08
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7.06
7.04
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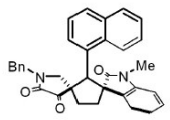
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Me

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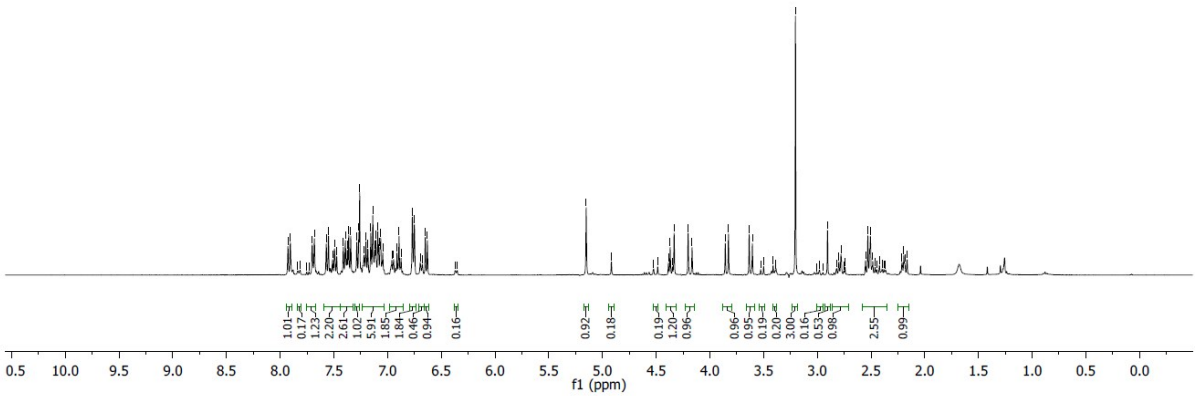
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0.92
0.18
0.19
1.20
0.96
0.96
0.95
0.19
0.20
3.00
0.16
0.53
0.98
2.55
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6.0
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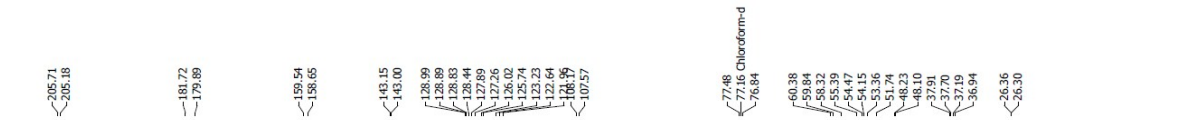
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126.02
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122.64
107.57
77.48
77.16 Chloroform-d
76.84
60.38
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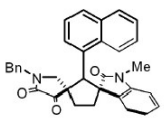
Bn
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O
O
O
N
Me

3ak

210
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100
90
80
70
60
50
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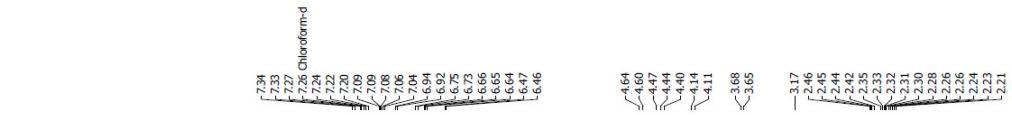
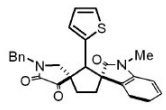
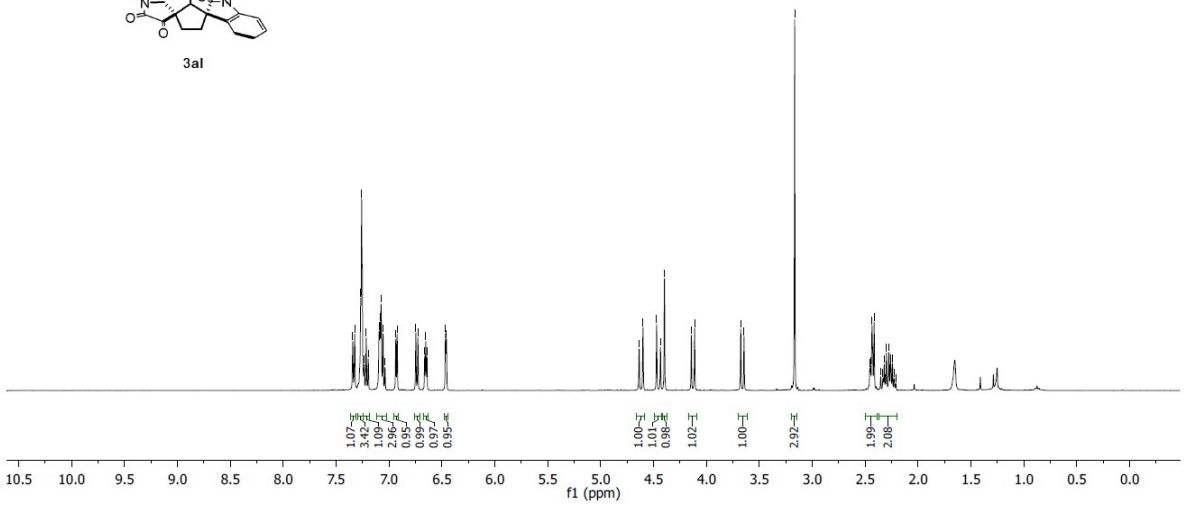
30
20
10
0
f1 (ppm)


7.34
7.33
7.27
Chloroform-d
7.26
7.24
7.22
7.20
7.09
7.08
7.06
7.04
6.94
6.92
6.75
6.73
6.66
6.65
6.64
6.47
6.46
4.64
4.60
4.47
4.44
4.40
4.14
4.11
3.68
3.65
3.17
2.46
2.45
2.44
2.42
2.35
2.33
2.32
2.31
2.30
2.28
2.26
2.26
2.24
2.23
2.21


Bn
N
O
O
S
O
N
Me

3al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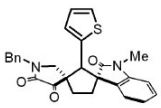
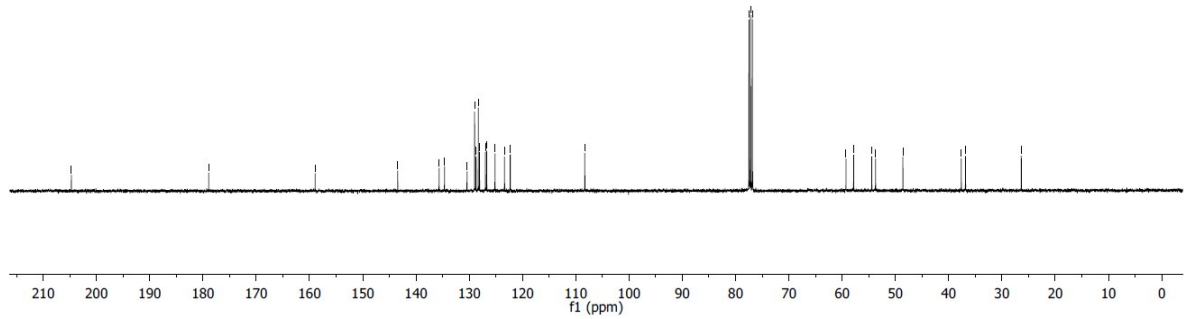
3.42
1.09
2.96
0.95
0.99
0.97
0.95
1.00
1.01
0.98
1.02
1.00
2.92
1.99
2.08
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


204.70
178.92
158.94
143.44
135.66
134.65
130.43
129.00
128.74
128.30
128.07
126.97
126.73
125.17
123.38
122.31
108.31
77.48
77.16
Chloroform-d
76.84
59.27
57.83
54.43
53.72
48.59
37.63
36.83
26.36


Bn
N
O
O
S
O
N
Me

3al


210
200
190
180
170
160
150
140
130
120
110
100
90
80
70
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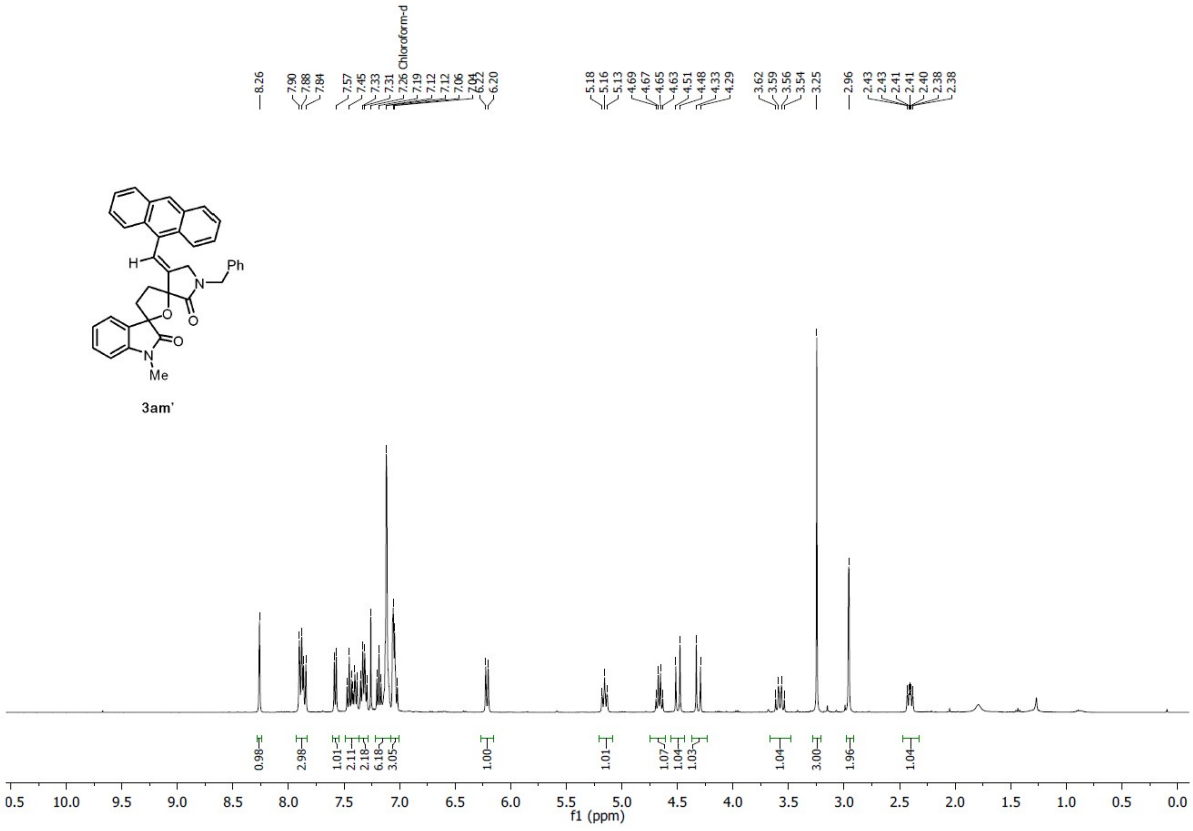
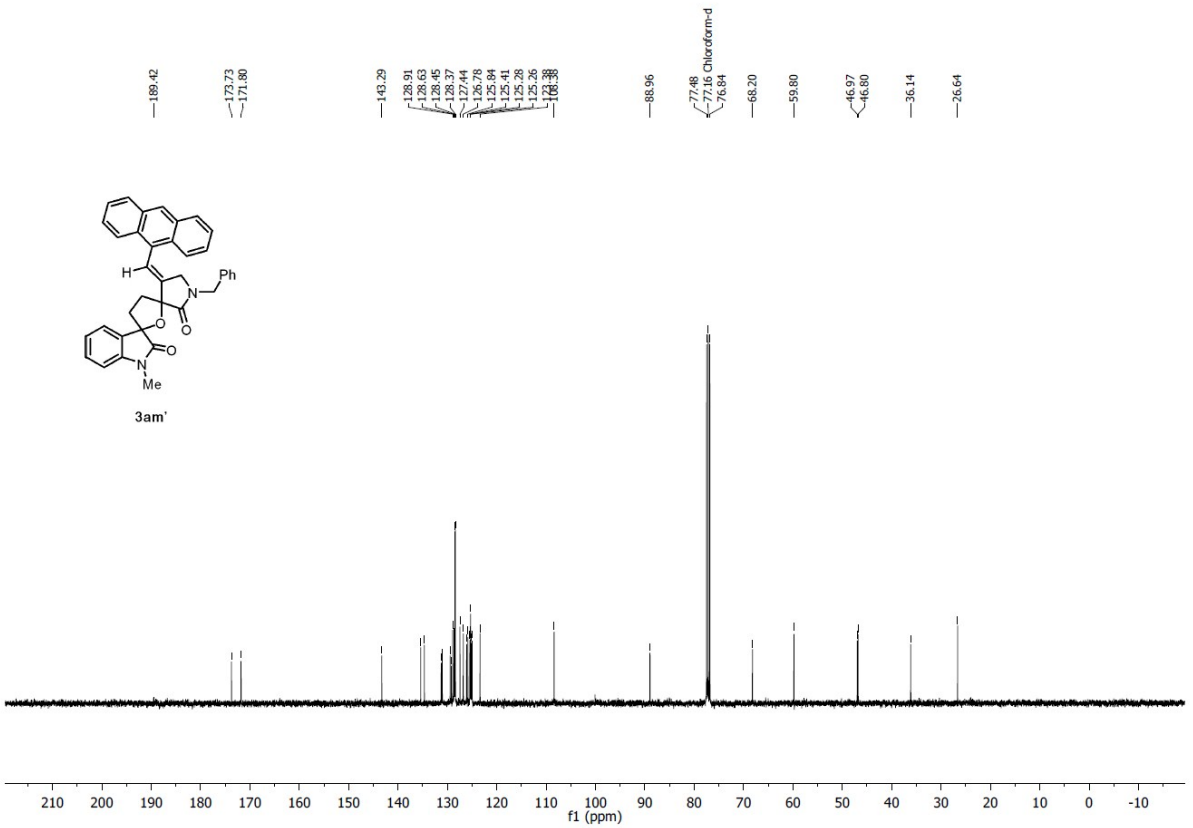
50
40
30
20
10
0
f1 (ppm)


8.26
7.90
7.88
7.84
7.57
7.45
7.33
7.31
Chloroform-d
7.26
7.19
7.12
7.12
7.06
6.22
6.20
5.18
5.16
5.13
4.69
4.67
4.65
4.63
4.51
4.48
4.33
4.29
3.62
3.59
3.56
3.54
3.25
2.96
2.43
2.43
2.41
2.41
2.40
2.38
2.38
H
Ph
N
O
O
O
N
Me
3am'
0.98
2.98
1.01
2.11
2.18
6.18
3.05
1.00
1.01
1.07
1.04
1.03
1.04
3.00
1.96
1.04
f1 (ppm)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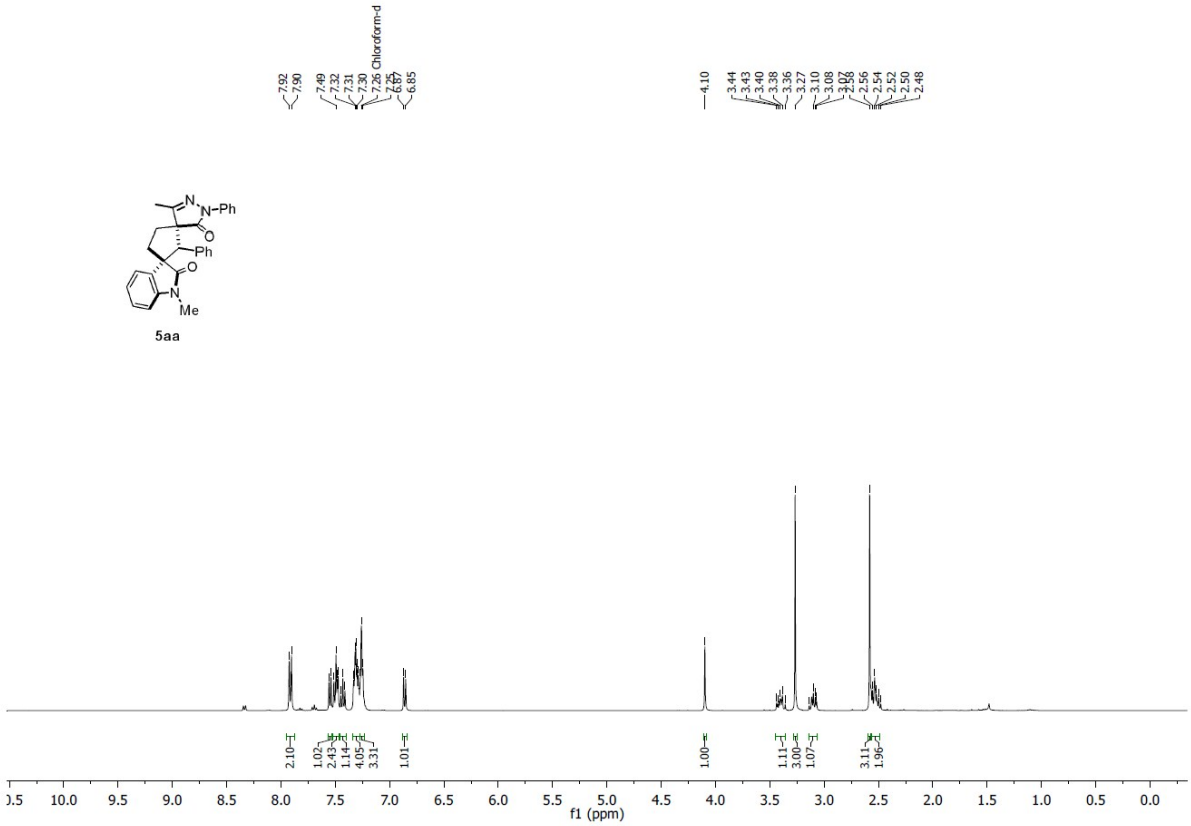
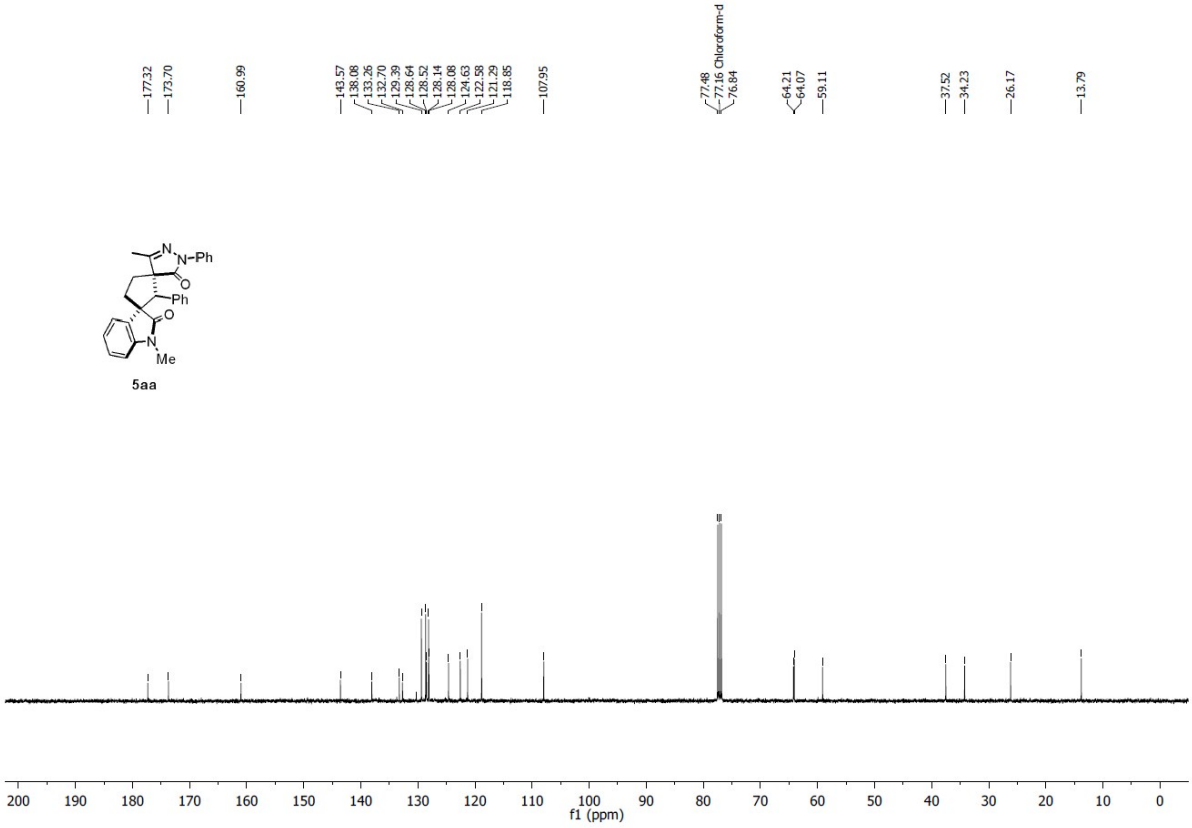
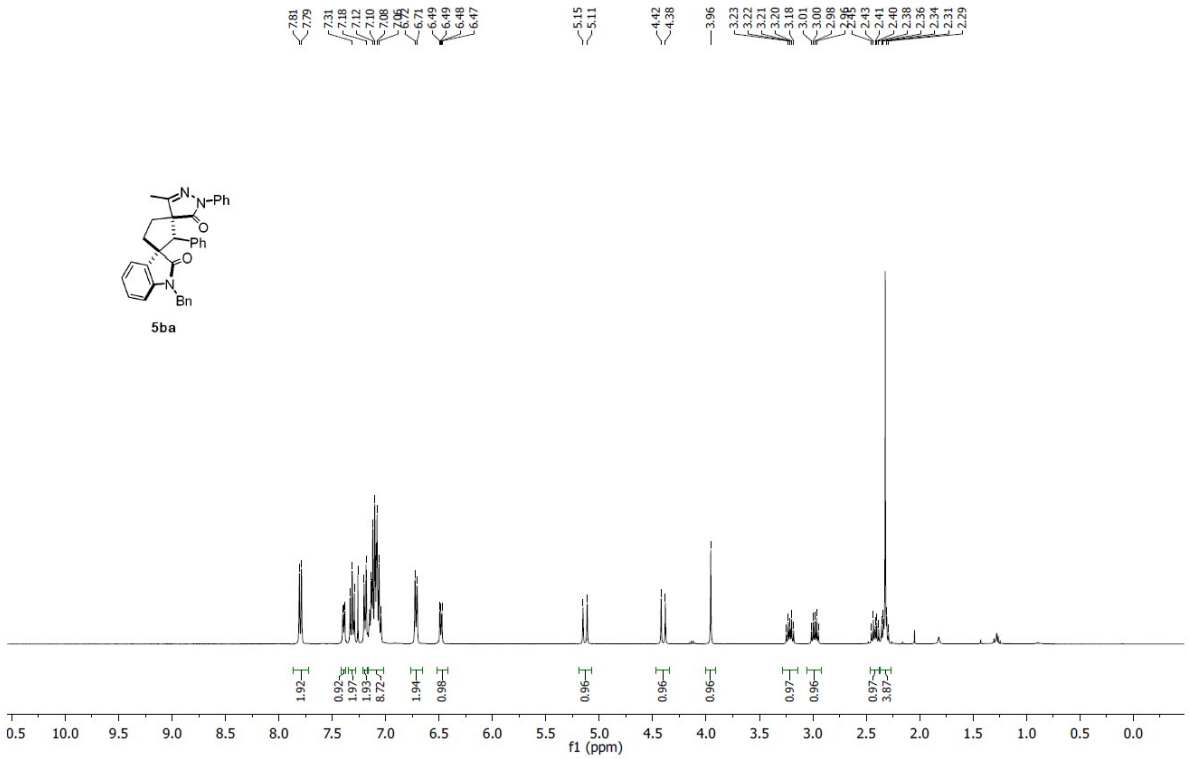
173.73
171.80
143.29
128.91
128.63
128.45
128.37
127.44
126.78
125.84
125.41
125.28
125.26
88.96
77.48
77.16
Chloroform-d
76.84
68.20
59.80
46.97
46.80
36.14
26.64
H
Ph
N
O
O
O
N
Me
3am'
f1 (ppm)

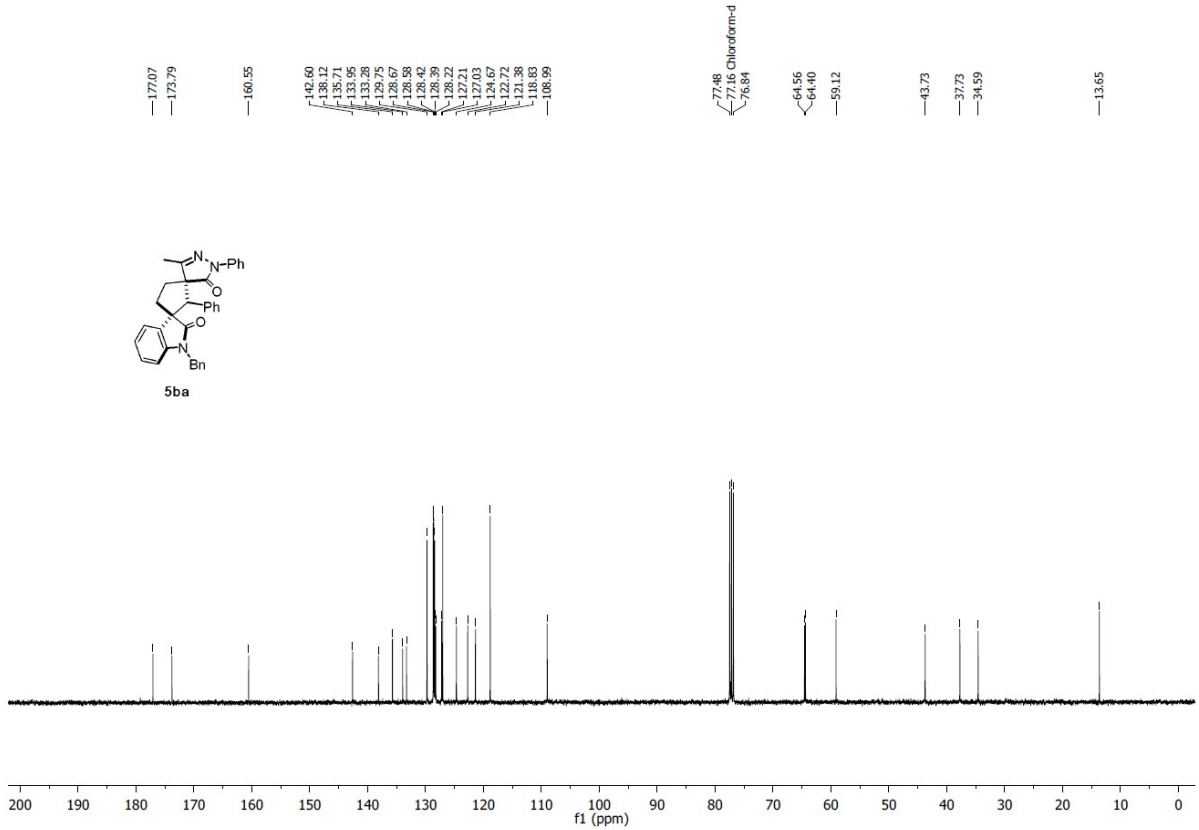

7.92
7.90
7.49
7.32
7.31
7.30
7.26 Chloroform-d
6.85
4.10
3.44
3.43
3.40
3.38
3.36
3.27
3.10
3.08
2.56
2.54
2.52
2.50
2.48
N-Ph
Ph
O
N
Me
2.10
1.02
2.43
1.14
4.05
3.31
1.01
1.00
1.11
3.00
1.07
3.11
1.96
f1 (ppm)

5aa


177.32
173.70
160.99
143.57
138.08
133.26
132.70
129.39
128.64
128.52
128.14
128.08
124.63
122.58
121.29
118.85
107.95
77.48
77.16 Chloroform-d
76.84
64.21
64.07
59.11
37.52
34.23
26.17
13.79
N-Ph
Ph
O
N
Me
f1 (ppm)

5aa


7.81
7.79
7.31
7.18
7.12
7.10
7.08
6.71
6.49
6.48
6.47
5.15
5.11
4.42
4.38
3.96
3.23
3.22
3.21
3.20
3.18
3.01
3.00
2.98
2.43
2.41
2.40
2.38
2.36
2.34
2.31
2.29
N-Ph
O
Ph
O
N
Bn
5ba
1.92
0.92
1.97
1.93
8.72
1.94
0.98
0.96
0.96
0.96
0.97
0.96
0.97
3.87
f1 (ppm)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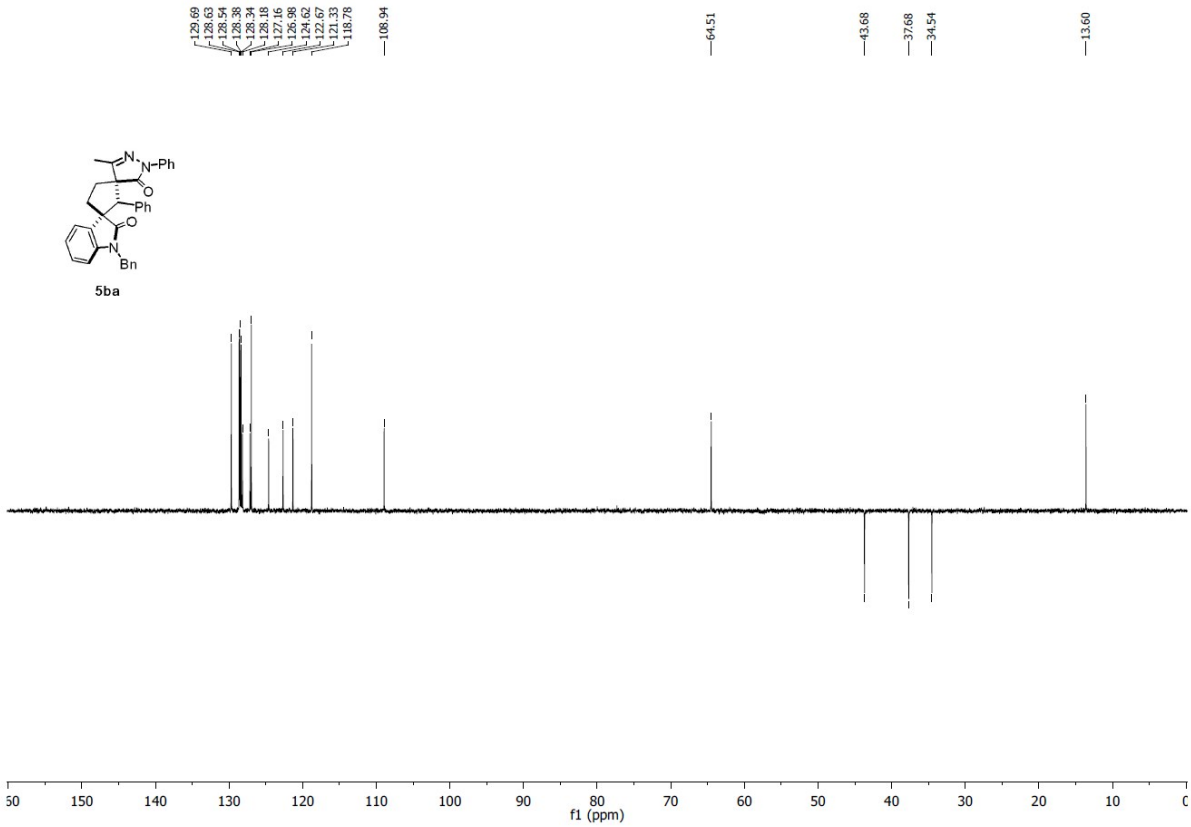
173.79
160.55
142.60
138.12
135.71
133.95
133.28
129.75
128.67
128.58
128.42
128.39
128.22
127.21
127.03
124.67
122.72
121.38
118.83
108.99
77.48
77.16 Chloroform-d
76.84
64.56
64.40
59.12
43.73
37.73
34.59
13.65
N-Ph
O
Ph
O
N
Bn
5ba
f1 (ppm)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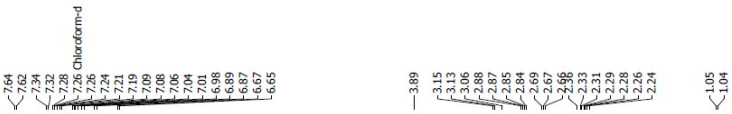
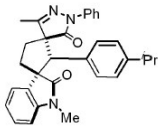
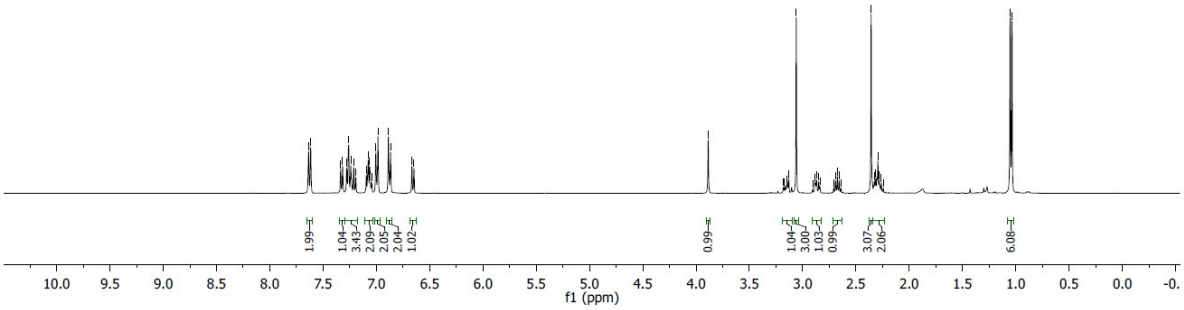
128.63
128.54
128.38
128.34
128.18
127.16
126.98
124.62
122.67
121.33
118.78
108.94
64.51
43.68
37.68
34.54
13.60
N
N–Ph
O
Ph
O
N
Bn
5ba
50
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

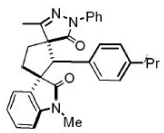

7.64
7.62
7.34
7.32
7.28
Chloroform-d
7.26
7.24
7.21
7.19
7.09
7.08
7.06
7.04
7.01
6.98
6.89
6.87
6.67
6.65
3.89
3.15
3.13
3.06
2.88
2.87
2.85
2.84
2.69
2.67
2.66
2.33
2.31
2.29
2.28
2.26
2.24
1.05
1.04


N
N-Ph
O
O
iPr
N
Me

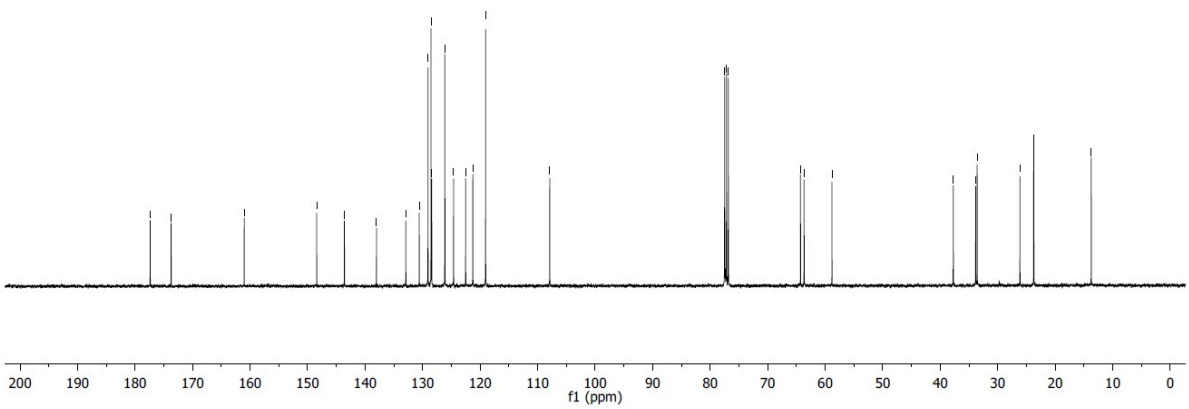
5ab

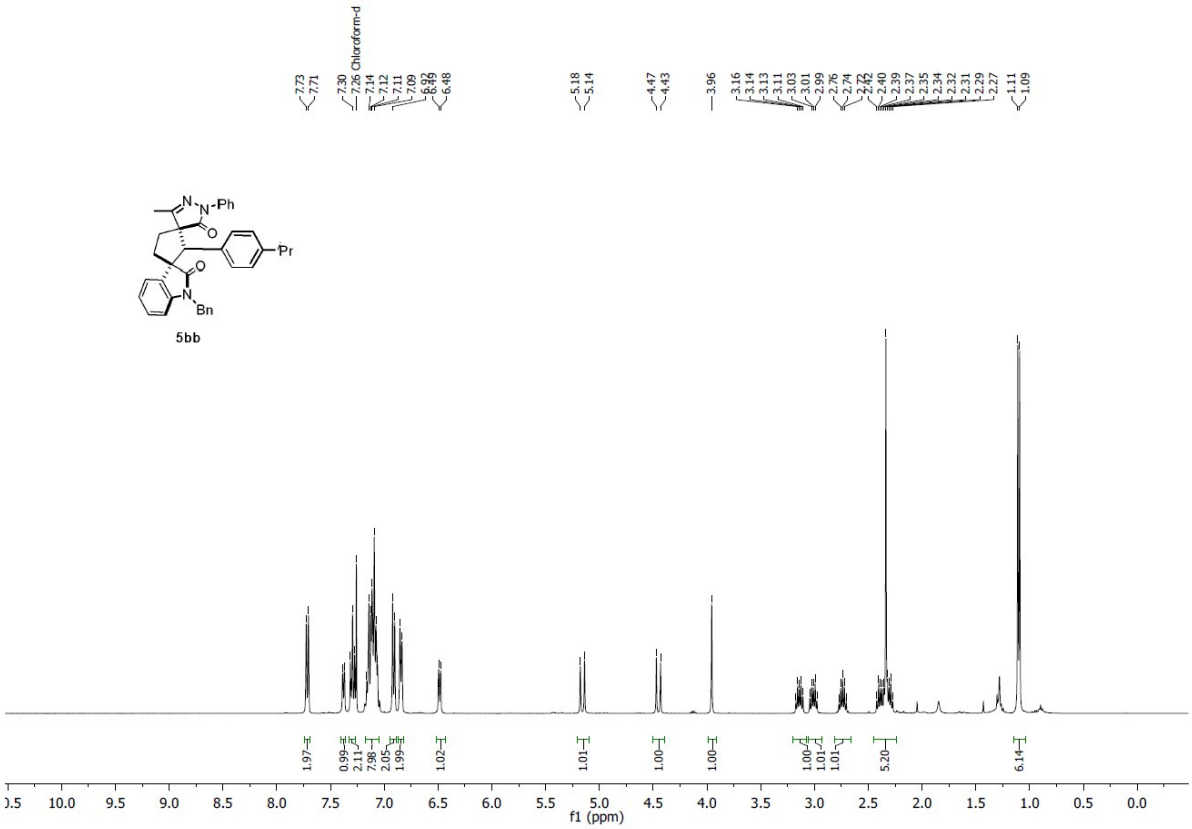

1.99
1.04
3.43
2.09
2.05
2.04
1.02
0.99
1.04
3.00
1.03
0.99
3.07
2.06
6.08
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
-0.
f1 (ppm)

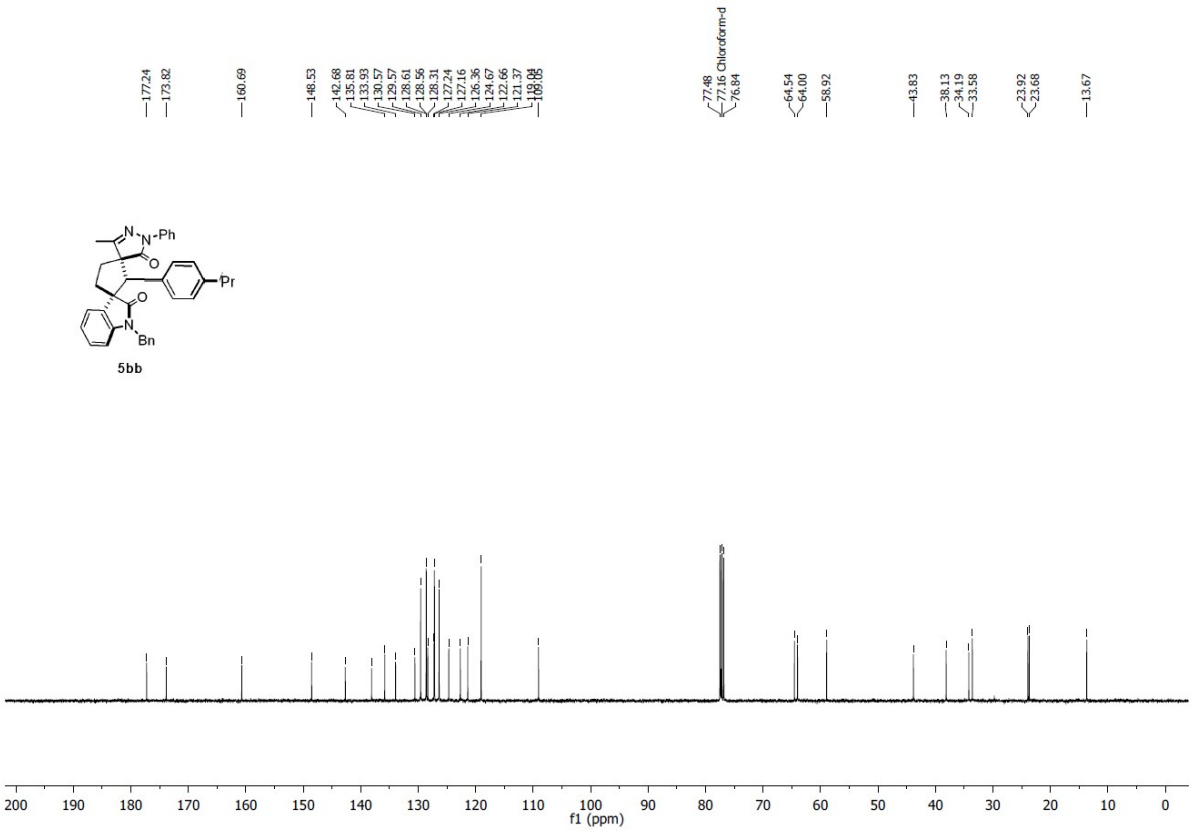

177.36
173.73
161.02
148.41
143.60
138.02
132.92
130.58
129.06
128.54
128.41
126.13
124.61
122.52
121.27
119.05
107.90
77.48
77.16
Chloroform-d
76.84
64.29
63.67
58.82
37.72
33.83
33.57
26.14
23.75
23.72
13.74

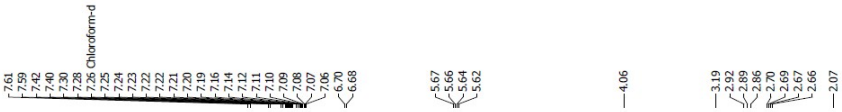

N
N-Ph
O
O
iPr
N
Me

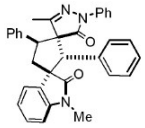
5ab


200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

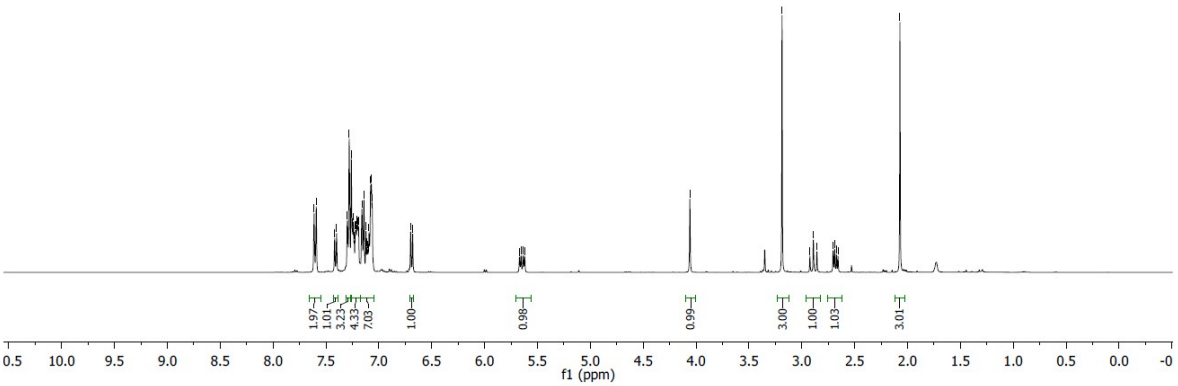

7.73
7.71
7.30
7.26 Chloroform-d
7.14
7.12
7.11
7.09
6.52
6.49
6.48
5.18
5.14
4.47
4.43
3.96
3.16
3.14
3.13
3.11
3.03
3.01
2.99
2.76
2.74
2.72
2.42
2.40
2.39
2.37
2.35
2.34
2.32
2.31
2.29
2.27
1.11
1.09
Ph
Pr
Bn
5bb
1.97
0.99
2.11
7.98
2.05
1.99
1.02
1.01
1.00
1.00
1.00
1.01
1.01
5.20
6.14
f1 (ppm)

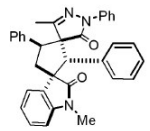

177.24
173.82
160.69
148.53
142.68
135.81
133.93
130.57
129.57
128.61
128.56
128.31
127.24
127.16
126.36
124.67
122.66
121.37
77.48
77.16 Chloroform-d
76.84
64.54
64.00
58.92
43.83
38.13
34.19
33.58
23.92
23.68
13.67
Ph
Pr
Bn
5bb
f1 (ppm)



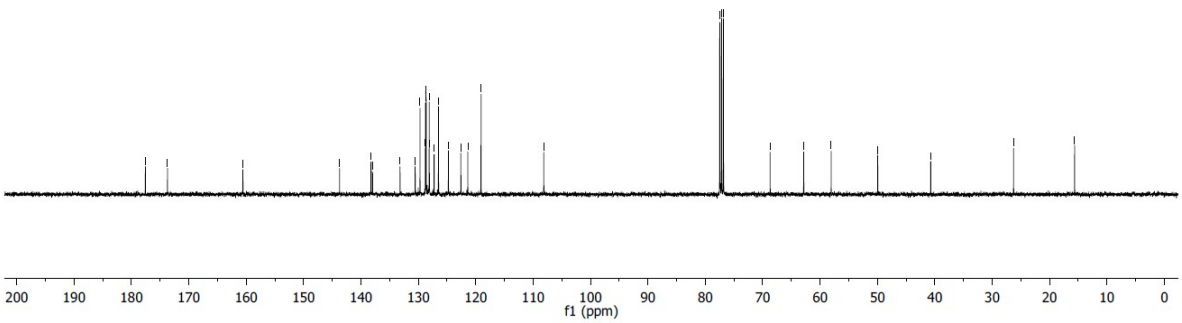


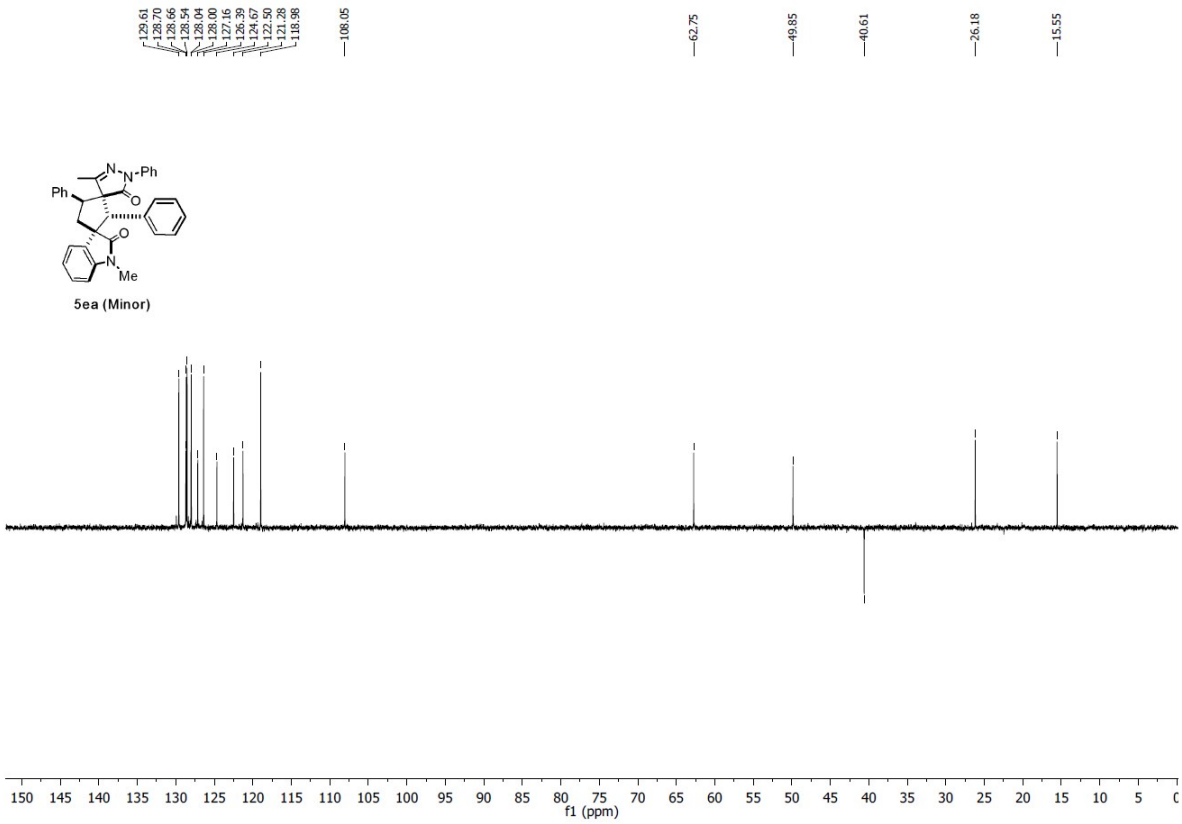
5ea (Minor)





5ea (Minor)

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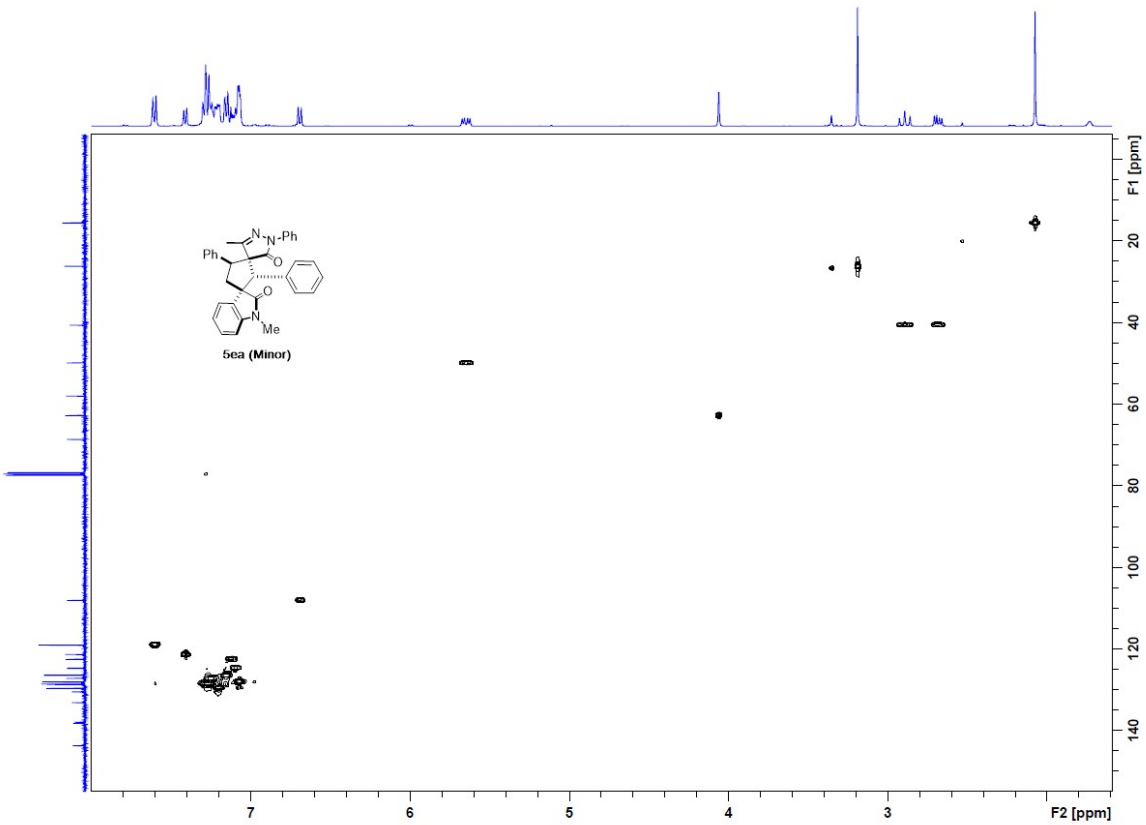
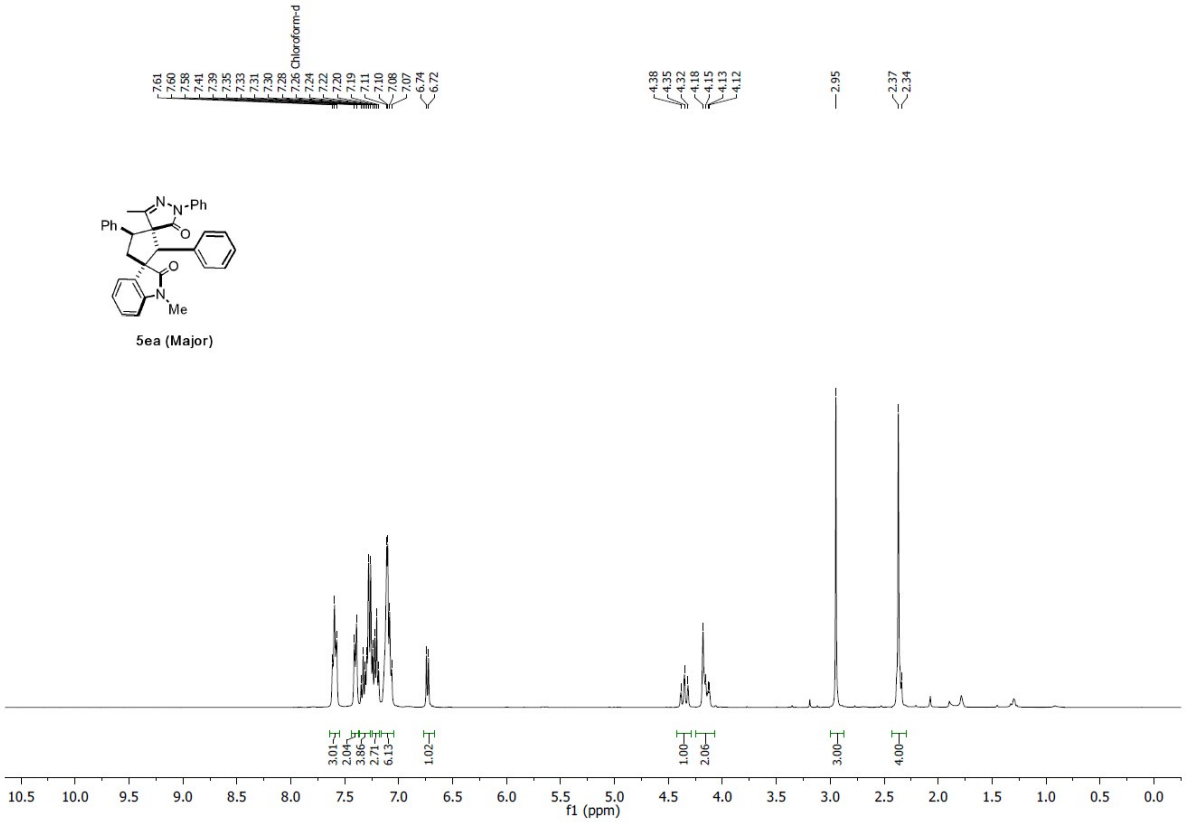
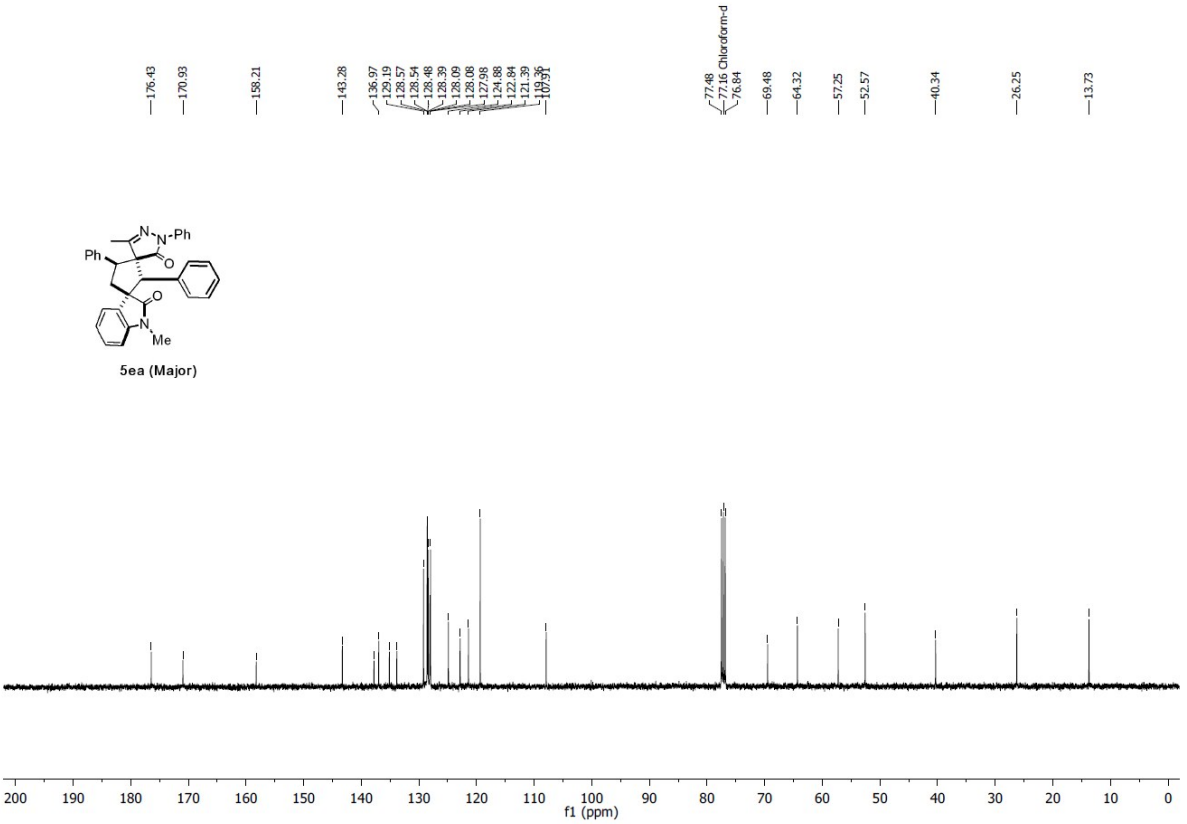
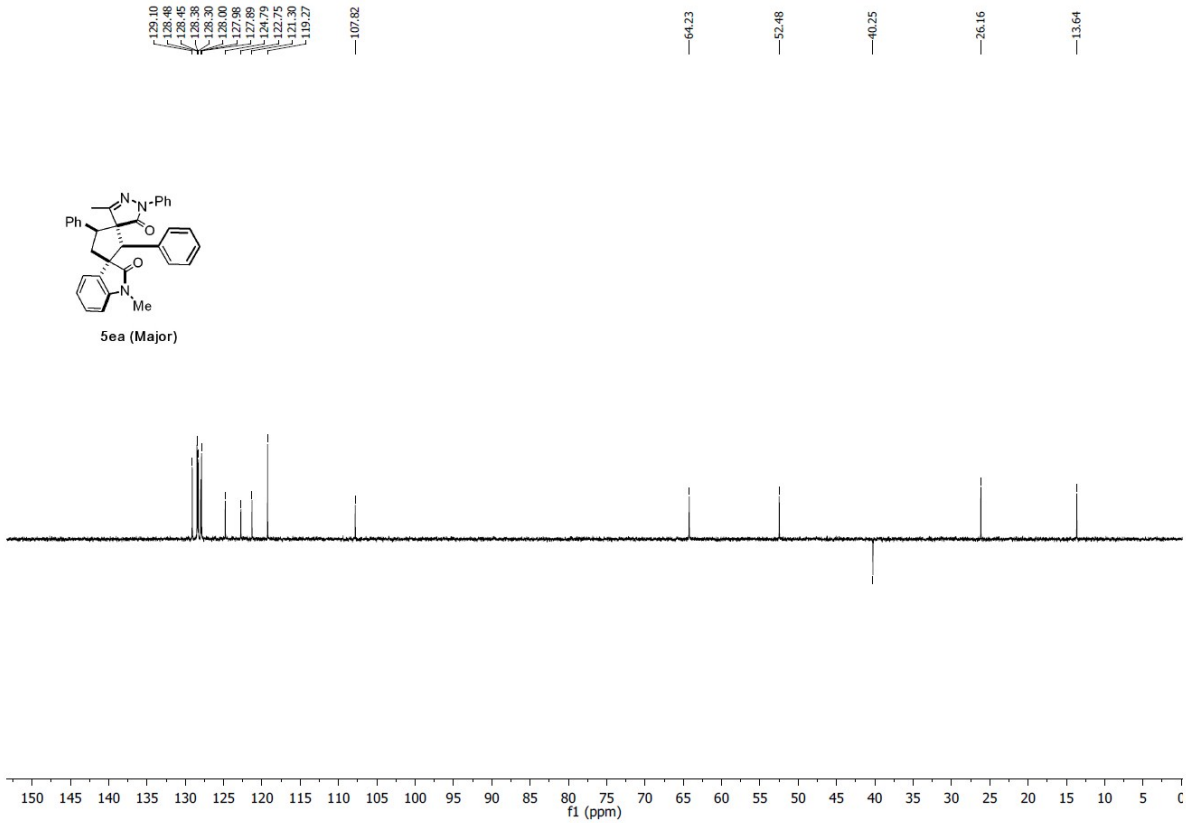
128.70
128.66
128.54
128.04
128.00
127.16
126.39
124.67
122.50
121.28
118.98
108.05
62.75
49.85
40.61
26.18
15.55
Ph
N
N-Ph
O
O
N
Me
5ea (Minor)
150 145 140 135 130 125 120 115 110 105 100 95 90 85 80 75 70 65 60 55 50 45 40 35 30 25 20 15 10 5 0
f1 (ppm)


Ph
N
N-Ph
O
O
N
Me
5ea (Minor)
F1 [ppm]
20
40
60
80
100
120
140
7
6
5
4
3
F2 [ppm]


7.61
7.60
7.58
7.41
7.39
7.35
7.33
7.31
7.30
7.28
Chloroform-d
7.26
7.24
7.22
7.20
7.19
7.11
7.10
7.08
7.07
6.74
6.72
4.38
4.35
4.32
4.18
4.15
4.13
4.12
2.95
2.37
2.34
Ph
N
N-Ph
O
O
N
Me
5ea (Major)
3.01
2.04
2.06
2.71
2.13
6.13
1.02
1.00
2.06
3.00
4.00
10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)


176.43
170.93
158.21
143.28
136.97
129.19
128.57
128.54
128.48
128.39
128.09
128.06
127.98
124.88
122.84
121.39
119.36
107.91
77.48
77.16
Chloroform-d
76.84
69.48
64.32
57.25
52.57
40.34
26.25
13.73
Ph
N
N-Ph
O
O
N
Me
5ea (Major)
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)


129.10
128.48
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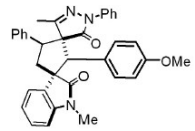
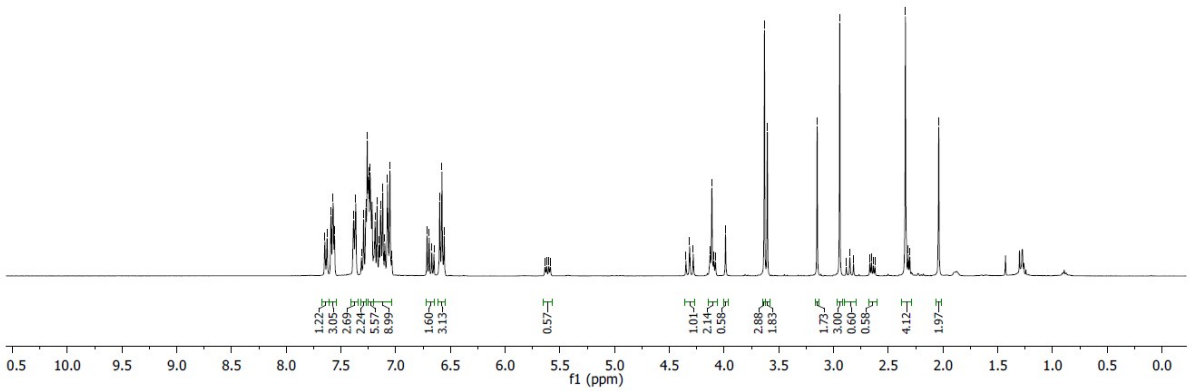
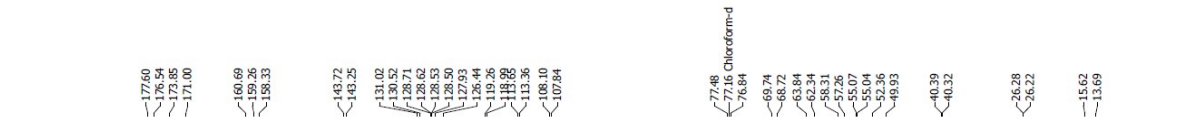
128.38
128.30
128.00
127.98
127.89
124.79
122.75
121.30
119.27
107.82
64.23
52.48
40.25
26.16
13.64
Ph
N
N
Ph
O
O
N
Me
5ea (Major)
150 145 140 135 130 125 120 115 110 105 100 95 90 85 80 75 70 65 60 55 50 45 40 35 30 25 20 15 10 5 0
f1 (ppm)

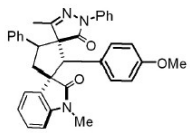

Chloroform-d


N-Ph
Ph
O
OMe
N
Me

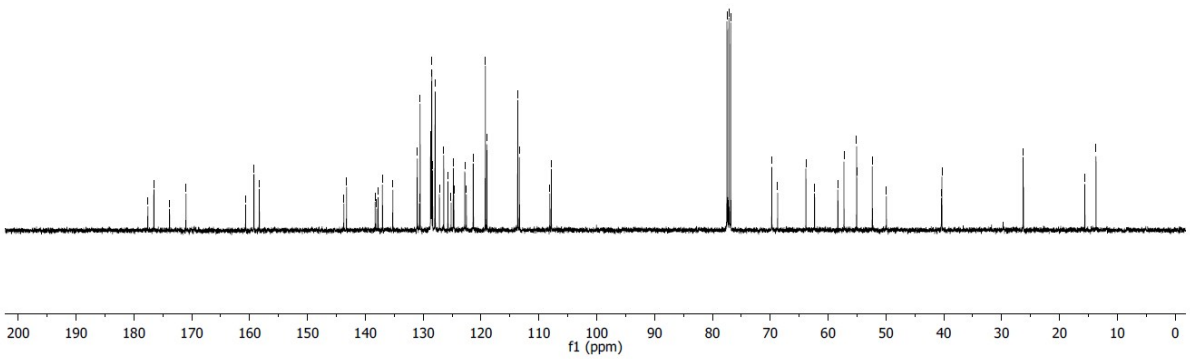
5ec


f1 (ppm)


Chloroform-d


N-Ph
Ph
O
OMe
N
Me

5ec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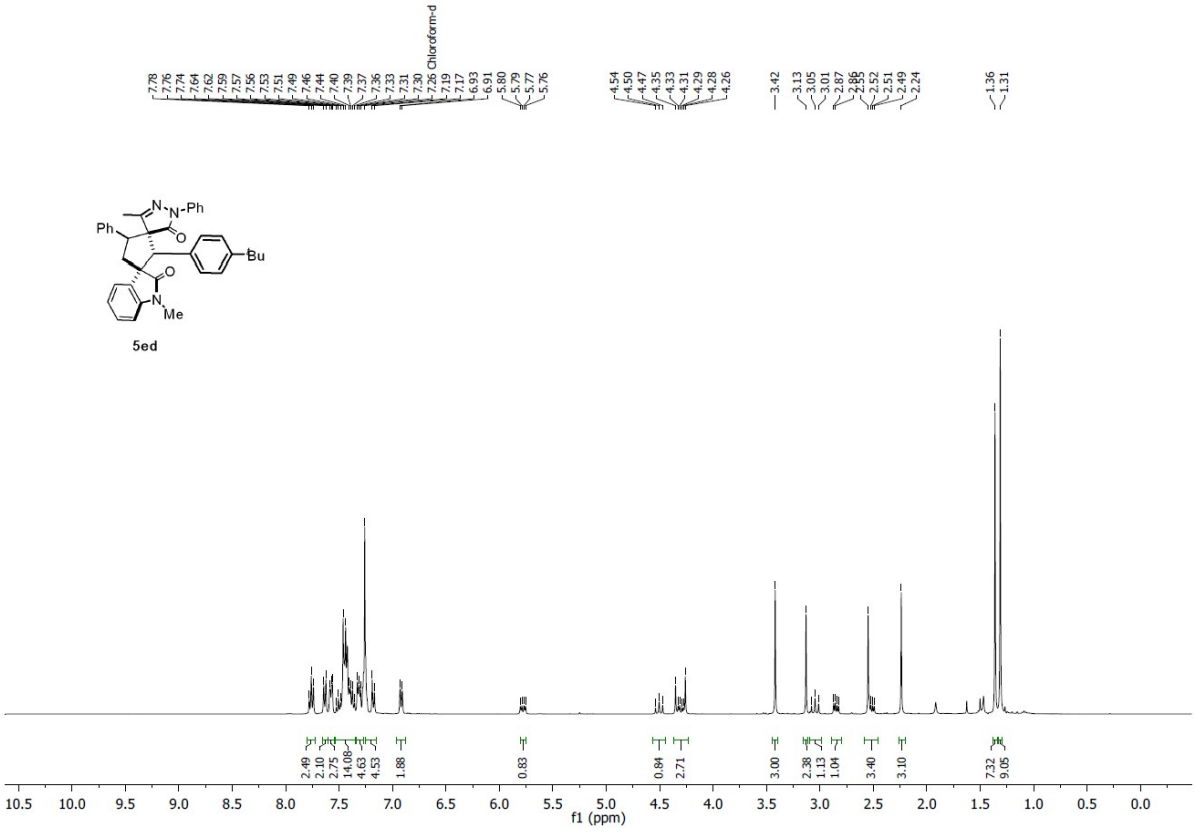
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7.44
7.40
7.39
7.37
7.36
7.33
7.31
7.30
Chloroform-d
7.19
7.17
6.93
6.91
5.80
5.79
5.77
5.76
4.54
4.50
4.47
4.35
4.33
4.31
4.29
4.28
4.26
3.42
3.13
3.05
3.01
2.87
2.52
2.51
2.49
2.24
1.36
1.31
N-Ph
Ph
O
Bu
N
Me
5ed
2.49
2.10
2.75
14.08
4.63
4.53
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1.13
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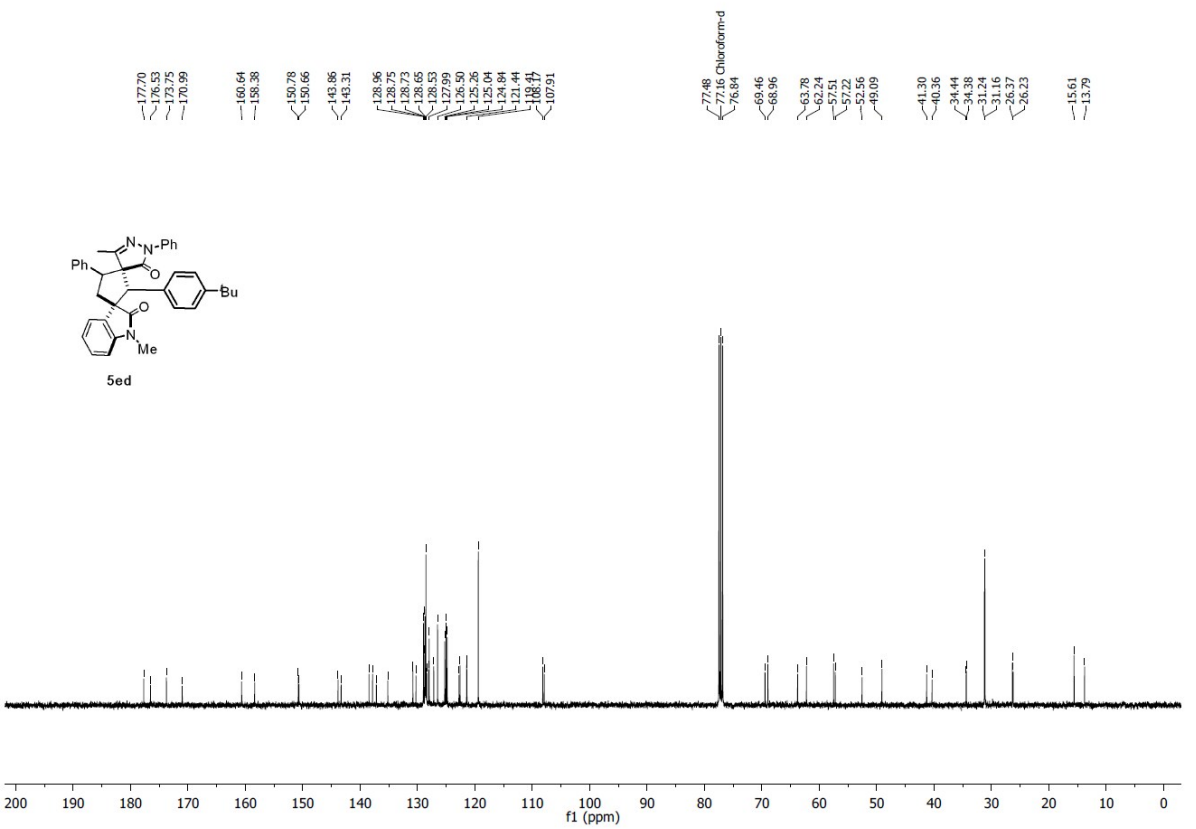
3.10
7.32
9.05
f1 (ppm)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150.66
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143.31
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128.75
128.73
128.65
128.53
127.99
126.50
125.26
125.04
124.84
121.44
108.41
107.91
77.48
77.16
Chloroform-d
76.84
69.46
68.96
63.78
62.24
57.51
57.22
52.56
49.09
41.30
40.36
34.44
34.38
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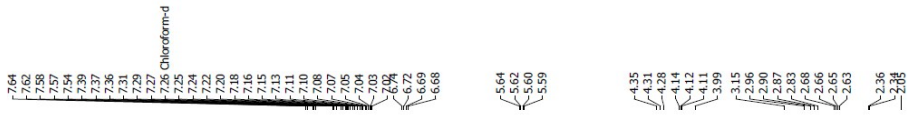
26.23
15.61
13.79
N-Ph
Ph
O
Bu
N
Me
5ed
f1 (ppm)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7.62
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7.36
7.31
7.29
7.27
Chloroform-d
7.26
7.25
7.24
7.22
7.20
7.18
7.16
7.15
7.13
7.11
7.10
7.08
7.07
7.05
7.04
7.03
7.02
6.74
6.72
6.69
6.68
5.64
5.62
5.60
5.59
4.35
4.31
4.28
4.14
4.12
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3.99
3.15
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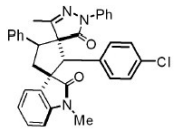
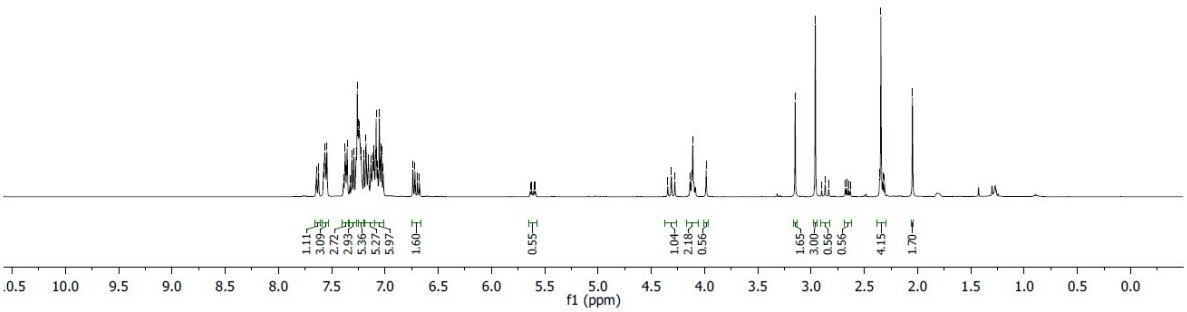
2.68
2.66
2.65
2.63
2.36


N
N-Ph
Ph
O
Cl
O
N
Me

5ee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2.93
5.36
5.27
5.97
1.60
0.55
1.04
2.18
0.56
1.65
3.00
0.56
0.56
4.15
1.70
.0.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


177.29
176.23
173.34
170.74
160.48
158.01
143.60
143.16
131.33
130.80
128.78
128.73
128.62
128.59
128.58
128.56
128.24
127.91
119.28
108.38
108.03
77.48
77.16
Chloroform-d
76.84
69.58
68.47
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58.31
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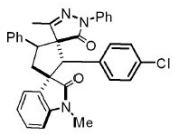
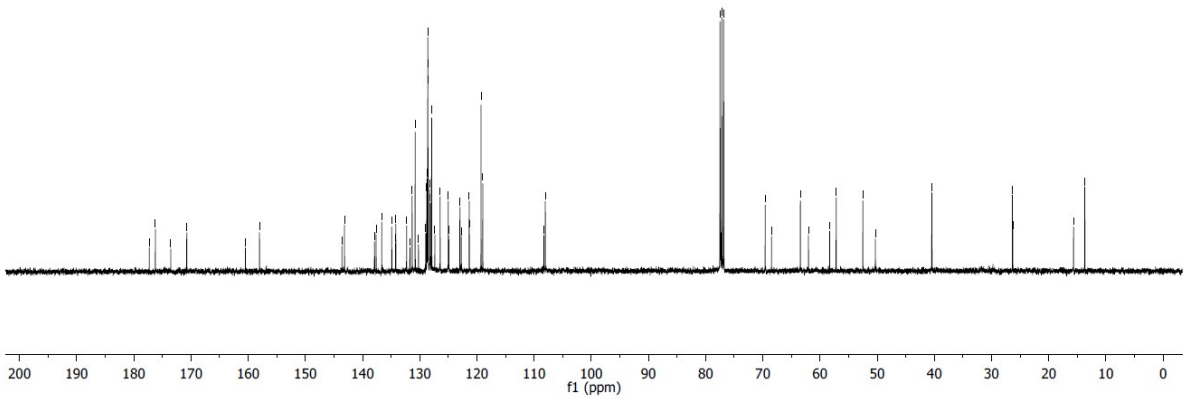
40.44
26.34
26.27
15.65
13.69


N
N-Ph
Ph
O
Cl
O
N
Me

5ee


200
190
180
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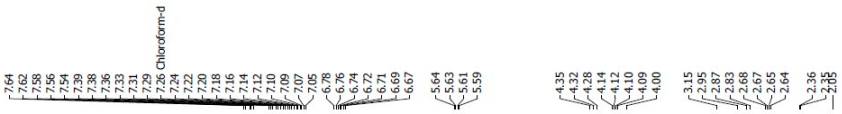
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


7.64
7.62
7.58
7.56
7.54
7.39
7.38
7.36
7.33
7.31
7.29
7.26 Chloroform-d
7.24
7.22
7.20
7.18
7.16
7.14
7.12
7.10
7.09
7.07
7.05
6.78
6.76
6.74
6.72
6.71
6.69
6.67
5.64
5.63
5.61
5.59
4.35
4.32
4.28
4.14
4.12
4.10
4.09
4.00
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2.87
2.83
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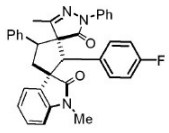
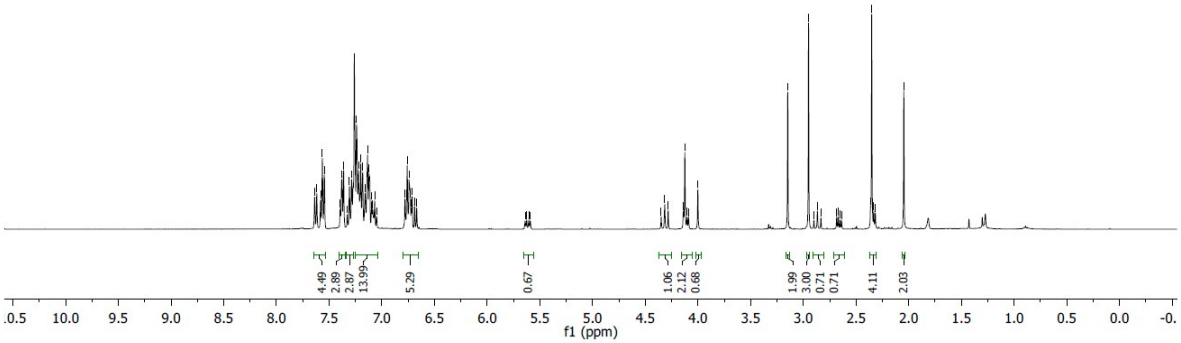
2.67
2.65
2.64
2.36
2.35
2.05


N
N-Ph
Ph
O
F
O
N
Me

5ef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0.67
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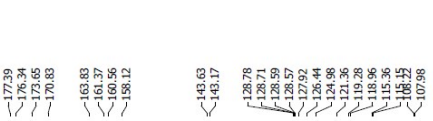
2.12
0.68
1.99
3.00
0.71
0.71
4.11
2.03
f1 (ppm)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170.83
163.83
161.37
160.56
158.12
145.63
143.17
128.78
128.71
128.59
128.57
127.92
126.44
124.98
121.36
119.28
118.96
115.36
77.48
77.16 Chloroform-d
76.84
69.72
68.57
63.51
62.06
58.37
57.23
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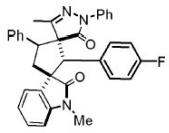
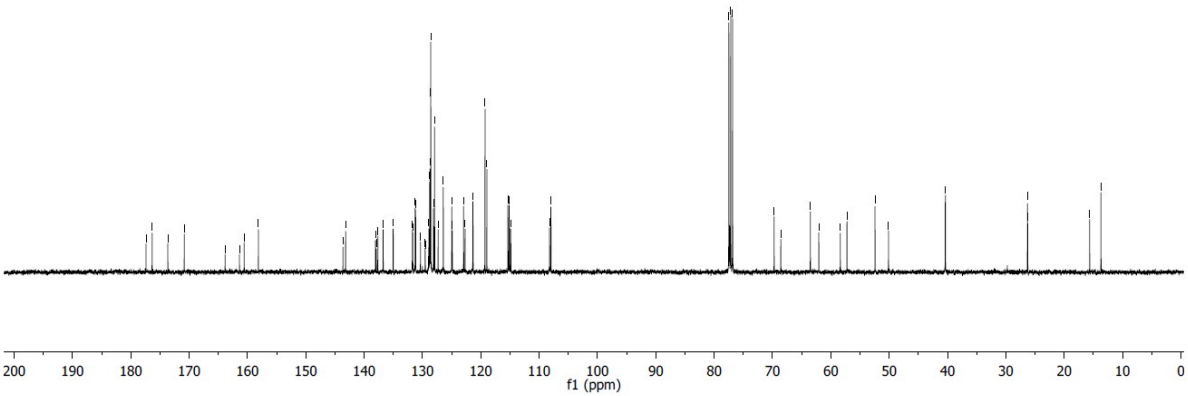
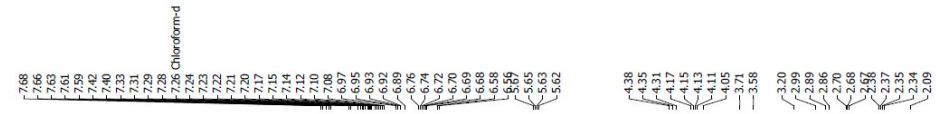
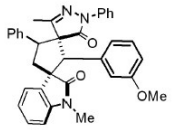
50.10
40.37
26.29
26.25
15.63
13.69


N
N-Ph
Ph
O
F
O
N
Me

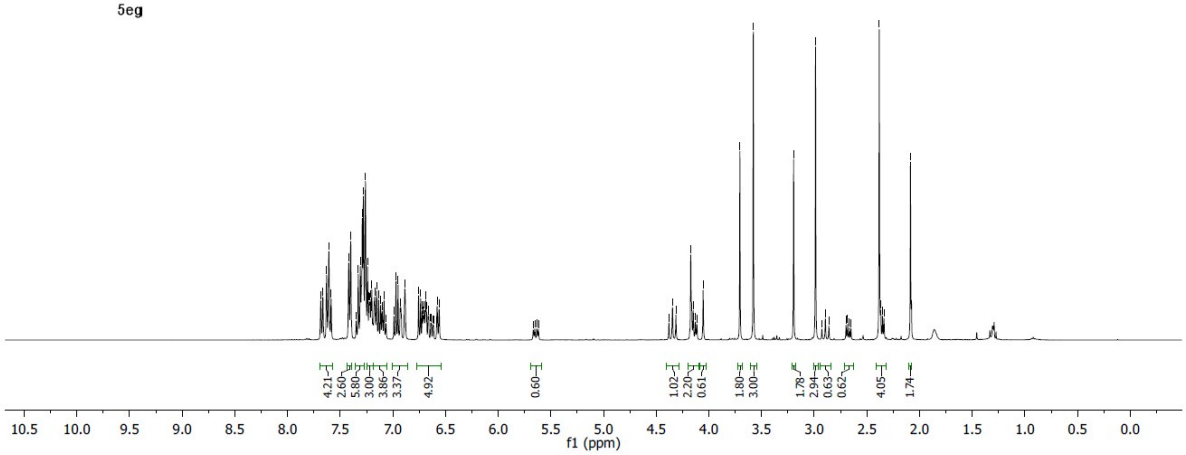
5ef


f1 (ppm)


Chloroform-d


Ph
N
N-Ph
O
O
N
Me
OMe

5eg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3.37
4.92
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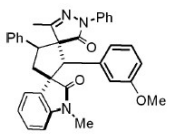
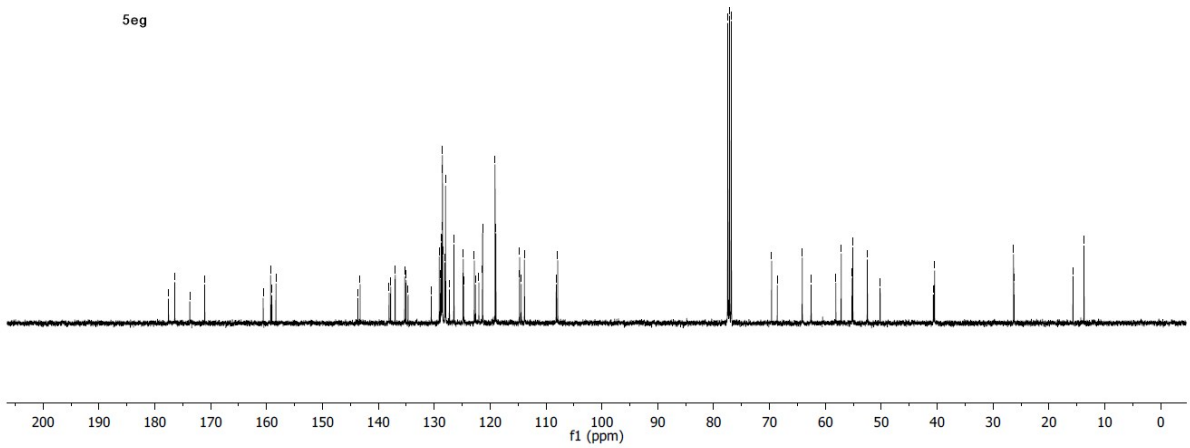
1.02
2.20
0.61
1.80
3.00
1.78
2.94
0.63
0.62
4.05
1.74
10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)


177.57
176.43
173.74
171.08
160.61
159.27
159.06
158.32
143.67
143.26
128.75
128.66
128.55
128.52
128.47
127.97
126.47
121.35
121.34
119.11
118.96
114.72
108.16
107.91
77.48
77.16 Chloroform-d
76.84
69.63
68.60
64.14
62.55
58.13
57.17
55.23
55.10
52.49
50.21
40.68
40.47
26.32
26.24
15.69
13.72


Ph
N
N-Ph
O
O
N
Me
OMe

5eg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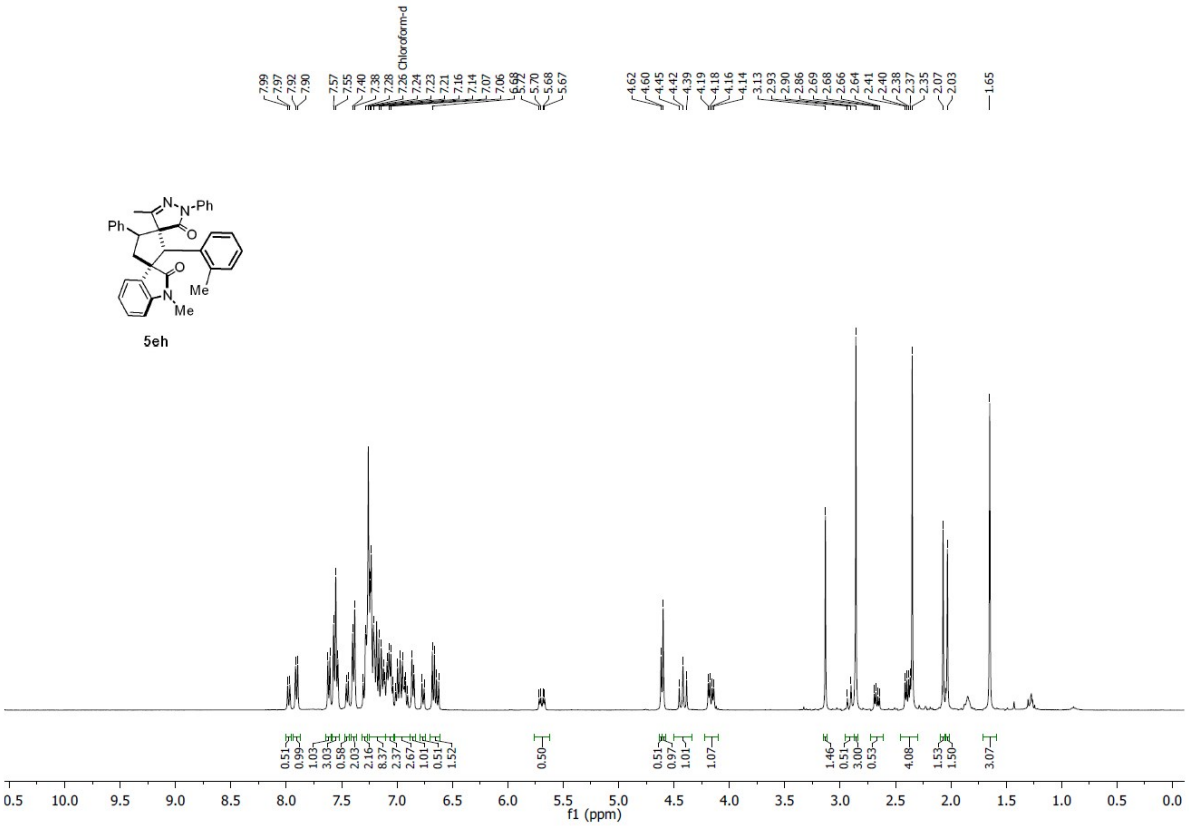
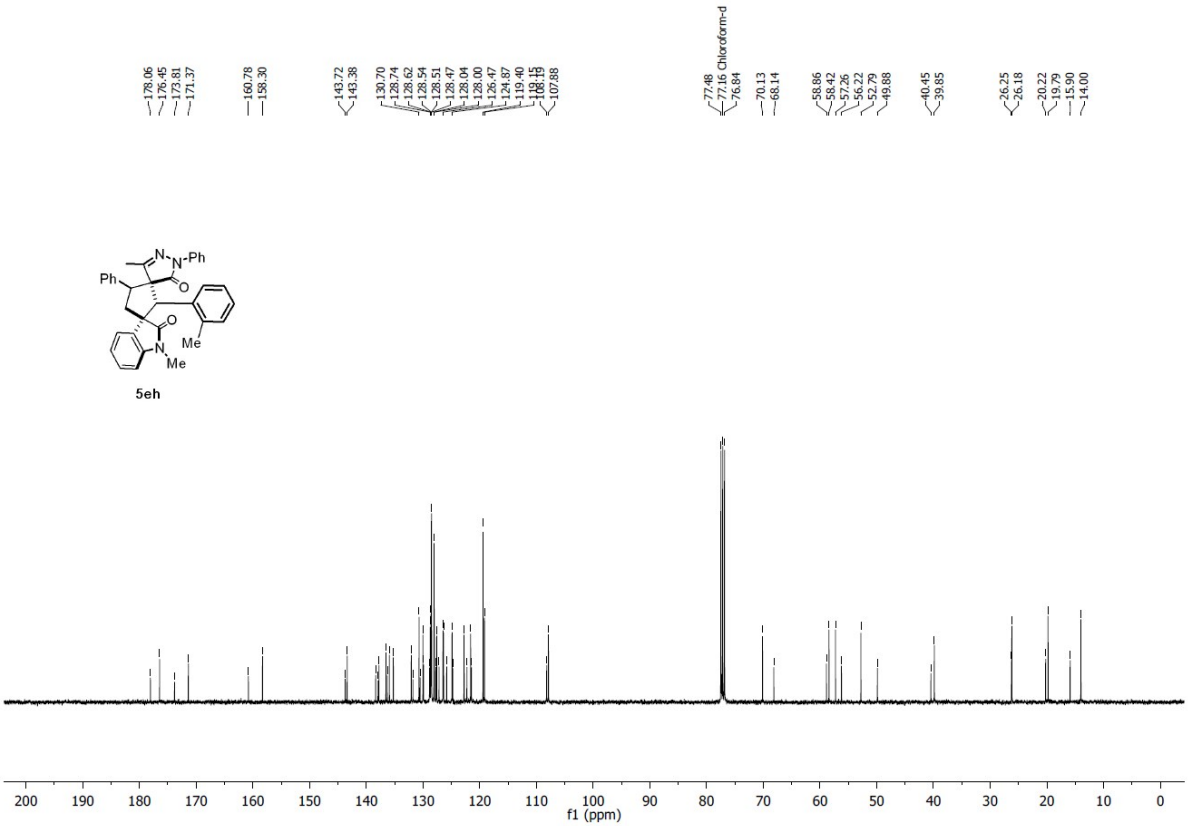
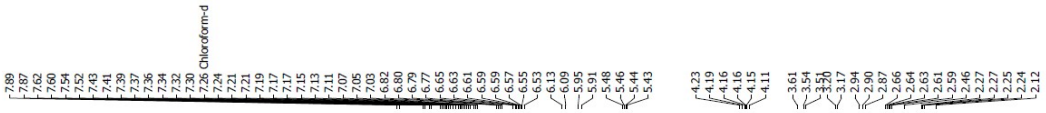
f1 (ppm)


Chloroform-d
5eh
f1 (ppm)


Chloroform-d
5eh
f1 (ppm)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7.52
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7.41
7.39
7.37
7.36
7.34
7.32
7.30
7.26 Chloroform-d
7.24
7.21
7.21
7.19
7.17
7.17
7.15
7.13
7.11
7.07
7.05
7.03
6.82
6.80
6.79
6.77
6.65
6.63
6.61
6.59
6.59
6.57
6.55
6.53
6.13
6.09
5.95
5.91
5.48
5.46
5.44
5.43
4.23
4.19
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4.16
4.15
4.11
3.61
3.54
3.52
3.50
3.17
2.94
2.90
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2.63
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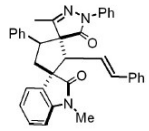
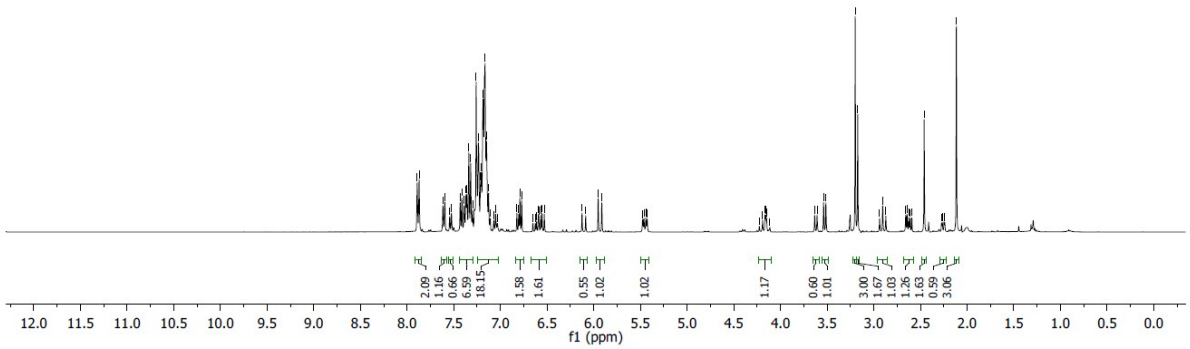
2.27
2.25
2.24
2.12


N
N-Ph
Ph
O
O
Ph
N
Me

5ei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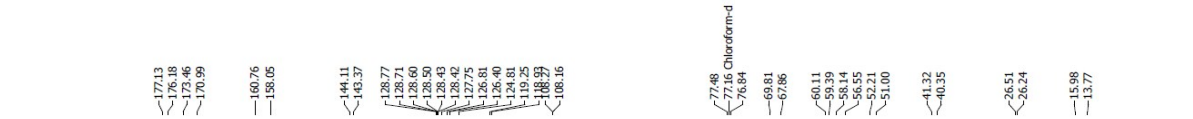
1.58
1.61
0.55
1.02
1.02
1.17
0.60
1.01
3.00
1.67
1.03
1.26
1.63
0.59
3.06
12.0
11.5
11.0
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


177.13
176.18
173.46
170.99
160.76
158.05
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143.37
128.77
128.71
128.60
128.50
128.43
128.42
127.75
126.81
126.40
124.81
119.25
118.93
118.27
108.16
77.48
77.16 Chloroform-d
76.84
69.81
67.86
60.11
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58.14
56.55
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51.00
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40.35
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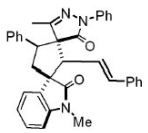
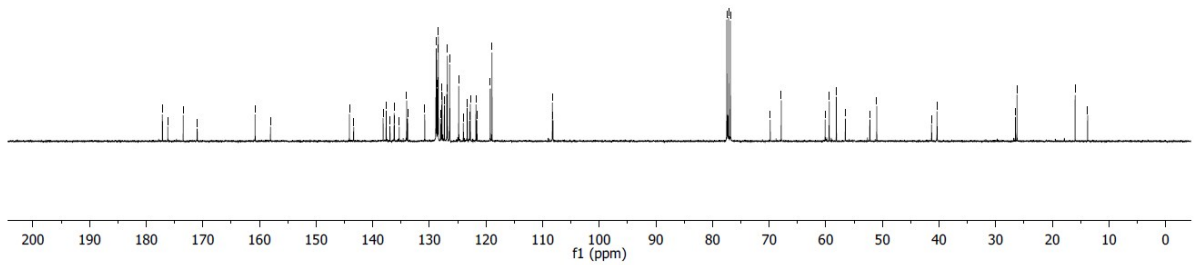
26.24
15.98
13.77


N
N-Ph
Ph
O
O
Ph
N
Me

5ei


200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


7.67
7.65
7.63
7.62
7.59
7.56
7.55
7.54
7.52
7.49
7.45
7.43
7.41
7.40
7.36
7.35
7.33
7.33
7.32
7.30
7.29
7.27
7.26 Chloroform-d
7.25
7.24
7.22
7.20
7.17
7.15
7.12
7.10
7.08
7.06
7.04
7.03
7.01
6.99
6.71
6.69
6.61
6.59
5.72
5.71
5.69
5.68
4.43
4.39
4.35
4.21
4.19
4.17
4.16
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2.68
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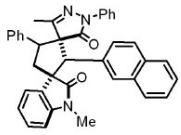
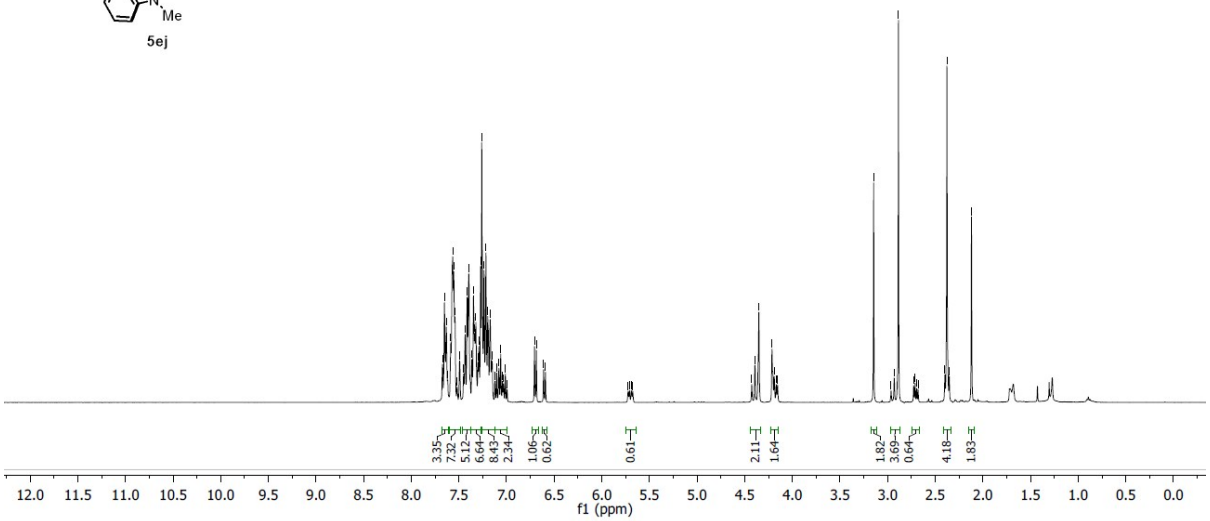
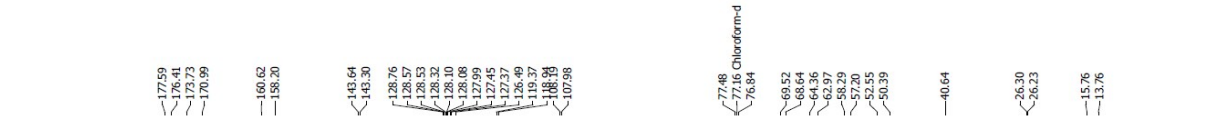
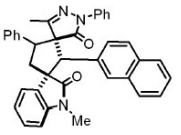
2.35
2.12


N
N
Ph
Ph
O
O
N
Me

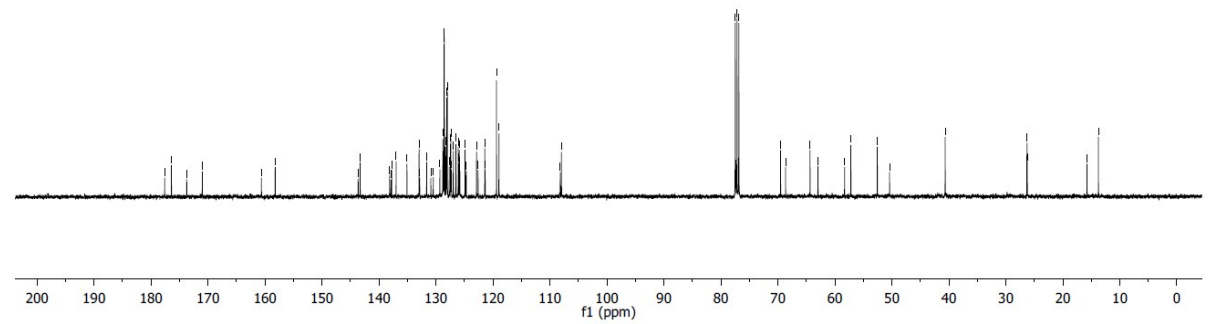
5ej

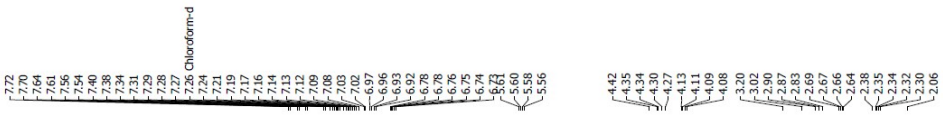

3.35
7.32
5.12
6.64
8.43
2.34
1.06
0.62
0.61
2.11
1.64
1.82
3.69
0.64
4.18
1.83
12.0 11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)

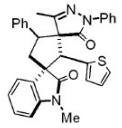

177.59
176.41
173.73
170.99
160.62
158.20
143.64
143.30
128.76
128.57
128.53
128.32
128.10
128.08
127.99
127.45
127.37
126.49
119.37
108.94
107.98
77.48
77.16 Chloroform-d
76.84
69.52
68.64
64.36
62.97
58.29
57.20
52.55
50.39
40.64
26.30
26.23
15.76
13.76


N
N
Ph
Ph
O
O
N
Me

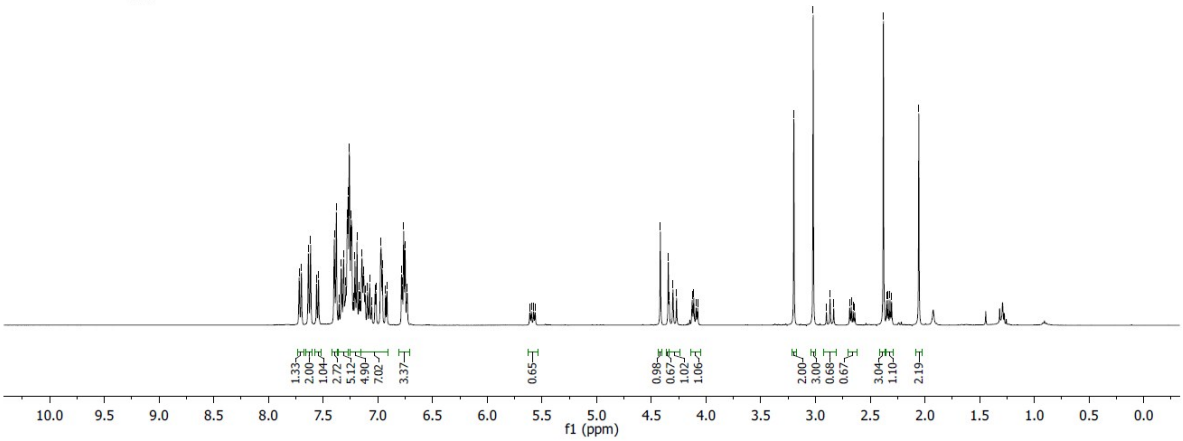
5ej

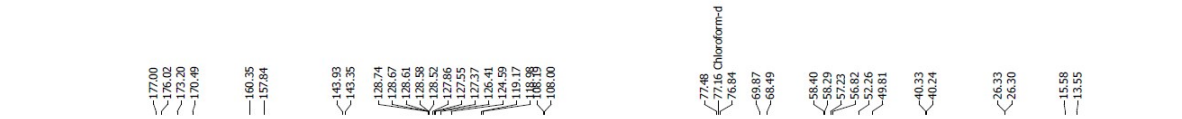

200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

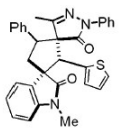

Chloroform-d


Ph
N
N-Ph
O
S
O
N
Me

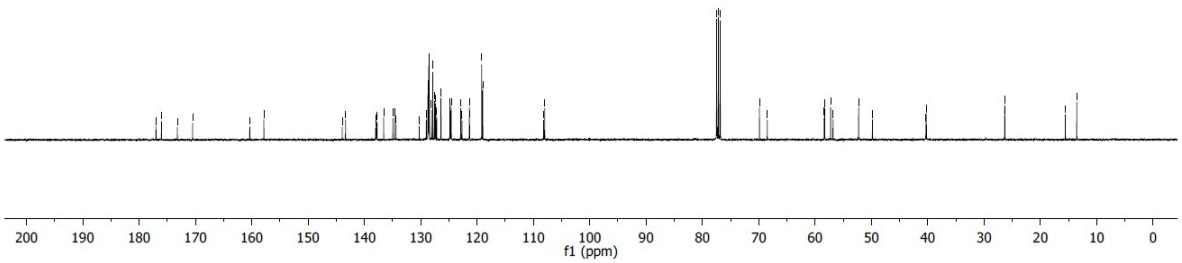
5ek

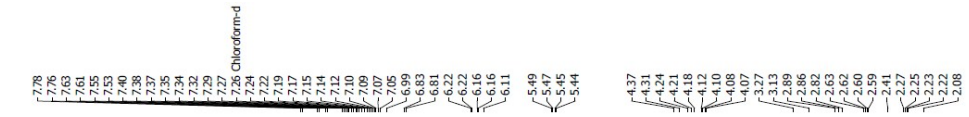

1.33
2.00
1.04
2.72
5.12
4.90
7.02
3.37
0.65
0.98
0.67
1.02
1.06
2.00
3.00
0.68
0.67
3.04
1.10
2.19
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

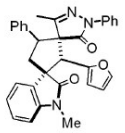

Chloroform-d
f1 (ppm)


Ph
N
N-Ph
O
S
O
N
Me

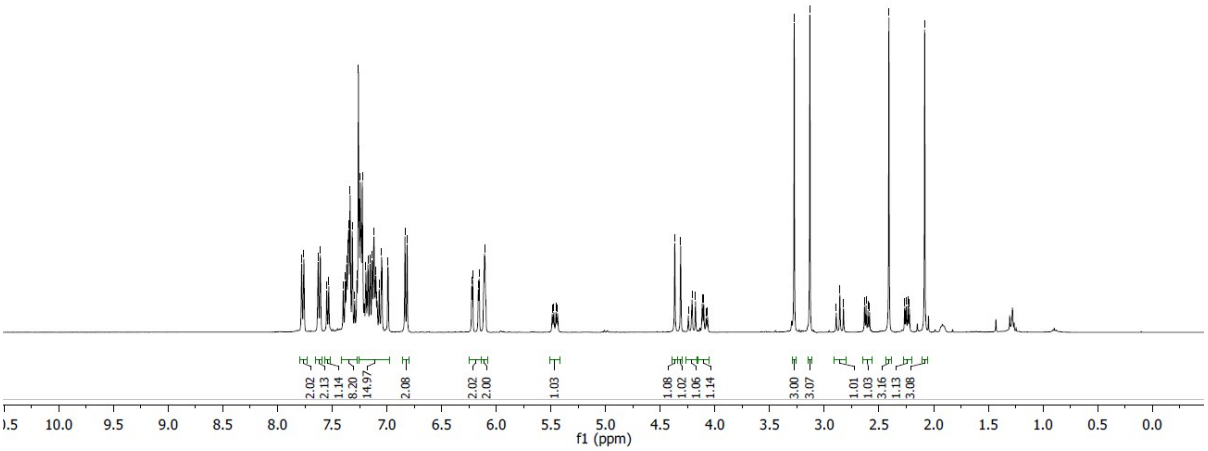
5ek


200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


Chloroform-d


Ph
N
N-Ph
O
O
O
N
Me

5el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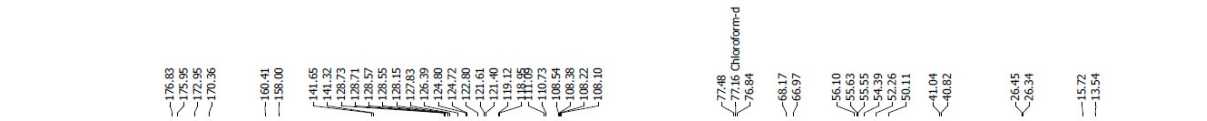
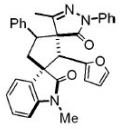
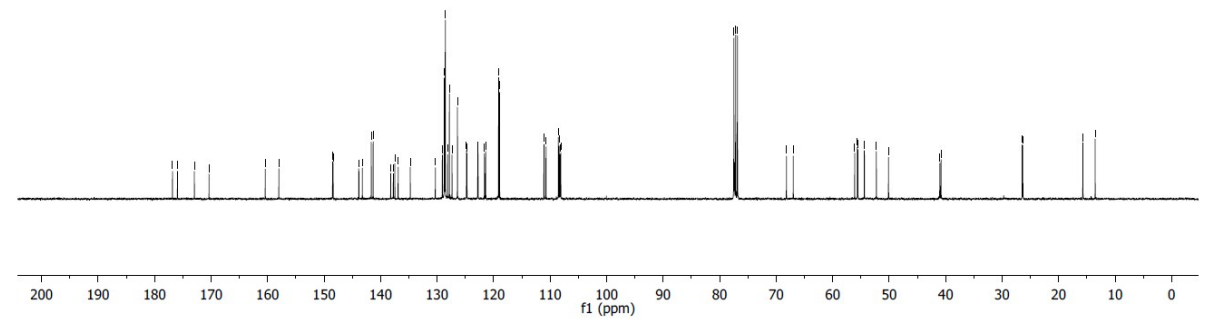
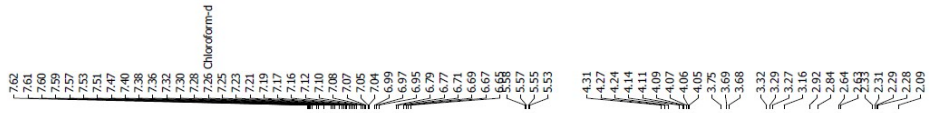
2.13
1.14
8.20
14.97
2.08
2.02
2.00
1.03
1.08
1.02
1.06
1.14
3.00
3.07
1.01
1.03
3.16
1.13
3.08
f1 (ppm)

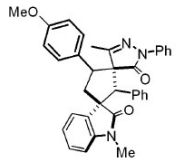

Chloroform-d


Ph
N
N-Ph
O
O
O
N
Me

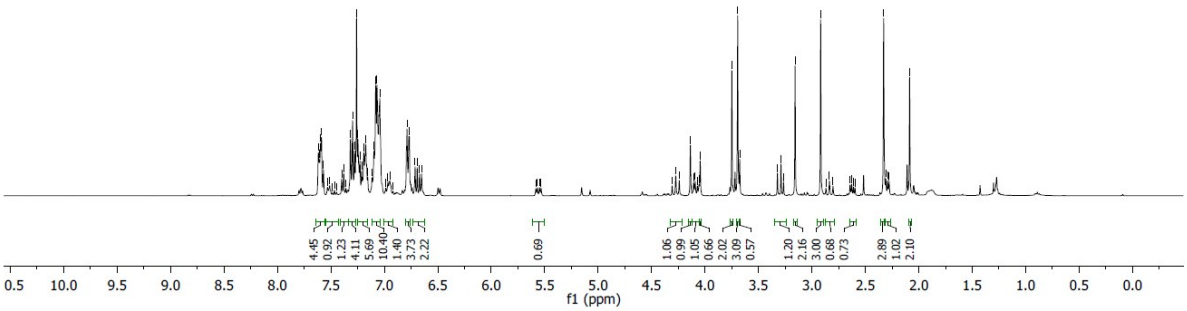
5el


f1 (ppm)

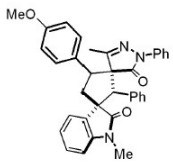

7.62
7.61
7.60
7.59
7.57
7.53
7.51
7.47
7.40
7.38
7.36
7.32
7.30
7.28
Chloroform-d
7.26
7.25
7.23
7.21
7.19
7.17
7.16
7.12
7.10
7.08
7.07
7.05
7.04
6.99
6.97
6.95
6.79
6.77
6.71
6.69
6.67
5.58
5.57
5.55
5.53
4.31
4.27
4.24
4.14
4.11
4.09
4.07
4.06
4.05
3.75
3.69
3.68
3.32
3.29
3.27
3.16
2.92
2.84
2.64
2.63
2.33
2.31
2.29
2.28
2.09


MeO
N
N-Ph
O
Ph
O
N
Me

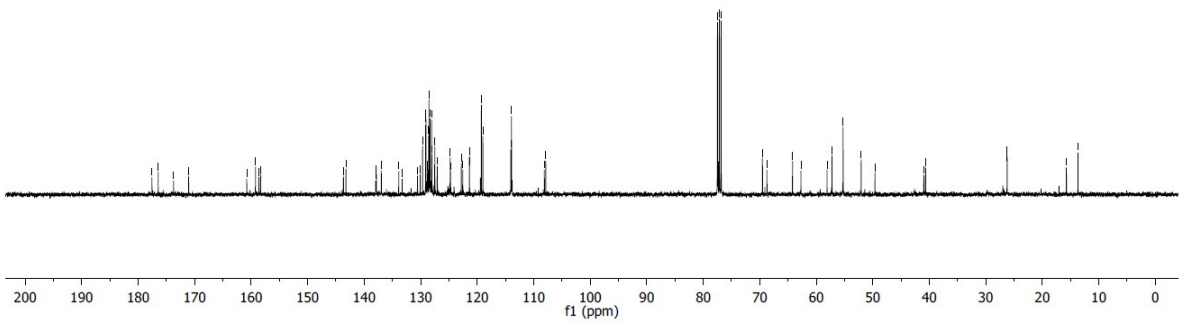
5ga

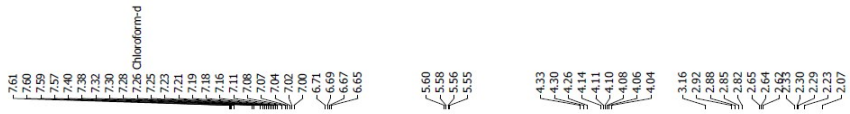

4.45
0.92
1.23
4.11
5.69
10.40
1.40
3.73
2.22
0.69
1.06
0.99
1.05
0.66
2.02
3.09
0.57
1.20
2.16
3.00
0.68
0.73
2.89
1.02
2.10
0.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)

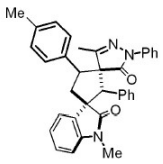

177.56
176.44
173.73
171.06
160.77
159.24
158.63
158.32
143.66
143.22
129.67
129.12
129.05
128.61
128.52
128.35
128.06
127.51
119.26
114.88
114.78
113.93
108.11
107.86
77.48
77.16
Chloroform-d
76.84
69.51
68.72
64.70
62.71
58.08
57.21
55.24
52.05
49.61
40.95
40.64
26.24
26.21
15.73
13.68


MeO
N
N-Ph
O
Ph
O
N
Me

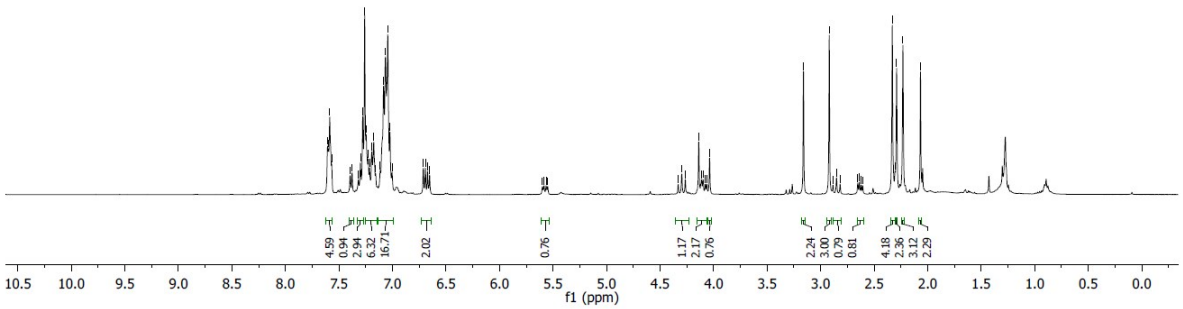
5ga


200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


7.61
7.60
7.59
7.57
7.40
7.38
7.32
7.30
7.28
7.26 Chloroform-d
7.25
7.23
7.21
7.19
7.18
7.16
7.11
7.08
7.07
7.04
7.02
7.00
6.71
6.69
6.67
6.65
5.60
5.58
5.56
5.55
4.33
4.30
4.26
4.14
4.11
4.10
4.08
4.06
4.04
3.16
2.92
2.88
2.85
2.82
2.65
2.64
2.33
2.30
2.29
2.23
2.07


Me
N
N-Ph
O
Ph
O
N
Me

5ha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0.94
2.94
6.32
16.71
2.02
0.76
1.17
2.17
0.76
2.24
3.00
0.79
0.81
4.18
2.36
3.12
2.29
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


177.60
176.47
173.77
171.06
160.73
158.30
143.72
143.28
129.71
129.41
129.27
129.19
128.62
128.51
128.36
128.07
127.85
126.35
119.33
119.08
118.11
107.87
77.48
77.16 Chloroform-d
76.84
69.47
68.74
64.39
62.80
58.10
57.27
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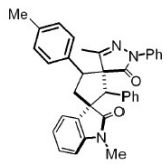
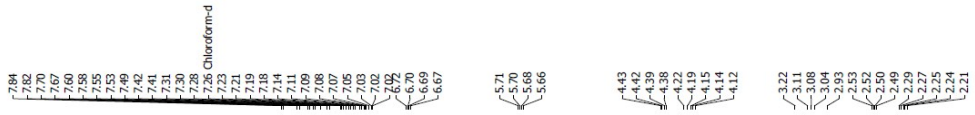
26.26
26.23
21.13
21.09
15.71
13.68


Me
N
N-Ph
O
Ph
O
N
Me

5ha

200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)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7.70
7.67
7.60
7.58
7.55
7.53
7.49
7.42
7.41
7.31
7.30
7.28
7.26 Chloroform-d
7.23
7.21
7.19
7.18
7.14
7.11
7.09
7.08
7.07
7.05
7.03
7.02
6.72
6.70
6.69
6.67
5.71
5.70
5.68
5.66
4.43
4.42
4.39
4.38
4.22
4.19
4.15
4.14
4.12
3.22
3.11
3.08
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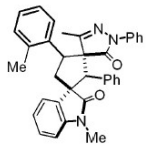
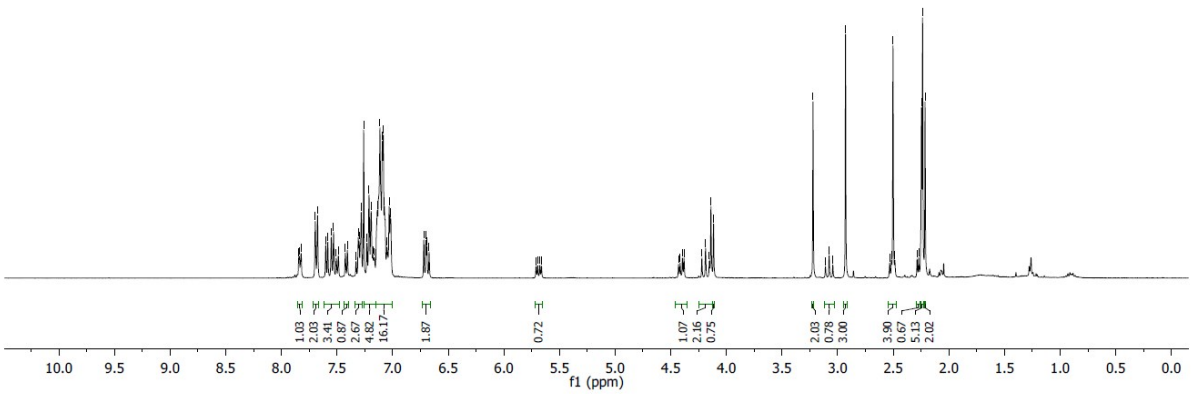
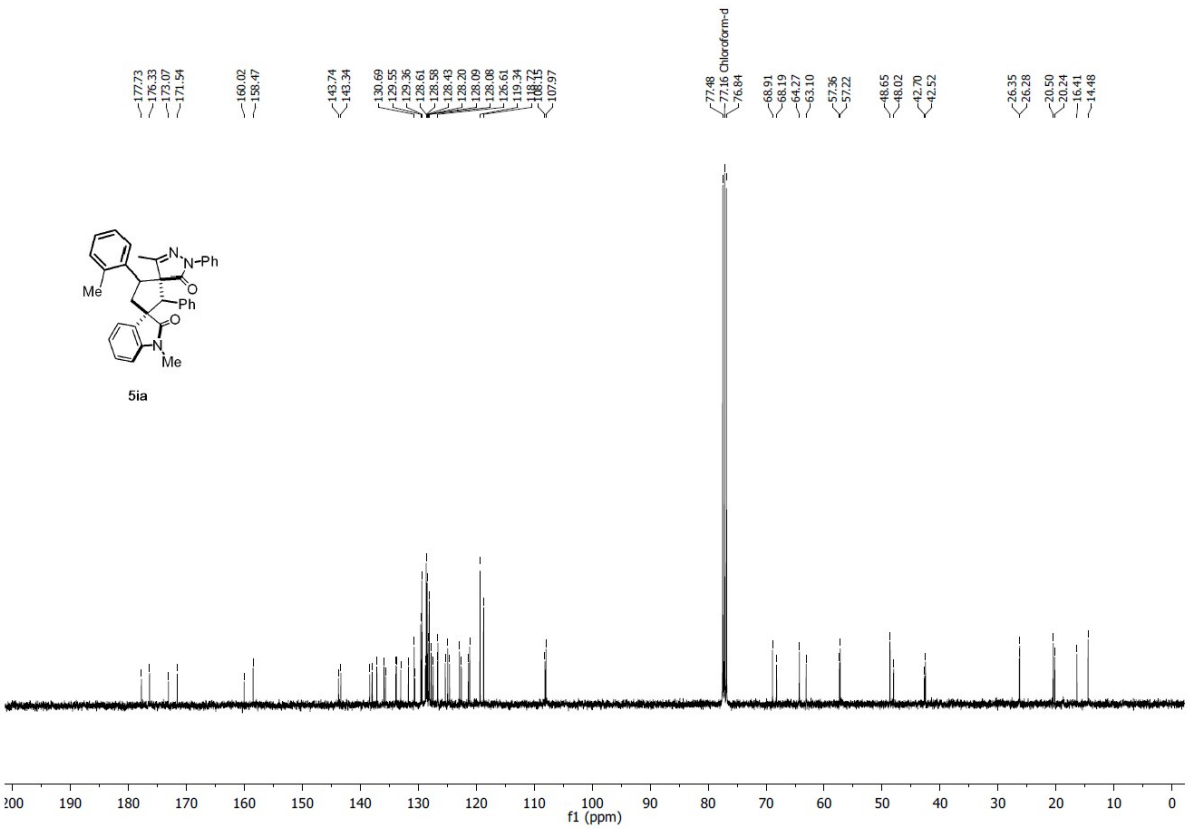
2.29
2.27
2.25
2.24
2.21


N
N-Ph
O
Me
Ph
O
N
Me

5ia


1.03
2.03
3.41
0.87
2.67
4.82
16.17
1.87
0.72
1.07
2.16
0.75
2.03
0.78
3.00
3.90
0.67
5.13
2.02
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)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171.54
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158.47
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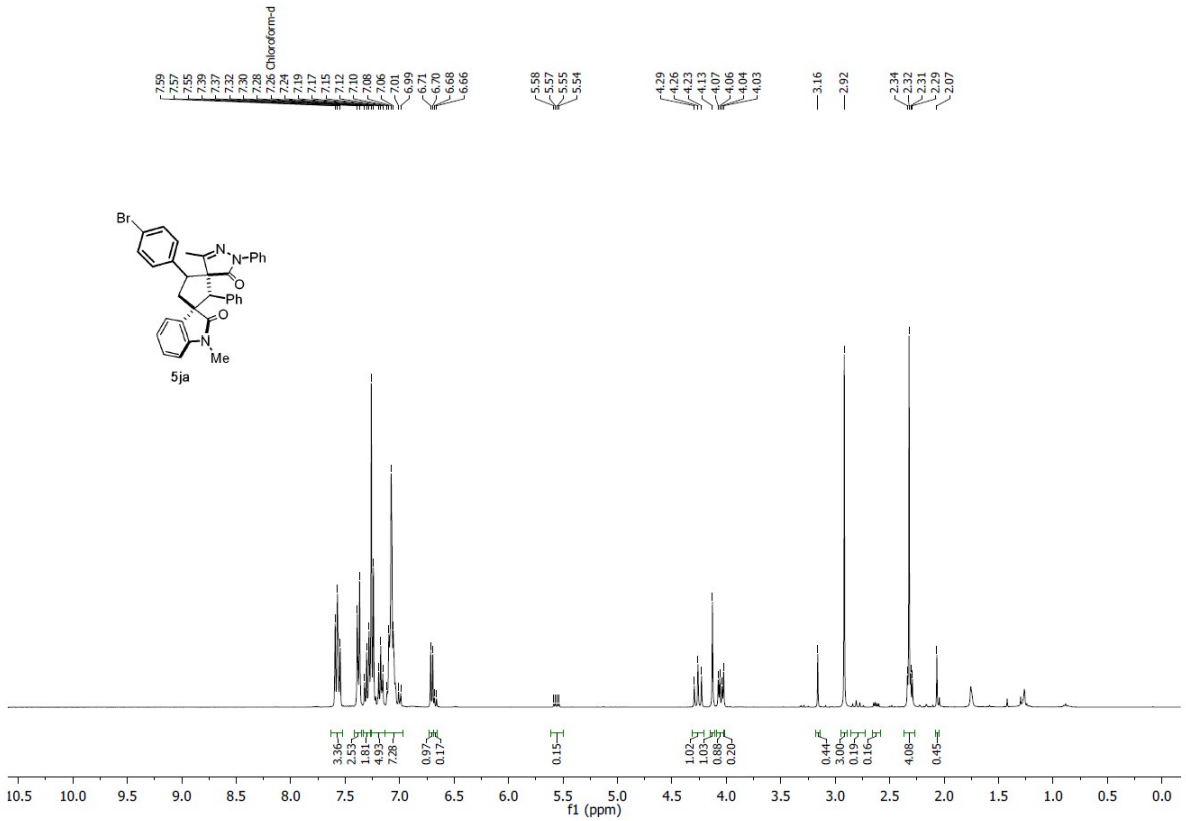
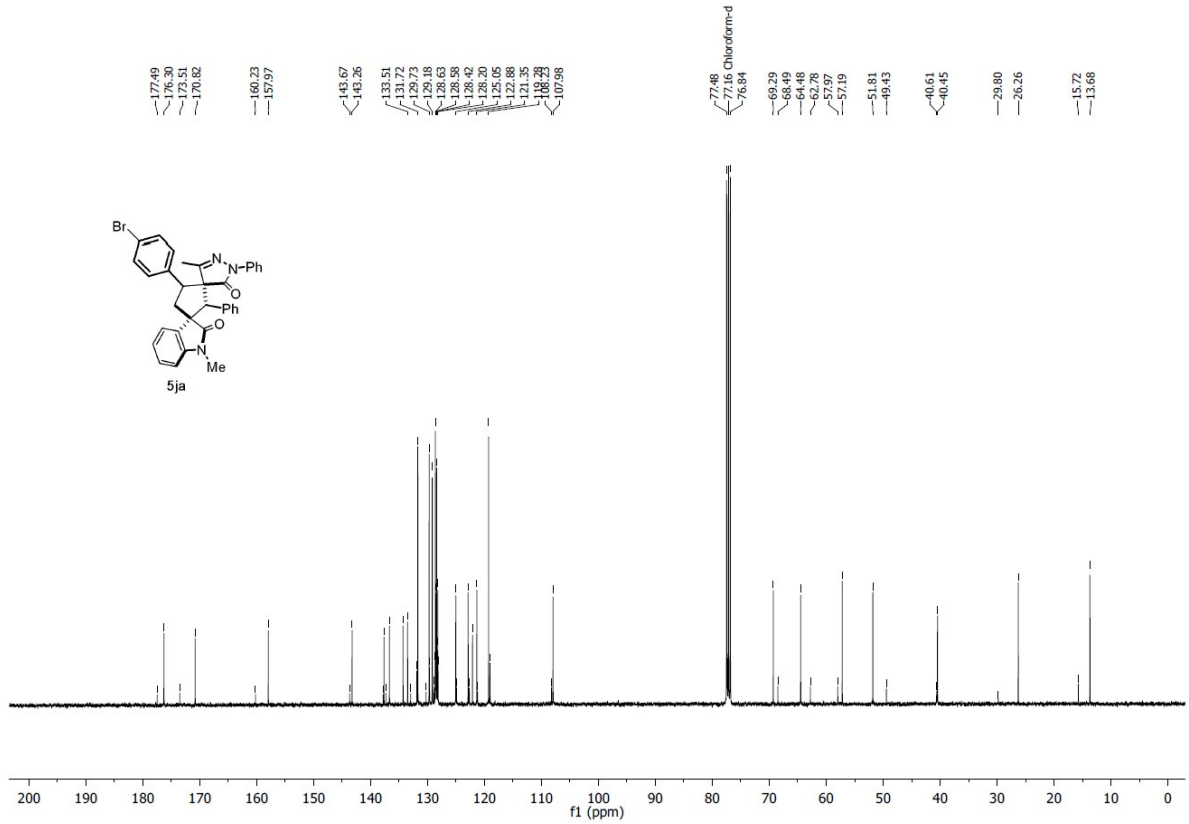
143.34
130.69
129.55
129.36
128.61
128.58
128.43
128.20
128.09
128.08
126.61
119.34
107.97
77.48
77.16 Chloroform-d
76.84
68.91
68.19
64.27
63.10
57.36
57.22
48.65
48.02
42.70
42.52
26.35
26.28
20.50
20.24
16.41
14.48
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

N
N-Ph
O
Me
Ph
O
N
Me

5ia


7.59
7.57
7.55
7.39
7.37
7.32
7.30
7.28 Chloroform-d
7.26
7.24
7.19
7.17
7.15
7.12
7.10
7.08
7.06
7.01
6.99
6.71
6.70
6.68
6.66
5.58
5.57
5.55
5.54
4.29
4.26
4.23
4.13
4.07
4.06
4.04
4.03
3.16
2.92
2.34
2.32
2.31
2.29
2.07
Br
N
N-Ph
O
Ph
O
N
Me
5ja
3.36
1.53
1.81
4.93
7.28
0.97
0.17
0.15
1.02
1.03
0.88
0.20
0.44
3.00
0.19
0.16
4.08
0.45
f1 (ppm)


177.49
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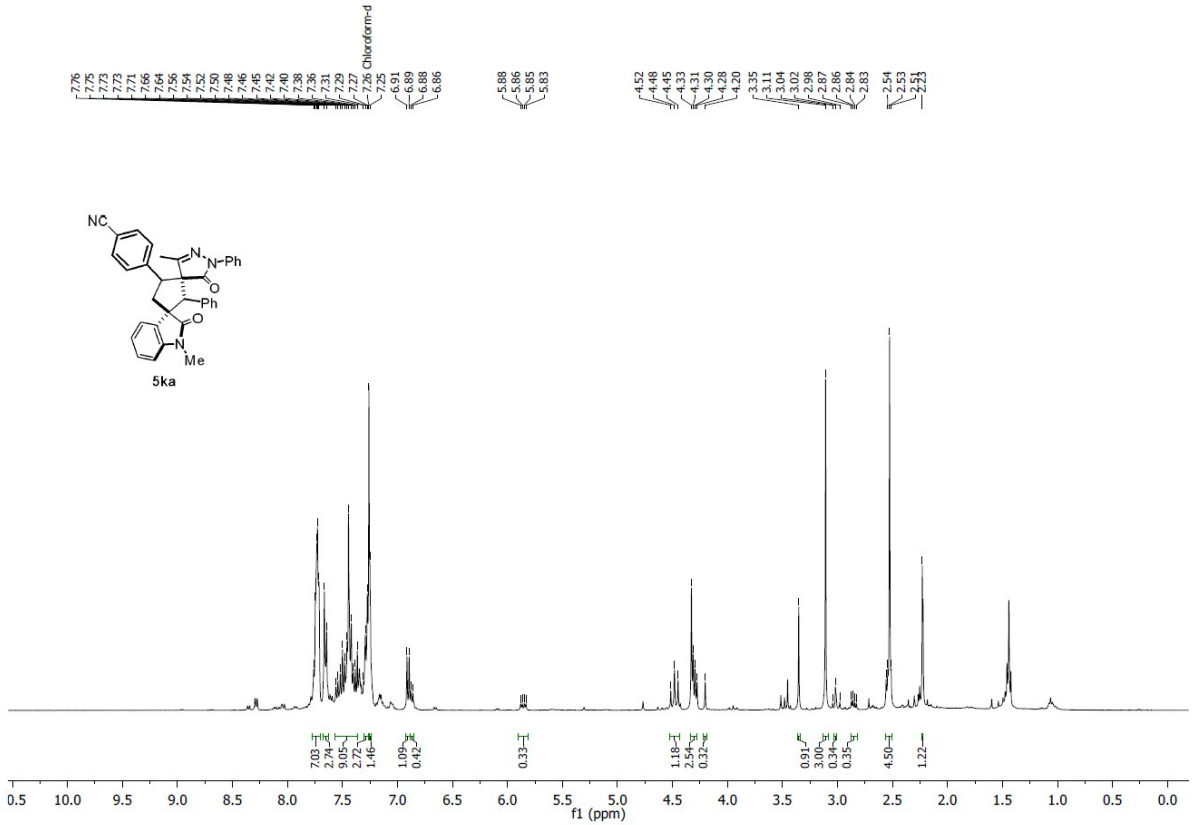
170.02
160.23
157.97
143.67
143.26
133.51
131.72
129.73
129.18
128.63
128.58
128.42
128.20
125.05
122.88
121.35
108.23
107.98
77.48
77.16 Chloroform-d
76.84
69.29
68.49
64.48
62.78
57.97
57.19
51.81
49.43
40.61
40.45
29.80
26.26
15.72
13.68
Br
N
N-Ph
O
Ph
O
N
Me
5ja
f1 (ppm)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7.75
7.73
7.73
7.71
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7.54
7.52
7.50
7.48
7.46
7.45
7.42
7.40
7.38
7.36
7.31
7.29
7.27
7.26 Chloroform-d
7.25
6.91
6.89
6.88
6.86
5.88
5.86
5.85
5.83
4.52
4.48
4.45
4.33
4.31
4.30
4.28
4.20
3.35
3.11
3.04
3.02
2.98
2.87
2.86
2.84
2.83
2.54
2.53
2.51
2.33
NC
N
N-Ph
O
Ph
O
N
Me
5ka
7.03
2.74
9.05
2.72
1.46
1.09
0.42
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2.54
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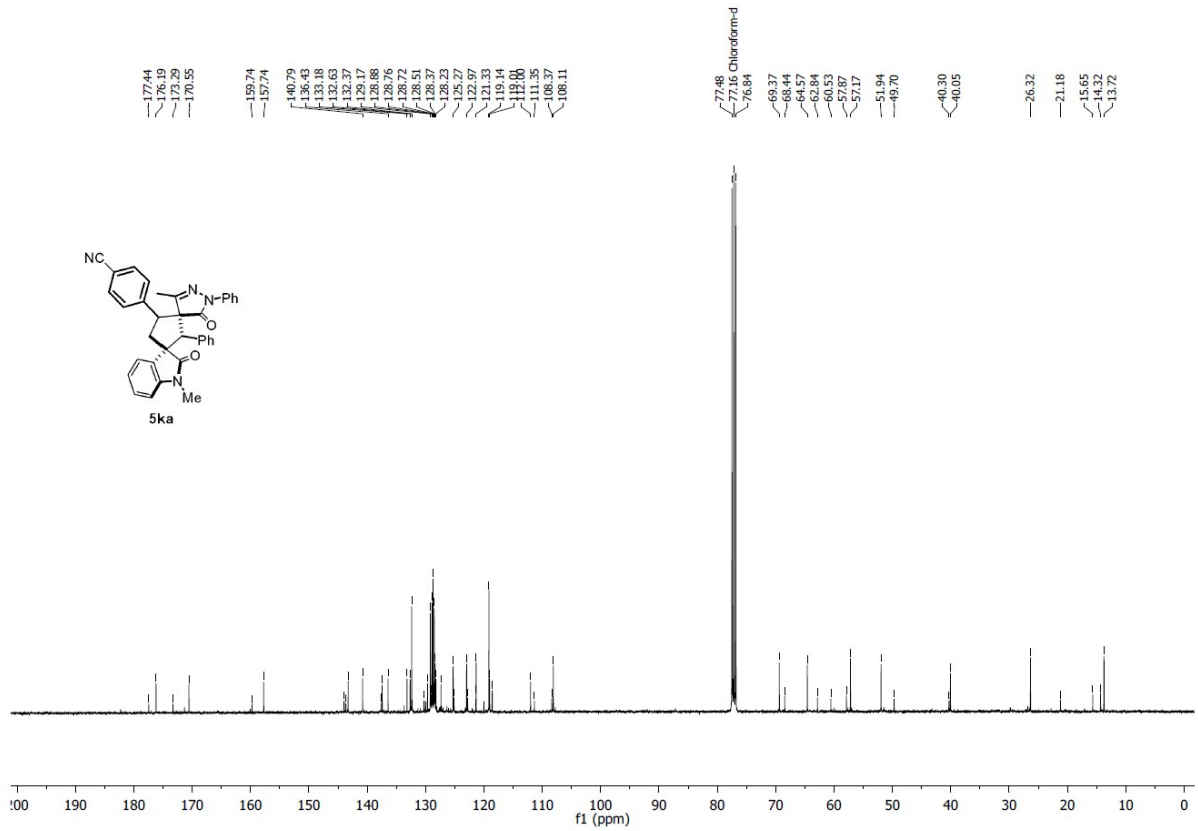
0.91
3.00
0.34
0.35
4.50
1.22
f1 (ppm)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176.19
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157.74
140.79
136.43
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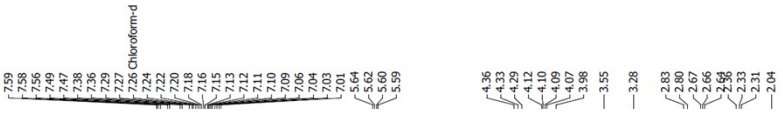
132.63
132.37
129.17
128.88
128.76
128.72
128.51
128.37
128.23
125.27
122.97
121.33
119.14
119.01
112.00
111.35
108.37
108.11
77.48
77.16 Chloroform-d
76.84
69.37
68.44
64.57
62.84
60.53
57.87
57.17
51.94
49.70
40.30
40.05
26.32
21.18
15.65
14.32
13.72
NC
N
N-Ph
O
Ph
O
N
Me
5ka
f1 (ppm)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7.58
7.56
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7.47
7.38
7.36
7.29
7.27
7.26 Chloroform-d
7.24
7.22
7.20
7.18
7.16
7.15
7.13
7.12
7.11
7.10
7.09
7.06
7.04
7.03
7.01
5.64
5.62
5.60
5.59
4.36
4.33
4.29
4.12
4.10
4.09
4.07
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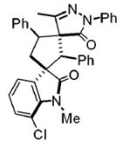
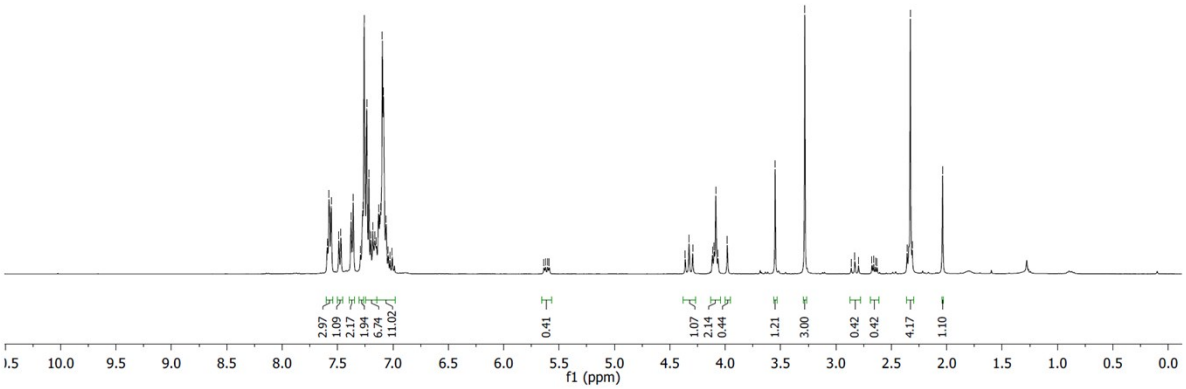
3.28
2.83
2.80
2.67
2.66
2.64
2.56
2.33
2.31
2.04


Ph
N
N-Ph
O
Ph
O
N
Me
Cl

5la


2.97
1.09
2.17
1.94
6.74
11.02
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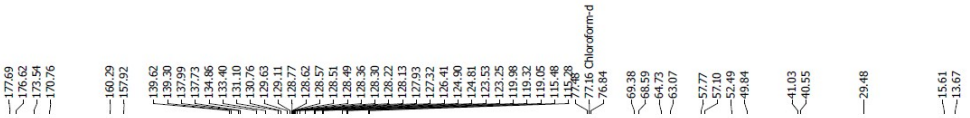
1.07
2.14
0.44
1.21
3.00
0.42
0.42
4.17
1.10
f1 (ppm)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173.54
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139.62
139.30
137.99
137.73
134.86
133.40
131.10
130.76
129.63
129.11
128.77
128.62
128.57
128.51
128.49
128.36
128.30
128.22
128.13
127.93
127.32
126.41
124.90
124.81
123.53
123.25
119.98
119.32
119.05
115.48
115.28
77.48
77.16 Chloroform-d
76.84
69.38
68.59
64.73
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57.77
57.10
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41.03
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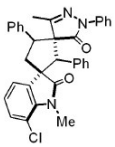
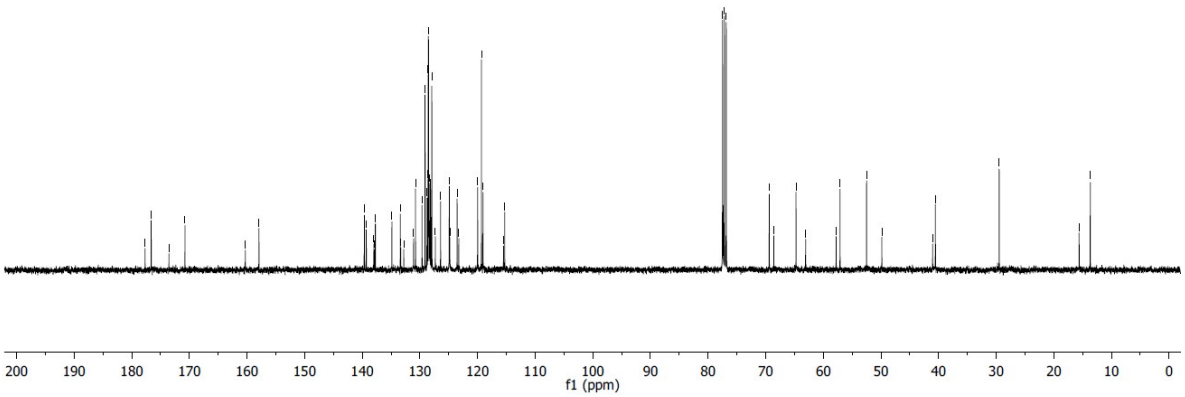
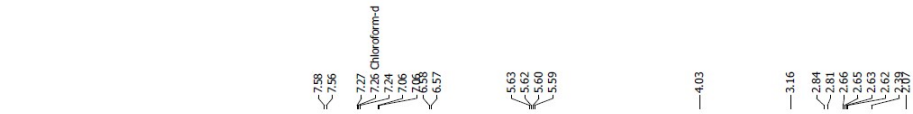
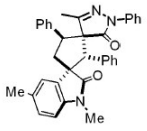
15.61
13.67


Ph
N
N-Ph
O
Ph
O
N
Me
Cl

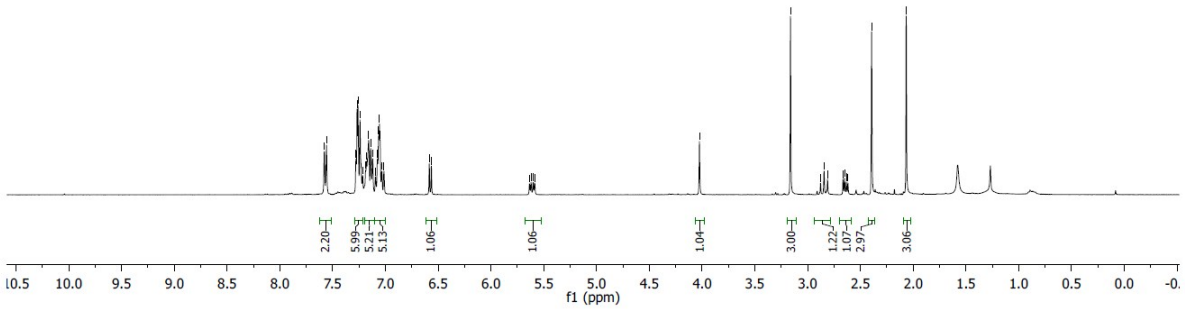
5la

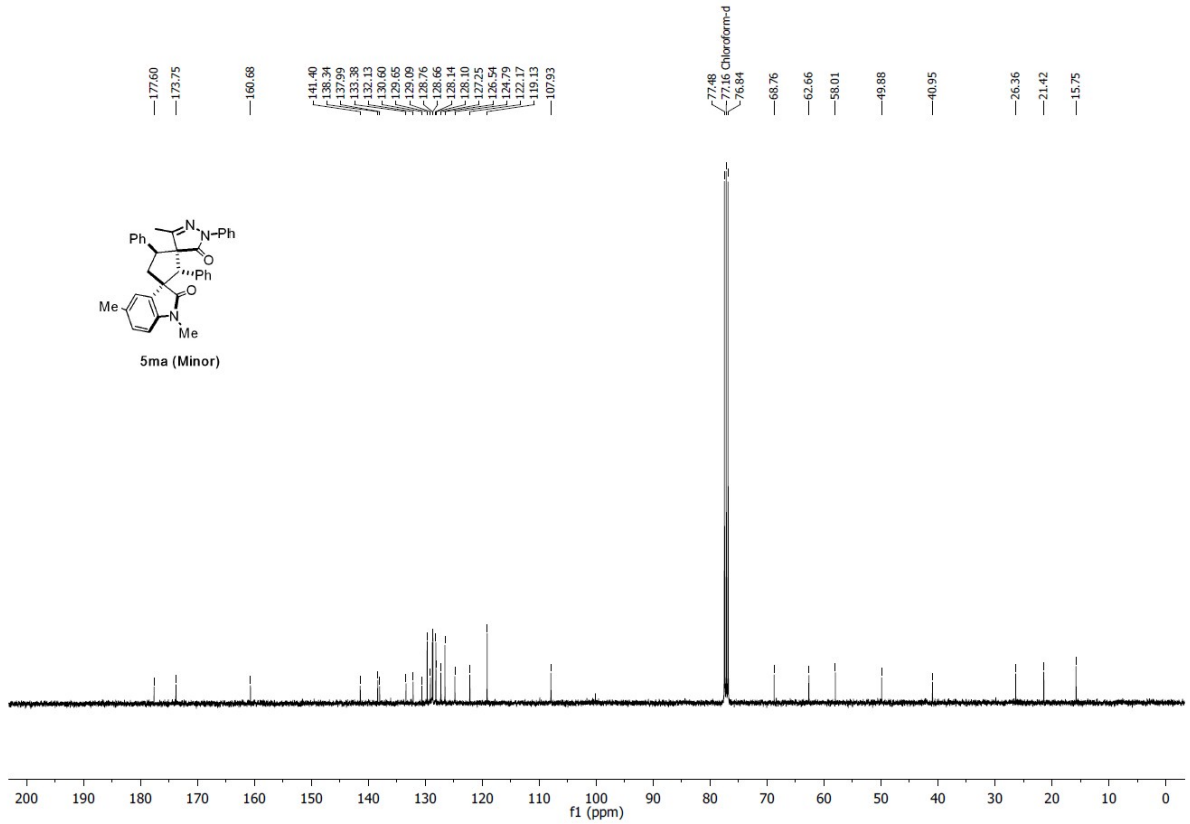

f1 (ppm)



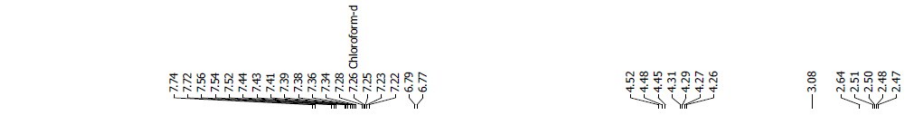


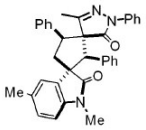
5ma (Minor)



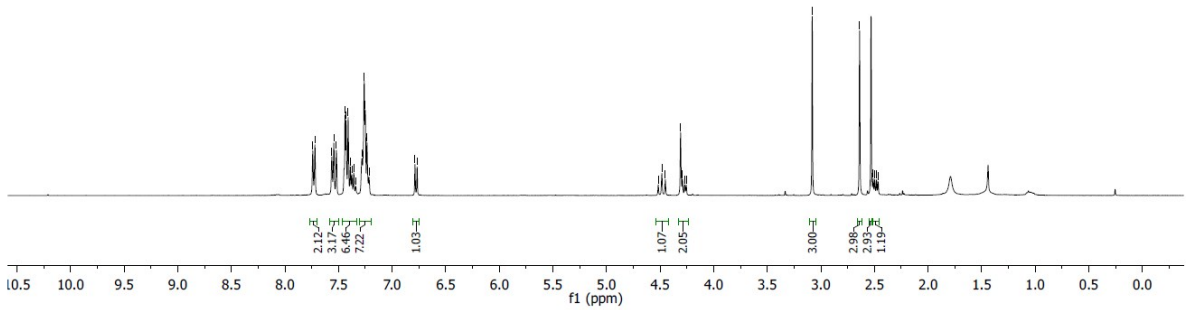


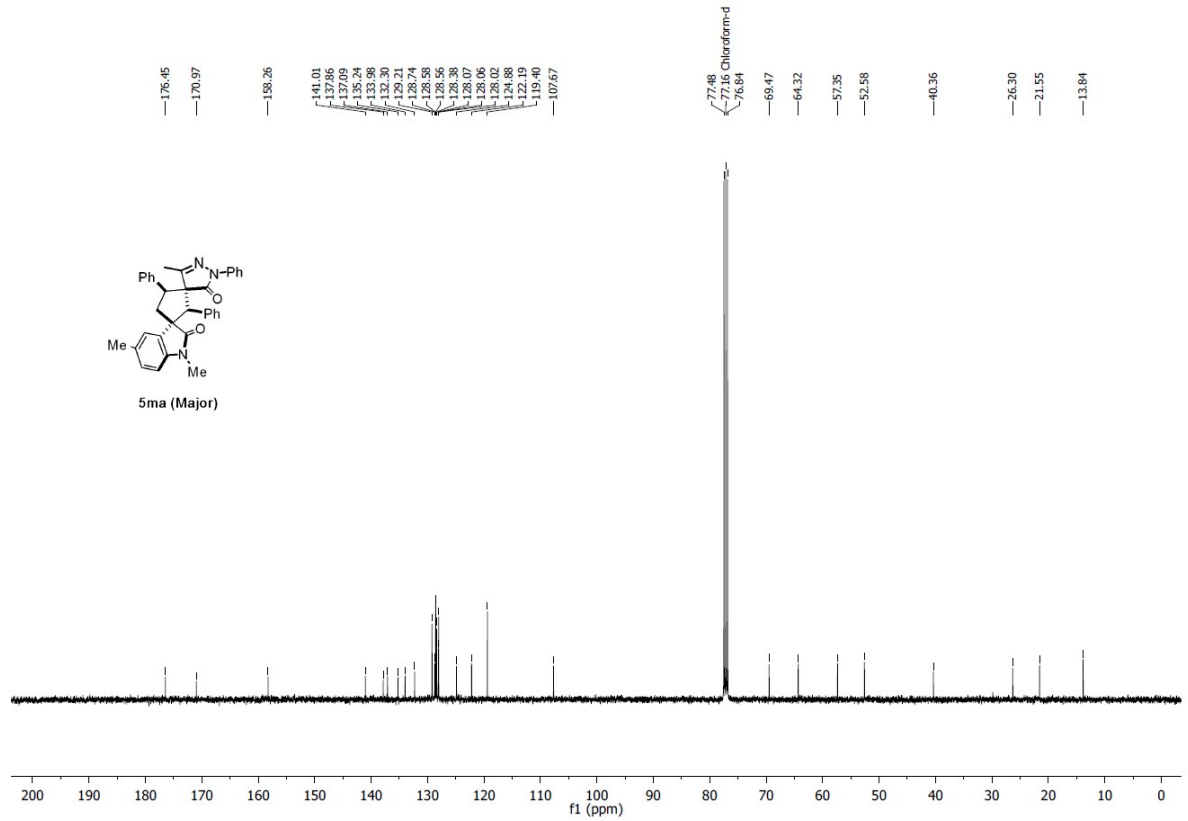
5ma (Minor)



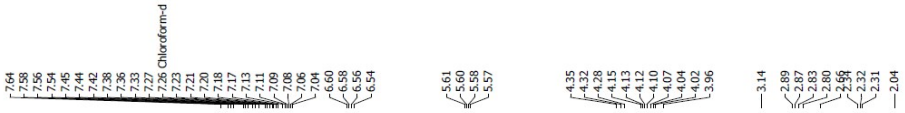


5ma (Major)





5ma (Major)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7.58
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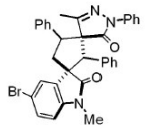
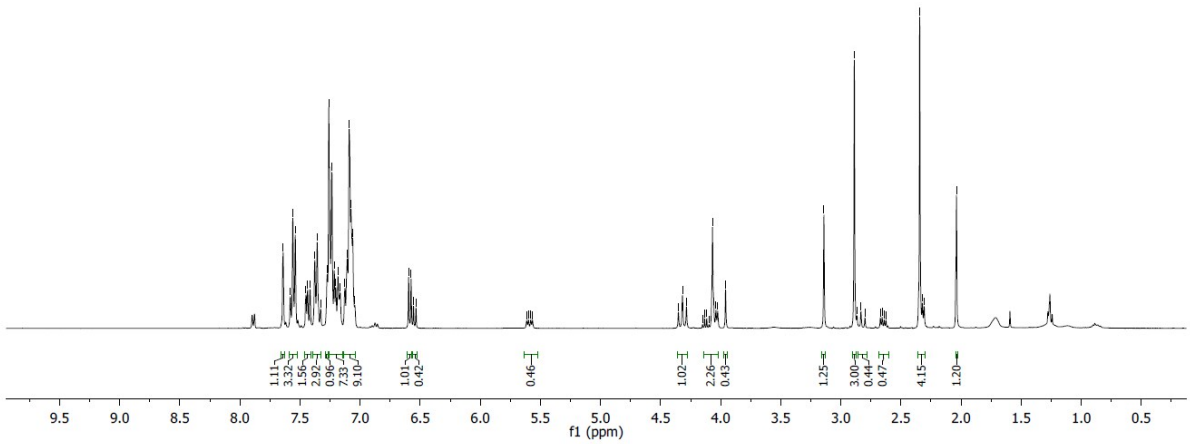
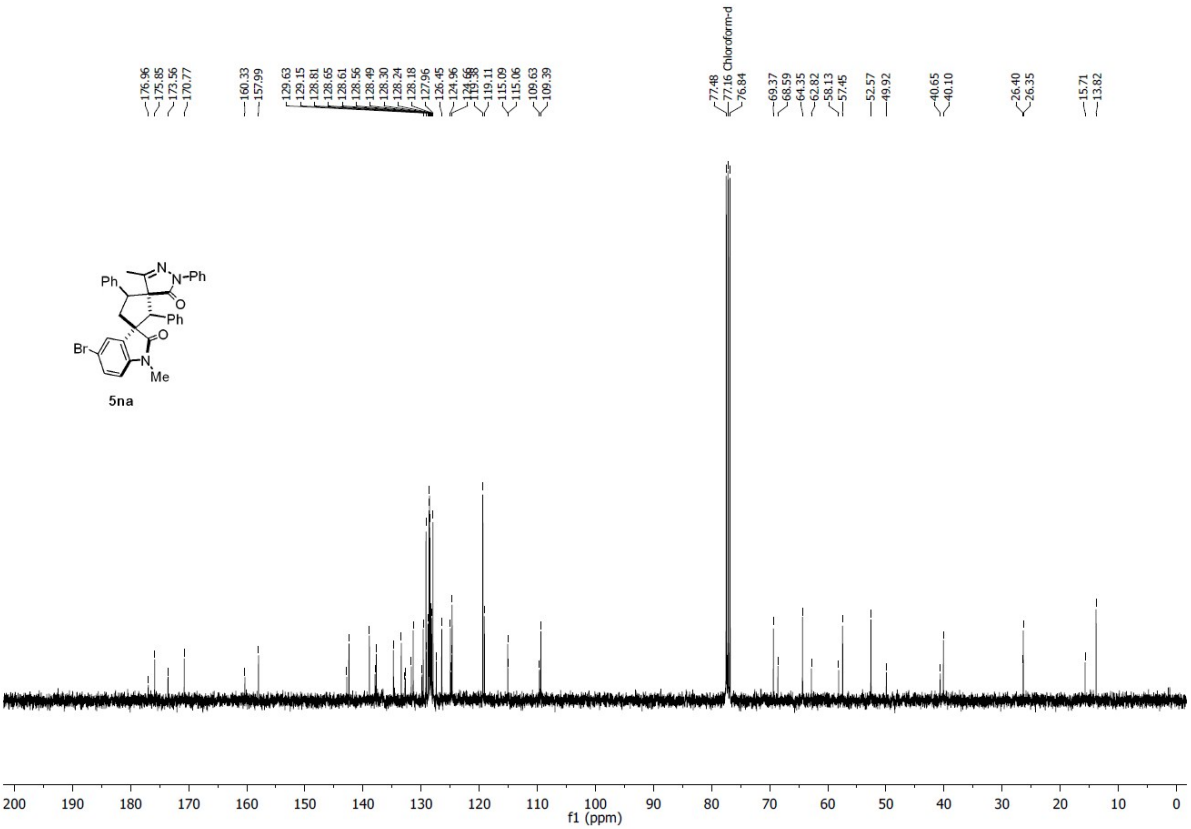
7.54
7.45
7.44
7.42
7.38
7.36
7.33
7.27
Chloroform-d
7.26
7.23
7.21
7.20
7.18
7.17
7.13
7.11
7.09
7.08
7.06
7.04
6.60
6.58
6.56
6.54
5.61
5.60
5.58
5.57
4.35
4.32
4.28
4.15
4.13
4.12
4.10
4.07
4.04
4.02
3.96
3.14
2.89
2.87
2.83
2.80
2.66
2.34
2.32
2.31
2.04


Ph
N
N-Ph
O
Ph
O
Br
N
Me

5na


1.11
3.32
1.56
2.92
0.96
7.33
9.10
1.01
0.42
0.46
1.02
2.26
0.43
1.25
3.00
0.44
0.47
4.15
1.20
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
f1 (ppm)


176.96
175.85
173.56
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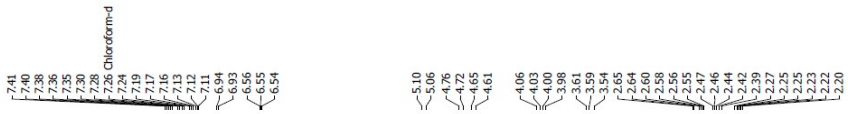
160.33
157.99
129.63
129.15
128.81
128.65
128.61
128.56
128.49
128.30
128.24
128.18
127.96
126.45
124.96
119.11
115.09
115.06
109.63
109.39
77.48
77.16
Chloroform-d
76.84
69.37
68.59
64.35
62.82
58.13
57.45
52.57
49.92
40.65
40.10
26.40
26.35
15.71
13.82
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

Ph
N
N-Ph
O
Ph
O
Br
N
Me

5na


7.41
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7.28
Chloroform-d
7.26
7.24
7.19
7.17
7.16
7.13
7.12
7.11
6.94
6.93
6.56
6.55
6.54
5.10
5.06
4.76
4.72
4.65
4.61
4.06
4.03
4.00
3.98
3.61
3.59
3.54
2.65
2.64
2.60
2.58
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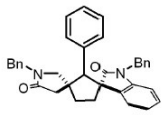
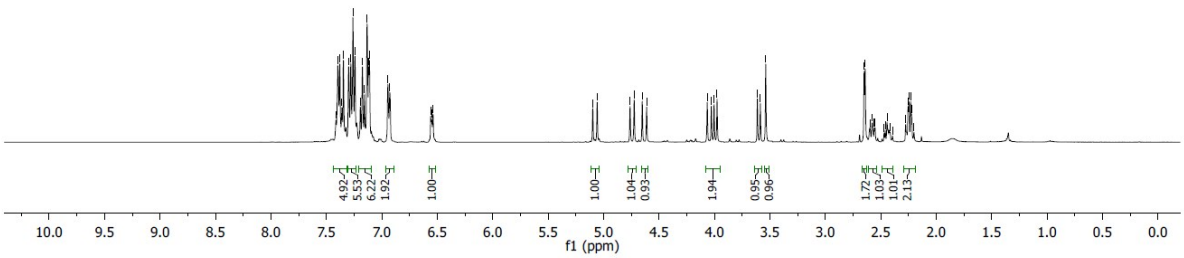
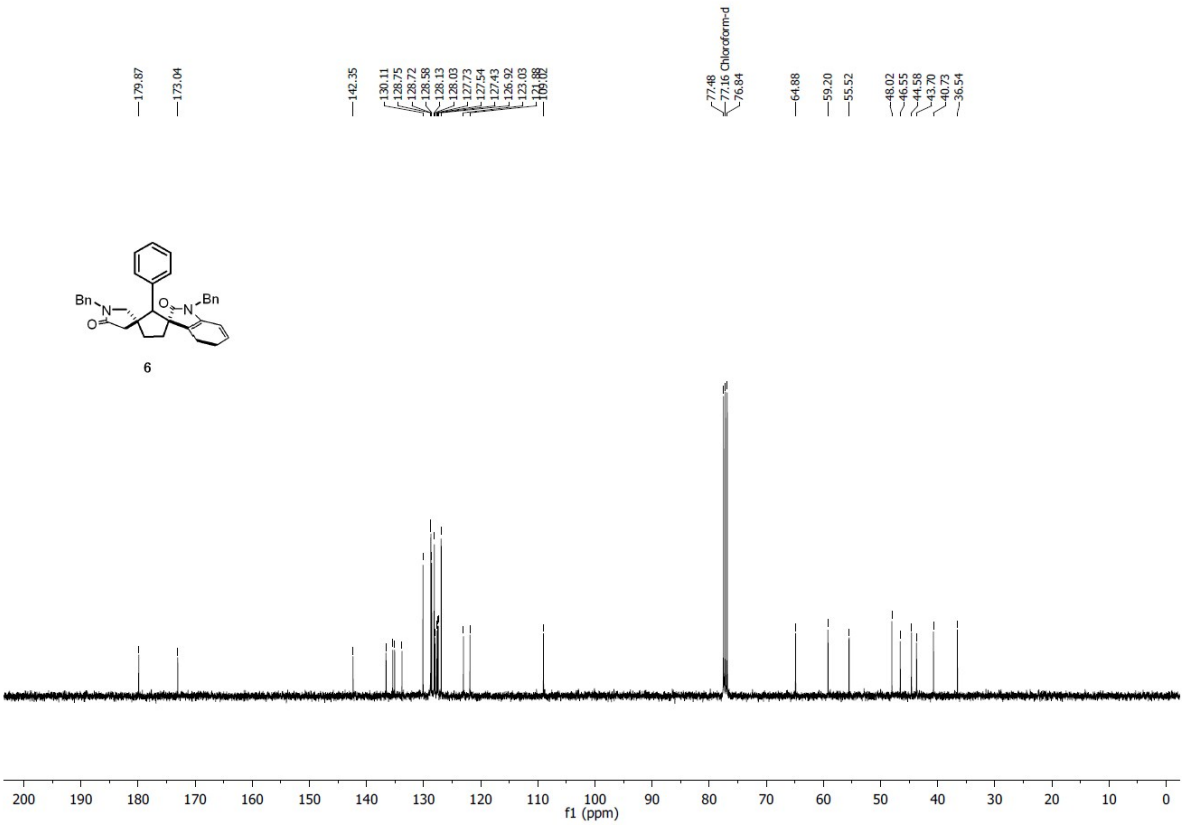
2.39
2.27
2.25
2.23
2.22
2.20


Bn
N
O
O
N
Bn

6


4.92
5.53
6.22
1.92
1.00
1.00
1.04
0.93
1.94
0.95
0.96
1.72
1.03
1.01
2.13
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)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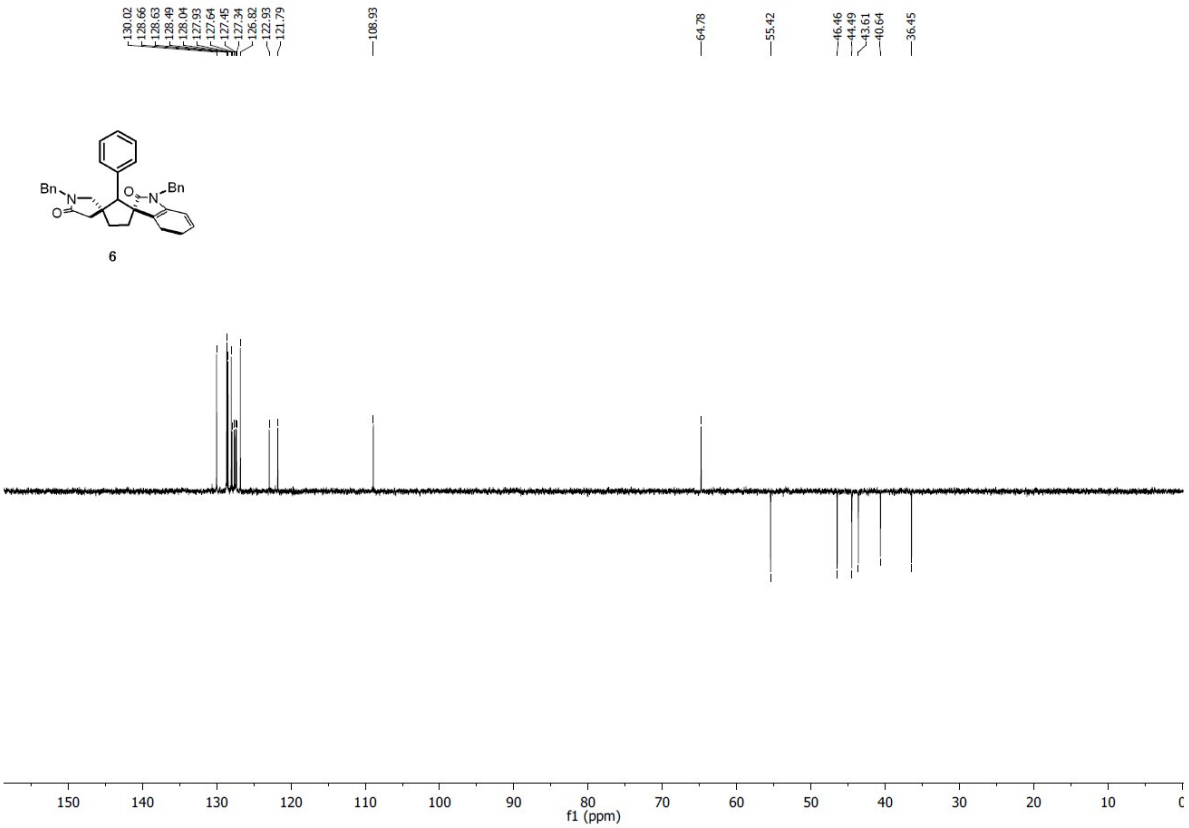
128.72
128.58
128.13
128.03
127.73
127.54
127.43
126.92
123.03
121.88
109.02
77.48
77.16
Chloroform-d
76.84
64.88
59.20
55.52
48.02
46.55
44.58
43.70
40.73
36.54
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

Bn
N
O
O
N
Bn

6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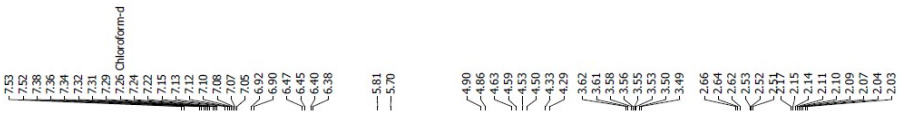
128.63
128.49
128.04
127.93
127.64
127.45
127.34
126.82
122.93
121.79
108.93
64.78
55.42
46.46
44.49
43.61
40.64
36.45
Bn
N
O
O
N
Bn
6
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
f1 (ppm)


7.53
7.52
7.38
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7.32
7.31
7.29
7.26 Chloroform-d
7.24
7.22
7.15
7.13
7.12
7.10
7.08
7.07
7.05
6.92
6.90
6.47
6.45
6.40
6.38
5.81
5.70
4.90
4.86
4.63
4.59
4.53
4.50
4.33
4.29
3.62
3.61
3.58
3.56
3.55
3.53
3.50
3.49
2.66
2.64
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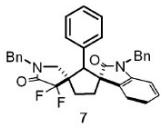
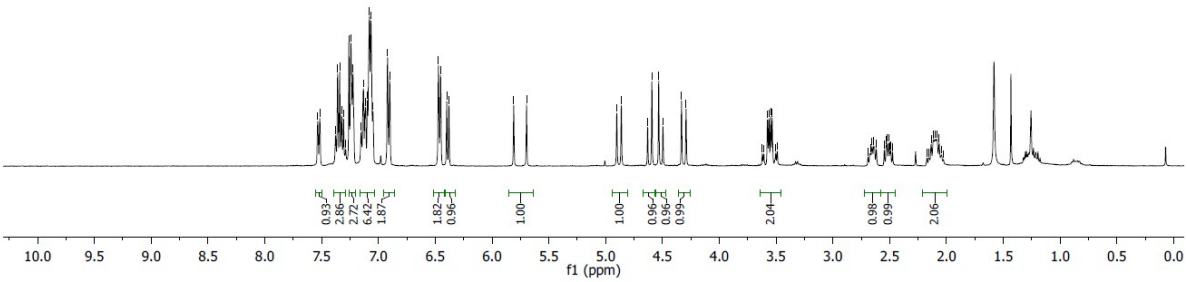
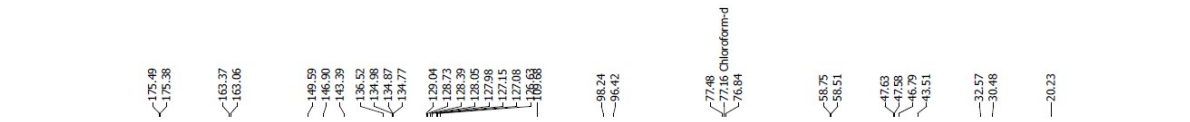
2.09
2.07
2.04
2.03


Bn
N
O
F
F
O
N
Bn

7


0.93
2.86
2.72
6.42
1.87
1.82
0.96
1.00
1.00
0.96
0.96
0.99
2.04
0.98
0.99
2.06
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
f1 (ppm)


175.49
175.38
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163.06
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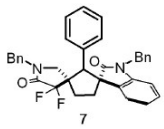
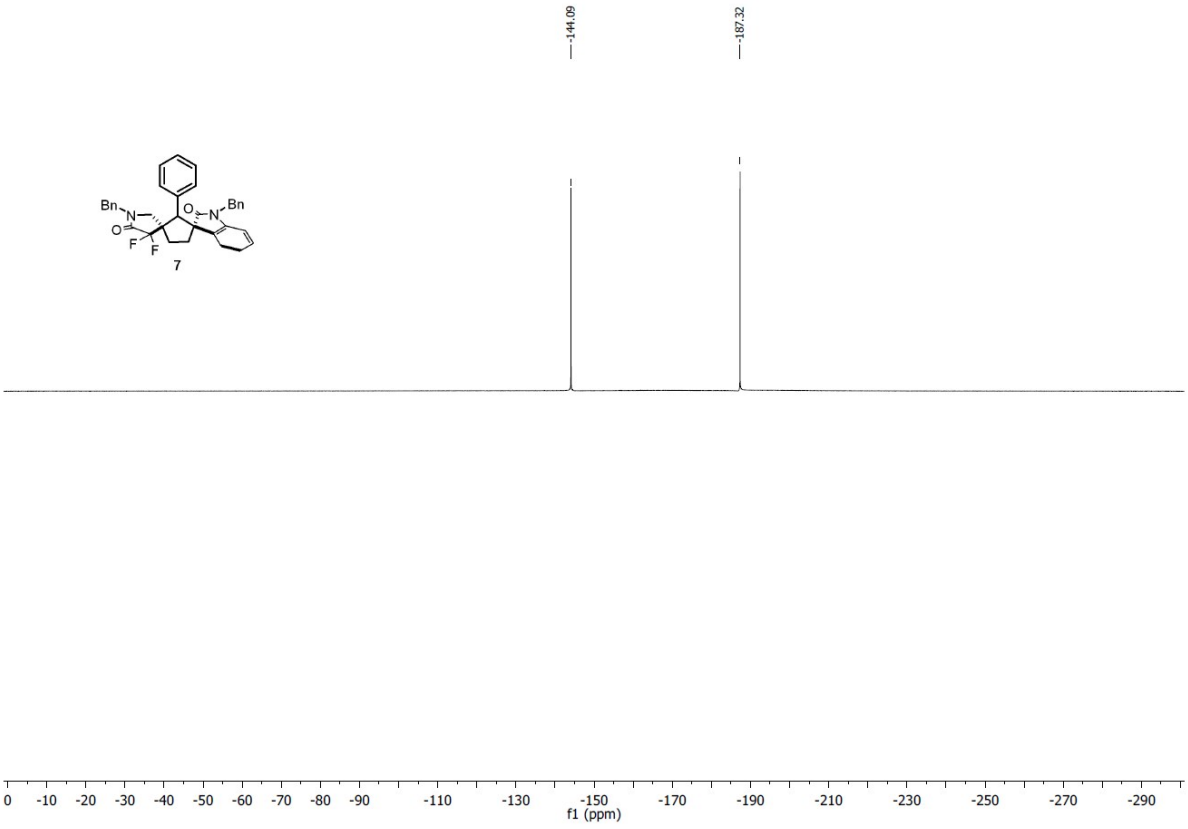
146.90
143.39
136.52
134.98
134.87
134.77
129.04
128.73
128.39
128.05
127.98
127.15
127.08
98.24
96.42
77.48
77.16 Chloroform-d
76.84
58.75
58.51
47.63
47.58
46.79
43.51
32.57
30.48
20.23

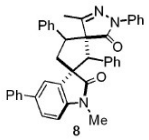

Bn
N
O
F
F
O
N
Bn

7

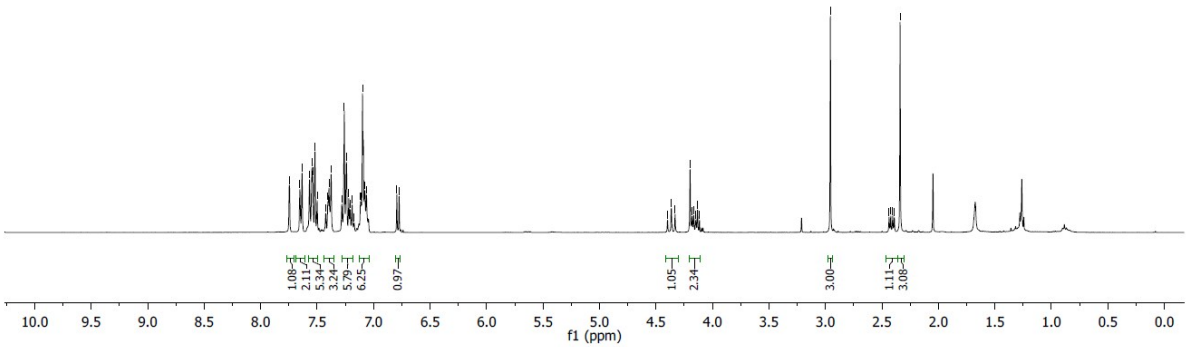
200
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

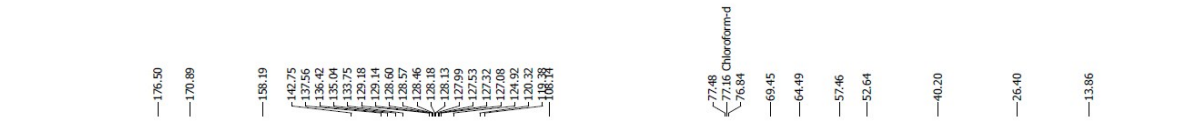

-144.09
-187.32
Bn
N
O
F
F
O
N
Bn
7
0
-10
-20
-30
-40
-50
-60
-70
-80
-90
-110
-130
-150
-170
-190
-210
-230
-250
-270
-290
f1 (ppm)

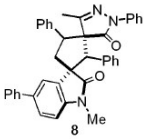

Chloroform-d


Ph
N
N-Ph
O
Ph
O
Ph
N
Me

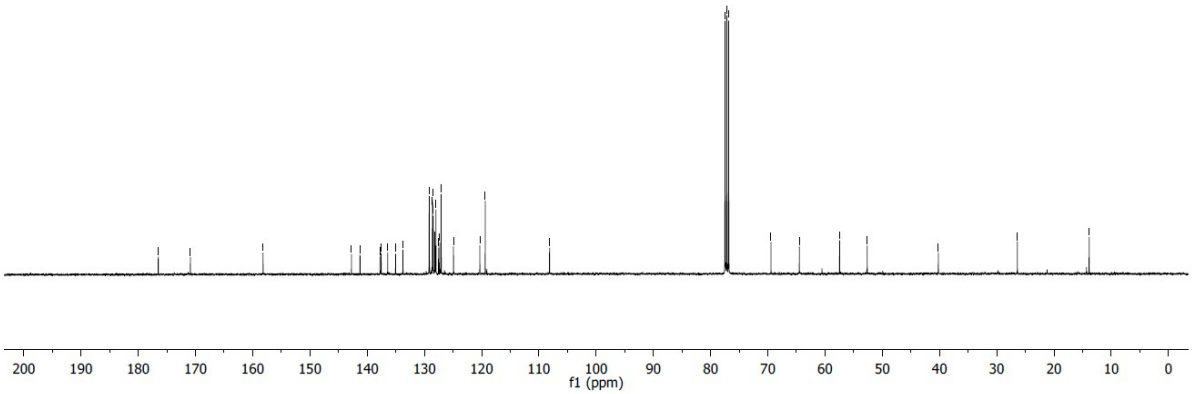
8


1.08
2.11
5.34
3.24
5.79
6.25
0.97
1.05
2.34
3.00
1.11
3.08
f1 (ppm)


Chloroform-d


Ph
N
N-Ph
O
Ph
O
Ph
N
Me

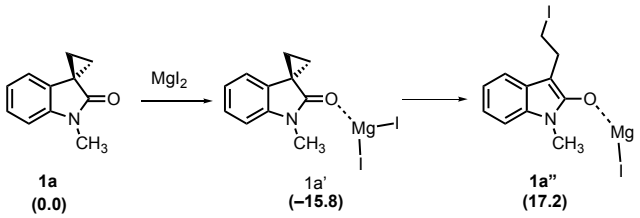
8


f1 (ppm)

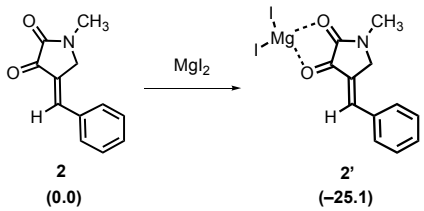
10. Computational Section

Computational Details

All optimizations of stationary points and vibrational analyses were carried out at OLYP[1]/TZ2P[2] using ADF.[3] The zeroth-order regular approximation (ZORA)[4] accounted for scalar relativistic effects. Solvation effects of tetrahydrofuran (THF) were included in all computations using the COnductor-like Screening MOdel (COSMO).[5] This level is referred to as COSMO(THF)-ZORA-OLYP/TZ2P. Energy minima and transition states were verified through vibrational analysis.[6] All minima were found to have zero imaginary frequencies, while all transition states had a single imaginary frequency.



Scheme S3. Ring opening of the representative spirocyclopropyl oxindole **1a** by MgI_2 to ring-opened intermediate **1a"**. Energies (in kcal mol^{-1}) computed at COSMO(THF)-ZORA-OLYP/TZ2P.



Scheme S4. Lewis acid activation of a representative 2,3-dioxopyrrolidine substrate **2** via coordination of MgI_2 is shown. Energies (in kcal mol^{-1}) computed at COSMO(THF)-ZORA-OLYP/TZ2P.

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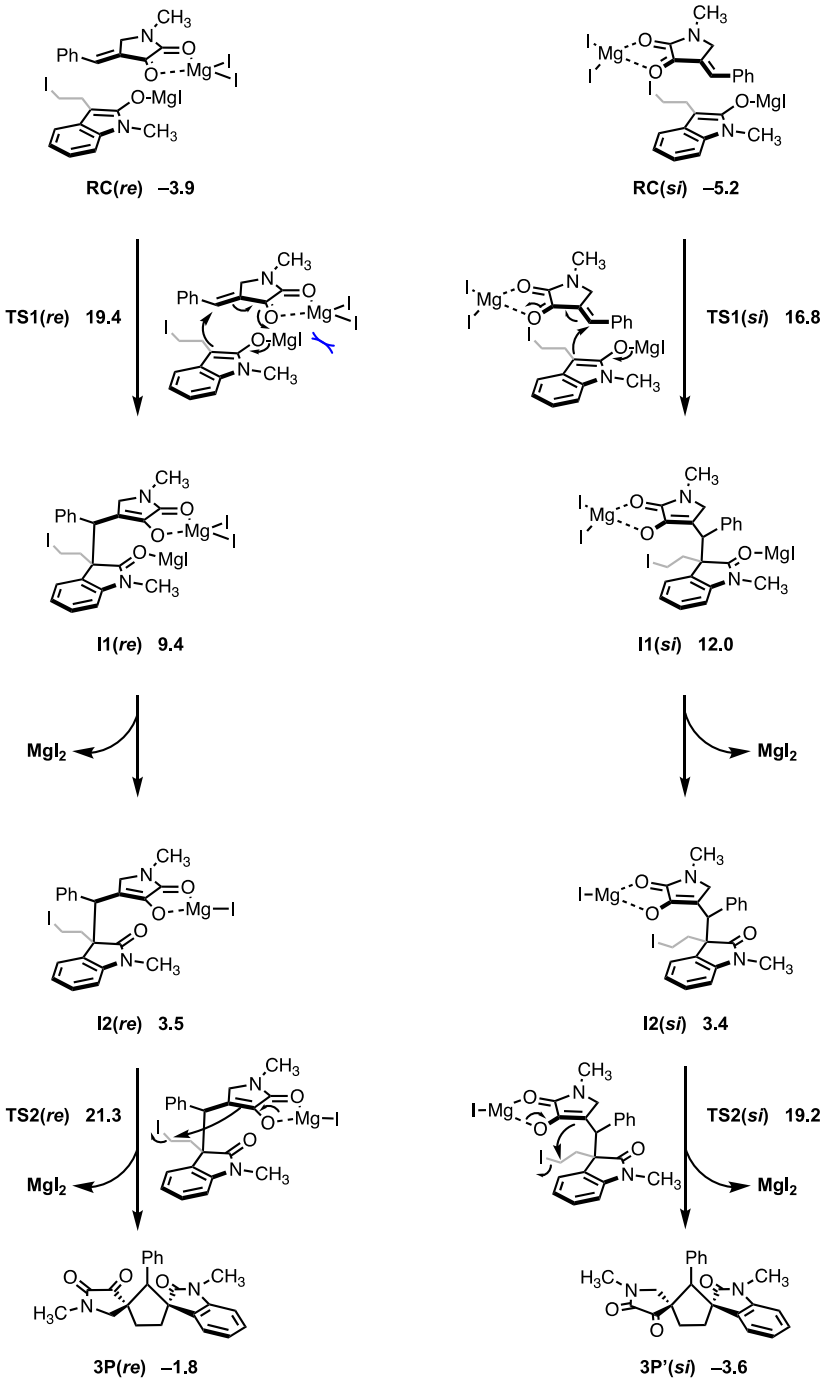


Scheme S5. Reaction pathways for a model [3+2] annulation reaction between DAC-**1a** with a representative 2,3-dioxopyrrolidine substrate **2** (see Scheme S1 and S2), with two different facial selectivity yielding diastereomeric products **3P** and **3P**' are shown. Energies (in kcal mol^{-1}) computed at COSMO(THF)-ZORA-OLYP/TZ2P. Where, RC = reactant complex, I = intermediate, TS = transition state, and 3P/3P' = products.

Table S1. Cartesian coordinates (Å), energies (kcal mol^{-1}), and imaginary vibrational frequencies of the optimized structures computed at COSMO(THF)-ZORA-OLYP/TZ2P.

MgI2
E = -142.80
N_{imag} = 0

Mg	0.000000	0.000000	0.000000
I	0.000000	0.000000	2.623989
I	0.000000	0.000000	-2.623989

1a
E = -3431.52
N_{imag} = 0

C	-1.847126	1.298057	-0.010139
C	-3.066973	0.640677	-0.091579
C	-3.091250	-0.760134	-0.045971
C	-1.904839	-1.484689	0.078985
C	-0.666210	-0.836917	0.162292
C	-0.658048	0.554092	0.115872
H	-3.996272	1.195939	-0.189047
H	-4.041286	-1.283884	-0.108770
H	-1.937172	-2.570731	0.113056
H	0.249913	-1.410170	0.259581
N	0.437979	1.423992	0.179995
C	-1.467251	2.725631	-0.025659
C	0.031878	2.743342	0.101742
O	0.776812	3.722384	0.132738
C	1.819718	1.008697	0.309815
H	2.441328	1.902986	0.328751
H	2.116744	0.382602	-0.537702
H	1.969749	0.447167	1.237600
C	-2.151486	3.799507	-0.865032
C	-2.277858	3.848474	0.613552
H	-3.001120	3.470819	-1.454122
H	-1.490662	4.512188	-1.347730
H	-1.704570	4.594906	1.153781
H	-3.216735	3.554332	1.070583

1a'
E = -3590.08
N_{imag} = 0
ATOMS

C	-1.872552	1.051463	-0.024306
C	-3.046116	0.311631	-0.092959
C	-2.971859	-1.081789	0.019633
C	-1.742072	-1.719283	0.198496
C	-0.551897	-0.987115	0.270624
C	-0.645217	0.394502	0.156101
H	-4.008638	0.796112	-0.231833
H	-3.882195	-1.672670	-0.032261
H	-1.703527	-2.801825	0.284321
H	0.399496	-1.489139	0.409912

N	0.393208	1.348964	0.196465
C	-1.583465	2.497316	-0.101838
C	-0.106380	2.601059	0.049810
O	0.578941	3.648002	0.044600
C	1.797465	1.019281	0.373188
H	2.379323	1.937310	0.369364
H	2.136215	0.374409	-0.442159
H	1.943698	0.503547	1.326153
C	-2.327298	3.485663	-1.003421
C	-2.486194	3.596433	0.463407
H	-3.132316	3.057690	-1.590641
H	-1.712414	4.217945	-1.514626
H	-1.983309	4.406746	0.979276
H	-3.404819	3.247426	0.922130
Mg	1.654346	5.299248	0.073330
I	4.314894	4.997606	0.353328
I	0.314028	7.616254	-0.173904

1a''

E = -3557.09

N_{imag} = 0

C	-1.858239	1.254667	0.531548
C	-3.139382	1.043008	1.069002
C	-3.675100	-0.245011	1.081316
C	-2.956809	-1.335155	0.563419
C	-1.680300	-1.157797	0.021381
C	-1.146840	0.130300	0.011177
H	-3.715523	1.871931	1.473326
H	-4.666753	-0.407959	1.497627
H	-3.395254	-2.329800	0.583071
H	-1.127854	-2.004749	-0.375512
N	0.074791	0.579954	-0.452244
C	-1.018920	2.404645	0.373376
C	0.157231	1.958490	-0.235684
O	1.220073	2.622032	-0.606338
C	1.102659	-0.243097	-1.052590
H	1.928570	0.396511	-1.360133
H	0.717242	-0.764185	-1.935433
H	1.478003	-0.987361	-0.341041
C	-2.031357	4.530651	-0.403983
C	-1.326532	3.821993	0.749185
H	-2.991502	4.082475	-0.654326
H	-1.411258	4.610051	-1.295468
H	-0.408435	4.363113	1.002776
H	-1.959173	3.851230	1.642347
I	-2.550838	6.642501	0.055526
Mg	2.686859	3.682599	-0.144051
I	4.731575	5.239828	0.299260

2

E = -3781.25

N_{imag} = 0

N	2.759214	0.668791	-1.062653
C	2.758244	-0.709972	-1.542164
C	4.123571	-1.232340	-1.195677

C	4.862392	-0.144975	-0.542545
C	3.916910	1.065976	-0.483673
H	1.955554	-1.273454	-1.048550
H	2.565771	-0.725048	-2.623174
C	4.688213	-2.452226	-1.388508
H	1.340608	1.615577	-2.283631
C	1.589833	1.506008	-1.222070
O	4.178934	2.169194	-0.002329
O	6.006452	-0.132601	-0.105393
H	1.800421	2.486980	-0.797211
H	0.730619	1.063038	-0.705852
H	1.550540	-5.170964	-3.556261
H	5.710179	-2.522242	-1.016290
H	3.123460	-7.094924	-3.558917
H	5.389248	-6.838143	-2.558805
H	6.064744	-4.680108	-1.571691
C	4.196522	-3.677948	-1.991877
C	2.915865	-3.846362	-2.564361
C	2.540399	-5.063596	-3.120447
C	3.424446	-6.146051	-3.122468
C	4.694828	-6.002185	-2.561668
C	5.074122	-4.786136	-2.005903
H	2.205035	-3.031059	-2.581638

2'

E = -3949.11

N_{imag} = 0

N	2.549479	0.502995	-1.164782
C	2.574941	-0.883716	-1.646515
C	3.955935	-1.385224	-1.304622
C	4.641132	-0.299689	-0.662723
C	3.696912	0.869914	-0.602635
H	1.784450	-1.452279	-1.144022
H	2.378354	-0.892395	-2.724494
C	4.553675	-2.599104	-1.508629
H	1.121504	1.439761	-2.373804
C	1.370327	1.335798	-1.313393
O	4.028543	1.963061	-0.089192
O	5.789155	-0.182437	-0.190309
H	1.573533	2.316964	-0.887299
H	0.524591	0.876586	-0.792743
I	7.758788	3.234683	-0.943353
H	5.577089	-2.647071	-1.139583
Mg	6.025317	1.808858	0.607019
I	6.133960	1.717733	3.331692
H	5.997745	-4.770148	-1.732273
C	4.091689	-3.821032	-2.117099
C	2.805675	-4.017011	-2.674325
C	2.461048	-5.235508	-3.240700
C	3.380717	-6.289377	-3.268246
C	4.655251	-6.119019	-2.723303
C	5.005968	-4.902162	-2.155584
H	2.070693	-3.223433	-2.669268
H	1.469912	-5.369894	-3.664779
H	3.101981	-7.240504	-3.714085

H	5.372020	-6.935035	-2.742612

RC(*si*)
E = -7503.61
N_{imag} = 0

C	-3.963564	2.017458	-3.316078
C	-5.320534	1.807616	-3.572121
C	-5.810654	0.500043	-3.609027
C	-4.959746	-0.592306	-3.405501
C	-3.592847	-0.407221	-3.177261
C	-3.122063	0.900025	-3.145212
H	-5.992907	2.642411	-3.746500
H	-6.866104	0.328622	-3.803824
H	-5.360914	-1.601774	-3.438336
H	-2.934268	-1.259083	-3.042924
N	-1.807615	1.360566	-3.000035
C	-3.144696	3.227485	-3.236715
C	-1.785573	2.722554	-3.117262
O	-0.698861	3.394036	-3.056096
C	-0.661950	0.500267	-2.774161
H	0.225422	1.111480	-2.625105
H	-0.500937	-0.153971	-3.636352
H	-0.819536	-0.112847	-1.882970
C	-2.488085	5.430082	-4.469710
C	-3.551034	4.399577	-4.126738
H	-1.616615	4.996152	-4.952070
H	-2.196236	6.064575	-3.636251
H	-4.415937	4.898688	-3.679107
H	-3.912910	3.961655	-5.063562
I	-3.217869	6.846767	-5.983515
Mg	1.016905	4.154589	-3.098545
I	3.024092	5.535326	-2.211155
N	-6.793222	5.119289	-0.247010
C	-5.853346	4.010257	-0.396287
C	-4.608387	4.632360	-0.986215
C	-4.861841	6.001184	-1.145090
C	-6.245109	6.270344	-0.634917
H	-5.671411	3.564412	0.588657
H	-6.300539	3.239199	-1.033162
C	-3.329411	4.032730	-1.149406
H	-8.690699	4.228909	-0.269007
C	-8.114735	4.946035	0.324124
O	-6.730541	7.429855	-0.589258
O	-4.160403	6.991896	-1.540099
H	-8.627448	5.907216	0.325105
H	-8.038028	4.575636	1.351709
I	-6.089574	10.288276	-3.305601
H	-2.570621	4.773483	-1.383160
Mg	-5.209717	8.760975	-1.198472
I	-4.067850	10.073705	0.948031
H	-1.020546	4.109677	0.155870
C	-2.825675	2.986453	-0.231122
C	-3.532126	1.822316	0.121240
C	-3.015179	0.924513	1.051994
C	-1.783541	1.165072	1.661721

C	-1.067713	2.315061	1.326683
C	-1.579159	3.207172	0.388616
H	-4.483212	1.596443	-0.344164
H	-3.580315	0.030079	1.302137
H	-1.387035	0.464875	2.392570
H	-0.110549	2.521657	1.799038

RC(*re*)
E = -7502.29
N_{imag} = 0

C	-3.656849	1.340339	-3.855813
C	-4.888968	0.839522	-4.308791
C	-5.261678	-0.465182	-3.984346
C	-4.425439	-1.288743	-3.213714
C	-3.194333	-0.819854	-2.746765
C	-2.825256	0.484327	-3.070162
H	-5.551615	1.455568	-4.912269
H	-6.214936	-0.852611	-4.337645
H	-4.736105	-2.303201	-2.976148
H	-2.552521	-1.459337	-2.147460
N	-1.684468	1.190177	-2.737887
C	-2.980074	2.596515	-3.984955
C	-1.770079	2.470763	-3.293567
O	-0.817125	3.339036	-3.094242
C	-0.589669	0.690453	-1.935142
H	0.153009	1.479075	-1.824541
H	-0.115707	-0.175286	-2.411104
H	-0.937111	0.395266	-0.939007
C	-3.092524	3.742026	-6.193876
C	-3.449731	3.821736	-4.711429
H	-3.566120	2.901682	-6.698620
H	-2.018915	3.722287	-6.376301
H	-2.999629	4.716701	-4.269643
H	-4.534519	3.935190	-4.609812
I	-3.785540	5.502730	-7.356482
Mg	0.491448	4.553514	-3.617952
I	1.902120	6.475747	-4.652305
N	-6.276334	5.559257	-1.239963
C	-5.837564	4.332108	-0.561482
C	-4.742899	4.779746	0.376133
C	-4.618400	6.200020	0.216175
C	-5.616903	6.636992	-0.821676
H	-6.689809	3.891234	-0.032759
H	-5.484470	3.612800	-1.309547
C	-3.956494	4.079209	1.255584
H	-7.082066	4.886562	-3.053622
C	-7.347204	5.544687	-2.220901
O	-5.729699	7.834223	-1.167705
O	-3.881778	7.053144	0.749339
H	-7.502434	6.556965	-2.590338
H	-8.267696	5.176196	-1.758620
I	-2.257175	9.815022	-1.634136
H	-3.246034	4.706367	1.792014
Mg	-4.337223	9.003538	-0.068255
I	-5.555980	10.544385	1.818827

H	-2.266231	3.077340	2.984507
C	-3.886184	2.682843	1.605014
C	-4.715037	1.667912	1.070240
C	-4.568615	0.350943	1.478445
C	-3.596779	0.006406	2.423676
C	-2.766632	0.990936	2.964508
C	-2.909892	2.310971	2.562008
H	-5.477565	1.899399	0.338694
H	-5.213080	-0.416220	1.058953
H	-3.488788	-1.028619	2.737029
H	-2.011396	0.726742	3.699230

TS1(*si*)
E = -7481.61
N_{imag} = 1, v = –255.1i

C	-3.957832	2.088977	-3.251867
C	-5.255245	1.854687	-3.705754
C	-5.715387	0.537313	-3.789877
C	-4.895327	-0.534369	-3.424085
C	-3.588974	-0.320404	-2.972926
C	-3.149223	0.995065	-2.900195
H	-5.906621	2.675202	-3.992788
H	-6.724184	0.344936	-4.145063
H	-5.272021	-1.551099	-3.495806
H	-2.953442	-1.156982	-2.702056
N	-1.886415	1.485679	-2.526185
C	-3.190051	3.329149	-3.038614
C	-1.856707	2.837324	-2.674510
O	-0.799661	3.524362	-2.458338
C	-0.785670	0.653039	-2.076823
H	-0.011191	1.280694	-1.641049
H	-0.365253	0.084561	-2.912056
H	-1.137391	-0.042370	-1.311951
C	-2.196317	5.296571	-4.436107
C	-3.408330	4.464648	-4.049433
H	-1.408033	4.711218	-4.904372
H	-1.797392	5.902221	-3.626876
H	-4.191638	5.129489	-3.683280
H	-3.801920	4.003688	-4.959705
I	-2.698117	6.778906	-5.979782
Mg	0.961100	4.172168	-2.667798
I	2.616163	6.160697	-2.586615
N	-3.451914	7.695711	-0.564906
C	-2.912907	6.339558	-0.470652
C	-3.956367	5.460564	-1.117396
C	-5.012342	6.284783	-1.505139
C	-4.665284	7.689114	-1.119698
H	-1.936250	6.304112	-0.968813
H	-2.751603	6.096362	0.586331
C	-4.004429	4.028729	-1.207654
H	-2.609354	8.746982	1.045046
C	-2.754887	8.851906	-0.035017
O	-5.449084	8.653454	-1.313241
O	-6.155346	6.068137	-2.041322
H	-3.350469	9.742398	-0.232893

H	-1.777497	8.958095	-0.516010
I	-9.224337	7.894309	-0.186545
H	-4.986600	3.724224	-1.555742
Mg	-7.234485	7.836799	-2.103076
I	-7.835225	8.761571	-4.620864
H	-5.365880	1.993295	-0.179535
C	-3.545045	3.150545	-0.098802
C	-2.330679	3.302439	0.595455
C	-2.010535	2.479934	1.672486
C	-2.894228	1.483181	2.089693
C	-4.101052	1.313540	1.411898
C	-4.417291	2.132460	0.330714
H	-1.614880	4.056955	0.294097
H	-1.065510	2.621122	2.191243
H	-2.644284	0.847496	2.935278
H	-4.801766	0.543648	1.725109

TS1(*re*)
E = -7478.94
N_{imag} = 1, v = –243.6i

C	-3.572949	1.945457	-3.168861
C	-4.822763	1.425738	-3.509505
C	-4.999614	0.040279	-3.535776
C	-3.940759	-0.823832	-3.237267
C	-2.673837	-0.326200	-2.919617
C	-2.517501	1.054160	-2.896541
H	-5.652495	2.080419	-3.758699
H	-5.970615	-0.371246	-3.798139
H	-4.096613	-1.898900	-3.265715
H	-1.850801	-1.002979	-2.715384
N	-1.355549	1.807668	-2.672929
C	-3.056699	3.317017	-3.083395
C	-1.625522	3.131274	-2.862181
O	-0.726999	4.036113	-2.769332
C	-0.061241	1.249039	-2.327366
H	0.530131	1.993940	-1.796646
H	0.479215	0.922099	-3.221617
H	-0.197870	0.395859	-1.662186
C	-2.777229	5.518634	-4.460844
C	-3.638915	4.330100	-4.067734
H	-1.825200	5.230661	-4.898162
H	-2.636023	6.249418	-3.668923
H	-4.594606	4.691376	-3.673299
H	-3.881435	3.771598	-4.977387
I	-3.728606	6.661833	-6.081609
Mg	0.982213	4.823058	-2.949741
I	3.548316	5.088898	-2.691018
N	-7.078074	5.200638	-0.740161
C	-6.132911	4.085271	-0.765870
C	-4.802807	4.713166	-1.109228
C	-5.013777	6.086301	-1.253800
C	-6.459928	6.357966	-0.975619
H	-6.128809	3.602190	0.218334
H	-6.469166	3.342314	-1.498273
C	-3.505749	4.114071	-1.100383

H	-8.937007	4.302461	-1.094509
C	-8.480032	5.026487	-0.413240
O	-6.934653	7.523441	-0.961309
O	-4.243367	7.080864	-1.486548
H	-8.987222	5.985442	-0.513485
H	-8.590613	4.665226	0.614717
I	-5.641301	10.628846	-3.226333
H	-2.738717	4.876258	-1.206422
Mg	-5.298879	8.844261	-1.168926
I	-4.567629	9.888526	1.289340
H	-1.364769	4.216164	0.464829
C	-3.119439	3.088643	-0.099467
C	-3.866290	1.934247	0.195366
C	-3.470974	1.063864	1.208325
C	-2.321529	1.321037	1.956234
C	-1.564006	2.458742	1.675881
C	-1.955845	3.325166	0.658928
H	-4.752747	1.692830	-0.377254
H	-4.065750	0.177106	1.412957
H	-2.019670	0.641918	2.749507
H	-0.668211	2.676248	2.252248

I1(*si*)
E = -7489.01
N_{imag} = 0

C	-3.723430	1.683117	-2.945796
C	-4.996780	1.221712	-3.250537
C	-5.191903	-0.151640	-3.433801
C	-4.127602	-1.047306	-3.319544
C	-2.837312	-0.596385	-3.024711
C	-2.673769	0.770657	-2.846563
H	-5.837085	1.901857	-3.349426
H	-6.185155	-0.523033	-3.669652
H	-4.297322	-2.110296	-3.465730
H	-2.010665	-1.293852	-2.947108
N	-1.478318	1.476530	-2.562299
C	-3.193704	3.090797	-2.730110
C	-1.712819	2.795252	-2.487402
O	-0.812910	3.655346	-2.257699
C	-0.189378	0.826279	-2.383910
H	0.537658	1.549323	-2.022797
H	0.154221	0.405280	-3.332138
H	-0.284261	0.027810	-1.645415
C	-2.601834	5.130036	-4.352789
C	-3.399024	3.863257	-4.095084
H	-1.532757	4.957813	-4.437016
H	-2.811011	5.937466	-3.659255
H	-4.466621	4.086155	-4.152264
H	-3.183982	3.150937	-4.895168
I	-3.102677	5.971461	-6.321895
Mg	0.839423	4.612632	-2.285681
I	2.122678	6.854041	-2.316835
N	-3.251267	7.334169	-0.342206
C	-2.786407	5.953726	-0.450654
C	-3.849807	5.249825	-1.252775

C	-4.839400	6.159552	-1.534226
C	-4.434622	7.471771	-0.949061
H	-1.797212	5.951330	-0.926405
H	-2.660525	5.536323	0.556305
C	-3.989587	3.773829	-1.511260
H	-2.353414	8.079128	1.403411
C	-2.514372	8.366047	0.359022
O	-5.152879	8.504676	-1.032571
O	-5.981945	6.088040	-2.145139
H	-3.091211	9.290180	0.328250
H	-1.542110	8.537669	-0.115067
I	-9.072066	7.883856	-0.262209
H	-5.015097	3.691117	-1.880177
Mg	-6.920248	7.890942	-2.019189
I	-7.292509	9.262432	-4.393067
H	-6.103455	2.696143	-0.225226
C	-3.962105	2.961128	-0.211713
C	-2.798673	2.699724	0.526070
C	-2.847114	1.991731	1.727950
C	-4.065242	1.535014	2.228834
C	-5.234635	1.797469	1.514959
C	-5.179470	2.499962	0.312358
H	-1.833148	3.055646	0.184995
H	-1.927025	1.804597	2.276587
H	-4.103550	0.985947	3.166239
H	-6.194922	1.454423	1.892416

I1(*re*)
E = -7486.33
N_{imag} = 0

C	-4.059545	1.919169	-3.138752
C	-5.433610	1.767706	-3.280163
C	-5.953032	0.507455	-3.596002
C	-5.108219	-0.585062	-3.794811
C	-3.720800	-0.442964	-3.702309
C	-3.233295	0.817124	-3.383689
H	-6.106396	2.609148	-3.163380
H	-7.027589	0.382844	-3.695377
H	-5.527545	-1.556821	-4.039695
H	-3.063874	-1.286529	-3.882690
N	-1.879829	1.217923	-3.282185
C	-3.196526	3.142066	-2.889895
C	-1.799777	2.524554	-2.995883
O	-0.709590	3.139584	-2.831953
C	-0.752183	0.315635	-3.462120
H	0.173176	0.883667	-3.410309
H	-0.820873	-0.165944	-4.439778
H	-0.757461	-0.447588	-2.679658
C	-2.525616	5.298438	-4.297141
C	-3.460984	4.117001	-4.102791
H	-1.490835	5.015763	-4.468085
H	-2.608148	6.055205	-3.522137
H	-4.483042	4.479964	-3.974150
H	-3.455721	3.501449	-5.005735
I	-3.043023	6.380821	-6.139976

Mg	0.909945	3.941615	-2.240951
I	2.423256	5.638523	-1.019136
N	-6.665587	5.117537	-0.155767
C	-5.782230	3.984629	-0.418164
C	-4.599450	4.580407	-1.142011
C	-4.791280	5.937903	-1.221086
C	-6.108922	6.250265	-0.592999
H	-5.500777	3.523861	0.537769
H	-6.331812	3.226259	-0.987131
C	-3.303420	3.887740	-1.470773
H	-8.605396	4.323035	-0.030376
C	-7.936819	4.987711	0.526762
O	-6.554970	7.426785	-0.497171
O	-4.077741	6.926329	-1.668357
H	-8.396469	5.972970	0.602678
H	-7.792901	4.581474	1.533717
I	-6.035591	10.133480	-3.420943
H	-2.592922	4.710119	-1.586996
Mg	-5.048822	8.675730	-1.286381
I	-3.778940	10.144064	0.701429
H	-1.460879	4.662786	0.333161
C	-2.779754	3.069979	-0.285533
C	-3.244045	1.792278	0.057797
C	-2.743552	1.120371	1.174388
C	-1.773458	1.712288	1.981965
C	-1.312500	2.990338	1.665412
C	-1.813498	3.656437	0.547565
H	-4.007918	1.306098	-0.537233
H	-3.121415	0.129436	1.414630
H	-1.386170	1.187394	2.851441
H	-0.564596	3.473397	2.289357

I2(*si*)
E = -7352.17
N_{imag} = 0

C	-3.616519	1.644300	-2.869340
C	-4.824704	1.101500	-3.280235
C	-4.919852	-0.280448	-3.492531
C	-3.809231	-1.101913	-3.299431
C	-2.579680	-0.568760	-2.896681
C	-2.508182	0.806061	-2.691513
H	-5.697675	1.728316	-3.438822
H	-5.865066	-0.712455	-3.809846
H	-3.893259	-2.172817	-3.466174
H	-1.718524	-1.213252	-2.754803
N	-1.398103	1.567459	-2.306525
C	-3.203809	3.082431	-2.603092
C	-1.715582	2.904701	-2.204781
O	-0.924366	3.784283	-1.877024
C	-0.081452	1.025260	-2.033745
H	0.578761	1.850147	-1.770258
H	0.314363	0.515183	-2.917333
H	-0.121567	0.315278	-1.201315
C	-2.560317	5.162033	-4.127947
C	-3.300210	3.843283	-3.981530

H	-1.480304	5.066569	-4.069254
H	-2.907868	5.944439	-3.463132
H	-4.364222	3.992172	-4.182876
H	-2.933404	3.155968	-4.748566
I	-2.891538	6.020407	-6.133414
H	-4.735786	0.922031	3.147708
H	-6.687744	1.396142	1.670132
N	-3.414946	7.275736	-0.277122
C	-2.960369	5.890128	-0.387603
C	-4.034246	5.195027	-1.182056
C	-5.011335	6.111910	-1.458511
C	-4.595518	7.414445	-0.878519
H	-1.977374	5.859537	-0.868939
H	-2.842916	5.473707	0.620499
C	-4.144701	3.720478	-1.490155
H	-2.533267	8.043954	1.466944
C	-2.662454	8.305456	0.411488
O	-5.308250	8.464304	-0.970850
O	-6.162739	6.054222	-2.087177
H	-3.206042	9.247315	0.340926
H	-1.675194	8.426072	-0.045938
I	-9.195569	9.099991	-2.782026
H	-5.129681	3.639993	-1.957506
Mg	-6.983029	7.856019	-1.986030
H	-2.477882	1.730012	2.477827
H	-6.378950	2.641233	-0.426470
C	-4.250352	2.903306	-0.194680
C	-3.165873	2.635005	0.653149
C	-3.338676	1.925265	1.842337
C	-4.602607	1.473006	2.219951
C	-5.693739	1.739413	1.392810
C	-5.515069	2.444305	0.203285
H	-2.169709	2.980172	0.402298

I2(*re*)
E = -7352.05
N_{imag} = 0

C	-4.016870	1.647379	-3.001663
C	-5.310920	1.404038	-3.442786
C	-5.685447	0.108944	-3.824449
C	-4.759309	-0.932686	-3.786019
C	-3.437542	-0.701358	-3.392191
C	-3.088338	0.592988	-3.017293
H	-6.039528	2.203848	-3.512195
H	-6.701881	-0.079985	-4.158926
H	-5.057945	-1.935056	-4.082037
H	-2.712808	-1.508553	-3.392967
N	-1.823805	1.063775	-2.650663
C	-3.305802	2.926391	-2.597127
C	-1.846793	2.419131	-2.413525
O	-0.872934	3.093602	-2.097220
C	-0.630228	0.245544	-2.559069
H	0.194303	0.883093	-2.243563
H	-0.390149	-0.199092	-3.530109
H	-0.769465	-0.554048	-1.824895

C	-2.503203	5.144905	-3.784375
C	-3.391676	3.912830	-3.816317
H	-1.441428	4.919255	-3.801802
H	-2.735284	5.840609	-2.984115
H	-4.436891	4.219972	-3.900877
H	-3.163029	3.330704	-4.713244
I	-2.796320	6.359375	-5.603005
H	-4.619803	-0.107863	1.561396
H	-3.046988	0.684632	3.321195
N	-7.231359	5.051914	-0.785075
C	-6.385653	3.872279	-0.950741
C	-4.993106	4.413851	-1.156433
C	-5.066140	5.778252	-1.073256
C	-6.486781	6.153878	-0.851263
H	-6.465700	3.247041	-0.053112
H	-6.765748	3.281737	-1.790478
C	-3.704935	3.626734	-1.217230
H	-9.146364	4.508012	-1.454143
C	-8.665723	4.978380	-0.589919
O	-6.865387	7.361684	-0.731603
O	-4.177214	6.742783	-1.124382
H	-9.058058	5.987890	-0.469777
H	-8.898619	4.394047	0.305981
I	-4.608671	11.063814	-0.903463
H	-2.929553	4.389622	-1.120927
Mg	-5.138220	8.458151	-0.877115
H	-1.786731	2.806718	2.963898
H	-2.094284	4.094426	0.891321
C	-3.533844	2.742033	0.024873
C	-4.237960	1.549280	0.246134
C	-4.062089	0.815164	1.419958
C	-3.181914	1.257455	2.406954
C	-2.478013	2.445082	2.206202
C	-2.654469	3.173344	1.030376
H	-4.930533	1.173835	-0.497393

TS2(*si*)
E = -7336.38
N_{imag} = 1, v = –373.8i

C	-3.706579	1.574424	-2.882315
C	-4.932052	1.059680	-3.278432
C	-5.043826	-0.310358	-3.550616
C	-3.933739	-1.146020	-3.428085
C	-2.688618	-0.641313	-3.036065
C	-2.600746	0.721485	-2.769278
H	-5.803887	1.699524	-3.383773
H	-6.000549	-0.721557	-3.860501
H	-4.030603	-2.207083	-3.643258
H	-1.829319	-1.297696	-2.949492
N	-1.474833	1.456429	-2.373041
C	-3.280492	2.986217	-2.547931
C	-1.777565	2.788050	-2.205104
O	-0.972378	3.654668	-1.878569
C	-0.148891	0.897708	-2.188675
H	0.516206	1.697561	-1.866971

H	0.223806	0.475353	-3.127081
H	-0.166507	0.113263	-1.425900
C	-3.137845	5.356650	-3.505304
C	-3.395234	3.912304	-3.801945
H	-2.197621	5.676243	-3.087746
H	-3.860348	6.105687	-3.776455
H	-4.393215	3.795297	-4.229730
H	-2.686955	3.554221	-4.551492
I	-1.978771	6.290982	-5.913676
H	-4.696903	1.186317	3.374546
H	-6.684499	1.683829	1.954265
N	-3.348780	7.238410	-0.378008
C	-2.894428	5.847948	-0.484854
C	-3.939650	5.157549	-1.339038
C	-4.983322	6.076286	-1.493501
C	-4.545047	7.389293	-0.930769
H	-1.884206	5.816582	-0.896981
H	-2.851310	5.418339	0.522100
C	-4.166667	3.658739	-1.443456
H	-2.366982	7.978245	1.318053
C	-2.564250	8.266505	0.281273
O	-5.276063	8.421687	-0.987962
O	-6.151831	6.000144	-2.016756
H	-3.120412	9.202815	0.264091
H	-1.610643	8.399927	-0.238272
I	-9.411527	8.884058	-2.323332
H	-5.171459	3.585892	-1.868669
Mg	-7.034616	7.820054	-1.858038
H	-2.430886	1.826380	2.566459
H	-6.403574	2.790725	-0.222708
C	-4.255586	2.951001	-0.089710
C	-3.149009	2.667939	0.723942
C	-3.307343	2.036155	1.957978
C	-4.576089	1.677601	2.412350
C	-5.687290	1.956333	1.617248
C	-5.524660	2.584321	0.382879
H	-2.147603	2.938621	0.410886

TS2(*re*)
E = -7334.31
N_{imag} = 1, ν = –379.4i

C	-3.896776	1.640267	-2.964318
C	-5.188512	1.365007	-3.396955
C	-5.509447	0.089238	-3.877942
C	-4.537748	-0.908140	-3.935547
C	-3.225244	-0.650528	-3.529638
C	-2.928547	0.624576	-3.057849
H	-5.967435	2.117040	-3.373657
H	-6.522749	-0.118832	-4.209810
H	-4.795195	-1.896170	-4.308175
H	-2.467030	-1.423705	-3.591622
N	-1.679207	1.103333	-2.642988
C	-3.225618	2.900420	-2.449971
C	-1.748922	2.426769	-2.294975
O	-0.810046	3.120449	-1.921984

C	-0.457428	0.321114	-2.622955
H	0.348270	0.959618	-2.264164
H	-0.213614	-0.036560	-3.628026
H	-0.563701	-0.537466	-1.953029
C	-4.502635	4.871491	-3.468611
C	-3.236571	4.072720	-3.479240
H	-4.471935	5.940700	-3.357569
H	-5.463041	4.420202	-3.656928
H	-3.071198	3.648687	-4.470956
H	-2.391511	4.730351	-3.263074
I	-4.774421	5.646080	-6.166521
H	-5.264409	-0.130502	1.446079
H	-3.789624	0.420753	3.374322
N	-6.985406	5.402499	-0.798182
C	-6.301190	4.133944	-1.065025
C	-4.833478	4.494283	-1.204646
C	-4.731982	5.838498	-0.825776
C	-6.109674	6.386724	-0.639706
H	-6.471208	3.458754	-0.219386
H	-6.749179	3.670021	-1.947157
C	-3.655475	3.542975	-1.079845
H	-8.901454	5.162703	-1.609897
C	-8.429194	5.502605	-0.683660
O	-6.315157	7.598809	-0.342904
O	-3.738244	6.626237	-0.642090
H	-8.697868	6.542075	-0.500558
H	-8.783730	4.882658	0.145482
I	-3.579509	10.871611	0.480070
H	-2.813823	4.212000	-0.880790
Mg	-4.457877	8.456239	-0.142218
H	-2.266797	2.388865	3.222726
H	-2.226494	3.768113	1.187023
C	-3.723614	2.620766	0.138780
C	-4.574672	1.510709	0.241921
C	-4.596292	0.725985	1.395454
C	-3.770594	1.033373	2.476398
C	-2.919607	2.134915	2.391098
C	-2.899159	2.915777	1.235880
H	-5.227111	1.238024	-0.578189

P(*si*)
E = -7216.35
N_{imag} = 0

C	-3.829522	1.590572	-2.851291
C	-5.112220	1.203118	-3.209753
C	-5.357842	-0.137500	-3.537585
C	-4.323098	-1.072374	-3.505031
C	-3.022291	-0.697637	-3.149091
C	-2.801036	0.638154	-2.827412
H	-5.927653	1.920689	-3.242009
H	-6.360513	-0.447896	-3.818640
H	-4.524066	-2.109552	-3.761112
H	-2.222522	-1.430448	-3.130241
N	-1.597488	1.252128	-2.454343
C	-3.263042	2.936590	-2.469198

C	-1.772254	2.597709	-2.220377
O	-0.875835	3.376061	-1.906157
C	-0.320756	0.573020	-2.352828
H	0.429329	1.303979	-2.054310
H	-0.037264	0.139298	-3.317019
H	-0.367244	-0.223671	-1.603634
C	-3.137223	5.352644	-2.873341
C	-3.377280	4.016440	-3.575934
H	-2.071188	5.574436	-2.823208
H	-3.618478	6.175632	-3.408772
H	-4.386153	3.972252	-3.994790
H	-2.674432	3.853100	-4.397595
C	-5.356361	2.108442	1.927995
H	-4.259891	1.308441	3.607105
H	-6.333879	1.895781	2.354050
N	-3.297923	7.160521	-0.115209
C	-2.750506	5.823480	-0.361062
C	-3.682050	5.178793	-1.404946
C	-4.933048	6.046755	-1.290630
C	-4.550382	7.353859	-0.577035
H	-1.720370	5.906908	-0.712899
H	-2.742064	5.266136	0.579379
C	-4.020587	3.648010	-1.292387
H	-2.387039	7.730575	1.686678
C	-2.561497	8.122079	0.677856
O	-5.267351	8.348583	-0.458831
O	-6.049469	5.802554	-1.696082
H	-3.139312	9.043563	0.742472
H	-1.592203	8.328895	0.212095
C	-5.267329	2.707510	0.671664
H	-5.059860	3.607164	-1.630637
H	-1.888353	2.877948	0.401700
H	-2.041888	1.812445	2.595438
H	-6.183408	2.949036	0.138693
C	-4.026684	2.998911	0.082041
C	-2.870679	2.665790	0.805899
C	-2.955258	2.061272	2.060251
C	-4.197086	1.779239	2.629180

P(*re*)
E = -7214.57
N_{imag} = 0

C	-3.764571	1.417645	-2.832119
C	-5.021274	0.942401	-3.183078
C	-5.191533	-0.413108	-3.493120
C	-4.103062	-1.284202	-3.465911
C	-2.821647	-0.820828	-3.150865
C	-2.676505	0.530032	-2.847838
H	-5.881310	1.602986	-3.225447
H	-6.177759	-0.784293	-3.758096
H	-4.243824	-2.335044	-3.705706
H	-1.973470	-1.497100	-3.157778
N	-1.489976	1.221695	-2.570946
C	-3.259397	2.807079	-2.509078
C	-1.727799	2.565608	-2.417215

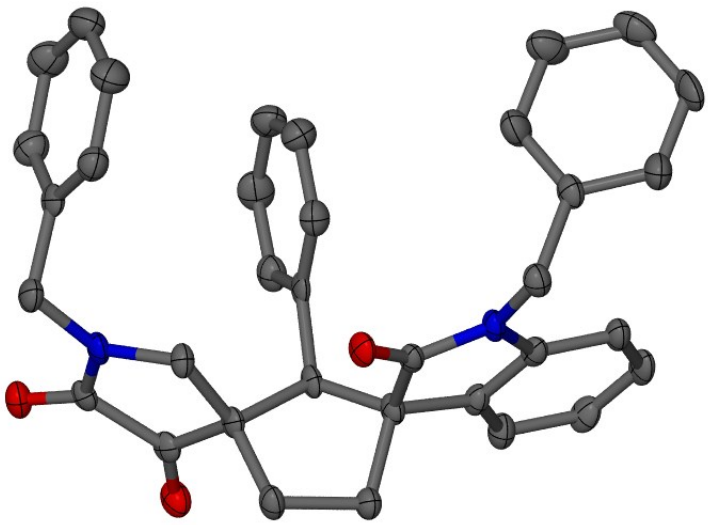
O	-0.870065	3.420387	-2.221587
C	-0.170251	0.621136	-2.537201
H	0.550534	1.399811	-2.292223
H	0.083725	0.188664	-3.510428
H	-0.127562	-0.163106	-1.775499
C	-4.081721	5.130214	-2.887498
C	-3.619618	3.859025	-3.607349
H	-3.226930	5.766096	-2.643807
H	-4.777714	5.721274	-3.488715
H	-4.421521	3.457507	-4.229910
H	-2.773514	4.061851	-4.268284
C	-3.245375	2.288136	2.416251
H	-5.821868	0.261520	1.539445
H	-4.353259	0.772474	3.483556
N	-6.958311	5.279355	-0.983190
C	-6.204404	4.254661	-1.702794
C	-4.715809	4.634396	-1.559258
C	-4.751453	5.783956	-0.557827
C	-6.212705	6.164676	-0.287493
H	-6.436442	3.281775	-1.265876
H	-6.528971	4.239075	-2.748102
C	-3.685363	3.526673	-1.156095
H	-8.772620	5.356216	-2.027287
C	-8.406353	5.269429	-0.998752
O	-6.597672	7.094622	0.421374
O	-3.805162	6.353250	-0.058111
H	-8.770034	6.110817	-0.409918
H	-8.784959	4.334451	-0.570688
C	-3.095397	2.973163	1.211507
H	-2.801295	4.131232	-0.936096
H	-5.544054	1.442466	-0.571137
H	-2.589555	2.520934	3.251686
H	-2.321330	3.731614	1.131656
C	-3.919334	2.708740	0.103310
C	-4.898543	1.713790	0.253051
C	-5.053572	1.026789	1.457470
C	-4.230502	1.310056	2.546577

11. Crystallographic Data

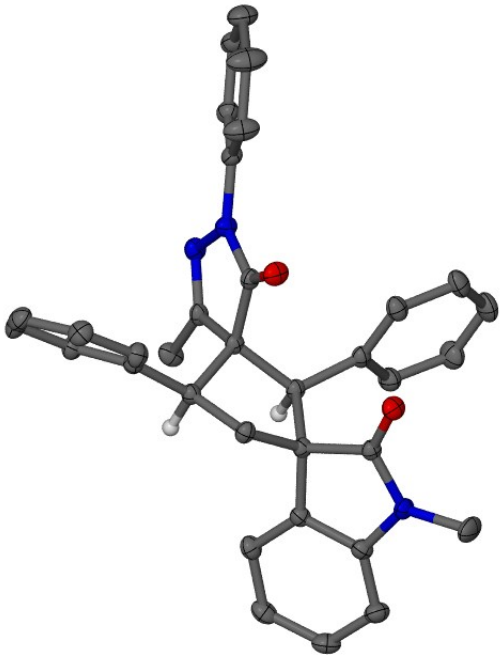
Crystal structures of all the compounds were determined by Single Crystal X-ray Diffraction (SCD) experiment. The structure of **3ba** was determined on a Bruker SMART APEX II CCD diffractometer with graphite monochromated Mo K_α (λ = 0.71073 Å) radiation. Structure of syn-**5ea** and anti-**5ea** were determined on Bruker D8 Quest equipped with a microfocus anode (Mo) and a PHOTON 100 CMOS detector. Integration and scaling of data were performed by SAINT[1] and SADABS program[2]. The structures were solved by direct methods using SHELXT-2018[3] and refined by full-matrix least-squares on F^2 using SHELXL-2018/3 version[3]. All non-hydrogen atoms were refined anisotropically and all hydrogen atoms were placed at calculated positions using riding models.

ORTEP diagram: Atoms are shown with 50% probability of thermal ellipsoids

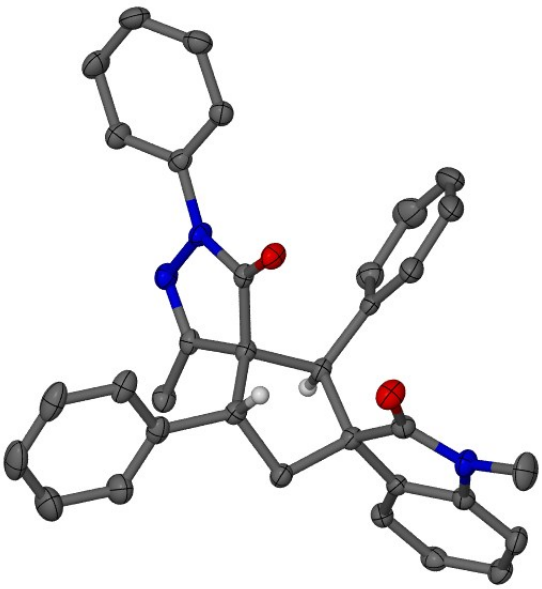
Compound: 3ba [CCDC No. 1911435]



Compound: *syn*-5ea [CCDC No. 1911436]



Compound: *anti*-5ea [CCDC No. 1911437]



References:

1. SAINT, Version 6.45; Bruker AXS Inc.: Madison, WI, 2003.

2. SADABS, Version 2.05; Bruker AXS Inc.: Madison, WI, 2002.

3. Sheldrick, G. M. *Acta Cryst. Sect. A.* **2015**, *71*, 3-8

Table S2. Crystal data and refinement parameters

Code	**3ba**	**syn-5ea**	**anti-5ea**
Empirical formula	$C_{35}H_{30}N_2O_3$, H_2O	$C_{34}H_{29}N_3O_2$	$C_{34}H_{29}N_3O_2$
Formula weight	544.62	511.60	511.60
Wavelength/ Å	0.71073	0.71073	0.71073
Crystal system	Triclinic	monoclinic	monoclinic
Space group	*P*-1	$P2_1/c$	$P2_1/c$
Crystal size ($mm^{3)}$	0.31 x 0.28 x 0.15	0.22 x 0.18 x 0.10	0.22 x 0.18 x 0.08
a/Å	9.854(3)	14.6308(9)	10.819(3)
b/Å	10.995(4)	11.7018(7)	18.779(4)
c/Å	13.297(5)	17.2952(10)	14.234(4)
α/(°)	96.31(2)	90	90
β/(°)	98.53(2)	103.047(2)	111.210(8)
γ/(°)	100.45(3)	90	90
V/$Å^3$	1387.2(8)	2884.6(3)	2696.1(11)
Z	2	4	4
D_{cal} /g cm^{-3}	1.304	1.178	1.260
T/K	100	100	100
μ/mm^{-1}	0.085	0.074	0.079
F_{000}	576	1080	1080
Theta ranges for data collection	1.9° to 25.3°	2.1° to 30.6°	2.3° to 33.2°
Index ranges	-11 ≤ h ≤ 11, -13 ≤ k ≤ 13, -15 ≤ l ≤ 15	-19 ≤ h ≤ 20, -16 ≤ k ≤ 16, -24 ≤ l ≤ 24	-16 ≤ h ≤ 16, -28 ≤ k ≤ 28, -21 ≤ l ≤ 19
Reflections measured	17877	40615	60389
Unique reflections	4941	8802	10227
Observed reflections	3717	7255	8168
Parameters	379	354	354
Data completeness	0.982	0.995	0.990
R_{int}	0.081	0.036	0.042
final *R* (I >2σ(I))	0.0667	0.0430	0.0456
final *R* (all data)	0.0854	0.0547	0.0622
final wR_2 (I >2σ(I))	0.1699	0.1104	0.1159
final wR_2 (all data)	0.1834	0.1205	0.1285
GOF on F^2	1.01	1.03	1.037

Highest peak and deepest hole	0.445 & -0.394	0.382 & -0.210	0.438 & -0.230
CCDC No.	1911435	1911436	1911437

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