# Supporting Information

# Substrate Selective Synthesis of Indole, Tetrahydroquinoline and Quinoline Derivatives *via* Intramolecular Addition of Hydrazones and Imines

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**Table of contents.**

## 1. General Information

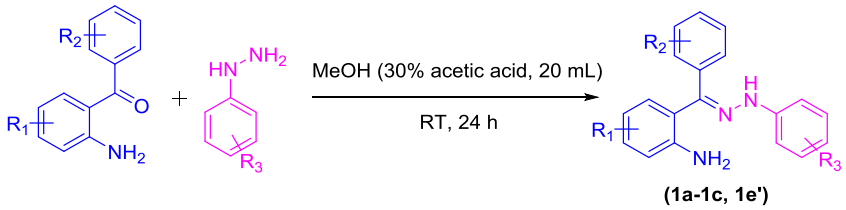
Melting points were determined by a capillary melting point apparatus and are uncorrected. All the compounds were fully characterized by $^{1}H$, $^{13}C$, IR and further confirmed through ESI-MS and ESI-HRMS analysis. $^{1}H$ NMR spectra were recorded on 400 and 500 MHz in $CDCl_3$ and DMSO-$d_6$ and $^{13}C$ NMR spectra recorded on 100 and 125 MHz in $CDCl_3$ and DMSO-$d_6$. Multiplicities are reported as follows: singlet (s), doublet (d), broad singlet (br s), doublet of doublets (dd), triplet (t), doublet of triplet (dt), triplet of doublet (td) multiplet (m) and quintet (quint). Chemical shift ($\delta$) and coupling constants ($J$) are reported in parts per million (ppm) relative to the residual signal of TMS in deuterated solvents and hertz, respectively. IR spectra were recorded using an FT-IR spectrophotometer and values are reported in $cm^{-1}$. HRMS were recorded using a Q-TOF mass spectrometer. Column chromatography was performed over silica gel (60-120, 100-200 and 230-400 mesh) by using EtOAc-*n*-hexane as eluent. All chemicals and reagents were purchased from commercial vendors and used without further purification.

## 2. Experimental Procedures

### General Experimental Procedure for the Preparation of Starting Materials 1.

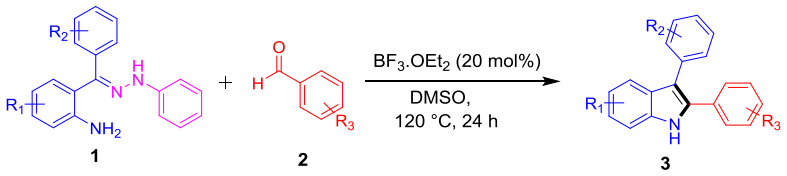
The starting materials hydrazones of 2-aminobenzophenones **1a, 1b, 1d, 1f and 1e′** (known compounds)[1] and **1c**, **1e**, **1b′-1d′** (unknown) were prepared by using known literature procedure.[1a, 1b] Most of the substrates were prepared by the experimental procedure mentioned below.

**Experimental procedure for the synthesis of 2-aminobenzophenone phenylhydrazone substrates (1a-1c, 1e').**



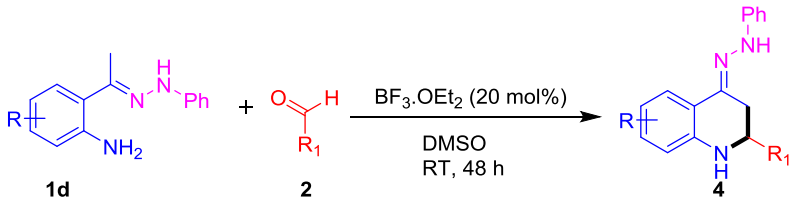

To a well-stirred solution of phenylhydrazine (2.0 mL; 1 equiv) in MeOH (30% acetic acid, 20 mL) was added 2-aminobenzophenones (1.0 equiv) at room temperature. The resulting mixture was allowed to stir for 24 h at room temperature. After completion of the reaction (monitored by TLC), MeOH was evaporated under reduced pressure and the residue was treated with saturated aq. $NaHCO_3$ solution (20 mL) and extracted with DCM (50 mL x 3). The organic layer was washed with brine (20 mL) and dried over anhydrous $Na_2SO_4$. Evaporation of the solvent under reduced pressure gave a crude product which was purified by column chromatography on silica gel (60-120 mesh) eluting with EtOAc-*n*-Hexane (3:97) to afford the solid products **1**. The Trans-geometry of **1a** was confirmed with the help of 2D-NOESY experiment.

**Experimental procedure for the synthesis of 2, 3-diaryl indole (3).**




*(E)*-2-(phenyl(2-phenylhydrazono)methyl)aniline **1** (300 mg, 1 equiv) was added to a well-stirred solution of benzaldehyde **2a** (1.1 equiv), $BF_3.OEt_2$ (0.2 equiv) and DMSO (10 mL) in 100 mL round bottom flask. The reaction mixture was allowed to stir for 24 h at 120 ºC. The completion of the reaction was monitored by TLC. The reaction mixture was allowed to cool at room temperature and quenched with saturated aq. $NaHCO_3$ (10 mL) solution and extracted with EtOAc (50 x 3 mL). The organic layer was washed with brine (10 mL) and dried over anhydrous $Na_2SO_4$. Evaporation of the solvent under reduced pressure gave a crude product which was purified by silica gel (100-200 mesh) column chromatography by using EtOAc-*n*-hexane (2:98) as an eluent to afford the respective products (**3**).

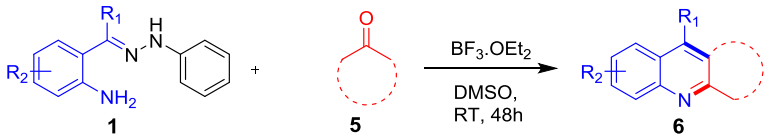
**Experimental procedure for the synthesis of 2-aryl-4-(2-phenylhydrazono)-1, 2, 3, 4-tetrahydroquinoline (4).**




*(E)*-2-(1-(2-phenylhydrazono)ethyl)aniline **1** (300 mg, 1equiv) was added to a well-stirred solution of benzaldehyde **2** (1.1 equiv), $BF_3.OEt_2$ (0.2 equiv) and DMSO (10 mL) in 100 mL round bottom flask. The reaction mixture was allowed to stir for 48 h at room temperature.

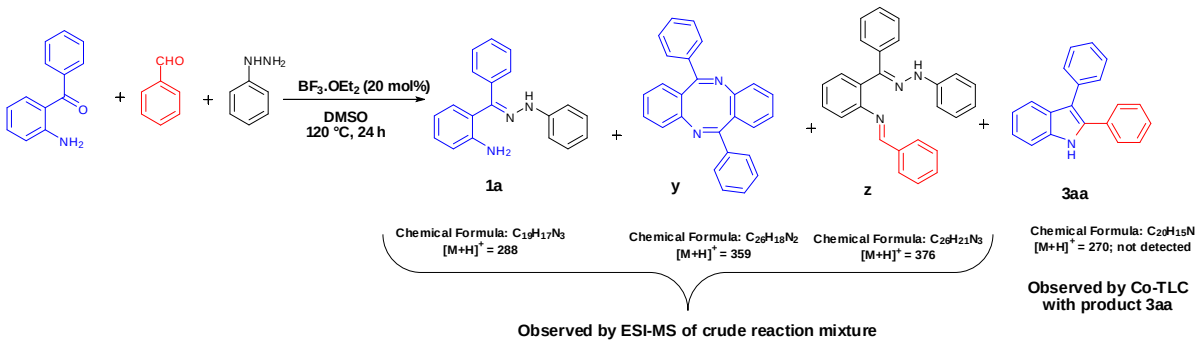
The completion of the reaction was monitored by TLC. The reaction mixture was quenched with 10 mL of saturated aq. $NaHCO_3$ solution and extracted with EtOAc (50 x 3 mL). The organic layer was washed with brine (10 mL) and dried over anhydrous $Na_2SO_4$. Evaporation of the solvent under reduced pressure gave a crude product which was purified by silica gel (100-200 mesh) column chromatography by using EtOAc-*n*-Hexane (2: 98) as an eluent to afford respective products (**4**). The Trans-geometry of **4de** was confirmed with the help of 2D-NOESY experiment.

**Experimental procedure for the synthesis of substituted quinoline (6).**



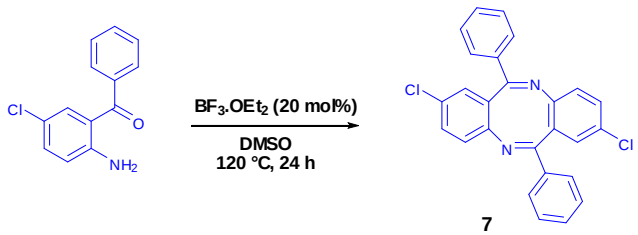

Corresponding (*E*)-2-(aryl/methyl(2-phenylhydrazono)methyl)aniline **1** (300 mg, 1 equiv) was added to a well-stirred solution of ketone **5** (1.1 equiv), $BF_3.OEt_2$ (0.2 equiv) and DMSO (10 mL) in 100 mL round bottom flask. The reaction mixture was allowed to stir for 48 h at room temperature. The completion of the reaction was monitored by TLC. The reaction mixture was quenched with 10 mL of saturated aq. $NaHCO_3$ solution and extracted with EtOAc (50 x 3 mL). The organic layer was washed with brine (10 mL) and dried over anhydrous $Na_2SO_4$. Evaporation of the solvent under reduced pressure gave a crude product which was purified by silica gel (100-200 mesh) column chromatography by using EtOAc-*n*-Hexane (1: 99) as an eluent to afford respective products (**6**).

**Experimental procedure for three component synthesis of 3aa in one pot *via* successive addition:**




2-aminobenzophenone (300 mg, 1 equiv) and phenylhydrazine (1 equiv) was stirred in DMSO (10 mL) at 120 ºC for 5 h in 100 mL round bottom flask followed by addition of aldehyde (1.1 equiv) and $BF_3.OEt_2$ (0.2 equiv) and subsequent stirring of reaction mixture at 120 ºC for another 19 h. The progress of the reaction was monitored by TLC. The reaction mixture was allowed to cool at room temperature and quenched with saturated aq. $NaHCO_3$ (10 mL) solution and extracted with EtOAc (50 x 3 mL). The organic layer was washed with brine (10 mL) and dried over anhydrous $Na_2SO_4$. Evaporation of the solvent under reduced pressure gave a crude mixture of products which was further analyzed by ESI-MS data.

**Experimental procedure for the synthesis of 1, 5-diazocine side product (7):**




To a well stirred solution of 2-amino-5-chlorobenzophenone (300 mg, 1 equiv) in DMSO (10 mL) was added $BF_3.OEt_2$ (0.2 equiv) in 100 mL round bottom flask. The reaction mixture was allowed to stir for 24 h at 120 ºC. The completion of the reaction was monitored by TLC. The reaction mixture was allowed to cool at room temperature and quenched with saturated aq.

$NaHCO_3$ (10 mL) solution and extracted with EtOAc (50 x 3 mL). The organic layer was washed with brine (10 mL) and dried over anhydrous $Na_2SO_4$. Evaporation of the solvent under reduced pressure gave a crude product which was purified by silica gel (100-200 mesh) column chromatography by using EtOAc-*n*-hexane (1:99) as an eluent to afford the respective products (**7**) in 40% yield.

## 3. Characterization data for compounds

***2-(Phenyl(2-phenylhydrazono)methyl)aniline (1a)*:** White solid (816 mg, 56%), mp 160-163 $^oC$; $^1H$ NMR (400 MHz, DMSO-$d_6$): $\delta$ 8.29 (s, 1H), 7.61(t, 2H, $J$ = 6.9 Hz), 7.56-7.52 (m, 1H), 7.30-7.28 (m, 2H), 7.21 (t, 2H, $J$ = 8.3 Hz), 7.05 (d, 2H, $J$ = 7.72 Hz), 7.00-6.96 (m, 1H), 6.91 (s, 2H), 6.80-6.75 (m, 2H), 6.46 (dd, 1H, $J$ = 7.9, 1.3 Hz), 6.39-6.35 (m, 1H) ppm. $^{13}C$ NMR (100 MHz, DMSO-$d_6$): $\delta$ 148.9 (C), 147.4 (C), 145.5 (C), 134.1 (C), 130.4 (CH), 129.9 (2xCH), 129.5 (2xCH), 129.38 (CH), 129.31 (2xCH), 128.7 (CH), 119.6 (CH), 119.2 (C), 116.2 (CH), 115.1 (CH), 112.9 (2xCH). FT-IR (KBr, $\nu$max/$cm^{-1}$) 3465, 3332, 1602, 1501, 1448, 1252, 1119, 1068, 750; HRMS (ESI): calcd for $C_{19}H_{18}N_3$ $[M+H]^+$ 288.1501, found: 288.1493.

***(E)-4-Chloro-2-(phenyl(2-phenylhydrazono)methyl)aniline (1b)*:** White solid (241 mg, 58%), mp 151-153 $^oC$; $^1H$ NMR (400 MHz, DMSO-$d_6$): $\delta$ 8.52 (s, 1H), 7.61-7.51(m, 3H), 7.28-7.21 (m, 4H), 7.08-7.05 (m, 2H), 6.99-6.95 (m, 1H), 6.82 (br s, 2H) 6.77 (dd, 1H, $J$ = 8.1, 0.9 Hz), 6.43 (dd, 1H, $J$ = 8.0, 1.5 Hz), 6.37-6.33 (m, 1H). $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ 146.7 (C), 145.7 (C), 144.8 (C), 132.9 (C), 129.6 (2xCH), 129.2 (CH), 129.0 (2xCH), 128.8 (2xCH), 128.5 (CH), 127.6 (CH), 120.0 (C), 119.4 (CH), 117.7 (C), 117.2 (CH), 112.5

(2xCH).FT-IR (KBr, *v*max/cm$^{-1}$) 3468, 3331, 1602, 1501, 1251, 1176, 750; HRMS (ESI): calcd for $C_{19}H_{17}ClN_3$ $[M+H]^+$ 322.1111, found: 322.1103.

***(E)-4-Chloro-2-((2-fluorophenyl)(2-phenylhydrazono)methyl)aniline (1c)*:** Yellow solid (159 mg, 39%), mp 155-157 $^oC$; $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ 7.63-7.57(m, 1H), 7.41 (dt, 1H, *J* = 7.5, 1.0), 7.37-7.26 (m, 4H), 7.21 (br s, 1H), 6.82 (br s, 2H) 7.04 (dd, 1H, *J* = 8.6, 2.4 Hz), 6.96-6.88 (m, 3H), 6.72 (d, 1H, *J* = 8.6 Hz), 6.64 (dd, 1H, *J* = 2.3, 0.5 Hz), 6.11 (br s, 1H). $^{13}C$ NMR (100 MHz, DMSO-$d_6$): $\delta$ 159.2 (d, *J* = 244 Hz, C), 144.5 (C), 144.8 (C), 140.6 (C), 131.9 (d, *J* = 8.0 Hz, CH), 131.1 (d, *J* = 4.0 Hz, CH), 129.0 (2xCH), 127.7 (CH), 127.5 (CH), 125.7 (d, *J* = 3.0 Hz, CH), 120.2 (d, *J* = 18 Hz, C), 119.6 (CH), 119.4 (CH), 118.1 (C), 117.3 (CH), 116.7 (d, *J* = 21 Hz, C), 112.6 (2xCH). FT-IR (KBr, *v*max/cm$^{-1}$) 3471, 3334, 3056, 1603, 1501, 1403, 1252, 1222, 1176, 1096, 756; HRMS (ESI): calcd for $C_{19}H_{16}ClFN_3$ $[M+H]^+$ 340.1017, found: 340.1006.

***(E)-2-(1-(2-Phenylhydrazono)ethyl)aniline (1d)*:** Yellow solid (375 mg, 75%), mp 102-103 $^oC$; $^1H$ NMR (400 MHz, DMSO-$d_6$): $\delta$ 9.09 (s, 1H), 7.34(d, 1H, *J* = 7.9 Hz), 7.22 (t, 2H, *J* = 7.4 Hz), 7.08 (d, 2H, *J* = 8.4 Hz), 6.99 (t, 1H, *J* = 8.0 Hz), 6.76 (t, 1H, *J* = 7.2 Hz) 6.71 (d, 1H, *J* = 8.0 Hz), 6.63 (br s, 2H), 6.56 (t, 1H, *J* = 7.9 Hz), 2.29 (s, 3H). $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ 146.7 (C), 146.0 (C), 145.7 (C), 129.0 (2xCH), 128.1 (CH), 128.0 (CH), 119.8 (C), 118.7 (CH), 115.7 (CH), 115.1 (CH), 112.3 (2xCH), 14.4. FT-IR (KBr, *v*max/cm$^{-1}$) 3453, 1603, 1496, 1252, 1216, 1159, 751; HRMS (ESI): calcd for $C_{14}H_{16}N_3$ $[M+H]^+$ 226.1344, found: 226.1336.

***2, 3-Diphenyl-1H-indole (3aa)*:[2]** White solid (227 mg, 81%), mp 113-114 (lit.[5e] 108-110 $^oC$); $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ = 8.24 (br s, 1H), 7.69 (d, 1H, *J* = 8.0 Hz), 7.47-7.42 (m, 5H),

7.40-7.24 (m, 7H), 7.18-7.14 (m, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 136.0 (C), 135.2 (C), 134.2 (C), 132.8 (C), 130.3 (2xCH), 128.9 (C), 128.8 (2xCH), 128.6 (2xCH), 128.3 (2xCH), 127.8 (CH), 126.3 (CH), 122.8 (CH), 120.5 (CH), 119.8 (CH), 115.2 (C), 111.0 (CH). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3398, 3019, 2399, 1645, 1403, 1156, 1069, 928, 669; HRMS (ESI): calcd for $C_{20}H_{16}N$ $[M+H]^+$ 270.1283, found: 270.1273.

***2-(4-Fluorophenyl)-3-phenyl-1H-indole (3ab)*:[3]** White solid (230 mg, 77%), mp 176-180 (lit.[15] 174-176 $^o$C); $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 8.19 (br s, 1H), 7.68 (d, 1H, $J$ = 8.0 Hz), 7.44-7.36 (m, 7H), 7.31-7.24 (m, 2H), 7.18-7.14 (m, 1H), 7.06-7.00 (m, 2H).$^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 162.4 (d, $J$ = 246 Hz, C), 136.0 (C), 134.9 (C), 133.3 (C), 130.2 (2xCH), 130.0 (d, $J$ = 8 Hz, CH), 129.0 (d, $J$ = 2.8 Hz, C), 128.8 (C), 128.7 (2xCH), 126.4 (CH), 122.9 (CH), 120.6 (CH), 119.8 (CH), 115.9 (d, $J$ = 22 Hz, CH), 115.2 (C), 111.0 (CH). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3745, 3392, 3019, 2924, 1645, 1512, 1403, 1158, 1047, 928, 839, 669; HRMS (ESI): calcd for $C_{20}H_{15}FN$ $[M+H]^+$ 288.1189, found: 288.1191.

***2-(4-Chlorophenyl)-3-phenyl-1H-indole (3ac)*:[4]** White solid (253 mg, 80%), mp 197-200 $^o$C; $^1$H NMR (400 MHz, $CDCl_3$): $\delta$8.20 (br s, 1H), 7.67 (d, 1H, $J$ = 8.0 Hz), 7.44-7.24 (m, 11H), 7.18-7.14 (m, 1H).$^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 136.1 (C), 134.8 (C), 133.7 (C), 132.9 (C), 131.3 (C), 130.2 (2xCH), 129.4 (2xCH), 129.0 (2xCH), 128.7 (CH), 126.6 (CH), 123.1 (CH), 120.7 (CH), 119.9 (CH), 115.7 (C), 111.0 (CH). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3399, 3019, 1644, 1215, 1155, 1069, 928, 669; HRMS (ESI): calcd for $C_{20}H_{15}ClN$ $[M+H]^+$ 304.0893, found: 304.0888.

***2-(4-Bromophenyl)-3-phenyl-1H-indole (3ae)*:[3]** White solid (272 mg, 75%), mp 140-143 $^o$C; $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 8.19 (br s, 1H), 7.67 (d, 1H, $J$ = 8.0 Hz), 7. 46-7.37 (m, 7H), 7.33-7.24 (m, 4H), 7.18-7.14 (m, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$) $\delta$ = 136.1 (C), 134.8 (C),

132.9 (C), 132.0 (2xCH), 131.7 (C), 130.2 (2xCH), 129.7 (2xCH), 128.88 (C), 128.80 (2xCH), 126.6 (CH), 123.2 (CH), 121.9 (C), 120.7 (CH), 119.9 (CH), 115.8 (C), 111.0 (CH).FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3398, 3019, 1645, 1402, 1216, 1155, 1068, 669; HRMS (ESI): calcd for $C_{20}H_{15}BrN$ $[M+H]^+$ 348.0388, found: 348.0375.

***2-(3-Bromophenyl)-3-phenyl-1H-indole (3af):***[4a] White solid (254 mg, 70%), mp 124-126 $^oC$; $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ 8.24 (br s, 1H), 7.67 (d, 1H, $J$ = 6.4 Hz), 7.61 (t, 1H, $J$ = 1.4 Hz), 7.44-7.38 (m, 6H), 7.33-7.25 (m, 3H), 7.18-7.13 (m, 2H). $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ 136.1 (C), 134.9 (C), 134.6 (C), 132.4 (C), 130.7 (CH), 130.6 (CH), 130.28 (CH), 130.24 (2xCH), 128.7 (2xCH), 127.1 (CH), 126.7 (CH), 123.3 (CH), 122.8 (C), 120.7 (CH), 120.0 (CH), 116.1 (C), 111.1 (CH). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3399, 3019, 2400, 1644, 1402, 1215, 1155, 1069, 928, 669; HRMS (ESI): calcd for $C_{20}H_{15}BrN$ $[M+H]^+$ 348.0388, found: 348.0380.

***2-(2, 4-Dichlorophenyl)-3-phenyl-1H-indole (3ag)*:** White solid (254 mg, 72%), mp 128-130 $^oC$; $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ 8.35 (br s, 1H), 7.80 (d, 1H, $J$ = 8.0 Hz), 7.50 (d, 1H, $J$ = 1.8 Hz), 7.45 (d, 1H, $J$ = 8.1), 7.35-7.25 (m, 6H), 7.23-7.13 (m, 3H). $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ 136.0 (C), 134.9 (C), 134.6 (C), 134.4 (C), 134.1 (CH), 130.4 (C), 130.3 (C), 130.2 (CH), 129.7 (2xCH), 128.6 (2xCH), 127.3 (CH), 126.4 (CH), 123.3 (CH), 120.6 (CH), 120.0 (CH), 117.6 (C), 111.1 (CH). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3393, 3019, 1644, 1216, 1155, 1069, 1023, 928, 771, 669; HRMS (ESI): calcd for $C_{20}H_{14}Cl_2N$ $[M+H]^+$ 338.0503, found: 338.0512.

***2-(2, 3-Dichlorophenyl)-3-phenyl-1H-indole (3ah)*:** White solid (250 mg, 71%), mp 137-139 $^oC$; $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ 8.36 (br s, 1H), 7.81 (d, 1H, $J$ = 7.9 Hz), 7.46 (d, 2H, $J$ = 8.6 Hz), 7.34-7.28 (m, 5H), 7.26-7.16 (m, 3H), 7.10 (t, 1H, $J$ = 7.8 Hz). $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ 135.9 (C), 134.6 (C), 134.2 (C), 133.9 (C), 132.4 (C), 131.7 (CH), 131.0 (C), 130.5

(CH), 129.7 (2xCH), 128.6 (2xCH), 127.4 (CH), 127.2 (C), 126.3 (CH), 123.3 (CH), 120.6 (CH), 120.1 (CH), 117.4 (C), 111.2 (CH).FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3461, 3019, 2343, 1602, 1522, 1475, 1021, 848, 669; HRMS (ESI): calcd for $C_{20}H_{14}Cl_2N$ $[M+H]^+$ 338.0503, found: 338.0506.

***2-(2, 6-Dichlorophenyl)-3-phenyl-1H-indole (3ai)*:** White solid (240 mg, 68%), mp 194-197 $^oC$; $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ 8.11 (br s, 1H), 7.85 (dd, 1H, $J$ = 7.9, 0.8 Hz), 7.47-7.45 (m, 1H), 7.38-7.36 (m, 4H), 7.32-7.27 (m, 4H), 7.25-7.19 (m, 2H). $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ = 137.2 (2xC), 136.2 (C), 134.8 (C), 131.6 (C), 130.7 (CH), 128.8 (2xCH), 128.6 (C), 128.4 (2xCH), 128.2 (2xCH), 127.1 (C), 126.2(CH), 123.0 (CH), 120.4 (CH), 120.1 (CH), 118.1 (C), 111.3 (CH). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3390, 1643, 1402, 1068, 831, 769; HRMS (ESI): calcd for $C_{20}H_{14}Cl_2N$ $[M+H]^+$ 338.0503, found: 338.0495.

***2-(2-Chloro-6-fluorophenyl)-3-phenyl-1H-indole (3aj)*:** White solid (218 mg, 65%), mp 137-139 $^oC$; $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ 8.21 (br s, 1H), 7.84 (d, 1H, $J$ = 8.0 Hz), 7.46 (d, 1H, $J$ = 8.1 Hz), 7.38-7.18 (m, 9H), 7.04-7.00 (m, 1H). $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ 161.5 (d, $J$ = 250 Hz, C), 136.3 (C), 134.9 (C), 130.9 (d, $J$ = 10 Hz, C), 128.9 (2xCH), 128.5 (2xCH), 127.1 (C), 126.3 (CH), 125.6 (d, $J$ = 3 Hz, CH), 124.6 (C), 123.1 (CH), 121.0 (d, $J$ = 19 Hz, C), 120.4 (CH), 120.0 (CH), 118.9 (C), 114.5 (d, $J$ = 23 Hz, CH), 111.2 (CH). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3399, 3021, 1611, 1216, 1068, 767, 670; HRMS (ESI): calcd for $C_{20}H_{14}ClFN$ $[M+H]^+$ 322.0799, found: 322.0794.

***4-(3-Phenyl-1H-indol-2-yl)benzonitrile (3ak)*:[5]** Yellow solid (252 mg, 82%), mp 176-179 (lit.[5a] 168-170 $^oC$); $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$8.30 (br s, 1H), 7.61 (d, 1H, $J$= 8.1), 7.55-7.53 (m, 2H), 7.48-7.45 (m, 2H), 7.42 (d, 1H, $J$= 8.2), 7.38-7.35 (m, 4H), 7.33-7.29 (m, 1H), 7.28-7.22 (m, 1H), 7.15-7.11 (m, 1H). $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$137.3 (C), 136.5 (C),

134.3 (C), 132.5 (2xCH), 131.7 (C), 130.2 (2xCH), 129.0 (2xCH), 128.9 (C), 128.3 (2xCH), 127.1 (CH), 123.9 (CH), 121.0 (CH), 120.3 (CH), 118.8 (C), 117.7 (C), 111.2 (CH), 110.8 (C). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3745, 3390, 3019, 2227, 1607, 1511, 1329, 1216, 1155, 1070, 928, 842, 669; HRMS (ESI): calcd for $C_{21}H_{15}N_2$ $[M+H]^+$ 295.1235, found: 295.1233

***2-(4-Nitrophenyl)-3-phenyl-1H-indole(3al)*:[6]** Red solid (239 mg, 73%), mp 168-171 $^oC$ (lit.[2g] 173-175 $^oC$); $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ 8.36 (br s, 1H), 8.17-8.14 (m, 2H), 7.65 (d, 1H, $J$= 7.9), 7.57-7.54 (m, 2H), 7.48-7.34 (m, 6H), 7.33-7.29 (m, 1H), 7.20-7.16 (m, 1H). $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ 146.7 (C), 139.2 (C), 136.7 (C), 134.2 (C), 131.4 (C), 130.2 (2xCH), 129.0 (2xCH), 128.9 (C), 128.3 (2xCH), 127.2 (CH), 124.24 (CH), 124.21 (CH), 121.1 (CH), 120.4 (CH), 118.4 (C), 111.3 (CH). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3390, 1645, 1402, 1068, 770; HRMS (ESI): calcd for $C_{20}H_{15}N_2O_2$ $[M+H]^+$ 315.1134, found: 315.1134.

***3-Phenyl-2-(p-tolyl)-1H-indole (3am)*:[6b, 7]** Yellow oil (236 mg, 59%); $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ 8.18 (br s, 1H), 7.71 (d, 1H, $J$= 7.9), 7.49-7.14 (m, 12H), 2.38 (s, 3H). $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ = 137.7 (C), 135.9 (C), 135.3 (C), 134.3 (C), 130.3 (2xCH), 129.9 (C), 129.5 (2xCH), 128.9 (C), 128.6 (2xCH), 128.1 (2xCH), 126.2 (CH), 122.6 (CH), 120.5 (CH), 119.7 (CH), 114.7 (C), 110.9 (CH), 21.3. FT-IR (neat, $\nu_{max}$/cm$^{-1}$) 3400, 3019, 1642, 1402, 1216, 1069, 669; HRMS (ESI): calcd for $C_{21}H_{18}N$ $[M+H]^+$ 284.1439, found: 284.1428.

***Ethyl-2-(4-(3-phenyl-1H-indol-2-yl)phenoxy)acetate (3an)*:** White solid (217 mg, 56%), mp 141-144 $^oC$; $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ 8.21 (br s, 1H), 7.67 (d, 1H, $J$ = 7.6 Hz), 7.43-7.12 (m, 10H), 6.86 (d, 2H, $J$ = 8.2 Hz), 4.62 (s, 2H), 4.27 (q, 2H, $J$ = 6.7 Hz), 1.30 (t, 3H, $J$ = 6.9 Hz). $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ 168.9 (C), 157.5 (C), 135.9 (C), 135.2 (C), 133.9 (C), 130.2 (2xCH), 129.6 (2xCH), 128.9 (C), 128.6 (2xCH), 126.4 (C), 126.2 (CH), 122.6 (CH), 120.5 (CH), 119.6 (CH), 115.0 (2xCH), 114.5 (C), 110.9 (CH), 65.5 ($CH_2$), 61.6 ($CH_2$),

14.3. FT-IR (KBr, $\nu_{max}/cm^{-1}$) 3390, 3019, 1642, 1402, 1216, 1069, 668; HRMS (ESI): calcd for $C_{24}H_{22}NO_3$ $[M+H]^+$ 372.1600, found: 372.1595.

***3-Phenyl-2-(3, 4, 5-trimethoxyphenyl)-1H-indole (3aq):***[5a] White solid (232 mg, 62%), mp 235-238 $^oC$; $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ 8.34 (br s, 1H), 7.65 (d, 1H, $J$ = 7.9 Hz), 7.48-7.37 (m, 5H), 7.32-7.24 (m, 2H), 7.18-7.14 (m, 1H), 6.64 (s, 2H), 3.88 (s, 3H), 3.67 (s, 6H). $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ 153.4 (2xC), 137.8 (C), 135.9 (C), 135.3 (C), 134.0 (C), 130.5 (2xCH), 129.0 (C), 128.6 (2xCH), 128.1 (C), 126.4 (CH), 122.8 (CH), 120.6 (CH), 119.7 (CH), 115.2 (C), 110.9 (CH), 105.4 (2xCH), 61.0, 56.0. FT-IR (KBr, $\nu_{max}/cm^{-1}$) 3684, 3019, 1602, 1407, 1128, 1032, 928, 669; HRMS (ESI): calcd for $C_{23}H_{22}NO_3$ $[M+H]^+$ 360.1600, found: 360.1586.

***2-(Furan-2-yl)-3-phenyl-1H-indole(3ar):***[3] Yellow Oil (108 mg, 40%); $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ 8.64 (br s, 1H), 7.60-7.57 (m, 3H), 7.50 (t, 2H, $J$ = 7.4 Hz), 7.44-7.38 (m, 3H), 7.27-7.23 (m, 1H), 7.14 (t, 1H, $J$ = 7.4 Hz), 6.39-6.37 (m, 2H). $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ = 147.2 (C), 141.4 (CH), 135.6 (C), 134.7 (C), 130.3 (2xCH), 128.9 (C), 128.7 (2xCH), 127.0 (CH), 125.3 (C), 123.0 (CH), 120.5 (CH), 119.6 (CH), 114.6 (C), 111.9 (CH), 110.9 (CH), 106.9 (CH). FT-IR (neat, $\nu_{max}/cm^{-1}$) 3391, 3019, 1644, 1403, 1215, 1069, 928, 669; HRMS (ESI): calcd for $C_{18}H_{14}NO$ $[M+H]^+$ 260.1075, found: 260.1068.

***3-Phenyl-2-(thiophen-2-yl)-1H-indole (3as):***[3, 8] White solid (195 mg, 68%), mp 120-123 $^oC$; $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ 8.23 (br s, 1H), 7.59 (d, 1H, $J$ = 8.0 Hz), 7.55-7.53 (m, 2H), 7.48-7.37 (m, 4H), 7.29-7.26 (m, 2H), 7.18-7.14 (m, 1H), 7.12 (d, 1H, $J$ = 3.5 Hz), 7.03-7.01 (m, 1H). $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ 135.8 (C), 134.68 (C), 134.61 (C), 130.6 (2xCH), 129.1 (C), 128.6 (2xCH), 128.4 (C), 127.6 (CH), 126.9 (CH), 125.49 (CH), 125.43 (CH), 123.1 (CH), 120.6 (CH), 119.8 (CH), 115.9 (C), 110.8 (CH). FT-IR (KBr, $\nu_{max}/cm^{-1}$) 3399,

3019, 1652, 1402, 1215, 1068, 668; HRMS (ESI): calcd for $C_{18}H_{14}NS$ $[M+H]^+$ 276.0847, found: 276.0838.

***3-Phenyl-1H, 1'H-2, 3'-biindole (3at)*:**[4a, 9] Yellow solid (206 mg, 64%), mp 212-214 °C (lit.[10] 220-221 °C); $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 9.80 (br s, 1H), 9.52 (br s, 1H), 7.66 (d, 1H, $J$ = 7.3 Hz), 7.47-6.96 (m, 13H). $^{13}$C NMR (100 MHz, $CDCl_3$):$\delta$ 136.2 (C), 136.0 (C), 130.2 (C), 129.6 (2xCH), 128.1 (3xCH), 125.9 (C), 125.3 (CH), 124.5 (CH), 121.8 (CH), 121.3 (C), 120.1 (CH), 119.7 (CH), 119.6 (CH), 118.5 (CH), 113.2 (C), 111.4 (CH), 110.9 (CH), 108.2 (C). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3464, 3019, 1602, 1406, 1334, 1156, 1053, 928, 669; HRMS (ESI): calcd for $C_{22}H_{17}N_2$ $[M+H]^+$ 309.1392, found: 309.1384.

***1-Benzyl-2, 3-diphenyl-1H-indole (3ea)*:**[11] White solid (154 mg, 54%), mp 155-158 °C; $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 7.85-7.83 (m, 1H), 7.36-7.17 (m, 16H), 7.03 (d, 2H, $J$ = 6.8 Hz), 5.31 (s, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 138.2 (C), 138.0 (C), 137.1 (C), 135.2 (C), 131.9 (C), 131.2 (2xCH), 130.0 (2xCH), 128.8 (2xCH), 128.5 (2xCH), 128.3 (3xCH), 127.5 (C), 127.3 (CH), 126.2 (2xCH), 125.7 (CH), 122.5 (CH), 120.5 (CH), 119.8 (CH), 115.8 (C), 110.6 (CH), 47.7 ($CH_2$). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3399, 3019, 1660, 1601, 1475, 1420, 1027, 928, 669; HRMS (ESI): calcd for $C_{27}H_{22}N$ $[M+H]^+$ 360.1752, found: 360.1778.

***5-Chloro-2, 3-diphenyl-1H-indole (3ba)*:**[10, 12] White solid (243 mg, 86%), mp 113-114 °C (lit.[22a] 110-112 °C); $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 8.26 (br s, 1H), 7.64 (d, 1H, $J$ = 1.4 Hz), 7.42-7.39 (m, 6H), 7.34-7.32 (m, 5H), 7.19 (dd, 1H, $J$ = 8.6, 1.9 Hz). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 135.5 (C), 134.5 (C), 134.3 (C), 132.3 (C), 130.1 (2xCH), 130.0 (C), 128.89 (2xCH), 128.81 (2xCH), 128.2 (2xCH), 128.1 (CH), 126.6 (CH), 126.3 (C), 123.0 (CH), 119.2 (CH), 114.9 (C), 112.0 (CH). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3391, 3019, 1644, 1404, 1215, 1156, 1026, 929, 669; HRMS (ESI): calcd for $C_{20}H_{15}ClN$ $[M+H]^+$ 304.0893, found: 304.0886.

***5-Chloro-2-(2-chlorophenyl)-3-phenyl-1H-indole (3bd)*:** White solid (262 mg, 83%), mp 202-205 $^{o}$C; $^{1}$H NMR (400 MHz, $CDCl_3$): $\delta$ 8.44 (br s, 1H), 7.77 (d, 1H, $J$ = 1.8 Hz), 7.49 (dd, 1H, $J$ = 8.0, 0.9 Hz), 7.38 (d, 1H, $J$ = 8.6 Hz), 7.34-7.31 (m, 4H), 7.30-7.23 (m, 4H), 7.19 (td, 1H, $J$ = 7.6, 1.1 Hz). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 134.29 (C), 134.22 (C), 133.7 (C), 133.3 (CH), 132.8 (C), 131.3 (C), 130.3 (CH), 129.9 (CH), 129.6 (2xCH), 128.6 (2xCH), 128.5 (C), 126.9 (CH), 126.5 (CH), 126.3 (C), 123.3 (CH), 119.4 (CH), 116.8 (C), 112.1 (CH). FT-IR (KBr, $\nu$max/cm$^{-1}$) 3461, 3019, 1602, 1457, 1334, 1067, 929, 668; HRMS (ESI): calcd for $C_{20}H_{14}Cl_2N$ $[M+H]^+$ 338.0503, found: 338.0505.

***5-Chloro-2-(2, 3-dichlorophenyl)-3-phenyl-1H-indole (3bh)*:** White solid (281 mg, 81%), mp 137-139 $^{o}$C; $^{1}$H NMR (400 MHz, $CDCl_3$): $\delta$ 8.41 (br s, 1H), 7.77 (d, 1H, $J$ = 1.8 Hz), 7.50 (dd, 1H, $J$ = 7.8, 1.8 Hz), 7.40-7.31 (m, 5H), 7.30-7.25 (m, 2H), 7.19-7.11 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 134.24 (C), 134.0 (C), 133.9 (C), 133.6 (C), 132.3 (C), 131.5 (CH), 130.8 (CH), 129.6 (2xCH), 128.7 (2xCH), 128.3 (C), 127.4 (CH), 126.7 (CH), 126.4 (C), 123.6 (CH), 119.5 (CH), 117.2 (C), 112.2 (CH). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3390, 3019, 1645, 1403, 1215, 1155, 669; HRMS (ESI): calcd for $C_{20}H_{13}Cl_3N$ $[M+H]^+$ 372.0111, found: 372.0114.

***4-(5-Chloro-3-phenyl-1H-indol-2-yl)benzonitrile (3bk)*:** Yellow solid (257 mg, 84%), mp 236-240 $^{o}$C; $^{1}$H NMR (400 MHz, $CDCl_3$): $\delta$ 8.39 (br s, 1H), 7.59-7.57 (m, 3H), 7.50-7.48 (m, 2H), 7.44-7.40 (m, 2H), 7.38-7.34 (m, 4H), 7.26-7.22 (m, 2H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 136.8 (C), 134.8 (C), 133.7 (C), 133.0 (C), 132.6 (2xCH), 130.1 (2xCH), 129.9 (C), 129.1 (2xCH), 128.4 (2xCH), 127.4 (CH), 126.8 (C), 124.2 (CH), 119.7 (CH), 118.7 (C), 117.3 (C), 112.3 (CH), 111.2 (C). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3391, 3019, 2229, 1607, 1403, 1215, 1068, 669; HRMS (ESI): calcd for $C_{21}H_{15}N_2$ $[M+H]^+$ 329.0846, found: 329.0846.

***5-Chloro-2-(4-nitrophenyl)-3-phenyl-1H-indole (3bl)***: Yellow solid (282 mg, 87%), mp 242-244 °C; $^{1}$H NMR (400 MHz, $CDCl_3$): $\delta$ 12.07 (br s, 1H), 8.22 (d, 2H, $J$ = 8.9 Hz), 7.67 (d, 2H, $J$ = 8.9 Hz), 7.51 (d, 1H, $J$ = 8.6 Hz), 7.47-7.42 (m, 3H), 7.39-7.34 (m, 3H), 7.23 (dd, 1H $J$ = 8.6, 2.0 Hz). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 146.3 (C), 138.4 (C), 135.0 (C), 133.7 (C), 133.1 (C), 129.7 (2xCH), 129.0 (2xCH), 128.9 (C), 128.8 (2xCH), 127.0 (CH), 124.8 (C), 123.8 (2xCH), 123.2 (CH), 118.0 (CH), 115.7 (C), 113.5 (CH). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3398, 3019, 1644, 1403, 1216, 1068, 669; HRMS (ESI): calcd for $C_{20}H_{14}ClN_2O_2$ $[M+H]^+$ 349.0744, found: 349.0744.

***5-Chloro-3-phenyl-2-(p-tolyl)-1H-indole(3bm):***[7b] Yellow solid (201 mg, 68%), mp 142-143 °C (lit.[7b] 140-142 °C);$^{1}$H NMR (400 MHz, $CDCl_3$): $\delta$ 8.22 (br s, 1H), 7.62 (d, 1H, $J$ = 1.9 Hz), 7.41-7.36 (m, 4H), 7.33-7.29 (m, 4H), 7.19-7.13 (m, 3H), 2.36 (s, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 138.1 (C), 135.7 (C), 134.6 (C), 134.2 (C), 130.1 (2xCH), 129.6 (2xCH), 129.4 (C), 128.7 (2xCH), 128.1 (2xCH), 126.6 (CH), 126.2 (C), 122.8 (CH), 119.1 (CH), 114.5 (C), 111.9 (CH), 21.4. FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3391, 3019, 1644, 1403, 1215, 1068, 669; HRMS (ESI): calcd for $C_{21}H_{17}ClN$ $[M+H]^+$ 318.1050, found: 318.1045.

***5-Chloro-2-(2, 4-dimethoxyphenyl)-3-phenyl-1H-indole (3bp)*:** White solid (248 mg, 73%), mp 187-190 °C; $^{1}$H NMR (400 MHz, $CDCl_3$): $\delta$ 8.96 (br s, 1H), 7.62 (s, 1H), 7.39-7.25 (m, 6H), 7.15-7.12 (m, 2H), 6.55 (s, 1H), 6.34 (d, 1H, $J$ = 7.9 Hz), 3.84 (s, 3H) 3.80 (s, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 160.9 (C), 158.0 (C), 135.5 (C), 133.7 (C), 132.87 (C), 132.80 (CH), 130.1 (2xCH), 129.2 (C), 128.7 (2xCH), 126.3 (CH), 125.7 (C), 122.3 (CH), 118.6 (CH), 114.6 (C), 113.3 (C), 111.7 (CH), 105.1 (CH), 99.3 (CH), 55.8, 55.5. FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3391, 3019, 1610, 1522, 1215, 1028, 928, 669; HRMS (ESI): calcd for $C_{22}H_{19}ClNO_2$ $[M+H]^+$ 364.1104, found: 364.1091.

***5-Chloro-3-phenyl-2-(3, 4, 5-trimethoxyphenyl)-1H-indole (3bq)*:** White solid (253 mg, 69%), mp 235-237 ºC; $^{1}$H NMR (400 MHz, $CDCl_3$): $\delta$ 8.36 (br s, 1H), 7.58 (s, 1H), 7.44-7.38 (m, 4H), 7.35-7.28 (m, 2H), 7.18 (d, 1H, $J$ = 8.2 Hz), 6.60 (s, 2H), 3.86 (s, 3H), 3.66 and 3.65 (s, 6H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 153.4 (2xC), 138.0 (C), 135.4 (C), 134.6 (C), 134.2 (C), 130.3 (2xCH), 130.1 (C), 128.7 (2xCH), 127.6 (C), 126.8 (CH), 126.3 (C), 123.0 (CH), 119.1 (CH), 114.9 (C), 112.0 (CH), 105.4 (2xCH), 61.0, 56.0. FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3369, 3019, 1651, 1403, 1217, 1068, 668; HRMS (ESI): calcd for $C_{23}H_{21}ClNO_3$ $[M+H]^+$ 394.1210, found: 394.1205.

***5-Chloro-3-(2-fluorophenyl)-2-phenyl-1H-indole (3ca)*:** White solid (165 mg, 58%), mp 142-144 ºC; $^{1}$H NMR (400 MHz, $CDCl_3$): $\delta$ 8.37 (br s, 1H), 7.47 (s, 1H), 7.40-7.31 (m, 8H), 7.20-7.13 (m, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 160.5 (d, $J$ = 246 Hz, C), 136.7 (C), 134.2 (C), 132.8 (CH), 132.3 (C), 130.2 (C), 129.0 (CH), 128.9 (2xCH), 128.3 (CH), 127.5 (2xCH), 126.3 (C), 124.4 (CH), 123.1 (CH), 122.1 (d, $J$ = 16 Hz, C), 119.4 (CH), 116.2 (d, $J$ = 22 Hz, CH), 112.0 (CH), 108.3 (C). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3390, 3019, 1644, 1402, 1215, 1067, 669; HRMS (ESI): calcd for $C_{20}H_{14}ClFN$ $[M+H]^+$ 322.0799, found: 322.0789.

***5-Chloro-2-(4-chlorophenyl)-3-(2-fluorophenyl)-1H-indole (3cc)*:** White solid (163 mg, 52%), mp 166-169 ºC; $^{1}$H NMR (400 MHz, $CDCl_3$): $\delta$ 8.34 (br s, 1H), 7.46 (s, 1H), 7.37-7.28 (m, 7H), 7.21-7.13 (m, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 160.4 (d, $J$ = 246 Hz, C), 135.5 (C) 134.3 (d, $J$ = 7 Hz, C), 132.7 (d, $J$ = 3 Hz, C), 130.8 (C), 130.1 (C), 129.2 (3xCH), 128.7 (3xCH), 126.5 (C), 124.5 (d, $J$ = 3 Hz, CH), 123.4 (CH), 121.8 (d, $J$ = 16 Hz, C), 119.4 (CH), 116.3 (d, $J$ = 22 Hz, CH), 112.1 (CH), 108.7 (C). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3749, 3399, 3019, 1644, 1216, 1154, 1066, 669; HRMS (ESI): calcd for $C_{20}H_{13}Cl_2FN$ $[M+H]^+$ 356.0409, found: 356.0413.

***4-(5-Chloro-3-(2-fluorophenyl)-1H-indol-2-yl)benzonitrile (3ck)*:** White solid (171 mg, 56%), mp 195-197 °C; $^{1}$H NMR (400 MHz, $CDCl_3$): $\delta$ 8.47 (br s, 1H), 7.60 (s, 2H, $J$ = 8.4 Hz), 7.49-7.47 (m, 3H), 7.41-7.34 (m, 3H), 7.26-7.22 (m, 2H), 7.20-7.14 (m, 1H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 160.2 (d, $J$ = 246 Hz, C), 136.8 (C) 134.7 (C), 134.2 (C), 132.7 (2xCH), 132.5 (d, $J$ = 3 Hz, CH), 130.0 (C), 129.6 (d, $J$ = 8 Hz, CH), 127.7 (2xCH), 126.9 (CH), 124.7 (d, $J$ = 4 Hz, CH), 124.3 (CH), 121.3 (d, $J$ = 16 Hz, C), 119.7 (d, $J$ = 2 Hz, CH), 118.7 (C), 116.5 (d, $J$ = 22 Hz, CH) 112.4 (CH), 111.5 (C), 110.4 (C). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3388, 3021, 2400, 1608, 1418, 1216, 1066, 671; HRMS (ESI): calcd for $C_{21}H_{13}ClFN_2$ $[M+H]^+$ 347.0751, found: 347.0748.

***(E)-2-Phenyl-4-(2-phenylhydrazono)-1, 2, 3, 4-tetrahydroquinoline (4da)*:** White solid (209 mg, 50%), mp 188-190 °C; $^{1}$H NMR (400 MHz, $CDCl_3$): $\delta$7.34-7.33 (m, 4H), 7.29-7.24 (m, 2H), 7.21-7.15 (m, 3H), 7.14-7.09 (m, 2H), 6.98 (d, 2H, $J$ = 7.8 Hz), 6.83-6.77 (m, 2H), 6.73-6.69 (m, 1H), 5.18 (dd, 1H, $J$ = 12.2, 7.3 Hz), 3.94 (dd, 1H, $J$ = 16.8, 12.2 Hz), 3.26 (dd, 1H, $J$ = 16.8, 7.3 Hz). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 149.0 (C), 145.3 (C), 144.7 (C), 142.6 (C), 129.5 (CH), 129.2 (2xCH), 129.1 (2xCH), 128.4 (CH), 127.7 (CH), 126.0 (2xCH), 119.2 (CH), 117.4 (CH), 116.4 (CH), 115.4 (C), 113.3 (2xCH), 63.2 (CH), 45.17 ($CH_2$). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3391, 3019, 1613, 1499, 1326, 1155, 1068, 669; HRMS (ESI): calcd for $C_{21}H_{20}N_3$ $[M+H]^+$ 314.1657, found: 314.1650.

***(E)-2-(4-Chlorophenyl)-4-(2-phenylhydrazono)-1, 2, 3, 4-tetrahydroquinoline (4dc)*:** White solid (280 mg, 60%), mp 202-203 °C; $^{1}$H NMR (400 MHz, DMSO-$d_6$): $\delta$ 7.81 (d, 2H, $J$ = 8.1 Hz), $\delta$ = 7.48 (d, 2H, $J$ = 8.1 Hz), 7.20-7.13 (m, 3H), 7.08 (t, 1H, $J$ = 7.8 Hz), 6.89 (d, 2H, $J$ = 8.1 Hz), 6.81 (d, 2H, $J$ = 8.1 Hz), 6.75-6.72 (m, 3H), 5.54 (t, 1H, $J$ = 7.4 Hz), 5.49 (dd, 1H, $J$ = 12.0, 6.0 Hz), 4.01 (dd, 1H, $J$ = 17.2, 12.1 Hz), 3.21 (dd, 1H, $J$ = 17.2, 6.1 Hz). $^{13}$C NMR

(100 MHz, DMSO-$d_6$): $\delta$ 149.8 (C), 147.0 (C), 144.0 (C), 141.5 (C), 131.8 (C), 129.3 (CH), 129.0 (2xCH), 128.9 (2xCH), 128.7 (CH), 127.9 (2xCH), 118.5 (CH), 115.1 (CH), 115.0 (CH) 113.0 (C), 112.6 (2xCH), 60.7 (CH), 44.3 ($CH_2$). FT-IR (KBr, $\nu_{max}$/$cm^{-1}$) 3387, 2361, 1597, 1512, 1450, 1342, 1265, 748; HRMS (ESI): calcd for $C_{21}H_{19}ClN_3$ $[M+H]^+$ 348.1268, found: 348.1259.

***(E)-2-(2, 4-Dichlorophenyl)-4-(2-phenylhydrazono)-1, 2, 3, 4-tetrahydroquinoline (4dg)*:** White solid (296 mg, 58%), mp 209-210 $^oC$; $^1H$ NMR (400 MHz, DMSO-$d_6$): $\delta$ 7.72 (d, 1H, $J$ = 2.1 Hz), 7.34 (dd, 1H, $J$ = 8.4, 2.1 Hz), 7.23-7.19 (m, 2H), 7.14 (dd, 1H, $J$ = 7.9, 1.4 Hz), 7.10-7.05 (m, 2H), 6.83-6.80 (m, 3H), 6.77-6.73 (m, 3H), 6.56-6.52 (m, 1H), 5.55 (dd, 1H, $J$ = 12.1, 6.0 Hz), 4.08 (dd, 1H, $J$ = 17.2, 12.2 Hz), 3.18 (dd, 1H, $J$ = 17.2, 6.1 Hz). $^{13}C$ NMR (100 MHz, DMSO-$d_6$): $\delta$ 150.1 (C), 147.0 (C), 143.6 (C), 138.0 (C), 132.8 (C), 132.2 (C), 129.49 (CH), 129.45 (CH), 129.2 (2xCH), 128.8 (CH), 128.6 (CH), 128.0 (CH), 118.7 (CH), 115.1 (CH), 115.0 (CH), 112.8 (C), 112.3 (2xCH), 58.4 (CH), 42.8 ($CH_2$). FT-IR (KBr, $\nu$max/$cm^{-1}$) 2361, 2353, 1597, 1497, 1389, 1327, 833; HRMS (ESI): calcd for $C_{21}H_{18}Cl_2N_3$ $[M+H]^+$ 382.0878, found: 382.0867.

***(E)-2-(3, 4-Dichlorophenyl)-4-(2-phenylhydrazono)-1, 2, 3, 4-tetrahydroquinoline (4dv)*:** White solid (293 mg, 57%), mp 184-185 $^oC$; $^1H$ NMR (400 MHz, DMSO-$d_6$): $\delta$ 7.60-7.56 (m, 2H), 7.24-7.17 (m, 3H), 7.14 (dd, 1H, $J$ = 7.8, 1.0 Hz), 7.10-7.06 (m, 1H), 6.91 (d, 2H, $J$ = 7.8 Hz), 6.82 (d, 2H, $J$ = 8.1 Hz), 6.76-6.72 (m, 3H), 6.57-6.53 (m, 1H), 5.41 (dd, 1H, $J$ = 11.9, 6.1 Hz), 3.97 (dd, 1H, $J$ = 17.2, 12.0 Hz), 3.24 (dd, 1H, $J$ = 17.3, 6.2 Hz).$^{13}C$ NMR (100 MHz, DMSO-$d_6$): $\delta$ 150.03 (C), 147.0 (C), 143.9 (C), 143.6 (C), 131.3 (C), 131.2 (CH), 129.8 (C), 129.4 (CH), 129.0 (2xCH), 128.7 (CH), 128.1 (CH), 126.2 (CH), 118.7 (CH), 115.1

(CH), 115.0 (CH) 112.9 (C), 112.6 (2xCH), 60.3 (CH), 44.1 ($CH_2$). FT-IR (KBr, $\nu_{max}$/$cm^{-1}$) 3387, 2924, 2361, 1597, 1327, 1265, 1126, 748; HRMS (ESI): calcd for $C_{21}H_{18}Cl_2N_3$ $[M+H]^+$ 382.0878, found: 382.0869.

***(E)-2-(4-Bromophenyl)-4-(2-phenylhydrazono)-1, 2, 3, 4-tetrahydroquinoline (4de)*:** White solid (302 mg, 58%), mp 203-205 °C; $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$ 7.52 (d, 2H, $J$ = 8.3 Hz), 7.24 (d, 2H, $J$ = 8.3 Hz), 7.18-7.12 (m, 3H), 7.08-7.04 (m, 1H), 6.89 (d, 2H, $J$ = 8.2 Hz), 6.80 (d, 1H, $J$ = 8.1 Hz), 6.73-6.69 (m, 3H), 6.54 (t, 1H, $J$ = 7.4 Hz), 5.37 (dd, 1H, $J$ = 12.0, 6.1 Hz), 3.97 (dd, 1H, $J$ = 17.2, 12.1 Hz), 3.17 (dd, 1H, $J$ = 17.2, 6.2 Hz). $^{13}$C NMR (100 MHz, DMSO-$d_6$): $\delta$ 149.8 (C), 146.9 (C), 144.0 (C), 141.9 (C), 131.8 (2xCH), 129.3 (CH), 129.0 (2xCH), 128.7 (CH), 128.2 (2xCH), 120.3 (C), 118.5 (CH), 115.1 (CH), 115.0 (CH), 113.0 (C), 112.6 (2xCH), 60.8 (CH), 44.26 ($CH_2$). FT-IR (KBr, $\nu_{max}$/$cm^{-1}$) 3389, 3020, 2401, 1595, 1494, 1384, 1120, 1067, 929, 670; HRMS (ESI): calcd for $C_{21}H_{19}BrN_3$ $[M+H]^+$ 392.0762, found: 392.0760.

***(E)-4-(4-(2-Phenylhydrazono)-1, 2, 3, 4-tetrahydroquinolin-2-yl)benzonitrile (4dk)*:** Yellow (318 mg, 70%), mp 193-194 °C; $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$ 7.39 (d, 2H, $J$ = 8.5 Hz), 7.30 (d, 2H, $J$ = 8.5 Hz), 7.19-7.13 (m, 3H), 7.09-7.05 (m, 1H), 6.90 (d, 2H, $J$ = 7.9 Hz), 6.82-6.70 (m, 4H), 6.55 (t, 1H, $J$ = 7.8 Hz) 5.39 (dd, 1H, $J$ = 11.9, 6.1 Hz), 3.97 (dd, 1H, $J$ = 17.2, 12.0 Hz), 3.18 (dd, 1H, $J$ = 17.2, 6.2 Hz). $^{13}$C NMR (100 MHz, DMSO-$d_6$): $\delta$ 149.9 (C), 148.1 (C), 147.0 (C), 143.9 (C), 132.9 (2xCH), 129.4 (CH), 129.1 (2xCH), 128.7 (CH), 127.1 (2xCH), 118.7 (CH), 118.6 (C), 115.1 (CH), 115.0 (CH), 112.9 (C), 112.6 (2xCH), 110.2 (C), 61.0 (CH), 44.1 ($CH_2$). FT-IR (KBr, $\nu_{max}$/$cm^{-1}$) 3397, 3294, 2361, 1597, 1497, 1327, 1265, 748; HRMS (ESI): calcd for $C_{22}H_{19}N_4$ $[M+H]^+$ 339.1610, found: 339.1604.

***(E)-2-(4-Nitrophenyl)-4-(2-phenylhydrazono)-1, 2, 3, 4-tetrahydroquinoline (4dl)*:** Orange solid (327 mg, 68%), mp 155-156 $^{o}$C; $^{1}$H NMR (400 MHz, DMSO-$d_6$): $\delta$ 8.21 (d, 2H, $J$ = 8.7 Hz), 7.57 (d, 2H, $J$ = 8.7 Hz), 7.20-7.10 (m, 3H), 7.08 (t, 1H, $J$ = 8.1 Hz), 6.90 (d, 2H, $J$ = 8.0 Hz), 6.82 (d, 1H, $J$ = 8.1 Hz), 6.76-6.72 (m, 3H), 5.55 (t, 1H, $J$ = 7.7 Hz), 5.56 (dd, 1H, $J$ = 12.0, 6.2 Hz), 4.04 (dd, 1H, $J$ = 17.2, 12.2 Hz), 3.24 (dd, 1H, $J$ = 17.3, 6.3 Hz).$^{13}$C NMR (100 MHz, DMSO-$d_6$): $\delta$ 150.1 (C), 149.9 (C), 147.0 (C), 146.8 (C), 143.9 (C), 129.4 (CH), 129.0 (2xCH), 128.7 (CH), 127.3 (2xCH), 124.2 (2xCH), 118.7 (CH), 115.1 (CH), 115.0 (CH) 112.8 (C), 112.6 (2xCH), 60.8(CH), 44.1 ($CH_2$). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3387, 2924, 2361, 1597, 1443, 1350, 1342, 1119, 748; HRMS (ESI): calcd for $C_{22}H_{19}N_4O_2$ $[M+H]^+$ 359.1508, found: 359.1498.

***(E)-4-(2-Phenylhydrazono)-2-(p-tolyl)-1, 2, 3, 4-tetrahydroquinoline (4dm)*:** White solid (183 mg, 42%), mp 155-156 $^{o}$C; $^{1}$H NMR (400 MHz, DMSO-$d_6$): $\delta$ 6.16-7.14 (m, 7H), 7.07 (t, 1H, $J$ = 7.8 Hz), 6.91 (d, 1H, $J$ = 7.8 Hz), 7.08 (t, 1H, $J$ = 8.1 Hz), 6.90 (d, 2H, $J$ = 8.0 Hz), 6.82 (d, 1H, $J$ = 8.1 Hz), 6.70(t, 1H, $J$ = 7.2 Hz), 6.55 (t, 1H, $J$ = 7.3 Hz), 5.24 (dd, 1H, $J$ = 11.6, 6.1 Hz), 3.95 (dd, 1H, $J$ = 17, 12.1 Hz), 3.14 (dd, 1H, $J$ = 17.0, 6.1 Hz). $^{13}$C NMR (100 MHz, DMSO-$d_6$): $\delta$ = 149.7 (C), 146.7 (C), 144.2 (C), 139.6 (C), 136.4 (C), 129.4 (2xCH), 129.2 (CH), 128.8 (2xCH), 128.6 (CH), 125.8 (2xCH), 118.3 (CH), 115.2 (CH), 115.0 (CH) 113.3 (C), 112.6 (2xCH), 61.2(CH), 44.5 ($CH_2$), 20.6. FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3672, 3394, 3032, 2908, 1597, 1497, 1327, 1018, 879; HRMS (ESI): calcd for $C_{22}H_{22}N_3$ $[M+H]^+$ 328.1814, found: 328.1817.

***(E)-2-(4-Methoxyphenyl)-4-(2-phenylhydrazono)-1, 2, 3, 4-tetrahydroquinoline (4do)*:** White solid (201 mg, 44%), mp 168-170 $^{o}$C; $^{1}$H NMR (400 MHz, $CDCl_3$): $\delta$ 7.26-7.24 (m,

2H), 7.21-7.17 (m, 2H), 7.15-7.09 (m, 2H), 6.98 (d, 2H, *J* = 7.8 Hz), 6.86 (d, 2H, *J* = 8.7 Hz), 6.80-6.76 (m, 2H), 6.70-6.66 (m, 1H), 5.95 (br s, 2H), 5.13 (dd, 1H, *J* = 12.1, 7.2 Hz), 3.91 (dd, 1H, *J* = 16.8, 12.2 Hz), 3.23 (dd, 1H, *J* = 16.8, 7.3 Hz). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 159.1 (C), 149.2 (C), 146.1 (C), 144.9 (C), 134.7 (C), 129.4 (CH), 129.0 (2xCH), 128.4 (CH), 127.2 (2xCH), 119.1 (CH), 116.8 (CH), 115.9 (CH), 115.0 (C), 114.6 (2xCH), 113.3 (2xCH), 62.7 (CH), 55.4, 45.2 ($CH_2$). FT-IR (KBr, $\nu_{max}$/cm$^{-1}$) 3400, 3019, 1644, 1402, 1216, 1068, 669; HRMS (ESI): calcd for $C_{22}H_{22}N_3O$ $[M+H]^+$ 344.1763, found: 344.1759.

***(E)-2-(2, 4-Dimethoxyphenyl)-4-(2-phenylhydrazineylidene)-1, 2, 3, 4-tetrahydroquinoline (4dp)*:** Yellow oil (215 mg, 43%); $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$ 7.18-7.12 (m, 3H), 7.07-7.03 (m, 1H), 6.85-6.78 (m, 4H), 6.71-6.68 (m, 3H), 6.63 (d, 1H, *J* = 2.3 Hz), 6.55-6.51 (m, 1H), 6.39 (dd, 1H, *J* = 8.5, 2.4 Hz), 5.42 (dd, 1H, *J* = 11.9, 5.8 Hz), 3.94-3.87 (m, 4H), 3.70 (s, 3H), 3.05 (dd, 1H, *J* = 17.1, 5.8 Hz). $^{13}$C NMR (100 MHz, DMSO-$d_6$): $\delta$ 159.8 (C), 157.0 (C), 150.1 (C), 146.8 (C), 144.0 (C), 129.1 (CH), 128.9 (CH), 129.45 (2xCH), 128.6 (CH), 126.7 (CH), 121.4 (C), 118.0 (CH), 115.1 (CH), 114.9 (CH), 113.3 (C), 112.2 (2xCH), 105.0 (CH), 98.8 (CH), 55.7 , 55.6 (CH), 55.1, 42.8 ($CH_2$). FT-IR (neat, $\nu_{max}$/cm$^{-1}$) 3379, 2924, 2361, 1612, 1504, 1203, 1119, 748; HRMS (ESI): calcd for $C_{23}H_{24}N_3O_2$ $[M+H]^+$ 374.1869, found: 374.1860.

***(E)-6-(4-Chlorophenyl)-8-(2-phenylhydrazono)-5, 6, 7, 8-tetrahydro- [1, 3]dioxolo [4, 5-g]quinolone (4fc)*:** Yellow solid (204 mg, 46%), mp 205-206 $^o$C; $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$ 7.39 (d, 2H, *J* = 8.4 Hz), 7.29 (d, 2H, *J* = 8.4 Hz), 7.15 (t, 2H, *J* = 7.5 Hz), 6.87 (br s, 2H), 6.72 (br s, 4H), 6.45 (s, 1H), 5.87 (br s, 2H), 5.32 (br s, 1H), 3.90 (dd, 1H, *J* = 17.2, 12.0 Hz), 3.13 (dd, 1H, *J* = 17.1, 6.0 Hz). $^{13}$C NMR (100 MHz, DMSO-$d_6$): $\delta$ = 150.4 (C),

149.0 (C), 144.7 (C), 144.4 (C), 142.1 (C), 138.4 (C), 132.2 (C), 129.4 (4xCH), 128.4 (2xCH), 118.7 (CH), 113.0 (2xCH), 107.6 (CH), 105.5 (C), 100.9 ($CH_2$), 96.5 (CH), 61.1 (CH), 45.2 ($CH_2$). FT-IR (KBr, $\nu_{max}$/$cm^{-1}$) 3425, 2924, 2361, 1597, 1412, 1234, 741; HRMS (ESI): calcd for $C_{22}H_{19}ClN_3O_2$ $[M+H]^+$ 392.1166, found: 392.1173.

***(E)-2-Ethyl-4-(2-phenylhydrazono)-1, 2, 3, 4-tetrahydroquinoline (4dw)*:** White solid (128 mg, 36%), mp 112-113 $^oC$; $^1H$ NMR (400 MHz, DMSO-$d_6$): $\delta$ 7.28-7.22 (m, 3H), 7.09-7.02 (m, 3H), 6.80-6.74 (m, 2H), 6.62-6.58 (m, 1H), 4.37-4.31 (m, 1H), 3.50 (dd, 1H, $J$ = 17.1, 11.2 Hz), 3.18-3.13 (m, 1H), 1.76-1.67 (m, 1H), 1.56-1.45 (m, 1H), 0.84 (t, 3H, $J$ = 7.4 Hz). $^{13}C$ NMR (100 MHz, DMSO-$d_6$): $\delta$ 150.4 (C), 146.7 (C), 143.9 (C), 129.1 (2xCH), 128.9 (CH), 128.5 (CH), 118.0 (CH), 115.0 (CH), 114.9 (CH) 113.5 (C), 112.5 (2xCH), 58.2 (CH), 38.4 ($CH_2$), 24.3 ($CH_2$), 8.5. FT-IR (KBr, $\nu_{max}$/$cm^{-1}$) 3371, 3032, 2955, 2361, 1597, 1497, 1388, 1296, 1337, 1134, 748; HRMS (ESI): calcd for $C_{17}H_{20}N_3$ $[M+H]^+$ 266.1657, found: 266.1658.

***9-Phenyl-1, 2, 3, 4-tetrahydroacridine (6aa)*:[13]** White solid (147 mg, 54%), mp 156-157 $^oC$; $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ 8.01 (d, 1H, $J$ = 8.4Hz), 7.61-7.57 (m, 1H), 7.54-7.50 (m, 2H), 7.48-7.44 (m, 1H), 7.33-7.30 (m, 2H), 7.24-7.22 (m, 2H), 3.20 (t, 2H, $J$ = 13.2, 6.6 Hz), 2.60 (t, 2H, $J$ = 13.0, 6.5 Hz), 2.00-1.93 (m, 2H), 1.82-1.76 (m, 2H).$^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ 159.2 (C), 146.6 (C), 146.4 (C), 137.2 (C), 129.2 (2xCH), 128.7 (2xCH), 128.49 (2xCH), 128.46 (C), 127.8 (CH), 126.8 (C), 125.9 (CH), 125.4 (CH), 34.3 ($CH_2$), 28.1 ($CH_2$), 23.1 ($CH_2$), 23.0 ($CH_2$). FT-IR (KBr, $\nu_{max}$/$cm^{-1}$) 3433, 2924, 2361, 1628, 1481, 1134, 671; HRMS (ESI): calcd for $C_{19}H_{18}N$ $[M+H]^+$ 260.1439, found: 260.1431.

***2-Methyl-4-phenylquinoline (6ab):[14]*** Yellow oil (115 mg, 50%); $^{1}$H NMR (400 MHz, $CDCl_3$): $\delta$ 8.10 (br d, 1H, $J$ = 8.4 Hz), 7.85 (dd, 1H, $J$ = 8.4, 0.9 Hz), 7.68 (ddd, 1H, $J$ = 8.3, 6.8, 1.4 Hz), 7.53-7.47 (m, 5H), 7.42 (ddd, 1H, 8.2, 6.9, 1.2 Hz), 7.23 (s, 1H), 2.77 (s, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 158.5 (C), 148.6 (C), 148.4 (C), 138.2 (C), 129.5 (2xCH), 129.4 (CH), 129.0 (CH), 128.6 (2xCH), 128.4 (CH), 125.8 (CH), 125.7 (CH), 125.1 (C), 122.3 (CH), 25.4. FT-IR (neat, $\nu_{max}$/cm$^{-1}$) 3402, 3063, 2361, 1597, 1489, 1404, 1196, 764; HRMS (ESI): calcd for $C_{16}H_{14}N$ $[M+H]^+$ 220.1126, found: 220.1118.

***9-Phenyl-2, 3-dihydro-1H-cyclopenta [b]quinoline (6ac):[15]*** Yellow oil (144 mg, 56%); $^{1}$H NMR (400 MHz, DMSO-$d_6$): $\delta$ 8.07-8.05 (m, 1H), 7.63-7.59 (m, 2H), 7.54-7.50 (m, 2H), 7.48-7.43 (m, 1H), 739-7.35 (m, 3H), 3.23 (t, 2H, $J$ = 7.64 Hz), 2.90 (t, 2H, $J$ = 7.36 Hz), 2.16 (quint, 2H, $J$ = 7.5 Hz). $^{13}$C NMR (100 MHz, DMSO-$d_6$): $\delta$ 167.5 (C), 148.0 (C), 142.8 (C), 136.9 (C), 133.7 (C), 129.4 (2xCH), 128.9 (CH), 128.6 (2xCH), 128.3 (CH), 128.1 (CH), 126.3 (C), 125.7 (CH), 125.6 (CH), 35.3 ($CH_2$), 30.4 ($CH_2$), 23.6 ($CH_2$). FT-IR (neat, $\nu_{max}$/cm$^{-1}$) 3433, 2962, 2924, 2361, 1597, 1389, 1342, 1126, 764; HRMS (ESI): calcd for $C_{18}H_{16}N$ $[M+H]^+$ 246.1283, found: 246.1274.

***6-Chloro-3-methyl-2, 4-diphenylquinoline (6bd):[16]*** Yellow oil (169 mg, 55%); $^{1}$H NMR (400 MHz, $CDCl_3$): $\delta$ 8.09 (d, 1H, $J$ = 9.0 Hz), 7.61-7.58 (m, 3H), 7.57-7.54 (m, 2H), 7.52-7.48 (m, 3H), 7.46-7.42 (m, 1H), 7.35 (d, 1H, $J$ = 2.2 Hz), 7.30-7.28 (m, 2H), 2.14 (s, 3H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 161.2 (C), 147.2 (C), 144.7 (C), 141.2 (C), 137.1 (C), 132.2 (C), 131.2 (CH), 129.6 (CH), 129.3 (2xCH), 129.0 (4xCH), 128.5 (2xCH), 128.4 (CH), 128.3 (CH), 127.98 (C), 127.95 (C), 124.8 (CH), 18.8. FT-IR (neat, $\nu_{max}$/cm$^{-1}$) 3402, 2932, 2361, 1628,

1381, 1196, 1119, 756; HRMS (ESI): calcd for $C_{22}H_{17}ClN$ $[M+H]^+$ 330.1050, found: 330.1044.

***2, 4-Dimethylquinoline (6da):***[17] Yellow oil (105 mg, 50%); $^1$H NMR (400 MHz, DMSO-$d_6$): $\delta$ 8.00 (dd, 1H, $J$ = 8.3, 0.5 Hz), 7.91 (dd, 1H, $J$ = 8.3, 1.0 Hz), 7.64 (ddd, 1H, $J$ = 8.3, 6.9, 1.4 Hz), 7.47 (ddd, 1H, 8.2, 6.9, 1.2 Hz), 7.09 (d, 1H, $J$ = 0.6 Hz), 2.67 (br s, 3H), 2.62 (d, 3H, $J$ = 0.9 Hz). $^{13}$C NMR (100 MHz, DMSO-$d_6$): $\delta$ 158.7 (C), 147.7 (C), 144.2 (C), 129.2 (CH), 129.1 (CH), 126.6 (C), 125.4 (CH), 123.6 (CH), 122.7 (CH), 25.2, 18.6. FT-IR (neat, $\nu_{max}$/cm$^{-1}$) 3703, 3402, 3063, 2950, 2361, 1612, 1574, 1389, 1342, 1196, 756; HRMS (ESI): calcd for $C_{11}H_{12}N$ $[M+H]^+$ 158.0970, found: 158.0959.

***9-Methyl-1, 2, 3, 4-tetrahydroacridine (6db):***[18] Yellow oil (128 mg, 49%); $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 7.98-7.95 (m, 2H), 7.66-7.57 (m, 1H), 7.48-7.43 (m, 1H), 3.12-3.10 (m, 2H), 2.92-2.89 (m, 2H), 2.56 (s, 3H), 1.95-1.92 (m, 4H). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ = 158.7 (C), 146.0 (C), 141.4 (C), 129.0 (CH), 128.8 (C), 128.2 (CH), 127.0 (C), 125.4 (CH), 123.4 (CH), 34.6 ($CH_2$), 27.2 ($CH_2$), 23.3 ($CH_2$), 22.9 ($CH_2$), 13.6. FT-IR (neat, $\nu_{max}$/cm$^{-1}$) 3433, 2962, 2361, 1597, 1402, 1226, 764; HRMS (ESI): calcd for $C_{14}H_{16}N$ $[M+H]^+$ 198.1283, found: 198.1284.

***9-Methyl-2, 3-dihydro-1H-cyclopenta [b]quinoline (6dc):***[19] Yellow oil (130 mg, 53%); $^1$H NMR (400 MHz, $CDCl_3$): $\delta$ 7.99 (dd, 1H, $J$ = 8.4, 0.6 Hz), 7.88 (dd, 1H, $J$ = 8.3, 1.1 Hz), 7.57 (ddd, 1H, $J$ = 8.3, 6.9, 1.4 Hz), 7.43 (ddd, 1H, $J$ = 8.2, 6.9, 1.2 Hz), 3.13 (t, 2H, $J$ = 7.7 Hz), 2.99 (t, 2H, $J$ = 7.5 Hz), 2.51 (s, 3H), 2.15 (quint, 2H, $J$ = 7.6 Hz). $^{13}$C NMR (100 MHz, $CDCl_3$): $\delta$ 166.9 (C), 147.4 (C), 138.1 (C), 134.0 (C), 129.1 (CH), 128.0 (CH), 127.0 (C), 125.2 (CH), 123.3 (CH), 35.11 ($CH_2$), 29.6 ($CH_2$), 22.9 ($CH_2$), 14.8. FT-IR (neat, $\nu_{max}$/cm$^{-1}$)

3402, 3063, 2955, 2361, 1612, 1443, 1342, 1119, 756; HRMS (ESI): calcd for $C_{13}H_{14}N$ $[M+H]^+$ 184.1126, found: 184.1126.

***2, 8-dichloro-6, 12-diphenyldibenzo [b, f] [1, 5]diazocine (7)*:[20]** Yellow solid (220 mg, 40%), mp 120-122 $^oC$; $^1H$ NMR (400 MHz, $CDCl_3$): $\delta$ 7.75-7.73 (m, 4H), 7.46-7.42 (m, 2H), 7.38-7.34 (m, 4H), 7.31 (dd, 2H, *J*= 2.4, 8.6 Hz), 7.00-6.96 (m, 4H). $^{13}C$ NMR (100 MHz, $CDCl_3$): $\delta$ 168.9 (C), 150.3 (C), 137.3 (C), 131.6 (CH), 130.2 (CH), 129.5 (2xCH), 129.2 (C), 128.5 (2xCH), 128.2 (C), 127.3 (CH), 122.6 (CH). HRMS (ESI): calcd for $C_{26}H_{17}Cl_2N_2$ $[M+H]^+$ 427.0769, found: 427.0763.

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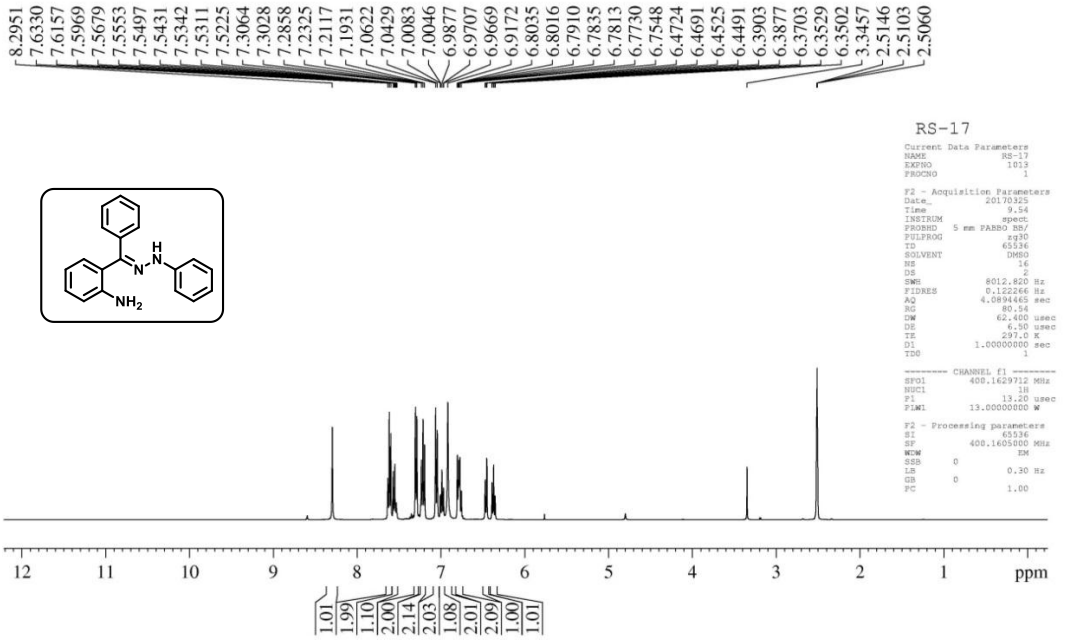
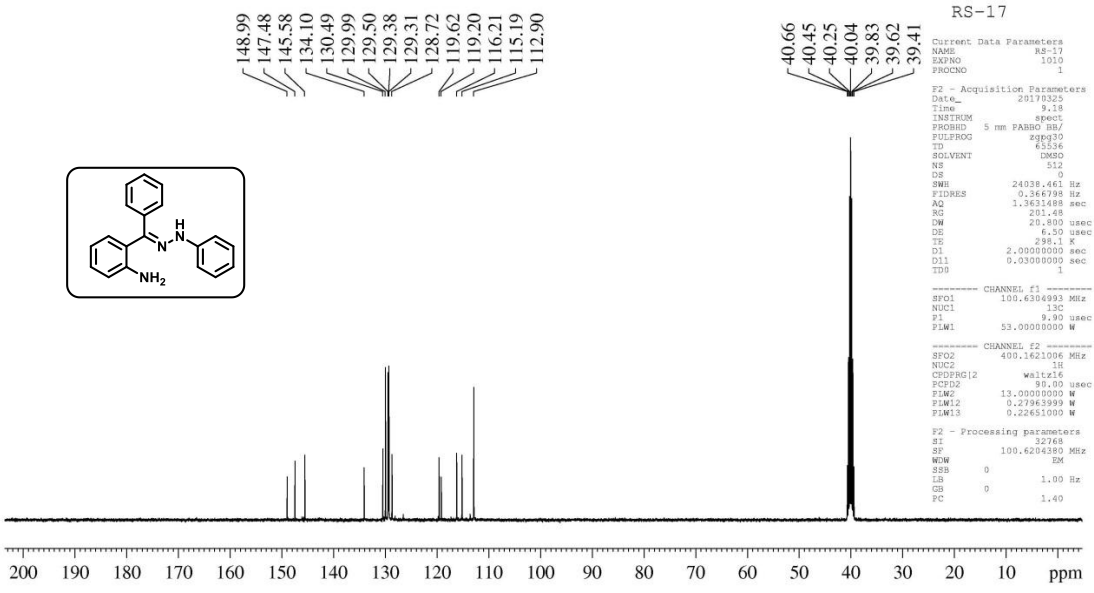
## Copies of $^{1}H$, $^{13}C$ NMR, HRMS spectra




**$^{1}H$ NMR spectrum of 1a**

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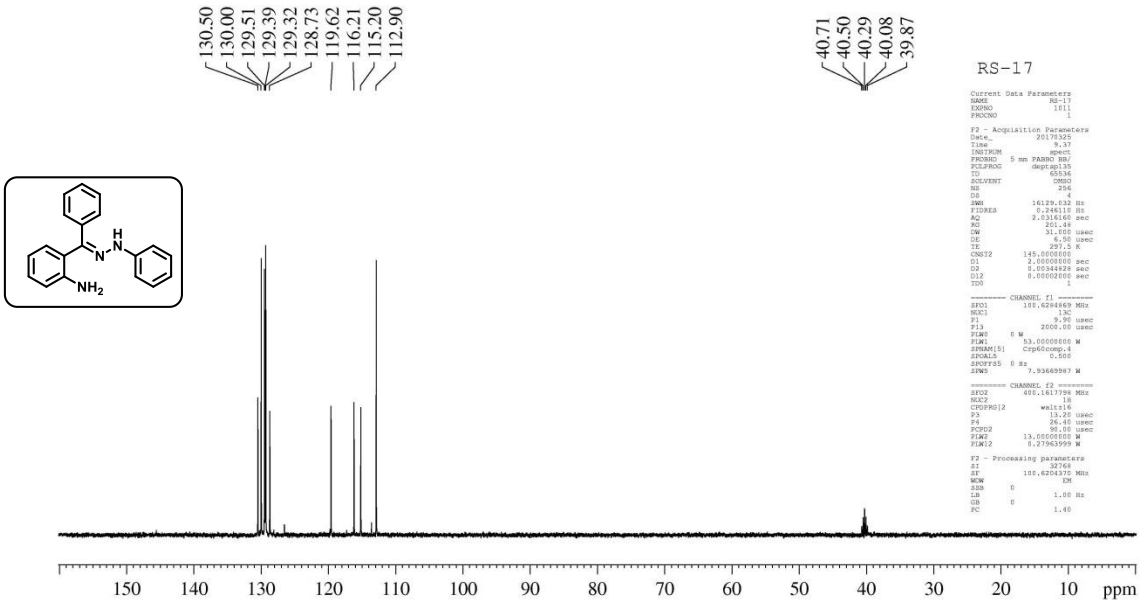
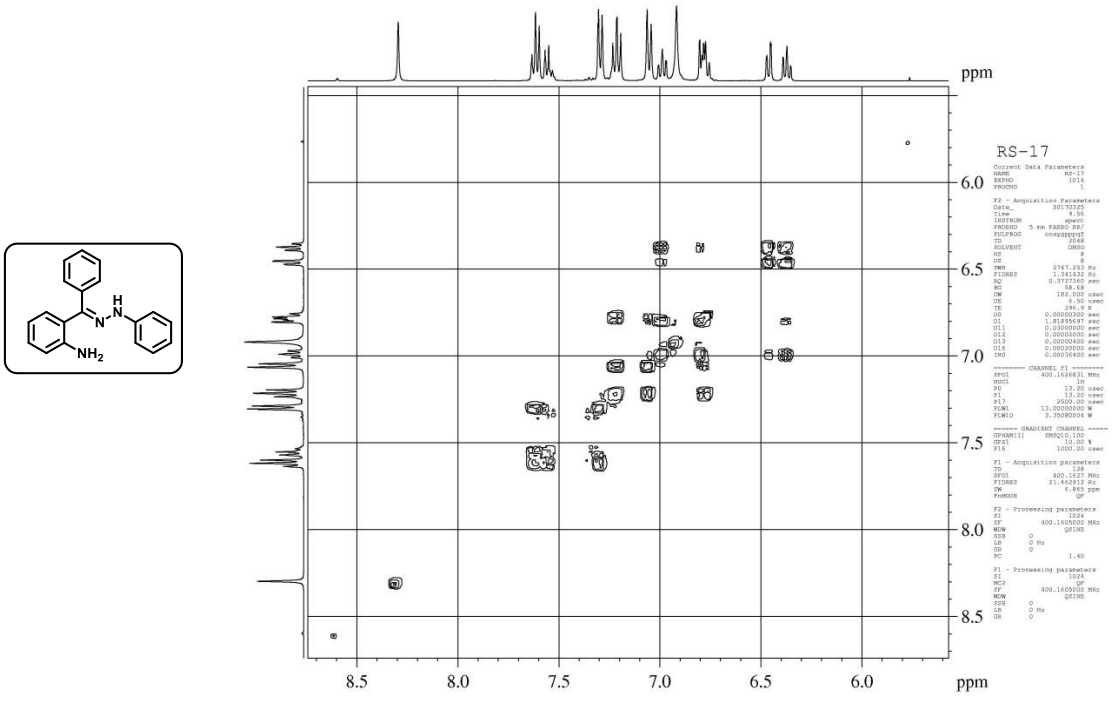
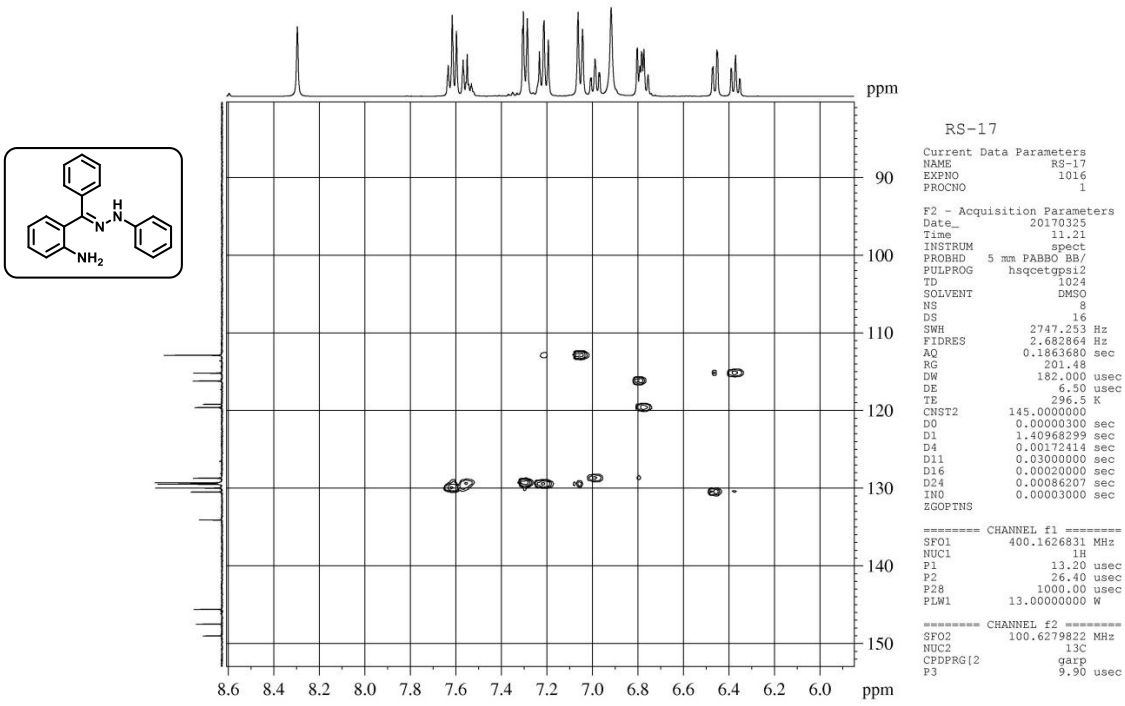



**DEPT 135 spectrum of 1a**




**Expanded COSY spectrum of 1a**




RS-17

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PROCNO                    1

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TD                    1024
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NS                       8
DS                      16
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FIDRES           2.682864 Hz
AQ               0.1863680 sec
RG                  201.48
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DE                   6.50 usec
TE                  296.5 K
CNST2           145.0000000
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D1               1.40968299 sec
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D16              0.00020000 sec
D24              0.00086207 sec
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ZGOPTNS

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NUC1                     1H
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P2                    26.40 usec
P28                 1000.00 usec
PLW1           13.00000000 W

======== CHANNEL f2 ========
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CPDPRG[2                garp
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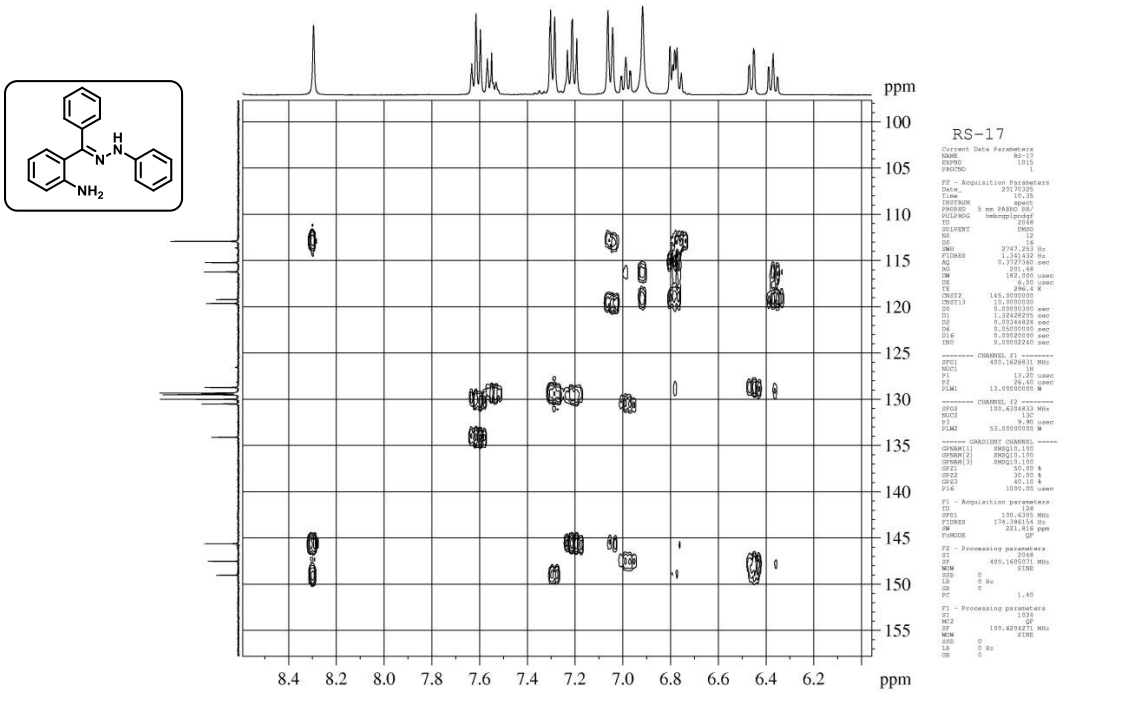
**Expanded HSQC spectrum of 1a**




RS-17


Current Data Parameters
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PROCNO                    1

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NS                      12
DS                      16
SWH              2747.253 Hz
FIDRES           1.341432 Hz
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RG                  201.48
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DE                   6.50 usec
TE                  296.4 K
CNST2           145.0000000
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D6               0.05000000 sec
DI4              0.00625000 sec
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NUC1                     1H
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PLW1           13.00000000 W

======== CHANNEL f2 ========
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NUC2                     13C
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PLW2           53.00000000 W

====== GRADIENT CHANNEL =====
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GPNAM[2]       SMSQ10.100
GPNAM[3]       SMSQ10.100
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GPZ2                 30.00 %
GPZ3                 40.10 %
P16                1000.00 usec

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SFO1          100.6355 MHz
FIDRES        176.386154 Hz
SW                221.816 ppm
FnMODE                  QF

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SSB                    0
LB                     0 Hz
GB                     0
PC                     1.40

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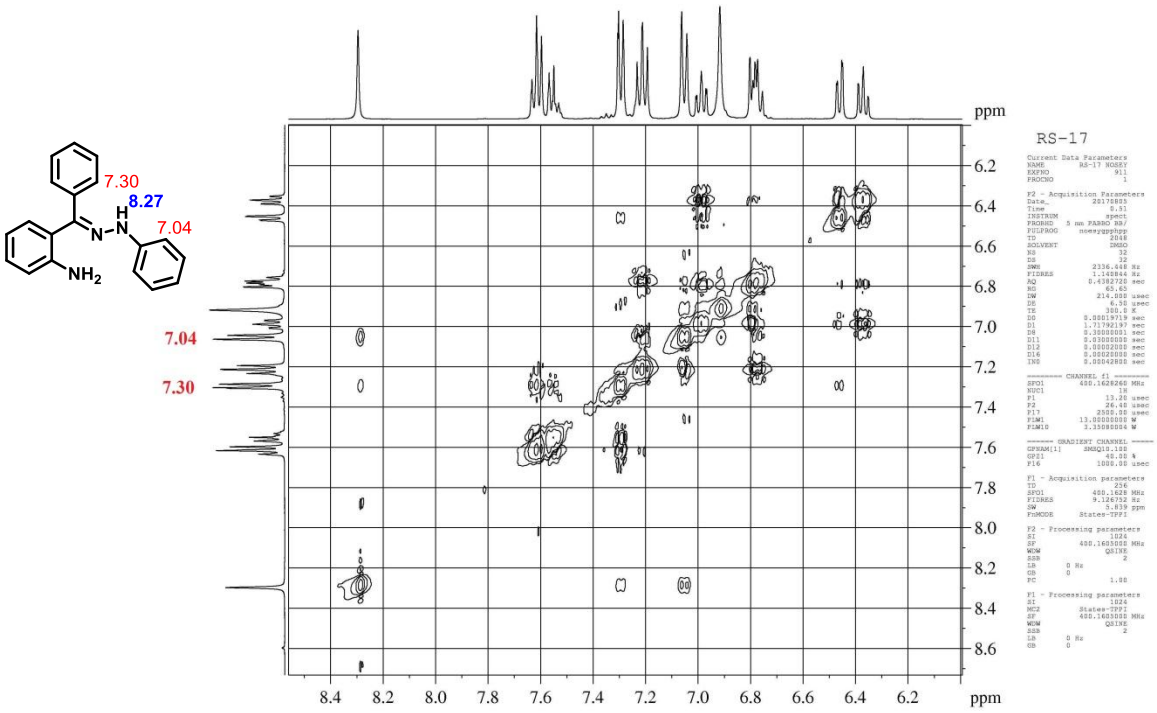
**Expanded HMBC spectrum of 1a**

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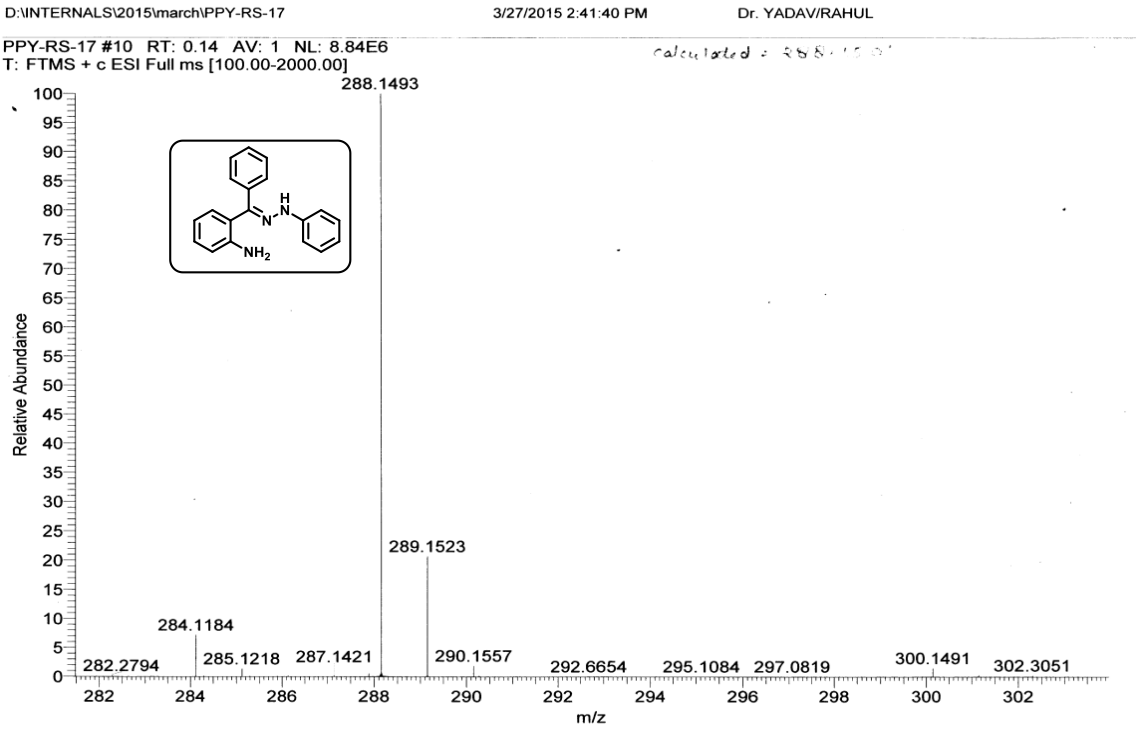
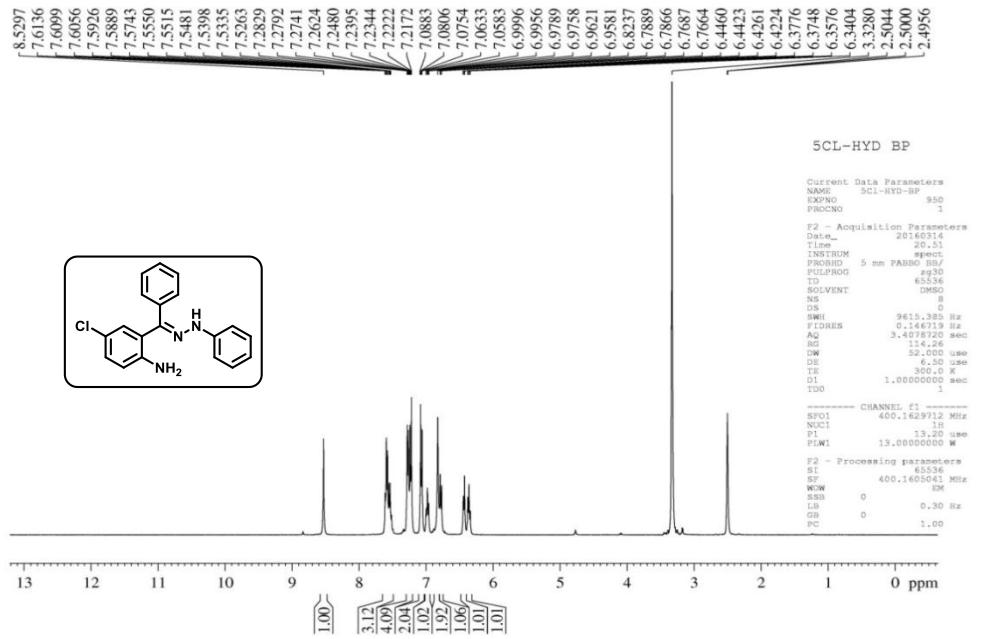
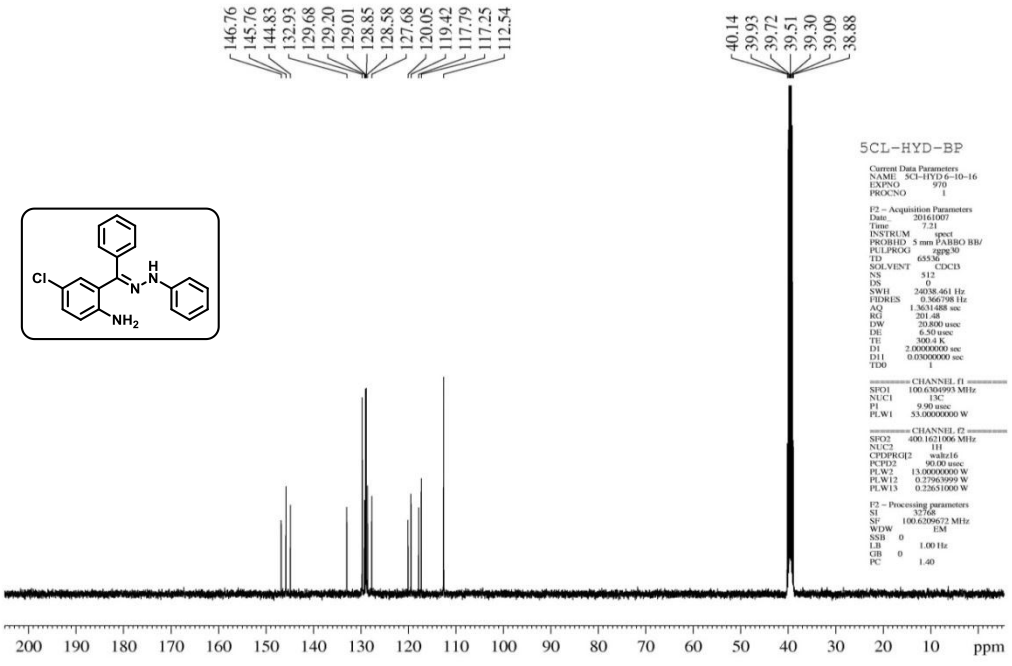


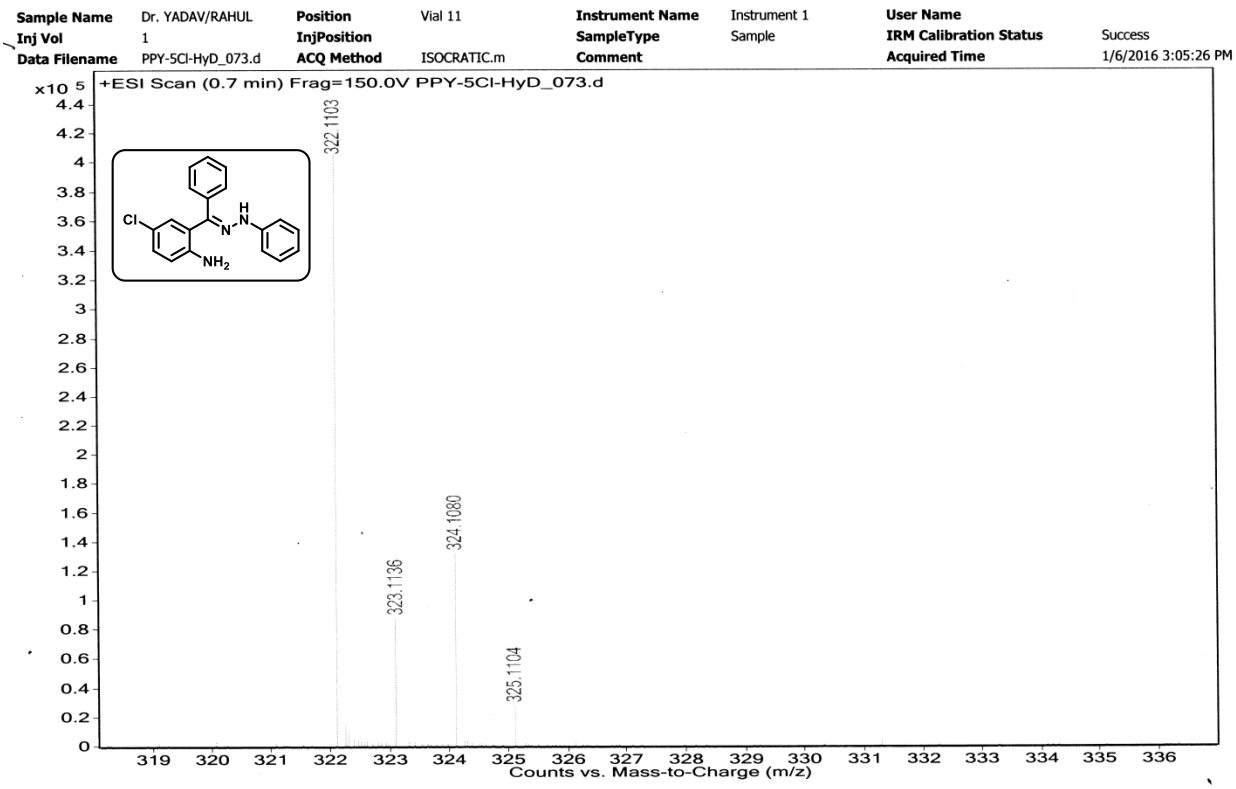

**HRMS spectrum of 1a**




**$^{1}$H NMR spectrum of 1b**




**$^{13}$C NMR spectrum of 1b**

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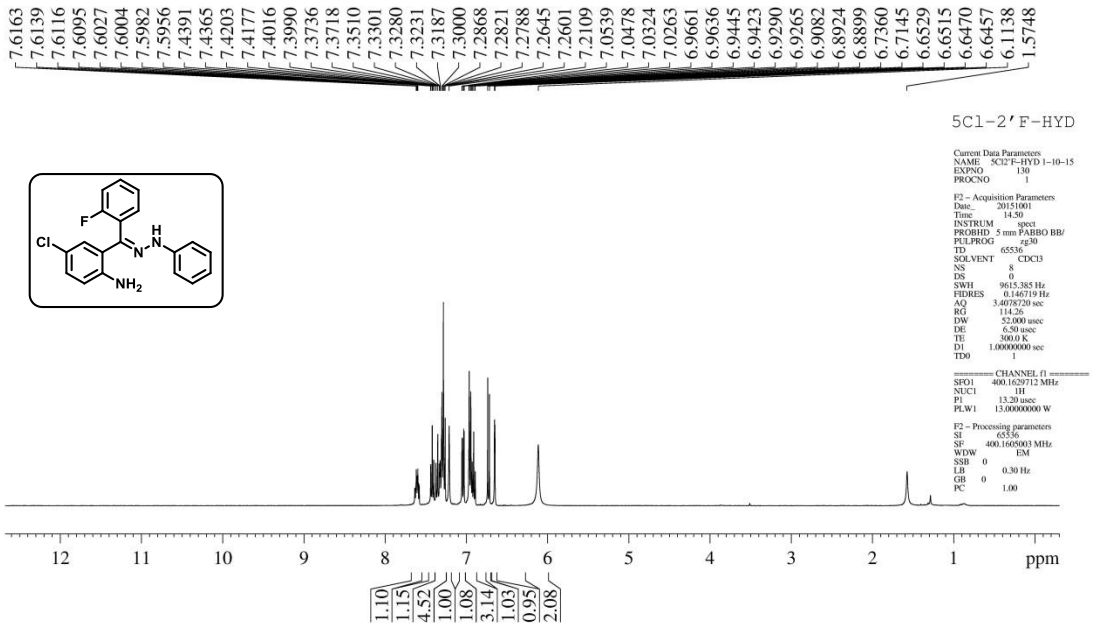
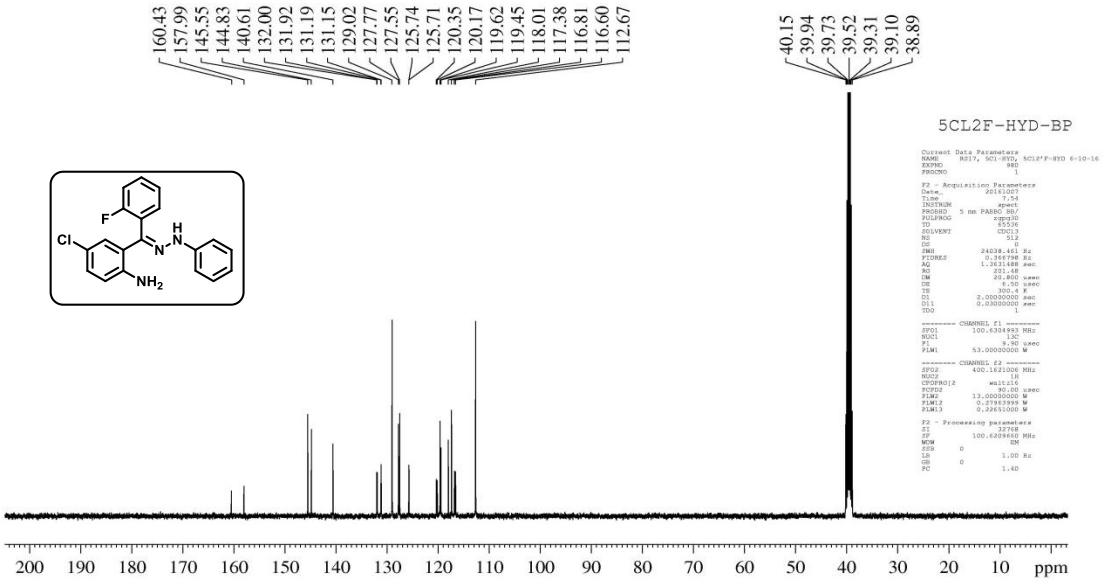
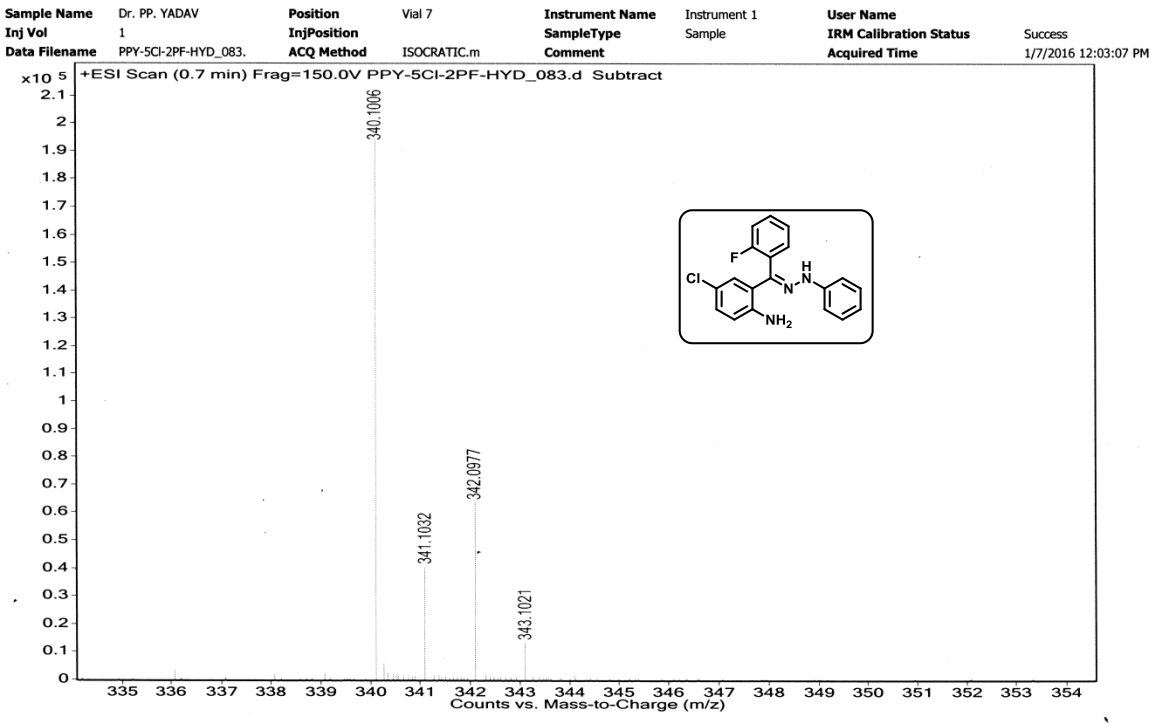


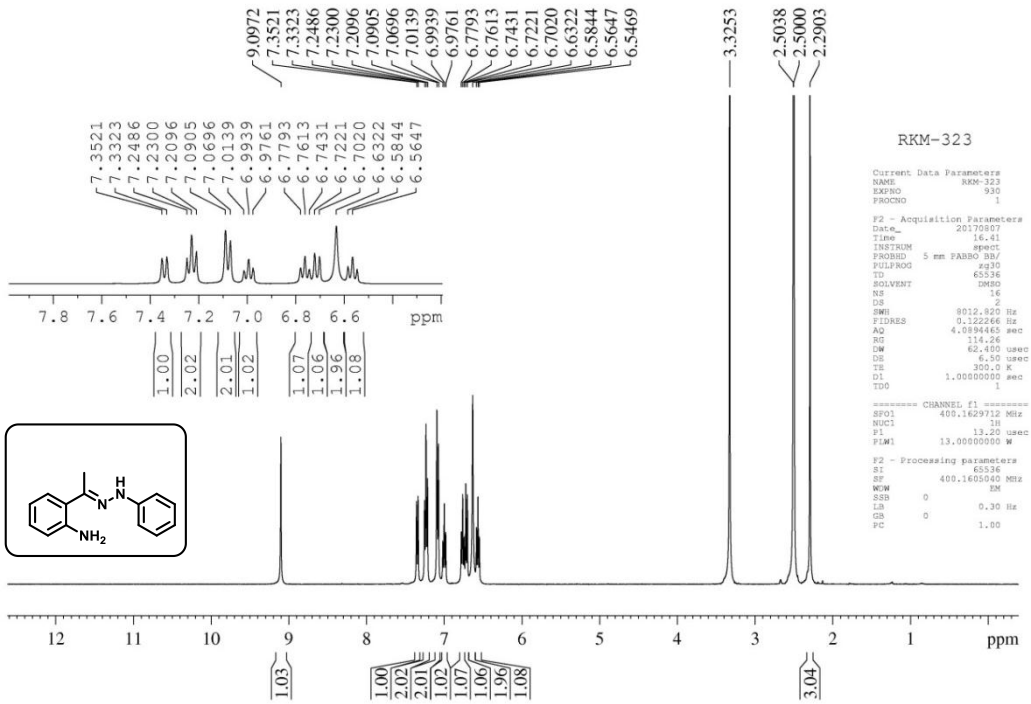

**$^{1}$H NMR spectrum of 1c**



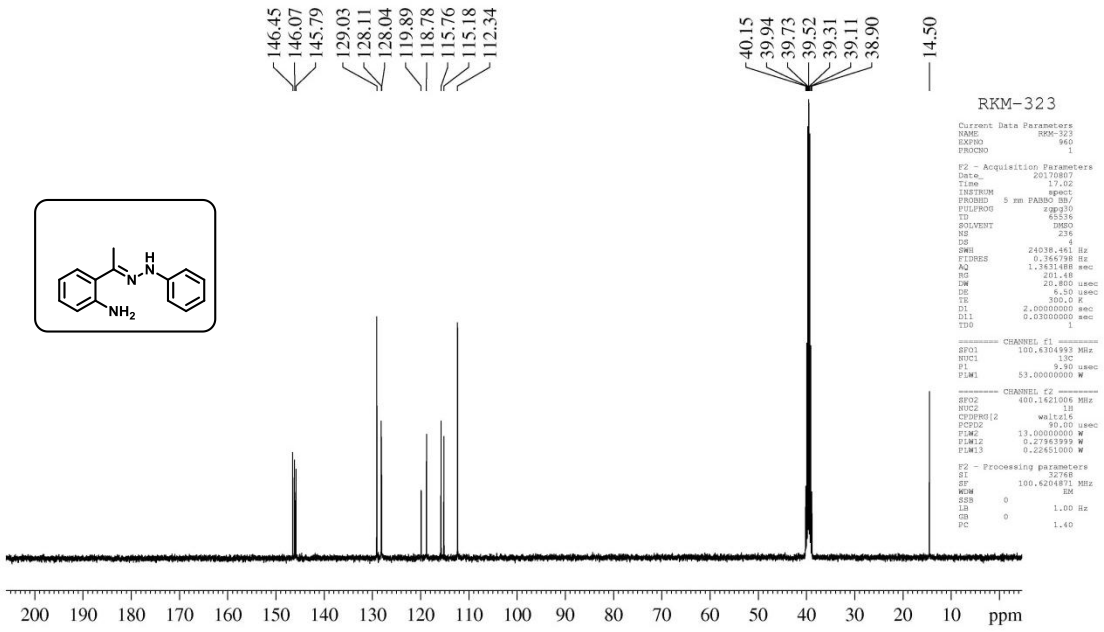

**$^{13}$C NMR spectrum of 1c**



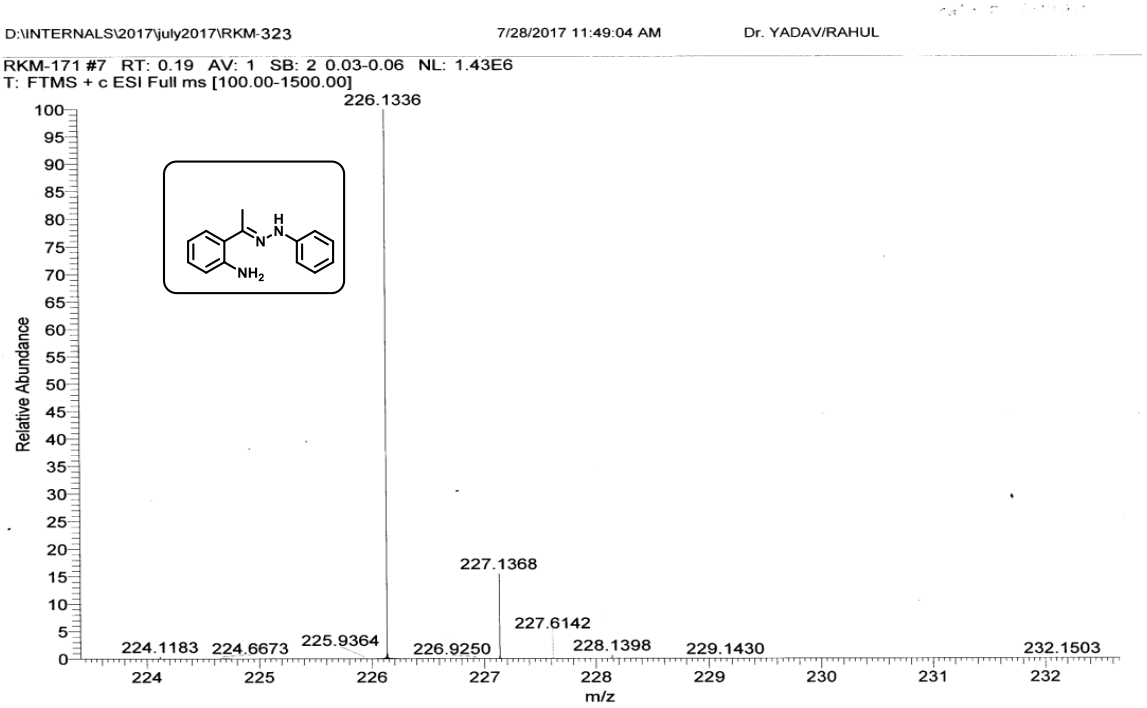

**HRMS spectrum of 1c**



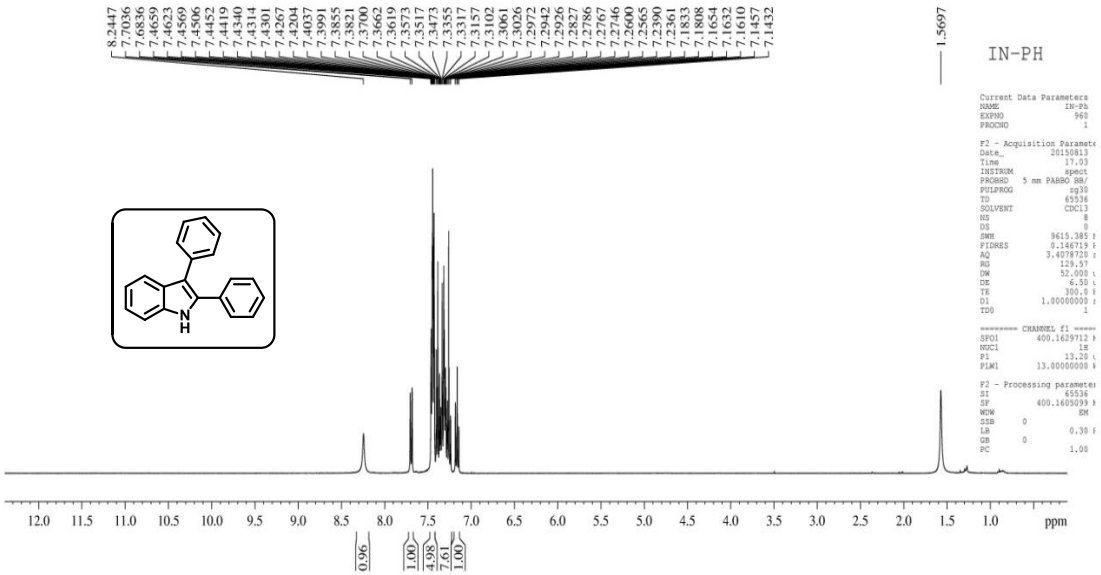

**$^{1}$H NMR spectrum of 1d**



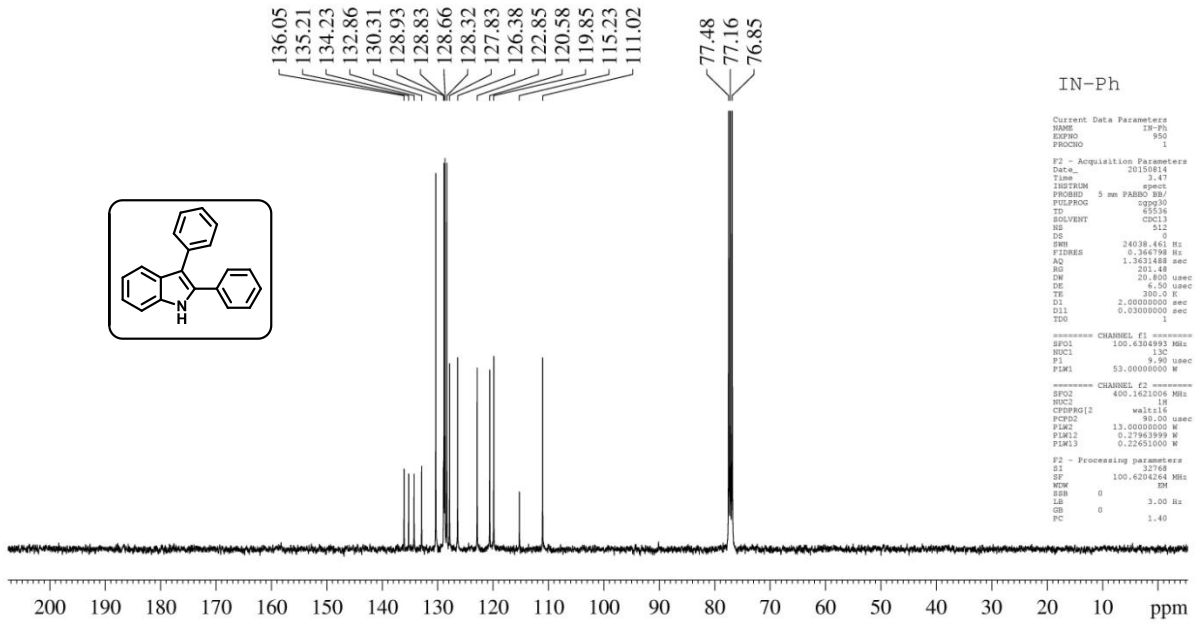

**$^{13}$C NMR spectrum of 1d**



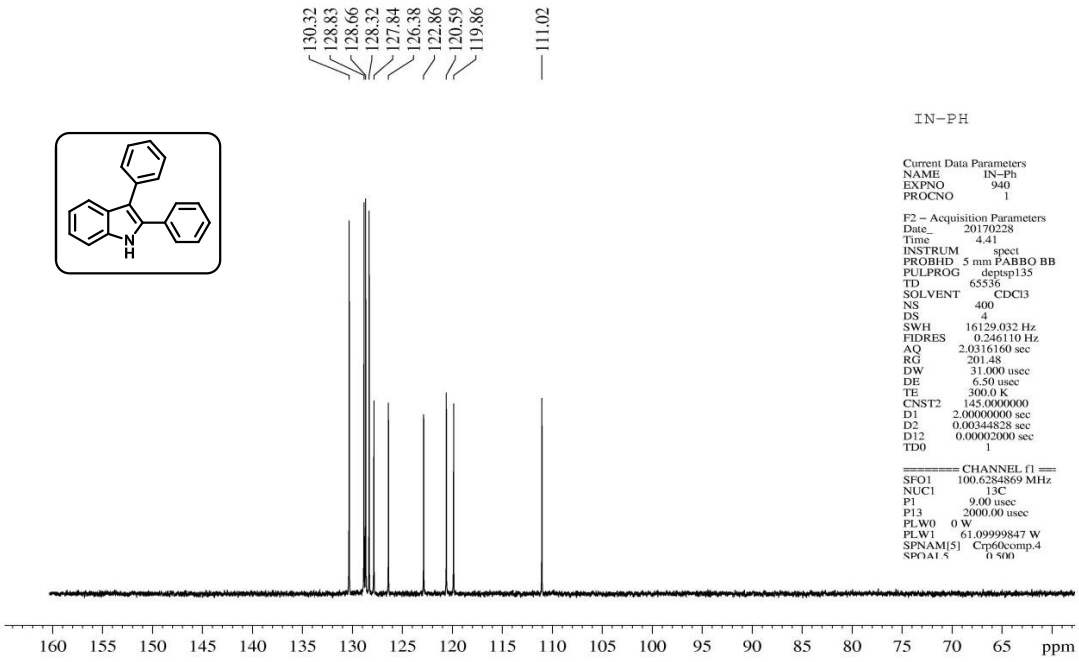

**HRMS spectrum of 1d**




**$^{1}$H NMR spectrum of 3aa**




**$^{13}$C NMR spectrum of 3aa**

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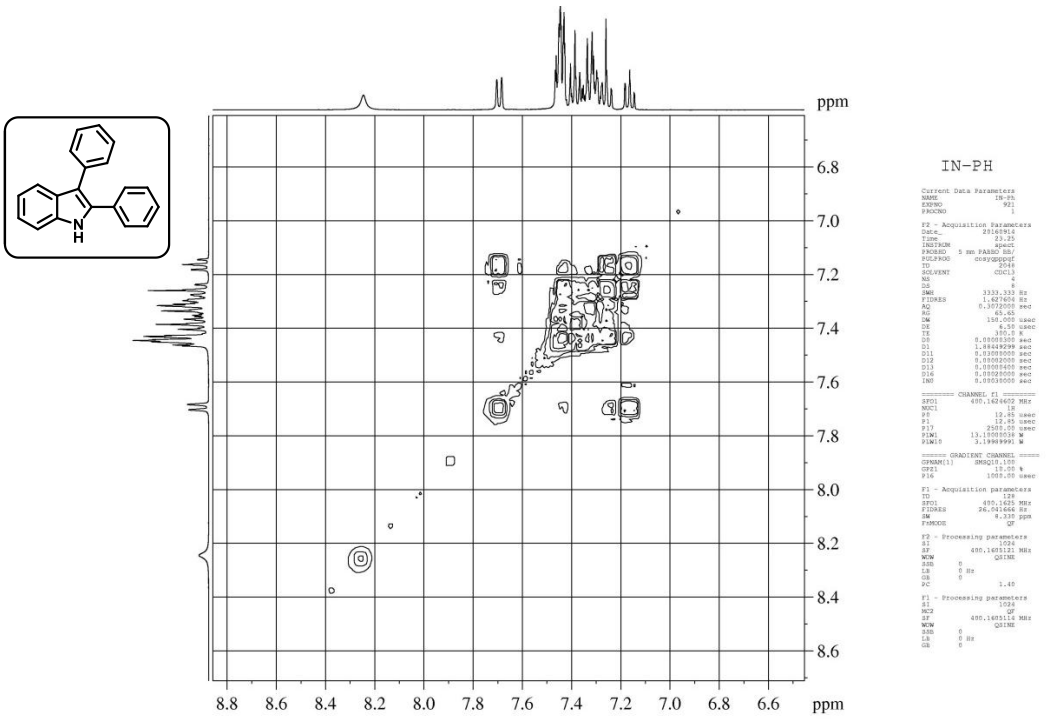
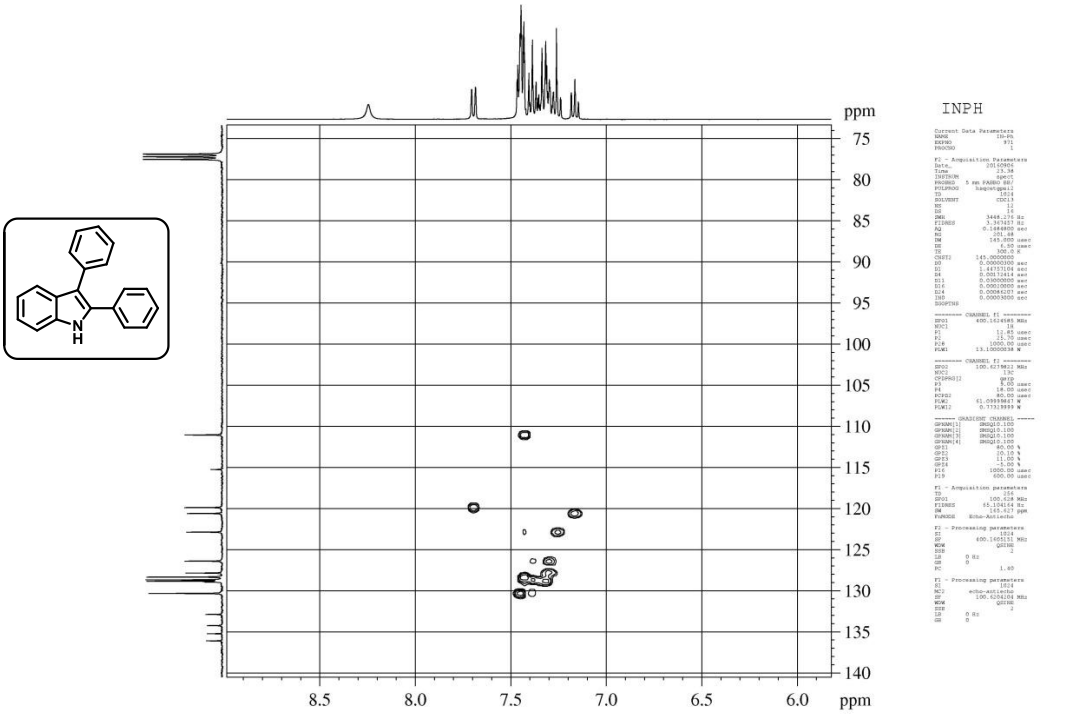
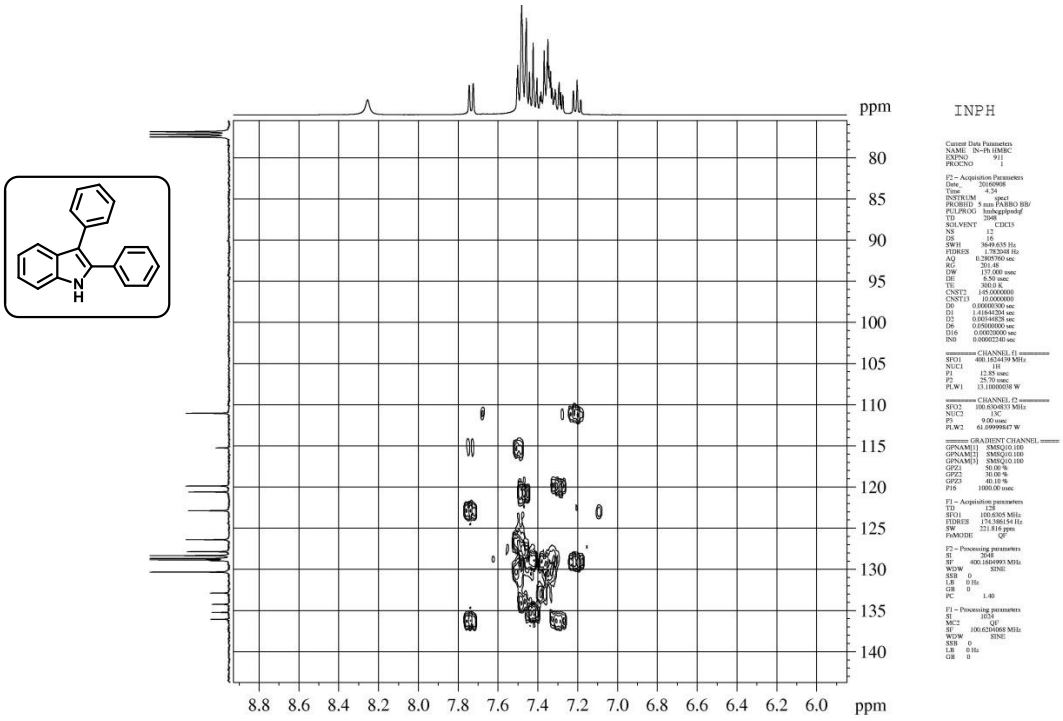



**Expanded COSY spectrum of 3aa**




**Expanded HSQC spectrum of 3aa**

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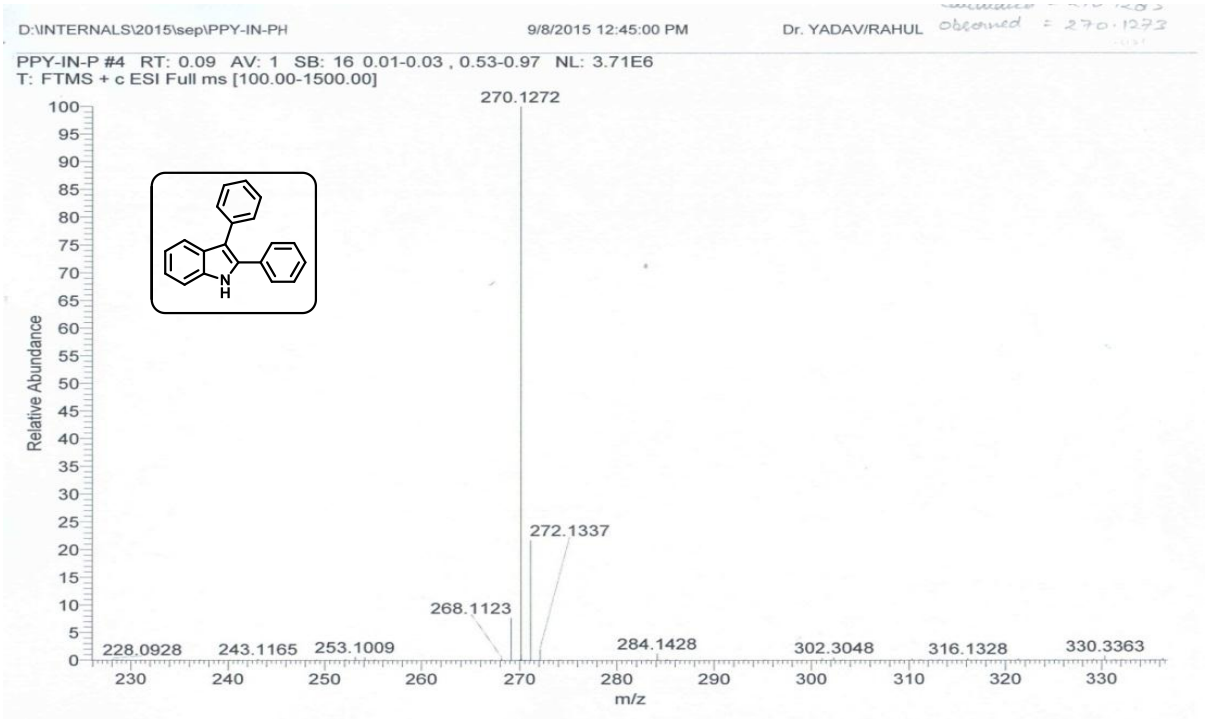
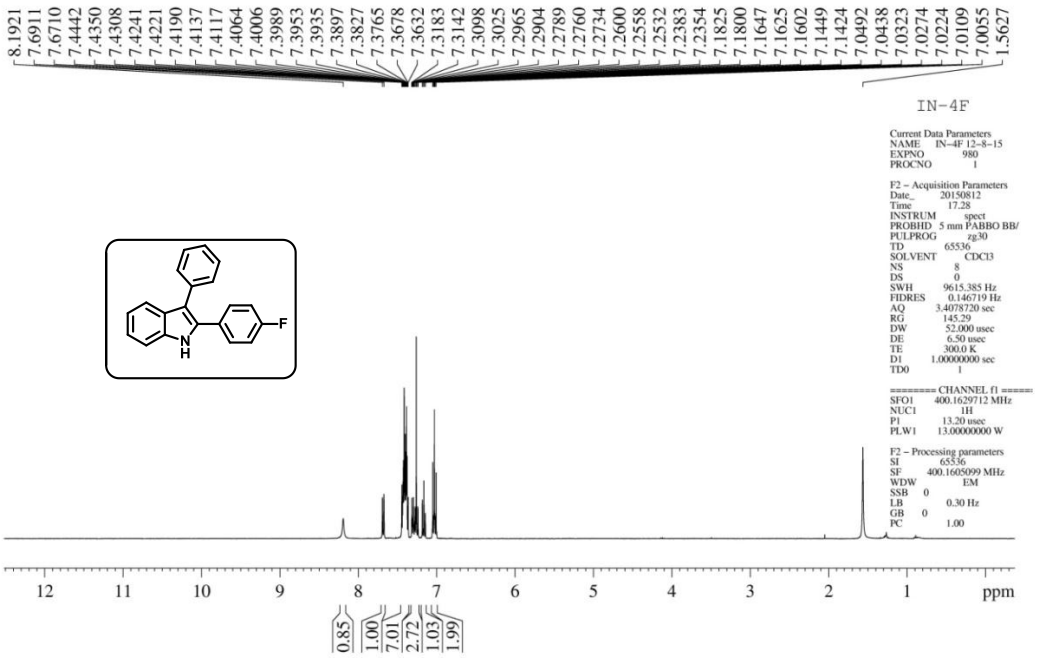
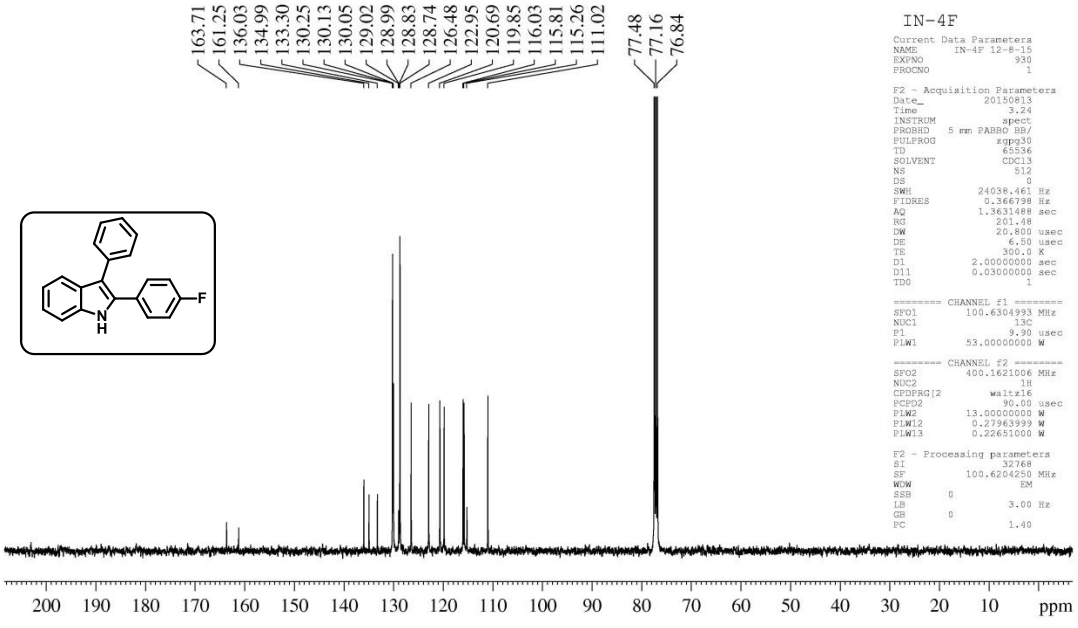



**HRMS spectrum of 3aa**




**$^{1}$H NMR spectrum of 3ab**

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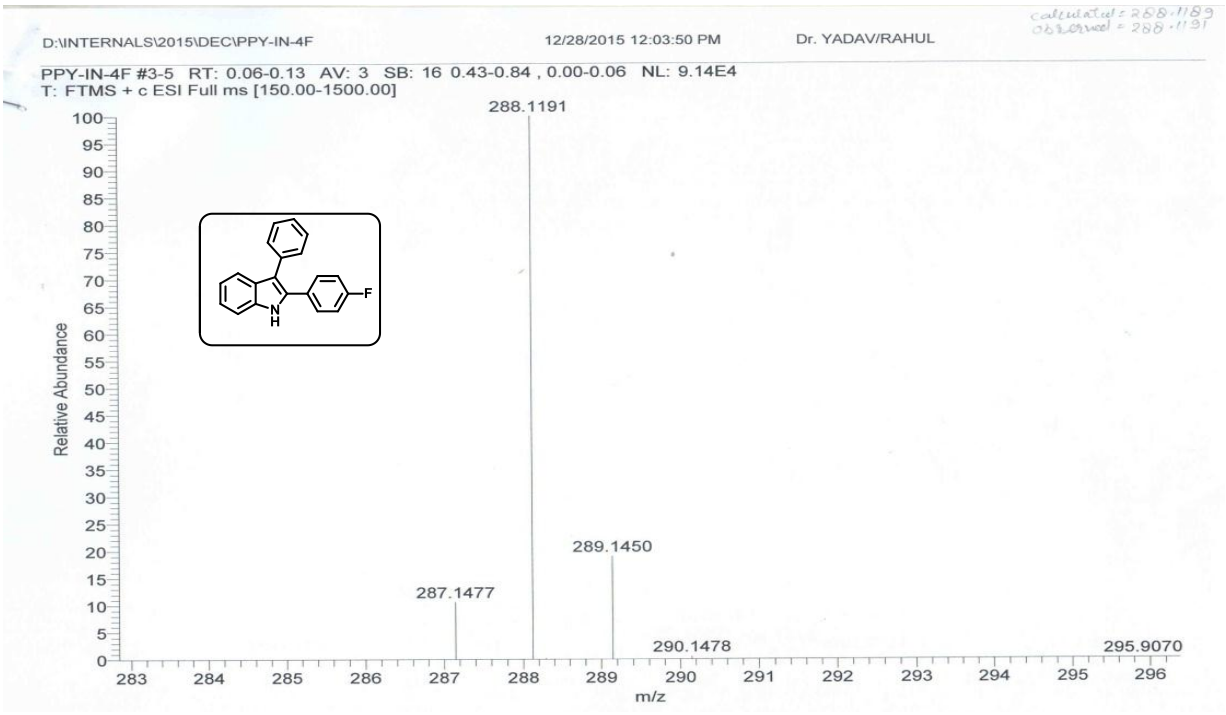
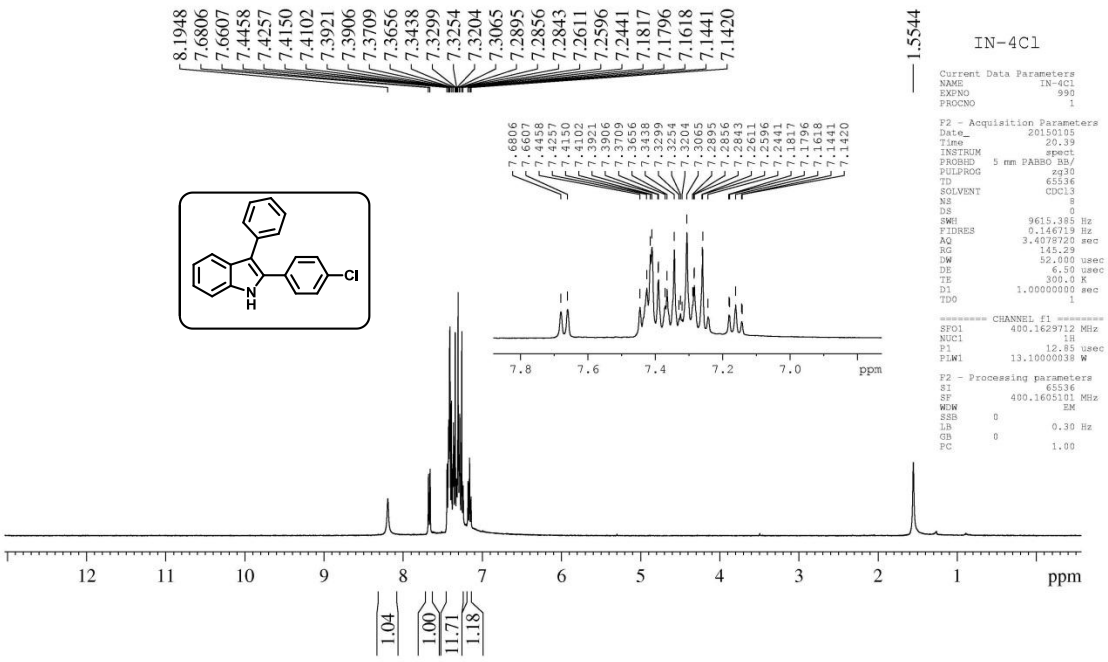
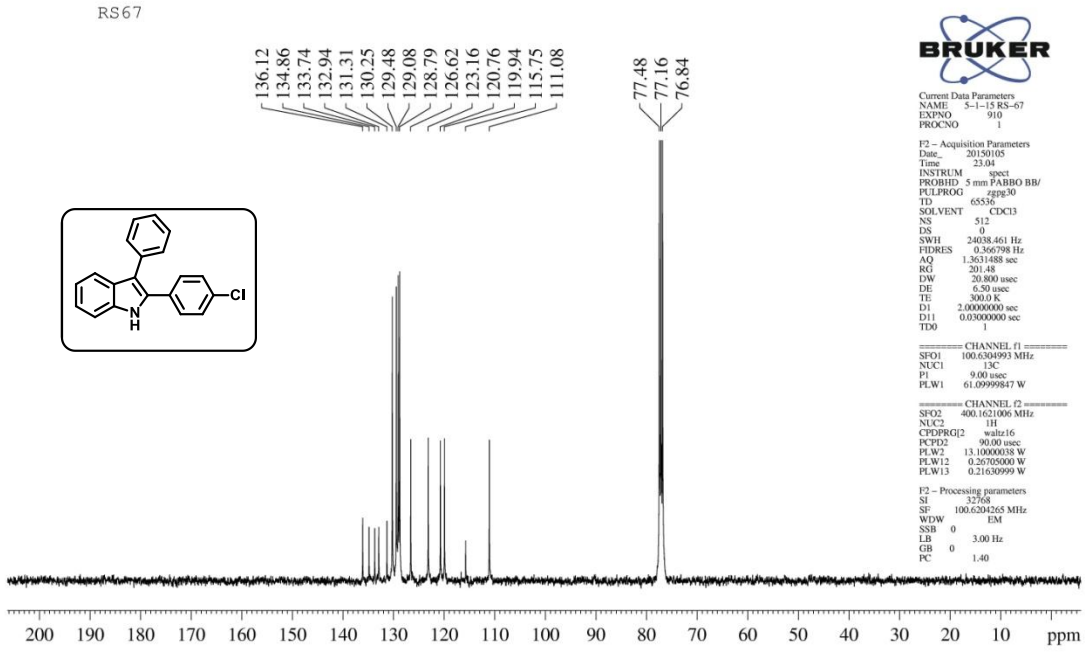


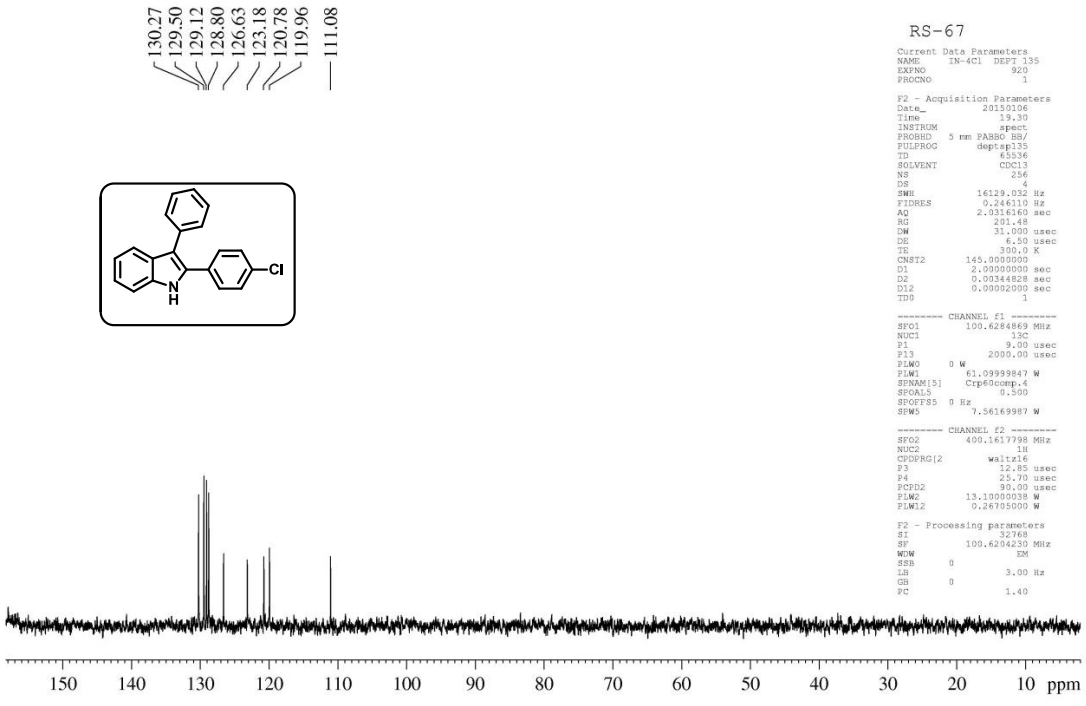

**HRMS spectrum of 3ab**



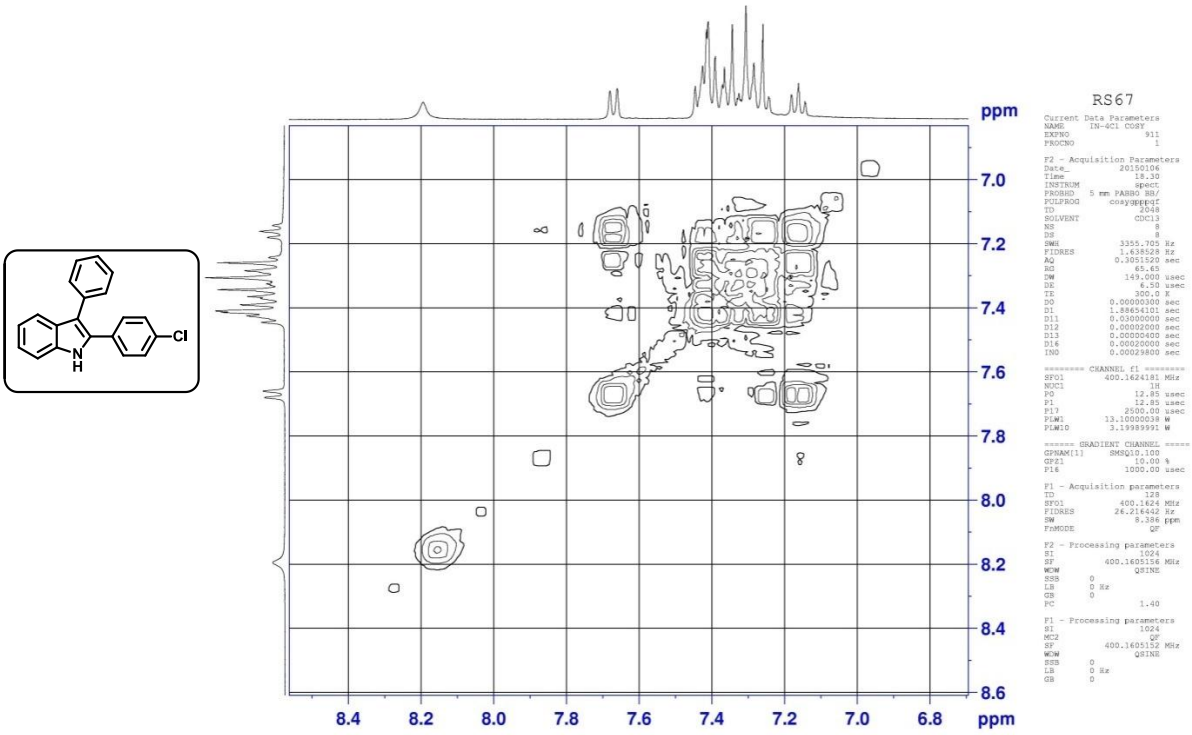

**$^{1}$H NMR spectrum of 3ac**



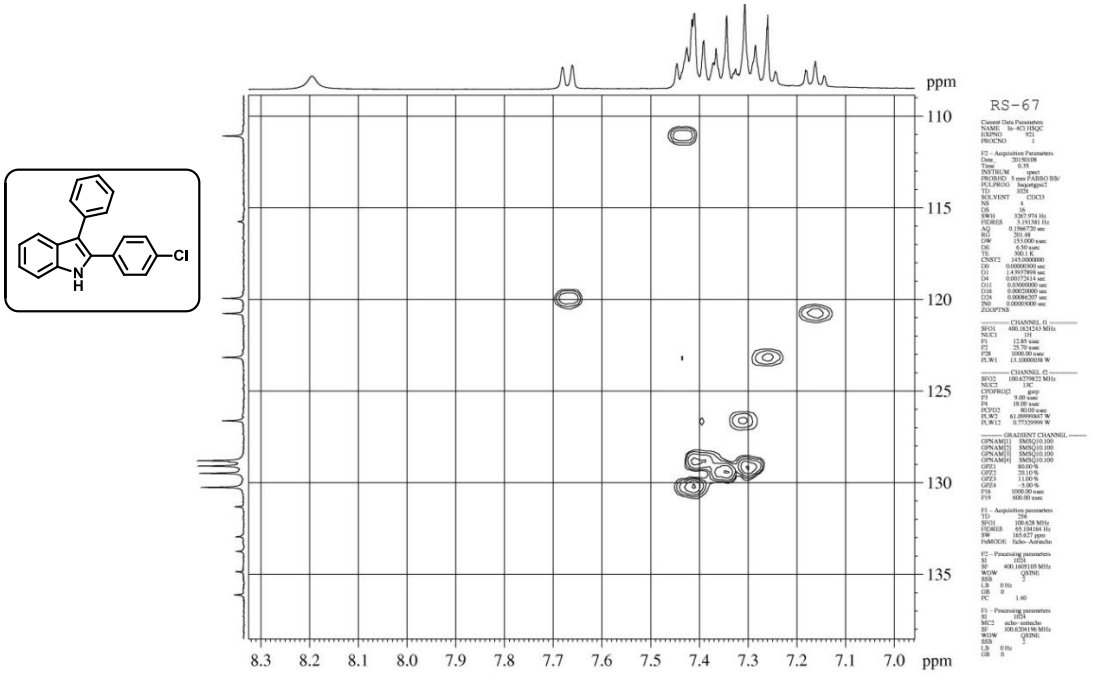

**$^{13}$C NMR spectrum of 3ac**



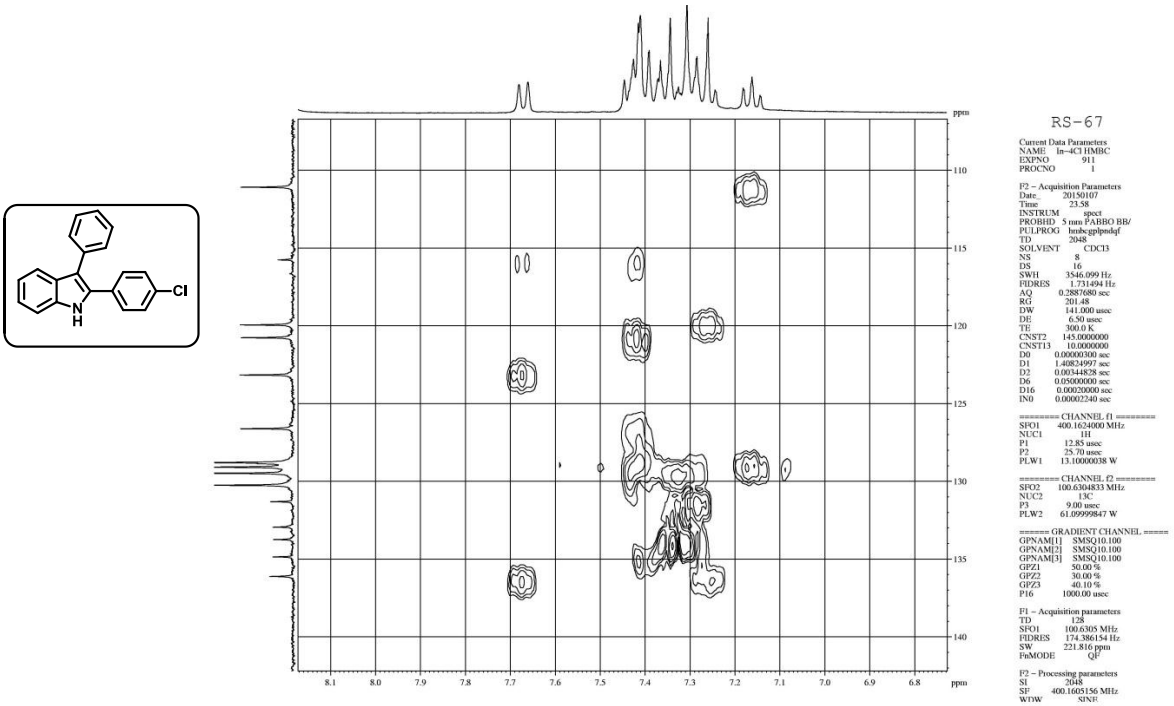

**DEPT 135 spectrum of 3ac**



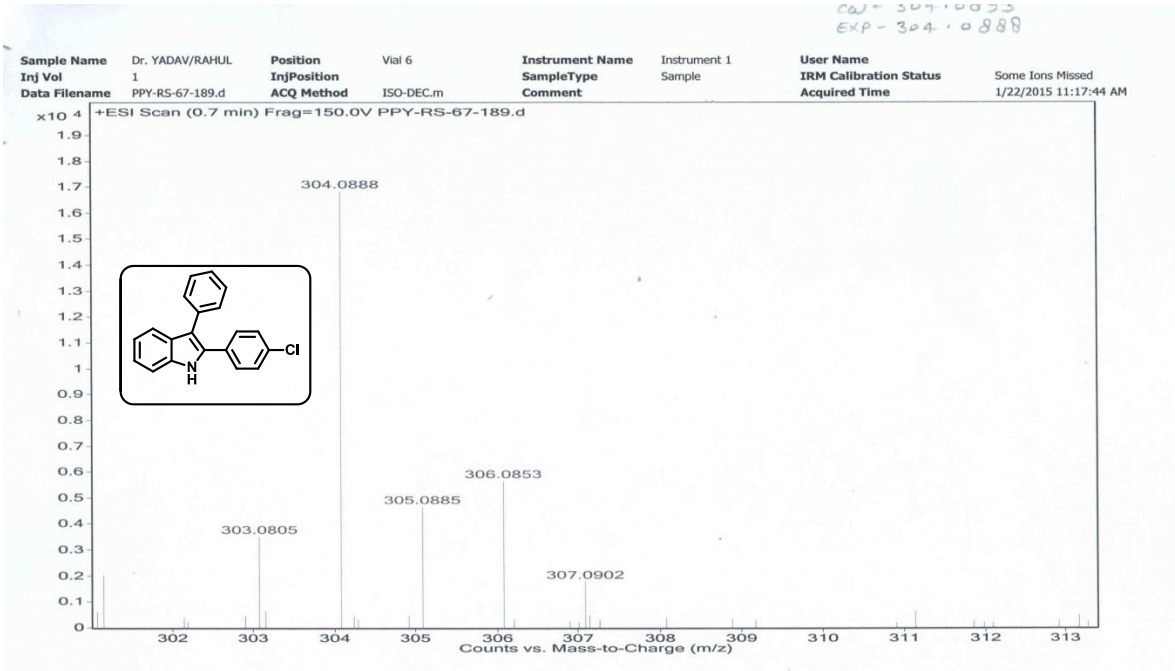

**Expanded COSY spectrum of 3ac**



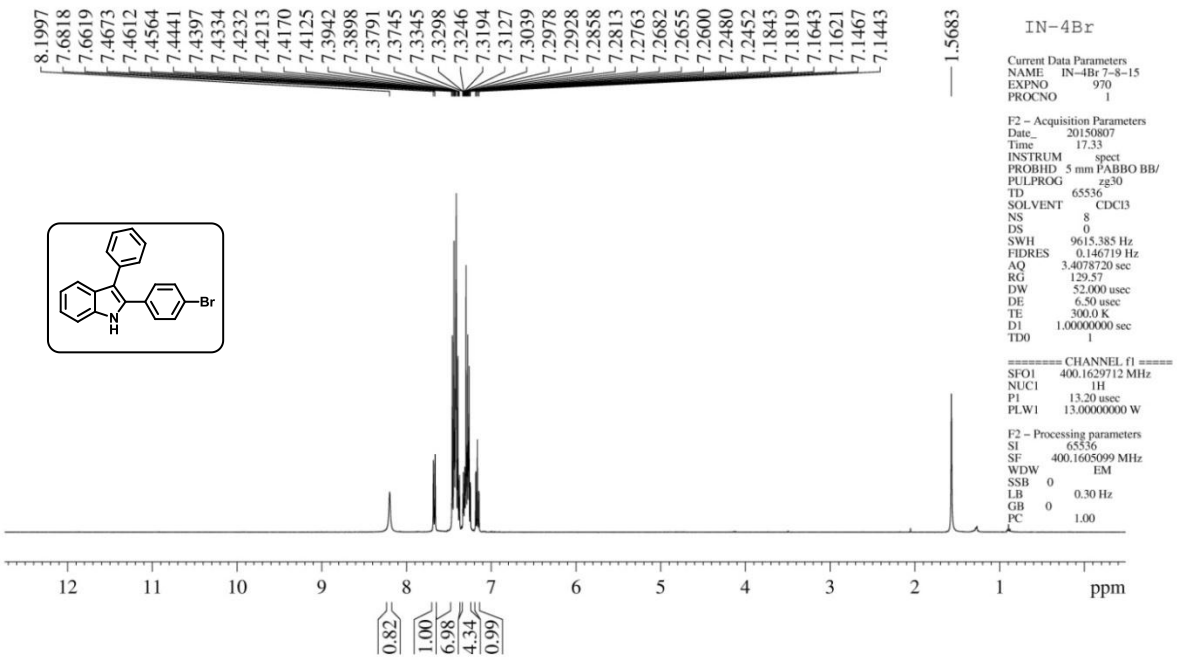

**Expanded HSQC spectrum of 3ac**



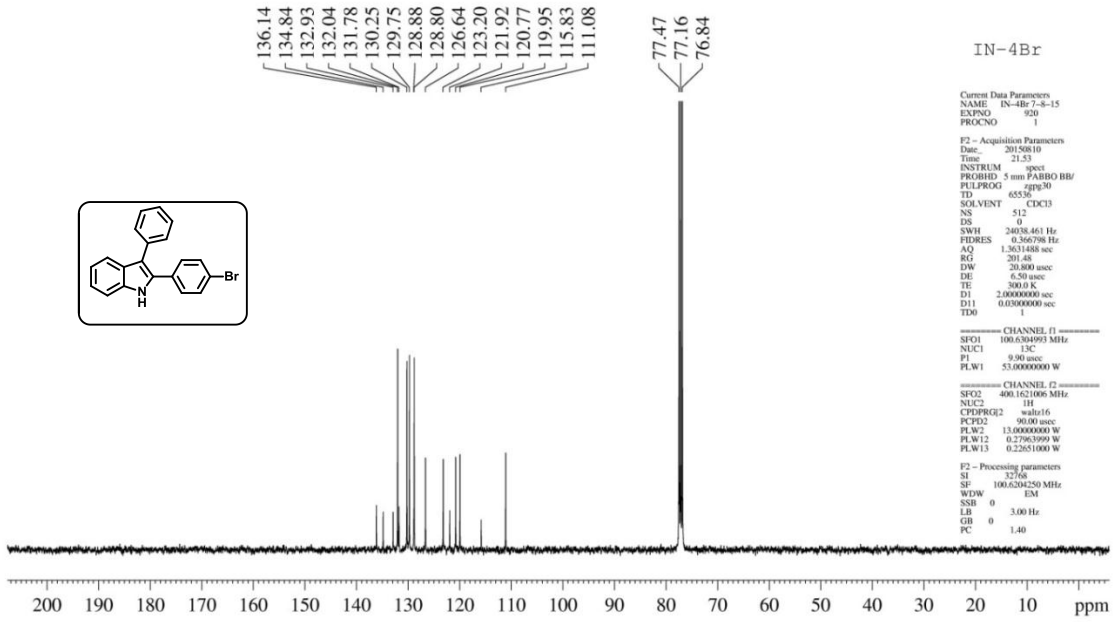

**Expanded HMBC spectrum of 3ac**



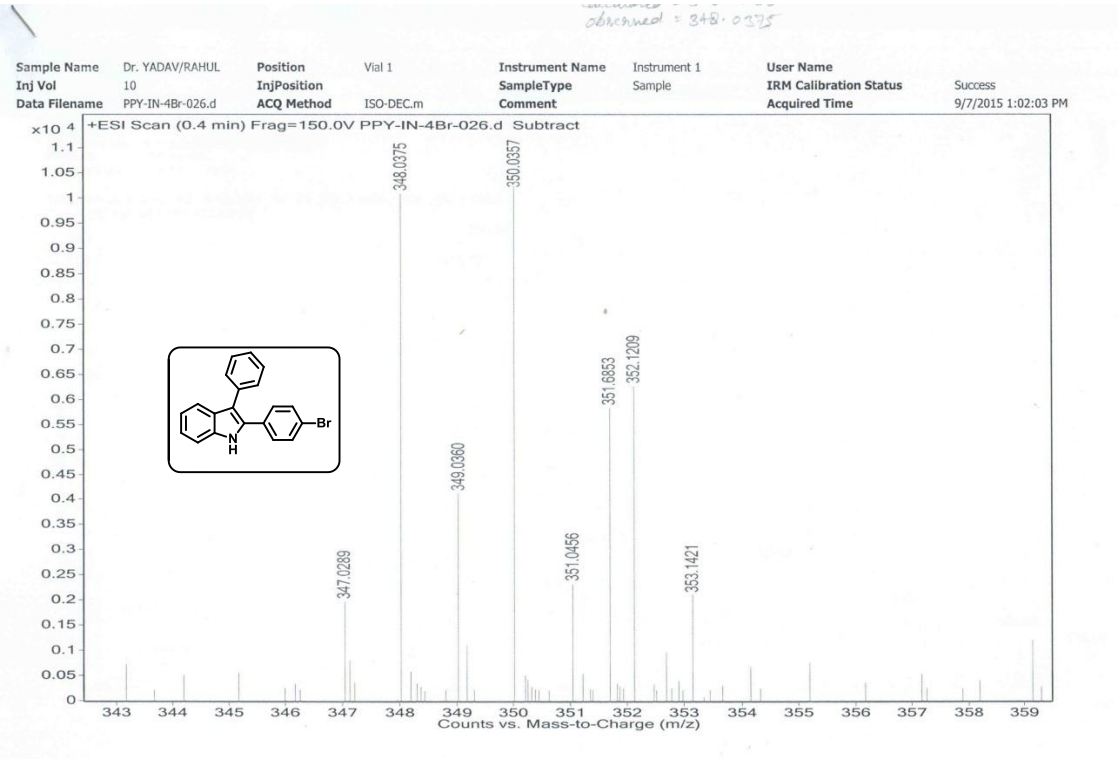

**HRMS spectrum of 3ac**



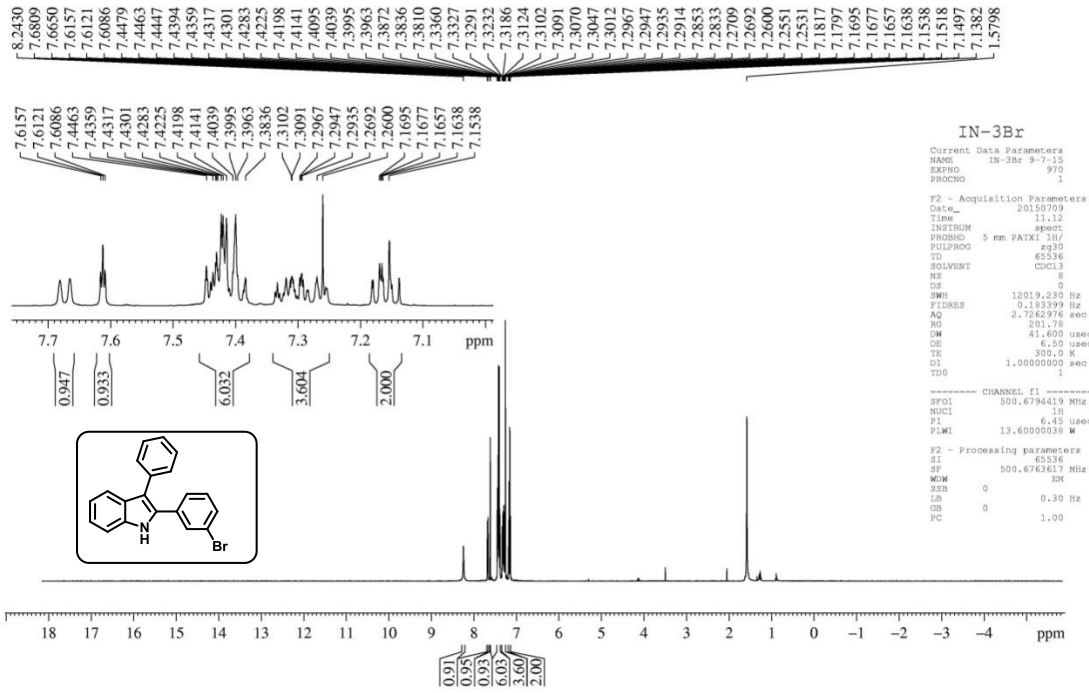

**$^{1}$H NMR spectrum of 3ae**



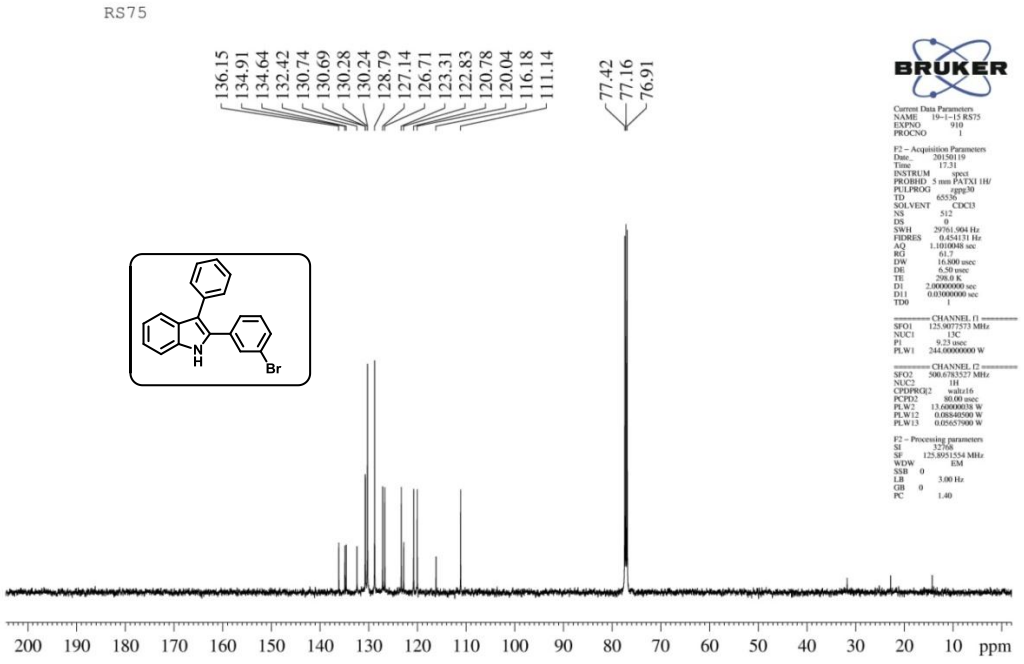

**$^{13}$C NMR spectrum of 3ae**



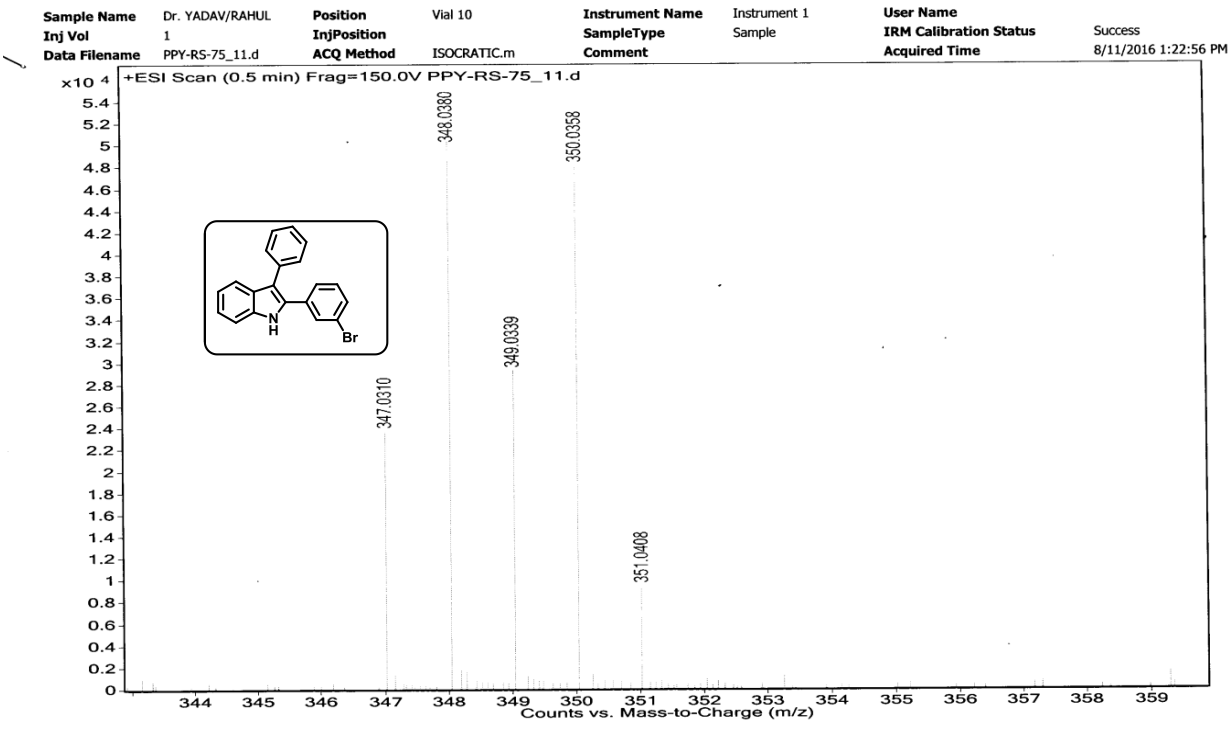

**HRMS spectrum of 3ae**



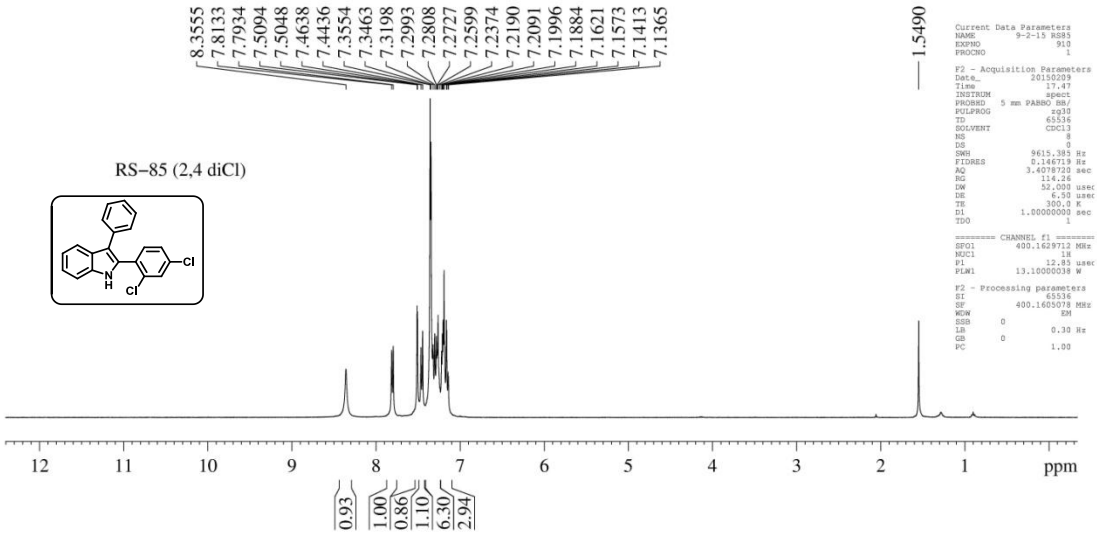

**$^{1}$H NMR spectrum of 3af**



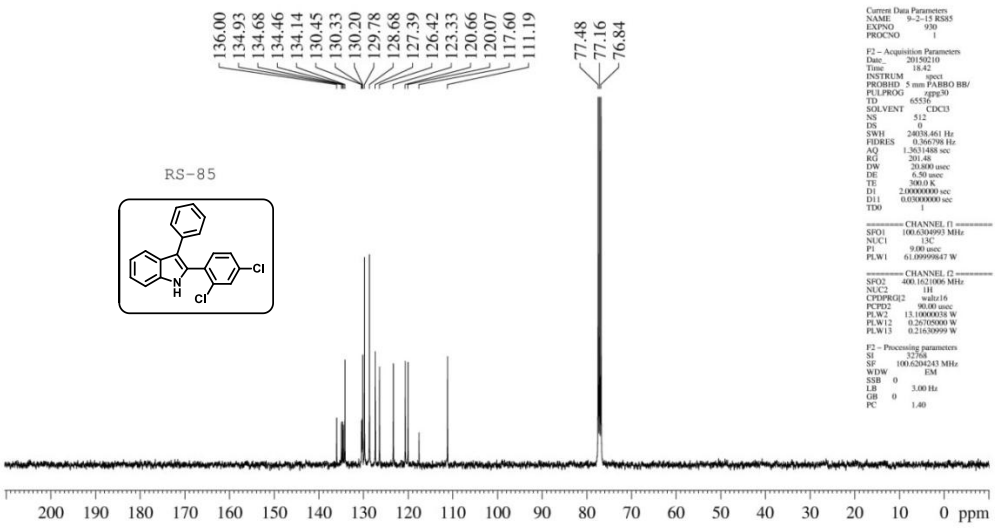

**$^{13}$C NMR spectrum of 3af**



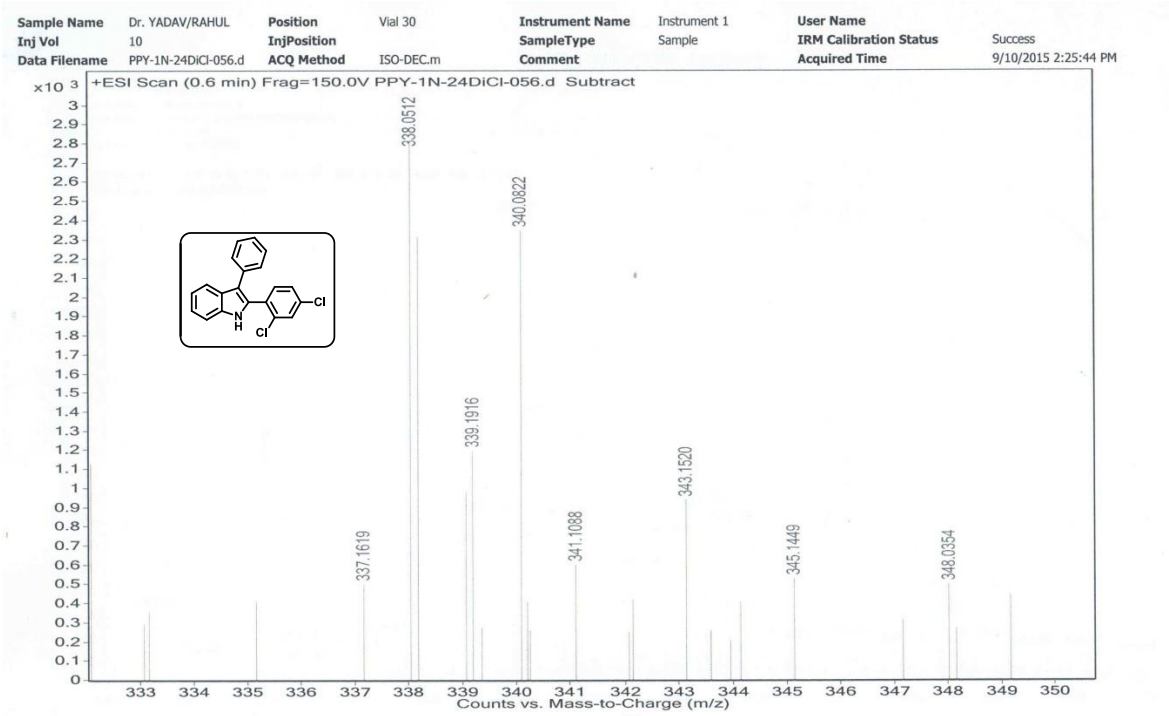

**HRMS spectrum of 3af**



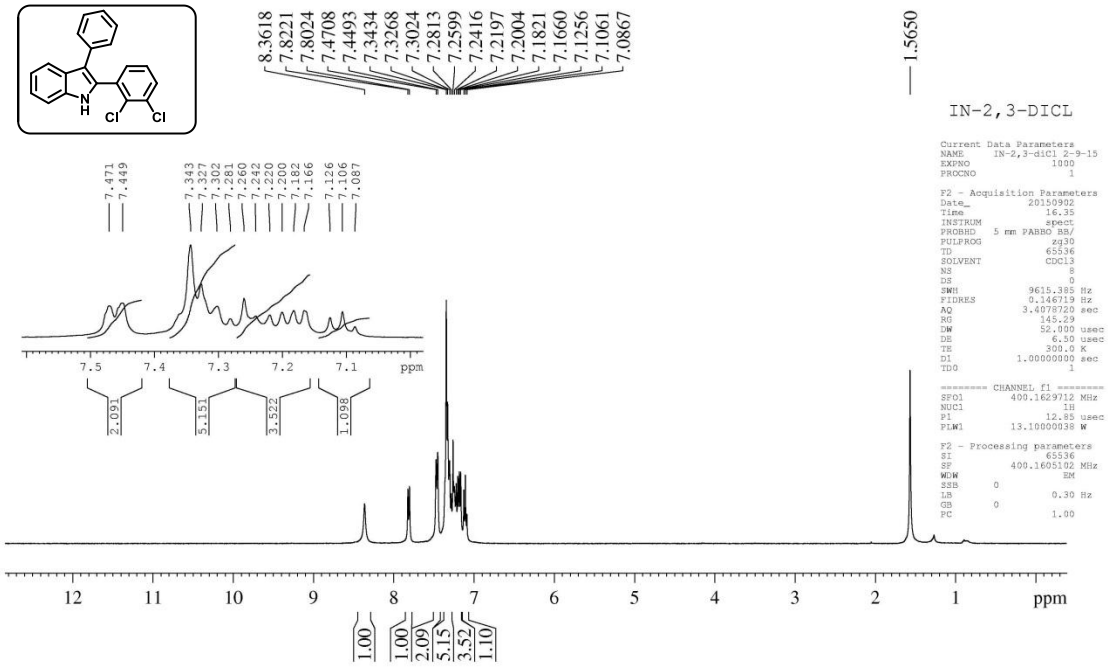

**$^{1}$H NMR spectrum of 3ag**



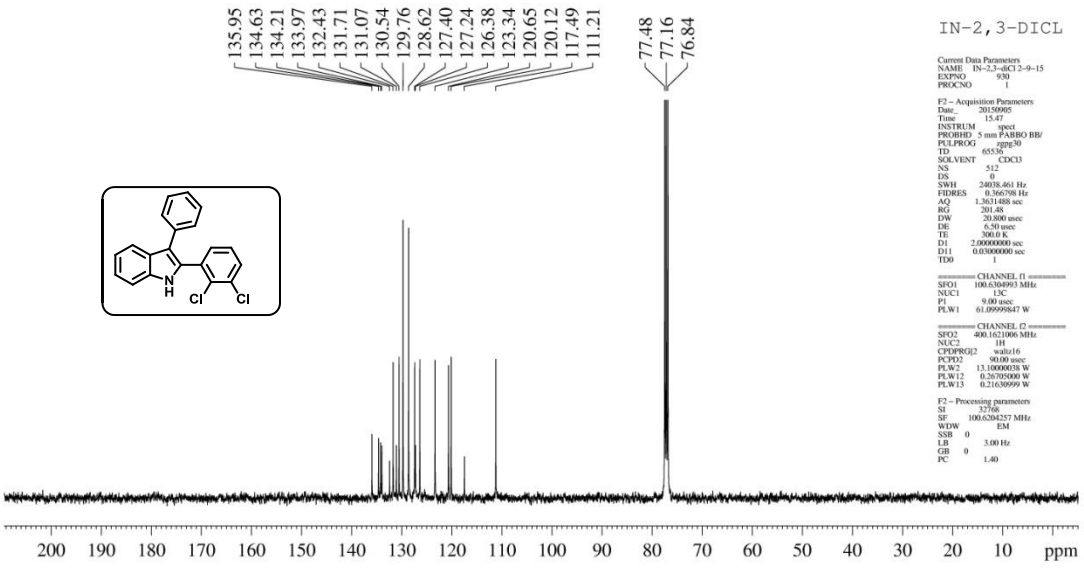

**$^{13}$C NMR spectrum of 3ag**



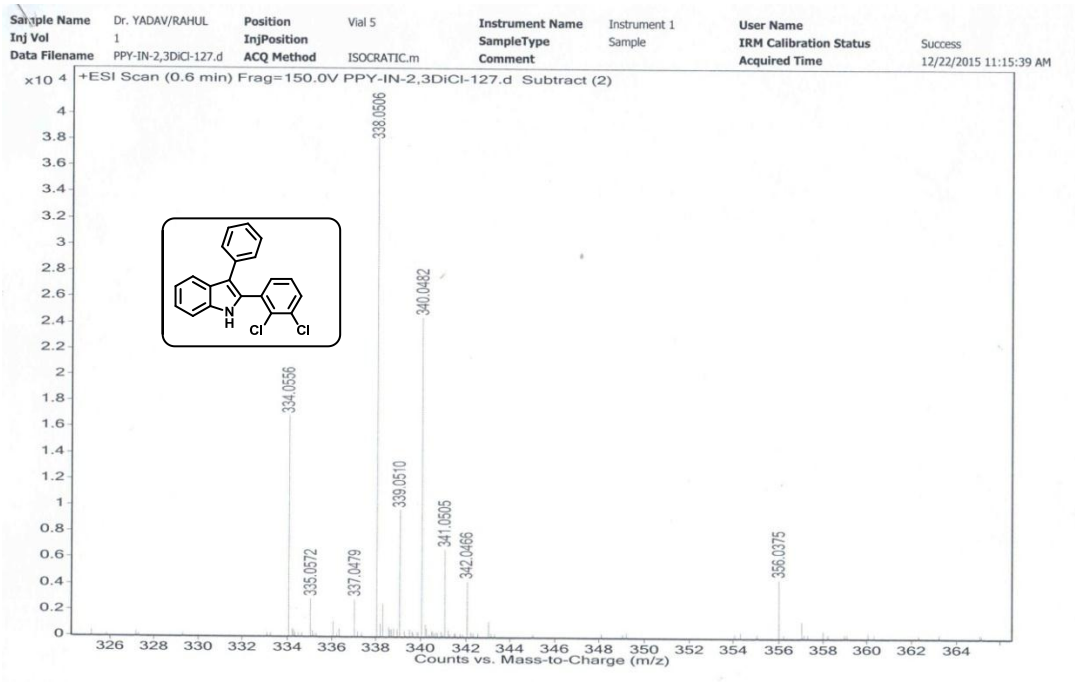

**HRMS spectrum of 3ag**



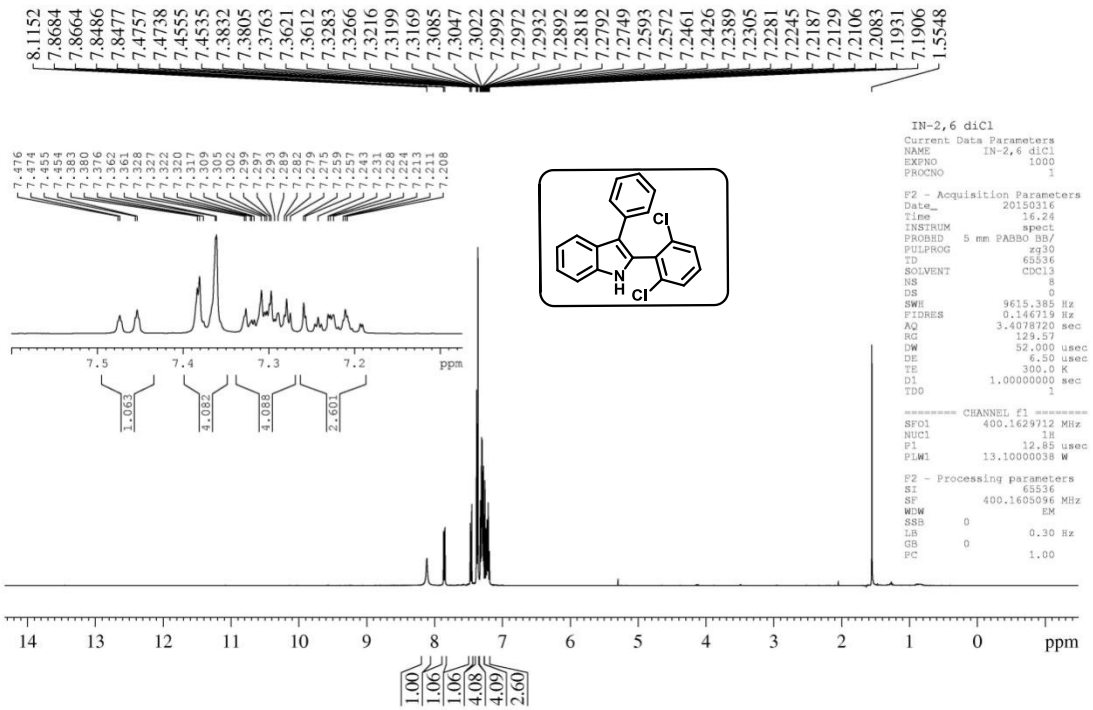

**$^{1}$H NMR spectrum of 3ah**



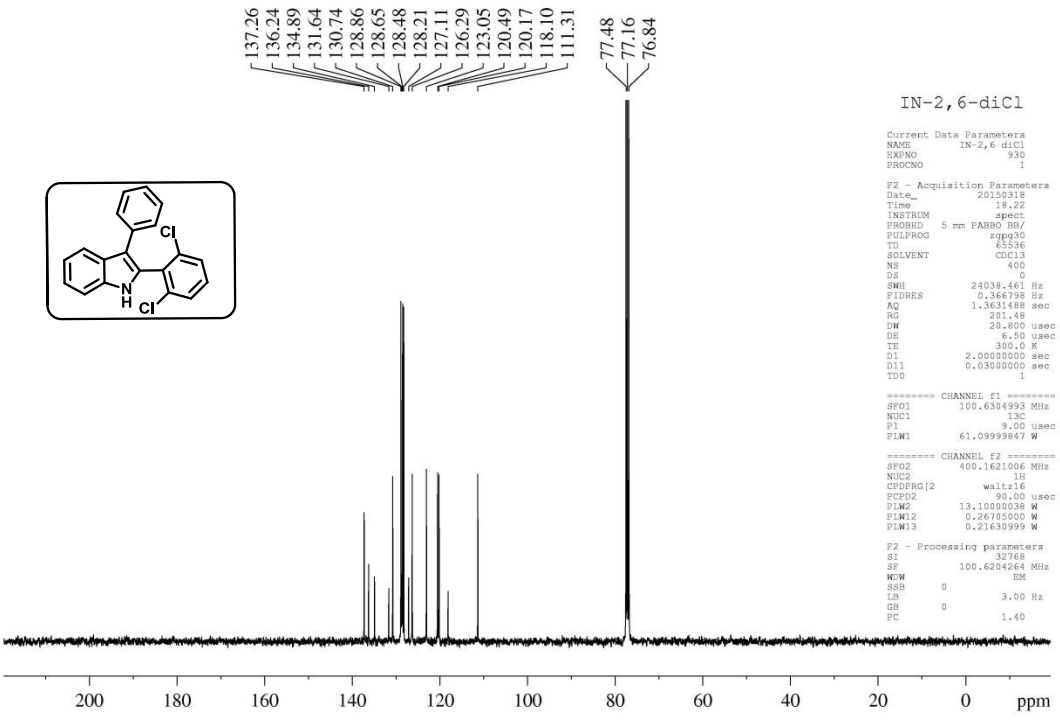

**$^{13}$C NMR spectrum of 3ah**



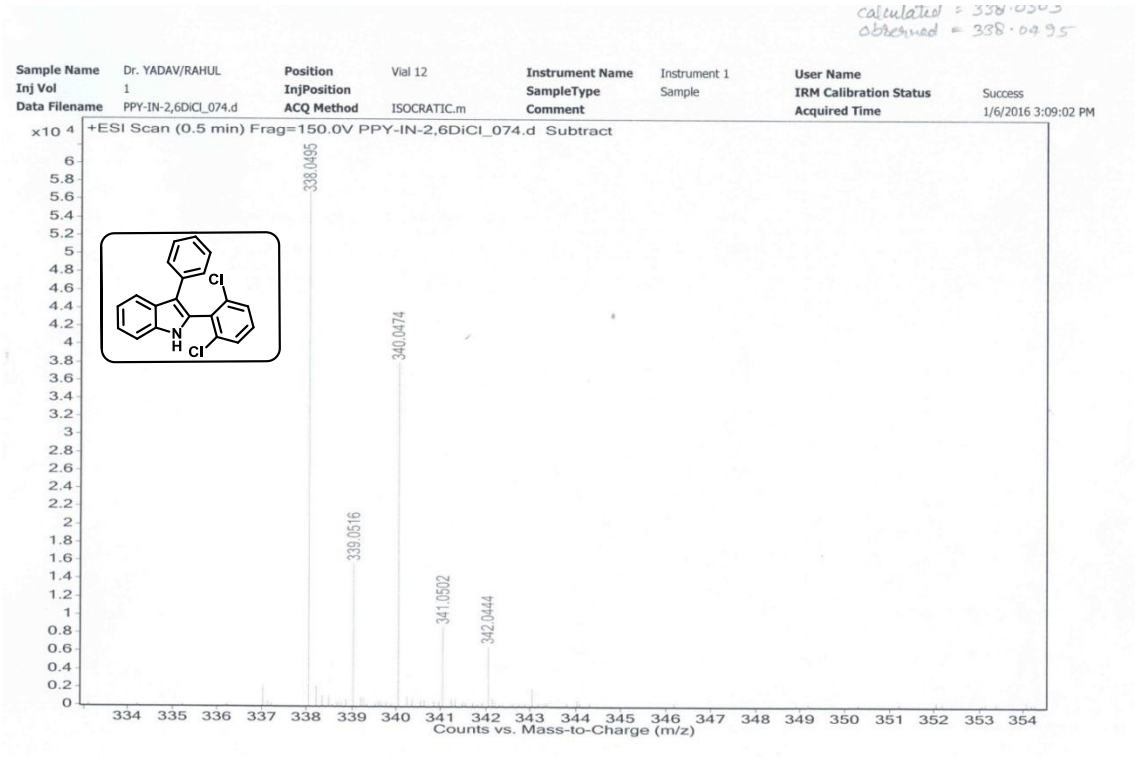

**HRMS spectrum of 3ah**



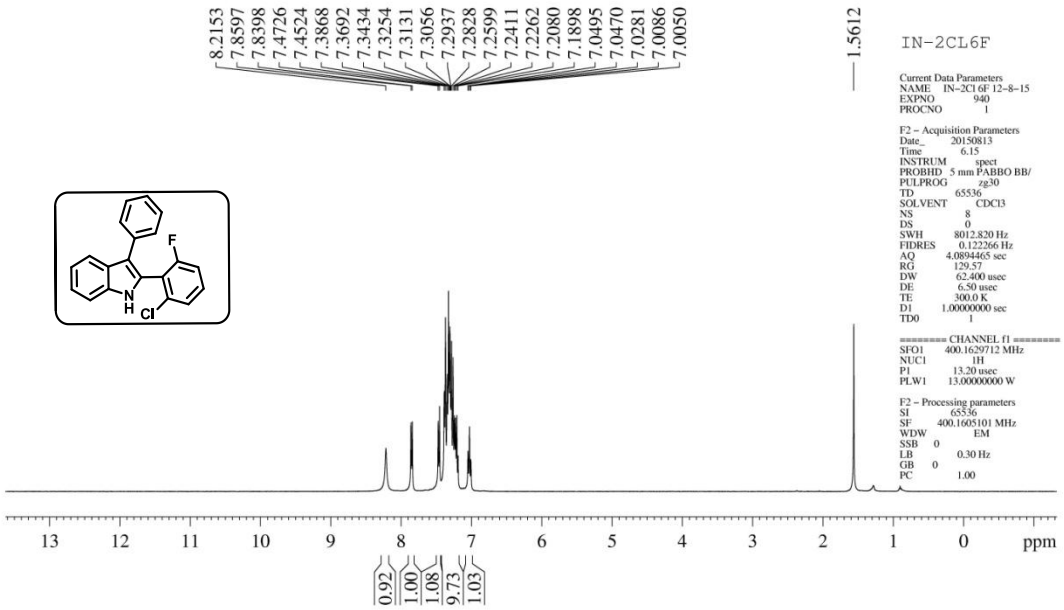

**$^{1}$H NMR spectrum of 3ai**



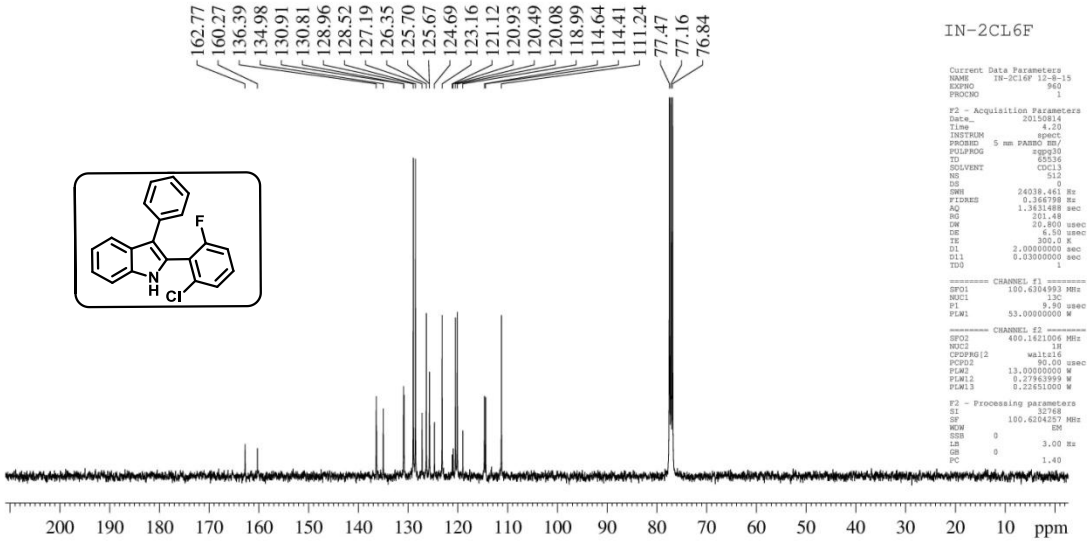

**$^{13}$C NMR spectrum of 3ai**



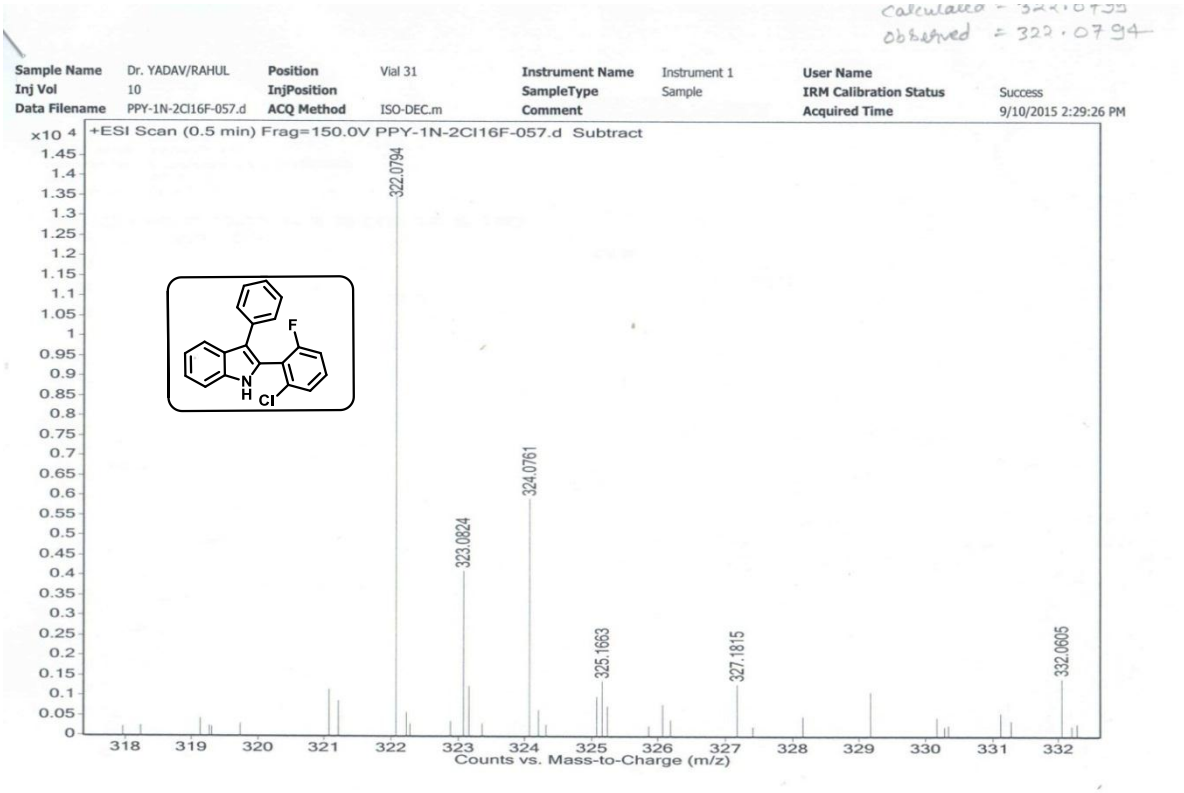

**HRMS spectrum of 3ai**



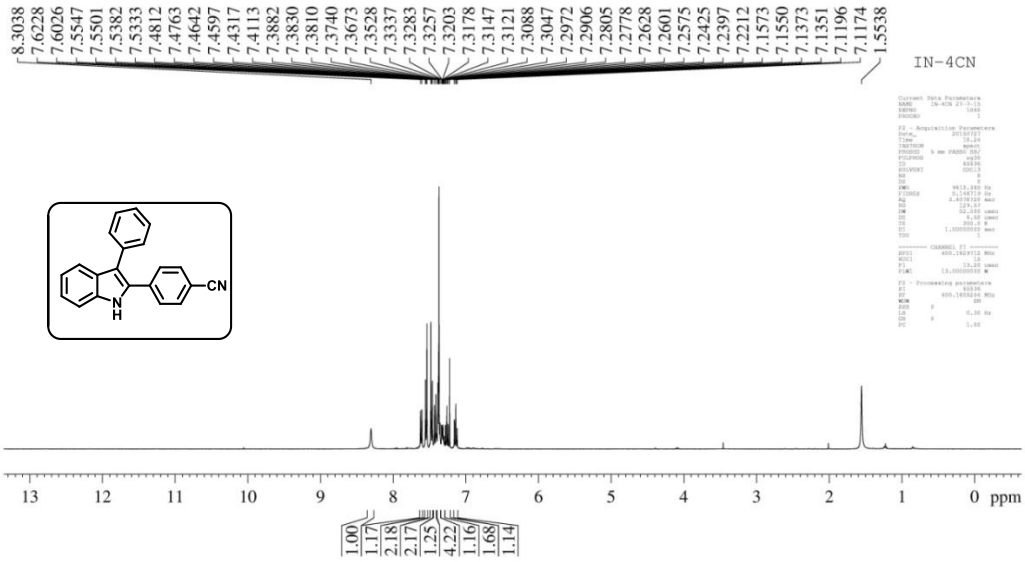

**$^{1}$H NMR spectrum of 3aj**



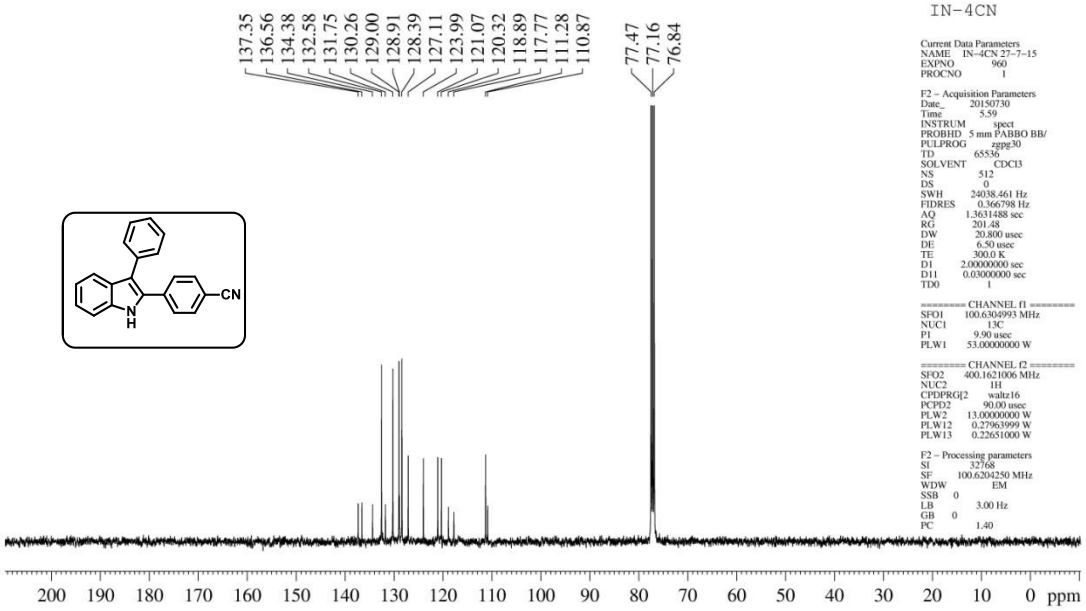

**$^{13}$C NMR spectrum of 3aj**



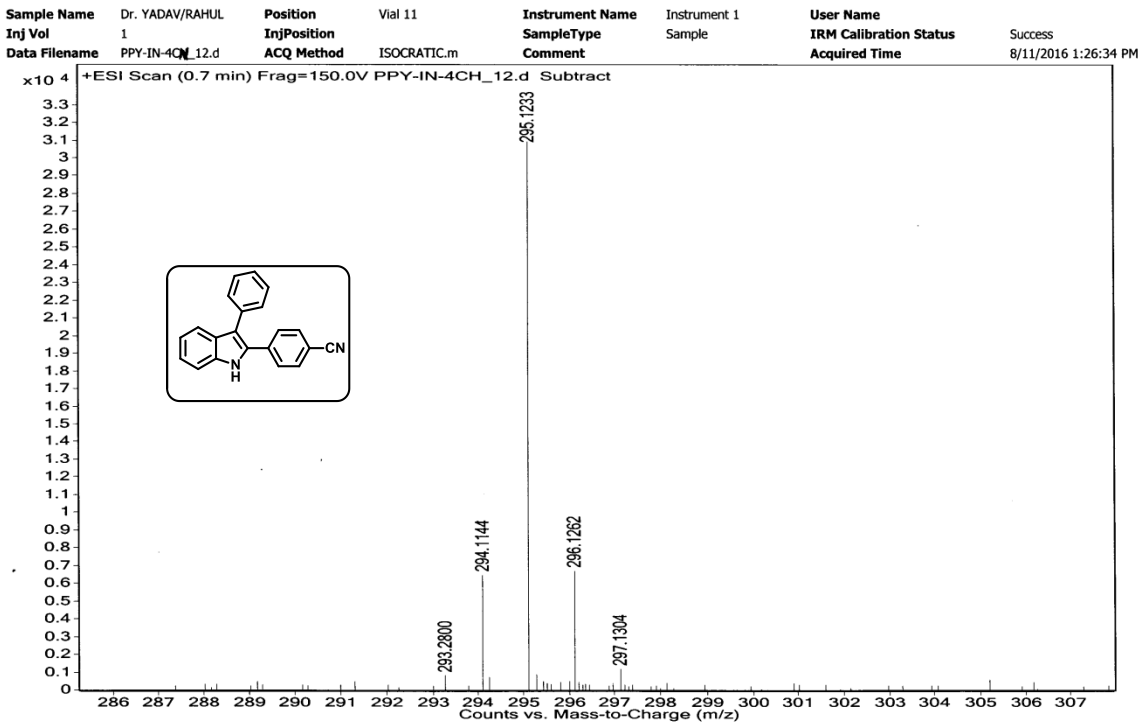

**HRMS spectrum of 3aj**



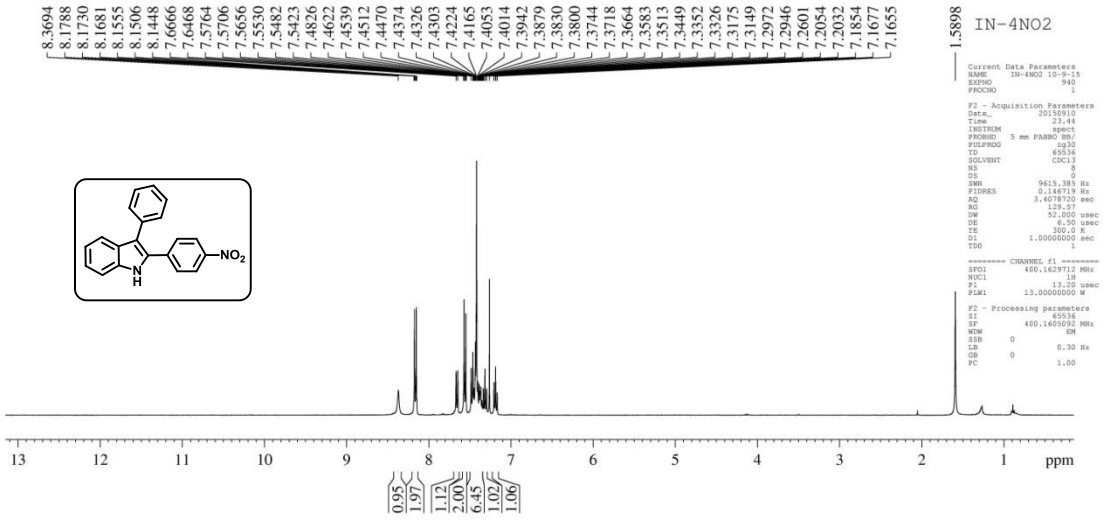

**[1]H NMR spectrum of 3ak**



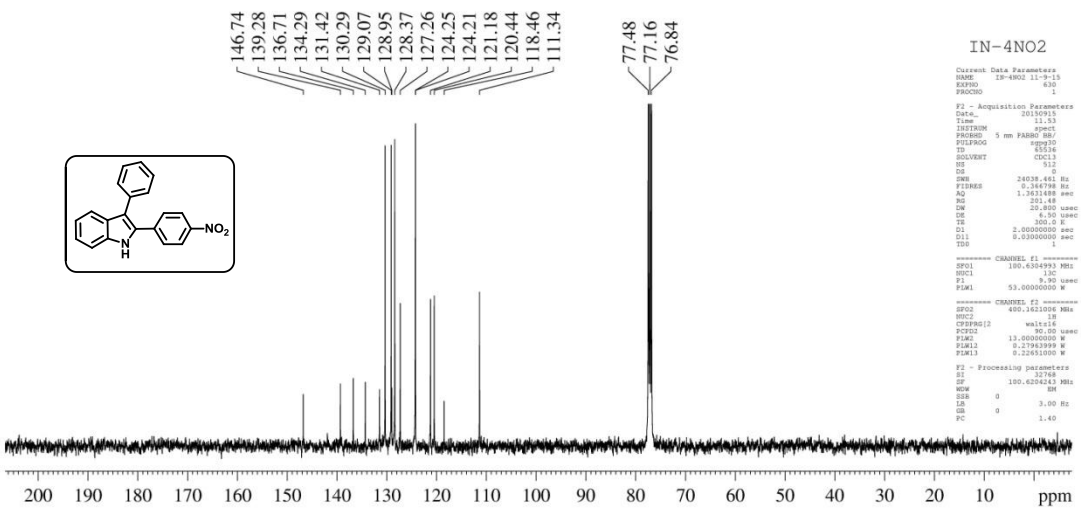

**[13]C NMR spectrum of 3ak**



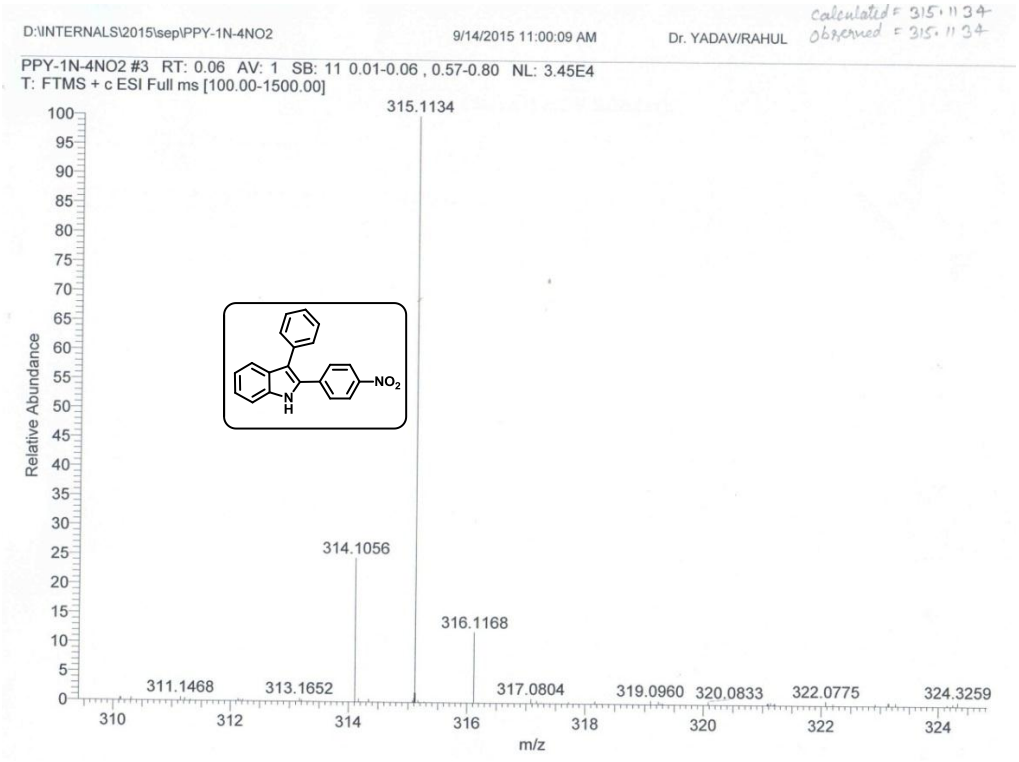

**HRMS spectrum of 3ak**



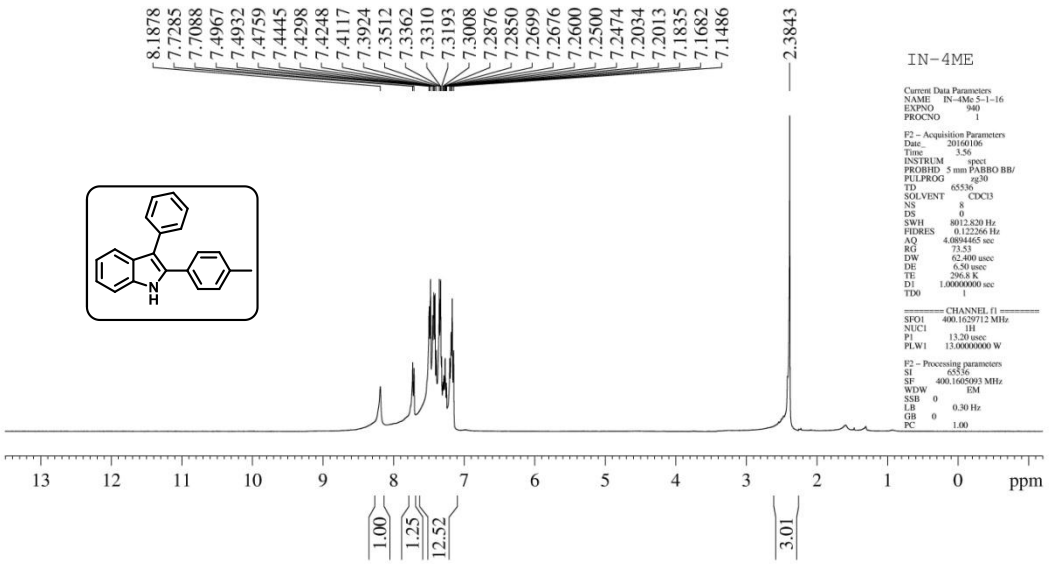

**$^{1}$H NMR spectrum of 3al**



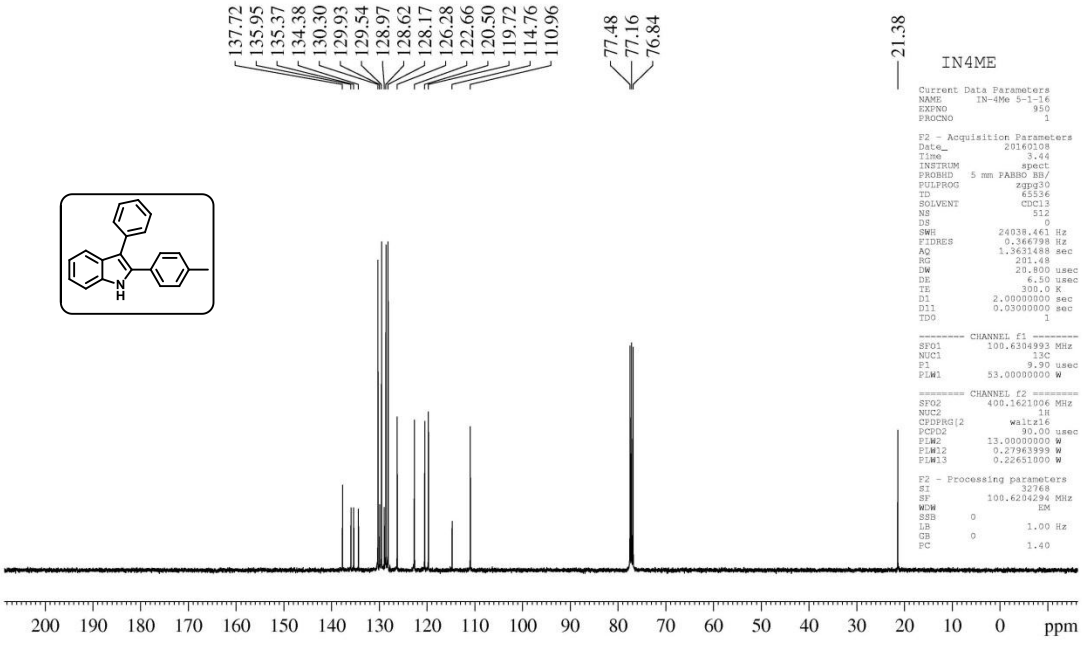

**$^{13}$C NMR spectrum of 3al**



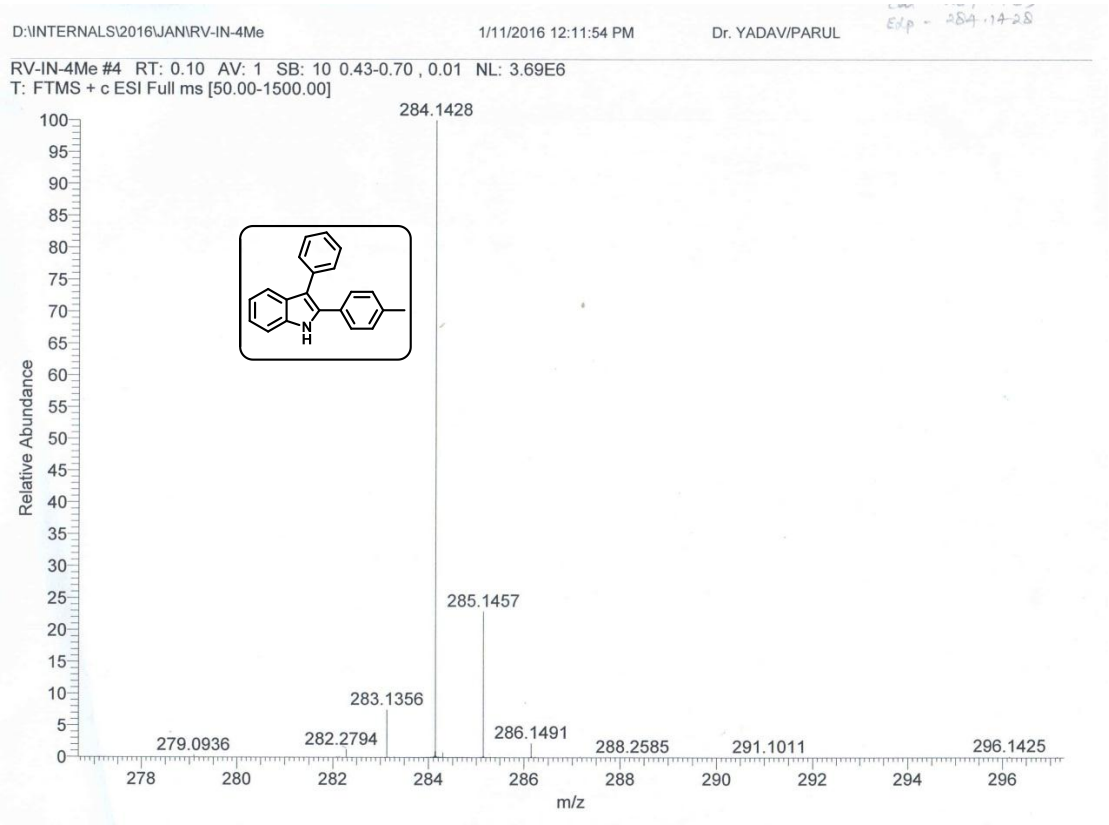

**HRMS spectrum of 3al**



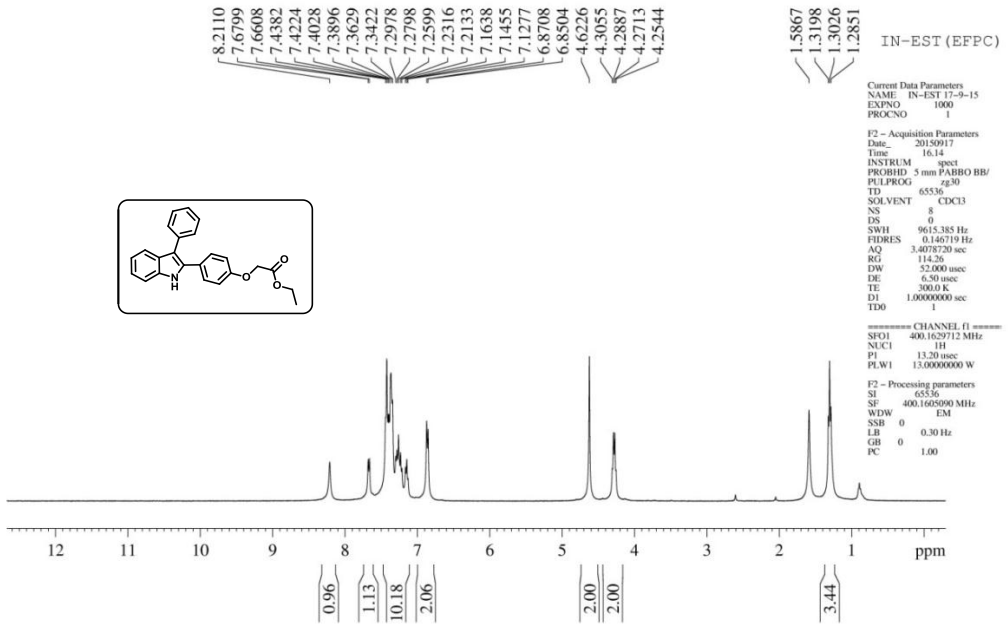

**$^{1}$H NMR spectrum of 3am**



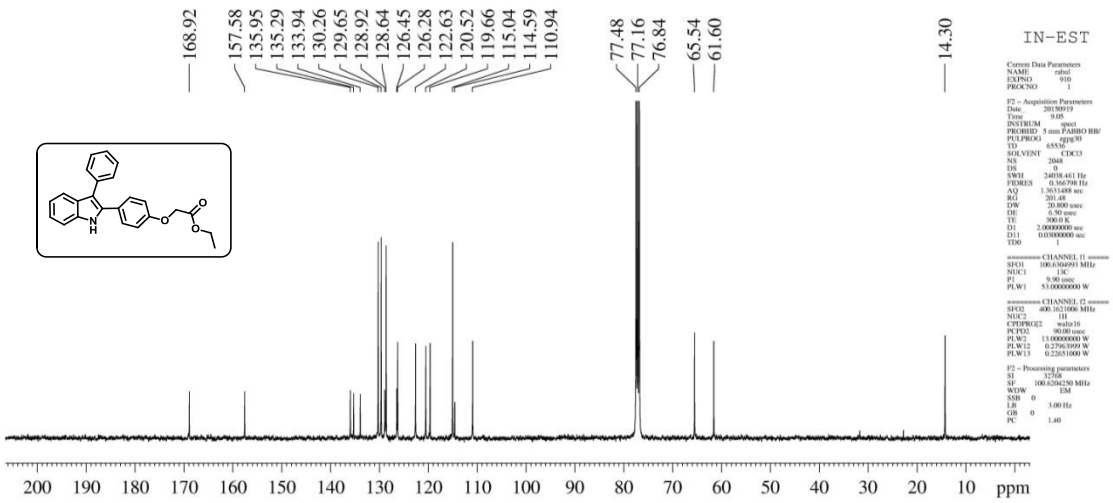

**$^{13}$C NMR spectrum of 3am**



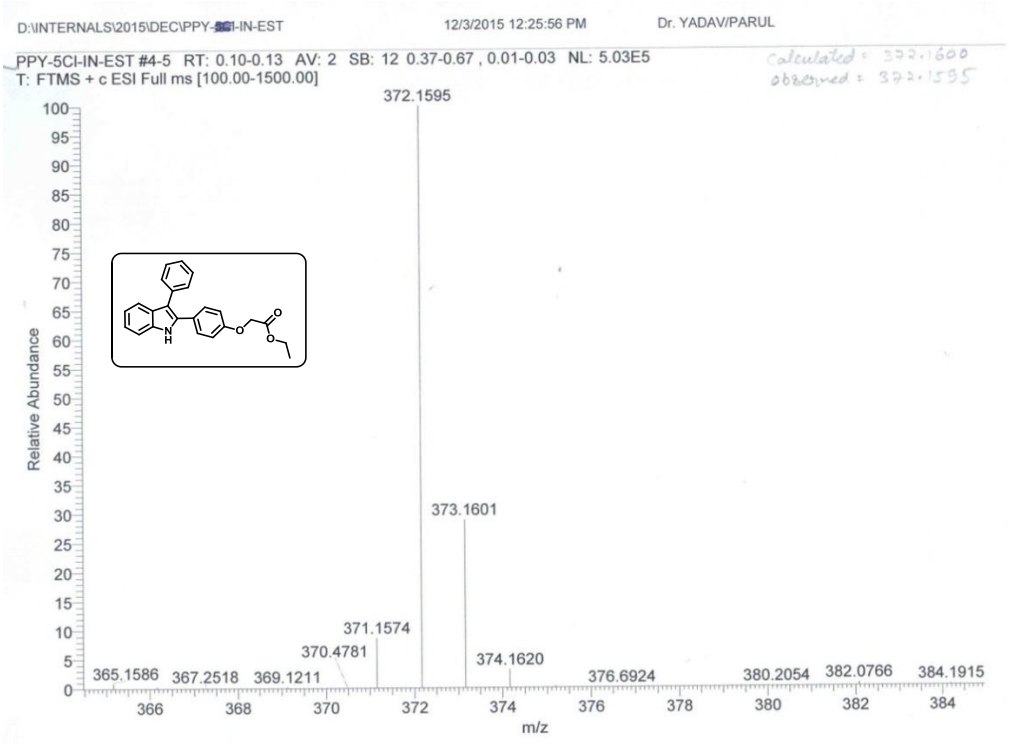

**HRMS spectrum of 3am**



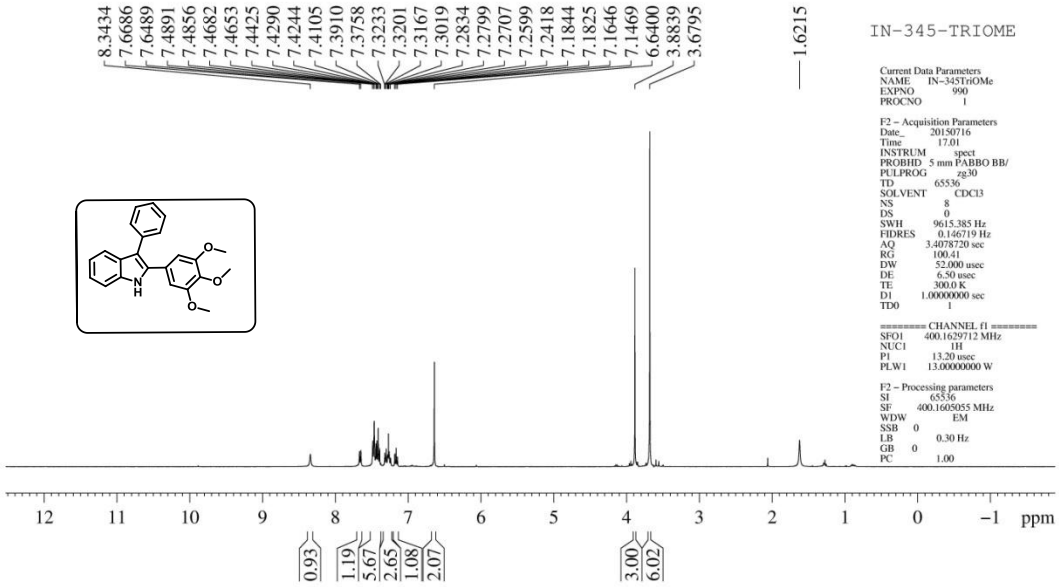

**$^{1}$H NMR spectrum of 3an**



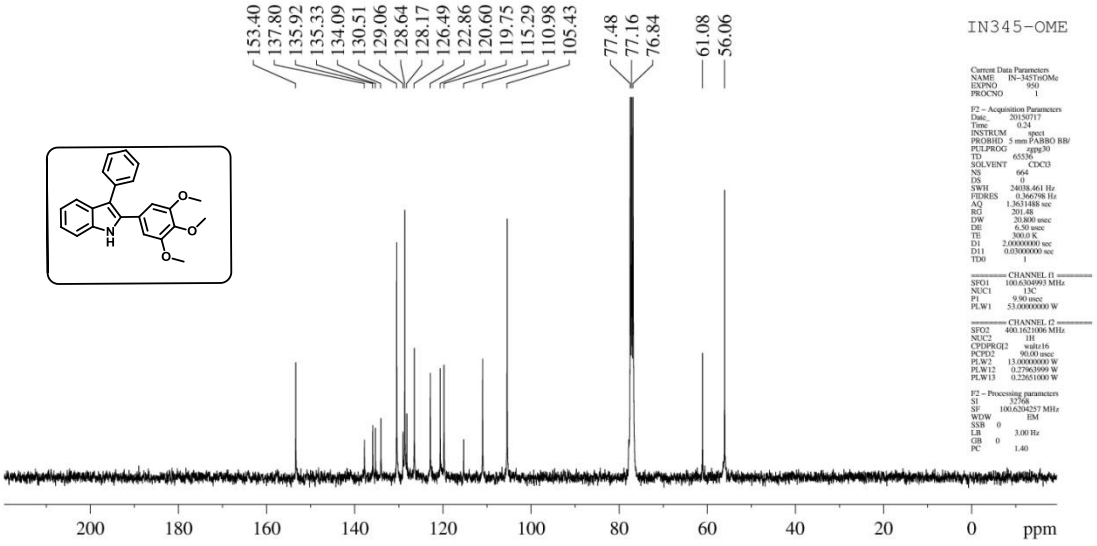

**$^{13}$C NMR spectrum of 3an**



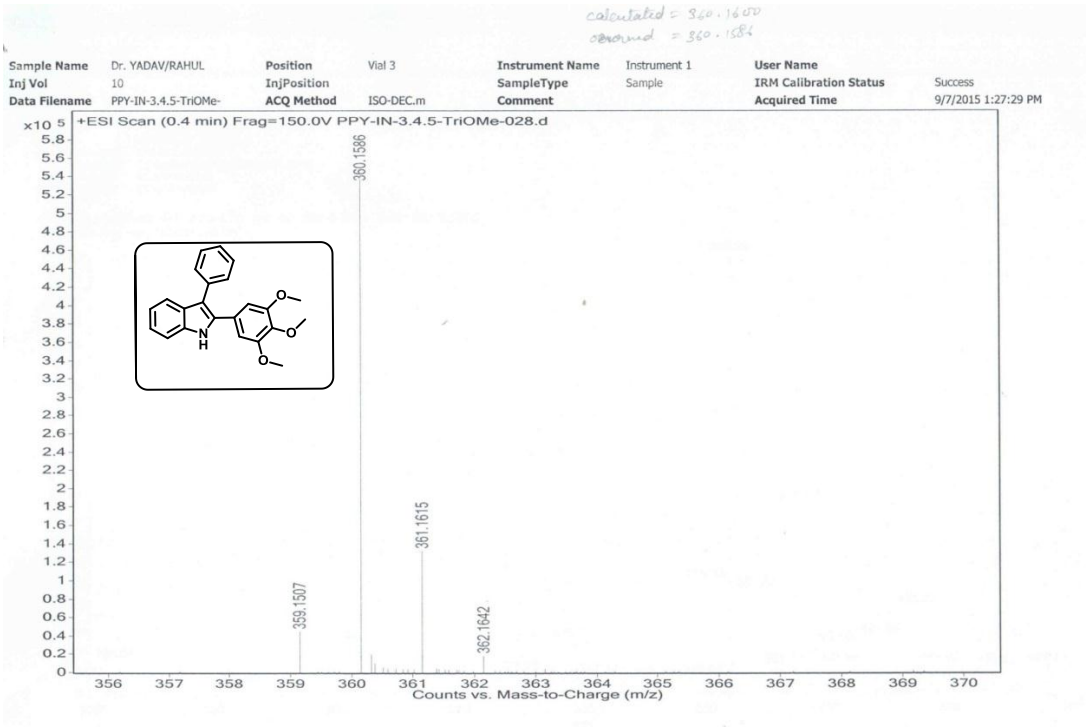

**HRMS spectrum of 3an**



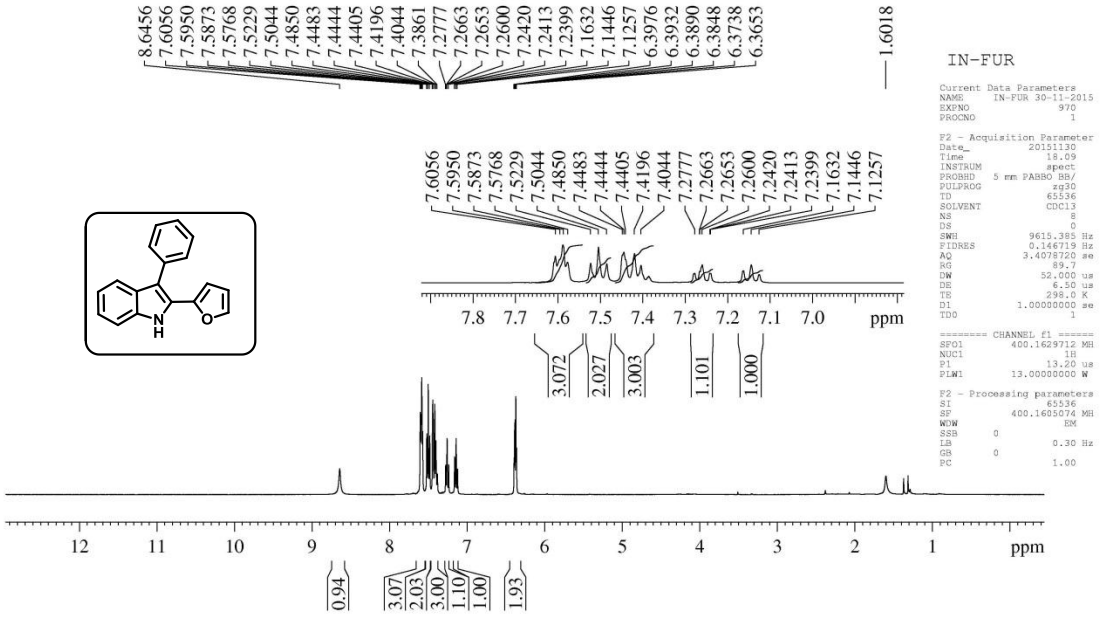

**$^{1}$H NMR spectrum of 3aq**



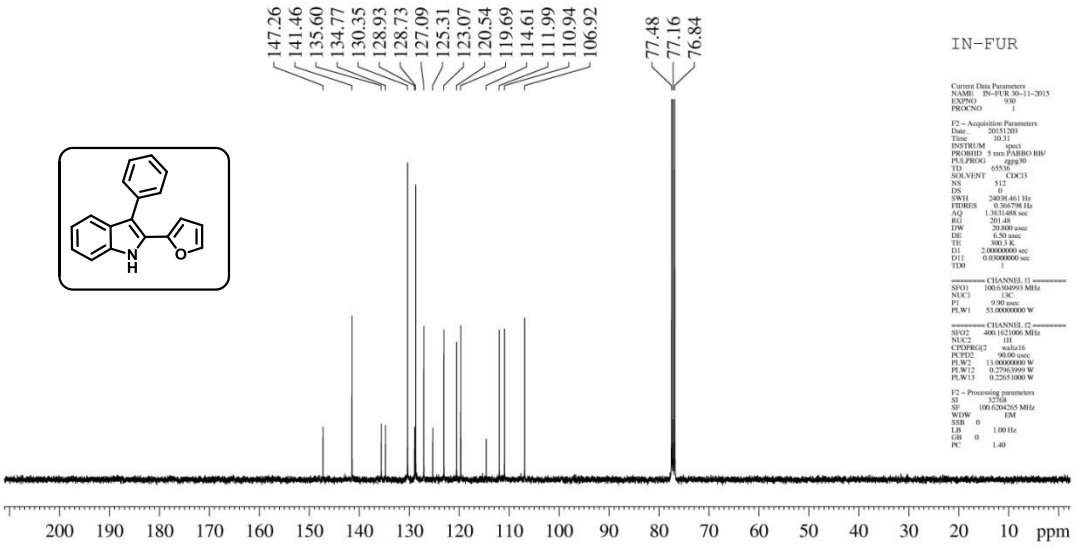

**$^{13}$C NMR spectrum of 3aq**



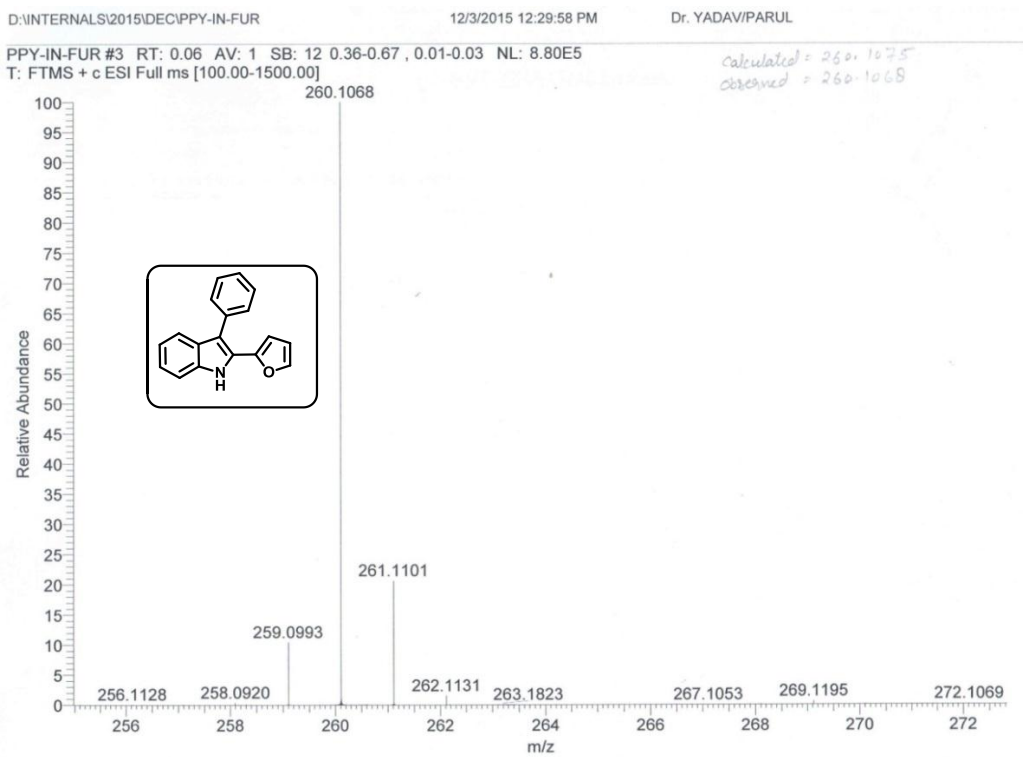

**HRMS spectrum of 3aq**



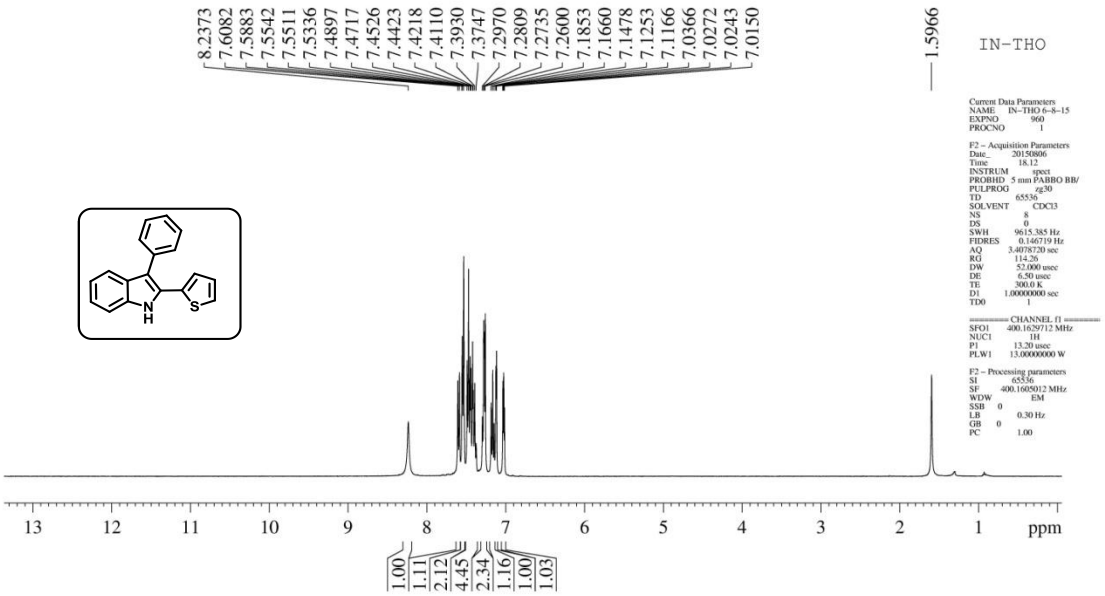

**$^{1}$H NMR spectrum of 3ar**



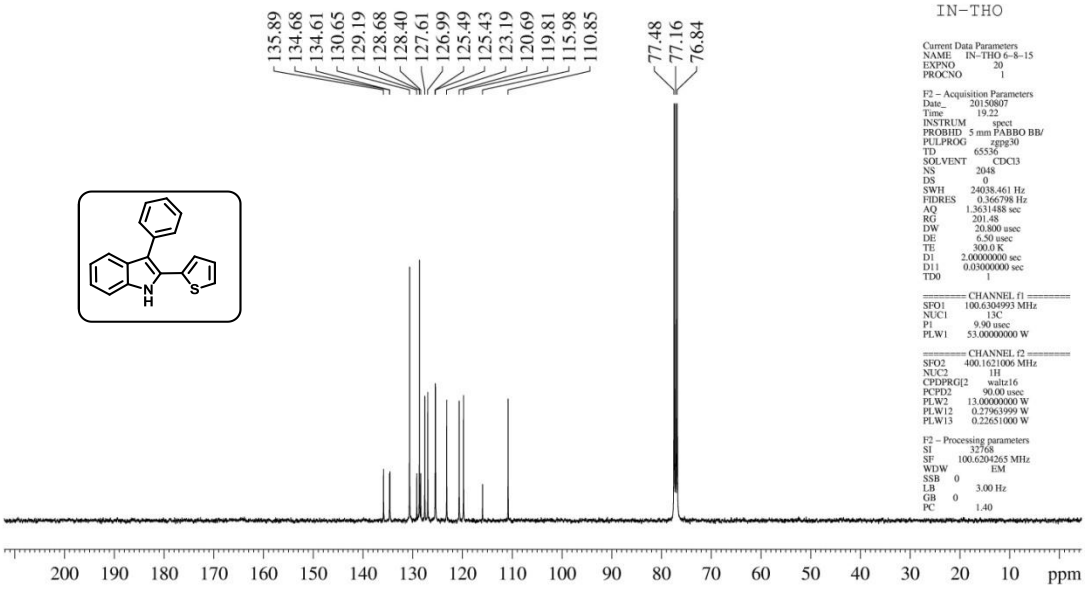

**$^{13}$C NMR spectrum of 3ar**



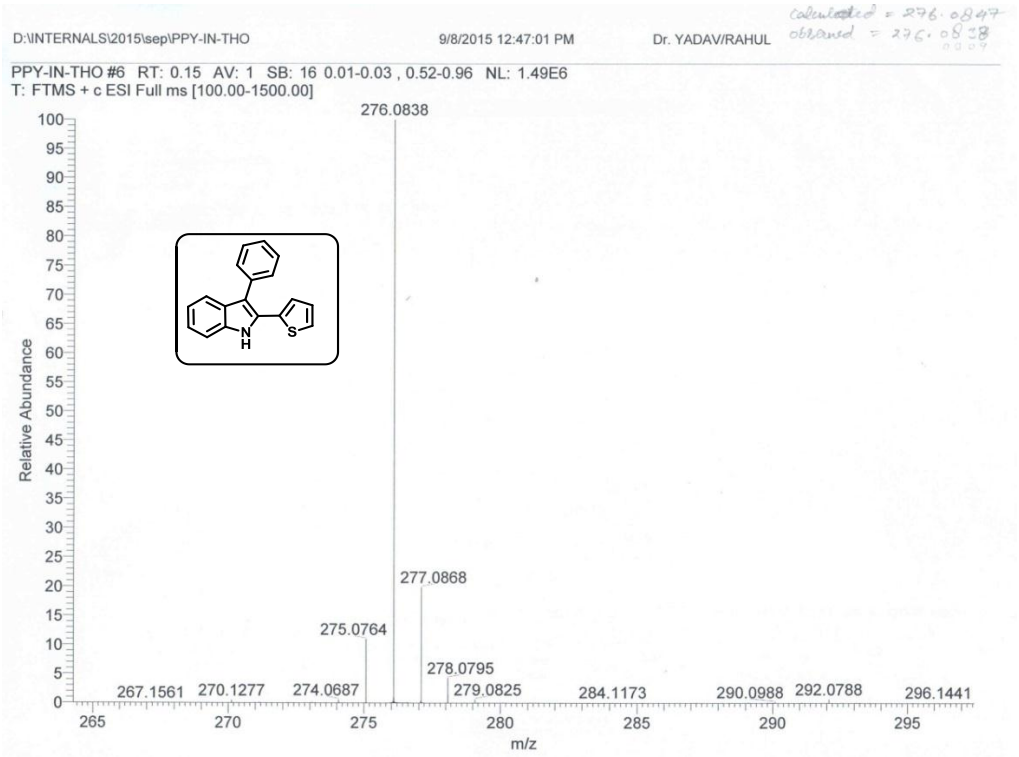

**HRMS spectrum of 3ar**



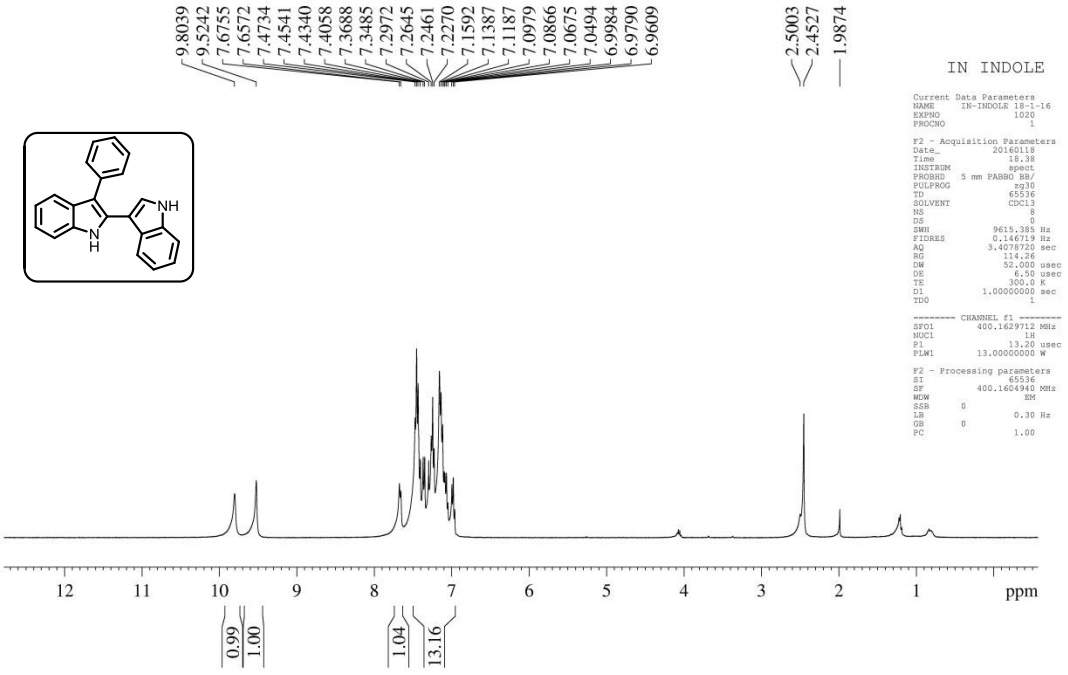

**$^{1}$H NMR spectrum of 3as**



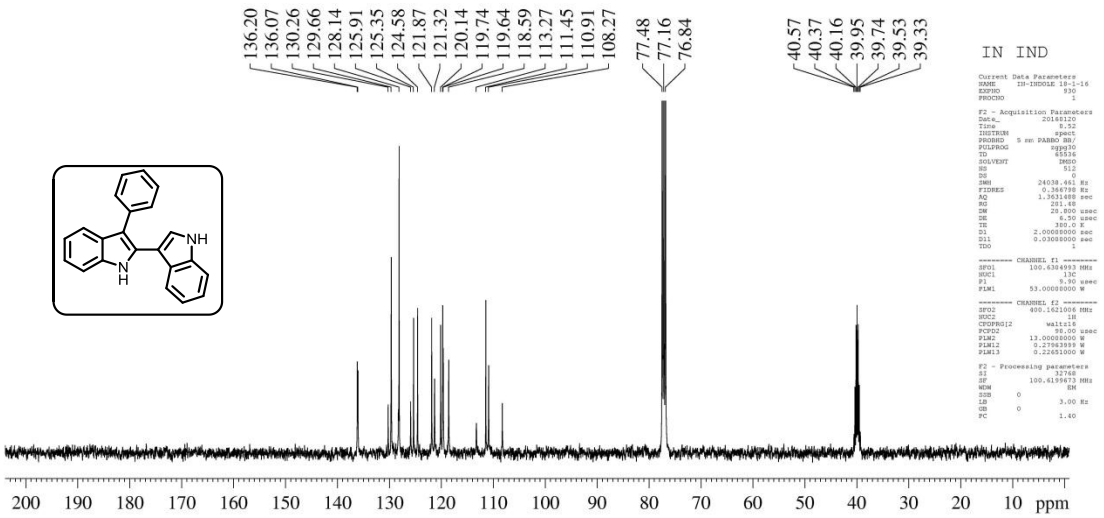

**$^{13}$C NMR spectrum of 3as**



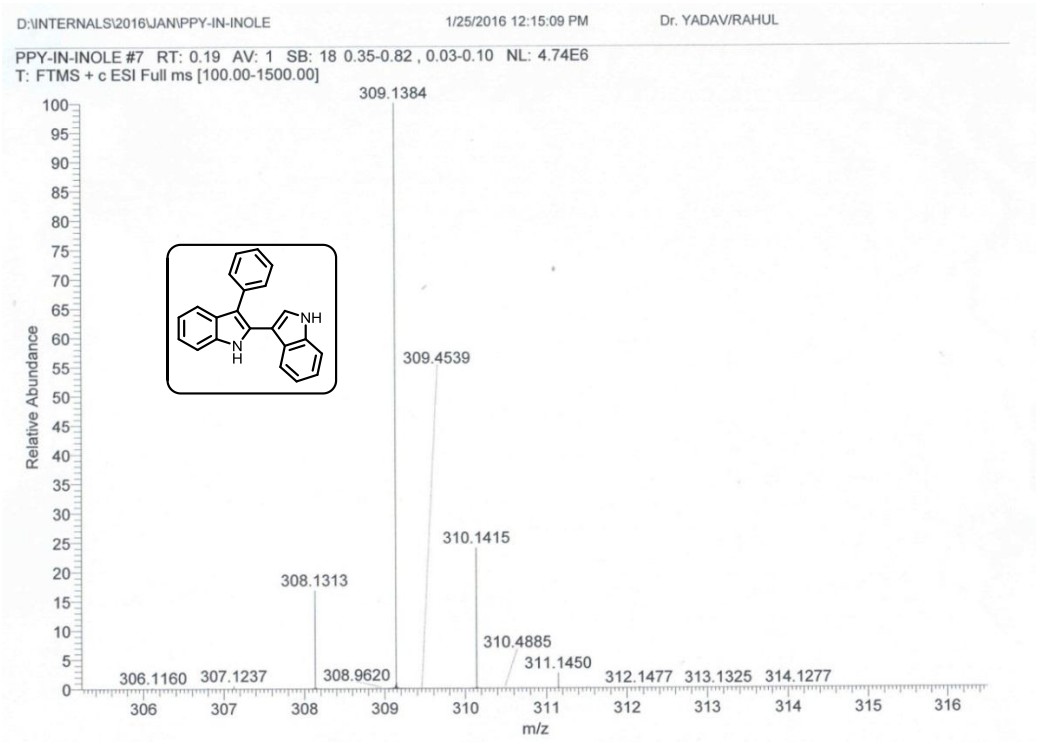

**HRMS spectrum of 3as**



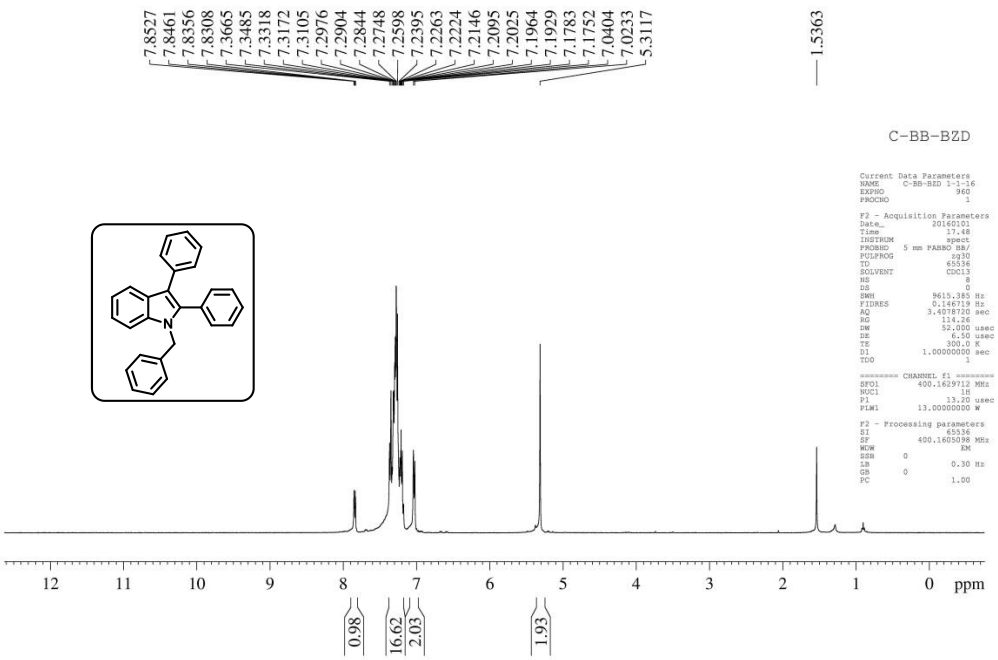

**$^{1}$H NMR spectrum of 3at**



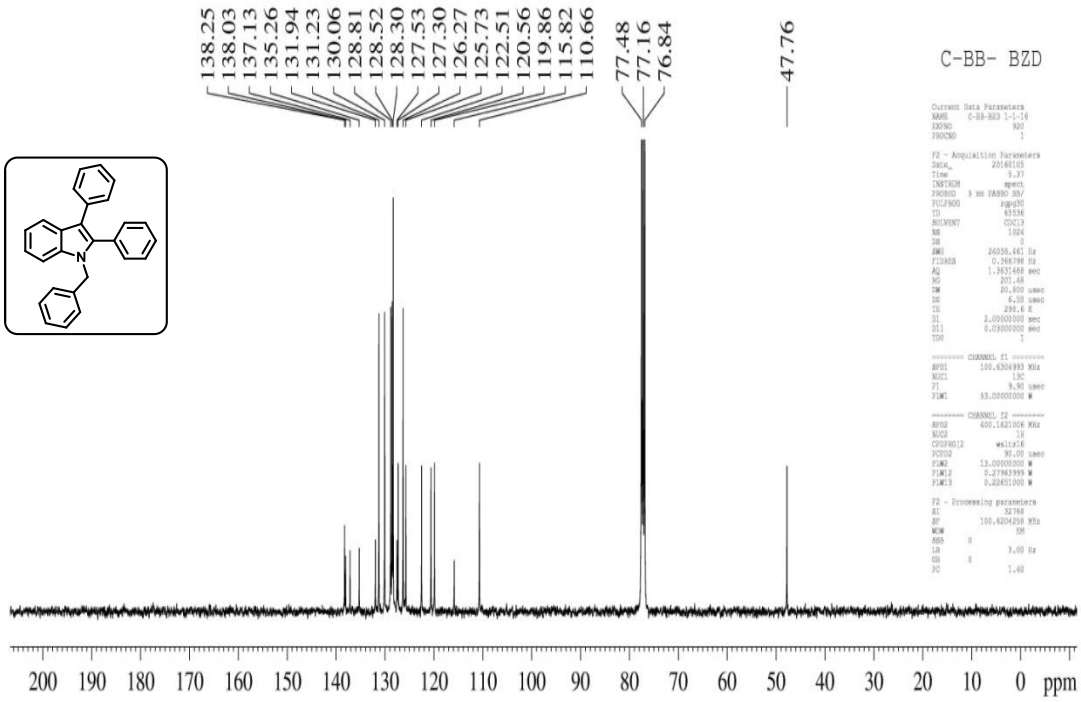

**$^{13}$C NMR spectrum of 3at**



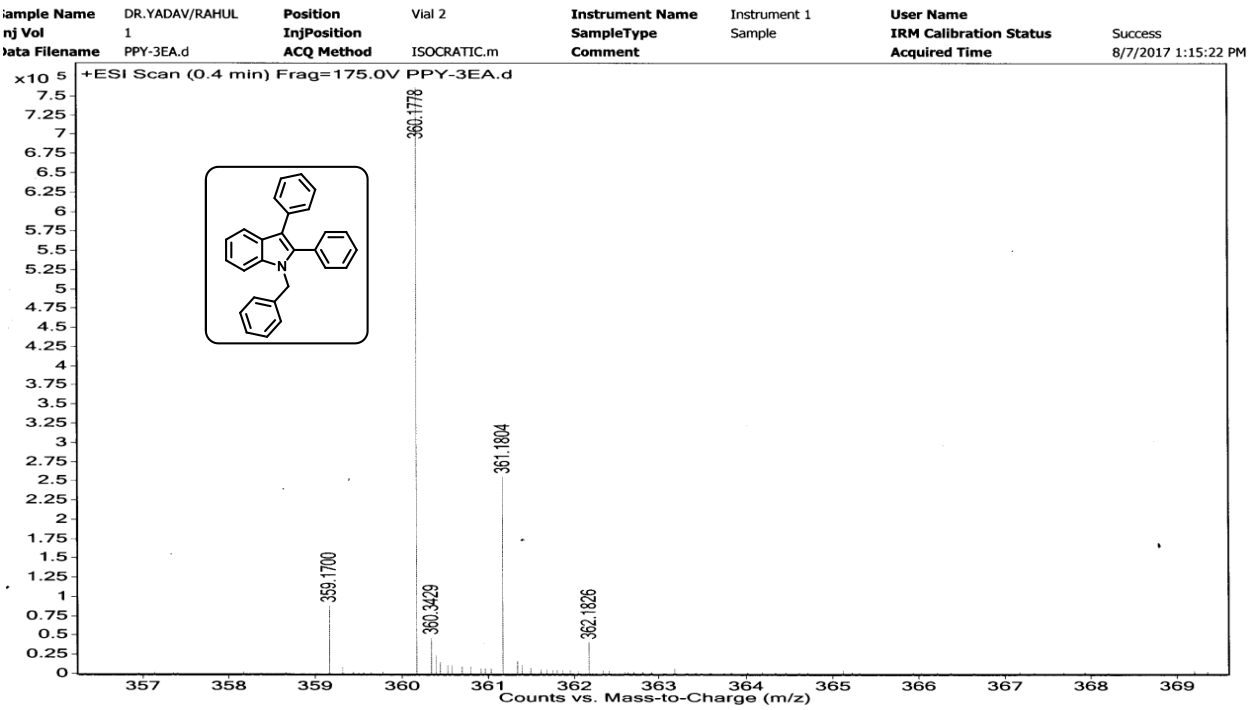

**HRMS spectrum of 3at**



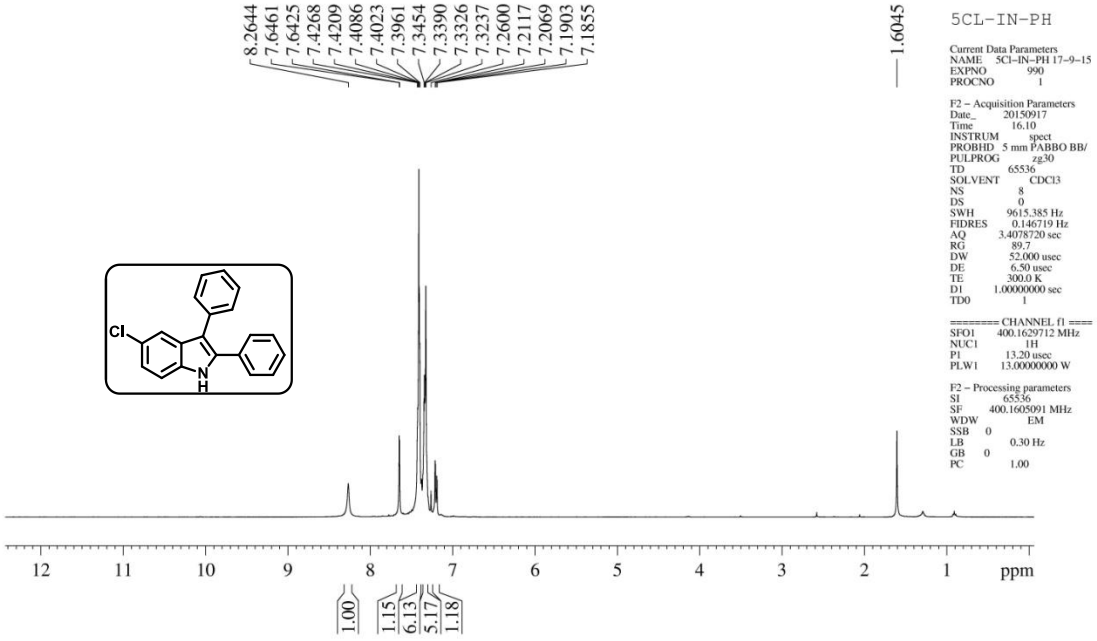

**$^{1}$H NMR spectrum of 3ea**



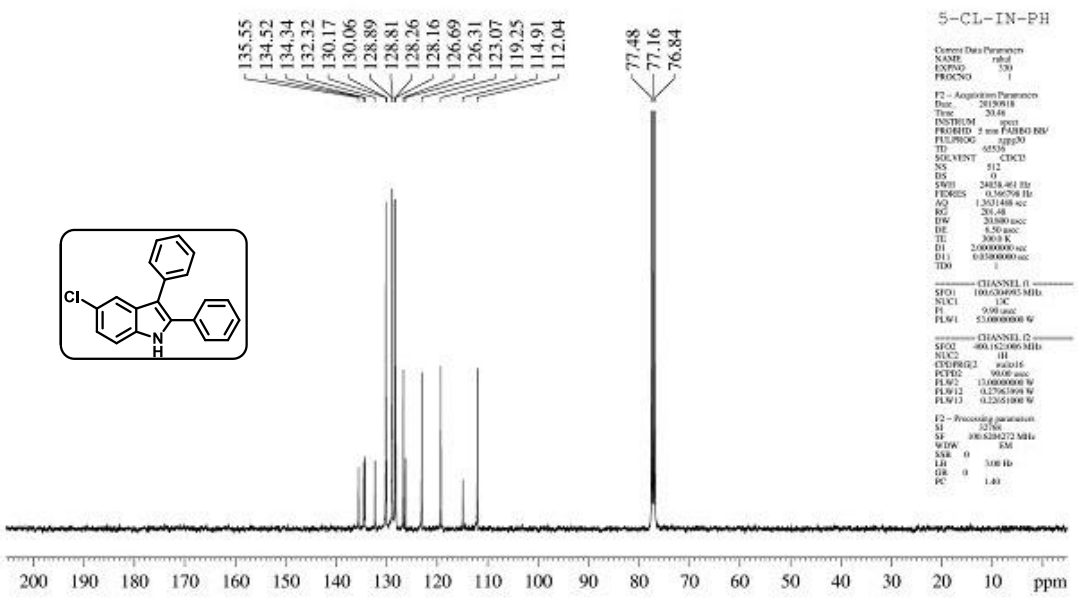

**$^{13}$C NMR spectrum of 3ea**



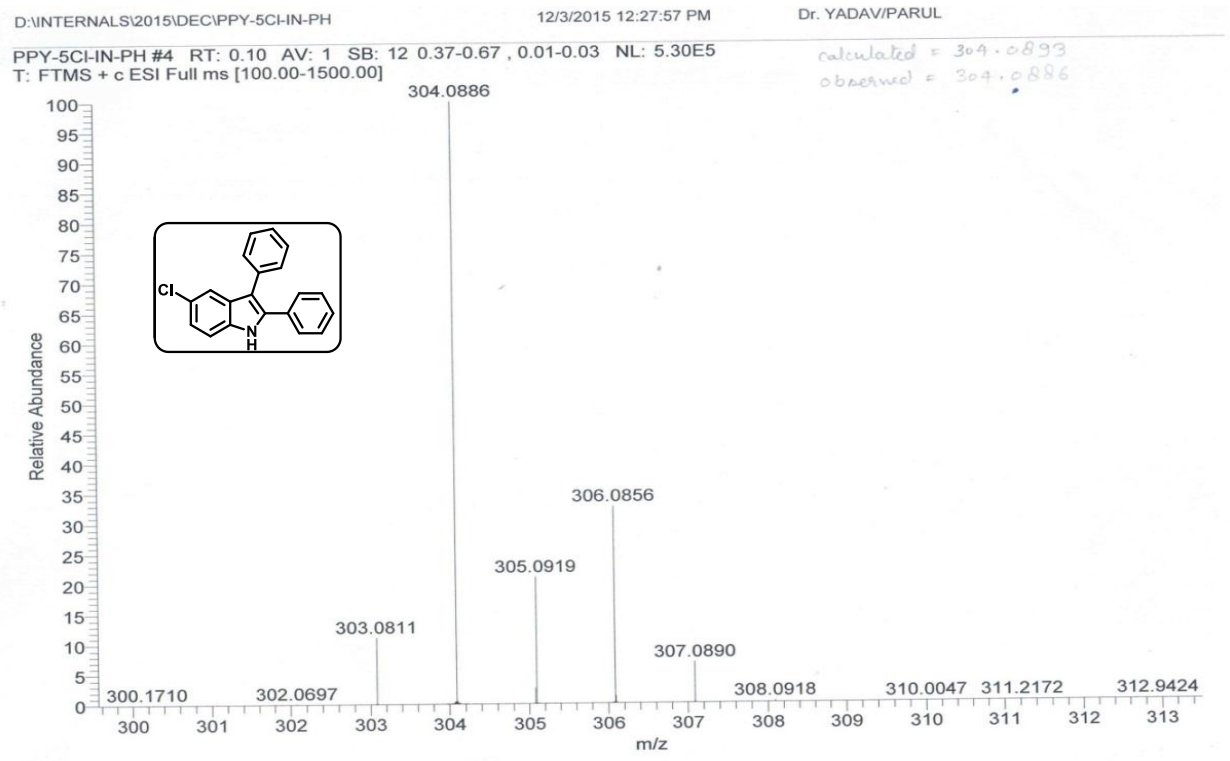

**HRMS spectrum of 3ea**



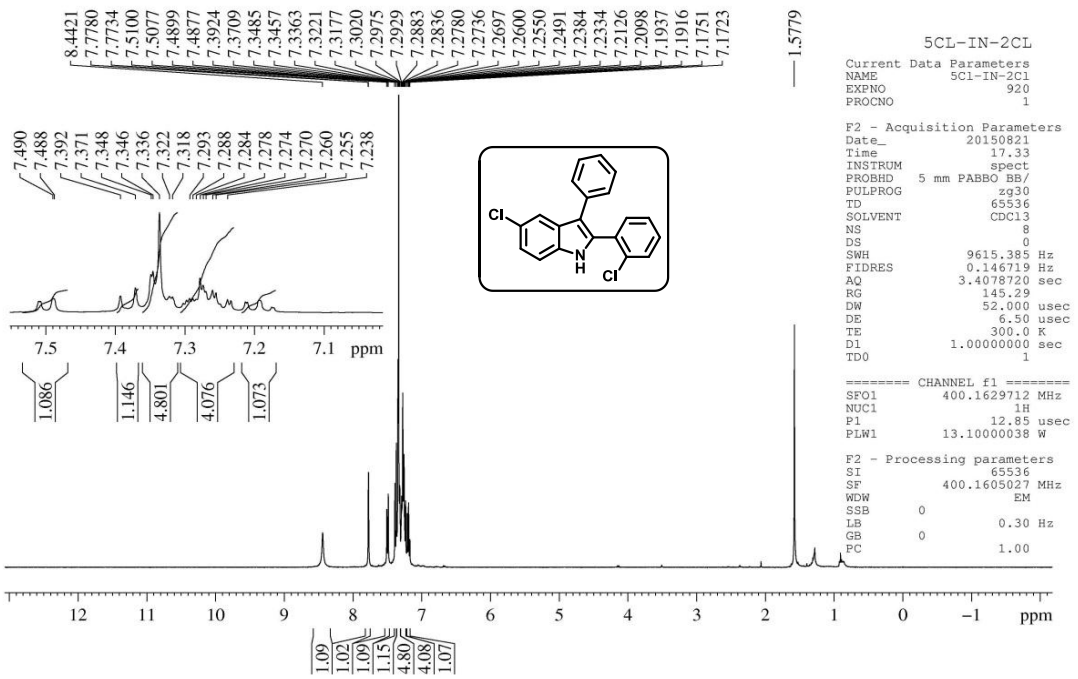

**$^{1}$H NMR spectrum of 3ba**



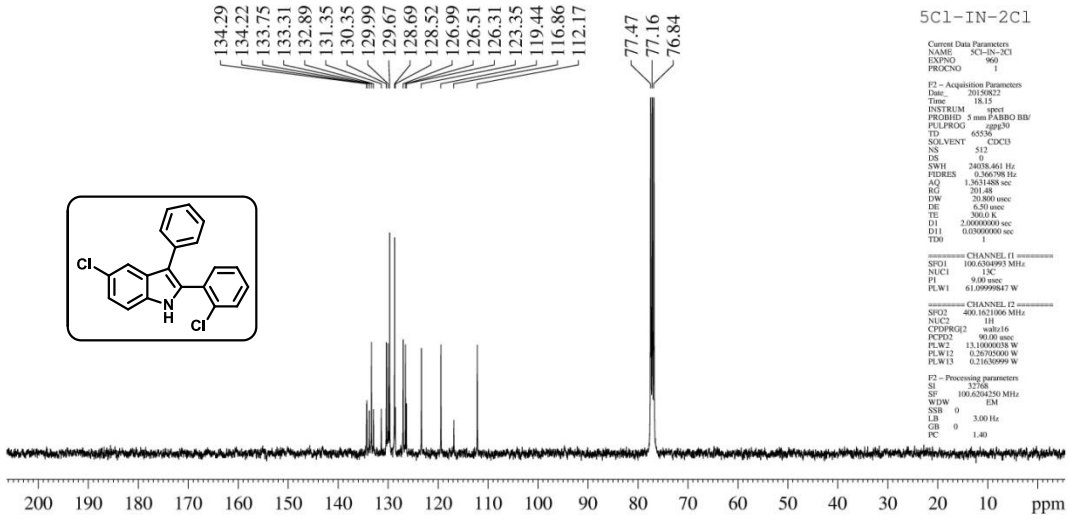

**$^{13}$C NMR spectrum of 3ba**



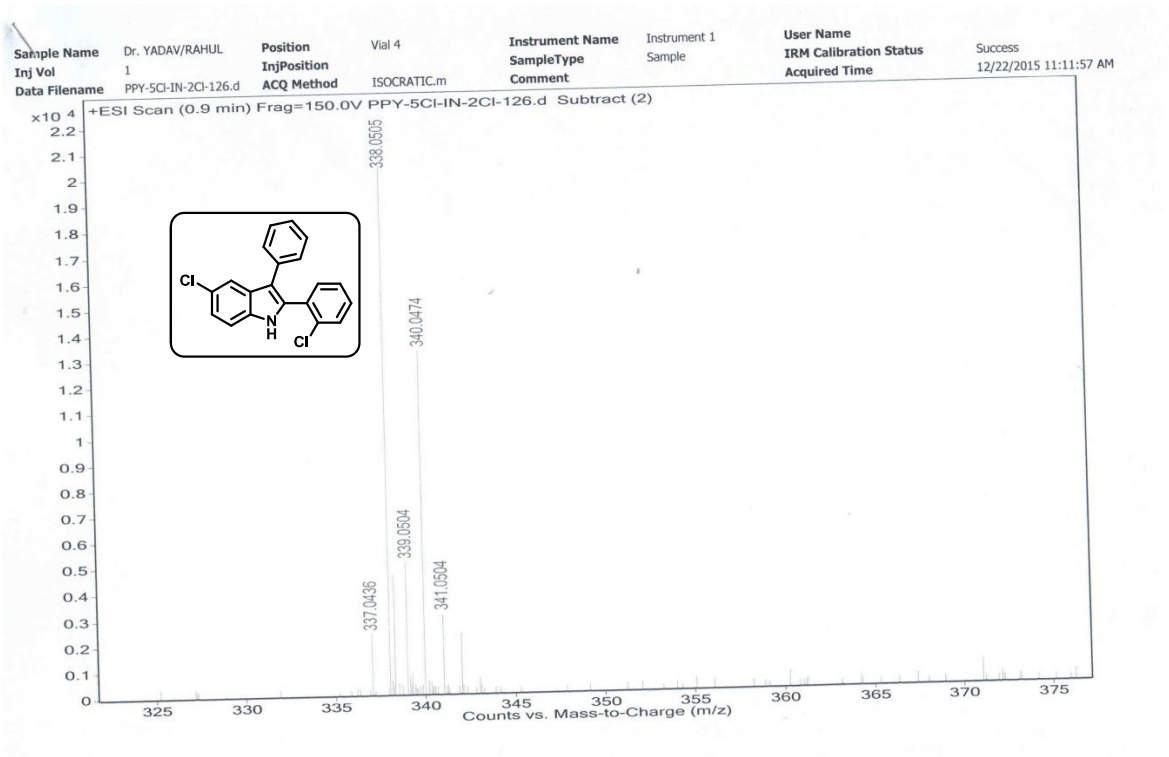

**HRMS spectrum of 3ba**



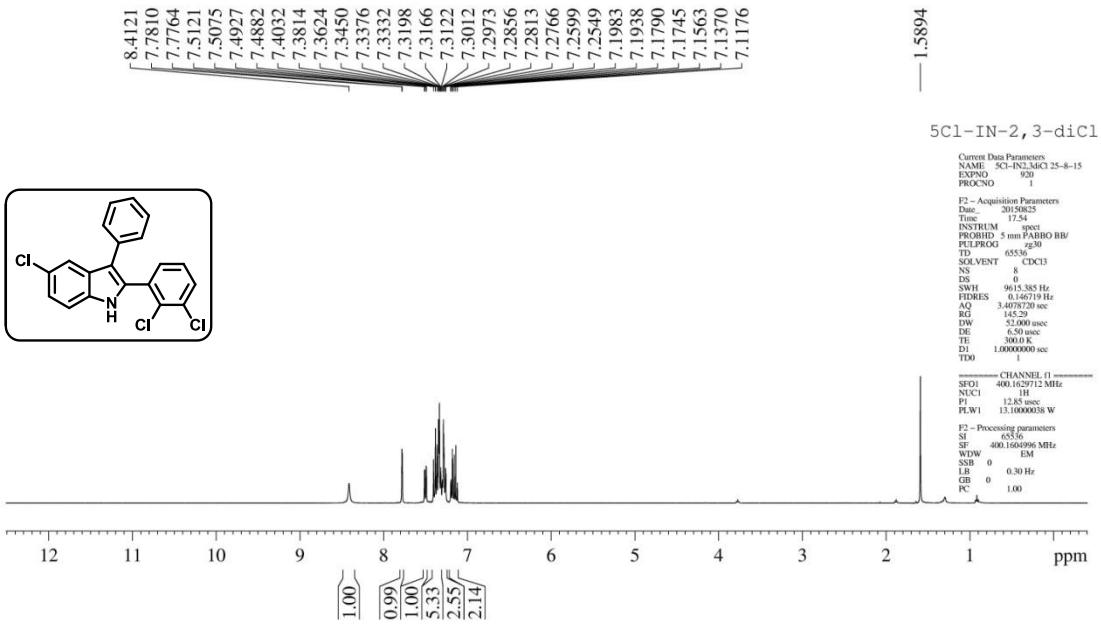
**$^{1}$H NMR spectrum of 3bd**



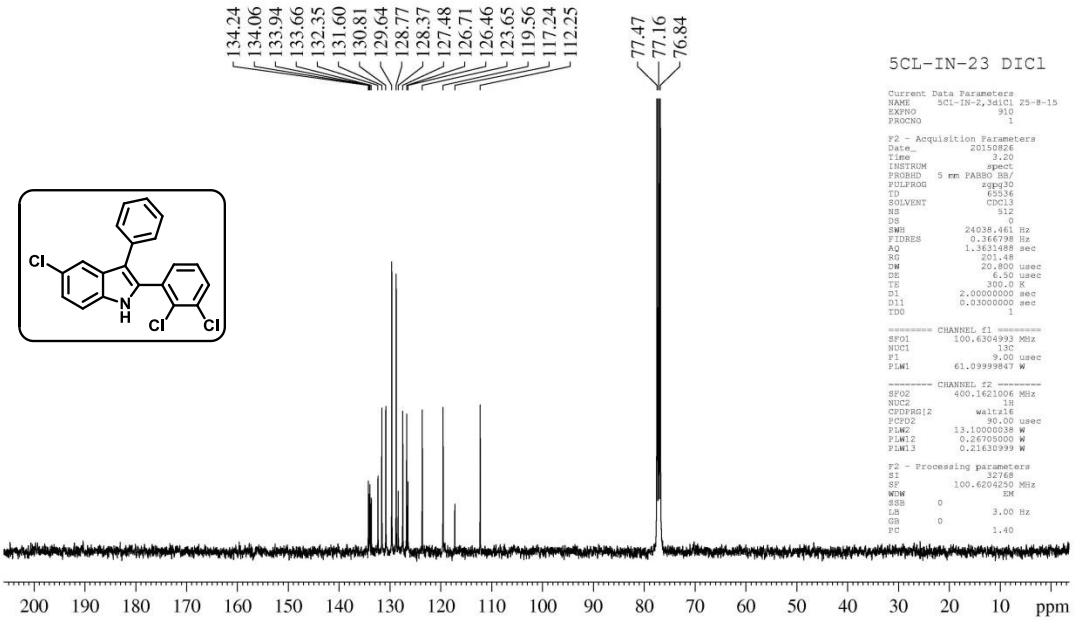
**$^{13}$C NMR spectrum of 3bd**



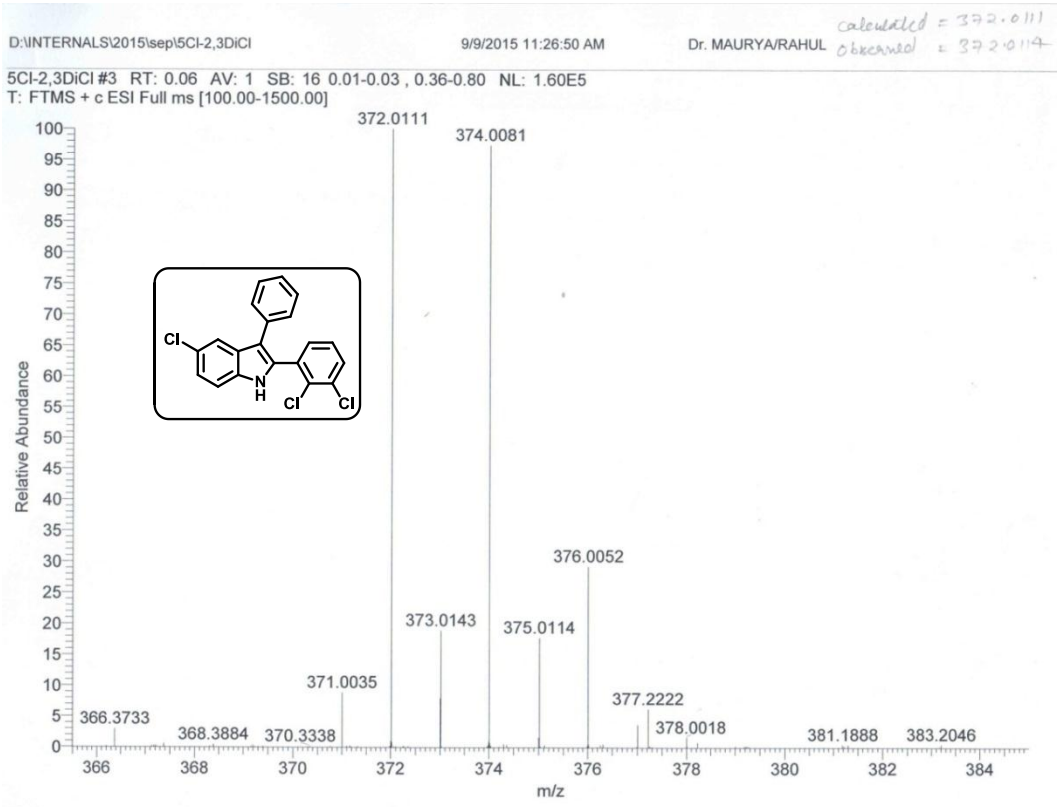

**HRMS spectrum of 3bd**



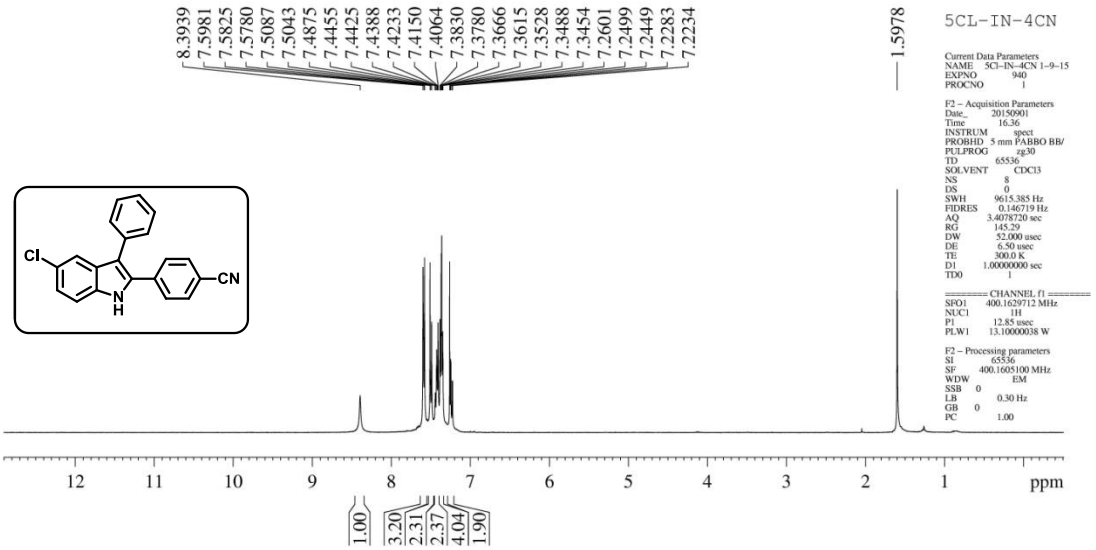

**$^{1}$H NMR spectrum of 3bh**



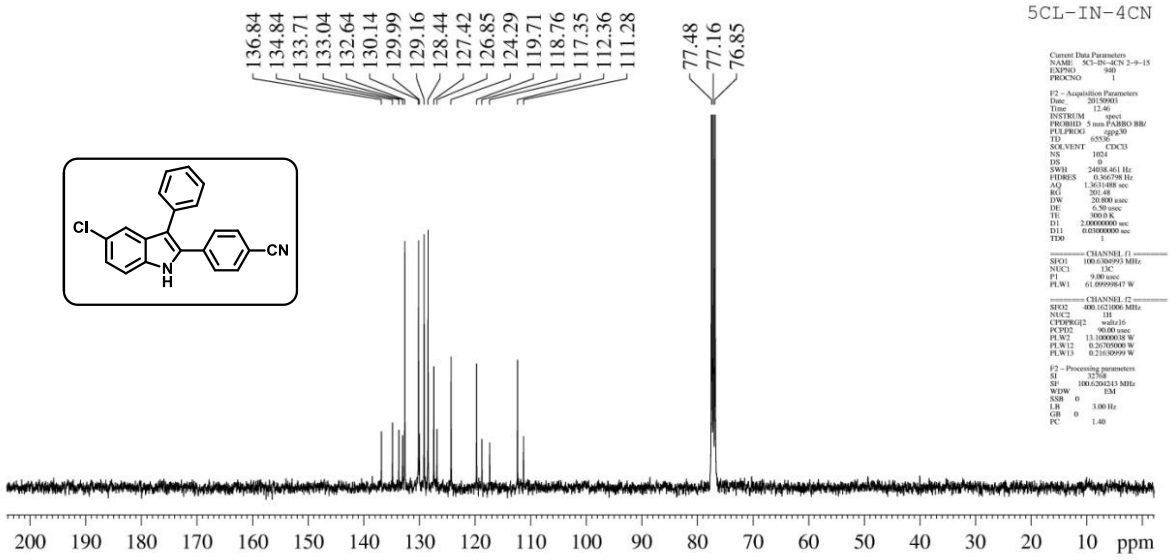

**$^{13}$C NMR spectrum of 3bh**



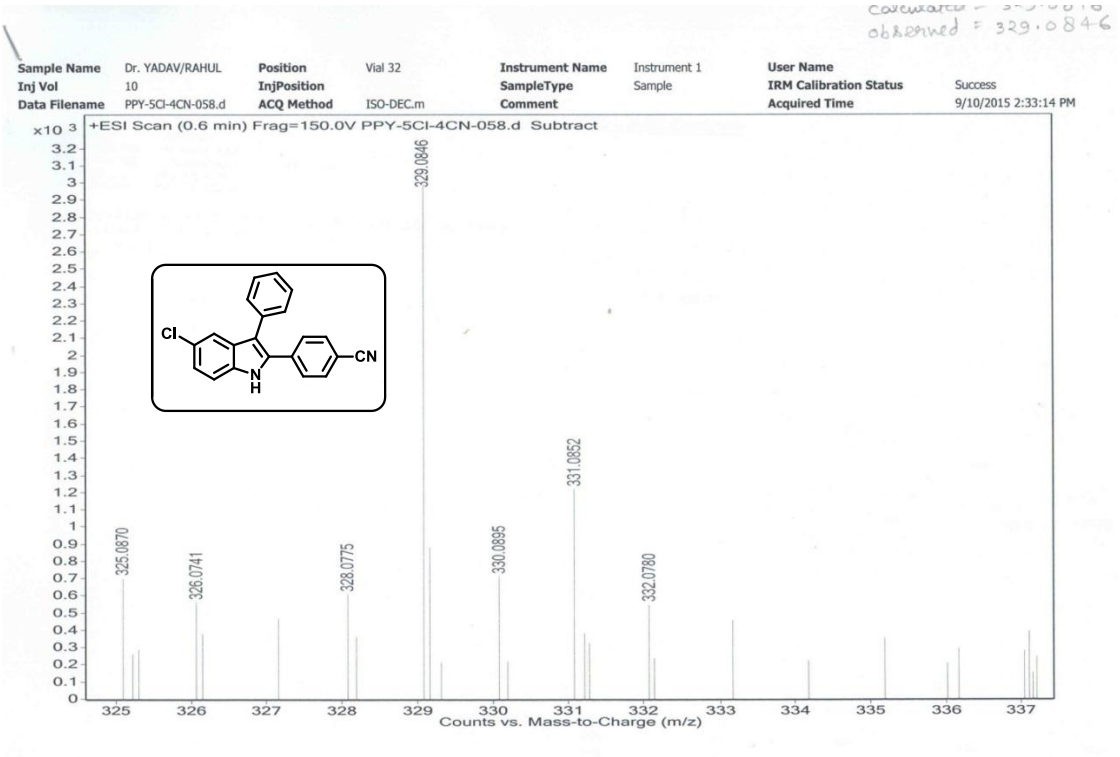

**HRMS spectrum of 3bh**



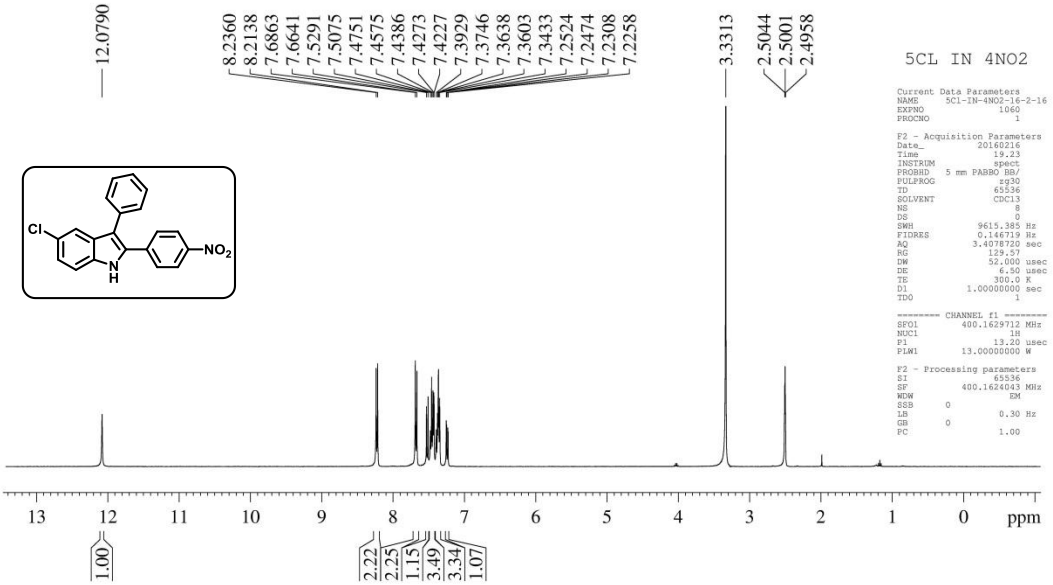

**[1]H NMR spectrum of 3bk**



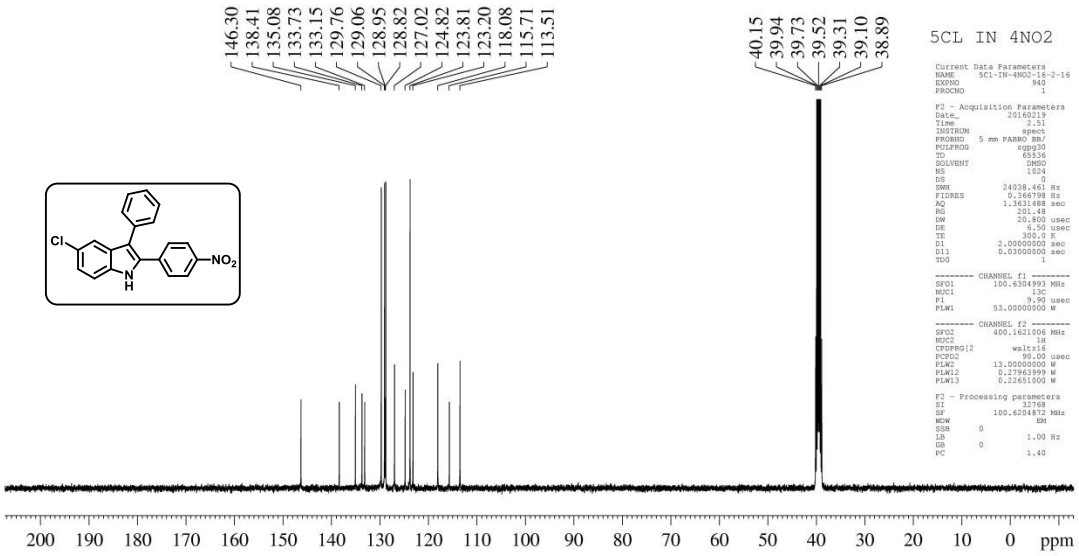

**[13]C NMR spectrum of 3bk**



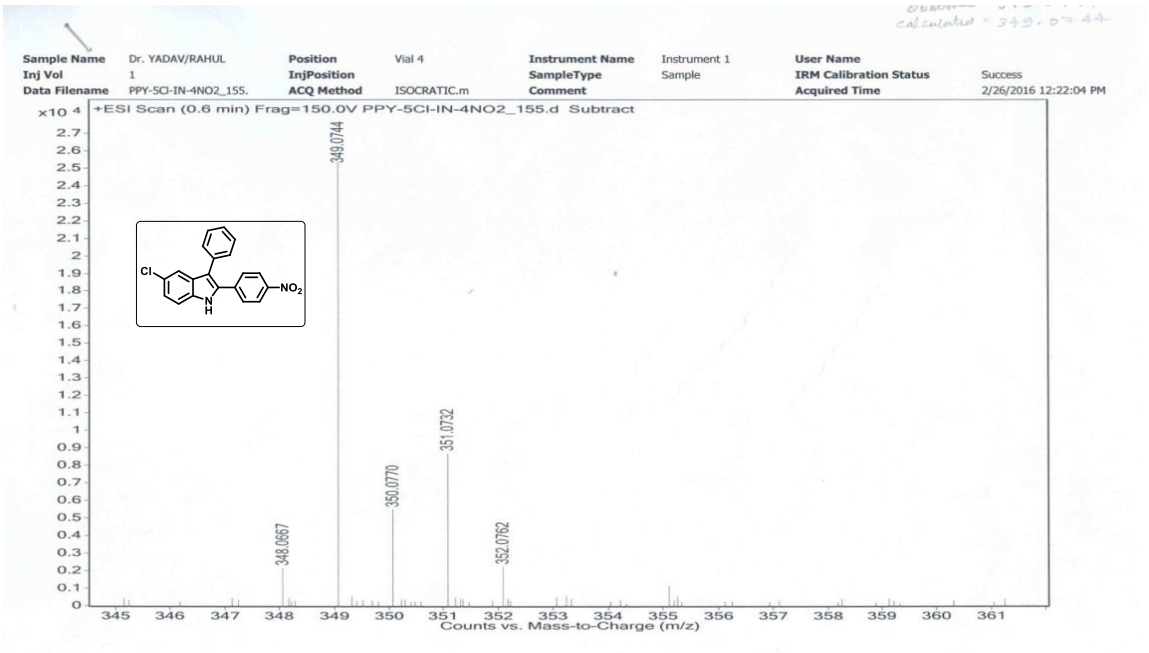

**HRMS spectrum of 3bk**



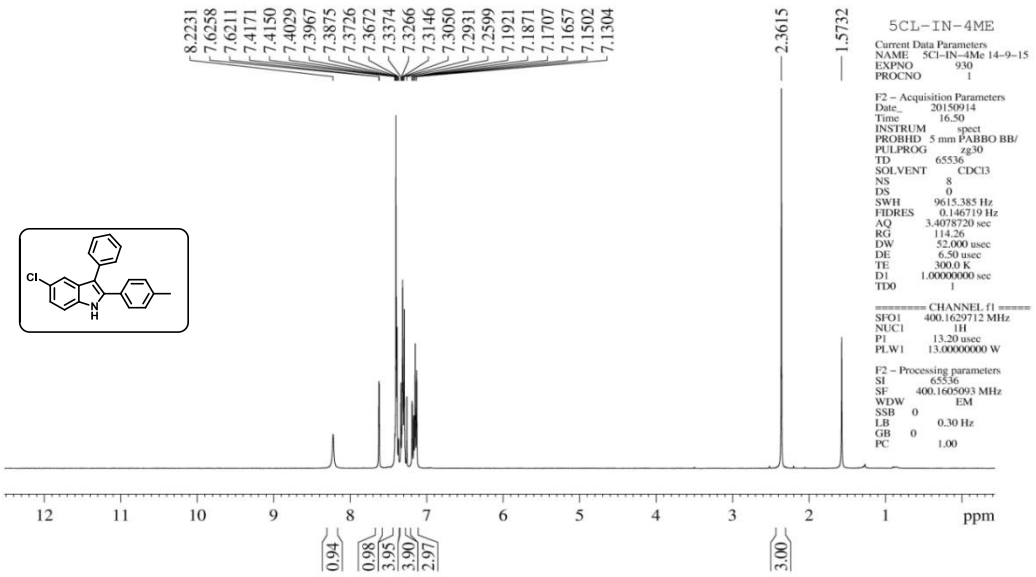

**$^{1}$H NMR spectrum of 3bl**



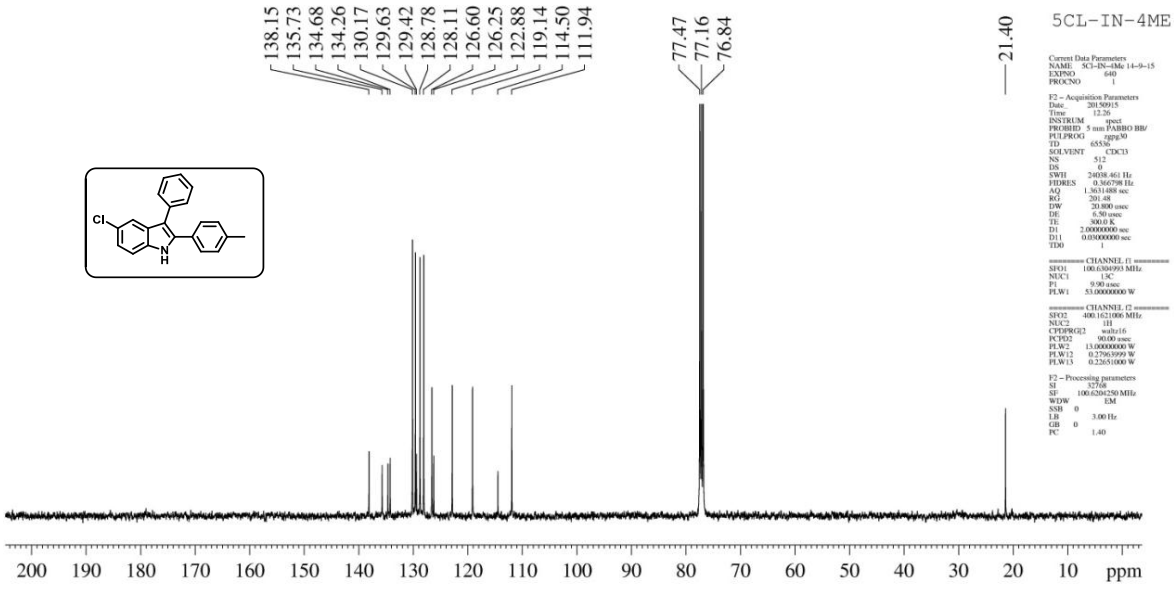

**$^{13}$C NMR spectrum of 3bl**



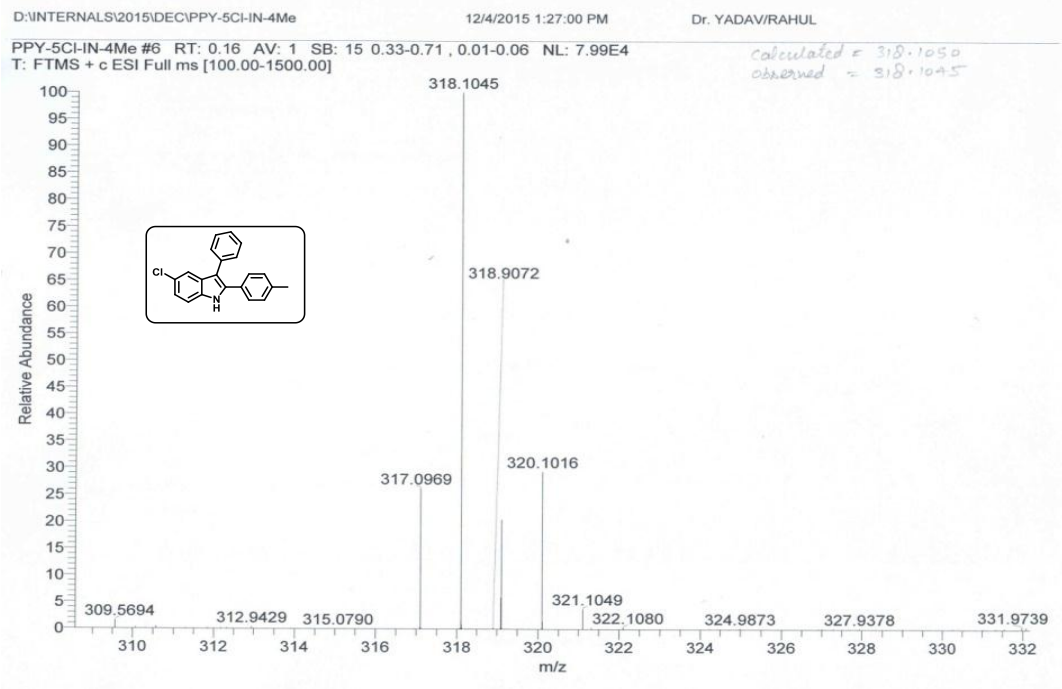

**HRMS spectrum of 3bl**



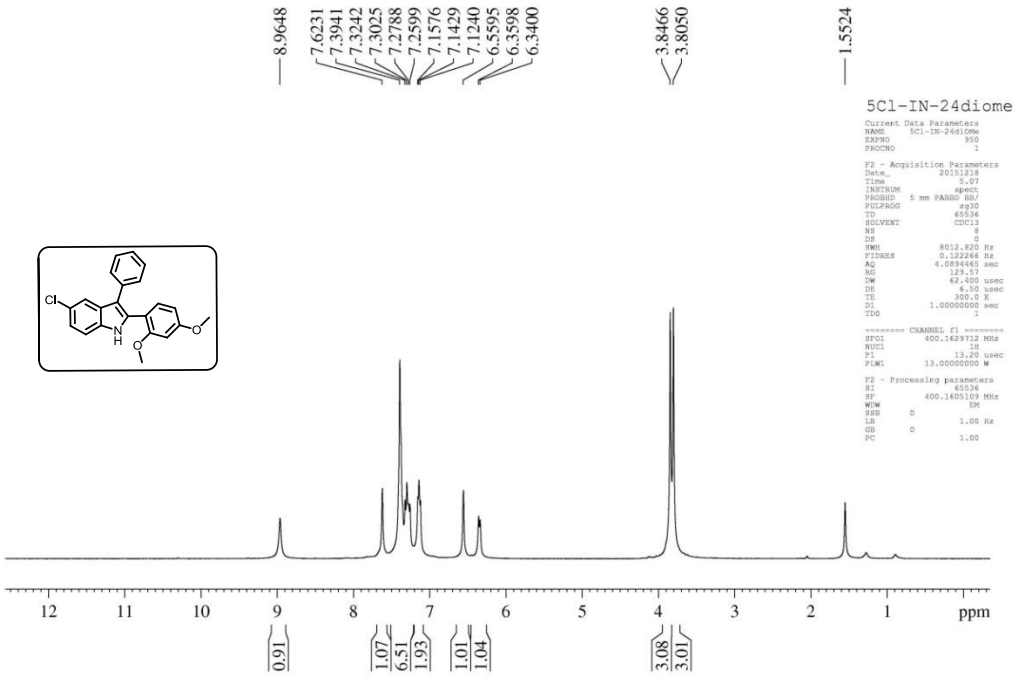

**$^{1}$H NMR spectrum of 3bm**



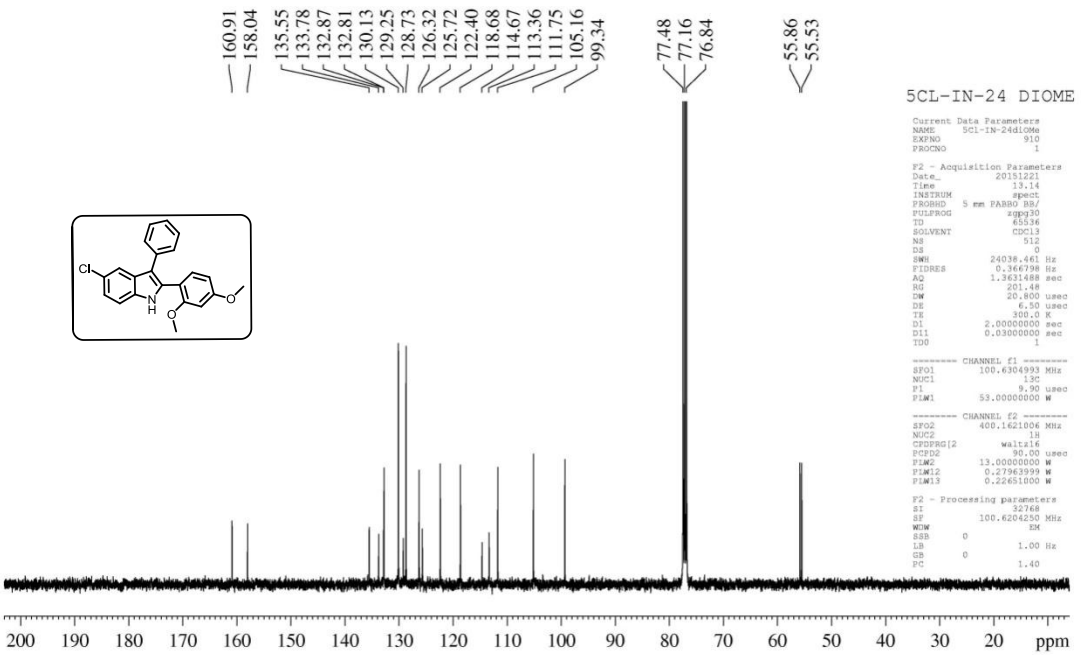

**$^{13}$C NMR spectrum of 3bm**



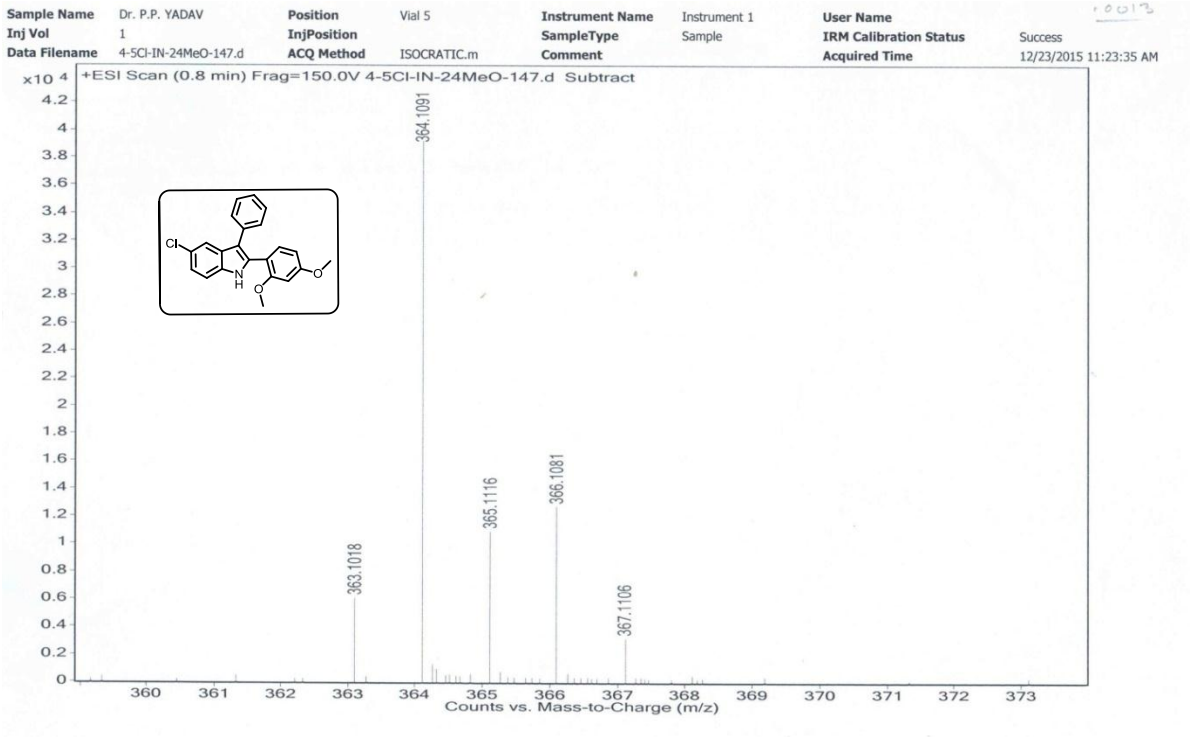

**HRMS spectrum of 3bm**



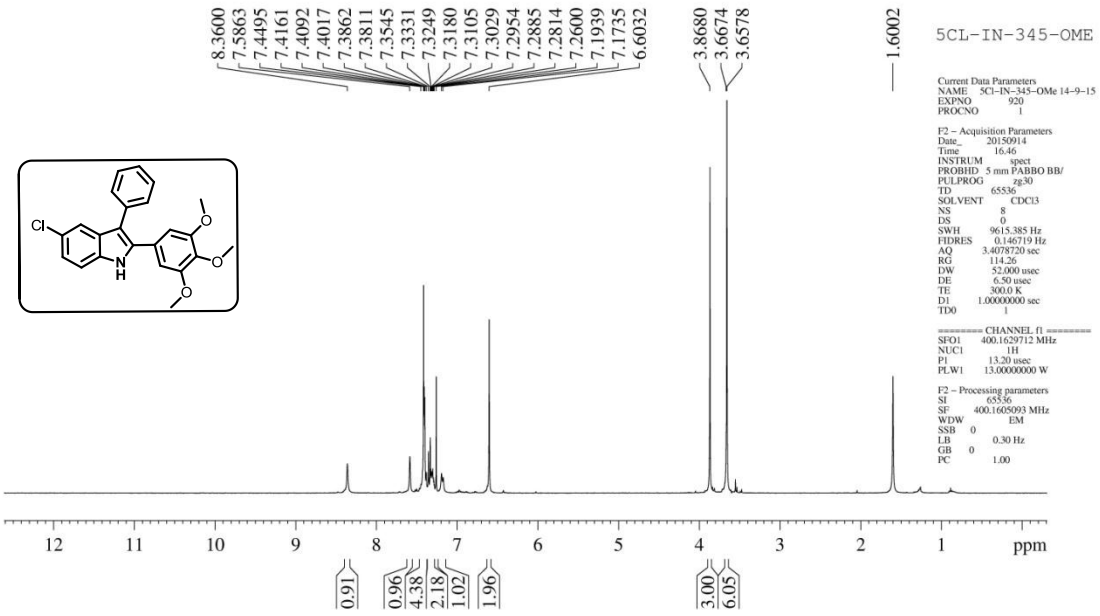

**$^{1}$H NMR spectrum of 3bp**



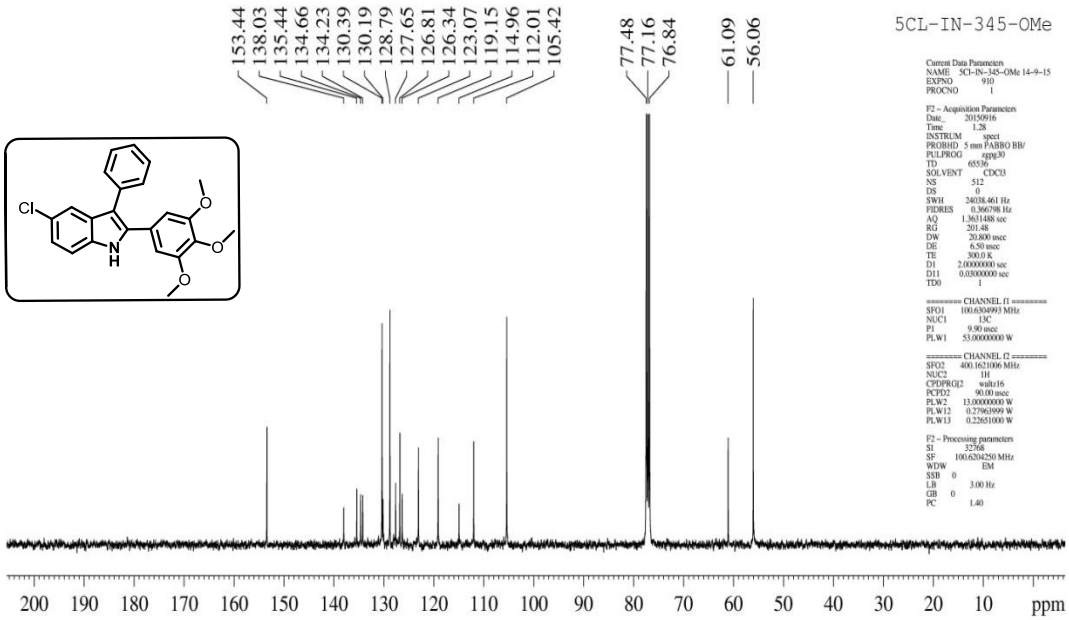

**$^{13}$C NMR spectrum of 3bp**



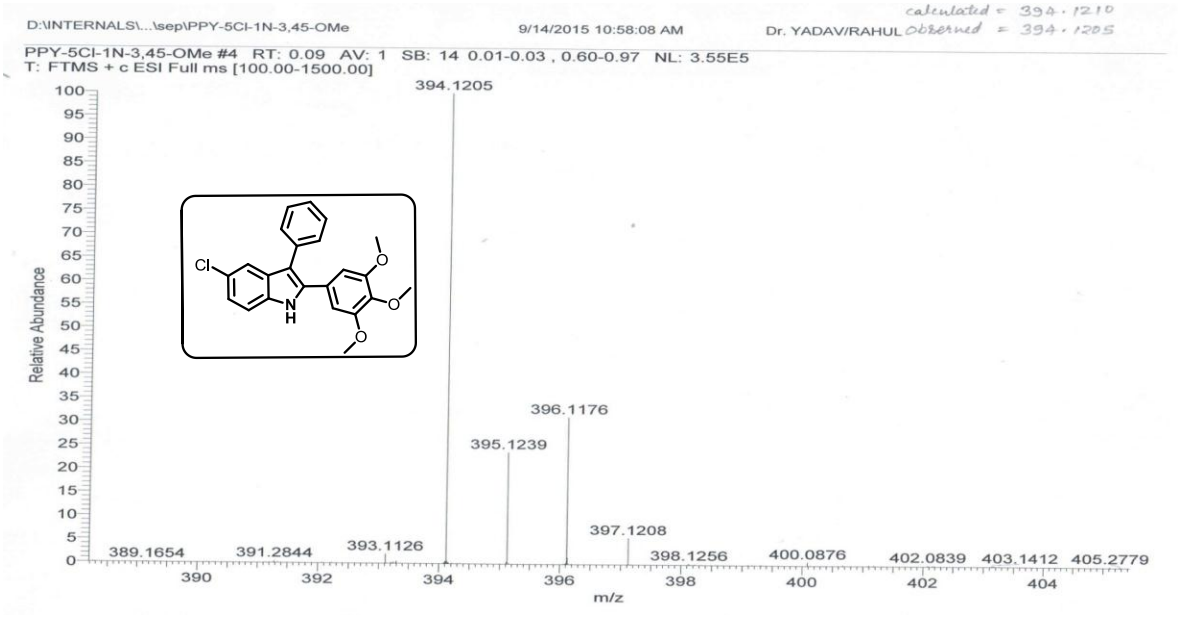

**HRMS spectrum of 3bp**



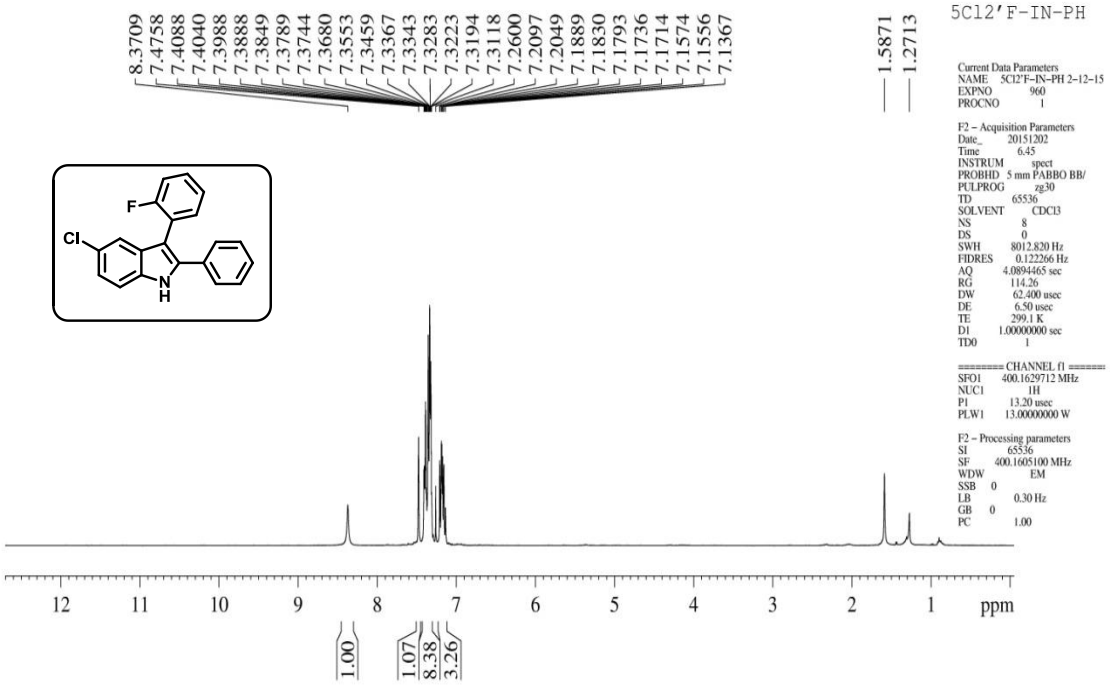

**$^{1}$H NMR spectrum of 3bq**



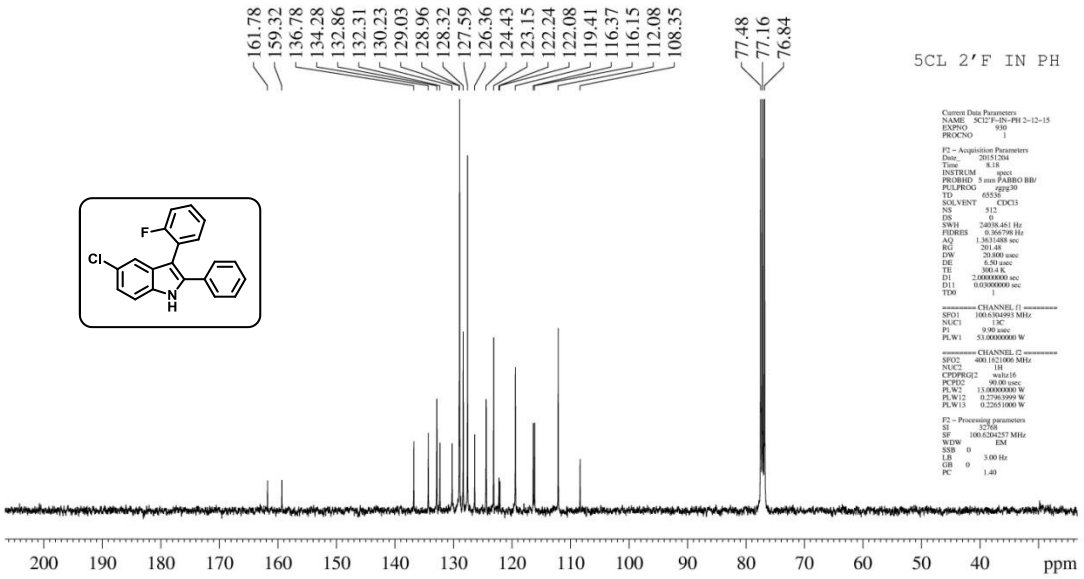

**$^{13}$C NMR spectrum of 3bq**



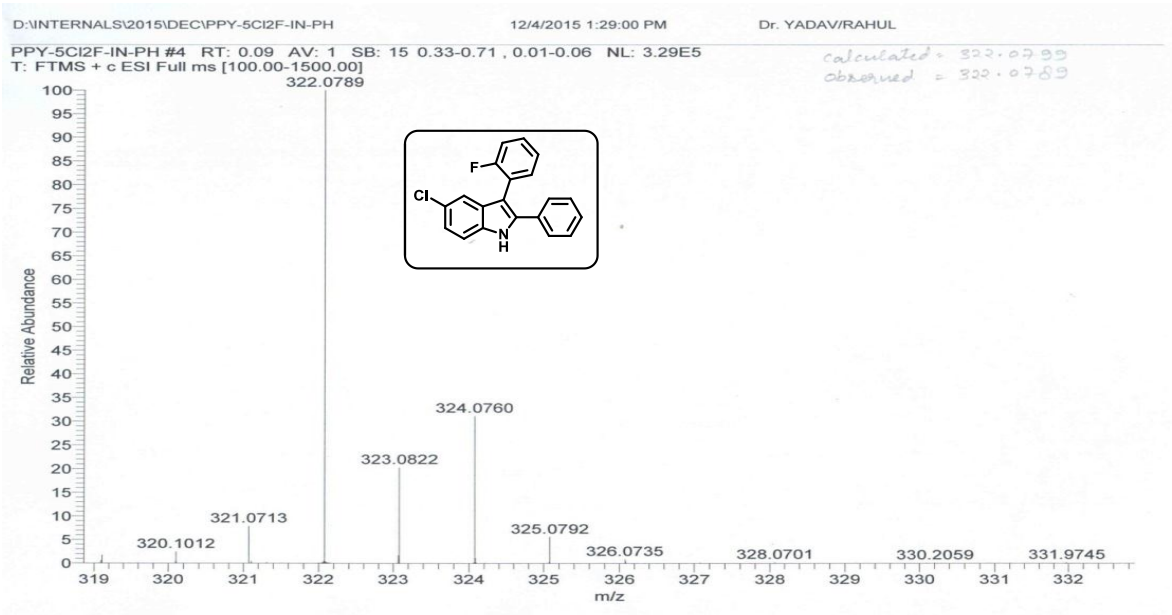

**HRMS spectrum of 3bq**



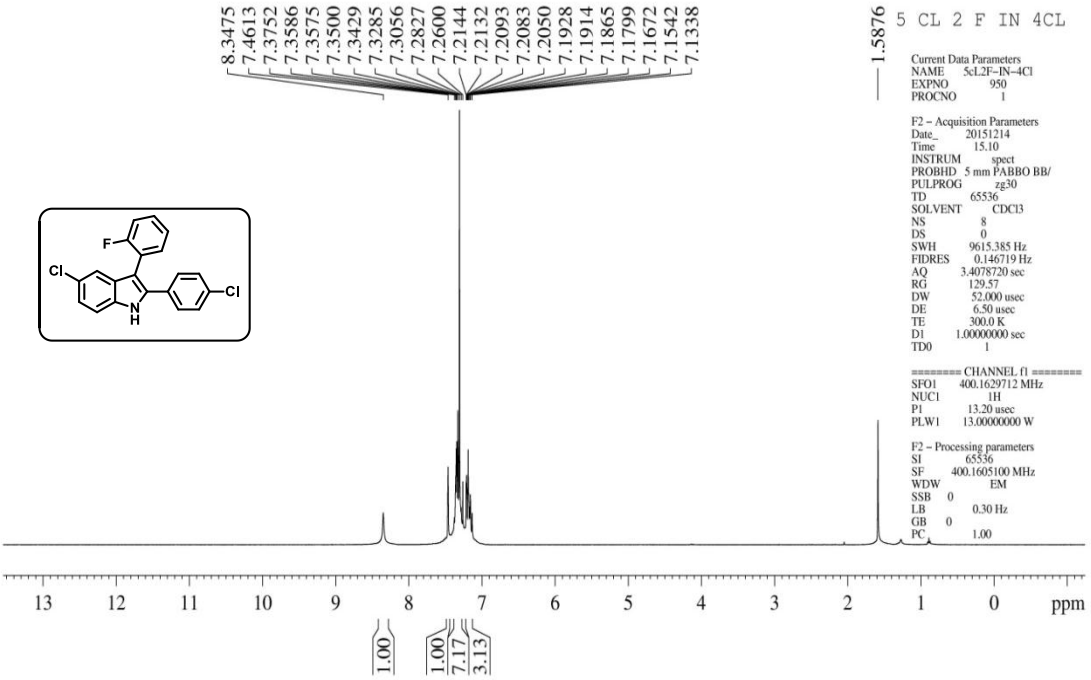

**$^{1}$H NMR spectrum of 3ca**



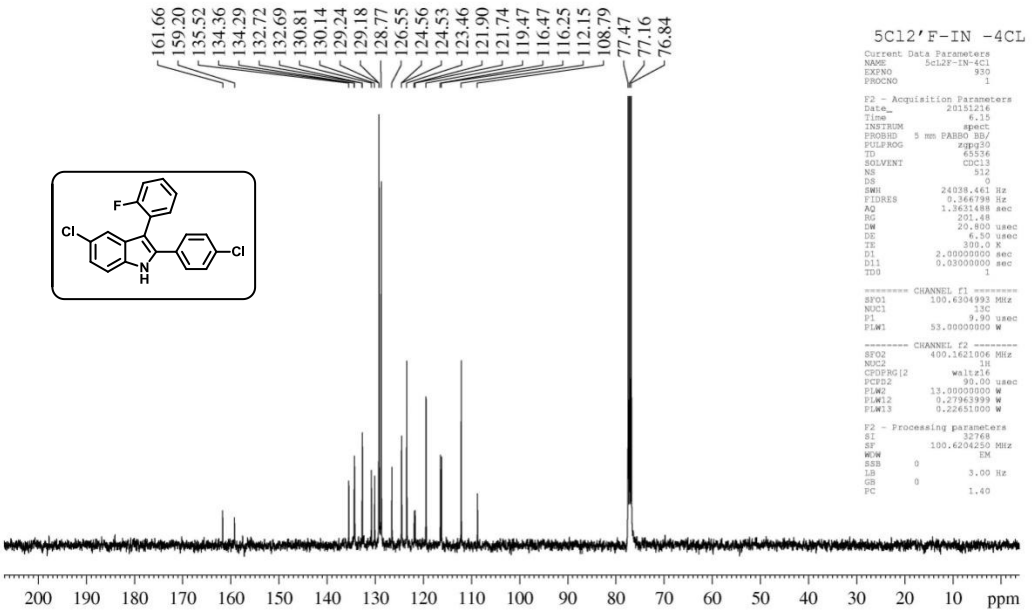

**$^{13}$C NMR spectrum of 3ca**



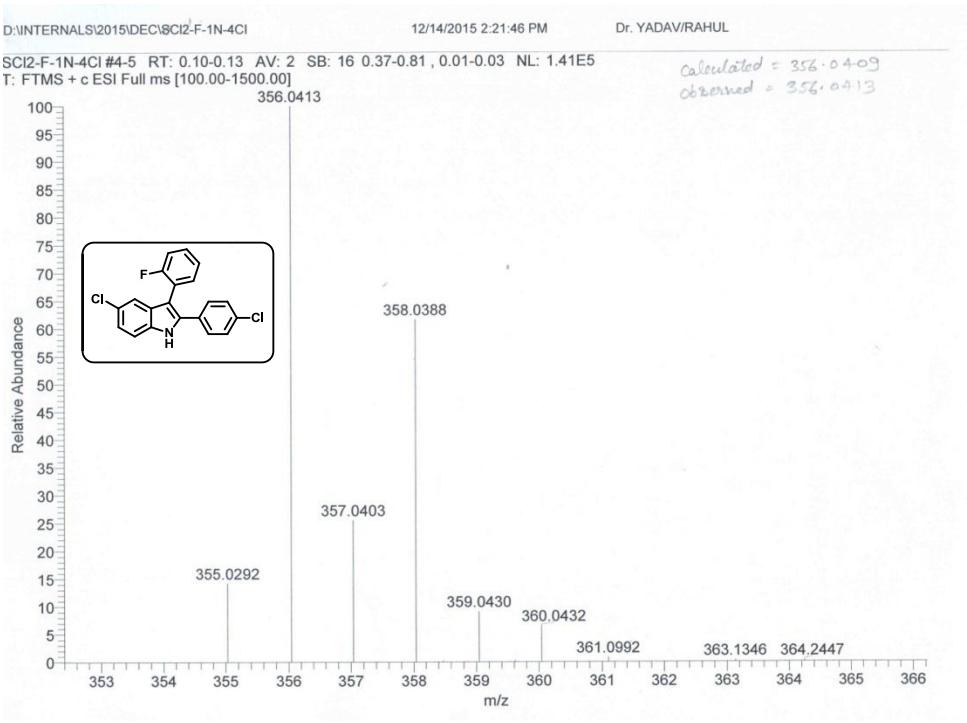

**HRMS spectrum of 3ca**



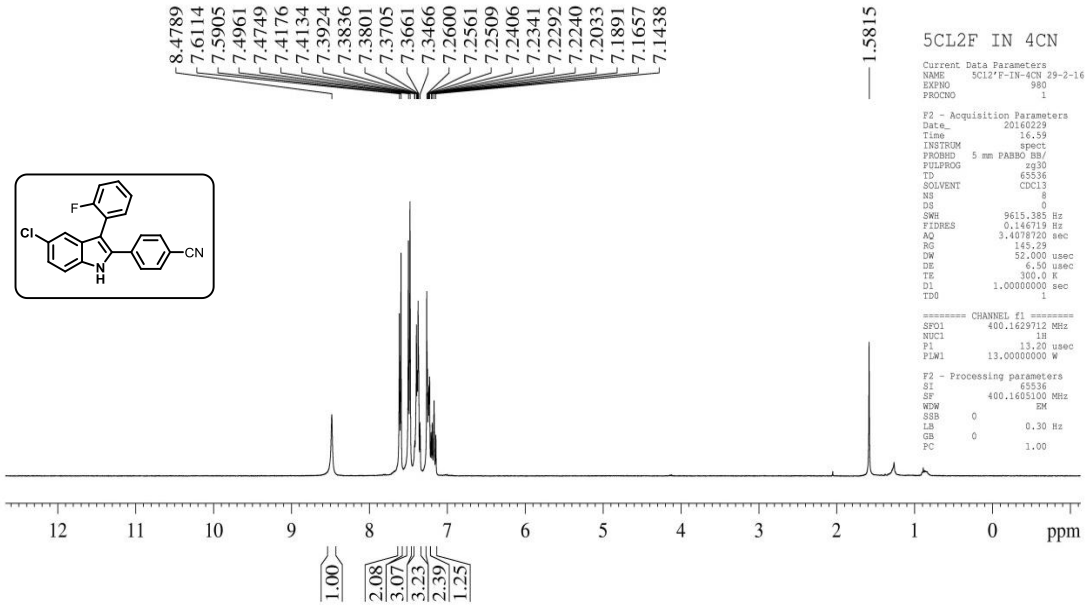

**$^{1}$H NMR spectrum of 3cc**



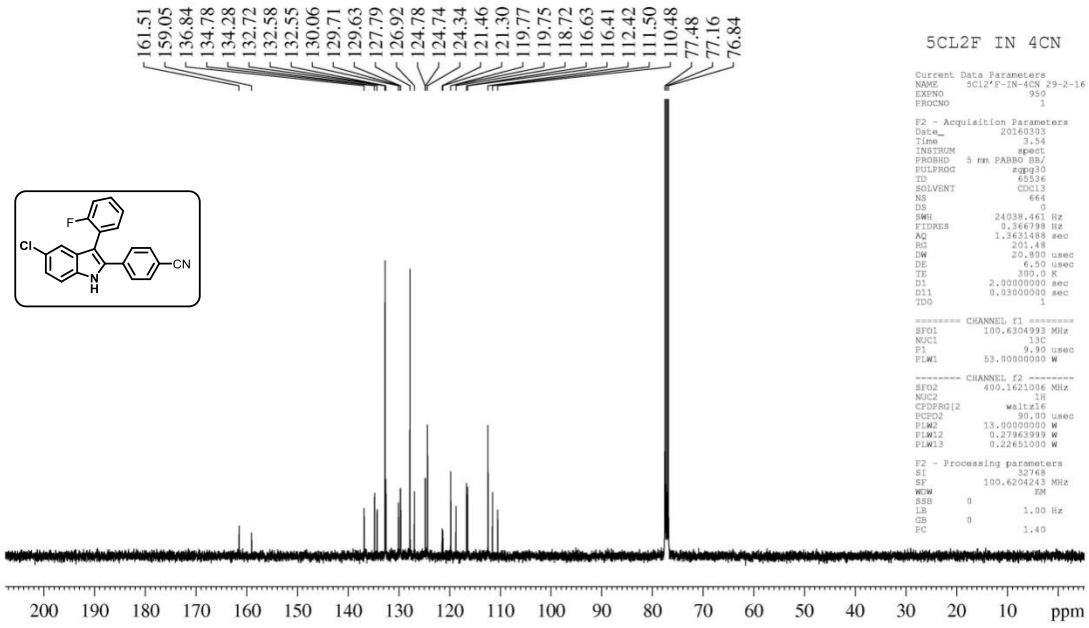

**$^{13}$C NMR spectrum of 3cc**



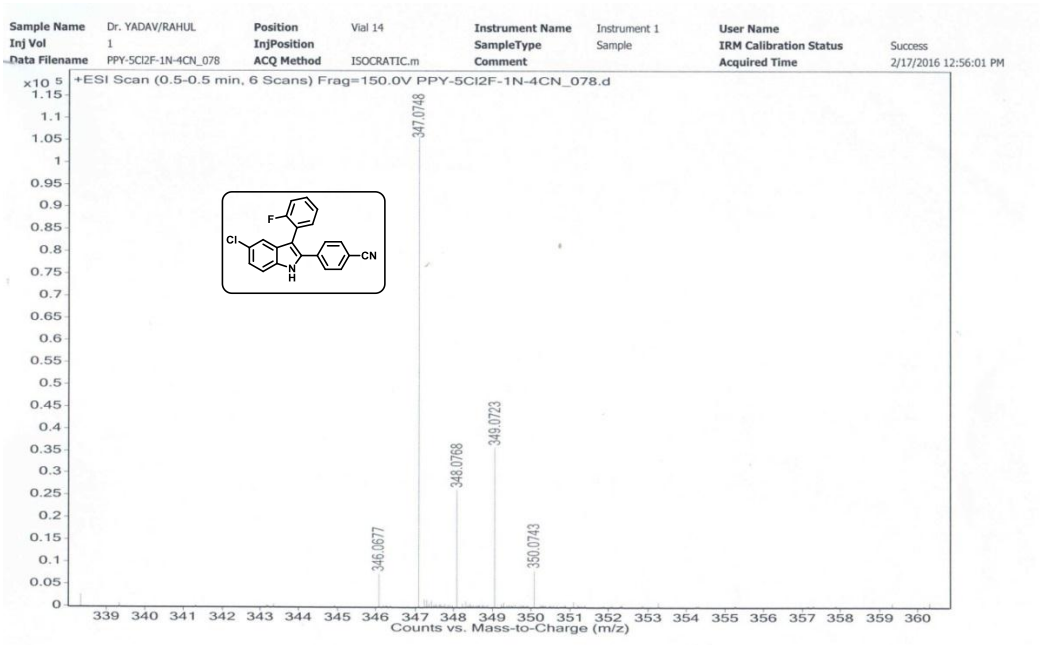

**HRMS spectrum of 3cc**



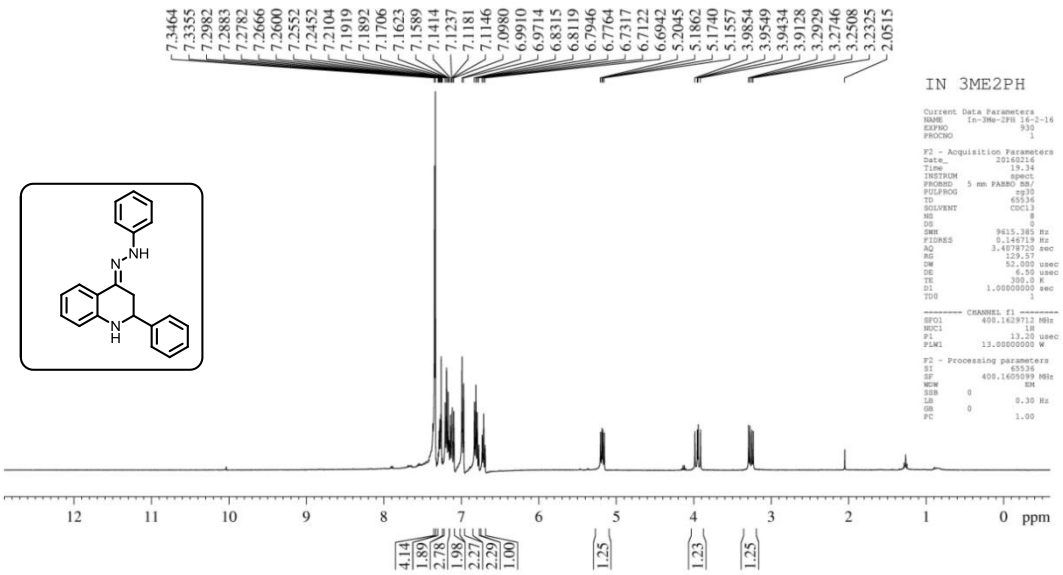

**$^{1}$H NMR spectrum of 3ck**



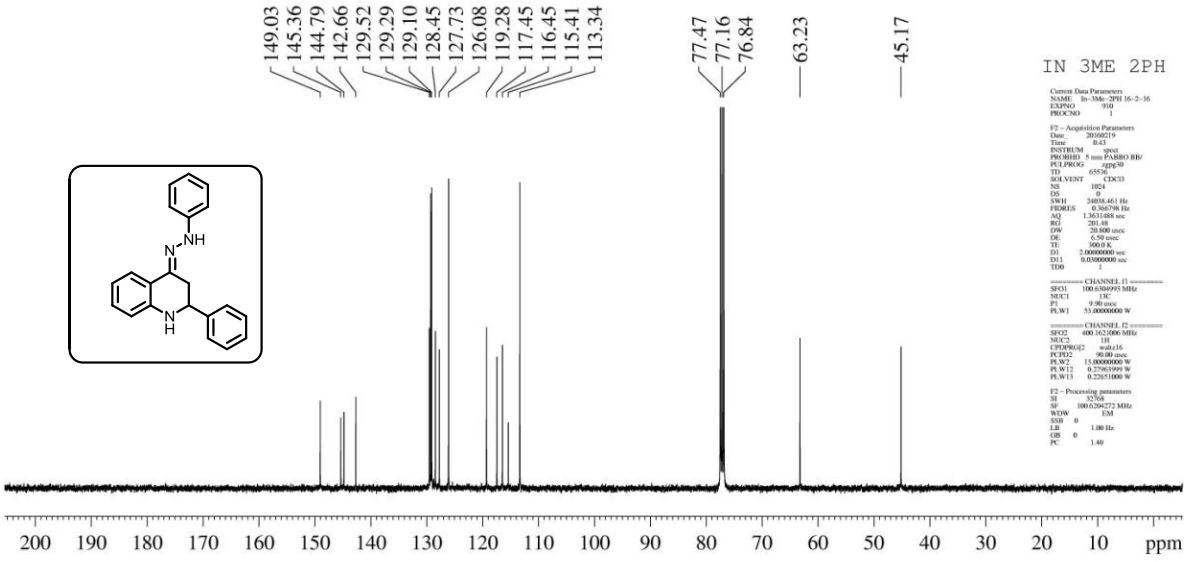

**$^{13}$C NMR spectrum of 3ck**



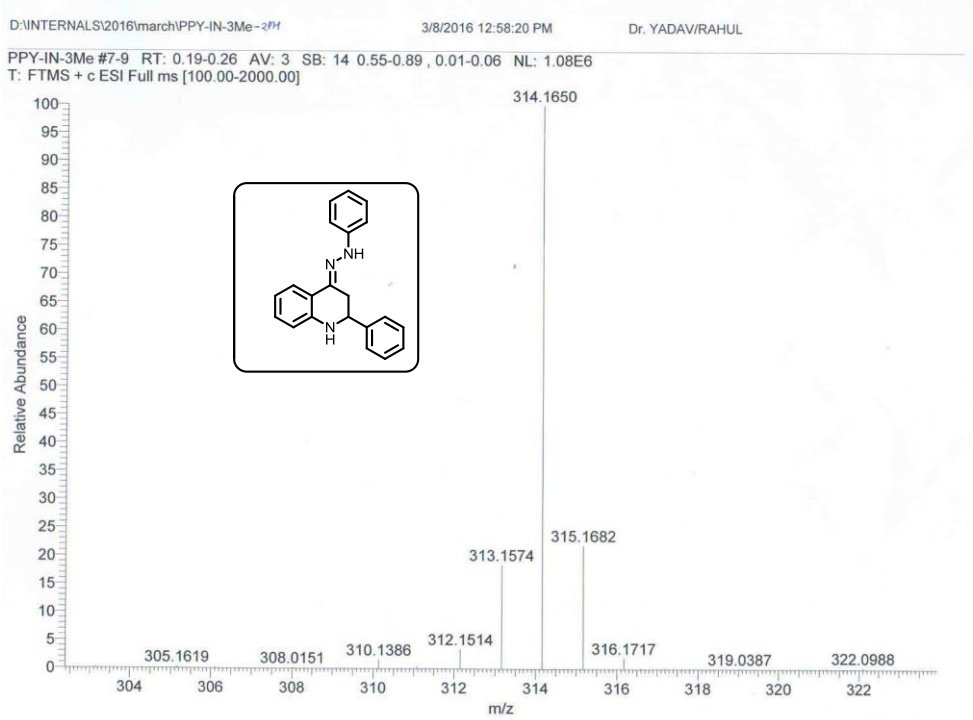

**HRMS spectrum of 3ck**



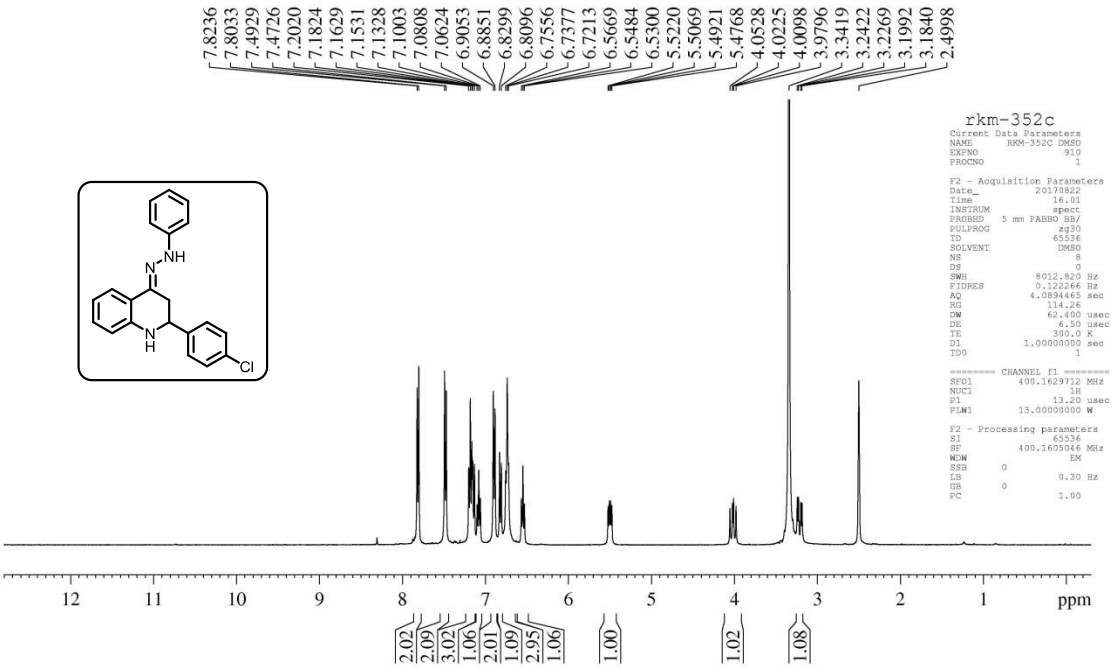

**$^{1}$H NMR spectrum of 4da**



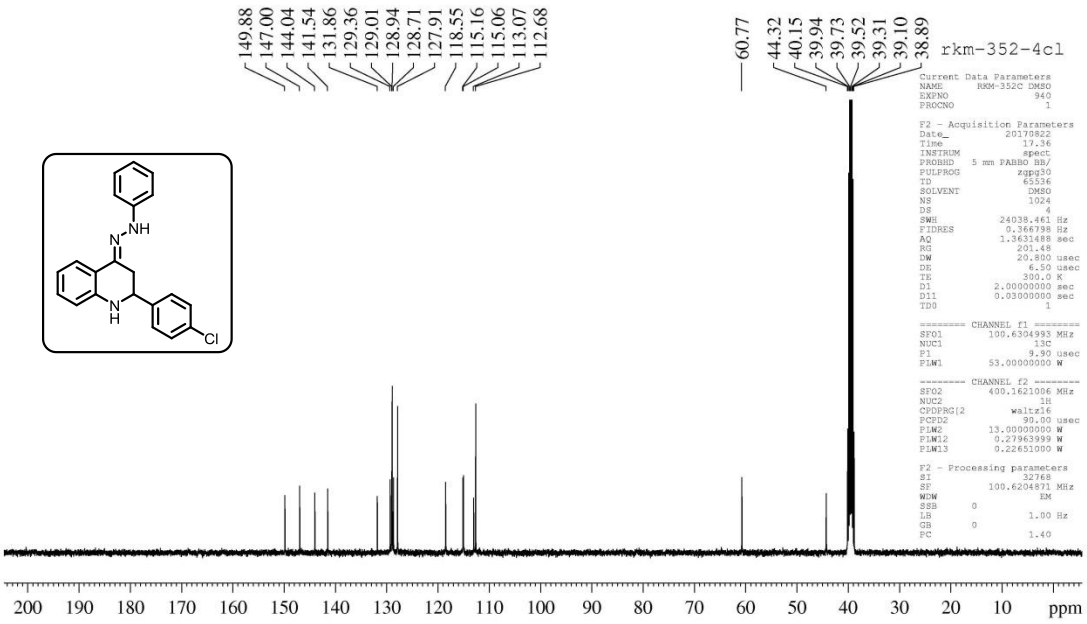

**$^{13}$C NMR spectrum of 4da**



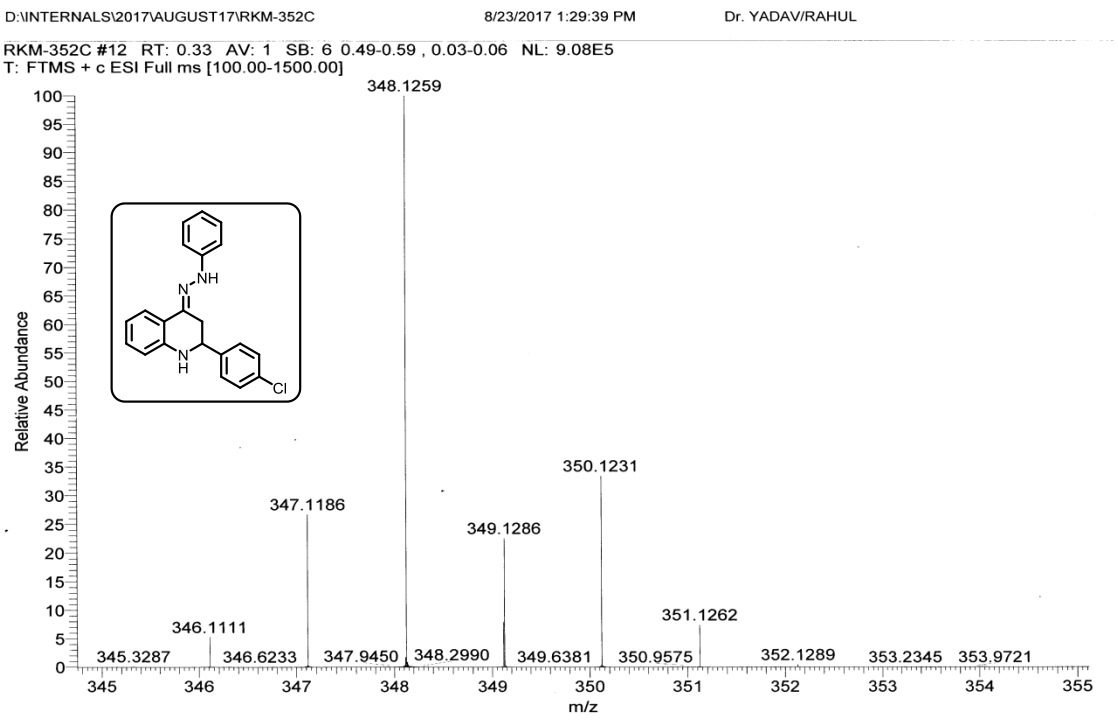

**HRMS spectrum of 4da**



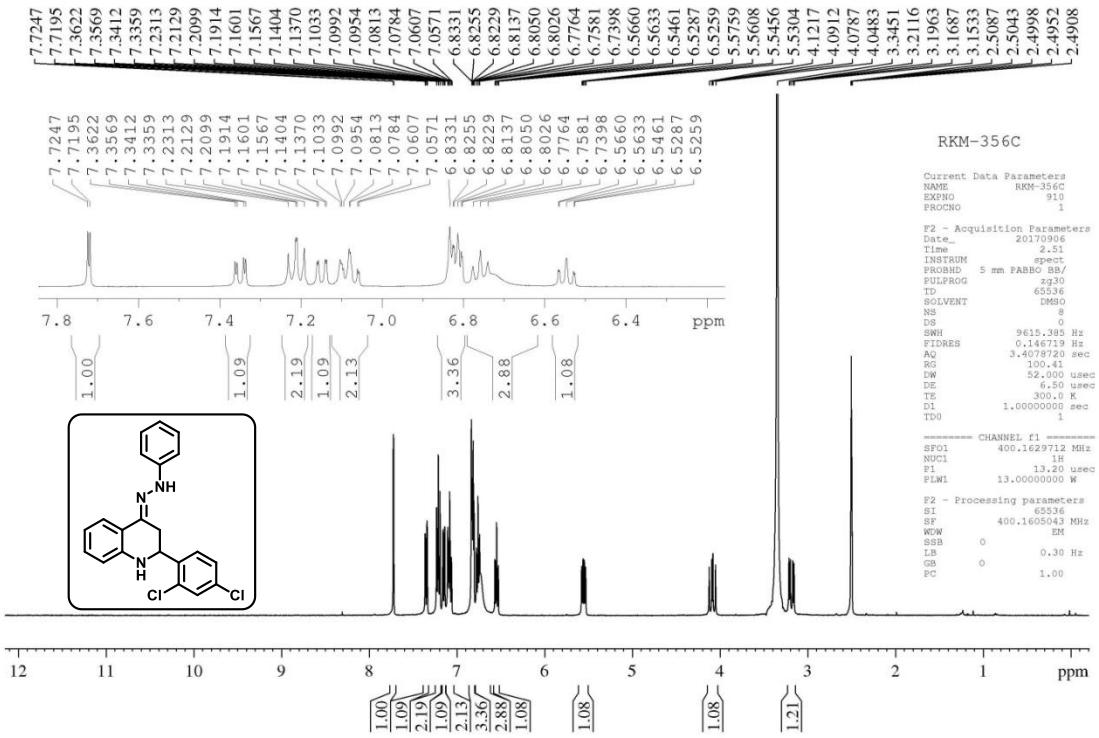

**$^{1}$H NMR spectrum of 4dc**



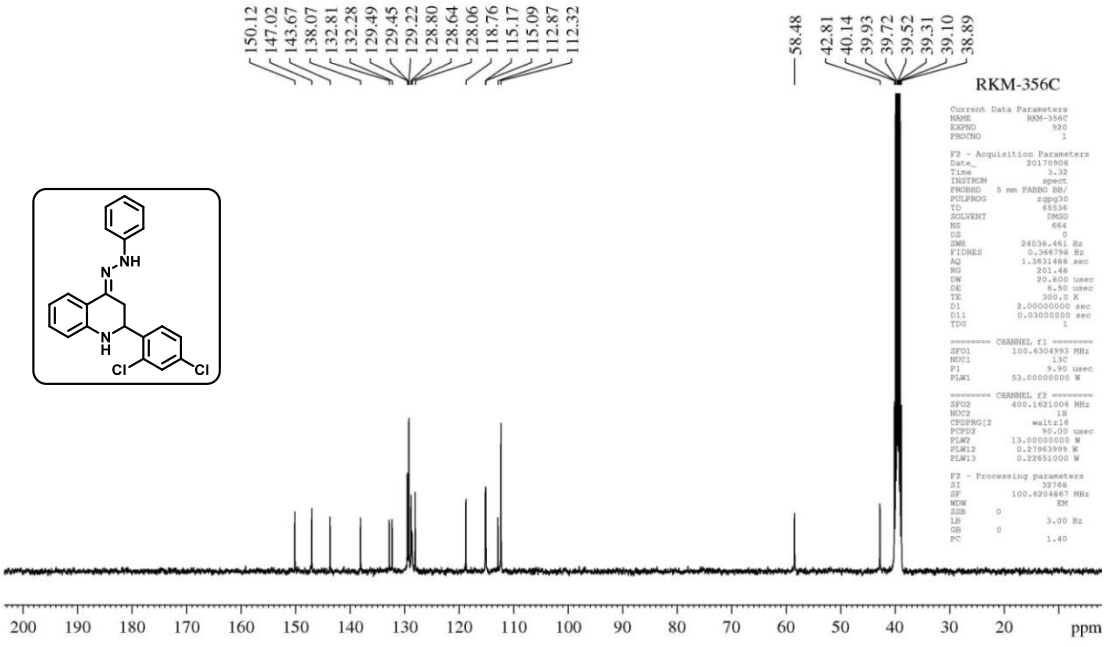

**$^{13}$C NMR spectrum of 4dc**



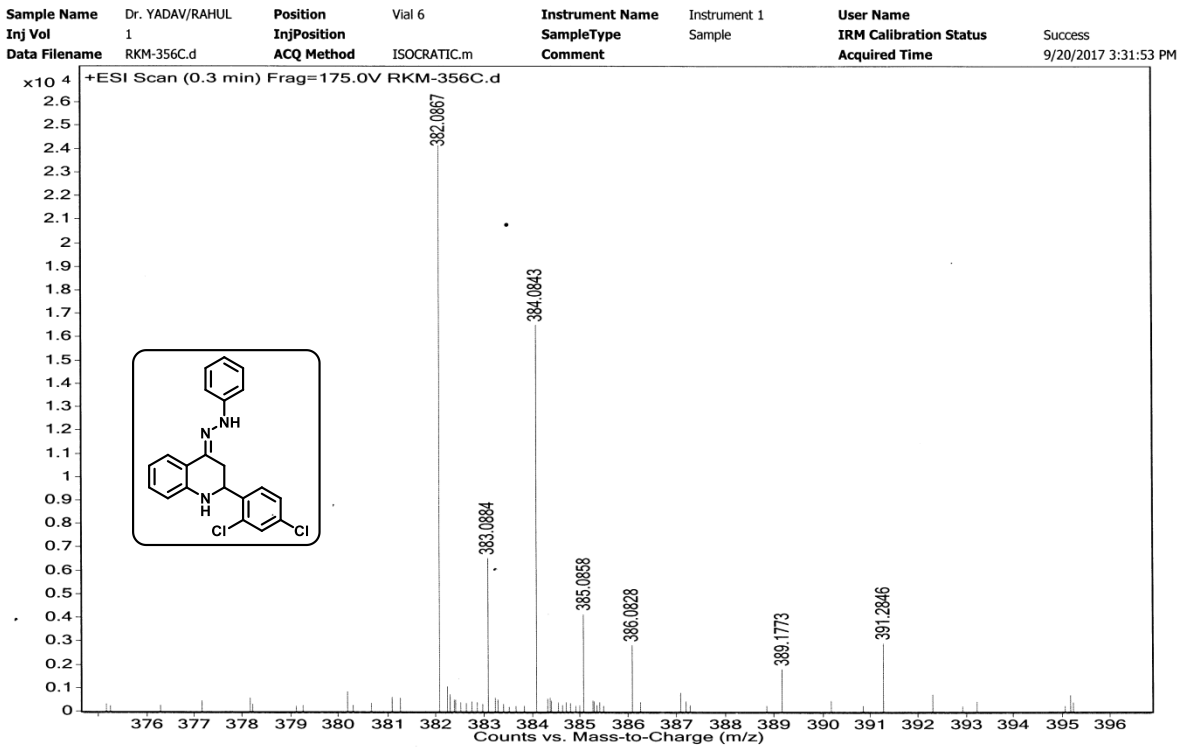

**HRMS spectrum of 4dc**



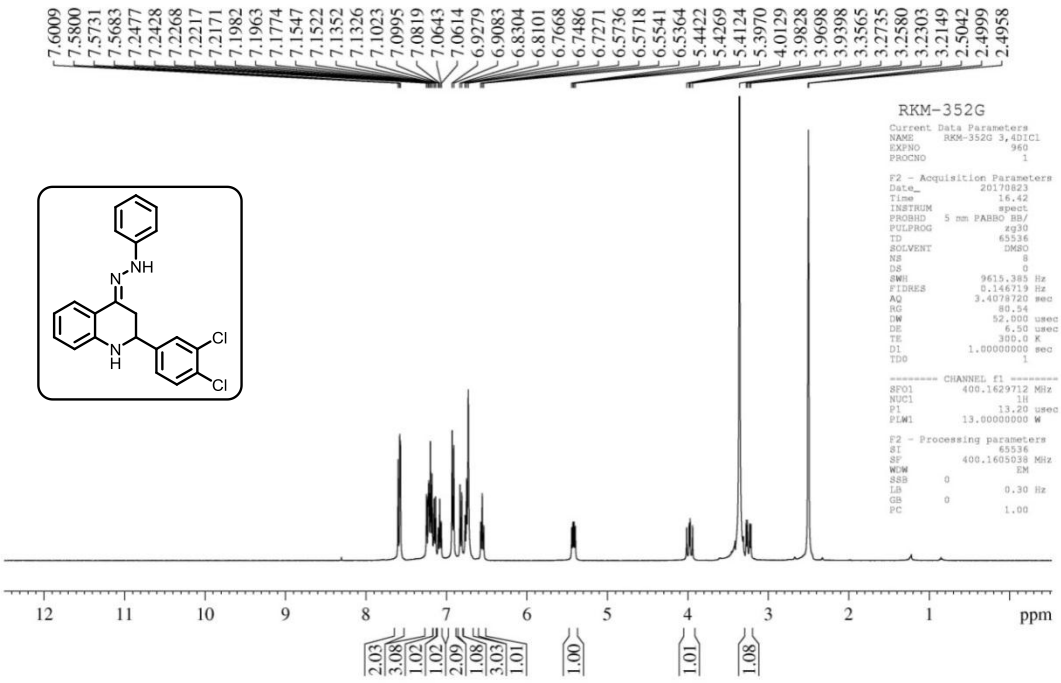

**$^{1}$H NMR spectrum of 4dg**



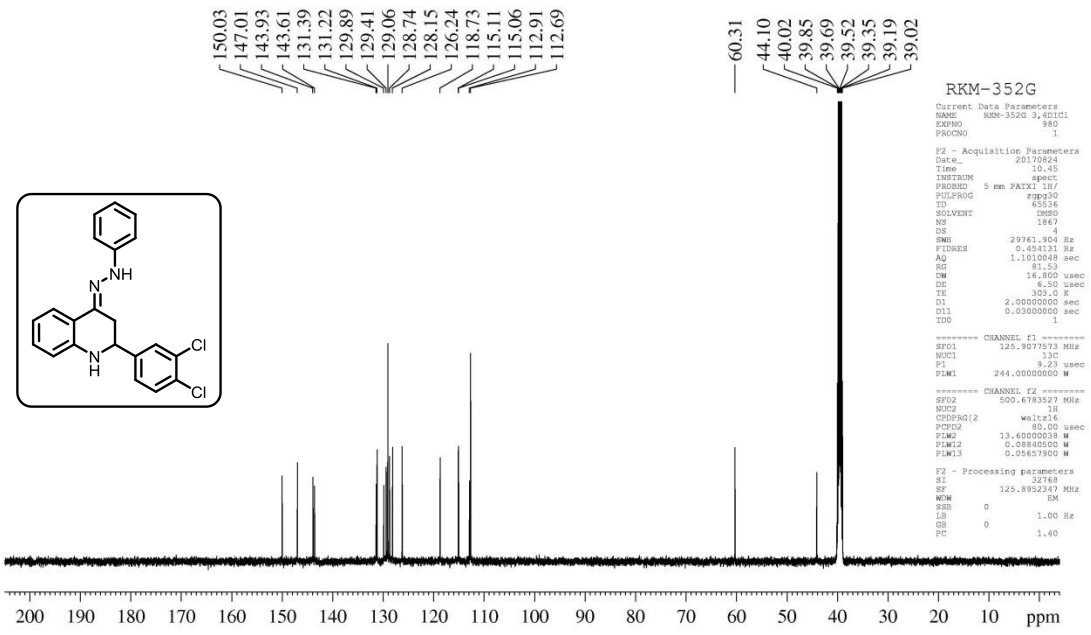

**$^{13}$C NMR spectrum of 4dg**



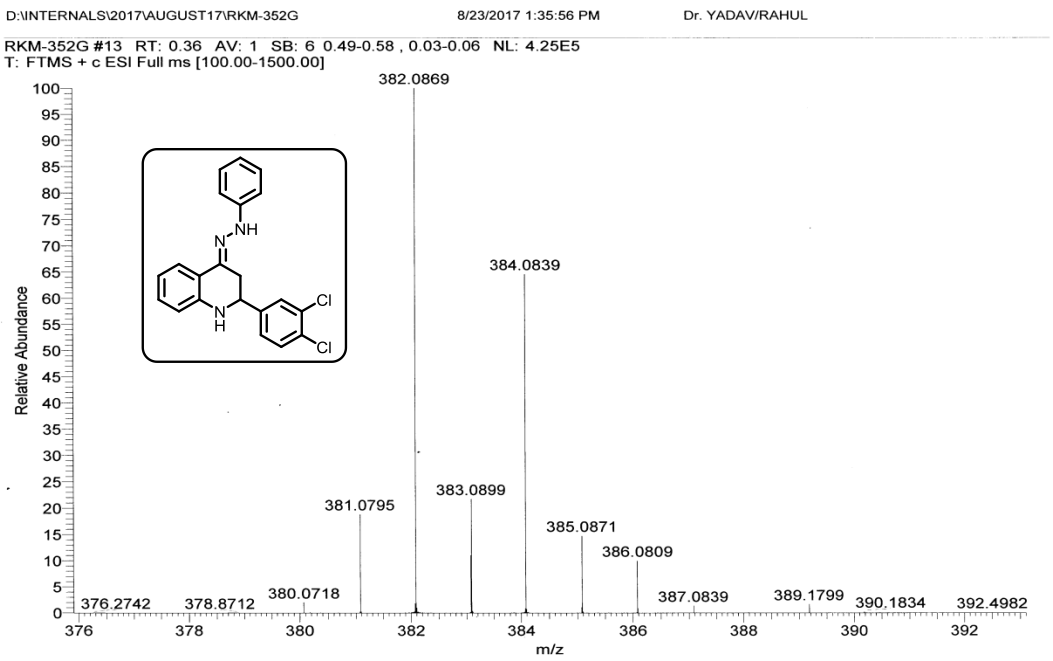

**HRMS spectrum of 4dg**



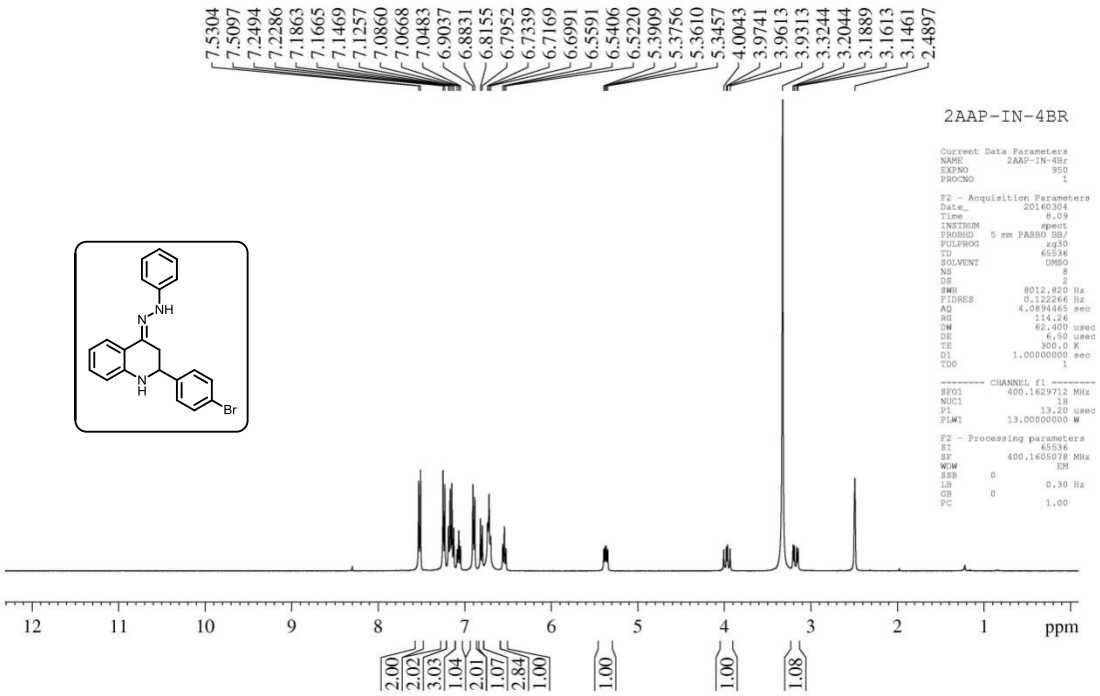

**$^{1}$H NMR spectrum of 4dv**



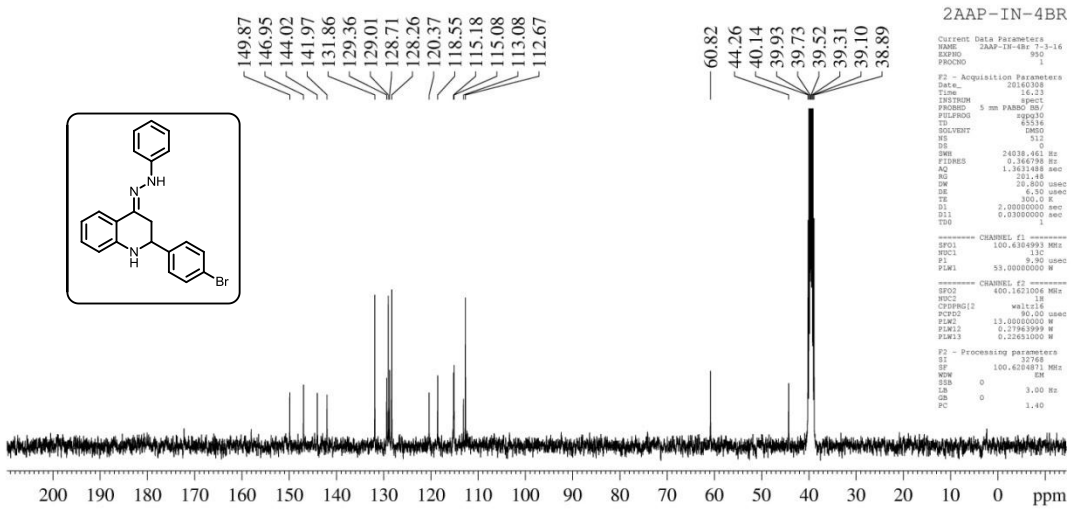

**$^{13}$C NMR spectrum of 4dv**



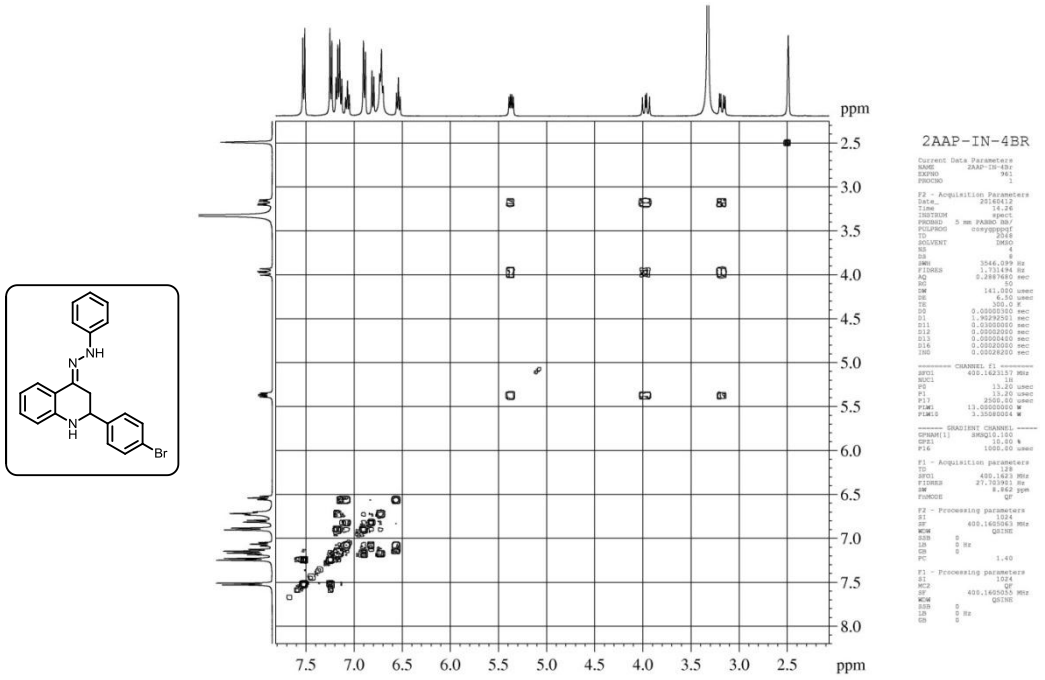

**HRMS spectrum of 4dv**



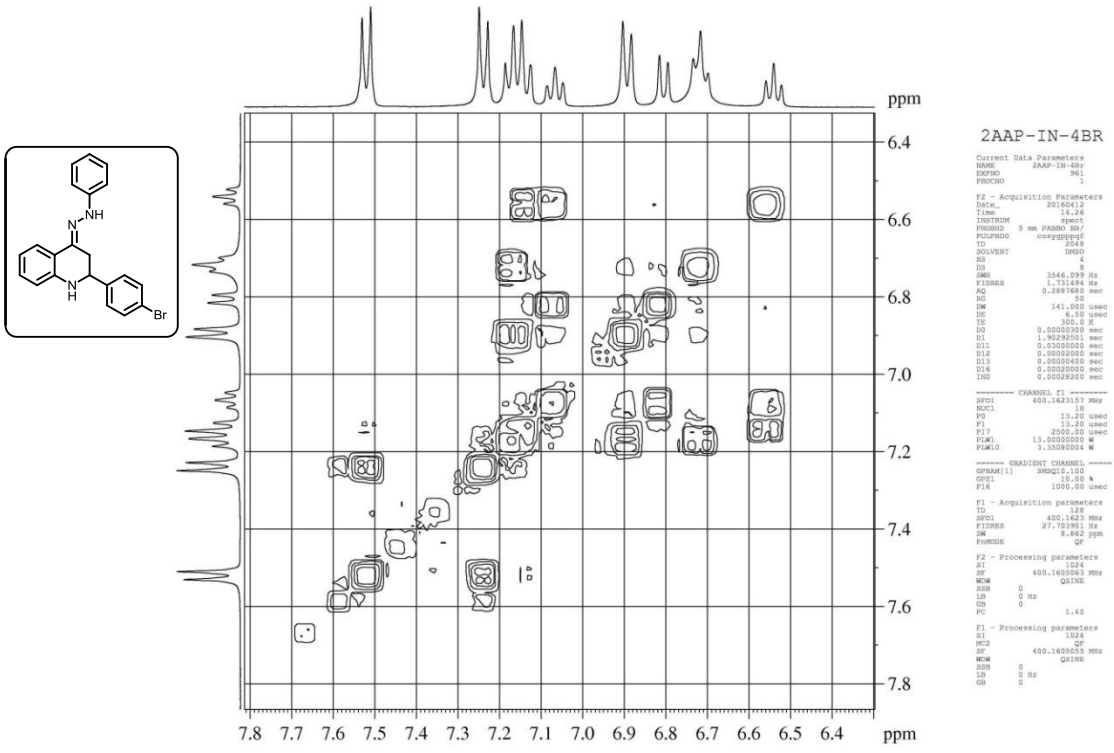

**$^{1}$H NMR spectrum of 4de**



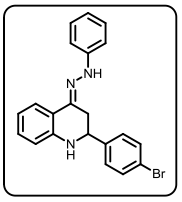

**$^{13}$C NMR spectrum of 4de**

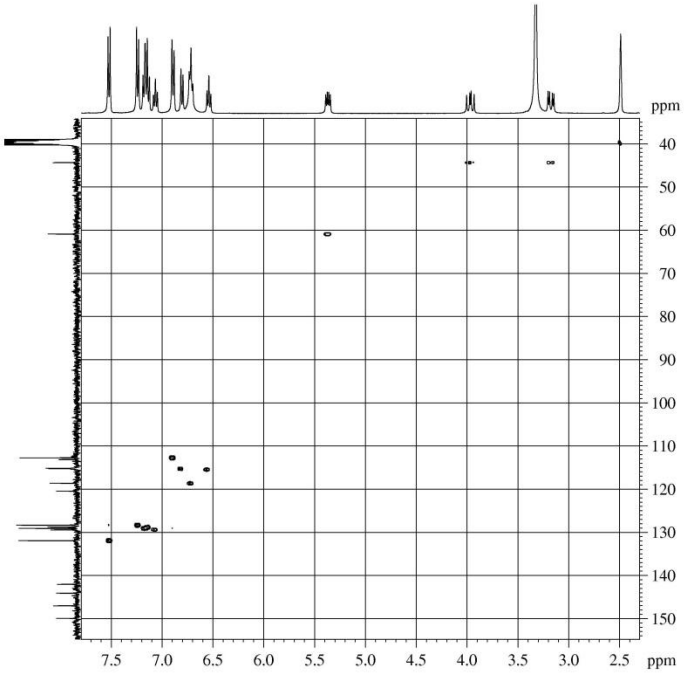



**COSY spectrum of 4de**



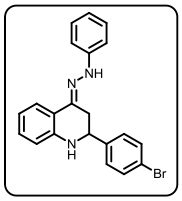

**Expanded COSY spectrum of 4de**

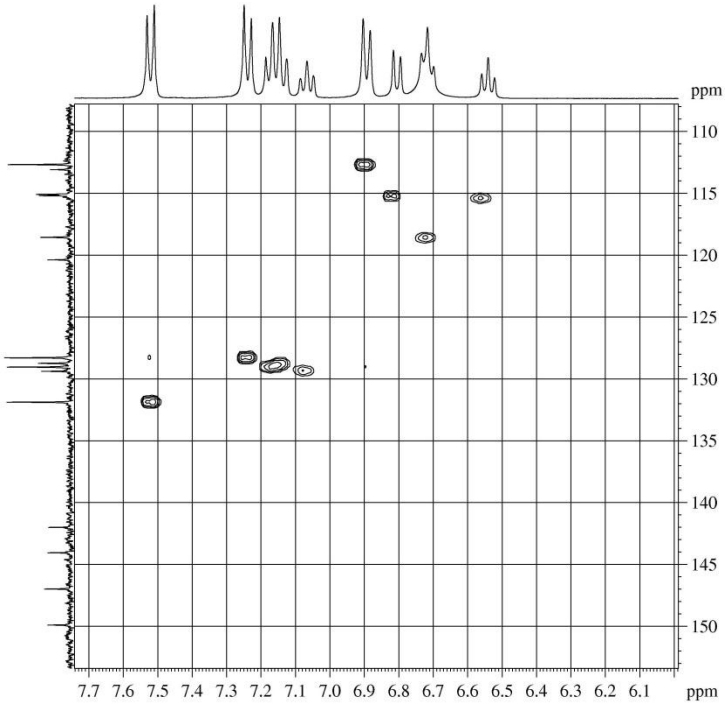




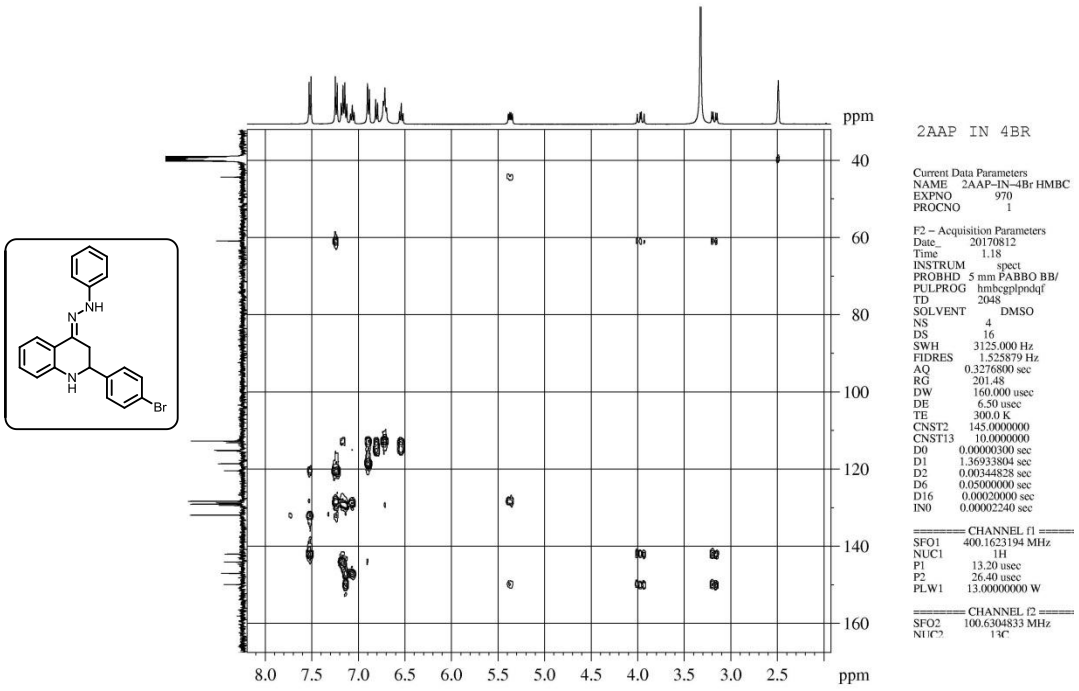
**HSQC spectrum of 4de**



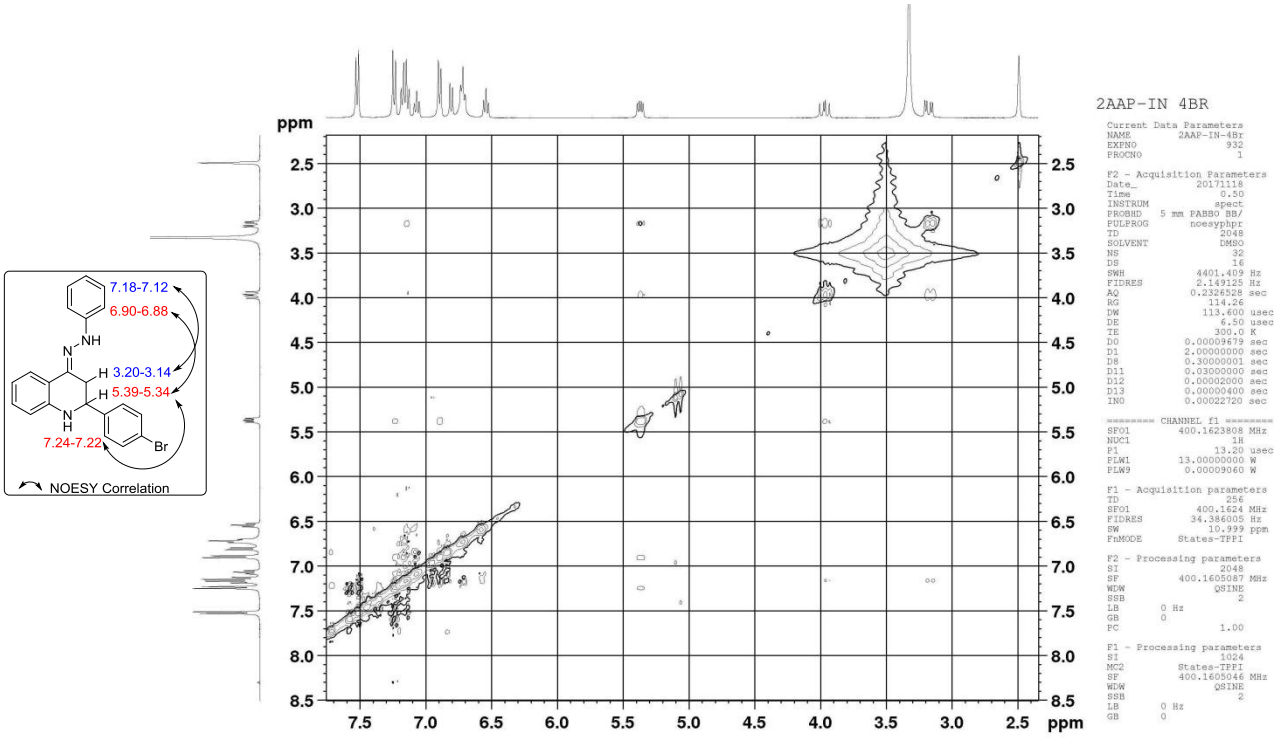





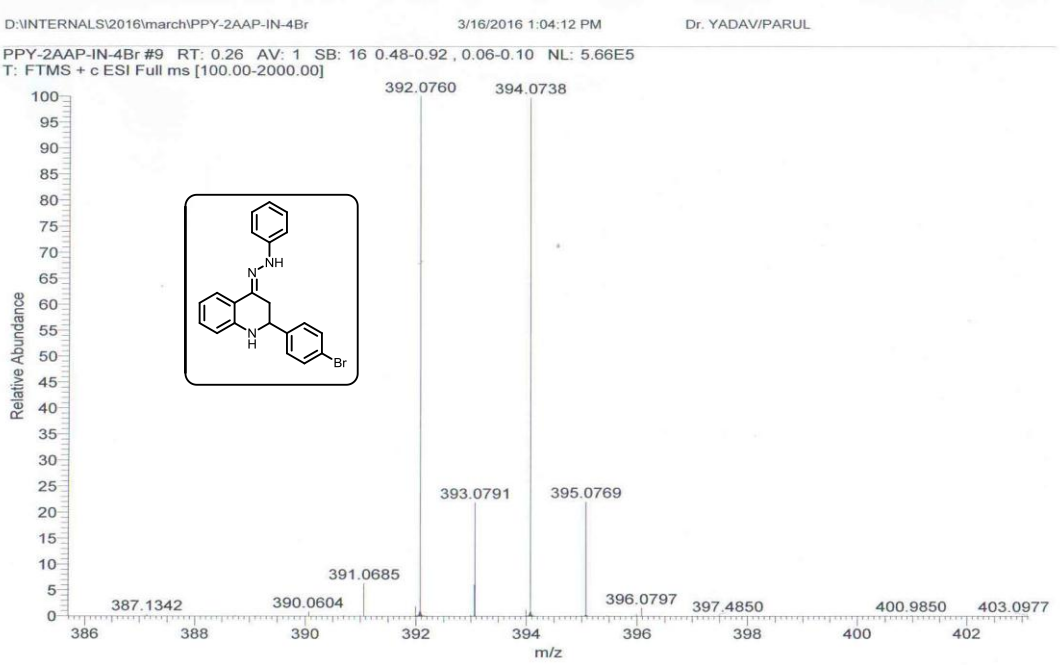
**Expanded HSQC spectrum of 4de**



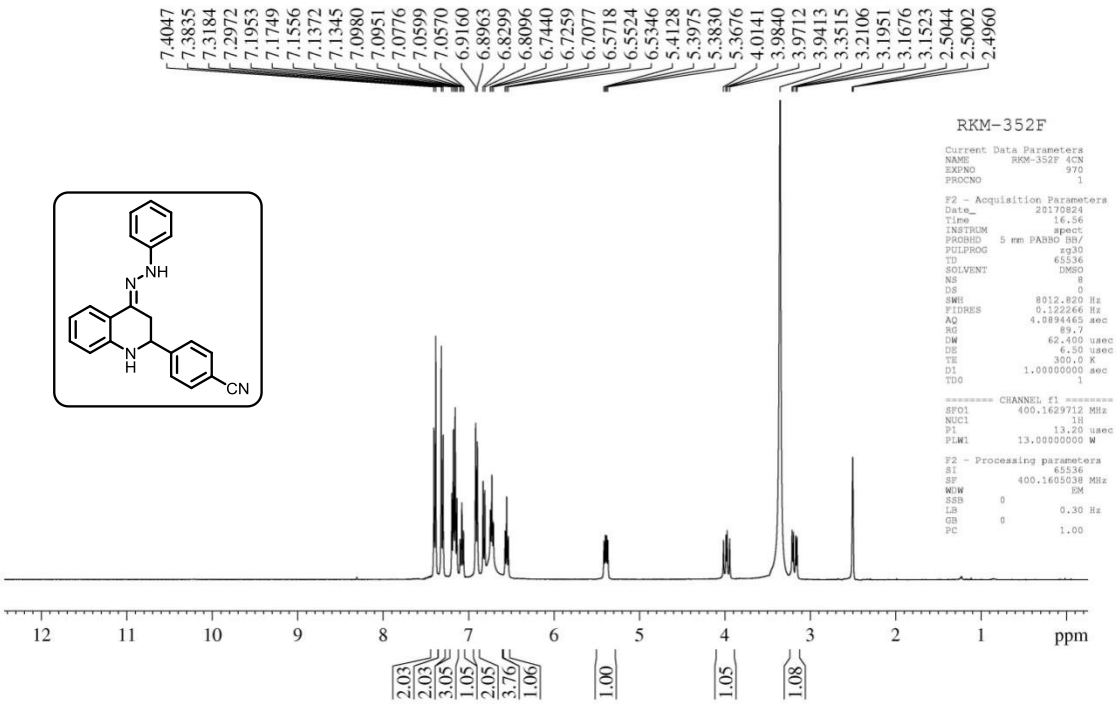

**HMBC spectrum of 4de**



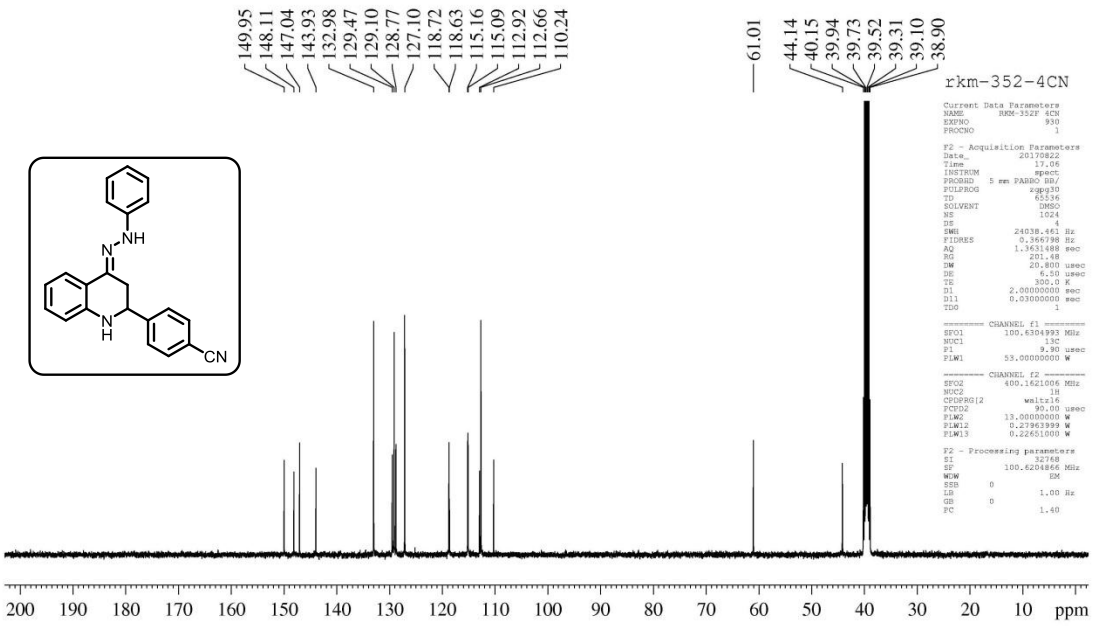

**NOESY spectrum of 4de**



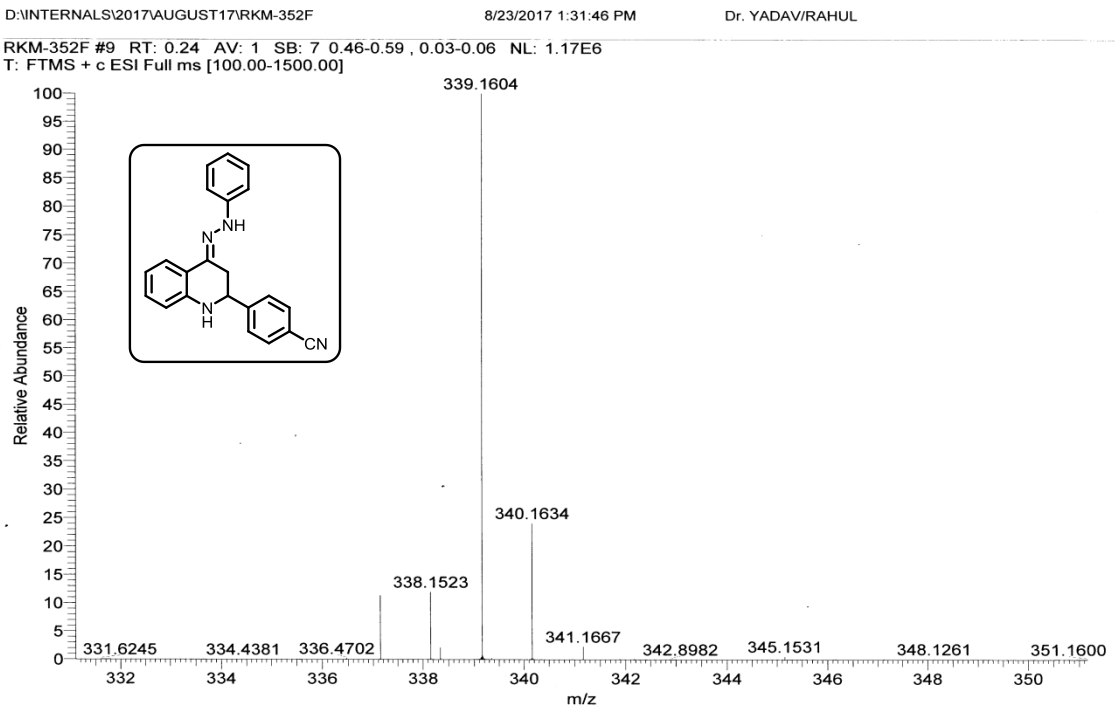

**HRMS spectrum of 4de**



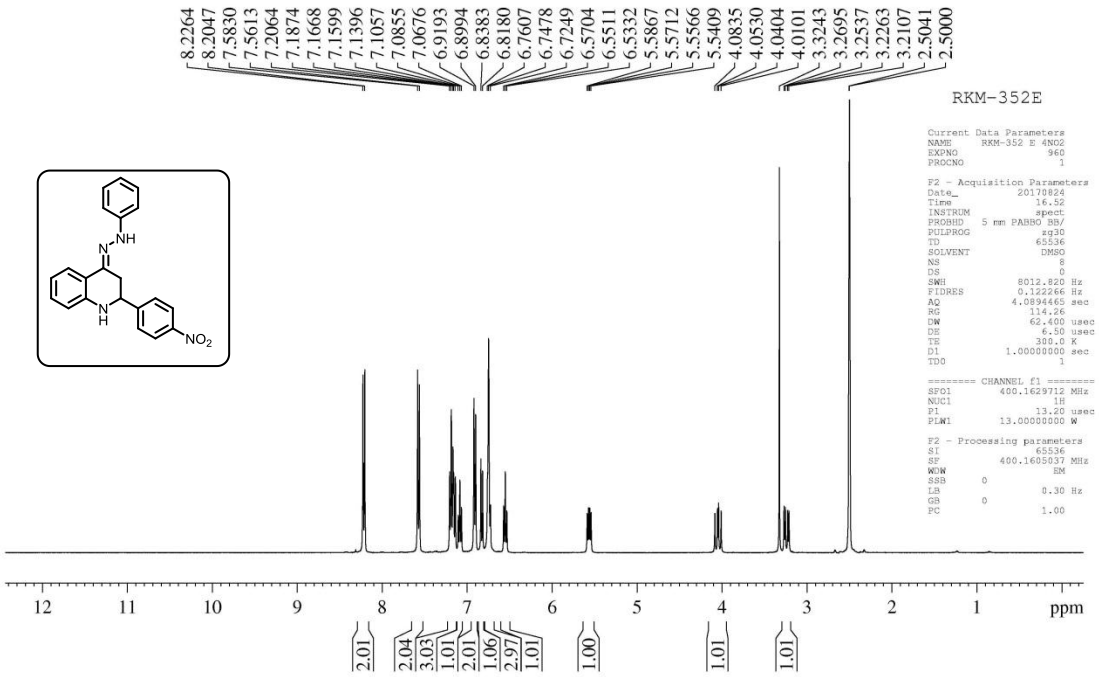

**$^{1}$H NMR spectrum of 4dk**



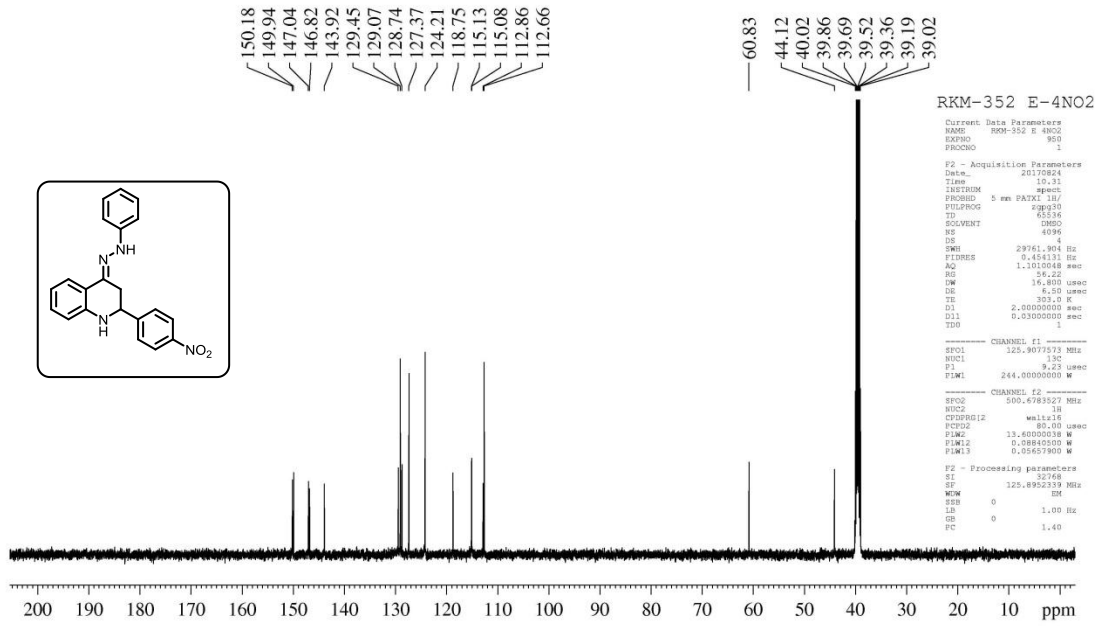

**$^{13}$C NMR spectrum of 4dk**



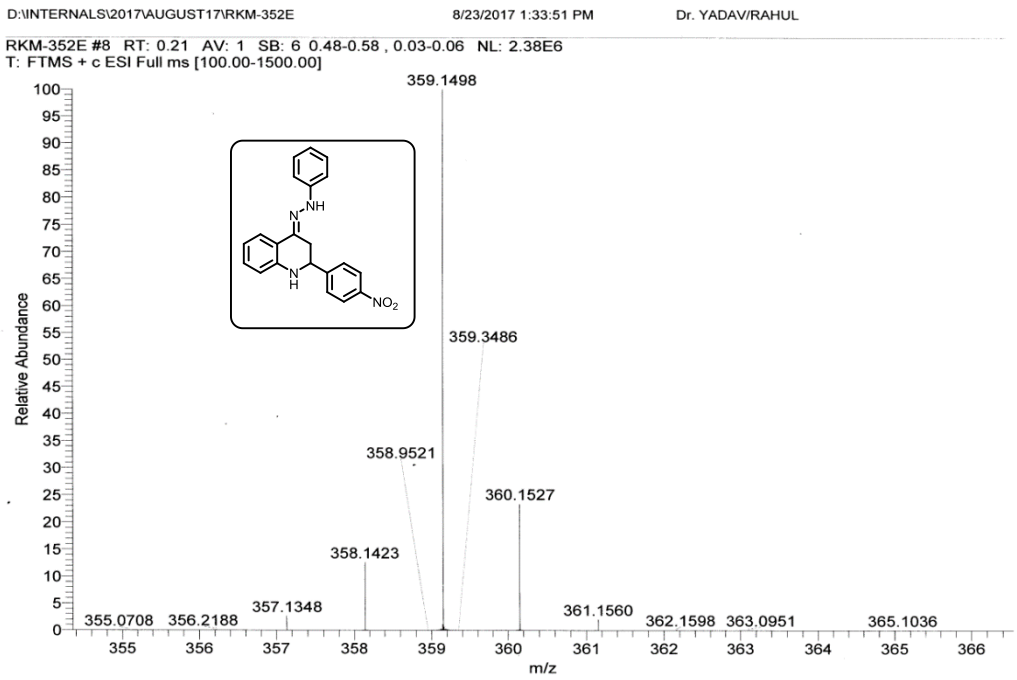

**HRMS spectrum of 4dk**



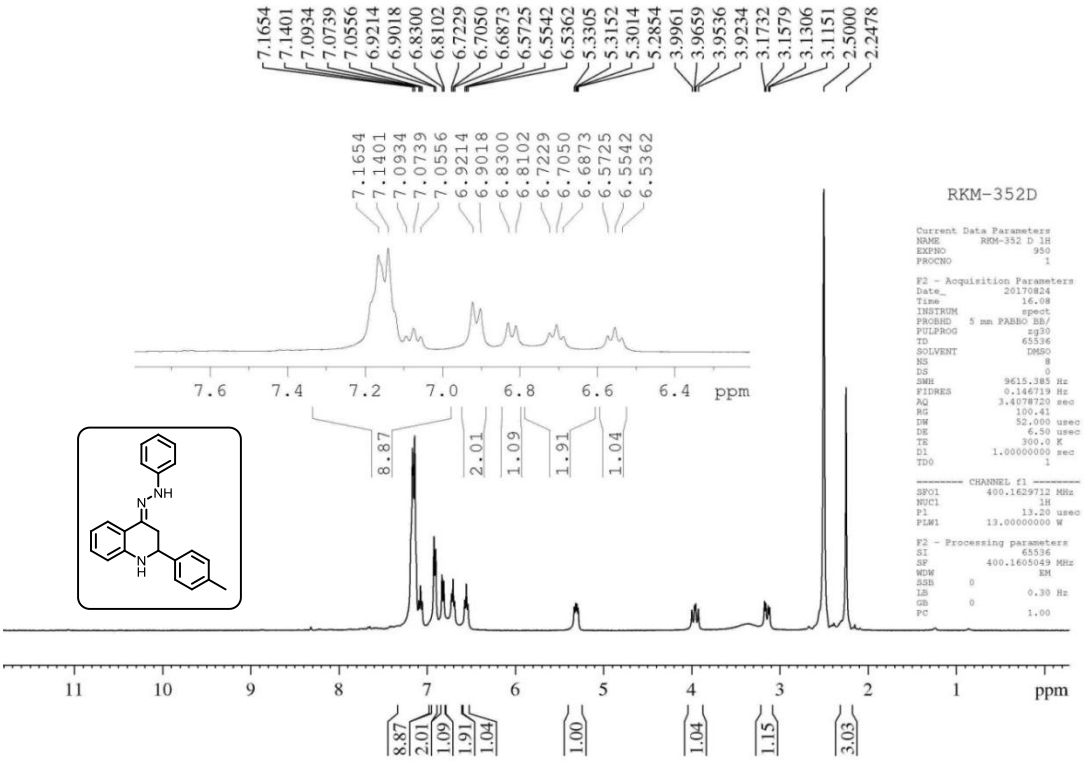

**[1]H NMR spectrum of 4dl**



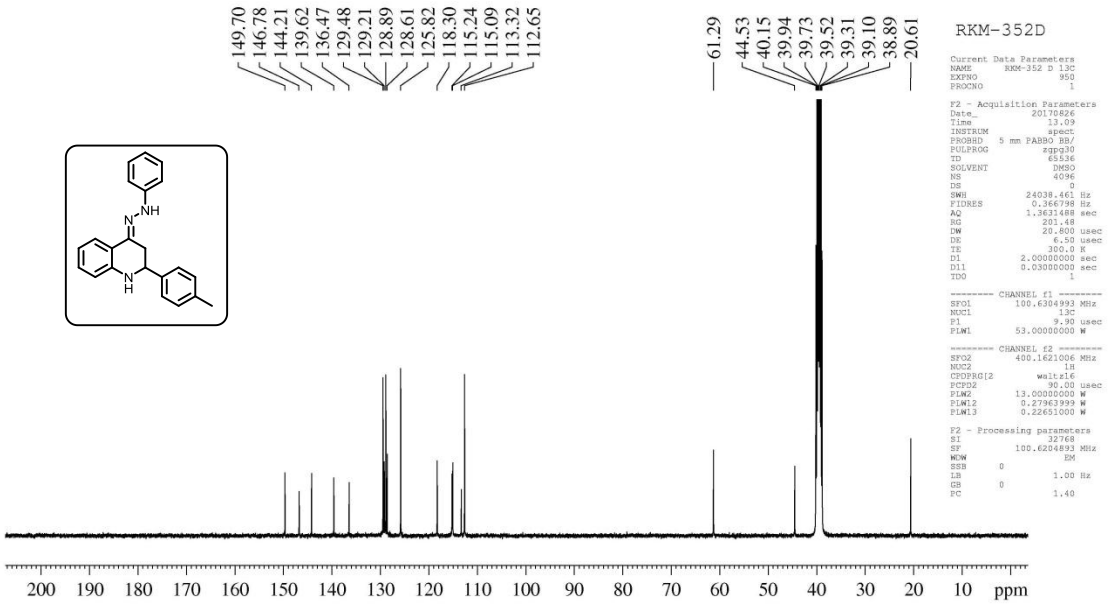

**[13]C NMR spectrum of 4dl**



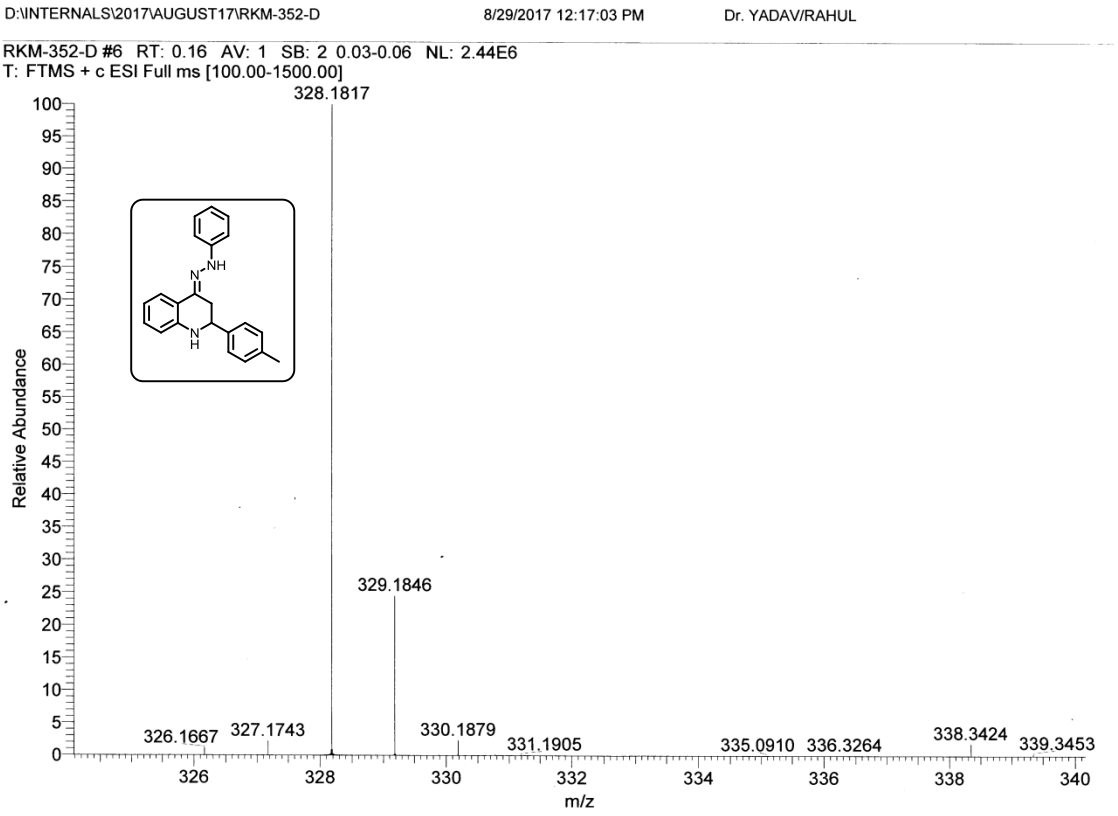

**HRMS spectrum of 4dl**



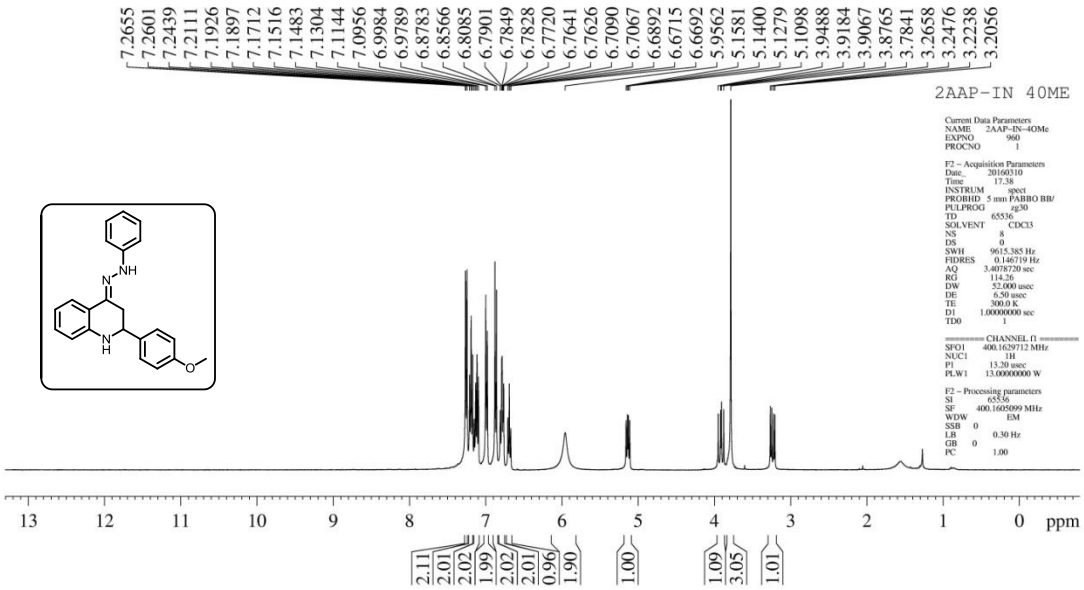

**$^{1}$H NMR spectrum of 4dm**



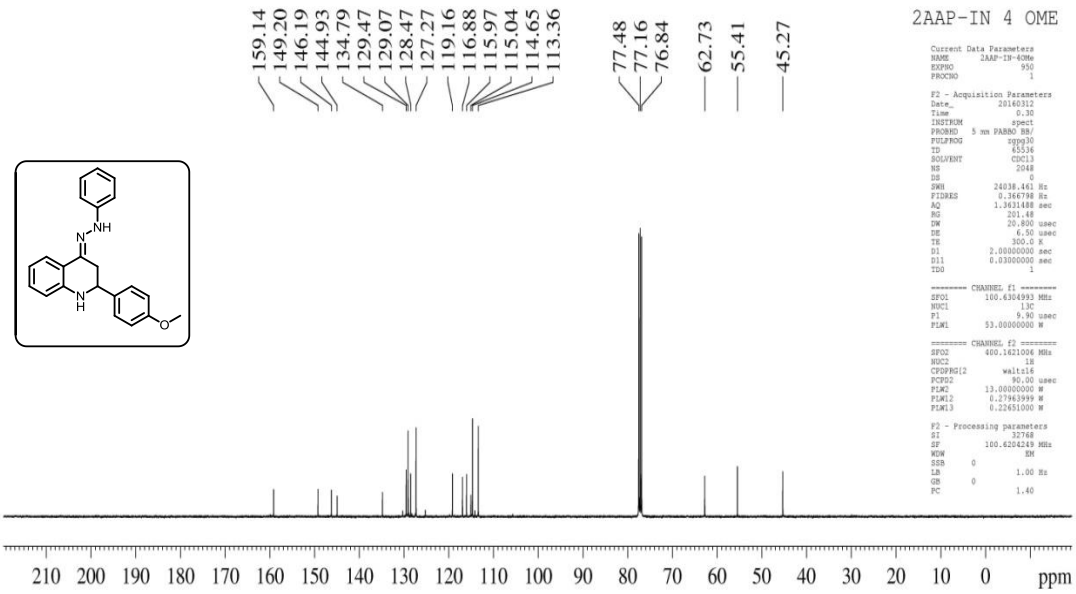

**$^{13}$C NMR spectrum of 4dm**



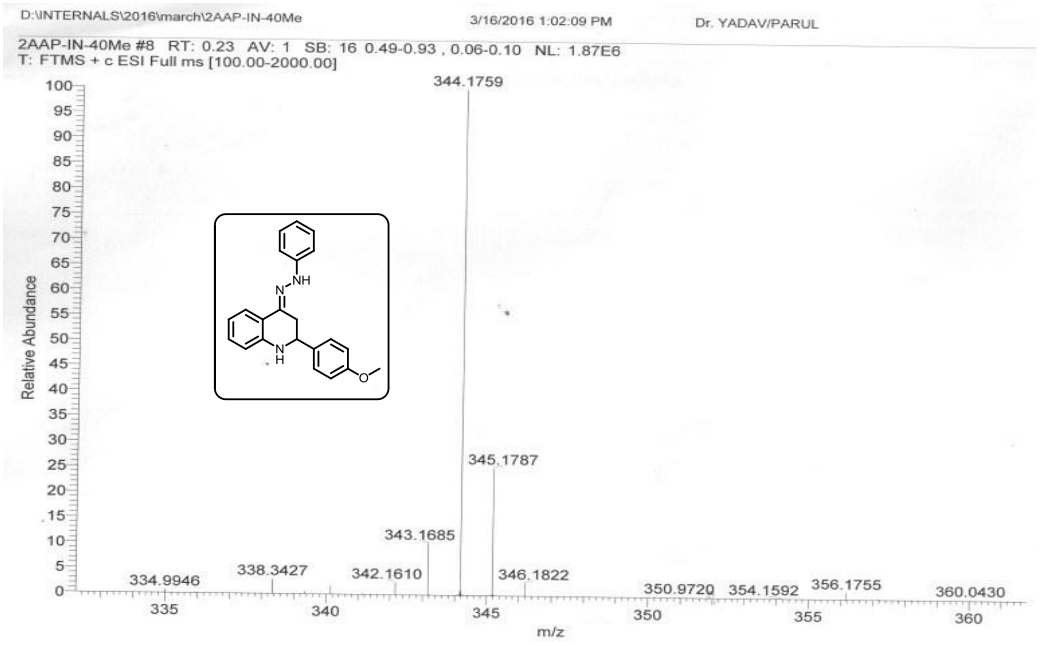

**HRMS spectrum of 4dm**



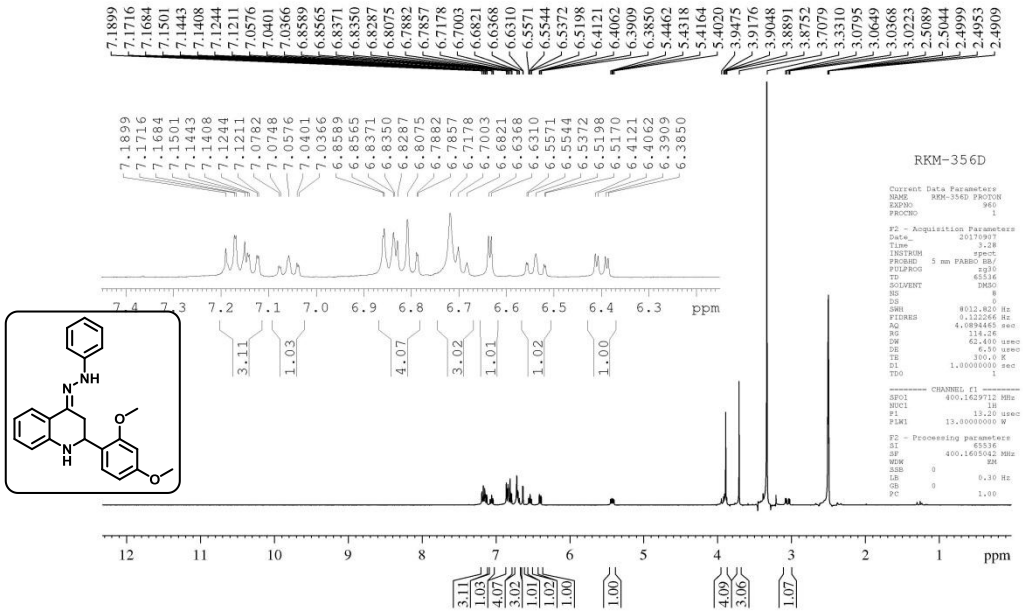

**[1]H NMR spectrum of 4do**



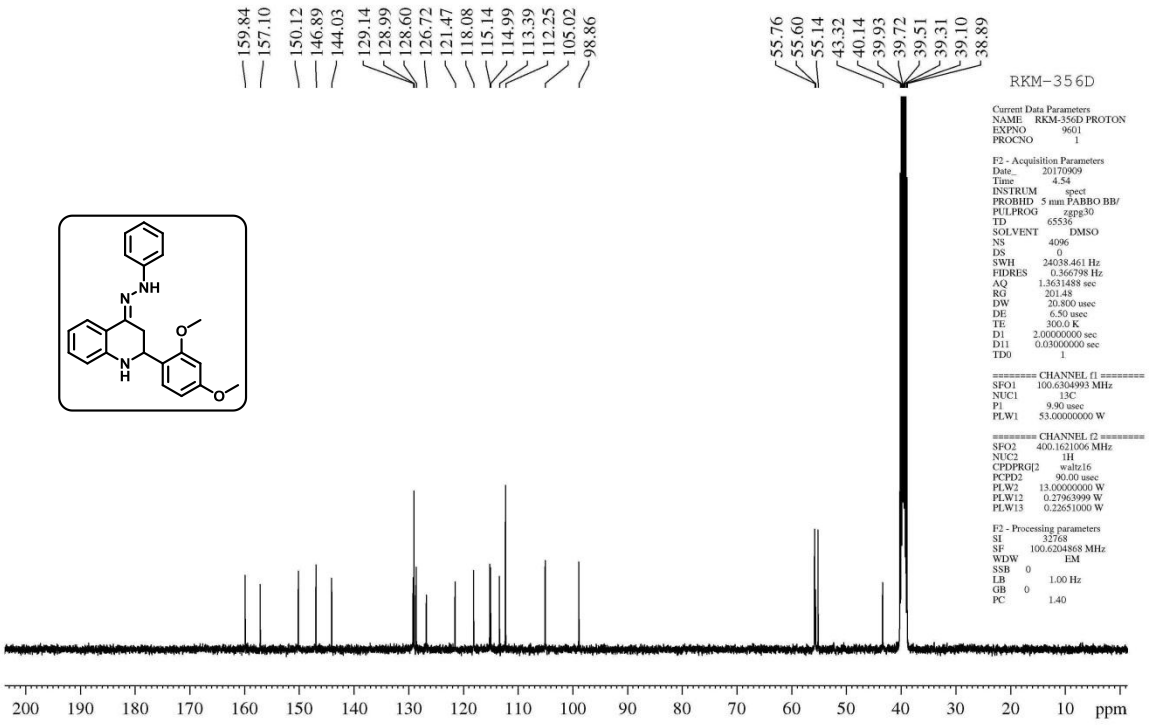

**[13]C NMR spectrum of 4do**



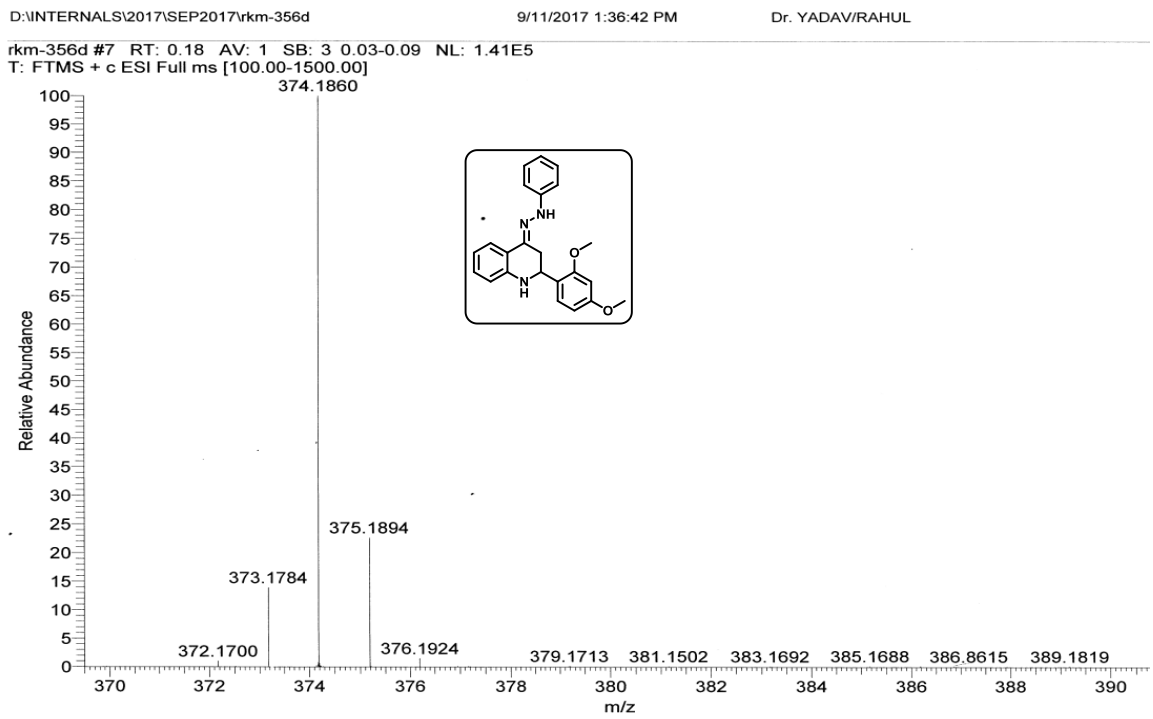

**HRMS spectrum of 4do**



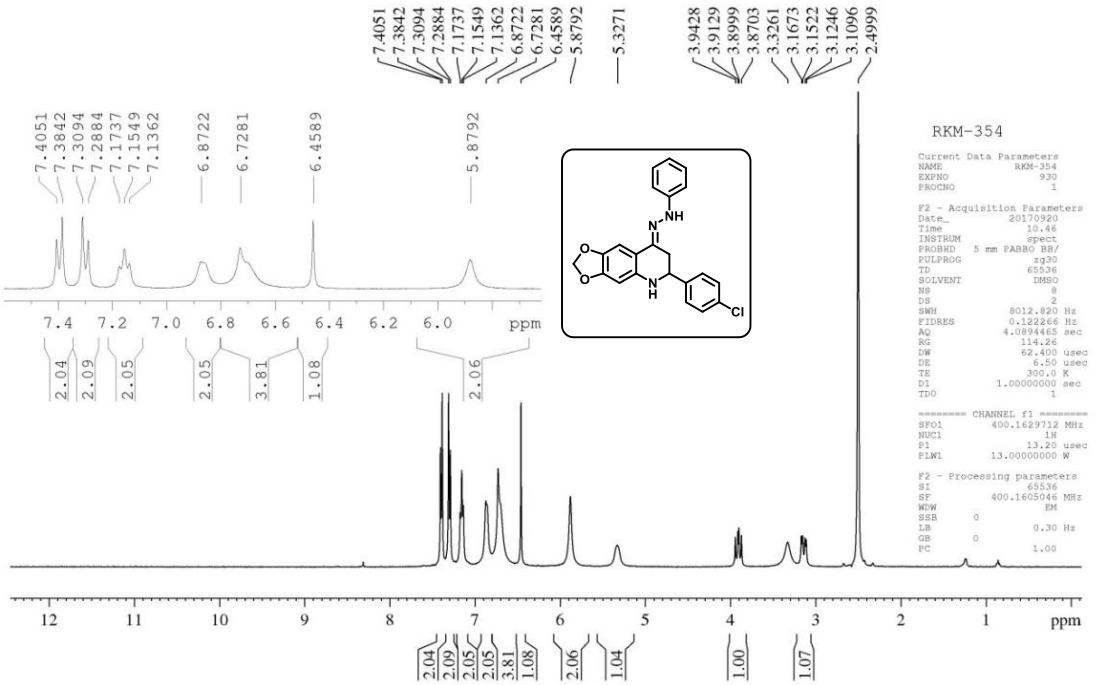

**$^{1}$H NMR spectrum of 4dp**



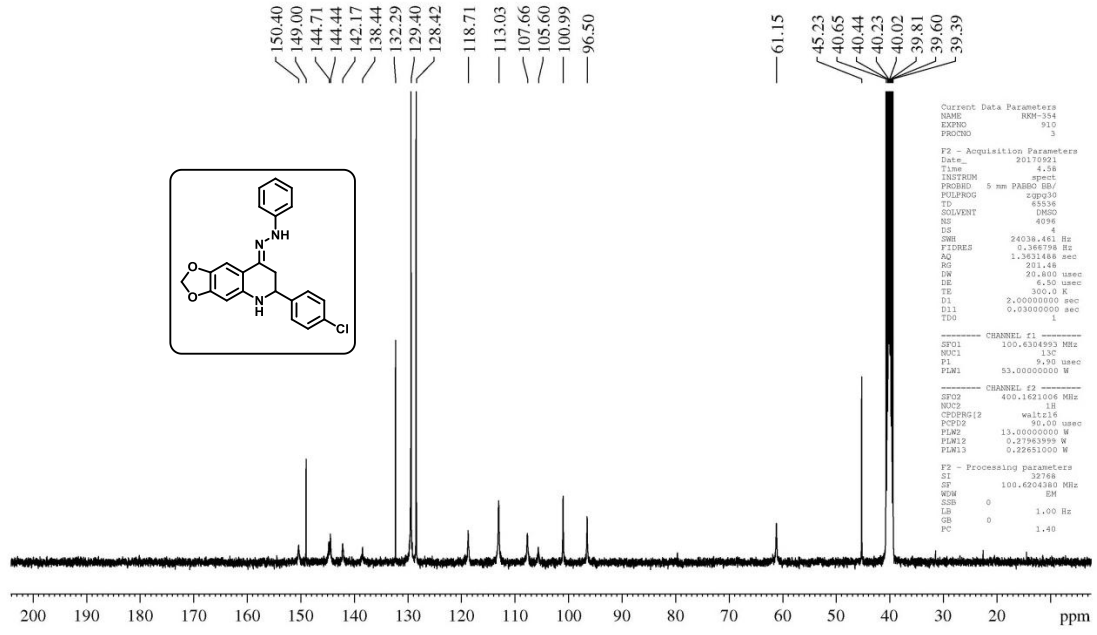

**$^{13}$C NMR spectrum of 4dp**



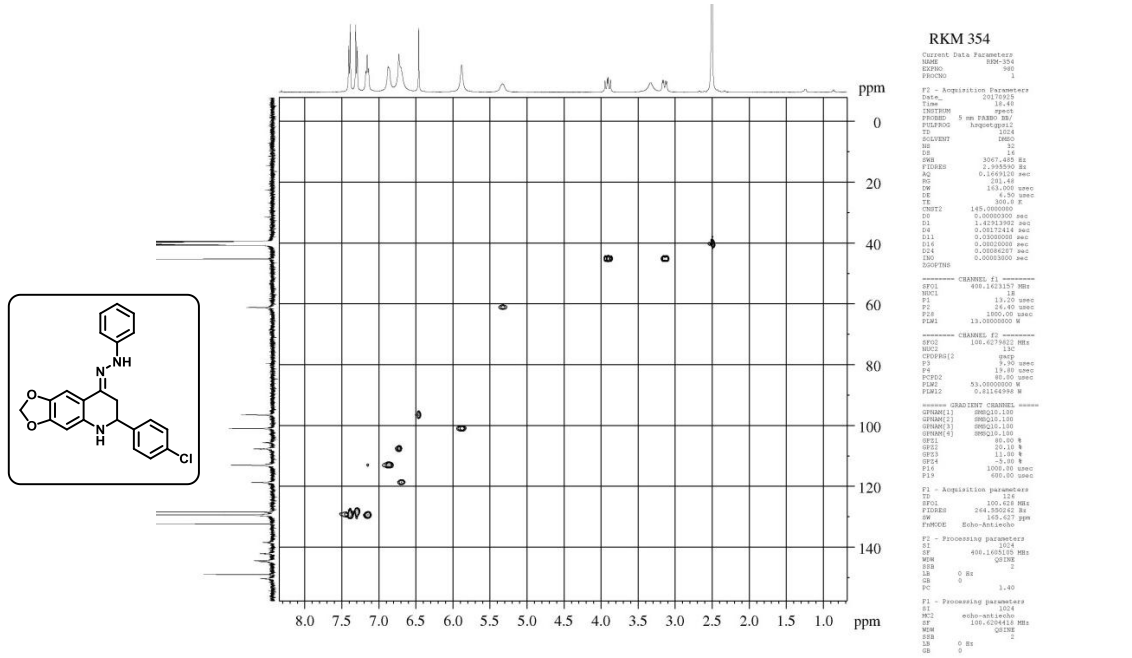

**HRMS spectrum of 4dp**



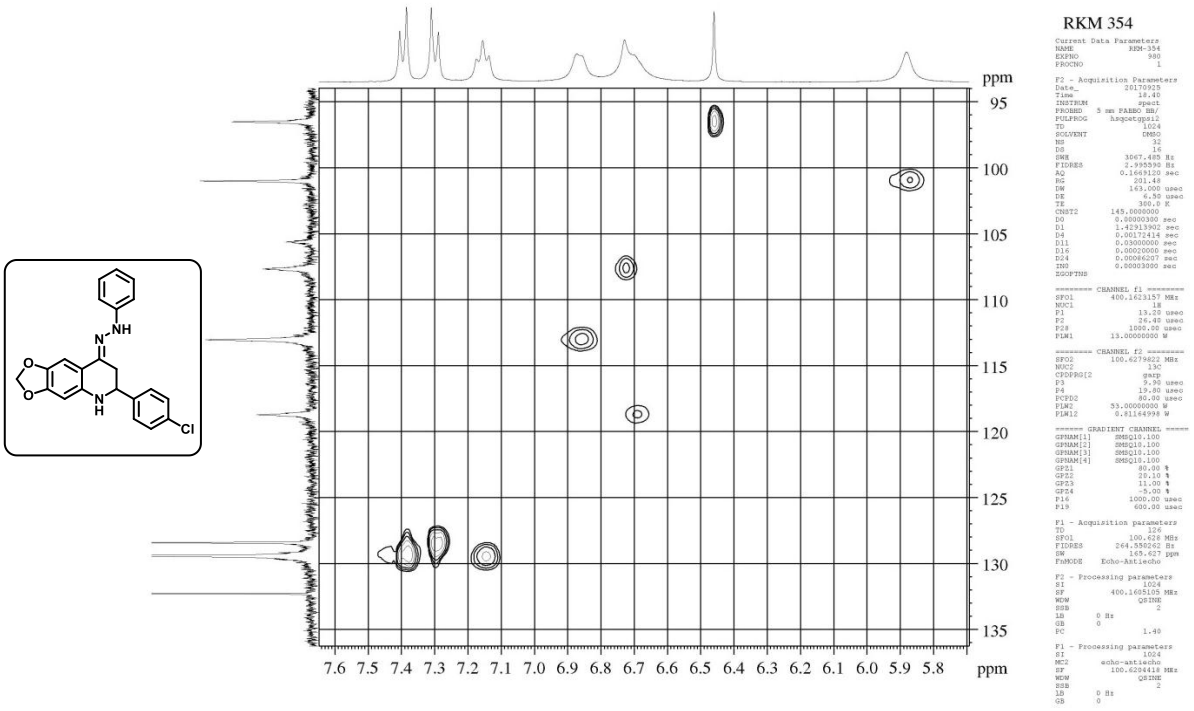

**$^{1}$H NMR spectrum of 4fc**



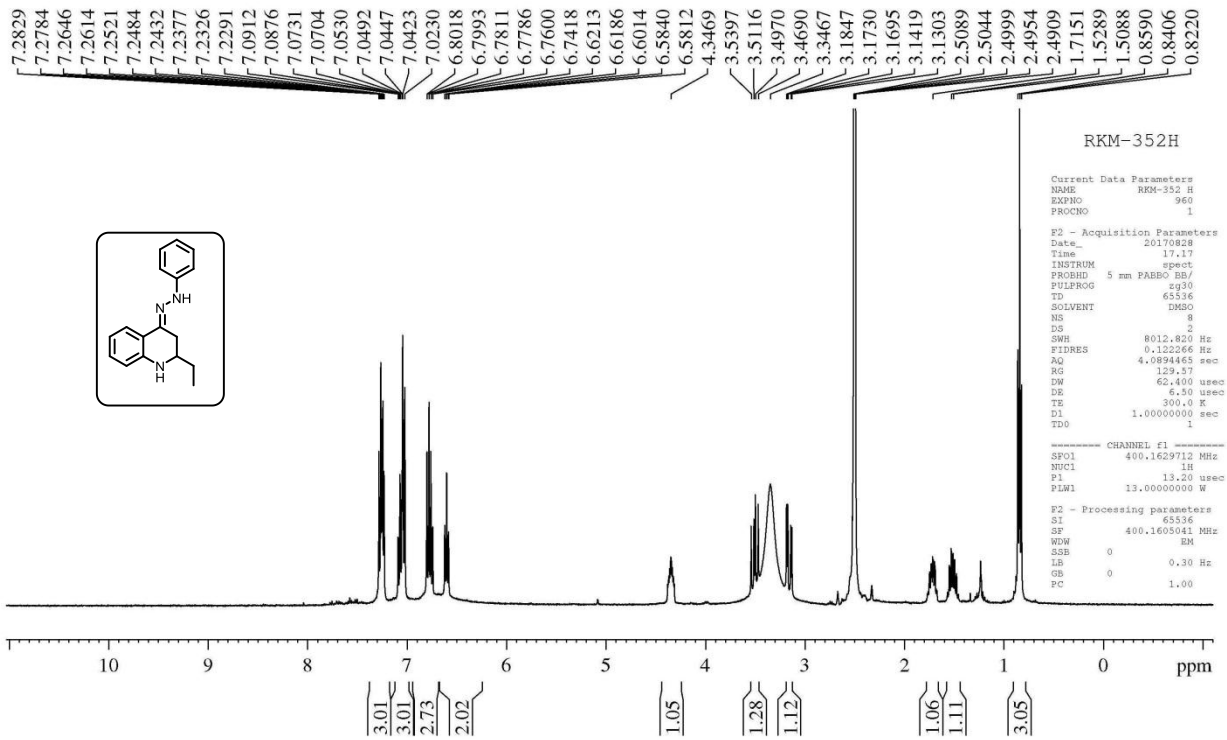

**$^{13}$C NMR spectrum of 4fc**



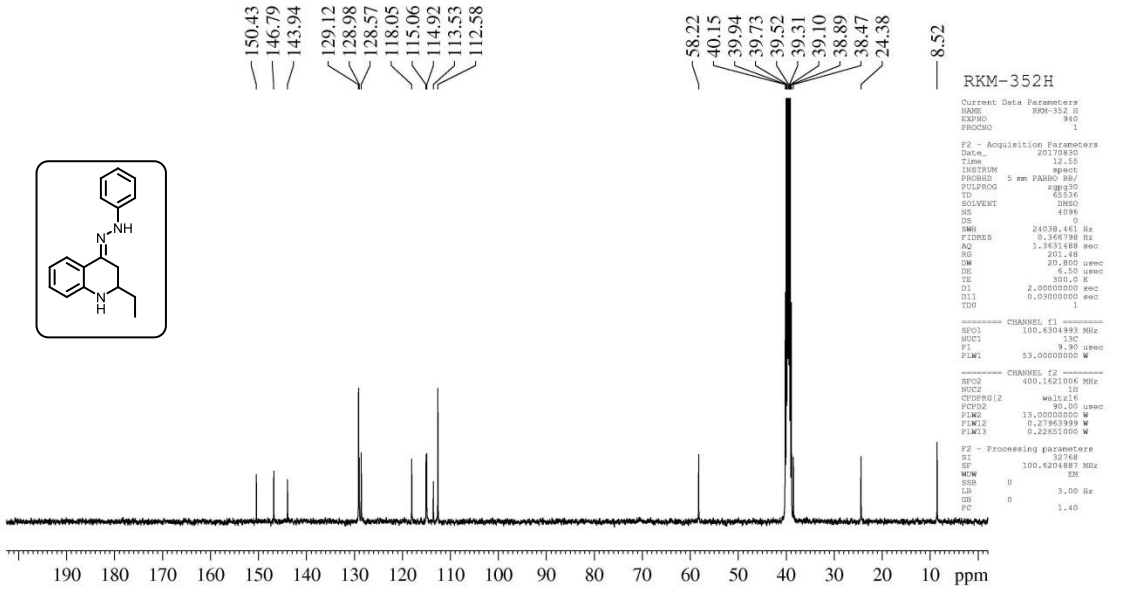

**HSQC spectrum of 4fc**



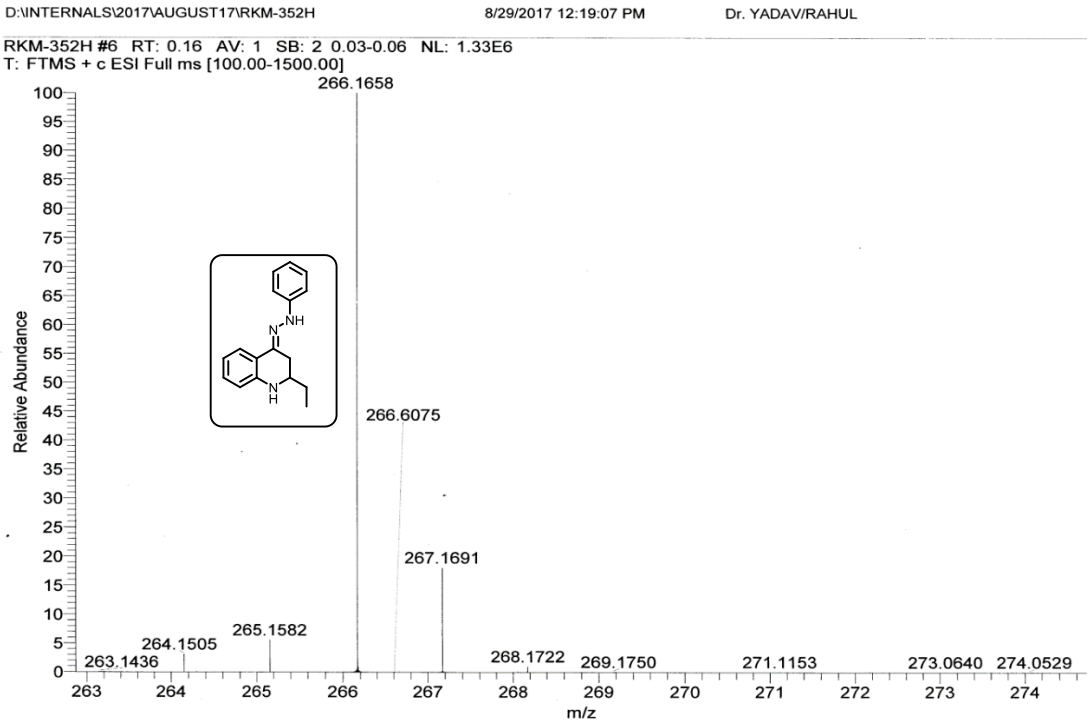

**Expanded HSQC spectrum of 4fc**



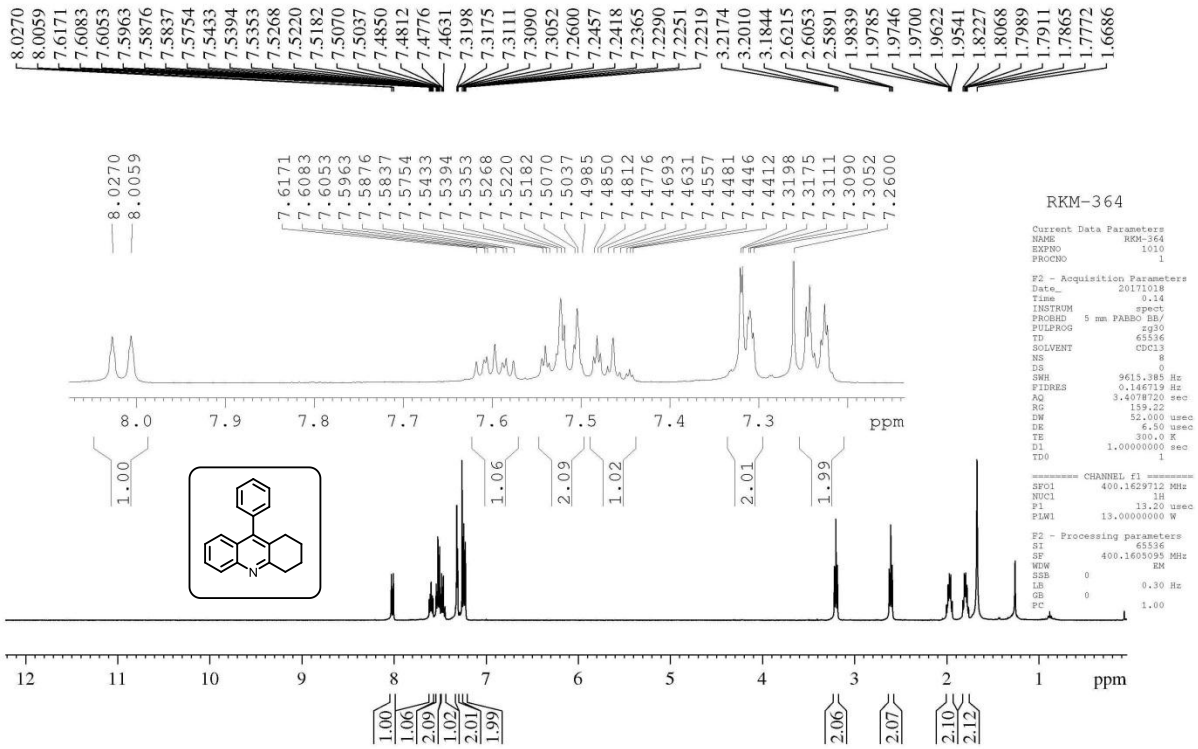

**$^{1}$H NMR spectrum of 4dw**



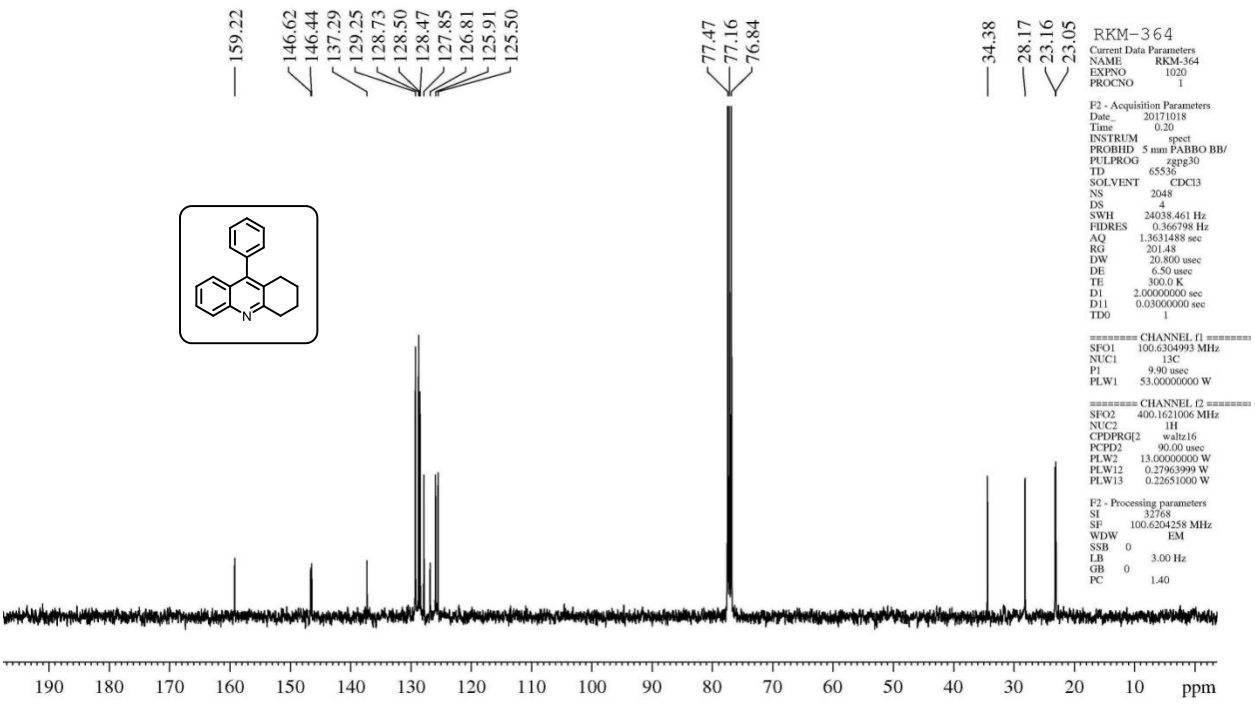

**$^{13}$C NMR spectrum of 4dw**



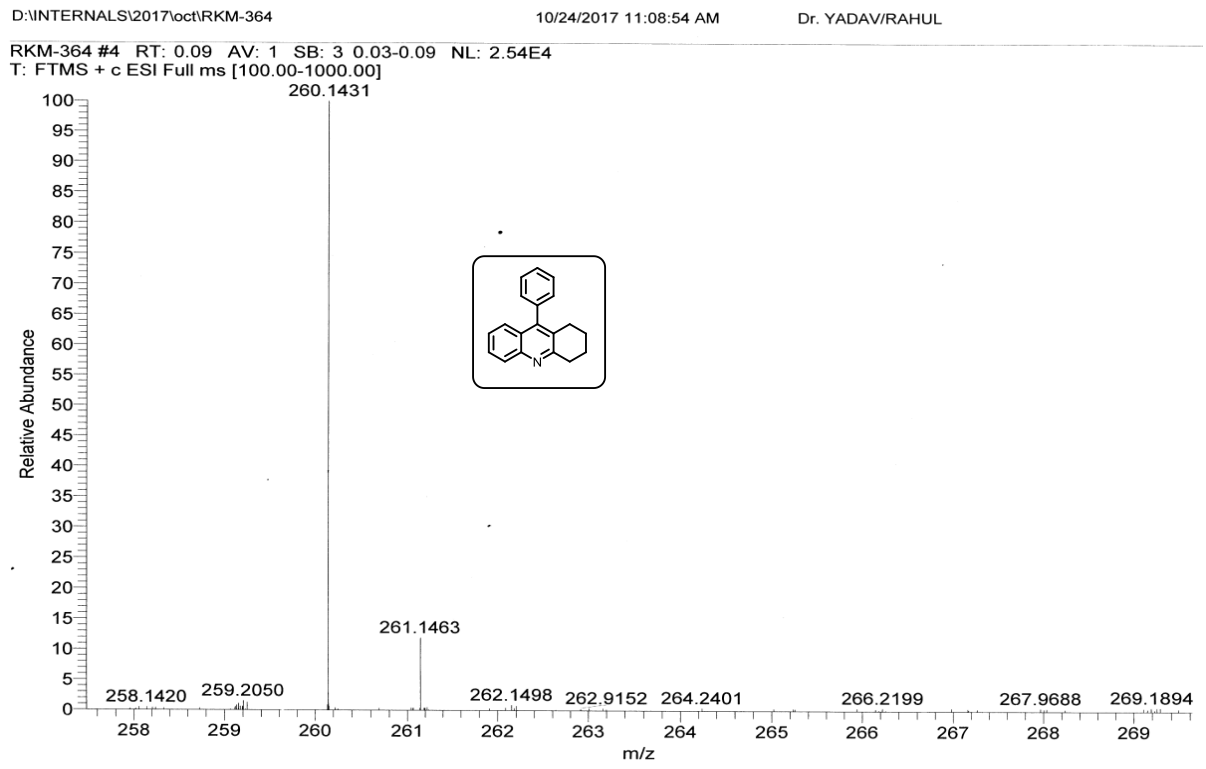

**HRMS spectrum of 4dw**



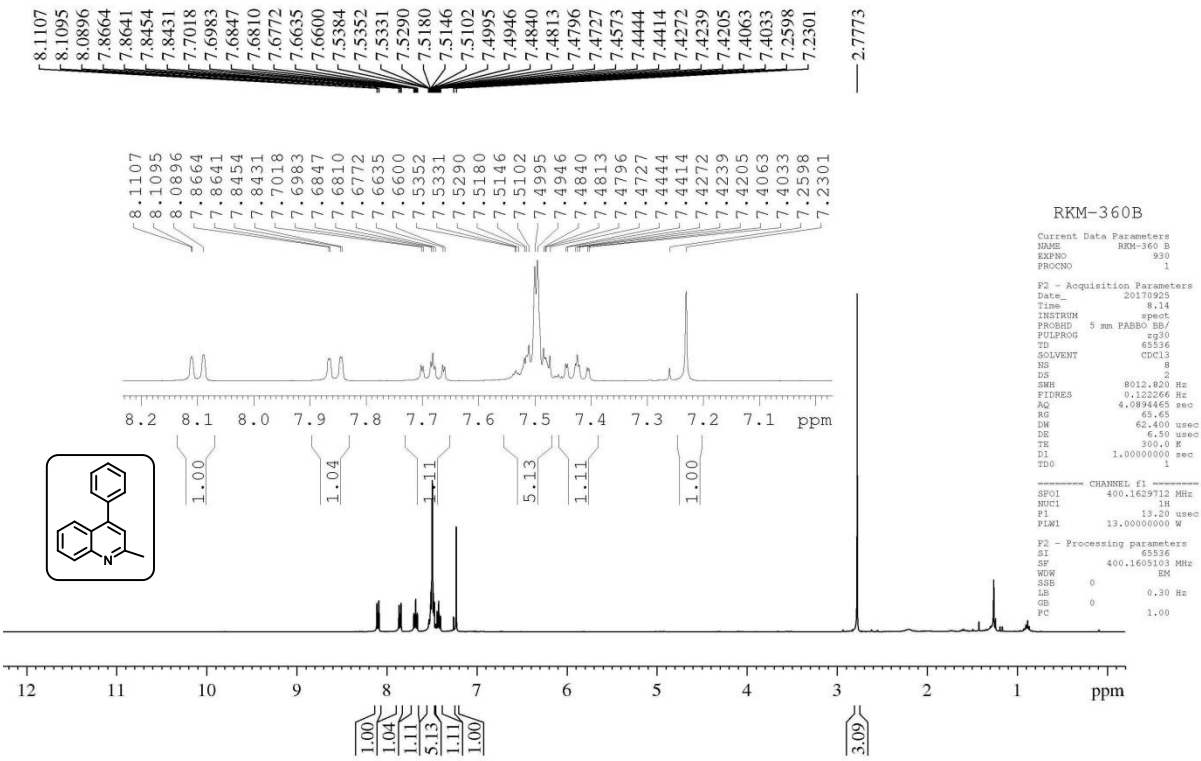

**$^{1}$H NMR spectrum of 6aa**



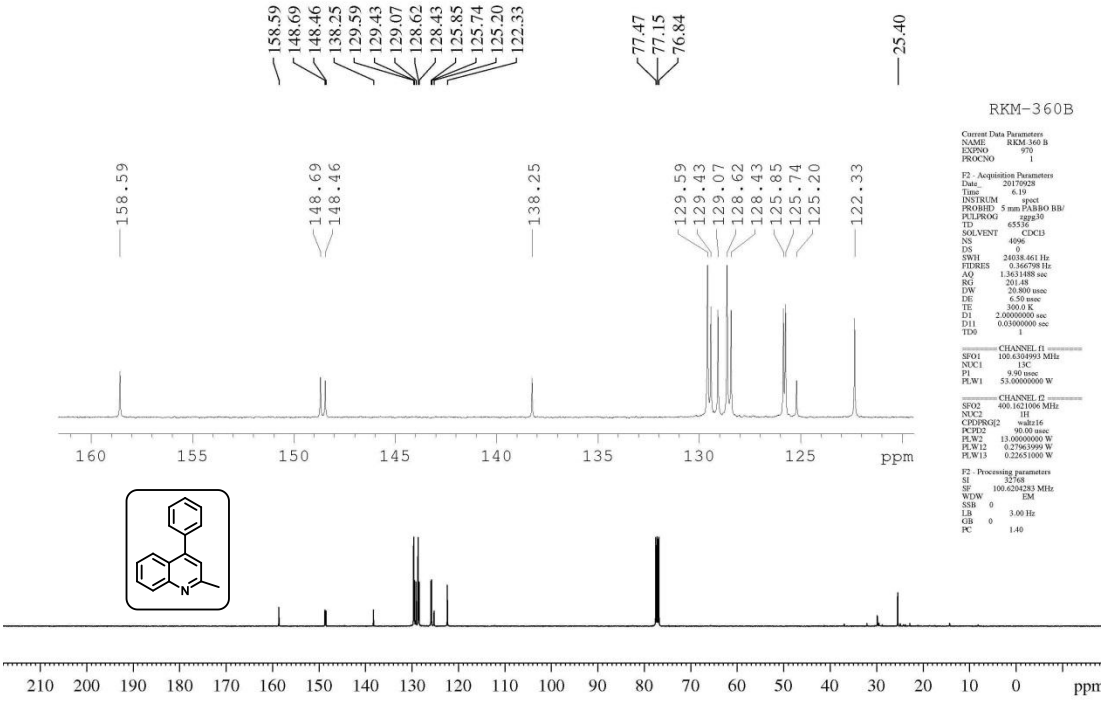

**$^{13}$C NMR spectrum of 6aa**



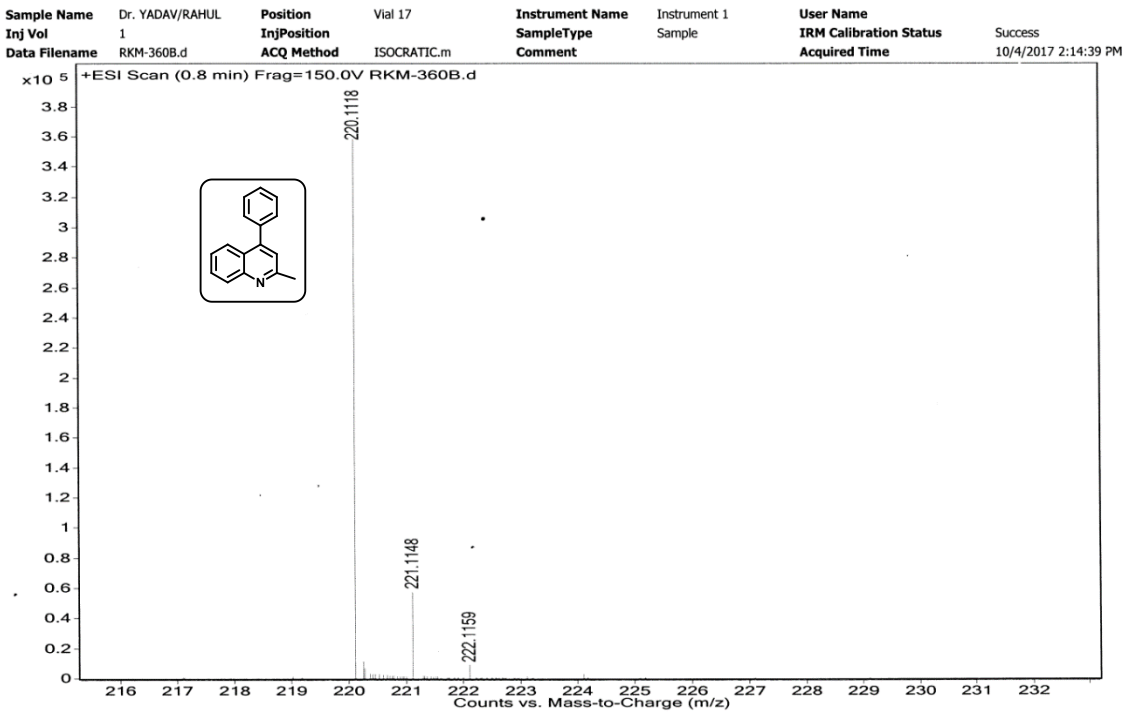

**HRMS spectrum of 6aa**



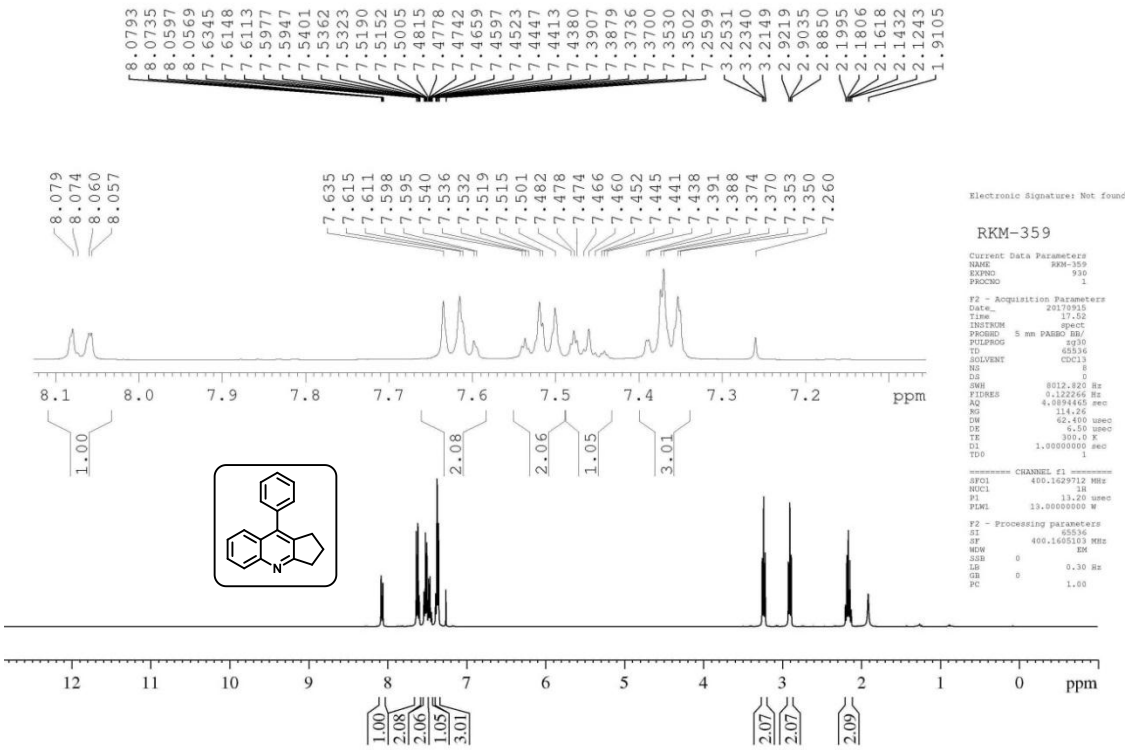

**[1]H NMR spectrum of 6ab**



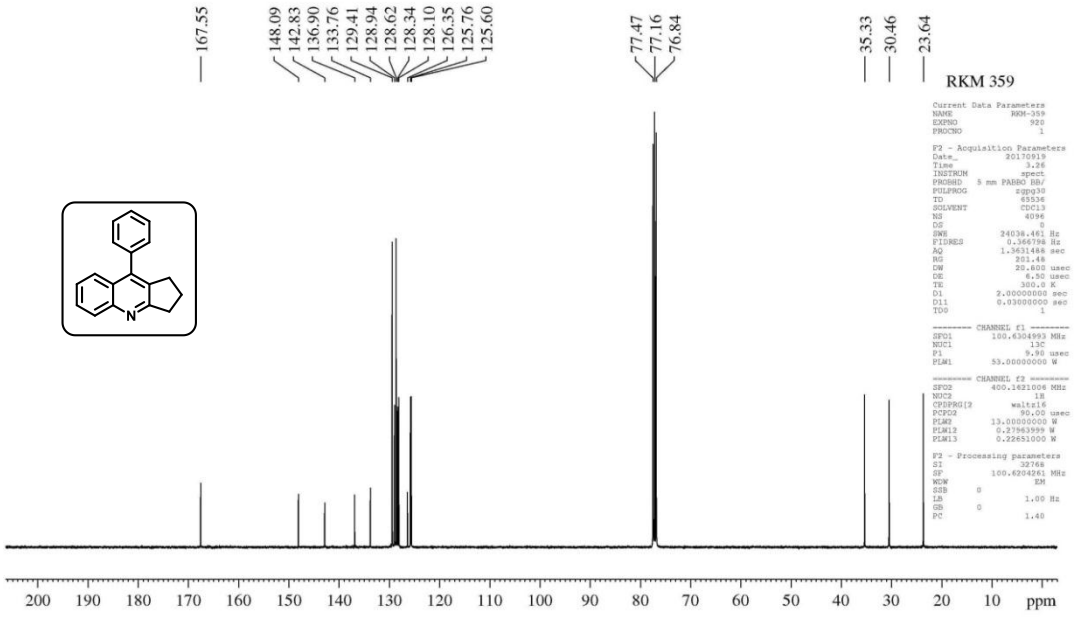

**[13]C NMR spectrum of 6ab**



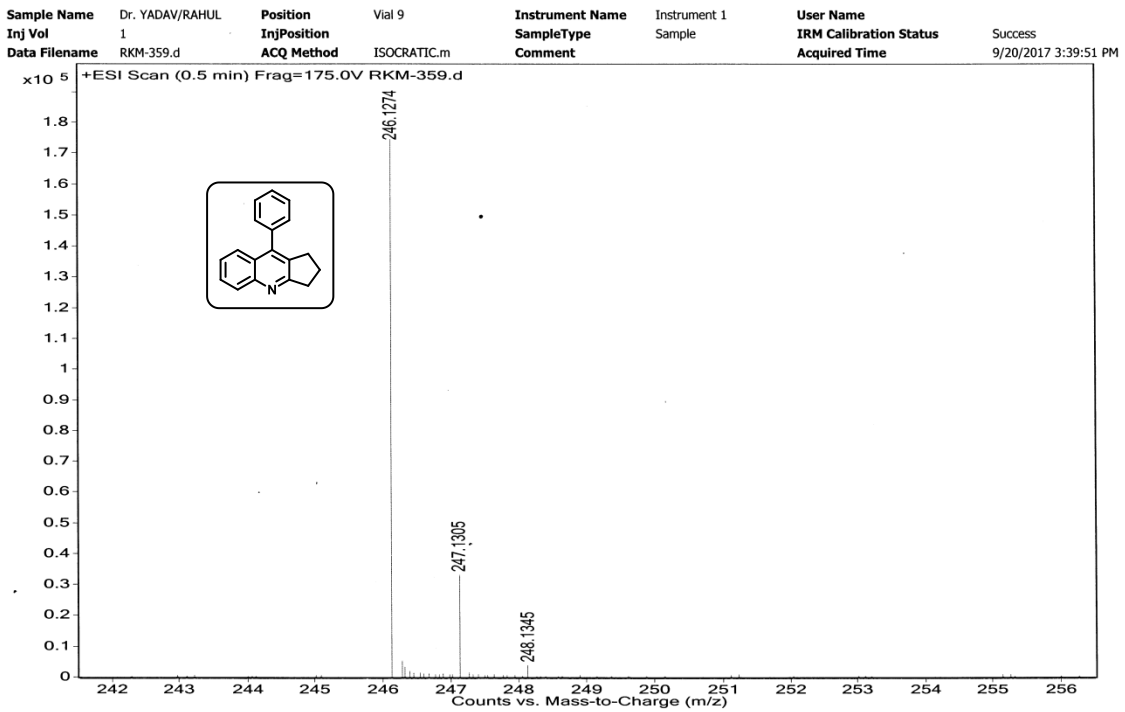

**HRMS spectrum of 6ab**



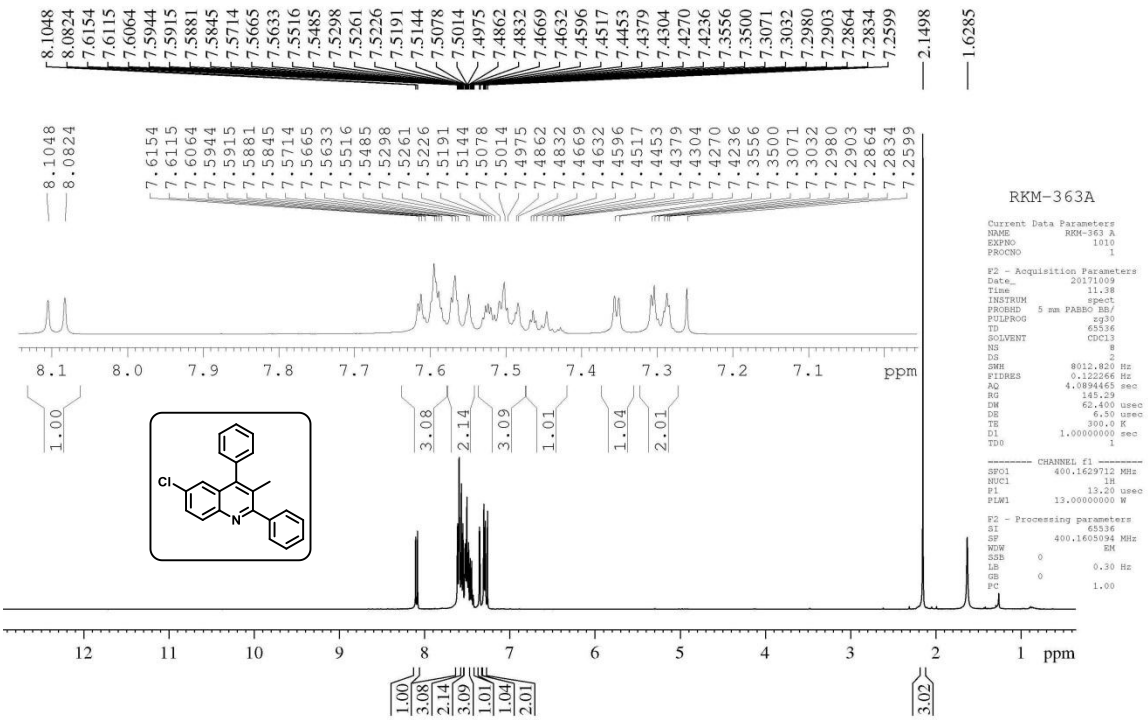

**$^{1}$H NMR spectrum of 6ac**



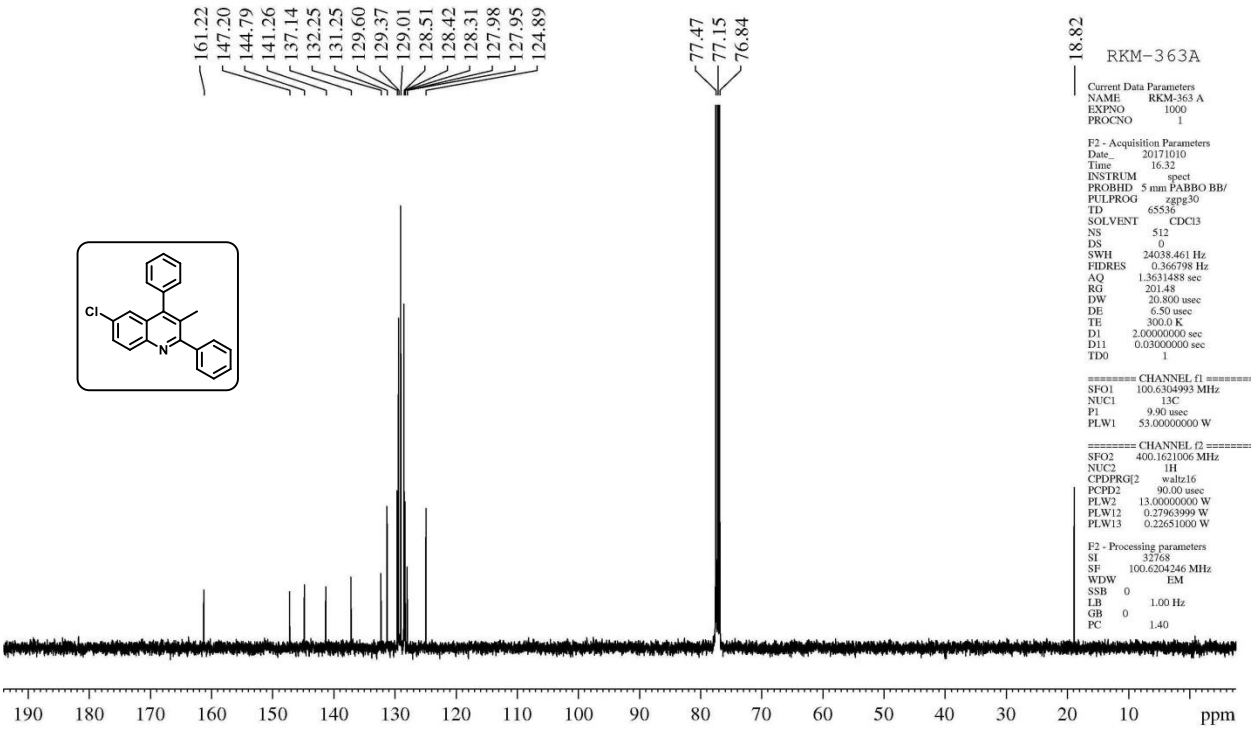

**$^{13}$C NMR spectrum of 6ac**



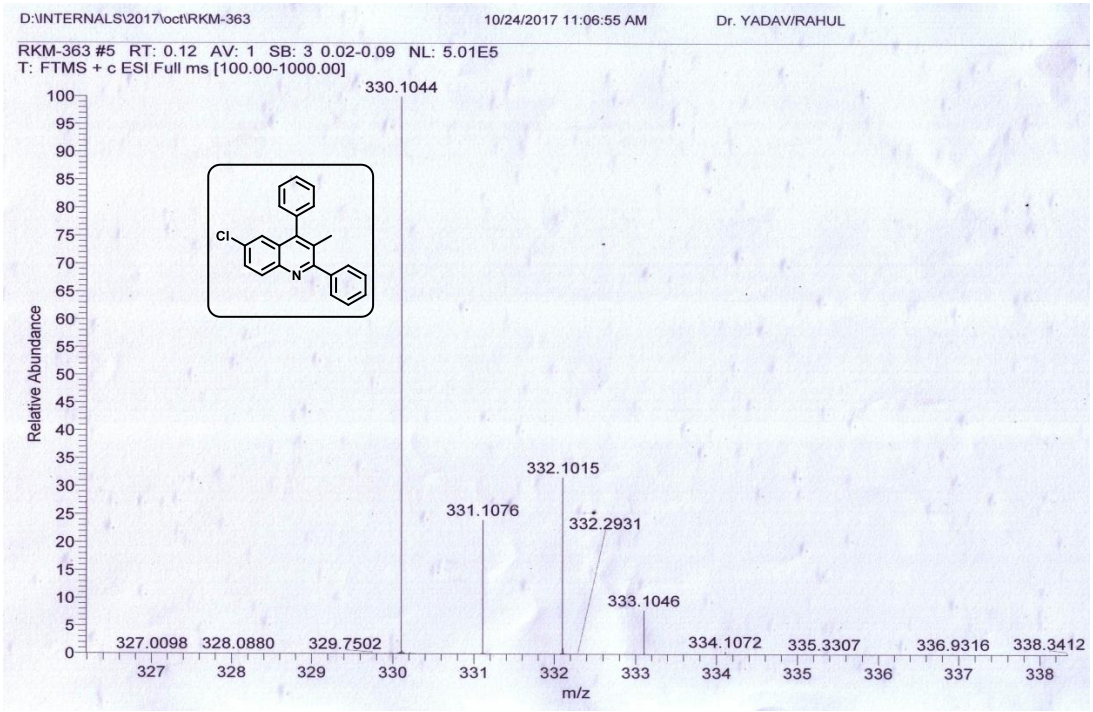

**HRMS spectrum of 6ac**



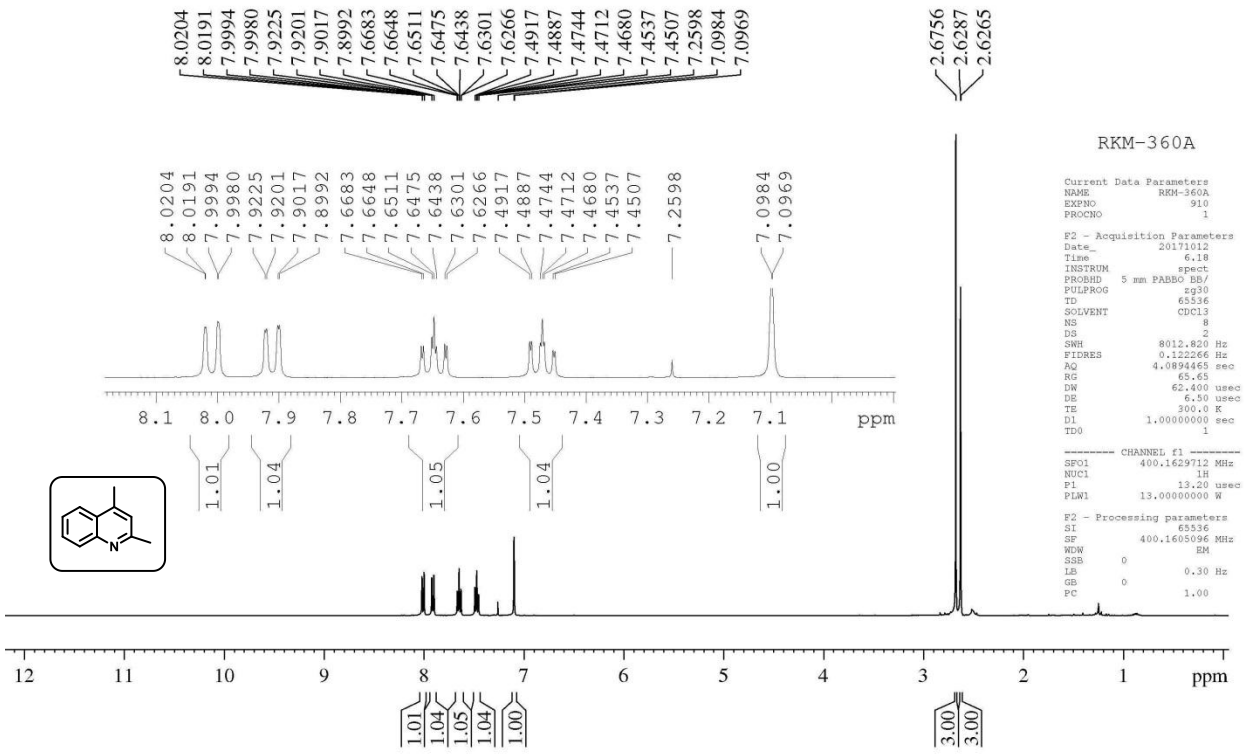

**$^{1}$H NMR spectrum of 6bd**



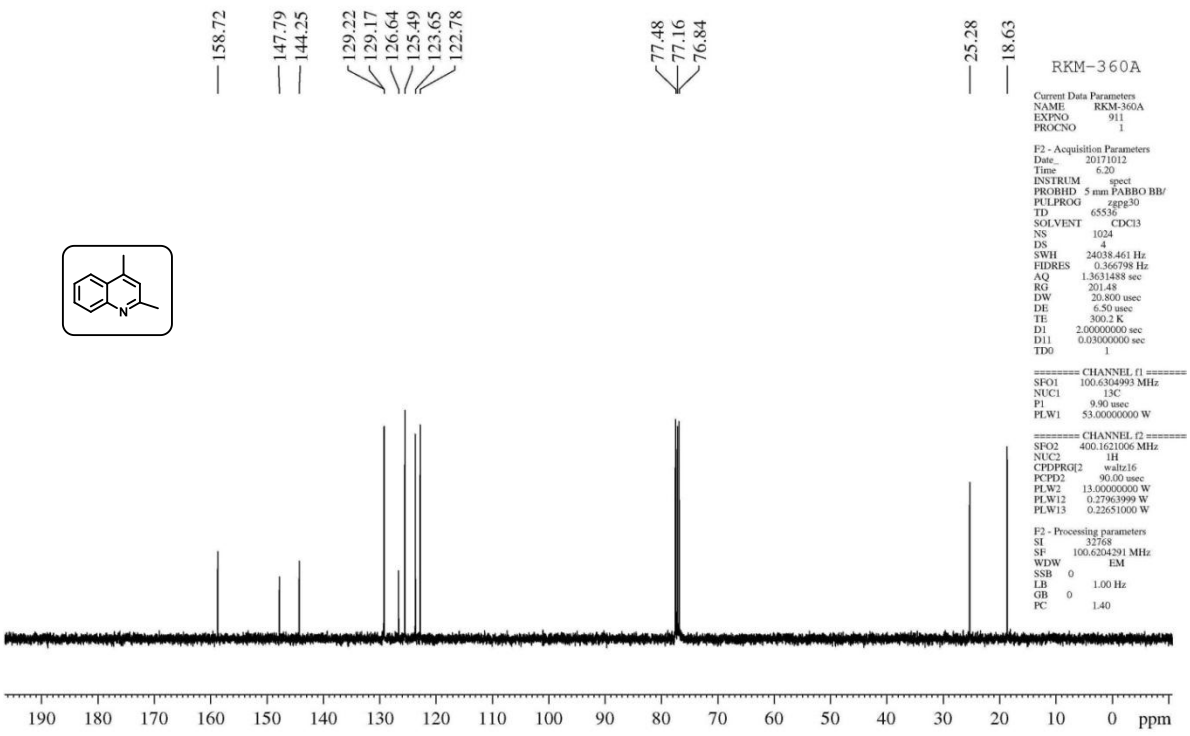

**$^{13}$C NMR spectrum of 6bd**



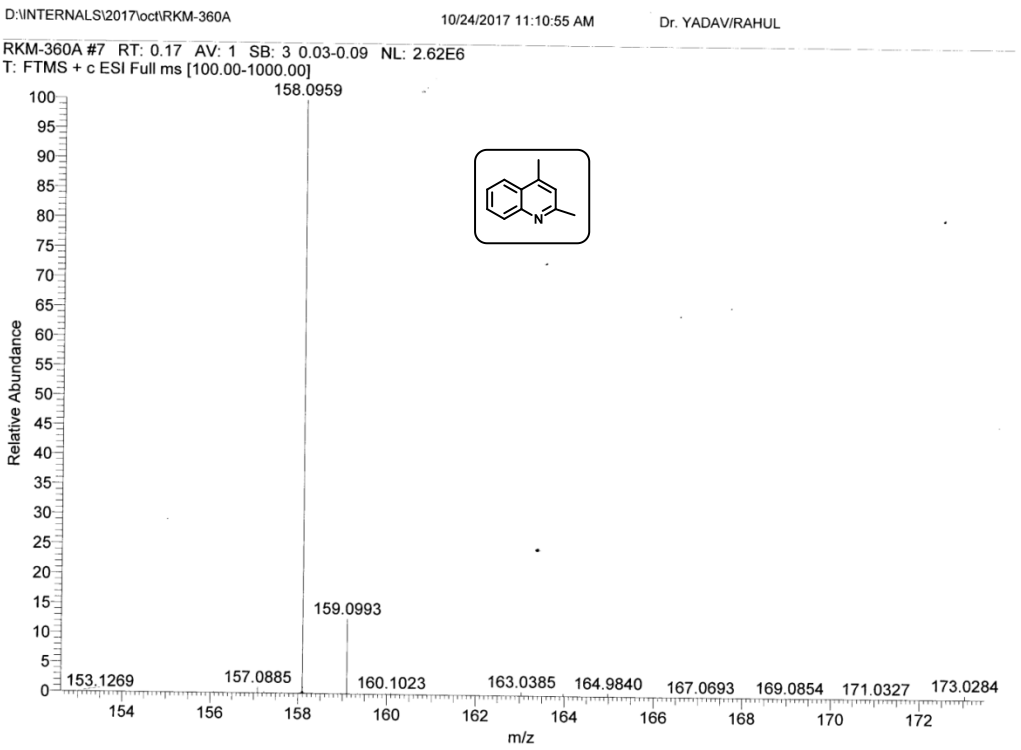

**HRMS spectrum of 6bd**



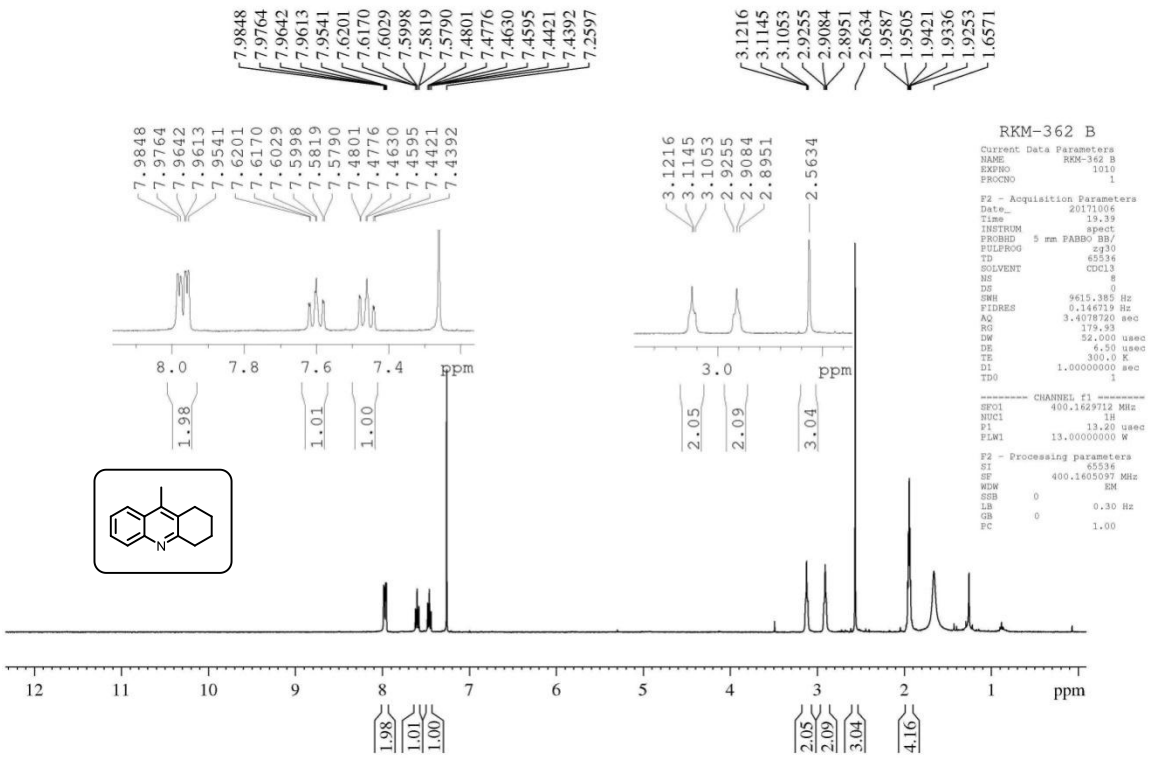

**$^{1}$H NMR spectrum of 6da**



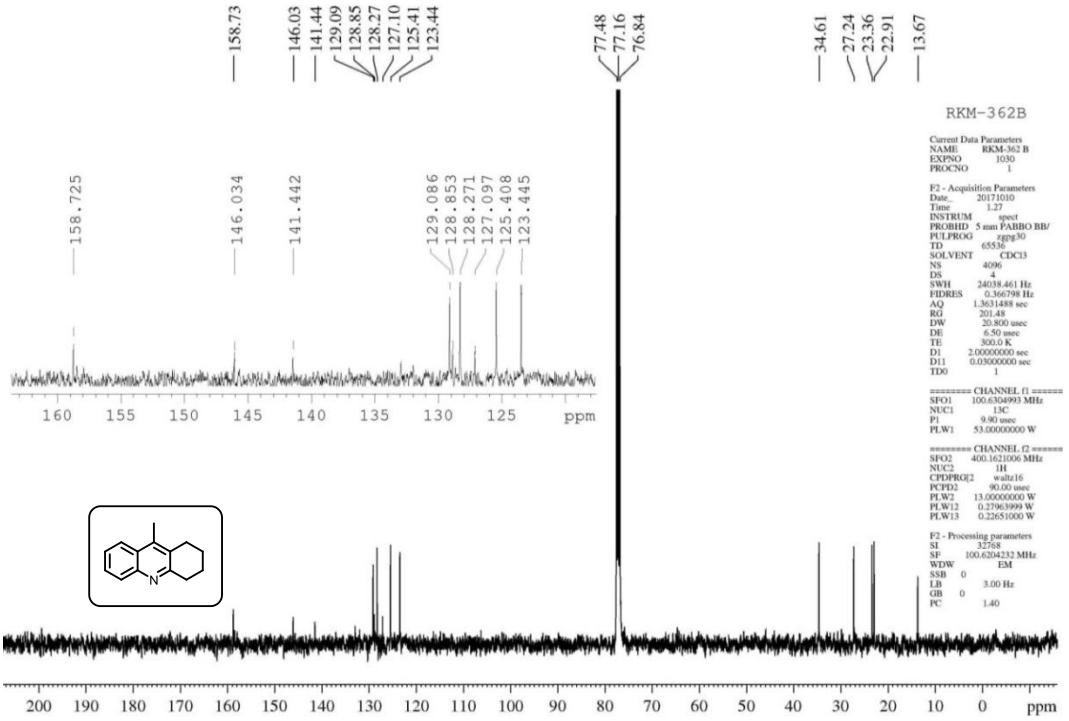

**$^{13}$C NMR spectrum of 6da**



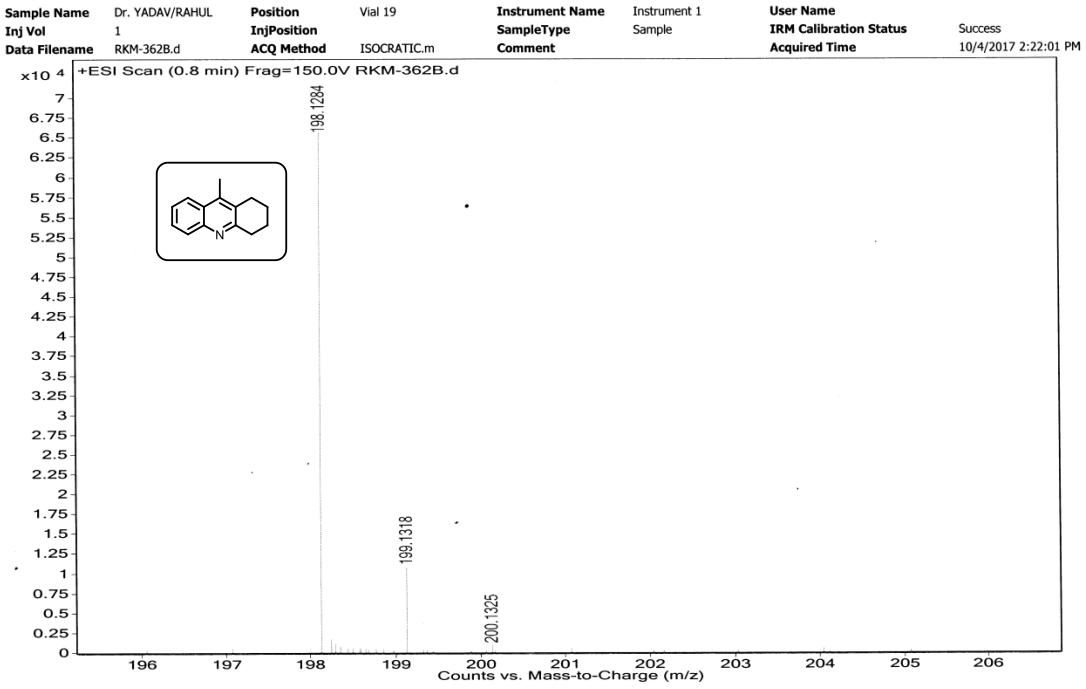

**HRMS spectrum of 6da**



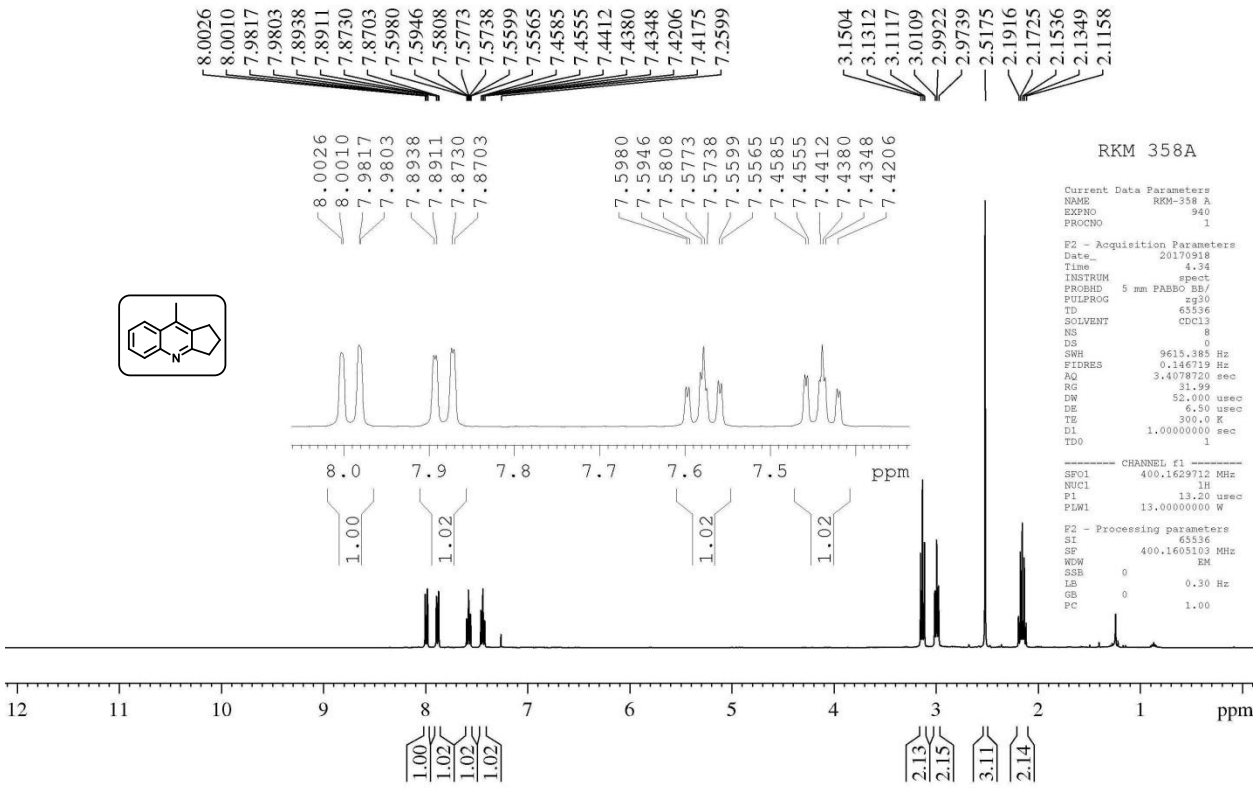

**$^{1}$H NMR spectrum of 6db**



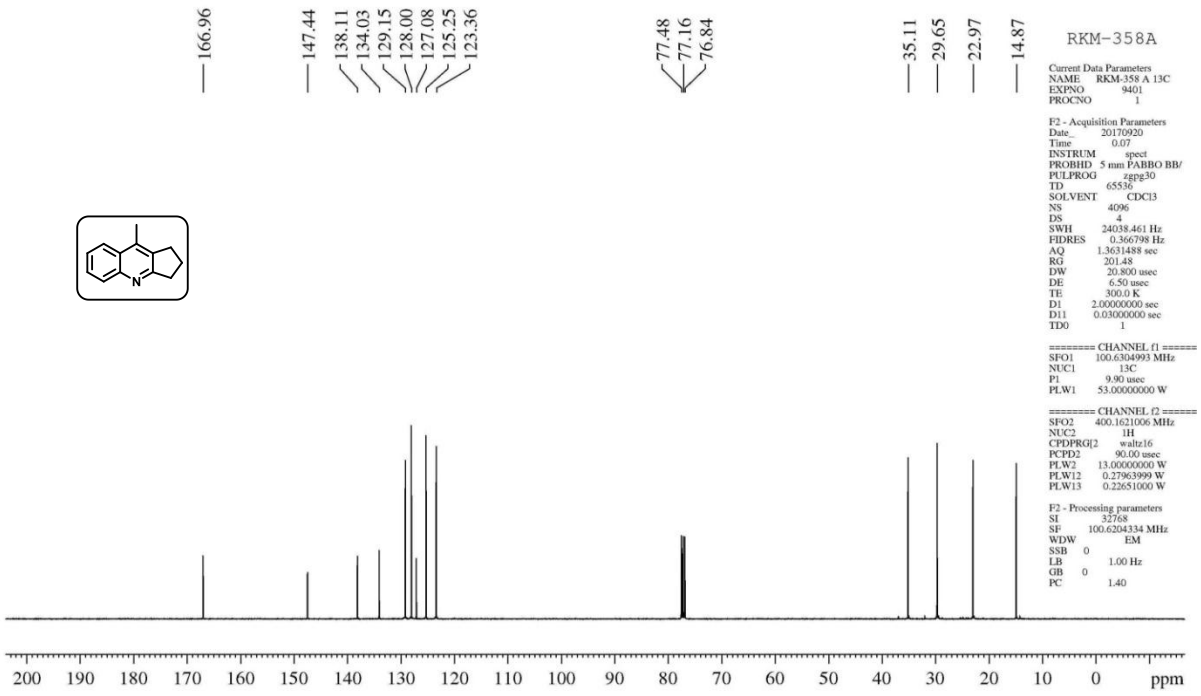

**$^{13}$C NMR spectrum of 6db**



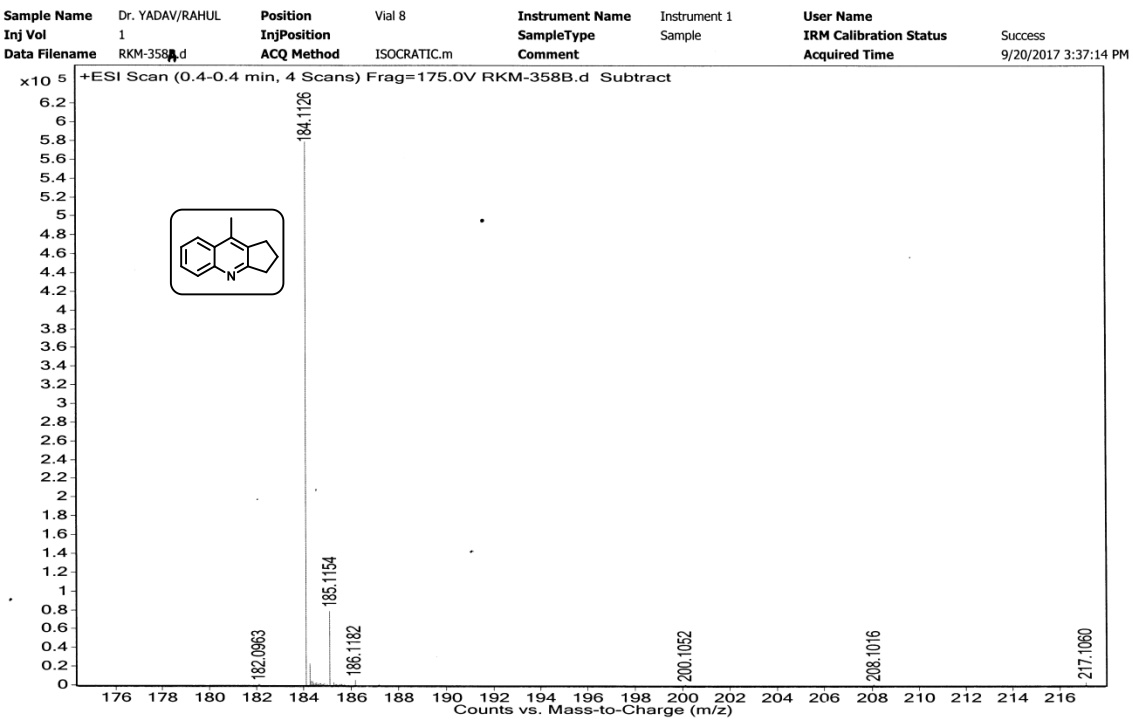

**HRMS spectrum of 6db**



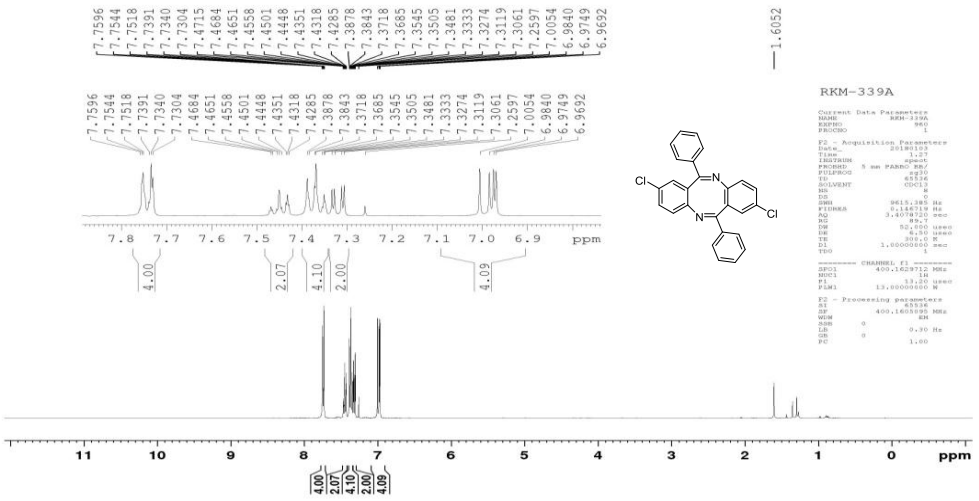

**$^{1}$H NMR spectrum of 6dc**



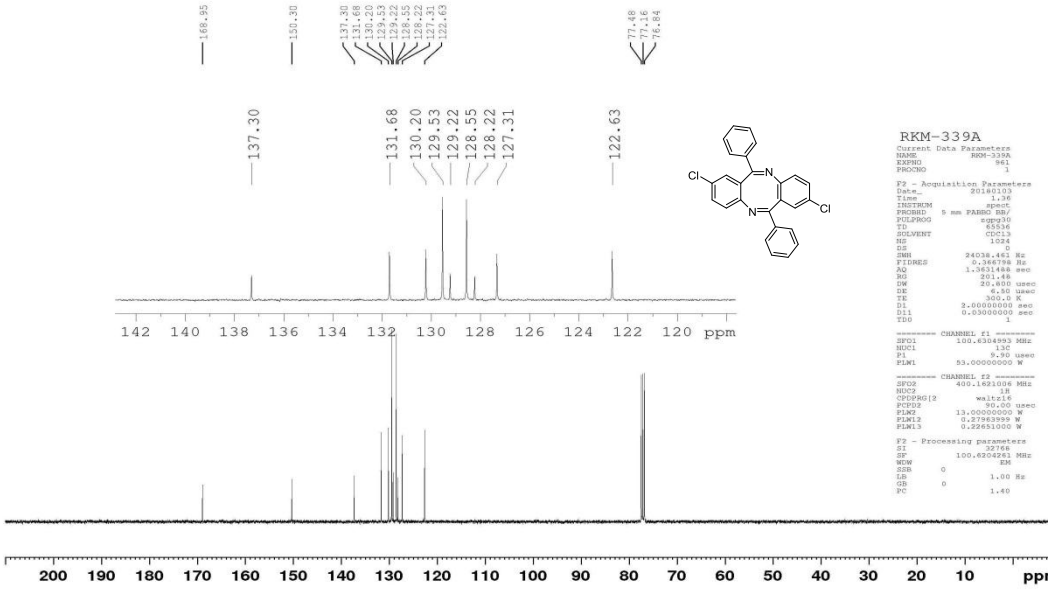

**$^{13}$C NMR spectrum of 6dc**



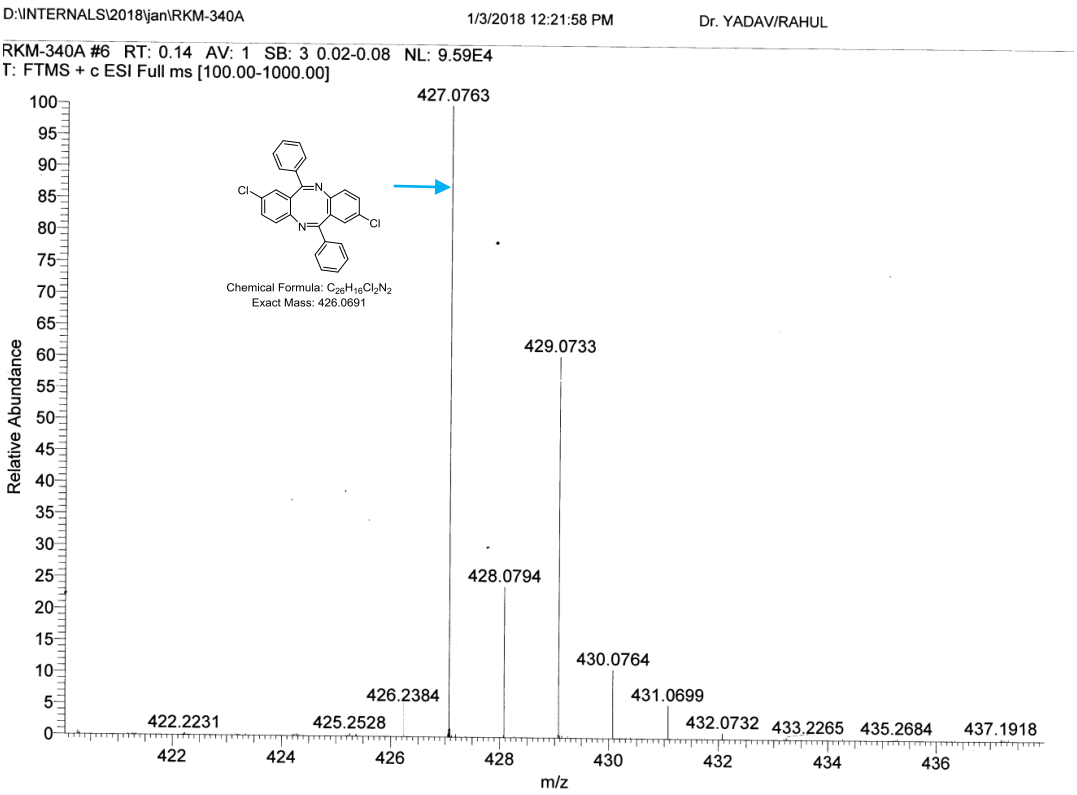

**HRMS spectrum of 6dc**



**$^{1}$H NMR Spectra of compound 7**

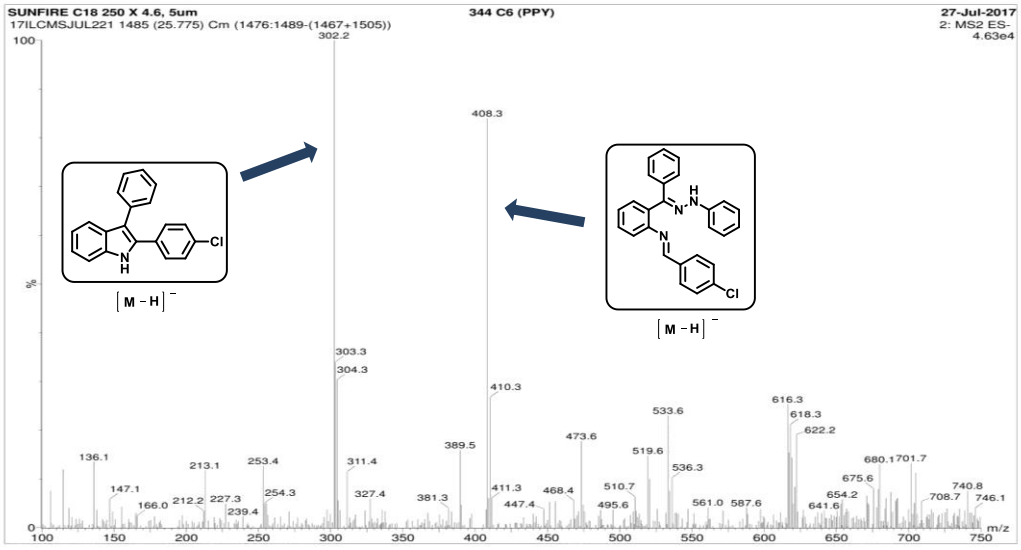


**$^{13}$C NMR Spectra of compound 7**

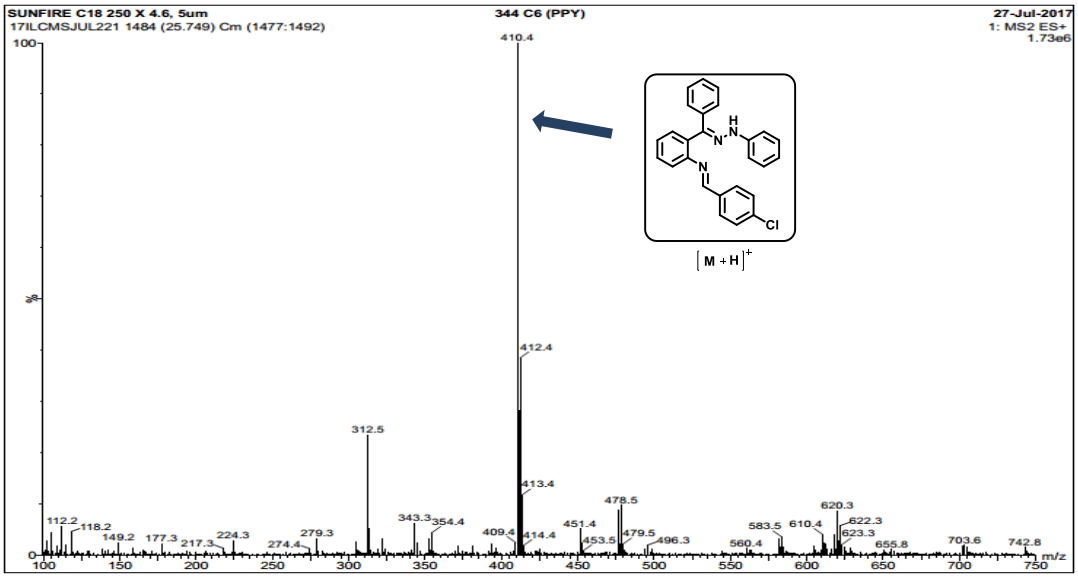



**HRMS Spectra of compound 7**

## 5. LC-ESIMS of crude reaction of Compound 3ac

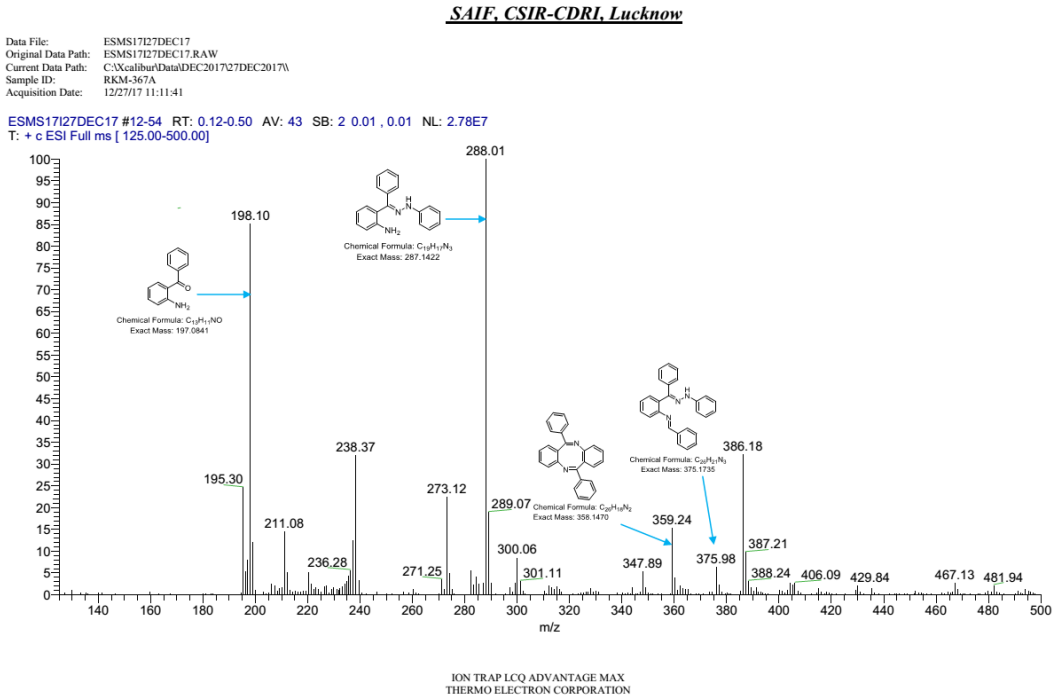



**LC-ESI-MS spectrum (-ve mode) of Crude Reaction mixture (8 h) of Compound 3ac**




**LC-ESI-MS spectrum (+ve mode) of Crude Reaction mixture (8 h) of Compound 3ac**

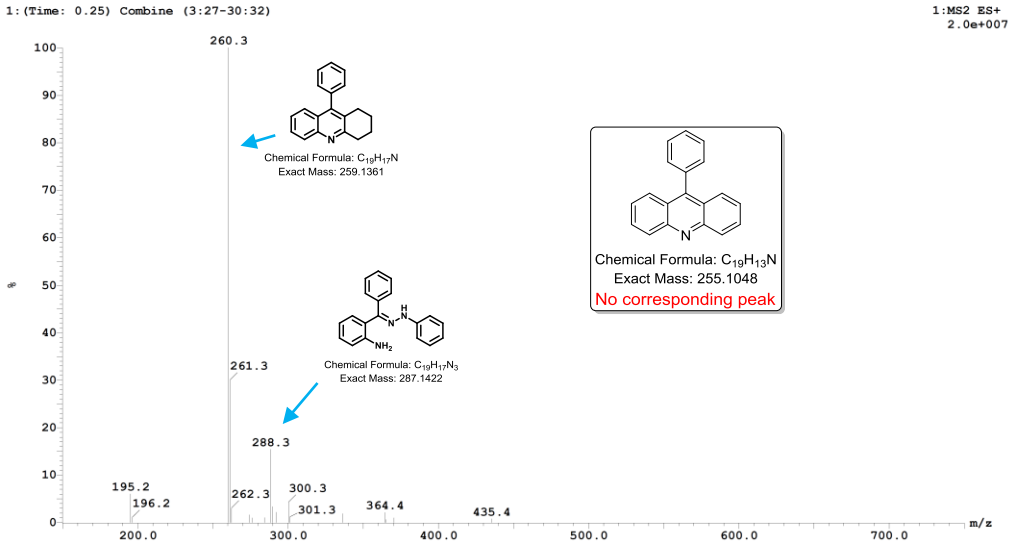
## 6. ESI-MS of crude reaction mixtures of 3aa and 6aa




**ESI-MS of crude reaction mixture for the synthesis of 3aa in one pot *via* successive addition**

**Openlynx Report SAIF, CSIR-CDRI, Lucknow** Page 1
Sample: 280 Vial:1:27 ID:ESMS17I12OCT27
File:ESMS17I12OCT27 Date:12-Oct-2017 Time:12:34:35
Description:RKM-364

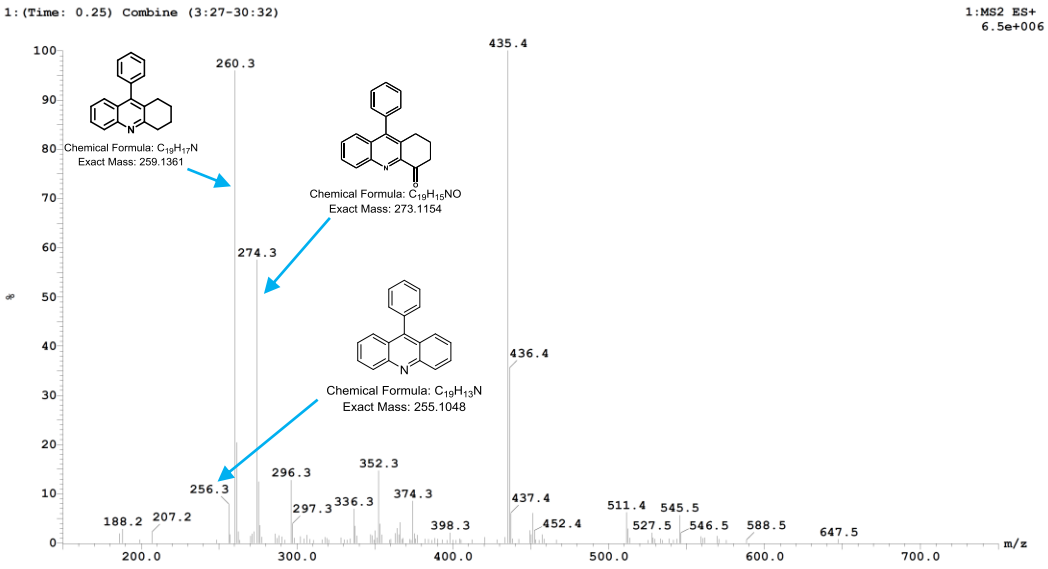
Printed: Thu Oct 12 14:13:38 2017




**ESI-MS of crude reaction mixture of 6aa (reaction at RT)**

**Openlynx Report SAIF, CSIR-CDRI, Lucknow** Page 1
Sample: 227 Vial:1:10 ID:ESMS17I11OCT10
File:ESMS17I11OCT10 Date:11-Oct-2017 Time:12:45:35
Description:RKM-364

Printed: Wed Oct 11 15:41:54 2017




**ESI-MS of crude reaction mixture of 6aa (reaction at 120 $^{0}$C)**