

Supporting Information

**Electrochemical Triamination of Alkynes: Controllable Synthesis of
Functionalized Indolines and Indoles**

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I. General Information

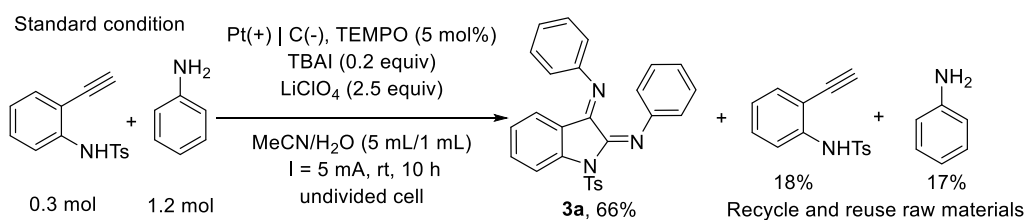
Unless otherwise stated, all glassware was oven-dried. All solvents were distilled from appropriate drying agents prior to use. All reagents were used as received from commercial suppliers unless otherwise indicated. The instrument for electrolysis is a domestic dual display DC stabilized power supply (UTP1303/UTP3305). Cyclic voltammograms were obtained on a CHI600E potentiostat. The anode electrode is platinum plates (1.0 cm × 1.0 cm × 0.2 mm) and the cathode electrode is graphite plates (1.0 cm × 1.0 cm × 0.2 cm). Reactions were monitored using thin layer chromatography (TLC) carried out on Merck silica gel plates (60F-254) using UV light as the visualizing agent and high performance liquid chromatography (HPLC) with UV detection at 254 nm. For HPLC yields, UV response factors relative to an internal standard (1-nitronaphthalene). Flash column chromatography was performed using silica gel 60 (200–300 mesh). HRMS data were recorded on ThermoFisher LTQ Orbitrap XL or Agilent 6500QTOFMS-ESI. GC spectra were recorded on a Shimadzu GC-7920 gas chromatograph instrument with an TCD detector. All ^1H NMR, ^{13}C NMR spectra were recorded on Bruker DRX600 and AMX-400 instruments. Chemical shifts were given in parts per million (ppm, δ), referenced to the solvent peak of CDCl_3 , defined at $\delta = 7.26$ (^1H NMR), defined at $\delta = 77.16$ (^{13}C NMR). Coupling constants were quoted in Hz (J). ^1H NMR spectroscopy splitting patterns were designated as singlet (s), doublet (d), triplet (t), quartet (q). Splitting patterns that could not be interpreted or easily visualized were designated as multiplet (m) or broad (br). The diastereomeric ratios were determined by ^1H NMR analysis.

II. Optimization of reaction conditions

Table S1. Optimization of reaction conditions

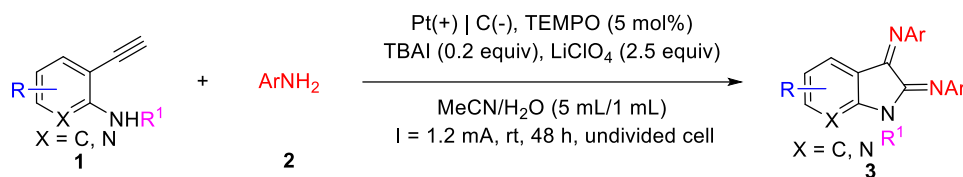
Entry	Electrolyte	AE (auxiliary electrolyte)	Solvent	Catalyst	Yield ^b
1	TBAI	—	CH ₃ CN/H ₂ O (5/1)	—	32%
2	TBAI	—	CH ₃ CN/H ₂ O (5/1)	TEMPO	38%
3	TBAI	LiClO ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	66%
4	NaI	LiClO ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	48%
5	KI	LiClO ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	34%
6	NH ₄ I	LiClO ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	32%
7	Me ₄ NI	LiClO ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	49%
8	TBAB	LiClO ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	0%
9	<i>n</i> -Bu ₄ NPF ₆	LiClO ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	0%
10	TBAI	<i>n</i> -Bu ₄ NBF ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	40%
11	TBAI	<i>n</i> -Bu ₄ NClO ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	25%
12	TBAI	NH ₄ PF ₆	CH ₃ CN/H ₂ O (5/1)	TEMPO	22%
13	TBAI	SDBS	CH ₃ CN/H ₂ O (5/1)	TEMPO	46%
14	TBAI	LiClO ₄	CH ₃ CN/H ₂ O (5.5/0.5)	TEMPO	40%
15	TBAI	LiClO ₄	CH ₃ CN/H ₂ O (4.5/1.5)	TEMPO	44%
16	TBAI	LiClO ₄	DMF/H ₂ O (5/1)	TEMPO	5%
17	TBAI	LiClO ₄	DCE/H ₂ O (5/1)	TEMPO	<1%
18	TBAI	LiClO ₄	DMSO/H ₂ O (5/1)	TEMPO	<1%
19	TBAI	LiClO ₄	THF/H ₂ O (5/1)	TEMPO	<1%
20	—	LiClO ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	0%
21	TBAI	LiClO ₄	CH ₃ CN (6)	TEMPO	0%
22 ^c	TBAI	LiClO ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	41%
23 ^d	TBAI	LiClO ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	20%
24 ^e	TBAI	LiClO ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	0%
25 ^f	TBAI	LiClO ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	29%
26 ^g	TBAI	LiClO ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	48%
27 ^h	TBAI	LiClO ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	65%
28 ⁱ	TBAI	LiClO ₄	CH ₃ CN/H ₂ O (5/1)	TEMPO	64%

^aStandard conditions: Pt anode, Graphite cathode, constant current = 1.2 mA
1a (0.3 mmol), **2a** (4.0 equiv), TBAI (0.2 equiv), LiClO₄ (2.5 equiv),
 TEMPO (5 mol%), H₂O (1.0 mL), MeCN (5.0 mL), rt, 48 h. ^bYields of **3a**
 were determined by HPLC analysis with naphthalene as the internal standard.
^cI = 5 mA. ^dI = 10 mA. ^eWithout electricity. ^f12 h. ^g24 h. ^h72 h. ⁱN₂ instead of air.



Scheme S1. Standard condition

III. Scope of indoline synthesis.

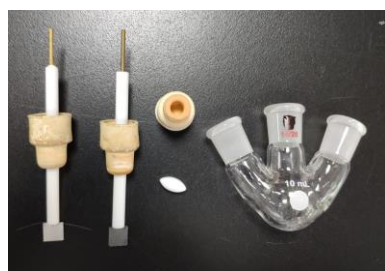


Scheme S2. General procedure for synthesis of functionalized Indolines

Procedure A. In an oven-dried undivided three-necked flask (10 mL) equipped with a stir bar, *o*-ethynylanilines^{1,2} **1** (0.3 mmol), amine **2** (4.0 equiv), LiClO₄ (2.5 equiv), *n*-Bu₄NI (0.2 equiv), TEMPO (5 mol %), H₂O (1 mL) and CH₃CN (5 mL) were combined and added. The flask was equipped with platinum plates (1.0 cm × 1.0 cm × 0.2 mm) and the cathode electrode is graphite plates (1.0 cm × 1.0 cm × 0.2 cm). The reaction mixture was stirred and electrolyzed at a constant current of 1.2 mA under room temperature for 48 h. When the reaction was finished and a red precipitate formed, then filtered with qualitative filter paper and dried under a vacuum to afford **3**.

Procedure B. In an oven-dried undivided three-necked flask (10 mL) equipped with a stir bar, *o*-ethynylanilines¹ **1** (0.3 mmol), amine **2** (4.0 equiv), LiClO₄ (2.5 equiv), *n*-Bu₄NI (0.2 equiv), TEMPO (5 mol %), H₂O (1 mL) and CH₃CN (5 mL) were combined and added. The flask was equipped with platinum plates (1.0 cm × 1.0 cm × 0.2 mm) and the cathode electrode is graphite plates (1.0 cm × 1.0 cm × 0.2 cm). The reaction mixture was stirred and electrolyzed at a constant current of 1.2 mA under room temperature for 48 h. When the reaction was finished and the solvent was removed with a rotary evaporator. The pure product was obtained by flash chromatography on silica gel using petroleum ether and ethyl acetate as the eluent.

Reaction and post-treatment apparatus



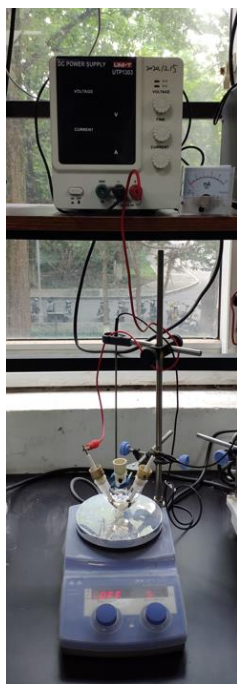
(A) Small scale



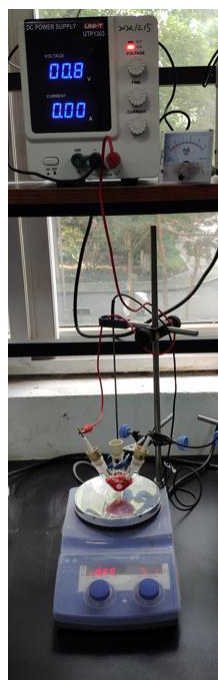
(B) Power supply



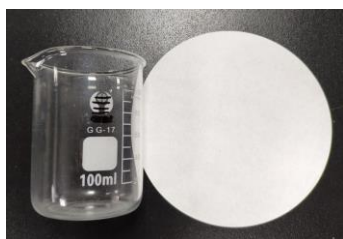
(C) Magnetic stirrer



(D) Before reaction



(E) The reaction mixture after 48 h of electrolysis.



(F) Filtration system



Before reaction



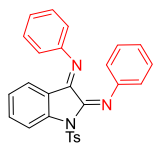
After Reaction



3b

(G) Filtration process

(2E,3E)-N²,N³-Diphenyl-1-tosylindoline-2,3-diimine (3a)



Following General Procedure A: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), aniline (110 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

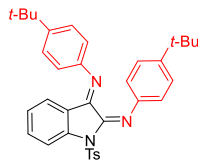
Red solid, 89.3 mg, 66% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.24 (d, *J* = 8.3 Hz, 1H), 8.01 (d, *J* = 6.5 Hz, 2H), 7.44 (t, *J* = 7.9 Hz, 1H), 7.34 (d, *J* = 8.0 Hz, 2H), 7.28 (t, *J* = 7.7 Hz, 2H), 7.22 (t, *J* = 7.6 Hz, 2H), 7.09 (t, *J* = 7.4 Hz, 1H), 6.98 (t, *J* = 7.3 Hz, 1H), 6.81 (t, *J* = 7.6 Hz, 1H), 6.74 (d, *J* = 5.5 Hz, 2H), 6.59 (d, *J* = 7.7 Hz, 3H), 2.47 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 149.7, 147.5, 145.6, 145.3, 145.0, 135.5, 134.0, 129.44, 129.42, 128.8, 128.5, 126.3, 124.6, 123.7, 123.2, 118.8, 116.7, 115.5, 21.8.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₂₇H₂₂N₃O₂S 452.1427, found 452.1417.

(2E,3E)-N²,N³-Bis(4-(*tert*-butyl)phenyl)-1-tosylindoline-2,3-diimine (3b)



Following General Procedure A: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

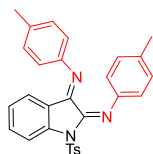
Red solid, 140.2 mg, 83% yield. (*R*_f = 0.25, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.24 (d, *J* = 8.0 Hz, 1H), 8.01 (s, 2H), 7.43 (t, *J* = 7.9 Hz, 1H), 7.33 (t, *J* = 9.1 Hz, 4H), 7.24 (m, 2H), 6.78 (m, 4H), 6.59 (d, *J* = 8.3 Hz, 2H), 2.47 (s, 3H), 1.33 (s, 9H), 1.31 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 147.8, 147.1, 146.2, 145.5, 145.1, 144.8, 144.4, 135.7, 133.7, 129.4, 128.8, 126.12, 126.06, 126.04, 125.2, 123.5, 119.1, 117.0, 115.5, 115.0, 34.5, 34.3, 31.51, 31.50, 21.8.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₃₅H₃₇N₃NaO₂S 586.2499, found 586.2487.

(2E,3E)-N²,N³-Di-*p*-tolyl-1-tosylindoline-2,3-diimine (3c)



Following General Procedure A: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), *p*-toluidine (130.0 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol),

TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

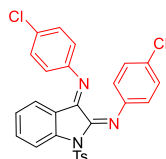
Red solid, 83.5 mg, 58% yield. (*R*_f = 0.2, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.21 (d, *J* = 3.6 Hz, 1H), 7.99 (s, 2H), 7.43 (t, *J* = 7.9 Hz, 1H), 7.32 (d, *J* = 7.9 Hz, 2H), 7.09 (d, *J* = 8.0 Hz, 2H), 7.01 (d, *J* = 7.5 Hz, 2H), 6.82 (t, *J* = 7.6 Hz, 1H), 6.70 (m, 3H), 6.52 (d, *J* = 8.1 Hz, 2H), 2.46 (s, 3H), 2.33 (s, 3H), 2.27 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 147.2, 145.50 145.1, 144.9, 144.6, 135.6, 134.2, 133.7, 132.8, 130.0, 129.3, 129.0, 128.8, 126.1, 123.5, 119.1, 116.9, 115.5, 21.8, 20.99, 20.96.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₂₉H₂₆N₃O₂S 480.1740, found 480.1734.

(2*E*,3*E*)-*N*²,*N*³-Bis(4-chlorophenyl)-1-tosylindoline-2,3-diimine (3d)



Following General Procedure A: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), 4-chloroaniline (156.2 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

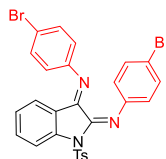
Red solid, 114.4 mg, 74% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.23 (d, *J* = 8.3 Hz, 1H), 7.97 (d, *J* = 6.5 Hz, 2H), 7.47 (t, *J* = 7.5 Hz, 1H), 7.34 (d, *J* = 8.1 Hz, 2H), 7.27 (m, 1H), 7.25 (m, 1H), 7.16 (d, *J* = 8.5 Hz, 2H), 6.86 (t, *J* = 7.6 Hz, 1H), 6.69 (d, *J* = 7.7 Hz, 1H), 6.64 (d, *J* = 6.7 Hz, 2H), 6.55 (d, *J* = 8.6 Hz, 2H), 2.46 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 147.7, 145.9, 145.7, 145.5, 145.2, 135.3, 134.5, 130.1, 129.6, 129.4, 128.7, 128.6, 128.4, 126.2, 123.8, 120.2, 118.4, 115.6, 21.8.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₂₇H₂₀Cl₂N₃O₂S 520.0648, found 520.0651.

(2*E*,3*E*)-*N*²,*N*³-Bis(4-bromophenyl)-1-tosylindoline-2,3-diimine (3e)



Following General Procedure A: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), 4-bromoaniline (210.0 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

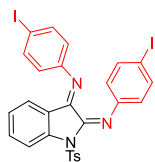
Red solid, 135.2 mg, 74% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.22 (d, *J* = 8.3 Hz, 1H), 7.97 (d, *J* = 6.7 Hz, 2H), 7.47 (t, *J* = 7.6 Hz, 1H), 7.40 (d, *J* = 8.5 Hz, 2H), 7.33 (d, *J* = 8.1 Hz, 2H), 7.30 (d, *J* = 8.5 Hz, 2H), 6.87 (t, *J* = 7.6 Hz, 1H), 6.69 (d, *J* = 7.7 Hz, 1H), 6.58 (d, *J* = 7.2 Hz, 2H), 6.49 (d, *J* = 8.5 Hz, 2H), 2.46 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 149.3, 148.1, 146.4, 145.8, 145.5, 145.1, 135.3, 134.5, 132.6, 131.5, 129.5, 128.7, 126.2, 123.9, 120.6, 118.8, 118.4, 117.8, 116.1, 115.6, 21.8.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₂₇H₂₀Br₂N₃O₂S 607.9637, found 607.9642.

(2*E*,3*E*)-*N*²,*N*³-Bis(4-iodophenyl)-1-tosylindoline-2,3-diimine (3f)



Following General Procedure B: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), 4-iodoaniline (268.2 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

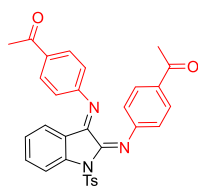
Red solid, 101.2 mg, 48% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 10/1).

¹H NMR (600 MHz, CDCl₃) δ 8.21 (d, *J* = 8.3 Hz, 1H), 7.96 (d, *J* = 6.4 Hz, 2H), 7.60 (d, *J* = 8.1 Hz, 2H), 7.47 (t, *J* = 8.9 Hz, 3H), 7.33 (d, *J* = 8.0 Hz, 2H), 6.87 (t, *J* = 7.6 Hz, 1H), 6.70 (d, *J* = 7.7 Hz, 1H), 6.45 (d, *J* = 6.9 Hz, 2H), 6.37 (d, *J* = 8.1 Hz, 2H), 2.46 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 148.8, 147.0, 145.8, 145.5, 144.9, 138.5, 137.4, 135.3, 134.5, 129.4, 128.7, 126.3, 123.9, 121.0, 119.1, 115.6, 88.3, 21.8.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₂₇H₁₉I₂N₃NaO₂S 725.9180, found 725.9176.

1,1'-((((2*E*,3*E*)-1-Tosylindoline-2,3-diylidene)bis(azaneylylidene))bis(4,1-phenylene))bis(ethan-1-one) (3g)



Following General Procedure B: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), 1-(4-aminophenyl)ethan-1-one (165.5 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

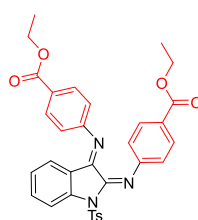
Red solid, 65.9 mg, 41% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 4/1).

¹H NMR (600 MHz, CDCl₃) δ 8.21 (d, *J* = 8.5 Hz, 1H), 7.97 (d, *J* = 8.2 Hz, 2H), 7.88 (d, *J* = 8.4 Hz, 2H), 7.83 (d, *J* = 8.4 Hz, 2H), 7.47 (t, *J* = 7.6 Hz, 1H), 7.35 (d, *J* = 8.2 Hz, 2H), 6.83 (t, *J* = 7.7 Hz, 1H), 6.70 (d, *J* = 8.4 Hz, 2H), 6.61 (d, *J* = 8.4 Hz, 2H), 6.56 (d, *J* = 7.5 Hz, 1H), 2.54 (s, 3H), 2.50 (s, 3H), 2.47 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 197.1, 196.9, 153.3, 152.4, 148.8, 145.9, 145.7, 144.5, 135.1, 134.7, 133.7, 132.3, 130.8, 130.2, 129.5, 128.7, 126.4, 124.1, 118.3, 118.1, 116.6, 115.6, 113.7, 26.4, 26.4, 21.8.

HRMS-ESI (m/z) [M+K]⁺ calculated for C₃₁H₂₅KN₃O₄S 574.1197, found 574.1189.

Diethyl 4,4'-((((2*E*,3*E*)-1-tosylindoline-2,3-diylidene)bis(azaneylylidene))dibenzoate (3h)



Following General Procedure B: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), ethyl 4-aminobenzoate (202.3 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture was stirred and electrolyzed at a constant current of 5 mA under room temperature for 24 h.

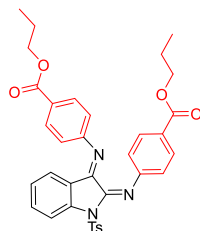
Red solid, 85.8 mg, 48% yield. (*R*_f = 0.30, petroleum ether/ethyl acetate = 10/1).

¹H NMR (600 MHz, CDCl₃) δ 8.22 (d, *J* = 8.5 Hz, 1H), 7.98 (d, *J* = 8.3 Hz, 2H), 7.96 (d, *J* = 8.5 Hz, 2H), 7.91 (d, *J* = 8.5 Hz, 2H), 7.47 (t, *J* = 7.5 Hz, 1H), 7.35 (d, *J* = 8.2 Hz, 2H), 6.82 (t, *J* = 7.7 Hz, 1H), 6.68 (d, *J* = 8.4 Hz, 2H), 6.59 (d, *J* = 8.5 Hz, 2H), 6.55 (d, *J* = 7.4 Hz, 1H), 4.35 (q, *J* = 7.1 Hz, 2H), 4.30 (q, *J* = 7.1 Hz, 2H), 2.47 (s, 3H), 1.36 (dt, *J* = 14.0, 7.1 Hz, 7H).

¹³C NMR (150 MHz, CDCl₃) δ 166.6, 166.1, 153.2, 152.0, 148.8, 145.9, 145.6, 144.5, 135.1, 134.7, 131.2, 130.5, 129.5, 128.7, 126.8, 126.4, 125.1, 124.0, 118.1, 116.4, 115.6, 113.8, 61.0, 60.6, 21.8, 14.36, 14.35.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₃₃H₃₀N₃O₆S 596.1850, found 596.1855.

Dipropyl 4,4'-(((2*E*,3*E*)-1-tosylindoline-2,3-diylidene)bis(azaneylylidene))dibenzoate (3i)



Following General Procedure B: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), propyl 4-aminobenzoate (220.1 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture was stirred and electrolyzed at a constant current of 5 mA under room temperature for 20 h.

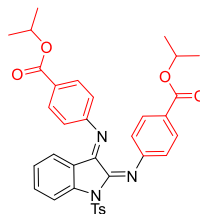
Red solid, 99.2 mg, 53% yield. (*R*_f = 0.30, petroleum ether/ethyl acetate = 10/1).

¹H NMR (600 MHz, CDCl₃) δ 8.22 (d, *J* = 8.5 Hz, 1H), 7.98 (d, *J* = 8.3 Hz, 2H), 7.96 (d, *J* = 8.6 Hz, 2H), 7.91 (d, *J* = 8.4 Hz, 2H), 7.47 (t, *J* = 8.0 Hz, 1H), 7.35 (d, *J* = 8.2 Hz, 2H), 6.83 (t, *J* = 7.7 Hz, 1H), 6.68 (d, *J* = 8.4 Hz, 2H), 6.60 (d, *J* = 8.4 Hz, 2H), 6.56 (d, *J* = 7.4 Hz, 1H), 4.25 (t, *J* = 6.6 Hz, 2H), 4.21 (t, *J* = 6.7 Hz, 2H), 2.47 (s, 3H), 1.80 – 1.71 (m, 4H), 0.99 – 1.02 (m, 6H).

¹³C NMR (150 MHz, CDCl₃) δ 166.6, 166.2, 153.2, 152.0, 148.8, 145.9, 145.6, 144.5, 135.1, 134.7, 131.2, 130.5, 129.5, 128.7, 126.8, 126.4, 125.2, 124.0, 118.1, 116.5, 115.6, 66.6, 66.3, 22.1, 22.1, 21.8, 10.6, 10.5.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₃₅H₃₄N₃O₆S 624.2163, found 624.2170.

Diisopropyl 4,4'-(((2*E*,3*E*)-1-tosylindoline-2,3-diylidene)bis(azaneylylidene))dibenzoate (3j)



Following General Procedure B: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), isopropyl 4-aminobenzoate (219.4 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture was stirred and electrolyzed at a constant current of 5 mA under room temperature for 24 h.

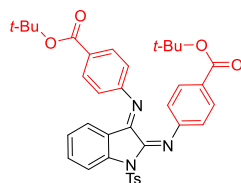
Red solid, 99.3 mg, 53% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 10/1).

¹H NMR (600 MHz, CDCl₃) δ 8.22 (d, *J* = 8.5 Hz, 1H), 7.98 (d, *J* = 8.3 Hz, 2H), 7.96 (d, *J* = 8.5 Hz, 2H), 7.90 (d, *J* = 8.5 Hz, 2H), 7.46 (t, *J* = 7.4 Hz, 1H), 7.34 (d, *J* = 8.2 Hz, 2H), 6.83 (t, *J* = 7.7 Hz, 1H), 6.68 (d, *J* = 8.4 Hz, 2H), 6.60 (d, *J* = 8.5 Hz, 2H), 6.57 (d, *J* = 7.5 Hz, 1H), 5.26 – 5.14 (m, 2H), 2.46 (s, 3H), 1.34 (d, *J* = 6.3 Hz, 6H), 1.32 (d, *J* = 6.3 Hz, 6H).

¹³C NMR (150 MHz, CDCl₃) δ 166.0, 165.6, 153.1, 151.9, 148.8, 145.8, 145.6, 144.5, 135.1, 134.7, 131.2, 130.5, 129.5, 128.7, 127.2, 126.5, 125.5, 124.0, 118.2, 118.1, 116.4, 115.6, 68.4, 67.9, 22.0, 22.0, 21.8.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₃₅H₃₄N₃O₆S 624.2163, found 624.2166.

Di-*tert*-butyl 4,4'-(((2*E*,3*E*)-1-tosylindoline-2,3-diylidene)bis(azaneylylidene))dibenzoate (3k)



Following General Procedure A: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), *tert*-butyl 4-aminobenzoate (236.0 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

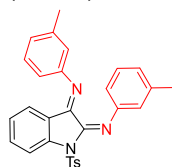
Red solid, 119.1 mg, 61% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.22 (d, *J* = 8.5 Hz, 1H), 7.98 (d, *J* = 8.2 Hz, 2H), 7.91 (d, *J* = 8.5 Hz, 2H), 7.85 (d, *J* = 8.5 Hz, 2H), 7.46 (t, *J* = 7.4 Hz, 1H), 7.34 (d, *J* = 8.1 Hz, 2H), 6.83 (t, *J* = 7.7 Hz, 1H), 6.67 (d, *J* = 8.4 Hz, 2H), 6.59 (d, *J* = 8.5 Hz, 3H), 2.46 (s, 3H), 1.58 (s, 9H), 1.55 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 165.7, 165.3, 152.9, 151.6, 148.8, 145.8, 145.6, 144.5, 135.2, 134.6, 131.1, 130.3, 129.5, 128.7, 128.3, 126.6, 126.5, 124.0, 118.2, 118.1, 116.3, 115.6, 81.0, 80.5, 28.3, 28.2, 21.8.

HRMS-ESI (m/z) [M+K]⁺ calculated for C₃₇H₃₇KN₃O₆S 690.2035, found 690.2020.

(2*E*,3*E*)-*N*²,*N*³-Di-*m*-tolyl-1-tosylindoline-2,3-diimine (3l)



Following General Procedure A: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), *p*-toluidine (130 μL, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

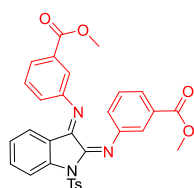
Red solid, 106.5 mg, 74% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.25 (d, *J* = 8.3 Hz, 1H), 8.02 (d, *J* = 6.6 Hz, 2H), 7.45 (t, *J* = 7.9 Hz, 1H), 7.35 (d, *J* = 7.9 Hz, 2H), 7.18 (t, *J* = 7.6 Hz, 1H), 7.12 (t, *J* = 7.6 Hz, 1H), 6.92 (d, *J* = 7.5 Hz, 1H), 6.83 (d, *J* = 7.6 Hz, 1H), 6.81 (d, *J* = 8.1 Hz, 1H), 6.67 – 6.59 (m, 2H), 6.56 – 6.55 (m, 1H), 6.42 – 6.41 (m, 2H), 2.48 (s, 3H), 2.30 (s, 3H), 2.29 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 149.9, 149.0, 147.4, 145.5, 145.2, 144.9, 139.4, 138.1, 135.6, 133.9, 129.4, 129.3, 128.8, 128.3, 126.4, 125.2, 124.1, 123.6, 119.6, 118.9, 117.1, 115.9, 115.5, 113.6, 21.8, 21.5, 21.5.

HRMS-ESI (m/z) [M+K]⁺ calculated for C₂₉H₂₅KN₃O₂S 518.1299, found 518.1282.

Dimethyl 3,3'-(((2*E*,3*E*)-1-tosylindoline-2,3-diylidene)bis(azaneylylidene))dibenzoate (3m)



Following General Procedure A: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), methyl 3-aminobenzoate (185.1 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

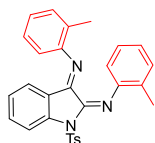
Red solid, 71.6 mg, 42% yield. (*R*_f = 0.30, petroleum ether/ethyl acetate = 10/1).

¹H NMR (600 MHz, CDCl₃) δ 8.22 (d, *J* = 8.4 Hz, 1H), 7.98 (d, *J* = 7.5 Hz, 2H), 7.76 (d, *J* = 7.7 Hz, 1H), 7.64 (d, *J* = 7.7 Hz, 1H), 7.45 (t, *J* = 8.0 Hz, 1H), 7.38 – 7.31 (m, 4H), 7.28 (d, *J* = 7.8 Hz, 1H), 7.23 (s, 1H), 6.94 (d, *J* = 7.1 Hz, 1H), 6.80 (t, *J* = 7.6 Hz, 1H), 6.75 (d, *J* = 7.8 Hz, 1H), 6.49 (d, *J* = 7.8 Hz, 1H), 3.87 (s, 3H), 3.84 (s, 3H), 2.47 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 167.1, 166.5, 149.5, 147.5, 145.8, 145.6, 145.4, 135.2, 134.5, 131.4, 130.5, 129.7, 129.5, 128.7, 128.6, 127.1, 126.3, 125.7, 124.5, 123.9, 123.6, 121.4, 120.0, 118.4, 117.7, 115.7, 52.2, 52.0, 21.8.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₃₁H₂₅N₃NaO₆S 590.1356, found 590.1351.

(2*E*,3*E*)-*N*²,*N*³-Di-*o*-tolyl-1-tosylindoline-2,3-diimine (3n)



Following General Procedure B: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), *o*-toluidine (129 μL, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

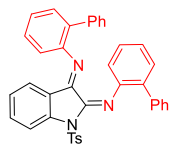
Red solid, 99.3 mg, 69% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.27 (d, *J* = 8.4 Hz, 1H), 8.07 (d, *J* = 8.2 Hz, 2H), 7.46 (t, *J* = 7.5 Hz, 1H), 7.35 (d, *J* = 8.2 Hz, 2H), 7.15 (d, *J* = 7.4 Hz, 1H), 7.12 – 6.98 (m, 4H), 6.89 (t, *J* = 7.1 Hz, 1H), 6.84 (t, *J* = 7.6 Hz, 1H), 6.64 (d, *J* = 7.7 Hz, 1H), 6.54 (d, *J* = 7.9 Hz, 1H), 6.52 (d, *J* = 7.9 Hz, 1H), 2.47 (s, 3H), 1.94 (s, 3H), 1.78 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 148.9, 148.4, 147.2, 145.6, 145.2, 144.7, 135.9, 134.0, 130.9, 129.9, 129.5, 128.7, 126.6, 126.5, 126.2, 126.0, 125.9, 124.8, 123.7, 122.7, 118.7, 117.3, 115.5, 115.2, 21.8, 17.6, 17.6.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₂₉H₂₅N₃NaO₂S 502.1560, found 502.1558.

(2*E*,3*E*)-*N*²,*N*³-di([1,1'-biphenyl]-2-yl)-1-tosylindoline-2,3-diimine (3o)



Following General Procedure B: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), [1,1'-biphenyl]-2-amine (209.0mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

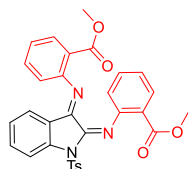
Red solid, 77.8 mg, 43% yield. (R_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.06 (d, *J* = 8.4 Hz, 1H), 7.92 (d, *J* = 8.1 Hz, 2H), 7.31 (t, *J* = 7.5 Hz, 2H), 7.27 (s, 1H), 7.26 – 7.16 (m, 10H), 7.16 – 7.07 (m, 4H), 6.92 (d, *J* = 7.0 Hz, 2H), 6.71 (t, *J* = 7.7 Hz, 1H), 6.51 (d, *J* = 7.8 Hz, 1H), 6.36 (d, *J* = 7.8 Hz, 1H), 6.31 (d, *J* = 7.6 Hz, 1H), 2.46 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 149.1, 147.8, 145.4, 145.1, 144.9, 143.6, 140.0, 138.5, 135.8, 133.7, 131.6, 130.5, 130.0, 129.8, 129.3, 129.2, 128.8, 128.7, 128.3, 128.0, 127.7, 127.2, 126.8, 126.5, 125.9, 125.1, 123.5, 123.4, 119.1, 118.6, 116.8, 114.8, 21.8.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₃₉H₃₀N₃O₂S 604.2053, found 604.2047.

Dimethyl 2,2'-(((2*E*,3*E*)-1-tosylindoline-2,3-diylidene)bis(azaneylylidene))dibenzoate (3p)



Following General Procedure B: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), methyl 2-aminobenzoate (185.1mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

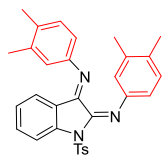
Red solid, 74.8 mg, 44% yield. (R_f = 0.30, petroleum ether/ethyl acetate = 10/1).

¹H NMR (600 MHz, CDCl₃) δ 8.20 (d, *J* = 8.4 Hz, 1H), 8.05 (d, *J* = 8.4 Hz, 2H), 7.85 (d, *J* = 6.7 Hz, 1H), 7.81 (d, *J* = 6.6 Hz, 1H), 7.41 (t, *J* = 7.3 Hz, 1H), 7.36 – 7.29 (m, 4H), 7.09 (t, *J* = 7.1 Hz, 1H), 6.94 (t, *J* = 7.0 Hz, 1H), 6.78 (t, *J* = 7.7 Hz, 1H), 6.67 (d, *J* = 7.6 Hz, 1H), 6.45 – 6.35 (m, 2H), 3.61 (s, 3H), 3.57 (s, 3H), 2.43 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 166.4, 165.7, 150.4, 149.4, 148.7, 145.6, 145.0, 144.5, 135.8, 134.0, 133.4, 132.7, 131.3, 130.7, 129.3, 128.9, 126.0, 123.9, 123.7, 122.3, 120.0, 118.7, 118.11, 118.08, 117.1, 115.2, 51.9, 51.5, 21.7.

HRMS-ESI (m/z) [M+K]⁺ calculated for C₃₁H₂₅KN₃O₆S 606.1096, found 606.1091.

(2E,3E)-N²,N³-Bis(3,4-dimethylphenyl)-1-tosylindoline-2,3-diimine (3q)



Following General Procedure A: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), 3,4-dimethylaniline (146.9 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

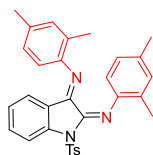
Red solid, 101.9 mg, 67% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.24 (d, *J* = 7.1 Hz, 1H), 8.01 (s, 2H), 7.44 (t, *J* = 7.9 Hz, 1H), 7.34 (d, *J* = 7.9 Hz, 2H), 7.06 (d, *J* = 7.9 Hz, 1H), 6.98 (d, *J* = 7.8 Hz, 1H), 6.83 (t, *J* = 7.6 Hz, 1H), 6.74 (d, *J* = 7.8 Hz, 1H), 6.64 (s, 1H), 6.56 (s, 1H), 6.43 (s, 1H), 6.39 (d, *J* = 7.8 Hz, 1H), 2.47 (s, 3H), 2.25 (s, 3H), 2.21 (s, 6H), 2.19 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 147.8, 145.4, 145.1, 144.9, 144.8, 137.8, 136.4, 135.7, 133.7, 132.7, 131.4, 130.4, 129.5, 129.4, 128.8, 126.2, 123.5, 120.7, 119.1, 118.1, 116.6, 115.4, 114.1, 21.8, 19.9, 19.3.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₃₁H₃₀N₃O₂S 508.2053, found 508.2043.

(2E,3E)-N²,N³-Bis(2,4-dimethylphenyl)-1-tosylindoline-2,3-diimine (3r)



Following General Procedure B: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), 2,4-dimethylaniline (150.0 μL, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

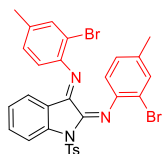
Red solid, 92.8 mg, 61% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.24 (d, *J* = 8.4 Hz, 1H), 8.03 (d, *J* = 8.1 Hz, 2H), 7.44 (t, *J* = 7.9 Hz, 1H), 7.32 (d, *J* = 8.1 Hz, 2H), 6.96 (s, 1H), 6.89 (d, *J* = 7.9 Hz, 1H), 6.86 (s, 1H), 6.85 – 6.80 (m, 2H), 6.74 (d, *J* = 7.7 Hz, 1H), 6.43 (d, *J* = 7.9 Hz, 1H), 6.41 (d, *J* = 7.9 Hz, 1H), 2.45 (s, 3H), 2.29 (s, 3H), 2.23 (s, 3H), 1.87 (s, 3H), 1.73 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 148.8, 145.9, 145.4, 145.0, 144.7, 144.5, 136.0, 134.4, 133.7, 132.0, 131.5, 130.6, 129.4, 128.7, 126.9, 126.8, 126.5, 126.1, 125.7, 123.5, 118.9, 117.3, 115.5, 115.1, 21.7, 20.9, 20.8, 17.5.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₃₁H₃₀N₃O₂S 508.2053, found 508.2065.

(2E,3E)-N²,N³-Bis(2-bromo-4-methylphenyl)-1-tosylindoline-2,3-diimine (3s)



Following General Procedure B: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), 2-bromo-4-methylaniline (227.8 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

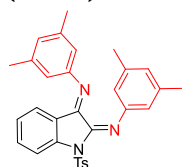
Red solid, 87.9 mg, 46% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 10/1).

¹H NMR (600 MHz, CDCl₃) δ 8.23 (d, *J* = 8.4 Hz, 1H), 8.04 (d, *J* = 8.0 Hz, 2H), 7.46 (t, *J* = 8.0 Hz, 1H), 7.35 (s, 1H), 7.31 (d, *J* = 8.1 Hz, 2H), 7.21 (s, 1H), 7.01 (d, *J* = 7.9 Hz, 1H), 6.95 (d, *J* = 7.9 Hz, 1H), 6.85 (t, *J* = 7.6 Hz, 1H), 6.72 (d, *J* = 8.0 Hz, 1H), 6.62 (d, *J* = 7.7 Hz, 1H), 6.52 (d, *J* = 8.0 Hz, 1H), 2.43 (s, 3H), 2.29 (s, 3H), 2.22 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 150.3, 146.1, 145.6, 145.3, 144.1, 135.7, 135.5, 134.4, 133.7, 133.6, 132.6, 129.5, 128.92, 128.88, 128.4, 126.1, 123.9, 119.8, 118.5, 117.5, 115.2, 112.2, 111.6, 21.8, 20.6, 20.5.

HRMS-ESI (*m/z*) [*M*+Na]⁺ calculated for C₂₉H₂₃Br₂N₃NaO₂S 657.9770, found 657.9758.

(2*E*,3*E*)-*N*²,*N*³-Bis(3,5-dimethylphenyl)-1-tosylindoline-2,3-diimine (3t)



Following General Procedure A: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), 3,5-dimethylaniline (150.0 μL, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

Red solid, 103.4 mg, 68% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

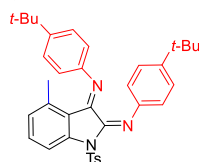
¹H NMR (600 MHz, CDCl₃) δ 8.24 (d, *J* = 8.2 Hz, 1H), 8.02 (d, *J* = 6.7 Hz, 2H), 7.44 (t, *J* = 7.8 Hz, 1H), 7.35 (d, *J* = 7.8 Hz, 2H), 6.84 (t, *J* = 7.5 Hz, 1H), 6.75 (s, 1H), 6.67 (d, *J* = 7.7 Hz, 1H), 6.64 (s, 1H), 6.40 (s, 2H), 6.24 (s, 2H), 2.48 (s, 3H), 2.26 (s, 12H).

¹³C NMR (150 MHz, CDCl₃) δ 150.1, 148.8, 147.3, 145.5, 145.1, 144.9, 139.2, 137.9, 135.6, 133.7, 129.4, 128.7, 126.4, 126.0, 125.0, 123.6, 119.0, 116.6, 115.5, 114.0, 21.8, 21.4, 21.4.

HRMS-ESI (*m/z*) [*M*+H]⁺ calculated for C₃₁H₃₀N₃O₂S 508.2053, found 508.2064.

Scope of 2-ethynylaniline

(2*E*,3*E*)-*N*²,*N*³-Bis(4-(*tert*-butyl)phenyl)-4-methyl-1-tosylindoline-2,3-diimine (3u)



Following General Procedure B: *N*-(2-Ethynyl-3-methylphenyl)-4-methylbenzenesulfonamide (87.4 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μL, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

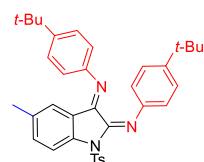
Red solid, 67.6 mg, 39% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 7.93 (d, *J* = 8.3 Hz, 1H), 7.88 (d, *J* = 8.1 Hz, 2H), 7.40 (t, *J* = 8.0 Hz, 1H), 7.22 (d, *J* = 8.2 Hz, 2H), 7.00 (d, *J* = 8.3 Hz, 3H), 6.94 (d, *J* = 8.2 Hz, 2H), 6.28 (d, *J* = 8.3 Hz, 2H), 6.25 – 6.15 (m, 2H), 2.57 (s, 3H), 2.38 (s, 3H), 1.30 (s, 9H), 1.28 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 150.6, 147.6, 147.3, 146.6, 144.8, 144.2, 144.0, 138.2, 135.1, 132.3, 129.4, 128.5, 127.0, 125.7, 125.4, 123.3, 119.7, 119.1, 112.8, 34.3, 34.2, 31.43, 31.43, 21.7, 19.5.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₃₆H₃₉N₃NaO₂S 600.2655, found 600.2656.

(2*E*,3*E*)-*N*²,*N*³-Bis(4-(*tert*-butyl)phenyl)-5-methyl-1-tosylindoline-2,3-diimine (3v)



Following General Procedure A: *N*-(2-Ethynyl-4-methylphenyl)-4-methylbenzenesulfonamide (87.4 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μL, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

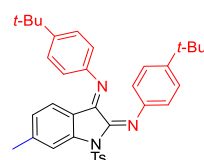
Red solid, 152.6 mg, 88% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.13 (d, *J* = 7.0 Hz, 1H), 8.01 (s, 2H), 7.35 (t, *J* = 8.2 Hz, 4H), 7.28 (d, *J* = 8.2 Hz, 2H), 7.25 (s, 1H), 6.80 (s, 2H), 6.59 (d, *J* = 8.1 Hz, 2H), 6.32 (s, 1H), 2.47 (s, 3H), 2.06 (s, 3H), 1.36 (s, 9H), 1.33 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 149.7, 147.9, 147.7, 146.2, 145.1, 145.0, 144.5, 143.4, 135.7, 134.5, 133.0, 129.4, 128.7, 126.5, 126.1, 125.2, 119.2, 116.9, 115.2, 34.5, 34.3, 31.6, 21.8, 20.8.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₃₆H₄₀N₃O₂S 578.2836, found 578.2823.

(2*E*,3*E*)-*N*²,*N*³-Bis(4-(*tert*-butyl)phenyl)-6-methyl-1-tosylindoline-2,3-diimine (3w)



Following General Procedure A: *N*-(2-Ethynyl-5-methylphenyl)-4-methylbenzenesulfonamide (87.4 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μL, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

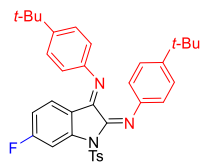
Red solid, 112.7 mg, 65% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.05 (s, 1H), 8.00 (s, 2H), 7.34 (d, *J* = 8.0 Hz, 2H), 7.30 (d, *J* = 8.3 Hz, 2H), 7.23 (d, *J* = 8.2 Hz, 2H), 6.74 (s, 2H), 6.64 (d, *J* = 8.0 Hz, 1H), 6.57 (d, *J* = 8.2 Hz, 3H), 2.47 (s, 3H), 2.42 (s, 3H), 1.32 (s, 9H), 1.29 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 147.6, 147.3, 146.1, 145.7, 145.14, 145.02, 144.97, 144.3, 135.8, 129.3, 128.8, 126.1, 125.8, 125.1, 124.4, 119.1, 117.1, 116.7, 115.7, 34.4, 34.3, 31.5, 31.5, 22.6, 21.8.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₃₆H₄₀N₃O₂S 578.2836, found 578.2831.

(2E,3E)-N²,N³-Bis(4-(*tert*-butyl)phenyl)-6-fluoro-1-tosylindoline-2,3-diimine (3y)



Following General Procedure A: *N*-(2-Ethynyl-5-fluorophenyl)-4-methylbenzenesulfonamide (88.6 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

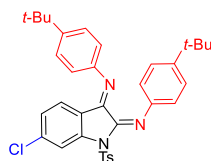
Red solid, 144.7 mg, 83% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.02 (d, *J* = 7.6 Hz, 2H), 7.98 (d, *J* = 10.0 Hz, 1H), 7.36 (d, *J* = 8.0 Hz, 2H), 7.32 (d, *J* = 8.3 Hz, 2H), 7.24 (d, *J* = 8.3 Hz, 2H), 6.76 (d, *J* = 7.7 Hz, 2H), 6.71 – 6.64 (m, 1H), 6.58 (d, *J* = 8.3 Hz, 2H), 6.51 (t, *J* = 7.8 Hz, 1H), 2.48 (s, 3H), 1.32 (s, 9H), 1.30 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 166.2, 164.6, 147.9, 147.2, 147.1, 146.9, 146.4, 145.4, 144.6, 143.9, 135.5, 129.4, 128.9, 128.0, 127.9, 126.2, 125.2, 119.1, 116.9, 115.4, 110.8, 110.6, 103.7, 103.5, 34.5, 34.3, 31.48, 31.46, 21.8.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₃₅H₃₇FN₃O₂S 582.2585, found 582.2578.

(2E,3E)-N²,N³-Bis(4-(*tert*-butyl)phenyl)-6-chloro-1-tosylindoline-2,3-diimine (3z)



Following General Procedure A: *N*-(5-Chloro-2-ethynylphenyl)-4-methylbenzenesulfonamide (93.6 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

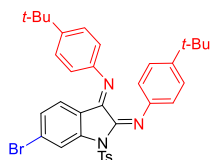
Red solid, 134.6 mg, 75% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.30 (s, 1H), 8.05 (d, *J* = 7.5 Hz, 2H), 7.37 (d, *J* = 8.1 Hz, 2H), 7.34 (d, *J* = 8.3 Hz, 2H), 7.28 (s, 2H), 6.86 – 6.74 (m, 3H), 6.68 – 6.63 (m, 1H), 6.60 (d, *J* = 8.3 Hz, 2H), 2.49 (s, 3H), 1.35 (s, 9H), 1.32 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 148.1, 146.9, 146.5, 146.2, 145.4, 144.4, 144.0, 139.7, 135.5, 129.4, 128.9, 126.9, 126.2, 126.1, 125.2, 123.8, 119.2, 116.9, 115.7, 115.0, 34.5, 34.4, 31.5, 31.5, 21.8.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₃₅H₃₆ClN₃NaO₂S 620.2109, found 620.2103.

(2E,3E)-6-Bromo-N²,N³-bis(4-(*tert*-butyl)phenyl)-1-tosylindoline-2,3-diimine (3aa)



Following General Procedure A: *N*-(5-Bromo-2-ethynylphenyl)-4-methylbenzenesulfonamide (116.7 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

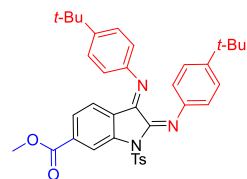
Red solid, 135.0 mg, 70% yield. (R_f = 0.20, petroleum ether/ethyl acetate = 20/1).

^1H NMR (600 MHz, CDCl_3) δ 8.46 (s, 1H), 8.03 (d, J = 7.1 Hz, 2H), 7.37 (d, J = 7.8 Hz, 2H), 7.33 (d, J = 8.0 Hz, 2H), 7.25 (d, J = 7.3 Hz, 2H), 6.97 (d, J = 8.3 Hz, 1H), 6.77 (d, J = 7.0 Hz, 2H), 6.64 – 6.47 (m, 3H), 2.49 (s, 3H), 1.33 (s, 9H), 1.31 (s, 9H).

^{13}C NMR (150 MHz, CDCl_3) δ 148.2, 146.9, 146.5, 146.1, 145.4, 144.3, 144.1, 135.5, 131.0, 129.4, 128.9, 128.3, 126.9, 126.7, 126.2, 125.2, 119.2, 118.5, 117.6, 116.9, 34.5, 34.3, 31.51, 31.49, 21.8.

HRMS-ESI (m/z) $[\text{M}+\text{Na}]^+$ calculated for $\text{C}_{35}\text{H}_{36}\text{BrN}_3\text{NaO}_2\text{S}$ 664.1604, found 664.1597.

Methyl (2*E*,3*E*)-2,3-bis((4-(*tert*-butyl)phenyl)imino)-1-tosylindoline-6-carboxylate (3ab)



Following General Procedure A: Methyl 4-ethynyl-3-((4-methylphenyl)sulfonamido)benzoate (100.7 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μL , 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO_4 (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H_2O (1 mL), CH_3CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

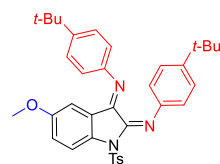
Red solid, 61.5 mg, 33% yield. (R_f = 0.30, petroleum ether/ethyl acetate = 10/1).

^1H NMR (600 MHz, CDCl_3) δ 8.83 (s, 1H), 8.02 (s, 2H), 7.51 (d, J = 8.1 Hz, 1H), 7.34 (d, J = 8.0 Hz, 2H), 7.32 (d, J = 8.3 Hz, 2H), 7.23 (d, J = 8.1 Hz, 2H), 6.82 – 6.68 (m, 3H), 6.57 (d, J = 8.2 Hz, 2H), 3.96 (s, 3H), 2.47 (s, 3H), 1.31 (s, 9H), 1.28 (s, 9H).

^{13}C NMR (150 MHz, CDCl_3) δ 165.9, 148.5, 146.7, 146.5, 145.4, 145.3, 144.4, 144.0, 135.5, 134.4, 129.4, 128.9, 126.2, 126.1, 125.8, 125.2, 124.7, 119.1, 116.9, 116.2, 114.9, 52.7, 34.5, 34.3, 31.5, 31.4, 21.8.

HRMS-ESI (m/z) $[\text{M}+\text{Na}]^+$ calculated for $\text{C}_{37}\text{H}_{39}\text{N}_3\text{NaO}_4\text{S}$ 644.2553, found 644.2557.

(2*E*,3*E*)-*N*²,*N*³-Bis(4-(*tert*-butyl)phenyl)-5-methoxy-1-tosylindoline-2,3-diimine (3ac)



Following General Procedure A: *N*-(2-Ethynyl-4-methoxyphenyl)-4-methylbenzenesulfonamide (92.3 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μL , 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO_4 (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H_2O (1 mL), CH_3CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

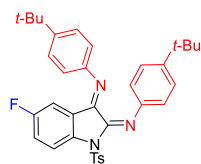
Red solid, 114.1 mg, 64% yield. (R_f = 0.20, petroleum ether/ethyl acetate = 10/1).

^1H NMR (600 MHz, CDCl_3) δ 8.11 (s, 1H), 7.96 (s, 2H), 7.35 (d, J = 8.4 Hz, 2H), 7.32 (d, J = 8.0 Hz, 2H), 7.27 – 7.24 (m, 2H), 7.00 (dd, J = 9.1, 2.5 Hz, 1H), 6.77 (s, 2H), 6.59 (d, J = 8.3 Hz, 2H), 6.04 (s, 1H), 3.36 (s, 3H), 2.46 (s, 3H), 1.31 (s, 9H), 1.31 (s, 9H).

^{13}C NMR (150 MHz, CDCl_3) δ 155.5, 147.7, 147.5, 146.2, 145.2, 145.0, 144.4, 139.5, 135.6, 129.4, 128.6, 126.1, 125.2, 121.4, 119.2, 116.9, 109.2, 55.0, 34.4, 34.3, 31.5, 31.5, 21.8.

HRMS-ESI (m/z) $[\text{M}+\text{H}]^+$ calculated for $\text{C}_{36}\text{H}_{40}\text{N}_3\text{O}_3\text{S}$ 594.2785, found 594.2776.

(2E,3E)-N²,N³-Bis(4-(*tert*-butyl)phenyl)-5-fluoro-1-tosylindoline-2,3-diimine (3ad)



Following General Procedure A: *N*-(2-Ethynyl-4-fluorophenyl)-4-methylbenzenesulfonamide (88.6 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

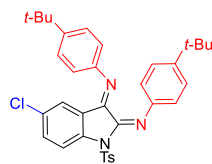
Red solid, 142.9 mg, 82% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.21 (s, 1H), 7.99 (s, 2H), 7.35 (d, *J* = 7.9 Hz, 4H), 7.30 – 7.26 (m, 2H), 7.16 (t, *J* = 7.7 Hz, 1H), 6.77 (s, 2H), 6.58 (d, *J* = 8.1 Hz, 2H), 6.35 (d, *J* = 6.6 Hz, 1H), 2.47 (s, 3H), 1.34 (s, 9H), 1.31 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 159.1, 157.5, 148.4, 146.6, 146.5, 145.3, 144.6, 144.1, 141.7, 135.5, 129.4, 128.8, 126.3, 125.2, 120.7, 120.5, 119.1, 116.8, 112.8, 112.6, 34.5, 34.3, 31.50, 31.45, 21.8.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₃₅H₃₆FN₃NaO₂S 604.2404, found 604.2391.

(2E,3E)-N²,N³-Bis(4-(*tert*-butyl)phenyl)-5-chloro-1-tosylindoline-2,3-diimine (3ae)



Following General Procedure A: *N*-(4-Chloro-2-ethynylphenyl)-4-methylbenzenesulfonamide (93.6 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

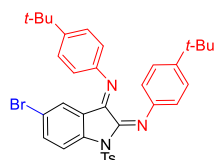
Red solid, 107.6 mg, 60% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.18 (d, *J* = 8.6 Hz, 1H), 7.99 (s, 2H), 7.45 – 7.30 (m, 5H), 7.25 (s, 2H), 6.77 (s, 2H), 6.57 (d, *J* = 8.1 Hz, 2H), 6.48 (s, 1H), 2.48 (s, 3H), 1.34 (s, 9H), 1.30 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 148.5, 147.0, 146.5, 145.3, 144.3, 144.0, 143.8, 135.4, 133.4, 129.4, 128.8, 126.3, 126.0, 125.2, 119.2, 116.7, 34.5, 34.3, 31.5, 31.5, 21.8.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₃₅H₃₆ClN₃NaO₂S 620.2109, found 620.2111.

(2E,3E)-5-Bromo-N²,N³-bis(4-(*tert*-butyl)phenyl)-1-tosylindoline-2,3-diimine (3af)



Following General Procedure A: *N*-(4-Bromo-2-ethynylphenyl)-4-methylbenzenesulfonamide (107.2 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

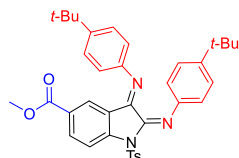
Red solid, 158.2 mg, 82% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.13 (d, *J* = 8.5 Hz, 1H), 8.00 (d, *J* = 5.0 Hz, 2H), 7.52 (d, *J* = 8.7 Hz, 1H), 7.36 (t, *J* = 7.2 Hz, 4H), 7.29 – 7.23 (m, 2H), 6.78 (s, 2H), 6.57 (d, *J* = 7.4 Hz, 3H), 2.48 (s, 3H), 1.35 (s, 9H), 1.31 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 148.5, 147.1, 146.6, 145.4, 144.3, 144.1, 144.0, 136.2, 135.5, 129.4, 129.0, 128.8, 126.3, 125.2, 120.3, 119.2, 116.99, 116.96, 116.6, 116.4, 34.5, 34.4, 31.50, 31.48, 21.8.

HRMS-ESI (m/z) [M+K]⁺ calculated for C₃₅H₃₆BrKN₃O₂S 680.1343, found 680.1348.

Methyl (2*E*,3*E*)-2,3-bis((4-(*tert*-butyl)phenyl)imino)-1-tosylindoline-5-carboxylate (3ag)



Following General Procedure A: Methyl 3-ethynyl-4-((4-methylphenyl)sulfonamido)benzoate (100.7 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μL, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

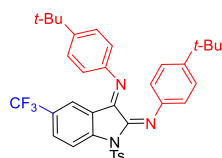
Red solid, 100.8 mg, 54% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 10/1).

¹H NMR (600 MHz, CDCl₃) δ 8.29 (d, *J* = 8.6 Hz, 1H), 8.12 (d, *J* = 8.6 Hz, 1H), 8.02 (d, *J* = 7.1 Hz, 2H), 7.45 – 7.32 (m, 4H), 7.28 – 7.24 (m, 3H), 6.78 (d, *J* = 7.1 Hz, 2H), 6.61 (d, *J* = 7.8 Hz, 2H), 3.73 (s, 3H), 2.48 (s, 3H), 1.36 (s, 9H), 1.30 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 165.6, 148.6, 148.5, 148.3, 147.1, 146.6, 145.5, 144.3, 143.8, 135.5, 135.2, 129.4, 128.9, 127.5, 126.3, 126.1, 125.2, 119.2, 118.6, 116.6, 115.0, 52.1, 34.5, 34.3, 31.6, 31.5, 21.8.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₃₇H₃₉N₃NaO₄S 644.2553, found 644.2543.

(2*E*,3*E*)-*N*²,*N*³-Bis(4-(*tert*-butyl)phenyl)-1-tosyl-5-(trifluoromethyl)indoline-2,3-diimine (3ah)



Following General Procedure A: *N*-(2-Ethynyl-4-(trifluoromethyl)phenyl)-4-methylbenzenesulfonamide (103.9 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μL, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

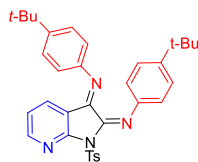
Red solid, 92.9 mg, 49% yield. (*R*_f = 0.30, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.35 (d, *J* = 8.7 Hz, 1H), 8.02 (d, *J* = 7.6 Hz, 2H), 7.65 (d, *J* = 8.7 Hz, 1H), 7.45 – 7.32 (m, 4H), 7.29 – 7.26 (m, 2H), 6.79 (d, *J* = 7.8 Hz, 2H), 6.66 (s, 1H), 6.59 (d, *J* = 8.0 Hz, 2H), 2.49 (s, 3H), 1.33 (s, 9H), 1.30 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 148.7, 147.6, 147.2, 146.8, 145.6, 144.0, 143.7, 135.4, 130.4, 129.5, 128.9, 126.4, 126.1, 125.8, 125.6, 125.4, 125.2, 124.3, 123.42, 123.40, 122.5, 119.2, 118.7, 116.4, 115.5, 34.5, 34.4, 31.5, 31.4, 21.8.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₃₆H₃₆F₃N₃NaO₂S 654.2373, found 654.2364.

(2E,3E)-N²,N³-Bis(4-(*tert*-butyl)phenyl)-1-tosyl-1H-pyrrolo[2,3-b]pyridine-2,3-diimine (3ai)



Following General Procedure A: *N*-(3-ethynylpyridin-2-yl)-4-methylbenzenesulfonamide (83.4 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

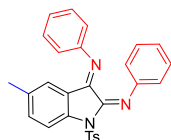
Red solid, 64.4 mg, 38% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.54 – 8.44 (m, 1H), 8.14 (s, 2H), 7.35 (t, *J* = 8.8 Hz, 4H), 7.26 (d, *J* = 8.4 Hz, 2H), 6.97 (d, *J* = 6.3 Hz, 1H), 6.91 – 6.78 (m, 3H), 6.66 (s, 2H), 2.47 (s, 3H), 1.34 (s, 9H), 1.32 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 152.0, 148.6, 146.8, 146.6, 145.0, 136.2, 133.9, 129.3, 128.8, 126.3, 125.3, 119.2, 117.2, 34.5, 34.3, 31.5, 31.4, 21.8.

HRMS-ESI (m/z) [M+K]⁺ calculated for C₃₄H₃₆KN₄O₂S 603.2191, found 603.2190.

(2E,3E)-5-Methyl-N²,N³-diphenyl-1-tosylindoline-2,3-diimine (3aj)



Following General Procedure A: *N*-(2-Ethynyl-4-methylphenyl)-4-methylbenzenesulfonamide (87.4 mg, 0.3 mmol), aniline (110 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

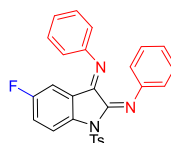
Red solid, 97.9 mg, 70% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.12 (d, *J* = 8.3 Hz, 1H), 8.05 – 7.87 (m, 2H), 7.34 (d, *J* = 8.0 Hz, 2H), 7.30 – 7.25 (m, 3H), 7.22 (t, *J* = 7.6 Hz, 2H), 7.10 (t, *J* = 7.4 Hz, 1H), 6.98 (t, *J* = 7.3 Hz, 1H), 6.74 (s, 2H), 6.58 (d, *J* = 7.7 Hz, 2H), 6.36 (s, 1H), 2.47 (s, 3H), 2.05 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 149.8, 147.6, 145.3, 145.2, 143.5, 135.5, 134.8, 133.3, 129.4, 129.4, 128.7, 128.5, 126.5, 124.5, 123.2, 118.88, 118.86, 116.8, 115.4, 115.4, 21.8, 20.8.

HRMS-ESI (m/z) [M+K]⁺ calculated for C₂₈H₂₃KN₃O₂S 504.1143, found 504.1148.

(2E,3E)-5-Fluoro-N²,N³-diphenyl-1-tosylindoline-2,3-diimine (3ak)



Following General Procedure B: *N*-(2-Ethynyl-4-fluorophenyl)-4-methylbenzenesulfonamide (88.6 mg, 0.3 mmol), aniline (110 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

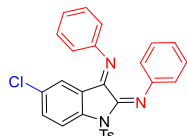
Red solid, 40.8 mg, 29% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.27 – 8.14 (m, 1H), 8.10 – 7.87 (m, 2H), 7.35 (d, *J* = 8.0 Hz, 2H), 7.29 (t, *J* = 7.7 Hz, 2H), 7.22 (t, *J* = 7.6 Hz, 2H), 7.16 (td, *J* = 8.9, 2.6 Hz, 1H), 7.11 (t, *J* = 7.4 Hz, 1H), 6.98 (t, *J* = 7.3 Hz, 1H), 6.80 – 6.62 (m, 2H), 6.56 (d, *J* = 7.7 Hz, 2H), 6.25 (d, *J* = 8.1 Hz, 1H), 2.47 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 159.2, 157.5, 149.1, 147.2, 145.4, 144.8, 141.6, 135.2, 129.6, 129.5, 128.8, 128.5, 125.0, 123.4, 121.0, 120.8, 119.5, 118.8, 116.9, 116.6, 112.9, 112.7, 21.8.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₂₇H₂₁FN₃O₂S 470.1333, found 470.1326.

(2*E*,3*E*)-5-Chloro-*N*²,*N*³-diphenyl-1-tosylindoline-2,3-diimine (3al)



Following General Procedure A: *N*-(4-Chloro-2-ethynylphenyl)-4-methylbenzenesulfonamide (93.6 mg, 0.3 mmol), aniline (110 μL, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

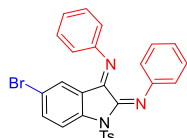
Red solid, 86.0 mg, 59% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.19 (d, *J* = 8.9 Hz, 1H), 7.99 (d, *J* = 7.3 Hz, 2H), 7.40 (d, *J* = 7.0 Hz, 1H), 7.35 (d, *J* = 8.1 Hz, 2H), 7.30 (t, *J* = 7.7 Hz, 2H), 7.22 (t, *J* = 7.7 Hz, 2H), 7.13 (t, *J* = 7.4 Hz, 1H), 6.99 (t, *J* = 7.3 Hz, 1H), 6.73 (d, *J* = 6.6 Hz, 2H), 6.58 (d, *J* = 7.7 Hz, 2H), 6.53 (d, *J* = 1.7 Hz, 1H), 2.48 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 149.1, 147.1, 145.5, 144.5, 144.0, 135.2, 133.7, 129.6, 129.5, 129.1, 128.8, 128.5, 125.9, 125.1, 123.5, 119.7, 118.8, 116.7, 116.6, 21.8.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₂₇H₂₀ClN₃NaO₂S 508.0857, found 508.0849.

(2*E*,3*E*)-5-Bromo-*N*²,*N*³-diphenyl-1-tosylindoline-2,3-diimine (3am)



Following General Procedure A: *N*-(4-Bromo-2-ethynylphenyl)-4-methylbenzenesulfonamide (107.2 mg, 0.3 mmol), aniline (110 μL, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

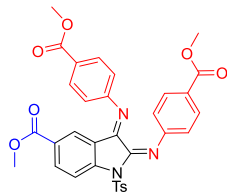
Red solid, 109.7 mg, 69% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.14 (d, *J* = 8.9 Hz, 1H), 7.99 (d, *J* = 7.5 Hz, 2H), 7.54 (dd, *J* = 8.9, 1.8 Hz, 1H), 7.35 (d, *J* = 8.1 Hz, 2H), 7.30 (t, *J* = 7.7 Hz, 2H), 7.22 (t, *J* = 7.7 Hz, 2H), 7.13 (t, *J* = 7.4 Hz, 1H), 6.99 (t, *J* = 7.3 Hz, 1H), 6.73 (d, *J* = 7.0 Hz, 2H), 6.67 (s, 1H), 6.58 (d, *J* = 7.7 Hz, 2H), 2.48 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 149.1, 147.8, 147.1, 145.6, 144.5, 144.4, 136.5, 135.2, 129.6, 129.5, 128.9, 128.8, 128.5, 125.1, 123.5, 120.1, 118.8, 117.1, 116.6, 116.5, 21.8.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₂₇H₂₀BrN₃NaO₂S 552.0352, found 552.0349.

Dimethyl 4,4'-(((2*E*,3*E*)-5-(methoxycarbonyl)-1-tosylindoline-2,3-diylidene)bis(azaneylylidene))dibenzoate (3an)



Following General Procedure A: Methyl 3-ethynyl-4-((4-methylphenyl)sulfonamido)benzoate (100.8 mg, 0.3 mmol), methyl 4-aminobenzoate (185.1, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

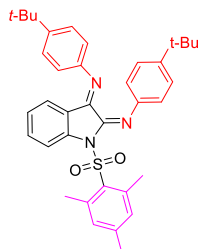
Red solid, 75.1 mg, 40% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 10/1).

¹H NMR (600 MHz, CDCl₃) δ 8.28 (d, *J* = 8.8 Hz, 1H), 8.13 (dd, *J* = 8.8, 1.8 Hz, 1H), 7.99 (d, *J* = 3.4 Hz, 2H), 7.98 (d, *J* = 8.4 Hz, 2H), 7.91 (d, *J* = 8.5 Hz, 2H), 7.36 (d, *J* = 8.1 Hz, 2H), 7.23 (s, 1H), 6.69 (d, *J* = 8.3 Hz, 2H), 6.61 (d, *J* = 8.5 Hz, 2H), 3.89 (s, 3H), 3.84 (s, 3H), 3.72 (s, 3H), 2.48 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 166.9, 166.4, 165.2, 152.7, 151.7, 148.8, 148.0, 146.1, 144.3, 135.9, 134.9, 131.4, 130.6, 129.6, 128.8, 127.8, 126.8, 125.9, 125.1, 118.1, 116.4, 115.1, 52.3, 52.1, 51.9, 21.8.

HRMS-ESI (m/z) [M+K]⁺ calculated for C₃₃H₂₇KN₃O₈S 664.1150, found 664.1143.

(2*E*,3*E*)-*N*²,*N*³-Bis(4-(*tert*-butyl)phenyl)-1-(mesitylsulfonyl)indoline-2,3-diimine (3ao)



Following General Procedure A: *N*-(2-Ethynylphenyl)-2,4,6-trimethylbenzenesulfonamide (91.7 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μL, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

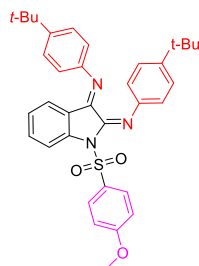
Red solid, 140.3 mg, 79% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.23 (d, *J* = 8.2 Hz, 1H), 7.43 (t, *J* = 7.7 Hz, 1H), 7.38 (d, *J* = 7.8 Hz, 2H), 7.20 (d, *J* = 7.9 Hz, 2H), 7.02 (s, 2H), 6.82 (t, *J* = 7.3 Hz, 1H), 6.77 (d, *J* = 7.5 Hz, 1H), 6.69 (d, *J* = 7.8 Hz, 2H), 6.64 (d, *J* = 7.9 Hz, 2H), 2.67 (s, 6H), 2.37 (s, 3H), 1.37 (s, 9H), 1.30 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 149.1, 147.9, 147.2, 146.3, 146.3, 144.8, 143.6, 143.3, 141.1, 134.2, 133.7, 131.8, 126.23, 126.18, 124.9, 122.9, 119.6, 118.3, 117.1, 115.6, 34.5, 34.3, 31.54, 31.51, 22.9, 21.2.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₃₇H₄₂N₃O₂S 592.2992, found 592.2991.

(2*E*,3*E*)-*N*²,*N*³-Bis(4-(*tert*-butyl)phenyl)-1-((4-methoxyphenyl)sulfonyl)indoline-2,3-diimine (3ap)



Following General Procedure A: *N*-(2-Ethynylphenyl)-4-methoxybenzenesulfonamide (87.9 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

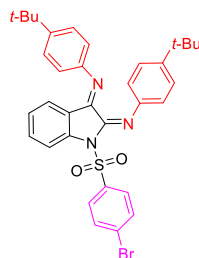
Red solid, 121.8 mg, 70% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 10/1).

¹H NMR (600 MHz, CDCl₃) δ 8.23 (d, *J* = 7.8 Hz, 1H), 8.16 – 7.99 (m, 2H), 7.42 (t, *J* = 7.8 Hz, 1H), 7.32 (d, *J* = 8.1 Hz, 2H), 7.25 (d, *J* = 6.6 Hz, 2H), 6.99 (d, *J* = 8.5 Hz, 2H), 6.87 – 6.74 (m, 3H), 6.72 (d, *J* = 7.6 Hz, 1H), 6.59 (d, *J* = 8.1 Hz, 2H), 3.89 (s, 3H), 1.33 (s, 9H), 1.31 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 164.1, 147.8, 147.1, 146.1, 145.6, 144.9, 144.5, 133.7, 131.1, 130.1, 126.1, 126.0, 125.2, 123.4, 119.1, 117.0, 115.4, 115.4, 114.9, 113.9, 55.8, 34.5, 34.3, 31.5, 31.5.

HRMS-ESI (*m/z*) [*M*+Na]⁺ calculated for C₃₅H₃₇N₃NaO₃S 602.2448, found 602.2443.

(2*E*,3*E*)-1-((4-bromophenyl)sulfonyl)-*N*²,*N*³-bis(4-(*tert*-butyl)phenyl)indoline-2,3-diimine (3aq)



Following General Procedure A: 4-Bromo-*N*-(2-ethynylphenyl)benzenesulfonamide (102.9 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

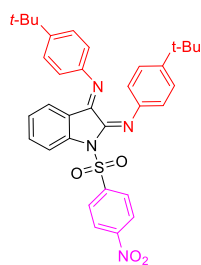
Red solid, 171.7 mg, 91% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.24 (d, *J* = 8.0 Hz, 1H), 8.12 – 7.95 (m, 2H), 7.68 (d, *J* = 8.1 Hz, 2H), 7.45 (t, *J* = 7.7 Hz, 1H), 7.35 (d, *J* = 7.9 Hz, 2H), 7.29 (d, *J* = 7.6 Hz, 2H), 6.87 – 6.75 (m, 4H), 6.63 (d, *J* = 7.9 Hz, 2H), 1.36 (s, 9H), 1.33 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 148.0, 145.0, 146.5, 145.1, 144.7, 144.1, 137.6, 133.9, 132.1, 130.3, 129.3, 126.2, 126.1, 125.3, 123.8, 119.2, 117.0, 115.3, 115.0, 34.5, 34.4, 31.7, 31.6.

HRMS-ESI (*m/z*) [*M*+H]⁺ calculated for C₃₄H₃₅BrN₃O₂S 628.1628, found 628.1621.

(2*E*,3*E*)-*N*²,*N*³-Bis(4-(*tert*-butyl)phenyl)-1-((4-nitrophenyl)sulfonyl)indoline-2,3-diimine (3ar)



Following General Procedure A: *N*-(2-Ethynylphenyl)-4-nitrobenzenesulfonamide (92.5 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

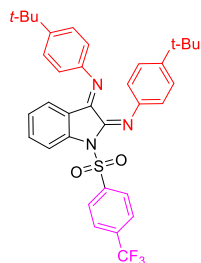
Red solid, 126.7 mg, 71% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.37 (d, *J* = 8.5 Hz, 2H), 8.32 (d, *J* = 8.3 Hz, 2H), 8.22 (d, *J* = 8.3 Hz, 1H), 7.47 (t, *J* = 7.8 Hz, 1H), 7.32 (d, *J* = 8.1 Hz, 2H), 7.24 (d, *J* = 8.2 Hz, 2H), 6.87 (t, *J* = 7.6 Hz, 1H), 6.77 (d, *J* = 7.7 Hz, 1H), 6.73 (d, *J* = 7.8 Hz, 2H), 6.59 (d, *J* = 8.1 Hz, 2H), 1.32 (s, 9H), 1.29 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 150.7, 148.39, 148.38, 148.2, 146.9, 146.8, 144.7, 144.5, 144.1, 143.4, 133.9, 130.2, 126.3, 126.2, 125.3, 124.1, 123.9, 119.2, 116.9, 115.1, 34.5, 34.4, 31.5, 31.5.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₃₄H₃₄N₄NaO₄S 617.2193, found 617.2195.

(2*E*,3*E*)-*N*²,*N*³-Bis(4-(*tert*-butyl)phenyl)-1-((4-(trifluoromethyl)phenyl)sulfonyl)indoline-2,3-diimine (3as)



Following General Procedure A: *N*-(2-Ethynylphenyl)-4-(trifluoromethyl)benzenesulfonamide (99.6 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

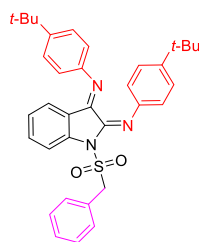
Red solid, 81.6 mg, 44% yield. (*R*_f = 0.30, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.28 (d, *J* = 7.1 Hz, 2H), 8.23 (d, *J* = 8.3 Hz, 1H), 7.82 (d, *J* = 8.2 Hz, 2H), 7.46 (t, *J* = 7.8 Hz, 1H), 7.33 (d, *J* = 8.3 Hz, 2H), 7.25 (d, *J* = 8.9 Hz, 2H), 6.86 (t, *J* = 7.6 Hz, 1H), 6.77 (d, *J* = 7.8 Hz, 1H), 6.73 (d, *J* = 7.1 Hz, 2H), 6.60 (d, *J* = 8.3 Hz, 2H), 1.33 (s, 9H), 1.30 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 148.6, 148.1, 146.9, 146.6, 144.9, 144.6, 143.8, 142.1, 135.8, 135.5, 135.3, 135.1, 133.8, 129.4, 126.21, 126.17, 125.92, 125.87, 125.84, 125.82, 125.3, 124.1, 123.9, 122.3, 120.5, 119.1, 116.9, 115.2, 77.3, 77.1, 76.9, 34.5, 34.3, 31.52, 31.46.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₃₅H₃₅F₃N₃O₂S 618.2397, found 618.2387.

(2*E*,3*E*)-1-(Benzylsulfonyl)-*N*²,*N*³-bis(4-(*tert*-butyl)phenyl)indoline-2,3-diimine (3at)



Following General Procedure A: *N*-(2-Ethynylphenyl)-1-phenylmethanesulfonamide (83.1 mg, 0.3 mmol), 4-(*tert*-butyl)aniline (193 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 48 h.

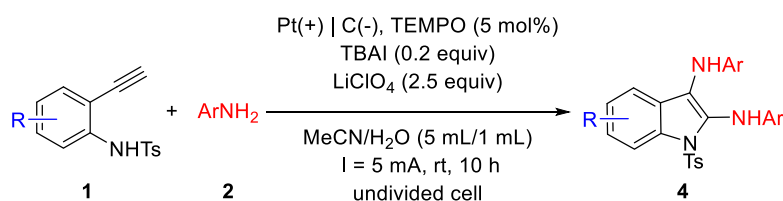
Red solid, 103.2 mg, 61% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 7.58 – 7.49 (m, 3H), 7.38 (d, *J* = 7.9 Hz, 4H), 7.34 (d, *J* = 6.3 Hz, 3H), 7.17 (d, *J* = 6.9 Hz, 1H), 7.14 (d, *J* = 8.3 Hz, 2H), 6.70 (d, *J* = 6.9 Hz, 4H), 5.05 (s, 2H), 1.36 (s, 18H).

¹³C NMR (150 MHz, CDCl₃) δ 149.3, 148.0, 147.0, 146.9, 146.3, 145.7, 144.0, 133.4, 130.9, 129.2, 128.9, 127.7, 126.2, 125.7, 125.4, 123.2, 119.8, 118.3, 117.1, 114.9, 60.0, 34.5, 34.4, 31.6, 31.5.

HRMS-ESI (*m/z*) [*M*+*K*]⁺ calculated for C₃₅H₃₇KN₃O₂S 602.2238, found 602.2233.

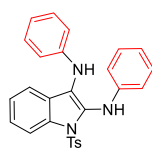
IV. Scope of indole synthesis.



Scheme S3. General procedure for synthesis of functionalized indoles

Procedure C. In an oven-dried undivided three-necked flask (10 mL) equipped with a stir bar, *o*-ethynylanilines¹ **1** (0.3 mmol), amine **2** (4.0 equiv), LiClO₄ (2.5 equiv), *n*-Bu₄NI (0.2 equiv), TEMPO (5 mol %), H₂O (1 mL) and CH₃CN (5 mL) were combined and added. The flask was equipped with platinum plates (1.0 cm × 1.0 cm × 0.2 mm) and the cathode electrode is graphite plates (1.0 cm × 1.0 cm × 0.2 cm). The reaction mixture was stirred and electrolyzed at a constant current of 5 mA under room temperature for 10 h. When the reaction was finished and the solvent was removed with a rotary evaporator. The pure product was obtained by flash chromatography on silica gel using petroleum ether and ethyl acetate as the eluent.

*N*²,*N*³-Diphenyl-1-tosyl-1*H*-indole-2,3-diamine (**4a**)



Following General Procedure C: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3

mmol), aniline (110 μ L, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), K₂CO₃ (2.0 equiv), H₂O (0.6 mL), CH₃CN (5.4 mL) were combined and added. The reaction mixture was stirred and electrolyzed at a constant current of 2 mA under room temperature for 12 h.

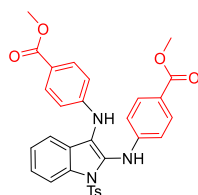
Red solid, 43.6 mg, 32% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.25 (d, *J* = 8.4 Hz, 1H), 7.53 (d, *J* = 8.3 Hz, 2H), 7.36 – 7.31 (m, 1H), 7.24 – 7.21 (m, 2H), 7.13 – 7.05 (m, 4H), 7.02 (d, *J* = 8.2 Hz, 2H), 6.84 (t, *J* = 7.3 Hz, 1H), 6.78 (t, *J* = 7.3 Hz, 1H), 6.53 (d, *J* = 7.5 Hz, 4H), 6.49 (s, 1H), 4.84 (s, 1H), 2.31 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 144.8, 144.4, 142.2, 134.9, 132.7, 129.6, 129.2, 129.0, 128.2, 126.9, 126.9, 124.1, 123.7, 120.5, 119.2, 119.1, 115.3, 115.02, 114.97, 114.0, 21.5.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₂₇H₂₃N₃NaO₂S 476.1403, found 476.1406.

Dimethyl 4,4'-((1-tosyl-1*H*-indole-2,3-diyl)bis(azanediyl))dibenzoate (4b)



Following General Procedure C: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), methyl 4-aminobenzoate (185.3 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 10 h.

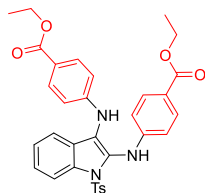
Yellow oil, 128.2 mg, 75% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 4/1).

¹H NMR (600 MHz, CDCl₃) δ 8.24 (d, *J* = 8.4 Hz, 1H), 7.75 (d, *J* = 8.8 Hz, 2H), 7.68 (d, *J* = 8.8 Hz, 2H), 7.48 (d, *J* = 8.4 Hz, 2H), 7.38 – 7.31 (m, 1H), 7.23 – 7.19 (m, 2H), 6.98 (d, *J* = 8.1 Hz, 2H), 6.77 (s, 1H), 6.48 (d, *J* = 8.8 Hz, 2H), 6.43 (d, *J* = 8.8 Hz, 2H), 5.37 (s, 1H), 3.83 (s, 3H), 3.82 (s, 3H), 2.25 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 167.1, 166.8, 148.1, 145.9, 145.4, 134.7, 132.7, 131.2, 131.2, 129.8, 127.5, 126.8, 126.1, 124.9, 124.0, 121.9, 120.5, 118.9, 115.1, 114.1, 113.6, 113.4, 51.8, 51.7, 21.5.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₃₁H₂₈N₃O₆S 570.1693, found 570.1683.

Diethyl 4,4'-((1-tosyl-1*H*-indole-2,3-diyl)bis(azanediyl))dibenzoate (4c)



Following General Procedure C: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), ethyl 4-aminobenzoate (202.3 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 10 h.

Yellow oil, 109.4 mg, 61% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 4/1).

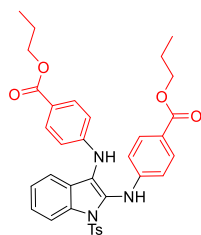
¹H NMR (600 MHz, CDCl₃) δ 8.24 (d, *J* = 8.4 Hz, 1H), 7.78 (d, *J* = 8.4 Hz, 2H), 7.71 (d, *J* = 8.3 Hz, 2H), 7.48 (d, *J* = 8.0 Hz, 2H), 7.36 (t, *J* = 7.5 Hz, 1H), 7.25 – 7.19 (m, 2H), 6.98 (d, *J* = 8.0 Hz,

2H), 6.73 (s, 1H), 6.49 (d, $J = 8.4$ Hz, 2H), 6.44 (d, $J = 8.3$ Hz, 2H), 5.26 (s, 1H), 4.34 – 4.27 (m, 4H), 2.26 (s, 3H), 1.37 – 1.33 (m, 6H).

^{13}C NMR (150 MHz, CDCl_3) δ 166.6, 166.3, 148.0, 145.8, 145.4, 134.7, 132.8, 131.2, 129.8, 127.5, 126.8, 126.0, 124.9, 124.0, 122.4, 121.0, 119.0, 115.1, 114.1, 113.6, 113.4, 60.6, 60.4, 21.5, 14.4, 14.4.

HRMS-ESI (m/z) $[\text{M}+\text{Na}]^+$ calculated for $\text{C}_{33}\text{H}_{31}\text{N}_3\text{NaO}_6\text{S}$ 620.1826, found 620.1815.

Dipropyl 4,4'-((1-tosyl-1*H*-indole-2,3-diyl)bis(azanediyl))dibenzoate (4d)



Following General Procedure C: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), propyl 4-aminobenzoate (220.1 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO_4 (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H_2O (1 mL), CH_3CN (5 mL) were combined and added. The reaction mixture under room temperature for 10 h.

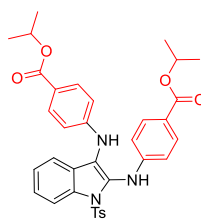
Yellow oil, 138.9 mg, 74% yield. ($R_f = 0.20$, petroleum ether/ethyl acetate = 4/1).

^1H NMR (600 MHz, CDCl_3) δ 8.24 (d, $J = 8.4$ Hz, 1H), 7.78 (d, $J = 8.7$ Hz, 2H), 7.71 (d, $J = 8.6$ Hz, 2H), 7.47 (d, $J = 8.3$ Hz, 2H), 7.38 – 7.32 (m, 1H), 7.25 – 7.20 (m, 2H), 6.98 (d, $J = 8.3$ Hz, 2H), 6.73 (s, 1H), 6.49 (d, $J = 8.7$ Hz, 2H), 6.44 (d, $J = 8.6$ Hz, 2H), 5.28 (s, 1H), 4.23 – 4.17 (m, 4H), 2.26 (s, 3H), 1.79 – 1.71 (m, 4H), 1.02 – 0.98 (m, 6H).

^{13}C NMR (150 MHz, CDCl_3) δ 166.7, 166.4, 148.0, 145.9, 145.3, 134.7, 132.8, 131.2, 129.8, 127.5, 126.8, 126.1, 124.9, 124.0, 122.3, 121.0, 119.0, 115.1, 114.1, 113.6, 113.5, 66.2, 66.0, 22.2, 22.2, 21.5, 10.6, 10.5.

HRMS-ESI (m/z) $[\text{M}+\text{K}]^+$ calculated for $\text{C}_{35}\text{H}_{35}\text{KN}_3\text{O}_6\text{S}$ 664.1878, found 664.1871.

Diisopropyl 4,4'-((1-tosyl-1*H*-indole-2,3-diyl)bis(azanediyl))dibenzoate (4e)



Following General Procedure C: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), isopropyl 4-aminobenzoate (219.4 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO_4 (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H_2O (1 mL), CH_3CN (5 mL) were combined and added. The reaction mixture under room temperature for 10 h.

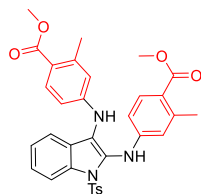
Yellow oil, 137.1 mg, 73% yield. ($R_f = 0.20$, petroleum ether/ethyl acetate = 4/1).

^1H NMR (600 MHz, CDCl_3) δ 8.24 (d, $J = 8.4$ Hz, 1H), 7.77 (d, $J = 8.8$ Hz, 2H), 7.69 (d, $J = 8.7$ Hz, 2H), 7.47 (d, $J = 8.4$ Hz, 2H), 7.37 – 7.31 (m, 1H), 7.24 – 7.17 (m, 2H), 6.97 (d, $J = 8.2$ Hz, 2H), 6.73 (s, 1H), 6.49 (d, $J = 8.8$ Hz, 2H), 6.43 (d, $J = 8.7$ Hz, 2H), 5.35 (s, 1H), 5.25 – 5.11 (m, 2H), 2.25 (s, 3H), 1.36 – 1.29 (m, 12H).

¹³C NMR (150 MHz, CDCl₃) δ 166.1, 165.8, 148.0, 145.9, 145.3, 134.7, 132.8, 131.2, 129.8, 127.5, 126.8, 126.0, 124.9, 124.0, 122.7, 121.3, 119.1, 115.1, 114.0, 113.7, 113.6, 67.9, 67.7, 22.0, 22.0, 21.5.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₃₅H₃₅N₃NaO₆S 648.2139, found 648.2136.

Dimethyl 4,4'-((1-tosyl-1*H*-indole-2,3-diyl)bis(azanediyl))bis(2-methylbenzoate) (4f)



Following General Procedure C: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), methyl 4-amino-2-methylbenzoate (202.3 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 10 h.

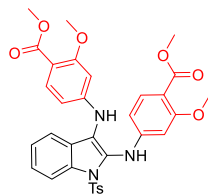
Yellow oil, 127.4 mg, 71% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 4/1).

¹H NMR (600 MHz, CDCl₃) δ 8.24 (d, *J* = 8.4 Hz, 1H), 7.74 (d, *J* = 9.2 Hz, 1H), 7.68 (d, *J* = 8.5 Hz, 1H), 7.46 (d, *J* = 8.4 Hz, 2H), 7.39 – 7.31 (m, 1H), 7.25 – 7.17 (m, 2H), 6.98 (d, *J* = 8.3 Hz, 2H), 6.63 (s, 1H), 6.38 – 6.30 (m, 3H), 6.19 – 6.07 (m, 1H), 5.19 (s, 1H), 3.81 (s, 3H), 3.80 (s, 3H), 2.44 (s, 3H), 2.26 (s, 3H), 2.23 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 167.6, 167.4, 147.4, 145.2, 144.9, 142.8, 142.7, 134.7, 132.8, 132.8, 132.7, 129.7, 127.6, 126.8, 126.3, 124.8, 124.1, 121.1, 119.6, 119.1, 117.3, 117.0, 115.2, 113.5, 111.9, 111.2, 51.5, 51.4, 22.3, 21.8, 21.5.

HRMS-ESI (m/z) [M+K]⁺ calculated for C₃₃H₃₁KN₃O₆S 636.1565, found 636.1566.

Dimethyl 4,4'-((1-tosyl-1*H*-indole-2,3-diyl)bis(azanediyl))bis(2-methoxybenzoate) (4g)



Following General Procedure C: *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), methyl 4-amino-2-methoxybenzoate (221.9 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 10 h.

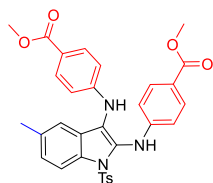
Yellow oil, 103.8 mg, 55% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 1/1).

¹H NMR (600 MHz, CDCl₃) δ 8.22 (d, *J* = 8.4 Hz, 1H), 7.63 (d, *J* = 8.5 Hz, 1H), 7.57 (d, *J* = 8.5 Hz, 1H), 7.43 (d, *J* = 8.2 Hz, 2H), 7.33 (t, *J* = 7.6 Hz, 1H), 7.25 – 7.19 (m, 2H), 6.96 (d, *J* = 8.1 Hz, 2H), 6.71 (s, 1H), 6.15 – 6.09 (m, 2H), 6.05 (d, *J* = 8.5 Hz, 1H), 5.88 (s, 1H), 5.73 (s, 1H), 3.77 (s, 3H), 3.75 (s, 3H), 3.57 (s, 3H), 3.36 (s, 3H), 2.23 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 166.1, 166.1, 161.4, 161.3, 149.4, 147.3, 145.4, 134.6, 133.8, 133.7, 132.9, 129.7, 127.0, 126.8, 125.8, 125.0, 124.0, 119.5, 115.2, 114.3, 111.1, 109.8, 106.4, 106.1, 98.0, 97.7, 55.6, 55.5, 51.6, 51.5, 21.5.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₃₃H₃₁N₃NaO₈S 652.1724, found 652.1712.

Dimethyl 4,4'-((5-methyl-1-tosyl-1*H*-indole-2,3-diyl)bis(azanediyl))dibenzoate (4h)



Following General Procedure C: *N*-(2-Ethynyl-4-methylphenyl)-4-methylbenzenesulfonamide (87.4 mg, 0.3 mmol), methyl 4-aminobenzoate (185.3 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 10 h.

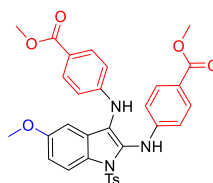
Yellow oil, 82.3 mg, 47% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 4/1).

¹H NMR (600 MHz, CDCl₃) δ 8.11 (d, *J* = 8.5 Hz, 1H), 7.77 (d, *J* = 8.8 Hz, 2H), 7.69 (d, *J* = 8.7 Hz, 2H), 7.46 (d, *J* = 8.4 Hz, 2H), 7.17 (dd, *J* = 8.5, 1.2 Hz, 1H), 7.00 (s, 1H), 6.98 (d, *J* = 8.2 Hz, 2H), 6.73 (s, 1H), 6.47 (d, *J* = 8.8 Hz, 2H), 6.42 (d, *J* = 8.7 Hz, 2H), 5.23 (s, 1H), 3.84 (s, 3H), 3.84 (s, 3H), 2.36 (s, 3H), 2.26 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 167.1, 166.8, 148.1, 145.9, 145.3, 134.7, 134.0, 131.3, 131.2, 130.9, 129.7, 127.6, 126.8, 126.3, 126.2, 121.9, 120.5, 118.7, 114.9, 114.2, 113.5, 113.1, 51.8, 51.7, 21.5, 21.4.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₃₂H₂₉N₃NaO₆S 606.1669, found 606.1662.

Dimethyl 4,4'-((5-methoxy-1-tosyl-1*H*-indole-2,3-diyl)bis(azanediyl))dibenzoate (4i)



Following General Procedure C: *N*-(2-Ethynyl-4-methoxyphenyl)-4-methylbenzenesulfonamide (92.3 mg, 0.3 mmol), methyl 4-aminobenzoate (185.3 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture was stirred and electrolyzed at a constant current of 1.2 mA under room temperature for 48 h.

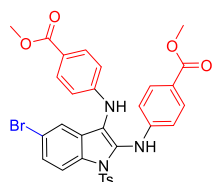
Yellow oil, 91.8 mg, 51% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 4/1).

¹H NMR (600 MHz, CDCl₃) δ 8.11 (d, *J* = 9.1 Hz, 1H), 7.75 (d, *J* = 8.8 Hz, 2H), 7.68 (d, *J* = 8.7 Hz, 2H), 7.44 (d, *J* = 8.4 Hz, 2H), 6.98 (d, *J* = 8.1 Hz, 2H), 6.94 (dd, *J* = 9.1, 2.6 Hz, 1H), 6.77 (s, 1H), 6.66 – 6.59 (m, 1H), 6.44 (d, *J* = 8.8 Hz, 2H), 6.42 (d, *J* = 8.8 Hz, 2H), 5.25 (s, 1H), 3.84 (s, 3H), 3.83 (s, 3H), 3.72 (s, 3H), 2.26 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 167.1, 166.8, 156.9, 148.0, 145.7, 145.3, 134.5, 131.24, 131.17, 129.7, 128.4, 127.5, 127.0, 126.8, 122.0, 120.5, 116.3, 114.3, 113.5, 113.2, 113.0, 101.6, 55.6, 51.8, 51.7, 21.5.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₃₂H₃₀N₃O₇S 600.1799, found 600.1794.

Dimethyl 4,4'-((5-bromo-1-tosyl-1H-indole-2,3-diyl)bis(azanediyl))dibenzoate (4j)



Following General Procedure C: *N*-(4-Bromo-2-ethynylphenyl)-4-methylbenzenesulfonamide (107.2 mg, 0.3 mmol), methyl 4-aminobenzoate (185.3 mg, 1.2 mmol), TBAI (23 mg, 0.06 mmol), LiClO₄ (83 mg, 0.75 mmol), TEMPO (2.2 mg, 5 mol %), H₂O (1 mL), CH₃CN (5 mL) were combined and added. The reaction mixture under room temperature for 10 h.

Yellow oil, 79.8 mg, 41% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 4/1).

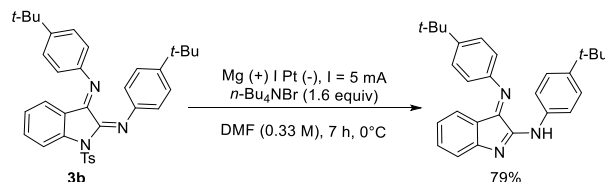
¹H NMR (600 MHz, CDCl₃) δ 8.10 (d, *J* = 8.8 Hz, 1H), 7.76 (d, *J* = 8.8 Hz, 2H), 7.70 (d, *J* = 8.7 Hz, 2H), 7.47 (d, *J* = 8.4 Hz, 2H), 7.44 (dd, *J* = 8.9, 2.0 Hz, 1H), 7.36 – 7.31 (m, 1H), 7.02 (d, *J* = 8.1 Hz, 2H), 6.83 (s, 1H), 6.44 – 6.42 (m, 2H), 6.42 – 6.38 (m, 2H), 5.17 (s, 1H), 3.85 (s, 3H), 3.84 (s, 3H), 2.28 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 167.0, 166.7, 147.7, 145.8, 145.1, 134.3, 131.3, 131.24, 131.19, 129.9, 129.1, 128.2, 127.6, 126.8, 122.5, 121.2, 120.9, 117.7, 116.5, 114.6, 113.4, 111.5, 51.9, 51.7, 21.6.

HRMS-ESI (m/z) [M+Na]⁺ calculated for C₃₁H₂₆BrN₃NaO₆S 670.0618, found 670.0604.

V. Derivatization of compound 3b

(*E*)-*N*-(4-(*Tert*-butyl)phenyl)-3-((4-(*tert*-butyl)phenyl)imino)-3*H*-indol-2-amine³ (5)



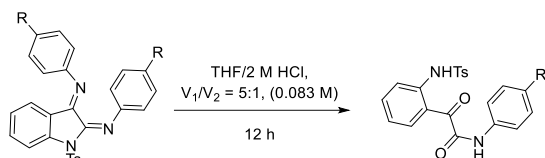
3b (0.2 mmol) was added to the solution of *n*-Et₄NBr (104 mg, 1.6 equiv) in anhydrous DMF (6 mL) placed in an undivided cell equipped with a Pt plate cathode (1.0 cm × 1.0 cm × 0.2 cm) and an Mg plate anode (1.0 cm × 1.0 cm × 0.2 cm). A constant current electrolysis was carried out at 0 °C under N₂ atmosphere for 7 h. TLC indicated complete conversion of the starting material. The reaction mixture was poured into aqueous NaHCO₃ (20 mL) and then extracted with ether (10 mL × 3). The combined organic layer was washed with H₂O (10 mL × 2) and dried over MgSO₄. Evaporation of the solvent gave a crude product. The pure product was obtained by flash chromatography on silica gel using petroleum ether and ethyl acetate as the eluent.

Red solid, 64.6 mg, 79% yield. (*R*_f = 0.42, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 7.89 (s, 1H), 7.77 (d, *J* = 8.7 Hz, 2H), 7.46 (d, *J* = 8.5 Hz, 2H), 7.42 (d, *J* = 8.7 Hz, 2H), 7.25 – 7.21 (m, 1H), 7.19 (d, *J* = 7.6 Hz, 1H), 7.03 (d, *J* = 8.5 Hz, 2H), 6.80 (d, *J* = 7.0 Hz, 1H), 6.66 (t, *J* = 7.0 Hz, 1H), 1.39 (s, 9H), 1.33 (s, 9H).

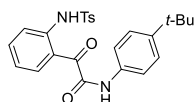
¹³C NMR (150 MHz, CDCl₃) δ 163.0, 159.8, 158.3, 149.0, 146.5, 146.0, 135.9, 134.4, 126.2, 126.1, 125.5, 122.5, 119.8, 119.12, 119.08, 119.0, 34.6, 34.4, 31.5, 31.5.

HRMS-ESI (m/z) [M+K]⁺ calculated for C₂₈H₃₁KN₃ 448.2150, found 448.2148.



Procedure D. 2 M HCl (0.2 mL) was added to a solution of **3** (0.1 mmol) in THF (1 mL) at room temperature. After 12 hours, the reaction mixture was concentrated under reduced pressure and purified by column chromatography (eluent: petroleum ether/ethyl acetate = 4/1) to afford product.

***N*-(4-(*Tert*-butyl)phenyl)-2-(2-((4-methylphenyl)sulfonamido)phenyl)-2-oxoacetamide (**6**)**



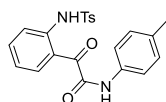
Following General Procedure D: Yellow oil, 32.8 mg, 73% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 4/1).

¹H NMR (600 MHz, CDCl₃) δ 10.44 (s, 1H), 8.82 (s, 1H), 8.45 (d, *J* = 8.0 Hz, 1H), 7.71 (d, *J* = 8.3 Hz, 1H), 7.68 (d, *J* = 8.2 Hz, 2H), 7.59 (d, *J* = 8.6 Hz, 2H), 7.55 (t, *J* = 7.8 Hz, 1H), 7.41 (d, *J* = 8.6 Hz, 2H), 7.20 (d, *J* = 8.1 Hz, 2H), 7.13 (t, *J* = 7.7 Hz, 1H), 2.31 (s, 3H), 1.33 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 190.3, 159.0, 148.7, 144.4, 140.9, 136.2, 135.9, 134.8, 133.9, 129.9, 127.3, 126.1, 123.3, 120.6, 120.4, 119.8, 34.6, 31.3, 21.5.

HRMS-ESI (m/z) [M+K]⁺ calculated for C₂₅H₂₆KN₂O₄S 489.1245, found 489.1244.

2-(2-((4-Methylphenyl)sulfonamido)phenyl)-2-oxo-*N*-(*p*-tolyl)acetamide (7**)**



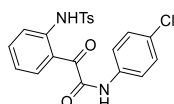
Following General Procedure D: Yellow oil, 23.7 mg, 58% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 4/1).

¹H NMR (600 MHz, CDCl₃) δ 10.40 (s, 1H), 8.75 (s, 1H), 8.47 (dd, *J* = 8.1, 1.4 Hz, 1H), 7.71 (d, *J* = 8.3 Hz, 1H), 7.67 (d, *J* = 8.3 Hz, 2H), 7.54 (d, *J* = 8.4 Hz, 3H), 7.22 – 7.17 (m, 4H), 7.14 (t, *J* = 7.7 Hz, 1H), 2.35 (s, 3H), 2.31 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 190.2, 158.8, 144.4, 140.9, 136.3, 135.9, 135.4, 134.8, 133.9, 129.8, 129.8, 127.3, 123.3, 120.6, 120.5, 120.0, 21.5, 21.0.

HRMS-ESI (m/z) [M+H]⁺ calculated for C₂₂H₂₁N₂O₄S 409.1217, found 409.1214.

***N*-(4-Chlorophenyl)-2-(2-((4-methylphenyl)sulfonamido)phenyl)-2-oxoacetamide (**8**)**



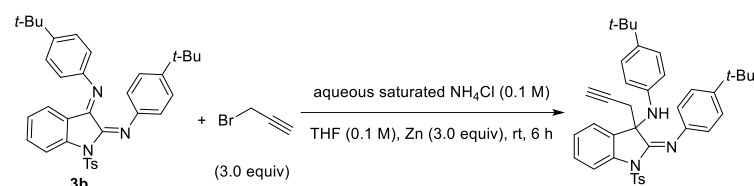
Following General Procedure D: Yellow oil, 36.5 mg, 85% yield. (*R*_f = 0.20, petroleum ether/ethyl acetate = 4/1).

¹H NMR (600 MHz, CDCl₃) δ 10.49 (s, 1H), 9.07 (s, 1H), 8.43 (dd, *J* = 8.0, 1.3 Hz, 1H), 7.71 (t, *J* = 9.0 Hz, 3H), 7.67 (d, *J* = 8.8 Hz, 2H), 7.57 (t, *J* = 7.2 Hz, 1H), 7.36 (d, *J* = 8.8 Hz, 2H), 7.22 (d, *J* = 8.2 Hz, 2H), 7.16 (t, *J* = 7.4 Hz, 1H), 2.33 (s, 3H).

¹³C NMR (150 MHz, CDCl₃) δ 190.0, 159.2, 144.6, 140.9, 136.4, 135.7, 135.2, 134.8, 130.6, 129.9, 129.3, 127.3, 123.3, 121.3, 120.4, 116.3, 21.5.

HRMS-ESI (m/z) [M+K]⁺ calculated for C₂₁H₁₇ClKN₂O₄S 467.0229, found 467.0235.

(*E*)-*N*-(4-(*Tert*-butyl)phenyl)-2-((4-(*tert*-butyl)phenyl)imino)-3-(prop-2-yn-1-yl)-1-tosylindolin-3-amine (9)



To a stirred solution of **3b** (0.1 mmol) in THF (1 mL), saturated aqueous NH₄Cl (1 mL) and 3-Bromopropyne (0.3 mmol) were added successively. To the resulting solution, Zn powder (0.3 mmol) was added portion wise at room temperature. This mixture was stirred for 6 hours. Then, the reaction mixture was transferred to the separatory funnel. Two liquid layers were separated, and the aqueous layer was extracted with 1:1 solution of EtOAc: THF (3 × 5 mL). Combined organic layers were washed with brine (10 mL) and dried over anhydrous Na₂SO₄. Evaporation of the solvent gave a crude product. The pure product was obtained by flash chromatography on silica gel using petroleum ether and ethyl acetate as the eluent.

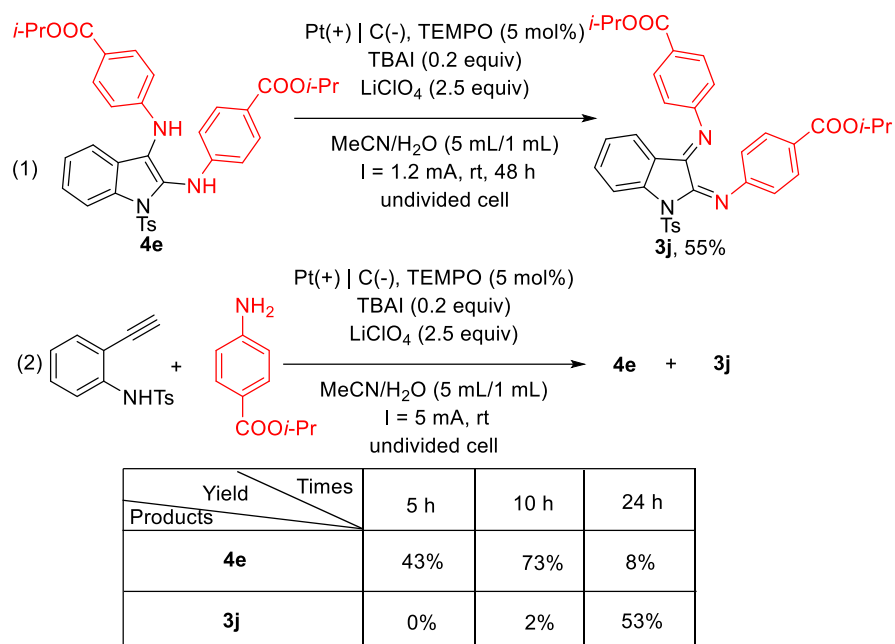
Pink solid, 36.8 mg, 61% yield. (*R*_f = 0.2, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 8.19 (d, *J* = 8.3 Hz, 1H), 8.02 (d, *J* = 8.3 Hz, 2H), 7.41 (t, *J* = 7.3 Hz, 1H), 7.29 (d, *J* = 8.1 Hz, 2H), 7.23 (dd, *J* = 7.5, 1.1 Hz, 1H), 7.08 (d, *J* = 8.6 Hz, 3H), 6.78 (d, *J* = 8.7 Hz, 2H), 6.16 (d, *J* = 8.5 Hz, 2H), 5.68 (d, *J* = 8.7 Hz, 2H), 3.48 (s, 1H), 2.86 (dd, *J* = 16.2, 2.6 Hz, 1H), 2.66 (dd, *J* = 16.2, 2.6 Hz, 1H), 2.45 (s, 3H), 1.88 (t, *J* = 2.6 Hz, 1H), 1.30 (s, 9H), 1.19 (s, 9H).

¹³C NMR (150 MHz, CDCl₃) δ 154.7, 145.7, 144.7, 142.8, 141.4, 141.3, 140.2, 135.7, 130.1, 129.5, 129.0, 125.4, 124.9, 124.4, 123.7, 118.1, 114.1, 114.0, 76.7, 72.7, 64.1, 34.2, 33.8, 33.5, 31.51, 31.45, 21.8.

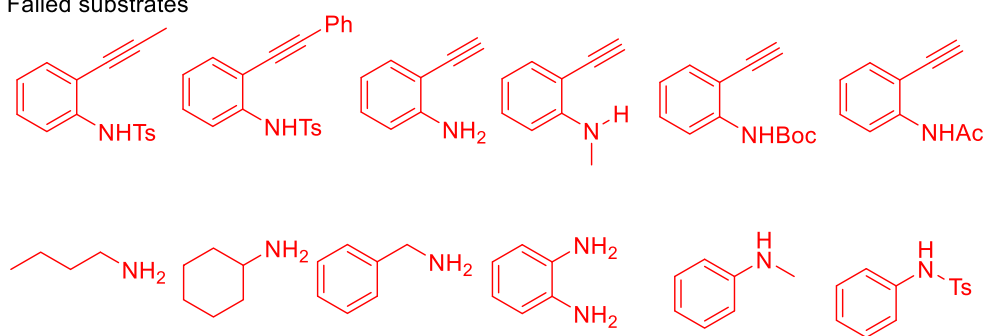
HRMS-ESI (m/z) [M+K]⁺ calculated for C₃₈H₄₁KN₃O₂S 642.2551, found 642.2549.

Controlled synthesis of **4e** and **3j**



Scheme S4. Controlled synthesis of **4e** and **3j**

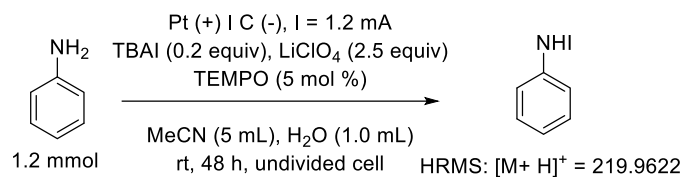
Failed substrates



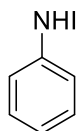
Scheme S5. Failed substrates

VI. Mechanistic studies

Byproduct-Trapping Study:



In an oven-dried undivided three-necked flask (10 mL) equipped with a stir bar, aniline (1.2 mmol, 4.0 equiv), LiClO₄ (2.5 equiv), *n*-Bu₄NI (0.2 equiv), TEMPO (5 mol %), H₂O (1 mL) and CH₃CN (5 mL) were combined and added. The flask was equipped with platinum plates (1.0 cm × 1.0 cm × 0.2 mm) and the cathode electrode is graphite plates (1.0 cm × 1.0 cm × 0.2 cm). The reaction mixture was stirred and electrolyzed at a constant current of 1.2 mA under room temperature for 48 h. The resulting mixture was analyzed by HRMS, and key possible intermediates could be detected.



HRMS-ESI (m/z) [M+H]⁺ calculated for C₆H₇IN 219.9618, found 219.9622.

0916-MXT15 #45 RT: 0.35 AV: 1 NL: 9.12E4
T: FTMS + p ESI Full ms [100.00-800.00]

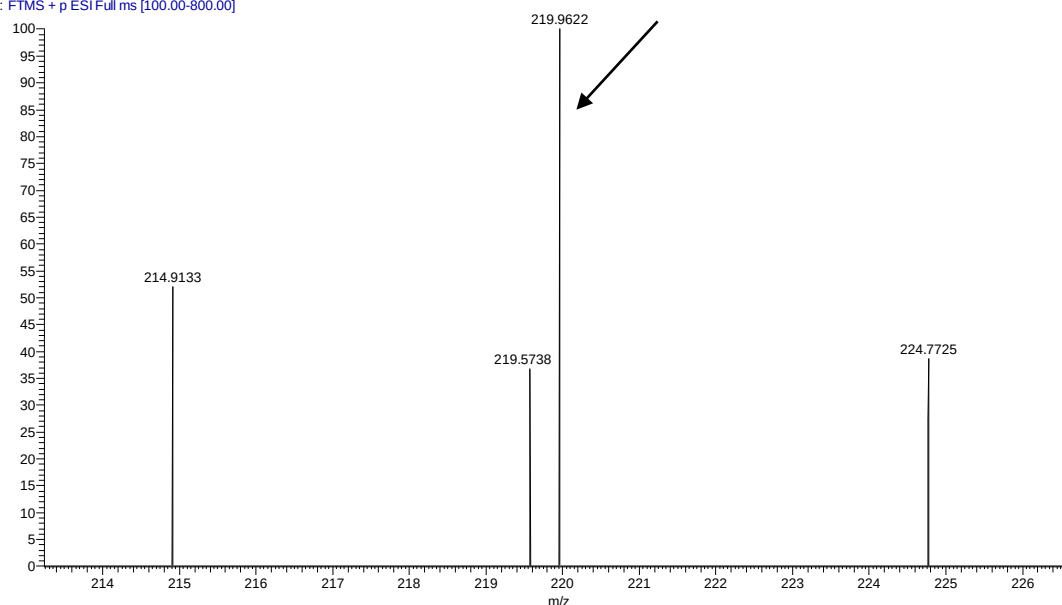
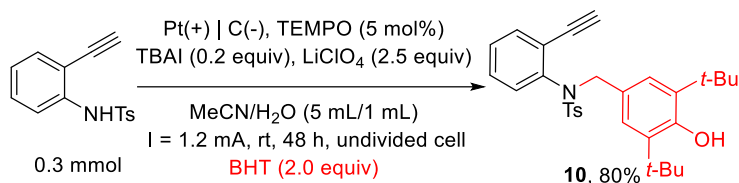
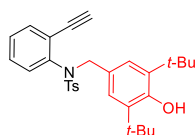


Figure S1. The HRMS spectra of compound **IV**.



In an oven-dried undivided three-necked flask (10 mL) equipped with a stir bar, *N*-(2-Ethynylphenyl)-4-methylbenzenesulfonamide (83 mg, 0.3 mmol), LiClO₄ (2.5 equiv), *n*-Bu₄NI (0.2 equiv), TEMPO (5 mol %), BHT (2.0 equiv), H₂O (1 mL) and CH₃CN (5 mL) were combined and added. The flask was equipped with platinum plates (1.0 cm × 1.0 cm × 0.2 mm) and the cathode electrode is graphite plates (1.0 cm × 1.0 cm × 0.2 cm). The reaction mixture was stirred and electrolyzed at a constant current of 1.2 mA under room temperature for 48 h. When the reaction was finished and the solvent was removed with a rotary evaporator. The pure product was obtained by flash chromatography on silica gel using petroleum ether and ethyl acetate as the eluent.

N-(3,5-di-*tert*-butyl-4-hydroxybenzyl)-*N*-(2-ethynylphenyl)-4-methylbenzenesulfonamide (**10**)



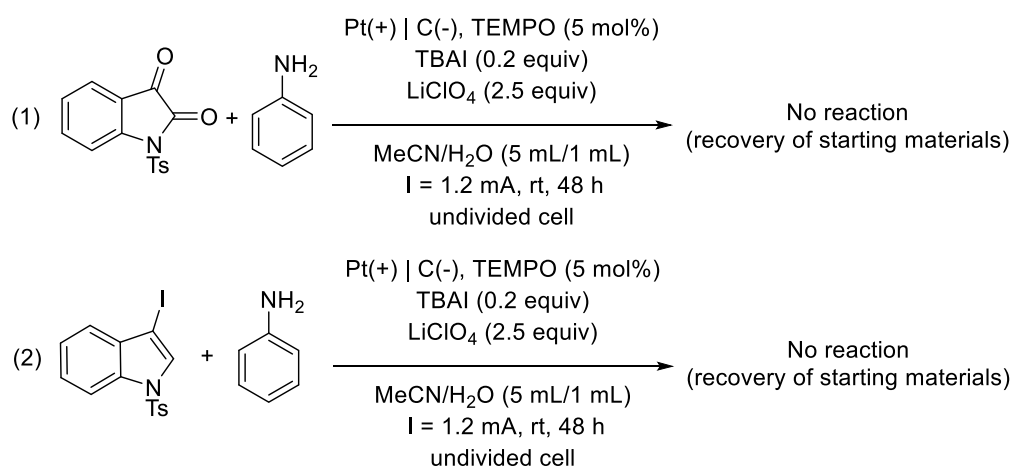
White solid, 117.5 mg, 80% yield. (*R*_f = 0.25, petroleum ether/ethyl acetate = 20/1).

¹H NMR (600 MHz, CDCl₃) δ 7.67 (d, *J* = 8.2 Hz, 2H), 7.45 – 7.37 (m, 1H), 7.26 (d, *J* = 8.2 Hz, 2H), 7.21 – 7.16 (m, 2H), 6.95 – 6.90 (m, 1H), 6.89 (s, 2H), 5.12 (s, 1H), 4.78 (s, 2H), 2.99 (s, 1H), 2.43 (s, 3H), 1.33 (s, 18H).

¹³C NMR (150 MHz, CDCl₃) δ 153.3, 143.2, 140.6, 137.4, 135.5, 134.0, 131.8, 129.5, 129.0, 127.91, 127.85, 126.2, 126.1, 123.7, 81.8, 80.5, 54.7, 34.2, 30.2, 21.6.

HRMS-ESI (m/z) [M+K]⁺ calculated for C₃₀H₃₅KNO₃S 528.1969, found 528.1970.

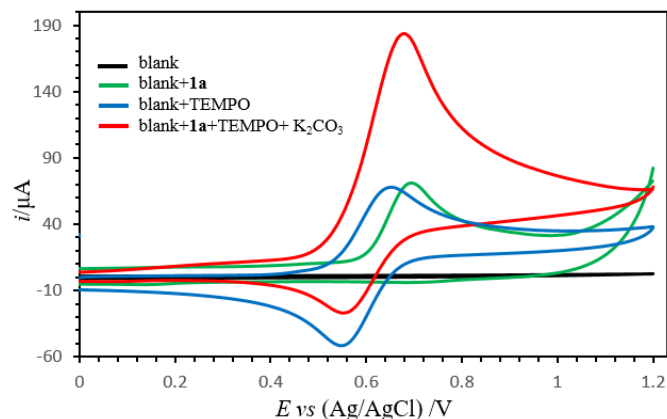
Control experiments



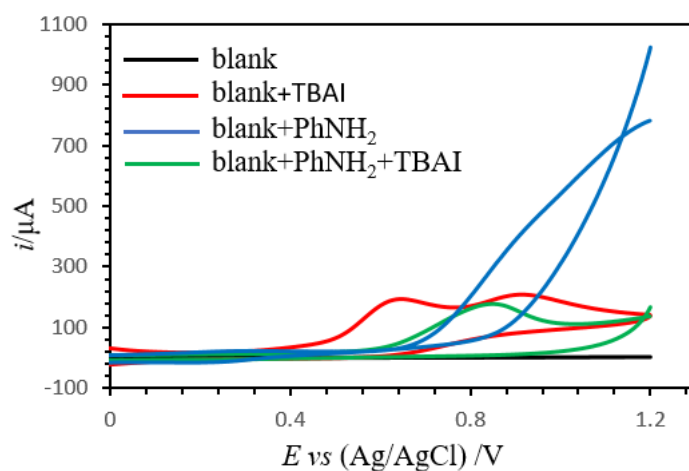
Scheme S6. Control experiments

Cyclic Voltammetry Studies.

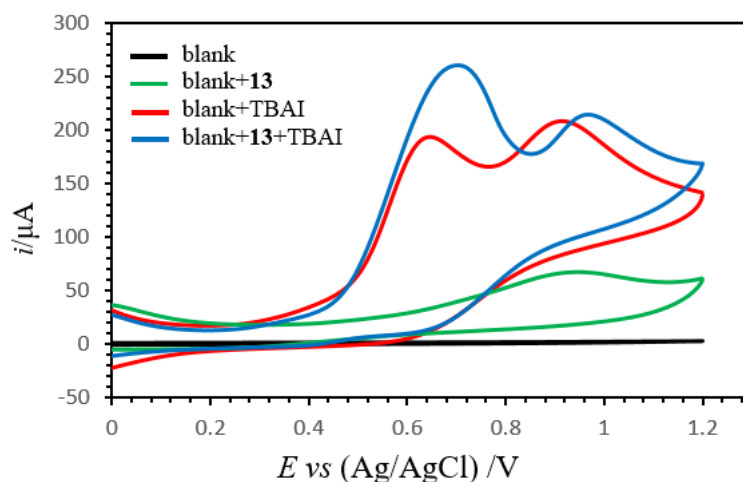
Cyclic voltammetry was obtained using a glassy carbon working electrode, a platinum counter electrode. The reference was an Ag/AgCl electrode submerged in saturated aqueous KCl solution, and separated from reaction by a salt bridge. The scan rate was 0.10 V/s, ranging from 0.0 V to 1.2 V.



Supplementary Figure S2. (a): (black line) blank; LiCO₄ (0.75 mmol), 1 mL H₂O and 5 mL CH₃CN were poured into the electrochemical cell in cyclic voltammetry experiments. (b): (green line) blank + **1a** (0.3 mmol). (c): (blue line) blank + TEMPO (0.015 mmol). (d): (red line) blank + **1a** (0.3 mmol) + TEMPO (0.015 mmol) + K₂CO₃ (0.6 mmol).



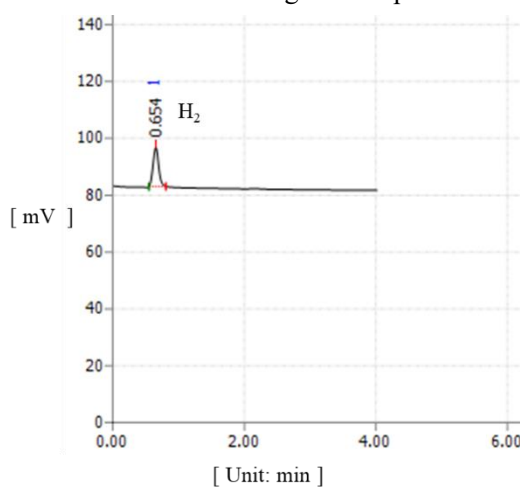
Supplementary Figure S3. (a): (black line) blank; LiCO₄ (0.75 mmol), 1 mL H₂O and 5 mL CH₃CN were poured into the electrochemical cell in cyclic voltammetry experiments. (b): (red line) blank + TBAI (0.06 mmol). (c): (blue line) blank + aniline (1.2 mmol). (d): (green line) blank + TBAI (0.06 mmol) + aniline (1.2 mmol).



Supplementary Figure S4. (a): (black line) blank; LiCO_4 (0.75 mmol), 1 mL H_2O and 5 mL CH_3CN were poured into the electrochemical cell in cyclic voltammetry experiments. (b): (green line) blank + **13** (0.3 mmol). (c): (red line) blank + TBAI (0.015 mmol). (d): (blue line) blank + **13** (0.3 mmol) + TBAI (0.015 mmol).

GC spectrum of the gaseous part in the reaction bottle

After the electrolysis reaction of **1a** and **2a** was finished, the headspace atmosphere was analyzed with GC. The formation of H_2 was confirmed through this experiment.

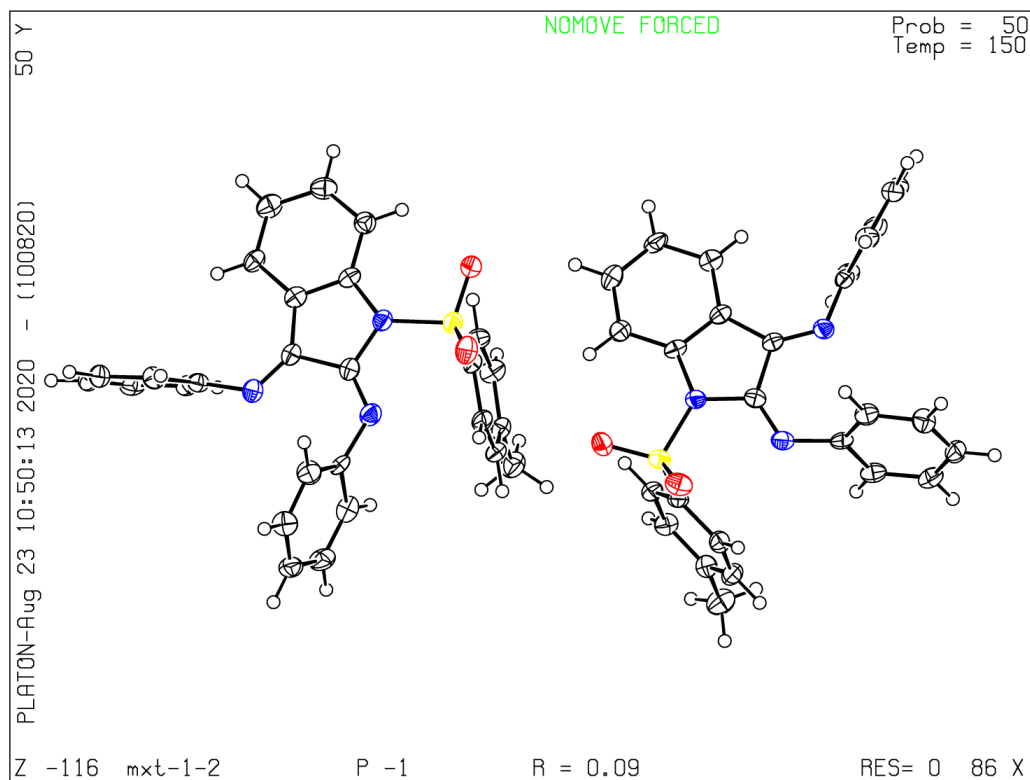


Supplementary Figure S5. GC trace of H_2 from the gas in headspace of reactor.

VII. References

1. M. He, N. Chen, T. Zhou, Q. Li, H. Li, M. Lang, J. Wang and S. Peng. *Org. Lett.*, 2019, **23**, 9559–9563.
2. Z.-Q. Zhang, Y.-H. Xu, J.-C. Dai, Y. Li, J. Sheng and X.-S Wang. *Org. Lett.*, 2021, **6**, 2194–2198.
3. H. Senboku, K. Nakahara, T. Fukuhara, S. Hara. *Tetrahedron Lett.*, 2010, **51**, 435-438.

VIII. X-ray crystal Data



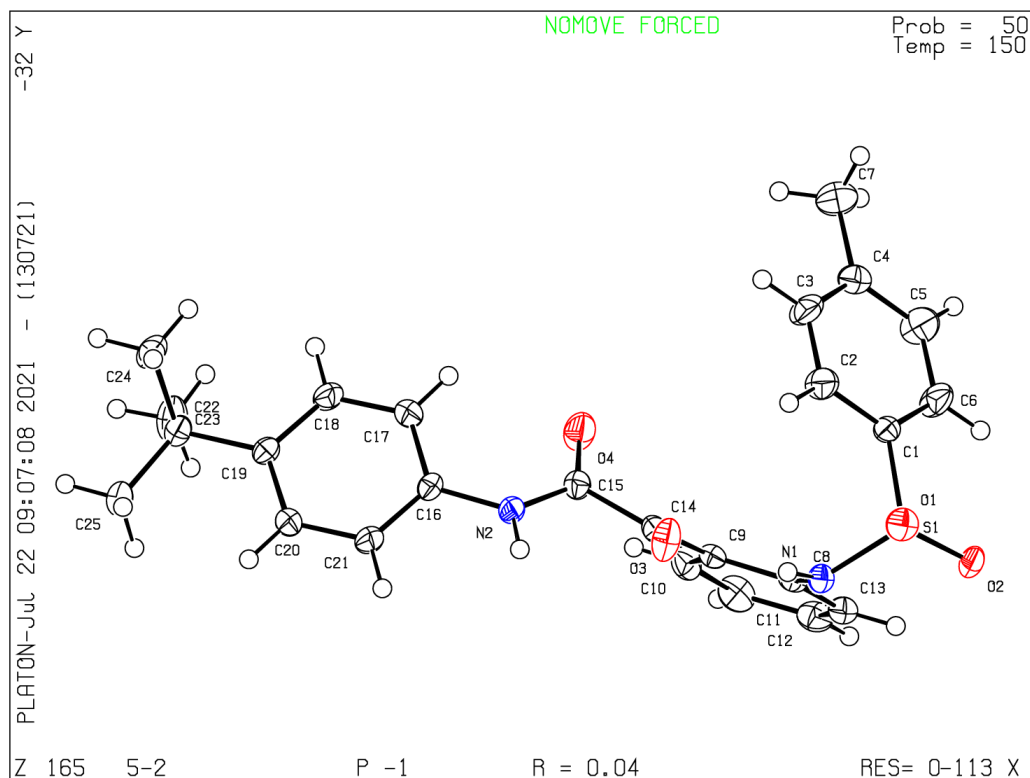
CCDC 2090357

X-ray structure and CCDC number of compound 3a.

Crystal data and structure refinement for 3a.

Identification code	3a
Empirical formula	C ₂₇ H ₂₁ N ₃ O ₂ S
Formula weight	451.53
Temperature/K	150.00(10)
Crystal system	triclinic
Space group	P-1
a/Å	9.8859(10)
b/Å	9.9067(9)
c/Å	23.766(2)
α /°	89.459(7)
β /°	81.983(8)
γ /°	73.840(8)
Volume/Å ³	2212.7(4)
Z	4
$\rho_{\text{calc}}/\text{cm}^3$	1.355

μ/mm^{-1}	0.177
F(000)	944.0
Crystal size/ mm^3	$0.14 \times 0.12 \times 0.11$
Radiation	Mo K α ($\lambda = 0.71073$)
2 Θ range for data collection/ $^\circ$	4.282 to 49.994
Index ranges	$-11 \leq h \leq 11, -11 \leq k \leq 10, -28 \leq l \leq 27$
Reflections collected	15379
Independent reflections	7780 [$R_{\text{int}} = 0.0448, R_{\text{sigma}} = 0.0812$]
Data/restraints/parameters	7780/0/597
Goodness-of-fit on F^2	1.068
Final R indexes [$I \geq 2\sigma(I)$]	$R_1 = 0.0888, wR_2 = 0.1899$
Final R indexes [all data]	$R_1 = 0.1101, wR_2 = 0.2028$
Largest diff. peak/hole / $\text{e } \text{\AA}^{-3}$	0.45/-0.44



CCDC 2123173

X-ray structure and CCDC number of compound 6.

Crystal data and structure refinement for 6.

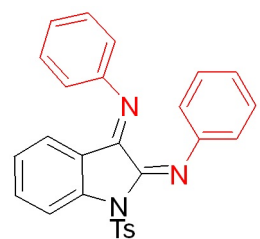
Identification code	6
Empirical formula	C ₂₅ H ₂₆ N ₂ O ₄ S
Formula weight	450.54
Temperature/K	149.99(10)
Crystal system	triclinic
Space group	P-1
a/Å	8.5563(10)
b/Å	11.6457(13)
c/Å	12.4602(16)
α/°	75.907(11)
β/°	77.454(11)
γ/°	74.029(10)
Volume/Å ³	1142.6(3)
Z	2
ρ _{calc} /cm ³	1.310
μ/mm ⁻¹	0.176
F(000)	476.0

Crystal size/mm ³	0.14 × 0.13 × 0.12
Radiation	Mo Kα (λ = 0.71073)
2θ range for data collection/°	4.524 to 49.986
Index ranges	-10 ≤ h ≤ 10, -12 ≤ k ≤ 13, -14 ≤ l ≤ 14
Reflections collected	6853
Independent reflections	4004 [R _{int} = 0.0274, R _{sigma} = 0.0481]
Data/restraints/parameters	4004/1/301
Goodness-of-fit on F ²	1.027
Final R indexes [I ≥ 2σ (I)]	R ₁ = 0.0426, wR ₂ = 0.1018
Final R indexes [all data]	R ₁ = 0.0535, wR ₂ = 0.1089
Largest diff. peak/hole / e Å ⁻³	0.36/-0.47

IX. NMR Spectra

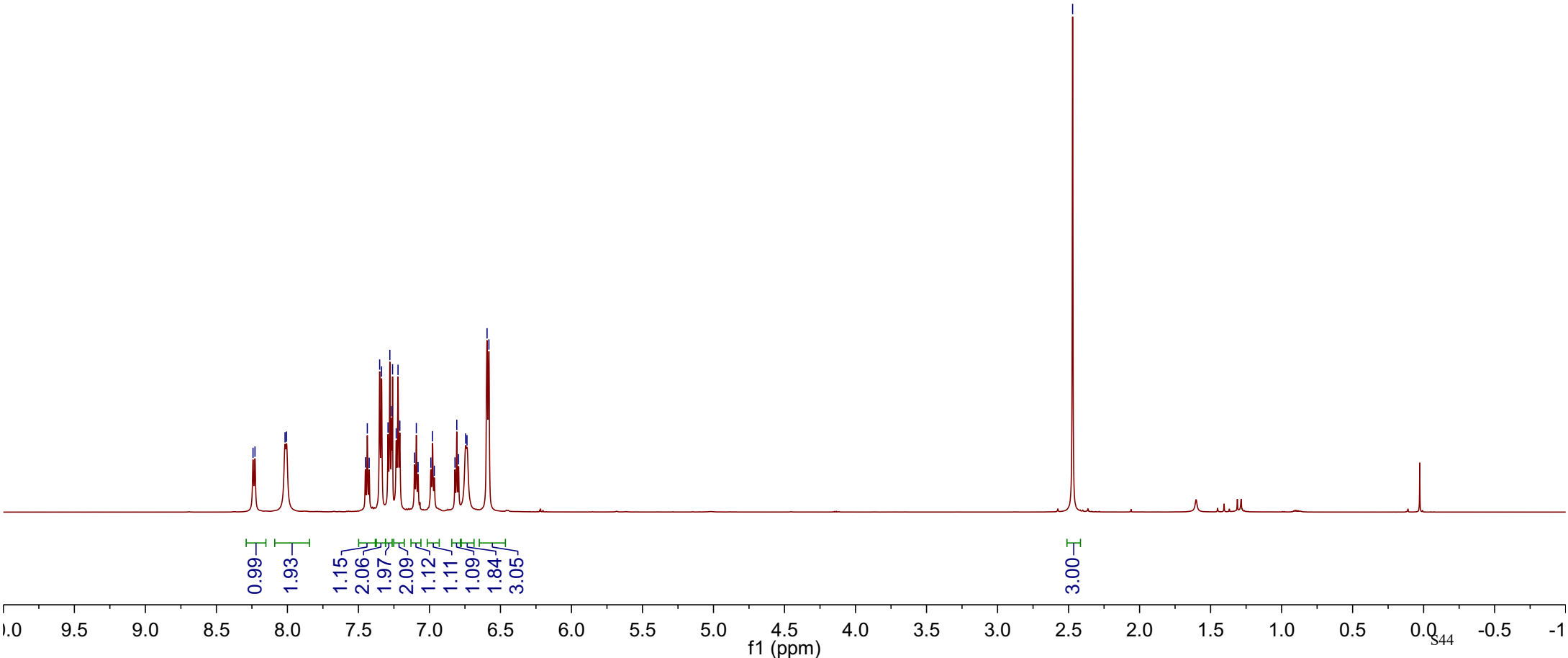
8.24
8.23
8.02
8.01
7.45
7.44
7.42
7.35
7.34
7.29
7.28
7.27
7.26
7.23
7.22
7.21
7.10
7.09
7.08
6.99
6.98
6.97
6.82
6.81
6.79
6.74
6.74
6.59
6.58

—2.47



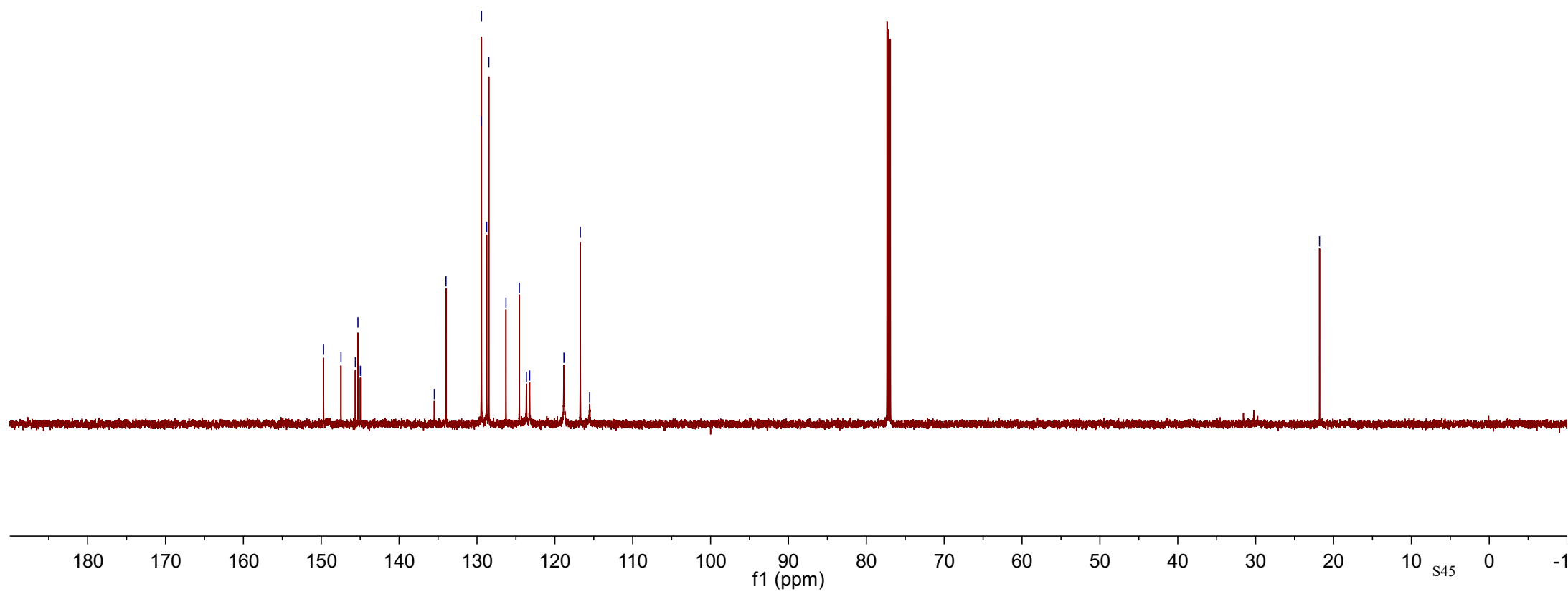
3a

Parameter	Value
Solvent	CDCl3
Spectrometer Frequency	600





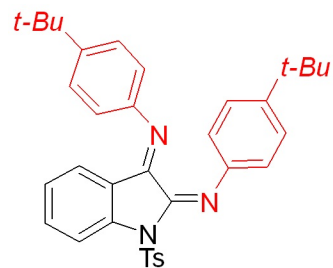
—21.80



8.24
8.23
8.01
7.35
7.33
7.32
7.26
7.24
6.84
6.82
6.81
6.77
6.73
6.72
6.60
6.58

2.47

1.33
1.31



3b

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	600

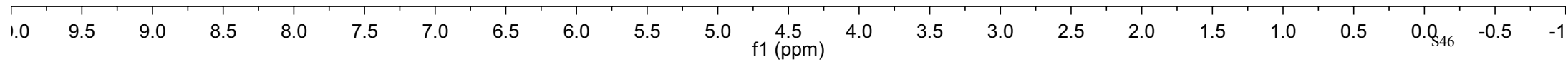
0.96
1.95

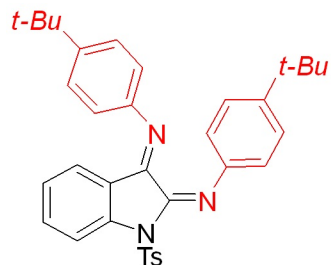
1.07
4.02
1.83

3.87
2.03

3.00

9.03
9.22



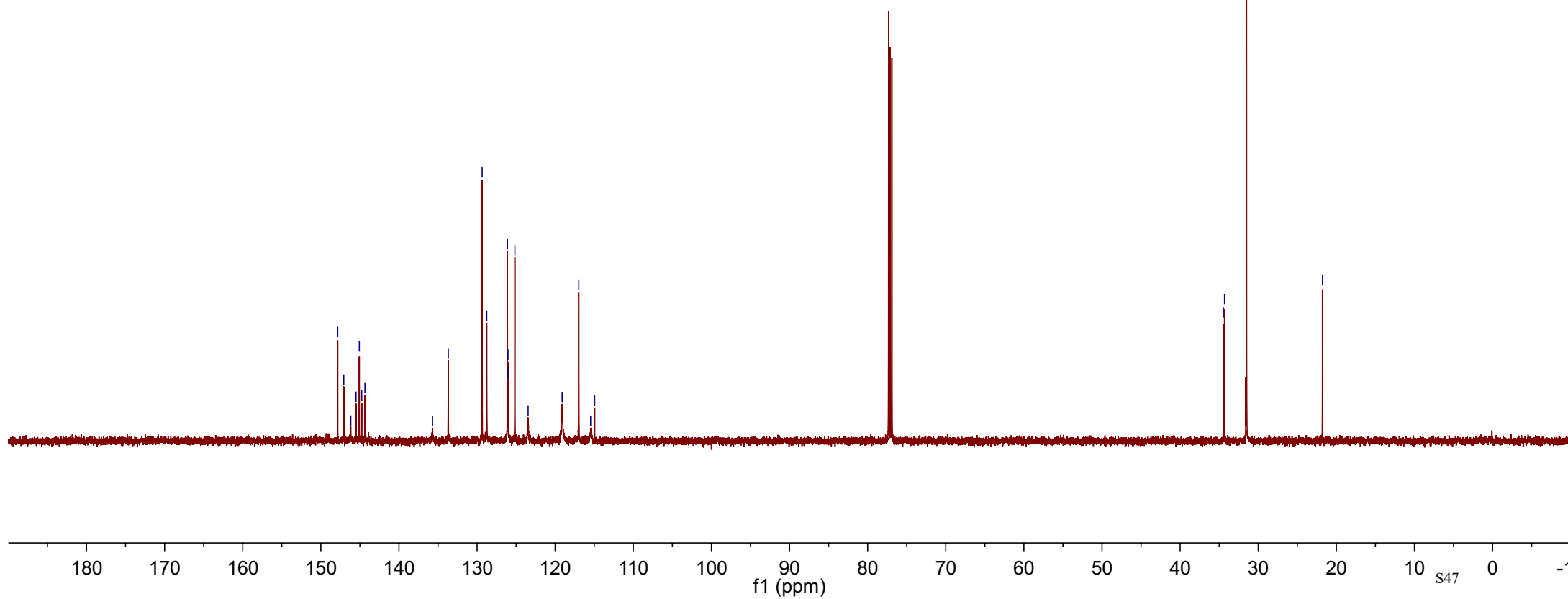


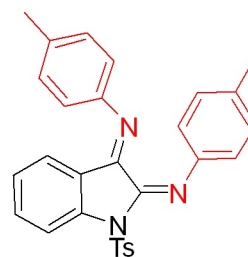
3b

147.84
147.06
146.16
145.50
145.08
144.77
144.36
135.70
133.69
129.35
128.77
126.12
126.06
126.04
125.16
123.46
119.11
116.98
115.46
114.95

34.46
34.32
31.51
31.50
— 21.78

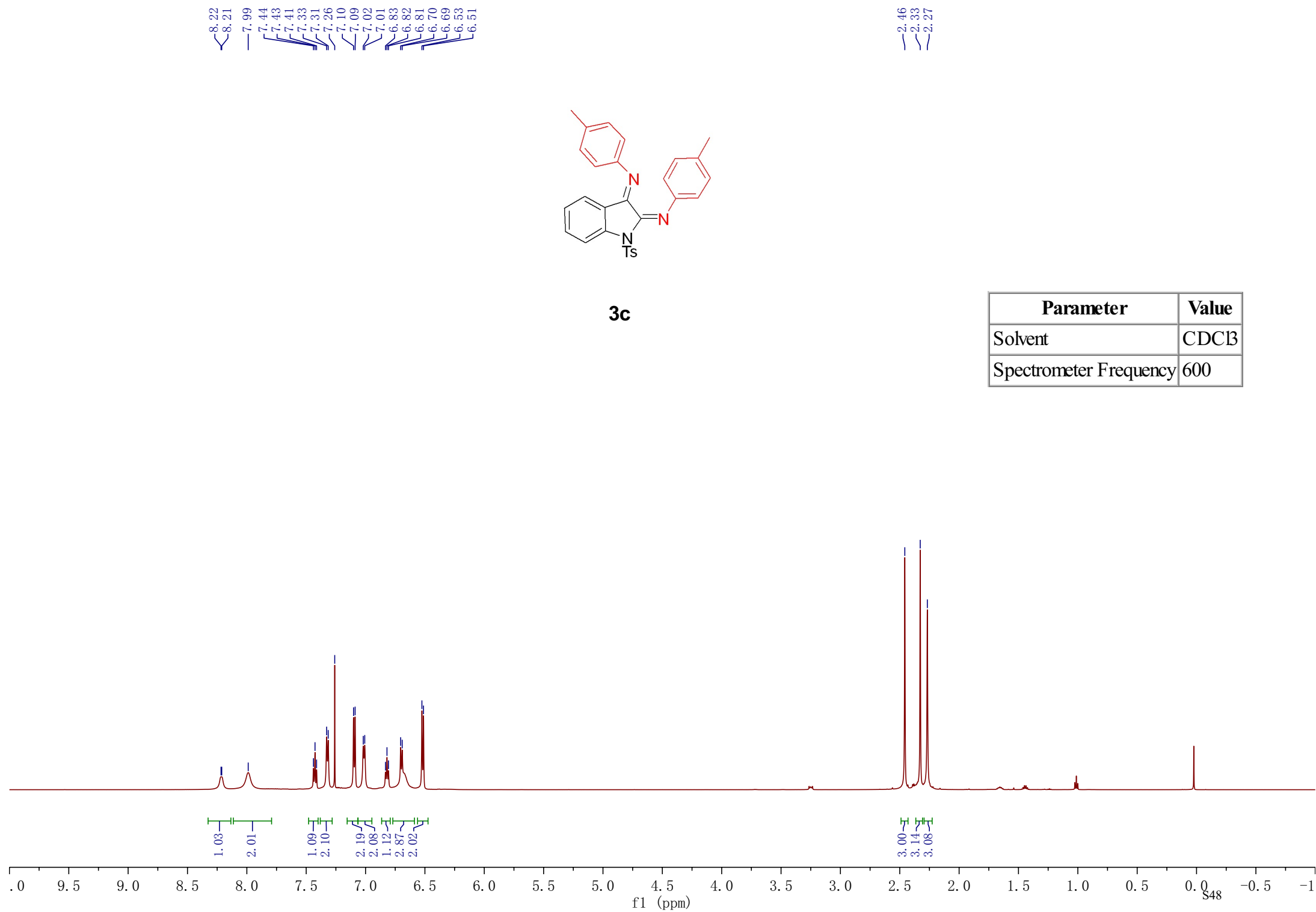
Parameter	Value
Solvent	CDCB
Spectrometer Frequency	150

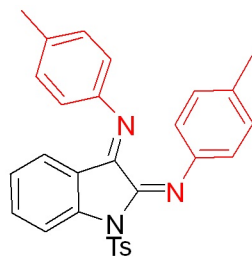




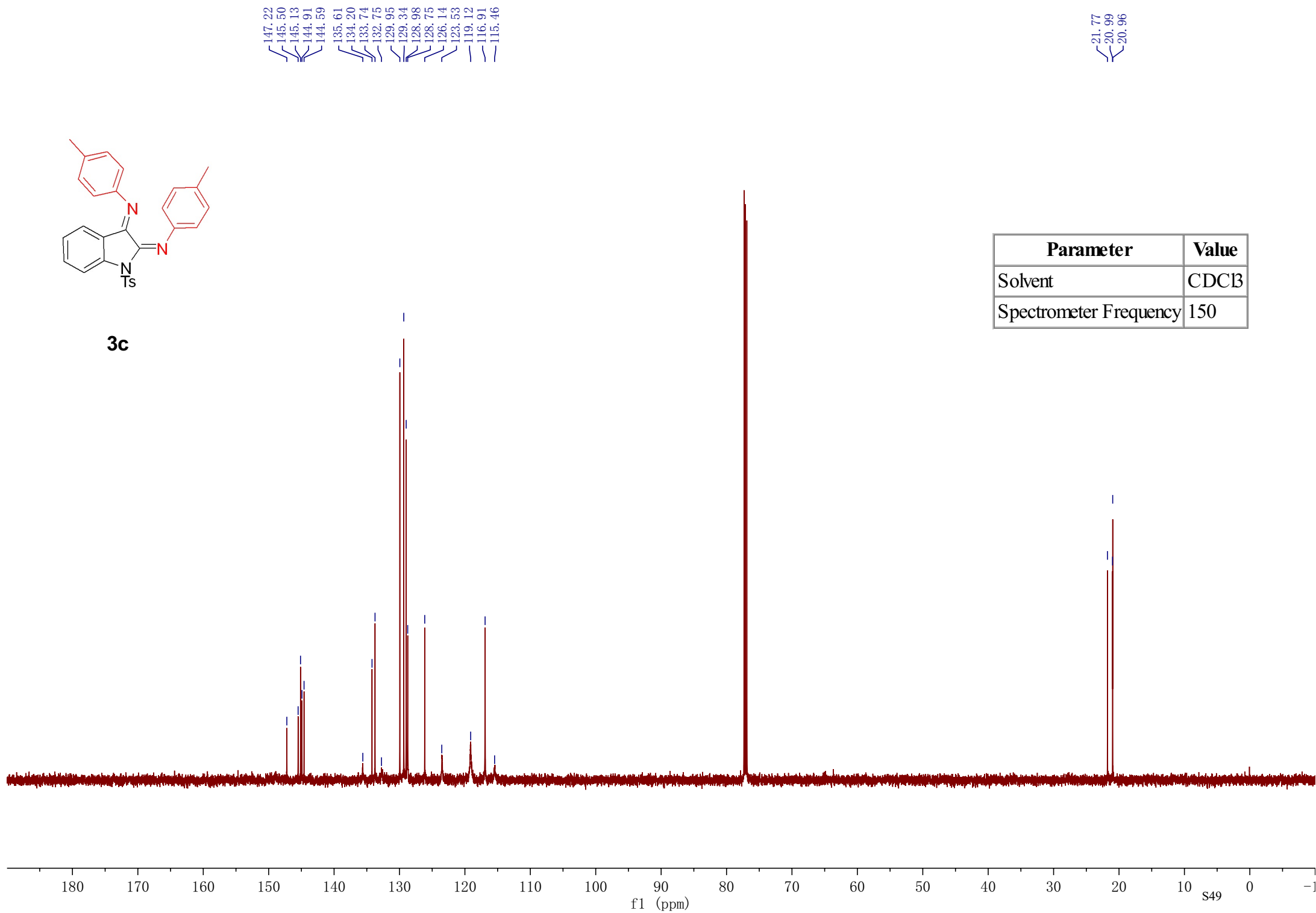
3c

Parameter	Value
Solvent	CDCB
Spectrometer Frequency	600

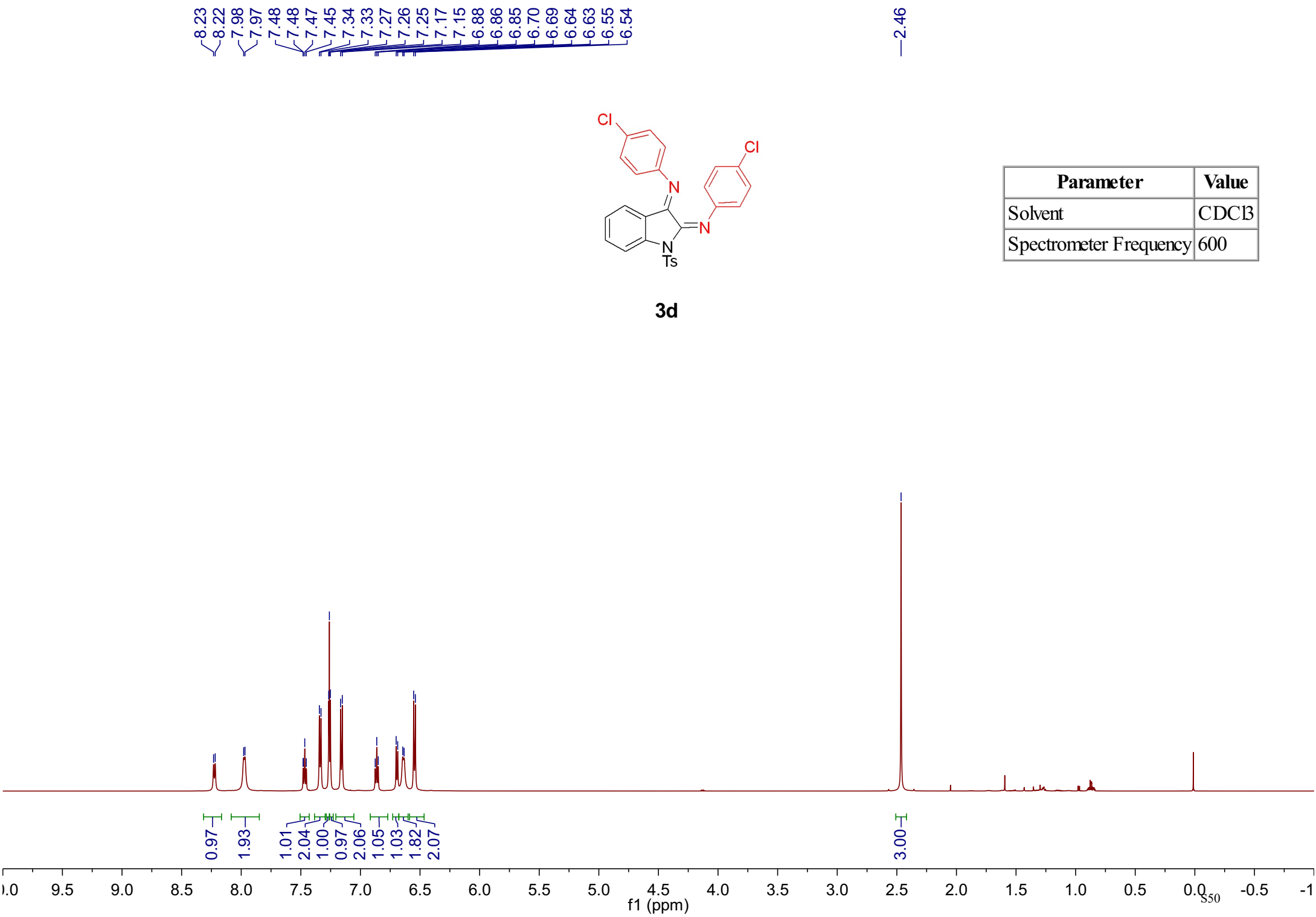


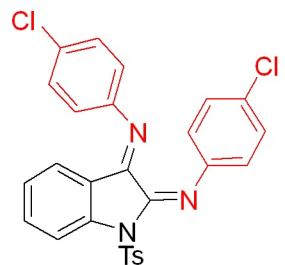


3c



Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150



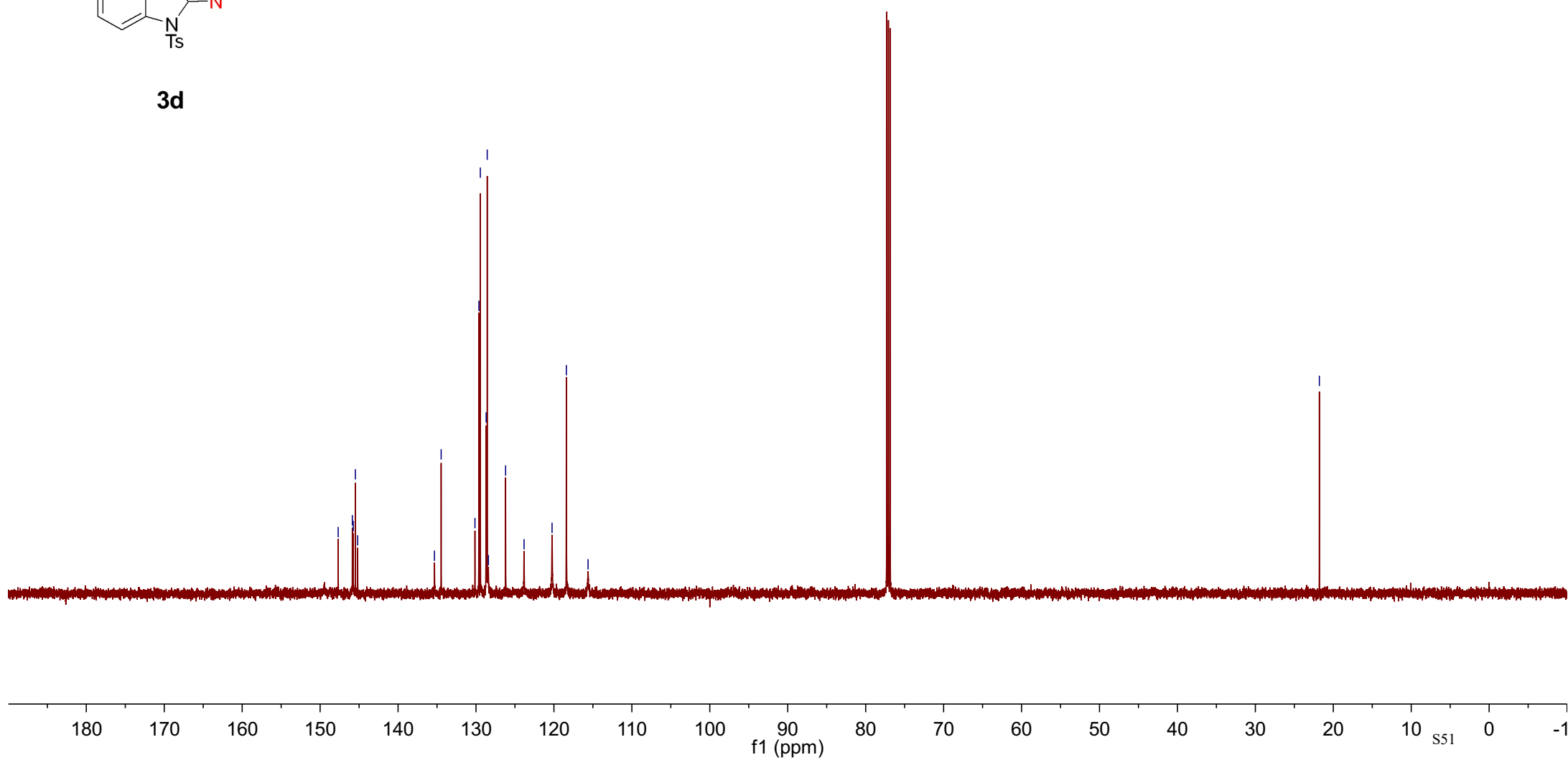


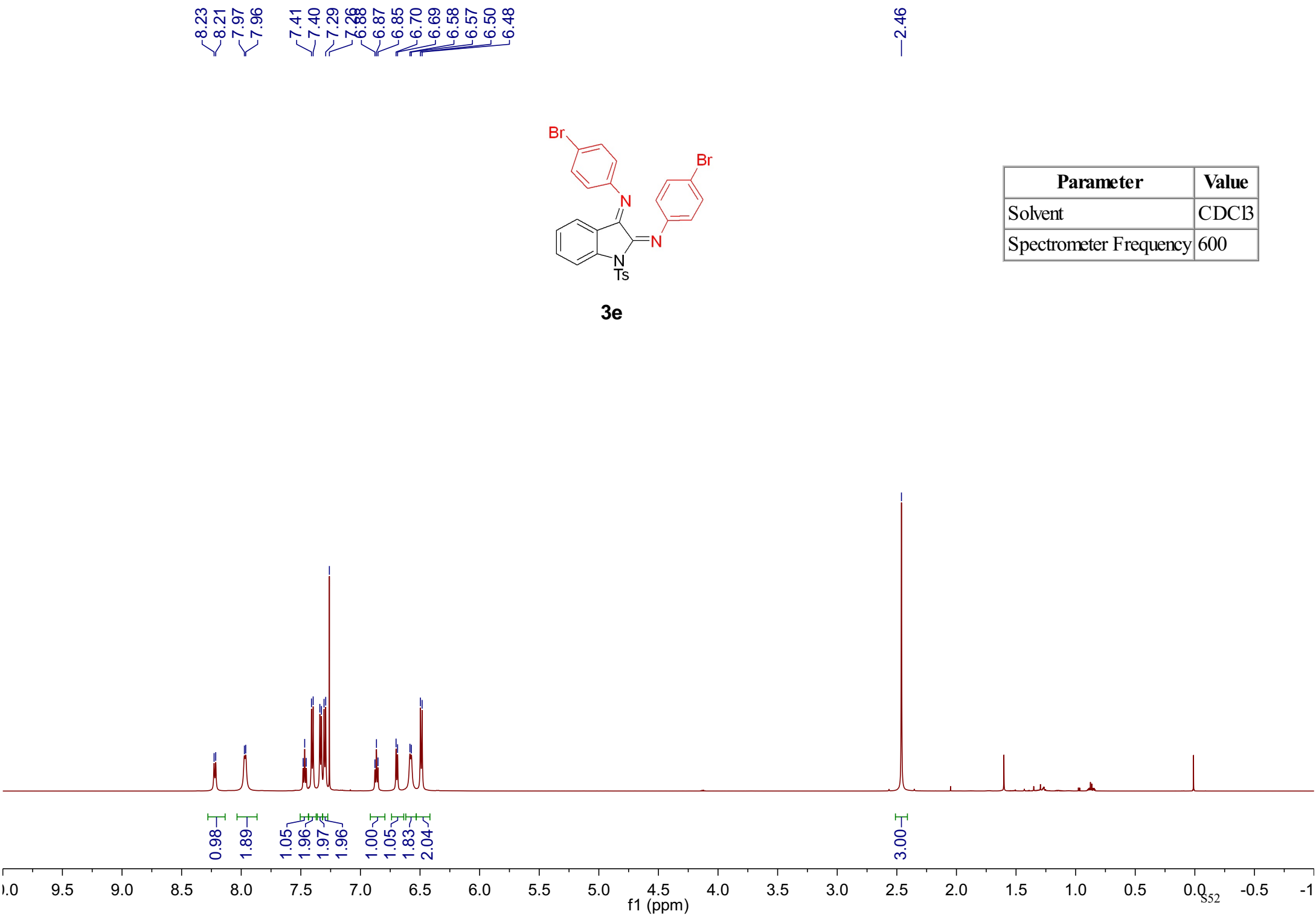
3d

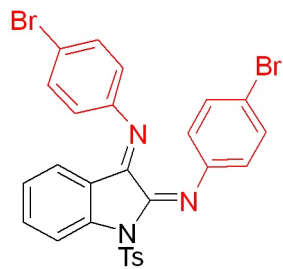
147.68
145.87
145.74
145.47
145.18
135.33
134.47
130.13
129.63
129.44
128.70
128.56
128.41
126.20
123.83
120.24
118.41
115.62

—21.79

Parameter	Value
Solvent	CDCI3
Spectrometer Frequency	150





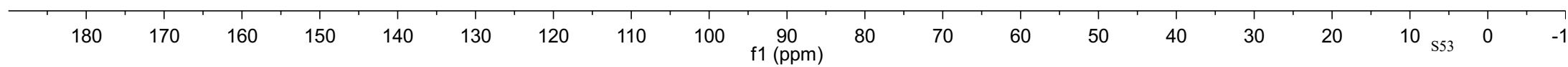


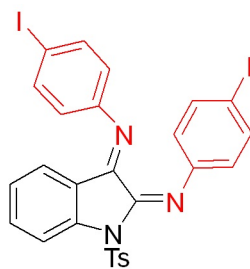
3e

149.28
148.13
146.37
145.75
145.49
145.08
135.30
134.51
132.57
131.50
129.45
128.70
126.23
123.87
120.62
118.76
118.35
117.77
116.10
115.62

—21.80

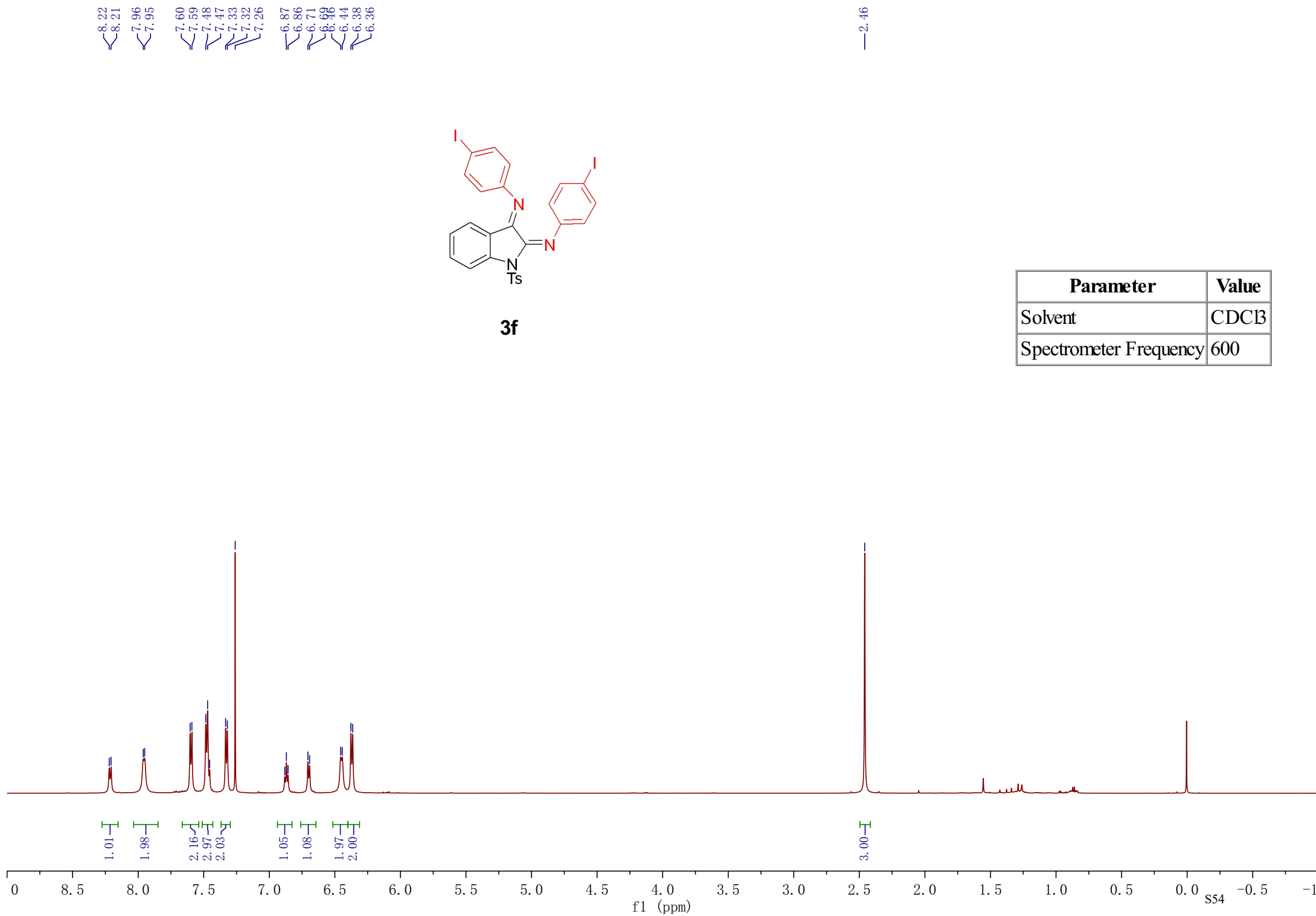
Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150

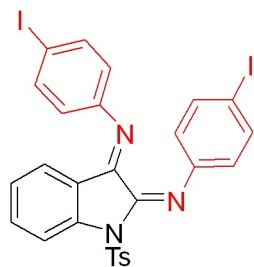




3f

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	600





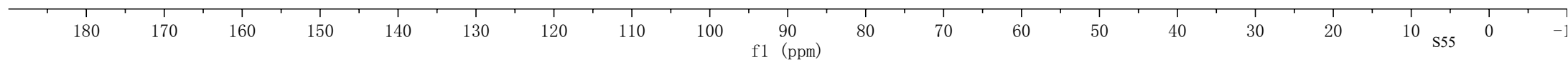
3f

148.78
147.04
145.75
145.45
144.94
138.48
137.43
135.28
134.49
129.43
128.70
126.24
123.88
120.98
119.10
115.61

—88.34

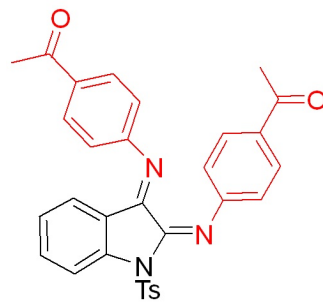
—21.79

Parameter	Value
Solvent	CDCI3
Spectrometer Frequency	150



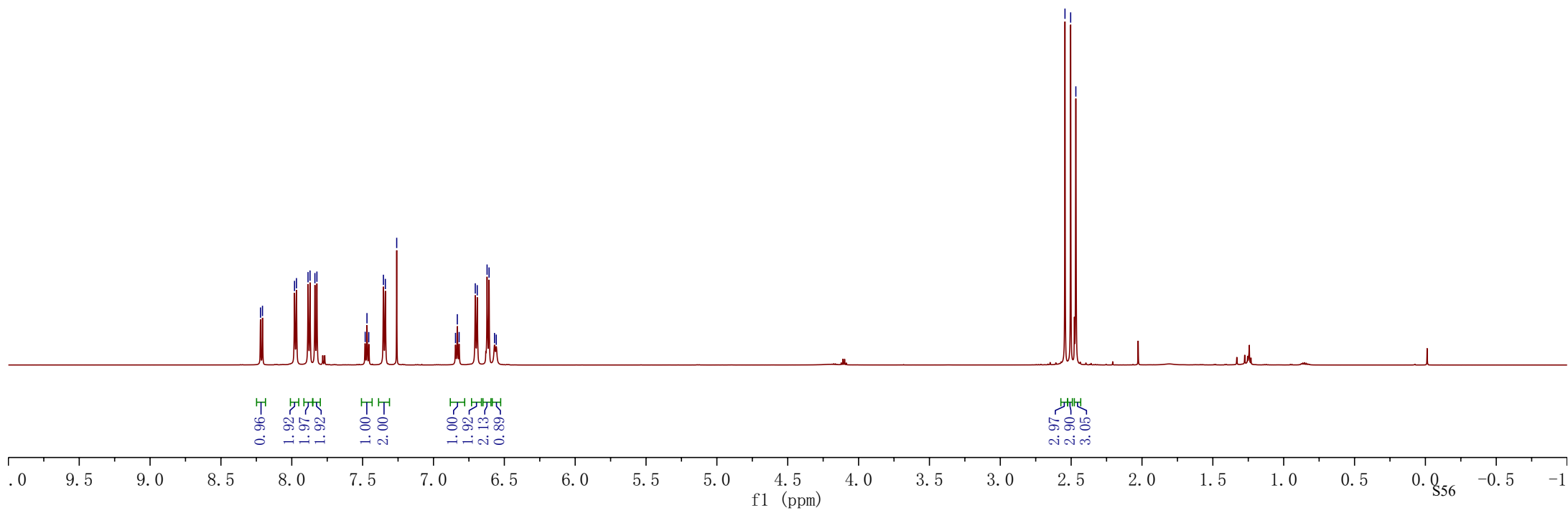
8.22
8.21
7.89
7.87
7.84
7.82
7.48
7.47
7.35
7.34
7.26
6.83
6.82
6.71
6.69
6.62
6.61
6.57
6.56

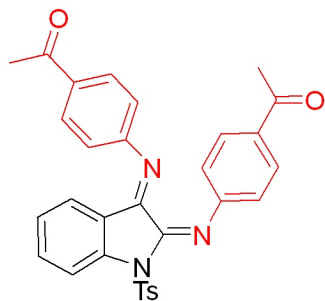
2.54
2.50
2.47



3g

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	600





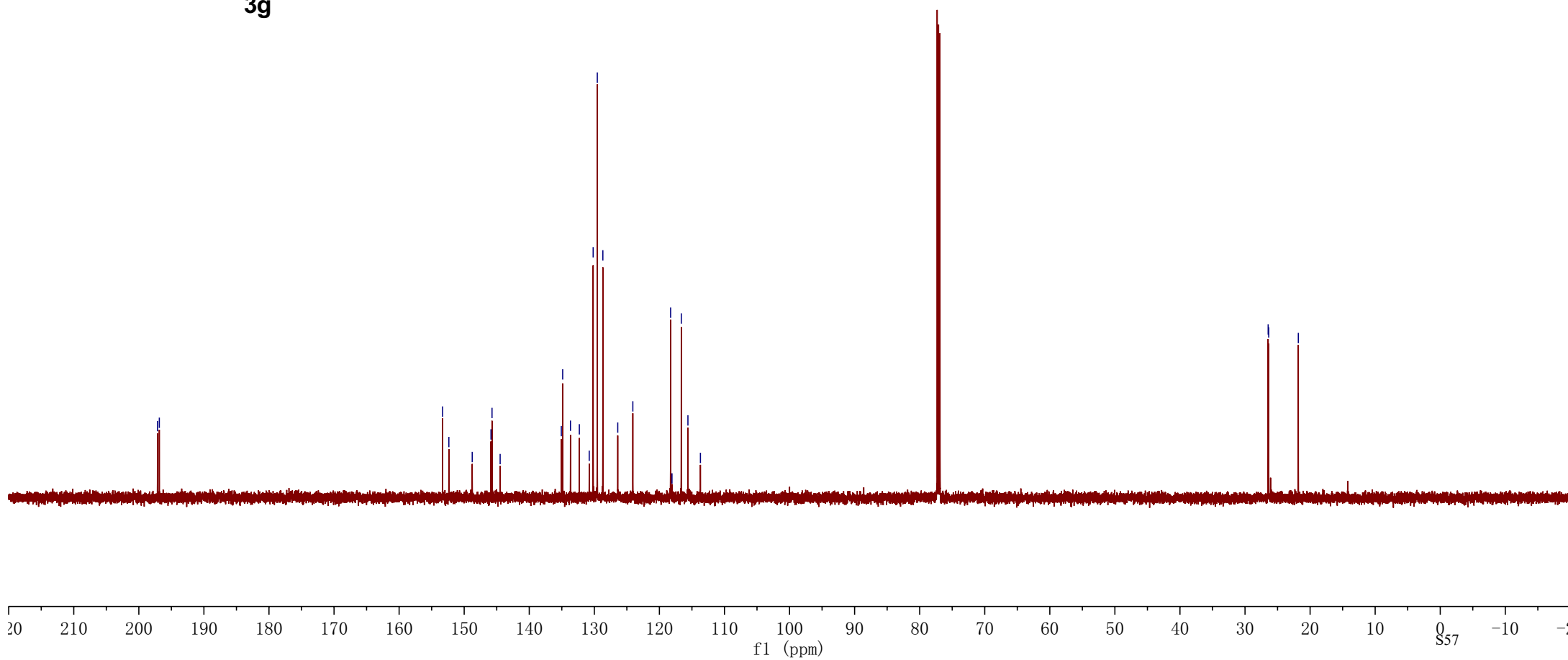
3g

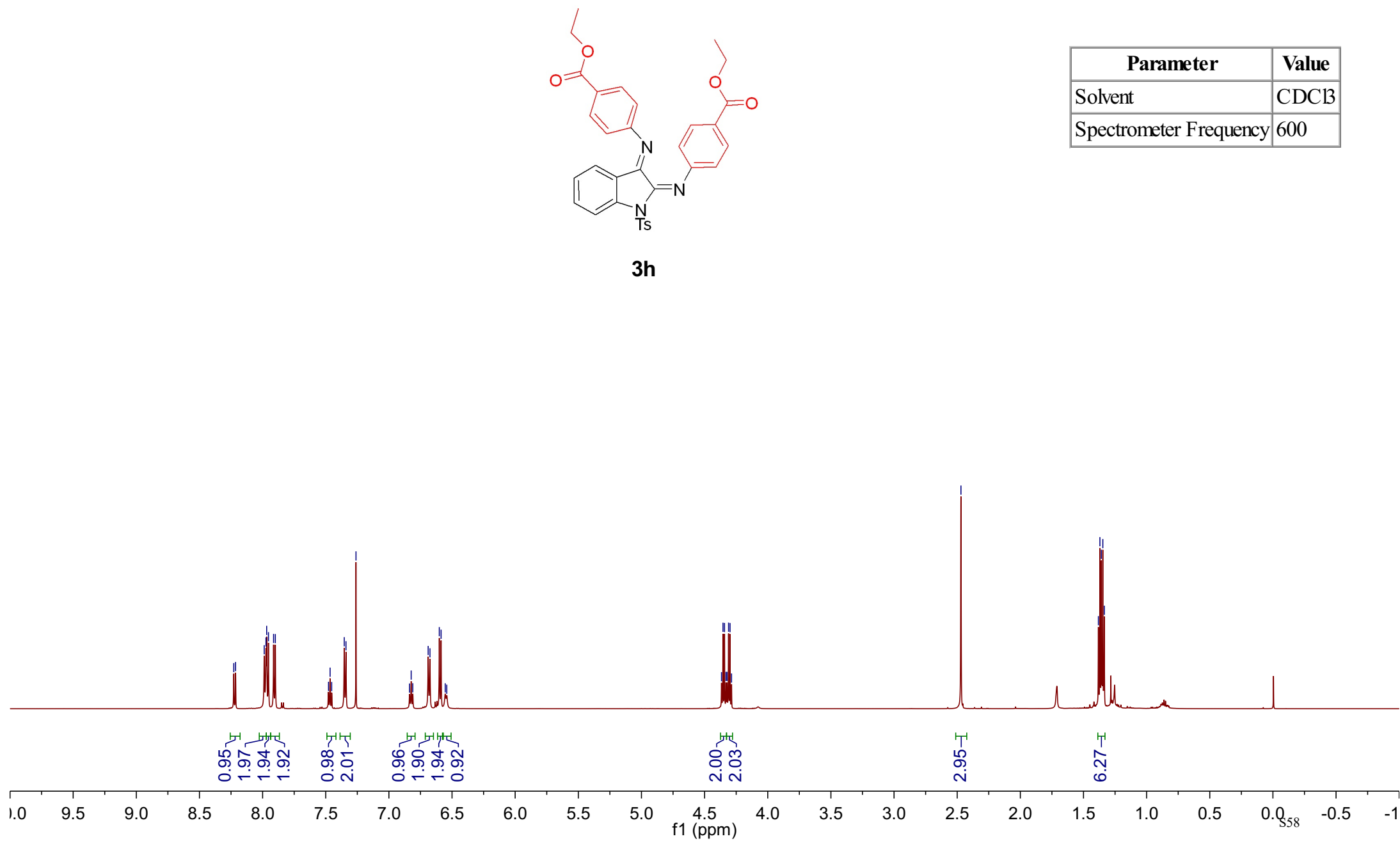
Parameter	Value
Solvent	CDCB
Spectrometer Frequency	150

197.14
196.85

153.32
152.35
148.76
145.88
145.72
144.47
134.86
133.67
132.30
130.18
129.54
128.69
126.40
124.97
118.06
116.63
115.62
113.69

26.44
26.36
21.81





166.56
166.10

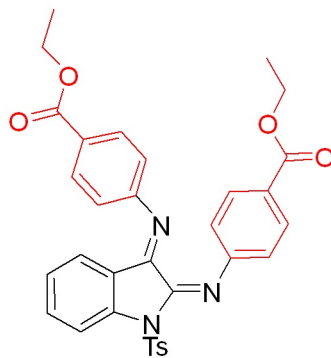
153.18
151.99
148.82
145.85
145.64
144.54

134.71
131.23
130.51
129.51
128.71
126.75
126.15
116.43
115.60
113.77

60.99
60.64

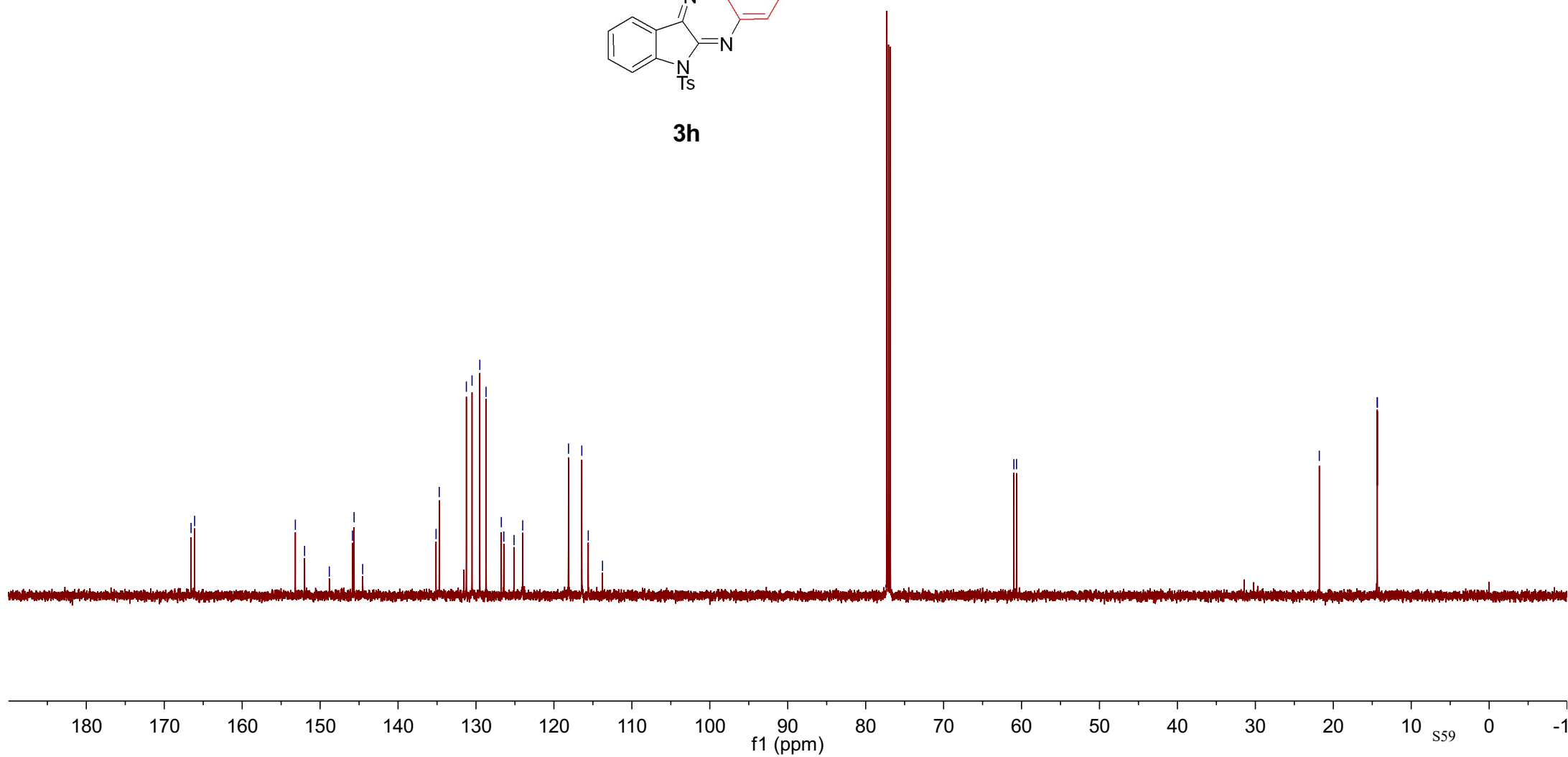
21.79

14.36
14.35



3h

Parameter	Value
Solvent	CDCl3
Spectrometer Frequency	150



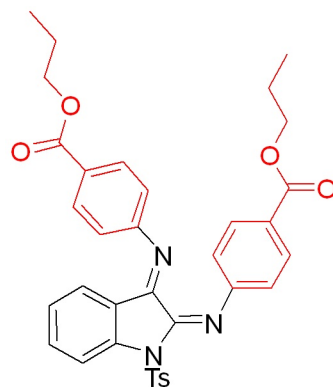
8.23
8.22
7.99
7.97
7.97
7.96
7.91
7.90

7.47
7.35
7.34
7.26
6.83
6.82
6.69
6.68
6.60
6.59
6.56
6.55

4.26
4.25
4.24
4.22
4.21
4.20

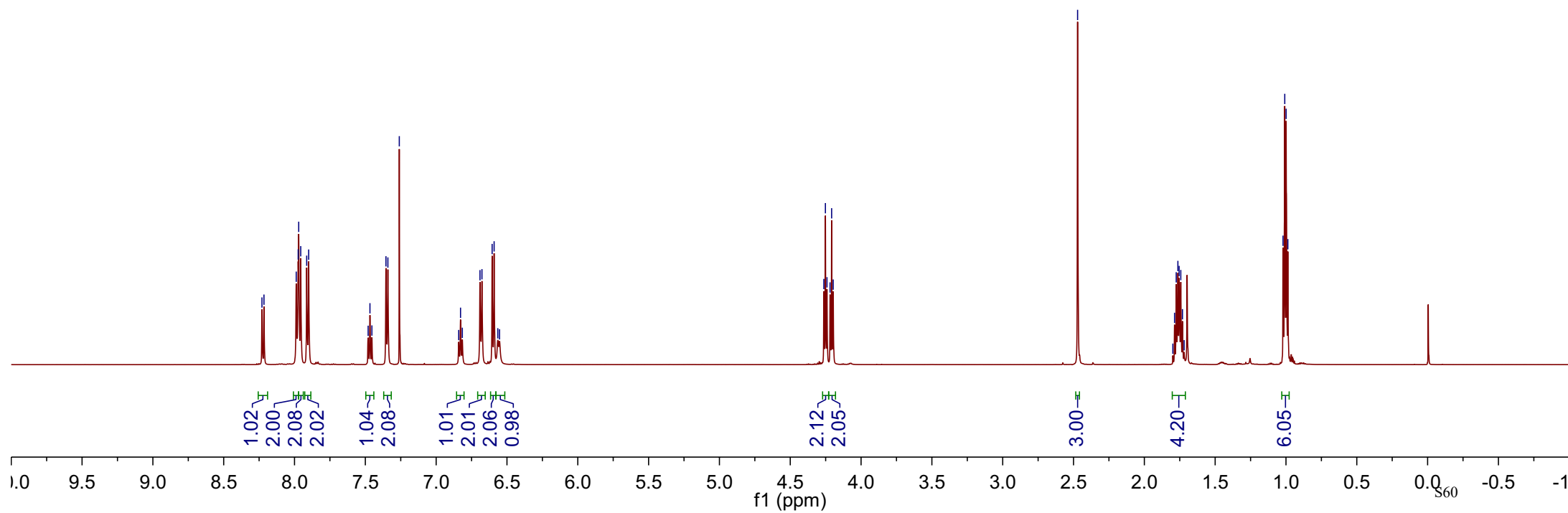
2.47

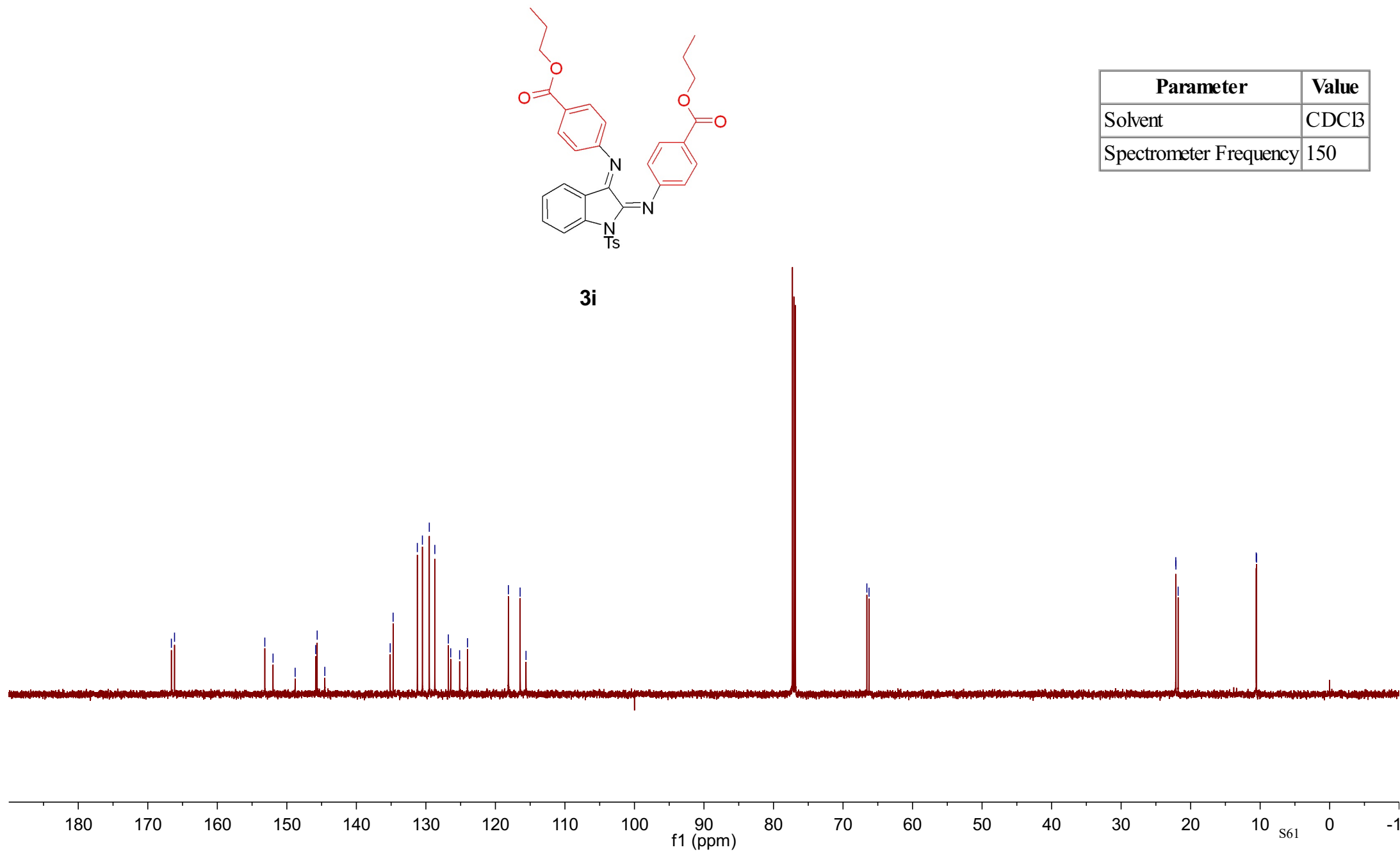
1.79
1.77
1.76
1.75
1.74
1.73
1.01
1.00
0.99



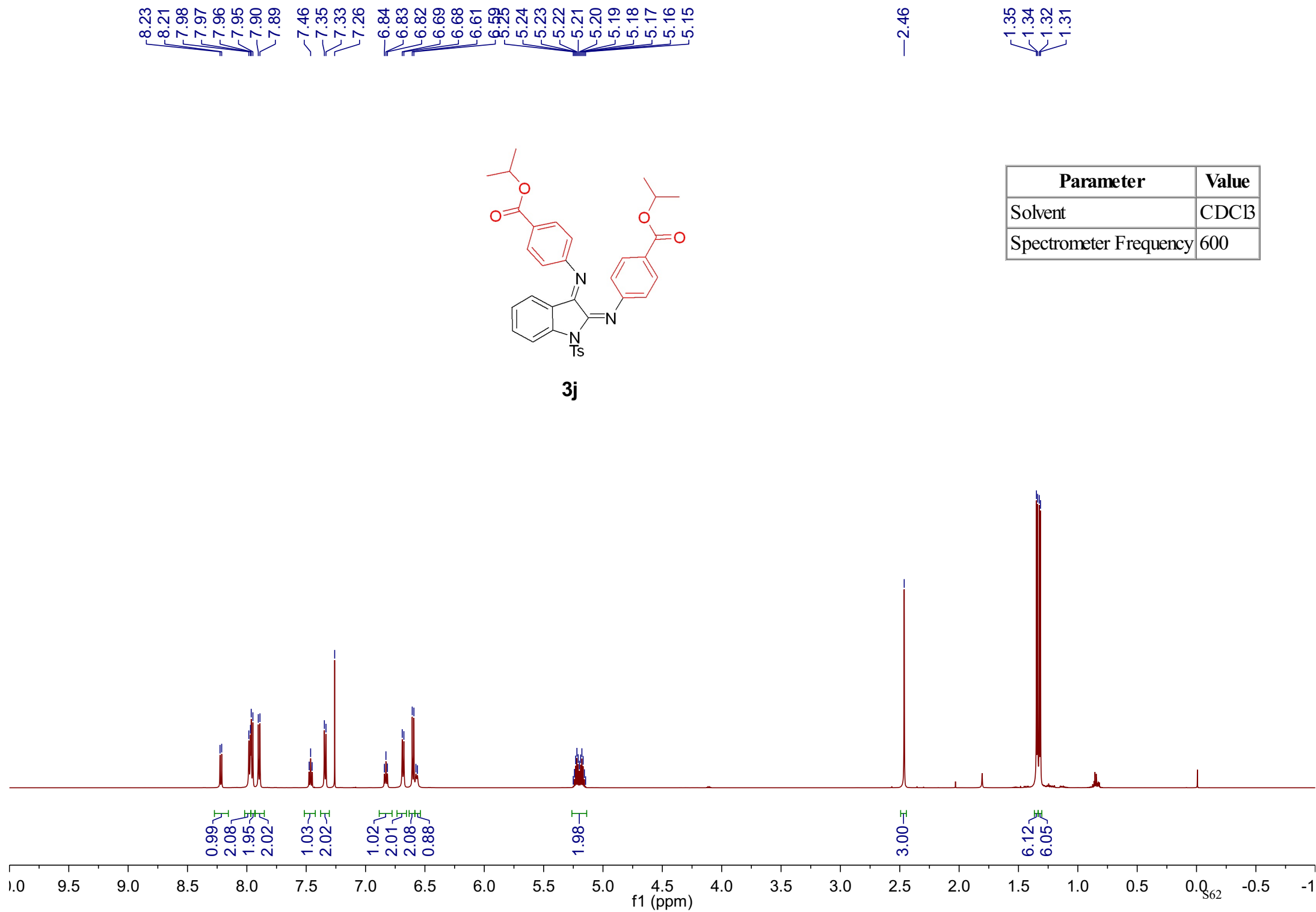
3i

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	600





Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150



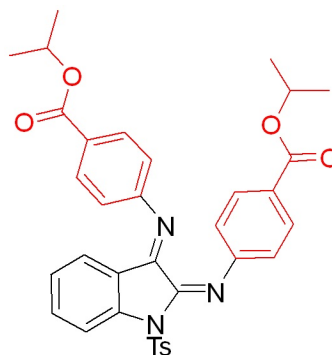
166.01
165.57

153.11
151.87
148.81
145.84
145.63
144.52

134.70
131.19
130.46
129.50
128.72
124.91
118.16
118.10
116.39
115.59

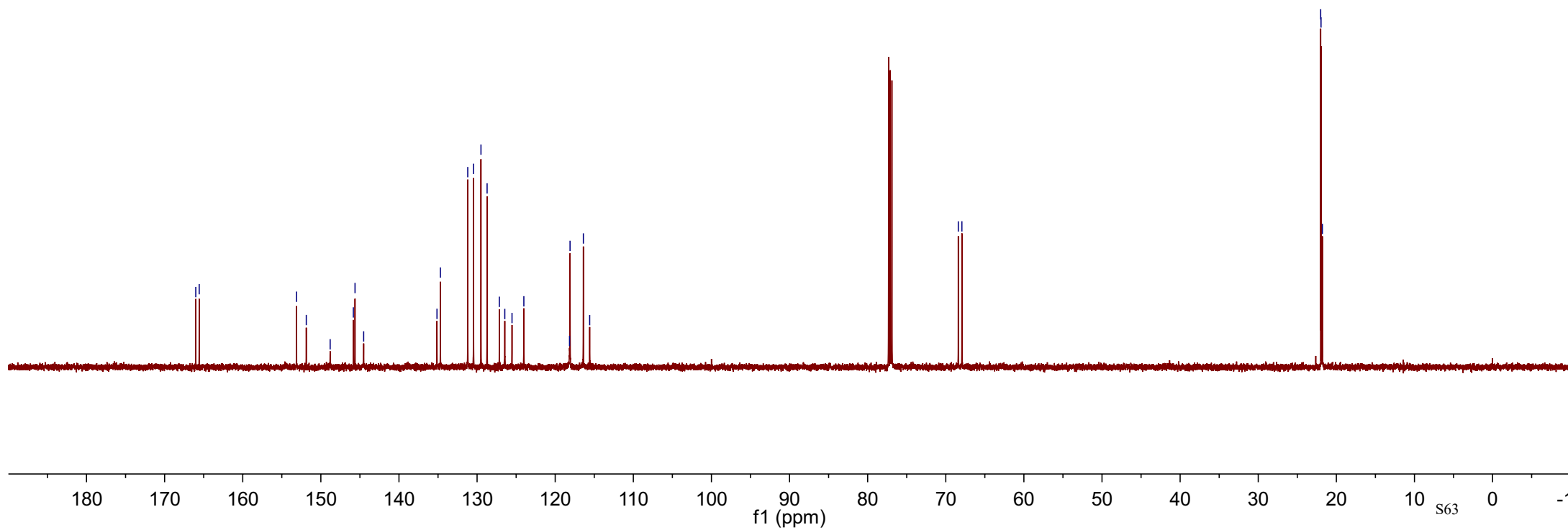
68.40
67.94

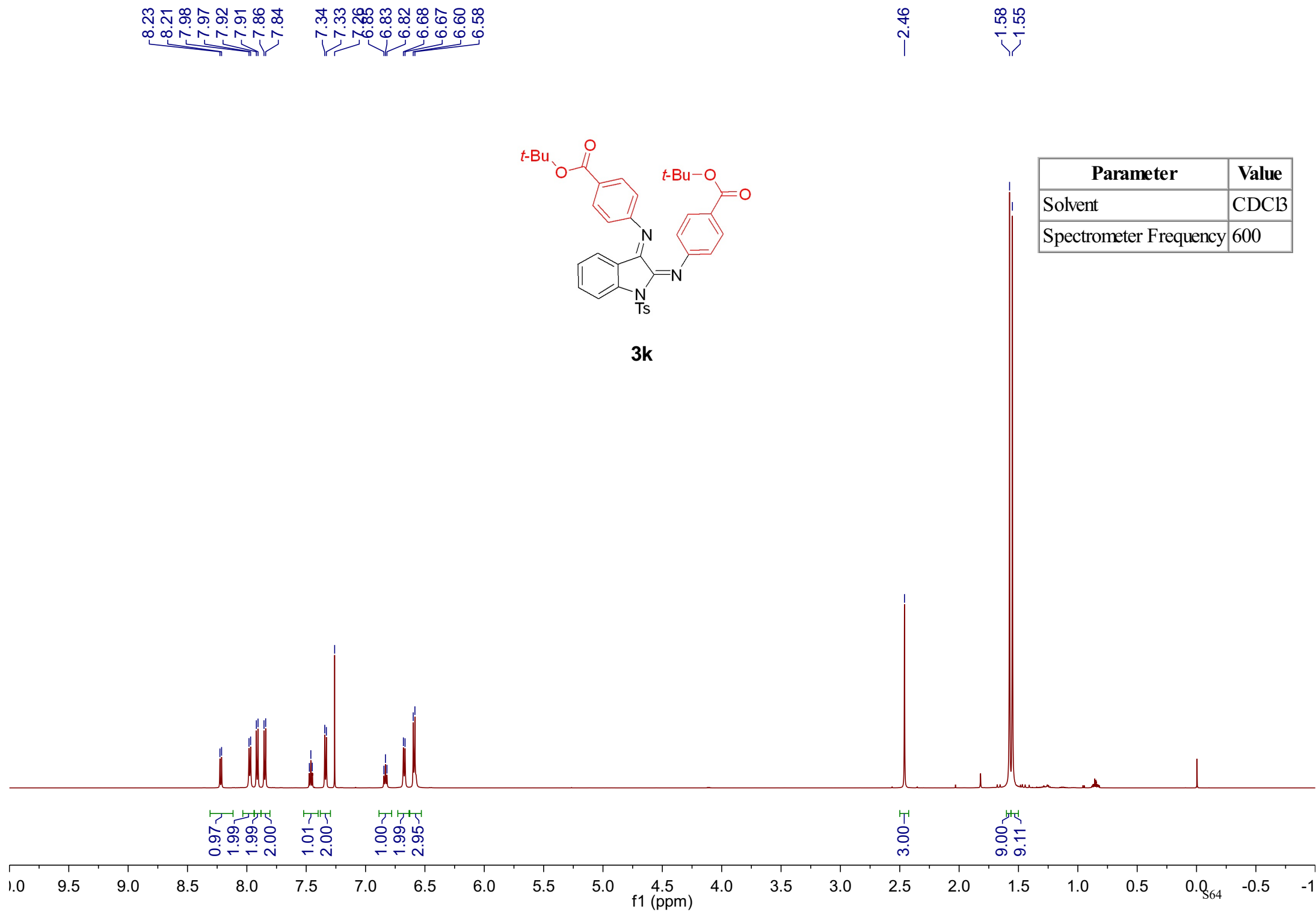
22.01
21.96
21.79



3j

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150





165.70
165.27

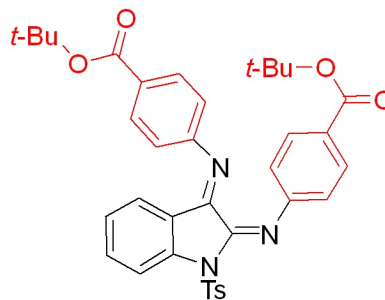
152.91
151.58
148.80
145.82
145.60
144.52

134.64
131.07
130.33
129.50
128.72
128.26
128.22
118.05
116.33
115.58

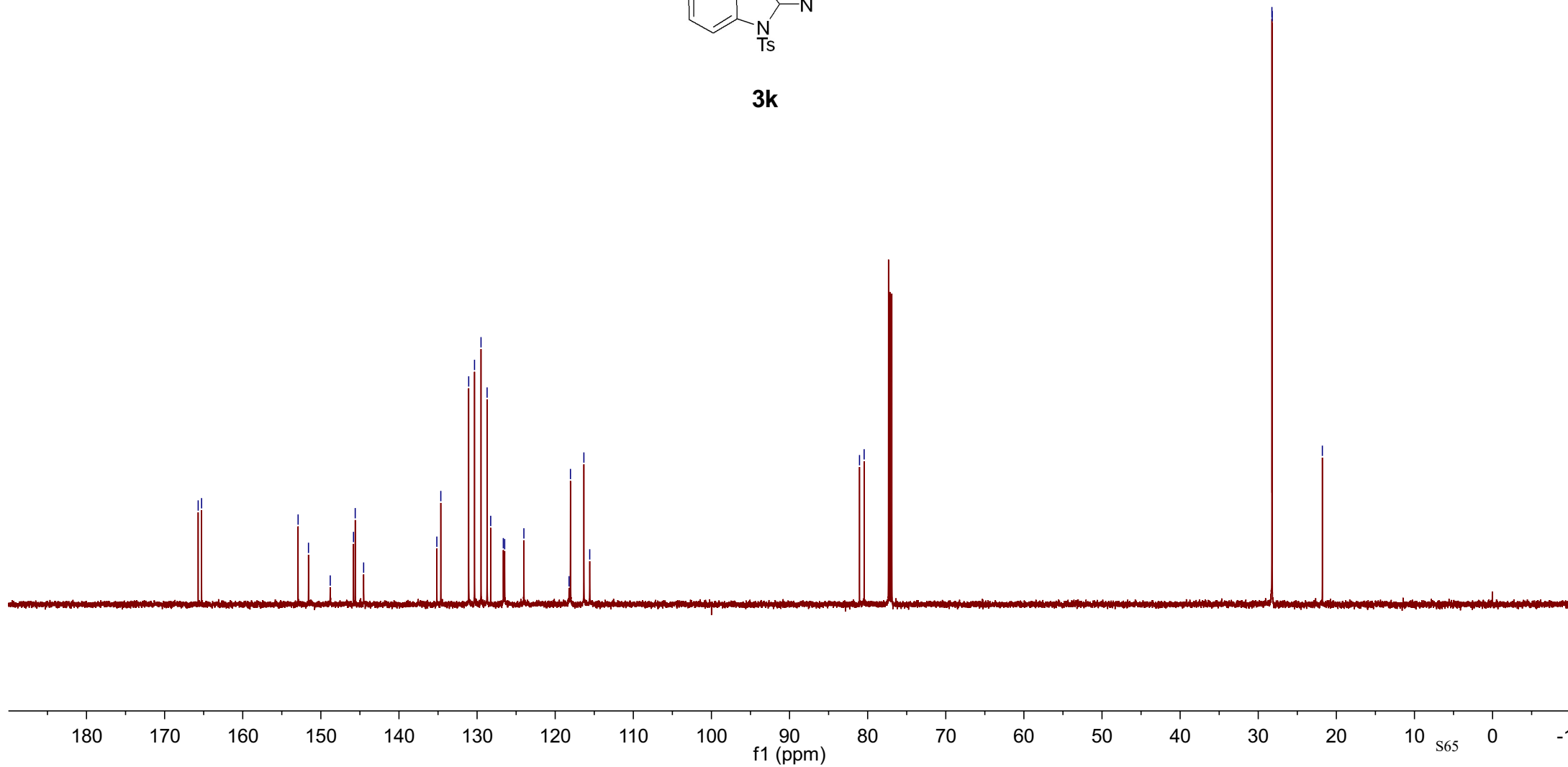
81.04
80.45

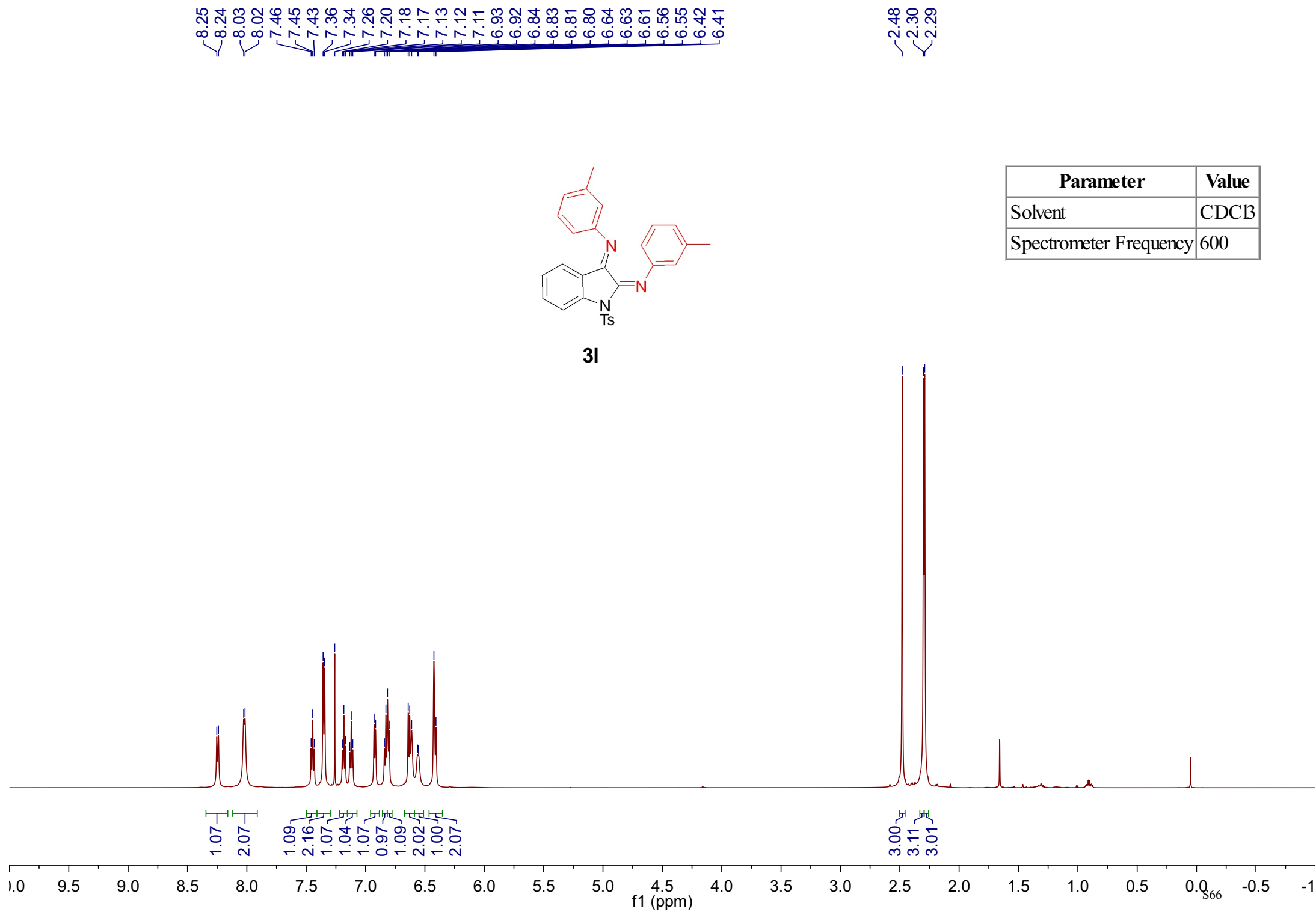
28.25
28.21
21.79

Parameter	Value
Solvent	CDCB
Spectrometer Frequency	150



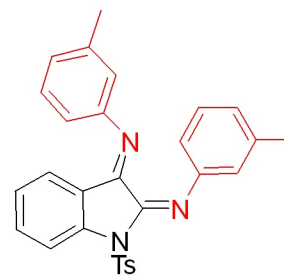
3k





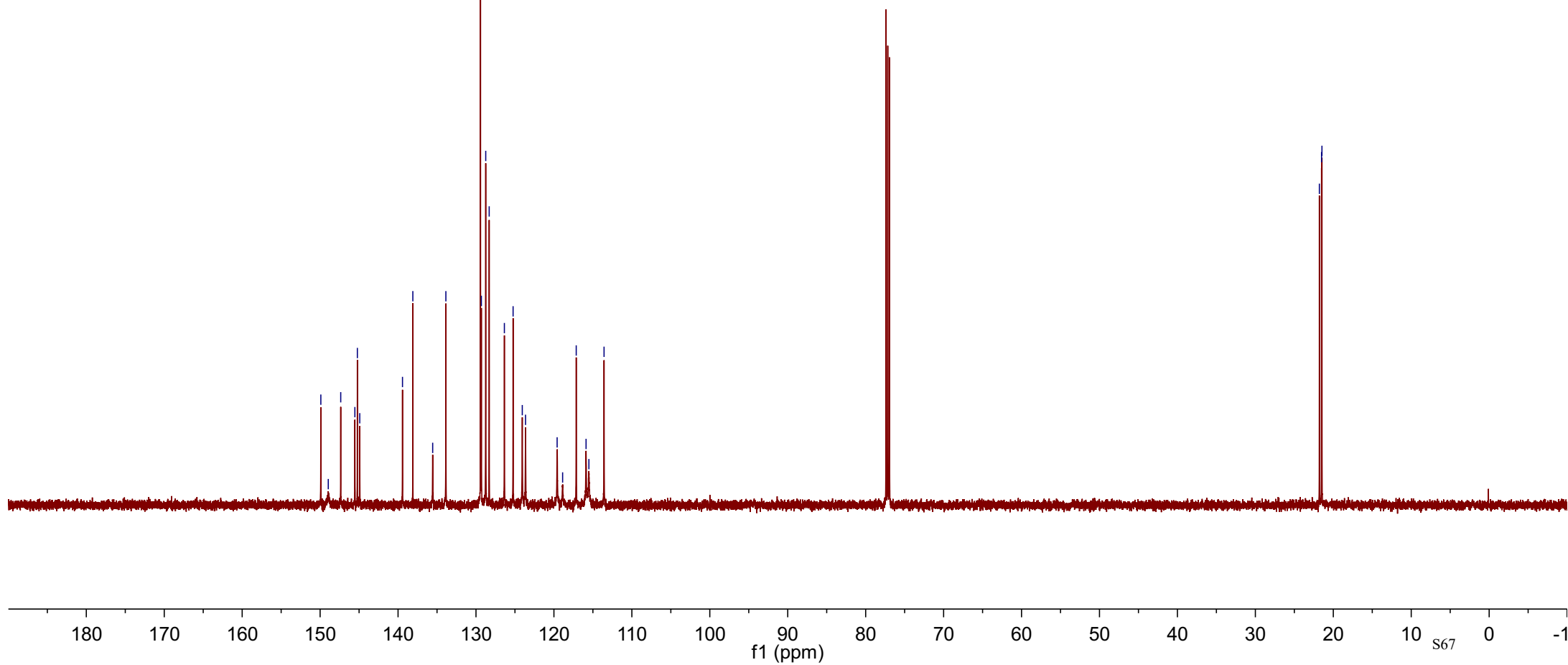
149.91
148.95
147.36
145.54
145.22
144.91
139.43
138.10
135.56
133.86
129.42
129.31
128.75
128.31
126.36
125.24
124.06
123.64
119.59
118.88
117.14
115.88
115.51
113.57

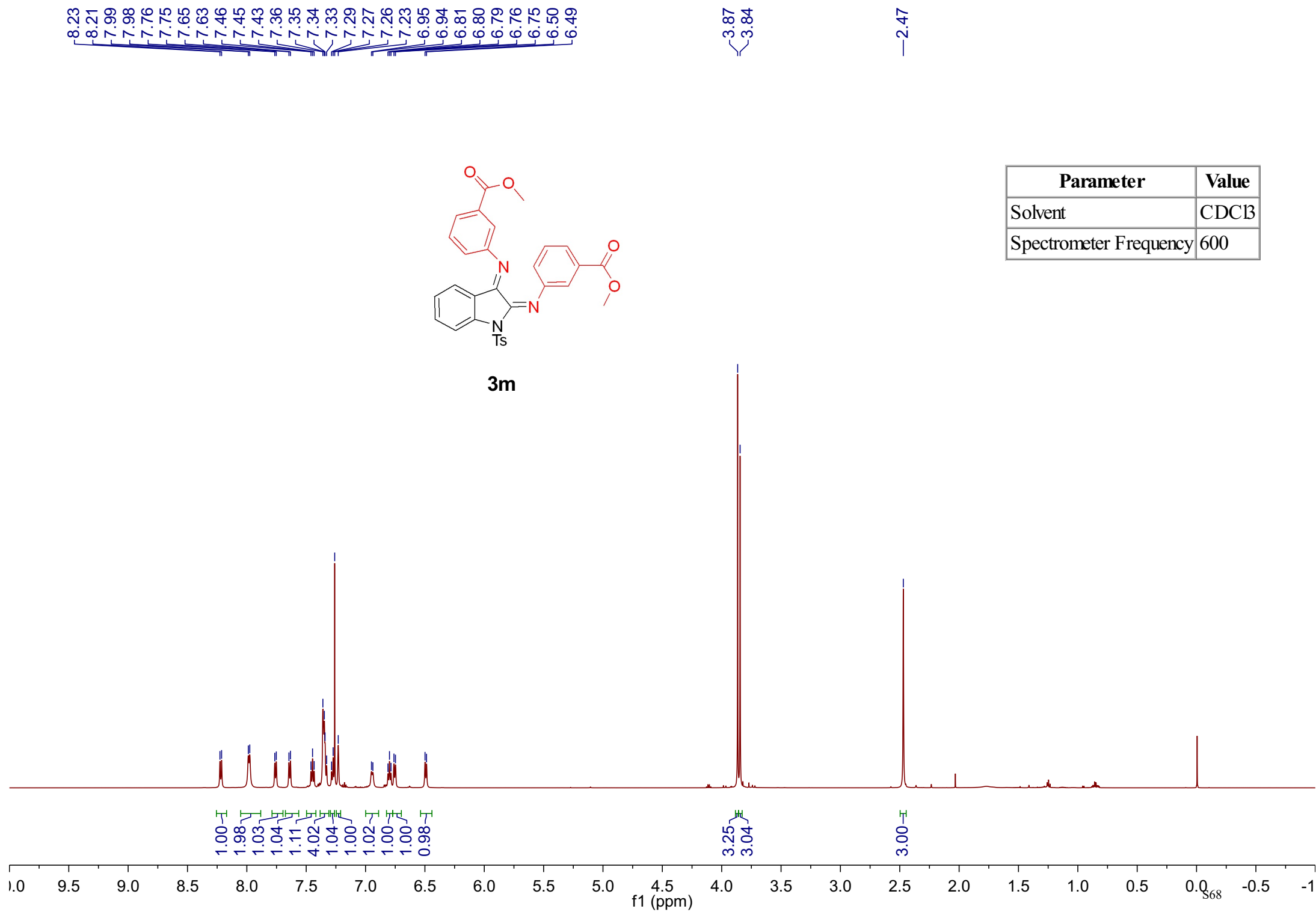
21.79
21.51
21.46



3I

Parameter	Value
Solvent	CDCI3
Spectrometer Frequency	150



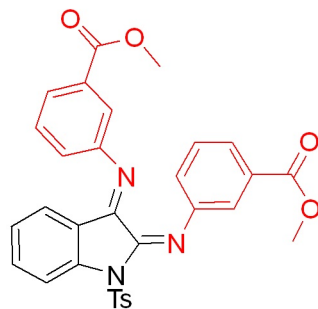


167.07
166.48

149.45
147.45
145.79
145.60
145.38
135.21
134.50
131.43
130.46
129.67
129.54
128.67
128.59
127.14
126.26
125.70
124.47
123.91
123.61
121.39
119.98
118.37
117.71
115.67

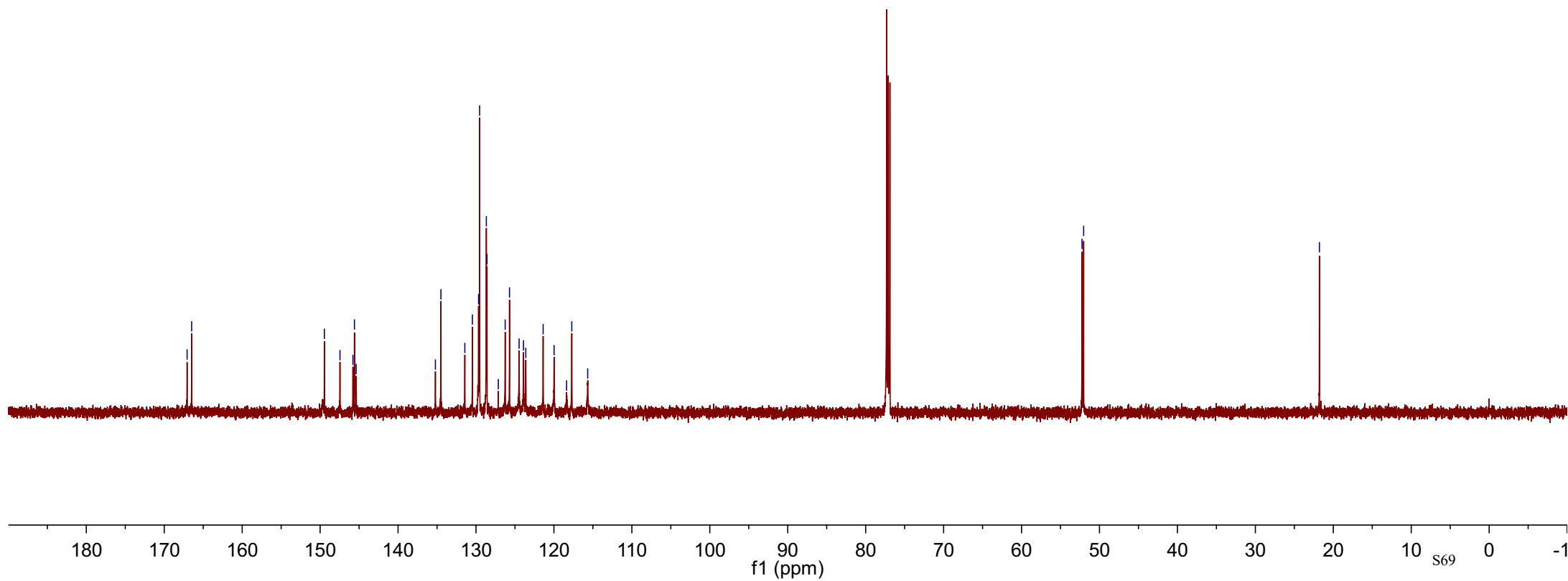
52.24
52.03

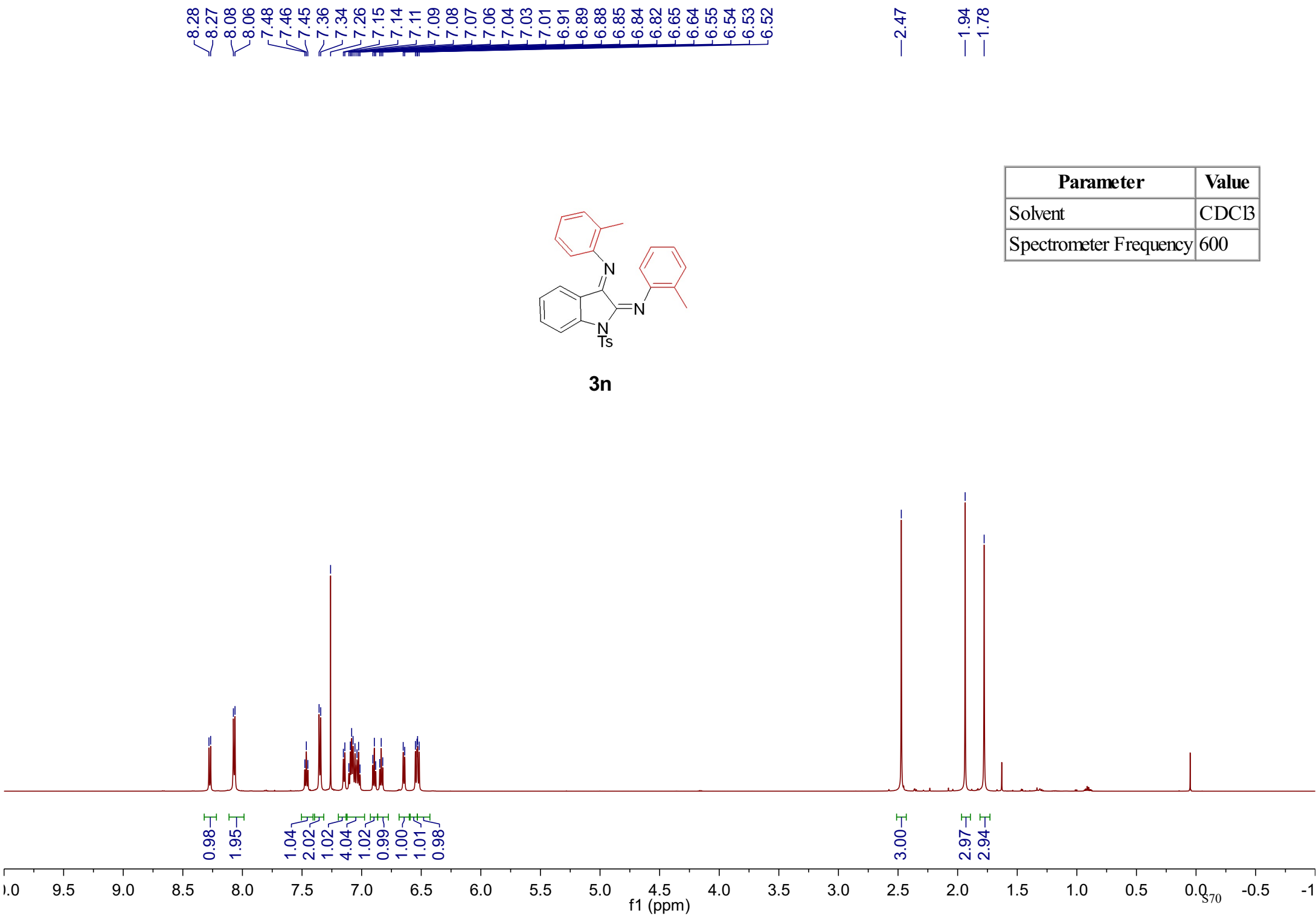
21.78

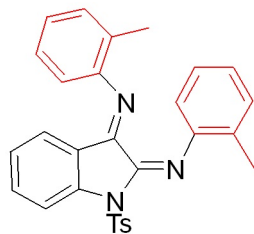


3m

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150





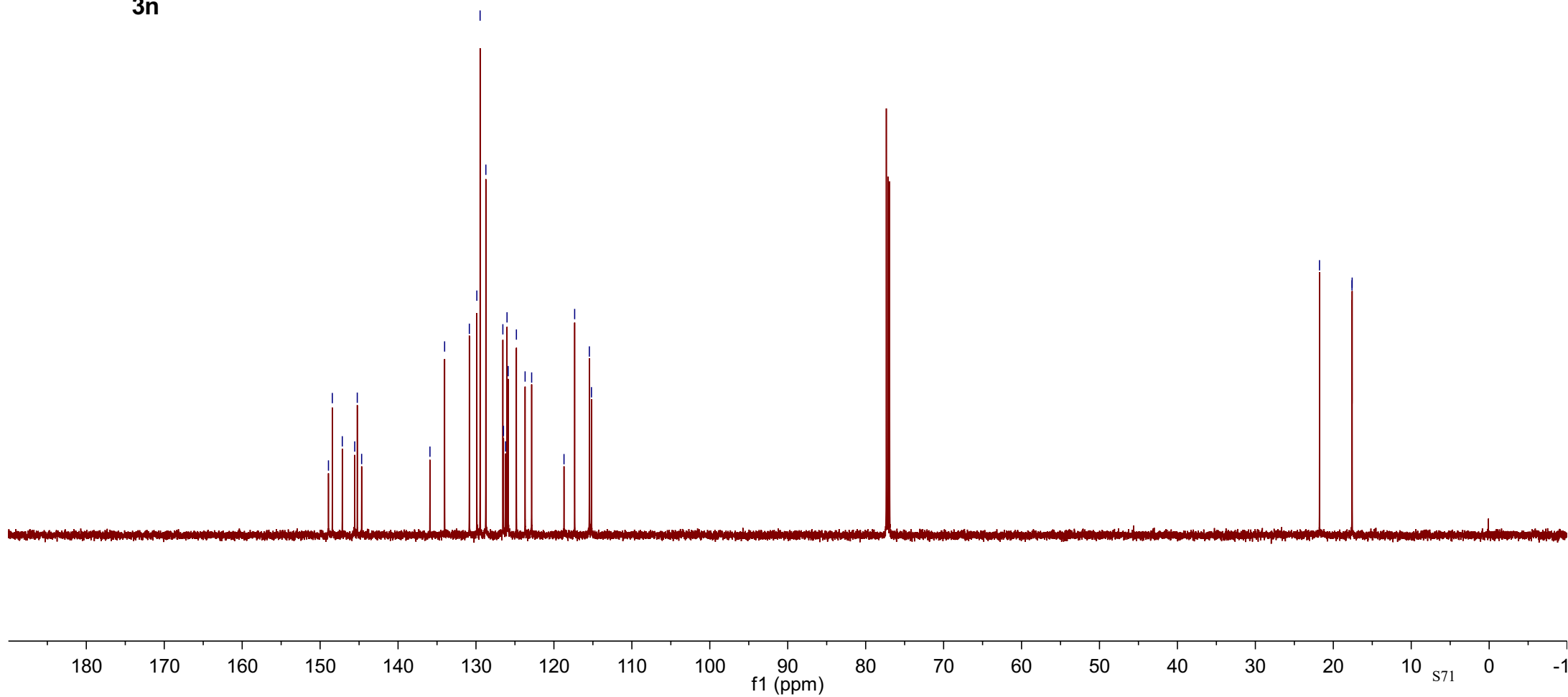


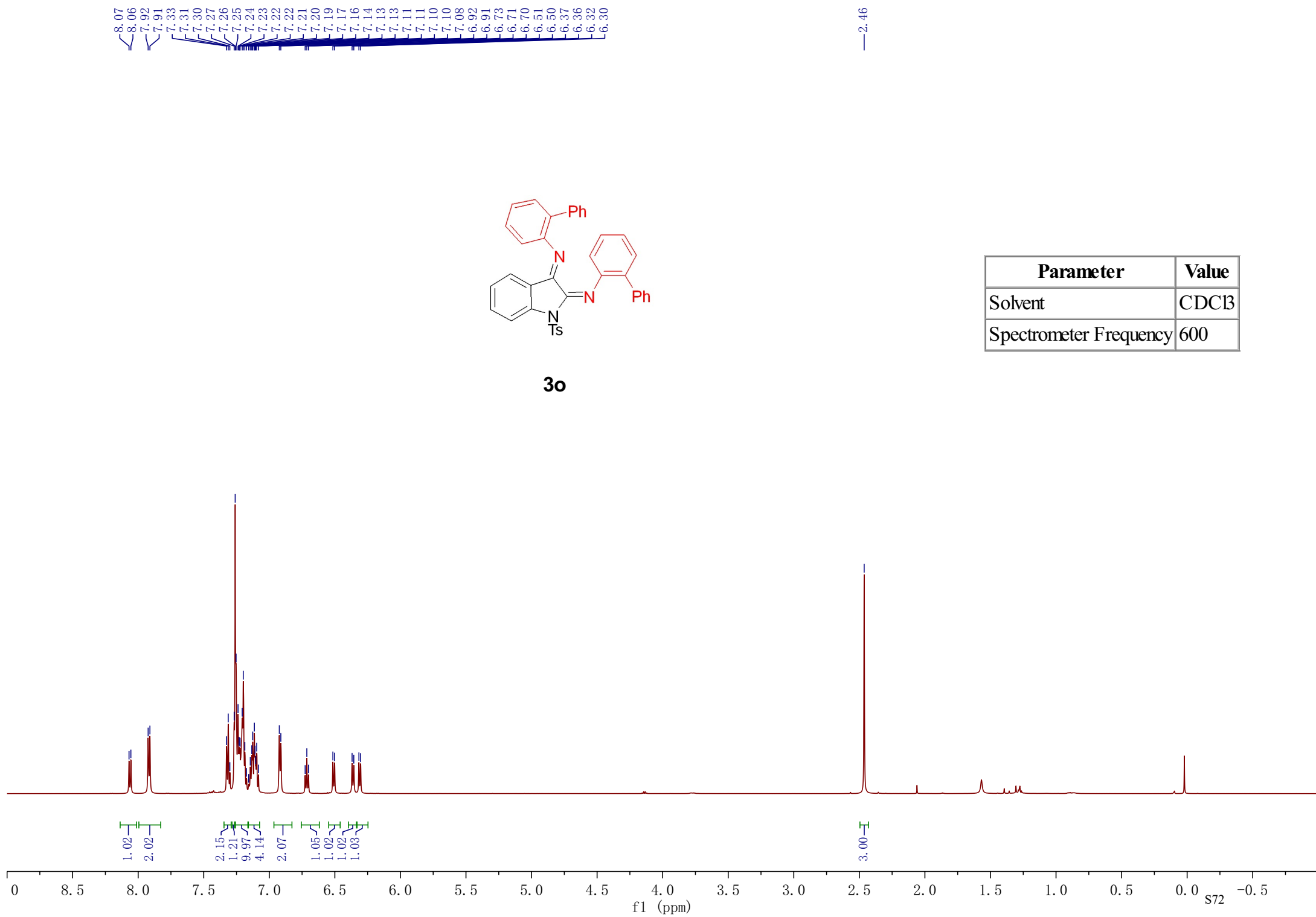
3n

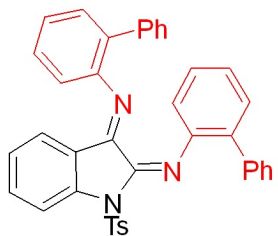
148.93
148.43
147.15
145.56
145.23
144.67
135.91
134.03
130.85
129.89
129.47
128.73
126.55
126.47
126.20
126.02
125.87
124.82
123.70
122.86
118.71
117.34
115.45
115.19

21.77
17.60
17.58

Parameter	Value
Solvent	CDCI3
Spectrometer Frequency	150

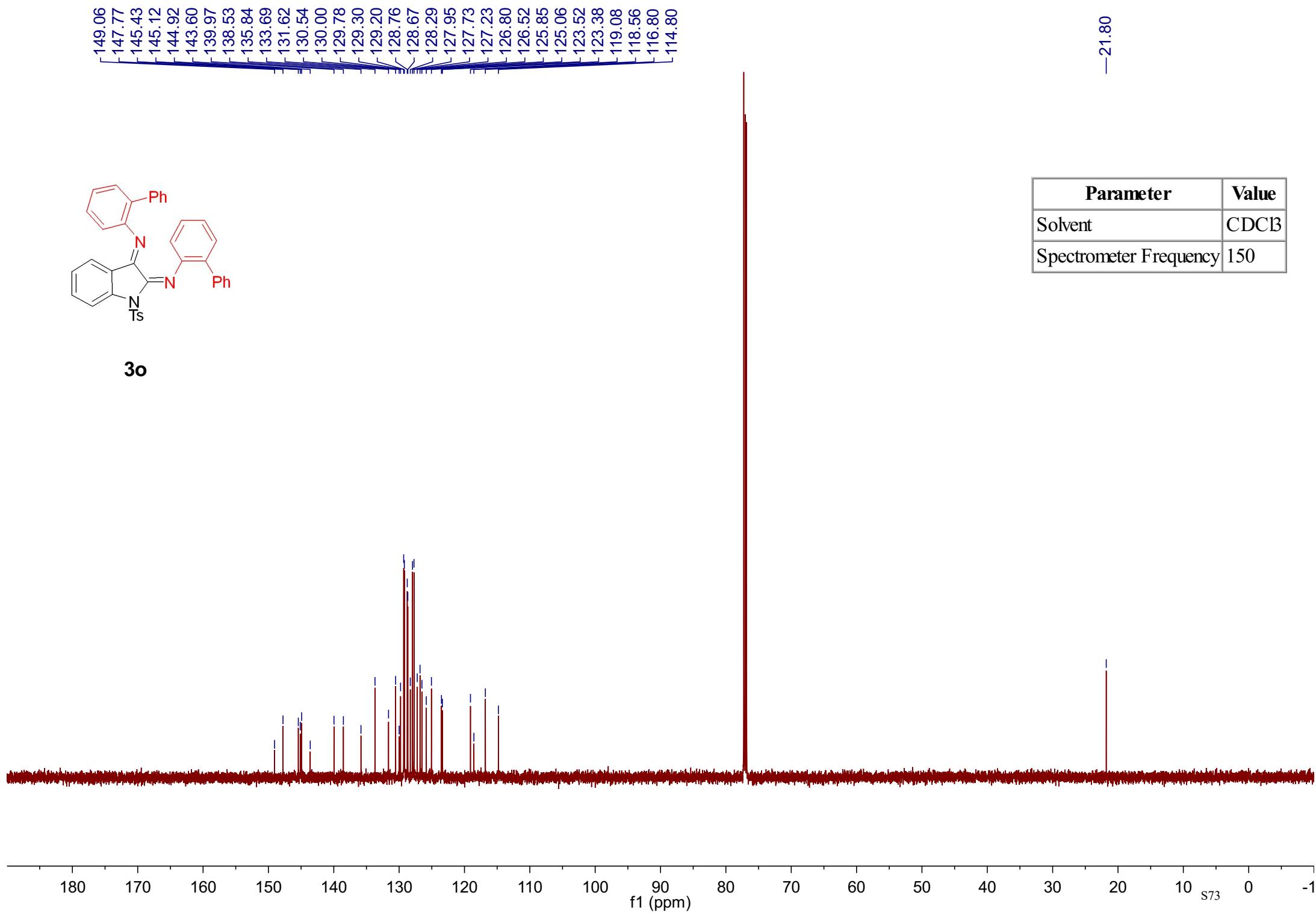


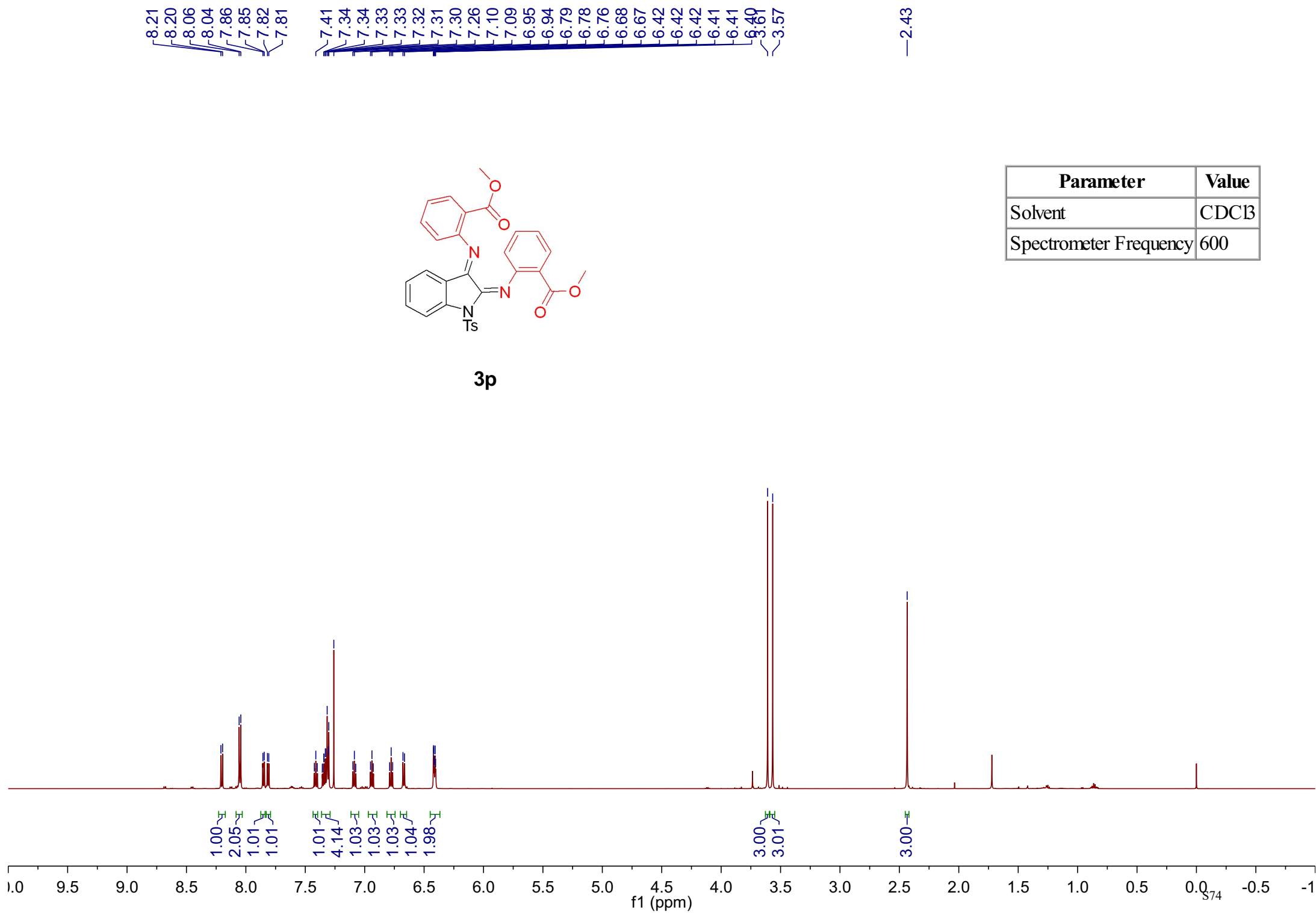


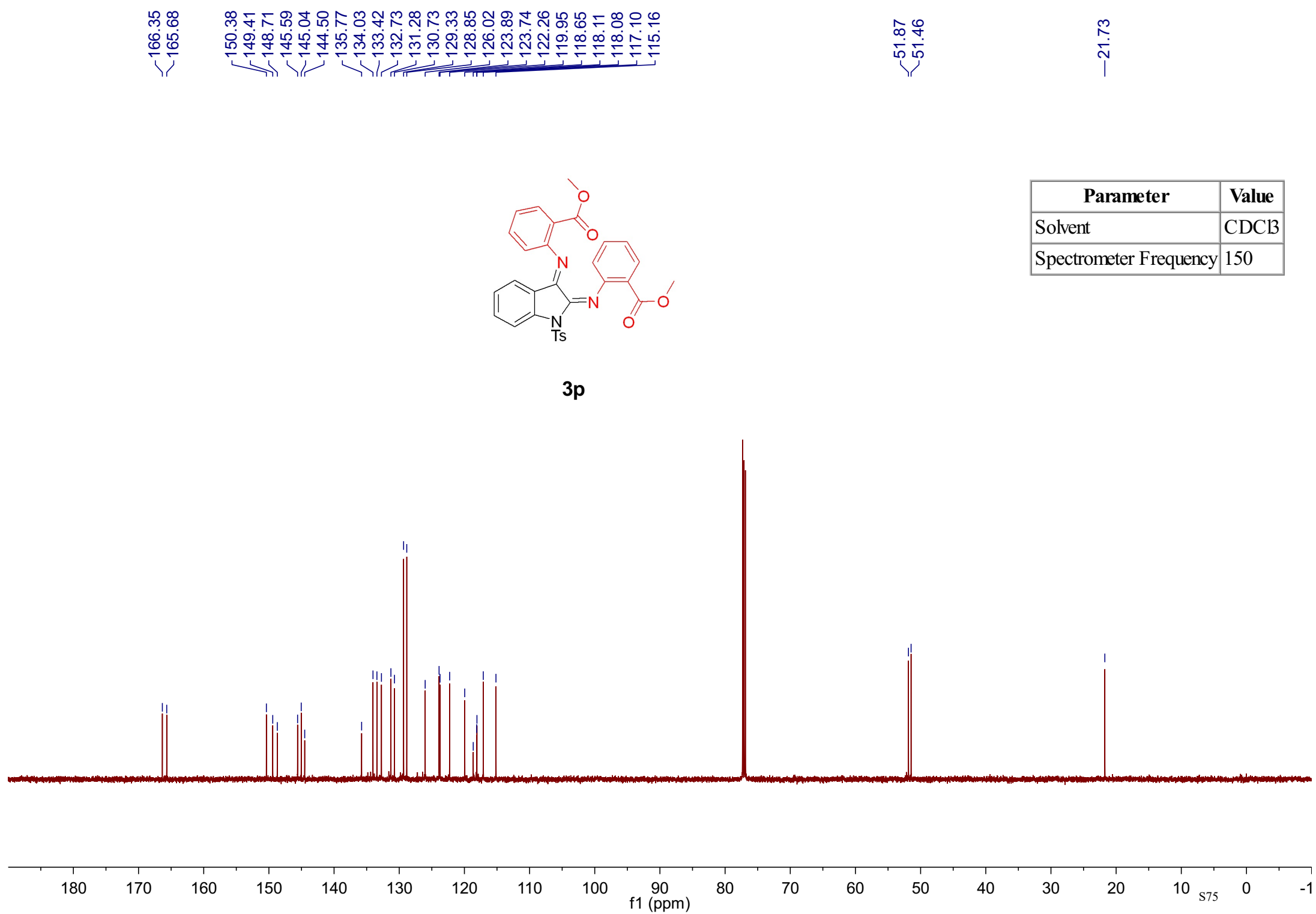


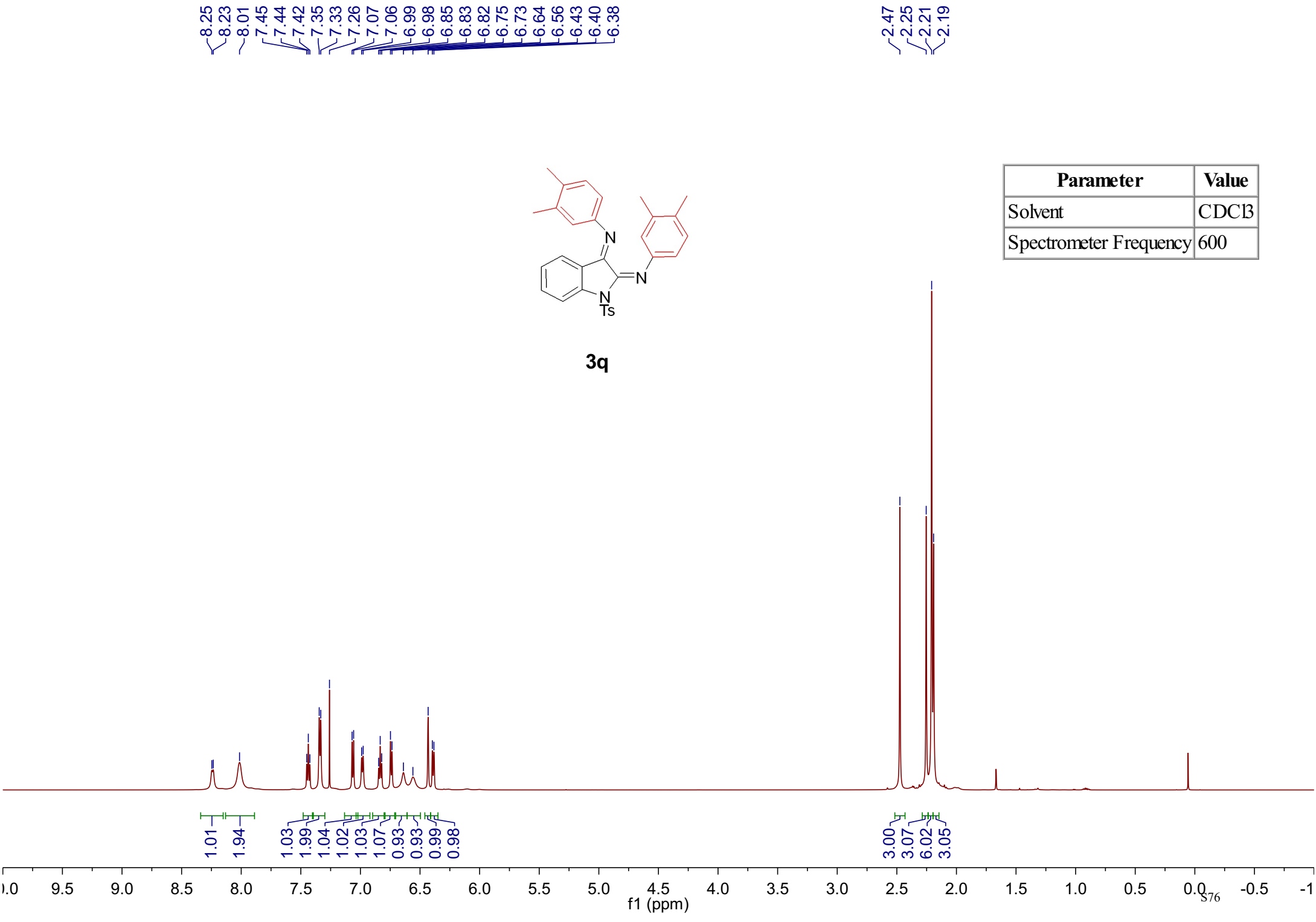
3o

Parameter	Value
Solvent	CDCI3
Spectrometer Frequency	150

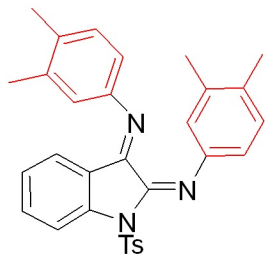








Parameter	Value
Solvent	CDCl3
Spectrometer Frequency	600

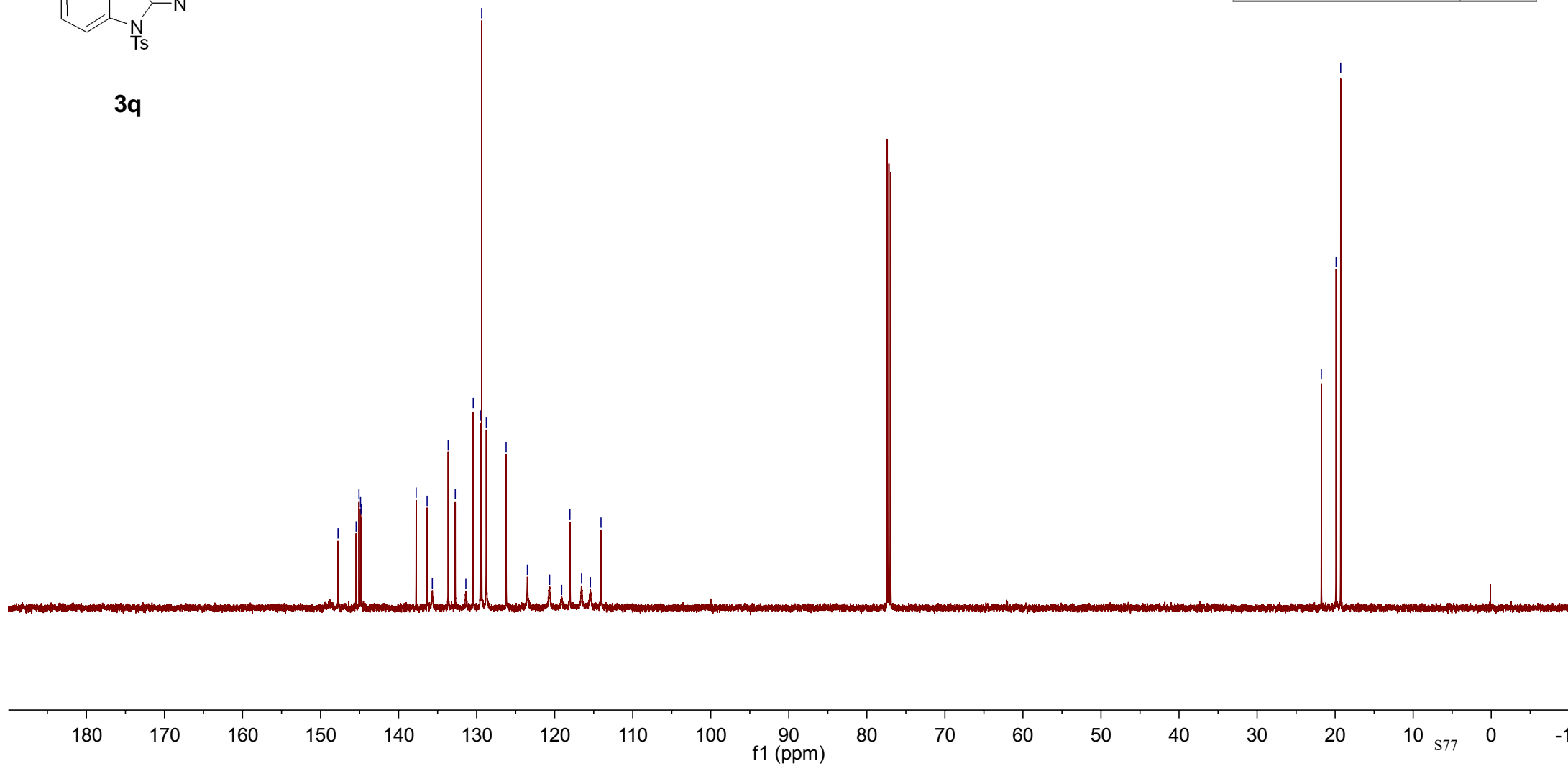


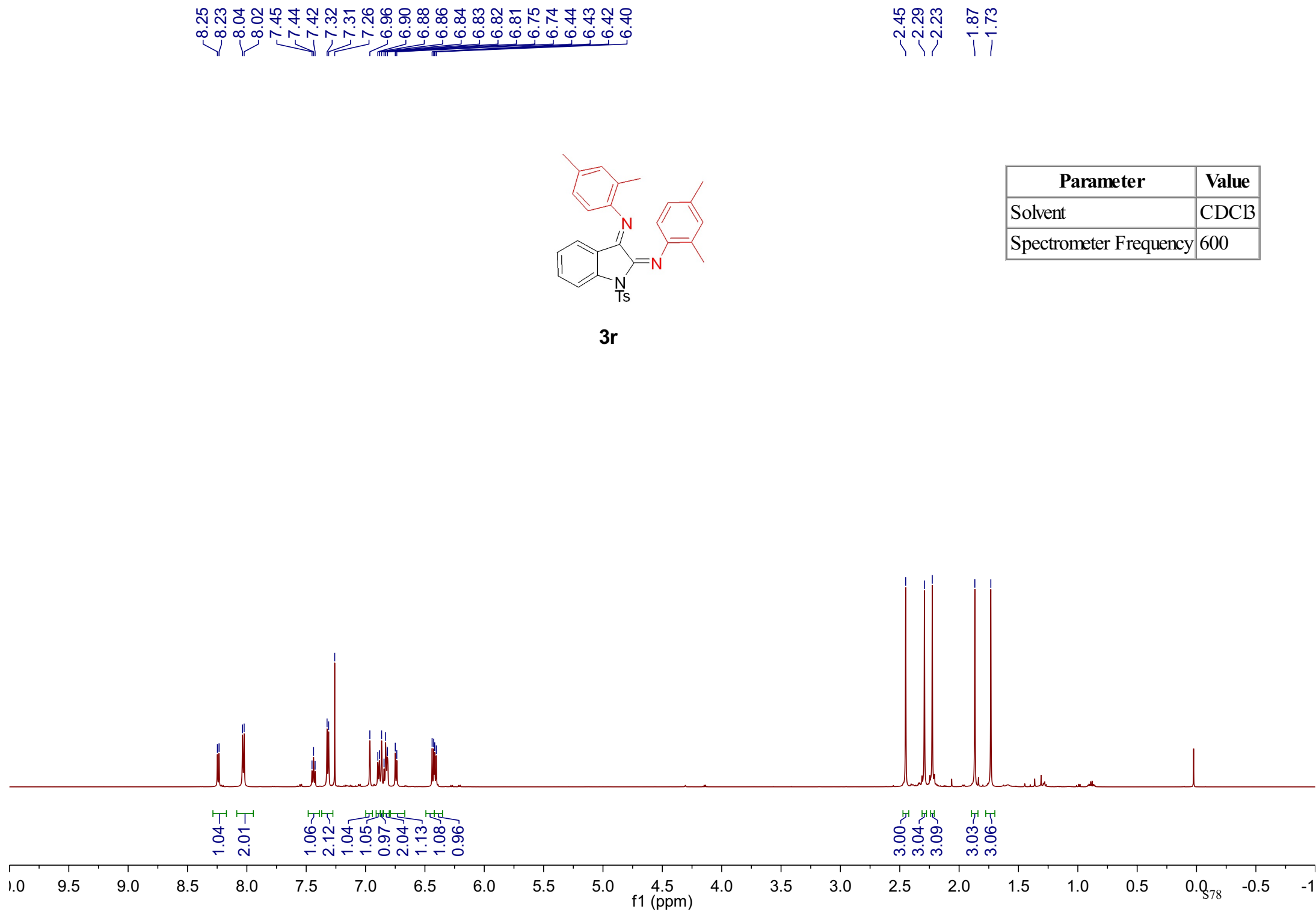
3q

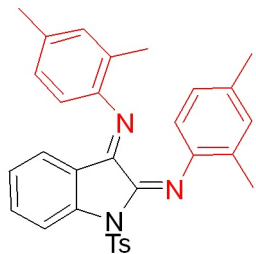
147.76
145.44
145.08
144.88
144.82
137.75
136.35
135.68
133.65
132.74
131.38
130.44
129.53
129.36
128.76
126.22
123.50
120.65
119.10
118.05
116.56
115.42
114.06

21.77
19.88
19.29

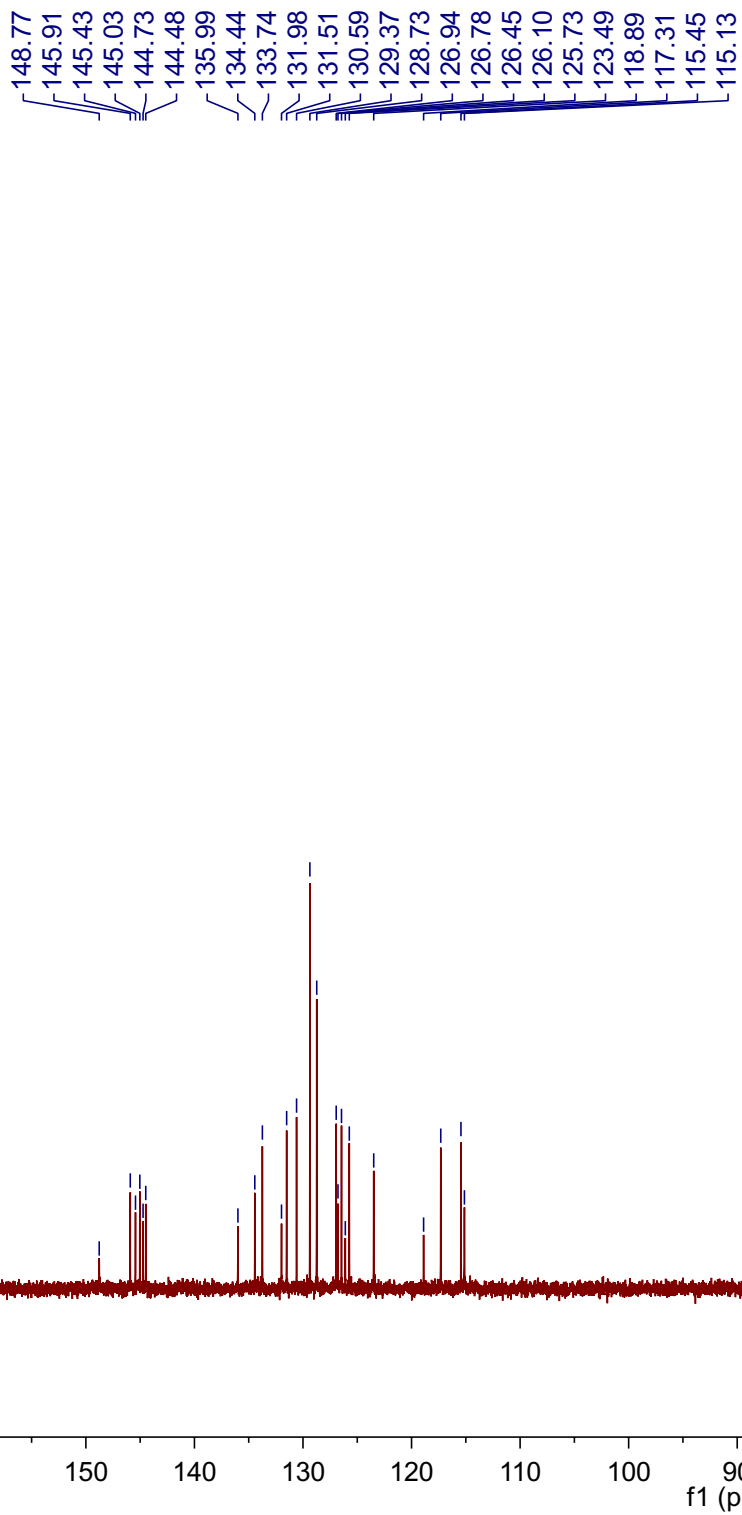
Parameter	Value
Solvent	CDCI3
Spectrometer Frequency	150





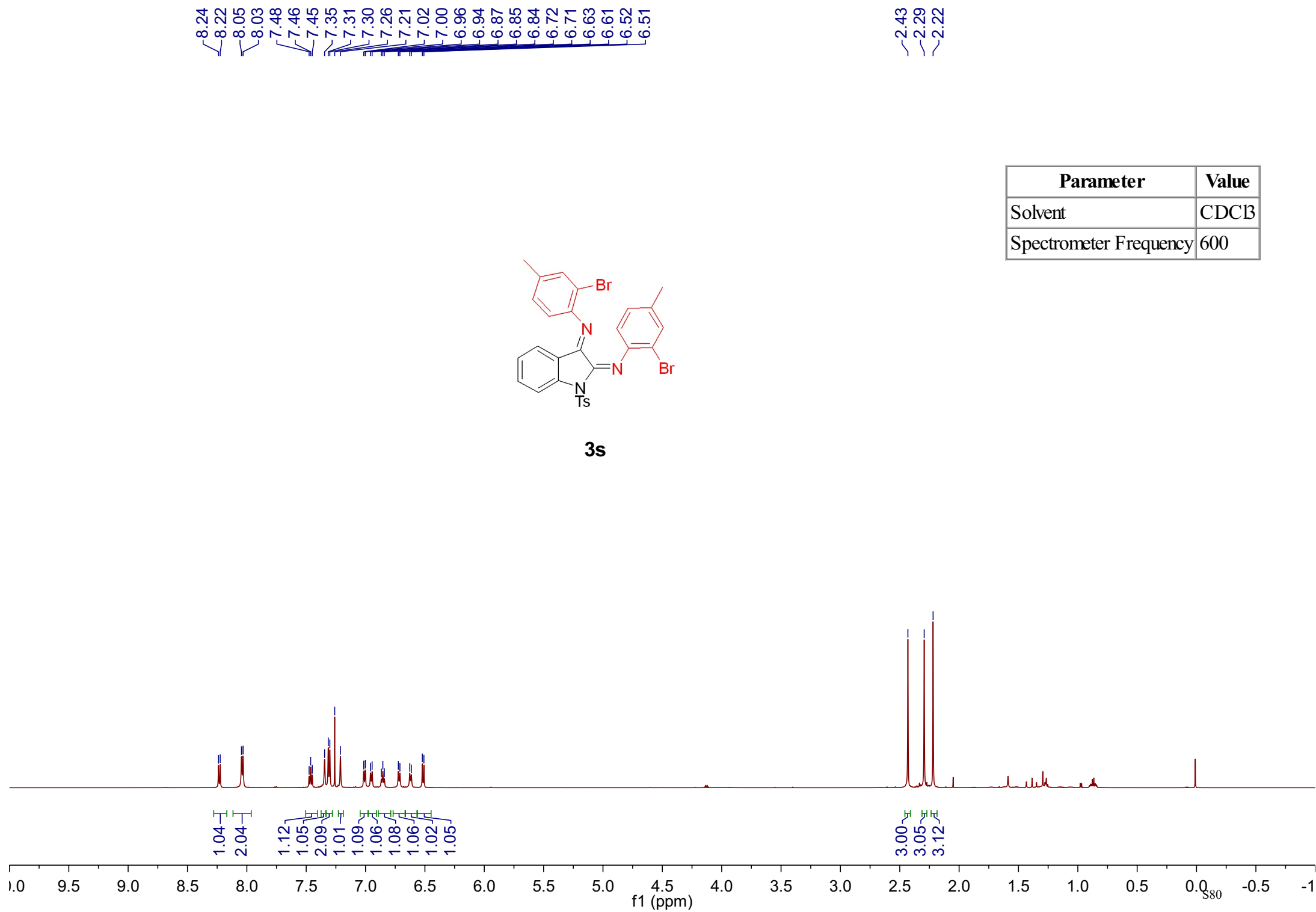


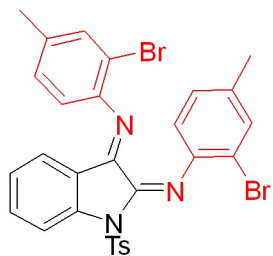
3r



21.72
20.87
20.81
17.51

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150



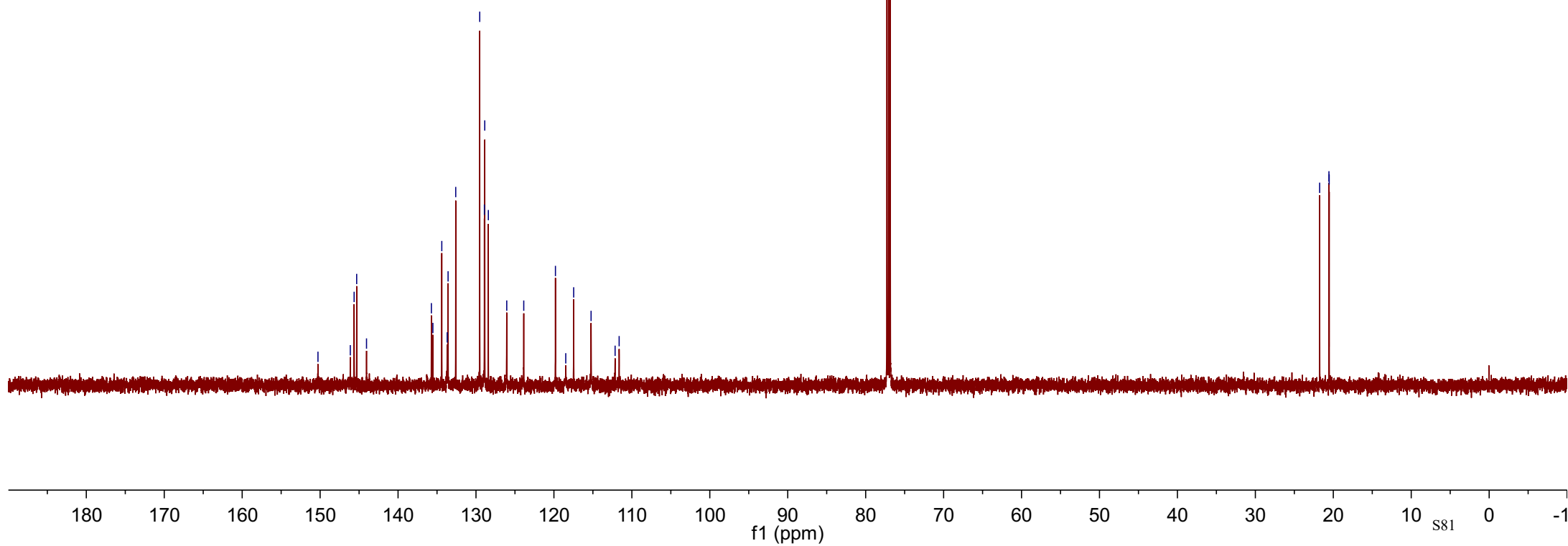


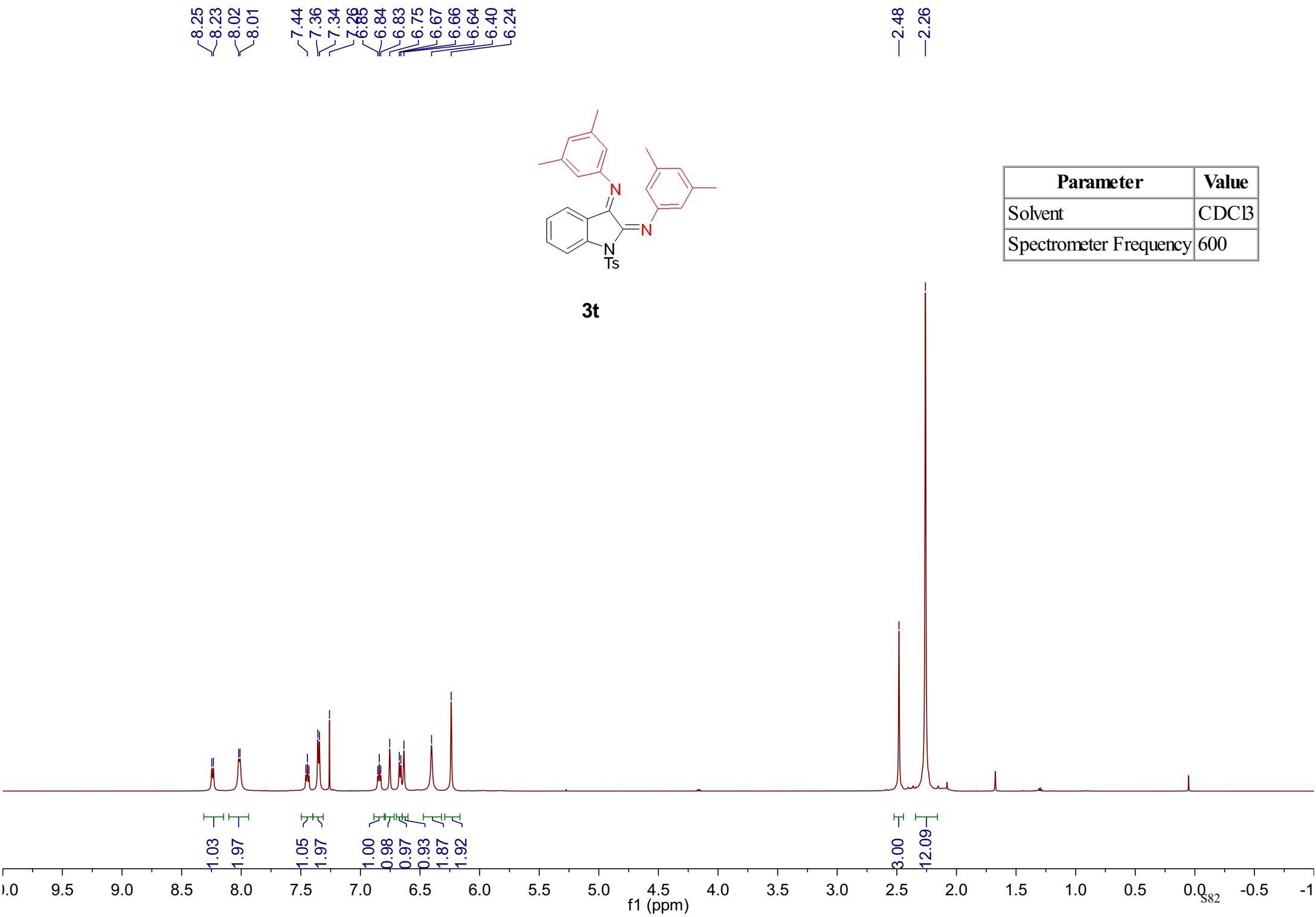
3s

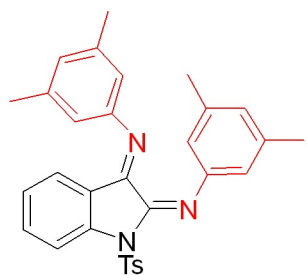
150.28
146.13
145.64
145.31
144.05
135.72
135.53
134.39
133.69
133.58
132.60
129.52
128.92
128.88
128.41
126.05
123.87
119.81
118.48
117.47
115.23
112.15
111.63

21.75
20.56
20.53

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150





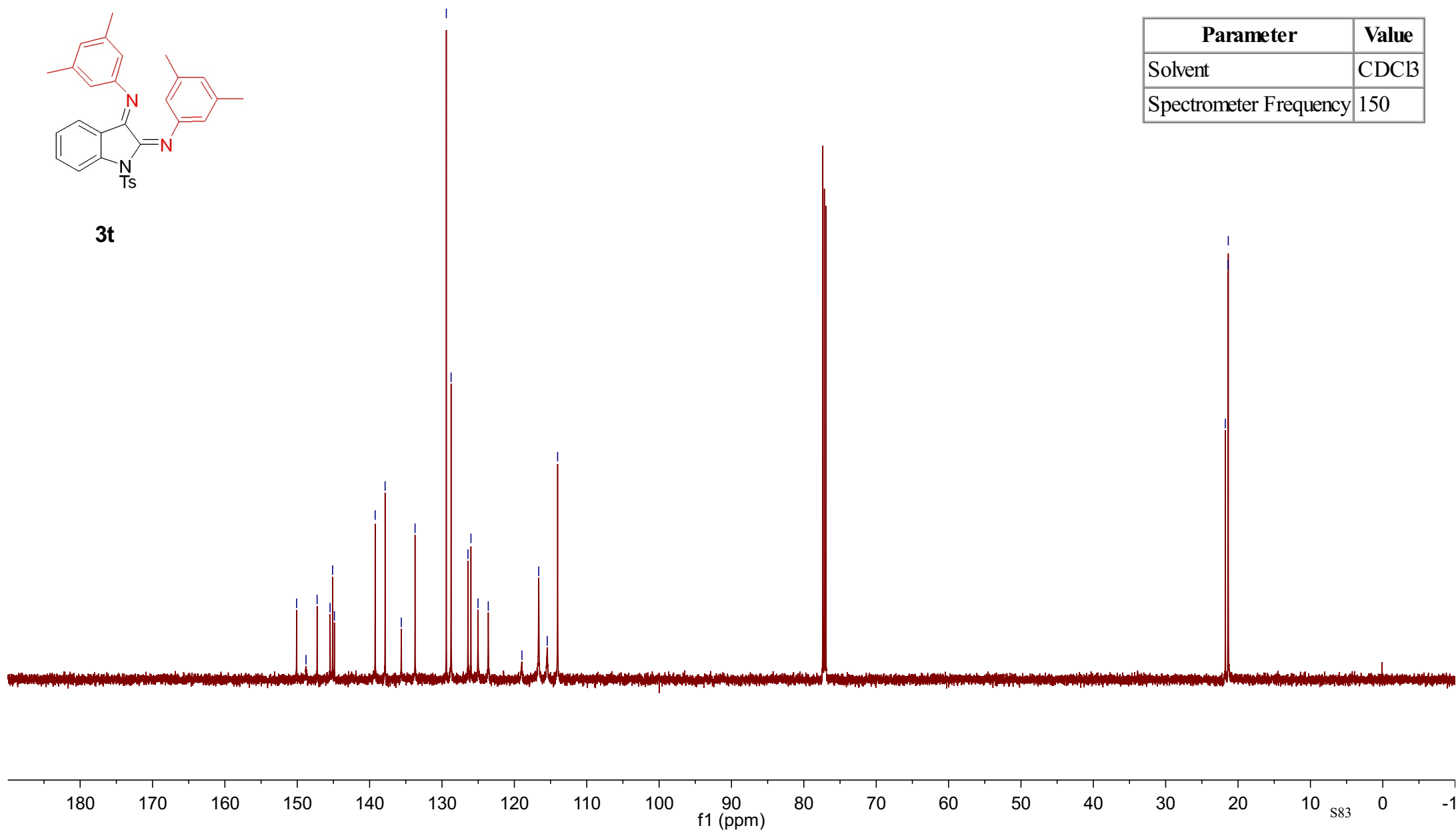


3t

150.09
148.79
147.26
145.46
145.12
144.86
139.23
137.86
135.63
133.72
129.40
128.74
126.41
126.00
125.02
123.61
118.95
116.63
115.46
114.02

21.76
21.40
21.35

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150



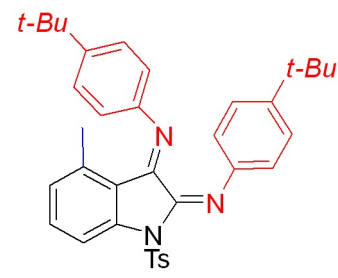
7.93
7.92
7.89
7.87

7.40
7.26
7.22
7.21
7.01
7.00
6.27
6.23
6.22

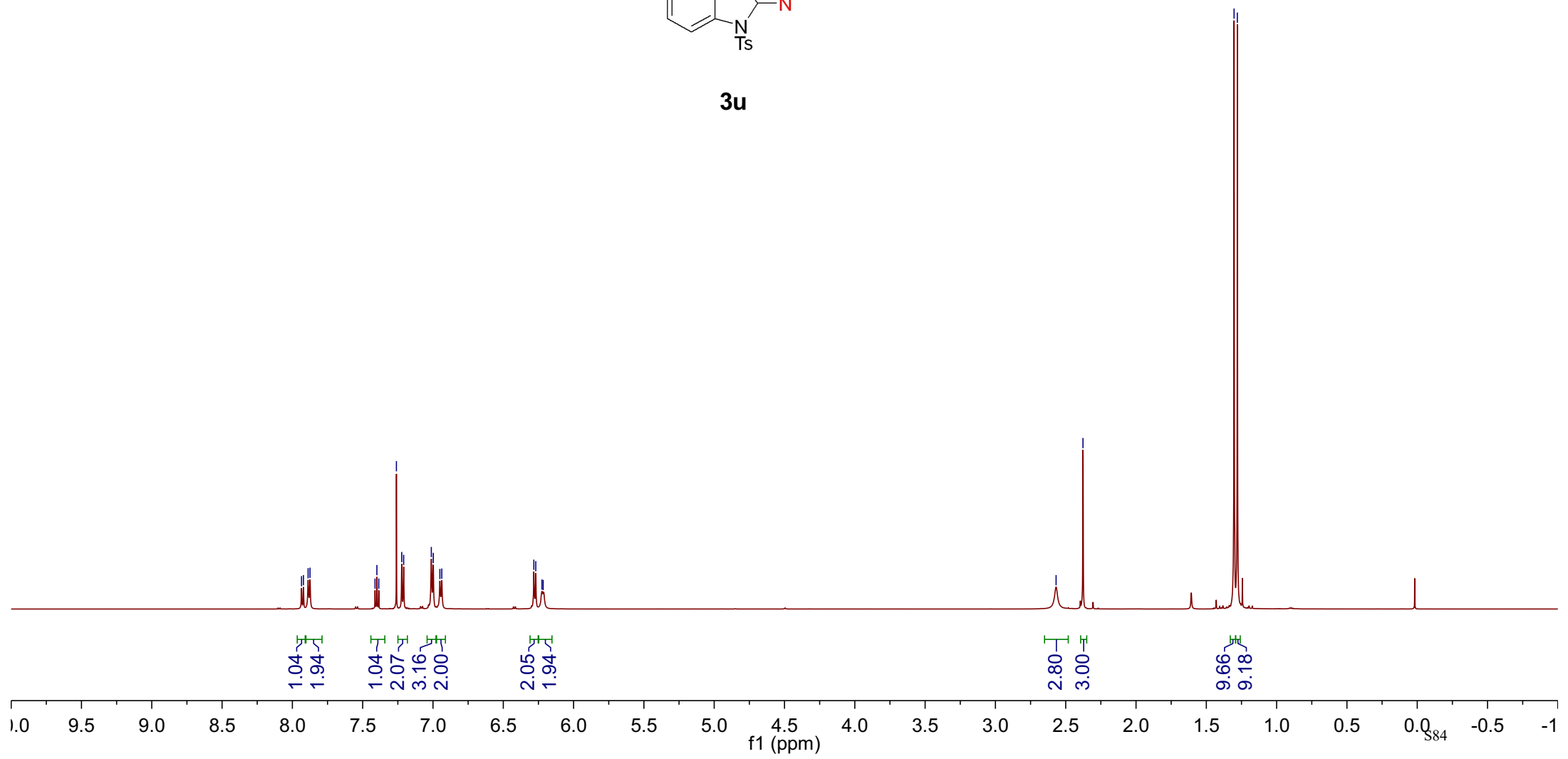
2.57
2.38

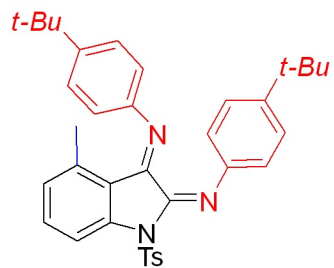
1.30
1.28

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	600



3u



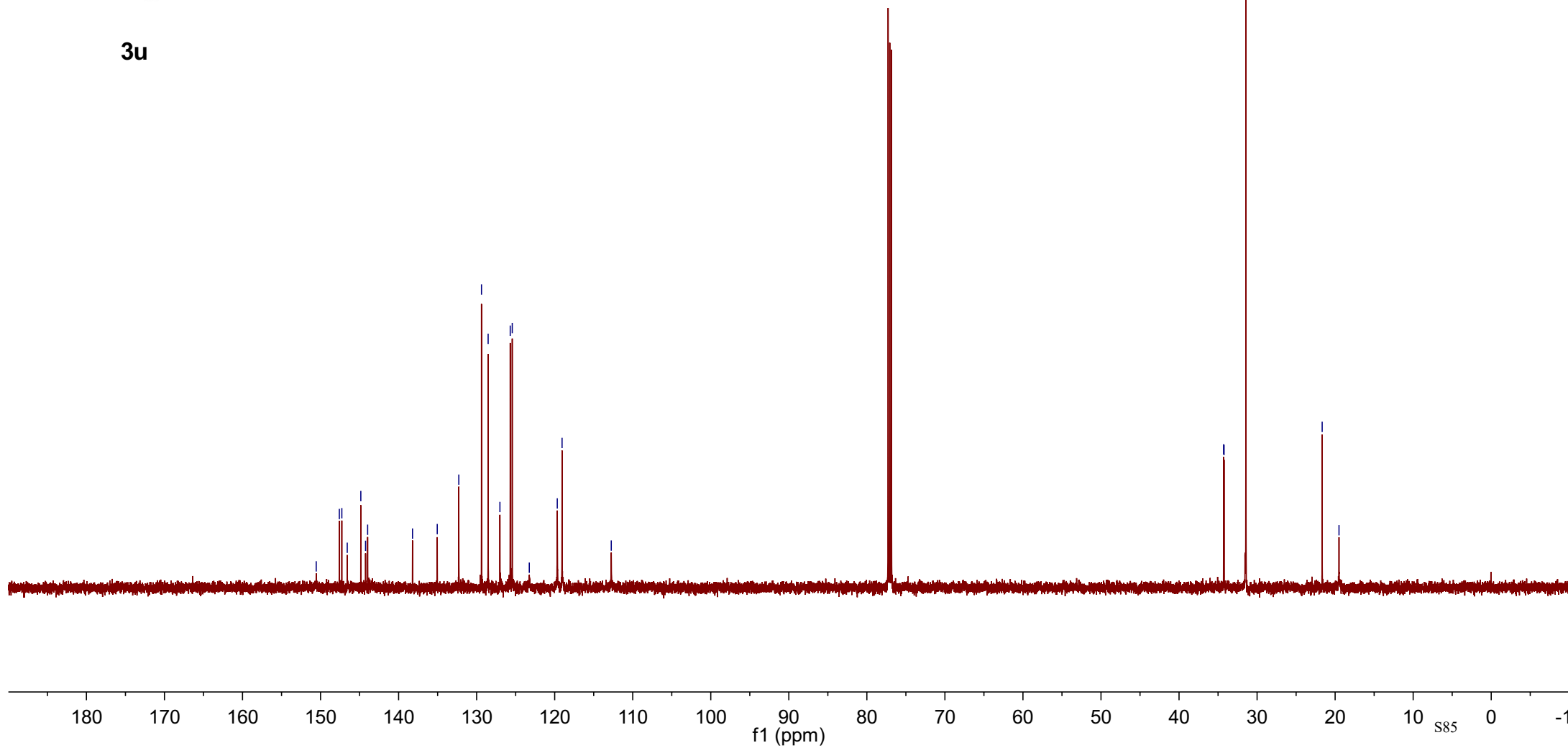


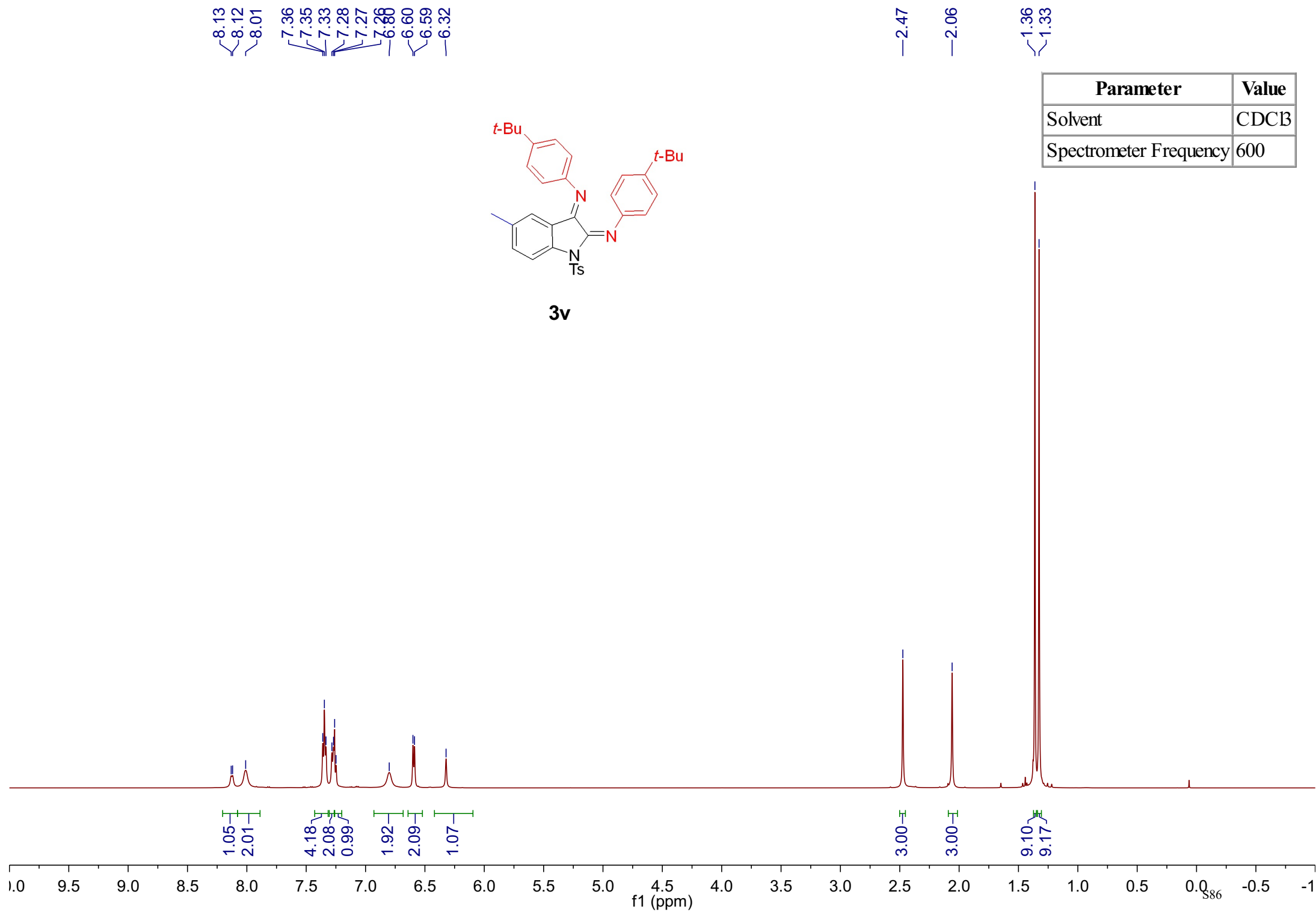
3u

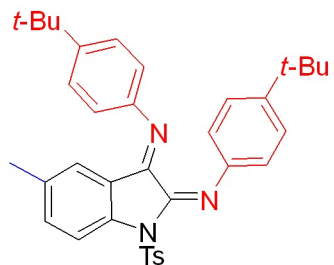
150.56
147.60
147.27
146.58
144.84
144.23
143.97
135.05
132.28
129.38
128.53
127.02
125.70
125.44
119.67
119.95

34.29
34.23
31.43
31.43
21.66
19.50

Parameter	Value
Solvent	CDCB
Spectrometer Frequency	150





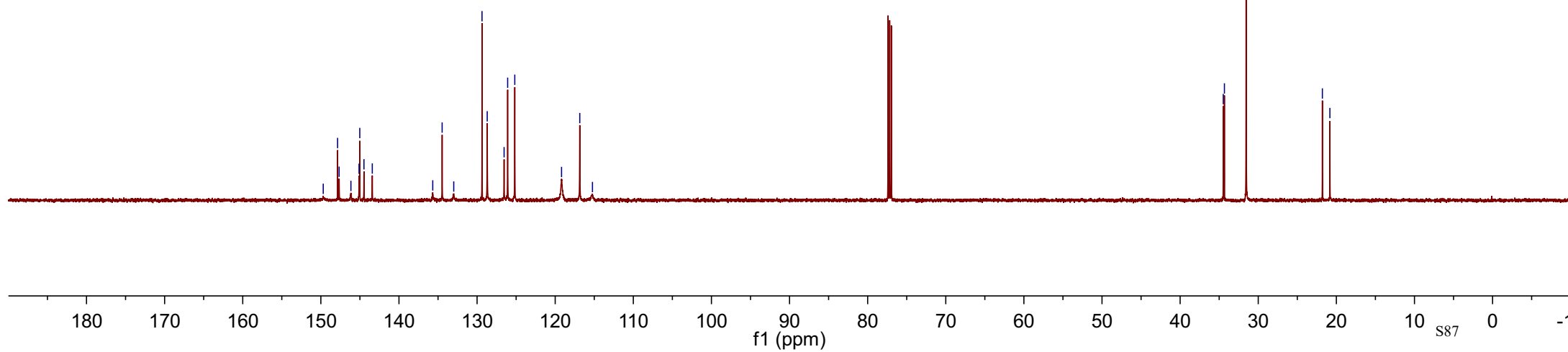


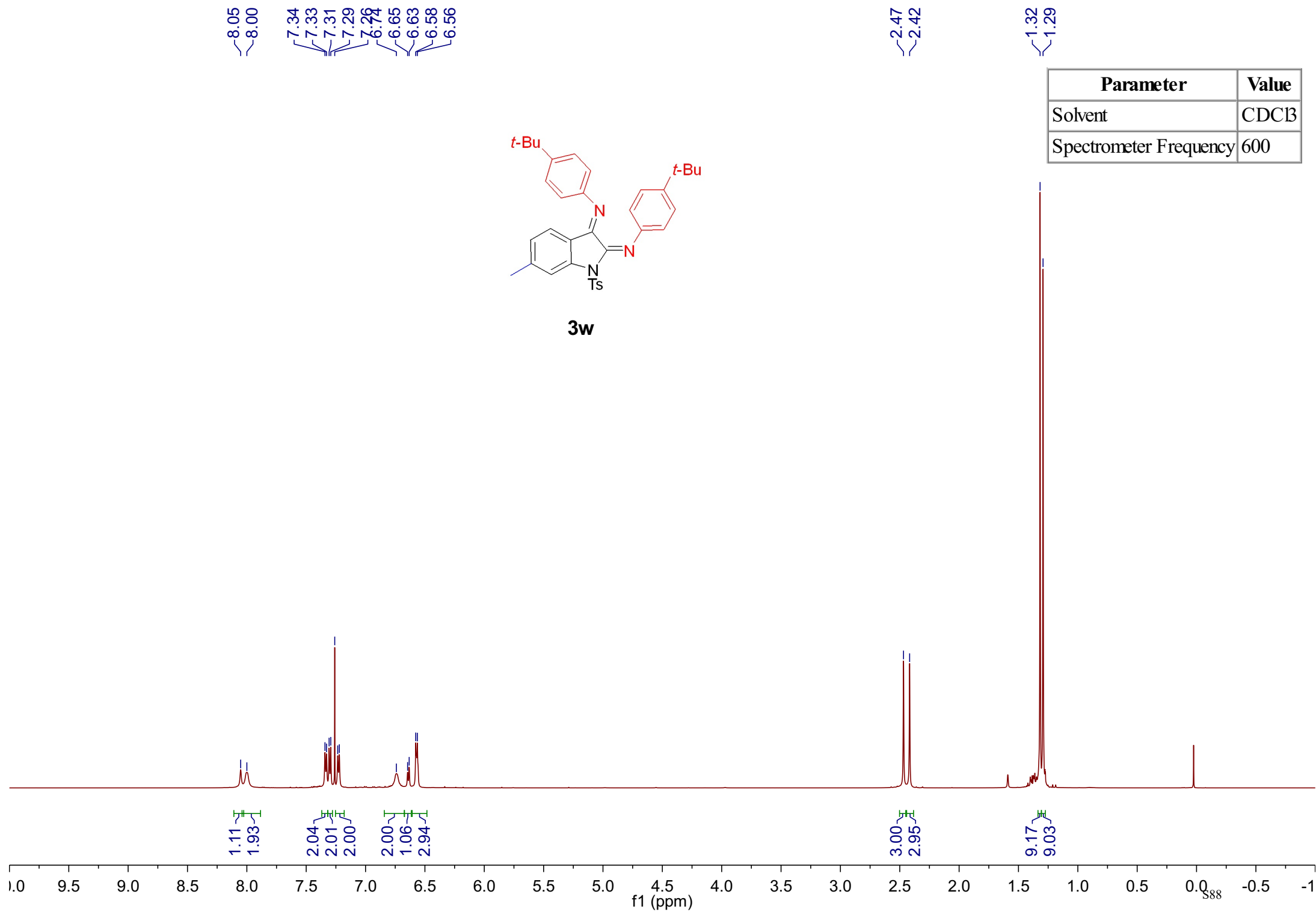
3v

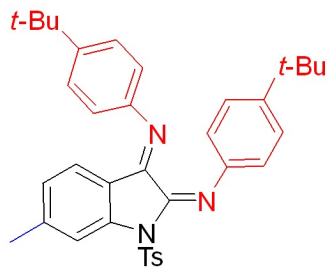
149.70
147.86
147.66
146.15
145.11
145.02
144.48
143.42
134.47
129.37
128.70
126.54
126.09
125.19
119.19
116.85
115.23

34.48
34.33
31.55
21.79
20.83

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150





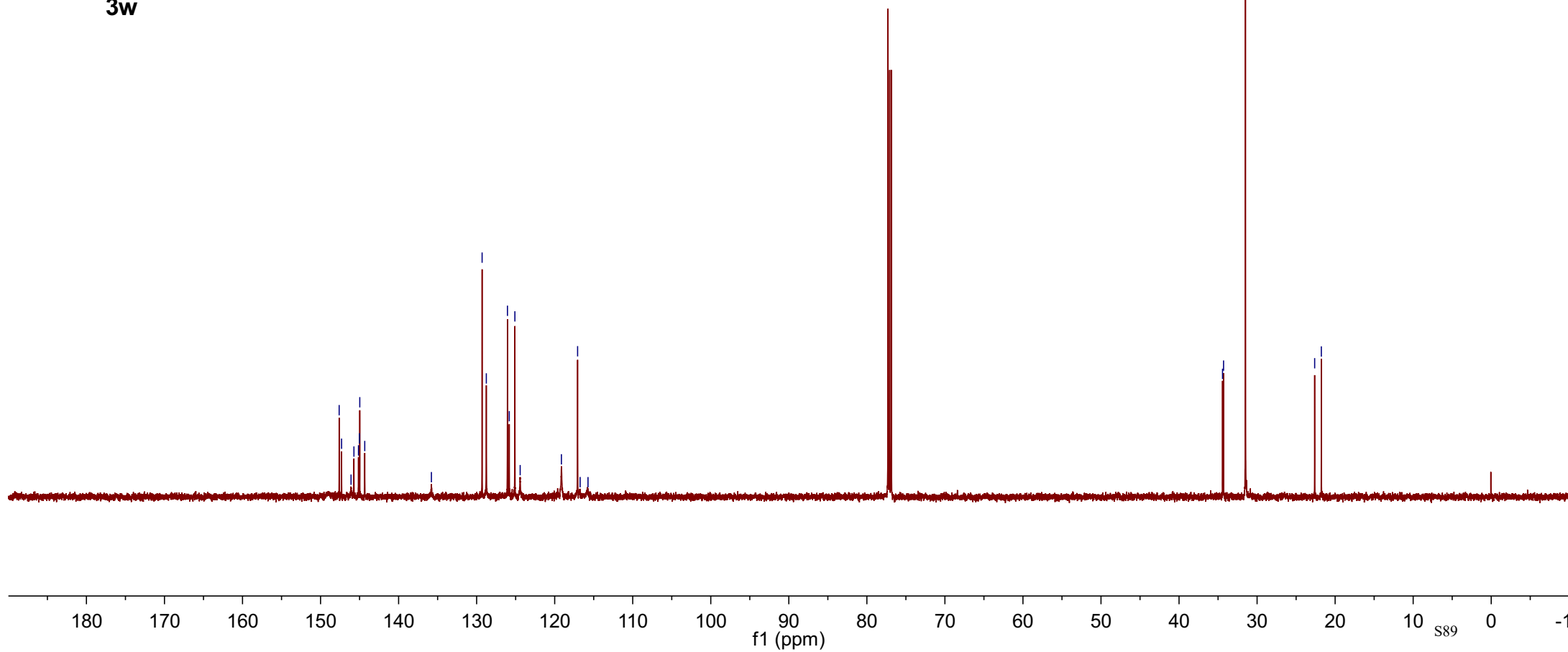


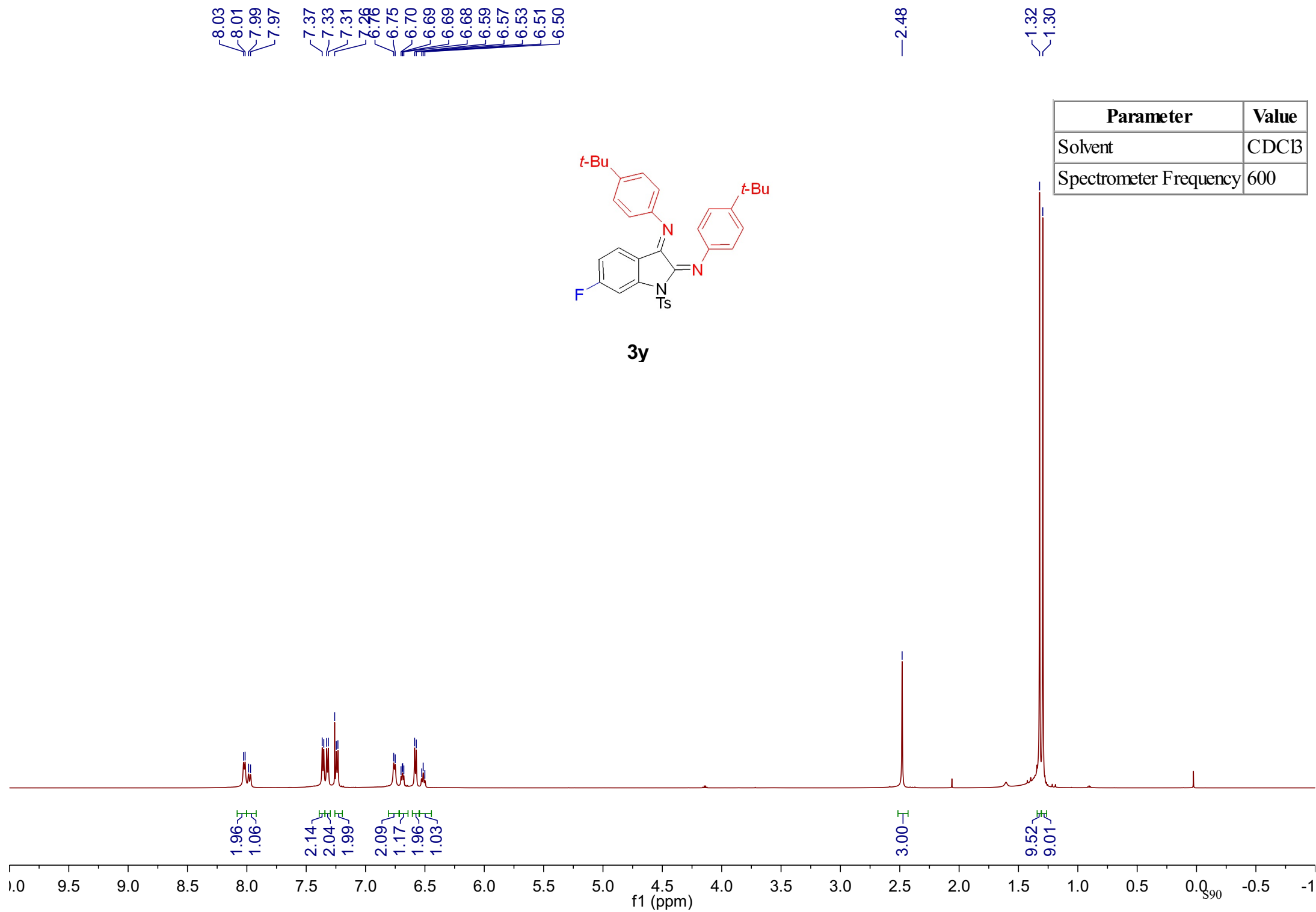
3w

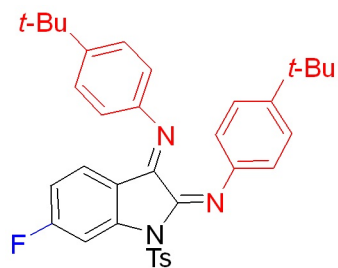
147.62
147.31
146.09
145.72
145.14
145.02
144.97
144.34
— 135.80
129.30
128.76
126.05
125.82
125.10
124.42
119.14
117.06
116.74
115.73

34.43
34.29
31.49
31.48
22.62
21.77

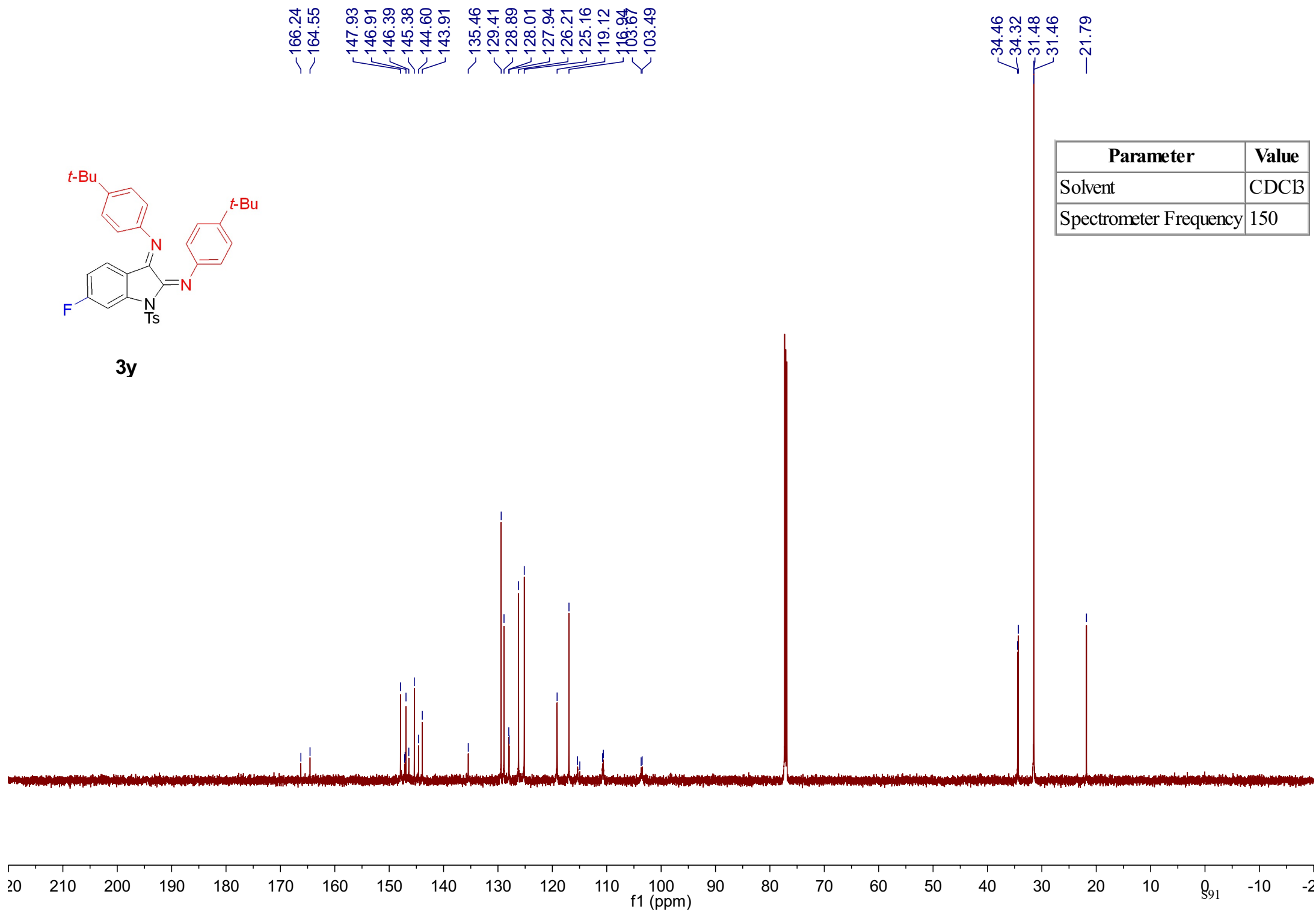
Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150



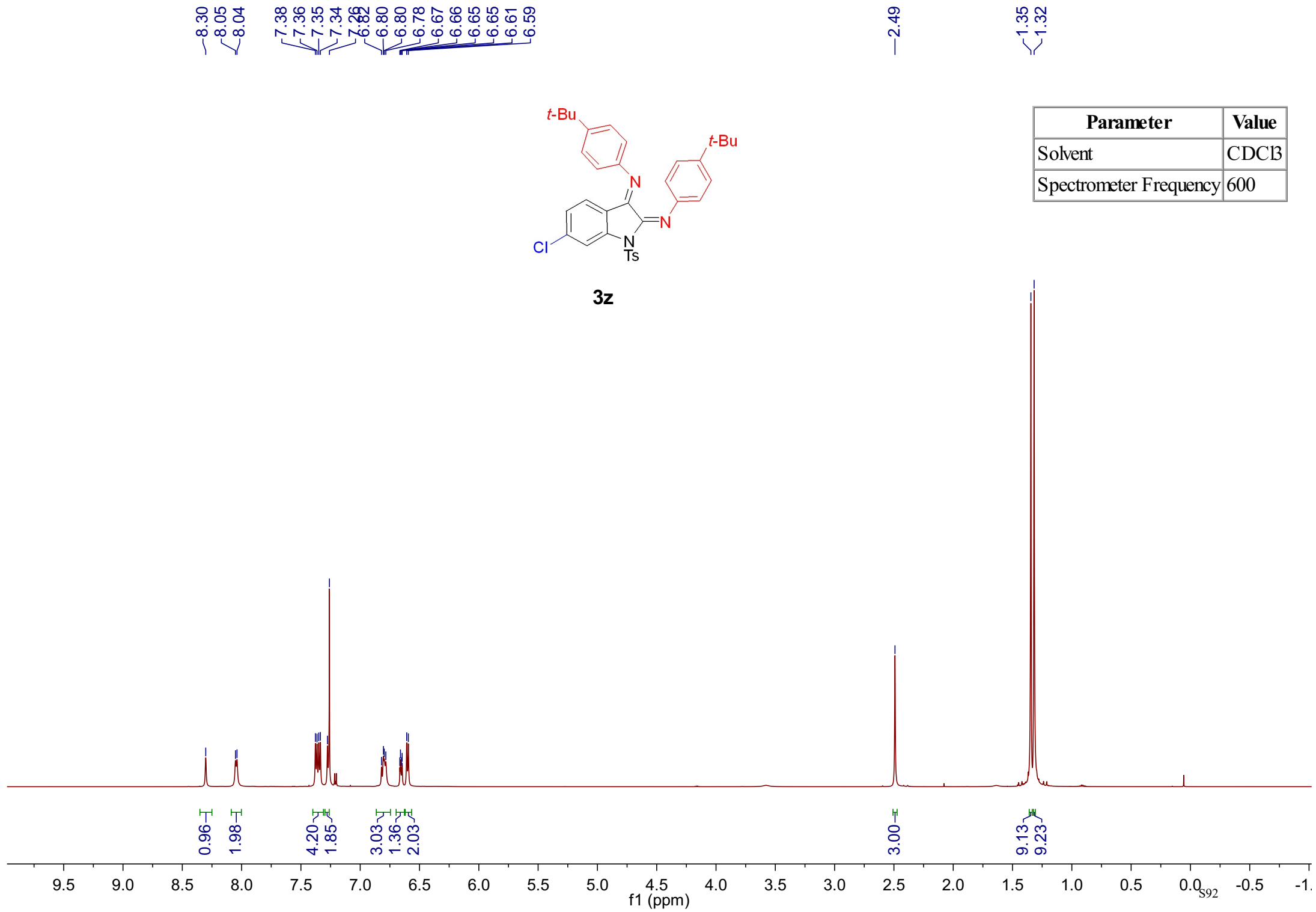




3y

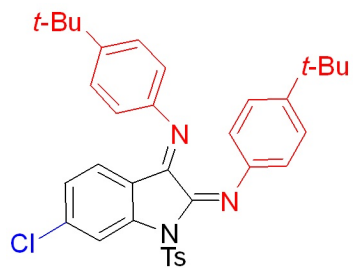


Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150



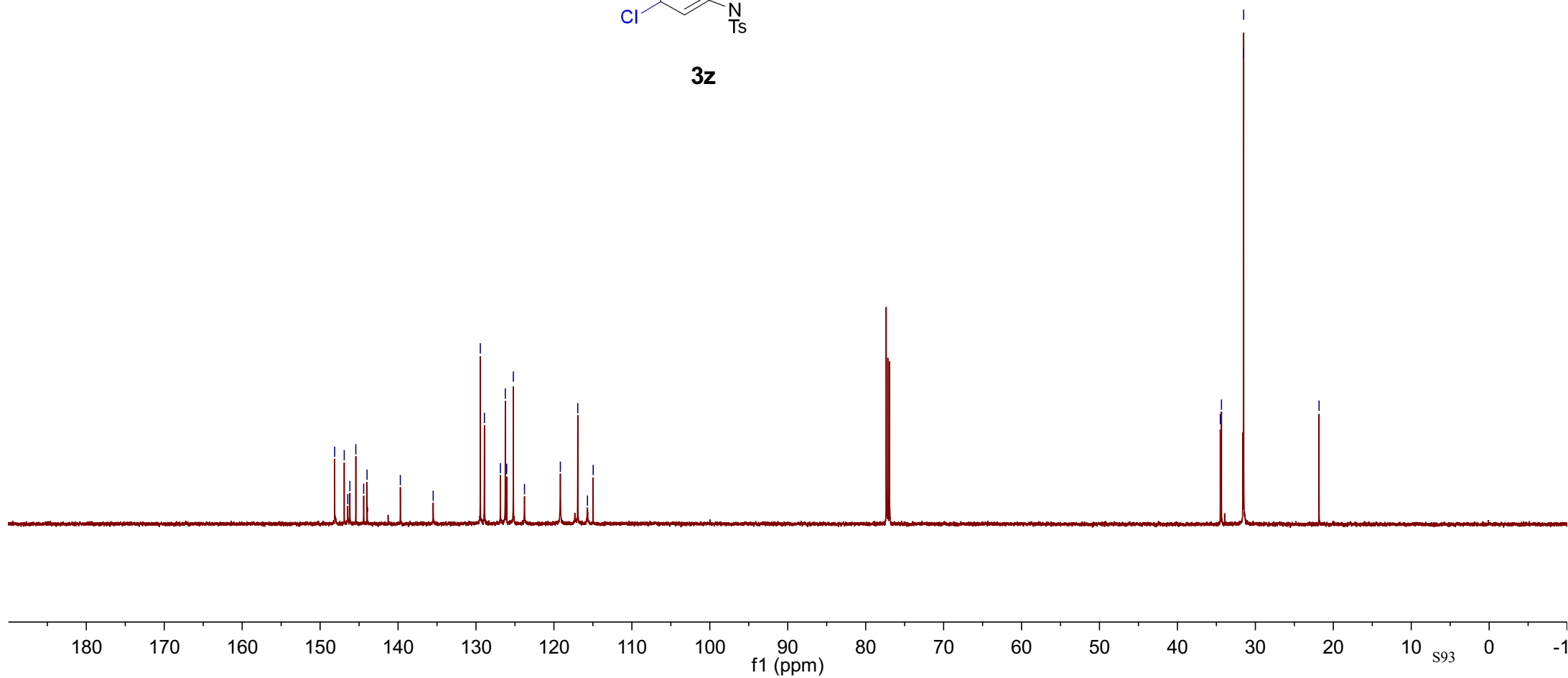
148.13
146.91
146.45
146.18
145.42
144.41
143.98
139.71
135.49
129.44
128.92
126.87
126.24
126.06
125.20
123.78
119.18
116.94
115.69
114.96

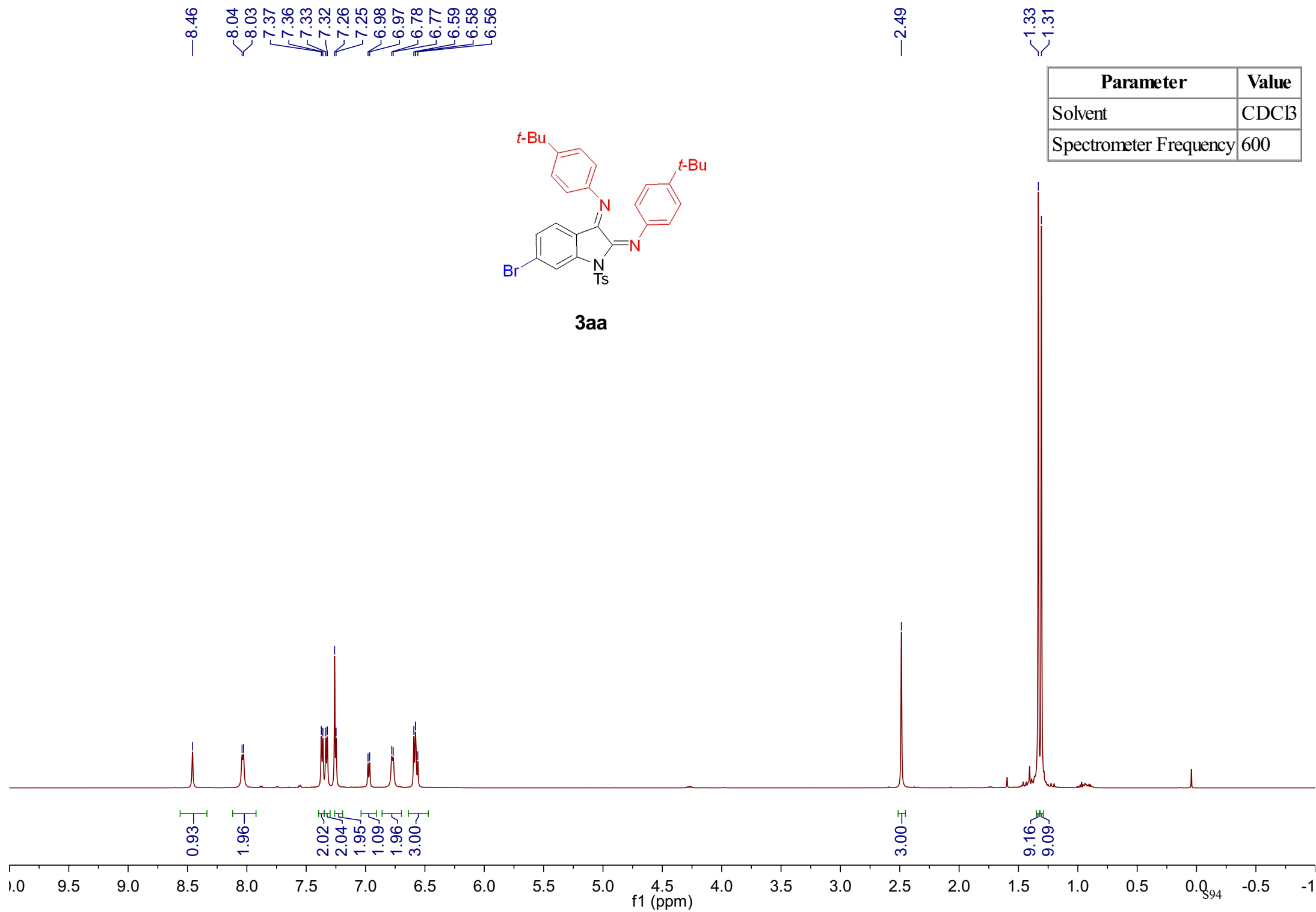
34.50
34.35
31.52
31.50
21.83

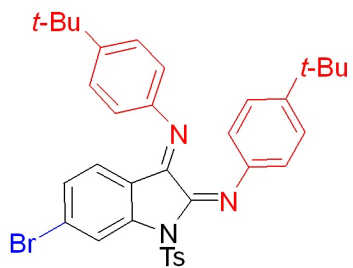


3z

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150





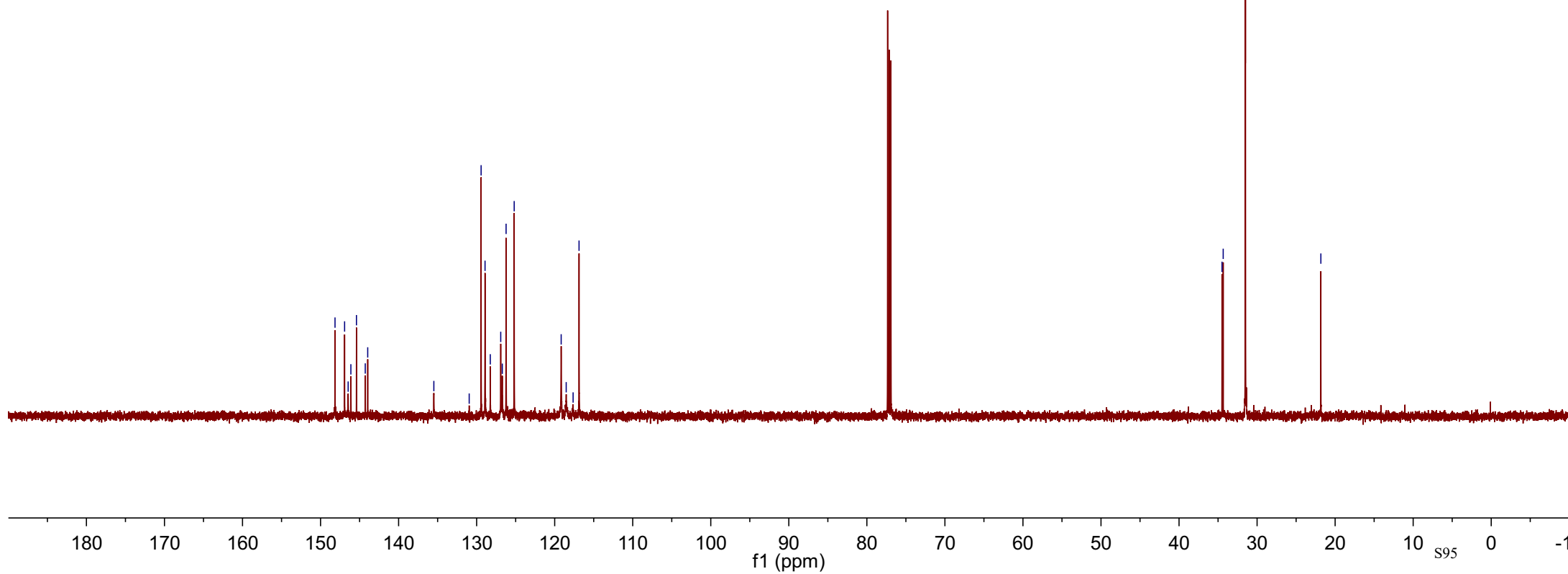


3aa

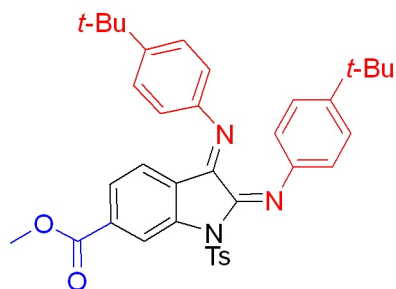
148.15
146.92
146.46
146.12
145.40
144.27
143.96
129.43
128.92
128.25
126.91
126.23
125.19
125.19
118.53
117.64
116.88

34.50
34.34
31.51
31.49
21.83

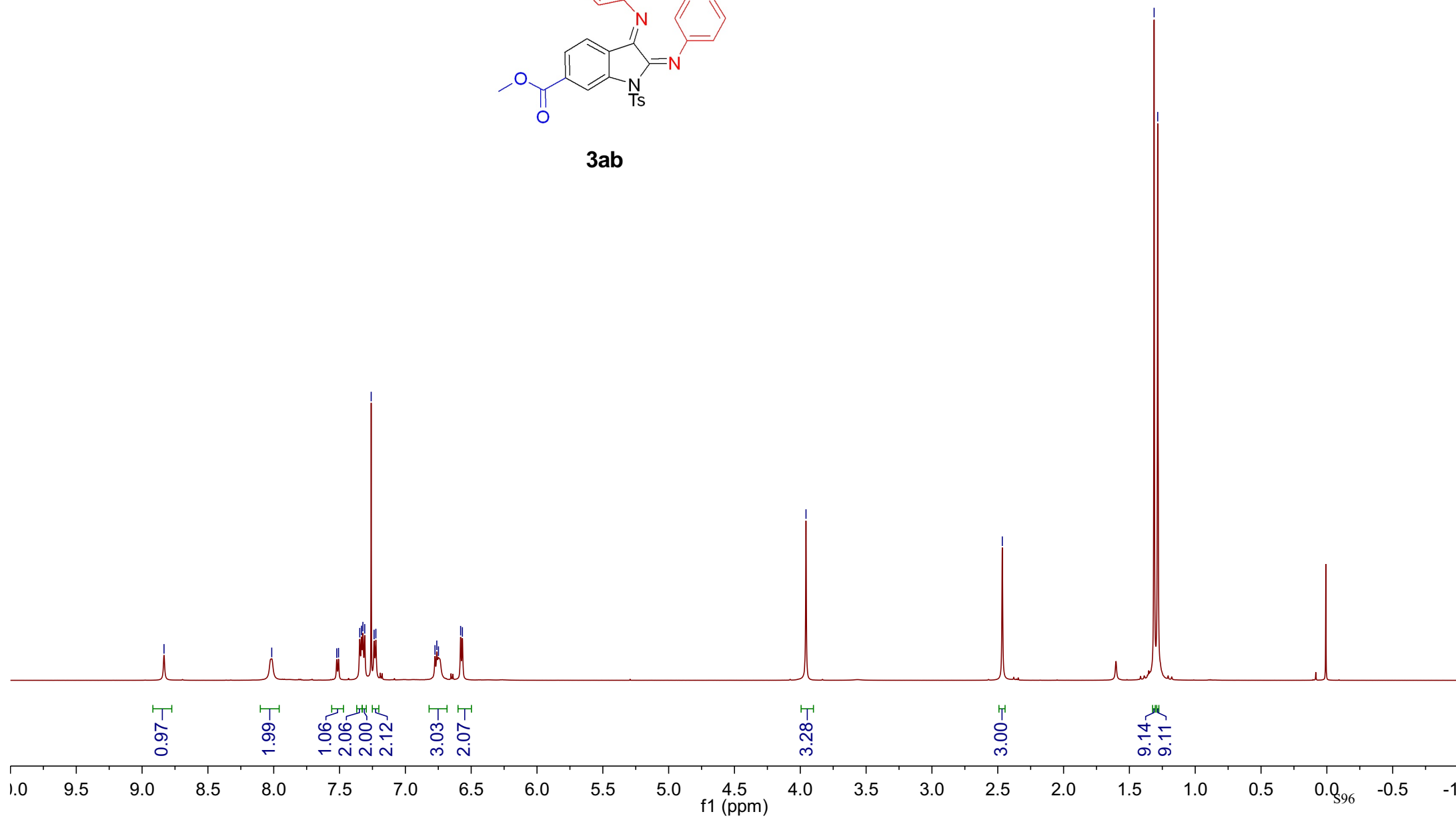
Parameter	Value
Solvent	CDCB
Spectrometer Frequency	150

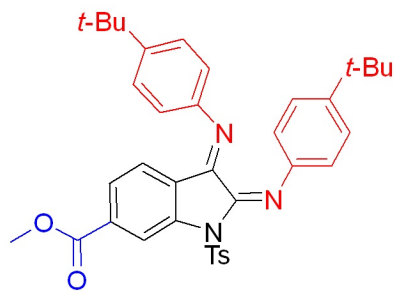


Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	600

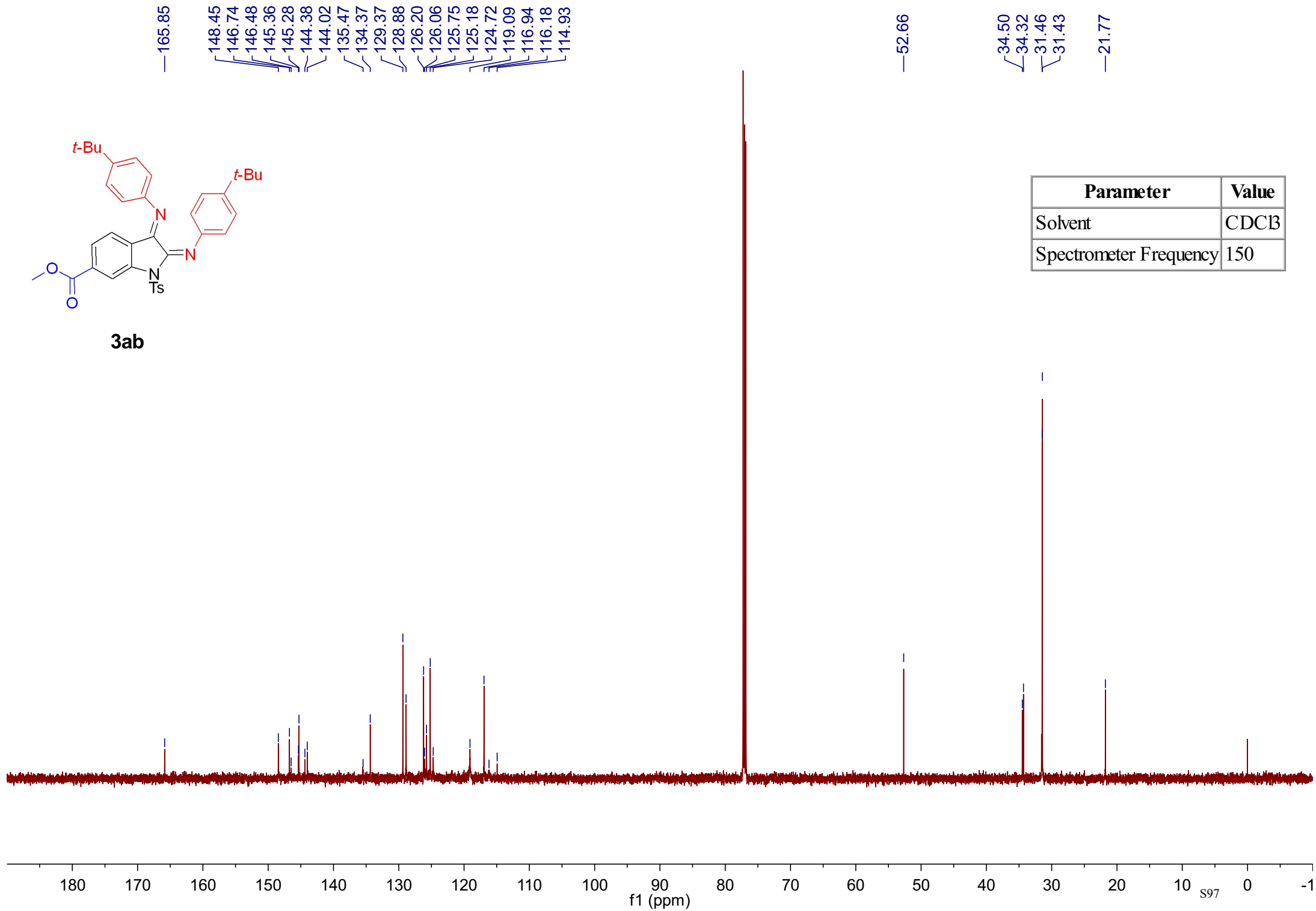


3ab

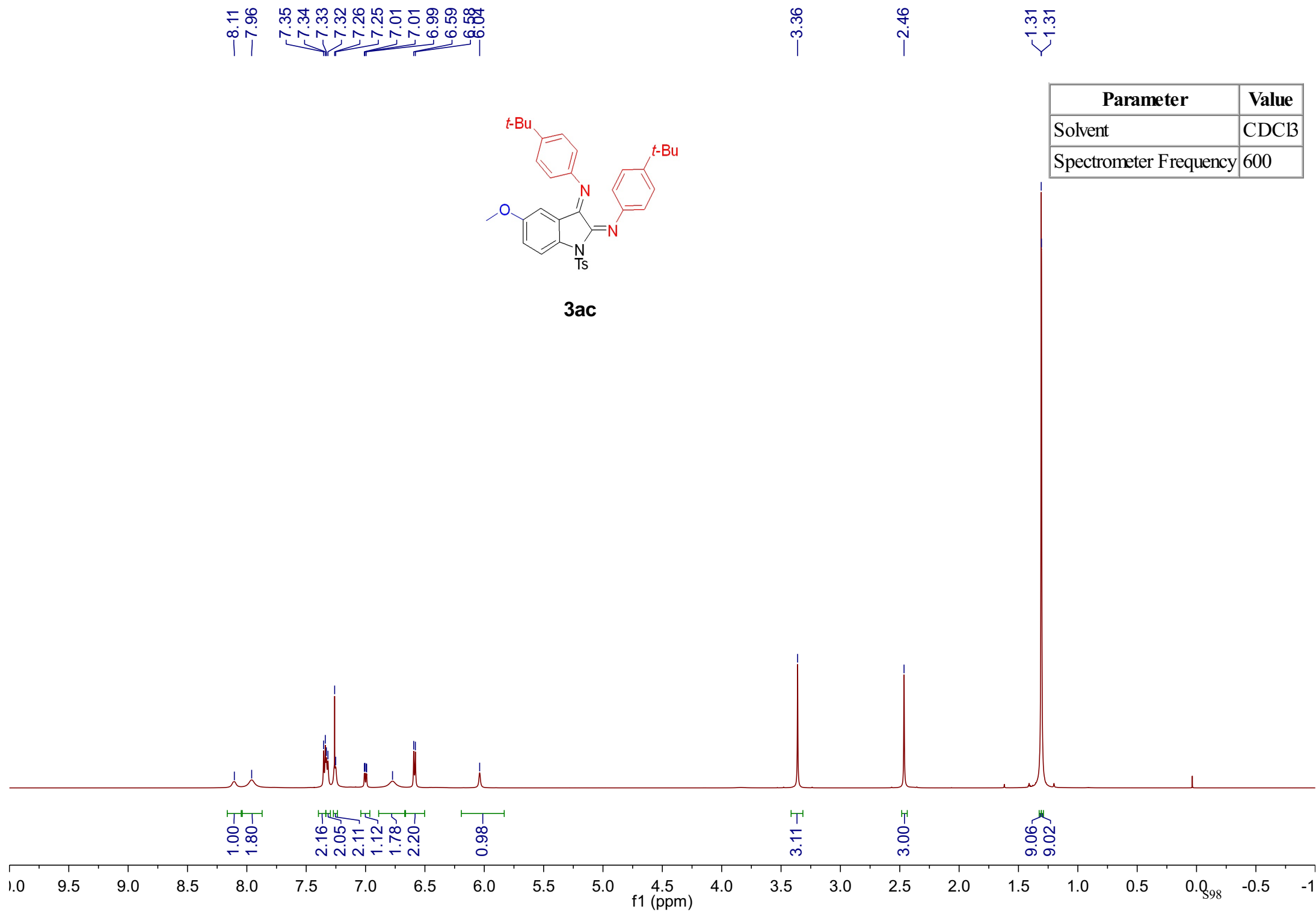


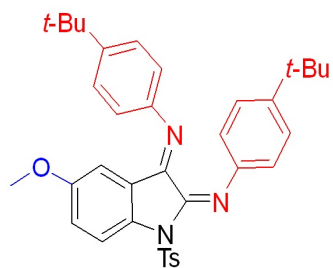


3ab



Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150





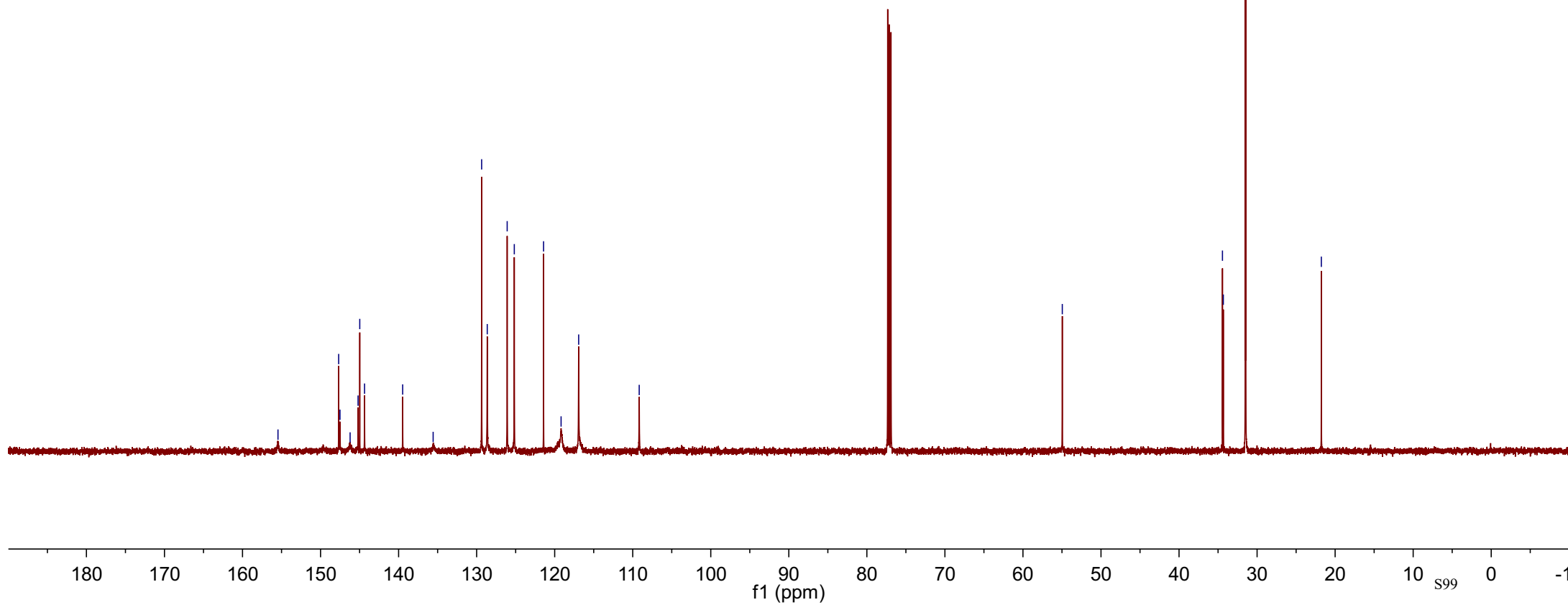
3ac

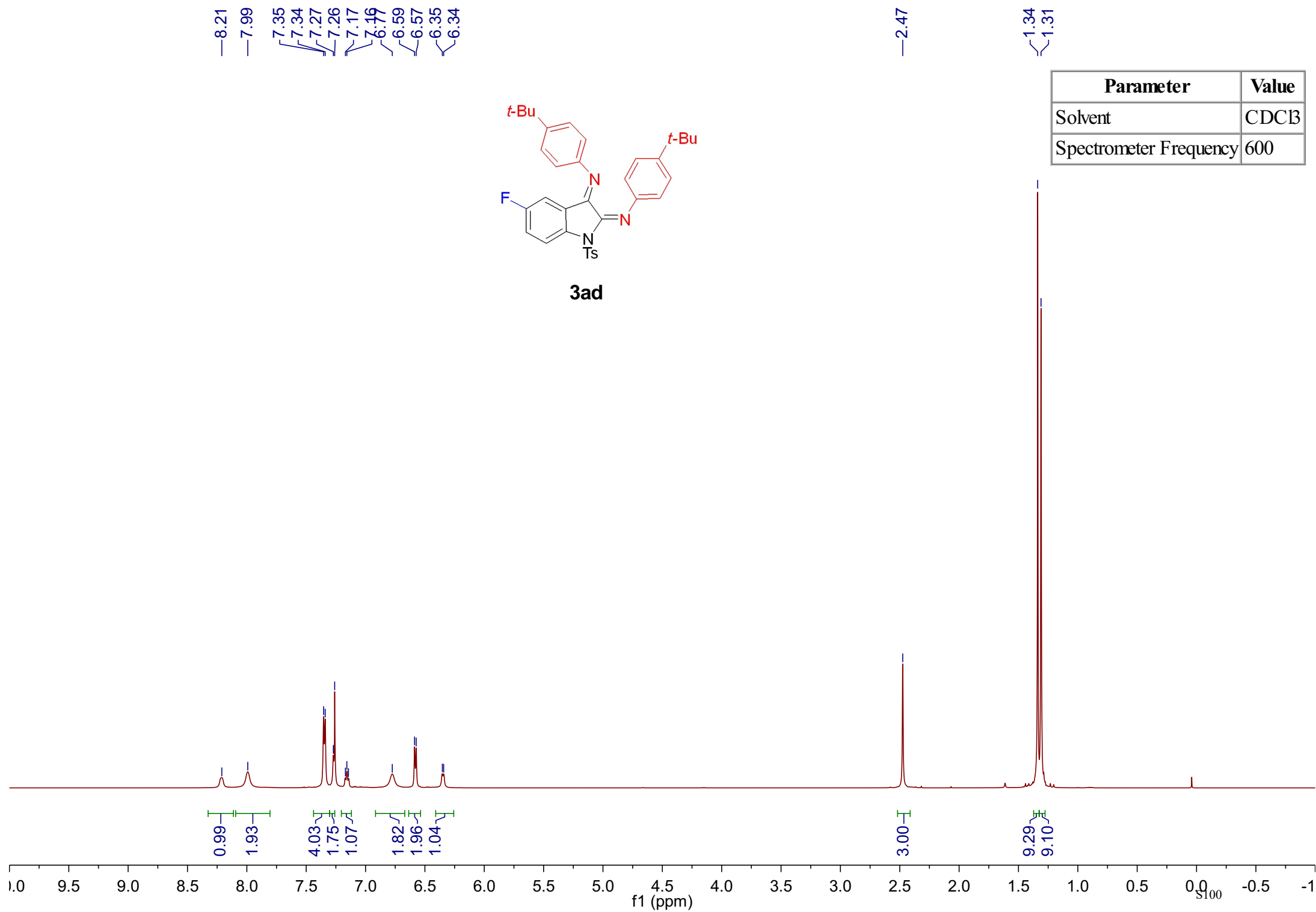
— 155.46
 147.68
 147.51
 145.19
 144.98
 144.36
 139.48
 135.57
 135.36
 128.64
 126.09
 125.18
 121.43
 119.18
 116.93
 — 109.17

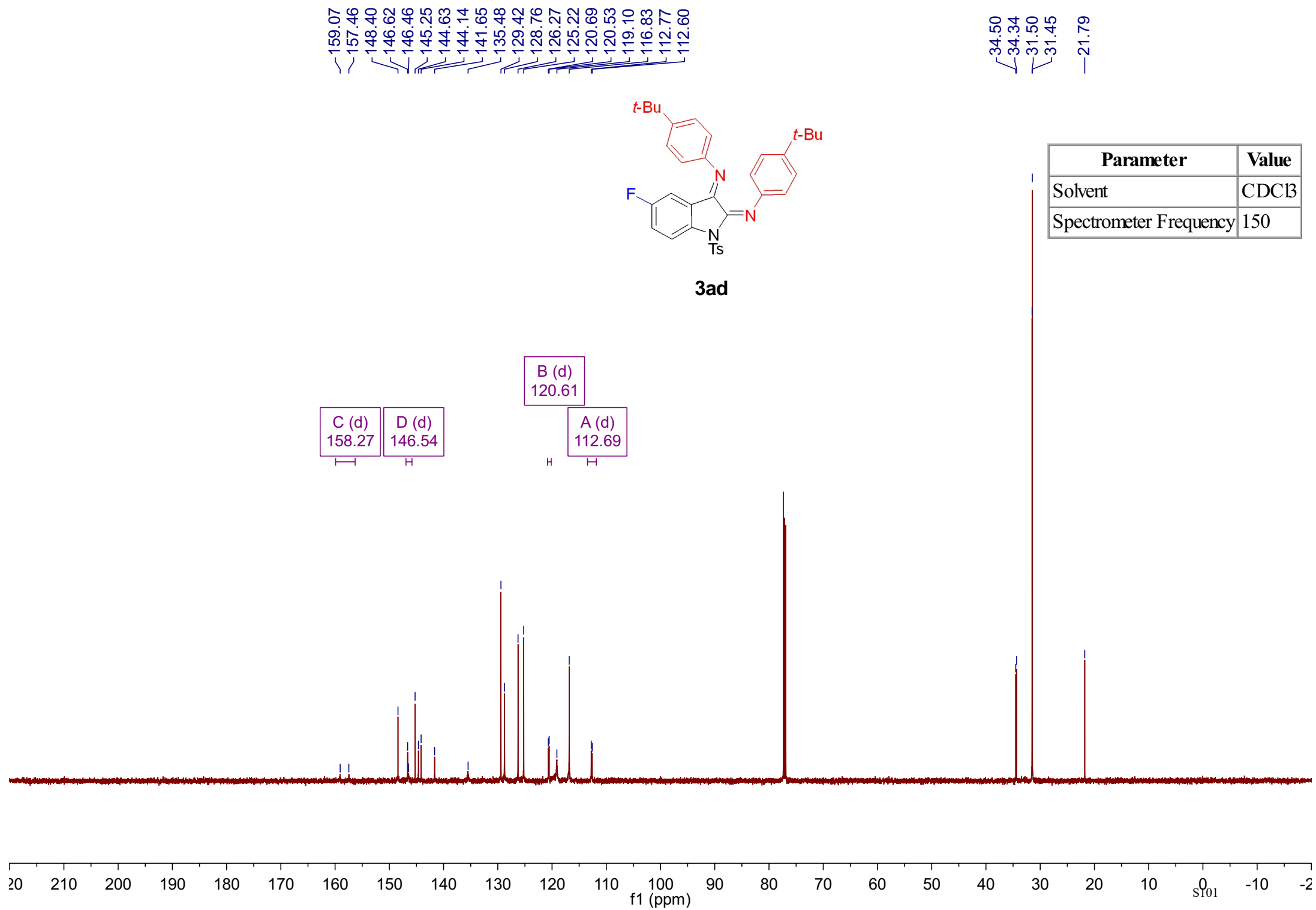
— 54.96

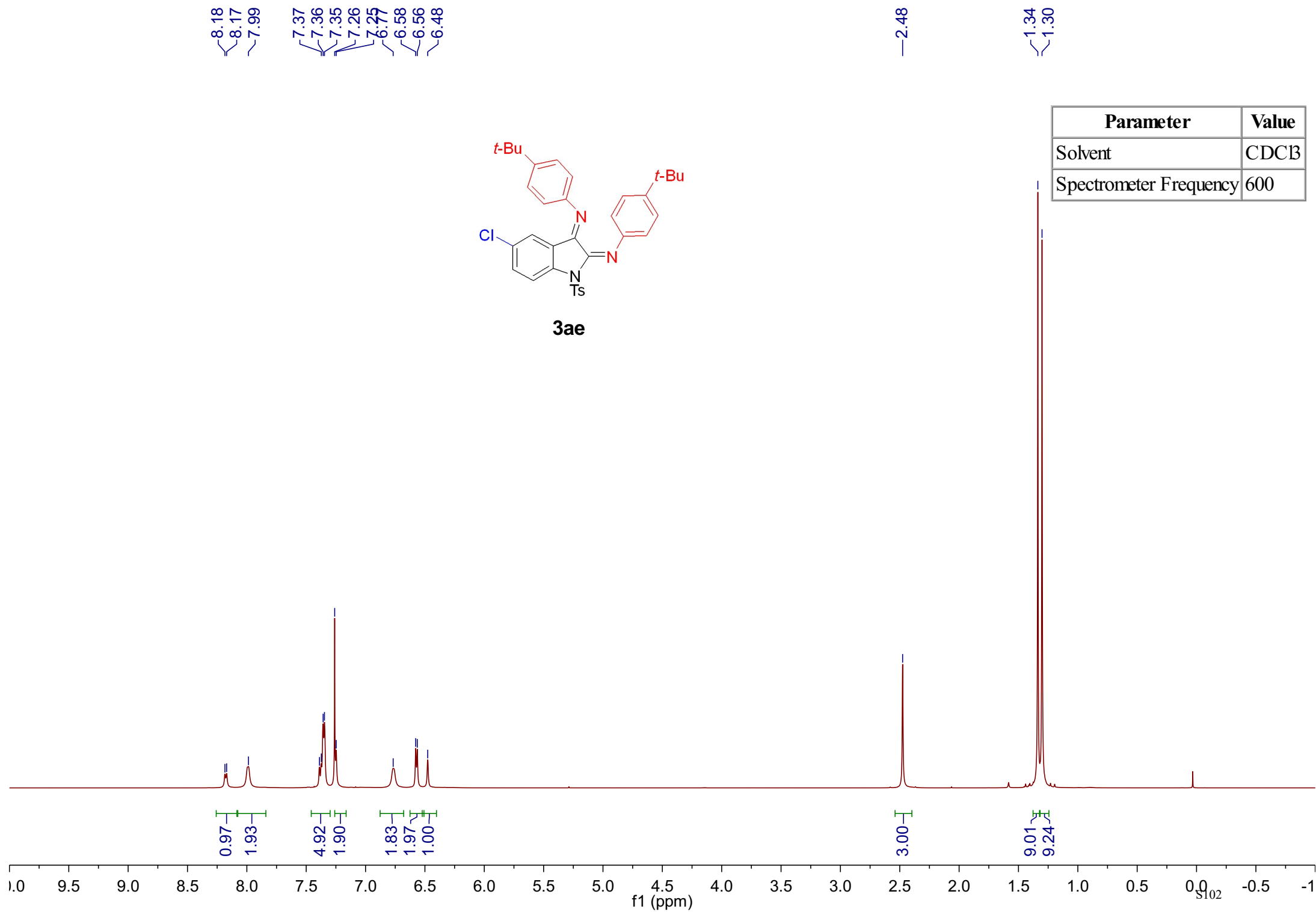
34.44
 34.32
 31.51
 31.45
 — 21.77

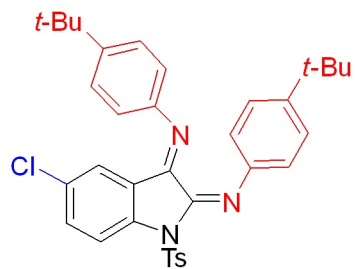
Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150









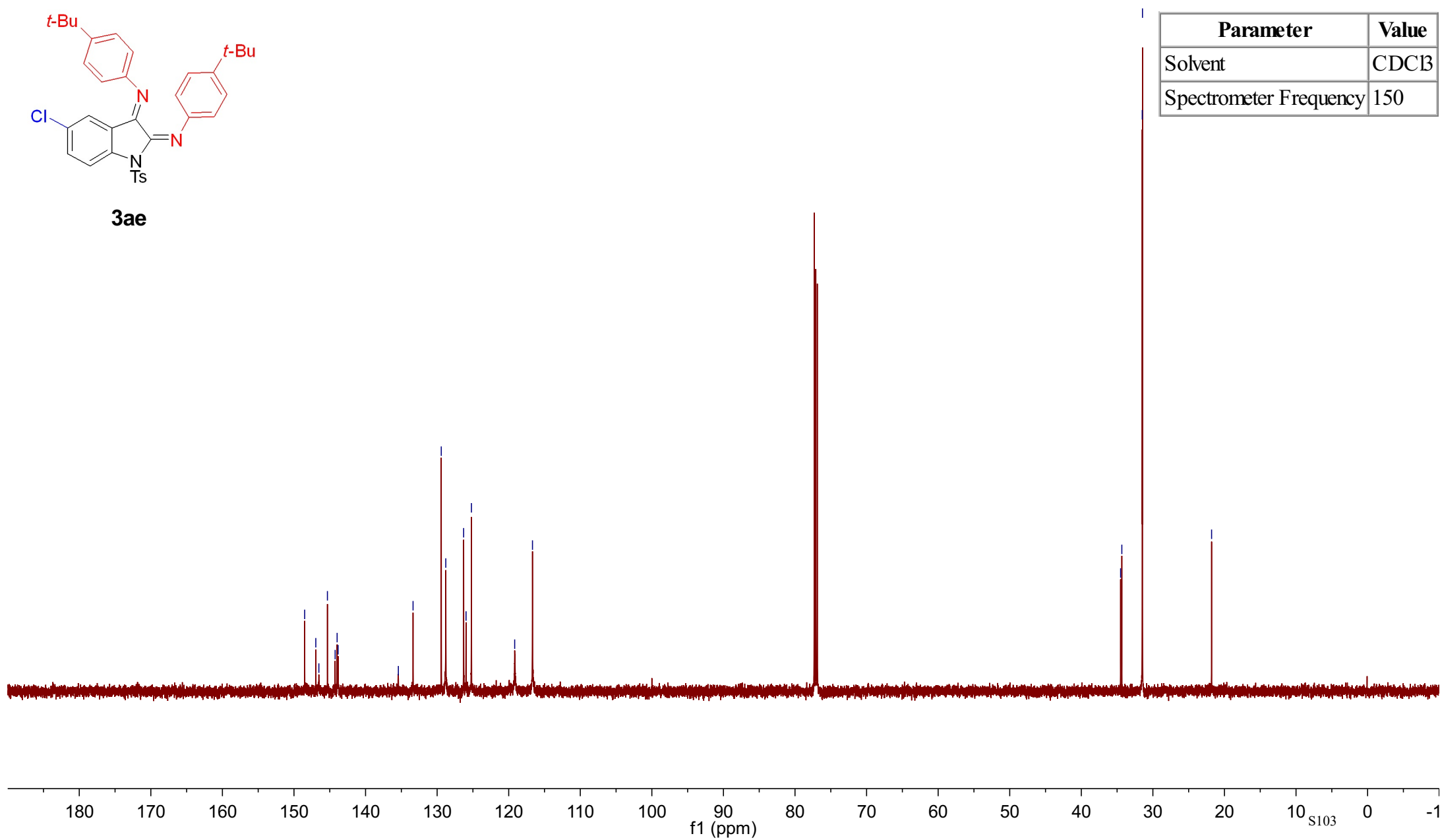


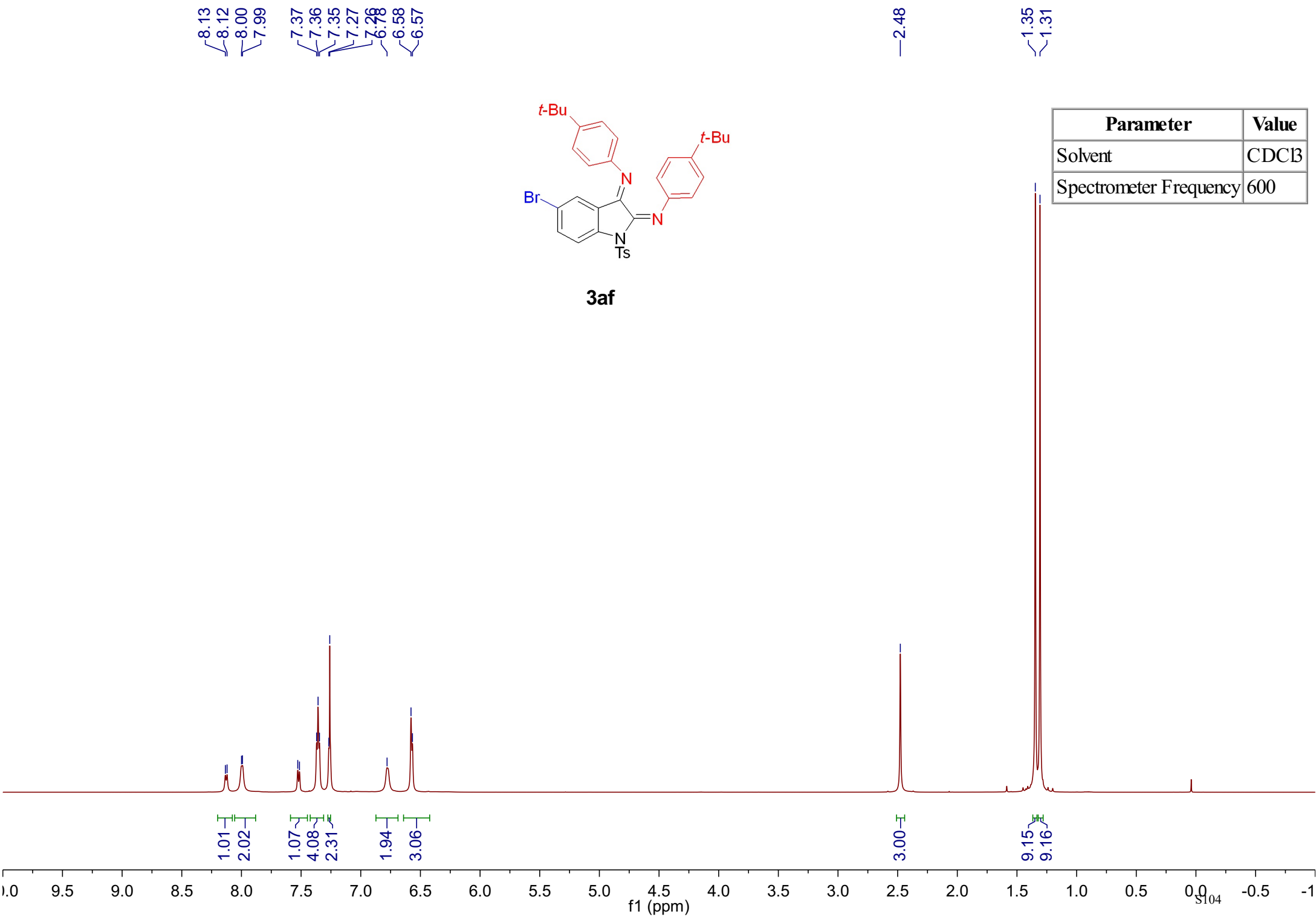
3ae

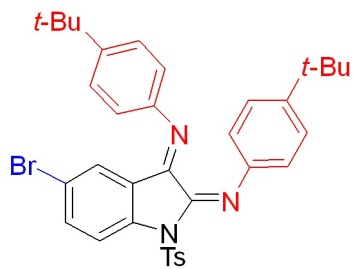
148.51
146.95
146.52
145.33
144.25
143.97
143.84
133.36
129.42
128.79
126.30
125.96
125.77
125.71
116.68

34.51
34.34
31.49
31.45
21.80

Parameter	Value
Solvent	CDC13
Spectrometer Frequency	150





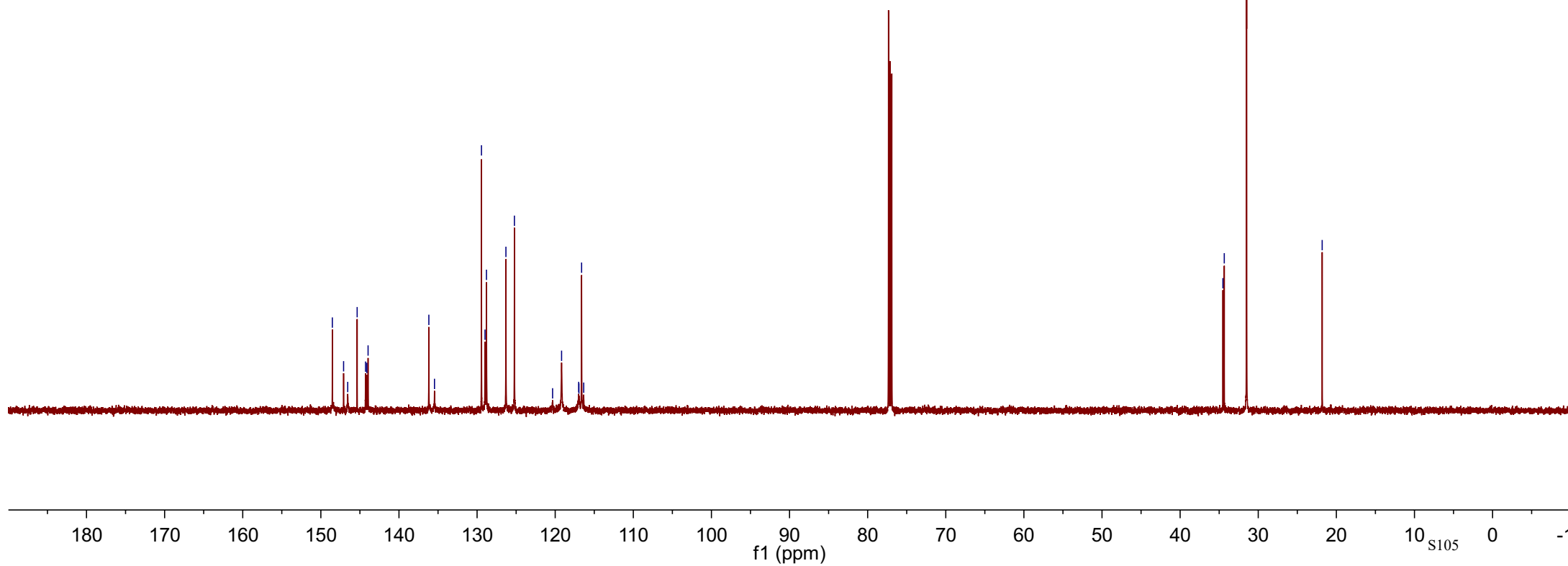


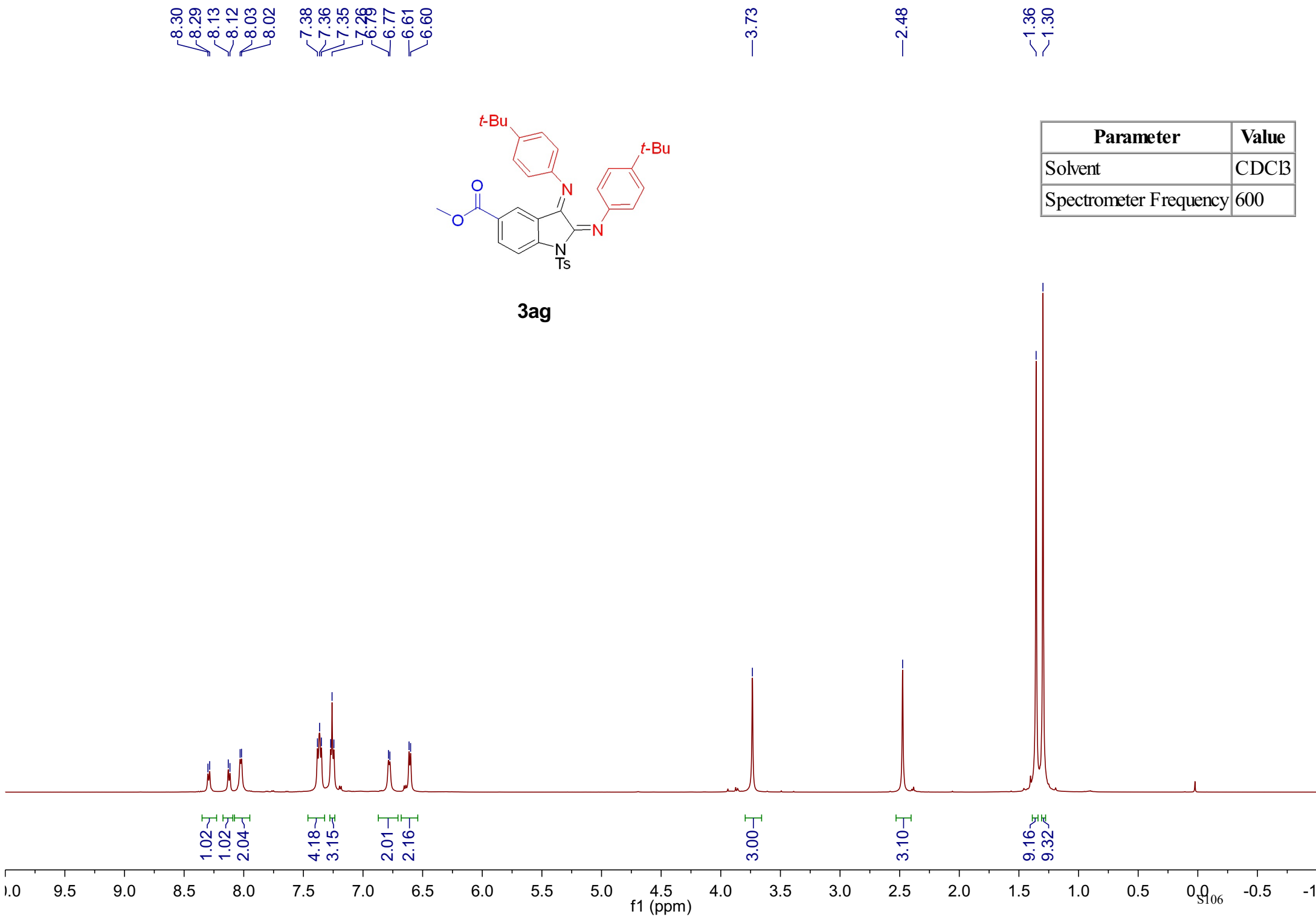
3af

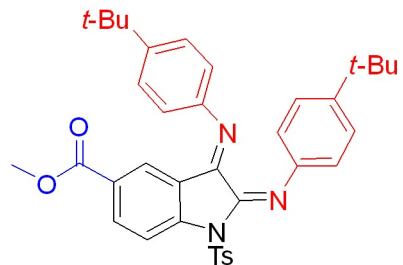
148.53
147.09
146.56
145.35
144.28
144.11
143.95
136.17
135.45
129.44
128.99
128.80
126.31
125.22
120.33
119.18
116.99
116.96
116.62
116.35

34.52
34.35
31.50
31.48
— 21.81

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150







3ag

165.56
148.64
148.48
148.28
147.06
146.58
145.45
144.32
143.79
135.49
135.15
129.43
126.29
125.19
119.21
118.62
116.62
114.94

52.09

34.50

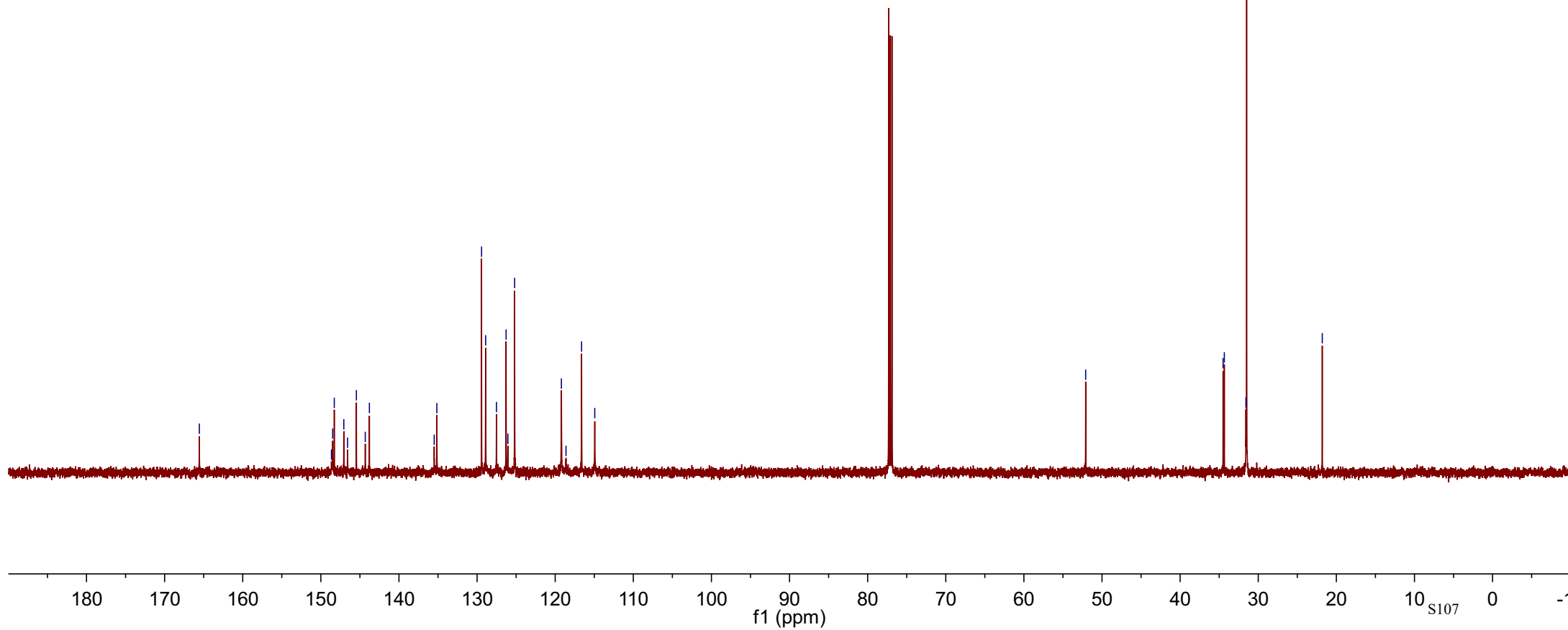
34.34

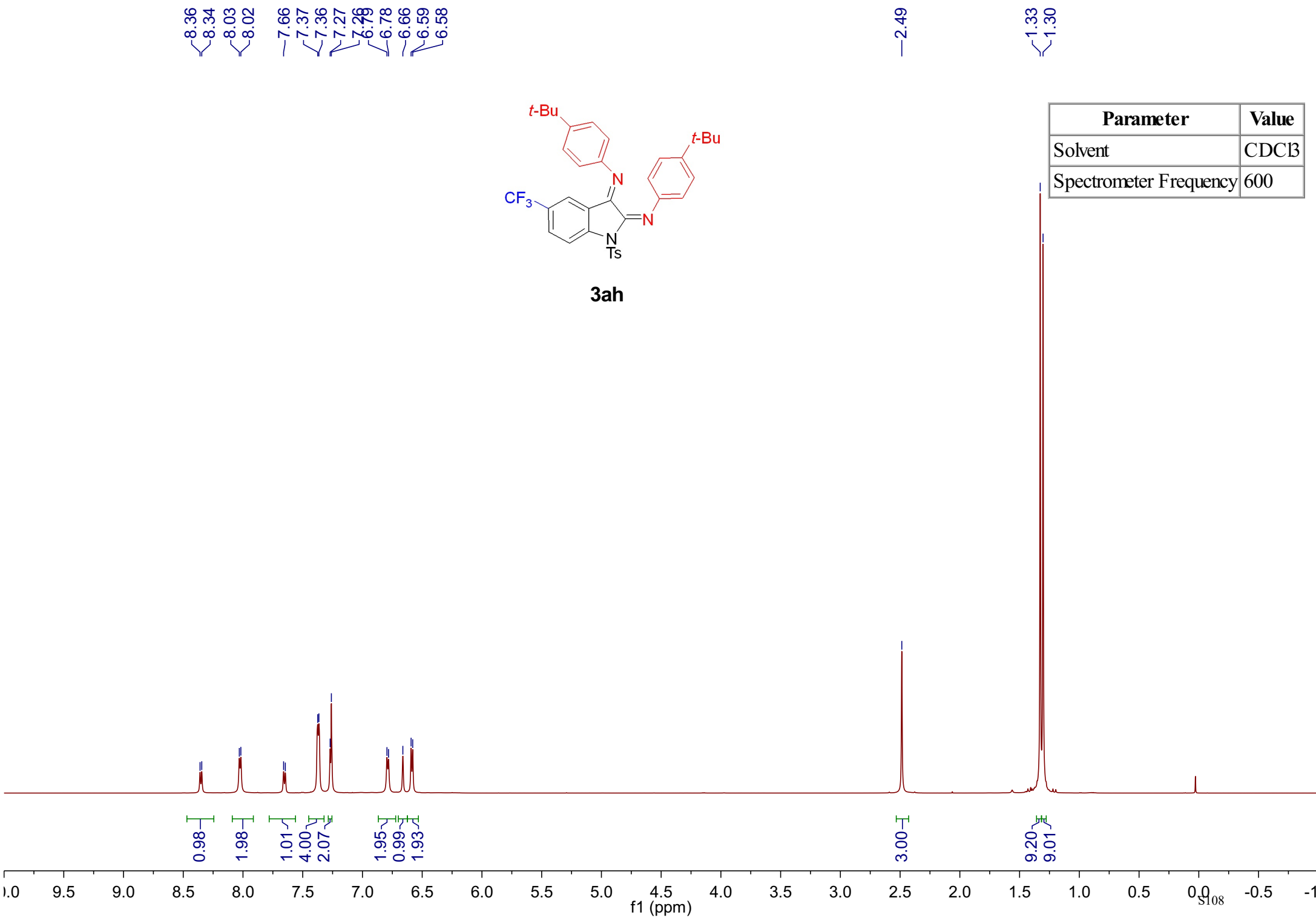
31.58

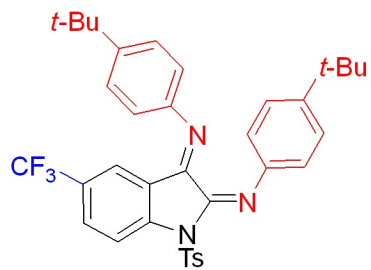
31.48

21.80

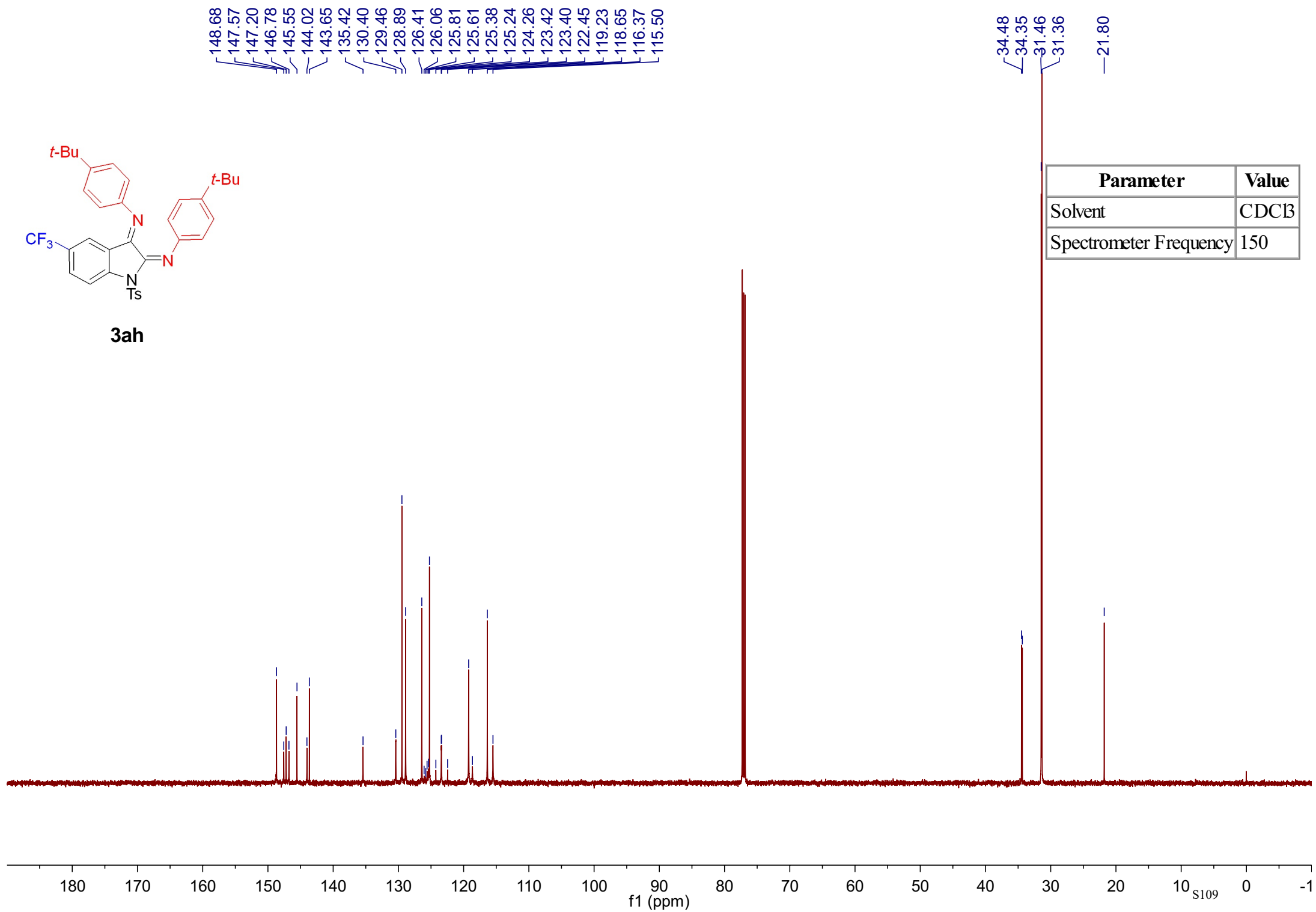
Parameter	Value
Solvent	CDCB
Spectrometer Frequency	150

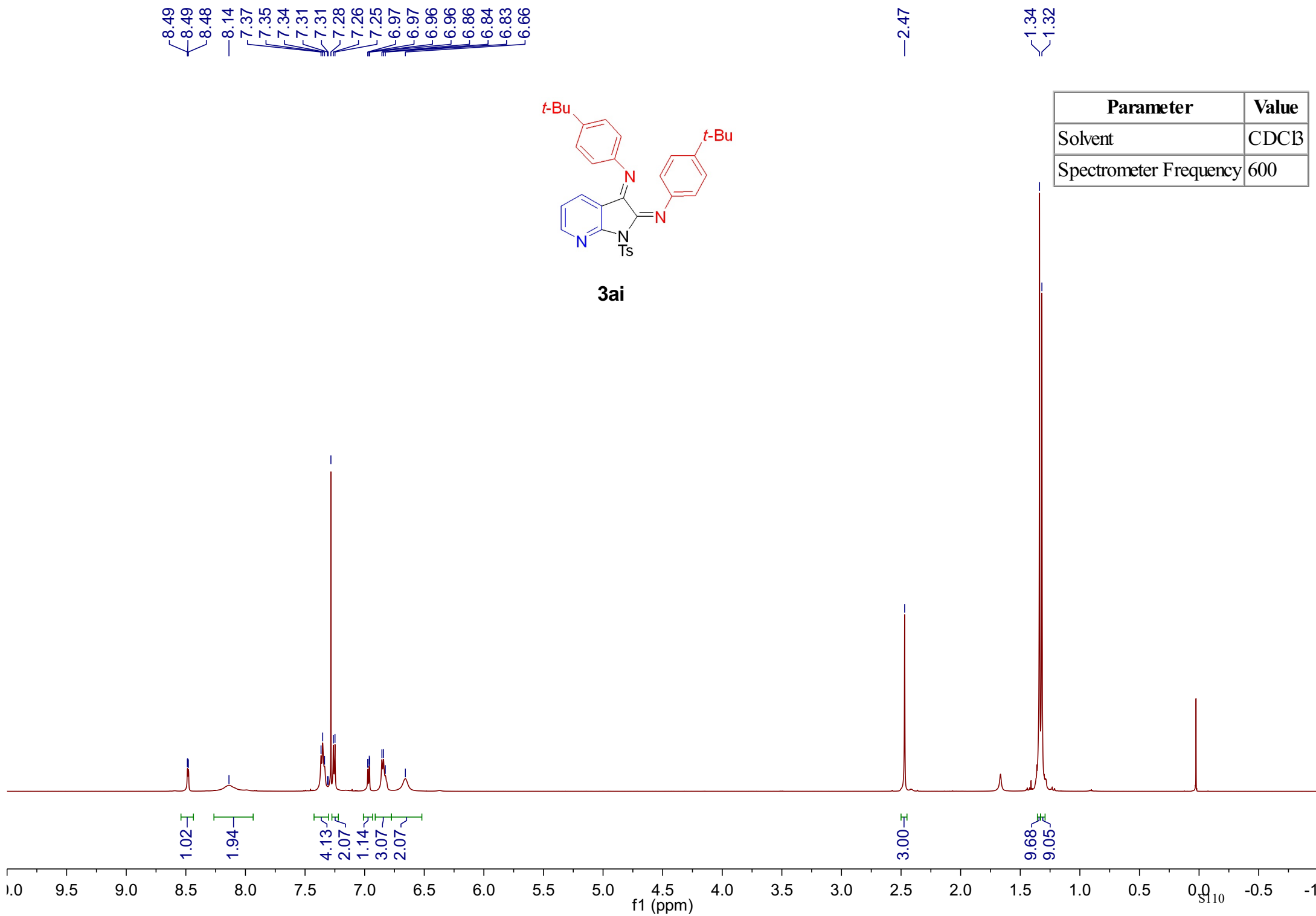


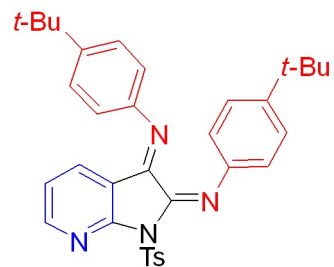




3ah





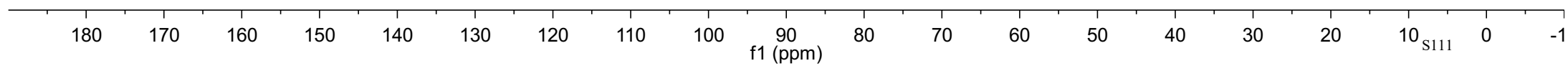


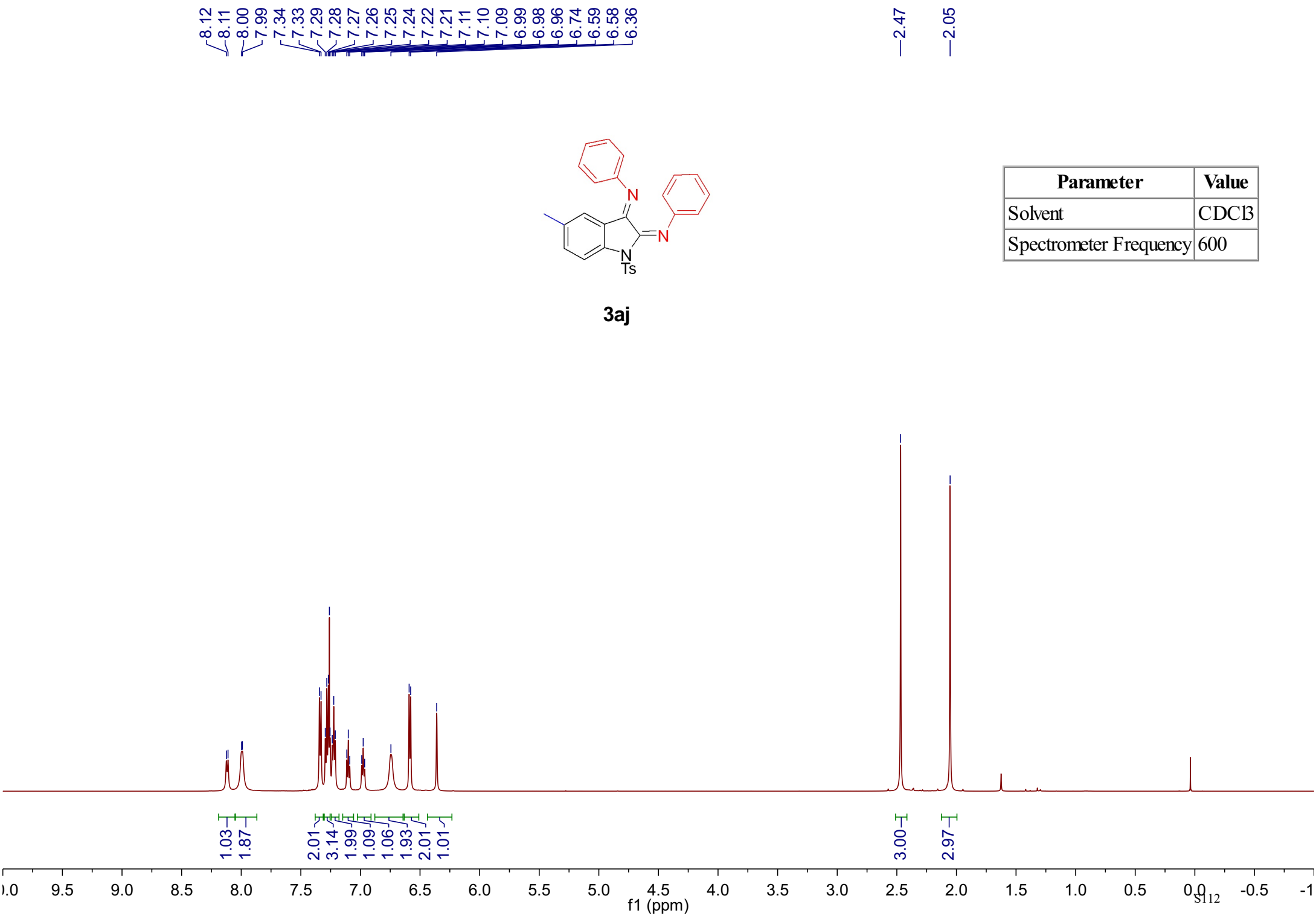
3ai

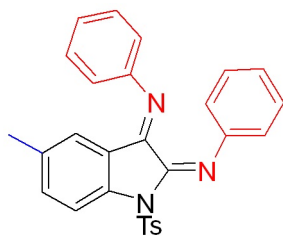
151.96
148.56
146.80
146.55
144.95
136.24
133.86
129.31
128.79
126.29
125.25
119.20
117.16

34.52
34.33
31.48
31.43
21.75

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150





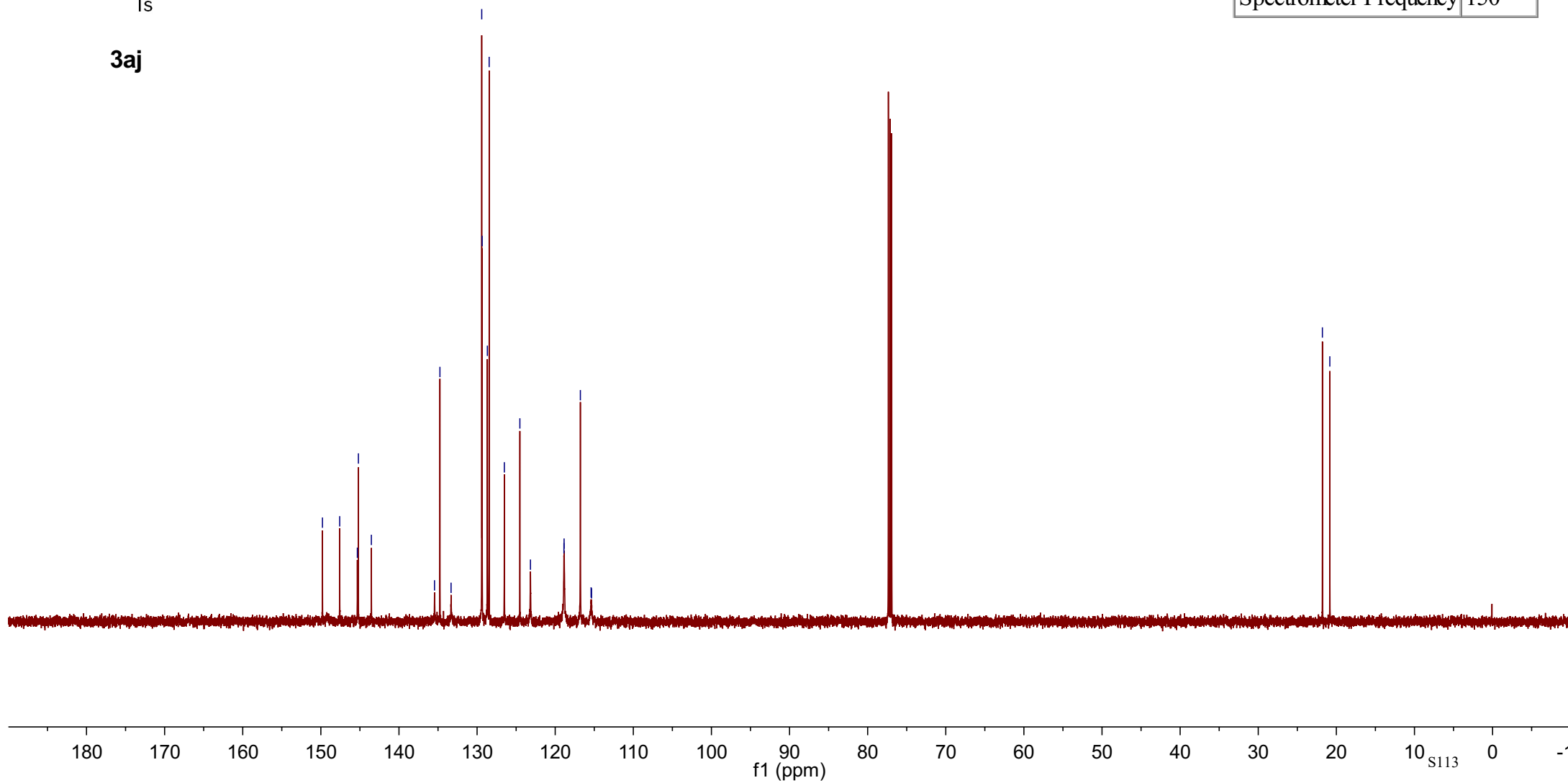


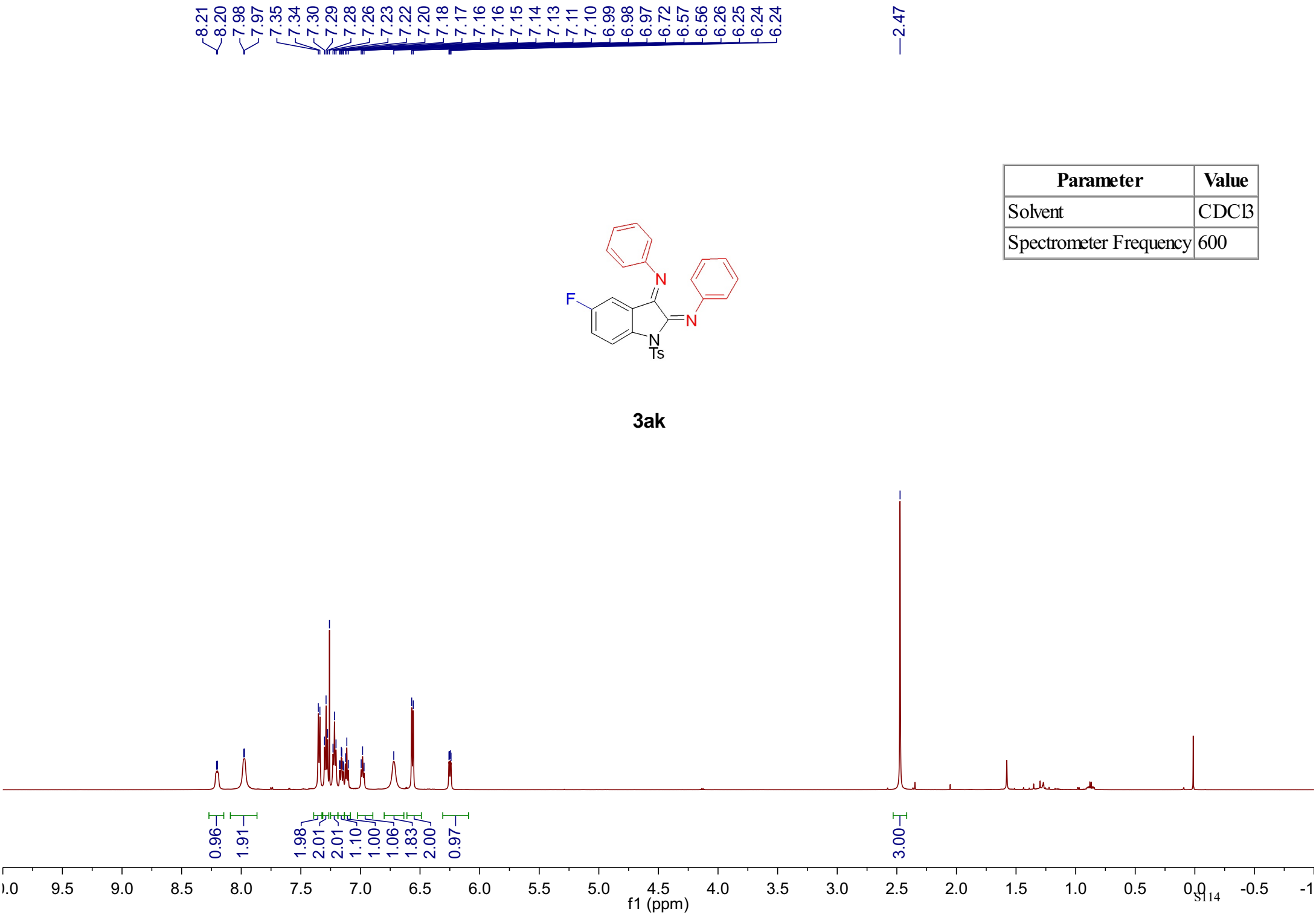
3aj

149.79
147.59
145.33
145.19
143.53
135.45
134.76
133.33
129.40
129.35
128.68
128.45
126.51
124.52
123.17
118.88
118.86
116.78
115.41
115.35

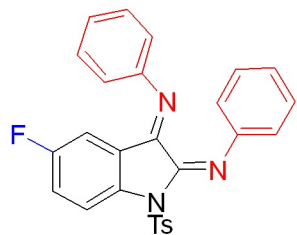
21.79
20.84

Parameter	Value
Solvent	CDCI3
Spectrometer Frequency	150

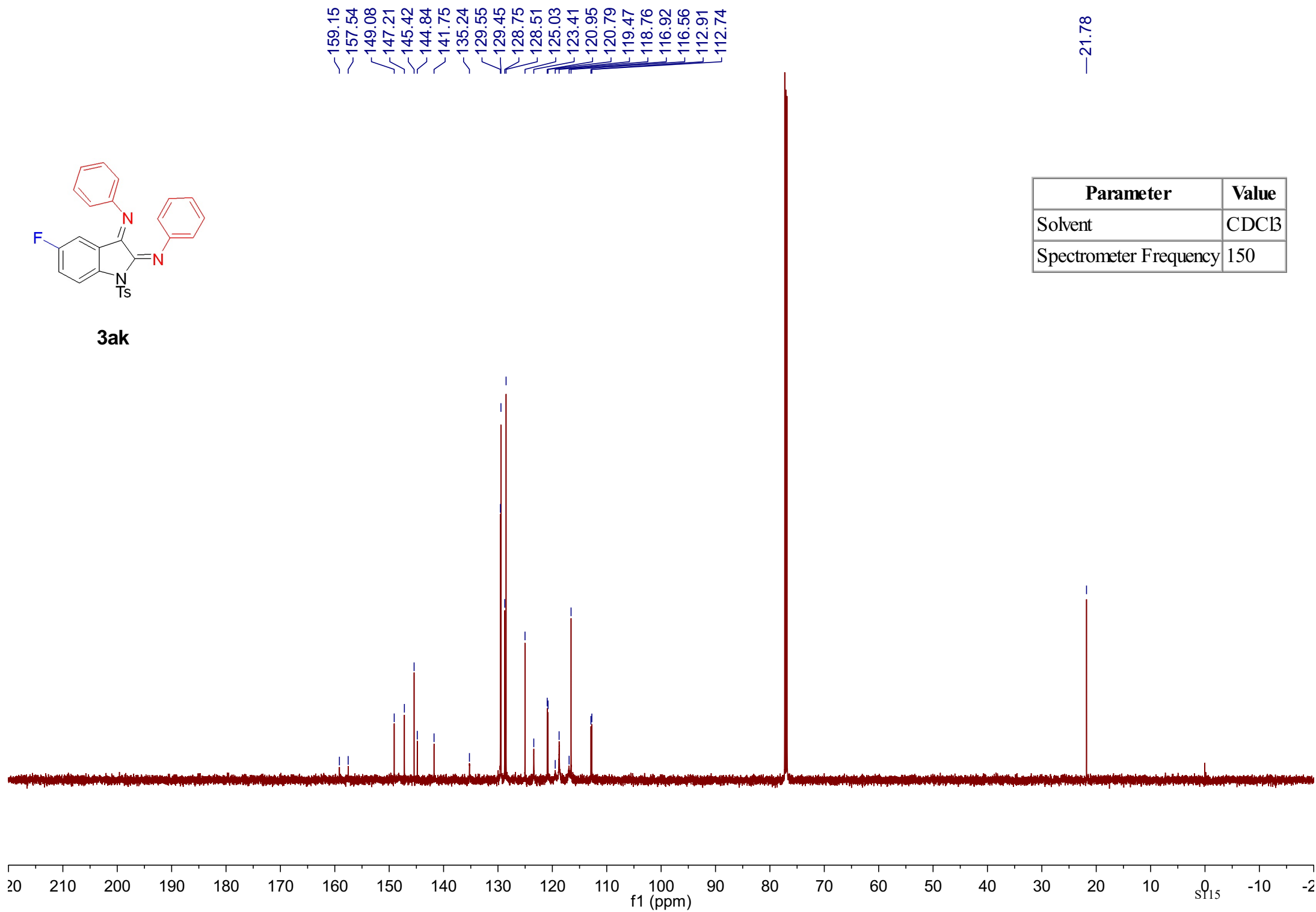


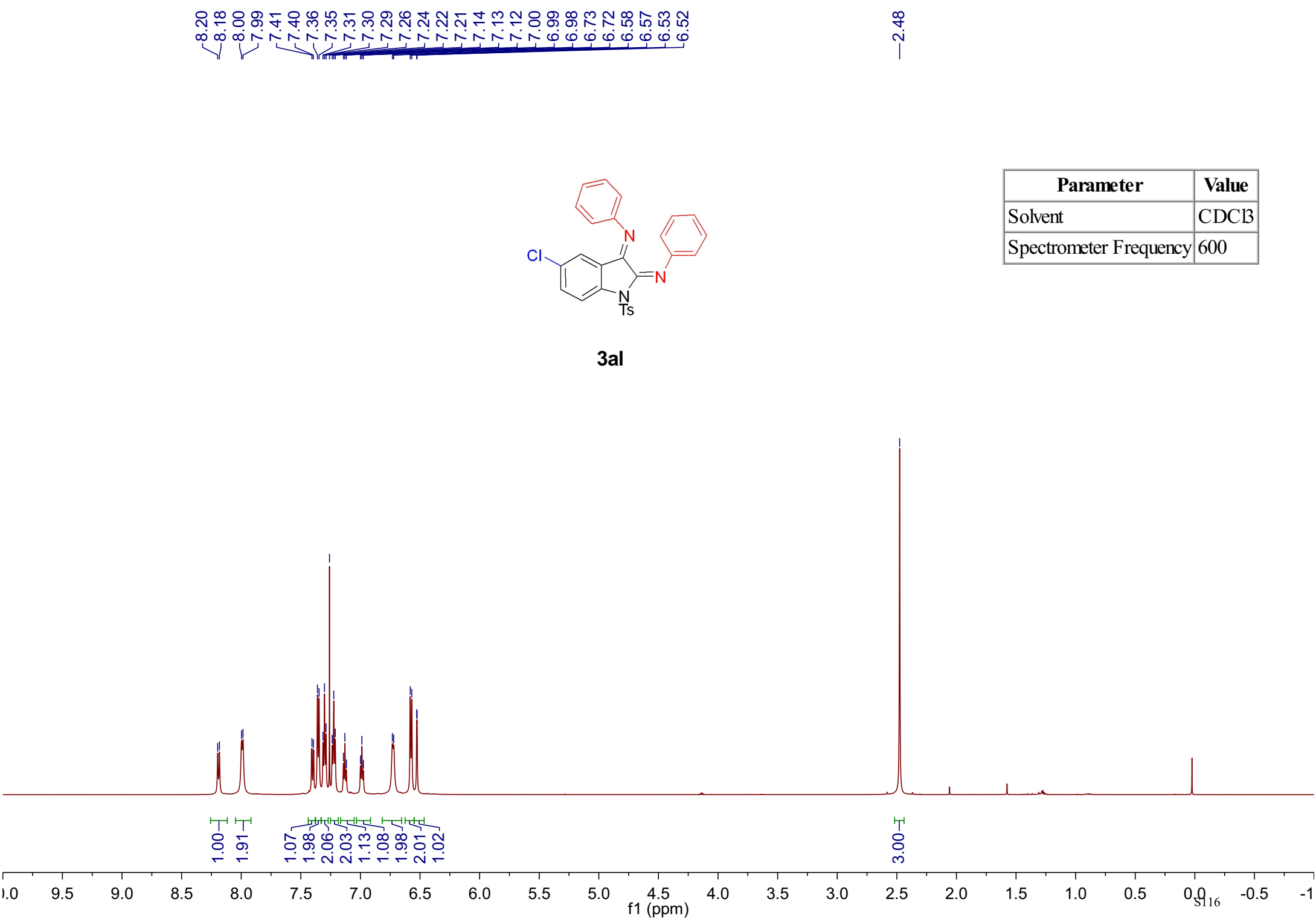


Parameter	Value
Solvent	CDCl3
Spectrometer Frequency	600

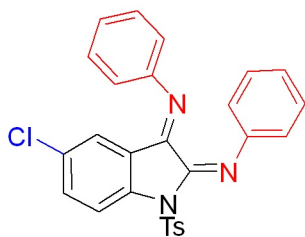


3ak





Parameter	Value
Solvent	CDCl3
Spectrometer Frequency	600

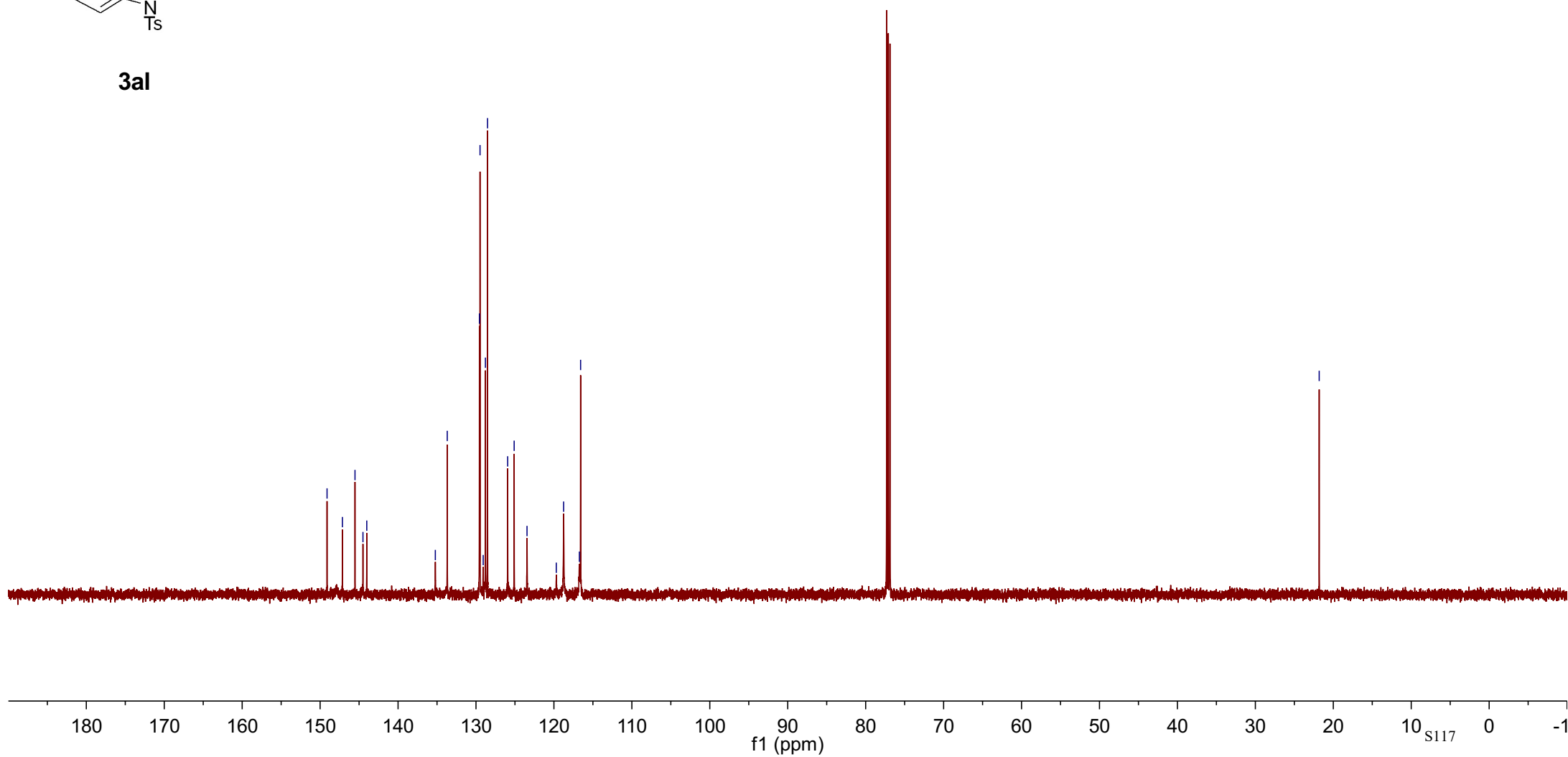


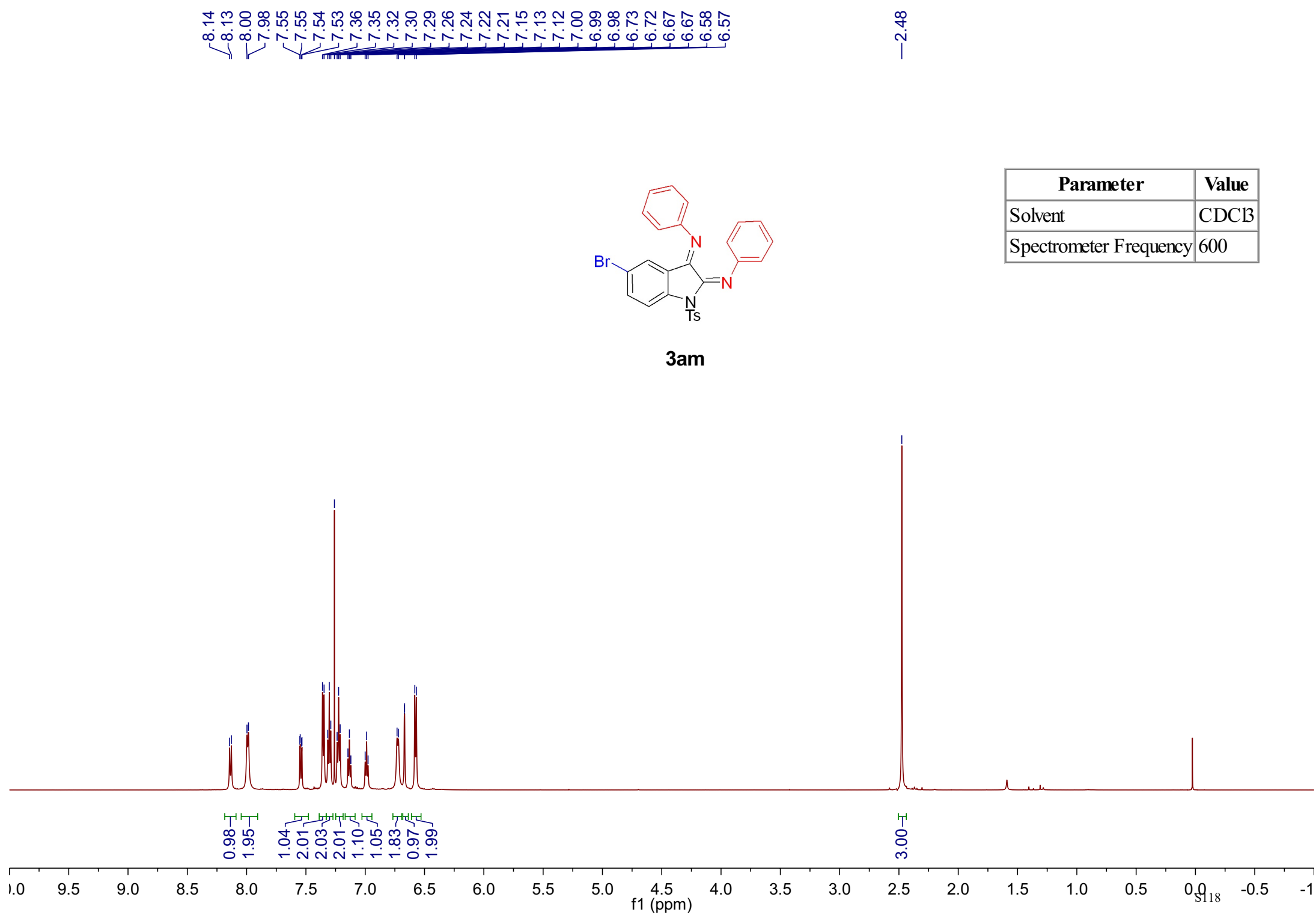
3al

149.11
147.14
145.53
144.50
144.01
135.22
133.69
129.56
129.48
129.08
128.79
128.52
125.94
125.10
123.45
119.70
118.76
116.73
116.58

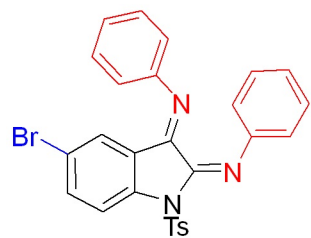
—21.81

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150





Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	600

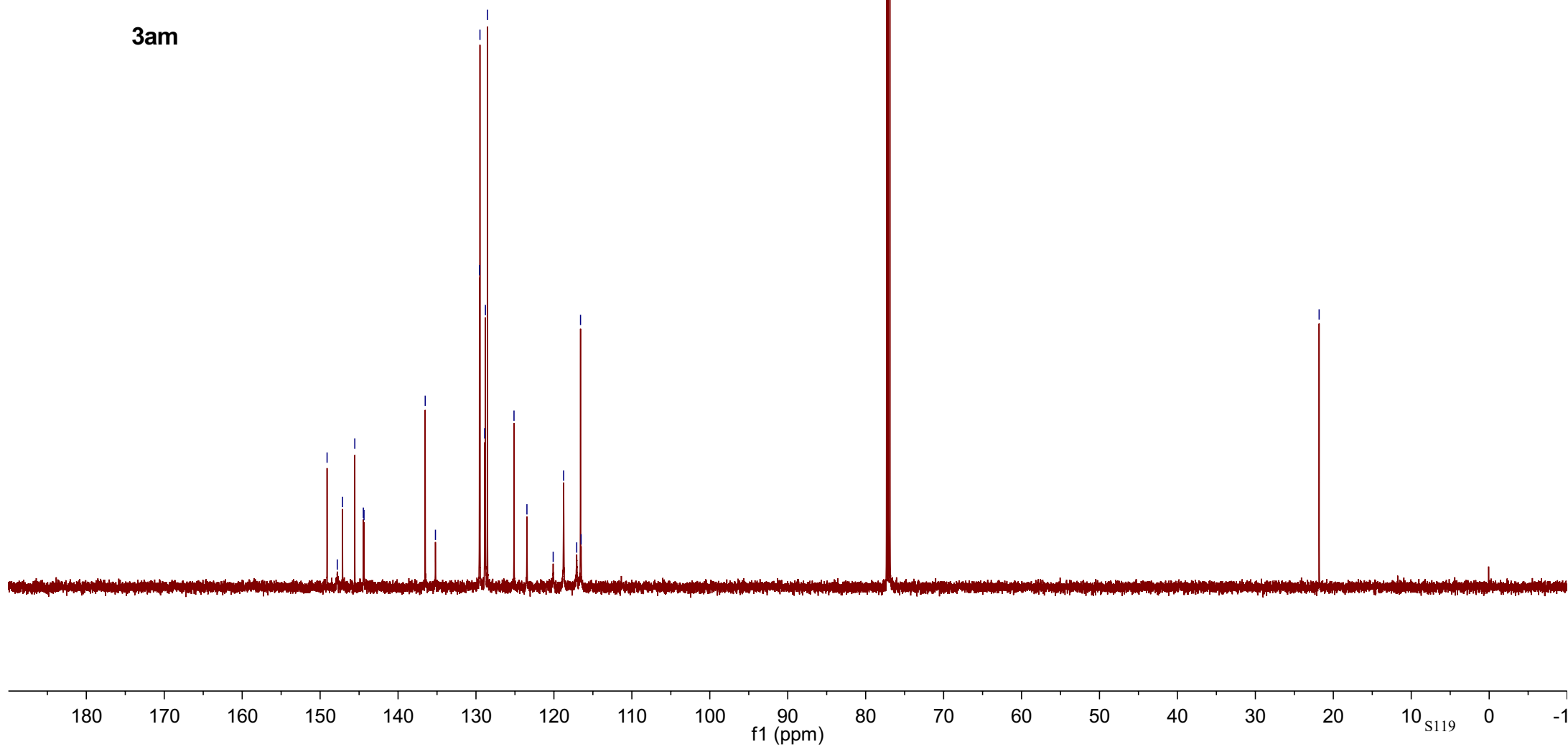


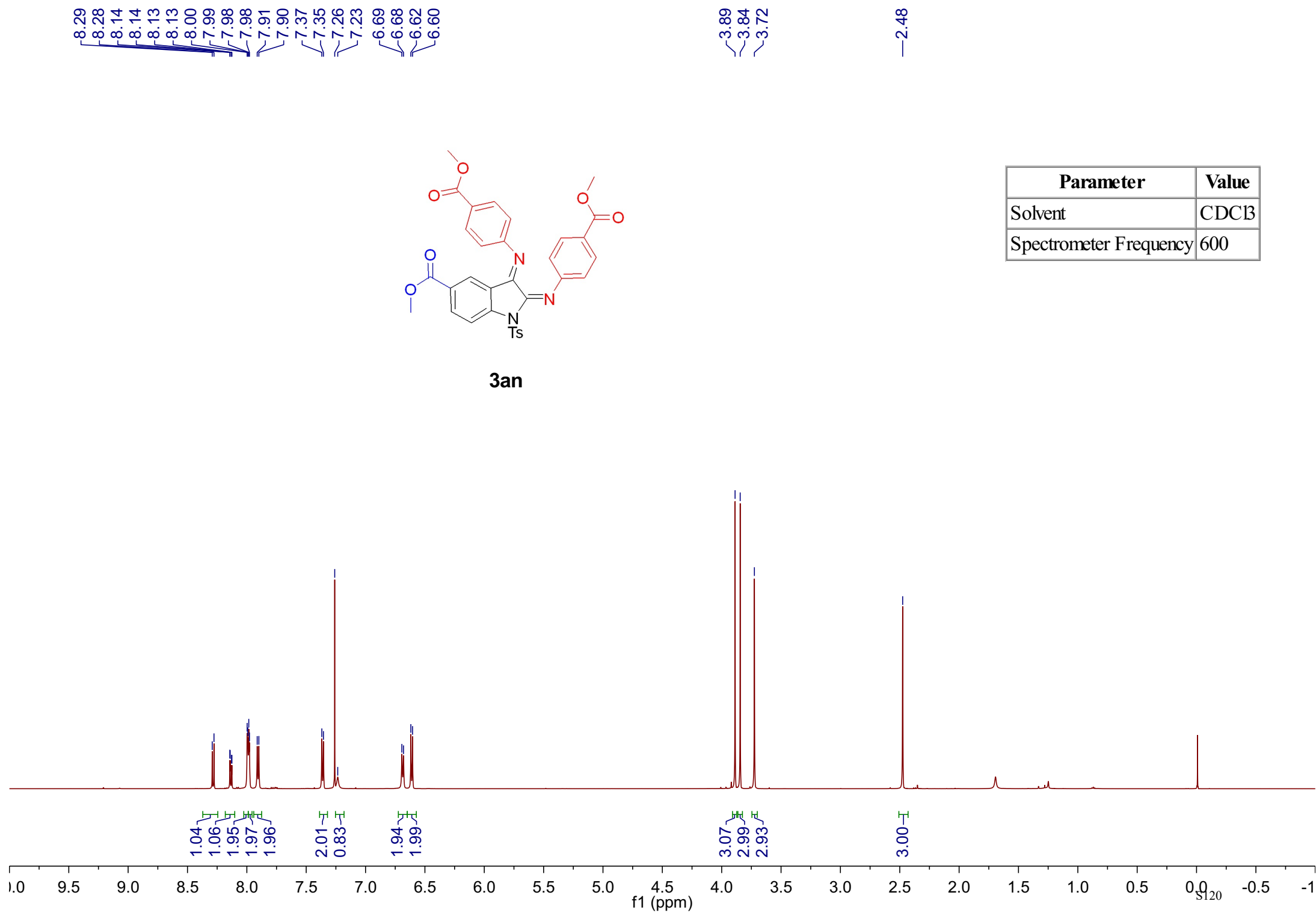
3am

149.11
147.80
147.13
145.55
144.46
144.37
136.51
135.20
129.55
129.49
128.89
128.79
128.53
125.12
123.46
120.10
118.76
117.08
116.59
116.51

—21.82

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150





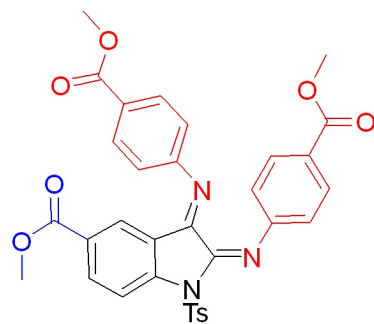
166.94
166.41
165.16

152.69
151.66
148.75
148.04
146.05
144.29

135.89
131.39
130.60
129.61
128.83
126.89
126.89
116.43
115.14

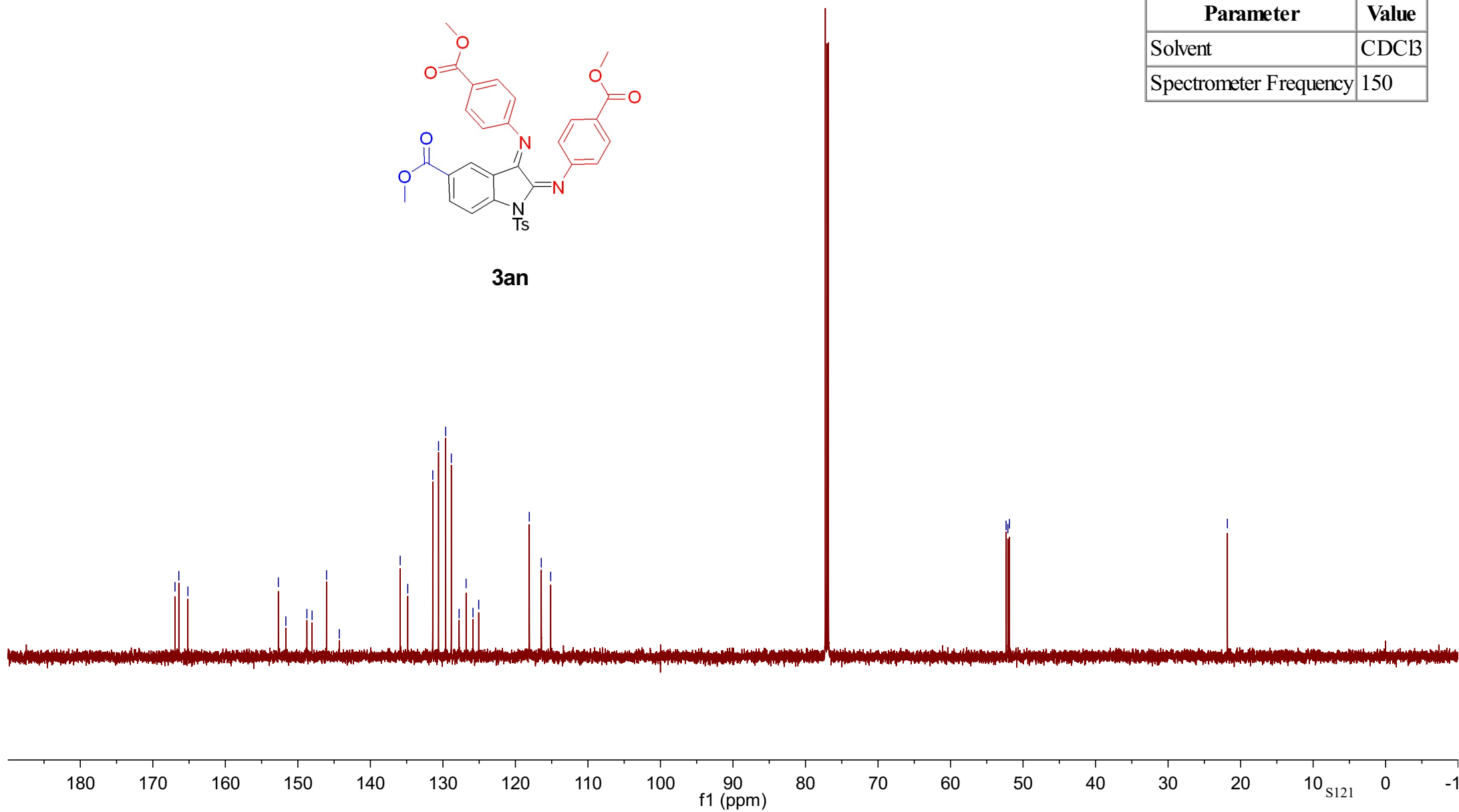
52.34
52.10
51.88

21.82



3an

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150



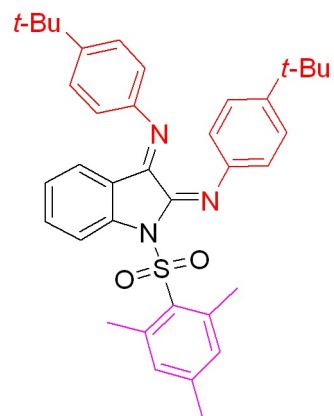
8.23
8.22
7.44
7.43
7.42
7.38
7.37
7.26
7.20
7.19
7.02
6.83
6.82
6.81
6.78
6.76
6.70
6.68
6.65
6.63

2.67

2.37

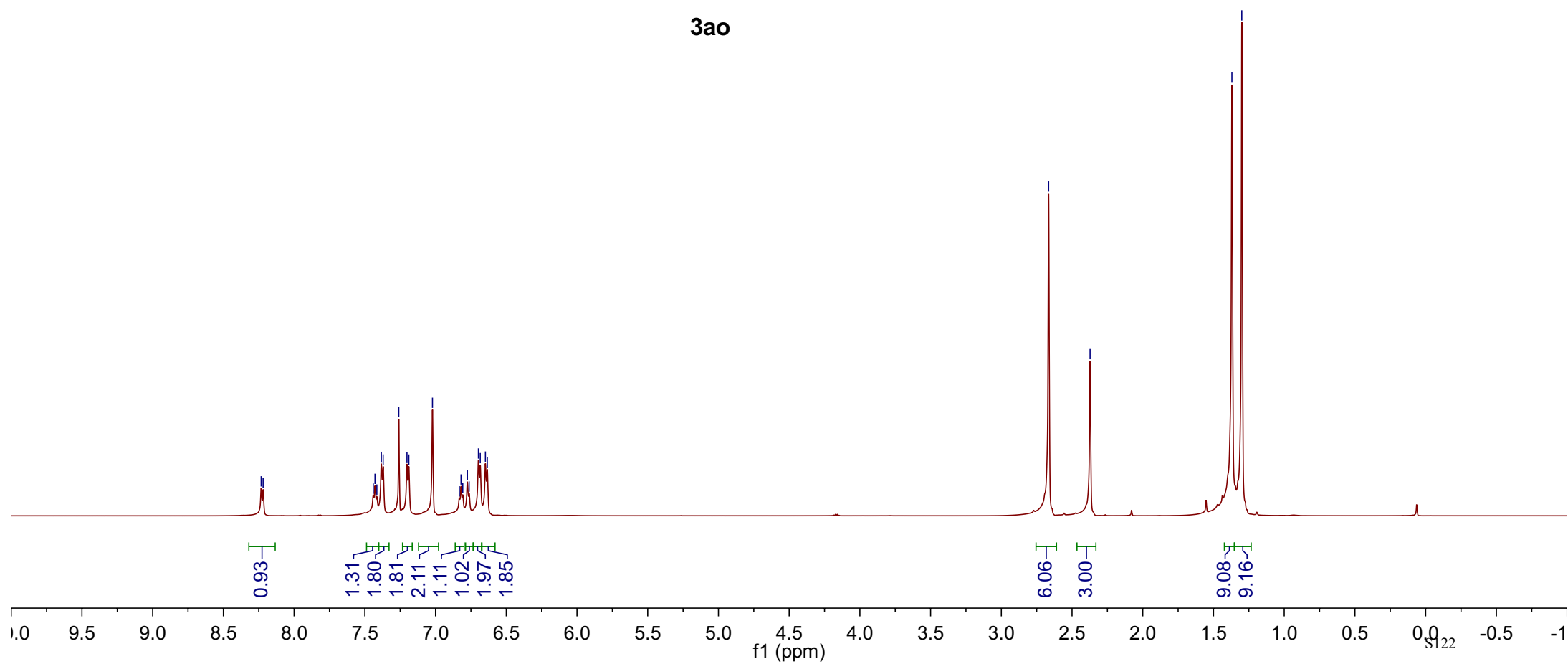
1.37

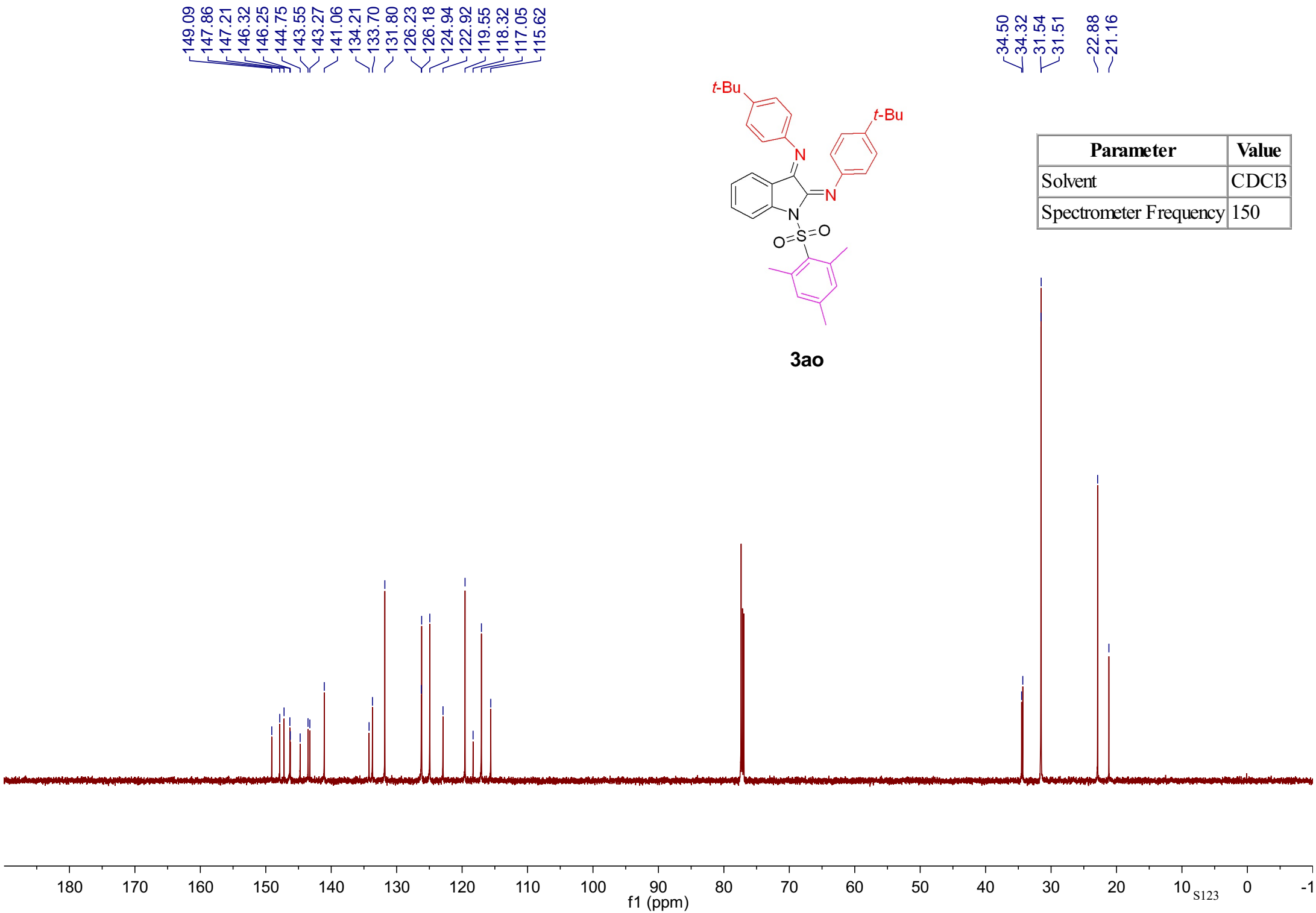
1.30

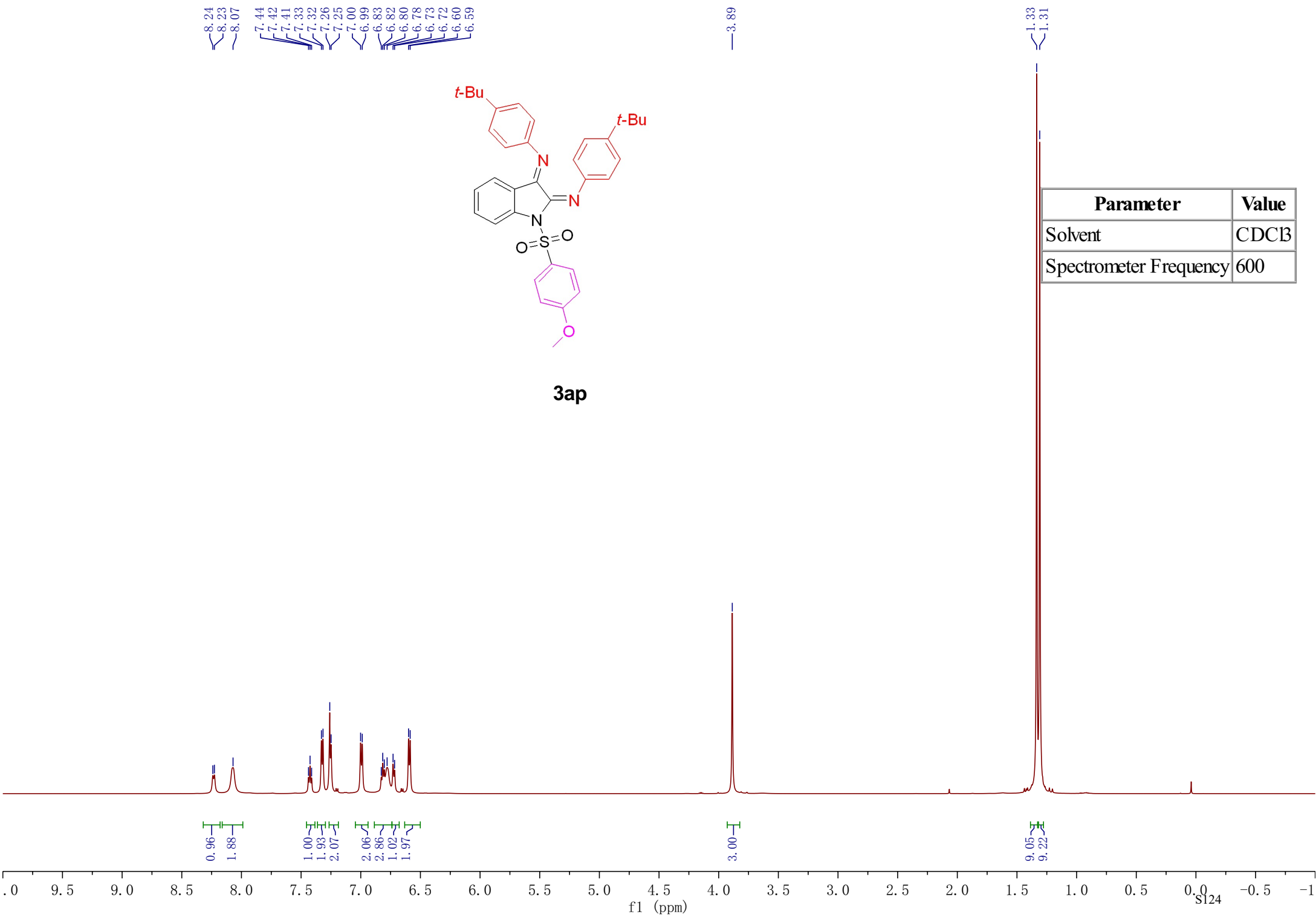


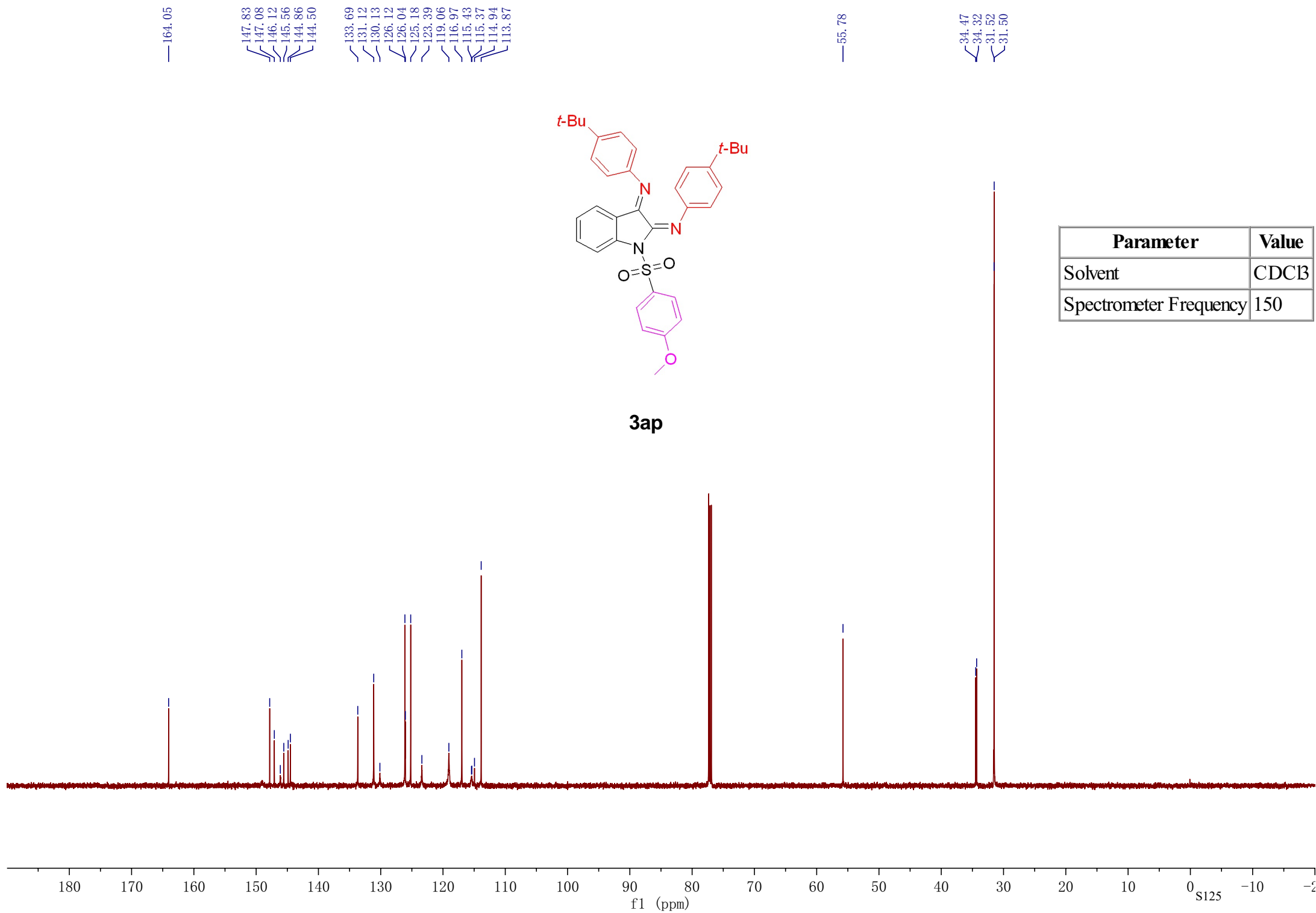
3ao

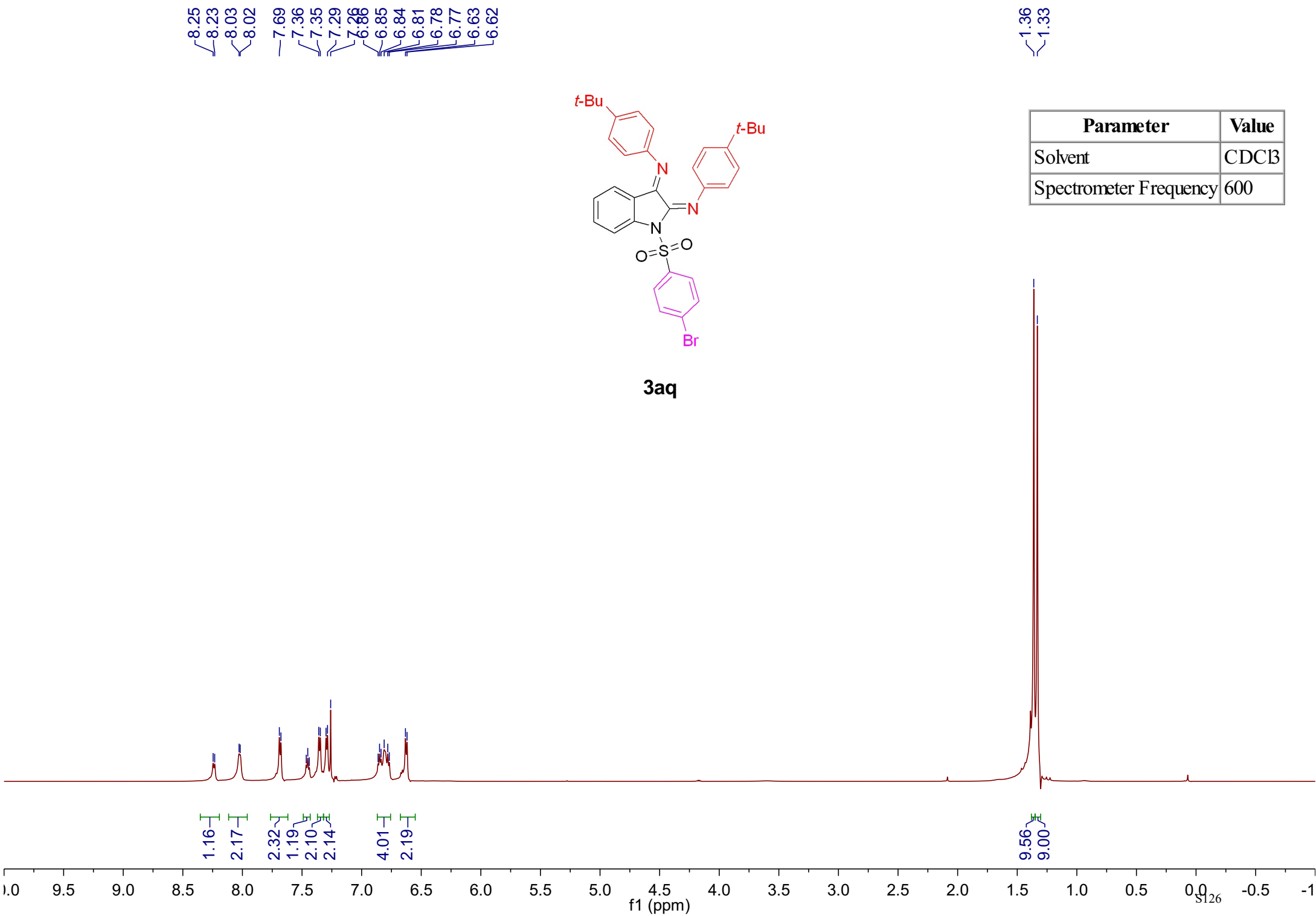
Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	600





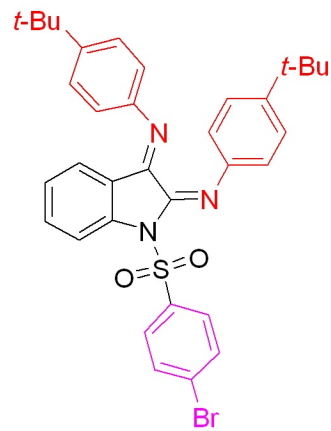






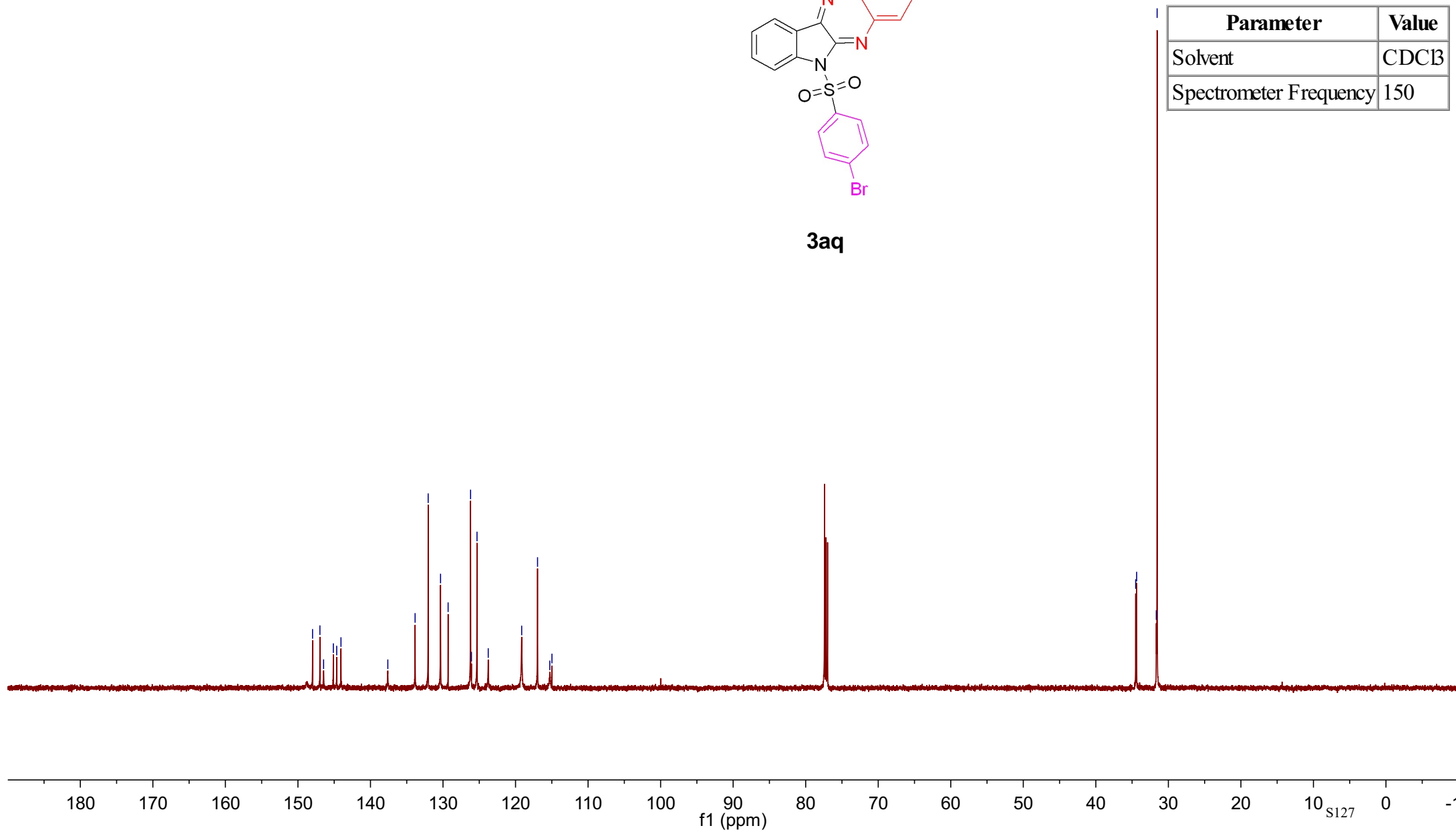
147.98
146.97
146.46
145.12
144.65
144.07
137.61
133.85
132.05
130.34
129.30
126.20
126.08
125.31
123.76
119.15
116.97
115.28
114.98

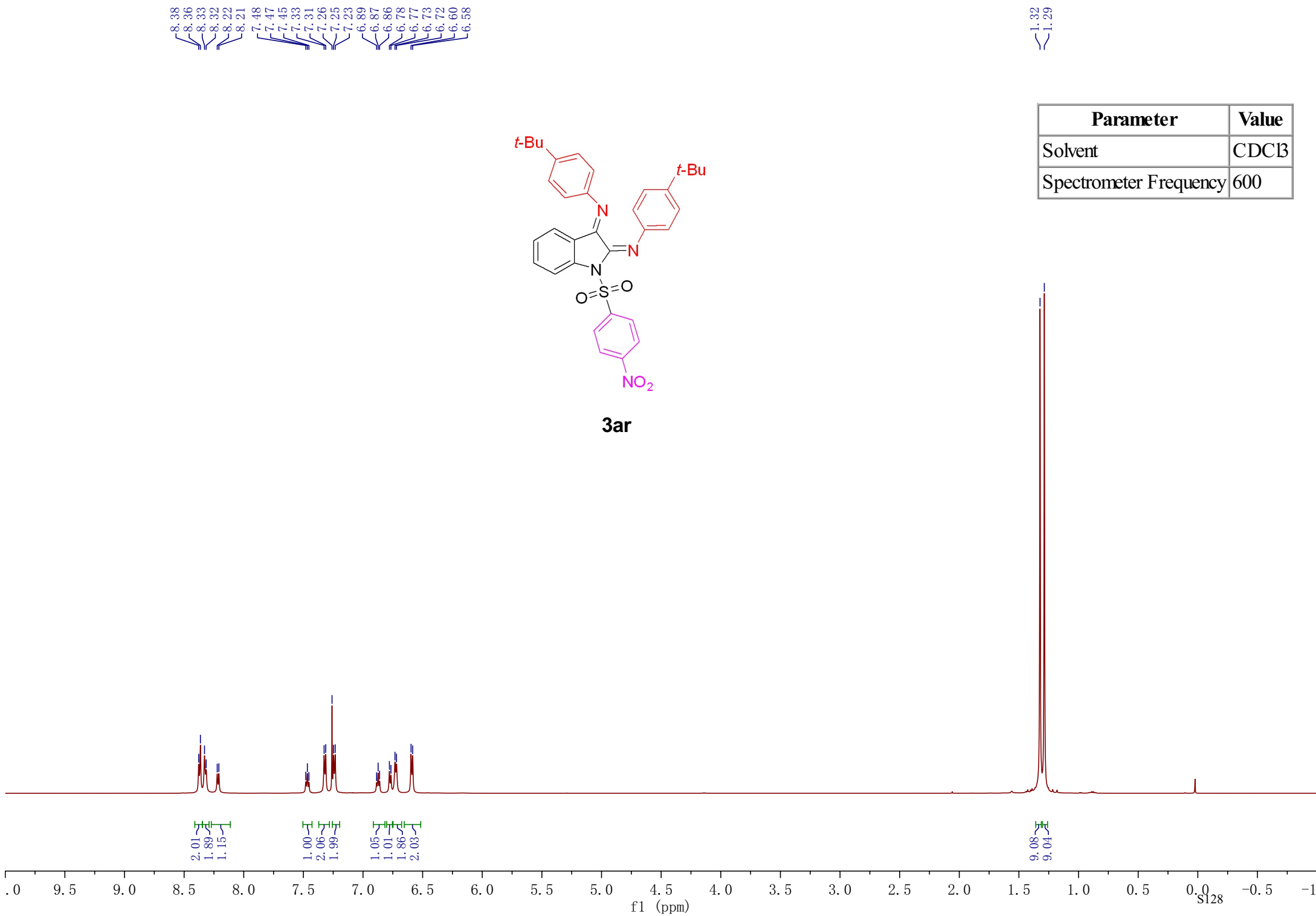
34.51
34.37
31.66
31.55

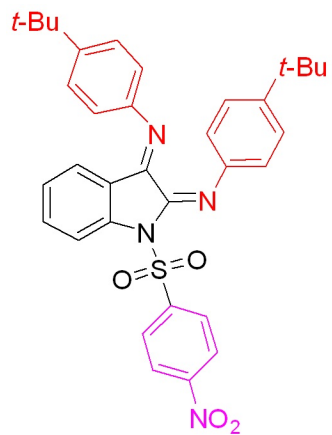


3aq

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150





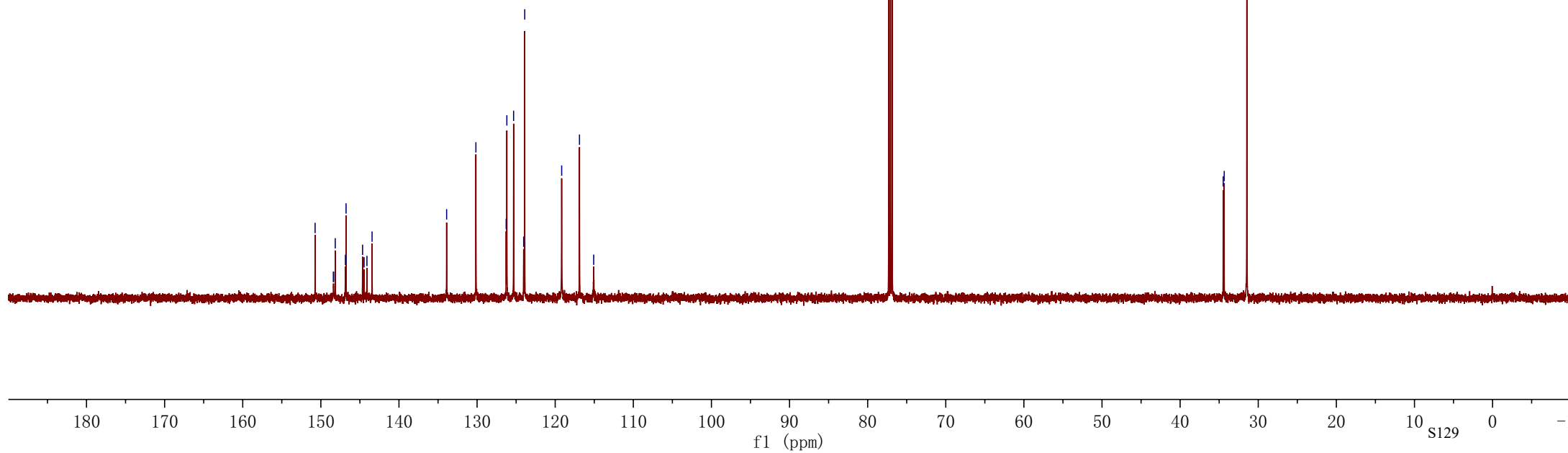


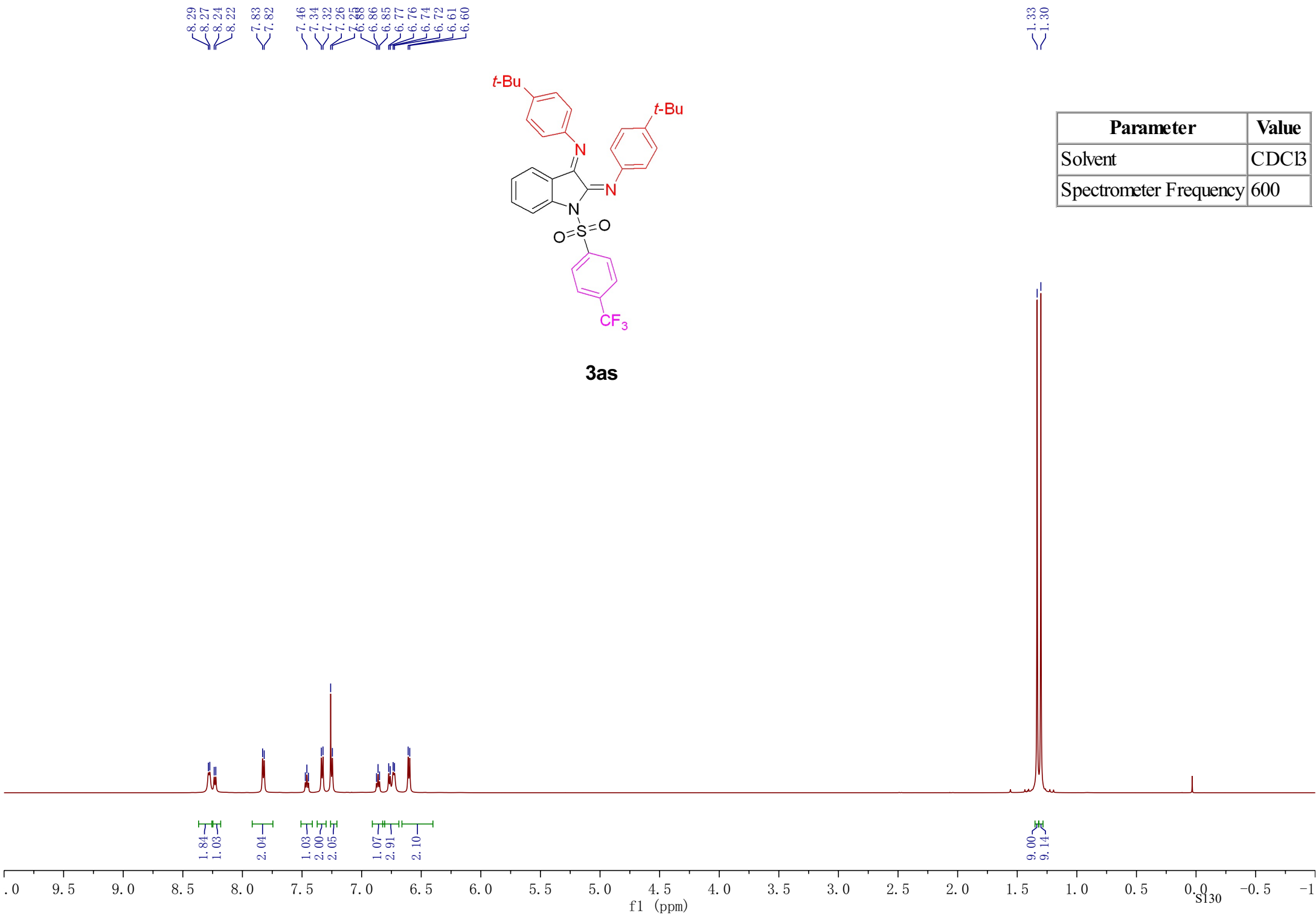
3ar

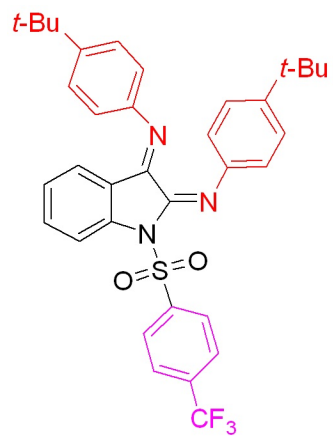
150.73
148.39
148.38
148.16
146.86
146.77
144.66
144.47
144.10
143.44
133.90
130.15
126.28
126.19
125.32
124.04
123.90
119.17
116.89
115.07

34.48
34.35
31.46
31.45

Parameter	Value
Solvent	CDCB
Spectrometer Frequency	150





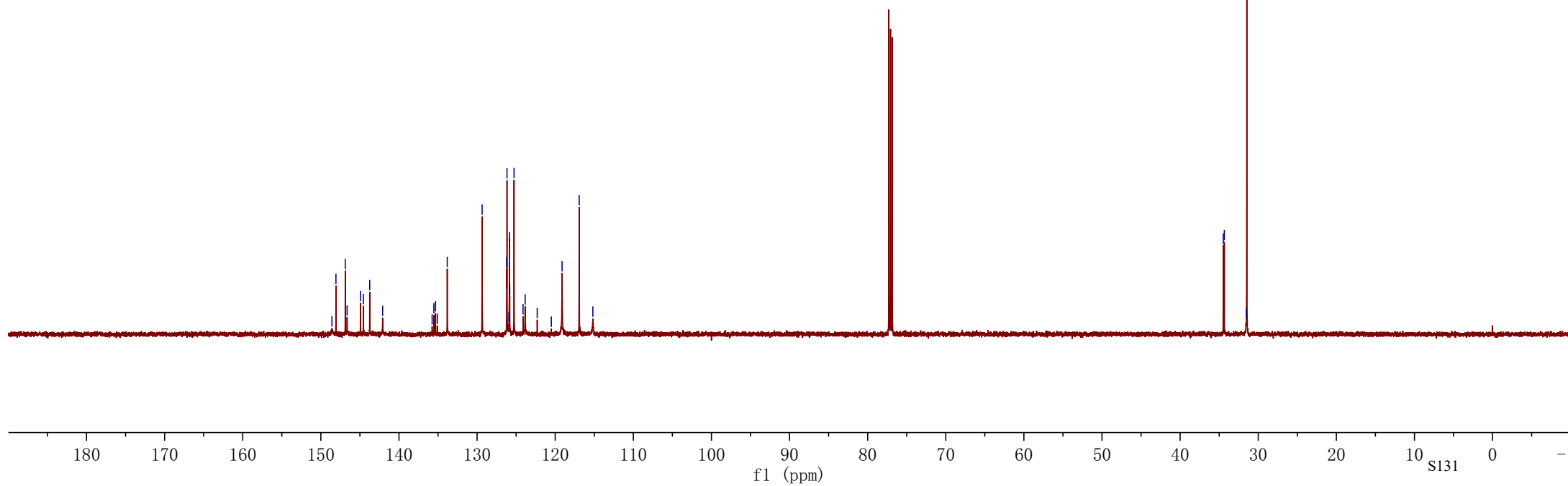


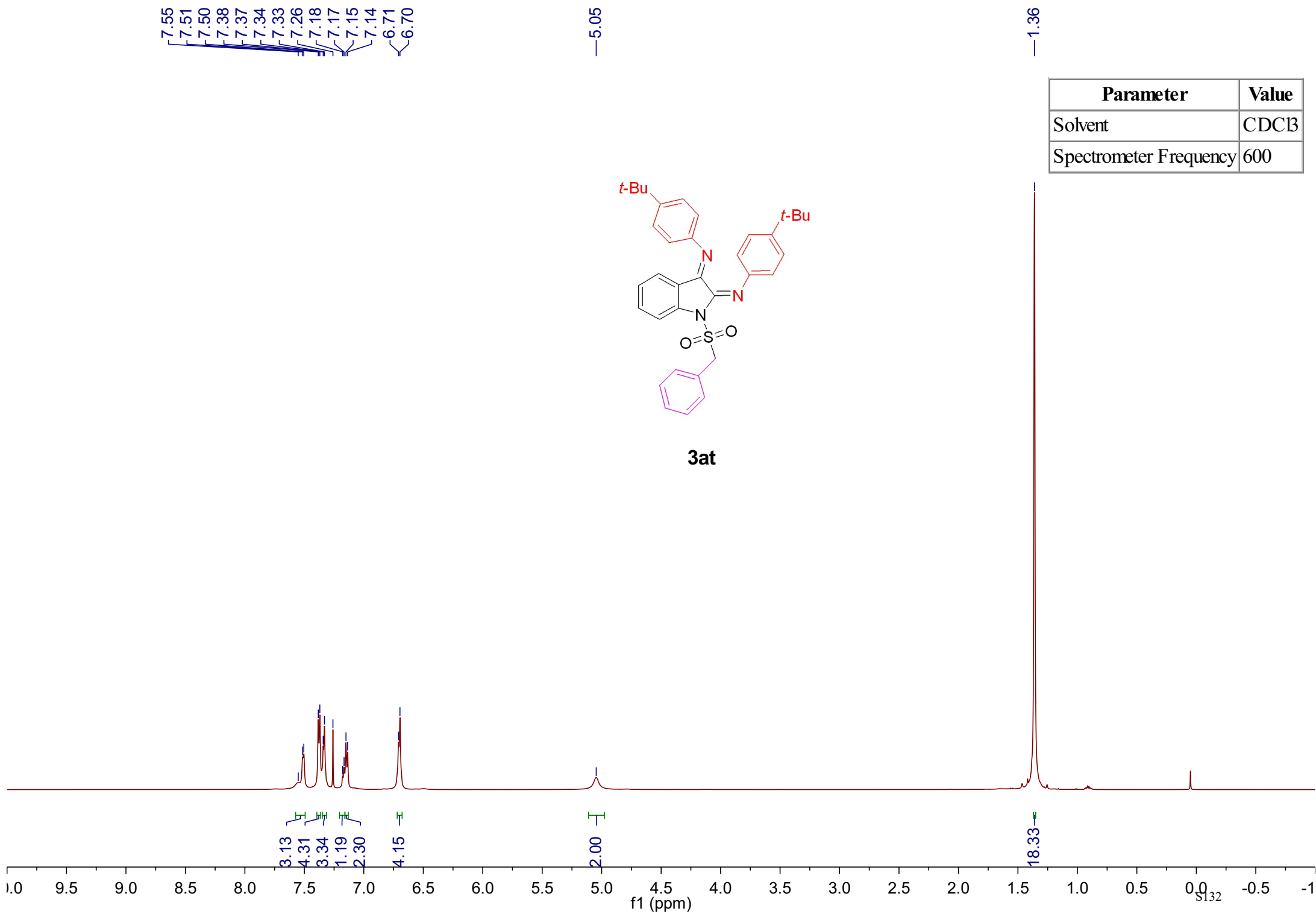
3as

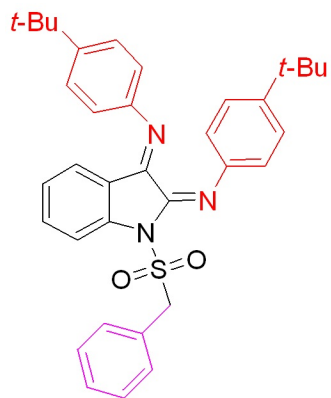
148.58
148.07
146.87
146.64
144.93
144.55
143.75
142.09
135.53
135.32
133.82
129.36
126.21
126.17
125.92
125.87
125.84
125.82
125.27
124.11
123.85
122.30
120.50
119.12
116.93
115.18

34.48
34.34
31.52
31.46

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150







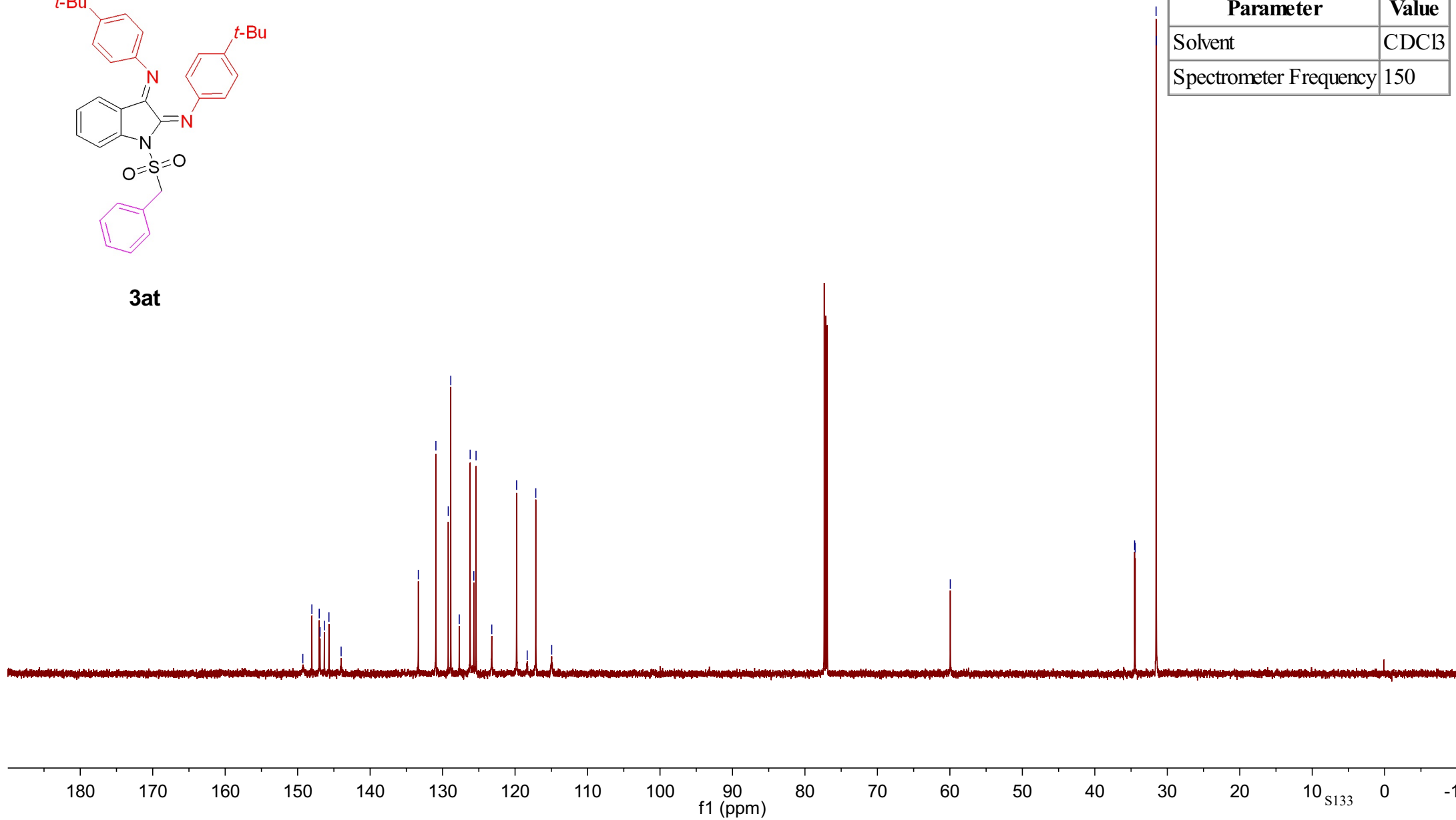
3at

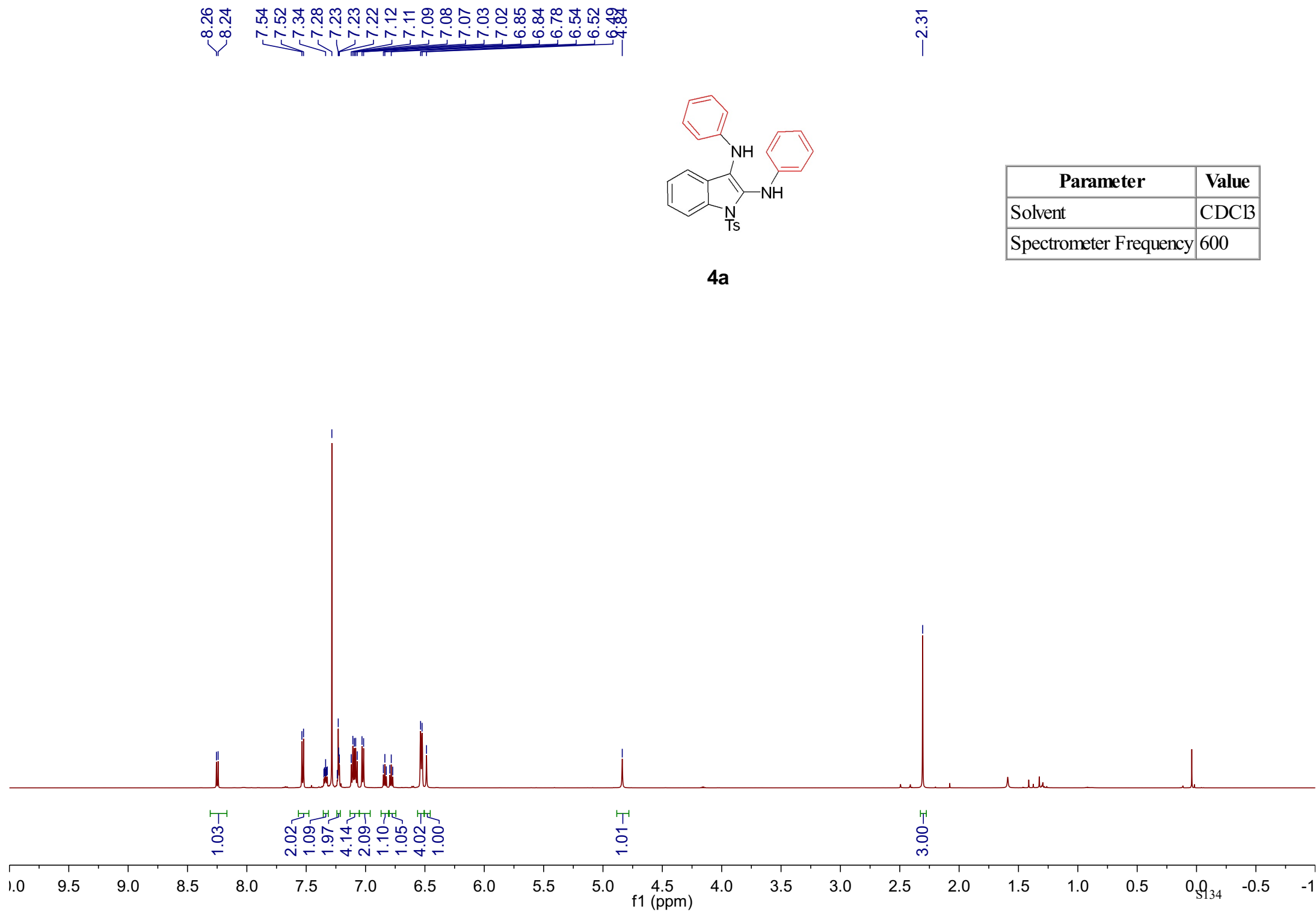
149.29
148.04
147.02
146.91
146.32
145.68
144.00
133.35
130.92
129.22
128.87
127.70
126.20
125.69
125.38
123.21
119.78
118.33
117.14
114.94

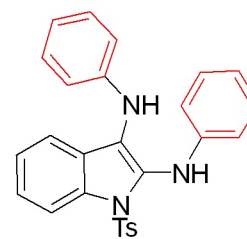
—59.95

34.51
34.43
31.55
31.52

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150

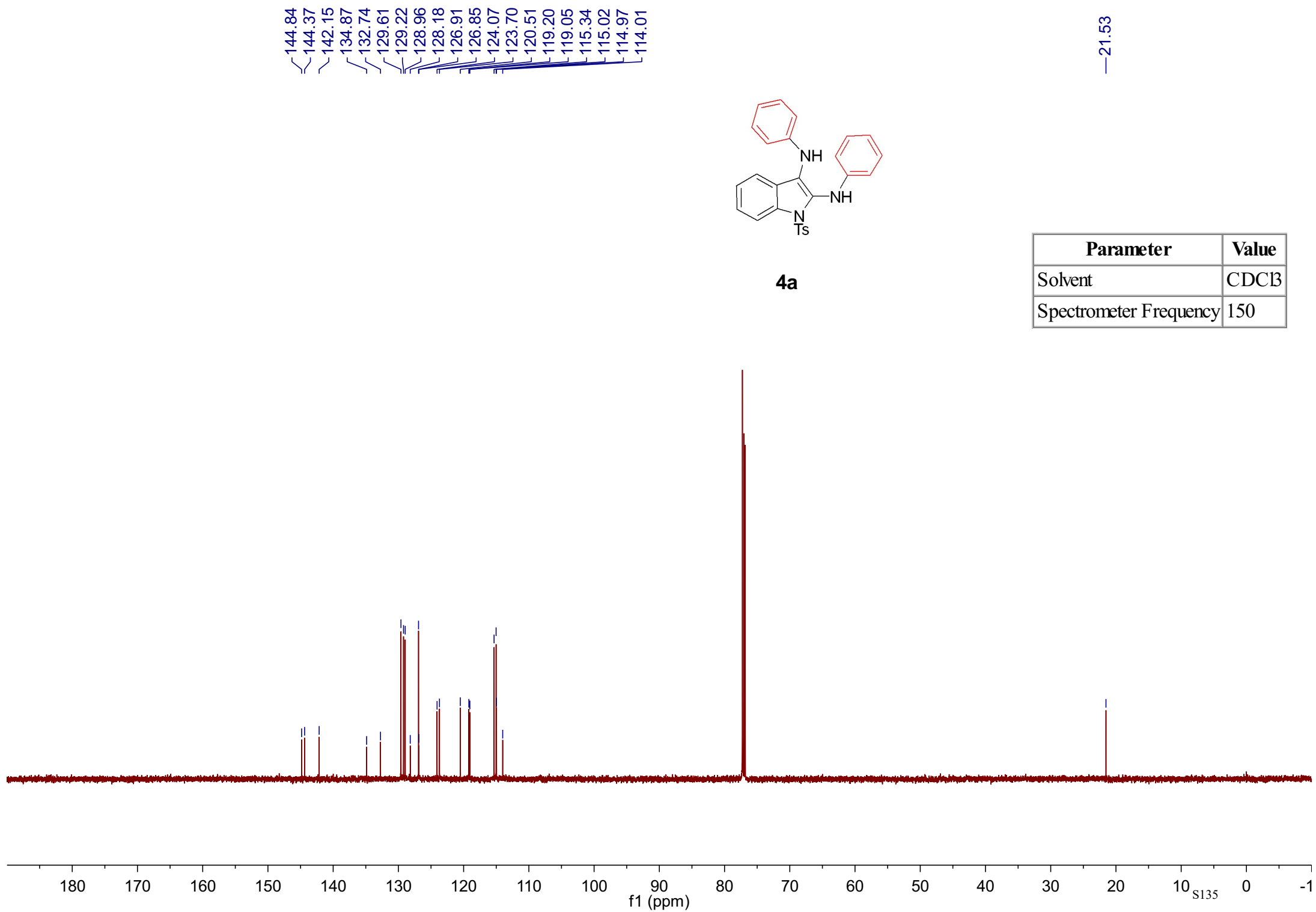






4a

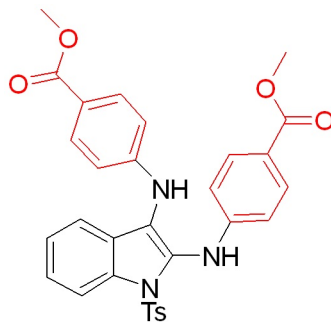
Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150



8.24
8.23
7.76
7.75
7.69
7.67
7.48
7.47
7.22
7.21
7.21
6.98
6.97
6.77
6.49
6.47
6.43
6.42

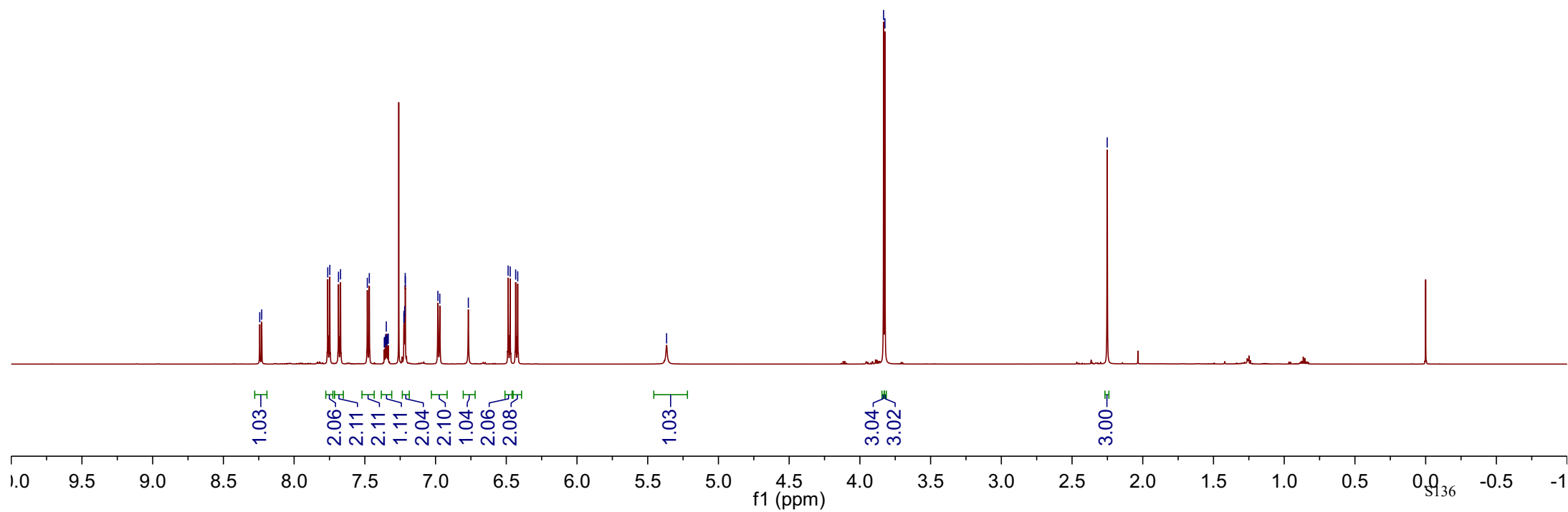
3.83
3.82

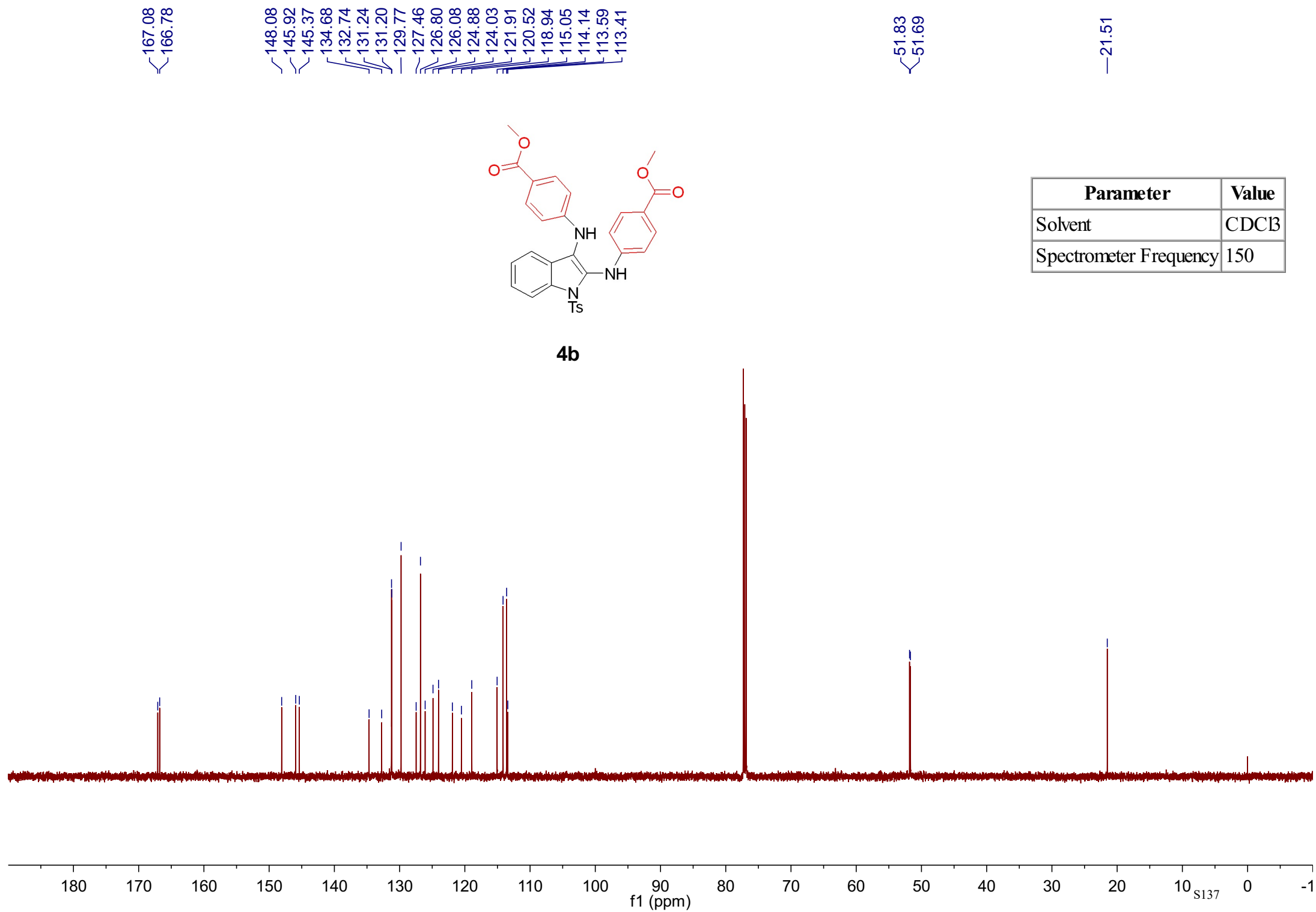
2.25

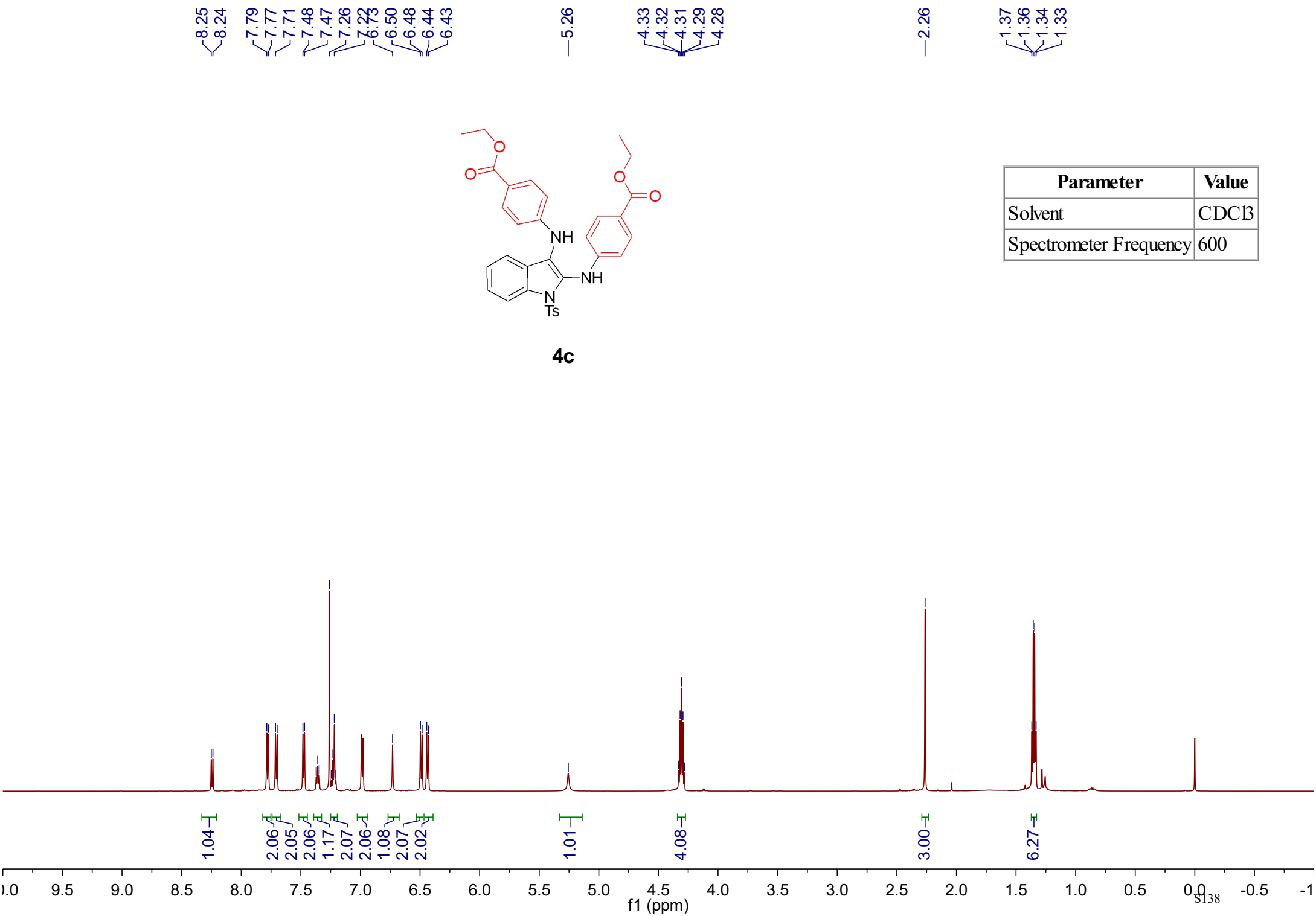


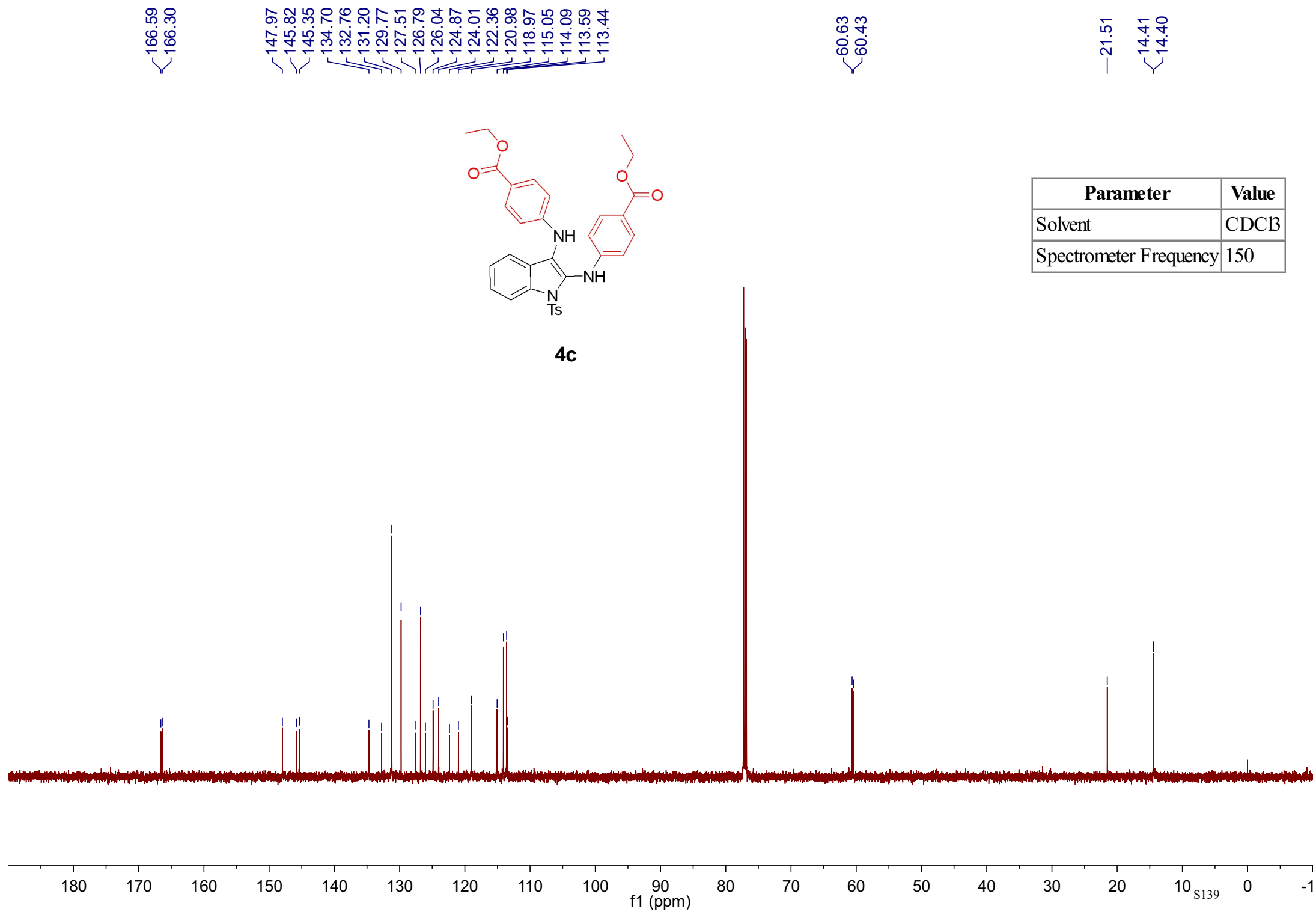
4b

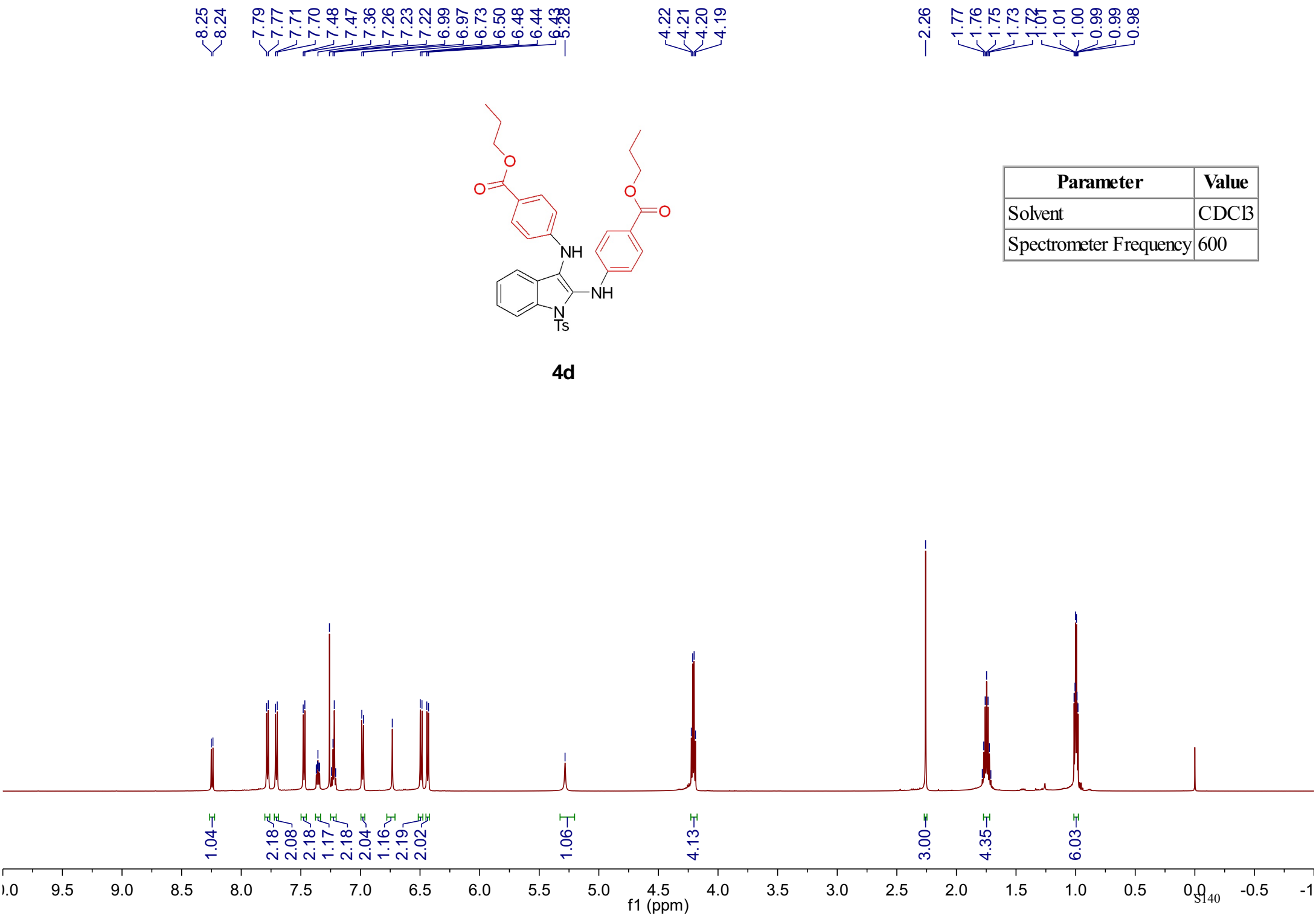
Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	600











166.66
166.37

148.00
145.86
145.34

134.70
132.78
131.20

129.77
127.52
126.79

126.05
124.87
124.02

122.34
120.98
118.98

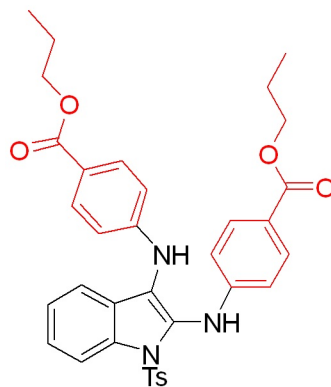
115.06
114.10
113.60

113.51

66.23
66.04

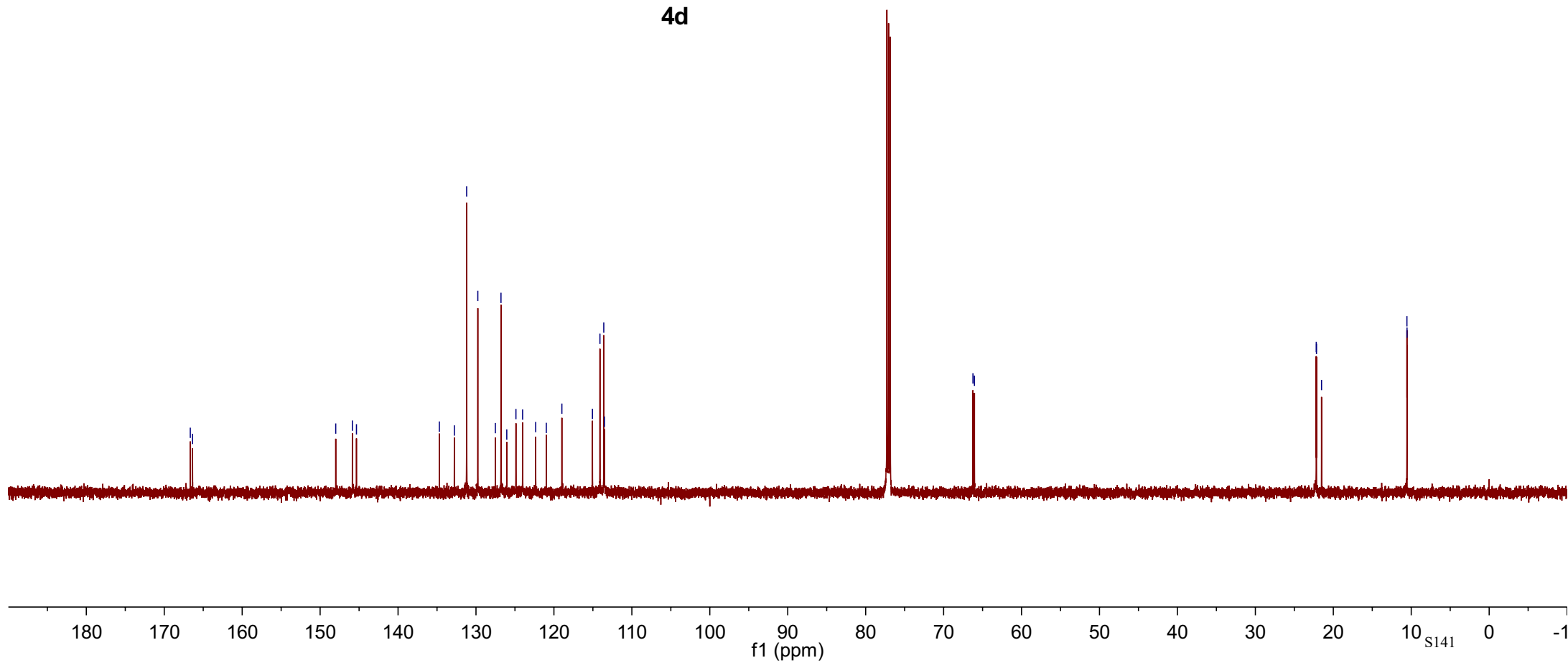
22.19
22.16
21.51

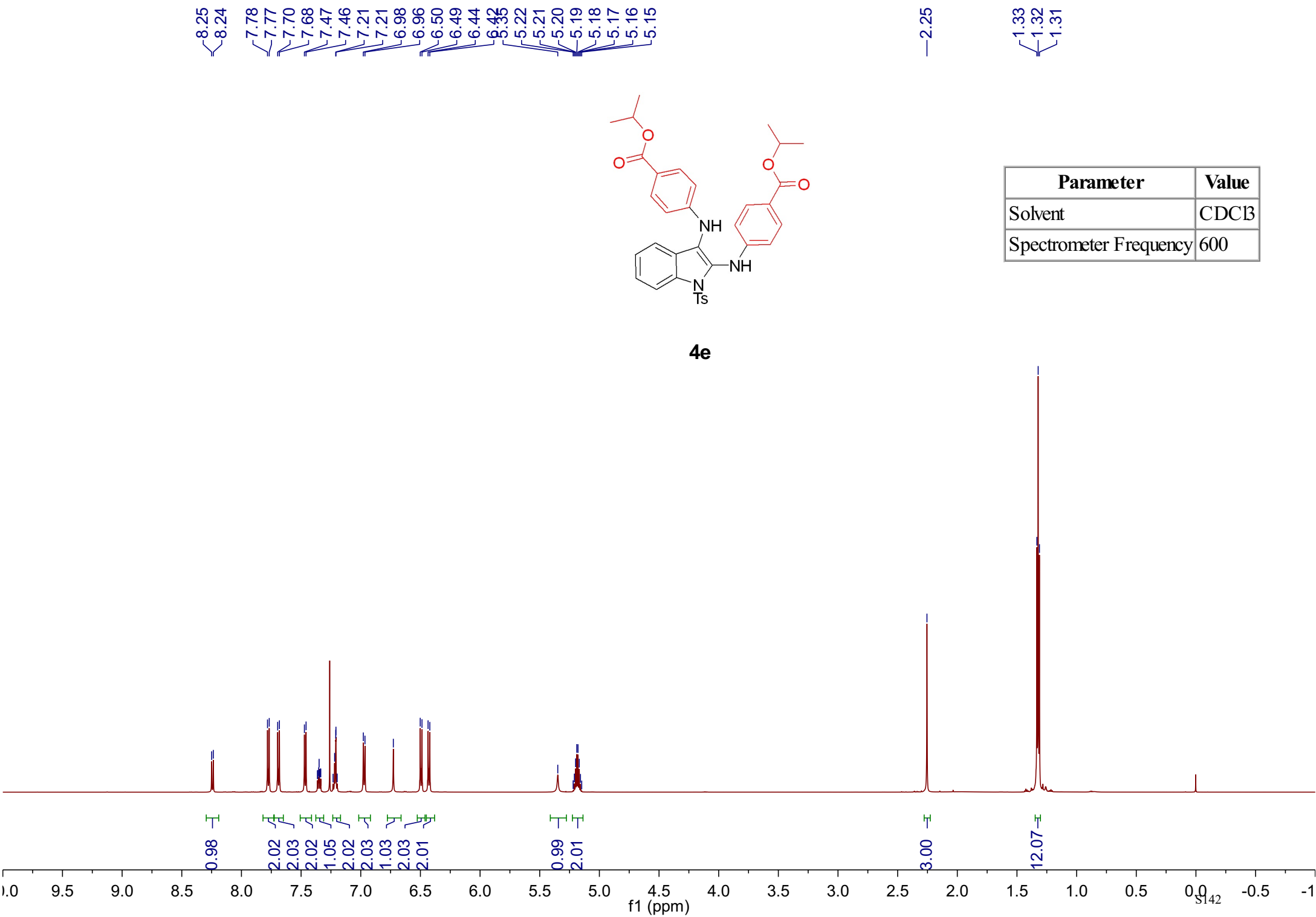
10.55
10.54



4d

Parameter	Value
Solvent	CDCl3
Spectrometer Frequency	150





166.13
165.84

147.96
145.87
145.30

134.70
132.80
131.16

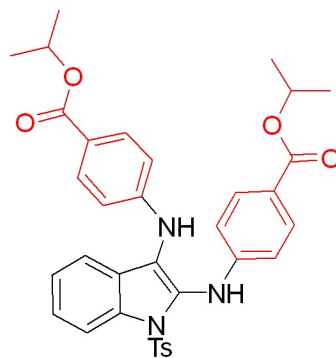
129.76
127.51
126.78

126.04
124.85
123.98

122.67
121.33
119.05
115.06
113.99
113.70
113.57

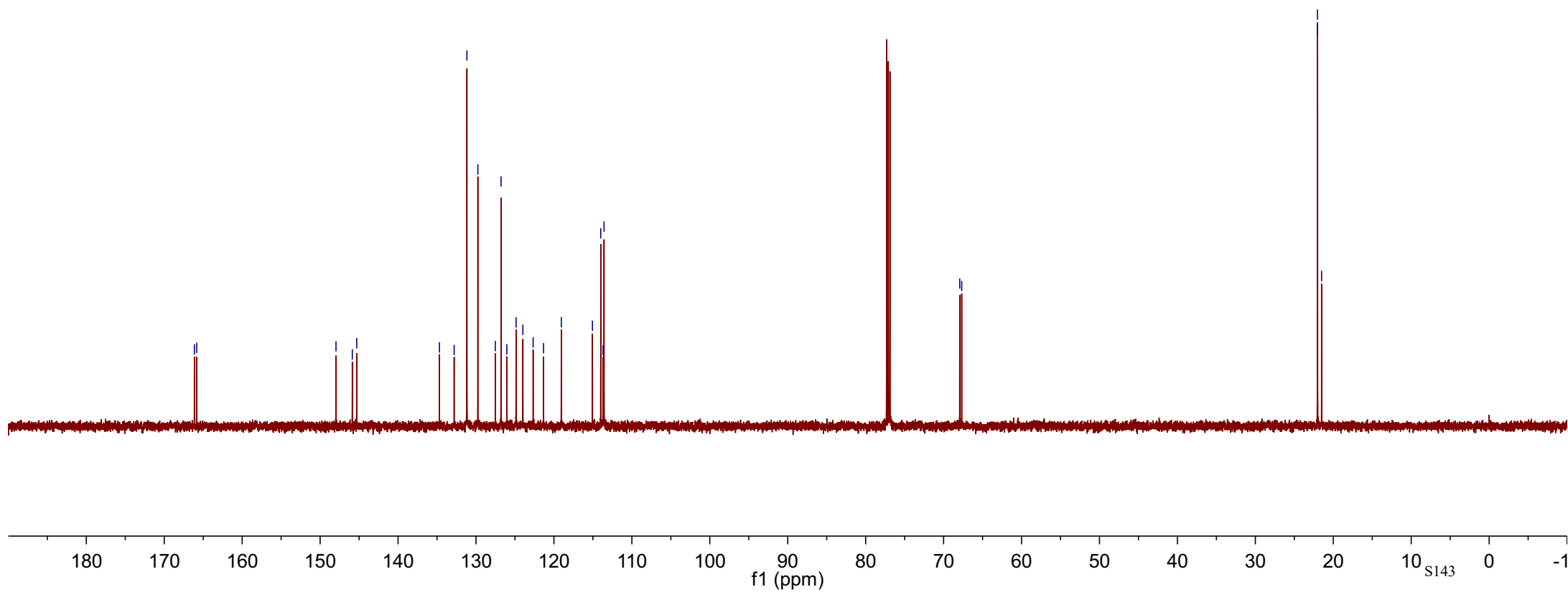
67.93
67.68

22.03
22.01
21.50

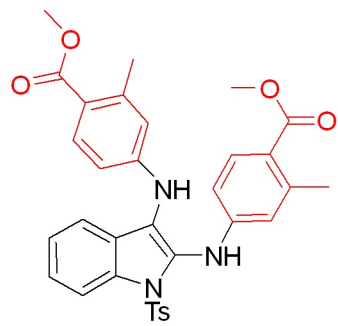


4e

Parameter	Value
Solvent	CDCI3
Spectrometer Frequency	150

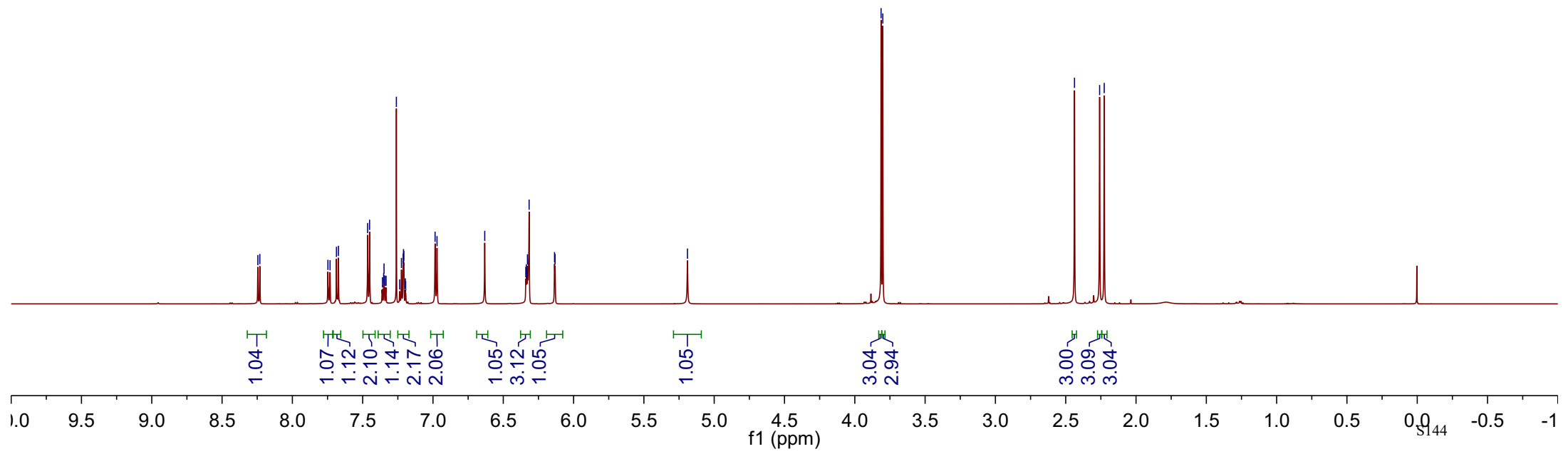


8.25
8.23
7.69
7.67
7.46
7.45
7.26
7.21
6.99
6.97
6.63
6.34
6.34
6.33
6.33
6.32
6.14
6.13
— 5.19
3.81
3.80
2.44
2.26
2.23



4f

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	600



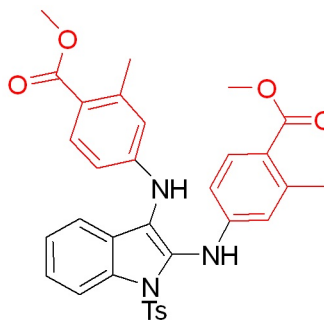
167.61
167.42

147.36
145.24
144.94
142.78
142.74
134.68
132.80
132.78
132.65
129.68
127.64
126.83
126.31
124.75
124.05
121.05
119.62
119.06
117.25
117.00
115.16
113.55
111.92
111.18

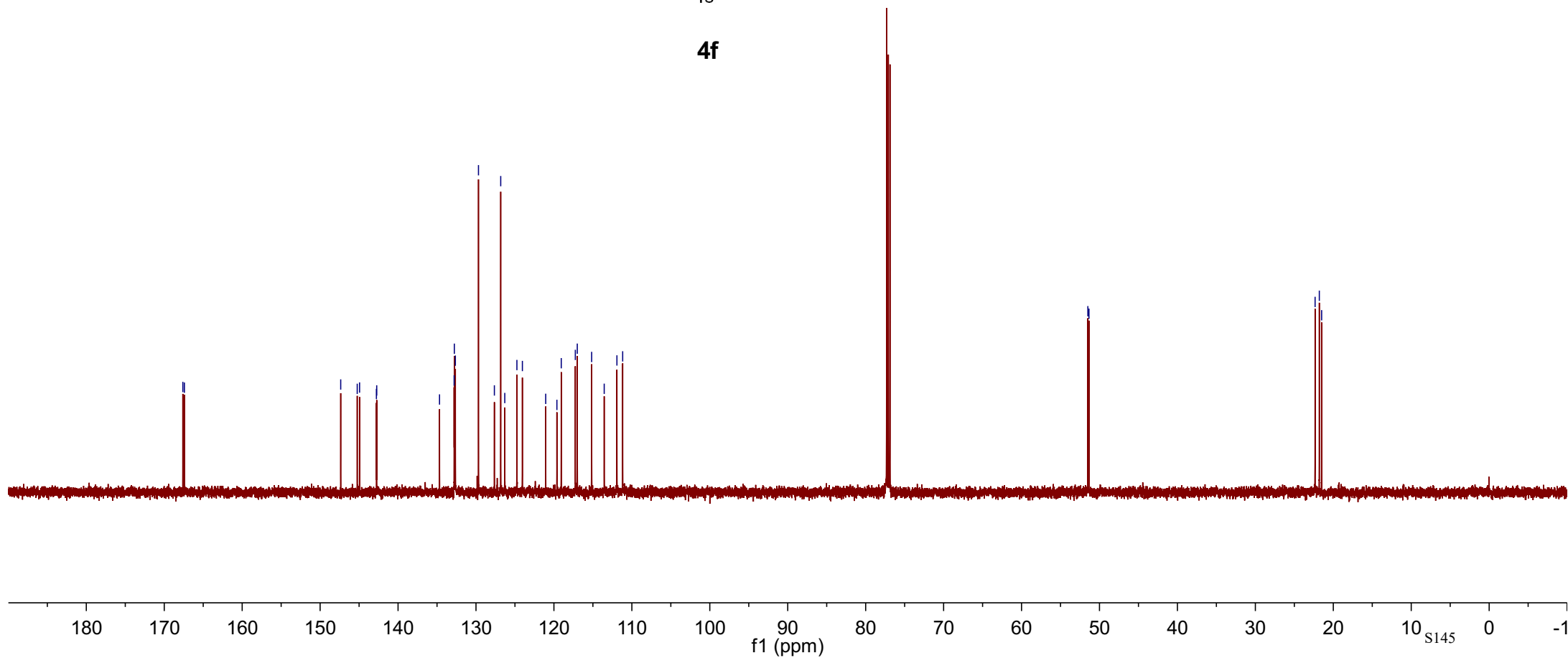
51.49
51.35

22.33
21.79
21.50

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150



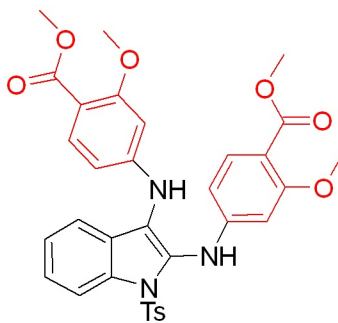
4f



8.23
8.21
7.63
7.56
7.43
7.42
7.26
7.22
7.22
6.96
6.95
6.71
6.71
6.12
6.12
6.11
6.05
6.04
5.88
5.73

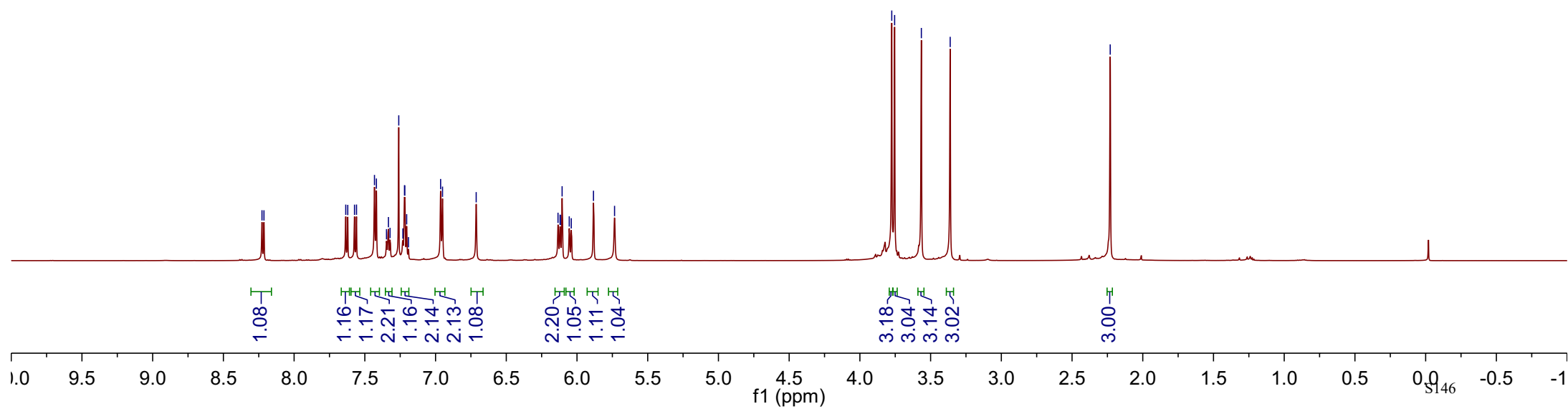
3.77
3.75
3.57
3.36

2.23



4g

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	600



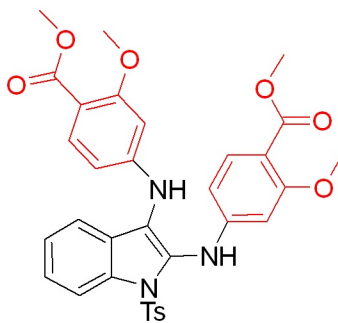
166.13
166.05
161.38
161.31

149.36
147.30
145.39

134.58
133.82
133.71
132.85
129.67
126.95
126.82
125.00
124.00
119.46
115.24
111.08
109.77
106.40
99.98
97.73

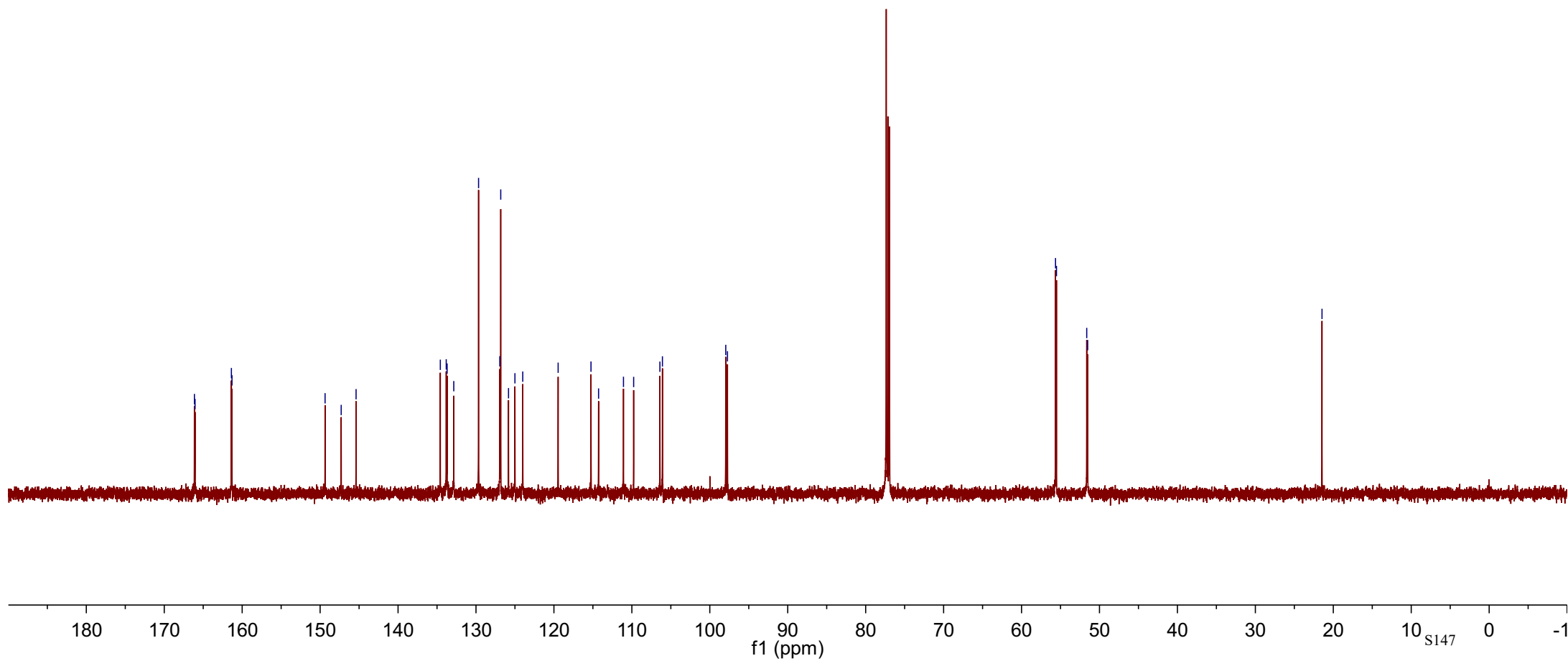
55.64
55.52
51.64
51.51

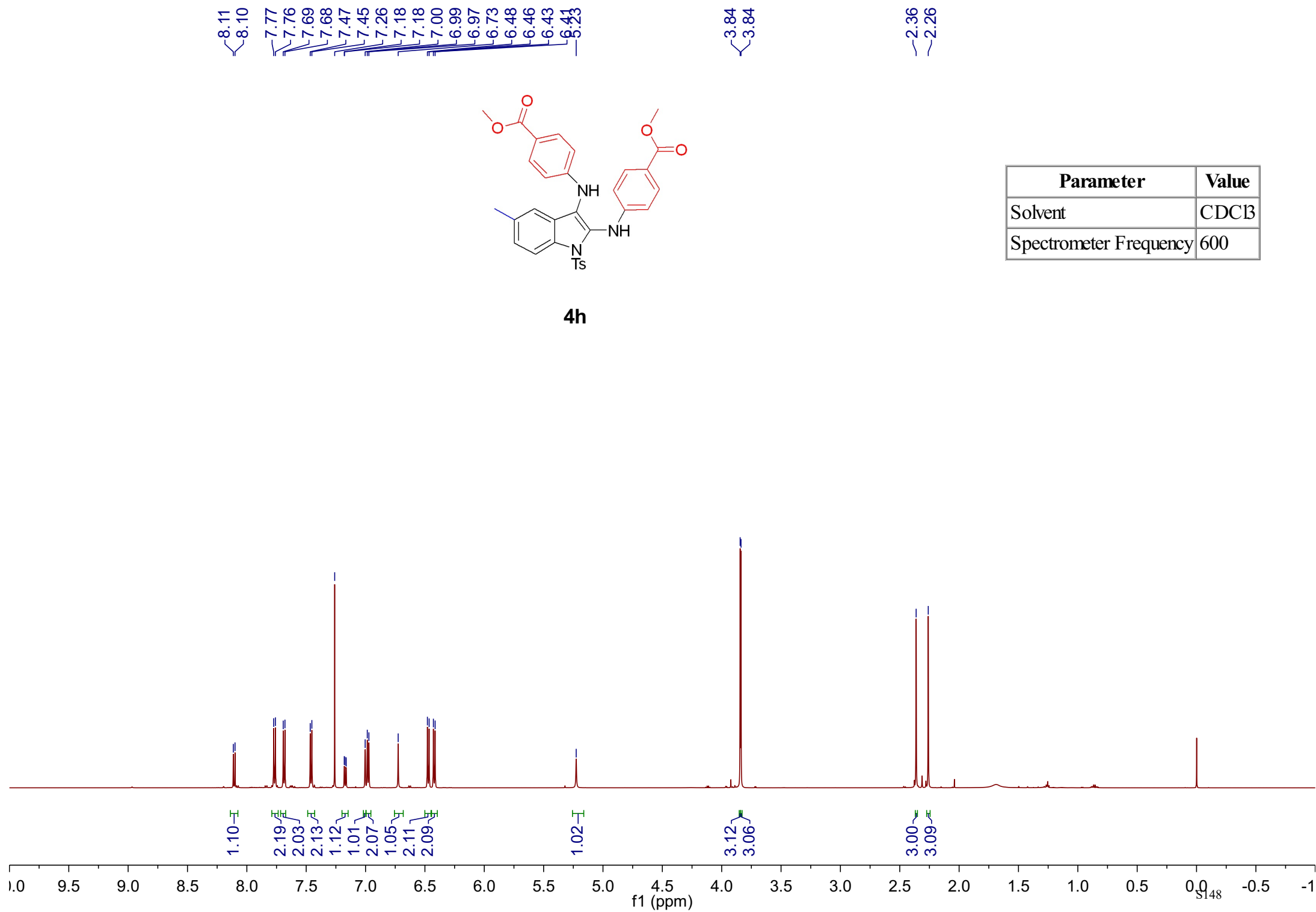
21.45

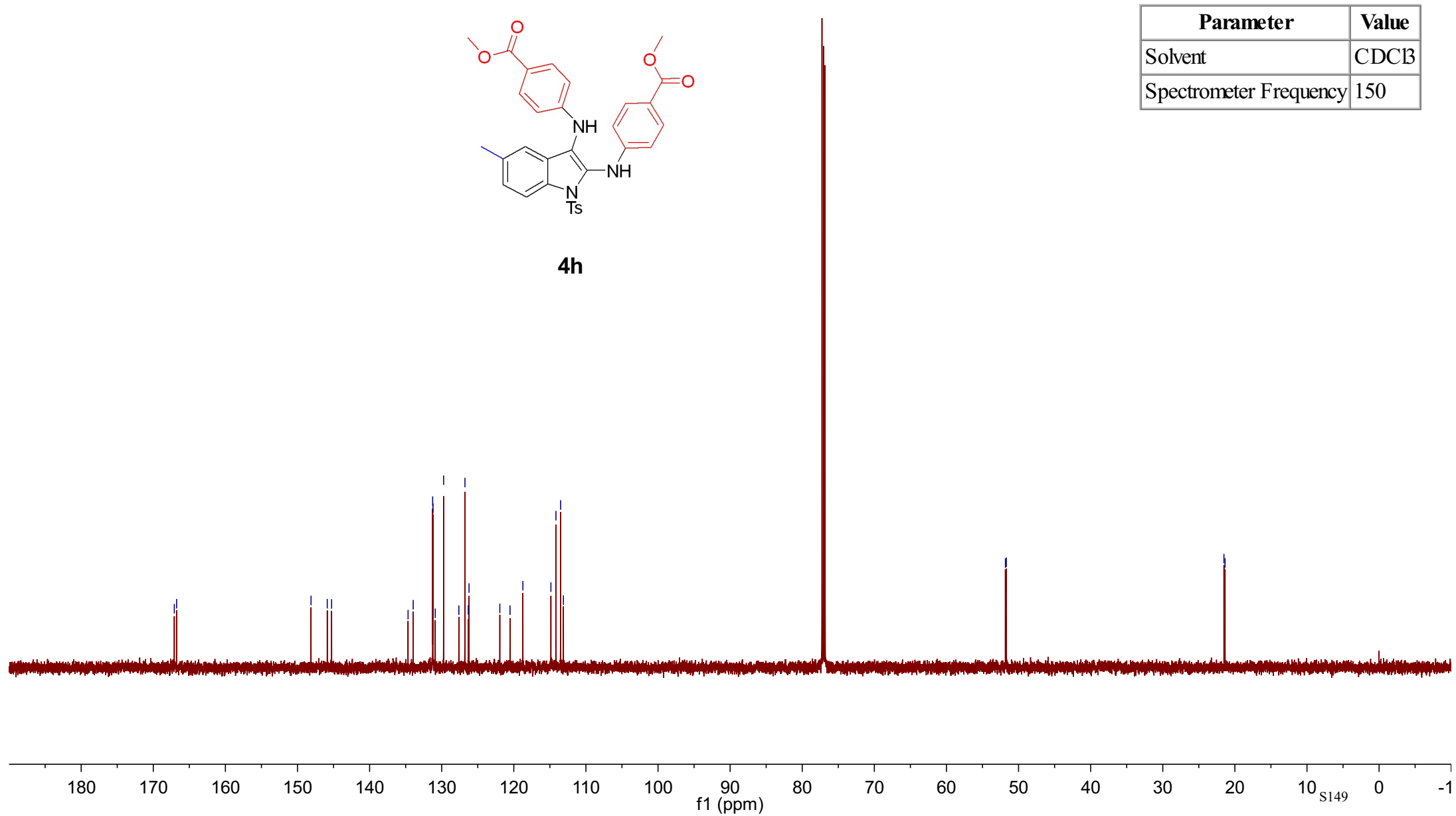


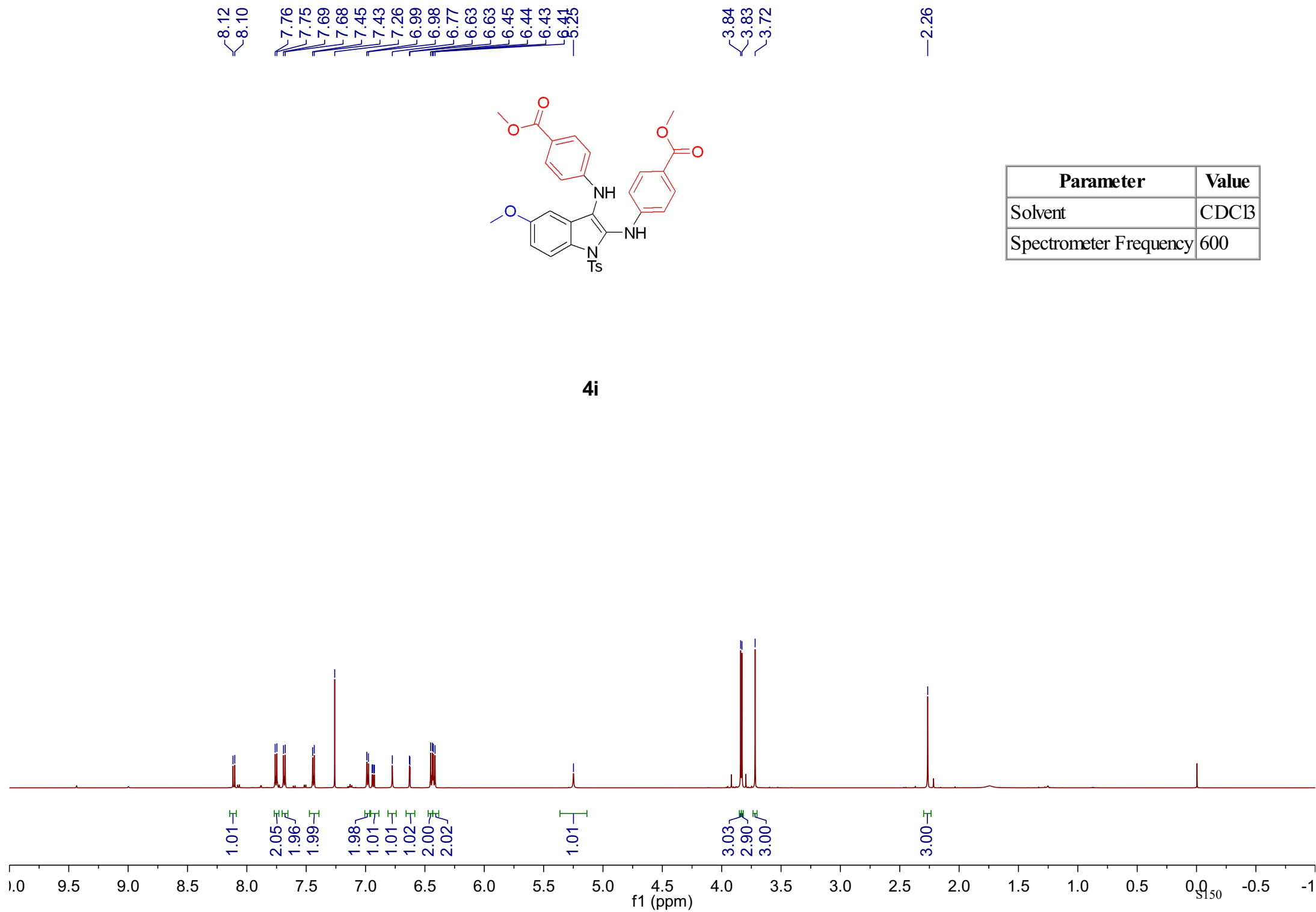
4g

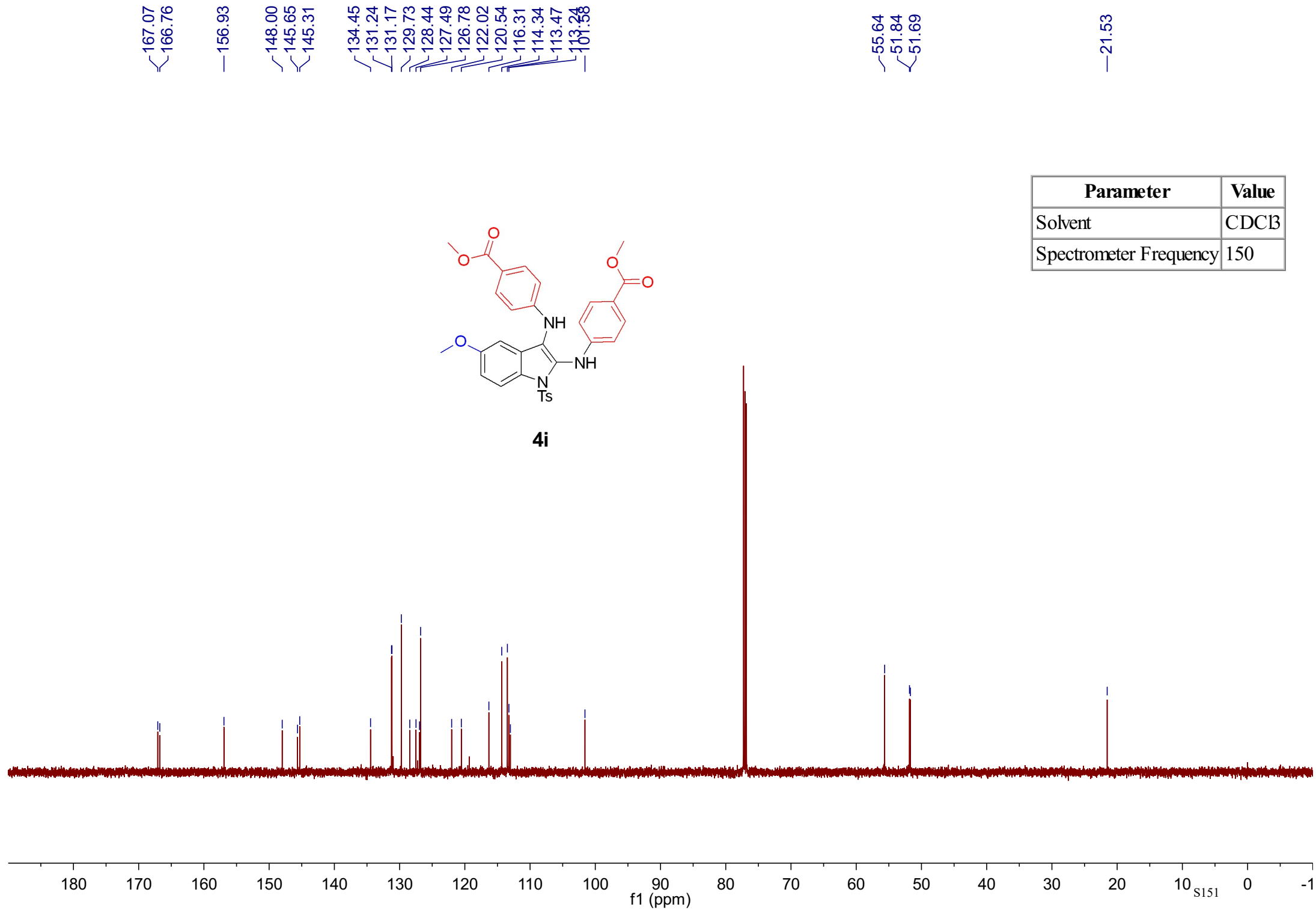
Parameter	Value
Solvent	CDCI3
Spectrometer Frequency	150

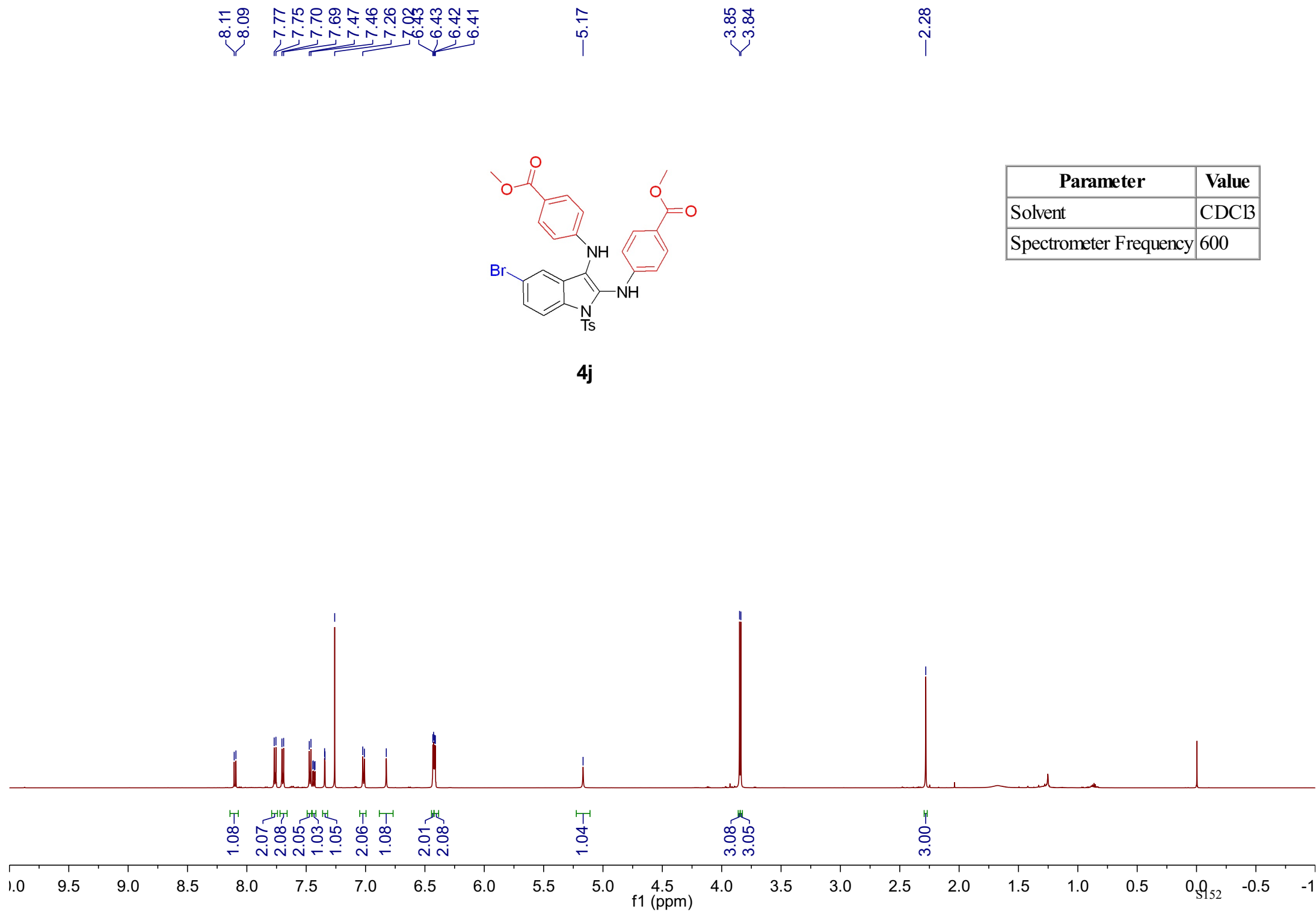


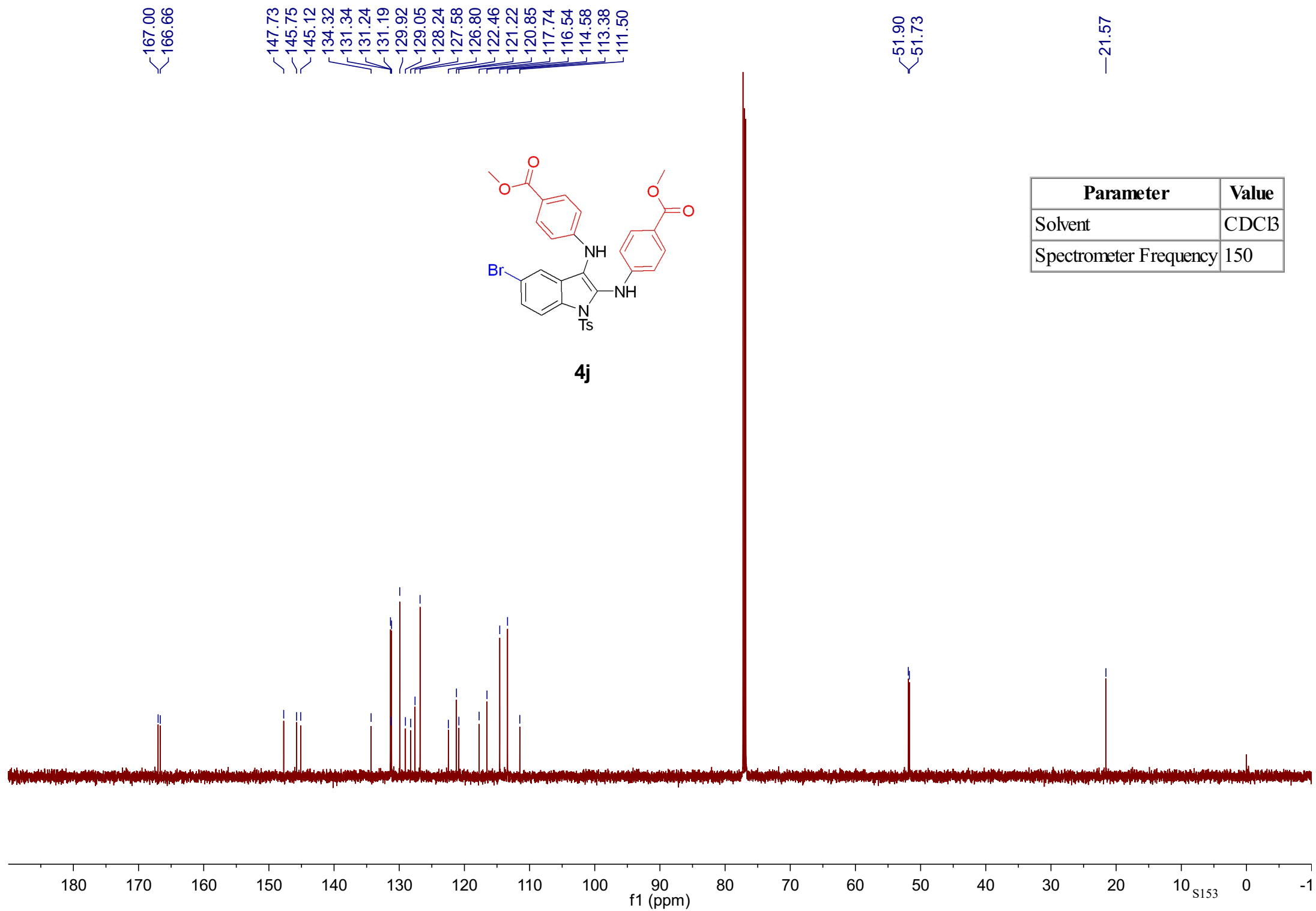






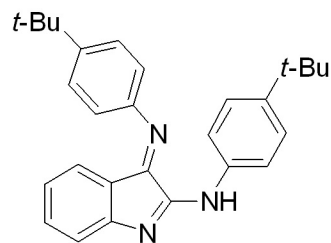






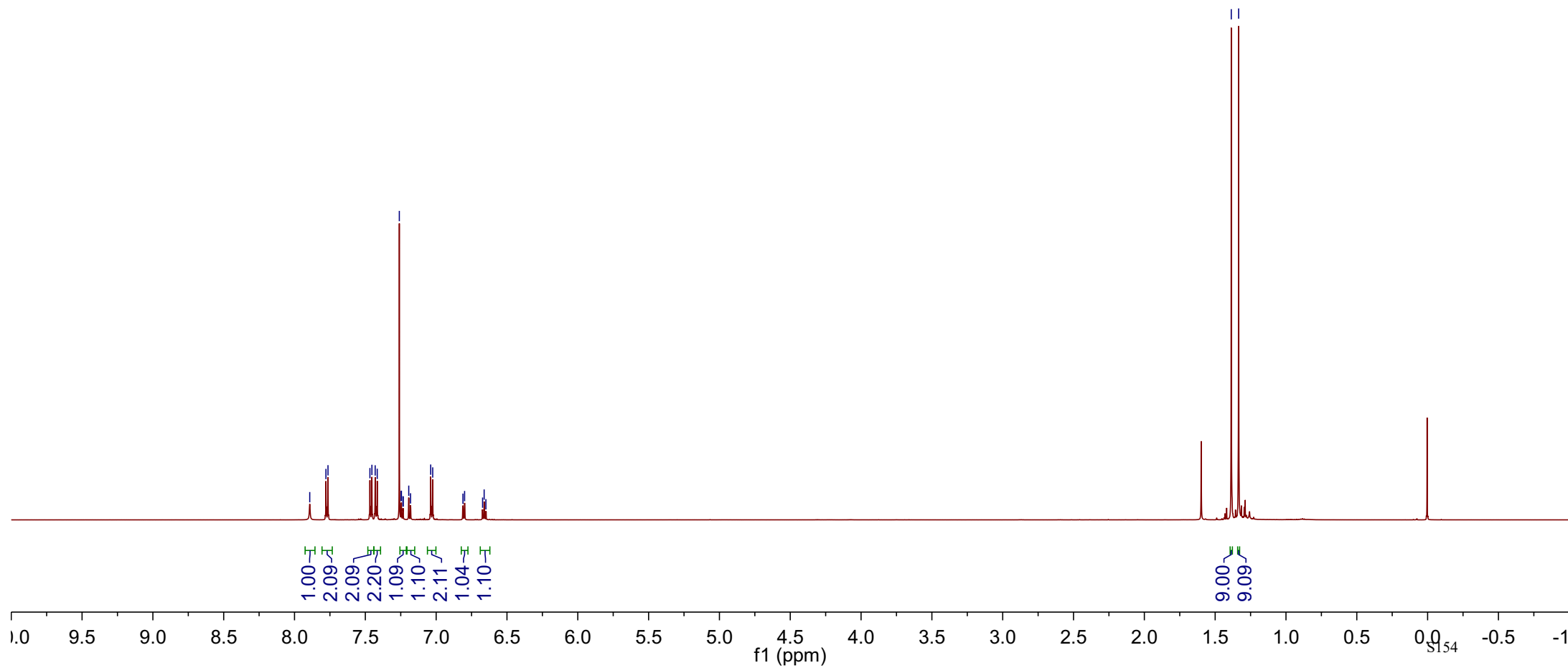
7.89
7.78
7.76
7.47
7.45
7.43
7.42
7.26
7.25
7.25
7.23
7.23
7.19
7.18
7.04
7.02
6.81
6.80
6.67
6.66
6.65

1.39
1.33



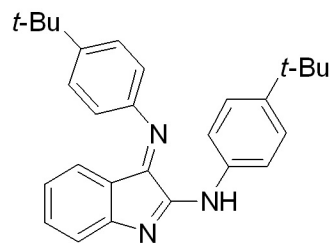
5

Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	600



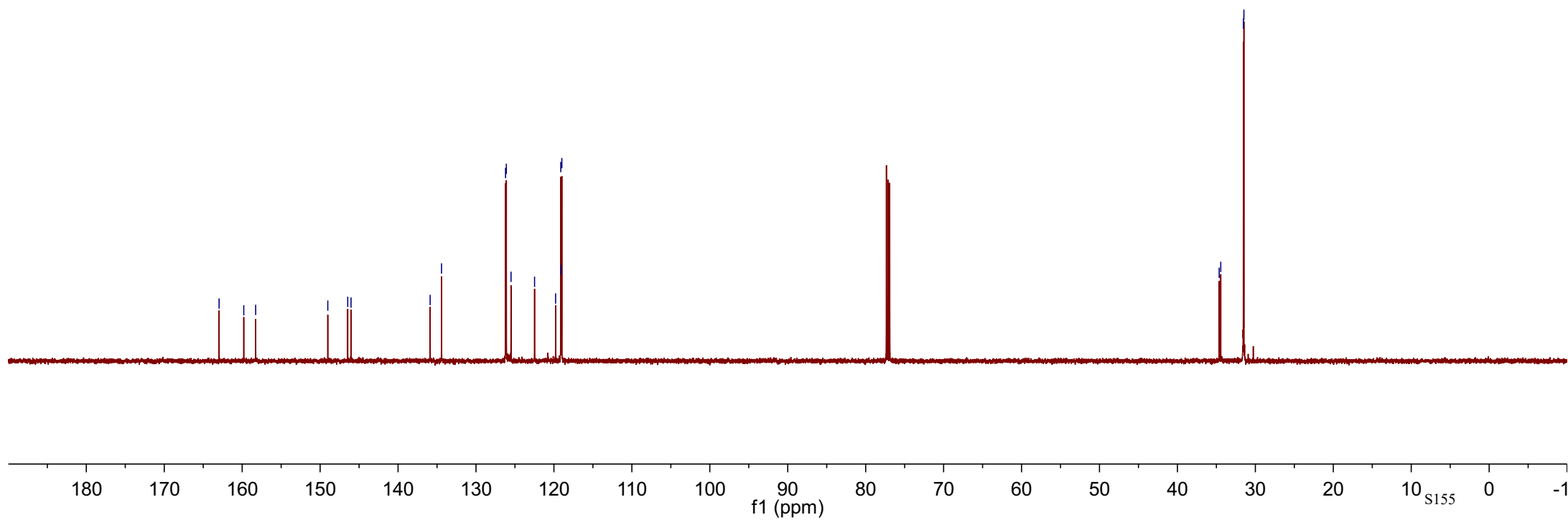
162.96
159.79
158.27
149.01
146.46
146.03
135.88
134.42
126.21
126.11
125.51
122.48
119.78
119.12
119.08
118.98

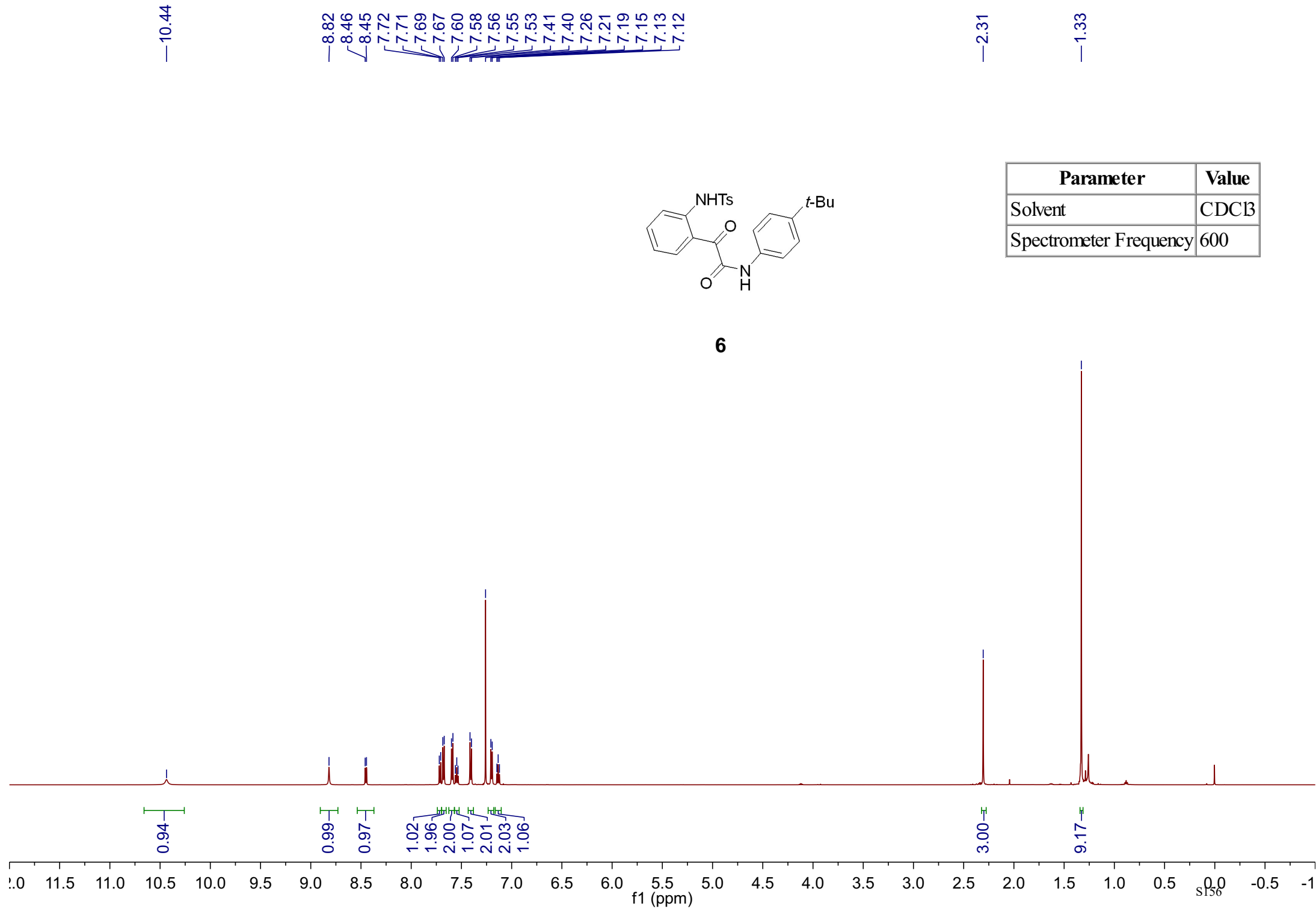
34.64
34.44
31.53
31.47

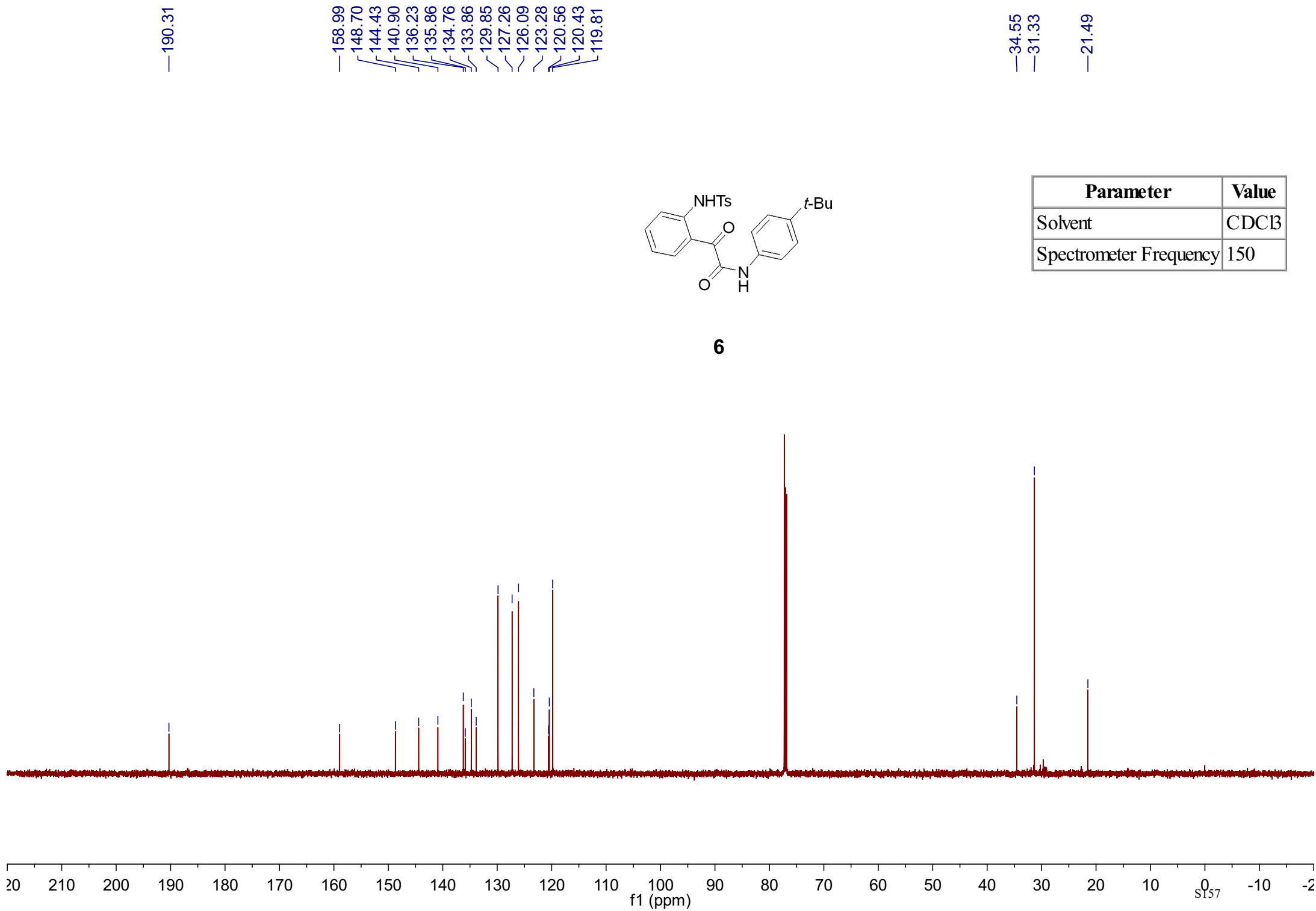


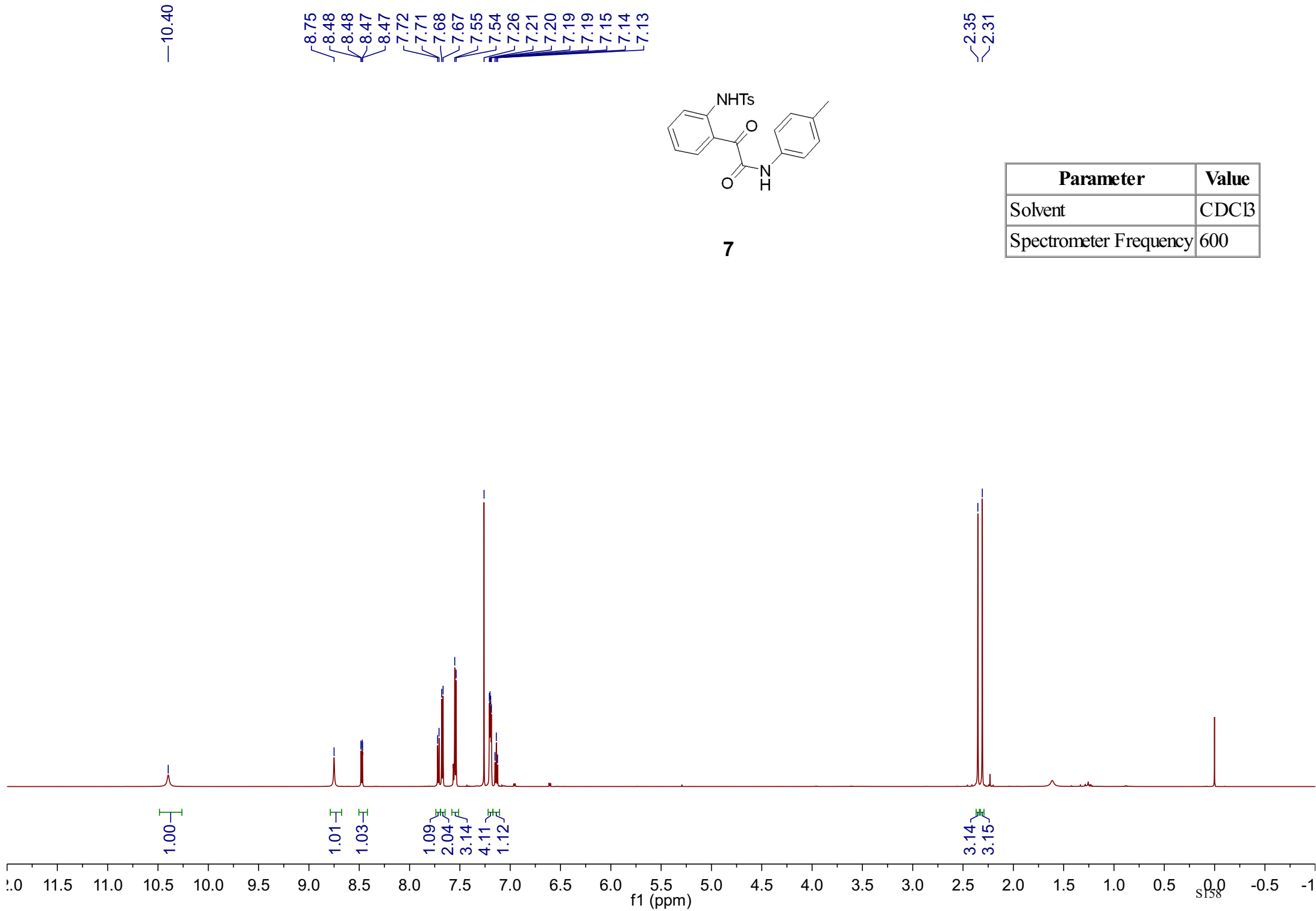
5

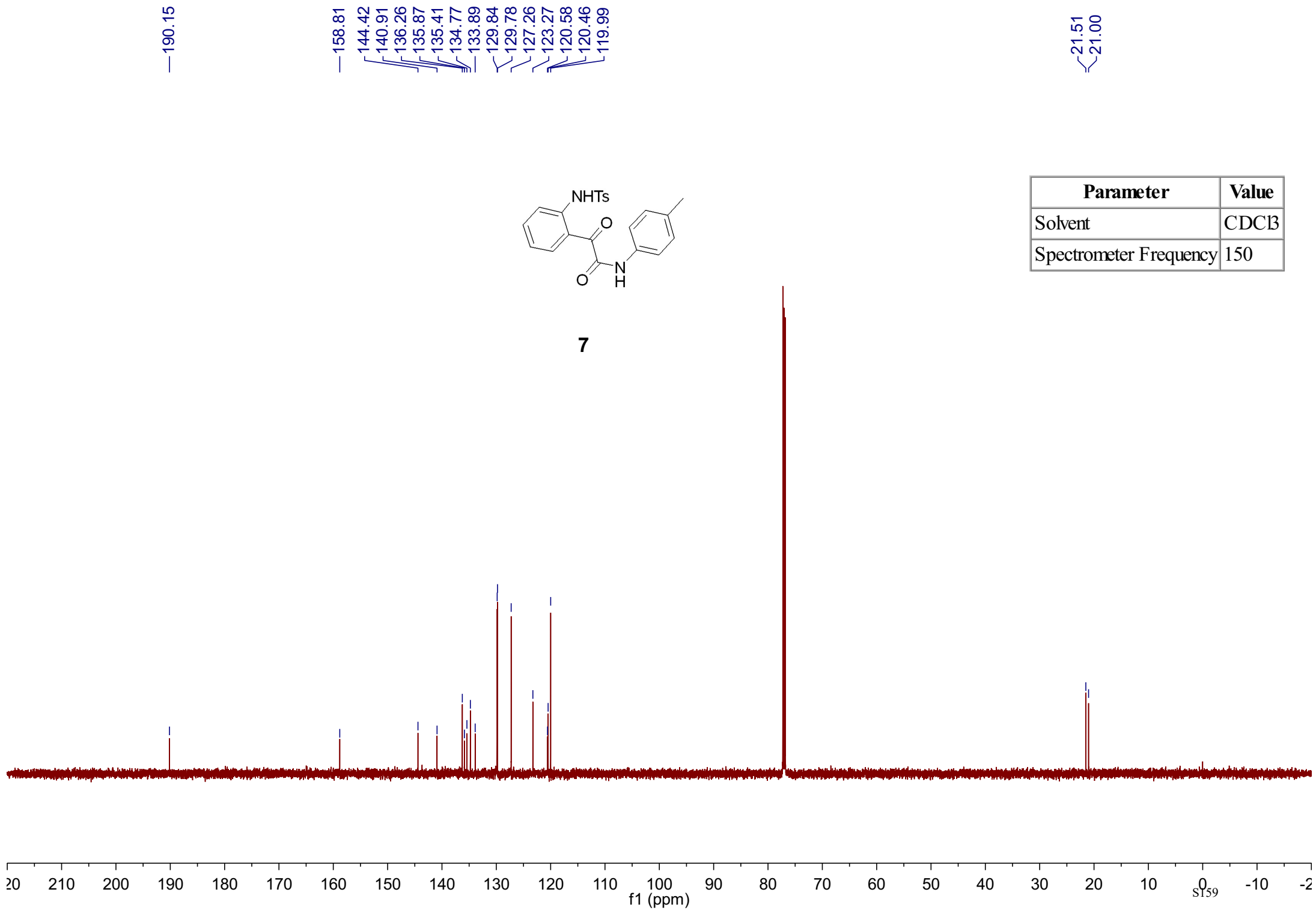
Parameter	Value
Solvent	CDCl ₃
Spectrometer Frequency	150

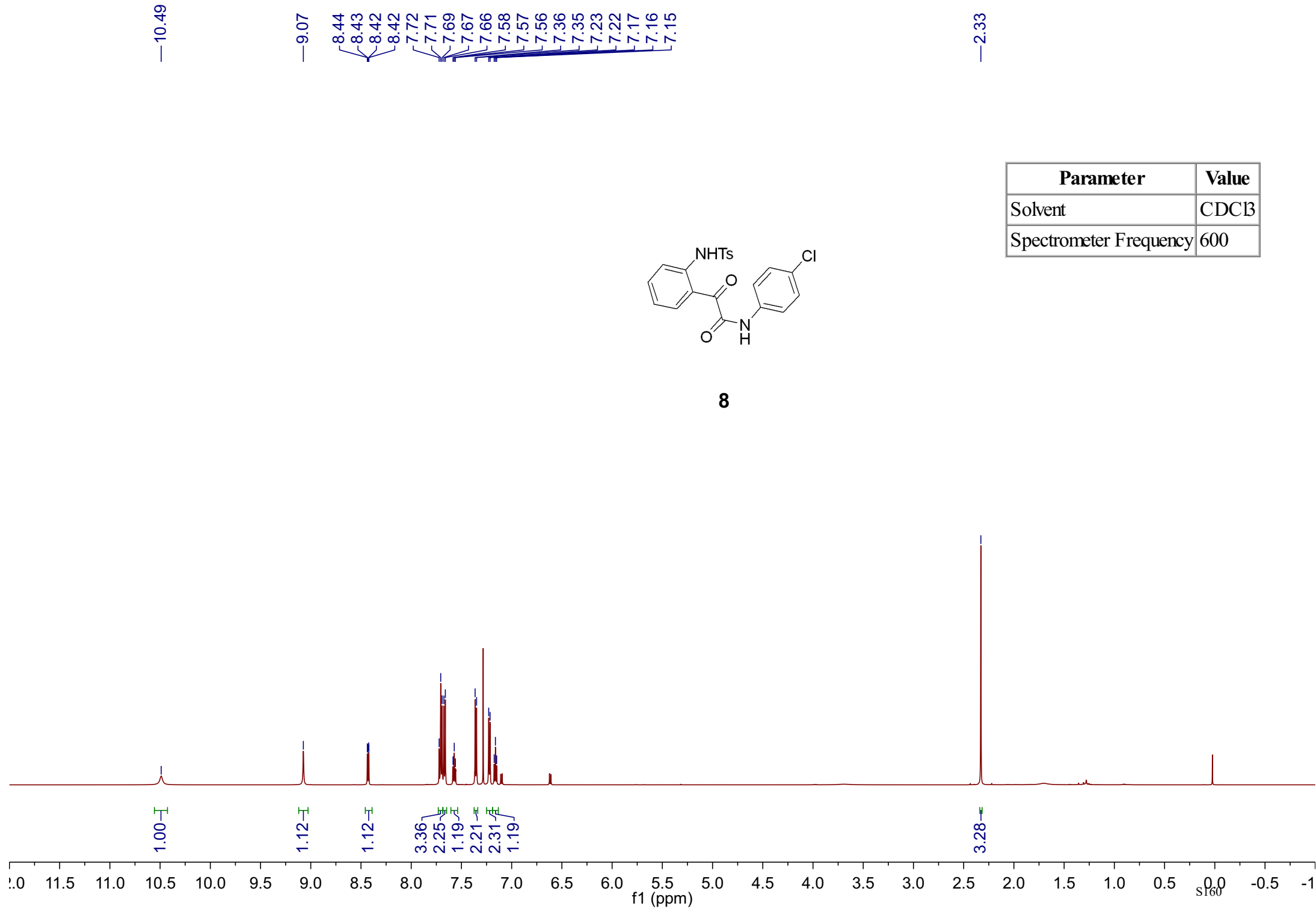


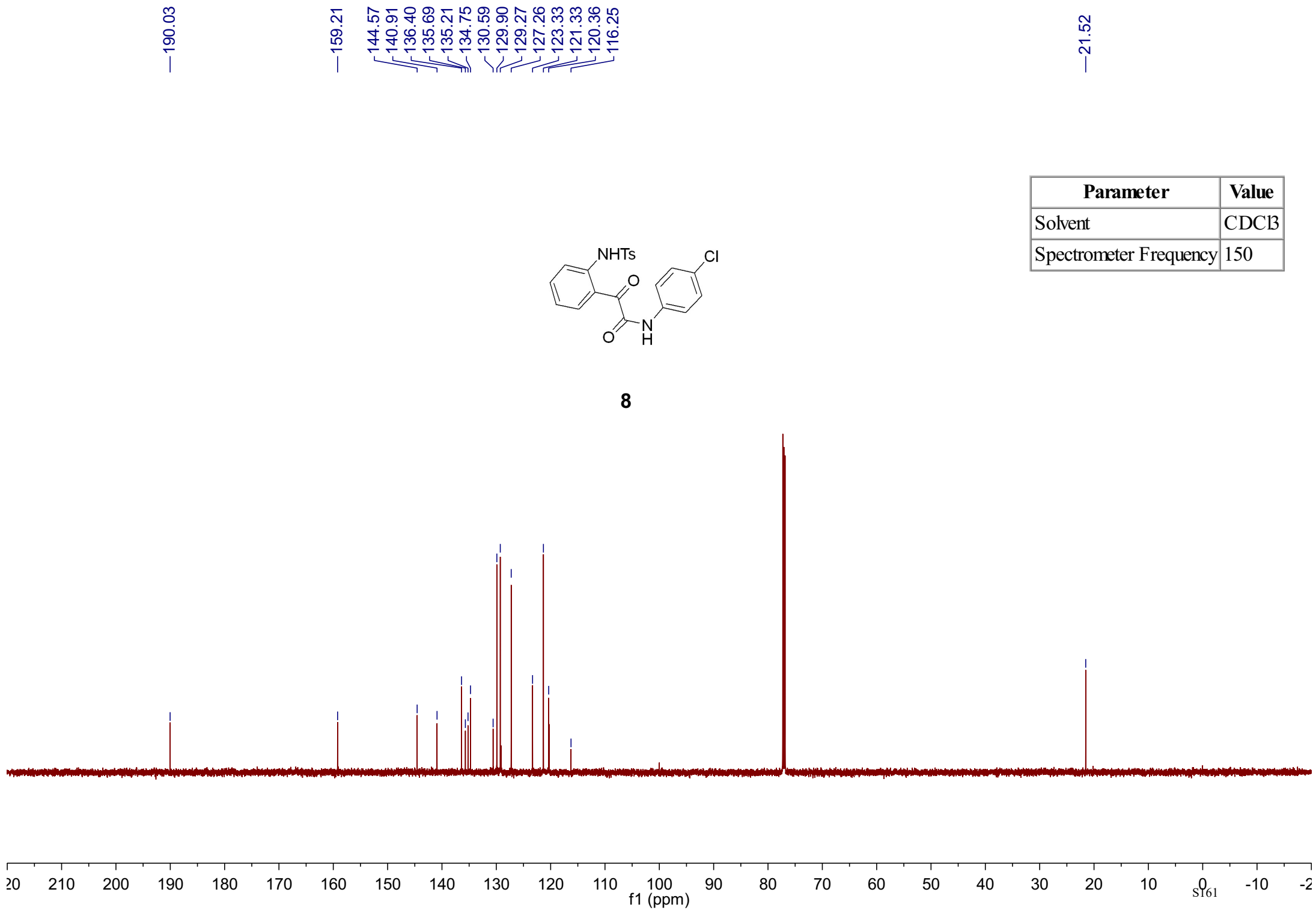


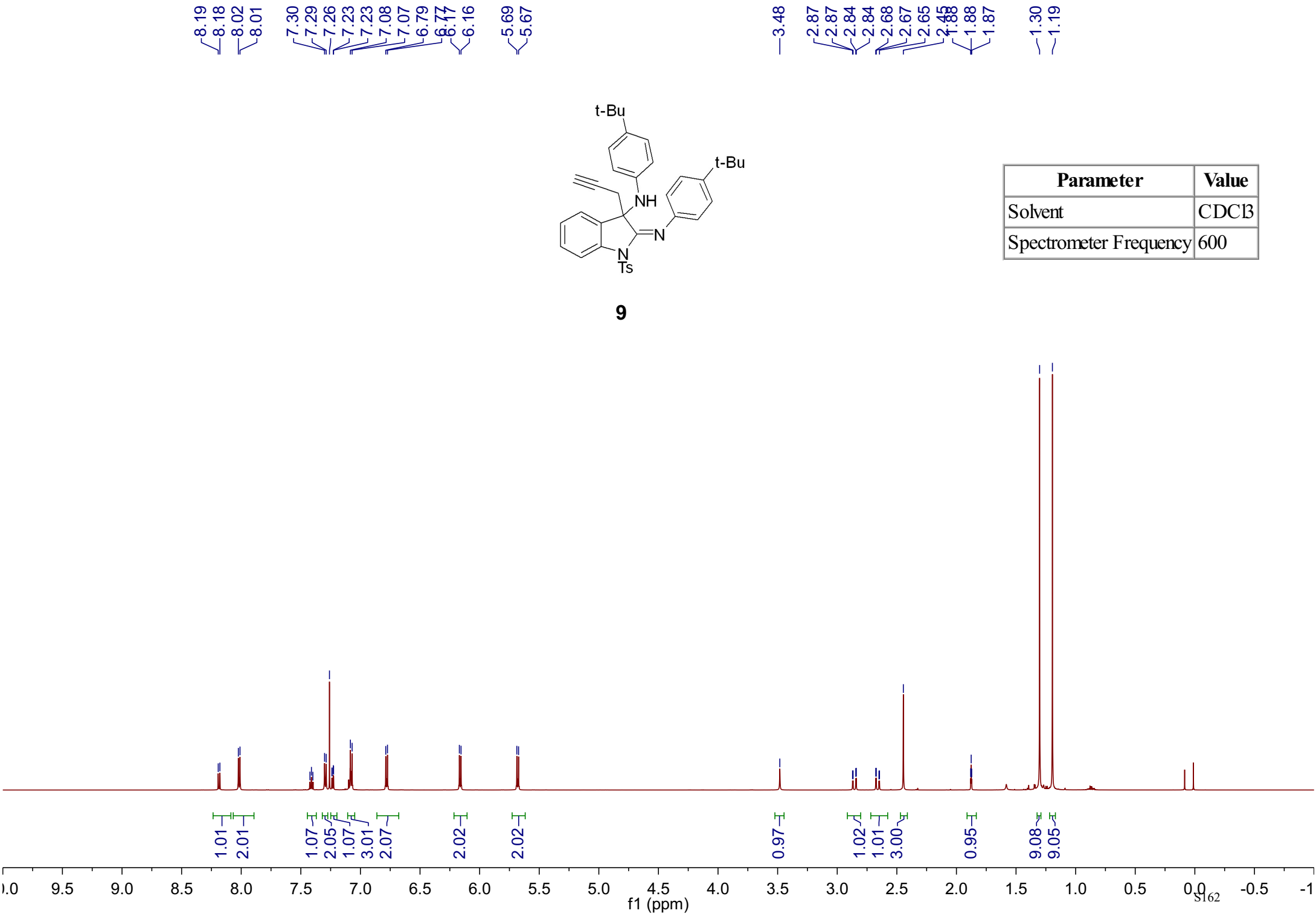








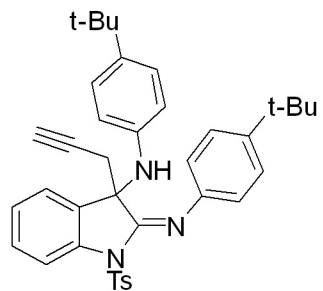




— 154.74
 — 145.68
 — 142.81
 — 141.44
 — 141.31
 — 130.13
 — 129.45
 — 129.00
 — 125.39
 — 124.86
 — 124.38
 — 123.74
 — 118.08
 — 114.05
 — 114.01

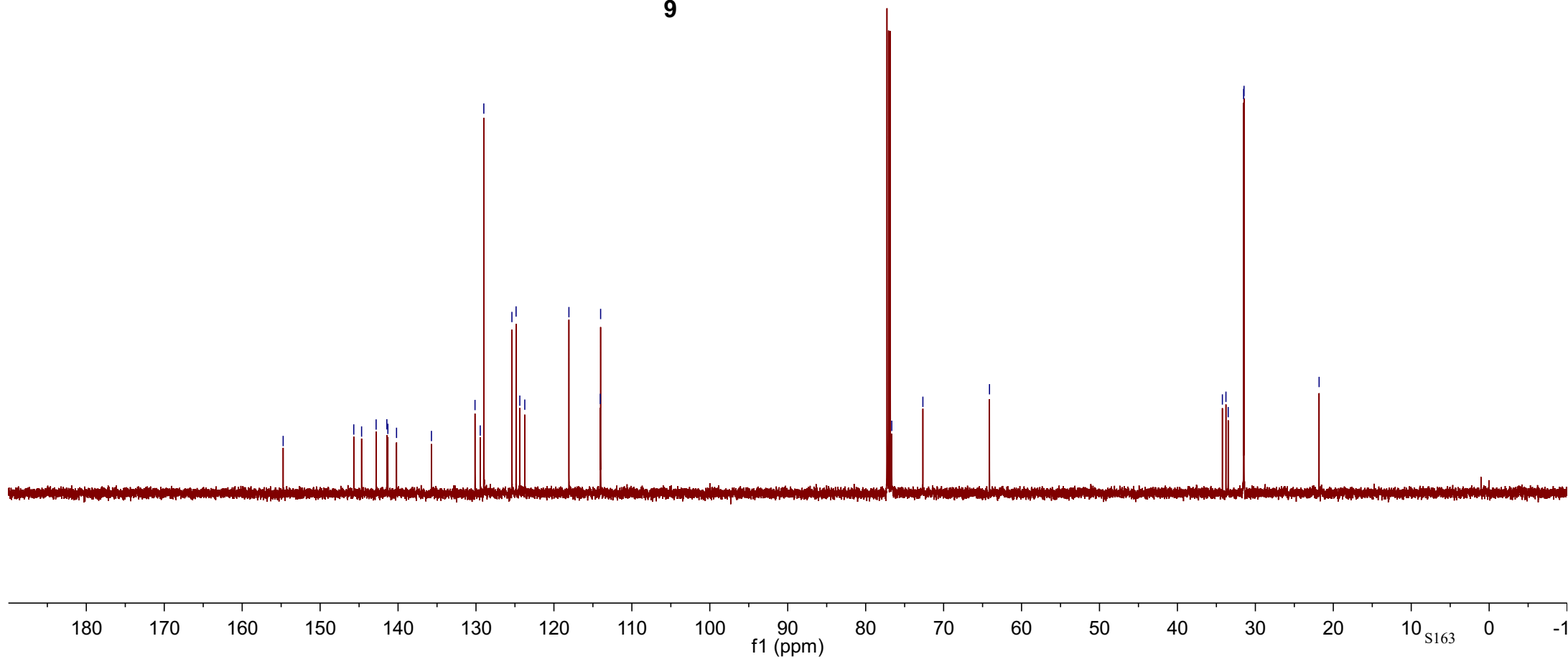
— 76.65
 — 72.66
 — 64.11

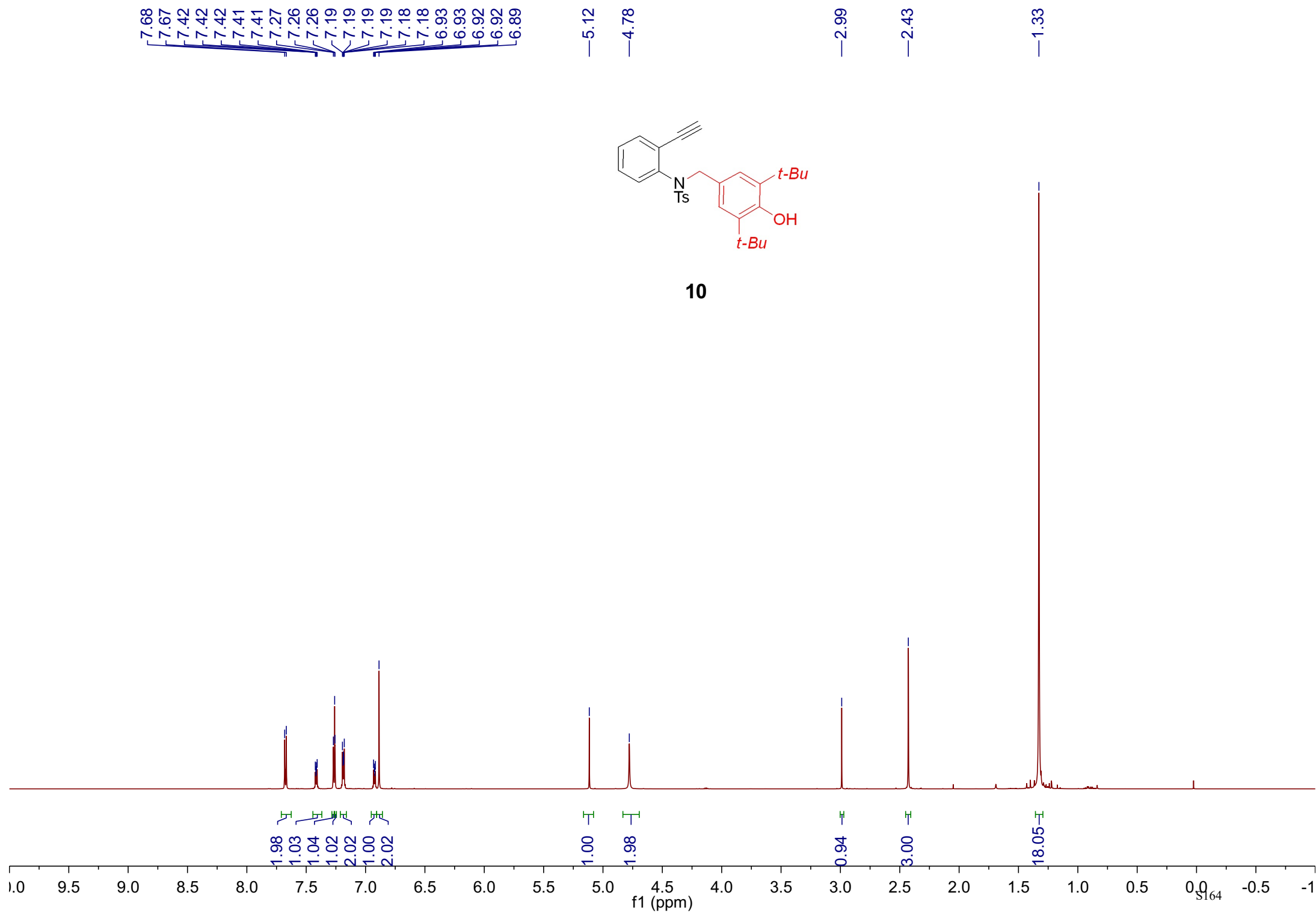
— 34.23
 — 33.77
 — 33.48
 — 31.51
 — 31.45
 — 21.82



9

Parameter	Value
Solvent	CDCI3
Spectrometer Frequency	150





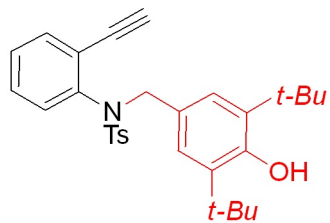
153.29
143.22
140.61
137.35
135.52
134.01
131.82
129.46
128.98
127.91
127.85
126.17
126.14
123.69

81.81
80.51

54.68

34.18
30.22

21.59



10

