

## Supporting Information

### Substrate-Controlled Rh(III)-catalyzed Regiodivergent Annulation to Fused and Spiro Benzimidazoles

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## General Methods

$^1\text{H}$  NMR (400 MHz and 600 Hz) and  $^{13}\text{C}$  NMR (101 MHz and 151 Hz) spectra were recorded on 400 and 600 MHz automated spectrometers. Chemical shifts are reported in parts per million (ppm) on the  $\delta$  scale from an internal standard (TMS). Analytical thin-layer chromatography (TLC) was performed using 0.25 mm silica gel-coated Kieselgel 60 F<sub>254</sub> plates. Flash chromatography was performed using the indicated solvent and silica gel 60 (Merck, 230-400 mesh). Melting points were recorded on a Fargo melting point apparatus and are uncorrected. High-resolution mass spectra (HRMS) were recorded in ESI mode using TOF mass spectrometer. IR spectra were recorded using a Bruker spectrophotometer. *N*-aryl benzimidazoles **1** and maleimides **2** were prepared according to the reported procedures.<sup>1,2</sup> All materials were purchased from commercial sources and used without further purification.

### A representative procedure for the synthesis of **3a**

To an oven-dried sealed tube was added 2-phenyl benzimidazole **1a** (0.1 g, 0.515 mmol), *N*-methylmaleimide **2a** (0.11 g, 1.03 mmol), [RhCp\*Cl<sub>2</sub>]<sub>2</sub> (0.0079 g, 0.012 mmol), AgOAc (0.27 g, 1.03 mmol) and 1,2-dichloroethane (3 mL). The reaction mixture was heated at 100 °C for 5 h. The reaction mixture was cooled down at room temperature and filtered through celite bed. The filtrate was concentrated under reduced pressure. The crude reaction mixture was purified by column chromatography (20% ethyl acetate/*n*-hexanes) to obtain 2-methyl-3*a*,13*a*-dihydro-1*H*-benzo[4,5]imidazo[2,1-*a*]pyrrolo[3,4-*c*]isoquinoline-1,3(2*H*)-dione **3a**.

Pale yellow solid, 0.212 g, 90%; mp 239-241 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 8.42 (d, *J* = 8.7 Hz, 1H), 7.92 (d, *J* = 8.1 Hz, 1H), 7.79 (dd, *J* = 19.6, 7.6 Hz, 2H), 7.50 (d, *J* = 4.0 Hz, 2H), 7.35 (s, 2H), 5.65 (d, *J* = 9.7 Hz, 1H), 4.68 (d, *J* = 9.7 Hz, 1H), 3.07 (s, 3H); <sup>13</sup>C NMR (101 MHz, CDCl<sub>3</sub>): δ 172.3, 145.5, 143.8, 134.9, 130.9, 129.2, 128.2, 126.2, 125.8, 124.4, 123.7, 123.5, 119.8, 111.1, 53.1, 41.6, 25.6; HRMS (ESI, *m/z*) calculated for C<sub>18</sub>H<sub>14</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 304.1086, found 304.1084.

### A representative procedure for the synthesis of **5aa**

To an oven-dried sealed tube was added 2-(*o*-tolyl)-benzimidazole **1h** (0.1 g, 0.481 mmol), *N*-methylmaleimide **2a** (0.1 g, 0.962 mmol), [RhCp\*Cl<sub>2</sub>]<sub>2</sub> (0.0074 g, 0.012 mmol), AgOAc (0.15 g, 0.962 mmol), AcOH (0.14 g, 2.405 mmol) and acetonitrile (3 mL). The reaction mixture was heated at 100 °C for 5 h. The reaction mixture was cooled down at room temperature and filtered through celite bed. The filtrate was concentrated under reduced pressure. The crude reaction mixture was purified by column chromatography (25% ethyl

acetate/n-hexanes) to obtain 1',4-dimethylspiro [benzo [4,5] imidazo [2,1-a] isoindole-11,3'-pyrrolidine]-2',5'-dione **5aa**.

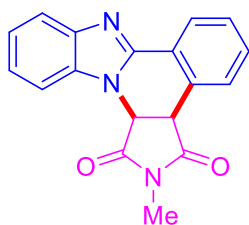
Pale yellow solid, 0.194 g, 65%; mp 261-263 °C; <sup>1</sup>H NMR (400 MHz, acetone-*d*<sub>6</sub>): δ 7.86 – 7.70 (m, 1H), 7.55 (d, J = 6.4 Hz, 1H), 7.52 – 7.39 (m, 3H), 7.38 – 7.19 (m, 2H), 3.75 (d, J = 18.5 Hz, 1H), 3.54 (d, J = 18.5 Hz, 1H), 3.18 (s, 3H); <sup>13</sup>C NMR (100 MHz, acetone-*d*<sub>6</sub>): δ 173.9, 173.6, 159.0, 150.0, 148.4, 135.7, 132.3, 131.5, 131.3, 127.9, 124.0, 123.2, 121.5, 120.5, 110.2, 67.5, 39.6, 26.0, 18.4; HRMS (ESI, m/z) calculated for C<sub>19</sub>H<sub>16</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 318.1243, found 318.1241.

## References

1. Y. S. Lee, Y. H. Cho., S. Lee, J. K. Bin, J. H. Yang, G. Chae, C. H. Cheon *Tetrahedron*, **2015**, *71*, 532-538.
2. R. Mandal, B. Emayavaramban, B. Sundararaju *Org. Lett.* **2018**, *20*, 2815-2838.

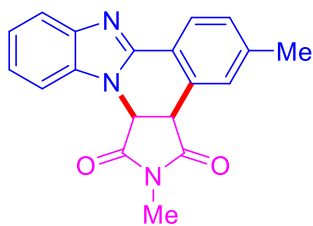
## Characterization data

### 2-methyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3a)



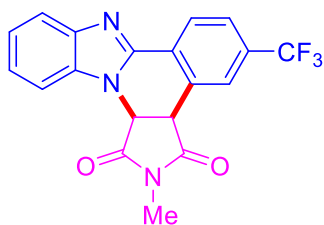
Pale yellow solid, 0.212 g, 90%; mp 239-241 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  8.42 (d,  $J = 8.7$  Hz, 1H), 7.92 (d,  $J = 8.1$  Hz, 1H), 7.79 (dd,  $J = 19.6, 7.6$  Hz, 2H), 7.50 (d,  $J = 4.0$  Hz, 2H), 7.35 (s, 2H), 5.65 (d,  $J = 9.7$  Hz, 1H), 4.68 (d,  $J = 9.7$  Hz, 1H), 3.07 (s, 3H);  $^{13}\text{C}$  NMR (101 MHz,  $\text{CDCl}_3$ ):  $\delta$  172.3, 145.5, 143.8, 134.9, 130.9, 129.2, 128.2, 126.2, 125.8, 124.4, 123.7, 123.5, 119.8, 111.1, 53.1, 41.6, 25.6; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{18}\text{H}_{14}\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  304.1086, found 304.1084.

### 2,5-dimethyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3b)



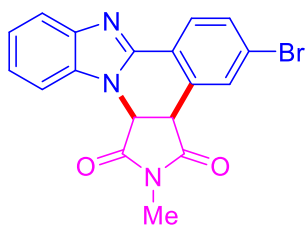
Pale yellow solid, 0.214 g, 87%; mp 237-239 °C;  $^1\text{H}$  NMR (600 MHz,  $\text{CDCl}_3$ ):  $\delta$  8.30 (d,  $J = 8.0$  Hz, 1H), 7.73 (dd,  $J = 7.9, 3.3$  Hz, 2H), 7.71 (s, 1H), 7.34 (t,  $J = 7.5$  Hz, 1H), 7.29 (t,  $J = 7.7$  Hz, 1H), 7.26 (s, 1H), 5.75 (d,  $J = 9.1$  Hz, 1H), 4.72 (d,  $J = 9.1$  Hz, 1H), 3.05 (s, 3H), 2.43 (s, 3H);  $^{13}\text{C}$  NMR (150 MHz,  $\text{CDCl}_3$ ):  $\delta$  173.9, 172.2, 145.2, 142.3, 134.2, 130.3, 128.6, 126.4, 126.2, 124.0, 118.70, 111.4, 53.3, 41.6, 25.7, 21.8; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{19}\text{H}_{16}\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  318.1243, found 318.1235.

### 2-methyl-5-(trifluoromethyl)-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3c)



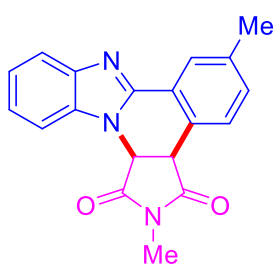
Pale yellow solid, 0.160 g, 75%; mp 265-267 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{acetone-}d_6$ ):  $\delta$  8.60 (d,  $J = 8.2$  Hz, 1H), 8.28 (s, 1H), 7.97 (d,  $J = 7.6$  Hz, 1H), 7.84 (d,  $J = 8.2$  Hz, 1H), 7.74 (d,  $J = 7.7$  Hz, 1H), 7.37 (dt,  $J = 19.5, 7.2$  Hz, 2H), 6.19 (d,  $J = 9.9$  Hz, 1H), 5.12 (d,  $J = 9.9$  Hz, 1H), 3.00 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{acetone-}d_6$ ):  $\delta$  174.2, 172.8, 144.2, 135.2, 129.2, 126.6 (q,  $J = 233$  Hz), 126.2, 125.7 (q,  $J = 4.1$  Hz), 125.4, 125.1 (q,  $J = 3.7$  Hz), 123.8, 123.5, 119.1, 112.7, 53.3, 41.4, 24.9; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{19}\text{H}_{13}\text{F}_3\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  372.0960, found 372.0957.

**5-bromo-2-methyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3d)**



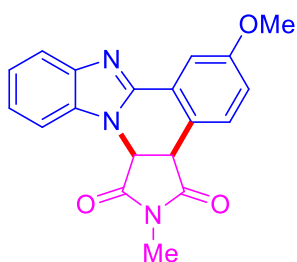
Orange solid, 0.168 g, 80%; mp 190-192 °C; <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ 8.18 (dd, *J* = 8.4, 3.5 Hz, 1H), 8.01 (s, 1H), 7.86 (d, *J* = 8.0 Hz, 1H), 7.72 (t, *J* = 8.1 Hz, 2H), 7.35 (dt, *J* = 22.6, 7.7 Hz, 2H), 6.08 (d, *J* = 9.8 Hz, 1H), 4.93 (d, *J* = 9.9 Hz, 1H), 2.88 (s, 3H); <sup>13</sup>C NMR (100 MHz, DMSO-*d*<sub>6</sub>): δ 174.9, 173.5, 145.1, 135.0, 132.0, 131.5, 130.9, 127.4, 124.5, 123.9, 123.3, 118.9, 113.1, 53.3, 41.3, 25.8; HRMS (ESI, *m/z*) calculated for C<sub>18</sub>H<sub>13</sub>BrN<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 382.0191, found 382.0186.

**2,6-dimethyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3e)**



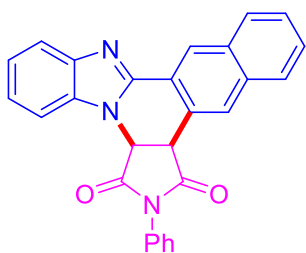
Yellow solid, 0.190 g, 83%; mp 210-212 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 8.23 (s, 1H), 7.77 (d, *J* = 7.7 Hz, 2H), 7.70 (d, *J* = 7.6 Hz, 1H), 7.41 – 7.28 (m, 3H), 5.59 (d, *J* = 9.6 Hz, 1H), 4.62 (d, *J* = 9.6 Hz, 1H), 3.04 (s, 3H), 2.41 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 174.2, 172.4, 145.7, 139.4, 134.8, 132.0, 128.1, 126.2, 123.9, 123.7, 123.6, 123.3, 119.6, 111.1, 53.2, 41.4, 25.6, 21.0; HRMS (ESI, *m/z*) calculated for C<sub>19</sub>H<sub>16</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 318.1243, found 318.1242.

**6-methoxy-2-methyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3f)**



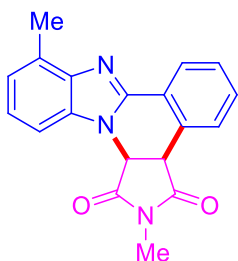
Yellow solid, 0.168 g, 81%; mp 234-236 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 7.96 (s, 1H), 7.83 (d, *J* = 8.7 Hz, 2H), 7.65 (d, *J* = 8.6 Hz, 1H), 7.40 (t, *J* = 7.6 Hz, 1H), 7.29 (d, *J* = 6.8 Hz, 1H), 7.07 (d, *J* = 8.6 Hz, 1H), 5.99 (d, *J* = 6.5 Hz, 1H), 4.88 (d, *J* = 6.5 Hz, 1H), 3.88 (s, 3H), 3.06 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 174.4, 172.4, 160.2, 145.6, 143.7, 135.0, 129.5, 125.5, 123.8, 123.5, 119.8, 119.0, 118.3, 111.1, 108.5, 55.8, 53.2, 41.2, 25.6; HRMS (ESI, *m/z*) calculated for C<sub>19</sub>H<sub>16</sub>N<sub>3</sub>O<sub>3</sub> (M + H)<sup>+</sup> 334.1192, found 334.1183.

**7-phenyl-5a,8a-dihydro-6H-benzo[*g*]benzo[4,5]imidazo[2,1-*a*]pyrrolo[3,4-*c*]isoquinoline-6,8(7*H*)-dione (3g)**



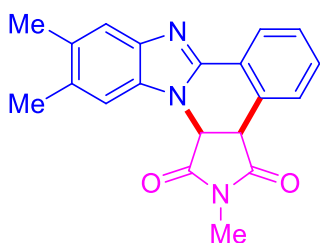
Yellow solid, 0.200 g, 80%; mp 205-207 °C; <sup>1</sup>H NMR (600 MHz, DMSO-*d*<sub>6</sub>): δ 9.18 (s, 1H), 8.49 (s, 2H), 8.13 – 8.07 (m, 1H), 8.04 – 8.00 (m, 1H), 7.95 (d, *J* = 7.3 Hz, 1H), 7.78 (d, *J* = 8.2 Hz, 1H), 7.67 – 7.60 (m, 2H), 7.46 (t, *J* = 7.4 Hz, 2H), 7.40 (dd, *J* = 18.3, 7.2 Hz, 2H), 7.36 (d, *J* = 8.5 Hz, 2H), 6.47 (d, *J* = 9.8 Hz, 1H), 5.34 (d, *J* = 9.3 Hz, 1H); <sup>13</sup>C NMR (150 MHz, DMSO-*d*<sub>6</sub>): δ 173.6, 171.9, 145.7, 134.4, 132.8, 132.2, 128.8, 128.7, 128.6, 128.1, 127.9, 127.4, 126.8, 124.6, 123.8, 123.5, 112.4, 53.8, 41.7, 29.4; HRMS (ESI, *m/z*) calculated for C<sub>27</sub>H<sub>18</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 416.1399, found 416.1395.

**2,9-dimethyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-*a*]pyrrolo[3,4-*c*]isoquinoline-1,3(2*H*)-dione (3h)**



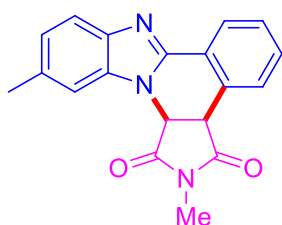
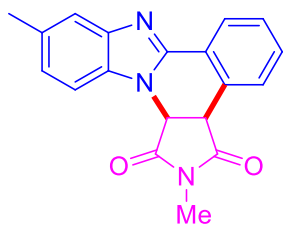
Yellow solid, 0.201 g, 88%; mp 232-234 °C; <sup>1</sup>H NMR (600 MHz, CDCl<sub>3</sub>): δ 8.49 (s, 1H), 7.88 (dd, *J* = 5.2, 3.8 Hz, 1H), 7.54 (d, *J* = 8.1 Hz, 1H), 7.47 (dd, *J* = 5.8, 3.4 Hz, 2H), 7.23 (d, *J* = 7.8 Hz, 1H), 7.11 (d, *J* = 7.3 Hz, 1H), 5.60 (d, *J* = 9.7 Hz, 1H), 4.65 (d, *J* = 9.6 Hz, 1H), 3.04 (s, 3H), 2.70 (s, 3H); <sup>13</sup>C NMR (150 MHz, CDCl<sub>3</sub>): δ 174.1, 172.4, 144.7, 134.6, 130.6, 130.0, 129.1, 128.1, 126.1, 126.0, 123.8, 123.6, 108.4, 53.2, 41.6, 25.6, 16.7; HRMS (ESI, *m/z*) calculated for C<sub>19</sub>H<sub>16</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 318.1243, found 318.1249.

**2,10,11-trimethyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-*a*]pyrrolo[3,4-*c*]isoquinoline-1,3(2*H*)-dione (3i)**



Yellow solid, 0.190 g, 85%; mp 215-217 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 8.43 – 8.32 (m, 1H), 7.94 – 7.83 (m, 1H), 7.56 (s, 1H), 7.51 – 7.45 (m, 3H), 5.57 (d, *J* = 9.7 Hz, 1H), 4.63 (d, *J* = 9.7 Hz, 1H), 3.06 (s, 3H), 2.42 (s, 3H), 2.39 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 174.2, 172.5, 144.6, 142.3, 133.3, 133.1, 132.6, 130.4, 129.1, 128.1, 125.9, 125.6, 119.7, 111.1, 53.1, 41.6, 28.2, 25.6, 20.7, 20.4; HRMS (ESI, *m/z*) calculated for C<sub>20</sub>H<sub>18</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 332.1399, found 332.1399.

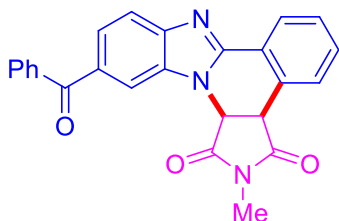
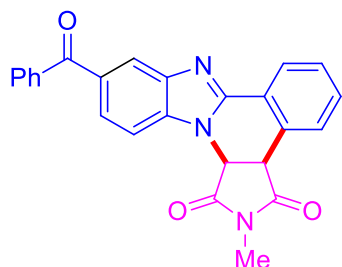
**2,10-dimethyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione & 2,11-dimethyl-3a,13a-dihydro-1H-benzo [4,5] imidazo[2,1-a] pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3j & 3j')**



Inseparable mixture; Pale yellow solid, 0.190 g, 83%; mp 232-234 °C; <sup>1</sup>H NMR (600 MHz, CDCl<sub>3</sub>) δ 8.41, 8.40, 8.40, 8.39, 8.39, 8.38, 8.38, 7.92, 7.91, 7.91, 7.90, 7.90, 7.69, 7.68, 7.63, 7.62, 7.59,

7.52, 7.50, 7.49, 7.49, 7.49, 7.48, 7.19, 7.17, 7.16, 5.59, 5.58, 5.57, 5.56, 4.66, 4.65, 4.64, 4.64, 3.07, 3.06, 2.54, 2.51; <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 174.1, 172.5, 172.4, 145.4, 145.1, 144.2, 142.0, 135.1, 133.9, 133.3, 133.0, 130.7, 130.6, 129.2, 128.2, 128.1, 126.0, 126.0, 125.8, 125.7, 125.3, 125.2, 124.6, 124.5, 119.6, 119.3, 110.8, 110.6, 53.1, 53.1, 41.60, 25.6, 22.0, 21.6; HRMS (ESI, *m/z*) calculated for C<sub>19</sub>H<sub>16</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 318.1243, found 318.1248.

**10-benzoyl-2-methyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione & 11-benzoyl-2-methyl-3a,13a-dihydro-1H-benzo [4,5]imidazo [2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3k & 3k')**

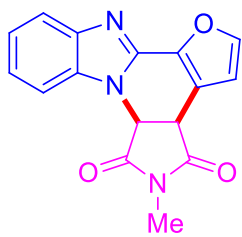


Inseparable mixture; Yellow solid, 0.154 g, 75%; mp 226-228 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 8.43, 8.41, 8.40, 8.38, 8.26, 8.20, 7.95, 7.94, 7.91, 7.86, 7.84, 7.82, 7.61, 7.59, 7.57, 7.55, 7.53, 7.51, 7.48,

7.47, 5.72, 5.69, 4.74, 4.71, 3.08, 3.05.; <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 196.5, 196.3, 173.8, 172.1, 148.3, 147.3, 147.1, 143.2, 138.1, 137.9, 134.7, 133.2, 132.8, 132.2, 131.6, 131.4, 130.1, 130.1, 129.4, 128.4, 128.3, 126.6, 126.3, 126.1, 126.1, 125.8, 123.9, 123.9, 123.2, 119.3, 114.1, 111.2, 53.1, 41.5, 29.7, 25.8; HRMS (ESI, *m/z*) calculated for C<sub>25</sub>H<sub>18</sub>N<sub>3</sub>O<sub>3</sub> (M + H)<sup>+</sup> 408.1348, found 408.1351.



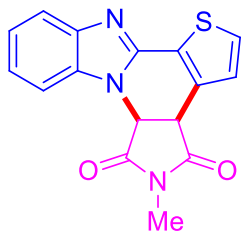
**5-methyl-3b,6a-dihydro-4H-benzo[4,5]imidazo[1,2-a]furo[2,3-c]pyrrolo[3,4-e]pyridine-4,6(5H)-dione (3m)**



Yellow solid, 0.175 g, 73%; mp 237-239 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 7.77 (d, *J* = 7.8 Hz, 1H), 7.62 (s, 1H), 7.60 (s, 1H), 7.32 (p, *J* = 7.2 Hz, 2H), 6.78 (s, 1H), 5.60 (d, *J* = 9.8 Hz, 1H), 4.64 (d, *J* = 9.8 Hz, 1H), 3.06 (s, 3H); <sup>13</sup>C NMR (101 MHz, CDCl<sub>3</sub>) δ 173.3, 172.3, 146.0, 143.6, 140.7, 139.4, 134.4, 124.1, 123.3, 120.2, 116.8, 110.6, 53.8, 39.8,

29.7, 25.7.; HRMS (ESI, *m/z*) calculated for C<sub>16</sub>H<sub>12</sub>N<sub>3</sub>O<sub>3</sub> (M + H)<sup>+</sup> 294.0879, found 294.0875.

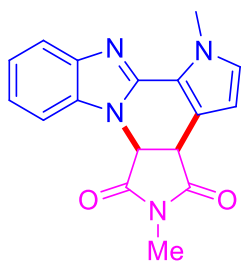
**5-methyl-3b,6a-dihydro-4H-benzo[4,5]imidazo[1,2-a]pyrrolo[3,4-e]thieno[2,3-c]pyridine-4,6(5H)-dione (3n)**



Yellow solid, 0.181 g, 78%; mp 242-244 °C; <sup>1</sup>H NMR (600 MHz, Acetone-*d*<sub>6</sub>): δ 7.77 (d, *J* = 8.1 Hz, 1H), 7.74 (d, *J* = 5.1 Hz, 1H), 7.62 (d, *J* = 7.9 Hz, 1H), 7.41 (d, *J* = 5.1 Hz, 1H), 7.30 (t, *J* = 8.1 Hz, 1H), 7.25 (t, *J* = 7.6 Hz, 1H), 6.11 (d, *J* = 9.8 Hz, 1H), 4.94 (d, *J* = 9.8 Hz, 1H), 2.98 (s, 3H); <sup>13</sup>C NMR (150 MHz, Acetone-*d*<sub>6</sub>): δ 173.9, 173.3,

144.0, 143.3, 135.0, 132.2, 129.0, 127.2, 123.0, 122.5, 119.1, 111.5, 54.0, 41.8, 24.7; HRMS (ESI, *m/z*) calculated for C<sub>16</sub>H<sub>12</sub>N<sub>3</sub>O<sub>2</sub>S (M + H)<sup>+</sup> 310.0650, found 310.0646.

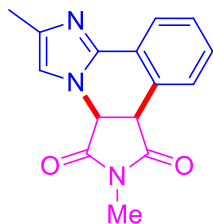
**1-methyl-5-phenyl-3b,6a-dihydrobenzo[4,5]imidazo[1,2-a]dipyrrolo[2,3-c:3',4'-e]pyridine-4,6(1H,5H)-dione (3o)**



Pale yellow solid, 0.181 g, 82%; mp 242-244 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 7.80 (d, *J* = 8.0 Hz, 1H), 7.62 (d, *J* = 8.1 Hz, 1H), 7.45 (t, *J* = 7.5 Hz, 2H), 7.40 (d, *J* = 7.2 Hz, 1H), 7.36 – 7.26 (m, 5H), 7.24 (d, *J* = 7.4 Hz, 1H), 6.85 (d, *J* = 2.6 Hz, 1H), 6.47 (d, *J* = 2.6 Hz, 1H), 4.15 (s, 3H); <sup>13</sup>C NMR (150 MHz, CDCl<sub>3</sub>): δ 171.7, 170.9, 135.5, 131.0, 130.6,

129.7, 129.4, 126.0, 125.7, 121.8, 114.3, 112.9, 112.1, 108.3, 55.1, 40.3, 34.6, 29.7; HRMS (ESI, *m/z*) calculated for C<sub>22</sub>H<sub>17</sub>N<sub>4</sub>O<sub>2</sub> (M + H)<sup>+</sup> 369.1352, found 369.1350.

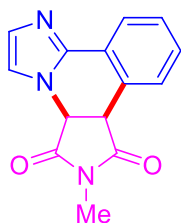
**2,6-dimethyl-4a,7a-dihydro-5H-imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-5,7(6H)-dione (3p)**



Pale yellow solid, 0.209 g, 82%; mp 231-233 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 8.10 (d, *J* = 7.2 Hz, 1H), 7.71 (d, *J* = 7.6 Hz, 1H), 7.46 – 7.28 (m, 2H), 7.08 (s, 1H), 5.22 (d, *J* = 9.6 Hz, 1H), 4.47 (d, *J* = 9.6 Hz, 1H), 3.05 (s, 3H), 2.29 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 174.3, 172.6, 140.4, 140.0, 129.1, 129.0, 128.9, 124.6, 123.8, 123.6, 115.7, 53.8, 41.9,

25.8, 13.8; HRMS (ESI, *m/z*) calculated for C<sub>15</sub>H<sub>14</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 268.1086, found 268.1081.

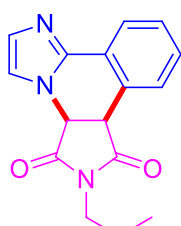
**6-methyl-4a,7a-dihydro-5H-imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-5,7(6H)-dione (3q)**



Orange solid, 0.221 g, 84%; mp 238-240 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 8.15 (d, *J* = 7.1 Hz, 1H), 7.75 (d, *J* = 8.0 Hz, 1H), 7.51 – 7.34 (m, 3H), 7.23 (s, 1H), 5.30 (d, *J* = 9.7 Hz, 1H), 4.52 (d, *J* = 9.6 Hz, 1H), 3.07 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 174.2, 172.4, 141.2, 130.6, 129.2, 128.9, 124.6, 123.9, 119.2, 54.0, 41.9, 25.8; HRMS (ESI, *m/z*) calculated for C<sub>14</sub>H<sub>12</sub>N<sub>3</sub>O<sub>2</sub>

(M + H)<sup>+</sup> 254.0930, found 254.0922.

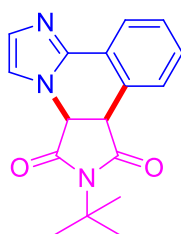
**6-propyl-4a,7a-dihydro-5H-imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-5,7(6H)-dione (3r)**



Off-white solid, 0.201 g, 90%; mp 140-142 °C; <sup>1</sup>H NMR (600 MHz, CDCl<sub>3</sub>): δ 8.15 (d, *J* = 7.5 Hz, 1H), 7.73 (d, *J* = 7.6 Hz, 1H), 7.42 (dt, *J* = 16.2, 7.3 Hz, 2H), 7.37 (s, 1H), 7.22 (s, 1H), 5.28 (d, *J* = 9.5 Hz, 1H), 4.49 (d, *J* = 9.6 Hz, 1H), 3.52 (t, *J* = 7.3 Hz, 2H), 1.58 (h, *J* = 7.4 Hz, 2H), 0.83 (t, *J* = 7.4 Hz, 3H); <sup>13</sup>C NMR (150 MHz, CDCl<sub>3</sub>): δ 174.2, 172.5, 141.2, 130.6, 129.2,

129.2, 128.9, 124.6, 124.1, 123.9, 119.2, 54.0, 41.9, 41.4, 20.8, 11.1; HRMS (ESI, *m/z*) calculated for C<sub>16</sub>H<sub>16</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 282.1243, found 282.1243.

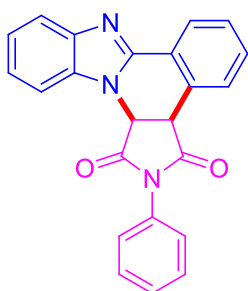
**6-(tert-butyl)-4a,7a-dihydro-5H-imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-5,7(6H)-dione (3s)**



Pale yellow solid, 0.239 g, 89%; mp 282-284 °C; <sup>1</sup>H NMR (600 MHz, CDCl<sub>3</sub>): δ 8.15 (d, *J* = 7.7 Hz, 1H), 7.68 (d, *J* = 7.7 Hz, 1H), 7.40 (dt, *J* = 23.4, 7.5 Hz, 2H), 7.32 (s, 1H), 7.21 (s, 1H), 5.12 (d, *J* = 9.7 Hz, 1H), 4.35 (d, *J* = 9.7 Hz,

1H), 1.54 (s, 9H); <sup>13</sup>C NMR (150 MHz, CDCl<sub>3</sub>): δ 175.0, 173.4, 141.3, 130.4, 129.1, 128.8, 124.7, 124.5, 123.8, 119.2, 59.9, 54.3, 42.2, 28.1; HRMS (ESI, *m/z*) calculated for C<sub>17</sub>H<sub>18</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 296.1399, found 296.1397.

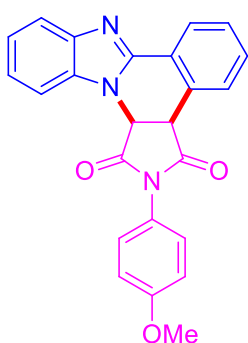
**2-phenyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3t)**



Yellow solid, 0.207 g, 73%; mp 255-257 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 8.44 (d, *J* = 7.3 Hz, 1H), 7.96 (d, *J* = 7.7 Hz, 1H), 7.79 (d, *J* = 7.8 Hz, 1H), 7.70 (d, *J* = 8.0 Hz, 1H), 7.55 – 7.27 (m, 9H), 6.13 (d, *J* = 8.2 Hz, 1H), 5.02 (d, *J* = 9.2 Hz, 1H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 172.7, 171.1, 144.6, 133.9, 132.1, 130.8, 129.6, 129.3, 128.3, 126.5, 126.1, 124.6, 118.2, 111.9, 53.6, 41.8; HRMS (ESI, *m/z*) calculated for

C<sub>23</sub>H<sub>16</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 366.1243, found 366.1246.

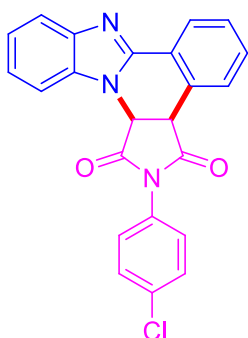
**2-(4-methoxyphenyl)-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione(3u)**



Pale yellow solid, 0.230 g, 75%; mp 241-243 °C; <sup>1</sup>H NMR (600 MHz, CDCl<sub>3</sub>) δ 8.52 – 8.44 (m, 1H), 7.99 – 7.90 (m, 1H), 7.81 (d, *J* = 7.5 Hz, 1H), 7.76 (d, *J* = 8.0 Hz, 1H), 7.59 – 7.46 (m, 2H), 7.39 – 7.30 (m, 2H), 7.19 (d, *J* = 9.0 Hz, 2H), 6.94 (d, *J* = 9.0 Hz, 2H), 5.86 (d, *J* = 9.7 Hz, 1H), 4.84 (d, *J* = 9.7 Hz, 1H), 3.80 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 173.2, 171.4, 159.9, 145.4, 134.7, 131.3, 129.4, 128.2, 127.3, 126.2, 124.0, 123.8, 123.4, 119.5, 114.5, 111.2, 55.5, 53.4, 41.7, 1.0;

HRMS (ESI, *m/z*) calculated for C<sub>24</sub>H<sub>18</sub>N<sub>3</sub>O<sub>3</sub> (M + H)<sup>+</sup> 396.1348, found 396.1343.

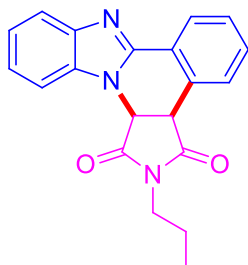
**2-(4-chlorophenyl)-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3v)**



Off-white solid, 0.210 g, 68%; mp 262-264 °C; <sup>1</sup>H NMR (600 MHz, acetone-*d*<sub>6</sub>): δ 8.38 (dd, *J* = 7.5, 1.8 Hz, 1H), 7.96 (d, *J* = 7.7 Hz, 1H), 7.85 (d, *J* = 7.4 Hz, 1H), 7.69 (d, *J* = 7.3 Hz, 1H), 7.59 – 7.48 (m, 4H), 7.40 (d, *J* = 8.7 Hz, 2H), 7.33 – 7.25 (m, 2H), 6.29 (d, *J* = 9.9 Hz, 1H), 5.12 (d, *J* = 9.8 Hz, 1H); <sup>13</sup>C NMR (150 MHz, acetone-*d*<sub>6</sub>): δ 173.6, 172.1, 145.9, 144.2, 135.6, 133.8, 131.0, 130.4, 129.0, 128.6, 128.4,

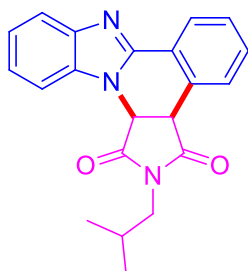
128.3, 127.7, 125.4, 125.1, 122.9, 122.7, 119.3, 111.8, 53.6, 41.9; HRMS (ESI,  $m/z$ ) calculated for  $C_{23}H_{15}ClN_3O_2$  ( $M + H$ )<sup>+</sup> 400.0853, found 400.0858.

**2-propyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3w)**



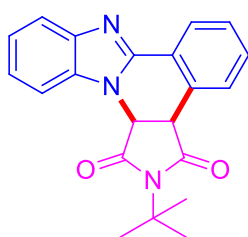
Pale yellow solid, 0.185 g, 80%; mp 162-164 °C; <sup>1</sup>H NMR (600 MHz, CDCl<sub>3</sub>): δ 8.35 (d,  $J = 9.2$  Hz, 1H), 7.84 (dd,  $J = 5.8, 2.6$  Hz, 1H), 7.80 – 7.73 (m, 1H), 7.69 – 7.58 (m, 1H), 7.51 – 7.37 (m, 2H), 7.34 – 7.24 (m, 2H), 5.46 (d,  $J = 9.7$  Hz, 1H), 4.54 (d,  $J = 9.7$  Hz, 1H), 3.54 – 3.37 (m, 2H), 1.54 (q,  $J = 7.4$  Hz, 2H), 0.78 (t,  $J = 7.4$  Hz, 3H); <sup>13</sup>C NMR (150 MHz, CDCl<sub>3</sub>): δ 174.1, 172.4, 145.5, 143.8, 134.9, 130.8, 129.1, 128.1, 126.3, 125.8, 124.3, 123.7, 123.4, 119.7, 53.0, 41.5, 41.3, 20.8, 11.1; HRMS (ESI,  $m/z$ ) calculated for  $C_{20}H_{18}N_3O_2$  ( $M + H$ )<sup>+</sup> 332.1399, found 332.1393.

**2-isobutyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3x)**



Yellow solid, 0.192 g, 75%; mp 192-194 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 8.31 (dd,  $J = 5.6, 3.7$  Hz, 1H), 7.78 (dd,  $J = 5.6, 4.0$  Hz, 1H), 7.76 – 7.70 (m, 1H), 7.56 (dd,  $J = 6.6, 2.1$  Hz, 1H), 7.44 – 7.36 (m, 2H), 7.29 – 7.18 (m, 2H), 5.42 (d,  $J = 9.7$  Hz, 1H), 4.49 (d,  $J = 9.8$  Hz, 1H), 3.35 – 3.21 (m, 2H), 1.91 (dp,  $J = 14.0, 6.9$  Hz, 1H), 0.73 (dd,  $J = 6.7, 4.5$  Hz, 6H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 174.3, 172.6, 145.4, 143.5, 134.8, 130.8, 129.1, 128.0, 126.4, 125.7, 124.0, 123.7, 123.5, 119.5, 111.1, 52.9, 46.7, 41.5, 26.9, 19.9, 19.8; HRMS (ESI,  $m/z$ ) calculated for  $C_{21}H_{20}N_3O_2$  ( $M + H$ )<sup>+</sup> 346.1556, found 346.1554.

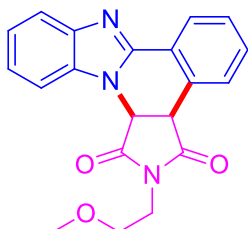
**2-(tert-butyl)-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3y)**



Pale yellow solid, 0.227 g, 85%; mp 144-146 °C; <sup>1</sup>H NMR (600 MHz, CDCl<sub>3</sub>): δ 8.37 (dd,  $J = 5.6, 3.5$  Hz, 1H), 7.85 – 7.80 (m, 1H), 7.81 – 7.75 (m, 1H), 7.63 – 7.57 (m, 1H), 7.46 (dd,  $J = 5.8, 3.3$  Hz, 2H), 7.30 (dd,  $J = 6.0, 3.1$  Hz, 2H), 5.39 (d,  $J = 9.8$  Hz, 1H), 4.44 (d,  $J = 9.8$  Hz, 1H), 1.53 (s, 9H); <sup>13</sup>C NMR (150 MHz, CDCl<sub>3</sub>): δ 174.9, 173.3, 145.7, 143.9, 135.1, 130.8, 129.1, 127.8, 126.6, 125.9, 124.5, 123.7, 123.3, 119.8, 110.8, 59.9, 53.3,

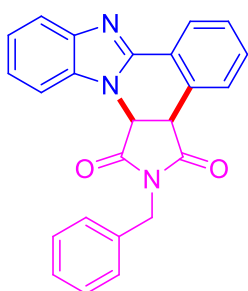
42.0, 28.2; HRMS (ESI,  $m/z$ ) calculated for  $C_{21}H_{20}N_3O_2$  ( $M + H$ )<sup>+</sup> 346.1556, found 346.1557.

**2-(2-methoxyethyl)-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3z)**



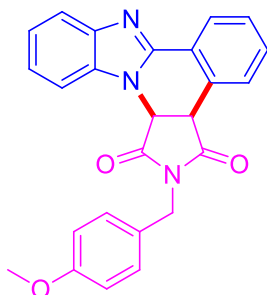
Pale yellow solid, 0.215 g, 80%; mp 174-176 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 8.37 (d,  $J = 7.4$  Hz, 1H), 7.86 (d,  $J = 7.4$  Hz, 1H), 7.72 (dd,  $J = 19.5, 7.7$  Hz, 2H), 7.44 (p,  $J = 7.4$  Hz, 2H), 7.36 – 7.23 (m, 2H), 5.66 (d,  $J = 9.8$  Hz, 1H), 4.67 (d,  $J = 9.8$  Hz, 1H), 3.81 – 3.65 (m, 2H), 3.52 (t,  $J = 5.4$  Hz, 2H), 3.21 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 173.9, 172.2, 145.3, 142.6, 134.6, 131.1, 129.2, 128.2, 126.5, 126.0, 123.9, 123.8, 123.6, 119.2, 111.4, 68.0, 58.5, 53.1, 41.5, 39.0, 30.9; HRMS (ESI,  $m/z$ ) calculated for  $C_{20}H_{18}N_3O_3$  ( $M + H$ )<sup>+</sup> 348.1348, found 348.1345.

**2-benzyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3za)**



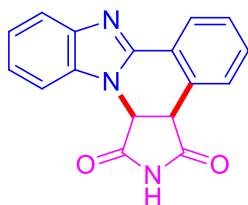
Off-white solid, 0.220 g, 75%; mp 225-227 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 8.47 – 8.38 (m, 1H), 7.91 (d,  $J = 7.7$  Hz, 1H), 7.76 (d,  $J = 9.3$  Hz, 2H), 7.54 – 7.40 (m, 2H), 7.39 – 7.26 (m, 7H), 5.73 (d,  $J = 9.8$  Hz, 1H), 4.74 (d,  $J = 9.8$  Hz, 1H), 4.68 (d,  $J = 2.1$  Hz, 2H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 173.4, 171.7, 144.7, 134.5, 134.0, 131.8, 129.4, 128.9, 128.9, 128.5, 128.3, 126.5, 126.4, 124.4, 118.5, 111.7, 53.3, 43.5, 41.6; HRMS (ESI,  $m/z$ ) calculated for  $C_{24}H_{18}N_3O_2$  ( $M + H$ )<sup>+</sup> 380.1399, found 380.1396.

**2-(4-methoxybenzyl)-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3zb)**



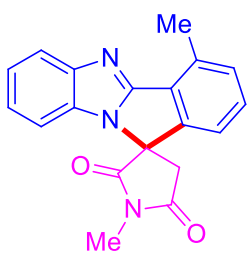
Pale yellow solid, 0.241 g, 76%; mp 243-245 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 8.39 (s, 1H), 7.87 (d,  $J = 5.2$  Hz, 1H), 7.78 (s, 1H), 7.69 (d,  $J = 4.9$  Hz, 1H), 7.53 – 7.42 (m, 2H), 7.29 (q,  $J = 7.7, 6.5$  Hz, 4H), 6.80 (d,  $J = 8.2$  Hz, 2H), 5.53 (d,  $J = 9.8$  Hz, 1H), 4.59 (s, 3H), 3.76 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 173.8, 172.0, 159.6, 145.5, 143.7, 134.9, 130.9, 130.5, 129.2, 128.2, 126.8, 126.1, 125.9, 124.3, 123.7, 123.5, 119.8, 114.1, 111.1, 55.3, 53.1, 42.9, 41.5; HRMS (ESI,  $m/z$ ) calculated for  $C_{25}H_{20}N_3O_3$  ( $M + H$ )<sup>+</sup> 410.1505, found 410.1507.

**3a, 13a-dihydro-1H-benzo [4, 5] imidazo [2, 1-a] pyrrolo [3, 4-c]isoquinoline-1, 3 (2H)-dione (3zc)**



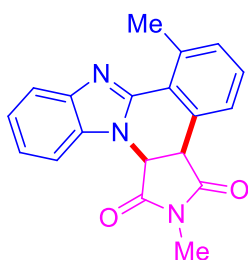
Yellow solid, 9 mg, 12%, <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ 8.27 – 8.22 (m, 1H), 7.80 (d, *J* = 7.5 Hz, 1H), 7.75 (d, *J* = 7.9 Hz, 1H), 7.69 (d, *J* = 7.8 Hz, 1H), 7.52 (p, *J* = 7.1 Hz, 2H), 7.28 (m, 2H), 6.12 (d, *J* = 9.6 Hz, 1H), 4.81 (d, *J* = 9.6 Hz, 1H). <sup>13</sup>C NMR (101 MHz, DMSO-*d*<sub>6</sub>) δ 176.4, 175.4, 145.9, 143.8, 135.5, 131.1, 129.1, 128.7, 128.3, 125.6, 124.6, 123.4, 123.2, 119.5, 112.2, 54.3, 43.2. HRMS (ESI, *m/z*) calculated for C<sub>17</sub>H<sub>11</sub>N<sub>3</sub>O<sub>2</sub> (M+H)<sup>+</sup> 290.0930, found 290.0926.

**1',4-dimethylspiro[benzo[4,5]imidazo[2,1-*a*]isoindole-11,3'-pyrrolidine]-2',5'-dione (5aa)**



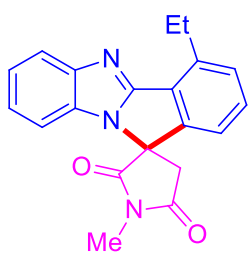
Pale yellow solid, 50 mg, 65%; mp 261-263 °C; <sup>1</sup>H NMR (400 MHz, acetone-*d*<sub>6</sub>): δ 7.79 (d, *J* = 7.6 Hz, 1H), 7.55 (d, *J* = 6.7 Hz, 1H), 7.48 (d, *J* = 8.9 Hz, 3H), 7.31 – 7.24 (m, 2H), 3.75 (d, *J* = 18.5 Hz, 1H), 3.54 (d, *J* = 18.5 Hz, 1H), 3.18 (s, 3H), 2.86 (s, 3H); <sup>13</sup>C NMR (100 MHz, acetone-*d*<sub>6</sub>): δ 173.9, 173.6, 159.0, 150.1, 148.4, 135.8, 132.3, 131.6, 131.3, 127.9, 124.1, 123.3, 121.6, 120.5, 110.2, 67.6, 39.6, 26.1, 18.5; HRMS (ESI, *m/z*) calculated for C<sub>19</sub>H<sub>16</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 318.1243, found 318.1241.

**2,7-dimethyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-*a*]pyrrolo[3,4-*c*]isoquinoline-1, 3(2H)-dione (3aa)**



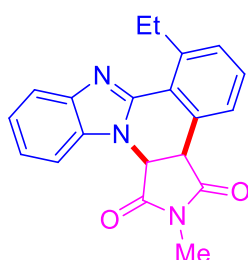
Pale yellow solid, 14 mg, 19%; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 7.87 (d, *J* = 8.0 Hz, 1H), 7.79 (d, *J* = 7.7 Hz, 1H), 7.70 (d, *J* = 8.0 Hz, 1H), 7.41 – 7.36 (m, 1H), 7.34 (d, *J* = 3.5 Hz, 1H), 7.33 – 7.30 (m, 1H), 7.28 (d, *J* = 7.7 Hz, 1H), 5.74 (s, 1H), 4.69 (d, *J* = 9.5 Hz, 1H), 3.04 (s, 3H), 2.99 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 174.2, 172.4, 139.6, 132.6, 130.5, 127.8, 125.9, 124.3, 123.7, 119.4, 110.9, 53.1, 42.3, 25.7, 24.0; HRMS (ESI, *m/z*) calculated for C<sub>19</sub>H<sub>16</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 318.1243, found 318.1238.

**4-ethyl-1'-methylspiro[benzo[4,5]imidazo[2,1-*a*]isoindole-11,3'-pyrrolidine]-2',5'-dione (5ab)**



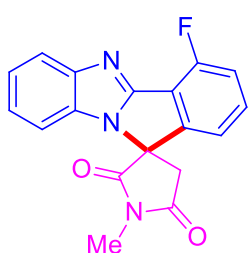
Pale yellow solid, 55 mg, 74%; mp 230-232 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.93 (d,  $J = 7.4$  Hz, 1H), 7.45 (d,  $J = 1.6$  Hz, 1H), 7.44 (s, 1H), 7.34 – 7.25 (m, 3H), 7.20-7.16 (m, 1H), 7.09 (d,  $J = 6.6$  Hz, 1H), 3.54 (d,  $J = 18.7$  Hz, 1H), 3.36 (m, 3H), 3.28 (s, 3H), 1.44 (t,  $J = 7.6$  Hz, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  172.9, 172.8, 157.9, 149.2, 146.6, 142.3, 130.7, 130.1, 130.0, 126.8, 123.6, 122.9, 121.5, 118.6, 108.1, 65.9, 39.2, 26.1, 25.6, 14.4; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{20}\text{H}_{18}\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  332.1399, found 332.1399.

**7-ethyl-2-methyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3ab)**



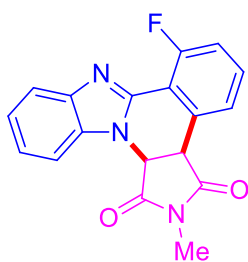
Pale yellow solid, 6 mg, 8%;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.84 (d,  $J = 7.8$  Hz, 1H), 7.78 (d,  $J = 7.5$  Hz, 1H), 7.70 – 7.66 (m, 1H), 7.43 – 7.27 (m, 4H), 5.65 (d,  $J = 9.6$  Hz, 1H), 4.66 (d,  $J = 9.6$  Hz, 1H), 3.68-3.63 (m, 1H), 3.54-3.49 (m, 1H), 3.05 (s, 3H), 1.33 (t,  $J = 7.4$  Hz, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  174.4, 172.5, 145.8, 134.0, 130.9, 130.3, 127.6, 125.8, 123.9, 123.3, 119.9, 110.4, 53.0, 42.4, 28.4, 25.6, 15.1; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{20}\text{H}_{18}\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  332.1399, found 332.1392.

**4-fluoro-1'-methylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5ac)**



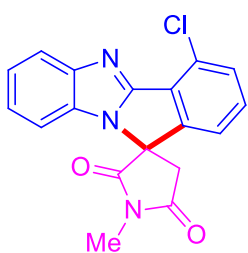
Pale yellow solid, 19 mg, 25%; mp 207-209 °C;  $^1\text{H}$  NMR (400 MHz, acetone- $d_6$ ):  $\delta$  7.99 (d,  $J = 7.7$  Hz, 1H), 7.57 (d,  $J = 6.4$  Hz, 1H), 7.40 – 7.31 (m, 3H), 7.21 (d,  $J = 7.5$  Hz, 1H), 7.12 (d,  $J = 7.5$  Hz, 1H), 3.61 (d,  $J = 18.4$  Hz, 1H), 3.44 (d,  $J = 18.4$  Hz, 1H), 3.29 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  172.1, 171.7, 158.6, 156.0, 153.5, 148.5, 133.2, 129.7, 124.6, 123.9 (d,  $J_{\text{C-F}} = 3.16$  Hz), 121.7, 118.3 (d,  $J_{\text{C-F}} = 18.2$  Hz), 117.5 (d,  $J_{\text{C-F}} = 3.1$  Hz), 108.3, 66.8, 39.0, 29.7, 26.3;  $^{19}\text{F}$  NMR (376 MHz,  $\text{CDCl}_3$ ):  $\delta$  -111.3; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{18}\text{H}_{13}\text{FN}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  322.0992, found 322.0988.

**7-fluoro-2-methyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3ac)**



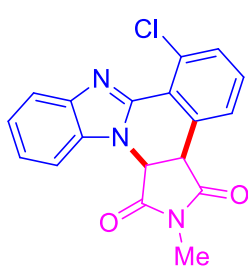
Pale yellow solid, 31 mg, 41%;  $^1\text{H NMR}$  (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.92 (d,  $J = 8.1$  Hz, 1H), 7.84 – 7.80 (m, 2H), 7.55 – 7.50 (m, 1H), 7.37 (t,  $J = 7.7$  Hz, 1H), 7.25 – 7.20 (m, 2H), 6.12 (d,  $J = 3.8$  Hz, 1H), 4.90 (d,  $J = 9.1$  Hz, 1H), 3.07 (s, 3H); HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{18}\text{H}_{13}\text{FN}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  322.0992, found 322.0986.

**4-chloro-1'-methylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5ad)**



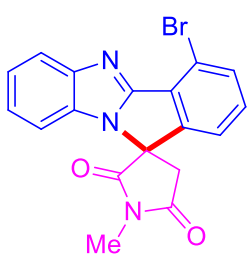
Pale yellow solid, 36 mg, 49%; mp 227-229 °C;  $^1\text{H NMR}$  (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  8.01 (d,  $J = 7.7$  Hz, 1H), 7.59 (d,  $J = 8.1$  Hz, 1H), 7.47 (t,  $J = 7.9$  Hz, 1H), 7.38 – 7.28 (m, 3H), 7.10 (d,  $J = 7.5$  Hz, 1H), 3.56 (d,  $J = 18.8$  Hz, 1H), 3.39 (d,  $J = 18.7$  Hz, 1H), 3.29 (s, 3H);  $^{13}\text{C NMR}$  (150 MHz,  $\text{CDCl}_3$ ):  $\delta$  173.4, 171.6, 144.5, 134.4, 132.0, 129.4, 128.9, 128.9, 128.5, 128.3, 126.6, 126.5, 124.6, 118.2, 111.9, 53.4, 43.5, 41.6; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{18}\text{H}_{13}\text{ClN}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  338.0696, found 338.0695.

**7-chloro-2-methyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3ad)**



Pale yellow solid, 7 mg, 9%;  $^1\text{H NMR}$  (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.93 (d,  $J = 7.7$  Hz, 1H), 7.88 (d,  $J = 7.9$  Hz, 1H), 7.70 (d,  $J = 7.9$  Hz, 1H), 7.58 (d,  $J = 8.0$  Hz, 1H), 7.54 – 7.49 (m, 1H), 7.41 – 7.35 (m, 2H), 5.64 (d,  $J = 9.9$  Hz, 1H), 4.68 (d,  $J = 9.7$  Hz, 1H), 3.08 (s, 3H); HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{18}\text{H}_{13}\text{ClN}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  338.0696.

**4-bromo-1'-methylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5ae)**



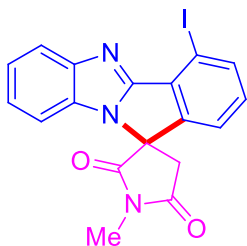
Off-white solid, 35 mg, 50%; mp 235-237 °C;  $^1\text{H NMR}$  (600 MHz,  $\text{CDCl}_3$ ):  $\delta$  8.03 (d,  $J = 7.6$  Hz, 1H), 7.77 (d,  $J = 7.8$  Hz, 1H), 7.36 (m, 4H), 7.09 (d,  $J = 7.4$  Hz, 1H), 3.56 (d,  $J = 18.6$  Hz, 1H), 3.39 (d,  $J = 18.6$  Hz, 1H), 3.28 (s, 3H);  $^{13}\text{C NMR}$  (150 MHz,  $\text{CDCl}_3$ ):  $\delta$  172.2, 171.9, 155.6, 148.1, 134.8, 131.5, 129.8, 129.0, 124.5, 123.5, 122.3,



120.2, 117.8, 108.2, 66.0, 39.0, 26.2; HRMS (ESI,  $m/z$ ) calculated for  $C_{18}H_{13}BrN_3O_2$  ( $M + H$ )<sup>+</sup> 382.0191, found 382.0183.

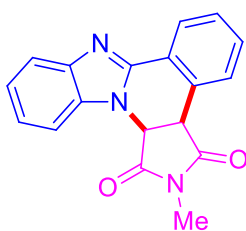
#### 4-iodo-1'-methylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione

(5af)



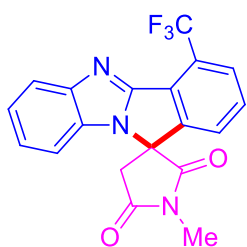
white solid, 13 mg, 19%; <sup>1</sup>H NMR (600 MHz, CDCl<sub>3</sub>): δ 8.06 – 8.00 (m, 2H), 7.36 – 7.29 (m, 3H), 7.21 (t,  $J = 7.8$  Hz, 1H), 7.08 (d,  $J = 6.5$  Hz, 1H), 3.54 (d,  $J = 18.7$  Hz, 1H), 3.37 (d,  $J = 18.7$  Hz, 1H), 3.28 (s, 3H); HRMS (ESI,  $m/z$ ) calculated for  $C_{18}H_{13}BrN_3O_2$  ( $M + H$ )<sup>+</sup> 430.0052, found 430.0046.

#### 2-methyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3a)



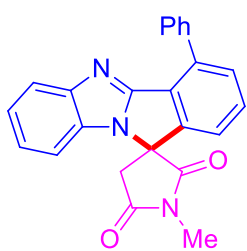
Off-white solid, 13 mg, 27%; <sup>1</sup>H NMR (600 MHz, CDCl<sub>3</sub>): δ 8.44-8.41 (m, 1H), 7.91 (d,  $J = 7.2$  Hz, 1H), 7.80 – 7.73 (m, 2H), 7.53 – 7.45 (m, 2H), 7.38-7.29 (m, 2H), 5.69 (d,  $J = 9.8$  Hz, 1H), 4.71 (d,  $J = 9.8$  Hz, 1H), 3.06 (s, 3H); <sup>13</sup>C NMR (150 MHz, CDCl<sub>3</sub>): δ 174.0, 172.3, 145.4, 134.8, 130.9, 129.3, 128.3, 126.2, 125.9, 124.2, 123.8, 123.6, 119.7, 111.1, 53.2, 41.6, 25.7; HRMS (ESI,  $m/z$ ) calculated for  $C_{18}H_{13}BrN_3O_2$  ( $M + H$ )<sup>+</sup> 304.1086, found 304.1080.

#### 1'-methyl-4-(trifluoromethyl)spiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5ag)



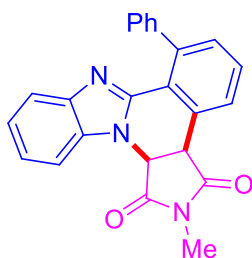
Pale yellow solid, 39 mg, 55%; mp 244-246 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 8.11 – 7.96 (m, 1H), 7.90 (d,  $J = 7.8$  Hz, 1H), 7.65 (t,  $J = 7.8$  Hz, 1H), 7.57 (d,  $J = 7.7$  Hz, 1H), 7.35 (tt,  $J = 6.8, 3.7$  Hz, 2H), 7.10 (d,  $J = 7.4$  Hz, 1H), 3.58 (d,  $J = 18.7$  Hz, 1H), 3.42 (d,  $J = 18.7$  Hz, 1H), 3.30 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 172.2, 171.8, 154.1, 148.7, 148.0, 130.6, 129.9, 127.7 (q,  $J_{C-CF} = 4.9$  Hz), 126.9, 126.1, 125.8, 125.5 (q,  $J_{C-F} = 271.73$  Hz), 124.8, 124.7, 123.7, 122.5, 108.2, 66.0, 38.9, 26.3; HRMS (ESI,  $m/z$ ) calculated for  $C_{19}H_{13}F_3N_3O_2$  ( $M + H$ )<sup>+</sup> 372.0960, found 372.0960.

**1'-methyl-4-phenylspiro[benzo[4,5]imidazo[2,1-*a*]isoindole-11,3'-pyrrolidine]-2',5'-dione (5ah)**



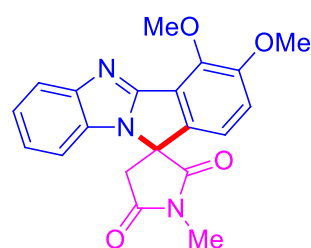
Pale yellow solid, 42 mg, 60%; mp 147-149 °C; <sup>1</sup>H NMR (600 MHz, CDCl<sub>3</sub>): δ 7.88 – 7.84 (m, 2H), 7.83 – 7.80 (m, 1H), 7.61 – 7.47 (m, 5H), 7.32 (d, *J* = 5.8 Hz, 1H), 7.26 (d, *J* = 5.7 Hz, 2H), 7.10 – 7.05 (m, 1H), 3.58 (d, *J* = 18.7 Hz, 1H), 3.42 (d, *J* = 18.7 Hz, 1H), 3.30 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 172.8, 172.8, 157.3, 148.8, 147.5, 139.5, 137.2, 132.1, 130.6, 130.1, 129.5, 128.8, 128.4, 125.9, 123.7, 122.9, 122.0, 119.9, 107.9, 65.6, 39.3, 26.1.; HRMS (ESI, *m/z*) calculated for C<sub>24</sub>H<sub>18</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 380.1399, found 380.1396.

**2-methyl-7-phenyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-*a*]pyrrolo[3,4-*c*]isoquinoline-1,3(2H)-dione (3ah)**



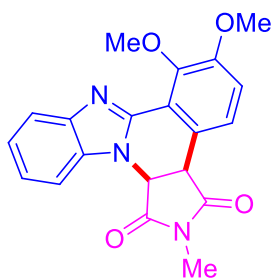
Off-white solid, 15 mg, 21%; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 7.91 (d, *J* = 7.8 Hz, 1H), 7.57 (d, *J* = 8.0 Hz, 1H), 7.49 (t, *J* = 7.8 Hz, 1H), 7.44 (d, *J* = 8.1 Hz, 1H), 7.37 (q, *J* = 6.1, 5.6 Hz, 6H), 7.27 – 7.23 (m, 2H), 7.18 – 7.15 (m, 1H), 5.64 (d, *J* = 9.6 Hz, 1H), 4.64 (d, *J* = 9.5 Hz, 1H), 3.07 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 174.3, 172.7, 144.7, 142.6, 141.7, 134.3, 132.7, 129.9, 129.5, 129.3, 128.4, 128.3, 127.9, 127.3, 123.9, 123.7, 123.1, 122.3, 120.3, 110.0, 53.1, 42.4, 25.7; HRMS (ESI, *m/z*) calculated for C<sub>24</sub>H<sub>18</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 380.1399, found 381.1392.

**3,4-dimethoxy-1'-methylspiro[benzo[4,5]imidazo[2,1-*a*]isoindole-11,3'-pyrrolidine]-2',5'-dione (5ai)**



Off-white solid, 33 mg, 46%; mp 239-241 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 7.91 (d, *J* = 8.3 Hz, 1H), 7.32 – 7.25 (m, 2H), 7.05 (d, *J* = 7.1 Hz, 1H), 6.98 (s, 2H), 4.28 (s, 3H), 3.91 (s, 3H), 3.54 (d, *J* = 18.6 Hz, 1H), 3.35 (d, *J* = 18.7 Hz, 1H), 3.24 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 172.8, 172.8, 155.6, 154.1, 148.8, 145.6, 138.9, 130.02, 123.78, 122.97, 121.64, 121.55, 116.4, 114.5, 108.0, 65.7, 61.9, 56.5, 39.4, 26.0; HRMS (ESI, *m/z*) calculated for C<sub>20</sub>H<sub>18</sub>N<sub>3</sub>O<sub>4</sub> (M + H)<sup>+</sup> 364.1297, found 364.1298.

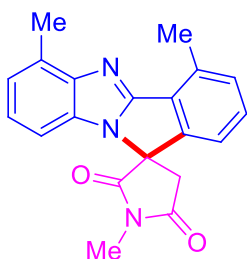
**6,7-dimethoxy-2-methyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3ai)**



Off-white solid, 23 mg, 32%;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.90 (d,  $J = 8.9$  Hz, 1H), 7.64 – 7.57 (m, 2H), 7.35 – 7.27 (m, 2H), 7.01 (d,  $J = 8.7$  Hz, 1H), 5.62 (d,  $J = 9.6$  Hz, 1H), 4.59 (d,  $J = 9.6$  Hz, 1H), 4.00 (s, 3H), 3.90 (s, 3H), 3.03 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  174.5, 172.5, 154.1, 148.4, 143.1, 133.9, 123.9, 123.8, 123.3, 120.5,

119.5, 114.7, 114.4, 110.4, 108.0, 61.0, 56.3, 53.1, 41.4, 25.6; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{20}\text{H}_{18}\text{N}_3\text{O}_4$  ( $\text{M} + \text{H}$ ) $^+$  364.1297, found 364.1296.

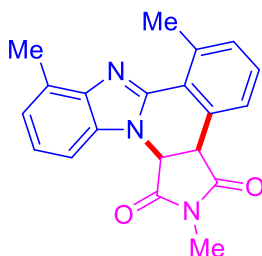
**1',4,6-trimethylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5aj)**



Yellow solid, 45 mg, 61%; mp 206-208 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.40 – 7.35 (m, 2H), 7.16 (t,  $J = 7.4$  Hz, 2H), 7.11 (d,  $J = 7.3$  Hz, 1H), 6.91 (d,  $J = 7.8$  Hz, 1H), 3.53 (d,  $J = 18.5$  Hz, 1H), 3.35 (d,  $J = 18.6$  Hz, 1H), 3.27 (s, 3H), 2.93 (s, 3H), 2.74 (s, 3H);  $^{13}\text{C}$  NMR (101 MHz,  $\text{CDCl}_3$ ):  $\delta$  172.9, 172.9, 157.3, 148.5, 146.5, 135.9, 131.9, 131.7, 130.1,

129.8, 127.6, 123.5, 123.4, 118.4, 105.5, 65.9, 39.2, 26.1, 18.7, 16.9; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{20}\text{H}_{18}\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  332.1399, found 332.1398.

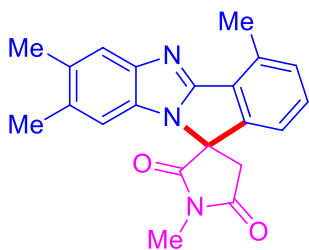
**2,7,9-trimethyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3aj)**



White solid, 10 mg, 14%;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.74 (d,  $J = 6.8$  Hz, 1H), 7.48 (d,  $J = 8.1$  Hz, 1H), 7.36 – 7.30 (m, 2H), 7.23 (d,  $J = 7.8$  Hz, 1H), 7.12 (d,  $J = 7.2$  Hz, 1H), 5.52 (d,  $J = 9.5$  Hz, 1H), 4.60 (d,  $J = 9.4$  Hz, 1H), 3.02 (s, 6H), 2.70 (s, 3H);  $^{13}\text{C}$  NMR (101 MHz,  $\text{CDCl}_3$ ):  $\delta$  174.5, 172.6, 145.1, 143.3, 139.5, 133.8, 132.5, 130.3,

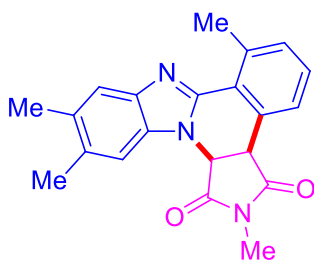
129.5, 127.2, 125.7, 123.7, 123.3, 123.3, 107.7, 53.1, 42.4, 25.6, 23.9, 16.5; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{20}\text{H}_{18}\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  332.1399, found 332.1391.

**1',4,7,8-tetramethylspiro[benzo[4,5]imidazo[2,1-*a*]isoindole-11,3'-pyrrolidine]-2',5'-dione (5ak)**



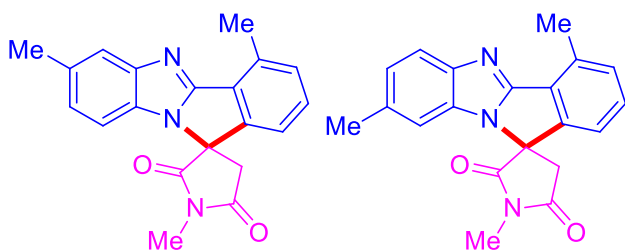
Yellow solid, 96 mg, 66%; mp 234-236 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 7.68 (s, 1H), 7.46 – 7.30 (m, 2H), 7.21 – 7.11 (m, 1H), 6.83 (s, 1H), 3.53 (d, *J* = 18.6 Hz, 1H), 3.34 (d, *J* = 18.6 Hz, 1H), 3.28 (s, 3H), 2.88 (s, 3H), 2.37 (s, 3H), 2.36 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 172.7, 172.7, 146.5, 136.0, 133.6, 132.5, 132.1, 130.5, 128.1, 127.0, 121.2, 118.4, 108.5, 66.2, 39.0, 26.2, 20.6, 20.4, 19.1; HRMS (ESI, *m/z*) calculated for C<sub>21</sub>H<sub>20</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 346.1556, found 346.1546.

**2,7,10,11-tetramethyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-*a*]pyrrolo[3,4-*c*]isoxaline-1,3(2H)-dione (3ak)**



Yellow solid, 19 mg, 13%; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 7.79 (d, *J* = 7.6 Hz, 1H), 7.68 (s, 1H), 7.47 (s, 1H), 7.36 (t, *J* = 7.6 Hz, 1H), 7.30 (d, *J* = 7.5 Hz, 1H), 5.84 (s, 1H), 4.69 (d, *J* = 8.6 Hz, 1H), 3.02 (s, 3H), 2.96 (s, 3H), 2.37 (s, 3H), 2.30 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 174.3, 172.6, 139.2, 133.8, 132.5, 131.9, 130.2, 127.5, 125.9, 119.2, 110.9, 53.1, 42.3, 25.6, 24.1, 20.7, 20.3; HRMS (ESI, *m/z*) calculated for C<sub>21</sub>H<sub>20</sub>N<sub>3</sub>O<sub>2</sub> (M + H)<sup>+</sup> 346.1556, found 346.1548.

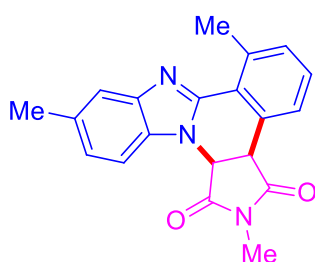
**1',4,7-trimethylspiro[benzo[4,5]imidazo[2,1-*a*]isoindole-11,3'-pyrrolidine]-2',5'-dione & 1',4,8-trimethylspiro [benzo[4,5]imidazo [2,1-*a*]isoindole-11,3'-pyrrolidine] -2',5'-dione (5al & 5al')**



Inseparable mixture; Pale yellow solid, 50 mg, 67%; mp 232-234 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 7.80, 7.78, 7.71, 7.39, 7.37, 7.17, 7.16, 7.15, 7.15, 7.15, 7.14, 7.12, 7.10, 7.08, 6.98, 6.96, 6.85, 3.57, 3.54, 3.53, 3.50, 3.38, 3.37, 3.33, 3.32, 3.29, 3.27, 2.89, 2.49, 2.47; <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 173.0, 172.9, 158.2, 157.7, 147.2, 146.5, 135.8, 133.9, 132.7, 131.9, 130.3, 130.2,

127.5, 125.0, 124.5, 121.5, 121.0, 118.5, 118.4, 108.1, 107.6, 65.9, 39.1, 26.1, 21.8, 21.6, 18.8; HRMS (ESI,  $m/z$ ) calculated for  $C_{20}H_{18}N_3O_4$  ( $M + H$ )<sup>+</sup> 332.1399, found 332.1397.

**2,7,10-trimethyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3al)**

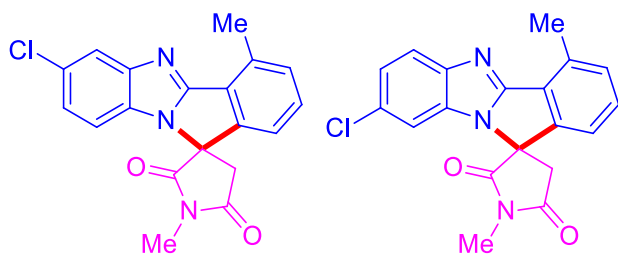


Yellow solid, 10 mg, 13%; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 7.75 (d,  $J$  = 7.1 Hz, 1H), 7.50 (d,  $J$  = 8.0 Hz, 1H), 7.38 – 7.31 (m, 2H), 7.27 (d,  $J$  = 9.9 Hz, 1H), 7.12 (d,  $J$  = 7.3 Hz, 1H), 5.61 (d,  $J$  = 9.0 Hz, 1H), 4.61 (d,  $J$  = 8.9 Hz, 1H), 3.03 (d,  $J$  = 3.5 Hz, 6H), 2.71 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 174.4, 172.5, 144.9, 139.7, 135.6,

133.7, 132.6, 130.1, 129.8, 127.4, 125.7, 123.9, 123.7, 107.8, 53.2, 42.4, 25.6, 23.8, 16.6;

HRMS (ESI,  $m/z$ ) calculated for  $C_{20}H_{18}N_3O_4$  ( $M + H$ )<sup>+</sup> 332.1399, found 332.1396.

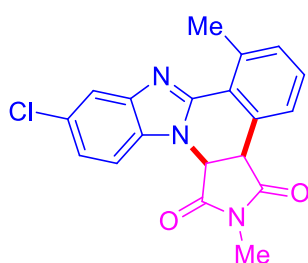
**7-chloro-1',4-dimethylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione & 8-chloro-1',4-dimethylspiro [benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5am & 5am')**



Inseparable mixture; Pale yellow solid, 50 mg, 69%; mp 239-241 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 7.93 – 7.82 (m, 1H), 7.46 – 7.38 (m, 2H), 7.32 – 7.26 (m, 1H), 7.18 (d,  $J$  = 7.2 Hz, 1H), 7.09 – 6.98 (m,

1H), 3.52 (dd,  $J$  = 18.7, 6.8 Hz, 1H), 3.37 (dd,  $J$  = 18.6, 5.1 Hz, 1H), 3.29 (d,  $J$  = 8.3 Hz, 3H), 2.90 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 172.5, 172.4, 159.5, 158.9, 150.0, 147.8, 146.5, 136.3, 136.2, 132.1, 132.1, 130.8, 130.8, 130.6, 129.3, 128.7, 128.6, 126.9, 126.8, 123.9, 123.6, 122.2, 121.4, 118.5, 118.5, 108.7, 108.3, 66.1, 66.1, 39.2, 39.1, 26.2, 26.2, 18.8; HRMS (ESI,  $m/z$ ) calculated for  $C_{19}H_{15}N_3O_2$  ( $M + H$ )<sup>+</sup> 352.0853, found 352.0847.

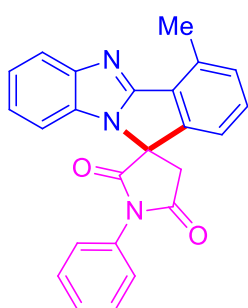
**10-chloro-2,7-dimethyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3am)**



White solid, 6 mg, 9%; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 7.77 (d,  $J$  = 7.7 Hz, 1H), 7.72 (d,  $J$  = 8.6 Hz, 1H), 7.69 (d,  $J$  = 1.9 Hz, 1H),

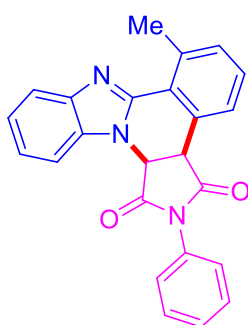
7.37 (t,  $J = 7.6$  Hz, 1H), 7.32 (d,  $J = 7.5$  Hz, 1H), 7.29 – 7.26 (m, 1H), 5.54 (d,  $J = 9.6$  Hz, 1H), 4.65 (d,  $J = 9.5$  Hz, 1H), 3.05 (s, 3H), 2.97 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  174.1, 172.3, 146.3, 139.7, 134.5, 132.7, 130.5, 129.9, 127.6, 126.1, 124.3, 120.5, 111.1, 53.2, 42.3, 25.7, 24.1; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{19}\text{H}_{15}\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  352.0853, found 352.0842.

**4-methyl-1'-phenylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5an)**



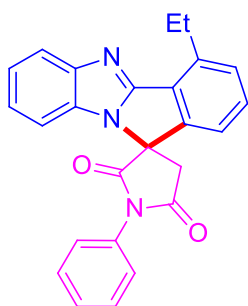
Pale yellow solid, 38 mg, 42%; mp 210-212 °C;  $^1\text{H}$  NMR (600 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.99 (d,  $J = 7.4$  Hz, 1H), 7.54 (t,  $J = 7.6$  Hz, 2H), 7.47 (t,  $J = 7.7$  Hz, 2H), 7.43 (d,  $J = 6.9$  Hz, 3H), 7.37 - 7.33 (m, 3H), 7.25 (s, 1H), 3.73 (d,  $J = 18.2$  Hz, 1H), 3.53 (d,  $J = 18.1$  Hz, 1H), 2.95 (s, 3H);  $^{13}\text{C}$  NMR (151 MHz,  $\text{CDCl}_3$ ):  $\delta$  171.5, 146.6, 136.4, 132.2, 131.2, 130.8, 129.5, 129.4, 129.3, 126.3, 126.1, 124.1, 123.3, 121.5, 118.5, 108.2, 66.2, 39.3, 19.1; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{24}\text{H}_{18}\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  379.1321, found 380.1395.

**7-methyl-2-phenyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3an)**



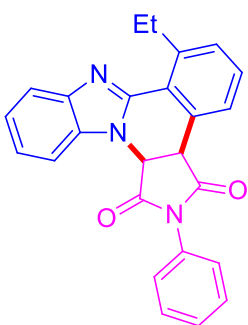
White solid, 26 mg, 29%;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.88 (d,  $J = 7.9$  Hz, 1H), 7.82 (d,  $J = 7.5$  Hz, 1H), 7.68 (d,  $J = 7.9$  Hz, 1H), 7.45 – 7.29 (m, 8H), 7.25 (d,  $J = 2.0$  Hz, 1H), 5.87 (d,  $J = 9.6$  Hz, 1H), 4.80 (d,  $J = 9.6$  Hz, 1H), 3.03 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  173.1, 171.3, 145.9, 143.7, 139.8, 134.9, 134.2, 132.9, 130.0, 129.4, 127.2, 126.9, 125.6, 124.0, 123.3, 123.2, 120.3, 110.2, 53.3, 42.6, 24.0; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{24}\text{H}_{18}\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  379.1321, found 380.1392.

**4-ethyl-1'-phenylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5ao)**



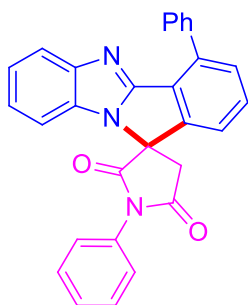
Red solid, 49 mg, 55%; mp 141-143 °C;  $^1\text{H}$  NMR (600 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.96 (d,  $J = 7.7$  Hz, 1H), 7.52 (t,  $J = 7.7$  Hz, 2H), 7.49 - 7.42 (m, 5H), 7.35 - 7.29 (m, 3H), 7.25 (d,  $J = 7.8$  Hz, 1H), 3.69 (d,  $J = 18.7$  Hz, 1H), 3.49 (d,  $J = 18.7$  Hz, 1H), 3.37 (hept,  $J = 7.3$  Hz, 2H), 1.45 (t,  $J = 7.6$  Hz, 3H);  $^{13}\text{C}$  NMR (150 MHz,  $\text{CDCl}_3$ ):  $\delta$  171.8, 171.7, 157.9, 149.2, 146.6, 142.4, 134.2, 131.3, 130.8, 130.2, 129.5, 129.4, 126.8, 126.2, 123.8, 122.9, 121.6, 118.6, 108.1, 66.0, 39.2, 25.6, 14.4; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{25}\text{H}_{20}\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  394.1556, found 394.1563.

**7-ethyl-2-phenyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinolin e-1,3(2H)-dione (3ao)**



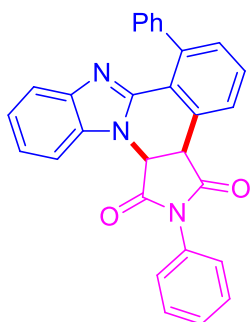
Pale yellow solid, 14 mg, 16%;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.85 (d,  $J = 9.0$  Hz, 1H), 7.79 (d,  $J = 6.5$  Hz, 1H), 7.66 (d,  $J = 7.0$  Hz, 1H), 7.47 - 7.26 (m, 9H), 5.74 (d,  $J = 9.6$  Hz, 1H), 4.74 (d,  $J = 9.5$  Hz, 1H), 3.66 - 3.57 (m, 2H), 1.37 (t,  $J = 7.4$  Hz, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  171.8, 171.7, 158.0, 149.2, 146.6, 142.4, 131.3, 130.8, 130.2, 129.5, 129.4, 126.8, 126.2, 123.8, 123.0, 121.6, 118.6, 108.1, 66.02, 39.24, 25.62, 14.43; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{25}\text{H}_{20}\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  394.1556, found 394.1551.

**1',4-diphenylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5ap)**



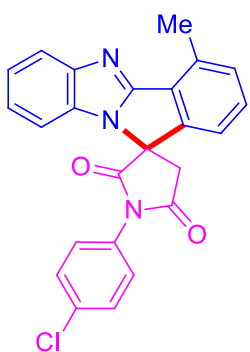
Pink solid, 29 mg, 36%; mp 144-146 °C;  $^1\text{H}$  NMR (600 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.90 - 7.86 (m, 2H), 7.85 - 7.82 (m, 1H), 7.61 (q,  $J = 7.8$  Hz, 2H), 7.63 - 7.53 (m, 4H), 7.52 - 7.44 (m, 4H), 7.31 - 7.27 (m, 2H), 7.24 - 7.22 (m, 1H), 3.75 (d,  $J = 18.7$  Hz, 1H), 3.58 (d,  $J = 18.7$  Hz, 1H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  171.8, 171.7, 157.3, 148.9, 147.5, 139.6, 137.2, 132.3, 131.3, 130.7, 130.2, 129.5, 129.5, 129.4, 128.8, 128.4, 126.1, 126.1, 123.9, 122.9, 122.1, 119.9, 107.9, 65.7, 39.4; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{29}\text{H}_{20}\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  442.1556, found 442.1559.

**2,7-diphenyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3ap)**



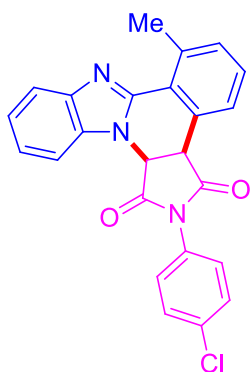
Off-white solid, 24 mg, 30%;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.93 (d,  $J = 7.8$  Hz, 1H), 7.57 (d,  $J = 8.2$  Hz, 1H), 7.52 (d,  $J = 7.7$  Hz, 1H), 7.49 (d,  $J = 3.2$  Hz, 1H), 7.46 (s, 1H), 7.43 (d,  $J = 7.6$  Hz, 2H), 7.40 (d,  $J = 5.8$  Hz, 6H), 7.29 (d,  $J = 7.4$  Hz, 2H), 7.24 (d,  $J = 7.6$  Hz, 1H), 7.16 (t,  $J = 7.6$  Hz, 1H), 5.84 (d,  $J = 9.2$  Hz, 1H), 4.73 (d,  $J = 9.1$  Hz, 1H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  173.2, 171.7, 144.7, 142.7, 141.6, 134.3, 132.9, 131.0, 130.2, 129.4, 129.3, 129.2, 129.1, 128.3, 127.9, 127.4, 127.1, 126.3, 126.1, 125.9, 123.9, 123.2, 120.3, 110.1, 53.4, 42.8 ; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{29}\text{H}_{20}\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  442.1556, found 442.1552.

**1'-(4-chlorophenyl)-4-methylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5aq)**



Off-white solid, 32 mg, 32%; mp 204-206 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.99 (d,  $J = 6.7$  Hz, 1H), 7.50 (d,  $J = 8.7$  Hz, 2H), 7.47 – 7.42 (m, 2H), 7.40 (d,  $J = 8.8$  Hz, 2H), 7.37 – 7.28 (m, 3H), 7.22 (d,  $J = 8.6$  Hz, 1H), 3.72 (d,  $J = 18.7$  Hz, 1H), 3.52 (d,  $J = 18.7$  Hz, 1H), 2.94 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  171.4, 171.2, 157.8, 146.4, 136.4, 135.3, 132.3, 130.8, 129.9, 129.7, 129.6, 127.3, 127.1, 124.1, 123.3, 121.5, 118.5, 108.1, 66.2, 39.1, 19.0; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{24}\text{H}_{17}\text{ClN}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  414.1009, found 414.1010.

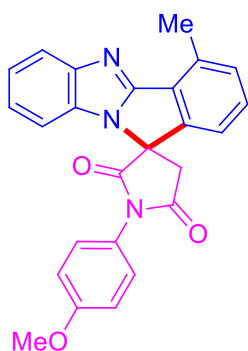
**2-(4-chlorophenyl)-7-methyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione(3aq)**



Pale red solid, 48 mg, 48%;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.86 (d,  $J = 8.9$  Hz, 1H), 7.78 (d,  $J = 6.8$  Hz, 1H), 7.65 (d,  $J = 7.0$  Hz, 1H), 7.43 – 7.30 (m, 6H), 7.23 (s, 2H), 5.75 (d,  $J = 9.5$  Hz, 1H), 4.77 (d,  $J = 9.4$  Hz, 1H), 3.03 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  173.3, 171.5, 145.9, 145.4, 134.2, 131.1, 131.0, 130.4, 129.2, 129.0, 127.4, 125.9, 125.6, 123.9, 123.3, 122.5, 120.1, 110.3, 53.3, 42.7, 28.4, 15.1; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{24}\text{H}_{17}\text{ClN}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  414.1009, found 414.0997.

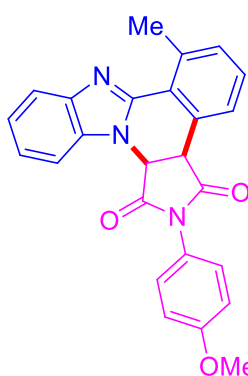


**1'-(4-methoxybenzyl)-4-methylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5ar)**



Off-White Solid, 57 mg, 58%; mp 206-208 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  8.06 (t,  $J = 5.1$  Hz, 1H), 7.51 – 7.43 (m, 2H), 7.41 – 7.27 (m, 6H), 7.04 – 6.99 (m, 2H), 3.83 (s, 3H), 3.72 (d,  $J = 18.0$  Hz, 1H), 3.52 (d,  $J = 17.7$  Hz, 1H), 2.97 (s, 3H);  $^{13}\text{C}$  NMR (101 MHz,  $\text{CDCl}_3$ ):  $\delta$  171.6, 171.3, 160.1, 146.7, 136.8, 132.4, 131.5, 129.4, 127.3, 126.2, 124.6, 123.9, 123.6, 120.9, 118.6, 114.8, 108.4, 66.6, 55.6, 38.9, 19.4; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{25}\text{H}_{20}\text{N}_3\text{O}_3$  ( $\text{M} + \text{H}$ ) $^+$  410.1505, found 410.1502.

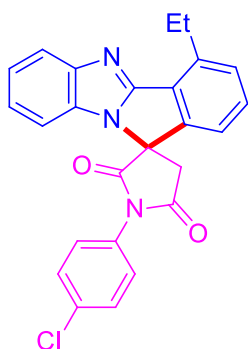
**2-(4-methoxyphenyl)-7-methyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3ar)**



Pale Yellow solid, 23 mg, 23%;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.87 – 7.83 (m, 1H), 7.79 (d,  $J = 6.8$  Hz, 1H), 7.69 – 7.64 (m, 1H), 7.38 – 7.31 (m, 4H), 7.16 (d,  $J = 9.0$  Hz, 2H), 6.92 (d,  $J = 9.0$  Hz, 2H), 5.73 (d,  $J = 9.6$  Hz, 1H), 4.74 (d,  $J = 9.6$  Hz, 1H), 3.79 (s, 3H), 3.03 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  173.6, 171.8, 159.8, 146.1, 143.9, 139.7, 134.3, 132.7, 129.9, 127.3, 127.2, 125.6, 123.9, 123.6,

123.3, 123.2, 120.3, 114.5, 110.2, 55.5, 53.2, 42.5, 24.0; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{26}\text{H}_{22}\text{N}_3\text{O}_3$  ( $\text{M} + \text{H}$ ) $^+$  410.1505, found 410.1496.

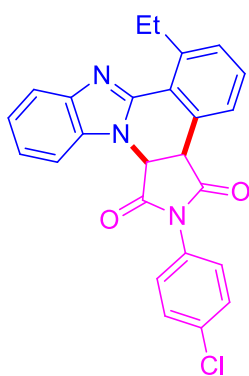
**1'-(4-chlorophenyl)-4-ethylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5as)**



Pale Yellow solid, 63 mg, 65%;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.98 (d,  $J = 8.9$  Hz, 1H), 7.49 (d,  $J = 8.8$  Hz, 3H), 7.44 – 7.37 (m, 3H), 7.34 (d,  $J = 7.2$  Hz, 1H), 7.29 (s, 1H), 7.22 (d,  $J = 8.8$  Hz, 1H), 6.83 (s, 1H), 3.71 (d,  $J = 18.7$  Hz, 1H), 3.50 (d,  $J = 18.7$  Hz, 1H), 3.39-3.36 (m, 2H), 1.45 (t,  $J = 7.6$  Hz, 3H);  $^{13}\text{C}$  NMR (101 MHz,  $\text{CDCl}_3$ ):  $\delta$  171.7, 171.6, 157.8,

148.8, 146.6, 142.5, 134.2, 131.3, 130.9, 130.2, 130.1, 129.5, 129.4, 129.1, 126.6, 126.1, 123.9, 123.1, 121.5, 118.6, 108.2, 66.1, 39.2, 25.7, 14.4; HRMS (ESI,  $m/z$ ) calculated for  $C_{25}H_{20}N_3O_3$  ( $M + H$ )<sup>+</sup> 428.1166, found 428.1158.

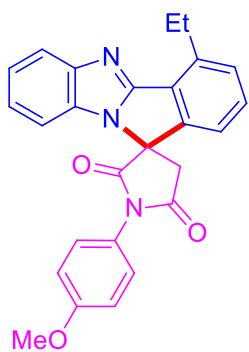
**2-(4-chlorophenyl)-7-ethyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3as)**



Off-White Solid, 11 mg, 12%; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 7.85 (d,  $J$  = 9.0 Hz, 1H), 7.80 – 7.76 (m, 1H), 7.66 (d,  $J$  = 6.8 Hz, 1H), 7.45 – 7.38 (m, 4H), 7.37 – 7.30 (m, 2H), 7.25 (d,  $J$  = 3.8 Hz, 2H), 5.79 (d,  $J$  = 9.4 Hz, 1H), 4.78 (d,  $J$  = 9.4 Hz, 1H), 3.67-3.55 (m, 2H), 1.36 (t,  $J$  = 7.4 Hz, 3H); <sup>13</sup>C NMR (101 MHz, CDCl<sub>3</sub>): δ 173.1, 171.3, 146.1, 145.4, 134.9, 134.2, 131.2, 130.3, 129.5, 129.4, 127.2, 127.1, 125.6, 123.9,

123.3, 122.7, 120.3, 110.1, 53.3, 42.7, 28.4, 15.1; HRMS (ESI,  $m/z$ ) calculated for  $C_{25}H_{20}N_3O_3$  ( $M + H$ )<sup>+</sup> 428.1166, found 428.1160.

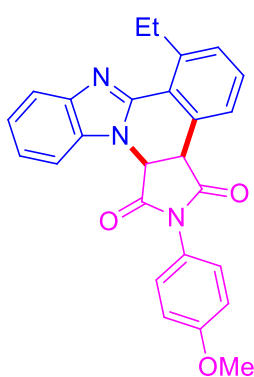
**4-ethyl-1'-(4-methoxyphenyl)spiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5at)**



Pale Yellow solid, 66 mg, 69%; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 7.98 (d,  $J$  = 8.4 Hz, 1H), 7.53 – 7.45 (m, 2H), 7.34 (d,  $J$  = 8.8 Hz, 5H), 7.23 (d,  $J$  = 8.8 Hz, 1H), 7.03 (d,  $J$  = 8.7 Hz, 2H), 3.84 (s, 3H), 3.71 (d,  $J$  = 18.6 Hz, 1H), 3.51 (d,  $J$  = 18.6 Hz, 1H), 3.40 - 3.37 (m, 2H), 1.46 (t,  $J$  = 7.5 Hz, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 171.9, 171.8, 160.0, 157.6, 146.7, 142.6, 134.1, 131.0, 130.2, 129.9, 127.6, 127.4, 123.9, 123.8,

123.2, 121.5, 118.6, 114.7, 114.5, 108.2, 66.1, 55.6, 39.2, 25.7, 14.5; HRMS (ESI,  $m/z$ ) calculated for  $C_{26}H_{22}N_3O_3$  ( $M + H$ )<sup>+</sup> 424.1661, found 424.1657.

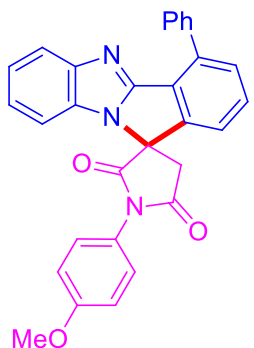
**7-ethyl-2-(4-methoxyphenyl)-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3at)**



Pale Yellow solid, 9 mg, 11%;  $^1\text{H NMR}$  (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.85 (d,  $J = 8.8$  Hz, 1H), 7.79 (d,  $J = 6.3$  Hz, 1H), 7.67 (d,  $J = 7.1$  Hz, 1H), 7.41 (d,  $J = 6.7$  Hz, 2H), 7.36 – 7.28 (m, 2H), 7.17 (d,  $J = 8.9$  Hz, 2H), 6.92 (d,  $J = 9.0$  Hz, 2H), 5.76 (d,  $J = 9.6$  Hz, 1H), 4.73 (d,  $J = 9.6$  Hz, 1H), 3.79 (s, 3H), 3.79 - 3.57 (m, 2H), 1.36 (t,  $J = 7.4$  Hz, 3H);  $^{13}\text{C NMR}$  (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  173.5, 171.7, 159.8, 145.9,

133.9, 130.9, 130.6, 127.7, 127.3, 125.7, 124.1, 123.5, 119.7, 114.5, 110.6, 55.5, 53.3, 42.6, 28.4, 14.9 ; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{26}\text{H}_{22}\text{N}_3\text{O}_3$  ( $\text{M} + \text{H}$ ) $^+$  424.1661, found 424.1663.

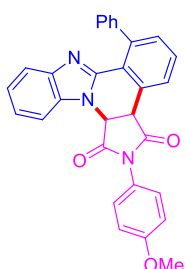
**1'-(4-methoxyphenyl)-4-phenylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5au)**



Off-White Solid, 38 mg, 44%;  $^1\text{H NMR}$  (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.89 – 7.81 (m, 3H), 7.62 – 7.44 (m, 6H), 7.38 – 7.34 (m, 2H), 7.32 – 7.26 (m, 2H), 7.24 – 7.21 (m, 1H), 7.06 – 7.01 (m, 2H), 3.84 (s, 3H), 3.71 (d,  $J = 18.7$  Hz, 1H), 3.55 (d,  $J = 18.7$  Hz, 1H);  $^{13}\text{C NMR}$  (101 MHz,  $\text{CDCl}_3$ ):  $\delta$  171.9, 171.9, 160.0, 157.2, 148.7, 147.5, 139.6, 137.2, 132.2, 130.7, 130.2, 129.5, 128.8, 128.4, 127.4, 125.9, 123.9, 123.8, 122.9, 122.1,

119.9, 114.7, 107.9, 65.7, 55.6, 39.3; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{25}\text{H}_{20}\text{N}_3\text{O}_3$  ( $\text{M} + \text{H}$ ) $^+$  472.1661, found 472.1655.

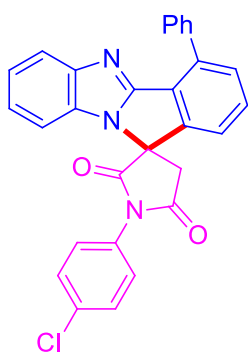
**2-(4-methoxyphenyl)-7-phenyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3au)**



Pale Yellow Solid, 25 mg, 29%;  $^1\text{H NMR}$  (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.96 – 7.93 (m, 1H), 7.61 (d,  $J = 8.0$  Hz, 1H), 7.55 – 7.47 (m, 3H), 7.39 (s, 6H), 7.20 (d,

$J = 8.8$  Hz, 3H), 7.13 (t,  $J = 7.6$  Hz, 1H), 6.94 (d,  $J = 8.9$  Hz, 2H), 5.96 (d,  $J = 9.2$  Hz, 1H), 4.76 (d,  $J = 9.1$  Hz, 1H), 3.80 (s, 3H);  $^{13}\text{C}$  NMR (101 MHz,  $\text{CDCl}_3$ ):  $\delta$  173.4, 171.8, 159.8, 144.5, 142.8, 141.3, 134.1, 132.9, 130.4, 129.4, 128.5, 128.0, 127.5, 127.3, 127.2, 124.1, 123.5, 123.3, 119.9, 114.5, 110.3, 55.5, 53.4, 42.7; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{25}\text{H}_{20}\text{N}_3\text{O}_3$  ( $\text{M} + \text{H}$ ) $^+$  472.1661, found 472.1655.

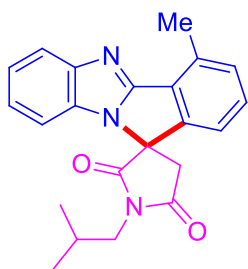
**1'-(4-chlorophenyl)-4-phenylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5av)**



Off-White Solid, 34 mg, 39%;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.86 (d,  $J = 6.8$  Hz, 3H), 7.64 – 7.50 (m, 7H), 7.47 – 7.40 (m, 3H), 7.31–7.28 (m, 2H), 7.22 – 7.18 (m, 1H), 3.76 (d,  $J = 18.7$  Hz, 1H), 3.58 (d,  $J = 18.7$  Hz, 1H);  $^{13}\text{C}$  NMR (101 MHz,  $\text{CDCl}_3$ ):  $\delta$  170.9, 170.7, 155.7, 147.4, 140.6, 136.6, 135.5, 132.7, 131.9, 129.8, 129.5, 129.3, 129.2, 129.1, 127.3, 125.0, 124.3, 121.6, 119.9, 108.2, 66.4, 39.0; HRMS (ESI,  $m/z$ )

calculated for  $\text{C}_{25}\text{H}_{20}\text{N}_3\text{O}_3$  ( $\text{M} + \text{H}$ ) $^+$  476.1166, found 476.1161.

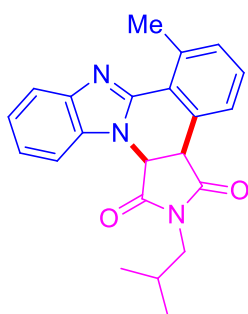
**1'-isobutyl-4-methylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5aw)**



Pale Yellow Solid, 58 mg, 67%; mp 229–231 °C;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.93 (d,  $J = 5.8$  Hz, 1H), 7.39 (d,  $J = 6.9$  Hz, 2H), 7.33 – 7.25 (m, 2H), 7.16 (d,  $J = 5.4$  Hz, 1H), 7.11 (d,  $J = 6.6$  Hz, 1H), 3.60 – 3.52 (m, 3H), 3.34 (d,  $J = 17.8$  Hz, 1H), 2.90 (s, 3H), 2.23 – 2.15 (m, 1H),

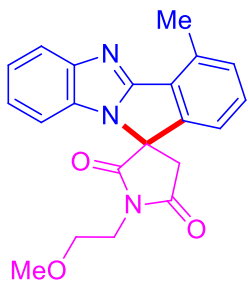
0.99 (d,  $J = 6.7$  Hz, 6H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  172.9, 158.2, 149.0, 146.9, 136.0, 131.9, 130.5, 130.1, 127.3, 123.7, 122.9, 121.5, 118.3, 108.1, 65.9, 47.3, 38.9, 27.4, 20.2, 20.2, 18.9; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{22}\text{H}_{22}\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  360.1712, found 360.1707.

**2-isobutyl-7-methyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3aw)**



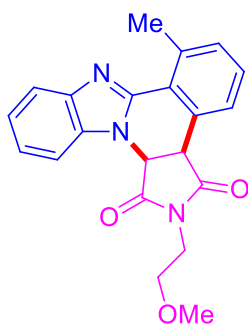
Red Solid, 15 mg, 17%;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.86 (d,  $J = 7.9$  Hz, 1H), 7.76 (d,  $J = 7.6$  Hz, 1H), 7.66 (d,  $J = 8.0$  Hz, 1H), 7.39 - 7.27 (m, 4H), 5.72 - 5.60 (m, 1H), 4.65 (d,  $J = 9.4$  Hz, 1H), 3.41 - 3.30 (m, 2H), 2.99 (s, 3H), 2.00 - 1.93 (m, 1H), 0.76 (d,  $J = 4.4$  Hz, 6H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  174.5, 172.6, 139.6, 133.8, 132.6, 130.3, 127.8, 125.7, 124.1, 123.5, 119.6, 110.6, 53.0, 46.8, 42.4, 26.9, 24.0, 19.9, 19.8; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{22}\text{H}_{22}\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  360.1712, found 360.1710.

**1'-(2-methoxyethyl)-4-methylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5ax)**



Pale yellow solid, 60 g, 69%; mp 139-141  $^\circ\text{C}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.92 (d,  $J = 8.3$  Hz, 1H), 7.39 (d,  $J = 8.4$  Hz, 2H), 7.30 (q,  $J = 8.6, 8.0$  Hz, 2H), 7.21 (t,  $J = 9.2$  Hz, 2H), 3.98 (q,  $J = 5.3$  Hz, 2H), 3.68 (s, 2H), 3.54 (d,  $J = 18.6$  Hz, 1H), 3.39 (s, 3H), 3.34 (d,  $J = 18.7$  Hz, 1H), 2.90 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  174.4, 172.5, 146.0, 143.8, 139.5, 134.2, 132.5, 129.7, 127.4, 125.8, 123.8, 123.1, 120.2, 110.4, 68.1, 58.5, 52.9, 42.4, 38.9, 24.0; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{21}\text{H}_{20}\text{N}_3\text{O}_3$  ( $\text{M} + \text{H}$ ) $^+$  362.1505, found 362.1501.

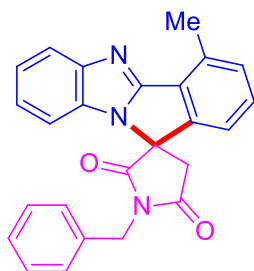
**2-(2-methoxyethyl)-7-methyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3ax)**



Pale yellow solid, 15 mg, 17%;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.83 (d,  $J = 8.1$  Hz, 1H), 7.75 (d,  $J = 7.5$  Hz, 1H), 7.66 (d,  $J = 7.7$  Hz, 1H), 7.38 - 7.29 (m, 4H), 5.58 (d,  $J = 9.6$  Hz, 1H), 4.62 (d,  $J = 9.6$  Hz, 1H), 3.76 - 3.70 (m, 2H), 3.53 (d,  $J = 5.5$  Hz, 2H), 3.22 (s, 3H), 3.00 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  172.7, 172.7, 158.2, 149.1, 146.9, 135.9,

131.9, 130.4, 130.1, 127.3, 123.6, 122.9, 121.4, 118.7, 108.3, 67.8, 65.8, 58.6, 39.2, 29.7, 18.8; HRMS (ESI,  $m/z$ ) calculated for  $C_{21}H_{20}N_3O_3$  ( $M + H$ )<sup>+</sup> 362.1505, found 362.1499.

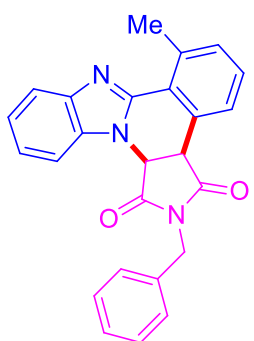
**1'-benzyl-4-methylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5ay)**



Off-white solid, 54 mg, 57%; mp 149-151 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 8.00 (d,  $J = 8.2$  Hz, 1H), 7.46 – 7.43 (m, 2H), 7.40 – 7.35 (m, 5H), 7.32 (d,  $J = 7.9$  Hz, 1H), 7.17 (t,  $J = 7.7$  Hz, 1H), 7.03 – 6.99 (m, 1H), 6.78 (d,  $J = 8.0$  Hz, 1H), 4.90 (s, 2H), 3.51 (d,  $J = 18.7$

Hz, 1H), 3.36 (d,  $J = 18.7$  Hz, 1H), 2.93 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 171.9, 146.6, 136.5, 134.9, 132.2, 131.2, 129.0, 129.0, 128.7, 124.3, 123.7, 120.8, 118.6, 108.4, 66.5, 43.9, 39.1, 19.3; HRMS (ESI,  $m/z$ ) calculated for  $C_{25}H_{20}N_3O_2$  ( $M + H$ )<sup>+</sup> 394.1556, found 394.1561.

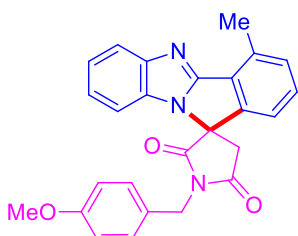
**2-benzyl-7-methyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3ay)**



Pale yellow solid, 22 mg, 23%; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ 7.83 (d,  $J = 8.9$  Hz, 1H), 7.75 (d,  $J = 7.3$  Hz, 1H), 7.64 (d,  $J = 7.1$  Hz, 1H), 7.37 – 7.26 (m, 9H), 5.53 (d,  $J = 9.6$  Hz, 1H), 4.65 (s, 2H), 4.60 (d,  $J = 9.5$  Hz, 1H), 3.00 (s, 3H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 174.1, 172.2, 145.9, 143.7, 139.6, 134.7, 134.1, 132.6, 129.8, 128.9, 128.8, 128.7, 128.3, 127.9, 127.3, 125.8, 123.8, 123.2, 122.9, 120.1, 110.4, 52.9, 43.3,

42.3, 24.1; HRMS (ESI,  $m/z$ ) calculated for  $C_{25}H_{20}N_3O_2$  ( $M + H$ )<sup>+</sup> 394.1556, found 394.1547.

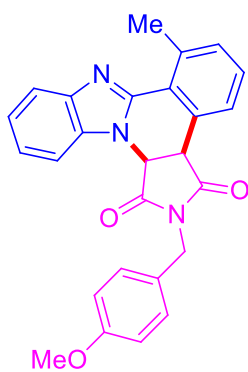
**1'-(4-methoxybenzyl)-4-methylspiro[benzo[4,5]imidazo[2,1-a]isoindole-11,3'-pyrrolidine]-2',5'-dione (5az)**



Pale Yellow solid, 71 mg, 70%; mp 85-87 °C; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>): δ <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>) δ 7.88 (d,  $J = 8.1$  Hz, 1H),

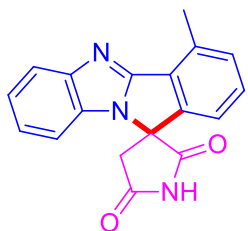
7.35 – 7.27 (m, 4H), 7.23 (t,  $J = 7.1$  Hz, 1H), 7.09 (t,  $J = 7.7$  Hz, 1H), 6.95 (d,  $J = 8.5$  Hz, 1H), 6.83 (d,  $J = 8.6$  Hz, 2H), 6.73 (d,  $J = 8.0$  Hz, 1H), 4.79 – 4.75 (m, 2H), 3.75 (s, 3H), 3.41 (d,  $J = 18.7$  Hz, 1H), 3.26 (d,  $J = 18.7$  Hz, 1H), 2.85 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  172.4, 172.3, 159.7, 157.8, 148.5, 146.5, 135.9, 131.9, 130.6, 130.5, 130.3, 129.9, 127.4, 127.1, 123.6, 122.9, 121.1, 118.7, 114.2, 114.0, 108.3, 66.1, 55.3, 43.1, 39.1, 18.9; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{26}\text{H}_{22}\text{N}_3\text{O}_3$  ( $\text{M} + \text{H}$ ) $^+$  424.1661, found 424.1662.

**2-(4-methoxybenzyl)-7-methyl-3a,13a-dihydro-1H-benzo[4,5]imidazo[2,1-a]pyrrolo[3,4-c]isoquinoline-1,3(2H)-dione (3az)**



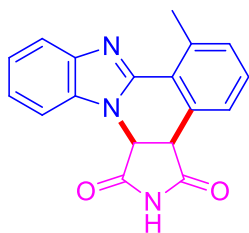
Pale Yellow solid, 14 mg, 14%;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.82 (d,  $J = 9.0$  Hz, 1H), 7.74 (d,  $J = 7.4$  Hz, 1H), 7.63 (d,  $J = 7.0$  Hz, 1H), 7.36 – 7.26 (m, 6H), 6.79 (d,  $J = 8.3$  Hz, 2H), 5.48 (d,  $J = 9.6$  Hz, 1H), 4.57 (d,  $J = 10.3$  Hz, 3H), 3.76 (s, 3H), 2.99 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  174.1, 172.3, 159.6, 145.9, 143.8, 139.5, 134.2, 132.5, 130.5, 129.7, 127.3, 126.9, 125.8, 123.8, 123.1, 123.0, 120.2, 114.1, 110.4, 55.3, 52.9, 42.8, 42.3, 24.1; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{26}\text{H}_{22}\text{N}_3\text{O}_3$  ( $\text{M} + \text{H}$ ) $^+$  424.1661, found 424.1651.

**4-methylspiro [benzo[4, 5] imidazo [2, 1-a] isoindole-11, 3'-pyrrolidine]-2',5'-dione (5bc)**



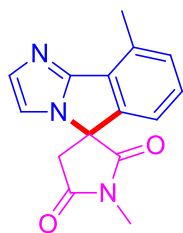
Pale yellow solid, 26 mg, 35%,  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO}-d_6$ ); HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{18}\text{H}_{12}\text{N}_3\text{O}_2$  ( $\text{M} + \text{H}$ ) $^+$  304.1086, found 304.1082.

**7-methyl-3a, 13a-dihydro-1H-benzo [4, 5] imidazo [2, 1-a] pyrrolo [3, 4-c] isoquinoline-1, 3 (2H)-dione (3bc)**



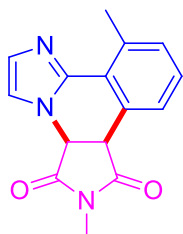
Pale yellow solid, 11 mg, 15%,  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO-d}_6$ )  $\delta$  11.93 (s, 1H), 7.75 – 7.65 (m, 3H), 7.42 (t,  $J = 7.6$  Hz, 1H), 7.38 – 7.24 (m, 3H), 6.14 (d,  $J = 9.5$  Hz, 1H), 4.77 (d,  $J = 9.5$  Hz, 1H), 2.90 (s, 3H).  $^{13}\text{C}$  NMR (101 MHz,  $\text{DMSO-d}_6$ )  $\delta$  176.5, 175.5, 146.3, 143.5, 138.6, 134.7, 132.3, 130.2, 129.9, 125.9, 123.6, 123.0, 123.0, 119.8, 111.6, 53.9, 43.8, 24.1. HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{18}\text{H}_{12}\text{N}_3\text{O}_2$  ( $\text{M}+\text{H}$ ) $^+$  304.1086, found 304.1082.

**1', 9-dimethylspiro [imidazo [2, 1-a] isoindole-5, 3'-pyrrolidine]-2', 5'-dione (5bd)**



Pale yellow solid, 78 mg, 46%,  $^1\text{H}$  NMR (400 MHz,  $\text{Acetone-d}_6$ )  $\delta$  7.51 (d,  $J = 1.4$  Hz, 1H), 7.36 – 7.33 (m, 1H), 7.31 – 7.24 (m, 3H), 7.18 (d,  $J = 1.4$  Hz, 1H), 3.50 (d,  $J = 18.3$  Hz, 1H), 3.39 (d,  $J = 18.3$  Hz, 1H), 3.04 (s, 3H), 2.68 (s, 3H).  $^{13}\text{C}$  NMR (101 MHz,  $\text{Acetone-d}_6$ )  $\delta$  173.6, 172.9, 153.5, 146.4, 134.3, 132.3, 131.1, 128.4, 128.1, 119.3, 114.4, 66.8, 40.6, 25.2, 17.6. HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{15}\text{H}_{13}\text{N}_3\text{O}_2$  ( $\text{M}+\text{H}$ ) $^+$  268.1086, found 268.1081.

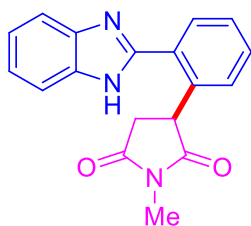
**6, 11-dimethyl-4aH-imidazo [2, 1-a] pyrrolo [3, 4-c] isoquinoline-5, 7 (6H, 7aH)-dione (3bd)**



Pale yellow solid, 59 mg, 35%,  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO-d}_6$ )  $\delta$  7.52 (d,  $J = 7.4$  Hz, 1H), 7.43 (d,  $J = 1.2$  Hz, 1H), 7.30 – 7.21 (m, 2H), 7.16 (d,  $J = 1.3$  Hz, 1H), 5.58 (d,  $J = 9.4$  Hz, 1H), 4.72 (d,  $J = 9.4$  Hz, 1H), 2.86 (s, 3H), 2.73 (s, 3H).  $^{13}\text{C}$  NMR (101 MHz,  $\text{DMSO-d}_6$ )  $\delta$  175.7, 174.0, 141.3, 135.9, 131.9, 129.8, 128.3, 127.4, 127.1, 123.2, 119.9, 54.2, 42.7, 25.7, 23.8. HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{15}\text{H}_{13}\text{N}_3\text{O}_2$  ( $\text{M}+\text{H}$ ) $^+$  268.1086, found 268.1080.



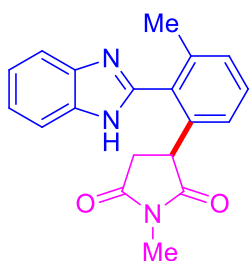
### 3-(2-(1H-benzo[d]imidazol-2-yl)phenyl)-1-methylpyrrolidine-2,5-dione (4a)



Yellow gummy mass,  $^1\text{H}$  NMR (400 MHz, acetone- $d_6$ ):  $\delta$  11.86 (s, 1H), 7.95 – 7.83 (m, 1H), 7.62 – 7.51 (m, 2H), 7.52 (s, 3H), 7.22 (p,  $J = 6.6$  Hz, 2H), 4.81 (dd,  $J = 9.4, 6.0$  Hz, 1H), 3.26 (dd,  $J = 17.5, 9.5$  Hz, 1H), 3.05 (dd,  $J = 17.5, 5.9$  Hz, 1H), 2.80 (s, 3H);  $^{13}\text{C}$  NMR (100 MHz, acetone- $d_6$ ):  $\delta$  178.19, 176.16, 137.50, 131.81, 129.83, 129.45, 127.85,

122.91, 121.76, 119.25, 111.18, 45.87, 37.98, 27.92, 24.15; HRMS (ESI,  $m/z$ ) calculated for  $\text{C}_{18}\text{H}_{16}\text{N}_3\text{O}_2$  (M) $^+$  306.1243, found 306.1237.

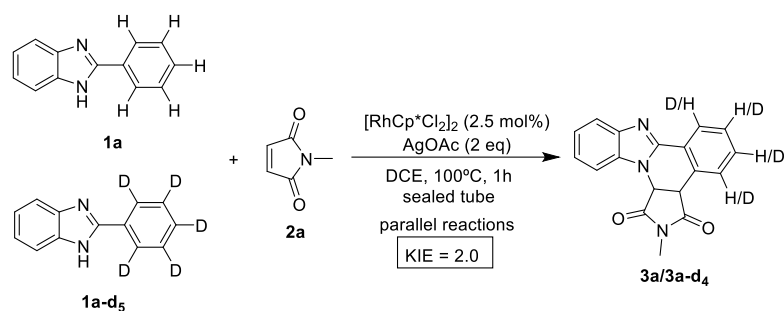
### 3-(2-(1H-benzo[d]imidazol-2-yl)-3-methylphenyl)-1-methylpyrrolidine-2,5-dione (6a)



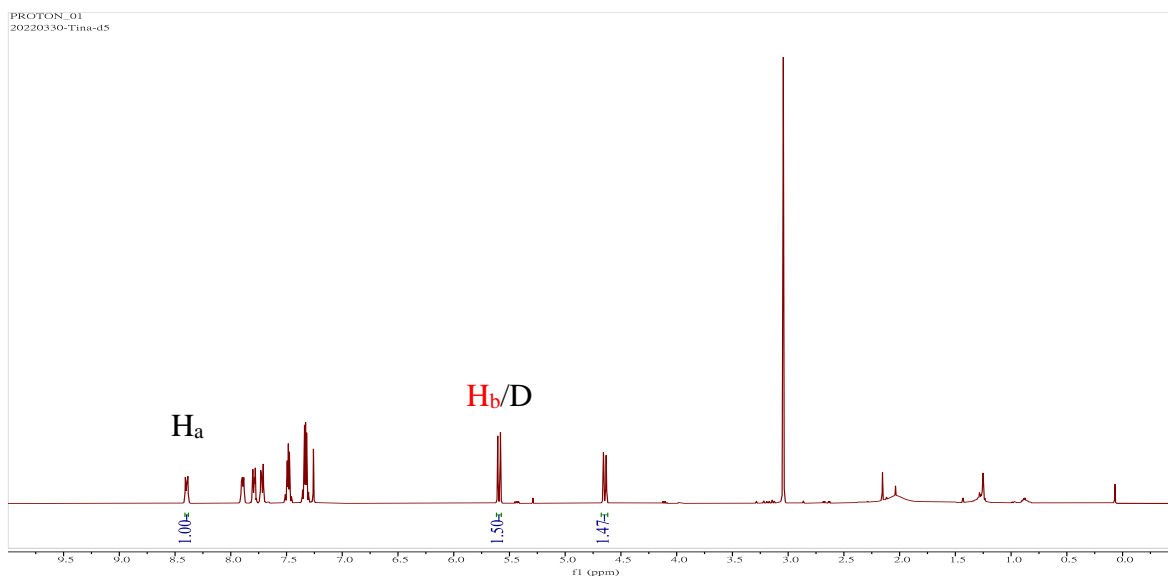
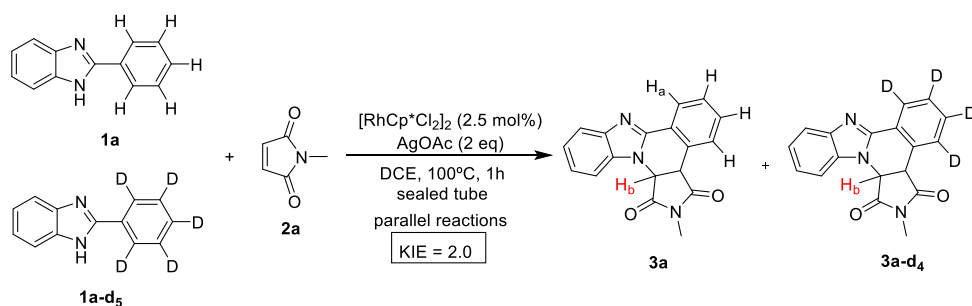
Pale yellow gummy mass,  $^1\text{H}$  NMR (600 MHz, DMSO- $d_6$ ):  $\delta$  8.27 (d,  $J = 6.5$  Hz, 1H), 7.91 (d,  $J = 7.9$  Hz, 1H), 7.82 (d,  $J = 7.9$  Hz, 1H), 7.68 (d,  $J = 7.7$  Hz, 1H), 7.58 (dt,  $J = 25.1, 7.1$  Hz, 2H), 7.27 (dd,  $J = 11.9, 7.6$  Hz, 1H), 5.93 (s, 1H), 4.28 (dd,  $J = 8.9, 6.9$  Hz, 1H), 2.94 (s, 3H), 2.73 (s, 3H), 2.43 – 2.36 (m, 1H), 2.02 (dd,  $J = 17.4, 6.8$  Hz, 1H;  $^{13}\text{C}$  NMR (150 MHz, DMSO- $d_6$ ):  $\delta$  176.24, 175.17, 172.20, 145.07, 143.70,

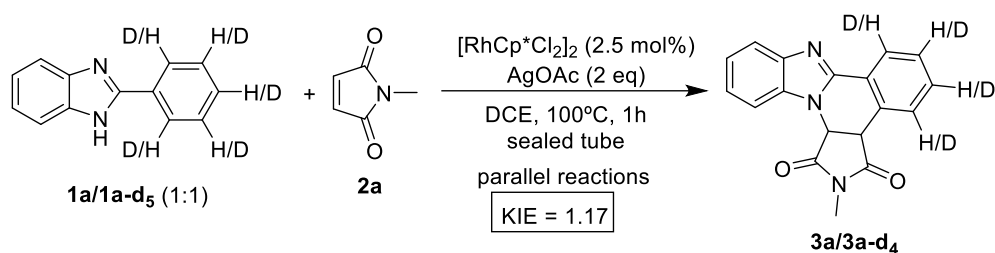
135.48, 131.66, 130.01, 127.91, 125.72, 124.79, 123.34, 119.42, 113.40, 55.97, 49.04, 31.44, 26.08, 25.10.

## KIE study



**Competitive reactions:** To an oven-dried sealed tube was added 2-phenyl benzimidazole **1a** (0.05 g, 0.258 mmol), **1a-d<sub>5</sub>** (0.05 g, 0.251 mmol), *N*-methylmaleimide **2a** (0.057 g, 0.52 mmol),  $[\text{RhCp}^*\text{Cl}_2]_2$  (3.9 mg, 0.006 mmol), AgOAc (0.085 g, 0.515 mmol) and 1,2-dichloroethane (3 mL). The reaction mixture was heated at 100 °C for 1 h. The reaction mixture was cooled down at room temperature and filtered through celite bed. The filtrate was concentrated under reduced pressure. The column chromatography (20% ethyl acetate/n-hexanes) purification afforded a mixture of **3a** and **3a-d<sub>4</sub>**. The KIE value was calculated from <sup>1</sup>H NMR spectrum ( $\text{K}_\text{H}/\text{K}_\text{D} = 1/(1.5-1) = 2.0$ ).



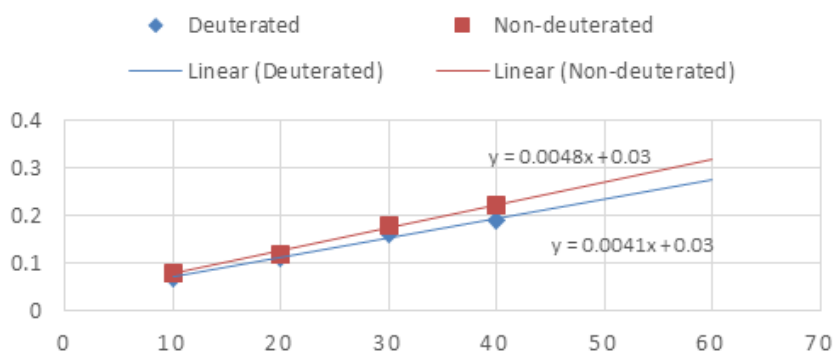


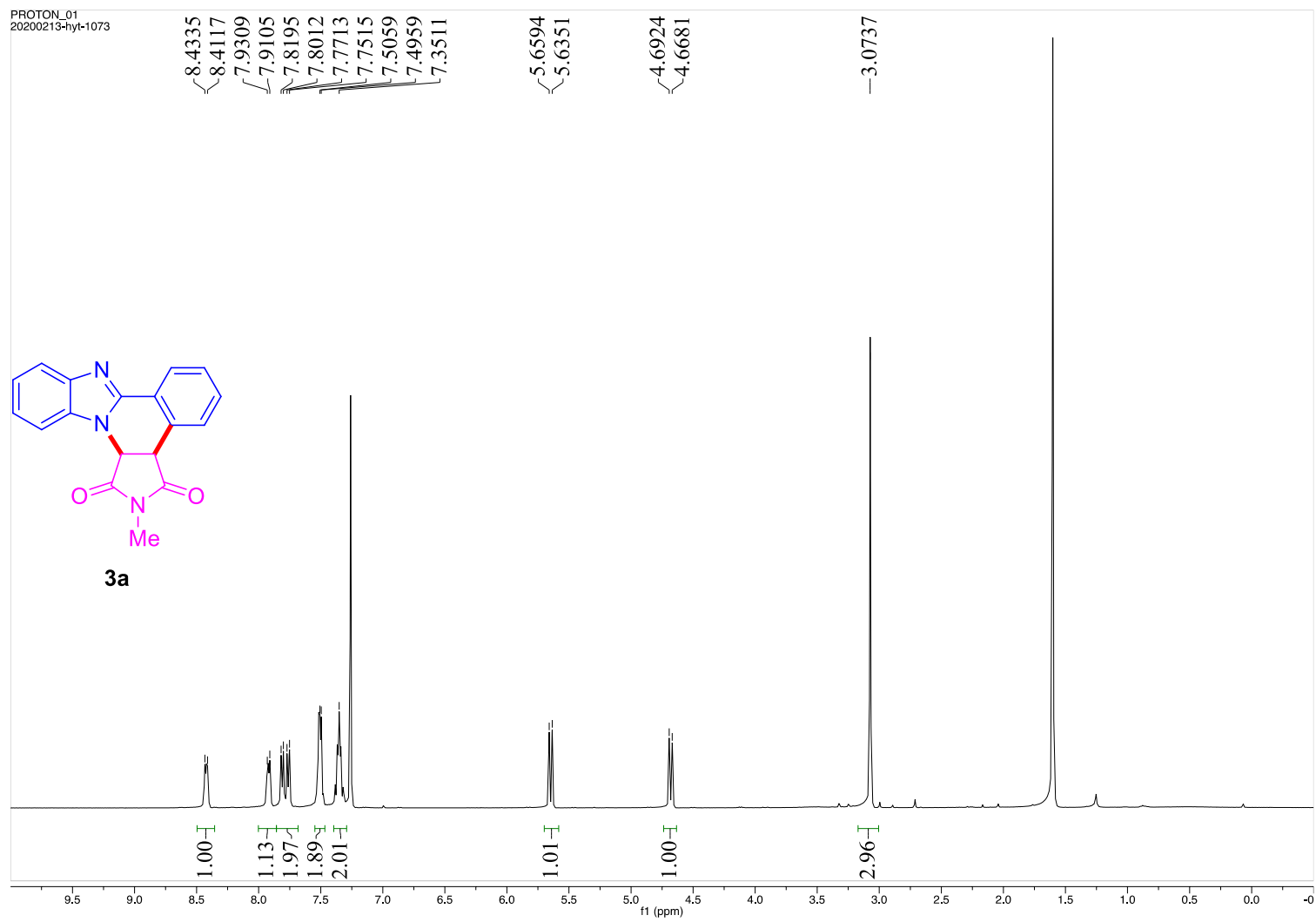
**Parallel reactions:** Two reactions were carried out. In the first oven-dried sealed tube 2-phenyl benzimidazole **1a** (0.05 g, 0.258 mmol) was added and in the second sealed tube **1a-d<sub>5</sub>** (0.05 g, 0.251 mmol) was added. After that *N*-methylmaleimide **2a** (0.057 g, 0.52 mmol),  $[\text{RhCp}^*\text{Cl}_2]_2$  (3.9 mg, 0.006 mmol), AgOAc (0.085 g, 0.515 mmol) and 1,2-dichloroethane (3 mL) were added in each of the sealed tube. The reaction mixture was heated at 100 °C for 1 h. The aliquots from both the reactions were taken at 10, 20, 30 and 40 min. and filtered through celite bed. The filtrate was concentrated *in vacuo*. The crude mixtures were analyzed by <sup>1</sup>H NMR spectroscopy.

Deuterated	
Time(min)	Ratio
10	0.07
20	0.11
30	0.16
40	0.19

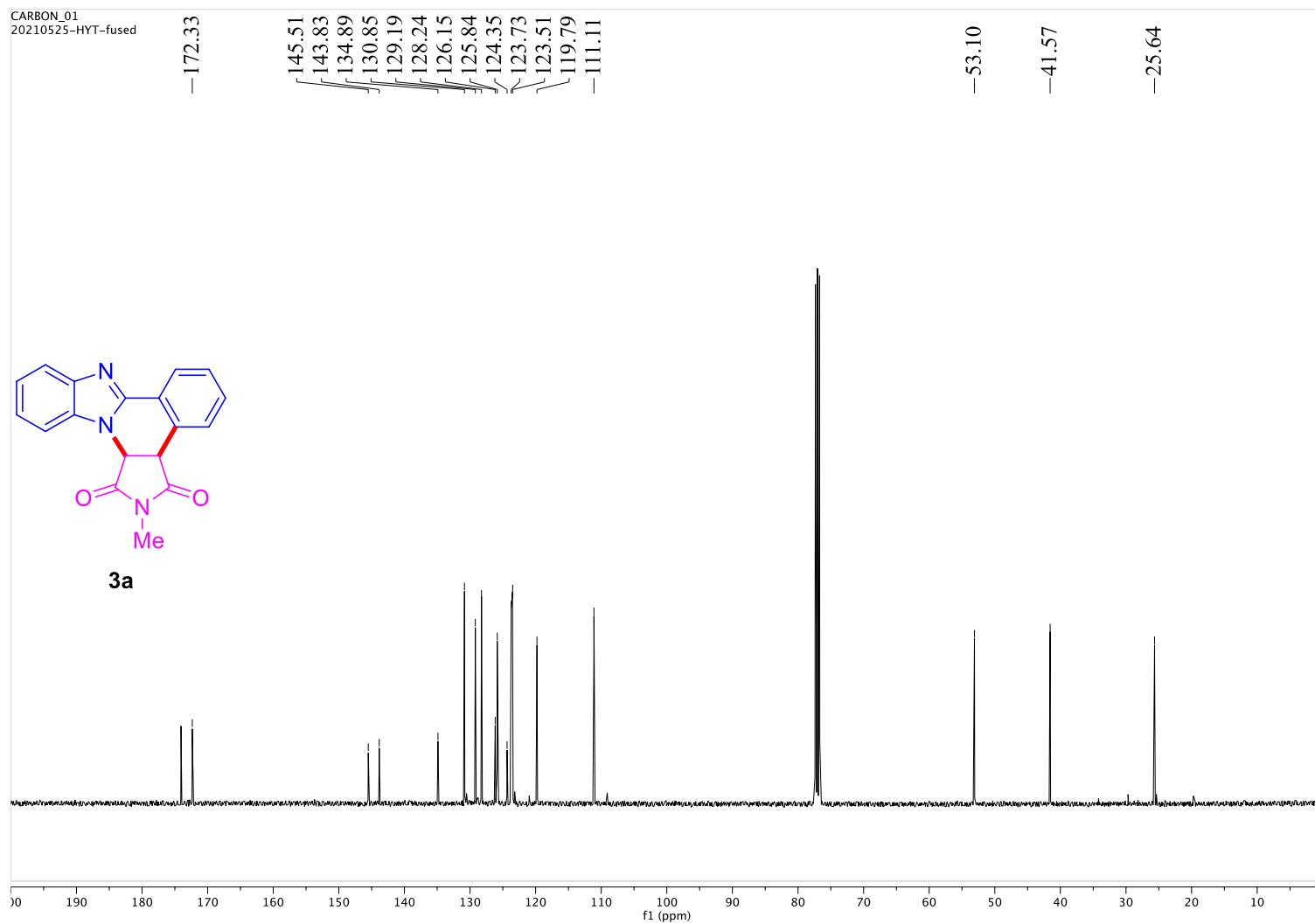
Non-deuterated	
Time(min)	Ratio
10	0.08
20	0.12
30	0.18
40	0.22

## KIE CALCULATION

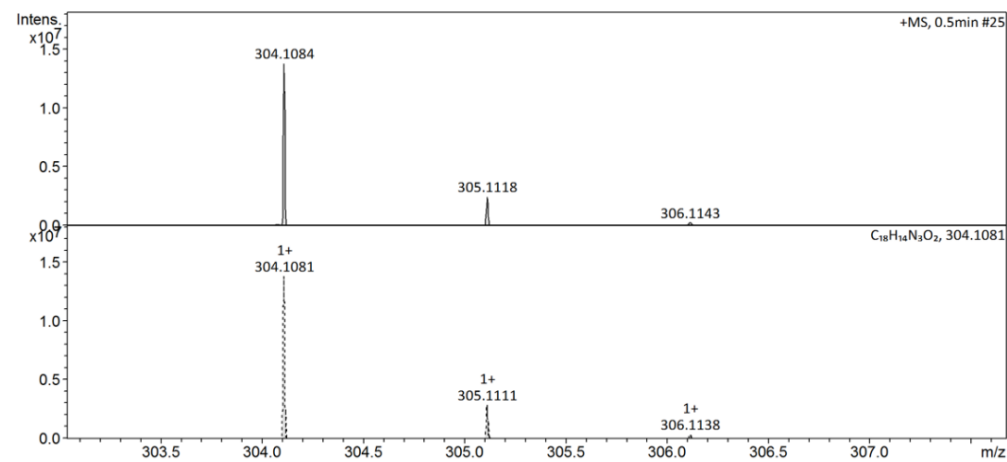
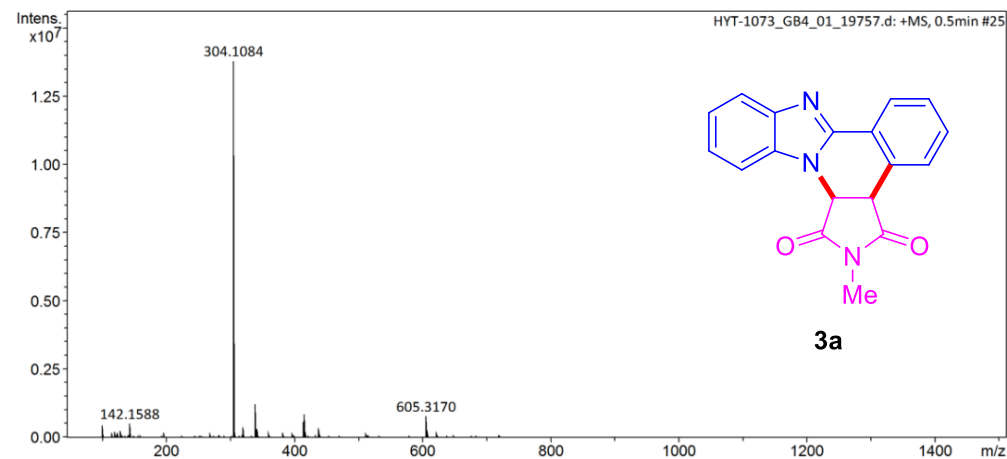




$^1\text{H}$  NMR spectrum (400 MHz) of compound **3a** in  $\text{CDCl}_3$ .



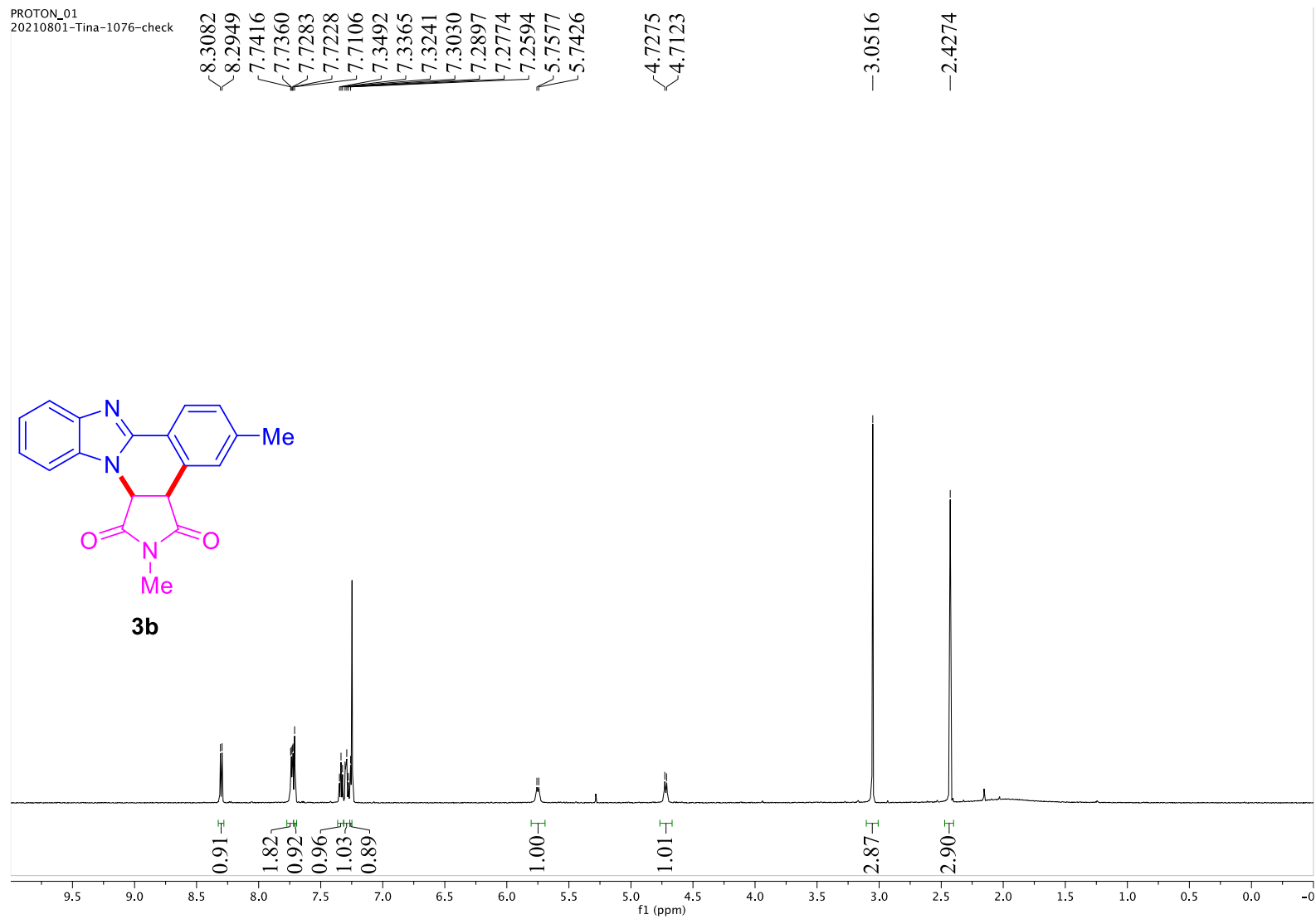
<sup>13</sup>C NMR spectrum (100 MHz) of compound **3a** in CDCl<sub>3</sub>.



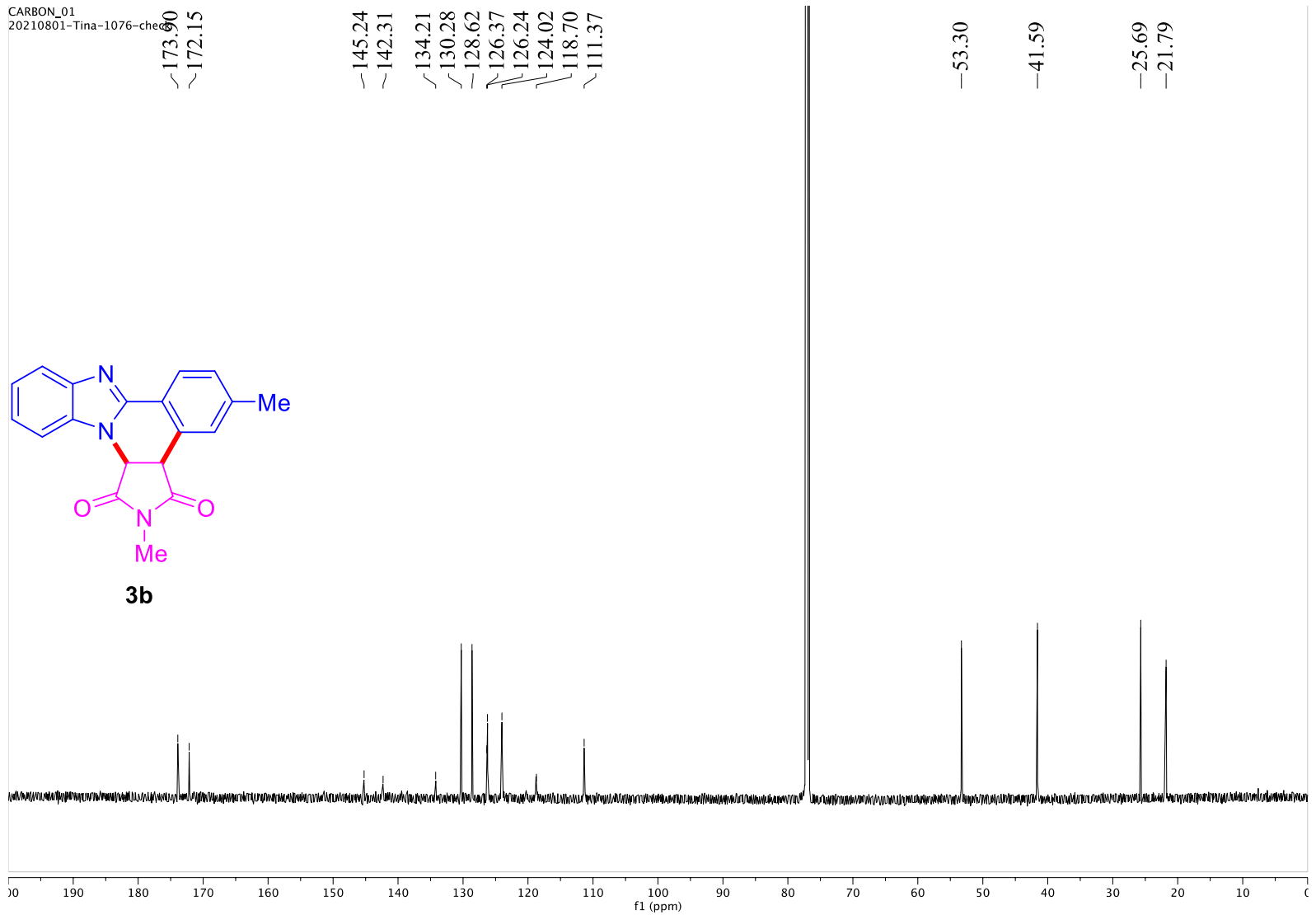
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
304.1084	1	C <sub>18</sub> H <sub>14</sub> N <sub>3</sub> O <sub>2</sub>	304.1081	-1.1	17.1	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **3a**.

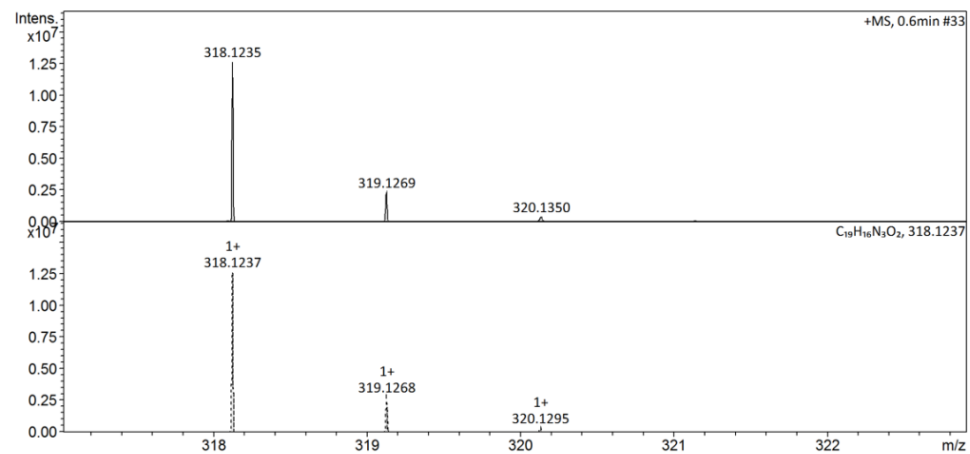
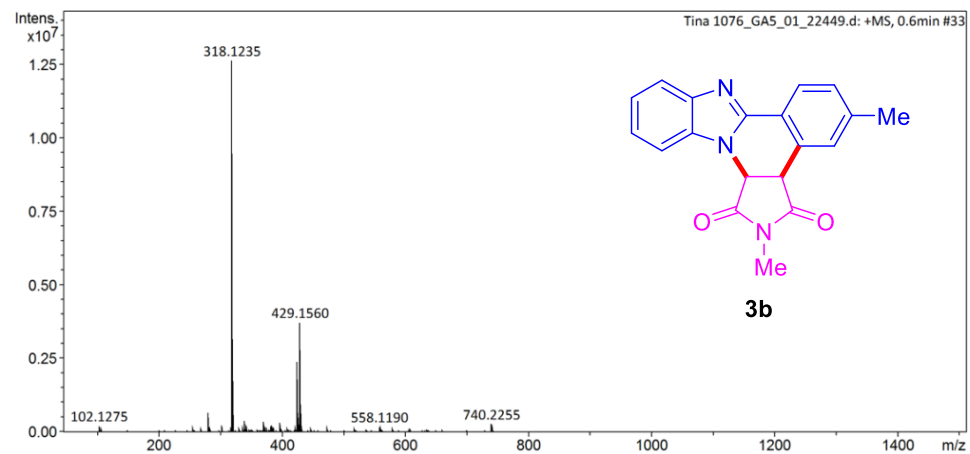


<sup>1</sup>H NMR spectrum (400 MHz) of compound **3b** in CDCl<sub>3</sub>.



$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3b** in  $\text{CDCl}_3$ .

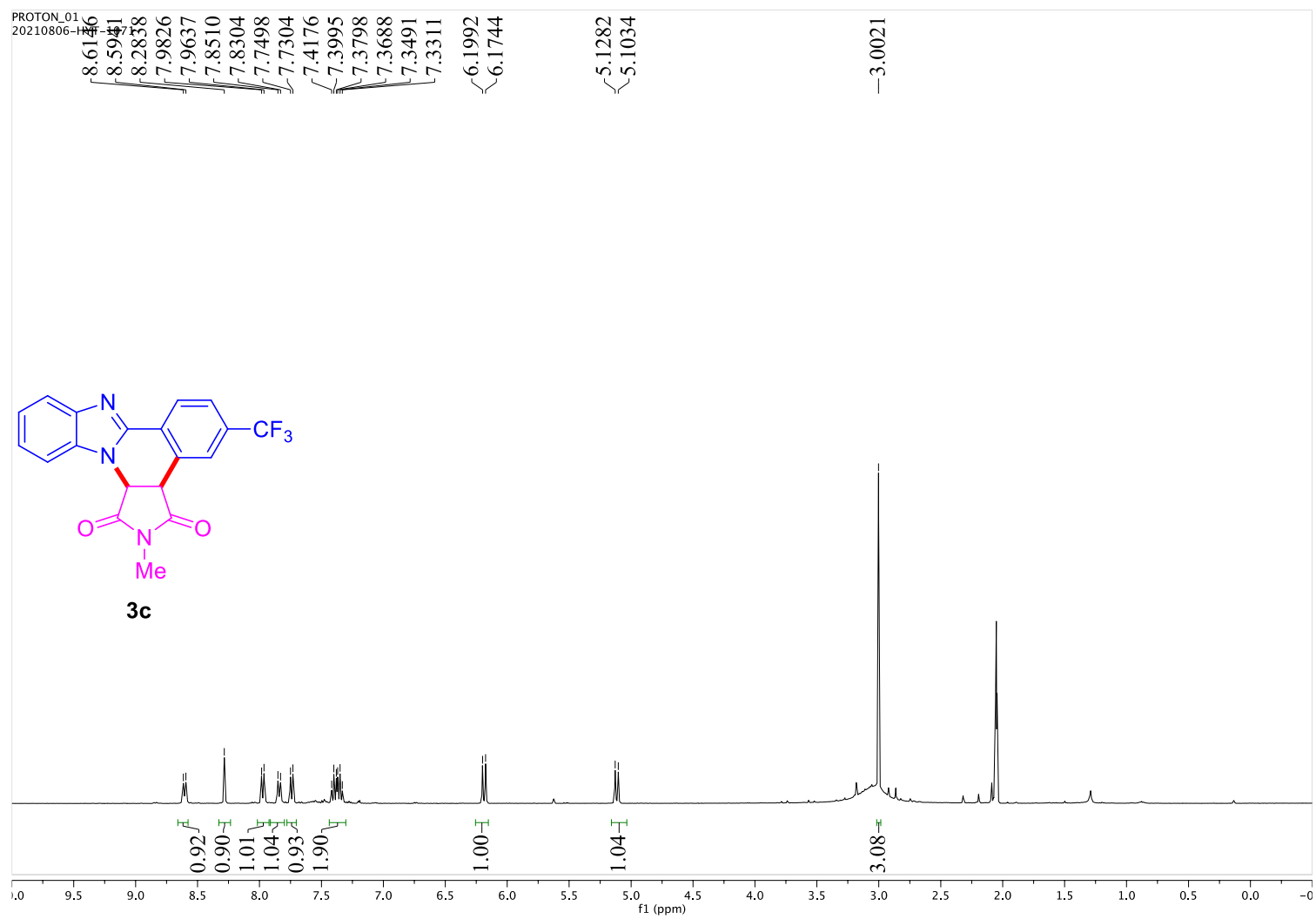




## Display Report

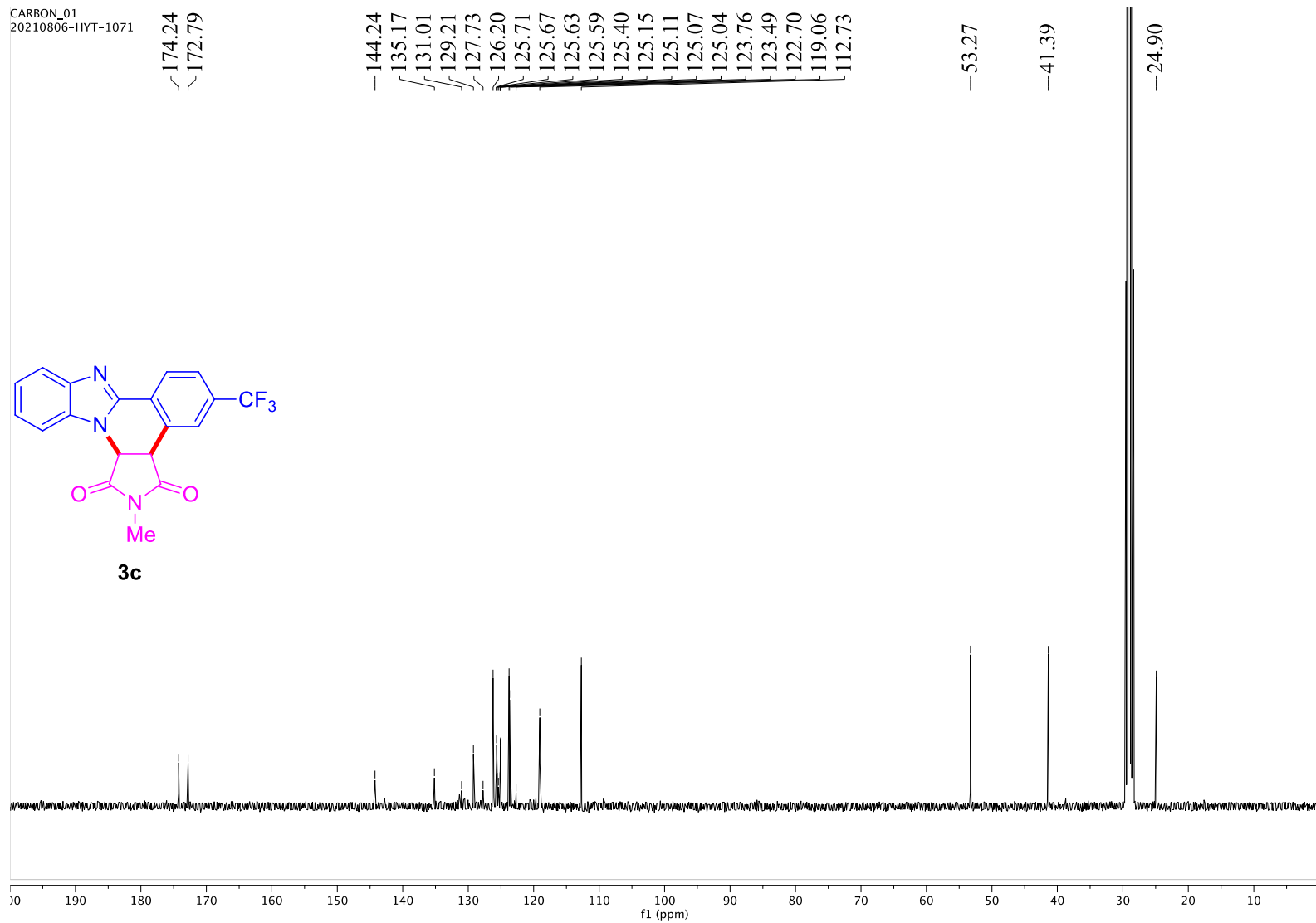
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
318.1235	1	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub>	318.1237	0.6	19.8	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **3b**.

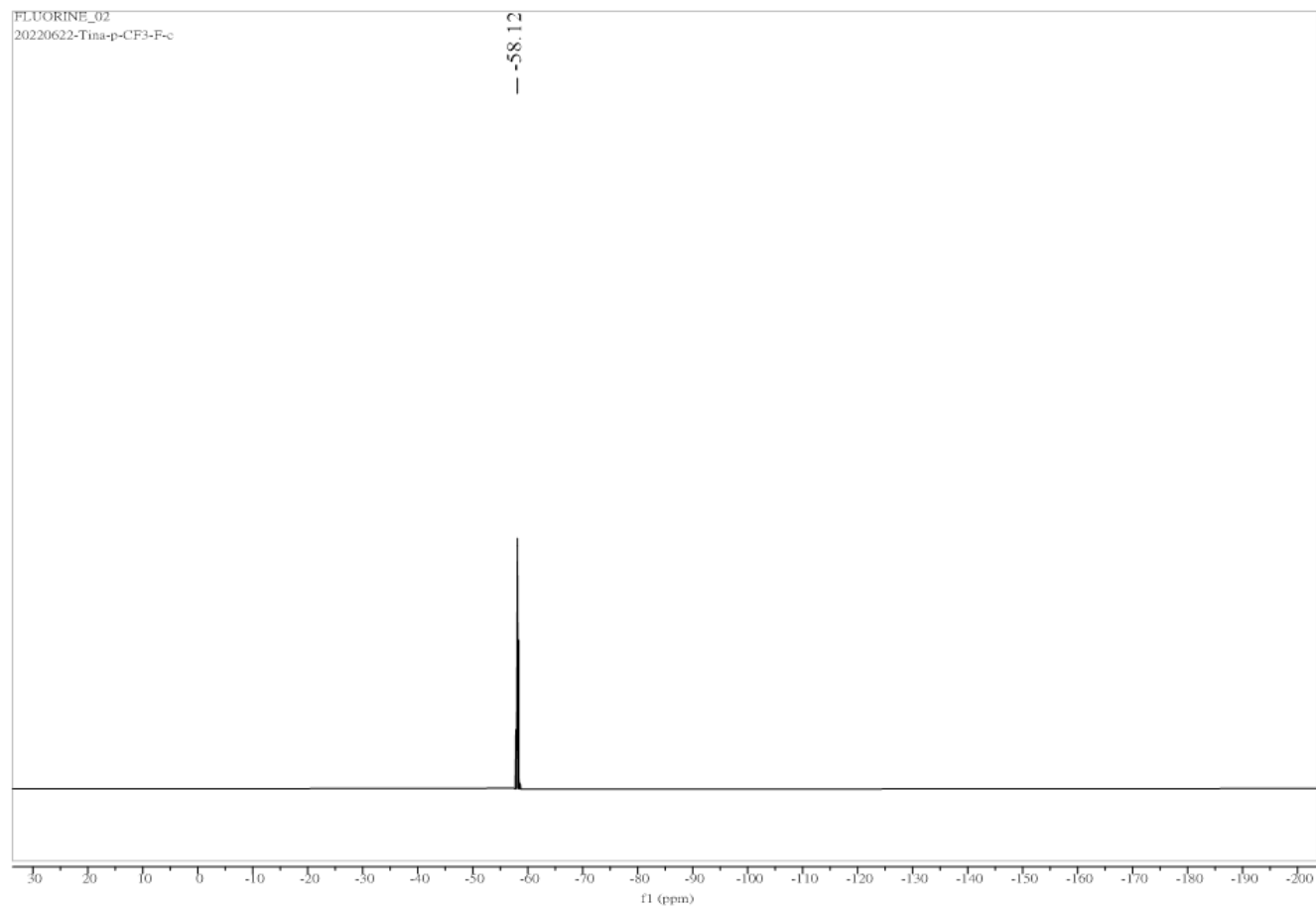


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3c** in acetone- $d_6$ .

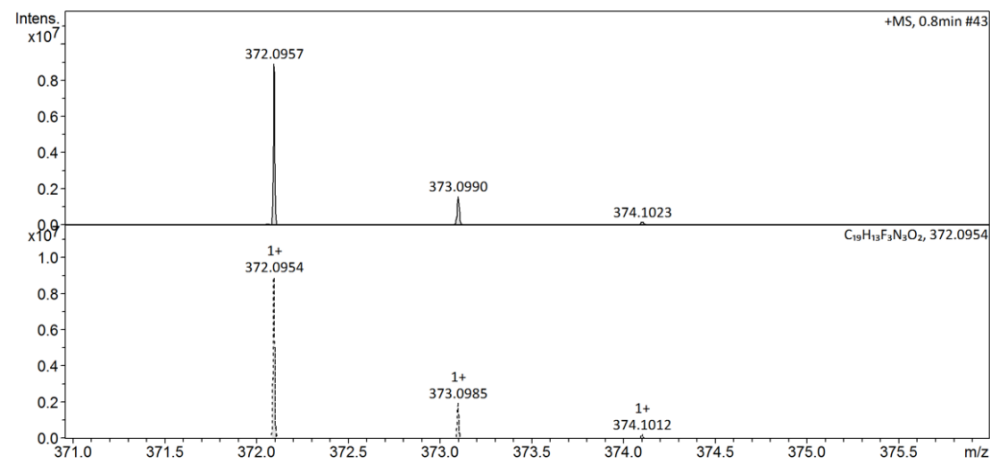
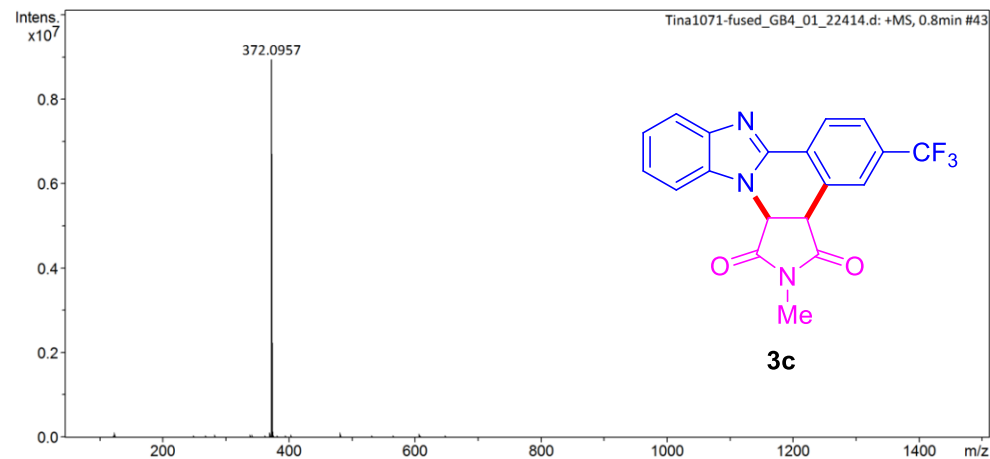
CARBON\_01  
20210806-HYT-1071



<sup>13</sup>C NMR spectrum (100 MHz) of compound **3c** in acetone-*d*<sub>6</sub>.



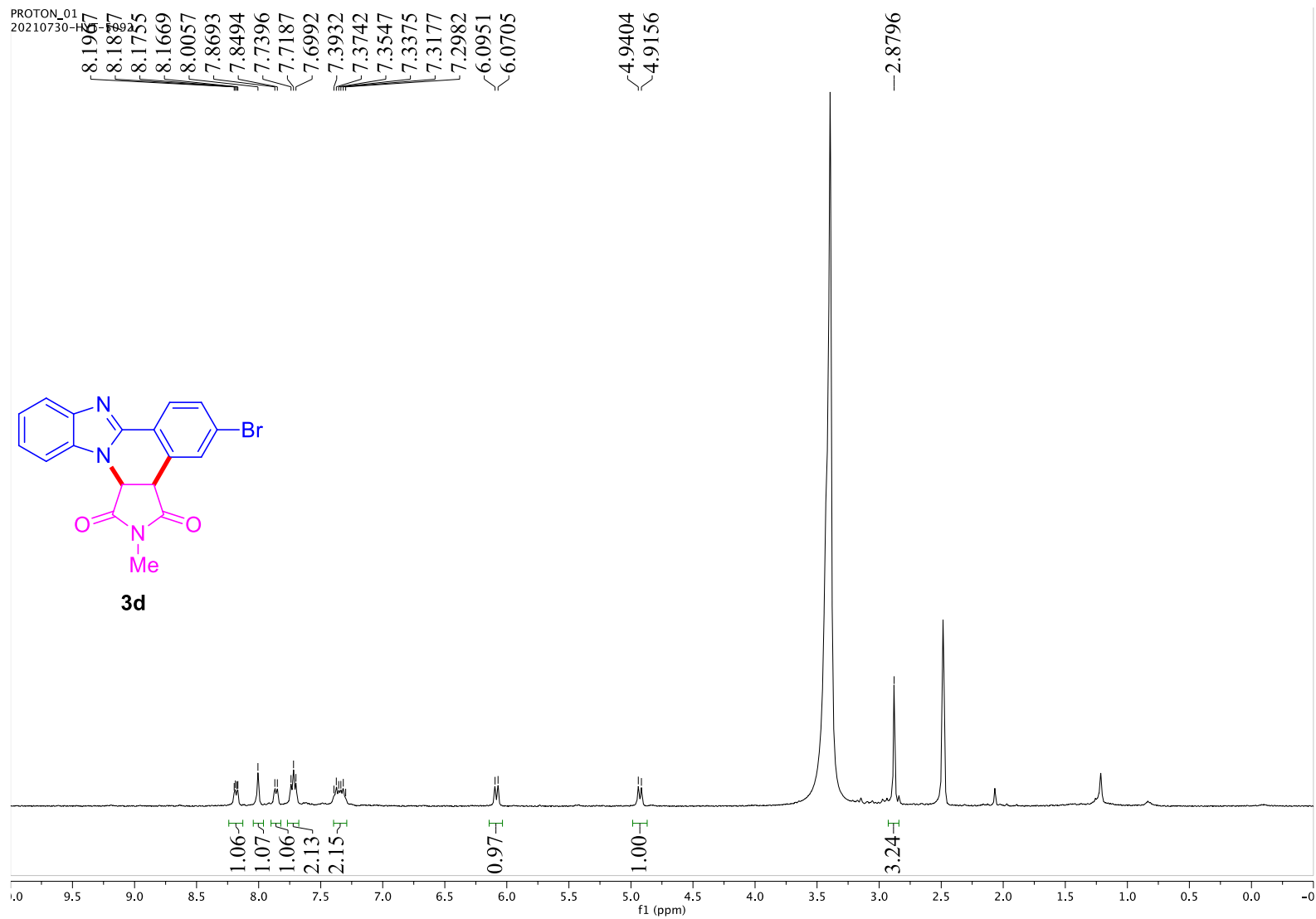
<sup>19</sup>F NMR spectrum (376 MHz) of compound **3c** in CDCl<sub>3</sub>.



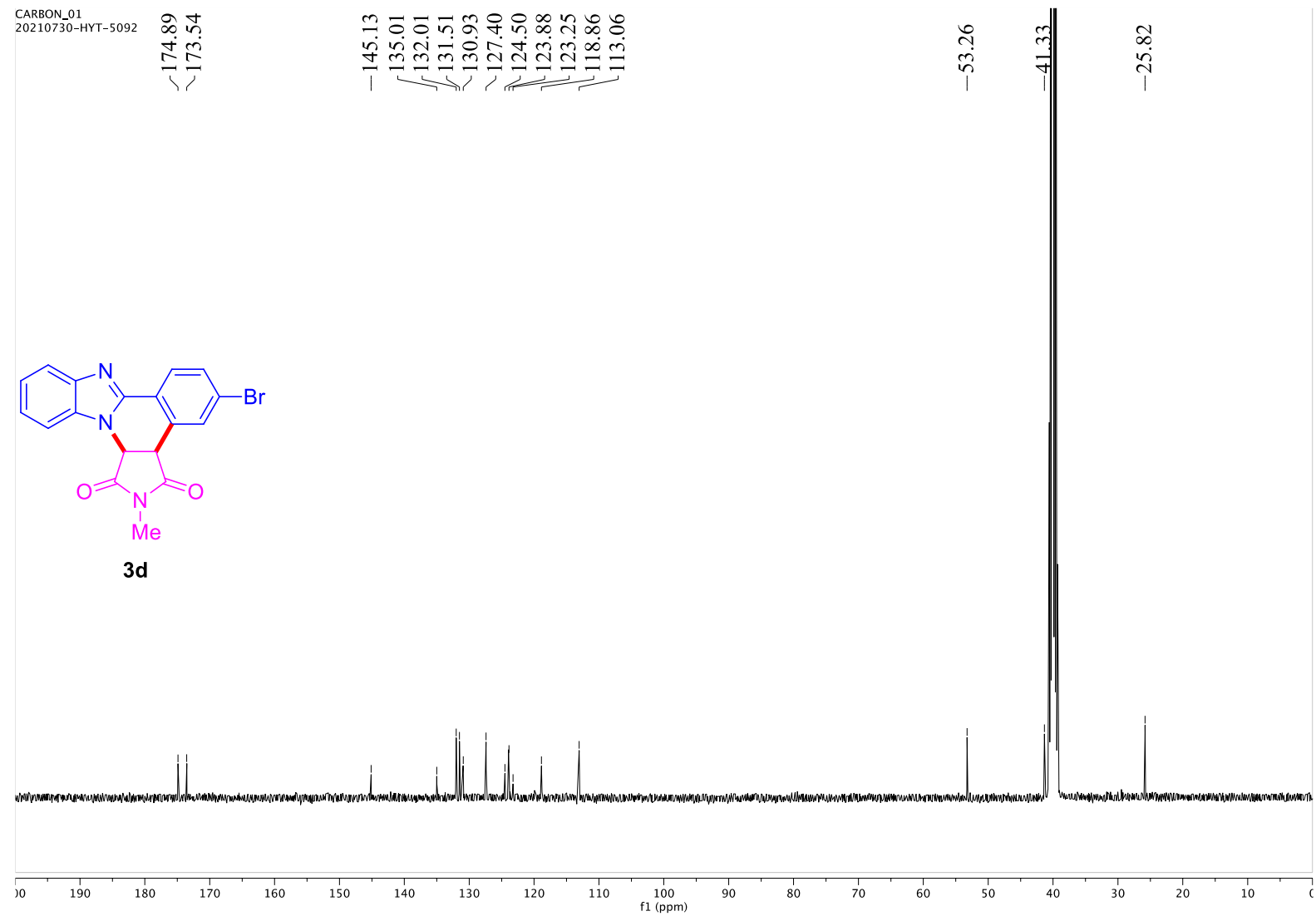
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
372.0957	1	C <sub>19</sub> H <sub>13</sub> F <sub>3</sub> N <sub>3</sub> O <sub>2</sub>	372.0954	0.8	23.2	1	100.00	13.5	even	ok	M+H

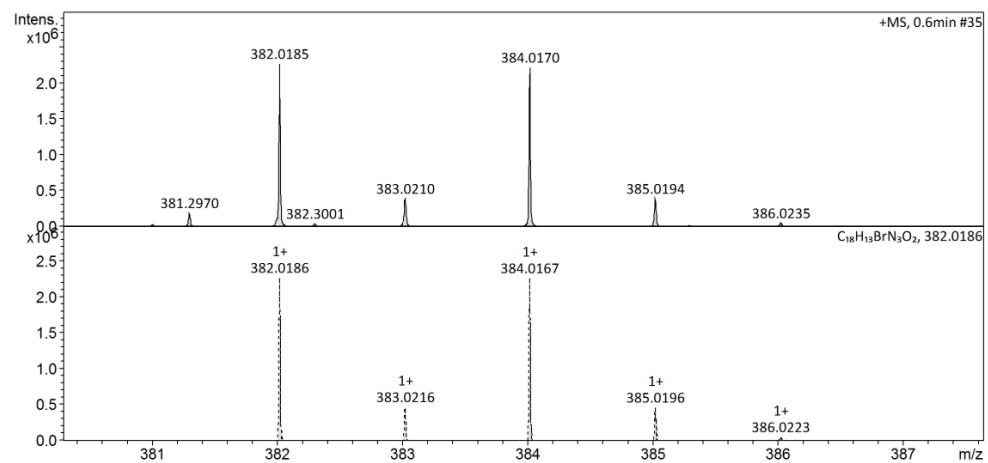
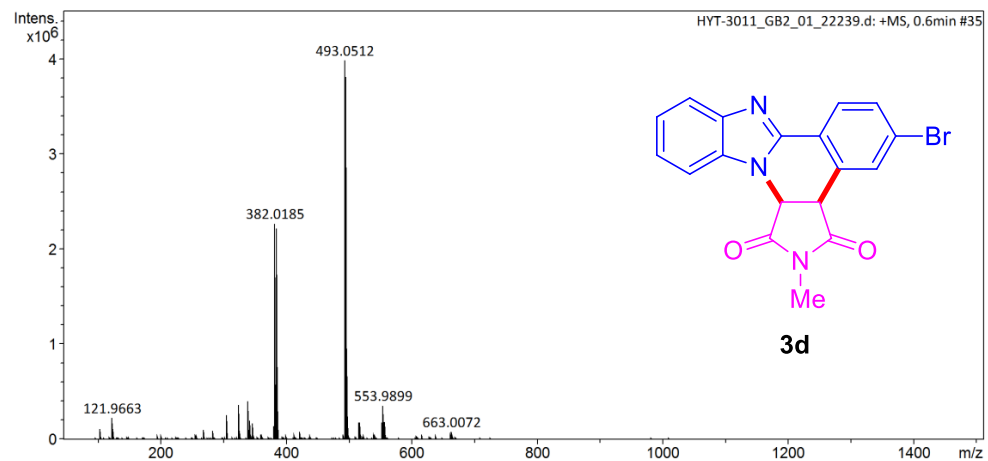
HRMS (ESI) of compound **3c**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3d** in  $\text{DMSO-}d_6$ .



$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3d** in  $\text{DMSO-}d_6$ .

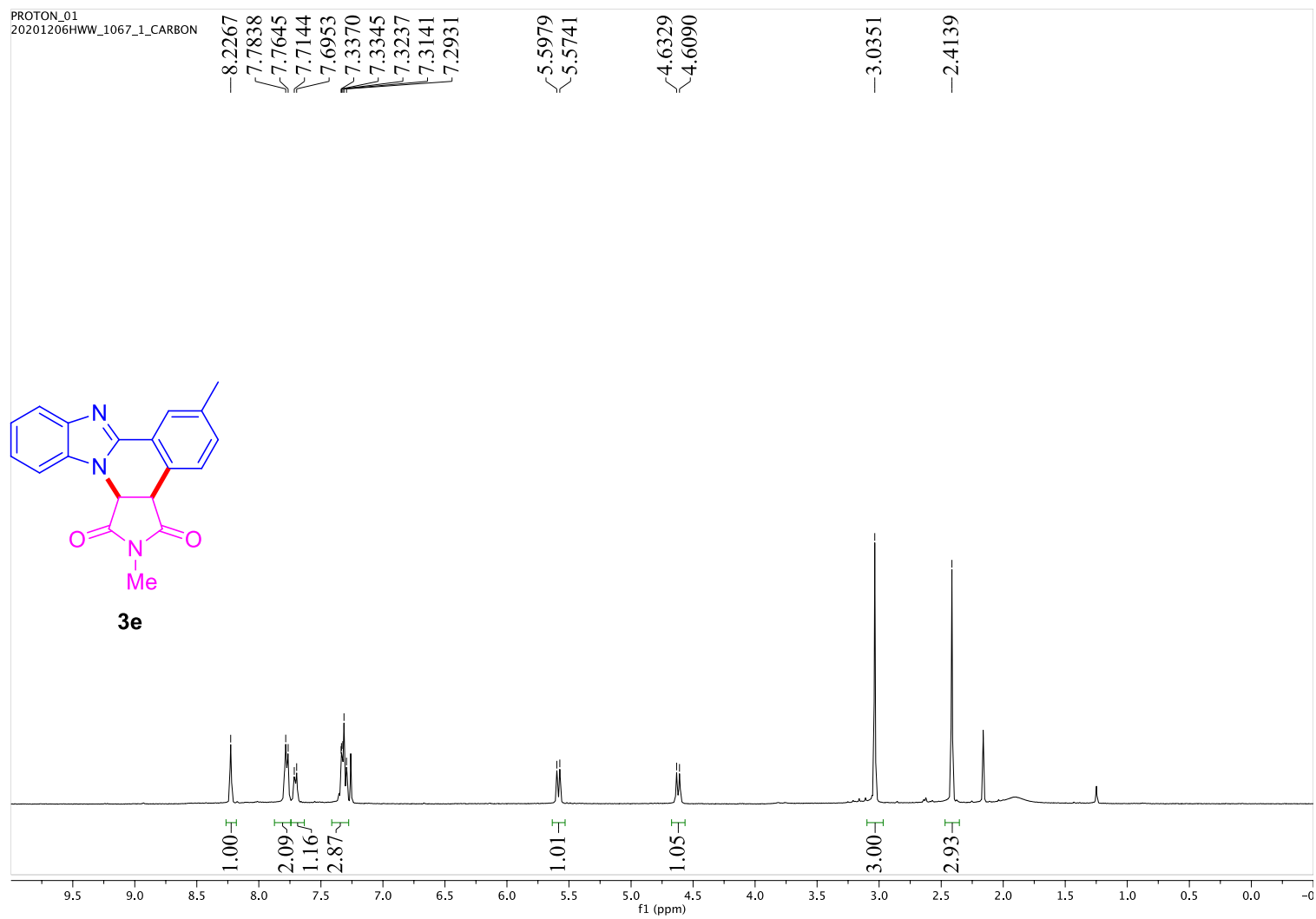


## Display Report

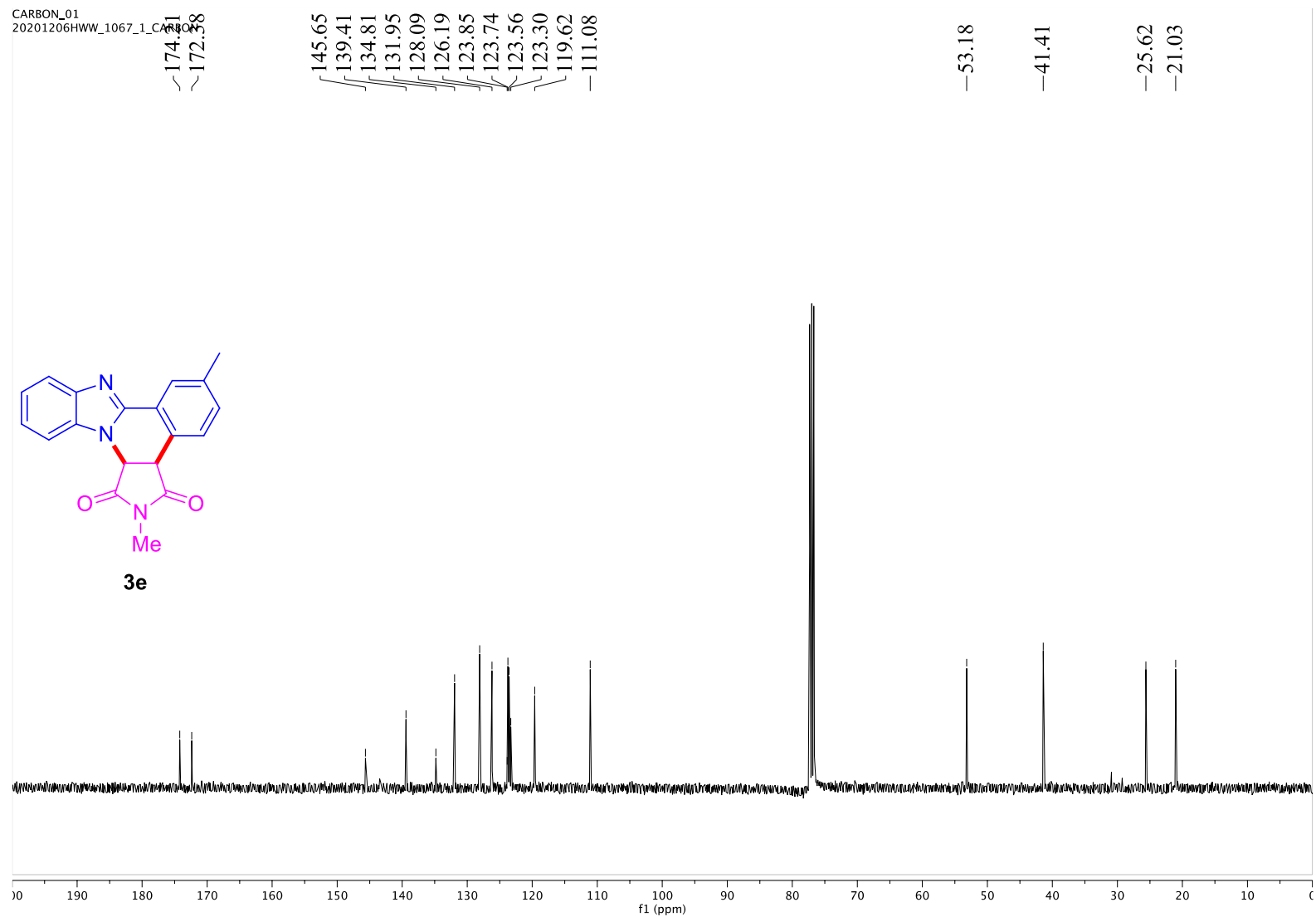
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
382.0185	1	C <sub>18</sub> H <sub>13</sub> BrN <sub>3</sub> O <sub>2</sub>	382.0186	-0.2	22.9	1	100.00	13.5	even	ok	M+H

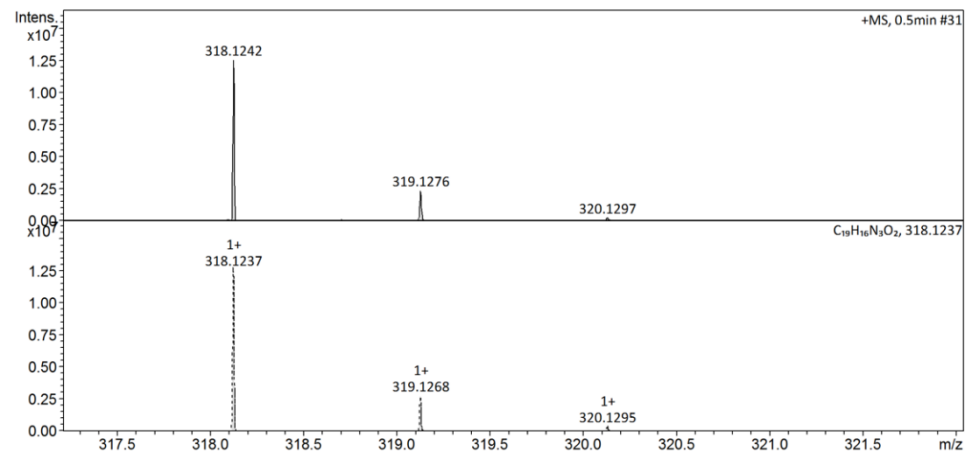
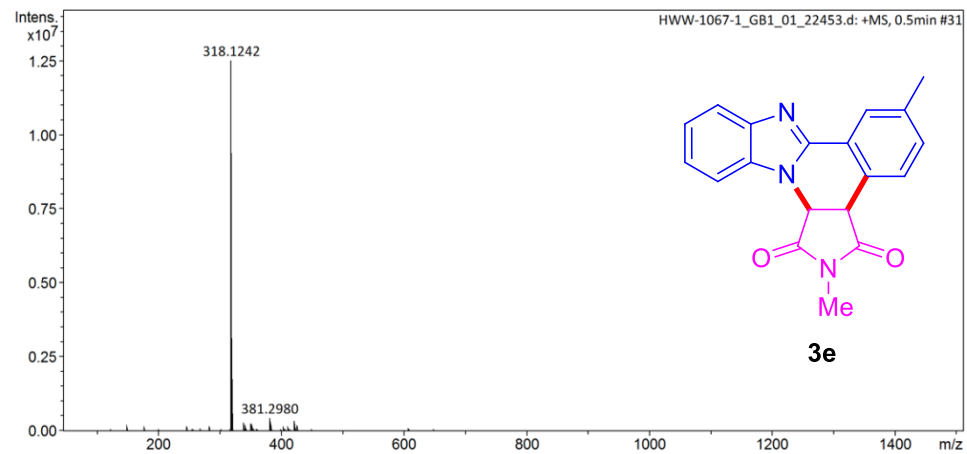
HRMS (ESI) of compound **3d**.





$^1\text{H}$  NMR spectrum (400 MHz) of compound **3e** in  $\text{CDCl}_3$ .

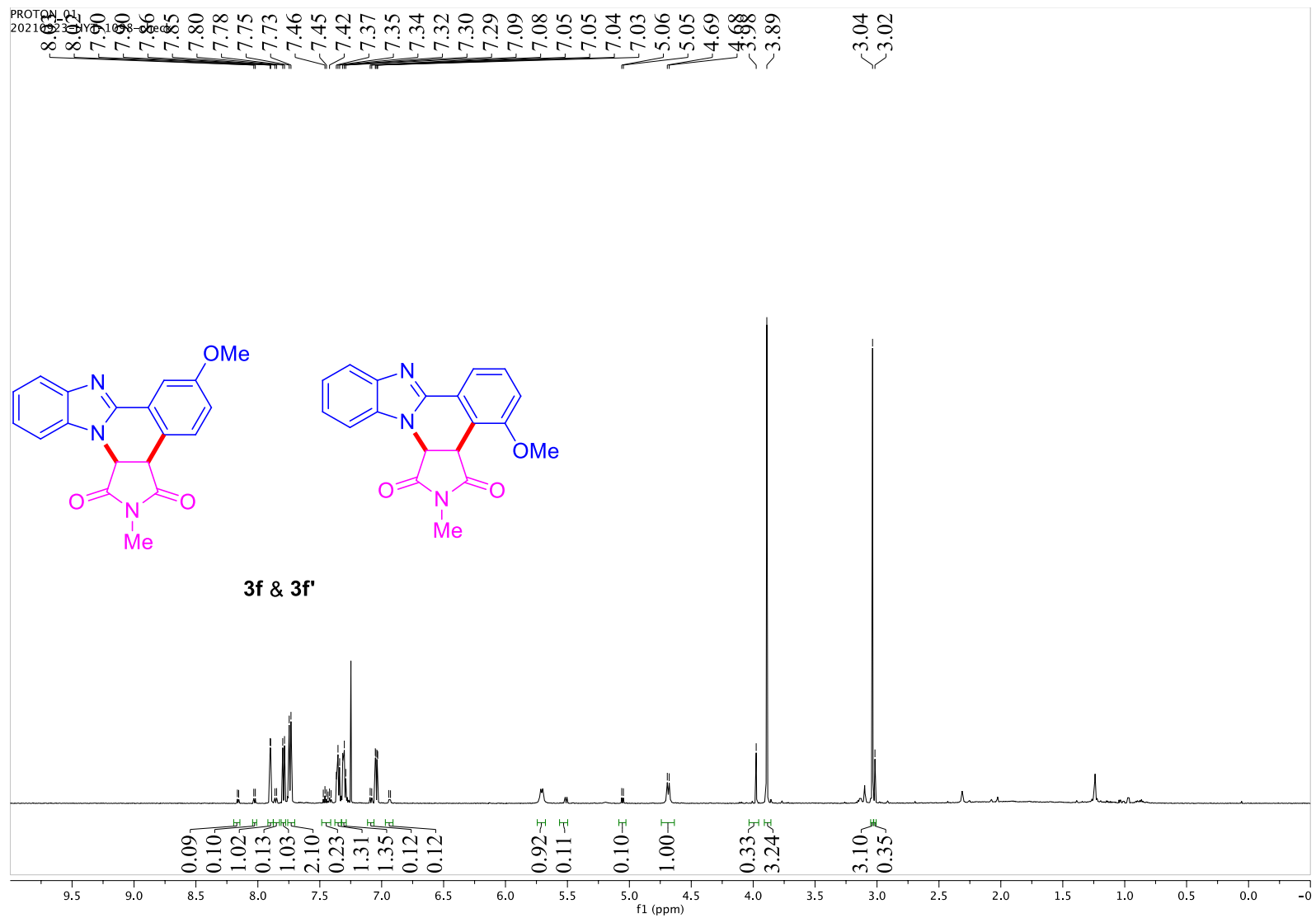




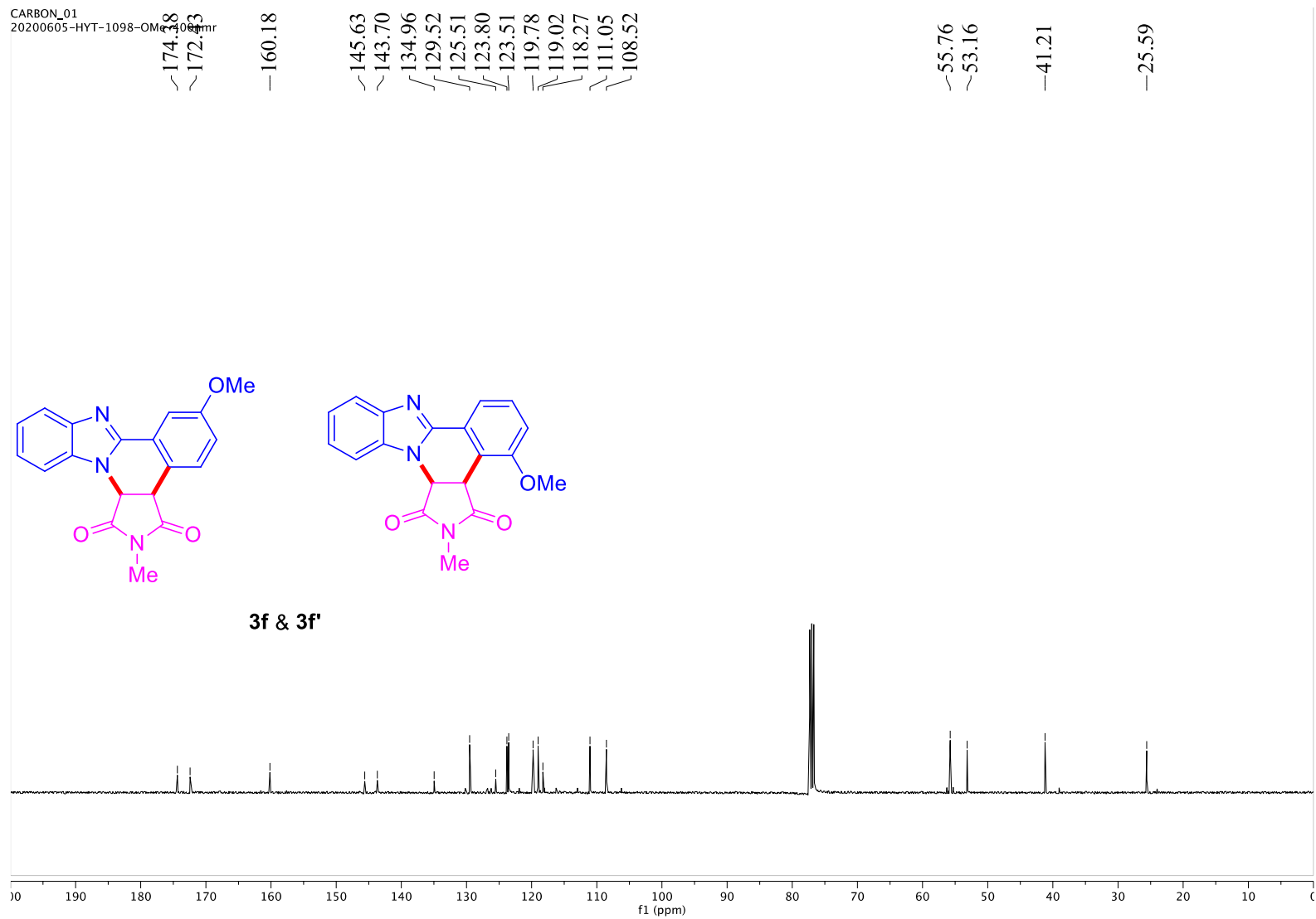
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
318.1242	1	C <sub>19</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub>	318.1237	-1.5	19.7	1	100.00	13.5	even	ok	M+H

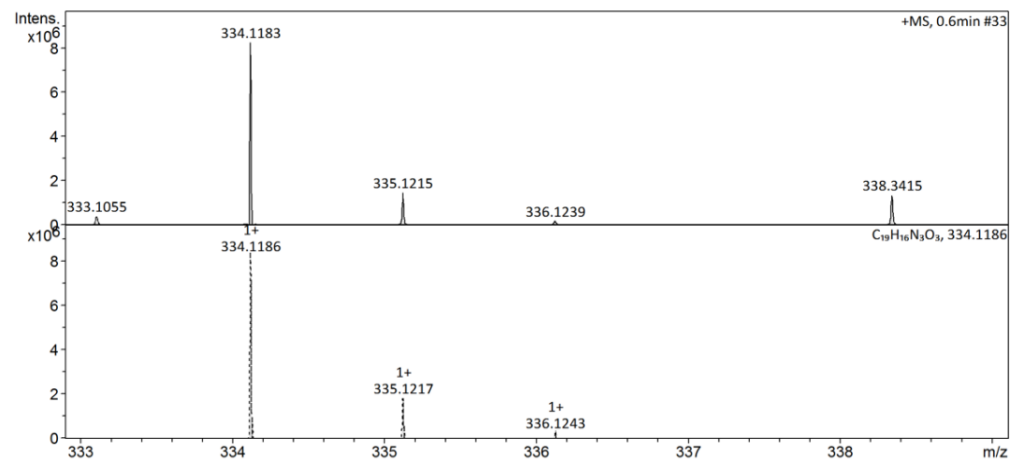
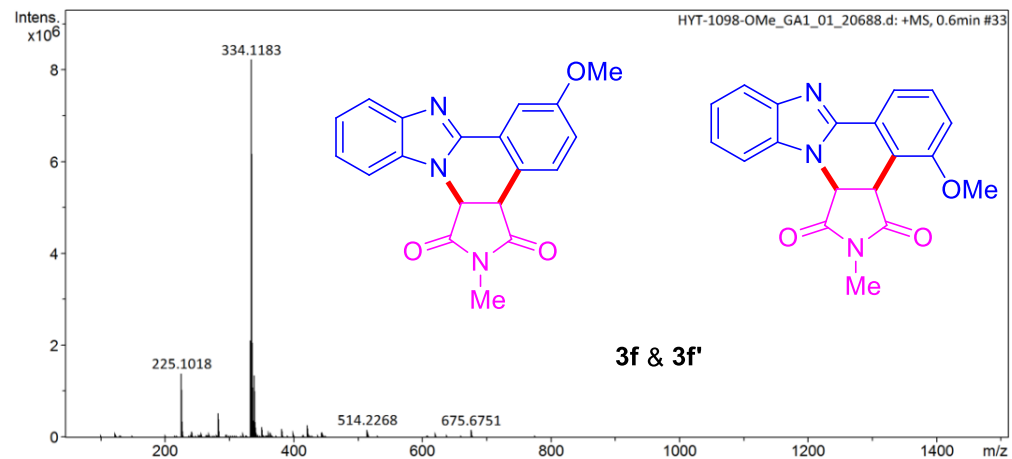
HRMS (ESI) of compound **3e**.



<sup>1</sup>H NMR spectrum (400 MHz) of compound **3f** & **3f'** in CDCl<sub>3</sub>.



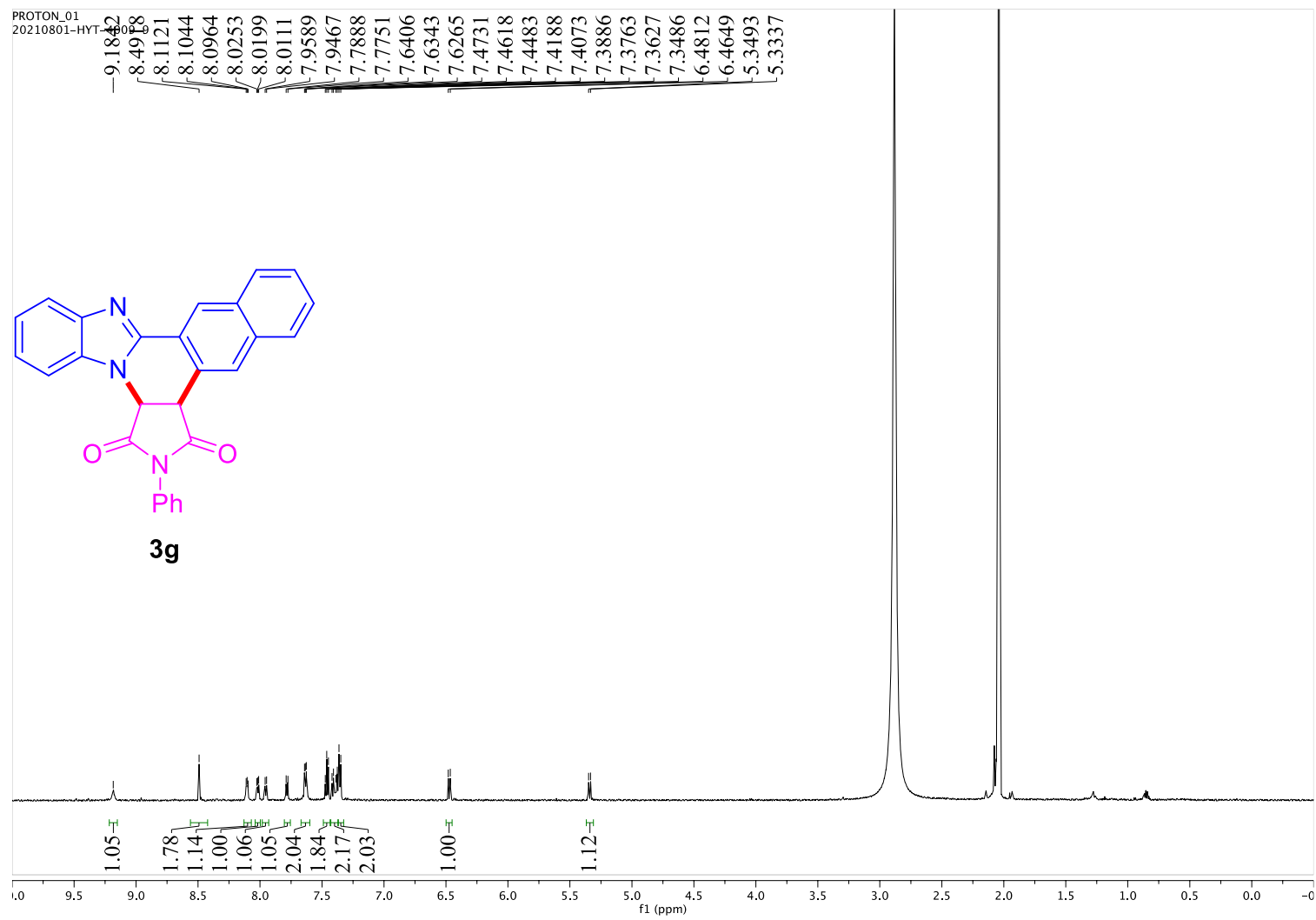
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3f** & **3f'** in  $\text{CDCl}_3$ .



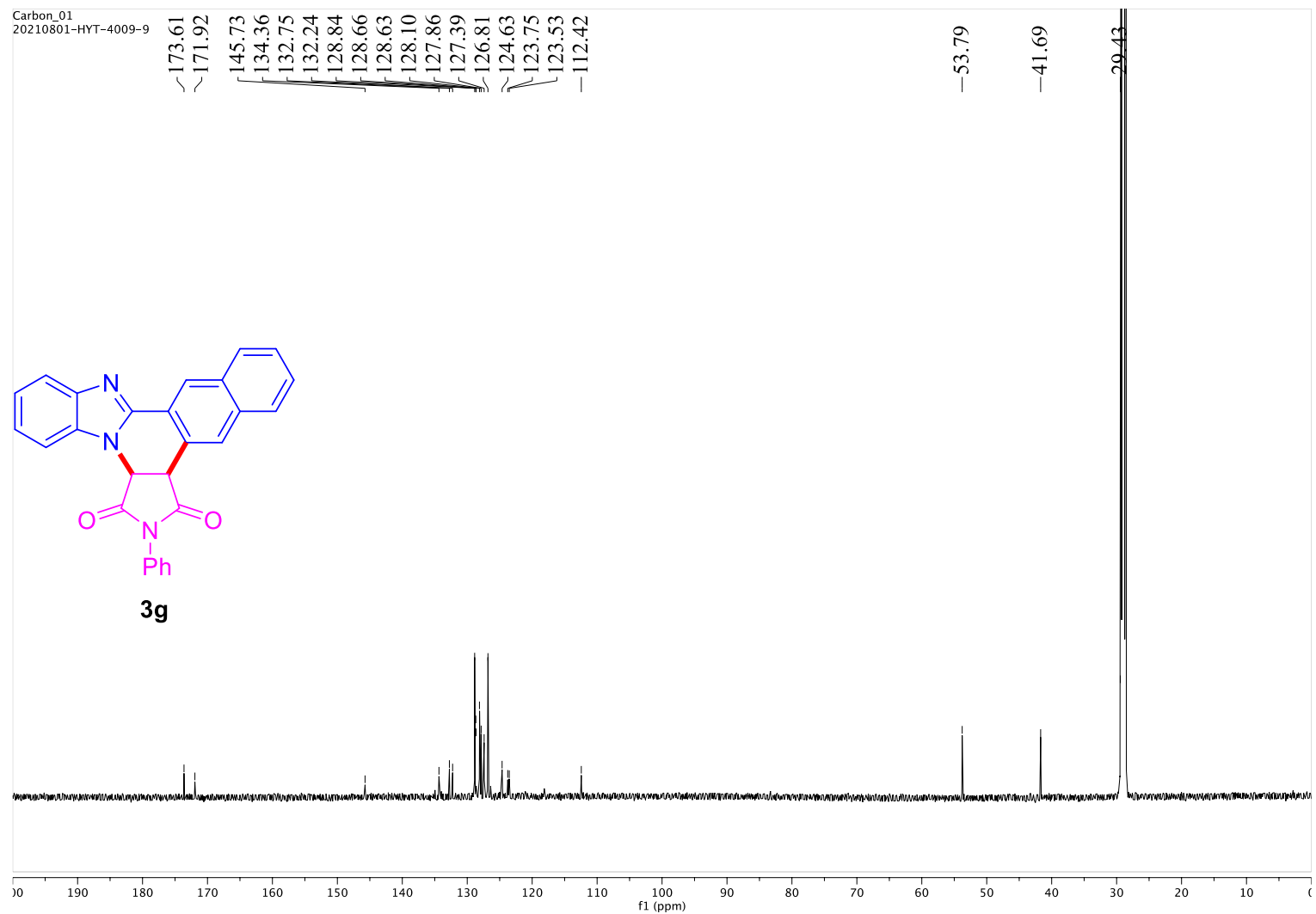
### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
334.1183	1	C <sub>19</sub> H <sub>16</sub> N <sub>3</sub> O <sub>3</sub>	334.1186	-0.9	25.9	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **3f & 3f'**.

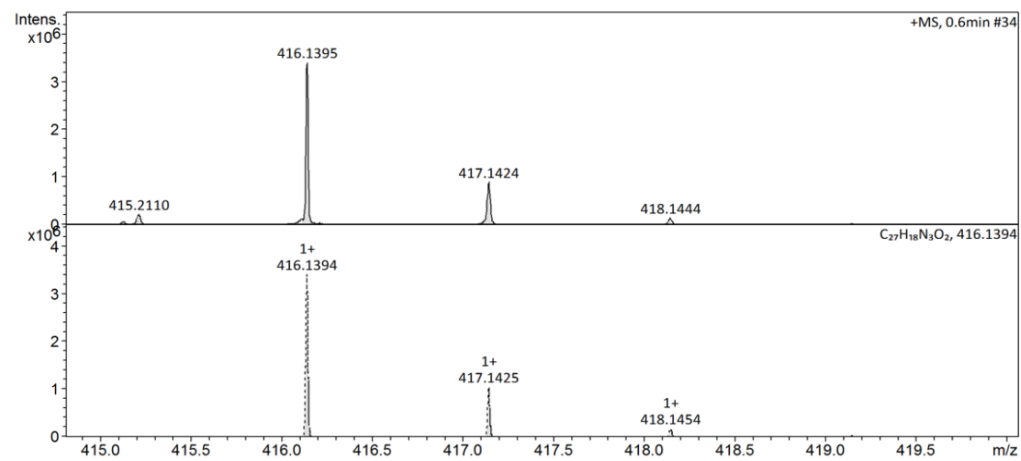
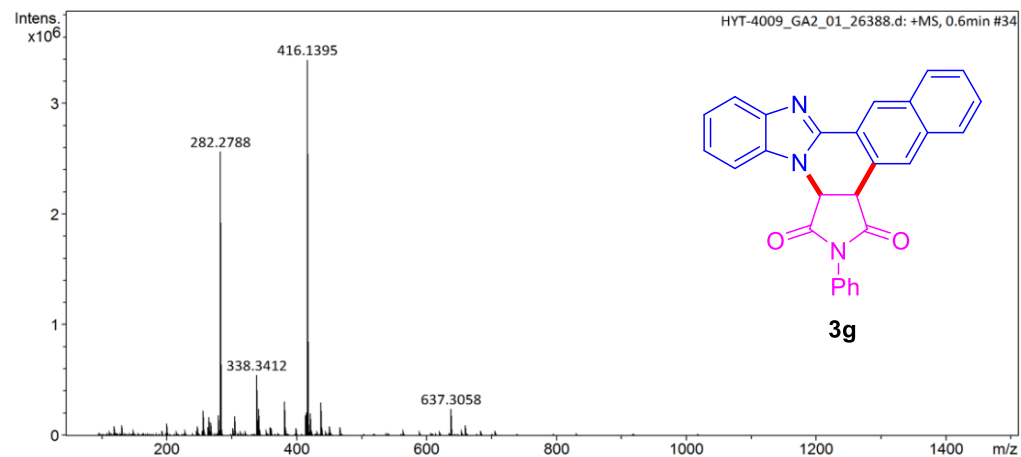


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3g** in  $\text{DMSO-}d_6$ .



$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3g** in  $\text{DMSO-}d_6$ .

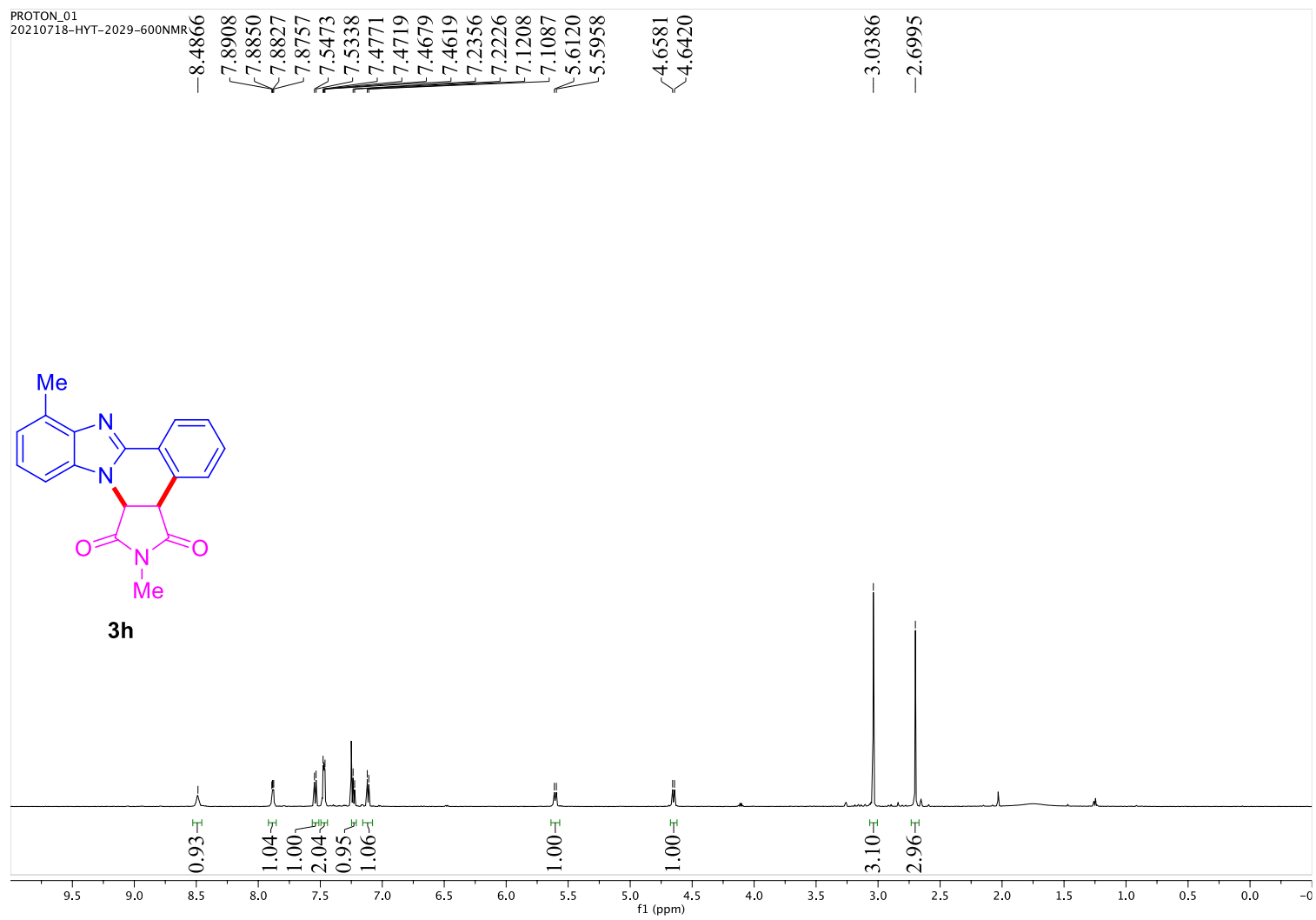




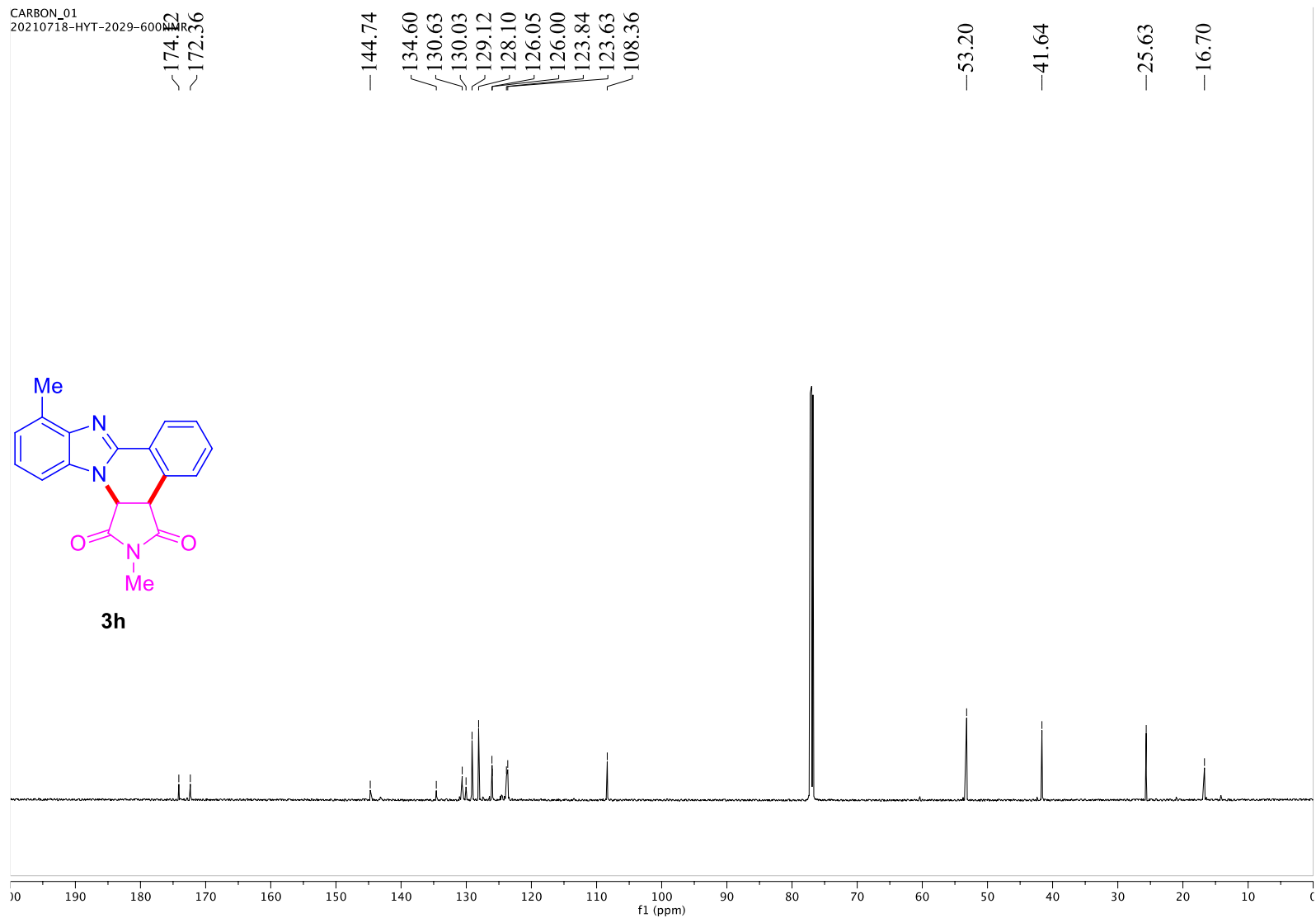
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
416.1395	1	C <sub>27</sub> H <sub>18</sub> N <sub>3</sub> O <sub>2</sub>	416.1394	0.3	23.6	1	100.00	20.5	even	ok	M

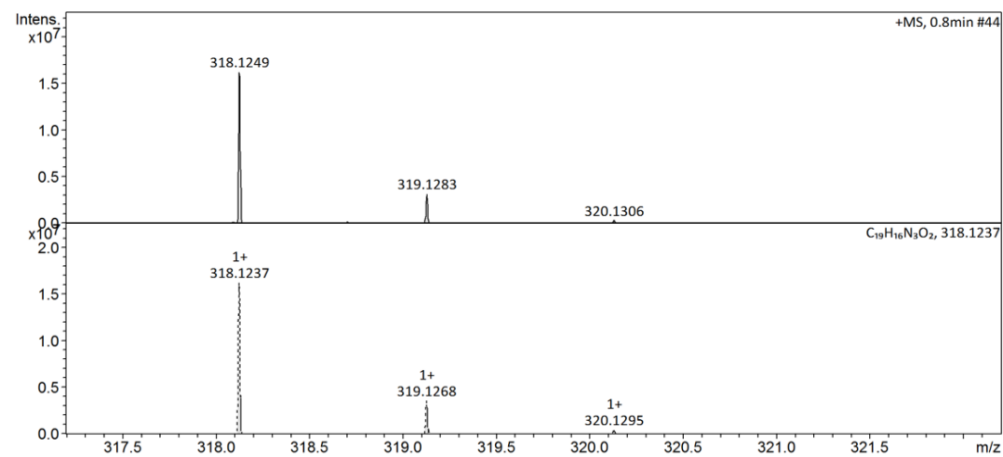
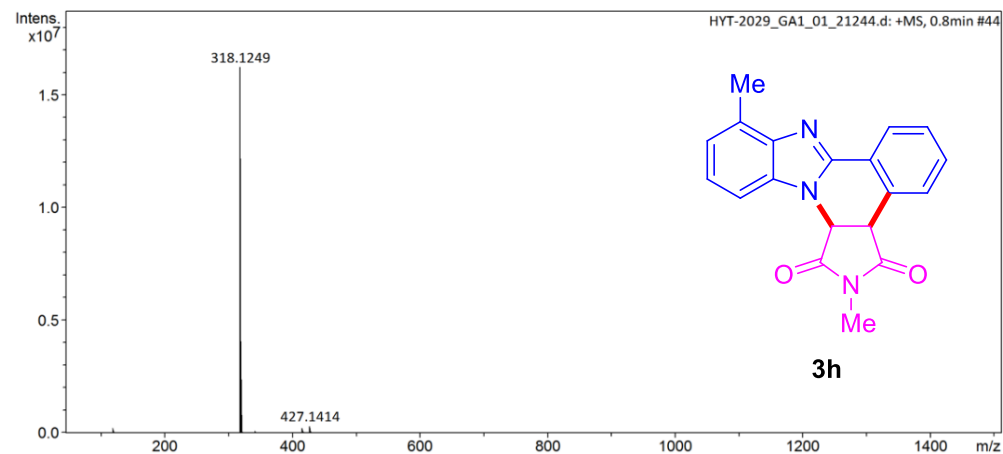
HRMS (ESI) of compound **3g**.



$^1\text{H}$  NMR spectrum (600 MHz) of compound **3h** in  $\text{CDCl}_3$ .



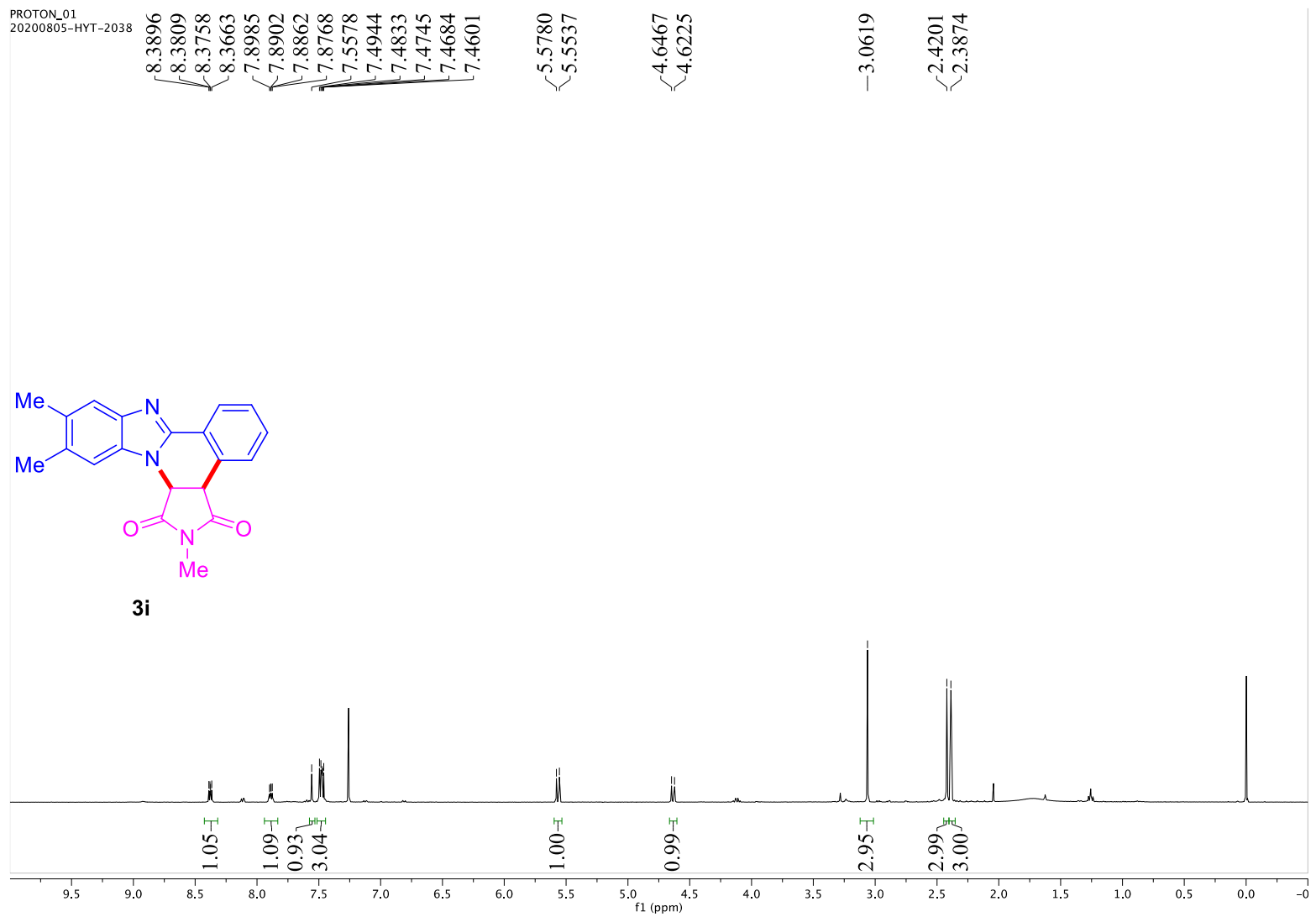
$^{13}\text{C}$  NMR spectrum (150 MHz) of compound **3h** in  $\text{CDCl}_3$ .



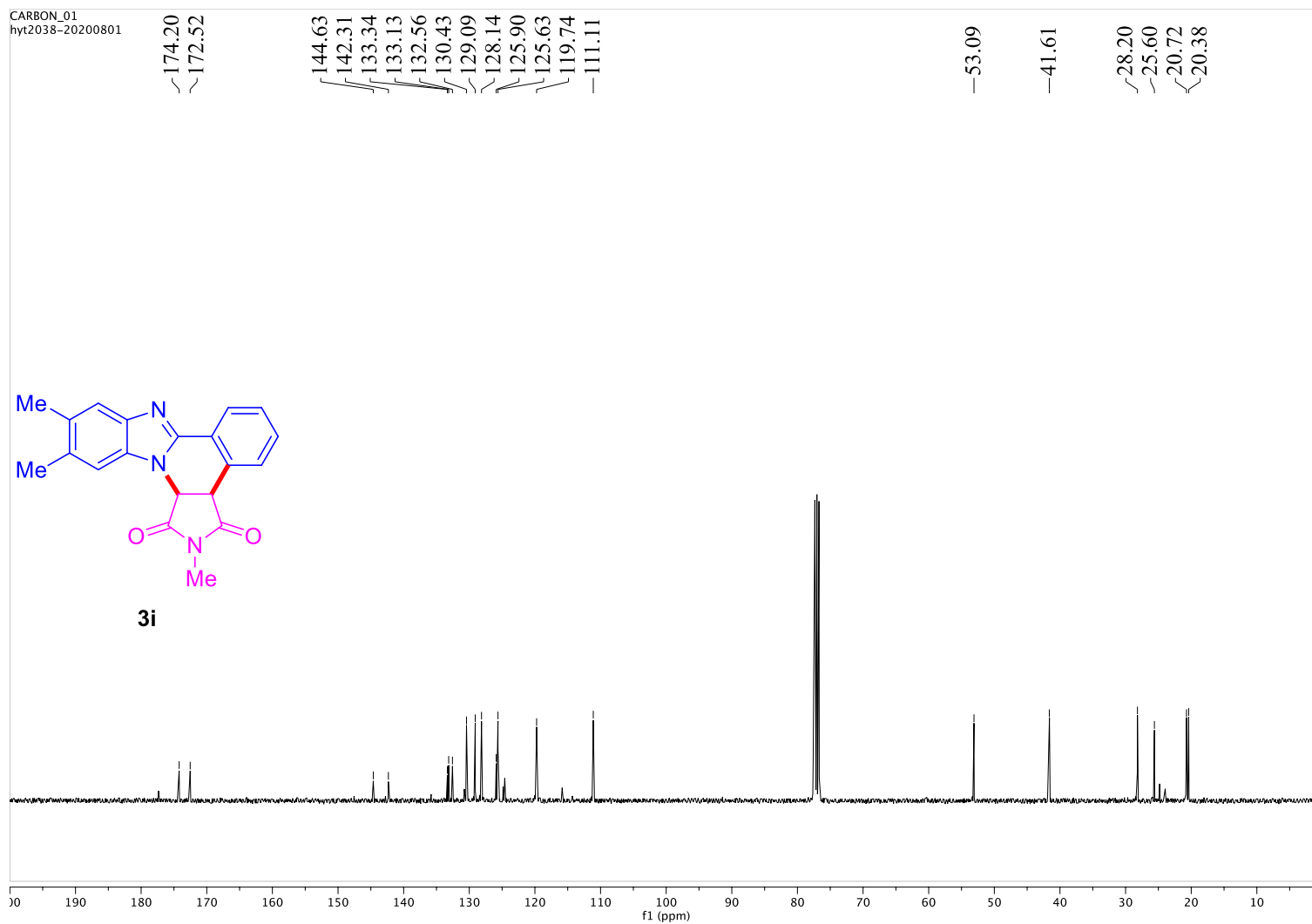
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
318.1249	1	C <sub>19</sub> H <sub>16</sub> N <sub>3</sub> O <sub>2</sub>	318.1237	3.9	15.1	1	100.00	13.5	even	ok	M+H

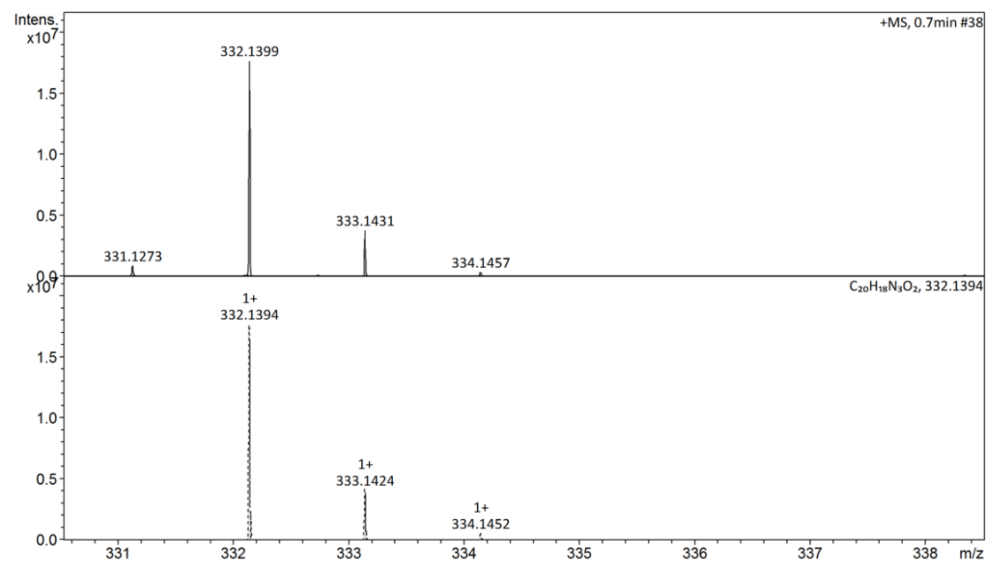
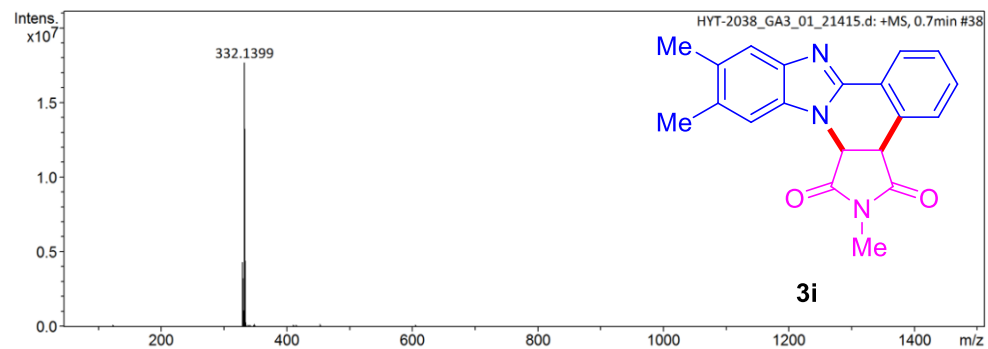
HRMS (ESI) of compound **3h**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3i** in  $\text{CDCl}_3$ .



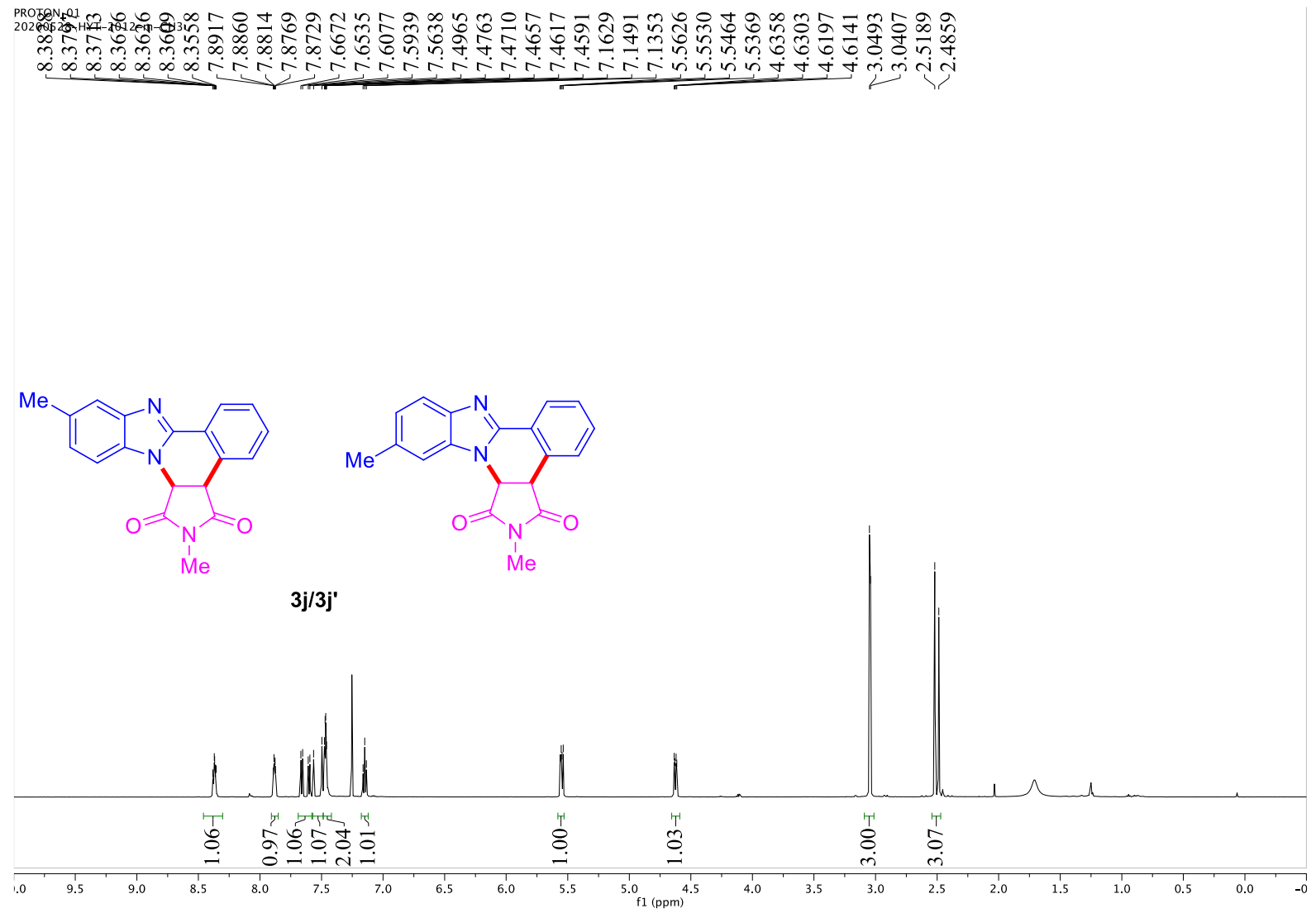
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3i** in  $\text{CDCl}_3$ .



## Display Report

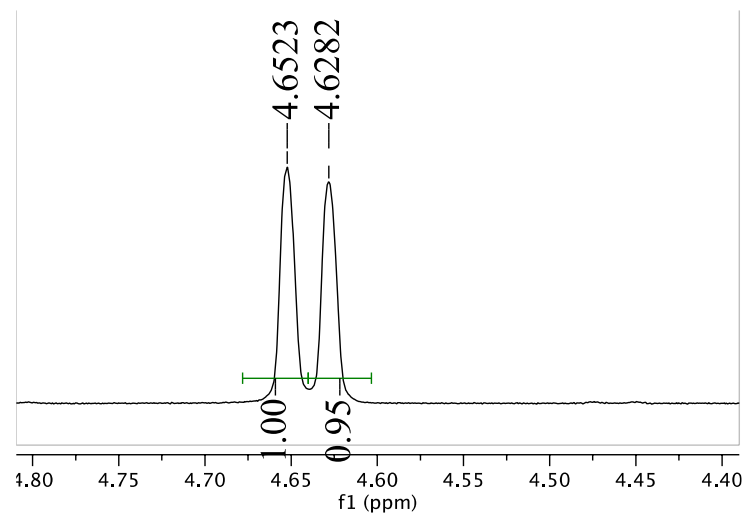
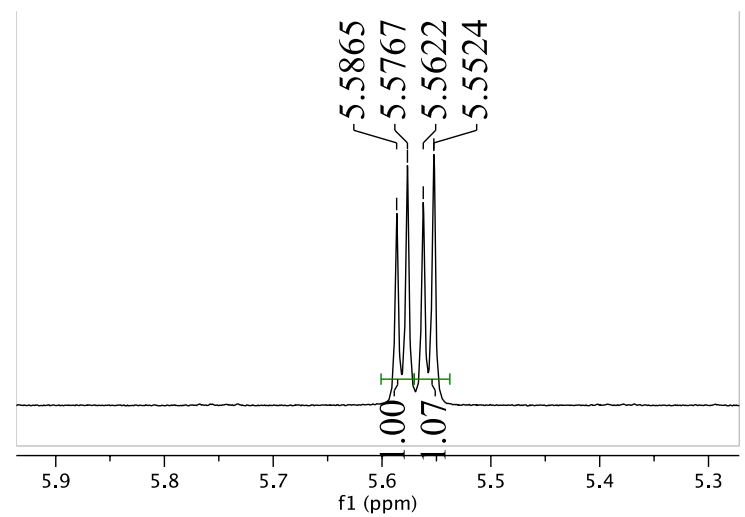
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
332.1399	1	C <sub>20</sub> H <sub>18</sub> N <sub>4</sub> O <sub>2</sub>	332.1394	-1.7	11.3	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **3i**.

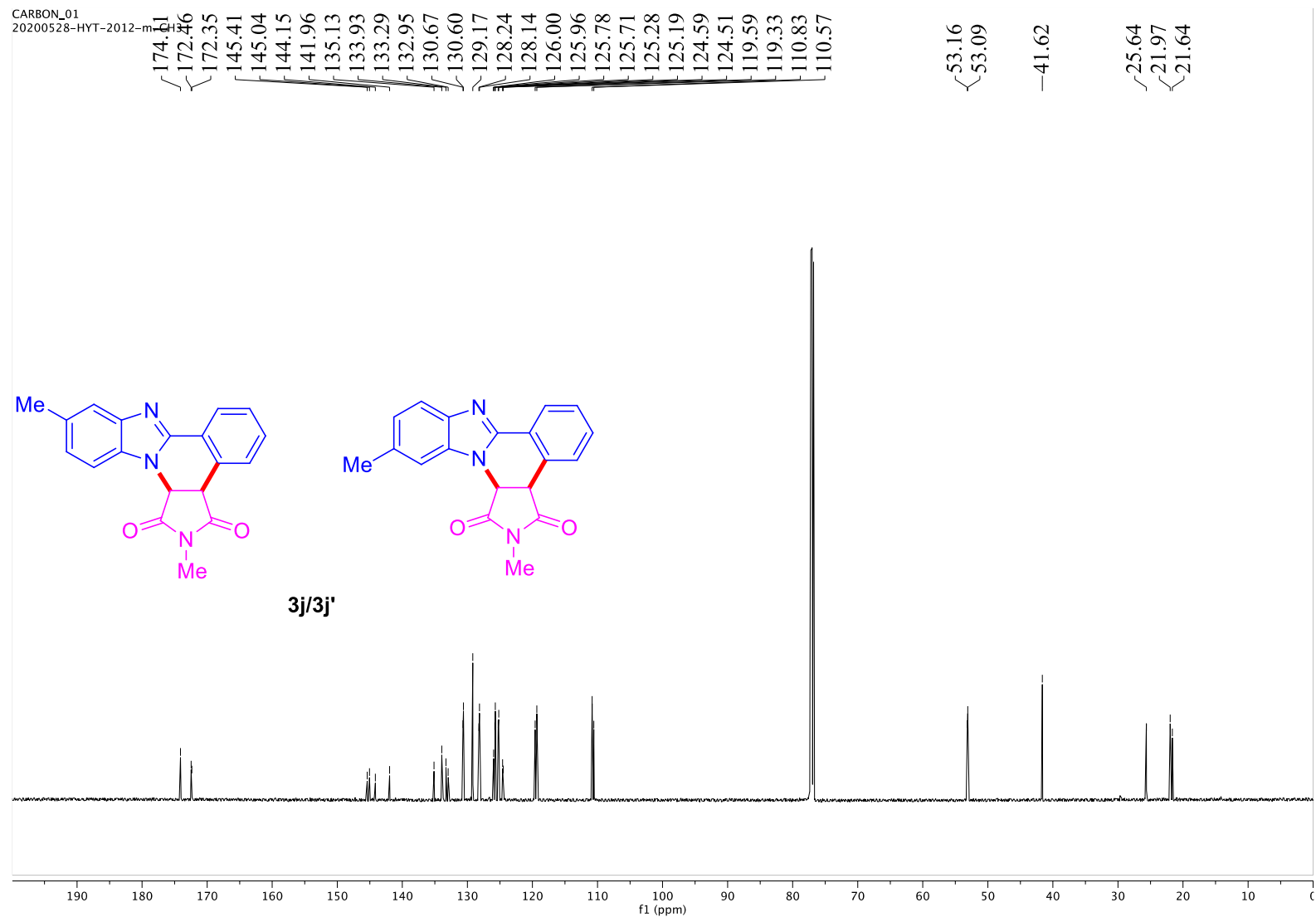


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3j/3j'** in  $\text{CDCl}_3$ .

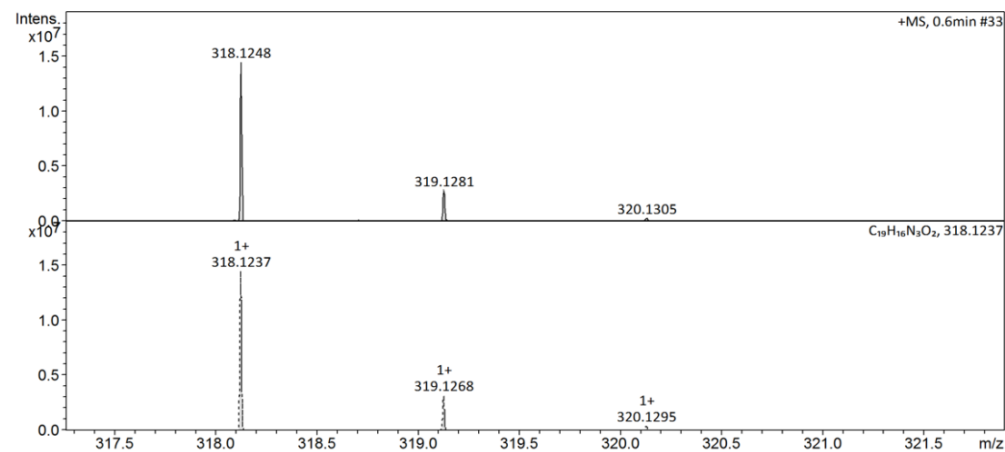
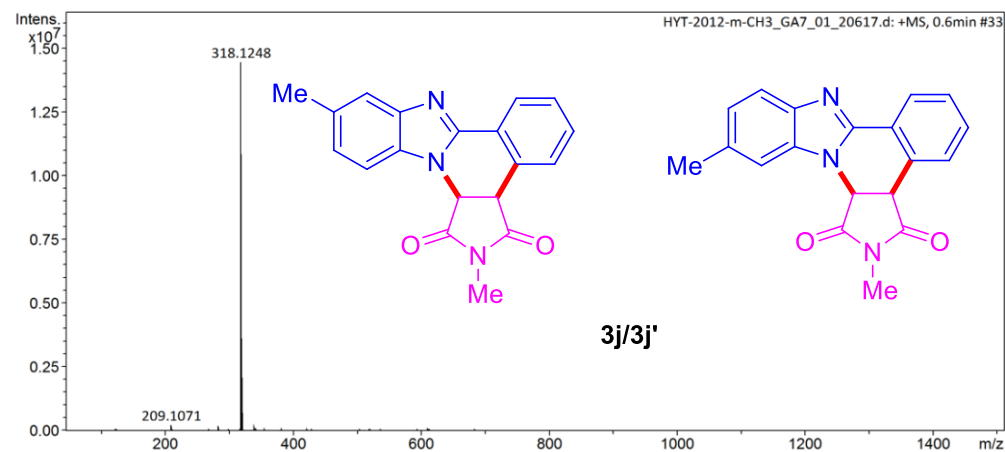




<sup>1</sup>H NMR spectrum (400 MHz) of compound **3j/3j'** in CDCl<sub>3</sub>.



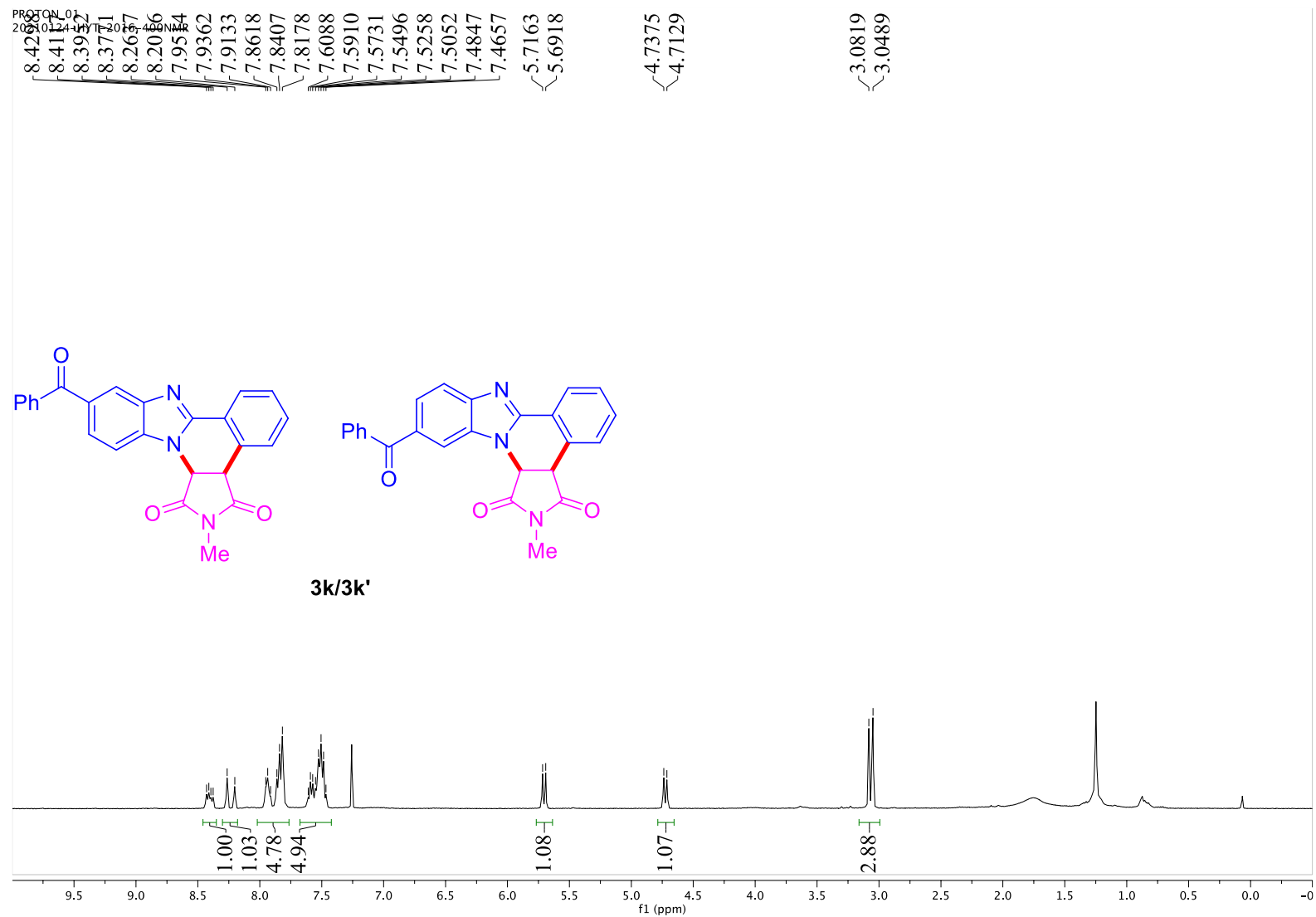
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3j/3j'** in  $\text{CDCl}_3$ .



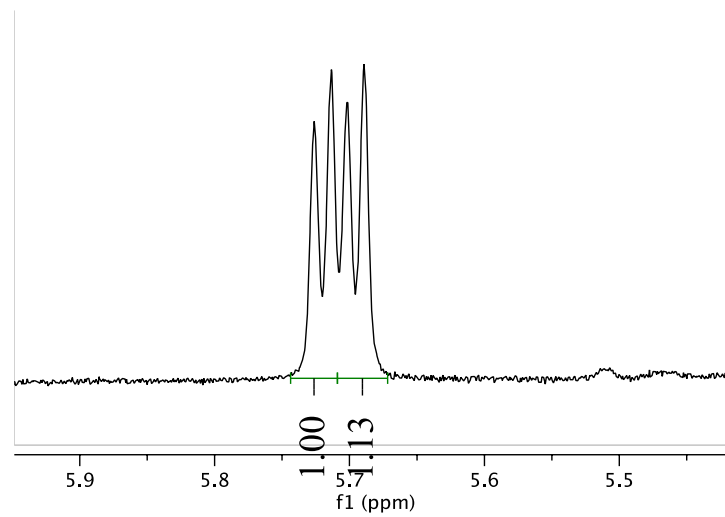
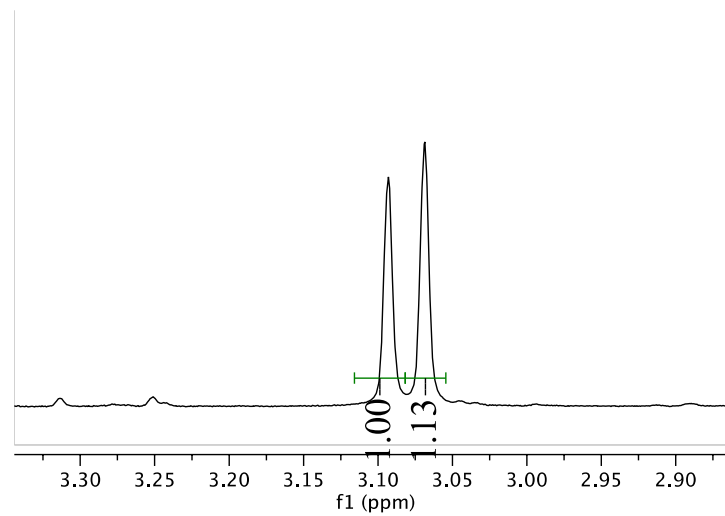
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
318.1248	1	C <sub>19</sub> H <sub>16</sub> N <sub>3</sub> O <sub>2</sub>	318.1237	3.3	17.2	1	100.00	13.5	even	ok	M+H

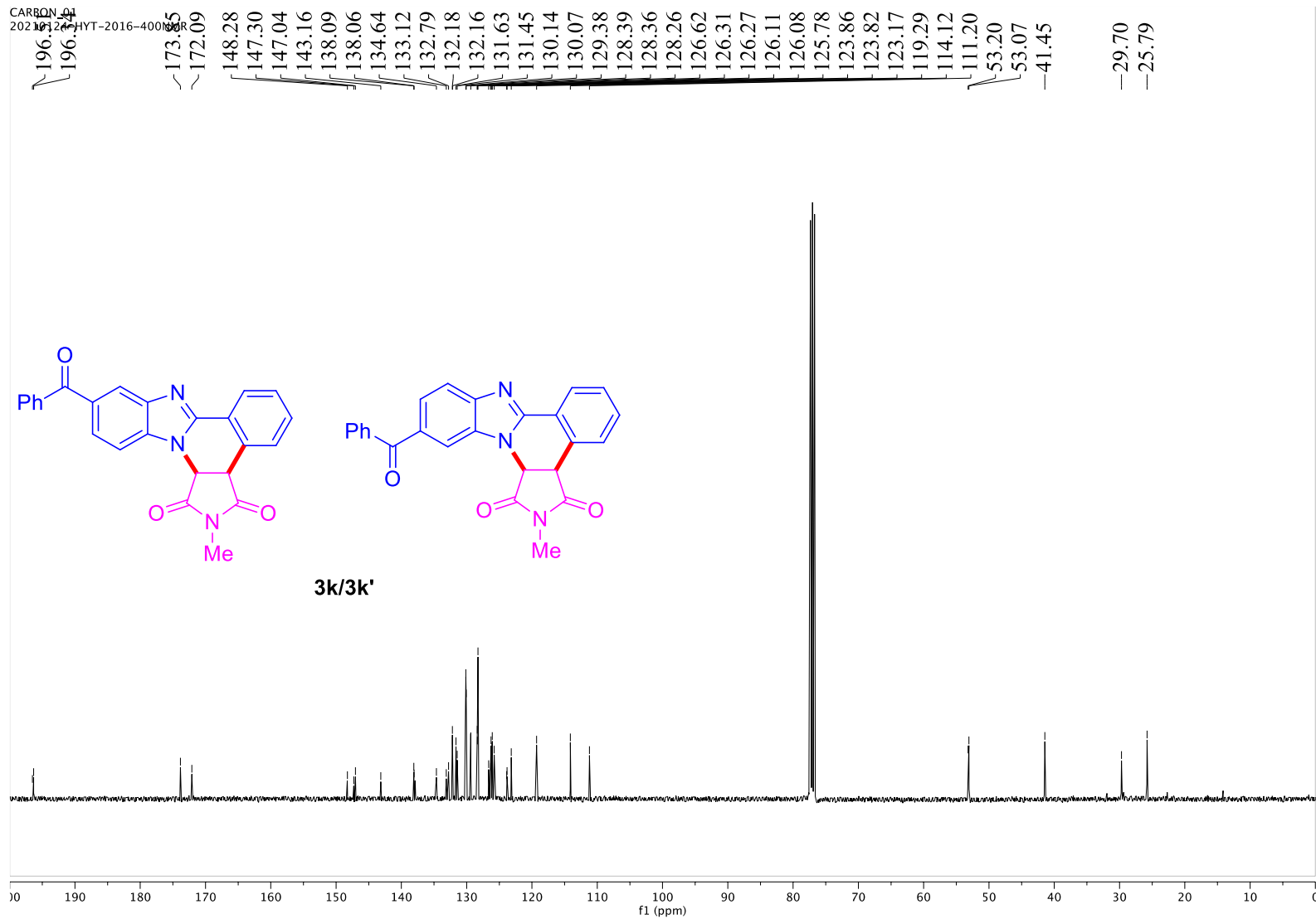
HRMS (ESI) of compound **3j/3j'**.



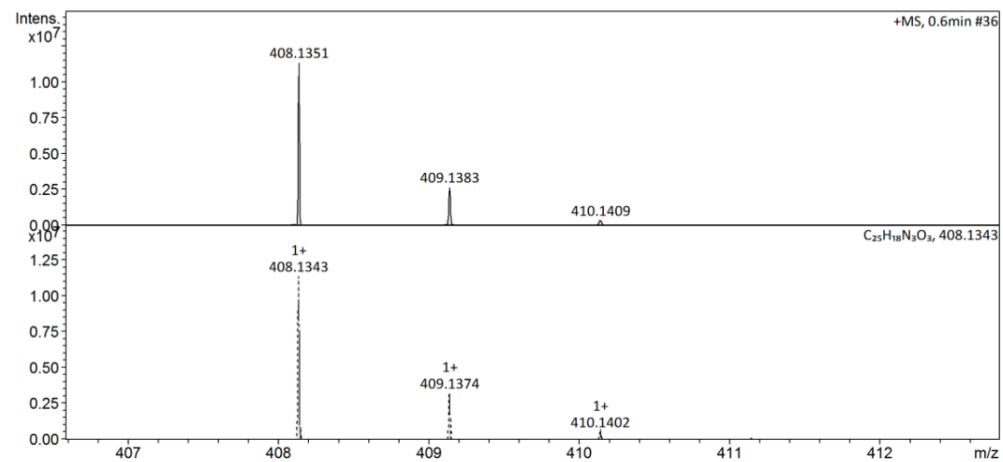
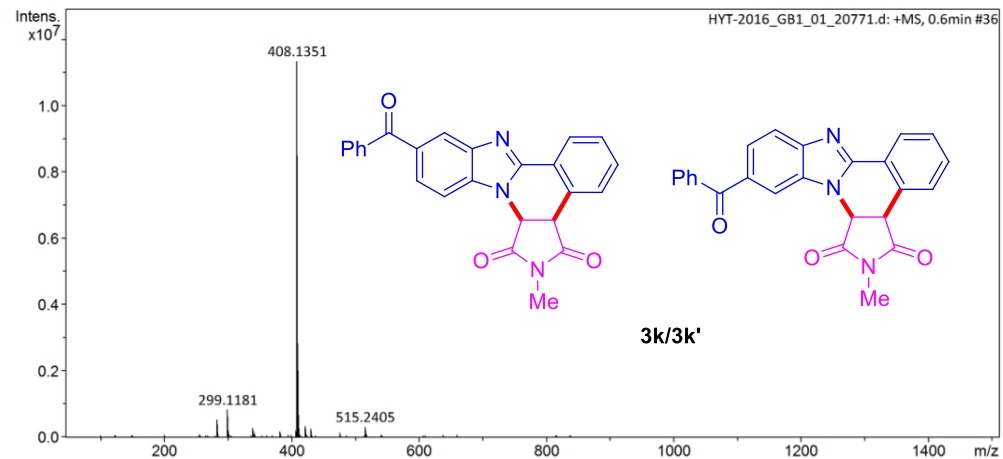
$^1\text{H}$  NMR spectrum (400 MHz) of compound **3k/3k'** in  $\text{CDCl}_3$ .



<sup>1</sup>H NMR spectrum (400 MHz) of compound **3k/3k'** in CDCl<sub>3</sub>.



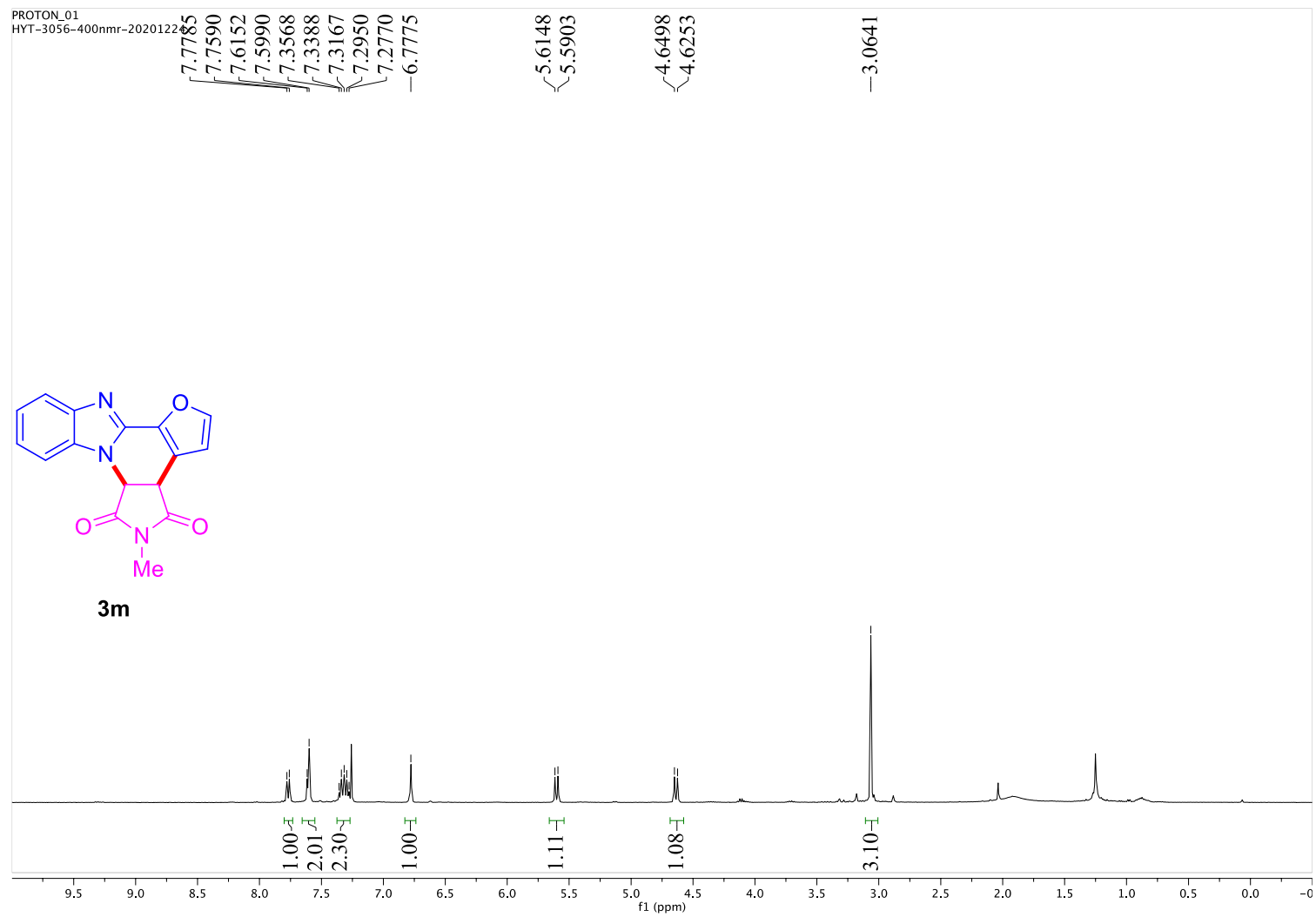
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3k/3k'** in  $\text{CDCl}_3$ .



## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
408.1351	1	C <sub>25</sub> H <sub>18</sub> N <sub>3</sub> O <sub>3</sub>	408.1343	-2.1	25.2	1	100.00	18.5	even	ok	M+H

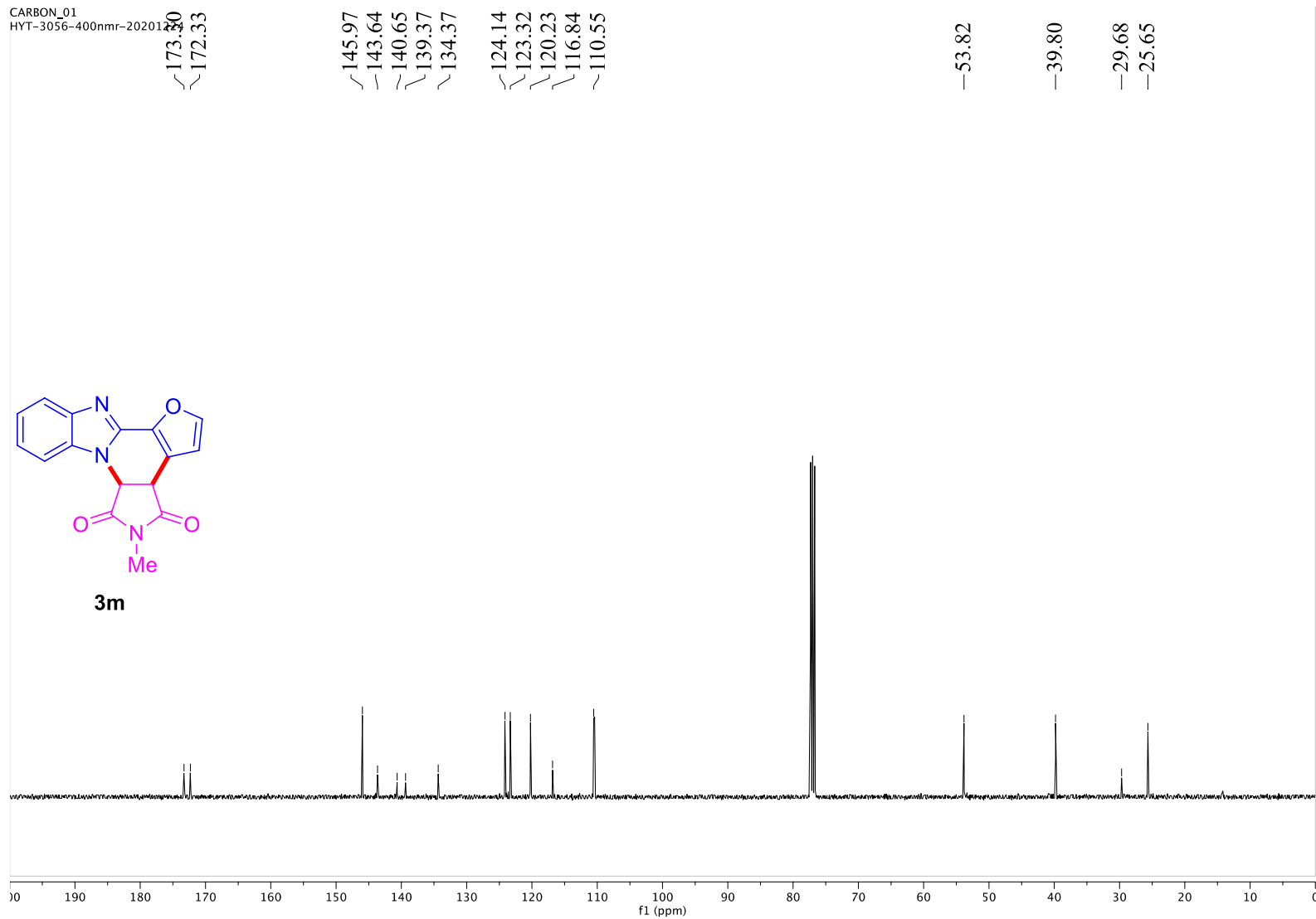
HRMS (ESI) of compound **3k/3k'**.



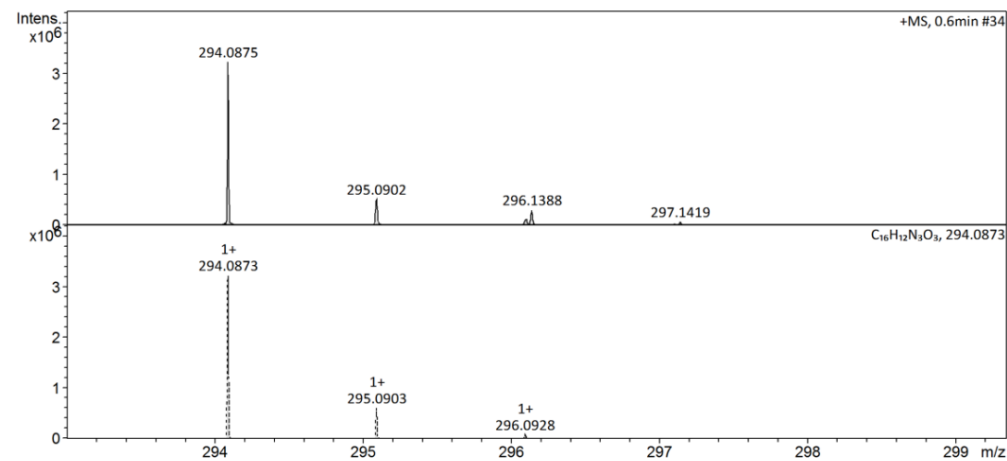
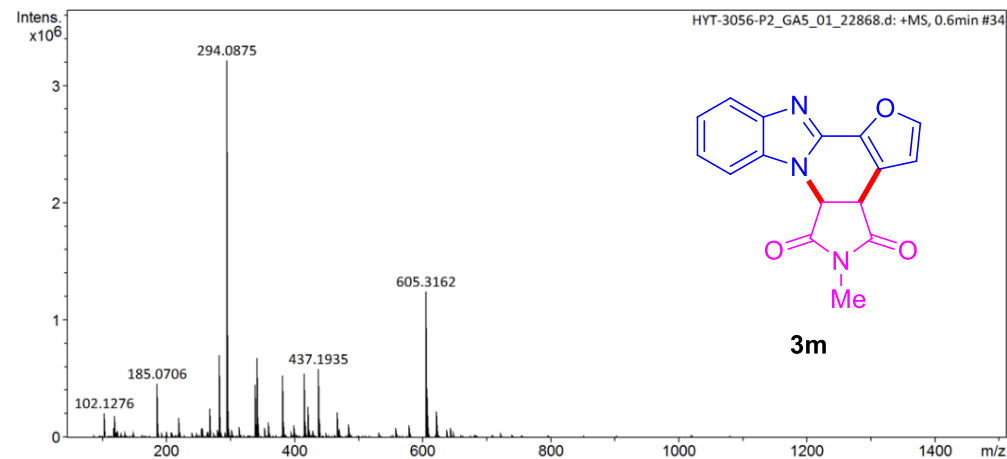
$^1\text{H}$  NMR spectrum (400 MHz) of compound **3m** in  $\text{CDCl}_3$ .



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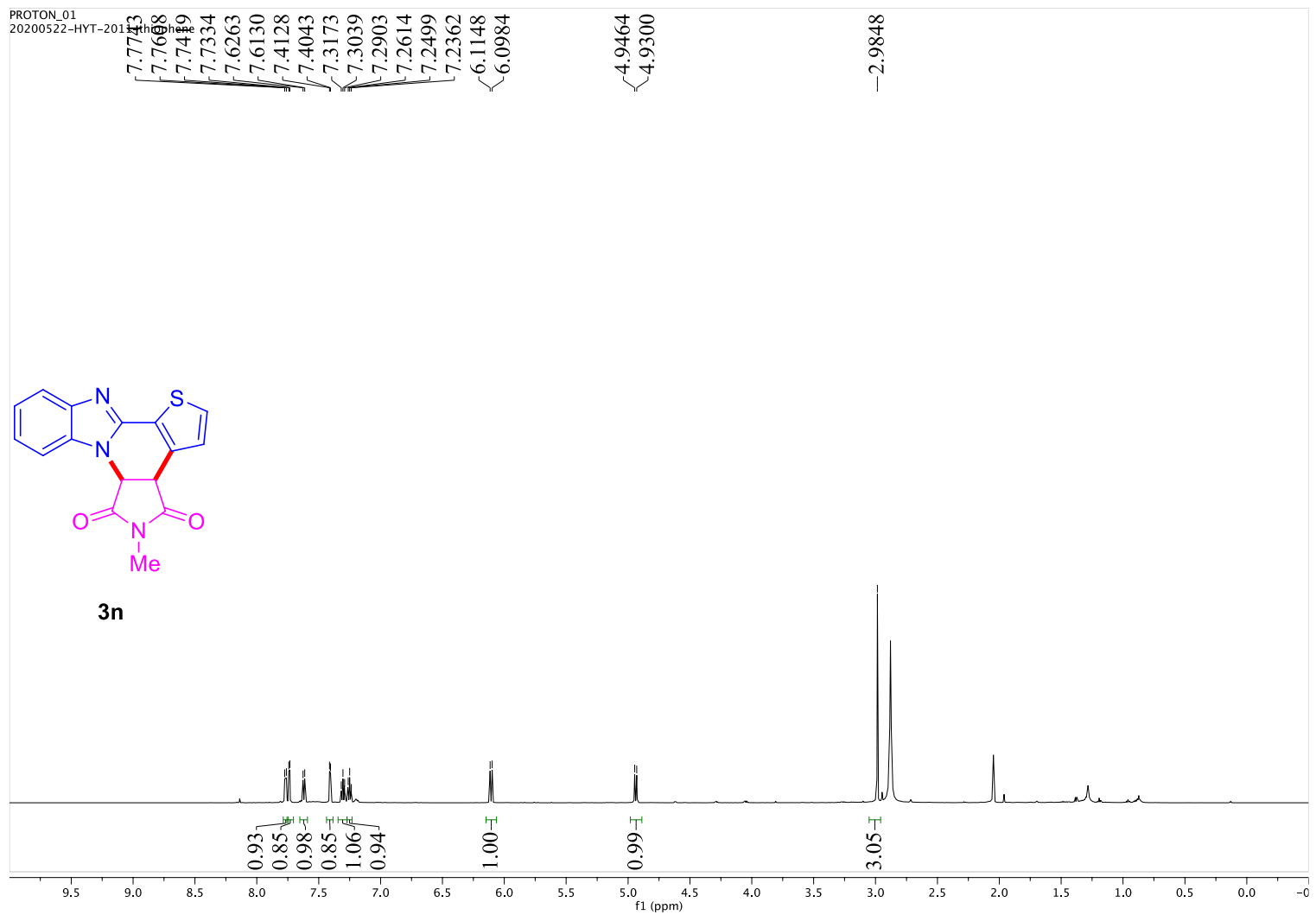
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3m** in  $\text{CDCl}_3$ .



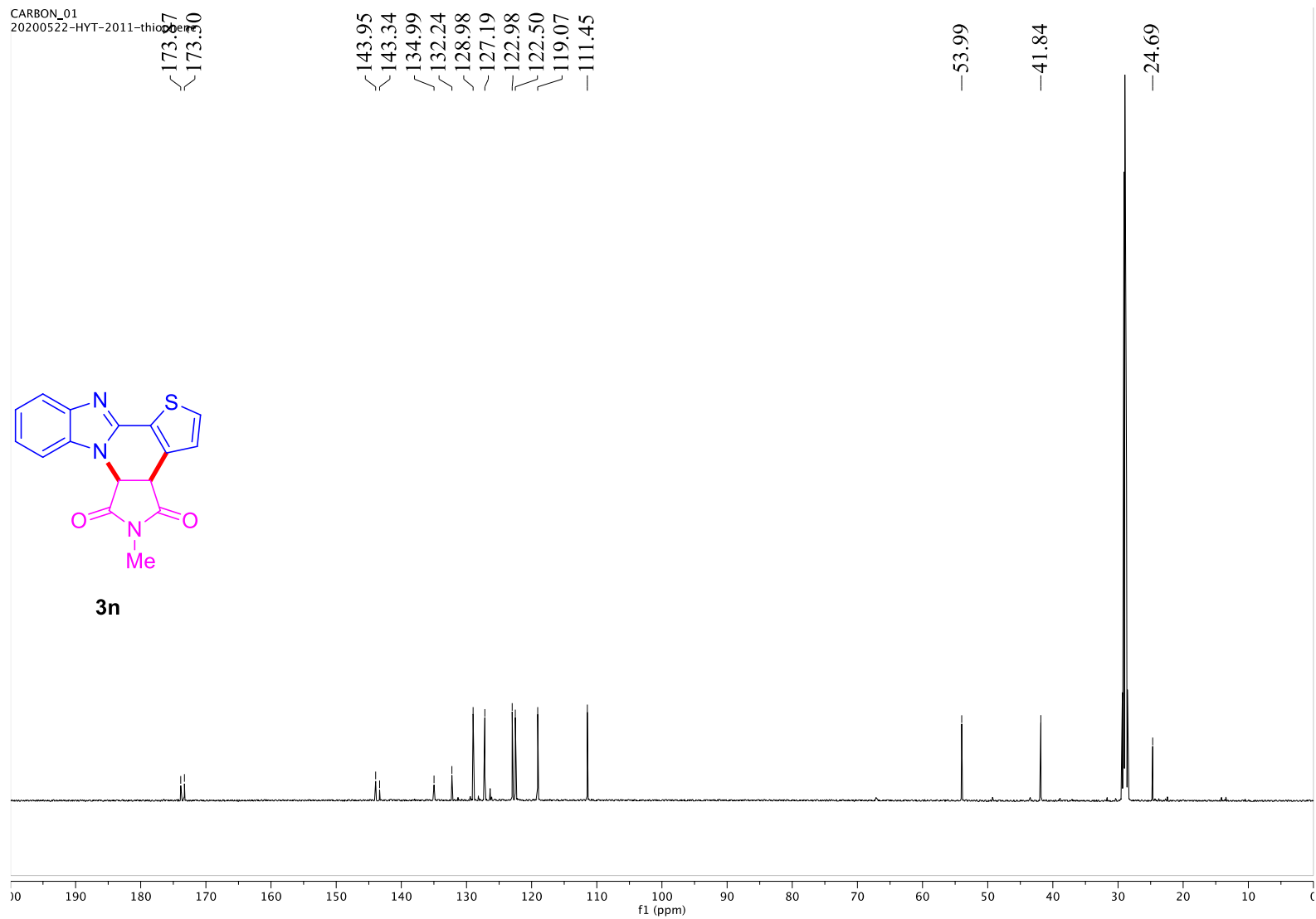
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
294.0875	1	C <sub>16</sub> H <sub>12</sub> N <sub>3</sub> O <sub>3</sub>	294.0873	0.5	18.1	1	100.00	12.5	even	ok	M+H

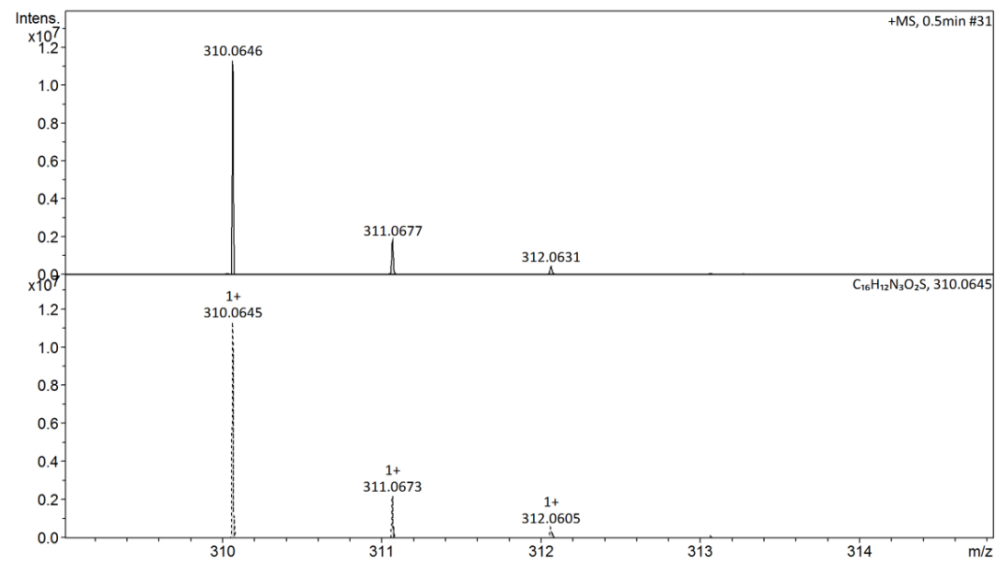
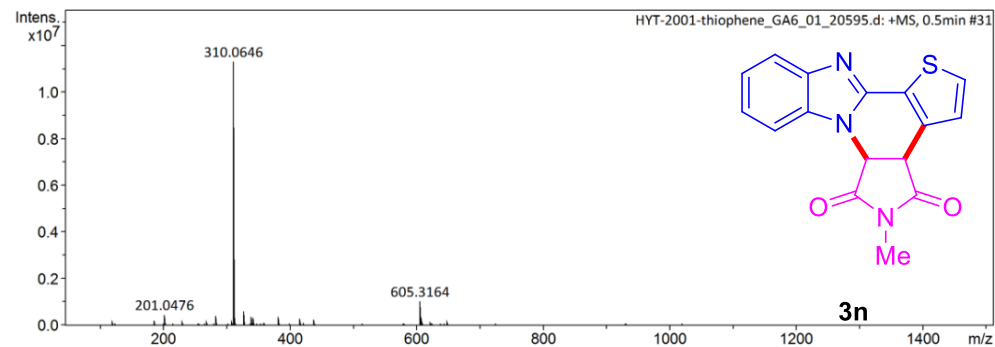
HRMS (ESI) of compound **3m**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3n** in acetone- $d_6$ .



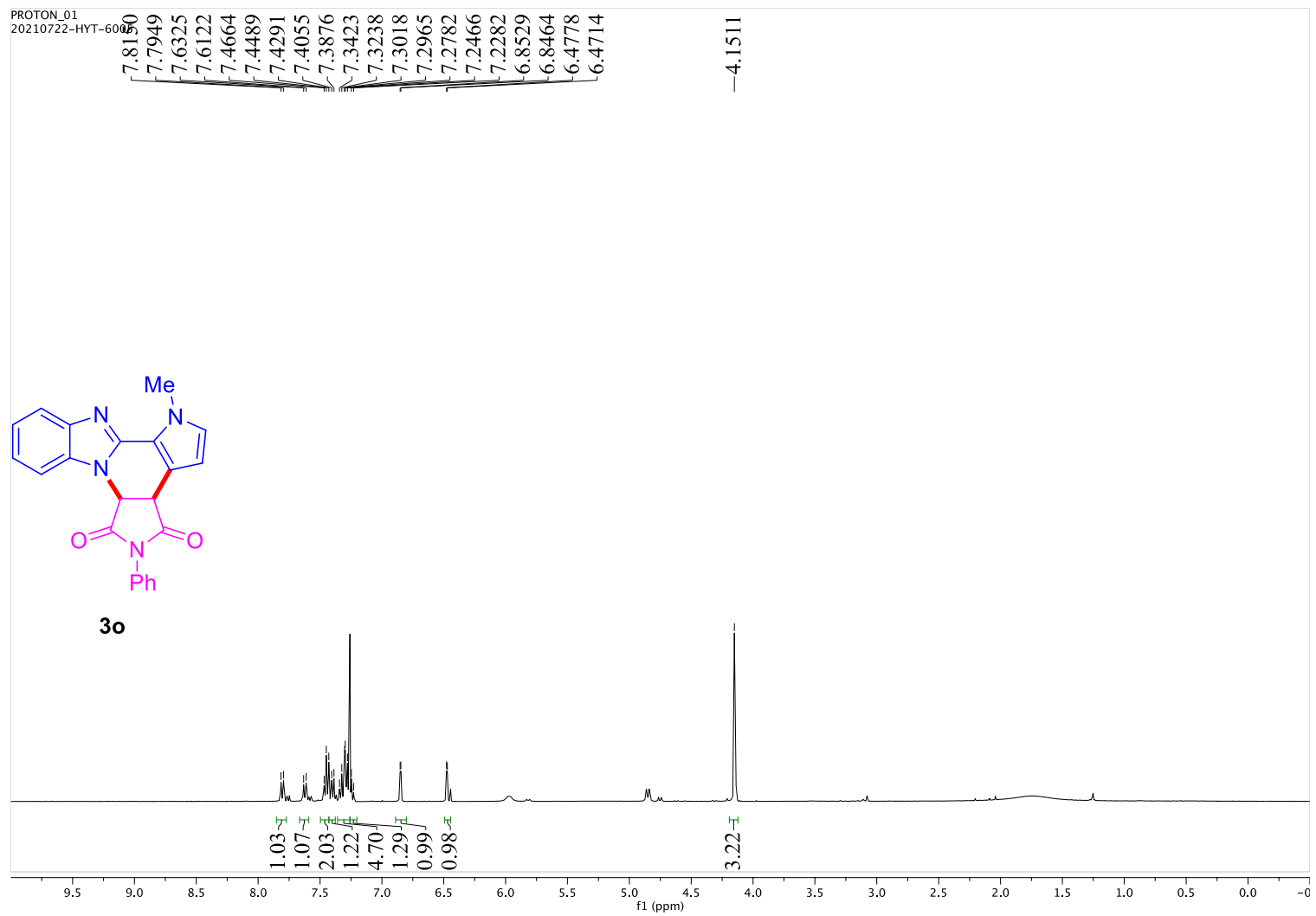
<sup>13</sup>C NMR spectrum (100 MHz) of compound **3n** in acetone-*d*<sub>6</sub>.



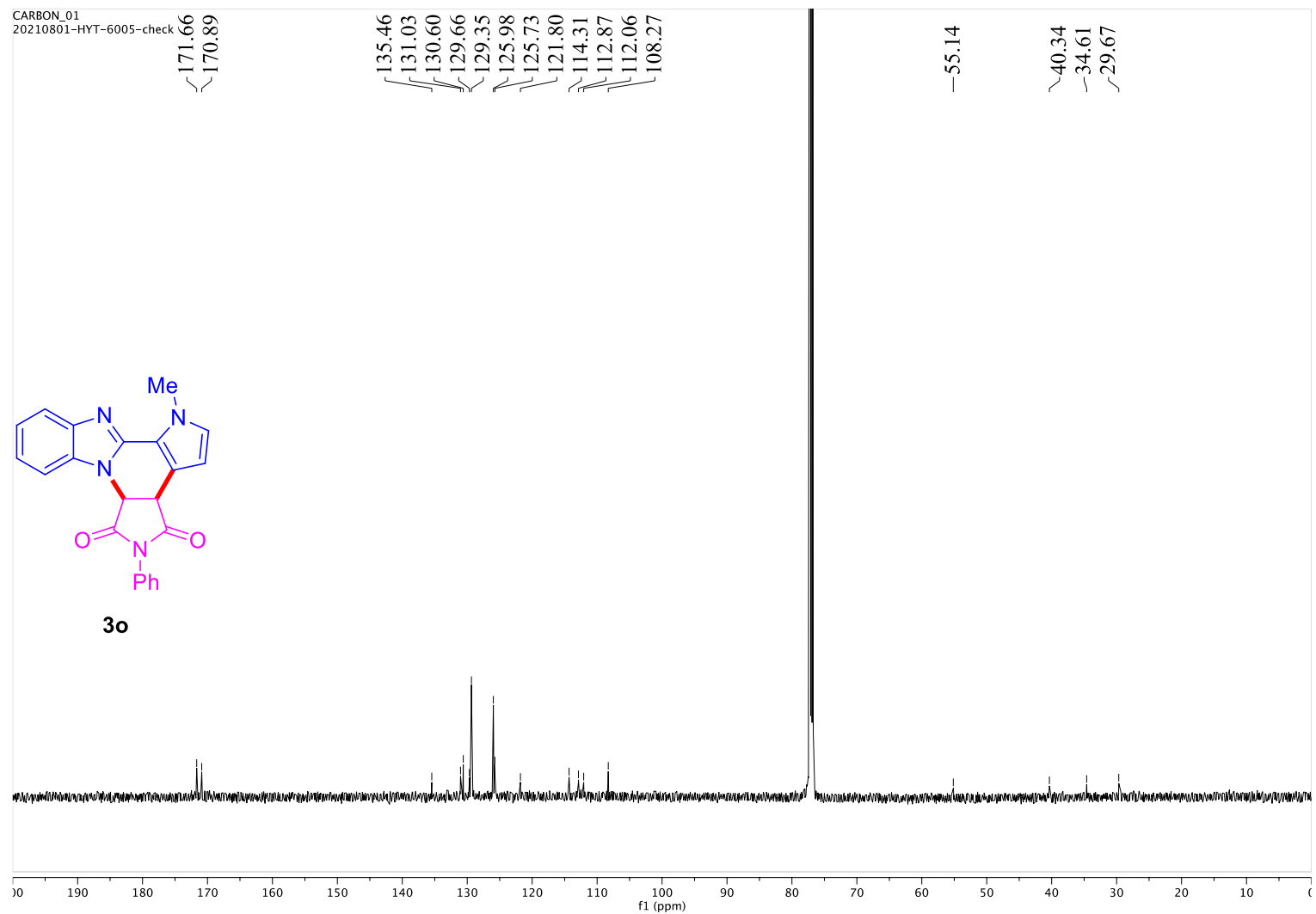
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
310.0646	1	C <sub>16</sub> H <sub>12</sub> N <sub>3</sub> O <sub>2</sub> S	310.0645	0.4	21.6	1	100.00	12.5	even	ok	M+H

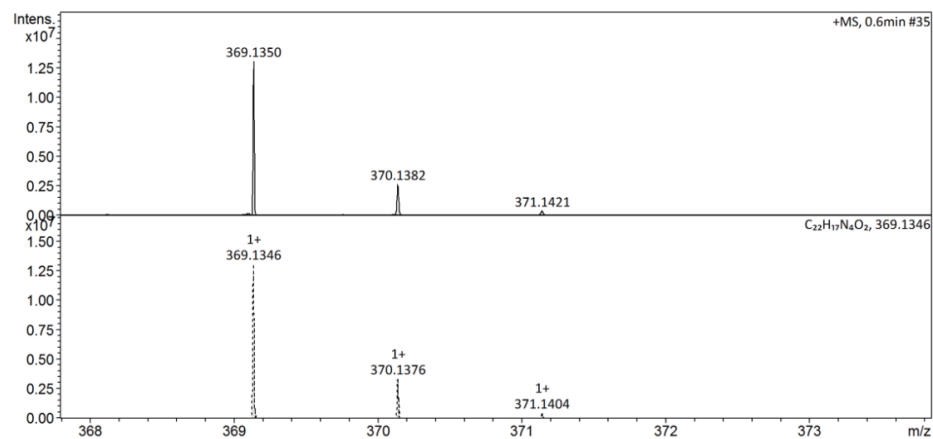
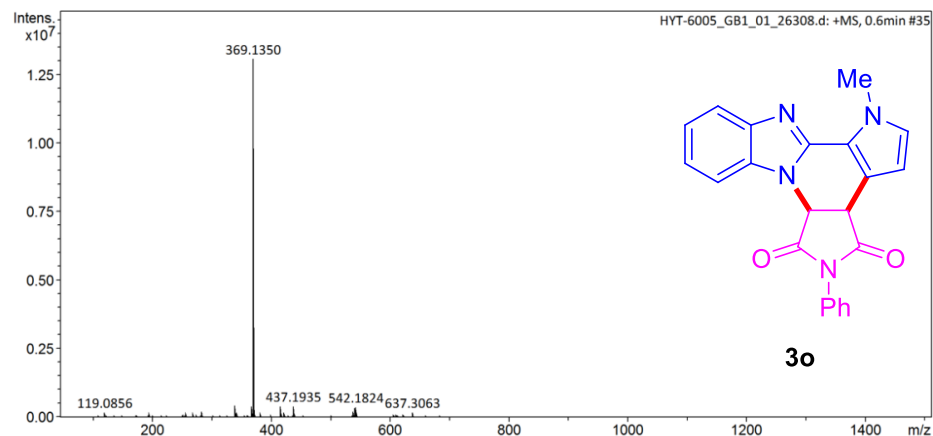
HRMS (ESI) of compound **3n**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3o** in  $\text{CDCl}_3$ .



$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3o** in  $\text{CDCl}_3$ .

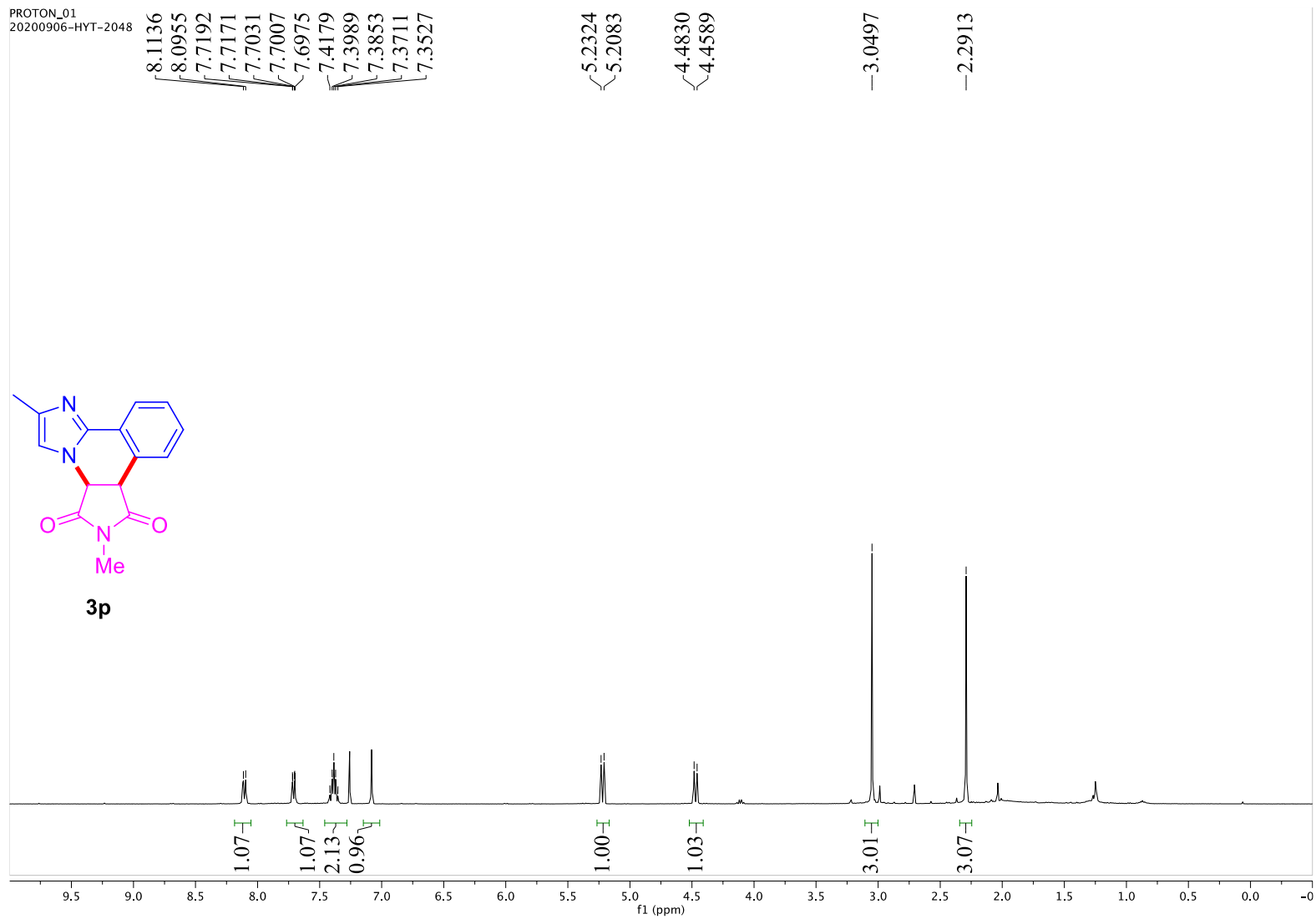


## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
369.1350	1	C <sub>22</sub> H <sub>17</sub> N <sub>4</sub> O <sub>2</sub>	369.1346	-1.1	31.5	1	100.00	16.5	even	ok	M+H

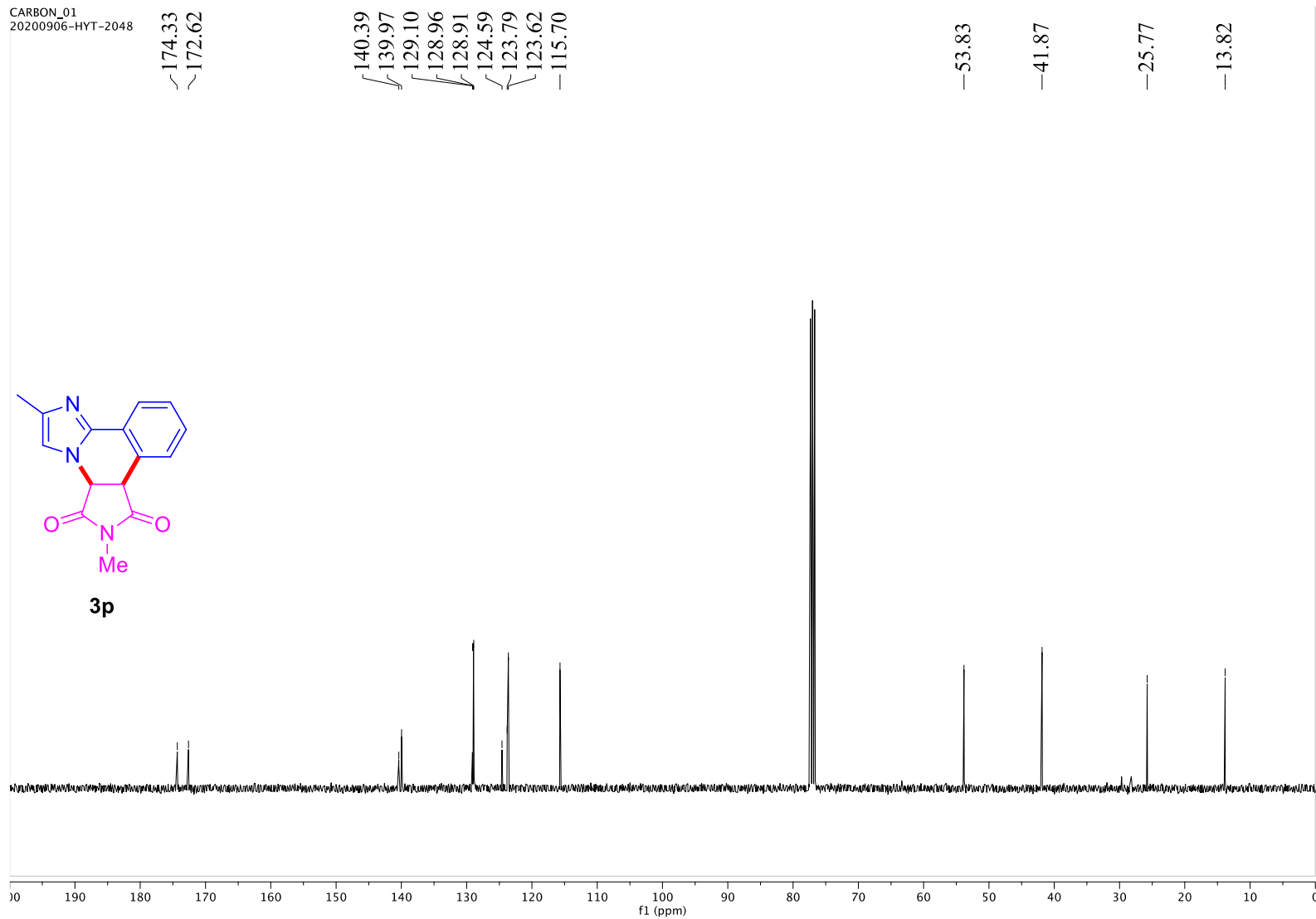
HRMS (ESI) of compound **3o**.



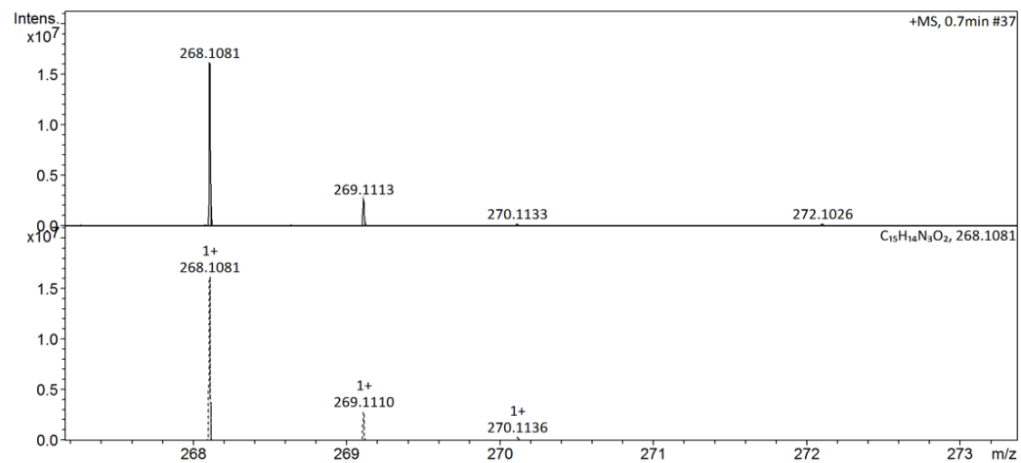
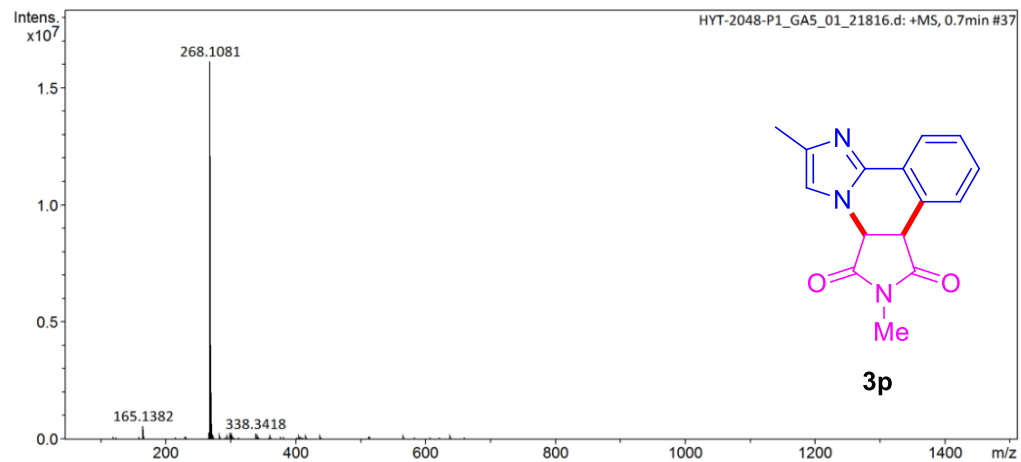


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3p** in  $\text{CDCl}_3$ .

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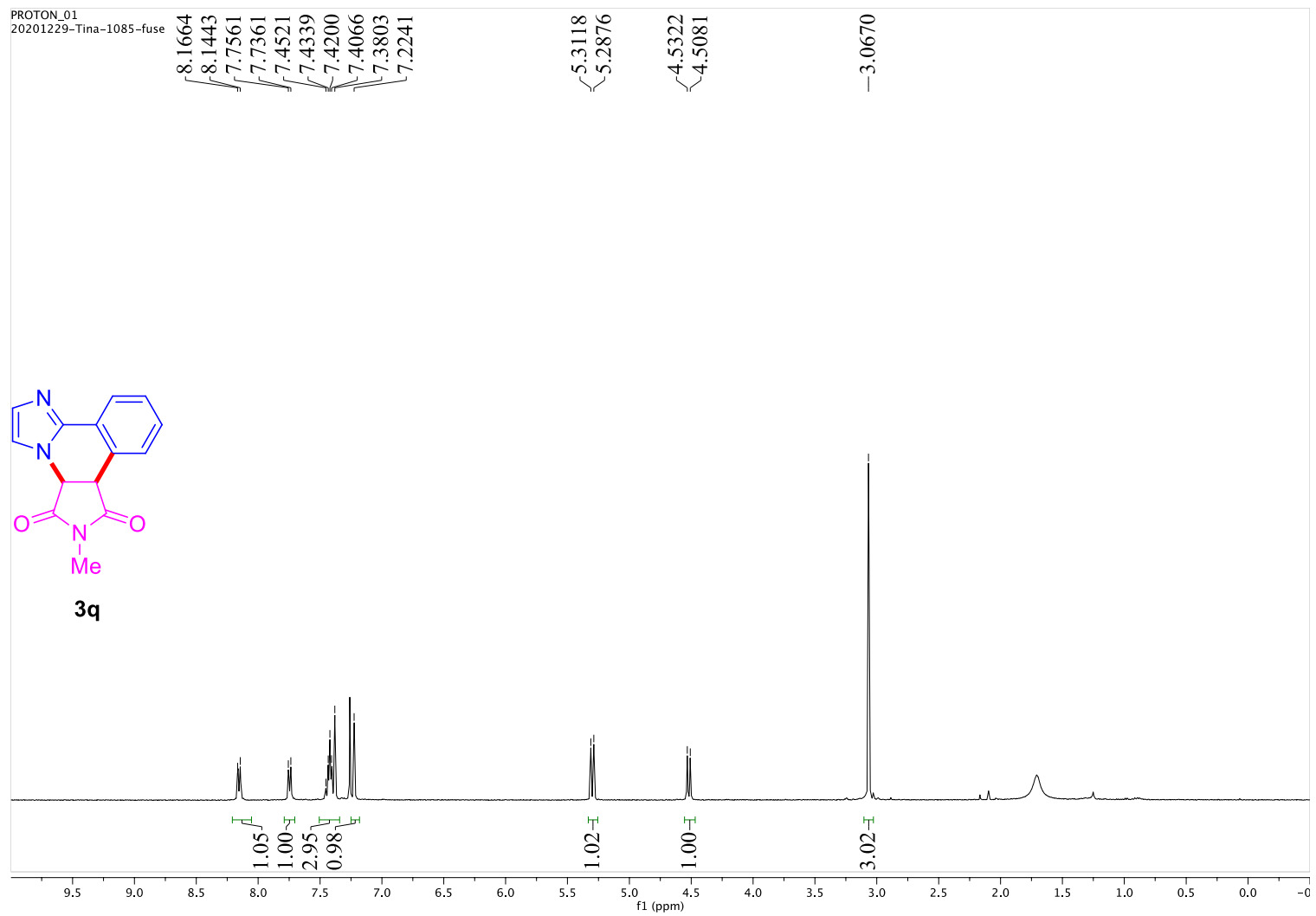
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3p** in  $\text{CDCl}_3$ .



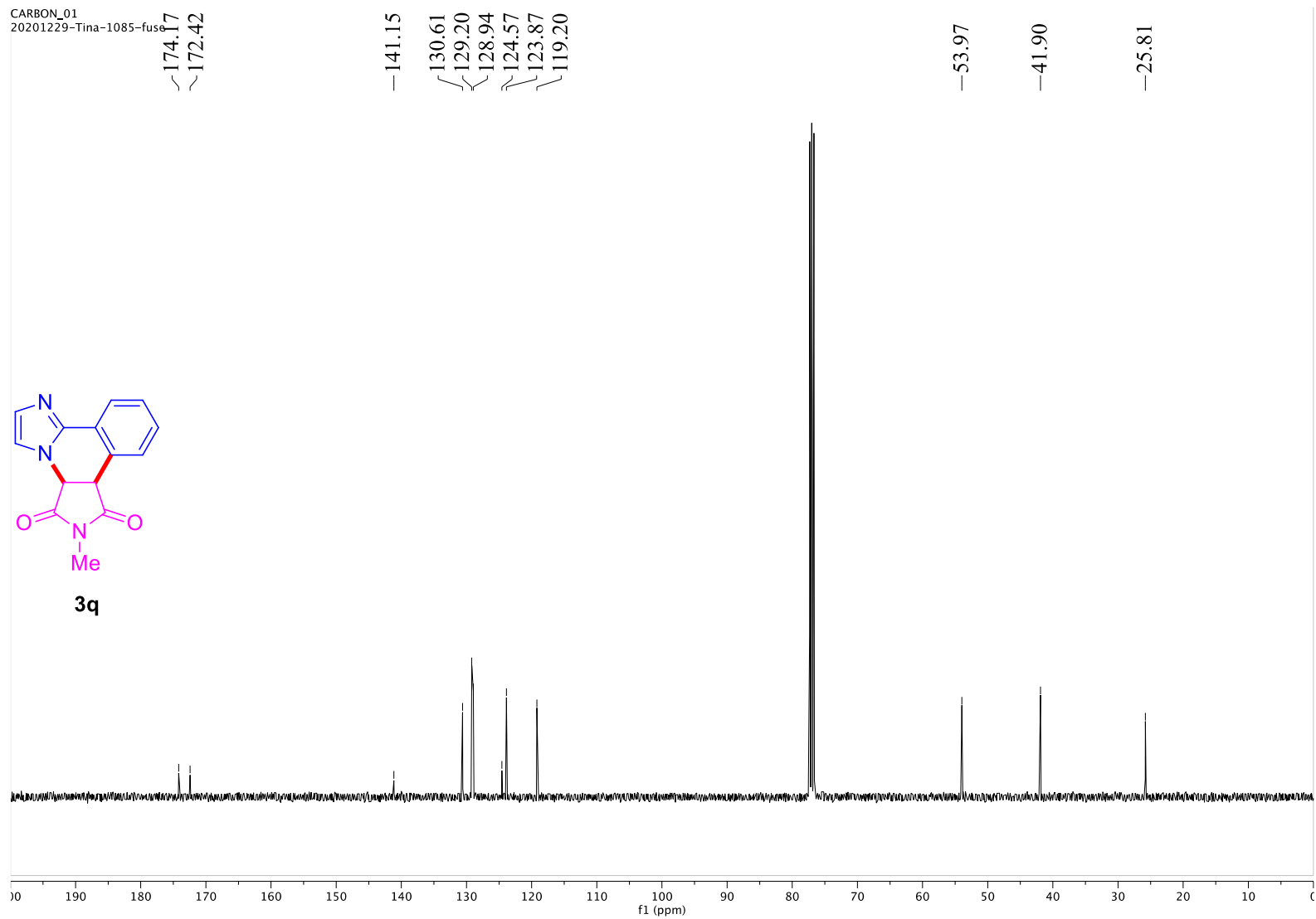
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
268.1081	1	C <sub>15</sub> H <sub>14</sub> N <sub>3</sub> O <sub>2</sub>	268.1081	-0.2	6.4	1	100.00	10.5	even	ok	M+H

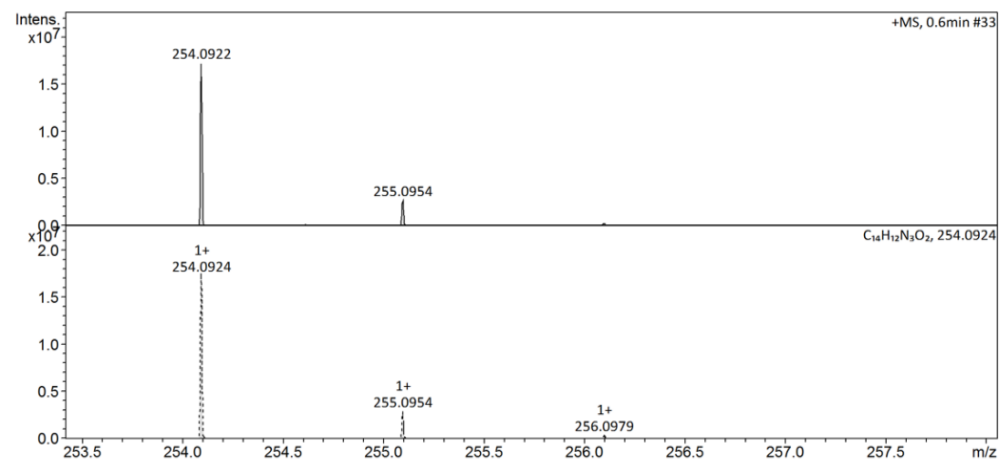
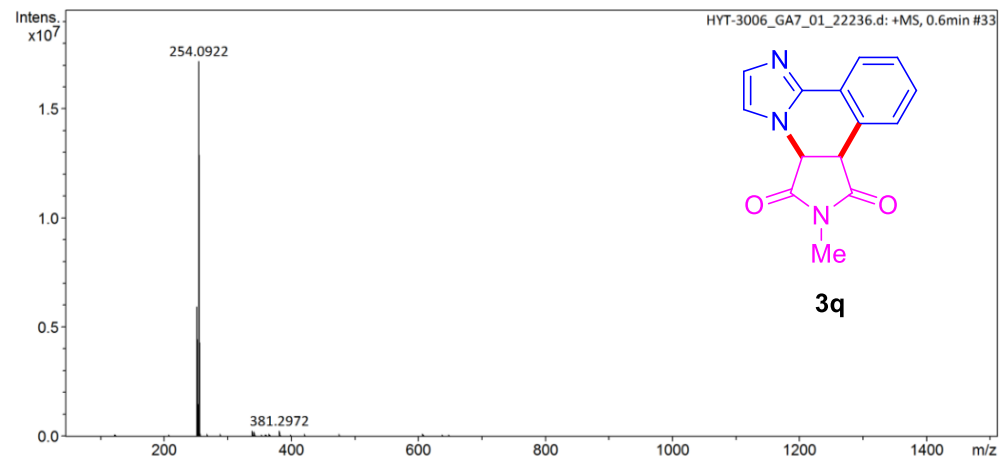
HRMS (ESI) of compound **3p**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3q** in  $\text{CDCl}_3$ .

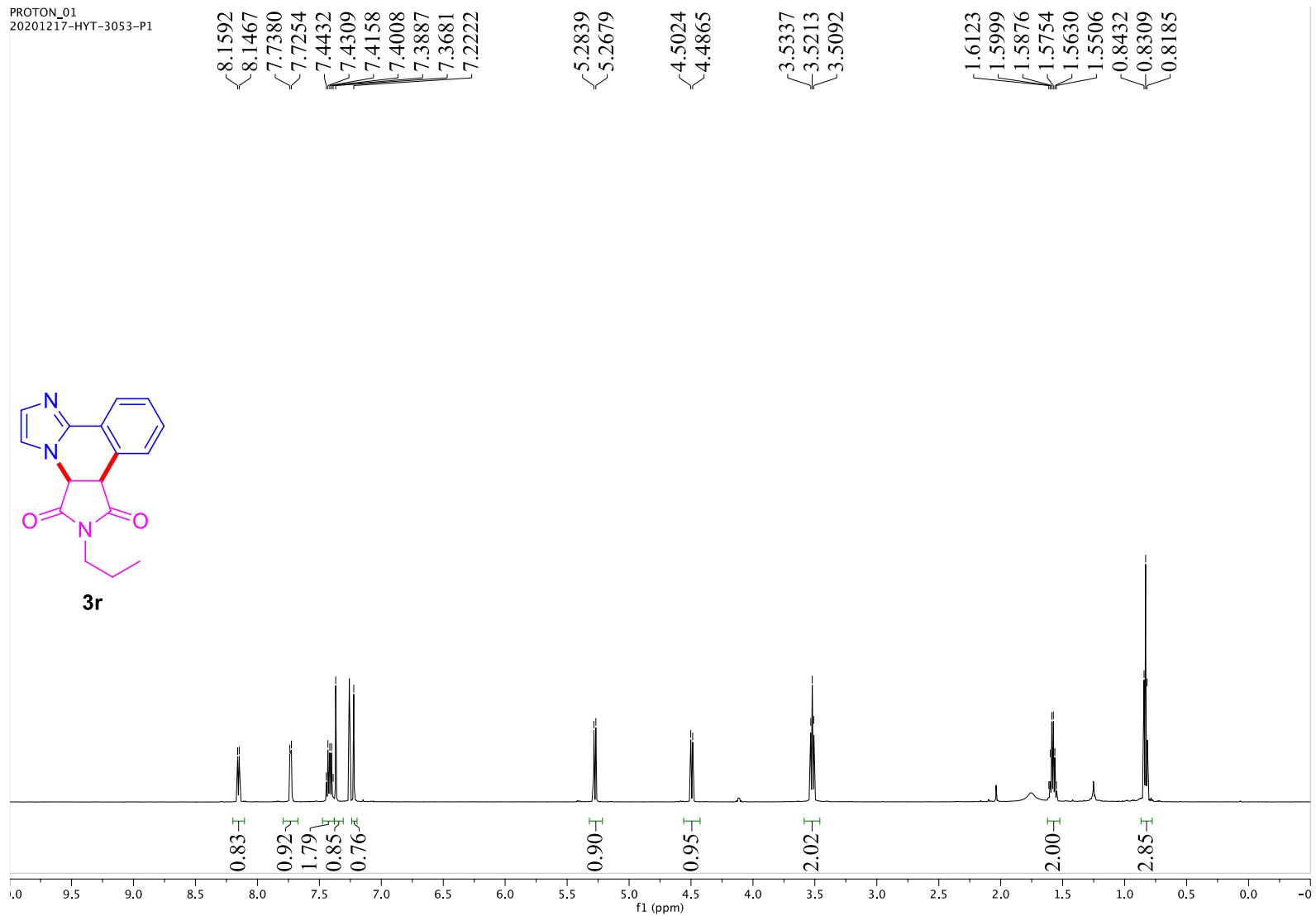


$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3q** in  $\text{CDCl}_3$ .



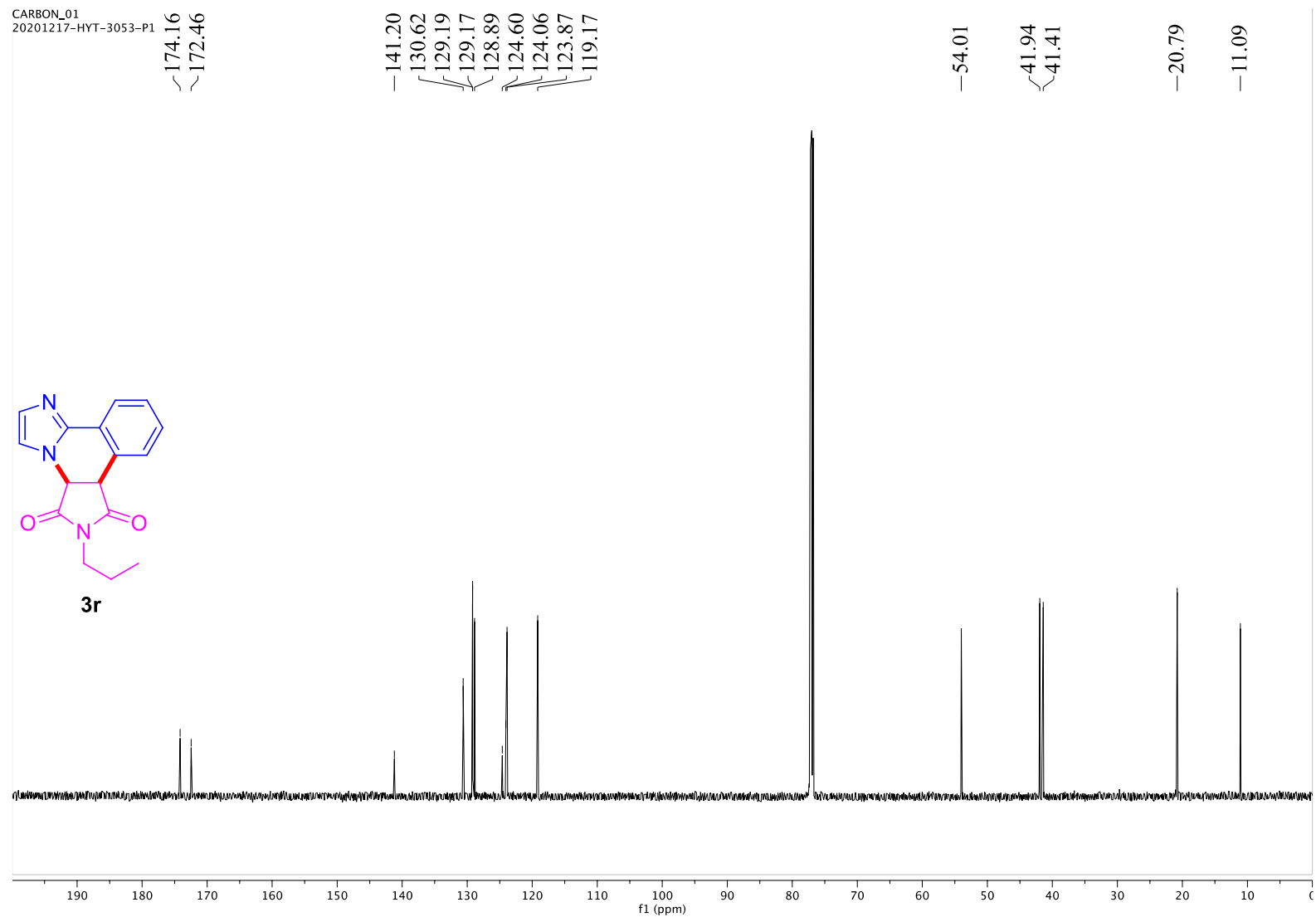
HRMS (ESI) of compound **3q**.

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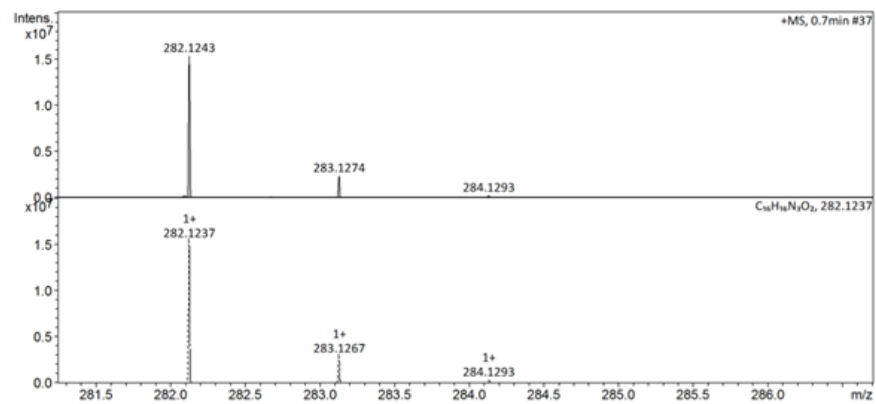
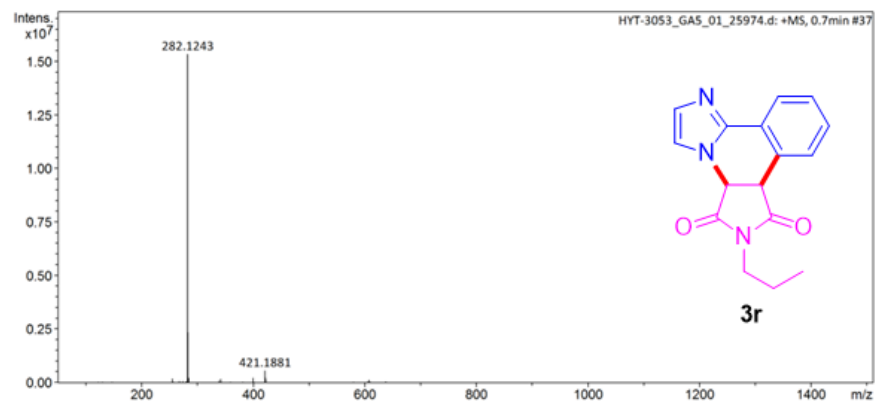
$^1\text{H}$  NMR spectrum (600 MHz) of compound **3r** in  $\text{CDCl}_3$ .

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$^{13}\text{C}$  NMR spectrum (150 MHz) of compound **3r** in  $\text{CDCl}_3$ .



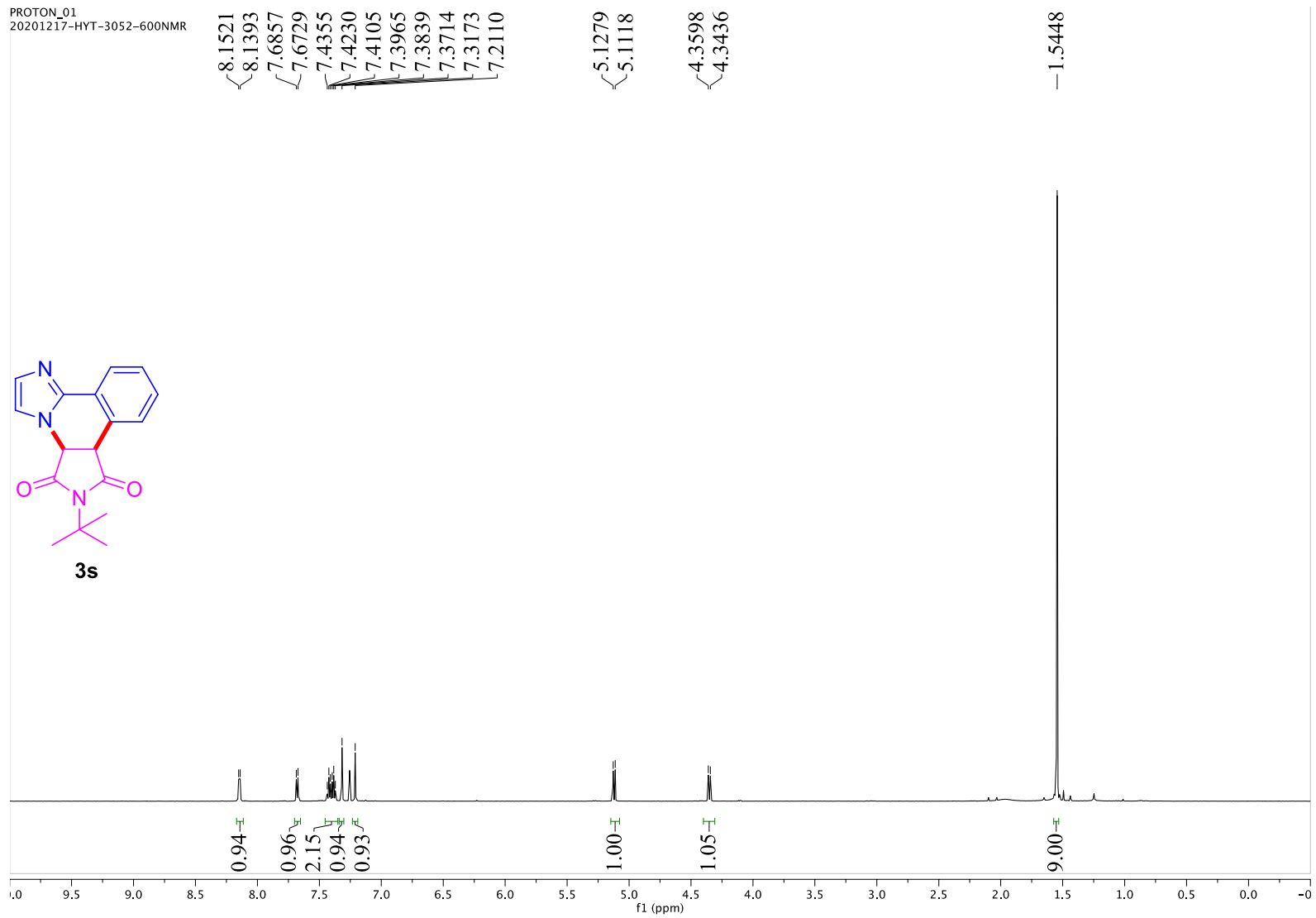


### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
282.1243	1	C <sub>16</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub>	282.1237	-2.2	19.7	1	100.00	10.5	even	ok	M+H

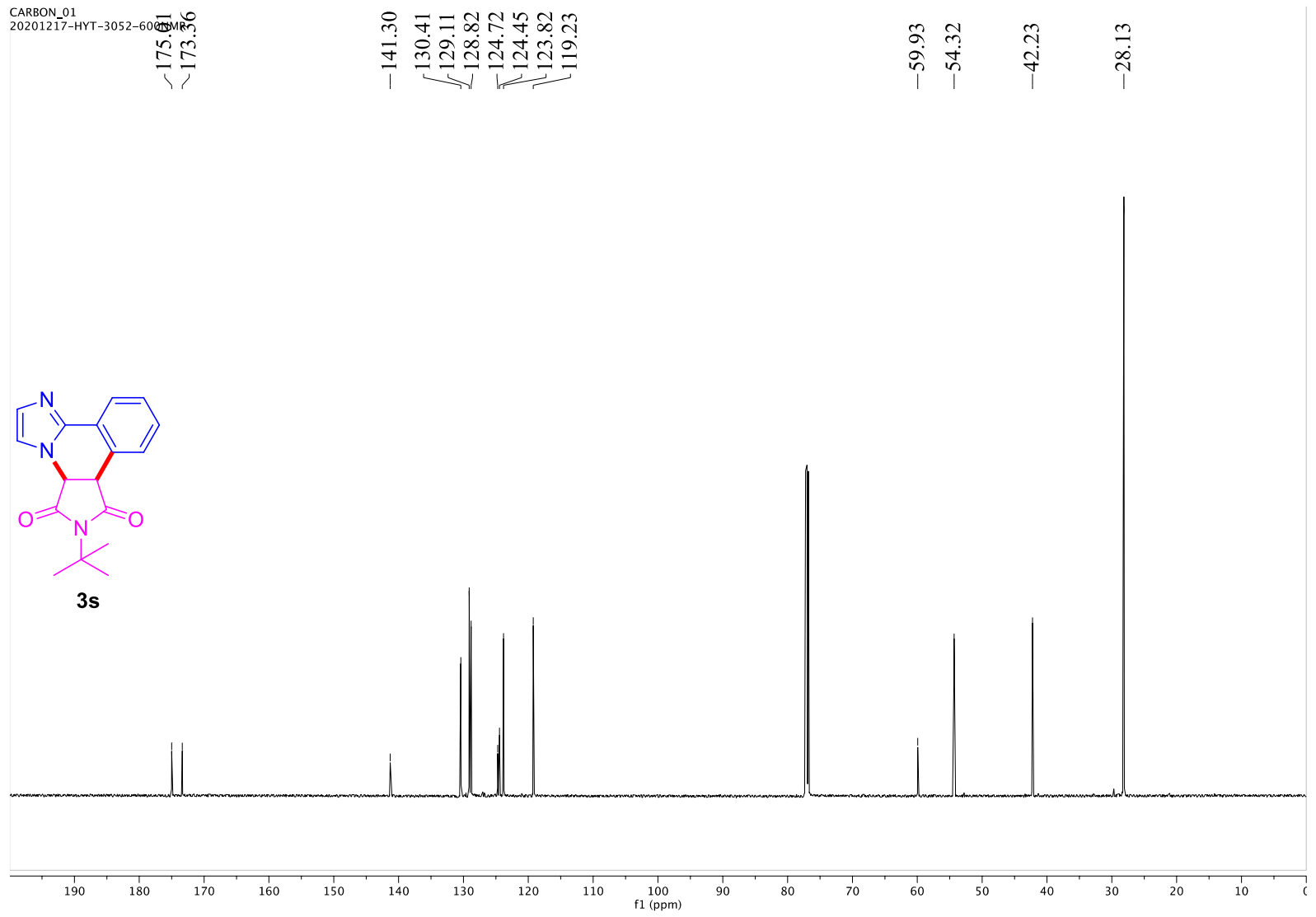
HRMS (ESI) of compound **3r**

PROTON\_01  
20201217-HYT-3052-600NMR

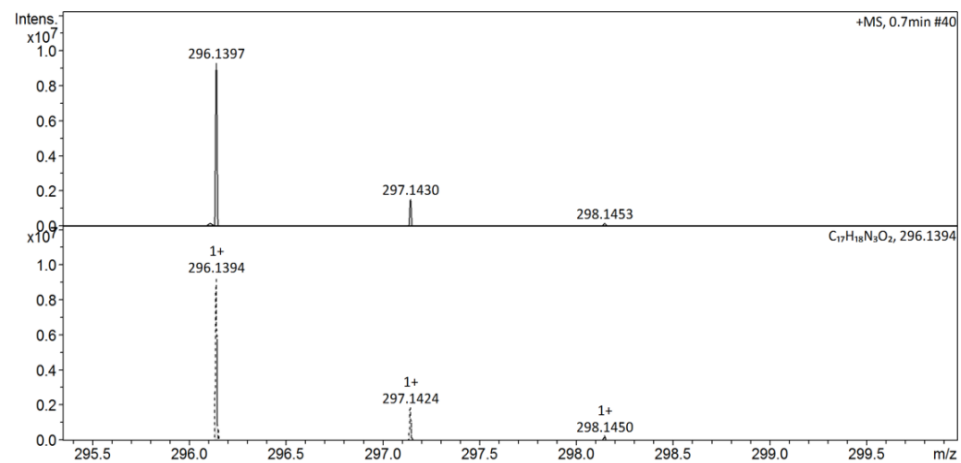
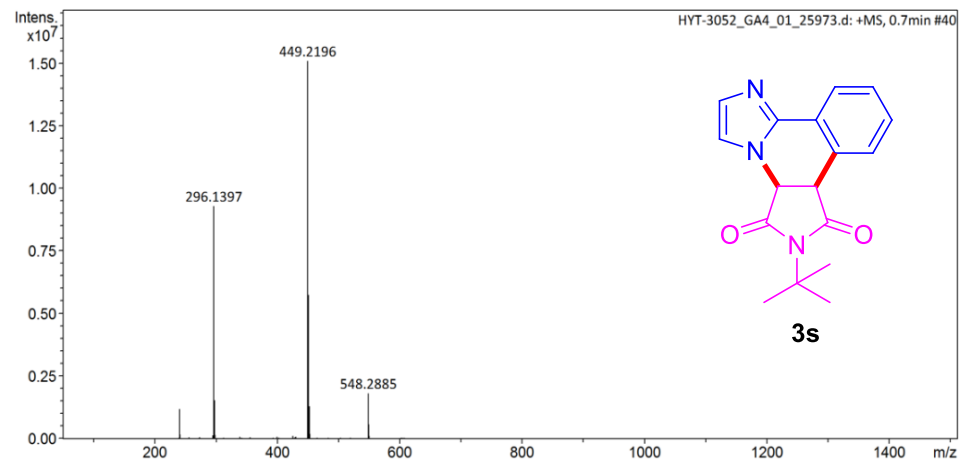


$^1\text{H}$  NMR spectrum (600 MHz) of compound **3s** in  $\text{CDCl}_3$ .

CARBON\_01  
20201217-HYT-3052-6000M



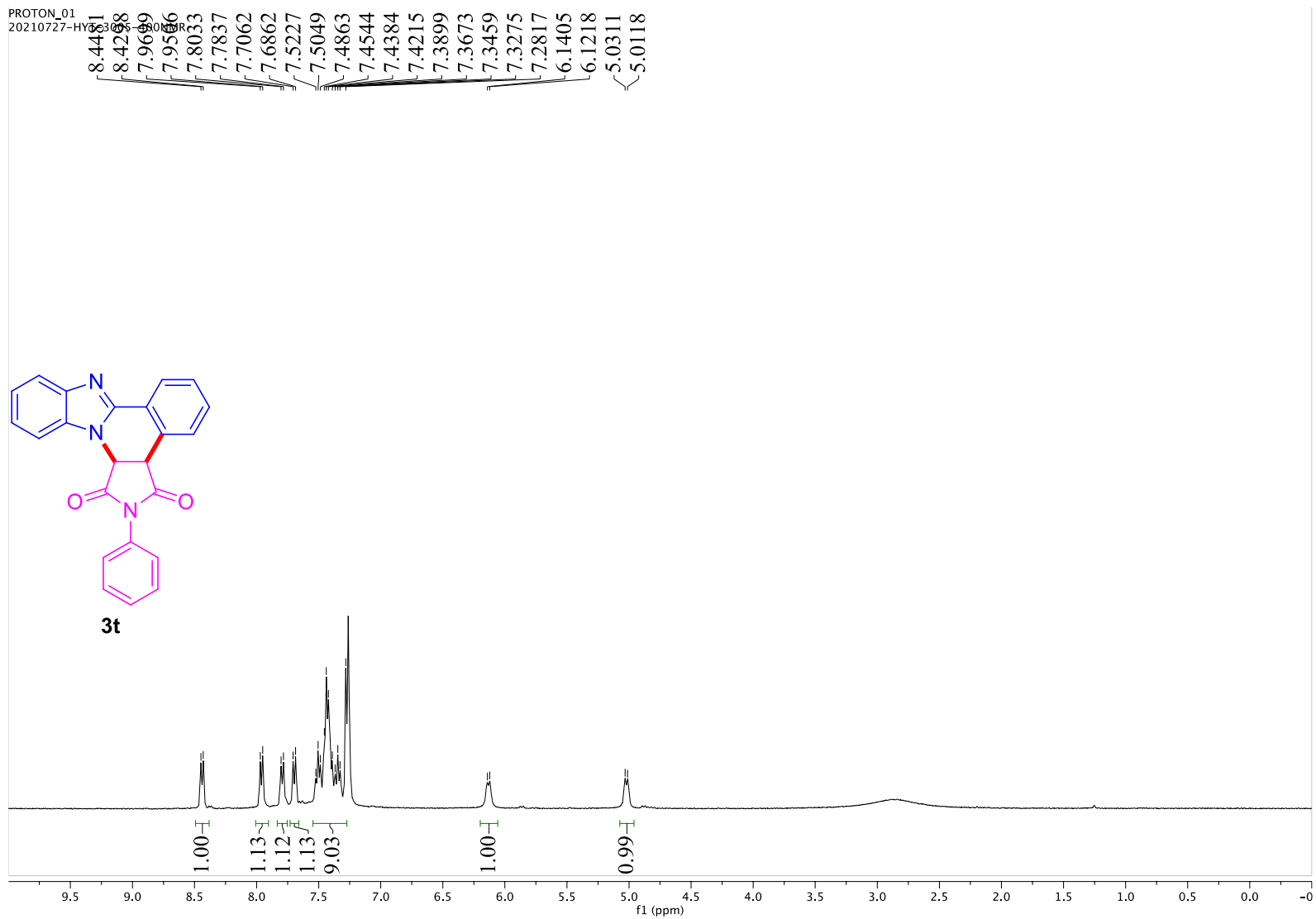
$^{13}\text{C}$  NMR spectrum (150 MHz) of compound **3s** in  $\text{CDCl}_3$ .



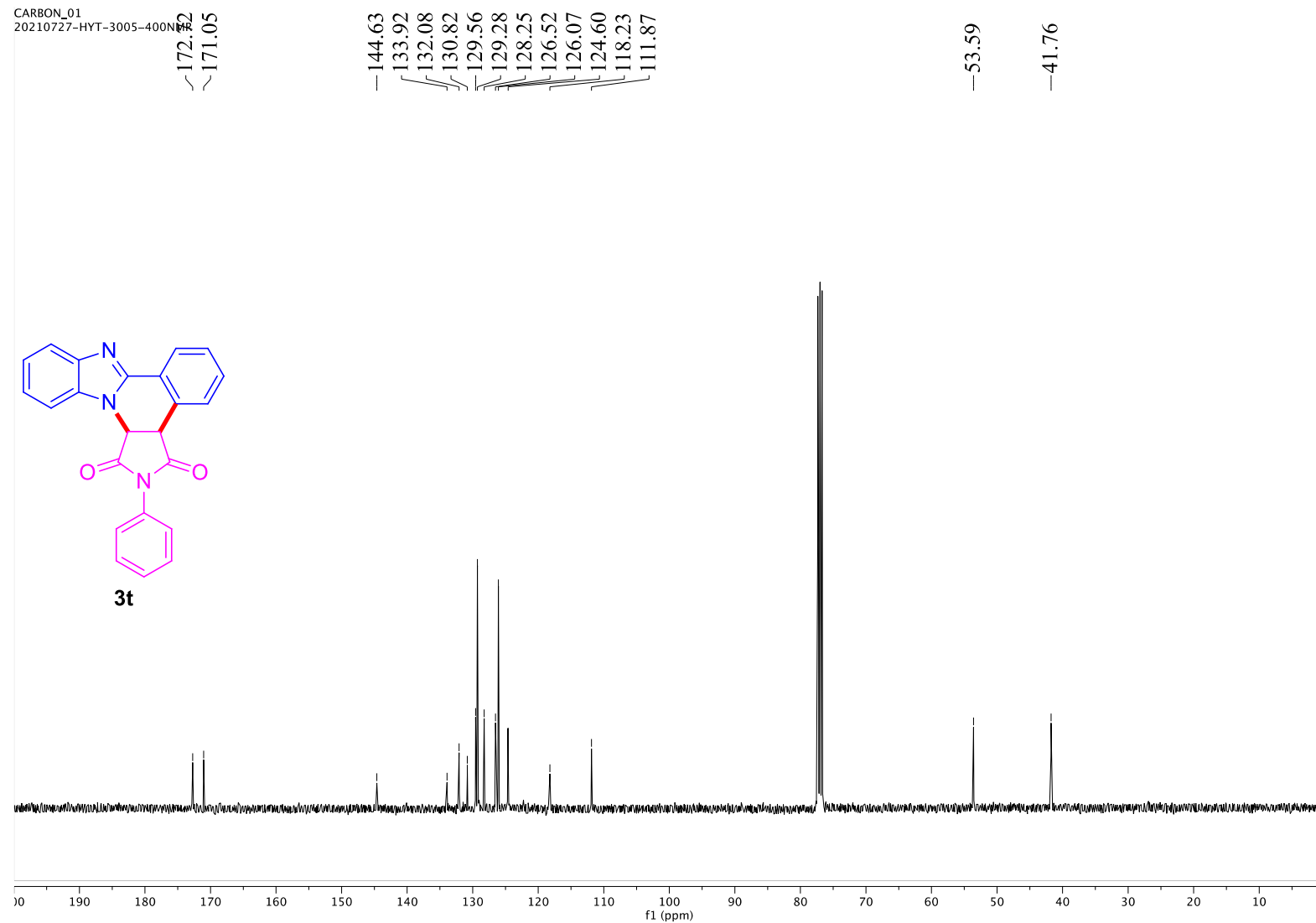
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
296.1397	1	C <sub>17</sub> H <sub>18</sub> N <sub>3</sub> O <sub>2</sub>	296.1394	-1.3	19.2	1	100.00	10.5	even	ok	M+H

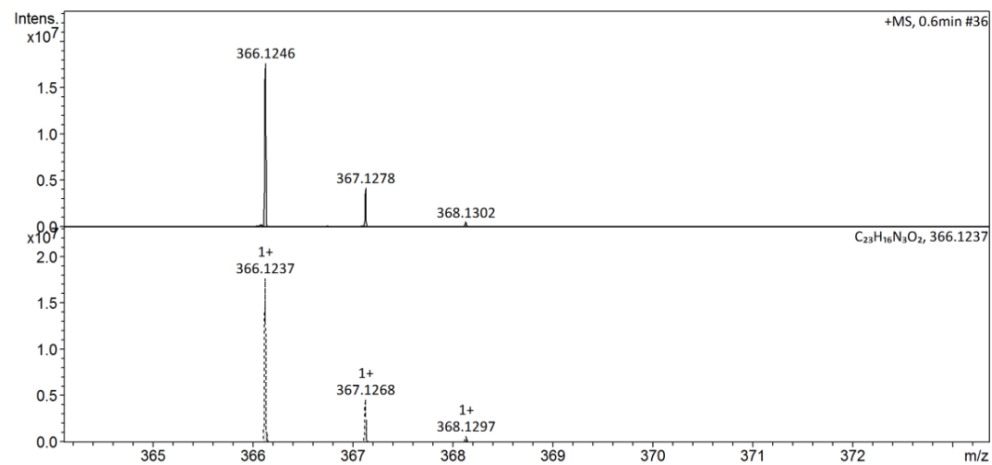
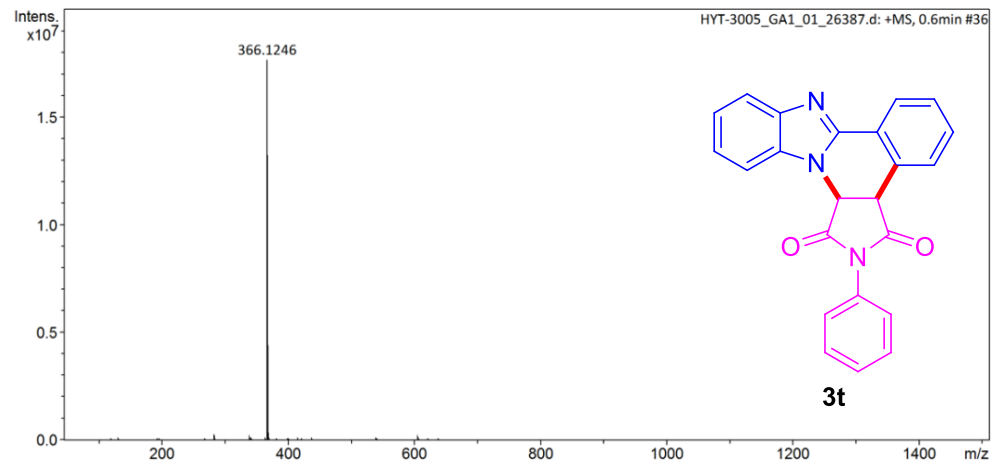
HRMS (ESI) of compound **3s**



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3t** in  $\text{CDCl}_3$ .



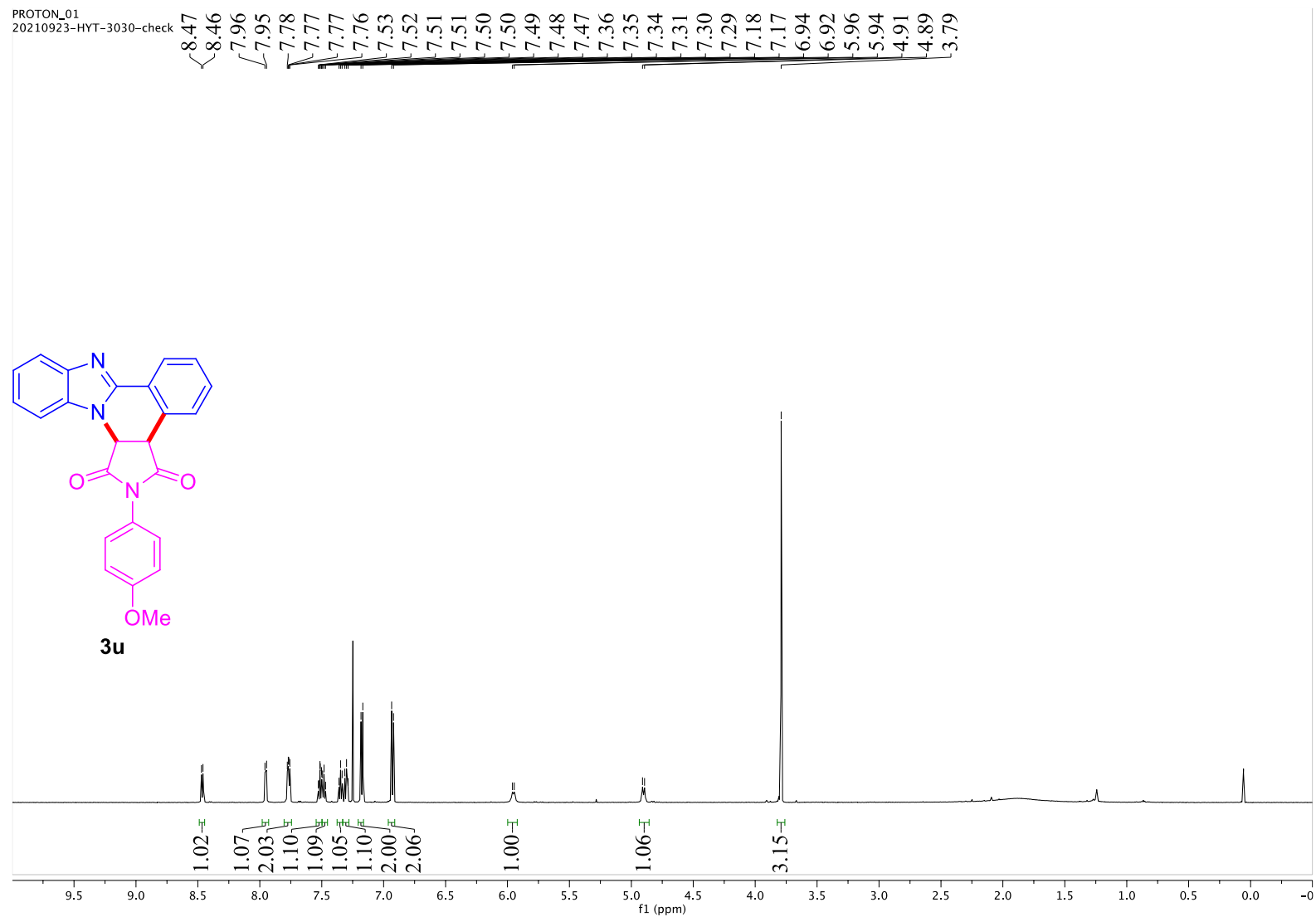
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3t** in  $\text{CDCl}_3$ .



## Display Report

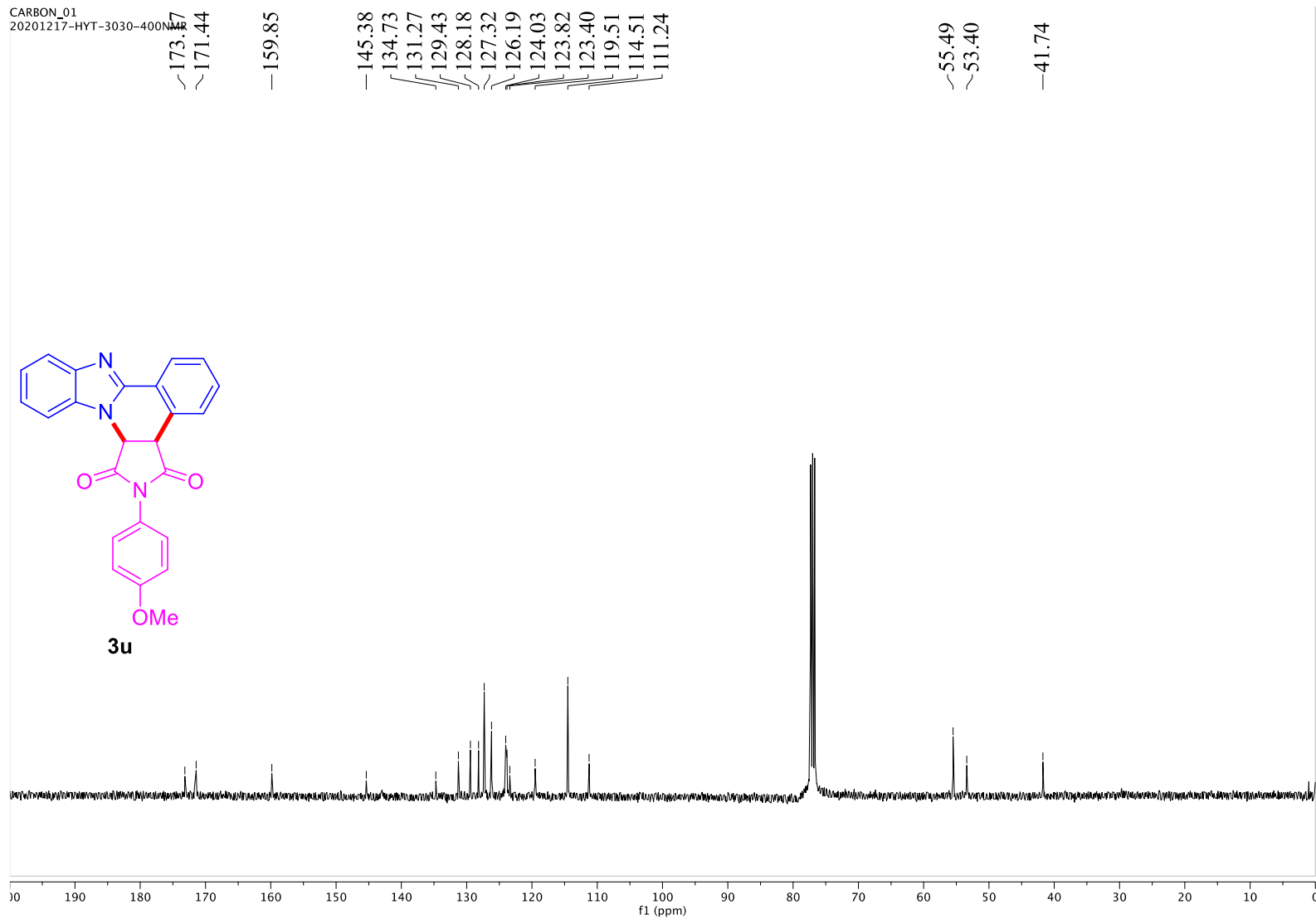
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
366.1246	1	C <sub>23</sub> H <sub>16</sub> N <sub>3</sub> O <sub>2</sub>	366.1237	2.6	12.9	1	100.00	17.5	even	ok	M

HRMS (ESI) of compound **3t**.

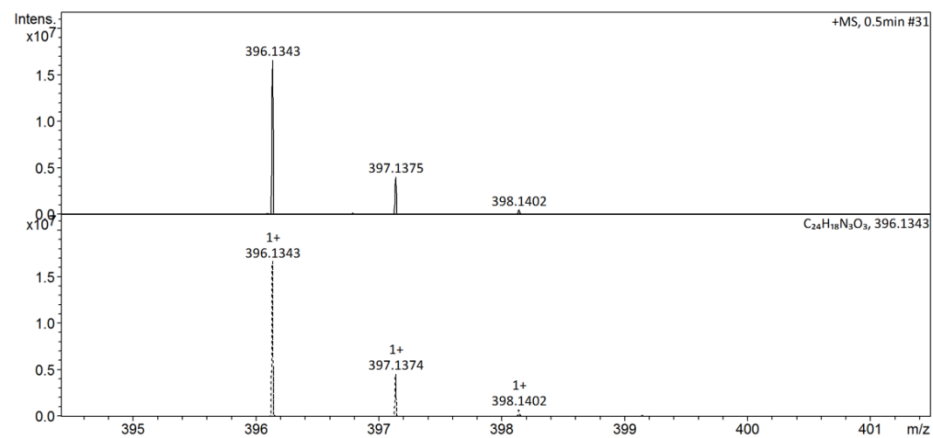
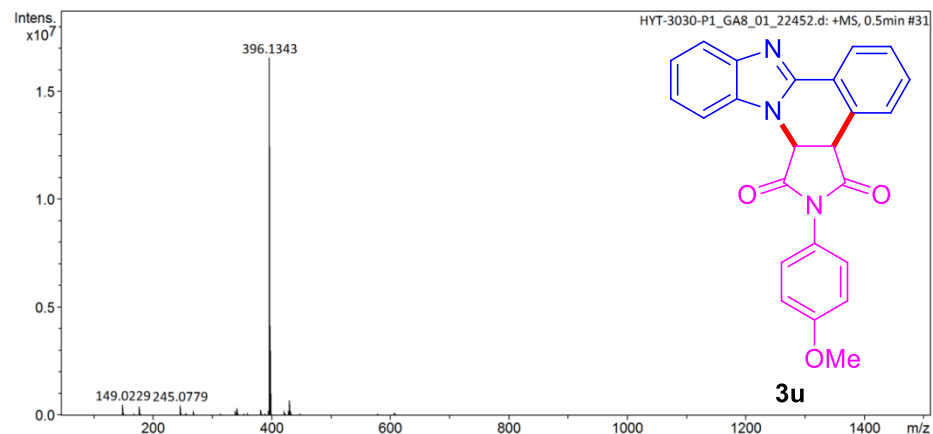


<sup>1</sup>H NMR spectrum (400 MHz) of compound **3u** in CDCl<sub>3</sub>.





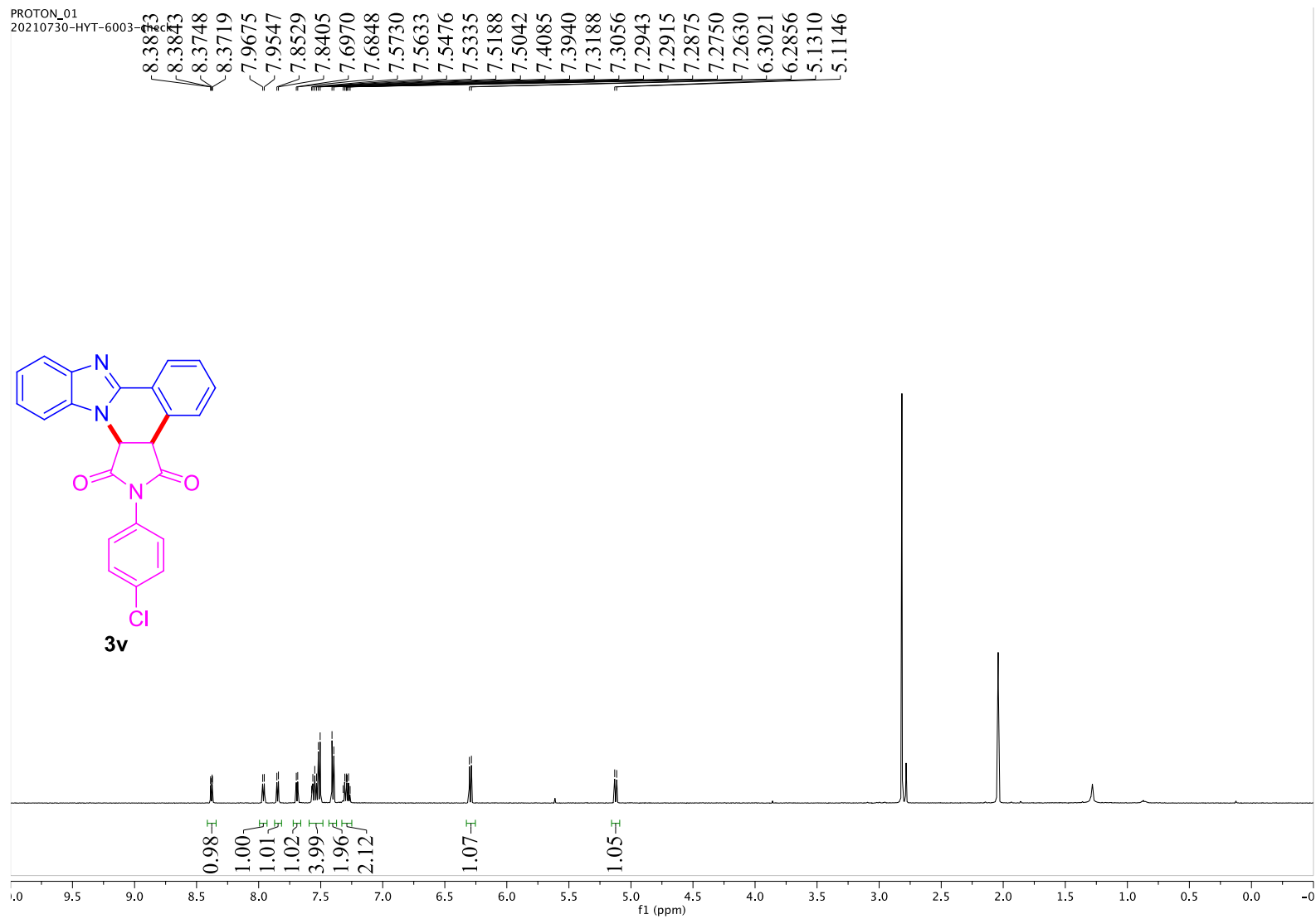
<sup>13</sup>C NMR spectrum (100 MHz) of compound **3u** in CDCl<sub>3</sub>.



## Display Report

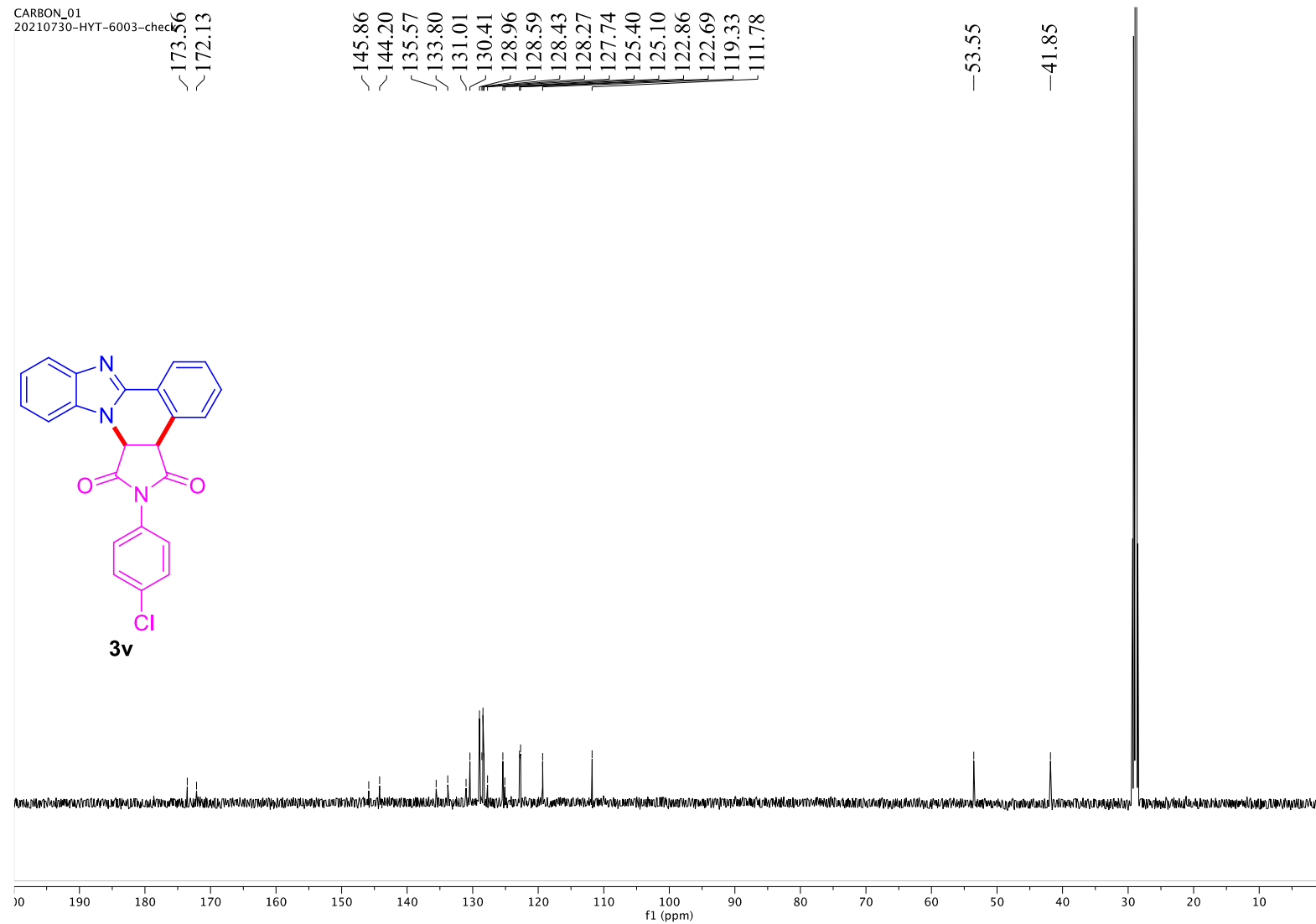
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
396.1343	1	C <sub>24</sub> H <sub>18</sub> N <sub>2</sub> O <sub>3</sub>	396.1343	0.0	17.3	1	100.00	17.5	even	ok	M+H

HRMS (ESI) of compound **3u**.

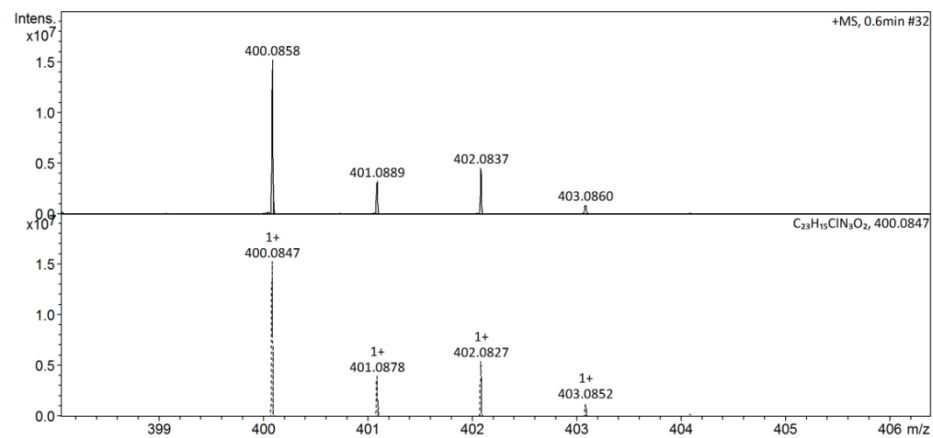
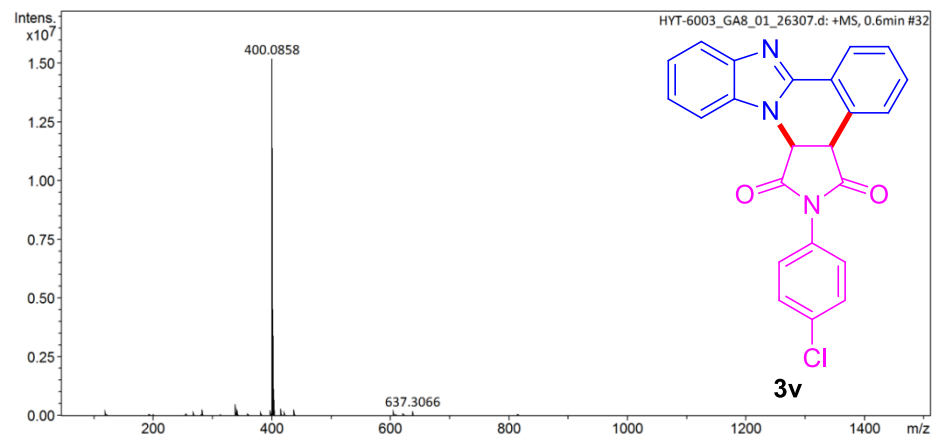


$^1\text{H}$  NMR spectrum (600 MHz) of compound **3v** in acetone- $d_6$ .

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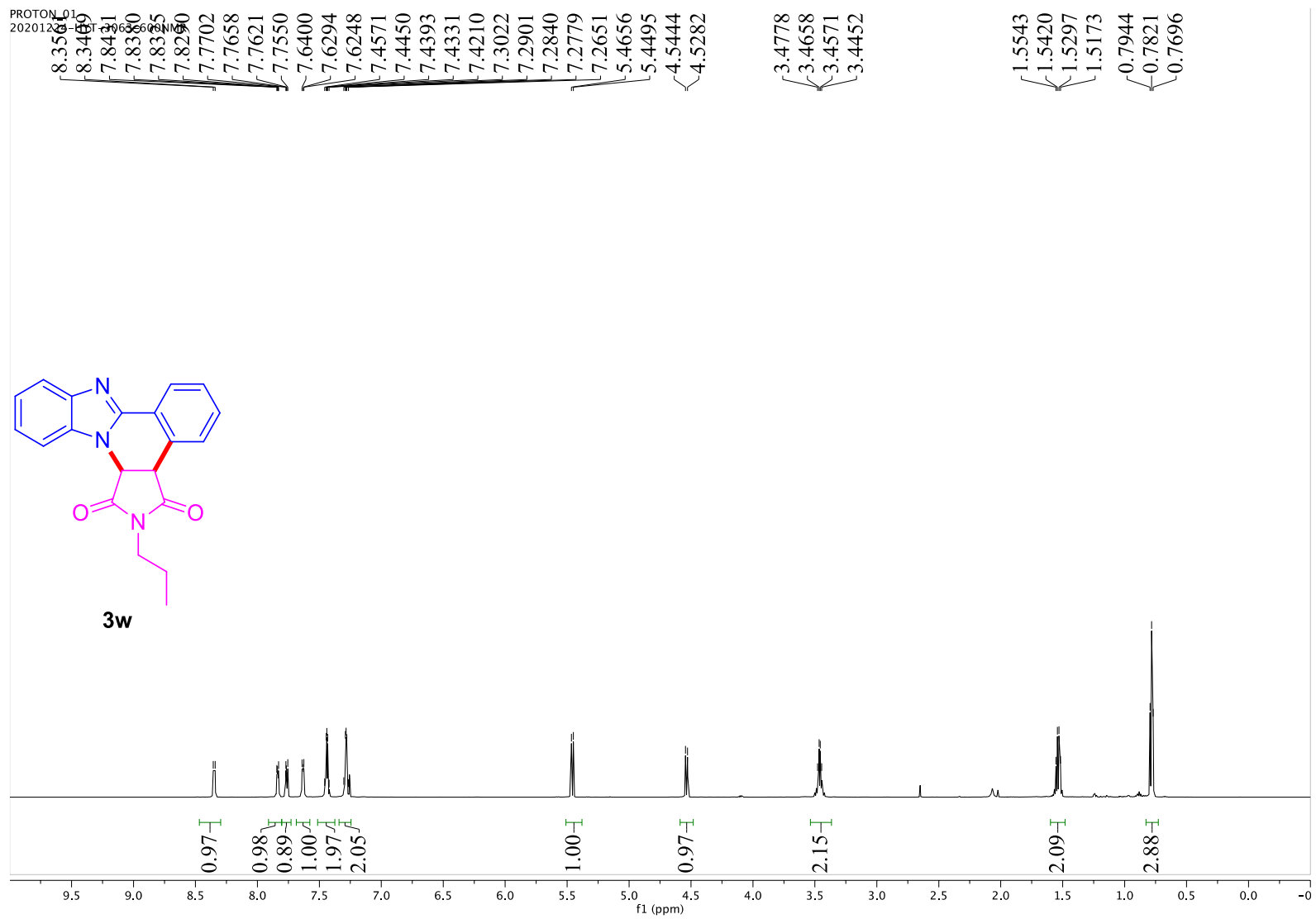
$^{13}\text{C}$  NMR spectrum (150 MHz) of compound **3v** in acetone- $d_6$ .



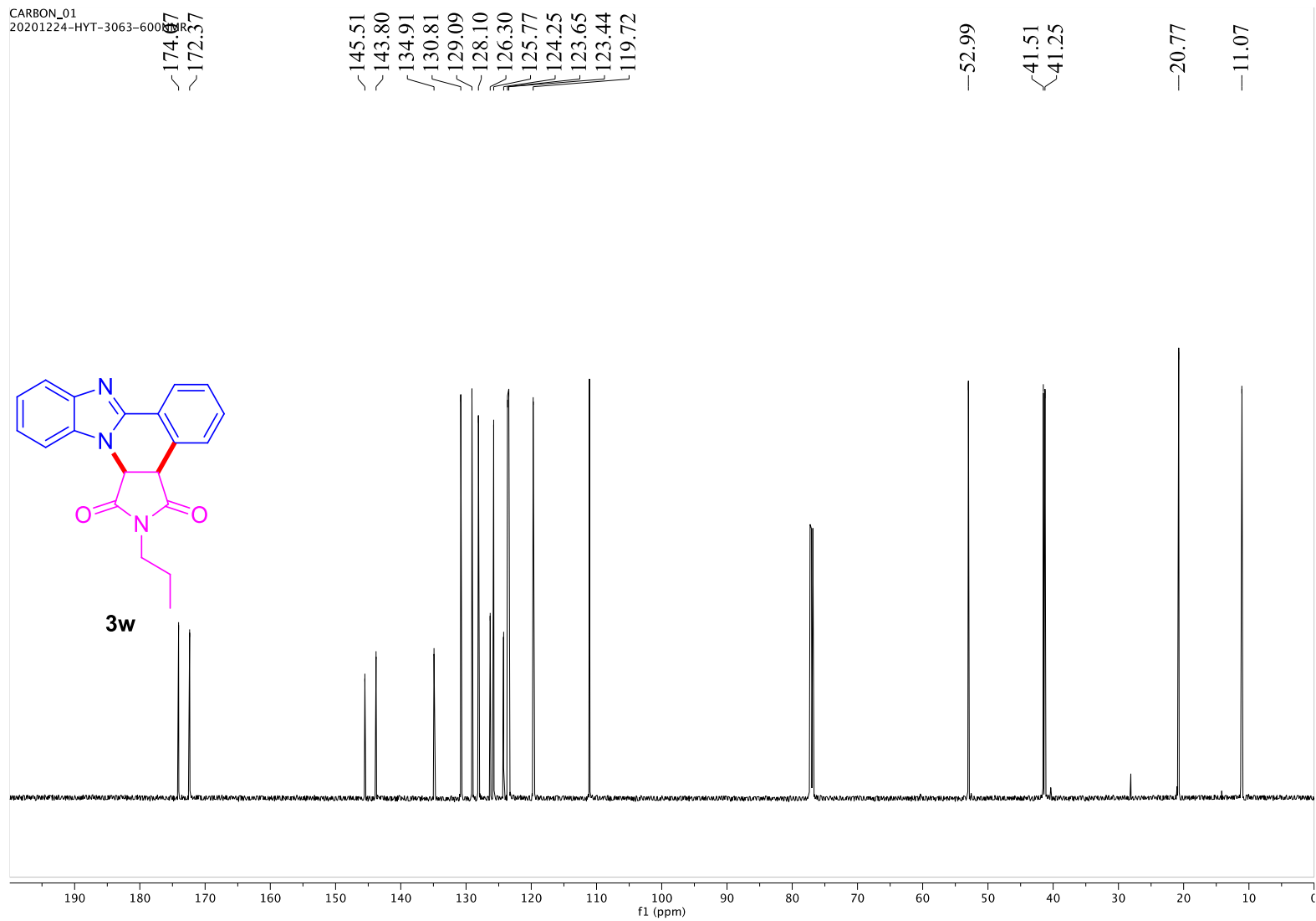
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
400.0858	1	C <sub>23</sub> H <sub>15</sub> ClN <sub>3</sub> O <sub>2</sub>	400.0847	-2.8	36.2	1	100.00	17.5	even	ok	M+H

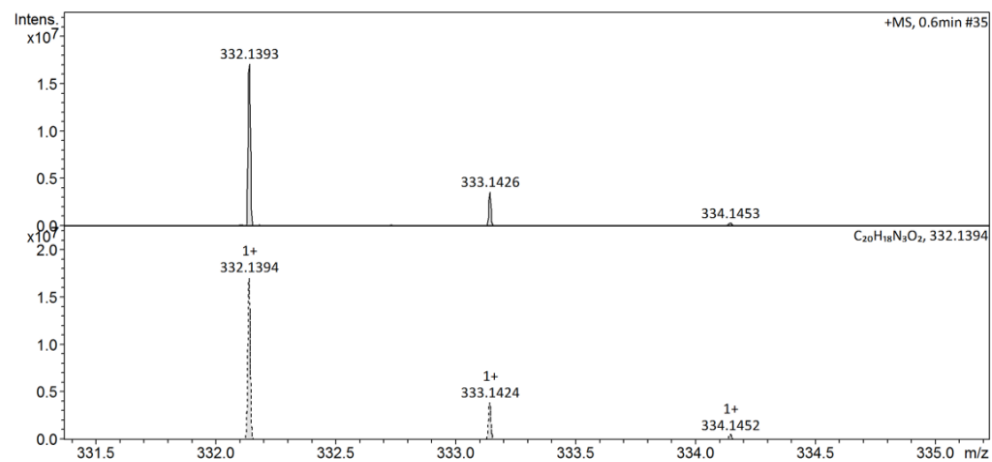
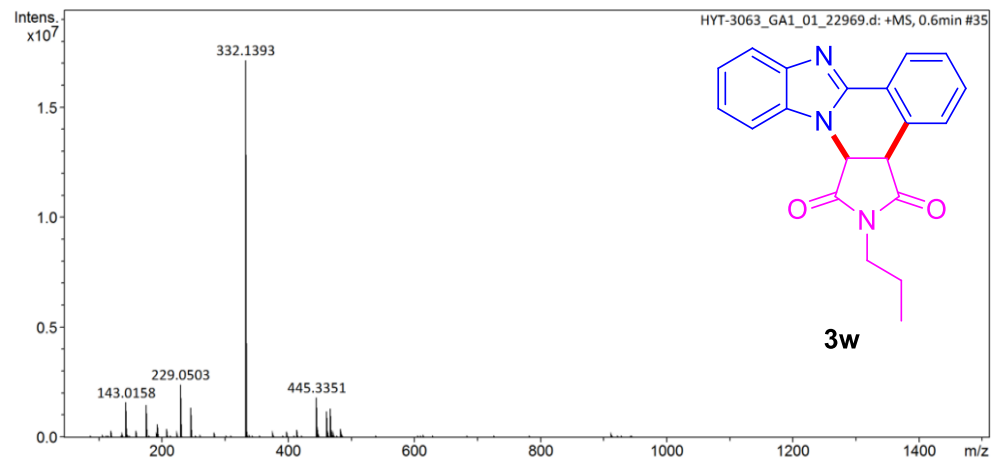
HRMS (ESI) of compound **3v**.



$^1\text{H}$  NMR spectrum (600 MHz) of compound **3w** in  $\text{CDCl}_3$ .



$^{13}\text{C}$  NMR spectrum (150 MHz) of compound **3w** in  $\text{CDCl}_3$ .

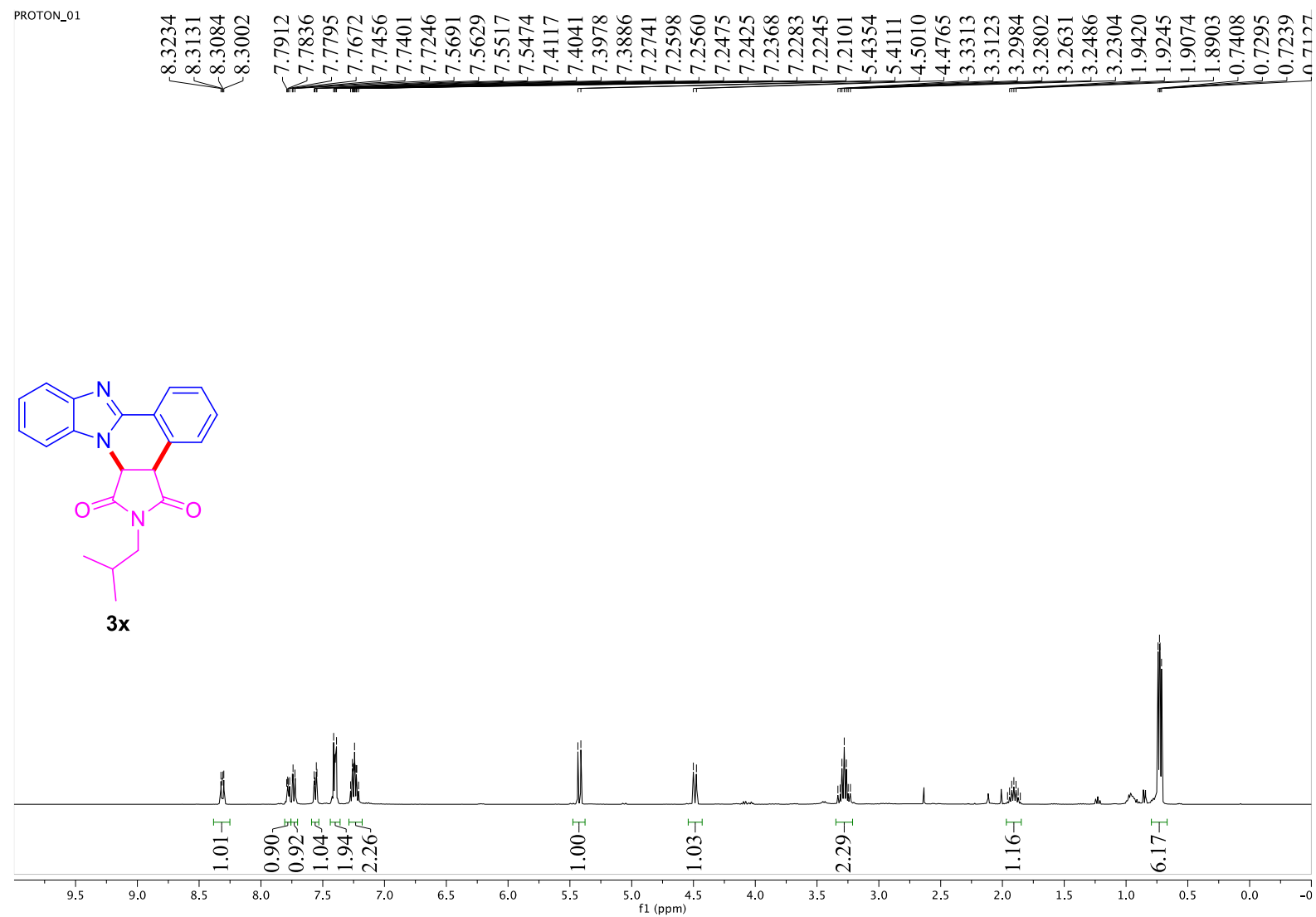


## Display Report

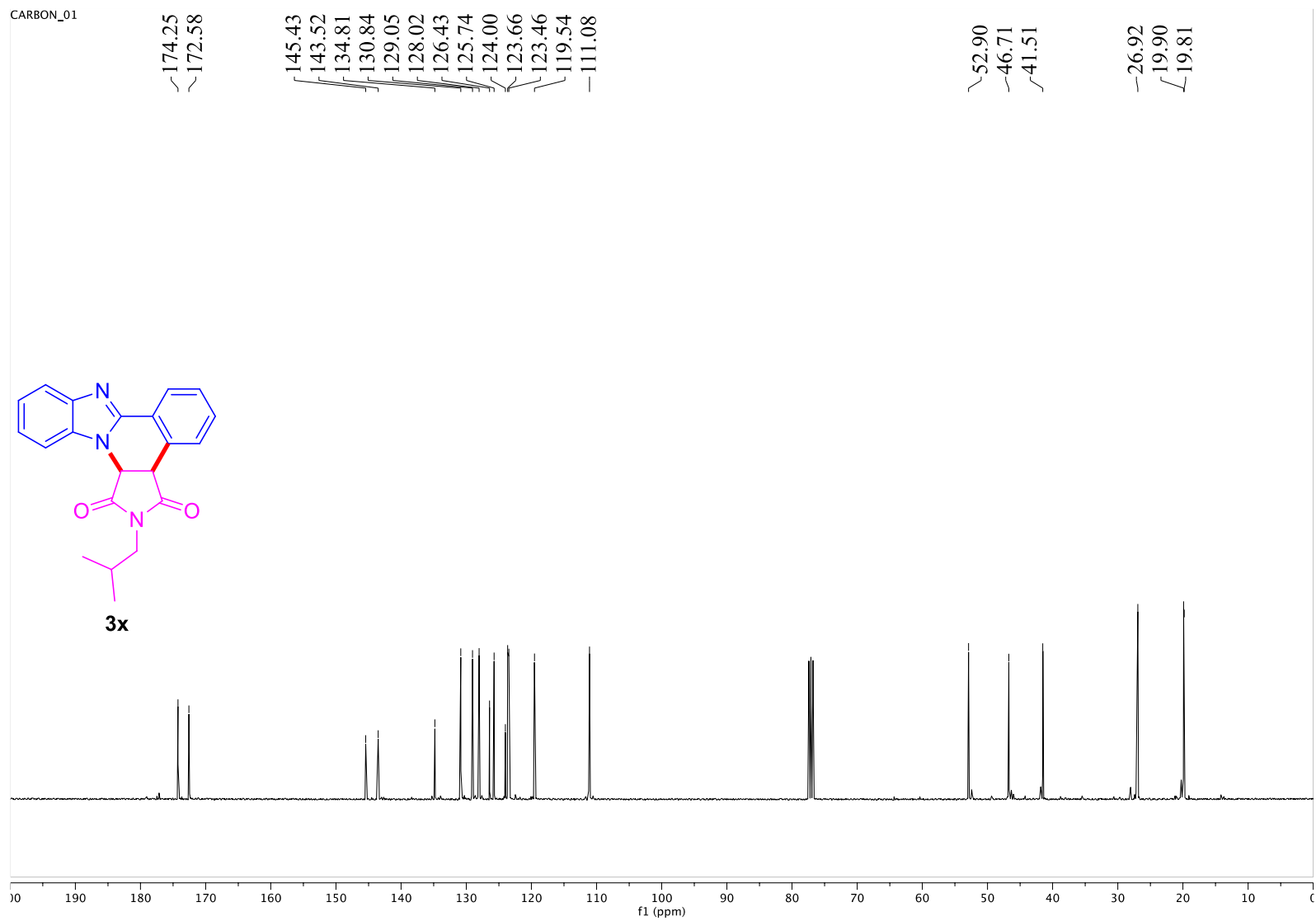
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
332.1393	1	C <sub>20</sub> H <sub>18</sub> N <sub>3</sub> O <sub>2</sub>	332.1394	0.1	11.5	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **3w**.

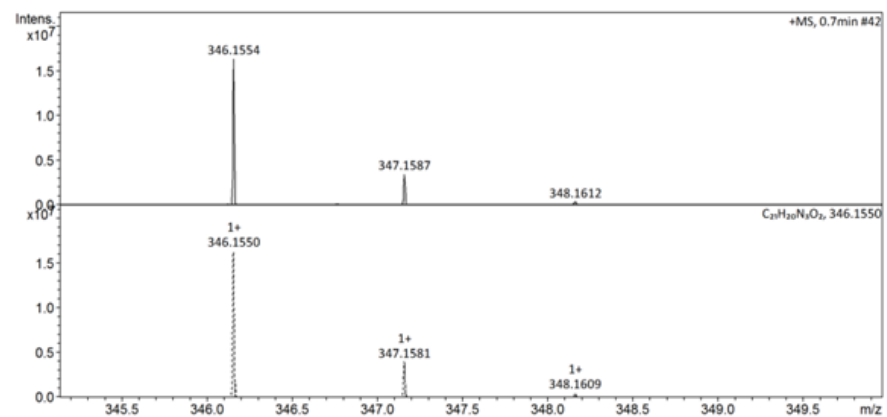
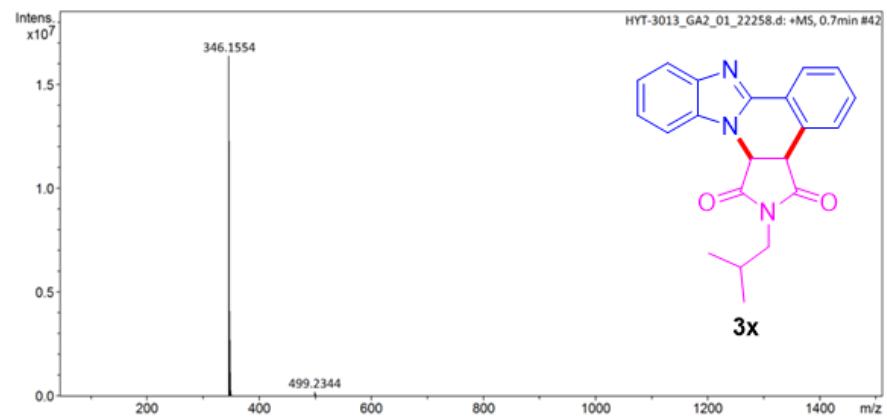




$^1\text{H}$  NMR spectrum (400 MHz) of compound **3x** in  $\text{CDCl}_3$ .



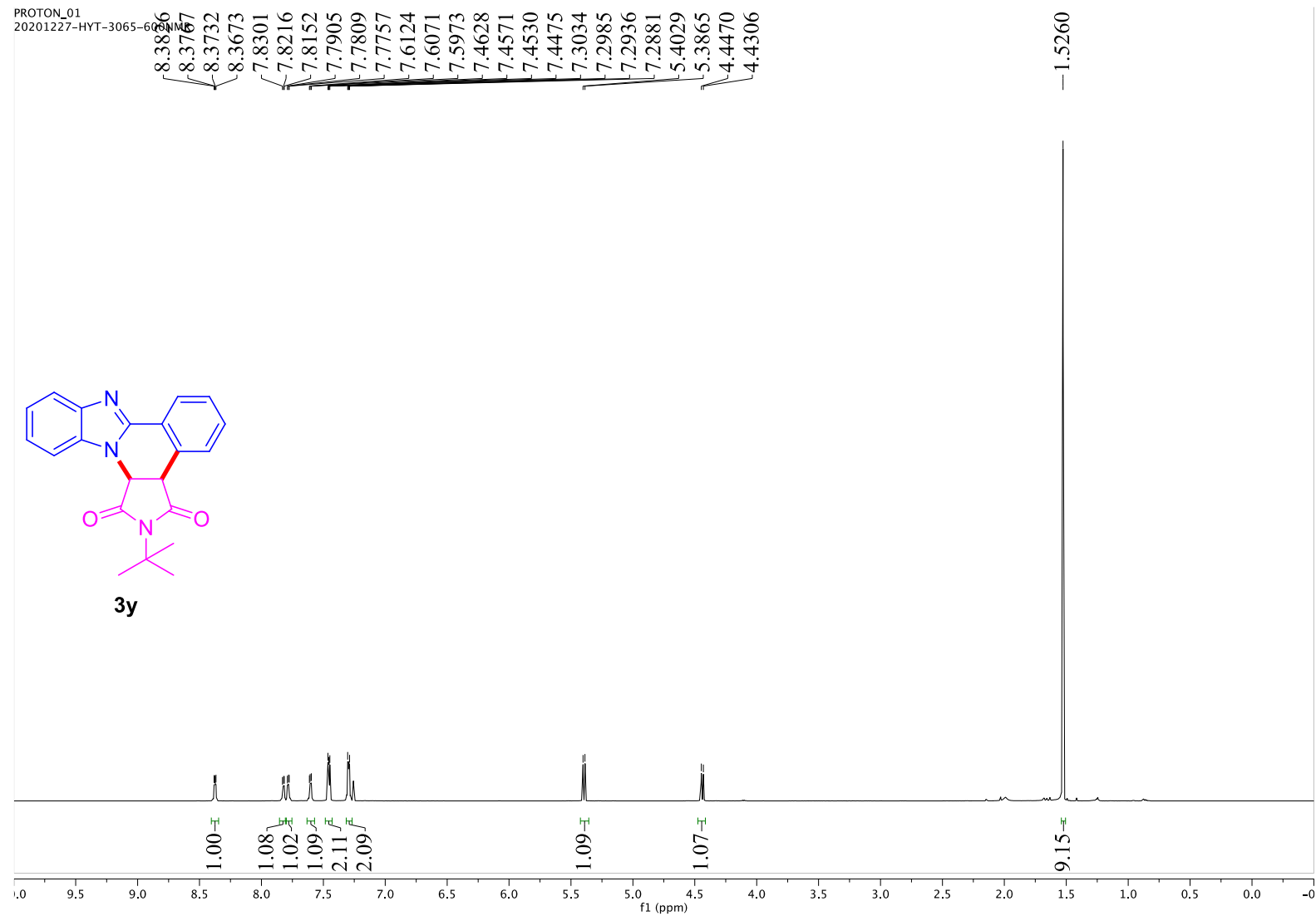
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3x** in  $\text{CDCl}_3$ .



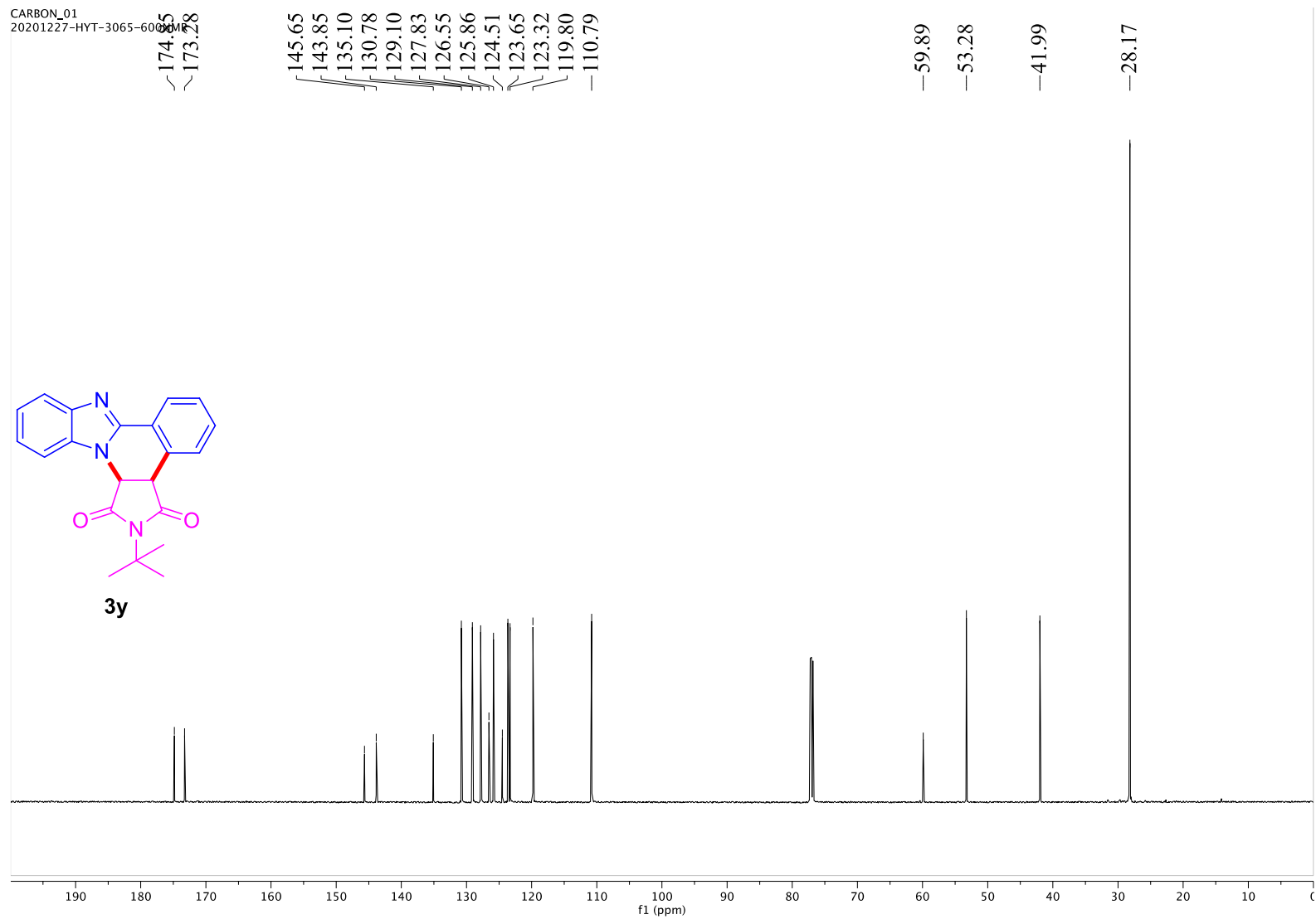
### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
346.1554	1	C <sub>21</sub> H <sub>20</sub> N <sub>3</sub> O <sub>2</sub>	346.1550	1.3	17.5	1	100.00	13.5	even	ok	M+H

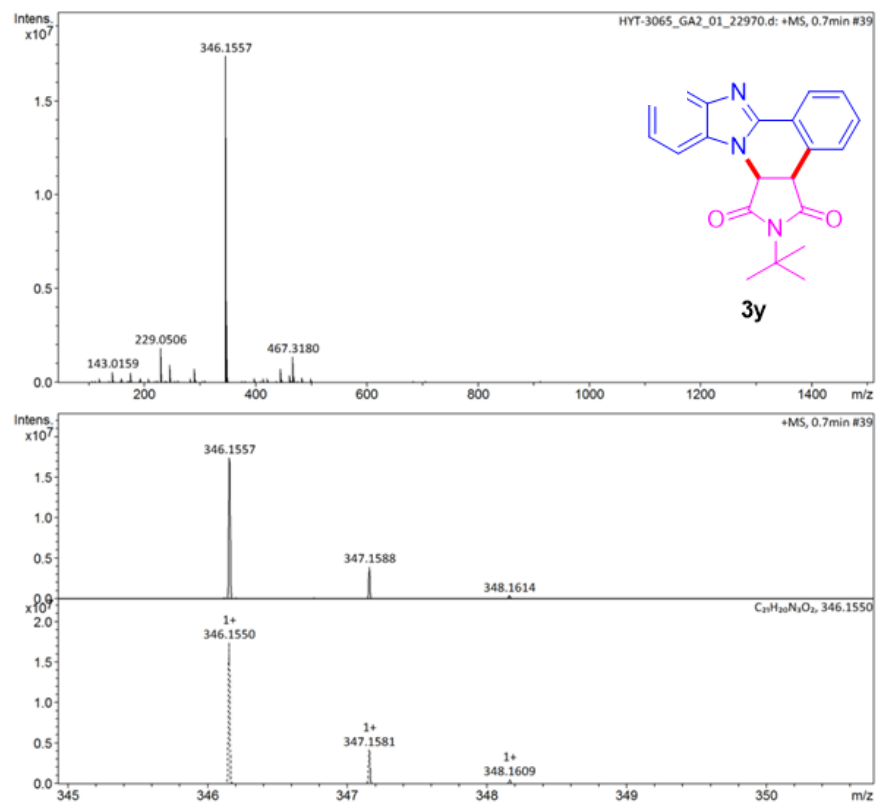
HRMS (ESI) of compound **3x**



$^1\text{H}$  NMR spectrum (600 MHz) of compound **3y** in  $\text{CDCl}_3$ .



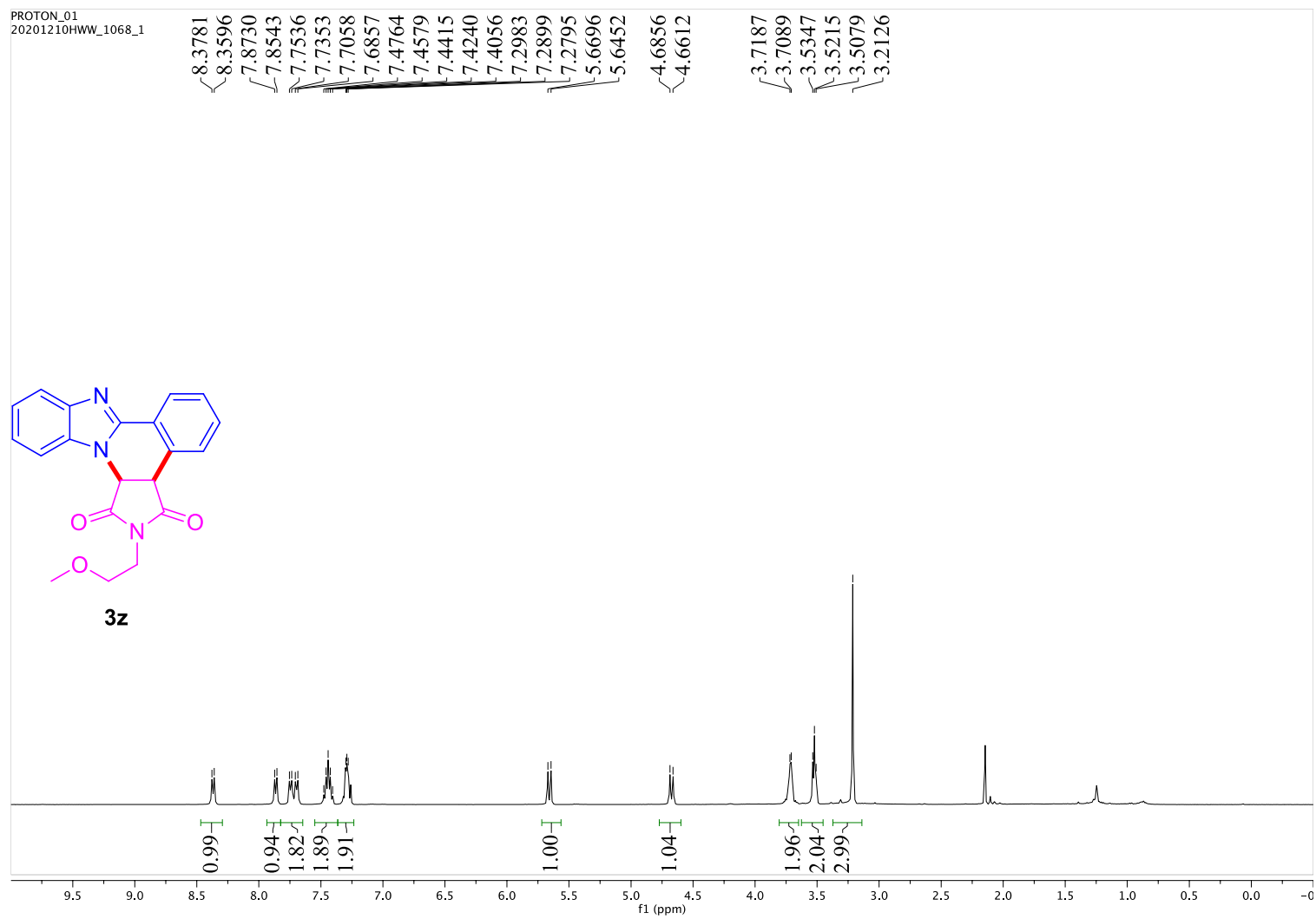
<sup>13</sup>C NMR spectrum (150 MHz) of compound **3y** in CDCl<sub>3</sub>.



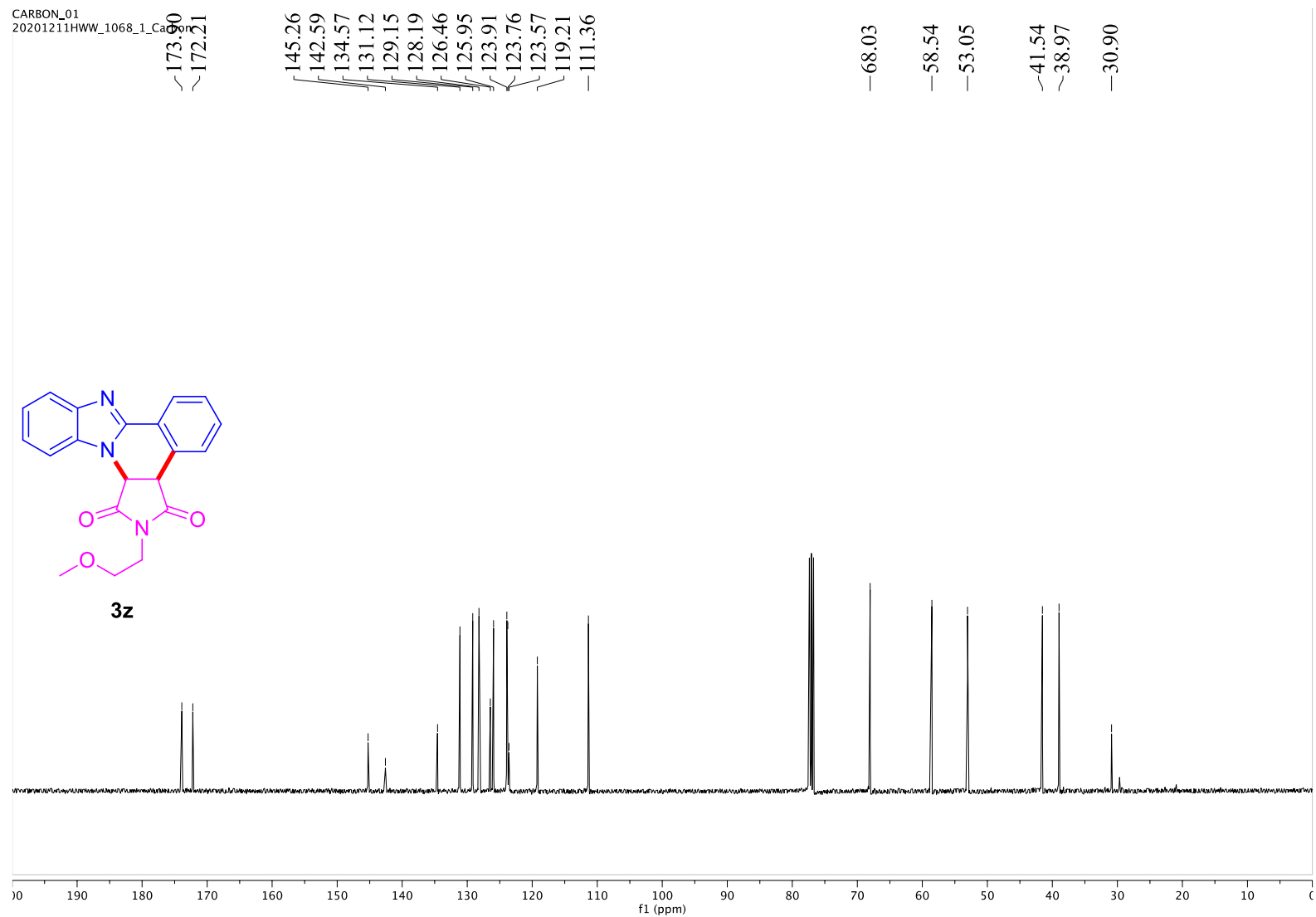
### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
346.1557	1	C <sub>21</sub> H <sub>20</sub> N <sub>3</sub> O <sub>2</sub>	346.1550	-1.9	8.8	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **3y**

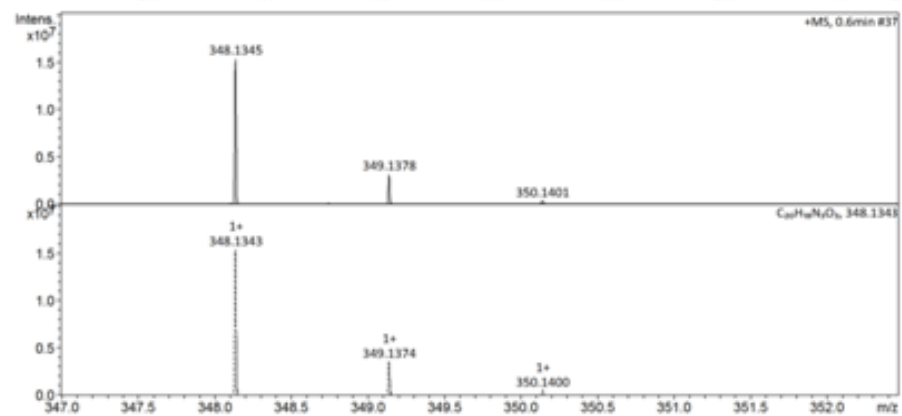
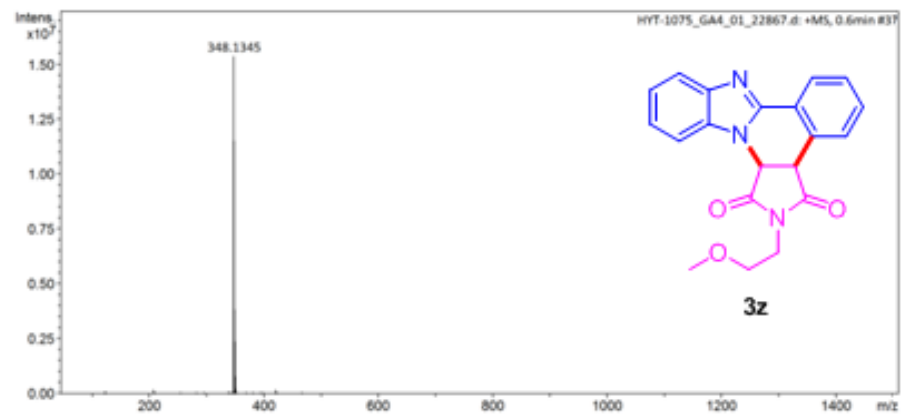


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3z** in  $\text{CDCl}_3$ .



<sup>13</sup>C NMR spectrum (100 MHz) of compound **3z** in CDCl<sub>3</sub>.



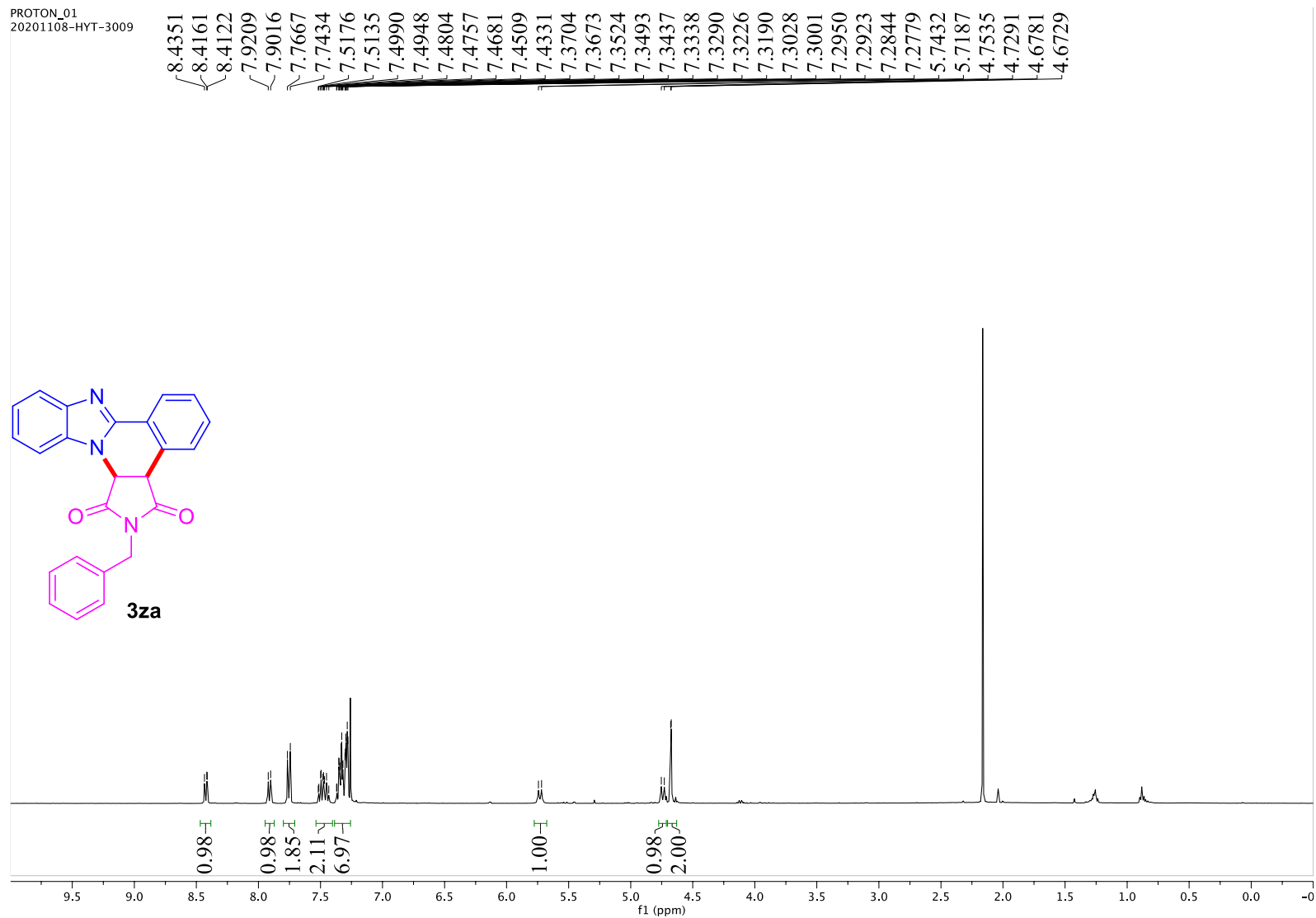


### Display Report

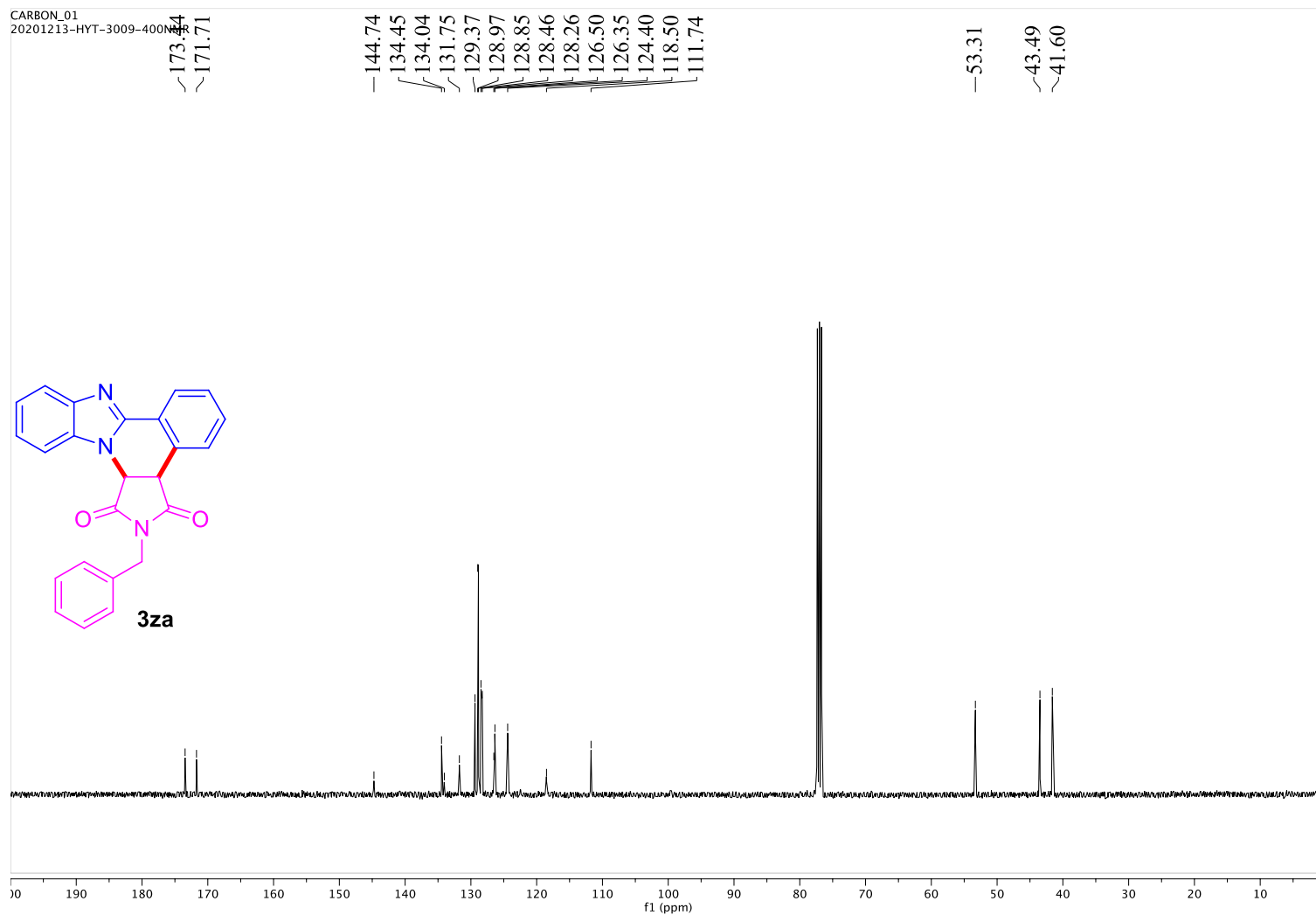
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
348.1345	1	C <sub>20</sub> H <sub>18</sub> N <sub>3</sub> O <sub>3</sub>	348.1343	0.7	14.7	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **3z**

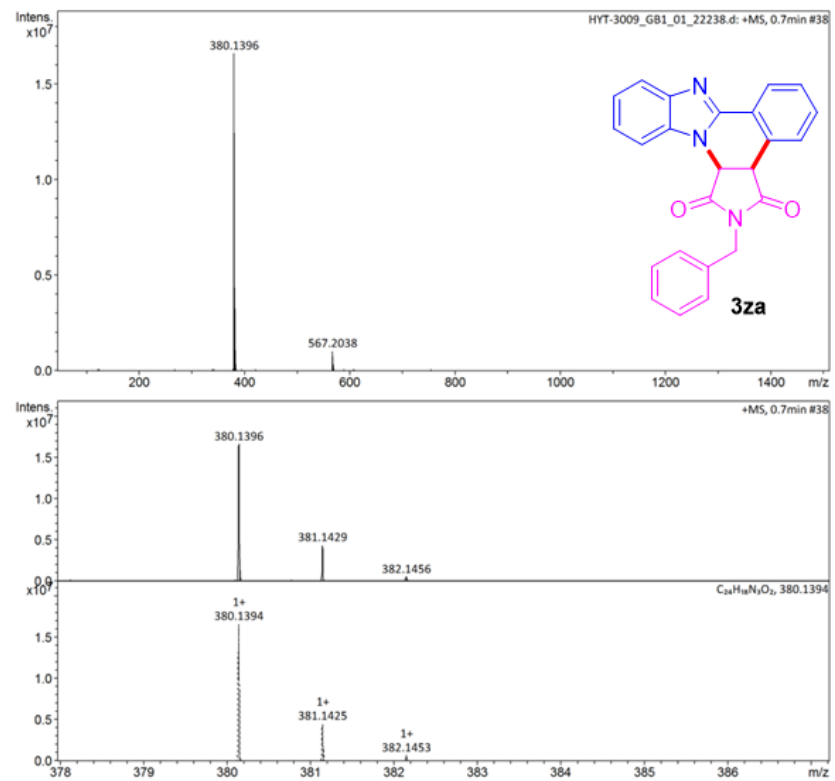
PROTON\_01  
20201108-HYT-3009



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3za** in  $\text{CDCl}_3$ .



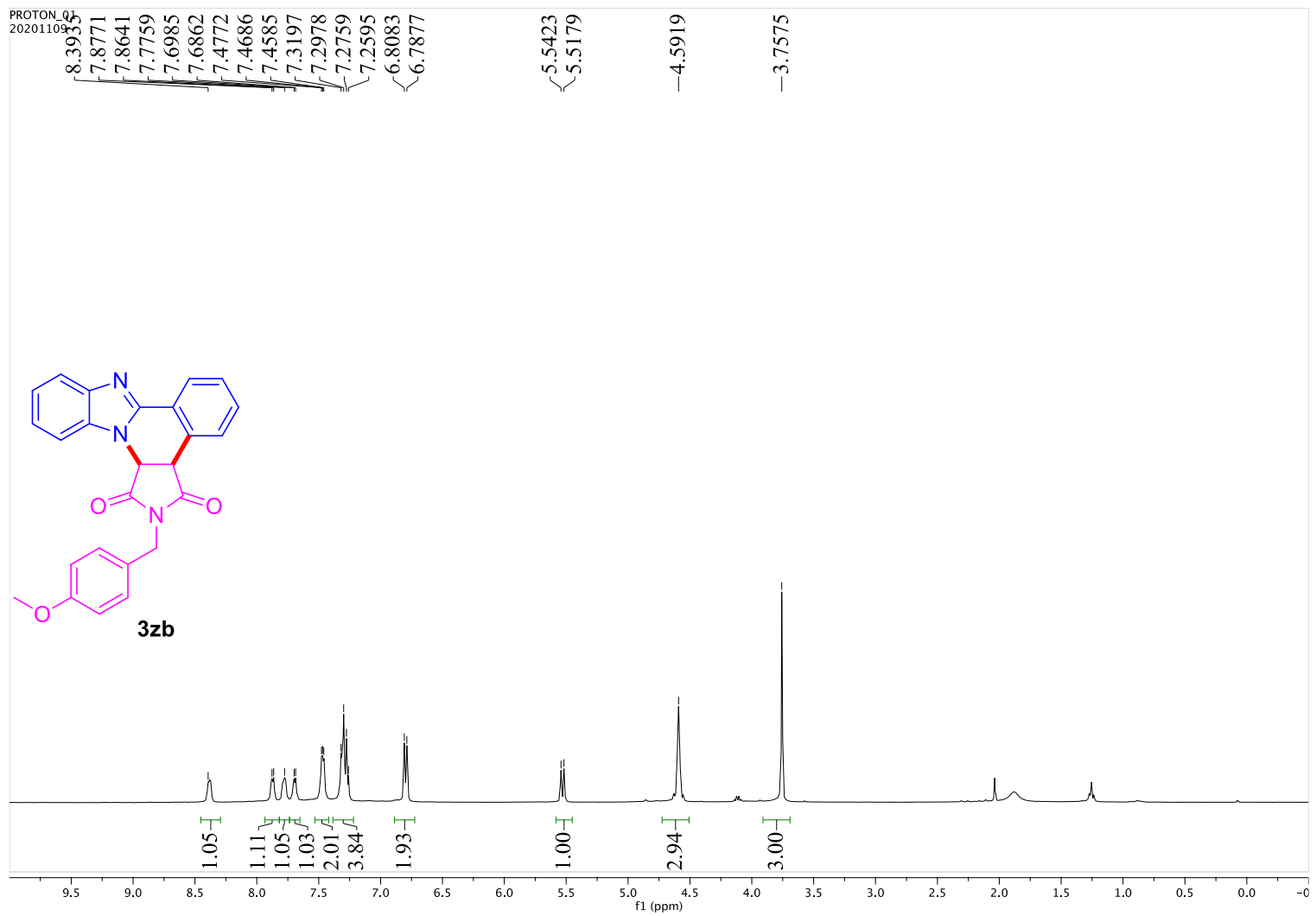
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3za** in  $\text{CDCl}_3$ .



### Display Report

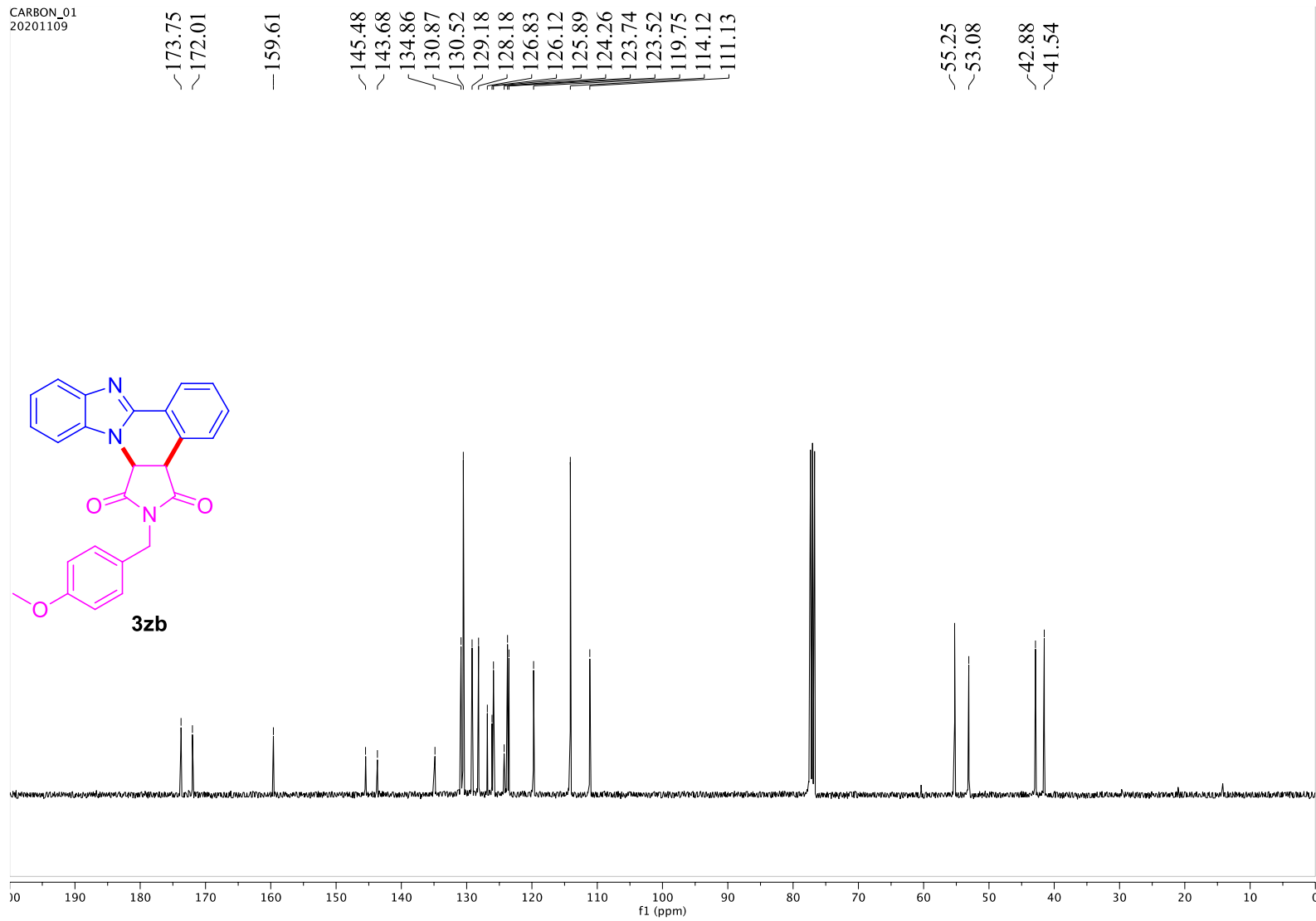
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
380.1396	1	C <sub>24</sub> H <sub>18</sub> N <sub>2</sub> O <sub>2</sub>	380.1394	0.6	7.0	1	100.00	17.5	even	ok	M+H

HRMS (ESI) of compound **3za**

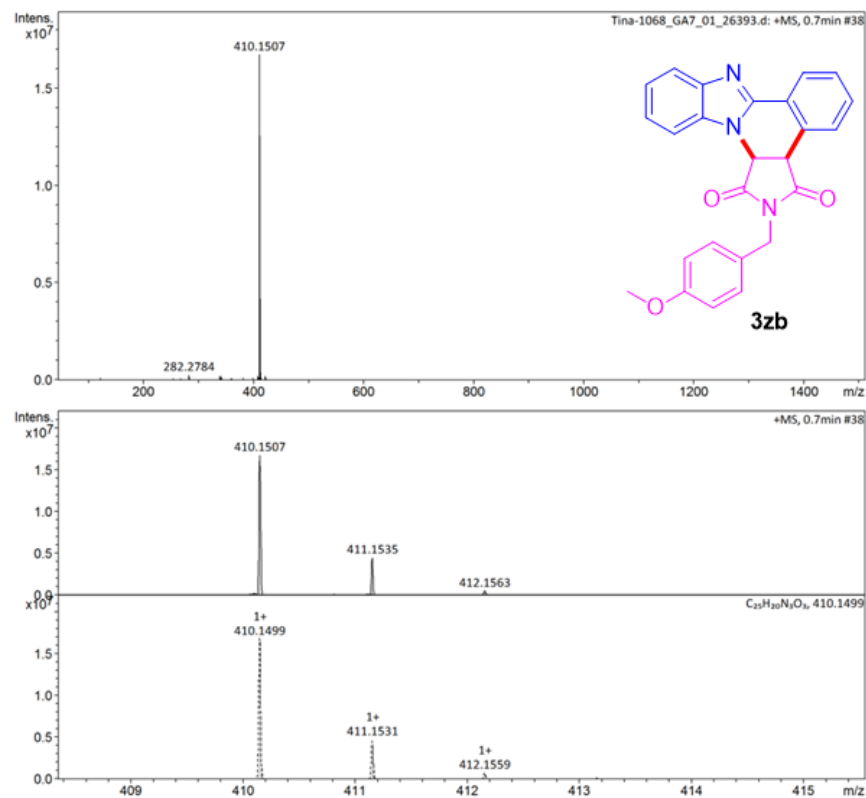


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3zb** in  $\text{CDCl}_3$ .

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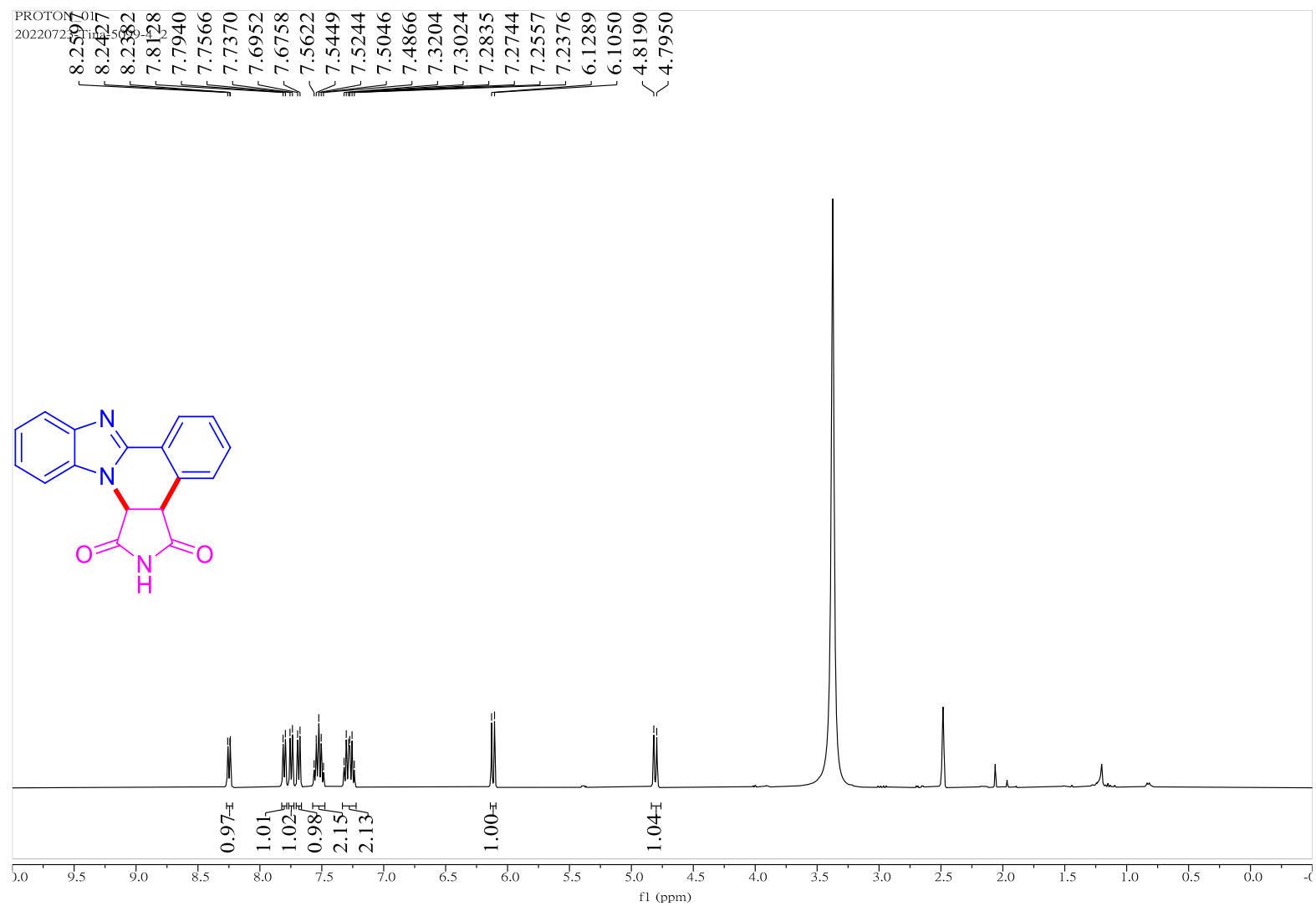
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3zb** in  $\text{CDCl}_3$ .



### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
410.1507	1	C <sub>25</sub> H <sub>20</sub> N <sub>3</sub> O <sub>3</sub>	410.1499	-1.8	9.7	1	100.00	17.5	even	ok	M+H

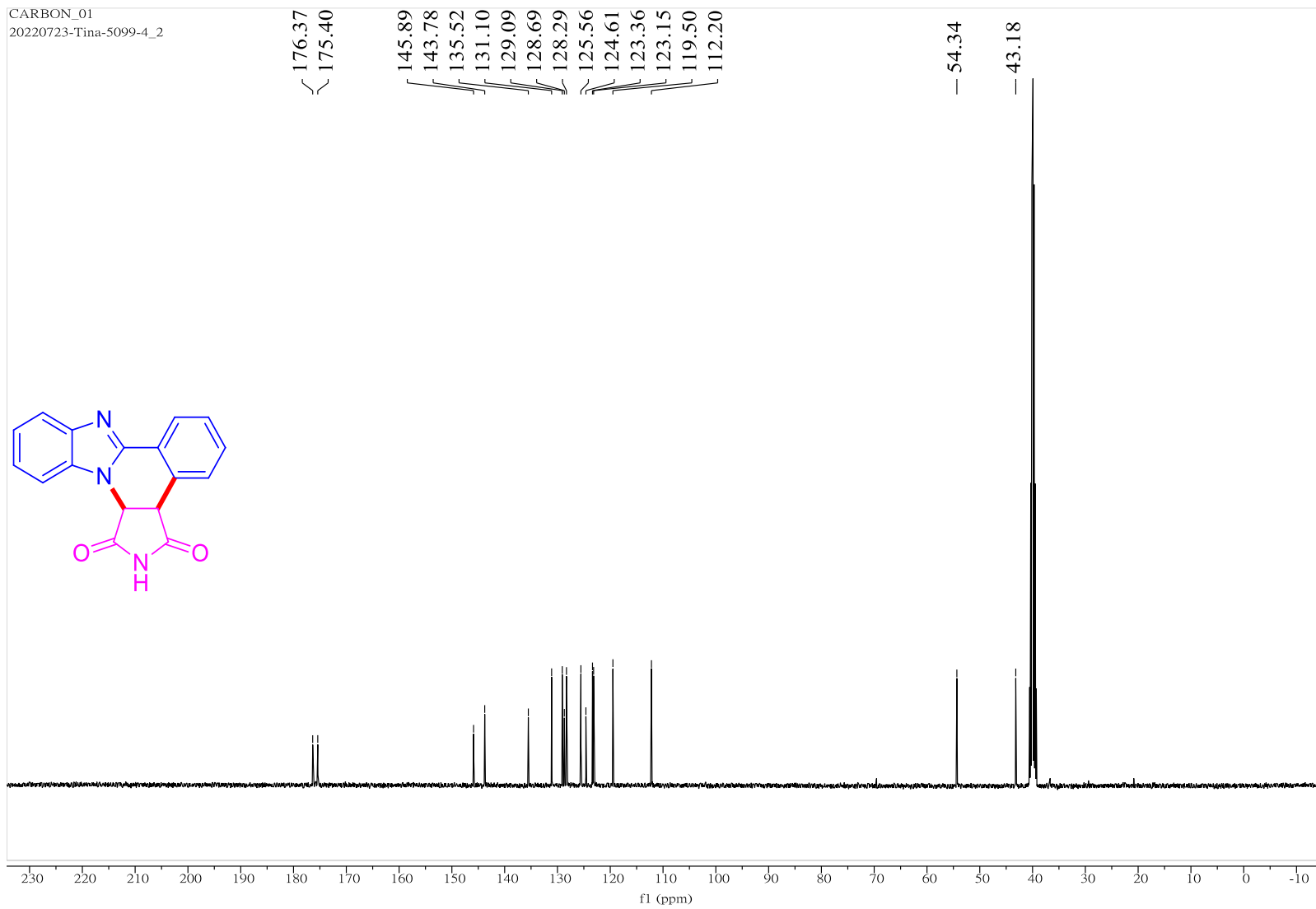
HRMS (ESI) of compound **3zb**



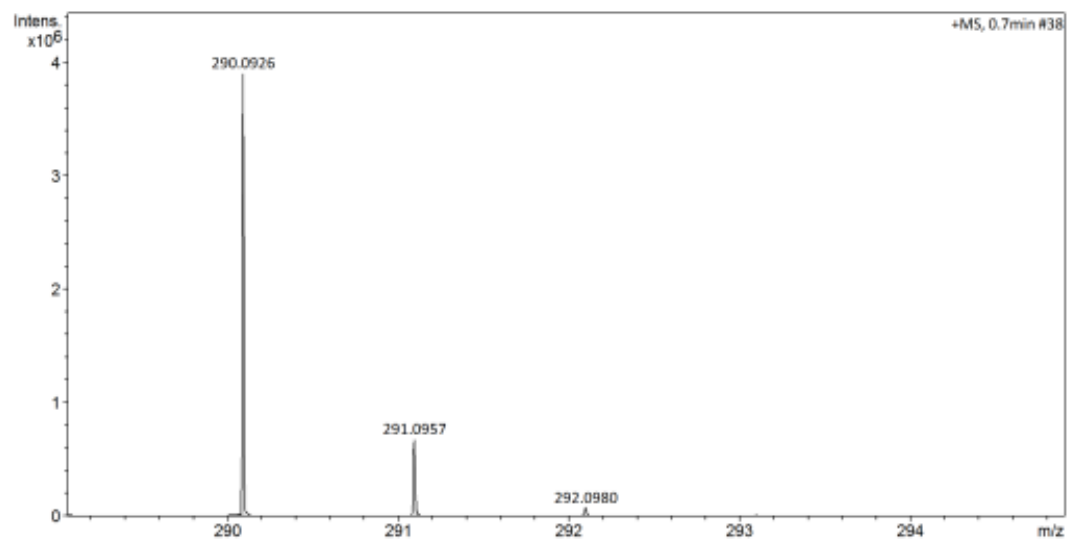
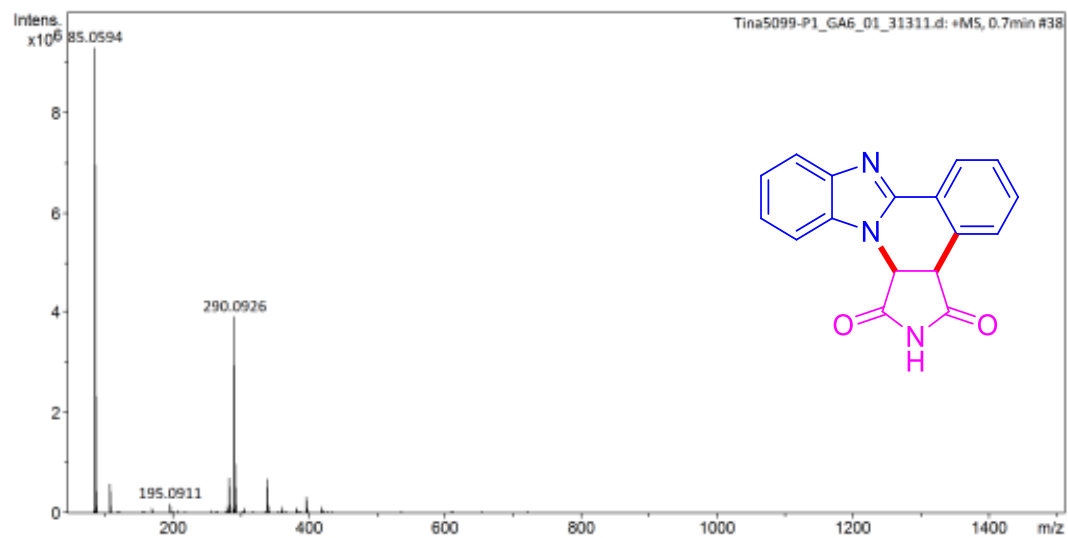
$^1\text{H}$  NMR spectrum (400 MHz) of compound **3zc** in  $\text{DMSO-}d_6$ .



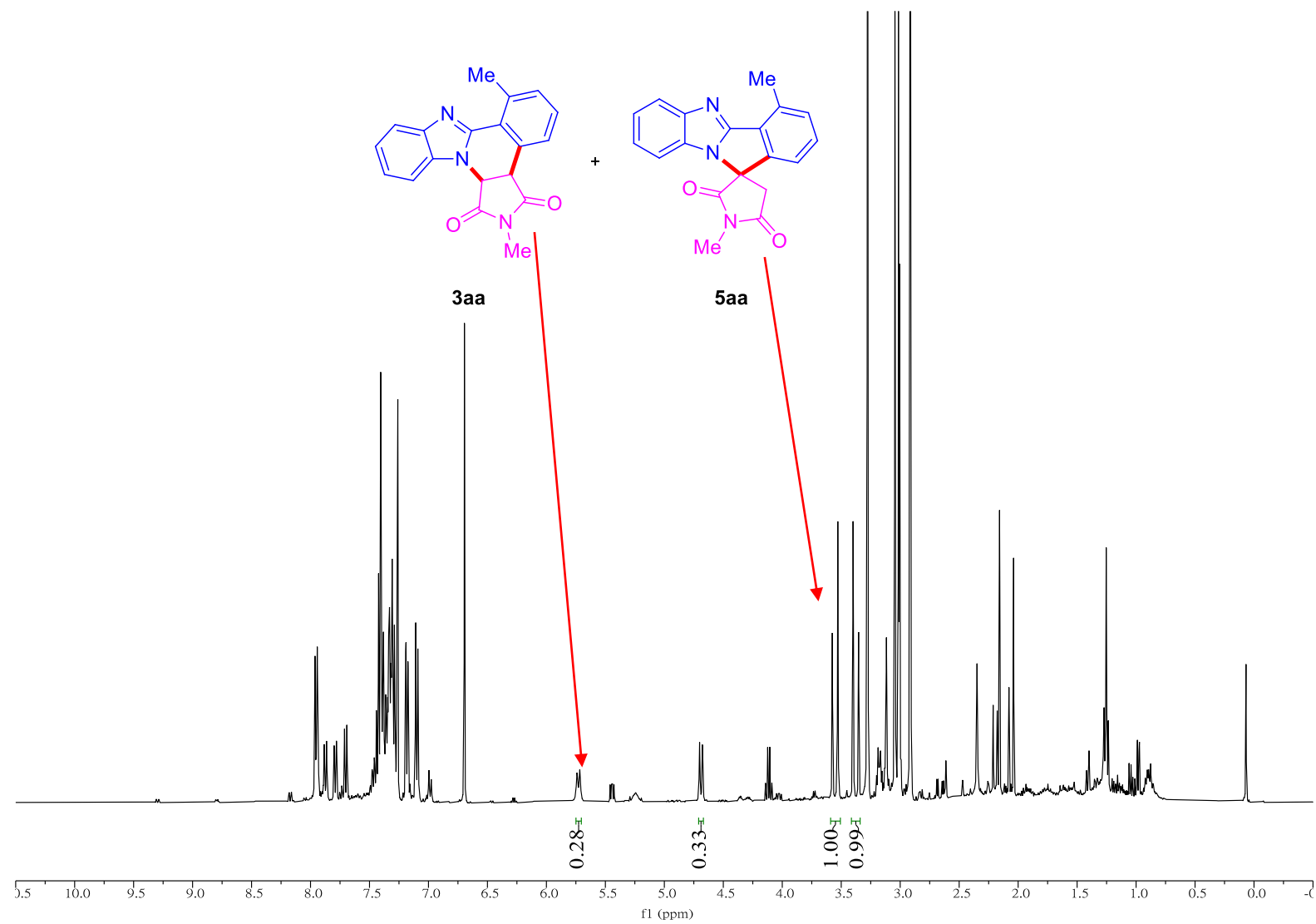
CARBON\_01  
20220723-Tina-5099-4\_2



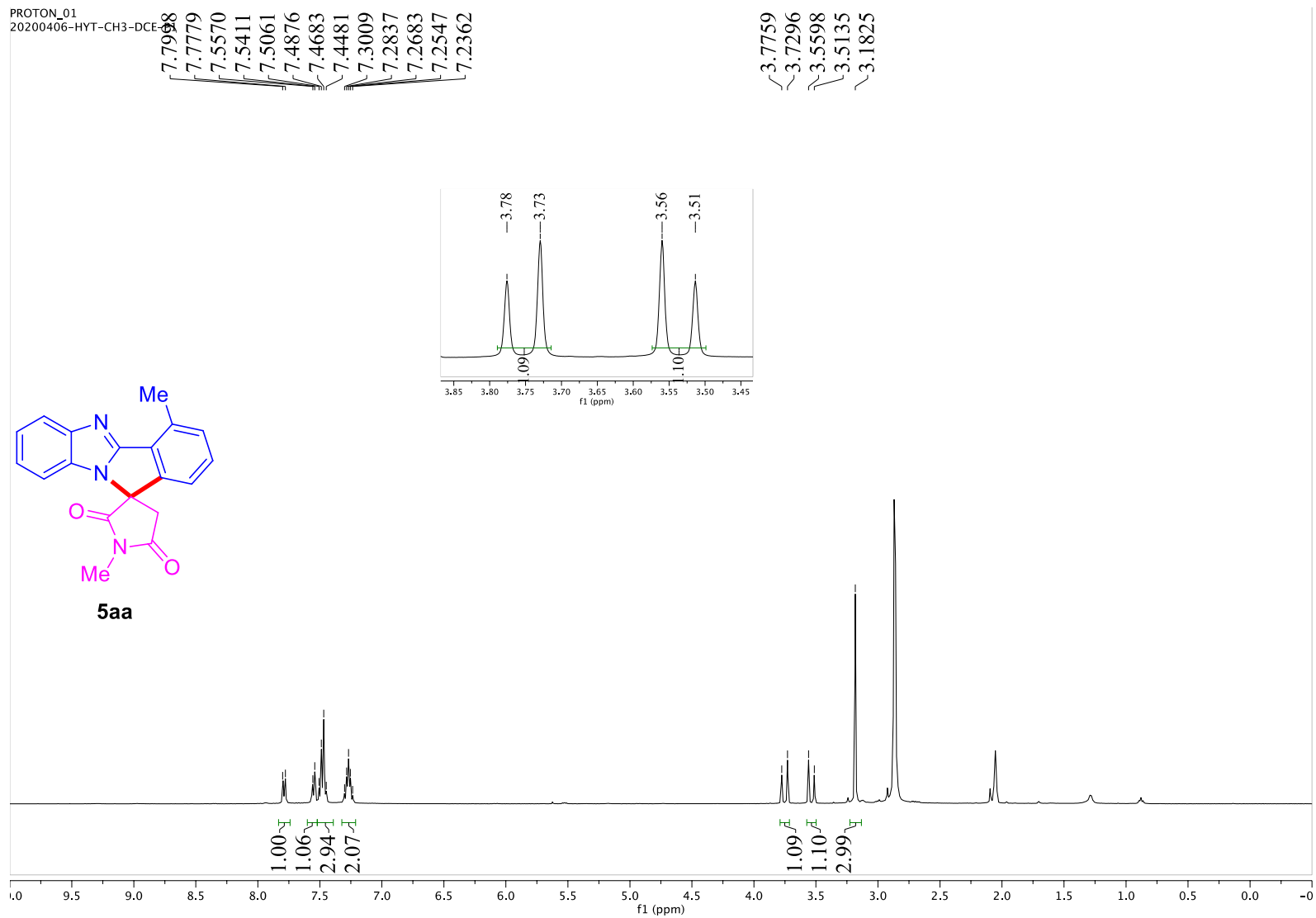
$^{13}\text{C}$  NMR spectrum (150 MHz) of compound **3zc** in  $\text{DMSO-}d_6$ .



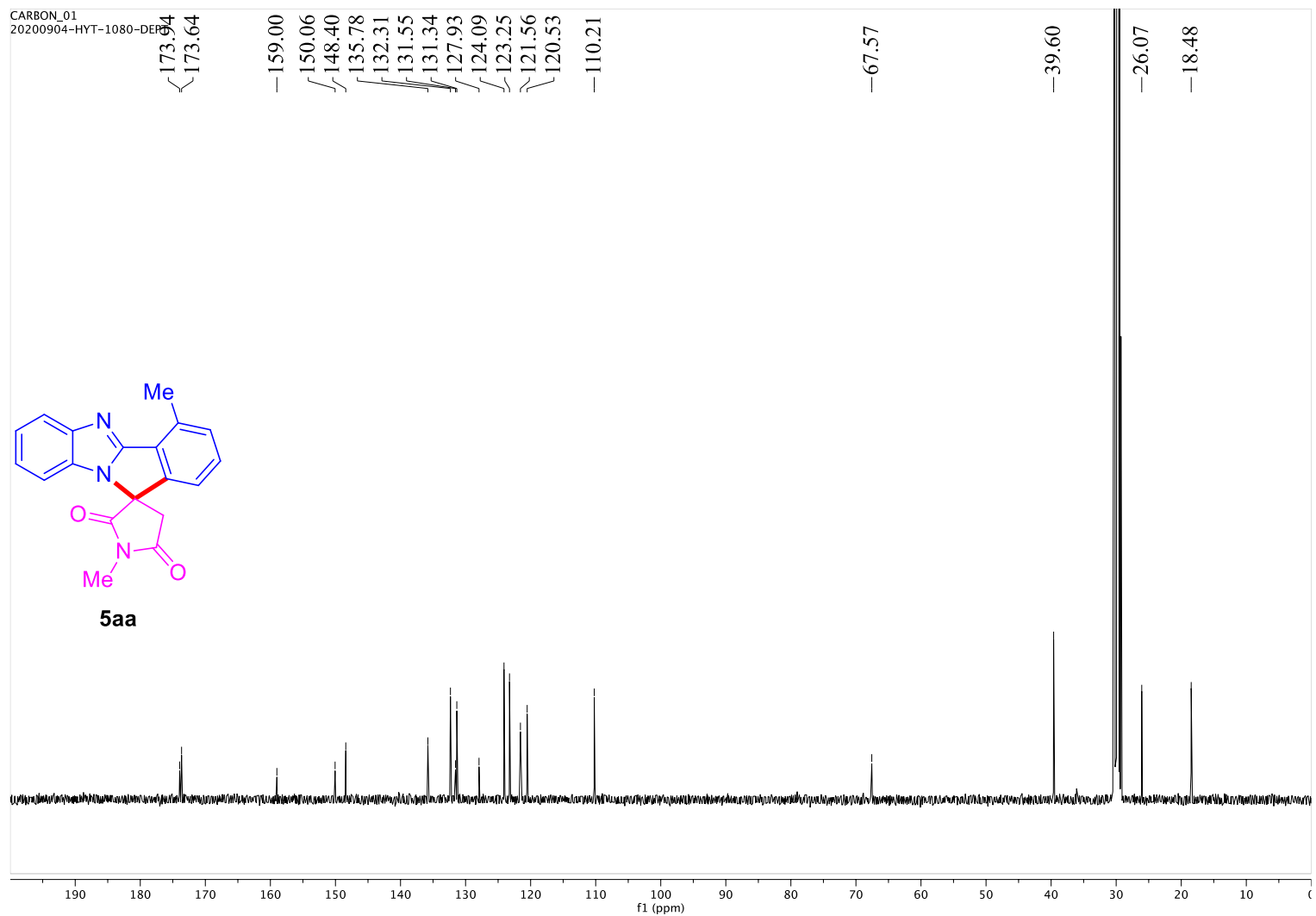
HRMS (ESI) of compound **3zc**



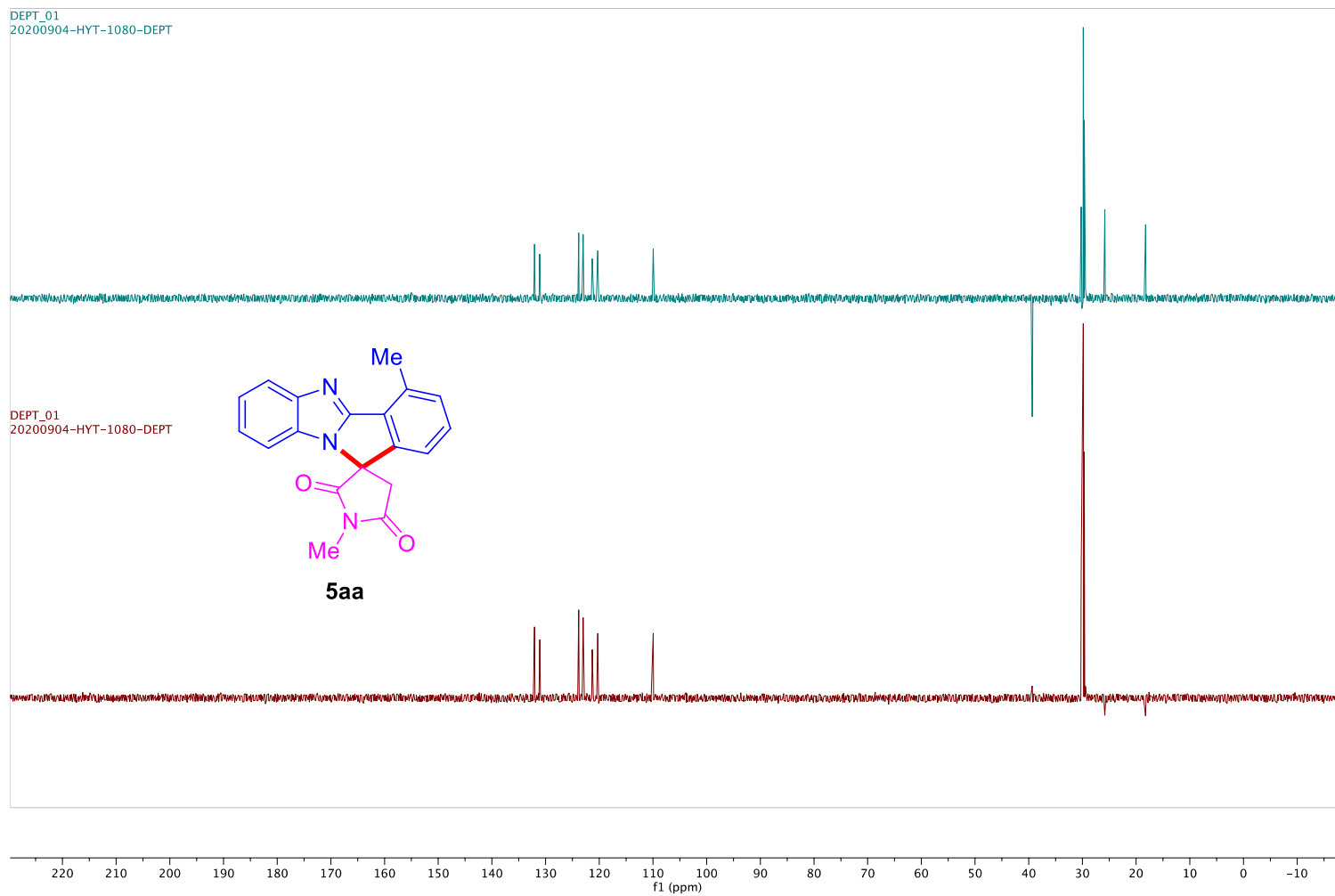
$^1\text{H}$  NMR spectrum (400 MHz) of compound **3aa** and **5aa** in  $\text{CDCl}_3$ .



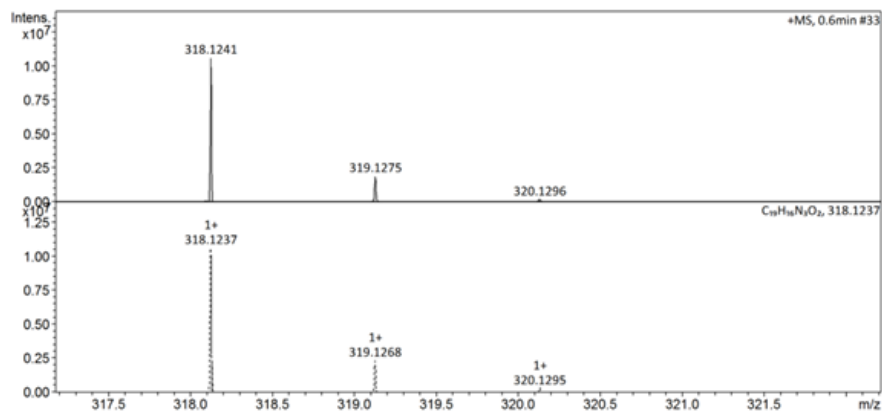
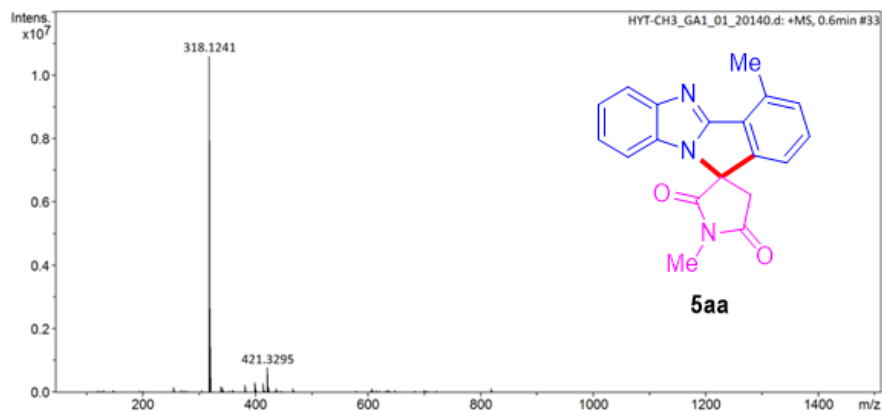
$^1\text{H}$  NMR spectrum (400 MHz) of compound **5aa** in acetone- $d_6$ .



$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5aa** in acetone- $d_6$ .



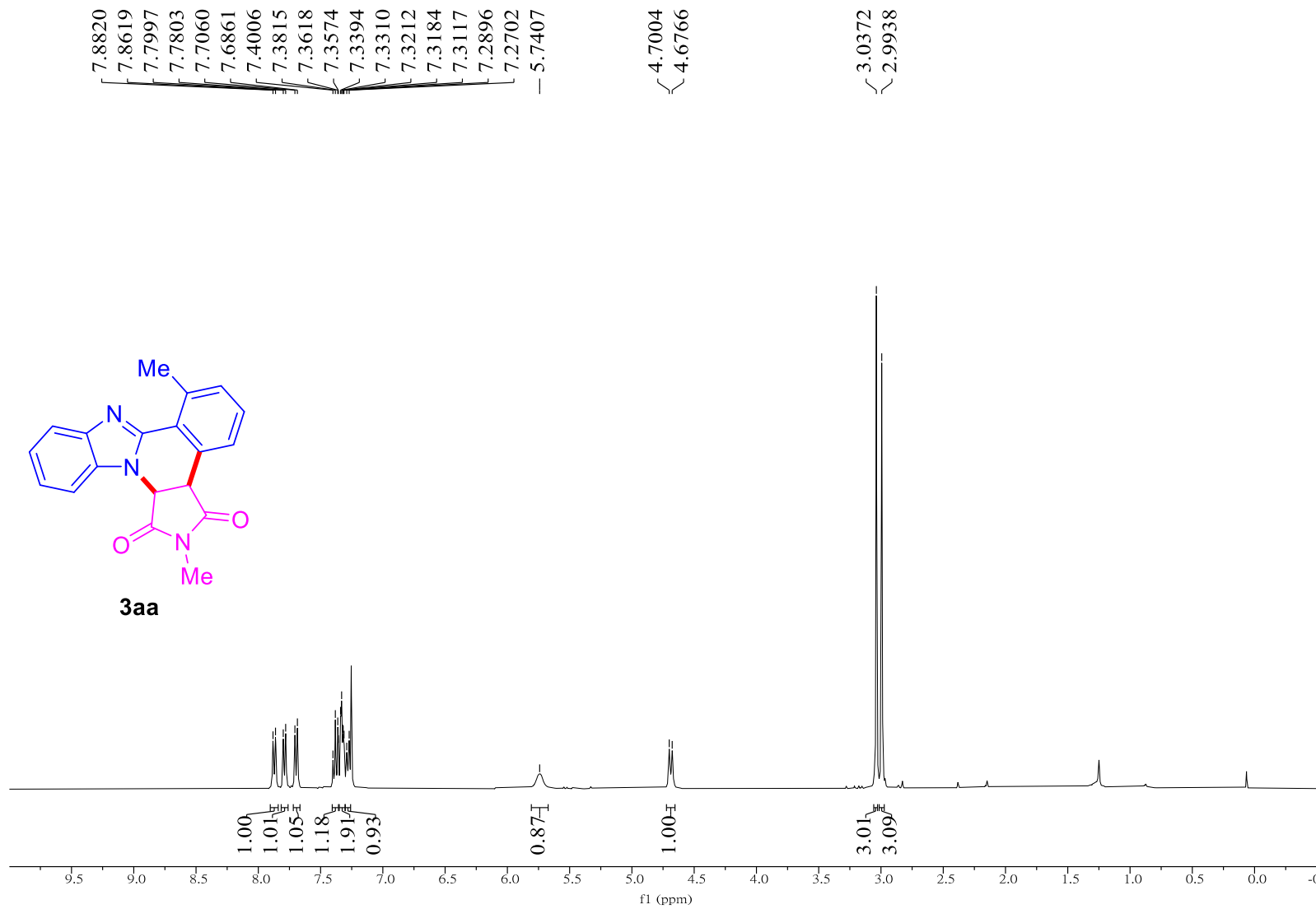
$^{13}\text{C}$ -DEPT spectrum (100 MHz) of compound **5aa** in acetone- $d_6$ .



### Display Report

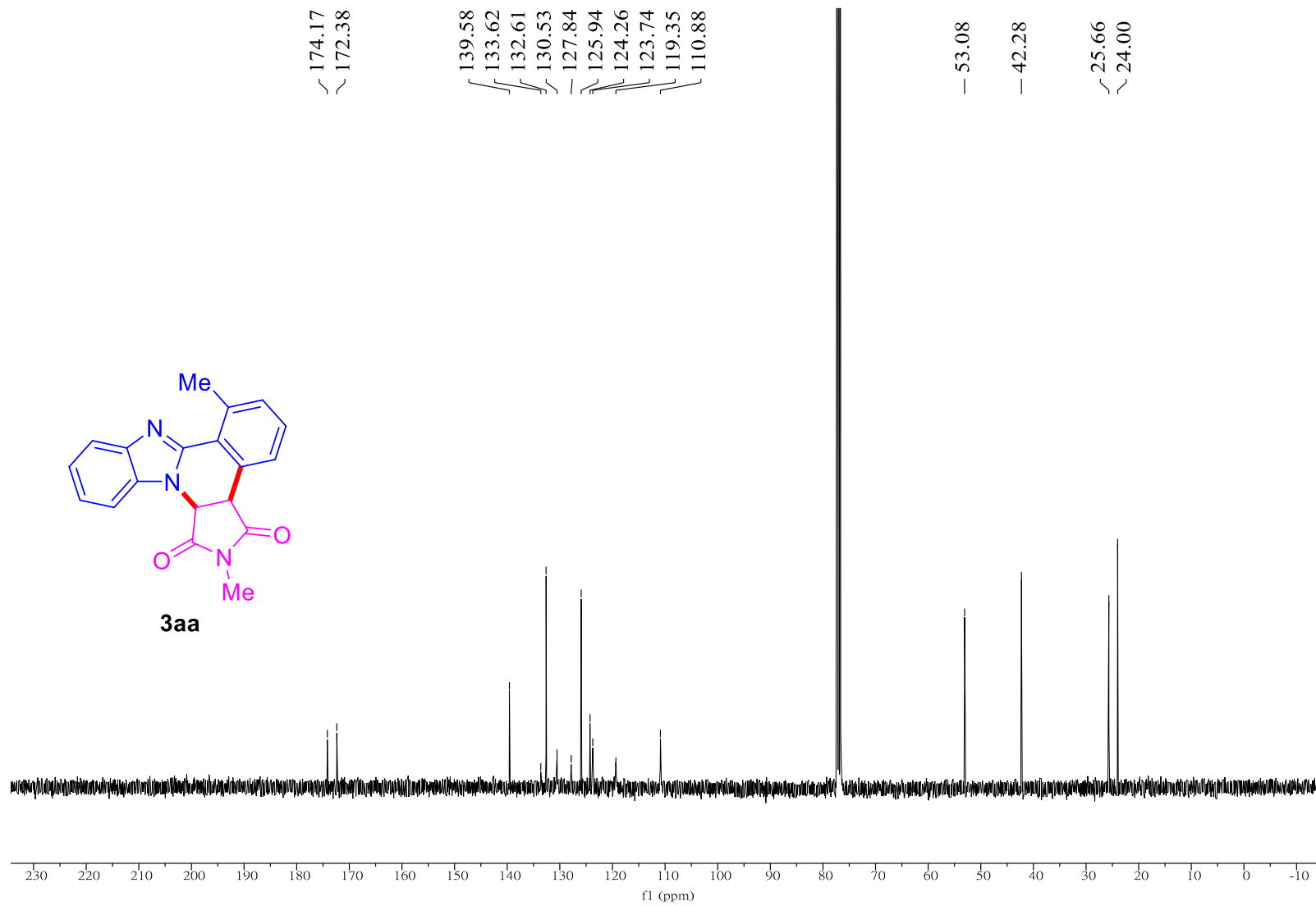
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
318.1241	1	C <sub>19</sub> H <sub>16</sub> N <sub>3</sub> O <sub>2</sub>	318.1237	1.2	23.5	1	100.00	13.5	even	ok	M

HRMS (ESI) of compound **5aa**

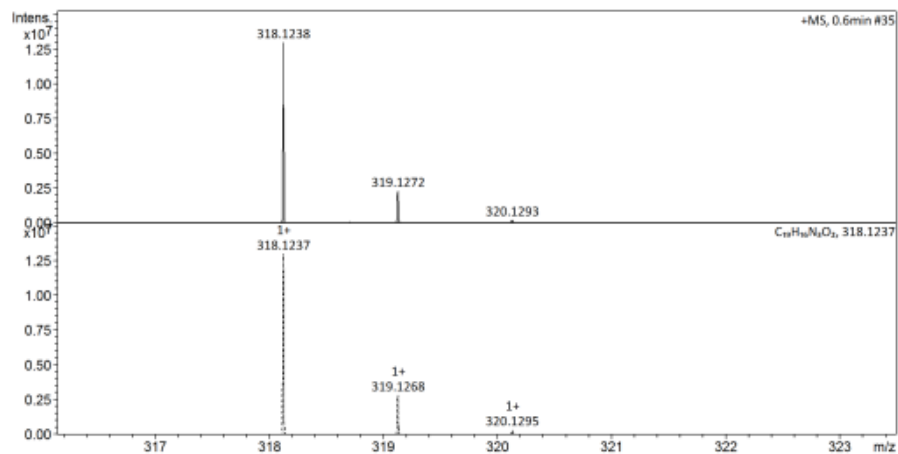
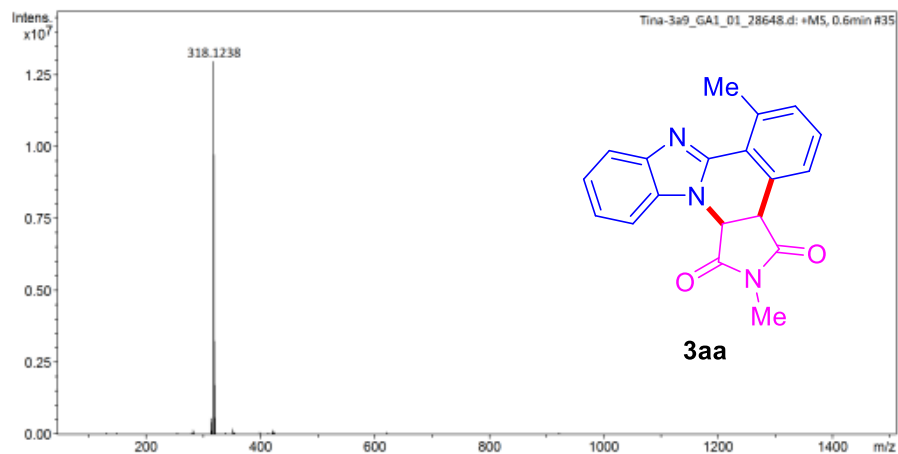


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3aa** in  $\text{CDCl}_3$ .





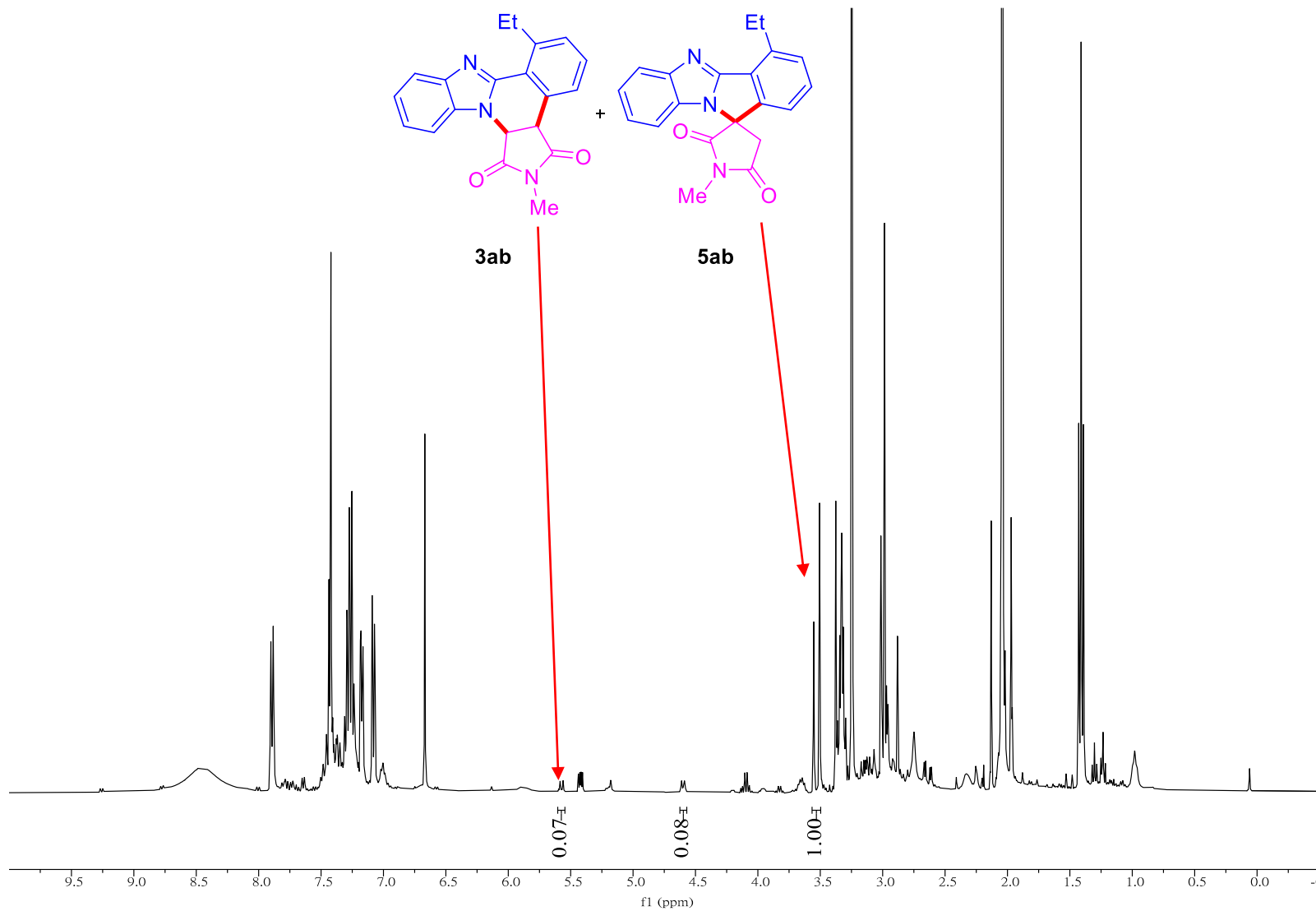
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3aa** in  $\text{CDCl}_3$ .



## Display Report

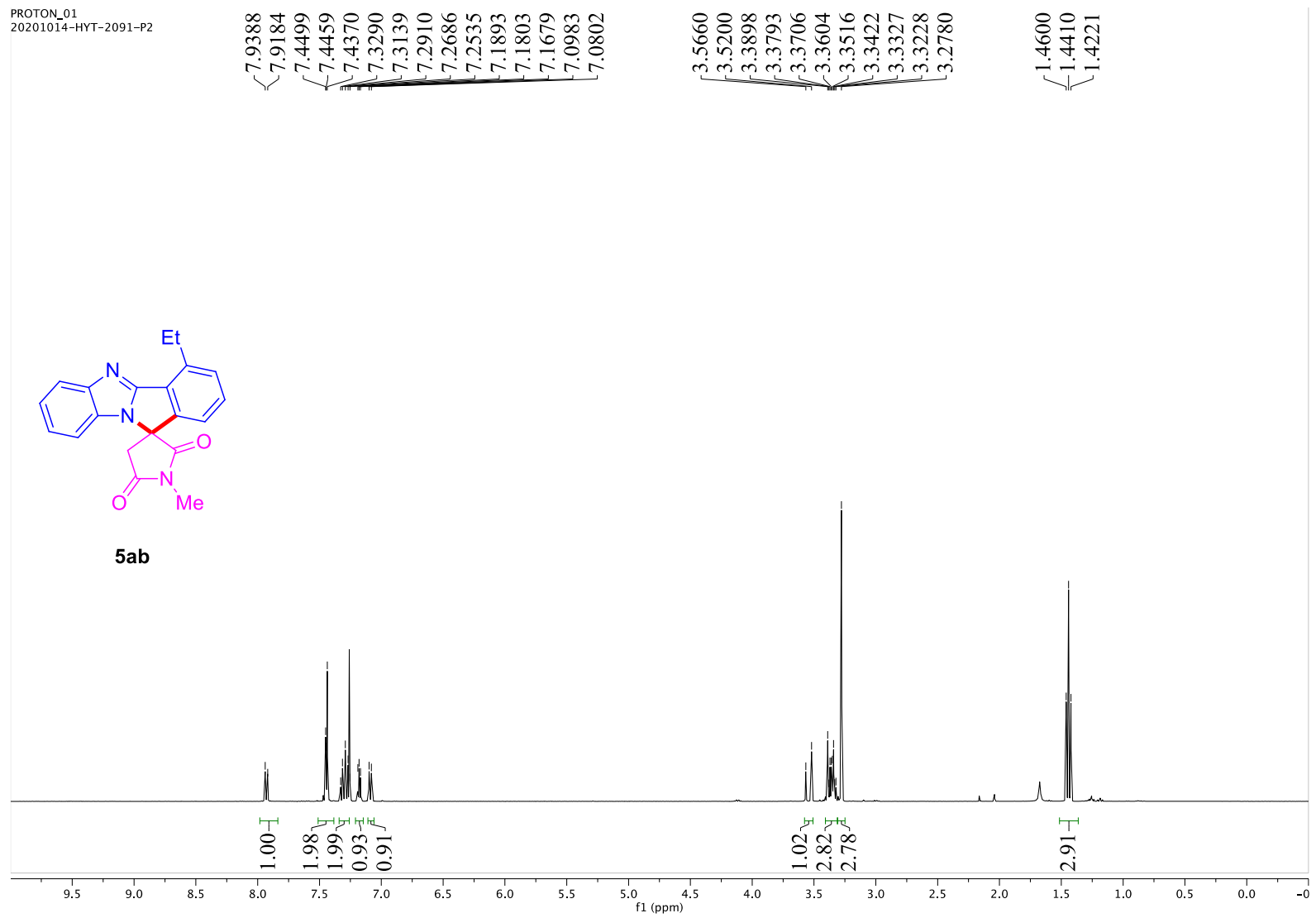
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
318.1238	1	C <sub>19</sub> H <sub>16</sub> N <sub>3</sub> O <sub>2</sub>	318.1237	-0.4	24.9	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **3aa**.



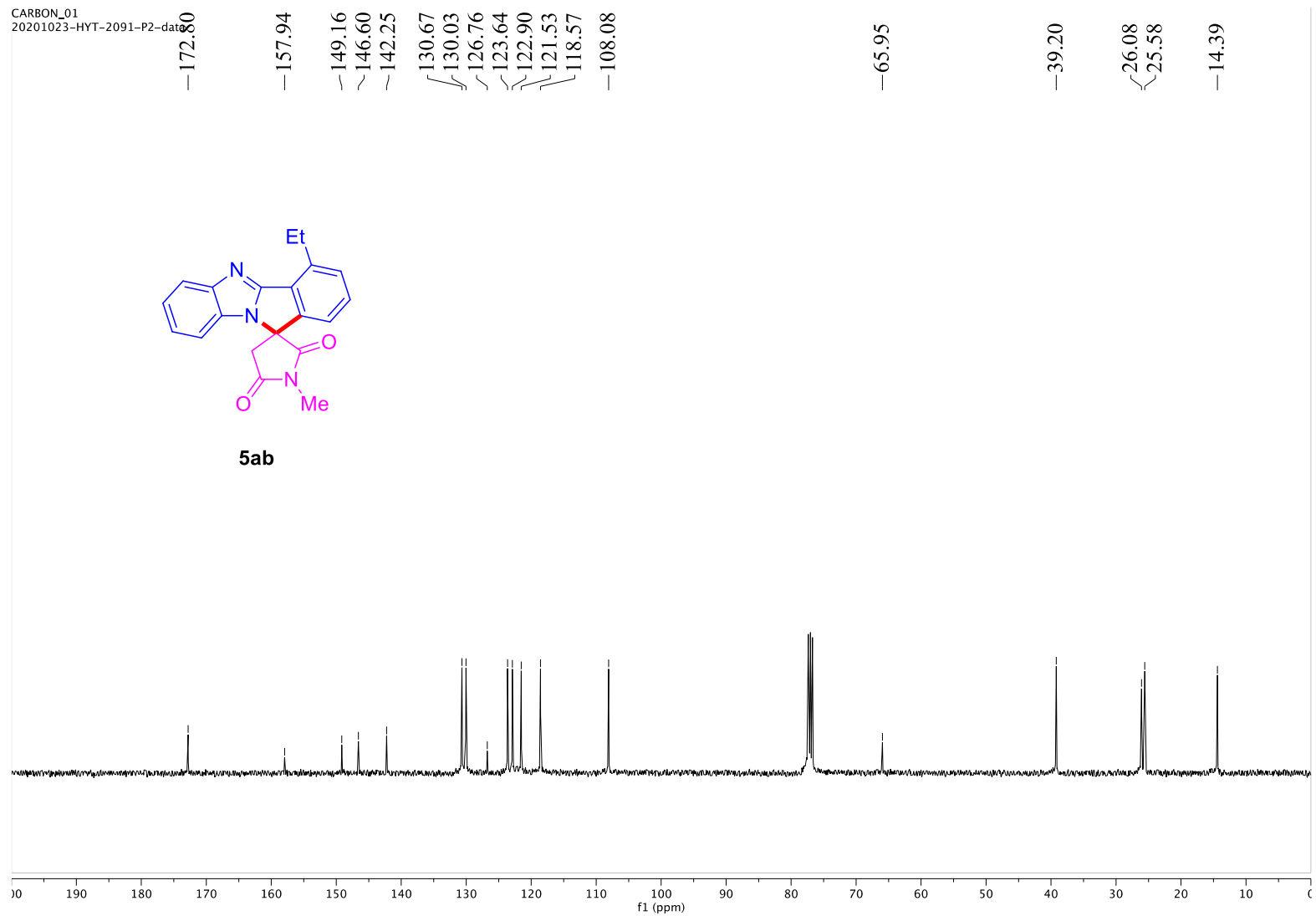
$^1\text{H}$  NMR spectrum (400 MHz) of compound **3ab** and **5ab** in  $\text{CDCl}_3$ .

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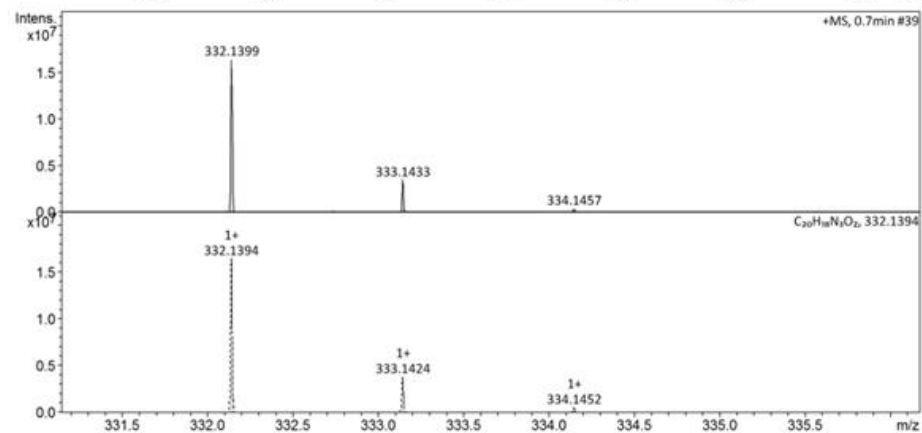
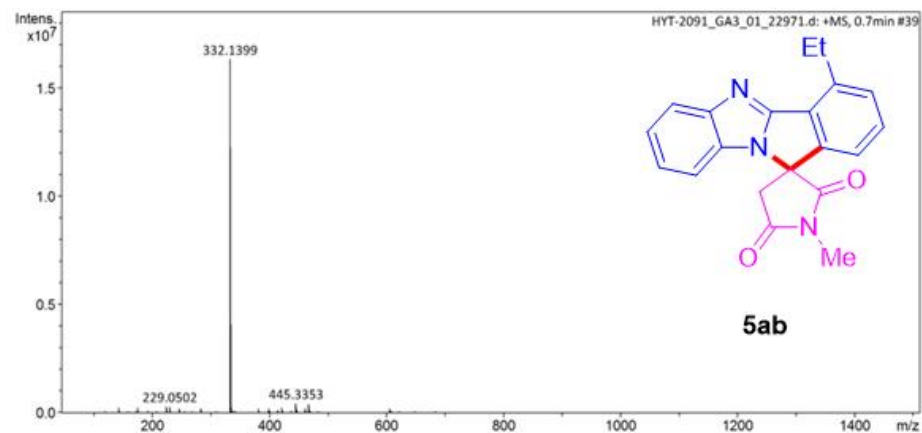


<sup>1</sup>H NMR spectrum (400 MHz) of compound **5ab** in CDCl<sub>3</sub>.

CARBON\_01  
20201023-HYT-2091-P2-data



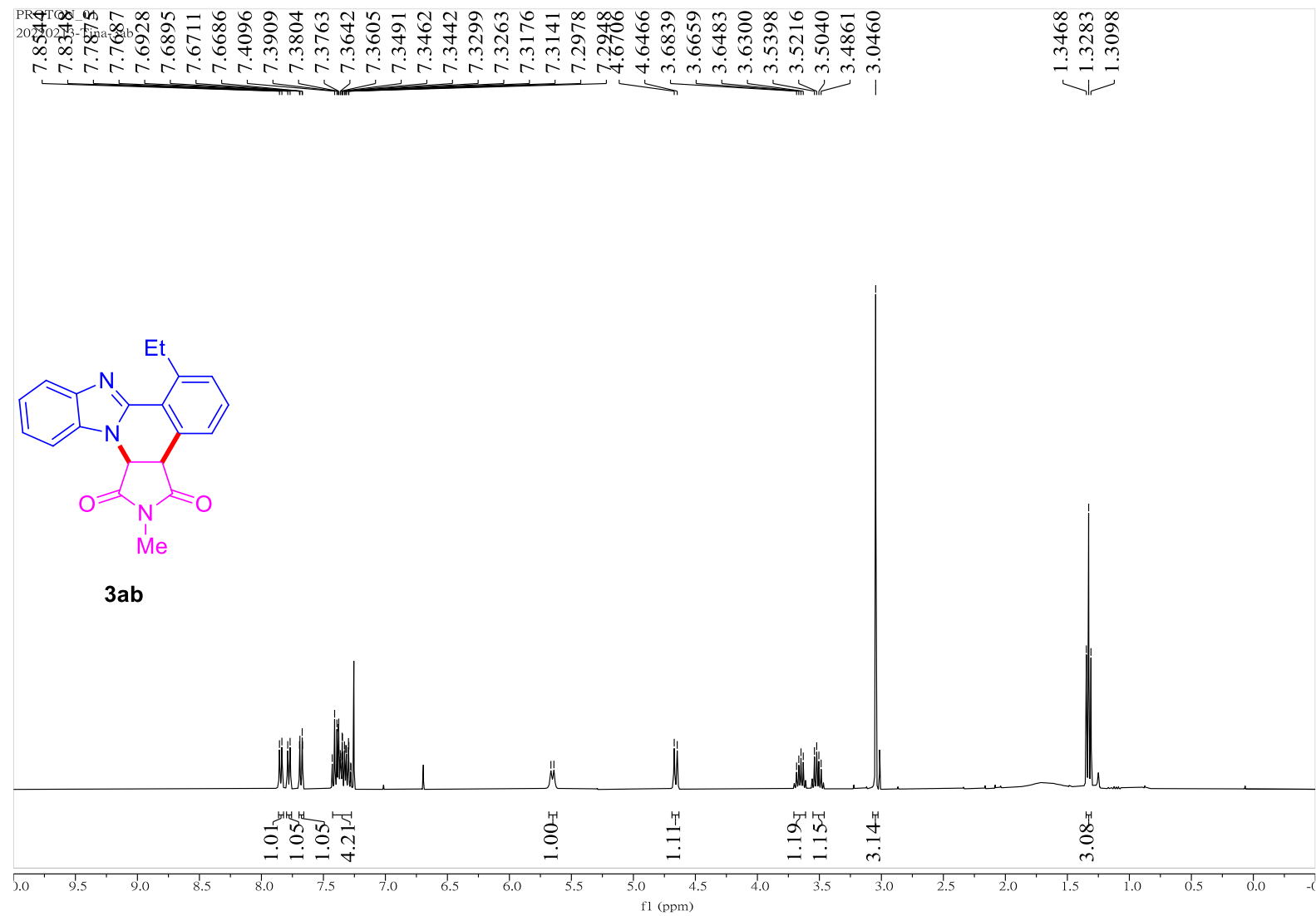
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5ab** in  $\text{CDCl}_3$ .



### Display Report

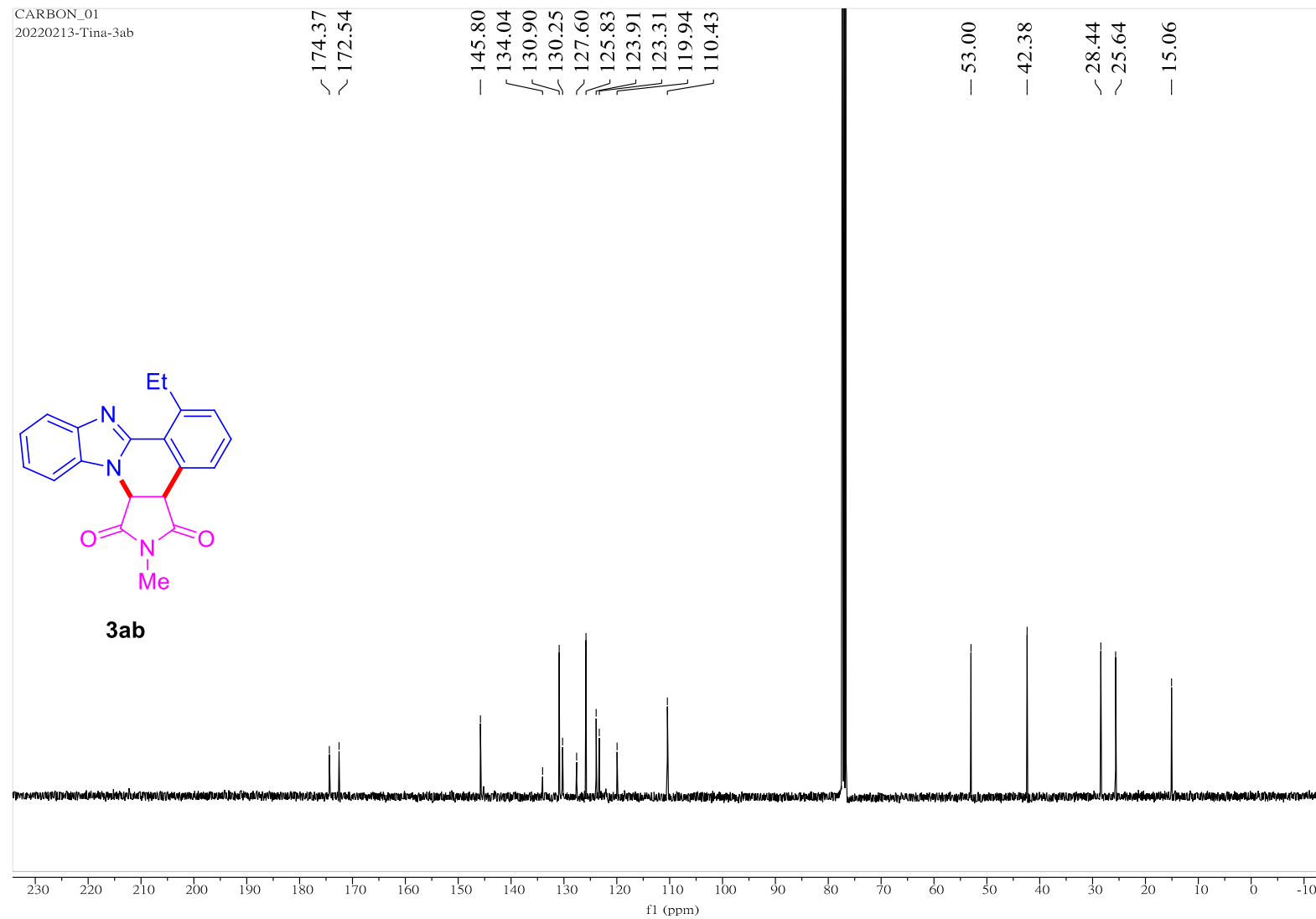
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
332.1399	1	C <sub>20</sub> H <sub>18</sub> N <sub>2</sub> O <sub>2</sub>	332.1394	1.7	13.8	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **5ab**



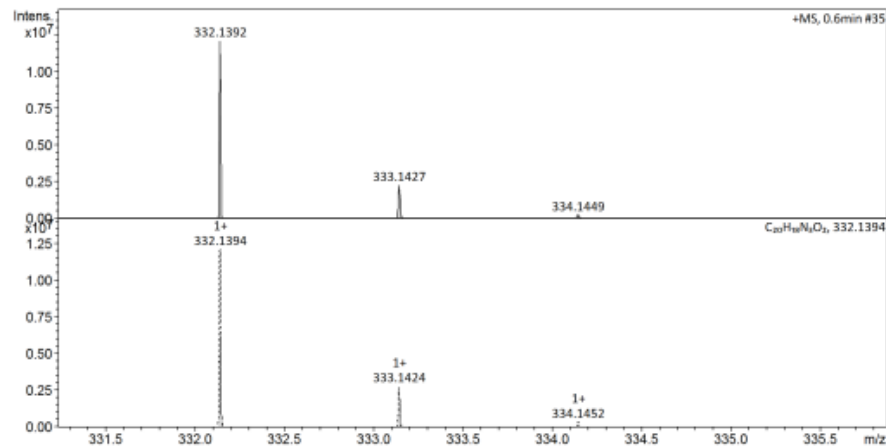
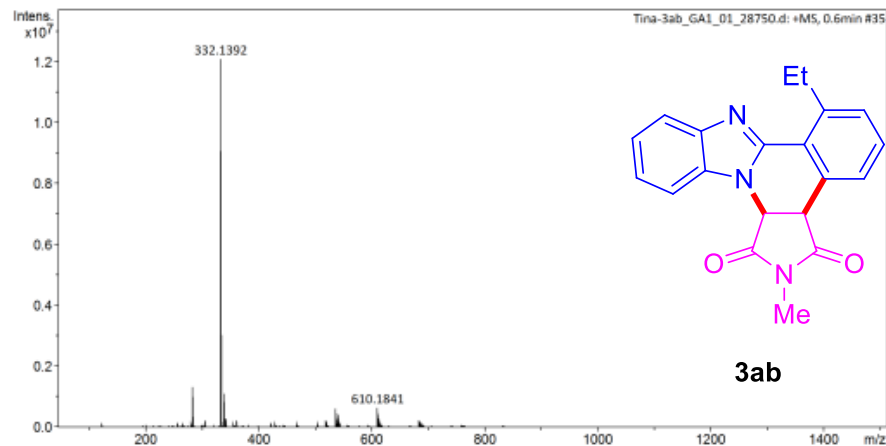
$^1\text{H}$  NMR spectrum (400 MHz) of compound **3ab** in  $\text{CDCl}_3$ .

CARBON\_01  
20220213-Tina-3ab



<sup>13</sup>C NMR spectrum (100 MHz) of compound **3ab** in CDCl<sub>3</sub>.

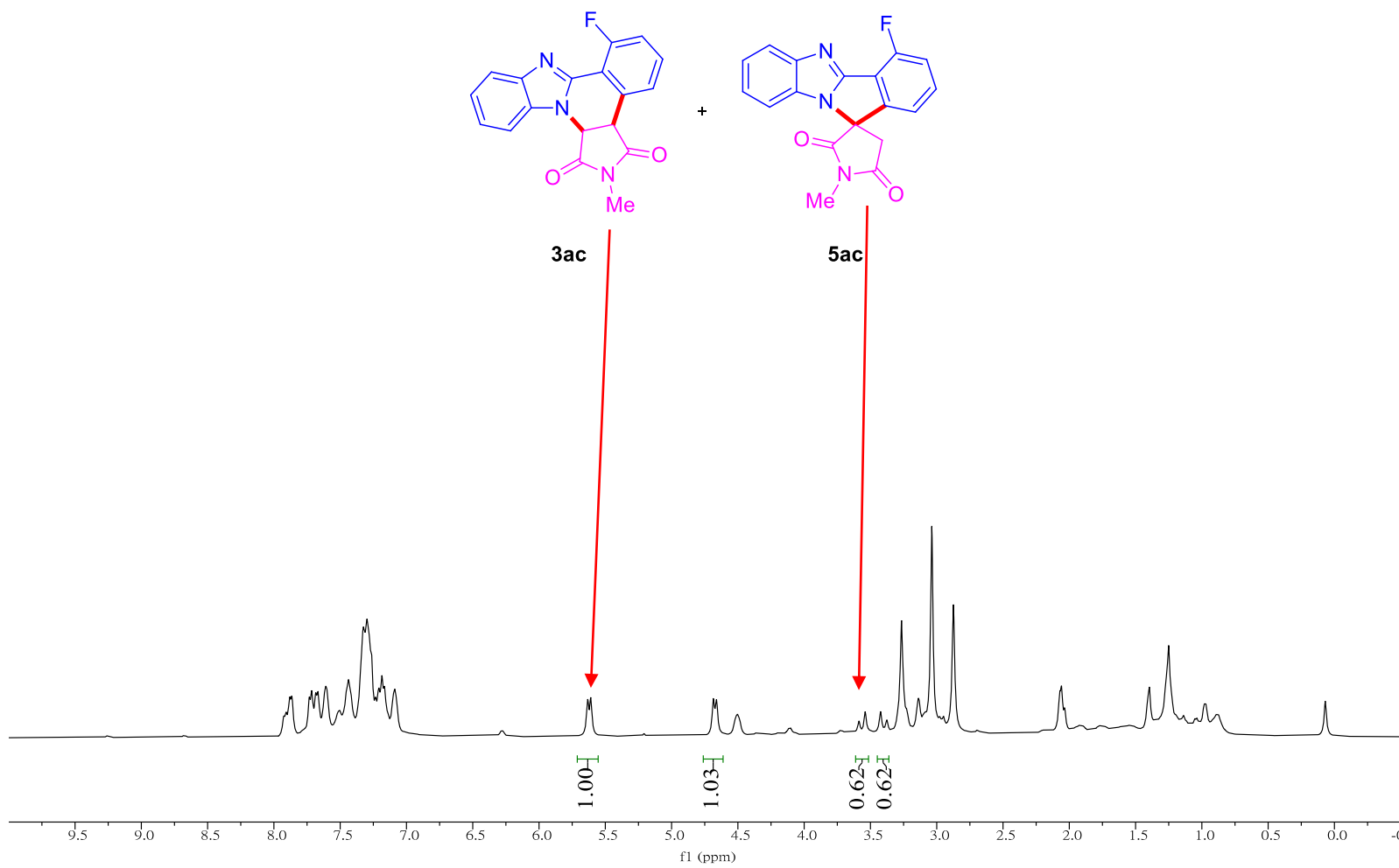




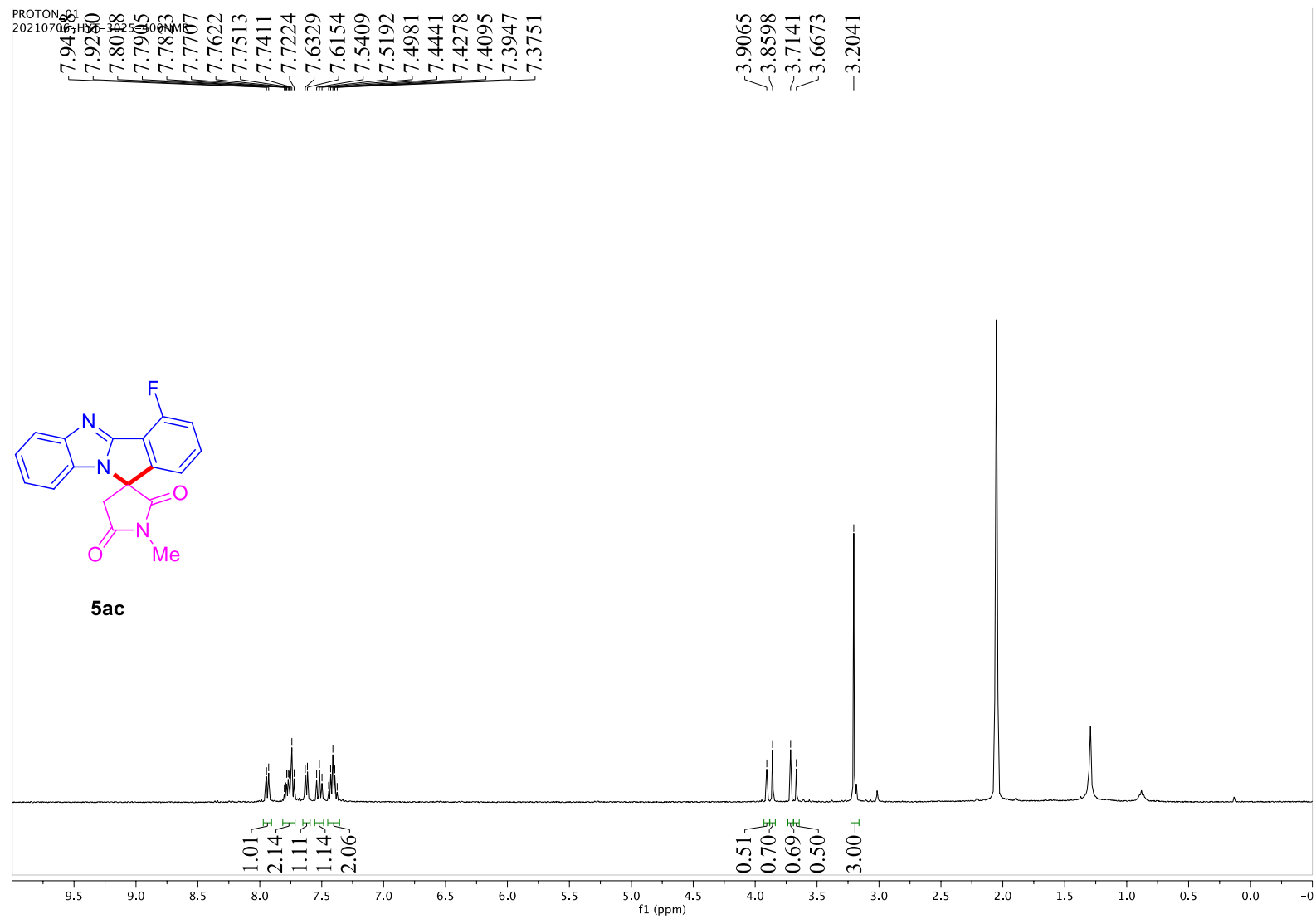
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
332.1392	1	C <sub>20</sub> H <sub>18</sub> N <sub>3</sub> O <sub>2</sub>	332.1394	-0.4	25.4	1	100.00	13.5	even	ok	M+H

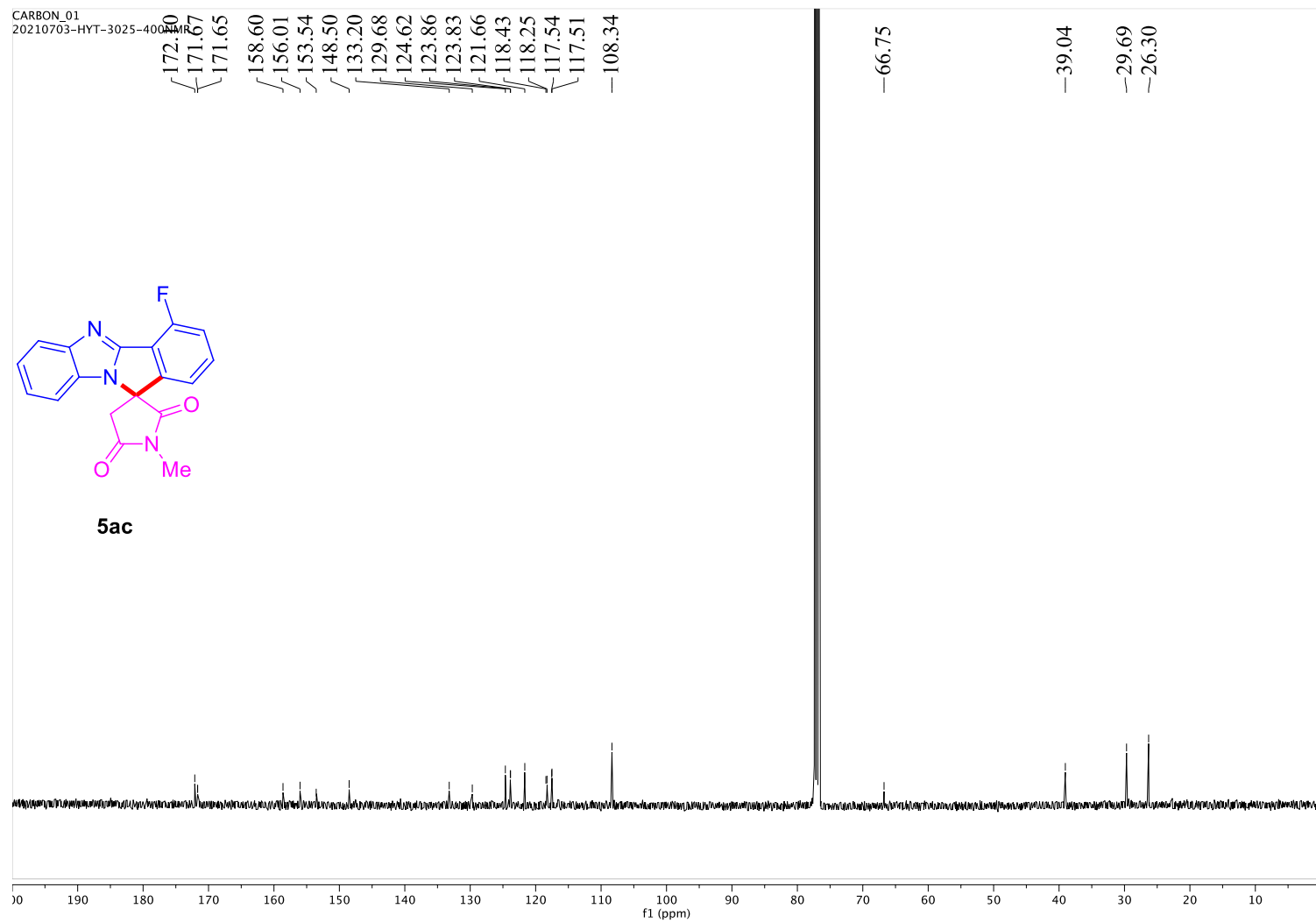
HRMS (ESI) of compound **3ab**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3ac** and **5ac** in  $\text{CDCl}_3$ .



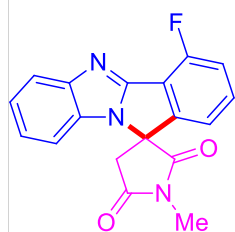
$^1\text{H}$  NMR spectrum (400 MHz) of compound **5ac** in acetone- $d_6$ .



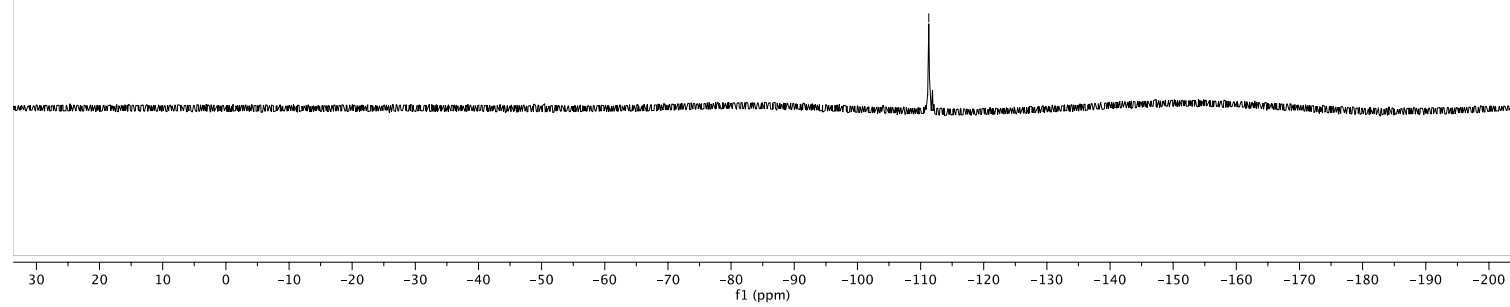
<sup>13</sup>C NMR spectrum (100 MHz) of compound **5ac** in CDCl<sub>3</sub>.

FLUORINE\_01  
20210703-HYT-3025-400NMR

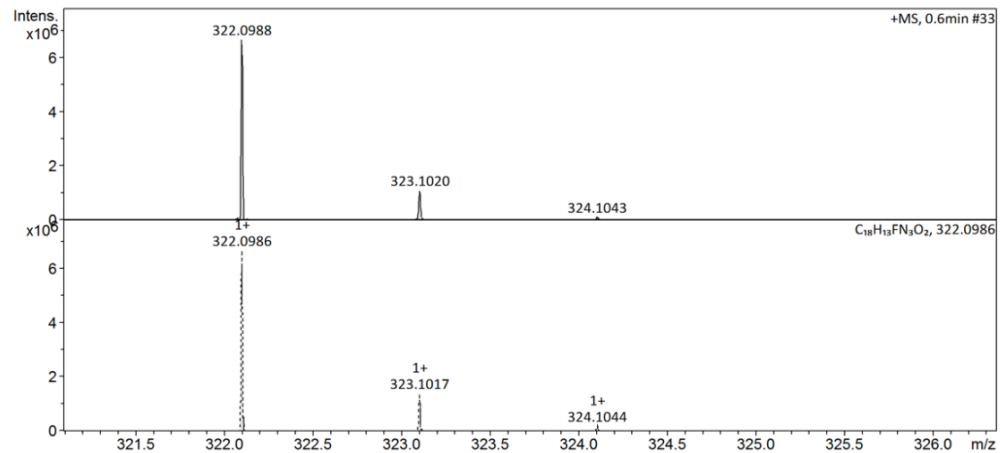
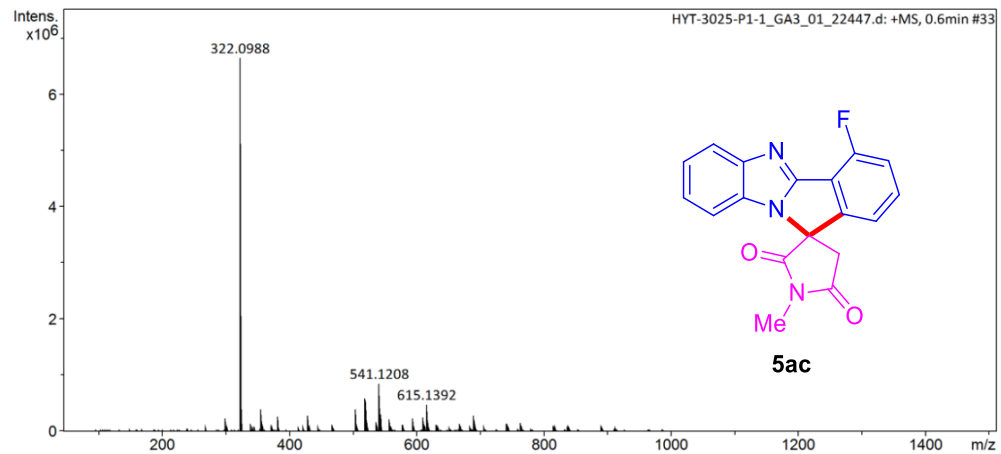
--111.30



**5ac**



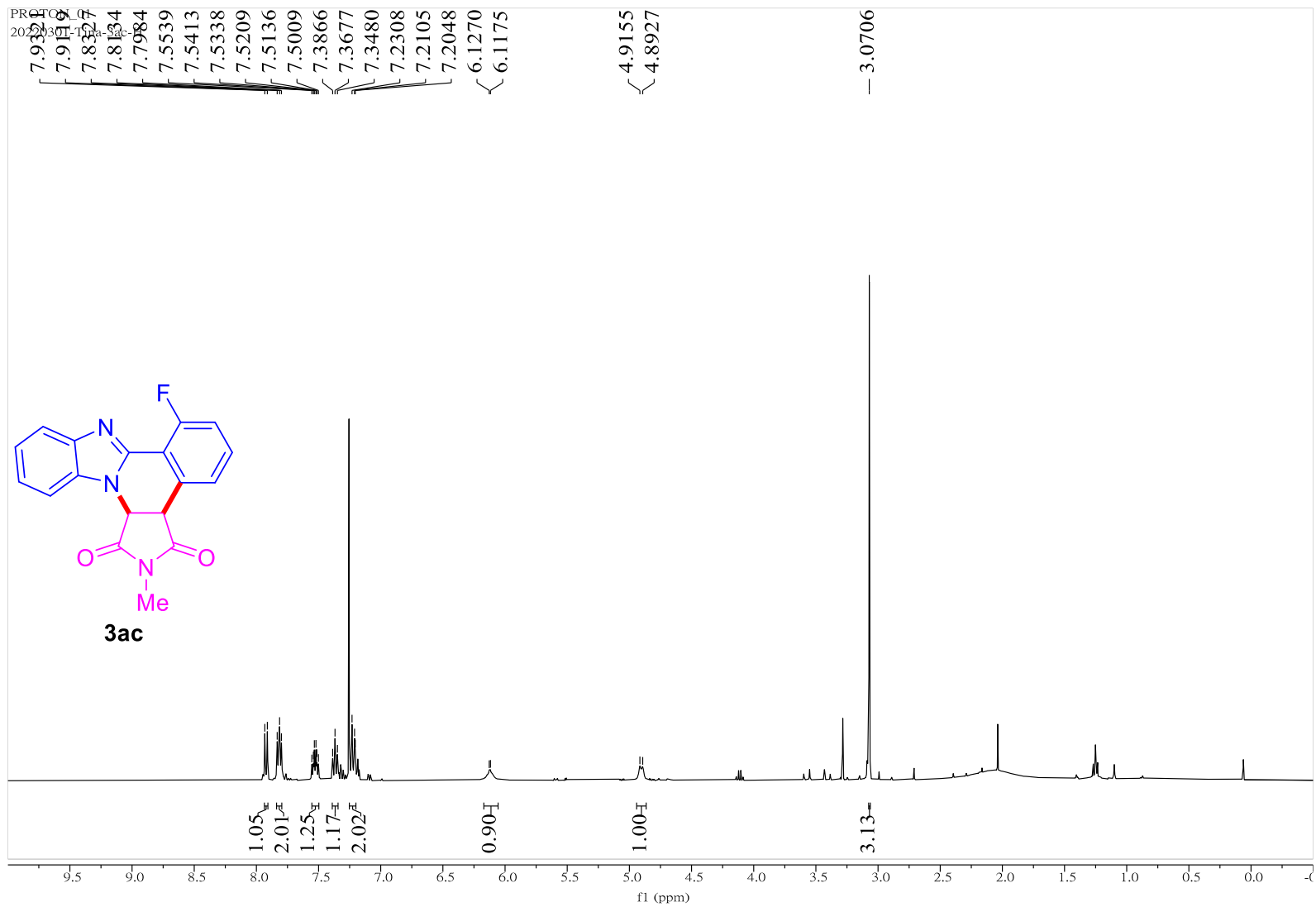
$^{19}\text{F}$  NMR spectrum (376 MHz) of compound **5ac** in  $\text{CDCl}_3$ .



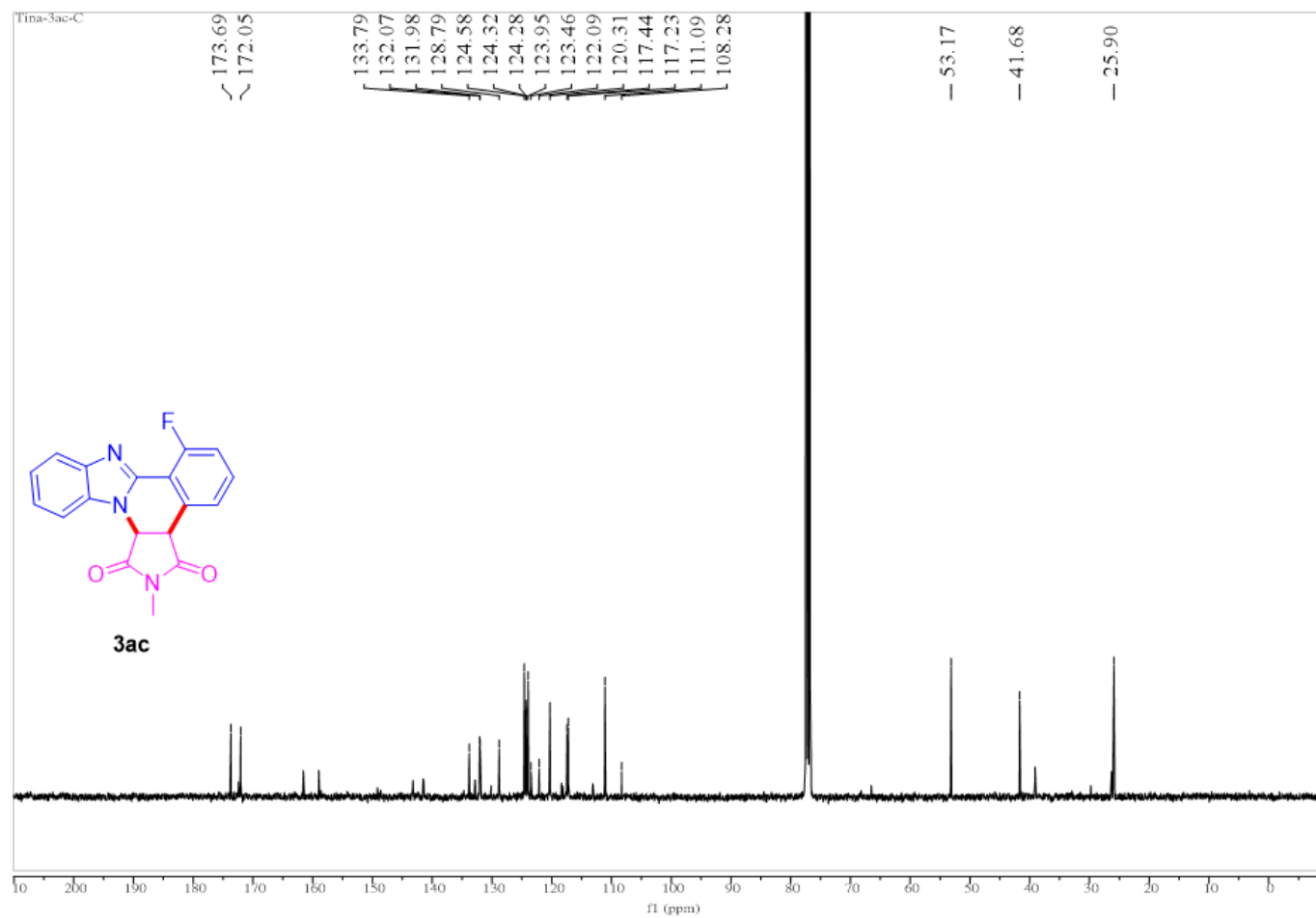
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
322.0988	1	C <sub>18</sub> H <sub>13</sub> FN <sub>3</sub> O <sub>2</sub>	322.0986	0.5	25.4	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **5ac**.

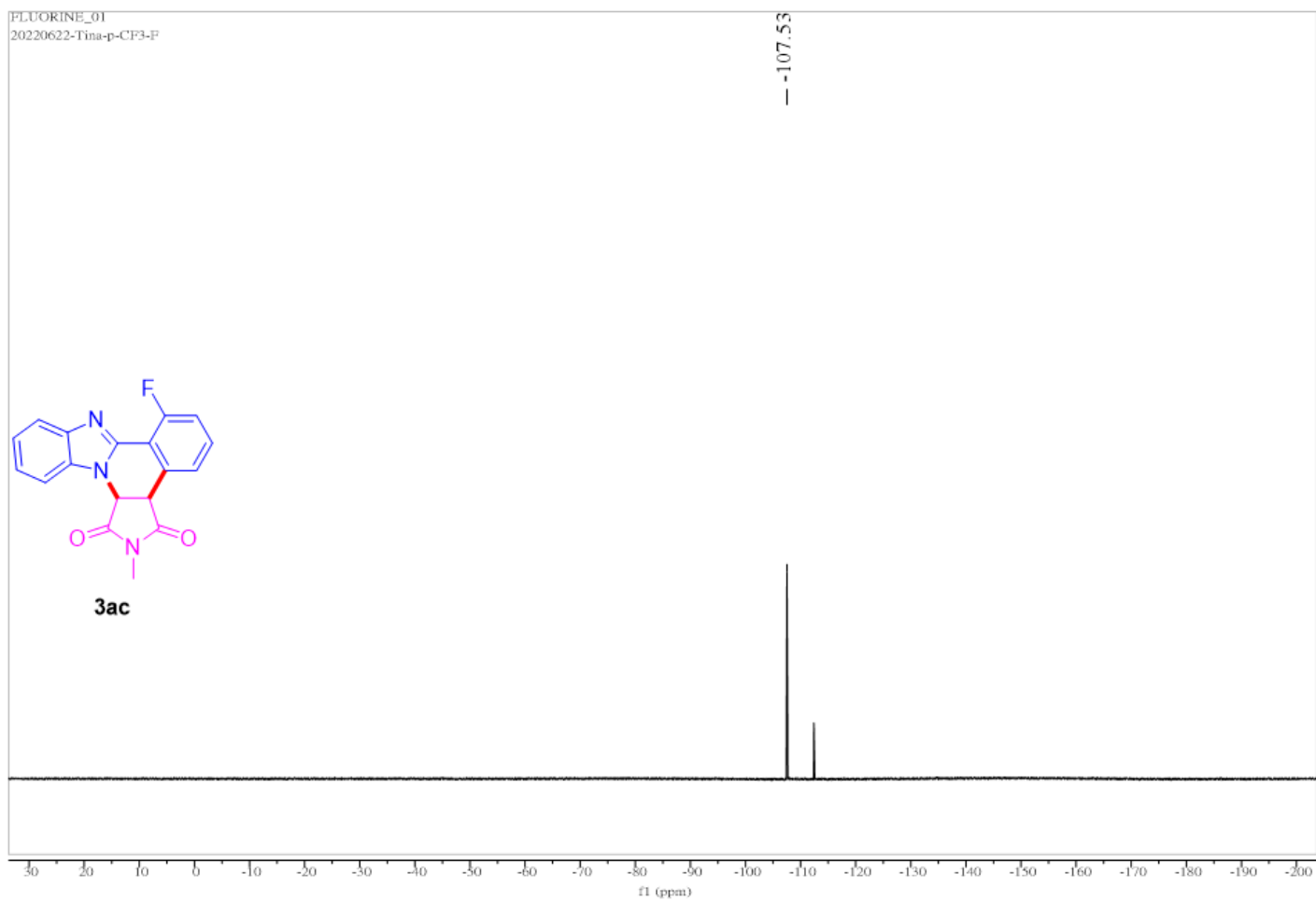


<sup>1</sup>H NMR spectrum (400 MHz) of compound **3ac** in CDCl<sub>3</sub>.

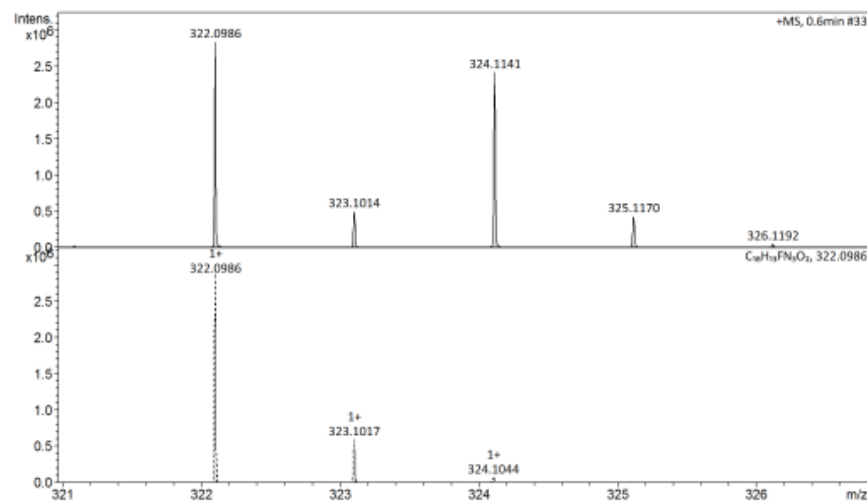
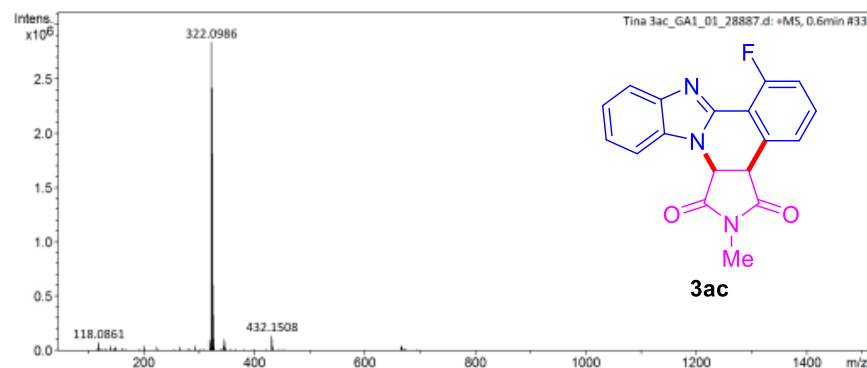


$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3ac** in  $\text{CDCl}_3$ .





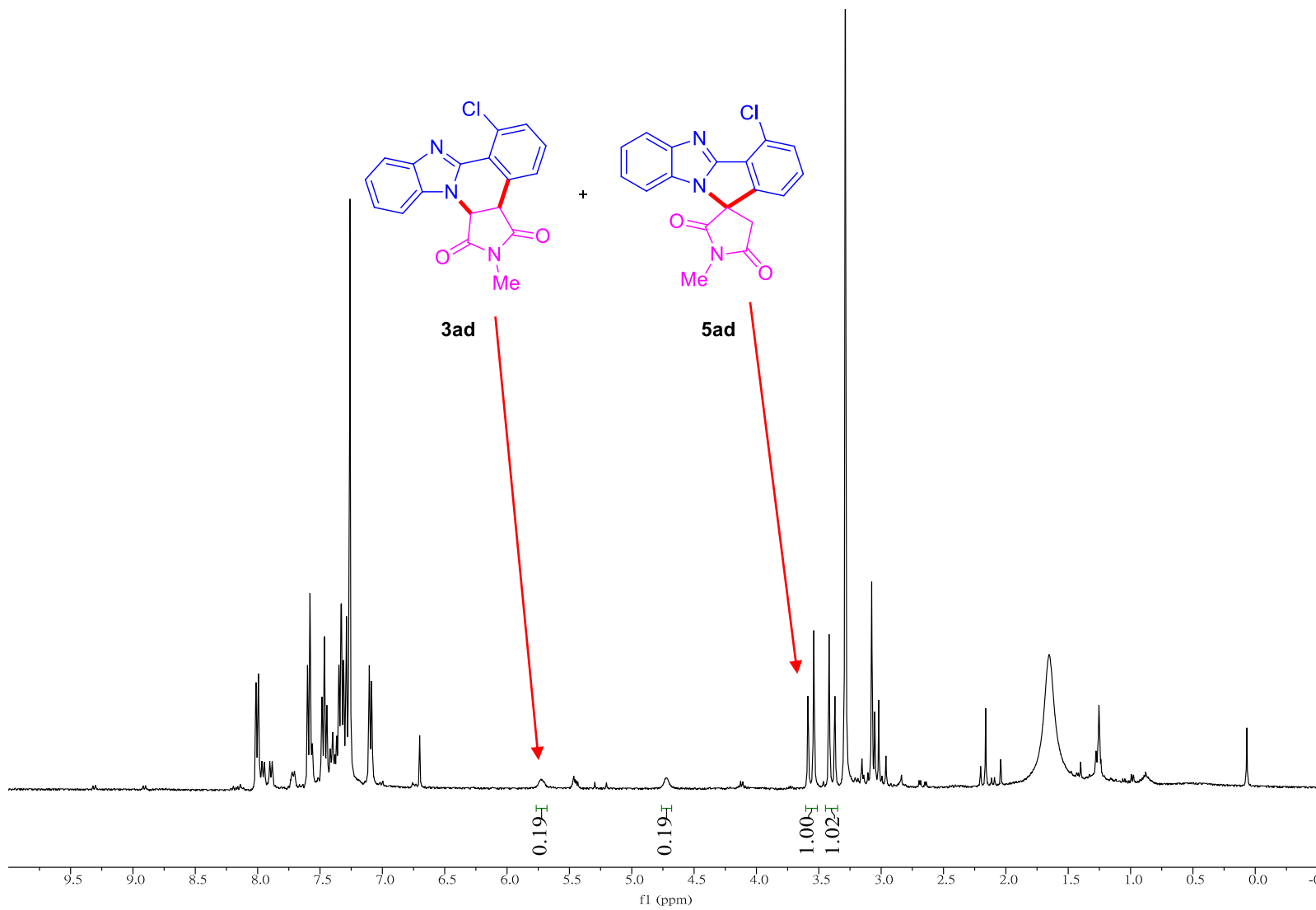
$^{19}\text{F}$  NMR spectrum (376 MHz) of compound **3ac** in  $\text{CDCl}_3$



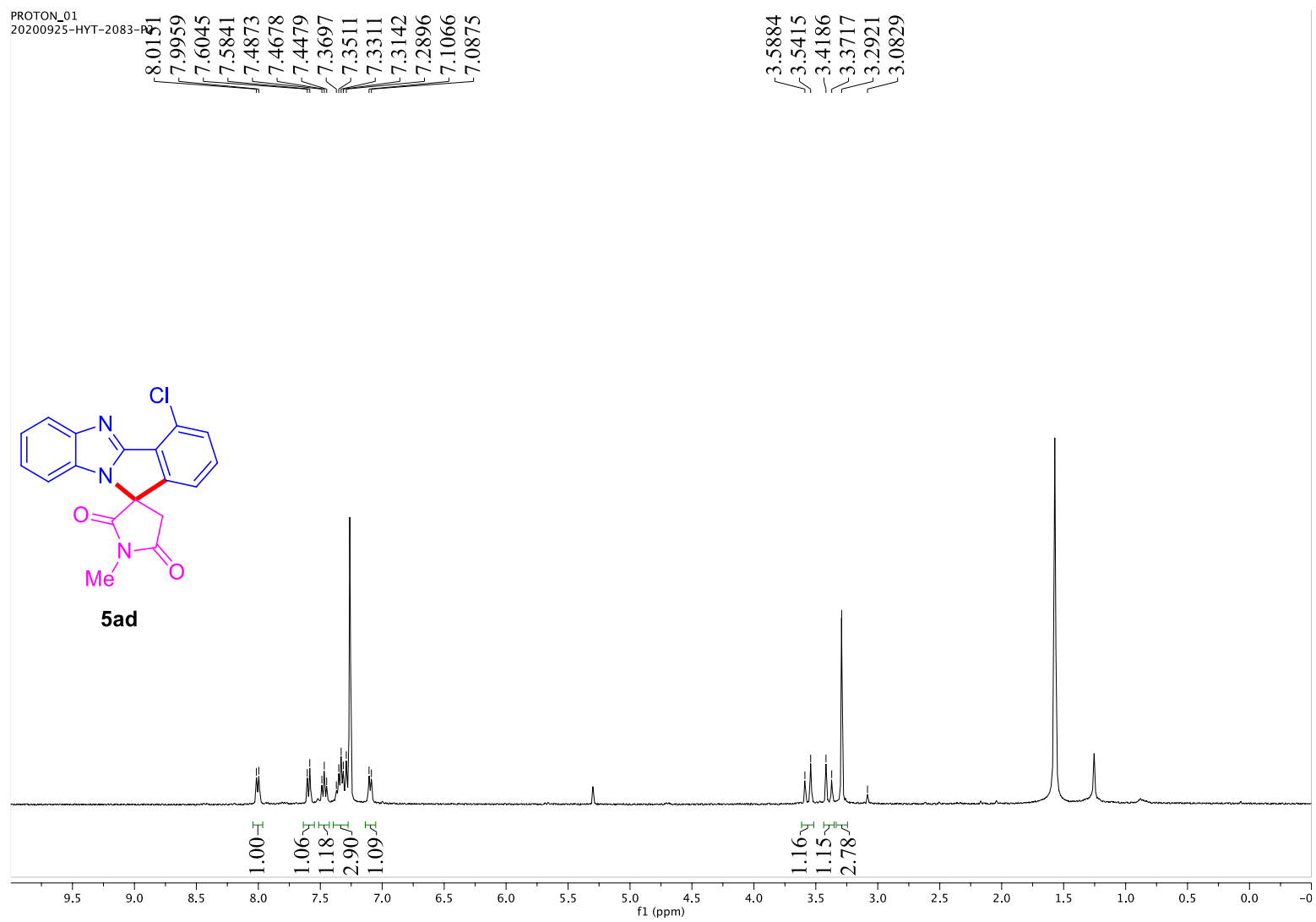
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
322.0986	1	C <sub>18</sub> H <sub>13</sub> FN <sub>3</sub> O <sub>2</sub>	322.0986	-0.1	23.3	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **3ac**.

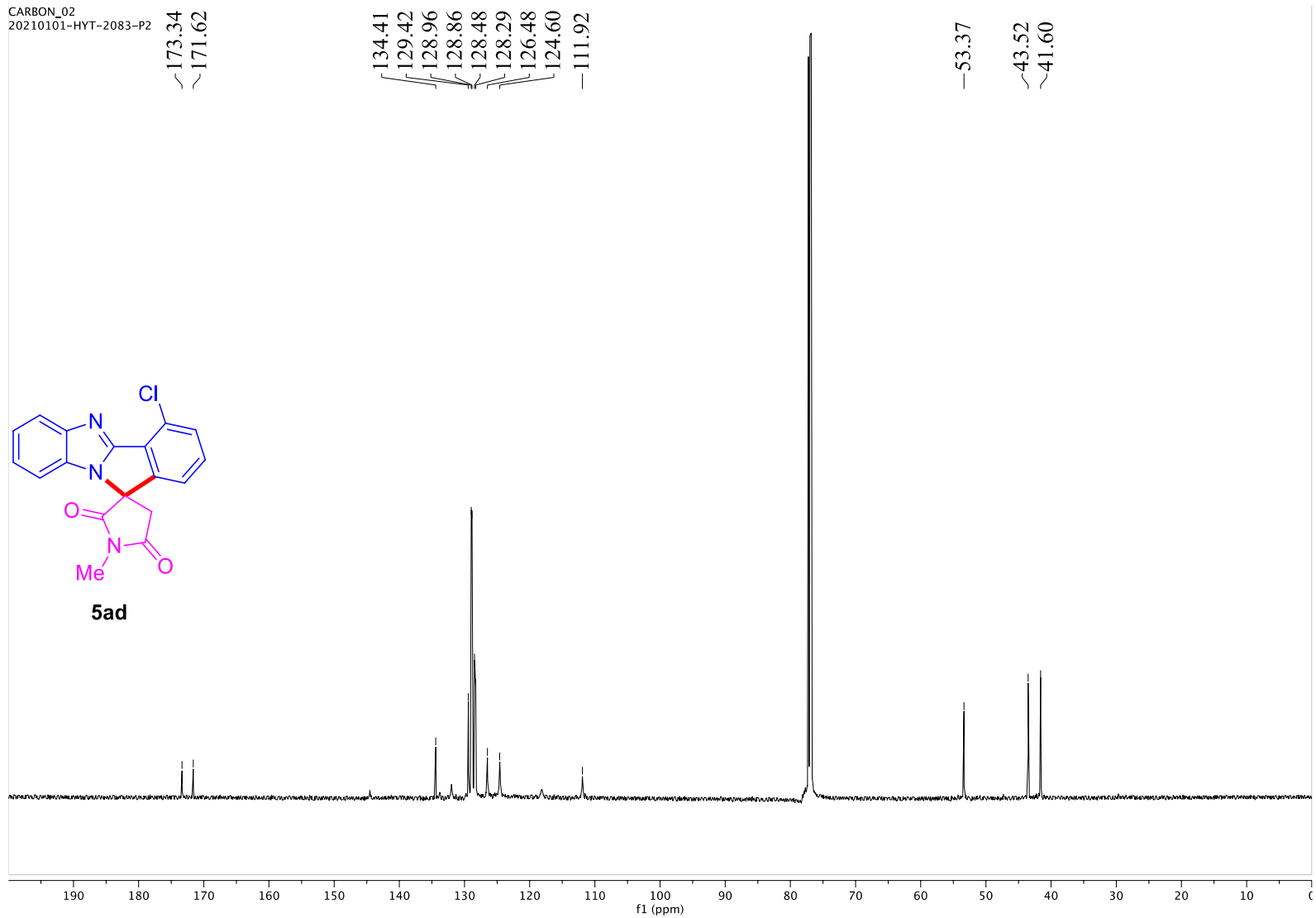


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3ad** and **5ad** (ratio) in  $\text{CDCl}_3$ .

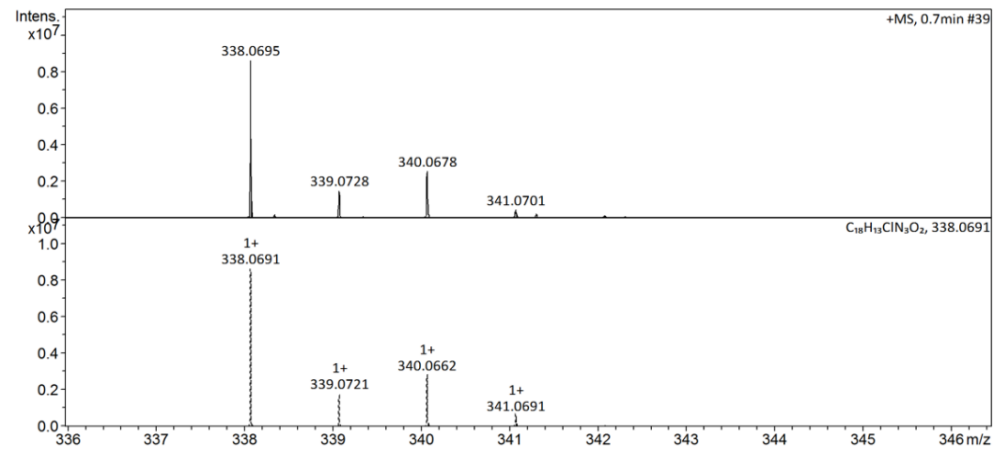
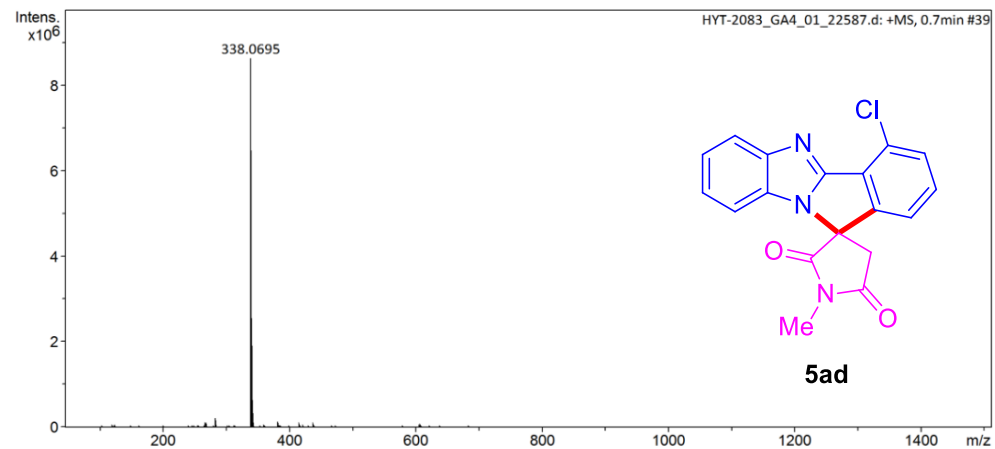


$^1\text{H}$  NMR spectrum (400 MHz) of compound **5ad** in  $\text{CDCl}_3$ .

CARBON\_02  
20210101-HYT-2083-P2



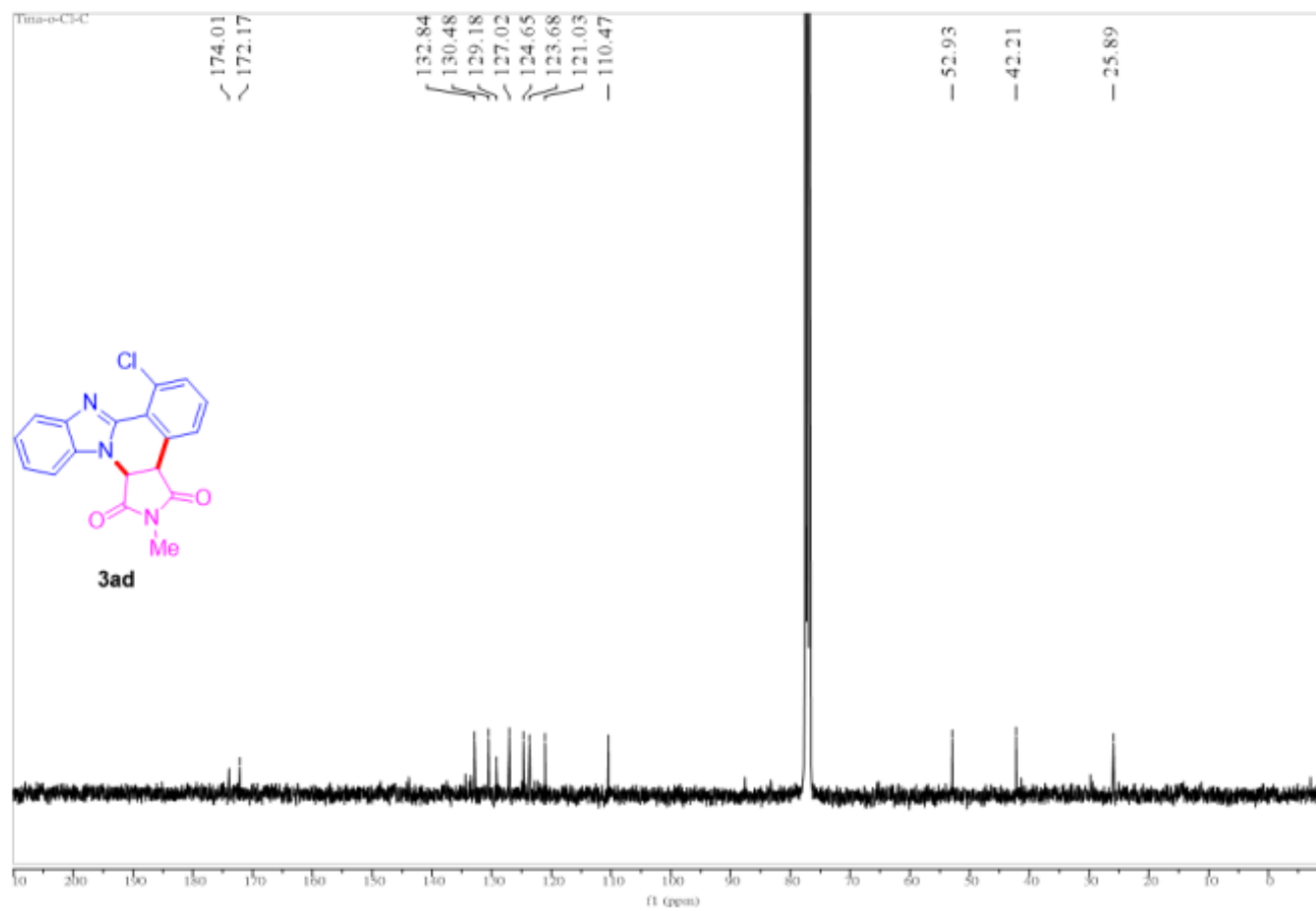
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5ad** in  $\text{CDCl}_3$ .



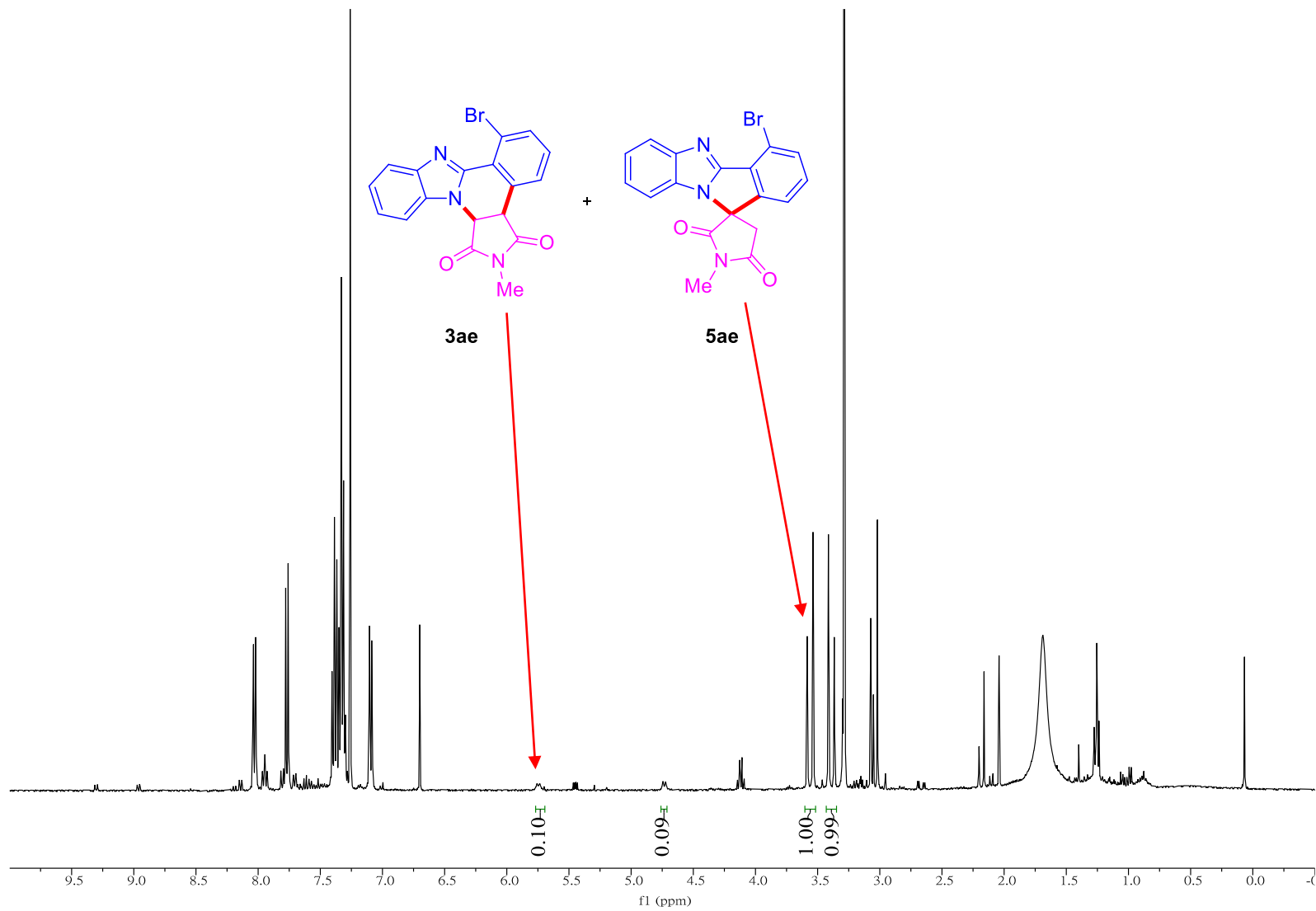
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
338.0695	1	C <sub>18</sub> H <sub>13</sub> ClN <sub>3</sub> O <sub>2</sub>	338.0691	1.4	27.8	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **5ad**.

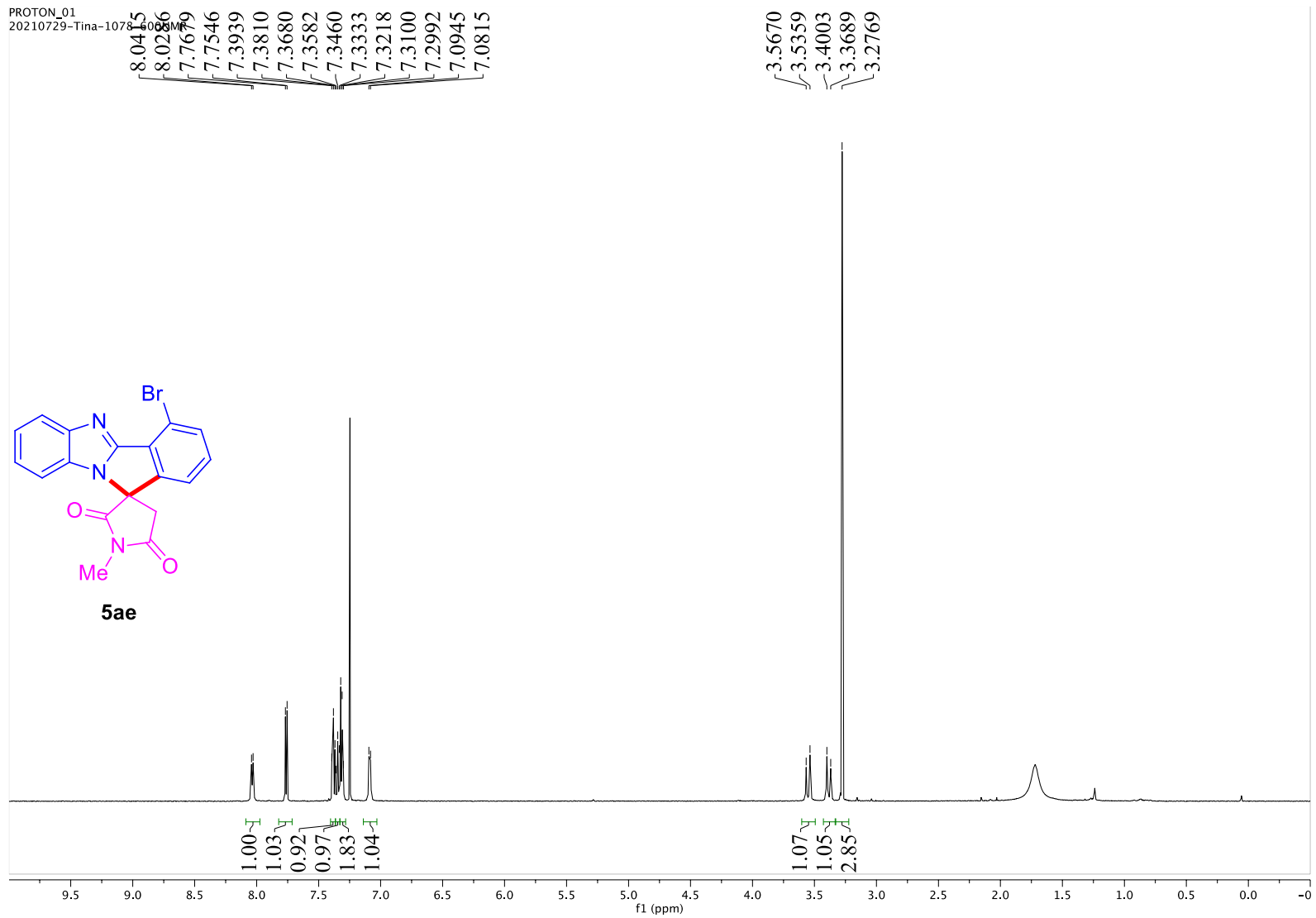


<sup>13</sup>C NMR spectrum (100 MHz) of compound **3ad** in CDCl<sub>3</sub>.

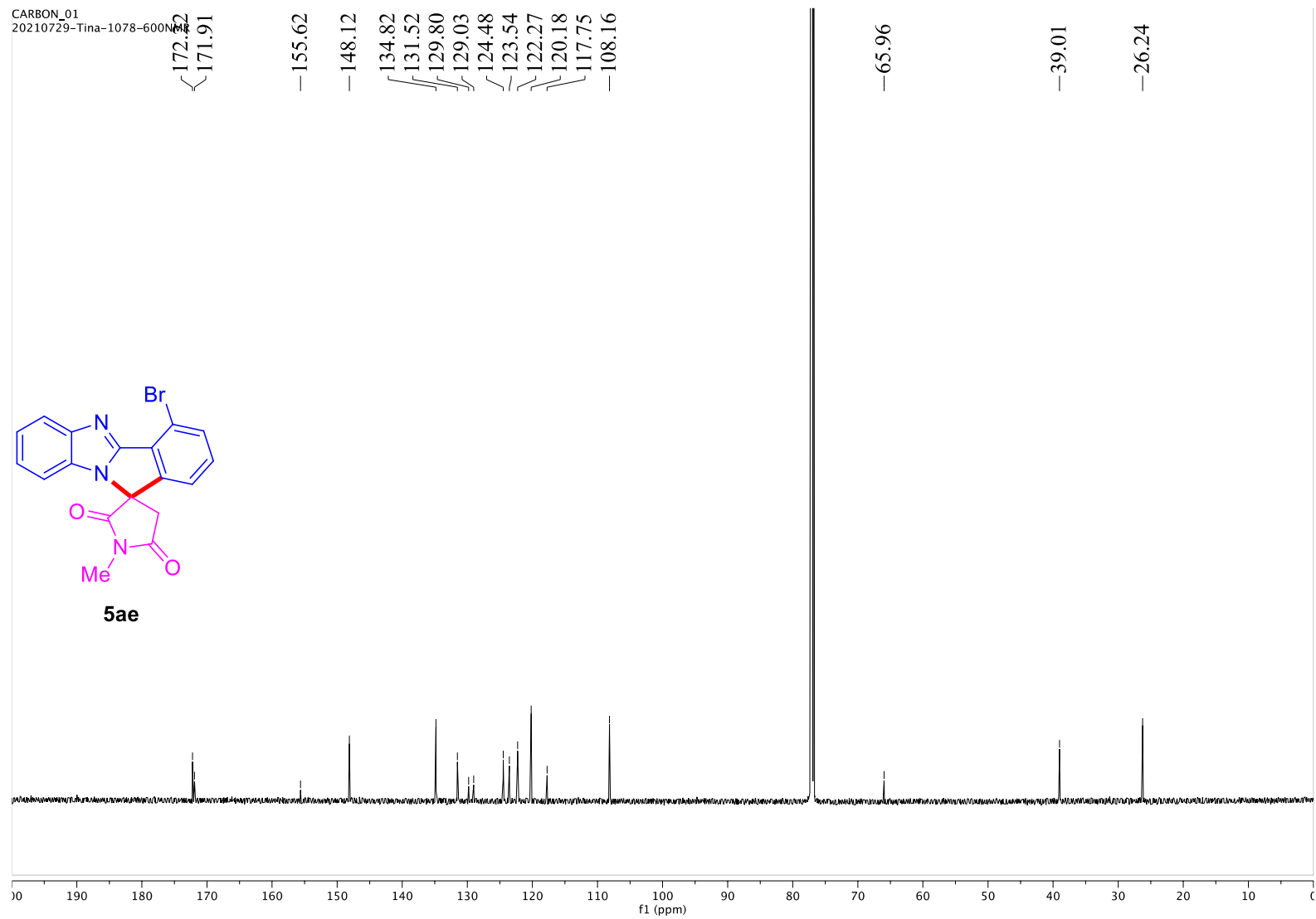


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3ae** and **5ae** (ratio) in  $\text{CDCl}_3$ .

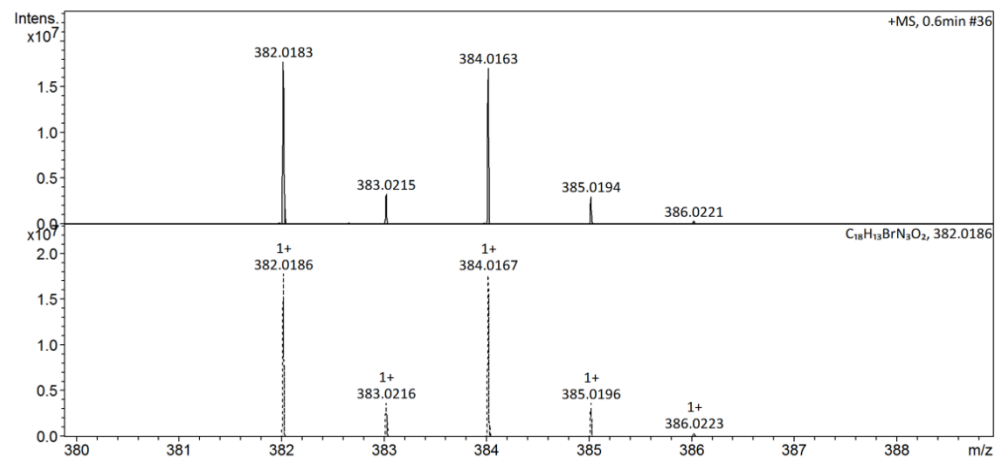
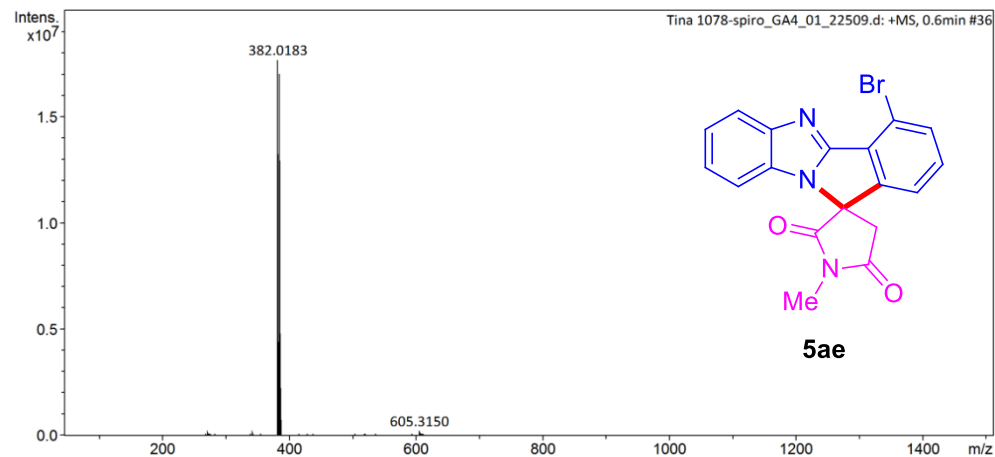




$^1\text{H}$  NMR spectrum (600 MHz) of compound **5ae** in  $\text{CDCl}_3$ .



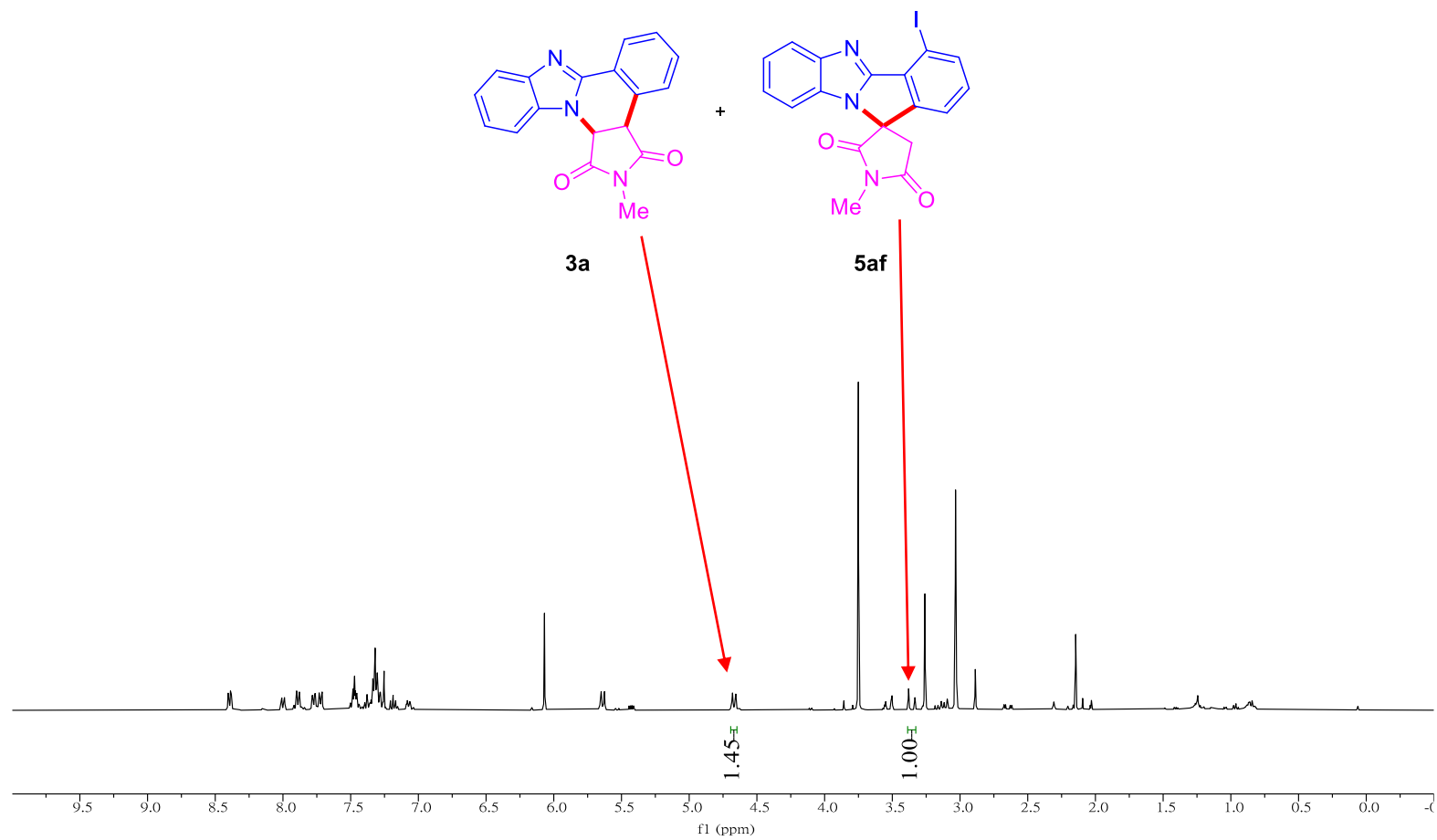
$^{13}\text{C}$  NMR spectrum (150 MHz) of compound **5ae** in  $\text{CDCl}_3$ .



## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
382.0183	1	C <sub>18</sub> H <sub>13</sub> BrN <sub>3</sub> O <sub>2</sub>	382.0186	-0.8	24.6	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **5ae**.

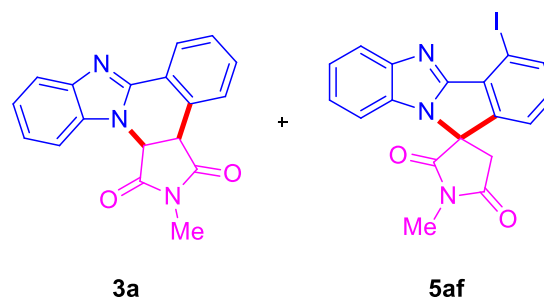
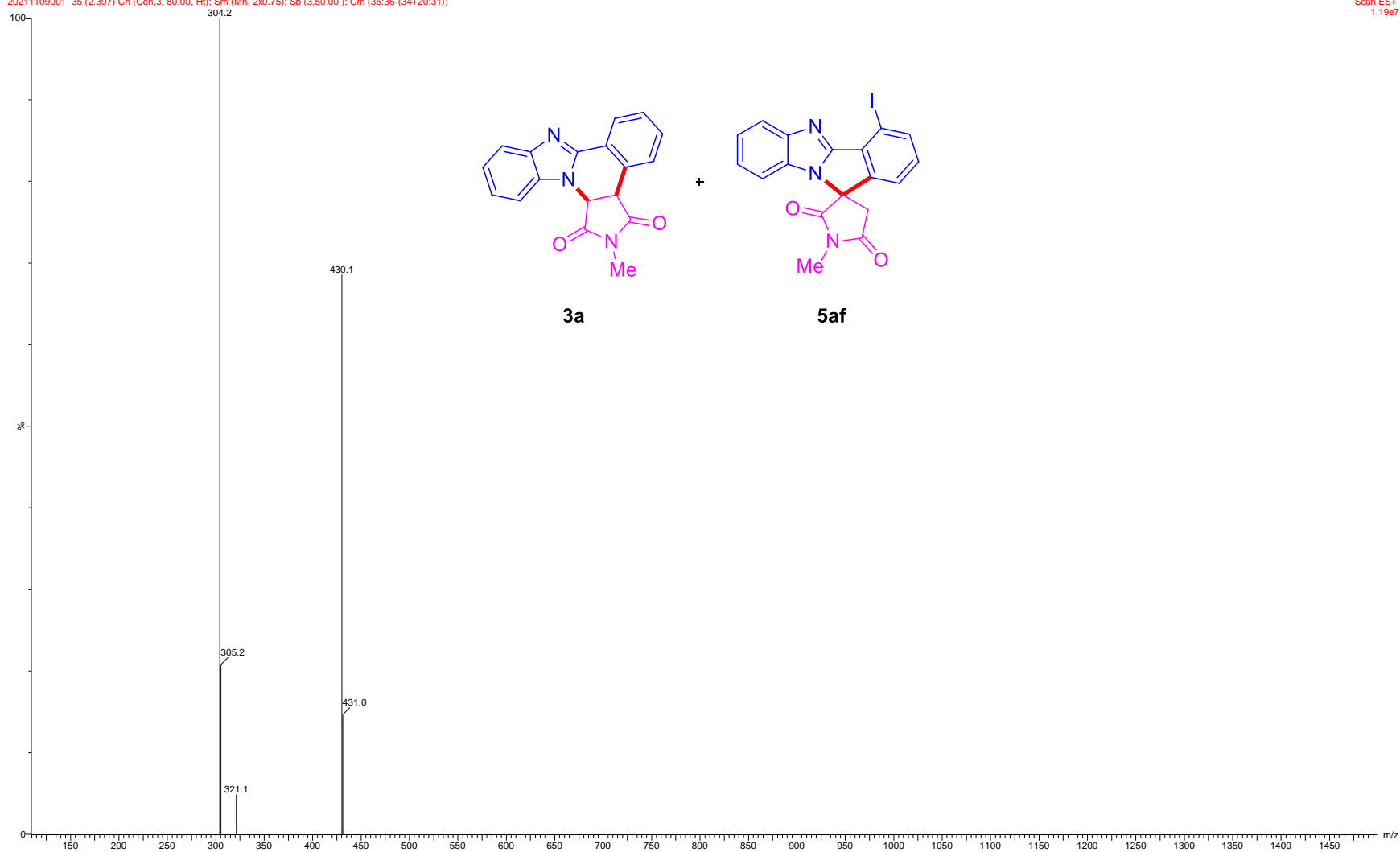


<sup>1</sup>H NMR spectrum (400 MHz) of compound **3a** and **5af** (ratio) in CDCl<sub>3</sub>.

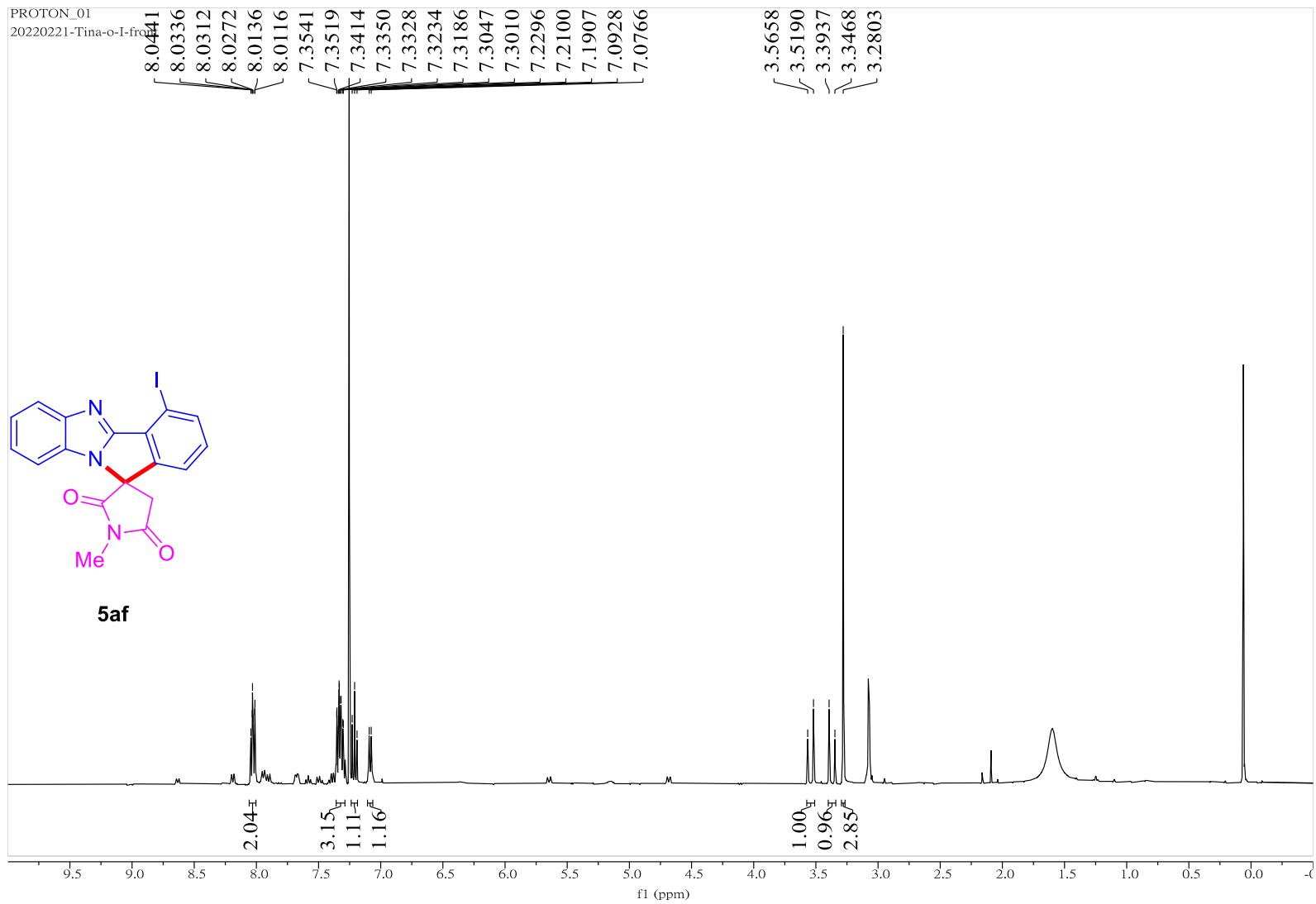
Tina-3095

20211109001 35 (2.397) Cn (Cen.3, 80.00, Ht); Sm (Mn, 2x0.75); Sb (3,50.00); Cm (35:36-(34+20:31))

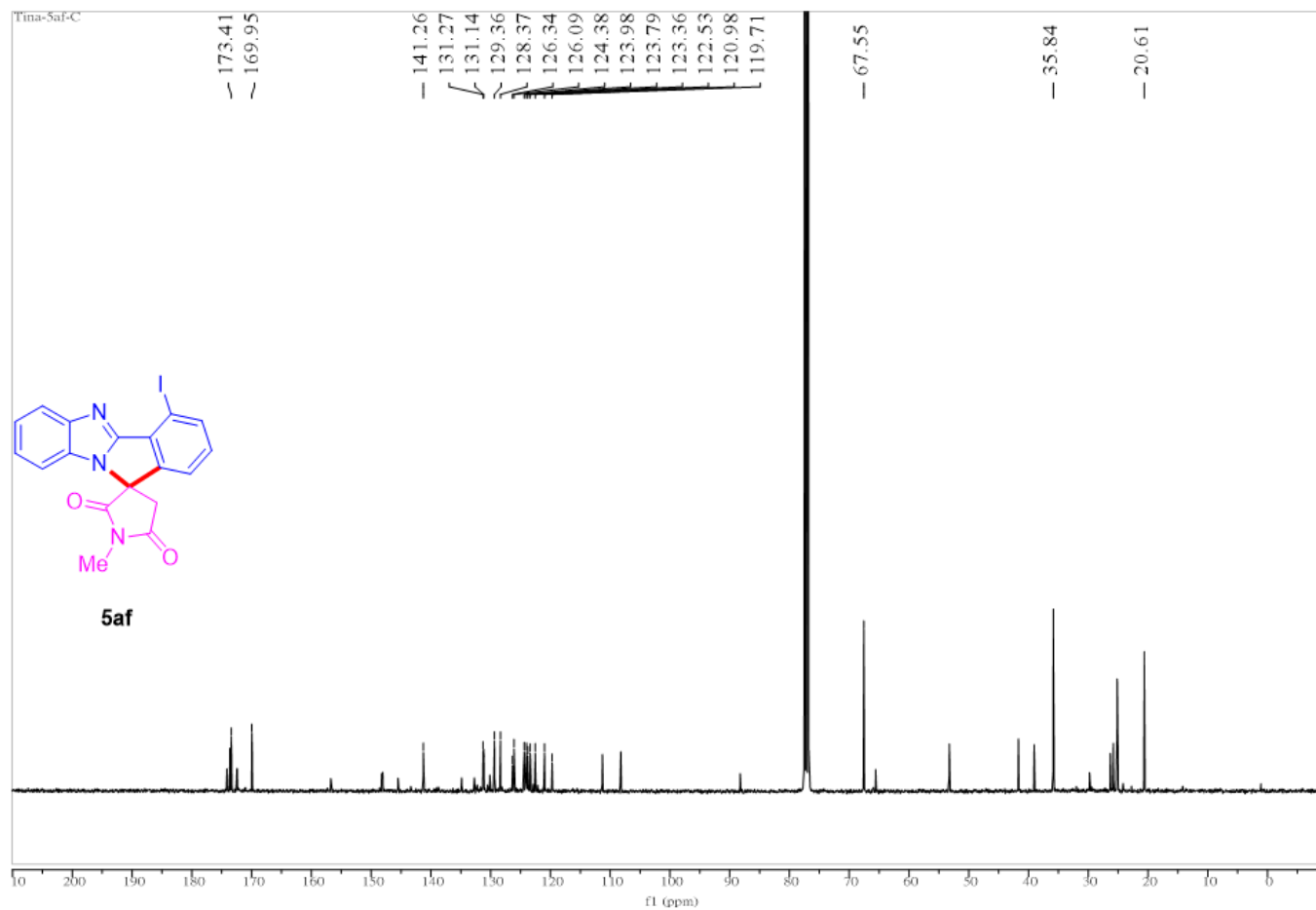
Scan ES+  
1.19e7

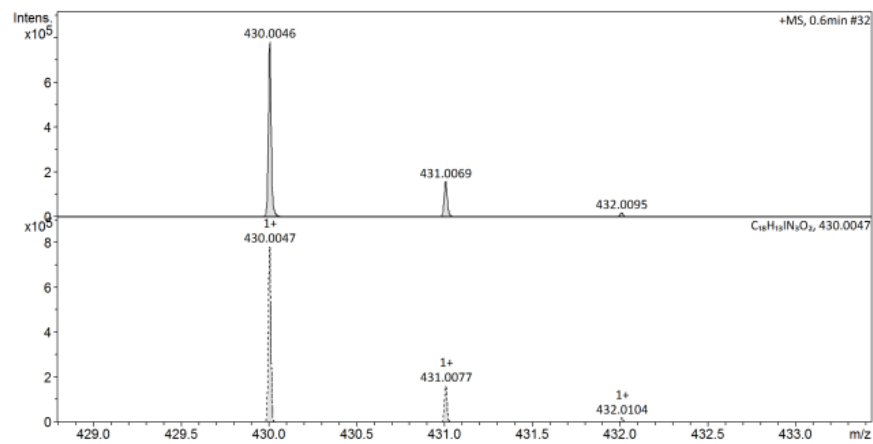
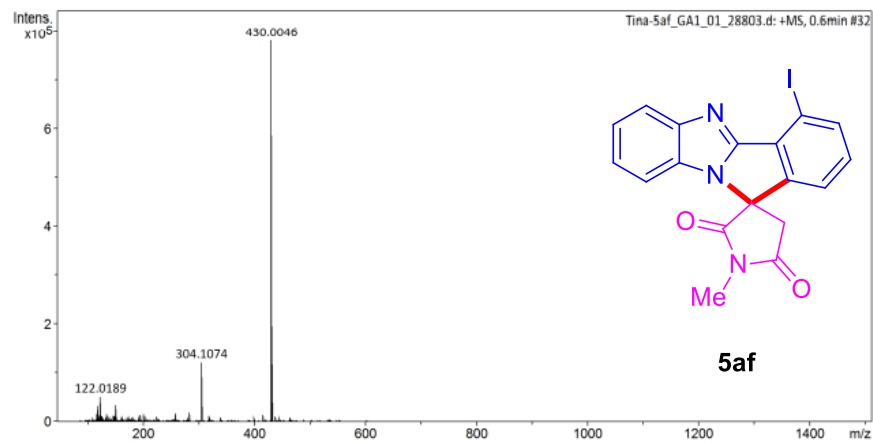


LRMS (ESI) of compound **3a** and **5af**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **5af** in  $\text{CDCl}_3$ .



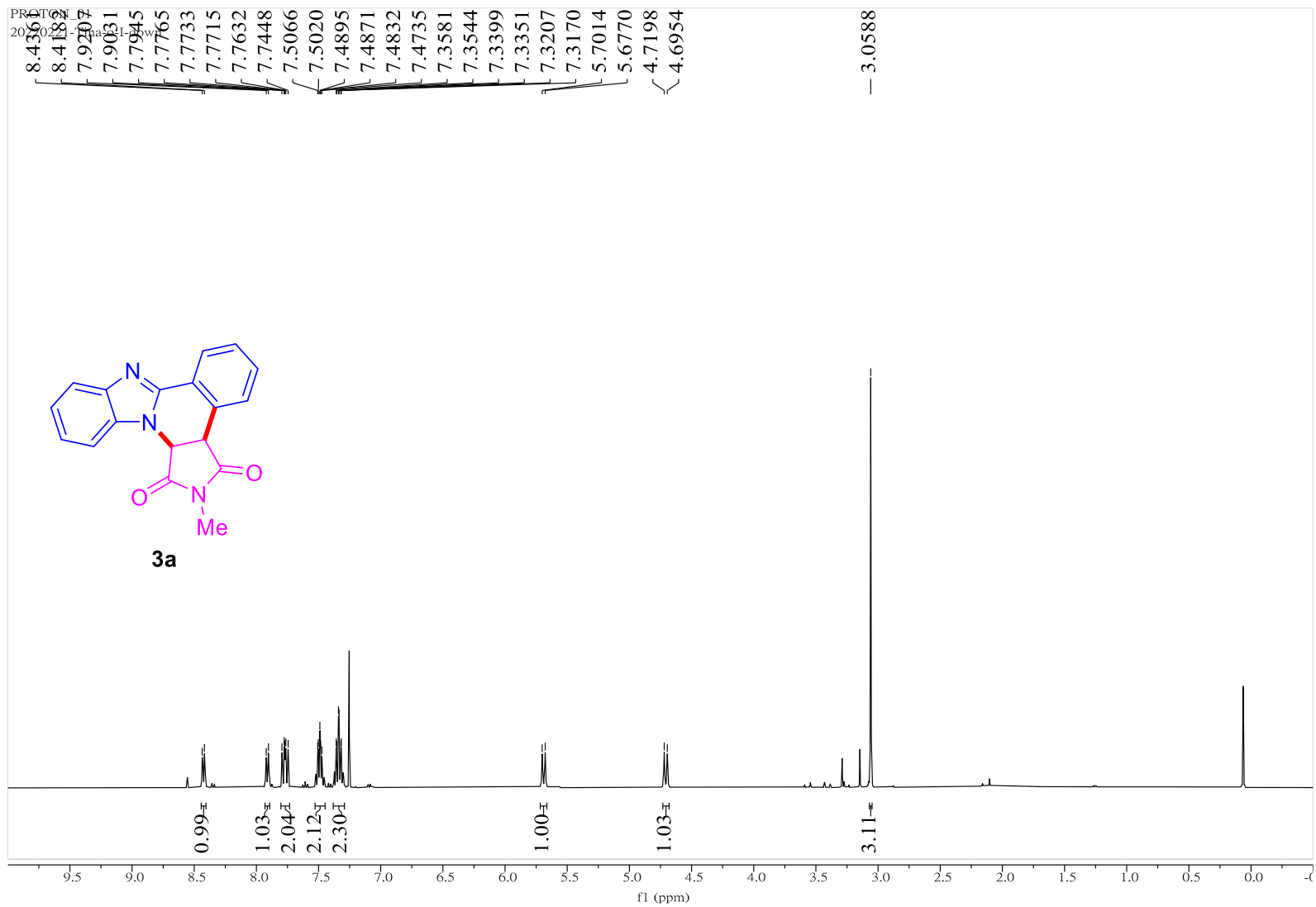


## Display Report

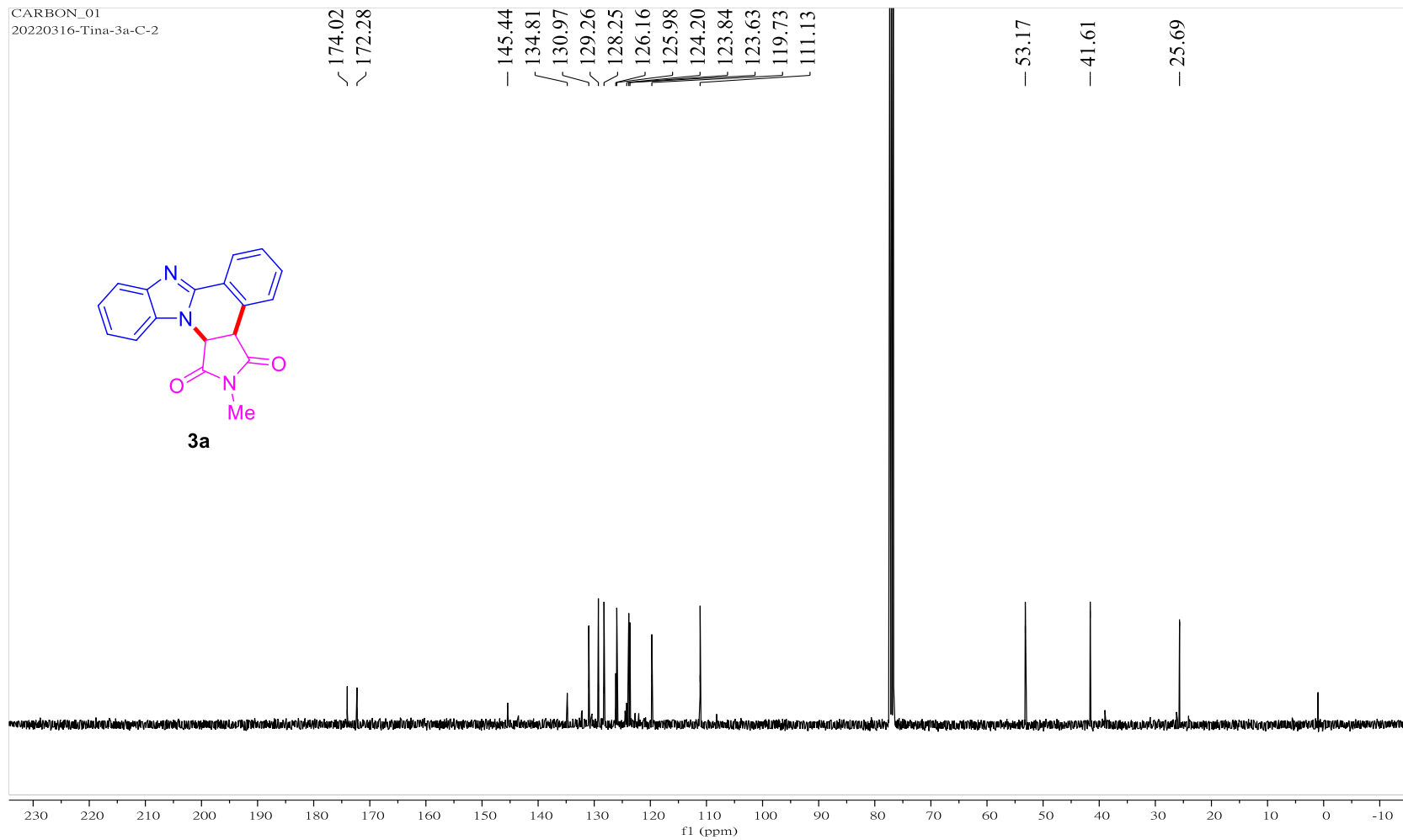
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
430.0046	1	C <sub>18</sub> H <sub>13</sub> IN <sub>3</sub> O <sub>2</sub>	430.0047	0.4	4.1	1	100.00	13.5	even	ok	M+H

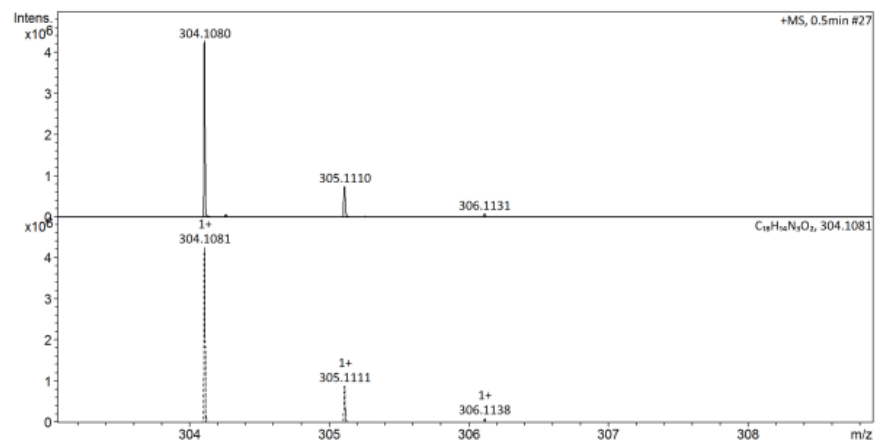
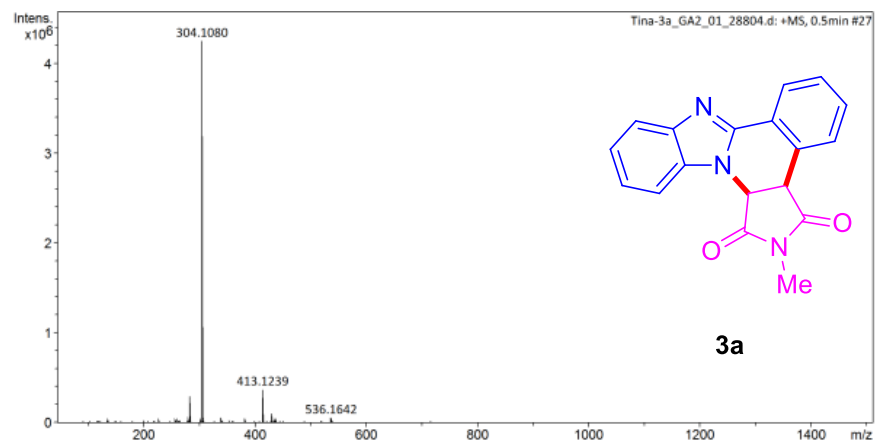
HRMS (ESI) of compound **5af**.





$^1\text{H}$  NMR spectrum (400 MHz) of compound **3a** in  $\text{CDCl}_3$ .

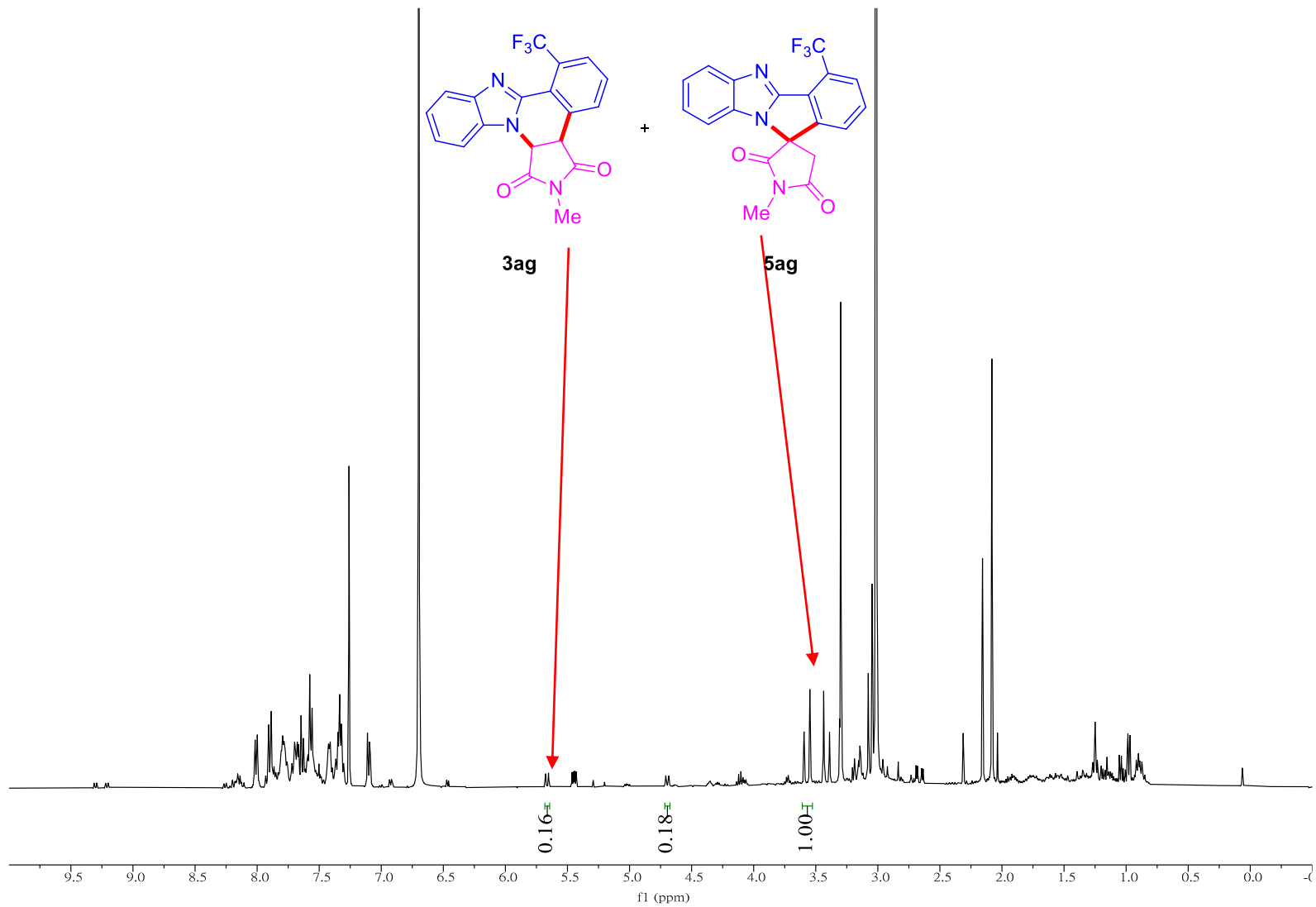




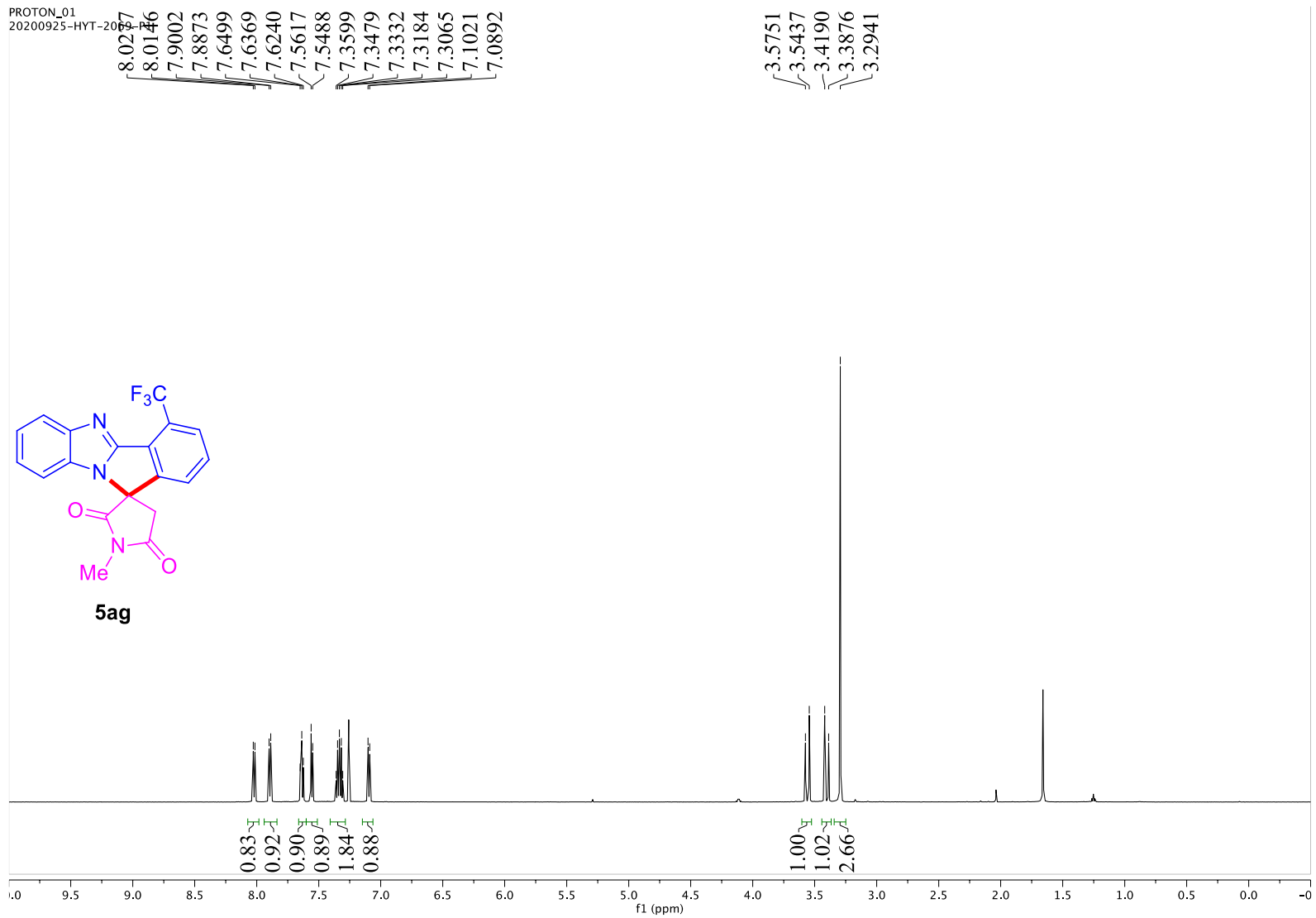
### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
304.1080	1	C <sub>18</sub> H <sub>14</sub> N <sub>3</sub> O <sub>2</sub>	304.1081	-0.2	18.7	1	100.00	13.5	even	ok	M+H

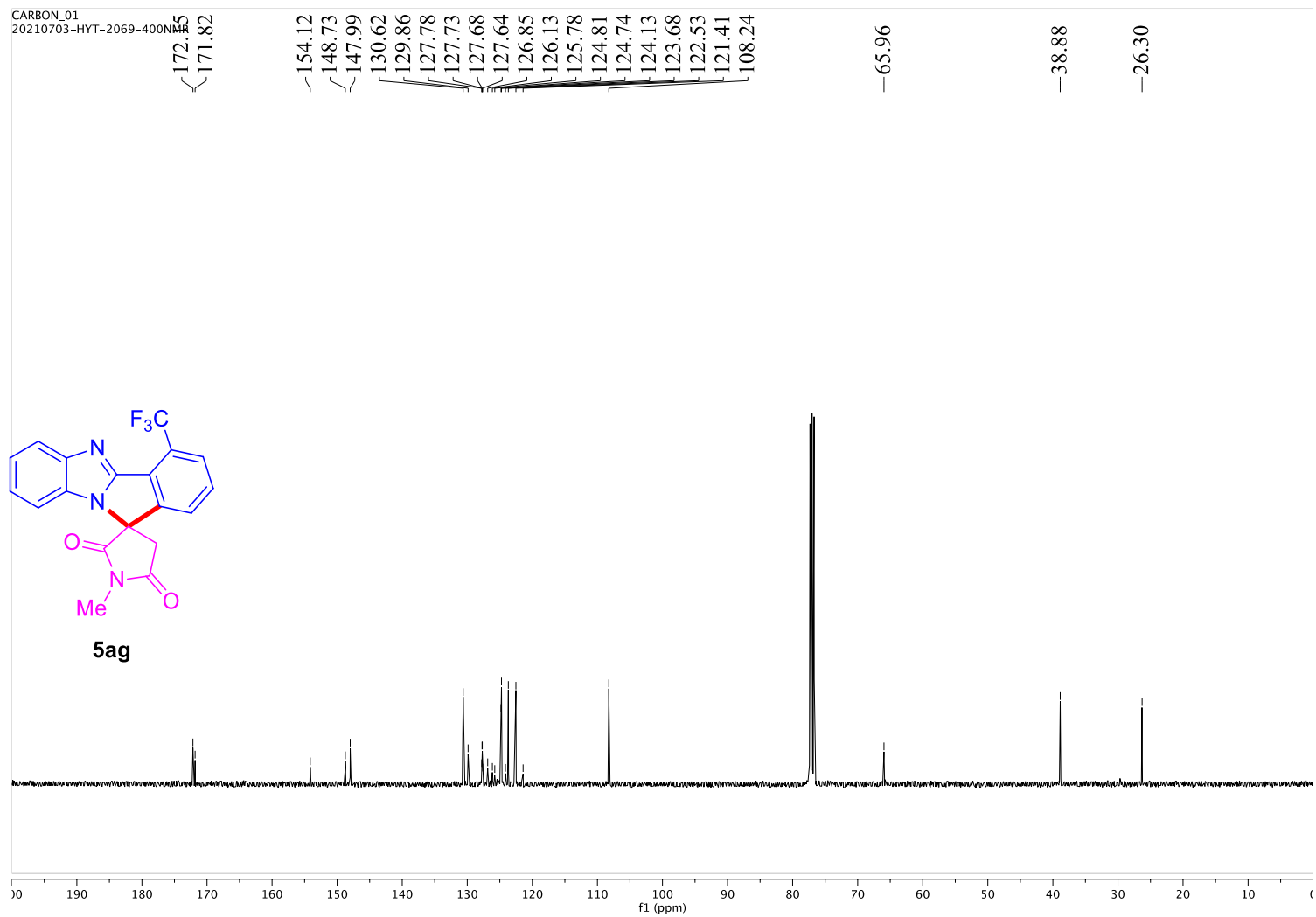
HRMS (ESI) of compound **3a**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3ag** and **5ag** in  $\text{CDCl}_3$ .

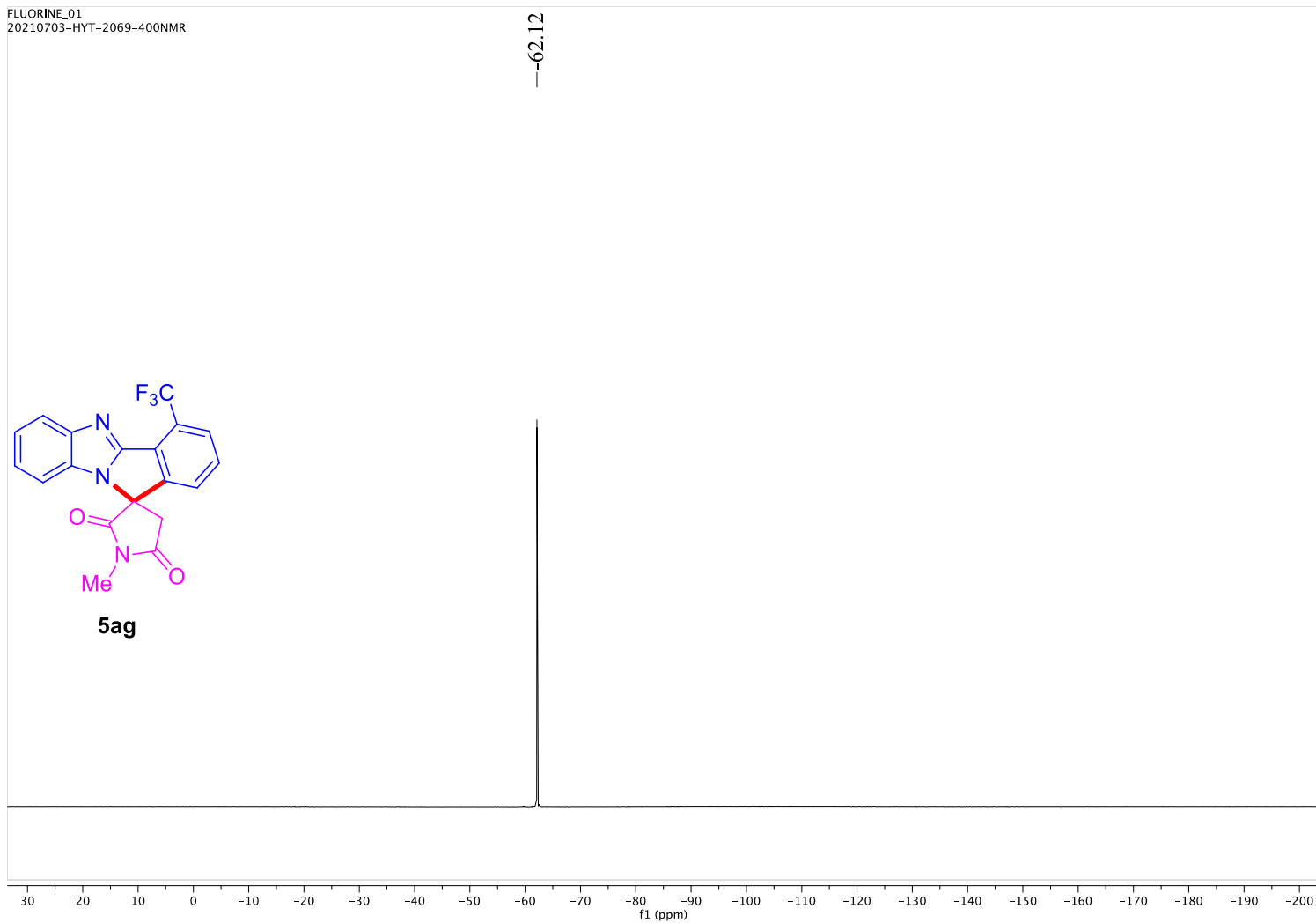


<sup>1</sup>H NMR spectrum (400 MHz) of compound **5ag** in CDCl<sub>3</sub>.

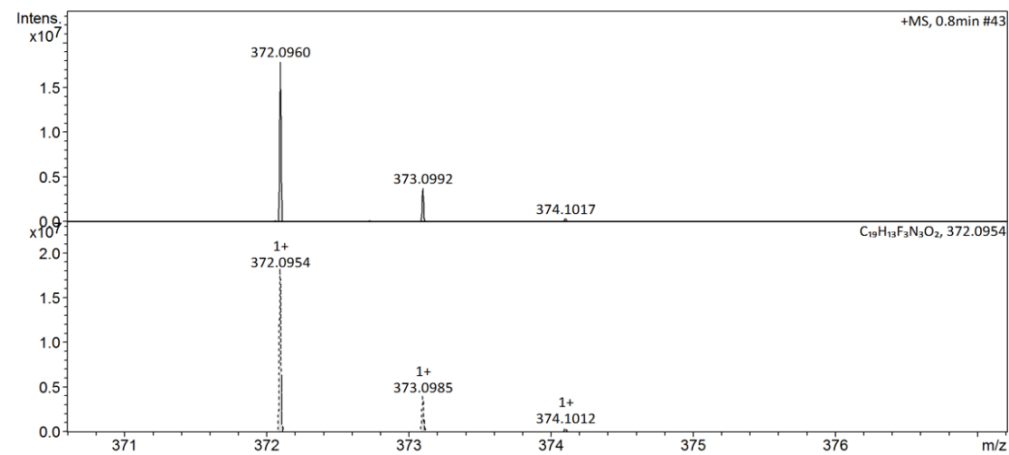
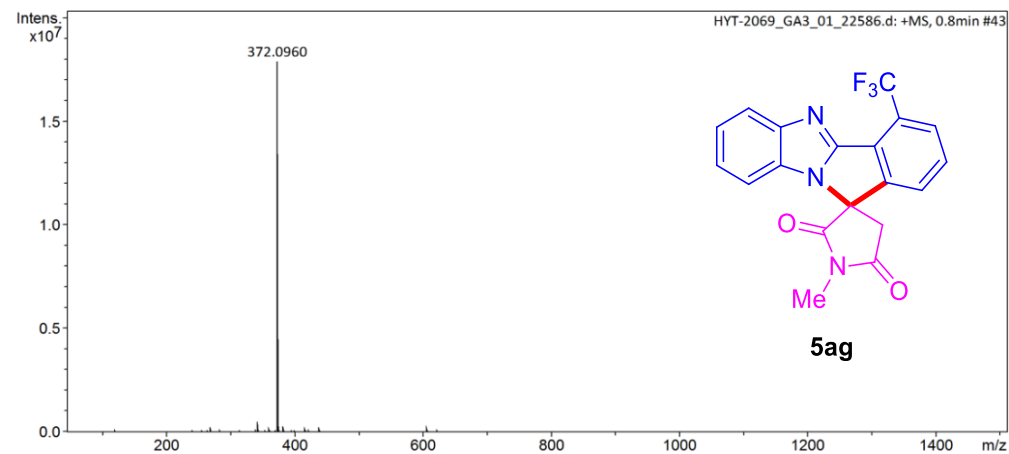


$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5ag** in  $\text{CDCl}_3$ .

FLUORINE\_01  
20210703-HYT-2069-400NMR



$^{19}F$  NMR spectrum (376 MHz) of compound **5ag** in  $CDCl_3$ .

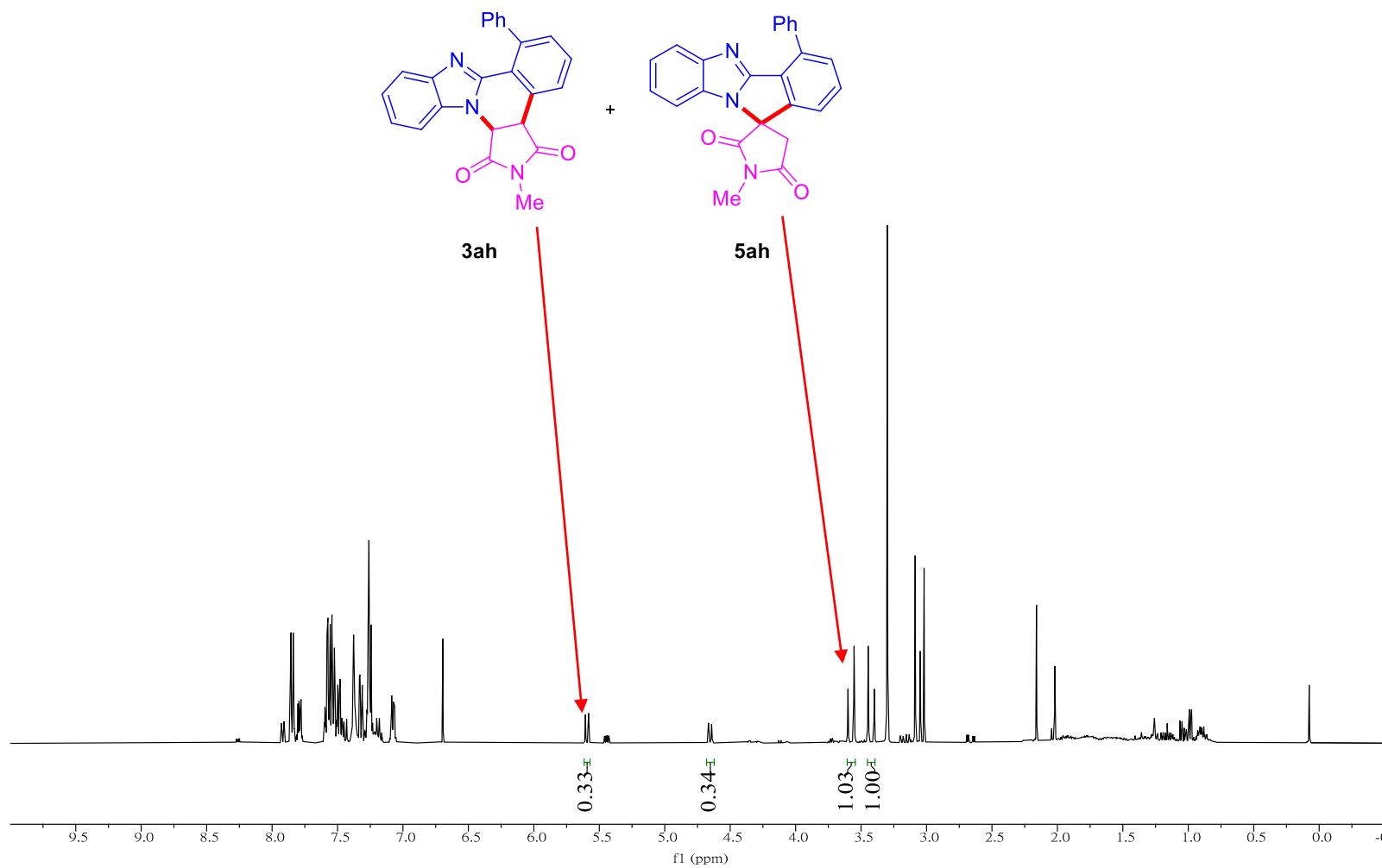


## Display Report

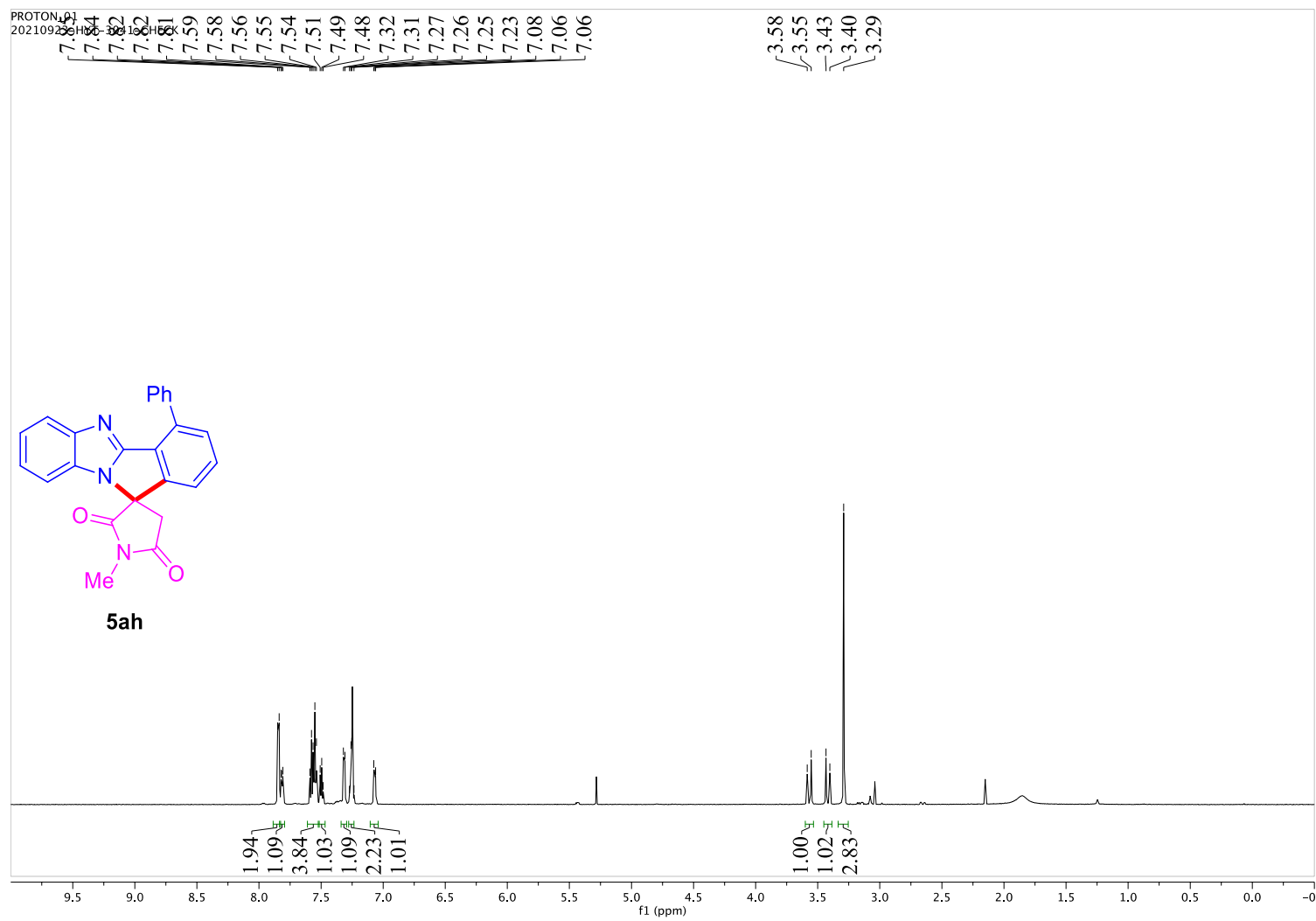
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
372.0960	1	C <sub>19</sub> H <sub>13</sub> F <sub>3</sub> N <sub>3</sub> O <sub>2</sub>	372.0954	-1.6	5.8	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **5ag**.

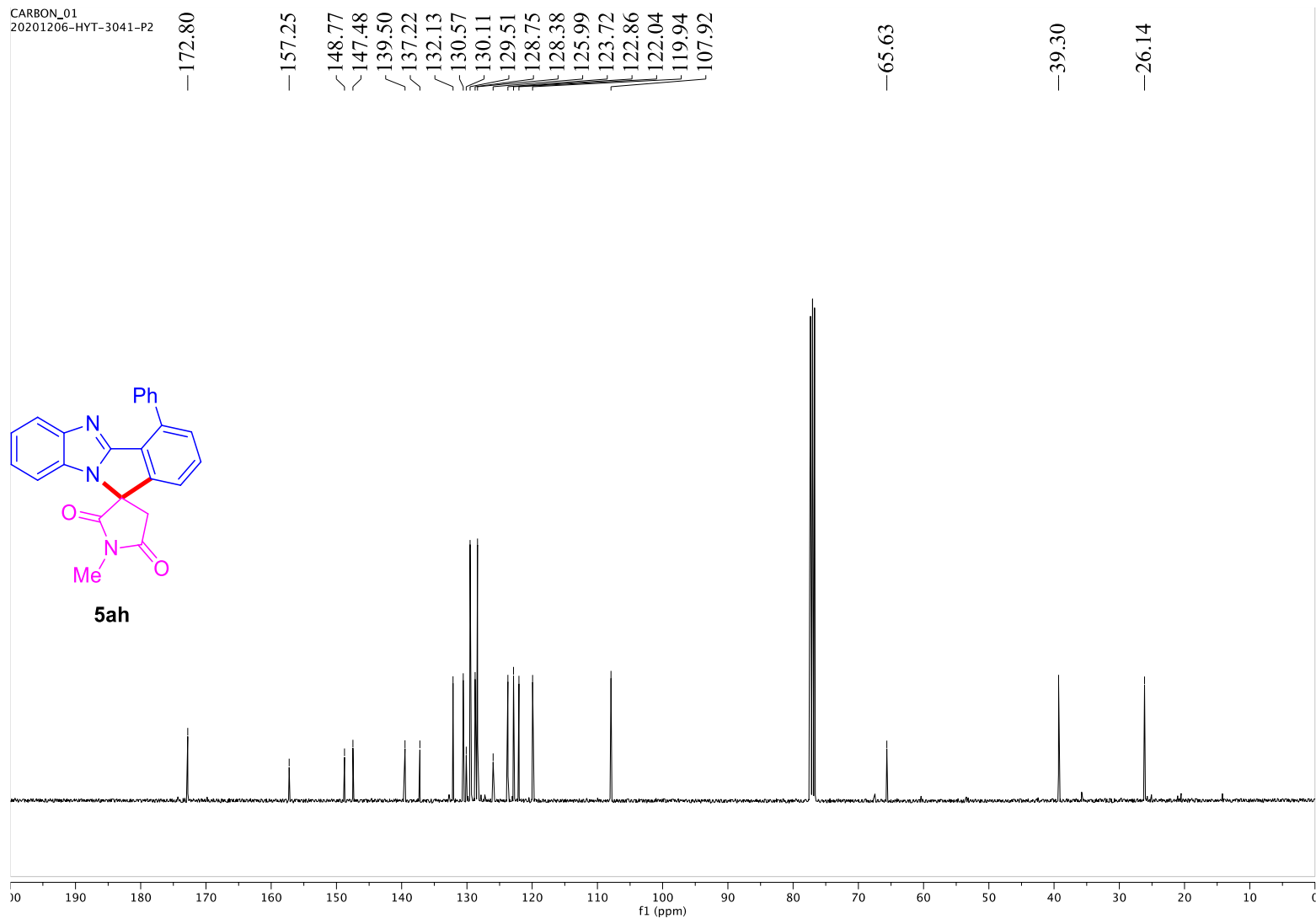




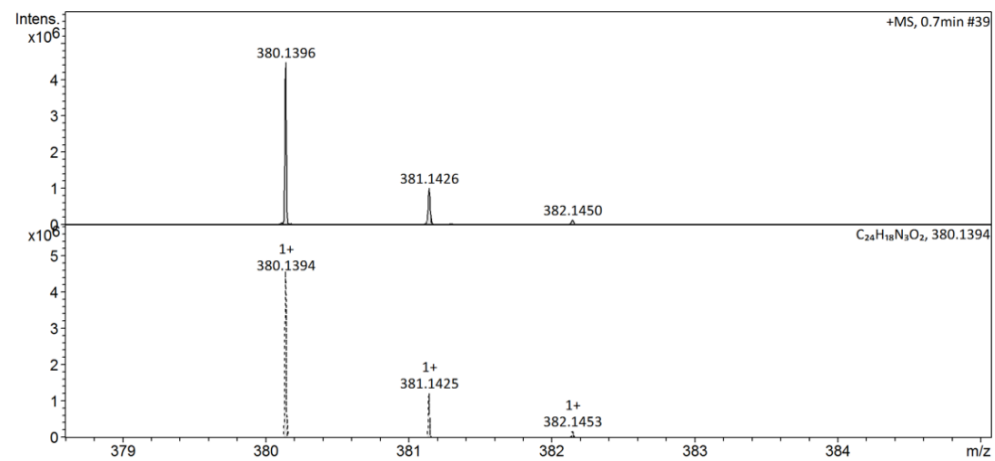
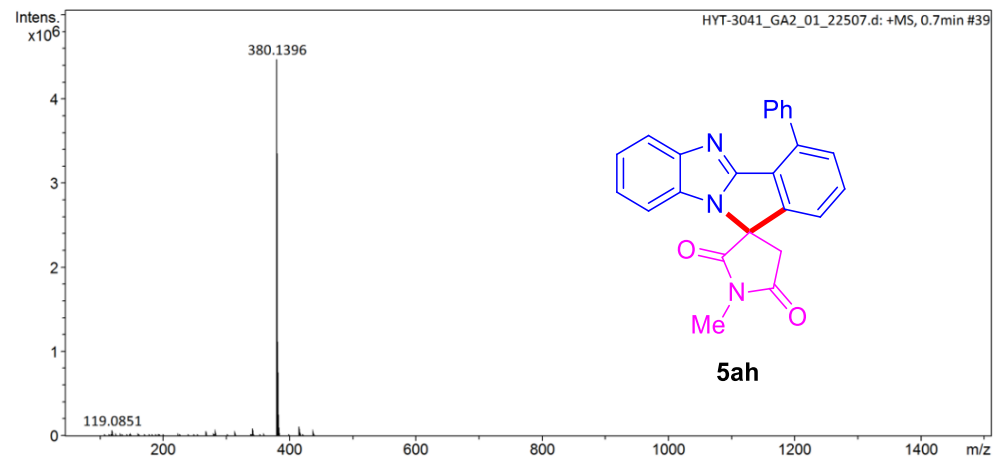
<sup>1</sup>H NMR spectrum (400 MHz) of compound **3ah** and **5ah** in CDCl<sub>3</sub>.



$^1\text{H}$  NMR spectrum (600 MHz) of compound **5ah** in  $\text{CDCl}_3$ .



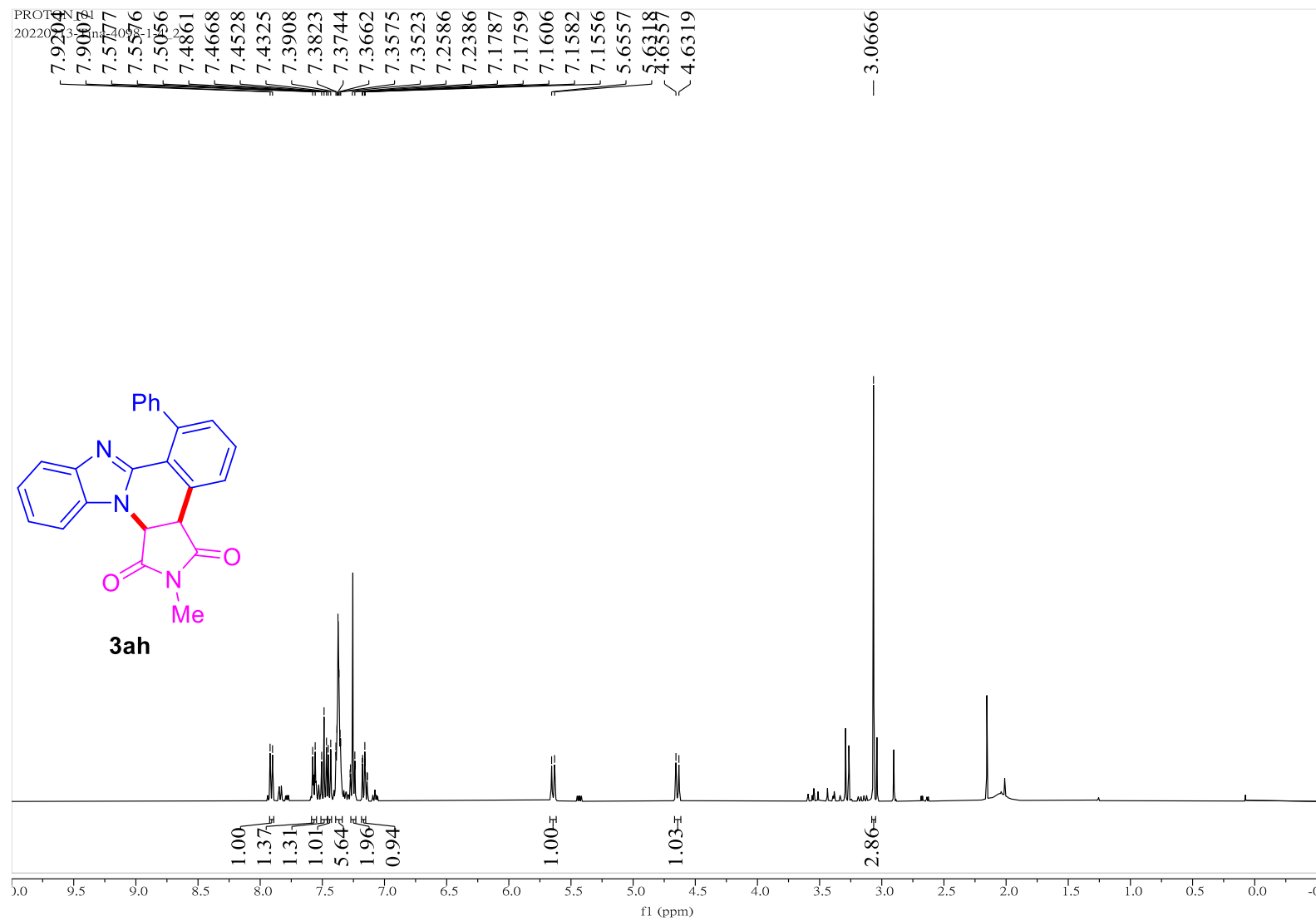
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5ah** in  $\text{CDCl}_3$ .



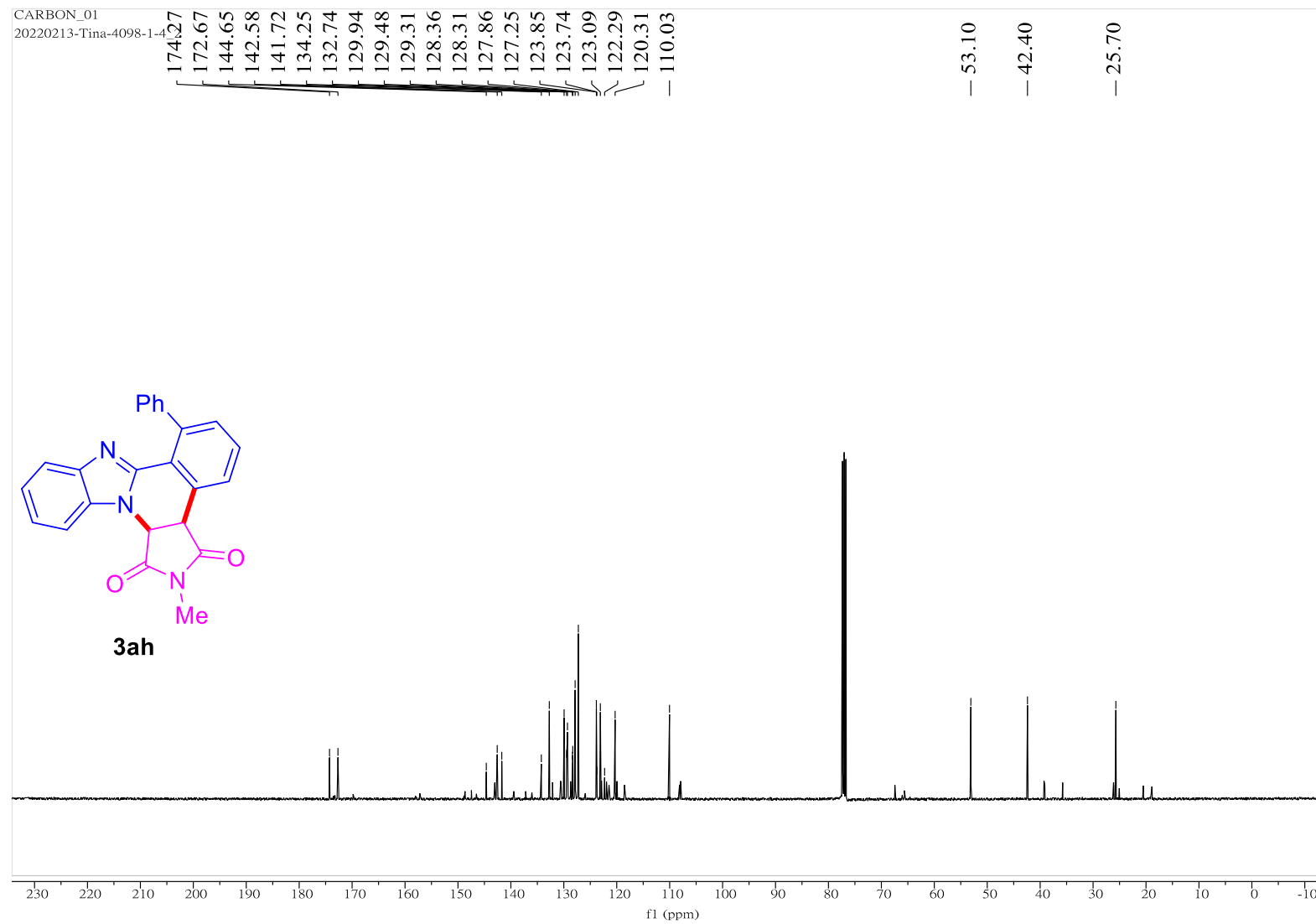
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
380.1396	1	C <sub>24</sub> H <sub>18</sub> N <sub>4</sub> O <sub>2</sub>	380.1394	0.6	24.3	1	100.00	17.5	even	ok	M+H

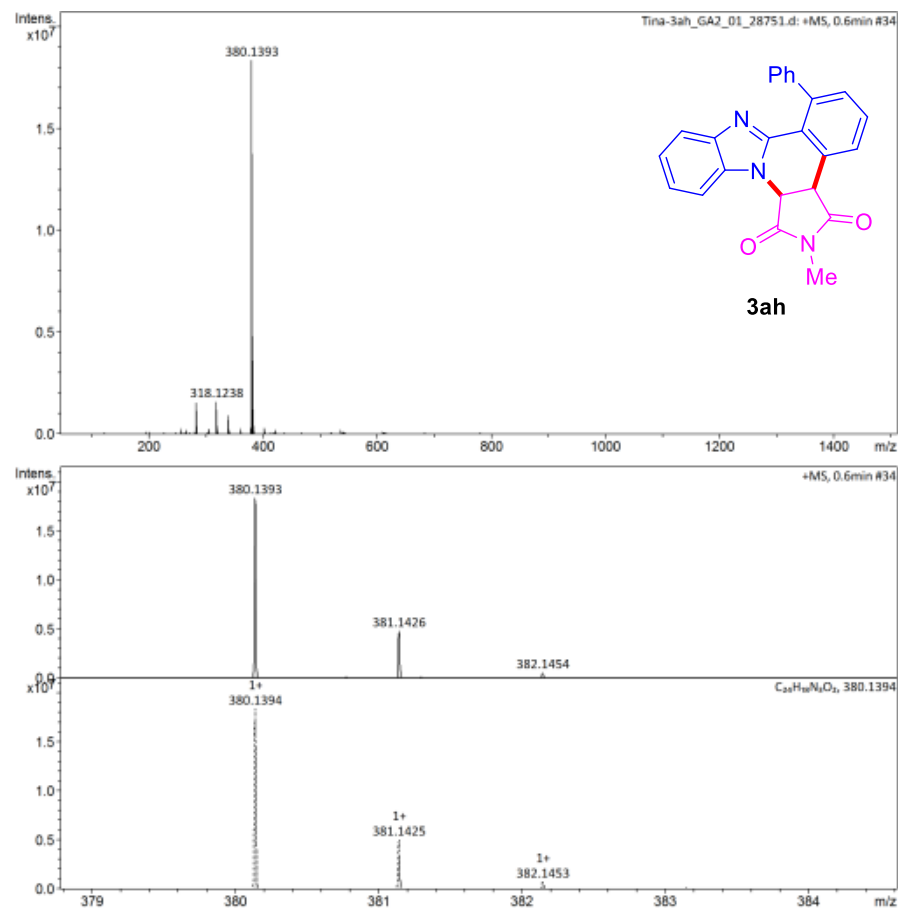
HRMS (ESI) of compound **5ah**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3ah** in  $\text{CDCl}_3$ .



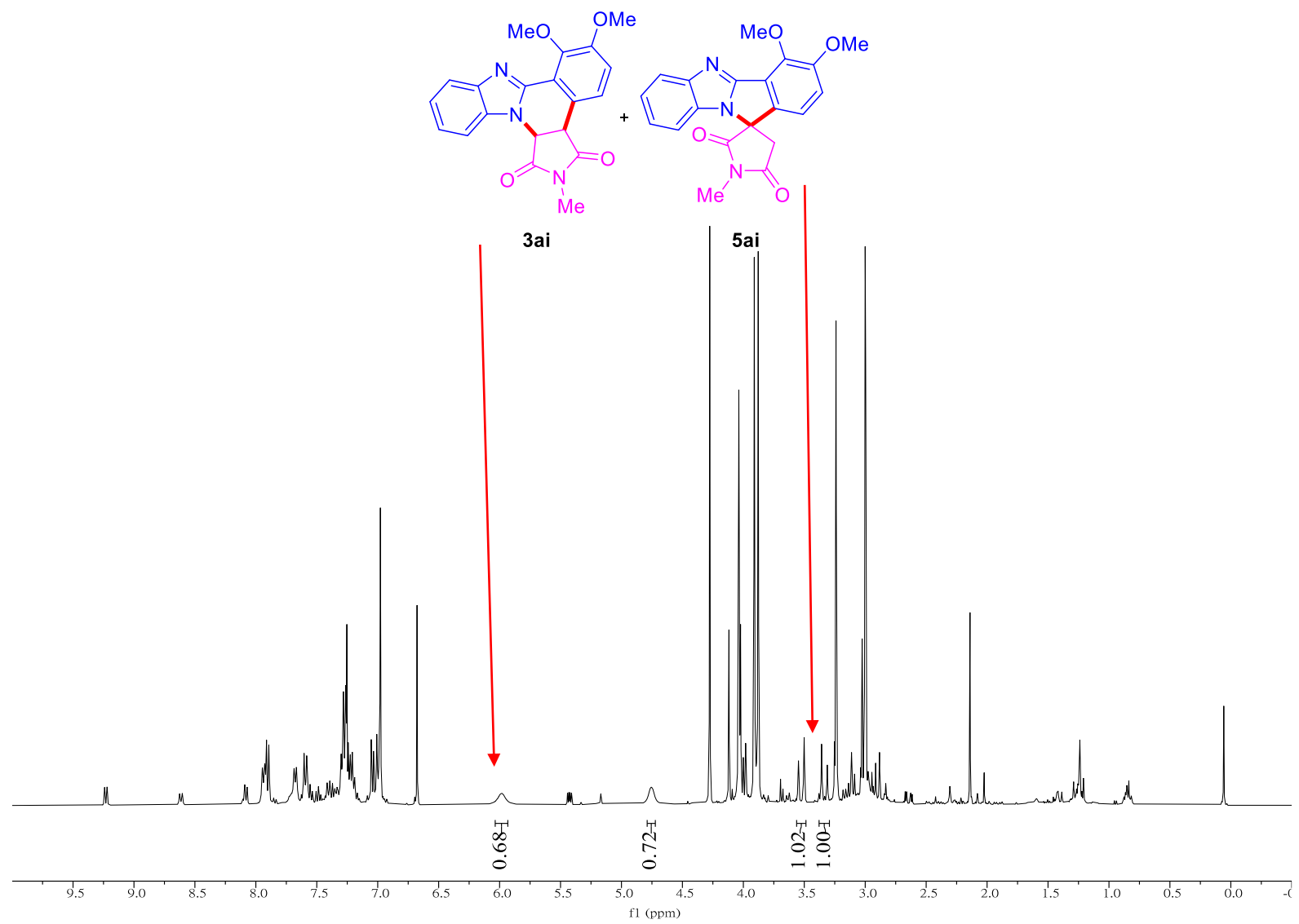
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3ah** in  $\text{CDCl}_3$ .



## Display Report

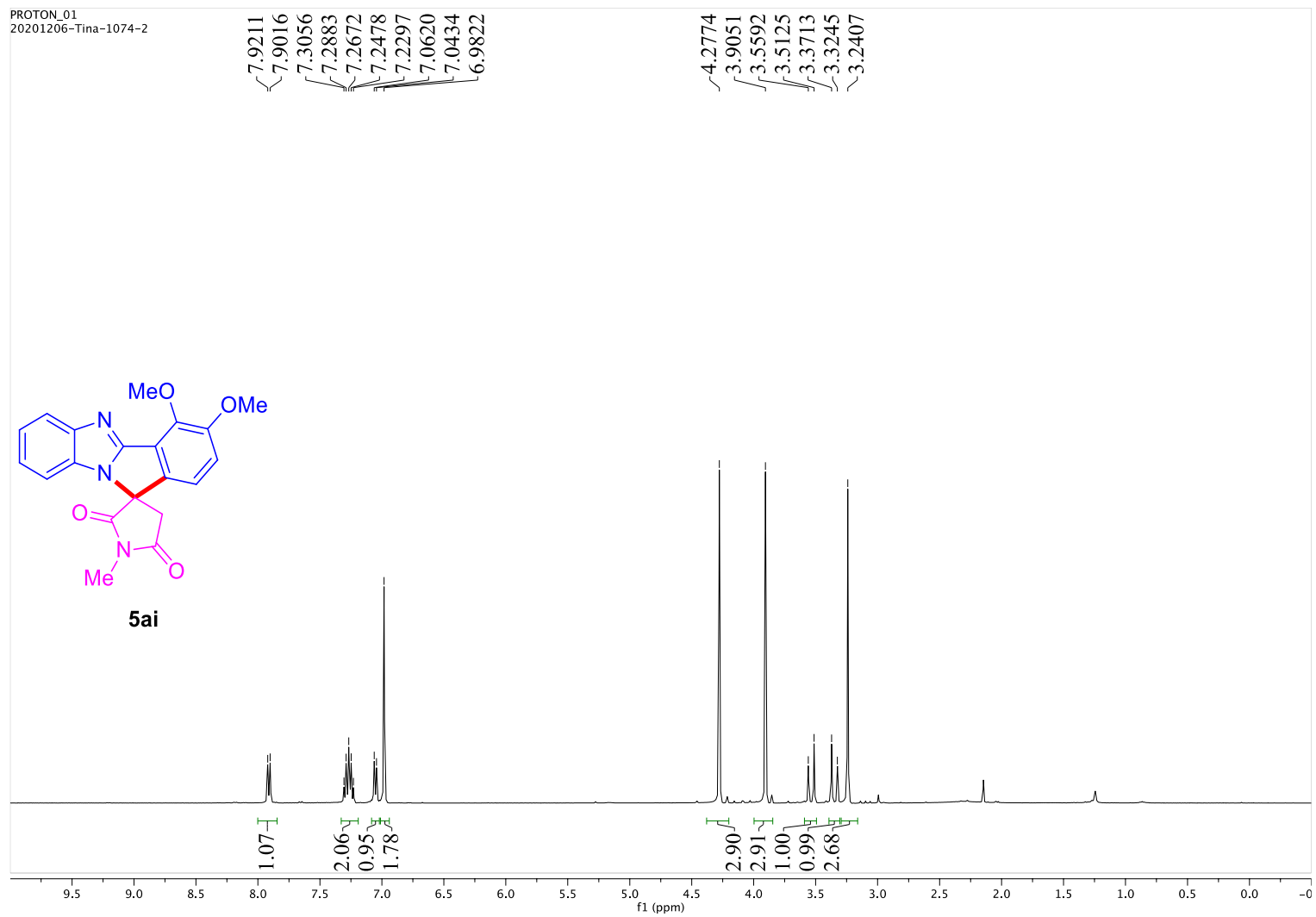
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
380.1393	1	C <sub>24</sub> H <sub>18</sub> N <sub>3</sub> O <sub>2</sub>	380.1394	-0.1	7.8	1	100.00	17.5	even	ok	M+H

HRMS (ESI) of compound **3ah**.

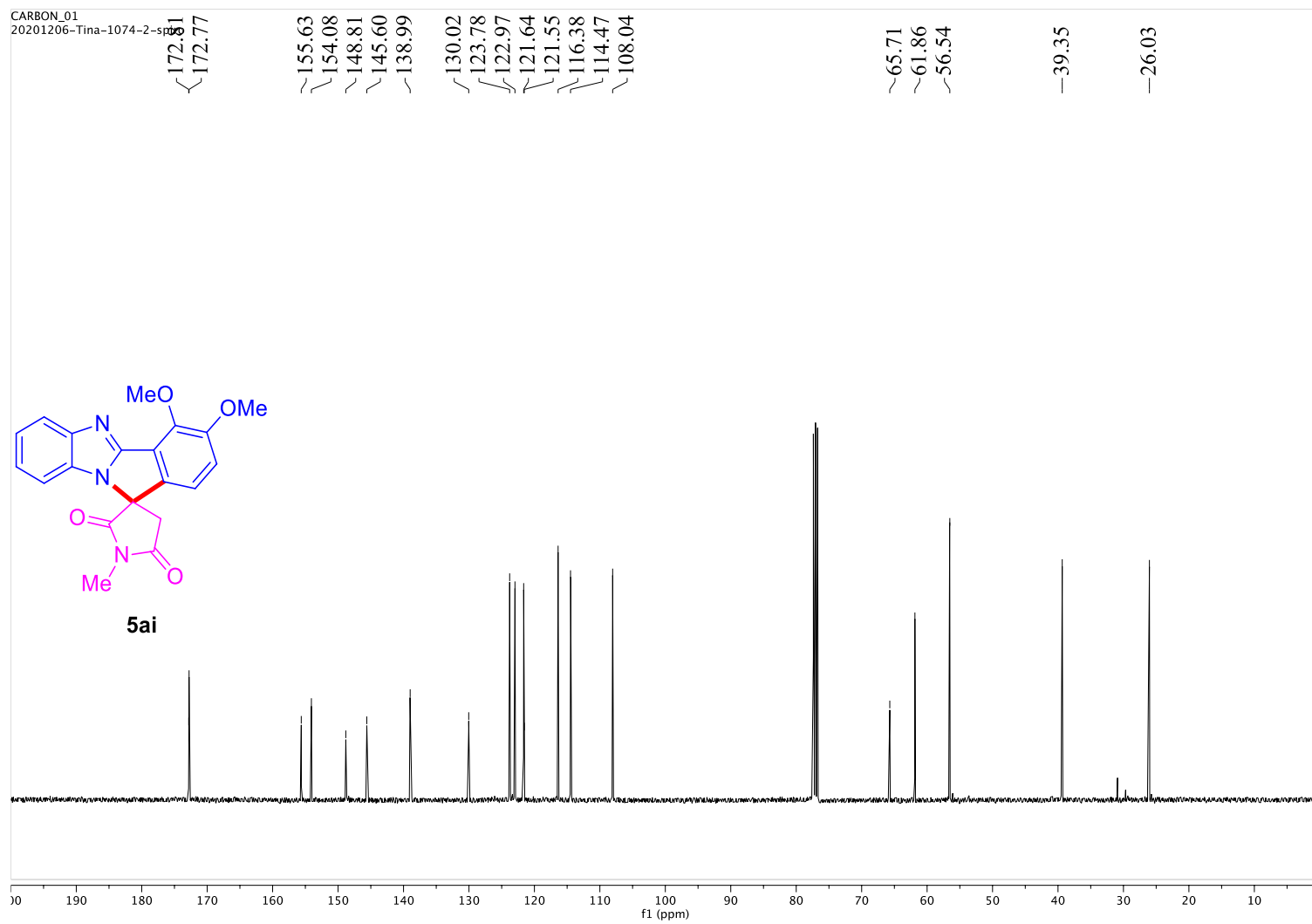


<sup>1</sup>H NMR spectrum (400 MHz) of compound **3ai** and **5ai** in CDCl<sub>3</sub>.

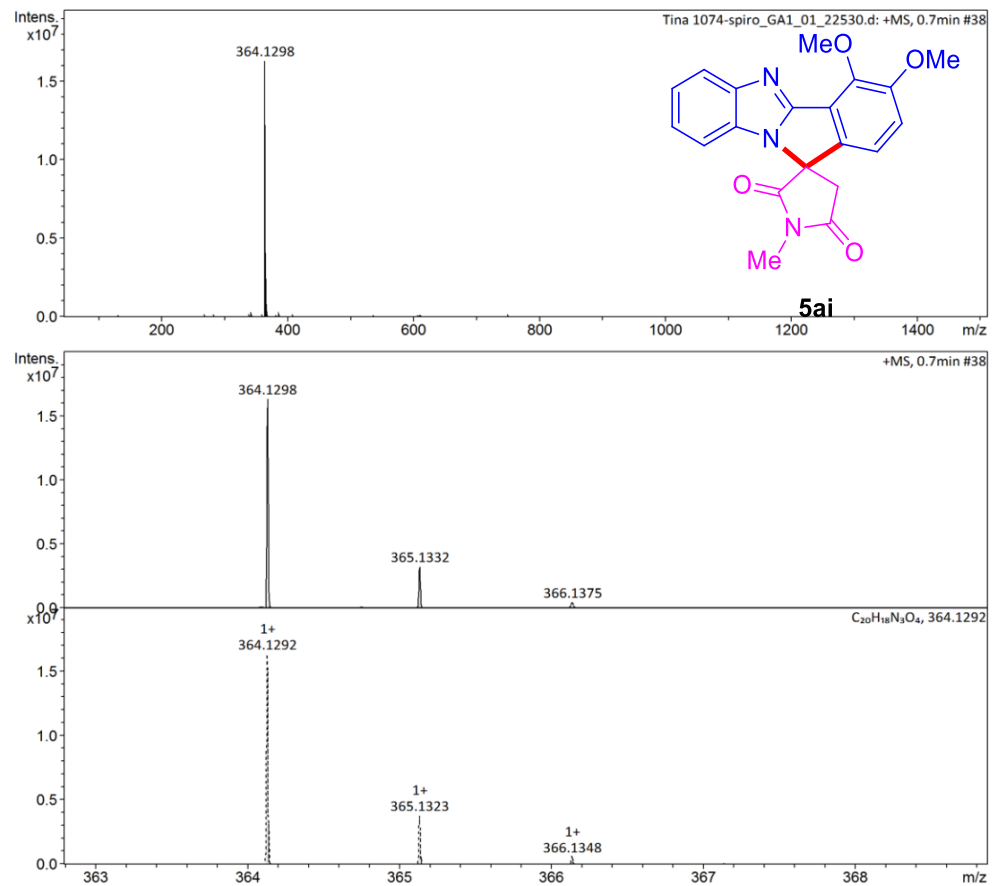




$^1\text{H}$  NMR spectrum (400 MHz) of compound **5ai** in  $\text{CDCl}_3$ .



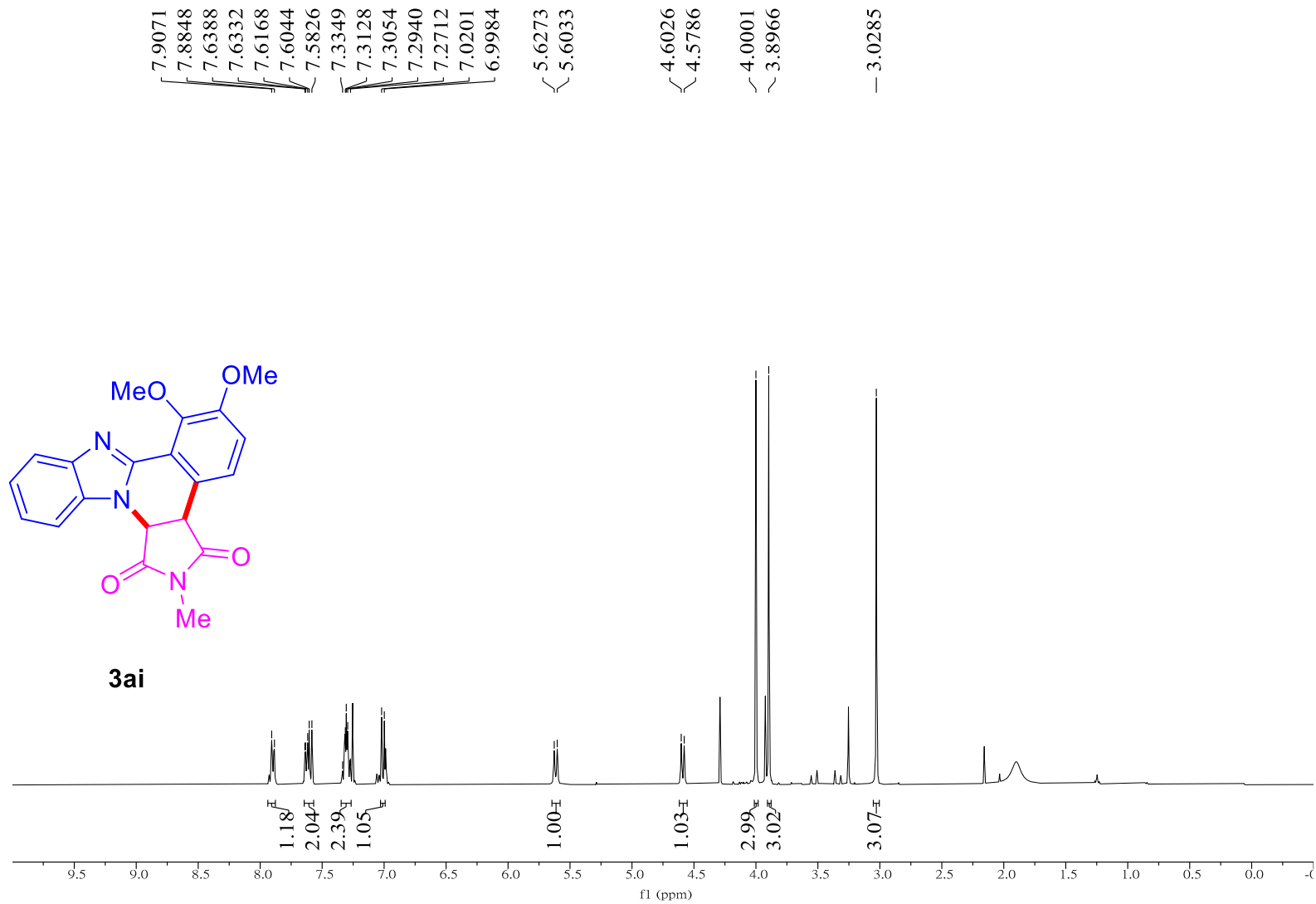
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5ai** in  $\text{CDCl}_3$ .



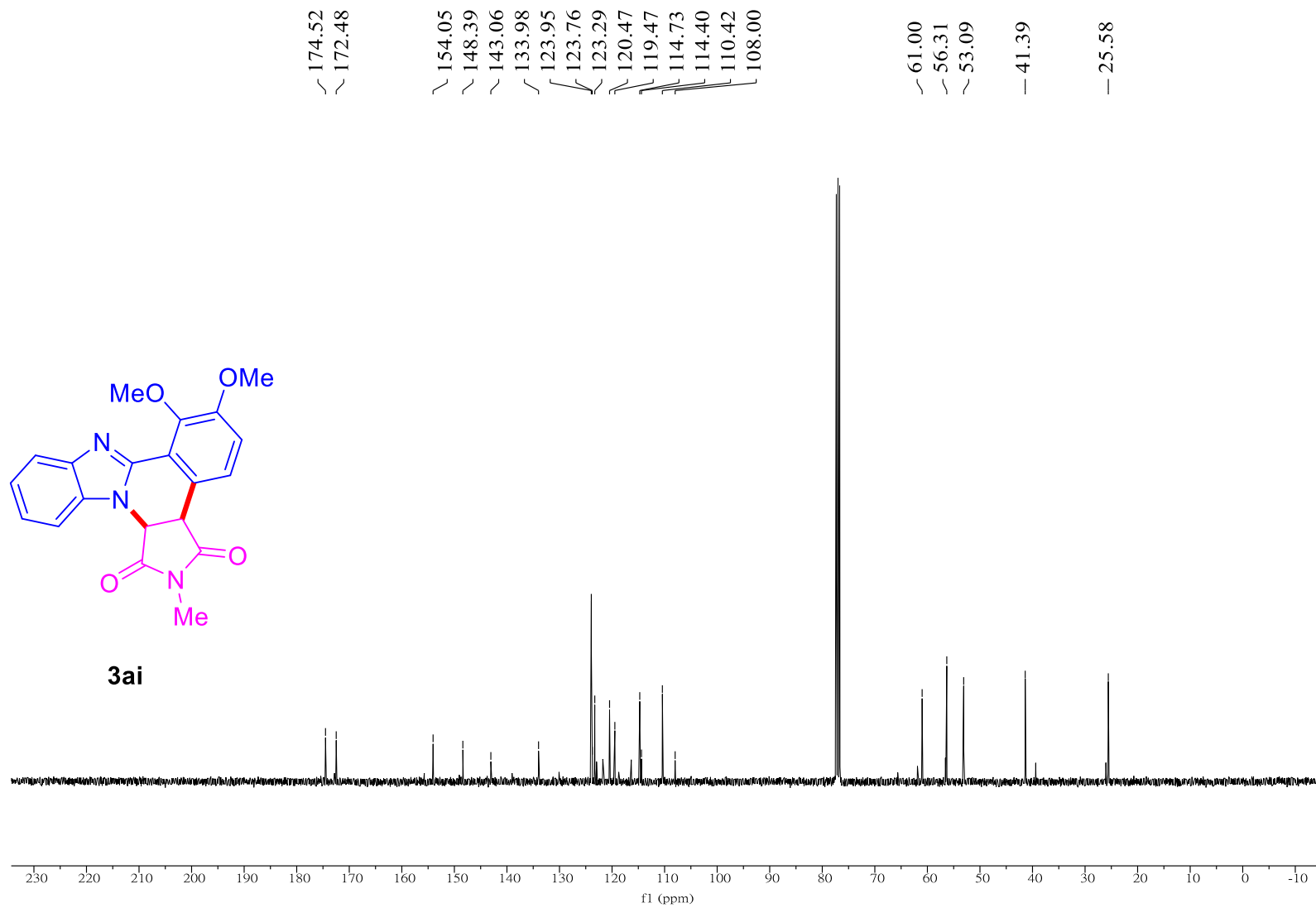
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
364.1298	1	C <sub>20</sub> H <sub>18</sub> N <sub>2</sub> O <sub>4</sub>	364.1292	-1.8	16.0	1	100.00	13.5	even	ok	M+H

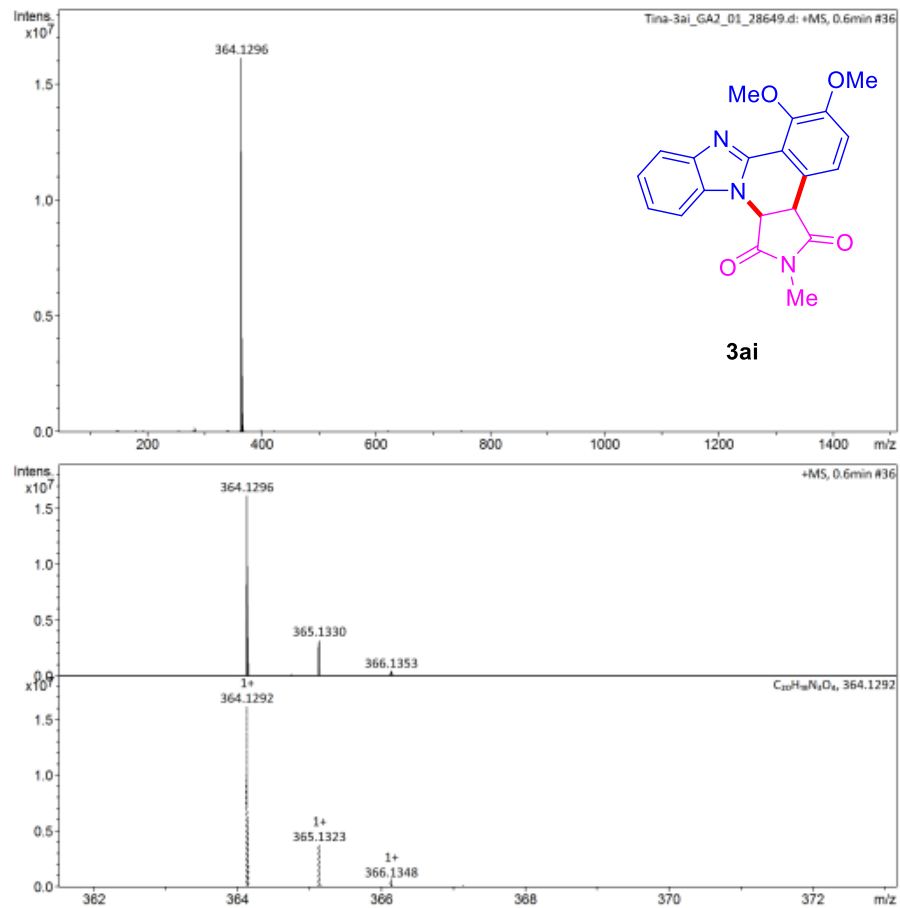
HRMS (ESI) of compound **5ai**.



<sup>1</sup>H NMR spectrum (400 MHz) of compound **3ai** in CDCl<sub>3</sub>.



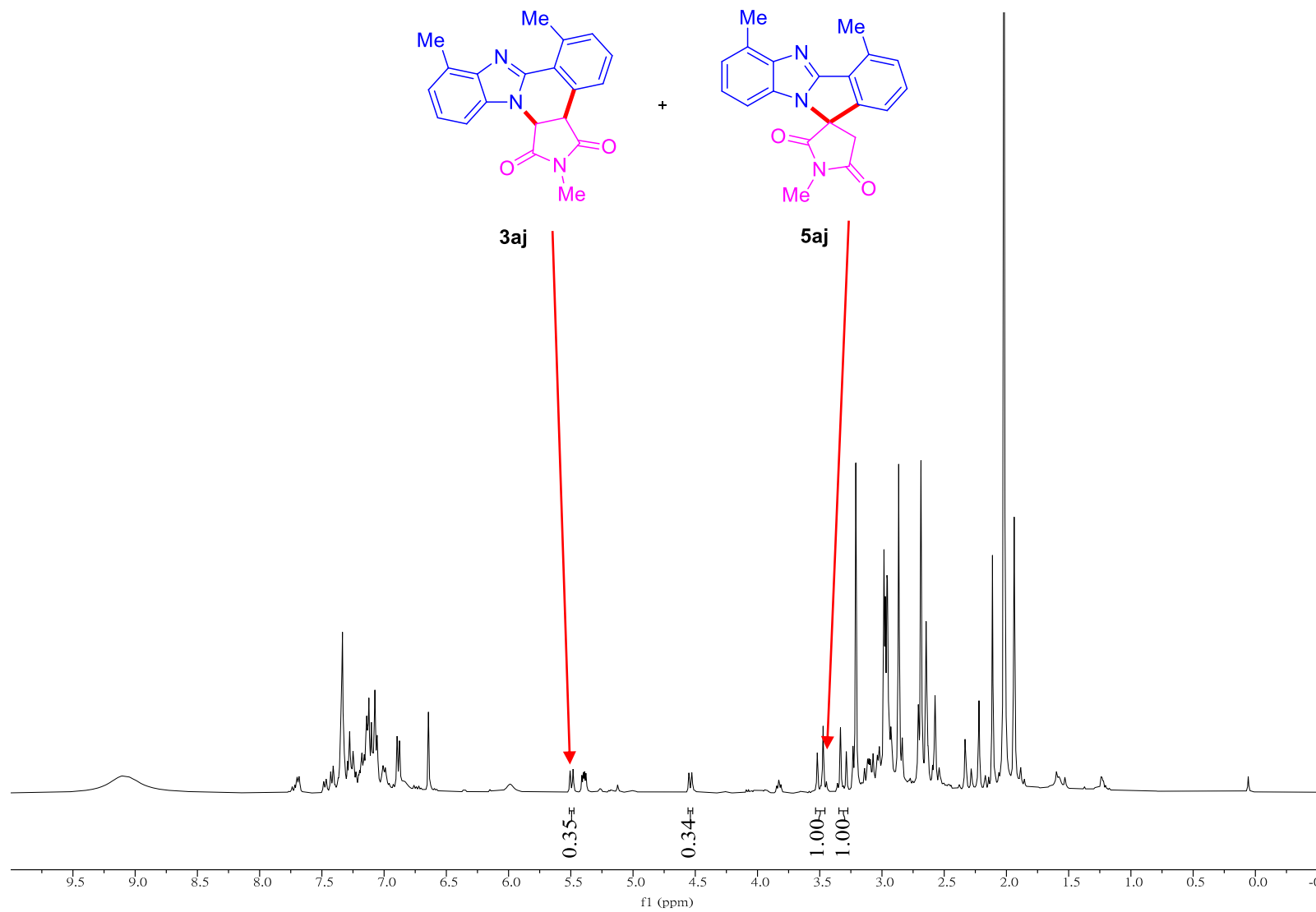
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3ai** in  $\text{CDCl}_3$ .



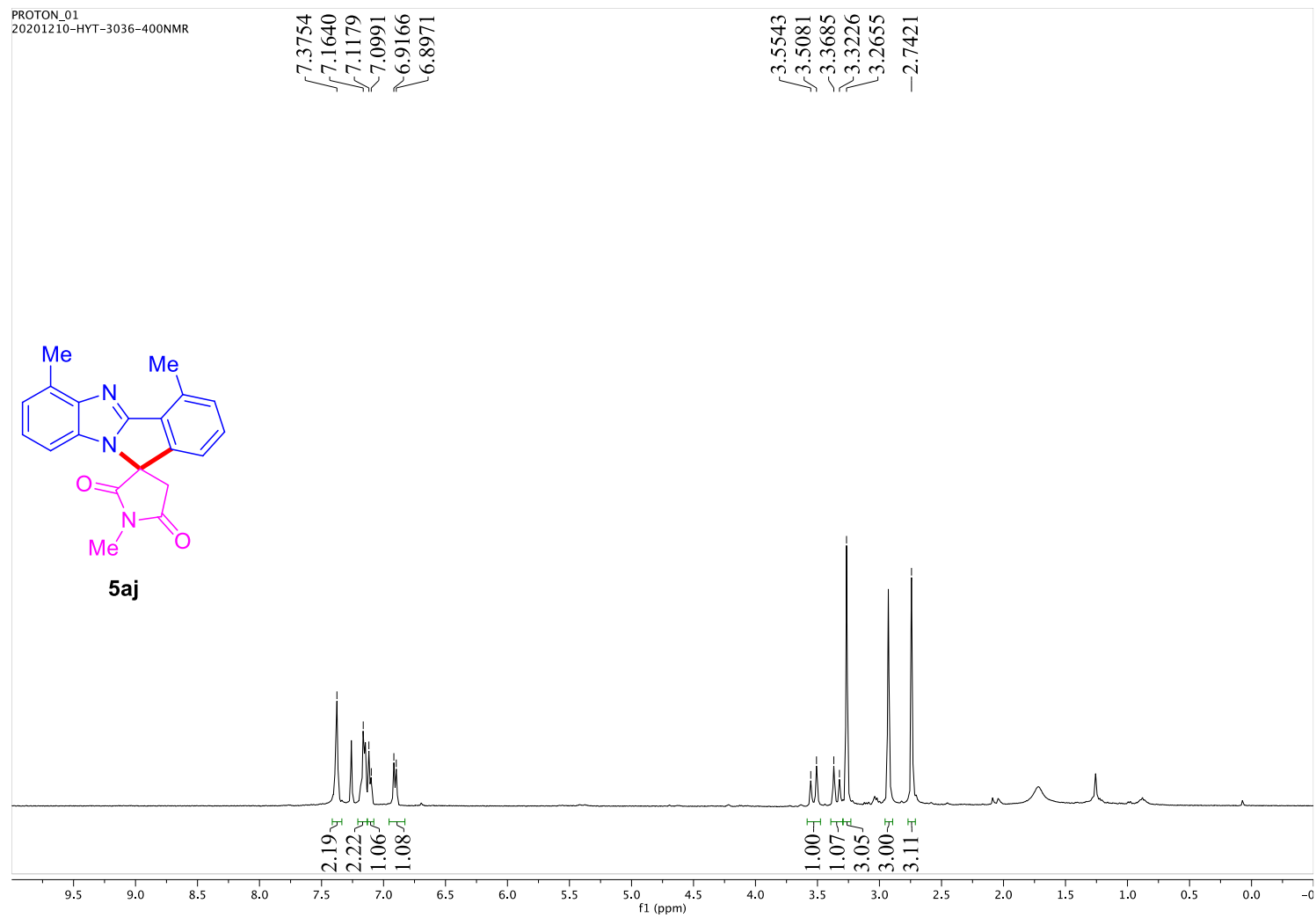
### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
364.1296	1	C <sub>20</sub> H <sub>18</sub> N <sub>3</sub> O <sub>4</sub>	364.1292	1.2	16.3	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **3ai**.

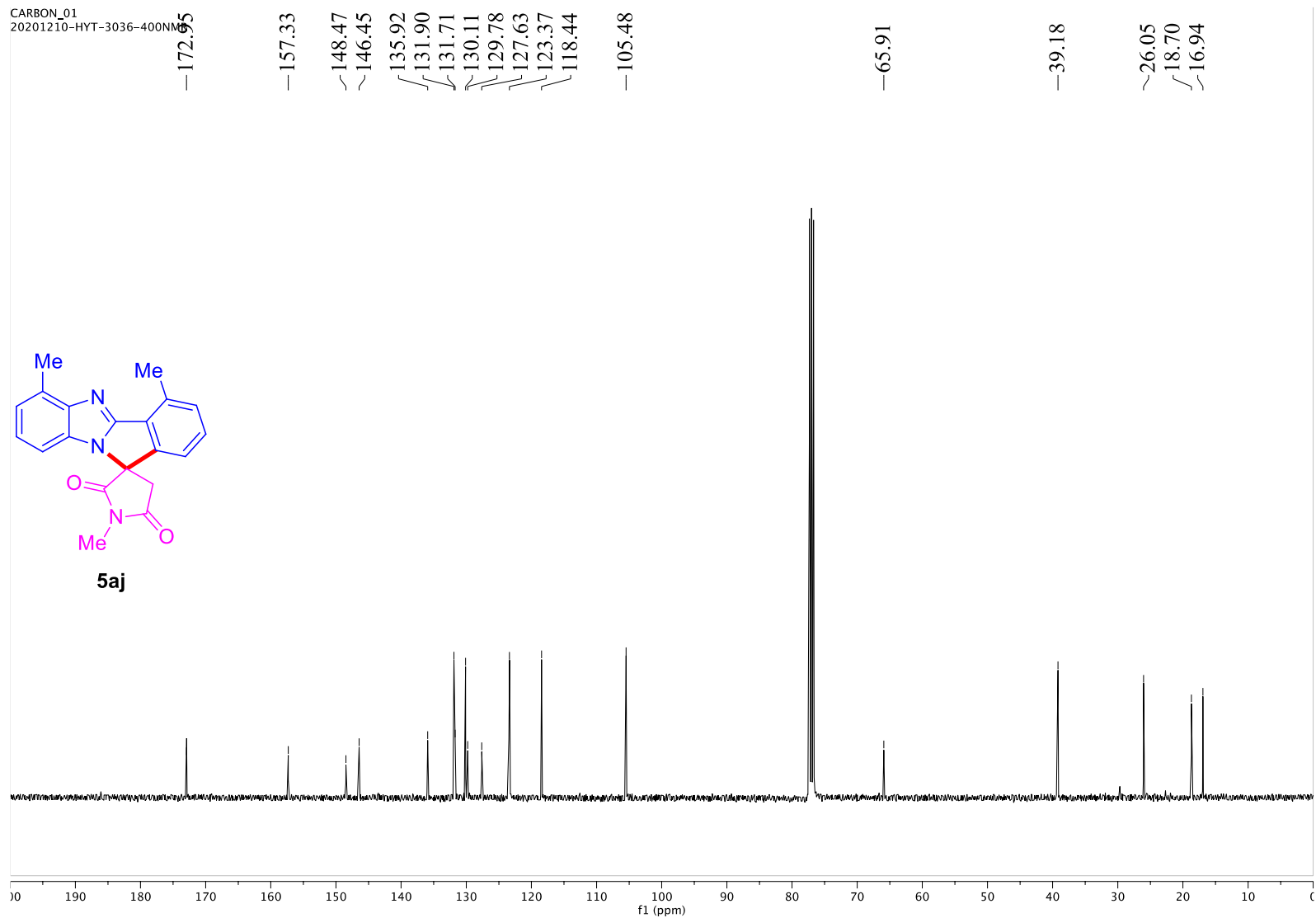


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3aj** and **5aj** in  $\text{CDCl}_3$ .

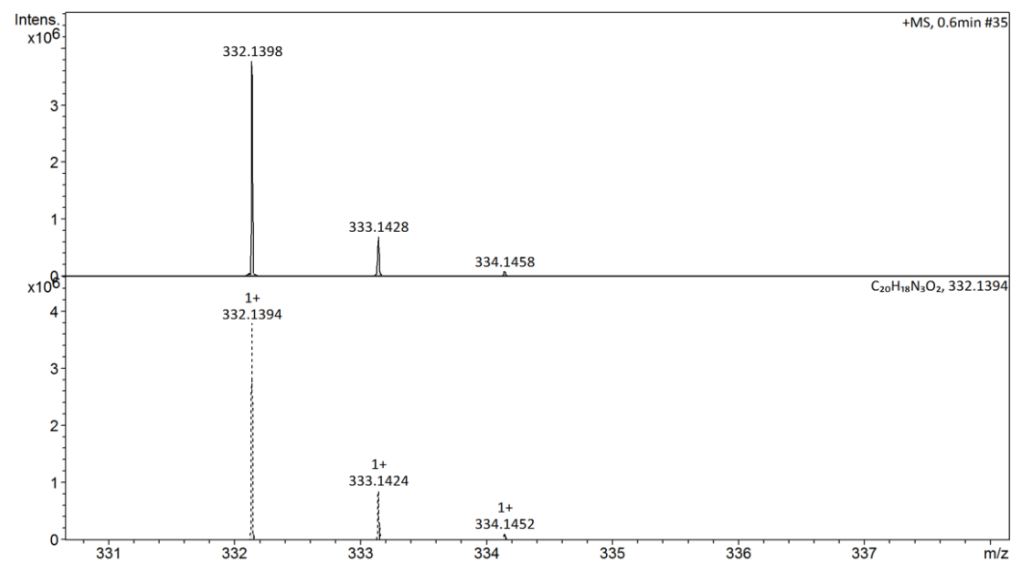
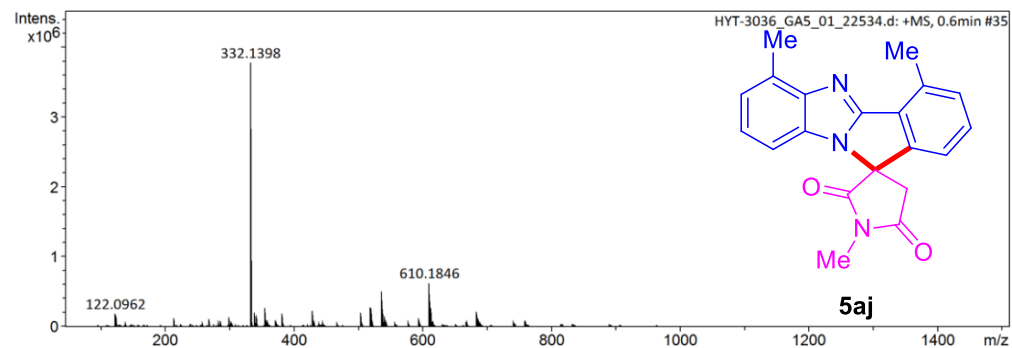


$^1\text{H}$  NMR spectrum (400 MHz) of compound **5aj** in  $\text{CDCl}_3$ .





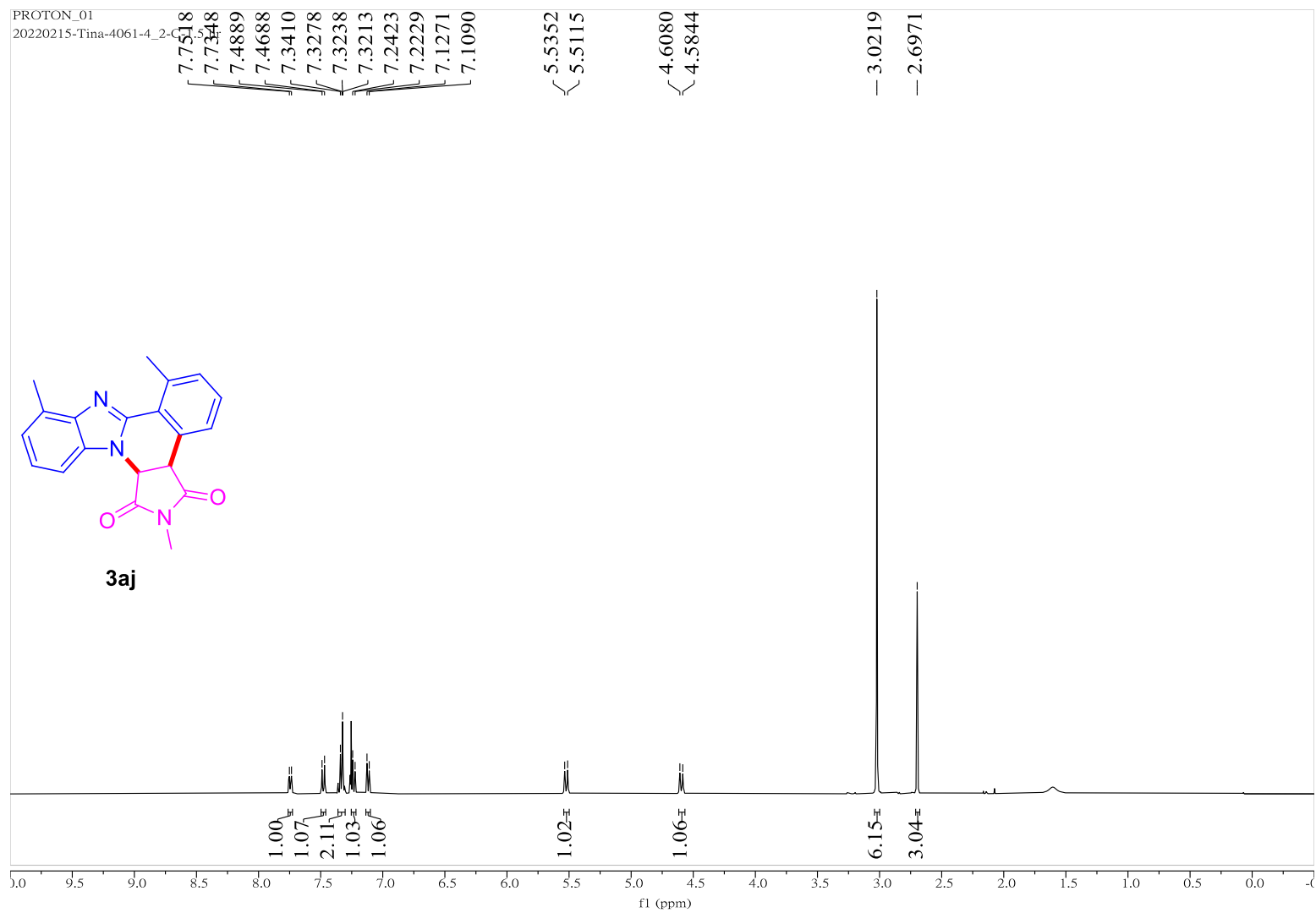
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5aj** in  $\text{CDCl}_3$ .



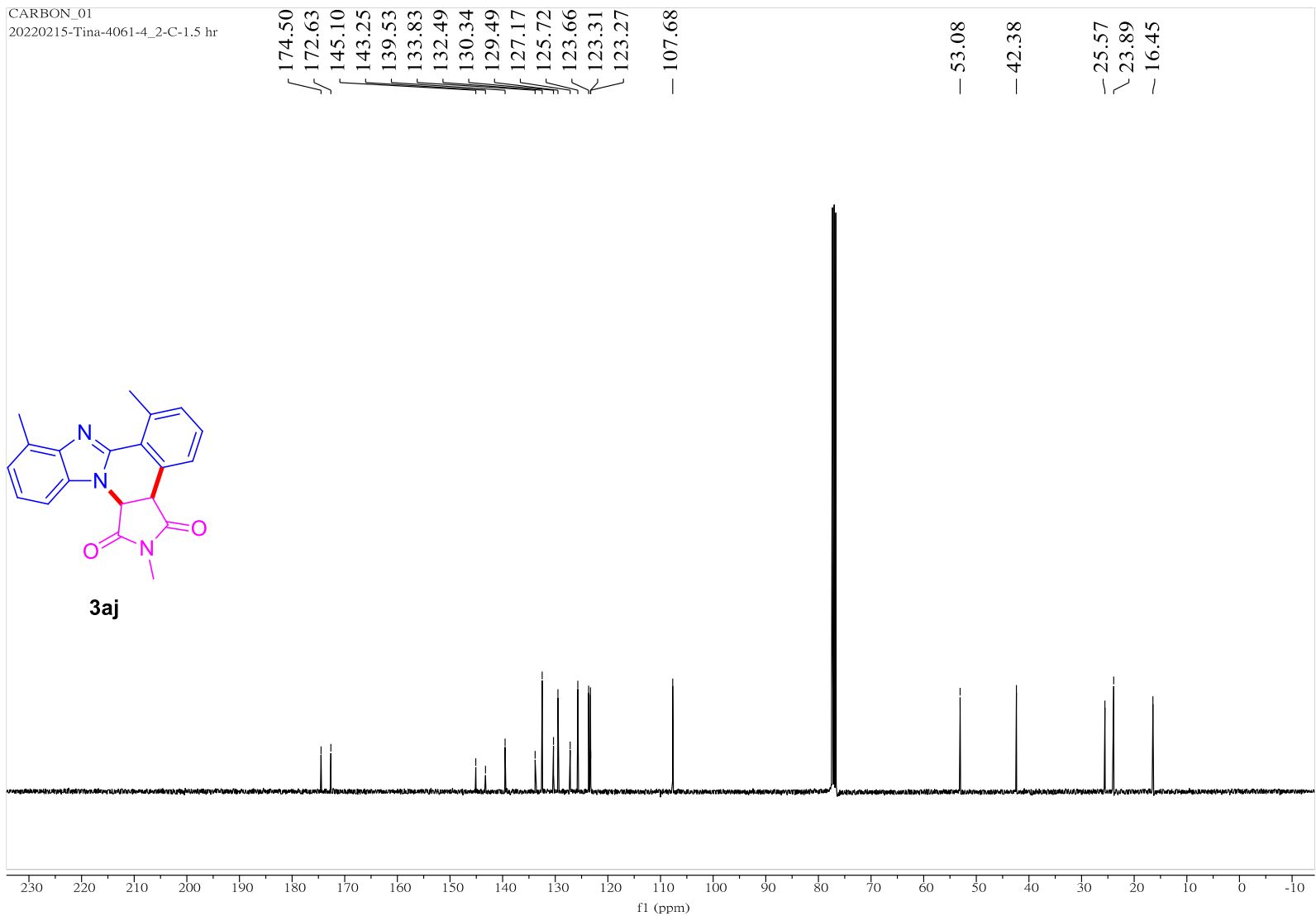
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
332.1399	1	C <sub>20</sub> H <sub>18</sub> N <sub>3</sub> O <sub>2</sub>	332.1394	-1.6	26.6	1	100.00	13.5	even	ok	M+H

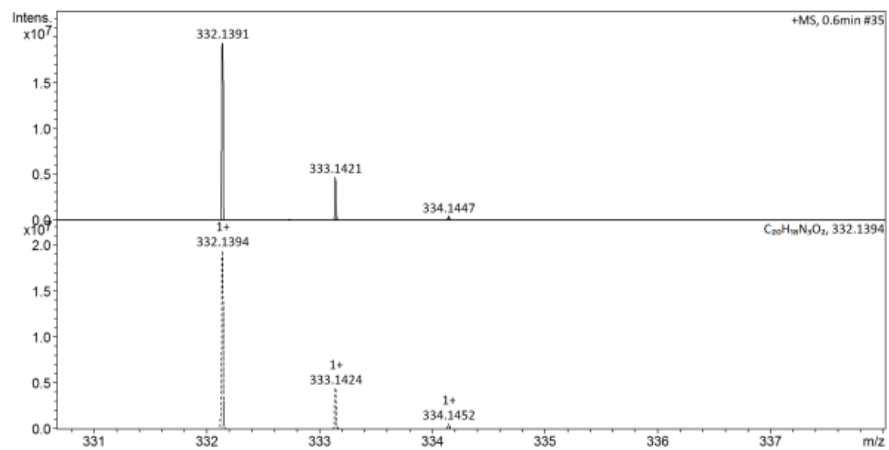
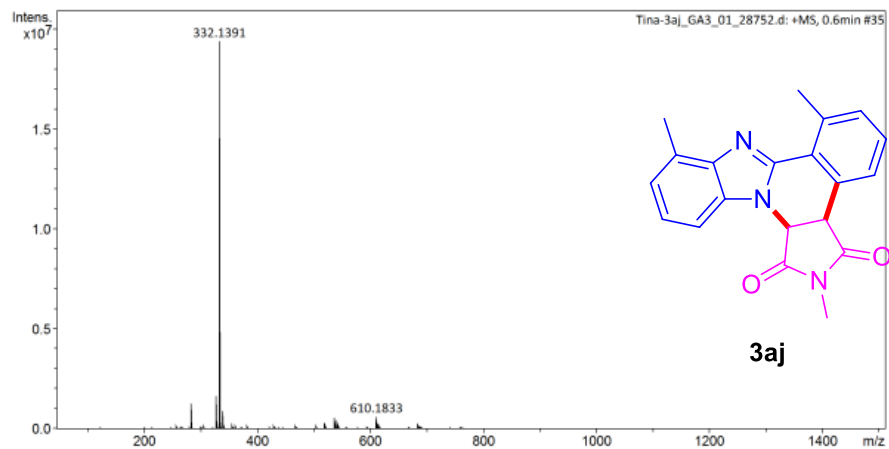
HRMS (ESI) of compound **5aj**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3aj** in  $\text{CDCl}_3$ .



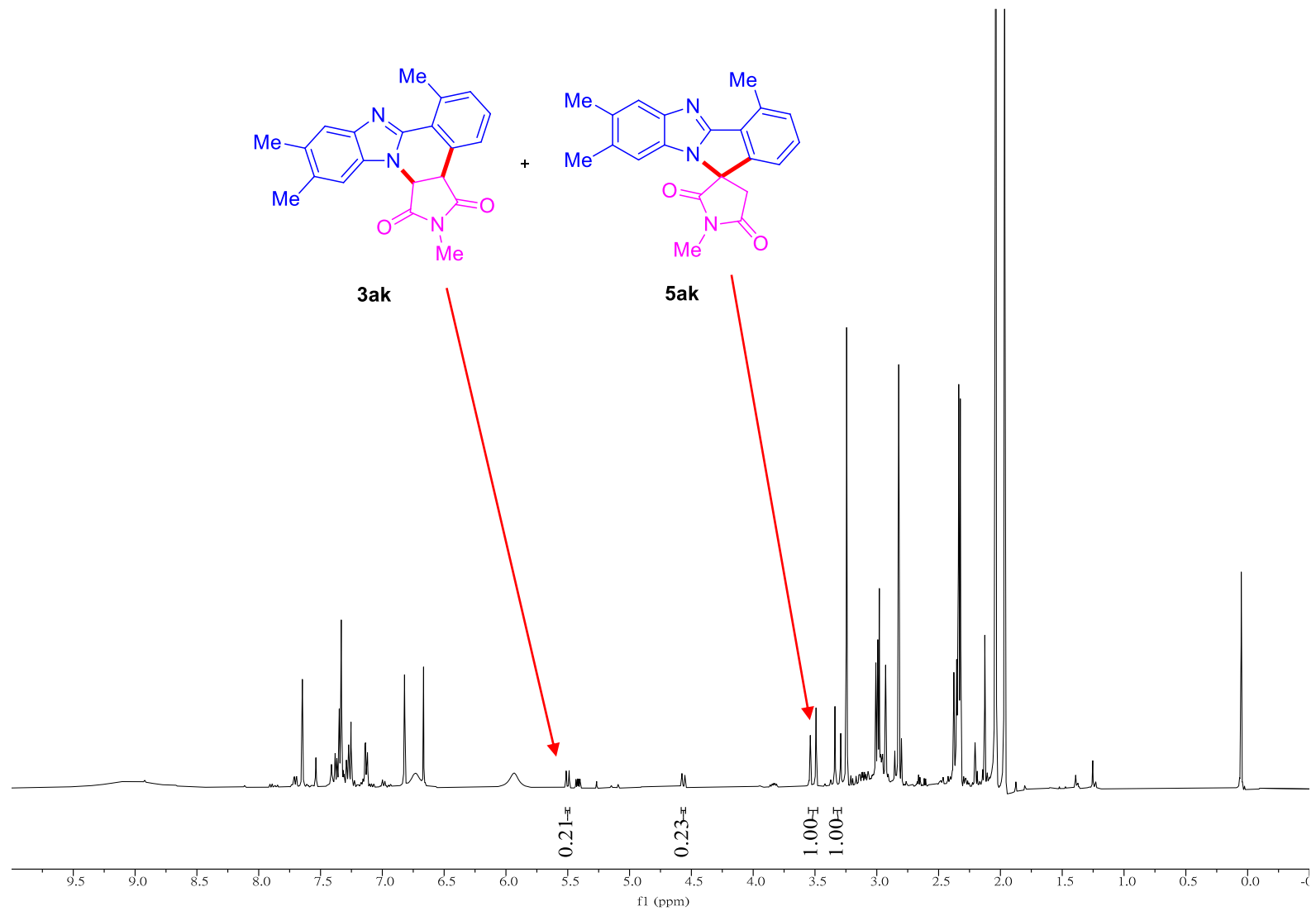
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3aj** in  $\text{CDCl}_3$ .



### Display Report

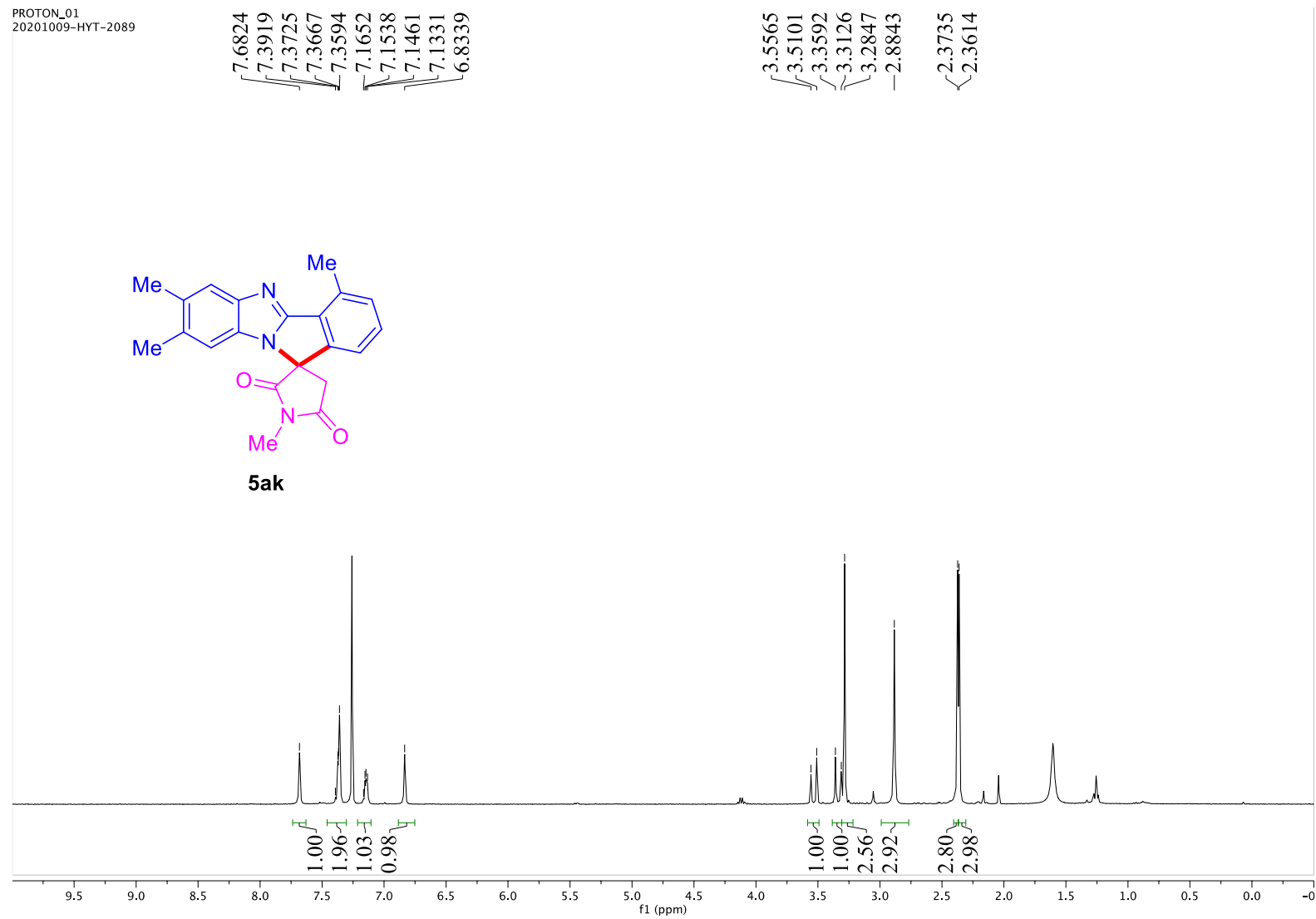
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
332.1391	1	C <sub>20</sub> H <sub>18</sub> N <sub>3</sub> O <sub>2</sub>	332.1394	0.8	9.1	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **3aj**.

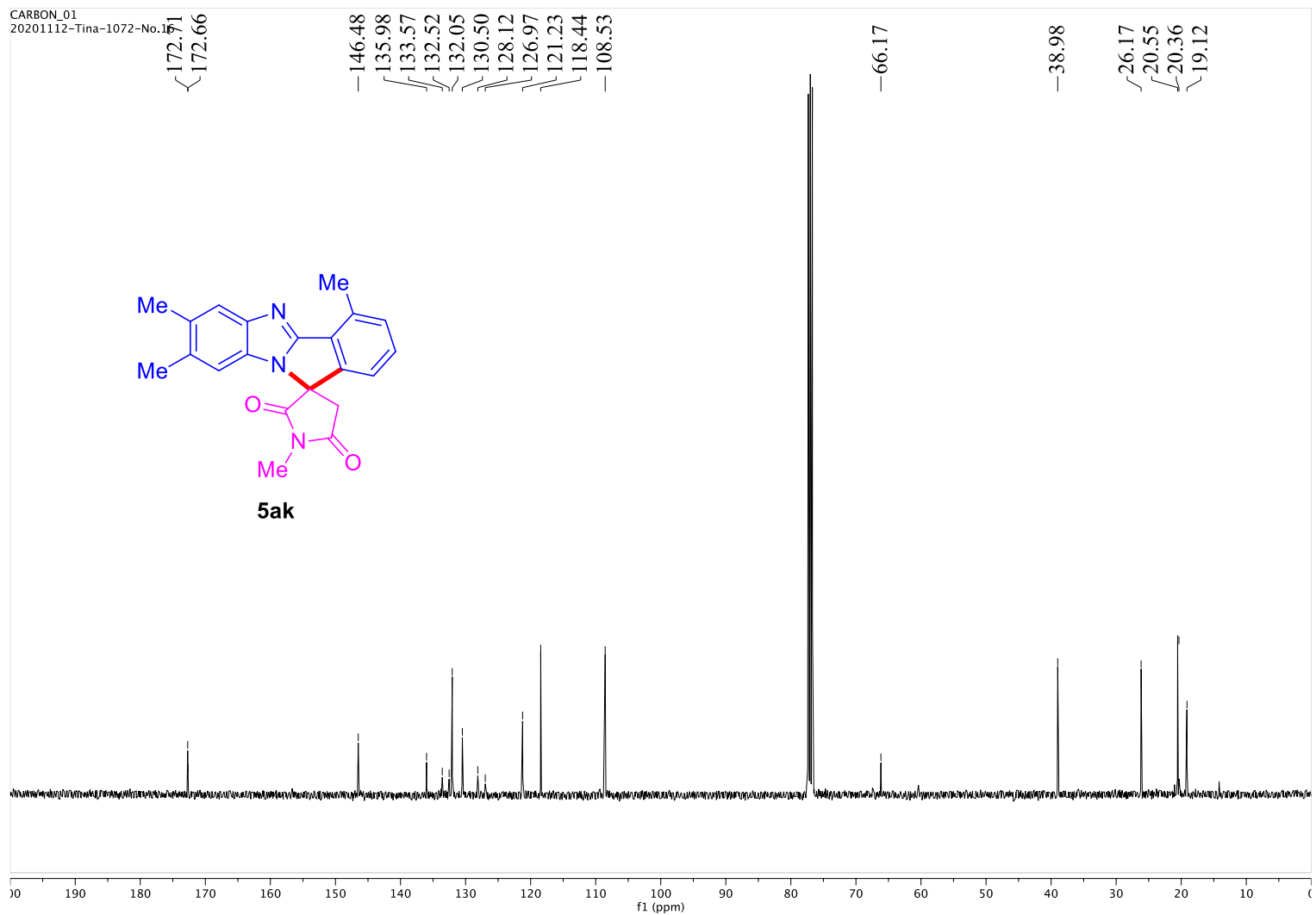


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3ak** and **5ak** in  $\text{CDCl}_3$ .

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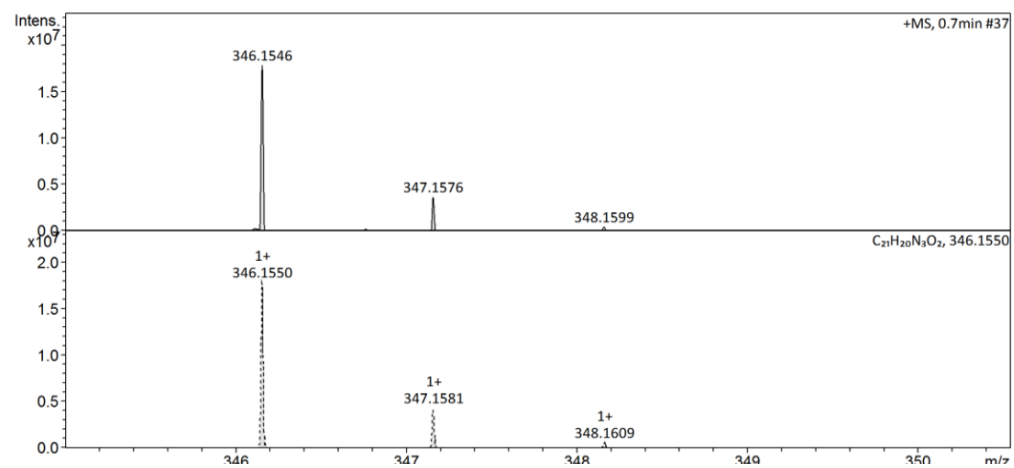
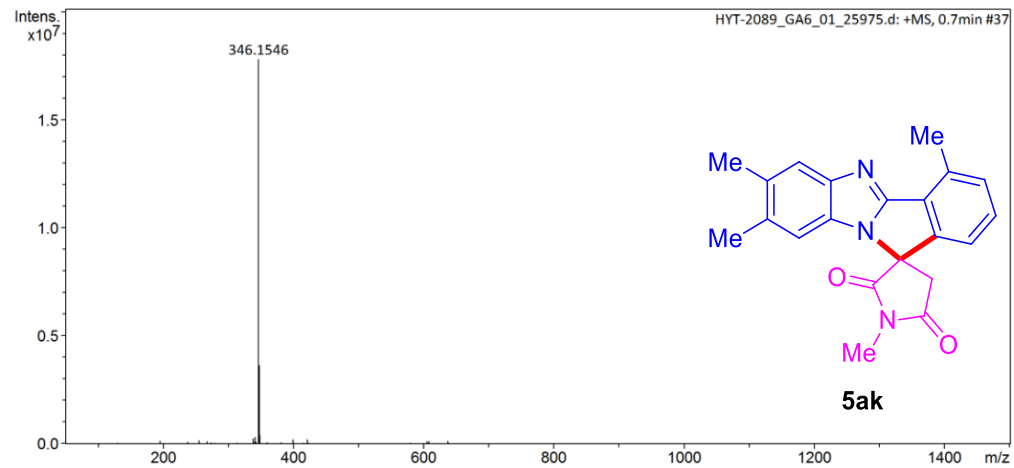


$^1\text{H}$  NMR spectrum (400 MHz) of compound **5ak** in  $\text{CDCl}_3$ .



$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5ak** in  $\text{CDCl}_3$ .

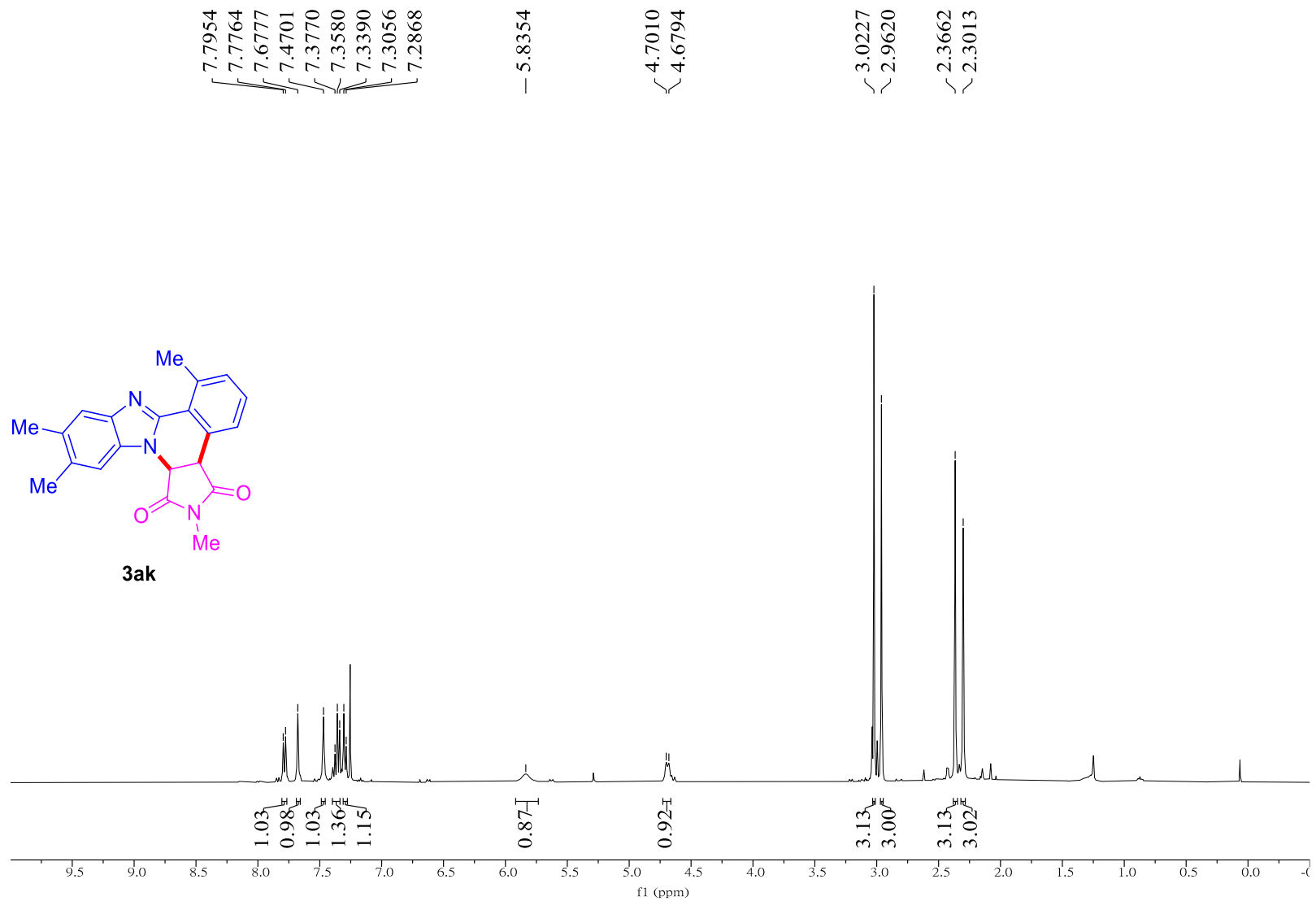




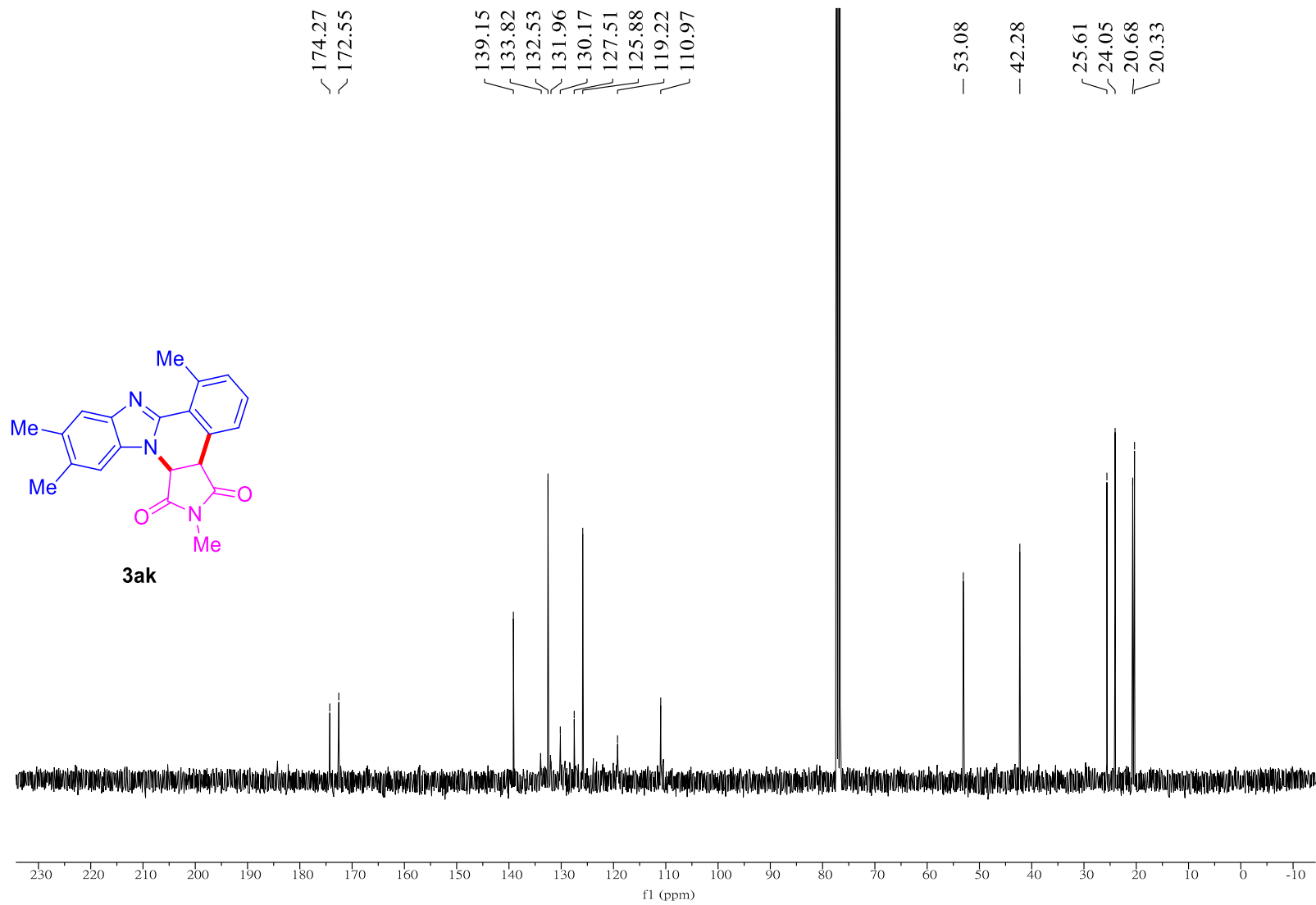
### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
346.1546	1	C <sub>21</sub> H <sub>20</sub> N <sub>3</sub> O <sub>2</sub>	346.1550	-1.2	22.5	1	100.00	13.5	even	ok	M+H

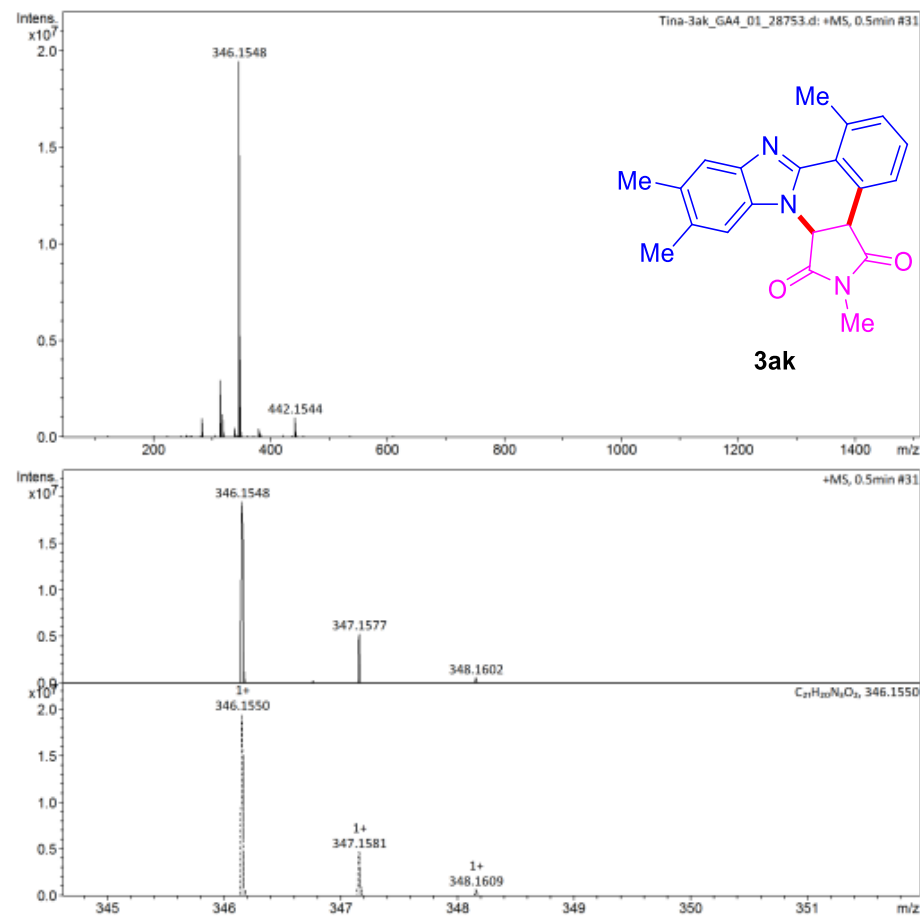
HRMS (ESI) of compound **5ak**.



**<sup>1</sup>H NMR spectrum (400 MHz) of compound **3ak** in CDCl<sub>3</sub>.**



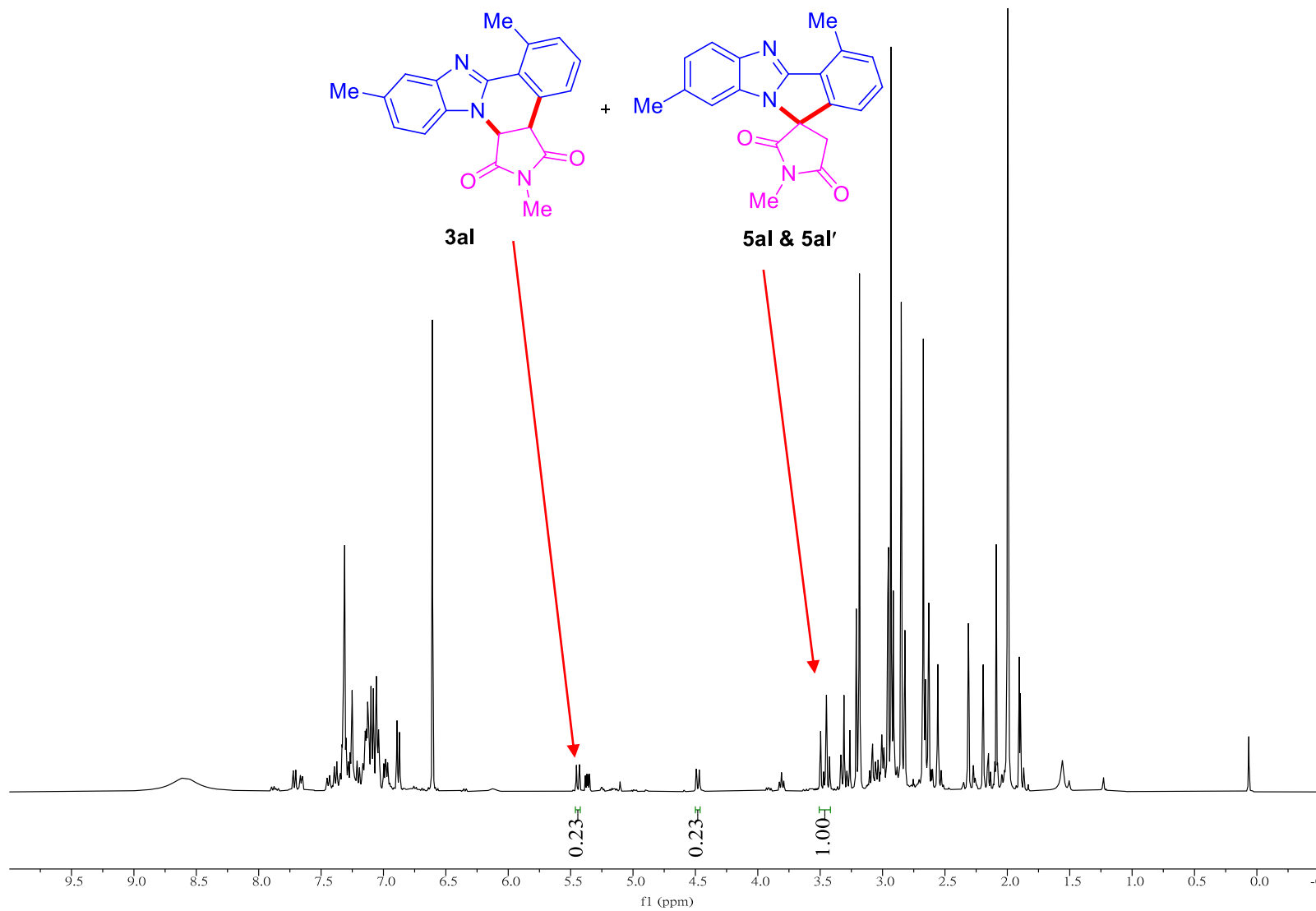
<sup>13</sup>C NMR spectrum (100 MHz) of compound **3ak** in CDCl<sub>3</sub>.



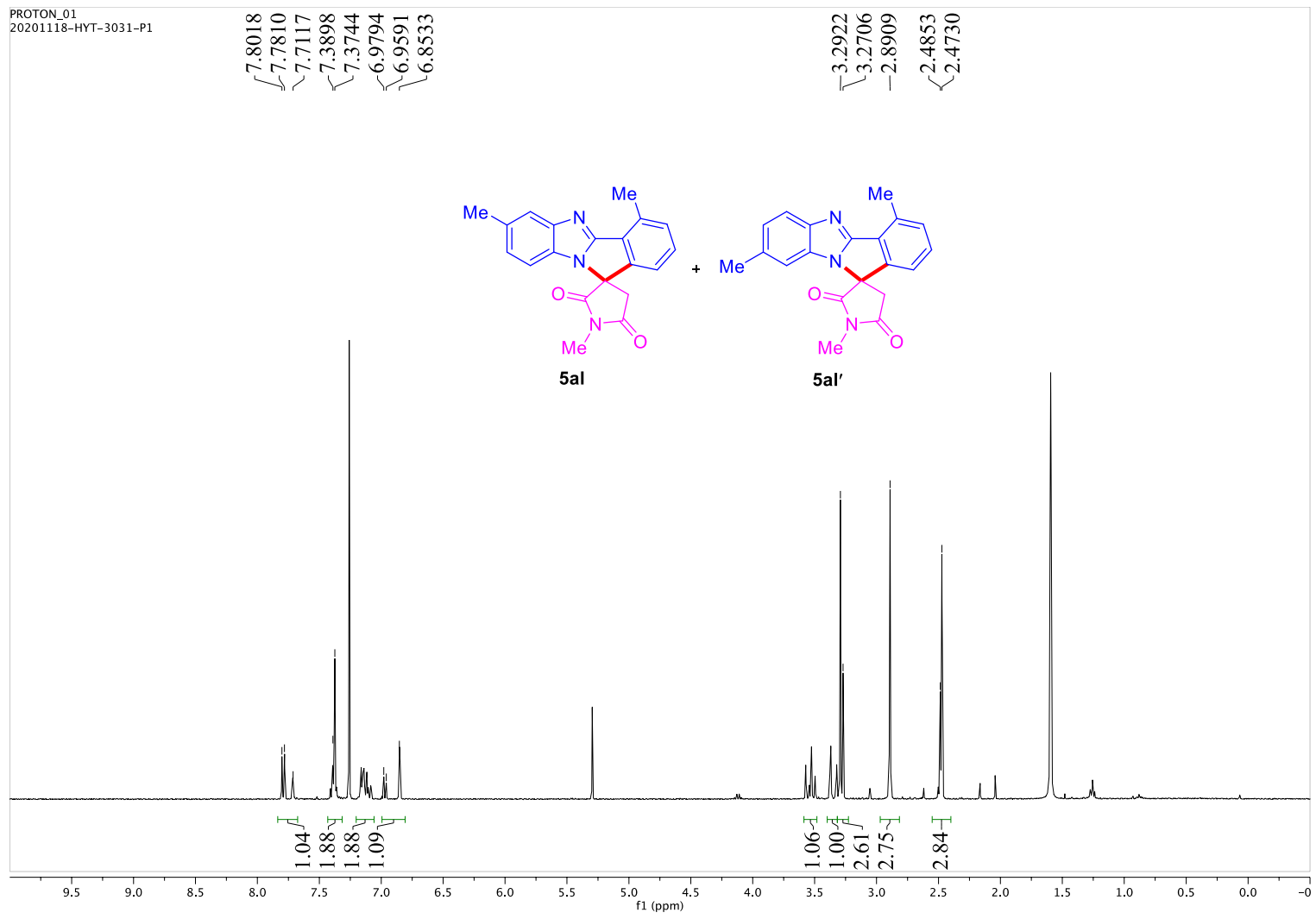
### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
346.1548	1	C <sub>21</sub> H <sub>20</sub> N <sub>3</sub> O <sub>2</sub>	346.1550	0.5	16.8	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **3ak**.

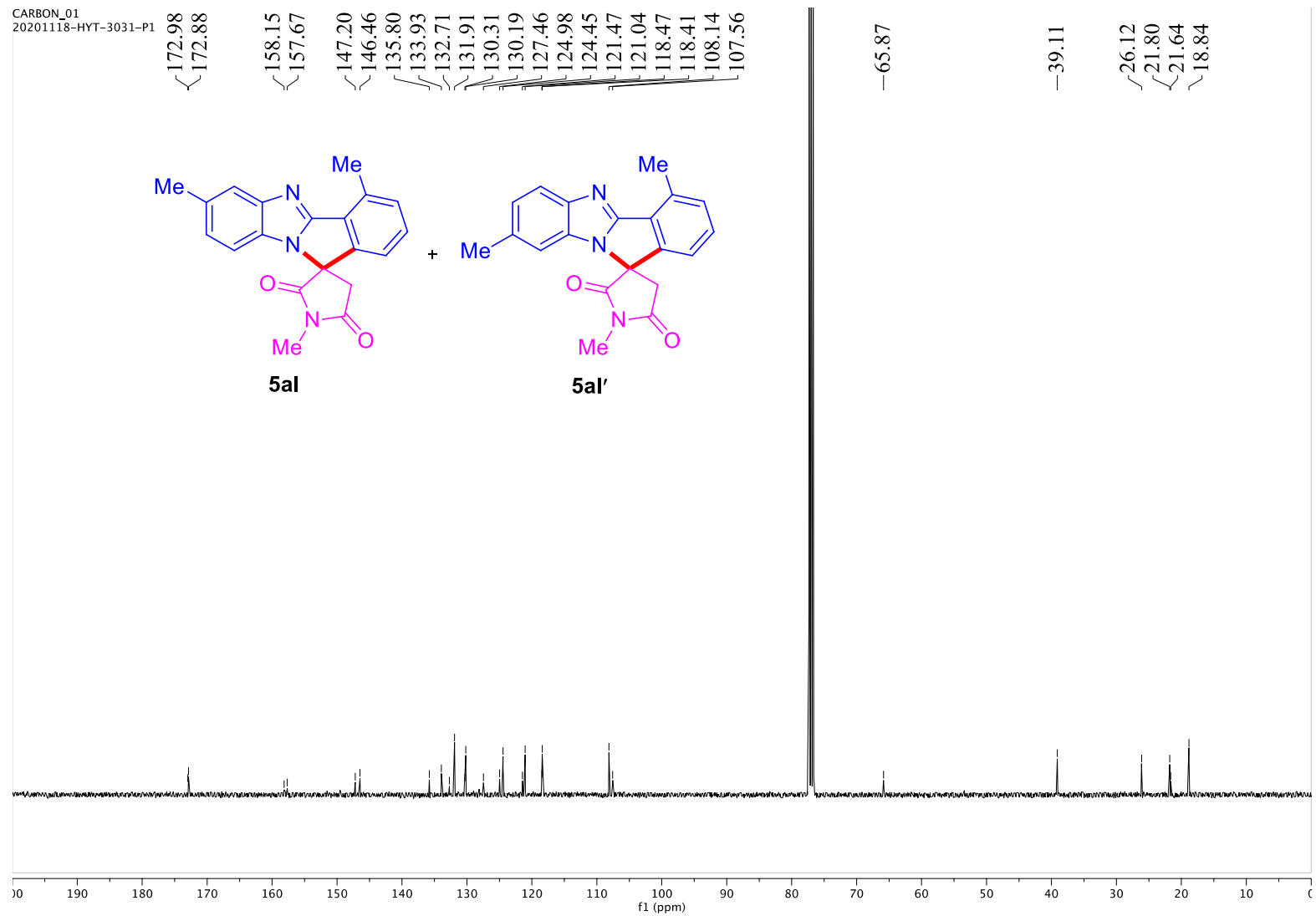


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3al** and **5al** and **5al'** in  $\text{CDCl}_3$ .

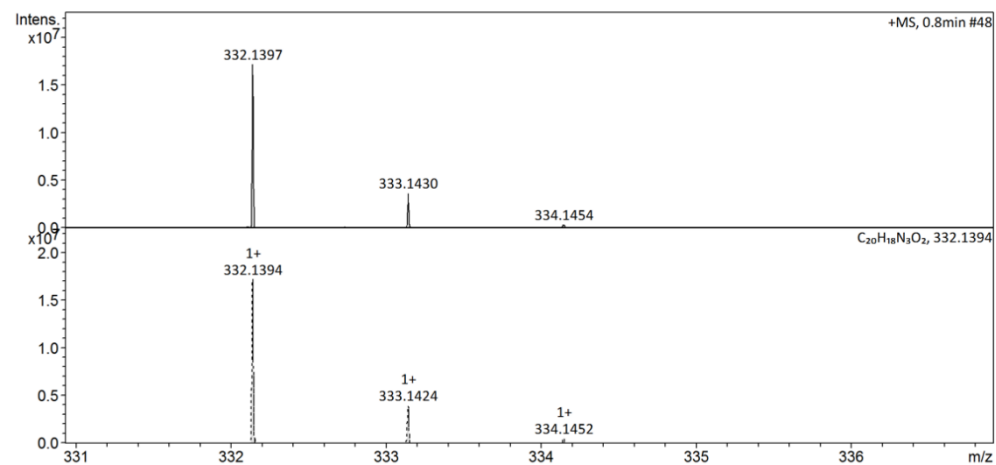
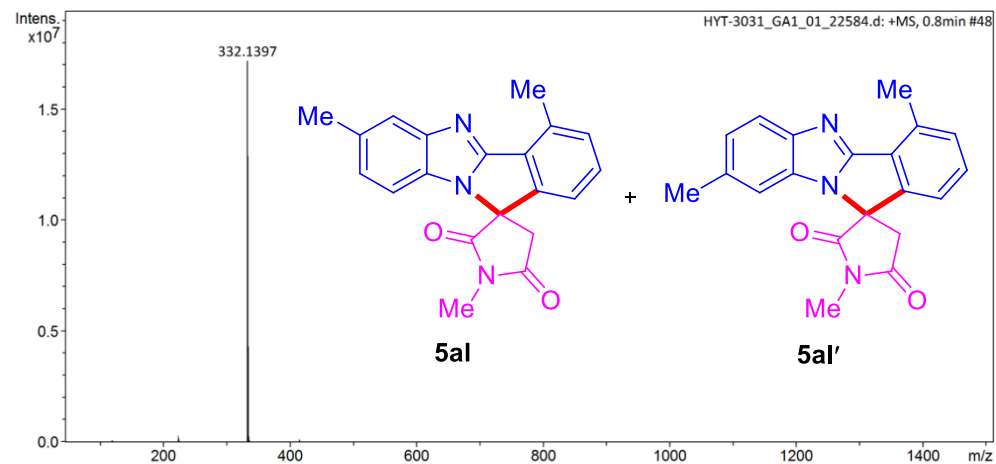


$^1\text{H}$  NMR spectrum (400 MHz) of compound **5al** and **5al'** in  $\text{CDCl}_3$ .

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$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5al** and **5al'** in  $\text{CDCl}_3$ .

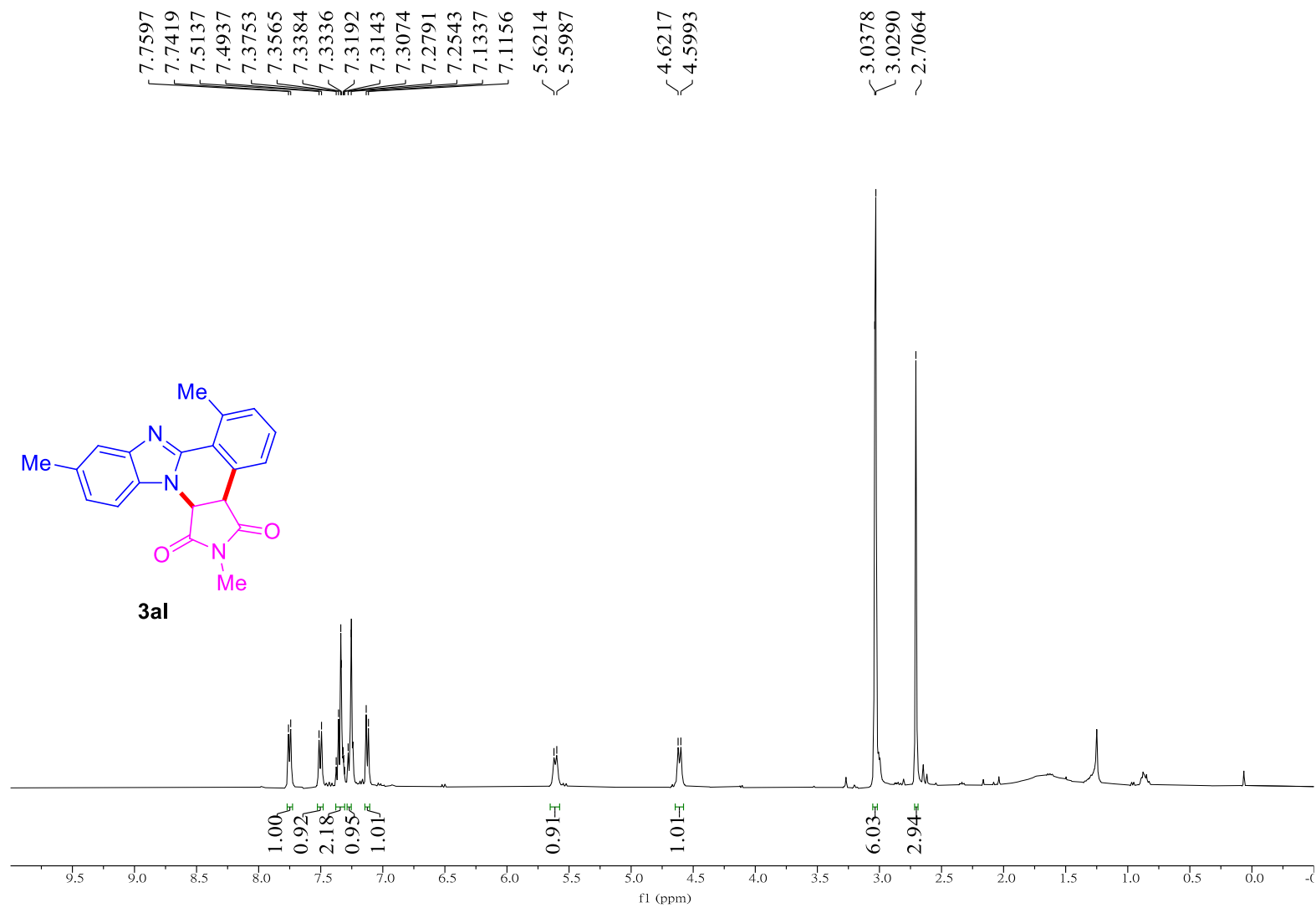


## Display Report

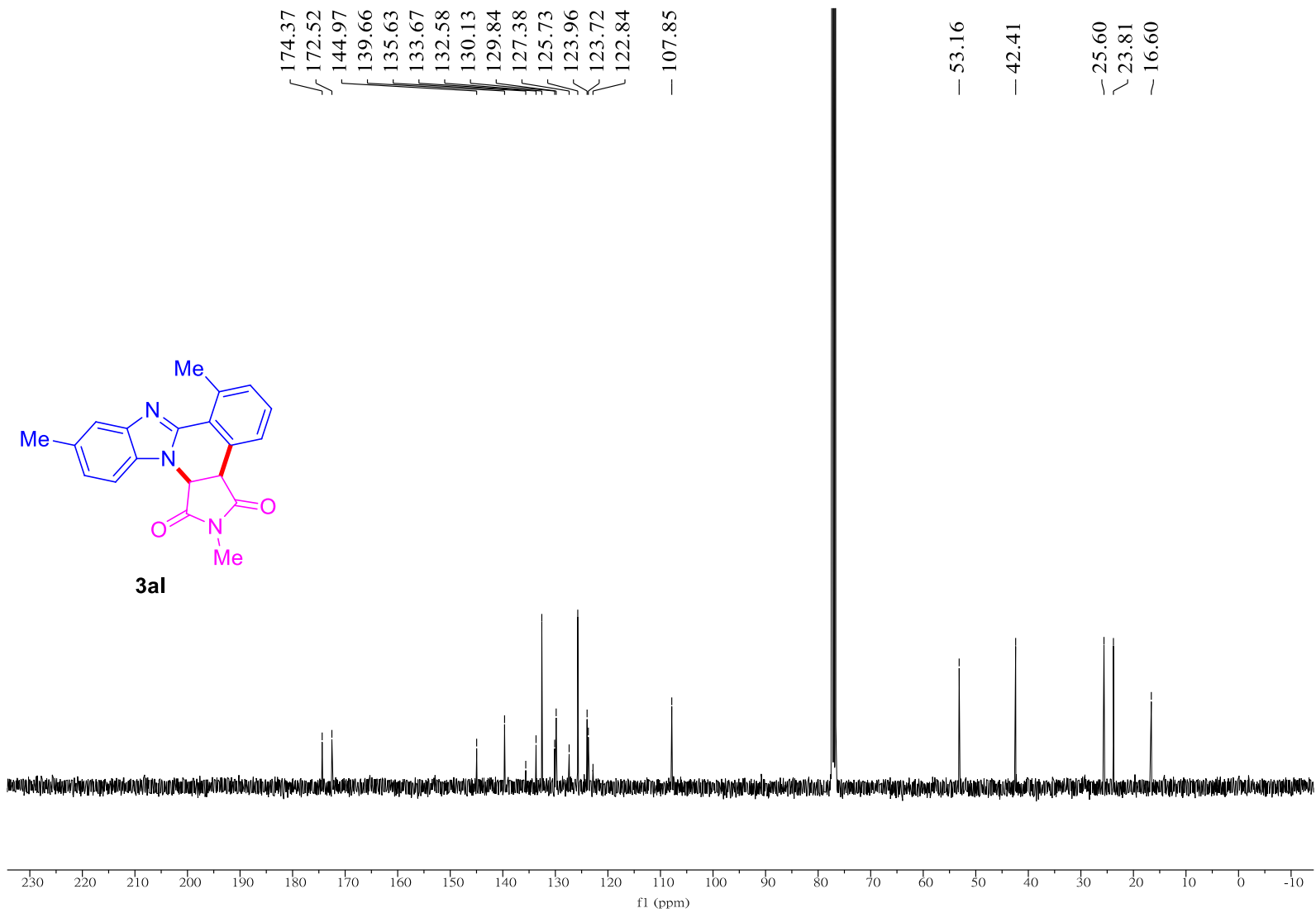
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
332.1397	1	C <sub>20</sub> H <sub>18</sub> N <sub>3</sub> O <sub>2</sub>	332.1394	-1.0	11.0	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **5al** and **5al'**.

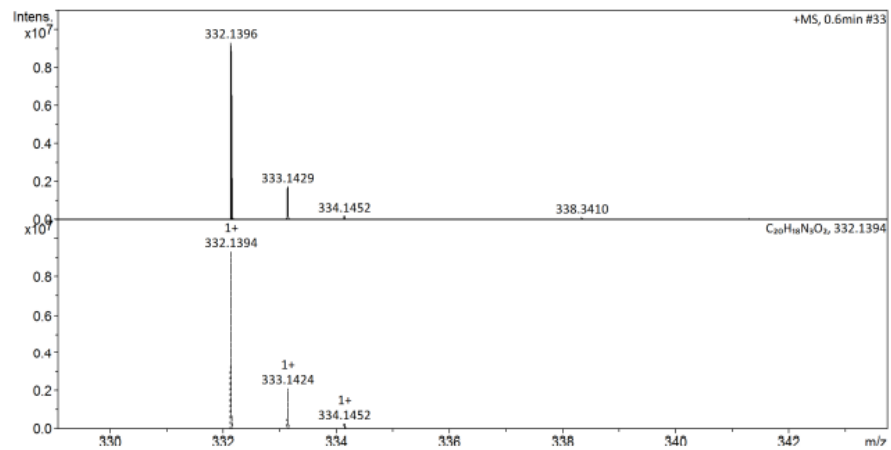
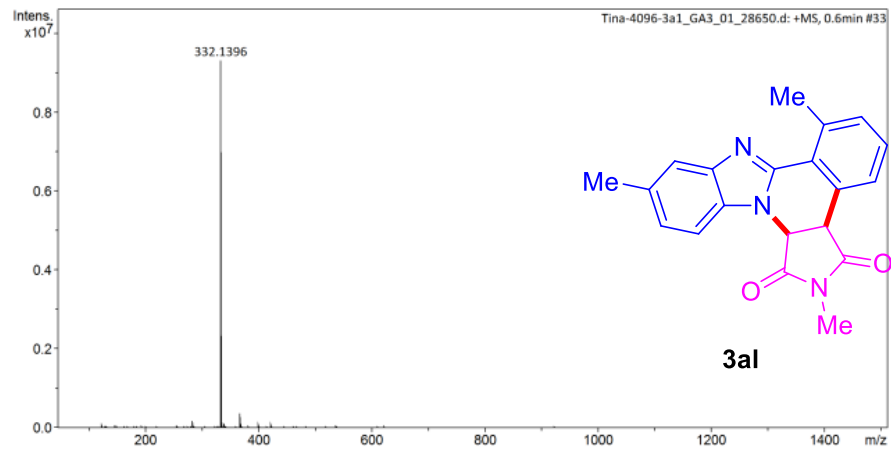




$^1\text{H}$  NMR spectrum (400 MHz) of compound **3al** in  $\text{CDCl}_3$ .



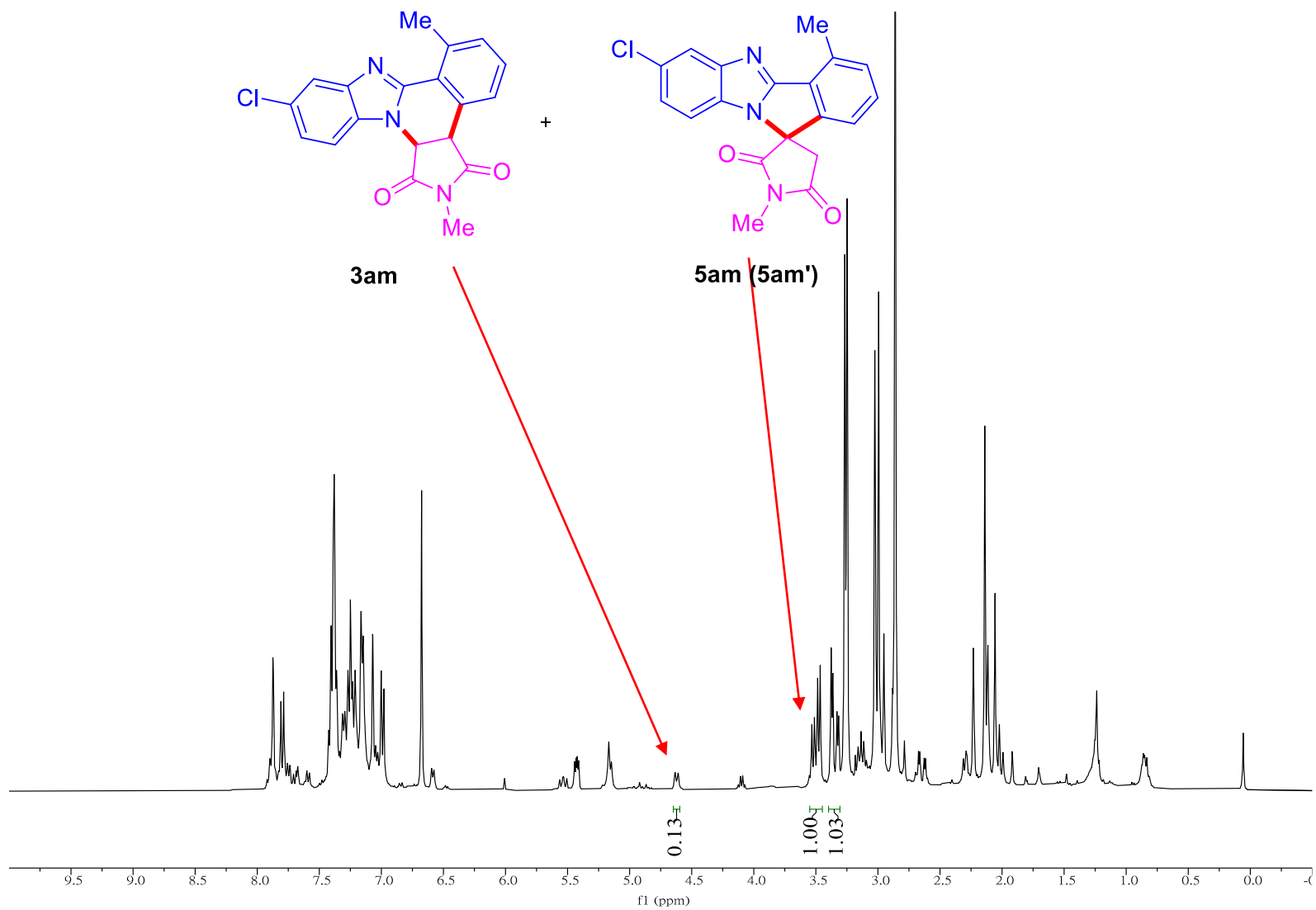
<sup>13</sup>C NMR spectrum (100 MHz) of compound **3al** in CDCl<sub>3</sub>.



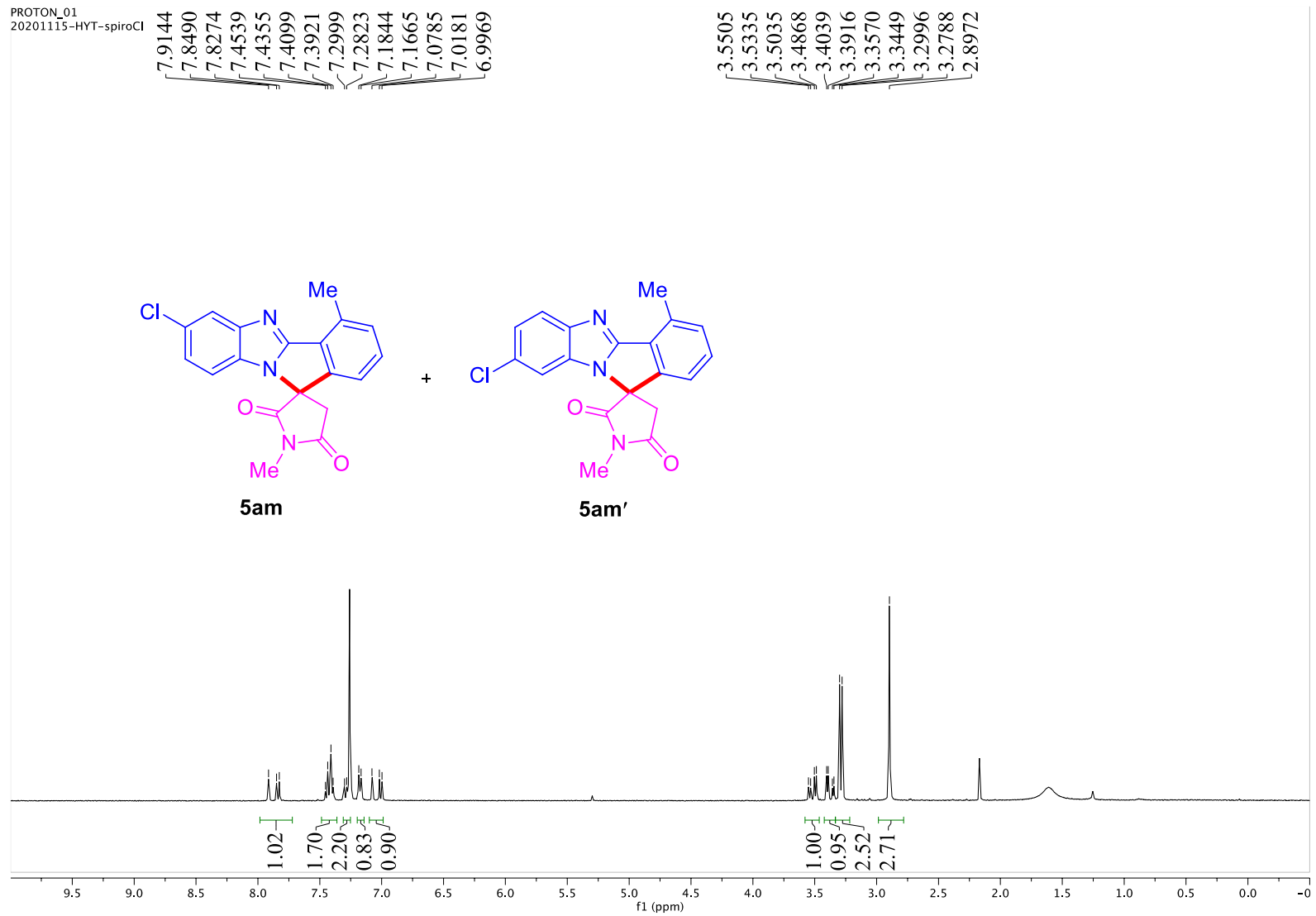
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
332.1396	1	C <sub>20</sub> H <sub>18</sub> N <sub>3</sub> O <sub>2</sub>	332.1394	-0.7	26.3	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **3a1**.

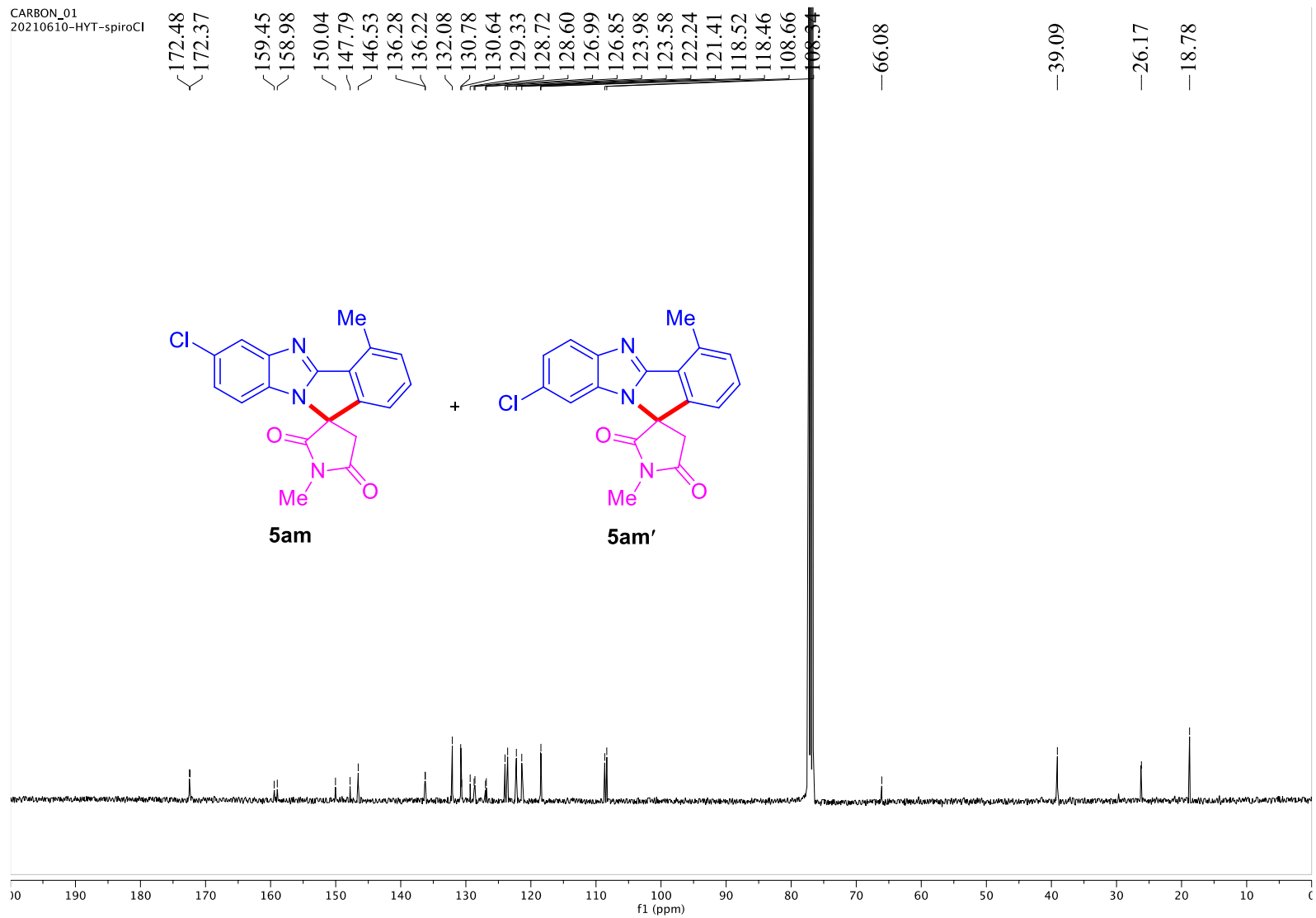


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3am** and **5am** in  $\text{CDCl}_3$ .

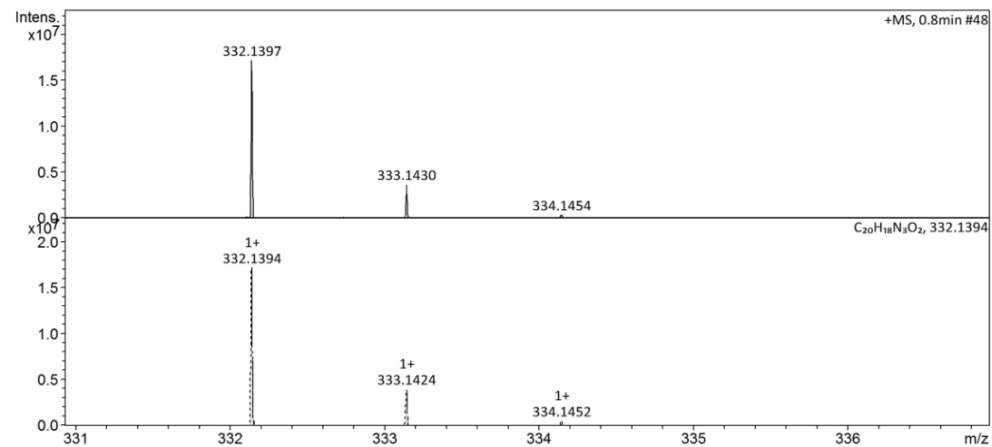
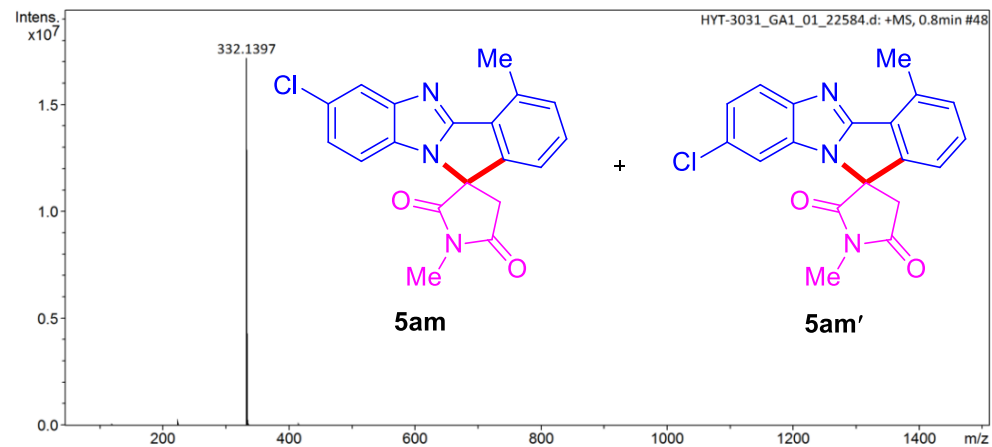


$^1\text{H}$  NMR spectrum (400 MHz) of compound **5am** and **5am'** in  $\text{CDCl}_3$ .

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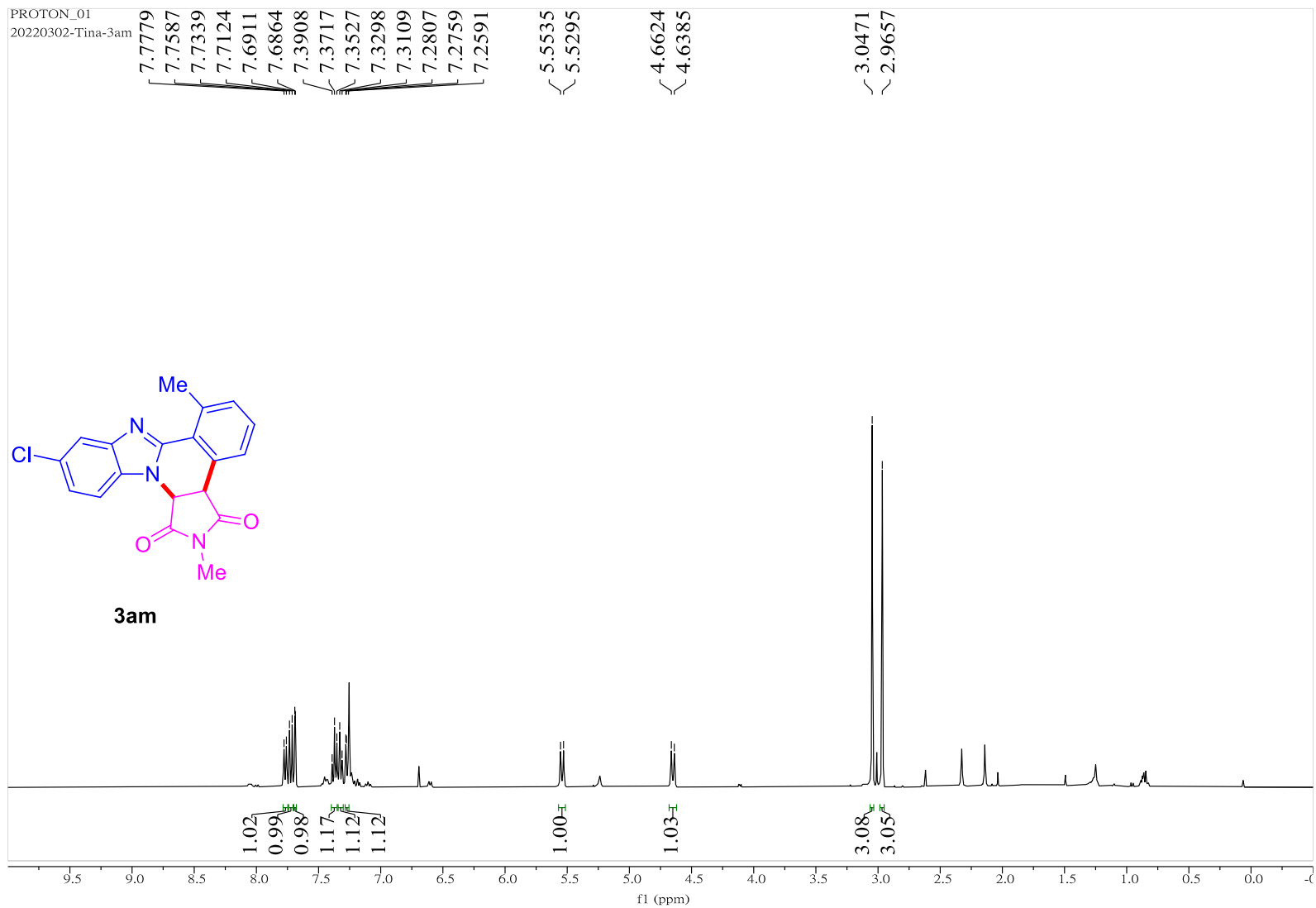
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5am** and **5am'** in  $\text{CDCl}_3$ .



## Display Report

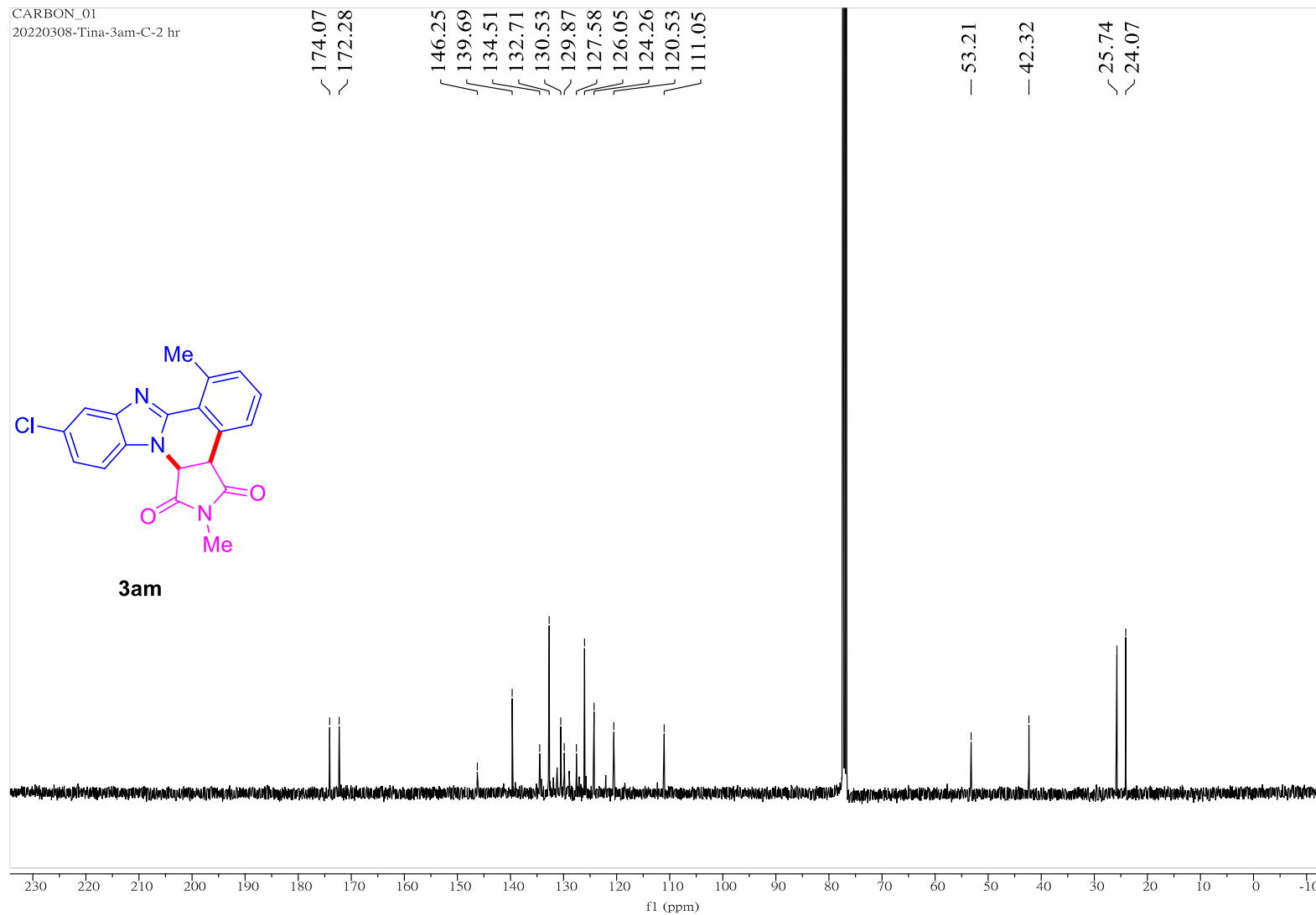
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
352.0847	1	C <sub>19</sub> H <sub>15</sub> ClN <sub>3</sub> O <sub>2</sub>	352.0847	-0.0	30.6	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **5am** and **5am'**.

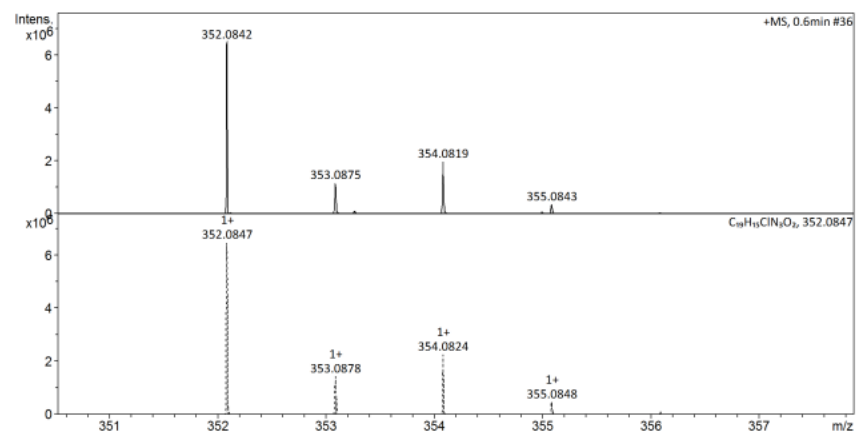
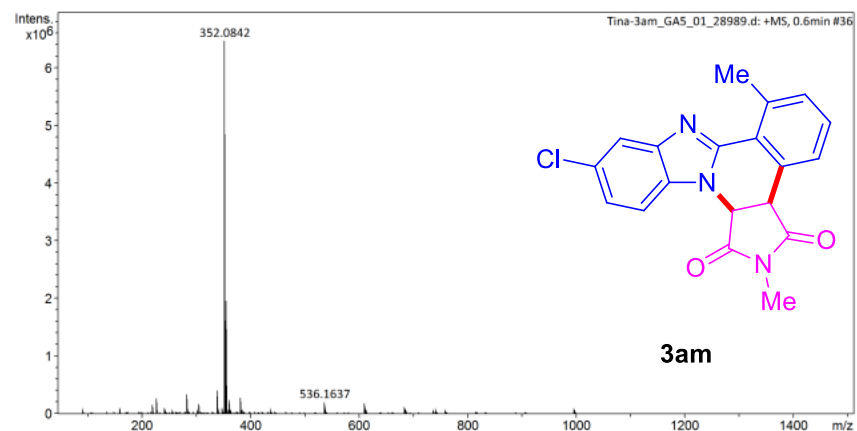


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3am** in  $\text{CDCl}_3$ .





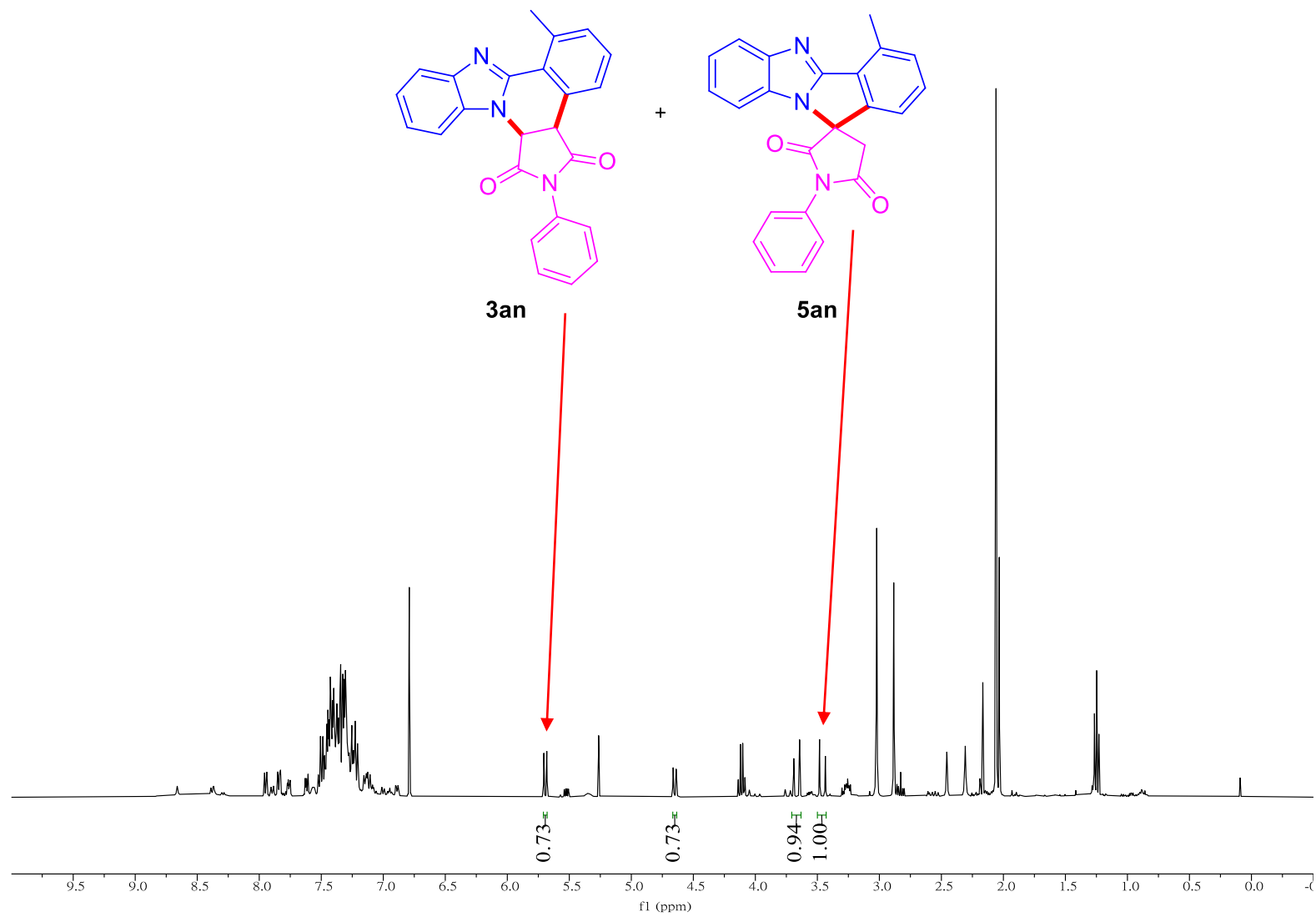
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3am** in  $\text{CDCl}_3$ .



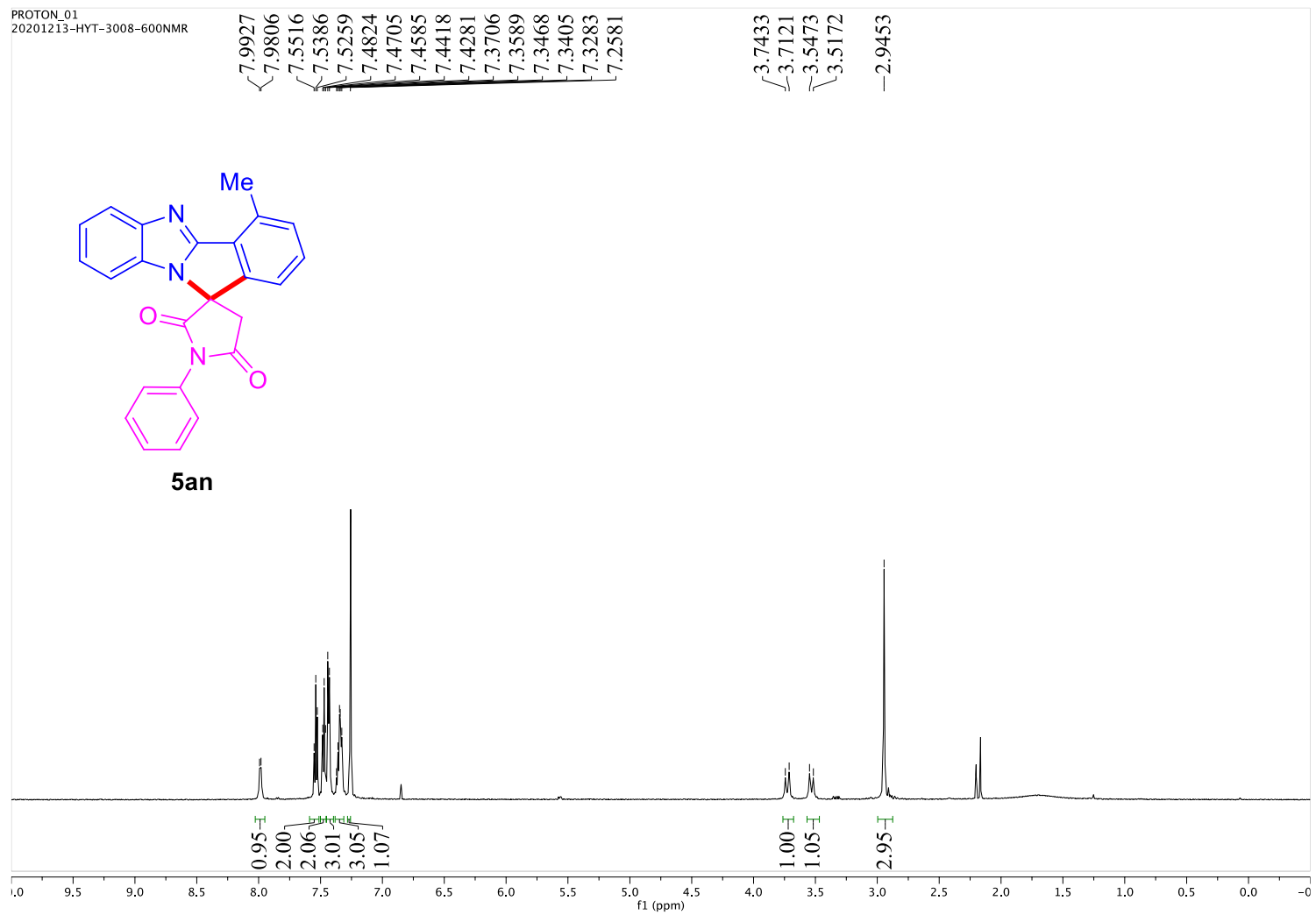
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
352.0842	1	C <sub>19</sub> H <sub>15</sub> ClN <sub>3</sub> O <sub>2</sub>	352.0847	1.5	27.5	1	100.00	13.5	even	ok	M+H

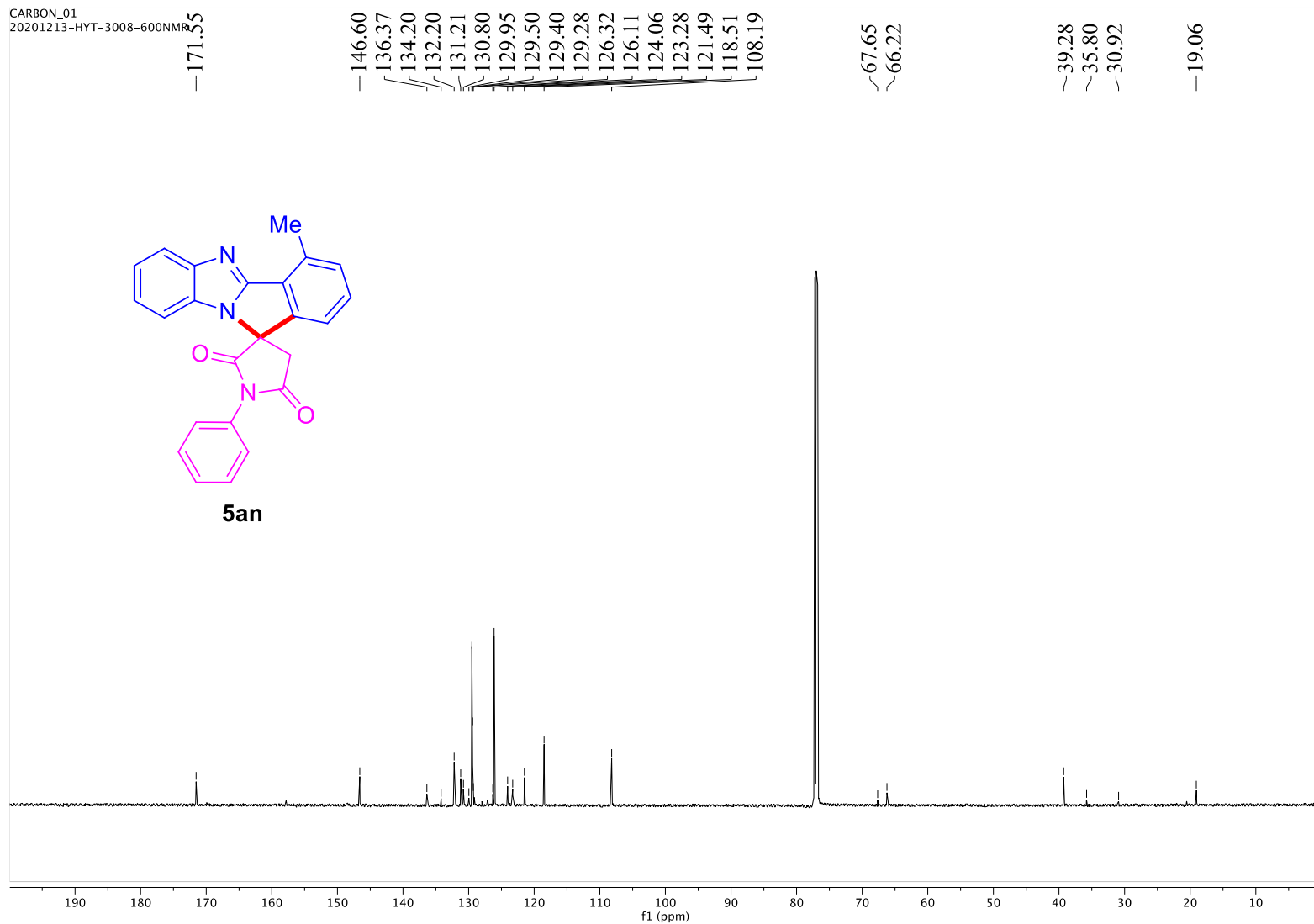
HRMS (ESI) of compound **3am**.



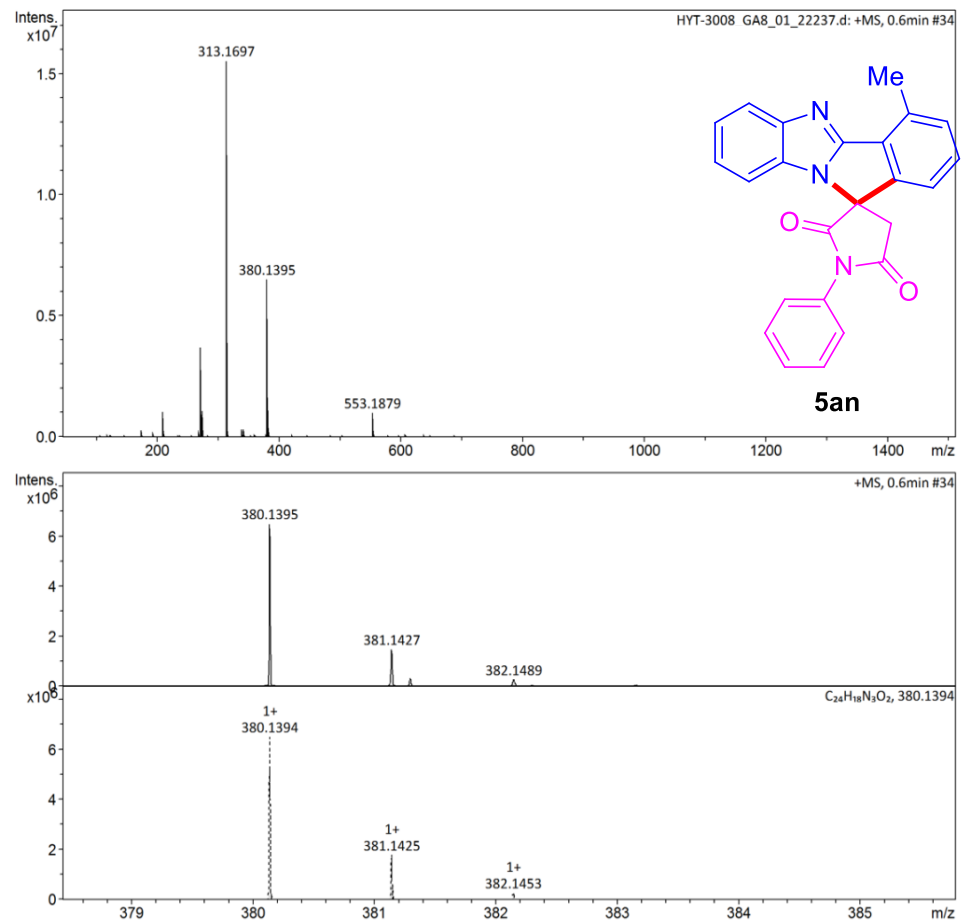
$^1\text{H}$  NMR spectrum (400 MHz) of compound **3an** and **5an** in  $\text{CDCl}_3$ .



$^1\text{H}$  NMR spectrum (600 MHz) of compound **5an** in  $\text{CDCl}_3$ .



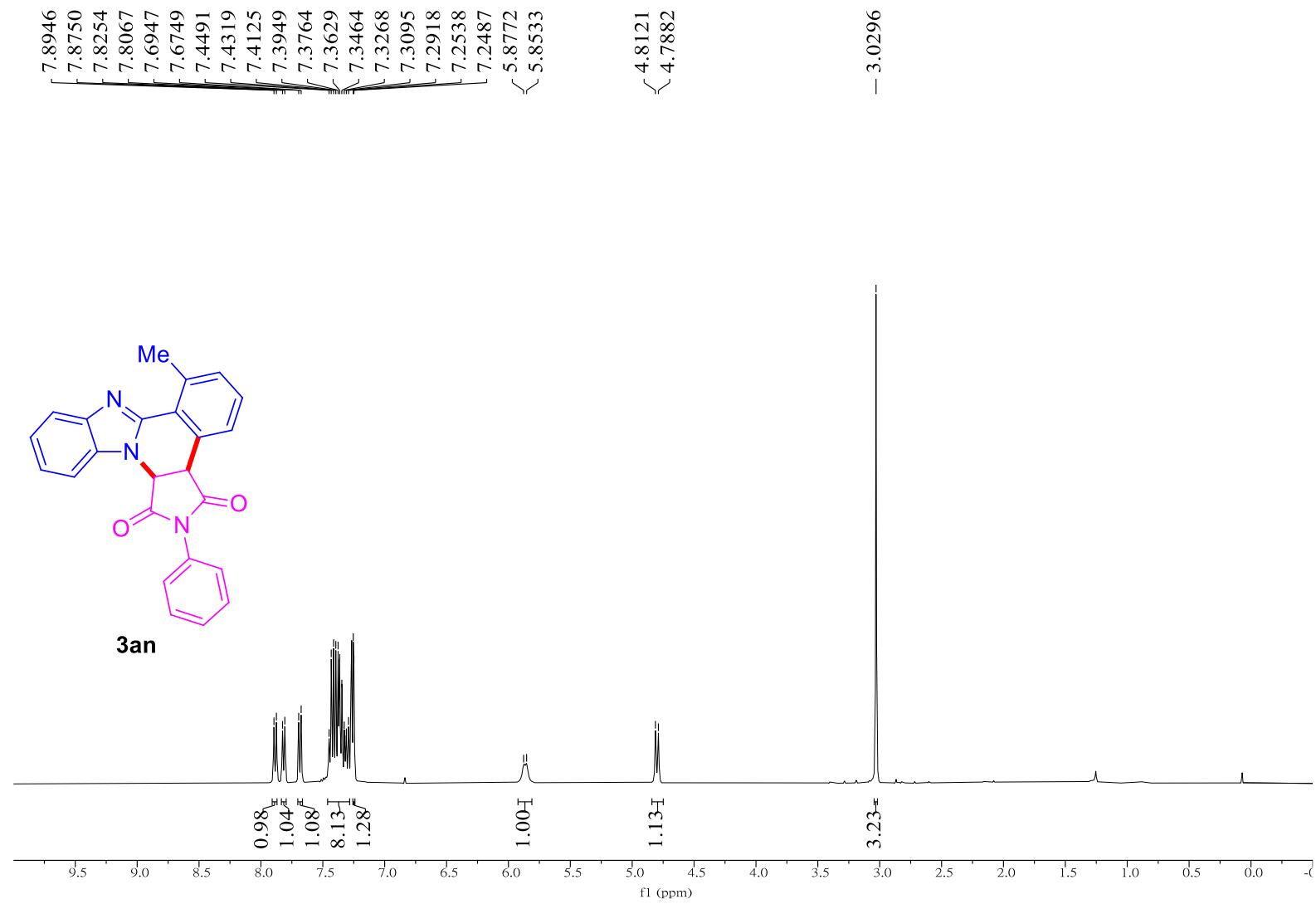
$^{13}\text{C}$  NMR spectrum (150 MHz) of compound **5an** in  $\text{CDCl}_3$ .



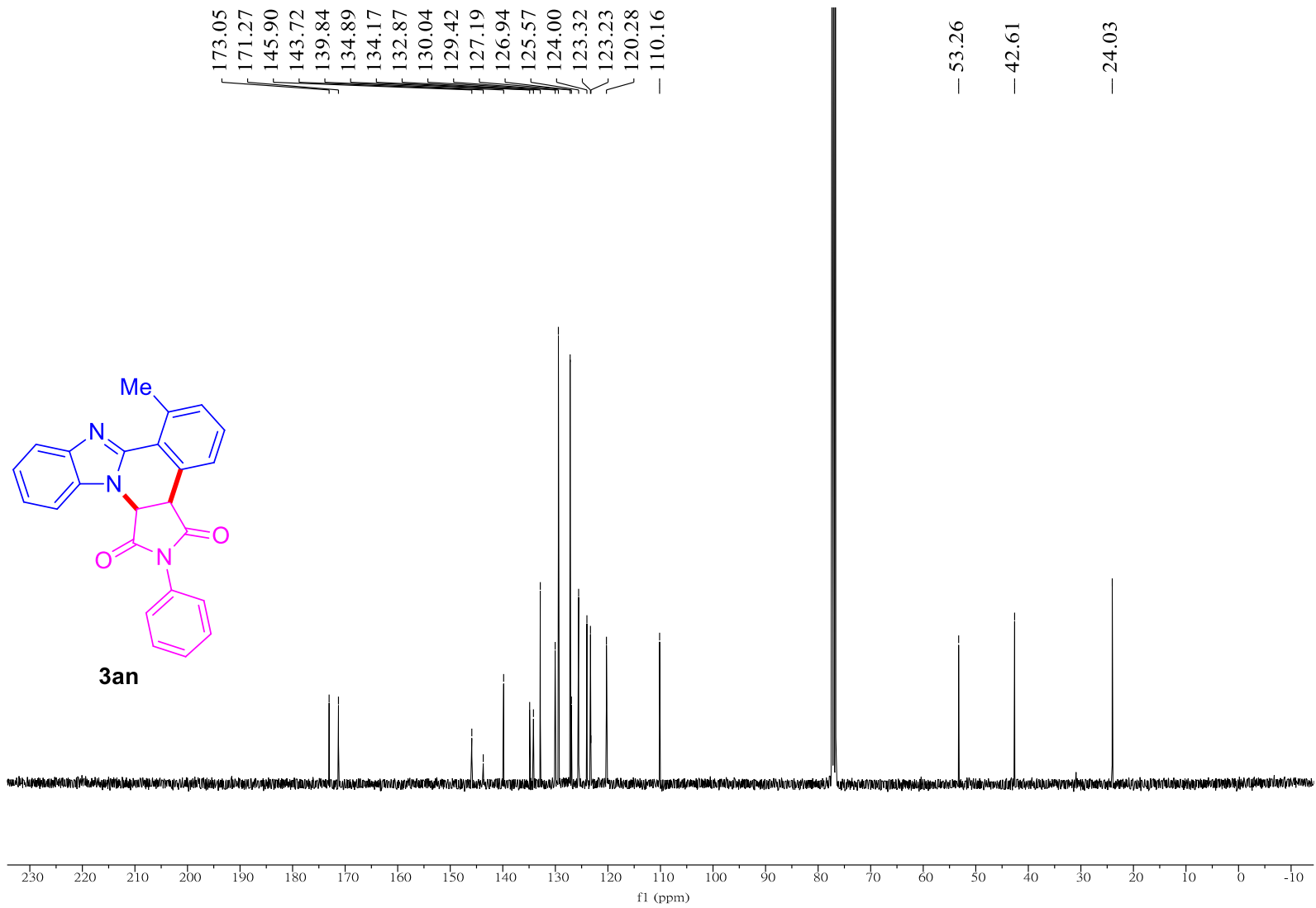
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
380.1395	1	C <sub>24</sub> H <sub>18</sub> N <sub>3</sub> O <sub>2</sub>	380.1394	-0.3	23.8	1	100.00	17.5	even	ok	M+H

HRMS (ESI) of compound **5an**.

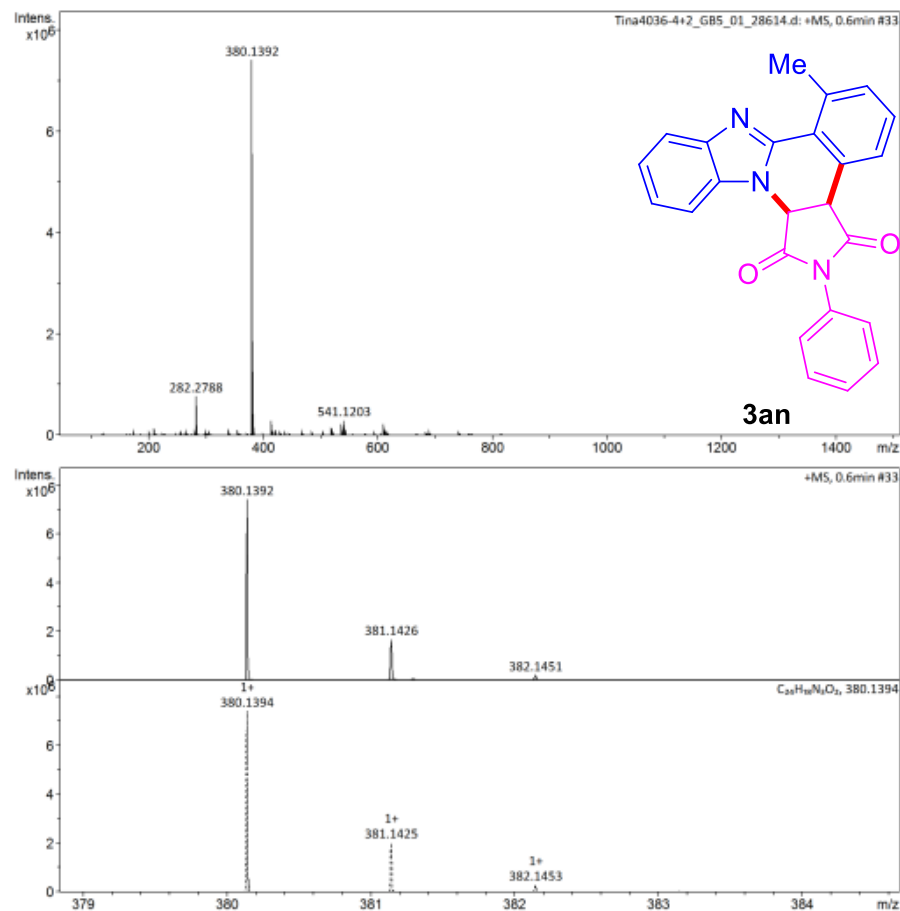


<sup>1</sup>H NMR spectrum (400 MHz) of compound **3an** in CDCl<sub>3</sub>.



$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3an** in  $\text{CDCl}_3$ .

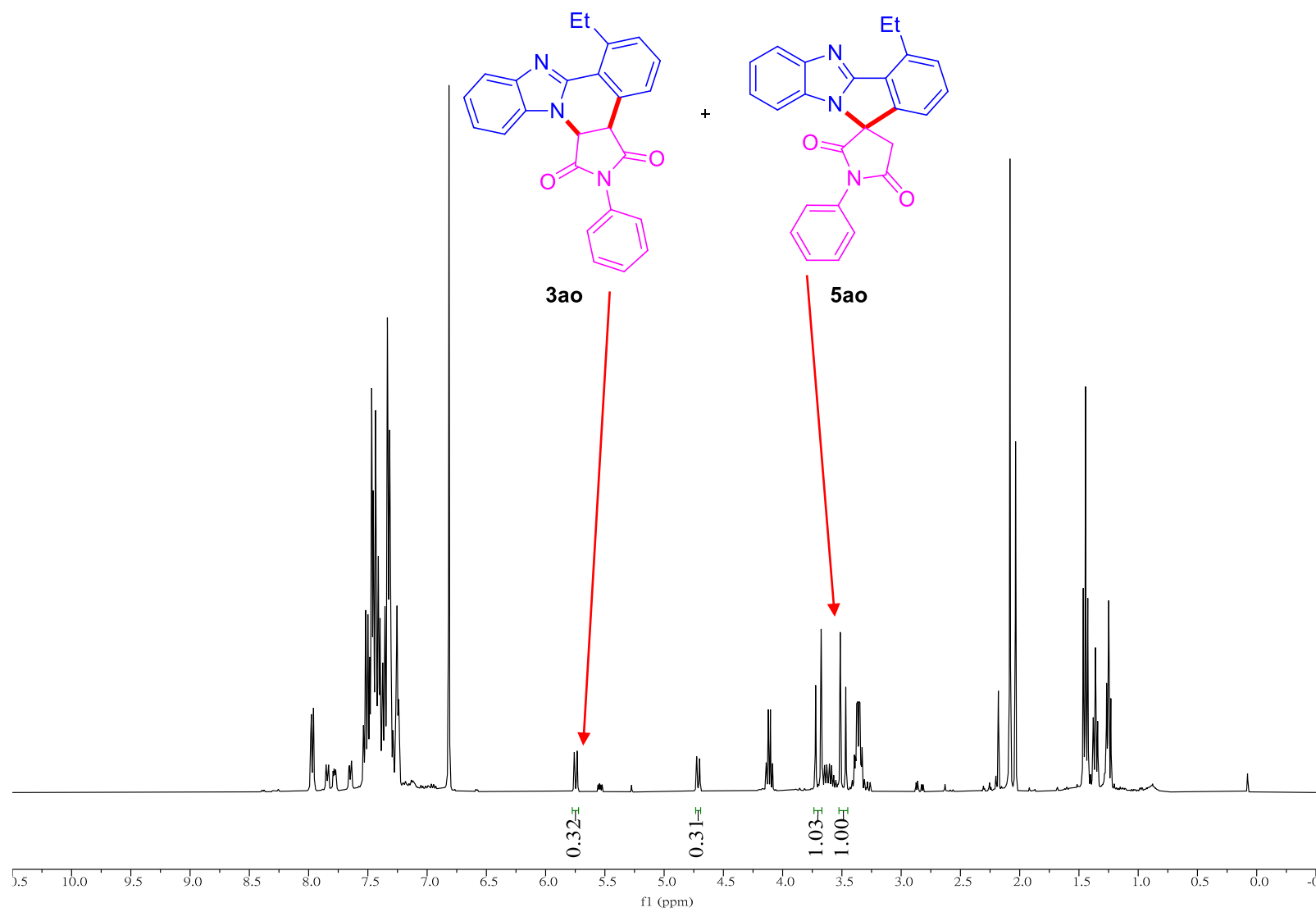




### Display Report

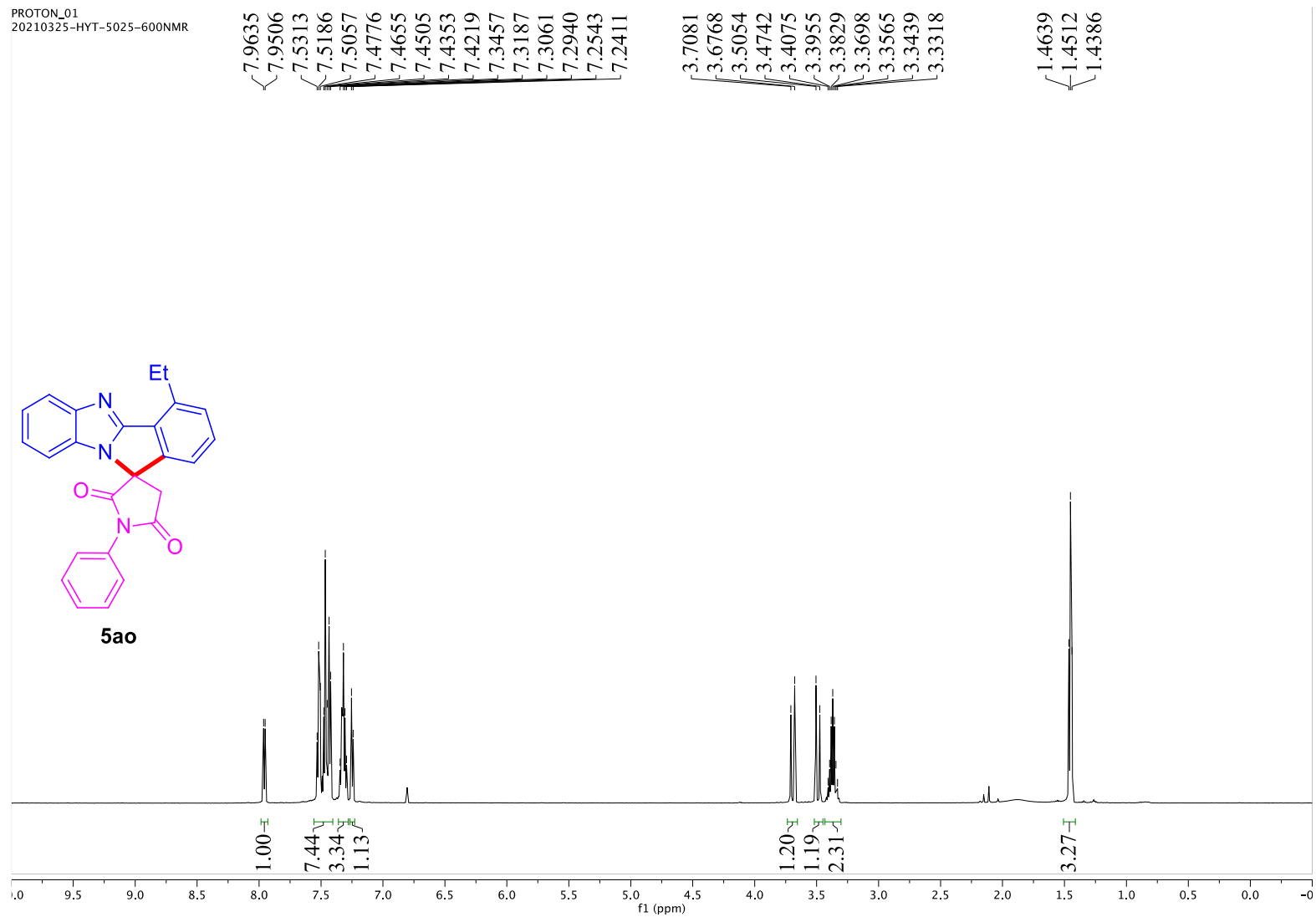
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
380.1392	1	C <sub>24</sub> H <sub>18</sub> N <sub>3</sub> O <sub>2</sub>	380.1394	0.4	24.3	1	100.00	17.5	even	ok	M+H

HRMS (ESI) of compound **3an**.

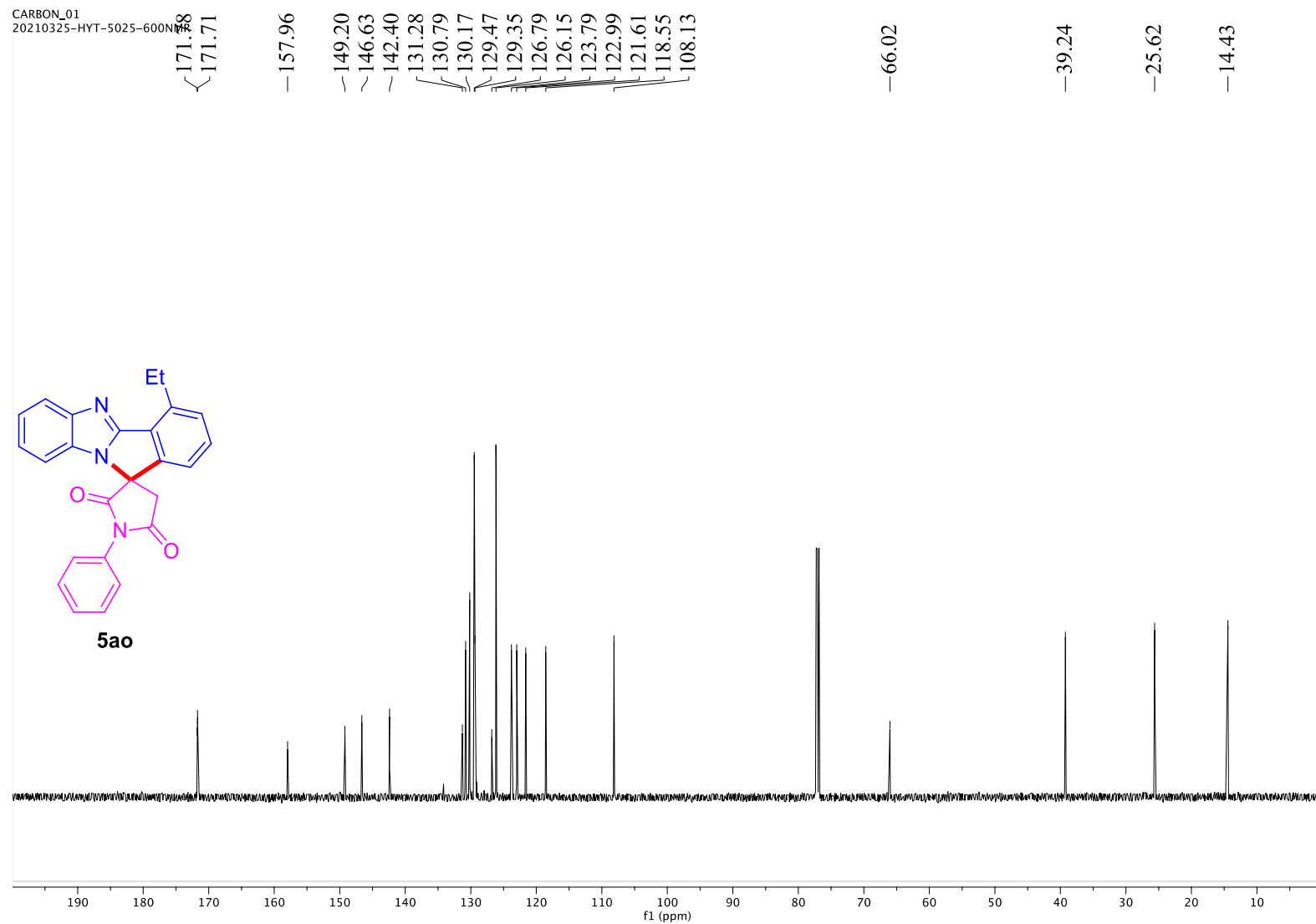


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3ao** and **5ao** in  $\text{CDCl}_3$ .

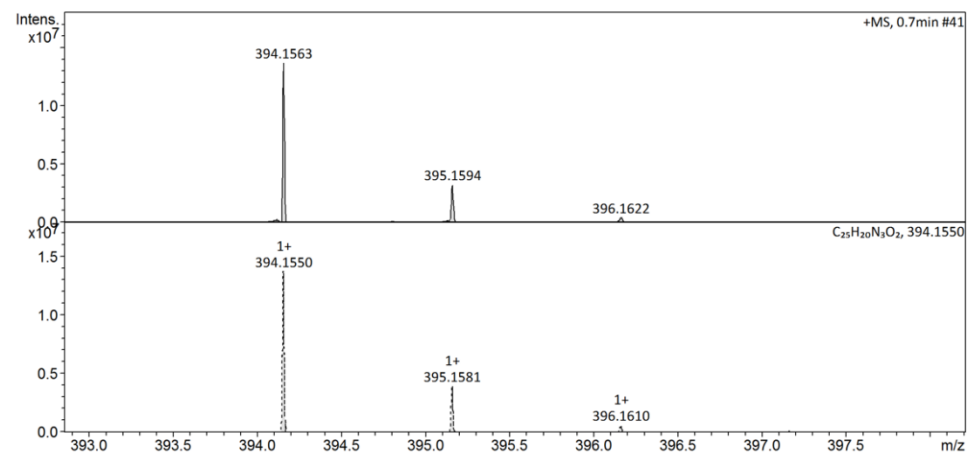
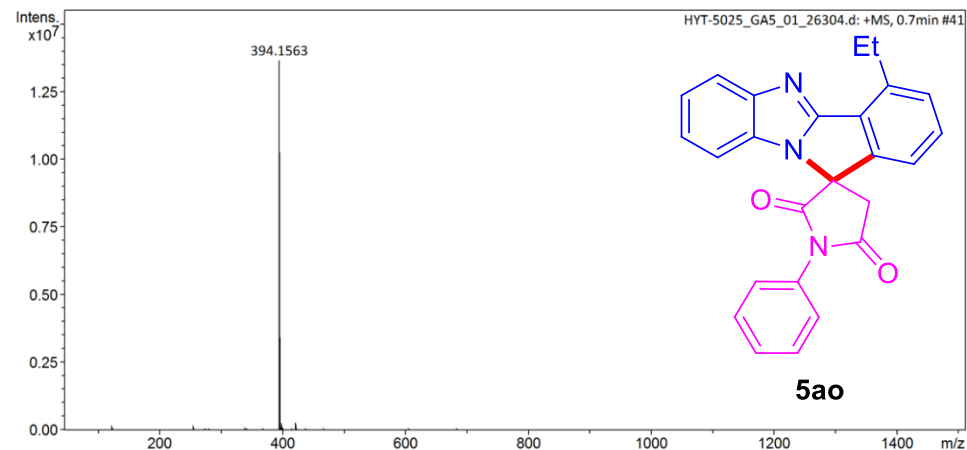
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<sup>1</sup>H NMR spectrum (600 MHz) of compound **5ao** in CDCl<sub>3</sub>.



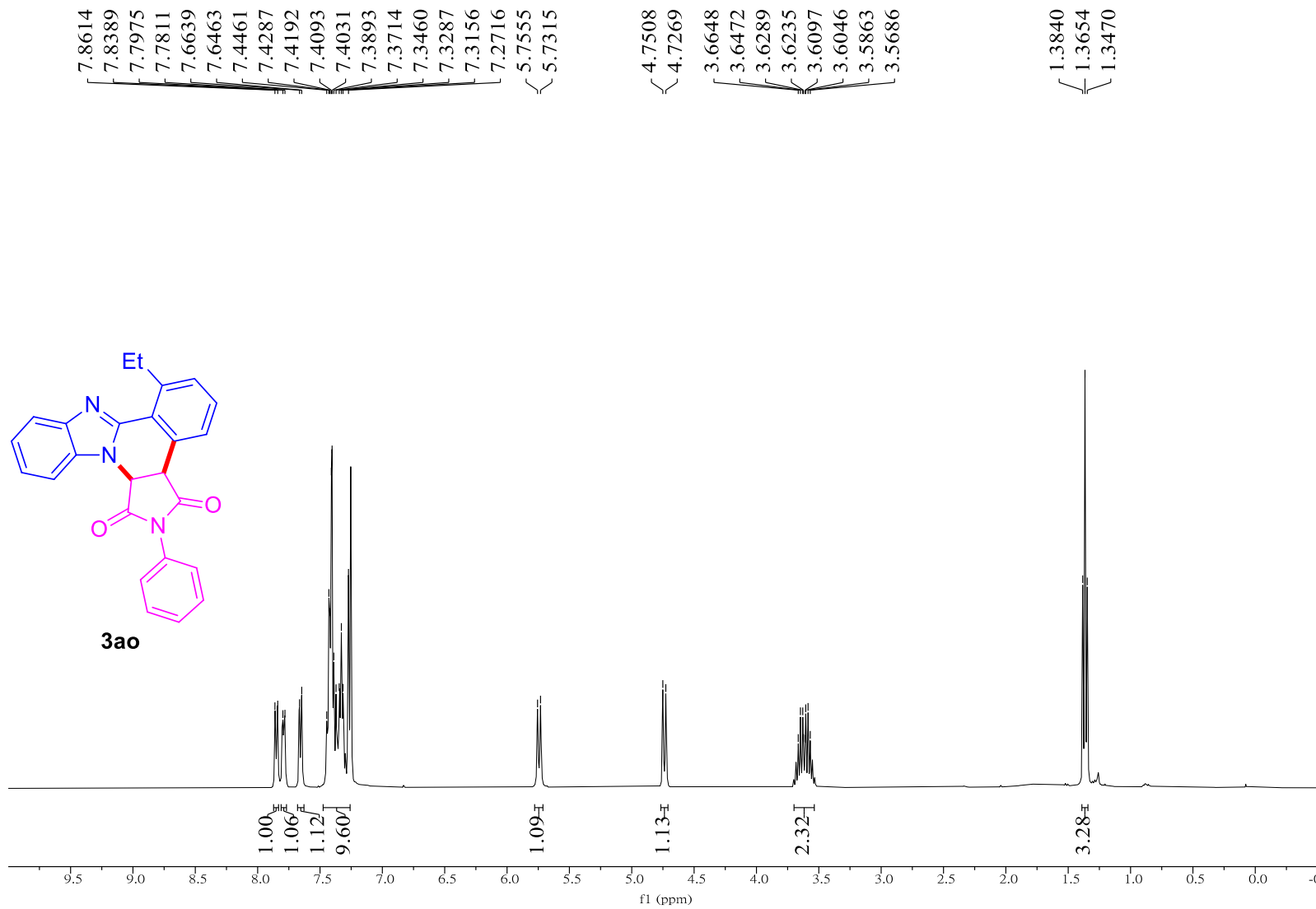
$^{13}\text{C}$  NMR spectrum (150 MHz) of compound **5ao** in  $\text{CDCl}_3$ .



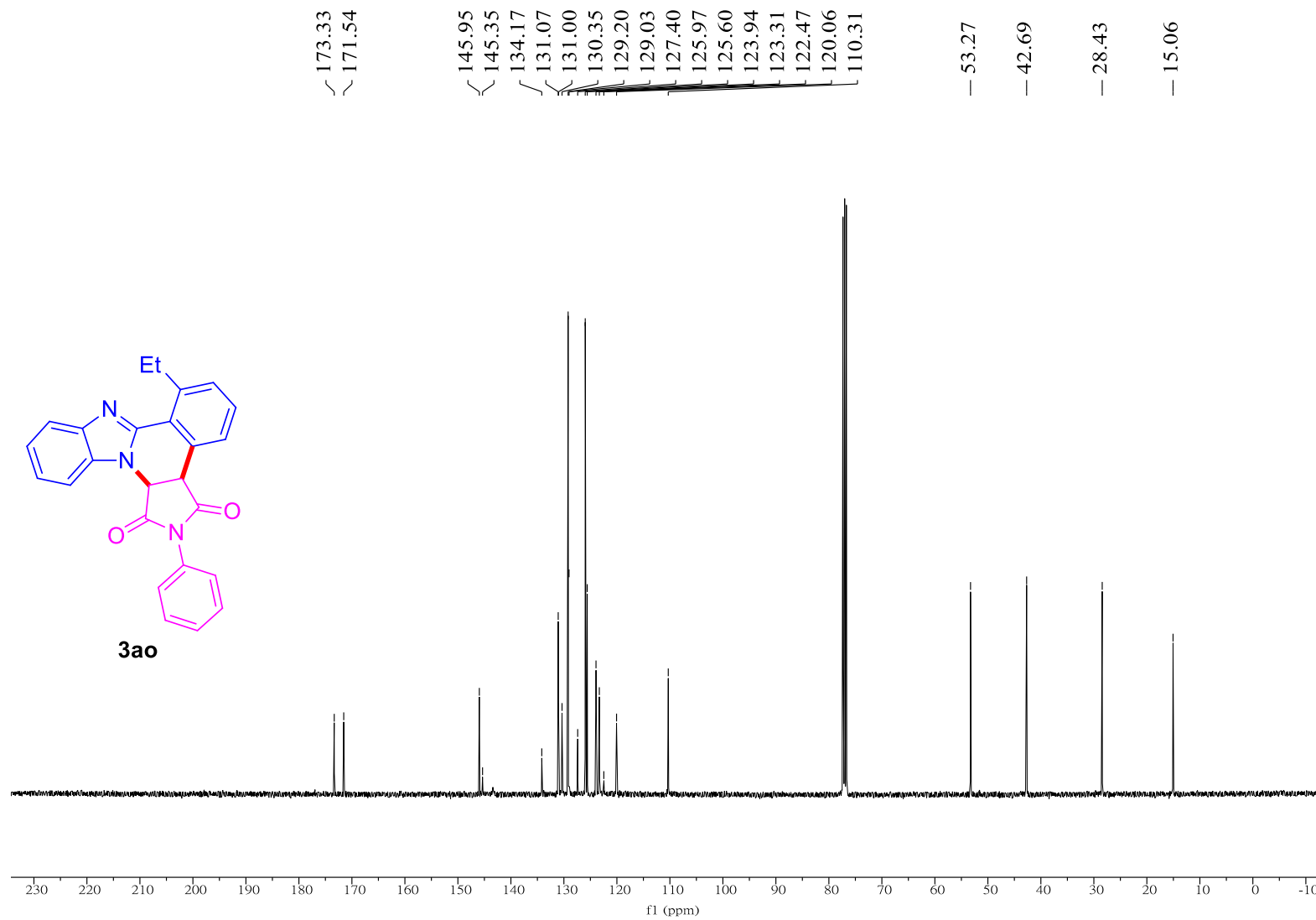
### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
394.1563	1	C <sub>25</sub> H <sub>20</sub> N <sub>3</sub> O <sub>2</sub>	394.1550	-3.2	25.6	1	100.00	17.5	even	ok	M+H

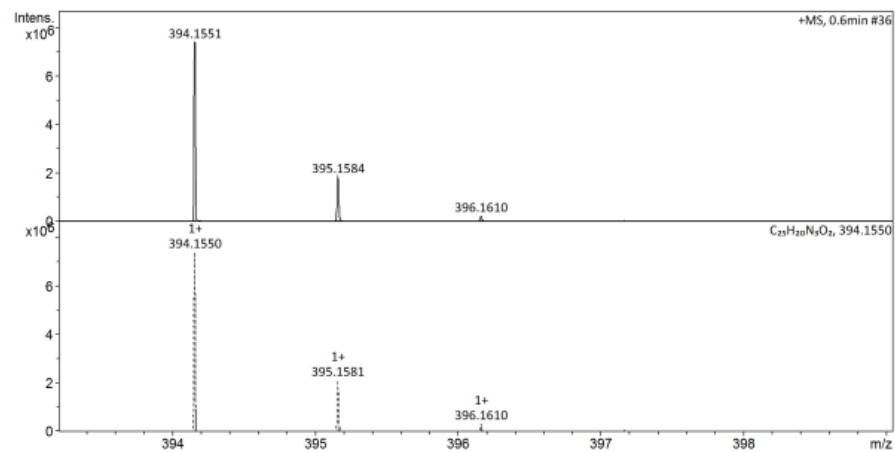
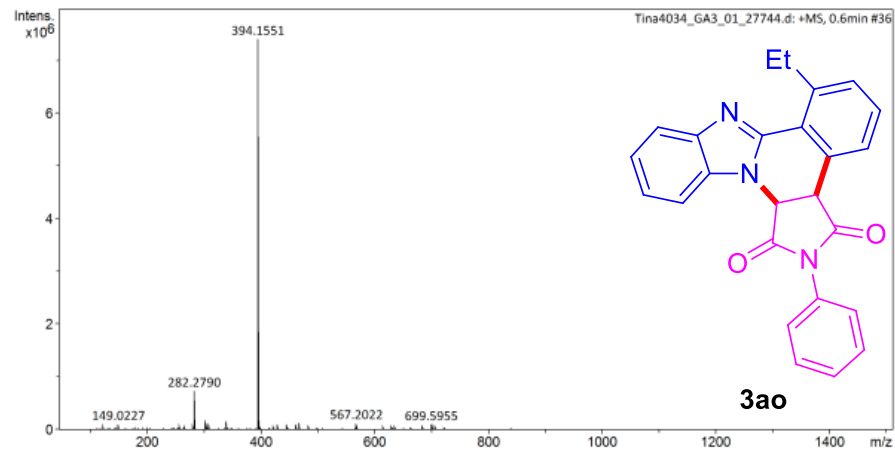
HRMS (ESI) of compound **5ao**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3ao** in  $\text{CDCl}_3$ .



<sup>13</sup>C NMR spectrum (100 MHz) of compound **3ao** in CDCl<sub>3</sub>.

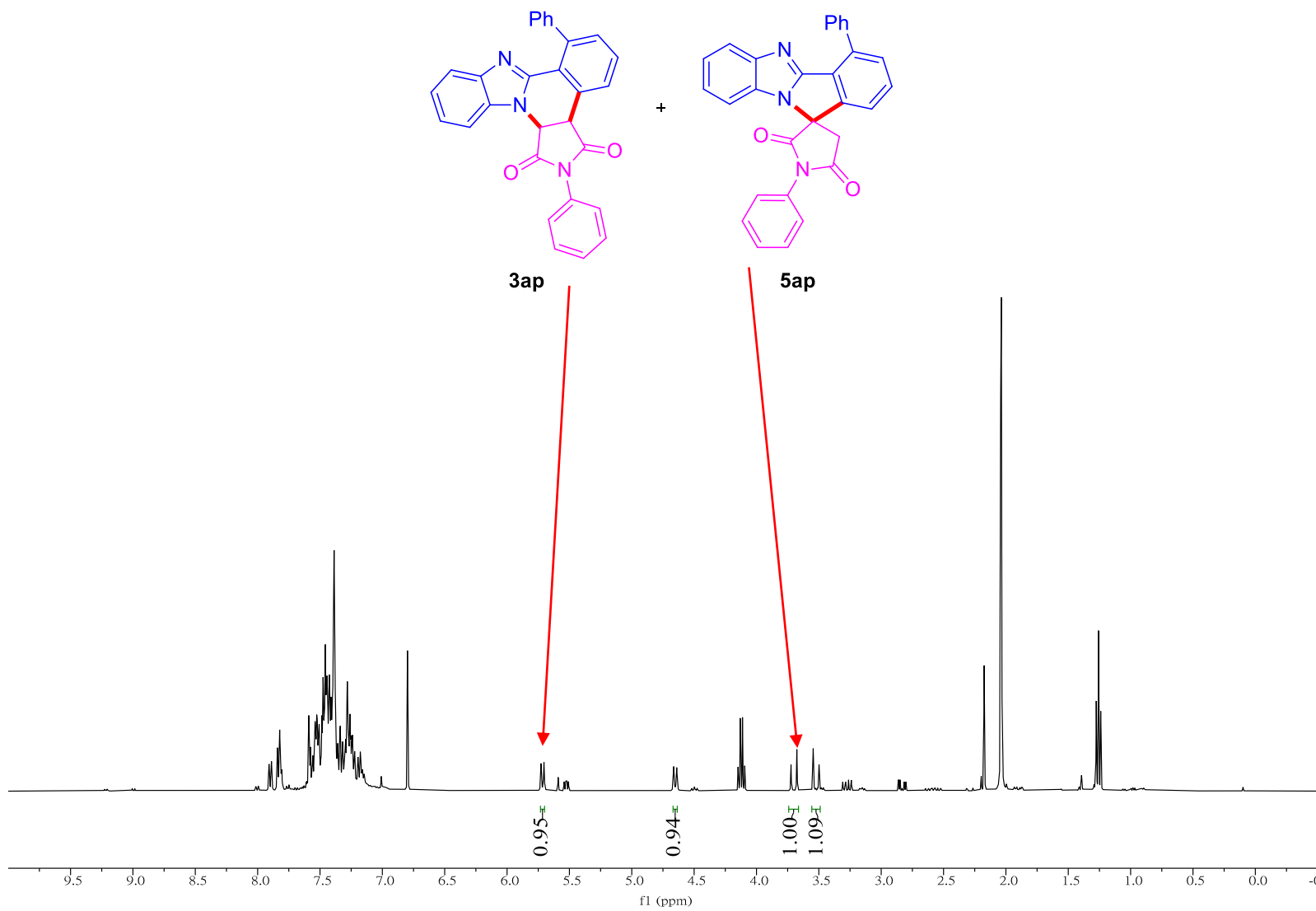


### Display Report

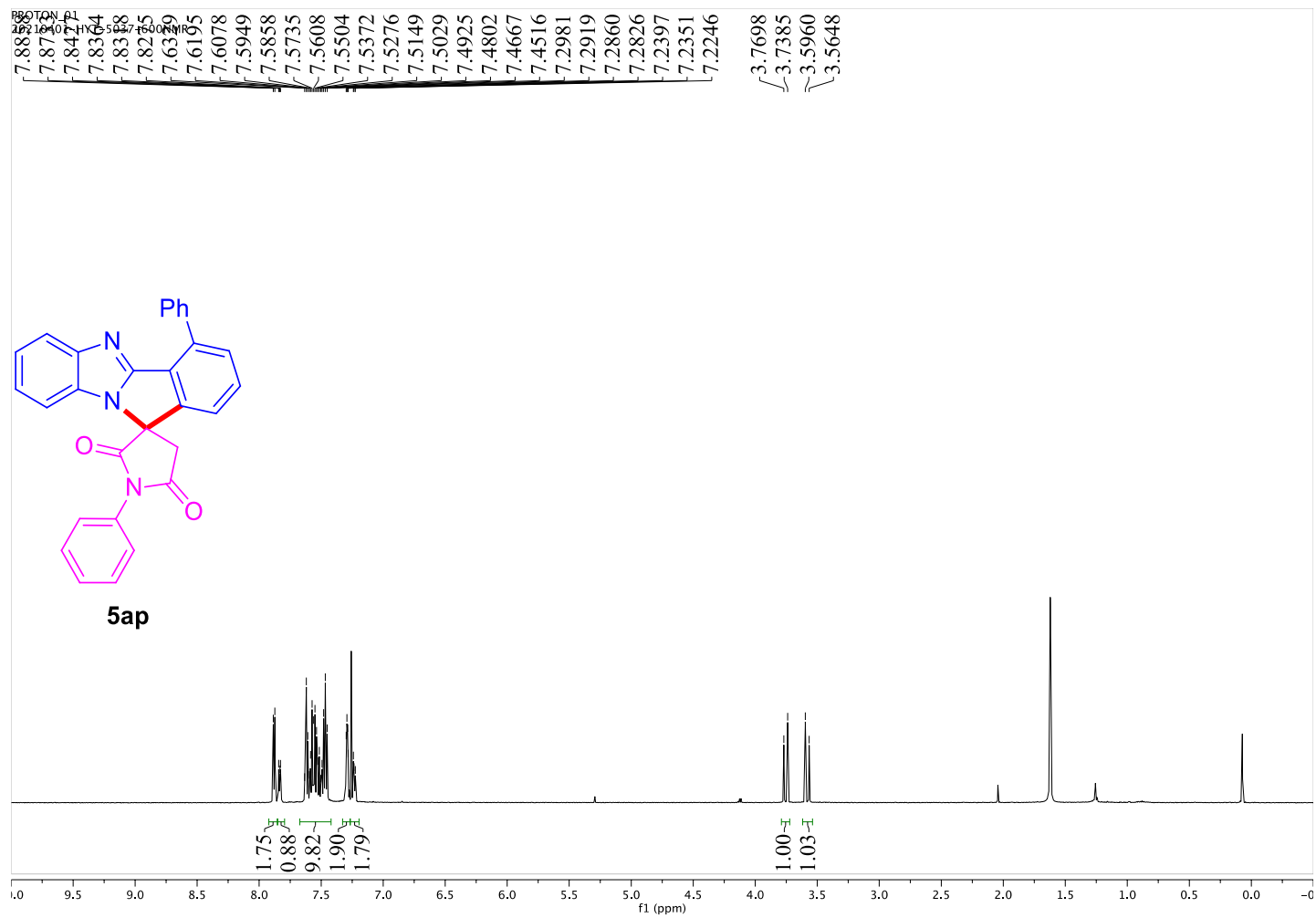
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
394.1551	1	C <sub>25</sub> H <sub>20</sub> N <sub>3</sub> O <sub>2</sub>	394.1550	0.2	19.7	1	100.00	17.5	even	ok	M+H

HRMS (ESI) of compound **3ao**.

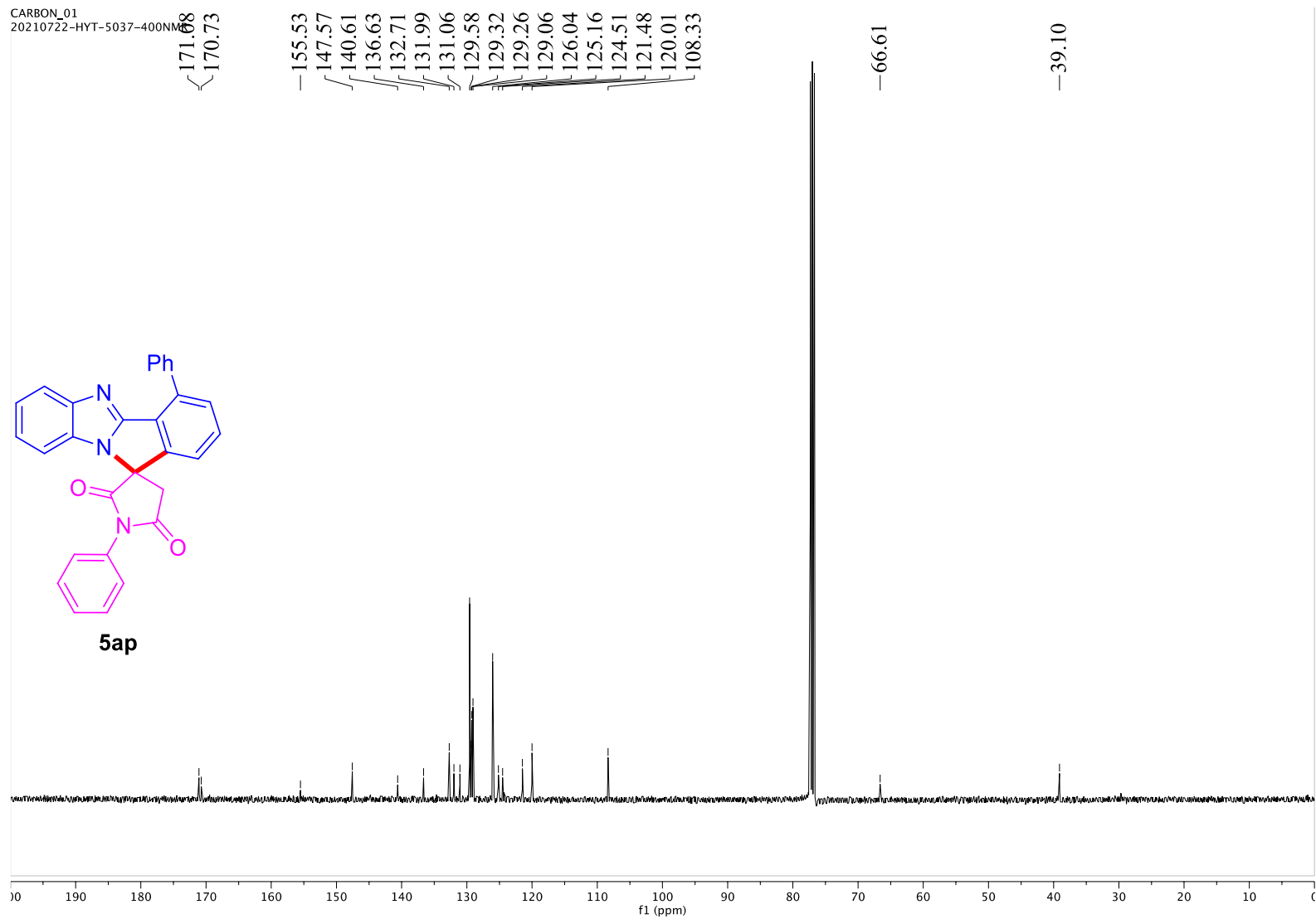




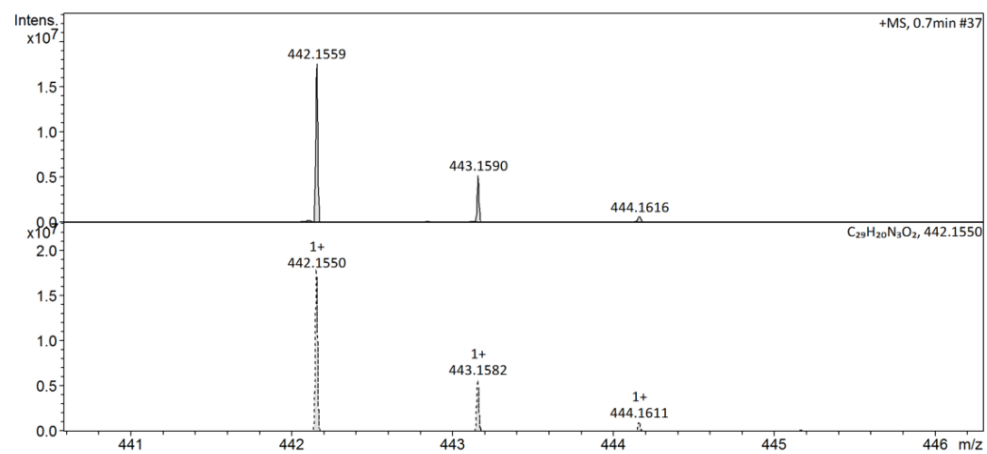
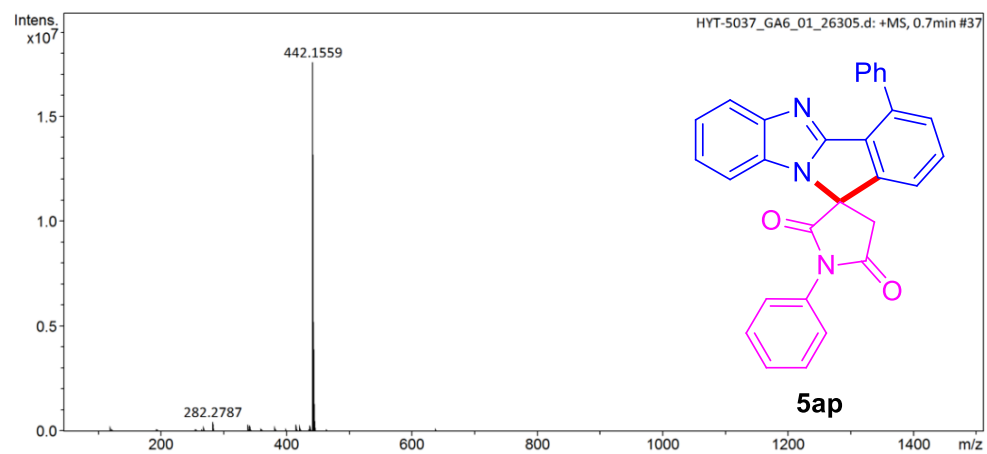
$^1\text{H}$  NMR spectrum (400 MHz) of compound **3ap** and **5ap** in  $\text{CDCl}_3$



<sup>1</sup>H NMR spectrum (600 MHz) of compound **5ap** in CDCl<sub>3</sub>.



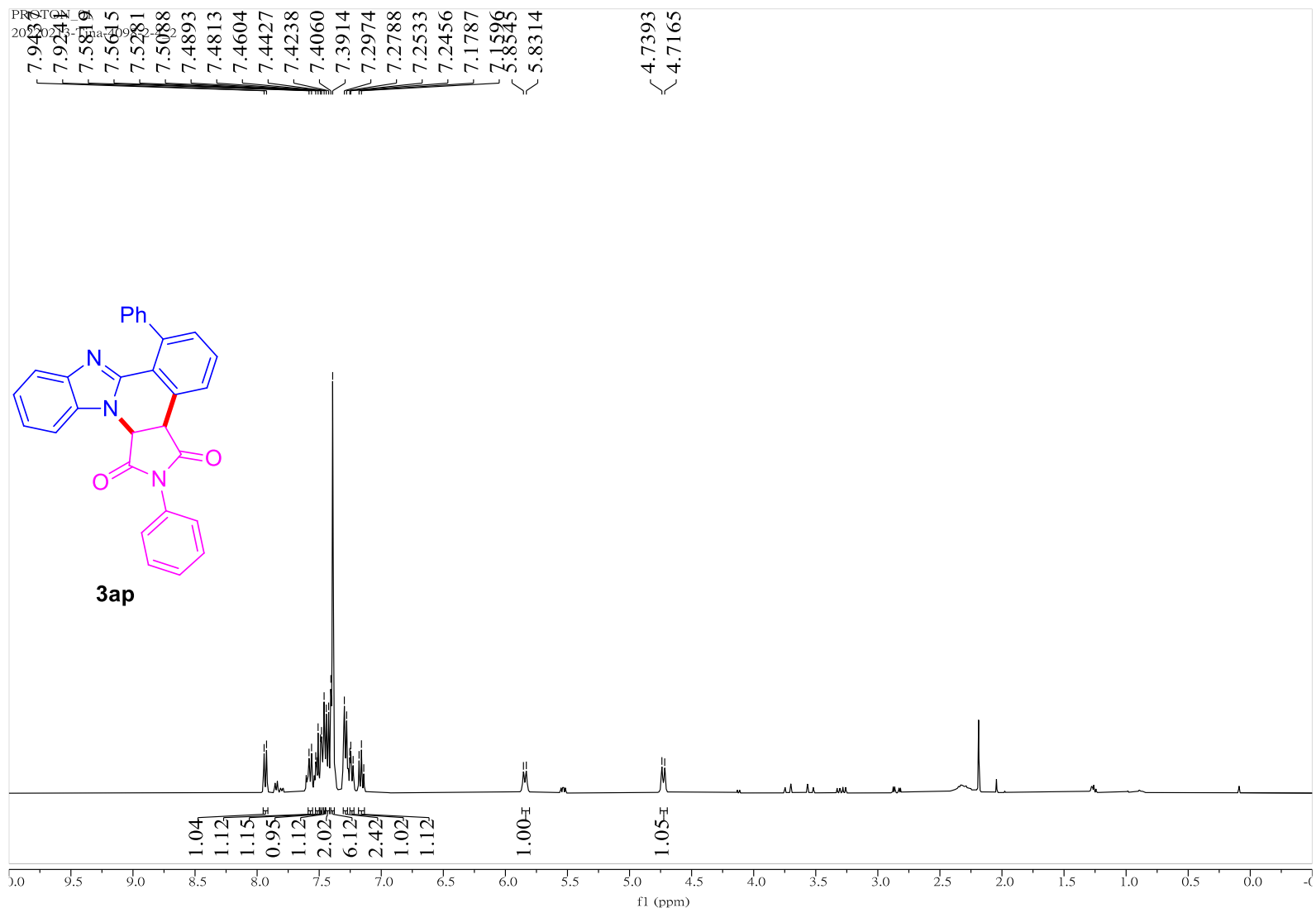
$^{13}\text{C}$  NMR spectrum (150 MHz) of compound **5ap** in  $\text{CDCl}_3$ .



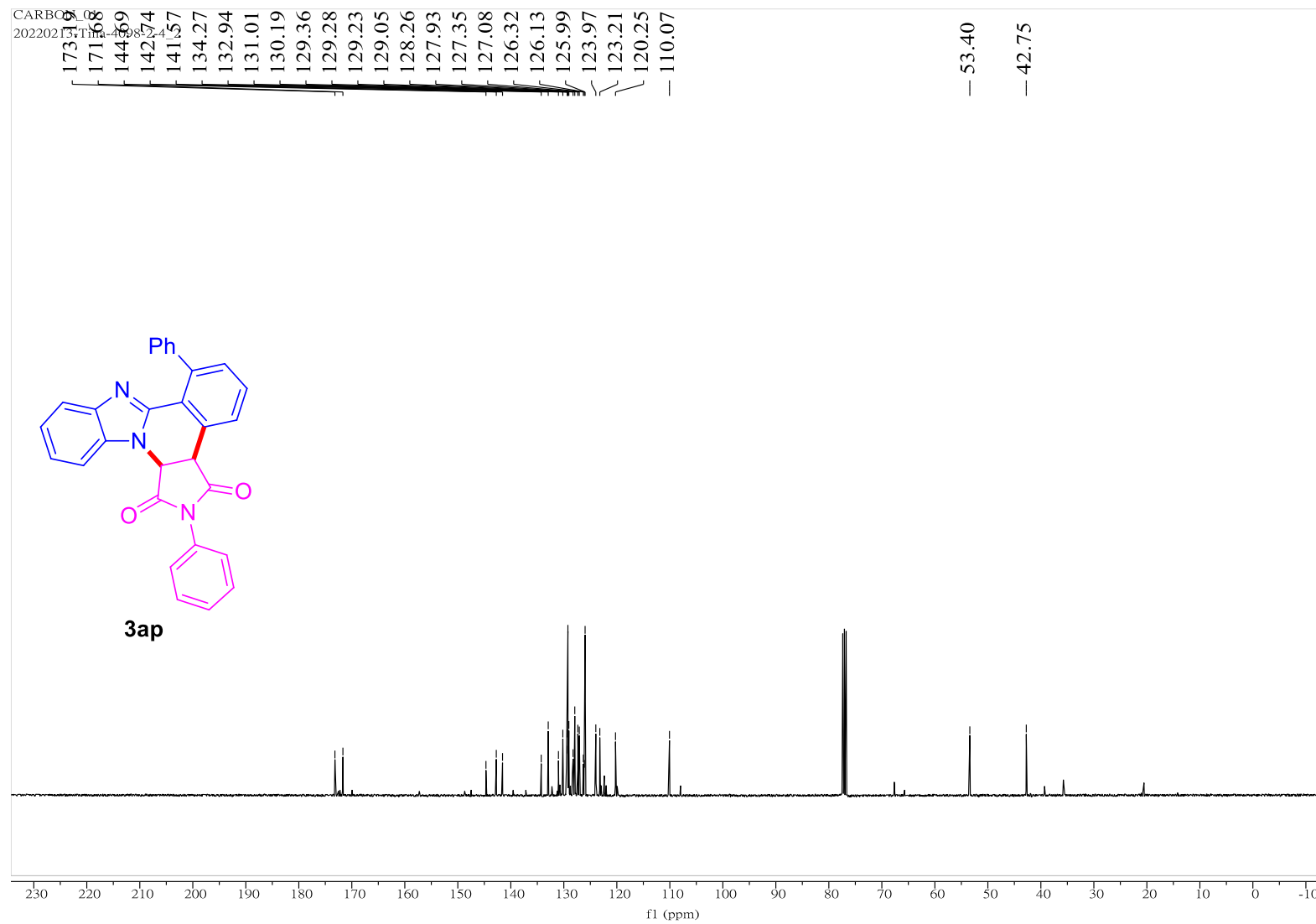
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
442.1559	1	C <sub>29</sub> H <sub>20</sub> N <sub>3</sub> O <sub>2</sub>	442.1550	-2.0	18.2	1	100.00	21.5	even	ok	M+H

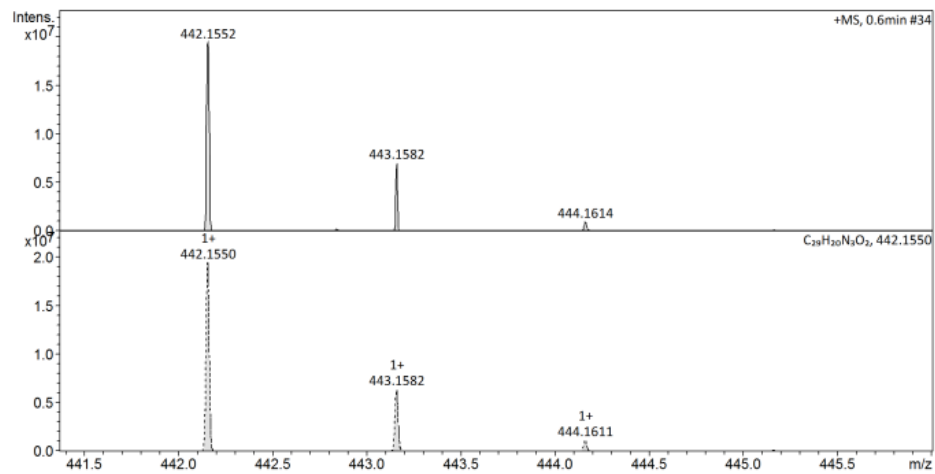
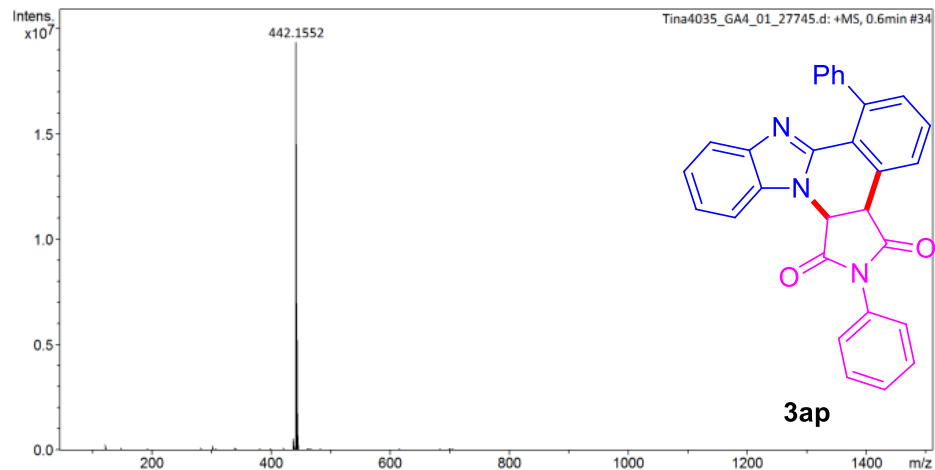
HRMS (ESI) of compound **5ap**.



$^1\text{H}$  NMR spectrum (600 MHz) of compound **3ap** in  $\text{CDCl}_3$ .



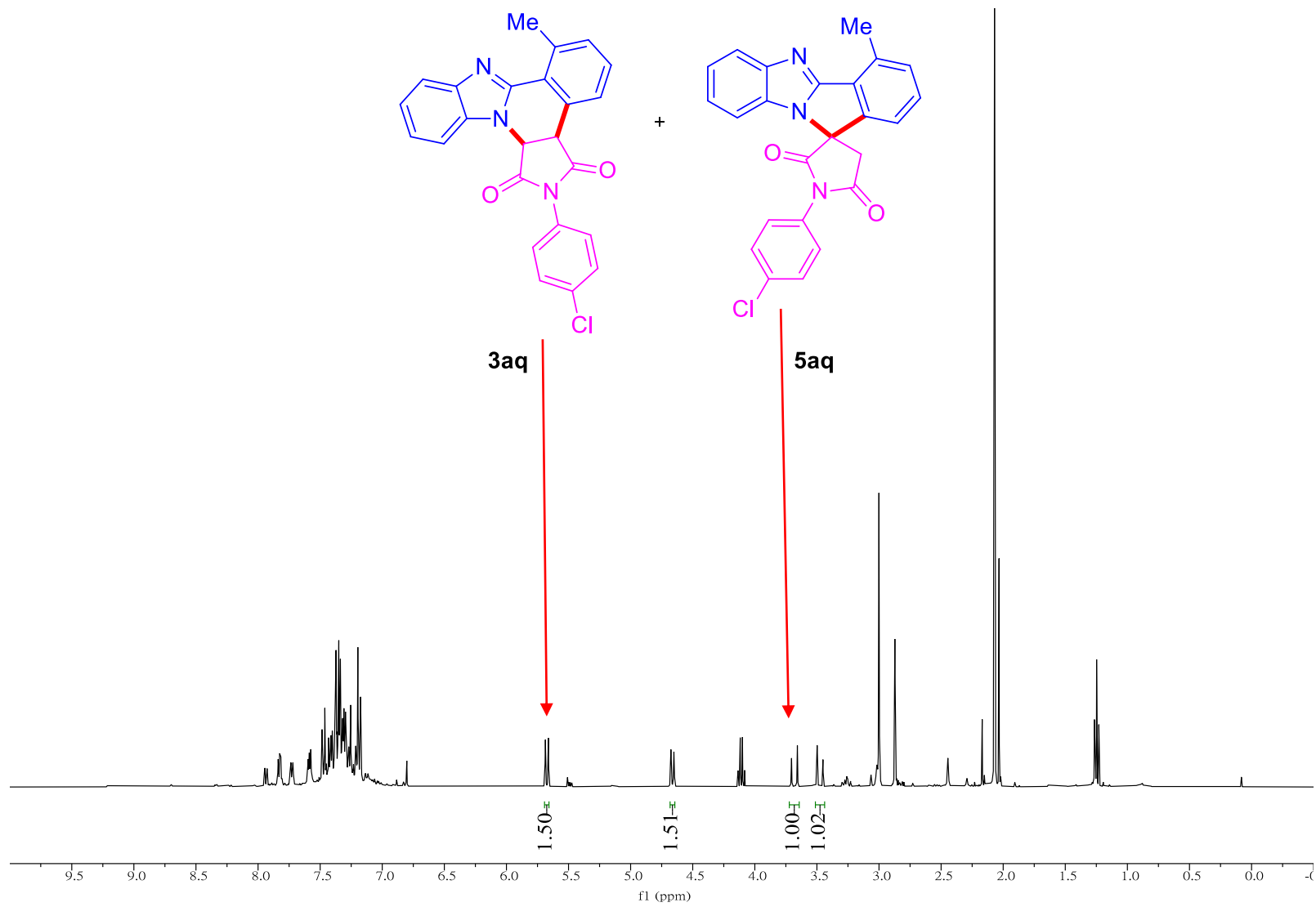
$^{13}\text{C}$  NMR spectrum (150 MHz) of compound **3ap** in  $\text{CDCl}_3$ .



### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
442.1552	1	C <sub>29</sub> H <sub>20</sub> N <sub>3</sub> O <sub>2</sub>	442.1550	0.4	16.9	1	100.00	21.5	even	ok	M+H

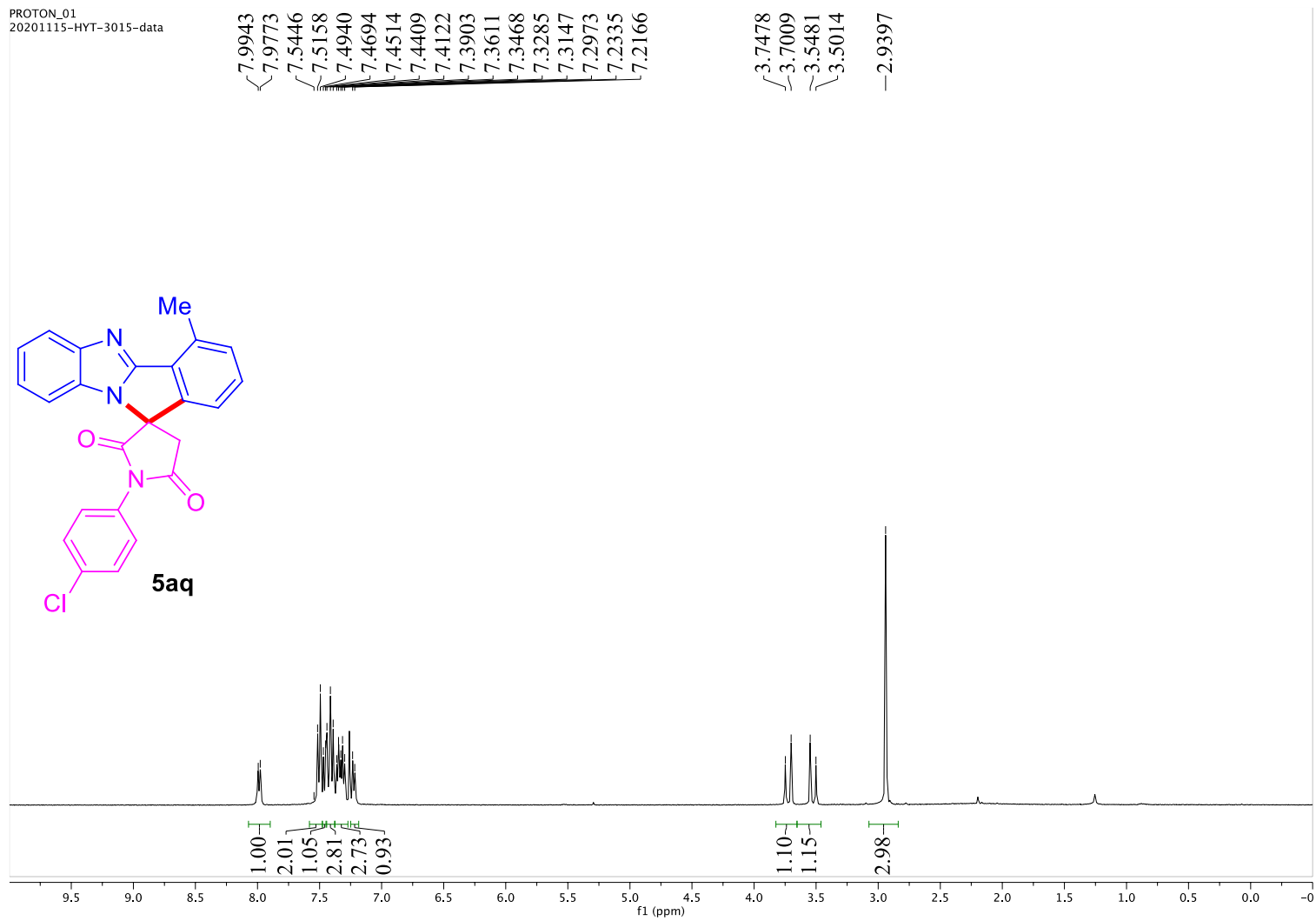
HRMS (ESI) of compound **3ap**.



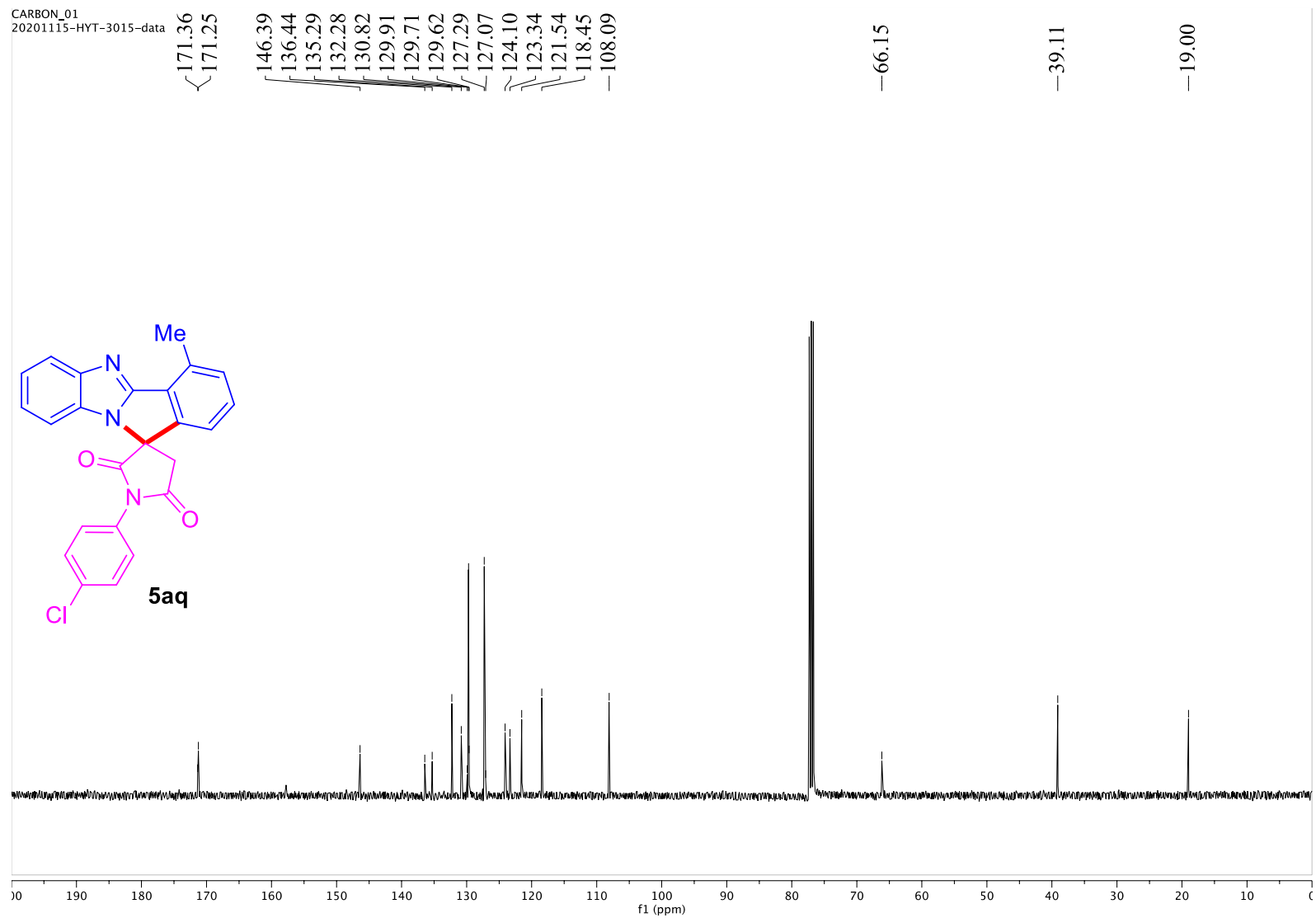
$^1\text{H}$  NMR spectrum (400 MHz) of compound **3aq** and **5aq** in  $\text{CDCl}_3$ .



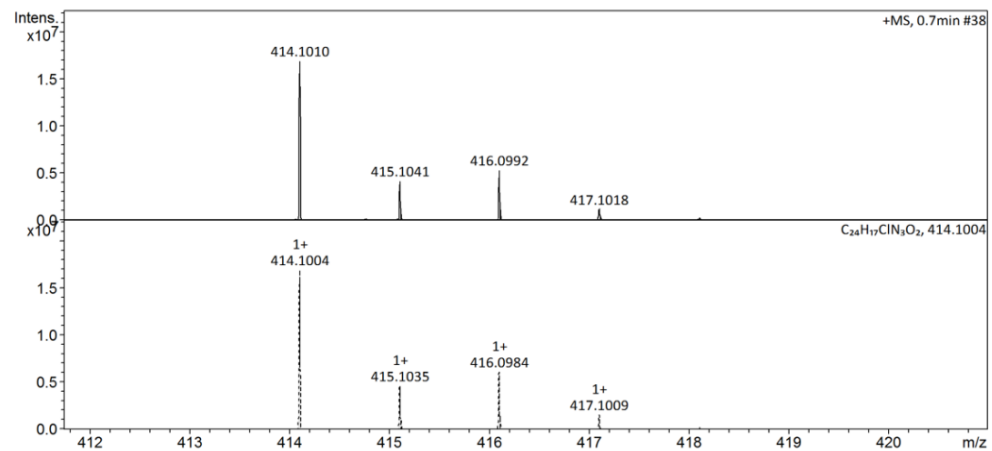
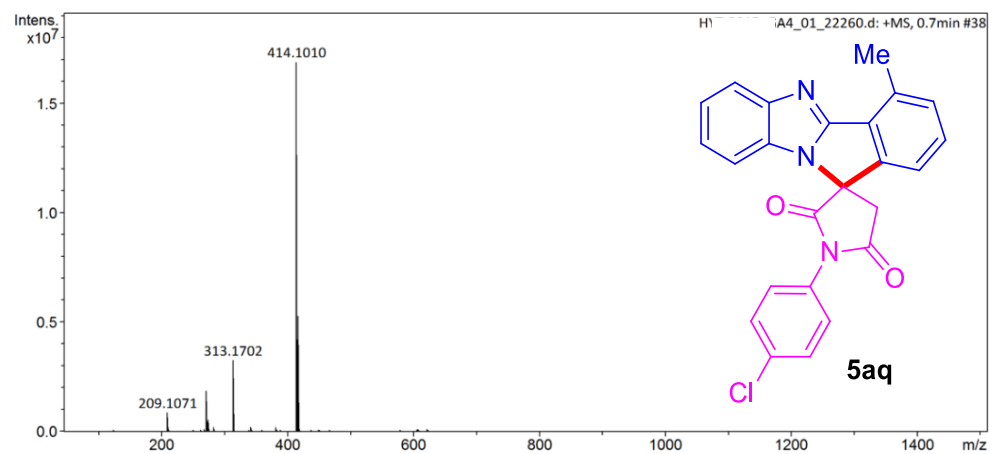
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<sup>1</sup>H NMR spectrum (400 MHz) of compound **5aq** in CDCl<sub>3</sub>.



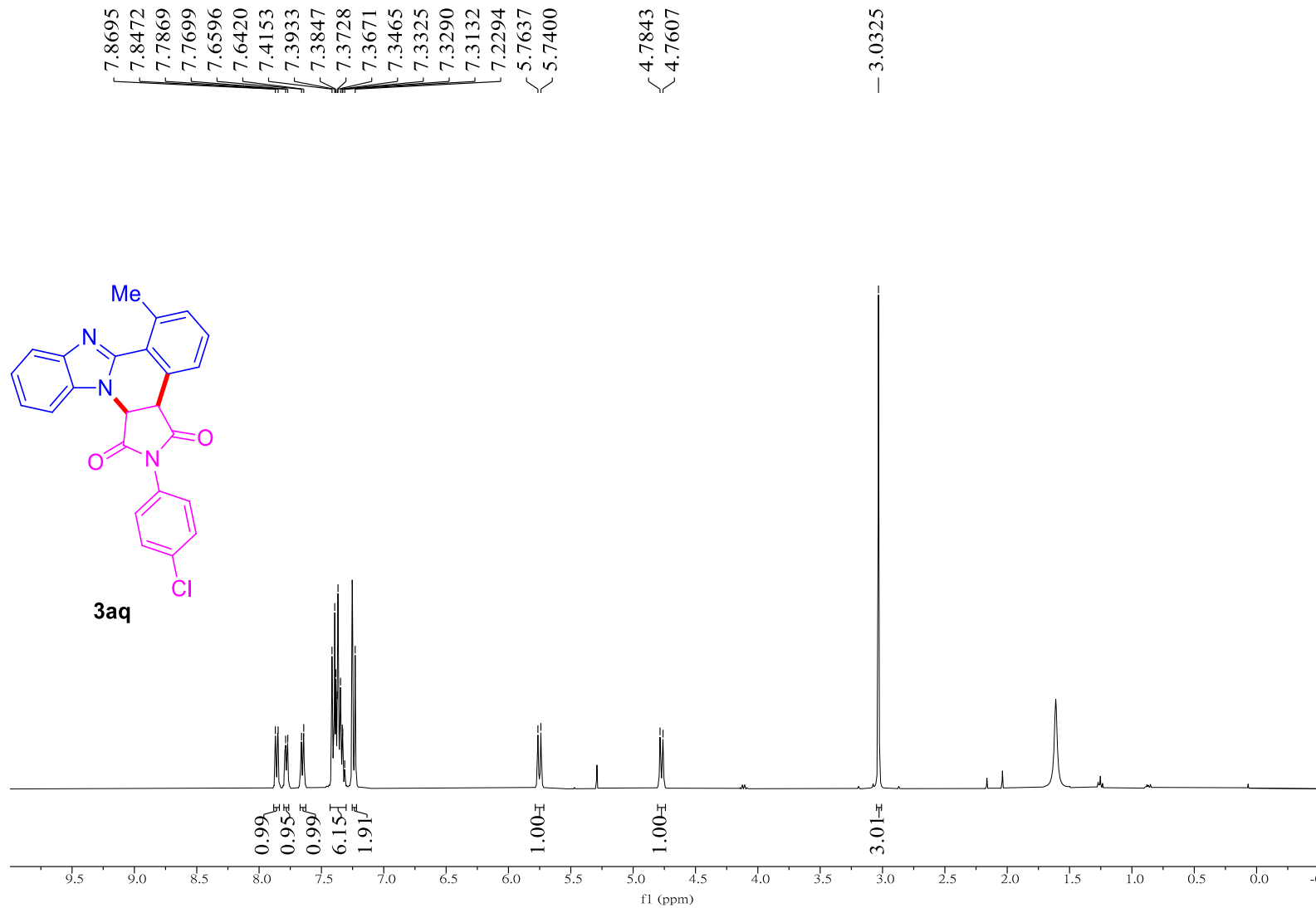
<sup>13</sup>C NMR spectrum (100 MHz) of compound **5aq** in CDCl<sub>3</sub>.



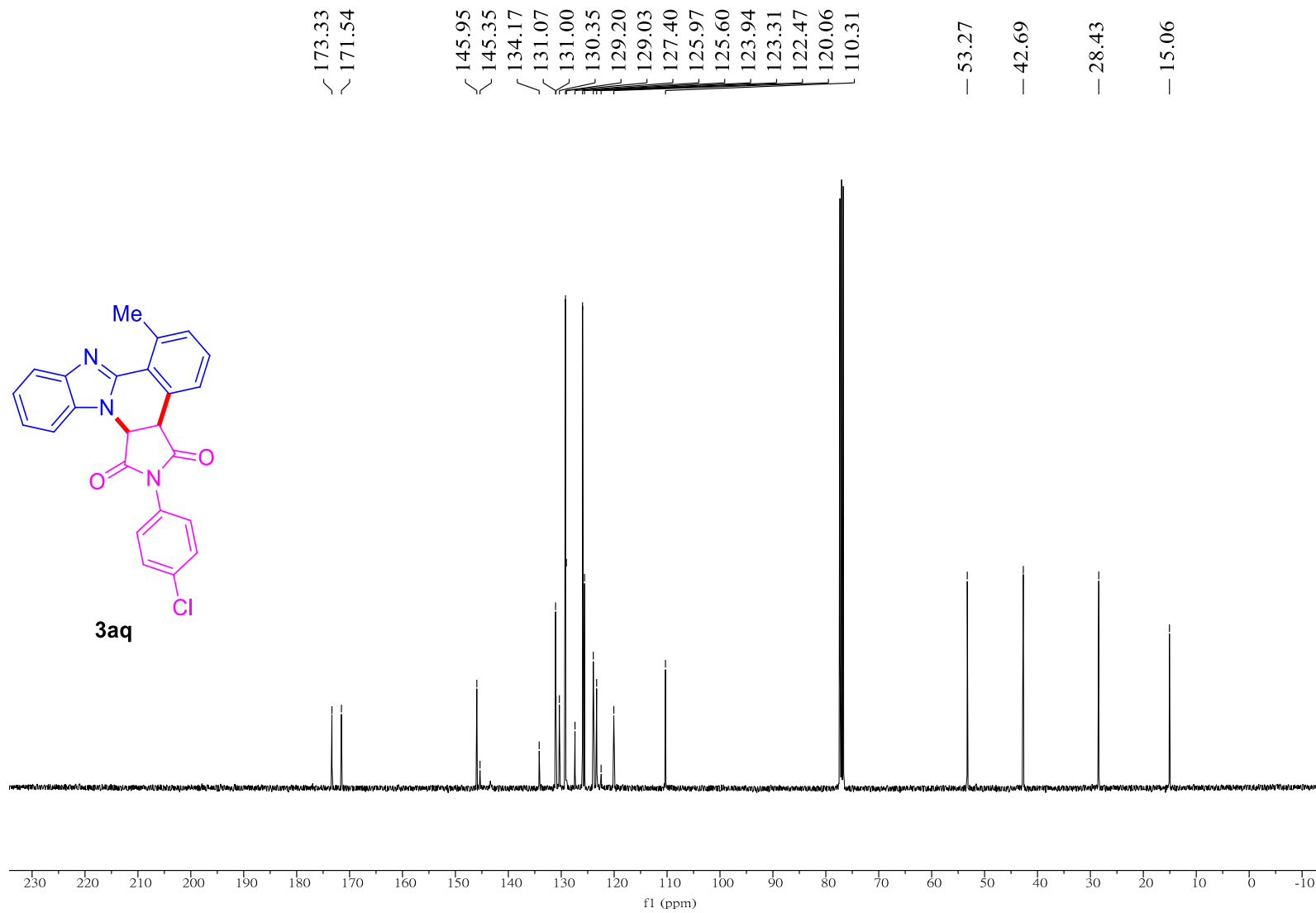
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
414.1010	1	C <sub>24</sub> H <sub>17</sub> ClN <sub>3</sub> O <sub>2</sub>	414.1004	1.4	26.3	1	100.00	17.5	even	ok	M+H

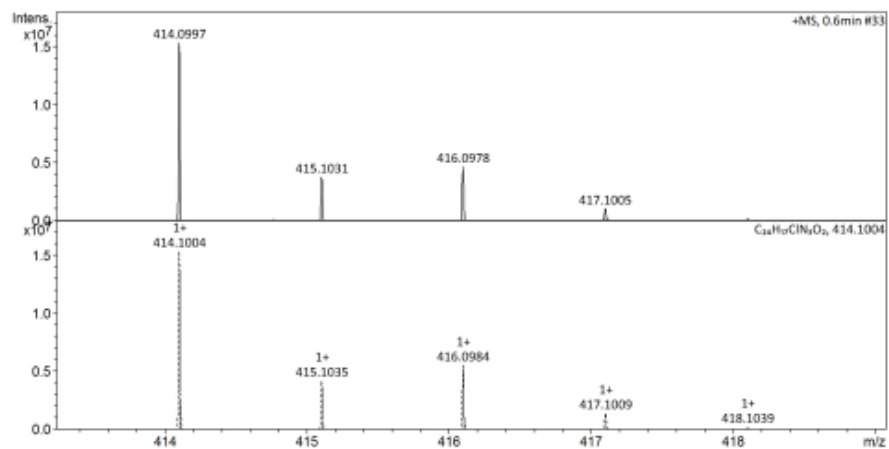
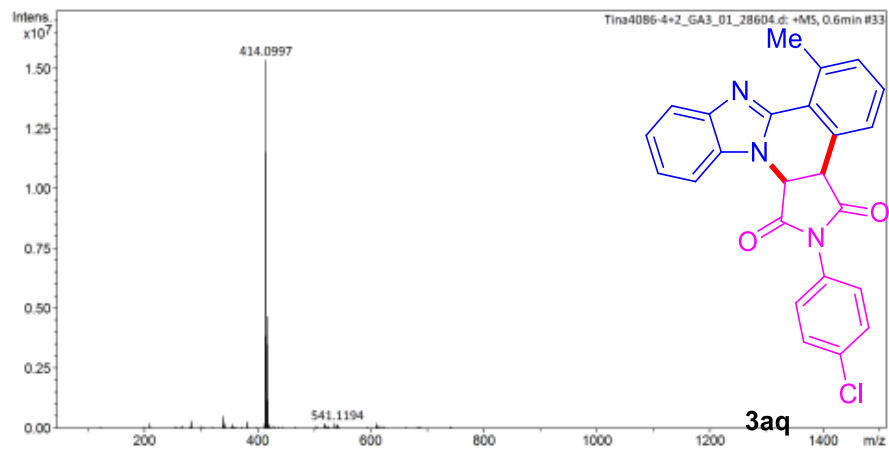
HRMS (ESI) of compound **5aq**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3aq** in  $\text{CDCl}_3$ .



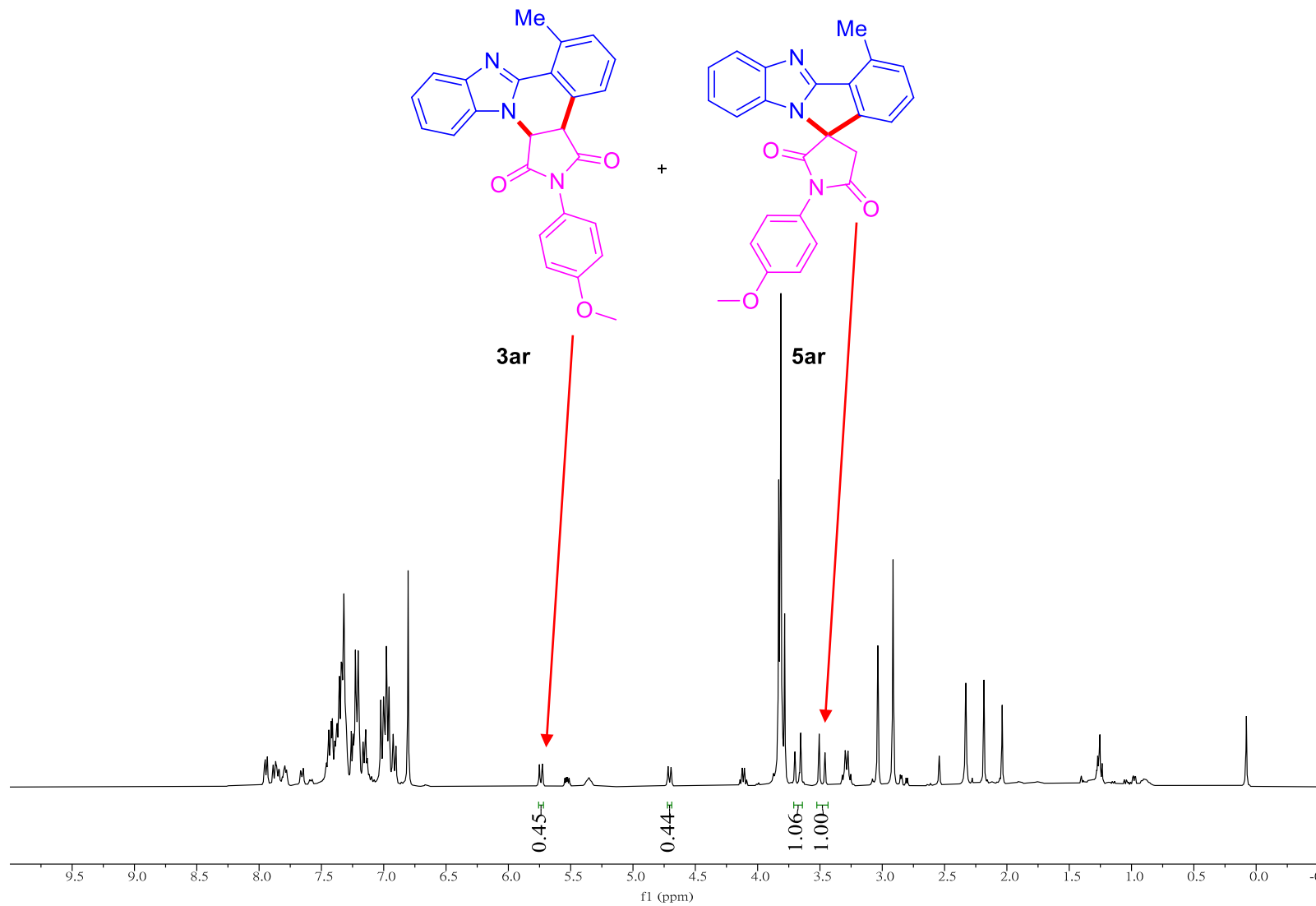
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3aq** in  $\text{CDCl}_3$ .



### Display Report

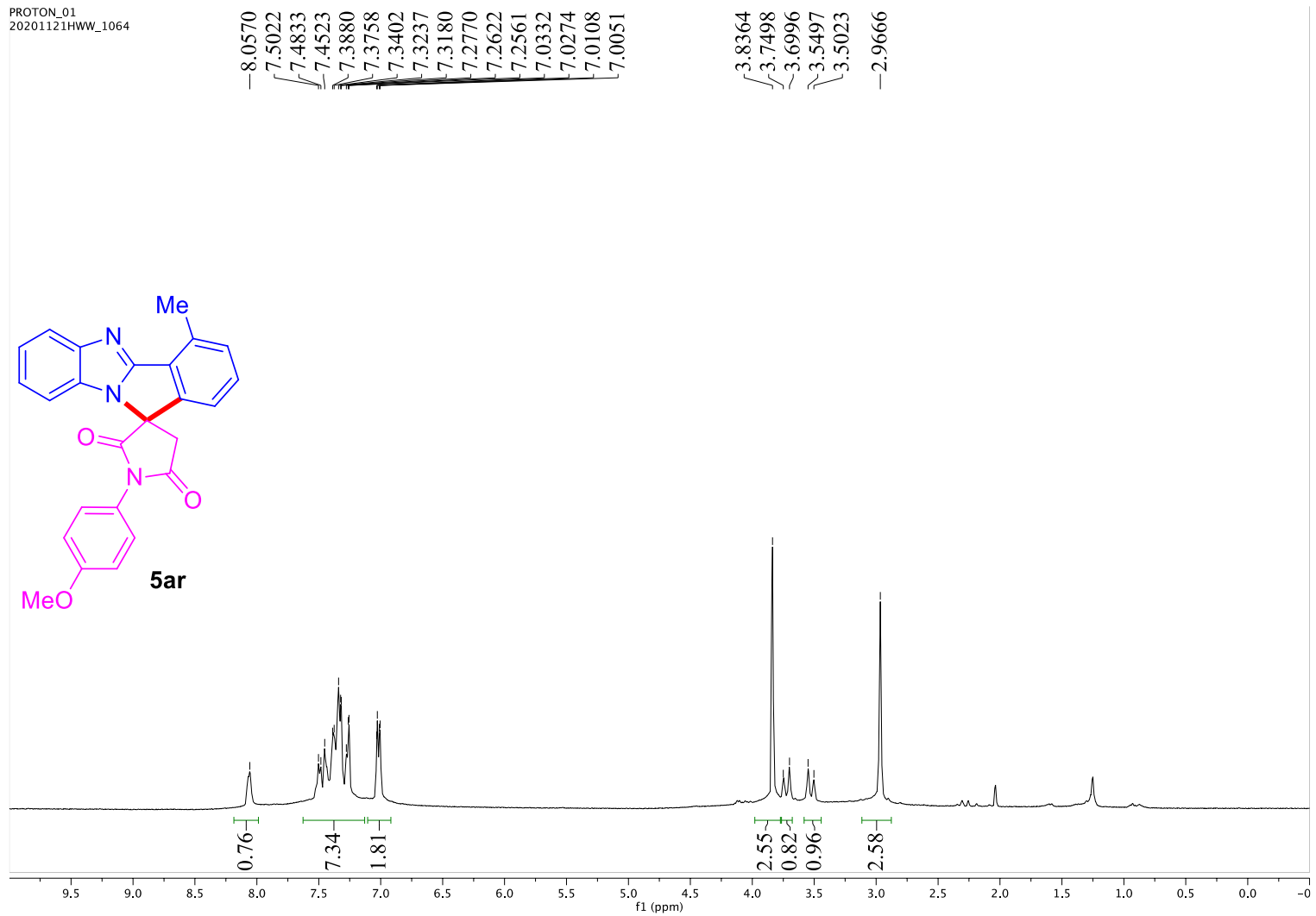
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
414.0997	1	C <sub>24</sub> H <sub>17</sub> ClN <sub>3</sub> O <sub>2</sub>	414.1004	-1.6	30.6	1	100.00	17.5	even	ok	M+H

HRMS (ESI) of compound **3aq**.



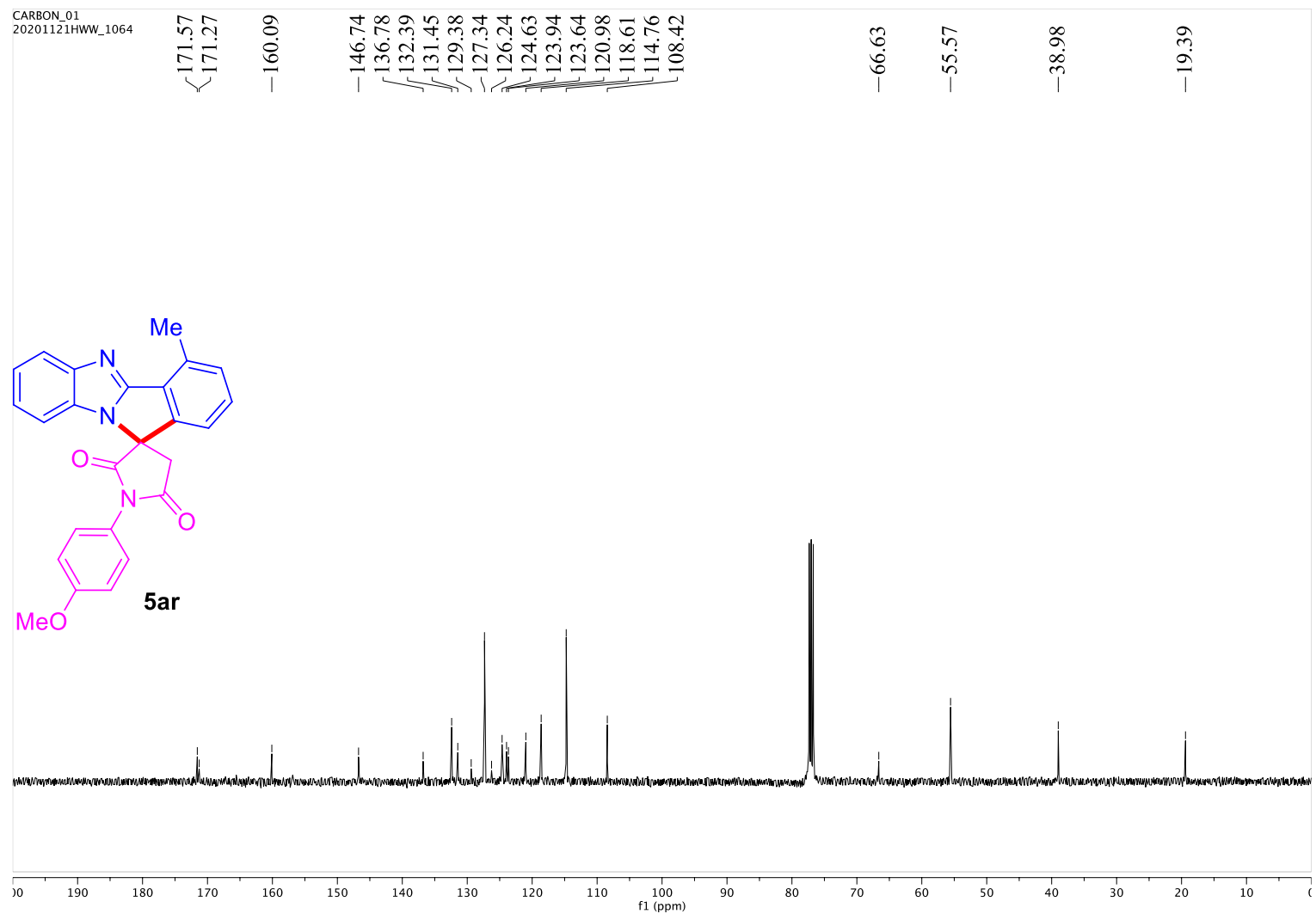
<sup>1</sup>H NMR spectrum (400 MHz) of compound **3ar** and **5ar** in CDCl<sub>3</sub>.

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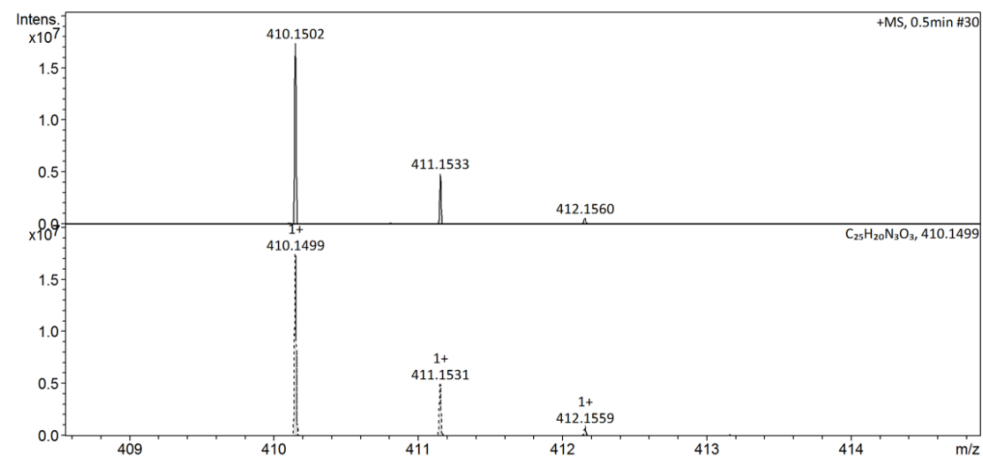
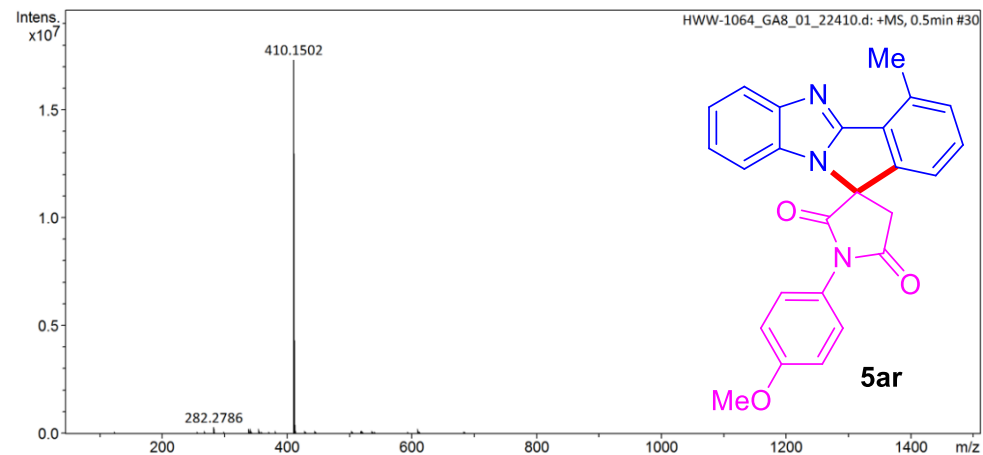


<sup>1</sup>H NMR spectrum (400 MHz) of compound **5ar** in CDCl<sub>3</sub>.





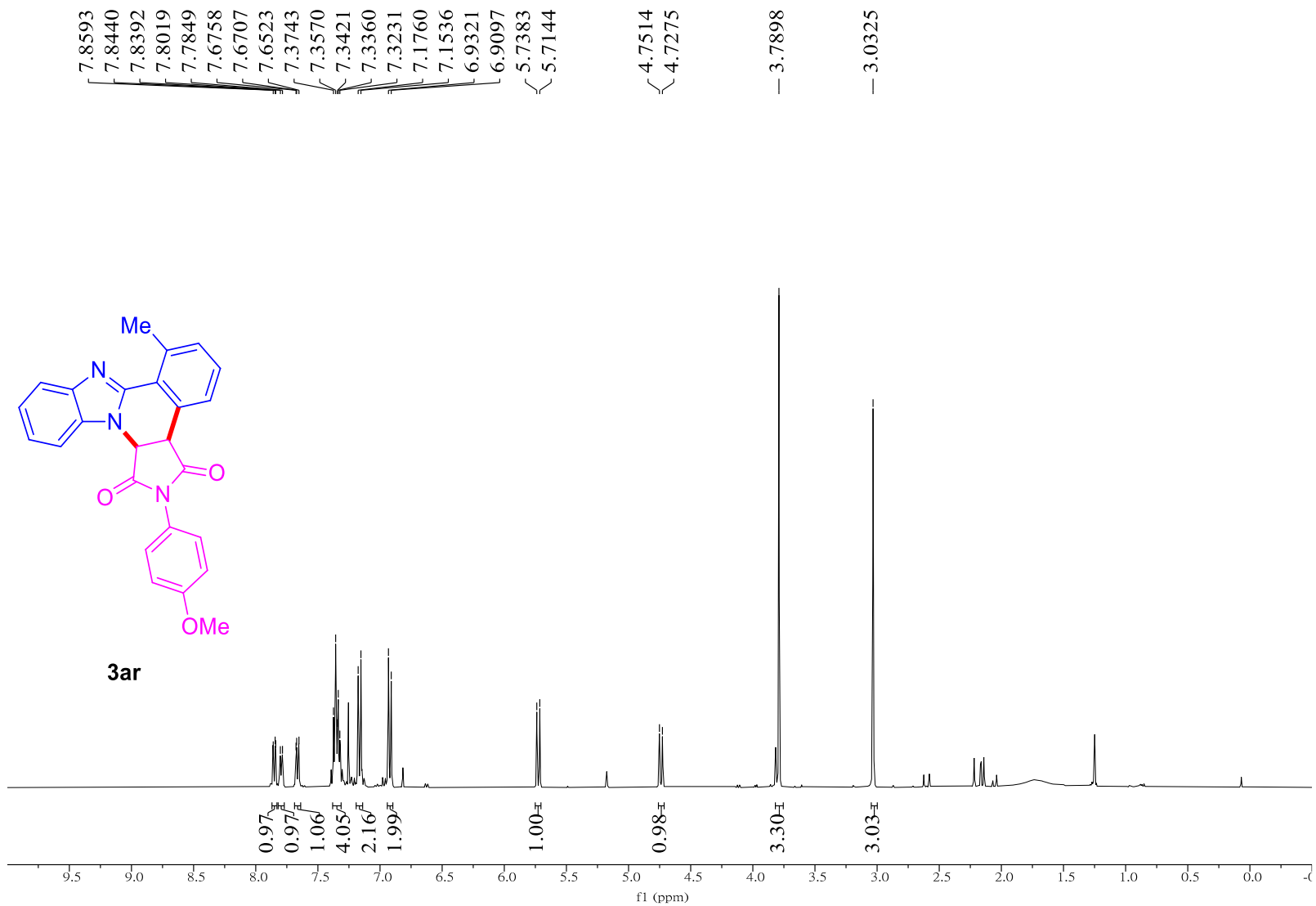
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5ar** in  $\text{CDCl}_3$ .



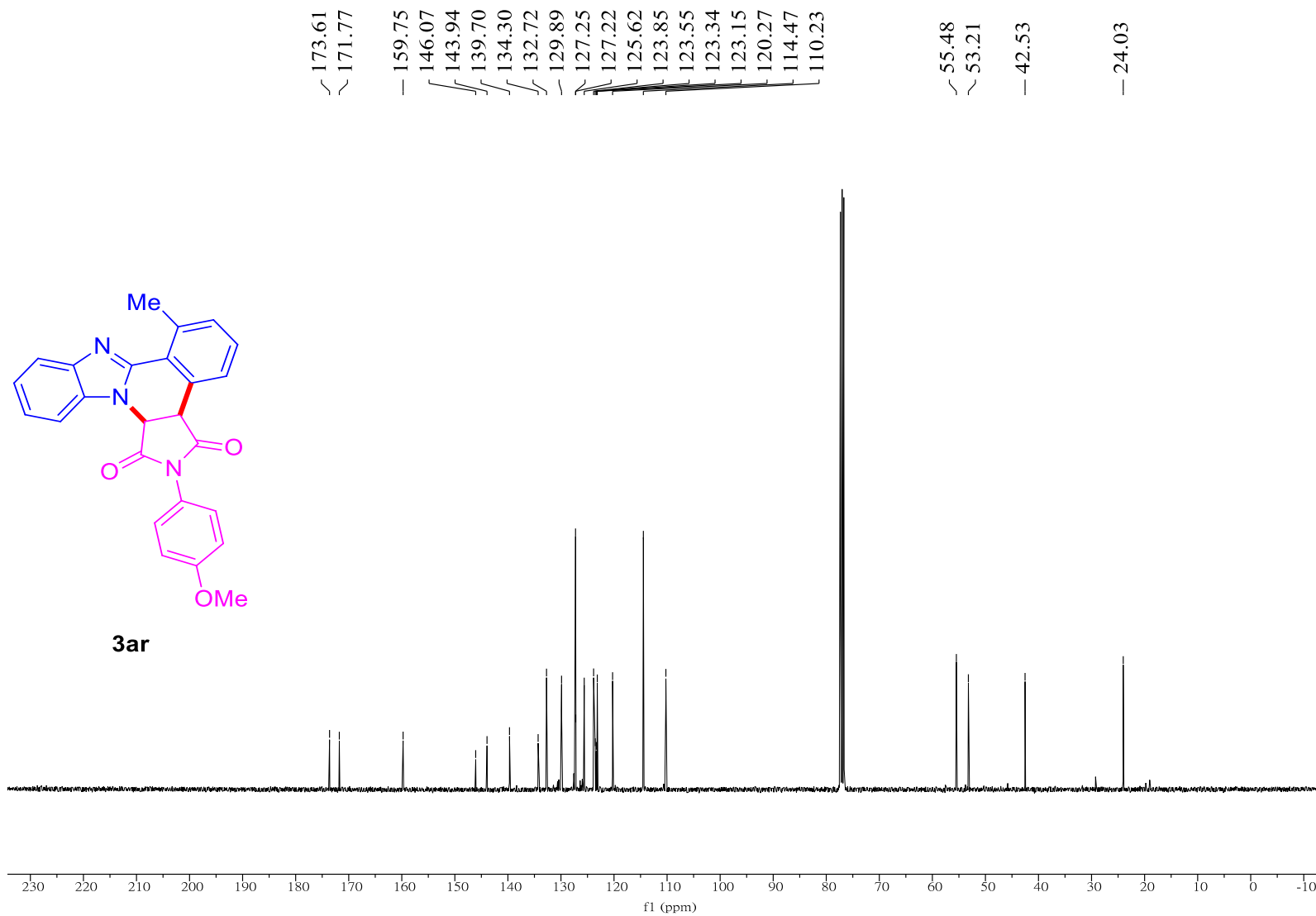
## Display Report

Meas. $m/z$	#	Ion Formula	$m/z$	err [ppm]	mSigma	# Sigma	Score	rdb	$e^-$ Conf	N-Rule	Adduct
410.1502	1	$C_{25}H_{20}N_4O_3$	410.1499	-0.6	6.4	1	100.00	17.5	even	ok	M+H

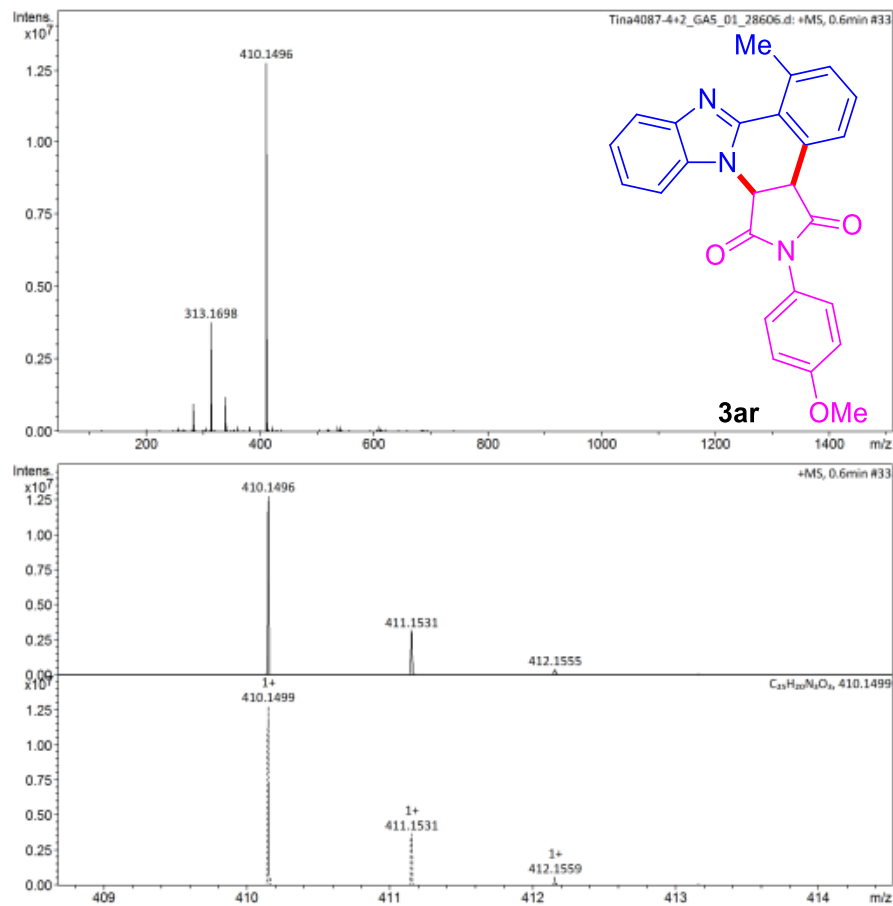
HRMS (ESI) of compound **5ar**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3ar** in  $\text{CDCl}_3$ .



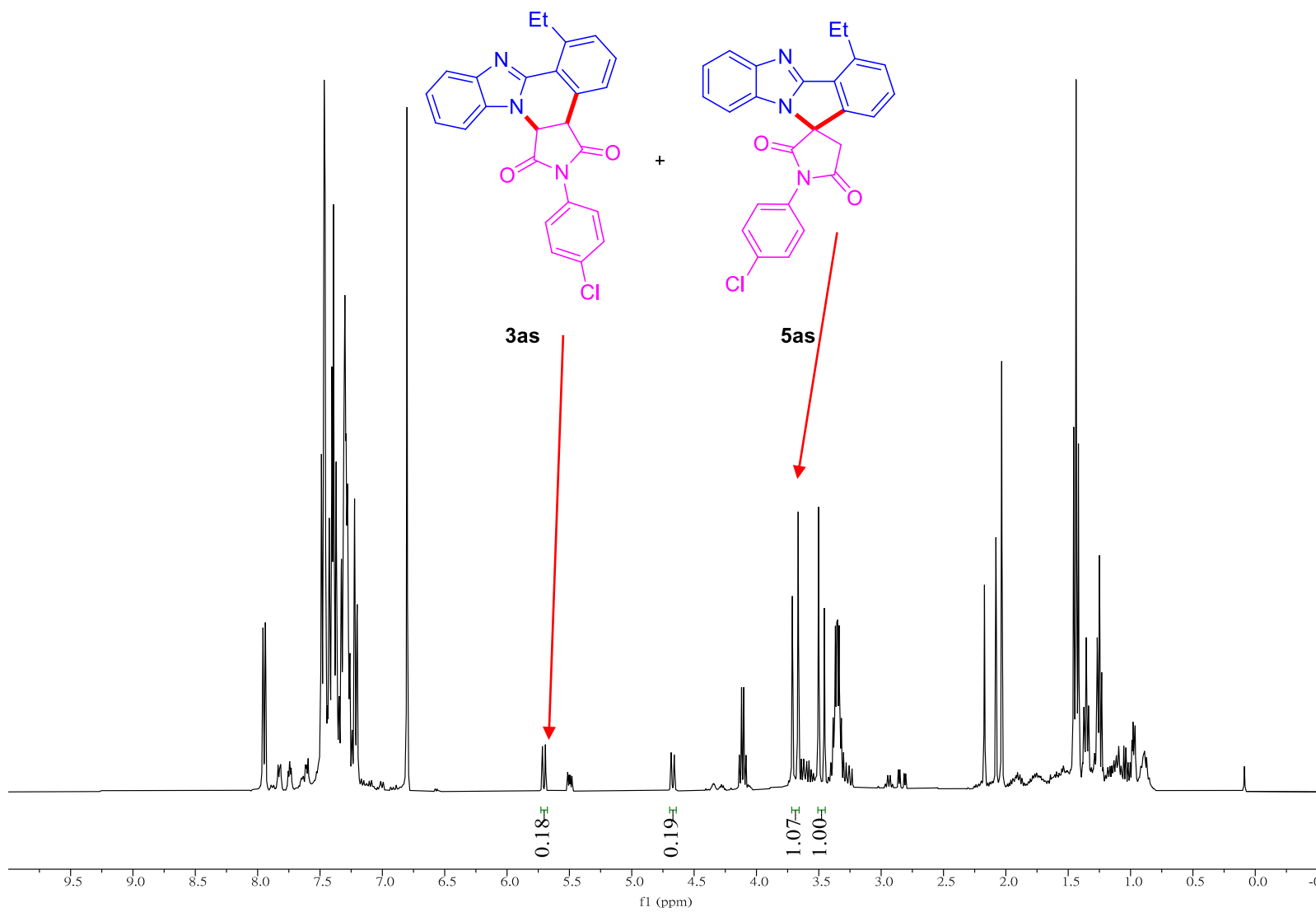
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3ar** in  $\text{CDCl}_3$ .



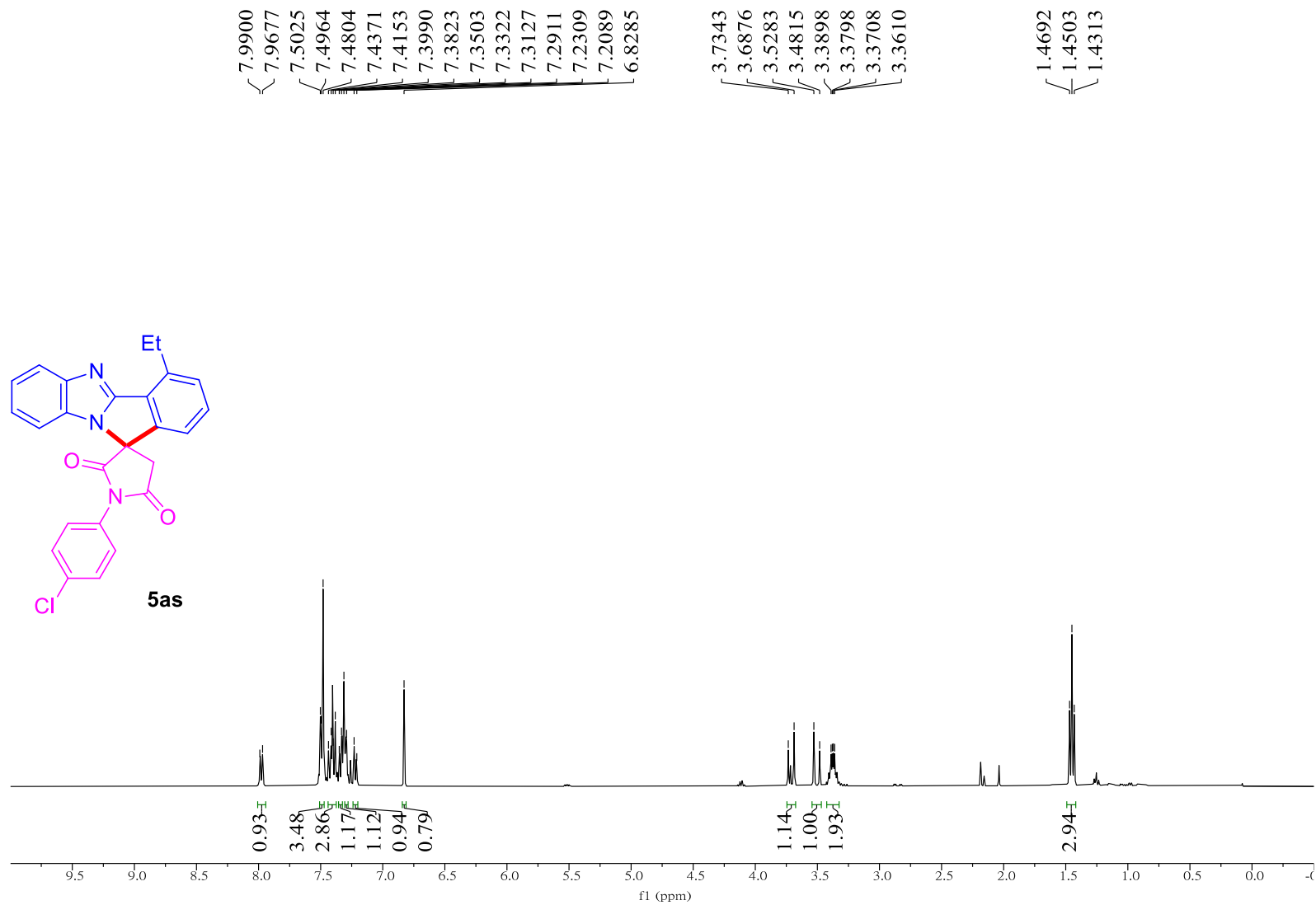
### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
410.1496	1	C <sub>25</sub> H <sub>20</sub> N <sub>3</sub> O <sub>3</sub>	410.1499	0.8	22.3	1	100.00	17.5	even	ok	M+H

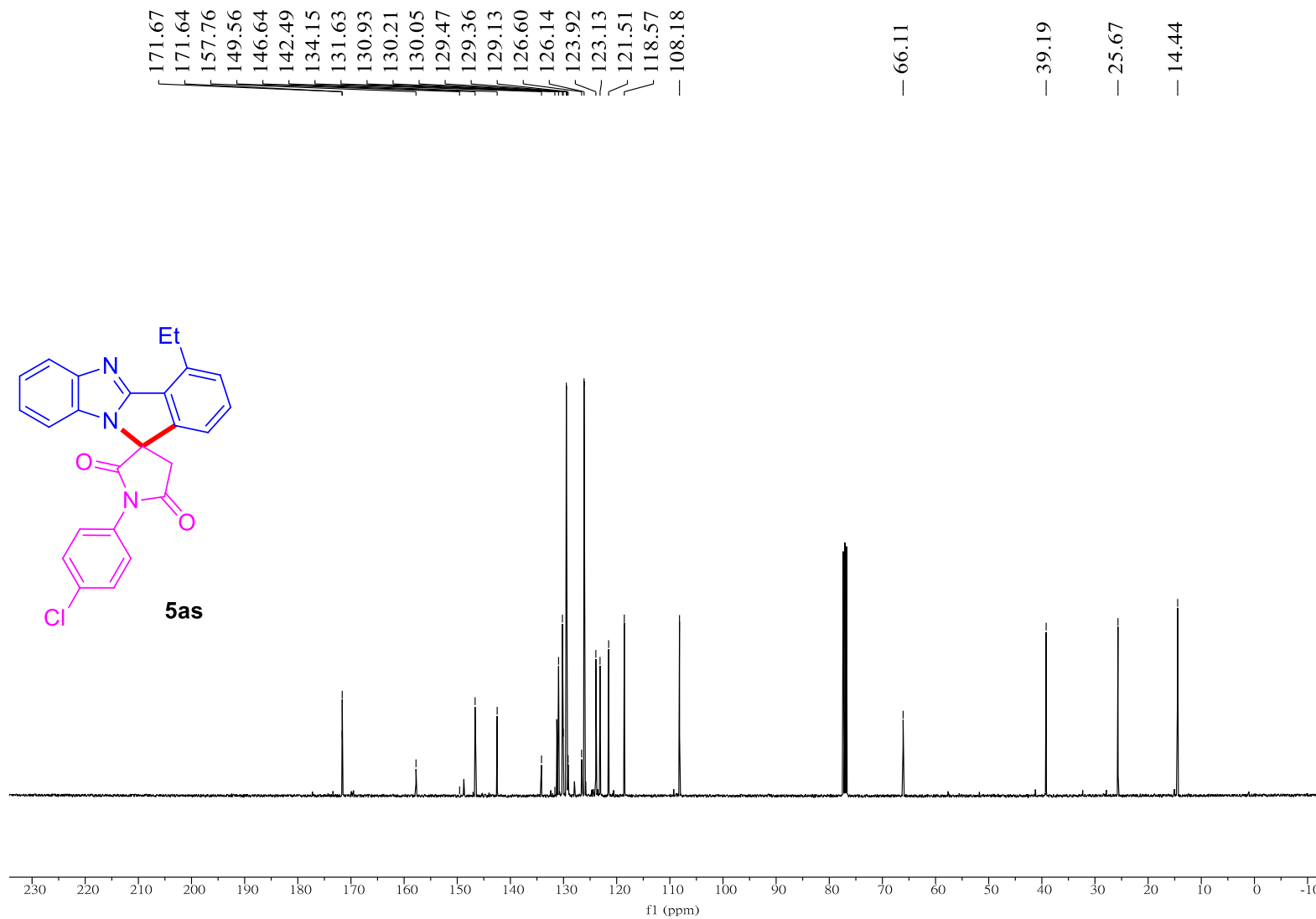
HRMS (ESI) of compound **3ar**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3as** and **5as** in  $\text{CDCl}_3$ .

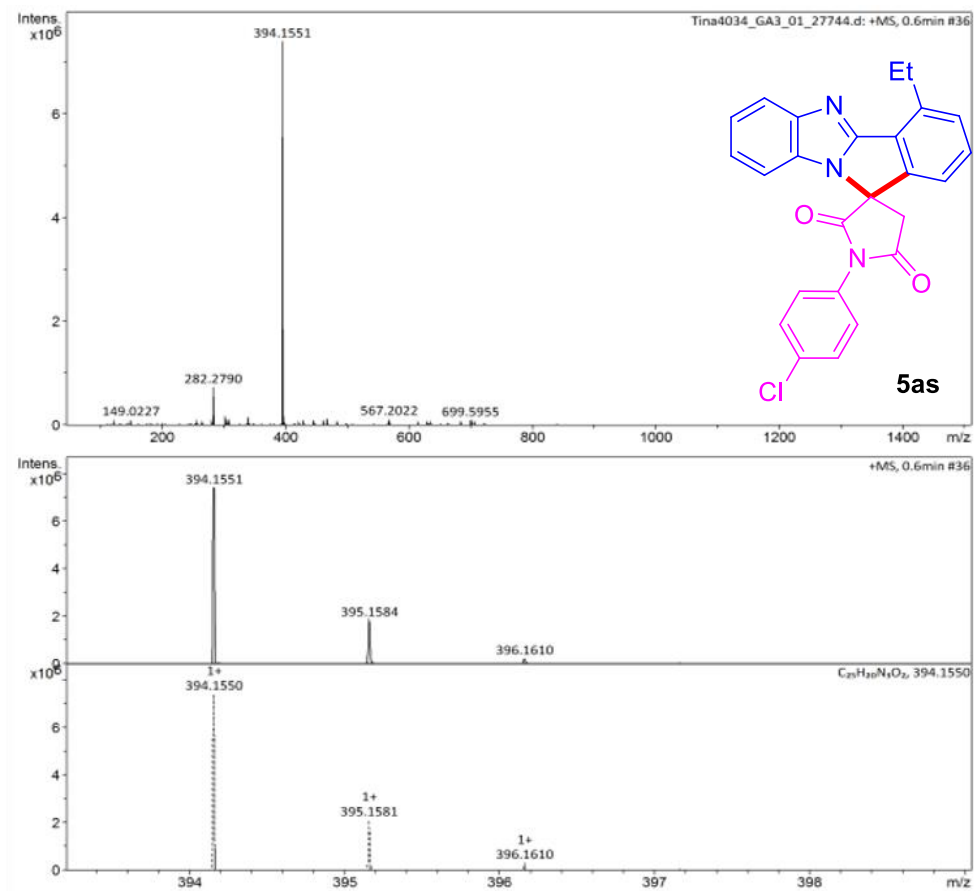


$^1\text{H}$  NMR spectrum (400 MHz) of compound **5as** in  $\text{CDCl}_3$ .



$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5as** in  $\text{CDCl}_3$ .

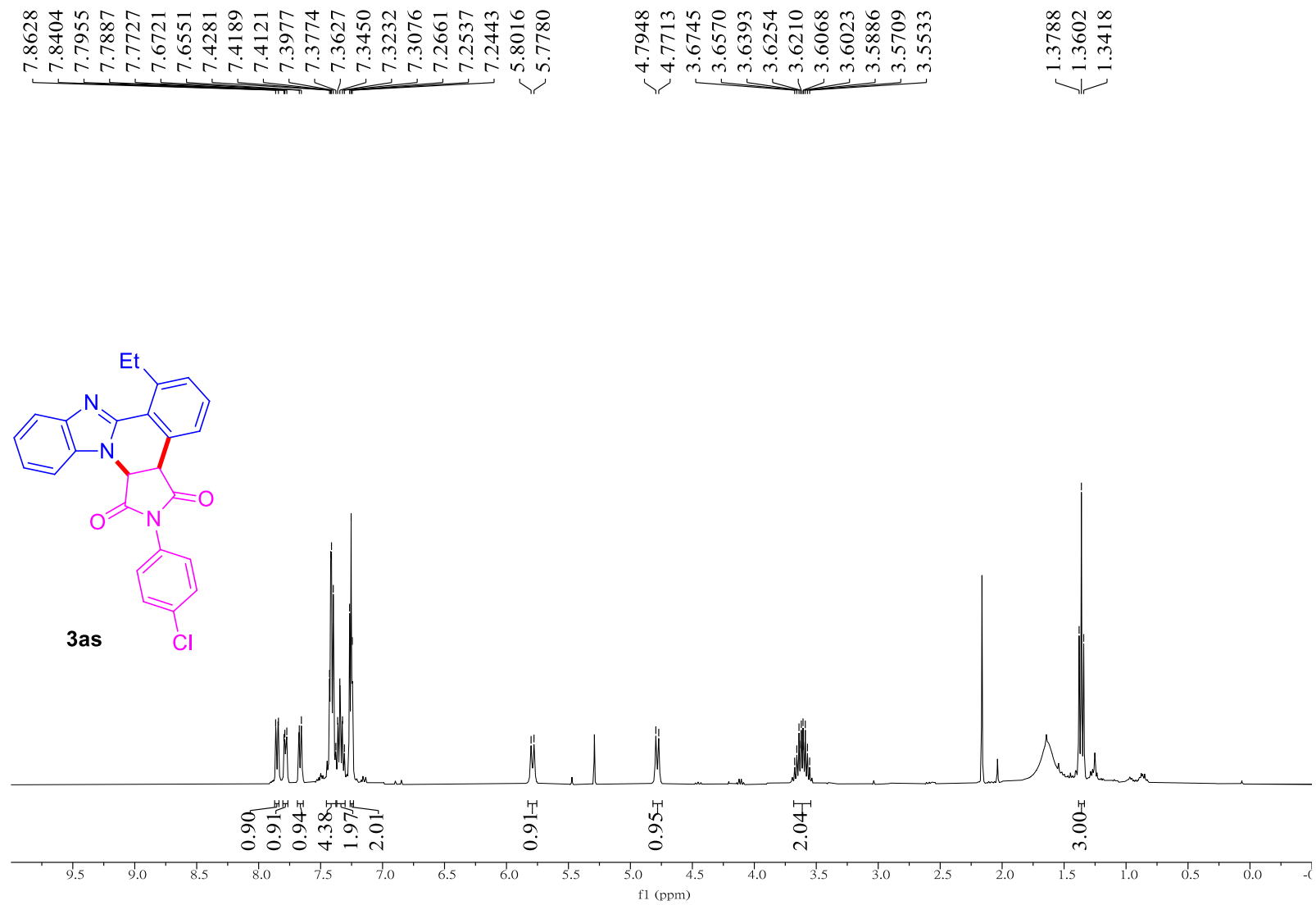




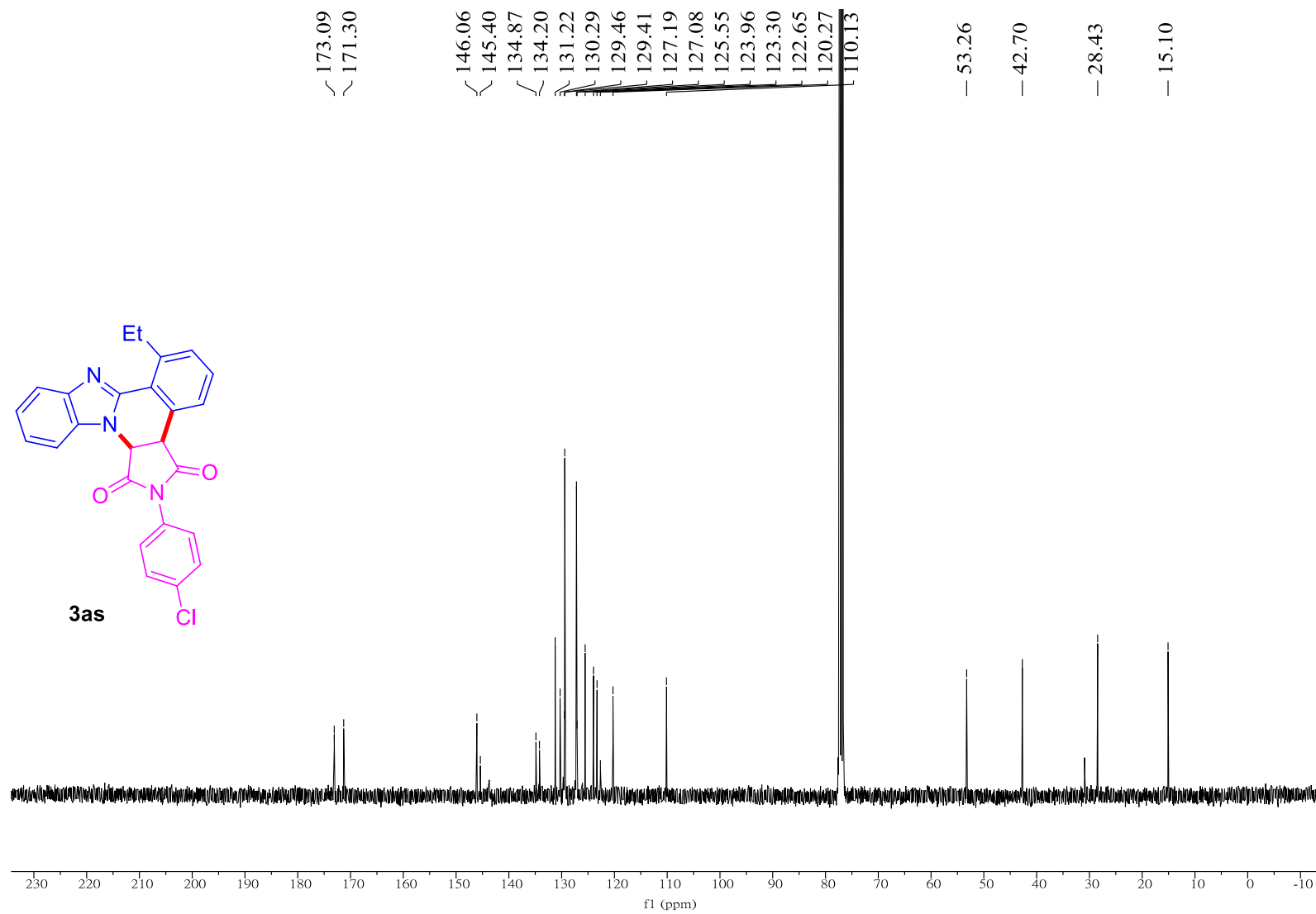
### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
394.1551	1	C <sub>25</sub> H <sub>20</sub> N <sub>4</sub> O <sub>2</sub>	394.1550	0.2	19.7	1	100.00	17.5	even	ok	M+H

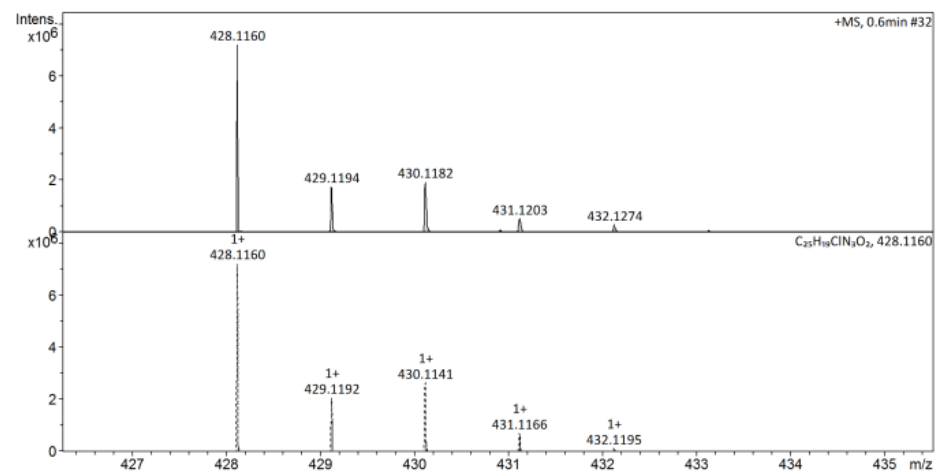
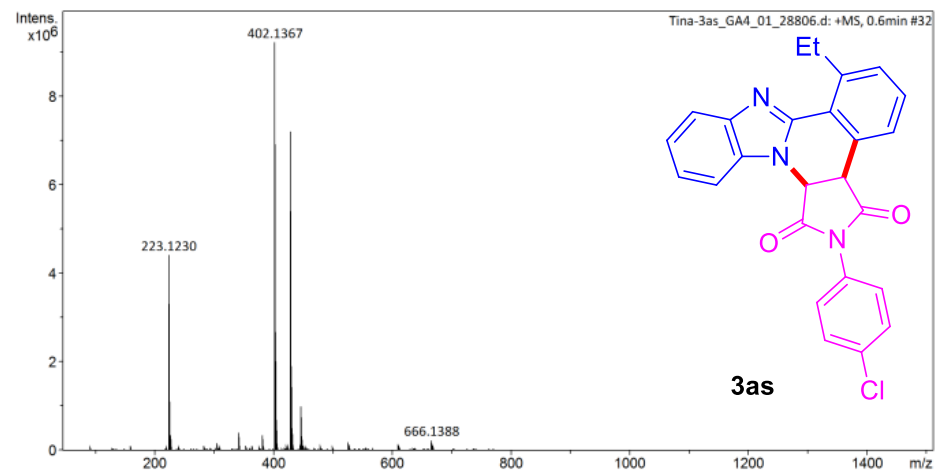
HRMS (ESI) of compound **5as**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3as** in  $\text{CDCl}_3$ .



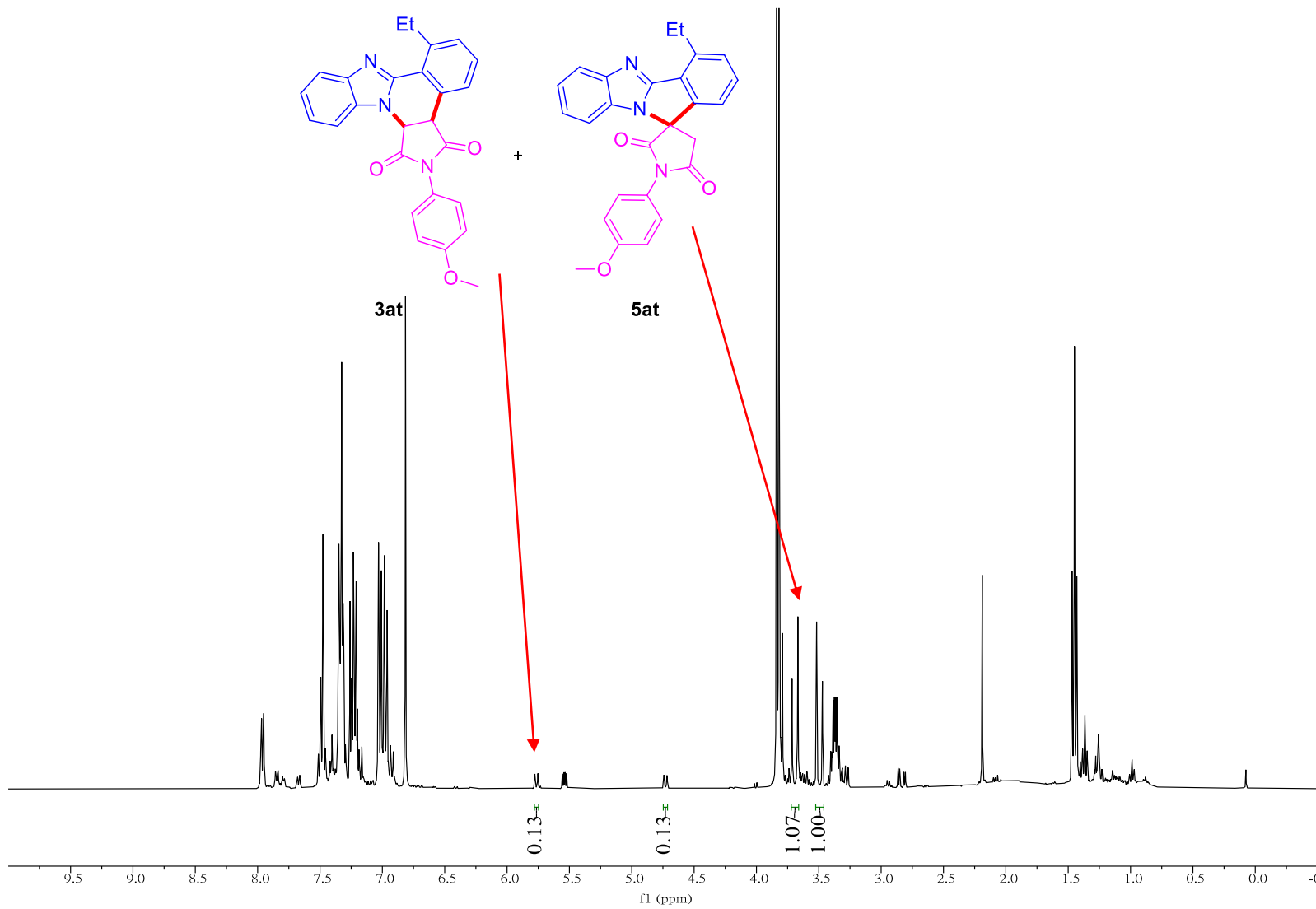
<sup>13</sup>C NMR spectrum (100 MHz) of compound **3as** in CDCl<sub>3</sub>.



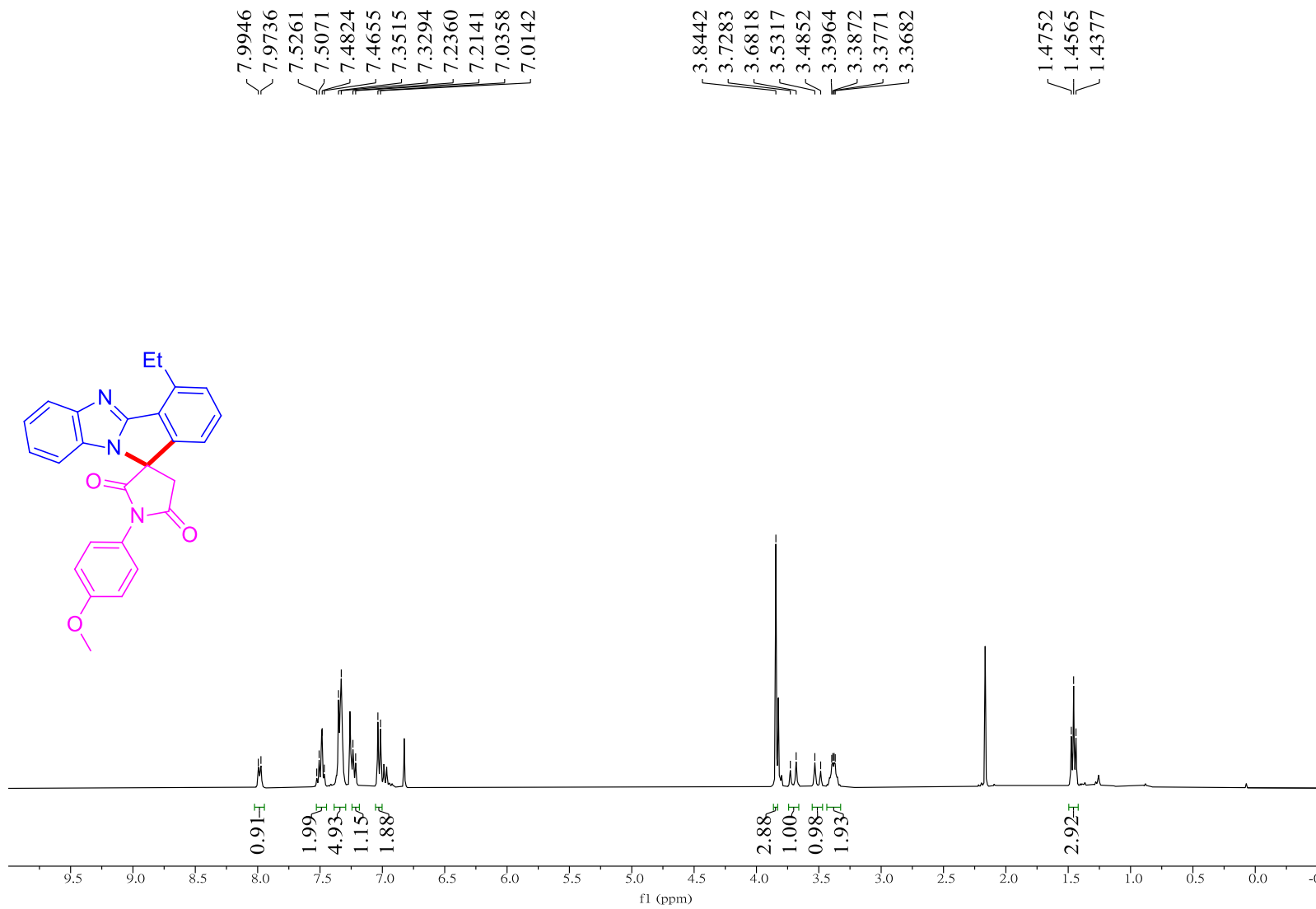
### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
428.1160	1	C <sub>25</sub> H <sub>19</sub> ClN <sub>3</sub> O <sub>2</sub>	428.1160	-0.0	50.2	1	100.00	17.5	even	ok	M+H

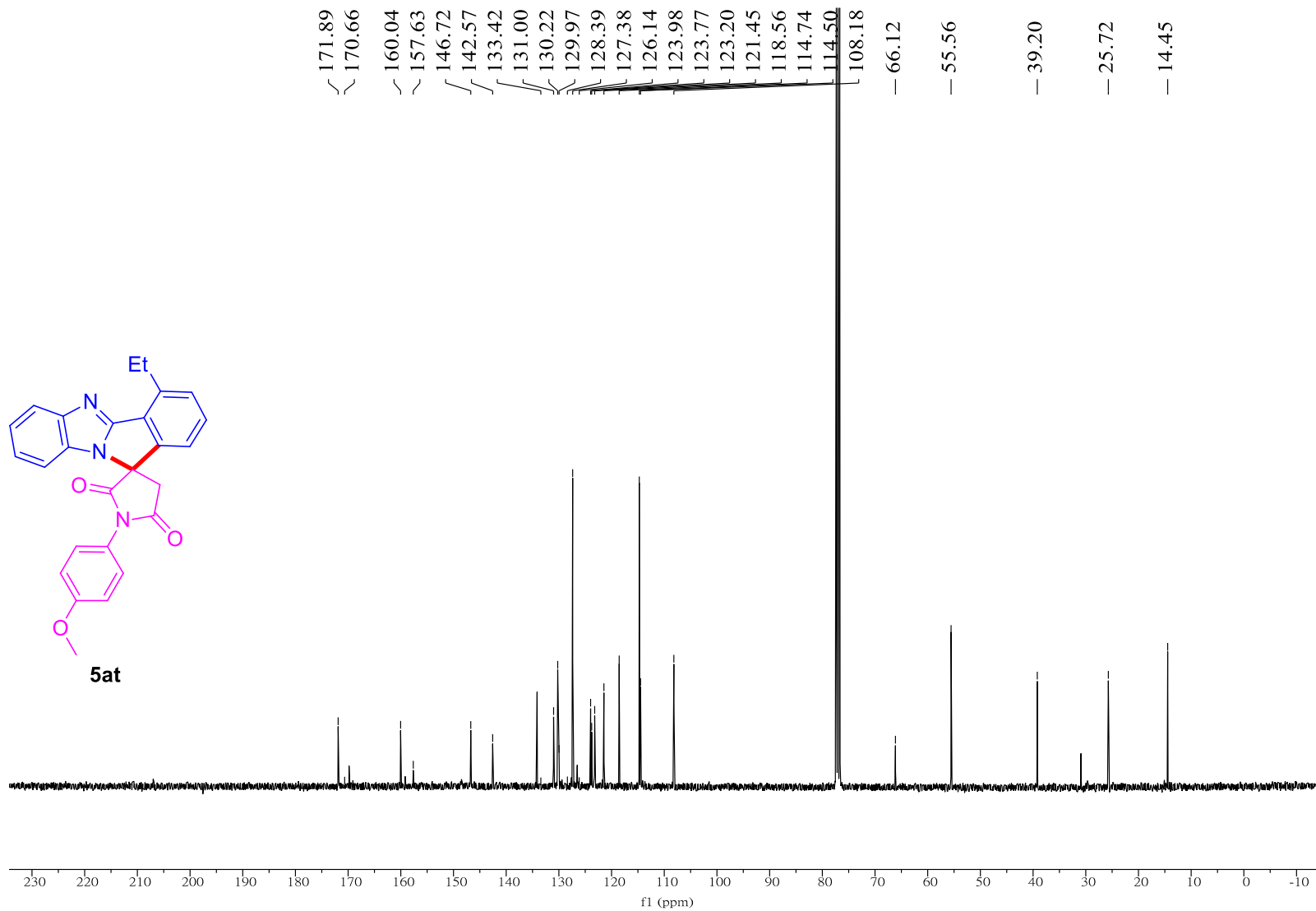
HRMS (ESI) of compound **3as**.



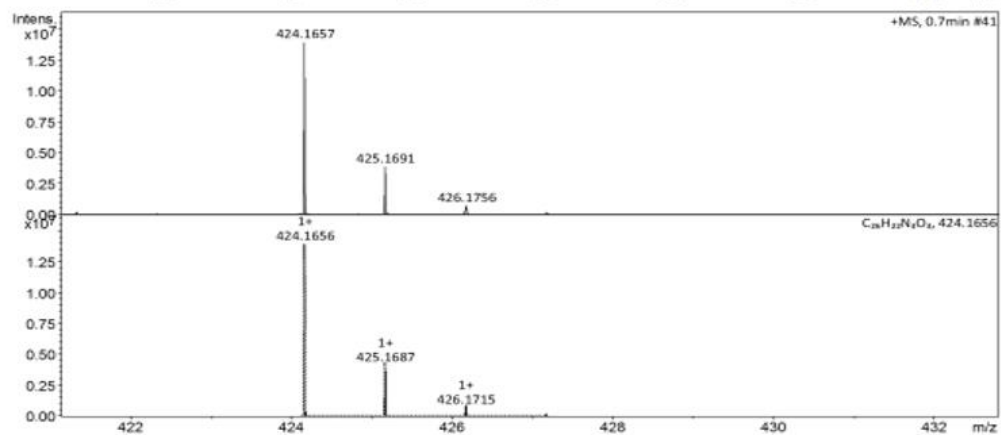
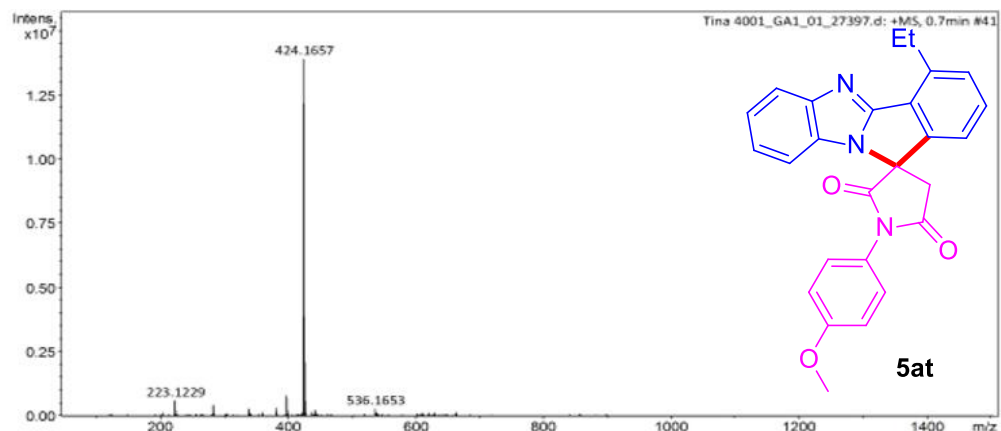
$^1\text{H}$  NMR spectrum (400 MHz) of compound **3at** and **5at** in  $\text{CDCl}_3$ .



<sup>1</sup>H NMR spectrum (400 MHz) of compound **5at** in CDCl<sub>3</sub>.



$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5at** in  $\text{CDCl}_3$ .

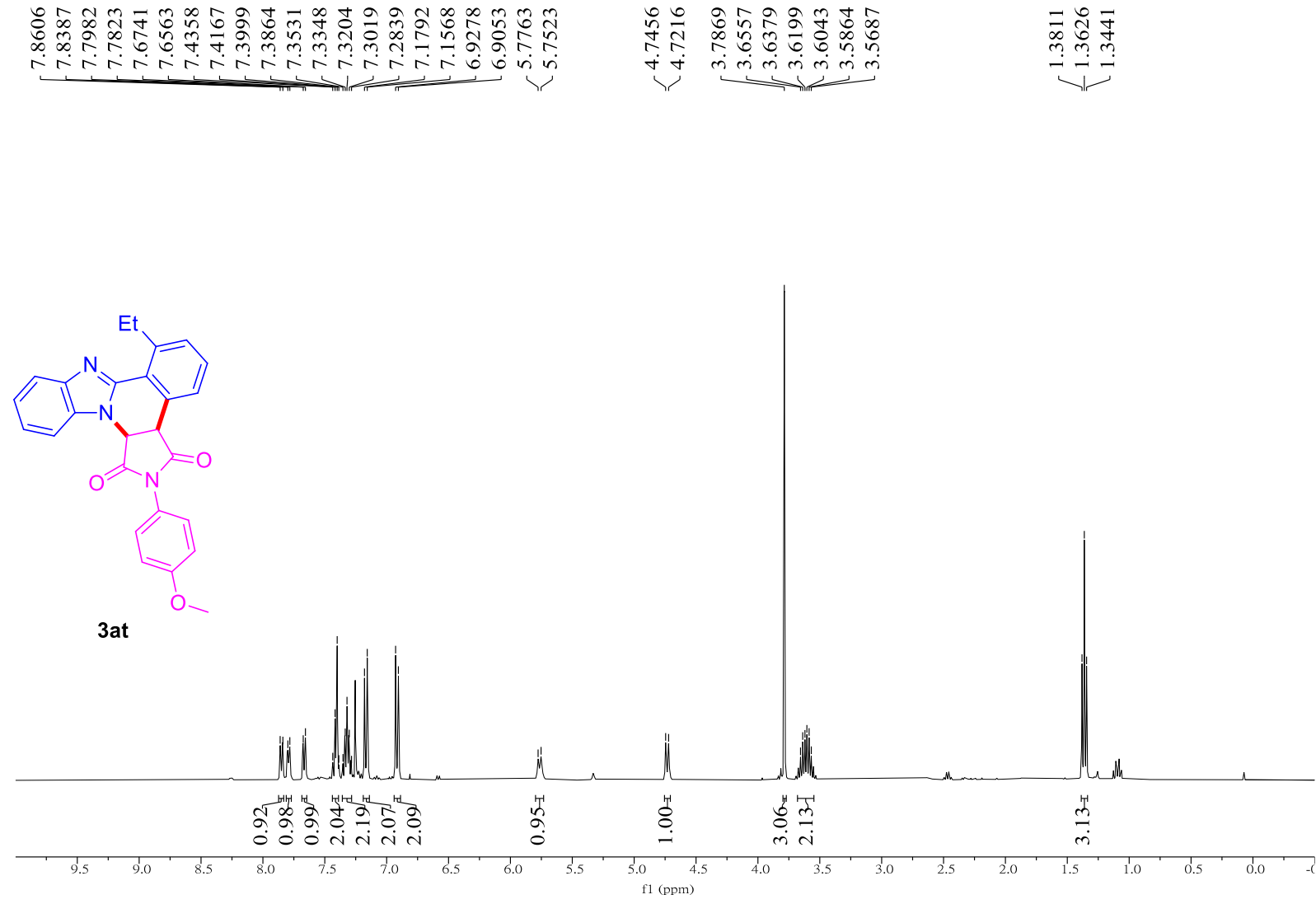


### Display Report

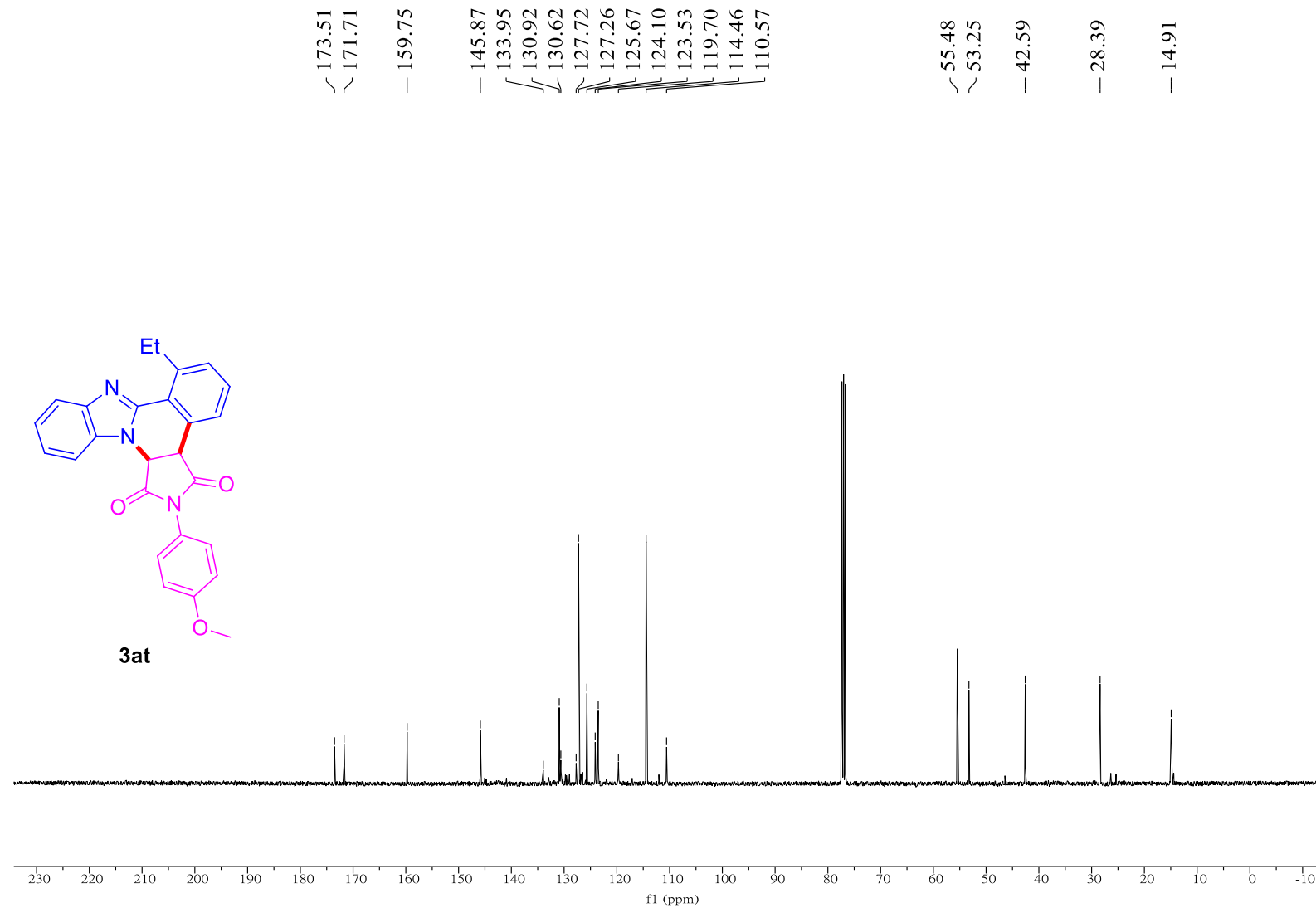
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
424.1657	1	C <sub>28</sub> H <sub>22</sub> N <sub>3</sub> O <sub>3</sub>	424.1656	0.4	8.9	1	100.00	17.5	even	ok	M+H

HRMS (ESI) of compound **5at**.

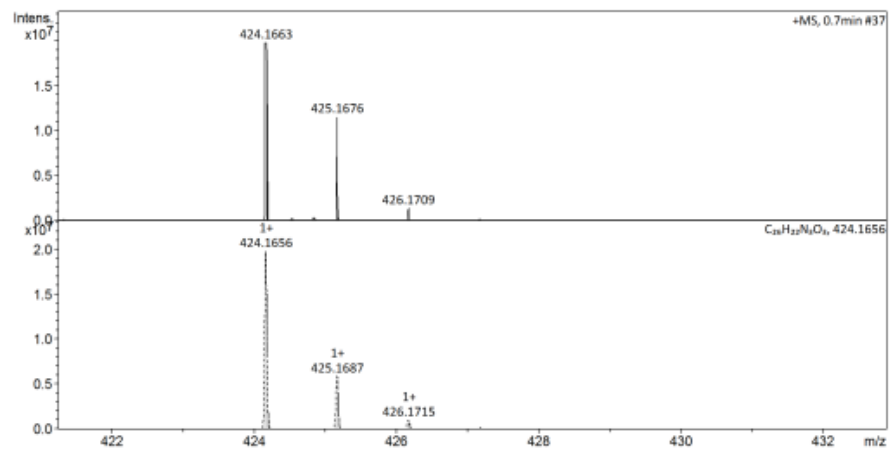
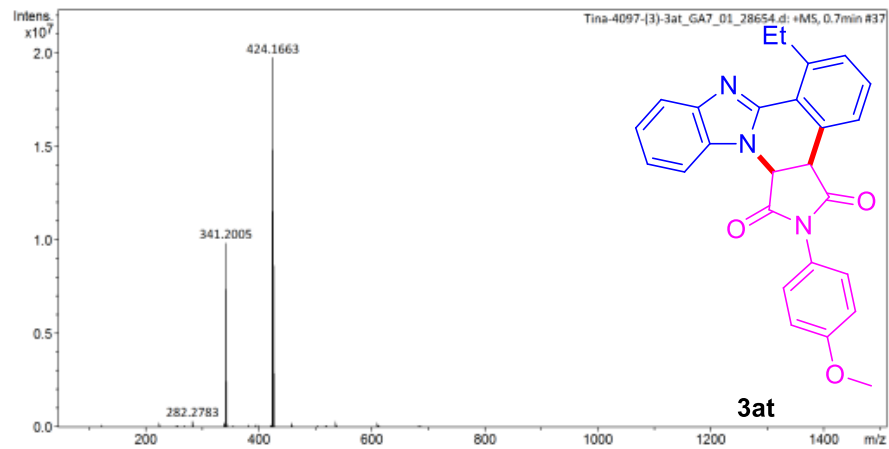




<sup>1</sup>H NMR spectrum (400 MHz) of compound **3at** in CDCl<sub>3</sub>.



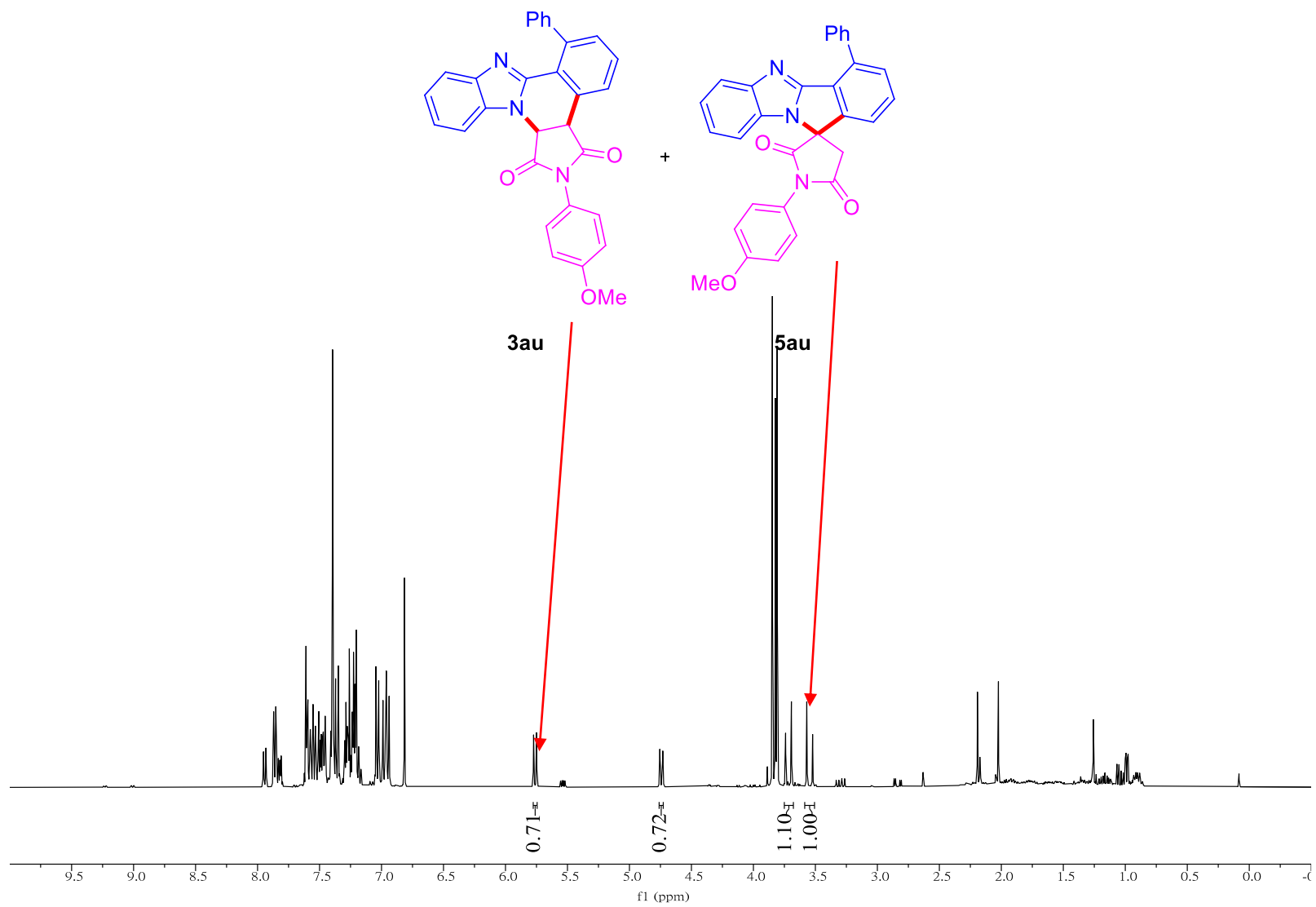
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3at** in  $\text{CDCl}_3$ .



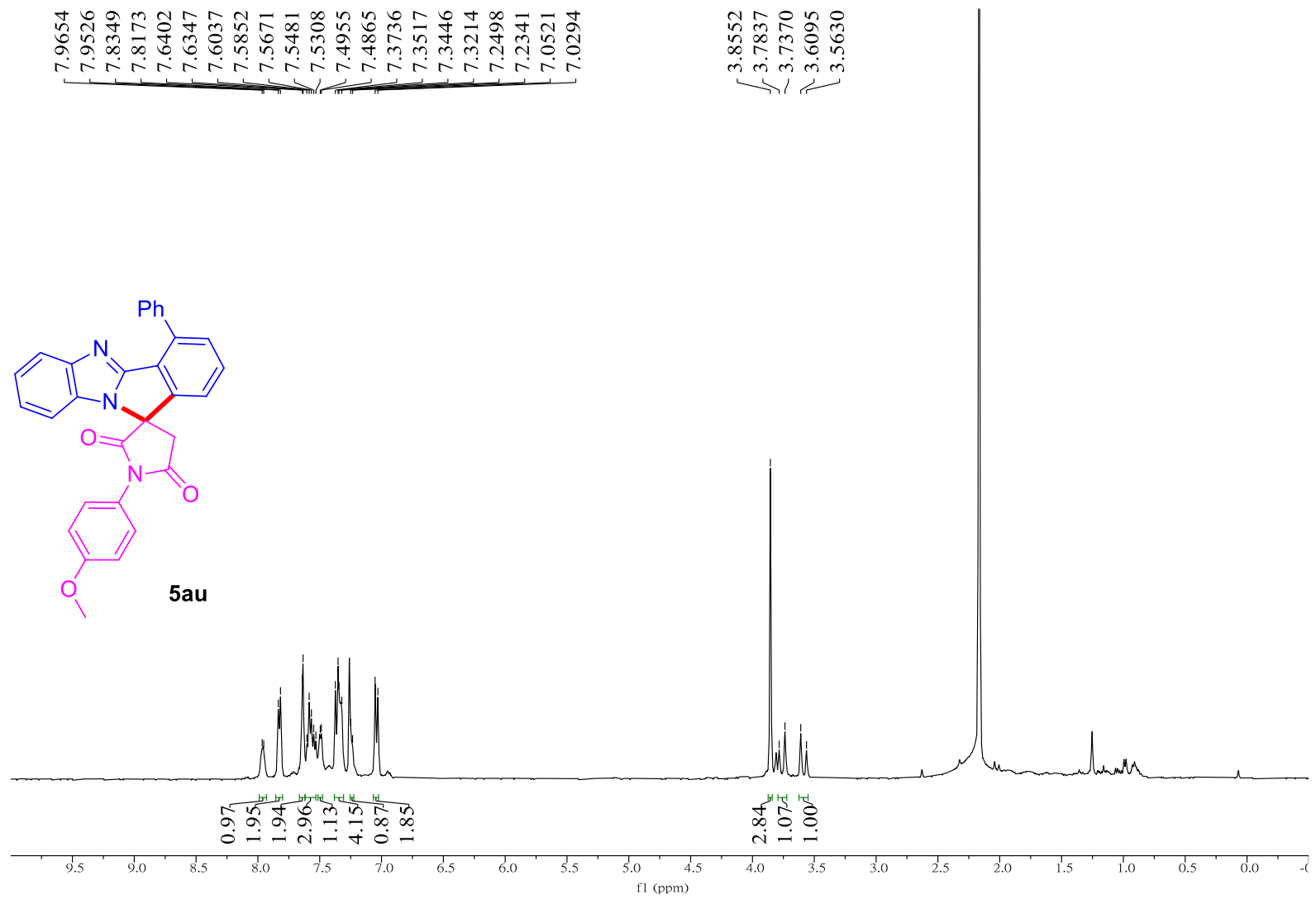
### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
424.1663	1	C <sub>26</sub> H <sub>22</sub> N <sub>3</sub> O <sub>3</sub>	424.1656	-1.7	143.9	1	100.00	17.5	even	ok	M+H

HRMS (ESI) of compound **3at**.

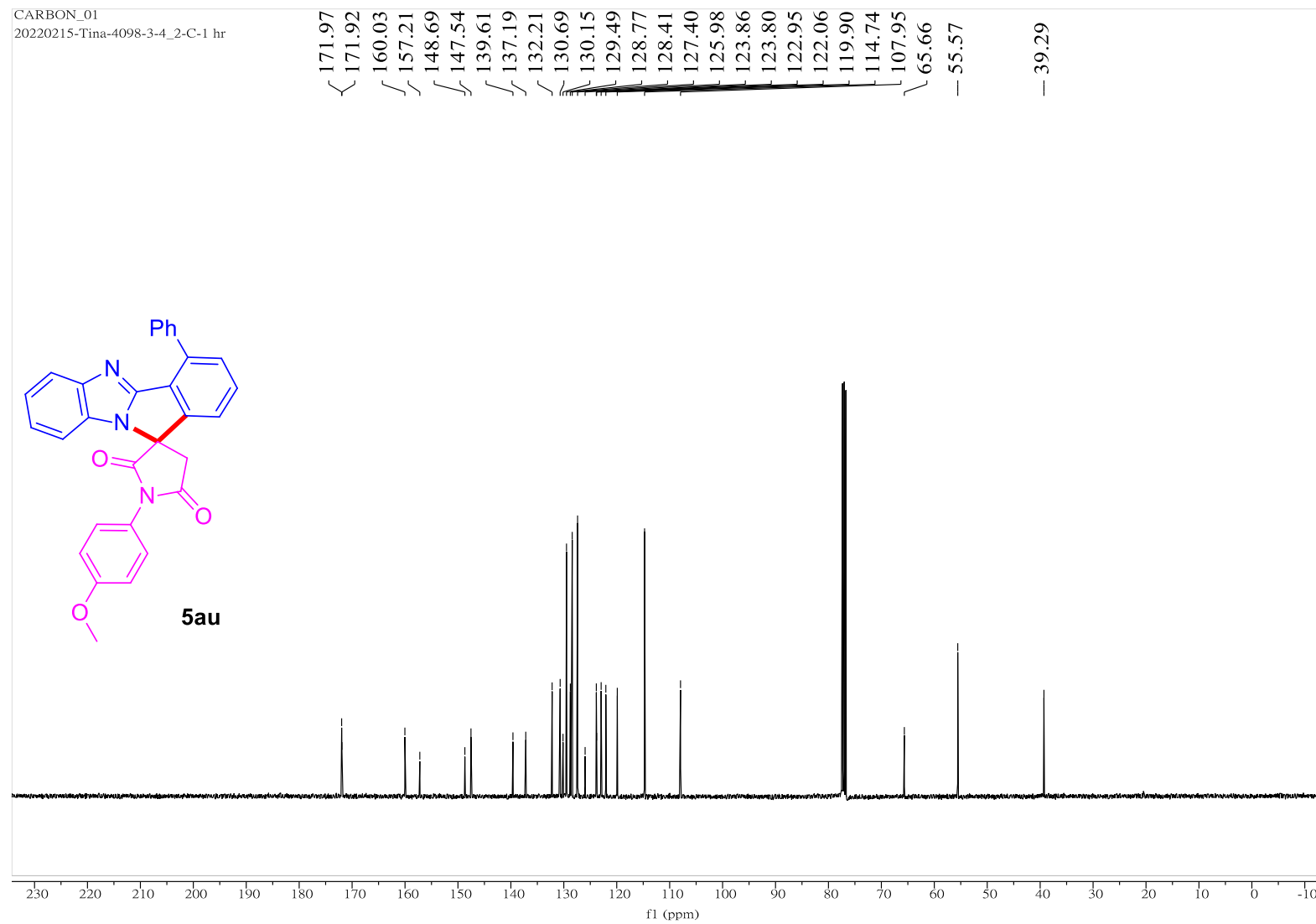


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3au** and **5au** in  $\text{CDCl}_3$ .

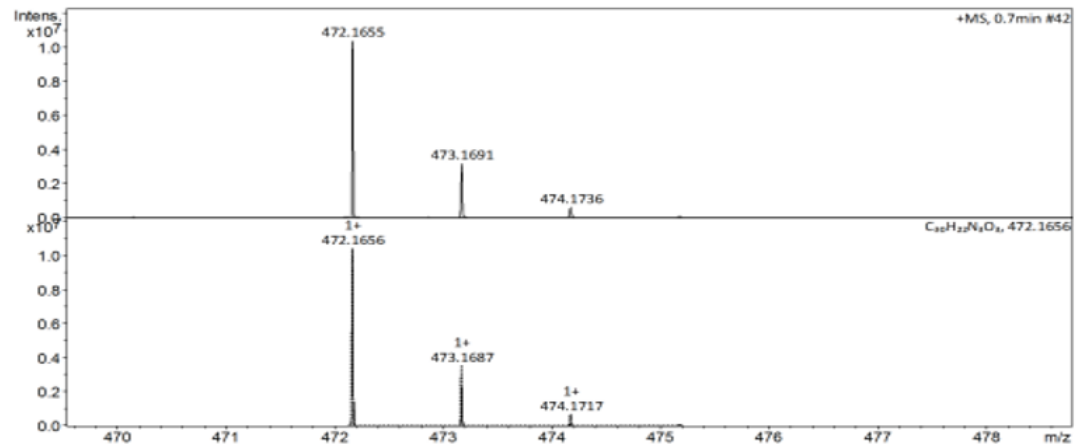
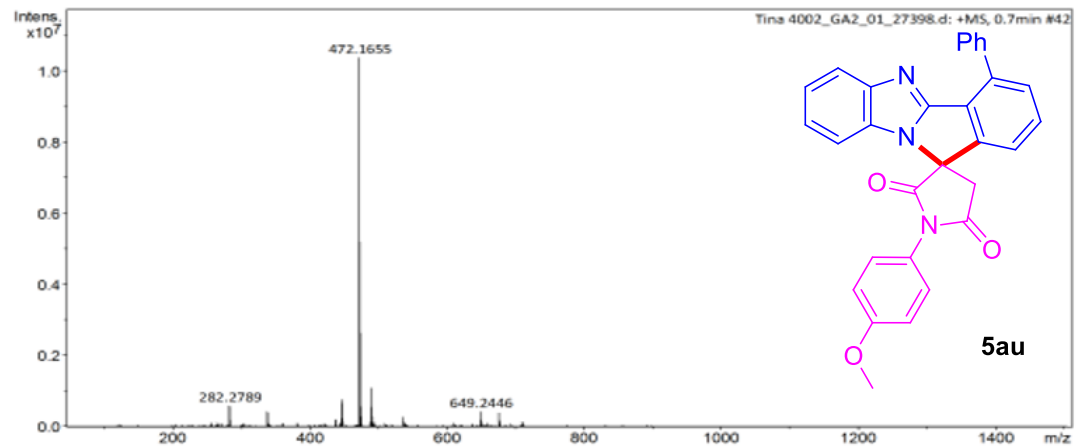


<sup>1</sup>H NMR spectrum (400 MHz) of compound **5au** in CDCl<sub>3</sub>.

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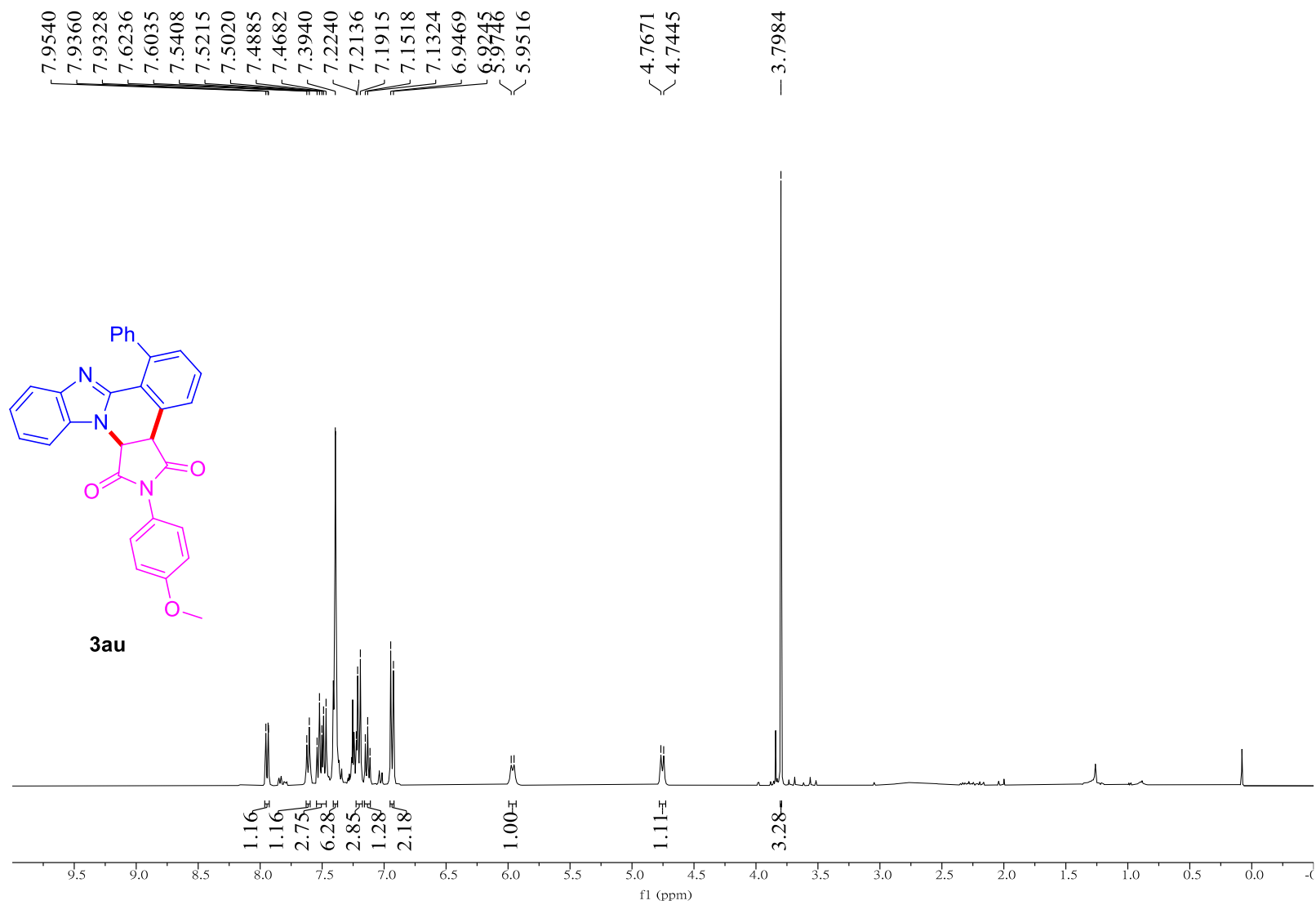
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5au** in  $\text{CDCl}_3$ .



### Display Report

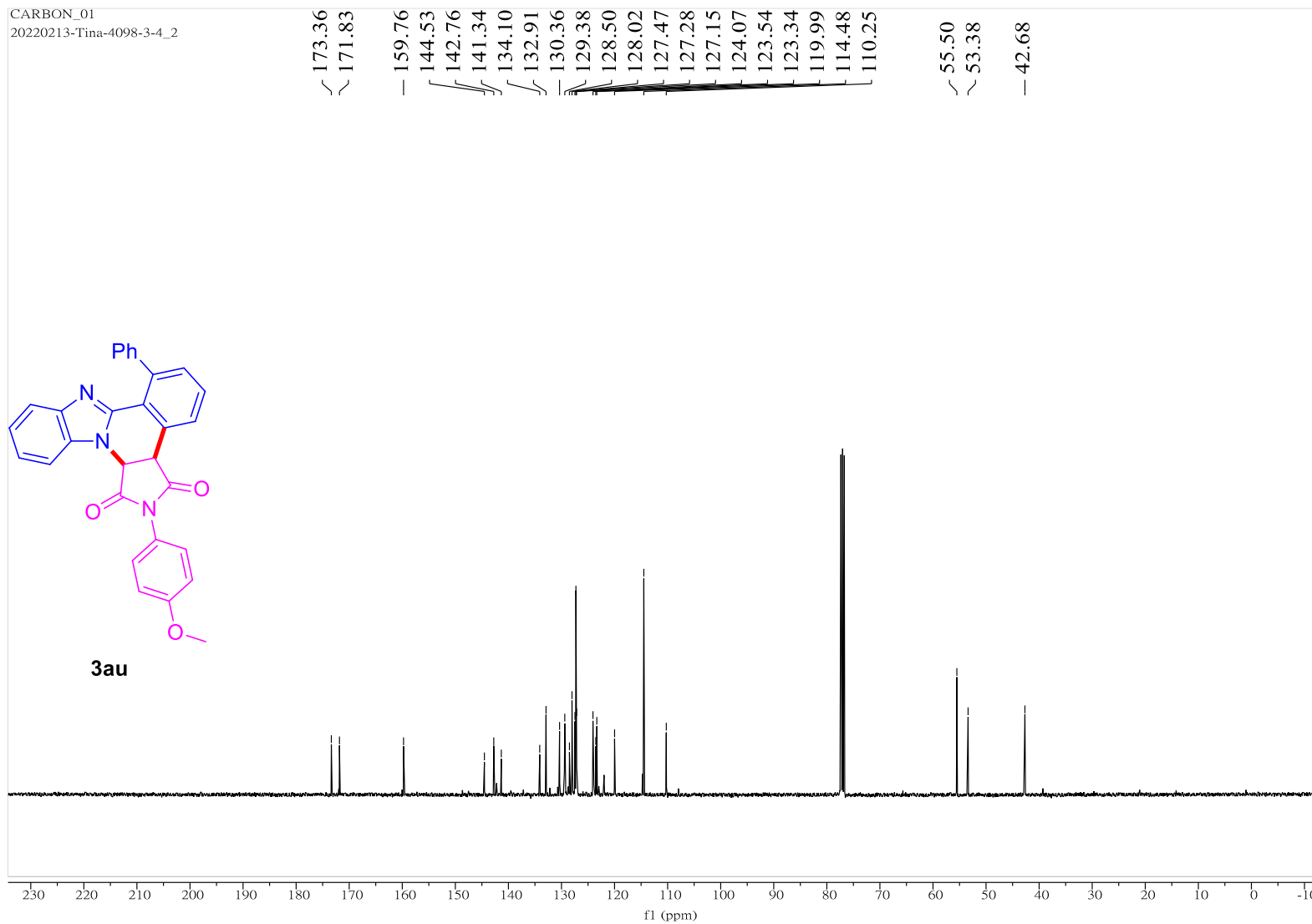
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
472.1655	1	C <sub>30</sub> H <sub>22</sub> N <sub>3</sub> O <sub>3</sub>	472.1656	-0.1	17.7	1	100.00	21.5	even	ok	M+H

HRMS (ESI) of compound **5au**.

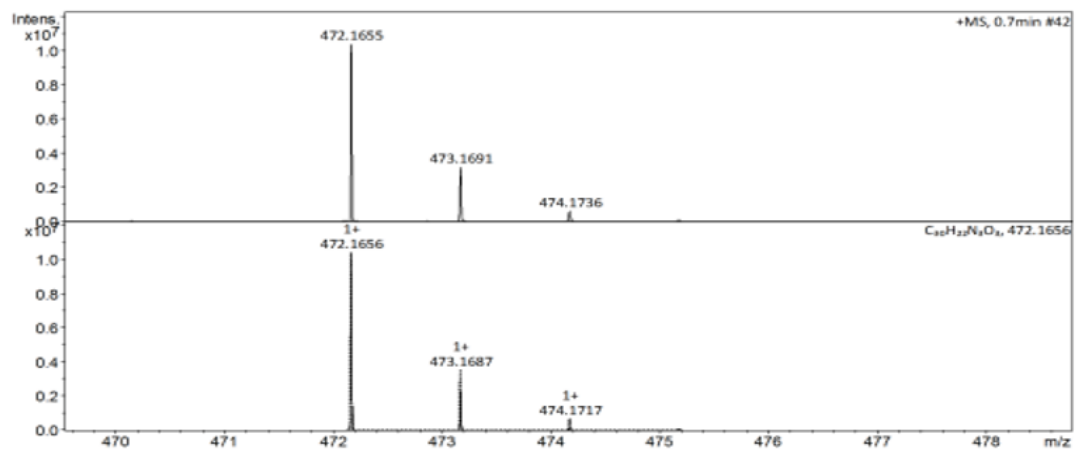
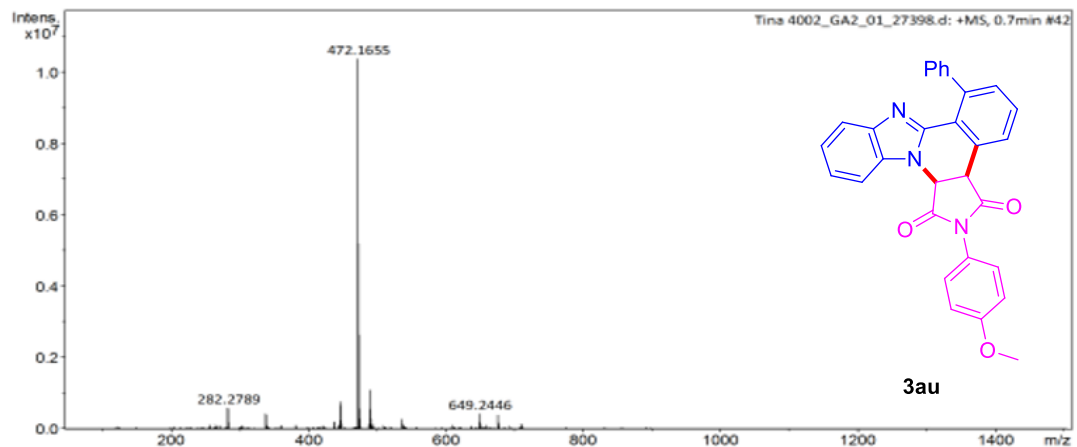


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3au** in  $\text{CDCl}_3$ .





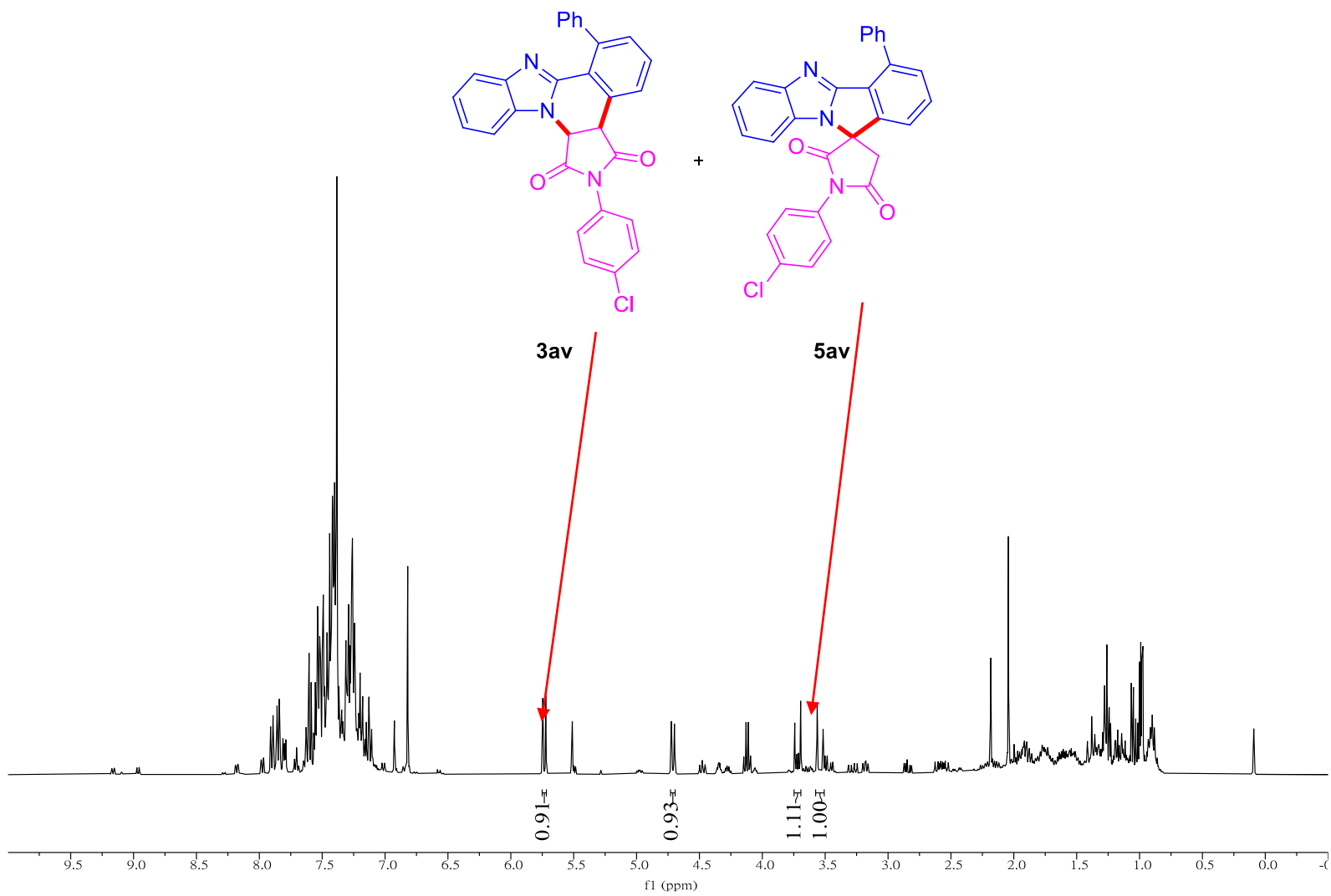
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3au** in  $\text{CDCl}_3$ .



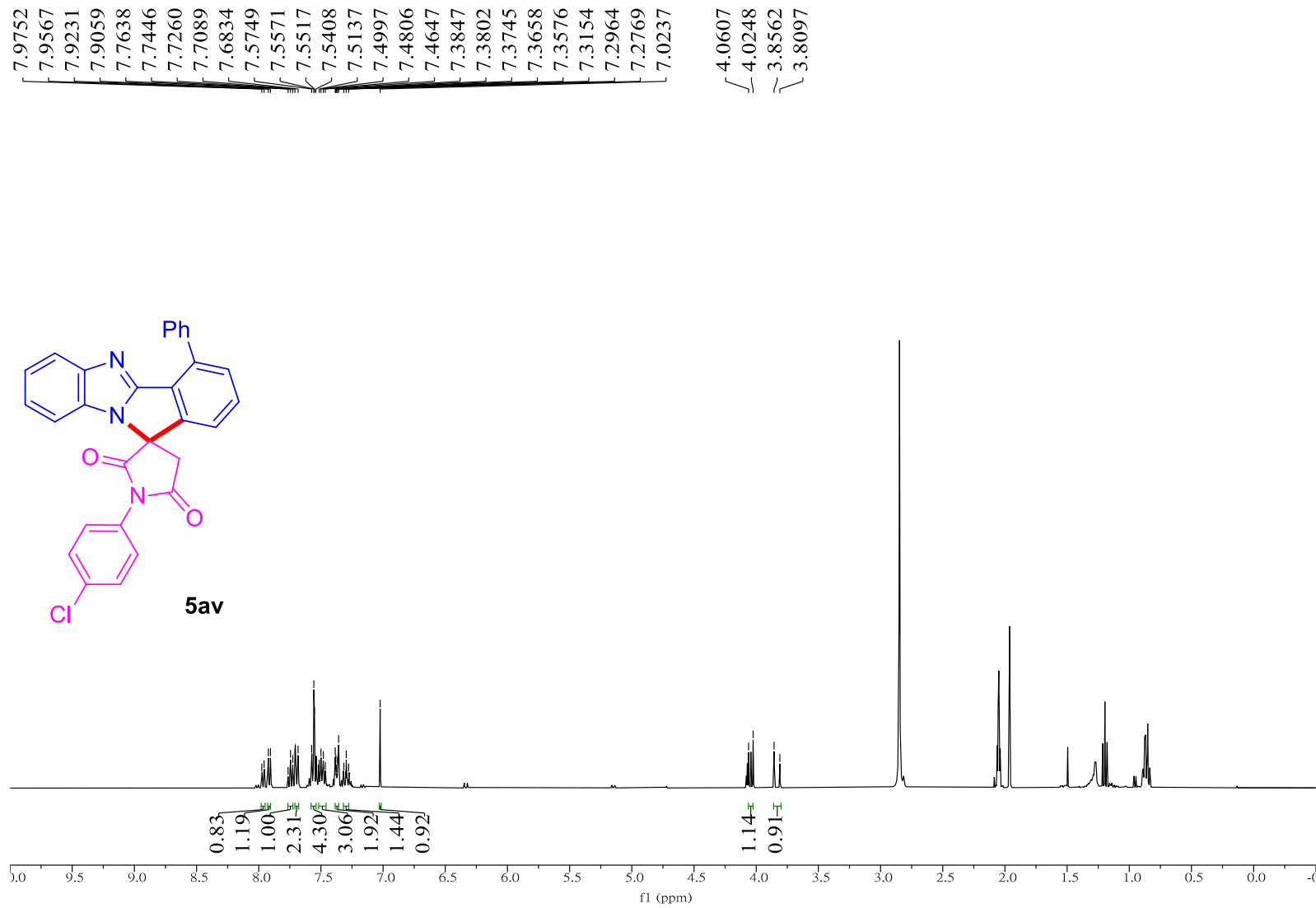
### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
472.1655	1	C <sub>30</sub> H <sub>22</sub> N <sub>3</sub> O <sub>3</sub>	472.1656	-0.1	17.7	1	100.00	21.5	even	ok	M+H

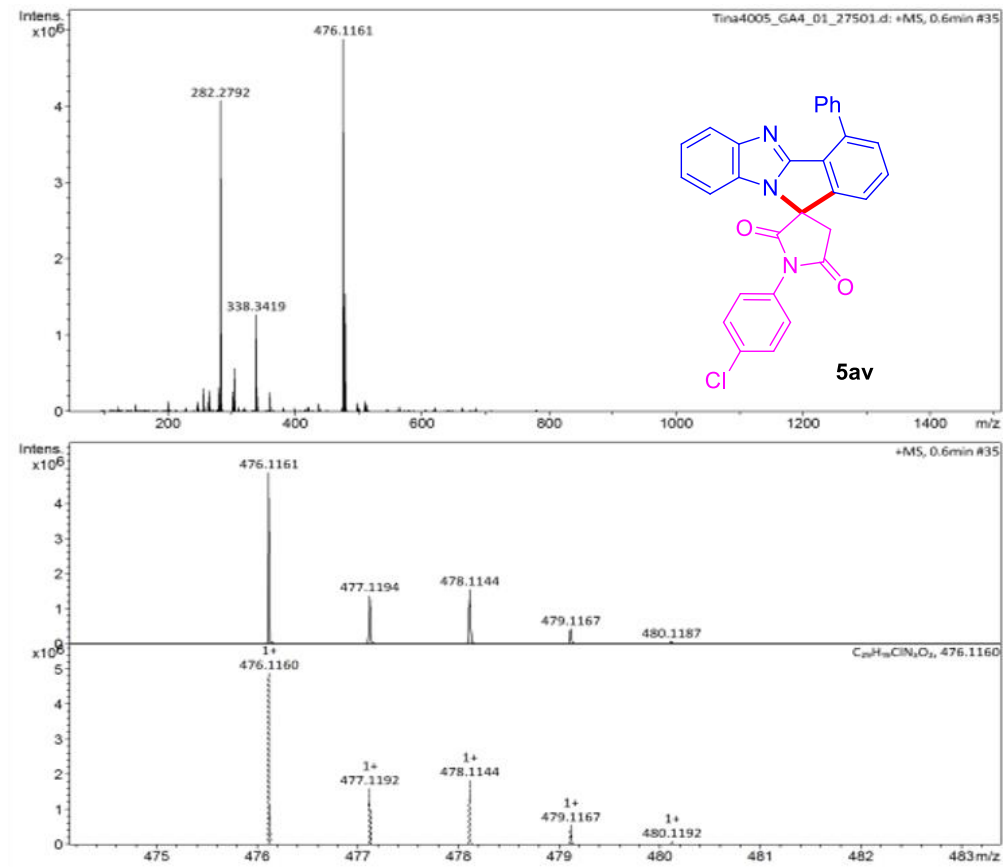
HRMS (ESI) of compound **3au**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3av** and **5av** in  $\text{CDCl}_3$ .



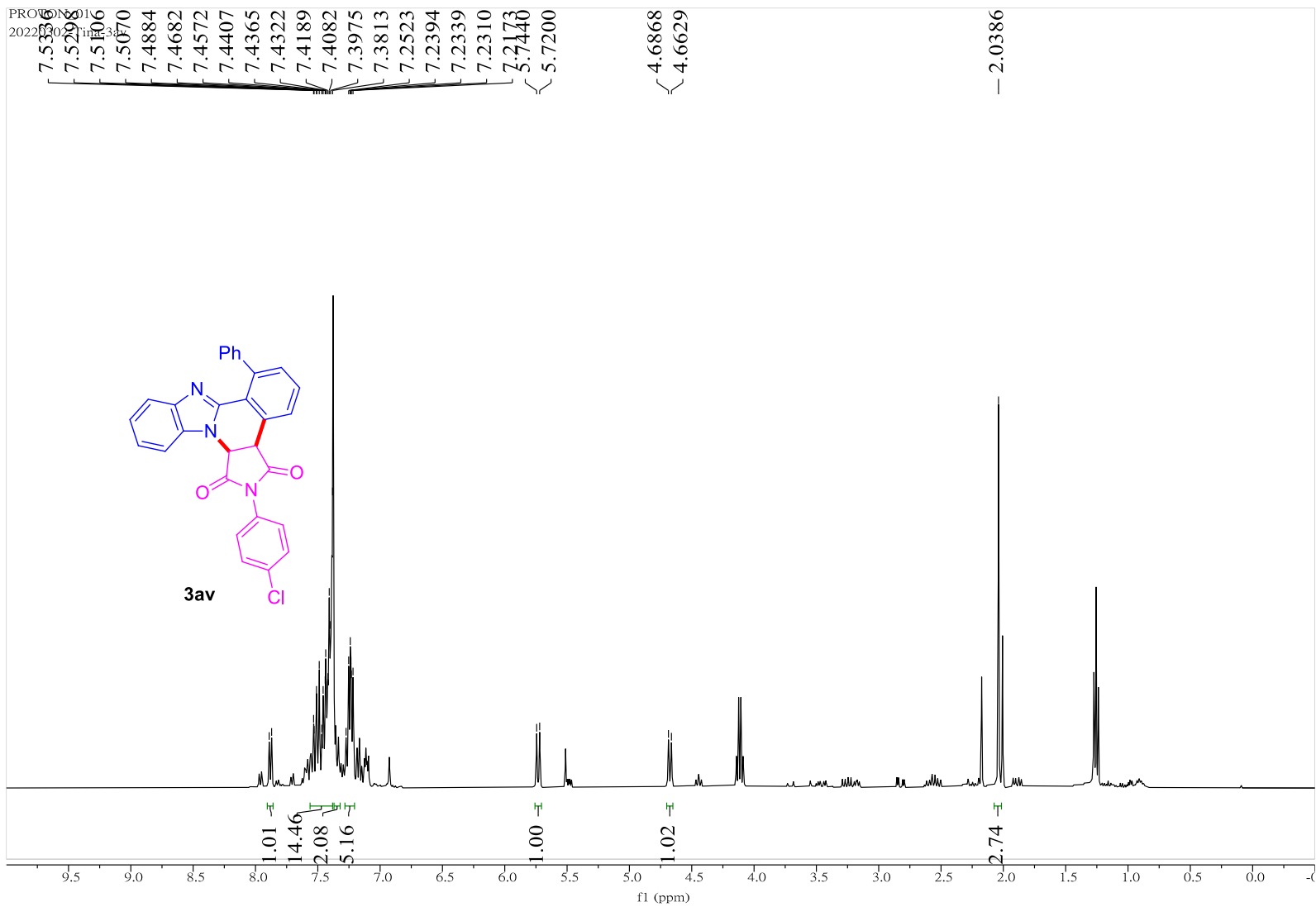
$^1\text{H}$  NMR spectrum (400 MHz) of compound **5av** in  $\text{CDCl}_3$ .



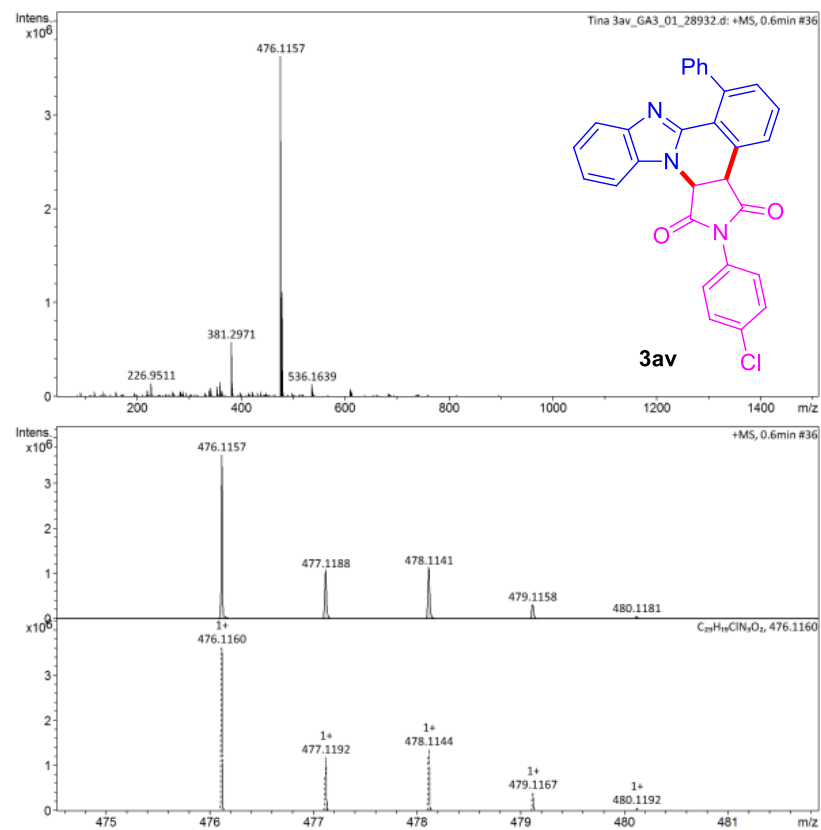
### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
476.1161	1	C <sub>29</sub> H <sub>19</sub> ClN <sub>3</sub> O <sub>2</sub>	476.1160	-0.1	35.2	1	100.00	21.5	even	ok	M+H

HRMS (ESI) of compound **5av**.



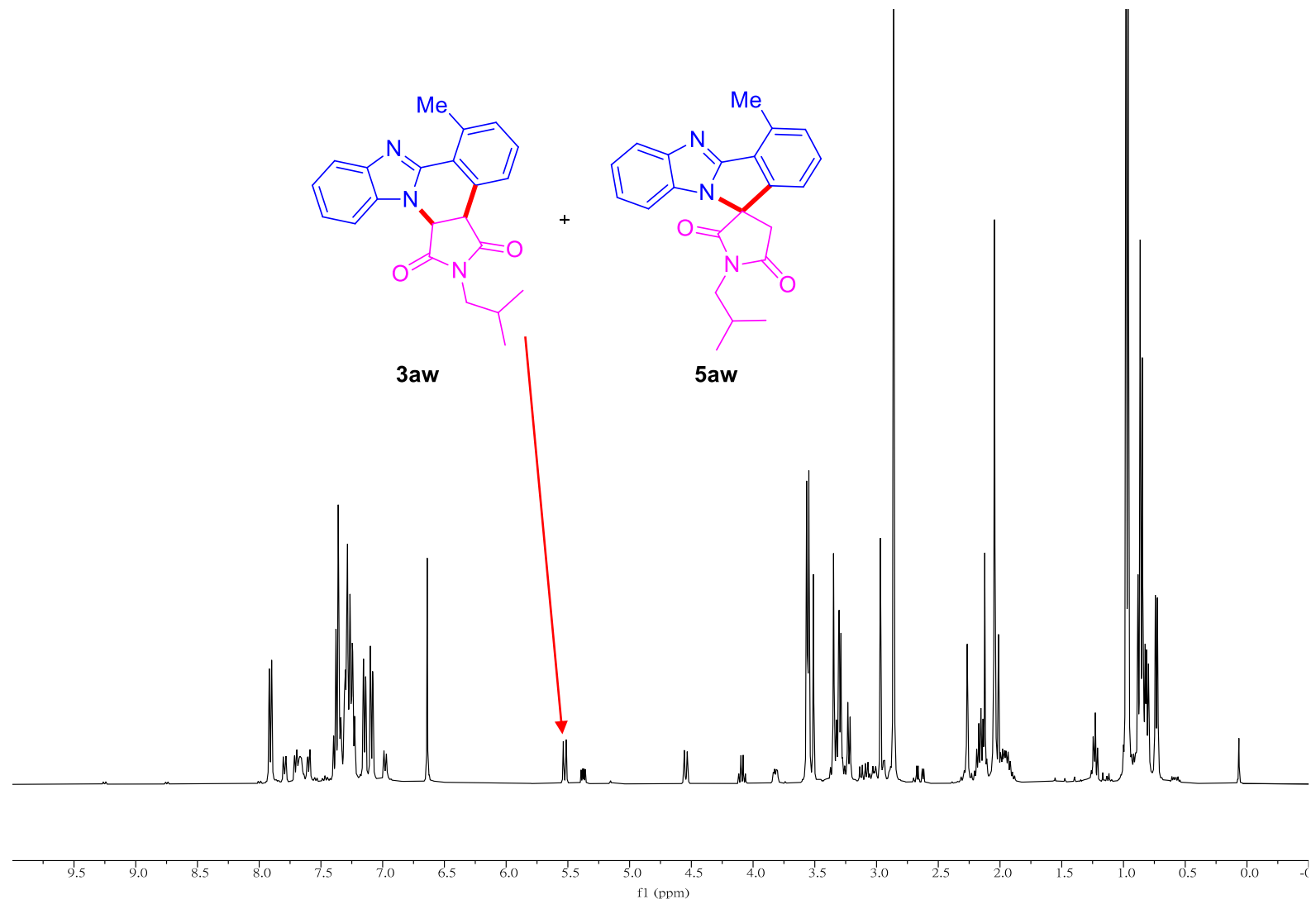
<sup>1</sup>H NMR spectrum (400 MHz) of compound **3av** in CDCl<sub>3</sub>.



## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
476.1157	1	C <sub>29</sub> H <sub>19</sub> ClN <sub>3</sub> O <sub>2</sub>	476.1160	-0.7	36.7	1	100.00	21.5	even	ok	M+H

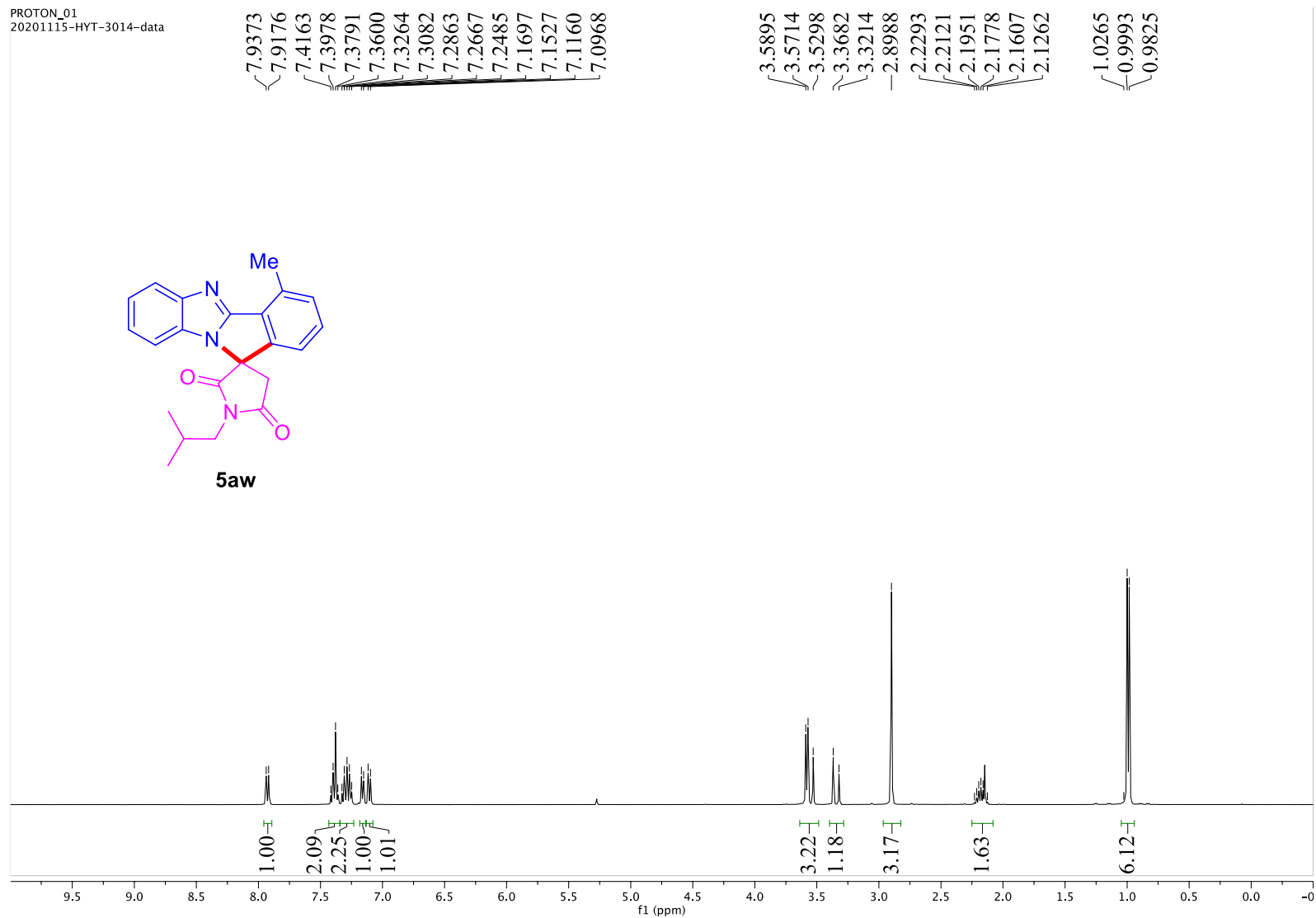
HRMS (ESI) of compound **3av**.



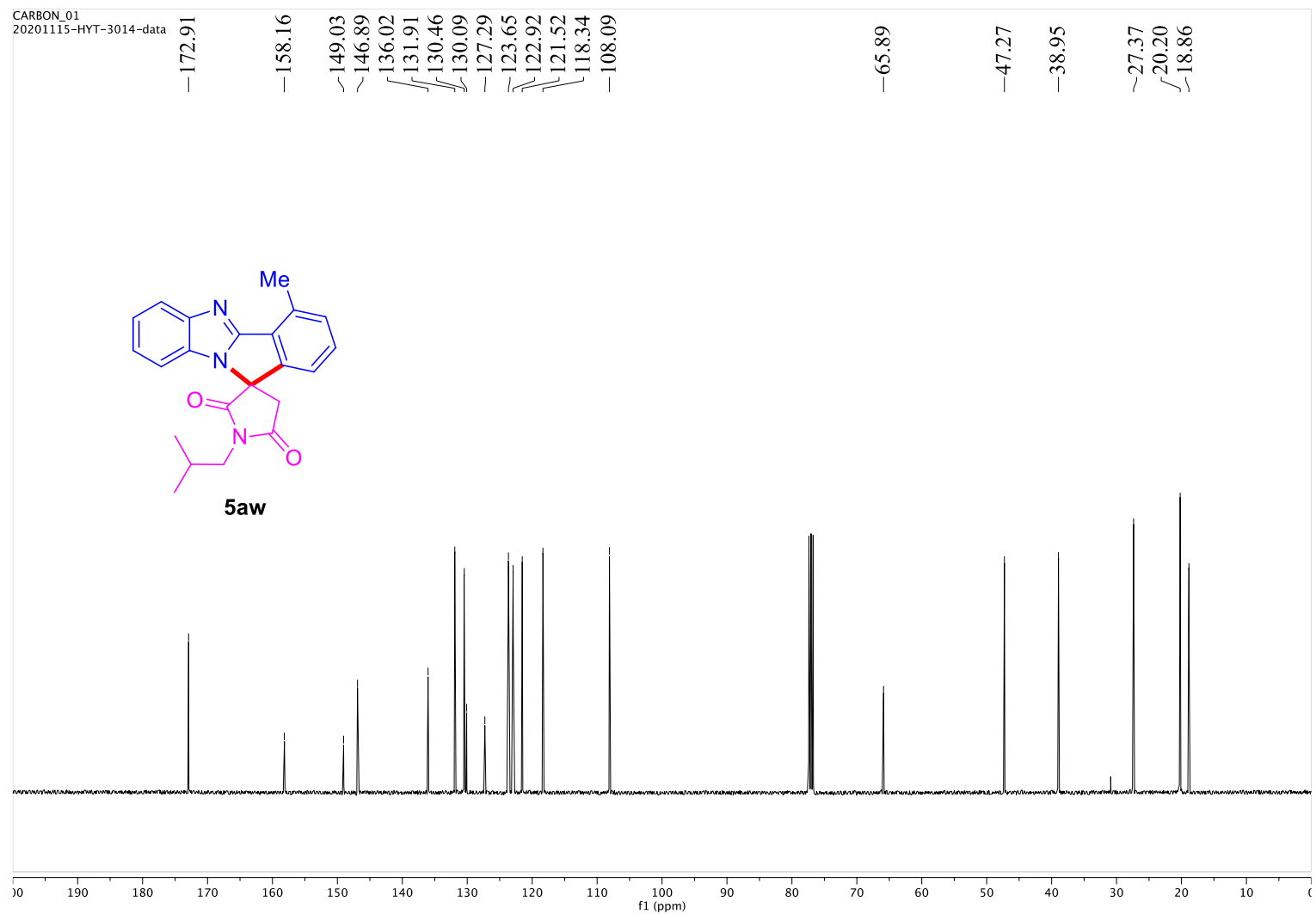
<sup>1</sup>H NMR spectrum (400 MHz) of compound **3aw** and **5aw** in CDCl<sub>3</sub>.



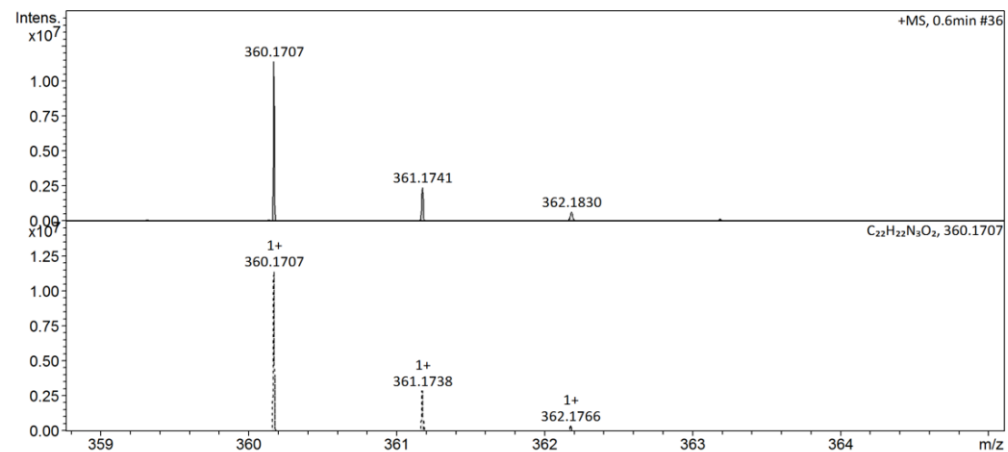
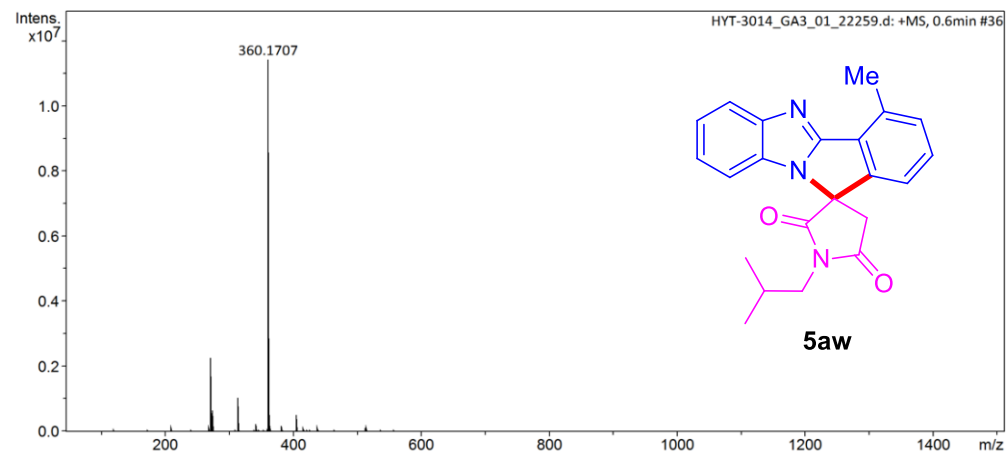
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<sup>1</sup>H NMR spectrum (400 MHz) of compound **5aw** in CDCl<sub>3</sub>.



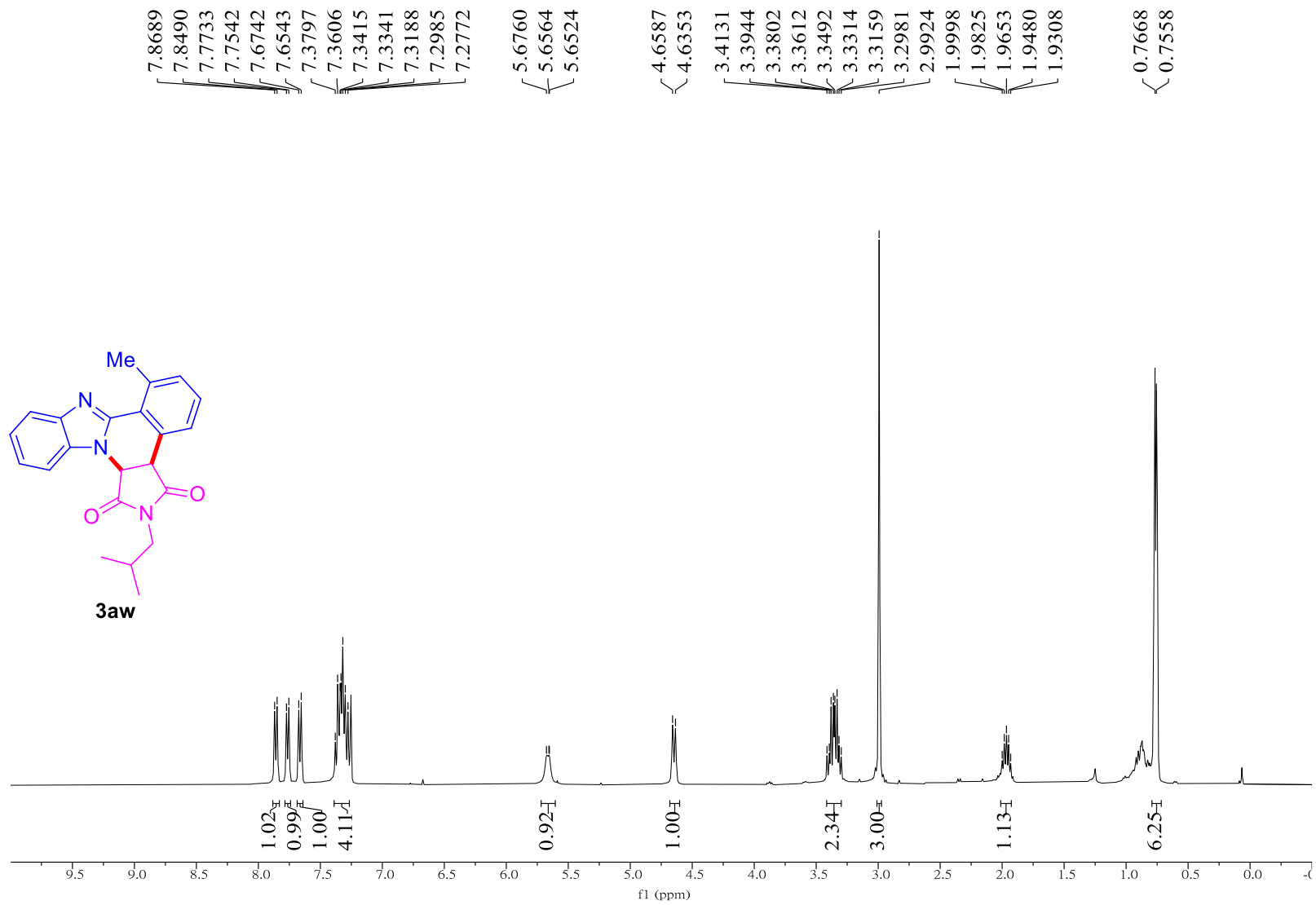
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5aw** in  $\text{CDCl}_3$ .



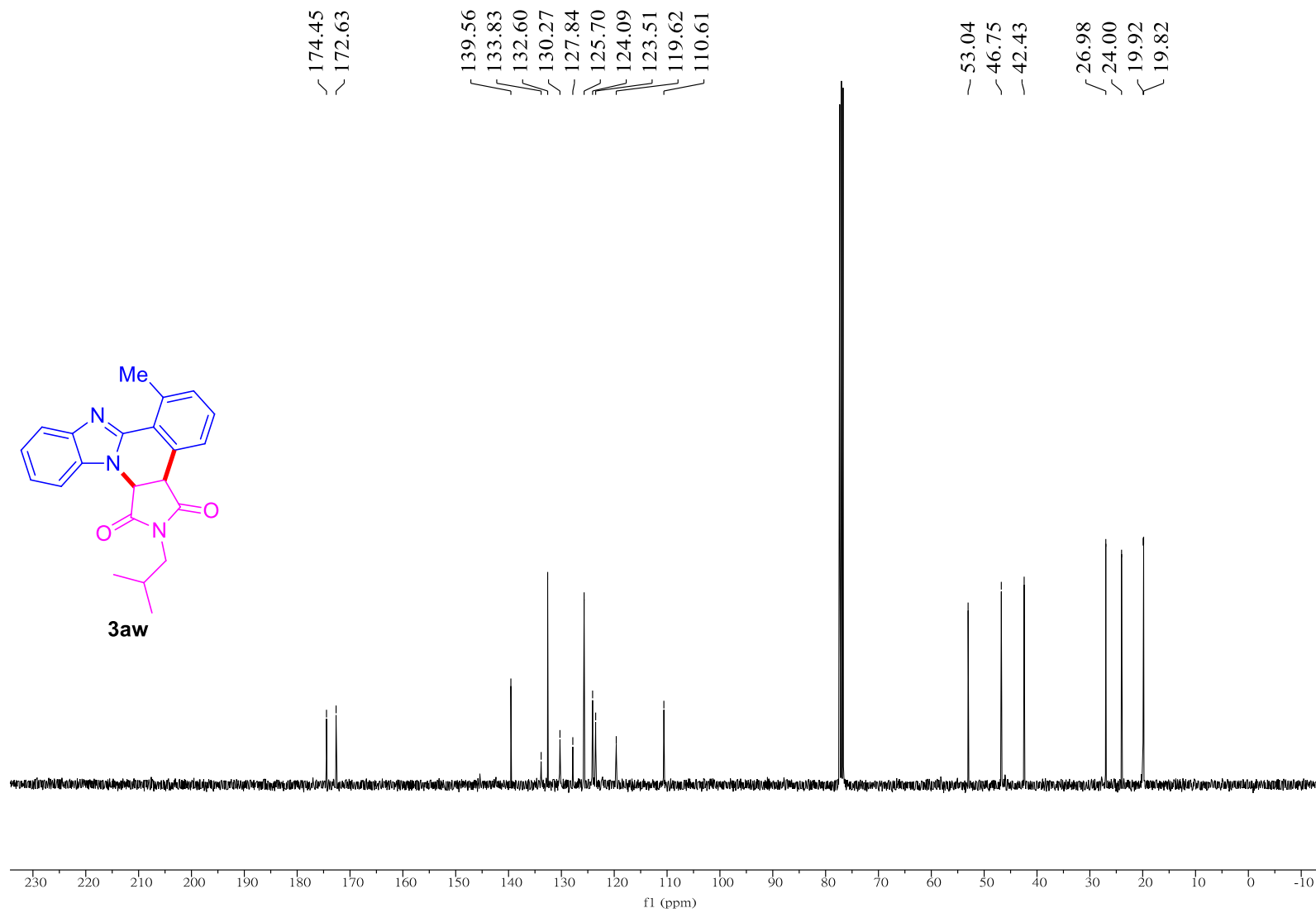
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
360.1707	1	C <sub>22</sub> H <sub>22</sub> N <sub>3</sub> O <sub>2</sub>	360.1707	-0.1	26.9	1	100.00	13.5	even	ok	M+H

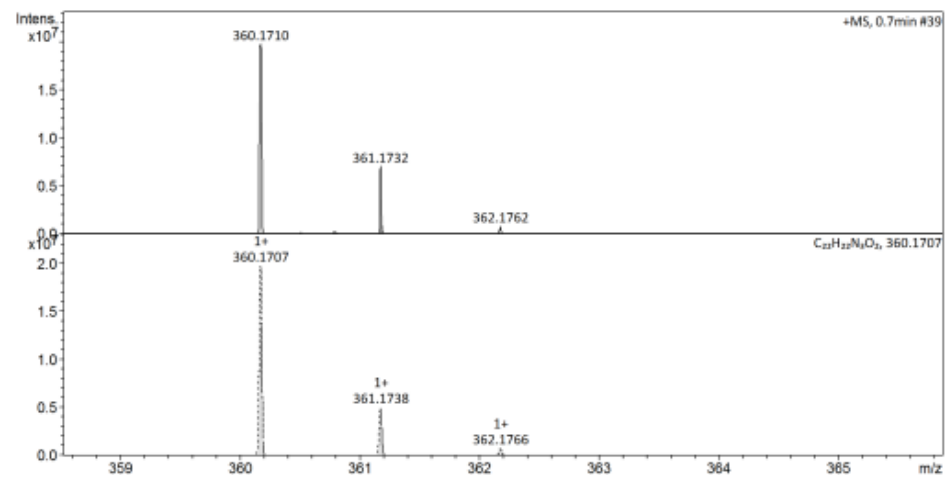
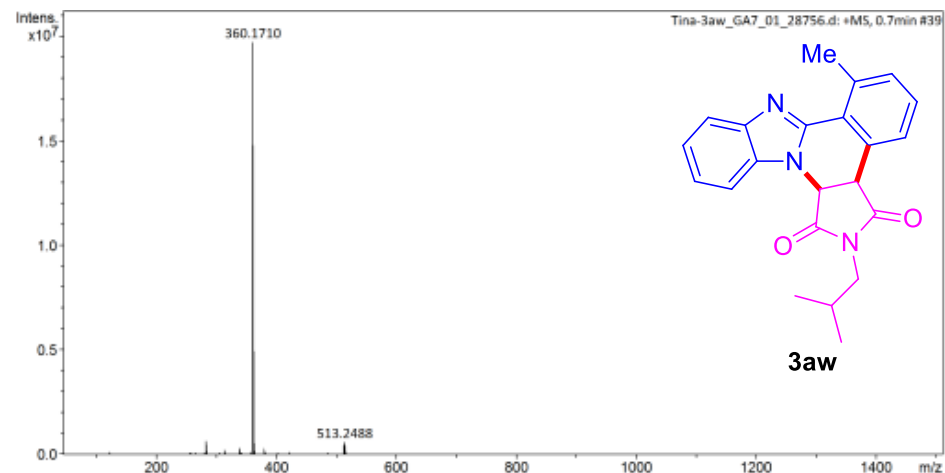
HRMS (ESI) of compound **5aw**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3aw** in  $\text{CDCl}_3$ .



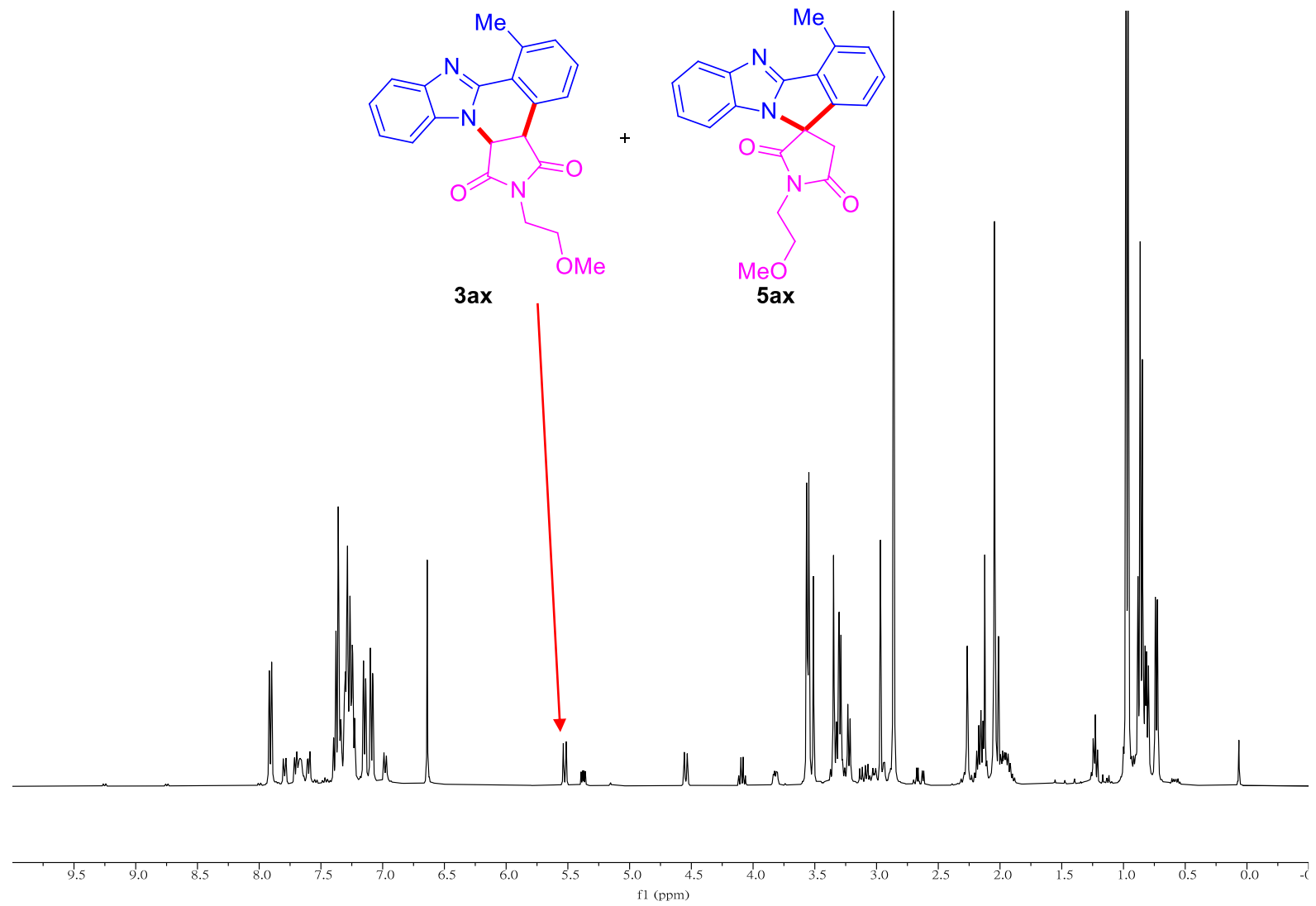
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3aw** in  $\text{CDCl}_3$ .



### Display Report

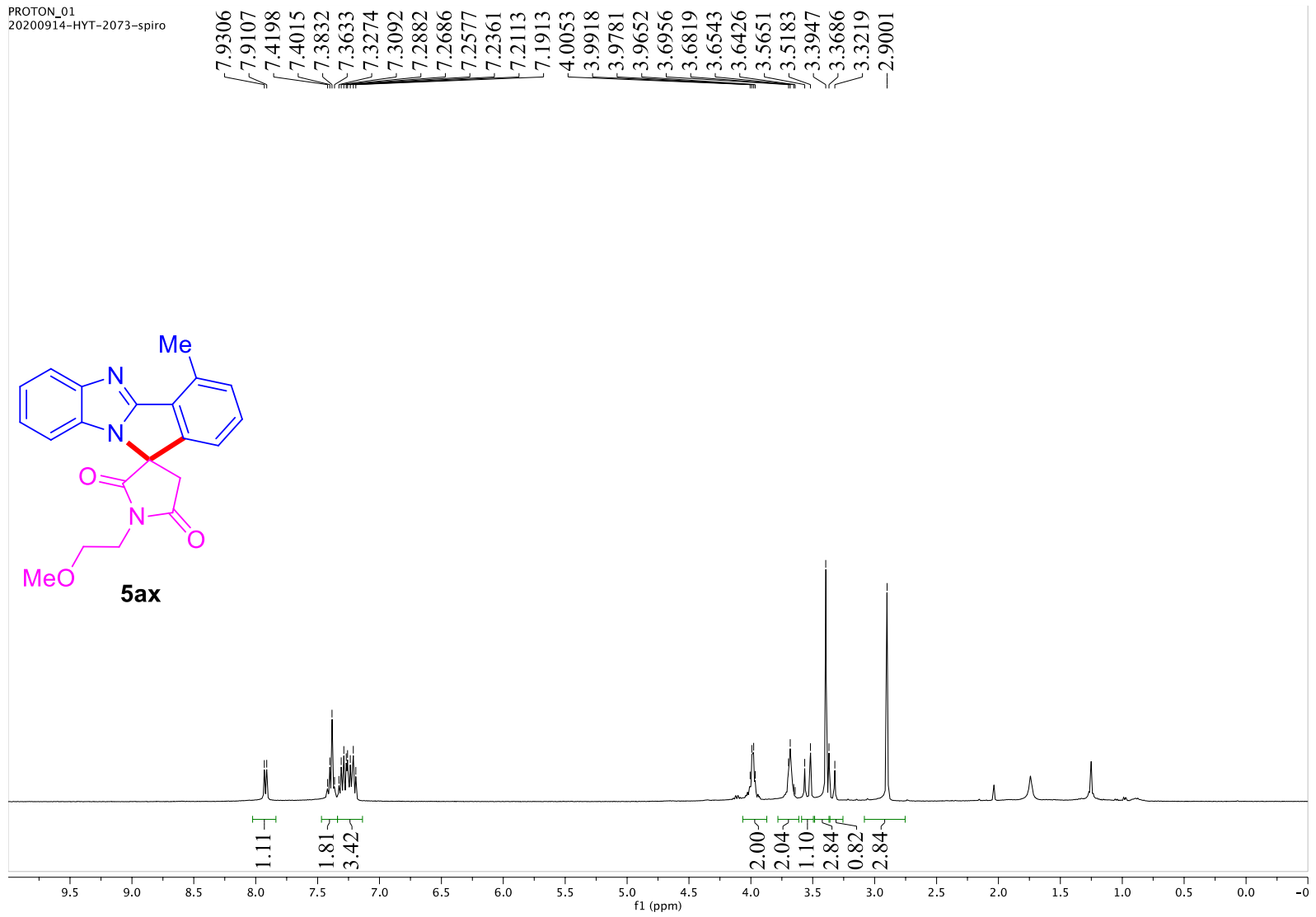
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdB	e <sup>-</sup> Conf	N-Rule	Adduct
360.1710	1	C <sub>22</sub> H <sub>22</sub> N <sub>3</sub> O <sub>2</sub>	360.1707	0.8	58.3	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **3aw**.



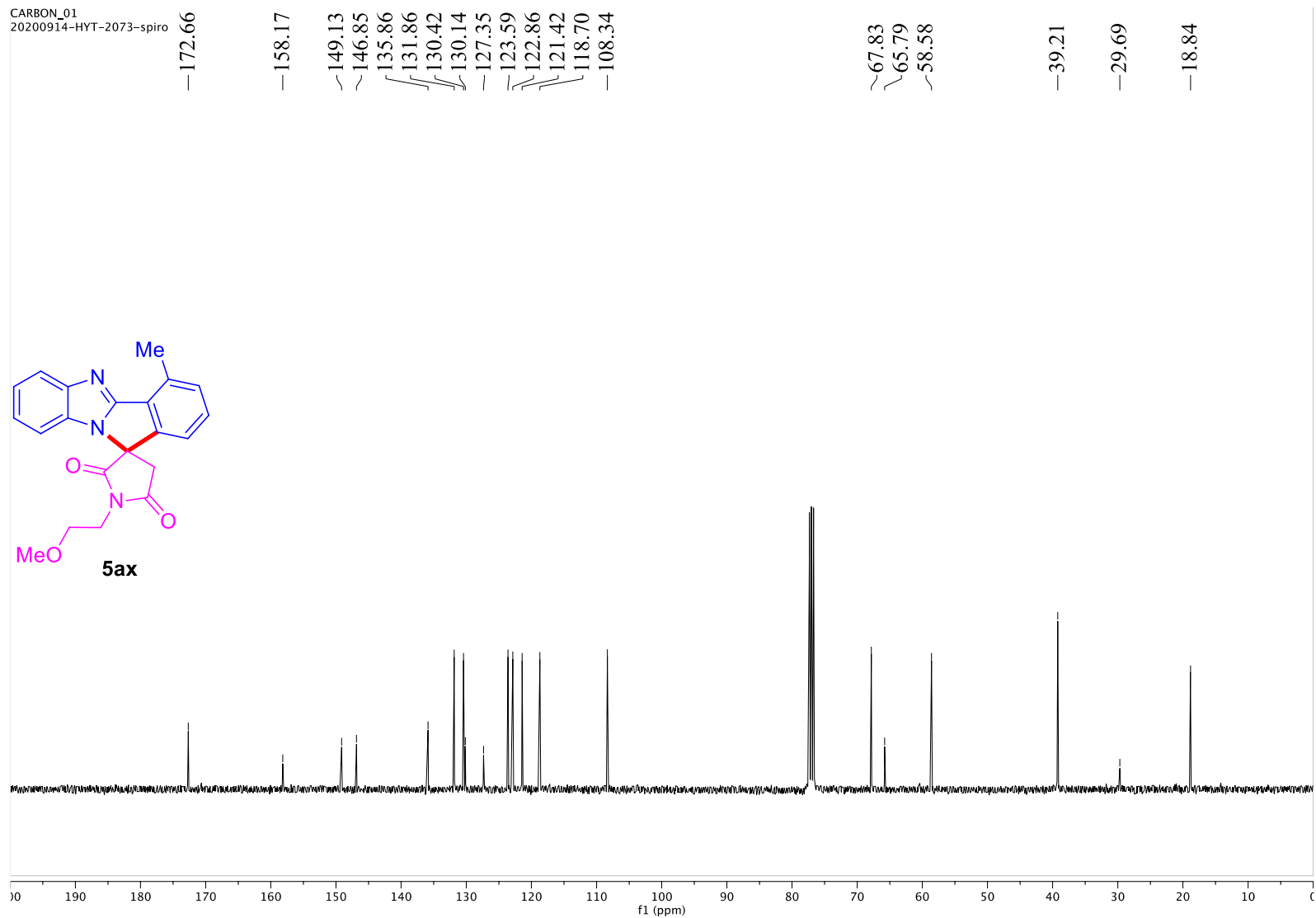
<sup>1</sup>H NMR spectrum (400 MHz) of compound **3ax** and **5ax** (ratio) in CDCl<sub>3</sub>.

PROTON\_01  
20200914-HYT-2073-spiro

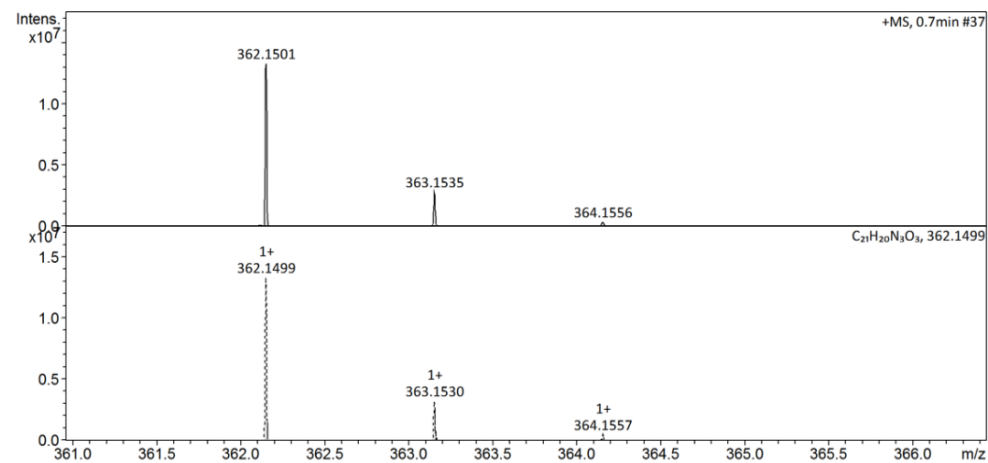
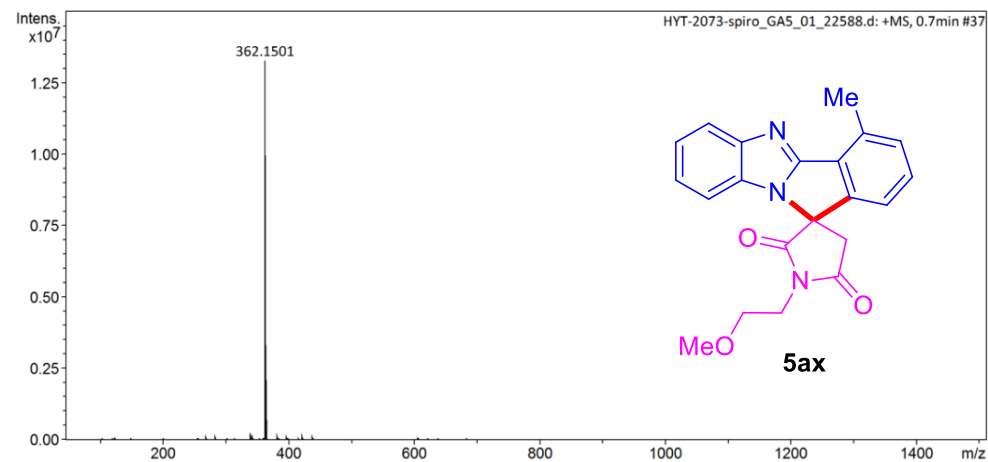


<sup>1</sup>H NMR spectrum (400 MHz) of compound **5ax** in CDCl<sub>3</sub>.





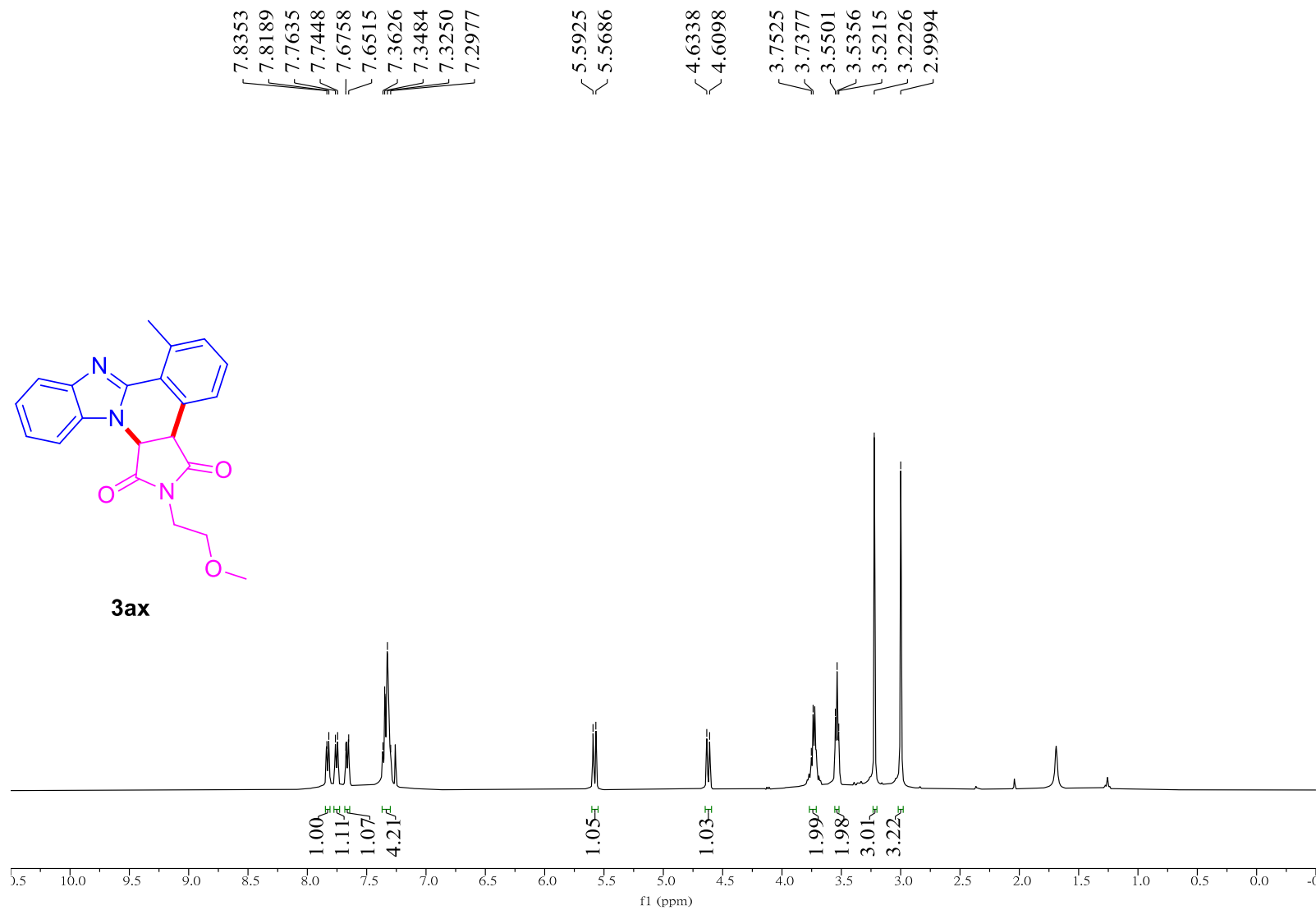
<sup>13</sup>C NMR spectrum (100 MHz) of compound **5ax** in CDCl<sub>3</sub>  
281



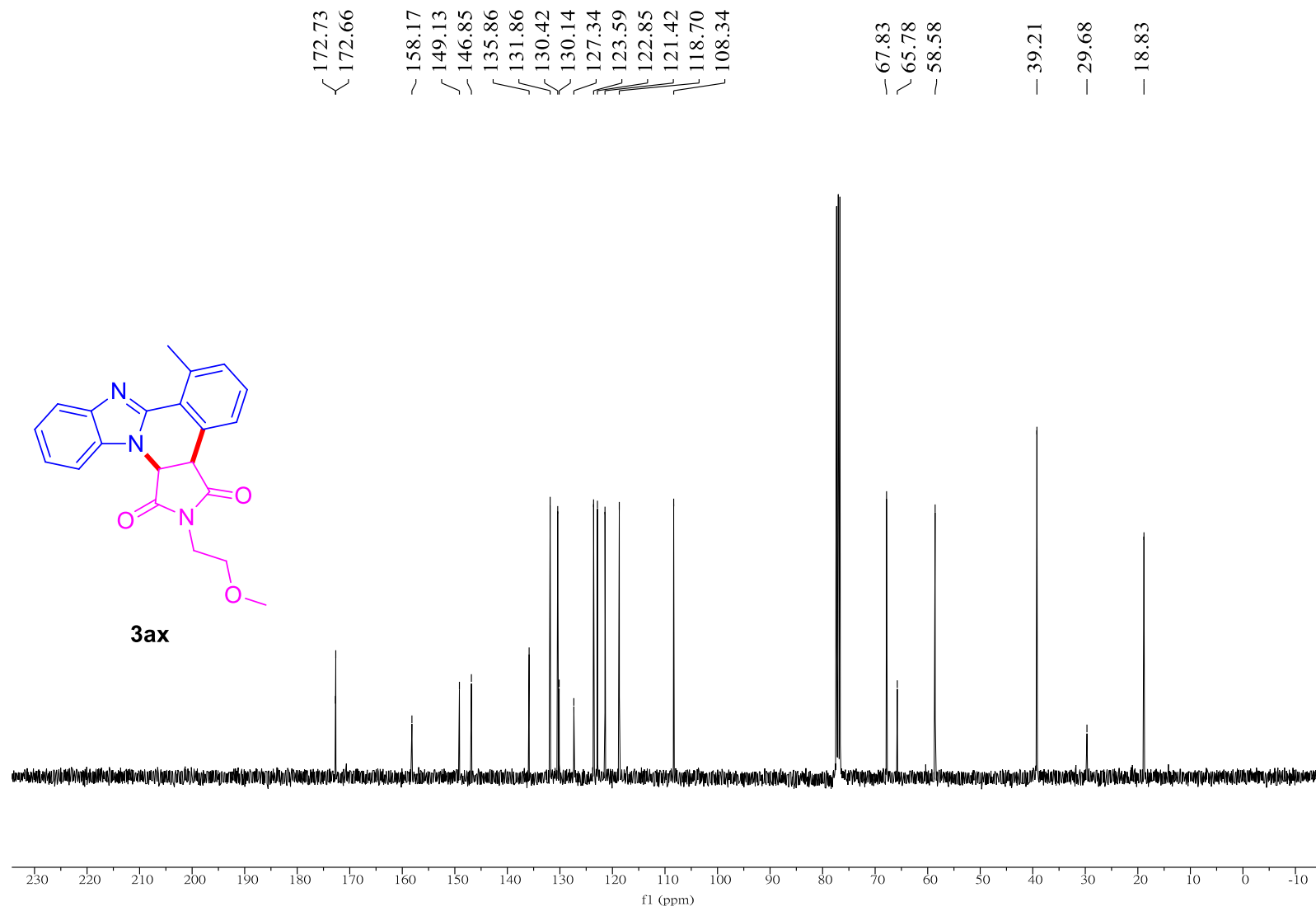
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
362.1501	1	C <sub>21</sub> H <sub>20</sub> N <sub>3</sub> O <sub>3</sub>	362.1499	-0.5	15.8	1	100.00	13.5	even	ok	M+H

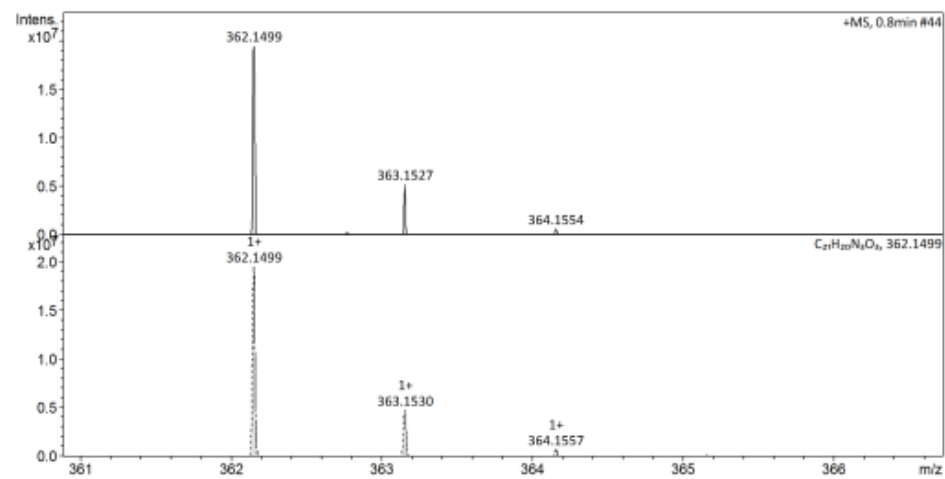
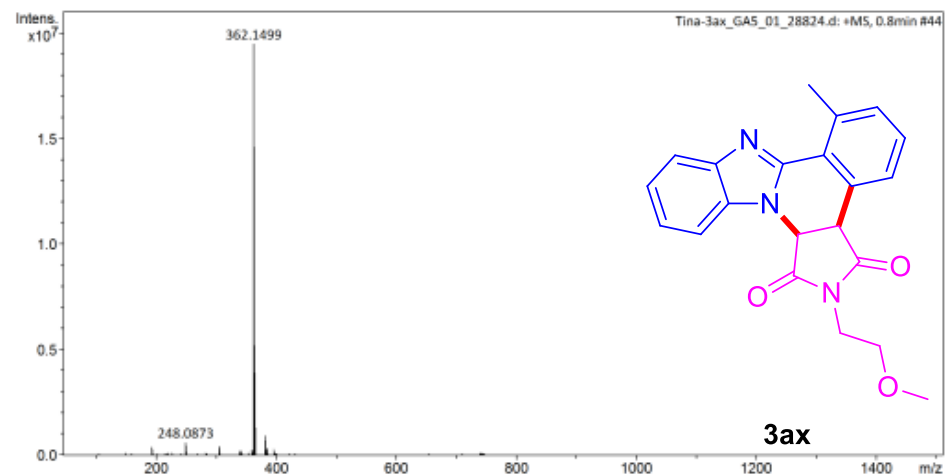
HRMS (ESI) of compound **5ax**.



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3ax** in  $\text{CDCl}_3$ .



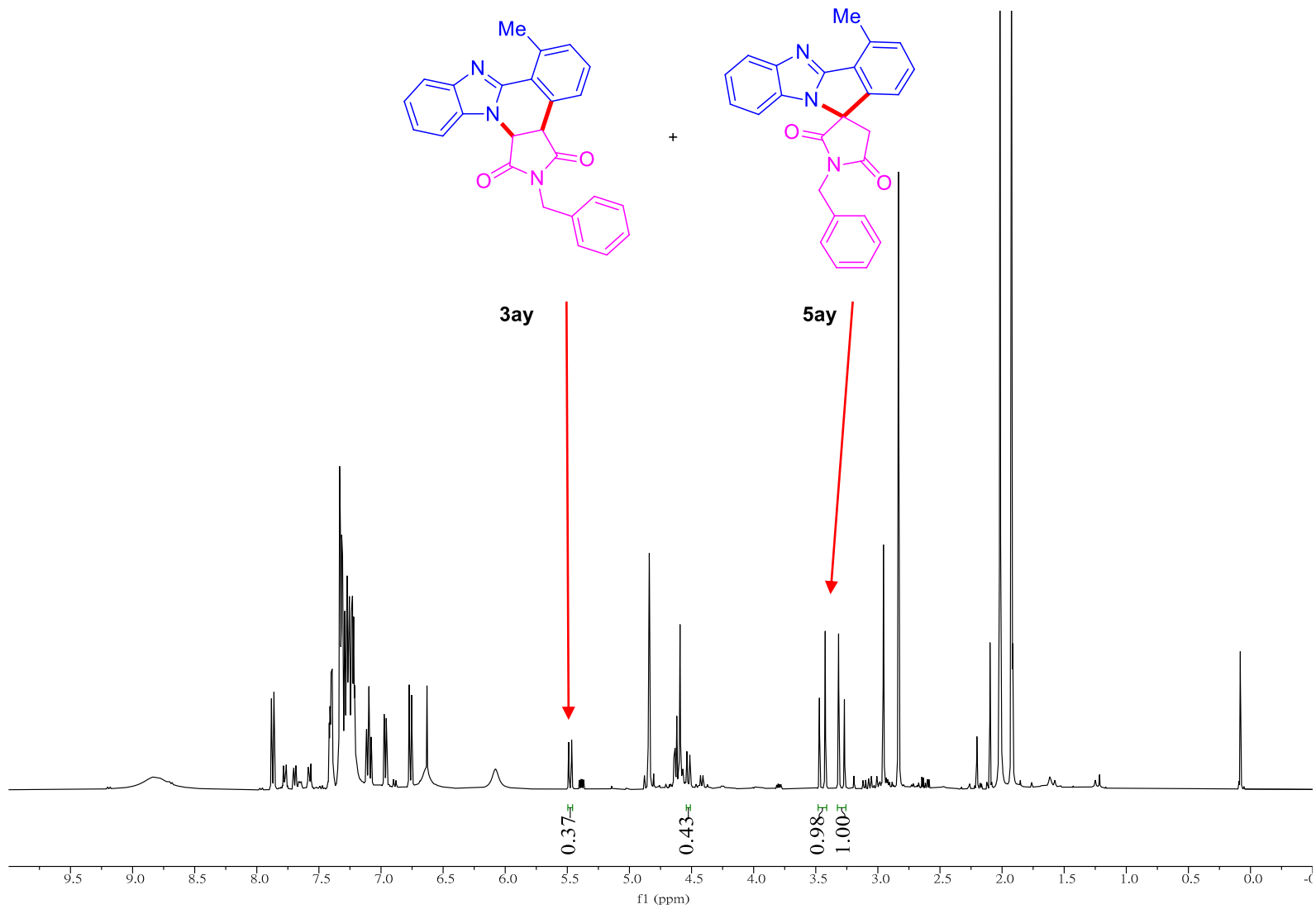
<sup>13</sup>C NMR spectrum (100 MHz) of compound **3ax** in CDCl<sub>3</sub>.



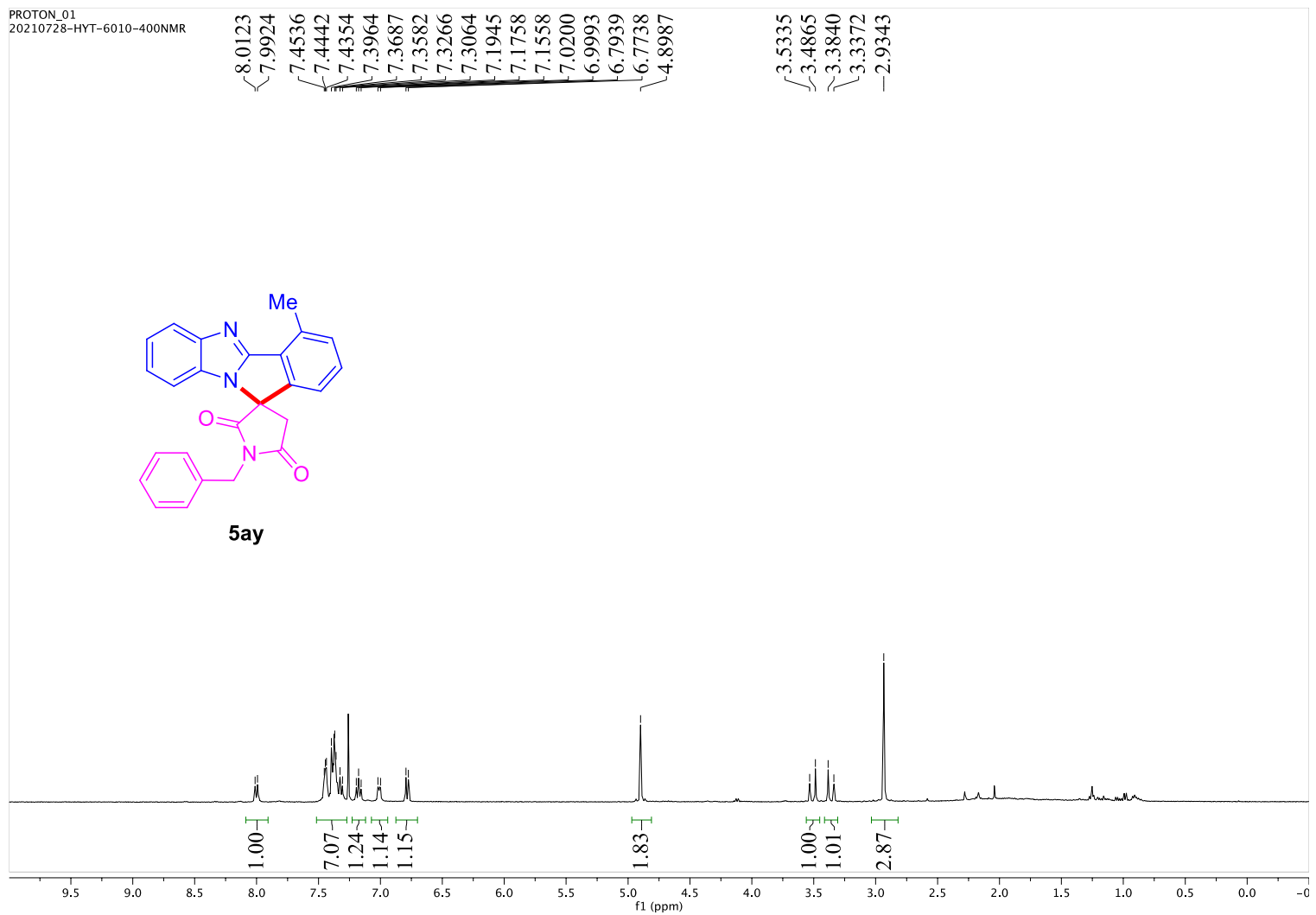
### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
362.1499	1	C <sub>21</sub> H <sub>20</sub> N <sub>2</sub> O <sub>3</sub>	362.1499	0.1	12.8	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **3ax**.

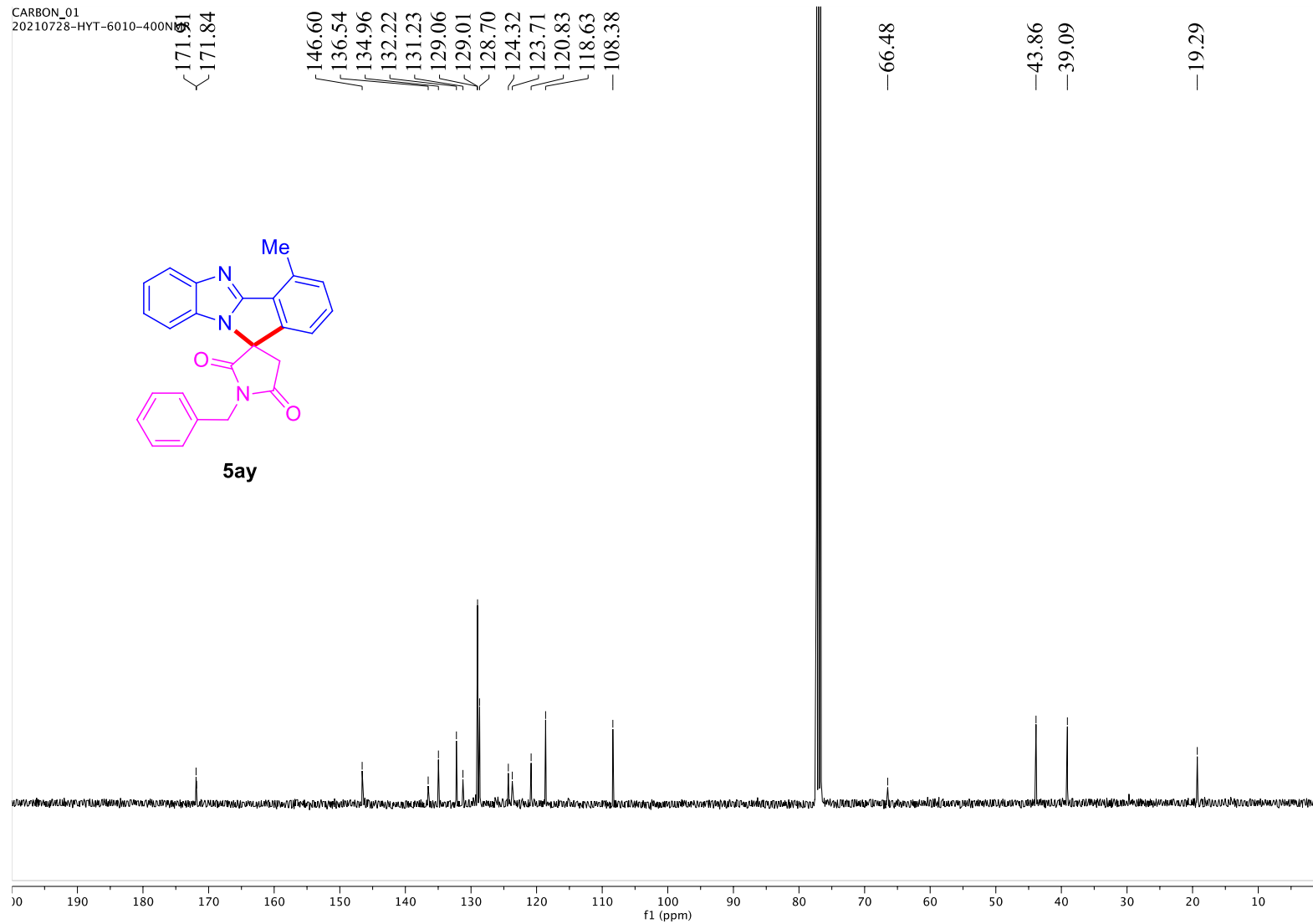


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3ay** and **5ay** in  $\text{CDCl}_3$ .



$^1\text{H}$  NMR spectrum (400 MHz) of compound **5ay** in  $\text{CDCl}_3$ .

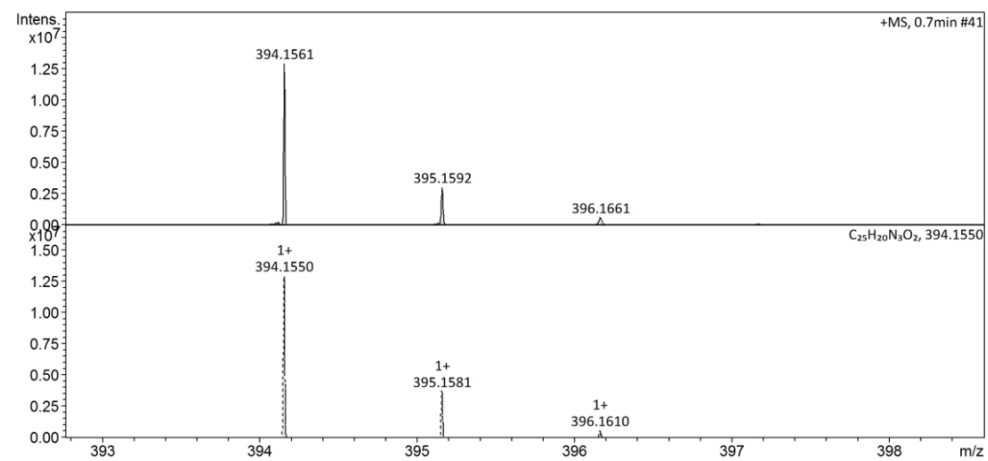
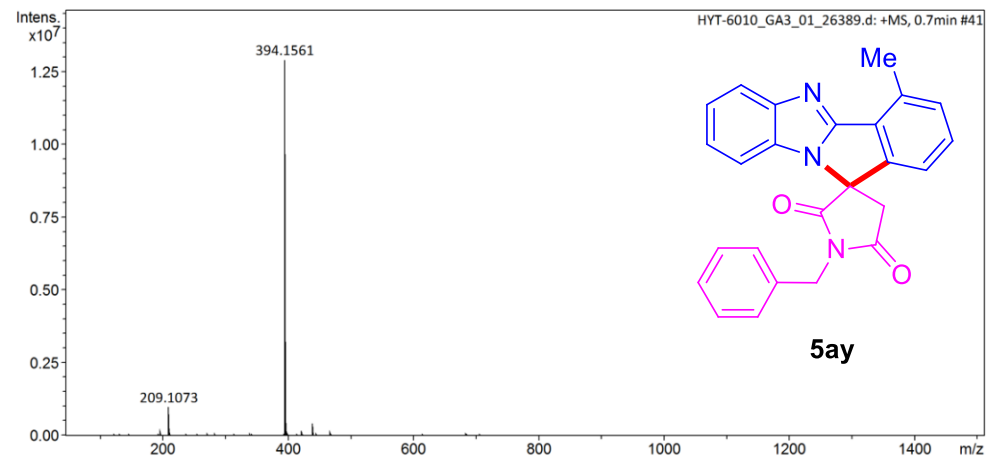
CARBON\_01  
20210728-HYT-6010-400N



$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5ay** in  $\text{CDCl}_3$ .

288

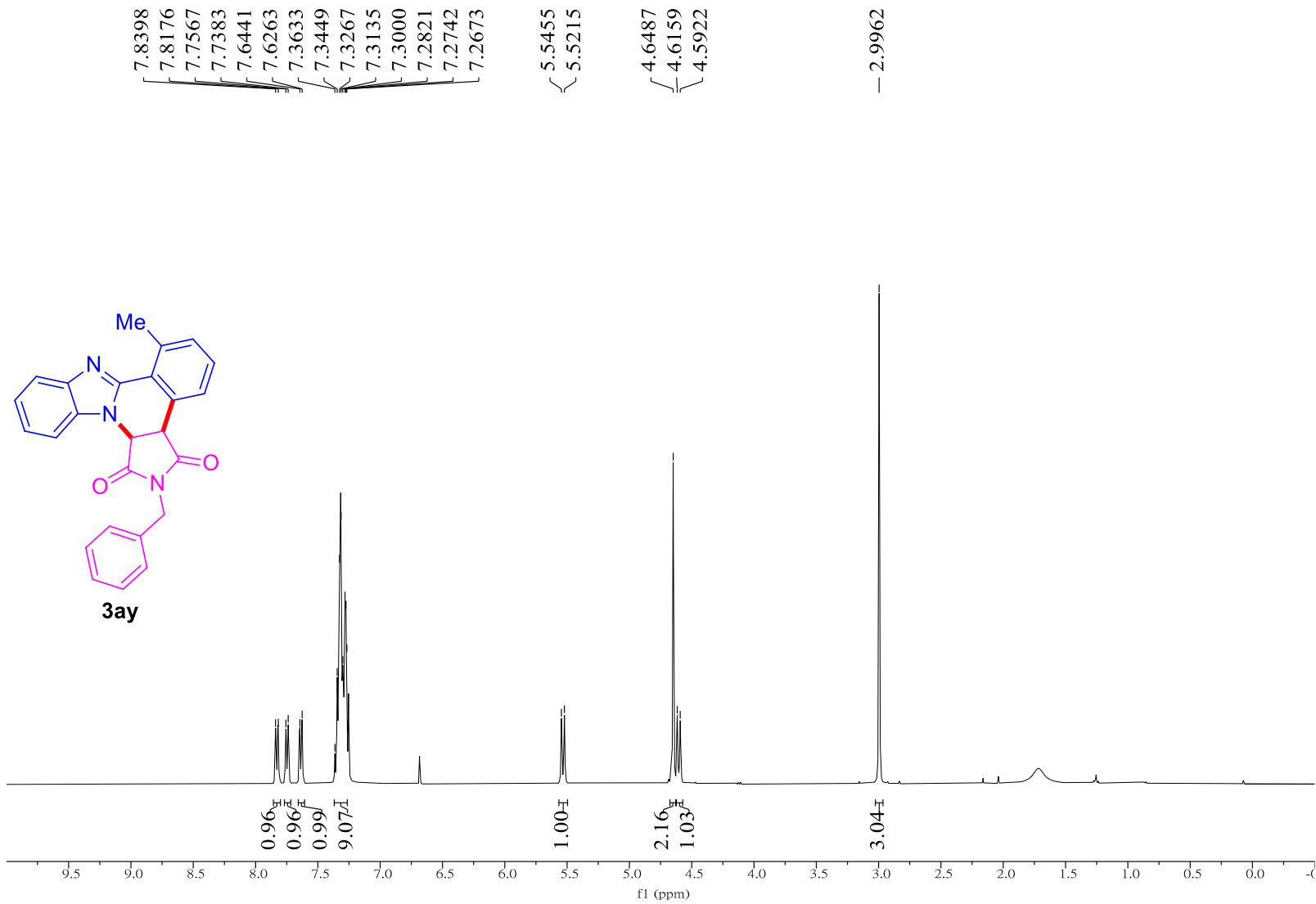




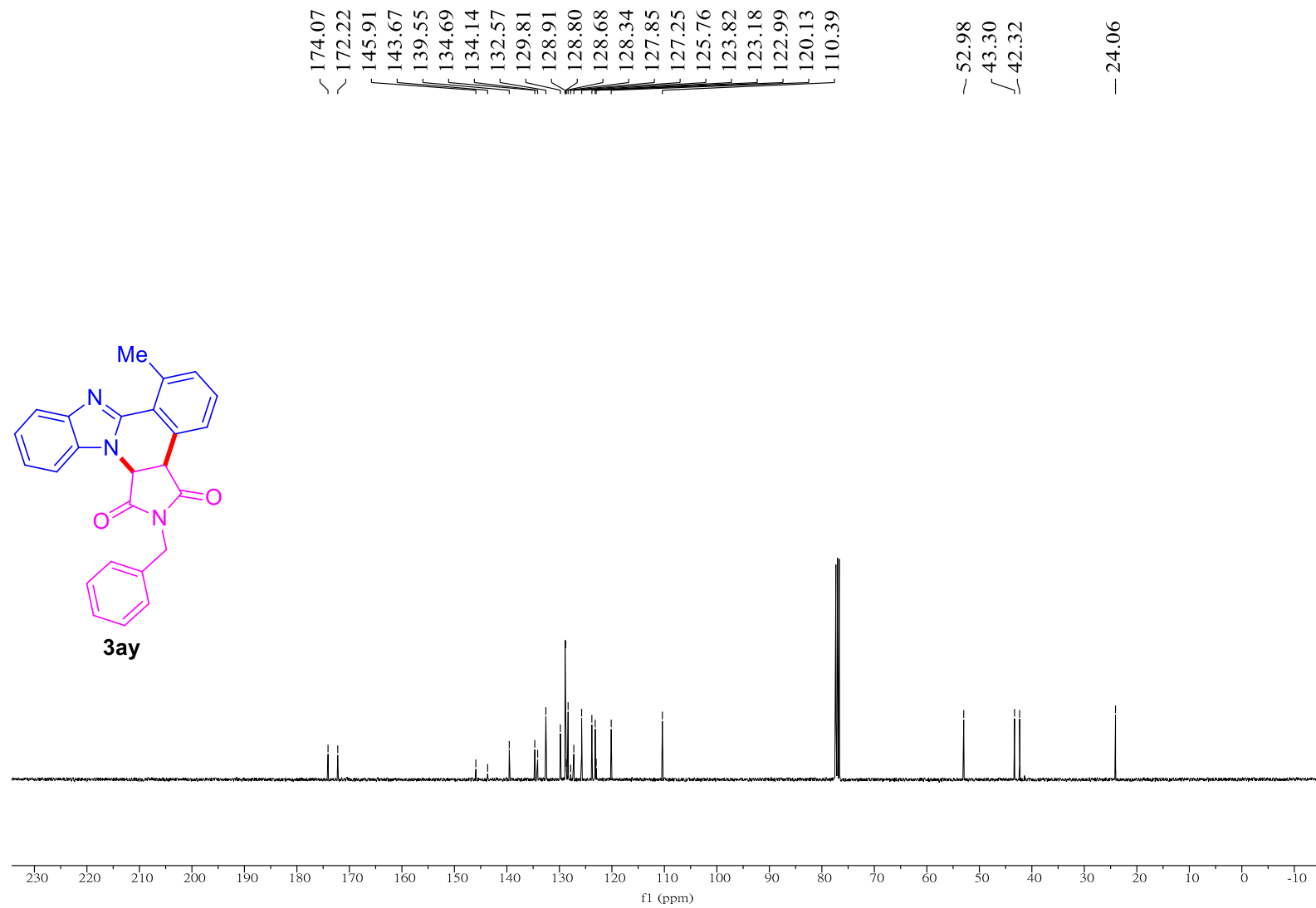
## Display Report

Meas. $m/z$	#	Ion Formula	$m/z$	err [ppm]	mSigma	# Sigma	Score	rdb	$e^-$ Conf	N-Rule	Adduct
394.1561	1	$C_{25}H_{20}N_4O_2$	394.1550	2.8	25.6	1	100.00	17.5	even	ok	M

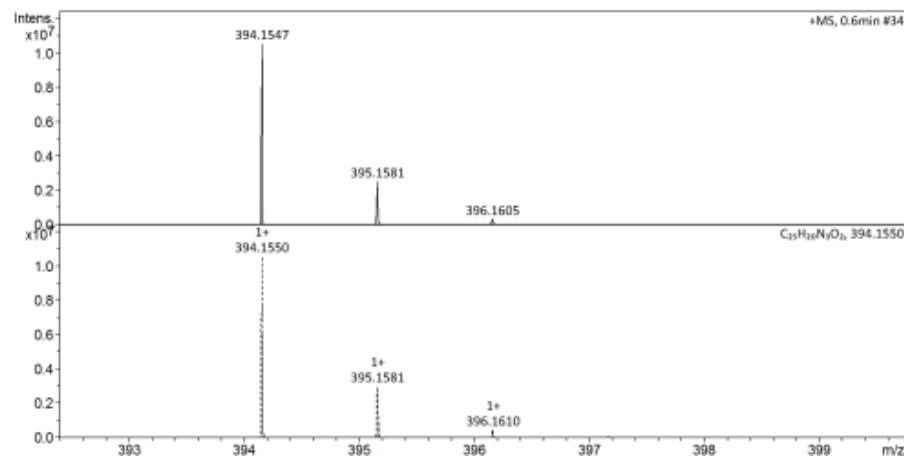
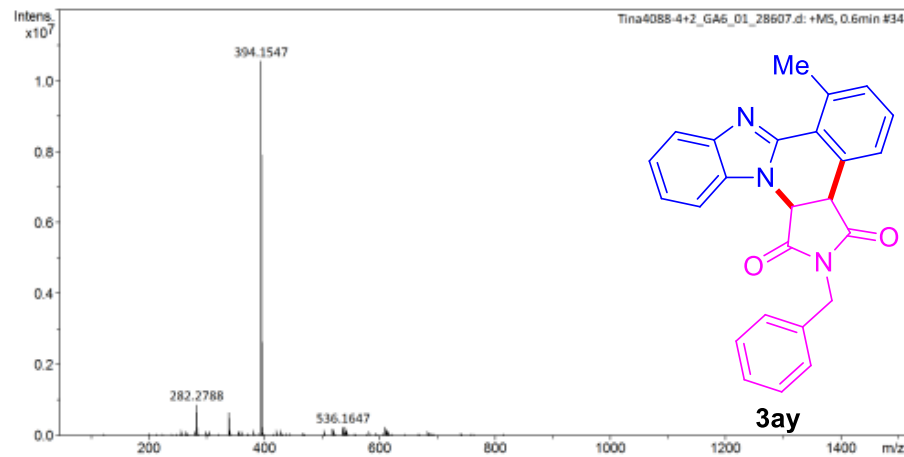
HRMS (ESI) of compound **5ay**.



**<sup>1</sup>H NMR spectrum (400 MHz) of compound **3ay** in CDCl<sub>3</sub>.**



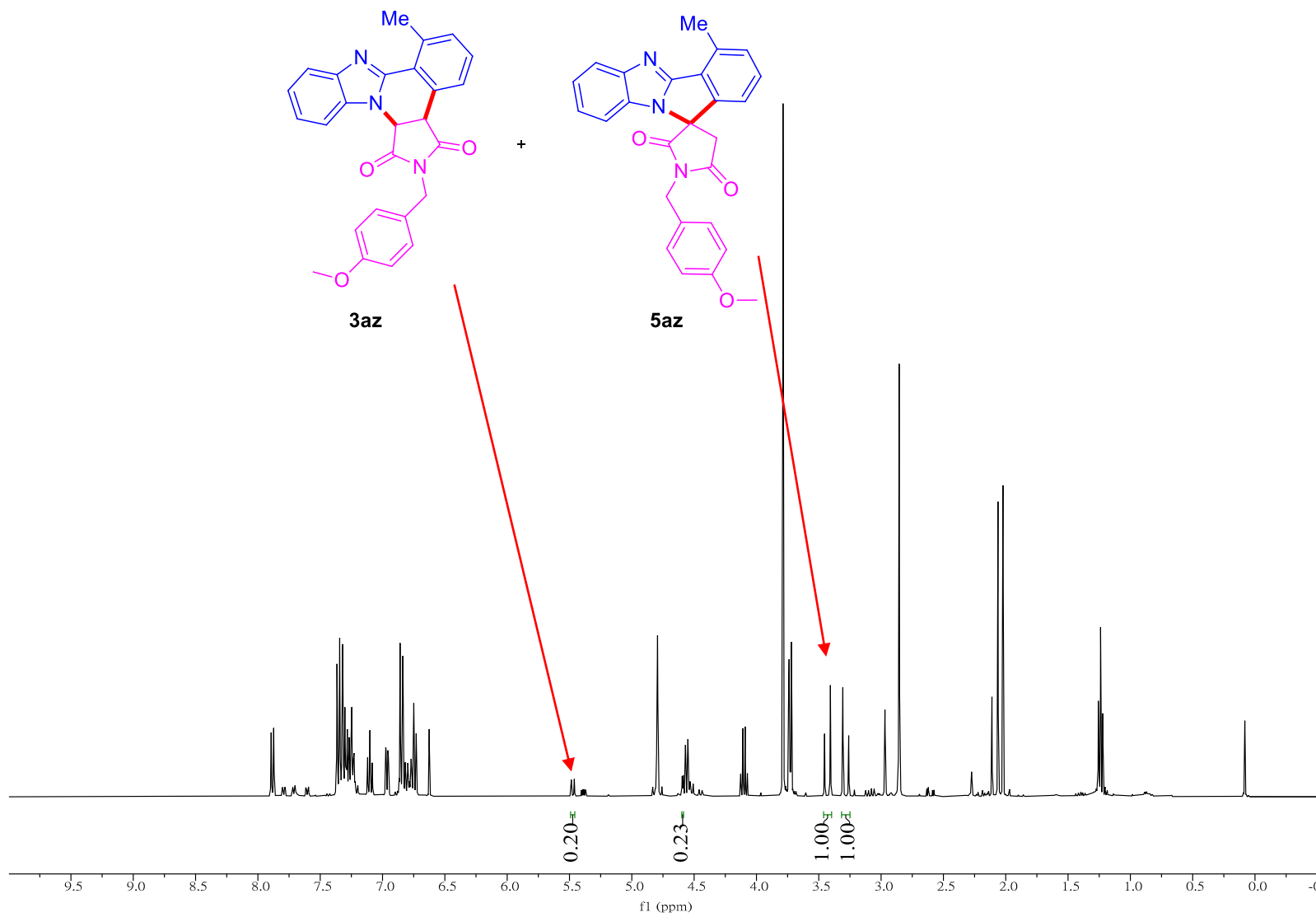
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3ay** in  $\text{CDCl}_3$ .



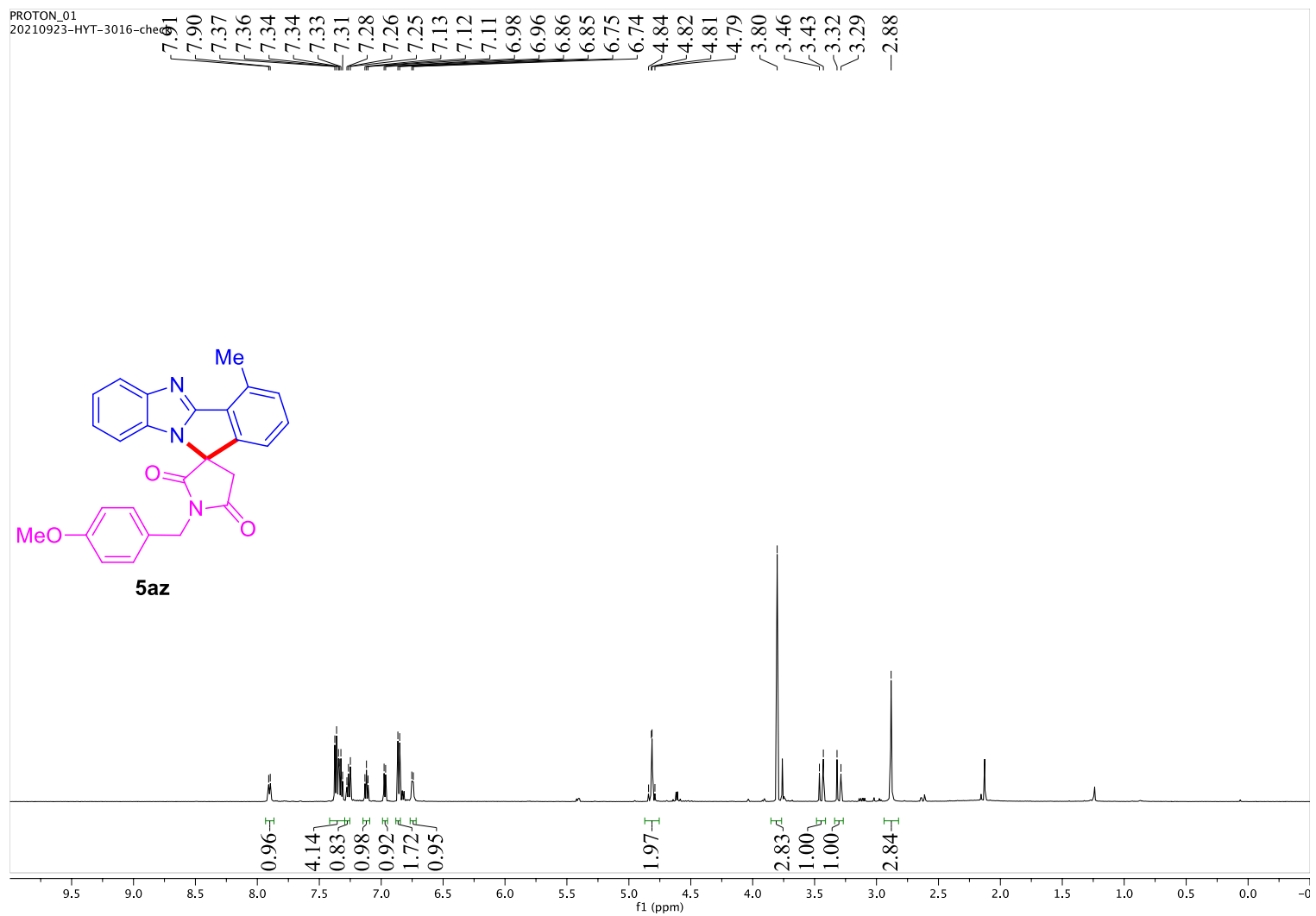
### Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
394.1547	1	C <sub>25</sub> H <sub>20</sub> N <sub>3</sub> O <sub>2</sub>	394.1550	0.8	24.2	1	100.00	17.5	even	ok	M+H

HRMS (ESI) of compound **3ay**.

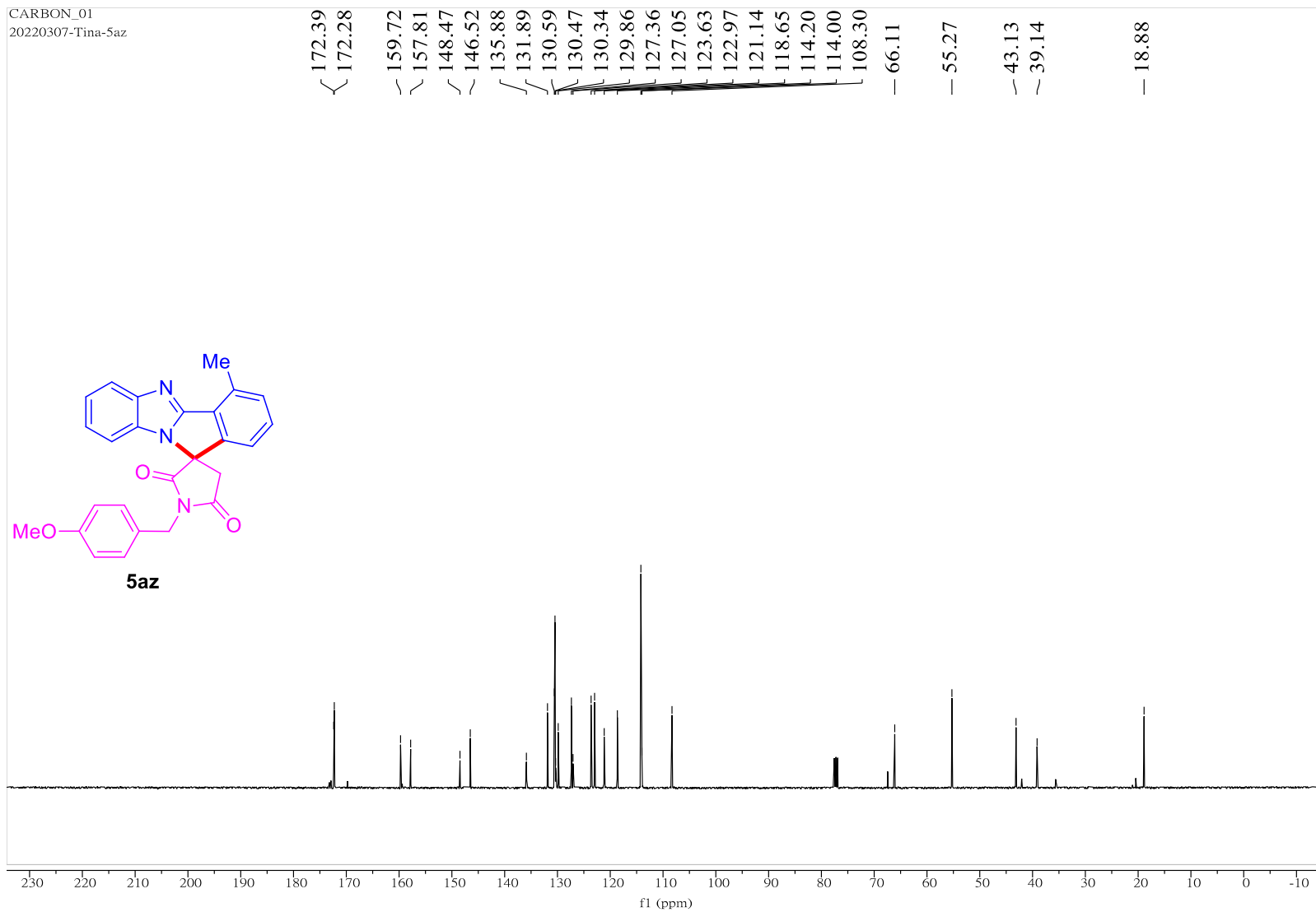


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3az** and **5az** in  $\text{CDCl}_3$ .

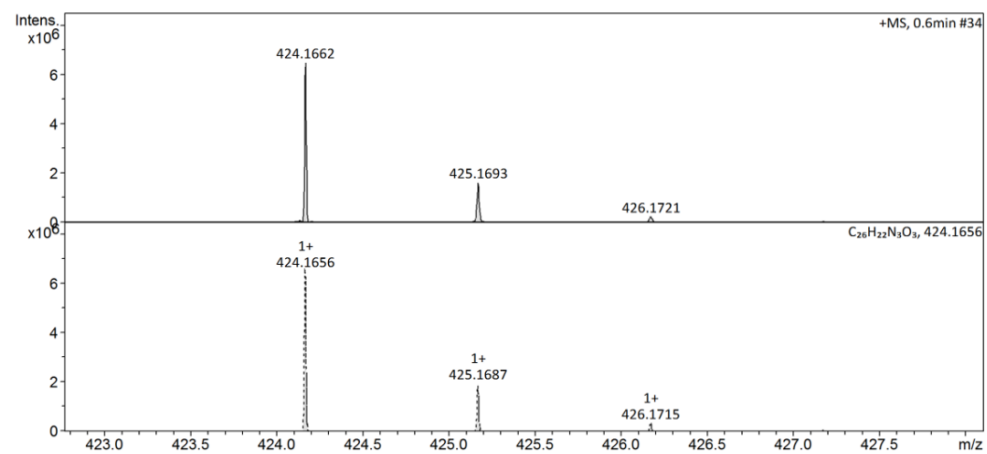
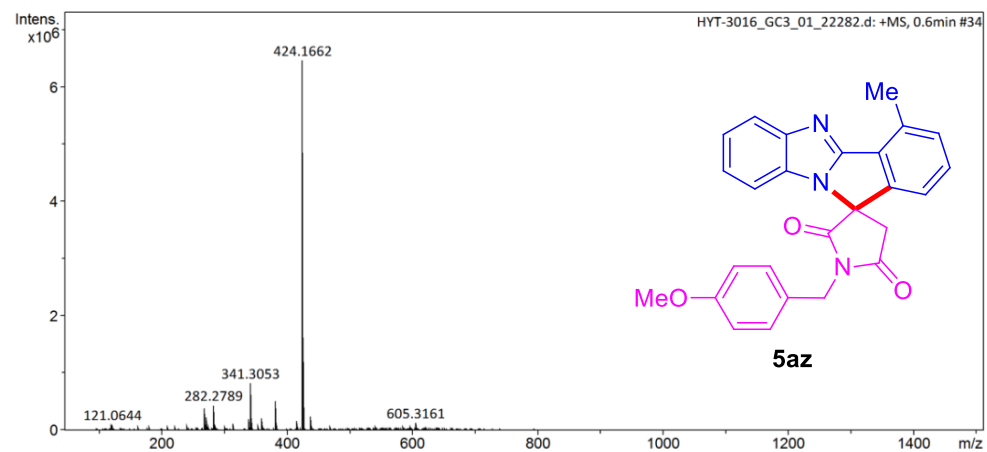


<sup>1</sup>H NMR spectrum (600 MHz) of compound **5az** in CDCl<sub>3</sub>.

CARBON\_01  
20220307-Tina-5az



<sup>13</sup>C NMR spectrum (100 MHz) of compound **5az** in CDCl<sub>3</sub>.

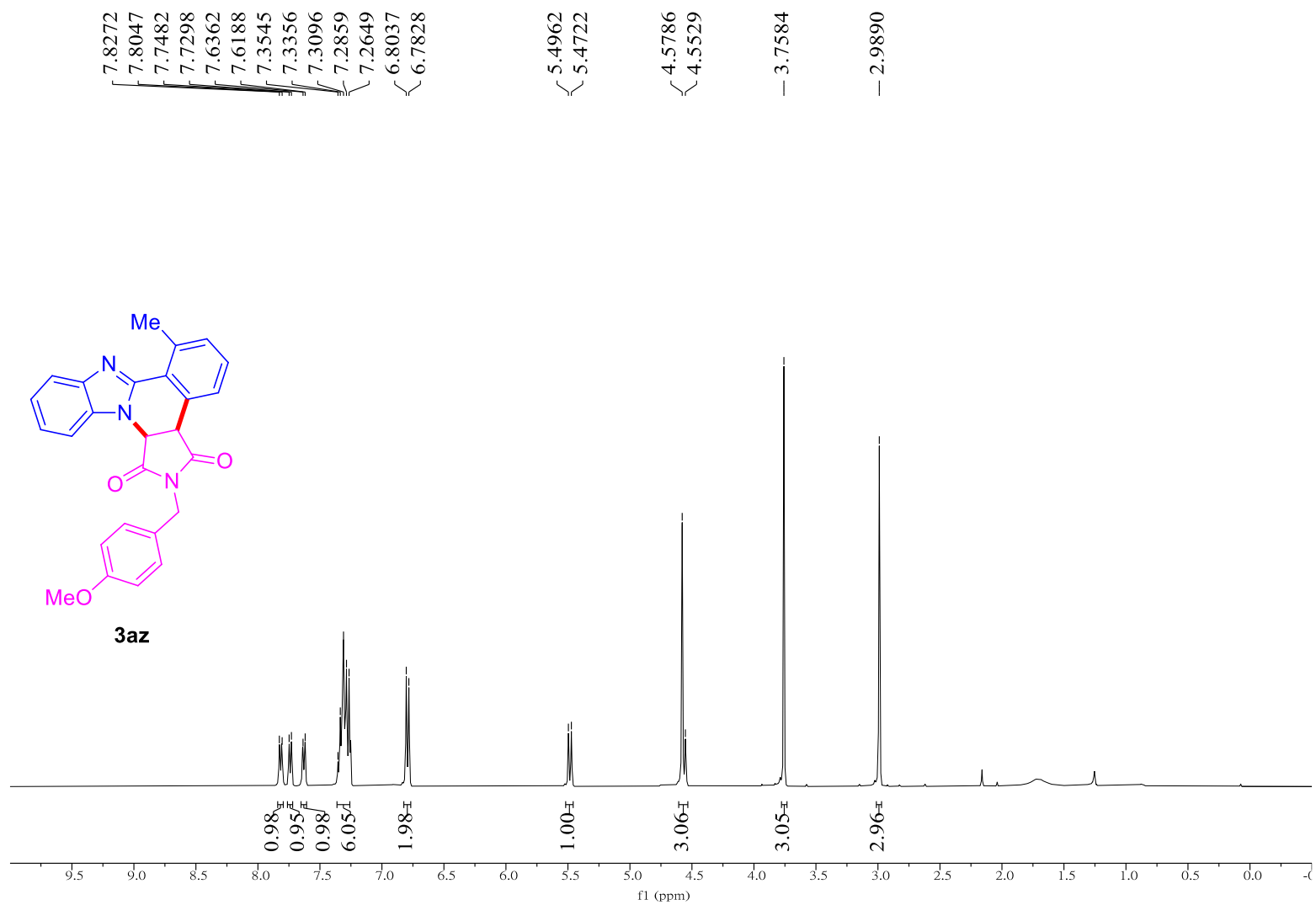


## Display Report

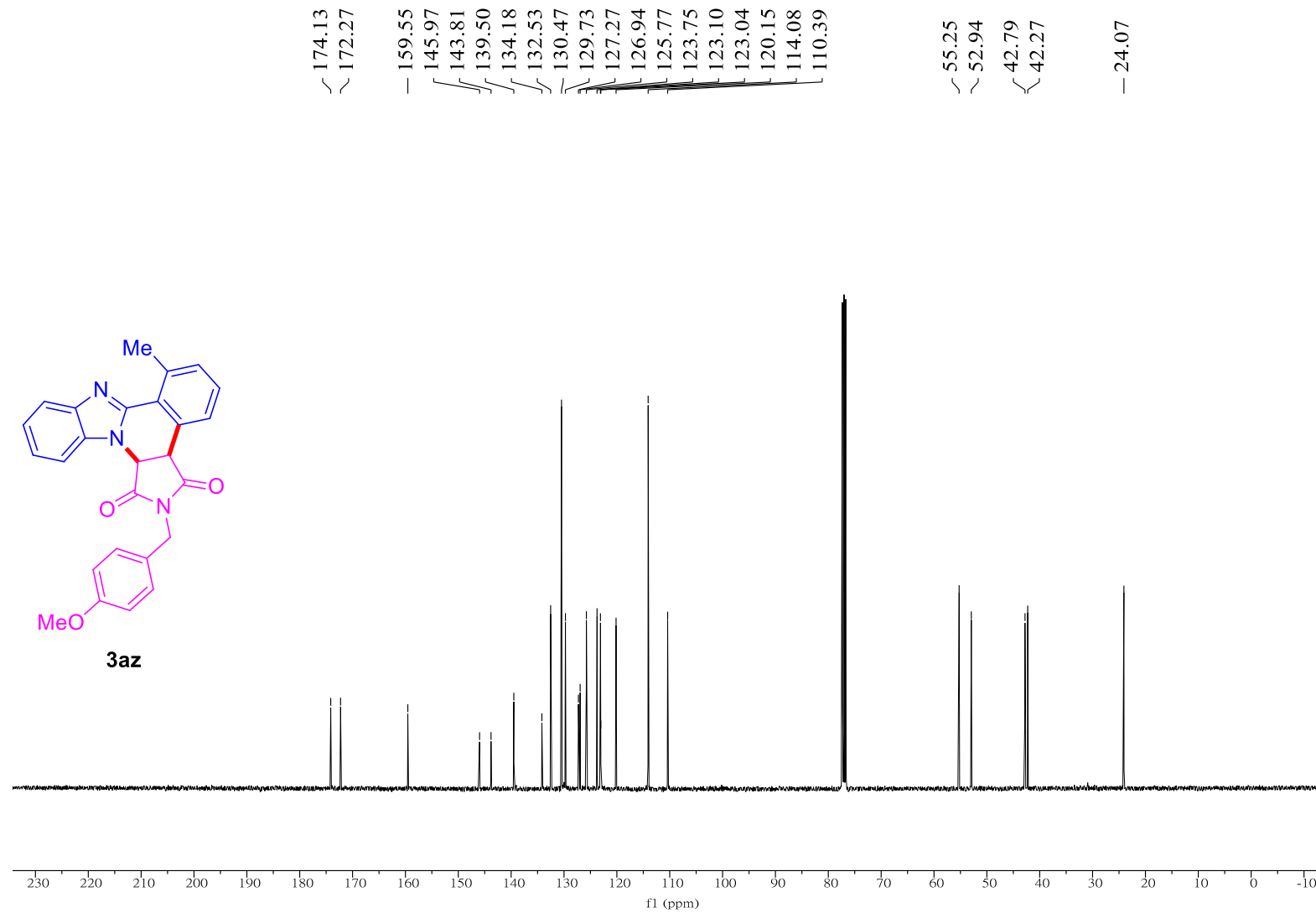
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
424.1662	1	C <sub>26</sub> H <sub>22</sub> N <sub>3</sub> O <sub>3</sub>	424.1656	1.5	25.1	1	100.00	17.5	even	ok	M+H

HRMS (ESI) of compound **5az**.

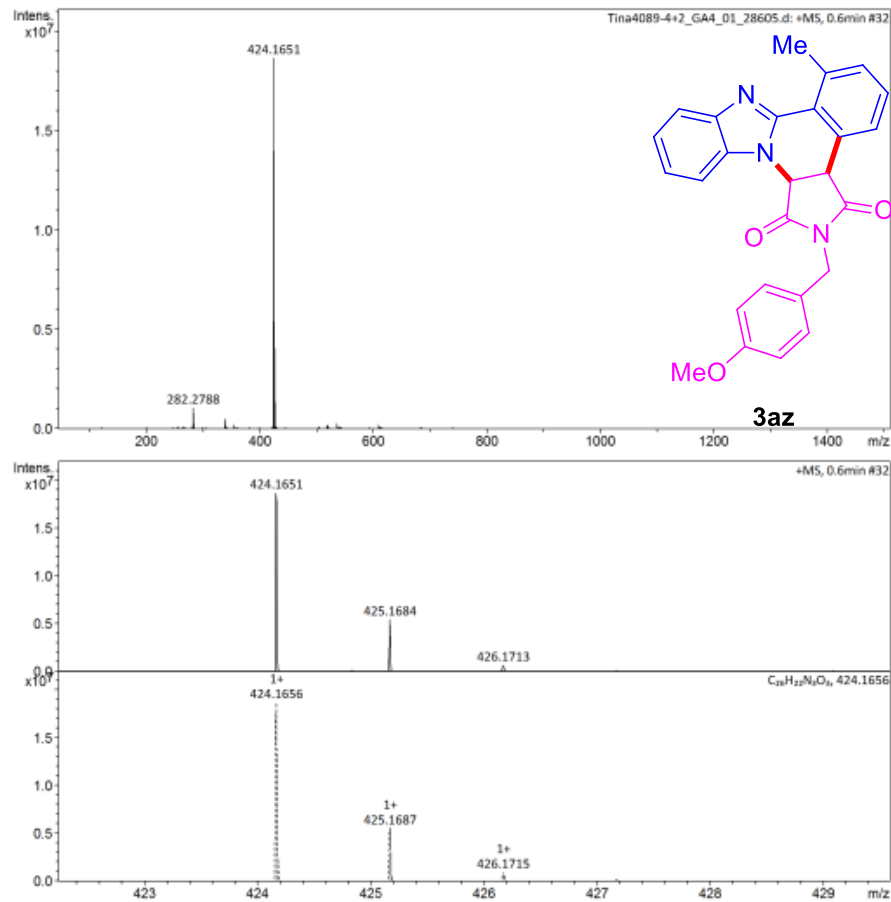




$^1\text{H}$  NMR spectrum (400 MHz) of compound **3az** in  $\text{CDCl}_3$ .



$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3az** in  $\text{CDCl}_3$ .

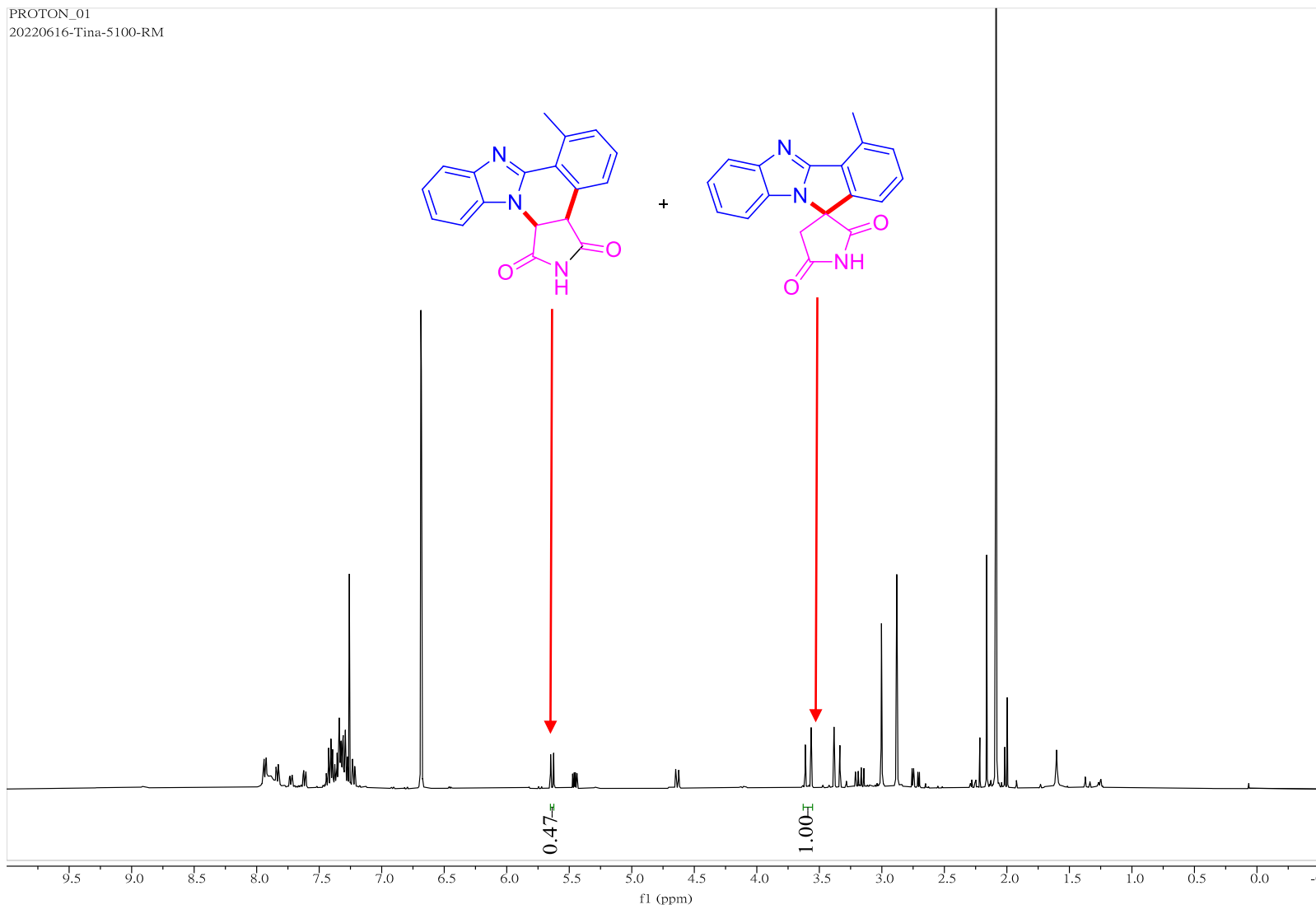


### Display Report

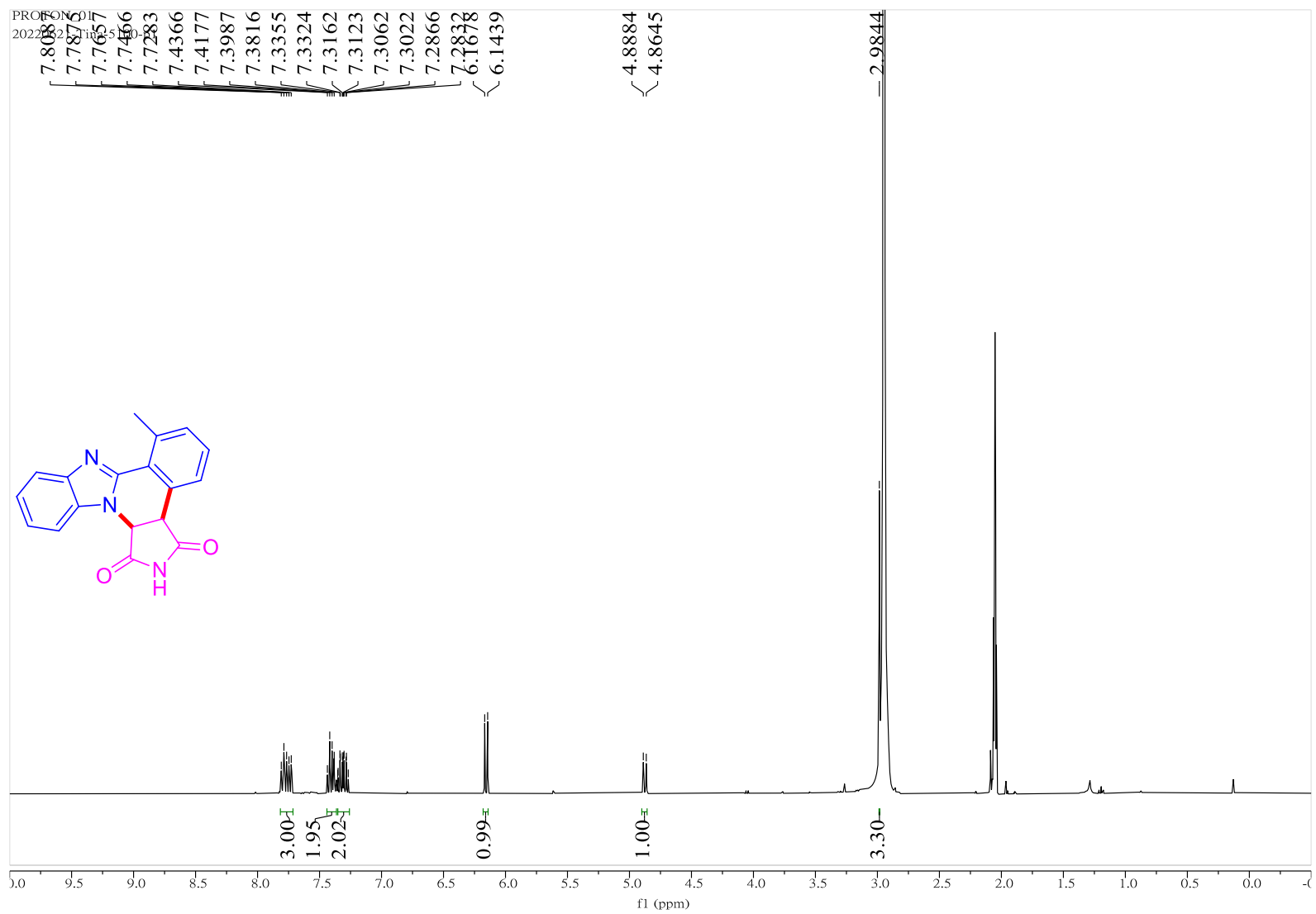
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdB	e <sup>-</sup> Conf	N-Rule	Adduct
424.1651	1	C <sub>26</sub> H <sub>22</sub> N <sub>3</sub> O <sub>3</sub>	424.1656	1.1	7.1	1	100.00	17.5	even	ok	M+H

HRMS (ESI) of compound **3az**.

PROTON\_01  
20220616-Tina-5100-RM

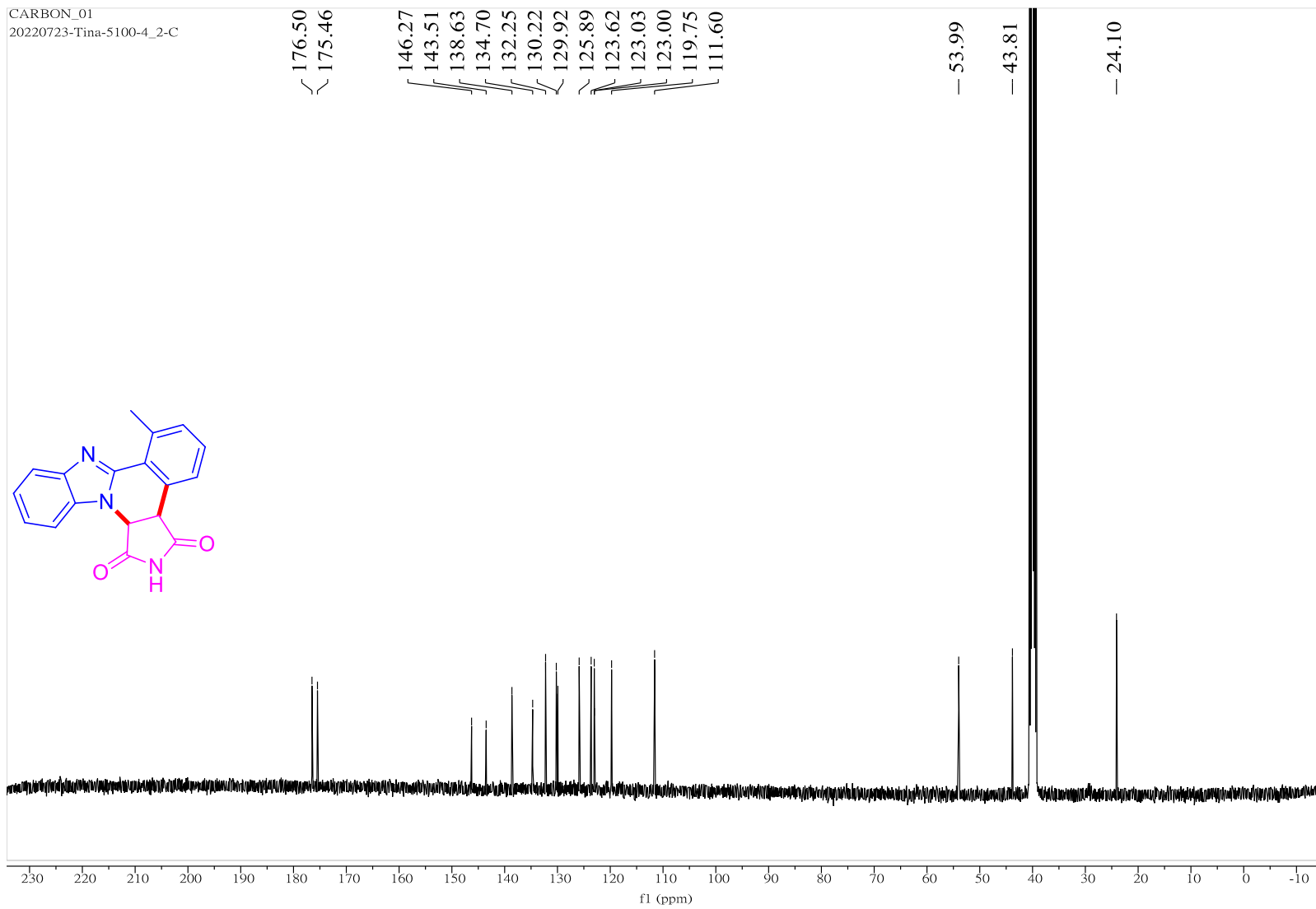


$^1\text{H}$  NMR spectrum (400 MHz) of compound **5bc** and **3bc** in  $\text{CDCl}_3$ .

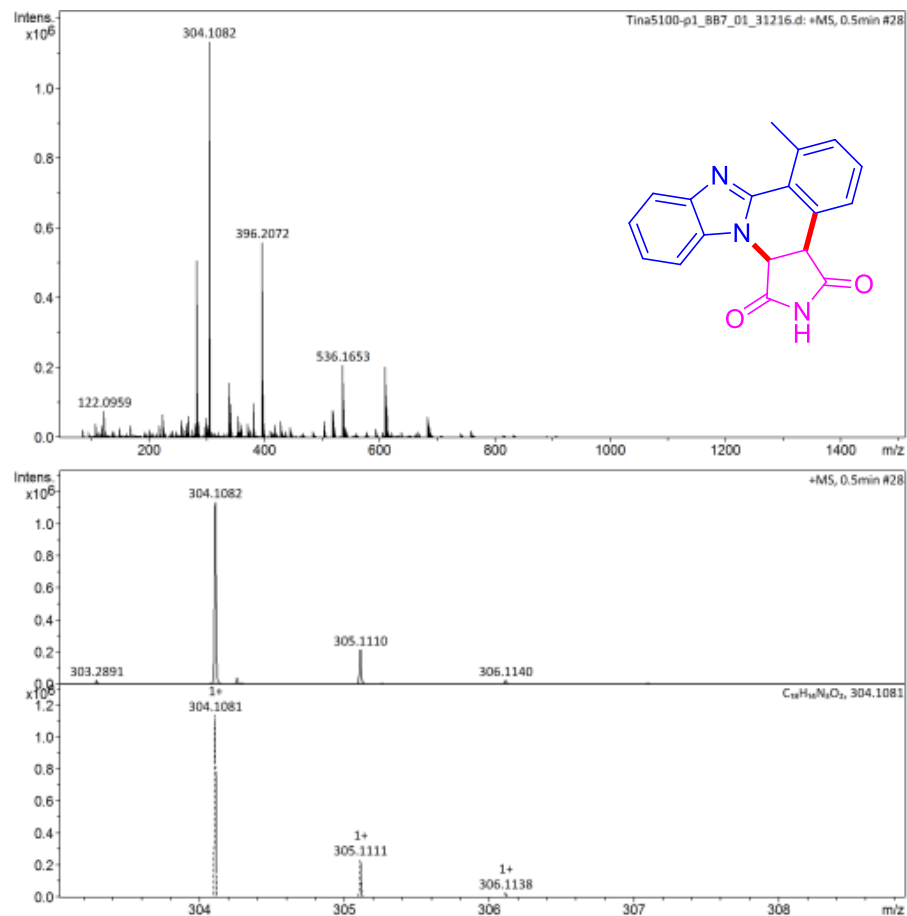


$^1\text{H}$  NMR spectrum (400 MHz) of compound **3bc** in acetone- $d_6$ .

CARBON\_01  
20220723-Tina-5100-4\_2-C



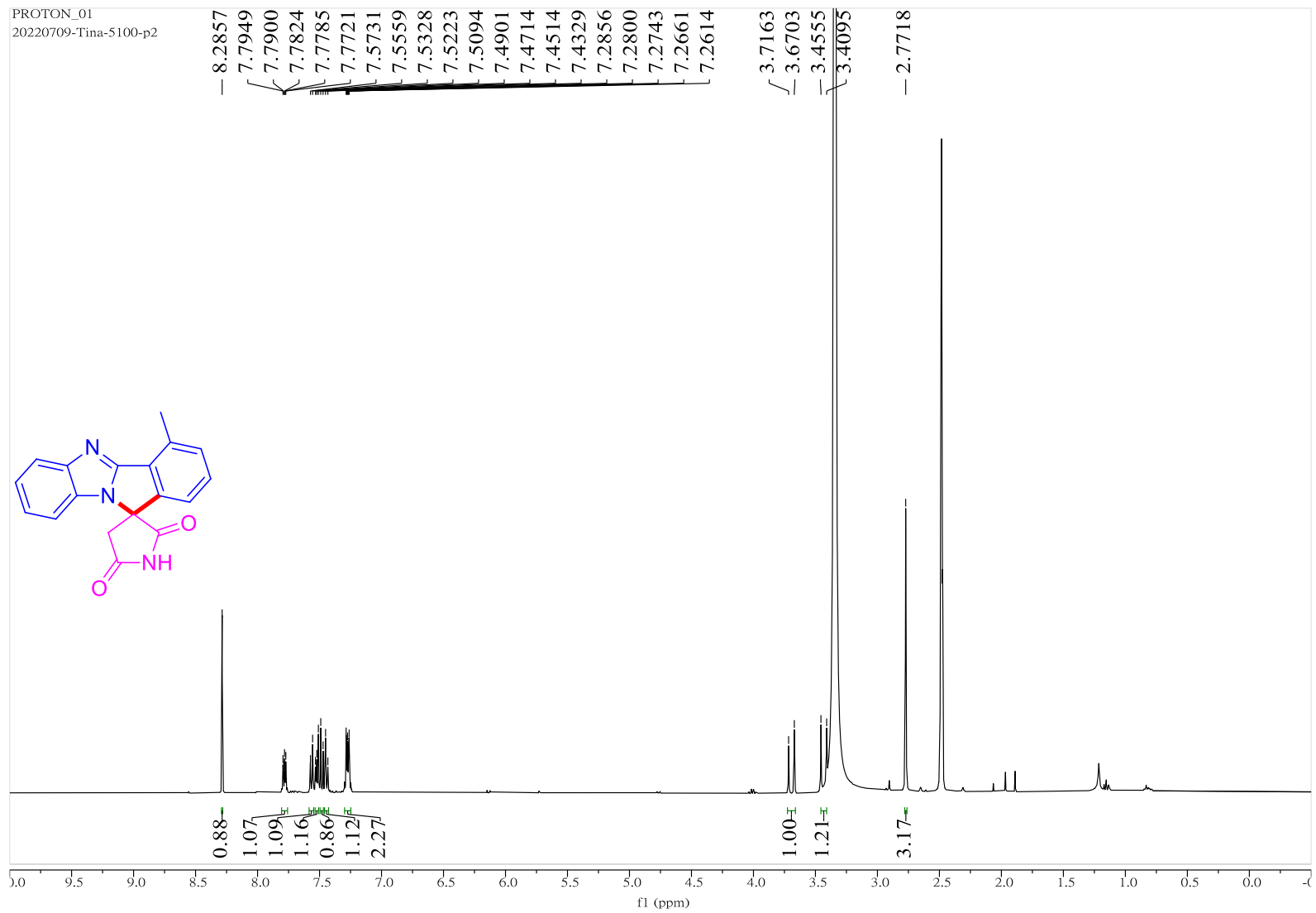
$^{13}\text{C}$  NMR spectrum (150 MHz) of compound **3bc** in  $\text{DMSO-}d_6$ .



### Display Report

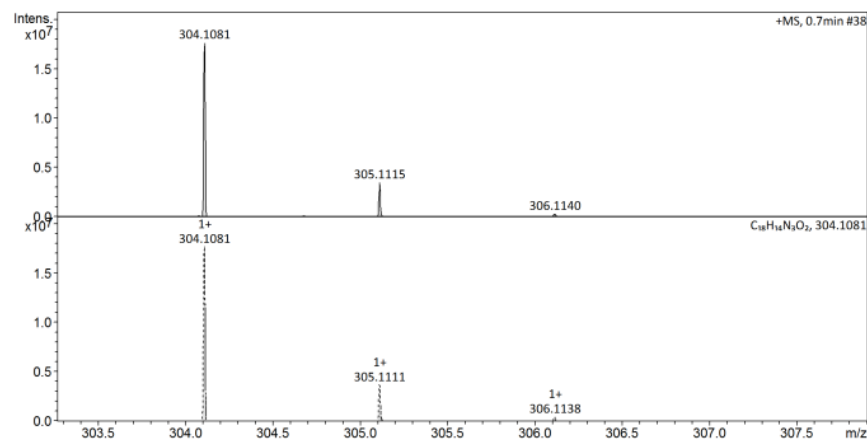
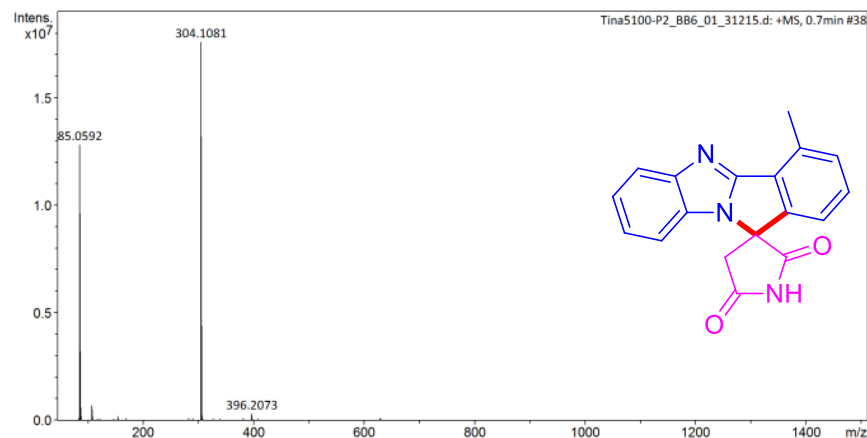
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
304.1082	1	C <sub>18</sub> H <sub>14</sub> N <sub>3</sub> O <sub>2</sub>	304.1081	-0.5	10.5	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **3bc**



$^1\text{H}$  NMR spectrum (400 MHz) of compound **5bc** in  $\text{DMSO-}d_6$ .



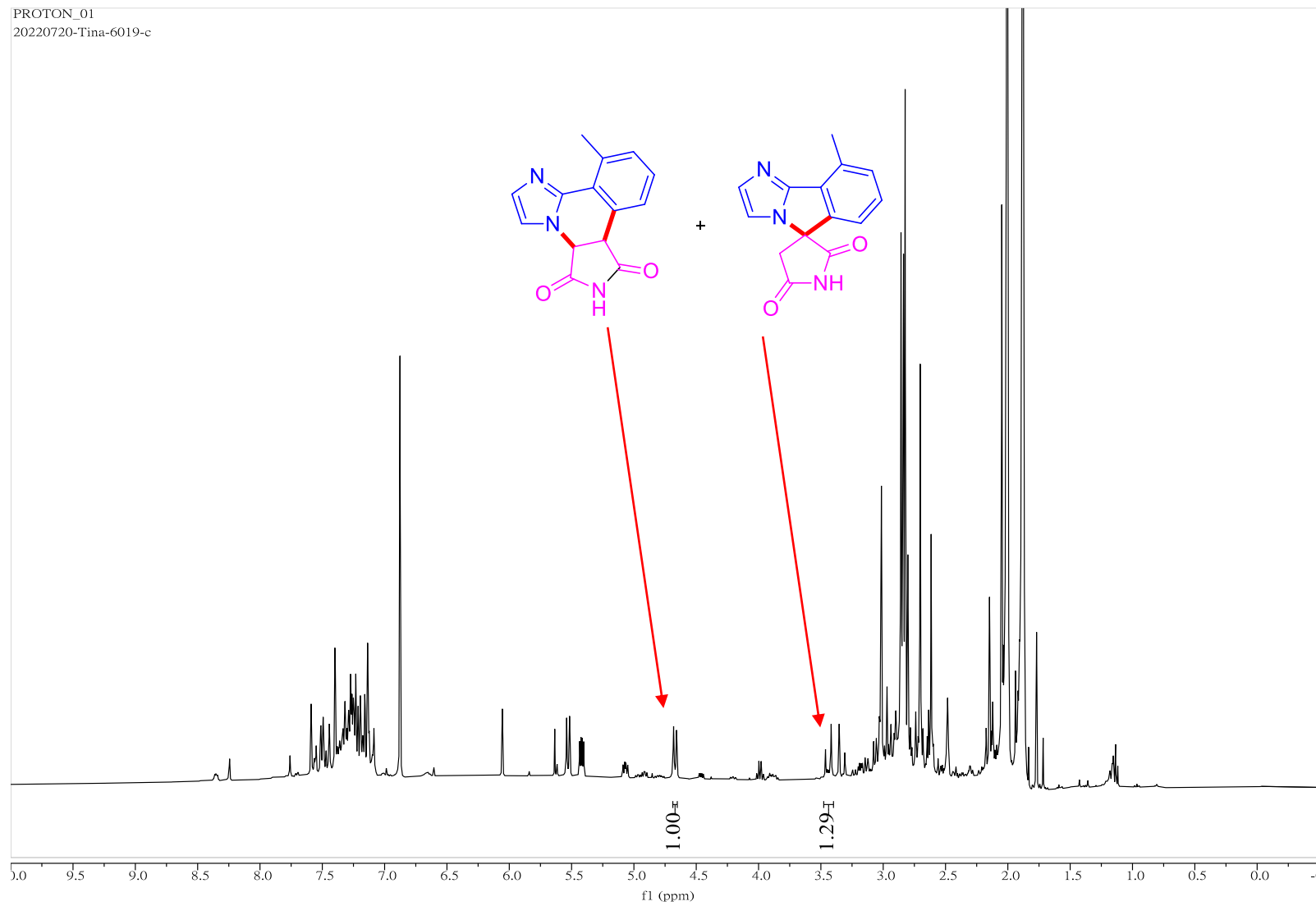


## Display Report

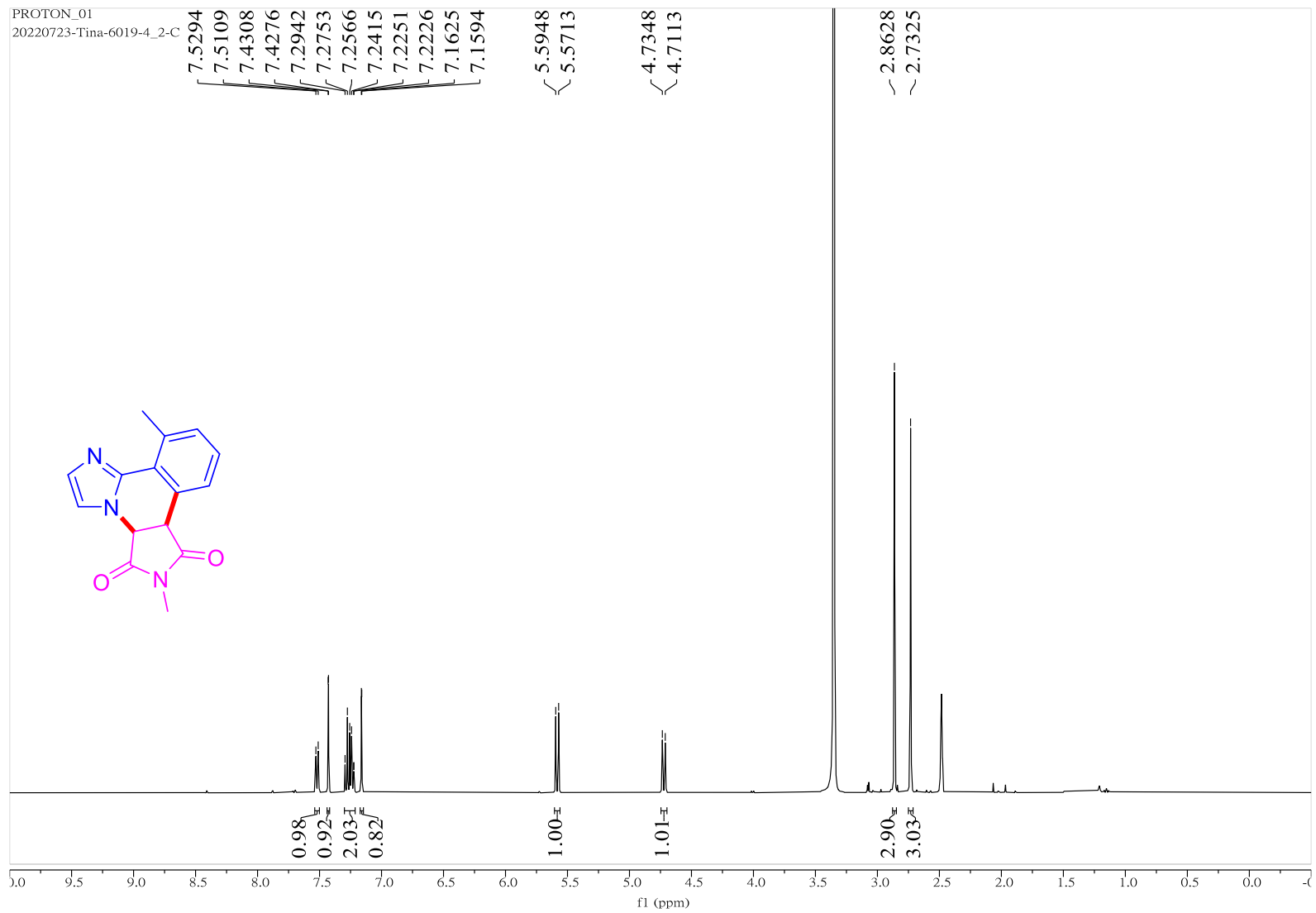
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
304.1081	1	C <sub>18</sub> H <sub>14</sub> N <sub>2</sub> O <sub>2</sub>	304.1081	0.2	8.2	1	100.00	13.5	even	ok	M+H

HRMS (ESI) of compound **5bc**

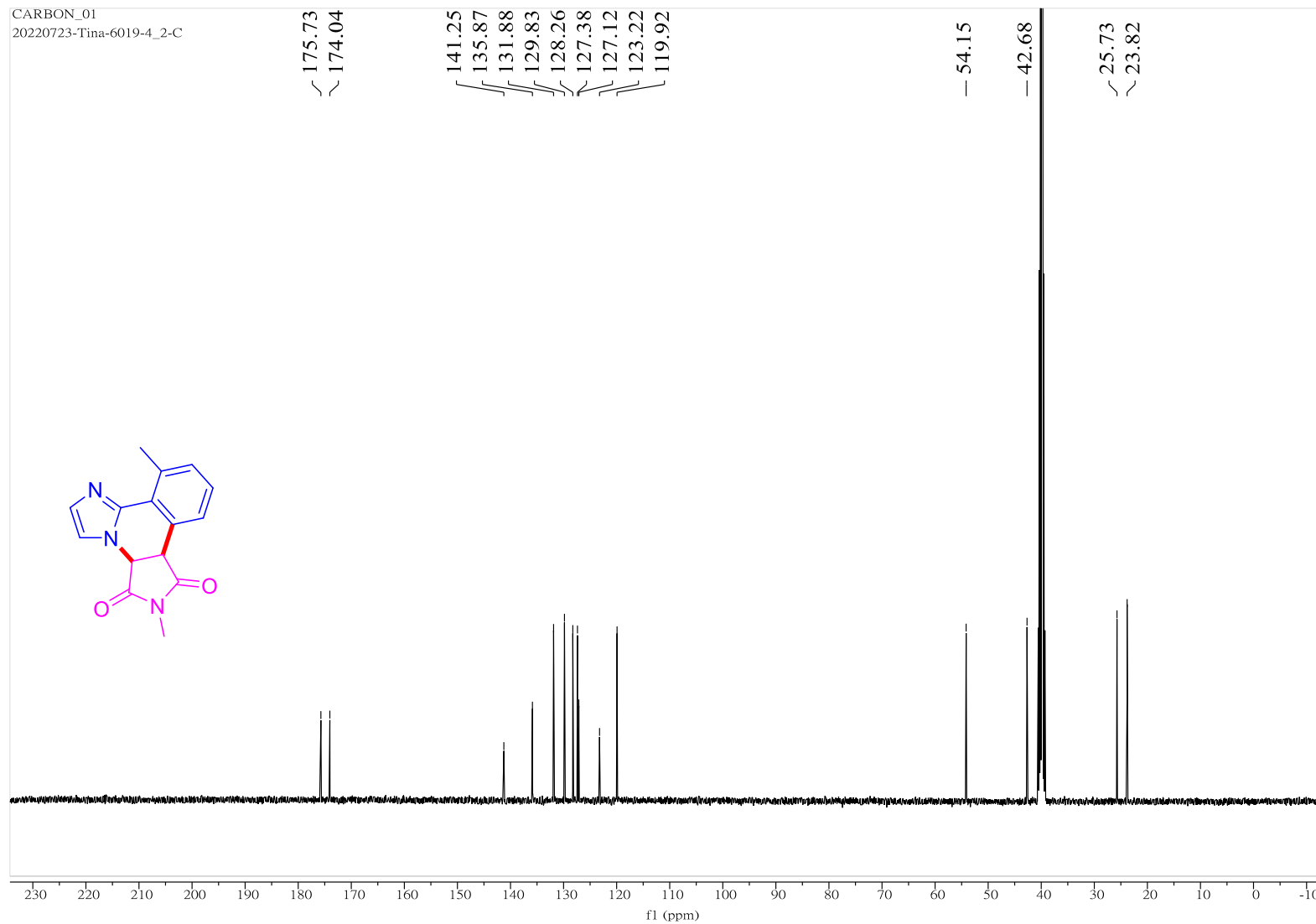
PROTON\_01  
20220720-Tina-6019-c



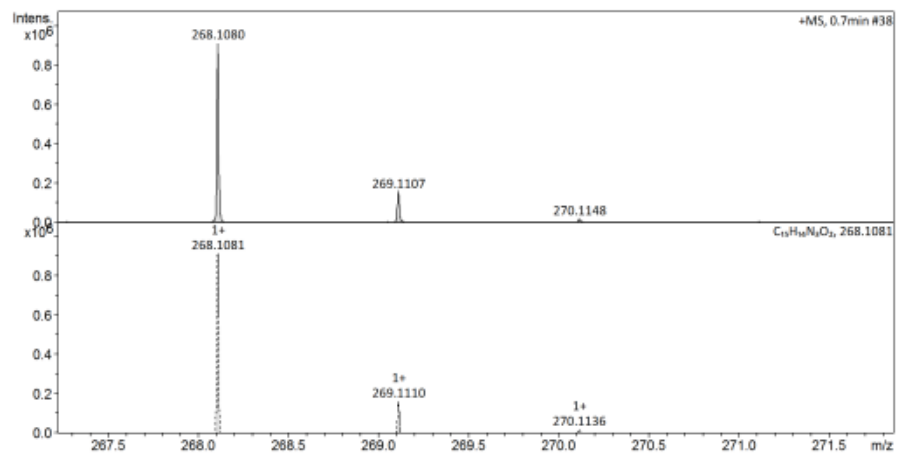
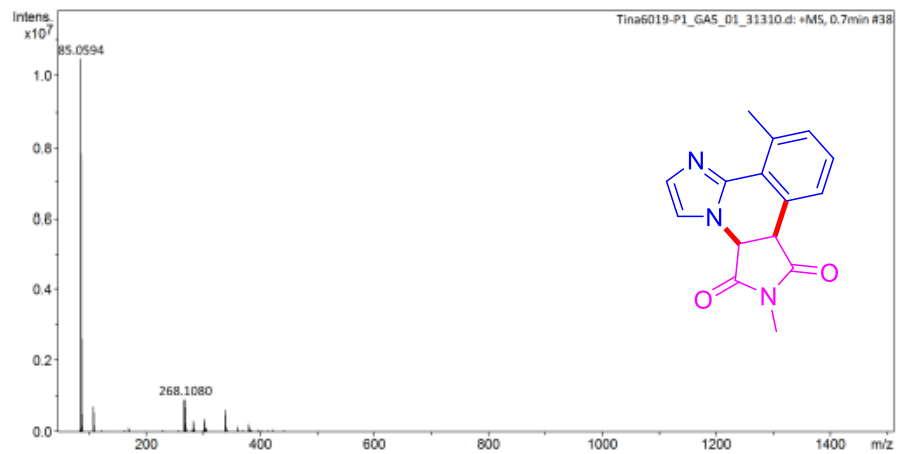
$^1\text{H}$  NMR spectrum (400 MHz) of compound **5bd** and **3bd** in  $\text{DMSO-}d_6$ .



$^1\text{H}$  NMR spectrum (400 MHz) of compound **3bd** in  $\text{DMSO-}d_6$ .



$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **3bd** in  $\text{DMSO-}d_6$ .

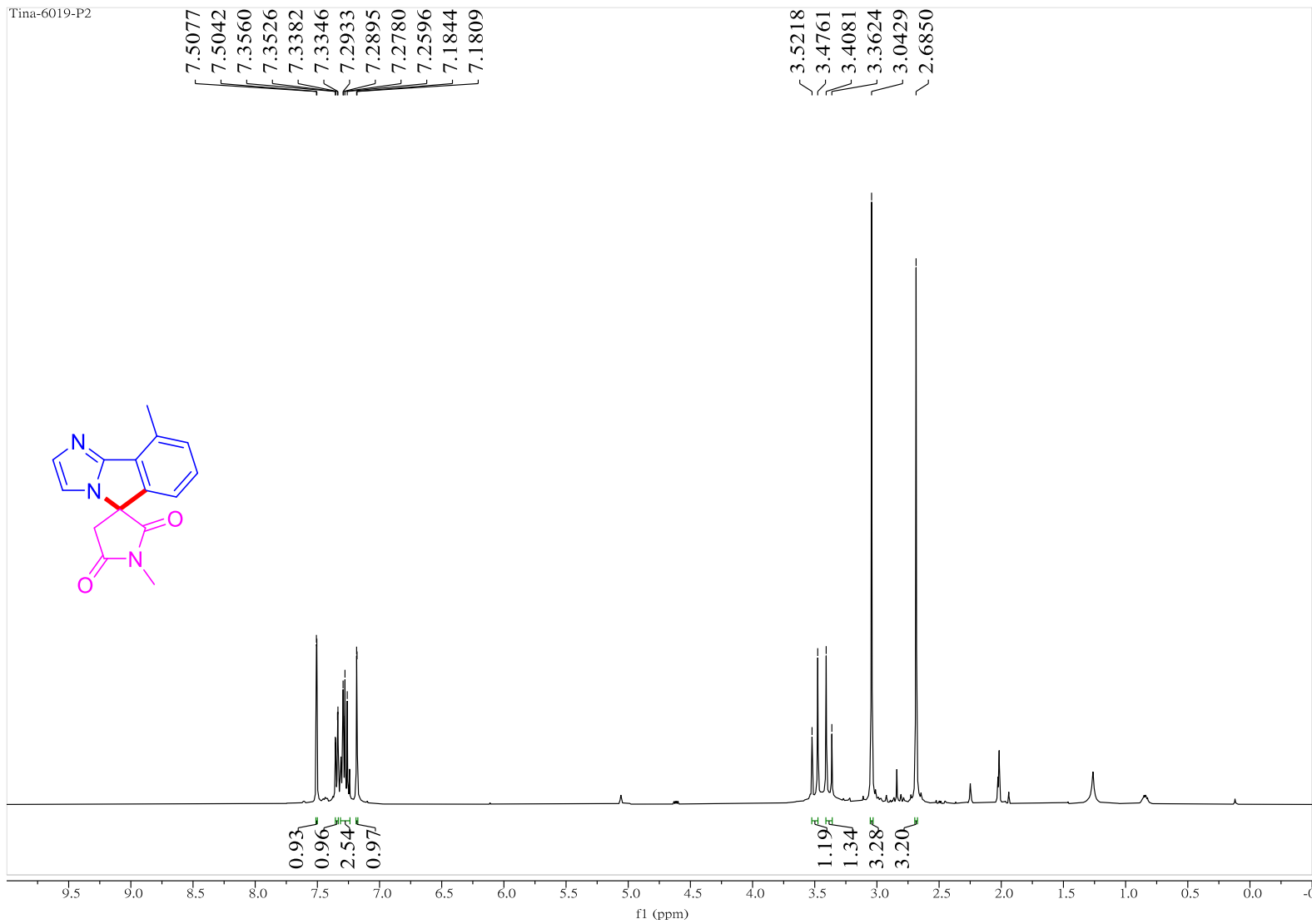


### Display Report

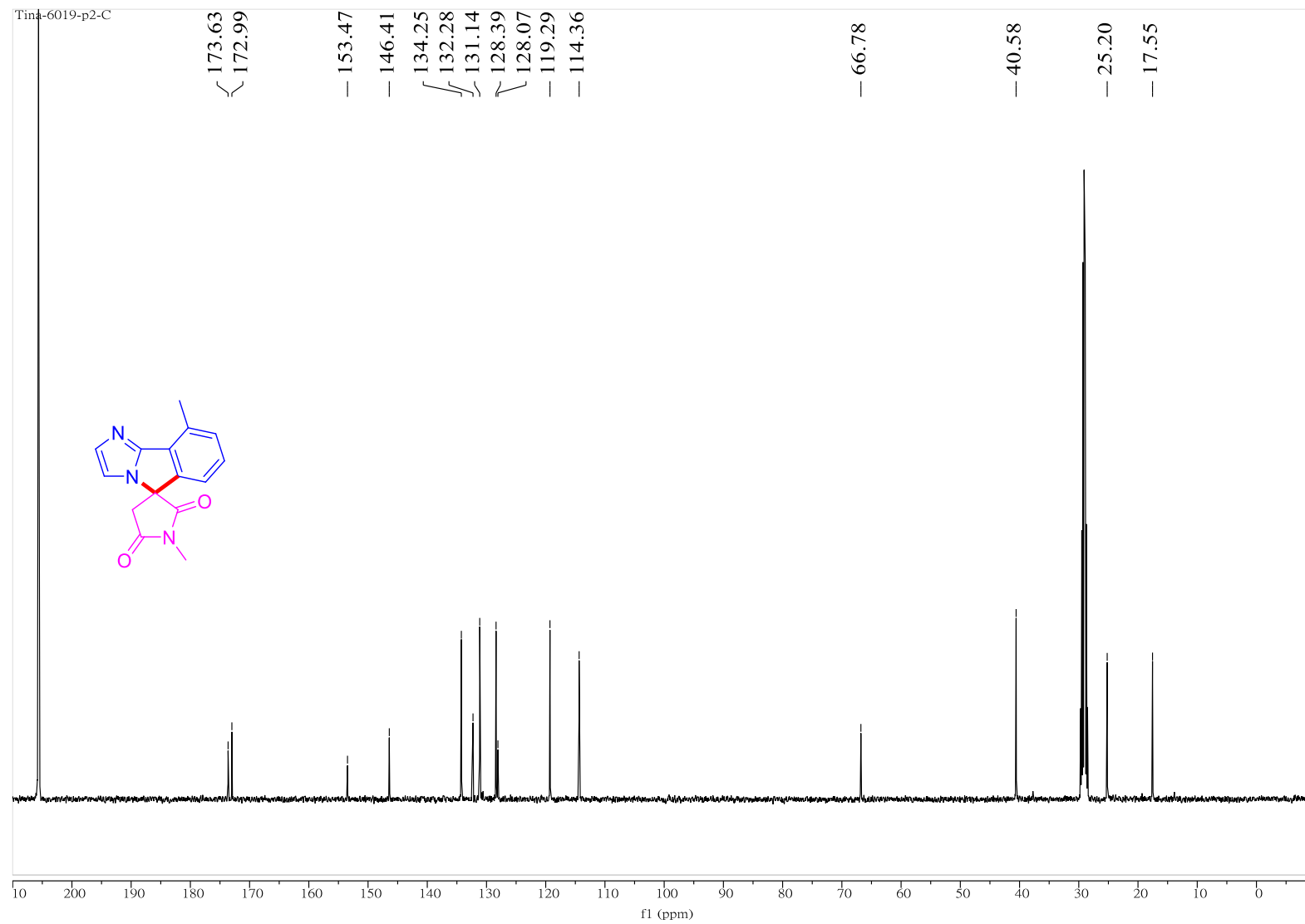
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
268.1080	1	C <sub>15</sub> H <sub>14</sub> N <sub>2</sub> O <sub>2</sub>	268.1081	0.1	4.4	1	100.00	10.5	even	ok	M+H

HRMS (ESI) of compound **3bd**

Tina-6019-P2

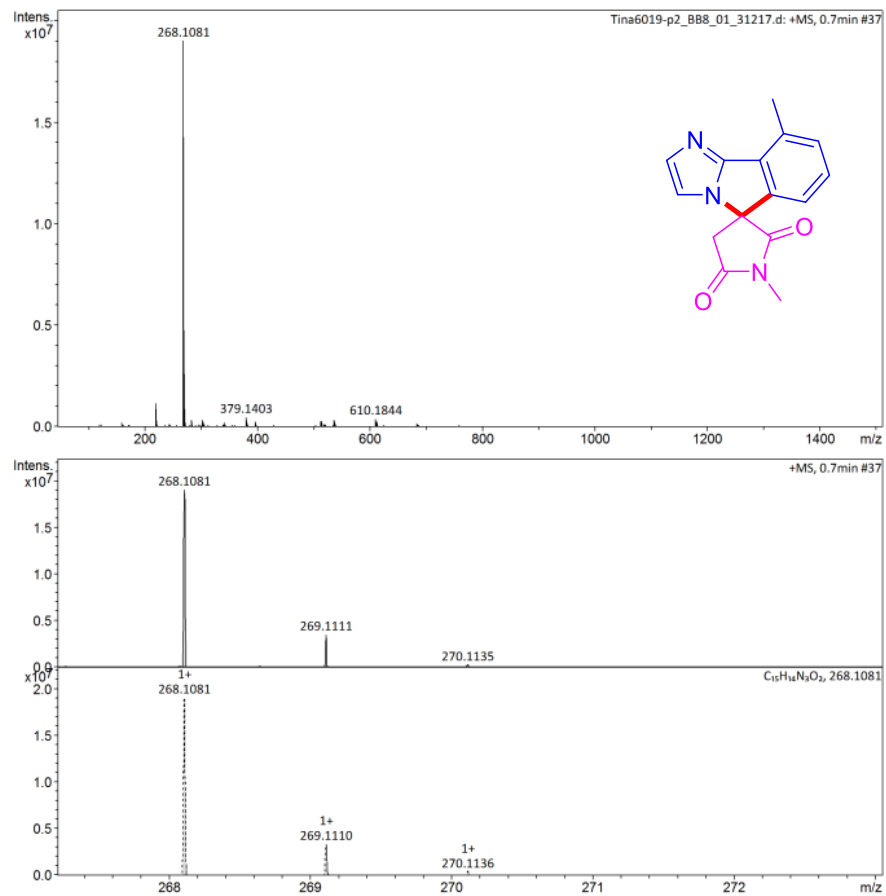


$^1\text{H}$  NMR spectrum (400 MHz) of compound **5bd** in  $d_6$ -acetone.





$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **5bd** in  $d_6$ -acetone.



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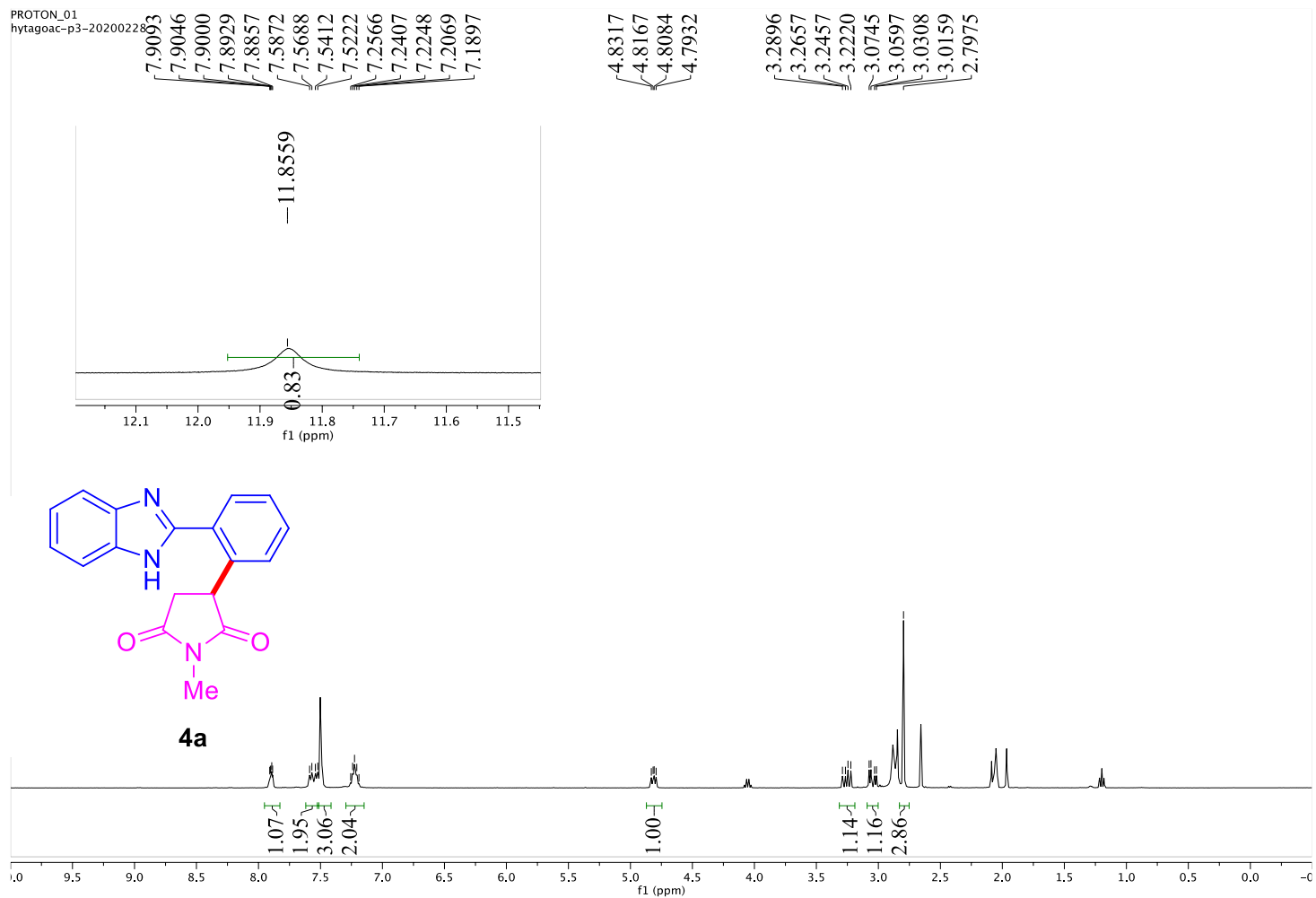
## Display Report

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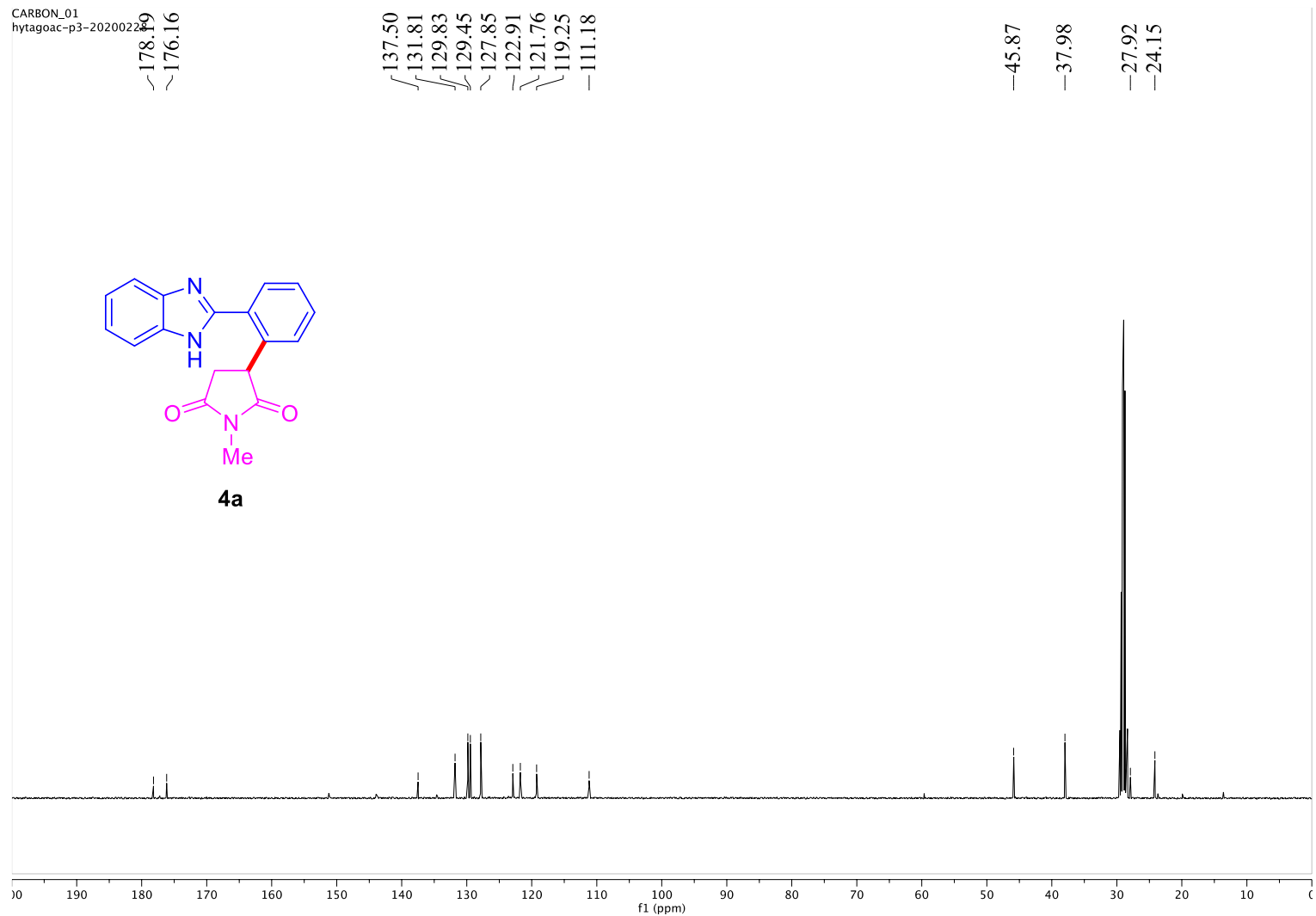
Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
268.1081	1	C15H14N3O2	268.1081	0.2	5.1	1	100.00	10.5	even	ok	M+H

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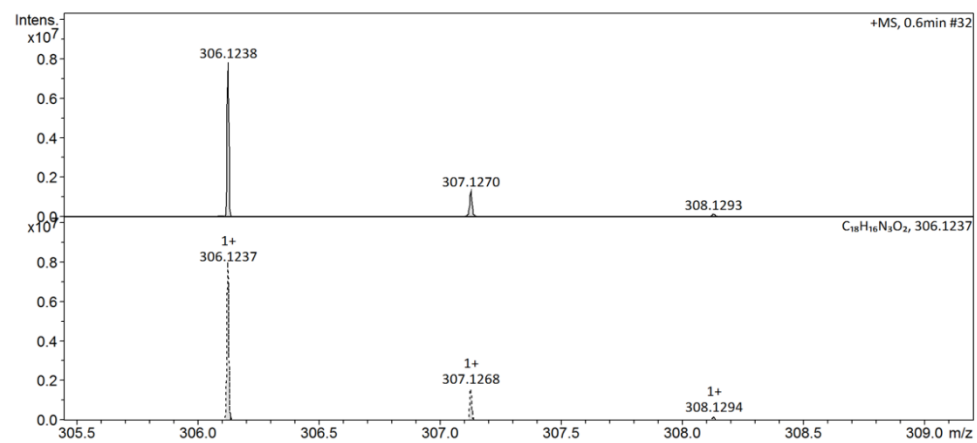
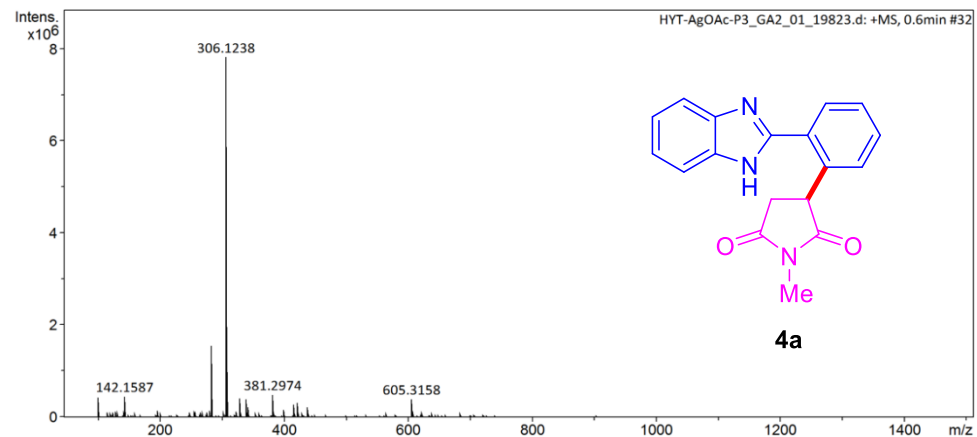
HRMS (ESI) of compound **5bd**



**<sup>1</sup>H NMR spectrum (400 MHz) of compound 4a in acetone-*d*<sub>6</sub>**



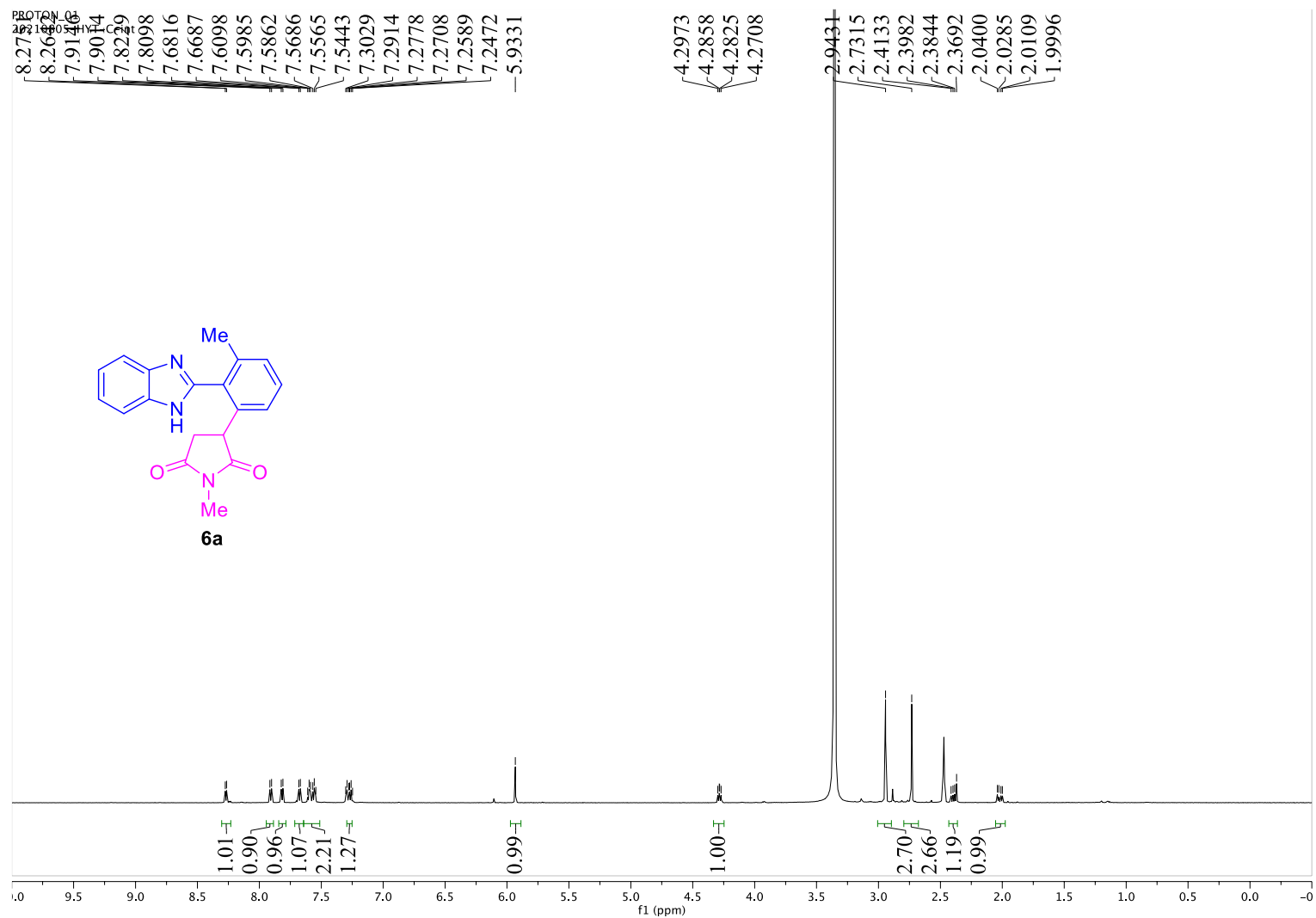
$^{13}\text{C}$  NMR spectrum (100 MHz) of compound **4a** in acetone- $d_6$ .



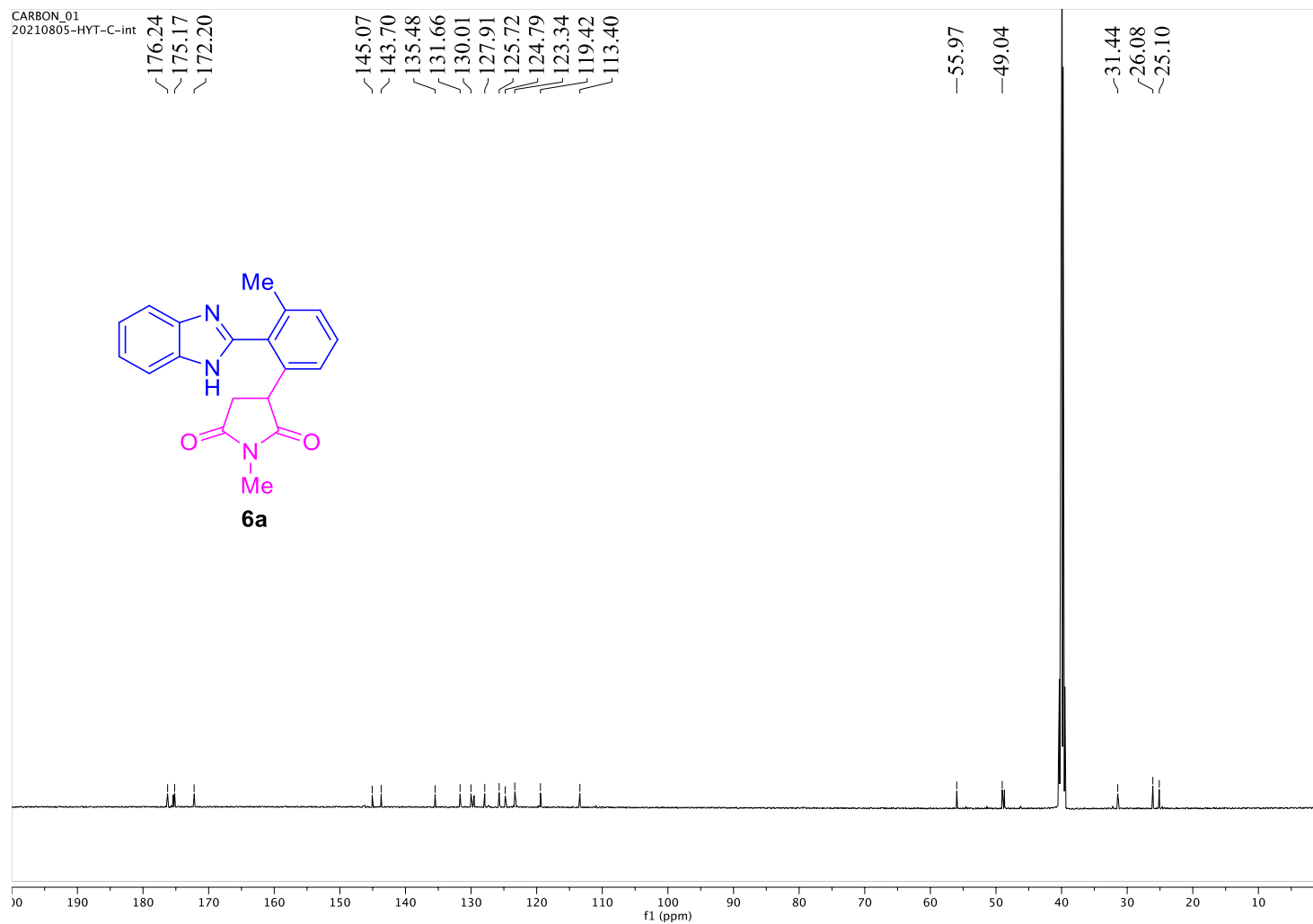
## Display Report

Meas. m/z	#	Ion Formula	m/z	err [ppm]	mSigma	# Sigma	Score	rdb	e <sup>-</sup> Conf	N-Rule	Adduct
306.1238	1	C <sub>18</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub>	306.1237	-0.3	24.2	1	100.00	12.5	even	ok	M+H

HRMS (ESI) of compound **4a**.

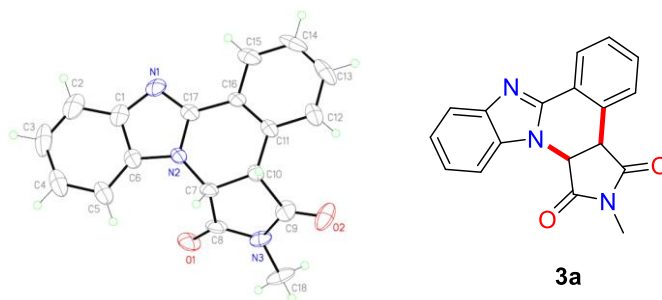


<sup>1</sup>H NMR spectrum (600 MHz) of compound **6a** in DMSO-*d*<sub>6</sub>.



$^{13}\text{C}$  NMR spectrum (150 MHz) of compound **6a** in  $\text{DMSO-}d_6$ .

## X-ray crystallographic data of compound 3a



**ORTEP diagram of compound 3a.** Atomic displacement ellipsoids are drawn at the 50% probability level

### CCDC No. 2133408

Table S1. Crystal data and structure refinement for 200218lt\_0m\_a\_pl.

Identification code	200218lt_0m_a_pl	
Empirical formula	C <sub>18</sub> H <sub>13</sub> N <sub>3</sub> O <sub>2</sub>	
Formula weight	303.31	
Temperature	100(2) K	
Wavelength	0.71073 Å	
Crystal system	Orthorhombic	
Space group	Pbca	
Unit cell dimensions	a = 16.2326(10) Å	α = 90°.
	b = 8.5326(5) Å	β = 90°.
	c = 20.3607(13) Å	γ = 90°.
Volume	2820.1(3) Å <sup>3</sup>	
Z	8	
Density (calculated)	1.429 Mg/m <sup>3</sup>	
Absorption coefficient	0.096 mm <sup>-1</sup>	
F(000)	1264	
Crystal size	0.12 x 0.11 x 0.04 mm <sup>3</sup>	
Theta range for data collection	2.000 to 26.452°.	
Index ranges	-20 ≤ h ≤ 19, -10 ≤ k ≤ 10, -25 ≤ l ≤ 25	
Reflections collected	18944	
Independent reflections	2877 [R(int) = 0.0534]	
Completeness to theta = 25.242°	100.0 %	
Absorption correction	Semi-empirical from equivalents	
Max. and min. transmission	0.7454 and 0.5458	



Refinement method	Full-matrix least-squares on F <sup>2</sup>
Data / restraints / parameters	2877 / 0 / 209
Goodness-of-fit on F <sup>2</sup>	1.205
Final R indices [I>2sigma(I)]	R1 = 0.1443, wR2 = 0.3270
R indices (all data)	R1 = 0.1653, wR2 = 0.3365
Extinction coefficient	n/a
Largest diff. peak and hole	0.425 and -0.563 e.Å <sup>-3</sup>

Table S2. Atomic coordinates ( x 10<sup>4</sup>) and equivalent isotropic displacement parameters (Å<sup>2</sup>x 10<sup>3</sup>) for 200218lt\_0m\_a\_pl. U(eq) is defined as one third of the trace of the orthogonalized U<sup>ij</sup> tensor.

	x	y	z	U(eq)
C(1)	953(4)	2124(9)	6047(3)	33(2)
C(2)	940(5)	1404(12)	6665(4)	53(3)
C(3)	1520(5)	1817(16)	7124(4)	75(4)
C(4)	2103(5)	2988(14)	6988(4)	59(3)
C(5)	2130(4)	3768(13)	6391(3)	49(3)
C(6)	1544(4)	3321(10)	5936(3)	35(2)
C(7)	1743(4)	5166(9)	4974(3)	28(2)
C(8)	2598(4)	4694(11)	4702(4)	36(2)
C(9)	1952(5)	6114(11)	3887(4)	40(2)
C(10)	1266(4)	5759(8)	4380(3)	26(1)
C(11)	628(4)	4663(10)	4091(3)	29(2)
C(12)	273(4)	5005(11)	3481(3)	40(2)
C(13)	-346(5)	4014(13)	3240(4)	52(3)
C(14)	-629(4)	2747(13)	3593(4)	51(3)
C(15)	-281(4)	2407(11)	4194(4)	40(2)
C(16)	348(4)	3377(9)	4450(3)	29(2)
C(17)	699(4)	2987(10)	5093(3)	30(2)
C(18)	3431(5)	5459(10)	3711(4)	46(2)
N(1)	437(3)	1948(8)	5513(3)	33(1)
N(2)	1361(3)	3859(7)	5315(2)	26(1)
N(3)	2676(3)	5436(8)	4104(3)	34(2)
O(1)	3099(3)	3883(7)	4962(3)	42(1)
O(2)	1891(4)	6865(7)	3381(3)	50(2)

Table S3. Bond lengths [Å] and angles [°] for 200218lt\_0m\_a\_pl.

---

C(1)-N(1)	1.381(9)
C(1)-C(2)	1.399(11)
C(1)-C(6)	1.420(11)
C(2)-C(3)	1.373(14)
C(2)-H(2)	0.9500
C(3)-C(4)	1.404(16)
C(3)-H(3)	0.9500
C(4)-C(5)	1.387(12)
C(4)-H(4)	0.9500
C(5)-C(6)	1.381(10)
C(5)-H(5)	0.9500
C(6)-N(2)	1.377(9)
C(7)-N(2)	1.453(9)
C(7)-C(10)	1.523(9)
C(7)-C(8)	1.548(9)
C(7)-H(7)	1.0000
C(8)-O(1)	1.192(9)
C(8)-N(3)	1.377(10)
C(9)-O(2)	1.217(10)
C(9)-N(3)	1.382(10)
C(9)-C(10)	1.530(9)
C(10)-C(11)	1.514(10)
C(10)-H(10)	1.0000
C(11)-C(16)	1.395(11)
C(11)-C(12)	1.399(9)
C(12)-C(13)	1.402(12)
C(12)-H(12)	0.9500
C(13)-C(14)	1.377(14)
C(13)-H(13)	0.9500
C(14)-C(15)	1.378(12)
C(14)-H(14)	0.9500
C(15)-C(16)	1.414(10)
C(15)-H(15)	0.9500
C(16)-C(17)	1.466(9)
C(17)-N(1)	1.304(9)
C(17)-N(2)	1.383(9)

C(18)-N(3)	1.465(8)
C(18)-H(18A)	0.9800
C(18)-H(18B)	0.9800
C(18)-H(18C)	0.9800
N(1)-C(1)-C(2)	130.6(8)
N(1)-C(1)-C(6)	111.3(6)
C(2)-C(1)-C(6)	118.0(8)
C(3)-C(2)-C(1)	119.4(9)
C(3)-C(2)-H(2)	120.3
C(1)-C(2)-H(2)	120.3
C(2)-C(3)-C(4)	120.6(8)
C(2)-C(3)-H(3)	119.7
C(4)-C(3)-H(3)	119.7
C(5)-C(4)-C(3)	122.4(8)
C(5)-C(4)-H(4)	118.8
C(3)-C(4)-H(4)	118.8
C(6)-C(5)-C(4)	115.7(9)
C(6)-C(5)-H(5)	122.1
C(4)-C(5)-H(5)	122.1
N(2)-C(6)-C(5)	132.3(8)
N(2)-C(6)-C(1)	103.9(6)
C(5)-C(6)-C(1)	123.8(7)
N(2)-C(7)-C(10)	114.6(5)
N(2)-C(7)-C(8)	110.8(6)
C(10)-C(7)-C(8)	105.0(5)
N(2)-C(7)-H(7)	108.8
C(10)-C(7)-H(7)	108.8
C(8)-C(7)-H(7)	108.8
O(1)-C(8)-N(3)	126.7(6)
O(1)-C(8)-C(7)	127.1(7)
N(3)-C(8)-C(7)	106.2(6)
O(2)-C(9)-N(3)	124.0(7)
O(2)-C(9)-C(10)	126.9(7)
N(3)-C(9)-C(10)	109.0(6)
C(11)-C(10)-C(7)	116.8(6)
C(11)-C(10)-C(9)	111.4(6)
C(7)-C(10)-C(9)	102.5(5)

C(11)-C(10)-H(10)	108.6
C(7)-C(10)-H(10)	108.6
C(9)-C(10)-H(10)	108.6
C(16)-C(11)-C(12)	119.6(7)
C(16)-C(11)-C(10)	120.4(6)
C(12)-C(11)-C(10)	119.9(7)
C(11)-C(12)-C(13)	118.7(8)
C(11)-C(12)-H(12)	120.6
C(13)-C(12)-H(12)	120.6
C(14)-C(13)-C(12)	122.0(7)
C(14)-C(13)-H(13)	119.0
C(12)-C(13)-H(13)	119.0
C(13)-C(14)-C(15)	119.4(7)
C(13)-C(14)-H(14)	120.3
C(15)-C(14)-H(14)	120.3
C(14)-C(15)-C(16)	120.0(9)
C(14)-C(15)-H(15)	120.0
C(16)-C(15)-H(15)	120.0
C(11)-C(16)-C(15)	120.2(7)
C(11)-C(16)-C(17)	121.3(6)
C(15)-C(16)-C(17)	118.5(7)
N(1)-C(17)-N(2)	113.9(6)
N(1)-C(17)-C(16)	127.9(6)
N(2)-C(17)-C(16)	118.2(7)
N(3)-C(18)-H(18A)	109.5
N(3)-C(18)-H(18B)	109.5
H(18A)-C(18)-H(18B)	109.5
N(3)-C(18)-H(18C)	109.5
H(18A)-C(18)-H(18C)	109.5
H(18B)-C(18)-H(18C)	109.5
C(17)-N(1)-C(1)	104.2(6)
C(6)-N(2)-C(17)	106.8(6)
C(6)-N(2)-C(7)	127.0(6)
C(17)-N(2)-C(7)	126.1(6)
C(8)-N(3)-C(9)	113.4(6)
C(8)-N(3)-C(18)	124.4(7)
C(9)-N(3)-C(18)	122.1(7)

Symmetry transformations used to generate equivalent atoms:

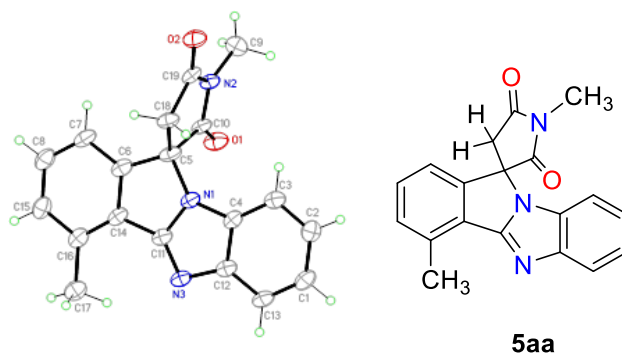
Table S4. Anisotropic displacement parameters ( $\text{\AA}^2 \times 10^3$ ) for 200218lt\_0m\_a\_pl. The anisotropic displacement factor exponent takes the form:  $-2\pi^2 [ h^2 a^{*2} U^{11} + \dots + 2 h k a^* b^* U^{12} ]$

	$U^{11}$	$U^{22}$	$U^{33}$	$U^{23}$	$U^{13}$	$U^{12}$
C(1)	30(3)	38(4)	31(4)	0(3)	13(3)	16(3)
C(2)	38(4)	71(7)	50(5)	19(5)	22(4)	23(4)
C(3)	44(5)	146(12)	35(5)	35(6)	21(4)	52(6)
C(4)	31(4)	121(9)	24(4)	-1(5)	4(3)	33(5)
C(5)	20(3)	103(8)	23(3)	-7(4)	5(3)	19(4)
C(6)	20(3)	64(5)	22(3)	-3(3)	7(3)	11(3)
C(7)	15(3)	40(4)	29(3)	-5(3)	3(3)	-1(3)
C(8)	13(3)	56(5)	40(4)	-4(4)	1(3)	-5(3)
C(9)	31(4)	56(5)	32(4)	-5(4)	11(3)	9(4)
C(10)	20(3)	33(4)	24(3)	0(3)	1(2)	4(3)
C(11)	14(3)	54(5)	20(3)	-9(3)	3(2)	11(3)
C(12)	22(3)	73(6)	25(3)	-4(4)	2(3)	22(4)
C(13)	26(4)	98(8)	32(4)	-24(5)	-10(3)	33(5)
C(14)	17(3)	92(8)	44(5)	-43(5)	-1(3)	-4(4)
C(15)	17(3)	68(6)	35(4)	-23(4)	7(3)	0(4)
C(16)	13(3)	53(5)	22(3)	-8(3)	6(2)	0(3)
C(17)	14(3)	52(5)	24(3)	-11(3)	7(2)	6(3)
C(18)	31(4)	42(5)	64(5)	-9(4)	30(4)	-8(4)
N(1)	24(3)	42(4)	34(3)	-2(3)	12(2)	1(3)
N(2)	15(2)	43(4)	19(3)	-4(2)	2(2)	4(2)
N(3)	20(3)	43(4)	40(3)	-2(3)	14(2)	0(3)
O(1)	17(2)	59(4)	50(3)	-1(3)	-2(2)	9(2)
O(2)	55(3)	55(4)	41(3)	14(3)	27(3)	19(3)

Table S5. Hydrogen coordinates ( $\times 10^4$ ) and isotropic displacement parameters ( $\text{\AA}^2 \times 10^{-3}$ ) for 200218lt\_0m\_a\_pl.

	x	y	z	U(eq)
H(2)	535	638	6765	64
H(3)	1526	1305	7539	90
H(4)	2493	3255	7317	70
H(5)	2526	4559	6301	59
H(7)	1817	6050	5291	34
H(10)	989	6766	4501	31
H(12)	448	5891	3236	48
H(13)	-577	4223	2821	63
H(14)	-1061	2113	3424	61
H(15)	-463	1520	4436	48
H(18A)	3320	5007	3277	69
H(18B)	3858	4842	3932	69
H(18C)	3621	6542	3660	69

## X-ray crystallographic data of compound 5aa



**ORTEP diagram of compound 5aa.** Atomic displacement ellipsoids are drawn at the 50% probability level

### CCDC No. 2131034

Table S6. Crystal data and structure refinement for 200739lt\_0m.

Identification code	200739LT_0m	
Empirical formula	C <sub>19</sub> H <sub>15</sub> N <sub>3</sub> O <sub>2</sub>	
Formula weight	317.34	
Temperature	100(2) K	
Wavelength	0.71073 Å	
Crystal system	Monoclinic	
Space group	C2/c	
Unit cell dimensions	a = 18.0754(12) Å	α = 90°.
	b = 15.6627(11) Å	β = 91.250(3)°.
	c = 11.5969(6) Å	γ = 90°.
Volume	3282.4(4) Å <sup>3</sup>	
Z	8	
Density (calculated)	1.284 Mg/m <sup>3</sup>	
Absorption coefficient	0.086 mm <sup>-1</sup>	
F(000)	1328	
Crystal size	0.15 x 0.02 x 0.01 mm <sup>3</sup>	
Theta range for data collection	1.721 to 26.336°.	
Index ranges	-22 ≤ h ≤ 22, -19 ≤ k ≤ 19, -14 ≤ l ≤ 9	
Reflections collected	10299	
Independent reflections	3328 [R(int) = 0.0471]	
Completeness to theta = 25.242°	99.9 %	

Absorption correction	Semi-empirical from equivalents
Max. and min. transmission	0.7454 and 0.6768
Refinement method	Full-matrix least-squares on F <sup>2</sup>
Data / restraints / parameters	3328 / 0 / 219
Goodness-of-fit on F <sup>2</sup>	1.060
Final R indices [I>2sigma(I)]	R1 = 0.0688, wR2 = 0.1769
R indices (all data)	R1 = 0.1152, wR2 = 0.2016
Extinction coefficient	n/a
Largest diff. peak and hole	1.131 and -0.244 e.Å <sup>-3</sup>

Table S7. Atomic coordinates ( x 10<sup>4</sup>) and equivalent isotropic displacement parameters (Å<sup>2</sup>x 10<sup>3</sup>) for 200739lt\_0m. U(eq) is defined as one third of the trace of the orthogonalized U<sup>ij</sup> tensor.

	x	y	z	U(eq)
O(1)	364(1)	611(2)	9011(2)	33(1)
O(2)	-684(1)	2698(2)	11183(2)	35(1)
N(1)	1528(2)	1920(2)	9354(2)	25(1)
N(2)	-318(2)	1570(2)	10062(2)	24(1)
N(3)	2677(2)	1706(2)	8677(2)	26(1)
C(1)	1976(2)	3159(2)	6338(3)	34(1)
C(2)	1264(2)	3299(2)	6758(3)	33(1)
C(3)	1036(2)	2912(2)	7773(3)	30(1)
C(4)	1553(2)	2396(2)	8347(3)	24(1)
C(5)	994(2)	1695(2)	10224(3)	28(1)
C(6)	1466(2)	1093(2)	10969(3)	27(1)
C(7)	1265(2)	653(2)	11970(3)	31(1)
C(8)	1782(2)	109(2)	12473(3)	34(1)
C(9)	-1050(2)	1247(2)	9747(3)	32(1)
C(10)	328(2)	1212(2)	9670(3)	25(1)
C(11)	2202(2)	1522(2)	9479(3)	25(1)
C(12)	2271(2)	2255(2)	7941(3)	26(1)
C(13)	2485(2)	2643(2)	6917(3)	30(1)
C(14)	2169(2)	977(2)	10514(3)	25(1)
C(15)	2472(2)	-17(2)	11996(3)	32(1)
C(16)	2685(2)	413(2)	10995(3)	29(1)
C(17)	3430(2)	261(2)	10466(3)	35(1)
C(18)	627(2)	2453(2)	10848(3)	28(1)



C(19)                      -195(2)                      2292(2)                      10748(3)                      25(1)

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Table S8. Bond lengths [Å] and angles [°] for 200739lt\_0m.

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O(1)-C(10)	1.214(4)
O(2)-C(19)	1.208(4)
N(1)-C(11)	1.373(4)
N(1)-C(4)	1.387(4)
N(1)-C(5)	1.455(4)
N(2)-C(10)	1.382(4)
N(2)-C(19)	1.397(4)
N(2)-C(9)	1.454(4)
N(3)-C(11)	1.310(4)
N(3)-C(12)	1.407(4)
C(1)-C(13)	1.387(5)
C(1)-C(2)	1.403(5)
C(1)-H(1)	0.9500
C(2)-C(3)	1.395(5)
C(2)-H(5)	0.9500
C(3)-C(4)	1.394(5)
C(3)-H(6)	0.9500
C(4)-C(12)	1.407(5)
C(5)-C(6)	1.529(5)
C(5)-C(18)	1.547(4)
C(5)-C(10)	1.549(4)
C(6)-C(14)	1.397(5)
C(6)-C(7)	1.404(4)
C(7)-C(8)	1.384(5)
C(7)-H(7)	0.9500
C(8)-C(15)	1.389(5)
C(8)-H(2)	0.9500
C(9)-H(3)	0.9800
C(9)-H(14)	0.9800
C(9)-H(15)	0.9800
C(11)-C(14)	1.476(4)
C(12)-C(13)	1.397(4)
C(13)-H(4)	0.9500

C(14)-C(16)	1.392(5)
C(15)-C(16)	1.404(5)
C(15)-H(11)	0.9500
C(16)-C(17)	1.511(5)
C(17)-H(8)	0.9800
C(17)-H(9)	0.9800
C(17)-H(10)	0.9800
C(18)-C(19)	1.510(5)
C(18)-H(13)	0.9900
C(18)-H(12)	0.9900

C(11)-N(1)-C(4)	106.7(3)
C(11)-N(1)-C(5)	114.8(3)
C(4)-N(1)-C(5)	138.4(3)
C(10)-N(2)-C(19)	113.0(3)
C(10)-N(2)-C(9)	123.1(3)
C(19)-N(2)-C(9)	123.9(3)
C(11)-N(3)-C(12)	103.0(3)
C(13)-C(1)-C(2)	121.8(3)
C(13)-C(1)-H(1)	119.1
C(2)-C(1)-H(1)	119.1
C(3)-C(2)-C(1)	121.0(3)
C(3)-C(2)-H(5)	119.5
C(1)-C(2)-H(5)	119.5
C(2)-C(3)-C(4)	116.6(3)
C(2)-C(3)-H(6)	121.7
C(4)-C(3)-H(6)	121.7
N(1)-C(4)-C(3)	132.8(3)
N(1)-C(4)-C(12)	104.2(3)
C(3)-C(4)-C(12)	123.0(3)
N(1)-C(5)-C(6)	99.8(3)
N(1)-C(5)-C(18)	115.9(3)
C(6)-C(5)-C(18)	116.7(3)
N(1)-C(5)-C(10)	110.7(2)
C(6)-C(5)-C(10)	110.6(3)
C(18)-C(5)-C(10)	103.5(3)
C(14)-C(6)-C(7)	120.2(3)
C(14)-C(6)-C(5)	111.6(3)

C(7)-C(6)-C(5)	128.2(3)
C(8)-C(7)-C(6)	117.7(3)
C(8)-C(7)-H(7)	121.1
C(6)-C(7)-H(7)	121.1
C(7)-C(8)-C(15)	121.6(3)
C(7)-C(8)-H(2)	119.2
C(15)-C(8)-H(2)	119.2
N(2)-C(9)-H(3)	109.5
N(2)-C(9)-H(14)	109.5
H(3)-C(9)-H(14)	109.5
N(2)-C(9)-H(15)	109.5
H(3)-C(9)-H(15)	109.5
H(14)-C(9)-H(15)	109.5
O(1)-C(10)-N(2)	125.4(3)
O(1)-C(10)-C(5)	126.0(3)
N(2)-C(10)-C(5)	108.6(3)
N(3)-C(11)-N(1)	114.7(3)
N(3)-C(11)-C(14)	138.1(3)
N(1)-C(11)-C(14)	107.2(3)
C(13)-C(12)-C(4)	119.4(3)
C(13)-C(12)-N(3)	129.2(3)
C(4)-C(12)-N(3)	111.4(3)
C(1)-C(13)-C(12)	118.2(3)
C(1)-C(13)-H(4)	120.9
C(12)-C(13)-H(4)	120.9
C(16)-C(14)-C(6)	122.5(3)
C(16)-C(14)-C(11)	130.8(3)
C(6)-C(14)-C(11)	106.6(3)
C(8)-C(15)-C(16)	121.8(3)
C(8)-C(15)-H(11)	119.1
C(16)-C(15)-H(11)	119.1
C(14)-C(16)-C(15)	116.2(3)
C(14)-C(16)-C(17)	122.3(3)
C(15)-C(16)-C(17)	121.5(3)
C(16)-C(17)-H(8)	109.5
C(16)-C(17)-H(9)	109.5
H(8)-C(17)-H(9)	109.5
C(16)-C(17)-H(10)	109.5

H(8)-C(17)-H(10)	109.5
H(9)-C(17)-H(10)	109.5
C(19)-C(18)-C(5)	105.4(3)
C(19)-C(18)-H(13)	110.7
C(5)-C(18)-H(13)	110.7
C(19)-C(18)-H(12)	110.7
C(5)-C(18)-H(12)	110.7
H(13)-C(18)-H(12)	108.8
O(2)-C(19)-N(2)	123.7(3)
O(2)-C(19)-C(18)	127.4(3)
N(2)-C(19)-C(18)	108.9(3)

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Symmetry transformations used to generate equivalent atoms:

Table S9. Anisotropic displacement parameters ( $\text{\AA}^2 \times 10^3$ ) for 200739lt\_0m. The anisotropic displacement factor exponent takes the form:  $-2\pi^2 [ h^2 a^{*2} U^{11} + \dots + 2 h k a^* b^* U^{12} ]$

	$U^{11}$	$U^{22}$	$U^{33}$	$U^{23}$	$U^{13}$	$U^{12}$
O(1)	34(1)	32(1)	34(1)	-14(1)	9(1)	-7(1)
O(2)	34(1)	36(1)	34(1)	-9(1)	10(1)	3(1)
N(1)	27(2)	27(1)	22(1)	-3(1)	6(1)	-5(1)
N(2)	24(1)	23(1)	25(1)	-1(1)	8(1)	-5(1)
N(3)	28(2)	24(1)	27(1)	-2(1)	8(1)	-4(1)
C(1)	42(2)	31(2)	28(2)	6(2)	9(2)	-7(2)
C(2)	40(2)	29(2)	31(2)	1(2)	-1(2)	-2(2)
C(3)	29(2)	31(2)	31(2)	-4(2)	6(2)	-5(2)
C(4)	31(2)	20(2)	21(2)	-4(1)	3(1)	-8(1)
C(5)	30(2)	28(2)	26(2)	-9(1)	12(1)	-8(1)
C(6)	33(2)	25(2)	24(2)	-8(1)	6(1)	-9(1)
C(7)	36(2)	35(2)	23(2)	-7(2)	11(2)	-12(2)
C(8)	44(2)	31(2)	27(2)	3(2)	4(2)	-10(2)
C(9)	30(2)	27(2)	38(2)	-4(2)	0(2)	-2(2)
C(10)	26(2)	26(2)	22(2)	-2(1)	10(1)	-6(1)
C(11)	27(2)	23(2)	24(2)	-9(1)	5(1)	-7(1)
C(12)	29(2)	22(2)	27(2)	-7(1)	7(1)	-5(1)
C(13)	30(2)	30(2)	30(2)	-1(2)	10(2)	-7(2)
C(14)	33(2)	22(2)	20(2)	-6(1)	6(1)	-10(1)

C(15)	43(2)	27(2)	25(2)	1(1)	-1(2)	-8(2)
C(16)	37(2)	25(2)	25(2)	-4(1)	3(2)	-8(2)
C(17)	40(2)	32(2)	32(2)	-3(2)	7(2)	-2(2)
C(18)	29(2)	27(2)	28(2)	-7(1)	10(2)	-4(1)
C(19)	33(2)	25(2)	18(2)	-1(1)	9(1)	-3(2)

Table S10. Hydrogen coordinates ( $\times 10^4$ ) and isotropic displacement parameters ( $\text{\AA}^2 \times 10^{-3}$ ) for 200739lt\_0m.

	x	y	z	U(eq)
H(1)	2113	3425	5637	41
H(5)	932	3664	6345	40
H(6)	552	2995	8059	36
H(7)	790	727	12291	38
H(2)	1661	-186	13159	41
H(3)	-1146	1342	8922	48
H(14)	-1423	1548	10193	48
H(15)	-1073	635	9913	48
H(4)	2966	2556	6625	36
H(11)	2808	-404	12358	38
H(8)	3370	219	9626	52
H(9)	3641	-273	10769	52
H(10)	3763	736	10661	52
H(13)	791	2474	11667	34
H(12)	757	3000	10475	34