Supporting Information

Rongalite-promoted self-dimerization of 3-acylidene-2-oxindoles: A diastereoselective route to synthesis of bispirooxindoles.

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1. General information

All reactions were carried out in either 10 mL reaction tubes or 50 mL pair-shaped round bottom flask. Reactions progressed was monitored by analytical thin layer chromatography (TLC). TLC performed on pre-coated silica gel plates. After development, TLC plates were visualized under UV illumination at 254 nm and further stanning with basic $KMnO_4$ solution or exposure to Iodine (I_2) chamber. ^{1}H, ^{13}C, 135 DEPT and ^{19}F NMR spectrum were recorded in deuterated solvents at room temperature on Bruker: Ultra shield 400/500/101 and 376 MHz respectively, chemical shifts (δ) are reported for ^{1}H NMR in ppm from TMS as an internal standard and for ^{13}C NMR from the residual solvent peak. ^{1}H NMR spectral data are reported as follows: chemical shift (δ ppm), multiplicity, coupling constant (Hz), and integration. Data for ^{13}C NMR spectra are reported in terms of chemical shift (δ ppm). High resolution mass spectral (HRMS) analysis was recorded by using a Thermo Scientific Q-Exactive, Accela 1250 pump. ESI TOF mass analyzer. FTIR spectra were recorded on a Perkin Elmer spectrometer. Melting points were measured with a YUHUA X-5 melting point instrument. All other solvents and chemicals were used without purification as commercially available.

2. Single crystal X-ray diffraction study

SC-XRD Experiments: The single crystals of **4Aa** and **4Bf** components were obtained from mixture of ethyl acetate : DCM solvent, by slow evaporation method. The X-ray diffraction measurements were performed to determine the crystal structure of all the two components at 150 K using APEX3 (Bruker, 2016; Bruker Smart Apex Duo) diffractometer having graphite-monochromatized (MoKα = 0.71073 Å). The X-ray generator was operated at 50 kV and 30 mA. A preliminary set of unit cell parameters and an orientation matrix were calculated from 36 frames, and the cell refinement was performed by SAINT-Plus (Bruker, 2016). An optimized strategy used for data collection consisted of different sets of φ and ω scans with 0.5^{o} steps φ/ω. The data were collected with a time frame of 10 sec for both the components by setting the sample to detector distance fixed at 40 cm. All the data points were corrected for Lorentzian, polarization, and absorption effects using SAINT-Plus and SADABS programs (Bruker, 2016). SHELXS-97 (Sheldrick, 2008) was used for structure solution, and full-matrix least-squares refinement on F^2.[1, 2] The molecular graphics of ORTEP diagrams were performed by Mercury software. The crystal symmetry of the components was cross-checked by running the cif files through PLATON (Spek, 2020) software and notified that no additional symmetry was observed. The Encifer software was used to correct the cif files. Crystallographic data files are for the **4Aa** and **4Bf** have deposited with Cambridge crystallographic data center: CCDC number : **2447899** (**4Aa**) and **2447900** (**4Bf**)

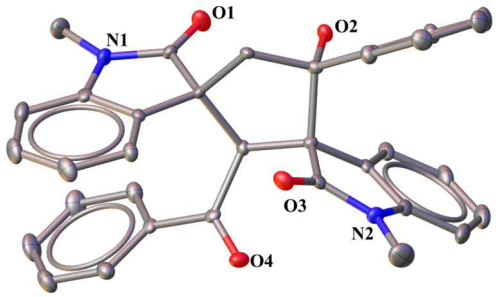


Figure 1. ORTEP diagram of compound **4Aa**, the asymmetric unit contains a single molecule. Herein, the ellipsoids are drawn with a 50% probability.

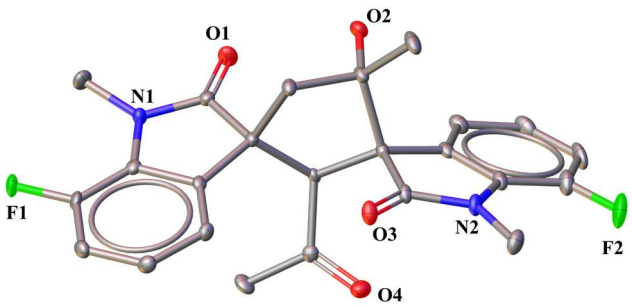


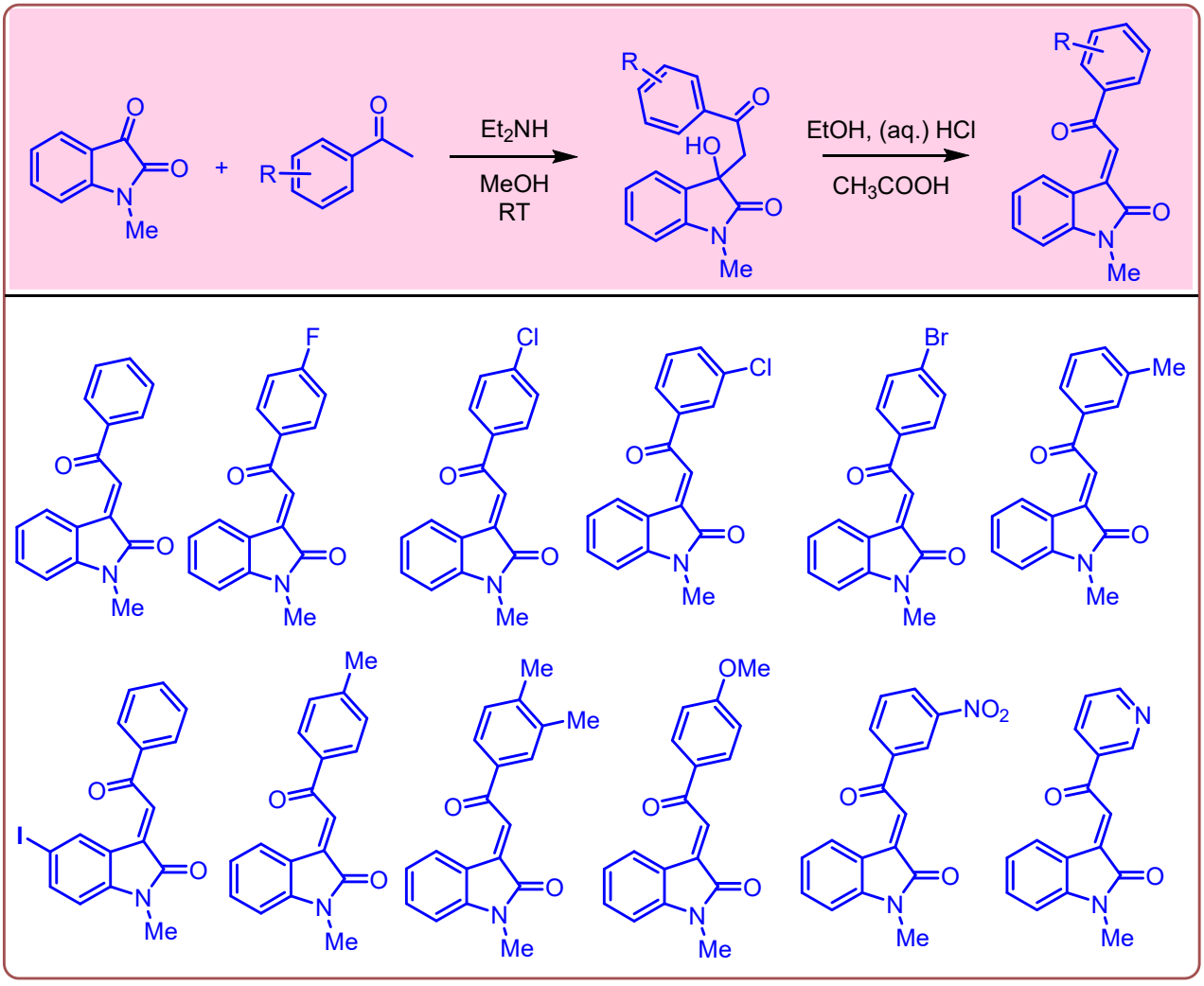
Figure 2. ORTEP diagram of compound **4Bf**, the asymmetric unit contains a single molecule. Herein, the ellipsoids are drawn with a 50% probability.

Table 1: Crystal data and structure refinement for 4Aa	
Identification code	4Aa
Empirical formula	$C_{34}H_{28}N_2O_4$
Formula weight	528.58
Temperature/K	150(2)
Crystal system	monoclinic
Space group	$P2_1/c$
a/Å	13.900(3)
b/Å	13.924(3)
c/Å	14.228(3)
α/°	90
β/°	102.004(6)
γ/°	90
Volume/Å^3	2693.4(9)
Z	4
ρ_{calc}g/cm^3	1.304
μ/mm^{-1}	0.086
F(000)	1112.0
Crystal size/mm^3	0.3 × 0.2 × 0.1
Radiation	MoKα (λ = 0.71073)
2Θ range for data collection/°	4.138 to 56.926
Index ranges	$-16 \leq h \leq 18$, $-17 \leq k \leq 18$, $-19 \leq l \leq 17$
Reflections collected	56471
Independent reflections	6764 [R_{int} = 0.2082, R_{sigma} = 0.1577]
Data/restraints/parameters	6764/240/366
Goodness-of-fit on F^2	1.116
Final R indexes [I>=2σ (I)]	R_1 = 0.1151, wR_2 = 0.1473
Final R indexes [all data]	R_1 = 0.2140, wR_2 = 0.1734
Largest diff. peak/hole / e Å^{-3}	0.30/-0.30

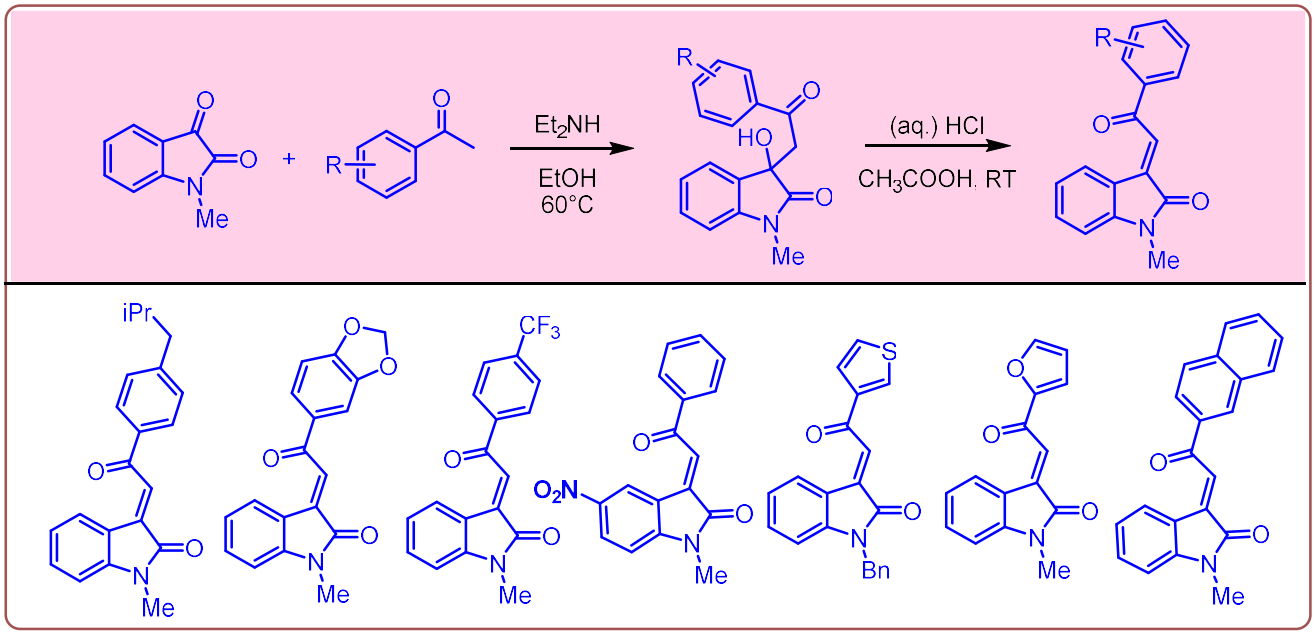
Table 2: Crystal data and structure refinement for 4Bf	
Identification code	4Bf
Empirical formula	$C_{24}H_{22}F_2N_2O_4$
Formula weight	440.15
Temperature/K	150(2)
Crystal system	trigonal
Space group	$P3_1$
a/Å	11.106(3)
b/Å	11.106(3)
c/Å	14.374(6)
α/°	90
β/°	90
γ/°	120
Volume/Å^3	1535.5(10)
Z	1
ρ_{calc}g/cm^3	1.429
μ/mm^{-1}	0.110
F(000)	690.0
Crystal size/mm^3	0.13 × 0.23 × 0.32
Radiation	MoKα (λ = 0.71073)
2Θ range for data collection/°	4.234 to 56.74
Index ranges	$-11 \leq h \leq 14$, $-14 \leq k \leq 14$, $-19 \leq l \leq 19$
Reflections collected	37274
Independent reflections	5101 [R_{int} = 0.0925, R_{sigma} = 0.0714]
Data/restraints/parameters	5101/1/295
Goodness-of-fit on F^2	0.958
Final R indexes [I>=2σ (I)]	R_1 = 0.0560, wR_2 = 0.1282
Final R indexes [all data]	R_1 = 0.0884, wR_2 = 0.1462
Largest diff. peak/hole / e Å^{-3}	0.27/-0.34
Flack parameter	0.5(4)

3. General procedure for synthesis of 3-Acylidene oxindole

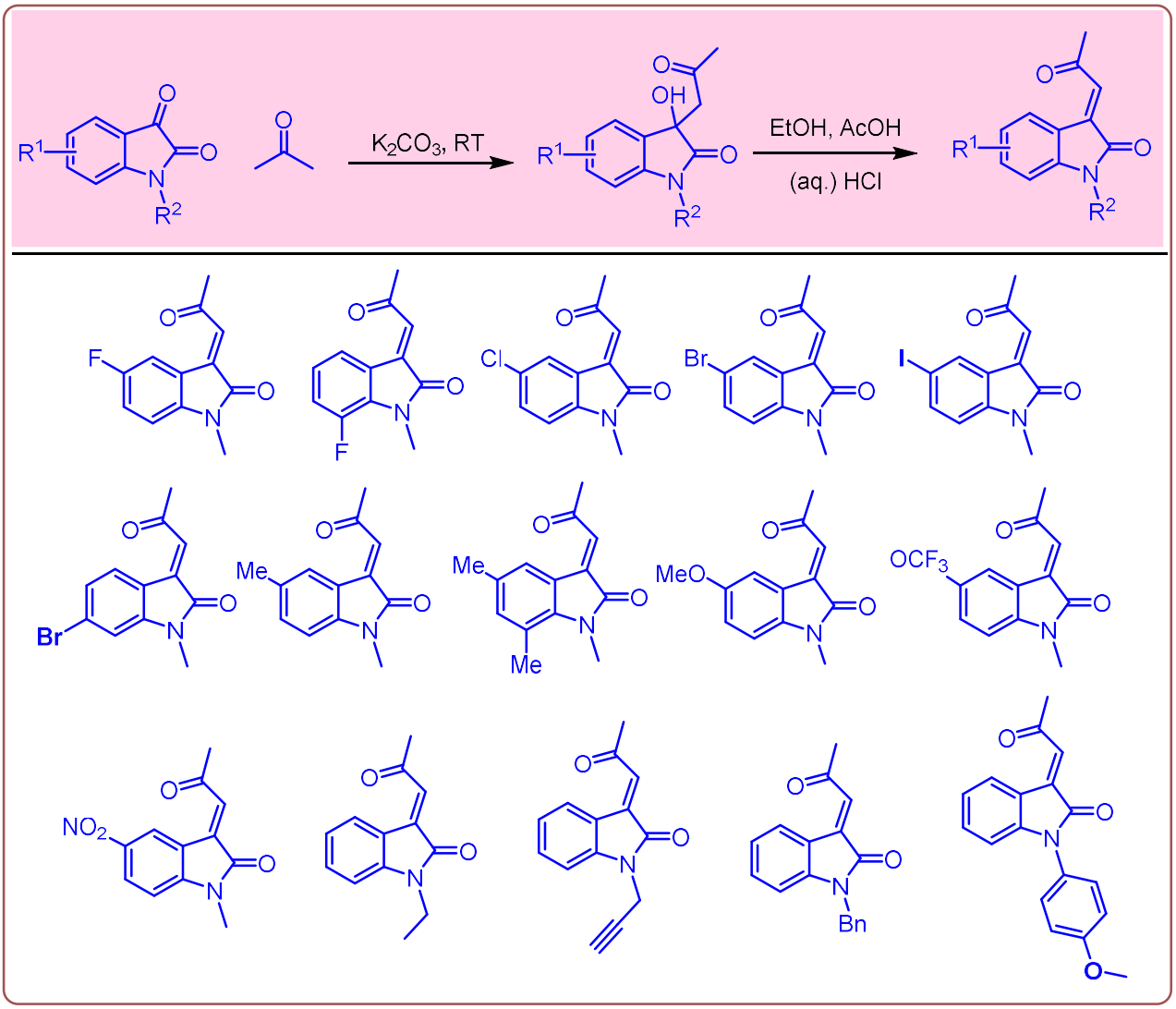
N-methylisatin (1 equiv.), substituted acetophenone (1.5 equiv.) and diethylamine (1 equiv.) in MeOH solvent were stirred at room temperature gave the intermediate crude aldol addition product. Dehydration of this intermediate in EtOH solvent using HCl (aq.) and glacial ethanoic acid and stirred at room temperature for 24 h. After that added ethyl acetate and water to the reaction mixture then organic layer was washed with H_2O and saturated aqueous $NaHCO_3$ solution. The crude reaction mixture was further purified by column chromatography to provided 3-phenacylidene oxindole. [3-4]



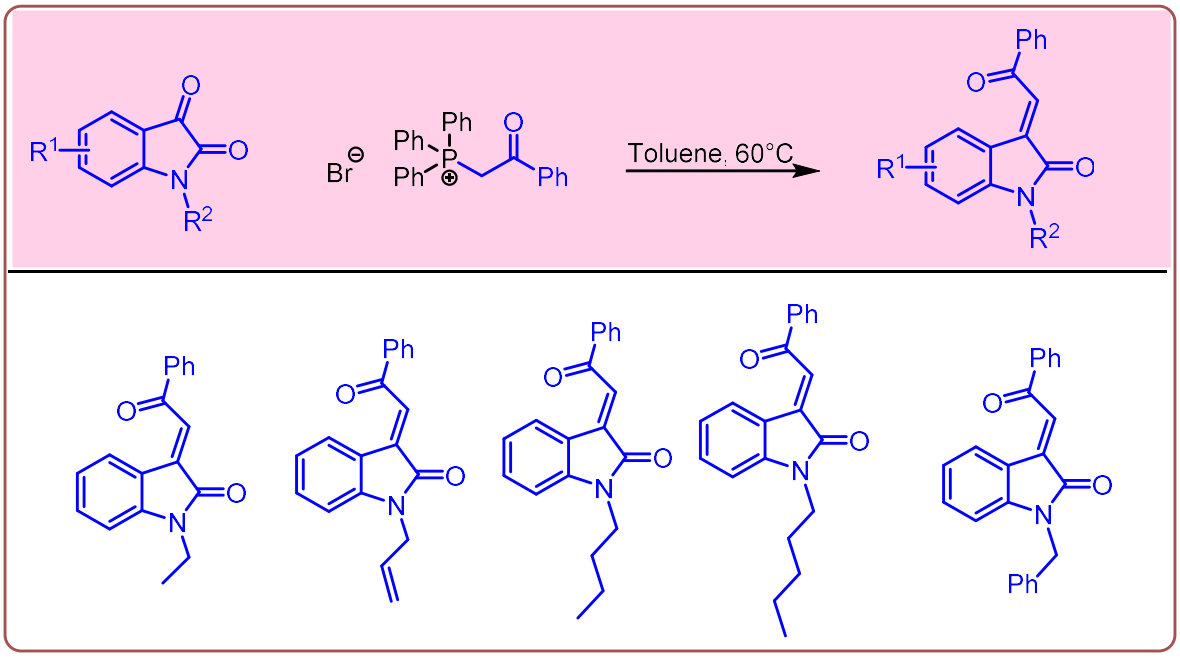
To a solution of N-methylisatin (1.0 equiv.) and ketone (1.0 equiv.) in EtOH (concentration 0.1 mmol N-methyl isatin/mL), diethylamine (1.1 equiv.) was added, and the mixture was stirred at 60° C until N-methylisatin was consumed (monitored by TLC). After being cooled to room temperature, CH_3COOH (1.2 equiv.) was added to the mixture, and the mixture was concentrated under vacuum. To the reaction mixture, water was added, and extracted with EtOAc. Organic layers were combined, washed with brine, dried over Na_2SO_4, filtered, and concentrated to give corresponding aldol product. This was dissolved in EtOH, and CH_3COOH (3 mL) and aqueous HCl solution (35%, 1 mL) were added at room temperature (25 °C). The mixture was stirred at same temperature until the aldol product was consumed (monitored by TLC). And the organic layer was washed with H_2O and saturated aqueous $NaHCO_3$ solution. The mixture was directly purified by flash column chromatography to provided 3-acylidene oxindole. [3-4]



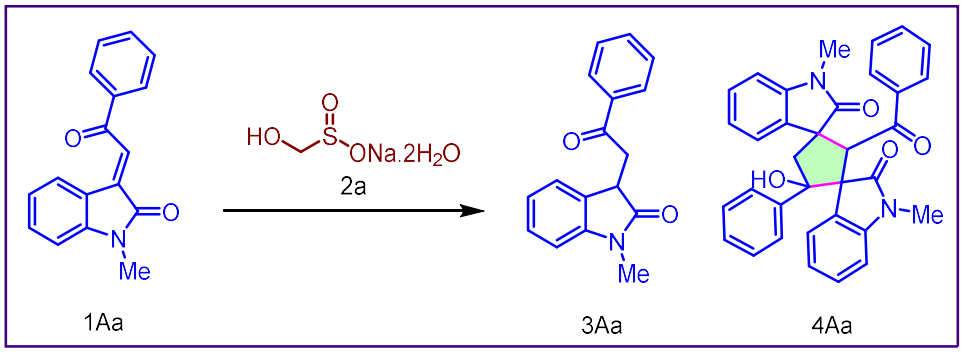
N-substituted isatin (1.0 equiv.) and K_2CO_3 (0.5 equiv.) in acetone were stirred at room temperature overnight. The resulting suspension was filtered and the solid washed with acetone then dried under vacuum to afford the Aldol product. This was dissolved in EtOH, and CH_3COOH and aqueous HCl solution (35%) were added at room temperature (25 °C). The mixture was stirred at same temperature until the aldol product was consumed (monitored by TLC). And the organic layer was washed with H_2O and saturated aqueous $NaHCO_3$ solution. The organic layer was dried over Na_2SO_4 and concentrated in vacuo. The crude reaction mixture was directly purified by flash column chromatography to provide 3-acylidene oxindole. [5]



To a solution of N-protected isatin (1.0 equiv.) in toluene (concentration 0.1 mmol N-protected isatin/mL), Witting reagent (1.2 equiv.) was added, and the mixture was stirred at 60° C, until the N-protected isatin was consumed (monitored by TLC). After being cooled to room temperature, the mixture was directly purified by flash chromatography or by crystallization to afford 3-acylidene oxindole. [6]



4. Table 1 Optimization of reaction conditions. [a]

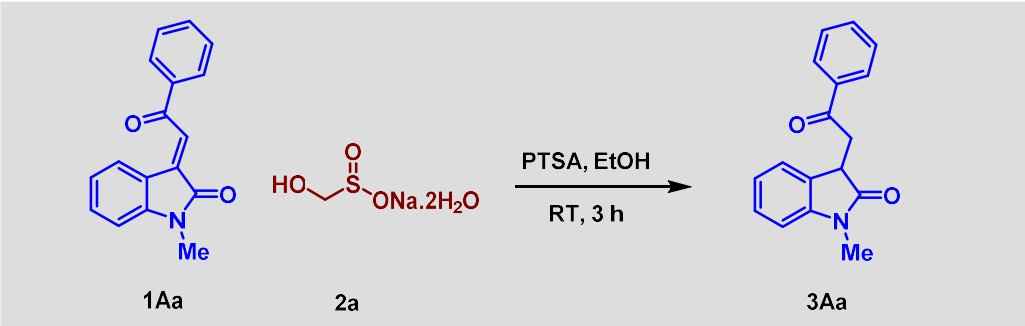


Entry	Additive (equiv.)	Solvent	Time h	(3Aa) [b]	(4Aa) [b]
1.	-	H_2O	24 h	NR	NR
2.	-	EtOH	12 h	52	ND
3.	**PTSA (2)**	**EtOH**	**3 h**	**85**	**ND**
4.	AcOH (2)	EtOH	7 h	74	ND
5.	Et_3N (2)	EtOH	24 h	10 [c]	54 [c]
6.	Et_3N (2)	H_2O	24 h	10 [c]	32 [c]
7.	Et_3N (2)	EtOH: H_2O (1:2)	1 h	ND	87
8.	-	EtOH: H_2O (1:2)	24 h	25 [c]	15 [c]
9.	DBU (2)	EtOH: H_2O (1:2)	24 h	ND	40
10.	Et_2NH (2)	EtOH: H_2O (1:2)	4 h	ND	62
11.	KOH (2)	EtOH: H_2O (1:2)	24	ND	65
12.	K_2CO_3 (2)	EtOH: H_2O (1:2)	2 h	ND	78
13.[d]	**Et_3N (1.5)**	**EtOH: H_2O (1:2)**	**2 h**	**ND**	**90**

[a] **Reaction condition**: (1a) 0.2 mmol, (2a) 0.4 mmol and Base 0.4 mmol were stirred in 3 mL solvent at room temperature, [b] Isolated yield), [c] NMR yield, [d] (2a) 0.3 mmol, 2.1 mL solvent & d. r. 99:1, ND= Not detected & NR= No Reaction.

5. Procedure A: Synthesis of 3Aa:

In 10 ml reaction tube sequential added **1Aa** 0.2mmol, **2a** 0.4 mmol and p-toluenesulphonic acid 0.4 mmol in ethanol solvent were stirred at room temperature until the starting material was completely converted to product (monitored by TLC). After that, added H_2O and EtOAc to separate the organic layer, dried over Na_2SO_4 and concentrated in vacuo. The crude reaction mixture was purified by flash column chromatography to obtain the desired product **3Aa**.



1Aa 2a 3Aa

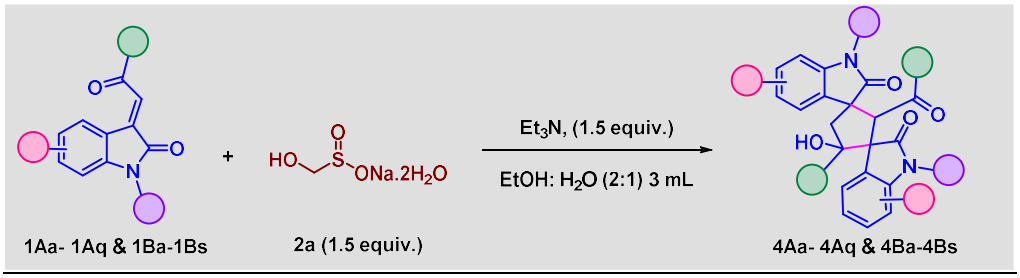
6. General procedure for dimerization reaction

Procedure B:

All reactions were carried out in 10 ml reaction tube. substituted 3-acylidene 2-oxindoles (0.2 mmol, 1 equiv.), (2a, 1.5 equiv.) and triethyl amine (1.5 equiv.) in [EtOH: H_2O (1:2)] 2.1 mL were stirred at room temperature until the starting material was completely converted to product (monitored by TLC). After that, the product was isolated by filtration and washed with H_2O 2-3 times and dry at 50° C under high vacuo to obtain the desired product.

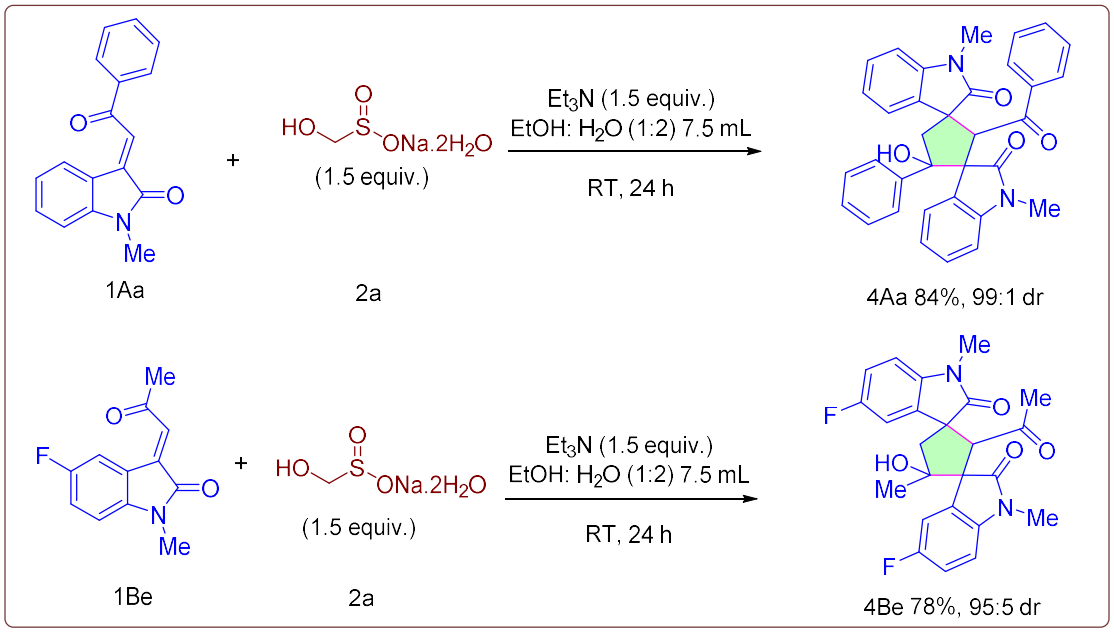
Procedure C:

All reactions were carried out in 10 ml reaction tube. substituted 3-acylidene 2-oxindoles (0.2 mmol, 1 equiv.), (2a, 1.5 equiv.) and triethyl amine (1.5 equiv.) in [EtOH: H_2O (1:2)] 2.1 mL were stirred at room temperature until the starting material was completely converted to product (monitored by TLC). After that, if precipitate was not formed then added H_2O and EtOAc to separate the organic layer, dried over Na_2SO_4 and concentrated in vacuo. The crude reaction mixture was purified by flash column chromatography to obtain the desired product.



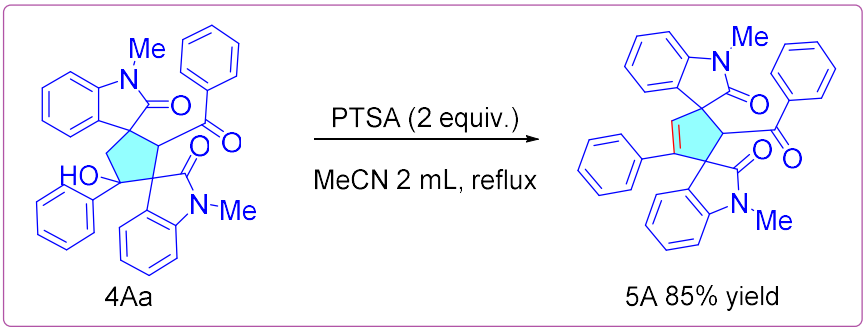
7. General procedure for gram-scale synthesis of 4Aa & 4Be

In 25 mL reaction tube, added sequentially 1Aa, (1 gm, 1 equiv.), 2a and triethyl amine (1.5 equiv.) in [EtOH: H_2O (1:2)] 4.5 mL solvent were stirred at room temperature for 10 min. under stirring second portion of remaining same solvent 3 mL was added slowly to the reaction mixture, continuously stirred for 2 h at room temperature until the starting material was completely converted to product (monitored by TLC). After that, the product was isolated by filtration and washed with H_2O 2-3 times and dry at 50° C under high vacuo to obtain the desired product.



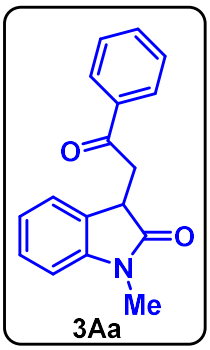
8. Procedure D: Synthetic transformation of 4Aa

An oven- dried 10 mL reaction tube was equipped with a magnetic stirrer, product 4Aa in (0.1 mmol, 1.0 equiv.) dissolved in acetonitrile at room temperature were added in p-toluensulfonic acid (p-TSA) (0.2 mmol, 2.0 equiv.), were stirred at 90 °C for 15 h. The reactions were stopped by the addition of H_2O and diluted by ethyl acetate (3 x 15 mL). The combined organic phases were dried over Na_2SO_4. The organic solvent was evaporated under reduced pressure. The crude product was purified by column chromatography (petroleum ether/ethyl acetate 4:1) to afford the elimination product 5A in 85% yield.



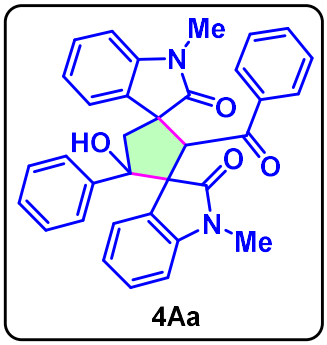
4Aa 5A 85% yield

9. Analytical data for synthesized compound



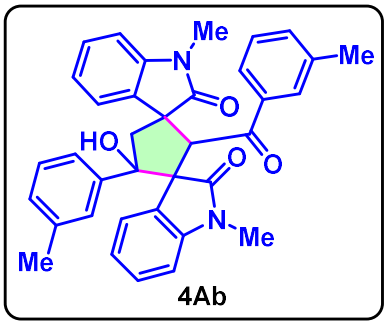
3Aa

1-methyl-3-(2-oxo-2-phenylethyl)indolin-2-one, (General Procedure A), obtained as a solid, 85% yield, mp 170-172 °C, **^{1}H NMR** (400 MHz, $CDCl_3$) δ = 7.90 (d, *J* = 7.8 Hz, 2 H), 7.56 - 7.46 (m, 1 H), 7.45 - 7.31 (m, 2 H), 7.25 - 7.11 (m, 2 H), 7.00 - 6.86 (m, 1 H), 6.78 (d, *J* = 7.8 Hz, 1 H), 4.01 (d, *J* = 9.0 Hz, 1 H), 3.76 (d, *J* = 18.3 Hz, 1 H), 3.40 - 3.25 (m, 1 H), 3.25 - 3.09 (m, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 197.0, 177.8, 144.3, 136.3, 133.4, 129.1, 128.7, 128.1, 128.1, 124.4, 122.5, 108.0, 41.2, 40.0, 26.4; **HRMS** (ESI) m/z calculated for $C_{17}H_{15}NO_2$+H 266.1181 found: 266.1176

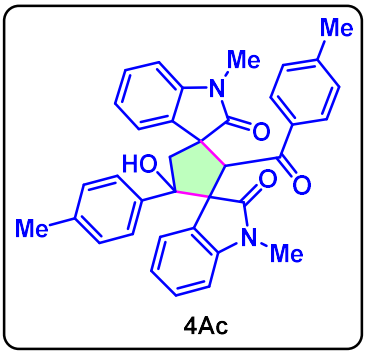


4Aa

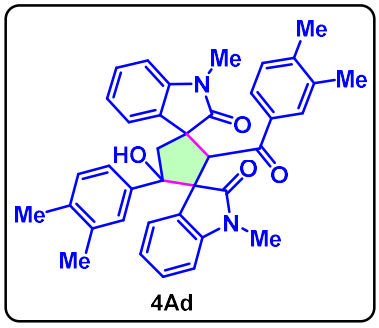
[4Aa] **2'-benzoyl-4'-hydroxy-1,1''-dimethyl-4'-phenyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (90% yield, 99:1 *d.r.*). mp 222-224 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.28 (d, *J* = 7.3 Hz, 1 H), 7.86 (d, *J* = 7.4 Hz, 1 H), 7.36 - 7.19 (m, 9 H), 7.14 - 7.05 (m, 3 H), 7.03 (d, *J* = 9.5 Hz, 1 H), 6.89 (d, *J* = 0.4 Hz, 1 H), 6.61 (d, *J* = 7.8 Hz, 1 H), 6.28 (d, *J* = 7.4 Hz, 1 H), 5.23 (s, 1 H), 4.36 (d, *J* = 14.0 Hz, 1 H), 3.09 (s, 3 H), 2.90 (s, 3 H), 2.45 (d, *J* = 13.9 Hz, 1 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 196.4, 183.5, 176.5, 144.4, 142.4, 137.8, 137.1, 132.2, 130.3, 128.6, 128.4, 127.6, 127.6, 127.5, 127.2, 127.0, 127.0, 126.9, 126.6, 125.6, 125.5, 124.2, 121.7, 107.5, 107.4, 84.6, 66.9, 64.5, 54.3, 45.5, 26.6, 25.8; **HRMS** (ESI) m/z calculated for $C_{34}H_{28}N_2O_4$+H 529.2127 found: 529.2122



[4Ab] **4'-hydroxy-1,1''-dimethyl-2'-(3-methylbenzoyl)-4'-(m-tolyl)dispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (90% yield, 96:4 *d.r.*), mp 183-185 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.20 (d, *J* = 7.1 Hz, 1 H), 7.80 (d, *J* = 7.4 Hz, 1 H), 7.24 - 7.14 (m, 3 H), 7.06 - 6.97 (m, 3 H), 6.95 (s, 1 H), 6.92 - 6.84 (m, 4 H), 6.83 - 6.74 (m, 3 H), 6.71 (s, 1 H), 6.55 (d, *J* = 7.8 Hz, 1 H), 6.22 (d, *J* = 7.4 Hz, 1 H), 5.14 (s, 1 H), 4.26 (d, *J* = 14.0 Hz, 1 H), 3.02 (s, 3 H), 2.83 (s, 3 H), 2.36 (d, *J* = 13.9 Hz, 1 H), 2.08 (s, 3 H), 2.04 (s, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 196.8, 183.5, 176.5, 144.5, 142.4, 137.7, 137.7, 137.3, 136.6, 132.8, 130.5, 128.5, 128.2, 128.1, 128.1, 127.6, 127.3, 127.0, 126.9, 126.7, 126.4, 125.5, 124.1, 124.0, 122.5, 121.7, 107.4, 107.2, 84.6, 66.8, 64.5, 54.4, 45.5, 26.6, 25.8, 21.3, 21.0; **HRMS** (ESI) m/z calculated for $C_{36}H_{32}N_2O_4$+H 557.2440 found: 557.2435

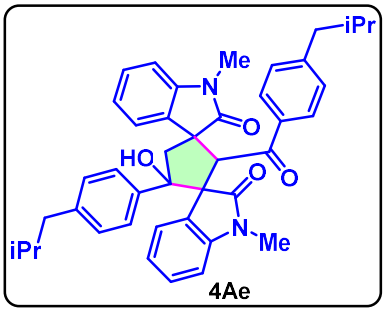


[4Ac] **4'-hydroxy-1,1''-dimethyl-2'-(4-methylbenzoyl)-4'-(p-tolyl)dispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (90% yield, 99:1 *d.r.*), mp 248-250 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.19 (d, *J* = 6.5 Hz, 1 H), 7.77 (d, *J* = 7.4 Hz, 1 H), 7.22 - 7.14 (m, 1 H), 7.05 - 6.99 (m, 1 H), 6.96 - 6.91 (m, 3 H), 6.89 - 6.84 (m, 2 H), 6.81 (d, *J* = 4.3 Hz, 3 H), 6.78 (br. s., 1 H), 5.13 (s, 1 H), 4.26 (d, *J* = 14.0 Hz, 1 H), 3.03 (s, 3 H), 2.85 (s, 3 H), 2.35 (d, *J* = 14.0 Hz, 1 H), 2.17 (s, 3 H), 2.14 (s, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 195.9, 183.6, 176.7, 144.4, 142.9, 142.4, 137.0, 135.0, 134.5, 130.4, 128.5, 128.3, 128.2, 127.8, 127.1, 126.9, 126.8, 125.5, 124.1, 121.7, 107.4, 107.3, 84.5, 66.8, 64.5, 54.4, 45.8, 26.6, 25.9, 21.5, 20.9; **HRMS** (ESI) m/z calculated for $C_{36}H_{32}N_2O_4$+H 557.2440 found: 557.2435



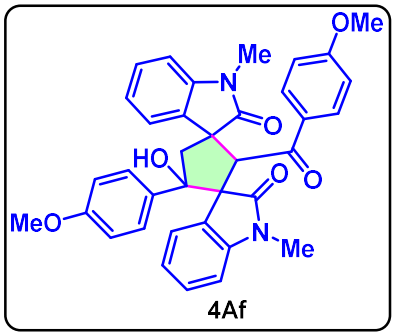
4Ad

[4Ad] **2'-(3,4-dimethylbenzoyl)-4'-(3,4-dimethylphenyl)-4'-hydroxy-1,1''-dimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (82% yield, 95:5 *d.r.*), mp 218-220 °C, **FT-IR** (KBr, cm^{-1}) 3305, 3056, 2935, 1705, 1611,1472, 1351,750; **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.20 (d, *J* = 7.1 Hz, 1 H), 7.80 (d, *J* = 7.4 Hz, 1 H), 7.24 - 7.14 (m, 1 H), 7.06 - 6.97 (m, 3 H), 6.97 - 6.83 (m, 5 H), 6.83 - 6.75 (m, 3 H), 6.71 (s, 1 H), 6.55 (d, *J* = 7.8 Hz, 1 H), 6.22 (d, *J* = 7.4 Hz, 1 H), 5.14 (s, 1 H), 4.26 (d, *J* = 14.0 Hz, 1 H), 3.02 (s, 3 H), 2.83 (s, 3 H), 2.36 (d, *J* = 13.9 Hz, 1 H), 2.06 (d, *J* = 14.3 Hz, 6 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 196.8, 183.5, 176.5, 144.5, 142.4, 137.7, 137.7, 137.3, 136.6, 132.8, 130.5, 128.5, 128.2, 128.1, 128.1, 127.6, 127.3, 127.0, 126.9, 126.7, 126.4, 125.5, 124.1, 124.0, 122.5, 121.7, 107.4, 107.2, 84.6, 66.8, 64.5, 54.4, 45.5, 26.6, 25.8, 21.3, 21.0; **HRMS** (ESI) m/z calculated for $C_{38}H_{36}N_2O_4$+H 585.2753 found: 585.2756



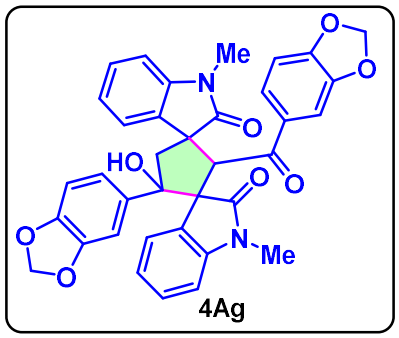
4Ae

[4Ae] **4'-hydroxy-2'-(4-isobutylbenzoyl)-4'-(4-isobutylphenyl)-1,1''-dimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure C), obtained as a solid, (78% yield, 90:10 *d.r.*), mp 222-224 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.25 (dd, *J* = 1.4, 7.1 Hz, 1 H), 7.84 (d, *J* = 6.9 Hz, 1 H), 7.26 (dd, *J* = 1.1, 15.4 Hz, 2 H), 7.16 - 7.07 (m, 1 H), 7.06 - 6.96 (m, 4 H), 6.95 - 6.88 (m, 2 H), 6.87 - 6.81 (m, 5 H), 6.60 (d, *J* = 7.6 Hz, 1 H), 6.30 - 6.23 (m, 1 H), 5.21 (s, 1 H), 4.34 (d, *J* = 13.9 Hz, 1 H), 3.11 (s, 3 H), 2.88 (s, 3 H), 2.44 (d, *J* = 13.9 Hz, 1 H), 2.41 - 2.24 (m, 4 H), 1.79 - 1.71 (m, 2 H), 0.87 - 0.75 (m, 12 H); **^{13}C NMR** (101MHz ,$CDCl_3$) δ = 196.0, 183.6, 176.7, 146.7, 144.5, 142.3, 140.8, 135.1, 134.8, 130.5, 128.6, 128.4, 128.2, 127.9, 126.9, 126.9, 126.7, 125.5, 125.2, 124.2, 121.6, 107.4, 107.2, 84.6, 66.9, 64.3, 54.4, 50.8, 45.5, 45.1, 44.8, 30.2, 30.1, 26.6, 25.7, 22.3, 22.2, 22.0; **HRMS** (ESI) m/z calculated for $C_{42}H_{44}N_2O_4$+H 641.3379 found: 641.3374



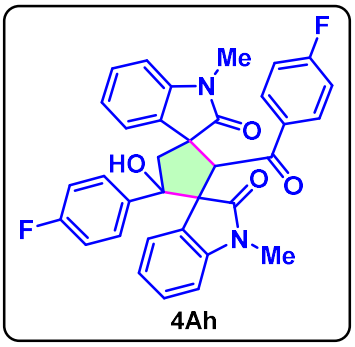
4Af

[4Af] **4'-hydroxy-2'-(4-methoxybenzoyl)-4'-(4-methoxyphenyl)-1,1''-dimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (90% yield, 96:4 *d.r.*), mp 248-250 °C, **FT-IR** (KBr, cm^{-1}) 3272, 3053, 1710, 1611, 1471, 1252, 753; **^{1}H NMR** (400MHz ,$CDCl_3$) δ = 8.18 (d, *J* = 7.5 Hz, 1 H), 7.76 (d, *J* = 7.4 Hz, 1 H), 7.18 (d, *J* = 15.1 Hz, 1 H), 7.08 - 6.88 (m, 7 H), 6.75 (s, 1 H), 6.66 - 6.41 (m, 5 H), 6.29 (d, *J* = 6.5 Hz, 1 H), 5.10 (s, 1 H), 4.26 (d, *J* = 13.9 Hz, 1 H), 3.64 (d, *J* = 9.8 Hz, 6 H), 3.07 (s, 3 H), 2.87 (s, 3 H), 2.36 (d, *J* = 13.9 Hz, 1 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 194.6, 183.6, 176.7, 162.8, 158.8, 144.4, 142.3, 130.4, 130.4, 130.0, 129.2, 128.5, 128.3, 126.9, 126.7, 125.5, 124.2, 121.6, 112.8, 112.5, 107.5, 107.4, 84.3, 66.8, 64.2, 55.4, 55.1, 54.5, 45.8, 26.7, 25.9; **HRMS** (ESI) m/z calculated for $C_{36}H_{32}N_2O_6$+H 589.2339 found: 589.2335

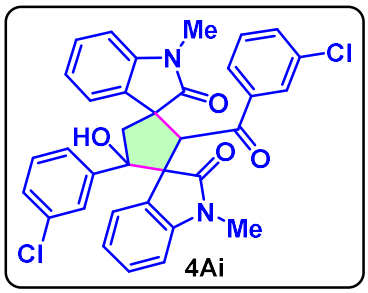


4Ag

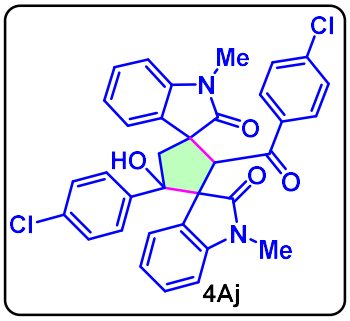
[4Ag] **4'-(benzo[d][1,3]dioxol-5-yl)-2'-(benzo[d][1,3]dioxole-5-carbonyl)-4'-hydroxy-1,1''-dimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (83% yield, 90:10 *d.r.*), mp 242-244 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.18 - 8.11 (m, 1 H), 7.74 (d, *J* = 7.4 Hz, 1 H), 7.24 - 7.19 (m, 1 H), 7.05 - 6.97 (m, 3 H), 6.80 (s, 1 H), 6.72 (dd, *J* = 1.6, 8.1 Hz, 1 H), 6.59 (d, *J* = 7.8 Hz, 1 H), 6.56 - 6.51 (m, 1 H), 6.50 - 6.41 (m, 4 H), 6.39 - 6.33 (m, 1 H), 5.80 (d, *J* = 2.4 Hz, 2 H), 5.75 (d, *J* = 6.6 Hz, 2 H), 5.02 (s, 1 H), 4.20 (d, *J* = 13.9 Hz, 1 H), 3.10 (s, 3 H), 2.93 (s, 3 H), 2.34 (d, *J* = 13.9 Hz, 1 H); **^{13}C NMR** (101MHz, $CDCl_3$) δ = 194.1, 183.6, 176.6, 151.1, 147.6, 146.7, 144.4, 142.4, 132.2, 131.8, 130.3, 128.7, 128.4, 126.9, 126.6, 125.4, 124.3, 122.7, 121.8, 119.2, 107.5, 107.4, 107.1, 106.8, 106.6, 101.6, 100.7, 84.3, 66.7, 64.5, 54.4, 46.0, 26.7, 26.0; **HRMS** (ESI) m/z calculated for $C_{36}H_{38}N_2O_8$+H 617.1924 found: 617.1918



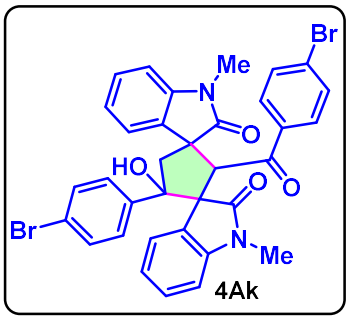
[4Ah] **2'-(4-fluorobenzoyl)-4'-(4-fluorophenyl)-4'-hydroxy-1,1''-dimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (88% yield, 97:3 d.r.), mp 242-242 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.16 (dd, *J* = 3.7, 5.1 Hz, 3 H), 7.76 (d, *J* = 7.6 Hz, 3 H), 7.25 - 7.17 (m, 4 H), 7.09 - 6.93 (m, 22 H), 6.81 (br. s., 3 H), 6.69 (dt, *J* = 2.0, 8.7 Hz, 12 H), 6.56 (d, *J* = 7.8 Hz, 3 H), 6.30 (dd, *J* = 2.9, 5.9 Hz, 3 H), 5.08 (s, 3 H), 4.25 (d, *J* = 13.9 Hz, 3 H), 3.07 (s, 9 H), 2.87 (s, 9 H), 2.37 (d, *J* = 13.9 Hz, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 194.8, 183.4, 176.5, 166.3, 163.8, 163.5, 161.0, 144.3, 142.3, 133.7, 133.7, 133.5, 133.5, 130.1, 129.6, 129.5, 128.9, 128.7, 127.6, 127.5, 126.9, 126.2, 125.5, 124.4, 121.9, 114.9, 114.7, 114.1, 113.9, 107.7, 107.7, 107.6, 84.2, 66.9, 64.4, 54.3, 45.7, 26.7, 25.9; **^{19}F NMR** (376 MHz, $CDCl_3$) δ -105.36, -115.14 ; **HRMS** (ESI) m/z calculated for $C_{34}H_{26}F_2N_2O_4$+H 565.1939 found: 565.1933



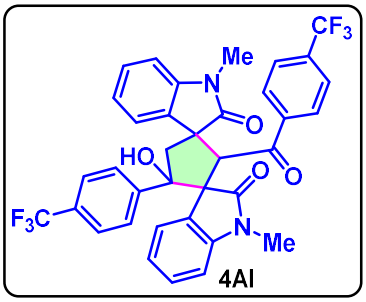
[4Ai] **2'-(3-chlorobenzoyl)-4'-(3-chlorophenyl)-4'-hydroxy-1,1''-dimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (87% yield, 98:2 *d.r.*), mp 184-186 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.15 (dd, *J* = 3.1, 5.7 Hz, 1 H), 7.77 (d, *J* = 7.4 Hz, 1 H), 7.26 - 7.20 (m, 1 H), 7.17 (d, *J* = 7.3 Hz, 1 H), 7.11 - 7.03 (m, 2 H), 7.03 - 6.99 (m, 2 H), 6.99 - 6.93 (m, 4 H), 6.92 (s, 2 H), 6.90 (br. s., 1 H), 6.59 (d, *J* = 7.6 Hz, 1 H), 6.30 (dd, *J* = 3.0, 5.8 Hz, 1 H), 5.03 (s, 1 H), 4.21 (d, *J* = 13.9 Hz, 1 H), 3.11 (s, 3 H), 2.87 (s, 3 H), 2.37 (d, *J* = 13.9 Hz, 1 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 195.1, 183.3, 176.2, 144.2, 142.4, 139.8, 138.6, 134.0, 133.3, 132.1, 129.9, 129.0, 128.7, 128.4, 127.7, 127.0, 126.8, 126.2, 125.9, 125.5, 125.0, 124.4, 123.8, 122.0, 107.7, 107.6, 84.0, 66.8, 64.5, 54.2, 45.4, 26.8, 25.9; **HRMS** (ESI) m/z calculated for $C_{34}H_{26}Cl_2N_2O_4$+H 597.1348 found: 597.1342



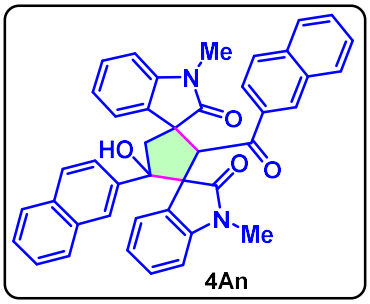
[4Aj] **2'-(4-chlorobenzoyl)-4'-(4-chlorophenyl)-4'-hydroxy-1,1''-dimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (84% yield, 98:2 *d.r.*), mp 250-252 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.20 - 8.10 (m, 1 H), 7.75 (d, *J* = 7.1 Hz, 1 H), 7.24 - 7.17 (m, 1 H), 7.05 - 6.98 (m, 4 H), 6.98 - 6.92 (m, 7 H), 6.82 (s, 1 H), 6.57 (d, *J* = 7.8 Hz, 1 H), 6.33 - 6.27 (m, 1 H), 5.06 (s, 1 H), 4.23 (d, *J* = 13.9 Hz, 1 H), 3.06 (s, 3 H), 2.88 (s, 3 H), 2.36 (d, *J* = 14.0 Hz, 1 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 195.1, 183.3, 176.3, 144.2, 142.3, 138.6, 136.3, 135.4, 133.5, 130.0, 128.9, 128.7, 128.3, 127.9, 127.3, 127.2, 126.9, 126.0, 125.5, 124.4, 121.9, 107.7, 107.6, 84.1, 66.8, 64.5, 54.3, 45.6, 26.7, 25.9; **HRMS** (ESI) m/z calculated for $C_{34}H_{26}Cl_2N_2O_4$+H 597.1348 found: 597.1342



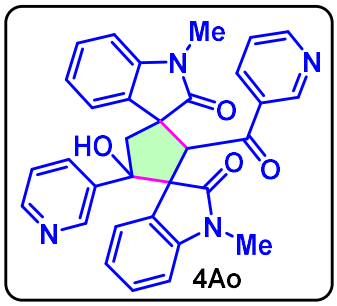
[4Ak] **2'-(4-bromobenzoyl)-4'-(4-bromophenyl)-4'-hydroxy-1,1''-dimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (82% yield, 92:8 *d.r.*), mp 232-234 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.29 - 8.16 (m, 1 H), 7.83 (d, *J* = 7.3 Hz, 1 H), 7.37 - 7.28 (m, 1 H), 7.22 (dd, *J* = 4.0, 8.5 Hz, 4 H), 7.15 - 7.02 (m, 3 H), 6.96 (dd, *J* = 5.8, 8.4 Hz, 4 H), 6.89 (s, 1 H), 6.66 (d, *J* = 7.8 Hz, 1 H), 6.43 - 6.31 (m, 1 H), 5.13 (s, 1 H), 4.29 (d, *J* = 13.9 Hz, 1 H), 3.14 (s, 3 H), 2.97 (s, 3 H), 2.43 (d, *J* = 13.9 Hz, 1 H); ^{13}C NMR (101MHz , $CDCl_3$) δ = 195.3, 183.3, 176.3, 144.2, 142.3, 136.9, 135.8, 130.9, 130.3, 130.0, 128.9, 128.8, 128.4, 127.6, 127.2, 126.9, 126.0, 125.5, 124.4, 121.9, 121.8, 107.7, 107.6, 84.1, 66.7, 64.5, 54.2, 45.5, 26.7, 26.0; **HRMS** (ESI) m/z calculated for $C_{34}H_{26}Br_2N_2O_4$+H 685.0338 found: 685.0332



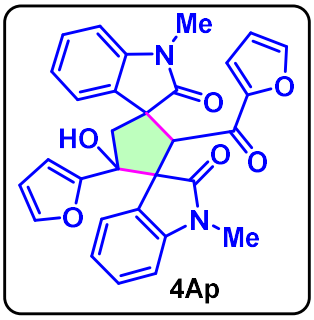
[4Al] **4'-hydroxy-1,1''-dimethyl-2'-(4-(trifluoromethyl)benzoyl)-4'-(4-(trifluoromethyl)phenyl)dispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (75% yield, 90:10 *d.r.*), mp 232-234 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.23 - 8.14 (m, 1 H), 7.80 (d, *J* = 7.4 Hz, 1 H), 7.33 - 7.20 (m, 5 H), 7.14 (d, *J* = 8.3 Hz, 2 H), 7.11 - 7.04 (m, 3 H), 7.03 - 6.96 (m, 2 H), 6.88 (s, 1 H), 6.58 (d, *J* = 7.8 Hz, 1 H), 6.20 (d, *J* = 8.4 Hz, 1 H), 5.12 (s, 1 H), 4.27 (d, *J* = 14.0 Hz, 1 H), 3.01 (s, 3 H), 2.85 (s, 3 H), 2.39 (d, *J* = 14.0 Hz, 1 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 195.7, 183.2, 176.1, 144.2, 142.3, 141.6, 140.0, 133.6, 133.3, 129.9, 129.1, 128.9, 127.2, 126.8, 126.2, 125.8, 125.6, 124.6, 124.6, 124.5, 124.5, 124.1, 124.1, 124.1, 124.1, 122.1, 107.7, 107.7, 84.2, 66.8, 64.9, 54.2, 45.5, 26.6, 25.9; **^{19}F NMR** (376 MHz, $CDCl_3$) δ = -62.62, -63.28; **HRMS** (ESI) m/z calculated for $C_{36}H_{26}F_6N_2O_4$+H 665.1875 found: 665.1870



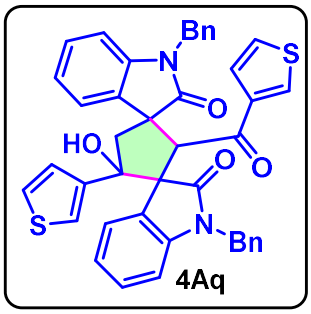
[4An] **2'-(2-naphthoyl)-4'-hydroxy-1,1''-dimethyl-4'-(naphthalen-2-yl)dispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (78% yield, 90:10 *d.r.*), mp 222-224 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.27 (d, *J* = 7.5 Hz, 1 H), 7.94 (d, *J* = 7.4 Hz, 1 H), 7.76 (d, *J* = 7.8 Hz, 1 H), 7.68 (s, 1 H), 7.65 - 7.57 (m, 3 H), 7.55 (s, 1 H), 7.49 - 7.41 (m, 3 H), 7.37 (d, *J* = 8.6 Hz, 1 H), 7.31 (t, *J* = 5.1 Hz, 2 H), 7.24 - 7.19 (m, 1 H), 7.09 (d, *J* = 7.3 Hz, 2 H), 7.04 - 6.97 (m, 3 H), 6.84 (t, *J* = 7.6 Hz, 1 H), 6.49 (d, *J* = 7.6 Hz, 1 H), 5.92 (d, *J* = 7.8 Hz, 1 H), 5.37 (s, 1 H), 4.46 (d, *J* = 13.9 Hz, 1 H), 2.81 (s, 3 H), 2.74 (s, 3 H), 2.49 (d, *J* = 14.0 Hz, 1 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 196.3, 183.6, 176.6, 144.4, 142.3, 135.5, 134.8, 134.5, 132.7, 132.4, 131.6, 130.3, 128.9, 128.7, 128.5, 128.4, 128.2, 127.8, 127.7, 127.1, 126.9, 126.7, 126.6, 125.8, 125.6, 125.5, 124.8, 124.2, 124.0, 123.2, 121.9, 107.5, 107.4, 84.7, 66.9, 64.7, 54.6, 45.8, 26.6, 25.9; **HRMS** (ESI) m/z calculated for $C_{42}H_{32}N_2O_4$+H 629.2440 found: 665.2435



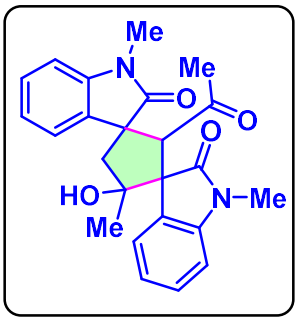
[4Ao] **4'-hydroxy-1,1''-dimethyl-2'-nicotinoyl-4'-(pyridin-3-yl)dispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (86% yield, 99:1 *d.r.*), mp 234-236 °C, **FT-IR** (KBr, cm^{-1}) 3334, 3062, 2931, 1694, 1677, 1612, 1472, 1382, 754; **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.62 - 8.19 (m, 4 H), 8.17 (d, *J* = 8.4 Hz, 1 H), 7.80 (d, *J* = 7.4 Hz, 1 H), 7.36 (d, *J* = 7.8 Hz, 1 H), 7.30 - 7.17 (m, 2 H), 7.06 (t, *J* = 7.7 Hz, 2 H), 7.00 (t, *J* = 5.6 Hz, 2 H), 6.97 (s, 1 H), 6.93 (dd, *J* = 4.8, 7.34Ba Hz, 1 H), 6.58 (d, *J* = 7.8 Hz, 1 H), 6.26 (d, *J* = 8.3 Hz, 1 H), 5.06 (s, 1 H), 4.28 (d, *J* = 13.9 Hz, 1 H), 3.10 (s, 3 H), 2.88 (s, 3 H), 2.41 (d, *J* = 14.0 Hz, 1 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 194.9, 183.0, 176.1, 152.7, 147.7, 144.0, 142.3, 134.3, 129.6, 129.3, 128.9, 126.7, 125.7, 125.3, 124.5, 122.2, 107.8, 66.8, 64.9, 54.3, 45.1, 26.8, 25.9; **HRMS** (ESI) m/z calculated for $C_{32}H_{36}N_4O_4$+H 531.2032 found: 531.2027



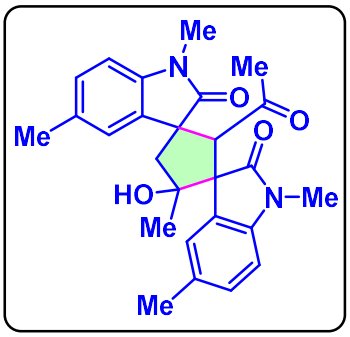
[4Ap] **2'-(furan-2-carbonyl)-4'-(furan-2-yl)-4'-hydroxy-1,1''-dimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure C), obtained as a solid, (71% yield, 90:10 *d.r.*), mp 230-232 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.14 (d, *J* = 6.8 Hz, 1 H), 7.75 (d, *J* = 7.1 Hz, 1 H), 7.33 - 7.27 (m, 1 H), 7.19 - 7.13 (m, 2 H), 7.12 - 7.00 (m, 3 H), 6.91 (s, 1 H), 6.76 (d, *J* = 7.6 Hz, 1 H), 6.61 (d, *J* = 4.0 Hz, 2 H), 6.20 - 6.10 (m, 2 H), 5.87 (d, *J* = 3.3 Hz, 1 H), 5.01 (s, 1 H), 4.15 (d, *J* = 13.8 Hz, 1 H), 3.34 (s, 3 H), 3.16 (s, 3 H), 2.57 (d, *J* = 14.1 Hz, 1 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 183.8, 183.5, 176.3, 152.2, 151.8, 144.7, 144.5, 142.6, 141.6, 129.7, 128.7, 128.4, 126.9, 126.5, 125.8, 124.2, 122.0, 116.0, 112.7, 110.0, 107.6, 107.4, 106.4, 81.7, 65.2, 63.9, 54.8, 46.4, 26.8, 26.1; **HRMS** (ESI) m/z calculated for $C_{30}H_{34}N_2O_6$+H 509.1713 found: 509.1720



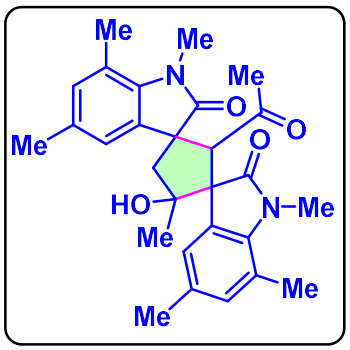
[4Aq] **1,1''-dibenzyl-4'-hydroxy-4'-(thiophen-3-yl)-2'-(thiophene-3-carbonyl)dispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure C), obtained as a solid, (74% yield, 9:1 *d.r.*), mp 228-230 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.18 (d, *J* = 6.6 Hz, 1 H), 7.91 (d, *J* = 7.4 Hz, 1 H), 7.49 - 7.27 (m, 7 H), 7.21 (7.14 - 7.24 (m, 3 H), 7.13 (d, *J* = 7.6 Hz, 1 H), 7.08 - 6.99 (m, 5 H), 6.86 (s, 1 H), 6.84 - 6.73 (m, 4 H), 6.66 (d, *J* = 4.8 Hz, 1 H), 6.48 (dd, *J* = 7.4, 14.3 Hz, 2 H), 5.20 (d, *J* = 15.3 Hz, 2 H), 5.12 (s, 1 H), 4.54 (dd, *J* = 3.6, 15.6 Hz, 2 H), 4.36 (d, *J* = 13.9 Hz, 1 H), 2.64 (d, *J* = 14.0 Hz, 1 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 189.6, 184.0, 176.7, 143.8, 141.5, 141.3, 140.1, 135.8, 134.9, 130.2, 130.0, 129.0, 128.7, 128.6, 128.4, 128.2, 127.9, 127.1, 126.9, 126.5, 126.5, 126.2, 126.0, 125.9, 124.7, 124.3, 122.0, 121.8, 108.7, 108.5, 83.4, 66.3, 66.2, 54.9, 48.0, 44.7, 43.9; **HRMS** (ESI) m/z calculated for $C_{42}H_{32}N_2O_4S_2$+H 693.1882 found: 693.1876



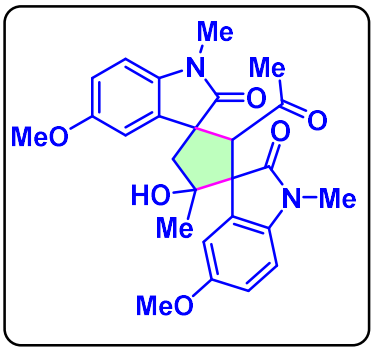
[4Ba] **2'-acetyl-4'-hydroxy-1,1'',4'-trimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (82% yield, 97:3 *d.r.*), mp 226-228 °C, **FT-IR** (KBr, cm^{-1}) 3371, 2931, 1704, 1609, 1349, 1049, 751; **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.07 (d, *J* = 7.5 Hz, 1 H), 7.47 (d, *J* = 7.3 Hz, 1 H), 7.32 - 7.17 (m, 2 H), 7.10 (t, *J* = 7.6 Hz, 1 H), 6.97 (t, *J* = 7.4 Hz, 1 H), 6.89 - 6.74 (m, 2 H), 6.00 (s, 1 H), 4.25 (s, 1 H), 3.37 (d, *J* = 14.0 Hz, 1 H), 3.29 (d, *J* = 18.9 Hz, 6 H), 2.17 (d, *J* = 14.1 Hz, 1 H), 1.15 (s, 3 H), 0.91 (s, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 201.7, 183.7, 178.0, 144.5, 142.8, 131.0, 128.8, 128.4, 127.1, 126.8, 125.7, 124.7, 122.0, 108.3, 107.6, 82.2, 69.9, 65.0, 54.2, 49.4, 28.2, 27.0, 26.3, 19.7; **HRMS** (ESI) m/z calculated for $C_{24}H_{24}N_2O_4$+H 405.1814 found: 405.1809



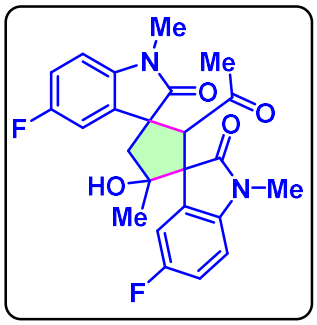
[4Bb] **2'-acetyl-4'-hydroxy-1,1'',4',5,5''-pentamethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (87% yield, 97:3 *d.r.*), mp 250-250 °C, **FT-IR** (KBr, cm^{-1}) 3381, 2985, 1703, 1676, 1485, 1356, 816; **^{1}H NMR** (400MHz , $CDCl_3$) δ = 7.88 (s, 1 H), 7.27 (s, 1 H), 7.03 (dd, *J* = 7.5, 15.6 Hz, 2 H), 6.73 (d, *J* = 7.9 Hz, 1 H), 6.68 (d, *J* = 7.8 Hz, 1 H), 6.05 (br. s., 1 H), 4.22 (s, 1 H), 3.34 (d, *J* = 14.0 Hz, 1 H), 3.29 (s, 3 H), 3.25 (s, 3 H), 2.29 (s, 3 H), 2.26 (s, 3 H), 2.14 (d, *J* = 14.0 Hz, 1 H), 1.17 (s, 3 H), 0.91 (s, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 201.8, 183.7, 177.9, 142.1, 140.4, 134.5, 131.3, 131.1, 129.1, 128.7, 127.3, 127.1, 126.6, 108.1, 107.2, 82.1, 69.9, 65.1, 54.3, 49.5, 28.2, 27.0, 26.4, 21.4, 21.2, 19.8; **HRMS** (ESI) m/z calculated for $C_{26}H_{28}N_2O_4$+H 433.2127 found: 433.2122



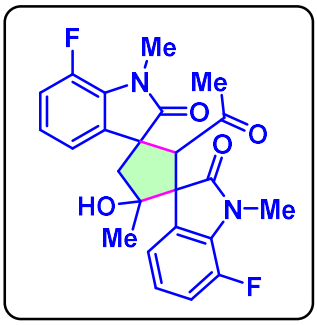
[4Bc] **2'-acetyl-4'-hydroxy-1,1'',4',5,5'',7,7''-heptamethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (83% yield, 95:5 *d.r.*) mp 280-282 °C, **FT-IR** (KBr, cm^{-1}) 3365, 2929, 1715, 1480, 1357, 751; **^{1}H NMR** (400MHz , $CDCl_3$) δ = 7.78 (br. s., 1 H), 7.11 (br. s., 1 H), 6.76 (d, *J* = 16.3 Hz, 2 H), 6.17 (br. s., 1 H), 4.17 (br. s., 1 H), 3.54 (d, *J* = 14.6 Hz, 6 H), 3.31 (d, *J* = 13.9 Hz, 1 H), 2.49 (br. s., 6 H), 2.21 (d, *J* = 12.3 Hz, 6 H), 2.11 (d, *J* = 13.9 Hz, 1 H), 1.19 (br. s., 3 H), 0.90 (br. s., 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 202.0, 184.4, 178.7, 139.8, 138.1, 134.2, 133.1, 132.8, 132.0, 130.9, 127.7, 125.1, 124.5, 119.5, 118.5, 82.1, 70.6, 64.4, 53.8, 50.1, 30.5, 29.7, 28.4, 21.1, 21.0, 19.8, 19.1, 18.8; **HRMS** (ESI) m/z calculated for $C_{28}H_{32}N_2O_4$+H 461.2440 found: 433.2435



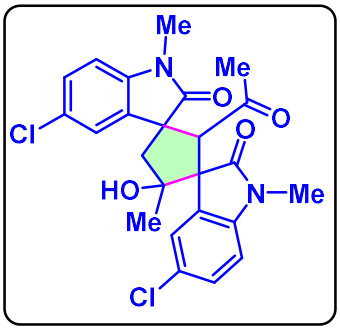
[4Bd] **2'-acetyl-4'-hydroxy-5,5''-dimethoxy-1,1'',4'-trimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (81% yield, 95:5 *d.r.*) mp 228-230 °C, **^{1}H NMR** (400MHz, $CDCl_3$) δ = 7.84 (s, 1 H), 7.10 (d, *J* = 2.1 Hz, 1 H), 6.84 - 6.62 (m, 4 H), 6.01 (br. s., 1 H), 4.21 (s, 1 H), 3.73 (d, *J* = 7.0 Hz, 6 H), 3.29 - 3.23 (m, 12 H), 2.16 (d, *J* = 14.0 Hz, 1 H), 1.19 (s, 3 H), 0.92 (s, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 201.5, 183.2, 177.5, 157.4, 155.3, 138.1, 136.1, 132.2, 128.4, 114.3, 113.2, 113.0, 108.8, 107.7, 82.1, 70.0, 65.3, 55.8, 55.8, 54.6, 49.5, 28.2, 27.1, 26.5, 19.7; **HRMS** (ESI) m/z calculated for $C_{26}H_{28}N_2O_6$+H 465.2026 found: 465.2020



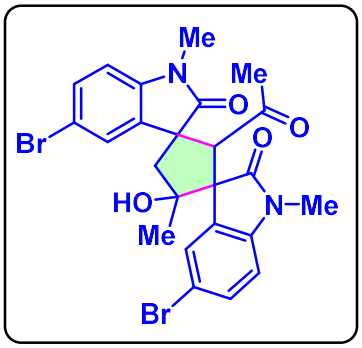
[4Be] **2'-acetyl-5,5''-difluoro-4'-hydroxy-1,1'',4'-trimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (84% yield, 97:3 *d.r.*) mp 260-262 °C, **FT-IR** (KBr, cm^{-1}) 3347, 3080, 1715, 1685, 1487, 1392, 686; **^{1}H NMR** (400MHz , $CDCl_3$) δ = 7.93 (dd, *J* = 2.6, 8.5 Hz, 1 H), 7.23 (dd, *J* = 2.6, 8.5 Hz, 1 H), 7.00 - 6.89 (m, 2 H), 6.78 (dd, *J* = 4.1, 8.5 Hz, 1 H), 6.71 (dd, *J* = 4.1, 8.5 Hz, 1 H), 5.92 (s, 1 H), 4.19 (s, 1 H), 3.34 (s, 1 H), 3.31 (s, 3 H), 3.25 (s, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 201.2, 183.2, 177.5, 161.5, 159.9, 159.0, 157.6, 140.4, 138.8, 132.6, 132.5, 128.6, 128.5, 115.5, 115.3, 115.0, 114.8, 114.6, 114.1, 113.9, 109.0, 108.9, 108.0, 107.9, 82.1, 69.9, 65.3, 54.4, 49.3, 28.2, 27.2, 26.5, 19.7; **^{19}F NMR** (376MHz, $CDCl_3$) δ -116.43, -120.99; **HRMS** (ESI) m/z calculated for $C_{24}H_{22}F_2N_2O_4$+H 441.1626 found:441.1620



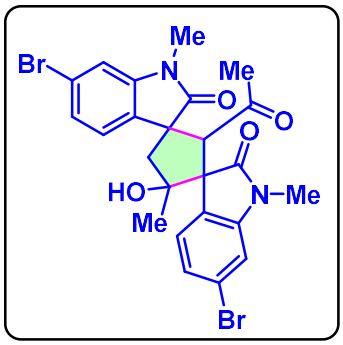
[4Bf] **2'-acetyl-7,7''-difluoro-4'-hydroxy-1,1'',4'-trimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (80% yield, 95:5 *d.r.*) mp 208-210 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 7.87 (dd, *J* = 1.4, 7.1 Hz, 1 H), 7.25 (dd, *J* = 0.8, 7.3 Hz, 1 H), 7.07 - 6.85 (m, 4 H), 5.92 (s, 1 H), 4.22 (s, 1 H), 3.52 (d, *J* = 2.8 Hz, 3 H), 3.47 (d, *J* = 2.6 Hz, 3 H), 3.34 (d, *J* = 14.1 Hz, 1 H), 2.18 (d, *J* = 14.1 Hz, 1 H), 1.22 (s, 3 H), 0.92 (s, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 201.3, 183.3, 177.6, 148.9, 148.6, 146.5, 146.2, 133.5, 133.5, 131.0, 131.0, 129.8, 129.8, 129.4, 129.3, 125.3, 125.3, 122.6, 122.5, 122.4, 122.3, 121.6, 121.6, 117.0, 116.8, 116.6, 116.4, 82.2, 70.4, 65.2, 65.2, 54.4, 49.5, 29.8, 29.7, 28.9, 28.8, 28.2, 19.8; **^{19}F NMR** (376 MHz, $CDCl_3$) δ -136.48, -137.25; **HRMS** (ESI) m/z calculated for $C_{24}H_{22}F_2N_2O_4$+H 441.1626 found: 441.1620



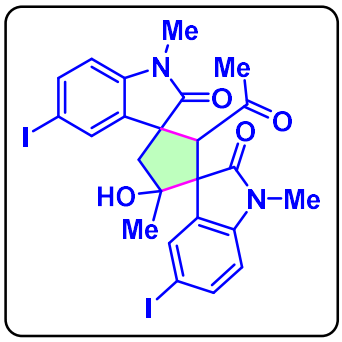
[4Bg] **2'-acetyl-5,5''-dichloro-4'-hydroxy-1,1'',4'-trimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (75% yield, 90:10 *d.r.*) mp 220-222 °C; **^{1}H NMR** (400MHz , $CDCl_3$) δ = 7.96 (d, *J* = 8.1 Hz, 1 H), 7.38 (d, *J* = 8.0 Hz, 1 H), 7.06 (d, *J* = 8.1 Hz, 1 H), 6.95 (d, *J* = 7.5 Hz, 1 H), 6.88 - 6.76 (m, 2 H), 5.80 (s, 1 H), 4.19 (s, 1 H), 3.30 (s, 3 H), 3.27 (br. s., 1 H), 3.24 (s, 3 H), 2.15 (d, *J* = 14.1 Hz, 1 H), 1.20 (s, 3 H), 0.89 (s, 3 H); **^{13}C NMR** (101MHz, $CDCl_3$) δ = 201.3, 183.6, 177.8, 145.6, 144.0, 134.9, 134.4, 129.1, 127.8, 126.6, 125.3, 124.5, 122.0, 109.3, 108.4, 82.1, 69.9, 64.7, 53.8, 49.3, 28.3, 27.2, 26.5, 19.7; **HRMS** (ESI) m/z calculated for $C_{24}H_{22}Cl_2N_2O_4$+H 473.1035 found: 473.1039



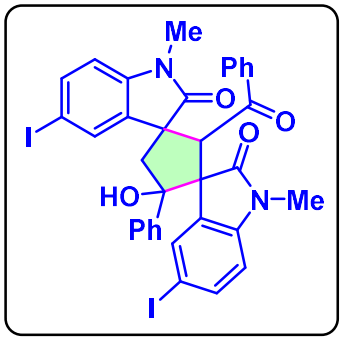
[4Bh] **2'-acetyl-5,5''-dibromo-4'-hydroxy-1,1'',4'-trimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (84% yield, 95:5 *d.r.*) mp 298-300 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.23 (d, *J* = 1.8 Hz, 1 H), 7.58 (d, *J* = 1.8 Hz, 1 H), 7.42 (dd, *J* = 1.5, 8.1 Hz, 1 H), 7.35 (dd, *J* = 1.8, 8.2 Hz, 1 H), 6.78 - 6.63 (m, 2 H), 5.83 (s, 1 H), 4.18 (s, 1 H), 3.30 (s, 3 H), 3.28 (d, *J* = 14.0 Hz, 1 H), 3.25 (s, 3 H), 2.15 (d, *J* = 14.1 Hz, 3 H), 1.23 (s, 8 H), 0.91 (s, 8 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 201.0, 183.0, 177.2, 143.6, 141.9, 132.8, 132.0, 131.3, 129.9, 128.9, 128.9, 117.6, 114.7, 109.8, 109.0, 82.1, 70.0, 65.0, 54.1, 49.3, 28.2, 27.2, 26.5, 19.7; **HRMS** (ESI) m/z calculated for $C_{24}H_{22}Br_2N_2O_4$+H 561.0025 found: 561.0019



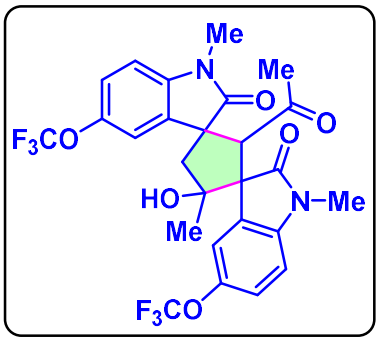
[4Bi] **2'-acetyl-6,6''-dibromo-4'-hydroxy-1,1'',4'-trimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (87% yield, 97:3 *d.r.*) mp 222-224 °C, **FT-IR** (KBr, cm^{-1}) 3334, 2930, 1710, 1602, 1490, 1368, 730; **^{1}H NMR** (400MHz , $CDCl_3$) δ = 7.98 (d, *J* = 8.0 Hz, 1 H), 7.39 (d, *J* = 8.0 Hz, 1 H), 7.29 (d, *J* = 8.3 Hz, 1 H), 7.18 (d, *J* = 9.0 Hz, 1 H), 7.11 - 6.97 (m, 2 H), 5.87 (s, 1 H), 4.26 (s, 1 H), 3.36 (s, 3 H), 3.34 (s, 1 H), 3.31 (s, 3 H), 2.22 (d, *J* = 14.3 Hz, 1 H), 1.27 (s, 3 H), 0.96 (s, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 201.3, 183.5, 177.7, 145.7, 144.1, 129.6, 128.1, 127.5, 127.0, 125.8, 124.9, 122.6, 122.3, 112.0, 111.2, 82.1, 69.8, 64.8, 53.8, 49.2, 28.3, 27.2, 26.5, 19.7; **HRMS** (ESI) m/z calculated for $C_{24}H_{22}Br_2N_2O_4$+H 561.0025 found: 561.0021



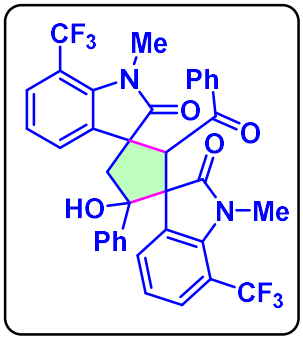
[4Bj] **2'-acetyl-4'-hydroxy-5,5''-diiodo-1,1'',4'-trimethyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (81% yield, 93:7 *d.r.*) mp 280-282 °C **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.37 (br. s., 1 H), 7.72 (br. s., 1 H), 7.62 (d, *J* = 6.9 Hz, 1 H), 7.54 (d, *J* = 7.0 Hz, 1 H), 6.68 - 6.52 (m, 2 H), 5.80 (br. s., 1 H), 4.16 (br. s., 1 H), 3.29 (br. s., 3 H), 3.24 (br. s., 3 H), 3.16 (d, *J* = 12.4 Hz, 1 H), 2.13 (d, *J* = 15.9 Hz, 1 H), 1.22 (br. s., 3 H), 0.90 (br. s., 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 201.0, 182.9, 177.0, 144.3, 142.6, 138.1, 137.3, 135.4, 134.4, 133.1, 132.2, 129.2, 110.4, 109.6, 87.6, 84.7, 82.1, 70.0, 64.9, 53.9, 49.3, 28.2, 27.1, 26.5, 19.7; **HRMS** (ESI) m/z calculated for $C_{24}H_{22}I_2N_2O_4$+H 656.9747 found: 656.9742



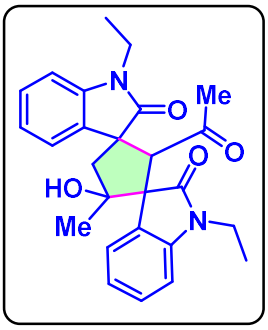
[4Bk] **2'-benzoyl-4'-hydroxy-5,5''-diiodo-1,1''-dimethyl-4'-phenyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (90% yield, 98:2 *d.r.*) mp 248-250 °C, **FT-IR** (KBr, cm^{-1}) 3293, 3059, 2926, 1703, 1603, 1488, 1347, 756; **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.55 (d, *J* = 1.1 Hz, 1 H), 8.11 (d, *J* = 1.0 Hz, 1 H), 7.58 (dd, *J* = 1.3, 8.1 Hz, 1 H), 7.37 (td, *J* = 0.9, 8.2 Hz, 1 H), 7.32 - 7.27 (m, 1 H), 7.21 - 6.98 (m, 9 H), 6.75 (s, 1 H), 6.39 (d, *J* = 8.1 Hz, 1 H), 6.05 (d, *J* = 8.1 Hz, 1 H), 5.10 (s, 1 H), 4.27 (d, *J* = 14.0 Hz, 1 H), 3.08 (s, 3 H), 2.87 (s, 3 H), 2.41 (d, *J* = 14.0 Hz, 1 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 196.1, 182.8, 175.7, 144.2, 142.1, 137.5, 137.2, 137.1, 135.4, 134.0, 132.5, 132.3, 128.8, 127.8, 127.8, 127.3, 127.0, 125.5, 109.5, 109.4, 87.0, 84.5, 84.2, 66.9, 64.7, 54.1, 45.3, 26.7, 25.9; **HRMS** (ESI) m/z calculated for $C_{34}H_{26}I_2N_2O_4$+H 779.9982 found: 779.9938



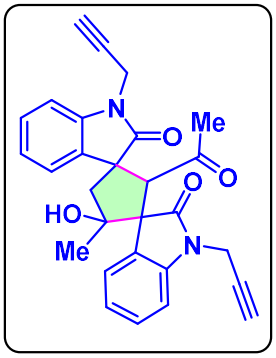
[4Bl] **2'-acetyl-4'-hydroxy-1,1'',4'-trimethyl-5,5''-bis(trifluoromethoxy)dispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (75% yield, 95:5 *d.r.*) mp 198-200 °C, **FT-IR** (KBr, cm^{-1}) 3345, 3072, 1706, 1619, 1497, 1253, 810; **^{1}H NMR** (400MHz, $CDCl_3$) δ = 8.06 (br. s., 1 H), 7.35 (br. s., 1 H), 7.18 - 7.07 (m, 2 H), 6.85 (d, *J* = 8.4 Hz, 1 H), 6.76 (d, *J* = 8.4 Hz, 1 H), 5.82 (s, 1 H), 4.20 (s, 1 H), 3.37 - 3.31 (m, 1 H), 3.33 (s, 3 H), 3.27 (s, 3 H), 2.18 (d, *J* = 14.1 Hz, 1 H), 1.20 (br. s., 3 H), 0.91 (s, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 200.9, 183.3, 177.5, 144.2, 143.2, 141.4, 132.4, 128.4, 121.9, 121.5, 120.9, 119.8, 108.9, 107.9, 82.2, 69.9, 65.2, 54.2, 49.3, 29.7, 28.1, 27.3, 26.6, 19.7; **^{19}F NMR** ($CDCl_3$) δ -58.15, -58.20; **HRMS** (ESI) m/z calculated for $C_{26}H_{22}F_6N_2O_6$+H 573.1460 found: 573.1455



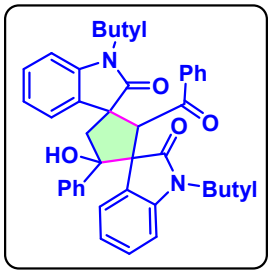
[4Bm] **2'-benzoyl-4'-hydroxy-1,1''-dimethyl-4'-phenyl-4,7''-bis(trifluoromethyl)dispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione,** (General Procedure B), obtained as a solid, (85% yield, 94:6 *d.r.*) mp 288-290 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.61 (d, *J* = 7.6 Hz, 1 H), 8.05 (d, *J* = 7.4 Hz, 1 H), 7.59 (d, *J* = 8.1 Hz, 1 H), 7.39 - 7.28 (m, 2 H), 7.23 - 7.15 (m, 3 H), 7.14 - 7.08 (m, 4 H), 7.07 - 7.02 (m, 2 H), 6.96 (d, *J* = 7.6 Hz, 2 H), 6.88 (s, 1 H), 5.21 (s, 1 H), 4.33 (d, *J* = 14.0 Hz, 1 H), 3.37 (br. s., 3 H), 3.04 (br. s., 3 H), 2.48 (d, *J* = 14.0 Hz, 1 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 195.9, 184.7, 177.5, 142.3, 140.1, 136.8, 136.6, 132.9, 132.7, 130.4, 128.7, 128.7, 128.1, 127.9, 127.3, 126.7, 125.2, 123.7, 121.0, 84.9, 65.6, 65.1, 52.8, 45.5; **^{19}F NMR** (377 MHz, $CDCl_3$) δ -53.23, -53.41; **HRMS** (ESI) m/z calculated for $C_{36}H_{26}F_6N_2O_4$+H 664.1797 found: 664.1790



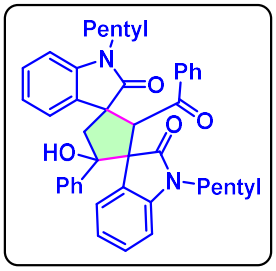
[4Bp] **2'-acetyl-1,1''-diethyl-4'-hydroxy-4'-methyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure C), obtained as a solid, (74% yield, 90:10 *d.r.*) mp 106-108 °C, **^{1}H NMR** (400MHz, $CDCl_3$) δ = 8.08 (d, *J* = 7.1 Hz, 1 H), 7.48 (d, *J* = 7.0 Hz, 1 H), 7.28 - 7.20 (m, 2 H), 7.08 (t, *J* = 7.4 Hz, 1 H), 6.96 (t, *J* = 7.3 Hz, 1 H), 6.85 (d, *J* = 7.8 Hz, 1 H), 6.80 (d, *J* = 7.8 Hz, 1 H), 6.02 (s, 1 H), 4.26 (s, 1 H), 3.96 - 3.72 (m, 4 H), 3.37 (d, *J* = 14.0 Hz, 1 H), 2.16 (d, *J* = 14.0 Hz, 1 H), 1.30 (dt, *J* = 3.9, 7.2 Hz, 6 H), 1.16 (s, 3 H), 0.91 (s, 3 H); **^{13}C NMR** (101MHz, $CDCl_3$) δ = 201.6, 183.3, 177.4, 143.4, 141.8, 131.2, 128.7, 128.3, 127.3, 127.0, 125.9, 124.5, 121.8, 108.4, 107.6, 82.2, 69.8, 64.9, 54.1, 49.4, 35.5, 34.6, 28.1, 19.6, 12.6, 12.1; **HRMS** (ESI) m/z calculated for $C_{26}H_{28}N_2O_4$+H 433.2127 found: 433.2130



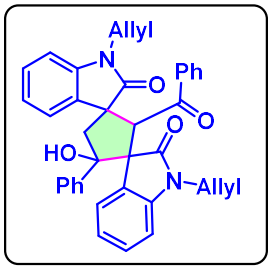
[4Bq] **2'-acetyl-4'-hydroxy-4'-methyl-1,1''-di(prop-2-yn-1-yl)dispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure C), obtained as a solid, (72% yield, 90:10 *d.r.*) mp 110-112 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.07 (d, *J* = 7.6 Hz, 1 H), 7.49 (d, *J* = 7.4 Hz, 1 H), 7.27 (td, *J* = 7.7, 12.9 Hz, 2 H), 7.16 - 7.08 (m, 1 H), 7.06 - 6.96 (m, 3 H), 5.82 (s, 1 H), 4.82 - 4.64 (m, 2 H), 4.43 (ddd, *J* = 2.4, 11.4, 17.6 Hz, 2 H), 4.27 (s, 1 H), 3.37 (d, *J* = 14.1 Hz, 1 H), 2.25 - 2.20 (m, 2 H), 2.19 (d, *J* = 3.4 Hz, 1 H), 1.18 (s, 3 H), 0.93 (s, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 201.6, 182.9, 177.0, 142.4, 140.9, 130.6, 128.8, 128.5, 126.9, 126.8, 125.9, 125.0, 122.4, 109.3, 108.5, 82.4, 76.0, 72.9, 71.9, 70.1, 64.9, 54.0, 49.3, 30.0, 29.3, 28.1, 19.6; **HRMS** (ESI) m/z calculated for $C_{26}H_{24}N_2O_4$+H 453.1814 found: 453.1809



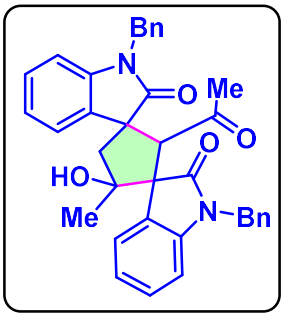
[4Br] 2'-benzoyl-1,1''-dibutyl-4'-hydroxy-4'-phenyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione, (General Procedure C), obtained as a solid, (70% yield, 90:10 *d.r.*) mp 188-190 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.21 (dd, *J* = 1.3, 7.1 Hz, 1 H), 7.89 (d, *J* = 7.3 Hz, 1 H), 7.33 - 7.20 (m, 3 H), 7.12 (d, *J* = 7.4 Hz, 4 H), 7.06 (d, *J* = 7.4 Hz, 5 H), 7.04 - 6.98 (m, 3 H), 6.94 (s, 1 H), 6.64 (d, *J* = 7.6 Hz, 1 H), 6.34 (d, *J* = 6.9 Hz, 1 H), 5.26 (s, 1 H), 4.41 (d, *J* = 13.9 Hz, 1 H), 3.92 - 3.68 (m, 2 H), 3.38 - 3.11 (m, 2 H), 2.45 (d, *J* = 13.9 Hz, 1 H), 1.56 (tt, *J* = 7.1, 14.6 Hz, 2 H), 1.44 - 1.30 (m, 2 H), 0.99 - 0.94 (m, 3 H), 0.82 - 0.75 (m, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 196.4, 183.5, 176.3, 144.1, 141.8, 138.0, 137.2, 132.0, 130.5, 128.5, 128.2, 127.8, 127.5, 127.3, 127.1, 127.0, 126.8, 125.8, 125.6, 123.9, 121.5, 107.8, 107.6, 84.5, 66.4, 64.4, 54.1, 46.2, 40.4, 39.7, 29.3, 28.9, 20.2, 20.0, 13.9, 13.6; **HRMS** (ESI) m/z calculated for $C_{40}H_{40}N_2O_4$+H 613.3066 found: 613.3061



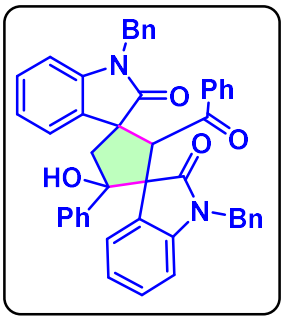
[4Bs] 2'-benzoyl-4'-hydroxy-1,1''-dipentyl-4'-phenyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione, (General Procedure C), obtained as a solid, (77% yield, 90:10 *d.r.*) mp 110-112 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.21 (dd, *J* = 2.4, 6.4 Hz, 1 H), 7.89 (d, *J* = 7.5 Hz, 1 H), 7.25 - 7.21 (m, 1 H), 7.16 - 6.99 (m, 12 H), 6.94 (s, 1 H), 6.64 (d, *J* = 7.8 Hz, 1 H), 6.39 - 6.28 (m, 1 H), 5.31 - 5.20 (m, 1 H), 4.41 (d, *J* = 13.6 Hz, 1 H), 3.86 - 3.70 (m, 2 H), 3.38 - 3.13 (m, 2 H), 2.45 (d, *J* = 13.9 Hz, 1 H), 1.64 - 1.46 (m, 2 H), 1.42 - 1.28 (m, 4 H), 1.25 - 1.16 (m, 2 H), 1.14 - 0.94 (m, 4 H), 0.91 (t, *J* = 6.9 Hz, 3 H), 0.86 - 0.81 (m, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 196.4, 183.4, 176.2, 144.0, 141.8, 138.0, 137.2, 132.0, 130.5, 128.5, 128.2, 127.8, 127.6, 127.3, 127.1, 127.0, 126.7, 125.8, 125.6, 123.9, 121.5, 107.8, 107.6, 84.5, 66.5, 64.3, 54.1, 46.1, 40.7, 39.9, 29.0, 28.9, 26.9, 26.5, 22.4, 22.3, 13.9, 13.8; **HRMS** (ESI) m/z calculated for $C_{42}H_{44}N_2O_4$+H 641.3379 found: 641.3374



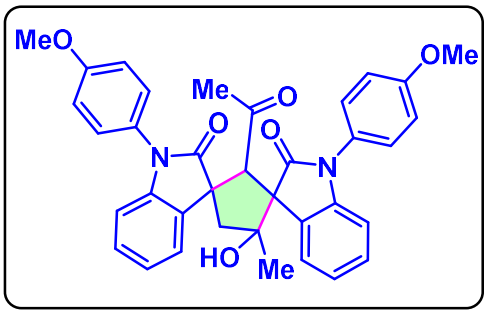
[4Bt] 1,1''-diallyl-2'-benzoyl-4'-hydroxy-4'-phenyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione, (General Procedure B), obtained as a solid, (80% yield, 96:4 *d.r.*), mp 215-217 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.16 (d, *J* = 7.3 Hz, 1 H), 7.83 (d, *J* = 7.4 Hz, 1 H), 7.24 - 7.13 (m, 2 H), 7.09 - 6.99 (m, 9 H), 6.98 - 6.89 (m, 2 H), 6.79 (s, 1 H), 6.53 (d, *J* = 7.6 Hz, 1 H), 6.29 (d, *J* = 7.6 Hz, 1 H), 5.65 (tdd, *J* = 5.6, 10.9, 16.6 Hz, 1 H), 5.22 (s, 1 H), 5.18 - 5.06 (m, 3 H), 4.78 (d, *J* = 10.4 Hz, 1 H), 4.50 (d, *J* = 17.3 Hz, 1 H), 4.42 (dd, *J* = 5.1, 16.0 Hz, 1 H), 4.37 - 4.26 (m, 2 H), 3.93 - 3.76 (m, 1 H), 2.41 (d, *J* = 14.0 Hz, 1 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 196.4, 183.4, 176.3, 143.7, 141.7, 138.0, 137.3, 132.2, 131.2, 130.6, 130.3, 128.6, 128.3, 127.9, 127.6, 127.5, 127.1, 126.5, 125.9, 125.6, 124.2, 121.8, 118.6, 116.6, 108.4, 108.4, 84.6, 66.8, 64.5, 54.2, 46.3, 43.2, 42.1; **HRMS** (ESI) m/z calculated for $C_{38}H_{32}N_2O_4$+H 581.2440 found: 581.2435



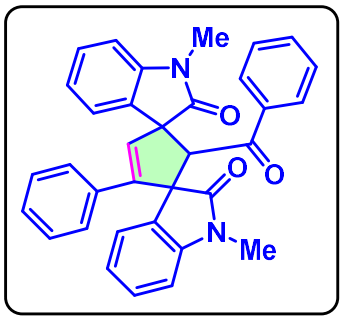
[4Bu] 2'-acetyl-1,1''-dibenzyl-4'-hydroxy-4'-methyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione, (General Procedure B), obtained as a solid, (82% yield, 93:7 *d.r.*), mp 120-122 °C **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.15 (d, *J* = 7.5 Hz, 1 H), 7.59 (d, *J* = 7.4 Hz, 1 H), 7.48 (d, *J* = 7.4 Hz, 2 H), 7.42 - 7.27 (m, 8 H), 7.22 (d, *J* = 7.9 Hz, 1 H), 7.19 - 7.09 (m, 2 H), 7.06 - 6.98 (m, 1 H), 6.88 (d, *J* = 7.6 Hz, 1 H), 6.71 (d, *J* = 7.8 Hz, 1 H), 6.11 (s, 1 H), 5.19 - 4.88 (m, 4 H), 4.43 (s, 1 H), 3.51 (d, *J* = 14.1 Hz, 1 H), 2.32 (d, *J* = 14.0 Hz, 21 H), 1.22 (s, 3 H), 1.05 (s, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 201.6, 183.9, 178.0, 143.7, 141.9, 136.2, 135.2, 131.0, 129.0, 128.7, 128.4, 128.2, 127.6, 127.4, 127.3, 127.0, 126.9, 125.8, 124.7, 122.0, 109.3, 108.7, 82.4, 69.9, 65.0, 54.1, 49.7, 44.6, 44.1, 28.3, 20.0; **HRMS** (ESI) m/z calculated for $C_{36}H_{32}N_2O_4$+H 557.2440 found: 557.2435



[4Bv] **2'-benzoyl-1,1''-dibenzyl-4'-hydroxy-4'-phenyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (76% yield, 90:10 *d.r.*), mp 223-225 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.14 (d, *J* = 7.3 Hz, 1 H), 7.90 (d, *J* = 7.1 Hz, 1 H), 7.22 (dd, *J* = 8.6, 18.9 Hz, 8 H), 7.12 - 6.97 (m, 10 H), 6.96 - 6.83 (m, 5 H), 6.46 (d, *J* = 6.5 Hz, 2 H), 6.32 (d, *J* = 7.0 Hz, 2 H), 6.23 (d, *J* = 7.5 Hz, 2 H), 5.31 (s, 1 H), 5.23 - 5.03 (m, 2 H), 4.51 - 4.30 (m, 2 H), 4.19 (d, *J* = 15.4 Hz, 1 H), 2.49 (d, *J* = 13.9 Hz, 1 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 196.4, 183.9, 176.8, 143.8, 141.6, 138.2, 137.1, 135.7, 134.9, 132.2, 130.3, 128.9, 128.6, 128.5, 128.3, 128.0, 127.8, 127.7, 127.6, 127.0, 126.8, 126.7, 126.4, 126.1, 125.7, 124.2, 121.9, 108.7, 108.5, 84.5, 66.6, 65.3, 54.3, 46.7, 44.5, 43.8; **HRMS** (ESI) m/z calculated for $C_{46}H_{36}N_2O_4$+H 681.2753 found: 681.2748



[4Bw] **2'-acetyl-4'-hydroxy-1,1''-bis(4-methoxyphenyl)-4'-methyldispiro[indoline-3,1'-cyclopentane-3',3''-indoline]-2,2''-dione**, (General Procedure B), obtained as a solid, (90% yield, 98:2 *d.r.*), mp 248-250 °C, **FT-IR** (KBr, cm^{-1}) 3325, 2925, 1714, 1610, 1513, 1250, 753; **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.18 (d, *J* = 7.5 Hz, 1 H), 7.59 (d, *J* = 7.4 Hz, 1 H), 7.48 (d, *J* = 8.6 Hz, 2 H), 7.37 (d, *J* = 8.8 Hz, 2 H), 7.25 - 7.17 (m, 2 H), 7.14 - 7.06 (m, 5 H), 6.84 - 6.66 (m, 2 H), 6.07 (s, 1 H), 4.50 (s, 1 H), 3.89 (d, *J* = 5.4 Hz, 6 H), 3.56 (d, *J* = 14.1 Hz, 1 H), 2.44 (d, *J* = 14.3 Hz, 1 H), 1.43 (s, 3 H), 1.18 (s, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 201.7, 183.8, 177.9, 159.8, 159.3, 145.1, 143.4, 130.6, 128.7, 128.5, 128.4, 127.8, 127.7, 127.6, 127.2, 126.8, 126.2, 126.0, 125.0, 122.3, 115.2, 115.1, 115.1, 109.5, 108.7, 82.5, 70.9, 65.1, 55.6, 55.6, 54.4, 49.8, 29.7, 28.3, 19.8; **HRMS** (ESI) m/z calculated for $C_{36}H_{32}N_2O_6$+H 589.2339 found: 589.2333



[5A] **2'-benzoyl-1,1''-dimethyl-4'-phenyldispiro[indoline-3,1'-cyclopentane-3',3''-indolin]-4'-ene-2,2''-dione**, (General Procedure D), obtained as a solid, mp 214-216 °C, **^{1}H NMR** (400MHz , $CDCl_3$) δ = 8.36 - 8.27 (m, 1 H), 7.51 (d, *J* = 7.3 Hz, 1 H), 7.25 - 7.21 (m, 1 H), 7.18 - 7.13 (m, 1 H), 7.06 - 7.01 (m, 3 H), 7.01 - 6.97 (m, 4 H), 6.95 (t, *J* = 7.1 Hz, 5 H), 6.79 (d, *J* = 7.8 Hz, 1 H), 6.23 (d, *J* = 8.8 Hz, 1 H), 5.86 (s, 1 H), 5.14 (s, 1 H), 3.36 (s, 3 H), 2.87 (s, 3 H); **^{13}C NMR** (101MHz , $CDCl_3$) δ = 197.6, 178.5, 177.7, 146.6, 144.2, 142.5, 137.5, 133.5, 131.6, 131.1, 130.8, 129.0, 128.6, 128.2, 128.2, 127.9, 127.4, 127.3, 127.1, 126.7, 123.9, 123.6, 122.8, 108.1, 107.2, 68.5, 63.0, 62.1, 26.9, 26.5; **HRMS** (ESI) m/z calculated for $C_{34}H_{26}N_2O_3$+H 511.2022 found: 511.2016

10. References:

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2. G. M. Sheldrick, *Acta Crystallogr., Sect. A: Found. Adv.*, 2015, **71**, 3.
3. S. J. Edeson, J. L. Jiang, S. Swanson, P. A. Procopiou, Adams, H.; Meijer, A.; Harrity, J. Studies on the stereochemical assignment of 3-acylidene 2-oxindoles. *Org. Biomol. Chem.* 2014, **12**, 3201.
4. P. Tehri, R. K. Peddinti, DBU-catalyzed [3+2] cycloaddition and Michael addition reactions of 3-benzylidenesuccinimides with 3-ylidene oxindoles and chalcones. *Org. Biomol.Chem.* 2019, **17**, 3964.
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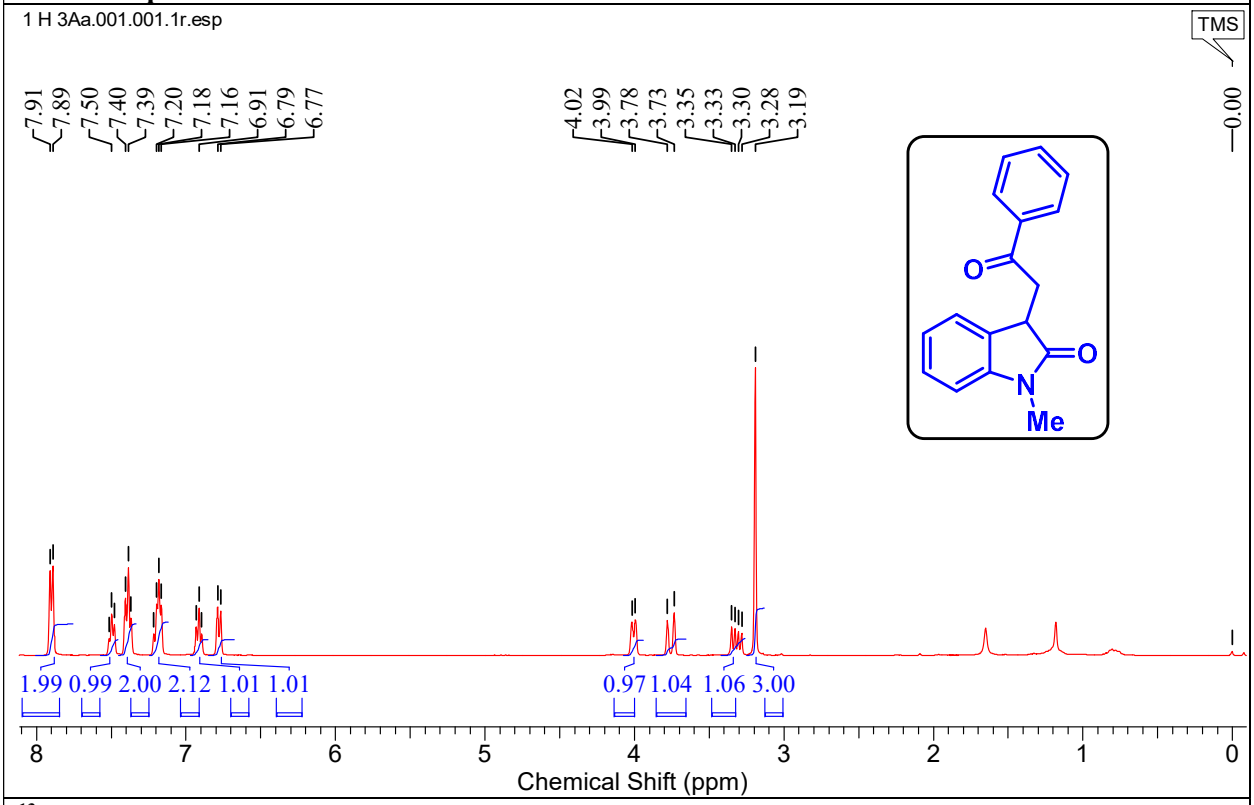
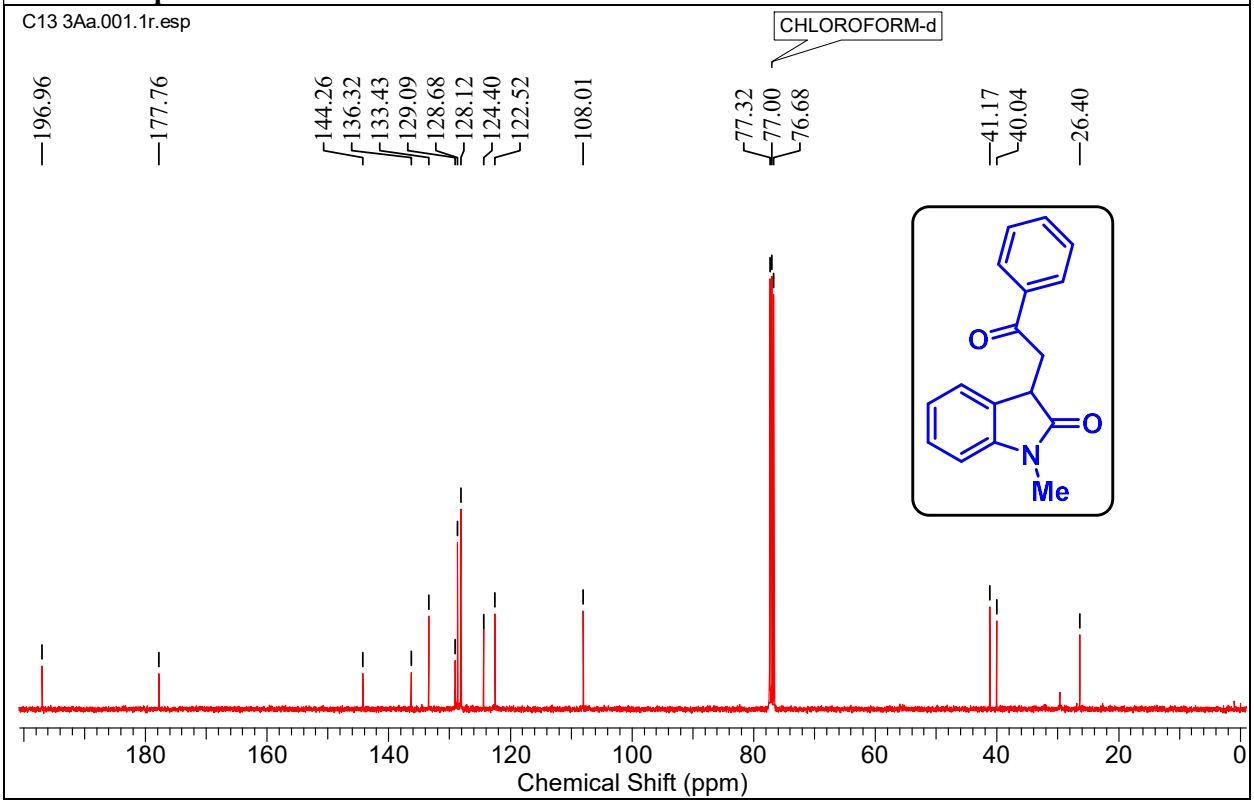
6. X. C. Zhang, S. H. Cao, Y. Wei, M. Shi, *Chem. Commun*. 2011, **47**, 1548.

11. NMR Spectra of Compounds

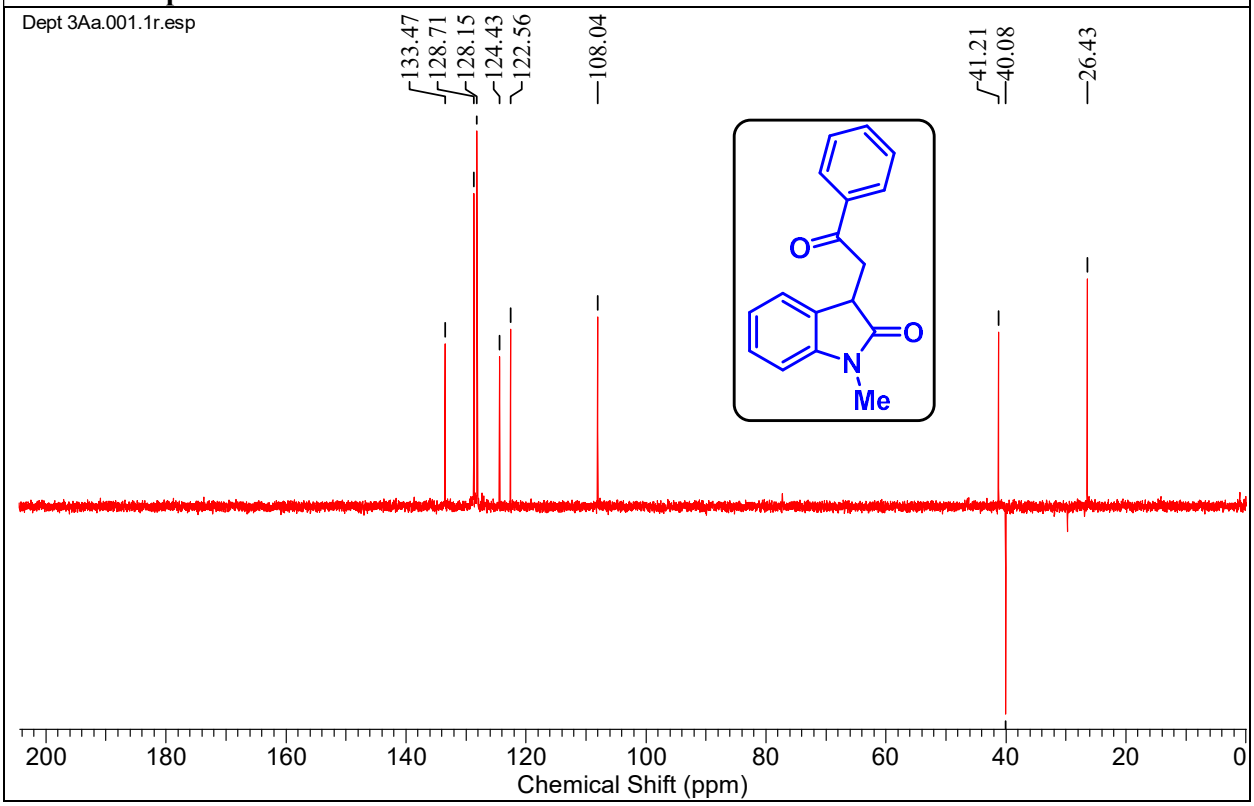
^{1}H NMR spectrum of 3Aa



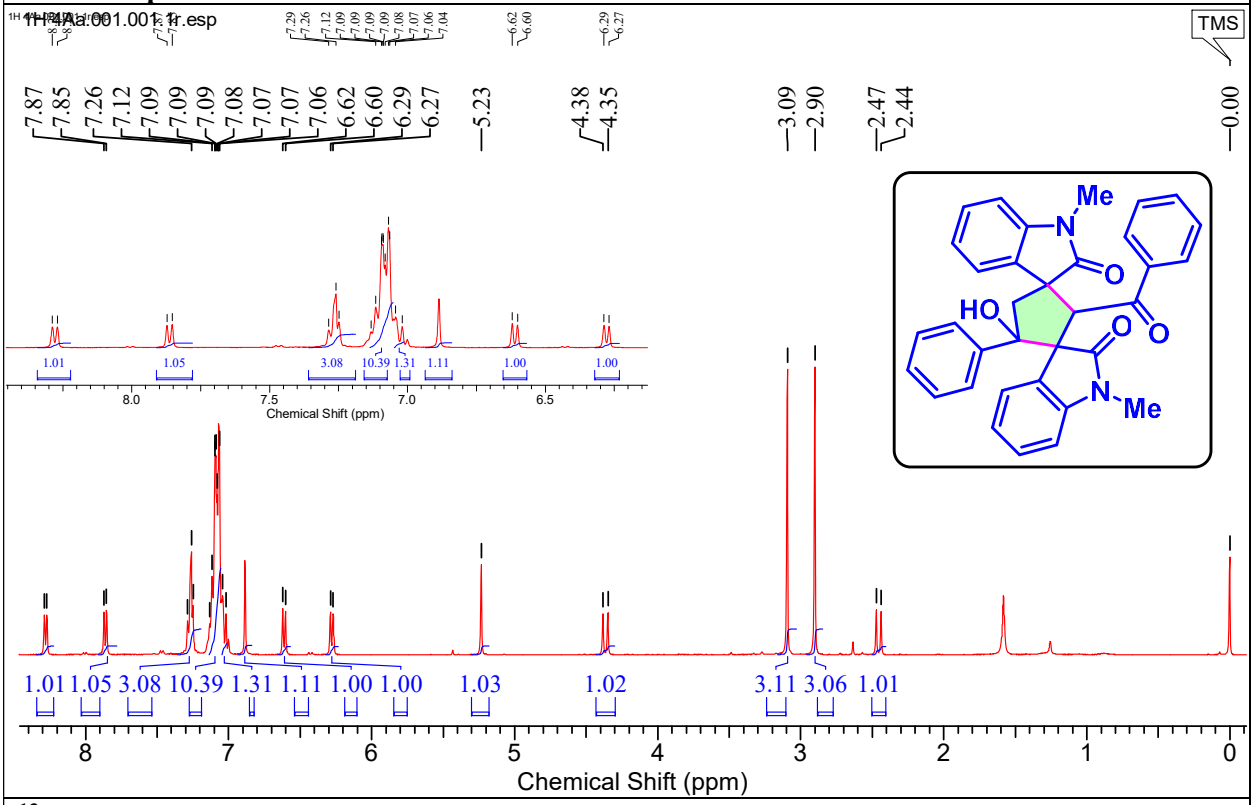
^{13}C NMR spectrum of 3Aa



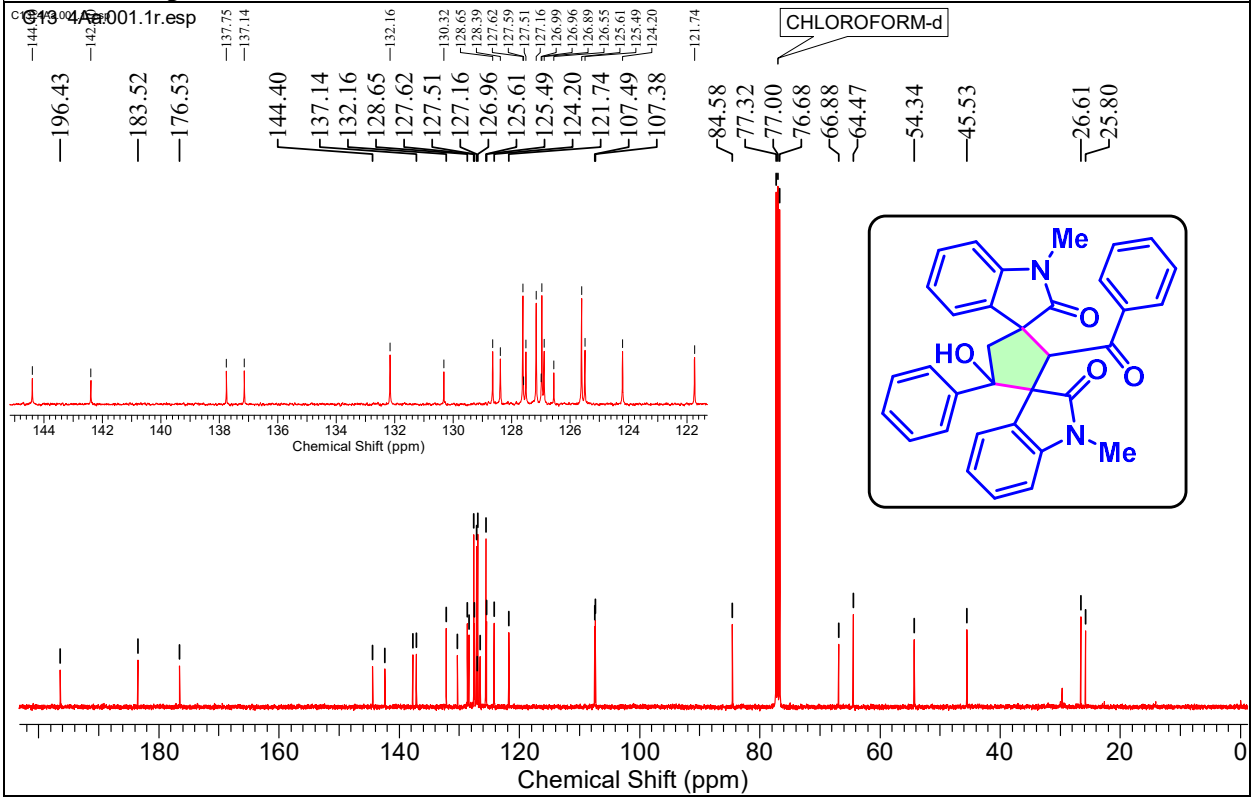
135 DEPT spectrum of 3Aa



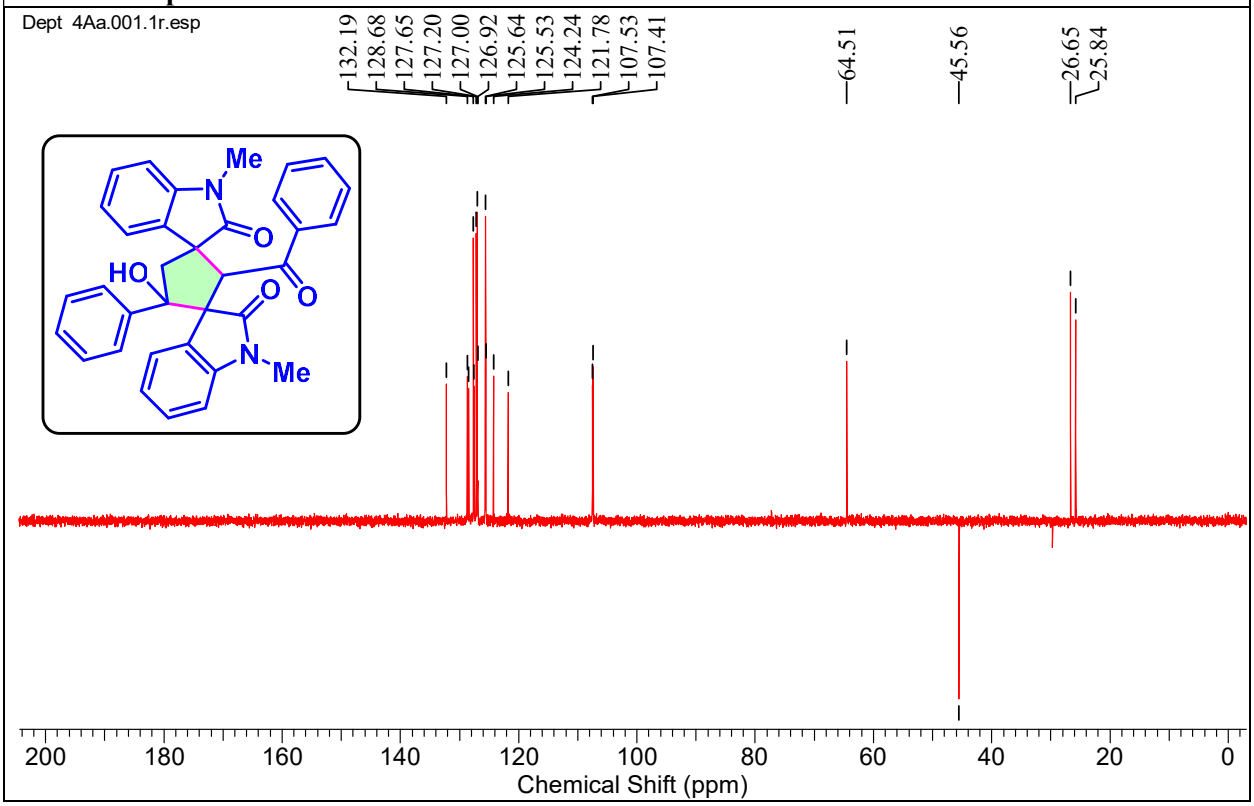
^{1}H NMR spectrum of 4Aa



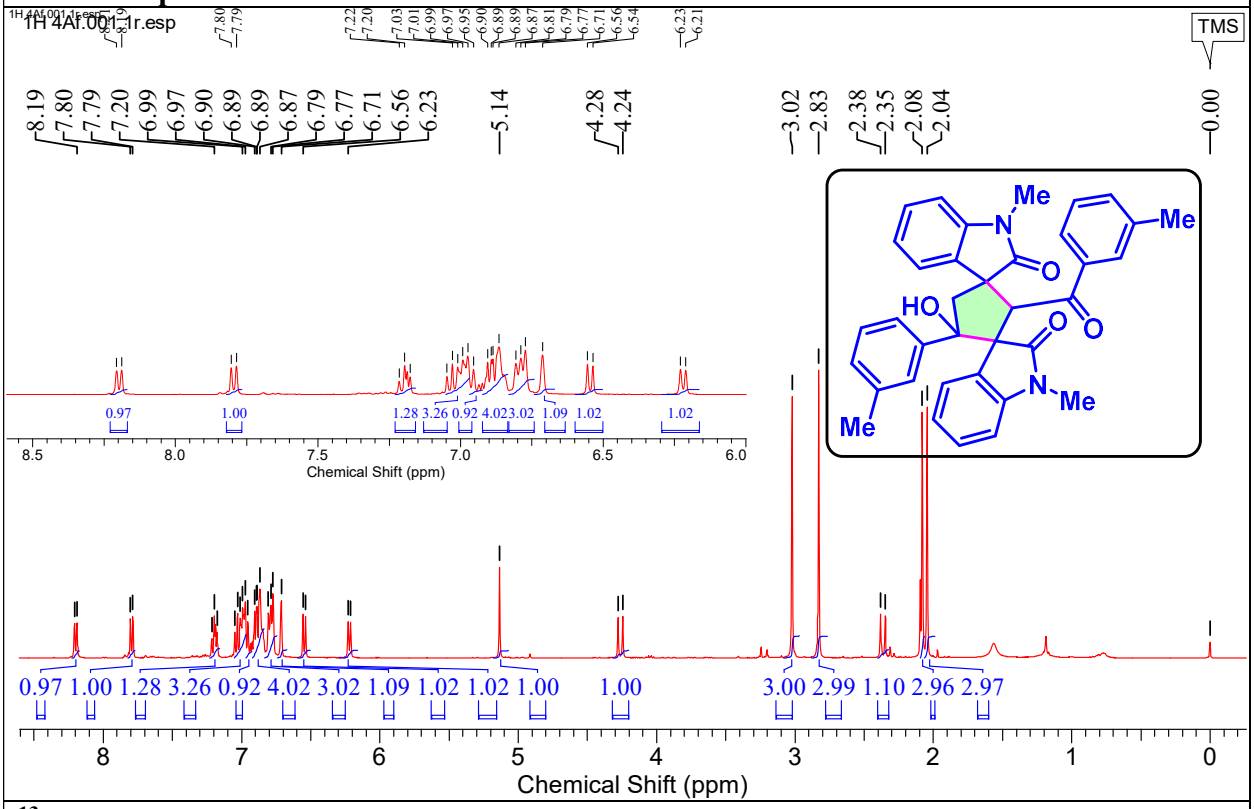
^{13}C NMR spectrum of 4Aa



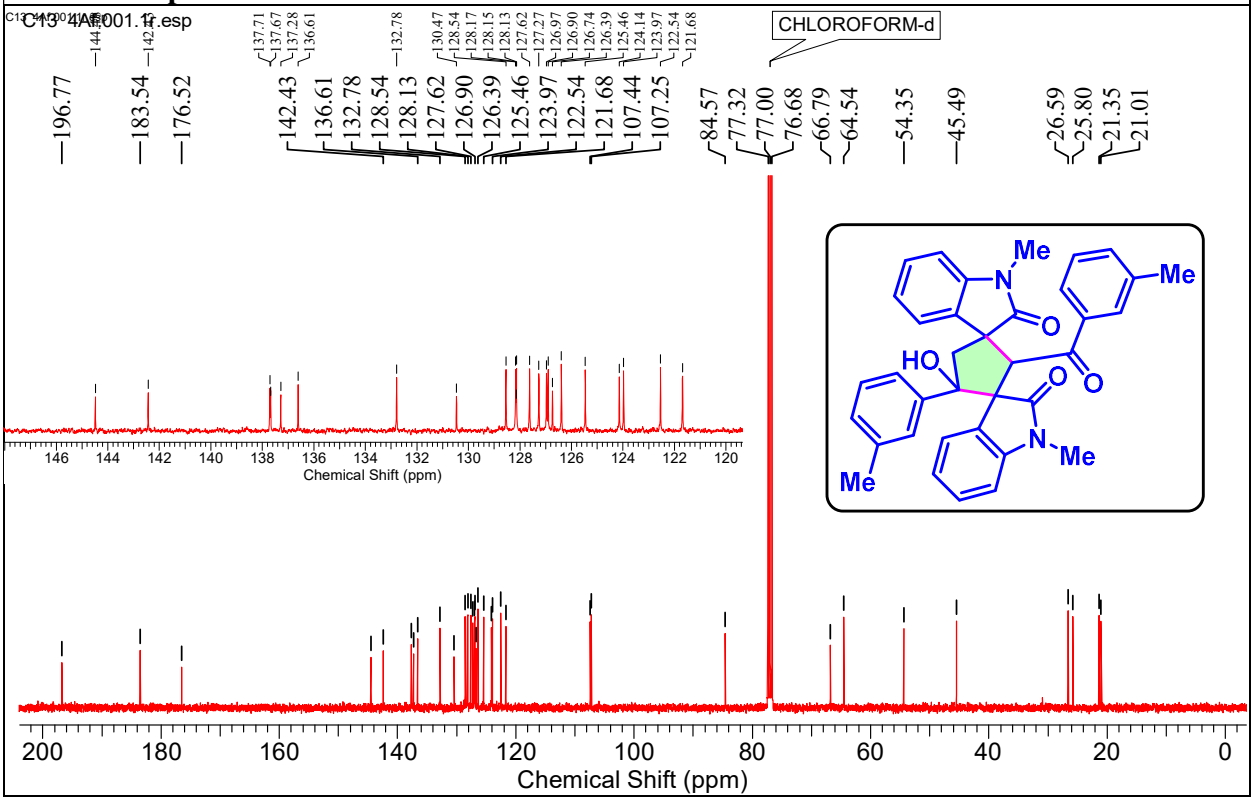
135 DEPT spectrum of 4Aa



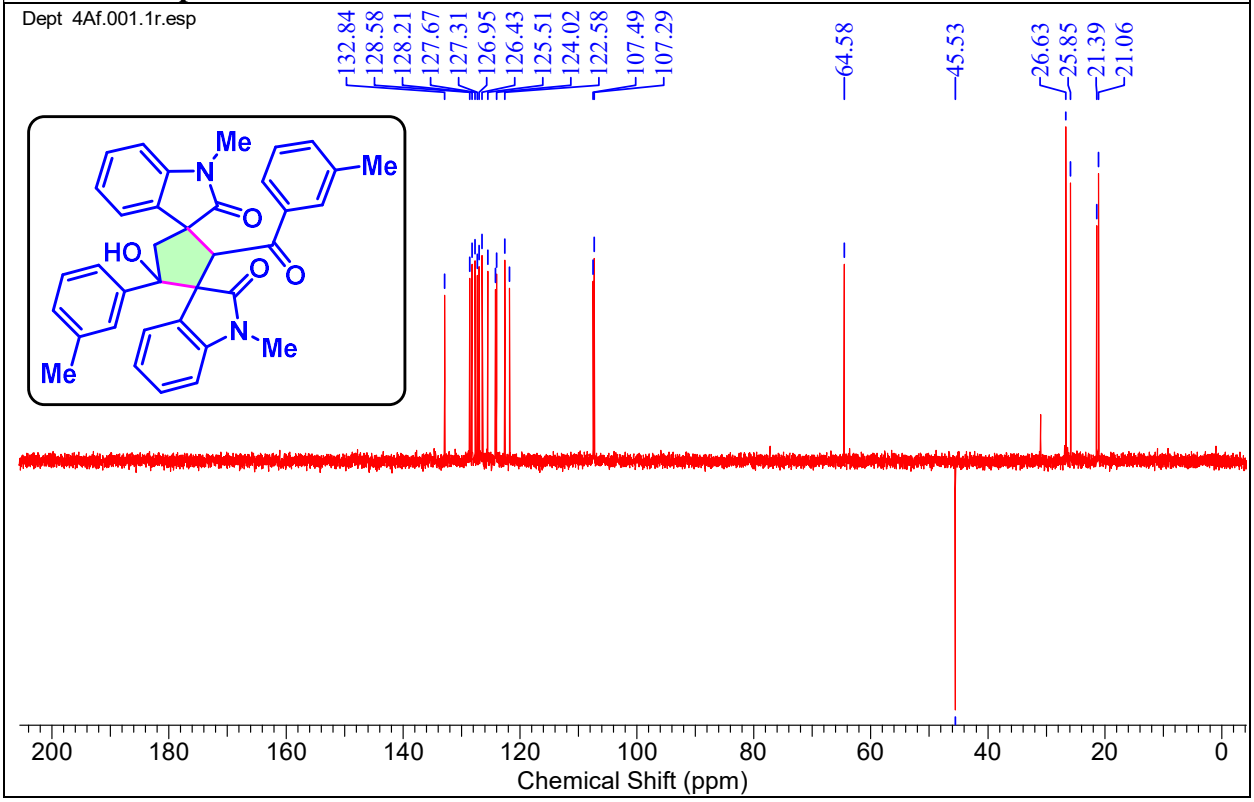
^{1}H NMR spectrum of 4Ab



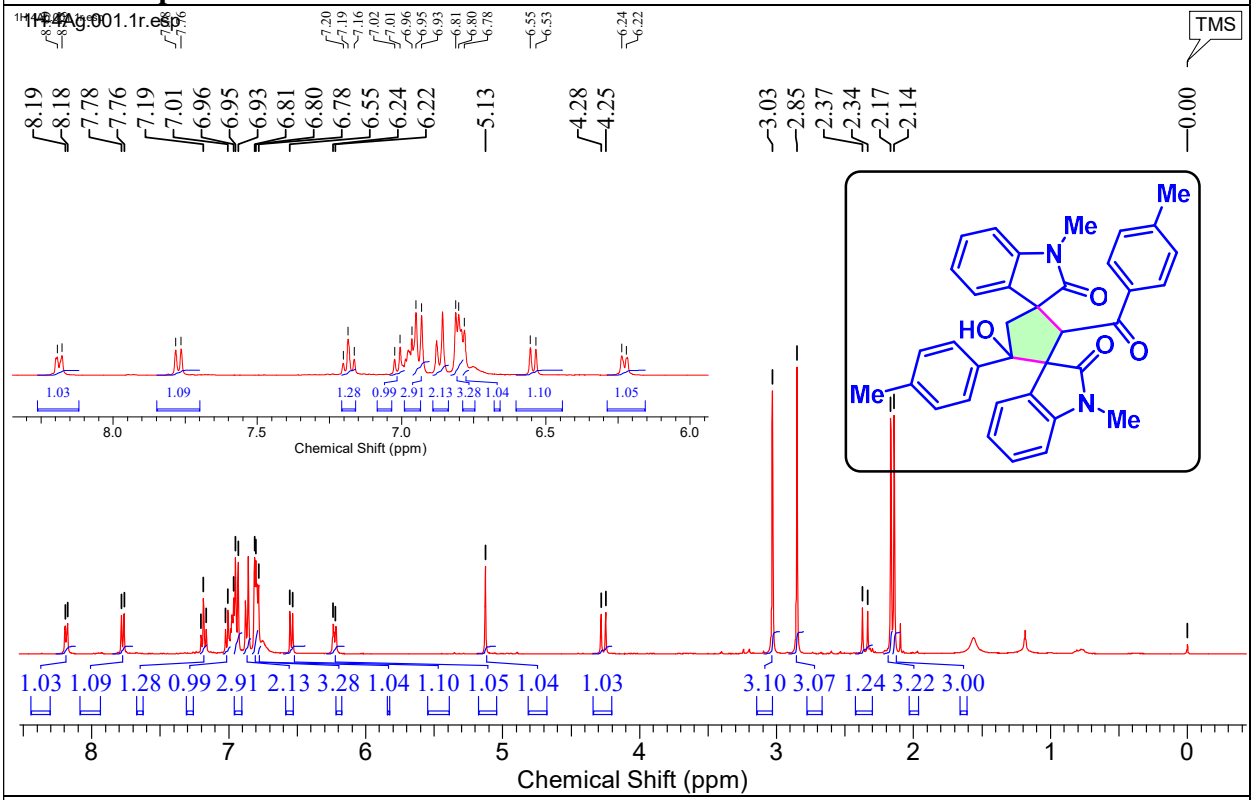
^{13}C NMR spectrum of 4Ab



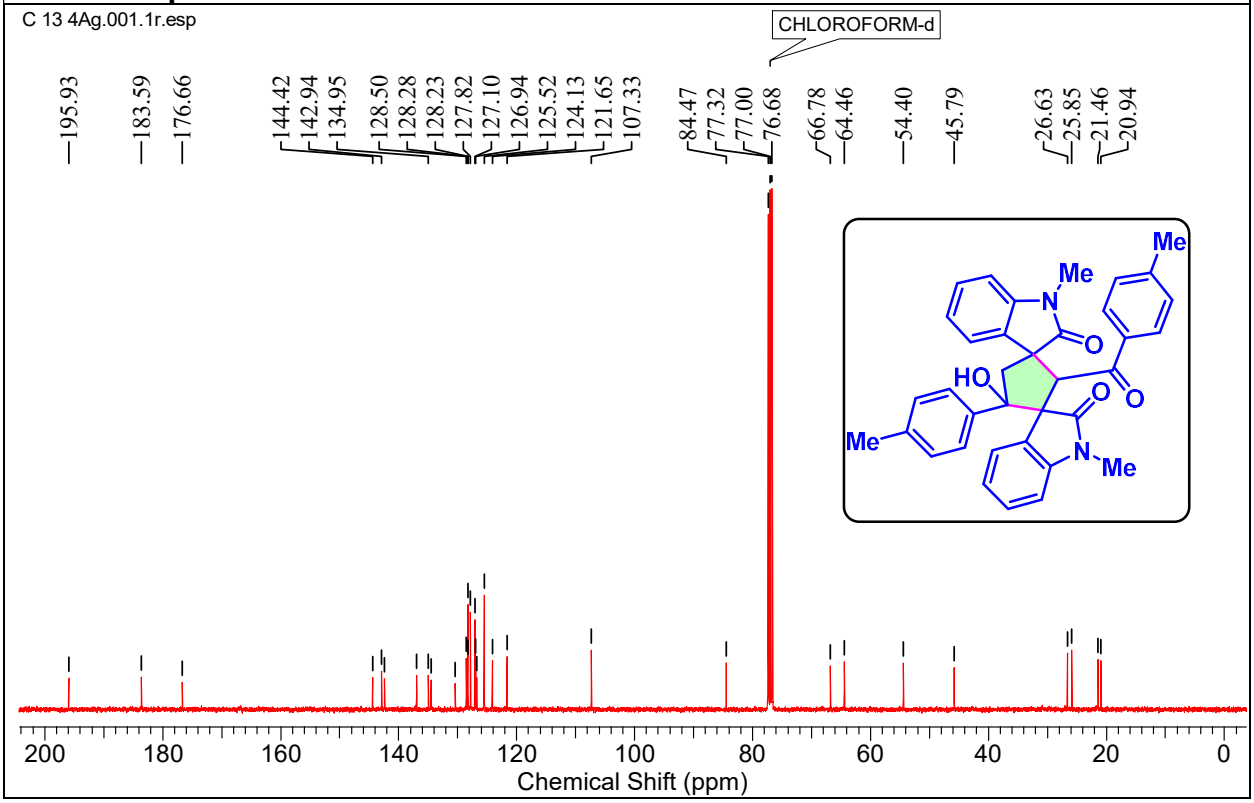
135 DEPT spectrum of 4Ab



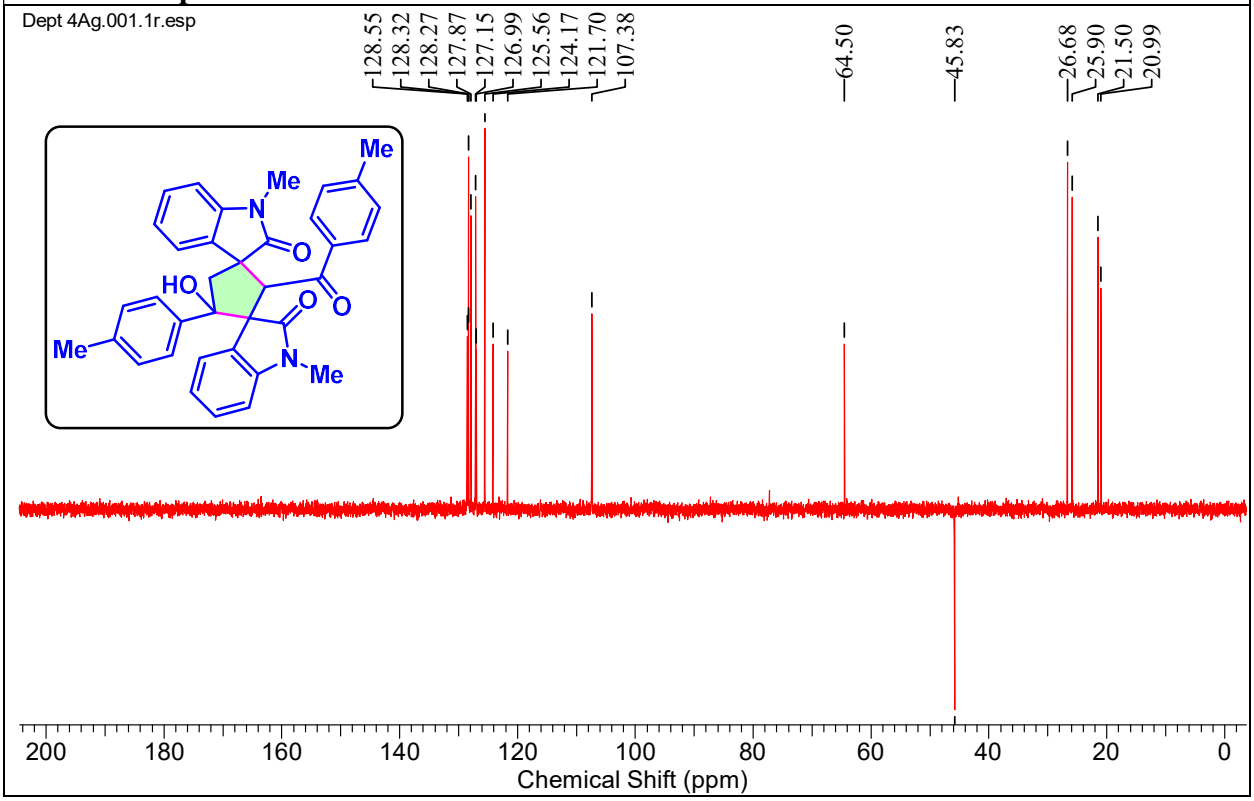
^{1}H NMR spectrum of 4Ac



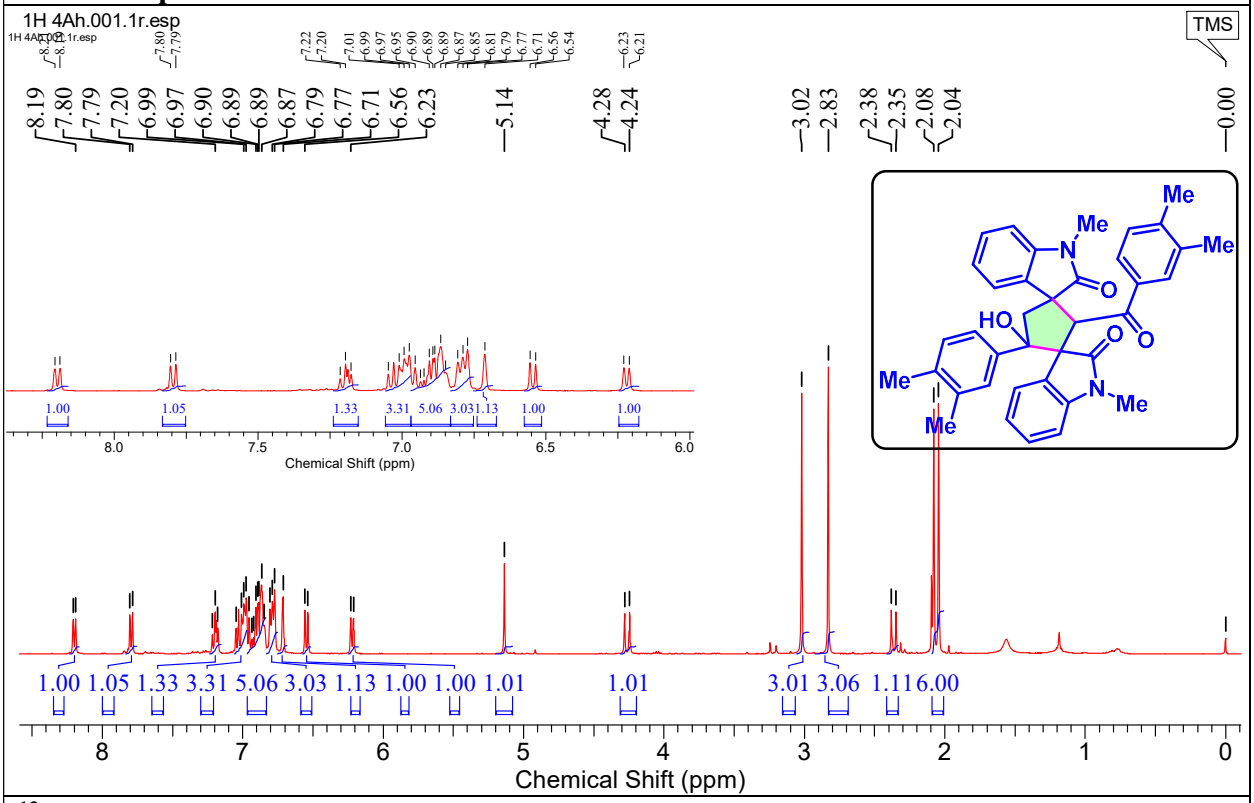
^{13}C NMR spectrum of 4Ac



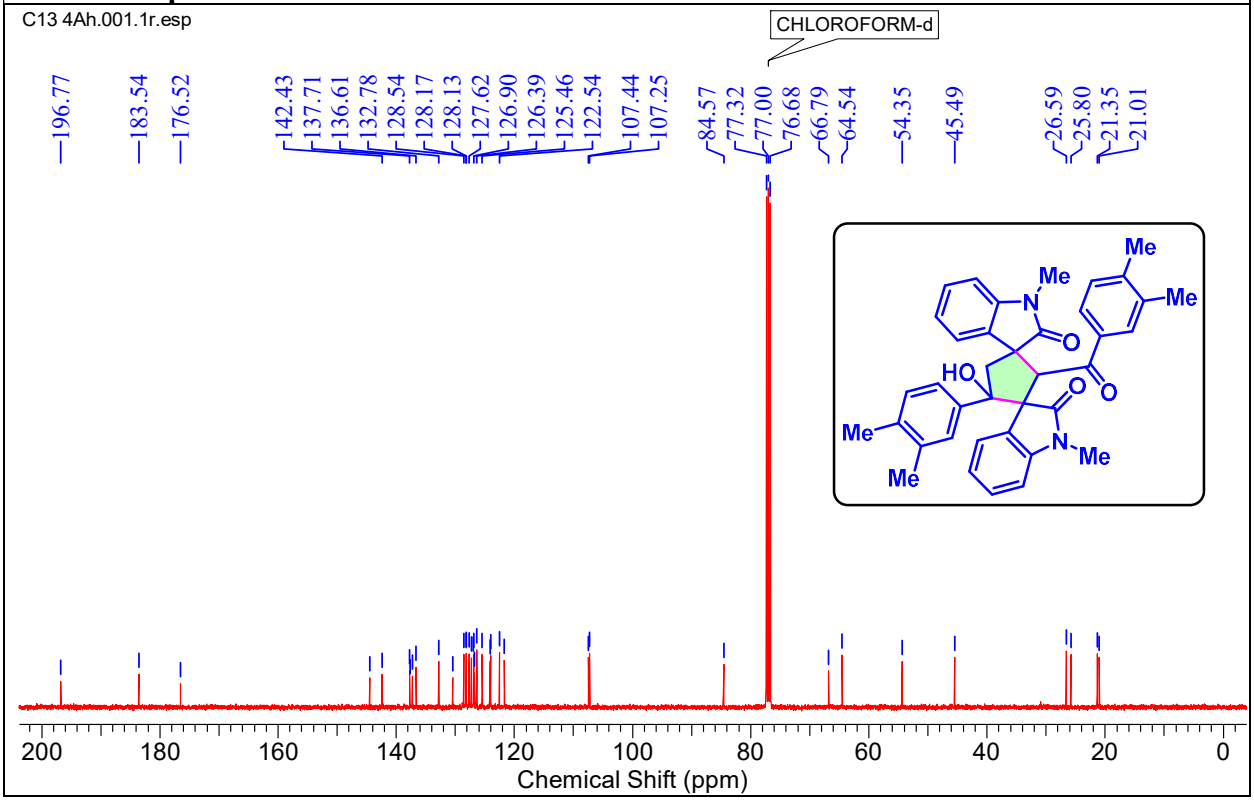
135 DEPT spectrum of 4Ac



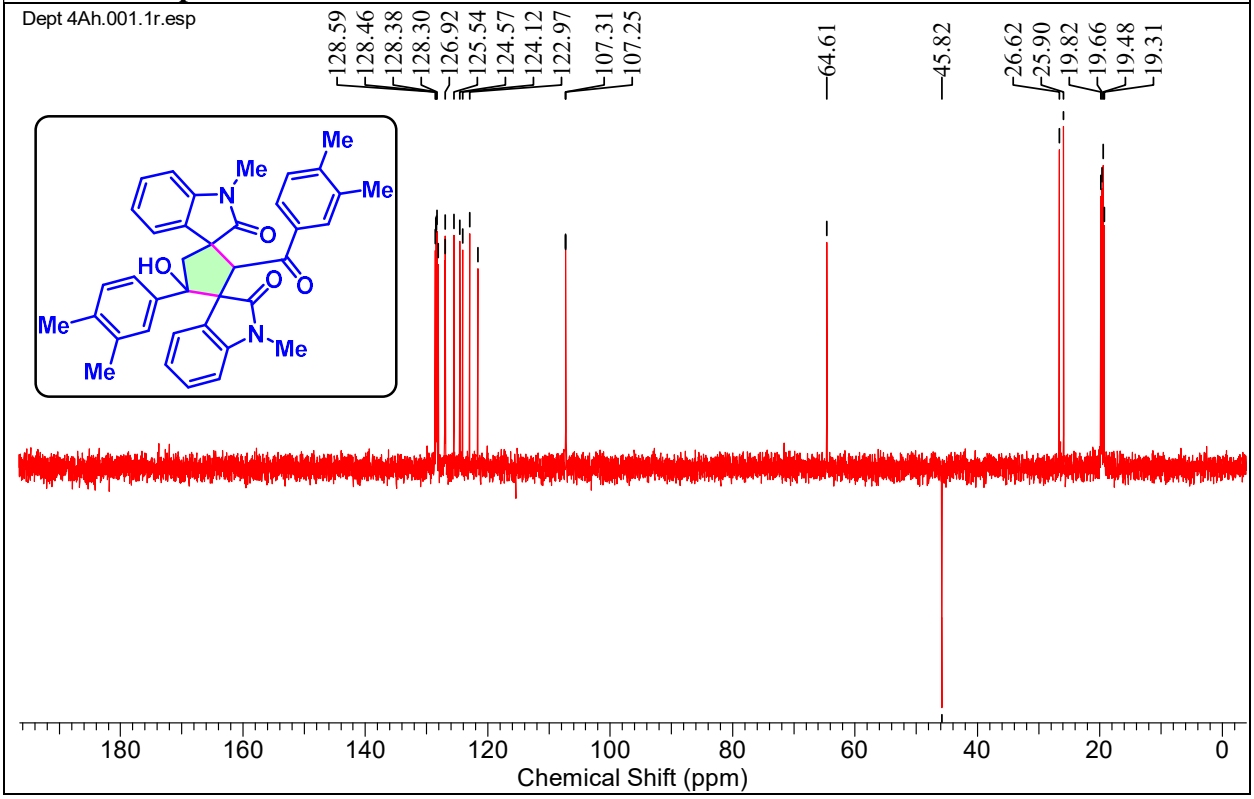
^{1}H NMR spectrum of 4Ad



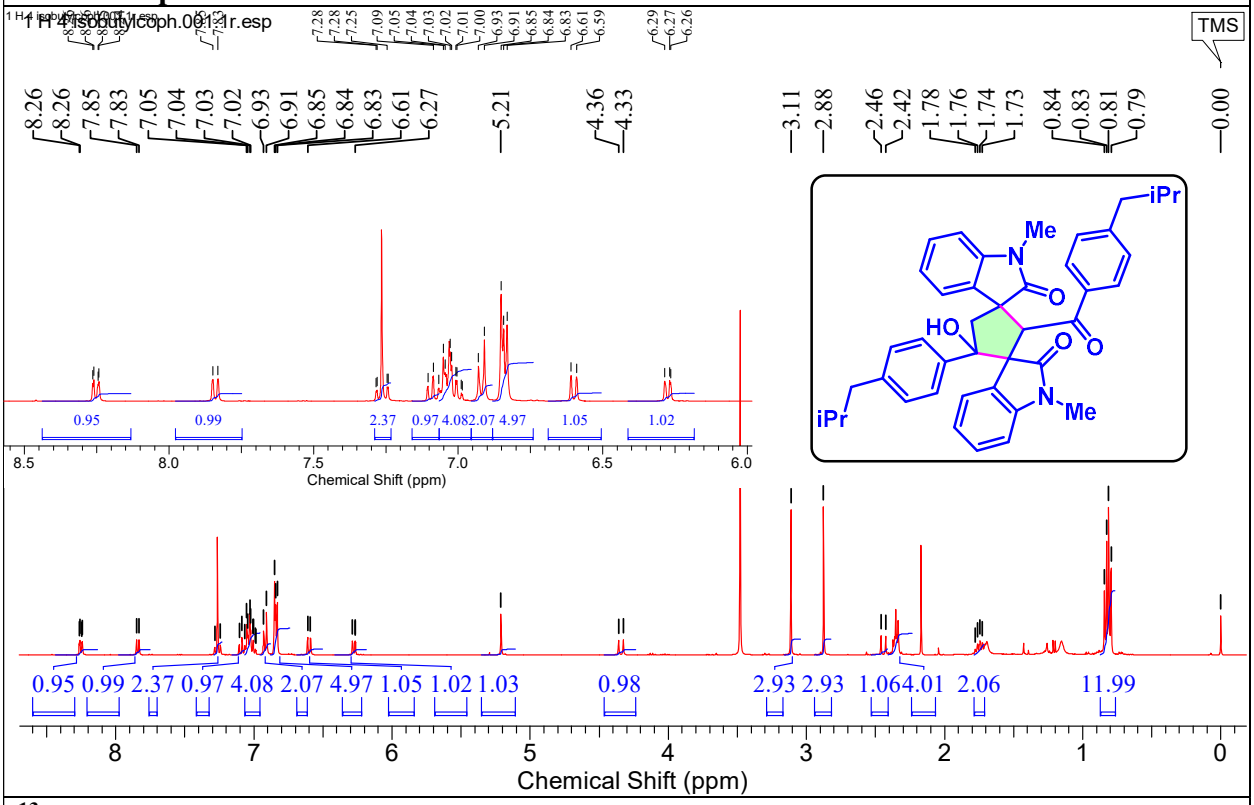
^{13}C NMR spectrum of 4Ad



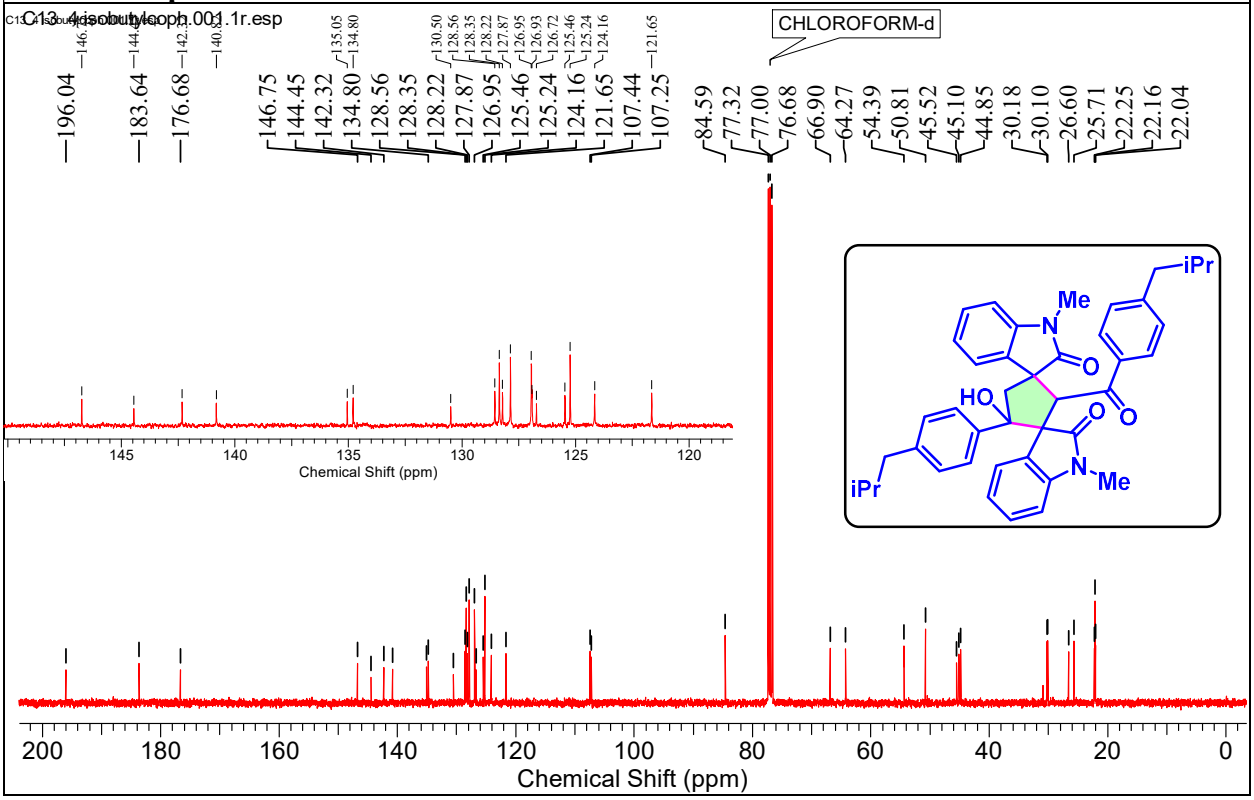
135 DEPT spectrum of 4Ad



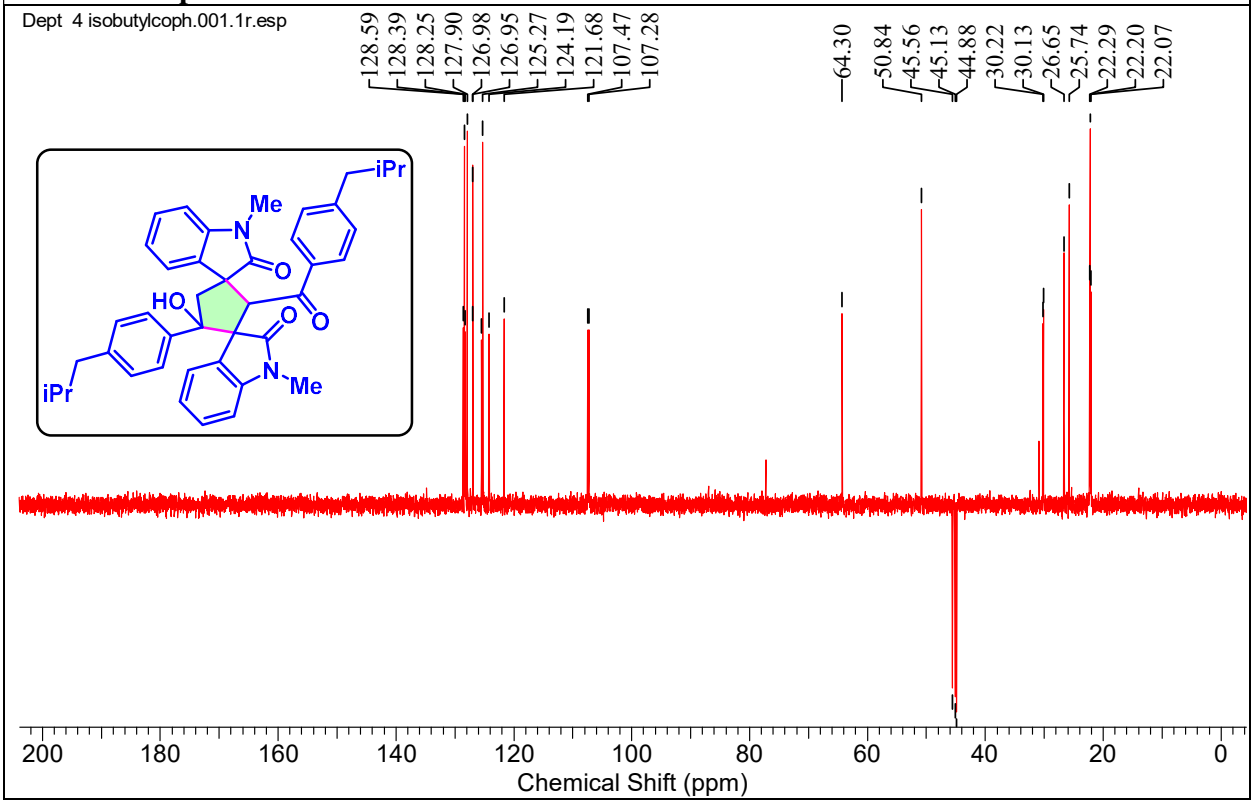
^{1}H NMR spectrum of 4Ae



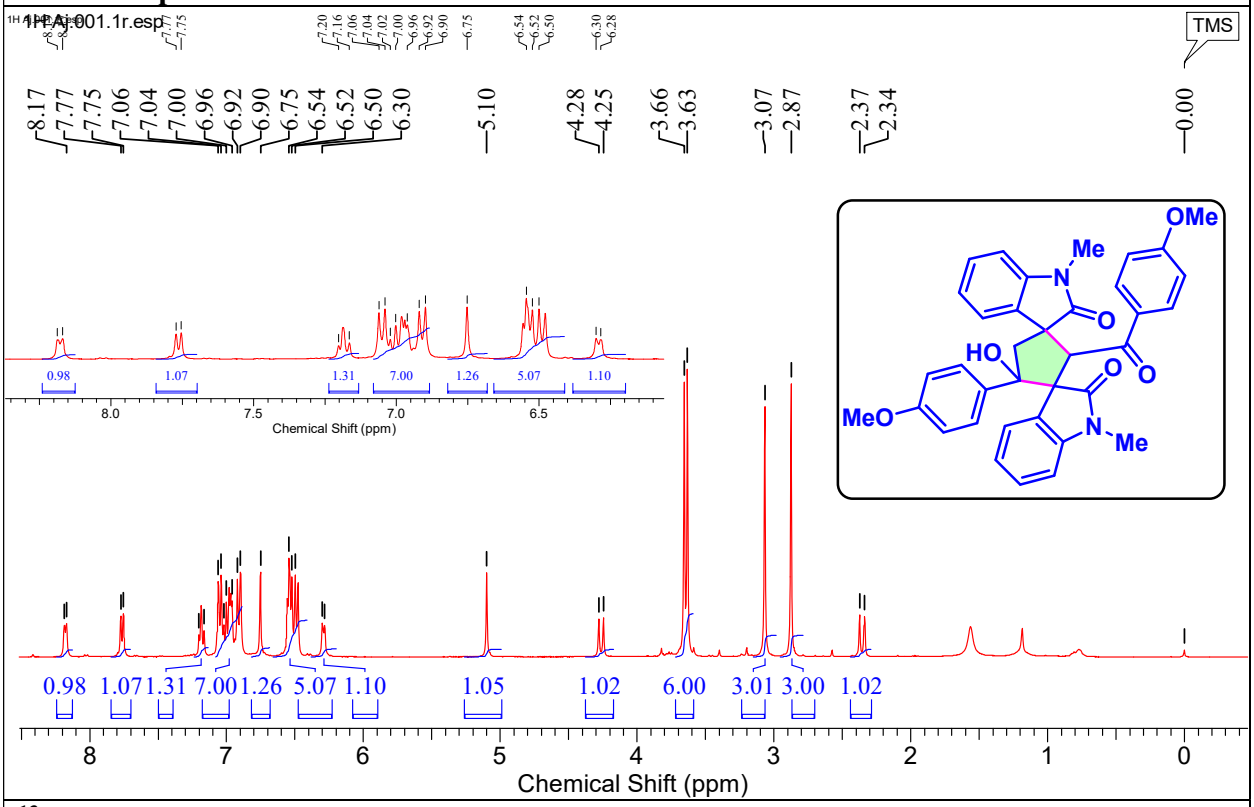
^{13}C NMR spectrum of 4Ae



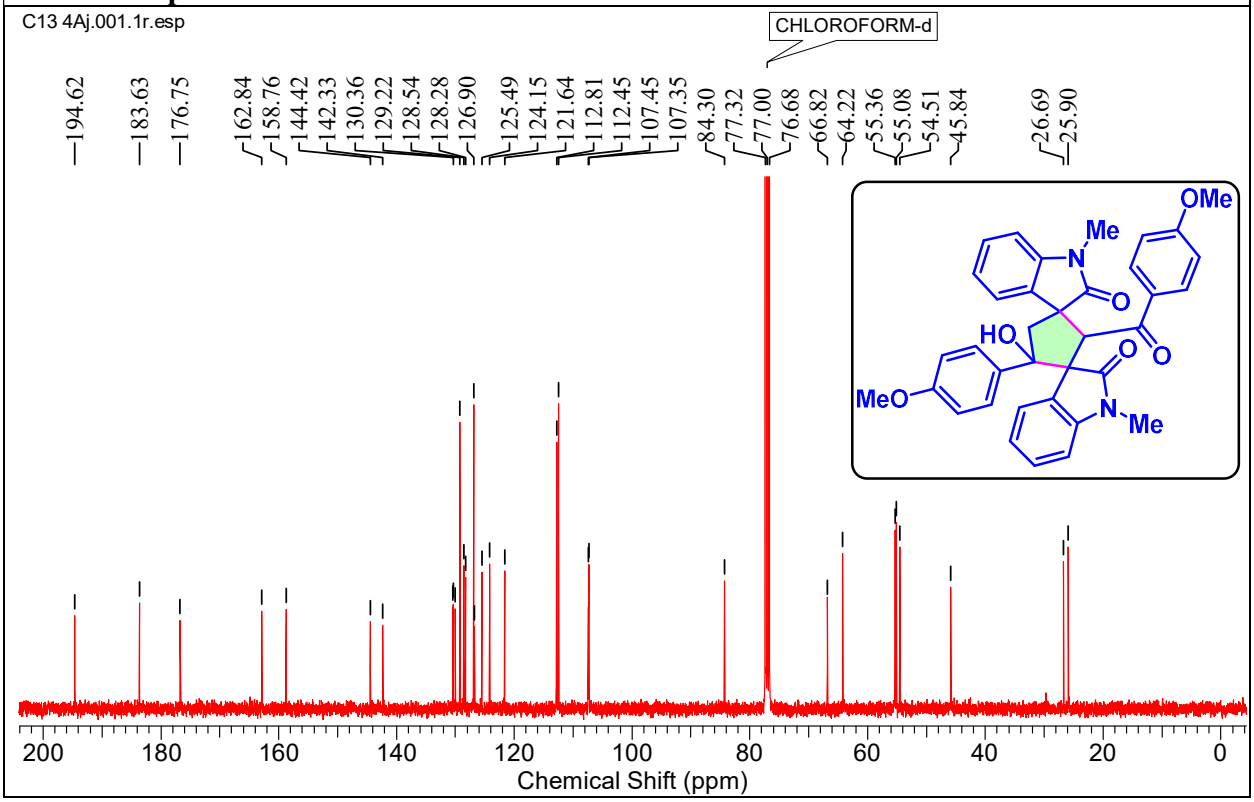
135 DEPT spectrum of 4Ae



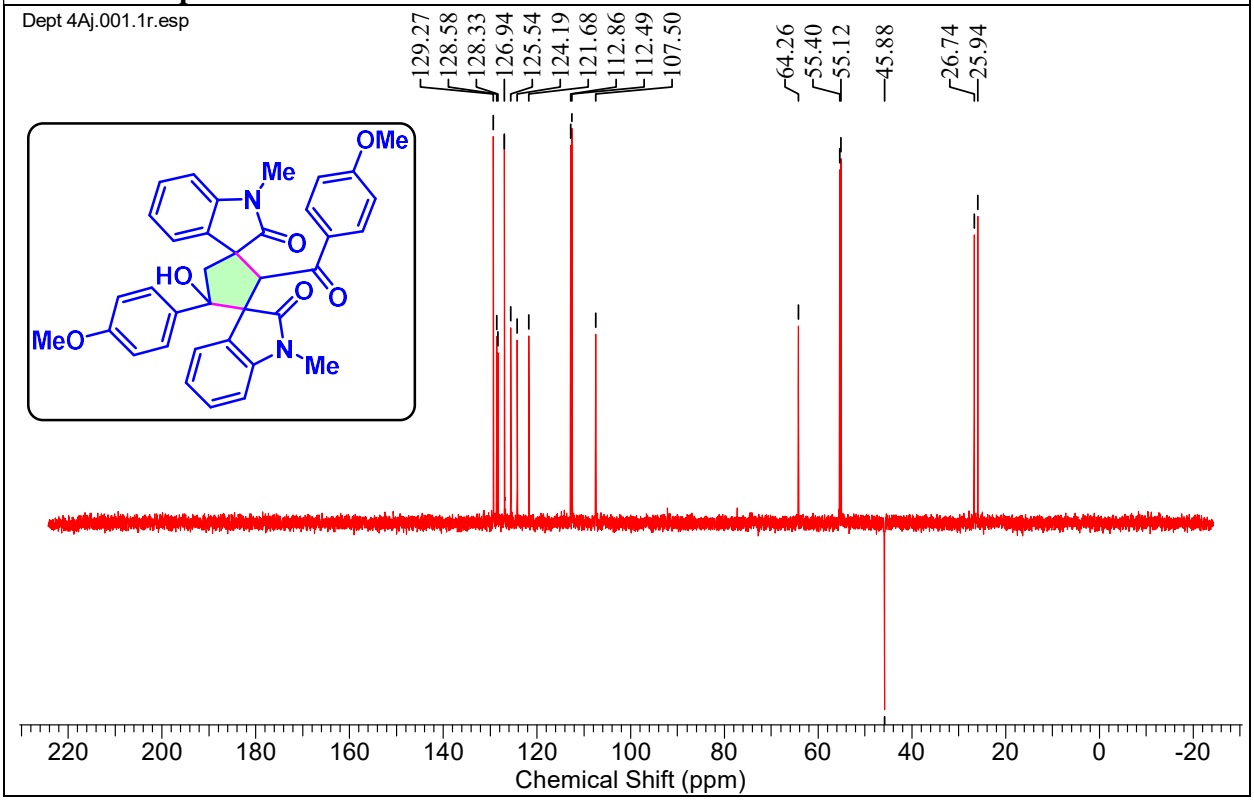
^{1}H NMR spectrum of 4Af



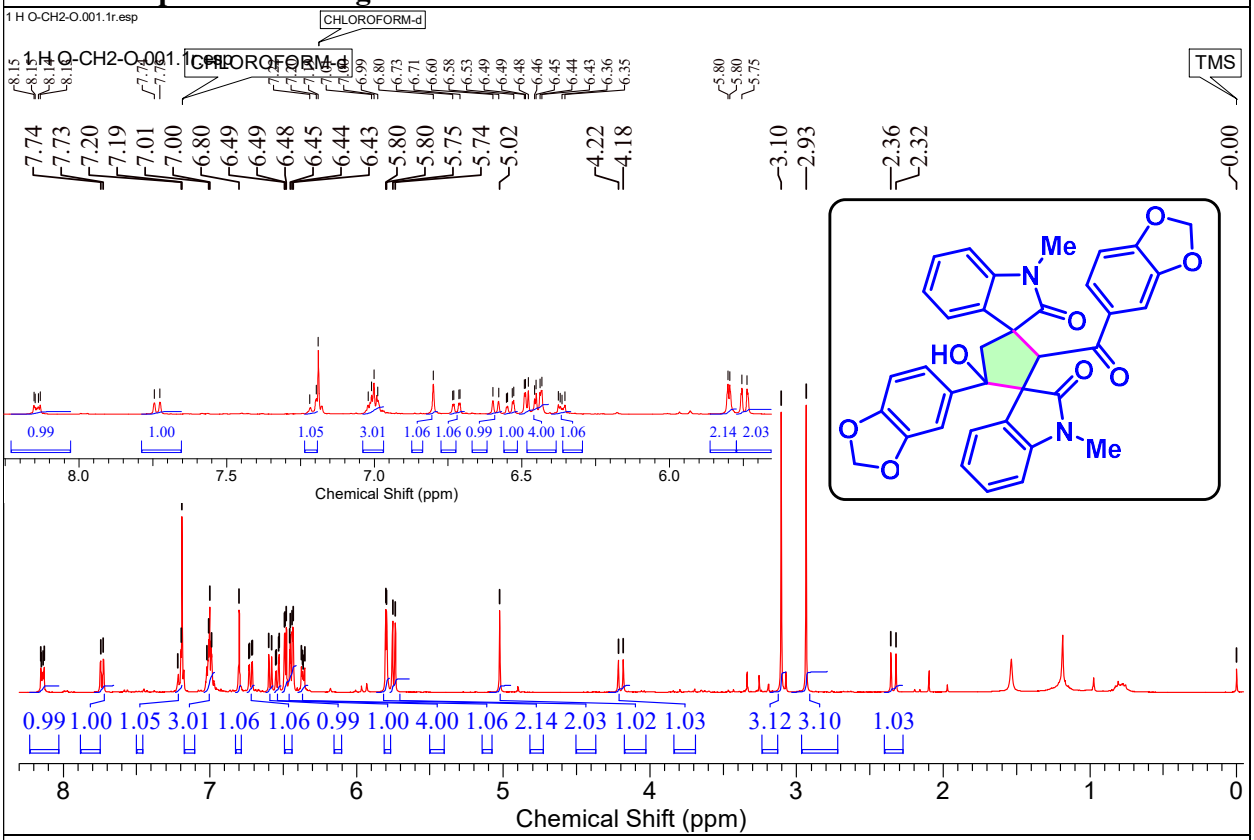
^{13}C NMR spectrum of 4Af



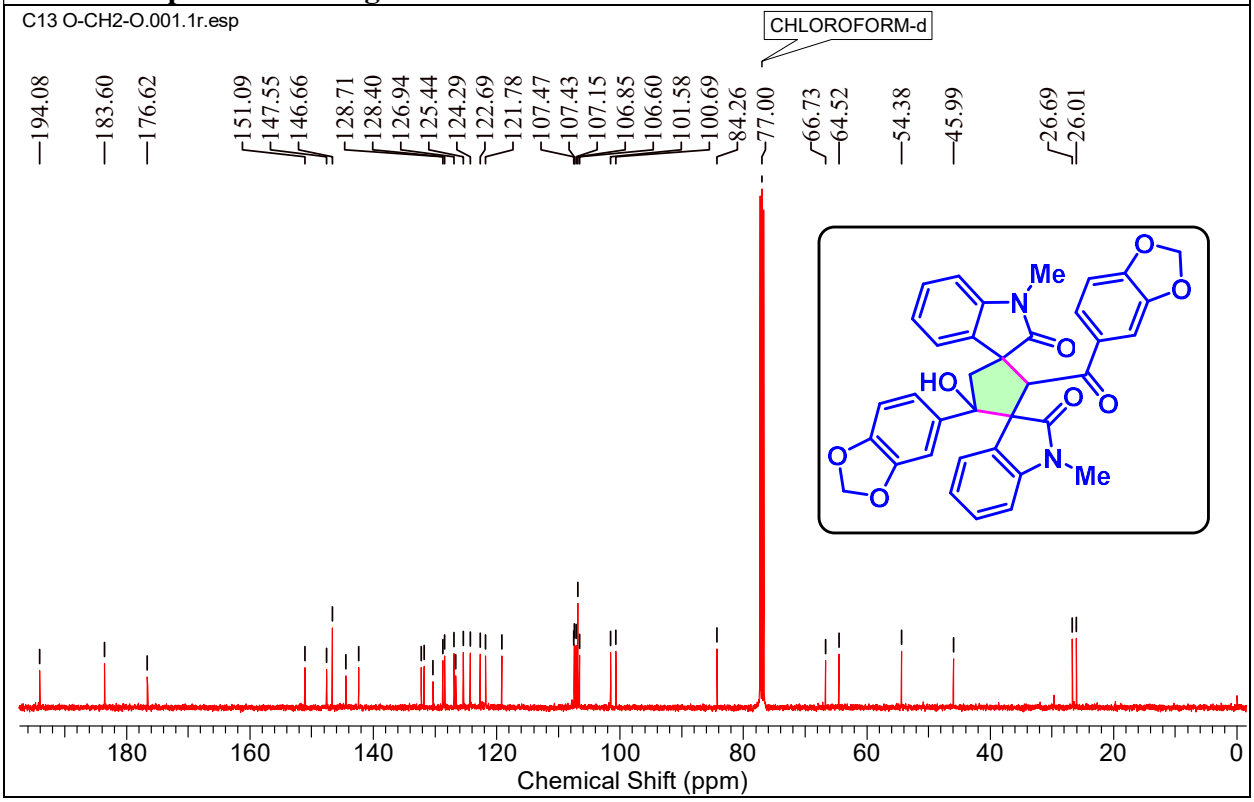
135 DEPT spectrum of 4Af



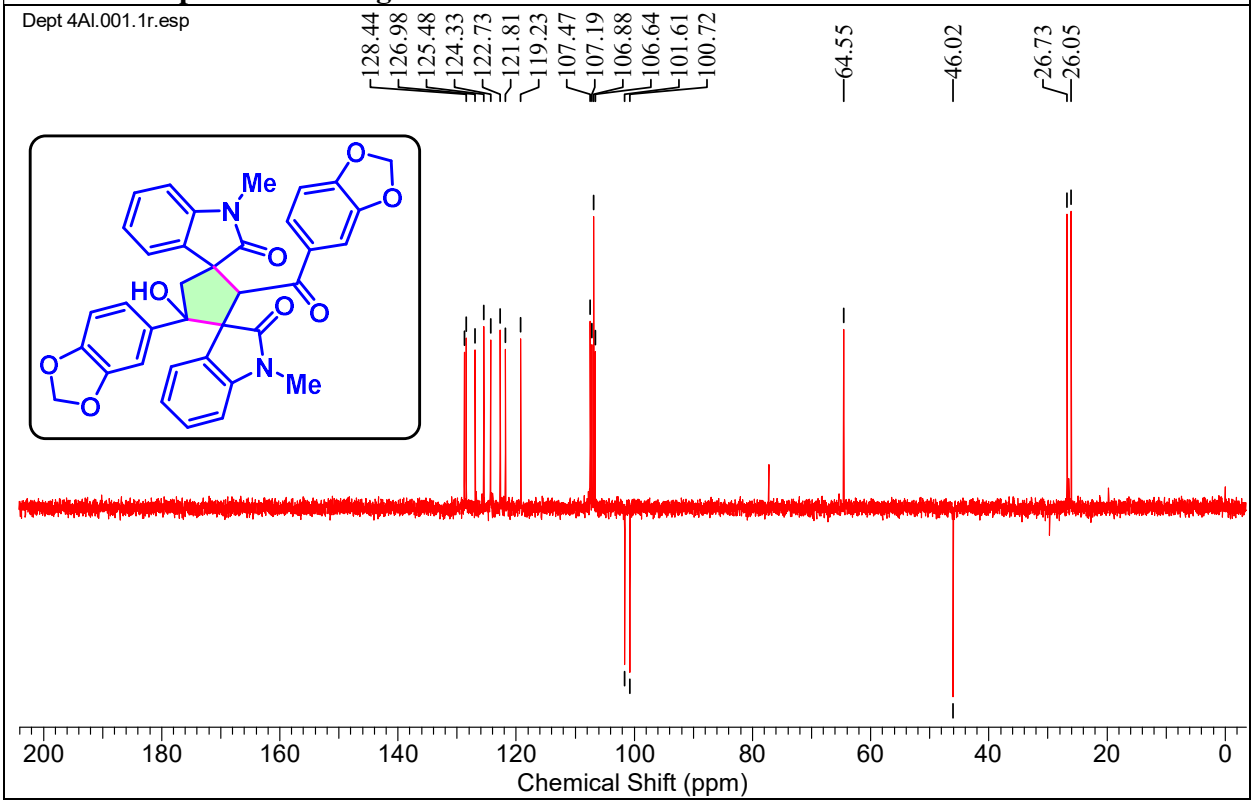
^{1}H NMR spectrum of 4Ag



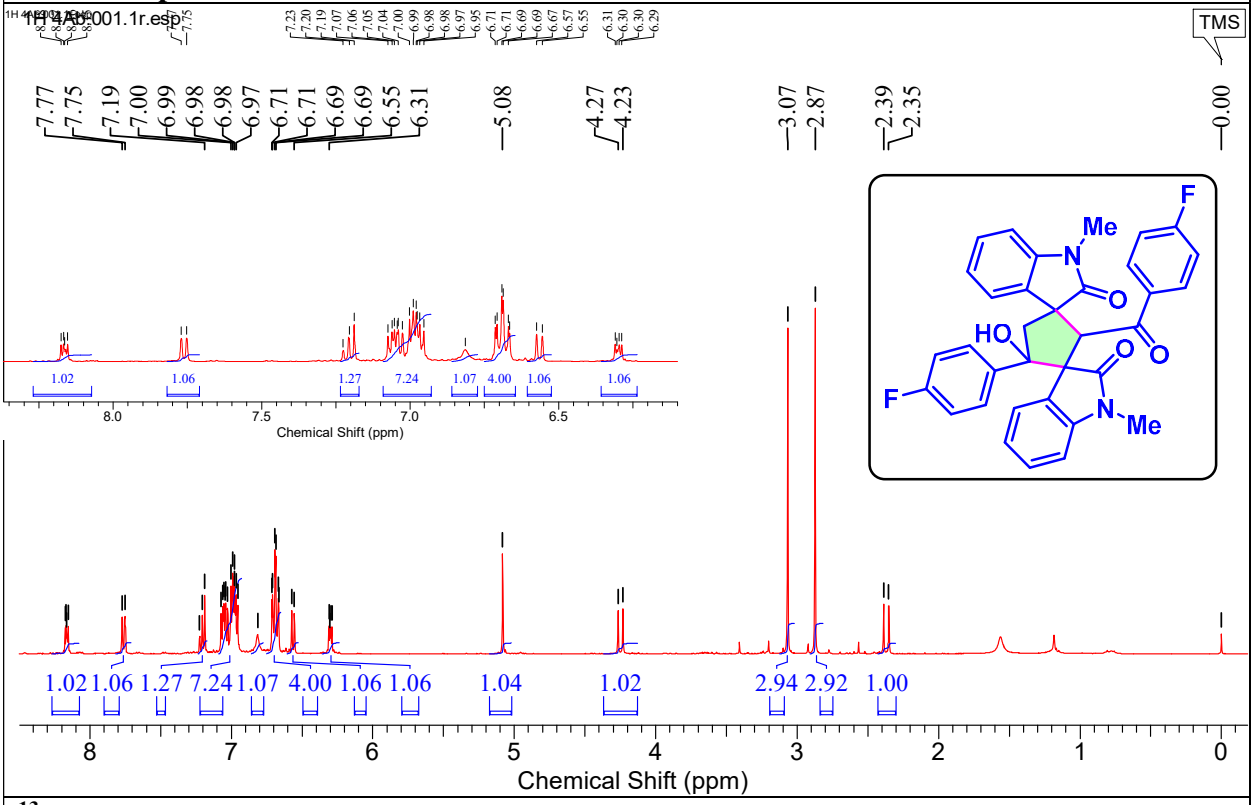
^{13}C NMR spectrum of 4Ag



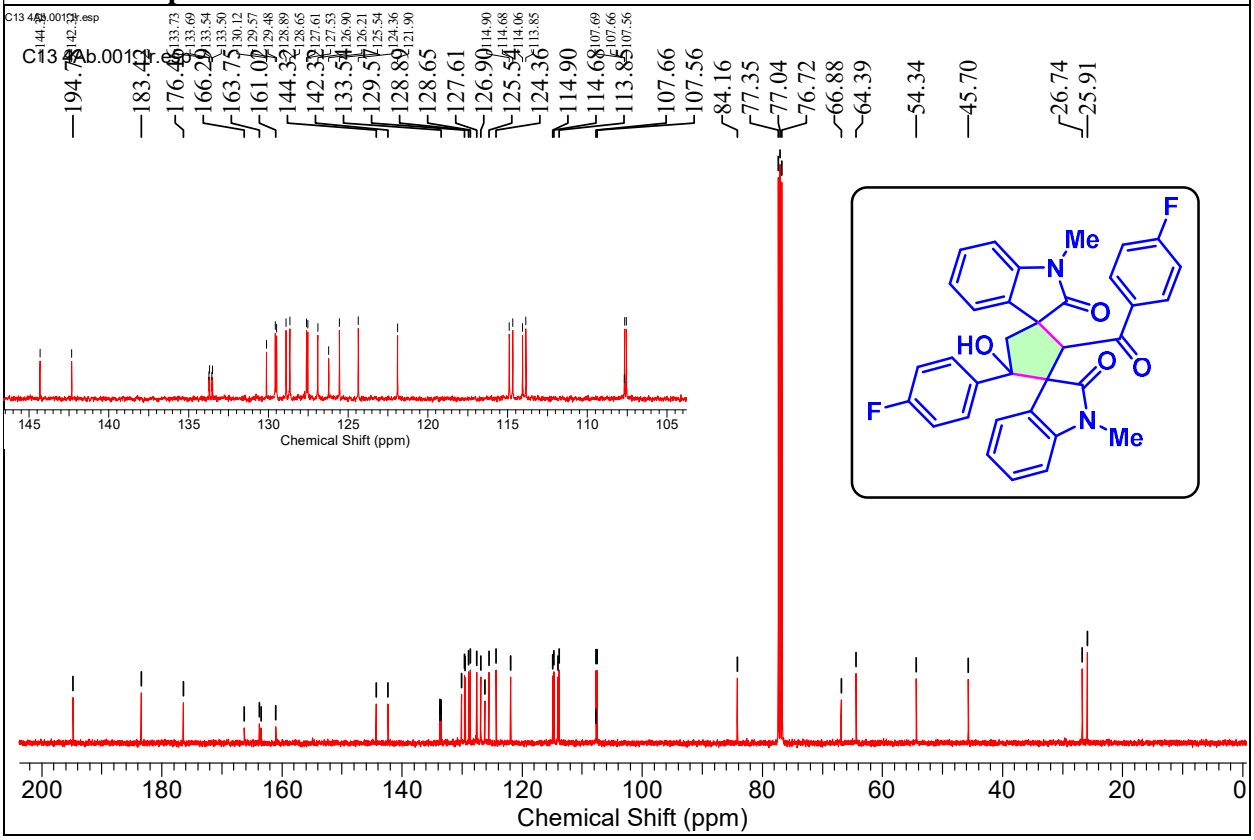
135 DEPT spectrum of 4Ag



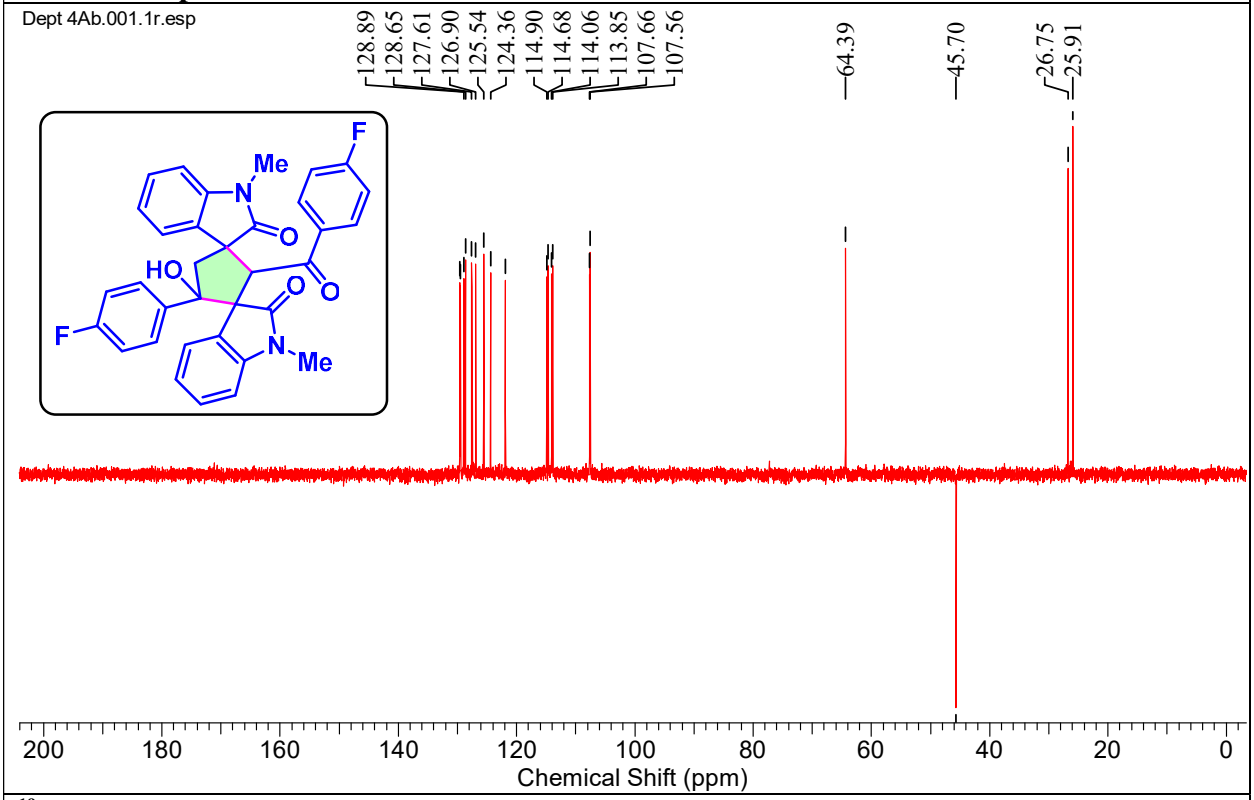
^{1}H NMR spectrum of 4Ah



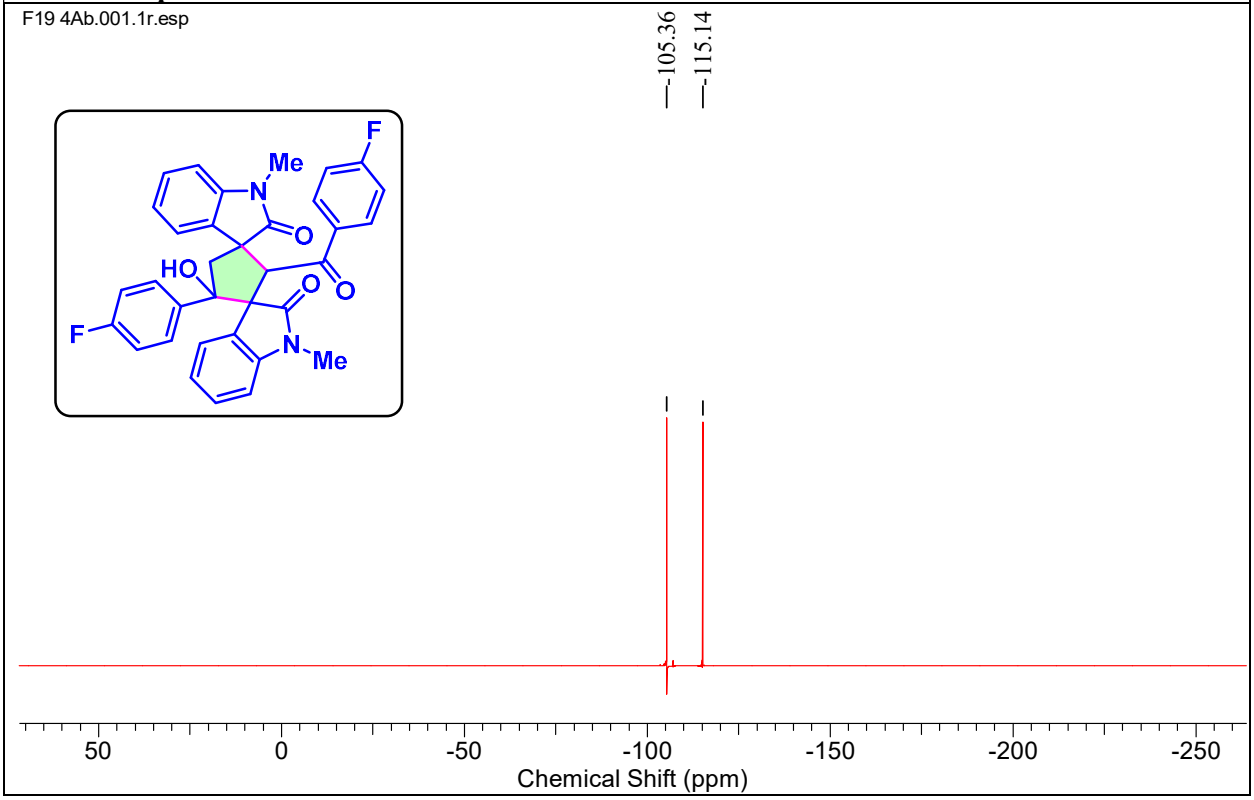
^{13}C NMR spectrum of 4Ah



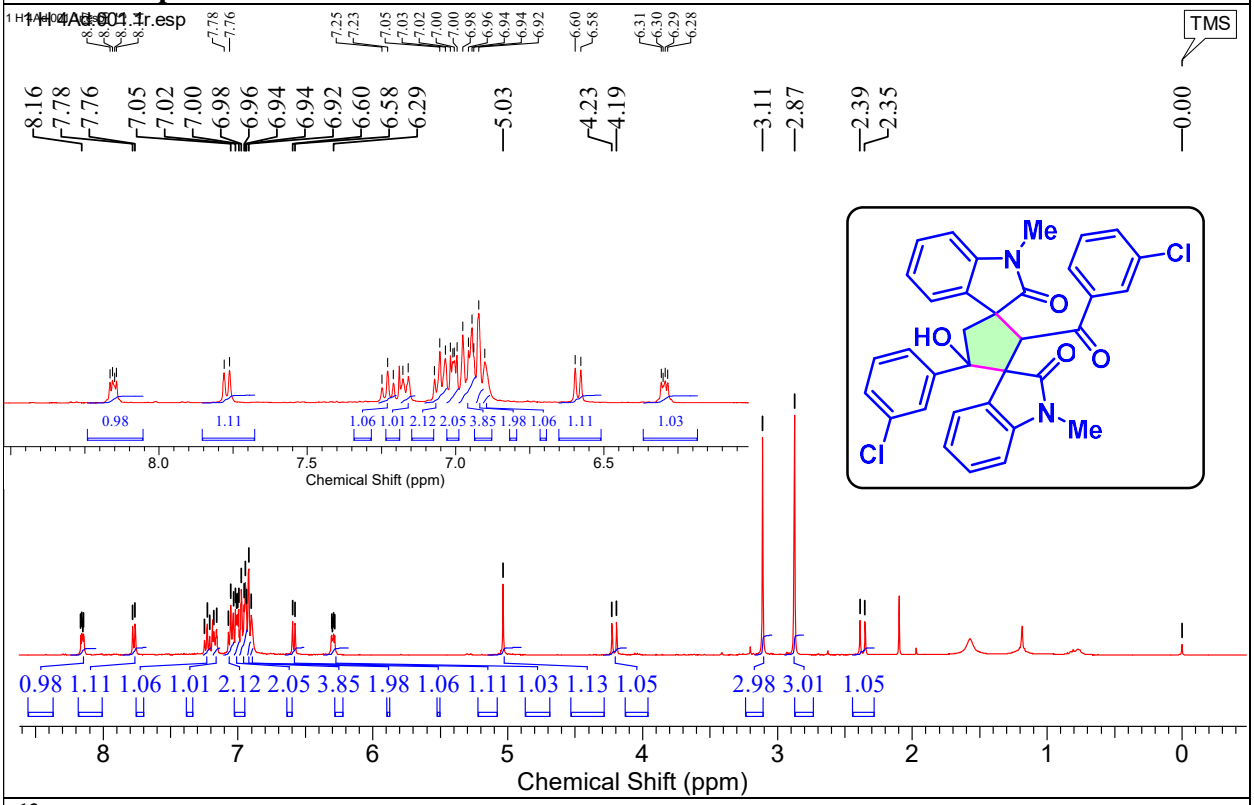
135 DEPT spectrum of 4Ah



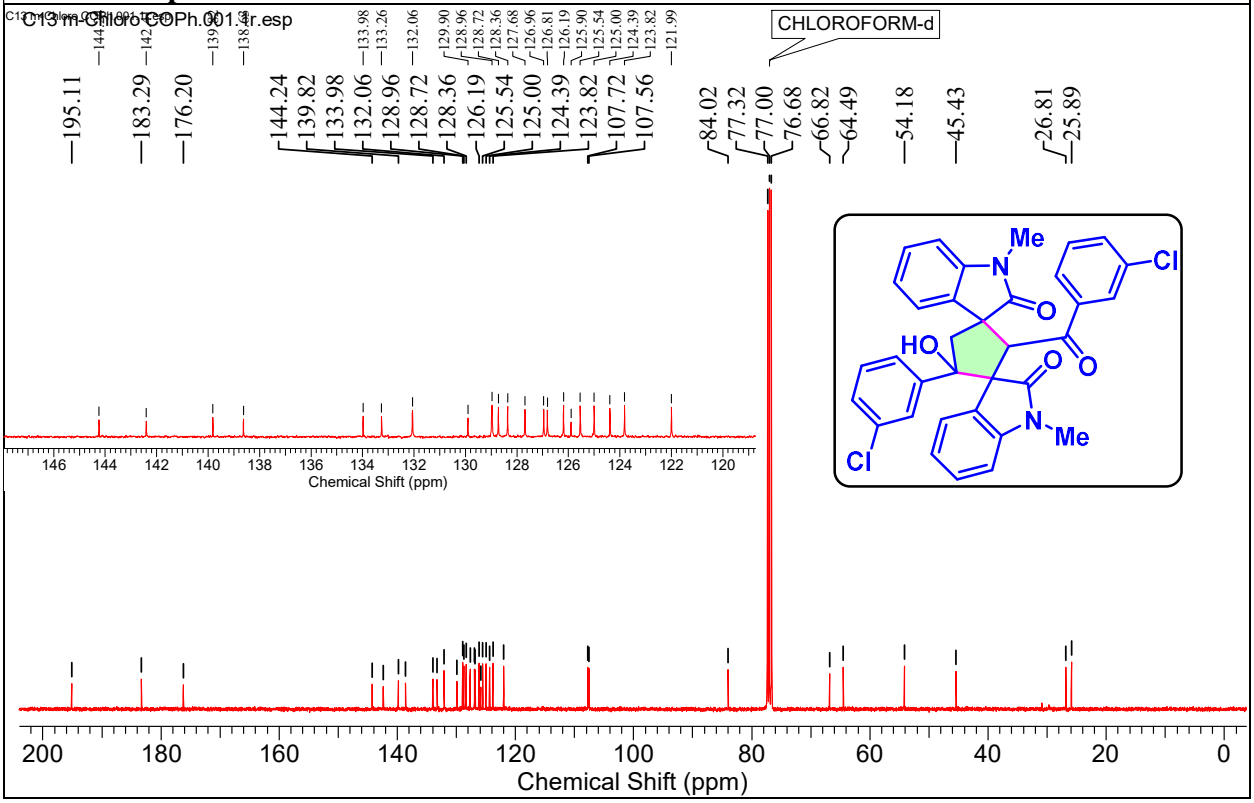
^{19}F NMR spectrum of 4Ah



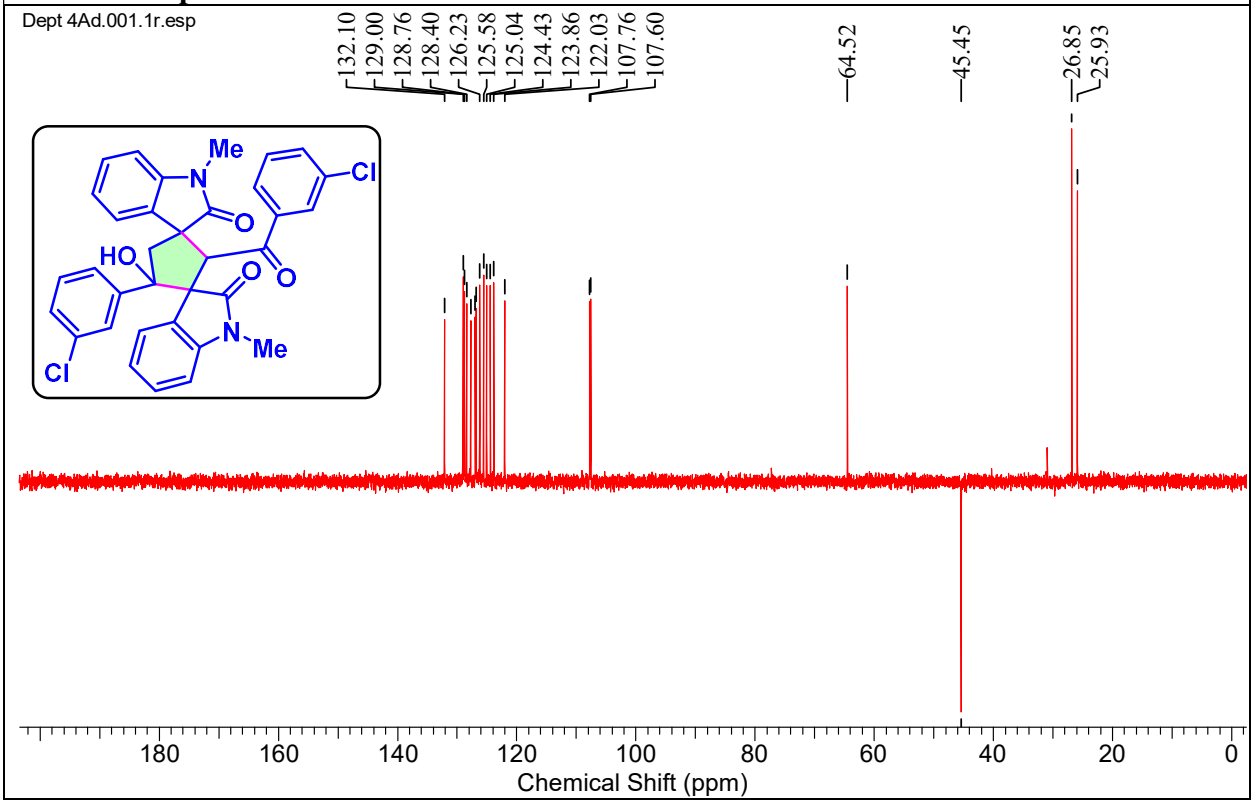
^{1}H NMR spectrum of 4Ai



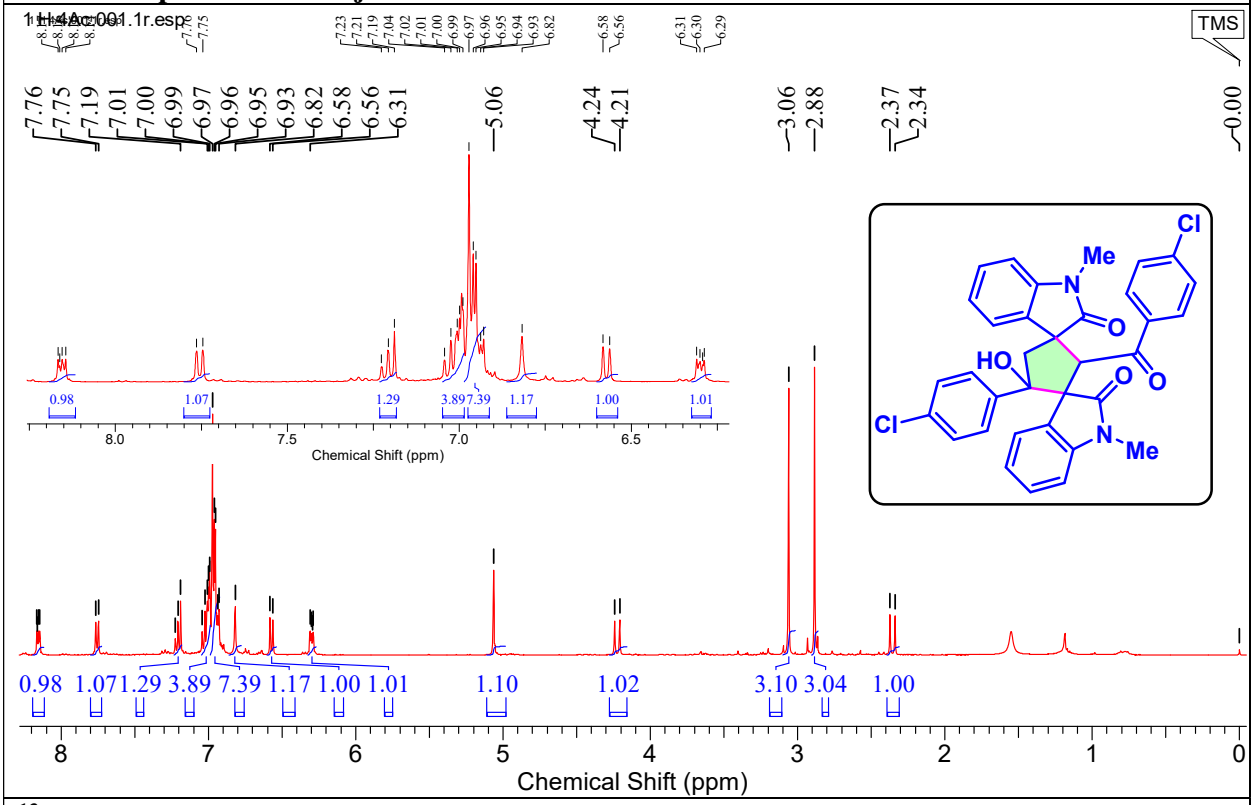
^{13}C NMR spectrum of 4Ai



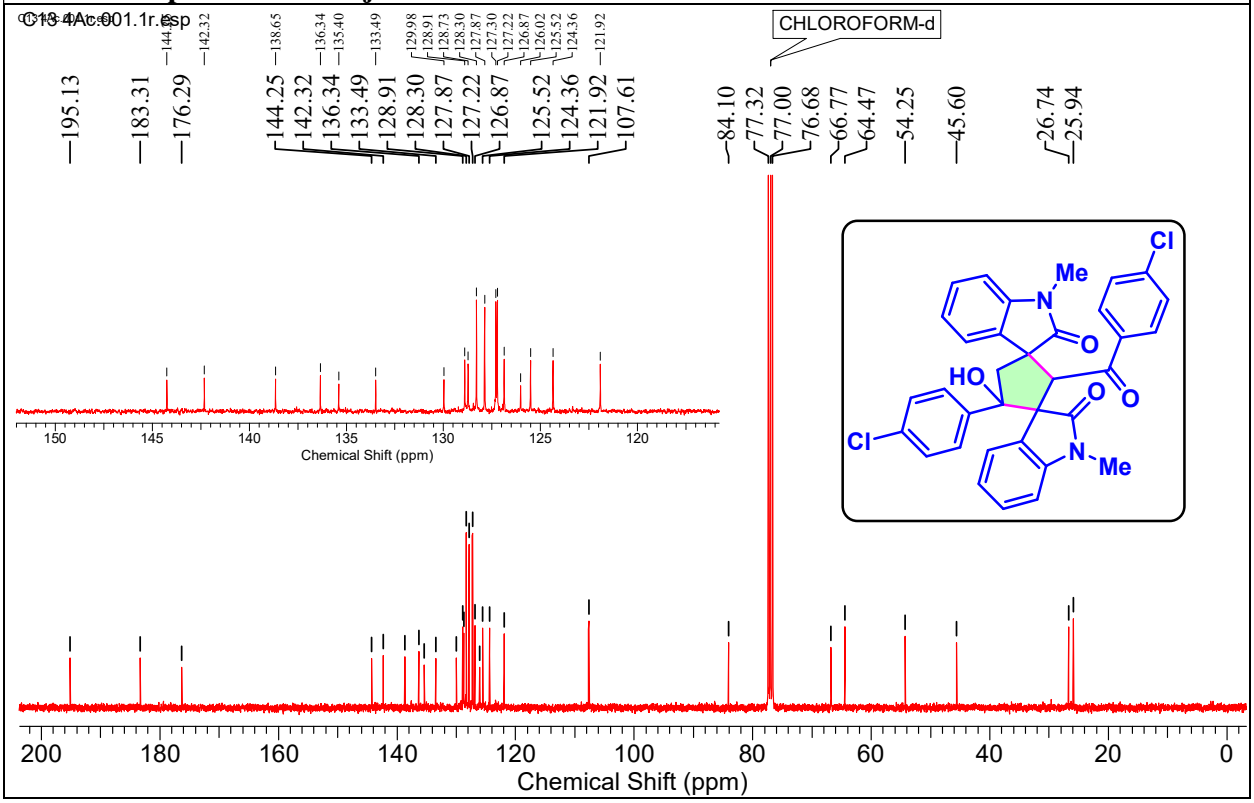
135 DEPT spectrum of 4Ai



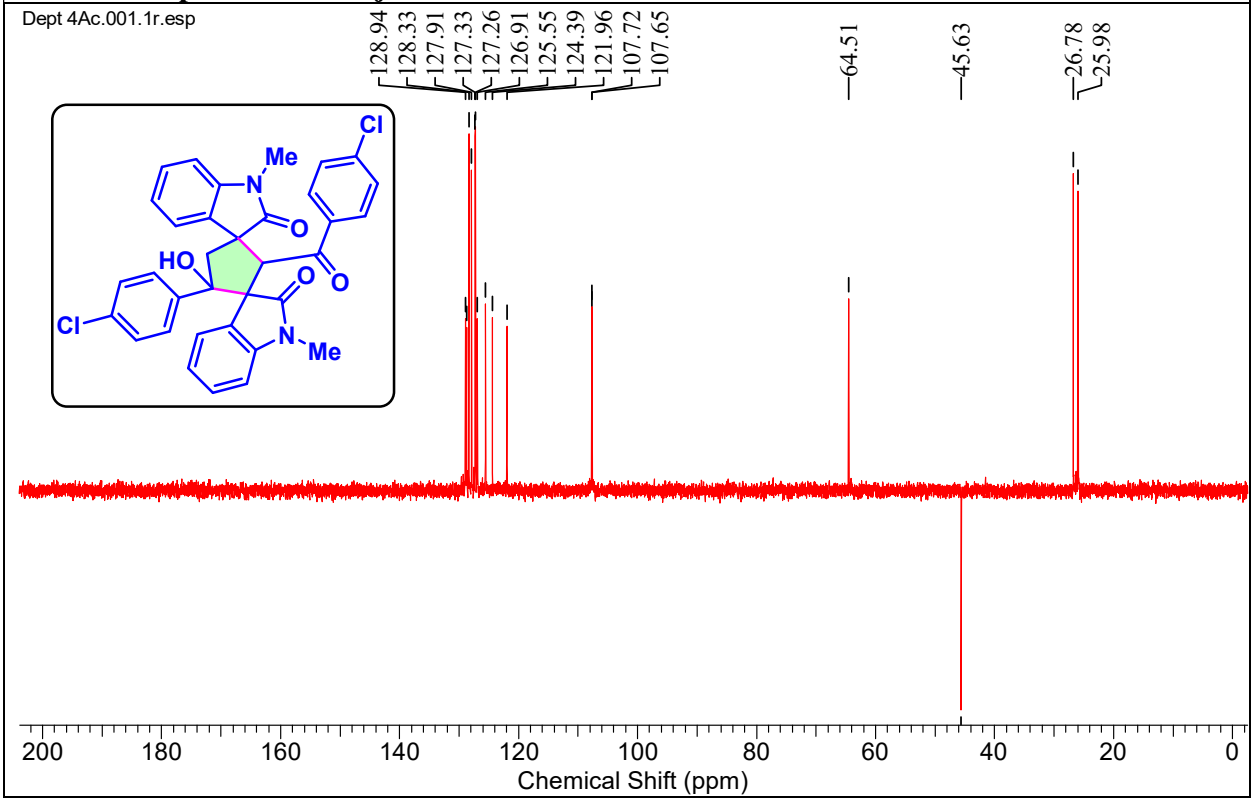
^{1}H NMR spectrum of 4Aj



^{13}C NMR spectrum of 4Aj



135 DEPT spectrum of 4Aj



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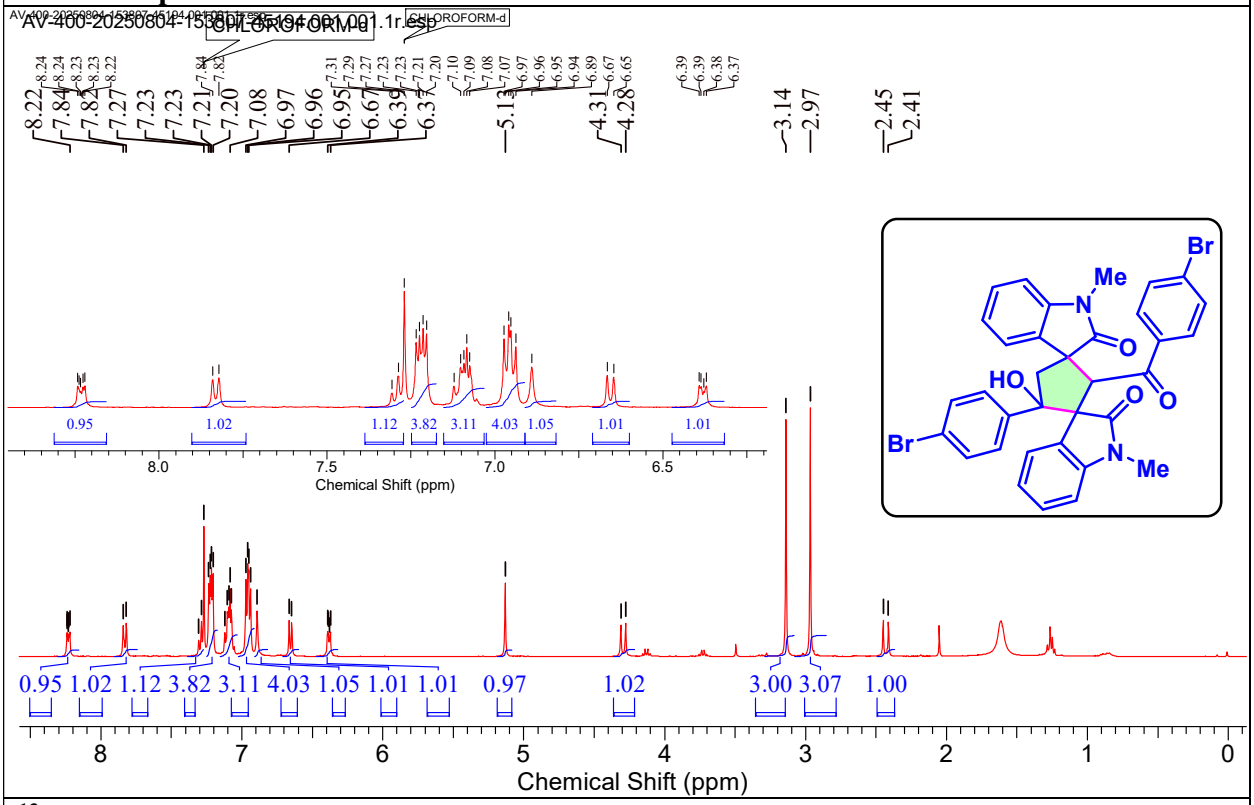
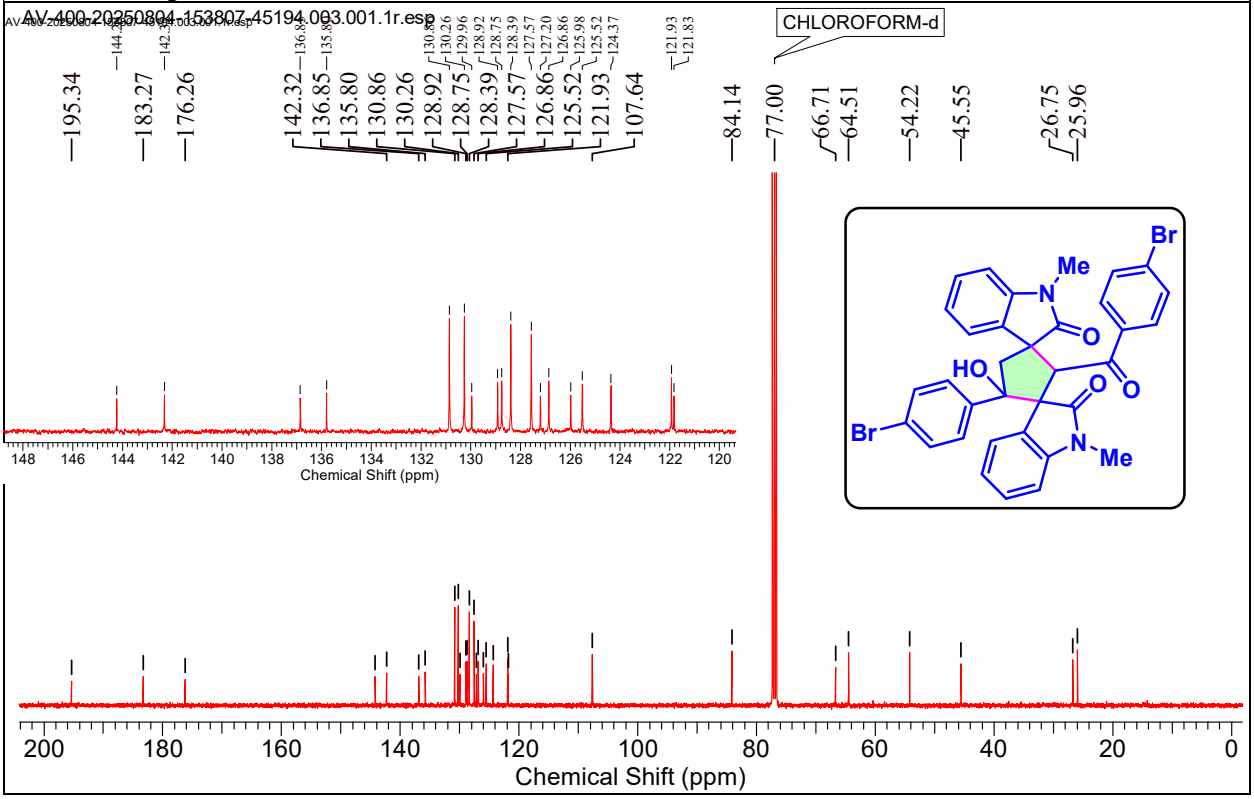
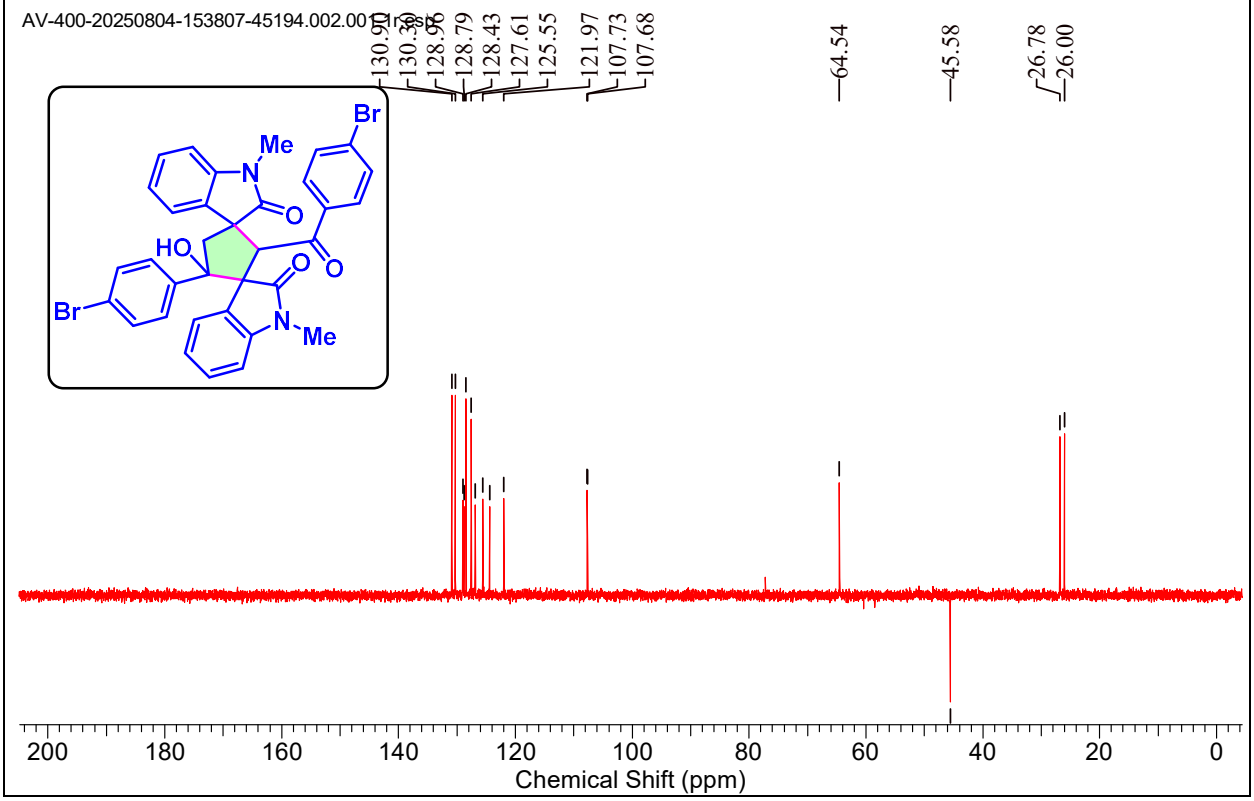


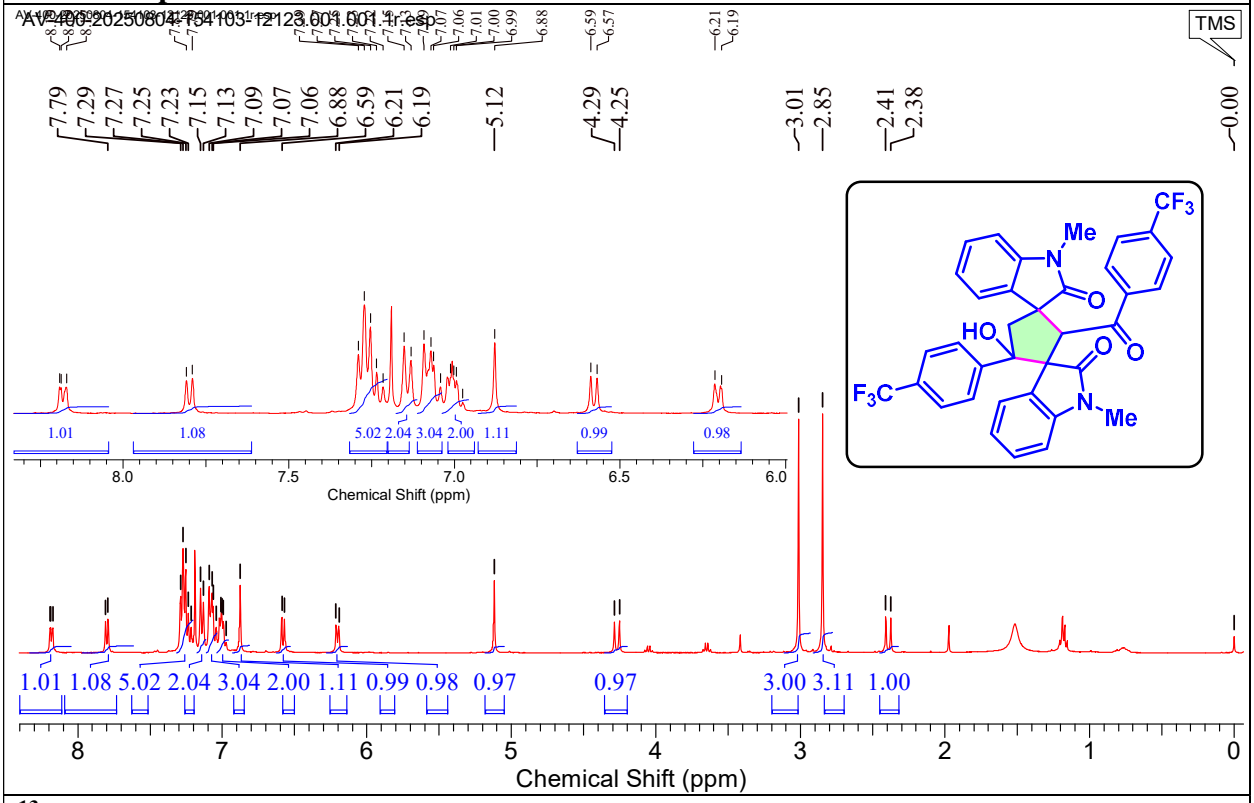
^{13}C NMR spectrum of 4Ak



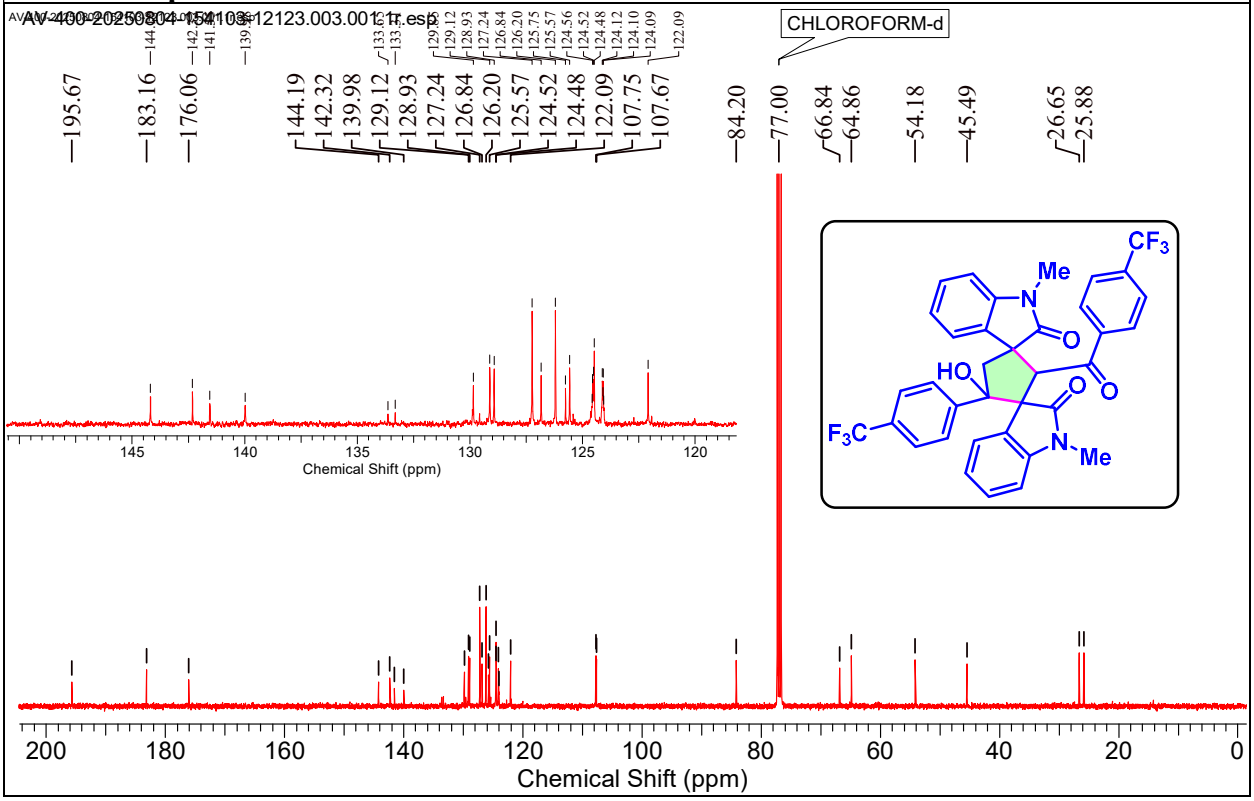
135 DEPT spectrum of 4Ak



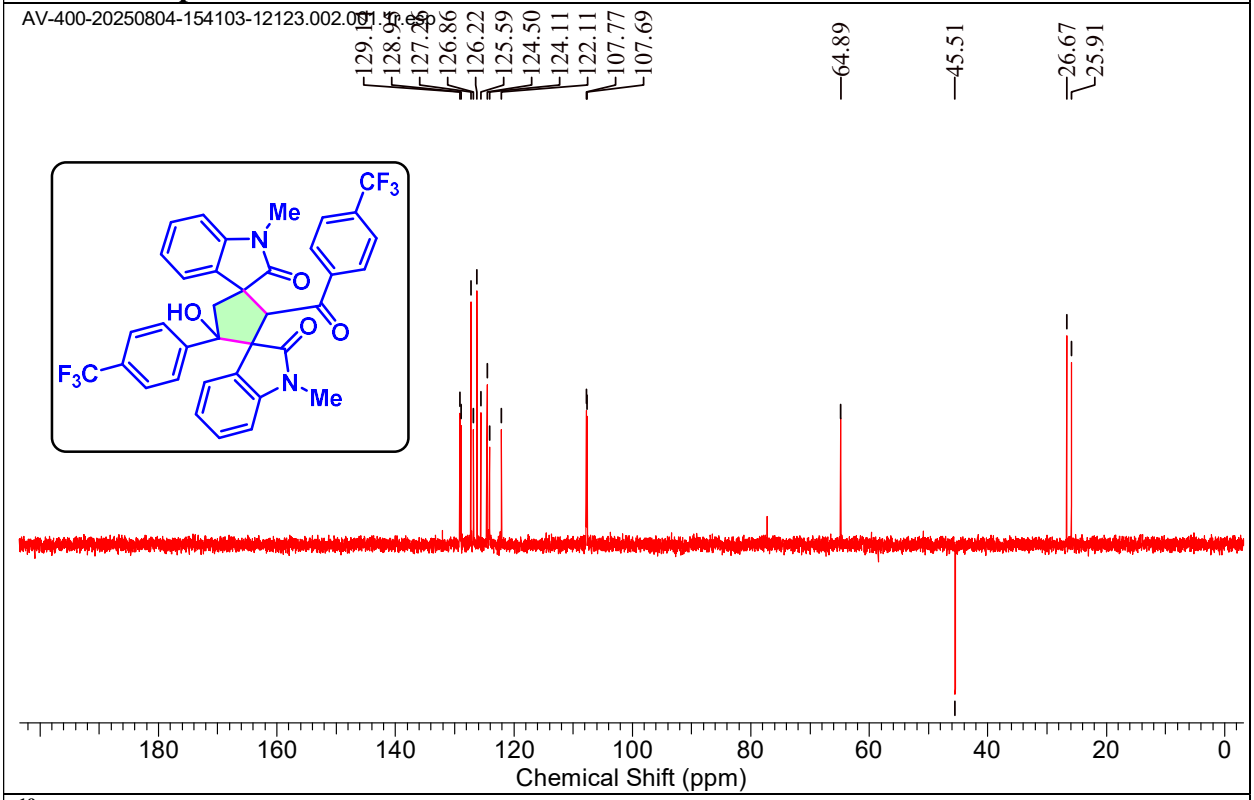
^{1}H NMR spectrum of 4Al



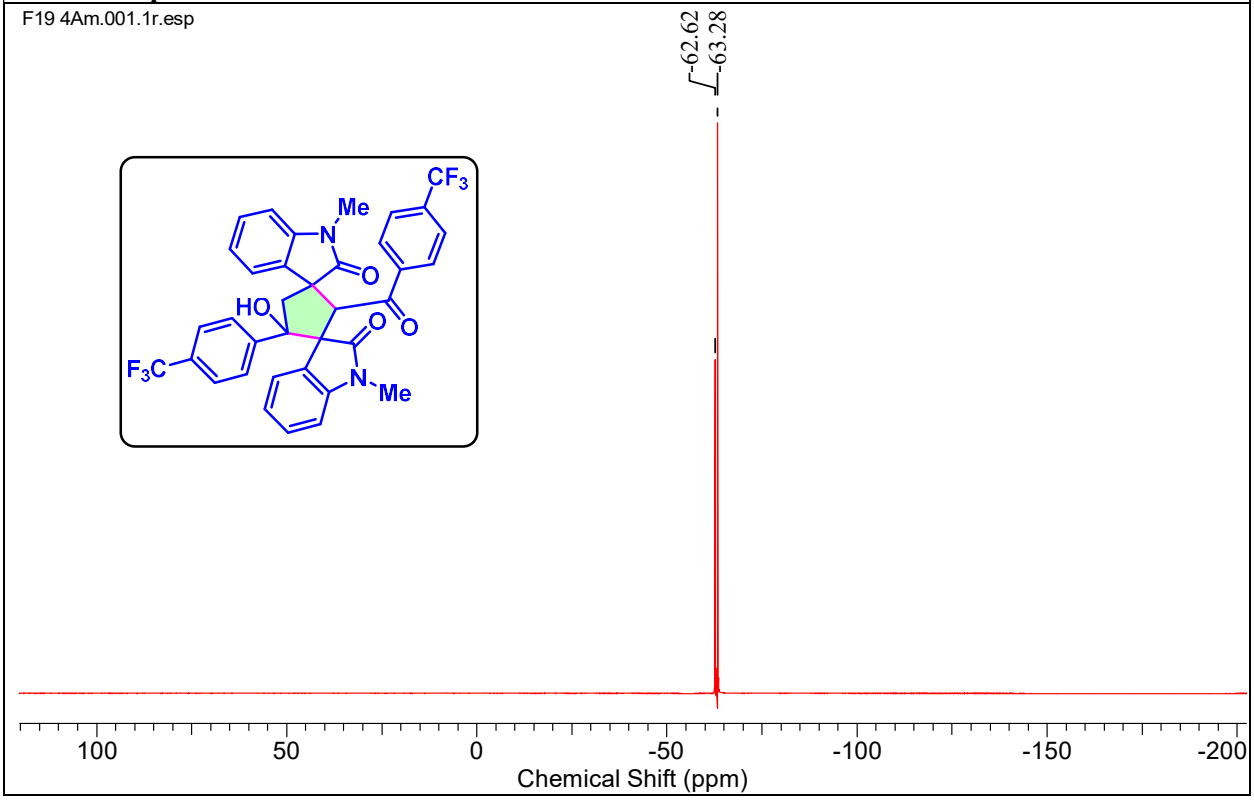
^{13}C NMR spectrum of 4Al



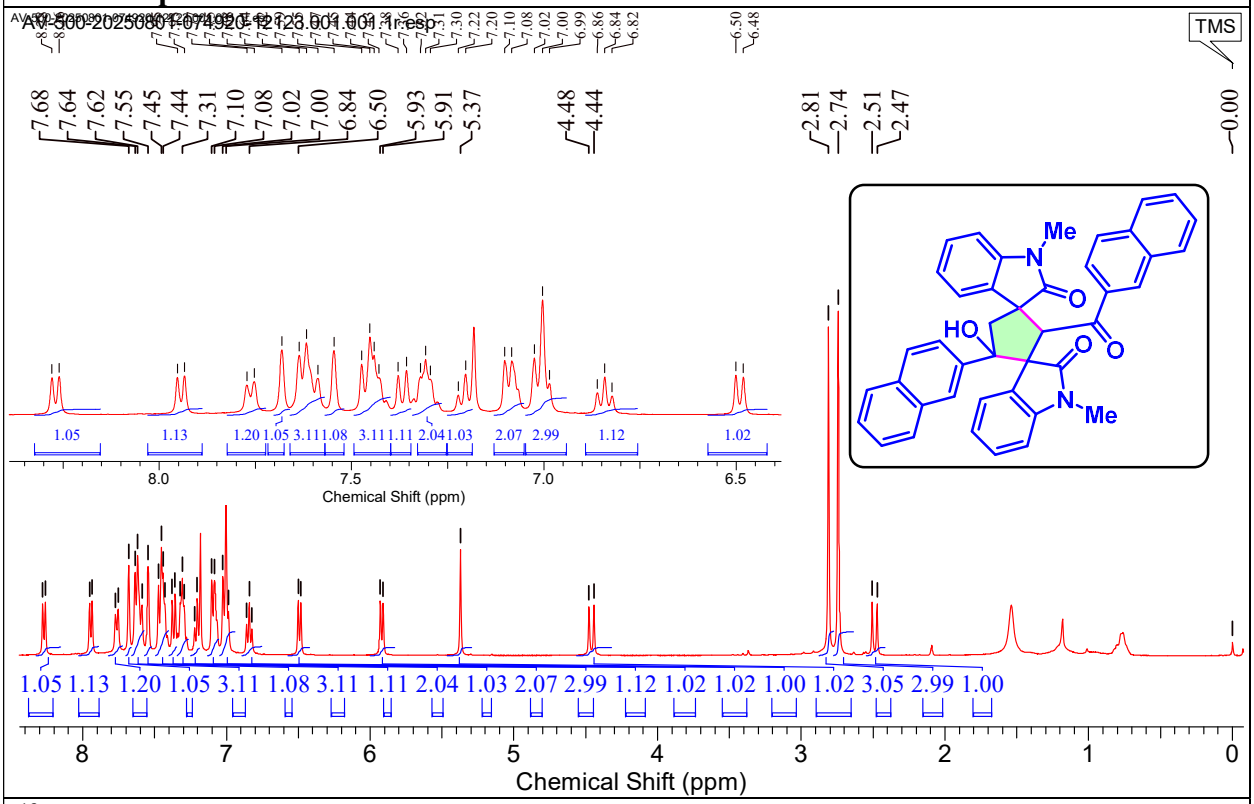
135 DEPT spectrum of 4Al



^{19}F NMR spectrum of 4Al



^{1}H NMR spectrum of 4An



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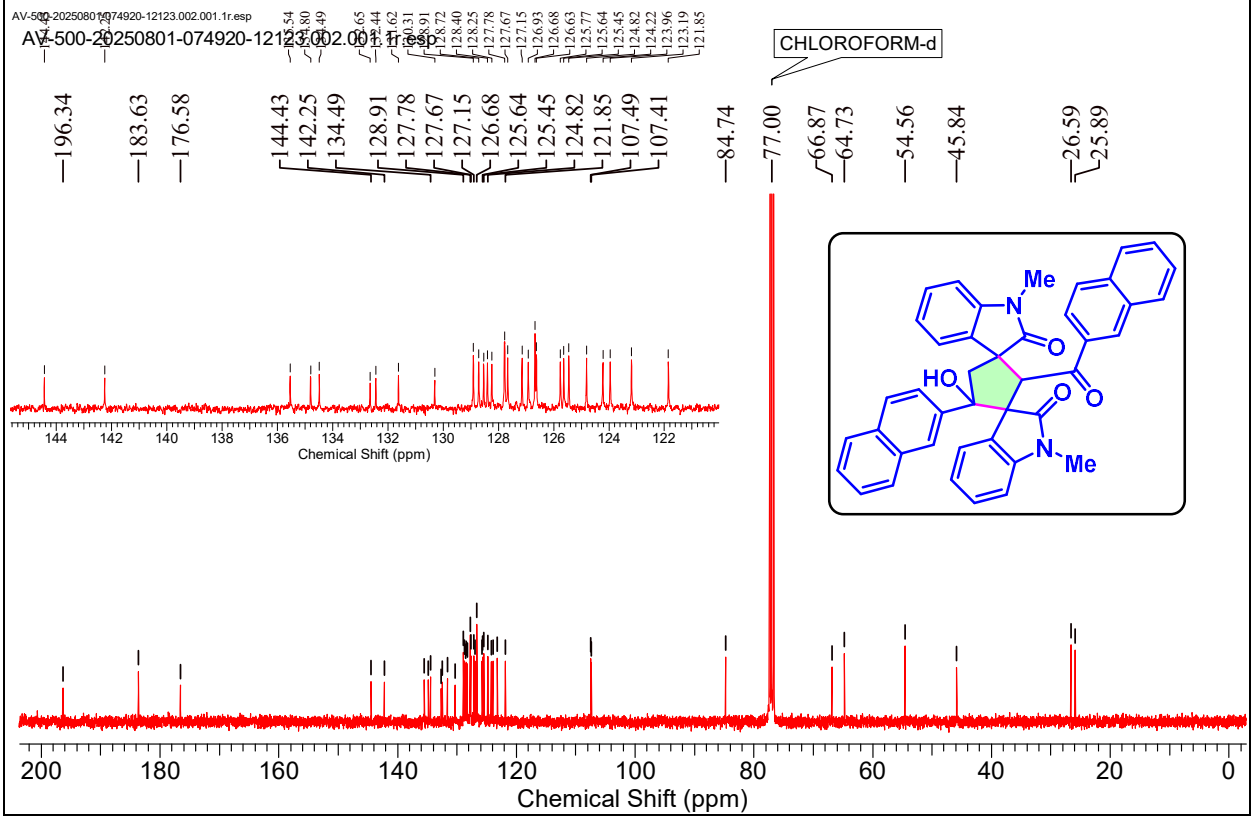
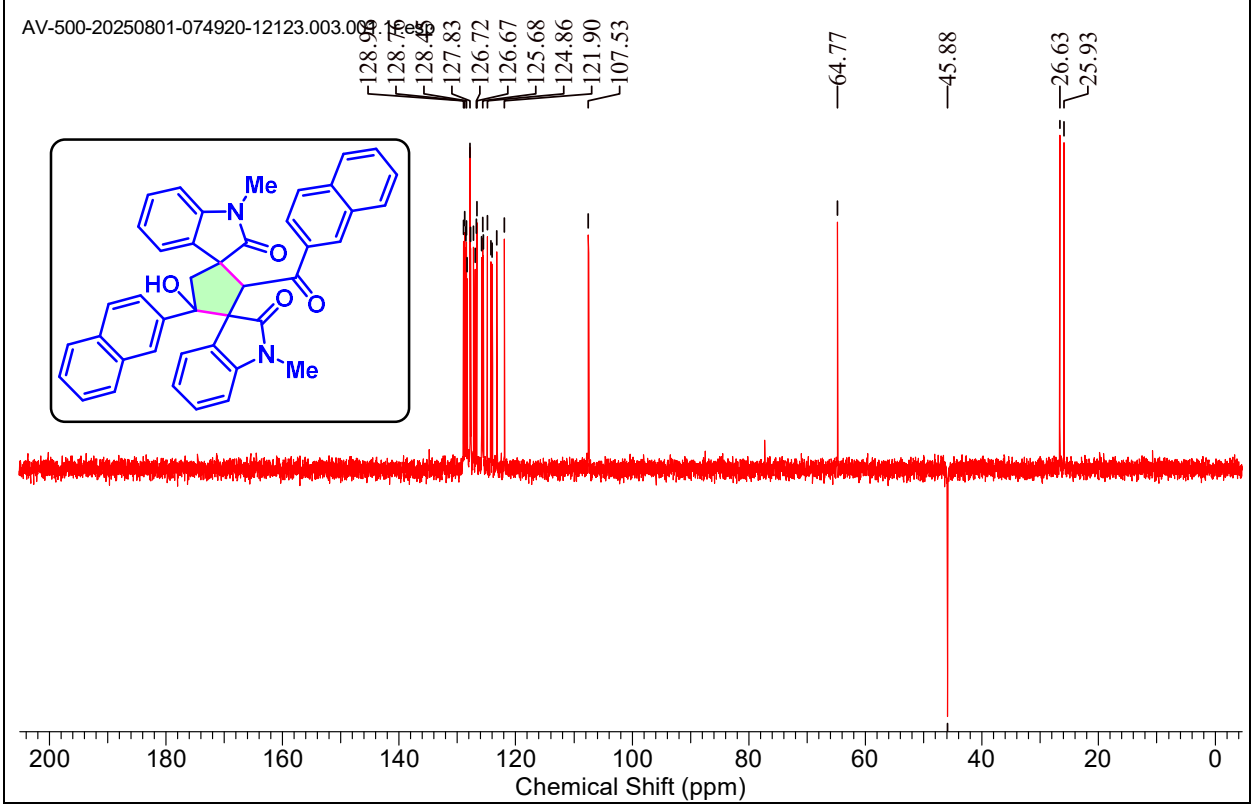
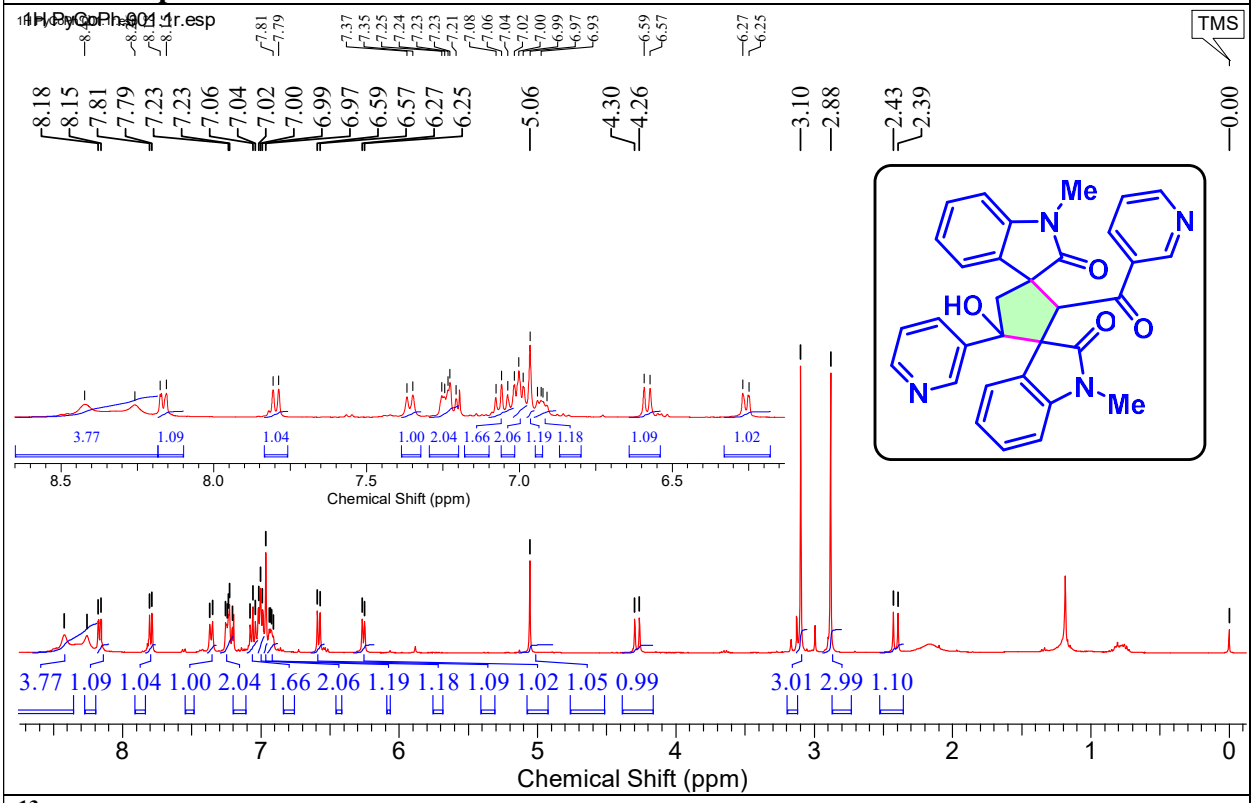


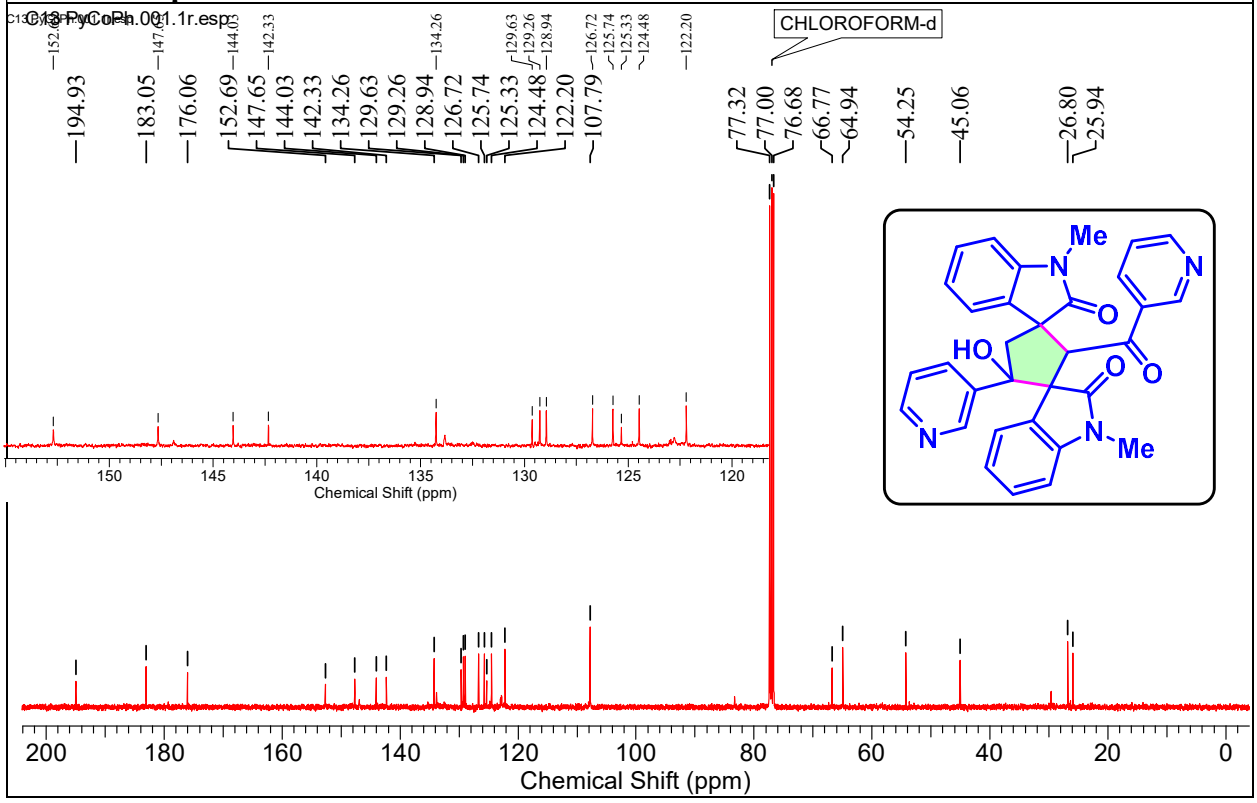
135 DEPT spectrum of 4An



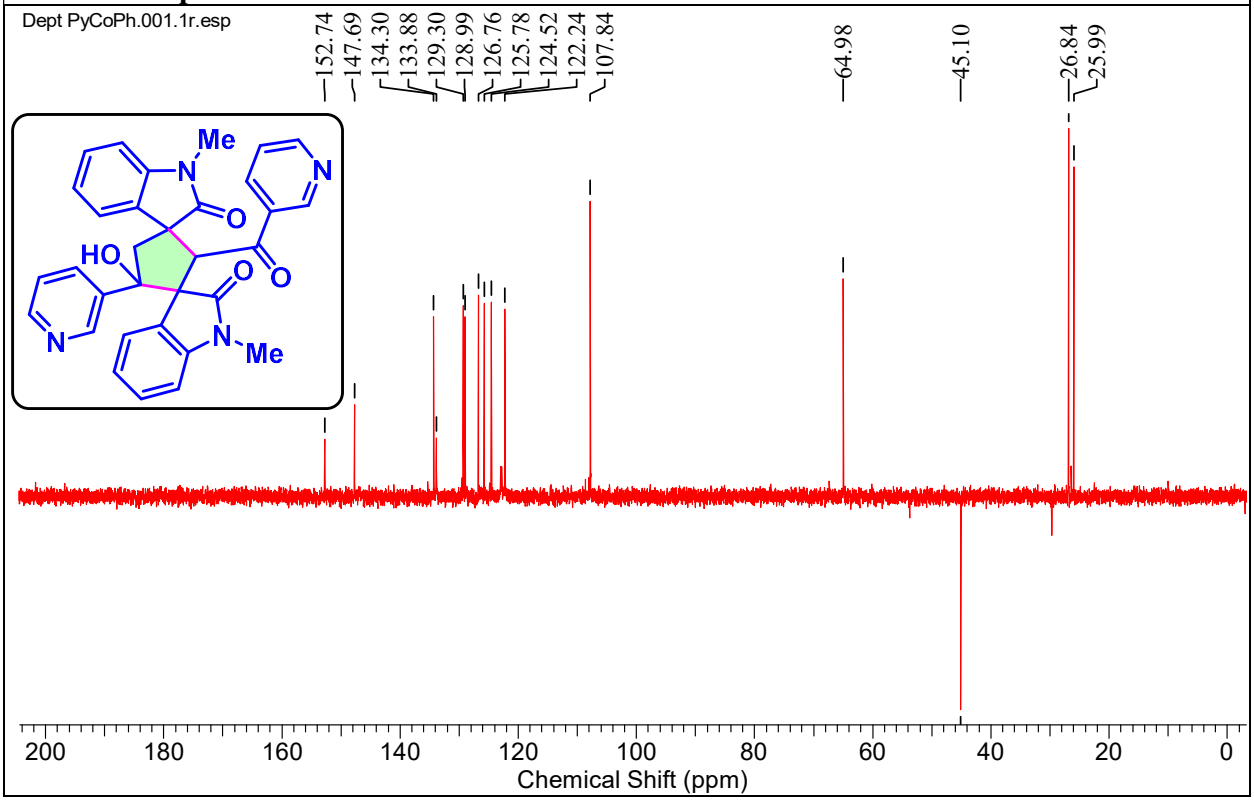
^{1}H NMR spectrum of 4Ao



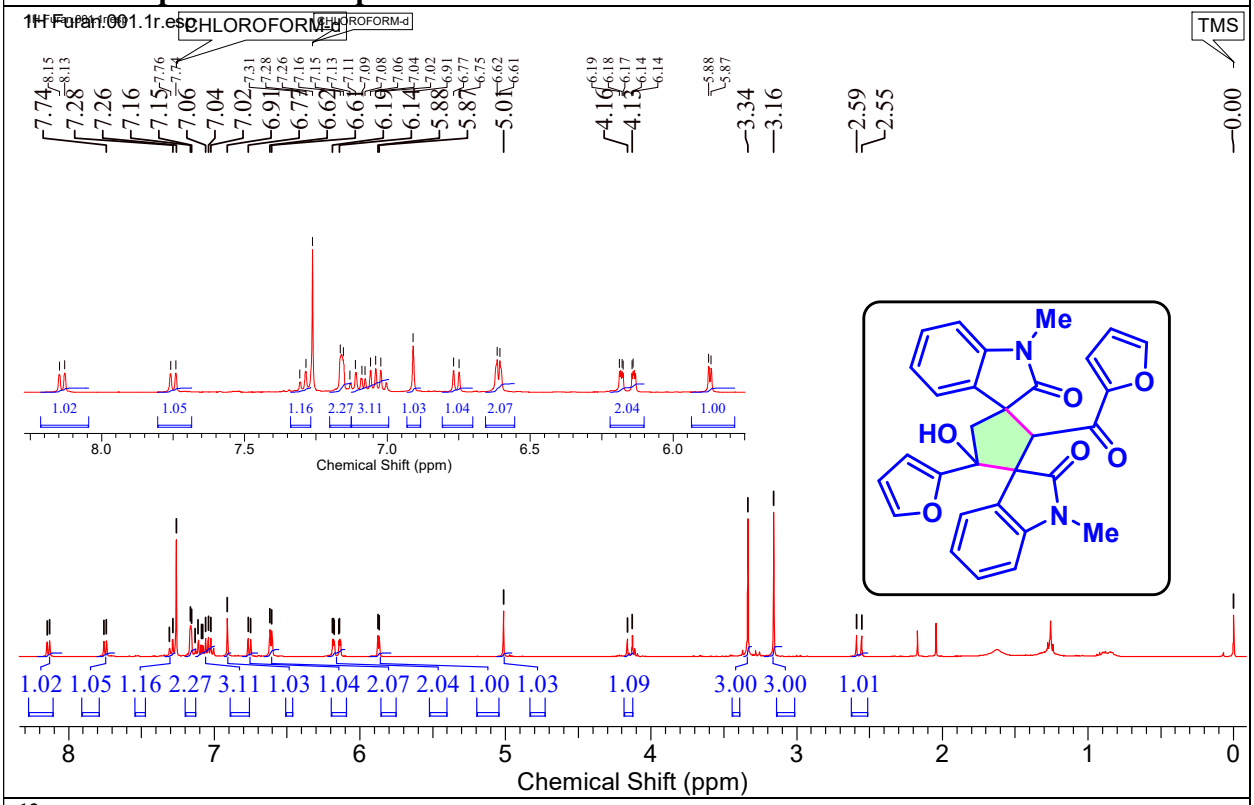
^{13}C NMR spectrum of 4Ao



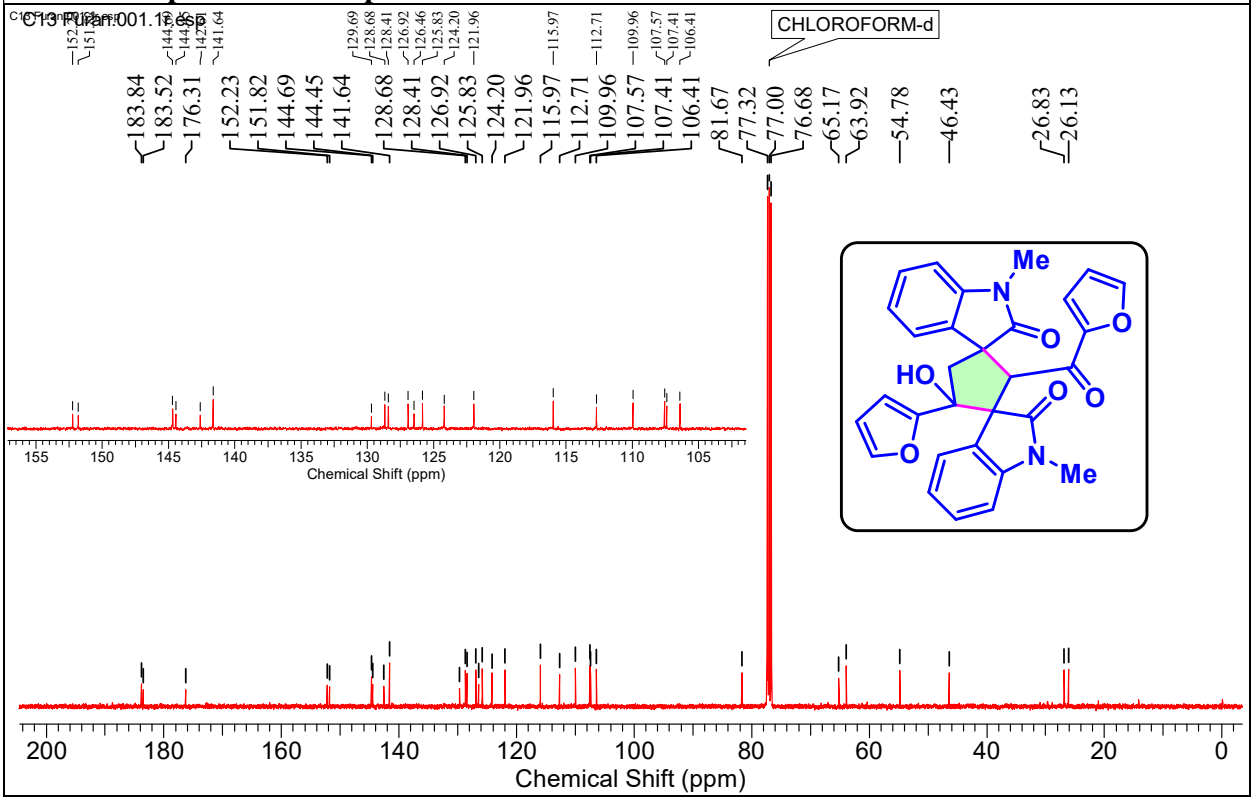
135 DEPT spectrum of 4Ao



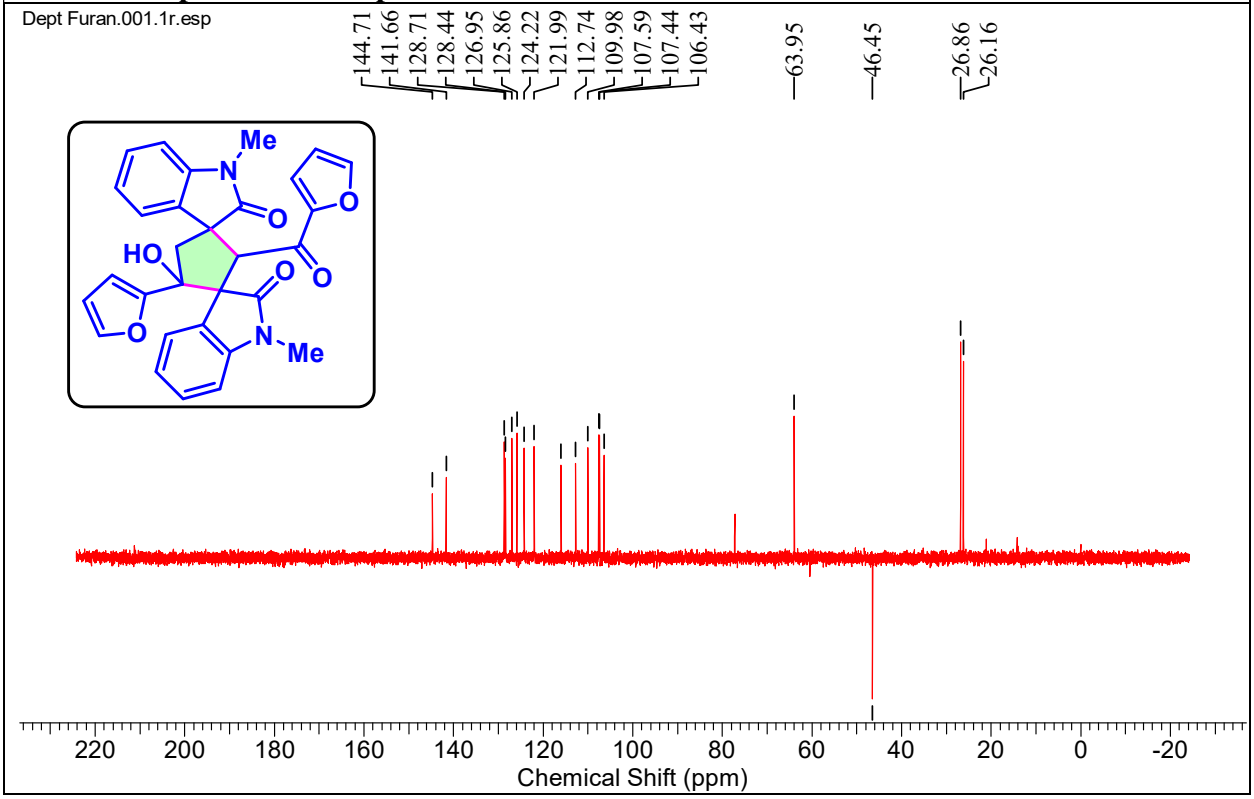
^{1}H NMR spectrum of 4Ap



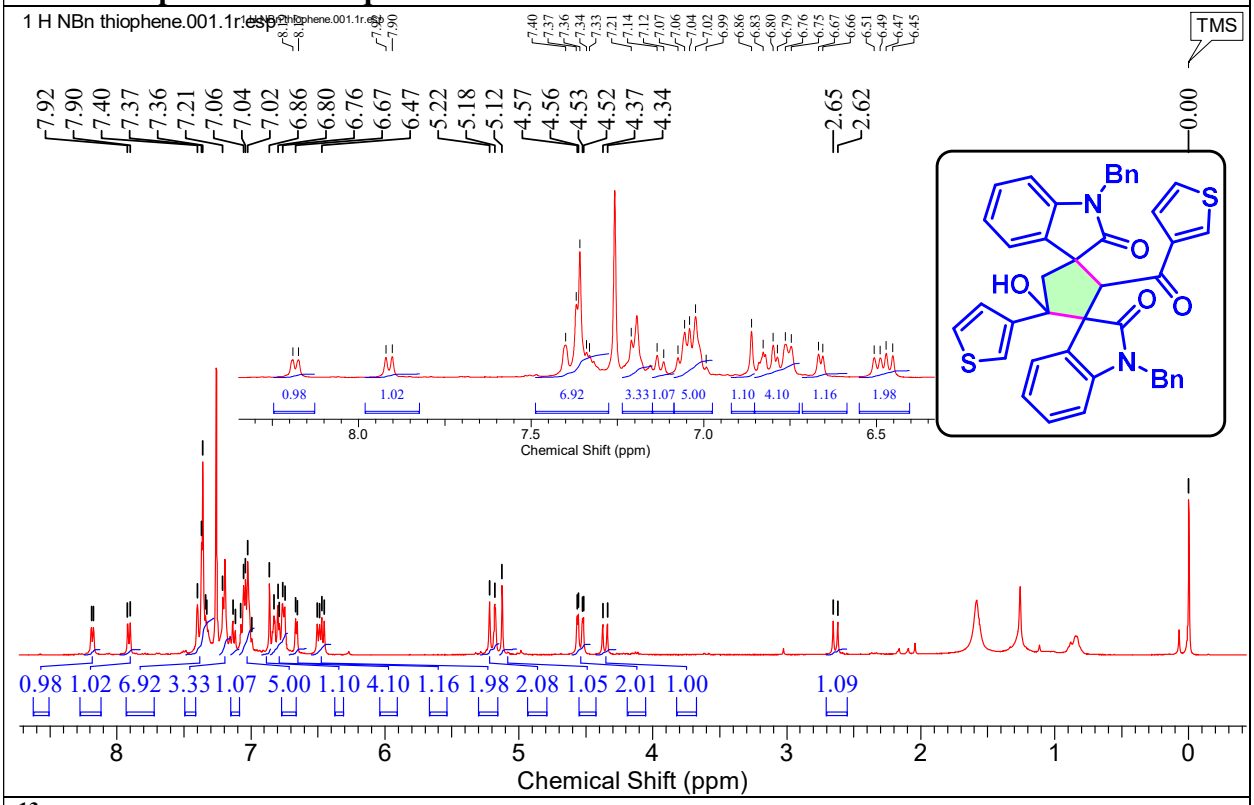
^{13}C NMR spectrum of 4Ap



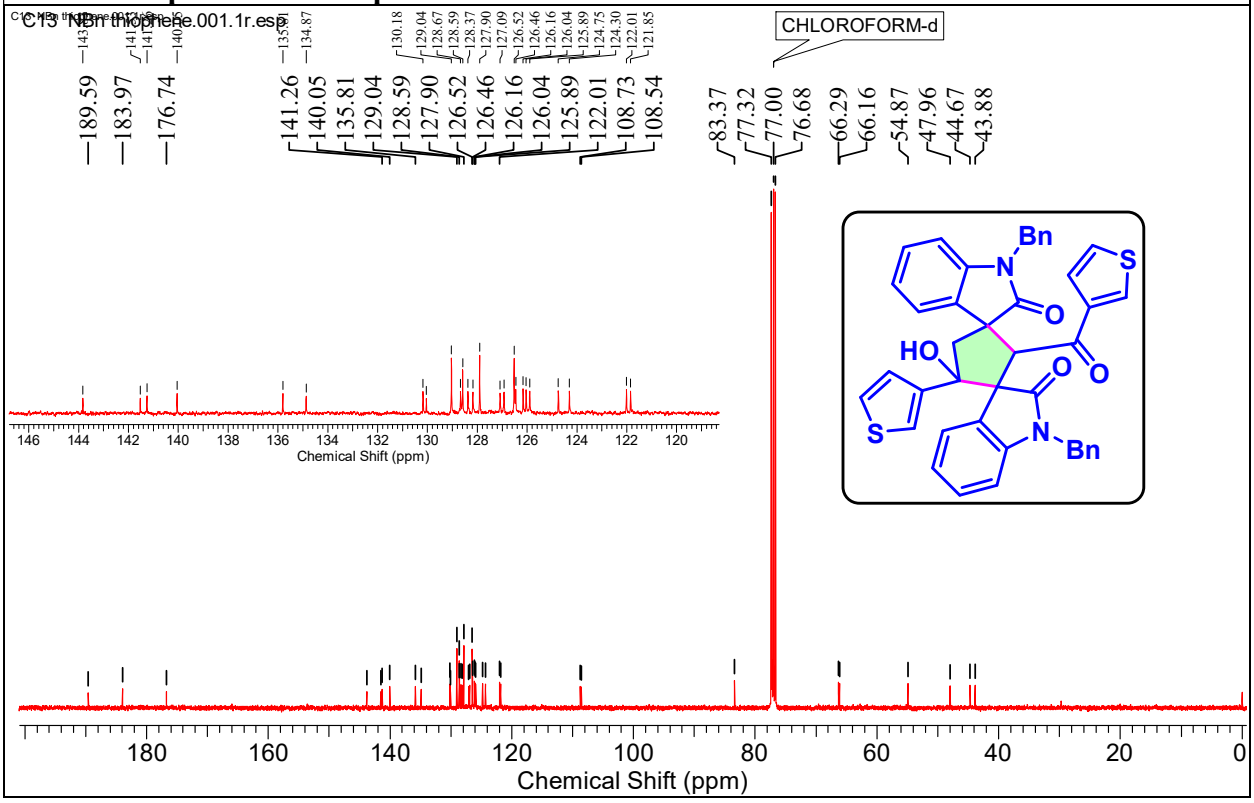
135 DEPT spectrum of 4Ap



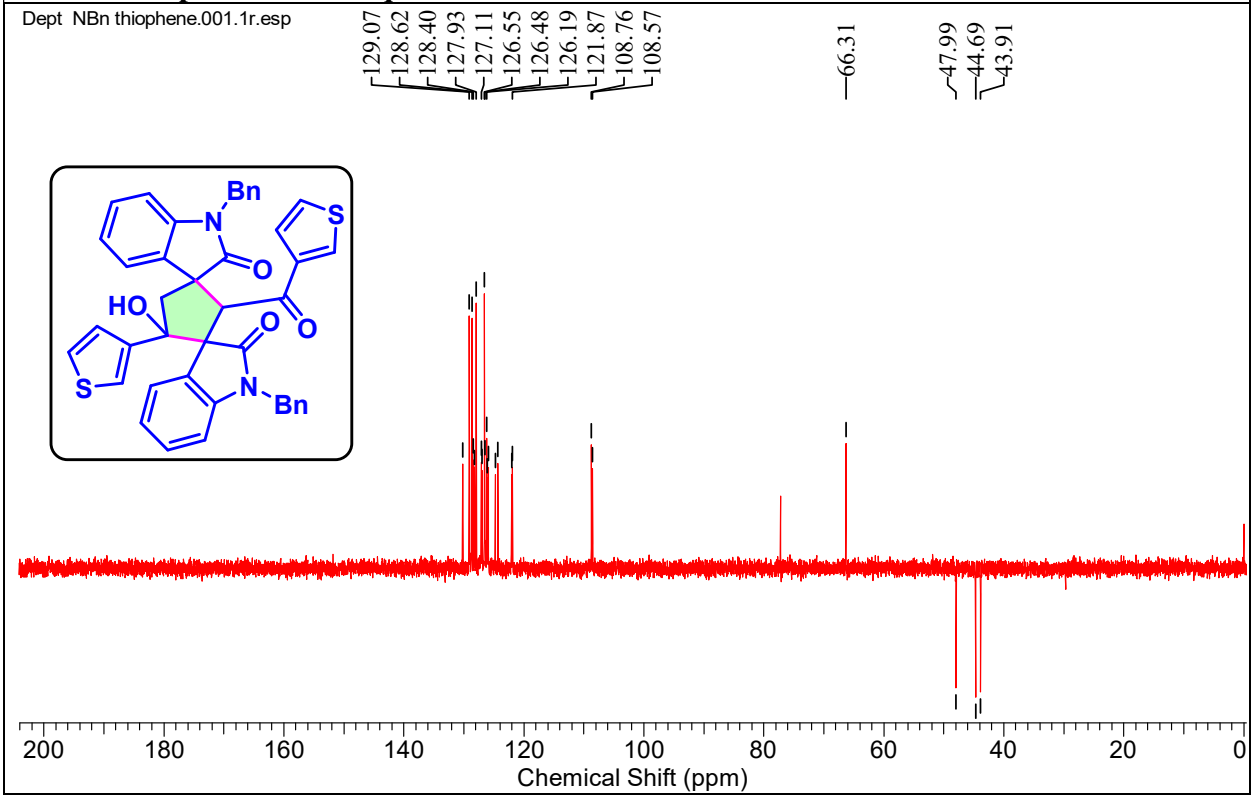
^{1}H NMR spectrum of 4Aq



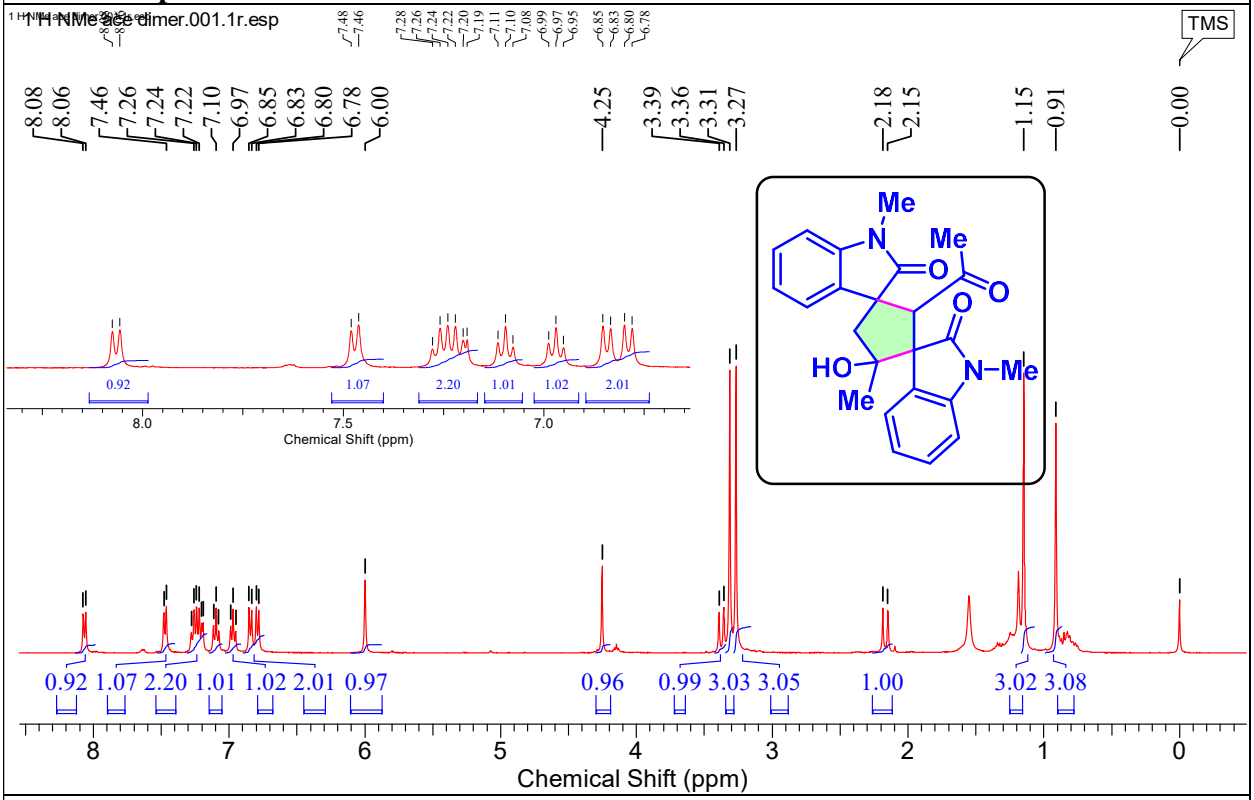
^{13}C NMR spectrum of 4Aq



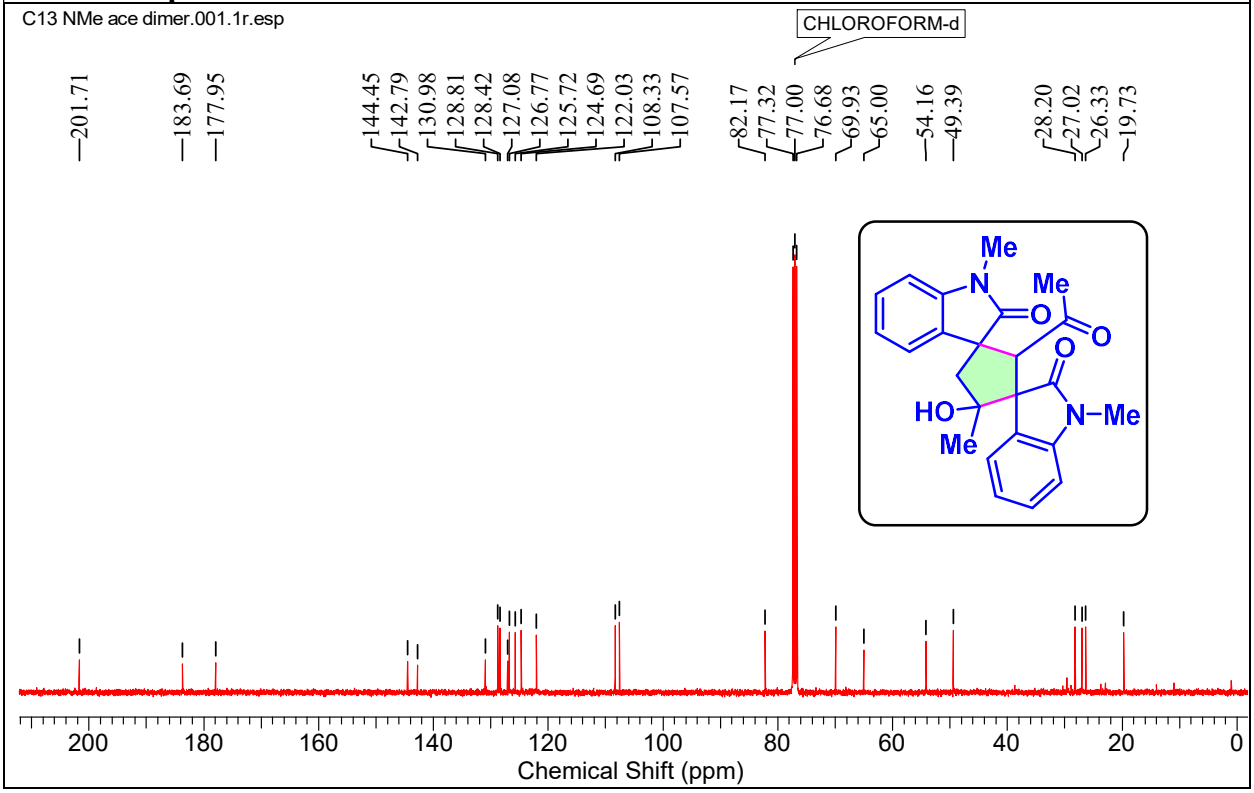
135 DEPT spectrum of 4Aq



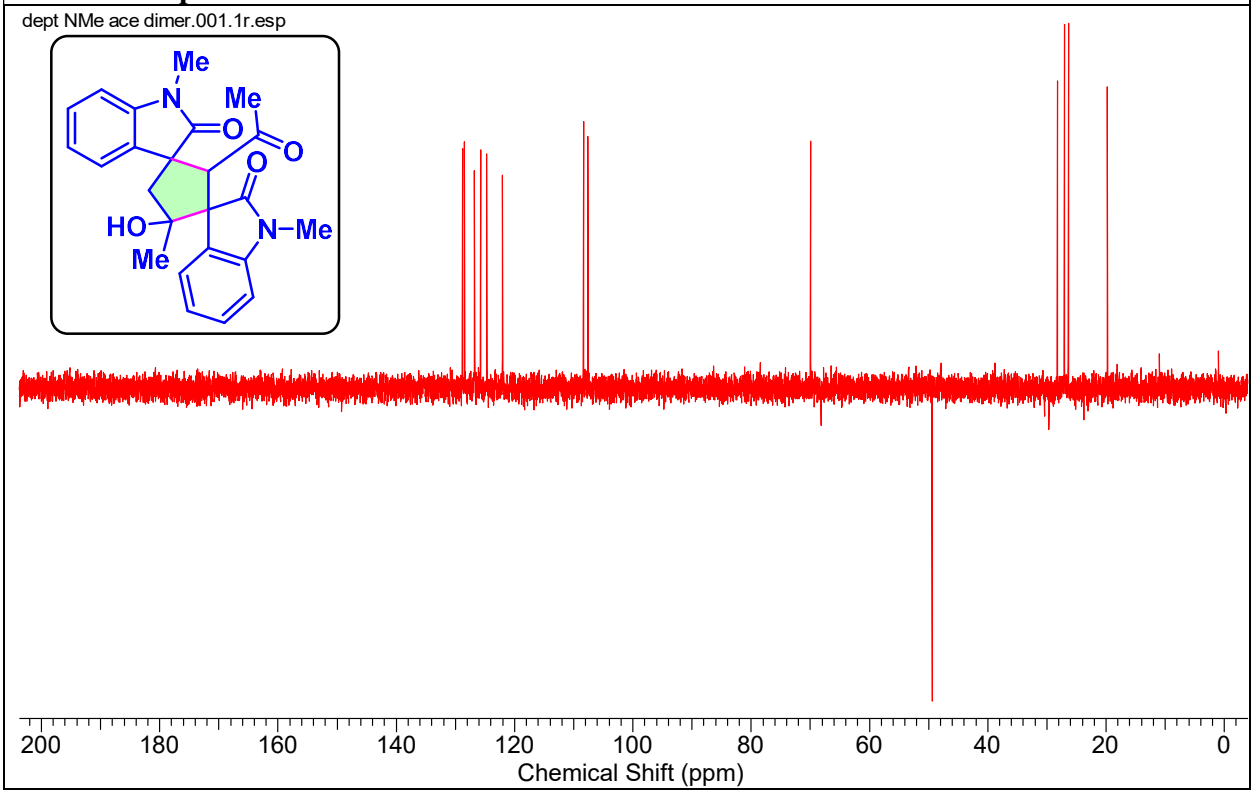
^{1}H NMR spectrum of 4Ba



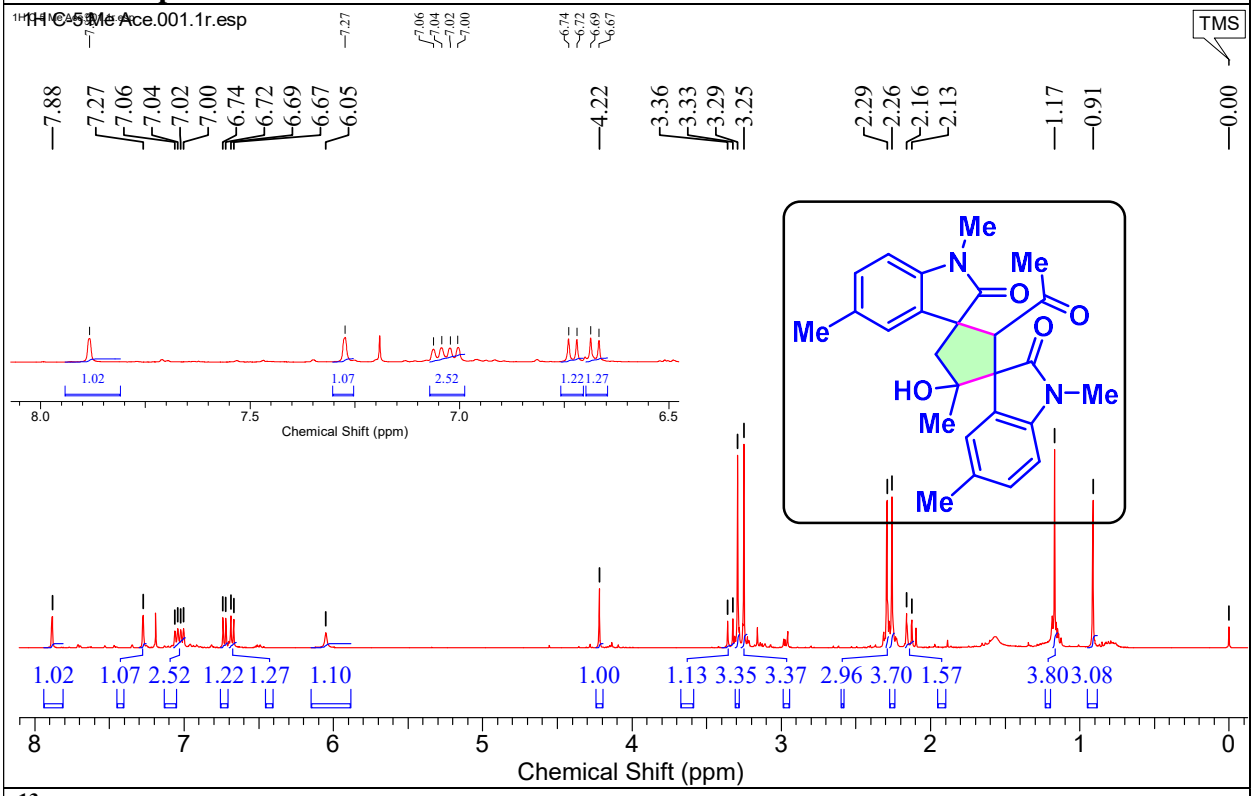
^{13}C NMR spectrum of 4Ba



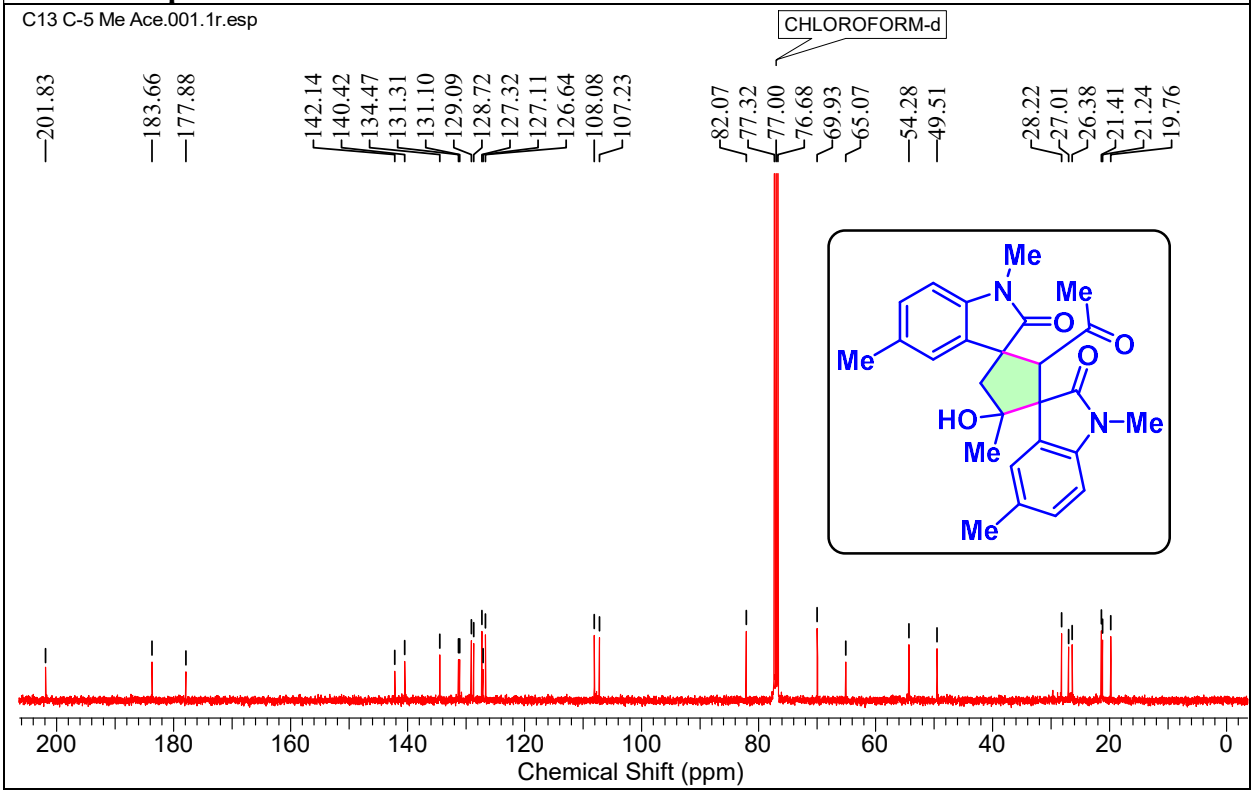
135 DEPT spectrum of 4Ba



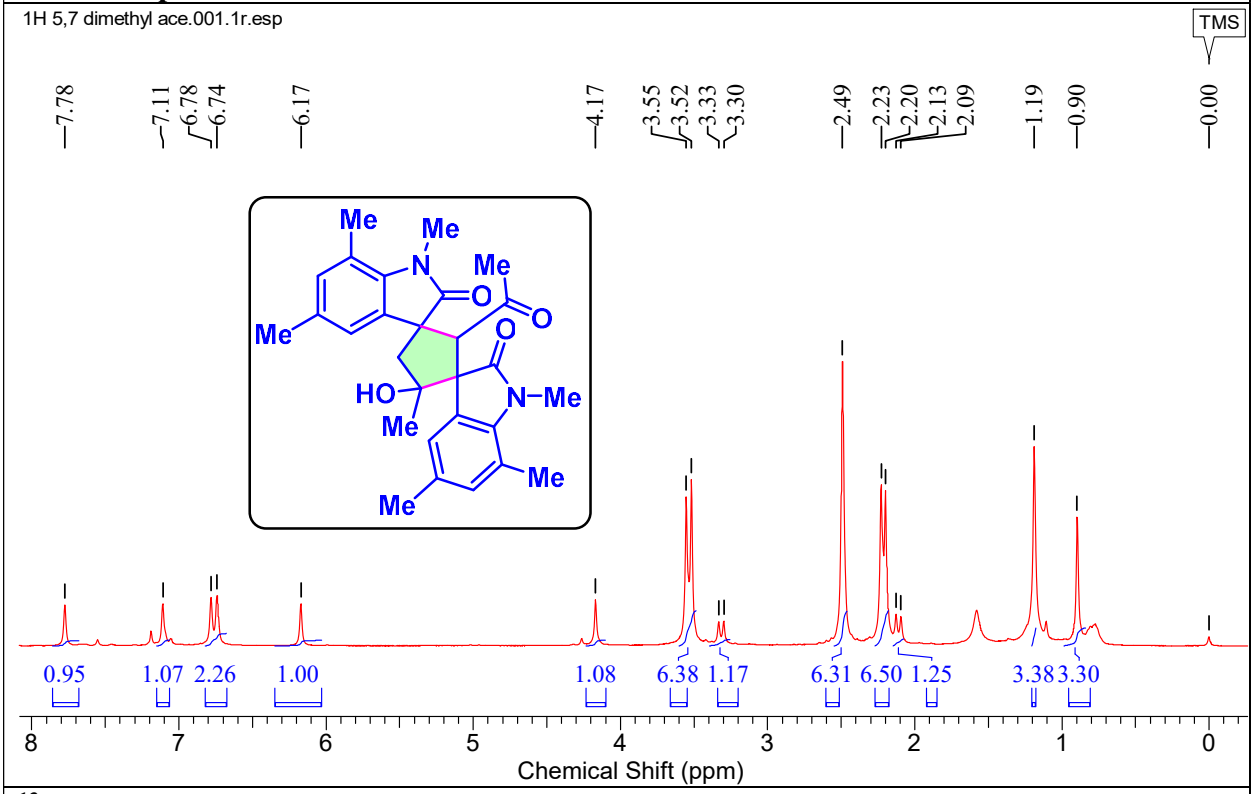
^{1}H NMR spectrum of 4Bb



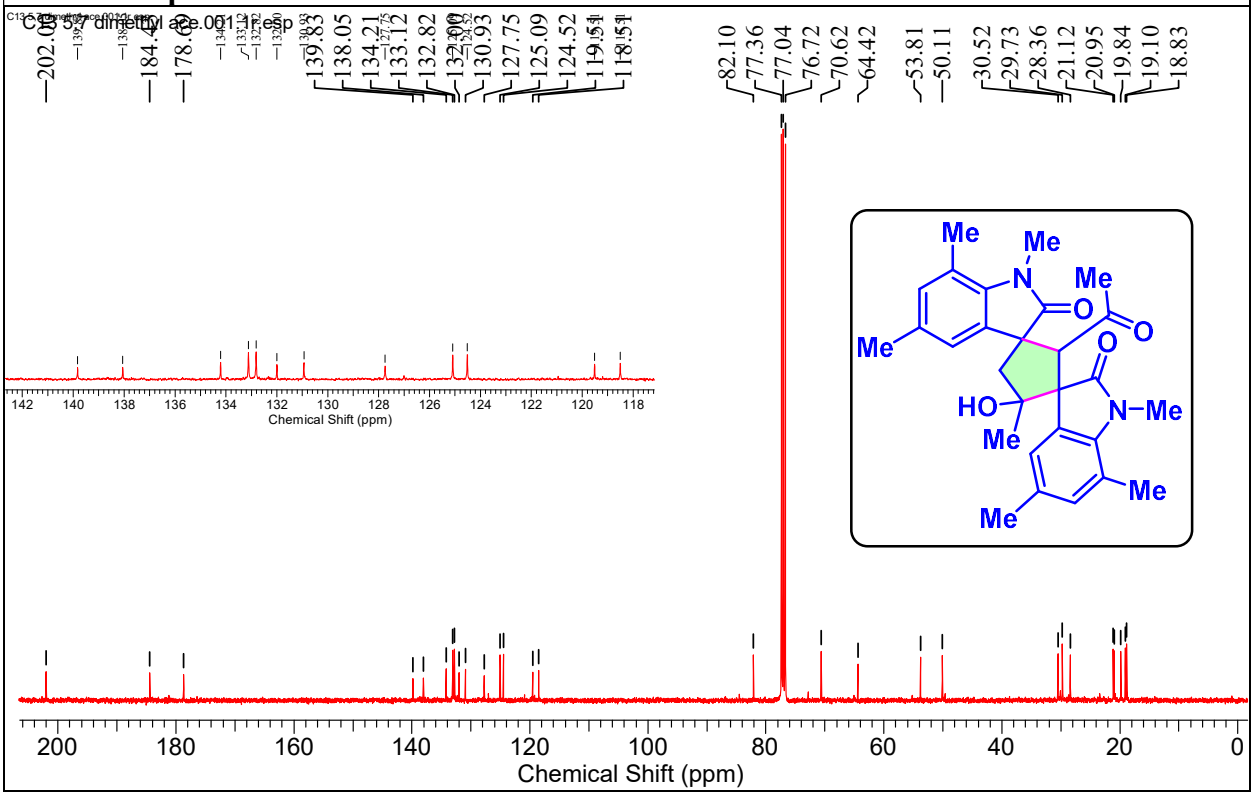
^{13}C NMR spectrum of 4Bb



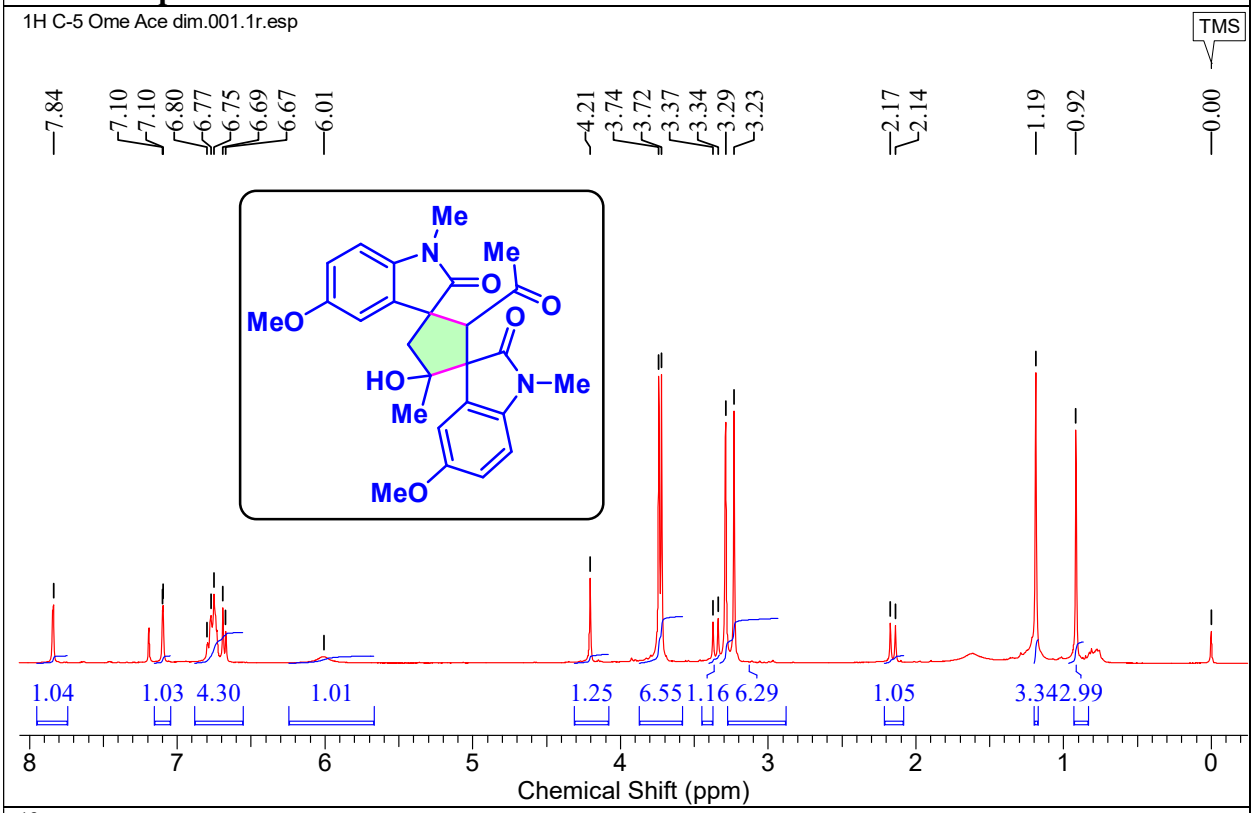
^{1}H NMR spectrum of 4Bc



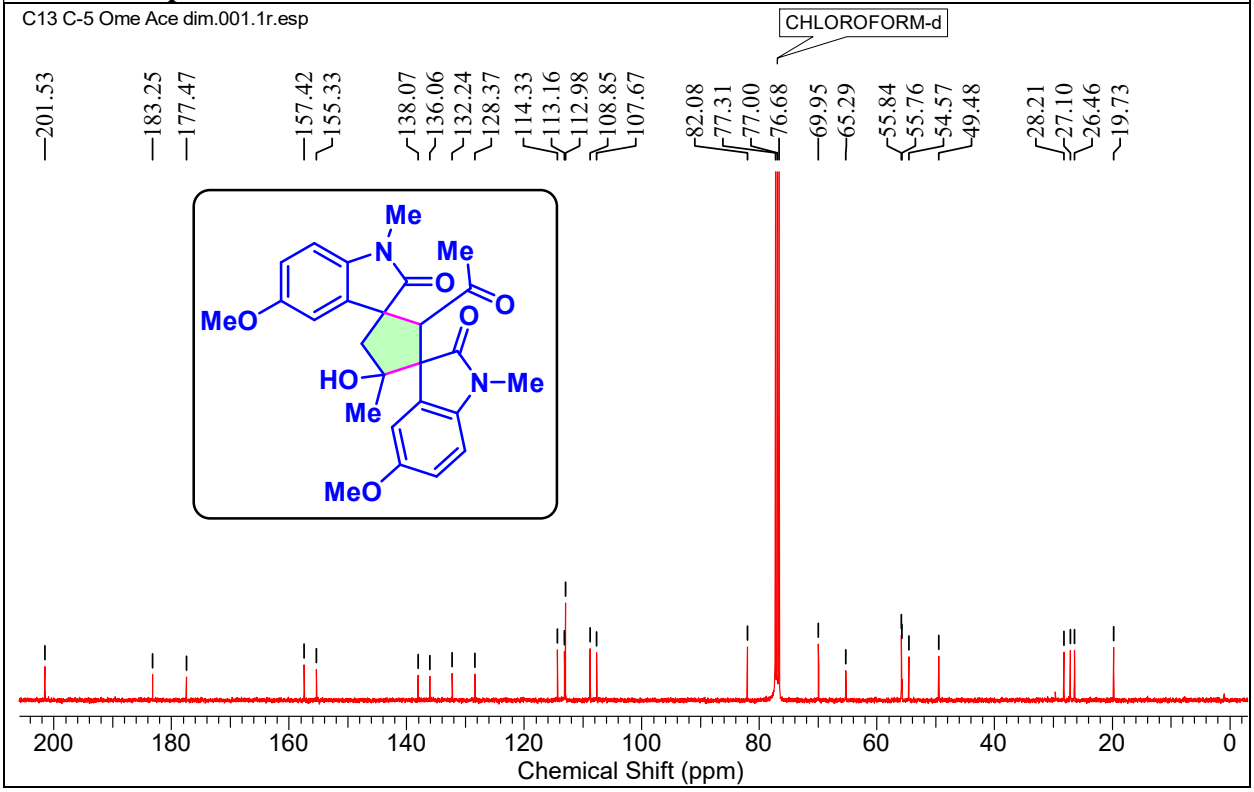
^{13}C NMR spectrum of 4Bc



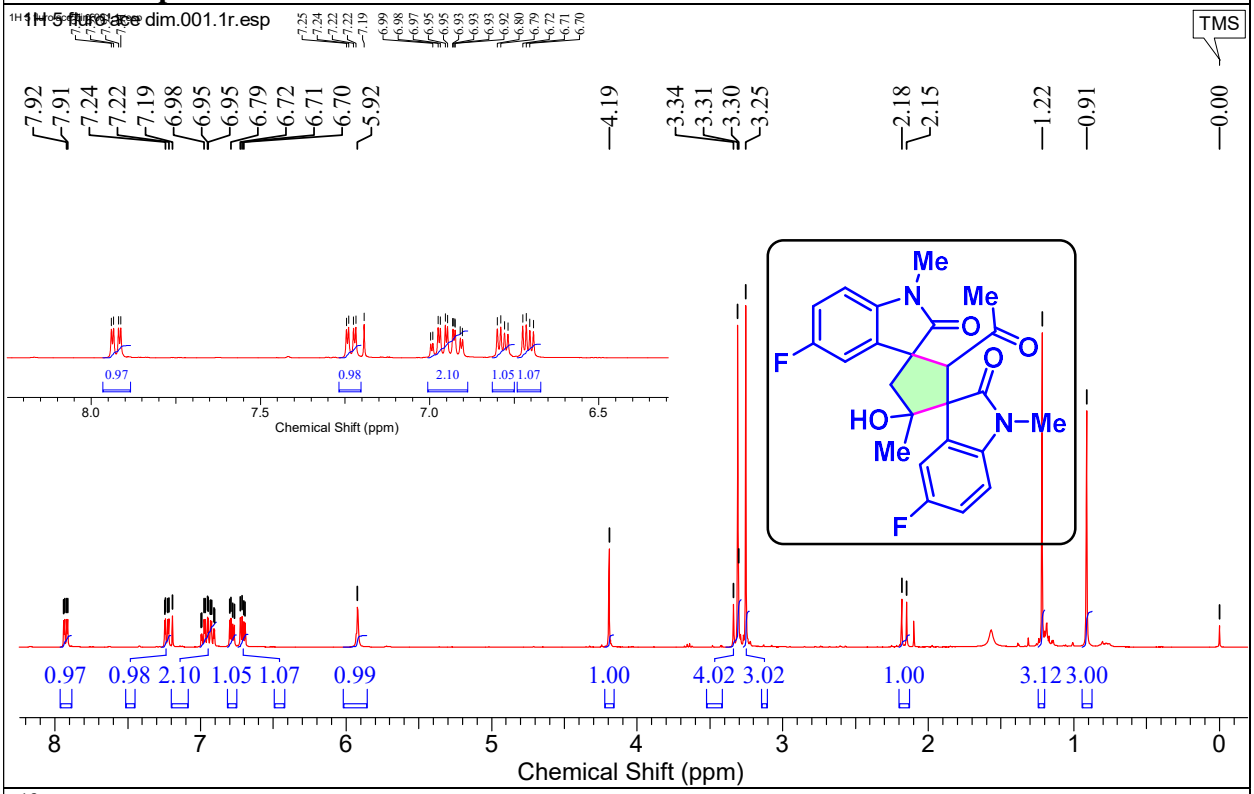
^{1}H NMR spectrum of 4Bd



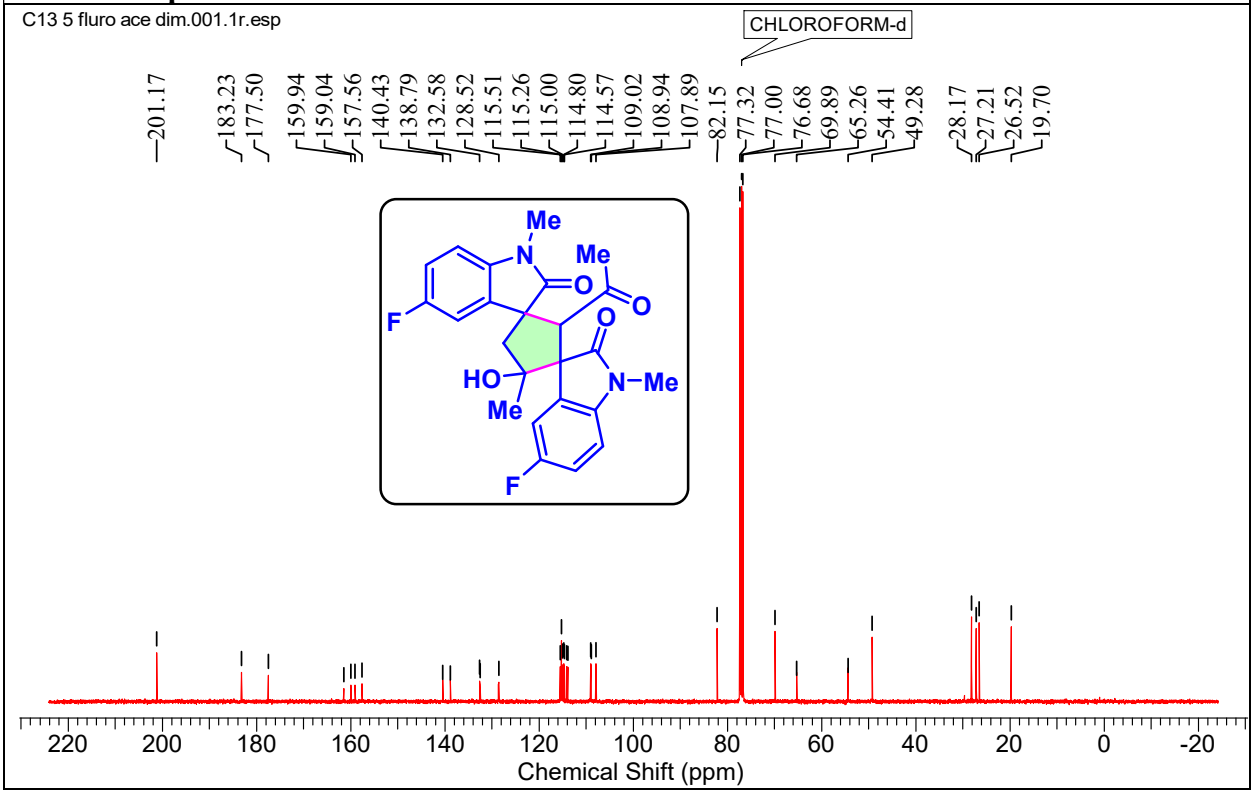
^{13}C NMR spectrum of 4Bd



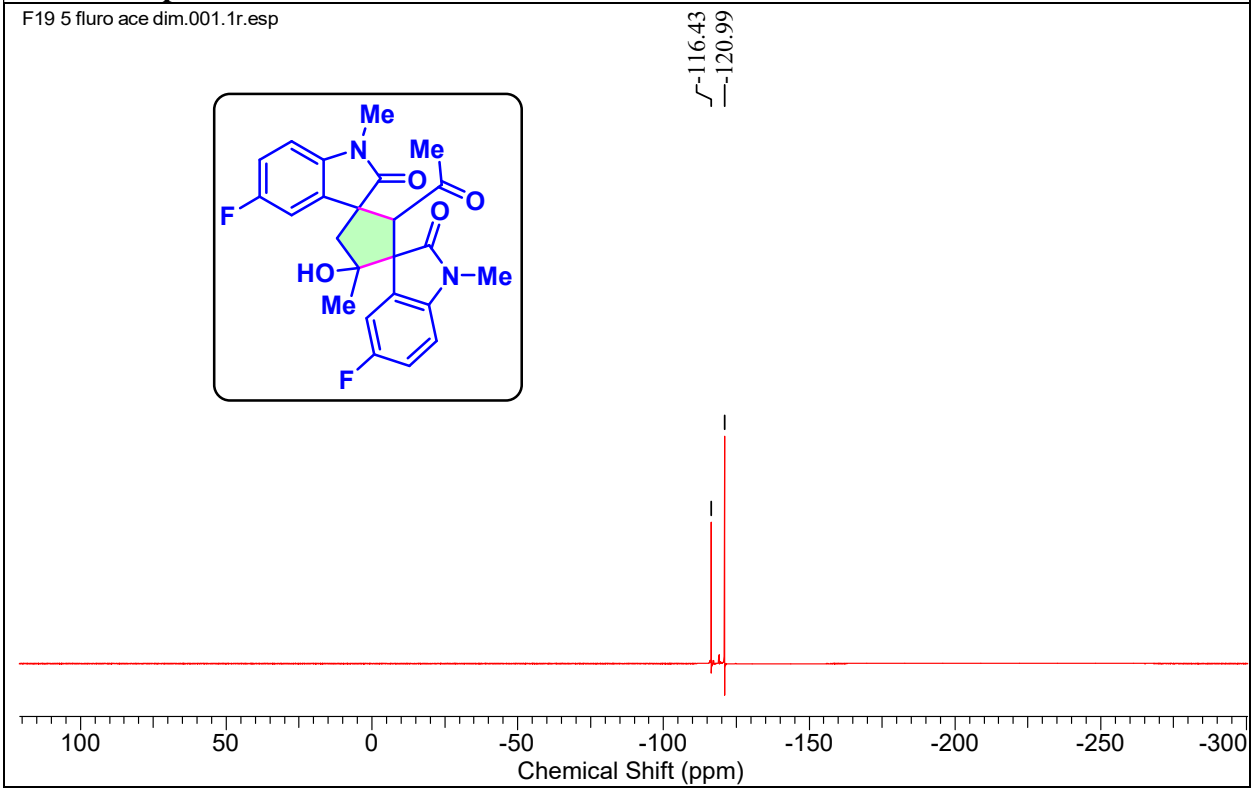
^{1}H NMR spectrum of 4Be



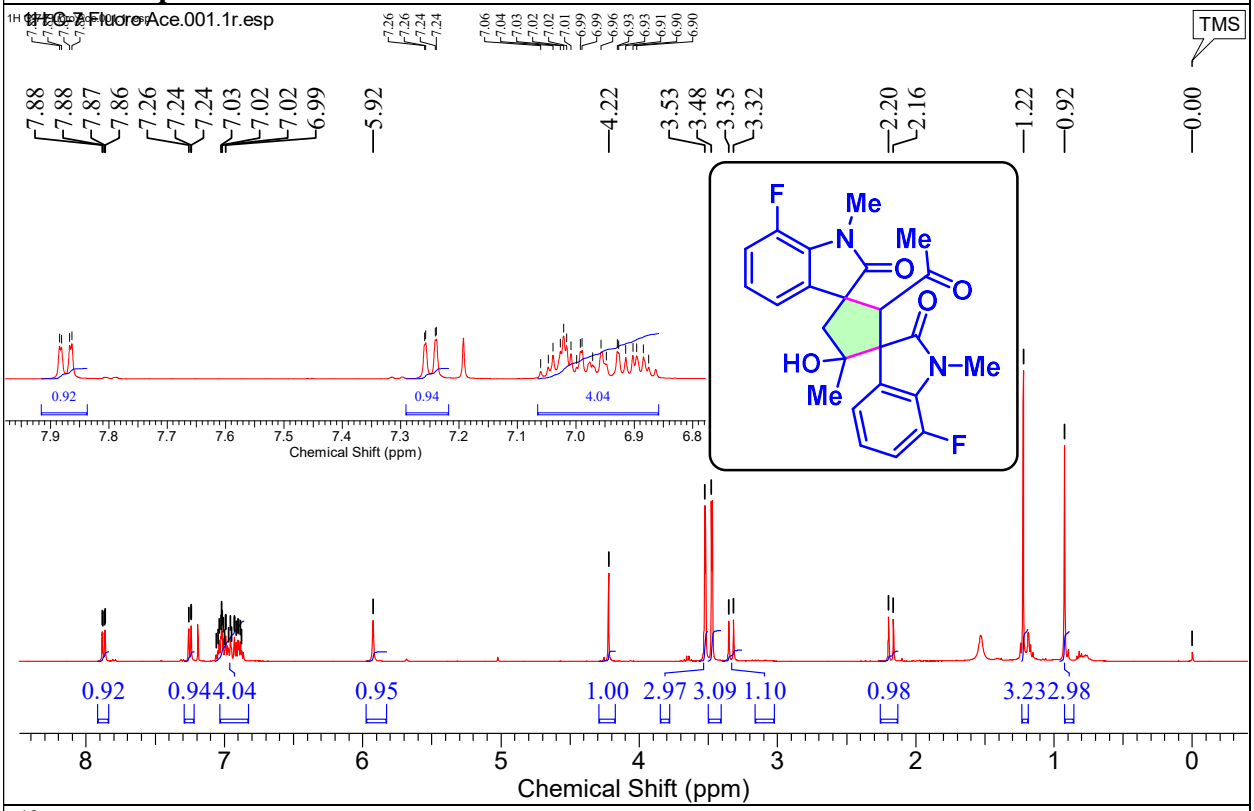
^{13}C NMR spectrum of 4Be



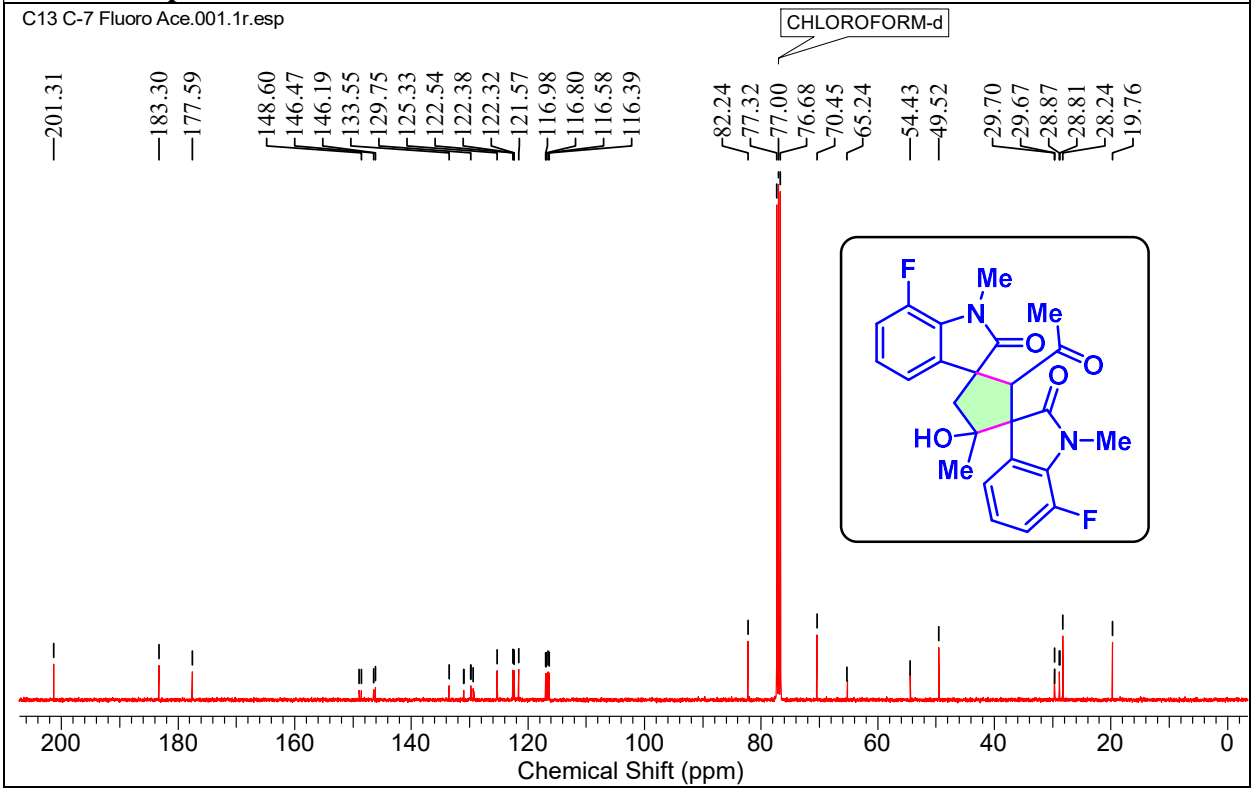
^{19}F NMR spectrum of 4Be



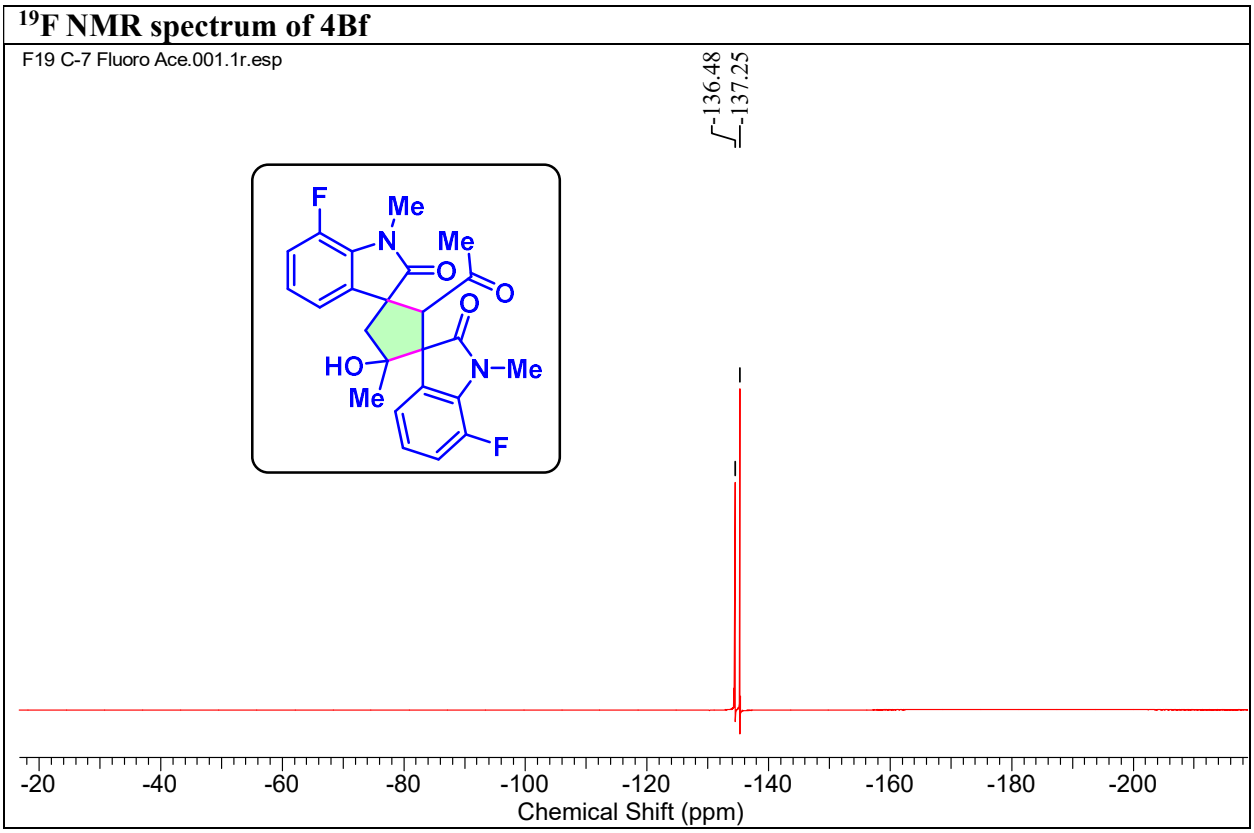
^{1}H NMR spectrum of 4Bf



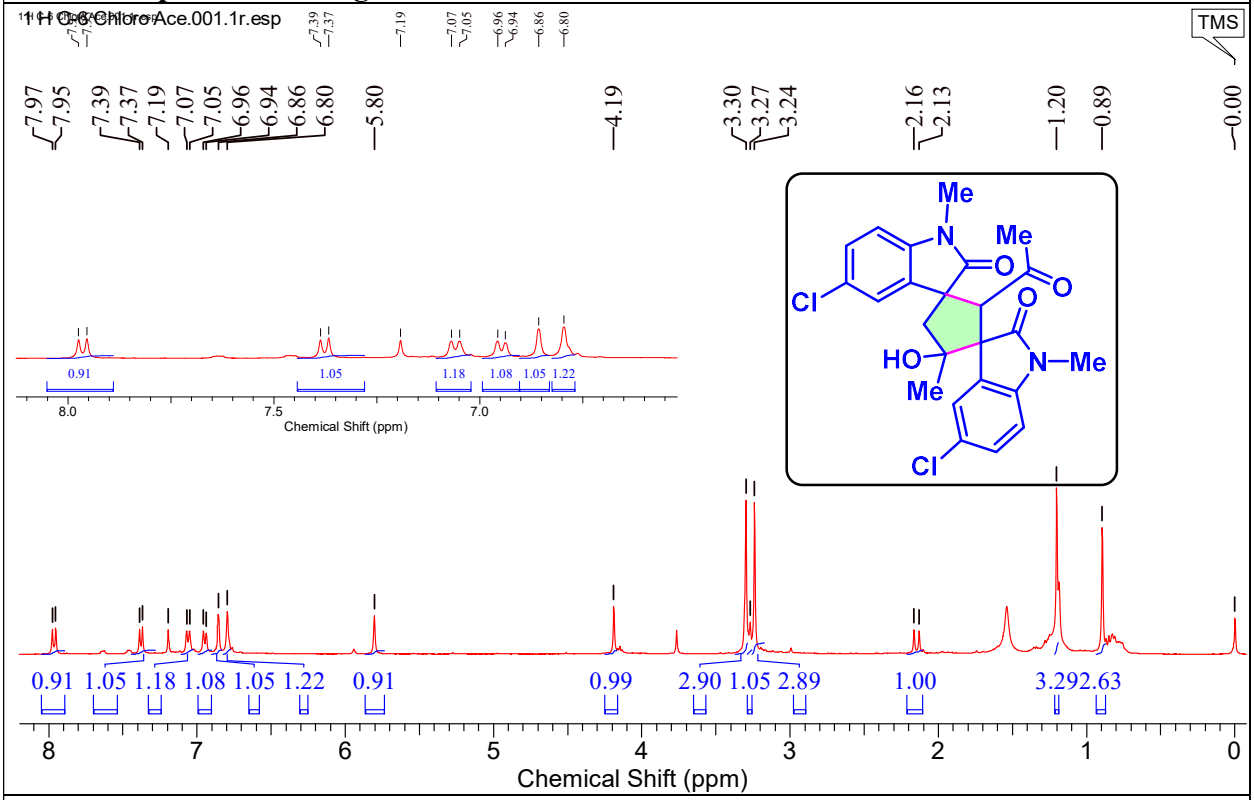
^{13}C NMR spectrum of 4Bf



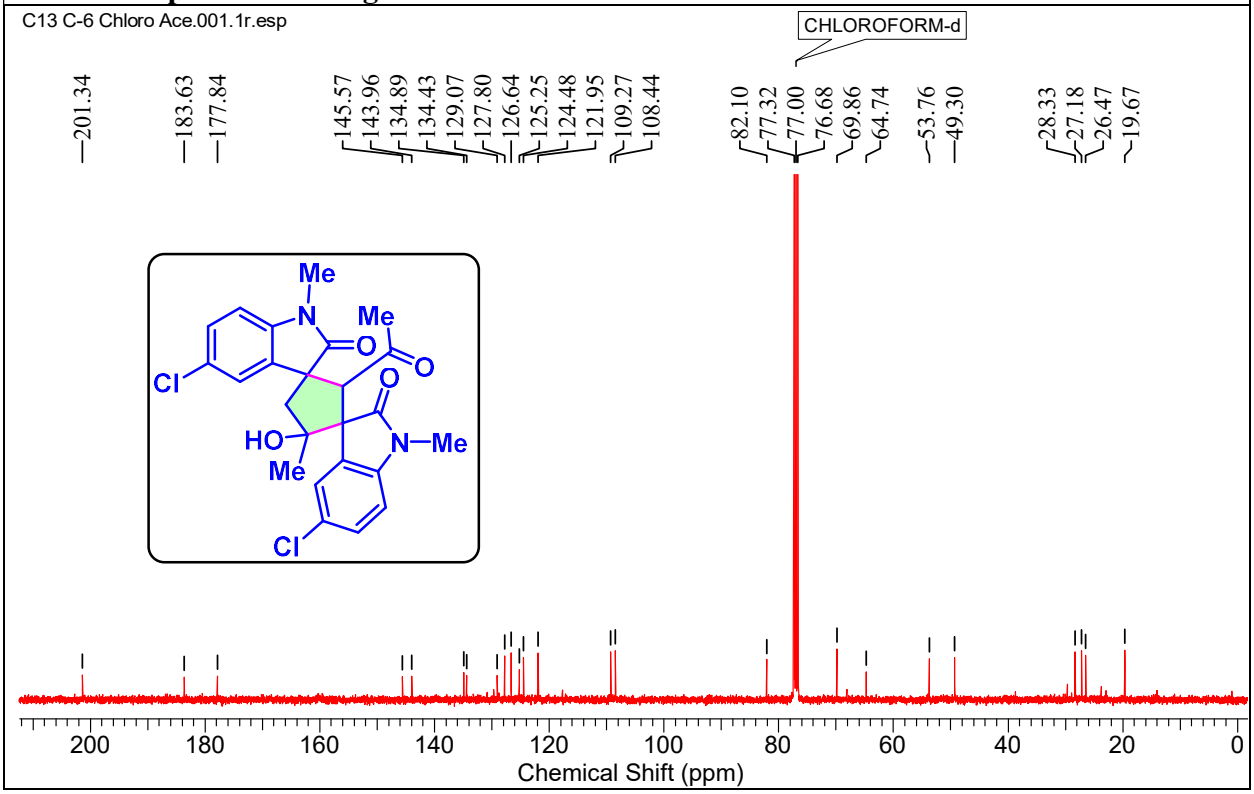
^{19}F NMR spectrum of 4Bf



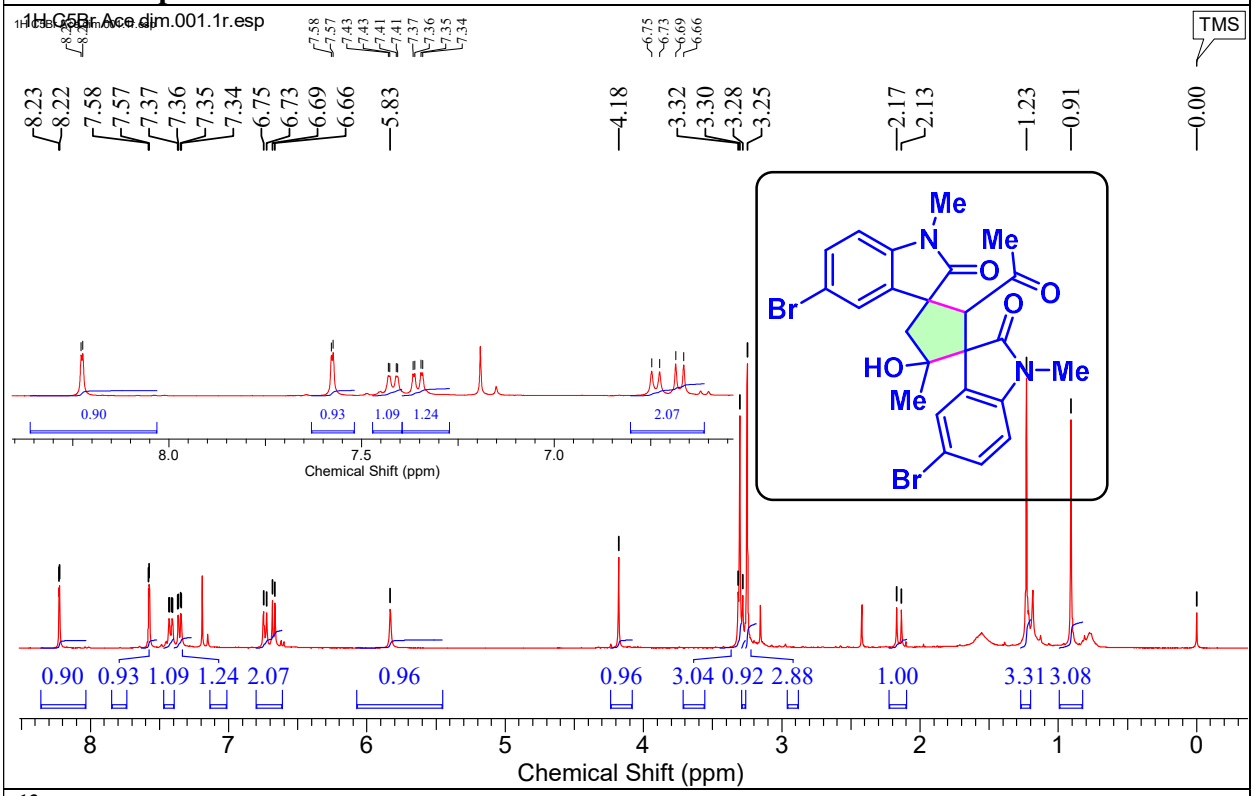
^{1}H NMR spectrum of 4Bg



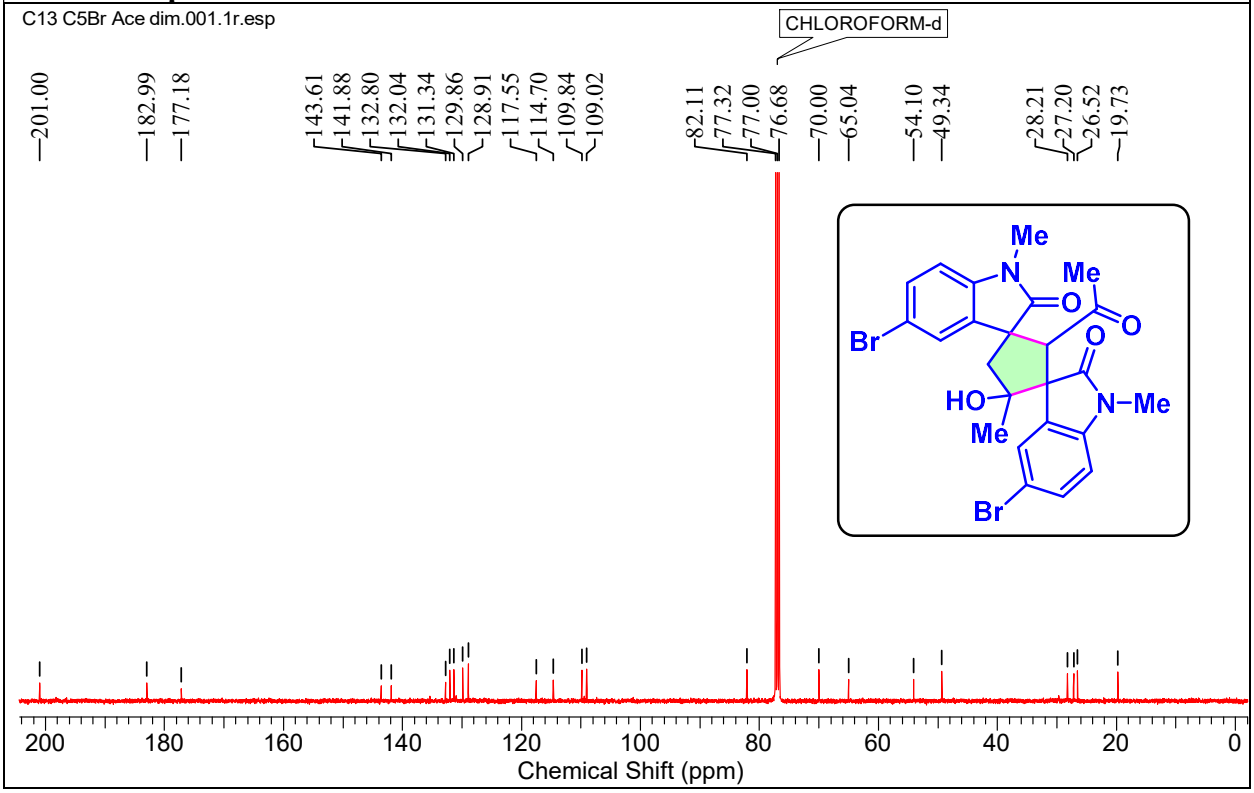
^{13}C NMR spectrum of 4Bg



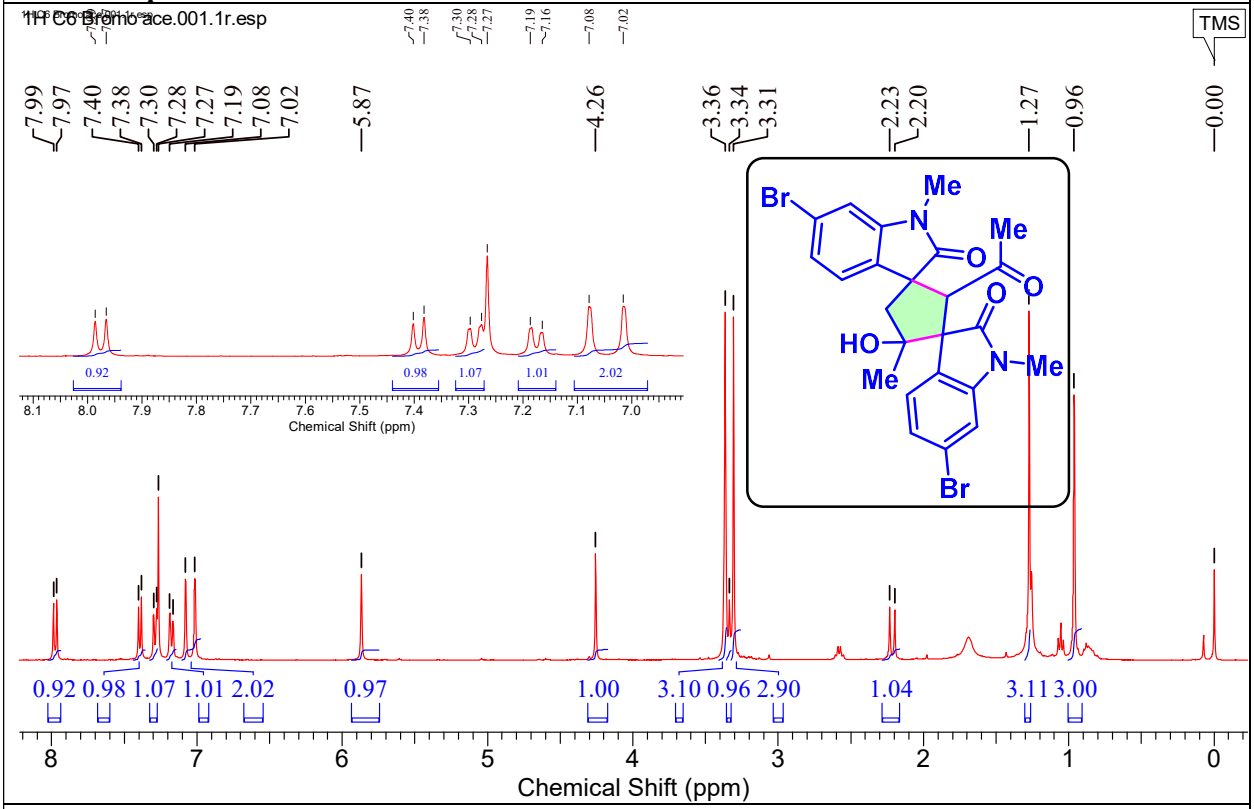
^{1}H NMR spectrum of 4Bh



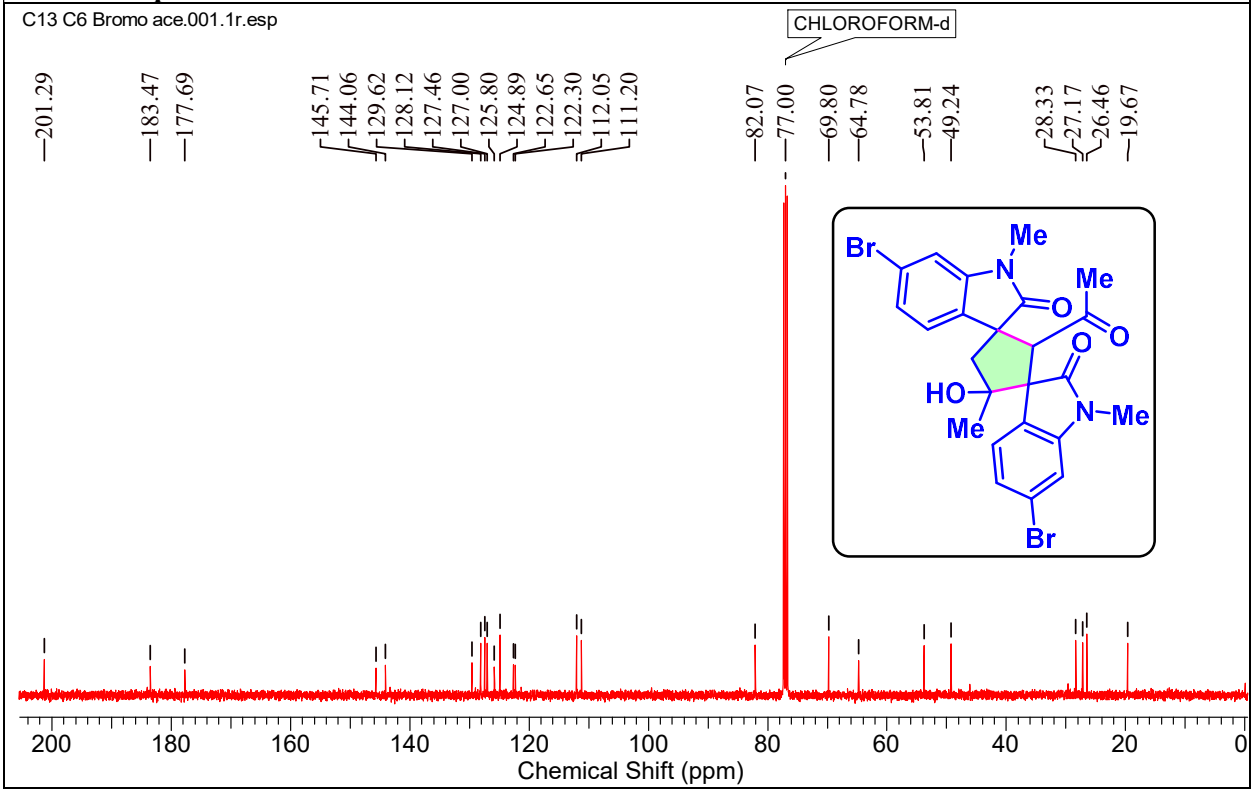
^{13}C NMR spectrum of 4Bh



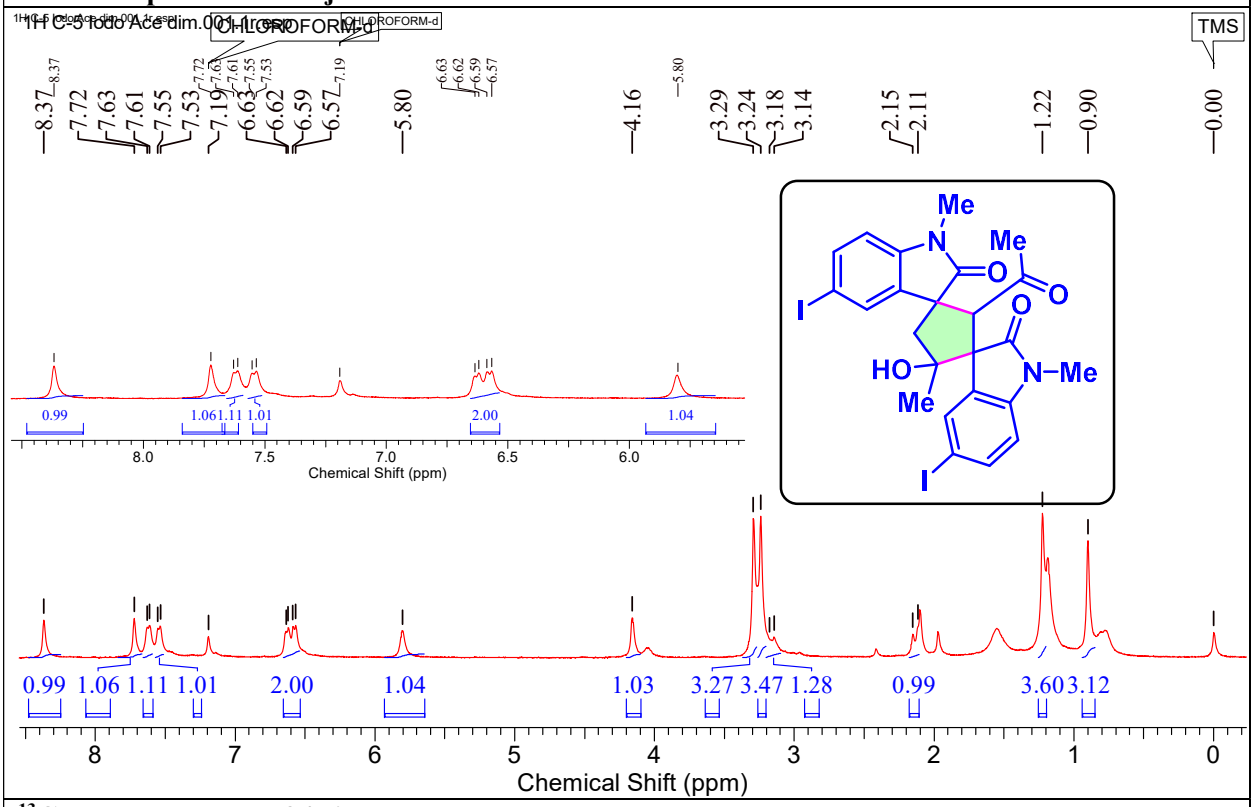
^{1}H NMR spectrum of 4Bi



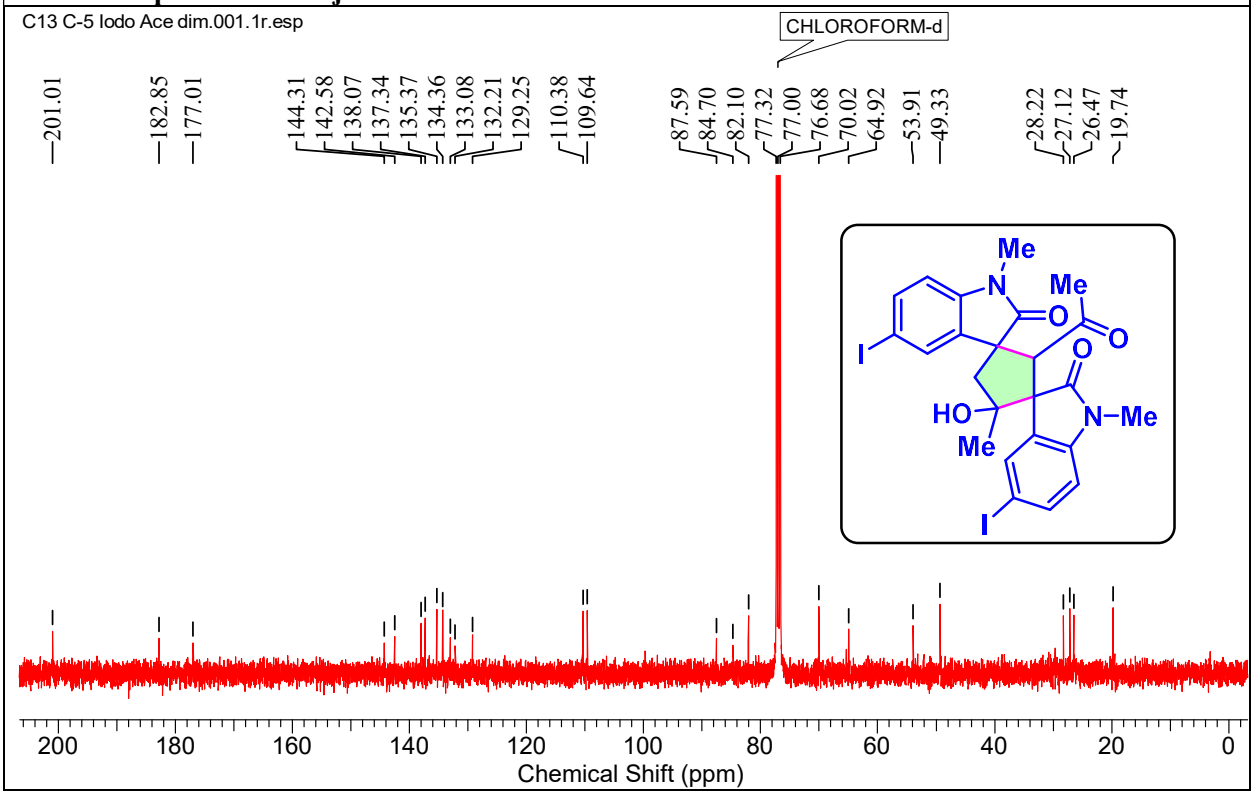
^{13}C NMR spectrum of 4Bi



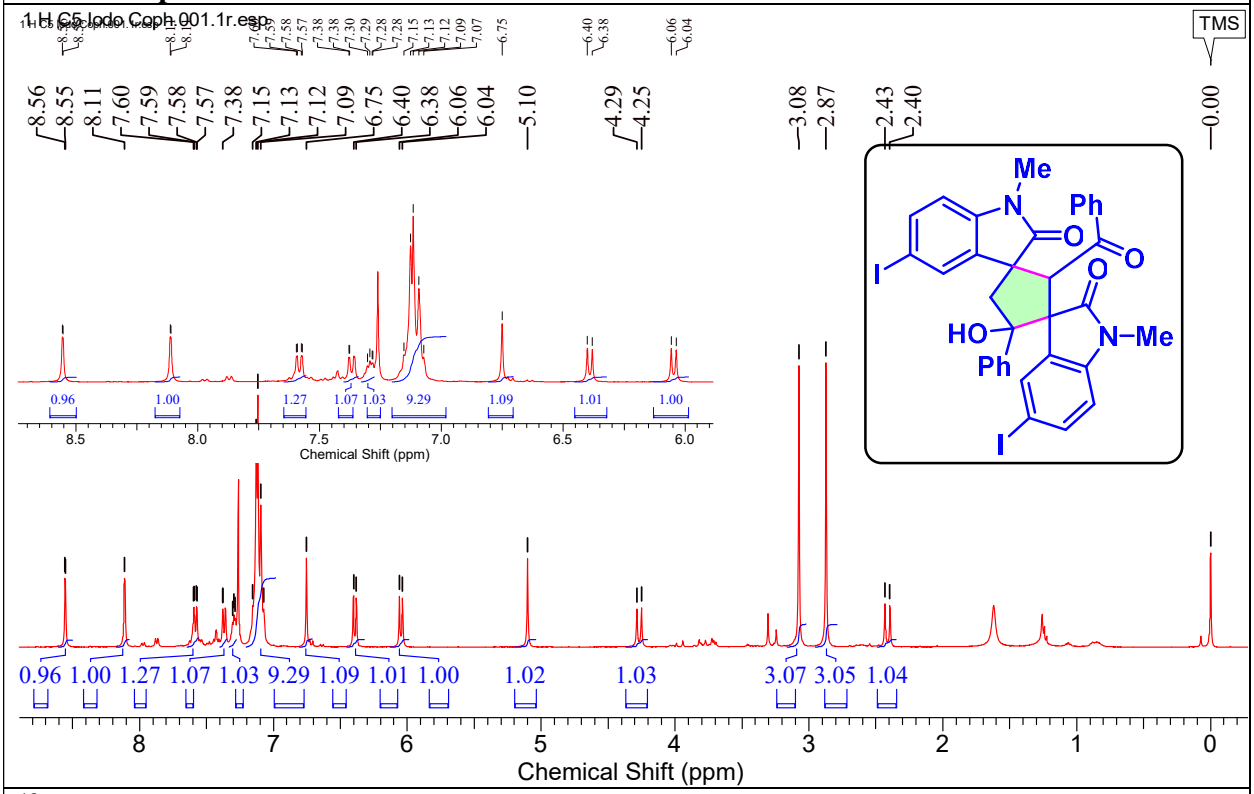
^{1}H NMR spectrum of 4Bj



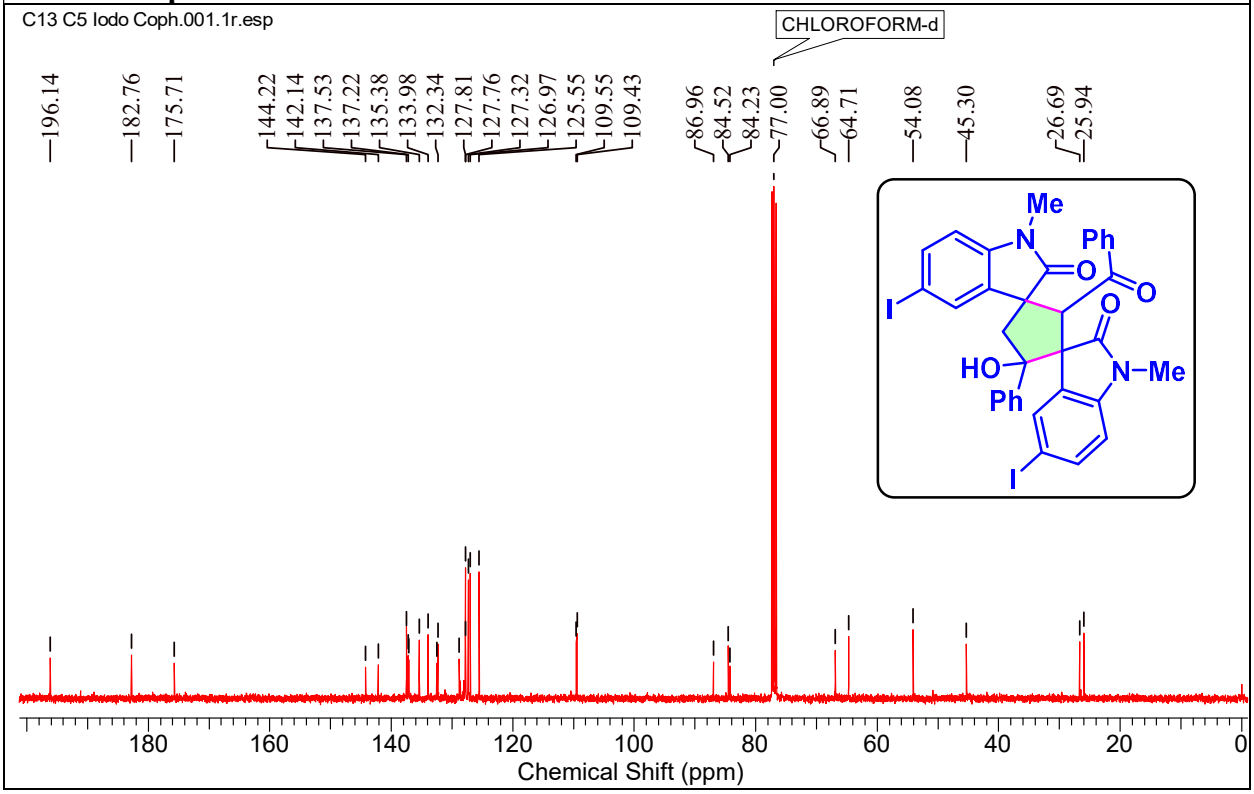
^{13}C NMR spectrum of 4Bj



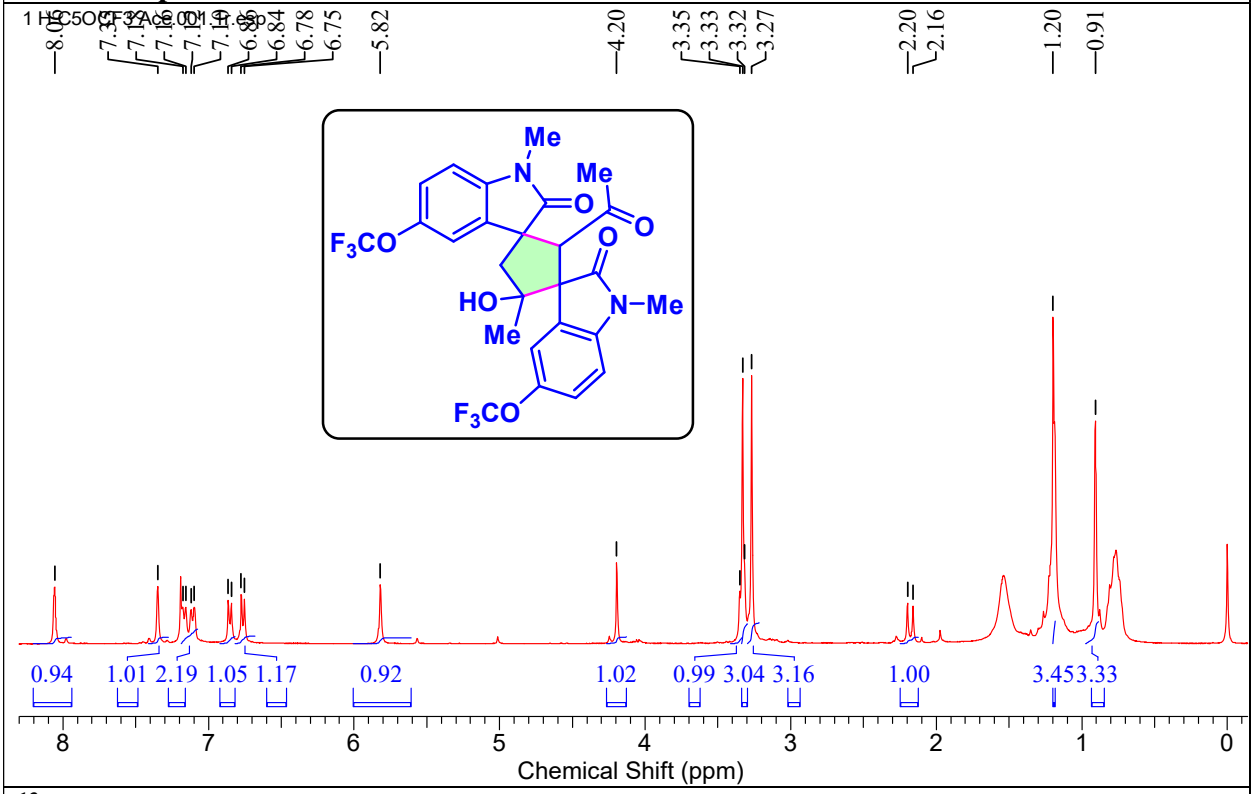
^{1}H NMR spectrum of 4Bk



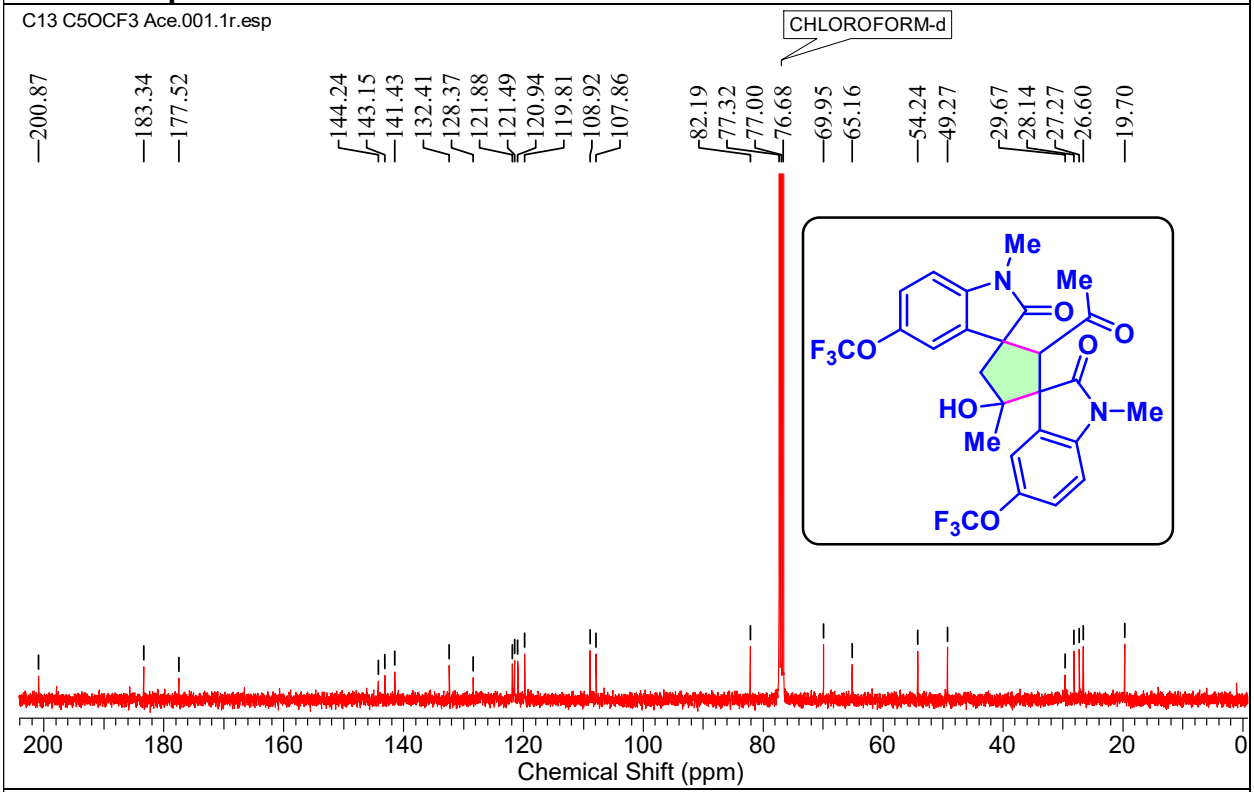
^{13}C NMR spectrum of 4Bk



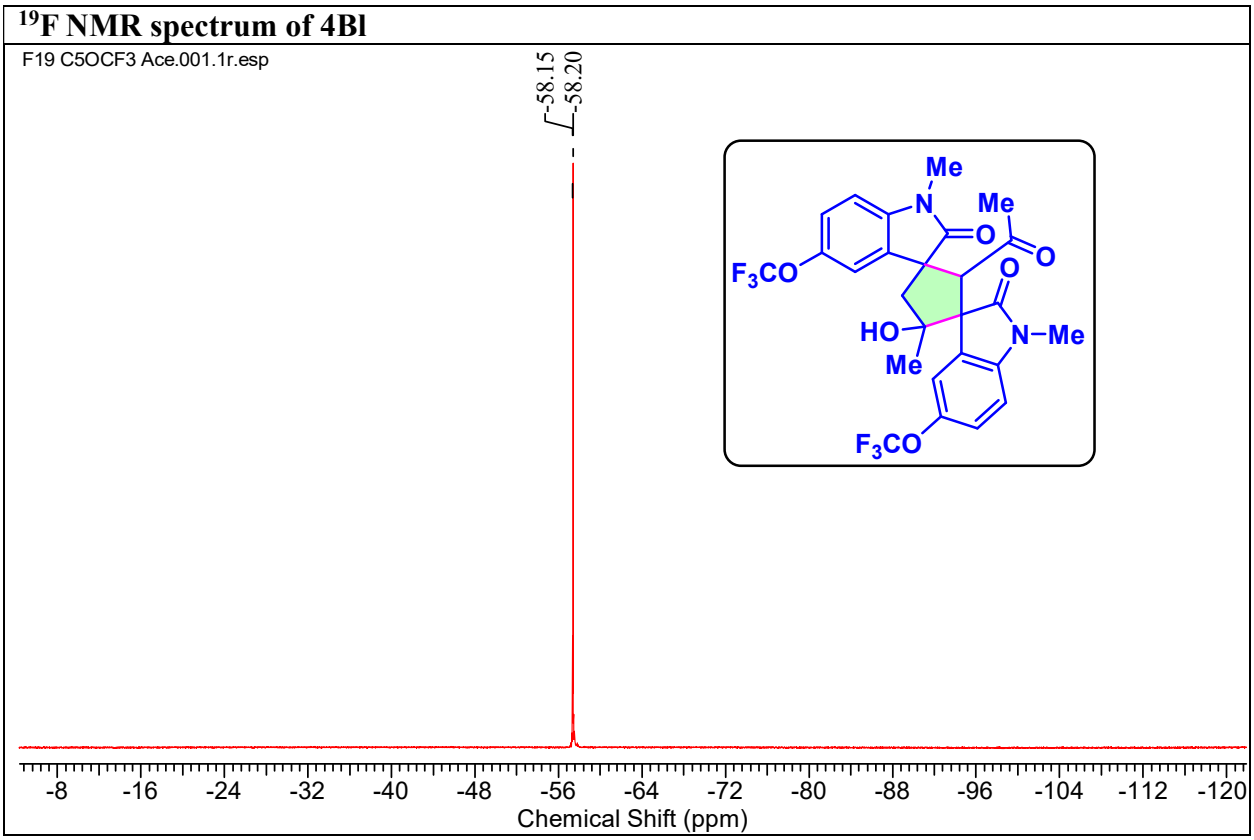
^{1}H NMR spectrum of 4Bl



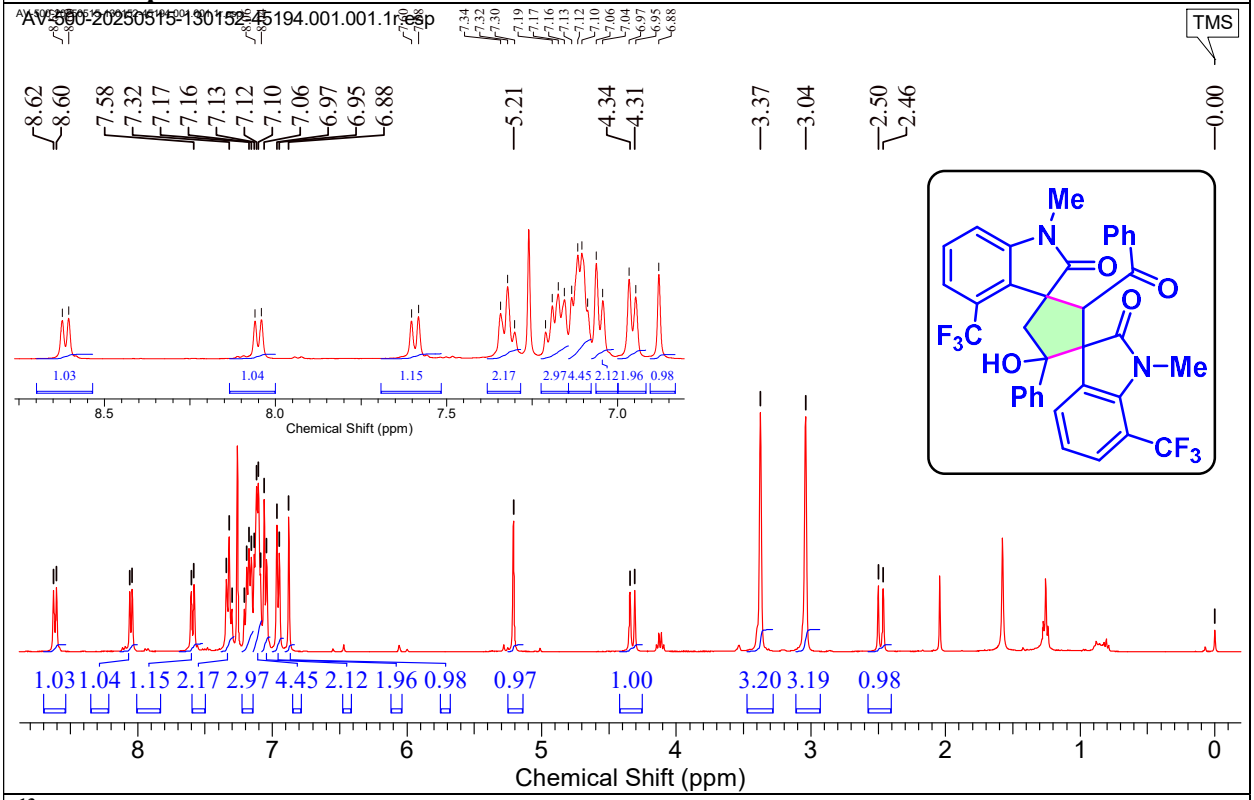
^{13}C NMR spectrum of 4Bl



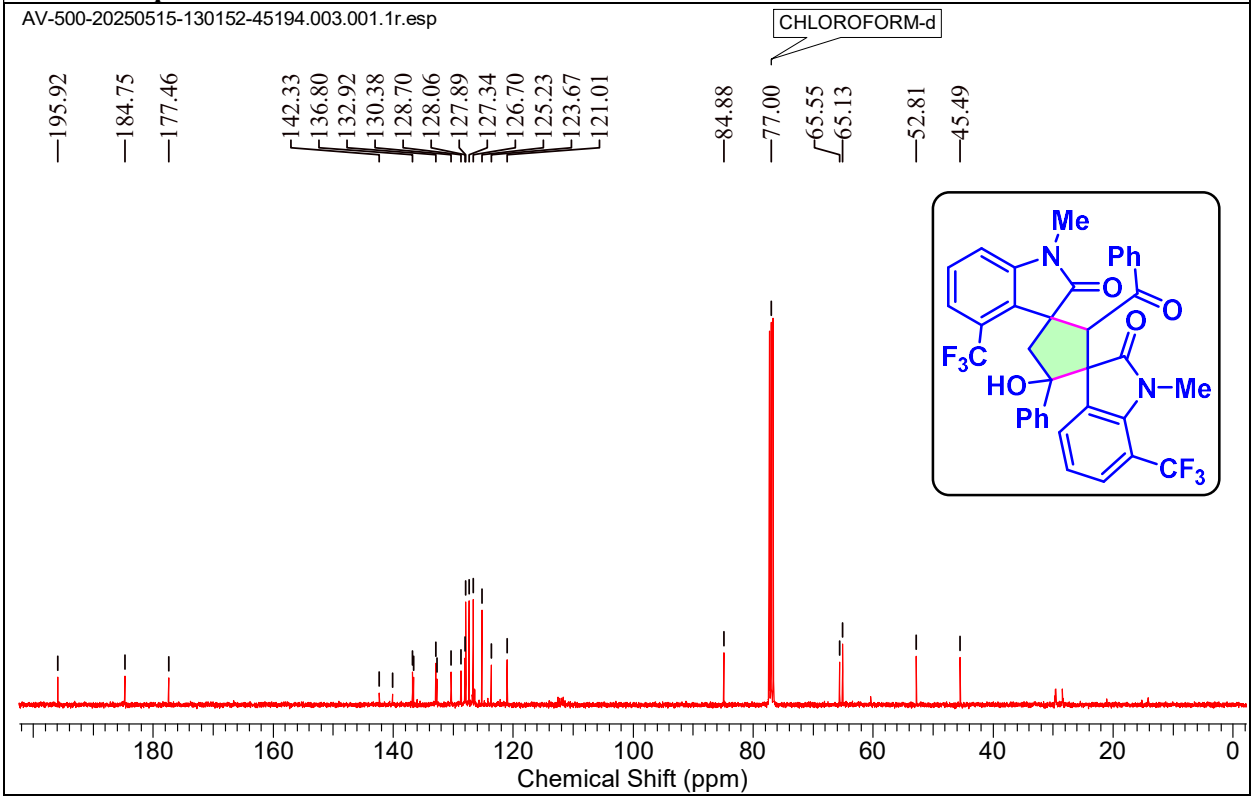
^{19}F NMR spectrum of 4Bl



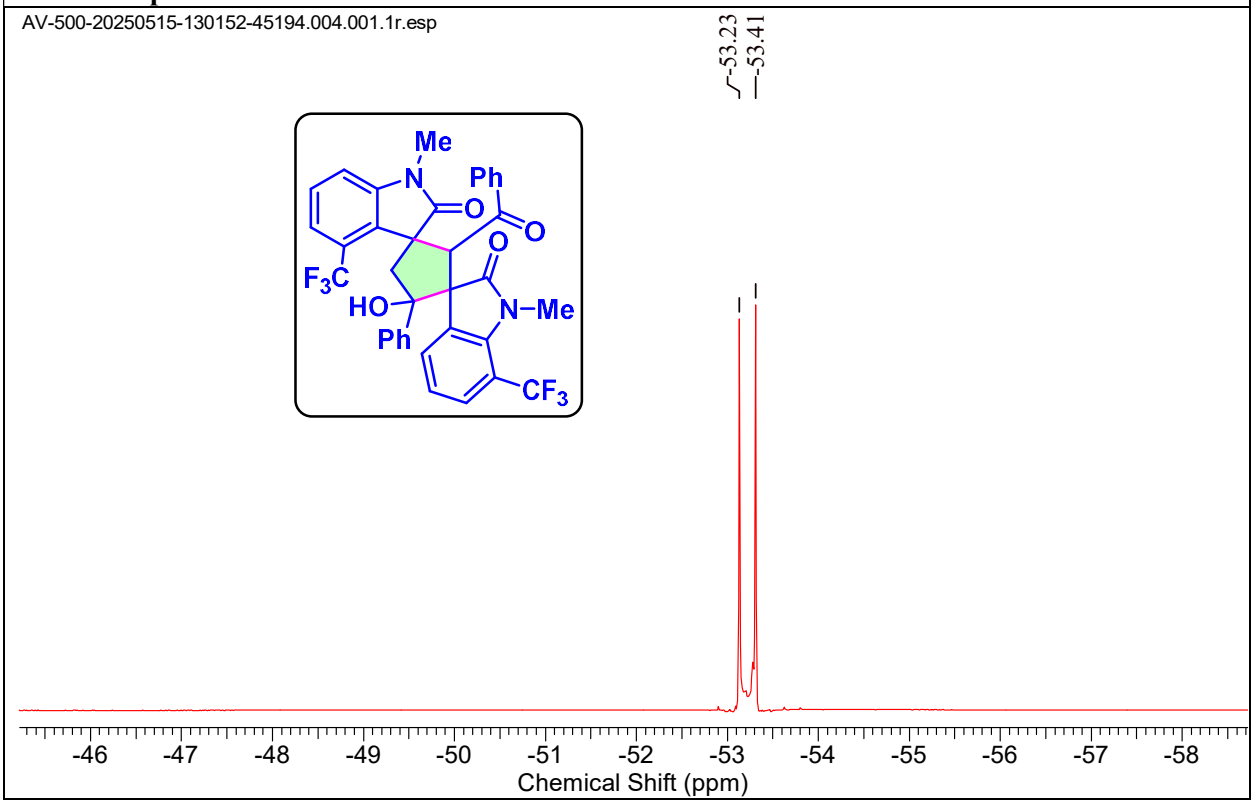
^{1}H NMR spectrum of 4Bm



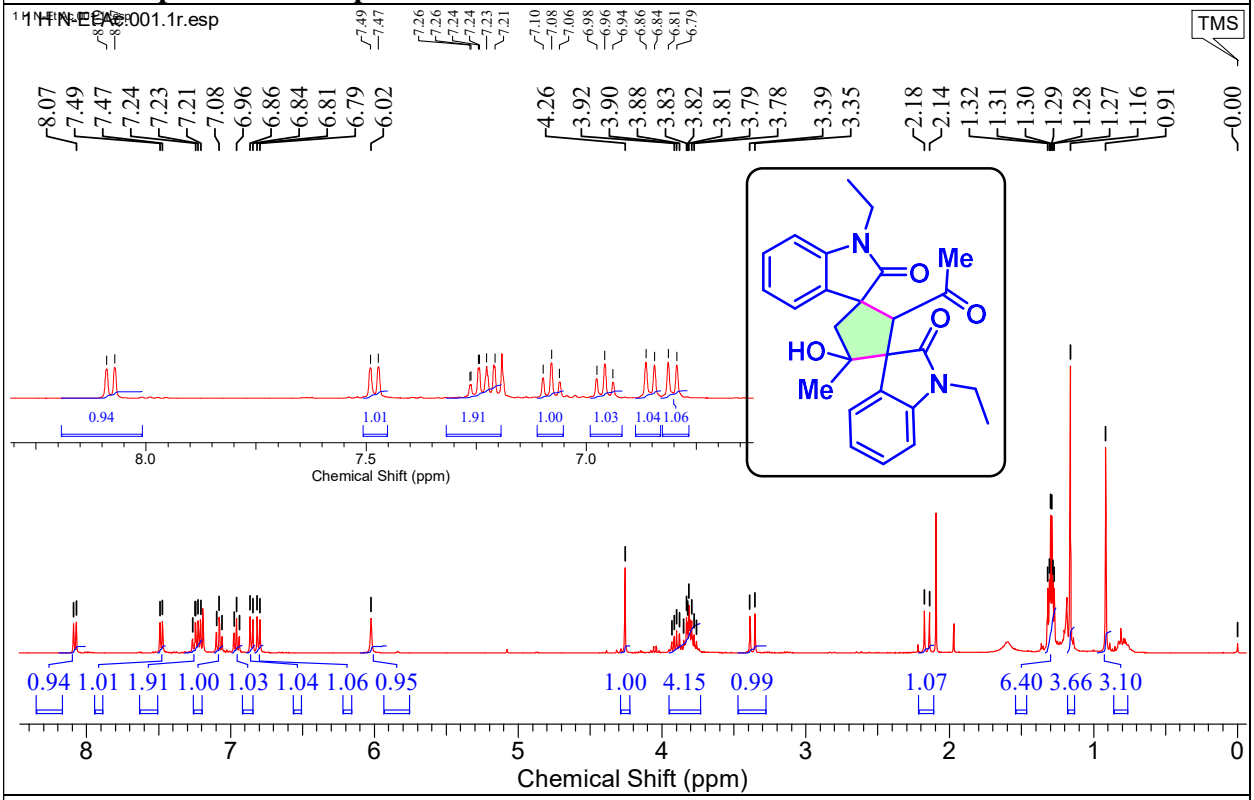
^{13}C NMR spectrum of 4Bm



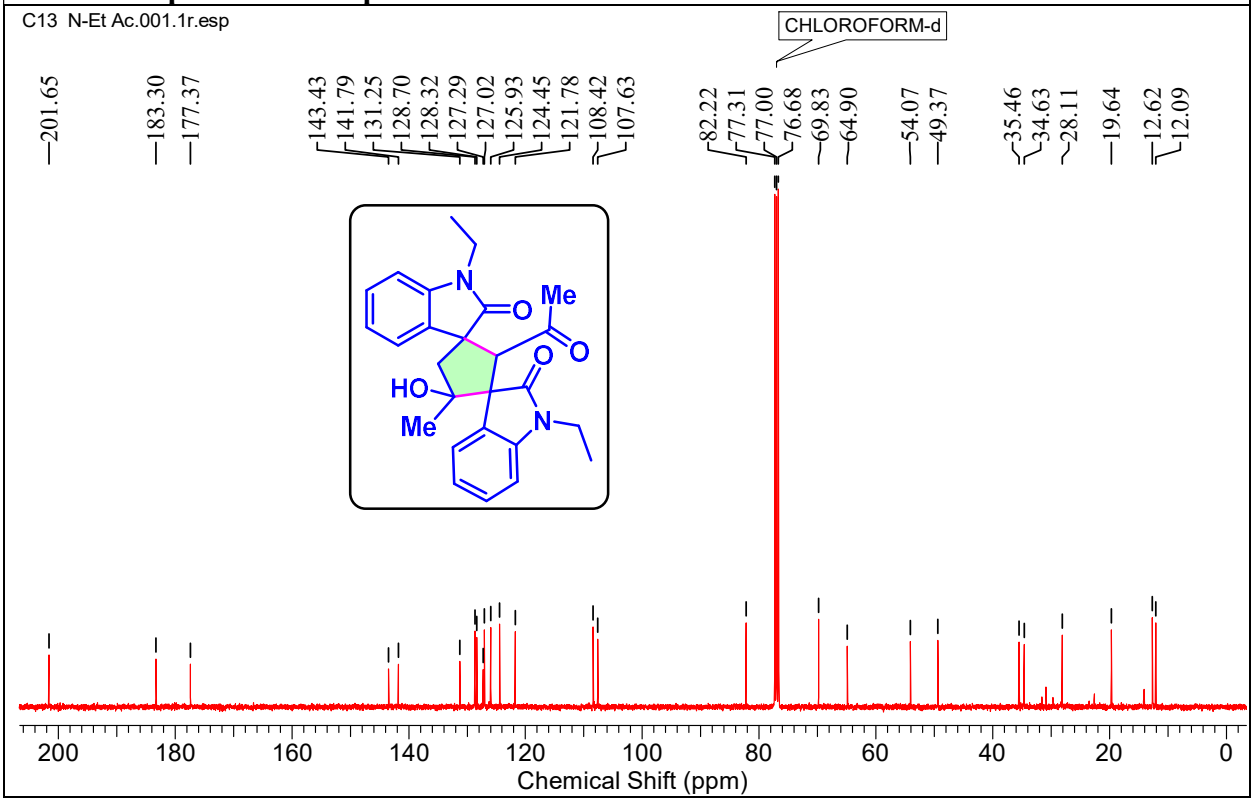
^{19}F NMR spectrum of 4Bm



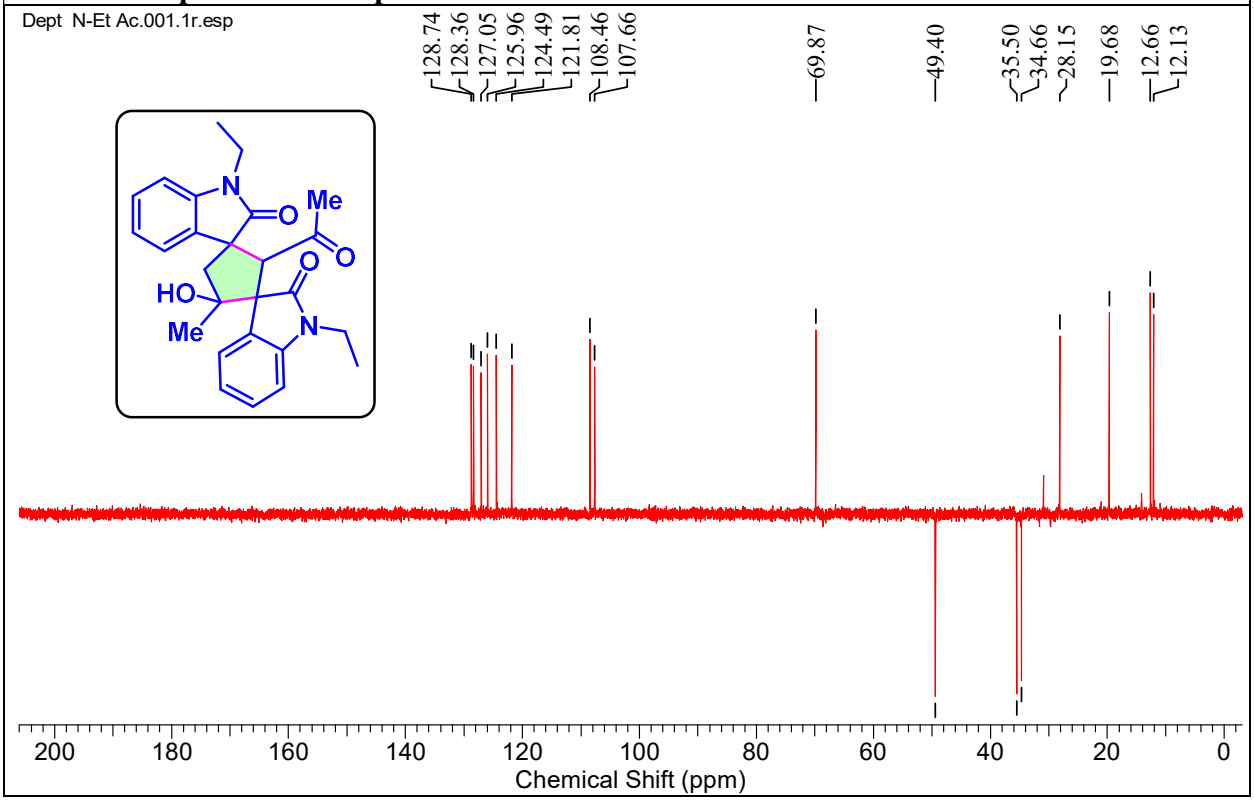
^{1}H NMR spectrum of 4Bp



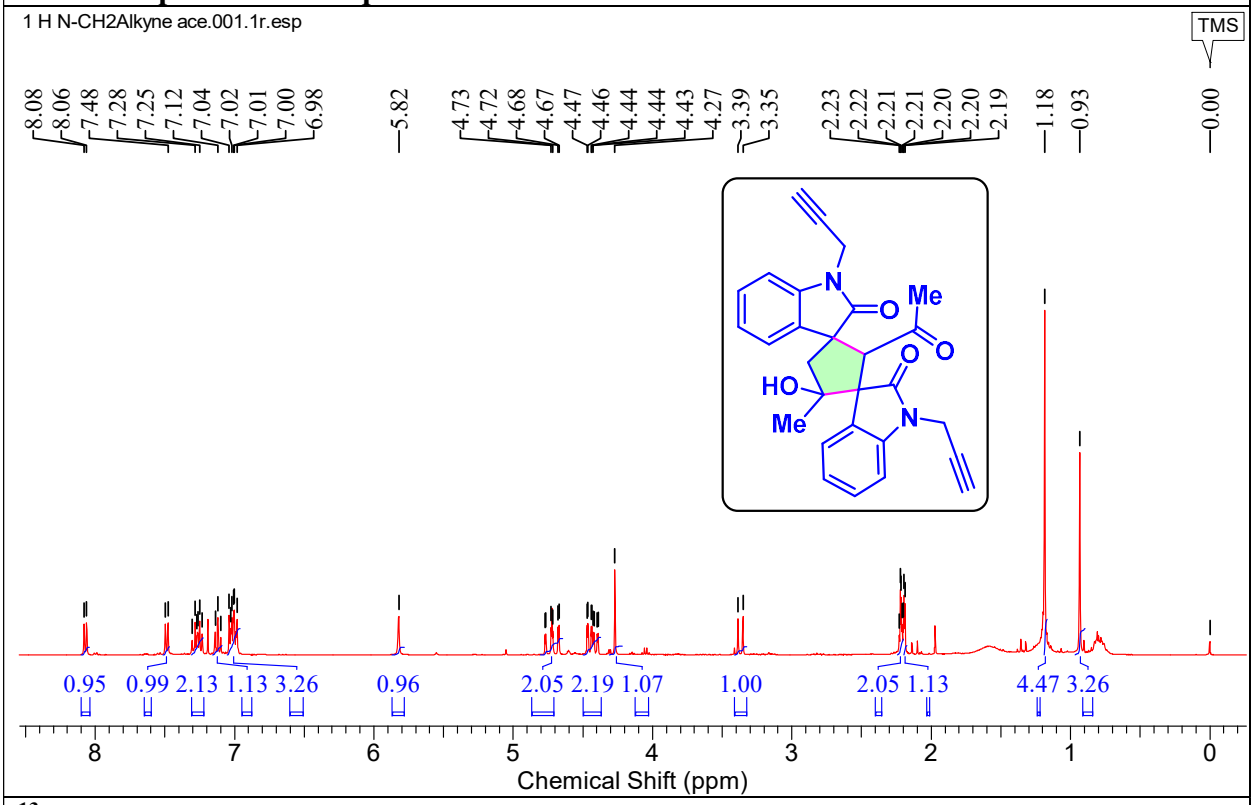
^{13}C NMR spectrum of 4Bp



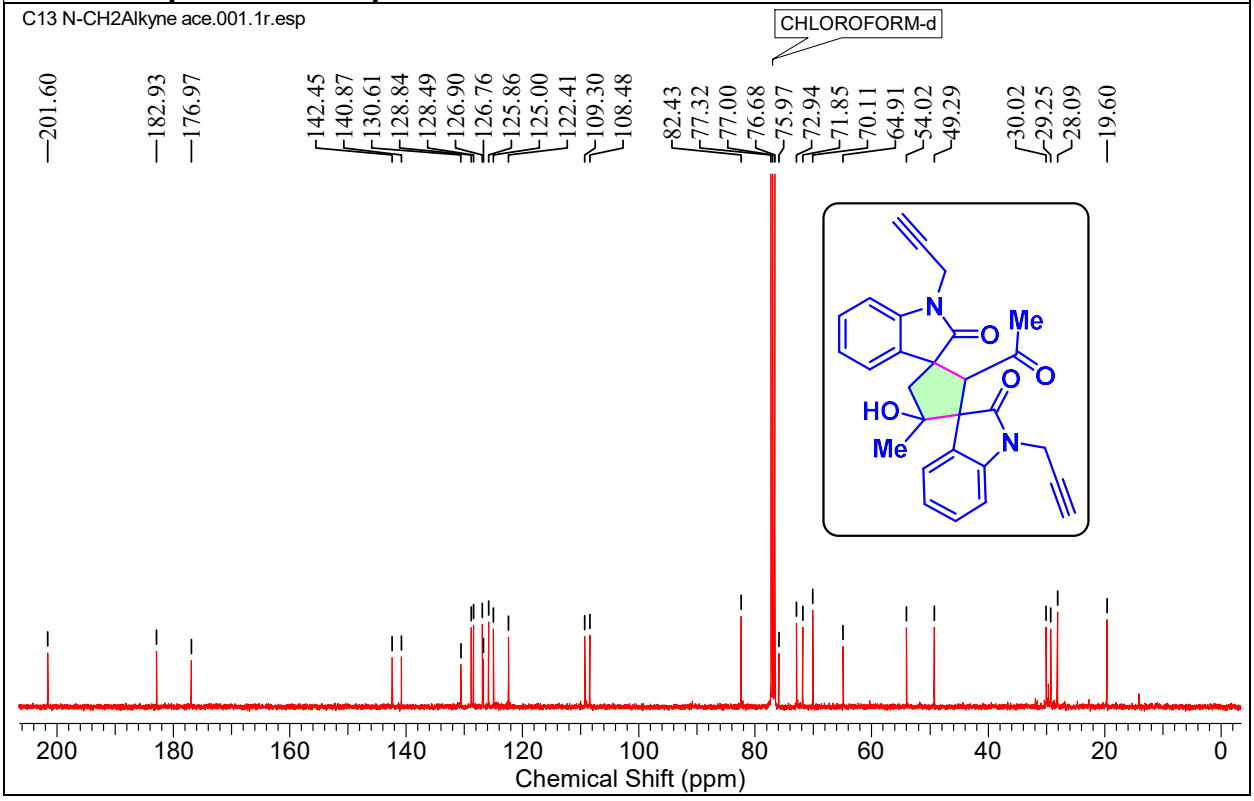
135 DEPT spectrum of 4Bp



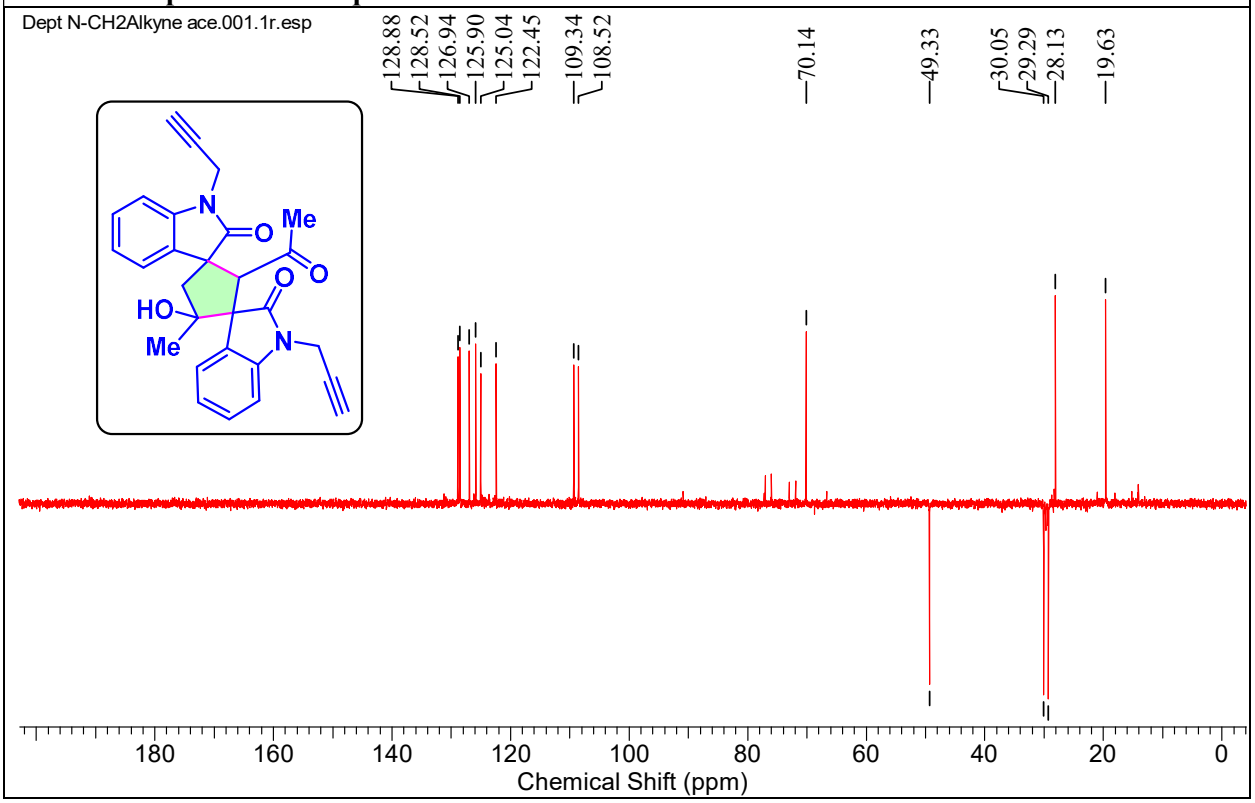
^{1}H NMR spectrum of 4Bq



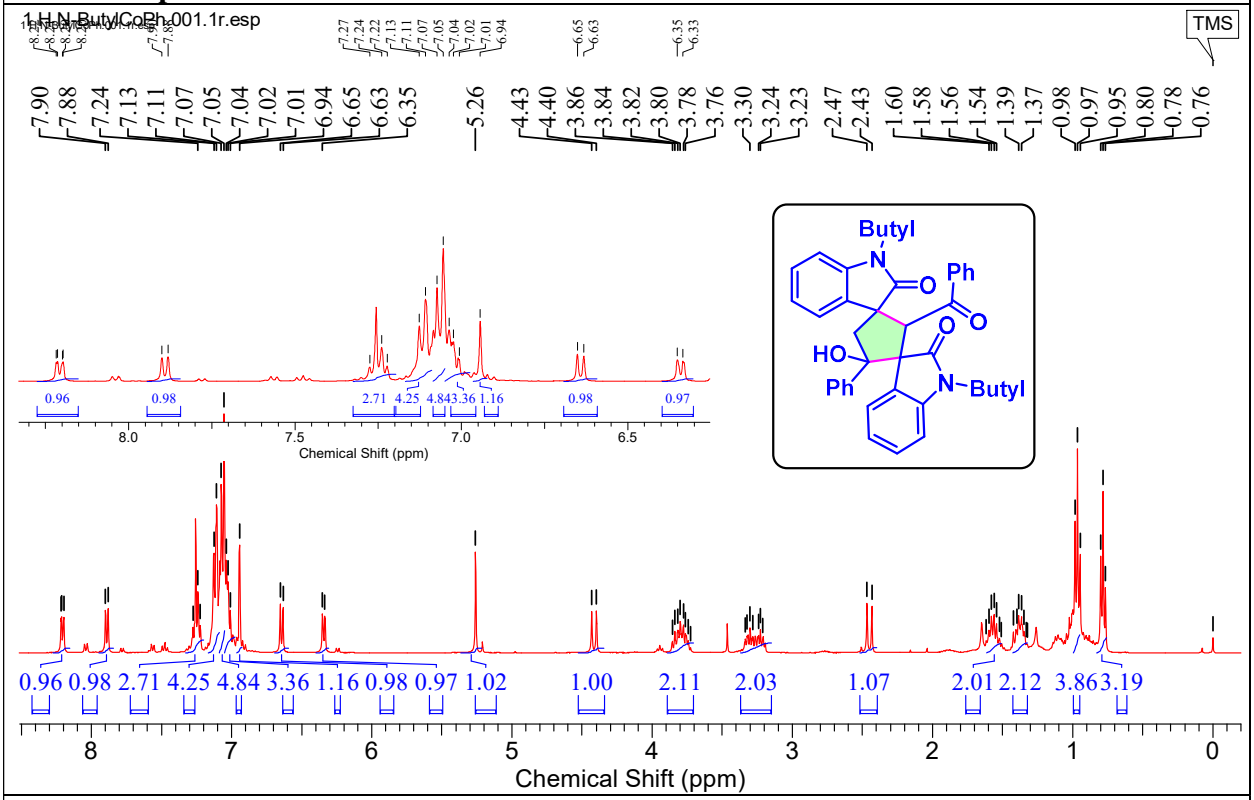
^{13}C NMR spectrum of 4Bq



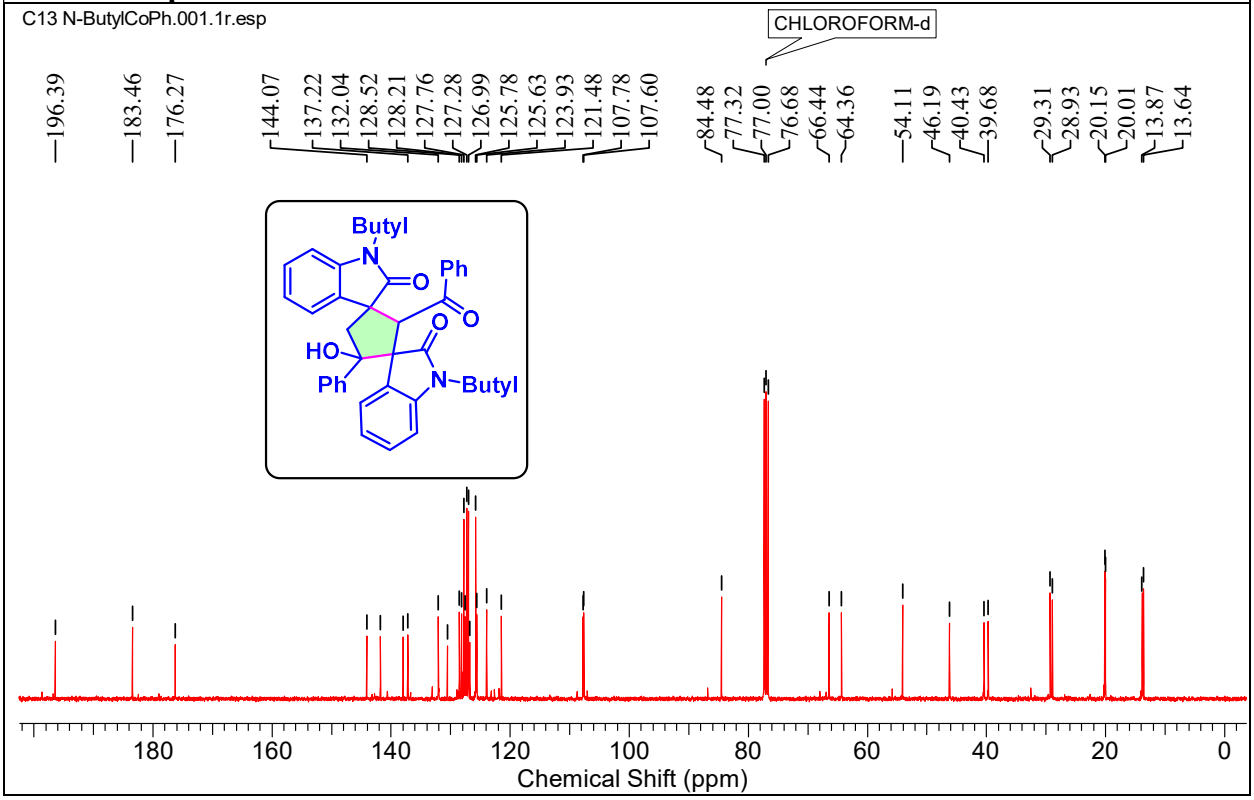
135 DEPT spectrum of 4Bq



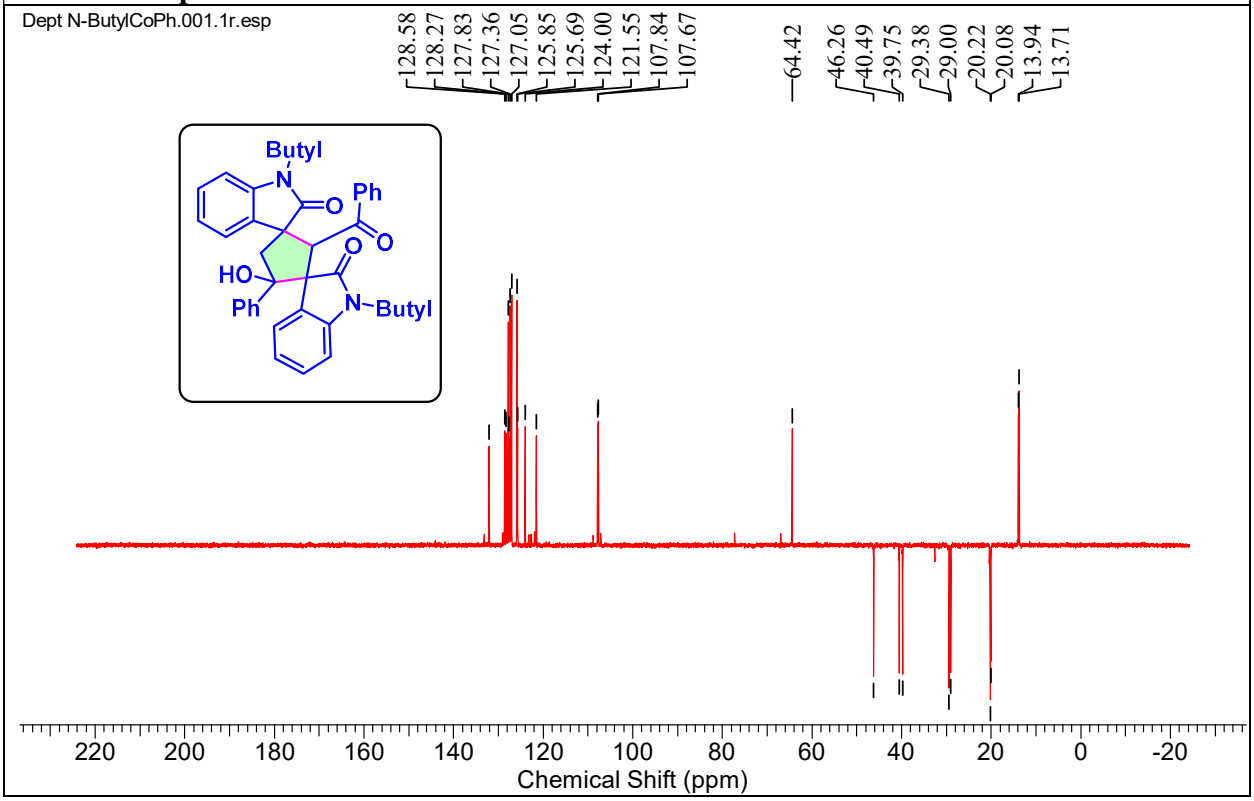
^{1}H NMR spectrum of 4Br



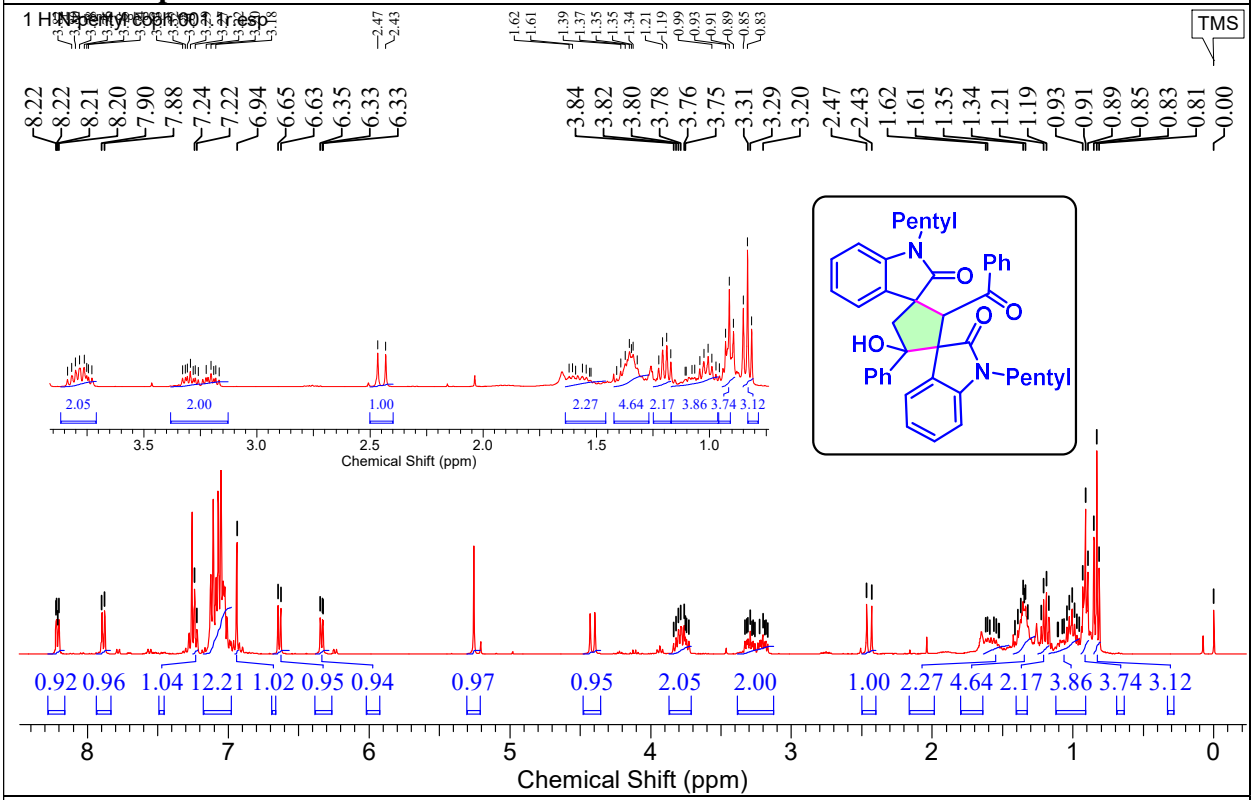
^{13}C NMR spectrum of 4Br



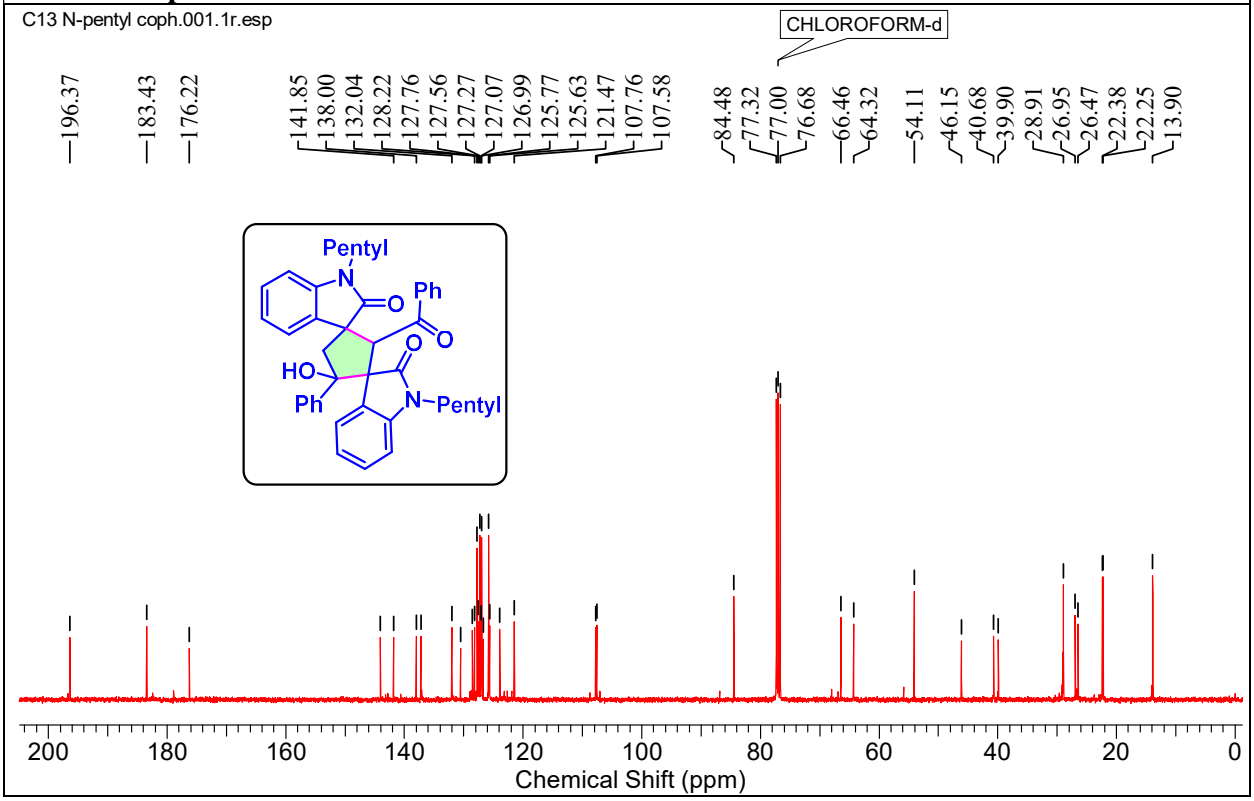
135 DEPT spectrum of 4Br



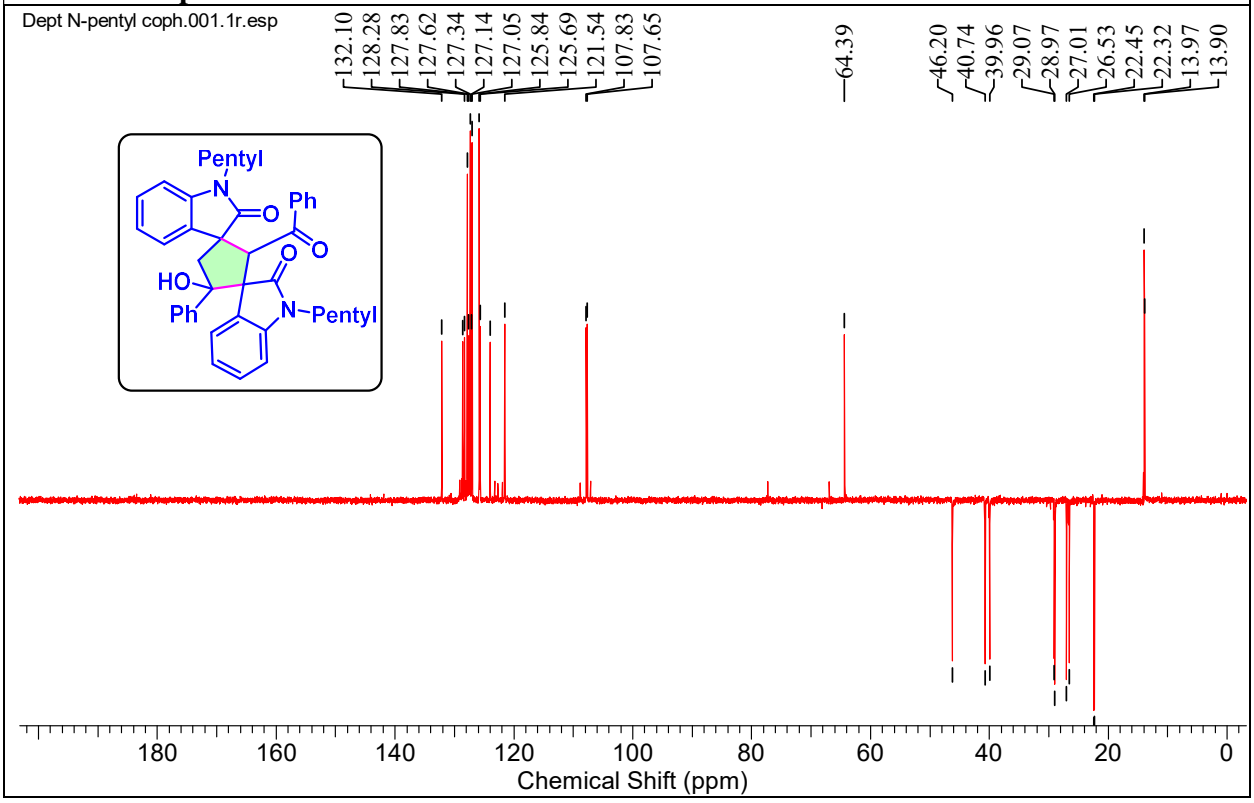
^{1}H NMR spectrum of 4Bs



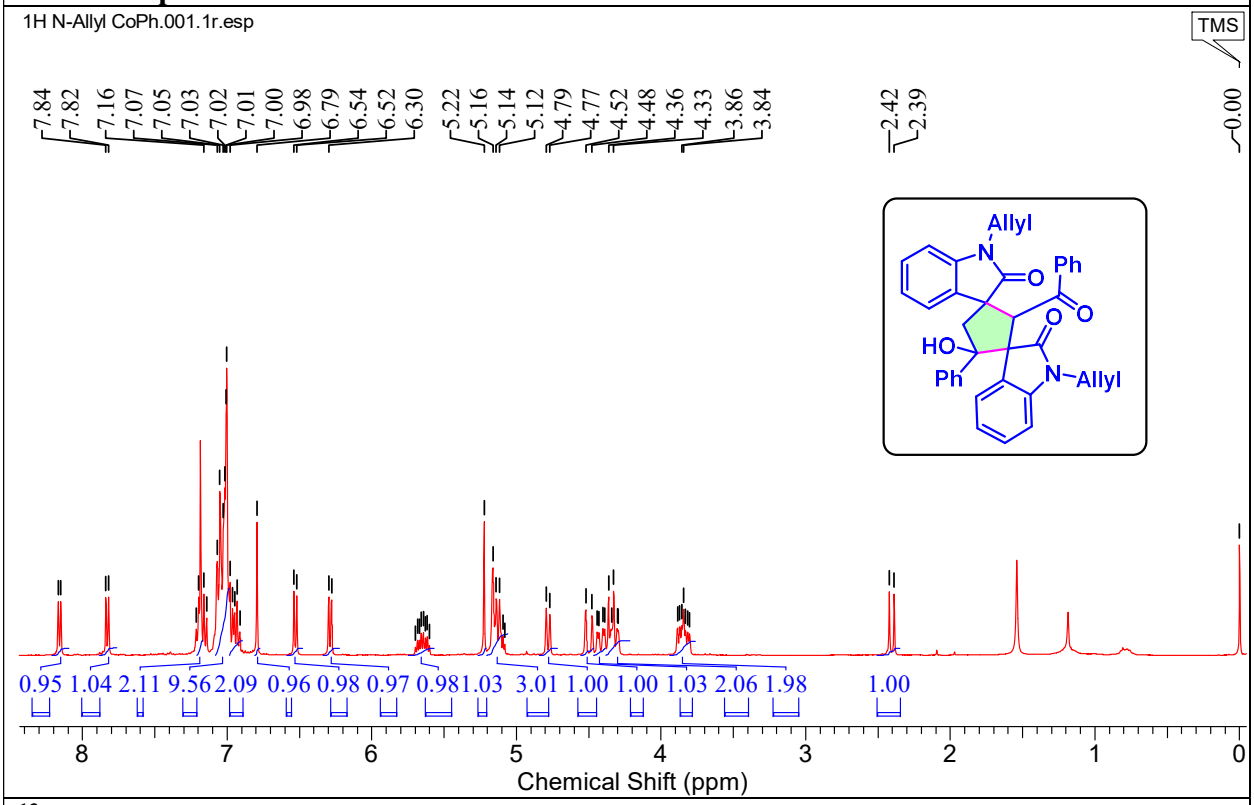
^{13}C NMR spectrum of 4Bs



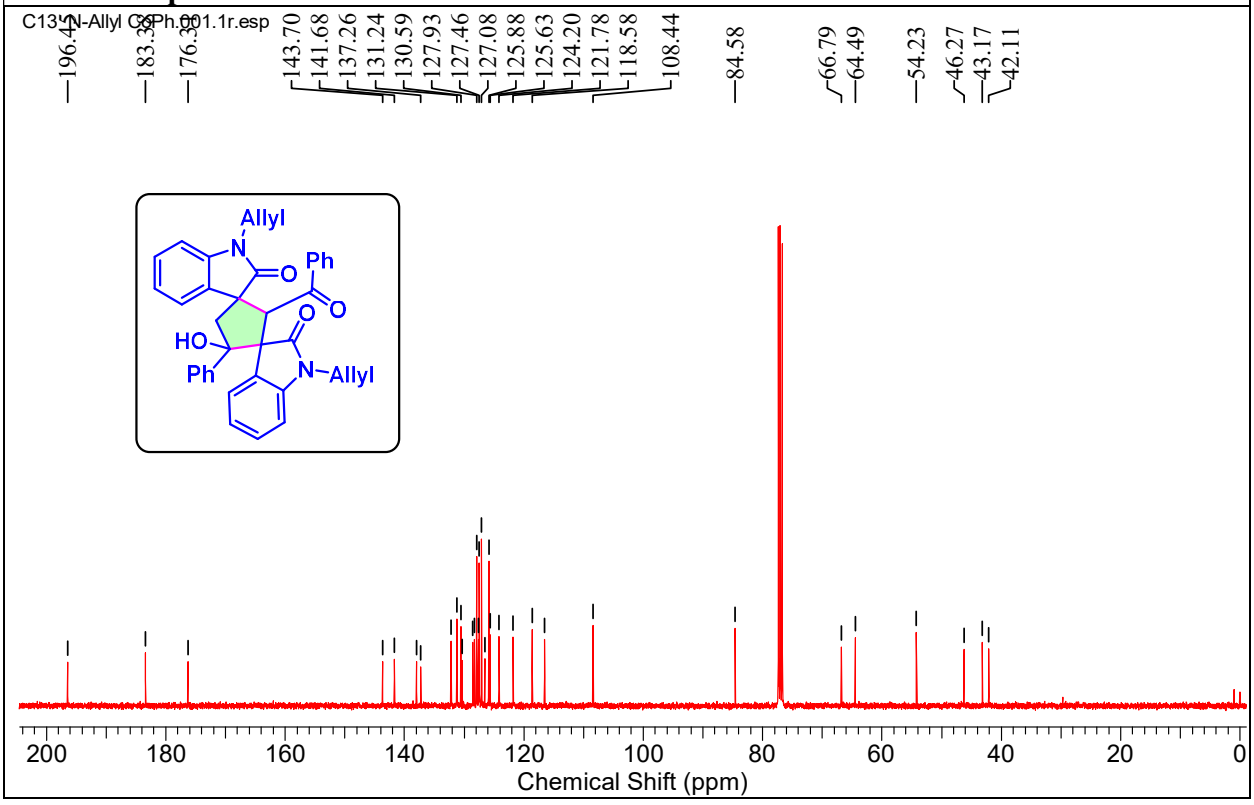
135 DEPT spectrum of 4Bs



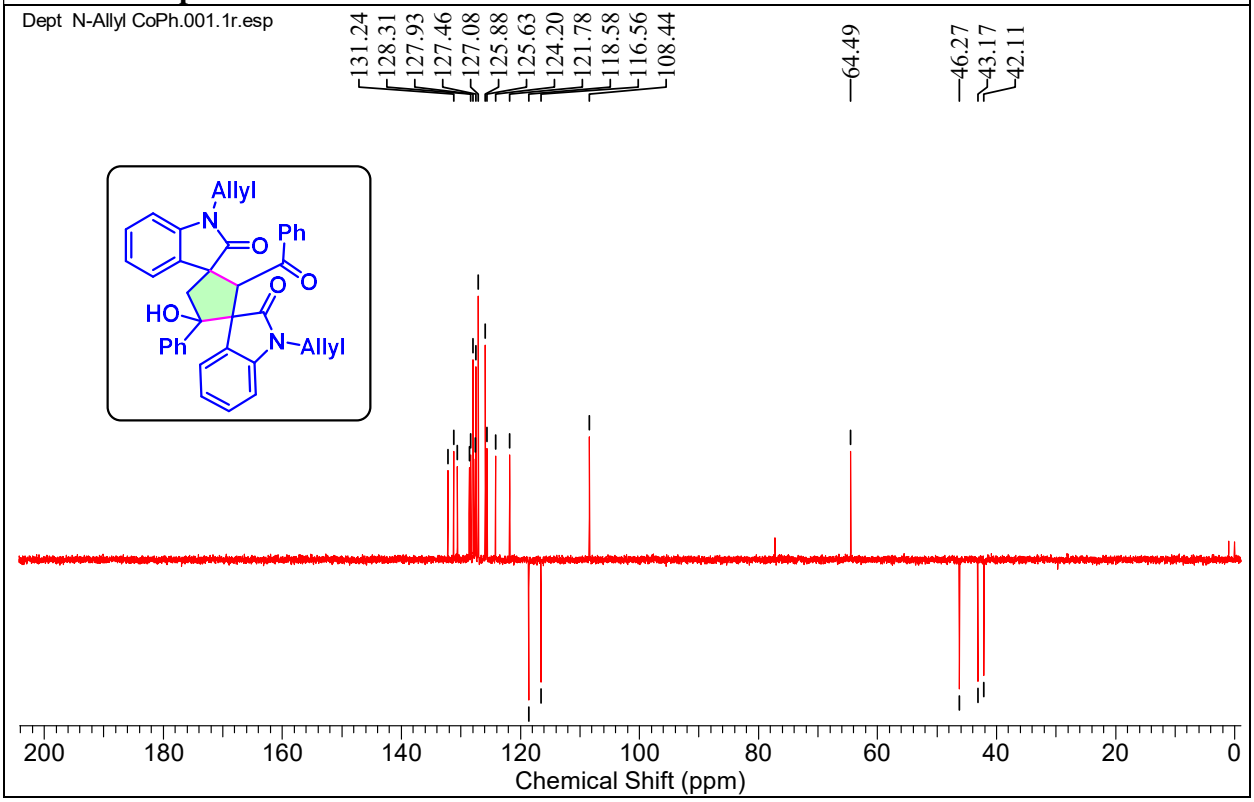
^{1}H NMR spectrum of 4Bt



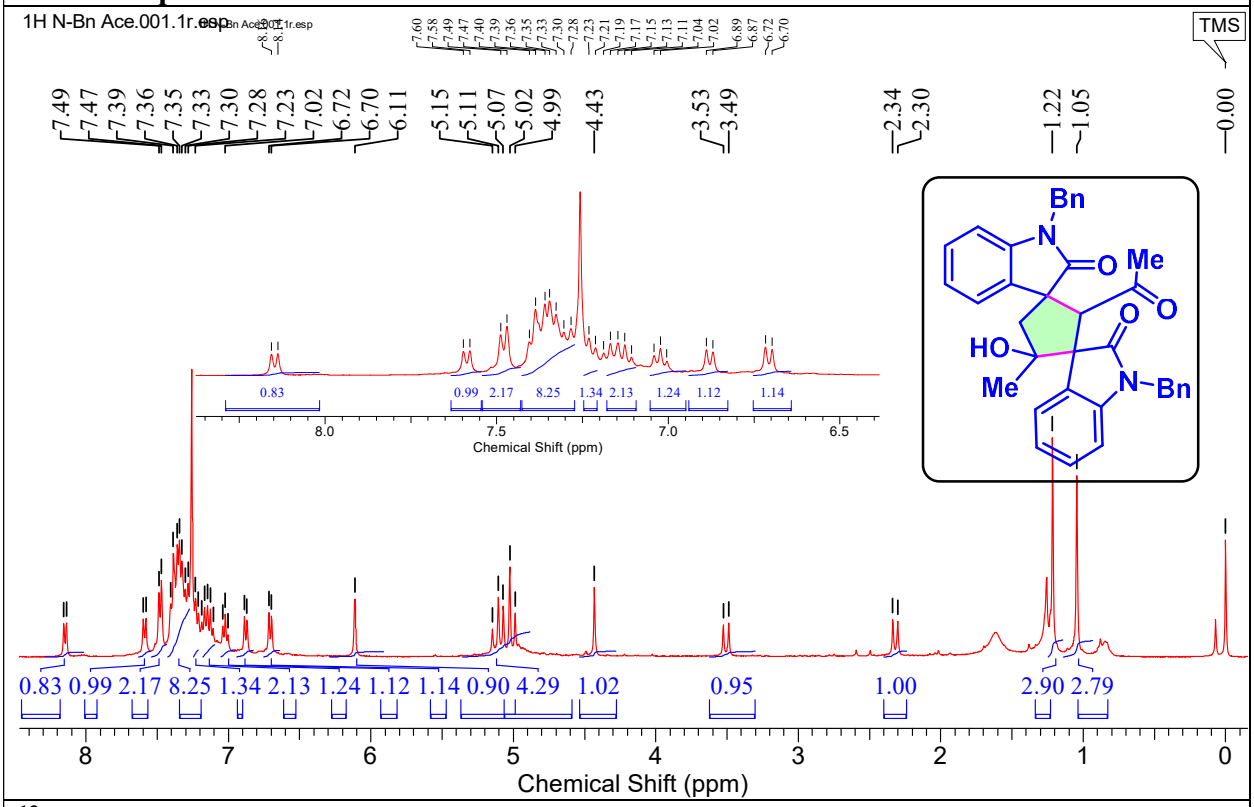
^{13}C NMR spectrum of 4Bt



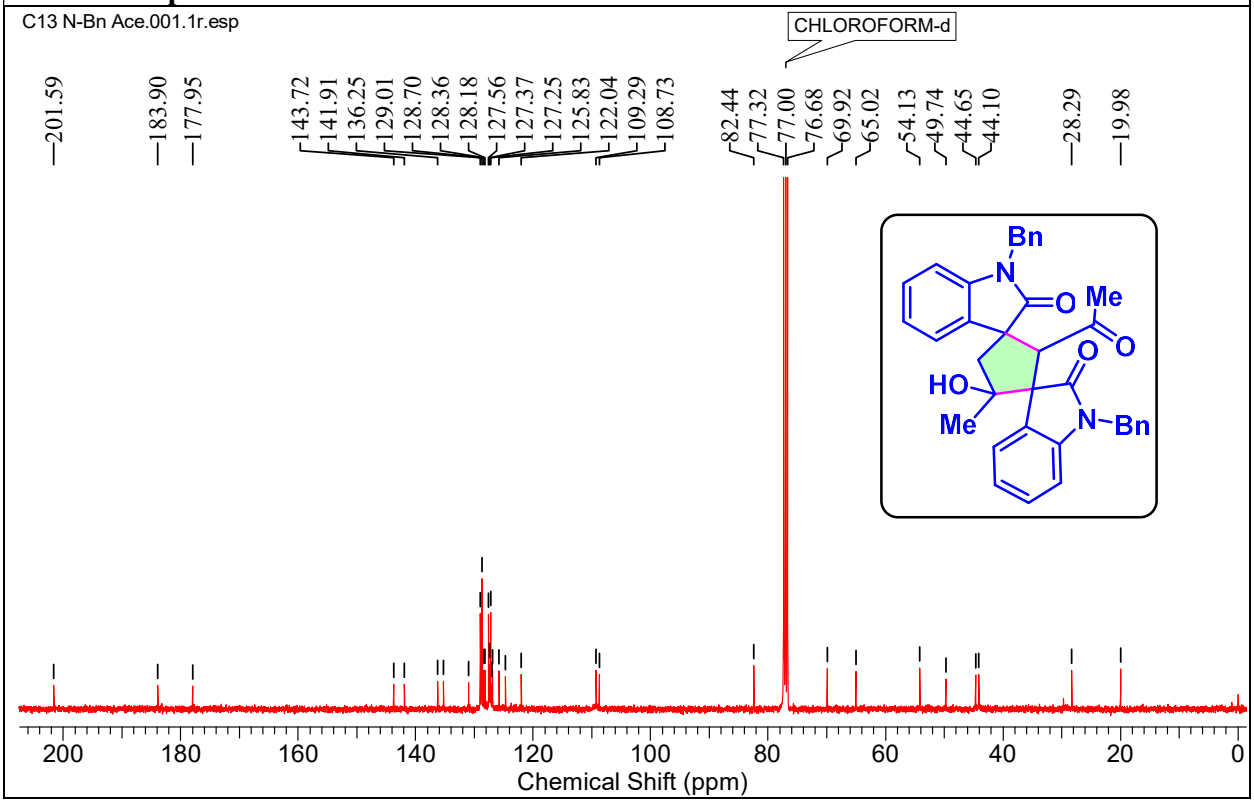
135 DEPT spectrum of 4Bt



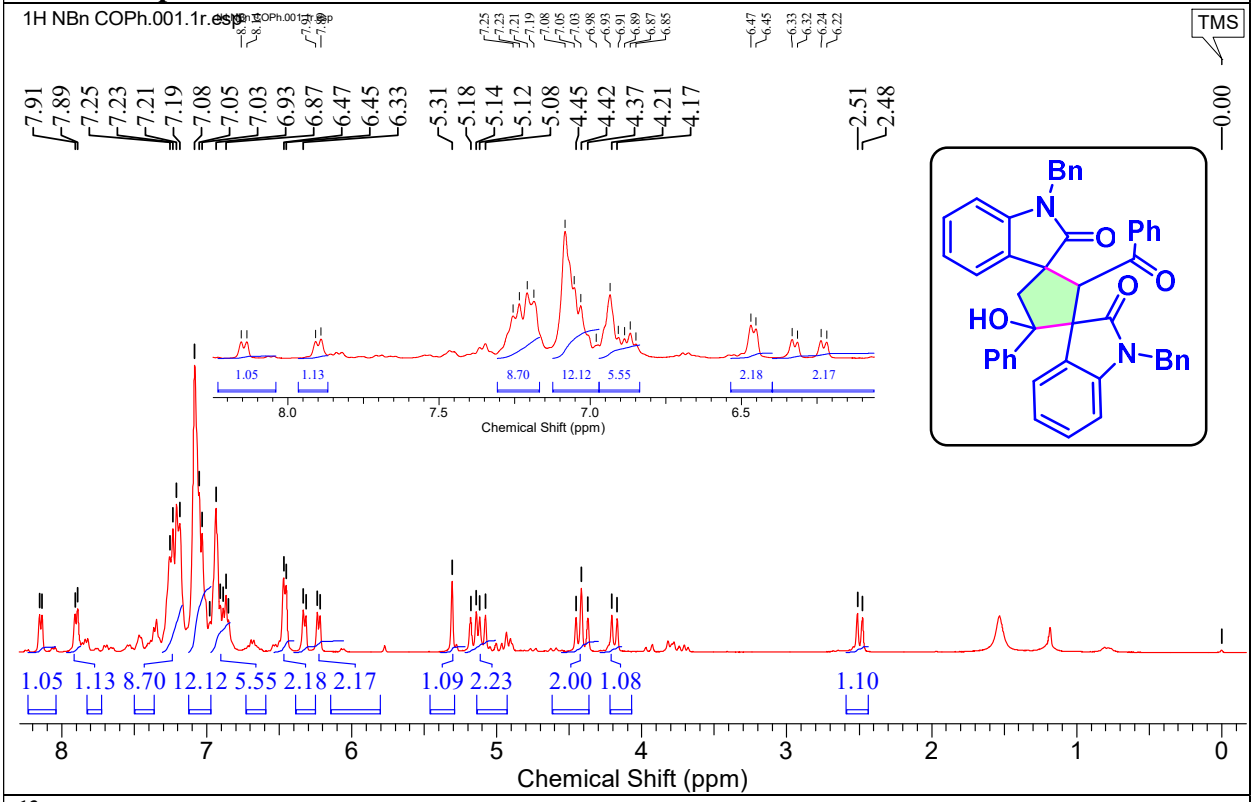
^{1}H NMR spectrum of 4Bu



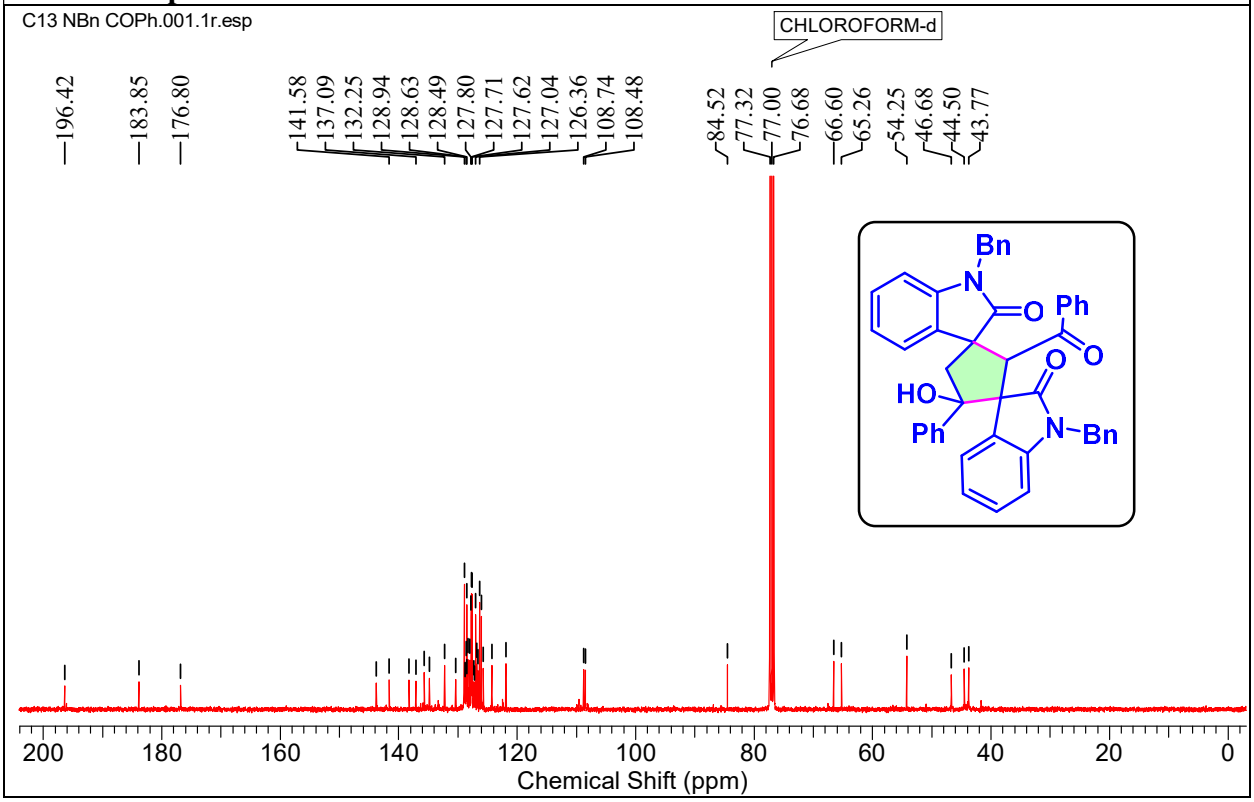
^{13}C NMR spectrum of 4Bu



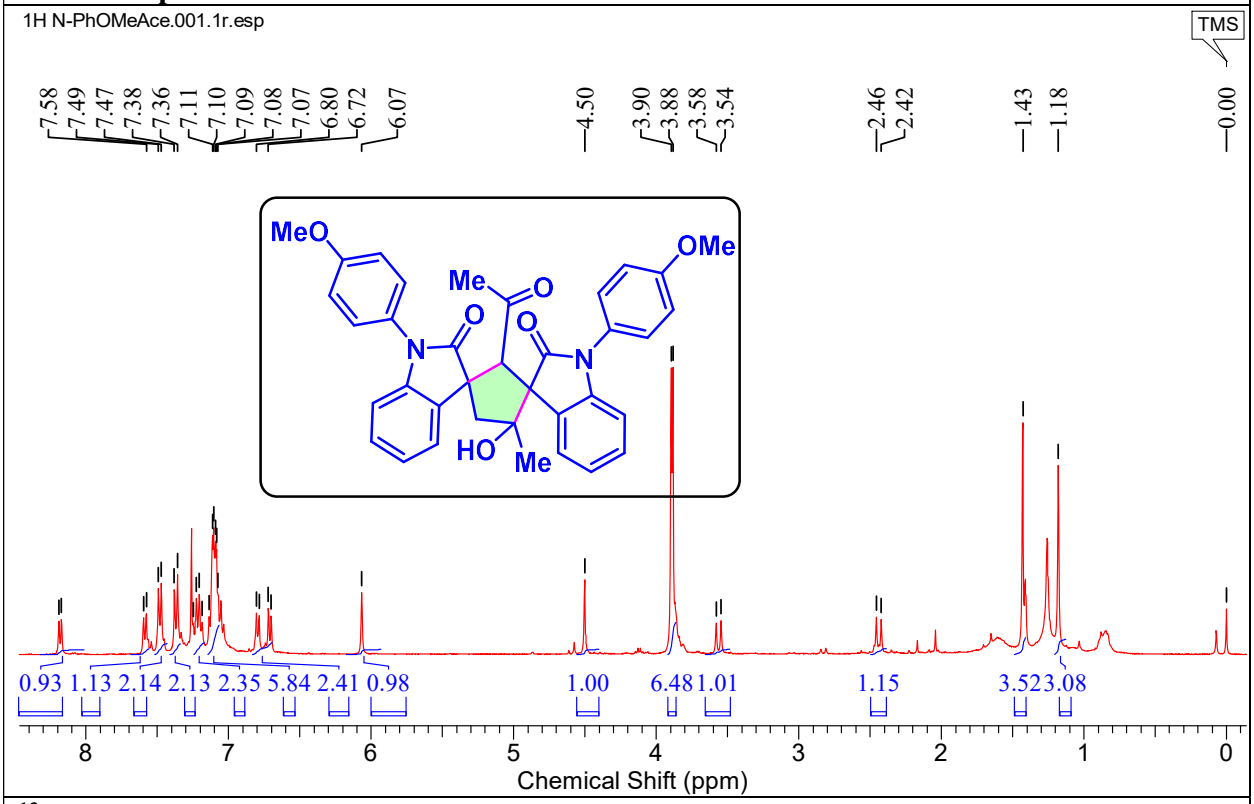
^{1}H NMR spectrum of 4Bv



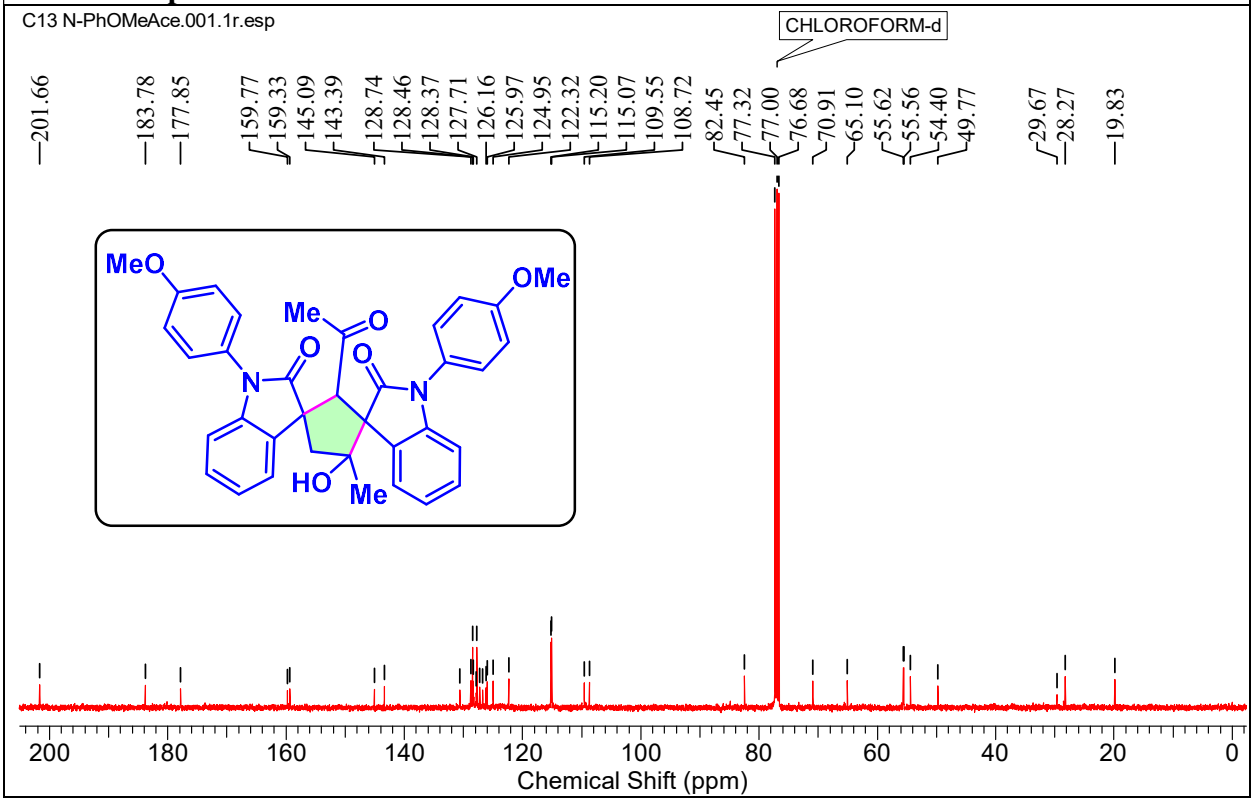
^{13}C NMR spectrum of 4Bv



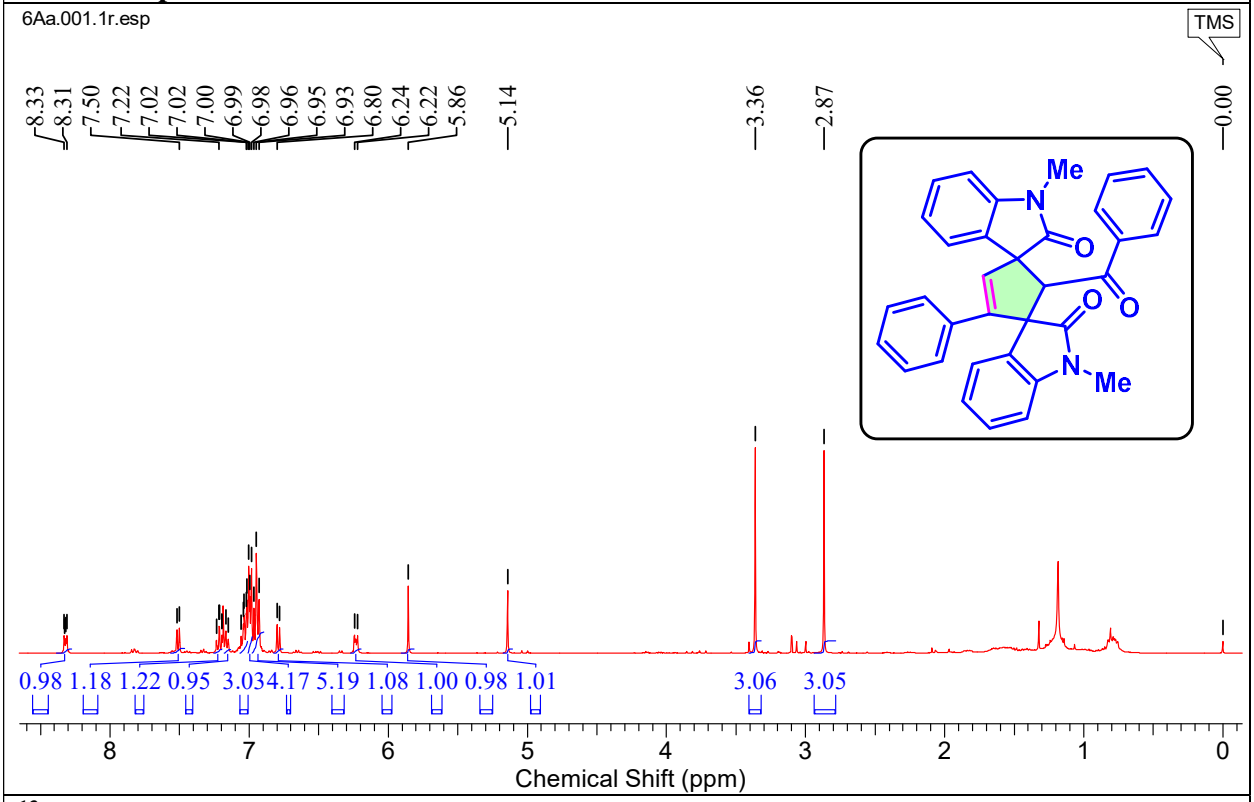
^{1}H NMR spectrum of 4Bw



^{13}C NMR spectrum of 4Bw



^{1}H NMR spectrum of 5A



^{13}C NMR spectrum of 5A