Electronic Supplementary Material (ESI) for Organic Chemistry Frontiers.

A facile approach to the azaindoline functionalized spirocarbocyclic scaffolds via Pd-catalyzed cascade cyclization/dearomatization process

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Supporting Information

Content:

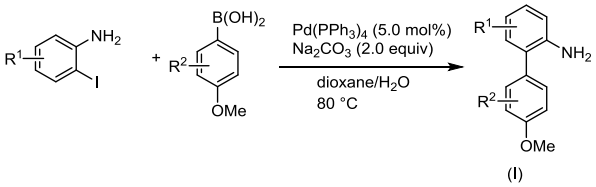
1. General considerations.

Unless stated otherwise, all reactions were carried out under a nitrogen atmosphere. Materials were obtained from commercial suppliers or prepared according to standard procedures unless otherwise noted. Solvents were purified and dried according to standard methods prior to use. For product purification by flash column chromatography, silica gel (200~300 mesh) and light petroleum ether (bp. 60~90) are used. ^{1}H NMR spectra were recorded on a Bruker advance III 500 MHz in $CDCl_3$ and ^{13}C NMR spectra were recorded on 126 MHz in $CDCl_3$ using TMS as internal standard, Data for ^{1}H NMR are recorded as follows: chemical shift (δ, ppm), multiplicity (s = singlet, d = doublet, t = triplet, m = multiplet or unresolved, br = broad singlet, coupling constant (s) in Hz, integration). Data for ^{13}C NMR is reported in terms of chemical shift (δ, ppm). High-resolution mass spectral analysis (HRMS) data were measured on a Bruker Apex II.

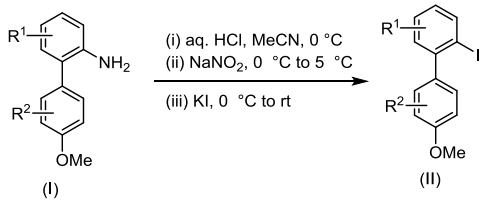
2. Preparation of starting materials.

Substrates **1** were prepared according to the known procedures.[1]

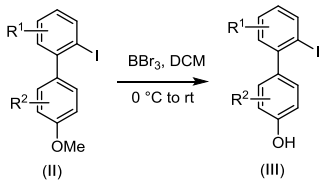
Substrates **2** were prepared according to the following procedure.[2]



A 100 mL round bottom flask was charged with $Pd(PPh_3)_4$ (0.29 g, 0.25 mmol), Na_2CO_3 (1.06 g, 10.0 mmol), boronic acid (7.5 mmol, 1.5 equiv.), ortho-iodoaniline derivatives (5.0 mmol), 20.0 ml deoxygenated 1,4-dioxane and 50.0 ml deoxygenated water. The mixture was stirred at 80 ℃ under argon conditions until the reaction was judged to be completed by TLC analysis. Water was added and extracted with EtOAc. The organic phase was dried over anhydrous $MgSO_4$ and concentrated under reduced pressure. The residue was then chromatographed on silica gel to afford the [1,1'-biphenyl]-2-amine derivative (**I**).

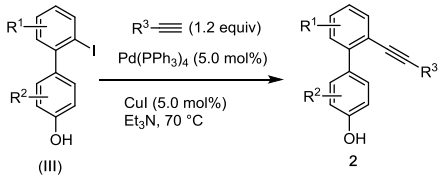


To a solution of the [1,1'-biphenyl]-2-amine derivative (**I**) (3.0 mmol) dissolved in MeCN (8.0 ml) was added aq. HCl (1.6 ml conc. HCl in 5.0 ml water), then the mixture was cooled to 0 ℃, and it was added a solution of $NaNO_2$ (0.25 g, 3.6 mmol) in 5.0 ml water. After addition, the reaction was kept at the temperature lower than 5 ℃ for 1 h and it was added a solution of KI (0.75 g, 4.5 mmol) in 5.0 ml water. The reaction was kept at room temperature overnight. Water was added and extracted with EtOAc. The organic phase was dried over anhydrous $MgSO_4$ and concentrated. The residue was then chromatographed on silica gel to afford the 2-iodo-1,1'-biphenyl derivative (**II**).



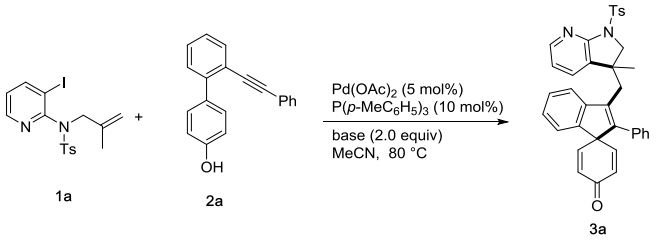
Under an argon atmosphere, a solution of BBr_3 (4.0 mmol) in dry DCM (3.0 ml) was slowly added to a solution of the above product (2.0 mmol) in dry DCM (15.0 ml) at 0 ℃ and the mixture was stirred at room

temperature for 3 h. After cooling to 0 ℃, the reaction was quenched with cold water and extracted with DCM. The organic phase was dried over anhydrous $MgSO_4$ and concentrated under reduced pressure. The residue was then chromatographed on silica gel to afford the desired product (**III**).



To a solution of substituted 2'-iodo-[1,1'-biphenyl]-4-ol (**III**) (5.0 mmol) in Et_3N (20 ml) was added terminal alkyne (6.0 mmol, 1.2 equiv.), $Pd(PPh_3)_4$ (0.25 mmol, 5 mol%), CuI (0.25 mmol, 5mol%), the mixture was then heated to 70 ℃ for 12 h under argon conditions. After cooling to room temperature, Water was added and extracted with EtOAc. The organic phase was dried over anhydrous $MgSO_4$ and concentrated. The crude product was purified by column chromatography to afford the desired product **2**.

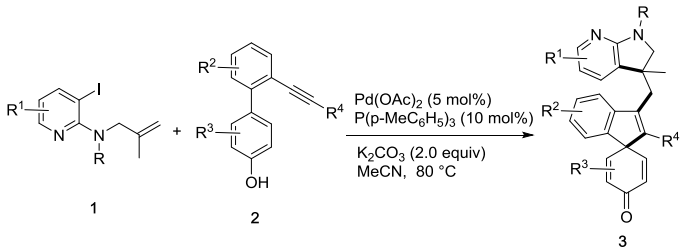
3. Optimization of reaction conditions: Screening of bases.



entry	base	yield (%)
1	K_2CO_3	71
2	Cs_2CO_3	57
3	Na_2CO_3	43
4	KOH	<10
5	^{t}BuOLi	<10
6	^{t}BuOK	<10
7	Et_3N	0
8	DBU	0

Unless otherwise noted, Reactions were carried out using 1a (0.2 mmol), 2 (0.26 mmol), $Pd(OAc)_2$ (5.0 mol%) and P(*p*-$MeC_6H_5)_3$ (10.0 mol%) in MeCN (2.0 ml)

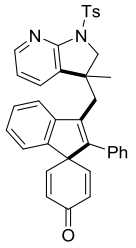
4. General procedure for the preparation of the product 3.



1 (0.2 mmol), **2** (0.26 mmol), $Pd(OAc)_2$ (5 mol%), P(*p*-$MeC_6H_5)_3$ (10 mol%), K_2CO_3 (2.0 equiv.) were added to a sealed tube, MeCN (2 ml) was added via syringe. The mixture was flushed with N_2 and stirred at 80 ℃ about 24 h until completion (monitored by TLC). After cooling at room temperature, the mixture was diluted with diethyl

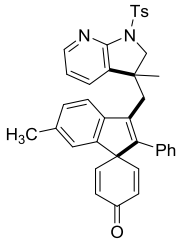
ether, washed with water, dried over magnesium sulfate and purified by flash chromatography on silica gel (petroleum ether/ethylacetate 5:1-2:1) to give the desired product **3**.

5. Spectra data of products.



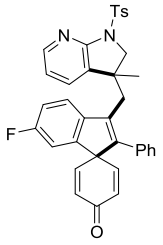
3a

3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3a): solid; m.p. 126-128 ℃; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.07 (dd, *J* = 5.2, 1.7 Hz, 1H), 7.77 (d, *J* = 8.1 Hz, 2H), 7.32-7.20 (m, 5H), 7.16 (d, *J* = 8.1 Hz, 2H), 7.12-7.04 (m, 2H), 7.02 (h, *J* = 4.3 Hz, 2H), 6.86 (dd, *J* = 7.4, 1.7 Hz, 1H), 6.56 (dd, *J* = 7.4, 5.1 Hz, 1H), 6.46 (dd, *J* = 9.9, 2.8 Hz, 1H), 6.36 (ddd, *J* = 16.2, 9.8, 1.7 Hz, 2H), 6.23 (dd, *J* = 9.8, 2.8 Hz, 1H), 3.94 (d, *J* = 9.7 Hz, 1H), 3.40 (d, *J* = 9.7 Hz, 1H), 3.18-3.00 (m, 2H), 2.34 (s, 3H), 1.22 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.14, 155.16, 148.48, 148.34, 147.23, 146.07, 145.81, 143.99, 140.79, 138.99, 134.62, 134.22, 131.27, 130.88, 130.78, 130.53, 129.22, 128.49, 128.47, 128.35, 128.05, 127.85, 126.57, 123.62, 120.65, 118.15, 61.61, 60.11, 42.69, 35.20, 26.22, 21.50. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{36}H_{30}N_2O_3SNa$: 593.1869; Found, 593.1869.



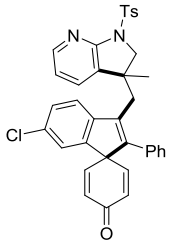
3b

6'-methyl-3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3b): viscous oil; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.07 (dd, *J* = 5.1, 1.6 Hz, 1H), 7.82-7.72 (m, 2H), 7.26 (dd, *J* = 4.3, 2.2 Hz, 3H), 7.16 (d, *J* = 8.1 Hz, 2H), 7.11 (dd, *J* = 7.8, 1.6 Hz, 1H), 7.07-6.99 (m, 2H), 6.97 (d, *J* = 7.8 Hz, 1H), 6.92-6.84 (m, 2H), 6.57 (dd, *J* = 7.4, 5.1 Hz, 1H), 6.47 (dd, *J* = 9.7, 2.9 Hz, 1H), 6.41-6.30 (m, 2H), 6.24 (dd, *J* = 9.7, 2.8 Hz, 1H), 3.93 (d, *J* = 9.7 Hz, 1H), 3.39 (d, *J* = 9.7 Hz, 1H), 3.15-2.97 (m, 2H), 2.35 (d, *J* = 4.7 Hz, 6H), 1.20 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.27, 155.18, 148.84, 148.67, 147.24, 144.96, 144.00, 143.30, 140.92, 138.90, 136.69, 134.84, 134.29, 131.26, 130.76, 130.69, 130.63, 129.22, 129.05, 128.56, 128.49, 128.11, 127.76, 124.43, 120.44, 118.14, 61.51, 60.14, 42.71, 35.31, 26.24, 21.53, 21.33. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{37}H_{32}N_2O_3SNa$: 607.2026; Found, 607.2039.



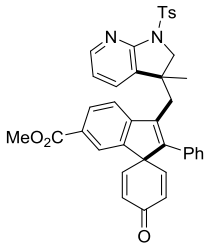
3c

6'-fluoro-3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3c): solid; m.p. 155-157 ℃; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.08 (dd, *J* = 5.2, 1.7 Hz, 1H), 7.81 (d, *J* = 8.1 Hz, 2H), 7.28 (dt, *J* = 4.5, 2.7 Hz, 3H), 7.19 (d, *J* = 8.0 Hz, 2H), 7.04 (dd, *J* = 6.6, 3.0 Hz, 2H), 6.99-6.88 (m, 3H), 6.80 (dd, *J* = 7.9, 2.4 Hz, 1H), 6.60 (dd, *J* = 7.4, 5.1 Hz, 1H), 6.51 (dd, *J* = 10.0, 2.8 Hz, 1H), 6.42-6.34 (m, 2H), 6.29 (dd, *J* = 10.0, 2.8 Hz, 1H), 3.94 (d, *J* = 9.7 Hz, 1H), 3.35 (d, *J* = 9.8 Hz, 1H), 3.09-2.99 (m, 2H), 2.35 (s, 3H), 1.21 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 185.84, 162.85, 160.88, 155.25, 147.67, 147.57, 147.36, 145.86, 145.83, 144.14, 143.21, 143.15, 141.59, 141.57, 138.22, 134.37, 134.21, 131.25, 131.21, 131.12, 130.53, 129.26, 129.22, 128.56, 128.54, 128.08, 127.97, 121.51, 121.45, 118.19, 115.34, 115.16, 111.52, 111.33, 61.31, 61.30, 60.11, 42.59, 35.75, 25.94, 21.51. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{36}H_{29}N_2O_3SFNa$: 611.1775; Found, 611.1776.



3d

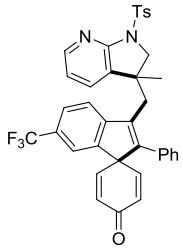
6'-chloro-3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3d): viscous oil; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.08 (dd, *J* = 5.1, 1.6 Hz, 1H), 7.80 (d, *J* = 8.1 Hz, 2H), 7.30-7.27 (m, 3H), 7.23 (dd, *J* = 8.2, 2.0 Hz, 1H), 7.19 (d, *J* = 8.0 Hz, 2H), 7.04 (dt, *J* = 6.6, 2.6 Hz, 3H), 6.94-6.83 (m, 2H), 6.59 (dd, *J* = 7.4, 5.1 Hz, 1H), 6.47 (dd, *J* = 9.9, 2.8 Hz, 1H), 6.38 (ddd, *J* = 10.1, 4.7, 1.6 Hz, 2H), 6.22 (dd, *J* = 9.9, 2.9 Hz, 1H), 3.93 (d, *J* = 9.7 Hz, 1H), 3.36 (d, *J* = 9.8 Hz, 1H), 3.12-2.94 (m, 2H), 2.35 (s, 3H), 1.21 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 185.78, 155.27, 147.42, 147.40, 147.32, 146.66, 144.17, 144.14, 142.72, 138.26, 134.20, 132.52, 131.29, 131.26, 131.18, 130.39, 129.27, 129.22, 128.59, 128.46, 128.09, 124.11, 121.45, 118.21, 61.28, 60.06, 42.61, 35.66, 25.98, 21.53. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{36}H_{29}N_2O_3SClNa$: 627.1480; Found, 627.1482.



3e

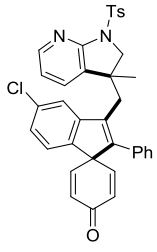
methyl

3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-4-oxo-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-diene-6'-carboxylate (3e): viscous oil; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.07 (dd, *J* = 5.1, 1.6 Hz, 1H), 7.96 (dd, *J* = 8.1, 1.6 Hz, 1H), 7.82-7.76 (m, 2H), 7.72 (d, *J* = 1.6 Hz, 1H), 7.30 (h, *J* = 2.5 Hz, 3H), 7.17 (d, *J* = 8.1 Hz, 2H), 7.07 (dd, *J* = 6.6, 3.0 Hz, 2H), 7.01 (d, *J* = 8.0 Hz, 1H), 6.94 (dd, *J* = 7.4, 1.7 Hz, 1H), 6.59 (dd, *J* = 7.4, 5.1 Hz, 1H), 6.52 (dd, *J* = 9.9, 2.9 Hz, 1H), 6.41 (ddd, *J* = 10.0, 3.0, 1.6 Hz, 2H), 6.27 (dd, *J* = 9.9, 2.8 Hz, 1H), 3.95 (d, *J* = 9.7 Hz, 1H), 3.90 (s, 3H), 3.35 (d, *J* = 9.7 Hz, 1H), 3.09 (s, 2H), 2.34 (s, 3H), 1.24 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 185.87, 166.52, 155.26, 150.00, 149.73, 147.42, 147.25, 147.18, 144.16, 141.14, 138.57, 134.13, 131.42, 131.33, 131.25, 130.29, 130.18, 129.26, 129.20, 128.61, 128.30, 128.24, 128.22, 128.06, 124.64, 120.32, 118.23, 61.42, 60.01, 52.12, 42.57, 35.77, 25.95, 21.49. HRMS-ESI (m/z) [M + H]$^{+}$ calcd for $C_{38}H_{33}N_2O_5S$: 629.2105; Found, 629.2114.



3f

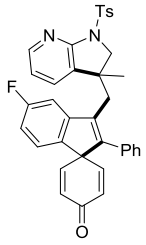
3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-phenyl-6'-(trifluoromethyl)spiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3f): ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.08 (dd, *J* = 5.1, 1.6 Hz, 1H), 7.79 (d, *J* = 8.1 Hz, 2H), 7.53 (d, *J* = 8.0 Hz, 1H), 7.30 (q, *J* = 2.6, 2.1 Hz, 4H), 7.17 (d, *J* = 8.1 Hz, 2H), 7.12-7.01 (m, 3H), 6.93 (dd, *J* = 7.4, 1.7 Hz, 1H), 6.60 (dd, *J* = 7.4, 5.1 Hz, 1H), 6.49 (dd, *J* = 10.0, 2.8 Hz, 1H), 6.41 (d, *J* = 9.9 Hz, 2H), 6.22 (dd, *J* = 10.0, 2.8 Hz, 1H), 3.94 (d, *J* = 9.8 Hz, 1H), 3.36 (d, *J* = 9.8 Hz, 1H), 3.09 (s, 2H), 2.35 (s, 3H), 1.23 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 185.63, 155.30, 149.51, 148.95, 147.48, 146.83, 146.81, 144.23, 141.85, 138.25, 134.19, 133.93, 131.64, 131.56, 131.27, 130.27, 129.27, 129.22, 128.72, 128.67, 128.46, 128.35, 128.11, 125.66, 125.63, 125.11, 120.73, 120.54, 120.51, 120.48, 118.24, 61.43, 60.06, 42.64, 35.69, 25.93, 21.50. HRMS-ESI (m/z) [M + Na]$^{+}$ calcd for $C_{37}H_{29}N_2O_3SF_3Na$: 661.1743; Found, 661.1755.



3g

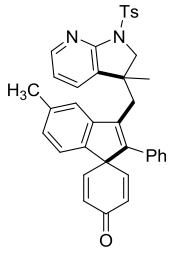
5'-chloro-3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3g): viscous oil; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.09 (dd, *J* = 5.2, 1.6 Hz, 1H), 7.83 (d, *J* = 8.1 Hz, 2H), 7.30 (dd, *J* = 5.1, 1.9 Hz, 3H), 7.22-7.13 (m, 3H), 7.10 (dd, *J* = 6.8, 2.9 Hz, 2H), 7.05 (dd, *J* = 7.4, 1.6 Hz, 1H), 6.99 (d, *J* = 8.0 Hz, 1H), 6.79 (d, *J* = 1.9 Hz, 1H), 6.68 (dd, *J* = 7.4, 5.1 Hz, 1H), 6.60 (dd, *J* = 9.9, 2.8 Hz, 1H), 6.39 (ddd, *J* = 18.6, 9.8, 1.7 Hz, 2H), 6.30 (dd, *J* = 9.9, 2.8 Hz, 1H), 3.90 (d, *J* = 9.8 Hz, 1H), 3.34 (d, *J* = 9.8 Hz, 1H), 3.00 (q, *J* = 13.8 Hz, 2H), 2.35 (s, 3H), 1.24 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 185.92, 155.25, 148.09, 147.61, 147.56, 147.09, 144.12, 139.22, 138.23, 134.24, 134.15, 134.12,

131.28, 131.15, 131.07, 130.42, 129.29, 128.60, 128.51, 128.15, 128.05, 126.45, 124.55, 120.92, 118.19, 61.13, 59.97, 42.48, 36.20, 26.03, 21.51. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{36}H_{29}N_2O_3SClNa$: 627.1480; Found, 627.1486.



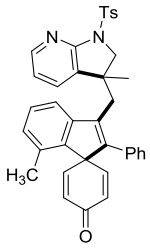
3h

5'-fluoro-3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3h): solid; m.p. 261-263 ℃; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.08 (dd, *J* = 5.2, 1.6 Hz, 1H), 7.87-7.76 (m, 2H), 7.33-7.23 (m, 3H), 7.19 (d, *J* = 8.1 Hz, 2H), 7.07 (dd, *J* = 6.6, 2.9 Hz, 2H), 7.05-6.96 (m, 2H), 6.89 (td, *J* = 8.6, 2.4 Hz, 1H), 6.62 (ddd, *J* = 16.0, 8.2, 3.8 Hz, 2H), 6.58-6.48 (m, 1H), 6.40-6.34 (m, 2H), 6.32 (dd, *J* = 10.1, 2.8 Hz, 1H), 3.91 (d, *J* = 9.8 Hz, 1H), 3.35 (d, *J* = 9.8 Hz, 1H), 3.02 (s, 2H), 2.35 (s, 3H), 1.24 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 185.97, 164.04, 162.08, 155.23, 148.29, 147.94, 147.88, 147.68, 147.61, 147.47, 144.12, 138.38, 138.35, 136.18, 136.16, 134.25, 134.16, 131.21, 130.97, 130.90, 130.46, 129.28, 128.57, 128.40, 128.08, 128.05, 124.73, 124.66, 118.20, 113.42, 113.23, 108.04, 107.85, 60.96, 59.99, 42.52, 35.95, 26.11, 21.49. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{36}H_{29}N_2O_3SFNa$: 611.1775; Found, 611.1789.



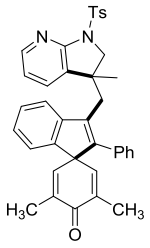
3i

5'-methyl-3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3i): solid; m.p. 124-126 ℃; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.08 (dd, *J* = 5.1, 1.6 Hz, 1H), 7.80 (d, *J* = 8.2 Hz, 2H), 7.28 (dt, *J* = 6.9, 4.2 Hz, 3H), 7.17 (d, *J* = 8.1 Hz, 2H), 7.11-7.00 (m, 3H), 7.01-6.87 (m, 2H), 6.82 (s, 1H), 6.61 (dd, *J* = 7.4, 5.1 Hz, 1H), 6.50 (dd, *J* = 10.0, 2.8 Hz, 1H), 6.36 (dd, *J* = 10.0, 2.2 Hz, 2H), 6.24 (dd, *J* = 9.9, 2.8 Hz, 1H), 3.95 (d, *J* = 9.7 Hz, 1H), 3.37 (d, *J* = 9.7 Hz, 1H), 3.15-2.91 (m, 2H), 2.35 (d, *J* = 2.9 Hz, 6H), 1.19 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.25, 155.21, 148.78, 148.67, 147.24, 146.27, 145.88, 144.01, 139.06, 138.36, 137.74, 134.78, 134.28, 131.34, 130.84, 130.69, 130.58, 129.25, 128.59, 128.49, 128.06, 127.85, 127.40, 123.29, 121.52, 118.02, 61.33, 60.13, 42.67, 35.34, 25.85, 21.51. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{37}H_{32}N_2O_3SNa$: 607.2026; Found, 607.2039.



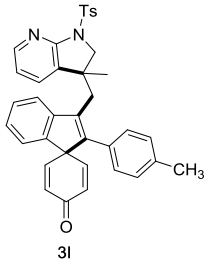
3j

7'-methyl-3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3j): viscous oil; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.07 (dd, *J* = 5.2, 1.5 Hz, 1H), 7.77 (d, *J* = 8.1 Hz, 2H), 7.30-7.21 (m, 4H), 7.16 (d, *J* = 8.0 Hz, 2H), 7.01 (d, *J* = 7.6 Hz, 1H), 6.98-6.88 (m, 3H), 6.85 (dd, *J* = 7.4, 1.6 Hz, 1H), 6.58 (dd, *J* = 7.4, 5.1 Hz, 1H), 6.49-6.35 (m, 3H), 6.14 (dd, *J* = 9.9, 2.7 Hz, 1H), 3.92 (d, *J* = 9.7 Hz, 1H), 3.41 (d, *J* = 9.7 Hz, 1H), 3.09-2.91 (m, 2H), 2.35 (s, 3H), 2.14 (s, 3H), 1.20 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 185.88, 155.16, 147.27, 147.20, 147.10, 145.99, 145.30, 143.96, 139.56, 138.49, 135.07, 134.36, 134.13, 132.20, 132.09, 131.30, 130.72, 129.20, 129.03, 128.96, 128.46, 128.34, 128.10, 127.83, 118.35, 118.18, 61.93, 60.13, 42.60, 35.39, 26.44, 21.52, 16.89. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{37}H_{32}N_2O_3SNa$: 607.2026; Found, 607.2028.



3k

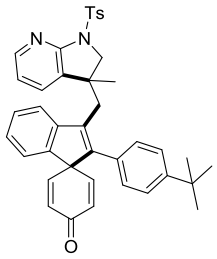
3,5-dimethyl-3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3k): solid; m.p. 293-295℃; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.07 (dt, *J* = 5.1, 1.7 Hz, 1H), 7.78 (dd, *J* = 8.3, 1.6 Hz, 2H), 7.31-7.23 (m, 4H), 7.21-7.13 (m, 3H), 7.09-6.97 (m, 4H), 6.94 (dd, *J* = 7.4, 1.7 Hz, 1H), 6.61 (dd, *J* = 7.4, 5.1 Hz, 1H), 6.25 (dd, *J* = 2.9, 1.5 Hz, 1H), 6.08 (dt, *J* = 2.9, 1.5 Hz, 1H), 3.89 (d, *J* = 9.7 Hz, 1H), 3.40 (d, *J* = 9.7 Hz, 1H), 3.05 (d, *J* = 1.7 Hz, 2H), 2.33 (d, *J* = 2.8 Hz, 3H), 1.88 (d, *J* = 1.5 Hz, 3H), 1.83 (d, *J* = 1.5 Hz, 3H), 1.18 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 187.39, 155.12, 147.19, 147.13, 145.60, 143.91, 142.81, 142.62,141.92, 138.02, 136.87, 134.96, 134.27, 131.31, 130.89, 129.18, 128.52, 128.33, 128.05, 127.85, 127.63, 126.22, 123.30, 120.49, 118.04, 61.49, 60.35, 42.71, 35.10, 26.04, 21.46, 16.23, 16.10. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{38}H_{34}N_2O_3SNa$: 621.2182; Found, 621.2189.



3l

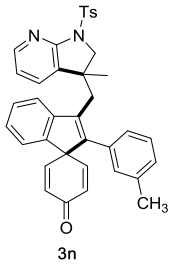
3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-(p-tolyl)spiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3l): viscous oil; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.07 (dd, *J* = 5.2, 1.6 Hz, 1H), 7.78

(d, *J* = 8.0 Hz, 2H), 7.28 (t, *J* = 7.5 Hz, 1H), 7.21 (t, *J* = 7.4 Hz, 1H), 7.16 (d, *J* = 8.0 Hz, 2H), 7.08 (d, *J* = 7.6 Hz, 3H), 7.04 (d, *J* = 7.6 Hz, 1H), 6.97-6.85 (m, 3H), 6.57 (dd, *J* = 7.4, 5.1 Hz, 1H), 6.45 (dd, *J* = 9.7, 2.8 Hz, 1H), 6.41-6.31 (m, 2H), 6.22 (dd, *J* = 9.7, 2.8 Hz, 1H), 3.94 (d, *J* = 9.7 Hz, 1H), 3.40 (d, *J* = 9.7 Hz, 1H), 3.18-2.94 (m, 2H), 2.34 (d, *J* = 2.0 Hz, 6H), 1.21 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.25, 155.21, 148.64, 148.53, 147.26, 146.31, 145.93, 144.00, 140.82, 138.75, 137.71, 134.37, 131.69, 131.34, 130.90, 130.79, 130.73, 129.24, 128.40, 128.34, 128.11, 126.48, 123.64, 120.61, 118.07, 61.70, 60.21, 42.77, 35.22, 26.23, 21.54, 21.24. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{37}H_{32}N_2O_3SNa$: 607.2026; Found, 607.2047.



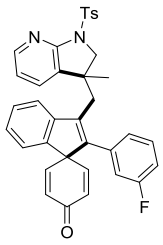
3m

2'-(4-(tert-butyl)phenyl)-3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)spiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3m): viscous oil; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.05 (dd, *J* = 5.1, 1.6 Hz, 1H), 7.77 (d, *J* = 8.1 Hz, 2H), 7.30 (td, *J* = 7.6, 1.1 Hz, 1H), 7.28-7.19 (m, 3H), 7.14 (d, *J* = 8.1 Hz, 2H), 7.10-7.01 (m, 2H), 6.94-6.85 (m, 2H), 6.72 (dd, *J* = 7.4, 1.6 Hz, 1H), 6.44 (ddd, *J* = 23.5, 8.6, 3.9 Hz, 2H), 6.35 (ddd, *J* = 13.0, 9.8, 1.7 Hz, 2H), 6.14 (dd, *J* = 9.7, 2.8 Hz, 1H), 4.01 (d, *J* = 9.8 Hz, 1H), 3.42 (d, *J* = 9.7 Hz, 1H), 3.18 (d, *J* = 13.8 Hz, 1H), 3.02 (d, *J* = 13.9 Hz, 1H), 2.34 (s, 3H), 1.32 (s, 9H), 1.22 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.35, 155.20, 150.76, 148.85, 148.64, 147.14, 146.18, 146.06, 143.95, 140.71, 138.65, 134.40, 131.62, 131.29, 130.87, 130.67, 130.46, 129.24, 128.35, 128.06, 128.02, 126.44, 125.35, 123.61, 120.50, 118.12, 61.62, 59.95, 42.71, 35.11, 34.54, 31.21, 26.50, 21.52. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{40}H_{38}N_2O_3SNa$: 649.2495; Found, 649.2491.



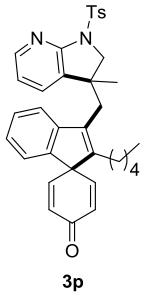
3n

3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-(m-tolyl)spiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3n): viscous oil; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.07 (dd, *J* = 5.1, 1.6 Hz, 1H), 7.77 (d, *J* = 8.2 Hz, 2H), 7.31 (td, *J* = 7.5, 1.2 Hz, 1H), 7.25-7.20 (m, 1H), 7.16 (dd, *J* = 8.1, 6.1 Hz, 3H), 7.12-7.03 (m, 3H), 6.89-6.80 (m, 2H), 6.76 (s, 1H), 6.55 (dd, *J* = 7.4, 5.1 Hz, 1H), 6.45-6.30 (m, 3H), 6.22 (dd, *J* = 9.9, 2.8 Hz, 1H), 3.93 (d, *J* = 9.7 Hz, 1H), 3.41 (d, *J* = 9.7 Hz, 1H), 3.11 (d, *J* = 13.8 Hz, 1H), 3.04 (d, *J* = 13.7 Hz, 1H), 2.34 (s, 3H), 2.29 (s, 3H), 1.23 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.23, 155.24, 148.58, 148.44, 147.14, 146.36, 145.93, 143.98, 140.83, 138.82, 138.02, 134.57, 134.37, 131.34, 130.87, 130.75, 130.59, 129.24, 129.04, 128.76, 128.37, 128.10, 126.54, 125.62, 123.66, 120.63, 118.03, 61.67, 60.22, 42.76, 35.24, 26.38, 21.53, 21.42. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{37}H_{32}N_2O_3SNa$: 607.2026; Found, 607.2020.



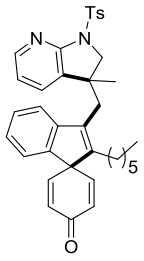
3o

2'-(3-fluorophenyl)-3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)spiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3o): solid; m.p. 93-94 ℃; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.08 (dd, *J* = 5.2, 1.6 Hz, 1H), 7.78 (d, *J* = 8.1 Hz, 2H), 7.33 (t, *J* = 7.5 Hz, 1H), 7.28-7.21 (m, 2H), 7.15 (dd, *J* = 15.2, 7.8 Hz, 3H), 7.10 (d, *J* = 7.4 Hz, 1H), 6.95 (td, *J* = 8.5, 2.5 Hz, 1H), 6.87 (dd, *J* = 7.4, 1.6 Hz, 1H), 6.80 (d, *J* = 7.7 Hz, 1H), 6.65 (dt, *J* = 9.9, 2.0 Hz, 1H), 6.57 (dd, *J* = 7.4, 5.1 Hz, 1H), 6.39 (dtd, *J* = 21.0, 9.8, 2.2 Hz, 3H), 6.24 (dd, *J* = 9.9, 2.8 Hz, 1H), 4.01 (d, *J* = 9.7 Hz, 1H), 3.38 (d, *J* = 9.8 Hz, 1H), 3.08 (q, *J* = 13.8 Hz, 2H), 2.35 (s, 3H), 1.27 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 185.98, 163.40, 161.44, 155.30, 148.09, 147.94, 147.36, 145.51, 144.45, 144.43, 144.09, 140.87, 139.71, 136.74, 136.68, 134.32, 131.22, 131.08, 131.02, 130.27, 130.20, 130.13, 129.28, 129.22, 128.47, 128.09, 126.94, 124.31, 124.28, 123.75, 120.80, 118.15, 115.55, 115.37, 114.97, 114.80, 61.52, 59.99, 42.63, 35.58, 26.26, 21.52. HRMS-ESI (m/z) [M + K]$^{+}$ calcd for $C_{36}H_{29}N_2O_3SFK$: 627.1515; Found, 627.1512.



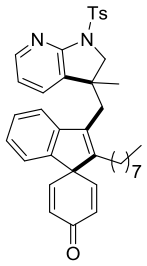
3p

3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-pentylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3p): solid; m.p. 245-247 ℃; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.18 (dd, *J* = 5.1, 1.7 Hz, 1H), 7.93 (d, *J* = 8.0 Hz, 2H), 7.24 (d, *J* = 7.7 Hz, 3H), 7.18-7.08 (m, 2H), 7.01 (dd, *J* = 22.5, 7.5 Hz, 2H), 6.76 (dd, *J* = 7.4, 5.1 Hz, 1H), 6.48 (ddd, *J* = 22.3, 9.8, 1.7 Hz, 2H), 6.35 (dd, *J* = 9.9, 2.8 Hz, 1H), 6.21 (dd, *J* = 9.8, 2.8 Hz, 1H), 4.19 (d, *J* = 9.7 Hz, 1H), 3.50 (d, *J* = 9.7 Hz, 1H), 2.96 (d, *J* = 14.0 Hz, 1H), 2.86 (d, *J* = 13.9 Hz, 1H), 2.37 (s, 3H), 1.93-1.80 (m, 1H), 1.70 (ddd, *J* = 14.2, 10.5, 5.6 Hz, 1H), 1.45 (s, 3H), 1.27-1.11 (m, 6H), 0.83 (t, *J* = 7.2 Hz, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.36, 155.35, 149.44, 149.16, 147.60, 147.31, 146.11, 144.16, 140.62, 137.42, 134.52, 131.61, 130.85, 130.72, 130.52, 129.33, 128.16, 128.12, 125.77, 123.38, 120.08, 117.82, 61.05, 60.90, 42.69, 35.72, 31.92, 29.05, 26.89, 25.13, 22.13, 21.51, 13.84. HRMS-ESI (m/z) [M + Na]$^{+}$ calcd for $C_{35}H_{36}N_2O_3SNa$: 587.2339; Found, 587.2366.



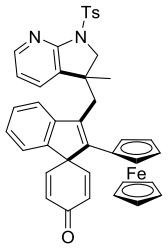
3q

2'-hexyl-3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)spiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3q): solid; m.p. 234-235 ℃; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.24-8.13 (m, 1H), 7.93 (d, *J* = 8.0 Hz, 2H), 7.24 (d, *J* = 7.7 Hz, 3H), 7.14 (t, *J* = 7.3 Hz, 2H), 7.03 (d, *J* = 7.4 Hz, 1H), 6.97 (d, *J* = 7.6 Hz, 1H), 6.76 (dd, *J* = 7.4, 5.1 Hz, 1H), 6.55-6.41 (m, 2H), 6.34 (dd, *J* = 9.9, 2.9 Hz, 1H), 6.20 (dd, *J* = 9.8, 2.8 Hz, 1H), 4.19 (d, *J* = 9.6 Hz, 1H), 3.50 (d, *J* = 9.7 Hz, 1H), 2.96 (d, *J* = 13.9 Hz, 1H), 2.86 (d, *J* = 13.9 Hz, 1H), 2.37 (s, 3H), 1.87 (td, *J* = 10.0, 5.1 Hz, 1H), 1.72 (ddd, *J* = 14.7, 10.2, 5.2 Hz, 1H), 1.44 (s, 3H), 1.27-1.14 (m, 8H), 0.85 (t, *J* = 7.2 Hz, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.38, 155.38, 149.46, 149.17, 147.63, 147.34, 146.13, 144.17, 140.64, 137.42, 134.56, 131.60, 130.88, 130.74, 130.54, 129.35, 128.18, 128.14, 125.79, 123.41, 120.08, 117.83, 61.05, 60.92, 42.71, 35.75, 31.32, 29.52, 29.38, 27.04, 25.18, 22.44, 21.53, 13.98. HRMS-ESI (m/z) [M + Na]$^{+}$ calcd for $C_{36}H_{38}N_2O_3SNa$: 601.2495; Found, 601.2521.



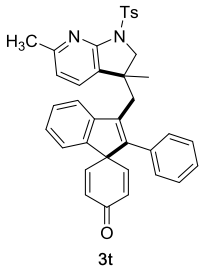
3r

3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-octylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3r): solid; m.p. 207-208 ℃; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.18 (dd, *J* = 5.1, 1.7 Hz, 1H), 7.97-7.88 (m, 2H), 7.23 (dd, *J* = 8.5, 7.0 Hz, 3H), 7.19-7.10 (m, 2H), 7.03 (d, *J* = 7.4 Hz, 1H), 6.97 (d, *J* = 7.6 Hz, 1H), 6.76 (dd, *J* = 7.3, 5.1 Hz, 1H), 6.48 (ddd, *J* = 22.4, 9.9, 1.7 Hz, 2H), 6.35 (dd, *J* = 9.8, 2.8 Hz, 1H), 6.21 (dd, *J* = 9.9, 2.8 Hz, 1H), 4.19 (d, *J* = 9.7 Hz, 1H), 3.50 (d, *J* = 9.6 Hz, 1H), 2.96 (d, *J* = 14.0 Hz, 1H), 2.86 (d, *J* = 13.9 Hz, 1H), 2.37 (s, 3H), 1.93-1.83 (m, 1H), 1.76-1.68 (m, 1H), 1.45 (s, 3H), 1.27-1.12 (m, 12H), 0.87 (t, *J* = 7.1 Hz, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.34, 155.34, 149.45, 149.17, 147.58, 147.28, 146.10, 144.14, 140.59, 137.40, 134.52, 131.59, 130.87, 130.69, 130.50, 129.32, 128.14, 128.10, 125.76, 123.36, 120.05, 117.82, 60.99, 60.89, 42.67, 35.73, 31.69, 29.82, 29.38, 29.08, 29.04, 27.02, 25.16, 22.54, 21.50, 14.01. HRMS-ESI (m/z) [M + H]$^{+}$ calcd for $C_{38}H_{43}N_2O_3S$: 607.2989; Found, 607.3008.



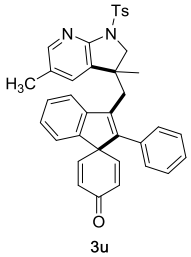
3s

3'-((3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-ferrocenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3s): viscous oil; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 8.09 (d, *J* = 5.2 Hz, 1H), 7.90 (d, *J* = 7.9 Hz, 2H), 7.18 (dd, *J* = 30.3, 7.4 Hz, 5H), 6.98 (d, *J* = 6.5 Hz, 1H), 6.82 (d, *J* = 6.6 Hz, 1H), 6.73-6.47 (m, 5H), 4.46 (s, 1H), 4.37-4.14 (m, 4H), 4.04 (s, 5H), 3.51 (d, *J* = 9.5 Hz, 1H), 3.36 (d, *J* = 14.0 Hz, 1H), 3.12 (d, *J* = 14.1 Hz, 1H), 2.36 (s, 3H), 1.44 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.37, 155.19, 152.14, 151.66, 147.58, 146.72, 144.68, 144.07, 136.02, 134.70, 131.65, 131.22, 129.80, 129.39, 129.33, 128.34, 128.01, 126.03, 123.17, 119.81, 117.85, 79.33, 69.60, 69.35, 68.98, 67.90, 67.84, 61.05, 60.87, 42.26, 36.53, 26.32, 21.55. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{40}H_{34}N_2O_3SFeNa$: 701.1532; Found, 701.1550.



3t

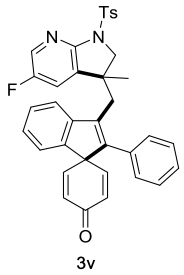
3'-((3,6-dimethyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3t): viscous oil; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.80 (d, *J* = 8.2 Hz, 2H), 7.32 (td, *J* = 7.5, 1.2 Hz, 1H), 7.28-7.21 (m, 4H), 7.15 (t, *J* = 7.8 Hz, 3H), 7.09 (d, *J* = 7.4 Hz, 1H), 7.01 (dd, *J* = 6.6, 3.0 Hz, 2H), 6.74 (d, *J* = 7.5 Hz, 1H), 6.45 (dd, *J* = 9.8, 2.8 Hz, 1H), 6.41-6.30 (m, 3H), 6.25 (dd, *J* = 9.8, 2.8 Hz, 1H), 3.92 (d, *J* = 9.7 Hz, 1H), 3.39 (d, *J* = 9.7 Hz, 1H), 3.05 (q, *J* = 13.7 Hz, 2H), 2.41 (s, 3H), 2.35 (s, 3H), 1.18 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.20, 156.57, 154.65, 148.66, 148.45, 145.99, 145.94, 143.86, 140.82, 139.21, 134.73, 134.40, 131.15, 130.91, 130.74, 128.99, 128.47, 128.45, 128.40, 128.34, 127.84, 127.17, 126.57, 123.64, 120.78, 117.27, 61.66, 60.40, 42.54, 35.24, 26.07, 24.10, 21.53. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{37}H_{32}N_2O_3SNa$: 607.2026; Found, 607.2044.



3u

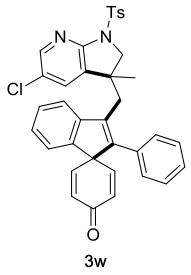
3'-((3,5-dimethyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3u): viscous oil; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.87 (d, *J* = 2.0 Hz, 1H), 7.80-7.69 (m, 2H), 7.34 (td, *J* = 7.6, 1.2 Hz, 1H), 7.30-7.19 (m, 4H), 7.15 (dd, *J* = 9.7, 7.7 Hz, 3H), 7.09 (d, *J* = 7.4

Hz, 1H), 7.02-6.90 (m, 2H), 6.62 (d, *J* = 2.1 Hz, 1H), 6.39 (dt, *J* = 9.9, 2.0 Hz, 2H), 6.30 (ddd, *J* = 19.1, 9.9, 2.2 Hz, 2H), 3.95 (d, *J* = 9.8 Hz, 1H), 3.36 (d, *J* = 9.8 Hz, 1H), 3.12 (d, *J* = 13.8 Hz, 1H), 3.03 (d, *J* = 13.7 Hz, 1H), 2.33 (s, 3H), 1.97 (s, 3H), 1.22 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.14, 153.25, 148.57, 148.42, 147.11, 145.95, 145.82, 143.83, 140.74, 138.89, 134.55, 134.16, 132.38, 130.80, 130.74, 130.01, 129.16, 128.36, 128.26, 128.03, 127.62, 127.43, 126.55, 123.61, 120.61, 61.60, 60.19, 42.57, 35.21, 26.31, 21.47, 17.83. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{37}H_{32}N_2O_3SNa$: 607.2026; Found, 607.2042.



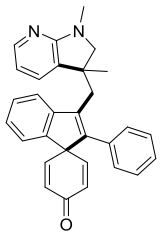
3v

3'-((5-fluoro-3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3v): solid; m.p. 158-160 ℃; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.88 (d, *J* = 2.6 Hz, 1H), 7.71 (d, *J* = 8.1 Hz, 2H), 7.39 (t, *J* = 7.5 Hz, 1H), 7.30-7.20 (m, 5H), 7.13 (dd, *J* = 22.7, 7.7 Hz, 3H), 6.99-6.89 (m, 2H), 6.47 (dd, *J* = 7.6, 2.8 Hz, 1H), 6.43-6.26 (m, 3H), 6.14 (dd, *J* = 9.8, 2.8 Hz, 1H), 4.01 (d, *J* = 9.8 Hz, 1H), 3.47 (d, *J* = 9.8 Hz, 1H), 3.16 (d, *J* = 13.9 Hz, 1H), 3.04 (d, *J* = 13.9 Hz, 1H), 2.35 (s, 3H), 1.26 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.06, 157.54, 155.55, 151.27, 148.30, 148.12, 146.25, 145.78, 144.18, 140.90, 138.47, 134.46, 134.37, 134.25, 134.00, 132.18, 132.15, 130.98, 130.84, 129.32, 128.58, 128.49, 128.19, 128.05, 128.01, 126.79, 123.82, 120.44, 119.94, 119.77, 61.58, 60.58, 42.67, 34.98, 26.48, 21.54. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{36}H_{29}N_2O_3SFNa$: 611.1775; Found, 611.1772.



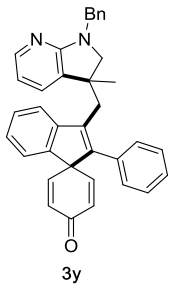
3w

3'-((5-chloro-3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3w) solid; m.p. 148-150 ℃; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.97 (t, *J* = 2.0 Hz, 1H), 7.80-7.67 (m, 2H), 7.38 (t, *J* = 7.6 Hz, 1H), 7.26 (h, *J* = 7.4 Hz, 5H), 7.17 (d, *J* = 8.0 Hz, 2H), 7.11 (d, *J* = 7.4 Hz, 1H), 6.95 (dt, *J* = 7.7, 1.8 Hz, 2H), 6.73 (t, *J* = 2.0 Hz, 1H), 6.45-6.30 (m, 3H), 6.21 (dt, *J* = 9.9, 2.2 Hz, 1H), 4.03 (dd, *J* = 9.8, 1.5 Hz, 1H), 3.44 (dd, *J* = 9.8, 1.5 Hz, 1H), 3.22-2.98 (m, 2H), 2.36 (s, 3H), 1.30-1.24 (m, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.08, 153.58, 148.28, 148.16, 146.37, 145.79, 145.73, 144.31, 140.93, 138.38, 134.21, 134.00, 131.99, 131.71, 130.97, 130.88, 129.36, 128.60, 128.46, 128.06, 128.03, 126.78, 126.11, 123.84, 120.47, 61.60, 60.51, 42.71, 35.14, 26.33, 21.55. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{36}H_{29}N_2O_3SClNa$: 627.1480; Found, 627.1490.



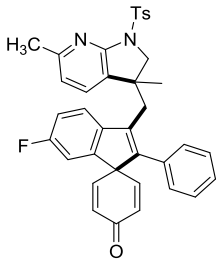
3x

3'-((1,3-dimethyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3x): solid; m.p. 167-168 ℃; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.83 (dd, *J* = 5.4, 1.6 Hz, 1H), 7.32-7.25 (m, 4H), 7.20 (tt, *J* = 5.3, 2.9 Hz, 2H), 7.16-7.05 (m, 3H), 6.82 (dd, *J* = 7.0, 1.6 Hz, 1H), 6.44 (dqd, *J* = 35.7, 9.8, 2.1 Hz, 4H), 6.27 (dd, *J* = 7.1, 5.3 Hz, 1H), 3.26 (d, *J* = 9.0 Hz, 1H), 3.09-2.95 (m, 2H), 2.86 (d, *J* = 9.0 Hz, 1H), 2.72 (s, 3H), 1.19 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.26, 162.50, 148.88, 148.72, 146.53, 146.44, 145.45, 140.74, 140.32, 135.08, 130.89, 130.85, 129.85, 129.15, 128.57, 128.42, 128.21, 127.80, 126.36, 123.46, 120.98, 112.42, 64.98, 61.71, 43.33, 35.14, 32.21, 25.12. HRMS-ESI (m/z) [M + Na]$^+$ calcd for $C_{30}H_{26}N_2ONa$: 453.1937; Found, 453.1937.



3y

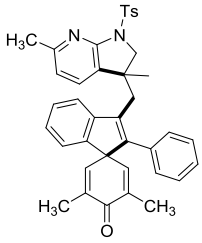
3'-((1-benzyl-3-methyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3y): viscous oil; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.88 (dd, *J* = 5.3, 1.6 Hz, 1H), 7.28-7.18 (m, 8H), 7.10 (td, *J* = 6.9, 6.4, 3.0 Hz, 4H), 6.98 (dd, *J* = 6.6, 3.0 Hz, 2H), 6.78 (dd, *J* = 6.9, 1.6 Hz, 1H), 6.41 (d, *J* = 1.6 Hz, 2H), 6.37-6.26 (m, 3H), 4.54 (d, *J* = 14.8 Hz, 1H), 4.21 (d, *J* = 14.8 Hz, 1H), 3.22 (d, *J* = 9.2 Hz, 1H), 3.00 (s, 2H), 2.84 (d, *J* = 9.3 Hz, 1H), 1.17 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.26, 161.90, 148.84, 148.61, 146.44, 146.36, 145.73, 140.82, 140.23, 137.73, 134.94, 130.89, 130.80, 129.59, 129.52, 128.54, 128.41, 128.37, 128.33, 128.21, 127.76, 127.06, 126.36, 123.51, 121.03, 112.79, 62.32, 61.67, 49.33, 43.15, 35.24, 25.93. HRMS-ESI (m/z) [M + Na]$^+$ calcd for $C_{36}H_{30}N_2ONa$: 529.2250; Found, 529.2248.



3aa

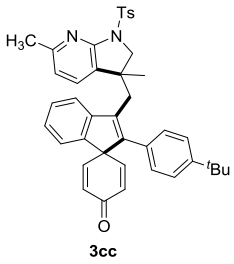
3'-((3,6-dimethyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-6'-fluoro-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3aa): viscous oil; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.89-7.79 (m, 2H), 7.27 (dt, *J* = 5.5, 2.0 Hz, 3H), 7.19 (d, *J* = 8.2 Hz, 2H), 7.03 (dt, *J* = 6.1, 1.9 Hz, 2H), 7.01-6.92 (m, 2H), 6.84-6.76

(m, 2H), 6.48 (dd, *J* = 10.2, 2.8 Hz, 1H), 6.45-6.35 (m, 3H), 6.30 (dd, *J* = 10.1, 2.8 Hz, 1H), 3.92 (d, *J* = 9.7 Hz, 1H), 3.35 (d, *J* = 9.8 Hz, 1H), 3.17-2.90 (m, 2H), 2.42 (s, 3H), 2.36 (s, 3H), 1.17 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 185.86, 162.85, 160.88, 156.70, 154.70, 147.84, 147.68, 145.67, 145.64, 143.97, 143.19, 143.12, 141.76, 141.74, 138.44, 134.45, 134.34, 131.16, 131.14, 131.03, 129.00, 128.95, 128.50, 128.43, 128.41, 127.92, 127.11, 121.65, 121.58, 117.29, 115.27, 115.09, 111.48, 111.29, 61.33, 61.32, 60.38, 42.40, 35.72, 25.83, 24.07, 21.50. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{37}H_{31}N_2O_3SFNa$: 625.1932; Found, 625.1952.



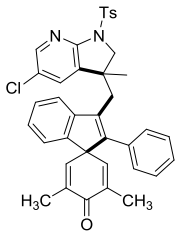
3bb

3'-((3,6-dimethyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-3,5-dimethyl-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3bb): viscous oil; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.84-7.72 (m, 2H), 7.26 (ddt, *J* = 4.5, 3.1, 1.3 Hz, 4H), 7.20 (td, *J* = 7.4, 1.1 Hz, 1H), 7.17 (d, *J* = 8.1 Hz, 2H), 7.09 (d, *J* = 7.5 Hz, 1H), 7.05 (dd, *J* = 7.7, 1.1 Hz, 1H), 7.01 (ddd, *J* = 6.4, 2.9, 1.6 Hz, 2H), 6.81 (d, *J* = 7.6 Hz, 1H), 6.45 (d, *J* = 7.5 Hz, 1H), 6.22 (dq, *J* = 2.7, 1.3 Hz, 1H), 6.07 (dq, *J* = 2.7, 1.3 Hz, 1H), 3.88 (d, *J* = 9.6 Hz, 1H), 3.40 (d, *J* = 9.7 Hz, 1H), 3.01 (d, *J* = 2.6 Hz, 2H), 2.41 (s, 3H), 2.35 (s, 3H), 1.88 (d, *J* = 1.4 Hz, 3H), 1.83 (d, *J* = 1.4 Hz, 3H), 1.15 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 187.47, 156.52, 154.70, 147.02, 145.84, 143.80, 142.92, 142.90, 141.99, 138.28, 136.94, 136.84, 135.09, 134.52, 131.23, 128.97, 128.56, 128.50, 128.28, 127.89, 127.64, 127.56, 126.26, 123.36, 120.63, 117.18, 61.56, 60.67, 42.60, 35.25, 25.90, 24.06, 21.52, 16.21, 16.17. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{39}H_{36}N_2O_3SNa$: 635.2339; Found, 635.2346.



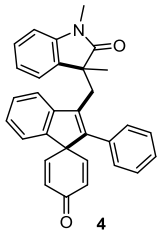
3cc

2'-(4-(tert-butyl)phenyl)-3'-((3,6-dimethyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)spiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3cc): viscous oil; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.86-7.76 (m, 2H), 7.32 (td, *J* = 7.6, 1.2 Hz, 1H), 7.26-7.21 (m, 3H), 7.14 (dd, *J* = 12.3, 7.8 Hz, 3H), 7.11-7.02 (m, 1H), 6.96-6.82 (m, 2H), 6.62 (d, *J* = 7.5 Hz, 1H), 6.46-6.26 (m, 4H), 6.19 (dd, *J* = 9.8, 2.8 Hz, 1H), 4.00 (d, *J* = 9.8 Hz, 1H), 3.40 (d, *J* = 9.7 Hz, 1H), 3.14 (d, *J* = 13.7 Hz, 1H), 3.00 (d, *J* = 13.7 Hz, 1H), 2.41 (s, 3H), 2.35 (s, 3H), 1.31 (s, 9H), 1.19 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 186.44, 156.41, 154.69, 150.70, 149.09, 148.80, 146.25, 145.99, 143.82, 140.71, 138.81, 134.58, 131.69, 131.14, 130.87, 130.60, 129.00, 128.98, 128.44, 128.32, 127.93, 127.05, 126.41, 125.21, 123.62, 120.59, 117.26, 61.68, 60.25, 42.51, 35.23, 34.53, 31.23, 26.29, 24.12, 21.57, 21.54. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{41}H_{40}N_2O_3SNa$: 663.2652; Found, 663.2657.



3dd

3'-((5-chloro-3-methyl-1-tosyl-2,3-dihydro-1H-pyrrolo[2,3-b]pyridin-3-yl)methyl)-3,5-dimethyl-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-4-one (3dd): viscous oil; ^{1}H NMR (500 MHz, Chloroform-*d*) δ 7.97 (d, *J* = 2.3 Hz, 1H), 7.73 (d, *J* = 8.1 Hz, 2H), 7.32 (t, *J* = 7.5 Hz, 1H), 7.28-7.20 (m, 4H), 7.17 (dd, *J* = 7.8, 5.0 Hz, 3H), 7.07 (d, *J* = 7.4 Hz, 1H), 6.94 (dt, *J* = 7.3, 3.7 Hz, 2H), 6.82 (d, *J* = 2.3 Hz, 1H), 6.31-6.15 (m, 1H), 6.05 (dd, *J* = 3.0, 1.6 Hz, 1H), 3.99 (d, *J* = 9.8 Hz, 1H), 3.44 (d, *J* = 9.8 Hz, 1H), 3.06 (s, 2H), 2.35 (s, 3H), 1.87 (d, *J* = 1.3 Hz, 3H), 1.84 (d, *J* = 1.3 Hz, 3H), 1.24 (s, 3H). ^{13}C NMR (126 MHz, Chloroform-*d*) δ 187.34, 153.54, 147.50, 145.68, 145.51, 144.21, 142.61, 142.46, 142.08, 137.42, 136.99, 136.97, 134.55, 134.10, 132.26, 131.74, 129.31, 128.39, 128.11, 128.05, 127.95, 127.74, 126.43, 125.96, 123.51, 120.34, 61.48, 60.85, 42.71, 35.01, 26.14, 21.50, 16.22, 16.12. HRMS-ESI (m/z) $[M + Na]^+$ calcd for $C_{38}H_{33}N_2O_3SClNa$: 655.1793; Found, 655.1798.



4

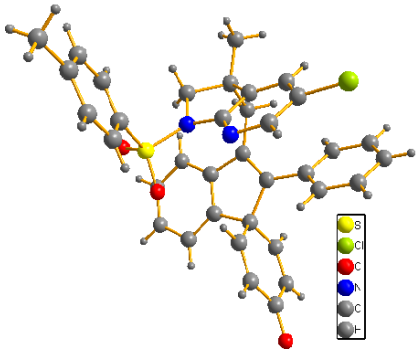
1,3-dimethyl-3-((4-oxo-2'-phenylspiro[cyclohexane-1,1'-indene]-2,5-dien-3'-yl)methyl)indolin-2-one (4): white solid; mp = 139-141 ℃; ^{1}H NMR (500 MHz, $CDCl_3$) δ 7.49 (d, *J* = 7.7 Hz, 1H), 7.34-7.24 (m, 4H), 7.15 (dtd, *J* = 14.7, 7.6, 1.2 Hz, 2H), 7.03-6.89 (m, 3H), 6.81 (td, *J* = 7.5, 1.0 Hz, 1H), 6.60 (d, *J* = 7.7 Hz, 1H), 6.48 (dd, *J* = 7.4, 1.3 Hz, 1H), 6.34 (dd, *J* = 9.9, 1.7 Hz, 1H), 6.28-6.14 (m, 2H), 5.69 (dd, *J* = 9.8, 2.7 Hz, 1H), 3.48-3.27 (m, 2H), 2.87 (s, 3H), 1.37 (s, 3H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 186.22, 180.40, 149.54, 148.51, 146.06, 144.96, 142.93, 140.24, 139.33, 134.64, 132.55, 130.75, 130.20, 128.87, 128.24, 128.09, 127.89, 127.63, 126.30, 123.98, 122.99, 122.17, 121.59, 107.30, 61.21, 48.76, 34.51, 25.95, 23.68. HRMS (ESI) calcd for $C_{31}H_{26}NO_2$ $[M+H]^+$: 444.1958, found: 444.1948.

6. References.

(1) T. T. Schempp, B. E. Daniels, S. T. Staben and C. E. Stivala, *Org. Lett.*, 2017, **19**, 3616.

(2) (a) W. Hu, H. Wang, L. Bai, J. Liu and X. Luan, *Org. Lett.*, 2018, **20**, 880; (b) L. Luo, H. Zheng, J. Liu, H. Wang, Y. Wang and X. Luan, *Org. Lett.*, 2016, **18**, 2082.

7. Crystallographic data of 3w.



Structure of 3w

Datablock:

Bond precision:	C-C = 0.0041 A	Wavelength=0.71073	
Cell:	a= 10.6142(4)	b=10.9358(5)	c=15.5922(7)
	alpha=79.650(2)	beta=72.028(1)	gamma=79.813(1)
Temperature:	293 K		
	Calculated	Reported	
Volume	1679.22(13)	1679.22(12)	
Space group	P -1	P -1	
Hall group	-P 1		
Moiety formula	C36 H29 N2O3SCl		
Sum formula	C36 H29 N2O3SCl	C36 H29 N2O3SCl	
Mr	605.12	605.12	
Dx,g cm-3	1.197	1.197	
Z	2	2	
Mu (mm-1)	0.212	0.212	
F000	632.0	632.0	
F000'	632.79		
h,k,lmax	12,13,18	12,13,18	
Nref	5908	5893	
Tmin,Tmax	0.995,0.998	0.694,0.746	
Tmin'	0.994		

Correction method= # Reported T Limits: Tmin=0.694 Tmax=0.746

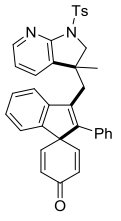
AbsCorr = NONE

Data completeness= 0.997 Theta(max)= 25.000

R(reflections)= 0.0578(4723) wR2(reflections)= 0.1790(5893)

S = 1.085 Npar= 390

8. Copies of ^{1}H and ^{13}C Spectra



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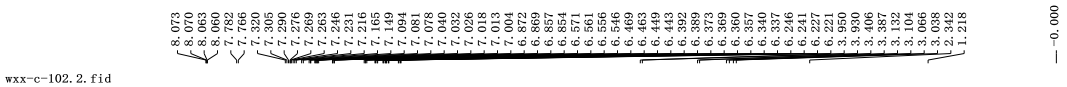

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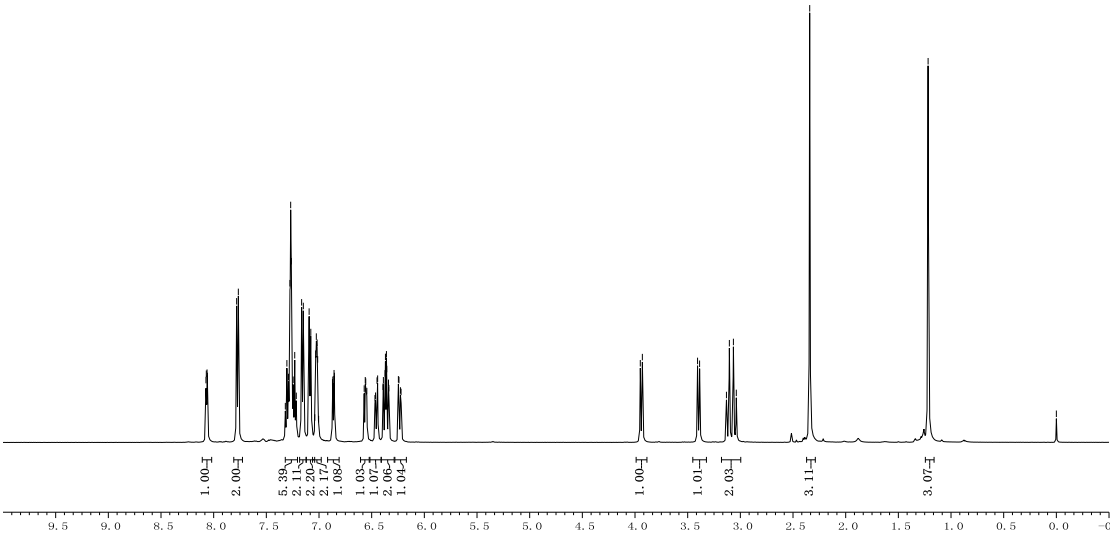

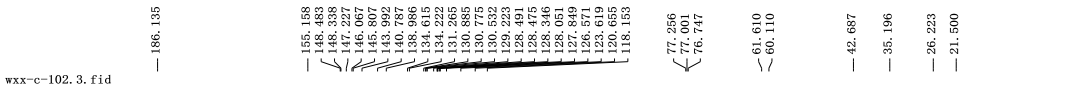

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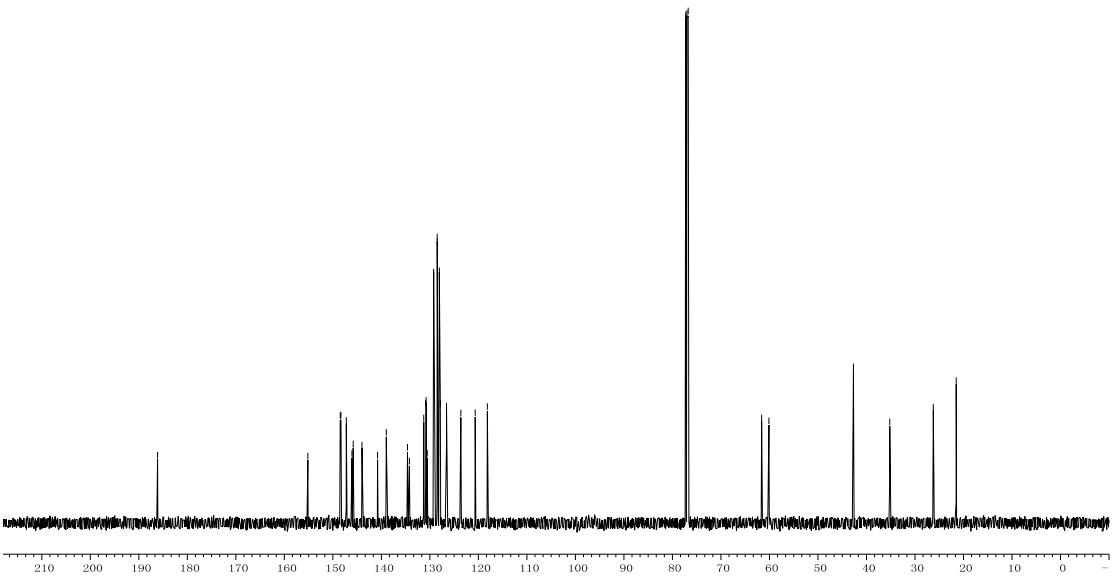

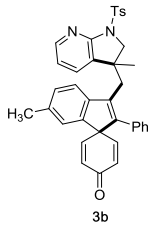

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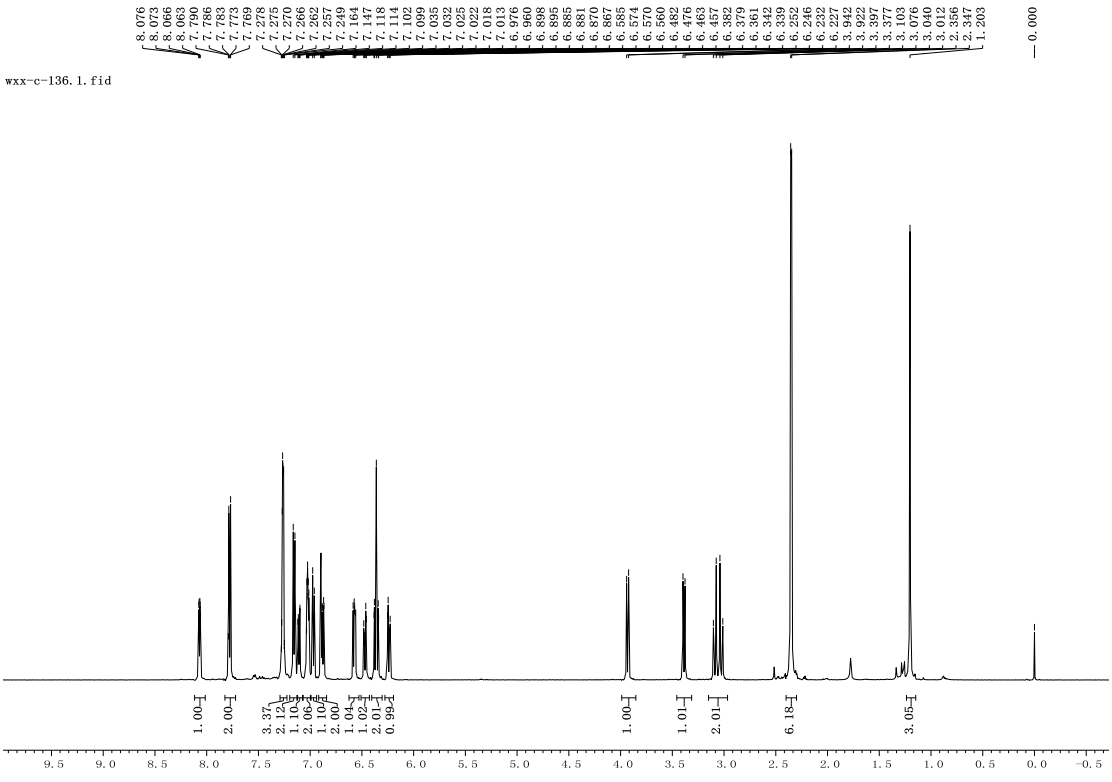

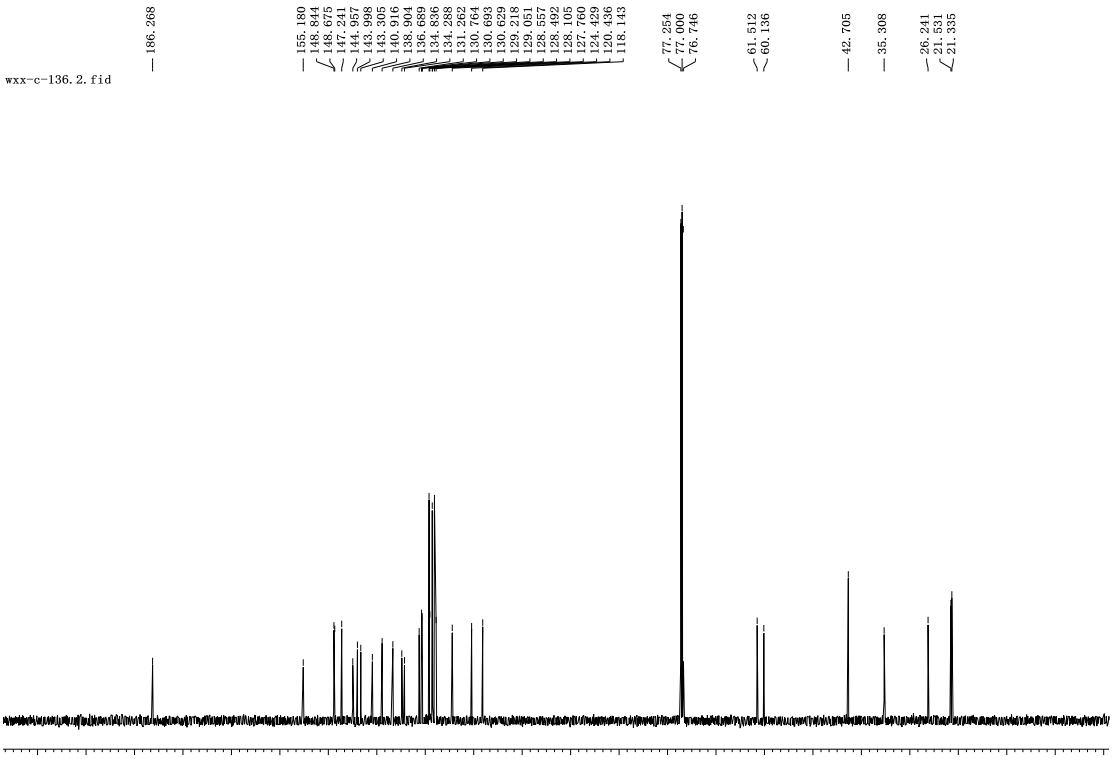

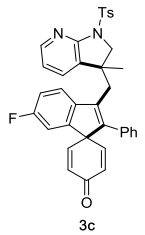

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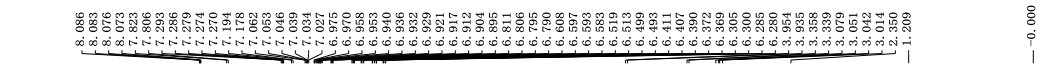

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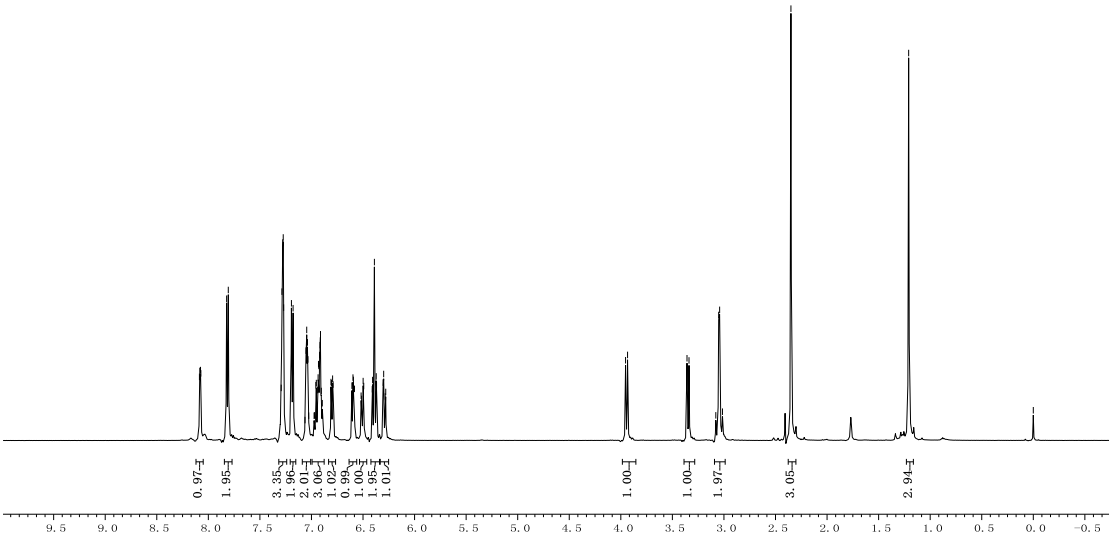

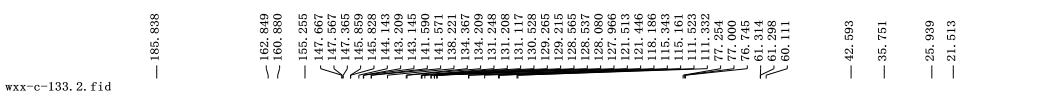

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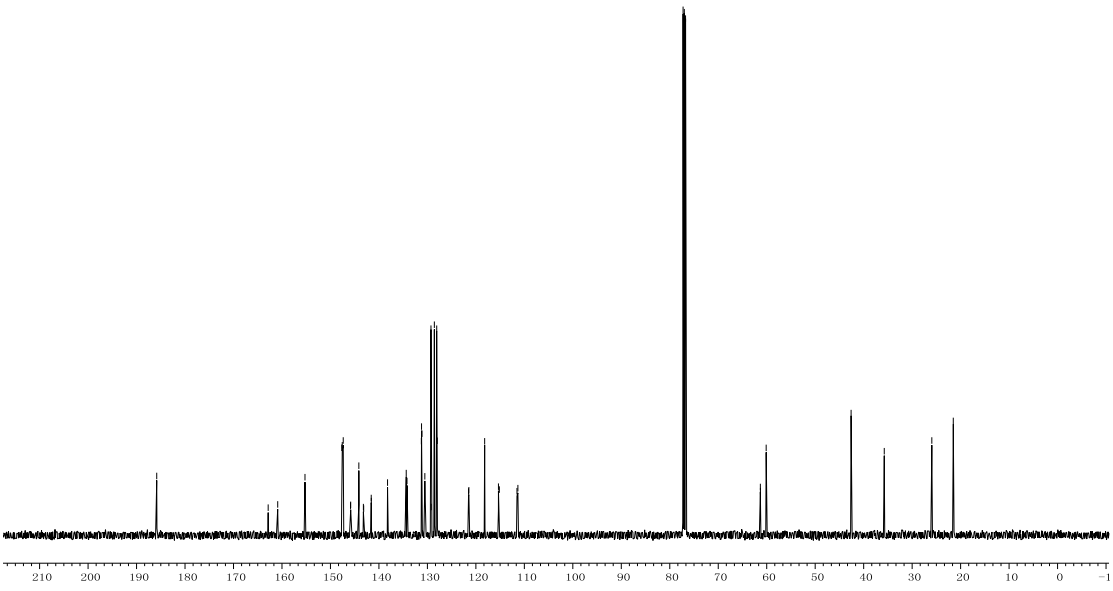

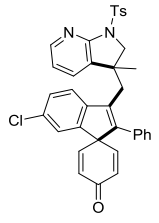

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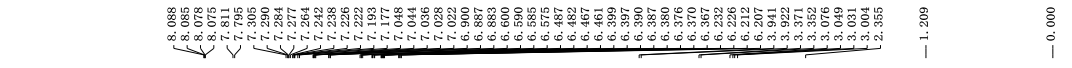

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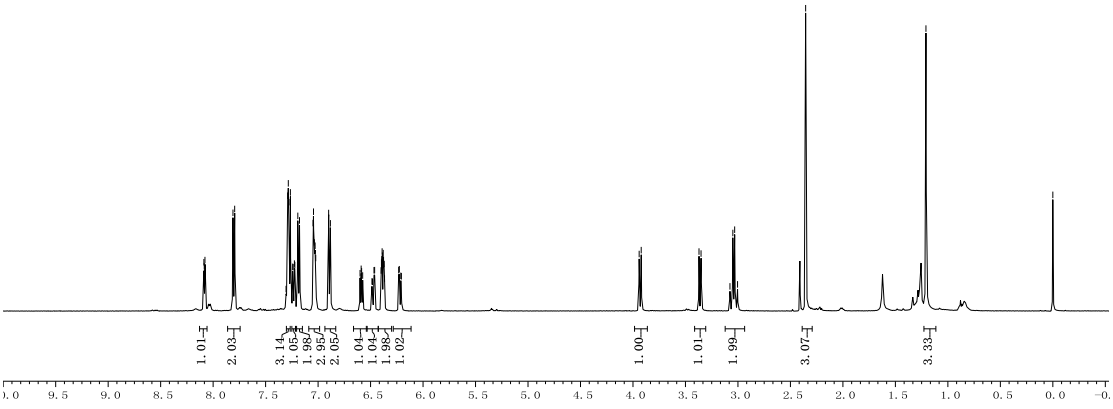

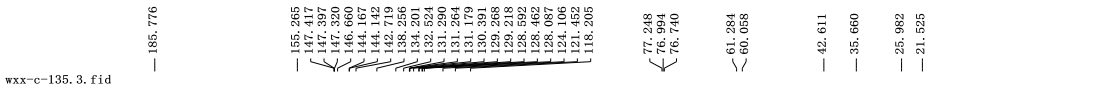

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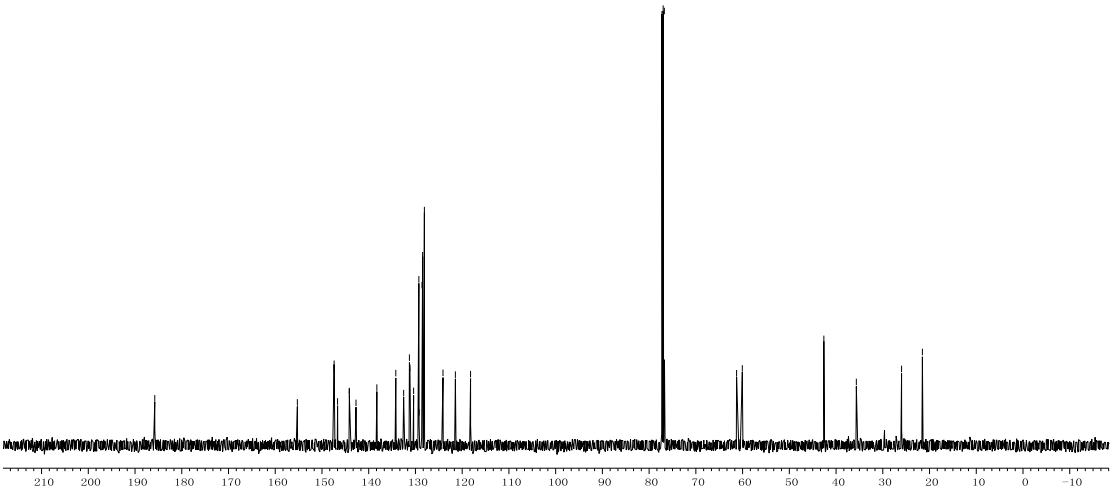

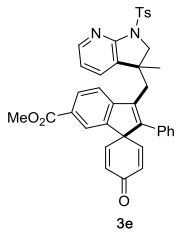

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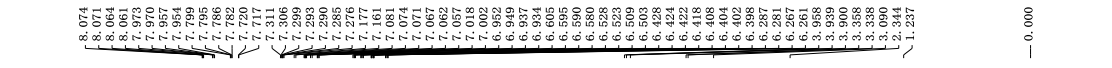

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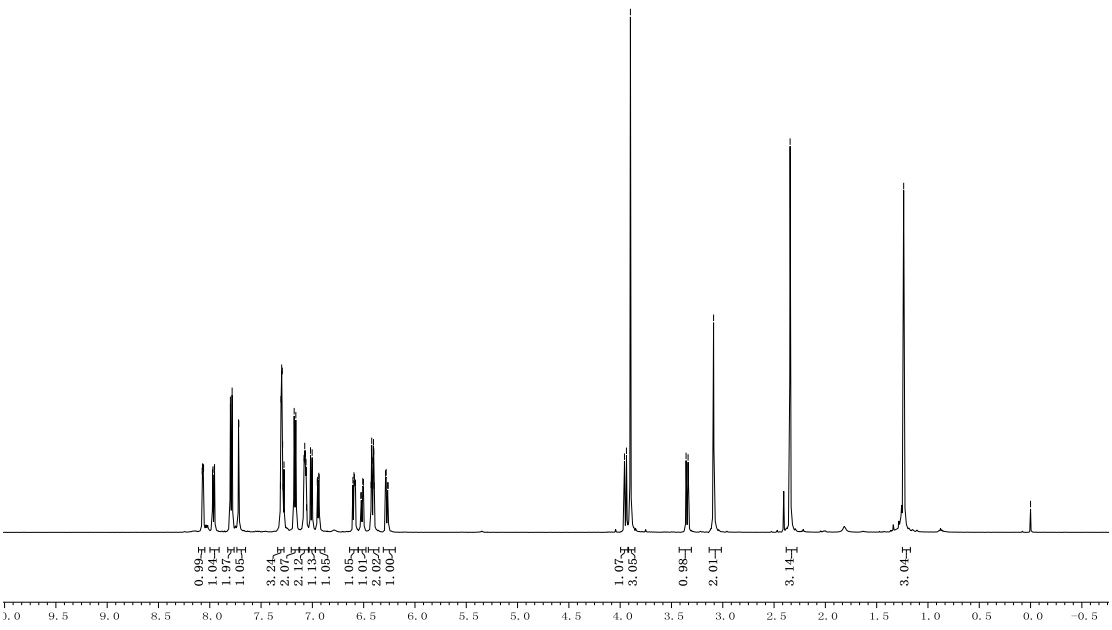

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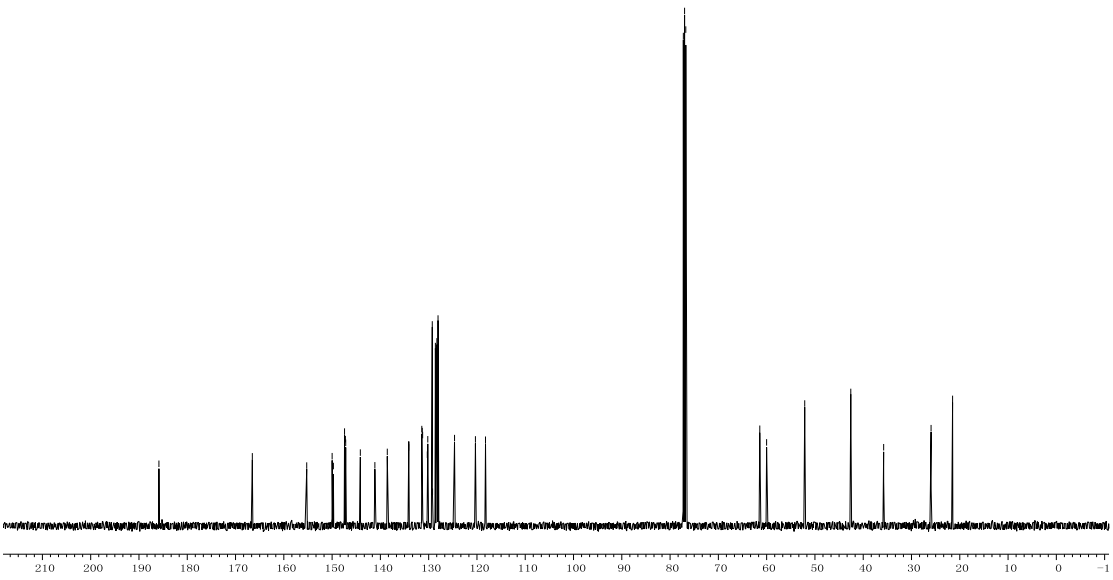

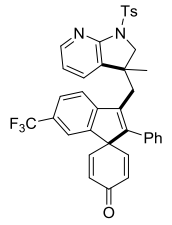

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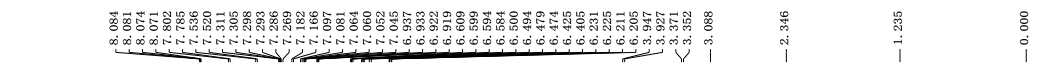

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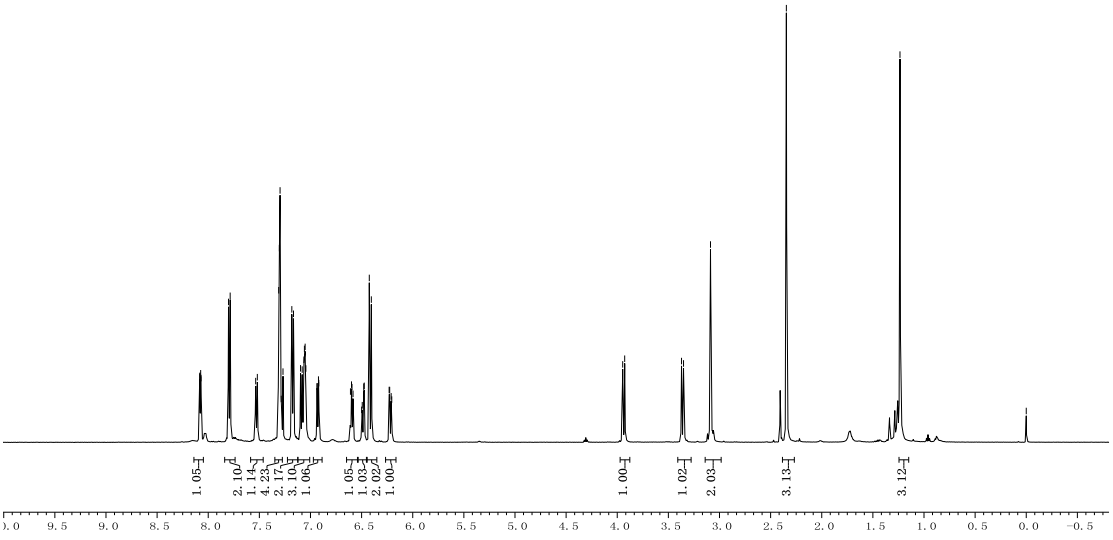

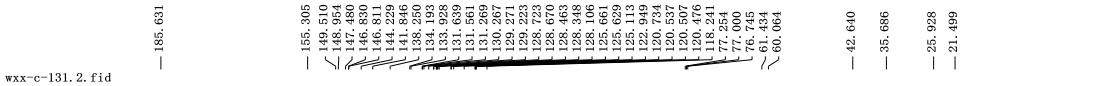

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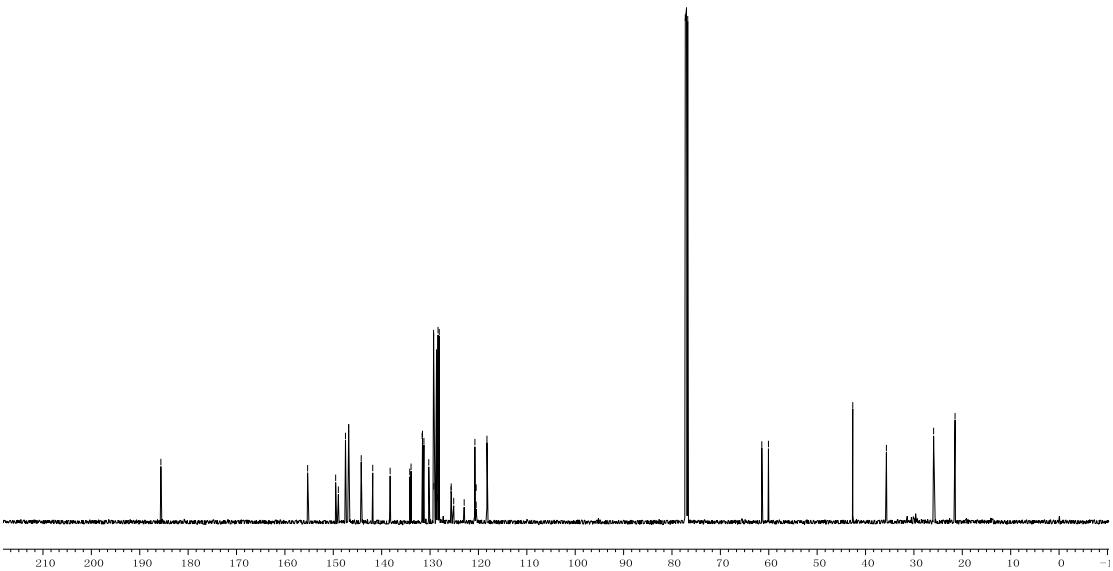

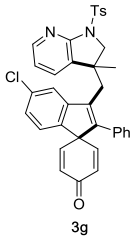
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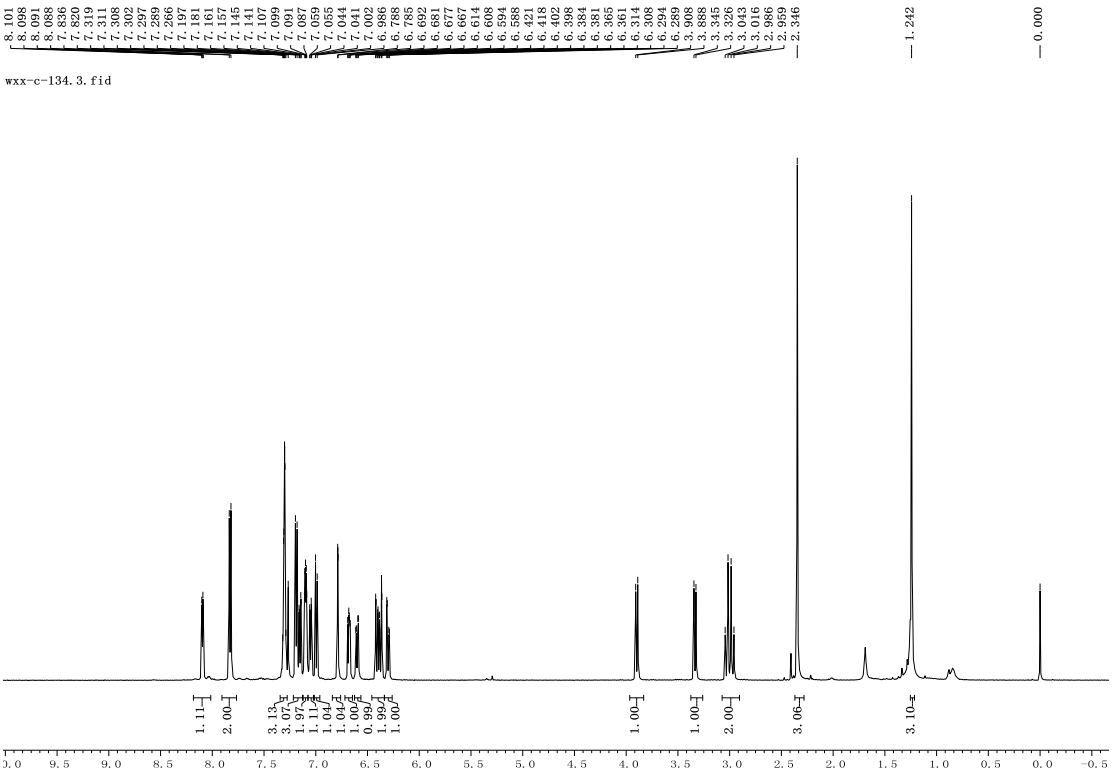
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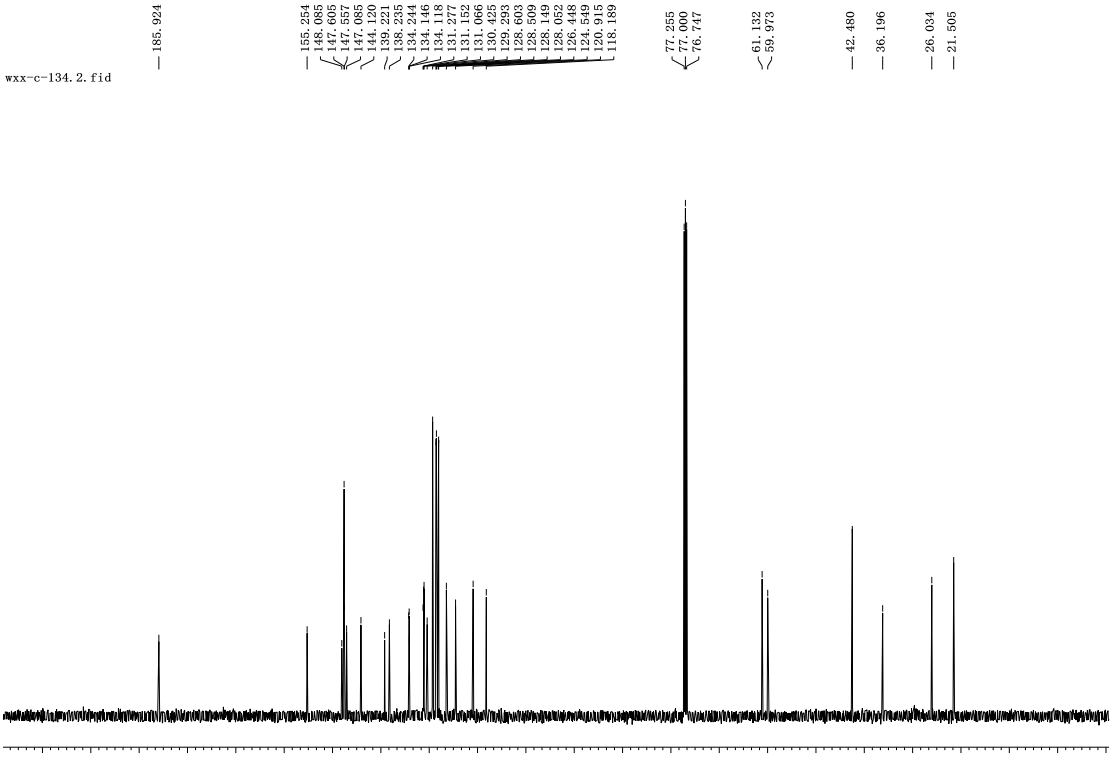
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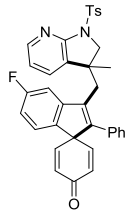

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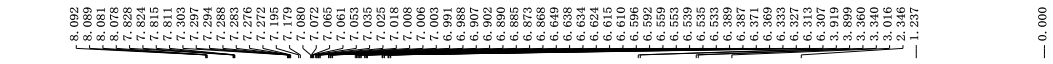

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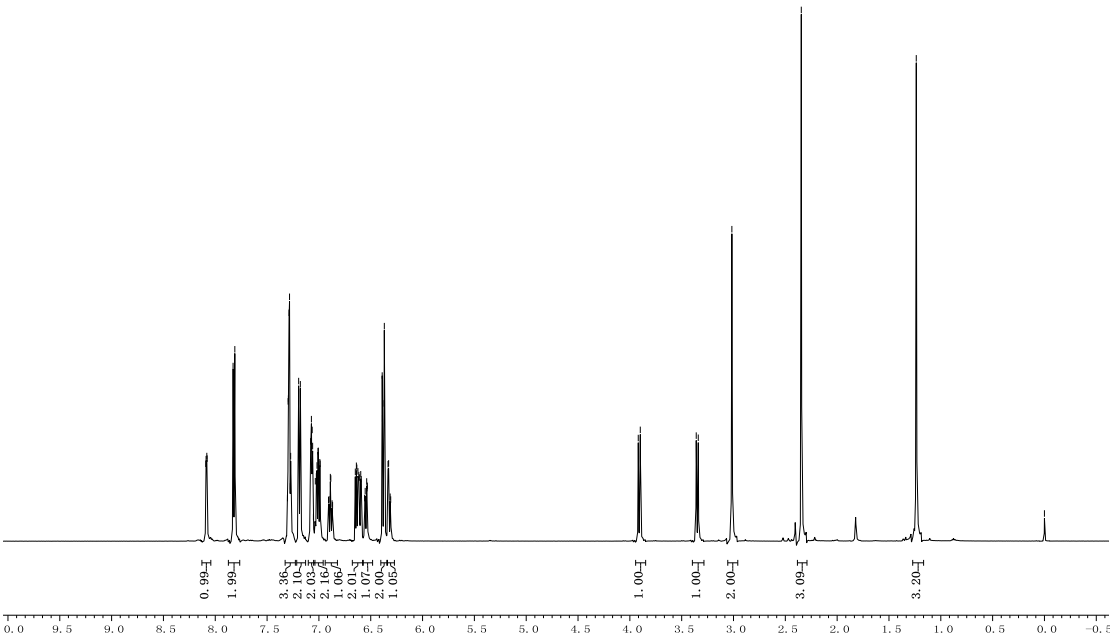

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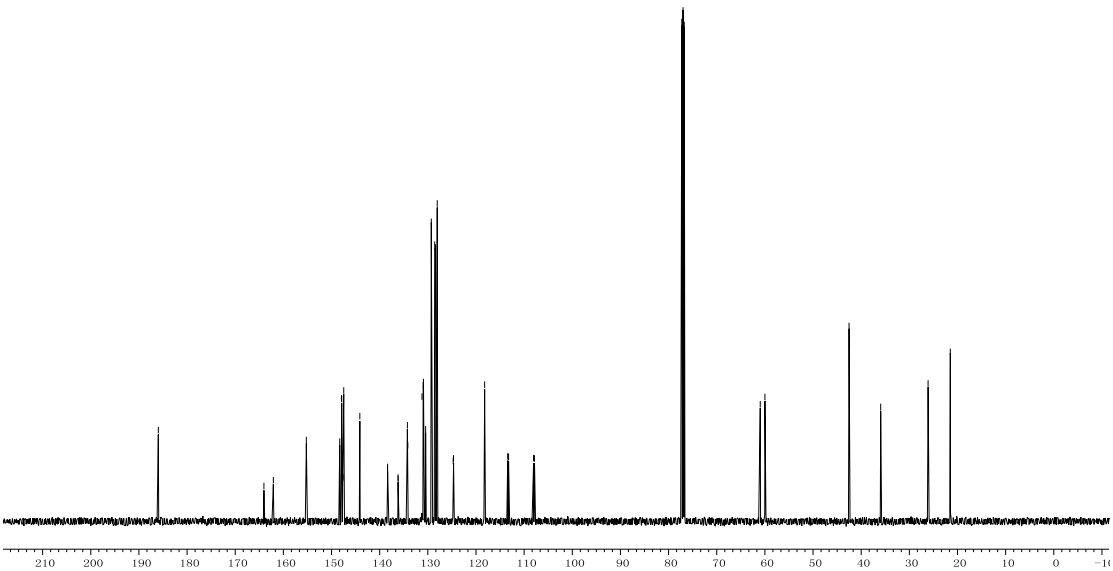

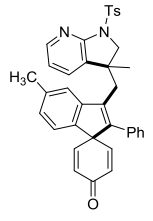

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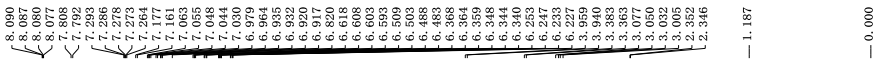

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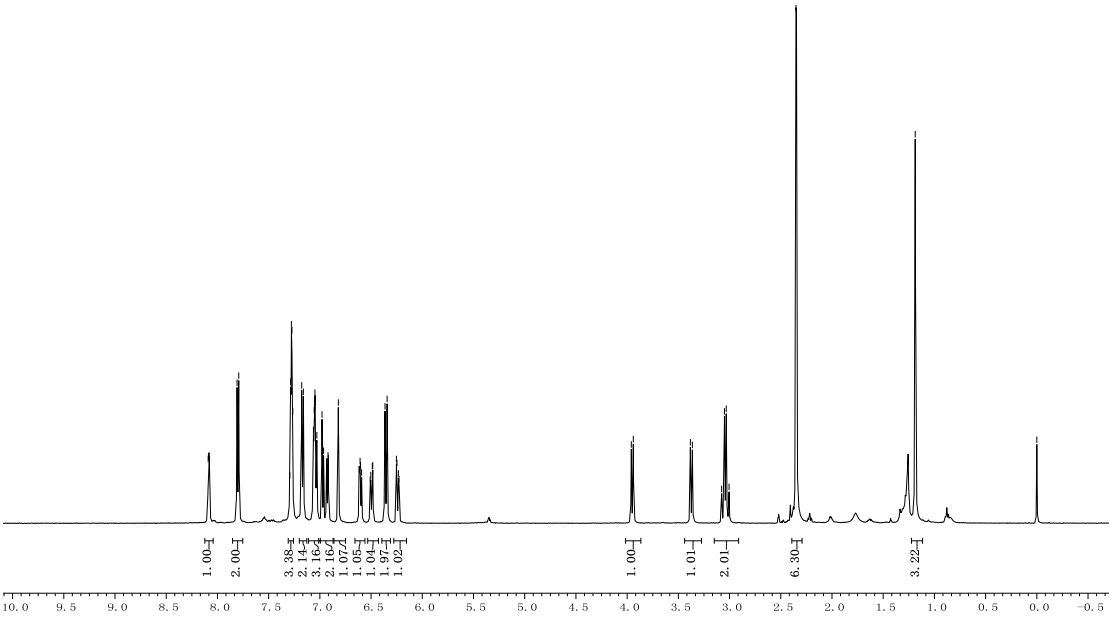

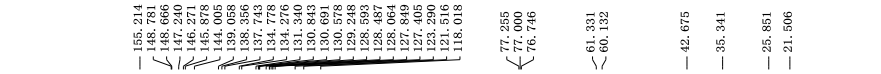

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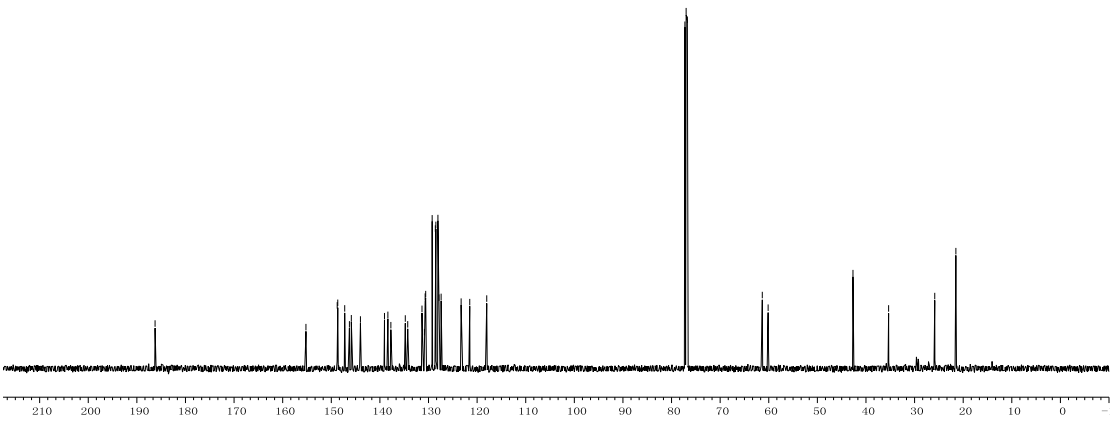

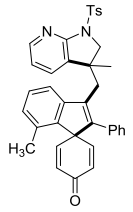

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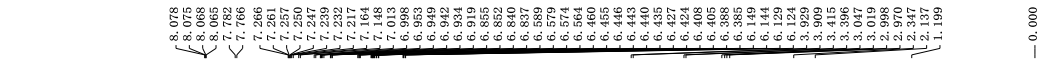

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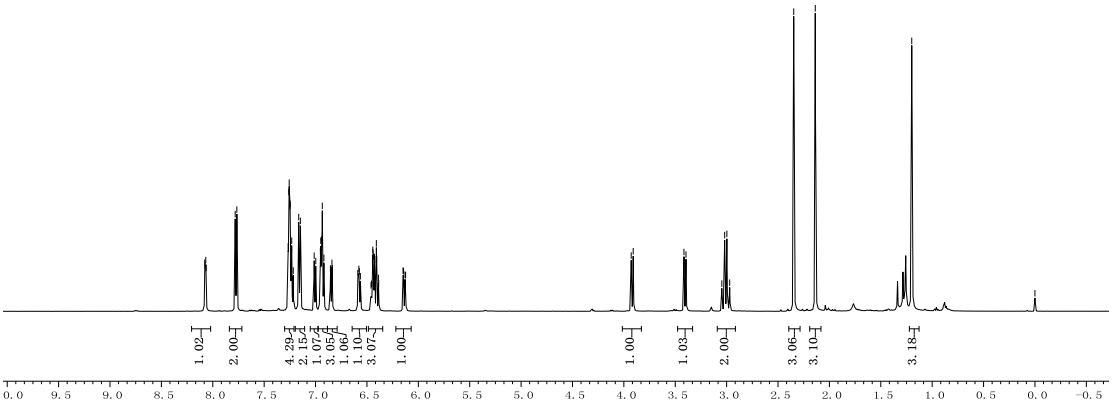

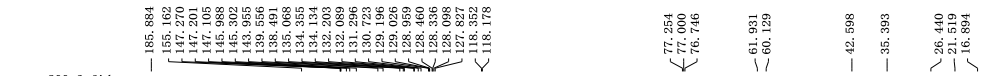

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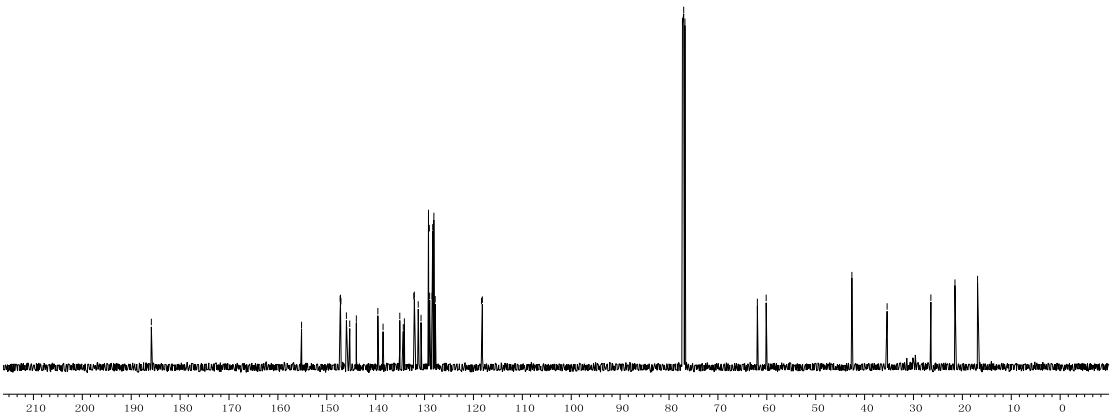

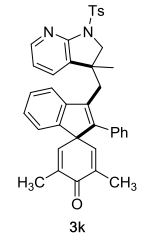

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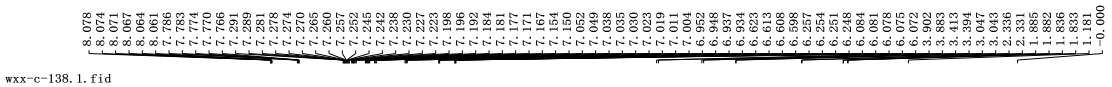

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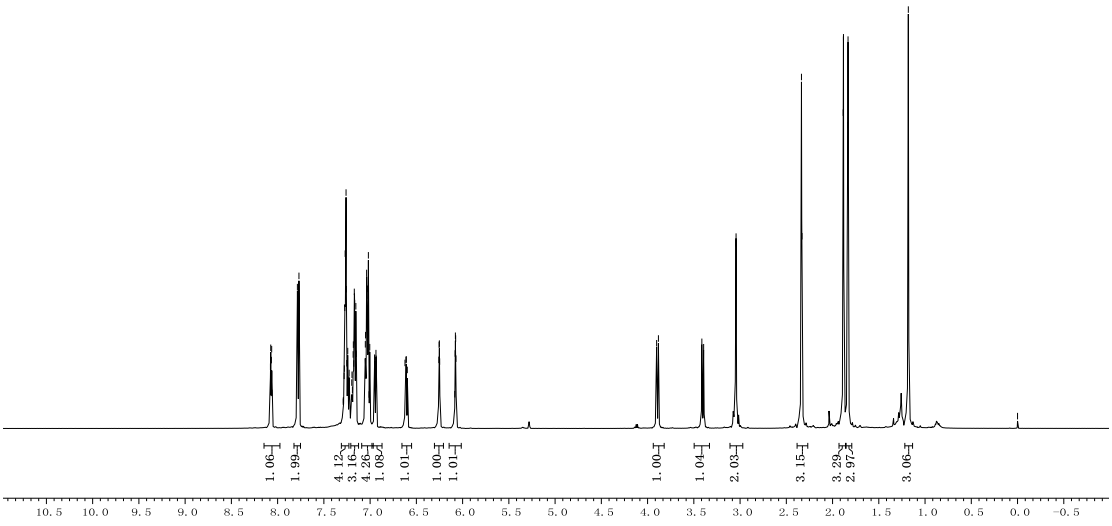

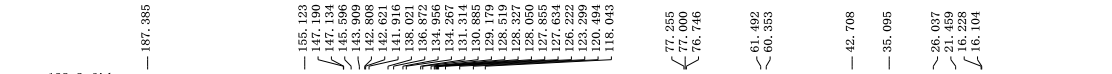

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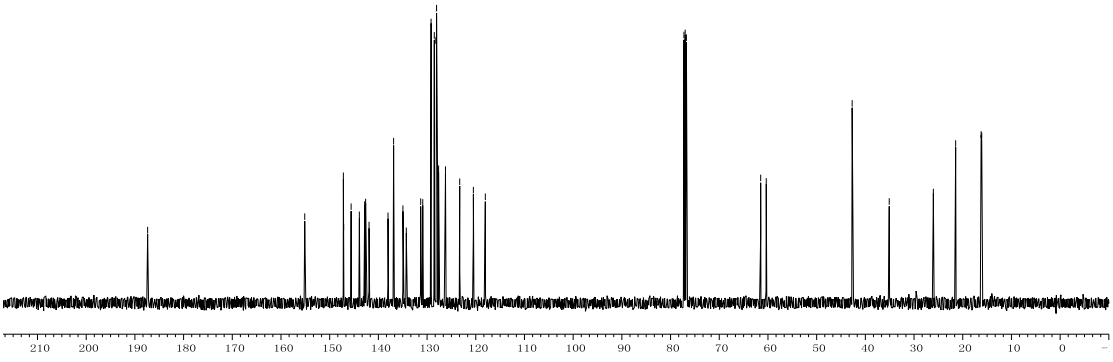

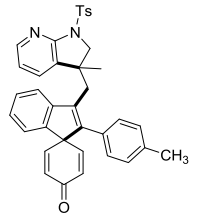

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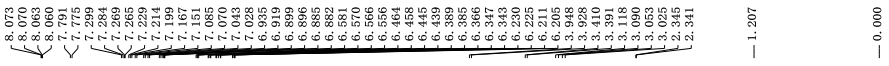

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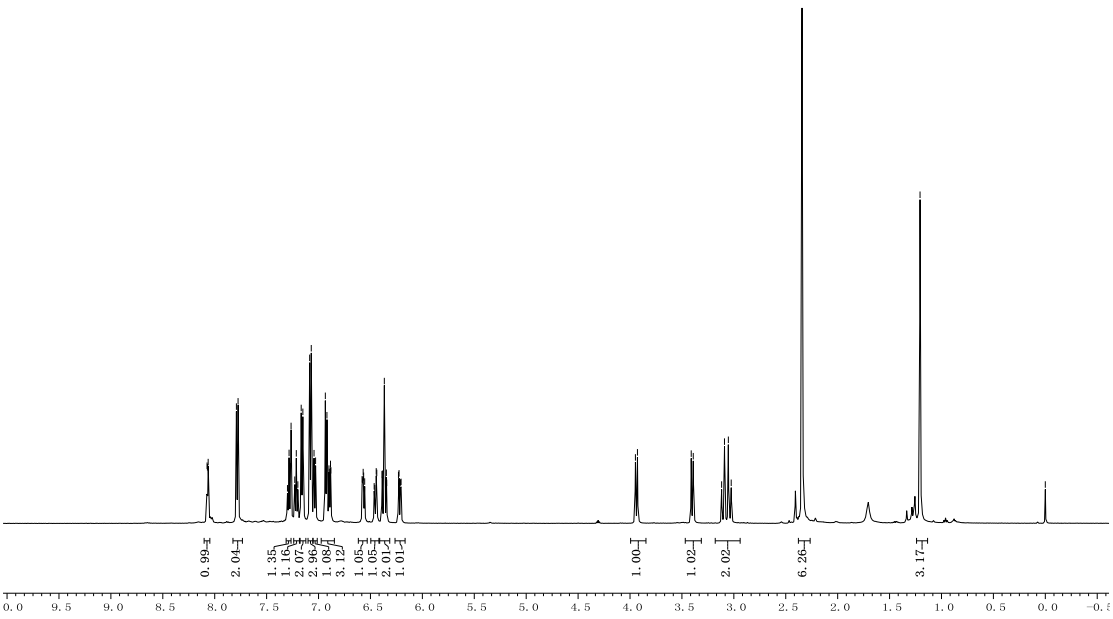

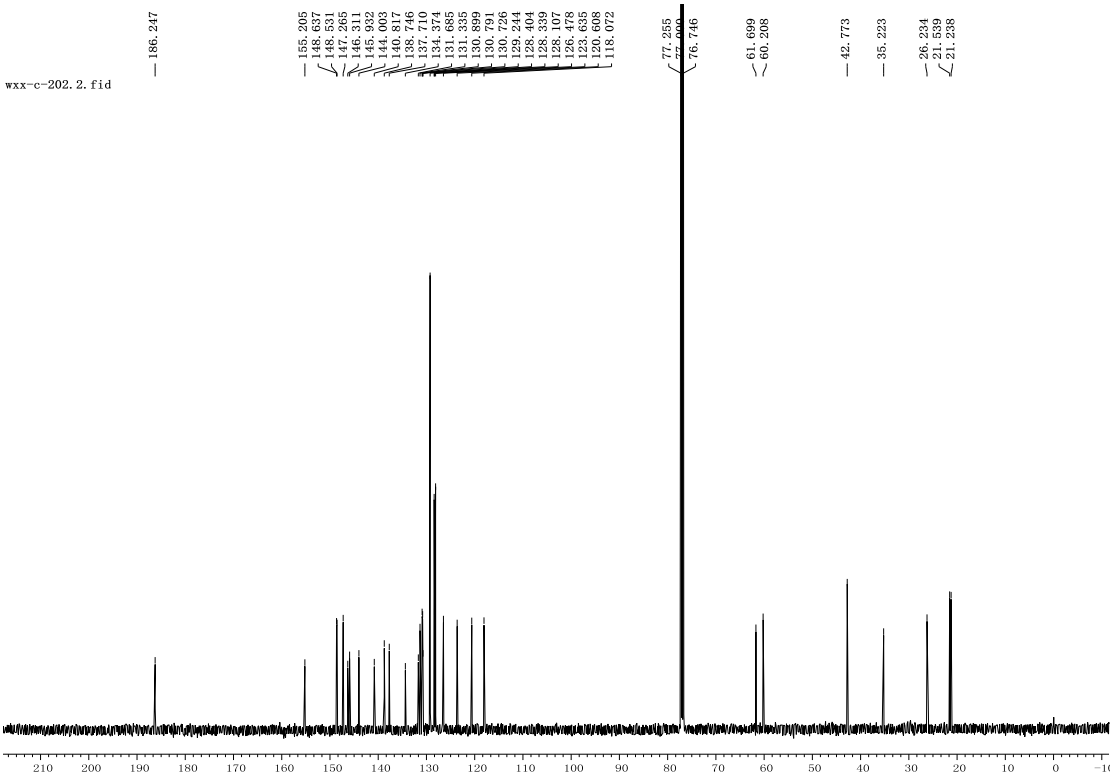

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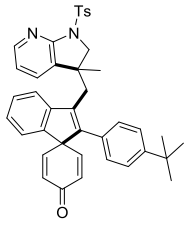

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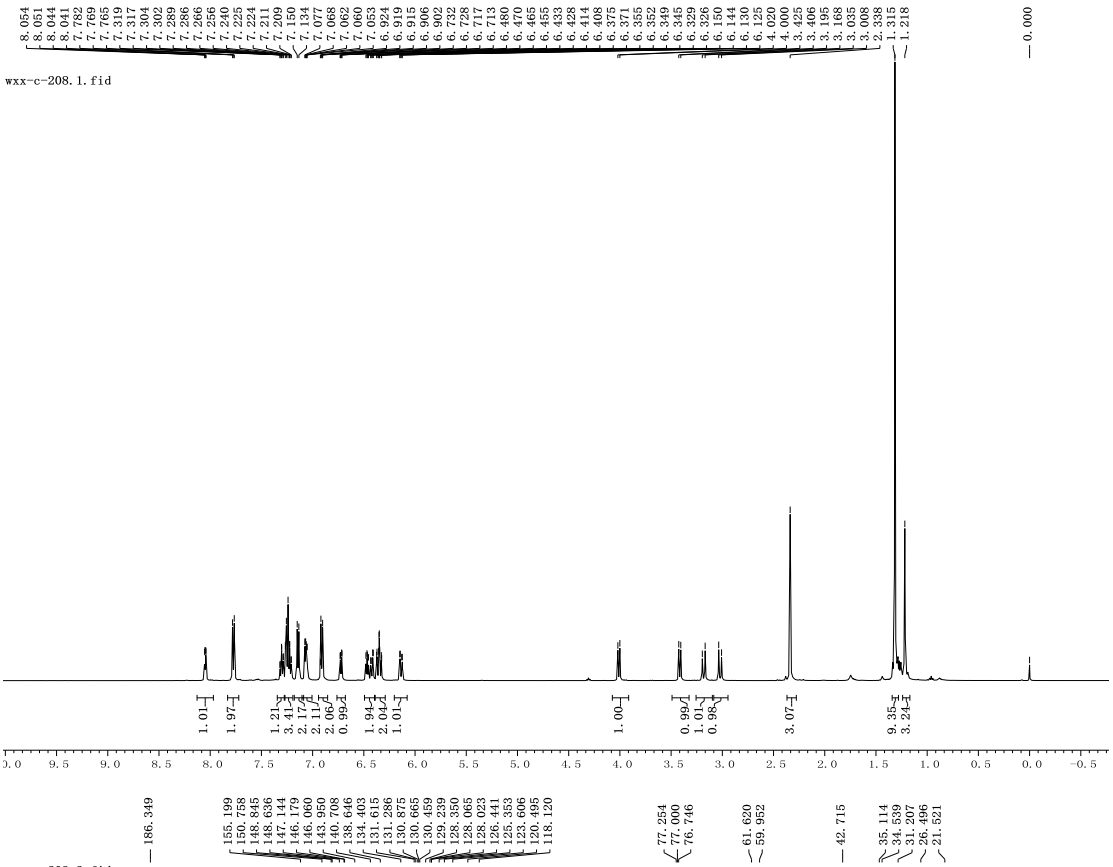

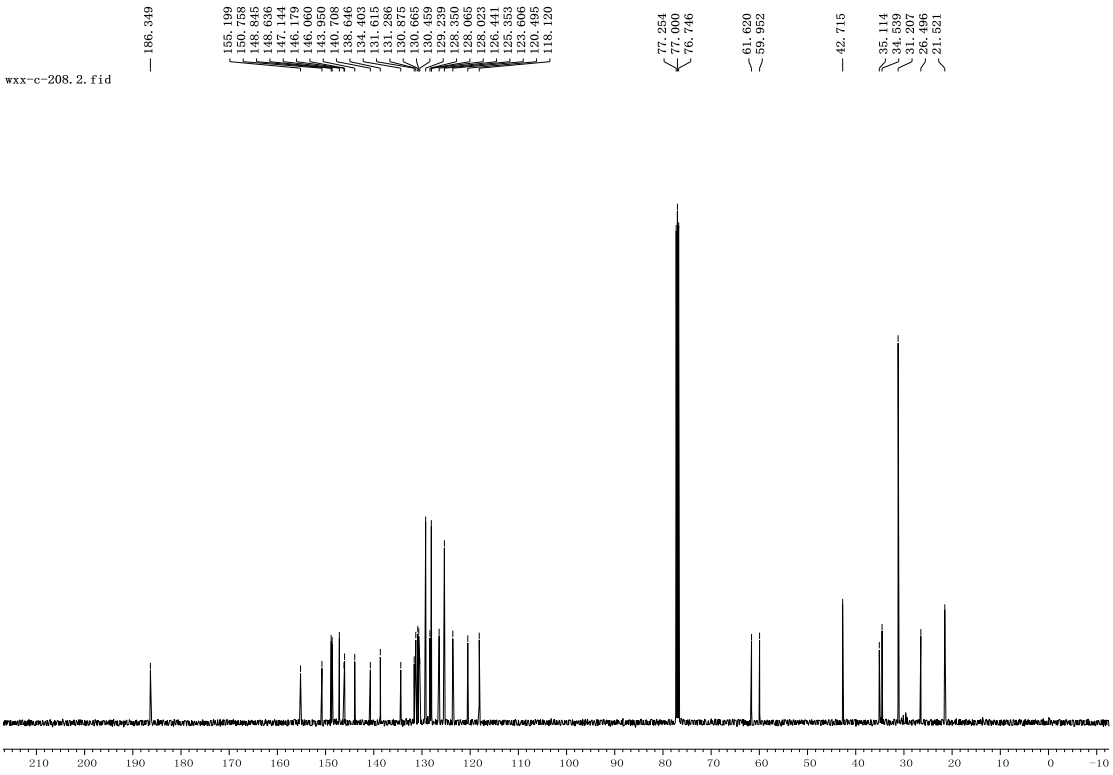

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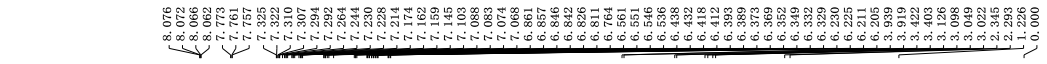

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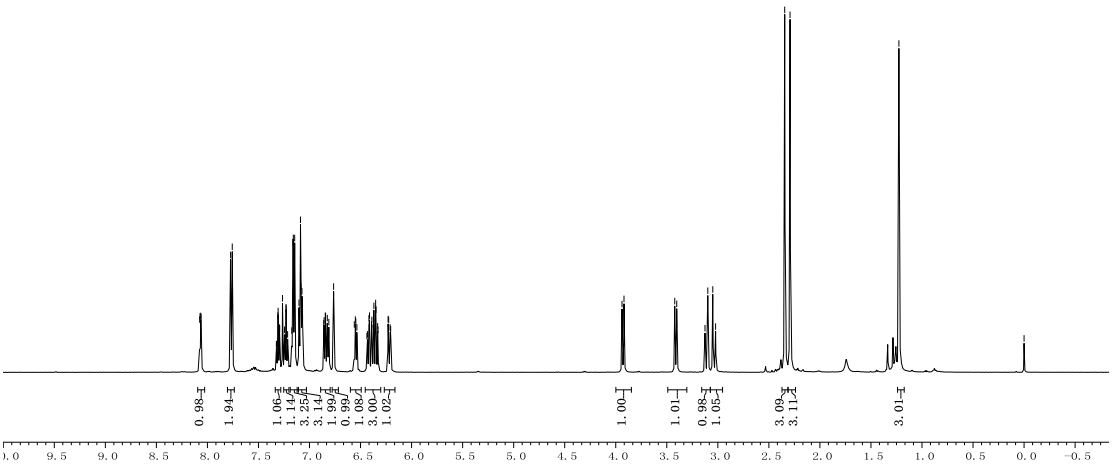

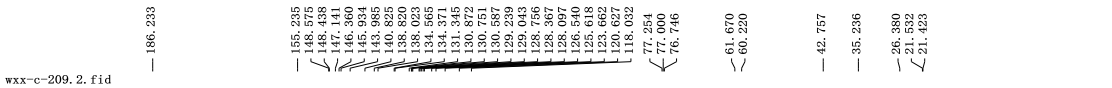

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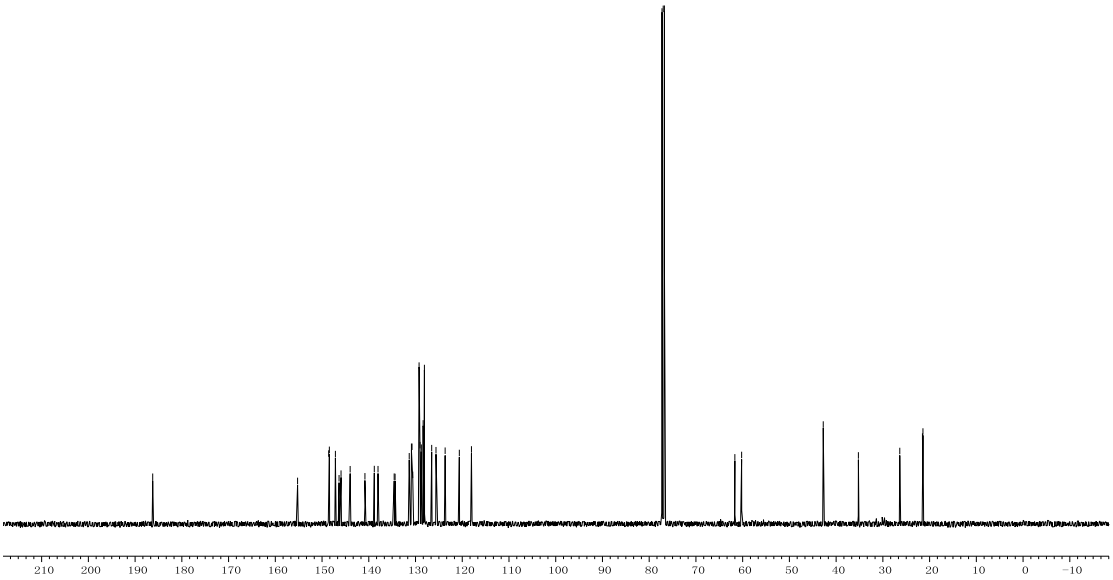

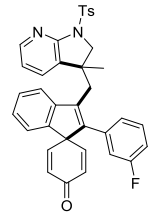

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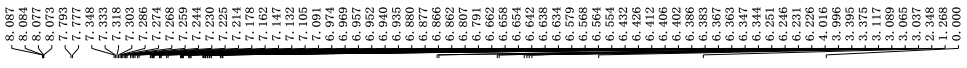

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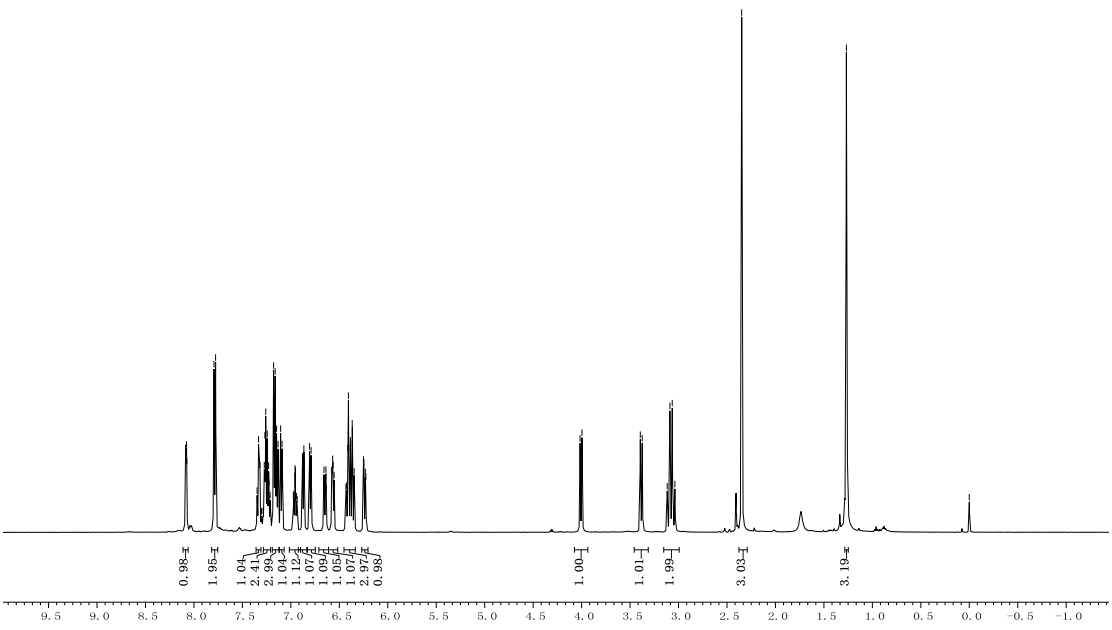

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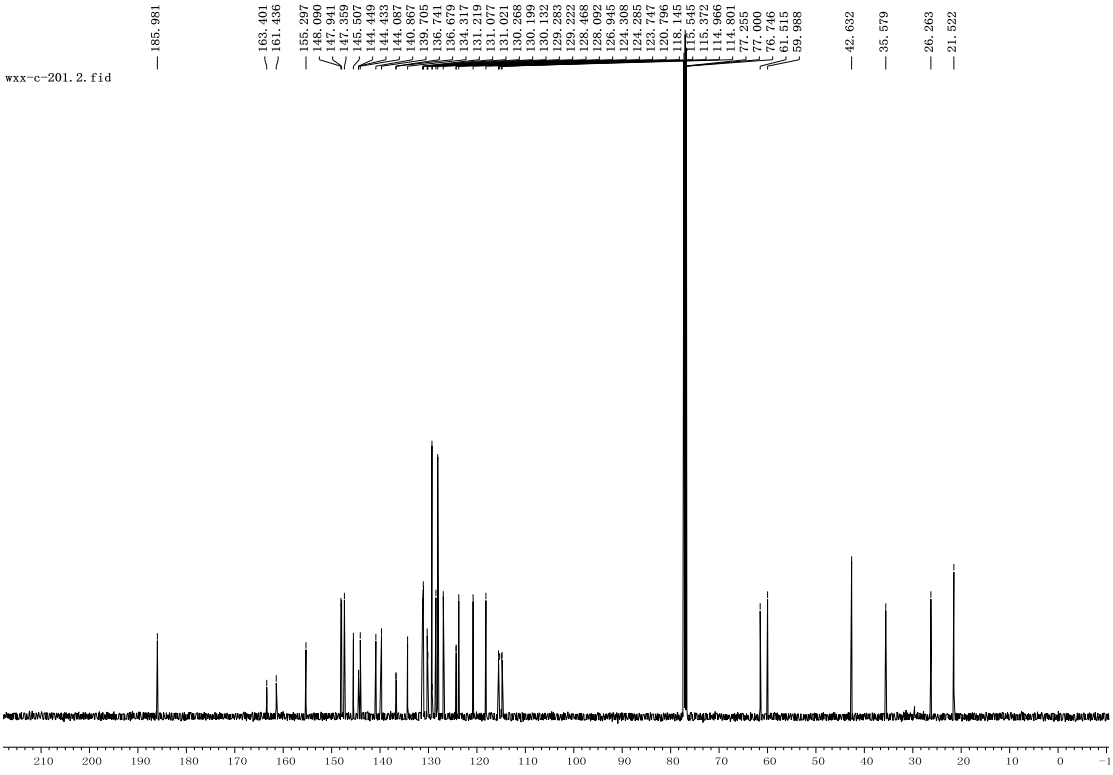

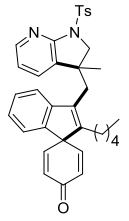

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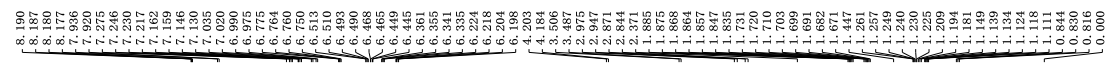

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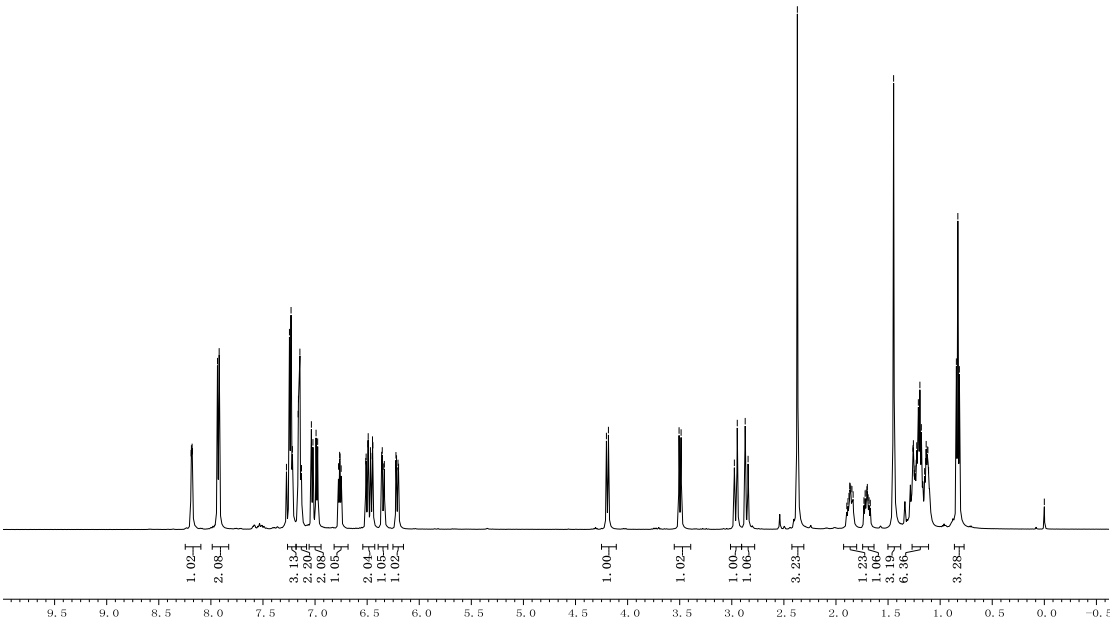

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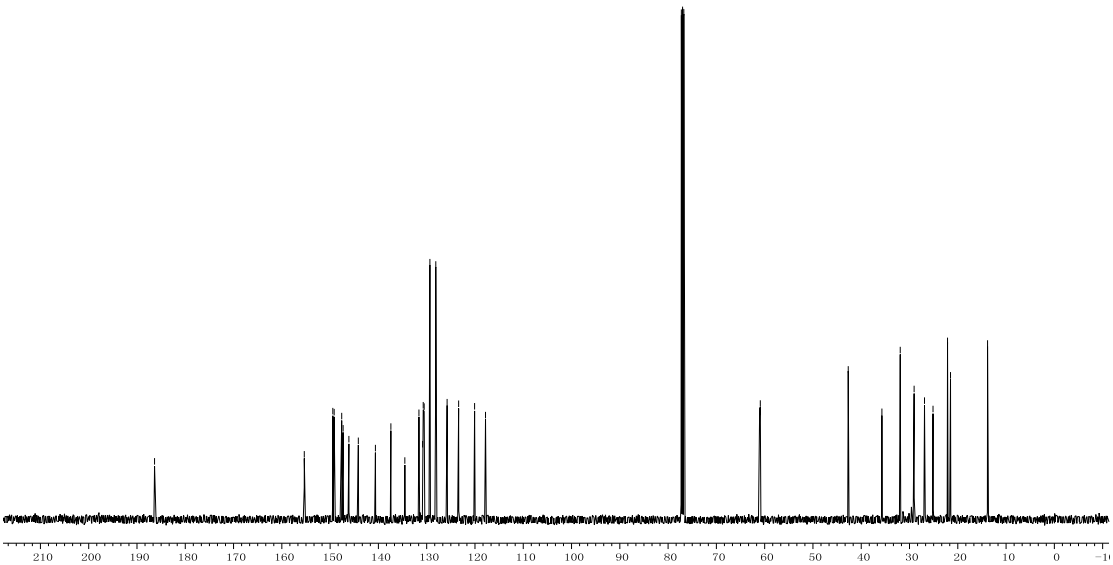

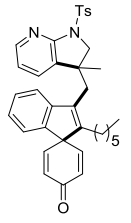

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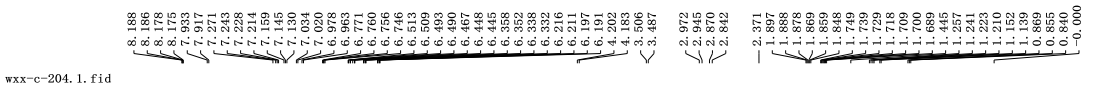

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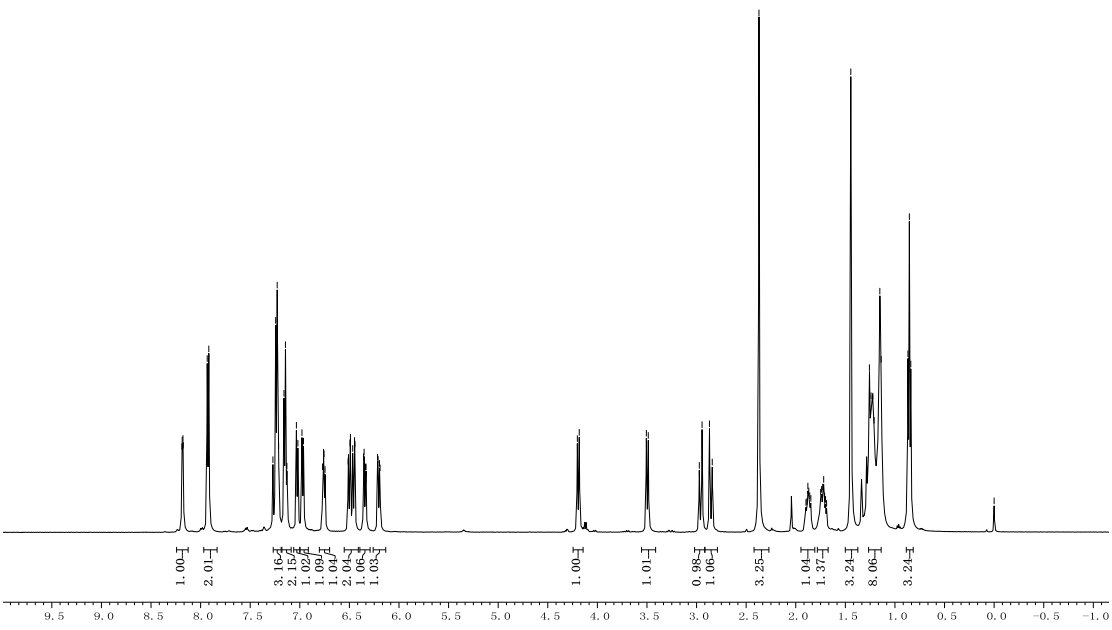

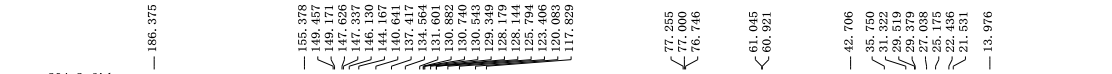

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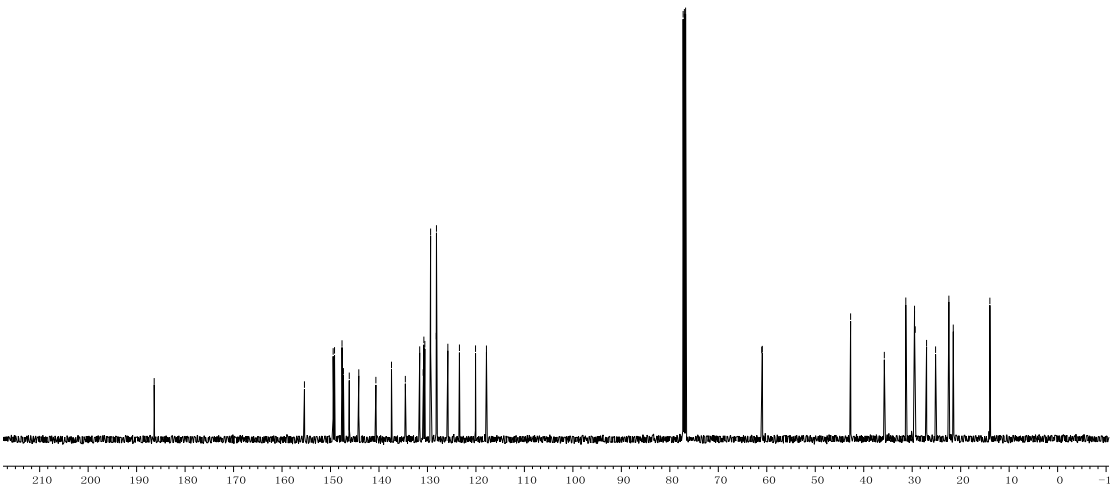

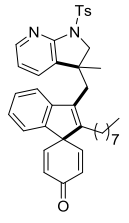

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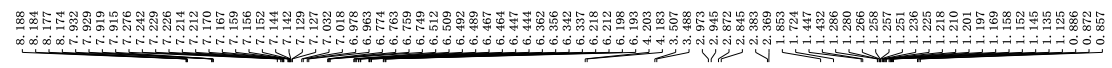

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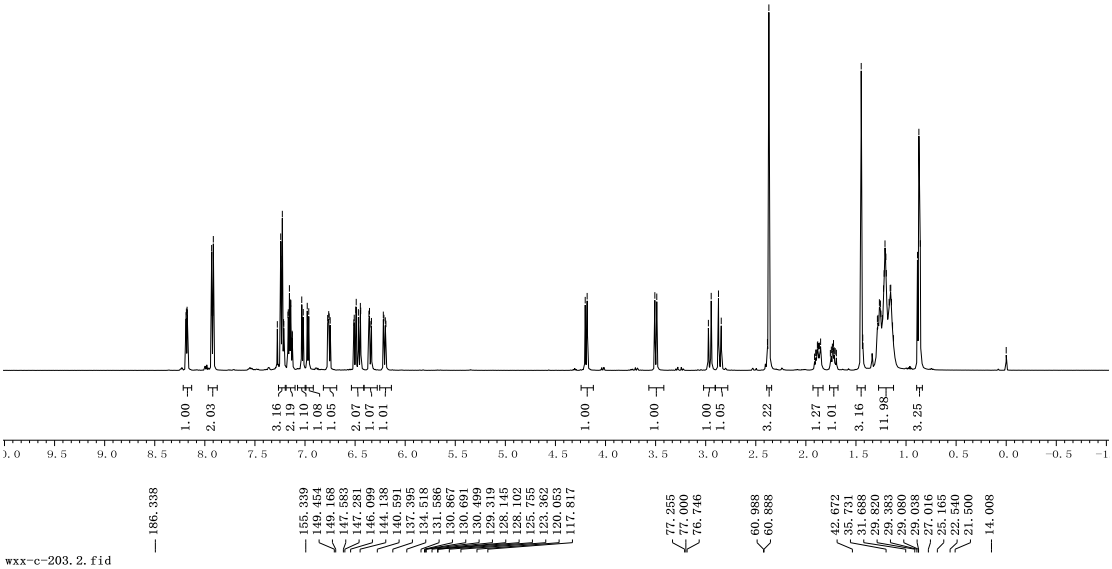

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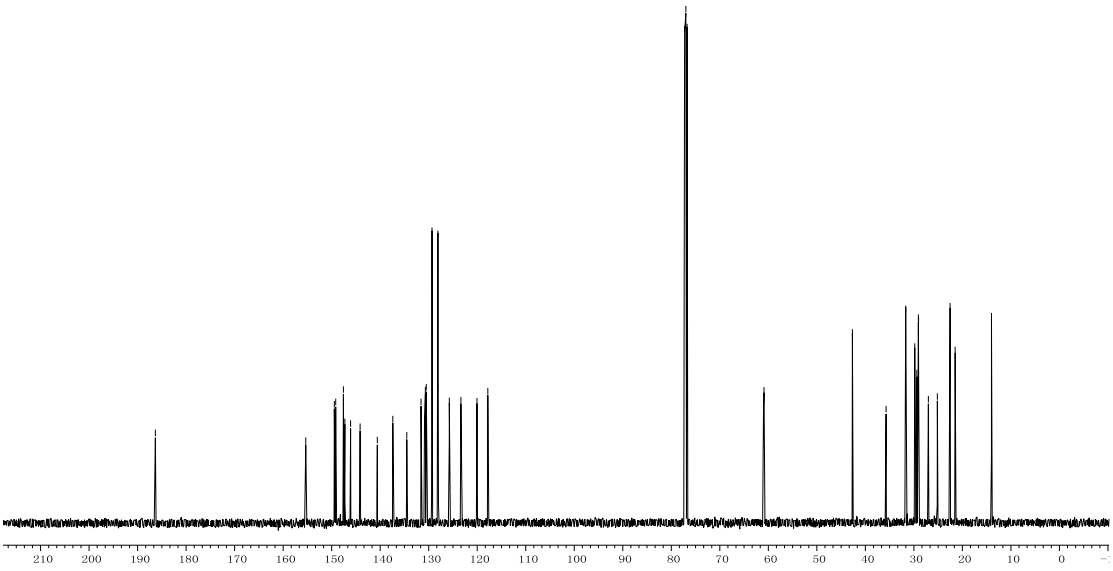

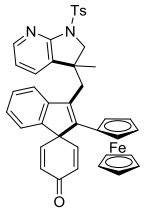

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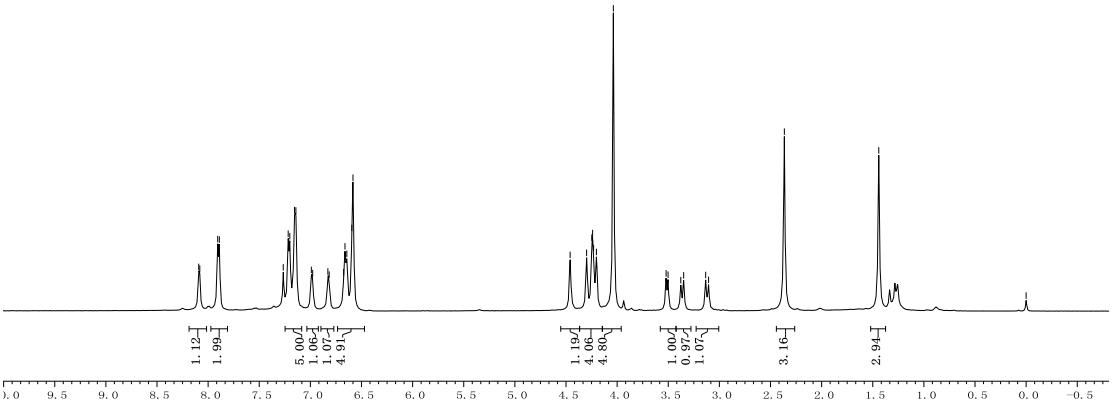

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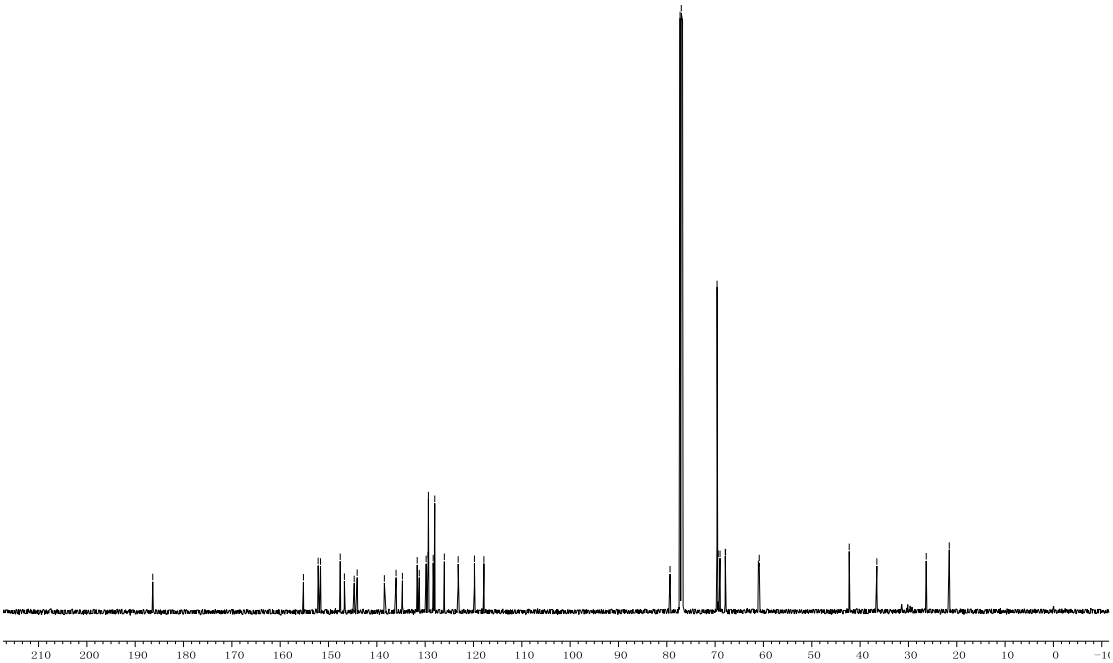

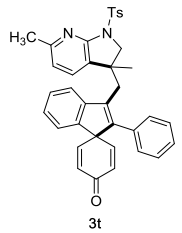

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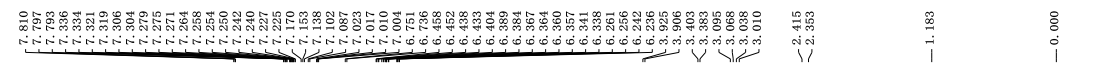

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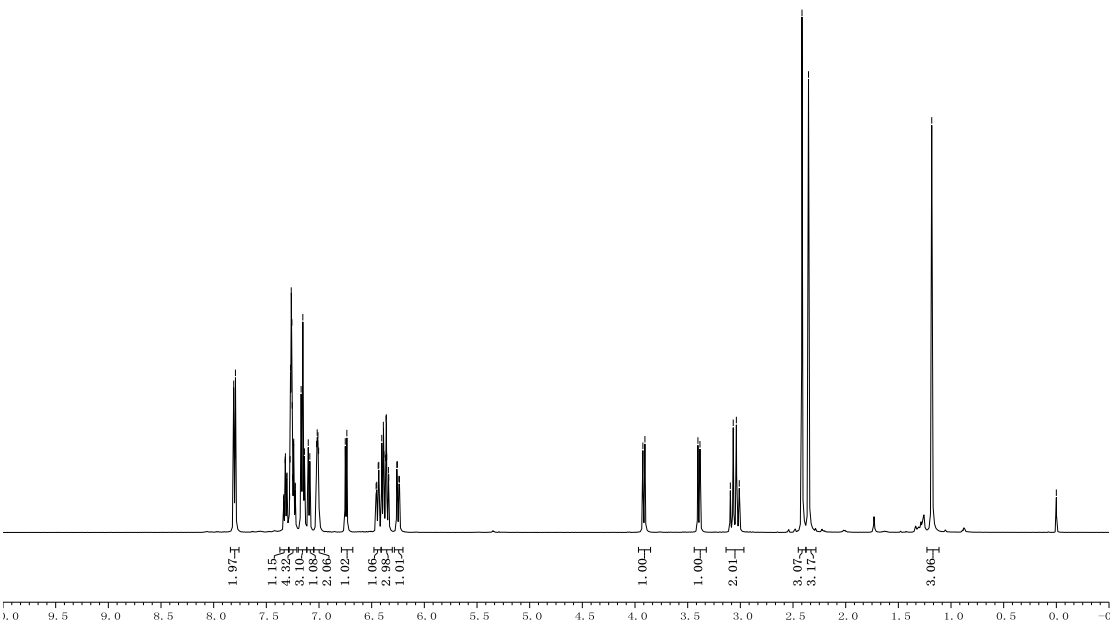

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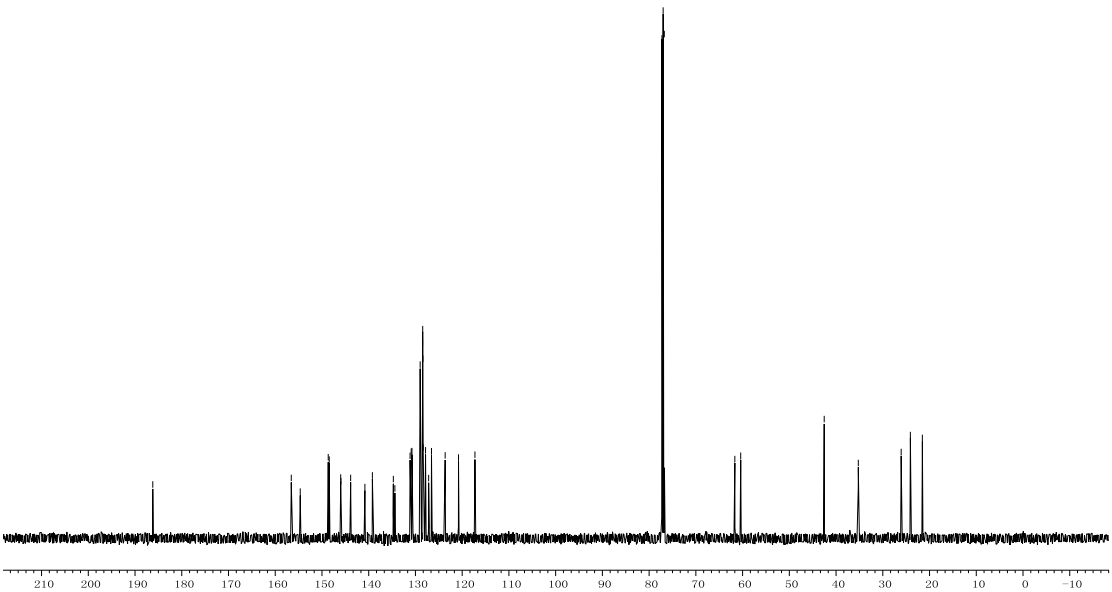

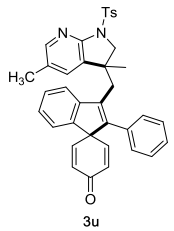

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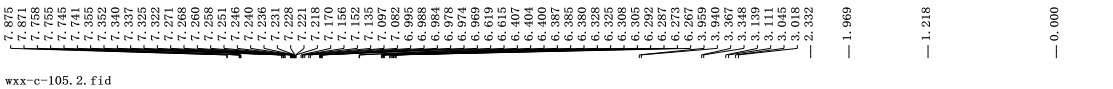

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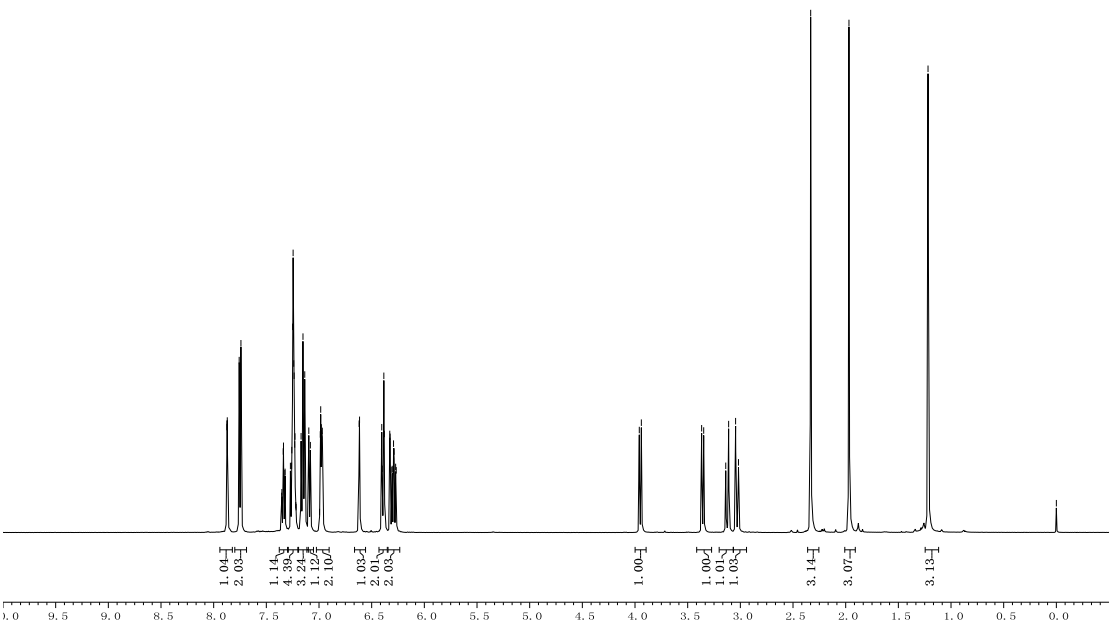

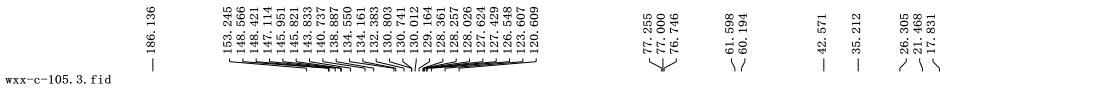

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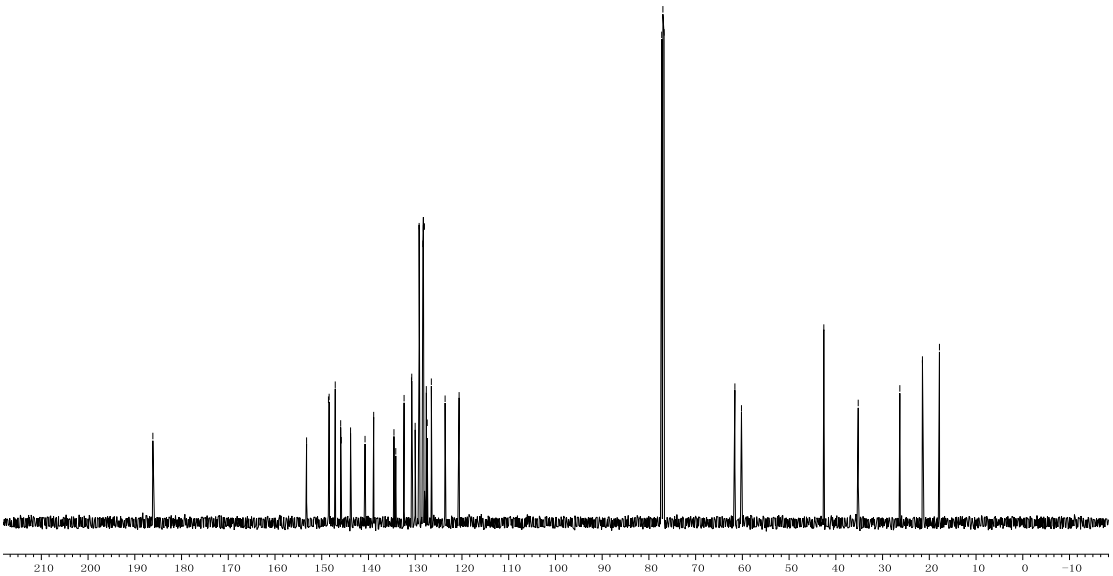

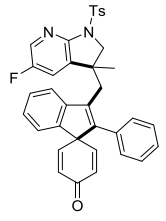

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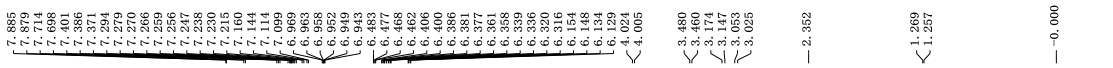

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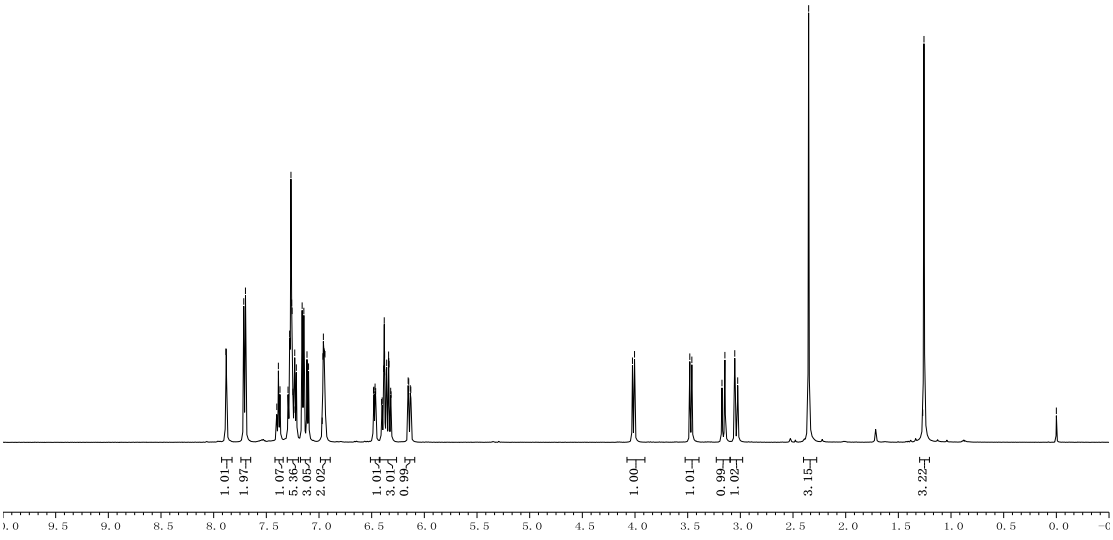

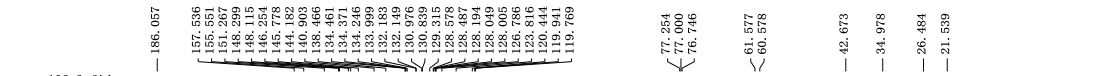

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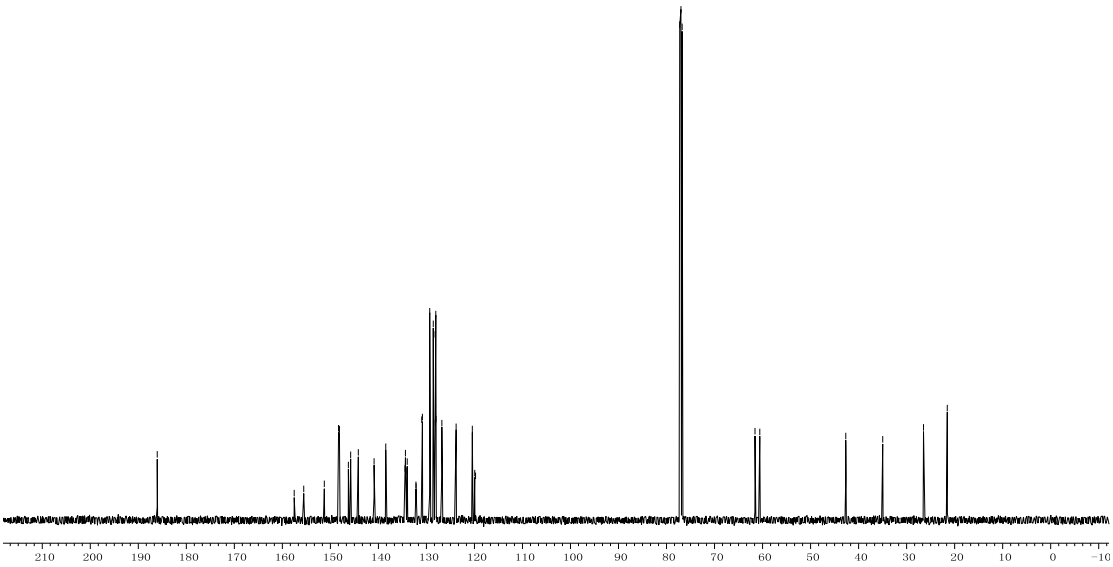

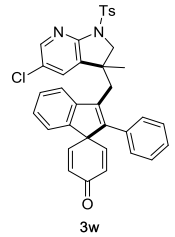

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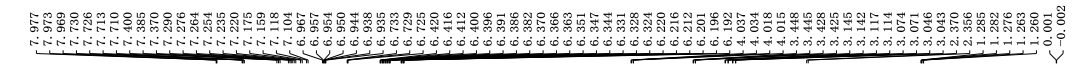

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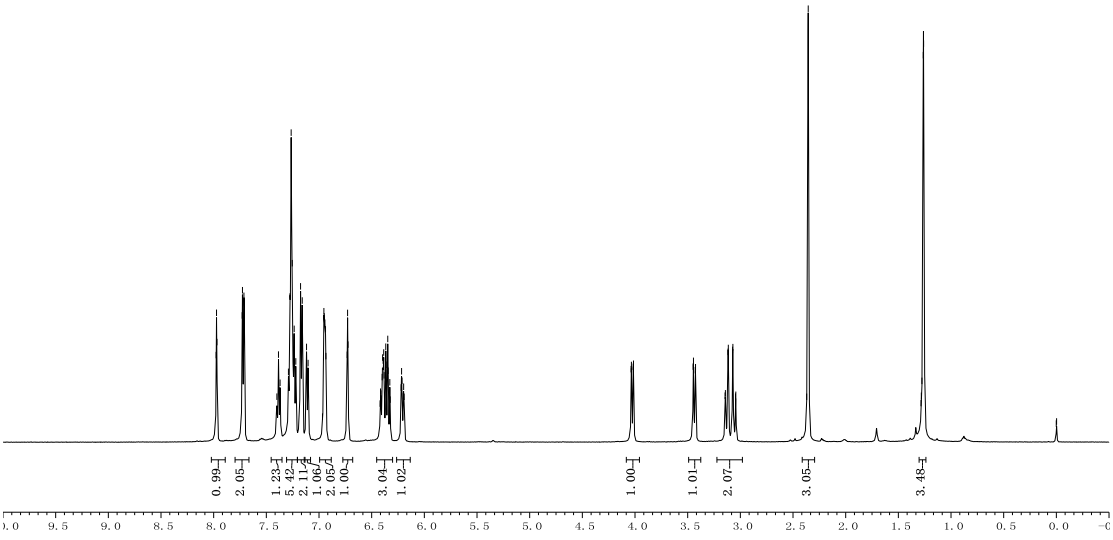

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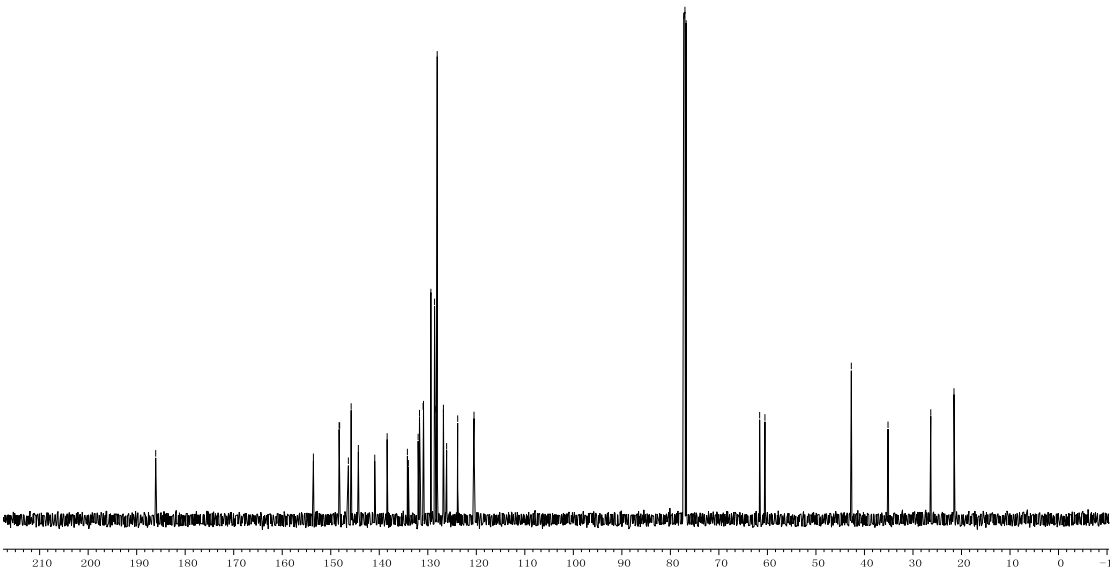

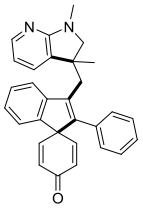

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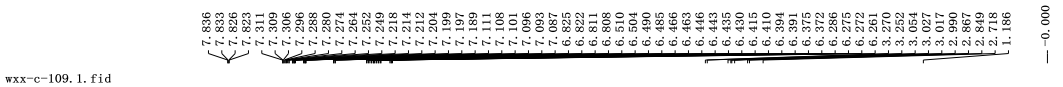

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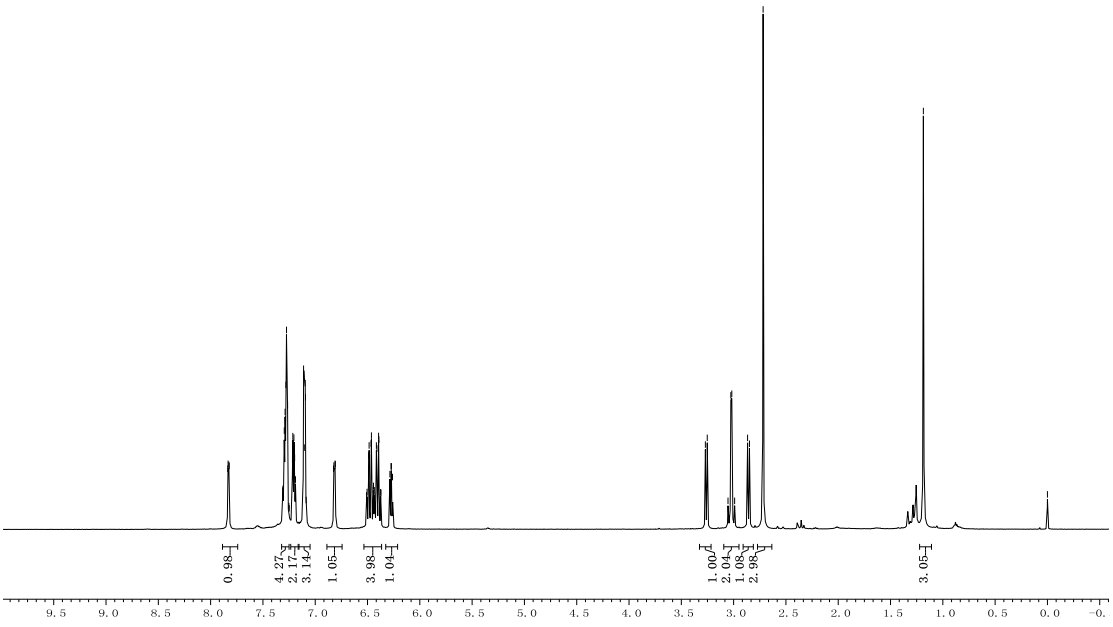

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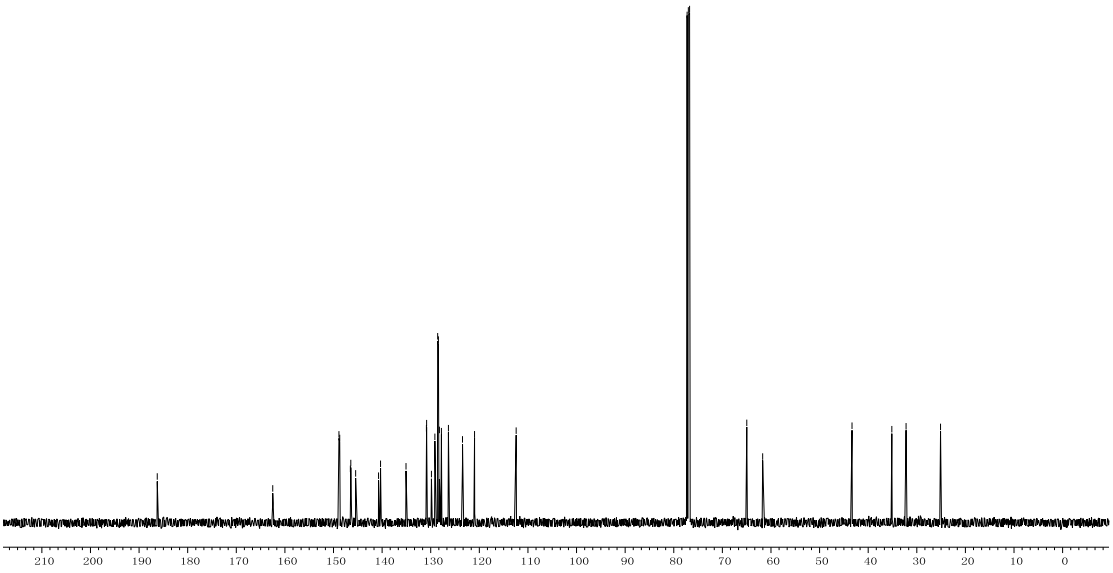

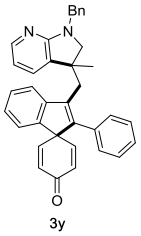

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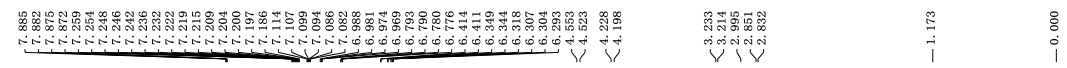

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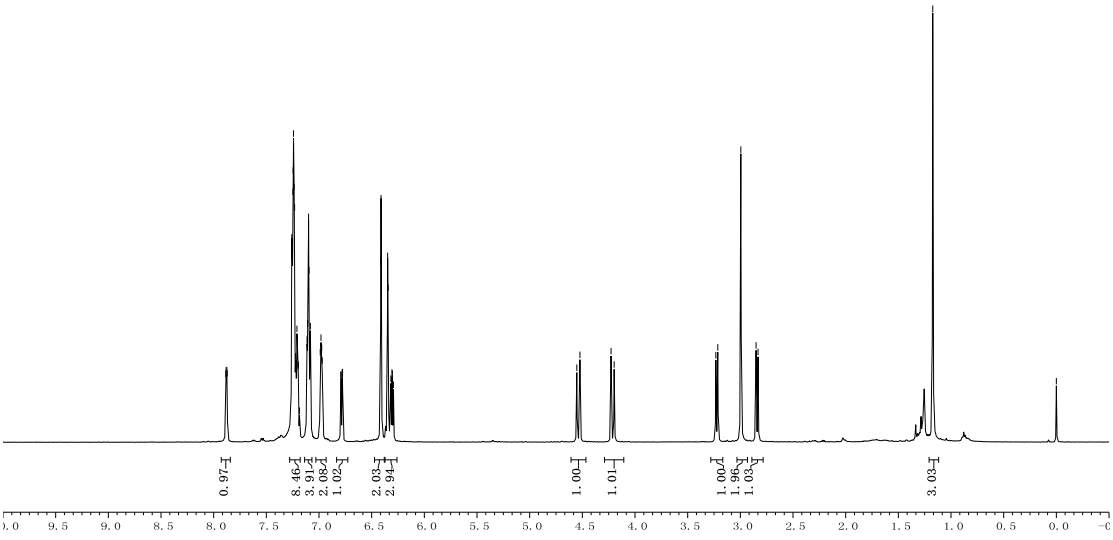

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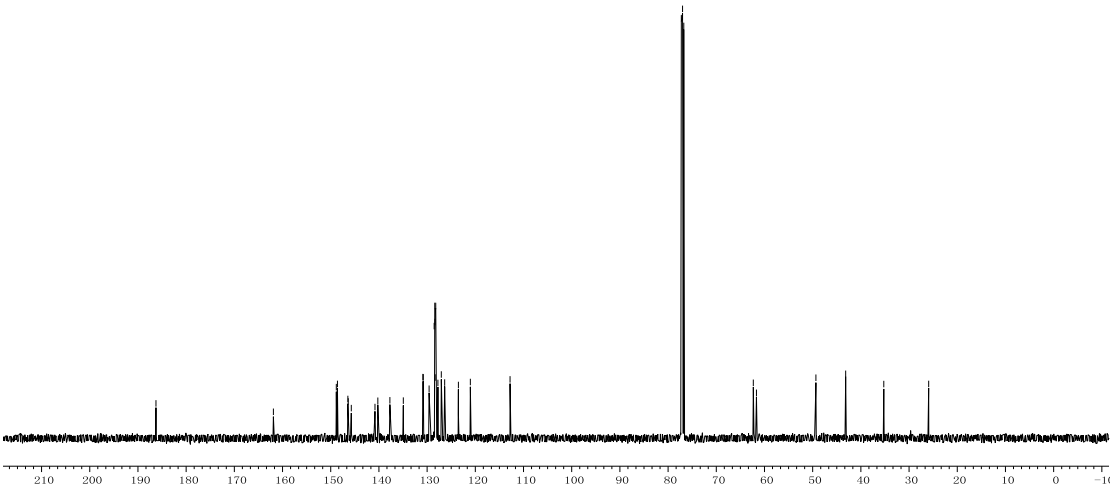

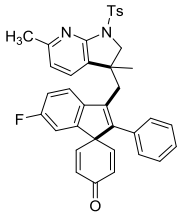

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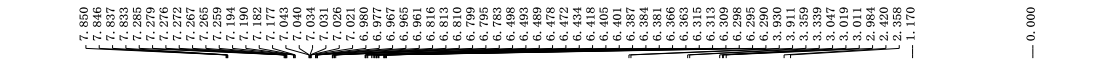

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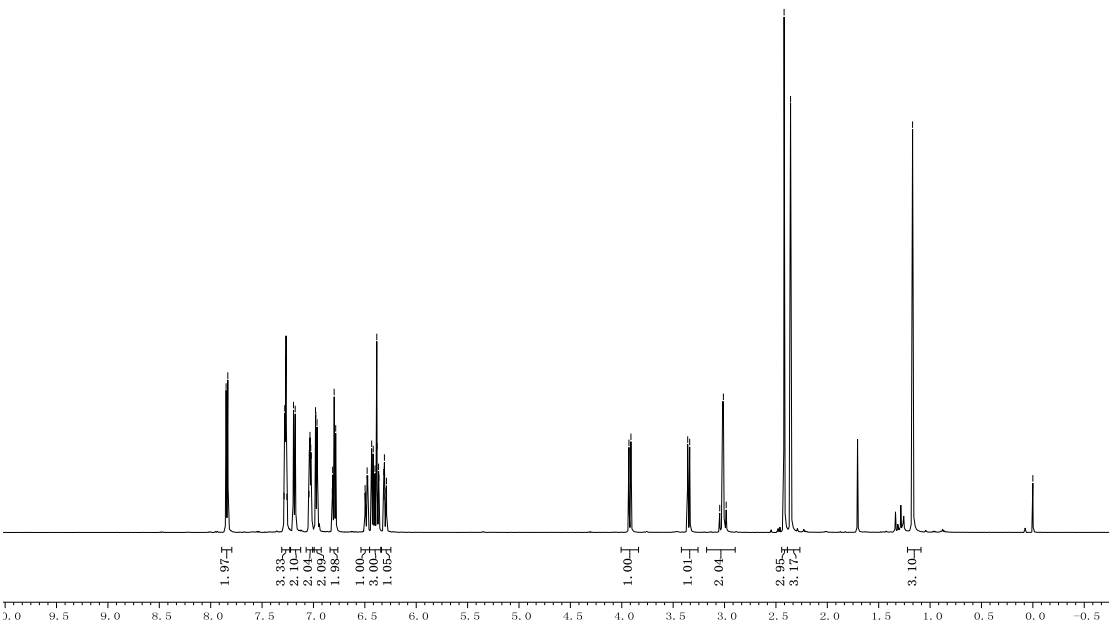

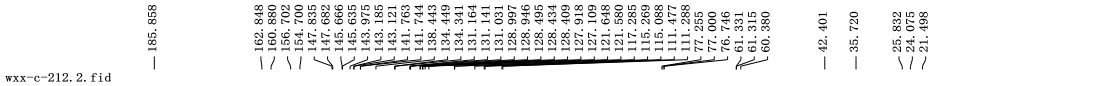

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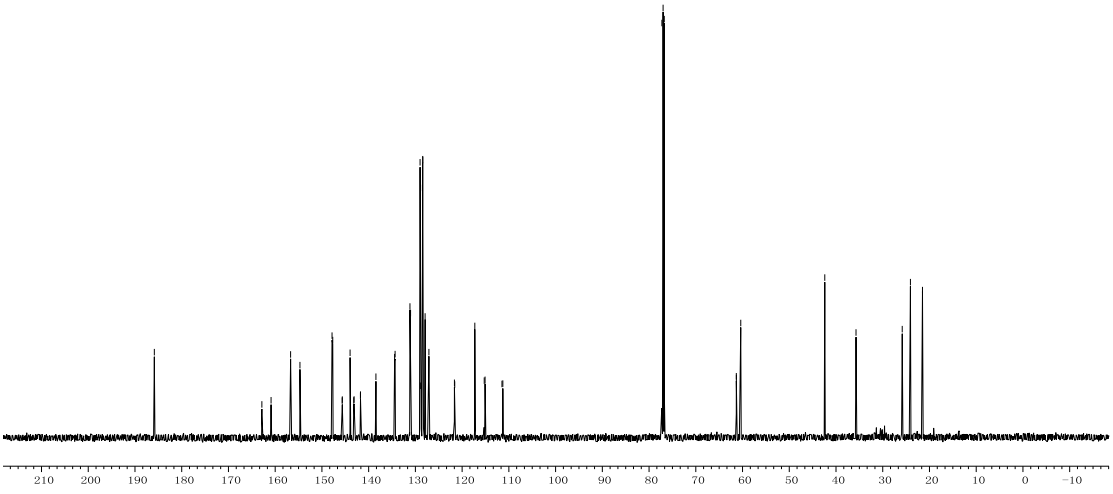

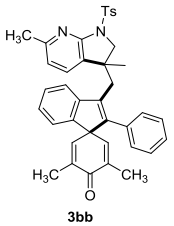

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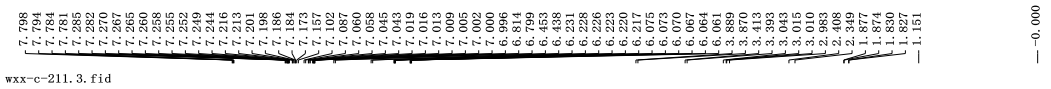

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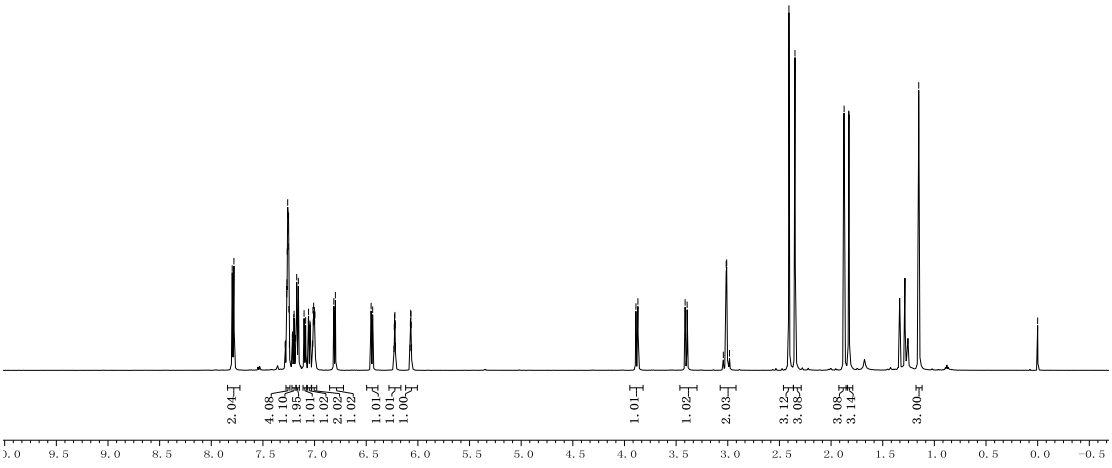

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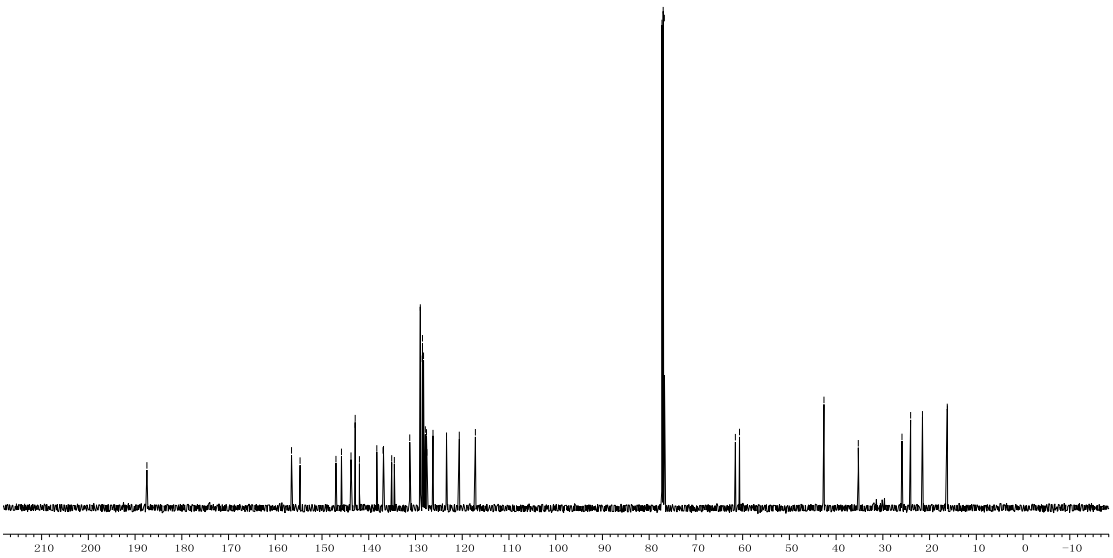

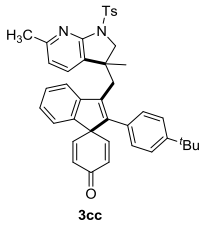

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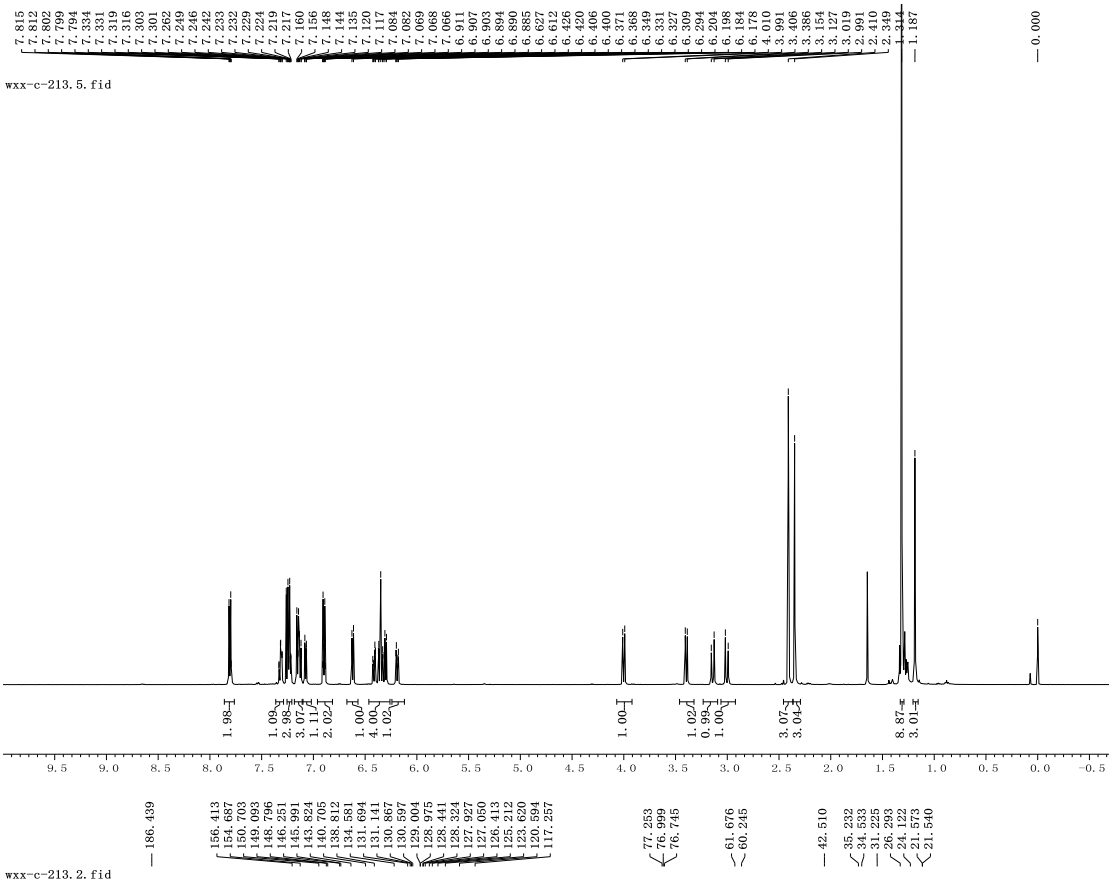

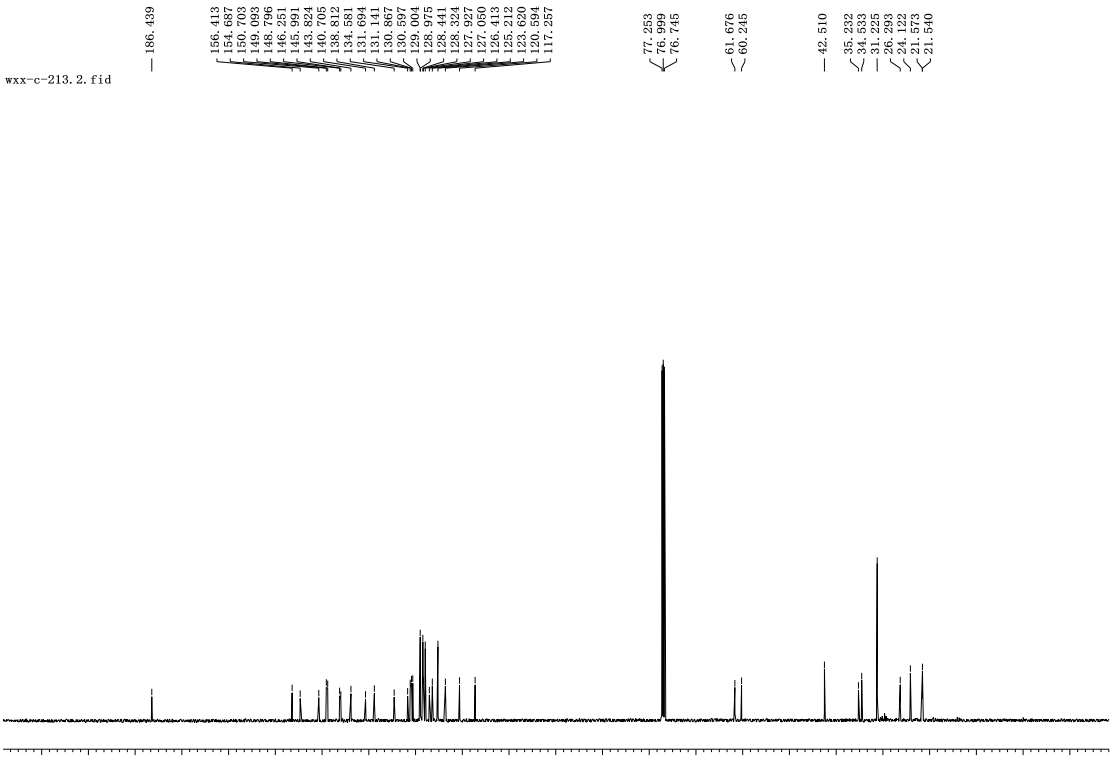

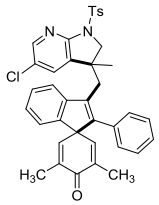

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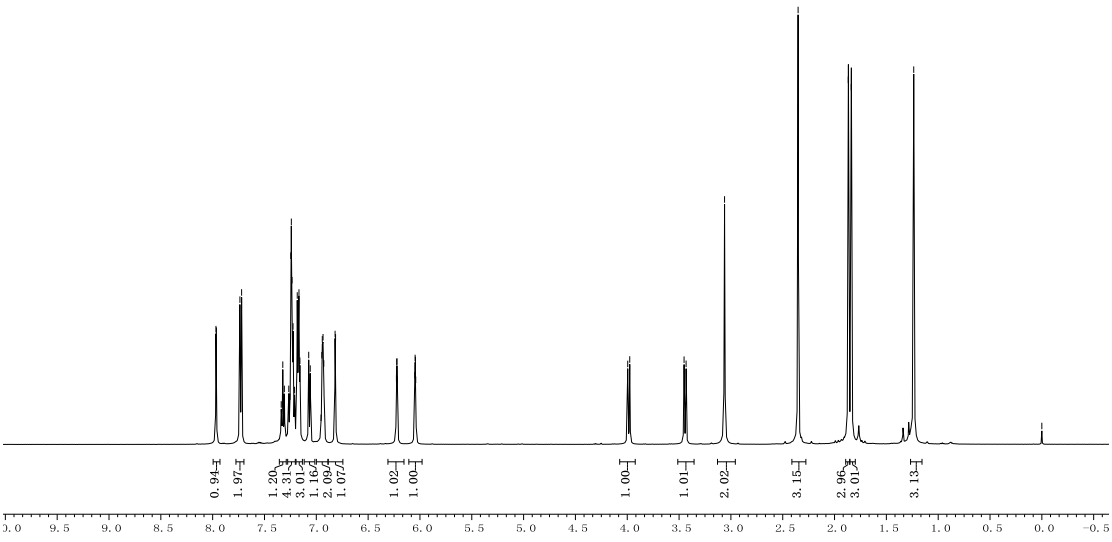

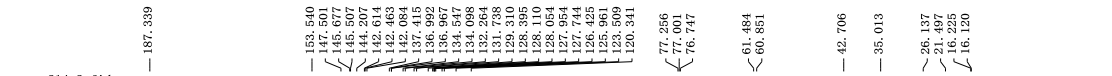

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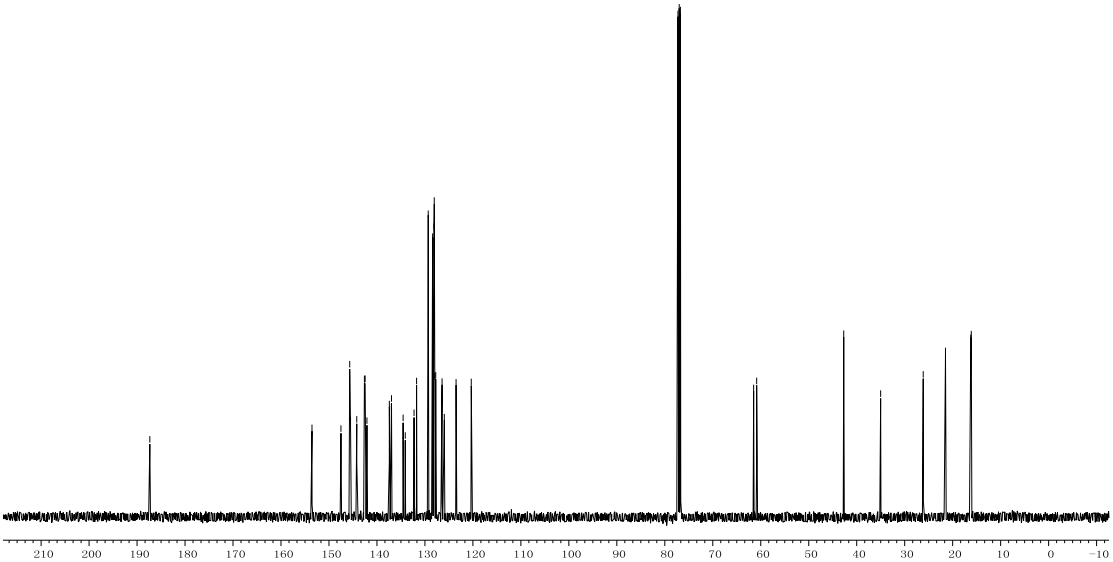

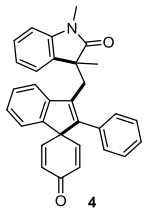

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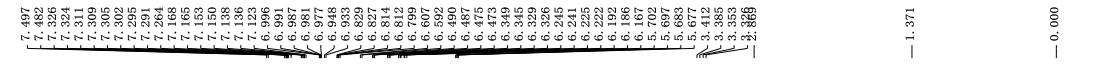

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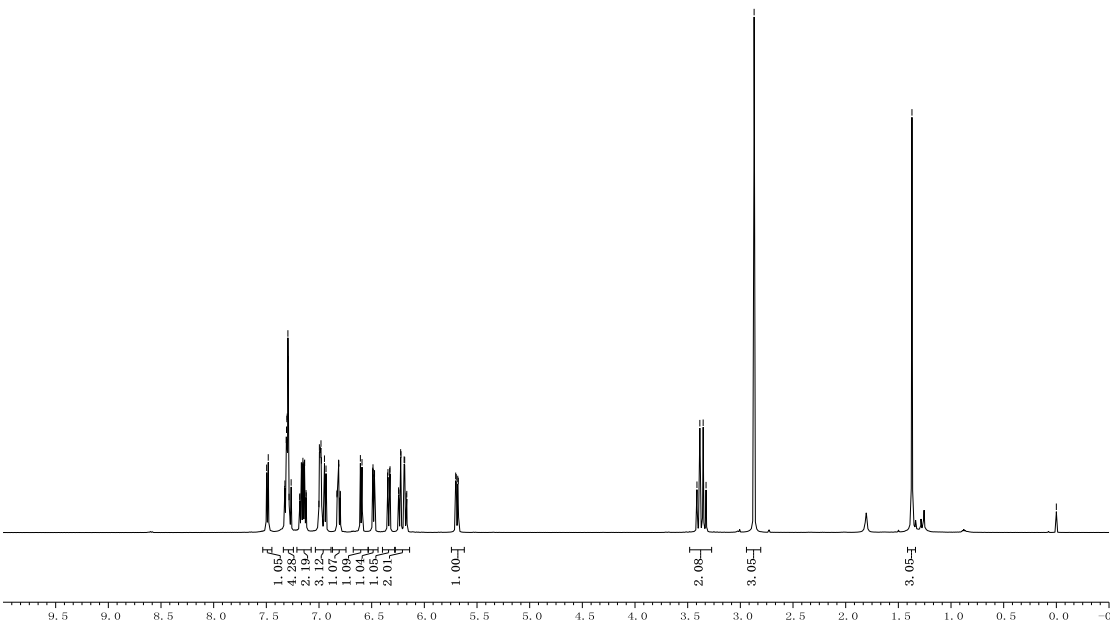

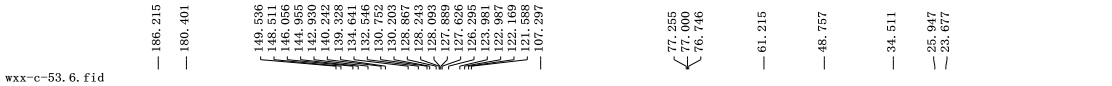

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