

Supplementary Materials

Synthesis of chiral *spiro*-indenones via Pd-catalyzed asymmetric (4 + 2) dipolar cyclization

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Table of Contents

1. General Information	S2
2. UV-vis Absorption Spectra	S3
3. Photoreactor Setup	S3
4. Details for Condition Optimizations	S4
5. General Procedures and Characterization of Products	S6
6. Demonstration of Synthetic Utility	S21
7. Mechanistic Investigations	S24
8. X-Ray Structure of Product 3aa	S44
9. Copies of NMR Spectra	S46
10. Copies of HPLC Spectra	S83

1. General Information

NMR spectra: ^1H NMR spectra were recorded on a 400 MHz spectrometer. Chemical shifts are reported in parts per million (ppm) and the spectra are calibrated to the resonance resulting from incomplete deuteration of the solvent (CDCl_3 : 7.26 ppm). ^{13}C NMR spectra were recorded on the same spectrometer with complete proton decoupling. Chemical shifts are reported in ppm with the solvent resonance as the internal standard ($^{13}\text{CDCl}_3$: 77.0 ppm). Data are reported as follows: chemical shift δ /ppm, integration (^1H only), multiplicity (s = singlet, d = doublet, t = triplet of doublets, m = multiplet or combinations thereof. ^{13}C signals are singlets unless otherwise stated), coupling constants J in Hz, assignment. ^{19}F NMR spectra were recorded on the same Spectrometer.

High Resolution Mass Spectrometry (HRMS): All were recorded on Bruker micrOTOF II ESI-TOF by ESI or APCI. Measured values are reported to 4 decimal places of the calculated value. The calculated values are based on the most abundant isotope.

Chromatography: Analytical thin layer chromatography was performed using Qingdao Puke Parting Materials Co. silica gel plates (Silicagel 60 F254). Visualisation was by ultraviolet fluorescence ($\lambda = 254$ nm) and/or staining with Phosphomolybdic acid or potassium permanganate (KMnO_4). Flash column chromatography was performed using 200-300 mesh silica gel. Optical rotations were measured with a polarimeter. $[\alpha]$. D values are reported at a given temperature ($^\circ\text{C}$) in degrees $\text{cm}^2 \text{ g}^{-1}$ with concentration in mg mL^{-1} .

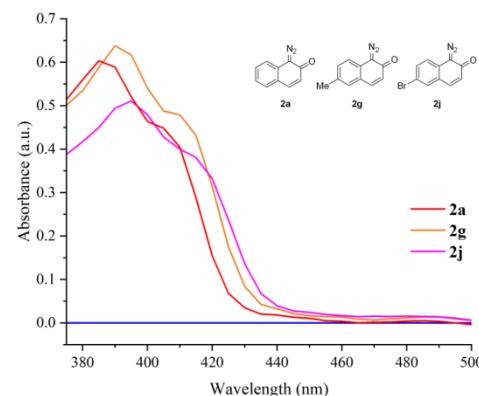
Chiral HPLC: Enantiomeric excesses (ee) values were determined by chiral HPLC with chiral AD-H, AZ-H, IA-H, IC-H, IF-H, IG-H columns with hexane and *i*-PrOH as solvents.

UV-vis: Measurements were made on a Shimadzu RF-6000 Spectro Fluorophotometer.

Materials: All the solvents were treated according to standard methods or through solvent purification systems before use. Substrates **1**^[1-3] and **2**^[4-6] were prepared according to previous methods and sulfides, copper salts and chiral ligands are commercially available.

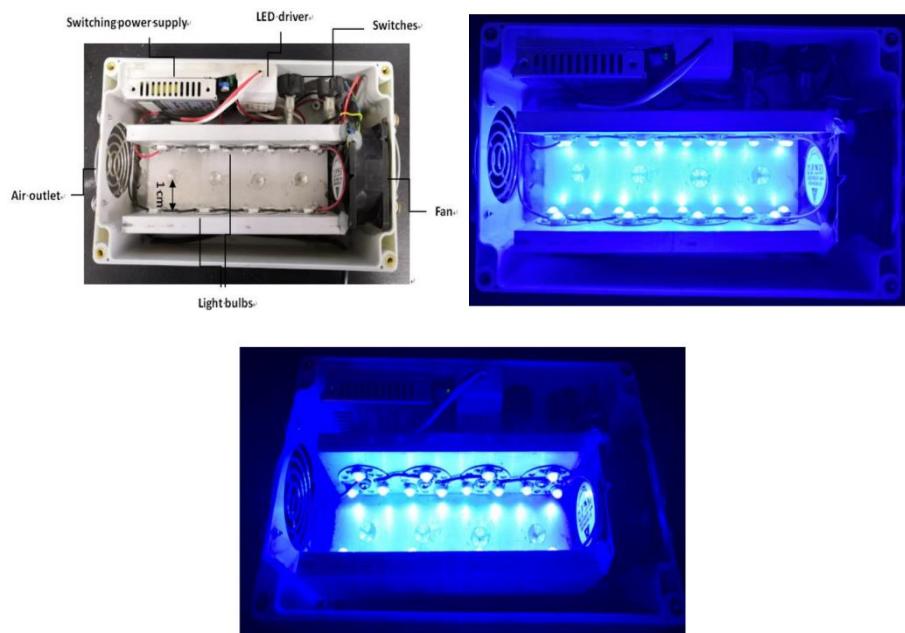
Photoreactor: The photoreactors used in this research were bought from GeAo Chem (blue LEDs, light intensity = 32.8 mw/cm², 1 W for every light bulb; every Schlenk tube was irradiated by 6 light bulbs from the side).

2. UV-vis Absorption Spectra



Supplementary Figure 1 UV-vis absorption spectra of **2a**, **2g** and **2j** (0.1 mmol/L in DCM)

3. Photoreactor Setup



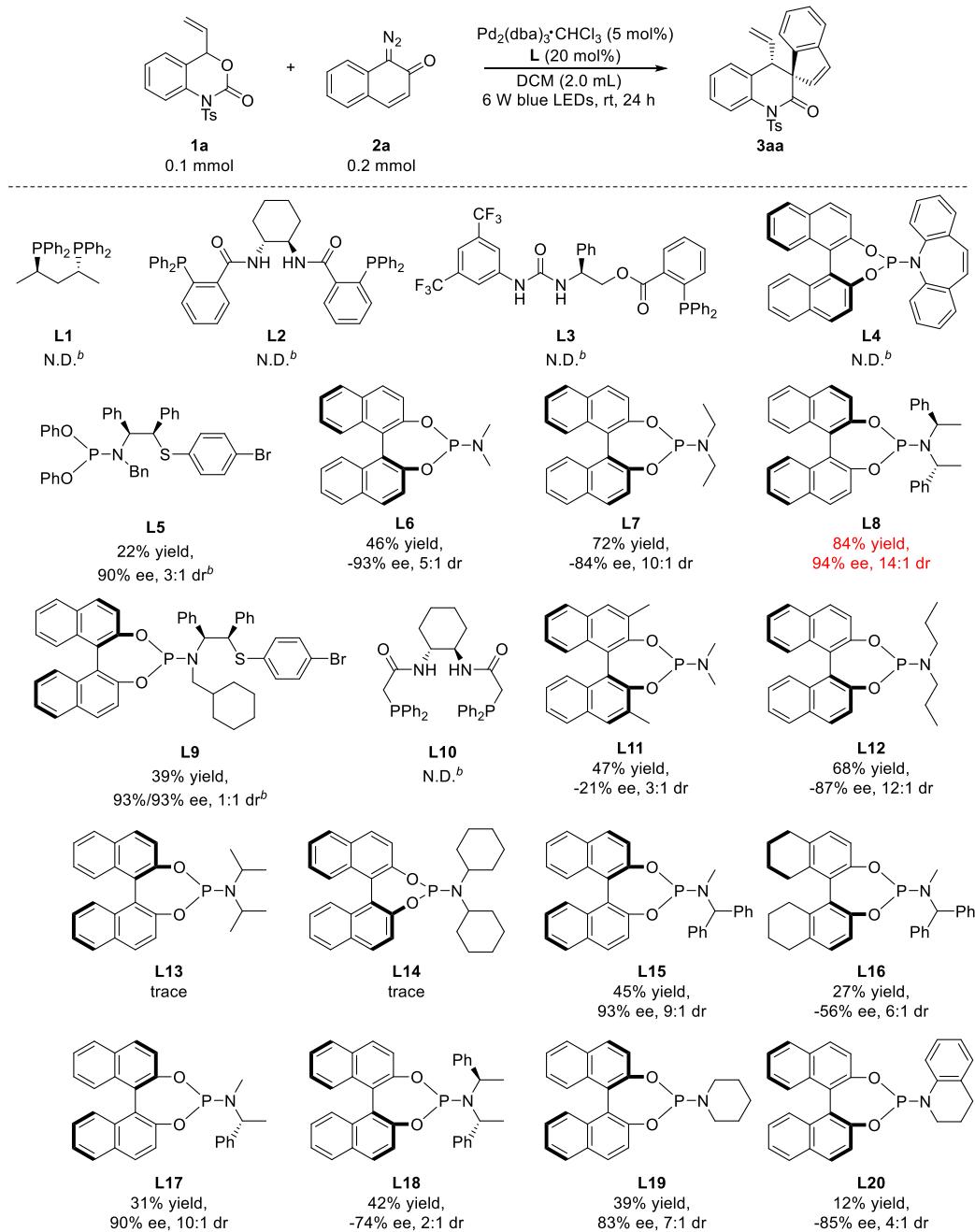
Supplementary Figure 2 Photoreactor used in this research (6 W blue LEDs, $\lambda_{\max} = 456$ nm)



Supplementary Figure 3 Photoreactor used in this research (20 W blue LEDs, $\lambda_{\max} = 456$ nm)

4. Details for Condition Optimizations

Supplementary Table 1. The Effect of Chiral Ligands^a



^aReaction conditions: **1a** (0.1 mmol), **2a** (0.2 mmol), $\text{Pd}_2(\text{dba})_3 \cdot \text{CHCl}_3$ (5 mol%) and chiral ligand (20 mol%) in DCM (2.0 mL) at rt for 24 h under the irradiation of 6 W blue LEDs ($\lambda_{\text{max}} = 456$ nm). Yield was determined by analyzing the ^1H NMR of reaction mixture with 1,3,5-trimethoxybenzene as an internal standard, N.D. = not detected. ee was determined by chiral HPLC and dr was determined by the ^1H NMR analysis of the reaction mixture. ^bLigand (10 mol%) was used instead.

Supplementary Table 2. The Effect of Light Sources^a

entry	light source	yield ^b (%)	dr ^c	ee ^d (%)
1	6 W 370 nm white LEDs	52	10:1	91
2	6 W 390 nm purple LEDs	57	11:1	92
3	6 W 456 nm blue LEDs	84	14:1	94

^aReaction conditions: **1a** (0.1 mmol), **2a** (0.2 mmol), $\text{Pd}_2(\text{dba})_3 \cdot \text{CHCl}_3$ (5 mol%) and **L8** (20 mol%) in DCM (2.0 mL) at rt for 24 h under the irradiation of 6 W different light source. ^bDetermined by analyzing the ¹H NMR of reaction mixture with 1,3,5-trimethoxybenzene as an internal standard.

^cDetermined by the ¹H NMR analysis of the reaction mixture. ^dDetermined by chiral HPLC.

Supplementary Table 3. The Effect of Solvents^a

entry	solvent	yield ^b (%)	dr ^c	ee ^d (%)
1	DMF	N.D.	-	-
2	DMSO	N.D.	-	-
3	DCM	84	14:1	94
4	CHCl ₃	41	8:1	93
5	Toluene	35	9:1	93
6	Acetone	38	10:1	94
7	EtOAc	51	11:1	91
8	Et ₂ O	44	9:1	90
9	THF	21	6:1	92

^aReaction conditions: **1a** (0.1 mmol), **II-2a** (0.2 mmol), $\text{Pd}_2(\text{dba})_3 \cdot \text{CHCl}_3$ (5 mol%) and **L8** (20 mol%) in solvent (2.0 mL) at rt for 24 h under the irradiation of 6 W blue LEDs ($\lambda_{\text{max}} = 456$ nm). ^bDetermined by analyzing the ¹H NMR of reaction mixture with 1,3,5-trimethoxybenzene as an internal standard, N.D. = not detected. ^cDetermined by the ¹H NMR analysis of the reaction mixture. ^dDetermined by chiral HPLC.

Supplementary Table 4. The Effect of Concentration^a

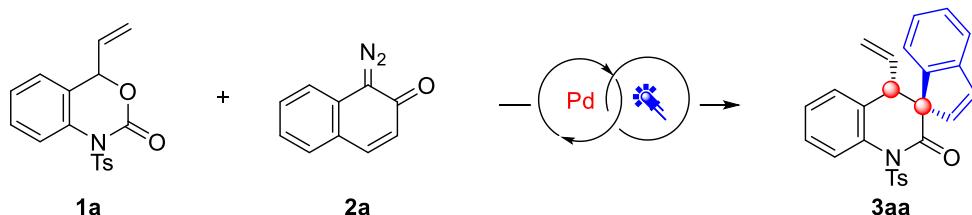
entry	2a (x mmol)	DCM (y mL)	yield ^b (%)	dr ^c	ee ^d (%)
1	0.1 mmol	2.0 mL	67	13:1	93
2	0.2 mmol	2.0 mL	84 (84) ^e	14:1	94
3	0.3 mmol	2.0 mL	82	14:1	94
4	0.2 mmol	1.5 mL	79	14:1	93
5	0.2 mmol	3.0 mL	81	13:1	93

^aReaction conditions: **1a** (0.1 mmol), **2a** (x mmol), $\text{Pd}_2(\text{dba})_3 \cdot \text{CHCl}_3$ (5 mol%) and **L8** (20 mol%) in DCM (y mL) at rt for 24 h under the irradiation of 6 W blue LEDs ($\lambda_{\text{max}} = 456$ nm). ^bDetermined by analyzing the ¹H NMR of reaction mixture with 1,3,5-trimethoxybenzene as an internal standard.

^cDetermined by the ¹H NMR analysis of the reaction mixture. ^dDetermined by chiral HPLC. ^eIsolated yield.

5. General Procedures and Characterization Data of Products

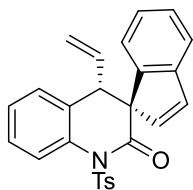
5.1 General Procedures



General procedure (one-pot procedure with product **3aa** as an example): Under argon atmosphere, a flame-dried 10 mL Schlenk tube was charged with $\text{Pd}_2(\text{dba})_3 \cdot \text{CHCl}_3$ (0.005 mmol, 5 mol%), **L8** (0.02 mmol, 20 mol%) and anhydrous DCM (1.0 mL) and the resulting solution was stirred for 30 min at room temperature. Then, vinylbenzoxazinanone **1a** (0.1 mmol, 1.0 equiv.), 1-diazonaphthalen-2(1H)-one **2a** (0.2 mmol, 2.0 equiv.) and anhydrous DCM (1.0 mL) were added to the reaction mixture. After that, the reaction solution was stirred under the irradiation of 6 W blue LEDs for 24 h at rt. The combined solution was concentrated under vacuum and the residue was purified by flash column chromatography on silica gel (petrol ether/ethyl acetate = 20/1 to 15/1) to afford the desired product **3aa**.

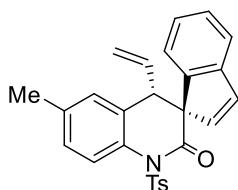
5.2 Characterization Data of Products

(1*S*,4'*S*)-1'-Tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3aa)



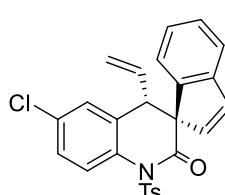
84% Isolated yield, colorless oil, $[\alpha]_D^{25} = -6.73$ ($c = 0.77$ in CHCl_3); 94% ee, 14:1 dr, determined by HPLC analysis (Chiralpak AD-H column, hexane/*i*-PrOH, 80:20 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 10.09 min, t_R (minor) = 14.73 min; **¹H NMR** (400 MHz, CDCl_3) δ (ppm) 8.07 – 7.96 (m, 2H), 7.79 (dd, $J = 8.2, 1.1$ Hz, 1H), 7.47 (td, $J = 7.9, 1.6$ Hz, 1H), 7.35 – 7.27 (m, 3H), 7.24 – 7.14 (m, 3H), 6.93 (td, $J = 7.4, 1.5$ Hz, 1H), 6.82 (d, $J = 5.6$ Hz, 1H), 6.25 (d, $J = 7.6$ Hz, 1H), 6.08 (d, $J = 5.6$ Hz, 1H), 5.71 – 5.62 (m, 1H), 5.17 – 5.07 (m, 2H), 3.59 (d, $J = 8.4$ Hz, 1H), 2.44 (s, 3H); **¹³C NMR** (100 MHz, CDCl_3) δ (ppm) 170.2, 145.1, 144.0, 143.8, 136.3, 134.9, 134.8, 134.6, 133.9, 131.3, 129.4, 129.1, 128.3, 127.9, 127.8, 126.7, 126.0, 123.5, 123.1, 121.8, 120.0, 65.9, 48.9, 21.7. **HRMS** (ESI) for $\text{C}_{26}\text{H}_{21}\text{NO}_3\text{S}$ [$\text{M}+\text{Na}$]⁺: calcd 450.1134, found 450.1137.

(1*S*,4'*S*)-6'-Methyl-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ab)



82% Isolated yield, colorless oil, $[\alpha]_D^{25} = 7.70$ ($c = 0.79$ in CHCl_3); 93% ee, 13:1 dr, determined by HPLC analysis (Chiralpak AD-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 0.5 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 29.71 min, t_R (minor) = 36.00 min; **¹H NMR** (400 MHz, CDCl_3) δ (ppm) 7.99 (d, $J = 8.1$ Hz, 2H), 7.67 (d, $J = 8.2$ Hz, 1H), 7.36 – 7.27 (m, 3H), 7.24 – 7.16 (m, 2H), 6.95 (d, $J = 6.6$ Hz, 2H), 6.81 (d, $J = 5.6$ Hz, 1H), 6.28 (d, $J = 7.7$ Hz, 1H), 6.09 (d, $J = 5.6$ Hz, 1H), 5.69 – 5.60 (m, 1H), 5.17 – 5.07 (m, 2H), 3.52 (d, $J = 8.4$ Hz, 1H), 2.44 (s, 3H), 2.37 (s, 3H); **¹³C NMR** (100 MHz, CDCl_3) δ (ppm) 170.2, 145.0, 144.0, 143.8, 136.7, 136.4, 134.7, 134.1, 132.5, 131.1, 129.4, 129.1, 129.0, 128.4, 128.3, 128.2, 126.0, 123.4, 123.2, 121.8, 119.8, 65.9, 48.9, 21.7, 21.0. **HRMS** (ESI) for $\text{C}_{27}\text{H}_{23}\text{NO}_3\text{S}$ [$\text{M}+\text{Na}$]⁺: calcd 464.1291, found 464.1289.

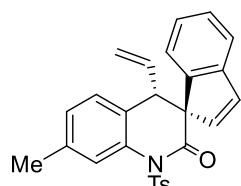
(1*S*,4'*S*)-6'-Chloro-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ac)



83% Isolated yield, colorless oil, $[\alpha]_D^{25} = -2.33$ ($c = 0.71$ in CHCl_3); 94% ee, 19:1 dr, determined by HPLC analysis (Chiralpak AD-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 14.02 min, t_R (minor) = 19.76 min; **¹H NMR** (400 MHz, CDCl_3) δ (ppm) 7.98 (d, $J = 8.3$ Hz, 2H, major+minor), 7.74 (d, $J = 8.7$ Hz, 1H, major+minor), 7.44 (dd, $J = 8.7, 2.4$ Hz, 1H, major+minor), 7.34 (d, $J = 8.1$ Hz, 2H, major+minor), 7.25 – 7.14 (m, 3H, major+minor), 7.01 (m, 1H, major+minor), 6.83 (d, $J = 5.6$ Hz, 1H, major), 6.76 (d, $J = 5.6$ Hz, 1H, minor), 6.41 (d, $J = 7.6$ Hz, 1H, major+minor), 6.00 (d, $J = 5.6$ Hz, 1H, major+minor), 5.56 – 5.48 (m, 1H, major+minor), 5.26 – 5.20 (m, 2H, minor), 5.17 – 5.08 (m, 2H, major), 3.63 (d, $J = 8.5$ Hz, 1H, major+minor), 2.45 (s, 3H, major+minor); **¹³C NMR** (100 MHz, CDCl_3) δ

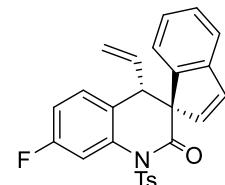
(ppm) 169.7, 145.4, 143.8, 143.7, 136.0, 135.3, 133.5, 133.4, 133.3, 132.7, 132.3, 129.5, 129.1, 128.4, 127.9, 127.6, 126.3, 124.8, 123.1, 122.0, 120.8, 65.7, 48.4, 21.7. **HRMS** (ESI) for C₂₆H₂₀ClNO₃S [M+Na]⁺: calcd 484.0745, found 484.0750.

(1*S*,4'*S*)-7'-Methyl-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ad)



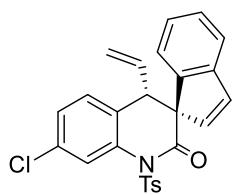
78% Isolated yield, white semi-solid, $[\alpha]_D^{25} = -7.73$ ($c = 0.81$ in CHCl₃); 94% ee, 17:1 dr, determined by HPLC analysis (Chiralpak IA-H column, hexane/i-PrOH, 95:5 v/v, flow rate 0.3 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 43.73 min, t_R (minor) = 54.77 min; **¹H NMR** (400 MHz, CDCl₃) δ (ppm) 7.99 (d, $J = 8.0$ Hz, 2H, major+minor), 7.61 (s, 1H, major+minor), 7.33 (d, $J = 8.1$ Hz, 2H, major+minor), 7.23 – 7.16 (m, 2H, major+minor), 7.09 (d, $J = 7.6$ Hz, 1H, major+minor), 7.02 (d, $J = 7.7$ Hz, 1H, major+minor), 6.94 (t, $J = 7.4$, 1.5 Hz, 1H, major+minor), 6.81 (d, $J = 5.6$ Hz, 1H, major), 6.73 (d, $J = 5.6$ Hz, 1H, minor), 6.25 (d, $J = 7.7$ Hz, 1H, major+minor), 6.08 (d, $J = 5.6$ Hz, 1H, major), 5.96 (d, $J = 5.6$ Hz, 1H, minor), 5.69 – 5.60 (m, 1H, major+minor), 5.21 – 5.17 (m, 2H, minor), 5.13 – 5.07 (m, 2H, major), 3.53 (d, $J = 8.3$ Hz, 1H, major+minor), 2.50 (s, 3H, major+minor), 2.45 (s, 3H, major+minor); **¹³C NMR** (100 MHz, CDCl₃) δ (ppm) 170.3, 145.0, 144.1, 143.8, 137.9, 136.4, 134.8, 134.7, 134.2, 129.4, 129.4, 129.1, 128.2, 128.2, 127.6, 127.4, 126.0, 124.1, 123.2, 121.8, 119.7, 66.0, 48.6, 21.7, 21.6. **HRMS** (ESI) for C₂₇H₂₃NO₃S [M+Na]⁺: calcd 464.1291, found 464.1290.

(1*S*,4'*S*)-7'-Fluoro-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ae)



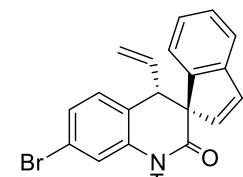
80% Isolated yield, colorless oil, $[\alpha]_D^{25} = -12.67$ ($c = 0.71$ in CHCl₃); 94% ee, 9:1 dr, determined by HPLC analysis (Chiralpak IA-H column, hexane/i-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 11.03 min, t_R (minor) = 13.76 min; **¹H NMR** (400 MHz, CDCl₃) δ (ppm) 8.00 (d, $J = 8.1$ Hz, 2H, major+minor), 7.59 (dd, $J = 10.2$, 2.5 Hz, 1H, major+minor), 7.34 (d, $J = 8.1$ Hz, 2H, major+minor), 7.24 – 7.19 (m, 2H, major+minor), 7.13 – 7.10 (m, 1H, major+minor), 7.02 – 6.96 (m, 2H, major+minor), 6.84 (d, $J = 5.6$ Hz, 1H, major), 6.77 (d, $J = 5.6$ Hz, 1H, minor), 6.33 (d, $J = 7.7$ Hz, 1H, major+minor), 6.08 (d, $J = 5.7$ Hz, 1H, major), 5.98 (d, $J = 5.7$ Hz, 1H, minor), 5.68 – 5.59 (m, 1H, major+minor), 5.24 – 5.19 (m, 2H, minor), 5.16 – 5.08 (m, 2H, major), 3.58 (d, $J = 8.3$ Hz, 1H, major+minor), 2.45 (s, 3H, major+minor). **¹³C NMR** (100 MHz, CDCl₃) δ (ppm) 169.8, 161.7 (d, $J = 244.8$ Hz), 145.4, 143.8, 136.0, 135.9 (d, $J = 10.6$ Hz), 135.0, 134.2, 133.7, 129.5, 129.1, 129.0, 128.9 (d, $J = 9.1$ Hz), 128.4, 127.0 (d, $J = 3.2$ Hz), 126.1, 123.0, 122.0, 120.2, 113.4 (d, $J = 21.2$ Hz), 111.3 (d, $J = 26.6$ Hz), 65.7, 48.3, 21.7. **¹⁹F NMR** (376 MHz, CDCl₃) δ (ppm) -112.15, -112.29. **HRMS** (ESI) for C₂₆H₂₀FNO₃S [M+Na]⁺: calcd 468.1040, found 468.1033.

(1*S*,4'*S*)-7'-Chloro-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3af)



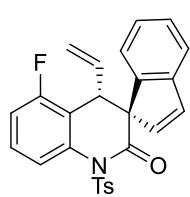
76% Isolated yield, white solid, $[\alpha]_D^{25} = -1.77$ ($c = 0.52$ in CHCl_3); 97% ee, 11:1 dr, determined by HPLC analysis (Chiralpak IA-H column, hexane/*i*-PrOH, 95:5 v/v, flow rate 0.3 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 46.43 min, t_R (minor) = 56.03 min; **$^1\text{H NMR}$** (400 MHz, CDCl_3) δ (ppm) 8.01 – 7.97 (m, 2H, major+minor), 7.82 (d, $J = 2.0$ Hz, 1H, major+minor), 7.34 (d, $J = 8.1$ Hz, 2H, major+minor), 7.24 – 7.18 (m, 2H, major+minor), 7.09 (d, $J = 8.2$ Hz, 1H, major+minor), 7.01 – 6.95 (m, 1H, major+minor), 6.83 (d, $J = 5.6$ Hz, 1H, major), 6.76 (d, $J = 5.6$ Hz, 1H, minor), 6.48 (d, $J = 7.6$ Hz, 1H, minor), 6.37 (d, $J = 7.6$ Hz, 1H, major), 6.05 (d, $J = 5.6$ Hz, 1H, major), 5.97 (d, $J = 5.6$ Hz, 1H, minor), 5.64 – 5.55 (m, 1H, major+minor), 5.25 – 5.18 (m, 2H, minor), 5.16 – 5.08 (m, 2H, major), 3.59 (d, $J = 8.4$ Hz, 1H, major+minor), 2.45 (s, 3H, major+minor); **$^{13}\text{C NMR}$** (100 MHz, CDCl_3) δ (ppm) 169.7, 145.4, 143.8, 143.7, 135.9, 135.8, 135.1, 133.9, 133.4, 133.3, 129.8, 129.5, 129.1, 128.7, 128.4, 126.7, 126.2, 123.6, 123.0, 122.0, 120.4, 65.6, 48.3, 21.7. **HRMS** (ESI) for $\text{C}_{26}\text{H}_{20}\text{ClNO}_3\text{S}$ [M+Na] $^+$: calcd 484.0745, found 484.0737.

(1*S*,4'*S*)-7'-Bromo-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ag)



81% Isolated yield, colorless oil, $[\alpha]_D^{25} = -2.55$ ($c = 0.46$ in CHCl_3); 96% ee, 13:1 dr, determined by HPLC analysis (Chiralpak IA-H column, hexane/*i*-PrOH, 97:3 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 40.43 min, t_R (minor) = 49.45 min; **$^1\text{H NMR}$** (400 MHz, CDCl_3) δ (ppm) 8.01 – 7.96 (m, 3H, major+minor), 7.42 (dd, $J = 8.1, 1.8$ Hz, 1H, major+minor), 7.35 (d, $J = 8.1$ Hz, 2H, major+minor), 7.24 – 7.19 (m, 2H, major+minor), 7.04 – 6.97 (m, 2H, major+minor), 6.83 (d, $J = 5.6$ Hz, 1H, major), 6.76 (d, $J = 5.6$ Hz, 1H, minor), 6.37 (d, $J = 7.7$ Hz, 1H, major+minor), 6.05 (d, $J = 5.6$ Hz, 1H, major), 5.96 (d, $J = 5.6$ Hz, 1H, minor), 5.63 – 5.54 (m, 1H, major+minor), 5.25 – 5.18 (m, 2H, minor), 5.16 – 5.08 (m, 2H, major), 3.58 (d, $J = 8.4$ Hz, 1H, major+minor), 2.45 (s, 3H); **$^{13}\text{C NMR}$** (100 MHz, CDCl_3) δ (ppm) 169.7, 145.4, 143.8, 143.7, 135.9, 135.9, 135.2, 133.9, 133.2, 130.3, 129.6, 129.5, 129.1, 129.0, 128.5, 126.4, 126.2, 123.1, 122.0, 121.1, 120.5, 65.6, 48.4, 21.7. **HRMS** (ESI) for $\text{C}_{26}\text{H}_{20}\text{BrNO}_3\text{S}$ [M+Na] $^+$: calcd 528.0239, found 528.0230.

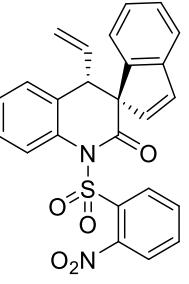
(1*S*,4'*R*)-5'-Fluoro-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ah)



68% Isolated yield, colorless oil, $[\alpha]_D^{25} = 107.77$ ($c = 1.02$ in CHCl_3); 81% ee, 5:1 dr, determined by HPLC analysis (Chiralpak AD-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 0.3 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 47.58 min, t_R (minor) = 38.78 min; **$^1\text{H NMR}$** (400 MHz, CDCl_3) δ (ppm) 8.01 – 7.96 (m, 2H, major+minor), 7.69 (d, $J = 8.3$ Hz, 1H, major), 7.63 (d, $J = 8.3$ Hz, 1H, minor), 7.51 – 7.45 (m, 1H, major), 7.42 – 7.37 (m, 1H, minor), 7.34 – 7.29 (m, 2H, major+minor), 7.25 – 7.15 (m, 2H, major+minor), 7.06

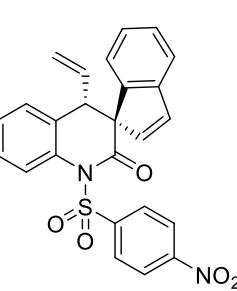
(td, $J = 8.4$, 1.0 Hz, 1H, major+minor), 6.89 (d, $J = 5.7$ Hz, 1H, major), 6.78 (td, $J = 7.6$, 1.3 Hz, 1H, major+minor), 6.72 (d, $J = 5.7$ Hz, 1H, minor), 6.44 (d, $J = 5.7$ Hz, 1H, major+minor), 6.09 – 5.95 (m, 1H, major+minor), 5.82 (d, $J = 5.7$ Hz, 1H, major), 5.70 (d, $J = 5.7$ Hz, 1H, minor), 5.28 – 5.10 (m, 2H, major+minor), 3.55 (d, $J = 7.7$ Hz, 1H, major), 3.47 (d, $J = 7.7$ Hz, 1H, minor), 2.45 (s, 3H, minor), 2.43 (s, 3H, major); ^{13}C NMR δ (ppm) 169.4, 160.0 (d, $J = 245.7$ Hz), 145.4, 143.6, 143.1, 137.1, 136.7 (d, $J = 5.4$ Hz), 135.9, 134.7, 134.3, 129.4, 129.3, 128.8 (d, $J = 9.1$ Hz), 128.7, 125.9, 122.2 (d, $J = 11.7$ Hz), 119.3, 119.3, 119.3, 118.8 (d, $J = 20.5$ Hz), 113.5 (d, $J = 21.7$ Hz), 64.3, 42.78, 21.7; ^{19}F NMR (376 MHz, CDCl_3) δ (ppm) -118.11, -118.83. HRMS (ESI) for $\text{C}_{26}\text{H}_{20}\text{FNO}_3\text{S} [\text{M}+\text{Na}]^+$: calcd 468.1040, found 468.1036.

(1*S*,4'*S*)-1'-(2-Nitrophenyl)sulfonyl)-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ai)



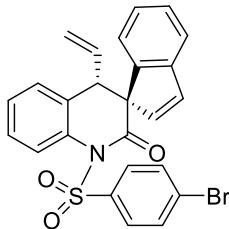
76% Isolated yield, colorless oil, $[\alpha]_D^{25} = -64.93$ ($c = 0.91$ in CHCl_3); 92% ee, 14:1 dr, determined by HPLC analysis (Chiralpak IC-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 0.5 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 32.63 min, t_R (minor) = 30.20 min; ^1H NMR (400 MHz, CDCl_3) δ (ppm) 8.45 (dd, $J = 7.6$, 1.9 Hz, 1H), 7.87 (dd, $J = 7.5$, 1.9 Hz, 1H), 7.81 – 7.74 (m, 3H), 7.47 – 7.42 (m, 1H), 7.34 – 7.29 (m, 1H), 7.26 – 7.20 (m, 3H), 7.06 (m, 1H), 6.83 (d, $J = 5.6$ Hz, 1H), 6.72 (d, $J = 7.6$ Hz, 1H), 5.93 (d, $J = 5.6$ Hz, 1H), 5.44 – 5.35 (m, 1H), 5.13 – 5.07 (m, 2H), 4.23 (d, $J = 9.2$ Hz, 1H); ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 170.0, 147.7, 144.1, 144.0, 135.6, 135.5, 134.7, 133.8, 133.6, 132.3, 132.1, 132.0, 131.3, 128.3, 127.7, 127.1, 126.9, 126.3, 124.6, 123.4, 122.9, 121.9, 121.0, 65.9, 47.6. HRMS (ESI) for $\text{C}_{25}\text{H}_{18}\text{N}_2\text{O}_5\text{S} [\text{M}+\text{Na}]^+$: calcd 481.0829, found 481.0831.

(1*S*,4'*S*)-1'-(4-Nitrophenyl)sulfonyl)-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3aj)



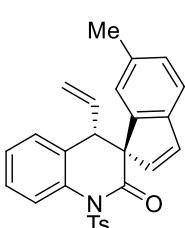
74% Isolated yield, colorless oil, $[\alpha]_D^{25} = -35.57$ ($c = 0.96$ in CHCl_3); 92% ee, 11:1 dr, determined by HPLC analysis (Chiralpak IA-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 18.15 min, t_R (minor) = 33.49 min; ^1H NMR (400 MHz, CDCl_3) δ (ppm) 8.39 – 8.35 (m, 2H, major+minor), 8.30 (m, 2H, major+minor), 7.79 (d, $J = 8.2$ Hz, 1H, major+minor), 7.52 (t, $J = 7.9$ Hz, 1H, major+minor), 7.34 (t, $J = 7.5$ Hz, 1H, major+minor), 7.21 (m, 3H, major+minor), 6.95 (t, $J = 7.5$ Hz, 1H, major+minor), 6.86 (d, $J = 5.6$ Hz, 1H, major), 6.77 (d, $J = 5.6$ Hz, 1H, minor), 6.25 (d, $J = 7.6$ Hz, 1H, major+minor), 6.10 (d, $J = 5.6$ Hz, 1H, major), 5.95 (d, $J = 5.6$ Hz, 1H, minor), 5.74 – 5.65 (m, 1H, major+minor), 5.28 – 5.25 (m, 2H, minor), 5.21 – 5.10 (m, 2H, major), 3.61 (d, $J = 8.0$ Hz, 1H, major+minor); ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 170.6, 150.6, 144.8, 143.7, 143.4, 135.3, 134.4, 134.0, 133.8, 131.2, 130.5, 128.6, 128.4, 128.2, 127.2, 126.2, 124.0, 123.2, 123.0, 122.1, 120.4, 65.6, 48.8. HRMS (ESI) for $\text{C}_{25}\text{H}_{18}\text{N}_2\text{O}_5\text{S} [\text{M}+\text{Na}]^+$: calcd 481.0829, found 481.0824.

(1*S*,4'*S*)-1'-(*(4*-Bromophenyl)sulfonyl)-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ak)



79% Isolated yield, colorless oil, $[\alpha]_D^{25} = -34.40$ ($c = 0.92$ in CHCl_3); 92% ee, 10:1 dr, determined by HPLC analysis (Chiralpak IA-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 12.72 min, t_R (minor) = 26.01 min; **¹H NMR** (400 MHz, CDCl_3) δ (ppm) 8.07 – 8.05 (m, 2H, minor), 8.00 – 7.96 (m, 2H, major), 7.76 (d, $J = 8.2$ Hz, 1H, major+minor), 7.69 – 7.66 (m, 2H, major+minor), 7.48 (t, $J = 7.8$ Hz, 1H, major+minor), 7.30 (t, $J = 7.5$ Hz, 1H, major+minor), 7.23 – 7.14 (m, 3H, major+minor), 6.95 (t, $J = 7.5$ Hz, 1H, major+minor), 6.84 (d, $J = 5.6$ Hz, 1H, major), 6.76 (d, $J = 5.6$ Hz, 1H, minor), 6.41 (d, $J = 7.6$ Hz, 1H, minor), 6.25 (d, $J = 7.6$ Hz, 1H, major), 6.09 (d, $J = 5.6$ Hz, 1H, major), 5.96 (d, $J = 5.6$ Hz, 1H, minor), 5.72 – 5.63 (m, 1H, major+minor), 5.25 – 5.19 (m, 2H, minor), 5.18 – 5.09 (m, 2H, major), 3.68 (d, $J = 8.2$ Hz, 1H, minor), 3.59 (d, $J = 8.2$ Hz, 1H, major); **¹³C NMR** (100 MHz, CDCl_3) δ (ppm) 170.3, 143.8, 143.8, 138.2, 135.0, 134.7, 134.3, 133.8, 132.2, 131.1, 130.6, 129.3, 128.4, 128.1, 128.0, 126.9, 126.1, 123.4, 123.1, 121.9, 120.2, 65.8, 48.8. **HRMS** (ESI) for $\text{C}_{25}\text{H}_{18}\text{BrNO}_3\text{S}$ [M+Na]⁺: calcd 514.0083, found 514.0086.

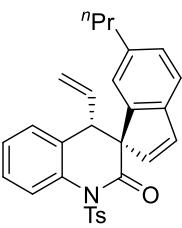
(1*S*,4'*S*)-6-Methyl-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ba)



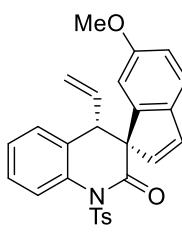
86% Isolated yield, colorless oil, $[\alpha]_D^{25} = -20.60$ ($c = 0.84$ in CHCl_3); 90% ee, 10:1 dr, determined by HPLC analysis (Chiralpak IA-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 13.95 min, t_R (minor) = 16.60 min; **¹H NMR** (400 MHz, CDCl_3) δ (ppm) 8.02 (d, $J = 8.2$ Hz, 1H, major+minor), 7.77 (d, $J = 8.2$ Hz, 1H, major+minor), 7.49 – 7.44 (m, 1H, major+minor), 7.34 (d, $J = 8.2$ Hz, 2H, major+minor), 7.29 (d, $J = 7.6$ Hz, 1H, major+minor), 7.17 – 7.09 (m, 2H, major+minor), 7.00 (d, $J = 7.7$ Hz, 1H, major+minor), 6.77 (d, $J = 5.6$ Hz, 1H, major), 6.70 (d, $J = 5.6$ Hz, 1H, minor), 6.09 (s, 1H, major+minor), 5.94 (d, $J = 5.6$ Hz, 1H, major), 5.85 (d, $J = 5.6$ Hz, 1H, minor), 5.64 – 5.56 (m, 1H, major+minor), 5.26 – 5.19 (m, 2H, minor), 5.14 – 5.08 (m, 2H, major), 3.63 (d, $J = 8.4$ Hz, 1H, major+minor), 2.45 (s, 3H, major+minor), 2.14 (s, 3H, major+minor); **¹³C NMR** (100 MHz, CDCl_3) δ (ppm) 170.5, 145.1, 144.4, 141.2, 136.3, 135.8, 134.9, 134.8, 133.6, 133.1, 131.5, 129.4, 129.2, 128.9, 127.7, 127.7, 126.6, 124.1, 123.5, 121.4, 120.1, 65.9, 48.7, 21.7, 21.5. **HRMS** (ESI) for $\text{C}_{27}\text{H}_{23}\text{NO}_3\text{S}$ [M+Na]⁺: calcd 464.1291, found 464.1298.

(1*S*,4'*S*)-6-Isopropyl-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ca)

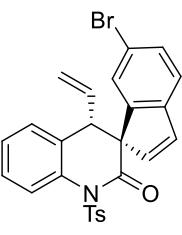
81% Isolated yield, colorless oil, $[\alpha]_D^{25} = -2.80$ ($c = 0.68$ in CHCl_3); 92% ee, 10:1 dr, determined by HPLC analysis (Chiralpak AD-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 12.88 min, t_R (minor) = 11.58 min; **¹H NMR** (400 MHz, CDCl_3) δ (ppm) 8.02 – 7.98 (d, $J = 8.2$ Hz, 1H, major+minor), 7.83 (d,


 $J = 8.2$ Hz, 1H, major+minor), 7.50 – 7.46 (m, 1H, major+minor), 7.34 – 7.27 (m, 3H, major+minor), 7.16 – 7.10 (m, 2H, major+minor), 7.02 (dd, $J = 7.8, 1.5$ Hz, 1H, major+minor), 6.79 (d, $J = 5.6$ Hz, 1H, major), 6.71 (d, $J = 5.6$ Hz, 1H, minor), 6.06 (d, $J = 5.7$ Hz, 1H, major+minor), 5.98 (d, $J = 5.6$ Hz, 1H, major), 5.92 (d, $J = 5.6$ Hz, 1H, minor), 5.76 – 5.67 (m, 1H, major+minor), 5.24 – 5.19 (m, 2H, minor), 5.15 – 5.07 (m, 2H, major), 3.48 (d, $J = 8.2$ Hz, 1H, major+minor), 2.68 – 2.61 (m, 1H, major+minor), 2.44 (s, 3H, major+minor), 1.00 (d, $J = 1.9$ Hz, 3H, major+minor), 0.99 (d, $J = 1.9$ Hz, 3H, major+minor); ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 170.3, 146.7, 145.0, 144.2, 141.4, 136.4, 135.2, 134.5, 134.3, 134.0, 131.4, 129.3, 129.2, 128.1, 127.9, 126.6, 126.6, 123.7, 121.4, 121.2, 119.7, 65.6, 49.3, 33.9, 24.1, 23.2, 21.7. HRMS (ESI) for $\text{C}_{29}\text{H}_{27}\text{NO}_3\text{S}$ [M+Na] $^+$: calcd 492.1604, found 492.1597.

(1*S*,4'*S*)-6-Methoxy-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3da)


 78% Isolated yield, colorless oil, $[\alpha]_D^{25} = -30.27$ ($c = 1.05$ in CHCl_3); 94% ee, 11:1 dr, determined by HPLC analysis (Chiralpak IA-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 0.6 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 27.29 min, t_R (minor) = 74.31 min; ^1H NMR (400 MHz, CDCl_3) δ (ppm) 8.01 (d, $J = 8.1$ Hz, 2H, major+minor), 7.79 (d, $J = 8.1$ Hz, 1H, major+minor), 7.49 – 7.44 (m, 1H, major+minor), 7.35 – 7.28 (m, 3H, major+minor), 7.17 (d, $J = 7.6$ Hz, 1H, major+minor), 7.11 (d, $J = 8.2$ Hz, 1H, major+minor), 6.77 – 6.70 (m, 2H, major+minor), 5.93 (d, $J = 5.6$ Hz, 1H, major+minor), 5.82 (s, 1H, major+minor), 5.69 – 5.60 (m, 1H, major+minor), 5.25 – 5.20 (m, 2H, minor), 5.16 – 5.10 (m, 2H, major), 3.58 (s, 1H, major+minor), 3.55 (s, 3H, major+minor), 2.44 (s, 3H, major+minor); ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 170.2, 158.3, 145.8, 145.1, 136.6, 136.3, 135.0, 134.4, 133.8, 132.2, 131.3, 129.4, 129.2, 128.0, 127.8, 126.7, 123.6, 122.1, 120.0, 113.5, 109.9, 65.8, 55.2, 48.9, 21.7. HRMS (ESI) for $\text{C}_{27}\text{H}_{23}\text{NO}_4\text{S}$ [M+Na] $^+$: calcd 480.1240, found 480.1247.

(1*S*,4'*S*)-6-Bromo-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ea)


 74% Isolated yield, colorless oil, $[\alpha]_D^{25} = -1.57$ ($c = 0.89$ in CHCl_3); 90% ee, 6:1 dr, determined by HPLC analysis (Chiralpak AD-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 0.5 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 39.61 min, t_R (minor) = 37.27 min; ^1H NMR (400 MHz, CDCl_3) δ (ppm) 8.04 (d, $J = 8.2$ Hz, 2H, major+minor), 7.77 (d, $J = 8.2$ Hz, 1H, major+minor), 7.51 – 7.47 (m, 1H, major+minor), 7.38 – 7.31 (m, 4H, major+minor), 7.17 (d, $J = 7.6$ Hz, 1H, major+minor), 7.08 (d, $J = 8.0$ Hz, 1H, major+minor), 6.76 (d, $J = 5.6$ Hz, 1H, major), 6.69 (d, $J = 5.6$ Hz, 1H, minor), 6.34 (s, 1H, major+minor), 6.01 (d, $J = 5.6$ Hz, 1H, major), 5.95 (d, $J = 5.6$ Hz, 1H, minor), 5.61 – 5.52 (m, 1H, major+minor), 5.28 – 5.21 (m, 2H, minor), 5.17 – 5.09 (m, 2H, major), 3.64 (d, $J = 8.6$ Hz, 1H, major+minor), 2.48 (s, 3H, major), 2.46 (s, 3H, minor); ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 169.6, 146.2, 145.4, 142.8, 136.0, 134.7, 134.4, 134.3,

132.7, 131.3, 131.0, 129.5, 129.2, 128.0, 127.5, 126.9, 126.6, 123.8, 122.9, 120.8, 120.1, 66.4, 48.4, 21.8. **HRMS** (ESI) for $C_{26}H_{20}BrNO_3S$ $[M+Na]^+$: calcd 528.0239, found 528.0233.

(1*S*,4'*S*)-2'-Oxo-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-6-yl acetate (3fa)

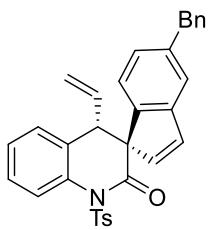
84% Isolated yield, colorless oil, $[\alpha]_D^{25} = -39.67$ ($c = 0.88$ in $CHCl_3$); 93% ee, 14:1 dr, determined by HPLC analysis (Chiralpak IA-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 22.53 min, t_R (minor) = 41.66 min; **1H NMR** (400 MHz, $CDCl_3$) δ (ppm) 8.03 – 7.99 (m, 2H, major+minor), 7.77 (dd, $J = 8.2, 1.1$ Hz, 1H, major+minor), 7.46 (td, $J = 7.9, 1.6$ Hz, 1H, major+minor), 7.36 – 7.28 (m, 3H, major+minor), 7.22 – 7.16 (m, 2H, major+minor), 6.94 (dd, $J = 8.1, 2.1$ Hz, 1H, major+minor), 6.78 (d, $J = 5.6$ Hz, 1H, major), 6.70 (d, $J = 5.6$ Hz, 1H, minor), 6.10 – 5.98 (m, 2H, major), 5.92 – 5.90 (m, 2H, minor), 5.68 – 5.59 (m, 1H, major+minor), 5.28 – 5.20 (m, 2H, minor), 5.18 – 5.12 (m, 2H, major), 3.64 (d, $J = 8.5$ Hz, 1H, major+minor), 2.45 (s, 3H, major+minor), 2.20 (s, 3H, major+minor); **13C NMR** (100 MHz, $CDCl_3$) δ (ppm) 169.8, 169.1, 149.0, 145.3, 145.2, 141.4, 136.3, 134.8, 134.2, 131.2, 129.6, 129.5, 129.1, 128.9, 127.9, 127.8, 126.8, 123.6, 122.0, 121.4, 120.5, 117.3, 66.3, 48.7, 21.7, 21.1. **HRMS** (ESI) for $C_{28}H_{23}NO_5S$ $[M+Na]^+$: calcd 508.1189, found 508.1194.

(1*S*,4'*S*)-5-Methyl-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ga)

80% Isolated yield, colorless oil, $[\alpha]_D^{25} = -22.53$ ($c = 0.94$ in $CHCl_3$); 93% ee, 14:1 dr, determined by HPLC analysis (Chiralpak IA-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 15.67 min, t_R (minor) = 28.49 min; **1H NMR** (400 MHz, $CDCl_3$) δ (ppm) 8.00 (d, $J = 8.0$ Hz, 2H, major+minor), 7.79 (d, $J = 8.2$ Hz, 1H, major), 7.67 (d, $J = 8.2$ Hz, 1H, minor), 7.46 (t, $J = 7.9$ Hz, 1H, major+minor), 7.33 – 7.27 (m, 3H, major+minor), 7.14 (d, $J = 7.5$ Hz, 1H, major+minor), 7.04 (s, 1H, major+minor), 6.78 – 6.73 (m, 2H, major), 6.70 – 6.69 (m, 2H, minor), 6.11 – 6.07 (m, 2H, major), 5.95 – 5.94 (m, 2H, minor), 5.73 – 5.64 (m, 1H, major+minor), 5.23 – 5.19 (m, 2H, minor), 5.16 – 5.09 (m, 2H, major), 3.55 (d, $J = 8.3$ Hz, 1H, major+minor), 2.44 (s, 3H, major+minor), 2.26 (s, 3H, major+minor); **13C NMR** (100 MHz, $CDCl_3$) δ (ppm) 170.4, 145.1, 144.1, 141.2, 138.2, 136.4, 135.0, 134.1, 131.4, 129.5, 129.4, 129.2, 129.1, 127.9, 127.9, 126.8, 126.7, 123.6, 122.8, 122.7, 119.9, 65.6, 49.1, 21.8, 21.4. **HRMS** (ESI) for $C_{27}H_{23}NO_3S$ $[M+Na]^+$: calcd 464.1291, found 464.1285.

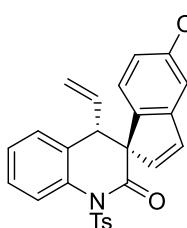
(1*S*,4'*S*)-5-Benzyl-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ha)

79% Isolated yield, colorless oil, $[\alpha]_D^{25} = -3.57$ ($c = 0.87$ in $CHCl_3$); 94% ee, 13:1 dr, determined by HPLC analysis (Chiralpak IA-H column, hexane/*i*-PrOH, 90:10 v/v, flow



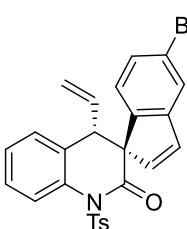
rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 26.93 min, t_R (minor) = 32.38 min; **¹H NMR** (400 MHz, CDCl₃) δ (ppm) 7.99 (d, $J = 8.4$ Hz, 2H, major+minor), 7.78 (dd, $J = 8.2, 1.1$ Hz, 1H, major), 7.69 (dd, $J = 8.2, 1.1$ Hz, 1H, minor), 7.45 (td, $J = 7.9, 1.6$ Hz, 1H, major+minor), 7.34 – 7.27 (m, 3H, major+minor), 7.26 – 7.23 (m, 2H, major+minor), 7.20 – 7.11 (m, 4H, major+minor), 7.04 (d, $J = 1.6$ Hz, 1H, major+minor), 6.77 (d, $J = 5.6$ Hz, 1H, major+minor), 6.74 – 6.70 (m, 1H, major+minor), 6.13 (d, $J = 5.7$ Hz, 1H, major+minor), 6.05 (d, $J = 7.9$ Hz, 1H, major), 6.00 (d, $J = 7.9$ Hz, 1H, minor), 5.77 – 5.69 (m, 1H, major+minor), 5.24 – 5.20 (m, 2H, minor), 5.17 – 5.09 (m, 2H, major), 3.89 (s, 2H, major+minor), 3.49 (d, $J = 8.2$ Hz, 1H, major+minor), 2.44 (s, 3H, major+minor); **¹³C NMR** (100 MHz, CDCl₃) δ (ppm) 170.2, 145.1, 144.2, 141.7, 141.5, 140.7, 136.3, 135.4, 135.0, 134.6, 134.4, 131.3, 129.4, 129.1, 129.0, 128.4, 128.1, 127.9, 126.7, 126.6, 126.1, 123.5, 123.0, 122.4, 119.7, 65.4, 49.2, 41.7, 21.7. **HRMS** (ESI) for C₃₃H₂₇NO₃O₂ [M+Na]⁺: calcd 540.1604, found 540.1601.

(1S,4'S)-5-Methoxy-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ia)



73% Isolated yield, colorless oil, $[\alpha]_D^{25} = -13.27$ ($c = 1.06$ in CHCl₃); 90% ee, 12:1 dr, determined by HPLC analysis (Chiralpak IA-H column, hexane/i-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 210$ nm, 25 °C), t_R (major) = 23.49 min, t_R (minor) = 48.61 min; **¹H NMR** (400 MHz, CDCl₃) δ (ppm) 8.00 (d, $J = 8.3$ Hz, 2H, major+minor), 7.79 (dd, $J = 8.2, 1.1$ Hz, 1H, major+minor), 7.49 – 7.44 (m, 1H, major+minor), 7.34 – 7.26 (m, 3H, major+minor), 7.15 (dd, $J = 7.6, 1.5$ Hz, 1H, major+minor), 6.77 (dd, $J = 4.2, 1.5$ Hz, 2H, major), 6.70 (dd, $J = 4.2, 1.5$ Hz, 2H, minor), 6.44 (dd, $J = 8.4, 2.5$ Hz, 1H, major+minor), 6.14 (d, $J = 5.7$ Hz, 1H, major), 6.07 (d, $J = 8.4$ Hz, 1H, major+minor), 6.00 (d, $J = 5.7$ Hz, 1H, minor), 6.14 (d, $J = 5.7$ Hz, 1H, major+minor), 6.07 (d, $J = 8.4$ Hz, 1H, major+minor), 5.75 – 5.66 (m, 1H, major+minor), 5.24 – 5.20 (m, 2H, minor), 5.17 – 5.08 (m, 2H, major), 3.72 (s, 3H, major+minor), 3.51 (d, $J = 8.3$ Hz, 1H, major+minor), 2.44 (s, 3H, major+minor); **¹³C NMR** (100 MHz, CDCl₃) δ (ppm) 170.3, 160.0, 145.4, 145.1, 136.4, 136.0, 135.0, 134.6, 134.2, 133.6, 131.4, 129.4, 129.1, 128.0, 127.9, 126.7, 123.6, 123.6, 119.8, 111.1, 107.9, 65.1, 55.3, 49.3, 21.7. **HRMS** (ESI) for C₂₇H₂₃NO₄S [M+Na]⁺: calcd 480.1240, found 480.1249.

(1S,4'S)-5-Bromo-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ja)



72% Isolated yield, colorless oil, $[\alpha]_D^{25} = -3.98$ ($c = 0.83$ in CHCl₃); 91% ee, 10:1 dr, determined by HPLC analysis (Chiralpak IA-H column, hexane/i-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 220$ nm, 25 °C), t_R (major) = 15.53 min, t_R (minor) = 32.59 min; **¹H NMR** (400 MHz, CDCl₃) δ (ppm) 8.02 – 7.98 (m, 2H, major+minor), 7.79 (d, $J = 8.2$ Hz, 1H, major+minor), 7.47 (td, $J = 7.8, 1.6$ Hz, 1H, major+minor), 7.36 – 7.27 (m, 4H, major+minor), 7.15 (dd, $J = 7.7, 1.5$ Hz, 1H, major+minor), 7.08 (dd, $J = 8.1, 1.9$ Hz, 1H, major+minor), 6.75 (d, $J = 5.6$ Hz, 1H, major), 6.68 (d, $J = 5.6$ Hz,

1H, minor), 6.14 (d, $J = 8.1$ Hz, 1H, major+minor), 6.09 (d, $J = 5.6$ Hz, 1H, major), 6.00 (d, $J = 5.6$ Hz, 1H, minor), 5.57 – 5.66 (m, 1H, major+minor), 5.25 – 5.19 (m, 2H, minor), 5.18 – 5.09 (m, 2H, major), 3.60 (d, $J = 8.5$ Hz, 1H, major+minor), 2.45 (s, 3H, major+minor); ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 169.5, 145.9, 145.2, 142.8, 136.2, 136.0, 134.8, 133.8, 133.3, 131.0, 129.4, 129.1, 128.8, 128.0, 127.7, 126.8, 125.0, 124.4, 123.6, 122.4, 120.4, 65.8, 48.6, 21.7. HRMS (ESI) for $\text{C}_{26}\text{H}_{20}\text{BrNO}_3\text{S}$ [M+Na] $^+$: calcd 528.0239, found 528.0232.

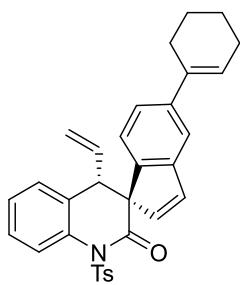
(1*S*,4'*S*)-5-Phenyl-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ka)

85% Isolated yield, colorless oil, $[\alpha]_D^{25} = -43.43$ ($c = 1.0$ in CHCl_3); 92% ee, 14:1 dr, determined by HPLC analysis (Chiralpak AD-H column, hexane/i-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 31.95 min, t_R (minor) = 51.13 min; ^1H NMR (400 MHz, CDCl_3) δ (ppm) 8.02 (d, $J = 8.3$ Hz, 2H, major+minor), 7.82 (d, $J = 8.1$ Hz, 1H, major+minor), 7.50 – 7.47 (m, 3H, major+minor), 7.42 (m, 2H, major+minor), 7.35 – 7.29 (m, 5H, major+minor), 7.18 – 7.14 (m, 2H, major+minor), 6.87 (d, $J = 5.6$ Hz, 1H, major), 6.69 (d, $J = 5.6$ Hz, 1H, minor), 6.28 (d, $J = 7.8$ Hz, 1H, major+minor), 6.15 (d, $J = 5.6$ Hz, 1H, major), 6.01 (d, $J = 5.6$ Hz, 1H, minor), 5.77 – 5.68 (m, 1H, major+minor), 5.27 – 5.22 (m, 2H, minor), 5.18 – 5.13 (m, 2H, major), 3.61 (d, $J = 8.3$ Hz, 1H, major+minor), 2.43 (s, 3H, major+minor); ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 170.1, 145.1, 144.5, 142.9, 141.6, 140.8, 136.3, 135.3, 135.0, 134.7, 133.9, 131.3, 129.6, 129.4, 129.1, 128.7, 127.9, 127.3, 127.1, 126.7, 125.1, 123.6, 123.3, 120.6, 120.0, 65.7, 49.0, 21.7. HRMS (ESI) for $\text{C}_{32}\text{H}_{25}\text{NO}_3\text{S}$ [M+Na] $^+$: calcd 526.1447, found 526.1457.

(1*S*,4'*S*)-5-(Thiophen-2-yl)-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3la)

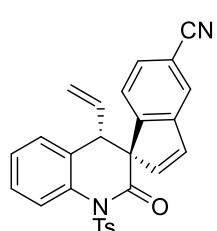
83% Isolated yield, colorless oil, $[\alpha]_D^{25} = -70.27$ ($c = 0.85$ in CHCl_3); 92% ee, 16:1 dr, determined by HPLC analysis (Chiralpak AD-H column, hexane/i-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 42.52 min, t_R (minor) = 70.79 min; ^1H NMR (400 MHz, CDCl_3) δ (ppm) 8.01 (d, $J = 8.4$ Hz, 2H, major+minor), 7.81 (dd, $J = 8.2, 1.1$ Hz, 1H, major+minor), 7.50 – 7.46 (m, 2H, major+minor), 7.34 – 7.28 (m, 3H, major+minor), 7.24 – 7.16 (m, 4H, major+minor), 7.03 (dd, $J = 5.0, 3.6$ Hz, 1H, major+minor), 6.84 (d, $J = 5.7$ Hz, 1H, major), 6.77 (d, $J = 5.7$ Hz, 1H, minor), 6.22 (d, $J = 8.0$ Hz, 1H, major+minor), 6.14 (d, $J = 5.7$ Hz, 1H, major), 6.02 (d, $J = 5.7$ Hz, 1H, minor), 5.74 – 5.65 (m, 1H, major+minor), 5.26 – 5.21 (m, 2H, minor), 5.18 – 5.11 (m, 2H, major), 3.59 (d, $J = 8.3$ Hz, 1H, major+minor), 2.44 (s, 3H, major+minor); ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 169.9, 145.2, 144.7, 144.0, 143.0, 136.3, 135.5, 134.9, 134.6, 134.5, 133.8, 133.8, 131.2, 129.4, 129.1, 127.9, 127.9, 126.7, 124.9, 123.9, 123.6, 123.4, 123.2, 120.1, 119.3, 65.8, 49.0, 21.7. HRMS (ESI) for $\text{C}_{30}\text{H}_{23}\text{NO}_3\text{S}_2$ [M+Na] $^+$: calcd 532.1012, found 532.1016.

(1*S*,4'*S*)-5-(Cyclohex-1-en-1-yl)-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ma)



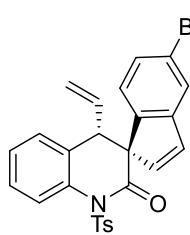
82% Isolated yield, colorless oil, $[\alpha]_D^{25} = -30.40$ ($c = 0.92$ in CHCl_3); 90% ee, 12:1 dr, determined by HPLC analysis (Chiralpak AD-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 22.14 min, t_R (minor) = 52.48 min; **1H NMR** (400 MHz, CDCl_3) δ (ppm) 8.00 (d, $J = 8.1$ Hz, 2H), 7.80 (d, $J = 8.1$ Hz, 1H), 7.49 – 7.45 (m, 1H), 7.35 – 7.27 (m, 3H), 7.23 (d, $J = 1.7$ Hz, 1H), 7.14 (dd, $J = 7.7, 1.5$ Hz, 1H), 6.92 (dd, $J = 8.0, 1.7$ Hz, 1H), 6.81 (d, $J = 5.6$ Hz, 1H), 6.12 – 6.01 (m, 3H), 5.76 – 5.67 (m, 1H), 5.16 – 5.09 (m, 2H), 3.52 (d, $J = 8.3$ Hz, 1H), 2.44 (s, 3H), 2.33 – 2.28 (m, 2H), 2.18 – 2.12 (m, 2H), 1.75 – 1.71 (m, 2H), 1.64 – 1.60 (m, 2H); **13C NMR** (100 MHz, CDCl_3) δ (ppm) 170.2, 145.1, 143.9, 143.1, 142.0, 136.3, 136.3, 135.0, 134.9, 134.2, 131.3, 129.4, 129.1, 128.9, 128.0, 127.9, 126.6, 125.1, 123.6, 122.8, 122.7, 119.8, 118.5, 65.6, 49.2, 27.4, 25.8, 23.0, 22.1, 21.7. **HRMS** (ESI) for $\text{C}_{32}\text{H}_{29}\text{NO}_3\text{S}$ [$\text{M}+\text{Na}$]⁺: calcd 530.1760, found 530.1766.

(1*S*,4'*S*)-2'-Oxo-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinoline]-5-carbonitrile (3na)



73% Isolated yield, colorless oil, $[\alpha]_D^{25} = -36.53$ ($c = 1.1$ in CHCl_3); 91% ee, 6:1 dr, determined by HPLC analysis (Chiralpak AD-H column, hexane/*i*-PrOH, 80:20 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 61.55 min, t_R (minor) = 128.33 min; **1H NMR** (400 MHz, CDCl_3) δ (ppm) 8.00 (m, 2H, major+minor), 7.80 (d, $J = 8.2$ Hz, 1H, major+minor), 7.50 (m, 1.8 Hz, 2H, major+minor), 7.37 – 7.28 (m, 4H, major+minor), 7.16 (dd, $J = 7.7, 1.5$ Hz, 1H, major+minor), 6.81 (d, $J = 5.7$ Hz, 1H, major), 6.74 (d, $J = 5.7$ Hz, 1H, minor), 6.61 (d, $J = 7.9$ Hz, 1H, minor), 6.49 (d, $J = 7.9$ Hz, 1H, major), 6.14 (d, $J = 5.7$ Hz, 1H, major), 6.07 (d, $J = 5.7$ Hz, 1H, minor), 5.57 – 5.49 (m, 1H, major+minor), 5.27 – 5.19 (m, 2H, minor), 5.18 – 5.09 (m, 2H, major), 3.71 (d, $J = 8.7$ Hz, 1H, major), 3.69 (d, $J = 8.7$ Hz, 1H, minor), 2.46 (s, 3H, major+minor); **13C NMR** (100 MHz, CDCl_3) δ (ppm) 168.9, 148.8, 145.4, 144.8, 136.4, 136.0, 134.6, 133.6, 132.4, 130.7, 130.2, 129.5, 129.0, 128.2, 127.4, 127.0, 124.9, 124.0, 123.6, 121.0, 118.7, 112.2, 66.8, 48.3, 21.7. **HRMS** (ESI) for $\text{C}_{27}\text{H}_{20}\text{N}_2\text{O}_3\text{S}$ [$\text{M}+\text{Na}$]⁺: calcd 475.1087, found 475.1089.

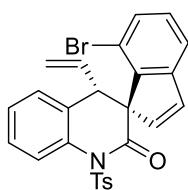
(1*S*,4'*S*)-5-(4,4,5,5-Tetramethyl-1,3,2-dioxaborolan-2-yl)-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3oa)



75% Isolated yield, colorless oil, $[\alpha]_D^{25} = -18.00$ ($c = 1.0$ in CHCl_3); 94% ee, 10:1 dr, determined by HPLC analysis (Chiralpak IA-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 0.5 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 24.65 min, t_R (minor) = 48.08 min; **1H NMR** (400 MHz, CDCl_3) δ (ppm) 8.00 (d, $J = 8.3$ Hz, 2H), 7.78 (d, $J = 8.2$ Hz, 1H), 7.67 (s, 1H), 7.46 (m, 2H), 7.33 (d, $J = 8.1$ Hz, 2H), 7.29 – 7.26 (m, 1H), 7.13 (d, $J = 7.7$ Hz, 1H), 6.81 (d, $J = 5.6$ Hz, 1H), 6.29 (d, $J = 7.6$ Hz, 1H),

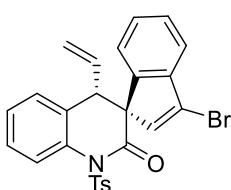
6.02 (d, $J = 5.6$ Hz, 1H), 5.66 – 5.57 (m, 1H), 5.14 – 5.06 (m, 2H), 3.61 (d, $J = 8.4$ Hz, 1H), 2.45 (s, 3H), 1.30 (s, 12H); **^{13}C NMR** (100 MHz, CDCl_3) δ (ppm) 170.0, 147.2, 145.1, 143.3, 136.3, 134.9, 134.9, 133.9, 133.5, 132.9, 131.3, 129.4, 129.1, 127.9, 127.9, 127.7, 126.7, 123.6, 122.6, 120.2, 83.8, 66.2, 48.7, 24.9, 24.8, 21.7. **HRMS** (ESI) for $\text{C}_{32}\text{H}_{32}\text{BNO}_5\text{S}$ [$\text{M}+\text{Na}]^+$: calcd 576.1986, found 576.1998.

(1*S*,4'*S*)-7-Bromo-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3pa)



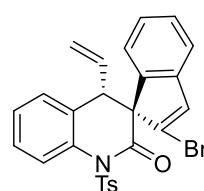
48% Isolated yield, colorless oil, $[\alpha]_D^{25} = -98.53$ ($c = 1.08$ in CHCl_3); 96% ee, 19:1 dr, determined by HPLC analysis (Chiralpak AD-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 11.87 min, t_R (minor) = 16.98 min; **^1H NMR** (400 MHz, CDCl_3) δ (ppm) 8.00 (d, $J = 8.4$ Hz, 2H), 7.79 (dd, $J = 8.1, 1.1$ Hz, 1H), 7.43 (td, $J = 7.7, 7.2, 1.3$ Hz, 1H), 7.35 – 7.28 (m, 3H), 7.24 – 7.15 (m, 3H), 7.12 – 7.06 (m, 1H), 6.70 (d, $J = 5.6$ Hz, 1H), 5.65 (d, $J = 5.6$ Hz, 1H), 5.28 – 5.12 (m, 2H), 5.04 (dd, $J = 9.6, 2.3$ Hz, 1H), 4.95 (d, $J = 8.8$ Hz, 1H), 2.46 (s, 3H); **^{13}C NMR** (100 MHz, CDCl_3) δ (ppm) 167.5, 147.0, 145.3, 142.7, 136.1, 135.0, 134.3, 132.1, 131.7, 130.3, 130.3, 130.0, 129.9, 129.3, 127.4, 126.6, 125.6, 123.7, 121.1, 120.7, 119.4, 68.8, 42.7, 21.7. **HRMS** (ESI) for $\text{C}_{26}\text{H}_{20}\text{BrNO}_3\text{S}$ [$\text{M}+\text{Na}]^+$: calcd 528.0239, found 528.0228.

(1*R*,4'*S*)-3-Bromo-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3qa)



53% Isolated yield, colorless oil, $[\alpha]_D^{25} = -6.17$ ($c = 0.90$ in CHCl_3); 93% ee, 9:1 dr, determined by HPLC analysis (Chiralpak IG-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 220$ nm, 25 °C), t_R (major) = 55.01 min, t_R (minor) = 98.06 min; **^1H NMR** (400 MHz, CDCl_3) δ (ppm) 7.99 (d, $J = 8.2$ Hz, 2H, major+minor), 7.79 (d, $J = 8.1$ Hz, 1H, major+minor), 7.51 – 7.46 (m, 1H, major+minor), 7.35 – 7.28 (m, 5H, major+minor), 7.16 (d, $J = 7.6$ Hz, 1H, major+minor), 7.05 (t, $J = 7.3$ Hz, 1H, major+minor), 6.31 (d, $J = 7.7$ Hz, 1H, major+minor), 6.18 (s, 1H, major), 6.04 (s, 1H, minor), 5.67 – 5.58 (m, 1H, major+minor), 5.28 – 5.20 (m, 2H, minor), 5.17 – 5.11 (m, 2H, major), 3.67 (d, $J = 8.5$ Hz, 1H, major+minor), 2.45 (s, 3H, major+minor); **^{13}C NMR** (100 MHz, CDCl_3) δ (ppm) 169.0, 145.3, 142.5, 142.2, 136.3, 134.8, 133.2, 132.6, 131.0, 129.5, 129.1, 128.7, 128.1, 127.8, 127.4, 126.9, 125.1, 123.7, 122.9, 121.2, 120.6, 65.9, 48.8, 21.7. **HRMS** (ESI) for $\text{C}_{26}\text{H}_{20}\text{BrNO}_3\text{S}$ [$\text{M}+\text{Na}]^+$: calcd 528.0239, found 528.0230.

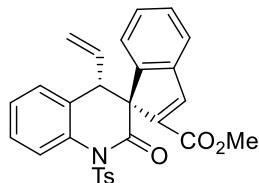
(1*R*,4'*S*)-2-Bromo-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ra)



58% Isolated yield, colorless oil, $[\alpha]_D^{25} = -79.50$ ($c = 0.94$ in CHCl_3); 87% ee, 4:1 dr, determined by HPLC analysis (Chiralpak AZ-H column, hexane/*i*-PrOH, 85:15 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 66.29 min, t_R (minor) = 52.24 min; **^1H NMR** (400 MHz, CDCl_3) δ (ppm) 8.04 – 8.00 (m, 2H), 7.78 (dd, $J = 8.3, 4.4$ Hz, 1H), 7.53 – 7.50 (m, 1H), 7.38 – 7.33 (m, 3H), 7.22 – 7.14 (m, 3H), 6.91 (t, $J = 2.7$ Hz,

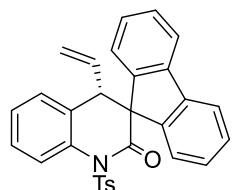
1H), 6.79 – 6.74 (m, 1H), 5.69 (d, J = 7.6 Hz, 1H), 5.45 – 5.38 (m, 1H), 5.27 – 5.22 (m, 1H), 4.95 – 4.85 (m, 1H), 4.29 (d, J = 9.4 Hz, 1H), 2.45 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 167.9, 145.3, 143.6, 140.1, 136.1, 135.7, 134.6, 130.2, 129.6, 129.5, 129.4, 129.3, 128.8, 128.1, 127.6, 126.8, 125.6, 123.5, 122.7, 121.9, 121.1, 68.0, 45.6, 21.7. HRMS (ESI) for $\text{C}_{26}\text{H}_{20}\text{BrNO}_3\text{S} [\text{M}+\text{Na}]^+$: calcd 528.0239, found 528.0227.

Methyl (1*S*,4*S*)-2'-oxo-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinoline]-2-carboxylate (3sa)



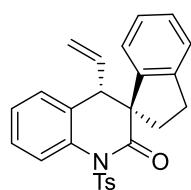
36%/37% Isolated yield, colorless oil, $[\alpha]_D^{25} = -1.73$ ($c = 0.47$ in CHCl_3); 78%/65% ee, 1:1 dr, determined by HPLC analysis (Chiralpak AD-H column, hexane/*i*-PrOH, 75:25 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 17.79 min, t_R (minor) = 22.26 min; ^1H NMR (400 MHz, CDCl_3) δ (ppm) 8.12 (d, J = 8.1 Hz, 2H), 7.70 – 7.66 (m, 2H), 7.43 – 7.36 (m, 4H), 7.25 – 7.21 (m, 2H), 7.09 (t, J = 7.8 Hz, 2H), 6.43 (d, J = 7.7 Hz, 1H), 5.87 – 5.77 (m, 1H), 4.96 – 4.85 (m, 2H), 3.75 (d, J = 9.6 Hz, 1H), 3.38 (s, 3H), 2.46 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 170.2, 163.8, 148.7, 145.1, 145.0, 140.7, 138.7, 136.7, 135.7, 133.7, 129.7, 129.6, 129.1, 128.9, 128.7, 127.9, 127.5, 126.1, 124.0, 122.5, 121.9, 118.4, 65.8, 51.5, 50.8, 21.7. HRMS (ESI) for $\text{C}_{28}\text{H}_{23}\text{NO}_5\text{S} [\text{M}+\text{Na}]^+$: calcd 508.1189, found 508.1198.

(*S*)-1'-Tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[fluorene-9,3'-quinolin]-2'-one (3ta)



74% Isolated yield, colorless oil, $[\alpha]_D^{25} = -55.87$ ($c = 1.05$ in CHCl_3); 79% ee, determined by HPLC analysis (Chiralpak AD-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 14.27 min, t_R (minor) = 30.80 min; ^1H NMR (400 MHz, CDCl_3) δ (ppm) 8.07 – 8.05 (m, 2H), 7.79 (dd, J = 8.1, 1.1 Hz, 1H), 7.64 – 7.60 (m, 2H), 7.53 (td, J = 7.8, 1.5 Hz, 1H), 7.39 – 7.27 (m, 5H), 7.18 (td, J = 7.6, 1.2 Hz, 2H), 6.90 (td, J = 7.7, 1.2 Hz, 1H), 6.74 (d, J = 7.6 Hz, 1H), 6.02 (d, J = 7.7 Hz, 1H), 5.10 – 4.94 (m, 3H), 4.19 (d, J = 8.5 Hz, 1H), 2.46 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 171.7, 145.4, 145.1, 142.1, 141.2, 141.0, 136.5, 135.2, 131.6, 130.9, 129.5, 129.0, 128.8, 128.4, 128.1, 128.0, 127.9, 127.1, 126.7, 124.6, 123.9, 123.3, 121.2, 120.2, 119.9, 64.0, 49.4, 21.7. HRMS (ESI) for $\text{C}_{30}\text{H}_{23}\text{NO}_3\text{S} [\text{M}+\text{Na}]^+$: calcd 500.1291, found 500.1292.

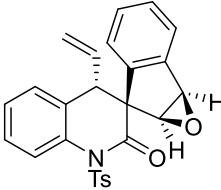
(1*S*,4*S*)-5-Phenyl-1'-tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (3ua)



79% Isolated yield, colorless oil, $[\alpha]_D^{25} = 150.67$ ($c = 1.05$ in CHCl_3); 93% ee, 12:1 dr, determined by HPLC analysis (Chiralpak IF-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 18.01 min, t_R (minor) = 19.98 min; ^1H NMR (400 MHz, CDCl_3) δ (ppm) 8.04 (d, J = 8.3 Hz, 2H, minor), 7.98 (d, J = 8.3 Hz, 2H, major), 7.76 (d, J = 8.2 Hz, 1H, major+minor), 7.45 – 7.41 (m, 1H, major+minor), 7.33 (d, J = 8.0 Hz, 2H, major+minor), 7.26 – 7.24 (m, 1H, major+minor), 7.17 – 7.08 (m, 3H, major+minor), 6.95 (t, J = 7.6 Hz, 1H, minor), 6.82 (t, J = 7.6 Hz, 1H, major), 5.94 (d, J

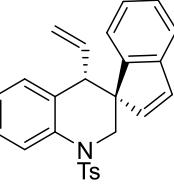
= 7.8 Hz, 1H, major+minor), 5.79 – 5.70 (m, 1H, major), 5.67 – 5.57 (m, 1H, minor), 5.24 – 5.21 (m, 2H, minor), 5.14 – 5.08 (m, 2H, major), 3.52 (d, J = 8.4 Hz, 1H, major+minor), 3.10 – 3.02 (m, 1H, major+minor), 2.92 – 2.82 (m, 1H, major+minor), 2.50 – 2.45 (m, 1H, major+minor), 2.44 (s, 3H, major+minor), 2.03 – 1.97 (m, 1H, major+minor); ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 173.3, 145.0, 144.6, 142.2, 136.5, 134.8, 134.4, 131.1, 129.3, 129.0, 128.5, 128.1, 127.7, 126.5, 126.4, 124.9, 123.9, 123.4, 119.3, 60.6, 51.6, 33.9, 30.8, 21.7. HRMS (ESI) for $\text{C}_{26}\text{H}_{23}\text{NO}_3\text{S}$ [M+Na] $^+$: calcd 452.1291, found 452.1298.

(1a*R*,4'*S*,6*S*,6a*S*)-1'-Tosyl-4'-vinyl-1a,1',4',6a-tetrahydro-2'H-spiro[indeno[1,2-b]oxirene-6,3'-quinolin]-2'-one (4a)



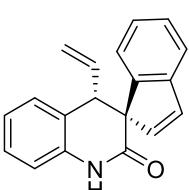
84% Isolated yield, colorless oil, $[\alpha]_D^{25} = 175.60$ ($c = 0.95$ in CHCl_3); 94% ee, 14:1 dr, determined by HPLC analysis (Chiralpak AD-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 30.72 min, t_R (minor) = 20.91 min; ^1H NMR (400 MHz, CDCl_3) δ (ppm) 7.99 – 7.97 (m, 2H), 7.84 (d, J = 8.4 Hz, 1H), 7.50 (t, J = 8.0 Hz, 1H), 7.43 (d, J = 7.4 Hz, 1H), 7.34 – 7.29 (m, 3H), 7.21 – 7.12 (m, 2H), 6.87 (t, J = 7.7 Hz, 1H), 6.12 – 6.02 (m, 1H), 5.57 (d, J = 7.9 Hz, 1H), 5.27 – 5.21 (m, 2H), 4.31 (t, J = 2.7 Hz, 1H), 4.20 (t, J = 2.6 Hz, 1H), 3.63 (d, J = 7.2 Hz, 1H), 2.45 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 170.6, 145.3, 143.1, 141.1, 136.1, 135.1, 134.7, 129.9, 129.4, 129.3, 129.3, 129.0, 128.3, 128.1, 126.9, 125.8, 124.9, 123.6, 119.3, 61.6, 59.6, 58.7, 48.2, 21.7. HRMS (ESI) for $\text{C}_{26}\text{H}_{21}\text{NO}_4\text{S}$ [M+Na] $^+$: calcd 466.1083, found 466.1081.

(1*S*,4'*S*)-4'-Vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (4b)



84% Isolated yield, colorless oil, $[\alpha]_D^{25} = 64.47$ ($c = 0.79$ in CHCl_3); 94% ee, 14:1 dr, determined by HPLC analysis (Chiralpak IC-H column, hexane/*i*-PrOH, 90:10 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 29.33 min, t_R (minor) = 19.39 min; ^1H NMR (400 MHz, CDCl_3) δ (ppm) 7.68 – 7.61 (m, 3H), 7.56 (d, J = 7.2 Hz, 1H), 7.34 – 7.28 (m, 3H), 7.25 – 7.17 (m, 5H), 6.88 (d, J = 5.7 Hz, 1H), 6.14 (d, J = 5.7 Hz, 1H), 5.21 – 5.12 (m, 1H), 4.39 (d, J = 10.2 Hz, 1H), 4.06 (d, J = 7.9 Hz, 1H), 3.79 (d, J = 17.0 Hz, 1H), 3.52 (d, J = 11.0 Hz, 1H), 2.90 (d, J = 11.0 Hz, 1H), 2.31 (s, 3H); ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 148.2, 143.8, 143.2, 137.6, 136.1, 135.7, 135.4, 135.3, 133.8, 129.8, 129.7, 127.4, 127.4, 126.9, 126.4, 126.0, 125.5, 123.7, 121.7, 116.9, 65.0, 62.1, 43.5, 21.3. HRMS (ESI) for $\text{C}_{26}\text{H}_{23}\text{NO}_2\text{S}$ [M+Na] $^+$: calcd 436.1342, found 436.1348.

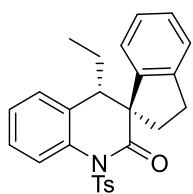
(1*S*,4'*R*)-1'-Tosyl-4'-vinyl-1',4'-dihydro-2'H-spiro[indene-1,3'-quinoline (4c)



84% Isolated yield, colorless oil, $[\alpha]_D^{25} = 117.67$ ($c = 0.86$ in CHCl_3); 94% ee, 14:1 dr, determined by HPLC analysis (Chiralpak AD-H column, hexane/*i*-PrOH, 92:8 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 38.83 min, t_R (minor) = 36.15 min; ^1H NMR (400 MHz, CDCl_3) δ (ppm) 8.55 (s, 1H), 7.31 (d, J = 7.4 Hz, 1H), 7.28 – 7.26 (m, 1H), 7.25 – 7.22 (m, 1H), 7.10 (d, J = 7.4 Hz, 1H), 7.06 – 7.02 (m,

2H), 6.93 – 6.89 (m, 2H), 6.84 (d, J = 7.8 Hz, 1H), 6.46 (d, J = 5.6 Hz, 1H), 5.89 – 5.80 (m, 1H), 5.08 (dd, J = 10.1, 1.4 Hz, 1H), 5.01 (d, J = 17.0 Hz, 1H), 3.65 (d, J = 8.8 Hz, 1H); ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 170.4, 145.1, 143.8, 136.4, 135.7, 135.4, 134.2, 128.3, 128.1, 125.9, 123.6, 123.0, 121.8, 118.4, 115.3, 62.6, 49.9. HRMS (ESI) for $\text{C}_{19}\text{H}_{15}\text{NO} [\text{M}+\text{Na}]^+$: calcd 296.1046, found 296.1045.

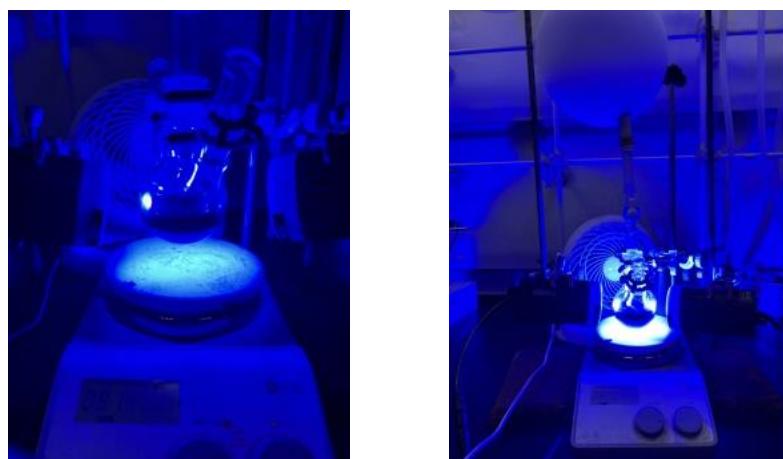
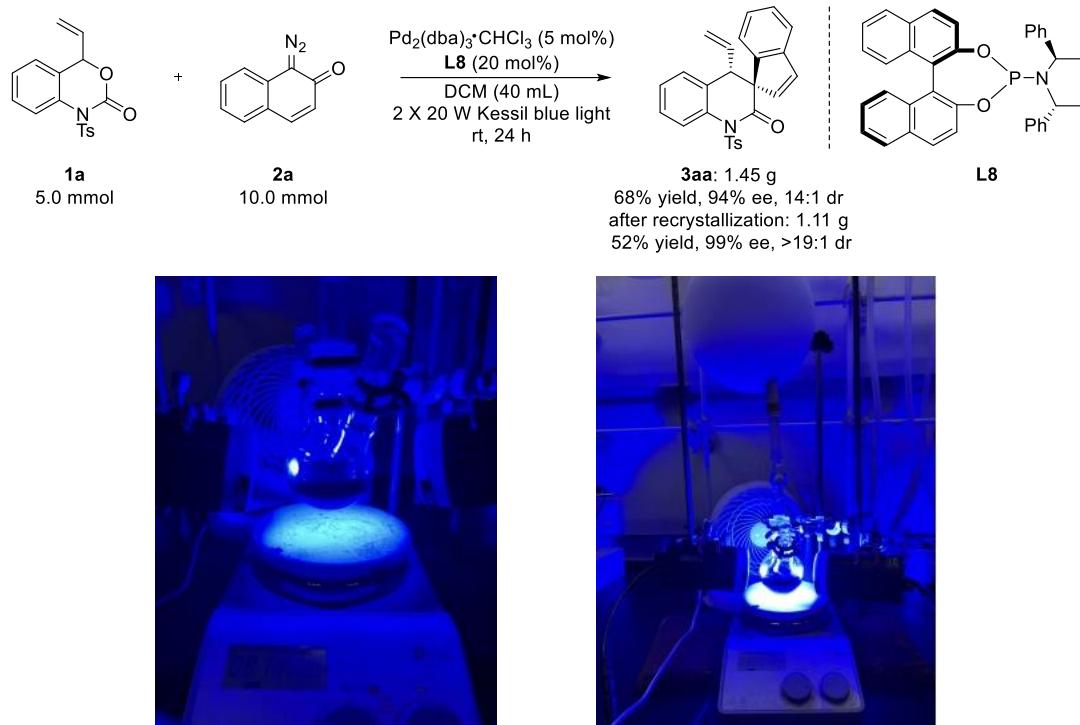
(1*S*,4'*S*)-4'-Ethyl-1'-tosyl-1',2,3,4'-tetrahydro-2'H-spiro[indene-1,3'-quinolin]-2'-one (4d)



84% Isolated yield, colorless oil, $[\alpha]_D^{25} = 73.60$ ($c = 0.75$ in CHCl_3); 94% ee, 14:1 dr, determined by HPLC analysis (Chiralpak IC-H column, hexane/*i*-PrOH, 85:15 v/v, flow rate 1.0 mL/min, $\lambda = 254$ nm, 25 °C), t_R (major) = 27.87 min, t_R (minor) = 34.22 min; ^1H NMR (400 MHz, CDCl_3) δ (ppm) 7.96 – 7.92 (m, 2H), 7.75 (dd, J = 8.2, 1.1 Hz, 1H), 7.46 – 7.41 (m, 1H), 7.33 – 7.26 (m, 3H), 7.13 (dd, J = 7.5, 1.8 Hz, 2H), 7.06 (td, J = 7.4, 1.1 Hz, 1H), 6.73 (t, J = 7.6 Hz, 1H), 5.60 (d, J = 7.7 Hz, 1H), 3.15 – 3.11 (m, 1H), 2.86 – 2.79 (m, 1H), 2.65 – 2.59 (m, 1H), 2.43 (s, 3H), 1.98 – 1.90 (m, 1H), 1.74 – 1.64 (m, 1H), 1.28 – 1.16 (m, 1H), 0.90 (t, J = 7.4 Hz, 3H); ^{13}C NMR (100 MHz, CDCl_3) δ (ppm) 173.4, 144.9, 144.9, 142.6, 136.4, 134.9, 132.1, 130.0, 129.3, 129.0, 128.0, 127.5, 126.3, 126.0, 125.0, 123.9, 123.8, 61.2, 49.4, 35.1, 31.0, 23.2, 21.7, 12.2. HRMS (ESI) for $\text{C}_{26}\text{H}_{25}\text{NO}_3\text{S} [\text{M}+\text{Na}]^+$: calcd 454.1447, found 454.1442.

6. Demonstration of the Synthetic Utility

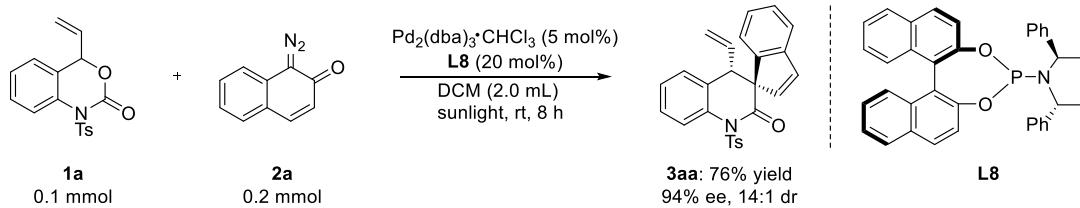
6.1 A Gram-Scale Reaction



Supplementary Figure 4 Set up of gram-scale reaction

Procedure I: Under argon atmosphere, a flame-dried 100 mL three-necked flask was charged with Pd₂(dba)₃·CHCl₃ (0.25 mmol, 5 mol%), L8 (1.0 mmol, 20 mol%) and anhydrous DCM (20 mL) and the resulting solution was stirred for 30 min at room temperature. Then, vinyl benzoxazinanone **1a** (5.0 mmol, 1.0 equiv.), 1-diazonaphthalen-2(1H)-one **2a** (10.0 mmol, 2.0 equiv.) and DCM (20 mL) were added to the reaction mixture. After that, the reaction solution was stirred under the irradiation of 2×20 W Kessil blue LEDs for 24 h at rt. The combined solution was concentrated under vacuum and the residue was purified by column chromatography to afford the desired product **3aa** in 68% yield, 94% ee and 14:1 dr. Then, it can be obtained by recrystallization with 52% yield, 99% ee and >19:1 dr.

6.2 A Sunlight-Driven Reaction

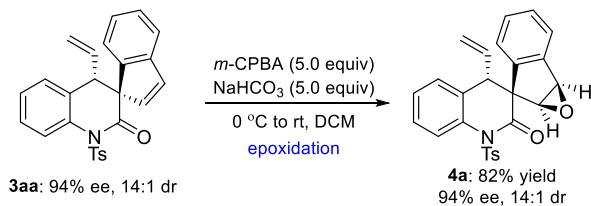




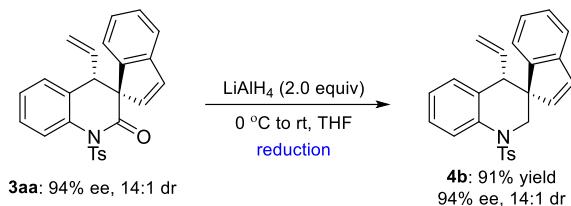
Supplementary Figure 5 Set up of sunlight-driven reaction

Procedure II: Under argon atmosphere, a flame-dried 10 mL Schlenk tube was charged with Pd₂(dba)₃·CHCl₃ (0.005 mmol, 5 mol%), **L8** (0.02 mmol, 20 mol%) and anhydrous DCM (1.0 mL) and the resulting solution was stirred for 30 min at room temperature. Then, vinyl benzoxazinanone **1a** (0.1 mmol, 1.0 equiv.), 1-diazonaphthalen-2(1H)-one **2a** (0.2 mmol, 2.0 equiv.) and DCM (1.0 mL) were added to the reaction mixture. After that, the reaction solution was stirred under sunlight from 8:00 am to 4:00 pm. Upon the completion of reaction as monitored by TLC, the solvent was removed by vacuum and the crude reaction mixture was purified by flash chromatography on silica gel (petrol ether/ethyl acetate = 20/1 to 15/1) to afford the product **3aa** in 76% yield, 94% ee and 14:1 dr.

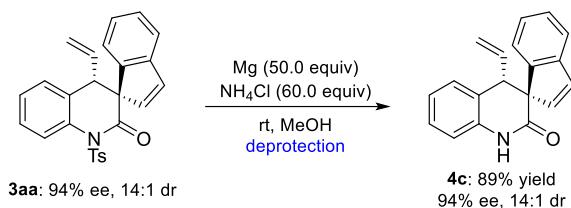
6.3 Synthetic Transformation



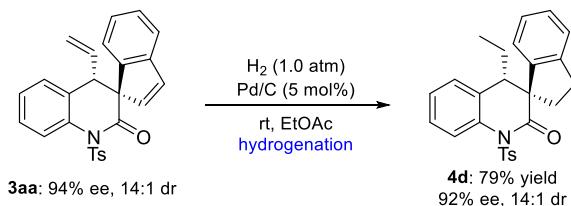
Procedure III: Under argon atmosphere, a flame-dried 10 mL Schlenk tube was charged with compound **3aa** (42.7 mg, 0.10 mmol), NaHCO₃ (42.0 mg, 0.50 mmol) and anhydrous DCM (2.0 mL) and cooled to 0 °C. To this solution, *m*-CPBA (86.3 mg, 0.50 mmol) was added dropwise, and the reaction mixture was stirred until the consumption of **3aa** as monitored by TLC. The reaction was quenched with saturated NH₄Cl aqueous solution and extracted with ethyl acetate. The combined organic layer was dried with Na₂SO₄, and evaporated under reduced pressure. The residue was purified by column chromatography to afford the desired product **4a** in 82% yield, 94% ee and 14:1 dr.



Procedure IV: Under argon atmosphere, a flame-dried 10 mL Schlenk tube was charged with compound **3aa** (42.7 mg, 0.10 mmol) and anhydrous THF (2.0 mL) and cooled to 0 °C. To this solution, LiAlH₄ (2.0 equiv., 1.0 M in THF) was added dropwise, and the reaction mixture was stirred at rt until the consumption of **3aa** as monitored by TLC. The reaction was quenched with saturated NH₄Cl aqueous solution and extracted with ethyl acetate. The combined organic layer was dried with Na₂SO₄, and evaporated under reduced pressure. The residue was purified by column chromatography to afford the desired product **4b** in 91% yield, 94% ee and 14:1 dr.

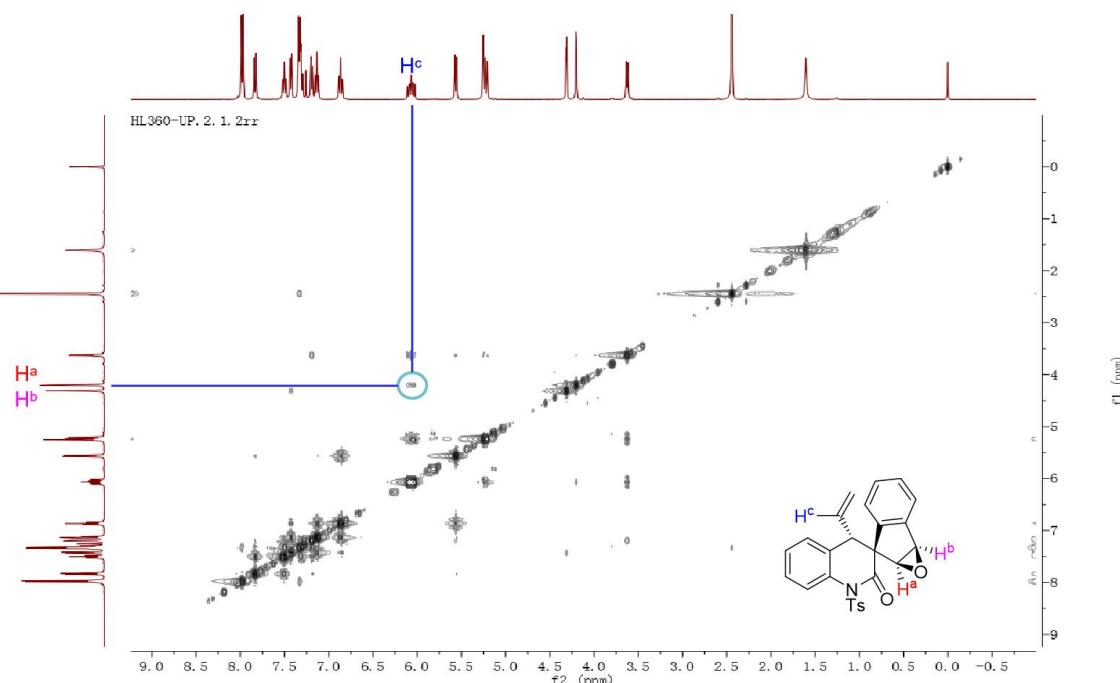


Procedure V: Under argon atmosphere, a flame-dried 10 mL Schlenk tube was charged with compound **3aa** (42.7 mg, 0.10 mmol) and Mg (120 mg, 5.0 mmol, 200-300 mesh) and NH₄Cl (321mg, 6.0 mmol) and anhydrous MeOH (4.0 mL). The resulting solution was in MW for 4 h. Then NH₄Cl (3.0 mL) was added to the reaction mixture to quench excess magnesium powder. The aqueous phase was extracted with ethyl acetate (4×5.0 mL). The combined organic layers were dried over Na₂SO₄, filtered and concentrated in vacuo. The residue was purified by column chromatography afford the desired product **4c** in 89% yield, 94% ee and 14:1 dr.



Procedure VI: In a flame-dried 10 ml Schlenk tube **3aa** (42.7 mg, 0.10 mmol), Pd/C (5.3 mg, 5 mol%, wt = 10%) were dissolved in EtOAc (2.0 mL). The reaction mixture was stirred at room temperature for about 4 h after pumped by H₂ balloon. Then the reaction was filtered by the diatomite and washed by ethyl acetate. The product was purified by flash column chromatography on silica gel (petrol ether/ethyl acetate = 20/1 to 10/1) to afford the desired product **4d** in 79% yield, 92% ee and 14:1 dr.

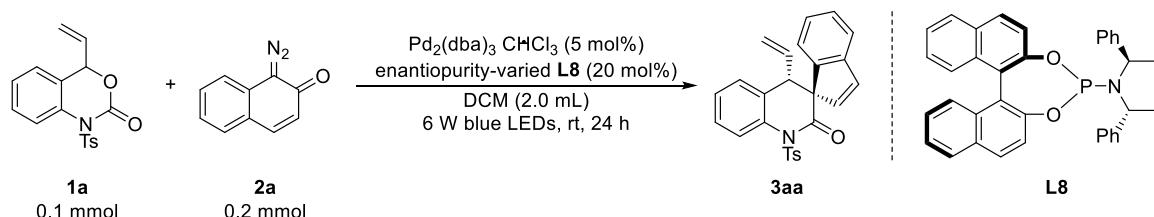
6.4 Cope of the NOESY Spectroscopy of Product 4a



Supplementary Figure 6 NOESY spectroscopy of 4a

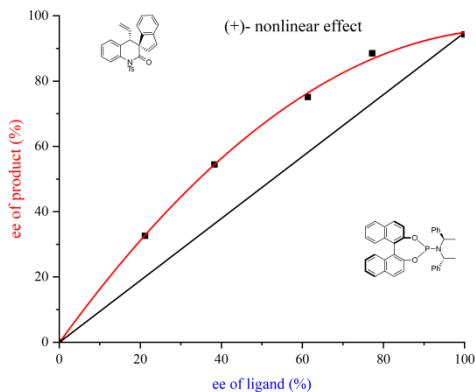
7. Mechanistic Investigations

7.1 Non-Linear Relationship



We carried out non-linear effect study with chiral phosphoramidite ligand **L8** under the optimized condition. Six different reactions were performed with chiral ligand **L8** with different levels of enantiopurity using the following procedure. Under argon atmosphere, a flame-dried 10 ml Schlenk tube was charged with $\text{Pd}_2(\text{dba})_3 \cdot \text{CHCl}_3$ (0.005 mmol, 5 mol%), chiral phosphoramidite ligand **L8** (0.02 mmol, 20 mol%, x% ee) with desired level of enantiopurity (prepared by mixing two enantiomers of the ligand **L8** in appropriate amount) and DCM (1.0 mL). The resulting solution was stirred for 30 mins at room temperature. Then vinylbenzoxazinanone **1a** (0.1 mmol, 1.0 equiv.) and 1-diazonaphthalen-2(1H)-one **2a** (0.2 mmol, 2.0 equiv.) in DCM (1.0 ml) were added. The resulting solution was stirred under the irradiation of 6W blue LEDs for 24 h at rt. After the reaction was over, the reaction mixture was purified by flash silica gel chromatography (petroleum/EtOAc = 10:1) to give the lactone product **3aa**. The pure product was dissolved in *i*-PrOH and small aliquot was injected onto a chiral HPLC for ee analysis. The enantioselectivity of the obtained product were plotted against the enantiopurity of the chiral ligand **L8** (Supplementary Figure 7).

ee of ligand (%)	0	21.17	38.29	61.34	77.22	99.99
ee of product (%)	0	32.60	54.45	75.13	88.52	94.32



Supplementary Figure 7 Relationship between ee of ligand (**L8**) and product (**3aa**)

7.2 A Preliminary Explanation for Selectivity of the Reaction via DFT Calculations

(1) Computational details

All of the calculations were performed using the Gaussian 16 program.¹ Structures were optimized at the B3LYP level of density functional theory² with Grimme's D3(BJ) dispersion correction³ in gas phase. For optimizations, Ahlrichs's def2SVP basis set was used for all atoms.⁴ Frequency calculations have been performed to verify the optimized structures as local minima or transition state and to obtain Gibbs free energy at 298 K. To reduce error caused by the breakdown of the harmonic oscillator approximation, Truhlar's quasi-harmonic correction was used to compute molecular entropies by setting all positive frequencies that are less than 100 cm⁻¹ to 100 cm⁻¹.⁵ Intrinsic reaction coordinate (IRC) calculations were carried out to make sure that every transition state links relevant intermediates.⁶ The electronic energies were further refined by carrying out single-point energy calculations using B3LYP functional with Grimme's D3(BJ) dispersion correction. The def2TZVP basis set was applied for all atoms.⁴ The SMD solvation model with DCM as the solvent was employed to account for solvation effect.⁷ The three-dimensional (3D) structures were depicted using CYLview software.

(2) References

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(3) B3LYP geometries for all the optimized compounds and transition states

3aa			6	2.62223300	-1.42021900	-0.01822100
E = -1682.65590180	G = -1682.300116		6	2.95860600	-1.99878700	1.19658300
6 -0.00782500	1.96760700	-0.14784600	1	2.55299500	-1.61281600	2.13441800
6 -0.88369400	2.91032800	-0.69484800	6	3.82353600	-3.10281800	1.19708100
1 -1.94605300	2.68850200	-0.76405900	1	4.10066000	-3.57196700	2.14394400
6 -0.38992600	4.13769500	-1.13873300	6	4.33152000	-3.61291100	-0.00399400
1 -1.08006500	4.87587600	-1.55297500	1	5.00315100	-4.47459000	0.01746000
6 0.97592700	4.41592000	-1.05951500	6	3.98396200	-3.03557500	-1.23127100
1 1.36300200	5.37722600	-1.40444100	1	4.37518200	-3.44009800	-2.16795200
6 1.85225300	3.45291300	-0.55265600	6	3.12388000	-1.93514200	-1.23103500
1 2.92376400	3.65393900	-0.50627400	6	2.57893300	-1.12702200	-2.32823500
6 1.37437000	2.22300300	-0.09443400	1	2.78754000	-1.30216700	-3.38506500
6 2.26798400	1.09246700	0.37599000	6	1.79550100	-0.14735000	-1.83353800
1 2.12356700	0.96126200	1.46383200	1	1.26028100	0.60975100	-2.40524100
6 1.74763400	-0.21812400	-0.30873100	6	-3.08074300	-0.28482000	0.55824100
6 0.30376100	-0.46184900	0.16902700	6	-3.10780000	-1.68173100	0.59834200
6 3.72819900	1.31343600	0.10725600	1	-2.33487800	-2.22093700	1.14304100
1 4.02030900	1.31490000	-0.94886000	6	-4.12119200	-2.34681700	-0.08624600
6 4.65231300	1.49280500	1.05212700	1	-4.15077100	-3.43931200	-0.06410000
1 4.39065900	1.48986800	2.11513700	6	-5.10554200	-1.64363300	-0.80294100
1 5.70549400	1.64088200	0.79970200	6	-5.05724900	-0.24150200	-0.80762300

1	-5.82375100	0.32248800	-1.34468500	6	-6.10434100	-2.76639400	-0.63011500
6	-4.05304200	0.44648500	-0.12593100	1	-6.49953300	-3.46403500	0.12746400
1	-4.03240000	1.53665600	-0.10253900	1	-5.90354100	-3.35697500	-1.53790000
6	-6.18174000	-2.38957500	-1.54699000	1	-6.89503400	-2.03833100	-0.86037300
1	-5.75143400	-2.97095800	-2.37956200	7	-0.23264600	0.31893400	-0.13777600
1	-6.93461900	-1.70675200	-1.96564700	8	-0.19375800	-1.06583200	2.04832100
1	-6.69670100	-3.10608300	-0.88714500	8	-1.41551300	1.17745700	2.04546100
7	-0.46643500	0.69870000	0.32055900	16	-1.04351300	-0.08151700	1.38950200
8	-0.13410400	-1.55831800	0.42572700	6	1.79943400	-0.98203800	-0.38165800
8	-1.37369400	-0.18174400	2.59206500	6	3.23434600	-1.28510700	-0.30003300
8	-2.26472800	1.98700500	1.62138200	6	4.34465700	-0.51402000	0.04391100
16	-1.82511400	0.60495000	1.45328400	1	4.22585100	0.53581300	0.31240400
				6	5.61087200	-1.11562000	0.03653200
				1	6.48973700	-0.52378200	0.30417800
3b				6	5.76452400	-2.46444800	-0.31016100
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6	-1.80136400	1.53545600	-1.60717000	1	4.77899700	-4.29876400	-0.92282800
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6	-2.20863500	2.74876600	-2.16919200	6	2.06371700	-3.19855100	-0.95772500
1	-3.07147500	2.77332400	-2.83851200	1	1.86934800	-4.22373400	-1.27455700
6	-1.50521600	3.92464900	-1.89051700	6	1.13357500	-2.21566300	-0.79865500
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6	0.01269100	2.69660800	-0.46761000				
6	1.17128700	2.52522900	0.49349400	3c			
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1	2.73932200	3.73793700	-0.46112800	1	1.26789200	-1.26146200	1.05802600
6	2.36943600	4.47508700	1.49003800	6	-0.23899200	-1.32596400	2.60199400
1	1.80926700	4.37577500	2.42522400	1	0.29643800	-2.05741500	3.21097600
1	3.09544800	5.28973300	1.42771500	6	-1.48148500	-0.83380300	2.99726300
6	-2.53074000	-0.86272700	0.81166900	1	-1.94154500	-1.17051800	3.92896300
6	-3.69565200	-0.10658600	0.68406600	6	-2.14823200	0.08872500	2.19396700
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6	-3.66450400	-2.82606300	0.02383000	6	-3.52308100	2.26876600	0.97028900
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6	3.70256100	-0.18089000	-1.14395200	6	-3.33895300	-2.72514000	0.11169500
1	3.28190500	-0.21678900	-2.14918800	1	-4.08995300	-3.07822000	0.82132100
6	4.82856800	-0.92195900	-0.79631600	6	-2.24673200	-1.99824000	0.58688300
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6	4.71096200	-0.09150500	1.46863100	1	3.40884600	-0.99322300	1.85277500
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8	0.90131000	-0.71871100	-2.60015400	1	6.12383900	2.51873500	-0.25047700
8	1.74977000	2.05409400	-1.92821200	1	6.11588100	2.03875900	-1.96634100
8	1.29325400	2.39521300	0.55939300	1	7.04414000	1.09761800	-0.77284800
16	1.62717900	1.53110400	-0.57237400	7	-0.11907300	-0.80894700	0.12644700
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6	-4.50892600	-1.22288000	-0.10079400	8	1.41450800	-2.57553800	1.41888100
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6	-4.41259000	-2.59460500	0.18410600	16	1.31343100	-1.92083800	0.11792300
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6	-3.28489100	-3.32724400	-0.20302100	6	-2.77553000	1.23425800	-0.72564000
1	-3.22592000	-4.39164900	0.03642300	1	-3.07570700	0.20169000	-0.57774800
6	-2.23340600	-2.71640900	-0.89933600	6	-3.35696000	1.95632600	-1.78027800
1	-1.35425800	-3.29007100	-1.20109800	1	-4.09257800	1.46212300	-2.41891500
6	-2.34045000	-1.36041800	-1.20432900	6	-3.00753500	3.28699900	-2.01946900
6	0.06328200	-0.35683500	-1.81793800	1	-3.47172900	3.83283000	-2.84446600
6	-3.20046000	0.83775400	-1.10063300	6	-2.06759500	3.92398500	-1.20130800
1	-4.07141400	1.44399600	-1.38310000	1	-1.79228100	4.96728200	-1.37410300
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6	-2.04170000	0.77080800	-2.04314900	6	-0.49723600	3.63011200	0.83643800
1	-1.53337600	1.66592400	-2.40475500	1	-0.06542700	4.63105700	0.88358500
				6	-0.21226300	2.60560800	1.65794800
3d				1	0.49883100	2.59393600	2.48078200
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6	-1.29156900	-1.50327700	-0.34157300	6	-1.02807000	1.36634700	1.30509900
6	-1.43978000	-1.75713200	-1.70608700	6	-2.08189300	-1.40667200	1.93654800
1	-0.70012300	-1.36396800	-2.40157000	1	-1.15566900	-1.63791300	2.47426800
6	-2.53273200	-2.49554300	-2.16366600	6	-2.68871500	-0.22080000	2.11386100
1	-2.64537700	-2.68864100	-3.23243000	1	-3.56688000	-0.00068300	1.50005000

6	-1.91675500	0.97479300	2.56623900	6	-6.70503800	-0.12236500	-3.46477800
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1	-2.53904600	1.84028600	2.83349400	6	-4.97823700	-1.38935300	-2.29482300
				6	-4.32655700	-1.62067800	-3.55377500
				6	-4.91542700	-1.11832900	-4.74664800
INT1				6	-6.08223600	-0.38746700	-4.70823300
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6	6.47527500	1.15921100	-2.57425800	6	-4.37262800	-1.88009500	-1.08579800
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1	7.21197700	1.08131000	-3.37765200	1	-2.59210000	-2.47833600	-4.54252700
1	6.36531600	-0.96148100	-2.29650800	1	-1.51159400	-3.21599200	-2.41072200
6	4.48211300	-1.08573000	-0.30393100	6	-4.97312600	-1.64534300	0.25299700
6	3.55250000	1.48607600	0.47288800	6	-6.29784600	-2.08153600	0.59933200
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6	2.99798100	0.35716600	1.03381600	6	-6.84557900	-1.72169200	1.87689200
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1	2.20635100	0.43247100	1.78080300	1	-6.66946500	-3.20290100	-1.22405200
6	4.90219500	-2.46603500	-0.66911200	6	-8.16103300	-2.14397500	2.21257100
6	6.24907100	-2.93136800	-0.48843400	6	-6.05660000	-0.96355000	2.78630800
6	3.96332200	-3.35814600	-1.17015100	1	-4.13342200	-0.04365100	3.15799800
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6	6.59987800	-4.26085700	-0.90291800	1	-8.93603300	-3.92190200	-0.58358100
6	4.29973400	-4.67026200	-1.57324000	1	-8.56916500	-1.85405400	3.18413300
6	8.53914900	-2.59951300	0.28060500	1	-6.48797700	-0.67570300	3.74795800
1	6.99272100	-1.12828800	0.46689100	1	-9.91292300	-3.23118100	1.61340200
6	7.93785800	-4.71150600	-0.73235600	8	-2.47993300	-2.88518400	-0.02313200
6	5.59911300	-5.10543100	-1.45679000	8	-2.92637300	-0.61990400	0.95969700
1	3.51222200	-5.30741100	-1.97932600	15	-1.65083500	-1.68137700	0.83378900
6	8.89097800	-3.89960500	-0.15771300	7	-1.42262200	-2.52950300	2.23799000
1	9.29246600	-1.96567200	0.75402100	6	-0.21509600	-2.35288900	3.03803200
1	8.19402100	-5.72216400	-1.06057800	1	0.30285300	-3.31877200	3.15150300
1	5.87467800	-6.11340000	-1.77610100	1	0.46620200	-1.63628700	2.56739800
1	9.91482100	-4.25823500	-0.02946500	1	-0.46184600	-1.95217600	4.03112500
8	2.77962100	-2.04069500	1.08026900	6	-2.47173200	-3.32308800	2.86558400
8	2.64342800	-2.96095700	-1.30818700	1	-2.06016500	-4.30117000	3.16850300
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1	-0.53911900	-4.24229500	-0.13675500	6	0.82966200	1.98419700	3.38669800
1	0.44087500	-5.73740800	-0.21470300	6	2.03866500	2.47072200	3.91625100
6	2.37107300	-4.84822500	1.52303700	1	2.59500400	3.23168700	3.36996400
1	1.92281500	-5.24731200	2.44877900	6	2.49788700	1.97680000	5.13719100
1	2.76204800	-5.69388600	0.93039500	1	3.43713900	2.35457200	5.55095500
1	3.20334700	-4.19083900	1.79620800	6	1.76924800	1.00416400	5.85211600
6	-0.81459100	0.76381100	-1.71092900	1	2.15029900	0.63785600	6.80951400
1	-1.28835000	0.05832700	-2.40427900	6	0.56631000	0.51311400	5.34924600
6	0.59264500	0.79683100	-1.72700800	1	-0.00306900	-0.23451400	5.91159000
1	1.13779700	1.56589300	-1.18568600	6	0.08584900	0.99066400	4.11699700
6	1.31251400	-0.31929300	-2.19374000	6	-1.08097900	0.66453900	3.34090700
1	0.89274800	-0.97138200	-2.96773300	1	-1.87161800	-0.02281000	3.63990100
1	2.39917300	-0.32722200	-2.10379500	6	-1.05284600	1.40668800	2.16926900
6	-1.75449000	1.73106700	-1.14321000	1	-1.83613900	1.37545000	1.42595100
6	-1.411157400	2.86951200	-0.37277700				
6	-3.12219000	1.47215400	-1.35807900				
6	-2.41563700	3.67900700	0.16734100	TS1arr			
6	-4.11107200	2.27587400	-0.81322100	E =	-4603.44170859	G = -4602.425559	
1	-3.40136000	0.60472600	-1.95475500	6	2.14670900	2.65237100	0.76461000
6	-3.76327800	3.38423200	-0.03712000	6	3.44983200	3.12364800	0.64238400
1	-2.11348900	4.53948000	0.76601000	1	3.63679500	4.02722700	0.06334500
1	-5.16602100	2.03325500	-0.99469800	6	4.49366300	2.42831800	1.26138800
1	-4.53386400	4.02197400	0.40163700	1	5.52025700	2.78704300	1.16163200
7	-0.05714400	3.22410900	-0.12594600	6	4.21904300	1.28350900	2.01248100
6	0.57767500	5.85361800	-0.84700200	1	5.02988700	0.74544700	2.50361100
6	-0.55433200	6.57306800	-1.23567400	6	2.91078900	0.81033400	2.12533700
1	-1.31188300	6.09337100	-1.85610600	1	2.71637400	-0.10942700	2.67779700
6	-0.67978900	7.90289000	-0.83024800	6	1.85772400	1.47816100	1.49179400
1	-1.56230400	8.47402300	-1.12966800	6	-0.01333900	4.53932700	-2.10645100
6	0.31246500	8.51972800	-0.05447200	6	-1.28642200	4.40010600	-2.65722900
6	1.44524700	7.76895100	0.30679600	1	-1.76850200	3.42301800	-2.66653500
1	2.22870100	8.23636200	0.90931800	6	-1.91737800	5.52752900	-3.17543800
6	1.58725500	6.43886600	-0.07825200	1	-2.91505300	5.42944200	-3.61104100
1	2.44580300	5.84253800	0.22265200	6	-1.29999500	6.78960000	-3.13773200
6	0.17680300	9.95212100	0.39238600	6	-0.01552300	6.89073500	-2.58086400
1	1.05308900	10.54793200	0.08922600	1	0.48326000	7.86260400	-2.55143200
1	0.10918400	10.01659100	1.49129100	6	0.63936500	5.77031900	-2.07021700
1	-0.72095500	10.42614200	-0.02980200	1	1.64258200	5.84985200	-1.64947000
8	-0.17813300	3.98042800	-2.52726000	6	-2.01650500	8.00685800	-3.66070700
8	2.10885400	3.76160900	-1.47291300	1	-2.74821000	8.37461300	-2.92078200
16	0.70341700	4.15514600	-1.36714500	1	-1.31789000	8.82938800	-3.87200200
8	1.60381200	3.87503600	1.29366100	1	-2.57390400	7.78019100	-4.58252700

7	1.03891900	3.37239400	0.20297200	1	-1.91225800	0.78610100	-1.53264100
8	-1.20883600	3.76628800	0.33217900	6	-5.16412200	-2.29022900	-0.26620500
8	0.02296300	1.90149100	-1.74602600	6	-6.44904300	-2.66682800	-0.79197500
8	2.20465900	3.13313300	-2.05429200	6	-4.36144700	-3.28620800	0.28073600
16	0.84251900	3.10137000	-1.51193600	6	-7.28061900	-1.75406900	-1.50090500
6	-1.36932000	2.85907600	3.13395700	6	-6.90832000	-4.01902600	-0.64065200
6	-2.66326000	2.51500500	2.74425400	6	-4.81096000	-4.62025900	0.42610000
1	-2.92903700	2.50045200	1.69213400	6	-8.50840800	-2.14261000	-1.99363900
6	-3.60976800	2.18970000	3.72401800	1	-6.93170200	-0.73460000	-1.66013100
1	-4.62312500	1.92074000	3.41750700	6	-8.18550800	-4.38310400	-1.14878000
6	-3.26856200	2.21141100	5.08427900	6	-6.06633100	-4.97323900	-0.00666100
1	-4.02079000	1.95527200	5.83494500	1	-4.13651200	-5.34099900	0.89127300
6	-1.97442700	2.55966000	5.48923400	6	-8.97544000	-3.46624200	-1.80698400
1	-1.71084900	2.57547400	6.55038900	1	-9.12463600	-1.42273600	-2.53744000
6	-1.02285200	2.88536100	4.51797700	1	-8.52505600	-5.41342100	-1.01437000
6	0.37922000	3.26984600	4.61599200	1	-6.42497400	-5.99846200	0.11328800
1	0.93011000	3.40330000	5.54760500	1	-9.95337200	-3.75920000	-2.19581800
6	0.88490700	3.43601800	3.35756600	8	-2.69815400	-1.71268300	-1.42760200
1	1.90463900	3.73036900	3.12101900	8	-3.08918100	-2.99751100	0.72011400
6	-0.18879000	3.49325400	0.94654200	15	-1.81927400	-2.62565300	-0.32577700
6	-0.15036600	3.12919500	2.36476900	7	-1.53900000	-3.94436000	-1.30945600
6	0.44737300	1.08361700	1.60467900	6	7.46883700	-1.04683400	2.76528200
1	-0.16916100	1.26451900	0.72450100	6	6.69559800	-1.10378000	1.62521800
6	-0.01202800	0.02135600	2.44316300	6	5.54102600	-1.93477800	1.55708500
1	0.63305400	-0.31369600	3.25910800	6	5.18404500	-2.68748000	2.72697400
6	-1.28723300	-0.55470000	2.29694400	6	6.00852000	-2.61257000	3.88382300
1	-2.06857300	-0.02473800	1.74855800	6	7.13123300	-1.81512100	3.90609900
1	-1.64213100	-1.27919400	3.03246700	1	8.34627500	-0.39639500	2.79076100
46	-0.13005400	-1.42145200	0.62526200	1	6.95785100	-0.49071200	0.76499000
6	-7.20714400	1.15465900	1.53693500	6	4.70246700	-2.00198700	0.39066300
6	-6.55599900	0.06328500	1.00076600	6	3.99578400	-3.46809700	2.72155300
6	-5.36946400	0.22152700	0.23104200	1	5.72550700	-3.19689500	4.76311000
6	-4.84560400	1.54742500	0.06011100	1	7.75370600	-1.76356100	4.80222400
6	-5.54853000	2.65163000	0.61506000	6	3.18229500	-3.49673400	1.61362300
6	-6.70852100	2.46445700	1.33425500	6	3.54426200	-2.76484100	0.45922200
1	-8.11170300	1.00575900	2.13136400	1	3.72932600	-4.03297100	3.61797000
1	-6.94488200	-0.93926800	1.17614100	1	2.25381900	-4.06945300	1.59533000
6	-4.65926300	-0.89270800	-0.33715600	6	4.98145100	-1.19937300	-0.83041900
6	-3.60270700	1.73124800	-0.60636500	6	6.21530200	-1.29470500	-1.55976900
1	-5.13045600	3.65212500	0.48216600	6	4.01676000	-0.30750800	-1.28734700
1	-7.23236300	3.31990000	1.76651200	6	7.19621200	-2.28553100	-1.27358300
6	-2.89269300	0.65182000	-1.07663300	6	6.46454800	-0.38676700	-2.64385800
6	-3.43410200	-0.64686500	-0.94134800	6	4.26363400	0.60647500	-2.33877800
1	-3.17835700	2.73359400	-0.68347000	6	8.37053900	-2.35489800	-1.99338700

1	7.00471100	-3.00337200	-0.47589900	6	-0.24463900	-0.04428600	2.42604800
6	7.69153500	-0.47733900	-3.35618000	1	0.41652700	-0.28428900	3.26214300
6	5.47432400	0.57565700	-2.98817600	6	-1.45085600	-0.75815900	2.24280400
1	3.48556800	1.33319400	-2.57684300	1	-2.28102900	-0.28667500	1.70885700
6	8.62967500	-1.43596600	-3.03904200	1	-1.74015800	-1.52621100	2.96359900
1	9.10513300	-3.12863300	-1.75773400	6	1.39421000	3.60840900	-2.30441200
1	7.87261800	0.22746200	-4.17193400	6	2.61786100	2.94609100	-2.40311600
1	5.68404000	1.28438800	-3.79319200	1	2.73511600	1.96829200	-1.93681100
1	9.56644600	-1.49662500	-3.59782800	6	3.66759300	3.57142900	-3.07479700
8	2.69656400	-2.81360600	-0.62783700	1	4.63549100	3.06795600	-3.14122400
8	2.77475300	-0.27552500	-0.70287400	6	3.50419500	4.83969300	-3.65445300
15	1.65195500	-1.51428300	-0.85794000	6	2.25296900	5.47263100	-3.54603700
7	1.36915000	-1.76055000	-2.48639500	1	2.10791700	6.45749000	-3.99742800
6	0.18404200	-1.16623200	-3.08810500	6	1.19299300	4.86696700	-2.87391600
1	-0.30880200	-1.89689300	-3.75149600	1	0.21796000	5.35004100	-2.79444000
1	-0.53357100	-0.87773000	-2.31382300	6	4.63236200	5.50595900	-4.39759600
1	0.43472100	-0.26152600	-3.66829300	1	4.45936200	5.46845900	-5.48672300
6	2.38916100	-2.18871200	-3.43512300	1	4.72069100	6.56833900	-4.12186500
1	1.95202300	-2.92511700	-4.13182600	1	5.59572600	5.01598900	-4.19569700
1	2.77664500	-1.33975200	-4.02495000	7	0.36196700	3.40428600	0.20467000
1	3.22509400	-2.66255900	-2.90936800	8	-1.21306400	3.34648200	-1.86172600
6	-0.29365200	-4.68350900	-1.18854600	8	0.32709900	1.37767200	-1.47242200
1	0.24107900	-4.70531900	-2.15379800	16	0.07379500	2.83253100	-1.40333300
1	0.36682600	-4.20124100	-0.45652100	6	-1.68017200	4.56805200	0.93547900
1	-0.47616800	-5.72420300	-0.86509500	6	-2.92657500	4.73088800	1.67962300
6	-2.50304500	-4.48563600	-2.25678200	6	-3.48700900	4.02832300	2.75162800
1	-2.03563000	-4.57976200	-3.25298900	1	-2.95006000	3.18597300	3.18791300
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6	2.76292700	3.52181000	0.70420300	1	-5.40282900	7.07755900	1.17414500
1	2.80536400	4.47303400	0.17325500	6	-3.61314100	5.85114000	1.11078800
6	3.89759200	2.97114100	1.30411200	6	-2.77223300	6.40204900	0.05883400
1	4.85577800	3.49083000	1.23393300	1	-3.01604400	7.27832900	-0.54276300
6	3.80348100	1.76472400	2.00496300	6	-1.63147700	5.64835600	-0.03353400
1	4.68684300	1.33400900	2.47712000	1	-0.80103200	5.82173500	-0.71424000
6	2.58124800	1.10076500	2.10554400	8	-0.79524700	2.72401600	2.11314100
1	2.52511300	0.14805500	2.63304400	46	-0.14832300	-1.41802600	0.66689500
6	1.43979400	1.63141100	1.49297900	6	-7.16422700	0.77508200	1.56085800
6	0.10400500	1.02212200	1.55238600	6	-6.47736400	-0.28724100	1.00957200
1	-0.50156500	1.11267100	0.65736500	6	-5.29552000	-0.07508300	0.24674300
6	-0.77067800	3.54840600	1.14102800	6	-4.81212300	1.27160300	0.10500500

6	-5.55199200	2.34395600	0.67385000	6	2.57664900	-3.14647900	2.23373000
6	-6.70864200	2.10302900	1.38223100	6	3.25206700	-2.63918400	1.09981000
1	-8.06666700	0.58666600	2.14786500	1	2.58230900	-3.30737300	4.37279900
1	-6.83651100	-1.30443900	1.16383400	1	1.64774900	-3.69891900	2.08497800
6	-4.55546200	-1.15299200	-0.35350400	6	5.04254800	-1.38427600	-0.05182200
6	-3.59024000	1.51363200	-0.58145000	6	6.39710700	-1.67754200	-0.43263000
1	-5.17339900	3.36043900	0.55975300	6	4.26312100	-0.61267700	-0.90900100
1	-7.25412800	2.93744400	1.82687900	6	7.22035000	-2.57432600	0.30553200
6	-2.86096400	0.46719800	-1.09900900	6	6.94295700	-1.08383700	-1.62027000
6	-3.35124200	-0.85044500	-0.97144800	6	4.79741500	-0.04577900	-2.09200400
1	-3.21861200	2.53522600	-0.67980600	6	8.51850500	-2.83271100	-0.08112500
1	-1.91022400	0.63580000	-1.60147800	1	6.80951200	-3.06780300	1.18573000
6	-5.01400000	-2.56807900	-0.31298600	6	8.28911800	-1.36109600	-1.98392900
6	-6.28594700	-2.97521200	-0.84767200	6	6.11339900	-0.25691600	-2.42578900
6	-4.18053500	-3.55084500	0.20858300	1	4.13807600	0.54652100	-2.72609000
6	-7.14792200	-2.07209600	-1.53194800	6	9.06703000	-2.21191300	-1.22947900
6	-6.69884100	-4.34572500	-0.73094400	1	9.12771500	-3.52779800	0.50135000
6	-4.58173800	-4.90261600	0.32058200	1	8.69284900	-0.89203000	-2.88492100
6	-8.36206500	-2.48916100	-2.03473600	1	6.53035600	0.18714900	-3.33309200
1	-6.83388000	-1.03735600	-1.66201800	1	10.09890200	-2.42017300	-1.52102500
6	-7.96359100	-4.73938700	-1.24842800	8	2.68347800	-2.83463400	-0.14571300
6	-5.82490300	-5.28708400	-0.12160700	8	2.94287500	-0.36377800	-0.62423700
1	-3.88372500	-5.61176400	0.76874000	15	1.73628300	-1.54880800	-0.69026400
6	-8.78381600	-3.83243300	-1.88270800	7	1.58409000	-2.01522900	-2.28225600
1	-9.00308100	-1.77628600	-2.55861100	6	0.44658600	-1.52951200	-3.05253300
1	-8.26865900	-5.78353700	-1.14056200	1	-0.04044200	-2.36686800	-3.57924000
1	-6.14868500	-6.32650000	-0.02754400	1	-0.29368000	-1.06323700	-2.39375100
1	-9.75184900	-4.14767500	-2.27891200	1	0.76151600	-0.77286000	-3.79203900
8	-2.57353100	-1.88891700	-1.47677900	6	2.65513000	-2.61978800	-3.06028100
8	-2.91636100	-3.22676700	0.65899500	1	2.25295400	-3.46742800	-3.64222300
15	-1.67640100	-2.76296100	-0.37515200	1	3.10142400	-1.89859200	-3.76761400
7	-1.29184200	-4.05238200	-1.36255300	1	3.44178300	-2.99918400	-2.39821000
6	6.57993900	-0.56736700	3.98138300	6	-0.07060400	-4.80322000	-1.12688400
6	6.11184400	-0.82892300	2.71091100	1	0.50491900	-4.90256700	-2.06326500
6	4.96256400	-1.64303300	2.50258100	1	0.56833800	-4.28524200	-0.40066900
6	4.28424300	-2.16197800	3.65734700	1	-0.28756700	-5.81754000	-0.74470900
6	4.79913000	-1.88009600	4.95291800	6	-2.21319500	-4.63805400	-2.32753700
6	5.92469300	-1.10333300	5.11647600	1	-1.69883000	-4.77543300	-3.29484200
1	7.46104200	0.06481300	4.11340300	1	-2.58204800	-5.62174400	-1.98724500
1	6.61900500	-0.39818500	1.84886600	1	-3.07087500	-3.97483300	-2.48529100
6	4.43257000	-1.91553100	1.19503500				
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1	4.27457600	-2.28618600	5.82142900				TS1c
1	6.30663000	-0.89011000	6.11723400	E =	-4603.42184408	G = -4602.406674	

6	1.86560400	2.24533200	0.52792800	6	0.59676400	4.02829600	2.14268900
6	3.21613000	2.49749400	0.25594300	6	0.08817000	0.92102700	1.68815000
1	3.47161200	3.32959500	-0.40142300	1	-0.61010100	1.44936100	1.04294100
6	4.21284600	1.73251700	0.84965600	6	-0.38740000	-0.38146700	2.11257300
1	5.26351900	1.94362500	0.64228300	1	0.30595600	-1.04943100	2.62964700
6	3.85568800	0.70105600	1.72412400	6	-1.74092500	-0.74123300	2.03486900
1	4.62469500	0.10827800	2.21798400	1	-2.50202500	0.01177400	1.80984200
6	2.51953000	0.42932700	1.97439200	1	-2.10117000	-1.63897300	2.54157000
1	2.26672300	-0.38197100	2.65574400	46	-0.88650900	-1.27369500	0.06439900
6	1.48517000	1.18242600	1.38066900	6	-6.13134900	2.37653800	2.86103700
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6	2.15625700	4.77525000	-2.73980600	6	-5.07831300	1.37714600	0.90017800
1	1.60308700	5.53199100	-2.18006800	6	-4.21177400	2.51991200	0.80700800
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1	4.13217700	1.99840000	-4.62837000	6	-4.93106600	0.30291500	-0.04438000
6	2.46966600	2.42739800	-3.32652300	6	-3.20333600	2.55528800	-0.19552100
1	2.14696200	1.39178100	-3.21826600	1	-3.70676100	4.45328600	1.64762300
6	5.18185000	4.49552600	-5.04835000	1	-5.39965300	4.33804600	3.46033700
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7	0.91284400	3.18004800	-0.02568300	1	-2.20633700	1.47203700	-1.76279100
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1	3.37994400	4.88319600	2.59939700	6	-9.22286600	0.69516600	-0.70898900
6	3.76992000	3.49988100	4.22312700	1	-7.22570400	1.45001900	-0.58104900
1	4.80231700	3.83361500	4.35491100	6	-9.52246900	-1.68788700	-0.45333300
6	3.26463400	2.47762400	5.04435700	6	-7.54718600	-3.14601300	-0.11543700
1	3.90835500	2.02837200	5.80445900	1	-5.72184400	-4.27825200	0.10266800
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6	4.54220300	-0.36628200	-1.87493600	6	-1.19435600	-0.81682700	0.62771700	
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1	0.41399300	-6.18351000	3.65084100	1	5.44827100	2.02129400	-2.69589100
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6	-2.39592100	-6.11360300	-0.14828300	1	7.79160000	2.22163500	-1.83480300
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6	-3.67050300	-2.68023900	-0.22480100	15	2.87513200	-0.86585600	-2.53947100
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6	-8.67846800	-3.57703800	-1.32004400	8	2.44099200	-3.30201900	0.02914600
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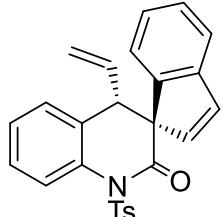
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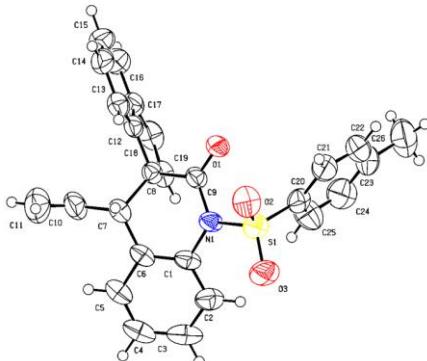
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1	3.19273000	2.88655300	2.19258600	1	5.39951500	-0.12760400	-3.55314800

8. X-Ray Structures of Product 3aa



3aa
CCDC: 2271263



Supplementary Figure 8 X-ray crystallography of **3aa**

Crystal data and structure refinement for mo_230601a_0m.

Identification code	mo_230601a_0m	
Empirical formula	C26 H21 N O3 S	
Formula weight	427.50	
Temperature	296.15 K	
Wavelength	0.71073 Å	
Crystal system	Monoclinic	
Space group	P 1 2 1 1	
Unit cell dimensions	a = 9.163(2) Å b = 12.687(3) Å c = 9.805(3) Å	a= 90° b= 104.805(4)° g = 90°
Volume	1102.1(5) Å ³	
Z	2	
Density (calculated)	1.288 Mg/m ³	
Absorption coefficient	0.174 mm ⁻¹	
F(000)	448	
Crystal size	0.3 x 0.2 x 0.2 mm ³	
Theta range for data collection	2.299 to 29.028°	
Index ranges	-12<=h<=12, -16<=k<=17, -13<=l<=13	
Reflections collected	10523	
Independent reflections	5579 [R(int) = 0.0418]	
Completeness to theta = 25.242?	99.7 %	
Absorption correction	Semi-empirical from equivalents	
Max. and min. transmission	0.7458 and 0.5818	
Refinement method	Full-matrix least-squares on F ²	
Data / restraints / parameters	5579 / 1 / 277	
Goodness-of-fit on F ²	0.974	

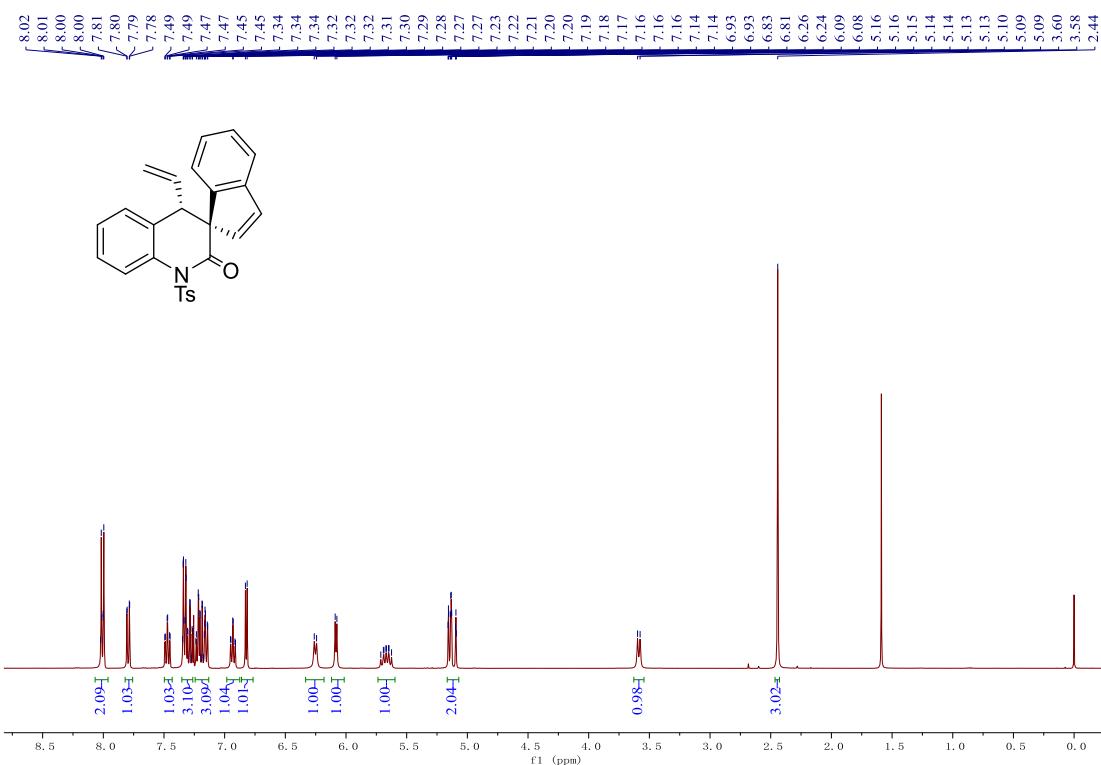
Final R indices [$I > 2\sigma(I)$]	R1 = 0.0487, wR2 = 0.1050
R indices (all data)	R1 = 0.0937, wR2 = 0.1244
Absolute structure parameter	0.09(6)
Extinction coefficient	0.012(3)
Largest diff. peak and hole	0.290 and -0.229 e. \AA^{-3}

References

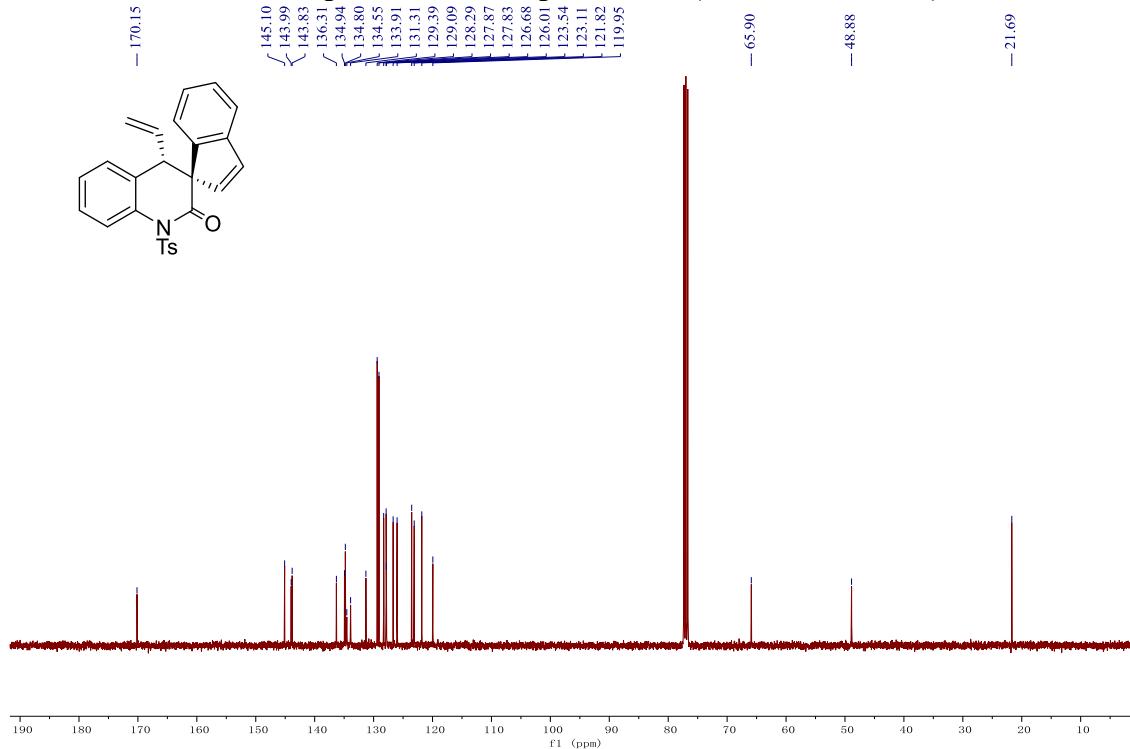
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9. Copies of NMR Spectra

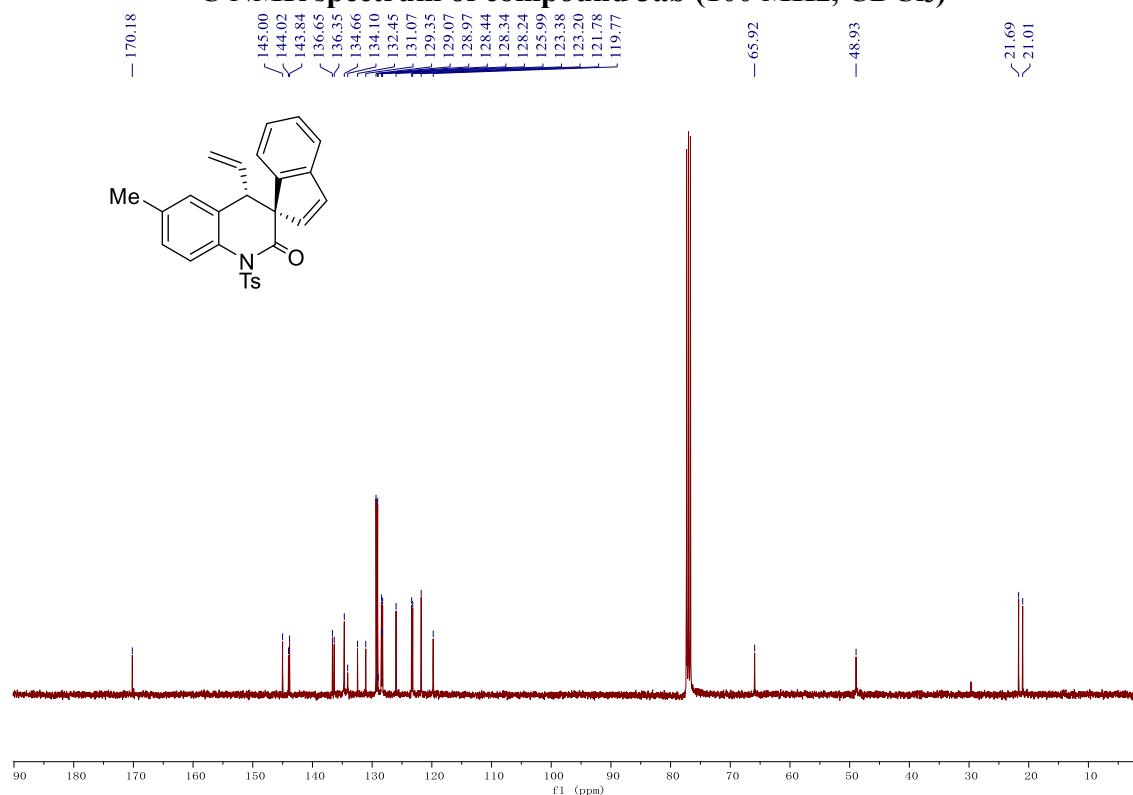
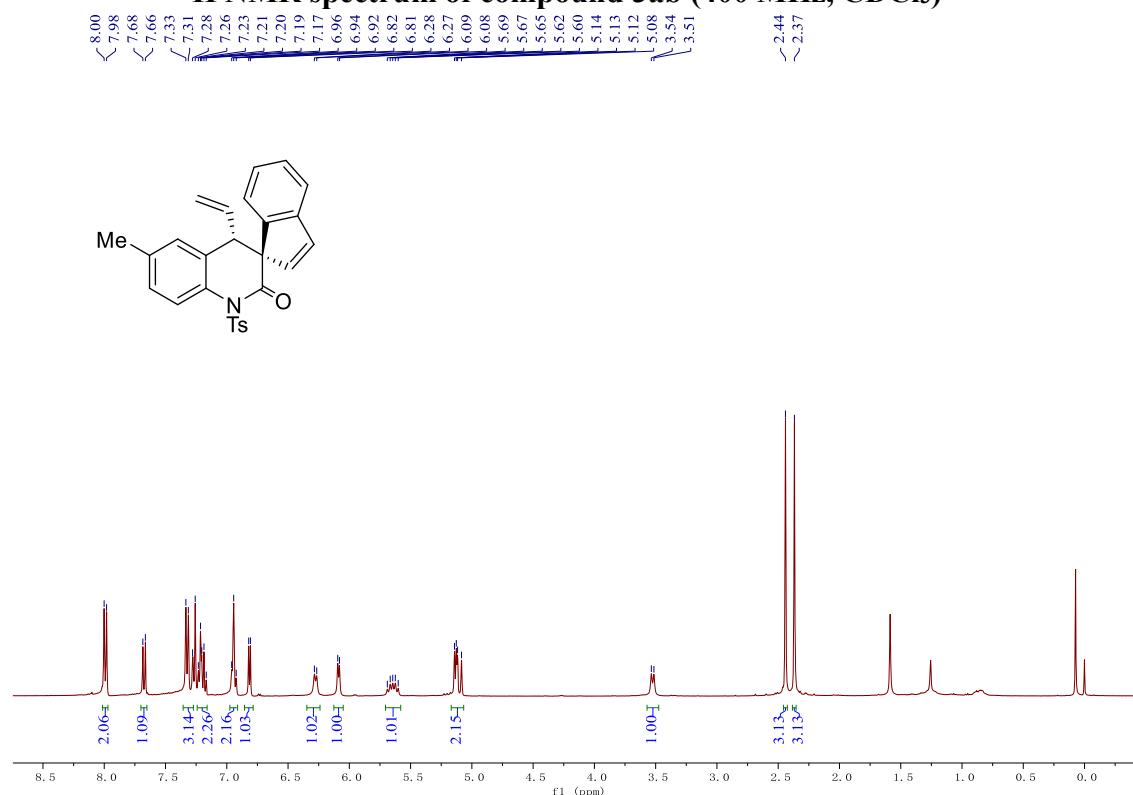
^1H NMR spectrum of compound 3aa (400 MHz, CDCl_3)



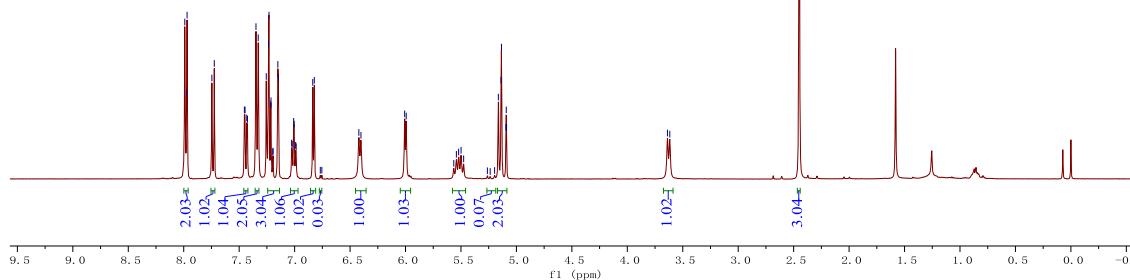
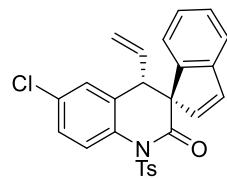
^{13}C NMR spectrum of compound 3aa (100 MHz, CDCl_3)



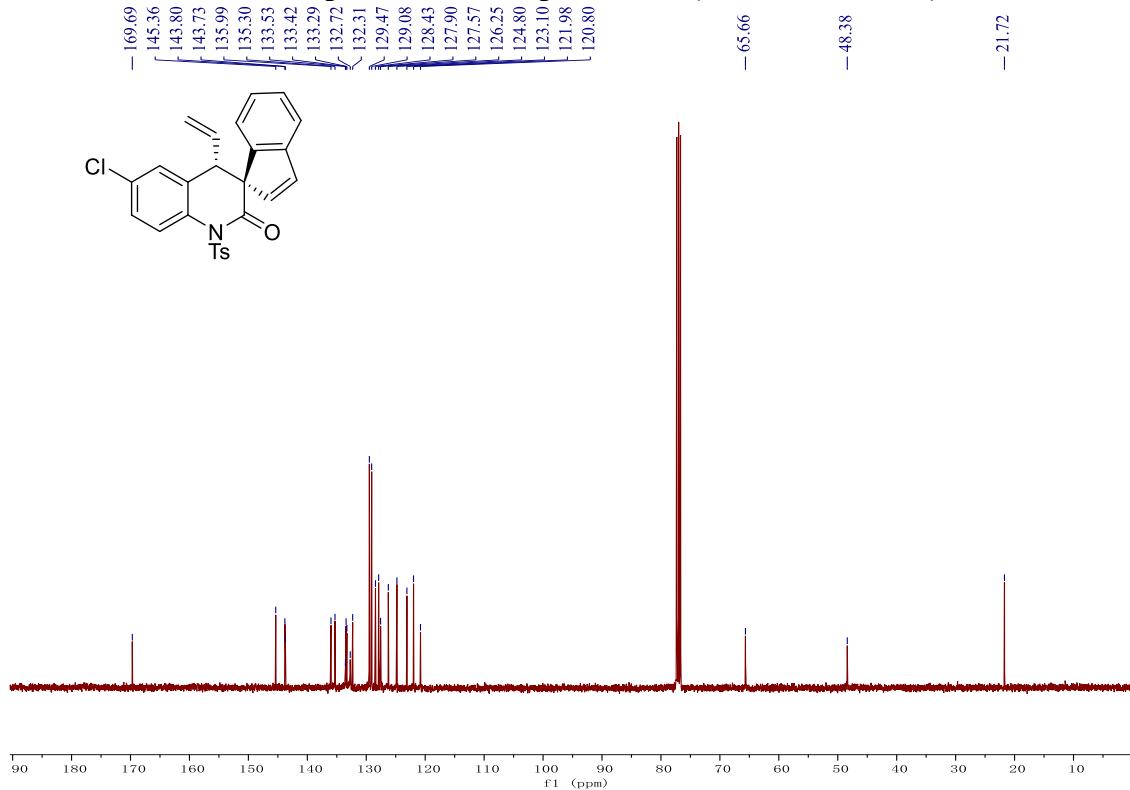
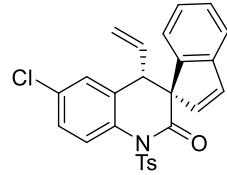
¹H NMR spectrum of compound 3ab (400 MHz, CDCl₃)



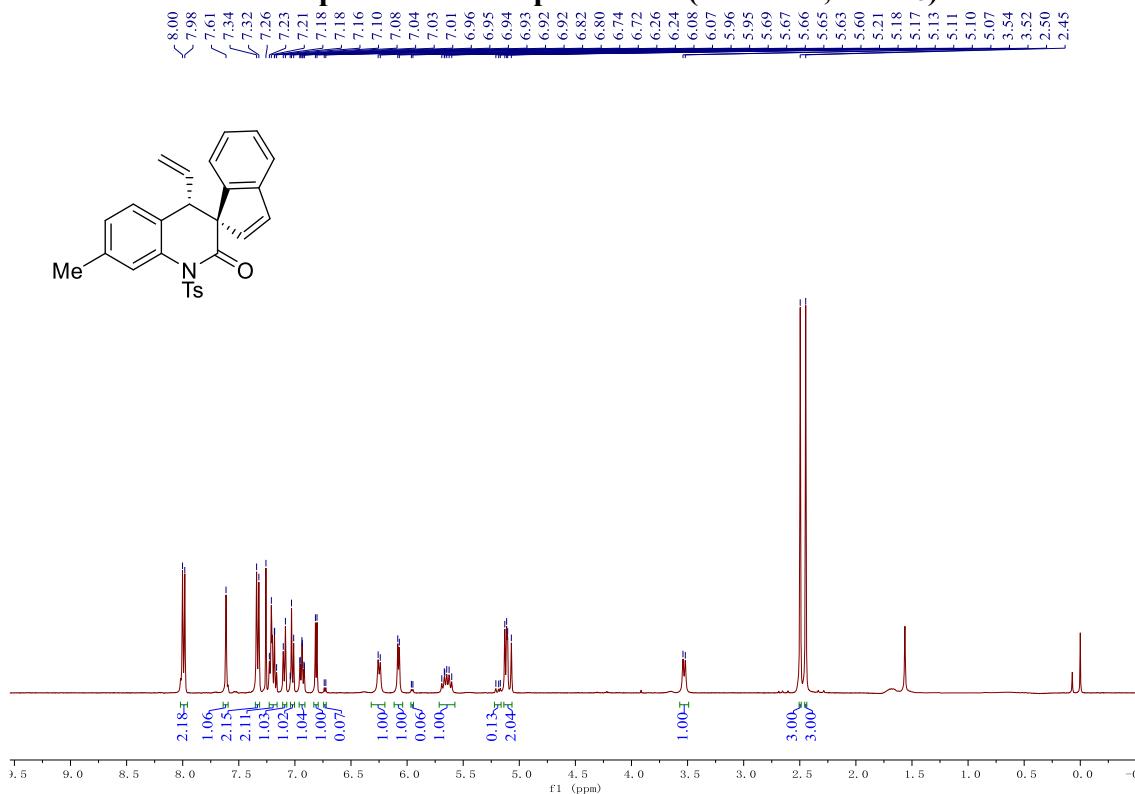
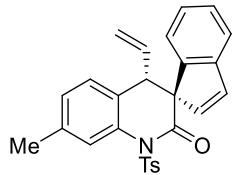
¹H NMR spectrum of compound 3ac (400 MHz, CDCl₃)



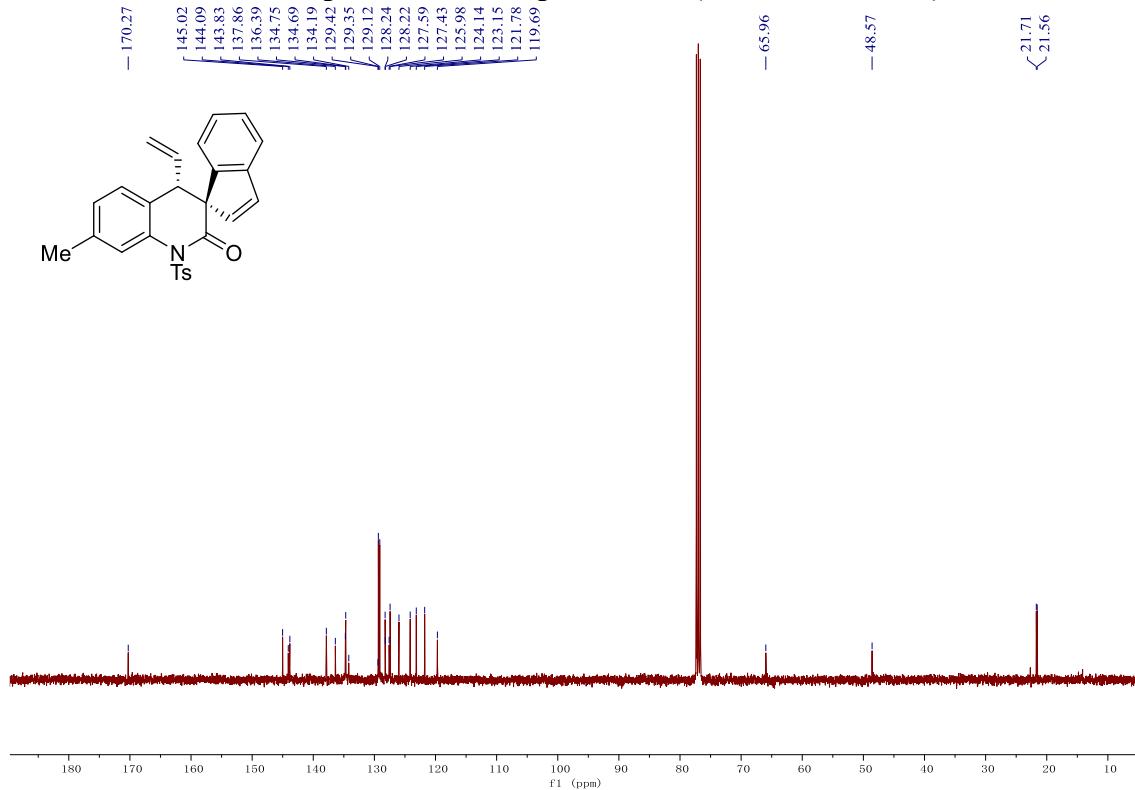
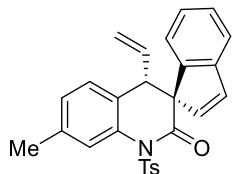
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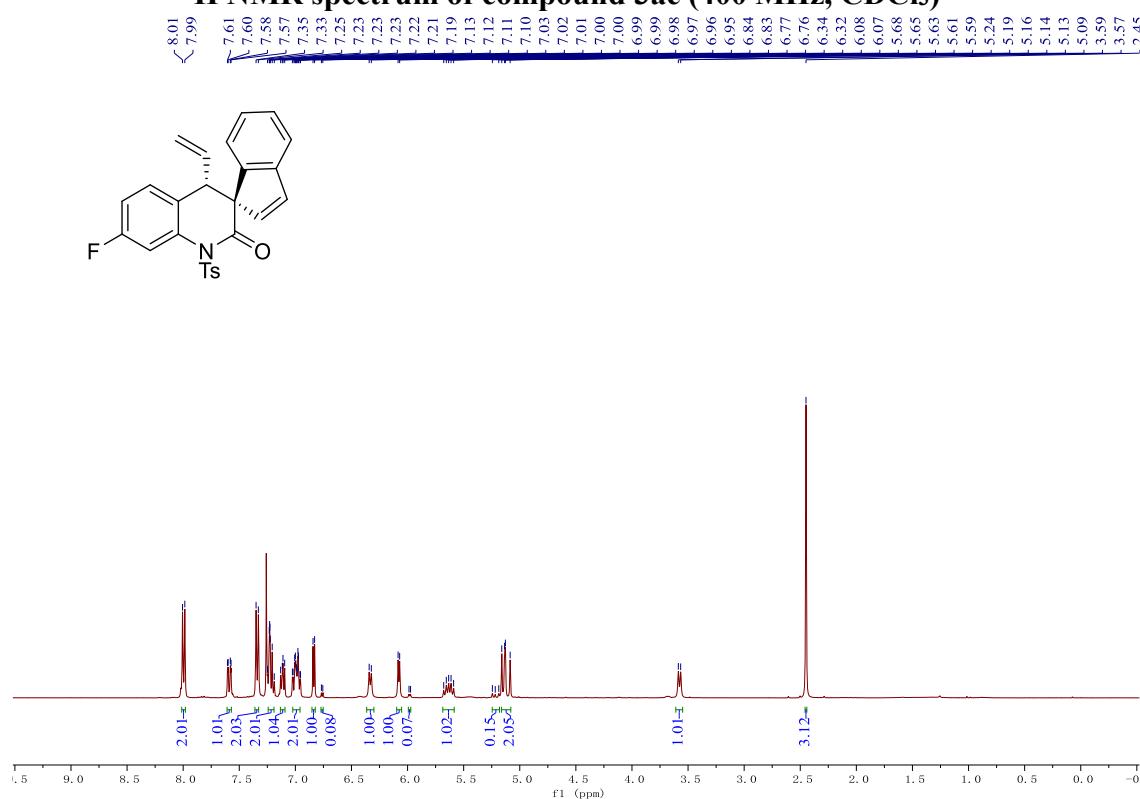
¹H NMR spectrum of compound 3ad (400 MHz, CDCl₃)



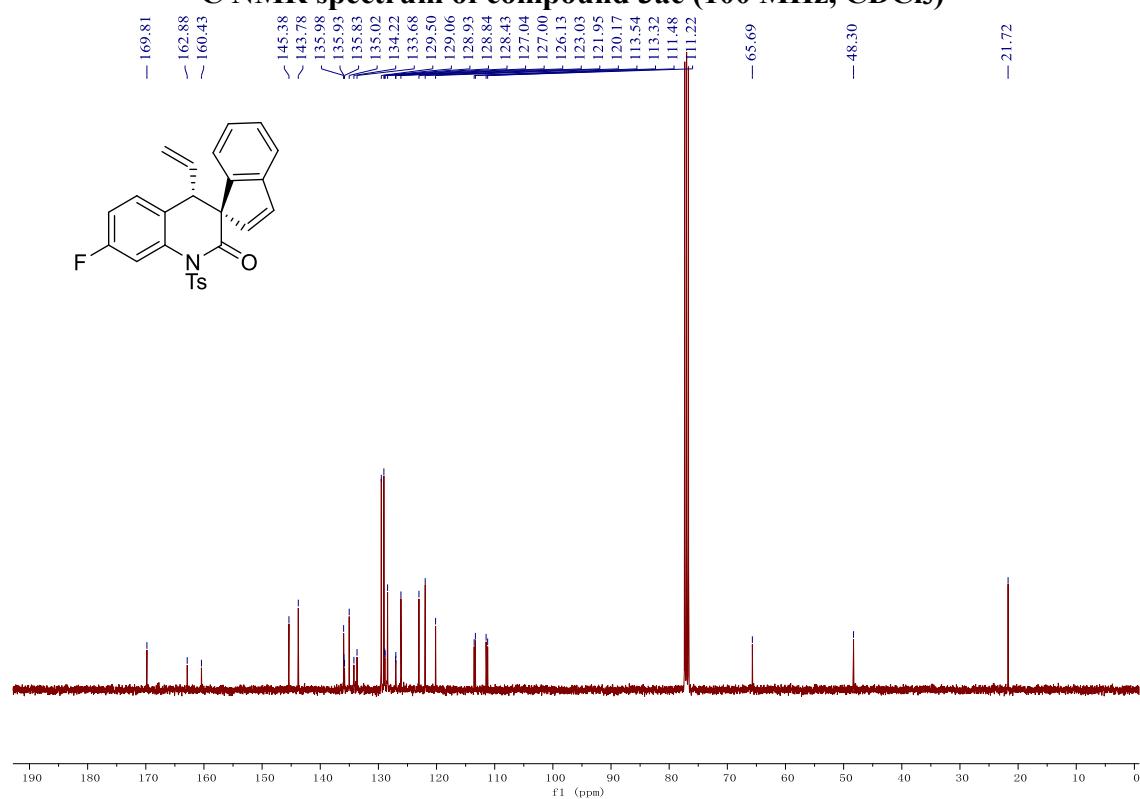
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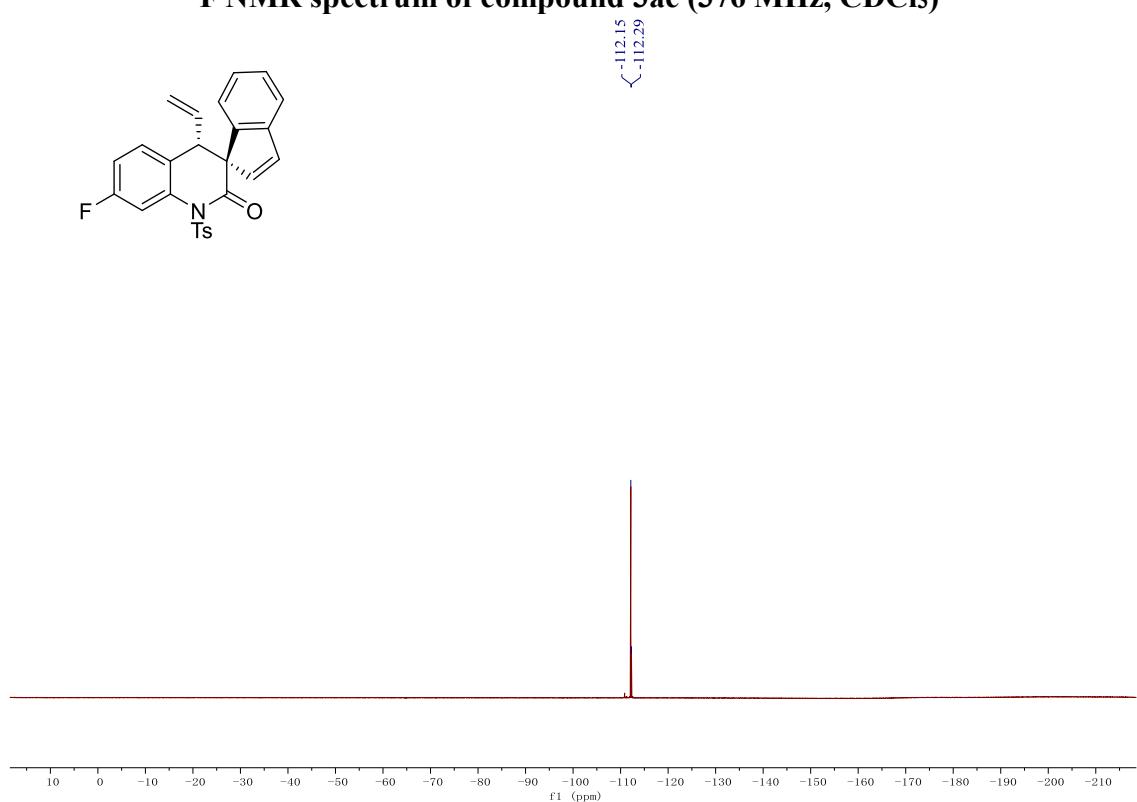
¹H NMR spectrum of compound 3ae (400 MHz, CDCl₃)



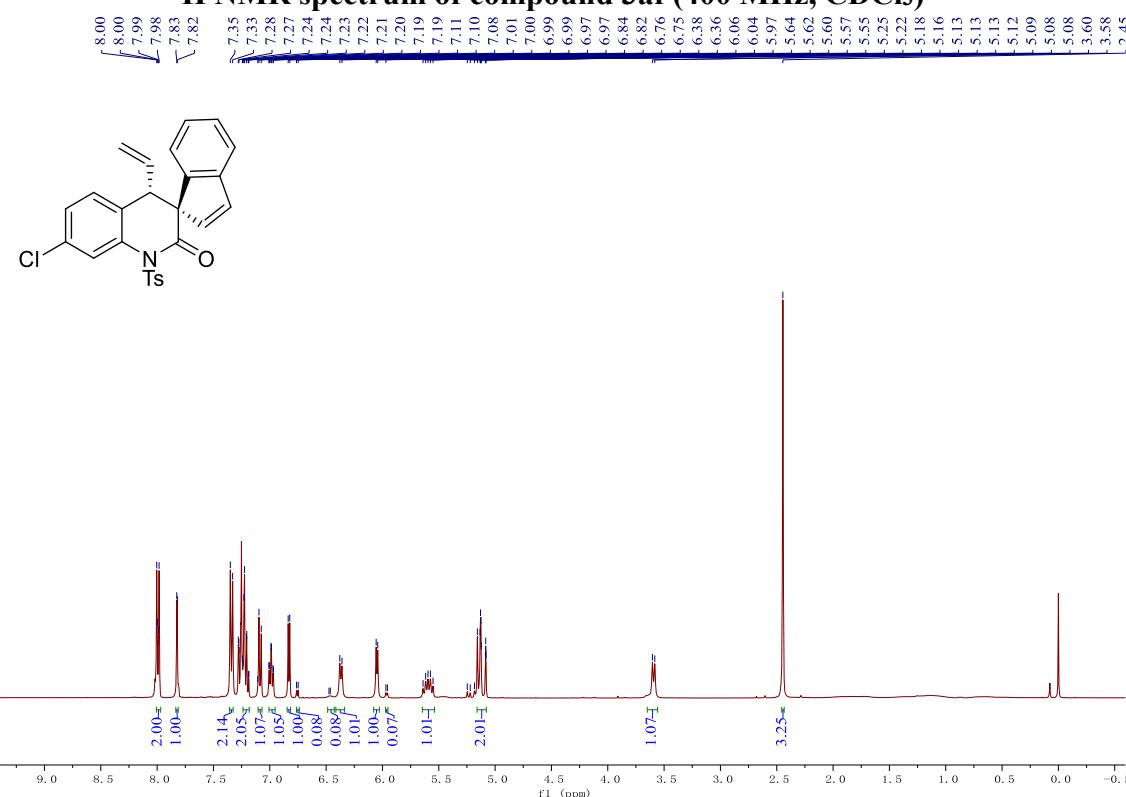
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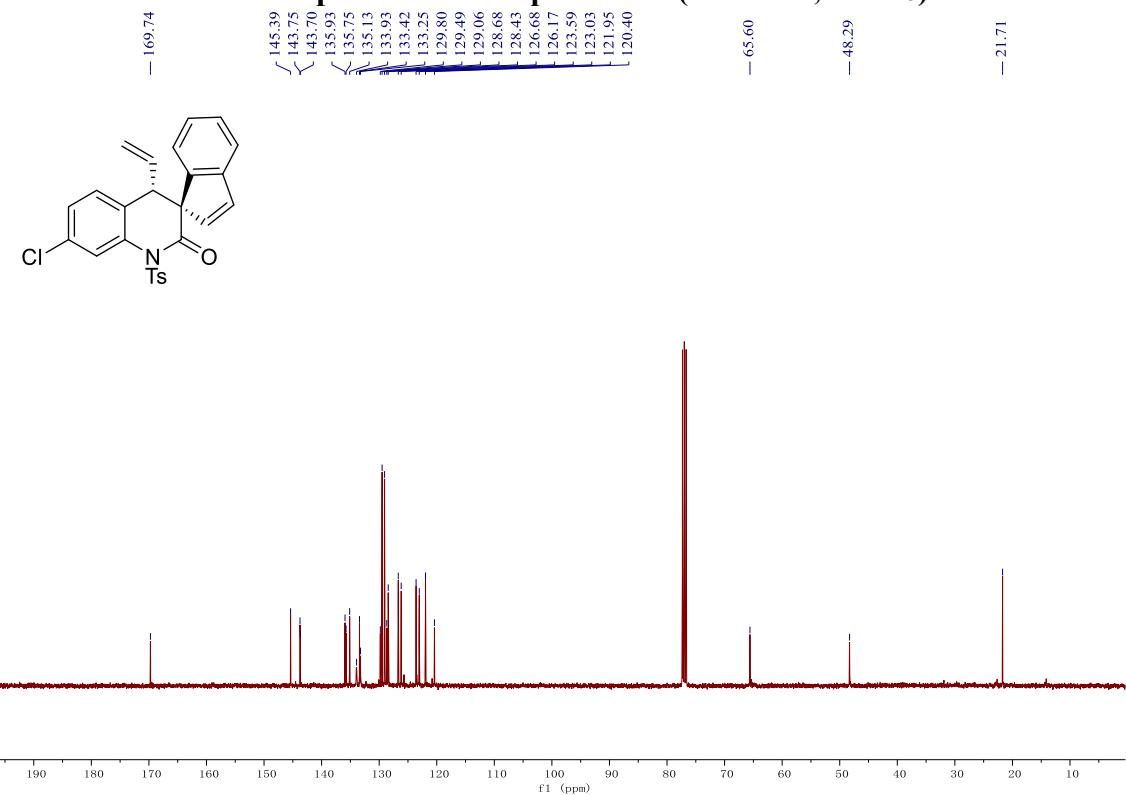
¹⁹F NMR spectrum of compound 3ae (376 MHz, CDCl₃)



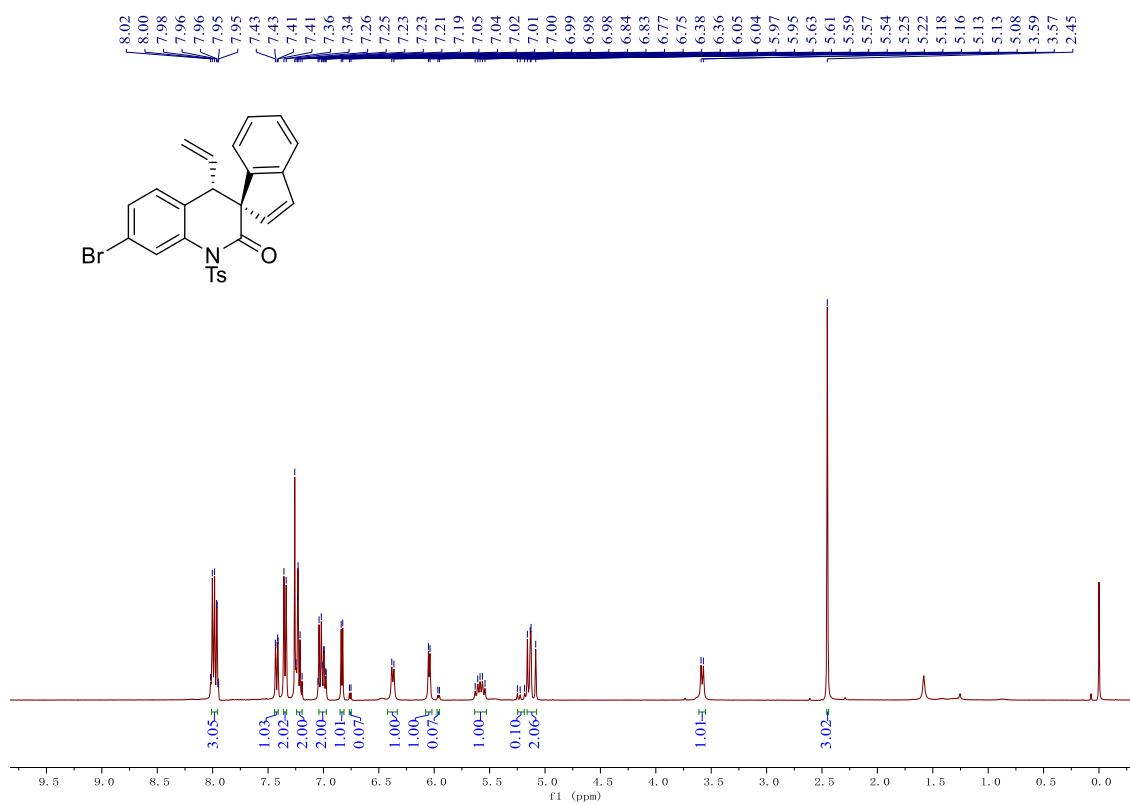
¹H NMR spectrum of compound 3af (400 MHz, CDCl₃)



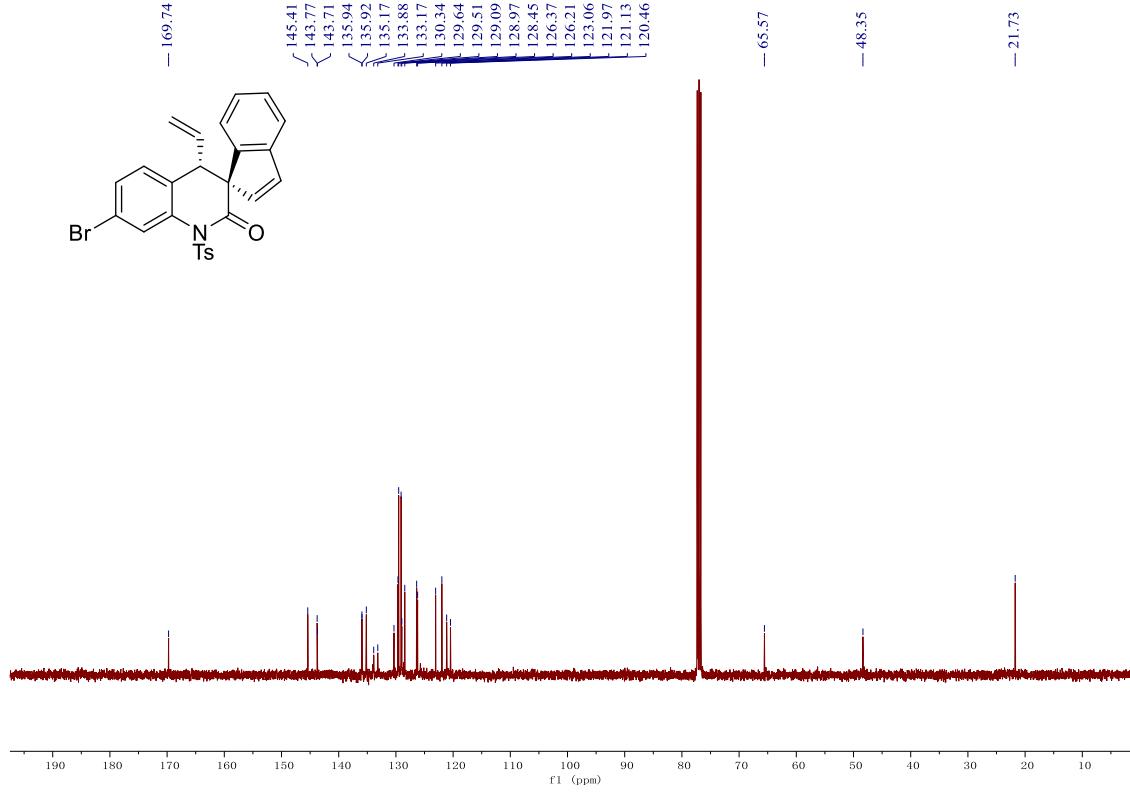
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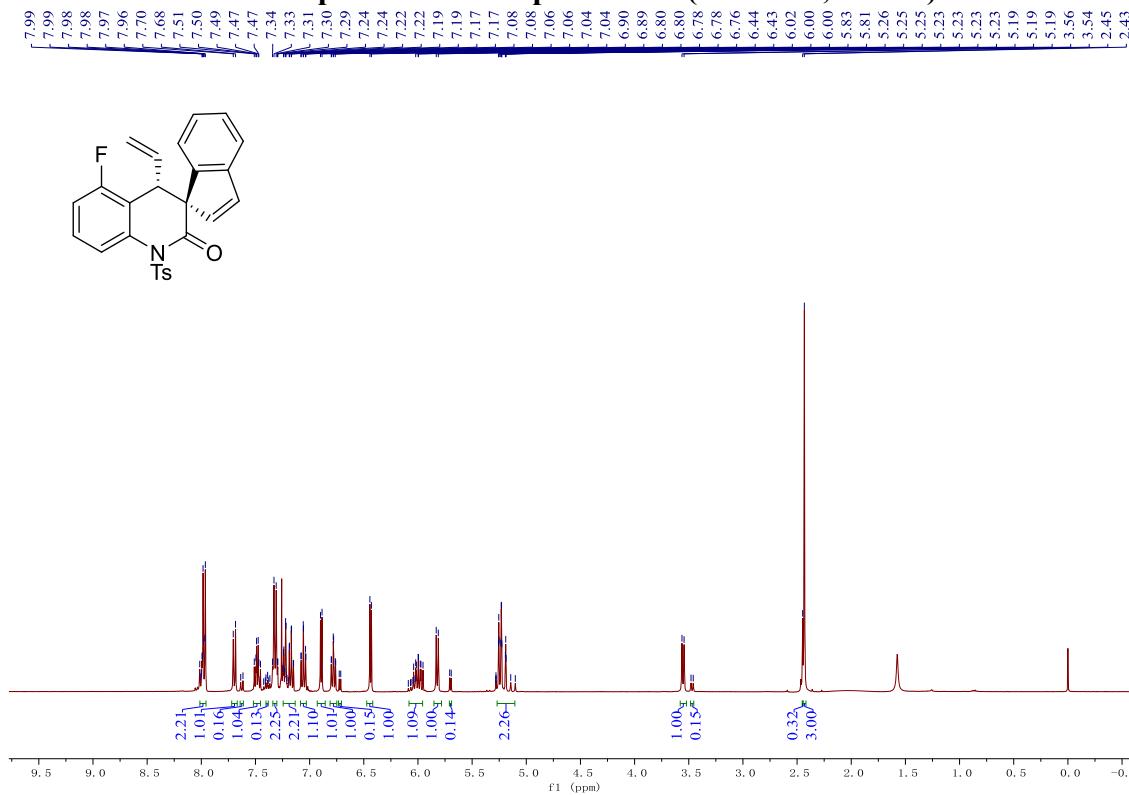
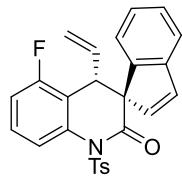
¹H NMR spectrum of compound 3ag (400 MHz, CDCl₃)



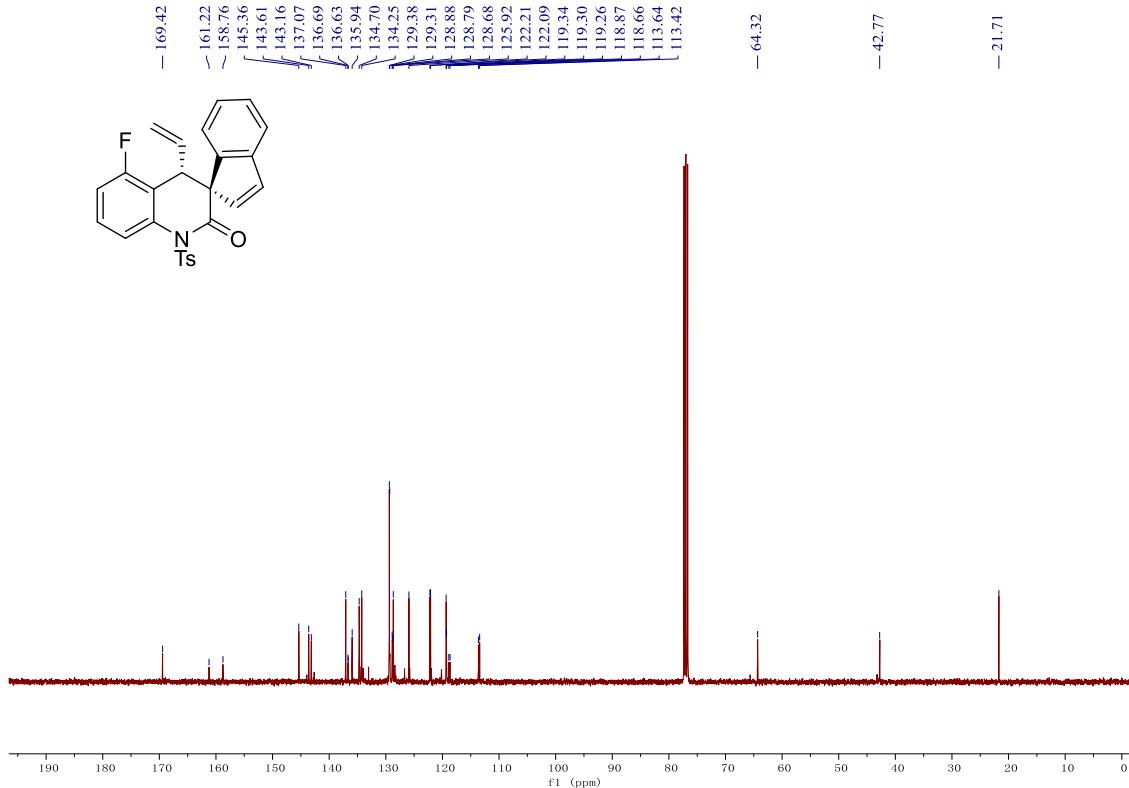
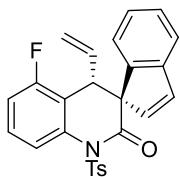
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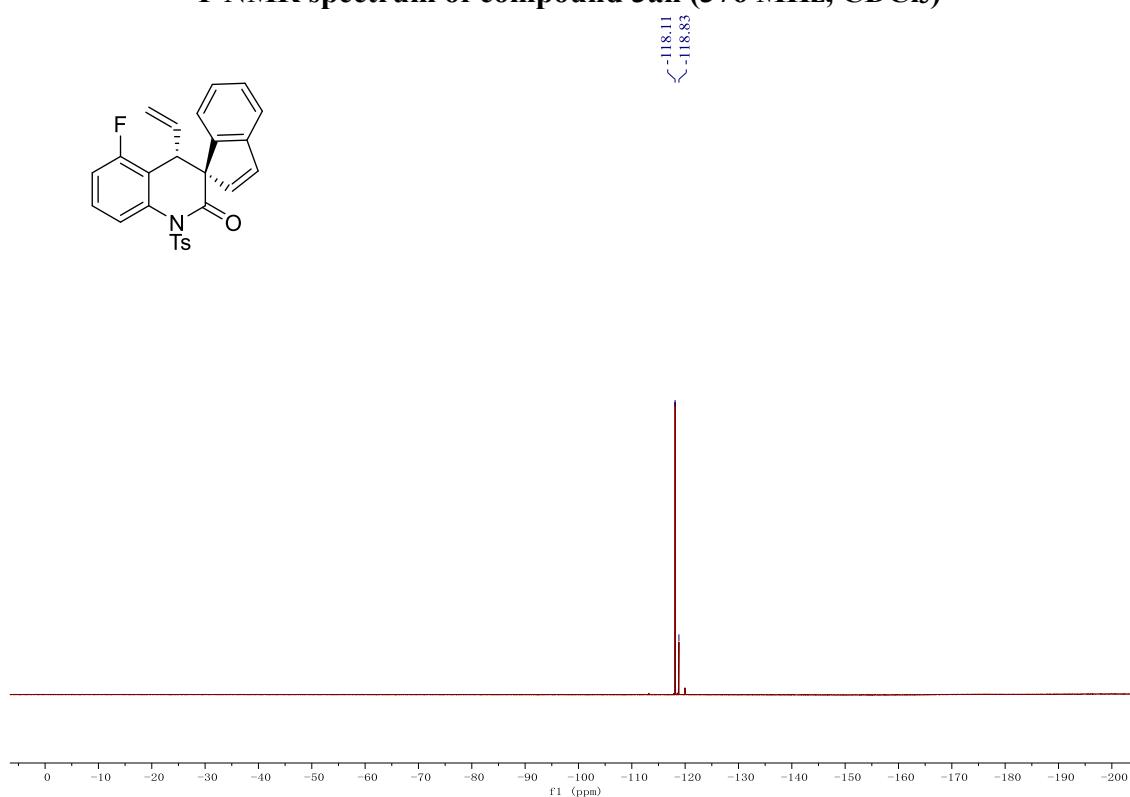
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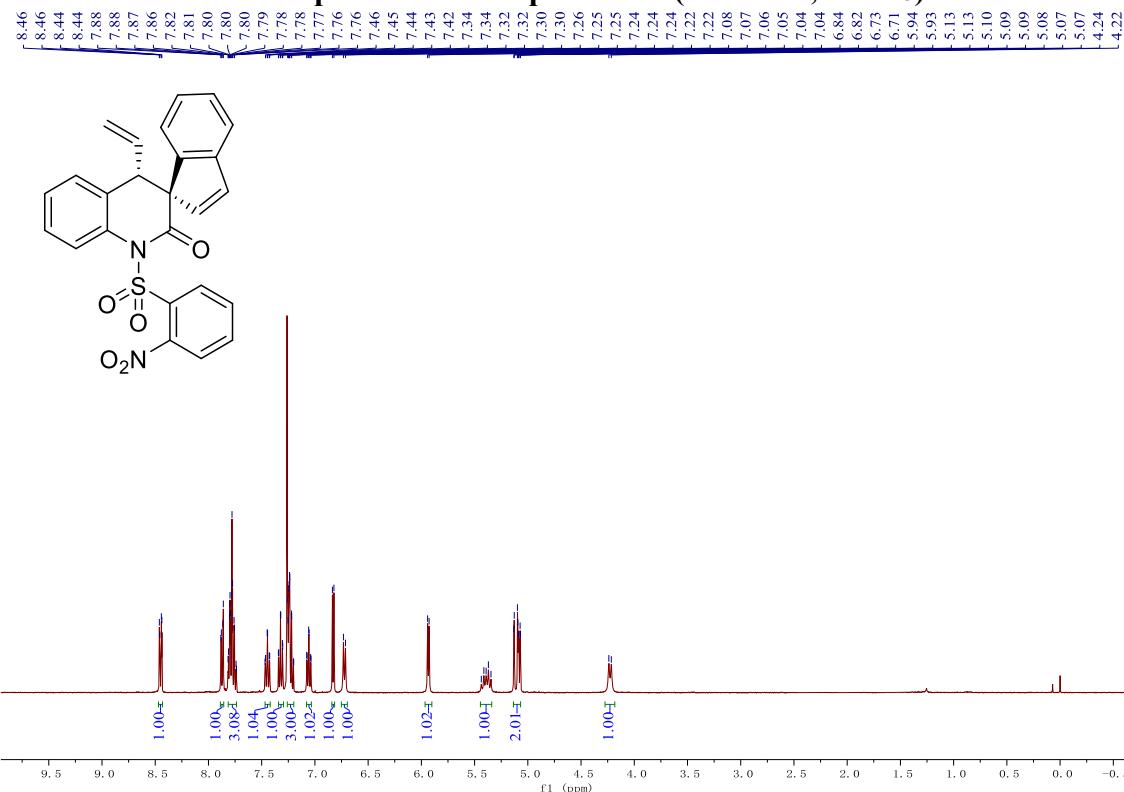
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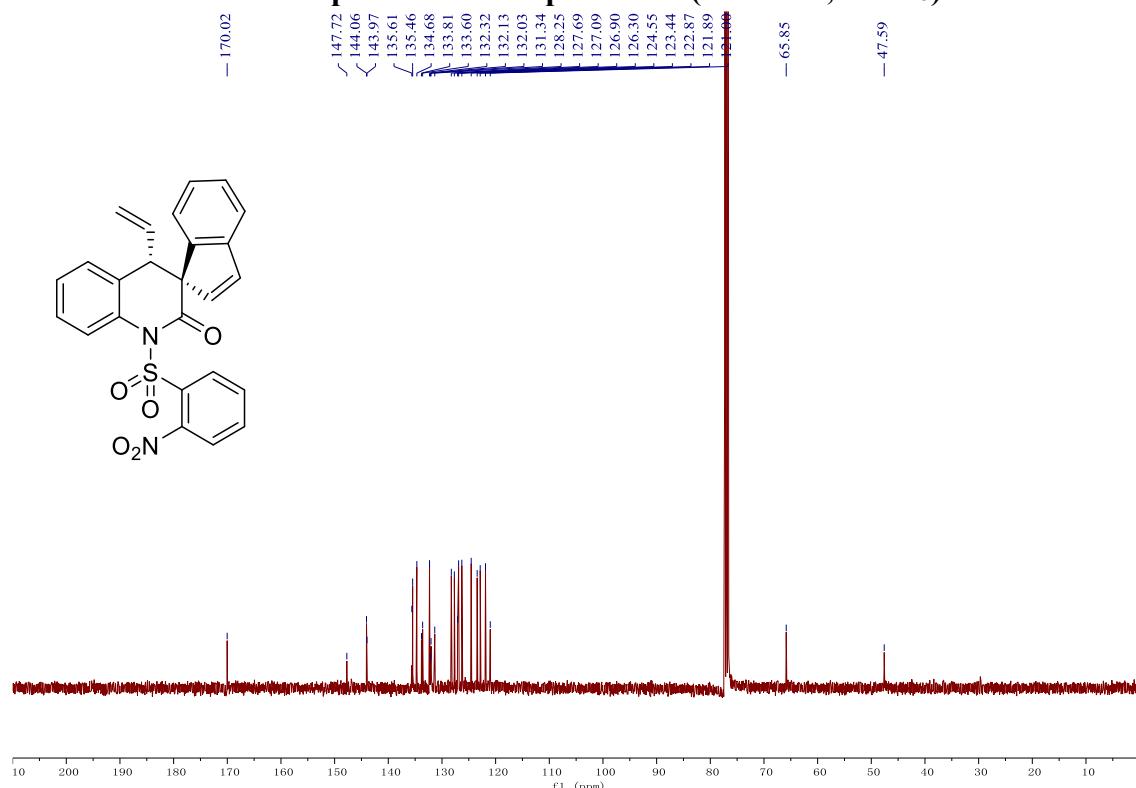
¹⁹F NMR spectrum of compound 3ah (376 MHz, CDCl₃)



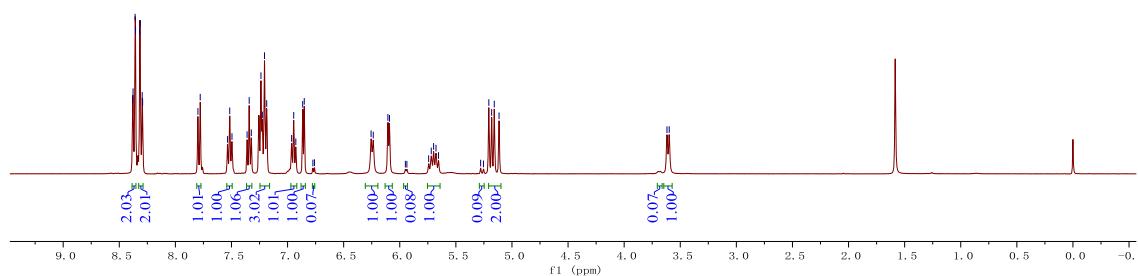
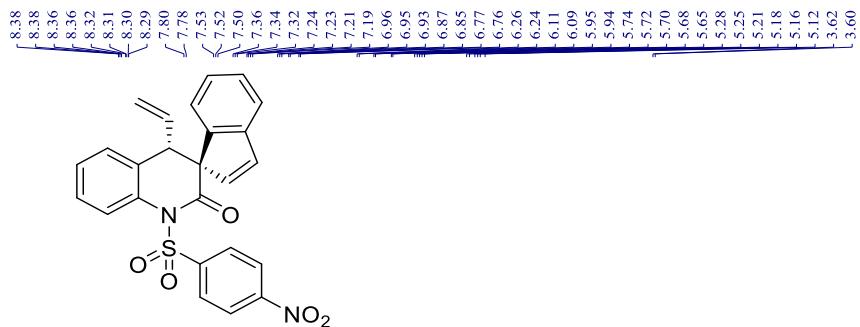
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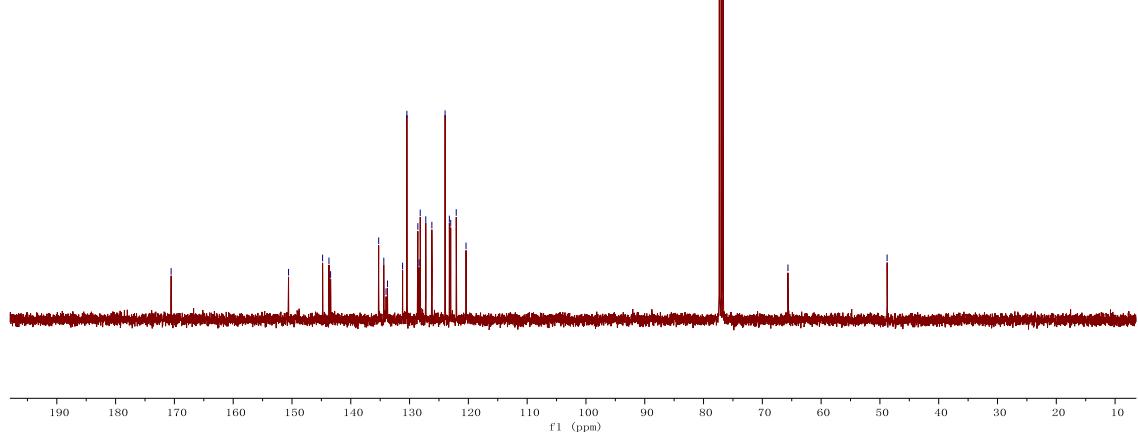
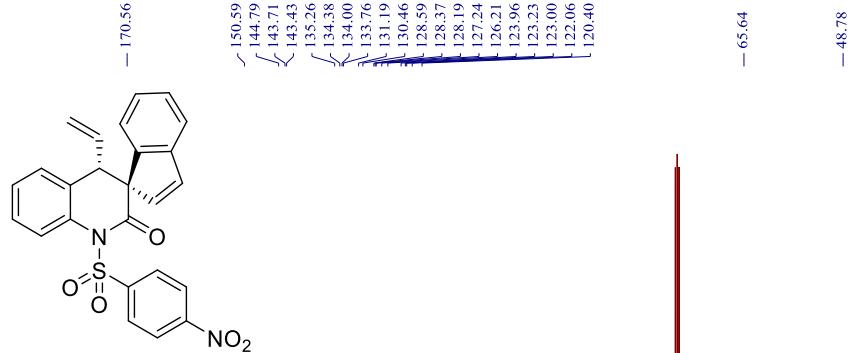
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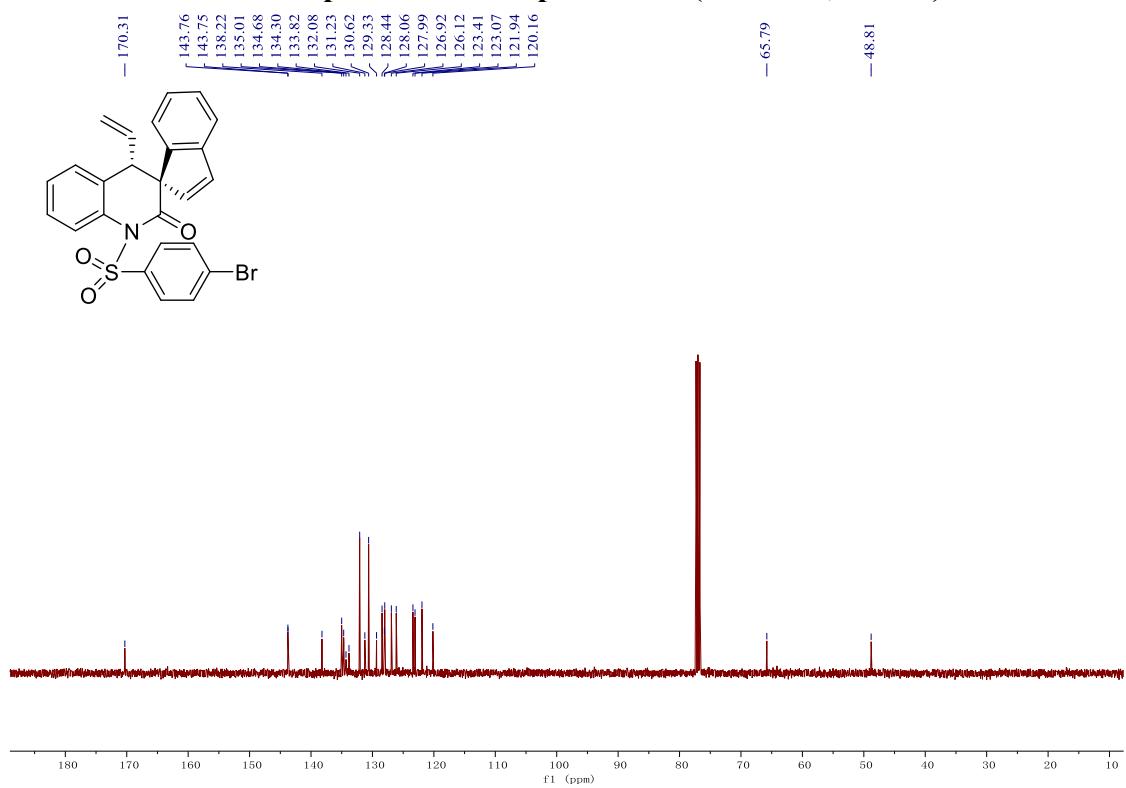
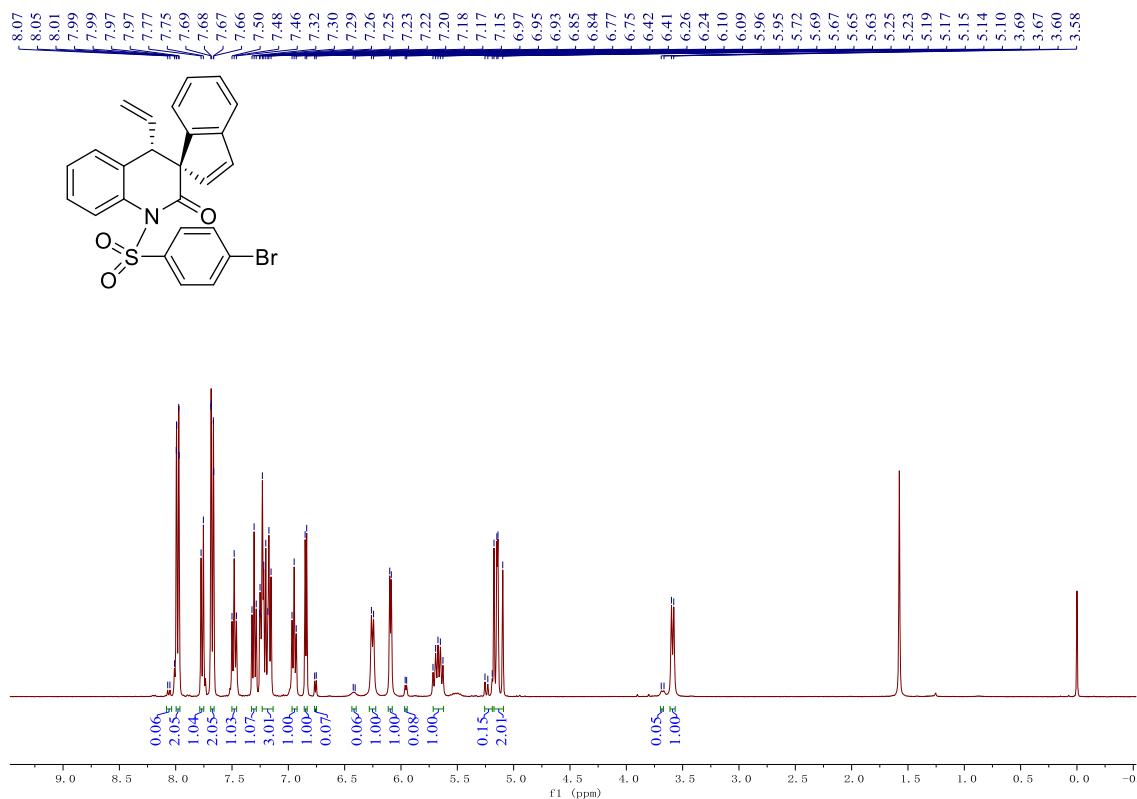
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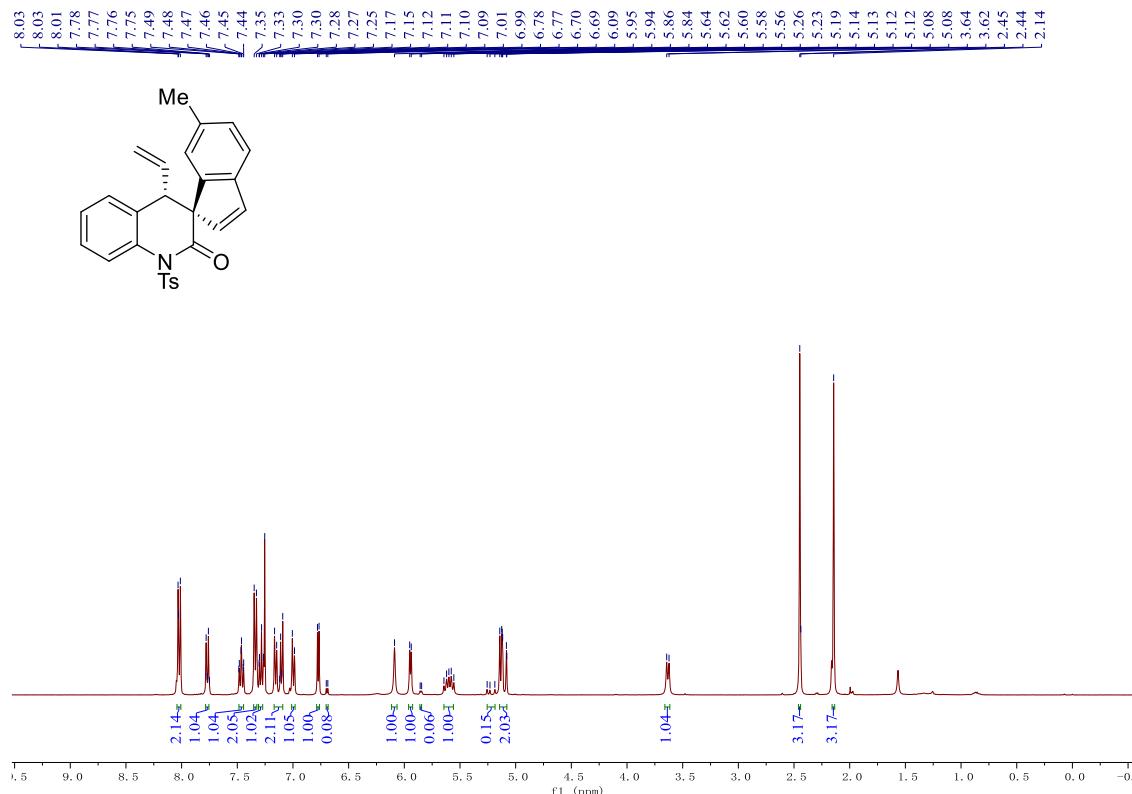
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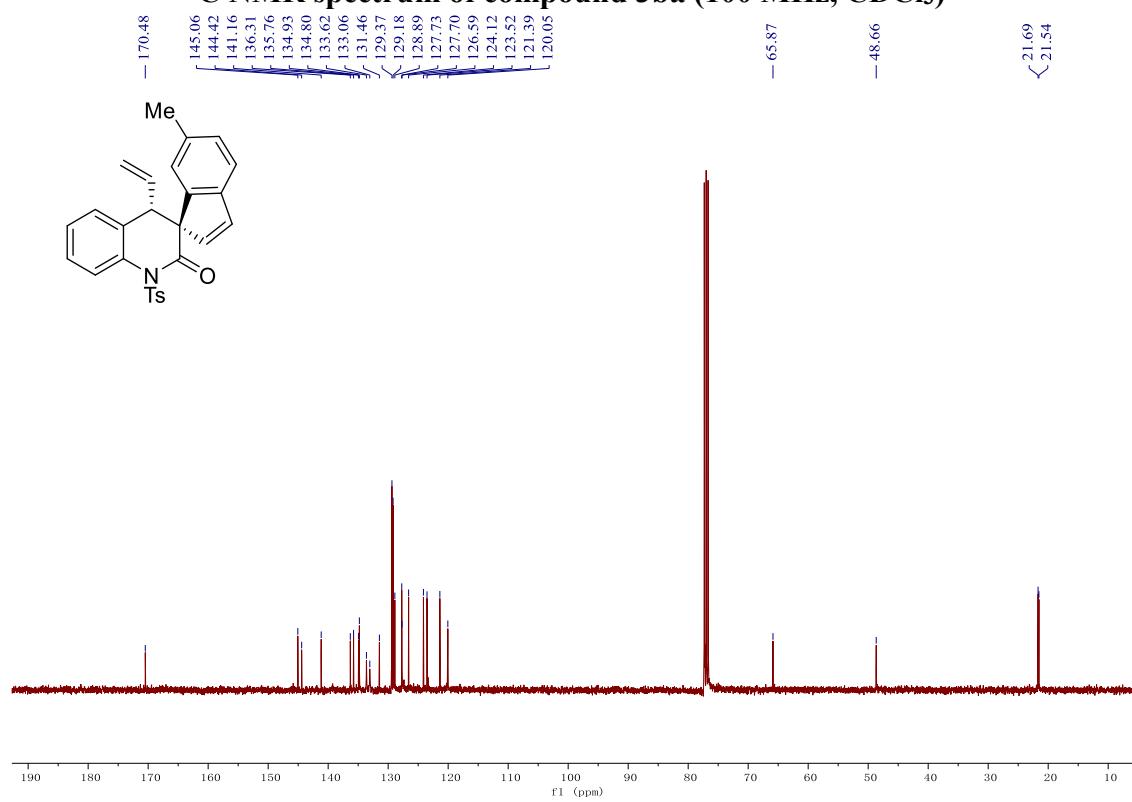
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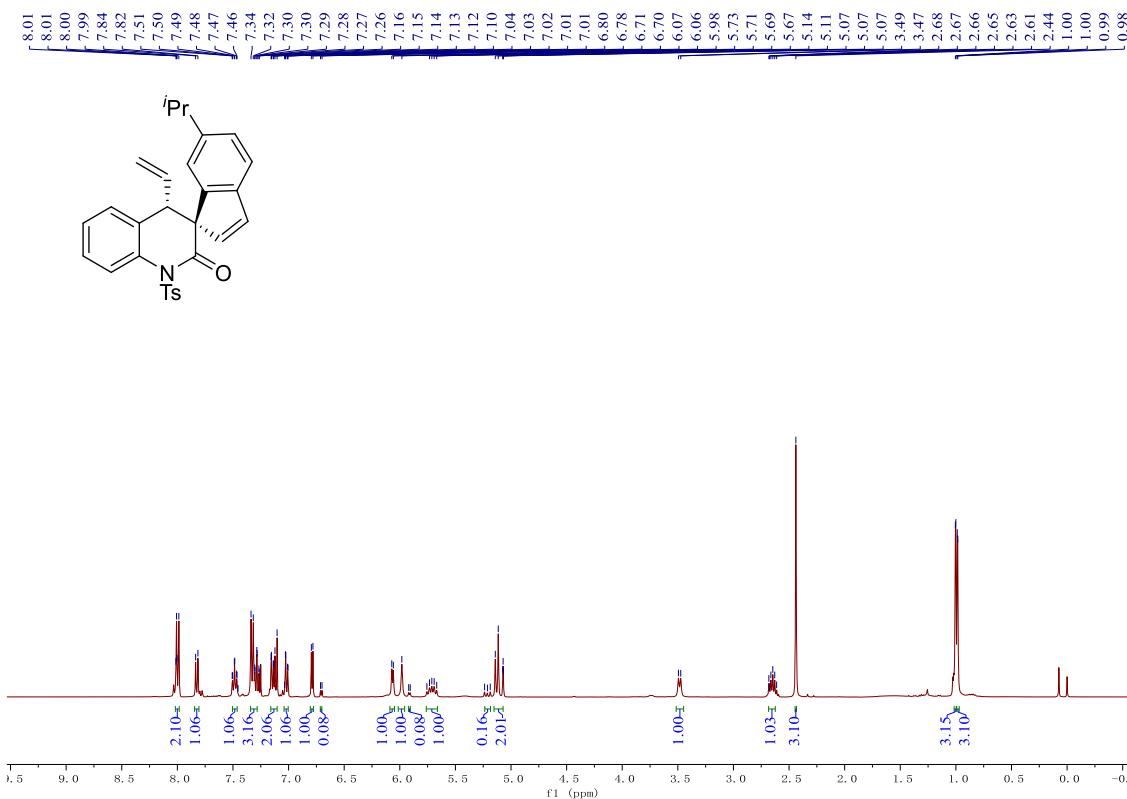
¹H NMR spectrum of compound 3ba (400 MHz, CDCl₃)



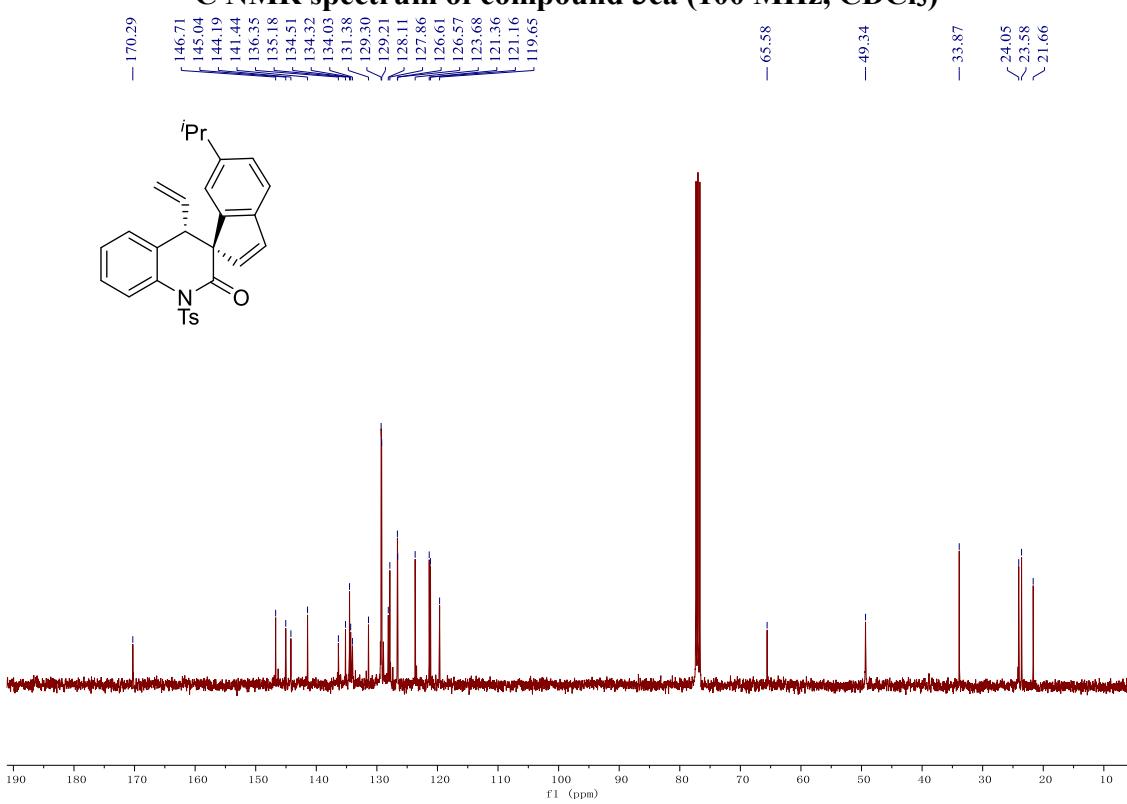
¹³C NMR spectrum of compound 3ba (100 MHz, CDCl₃)



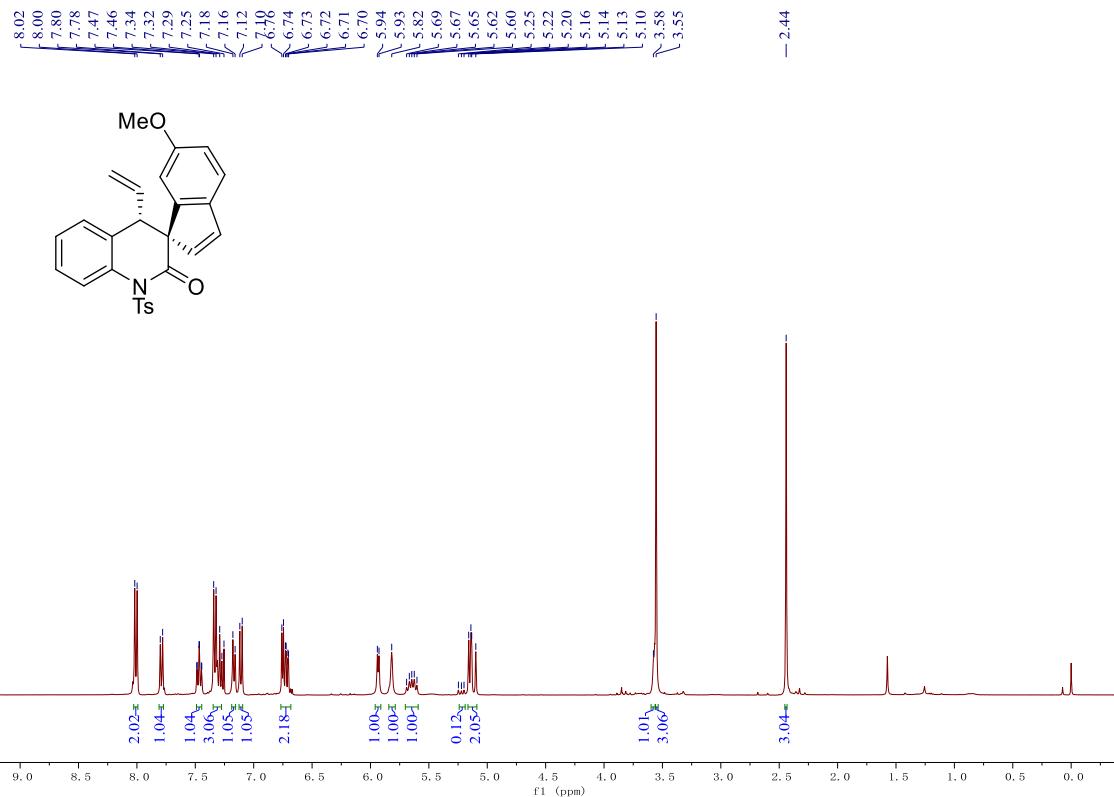
¹H NMR spectrum of compound 3ca (400 MHz, CDCl₃)



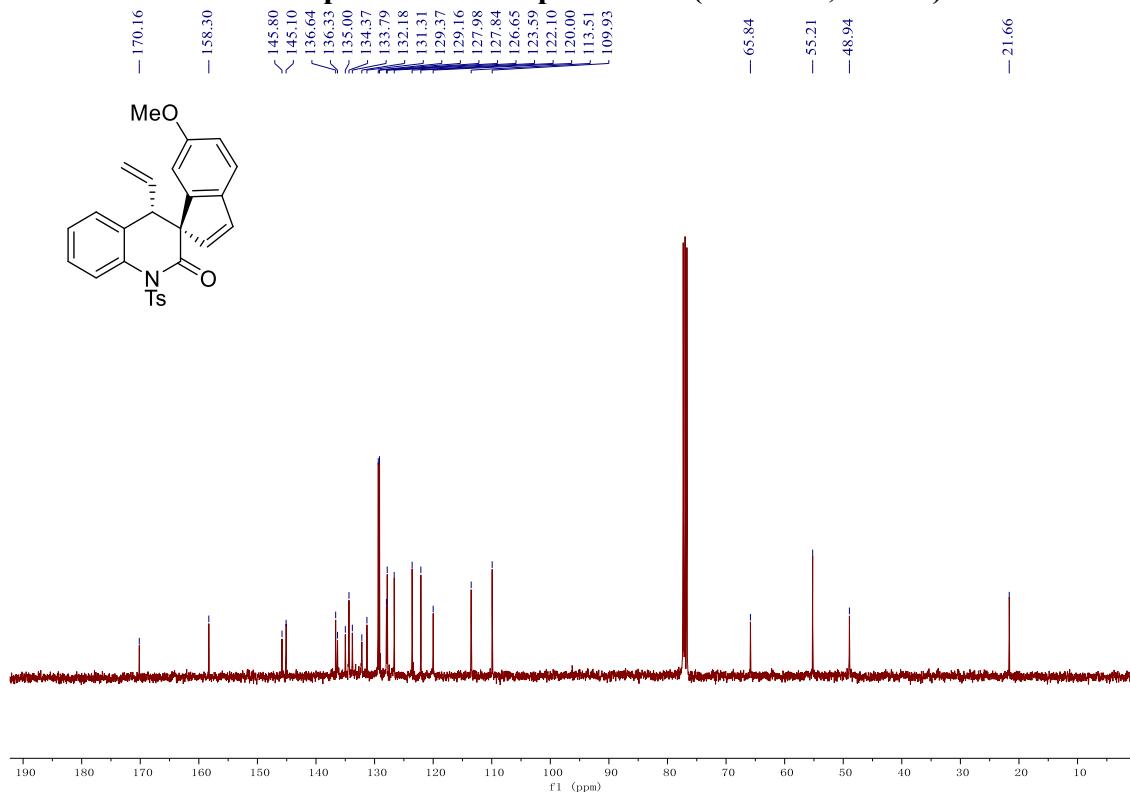
¹³C NMR spectrum of compound 3ca (100 MHz, CDCl₃)

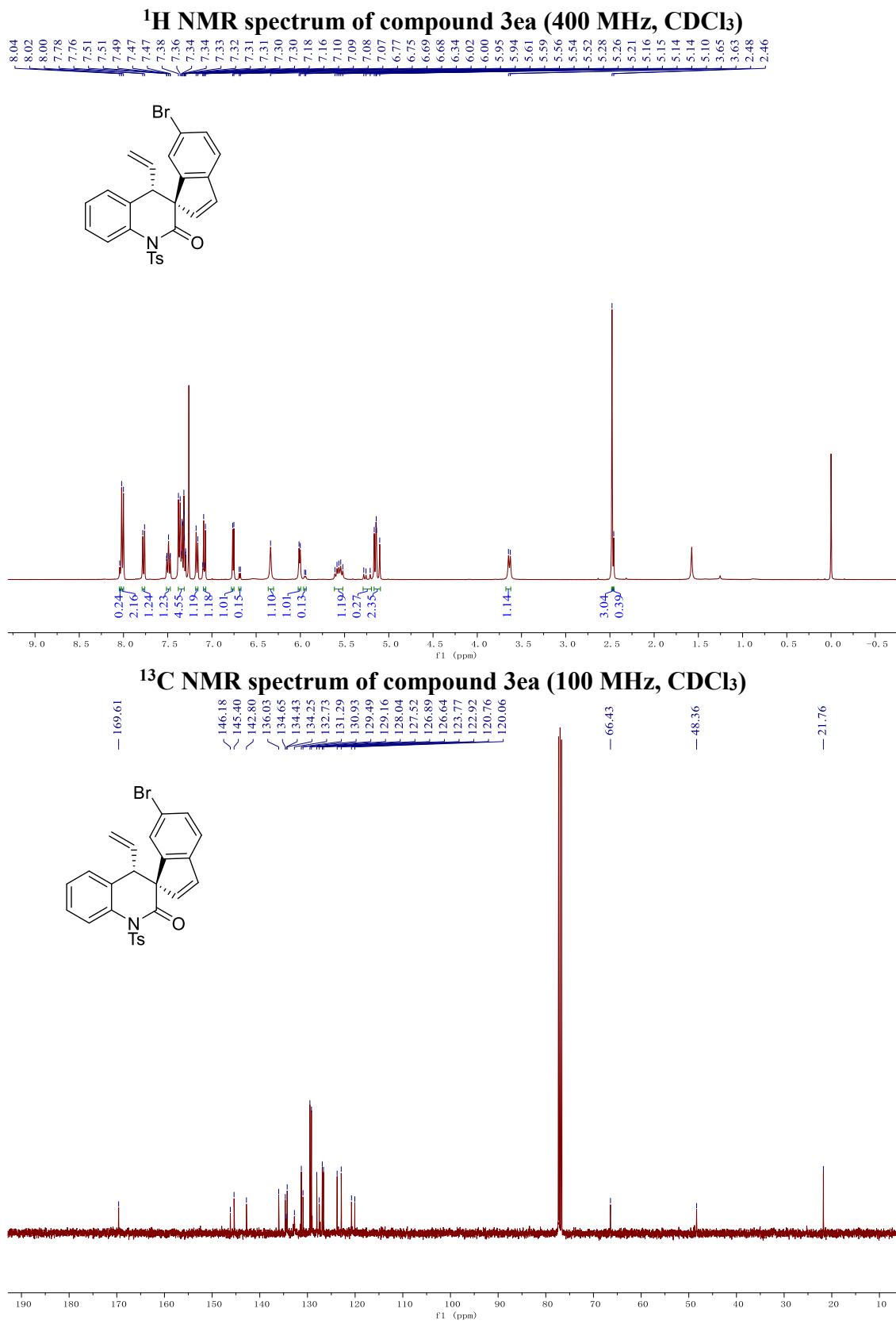


¹H NMR spectrum of compound 3da (400 MHz, CDCl₃)

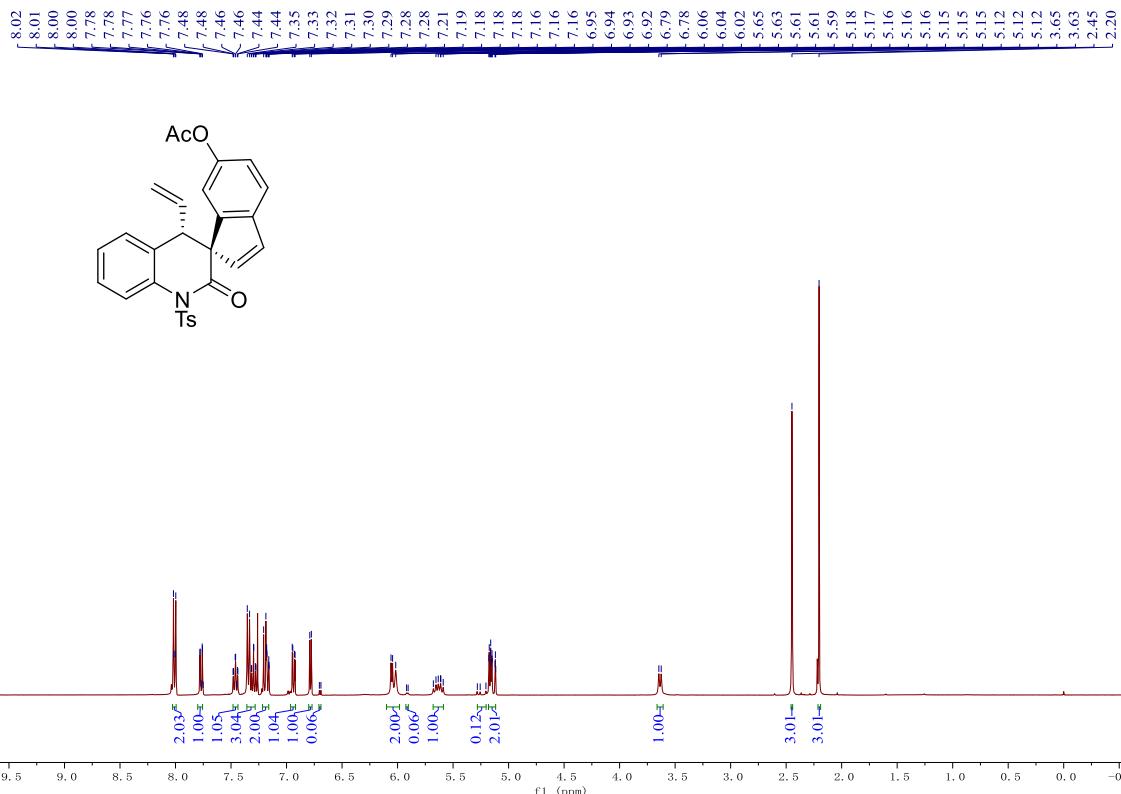


¹³C NMR spectrum of compound 3da (100 MHz, CDCl₃)

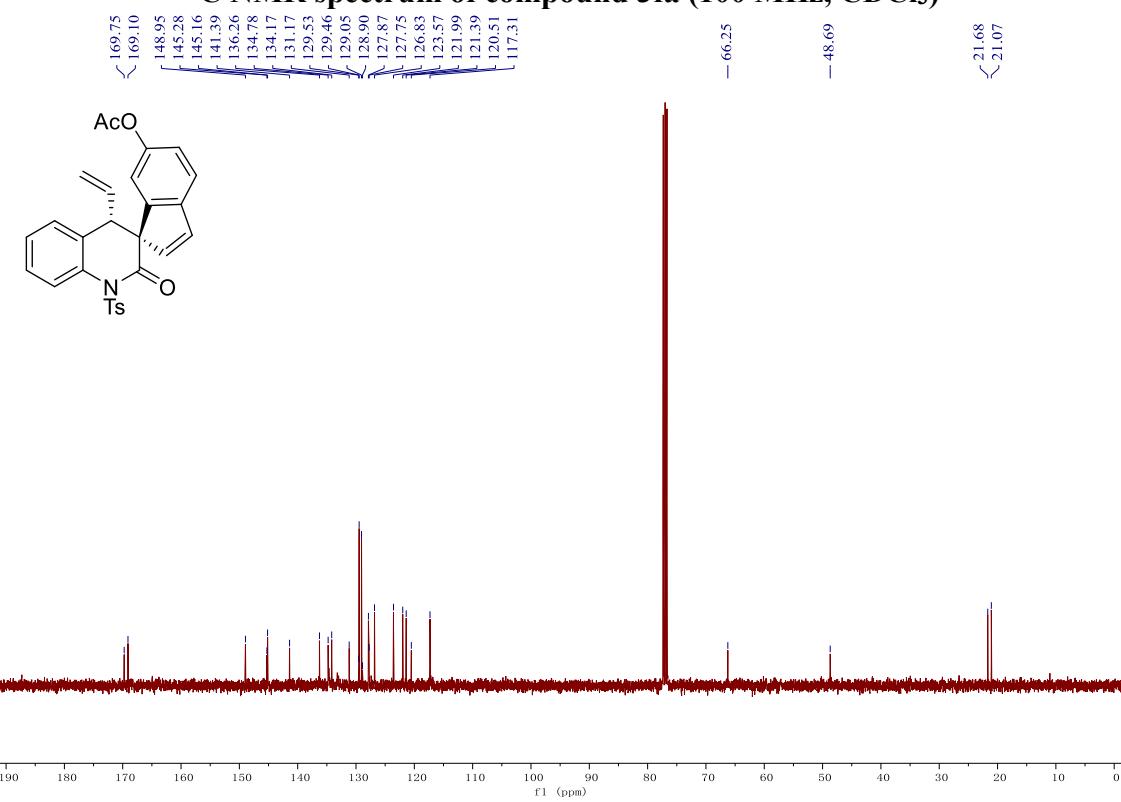




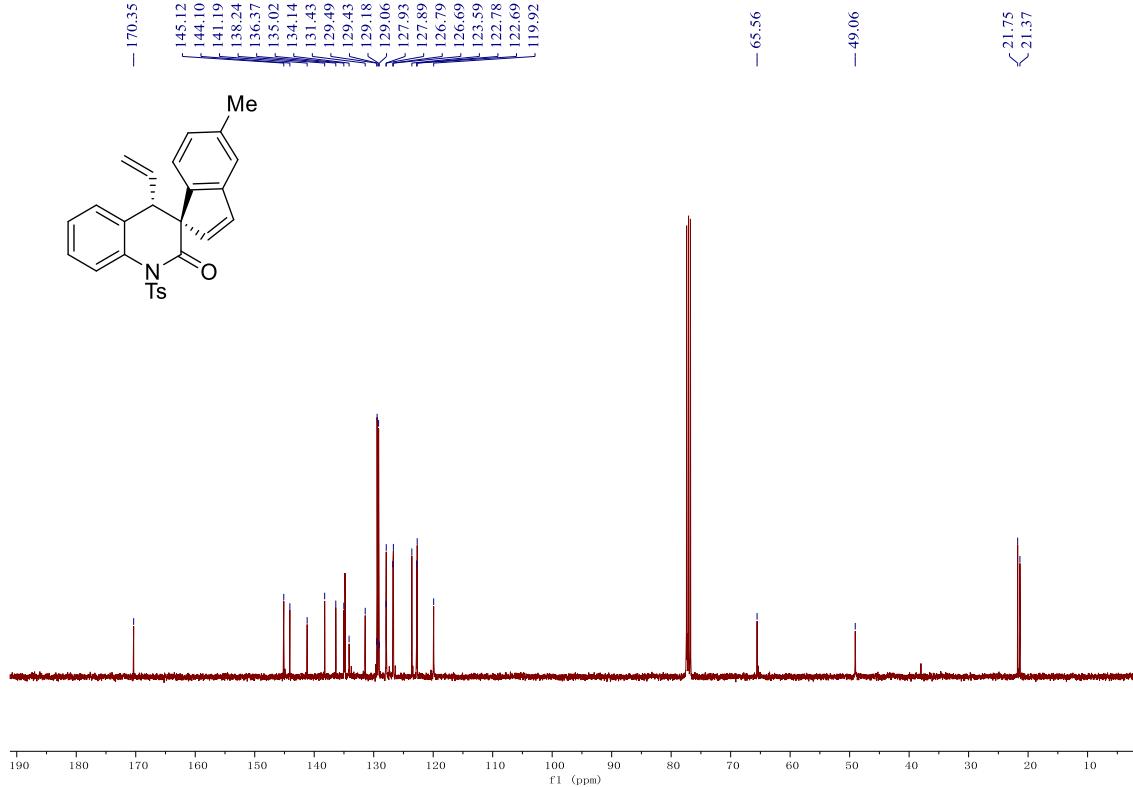
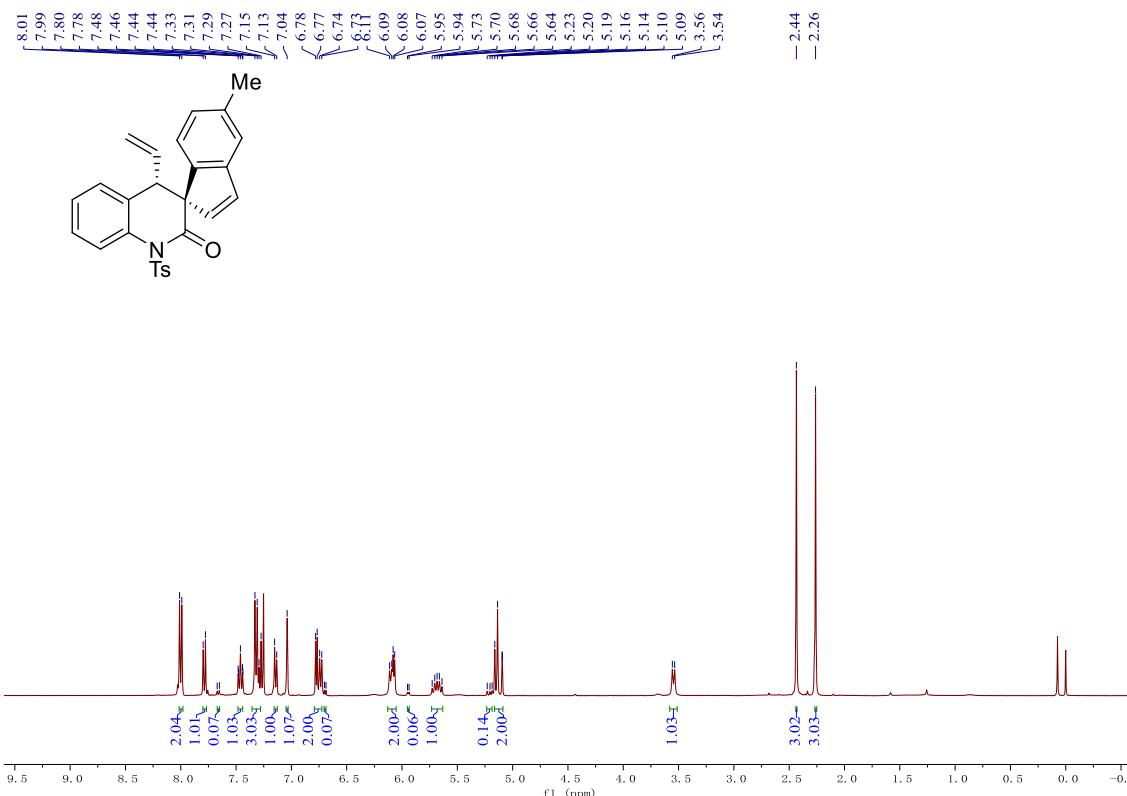
¹H NMR spectrum of compound 3fa (400 MHz, CDCl₃)

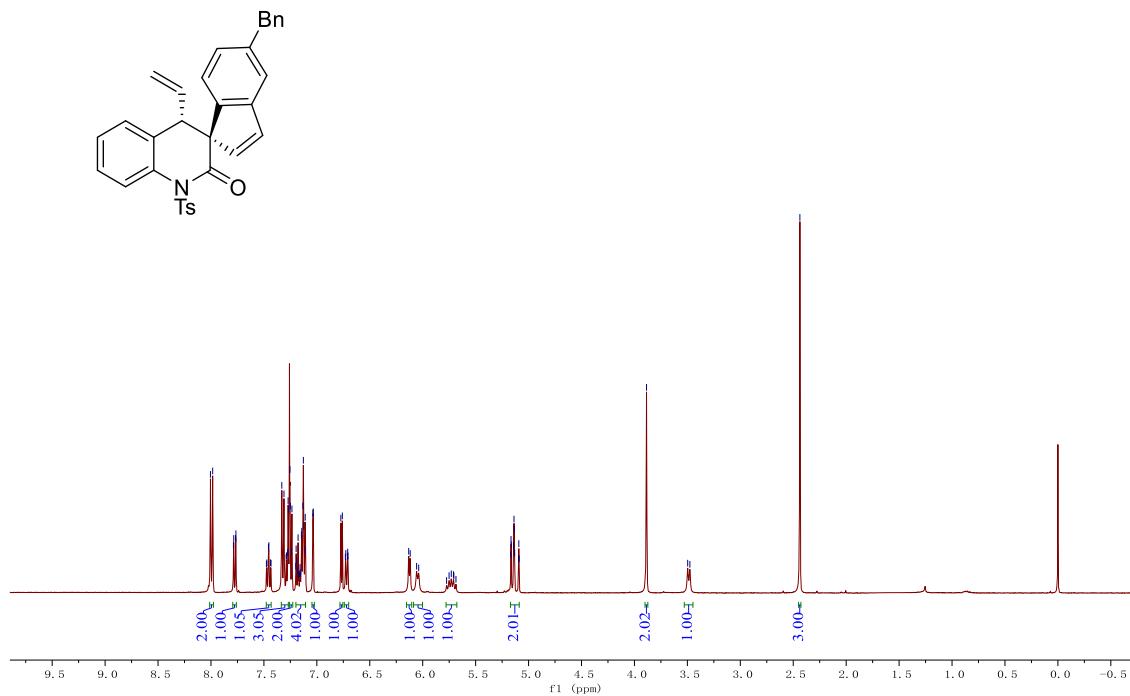


¹³C NMR spectrum of compound 3fa (100 MHz, CDCl₃)

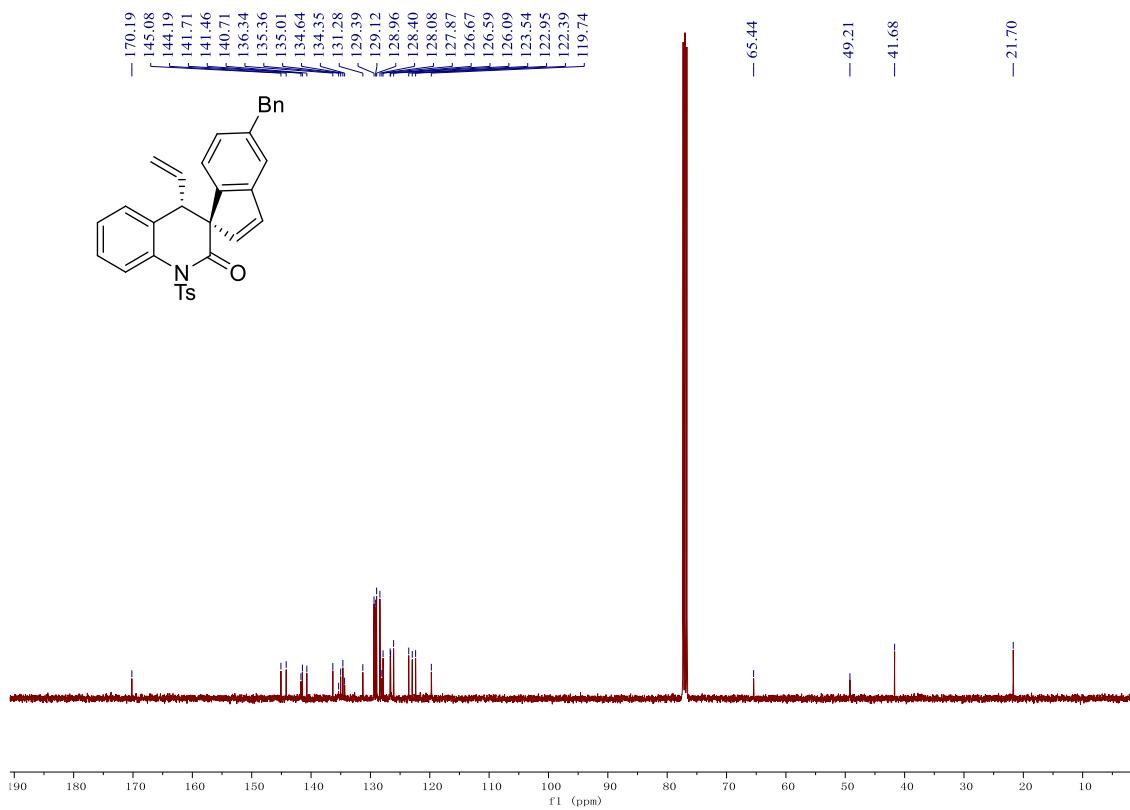


¹H NMR spectrum of compound 3ga (400 MHz, CDCl₃)

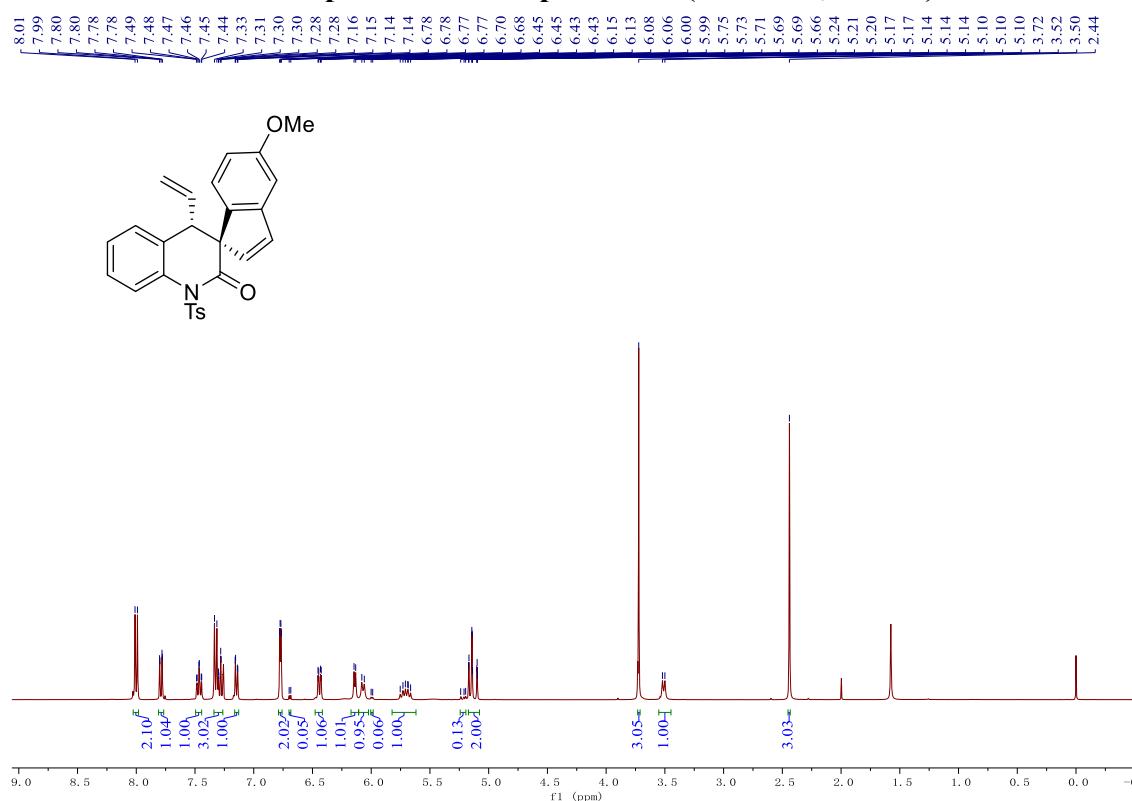




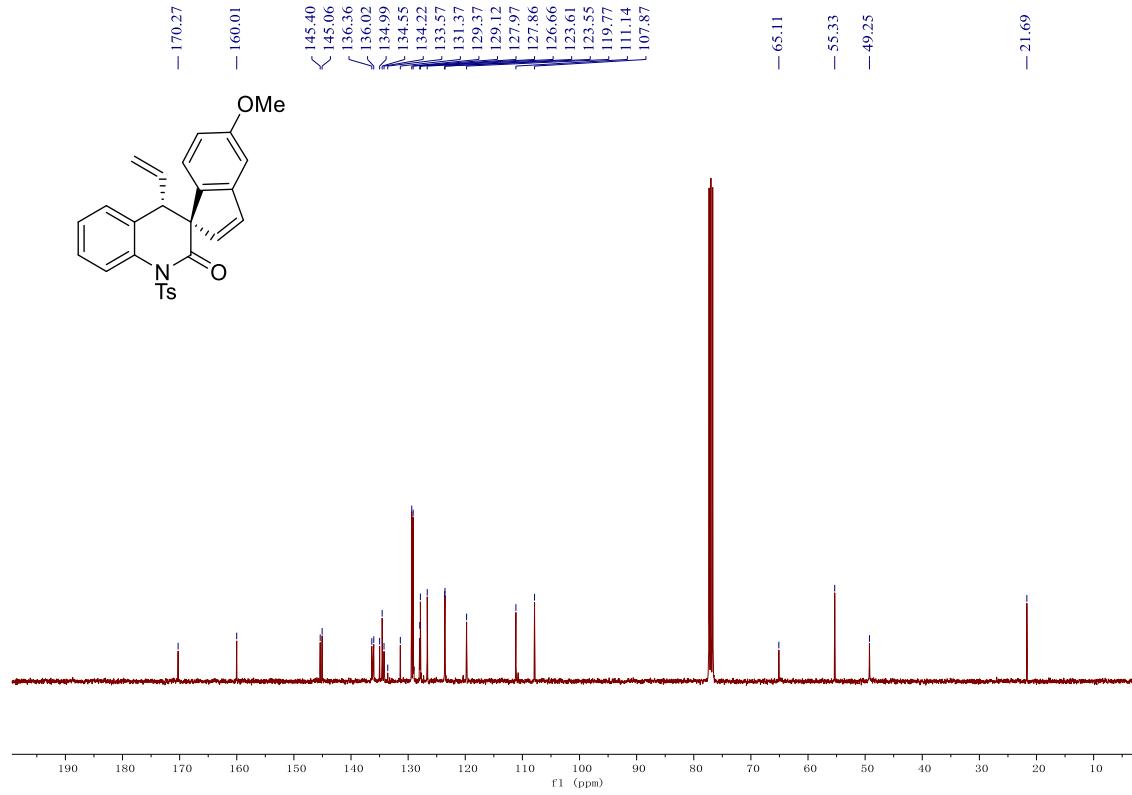
¹³C NMR spectrum of compound 3ma (100 MHz, CDCl₃)



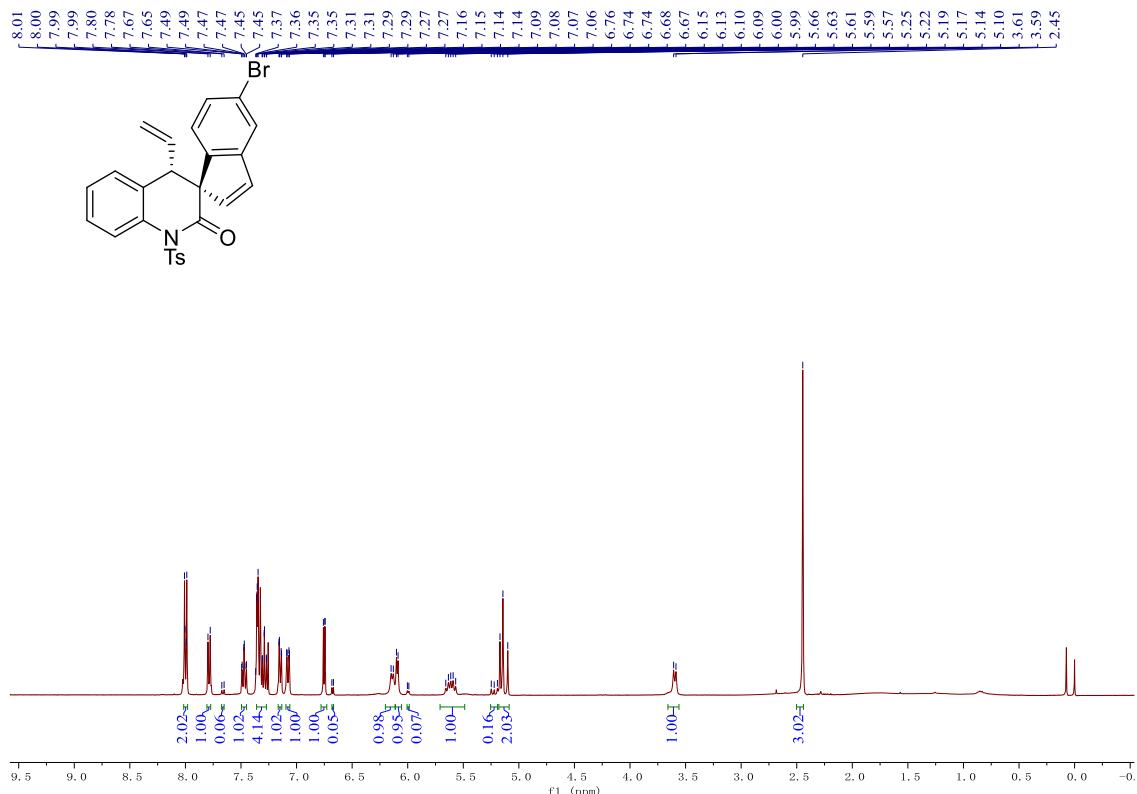
¹H NMR spectrum of compound 3ia (400 MHz, CDCl₃)



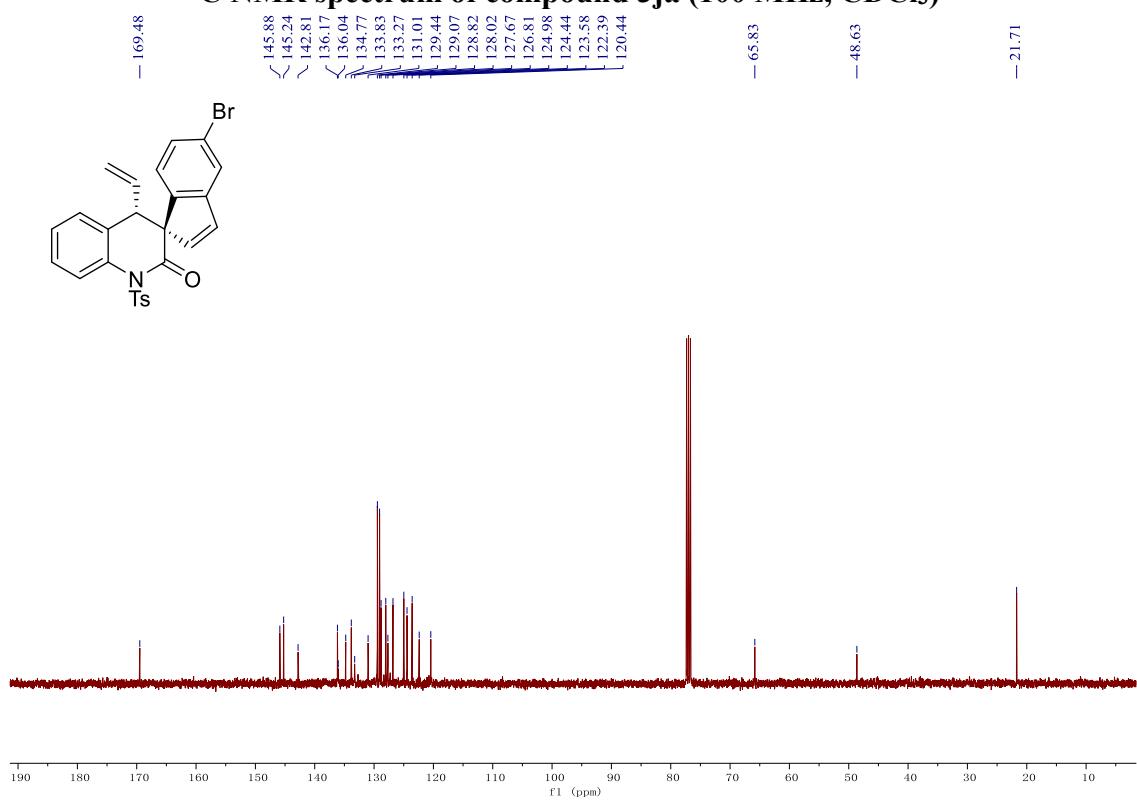
¹³C NMR spectrum of compound 3ia (100 MHz, CDCl₃)



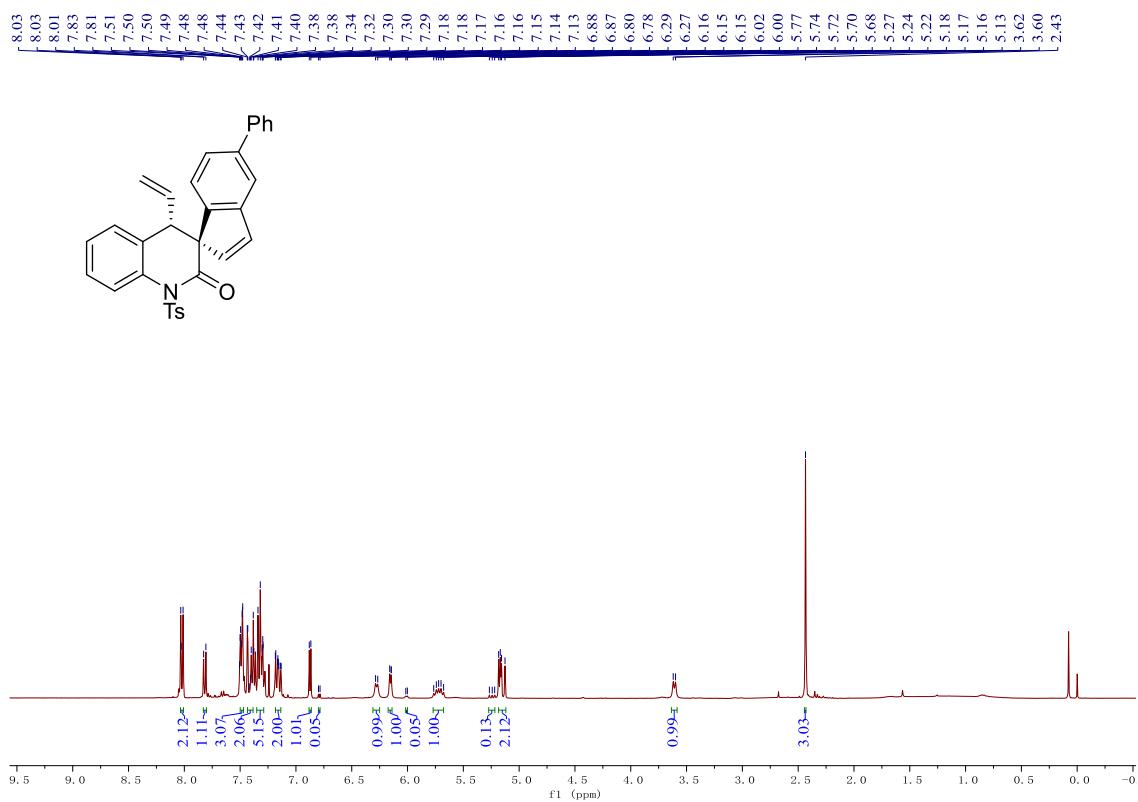
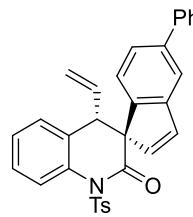
¹H NMR spectrum of compound 3ja (400 MHz, CDCl₃)



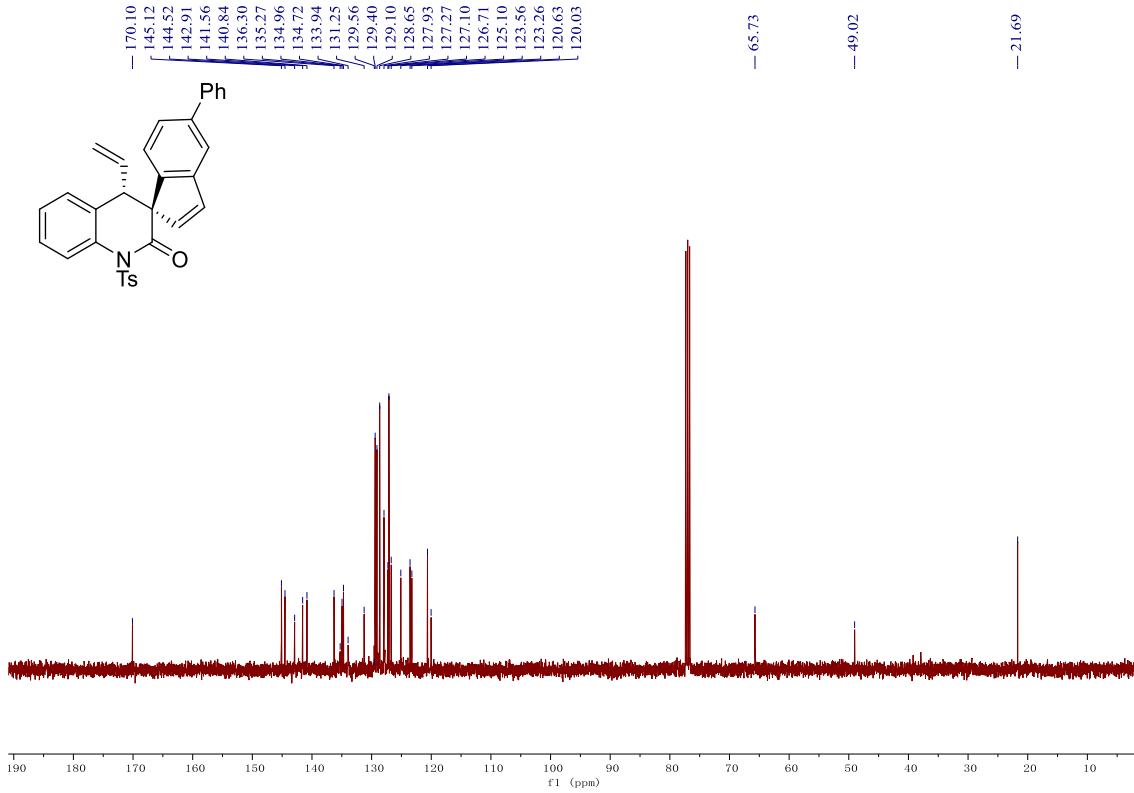
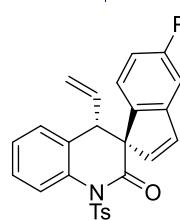
¹³C NMR spectrum of compound 3ja (100 MHz, CDCl₃)



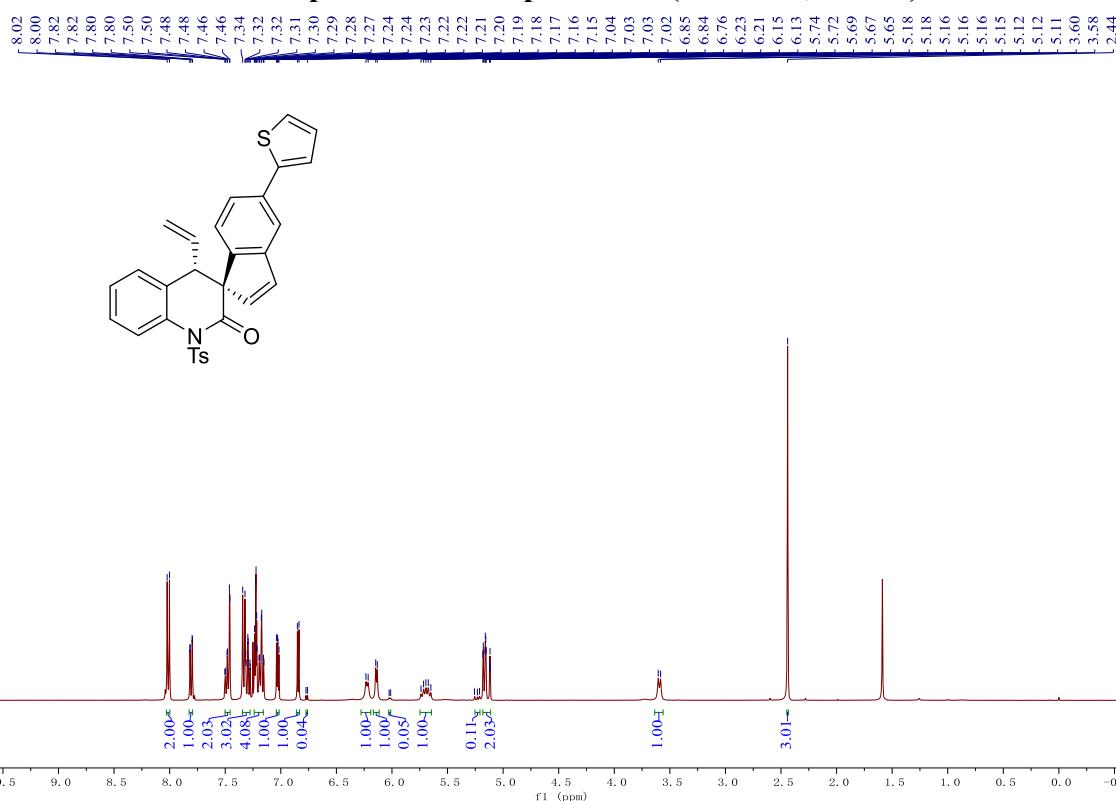
¹H NMR spectrum of compound 3ka (400 MHz, CDCl₃)



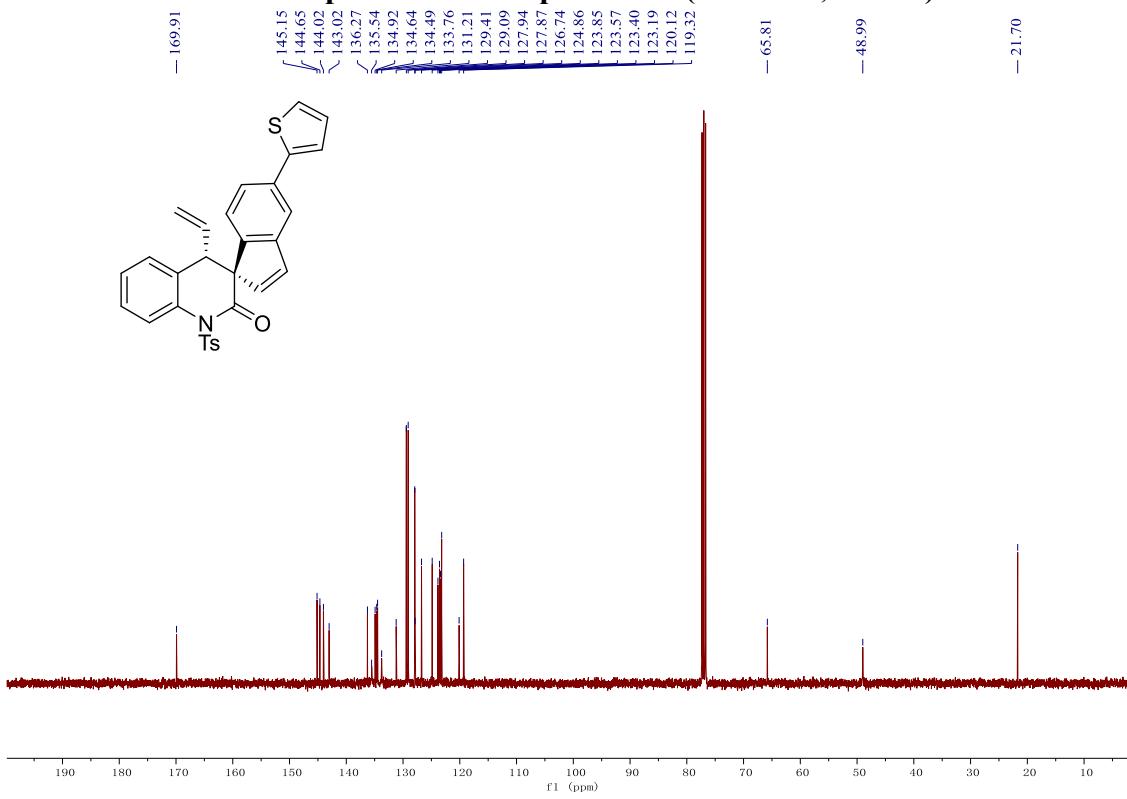
¹³C NMR spectrum of compound 3ka (100 MHz, CDCl₃)

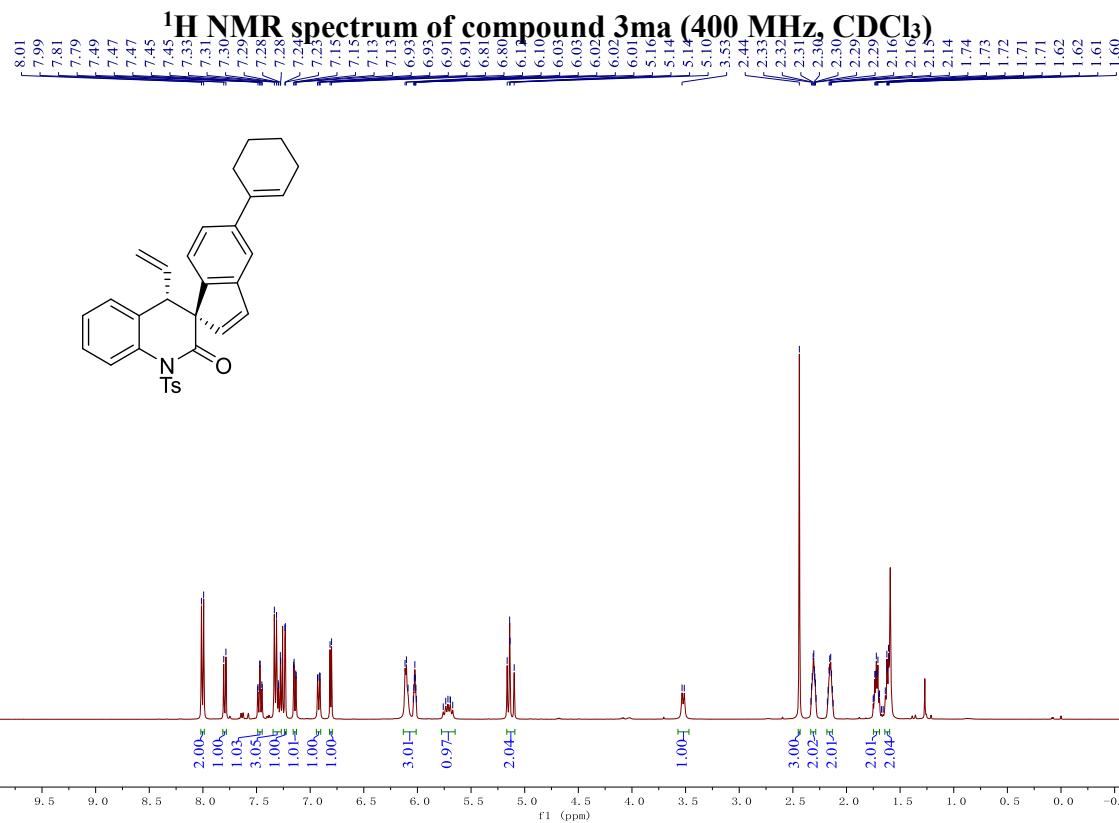


¹H NMR spectrum of compound 3la (400 MHz, CDCl₃)

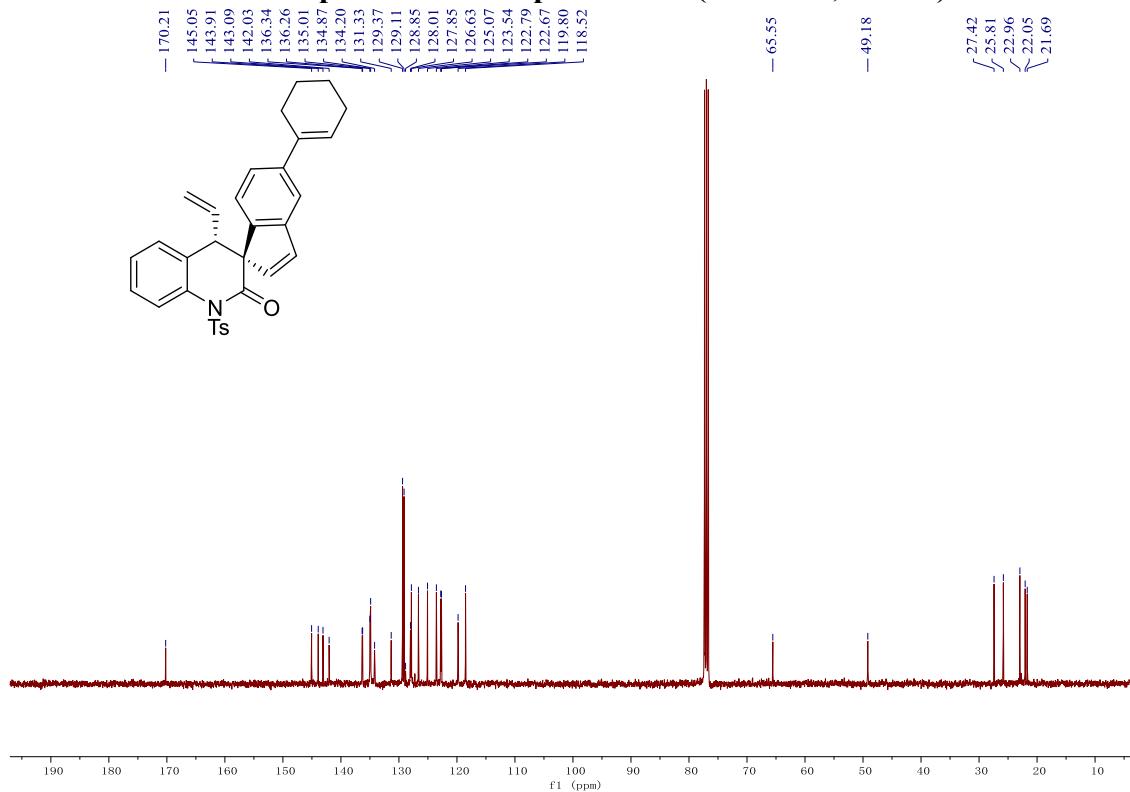
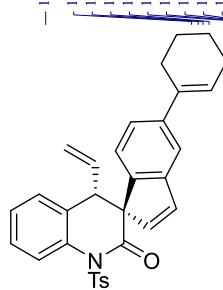


¹³C NMR spectrum of compound 3la (100 MHz, CDCl₃)

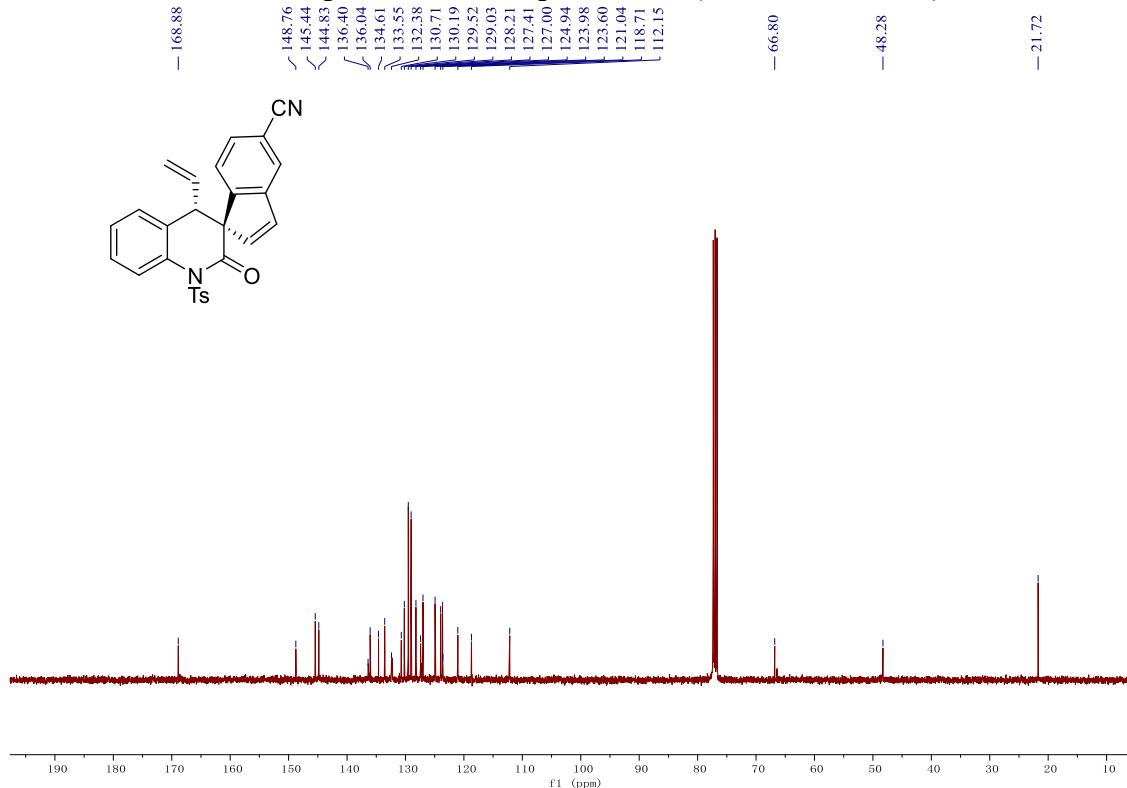
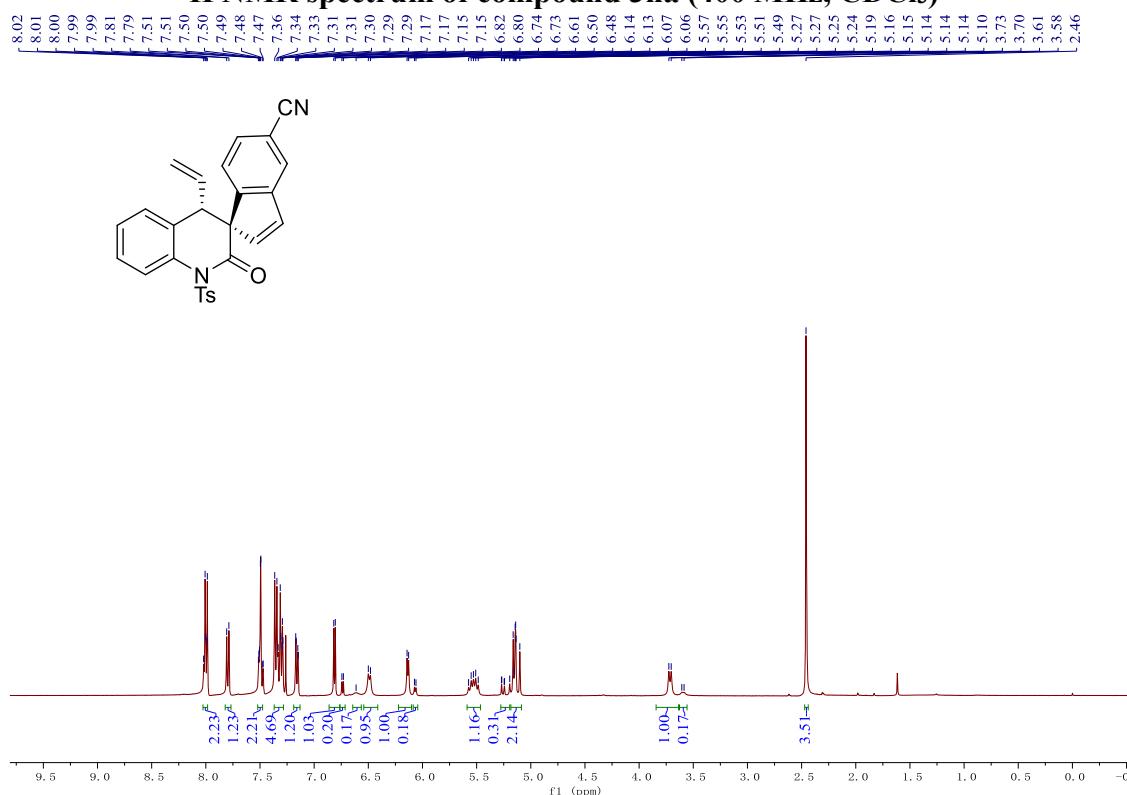




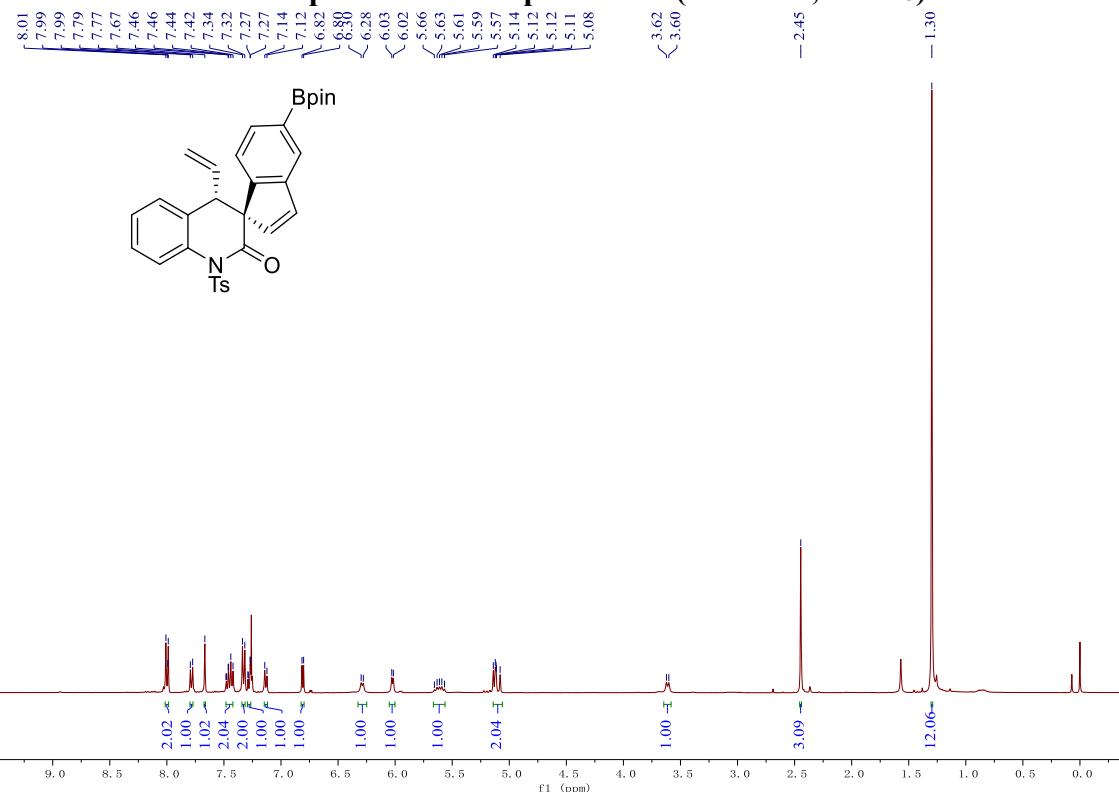
¹³C NMR spectrum of compound 3ma (100 MHz, CDCl₃)



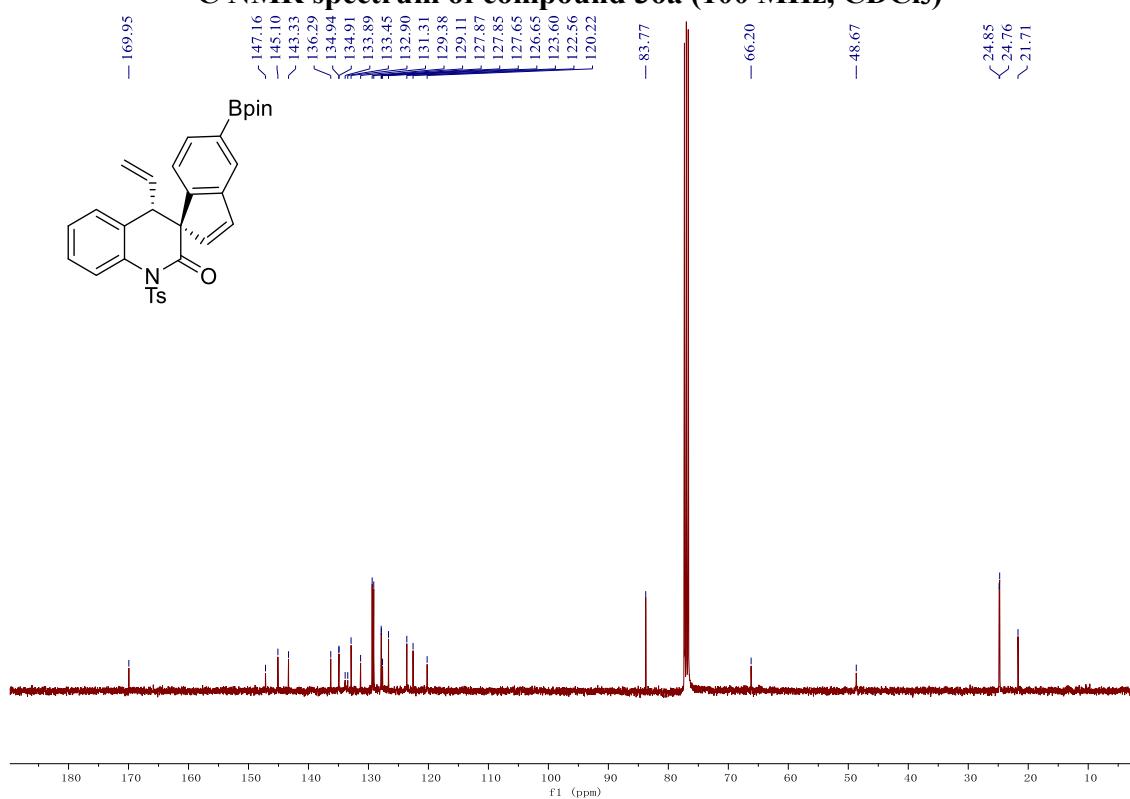
¹H NMR spectrum of compound 3na (400 MHz, CDCl₃)



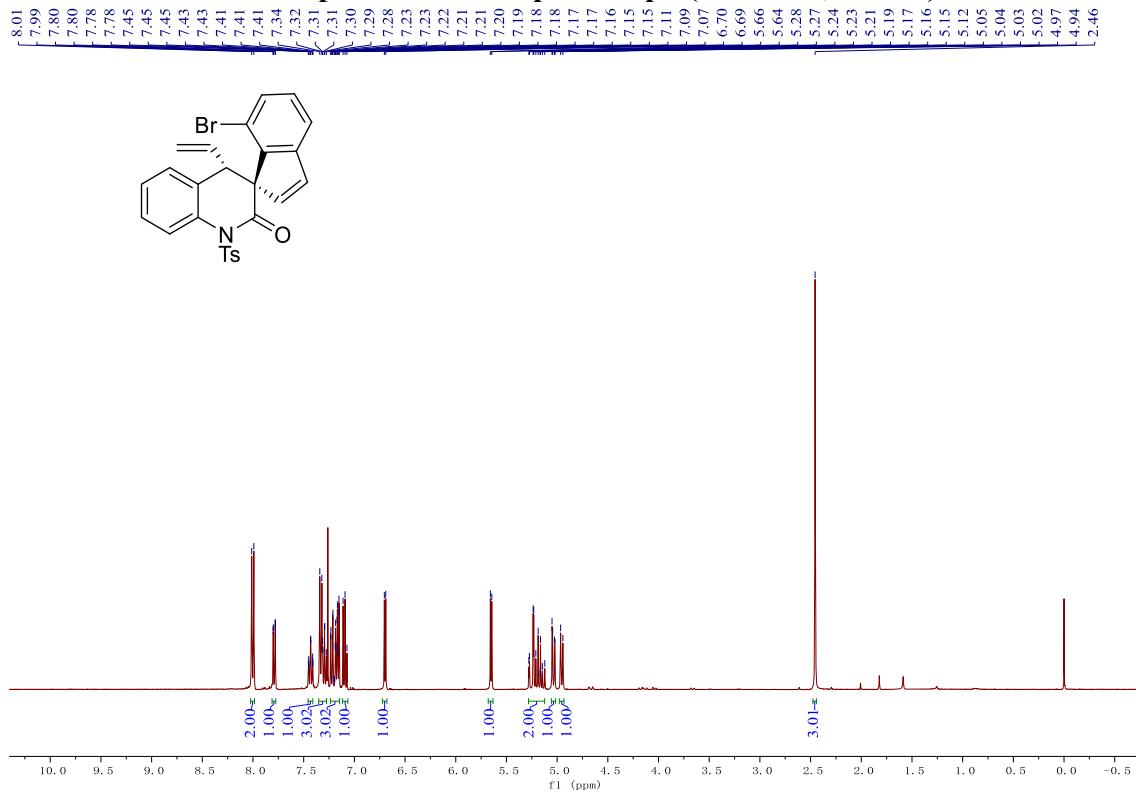
¹H NMR spectrum of compound 3oa (400 MHz, CDCl₃)



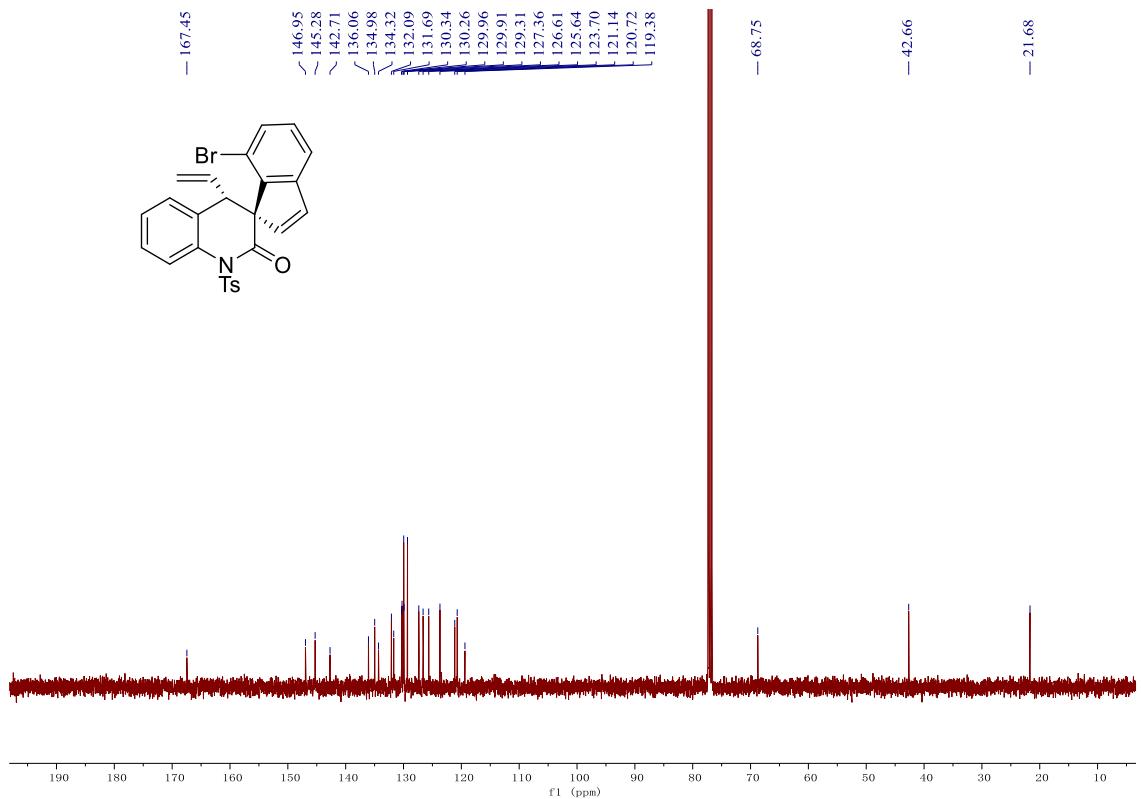
¹³C NMR spectrum of compound 3oa (100 MHz, CDCl₃)



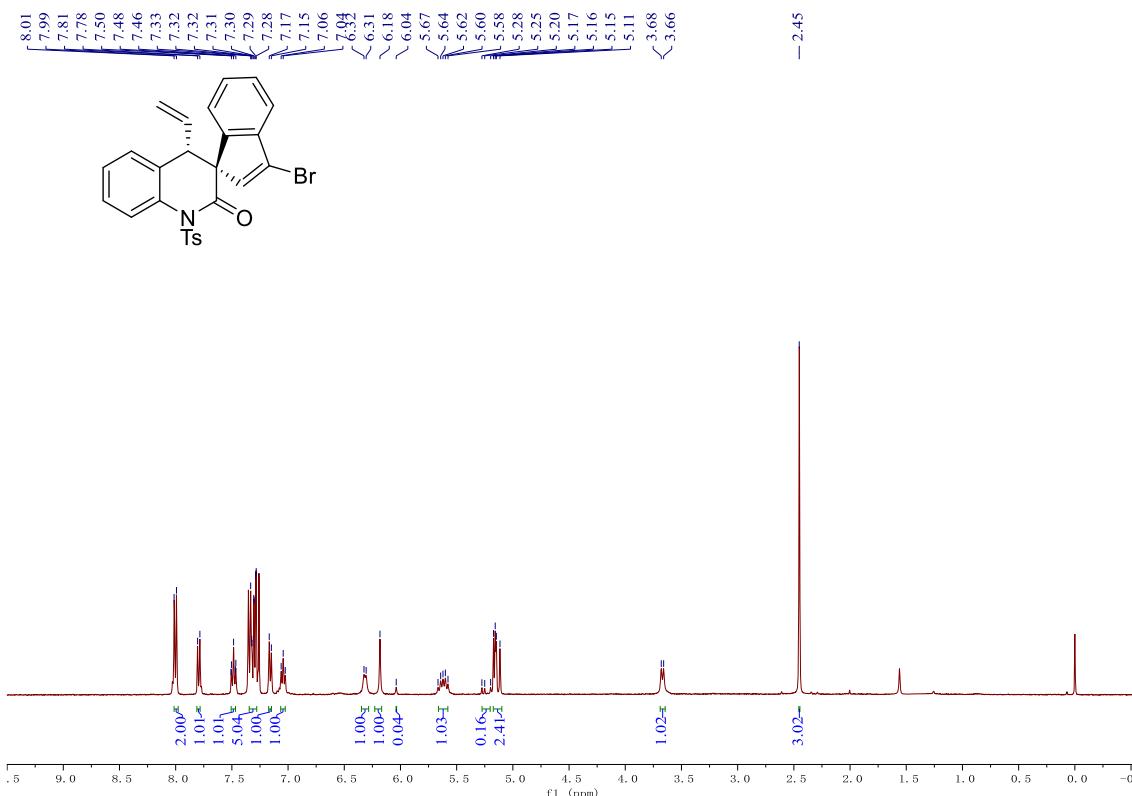
¹H NMR spectrum of compound 3pa (400 MHz, CDCl₃)



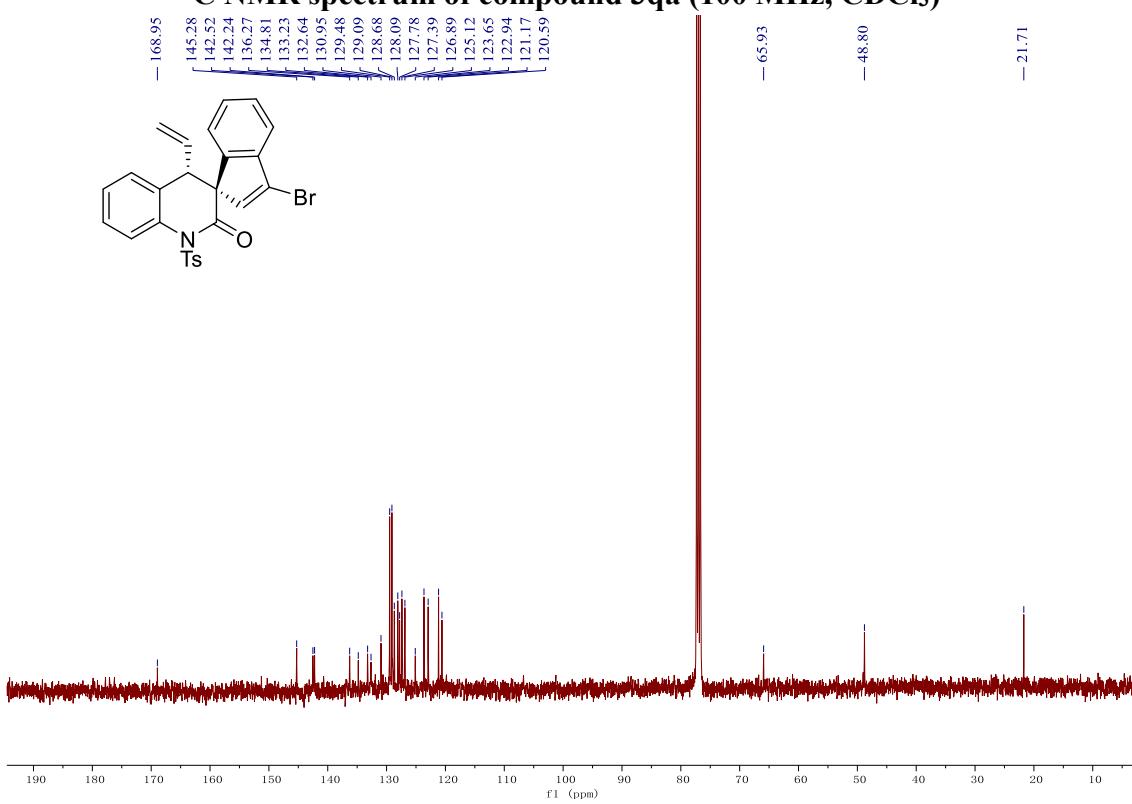
¹³C NMR spectrum of compound 3pa (100 MHz, CDCl₃)



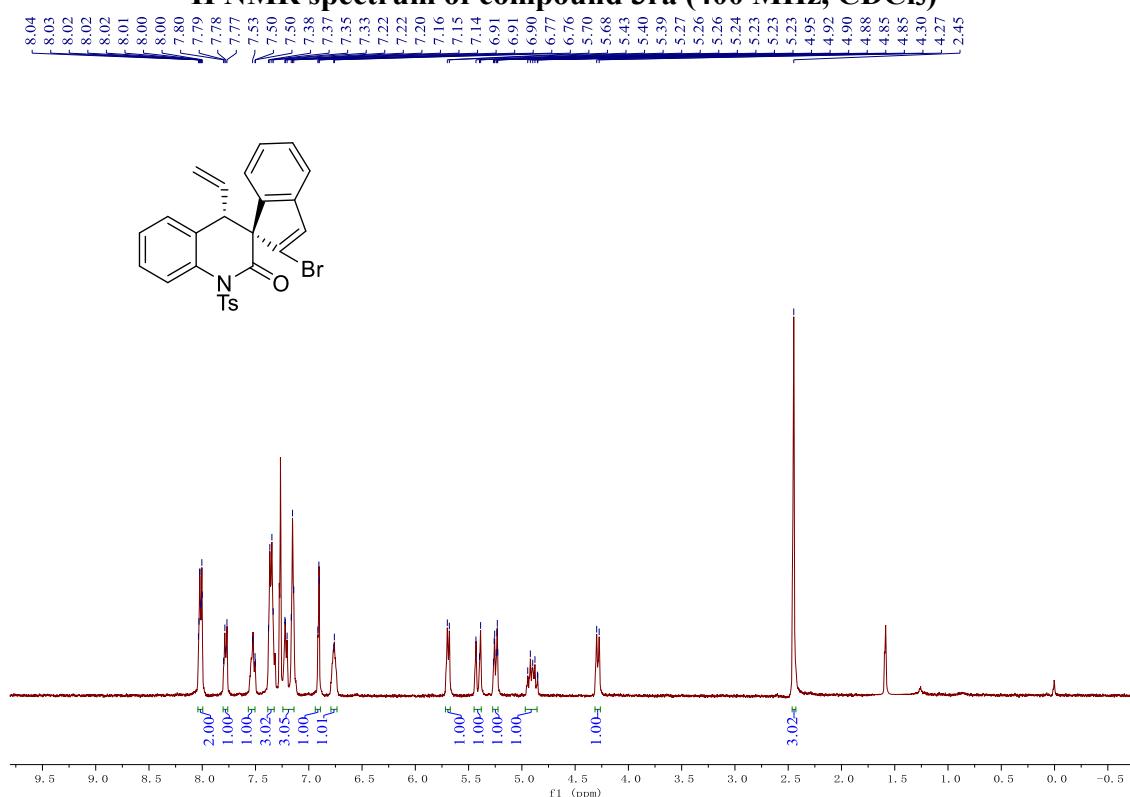
¹H NMR spectrum of compound 3qa (400 MHz, CDCl₃)



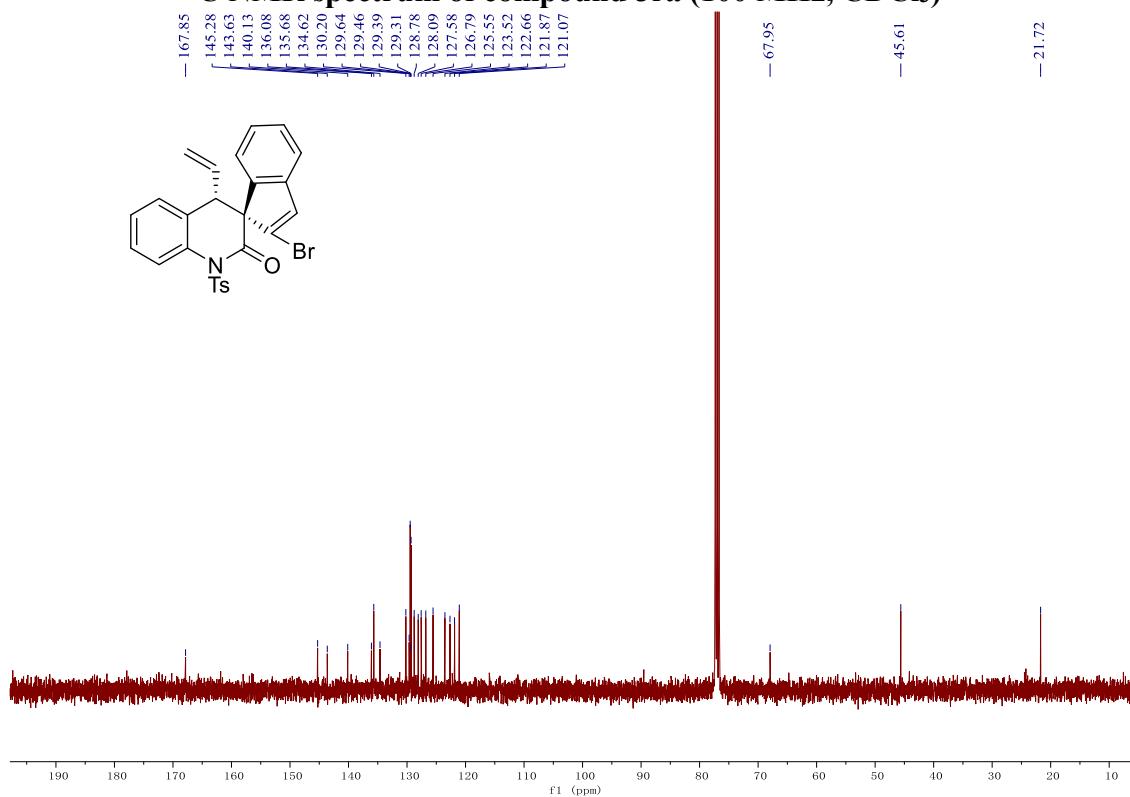
¹³C NMR spectrum of compound 3qa (100 MHz, CDCl₃)



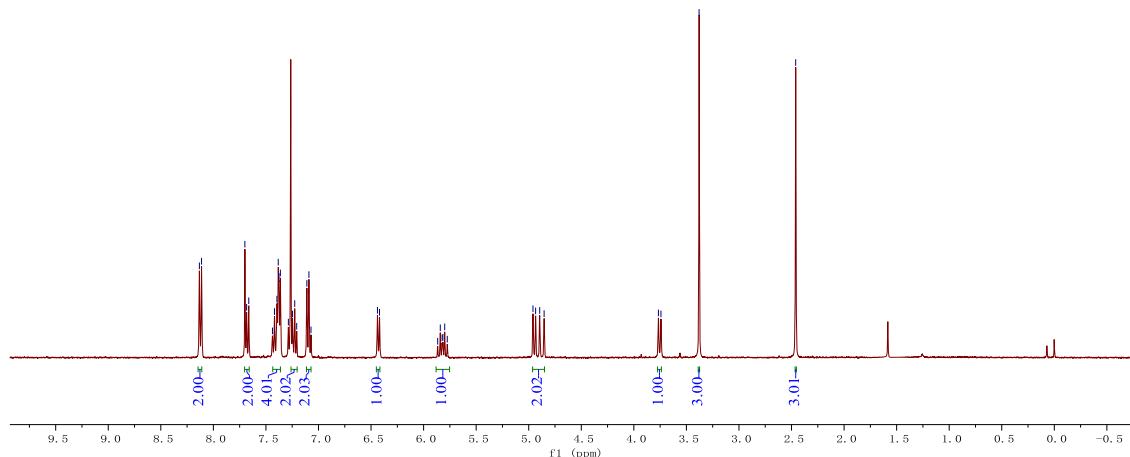
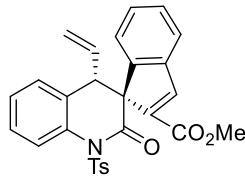
¹H NMR spectrum of compound 3ra (400 MHz, CDCl₃)



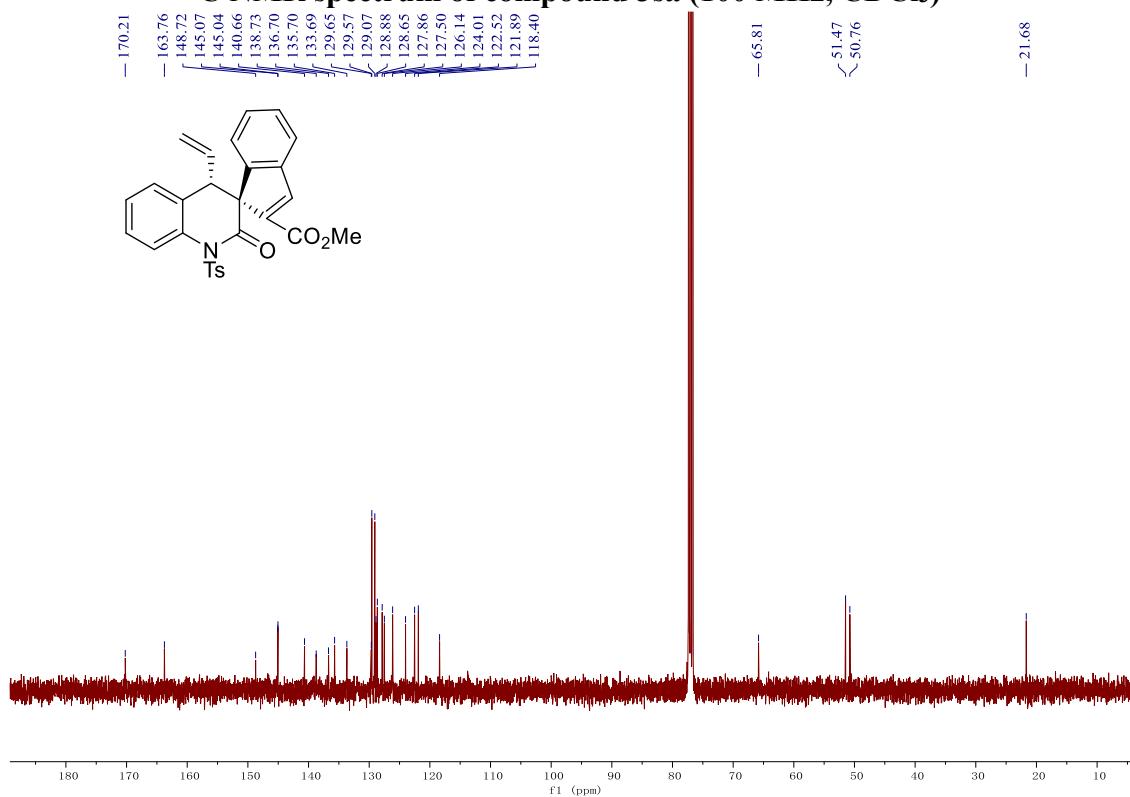
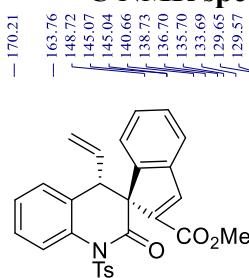
¹³C NMR spectrum of compound 3ra (100 MHz, CDCl₃)



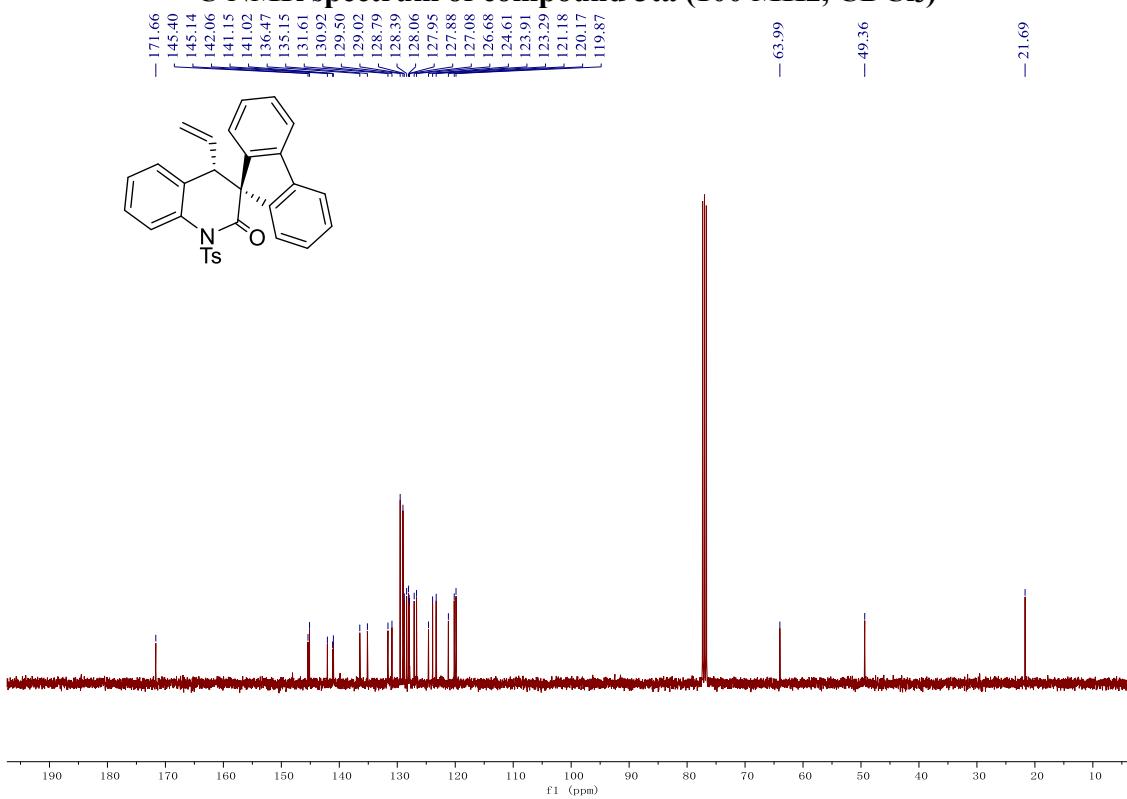
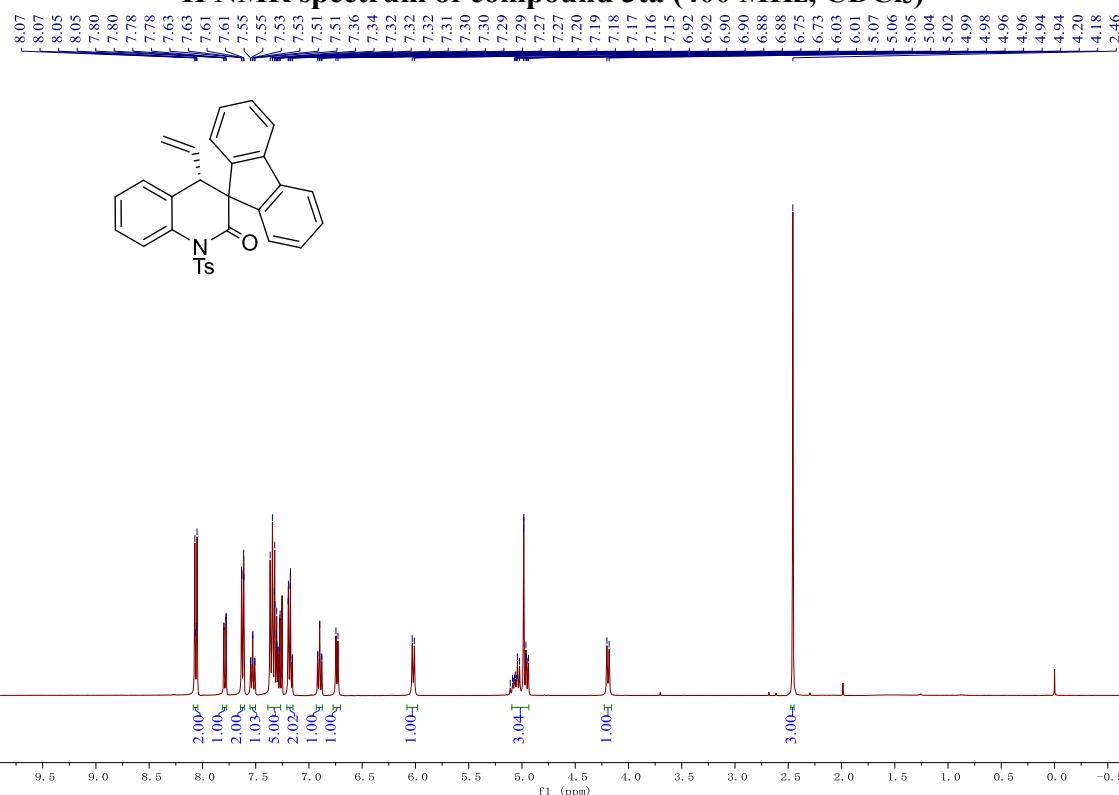
¹H NMR spectrum of compound 3sa (400 MHz, CDCl₃)



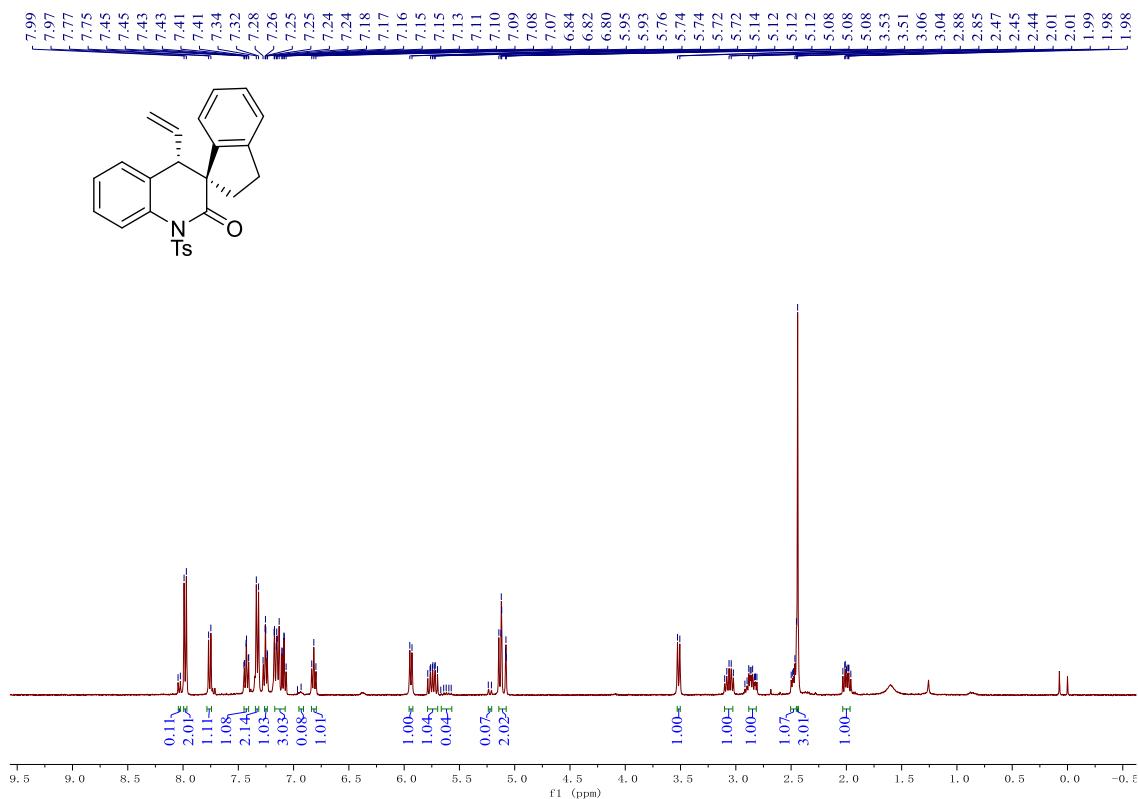
¹³C NMR spectrum of compound 3sa (100 MHz, CDCl₃)



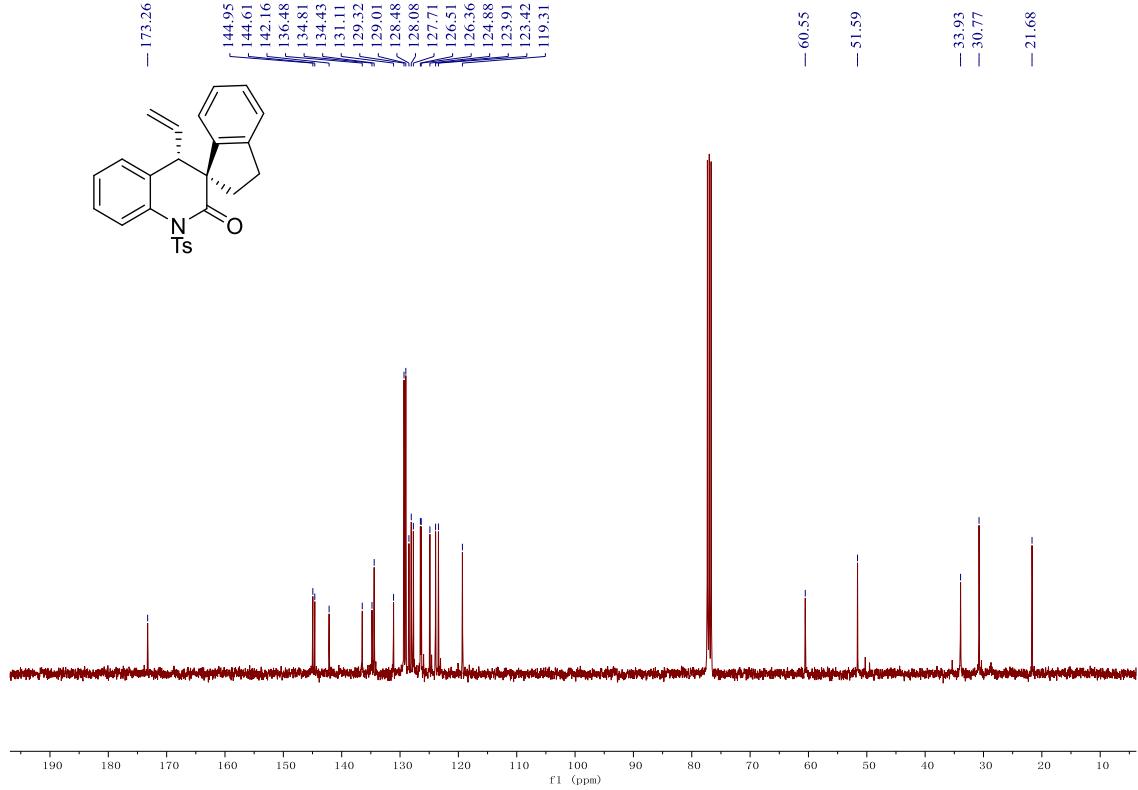
¹H NMR spectrum of compound 3ta (400 MHz, CDCl₃)



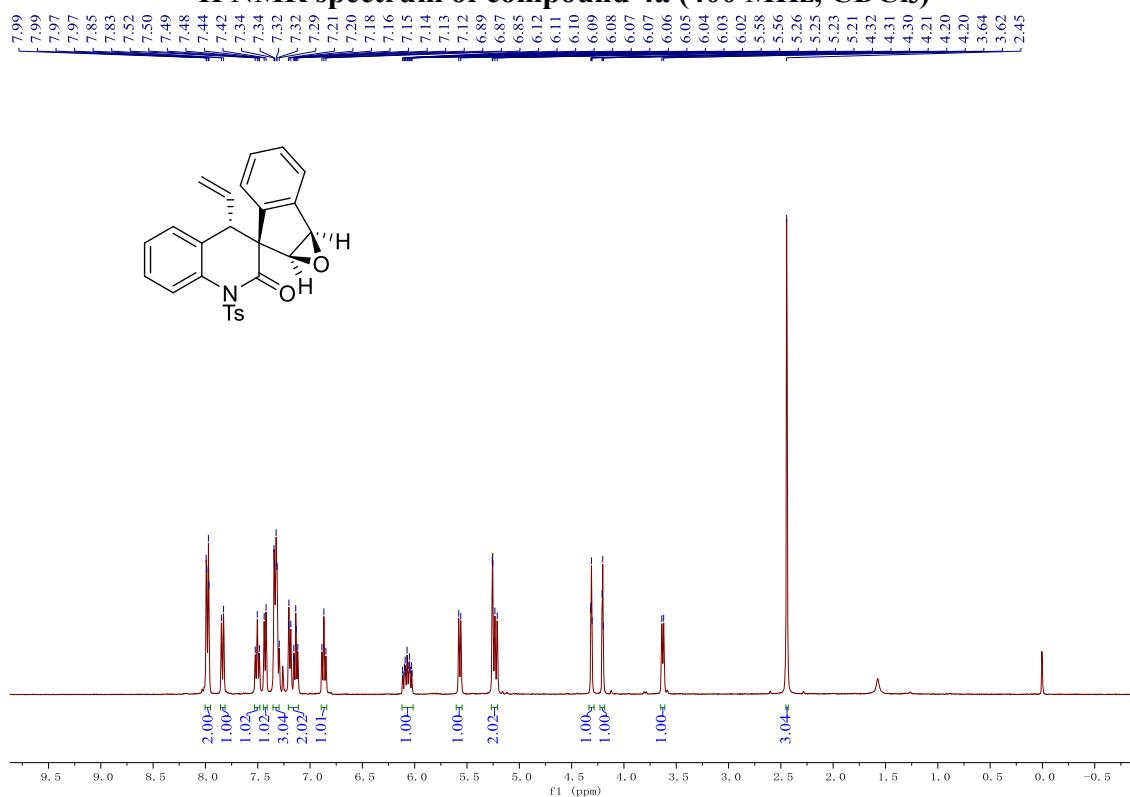
¹H NMR spectrum of compound 3ua (400 MHz, CDCl₃)



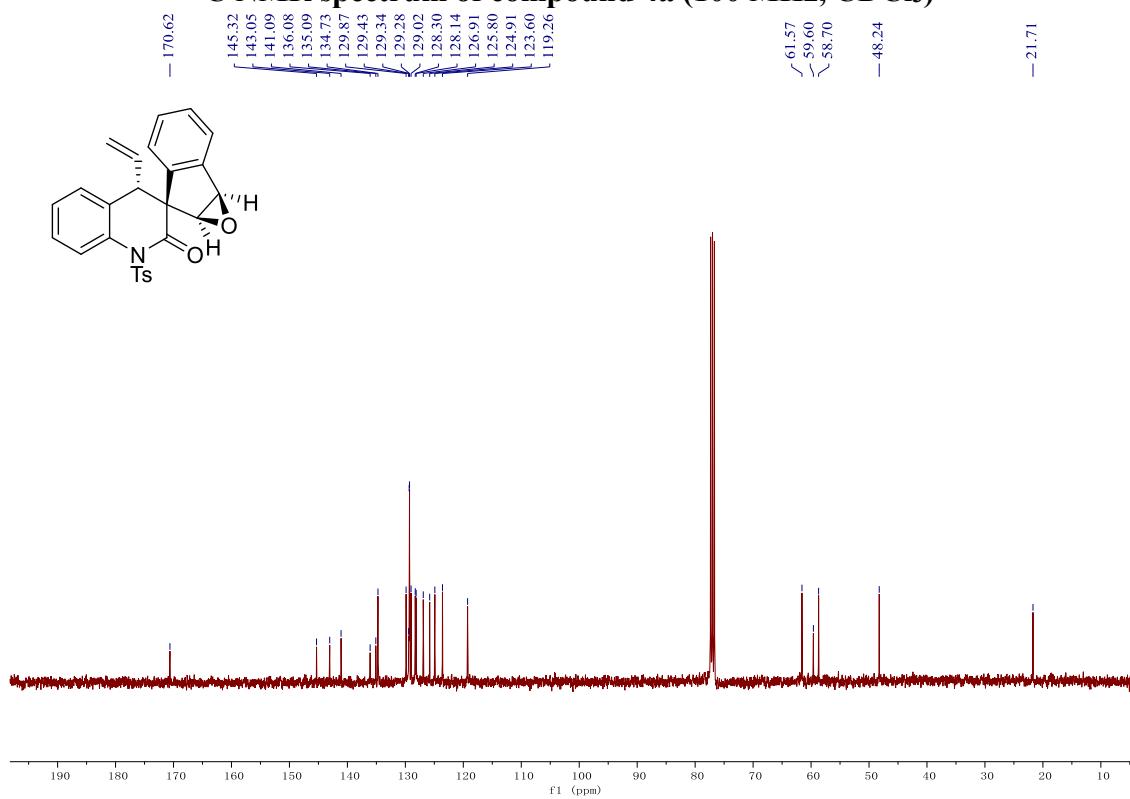
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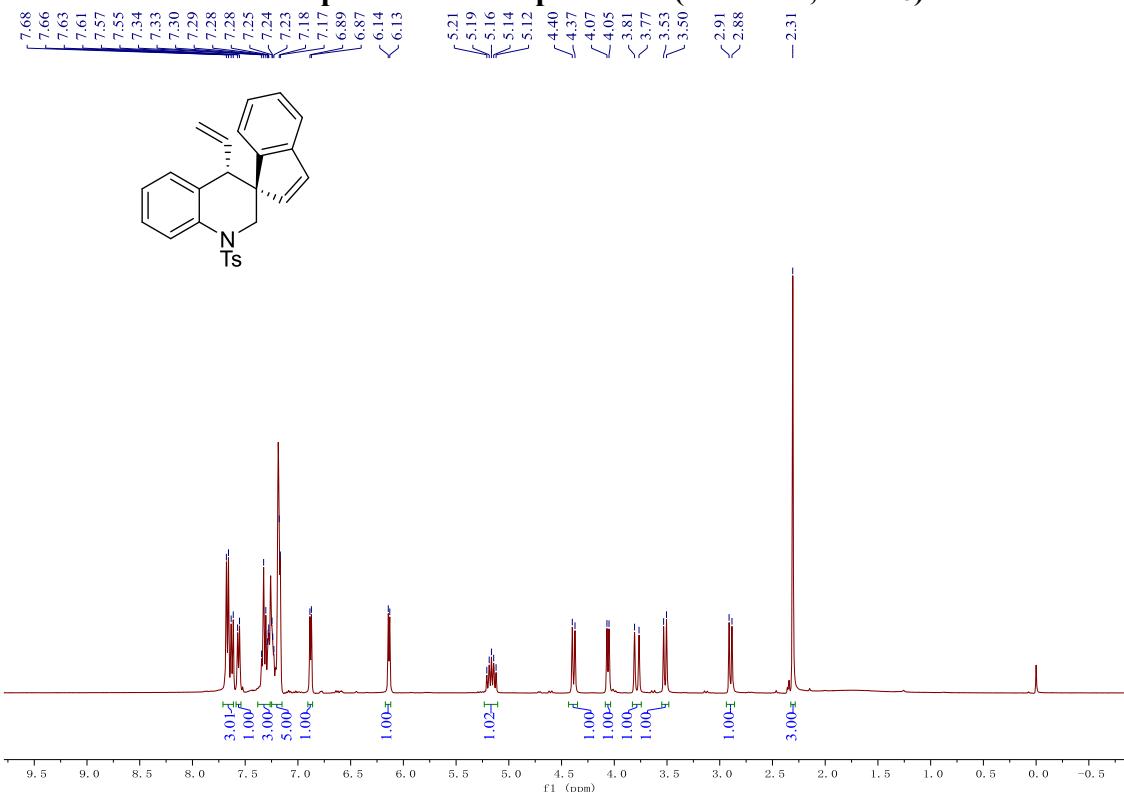
¹H NMR spectrum of compound 4a (400 MHz, CDCl₃)



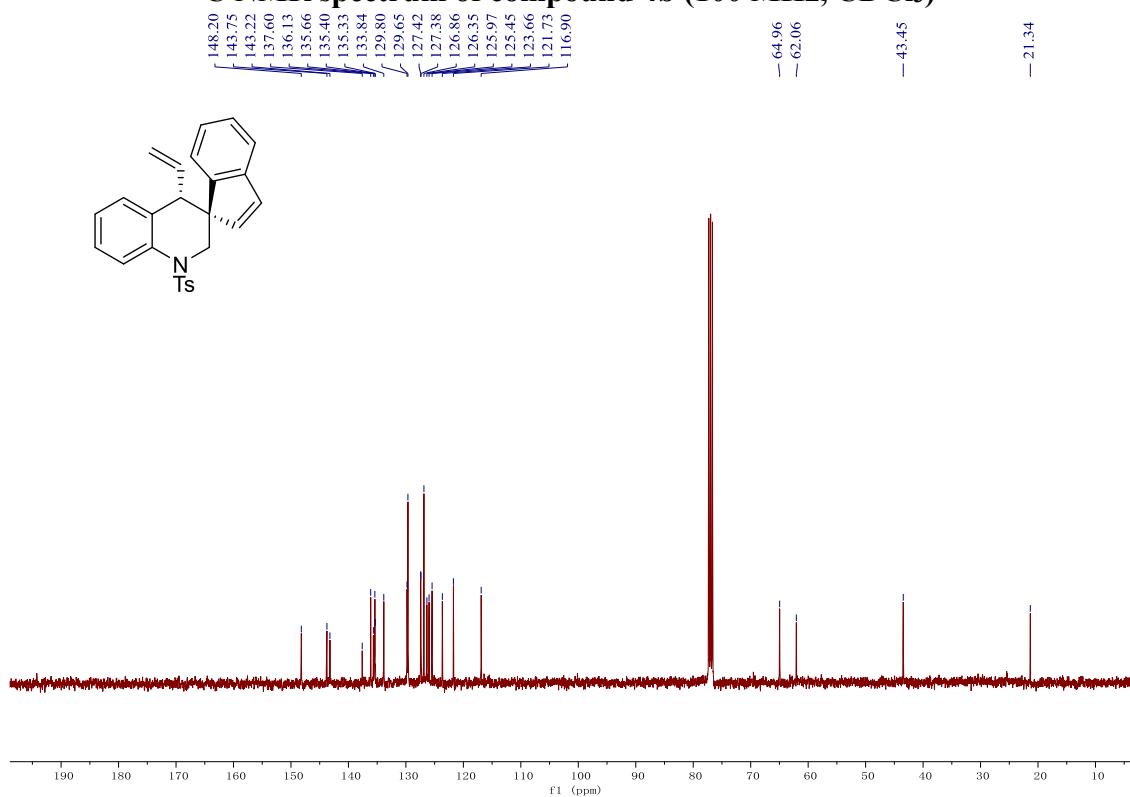
¹³C NMR spectrum of compound 4a (100 MHz, CDCl₃)



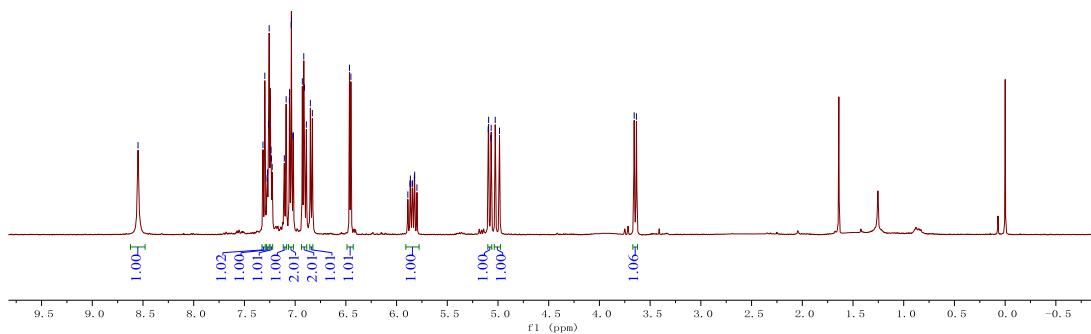
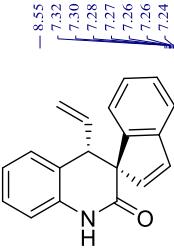
¹H NMR spectrum of compound 4b (400 MHz, CDCl₃)



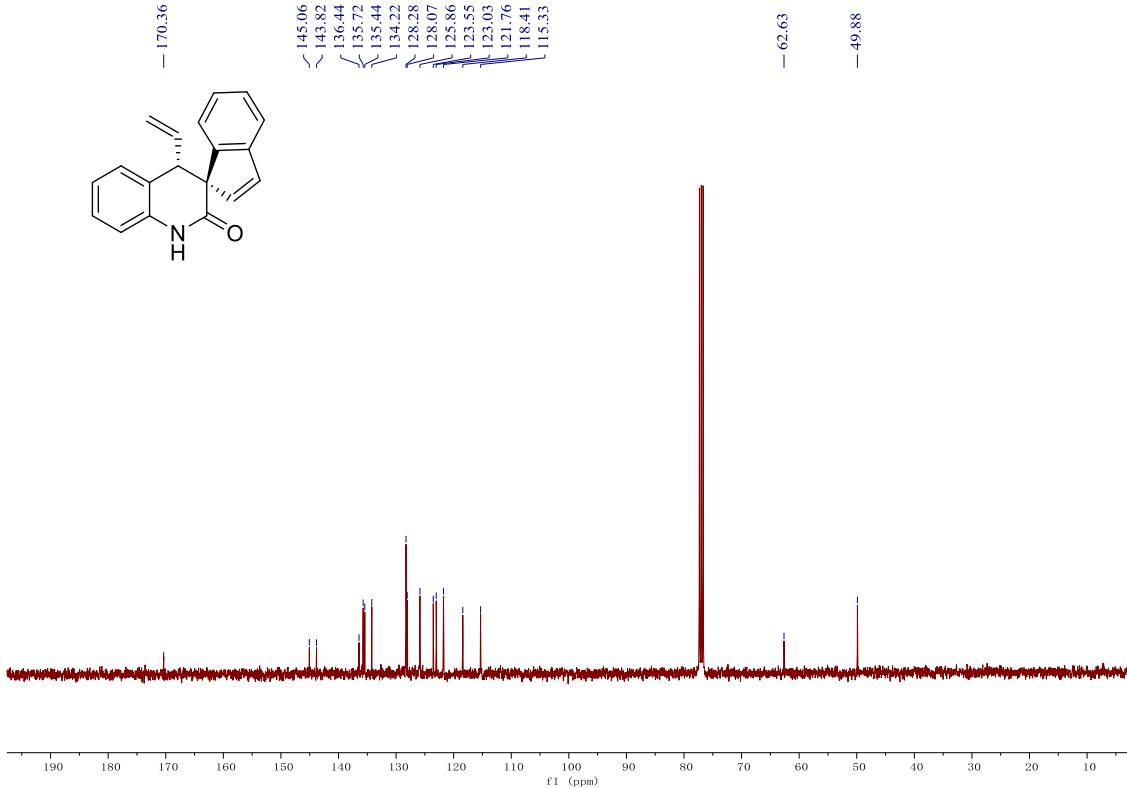
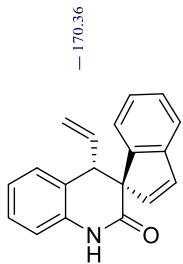
¹³C NMR spectrum of compound 4b (100 MHz, CDCl₃)



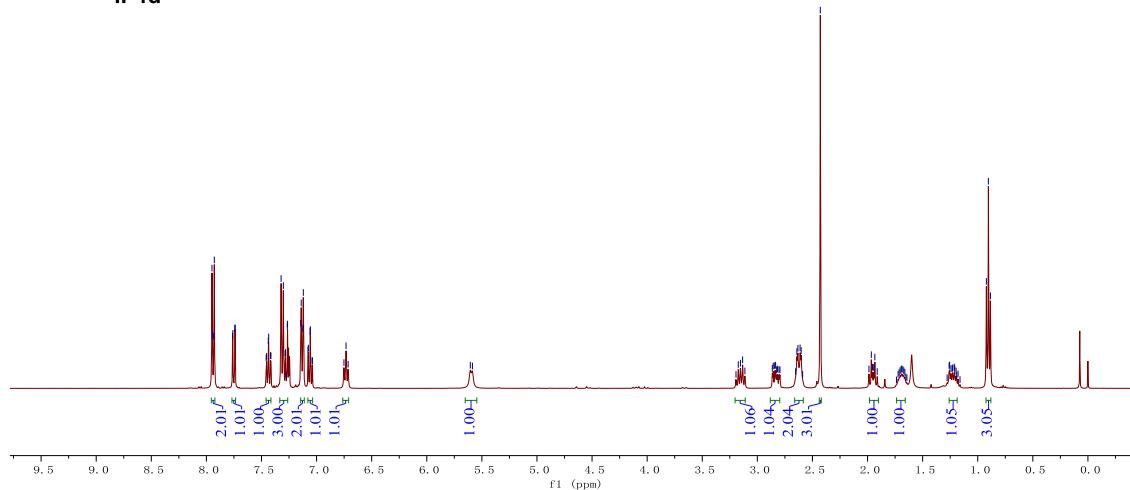
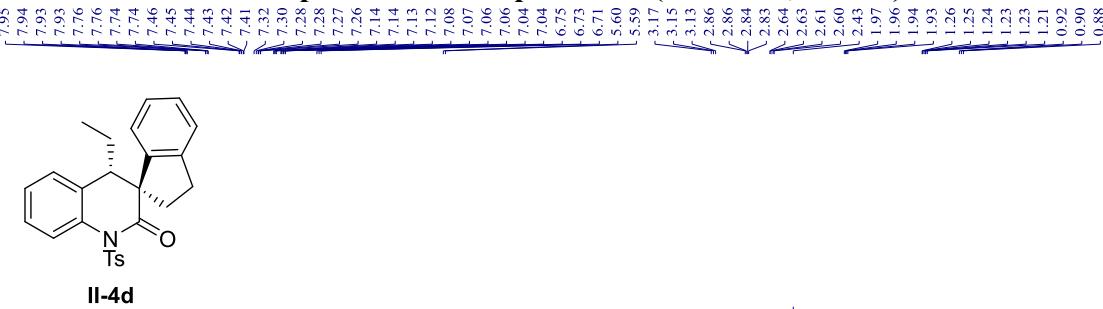
¹H NMR spectrum of compound 4c (400 MHz, CDCl₃)



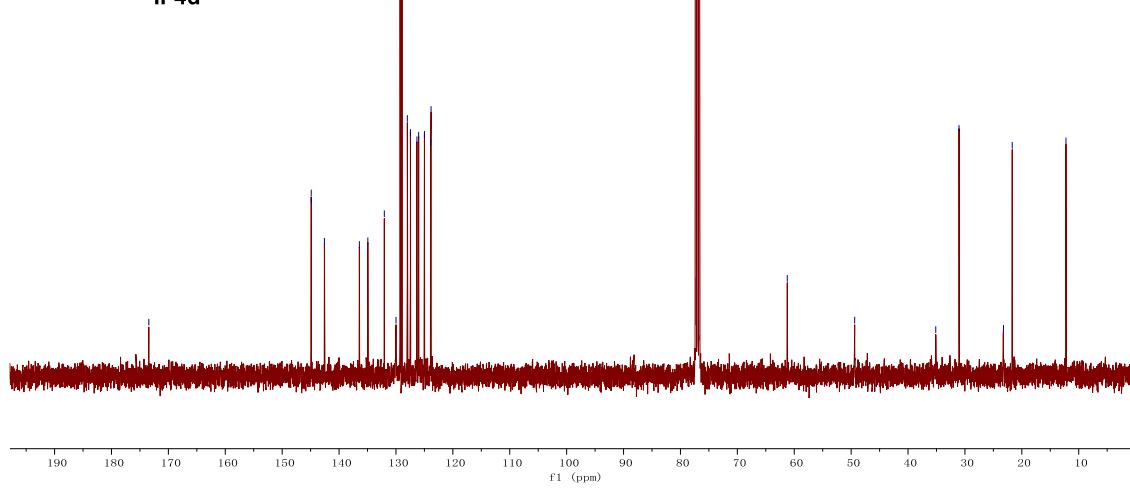
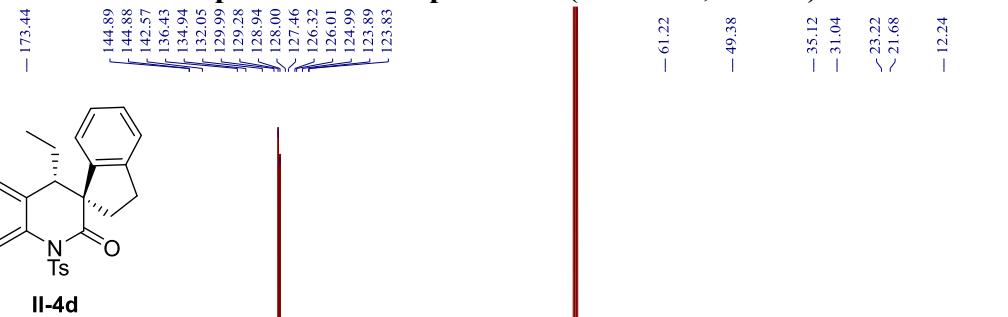
¹³C NMR spectrum of compound 4c (100 MHz, CDCl₃)



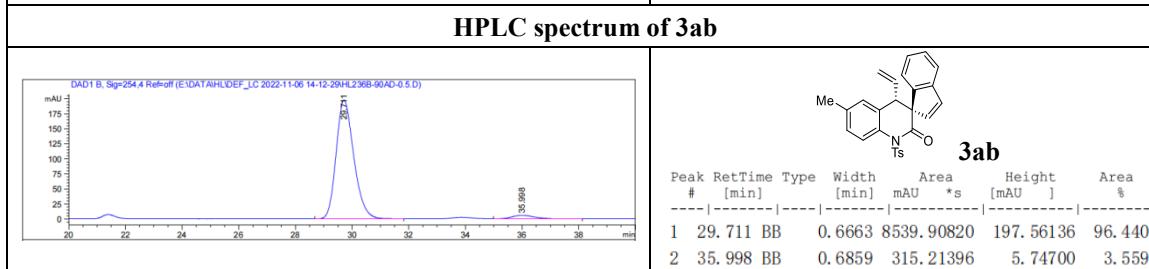
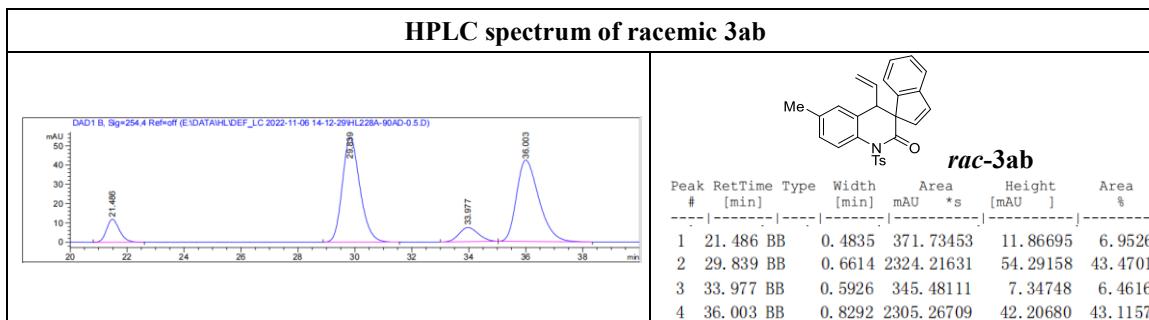
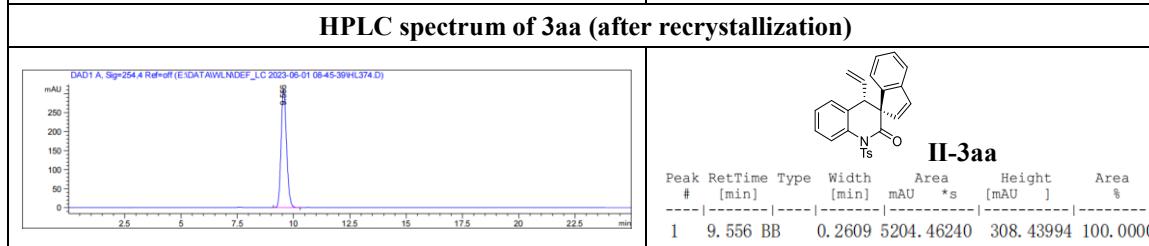
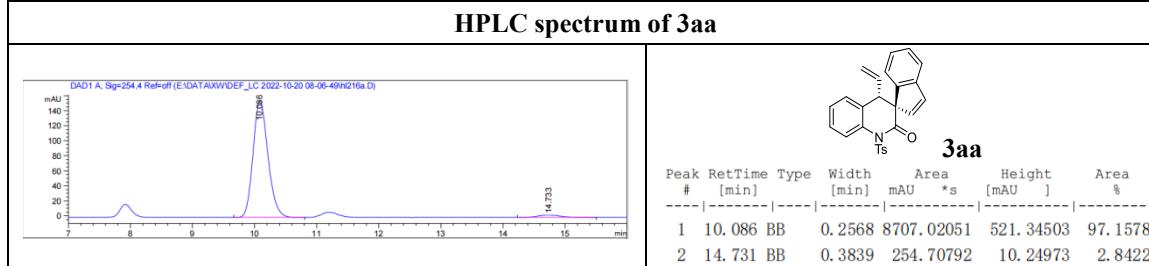
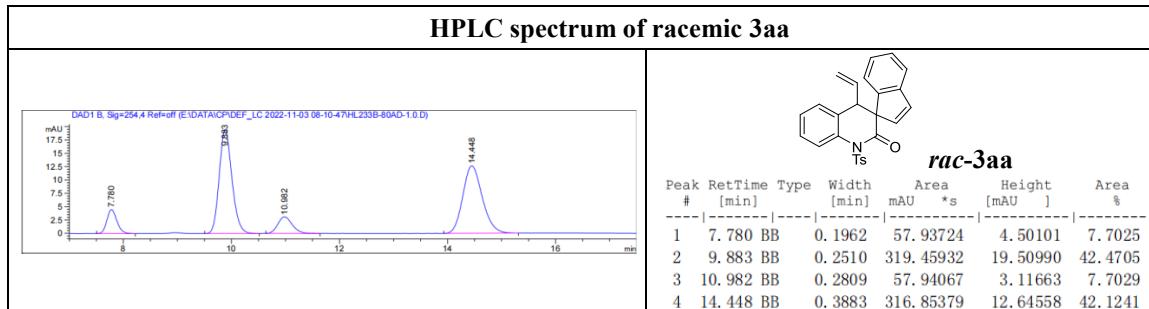
¹H NMR spectrum of compound 4d (400 MHz, CDCl₃)



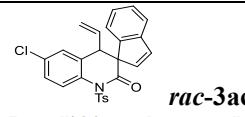
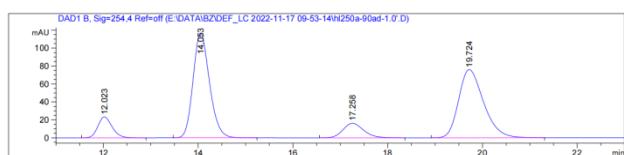
¹³C NMR spectrum of compound 4d (100 MHz, CDCl₃)



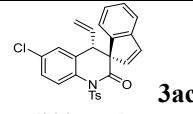
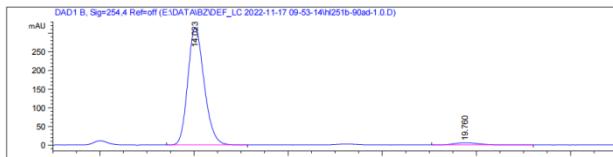
10. Copies of HPLC Spectra



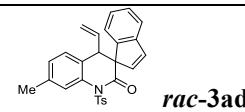
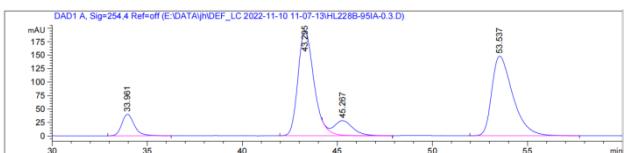
HPLC spectrum of racemic 3ac



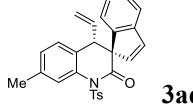
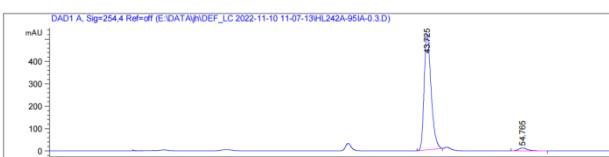
HPLC spectrum of 3ac



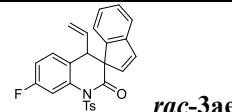
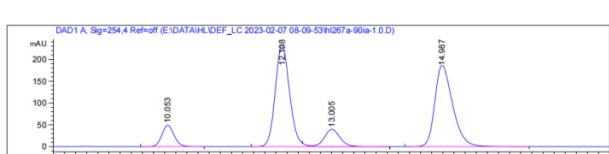
HPLC spectrum of racemic 3ad



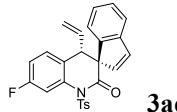
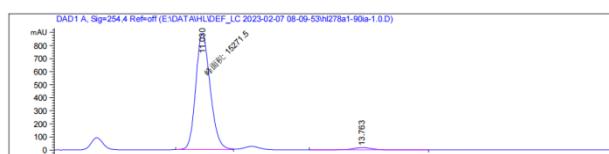
HPLC spectrum of 3ad



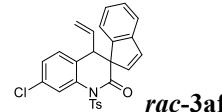
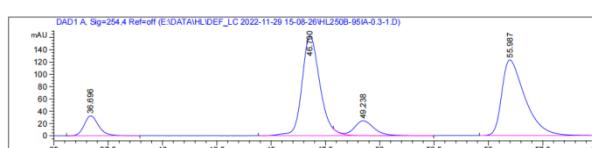
HPLC spectrum of racemic 3ae



HPLC spectrum of 3ae

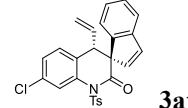
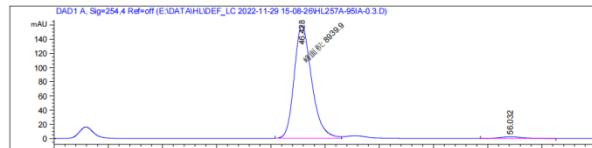


HPLC spectrum of racemic 3af



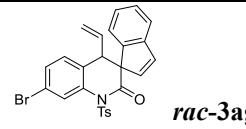
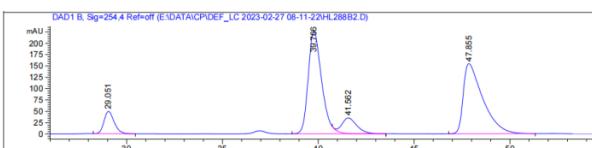
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2	46.790	BV R	0.8738	9296.68750		162.40465	43.4150
3	49.238	BV E	0.9416	1506.90234		24.03176	7.0371
4	55.987	BB	1.1165	9121.87402		122.81918	42.5986

HPLC spectrum of 3af



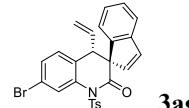
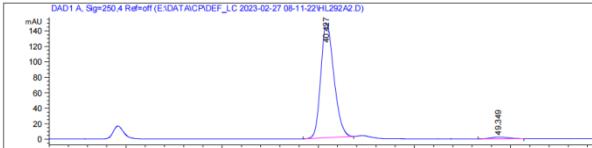
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1	46.428	MM	0.9381	8939.90430		158.82639	98.3270
2	56.032	BB	0.8330	152.11055		2.17225	1.6730

HPLC spectrum of racemic 3ag



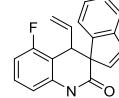
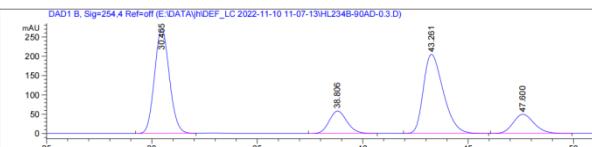
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1	29.051	BB	0.5583	1820.78198		49.13707	7.0944
2	39.766	BV R	0.7426	1.10563e4		225.28391	43.0791
3	41.562	BV E	0.6921	1841.92175		34.10275	7.1768
4	47.855	BB	1.0061	1.09461e4		154.58386	42.6498

HPLC spectrum of 3ag



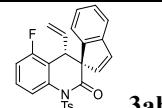
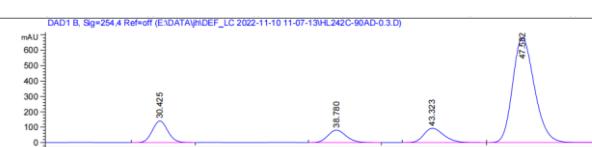
Peak #	RetTime [min]	Type	Width [min]	Area mAU	*s	Height [mAU]	Area %
1	40.427	BB	0.7322	7013.39355		148.16573	98.1715
2	49.349	BB	0.7294	130.62527		2.15439	1.8285

HPLC spectrum of racemic 3ah



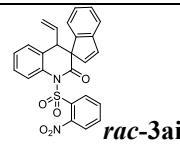
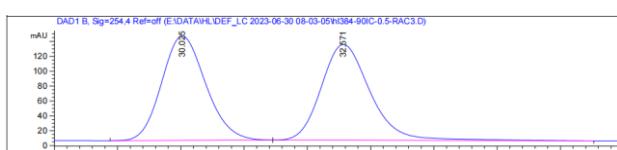
Peak #	RetTime [min]	Type	Width [min]	Area mAU	*s	Height [mAU]	Area %
1	30.465	BB	0.7637	1.32719e4		270.84818	39.6436
2	38.806	BB	0.8975	3390.18774		58.03740	10.1266
3	43.261	BB	1.0024	1.34025e4		204.56905	40.0336
4	47.600	BB	1.0394	3413.50000		49.32220	10.1962

HPLC spectrum of 3ah

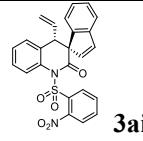
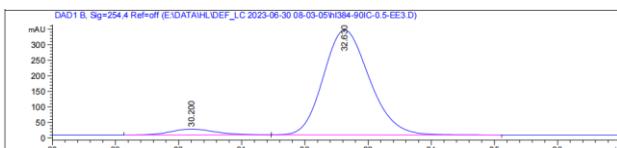


Peak #	RetTime [min]	Type	Width [min]	Area mAU	*s	Height [mAU]	Area %
1	30.425	BB	0.7945	7084.90918		139.98291	10.6524
2	38.780	BB	0.9262	4816.06494		80.71114	7.2411
3	43.323	BB	0.9942	6023.51611		92.68636	9.0565
4	47.590	BB	1.1081	4.85857e4		681.19043	73.0500

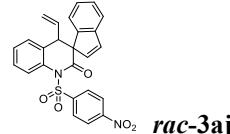
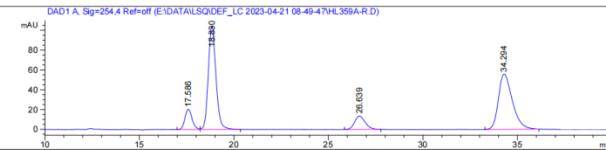
HPLC spectrum of racemic 3ai



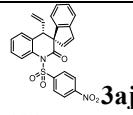
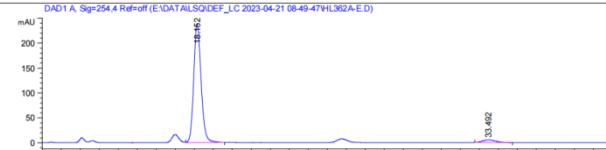
HPLC spectrum of 3ai



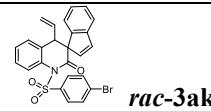
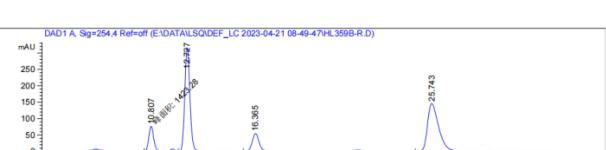
HPLC spectrum of racemic 3aj



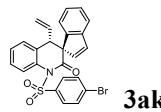
HPLC spectrum of 3aj



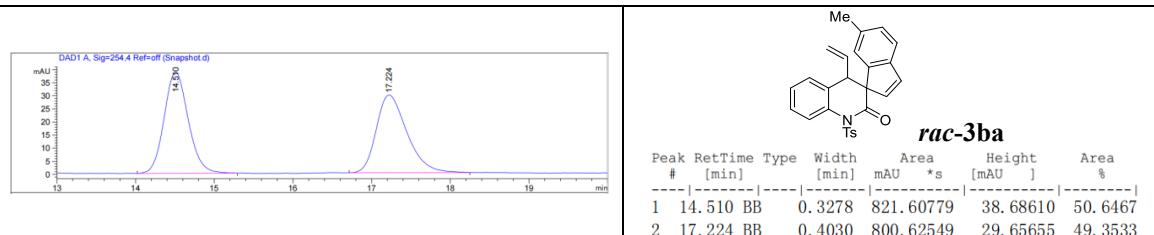
HPLC spectrum of racemic 3ak



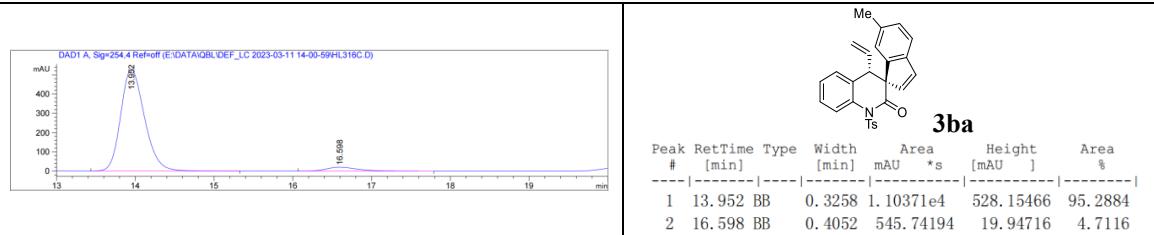
HPLC spectrum of 3ak



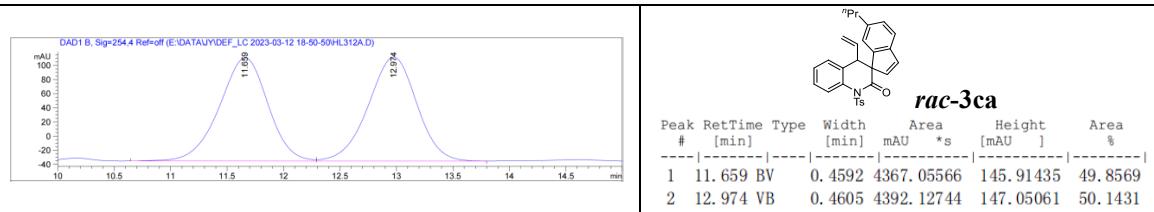
HPLC spectrum of racemic 3ba



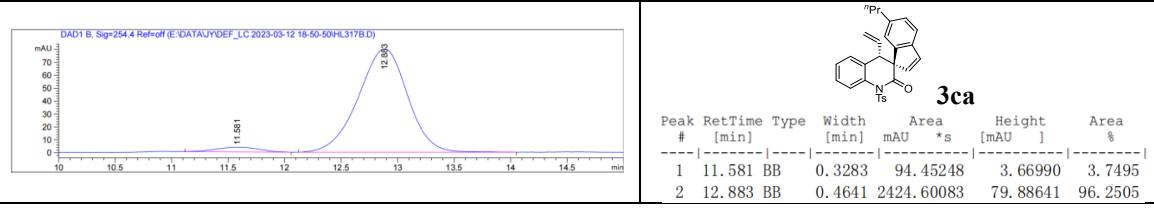
HPLC spectrum of 3ba



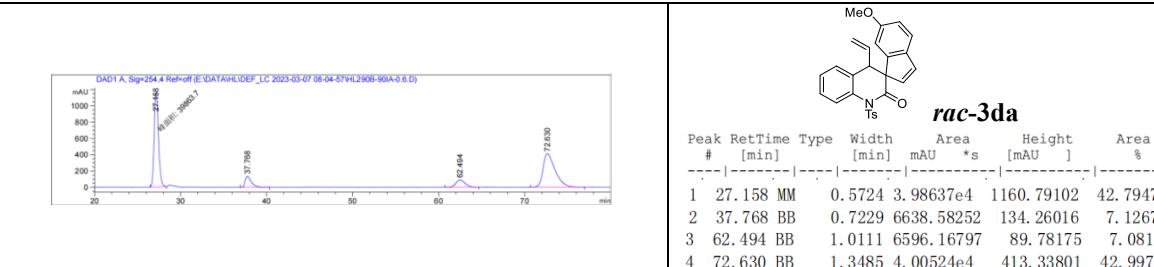
HPLC spectrum of racemic 3ca



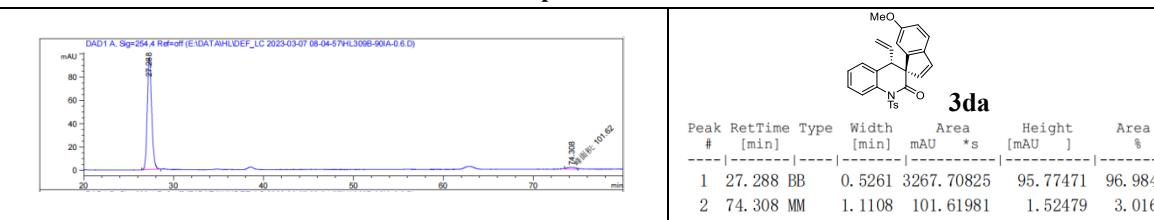
HPLC spectrum of 3ca

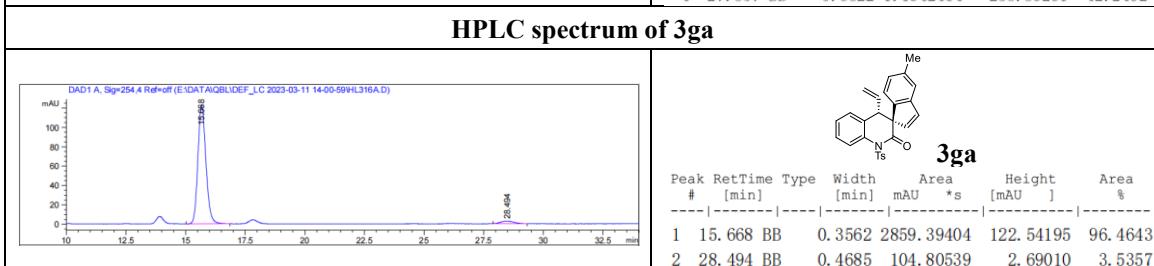
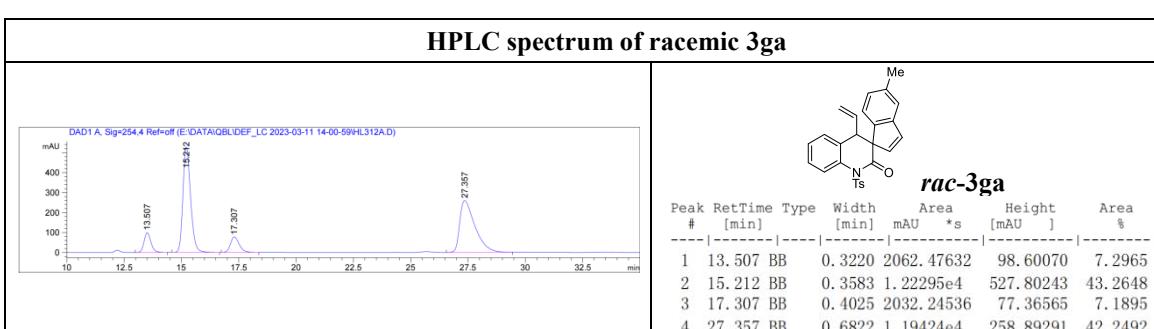
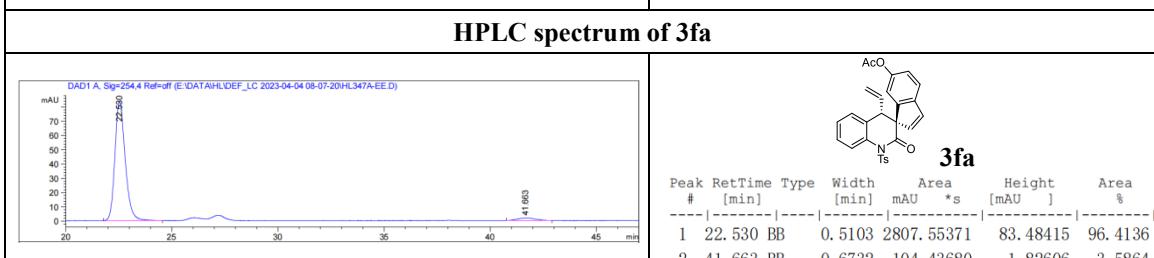
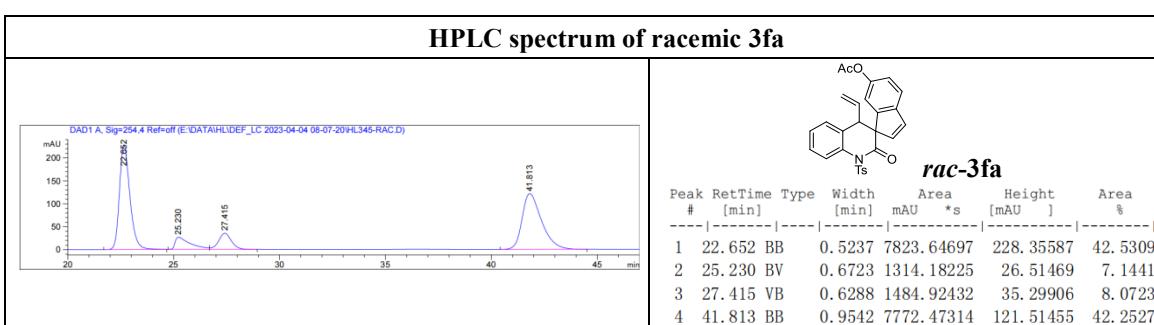
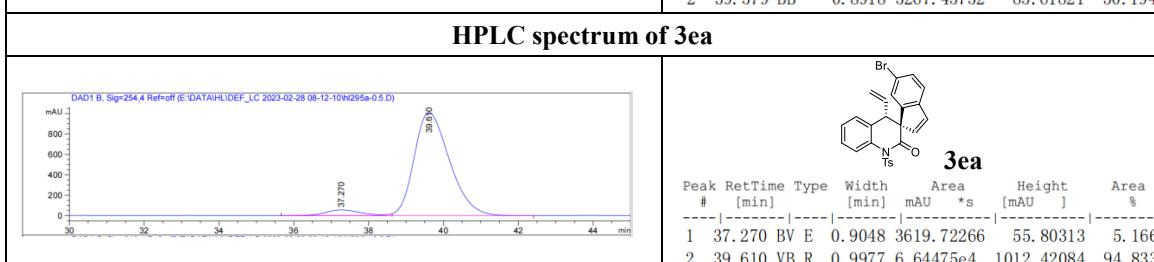
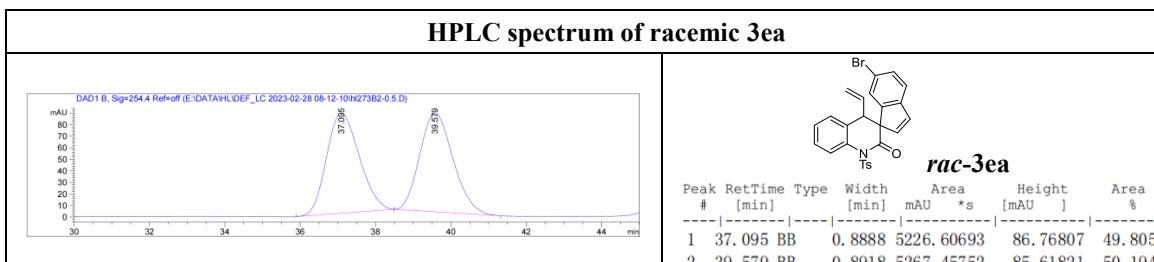


HPLC spectrum of racemic 3da

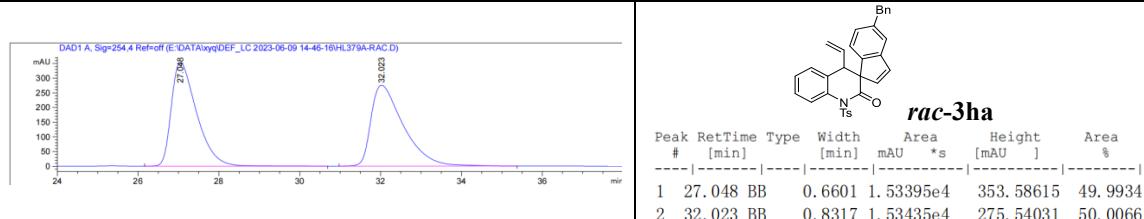


HPLC spectrum of 3da

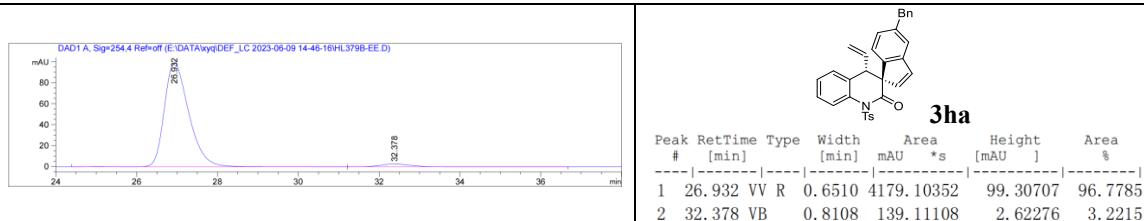




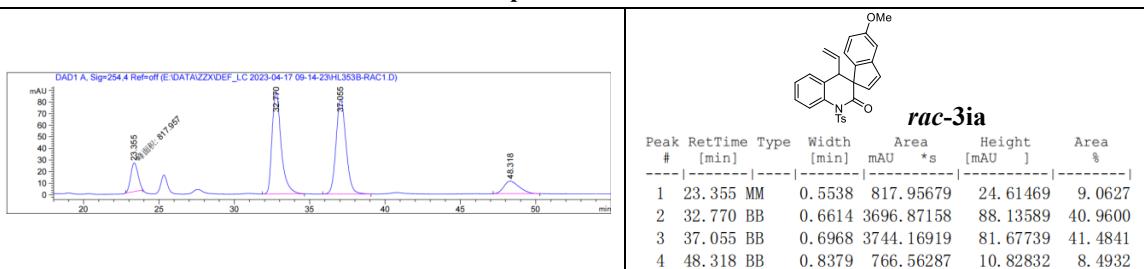
HPLC spectrum of racemic 3ha



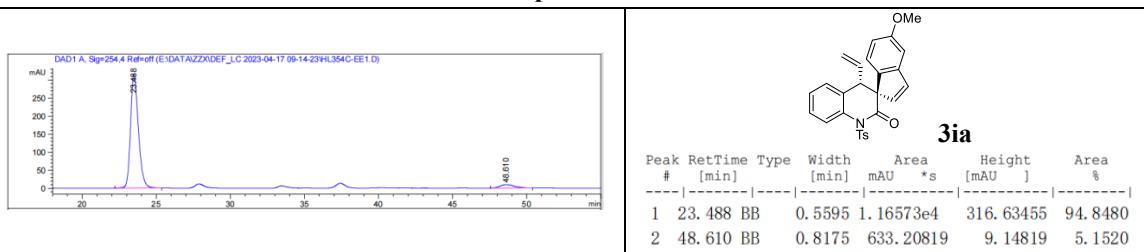
HPLC spectrum of 3ha



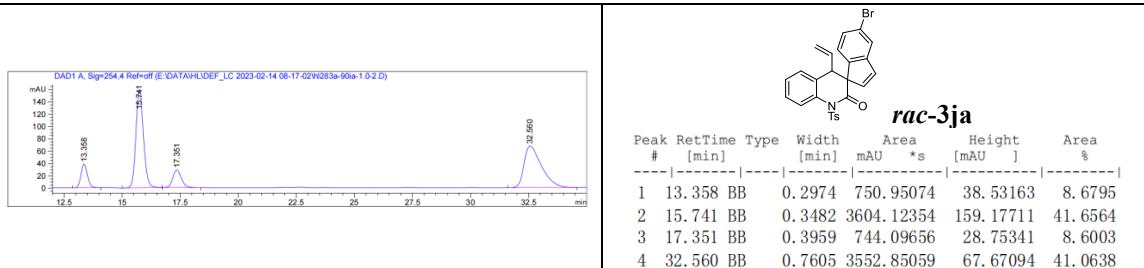
HPLC spectrum of racemic 3ia



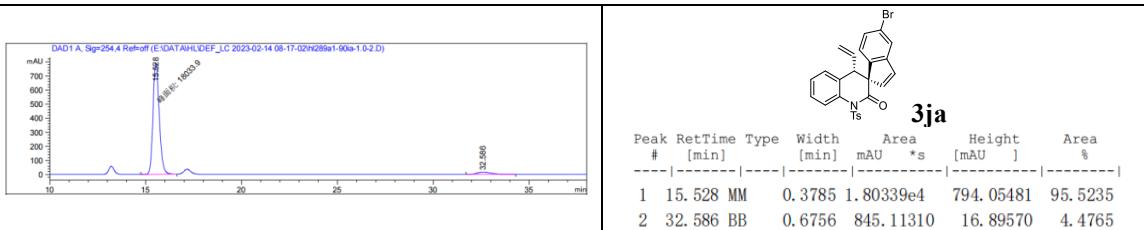
HPLC spectrum of 3ia



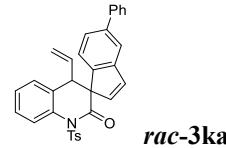
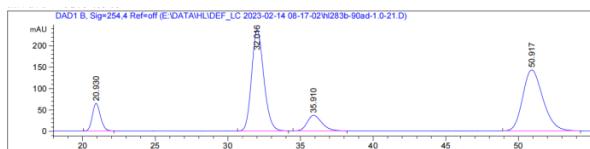
HPLC spectrum of racemic 3ja



HPLC spectrum of 3ja

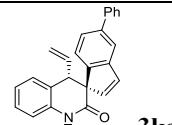
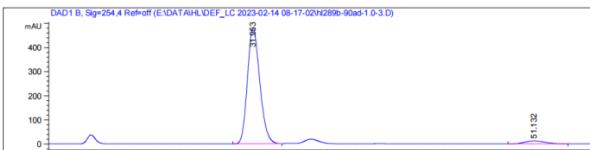


HPLC spectrum of racemic 3ka



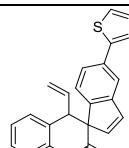
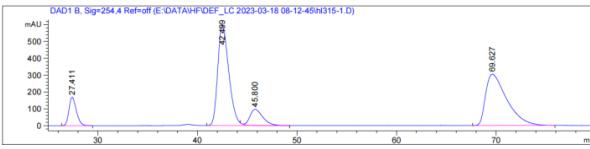
Peak #	RetTime [min]	Type	Width [min]	mAU	*s	Area [mAU]	Height [mAU]	Area %
1	20.930	BB	0.6003	2497.46460	64.42068	7.6866		
2	32.016	BB	0.8831	1.37506e4	236.21318	42.3208		
3	35.910	BB	0.9306	2512.97949	36.07872	7.7343		
4	50.917	BB	1.3447	1.37303e4	142.42239	42.2583		

HPLC spectrum of 3ka



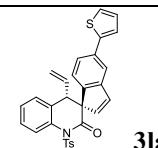
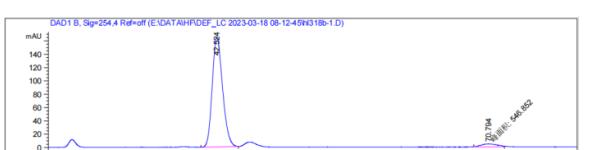
Peak #	RetTime [min]	Type	Width [min]	mAU	*s	Area [mAU]	Height [mAU]	Area %
1	31.953	BB	0.8979	2.81663e4	480.47113	96.0279		
2	51.132	BB	1.1272	1165.07983	12.21028	3.9721		

HPLC spectrum of racemic 3la



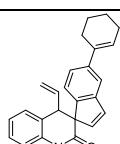
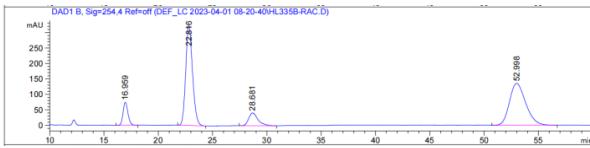
Peak #	RetTime [min]	Type	Width [min]	mAU	*s	Area [mAU]	Height [mAU]	Area %
1	27.411	BB	0.7960	8691.08887	168.42078	8.0206		
2	42.499	BV R	1.1517	4.61223e4	598.05658	42.5638		
3	45.800	BV E	1.2049	8607.98242	96.47797	7.9439		
4	69.627	BB	2.0279	4.49389e4	304.13571	41.4718		

HPLC spectrum of 3la



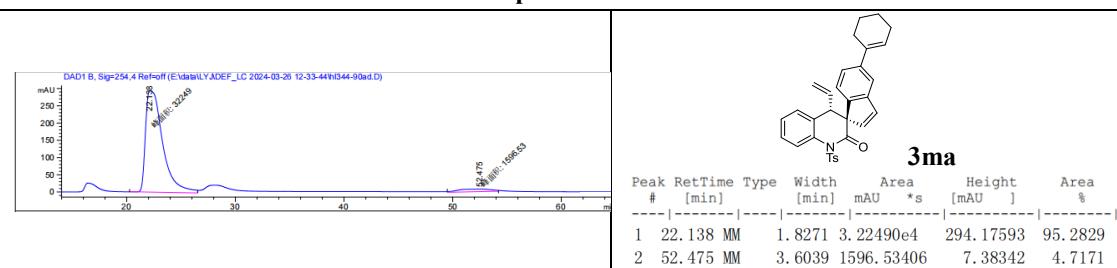
Peak #	RetTime [min]	Type	Width [min]	mAU	*s	Area [mAU]	Height [mAU]	Area %
1	42.524	BB	1.1305	1.22822e4	164.23651	95.7374		
2	70.794	MM	1.9702	546.85229	4.62612	4.2626		

HPLC spectrum of racemic 3ma

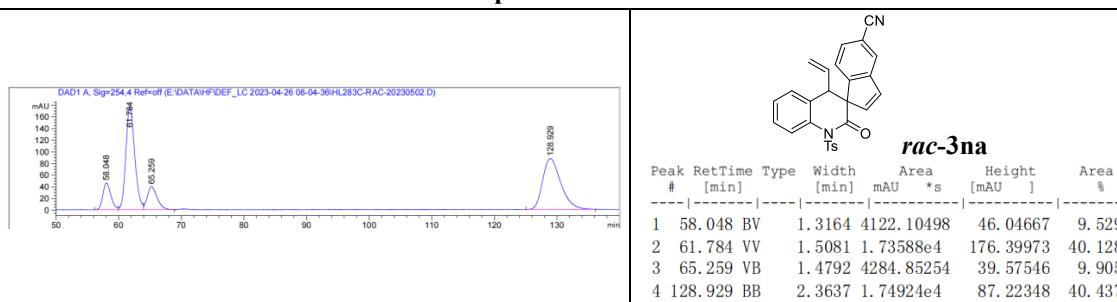


Peak #	RetTime [min]	Type	Width [min]	mAU	*s	Area [mAU]	Height [mAU]	Area %
1	16.699	BB	0.4834	2328.20215	74.33793	7.0734		
2	22.816	BB	0.6597	1.40419e4	322.67844	42.6610		
3	28.681	BB	0.9172	2468.67773	41.19395	7.5001		
4	52.998	BB	1.5400	1.40763e4	135.26993	42.7655		

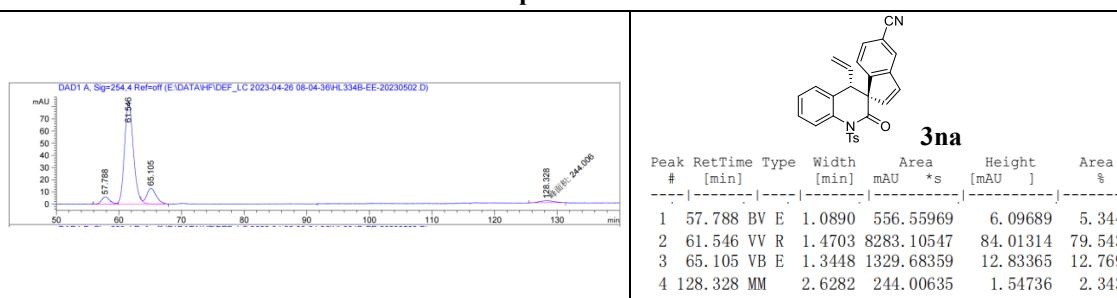
HPLC spectrum of 3ma



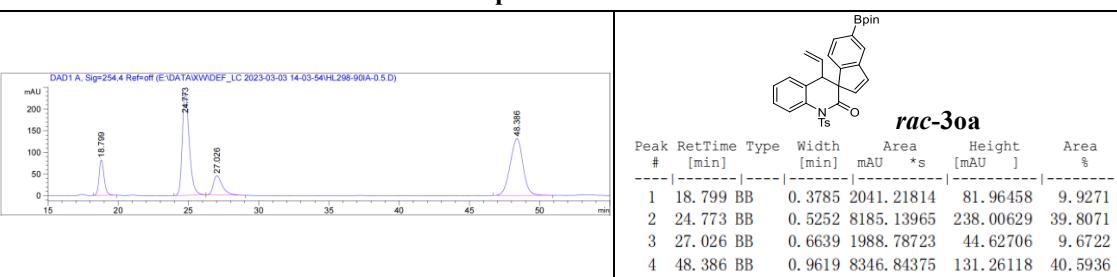
HPLC spectrum of racemic 3na



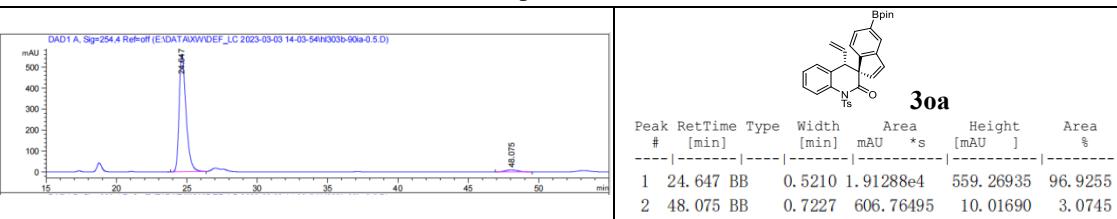
HPLC spectrum of 3na



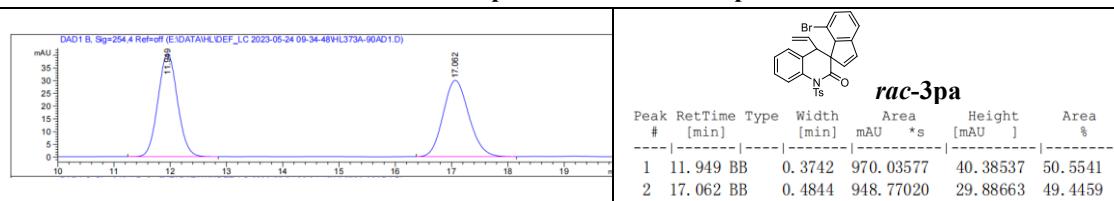
HPLC spectrum of racemic 3oa



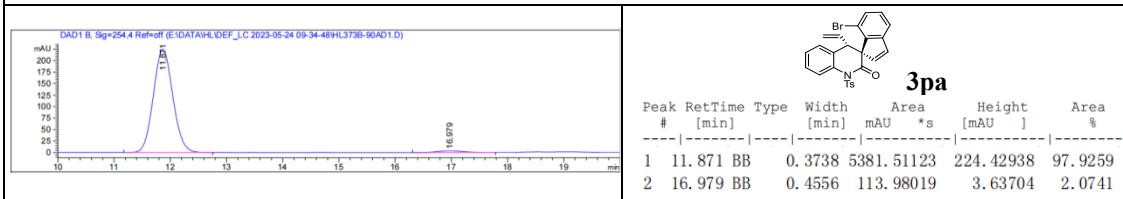
HPLC spectrum of 3oa



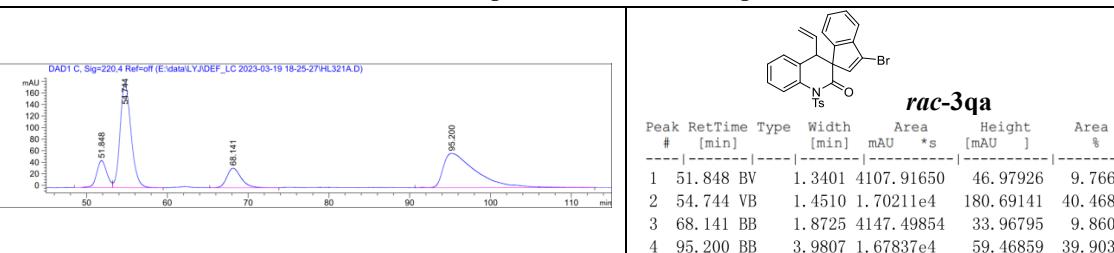
HPLC spectrum of racemic 3pa



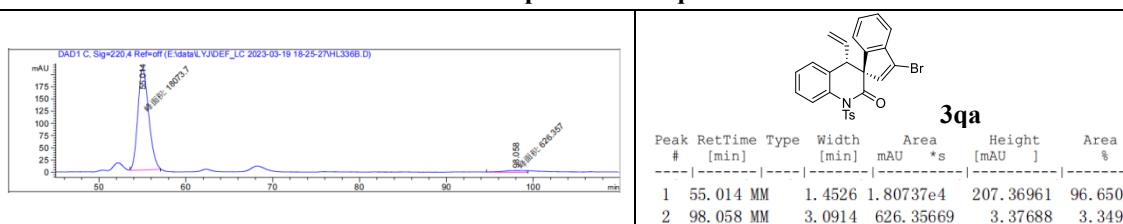
HPLC spectrum of 3pa



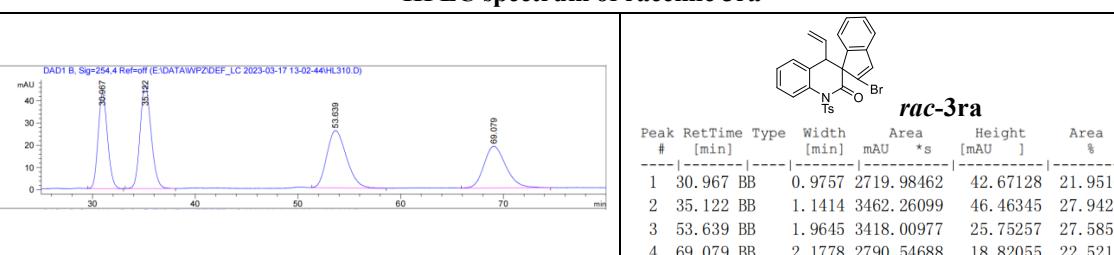
HPLC spectrum of racemic 3qa



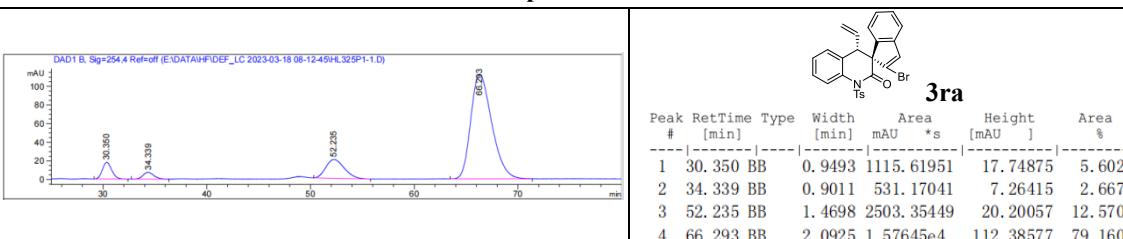
HPLC spectrum of 3qa



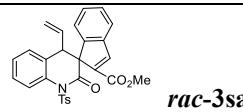
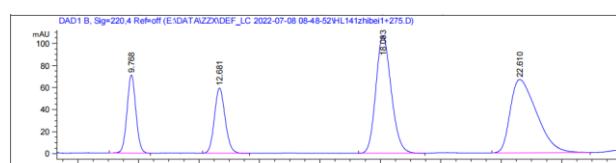
HPLC spectrum of racemic 3ra



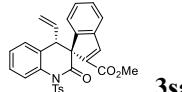
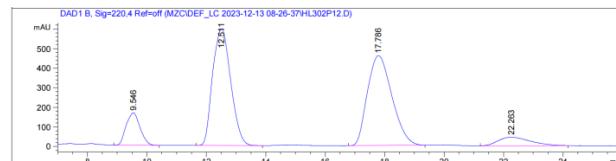
HPLC spectrum of 3ra



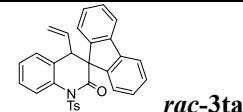
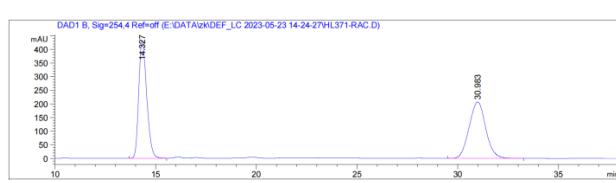
HPLC spectrum of racemic 3sa



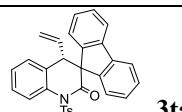
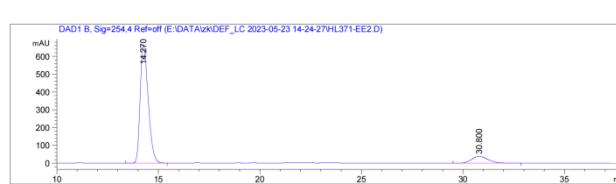
HPLC spectrum of 3sa



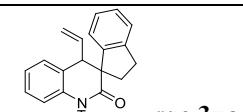
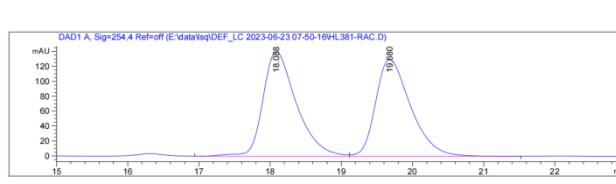
HPLC spectrum of racemic 3ta



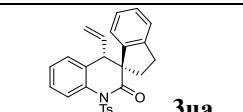
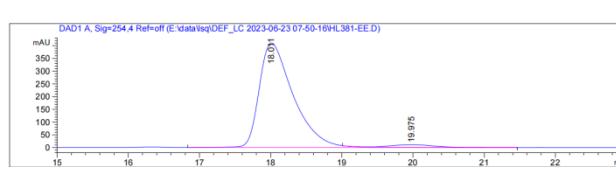
HPLC spectrum of 3ta



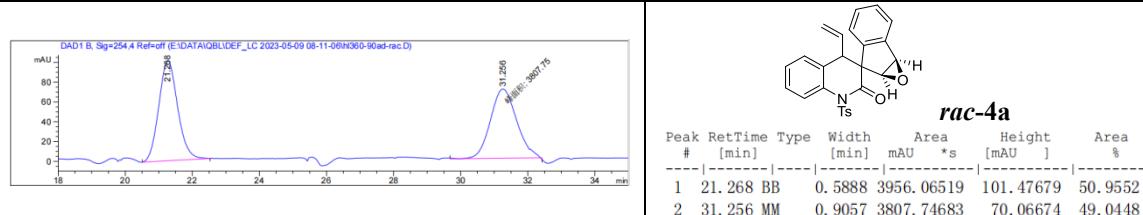
HPLC spectrum of racemic 3ua



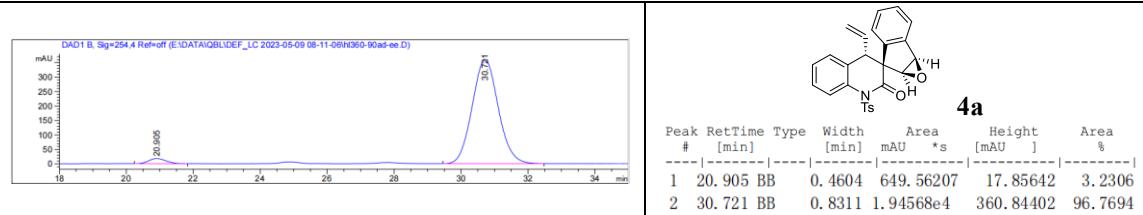
HPLC spectrum of 3ua



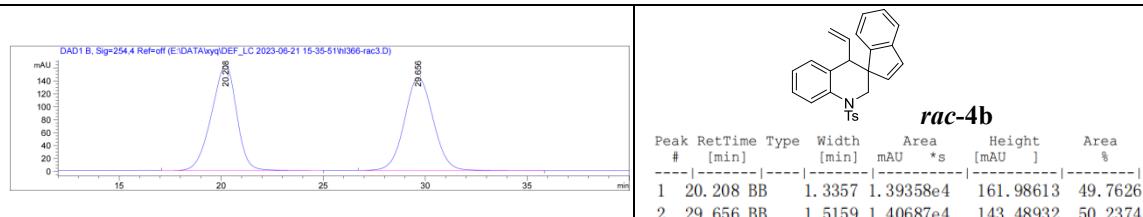
HPLC spectrum of racemic 4a



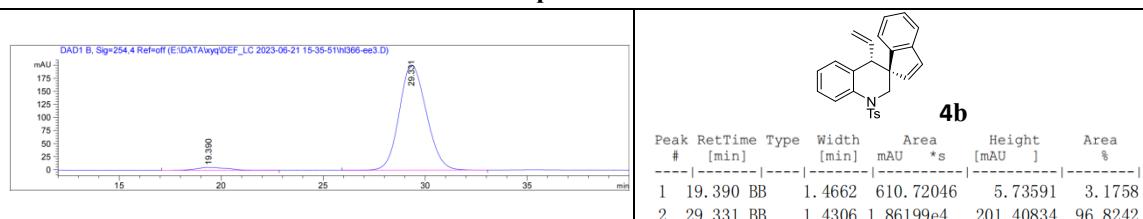
HPLC spectrum of 4a



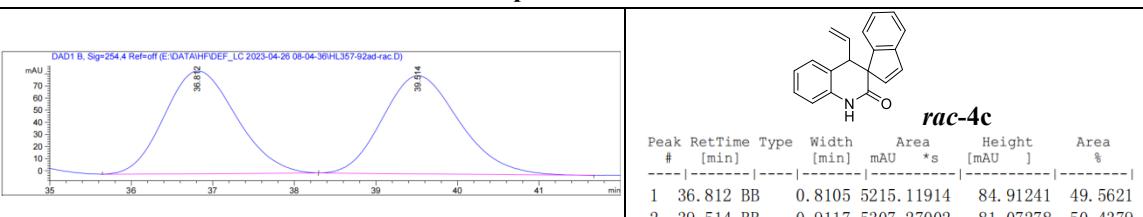
HPLC spectrum of racemic 4b



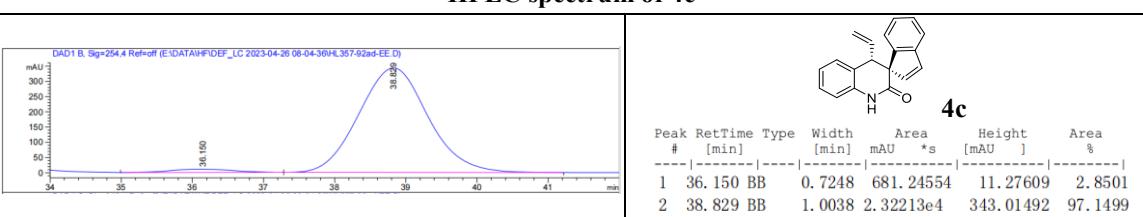
HPLC spectrum of 4b



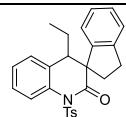
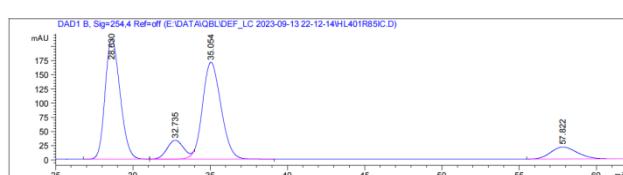
HPLC spectrum of racemic 4c



HPLC spectrum of 4c

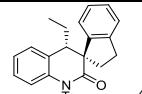
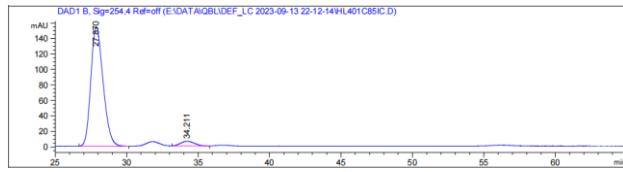


HPLC spectrum of racemic 4d



Peak #	RetTime [min]	Type	Width [min]	Area mAU	Height *s	Height [mAU]	Area %
1	28.630	BV	1.0491	1.42749e4	211.06267	42.5614	
2	32.735	VV E	1.1045	2430.67896	33.11080	7.2472	
3	35.054	VB R	1.3116	1.42840e4	170.81622	42.5887	
4	57.822	BB	1.6367	2549.88721	21.05850	7.6027	

HPLC spectrum of 4d



Peak #	RetTime [min]	Type	Width [min]	Area mAU	Height *s	Height [mAU]	Area %
1	27.870	BB	0.9014	9181.82715	154.04274	95.7462	
2	34.211	BB	0.8019	407.92734	6.04058	4.2538	