

Electronic Supporting Information for

B(C₆F₅)₃/I₂ Co-Mediated Cascade [4 + 2]/[3 + 2] Cyclization to Spiroimidazolines

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1. General information

All starting materials and reagents were purchased from commercial sources and used as received unless otherwise noted. Reactions were monitored using thin-layer chromatography (TLC) on commercial silica gel plates. Visualization of the developed plates was performed under UV light (254 nm). NMR spectra data were obtained on Avance (III) HD 400 MHz instruments. ¹H NMR and ¹³C NMR spectra were referenced to residual protic solvent peaks or TMS signal (0 ppm). ¹⁹F NMR chemical shifts were externally referenced to CCl₃F (0 ppm). Data for ¹H NMR are recorded as follows: chemical shift (δ, ppm), multiplicity (s = singlet, d = doublet, t = triplet, m = multiplet or unresolved, br = broad singlet, coupling constant (s) in Hz, integration). Data for ¹³C and ¹⁹F NMR are reported in terms of chemical shift (δ, ppm).

2. Experimental section

2.1 General synthesis of spiroimidazoline 4a–4ai.

A solution of 2-aminoacetophenone **1** (1 mmol, 1 equiv.), isocyanate **2** (1.2 mmol, 1.2 equiv.), benzimidamide **3** (1.4 mmol, 1.4 equiv.), B(C₆F₅)₃ (0.01 mmol, 0.01 equiv.) and molecular iodine (1.2 mmol, 1.2 equiv.) were dissolved in DCE (2 mL) under an air atmosphere. The mixture was stirred at 60 °C for 16 h. Then the reaction mixture was concentrated in vacuo and purified by flash column chromatography to afford the corresponding product **4**.

2.2 Gram-scale preparation of spiroimidazoline 4u.

A solution of 2-aminoacetophenone **1a** (675 mg, 5 mmol), (2-isocyanatoethyl)benzene **2u** (882 mg, 6 mmol), *N*-phenylbenzimidamide **3a** (1.37 g, 7 mmol), B(C₆F₅)₃ (26 mg, 0.05 mmol) and molecular iodine (1.52 g, 6 mmol) were dissolved in DCE (10 mL) under an air atmosphere. The mixture was stirred at 60 °C for 16 h. Then the reaction mixture was concentrated in vacuo and purified by flash column chromatography to afford the 3'-phenethyl-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one **4u** (1.82 g, 80%) as a white solid.

2.3 Synthesis of 5u.

A solution of 2-aminoacetophenone **1a** (135 mg, 1 mmol), 2-phenylethyl isocyanate **2u** (177 mg, 1.2 mmol) and B(C₆F₅)₃ (5 mg, 0.01 mmol) were dissolved in DCE (2 mL) under an air atmosphere. The mixture was stirred at 60 °C for 10 h. Then the reaction mixture was concentrated in vacuo and purified by flash column chromatography to afford the 4-methylene-3-phenethyl-3,4-dihydroquinazolin-2(1H)-one **5u** (232 mg, 88%) as a white solid.

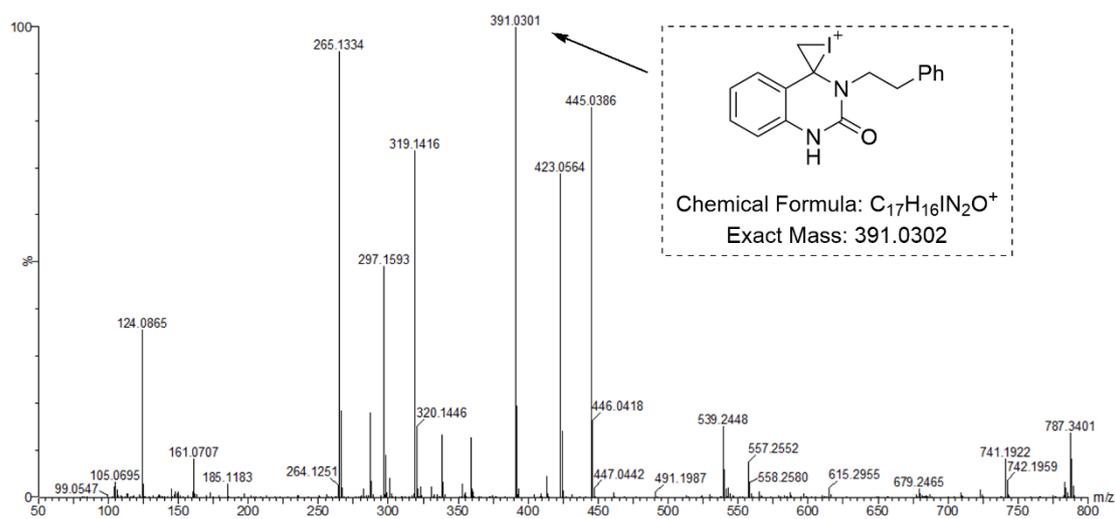
2.4 Synthesis of 6u'.

A suspension of **5u** (264 mg, 1 mmol) and NBS (214 mg, 1.2 mmol) were dissolved in DCE (2 mL) under an air atmosphere. The mixture was stirred at 60 °C for 10 h. Then the reaction mixture was

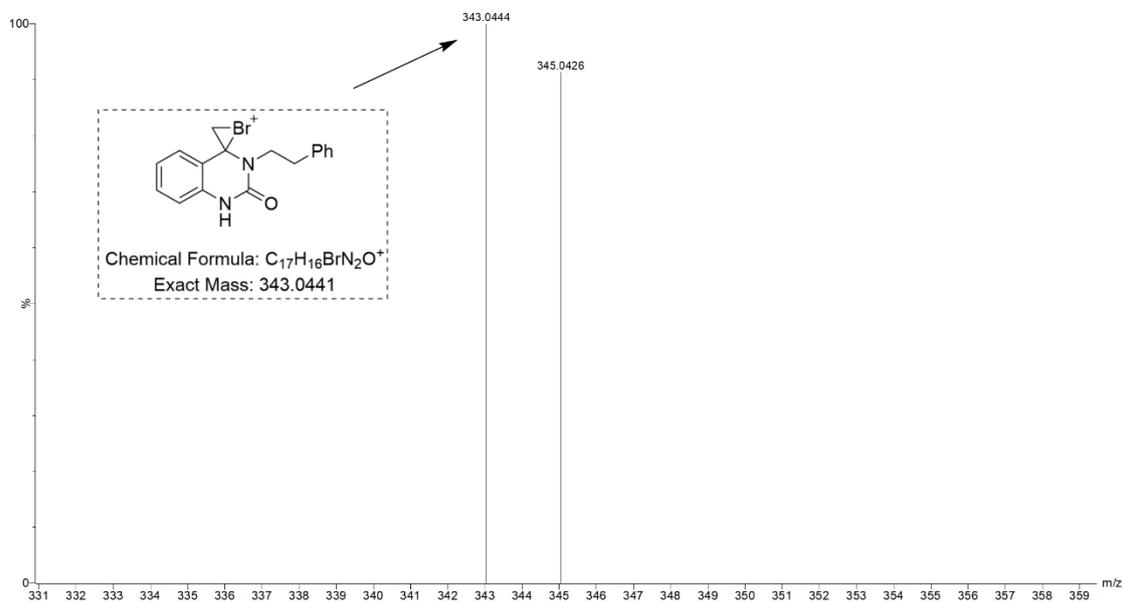
concentrated in vacuo and purified by flash column chromatography to afford the 2'-oxo-3'-phenethyl-2',3'-dihydro-1'H-spiro[bromirane-2,4'-quinazolin]-1-ium **6u'** as a yellow solid (206 mg, 60%).

3. HRMS and NMR spectra of **6u** and **6u'**

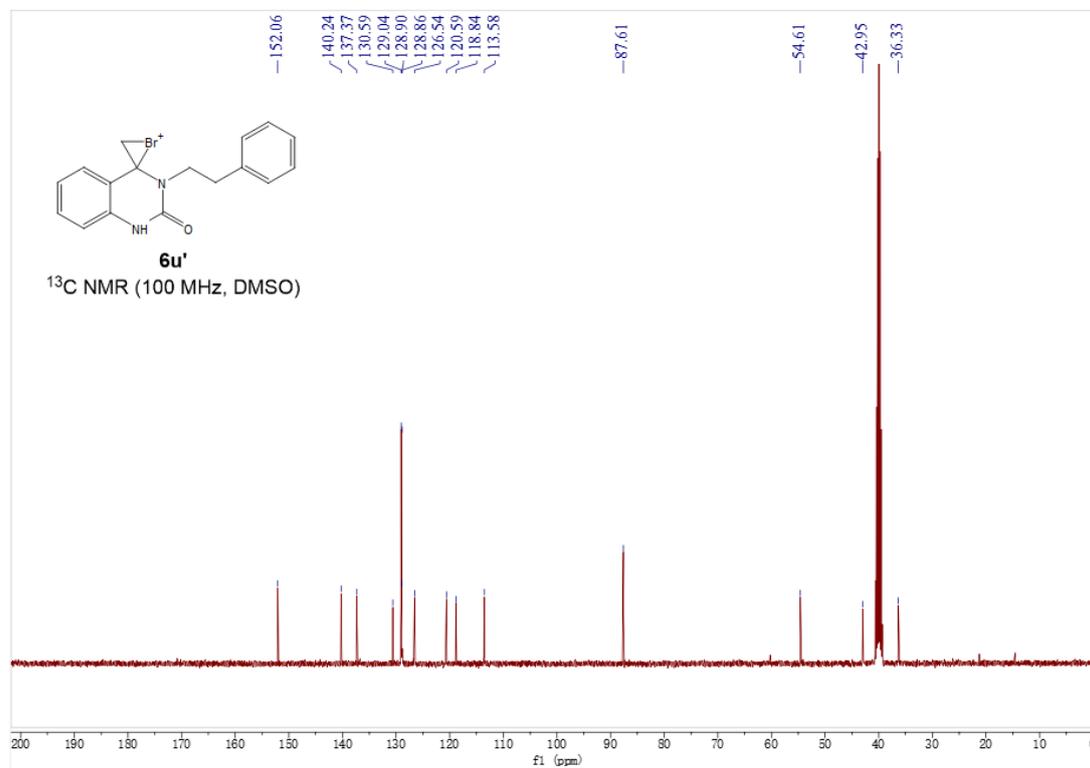
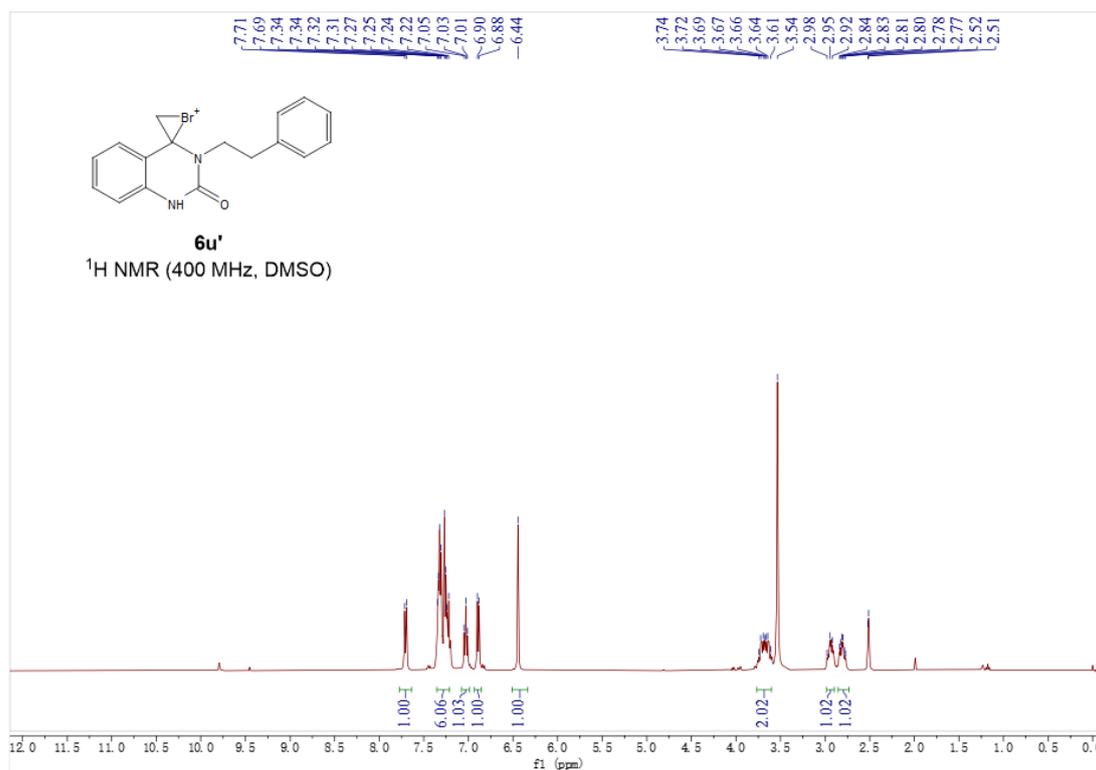
3.1 HRMS spectrum of **6u**



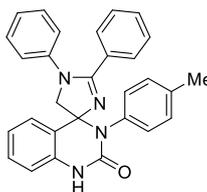
3.2 HRMS spectrum of **6u'**



3.3 NMR spectra of 6u'



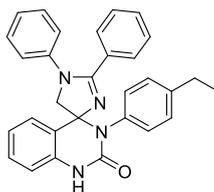
4. Characterization data of products



4a

1,2-Diphenyl-3'-(*p*-tolyl)-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4a):

Petroleum ether/ethyl acetate = 2:1; white solid, 86% yield, 382 mg; mp 216.0 – 217.4 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ 9.88 (s, 1H), 7.43 – 6.89 (m, 16H), 6.28 (d, *J* = 7.6 Hz, 2H), 4.52 (d, *J* = 12.0 Hz, 1H), 4.21 (d, *J* = 12.0 Hz, 1H), 2.23 (s, 3H); ¹³C {¹H} NMR (100 MHz, DMSO-*d*₆) δ 161.1, 151.3, 141.8, 137.4, 136.1, 135.6, 130.9, 130.4, 129.3, 129.3, 129.0, 128.9, 128.7, 127.5, 124.7, 124.4, 123.9, 121.9, 113.9, 86.5, 65.9, 21.0; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₂₉H₂₅N₄O⁺, 445.2023; found 445.2029.

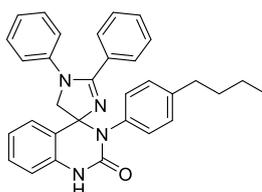


4b

3'-(4-Ethylphenyl)-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-

one (4b): Petroleum ether/ethyl acetate = 2:1; white solid, 70% yield, 321 mg; mp 199.6 – 201.1 °C;

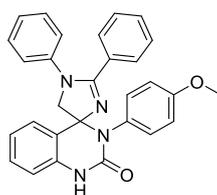
¹H NMR (400 MHz, CDCl₃) δ 7.65 (s, 2H), 7.4 – 7.35 (m, 4H), 7.28 – 7.23 (m, 4H), 7.08 – 6.99 (m, 6H), 6.75 (d, *J* = 8.0 Hz, 1H), 6.29 (d, *J* = 7.2 Hz, 2H), 4.53 (d, *J* = 11.6 Hz, 1H), 4.36 (d, *J* = 11.6 Hz, 1H), 2.62 (q, *J* = 7.6 Hz, 2H), 1.20 (t, *J* = 7.6 Hz, 3H); ¹³C {¹H} NMR (100 MHz, CDCl₃) δ 162.3, 152.3, 144.2, 141.1, 135.0, 134.9, 130.6, 129.2, 129.2, 128.6, 128.1, 126.7, 124.6, 123.6, 122.4, 114.2, 86.1, 66.9, 28.5, 15.7; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₃₀H₂₇N₄O⁺, 459.2179; found 459.2183.



4c

3'-(4-Butylphenyl)-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-

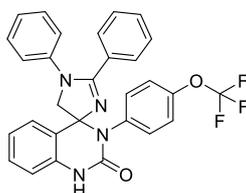
one (4c): Petroleum ether/ethyl acetate = 2:1; white solid, 68% yield, 330 mg; mp 183.3 – 185.0 °C; ^1H NMR (400 MHz, CDCl_3) δ 8.44 (s, 1H), 7.60 – 7.40 (m, 1H), 7.37 – 7.28 (m, 4H), 7.20 (dt, J = 15.6, 7.6 Hz, 4H), 7.13 – 6.91 (m, 6H), 6.71 (d, J = 8.0 Hz, 1H), 6.28 (d, J = 7.6 Hz, 2H), 4.46 (d, J = 11.6 Hz, 1H), 4.34 (d, J = 11.6 Hz, 1H), 2.64 – 2.49 (m, 2H), 1.54 (dt, J = 15.2, 7.6 Hz, 2H), 1.30 (q, J = 7.6 Hz, 2H), 0.89 (t, J = 7.6 Hz, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ 162.4, 152.1, 142.8, 141.1, 134.9, 130.5, 129.7, 129.2, 129.2, 128.6, 128.1, 126.7, 124.6, 124.3, 123.6, 122.5, 114.0, 86.1, 66.9, 35.3, 33.6, 22.3, 14.0; HRMS (ESI-TOF) m/z $[\text{M} + \text{H}]^+$ calcd for $\text{C}_{32}\text{H}_{31}\text{N}_4\text{O}^+$, 487.2492; found 487.2499.



4d

3'-(4-Methoxyphenyl)-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-

2'(3'H)-one (4d): Petroleum ether/ethyl acetate = 2:1; white solid, 88% yield, 405 mg; mp 187.4 – 189.3 °C; ^1H NMR (400 MHz, CDCl_3) δ 8.15 (s, 1H), 7.56 (s, 1H), 7.43 (d, J = 7.2 Hz, 2H), 7.34 (dd, J = 13.2, 7.6 Hz, 2H), 7.29 – 7.20 (m, 4H), 7.09 – 7.05 (m, 3H), 7.03 – 6.96 (m, 2H), 6.73 (d, J = 8.0 Hz, 2H), 6.34 (dd, J = 7.2, 1.6 Hz, 2H), 4.51 (d, J = 11.6 Hz, 1H), 4.37 (d, J = 11.6 Hz, 1H), 3.77 (s, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ 162.4, 159.2, 152.3, 141.3, 134.8, 130.6, 130.0, 129.6, 129.2, 129.2, 128.6, 128.2, 126.8, 124.5, 124.1, 123.5, 122.5, 114.1, 86.2, 67.0, 55.5; HRMS (ESI-TOF) m/z $[\text{M} + \text{H}]^+$ calcd for $\text{C}_{29}\text{H}_{25}\text{N}_4\text{O}_2^+$, 461.1972; found 461.1978.

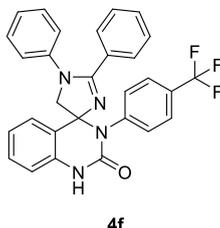


4e

1,2-Diphenyl-3'-(4-(trifluoromethoxy)phenyl)-1,5-dihydro-1'H-spiro[imidazole-4,4'-

quinazolin]-2'(3'H)-one (4e): Petroleum ether/ethyl acetate = 2:1; white solid, 63% yield, 324 mg; mp 212.8 – 213.5 °C; ^1H NMR (400 MHz, $\text{DMSO}-d_6$) δ 10.01 (s, 1H), 7.50 (s, 2H), 7.42 – 7.09 (m, 9H), 7.08 – 6.99 (m, 3H), 6.98 – 6.77 (m, 2H), 6.32 (d, J = 7.6 Hz, 2H), 4.51 (d, J = 12.0 Hz, 1H), 4.30 (d, J = 12.0 Hz, 1H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, $\text{DMSO}-d_6$) δ 161.5, 151.1, 147.9, 141.3, 137.5, 135.9, 130.9, 130.2, 129.4, 129.1(d, J = 25 Hz), 128.6, 127.6, 124.8, 124.3, 123.6, 122.1, 121.7,

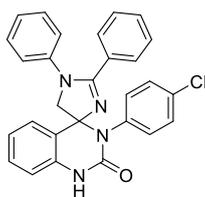
121.3, 114.0, 86.2, 66.0; $^{19}\text{F}\{^1\text{H}\}$ NMR (376 MHz, DMSO- d_6) δ -57.0; HRMS (ESI-TOF) m/z [$\text{M} + \text{H}$] $^+$ calcd for $\text{C}_{29}\text{H}_{22}\text{F}_3\text{N}_4\text{O}_2$, 515.1689; found 515.1685.



1,2-Diphenyl-3'-(4-(trifluoromethyl)phenyl)-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4f): Petroleum ether/ethyl acetate = 2:1; yellow solid, 74% yield, 369 mg; mp 199.2 – 200.4 °C; ^1H NMR (400 MHz, CDCl_3) δ 8.66 (t, $J = 27.2$ Hz, 1H), 7.61 (s, 4H), 7.39 (t, $J = 6.8$ Hz, 3H), 7.33 (d, $J = 7.6$ Hz, 1H), 7.27 (t, $J = 6.8$ Hz, 2H), 7.21 (d, $J = 7.6$ Hz, 1H), 7.04 (dt, $J = 23.2, 7.2$ Hz, 4H), 6.72 (d, $J = 8.0$ Hz, 1H), 6.24 (d, $J = 7.6$ Hz, 2H), 4.52 (d, $J = 11.6$ Hz, 1H), 4.33 (d, $J = 11.6$ Hz, 1H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ 162.8, 151.9, 140.9 (d, $J = 5$ Hz), 134.6, 130.9, 130.2 (d, $J = 32$ Hz), 129.4, 129.2, 129.0 (d, $J = 25$ Hz), 128.3, 126.7, 125.6, 124.8, 123.9, 123.1, 122.8, 114.3, 86.3, 67.0; $^{19}\text{F}\{^1\text{H}\}$ NMR (376 MHz, CDCl_3) δ -62.5; HRMS (ESI-TOF) m/z [$\text{M} + \text{H}$] $^+$ calcd for $\text{C}_{29}\text{H}_{22}\text{F}_3\text{N}_4\text{O}^+$, 499.1740; found 499.1747.

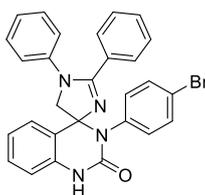


3'-(4-Fluorophenyl)-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4g): Petroleum ether/ethyl acetate = 2:1; yellow solid, 84% yield, 376 mg; mp 188.8 – 189.6 °C; ^1H NMR (400 MHz, CDCl_3) δ 9.11 (s, 1H), 7.64 (s, 1H), 7.43 – 7.34 (m, 3H), 7.34 – 7.25 (m, 3H), 7.23 – 7.06 (m, 5H), 7.02 (q, $J = 7.2, 6.4$ Hz, 2H), 6.92 (s, 1H), 6.69 (d, $J = 8.0$ Hz, 1H), 6.34 (d, $J = 8.0$ Hz, 2H), 4.50 (dd, $J = 11.6, 3.2$ Hz, 1H), 4.35 (d, $J = 11.6$ Hz, 1H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ 163.5, 162.6, 161.0, 152.3, 141.0, 134.7, 133.3 (d, $J = 3$ Hz), 130.8, 129.4, 129.3, 129.0 (d, $J = 31$ Hz), 128.3, 126.6, 124.8, 124.0, 123.3, 122.7, 115.5 (d, $J = 22$ Hz), 114.2, 86.1, 67.0; $^{19}\text{F}\{^1\text{H}\}$ NMR (376 MHz, CDCl_3) δ -113.8; HRMS (ESI-TOF) m/z [$\text{M} + \text{H}$] $^+$ calcd for $\text{C}_{28}\text{H}_{22}\text{FN}_4\text{O}^+$, 449.1772; found 449.1777.



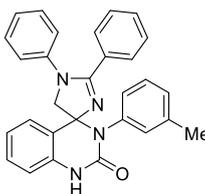
4h

3'-(4-Chlorophenyl)-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4h): Petroleum ether/ethyl acetate = 2:1; yellow solid, 76% yield, 353 mg; mp 198.8 – 199.2 °C; ^1H NMR (400 MHz, CDCl_3) δ 8.44 (s, 1H), 7.40 (dd, J = 13.2, 7.2 Hz, 4H), 7.34 – 7.21 (m, 6H), 7.17 – 6.97 (m, 5H), 6.71 (d, J = 8.0 Hz, 1H), 6.32 (d, J = 7.6 Hz, 2H), 4.51 (d, J = 11.6 Hz, 1H), 4.32 (d, J = 11.6 Hz, 1H); ^{13}C { ^1H } NMR (100 MHz, CDCl_3) δ 162.7, 151.9, 141.0, 136.0, 134.6, 134.1, 130.8, 129.3, 129.2, 128.8, 128.3, 126.7, 124.8, 124.0, 123.3, 122.7, 114.1, 86.2, 67.0; HRMS (ESI-TOF) m/z $[\text{M} + \text{H}]^+$ calcd for $\text{C}_{28}\text{H}_{22}\text{ClN}_4\text{O}^+$, 465.1477; found 465.1474.



4i

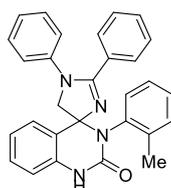
3'-(4-Bromophenyl)-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4i): Petroleum ether/ethyl acetate = 2:1; yellow solid, 77% yield, 391 mg; mp 189.3 – 190.7 °C; ^1H NMR (400 MHz, CDCl_3) δ 8.78 (s, 1H), 7.51 – 7.36 (m, 5H), 7.34 – 7.17 (m, 5H), 7.17 – 6.91 (m, 5H), 6.70 (d, J = 8.0 Hz, 1H), 6.32 (d, J = 7.6 Hz, 2H), 4.51 (d, J = 11.6 Hz, 1H), 4.31 (d, J = 11.6 Hz, 1H); ^{13}C { ^1H } NMR (100 MHz, CDCl_3) δ 162.7, 151.9, 141.0, 136.4, 134.6, 132.6, 131.8, 130.9, 130.5, 129.4, 129.2, 128.8, 128.5, 128.3, 127.4, 126.8, 124.8, 123.8, 123.3, 122.8, 122.3, 114.2, 86.1, 67.0; HRMS (ESI-TOF) m/z $[\text{M} + \text{H}]^+$ calcd for $\text{C}_{28}\text{H}_{22}\text{BrN}_4\text{O}^+$, 509.0972; found 509.0970.



4j

1,2-Diphenyl-3'-(m-tolyl)-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4j): Petroleum ether/ethyl acetate = 2:1; white solid, 77% yield, 342 mg, $d.r.$ = 4:1; mp 192.3 – 193.7 °C;

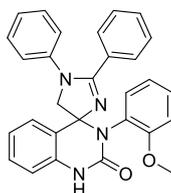
^1H NMR (400 MHz, DMSO- d_6) δ 9.97 and 9.79 (s, 1H), 7.47 – 7.29 (m, 3H), 7.29 – 7.15 (m, 5H), 7.14 – 7.08 (m, 3H), 7.07 – 7.00 (m, 1H), 7.00 – 6.91 (m, 5H), 6.51 – 6.45 (m, 2H), 4.62 (d, J = 11.6 Hz, 1H), 4.29 and 3.61 (d, J = 11.6 Hz, 1H), 2.27 and 2.05 (s, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO- d_6) δ 162.6, 160.3, 150.9, 142.2, 140.8, 139.5, 138.4, 138.1, 136.1, 135.9, 131.9, 130.8, 130.7, 130.6, 130.3, 130.2, 129.4, 129.2, 129.2, 129.0, 128.9, 128.8, 128.5, 128.2, 128.0, 126.7, 126.6, 126.2, 125.7, 125.3, 124.6, 124.4, 123.3, 122.2, 122.0, 114.1, 113.8, 87.0, 86.1, 67.9, 63.6, 19.3, 18.5; HRMS (ESI-TOF) m/z $[\text{M} + \text{H}]^+$ calcd for $\text{C}_{29}\text{H}_{25}\text{N}_4\text{O}^+$, 445.2023; found 445.2020.



4k

1,2-Diphenyl-3'-(o-tolyl)-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4k):

Petroleum ether/ethyl acetate = 2:1; yellow solid, 50% yield, 222 mg, $d.r.$ = 1:1; mp 198.8 – 199.2 °C; ^1H NMR (400 MHz, DMSO- d_6) δ 10.00 and 9.82 (s, 1H), 7.47 – 7.23 (m, 8H), 7.13 – 6.87 (m, 8H), 6.48 (dd, J = 19.6, 7.2 Hz, 2H), 4.66 (dd, J = 26.0, 11.6 Hz, 1H), 4.30 and 3.61 (d, J = 11.6 Hz, 1H), 2.28 and 2.05 (s, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO- d_6) δ 162.6, 160.3, 150.9, 142.2, 140.8, 139.5, 138.4, 138.1, 137.8, 136.1, 135.8, 131.9, 130.9, 130.7, 130.5, 130.5, 130.3, 130.2, 129.4, 129.2, 129.0, 128.9, 128.8, 128.6, 128.2, 128.0, 126.7, 126.6, 126.2, 125.7, 125.3, 124.6, 124.5, 124.4, 123.3, 122.2, 122.0, 114.1, 113.8, 86.9, 86.1, 67.9, 63.6, 19.3, 18.5; HRMS (ESI-TOF) m/z $[\text{M} + \text{H}]^+$ calcd for $\text{C}_{29}\text{H}_{25}\text{N}_4\text{O}^+$, 445.2023; found 445.2021.

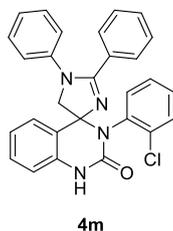


4l

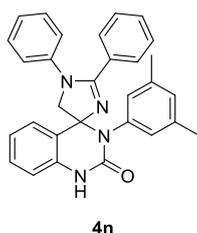
3'-(2-Methoxyphenyl)-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-

2'(3'H)-one (4l): Petroleum ether/ethyl acetate = 2:1; yellow solid, 55% yield, 253 mg, $d.r.$ = 9:1; mp 206.2 – 207.8 °C; ^1H NMR (400 MHz, CDCl_3) δ 8.58 (d, J = 49.2 Hz, 1H), 7.51 (dd, J = 7.6, 1.6 Hz, 1H), 7.44 – 7.31 (m, 2H), 7.24 (dd, J = 19.2, 7.6 Hz, 2H), 7.19 – 7.11 (m, 3H), 7.06 (s, 1H), 6.97 – 6.79 (m, 5H), 6.64 – 6.55 (m, 2H), 6.12 – 6.03 (m, 2H), 4.50 (d, J = 11.2 Hz, 1H), 4.23 (d, J = 11.2 Hz, 1H), 3.22 (s, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ 162.5, 156.4, 151.5, 151.5, 141.7,

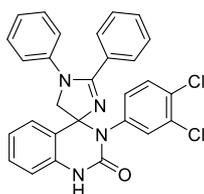
135.2, 133.8, 130.6, 130.4, 130.3, 129.8, 129.4, 129.4, 129.0, 129.0, 128.8, 128.7, 128.5, 128.1, 128.0, 126.8, 125.8, 124.3, 123.8, 122.8, 122.4, 122.2, 120.0, 114.0, 111.9, 86.5, 68.8, 67.2, 56.2, 54.9; HRMS (ESI-TOF) m/z $[M + H]^+$ calcd for $C_{29}H_{25}N_4O_2^+$, 461.1972; found 461.1968.



3'-(2-Chlorophenyl)-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4m): Petroleum ether/ethyl acetate = 2:1; yellow solid, 53% yield, 246 mg, *d.r.* = 3:2; mp 220.5 – 221.1 °C; 1H NMR (400 MHz, DMSO- d_6) δ 9.90 (d, J = 2.4 Hz, 1H), 7.56 (dd, J = 8.0, 1.6 Hz, 1H), 7.46 – 7.25 (m, 7H), 7.24 – 7.18 (m, 1H), 7.13 (td, J = 7.2, 5.2 Hz, 2H), 7.07 – 6.91 (m, 5H), 6.52 (d, J = 7.6 Hz, 1H), 6.43 (d, J = 7.6 Hz, 1H), 4.69 (dd, J = 11.6, 9.2 Hz, 1H), 4.28 (d, J = 11.6 Hz, 1H); $^{13}C\{^1H\}$ NMR (100 MHz, DMSO- d_6) δ 162.9, 160.9, 150.7, 150.2, 141.9, 140.7, 137.1, 136.0, 136.0, 135.7, 134.8, 134.4, 132.1, 131.0, 130.8, 130.4, 130.2, 130.1, 130.0, 129.6, 129.3, 129.2, 129.1, 129.0, 128.8, 128.6, 127.8, 127.4, 127.3, 126.7, 125.8, 125.2, 124.4, 124.1, 123.2, 122.3, 122.1, 114.2, 114.0, 87.1, 86.3, 68.1, 64.4; HRMS (ESI-TOF) m/z $[M + H]^+$ calcd for $C_{28}H_{22}ClN_4O^+$, 465.1477; found 465.1470.



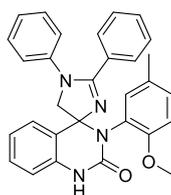
3'-(3,5-Dimethylphenyl)-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4n): Petroleum ether/ethyl acetate = 2:1; white solid, 79% yield, 362 mg; mp 211.3 – 212.4 °C; 1H NMR (400 MHz, $CDCl_3$) δ 7.42 – 7.35 (m, 4H), 7.27 (d, J = 9.6 Hz, 4H), 7.10 – 7.04 (m, 3H), 7.01 (d, J = 7.2 Hz, 1H), 6.92 (s, 1H), 6.75 (d, J = 8.0 Hz, 2H), 6.26 (d, J = 7.6 Hz, 2H), 4.50 (d, J = 11.6 Hz, 1H), 4.39 (d, J = 11.6 Hz, 1H), 2.36 (s, 3H), 2.04 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$) δ 162.5, 151.9, 141.1, 137.0, 134.9, 130.6, 129.9, 129.6, 129.3, 129.2, 128.6, 128.1, 127.8, 126.8, 124.3, 124.3, 123.3, 122.5, 113.9, 113.8, 86.0, 67.4, 21.1; HRMS (ESI-TOF) m/z $[M + H]^+$ calcd for $C_{30}H_{27}N_4O^+$, 459.2179; found 459.2170.



4o

3'-(3,4-Dichlorophenyl)-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-

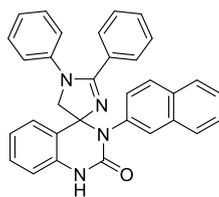
2'(3'H)-one (4o): Petroleum ether/ethyl acetate = 2:1; yellow solid, 60% yield, 299 mg, *d.r.* > 20:1; mp 203.2 – 204.3 °C; ¹H NMR (400 MHz, CDCl₃) δ 8.34 (s, 1H), 7.40 – 7.33 (m, 6H), 7.29 – 7.20 (m, 4H), 7.14 – 6.98 (m, 6H), 6.74 (d, *J* = 8.0 Hz, 1H), 6.28 (d, *J* = 7.6 Hz, 2H), 4.49 (d, *J* = 11.6 Hz, 1H), 4.39 (d, *J* = 11.6 Hz, 1H); ¹³C{¹H} NMR (100 MHz, CDCl₃) δ 162.5, 152.1, 141.1, 137.3, 134.9, 130.6, 129.6, 129.2, 129.2, 128.9, 128.6, 128.3, 128.1, 126.7, 124.5, 124.3, 123.4, 122.5, 114.0, 86.1, 67.2; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₂₈H₂₁Cl₂N₄O⁺, 499.1087; found 499.1080.



4p

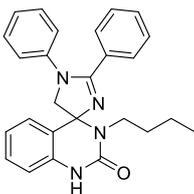
3'-(2-Methoxy-5-methylphenyl)-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-

quinazolin]-2'(3'H)-one (4p): Petroleum ether/ethyl acetate = 2:1; yellow solid, 82% yield, 389 mg, *d.r.* = 9:1; mp 188.2 – 189.4 °C; ¹H NMR (400 MHz, CDCl₃) δ 8.87 and 8.33 (s, 1H), 7.53 – 7.43 (m, 3H), 7.40 – 7.12 (m, 6H), 7.03 – 6.94 (m, 5H), 6.73 (d, *J* = 8.0 Hz, 1H), 6.57 and 6.45 (d, *J* = 8.4 Hz, 1H), 6.17 (d, *J* = 7.6 Hz, 2H), 4.62 (d, *J* = 11.6 Hz, 1H), 4.50 and 4.32 (d, *J* = 11.6 Hz, 1H), 3.84 and 3.27 (s, 3H), 2.34 and 1.99 (s, 3H); ¹³C{¹H} NMR (100 MHz, CDCl₃) δ 162.5, 154.1, 151.6, 141.6, 135.3, 134.4, 130.7, 129.7, 129.4, 129.1, 128.9, 128.7, 128.4, 128.1, 126.9, 125.3, 123.7, 122.8, 122.2, 114.1, 111.7, 86.4, 68.8, 67.2, 56.4, 54.9, 20.5; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₃₀H₂₇N₄O₂⁺, 475.2129; found 475.2125.



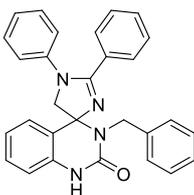
4r

3'-(Naphthalen-2-yl)-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4r): Petroleum ether/ethyl acetate = 2:1; yellow solid, 79% yield, 379 mg, *d.r.* > 20:1; mp 202.7 – 203.6 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ 9.99 (s, 1H), 8.08 – 7.59 (m, 4H), 7.56 – 7.17 (m, 10H), 7.03 (t, *J* = 7.6 Hz, 1H), 6.95 (d, *J* = 8.0 Hz, 1H), 6.75 (dd, *J* = 18.4, 7.6 Hz, 3H), 6.02 (d, *J* = 7.6 Hz, 2H), 4.57 (d, *J* = 12.0 Hz, 2H); ¹³C {¹H} NMR (100 MHz, DMSO-*d*₆) δ 161.3, 151.4, 141.5, 136.1, 132.7, 130.9, 130.2, 129.4, 129.3, 128.6, 128.1, 127.7, 127.5, 126.6, 124.5, 124.4, 123.6, 122.0, 114.0, 86.8; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₃₂H₂₅N₄O⁺, 481.2023; found 481.2020.



4s

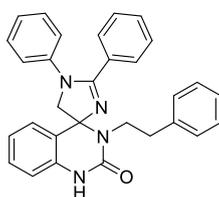
3'-Butyl-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4s): Petroleum ether/ethyl acetate = 2:1; white solid, 62% yield, 254 mg; mp 186.1 – 187.3 °C; ¹H NMR (400 MHz, CDCl₃) δ 8.58 (s, 1H), 7.69 – 7.60 (m, 2H), 7.44 (t, *J* = 7.6 Hz, 1H), 7.34 (t, *J* = 7.6 Hz, 2H), 7.24 – 7.12 (m, 4H), 7.07 (t, *J* = 7.6 Hz, 1H), 6.97 (td, *J* = 7.6, 1.2 Hz, 1H), 6.86 (d, *J* = 7.6 Hz, 2H), 6.78 (d, *J* = 7.6 Hz, 1H), 4.30 – 4.15 (m, 2H), 3.72 (ddd, *J* = 13.6, 9.6, 6.0 Hz, 1H), 3.13 (ddd, *J* = 13.6, 10.0, 5.6 Hz, 1H), 1.93 – 1.78 (m, 2H), 1.36 (qd, *J* = 7.6, 2.4 Hz, 2H), 0.91 (t, *J* = 7.6 Hz, 3H); ¹³C {¹H} NMR (100 MHz, CDCl₃) δ 162.7, 141.7, 134.5, 130.8, 129.9, 129.2, 129.2, 129.1, 128.4, 126.0, 124.8, 124.4, 123.6, 122.3, 113.6, 85.8, 68.1, 43.0, 32.2, 20.6, 13.9; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₂₆H₂₇N₄O⁺, 411.2179; found 411.2170.



4t

3'-Benzyl-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4t):

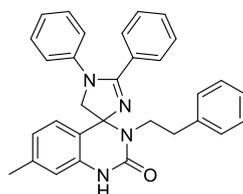
Petroleum ether/ethyl acetate = 2:1; yellow solid, 81% yield, 360 mg; mp 198.8 – 199.2 °C; ¹H NMR (400 MHz, CDCl₃) δ 9.03 (s, 1H), 7.55 – 7.47 (m, 4H), 7.44 (t, *J* = 7.2 Hz, 1H), 7.30 (ddd, *J* = 15.6, 10.8, 7.2 Hz, 5H), 7.23 – 7.13 (m, 4H), 7.09 – 6.98 (m, 2H), 6.77 (d, *J* = 8.0 Hz, 1H), 6.67 (d, *J* = 7.2 Hz, 2H), 4.98 (d, *J* = 15.6 Hz, 1H), 4.76 (d, *J* = 15.6 Hz, 1H), 4.17 (d, *J* = 11.2 Hz, 1H), 4.07 (d, *J* = 11.2 Hz, 1H); ¹³C {¹H} NMR (100 MHz, CDCl₃) δ 162.8, 153.5, 141.2, 139.8, 134.4, 130.8, 129.7, 129.3, 129.1, 129.0, 128.4, 128.3, 128.0, 126.9, 125.6, 124.8, 124.7, 123.6, 122.5, 114.0, 85.8, 67.3, 45.8; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₂₉H₂₅N₄O⁺, 445.2023; found 445.2020.



4u

3'-Phenethyl-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4u):

Petroleum ether/ethyl acetate = 2:1; yellow solid, 83% yield, 380 mg; mp 198.8 – 199.3 °C; ¹H NMR (400 MHz, CDCl₃) δ 7.98 (s, 1H), 7.72 (d, *J* = 7.2 Hz, 2H), 7.50 (t, *J* = 7.6 Hz, 1H), 7.40 (t, *J* = 7.6 Hz, 2H), 7.26 – 7.15 (m, 9H), 7.11 (t, *J* = 7.6 Hz, 1H), 7.04 (t, *J* = 7.6 Hz, 1H), 6.87 (d, *J* = 7.6 Hz, 2H), 6.80 (d, *J* = 8.0 Hz, 1H), 4.28 – 4.15 (m, 2H), 3.97 – 3.89 (m, 1H), 3.41 – 3.30 (m, 1H), 3.22 – 3.14 (m, 2H); ¹³C {¹H} NMR (100 MHz, CDCl₃) δ 162.8, 152.4, 141.7, 139.9, 134.4, 131.0, 129.9, 129.3, 129.2, 129.2, 129.0, 128.5, 126.2, 124.9, 124.1, 123.7, 122.5, 113.8, 86.2, 68.1, 45.3, 36.6; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₃₀H₂₇N₄O⁺, 459.2179; found 459.2170.

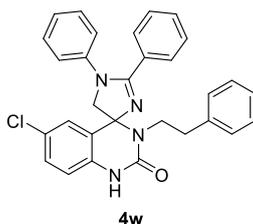


4v

7'-Methyl-3'-phenethyl-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-

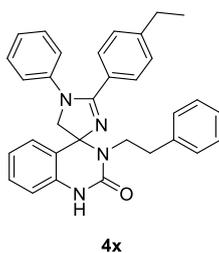
2'(3'H)-one (4v): Petroleum ether/ethyl acetate = 2:1; white solid, 78% yield, 368 mg, *d.r.* = 4:1; mp 198.8 – 199.3 °C; ¹H NMR (400 MHz, CDCl₃) δ 9.66 and 9.18 (s, 1H), 7.73 (d, *J* = 7.2 Hz, 2H), 7.48 (t, *J* = 7.6 Hz, 1H), 7.40 (d, *J* = 7.6 Hz, 2H), 7.25 – 7.20 (m, 7H), 7.09 (t, *J* = 7.2 Hz, 2H), 6.89 – 6.83 (m, 3H), 7.55 and 6.73 (s, 1H), 4.27 – 4.10 (m, 2H), 3.96 (ddd, *J* = 13.6, 10.8, 5.6 Hz, 1H),

3.38 (ddd, $J = 13.2, 9.2, 5.6$ Hz, 1H), 3.21 (dq, $J = 16.4, 5.6$ Hz, 2H), 2.39 and 2.33 (s, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ 163.1, 162.5, 152.7, 152.6, 142.3, 141.8, 141.5, 140.1, 139.9, 139.3, 136.1, 135.1, 134.4, 131.1, 130.9, 130.0, 129.7, 129.4, 129.3, 129.2, 129.2, 129.0, 128.9, 128.5, 128.5, 126.2, 126.2, 126.0, 125.0, 124.8, 124.0, 123.7, 123.6, 123.3, 121.3, 115.1, 114.2, 91.3, 86.2, 85.3, 68.1, 45.3, 45.1, 36.7, 36.6, 27.9, 21.2; HRMS (ESI-TOF) m/z $[\text{M} + \text{H}]^+$ calcd for $\text{C}_{31}\text{H}_{29}\text{N}_4\text{O}^+$, 473.2336; found 473.2344.



6'-Chloro-3'-phenethyl-1,2-diphenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-

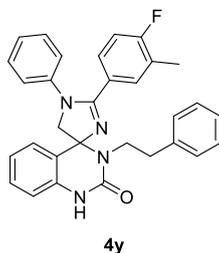
2'(3'H)-one (4w): Petroleum ether/ethyl acetate = 2:1; white solid, 71% yield, 349 mg, *d.r.* > 20:1; mp 200.8 – 201.3 °C; ^1H NMR (400 MHz, CDCl_3) δ 8.15 (s, 1H), 7.69 (d, $J = 7.6$ Hz, 2H), 7.49 (t, $J = 7.6$ Hz, 1H), 7.39 (t, $J = 7.6$ Hz, 2H), 7.24 – 7.17 (m, 6H), 7.16 – 7.07 (m, 4H), 6.85 (d, $J = 8.0$ Hz, 2H), 6.74 (d, $J = 8.4$ Hz, 1H), 4.24 – 4.11 (m, 2H), 3.90 (ddd, $J = 13.2, 10.8, 6.0$ Hz, 1H), 3.30 (ddd, $J = 13.2, 10.4, 5.6$ Hz, 1H), 3.14 (ddt, $J = 16.0, 10.4, 5.6$ Hz, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ 163.4, 151.8, 141.3, 139.6, 133.1, 131.2, 129.3, 129.3, 128.9, 128.6, 128.5, 127.3, 126.3, 125.8, 125.2, 123.8, 115.0, 85.7, 68.1, 45.2, 36.5; HRMS (ESI-TOF) m/z $[\text{M} + \text{H}]^+$ calcd for $\text{C}_{30}\text{H}_{26}\text{ClN}_4\text{O}^+$, 493.1790; found 493.1794.



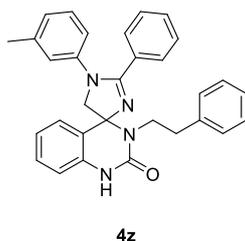
2-(4-Ethylphenyl)-3'-phenethyl-1-phenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-

2'(3'H)-one (4x): Petroleum ether/ethyl acetate = 2:1; white solid, 65% yield, 316 mg; mp 216.0 – 217.4 °C; ^1H NMR (400 MHz, CDCl_3) δ 7.63 (d, $J = 8.0$ Hz, 2H), 7.26 – 7.14 (m, 12H), 7.10 (t, $J = 7.6$ Hz, 1H), 7.03 (t, $J = 7.2$ Hz, 1H), 6.88 (d, $J = 7.6$ Hz, 2H), 6.74 (d, $J = 8.0$ Hz, 1H), 4.20 (q, $J = 11.2$ Hz, 2H), 3.99 – 3.81 (m, 1H), 3.40 – 3.25 (m, 1H), 3.17 (dt, $J = 10.8, 5.6$ Hz, 2H), 2.71 (q, $J = 7.6$ Hz, 2H), 1.27 (t, $J = 7.6$ Hz, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) δ 162.8, 152.3, 147.5, 141.9, 140.0, 134.4, 129.3, 129.2, 129.2, 129.0, 128.5, 128.0, 127.0, 126.2, 126.1, 124.8, 124.2, 123.7,

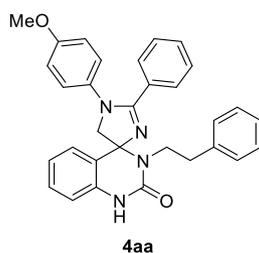
122.5, 113.7, 86.1, 68.3, 45.3, 36.6, 28.8, 15.2; HRMS (ESI-TOF) m/z $[M + H]^+$ calcd for $C_{32}H_{31}N_4O^+$, 487.2492; found 487.2490.



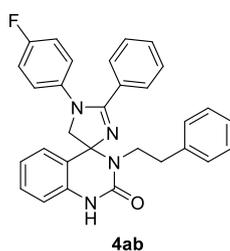
2-(4-Fluoro-3-methylphenyl)-3'-phenethyl-1-phenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4y): Petroleum ether/ethyl acetate = 2:1; yellow solid, 54% yield, 265 mg; mp 171.1 – 172.3 °C; 1H NMR (400 MHz, $CDCl_3$) δ 7.66 (s, 1H), 7.36 (d, $J = 10.4$ Hz, 2H), 7.27 – 7.15 (m, 11H), 7.04 (t, $J = 7.6$ Hz, 1H), 6.88 (d, $J = 7.6$ Hz, 2H), 6.77 (d, $J = 8.4$ Hz, 1H), 4.19 (q, $J = 11.6$ Hz, 2H), 3.93 – 3.87 (m, 1H), 3.33 – 3.28 (m, 1H), 3.24 – 3.08 (m, 2H), 2.34 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$) δ 162.1, 161.7 (d, $J = 3$ Hz), 159.7, 152.4, 141.6, 139.9, 134.4, 131.6 (d, $J = 6$ Hz), 129.3, 129.3, 129.2, 129.1, 128.9, 128.5, 128.4, 128.2, 126.2, 126.1, 125.2, 124.8 (d, $J = 4$ Hz), 124.0, 123.8, 122.5, 116.0 (d, $J = 25$ Hz), 113.8, 86.3, 68.3, 45.3, 36.5, 14.7 (d, $J = 3$ Hz); $^{19}F\{^1H\}$ NMR (376 MHz, $CDCl_3$) δ -116.2; HRMS (ESI-TOF) m/z $[M + H]^+$ calcd for $C_{31}H_{28}FN_4O^+$, 491.2242; found 491.2240.



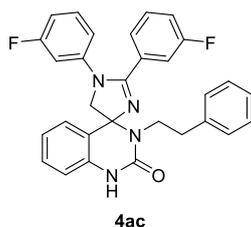
3'-Phenethyl-2-phenyl-1-(m-tolyl)-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4z): Petroleum ether/ethyl acetate = 2:1; yellow solid, 80% yield, 378 mg; mp 133.2 – 134.5 °C; 1H NMR (400 MHz, $CDCl_3$) δ 8.80 (s, 1H), 7.70 (d, $J = 7.6$ Hz, 2H), 7.45 (t, $J = 7.6$ Hz, 1H), 7.36 (t, $J = 7.6$ Hz, 2H), 7.19 (dd, $J = 16.0, 6.4$ Hz, 7H), 7.06 – 6.97 (m, 2H), 6.85 (dd, $J = 16.4, 7.6$ Hz, 2H), 6.68 (s, 1H), 6.61 (d, $J = 8.0$ Hz, 1H), 4.19 (q, $J = 11.2$ Hz, 2H), 3.98 – 3.85 (m, 1H), 3.36 – 3.26 (m, 1H), 3.18 (ddt, $J = 20.8, 11.2, 5.2$ Hz, 2H), 2.21 (s, 3H); $^{13}C\{^1H\}$ NMR (100 MHz, $CDCl_3$) δ 162.8, 152.4, 141.6, 140.0, 139.2, 134.5, 130.9, 129.9, 129.3, 129.2, 129.0, 128.9, 128.5, 128.4, 126.2, 126.2, 125.7, 124.3, 124.2, 122.4, 120.8, 113.8, 86.2, 68.2, 45.3, 36.6, 21.4; HRMS (ESI-TOF) m/z $[M + H]^+$ calcd for $C_{31}H_{29}N_4O^+$, 473.2336; found 473.2340.



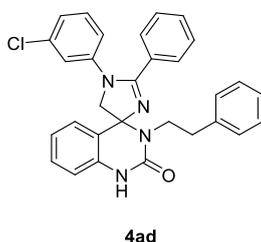
1-(4-Methoxyphenyl)-3'-phenethyl-2-phenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4aa): Petroleum ether/ethyl acetate = 2:1; white solid, 70% yield, 342 mg; mp 141.3.2 – 142.8 °C; ¹H NMR (400 MHz, CDCl₃) δ 7.71 (d, *J* = 7.2 Hz, 2H), 7.47 (t, *J* = 7.6 Hz, 1H), 7.38 (t, *J* = 7.6 Hz, 2H), 7.28 – 7.18 (m, 7H), 7.04 (t, *J* = 7.6 Hz, 1H), 6.93 – 6.67 (m, 5H), 4.22 – 4.09 (m, 2H), 3.97 – 3.93 (m, 1H), 3.77 (s, 3H), 3.44 – 3.31 (m, 1H), 3.29 – 3.06 (m, 2H); ¹³C{¹H} NMR (100 MHz, CDCl₃) δ 163.3, 157.2, 139.9, 134.8, 134.2, 130.9, 129.4, 129.2, 129.0, 128.5, 128.4, 126.3, 126.2, 125.8, 122.6, 114.5, 113.5, 68.8, 55.4, 45.4, 36.5; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₃₁H₂₉N₄O₂⁺, 489.2285; found 489.2284.



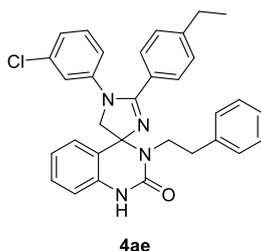
1-(4-Fluorophenyl)-3'-phenethyl-2-phenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4ab): Petroleum ether/ethyl acetate = 2:1; yellow solid, 51% yield, 243 mg; mp 197.2 – 198.5 °C; ¹H NMR (400 MHz, CDCl₃) δ 8.98 (d, *J* = 11.2 Hz, 1H), 7.70 (d, *J* = 7.6 Hz, 2H), 7.50 (t, *J* = 7.6 Hz, 1H), 7.40 (t, *J* = 7.6 Hz, 2H), 7.27 – 7.17 (m, 7H), 7.03 (t, *J* = 7.5 Hz, 1H), 6.94 – 6.84 (m, 5H), 4.21 – 4.13 (m, 2H), 3.96 (dt, *J* = 12.0, 6.4 Hz, 1H), 3.40 – 3.31 (m, 1H), 3.20 (ddd, *J* = 26.8, 12.0, 6.0 Hz, 2H); ¹³C{¹H} NMR (100 MHz, CDCl₃) δ 162.9, 161.1, 158.7, 152.4, 139.9, 138.0, 138.0, 134.4, 131.1, 129.5, 129.3, 128.9, 128.5 (d, *J* = 5 Hz), 126.2, 126.0, 125.6 (d, *J* = 8 Hz), 124.1, 122.5, 116.1 (d, *J* = 22 Hz), 113.8, 86.3, 68.5, 45.3, 36.6; ¹⁹F{¹H} NMR (376 MHz, CDCl₃) δ -116.5; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₃₀H₂₆FN₄O⁺, 477.2085; found 477.2080.



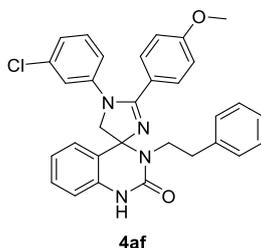
1,2-Bis(3-fluorophenyl)-3'-phenethyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4ac): Petroleum ether/ethyl acetate = 2:1; yellow solid, 43% yield, 212 mg, *d.r.* > 20:1; mp 197.2 – 199.6 °C; ¹H NMR (400 MHz, CDCl₃) δ 8.74 (s, 1H), 7.49 – 7.38 (m, 3H), 7.28 – 7.12 (m, 9H), 7.04 (t, *J* = 7.6 Hz, 1H), 6.90 – 6.79 (m, 2H), 6.59 (ddd, *J* = 16.4, 8.8, 2.0 Hz, 2H), 4.24 – 4.13 (m, 2H), 3.94 – 3.87 (m, 1H), 3.37 – 3.30 (m, 1H), 3.26 – 3.19 (m, 1H), 3.14 – 3.08 (m, 1H); ¹³C{¹H} NMR (100 MHz, CDCl₃) δ 163.9 (d, *J* = 31 Hz), 161.4 (d, *J* = 30 Hz), 161.0 (d, *J* = 3 Hz), 161.0, 152.3, 142.9, 142.8, 139.8, 134.4, 130.4 (q, *J* = 13 Hz), 129.5, 128.7 (d, *J* = 32 Hz), 126.3, 125.9, 124.9 (d, *J* = 4 Hz), 123.5, 122.6, 118.9 (d, *J* = 3 Hz), 118.3 (d, *J* = 21 Hz), 116.2 (d, *J* = 24 Hz), 113.9, 112.0 (d, *J* = 21 Hz), 110.6 (d, *J* = 24 Hz), 86.4, 67.7, 45.4, 36.4; ¹⁹F{¹H} NMR (376 MHz, CDCl₃) δ -110.5 (d, *J* = 8.4 Hz), -111.4 (d, *J* = 7.2 Hz); HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₃₀H₂₅F₂N₄O⁺, 495.1991; found 495.1990.



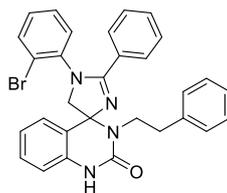
1-(3-Chlorophenyl)-3'-phenethyl-2-phenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4ad): Petroleum ether/ethyl acetate = 2:1; yellow solid, 54% yield, 266 mg, *d.r.* > 20:1; mp 198.5 – 199.6 °C; ¹H NMR (400 MHz, CDCl₃) δ 7.89 (s, 1H), 7.70 (d, *J* = 7.2 Hz, 2H), 7.53 (t, *J* = 7.2 Hz, 1H), 7.43 (t, *J* = 7.2 Hz, 2H), 7.28 – 7.18 (m, 4H), 7.15 (d, *J* = 8.0 Hz, 3H), 7.12 – 6.99 (m, 3H), 6.87 (s, 1H), 6.80 (d, *J* = 8.0 Hz, 1H), 6.67 (d, *J* = 8.0 Hz, 1H), 4.25 – 4.11 (m, 2H), 3.89 (ddd, *J* = 13.2, 10.8, 5.6 Hz, 1H), 3.37 – 3.26 (m, 1H), 3.20 (td, *J* = 11.6, 11.6, 5.6 Hz, 1H), 3.12 (dd, *J* = 10.8, 4.4 Hz, 1H). ¹³C{¹H} NMR (100 MHz, CDCl₃) δ 162.1, 152.4, 142.8, 139.9, 134.7, 134.5, 131.3, 130.0, 129.5, 129.4, 129.2, 128.9, 128.7, 128.5, 126.3, 126.0, 124.7, 123.7, 123.2, 122.5, 121.3, 113.9, 86.3, 67.7, 45.4, 36.5; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₃₀H₂₆ClN₄O⁺, 493.1790; found 493.1798.



1-(3-Chlorophenyl)-2-(4-ethylphenyl)-3'-phenethyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4ae): Petroleum ether/ethyl acetate = 2:1; yellow solid, 58% yield, 302 mg, *d.r.* > 20:1; mp 182.2 – 183. °C; ¹H NMR (400 MHz, CDCl₃) δ 9.43 (s, 1H), 7.51 (d, *J* = 8.0 Hz, 2H), 7.15 – 7.00 (m, 11H), 6.88 (t, *J* = 7.6 Hz, 1H), 6.79 (d, *J* = 8.0 Hz, 1H), 6.67 (d, *J* = 8.8 Hz, 2H), 4.04 (q, *J* = 11.2 Hz, 2H), 3.89 – 3.75 (m, 1H), 3.24 – 2.97 (m, 3H), 2.60 (q, *J* = 7.6 Hz, 2H), 1.16 (t, *J* = 7.6 Hz, 3H); ¹³C {¹H} NMR (100 MHz, CDCl₃) δ 162.4, 152.8, 147.8, 140.6, 140.0, 134.6, 130.0, 129.3, 129.3, 129.0, 128.5, 128.2, 126.8, 126.2, 126.1, 124.6, 123.9, 122.4, 114.0, 86.3, 68.1, 45.3, 36.6, 28.9, 15.3; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₃₂H₃₀ClN₄O⁺, 521.2103; found 521.2100.

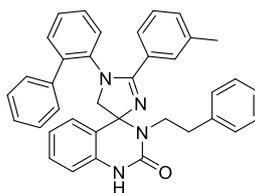


1-(3-Chlorophenyl)-2-(4-methoxyphenyl)-3'-phenethyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4af): Petroleum ether/ethyl acetate = 2:1; yellow solid, 74% yield, 386 mg, *d.r.* > 20:1; mp 171.3 – 173.2 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ 9.73 (s, 1H), 7.54 (d, *J* = 8.8 Hz, 2H), 7.27 – 7.15 (m, 5H), 7.10 (d, *J* = 9.6 Hz, 2H), 7.05 – 6.92 (m, 6H), 6.83 (dd, *J* = 17.6, 8.0 Hz, 2H), 4.22 (s, 2H), 3.80 (s, 3H), 3.59 (ddd, *J* = 13.2, 10.0, 6.4 Hz, 1H), 3.09 (ddd, *J* = 13.2, 10.0, 6.0 Hz, 1H), 2.99 – 2.81 (m, 2H); ¹³C {¹H} NMR (100 MHz, DMSO-*d*₆) δ 161.7, 160.6, 151.3, 143.7, 140.0, 135.4, 133.7, 131.0, 130.9, 129.3, 128.9, 128.9, 127.0, 126.6, 124.3, 124.2, 123.2, 122.1, 122.0, 114.5, 113.7, 86.3, 67.1, 55.8, 45.2, 36.4; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₃₁H₂₈ClN₄O₂⁺, 523.1895; found 523.1890.



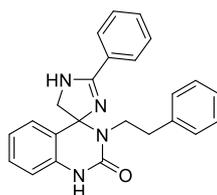
4ag

1-(2-Bromophenyl)-3'-phenethyl-2-phenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4ag): Petroleum ether/ethyl acetate = 2:1; yellow solid, 44% yield, 236 mg, *d.r.* = 1:1; mp 190.2 – 191.4 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ 9.75 and 9.73 (s, 1H), 7.70 (s, 1H), 7.57 – 7.46 (m, 2H), 7.45 – 7.32 (m, 3H), 7.13 (dt, *J* = 92.0, 7.2 Hz, 11H), 6.92 – 6.84 (m, 1H), 4.46 and 4.26 (s, 1H), 3.81 (s, 2H), 3.37 (s, 1H), 3.02 (s, 2H); ¹³C {¹H} NMR (100 MHz, DMSO-*d*₆) δ 163.6, 162.2, 163.6, 151.4, 140.1, 135.3, 134.0, 131.2, 129.9, 129.7, 129.4, 129.0, 128.9, 126.6, 124.4, 122.1, 113.8, 87.8, 87.2, 66.9, 66.5, 45.1, 36.5; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₃₀H₂₆BrN₄O⁺, 537.1285; found 537.1280.



4ah

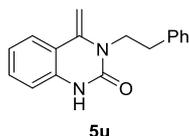
1-([1,1'-Biphenyl]-2-yl)-3'-phenethyl-2-(m-tolyl)-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4ah): Petroleum ether/ethyl acetate = 2:1; white solid, 50% yield, 274 mg, *d.r.* > 20:1; mp 162.2 – 164.5 °C; ¹H NMR (400 MHz, CDCl₃) δ 7.67 (s, 1H), 7.43 – 7.11 (m, 17H), 7.04 – 6.86 (m, 3H), 6.74 (s, 1H), 3.86 – 3.53 (m, 3H), 3.16 (s, 3H), 2.37 (s, 3H); ¹³C {¹H} NMR (100 MHz, CDCl₃) δ 152.2, 140.1, 139.2, 138.3, 138.2, 134.2, 131.6, 131.3, 130.1, 129.5, 129.0, 129.0, 128.7, 128.5, 128.4, 128.3, 128.0, 127.7, 127.5, 126.9, 126.5, 126.2, 126.1, 124.2, 122.2, 113.5, 67.5, 45.4, 36.4, 21.4; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₃₇H₃₃N₄O⁺, 549.2649; found 549.2655.



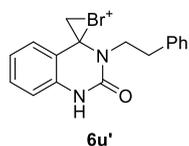
4ai

3'-Phenethyl-2-phenyl-1,5-dihydro-1'H-spiro[imidazole-4,4'-quinazolin]-2'(3'H)-one (4ai):

Petroleum ether/ethyl acetate = 2:1; white solid, 69% yield, 264 mg; mp 189.2 – 190.5 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ 9.65 (s, 1H), 8.01 (d, *J* = 6.8 Hz, 2H), 7.56 – 7.50 (m, 3H), 7.20 – 7.12 (m, 5H), 7.00 – 6.90 (m, 3H), 6.85 (d, *J* = 8.0 Hz, 1H), 4.19 – 3.52 (m, 3H), 3.03 – 2.90 (m, 3H); ¹³C {¹H} NMR (100 MHz, DMSO-*d*₆) δ 168.4, 151.2, 140.1, 135.4, 131.5, 130.1, 129.2, 129.0, 128.8, 127.98, 126.8, 126.5, 121.9, 113.6, 79.7, 44.9, 36.7; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₂₄H₂₃N₄O⁺, 383.1866; found 383.1869.



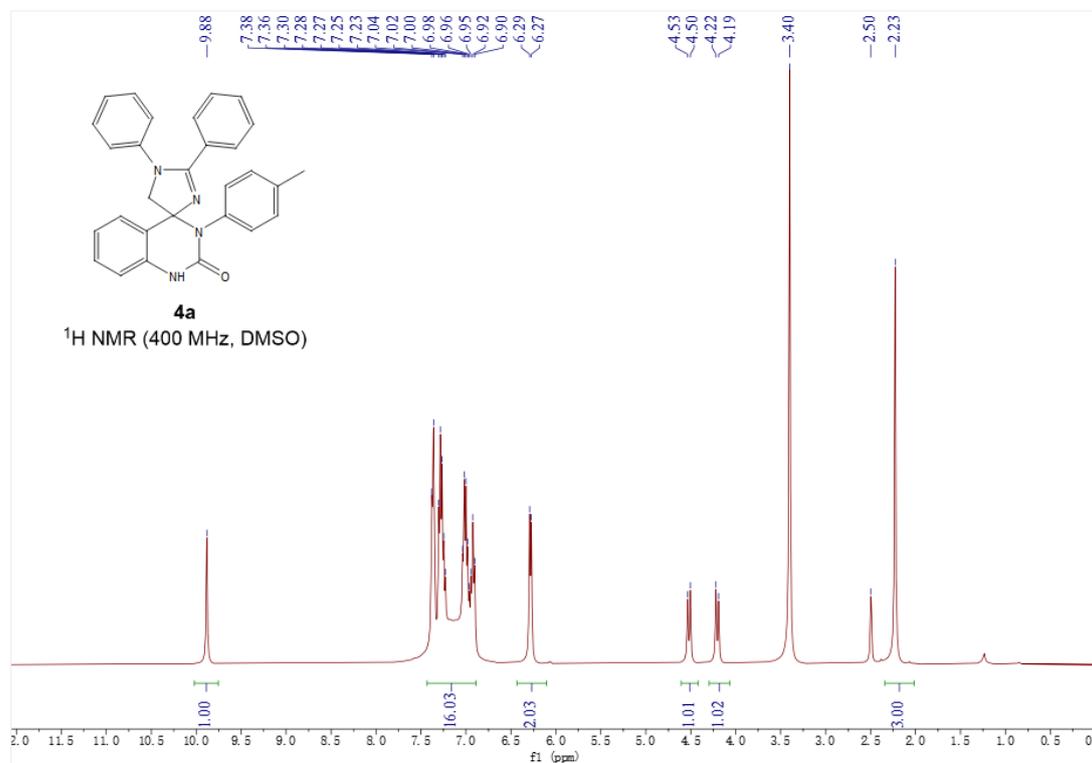
4-Methylene-3-phenethyl-3,4-dihydroquinazolin-2(1H)-one (5u): white solid, 90% yield, 2.37 g; mp 172.8 – 173.9 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.16 (s, 1H), 7.69 (d, *J* = 8.0 Hz, 1H), 7.32 – 7.21 (m, 6H), 6.97 (t, *J* = 7.6 Hz, 1H), 6.90 (d, *J* = 8.0 Hz, 1H), 4.96 (d, *J* = 2.6 Hz, 1H), 4.42 (d, *J* = 2.6 Hz, 1H), 3.98 – 3.90 (m, 2H), 2.91 – 2.84 (m, 2H); ¹³C {¹H} NMR (100 MHz, DMSO-*d*₆) δ 149.9, 140.2, 139.3, 136.1, 130.6, 129.1, 128.9, 126.8, 124.5, 122.5, 116.5, 115.0, 84.3, 44.3, 31.8; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₁₇H₁₇N₂O⁺, 265.1335; found 265.1337.



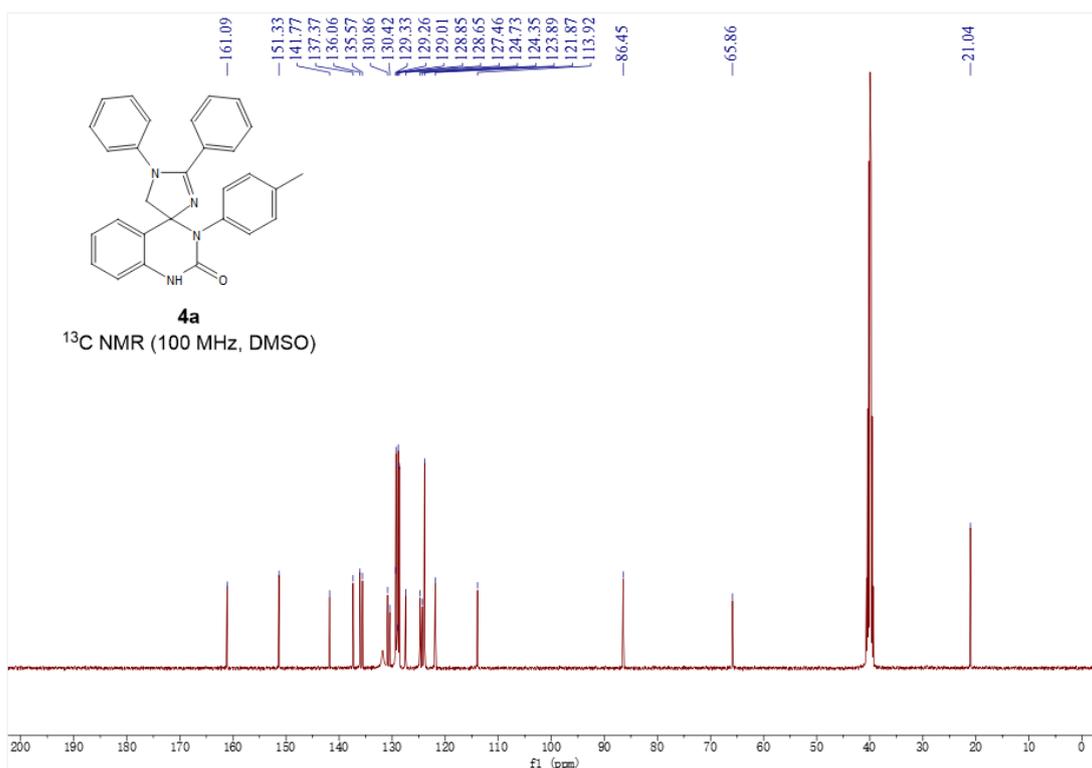
2'-oxo-3'-phenethyl-2',3'-dihydro-1'H-spiro[bromirane-2,4'-quinazolin]-1-ium (6u'): Petroleum ether/ethyl acetate = 2:1; yellow solid, 60% yield, 205 mg; mp 213.4 – 215.2 °C; ¹H NMR (400 MHz, DMSO-*d*₆) δ 7.70 (d, *J* = 7.8 Hz, 1H), 7.35 – 7.21 (m, 6H), 7.03 (t, *J* = 7.6 Hz, 1H), 6.89 (d, *J* = 8.0 Hz, 1H), 6.44 (s, 1H), 3.77 – 3.60 (m, 2H), 2.98 – 2.92 (m, 1H), 2.84 – 2.77 (m, 1H); ¹³C {¹H} NMR (100 MHz, DMSO-*d*₆) δ 152.1, 140.2, 137.4, 130.6, 129.0, 128.9, 128.9, 126.5, 120.6, 118.8, 113.6, 87.6, 54.6, 43.0, 36.3; HRMS (ESI-TOF) *m/z* [M + H]⁺ calcd for C₁₇H₁₆BrN₂O⁺, 343.0441; found 343.0447.

5. ^1H NMR, ^{13}C NMR and ^{19}F NMR spectra of products

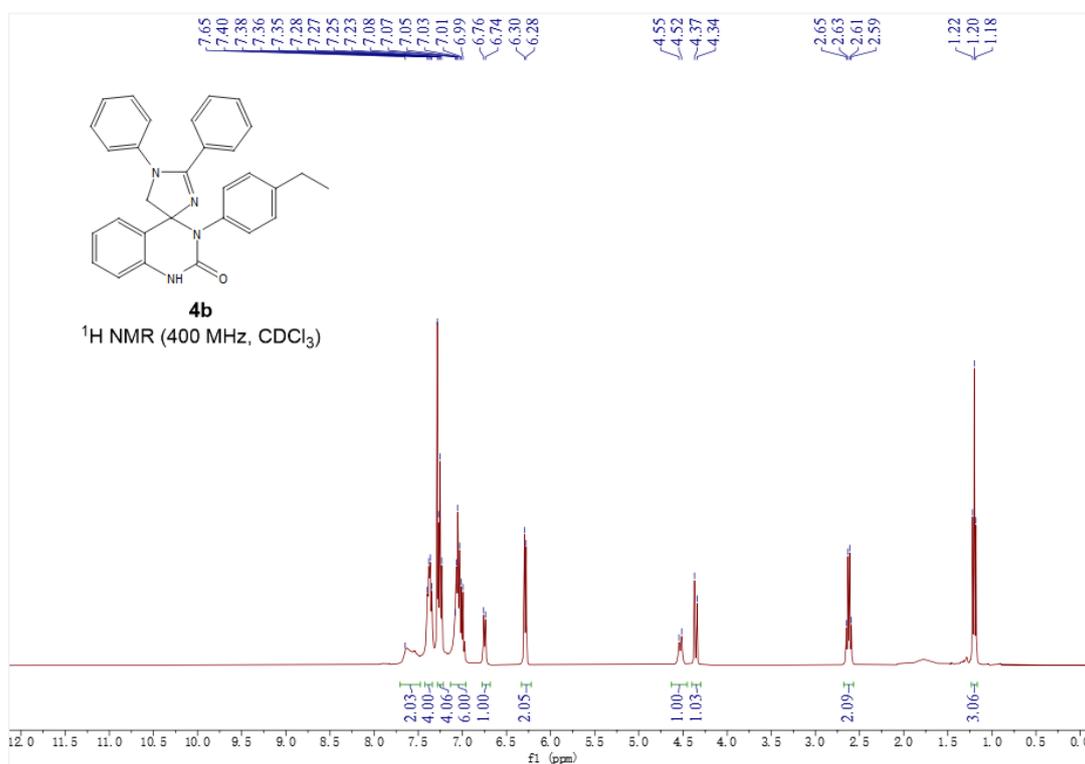
^1H NMR (400 MHz, DMSO) spectrum of **4a**



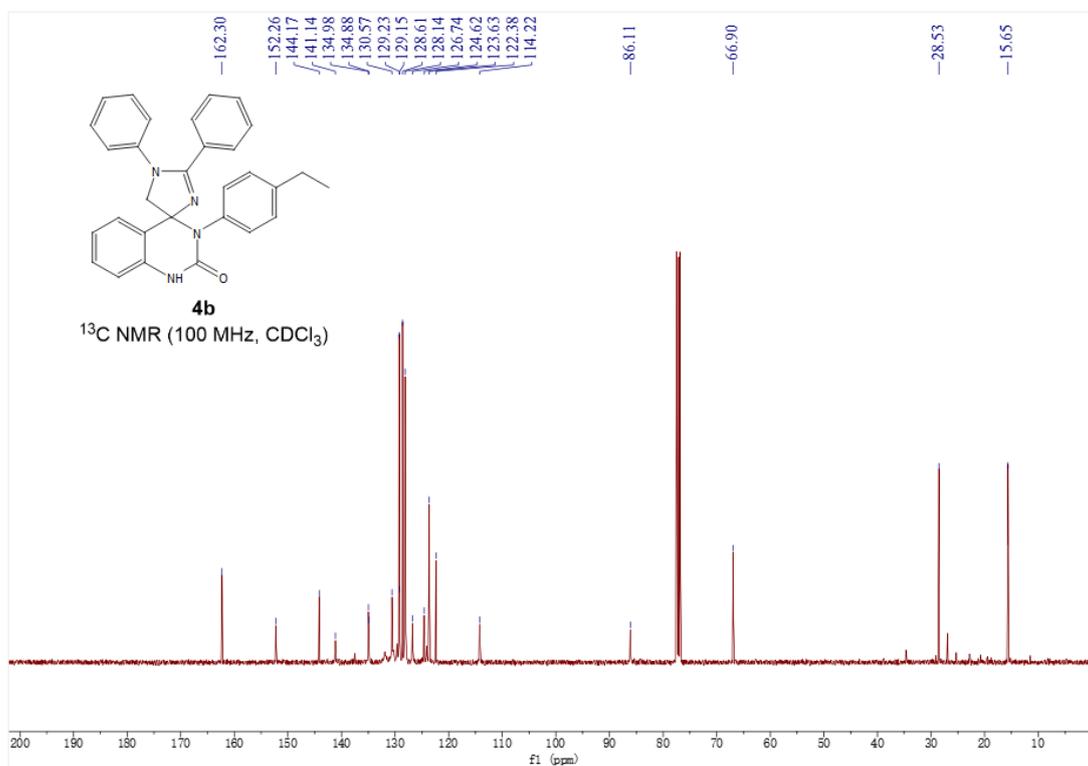
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO) spectrum of **4a**



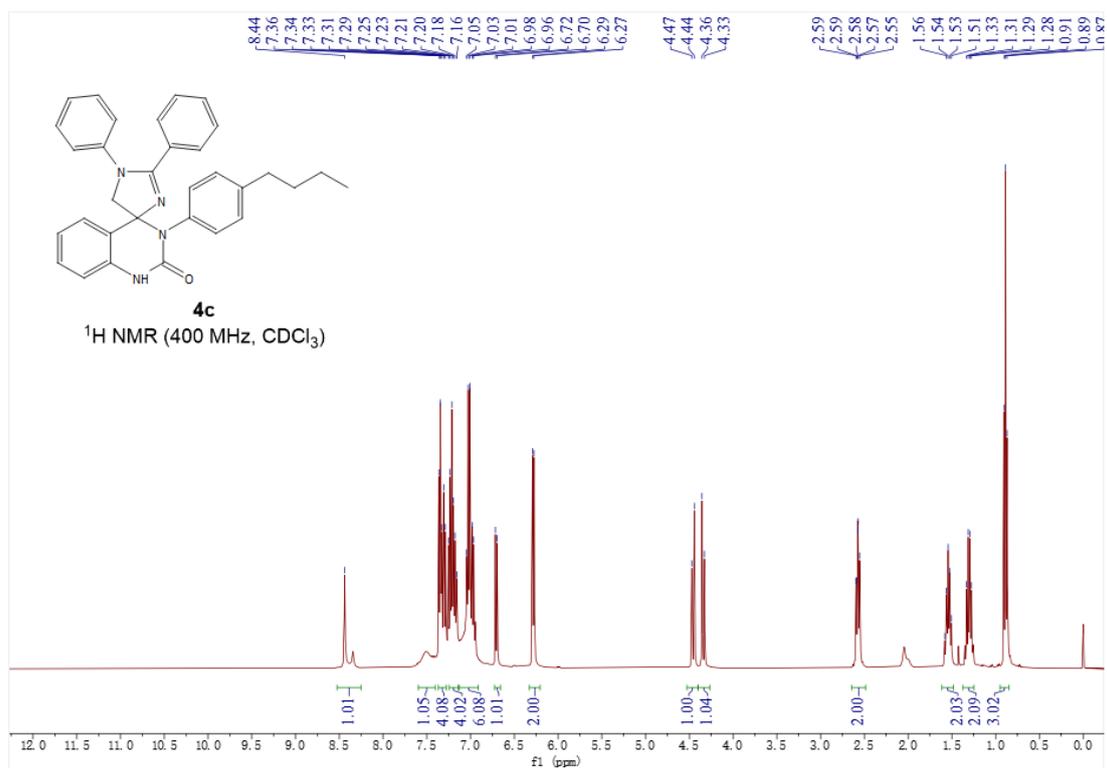
^1H NMR (400 MHz, CDCl_3) spectrum of **4b**



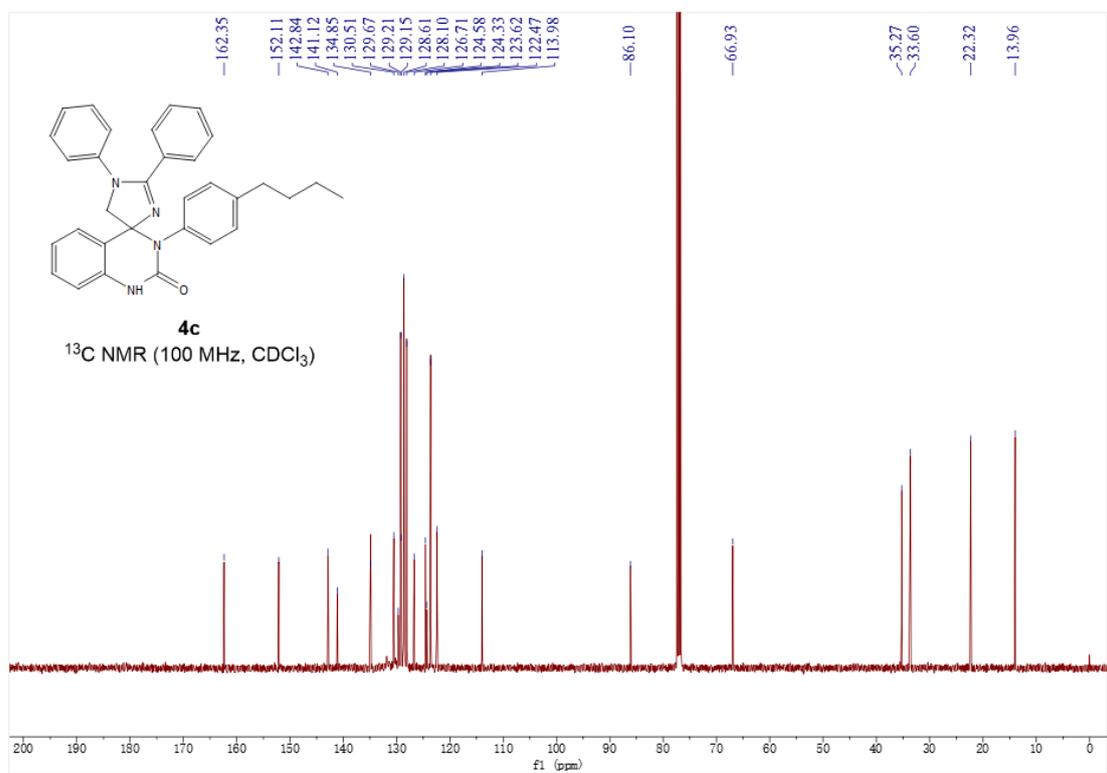
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4b**



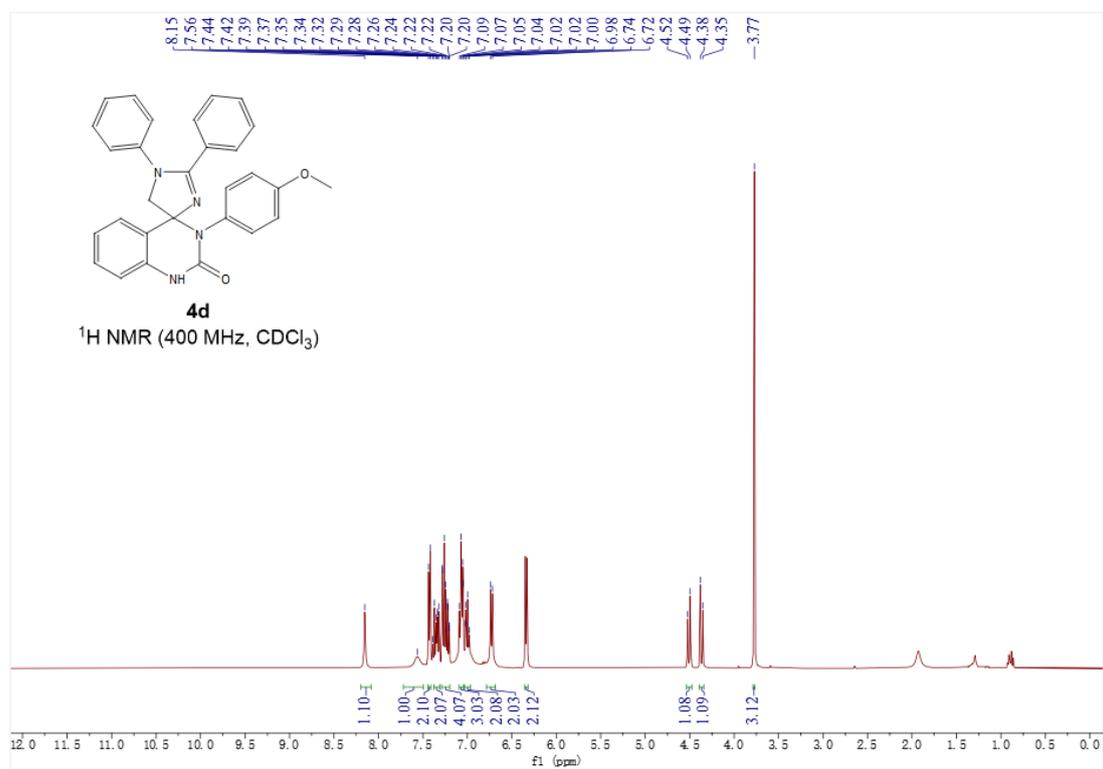
^1H NMR (400 MHz, CDCl_3) spectrum of **4c**



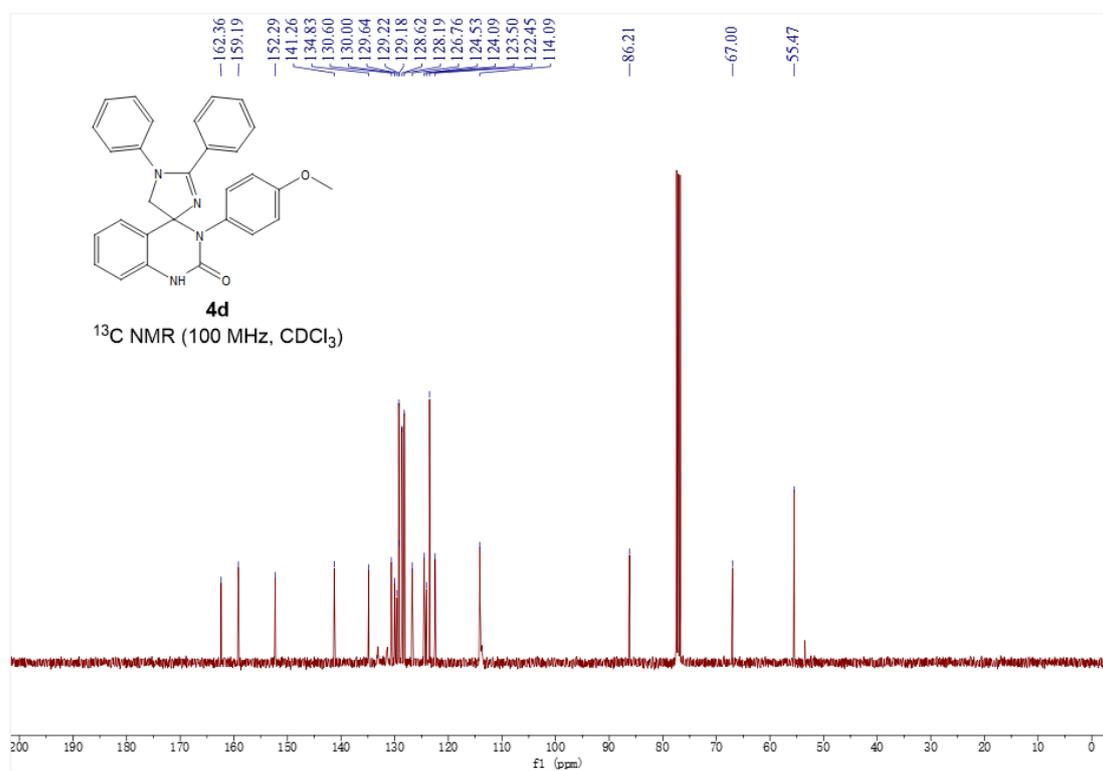
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4c**



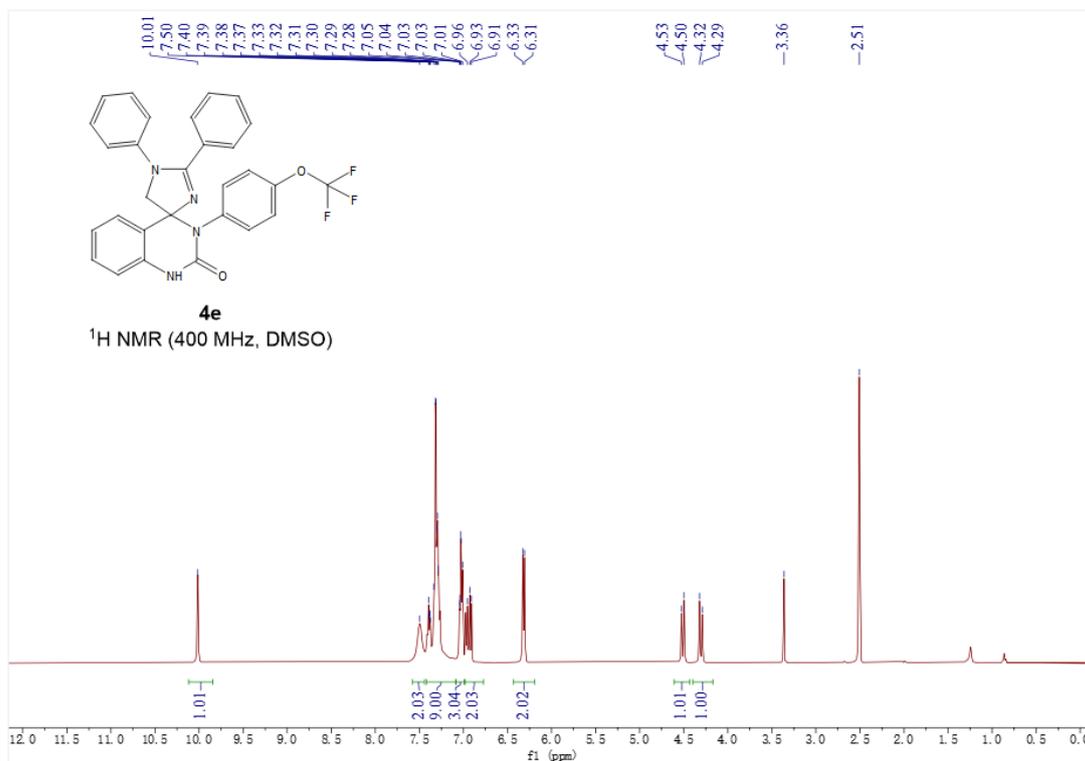
^1H NMR (400 MHz, CDCl_3) spectrum of **4d**



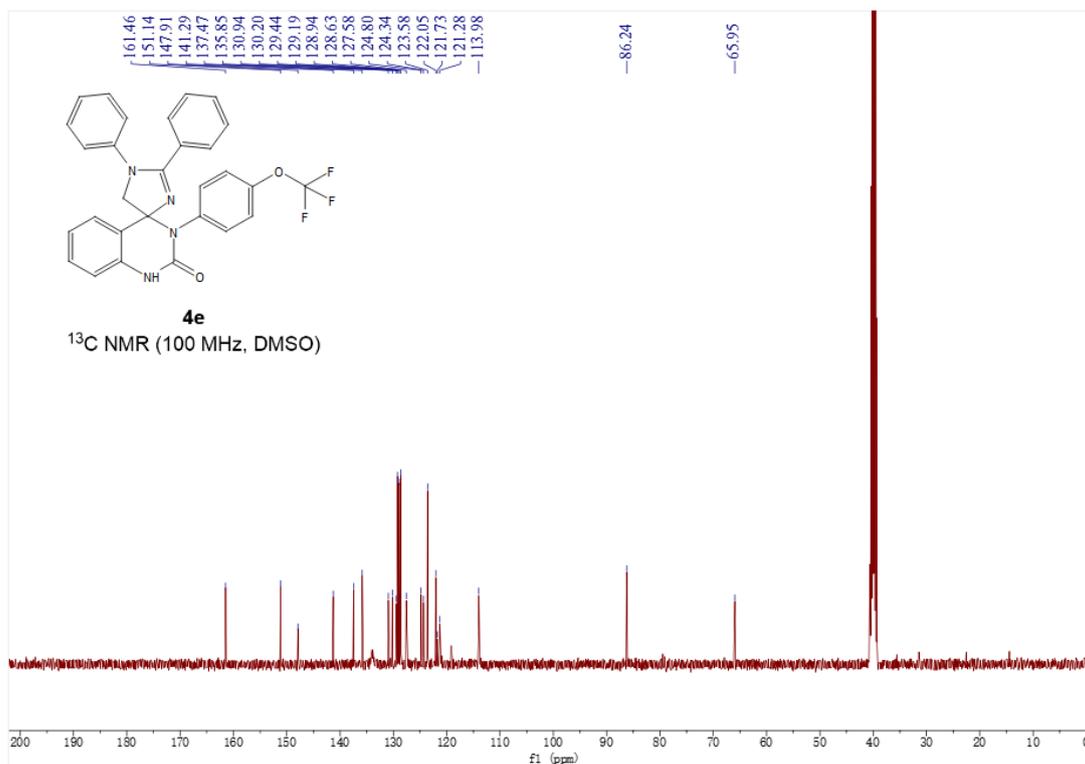
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4d**



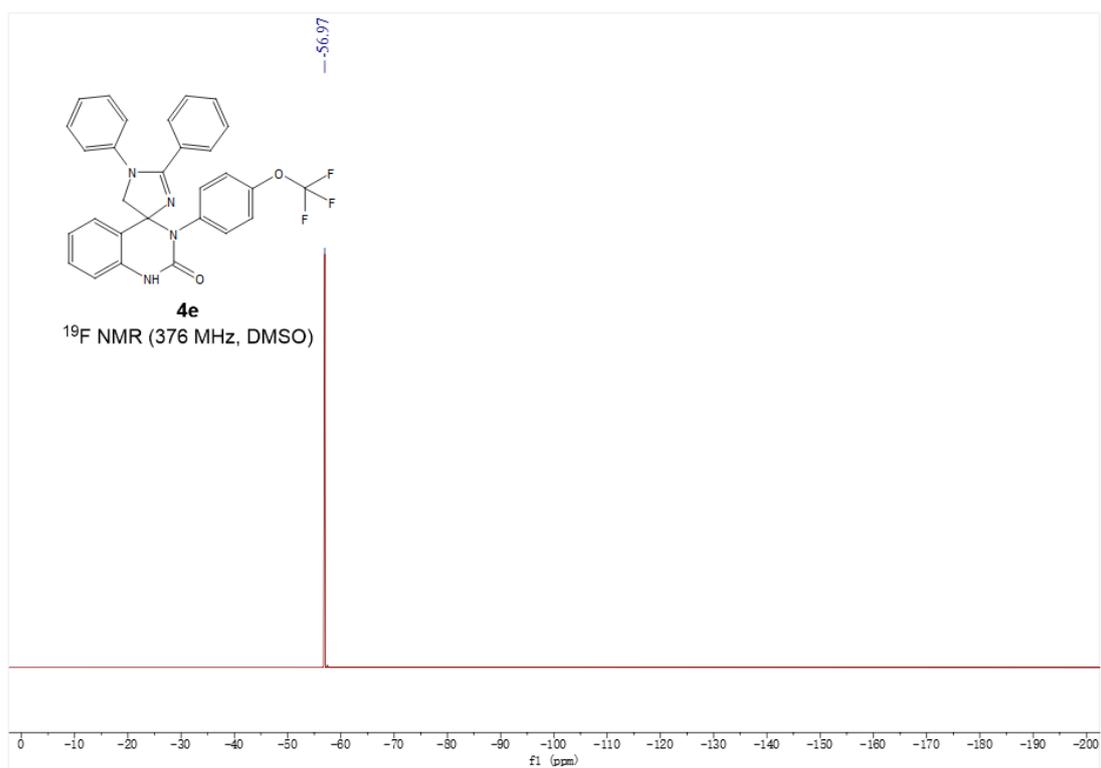
^1H NMR (400 MHz, DMSO) spectrum of **4e**



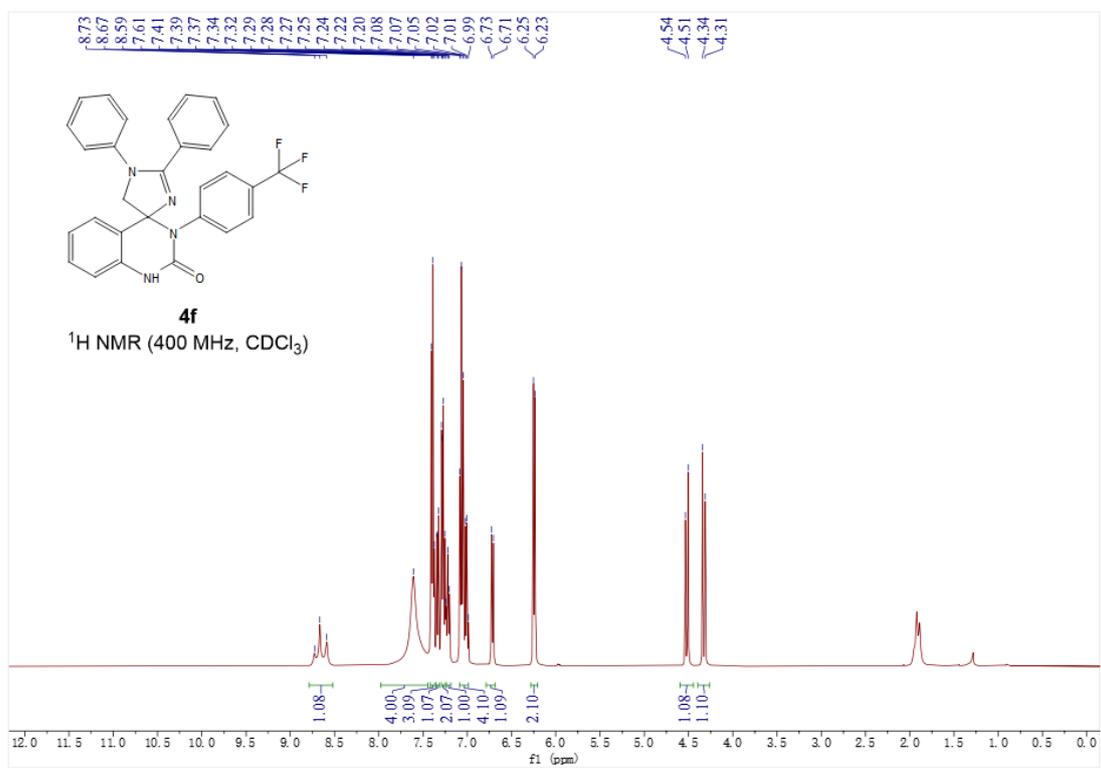
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO) spectrum of **4e**



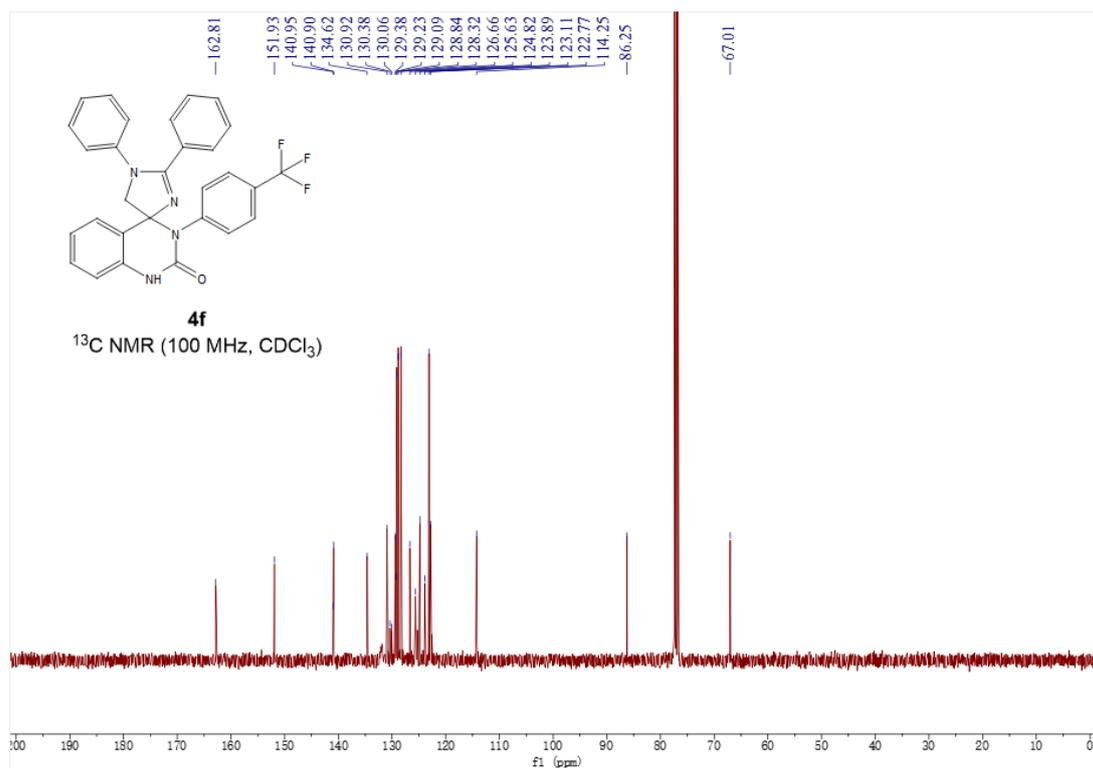
^{19}F $\{^1\text{H}\}$ NMR (376 MHz, DMSO) spectrum of **4e**



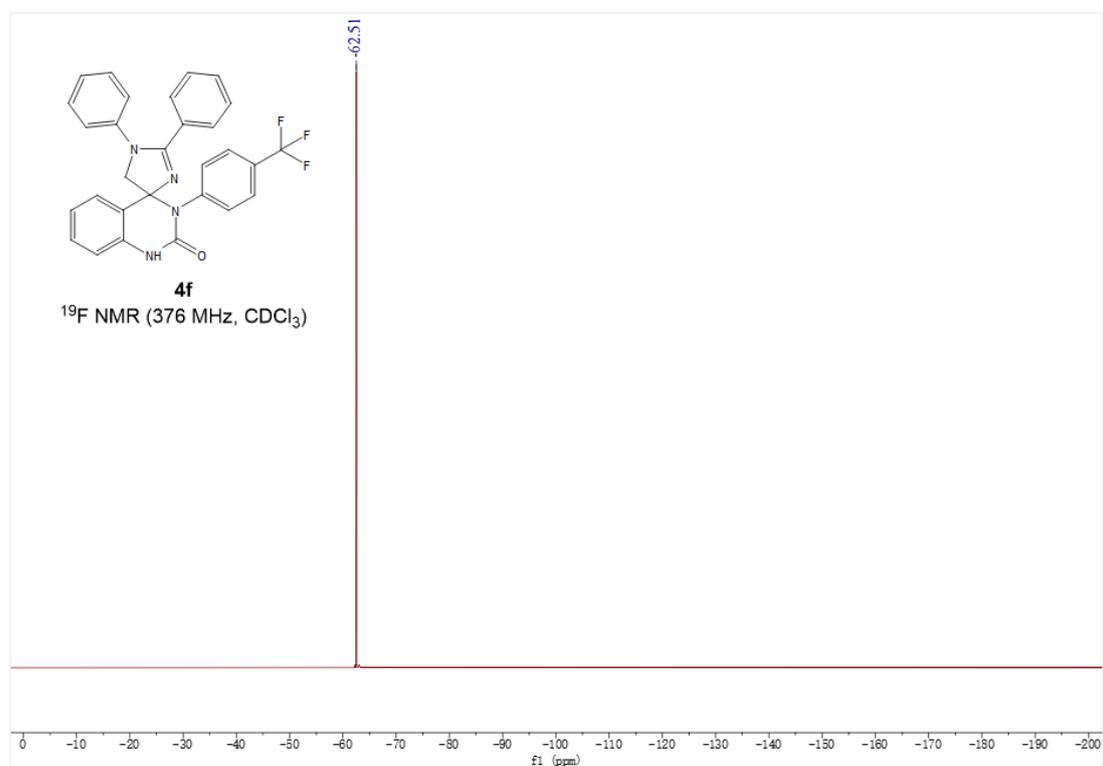
^1H NMR (400 MHz, CDCl_3) spectrum of **4f**



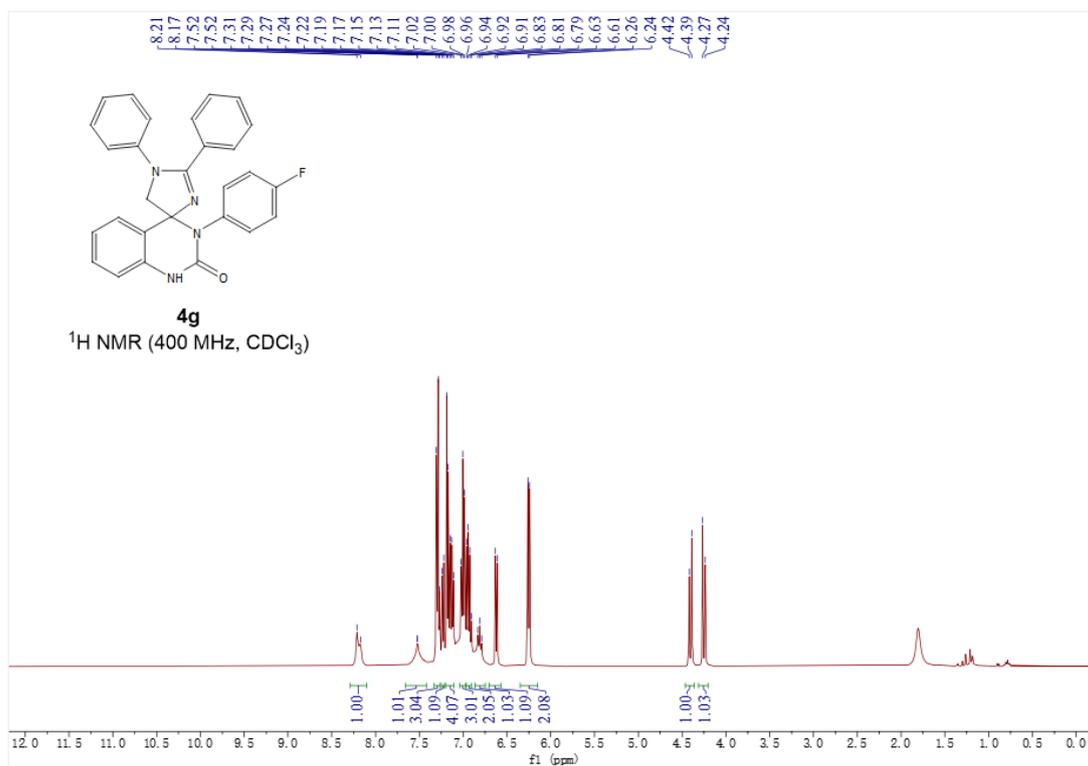
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4f**



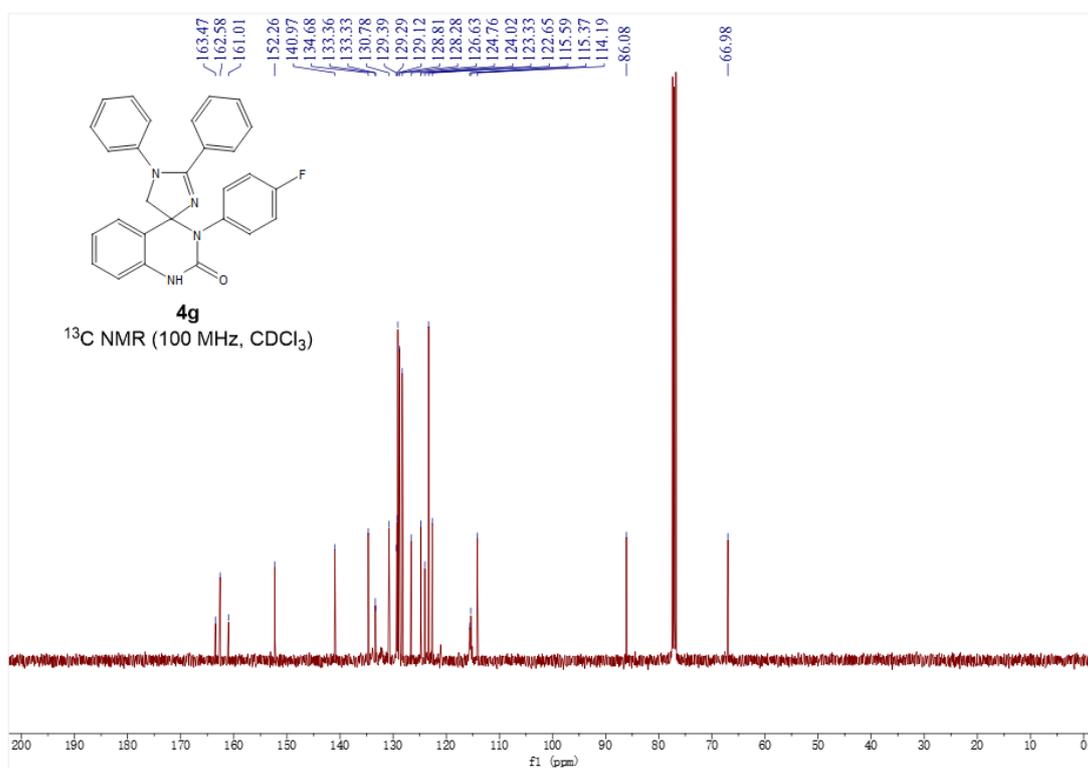
$^{19}\text{F}\{^1\text{H}\}$ NMR (376 MHz, CDCl_3) spectrum of **4f**



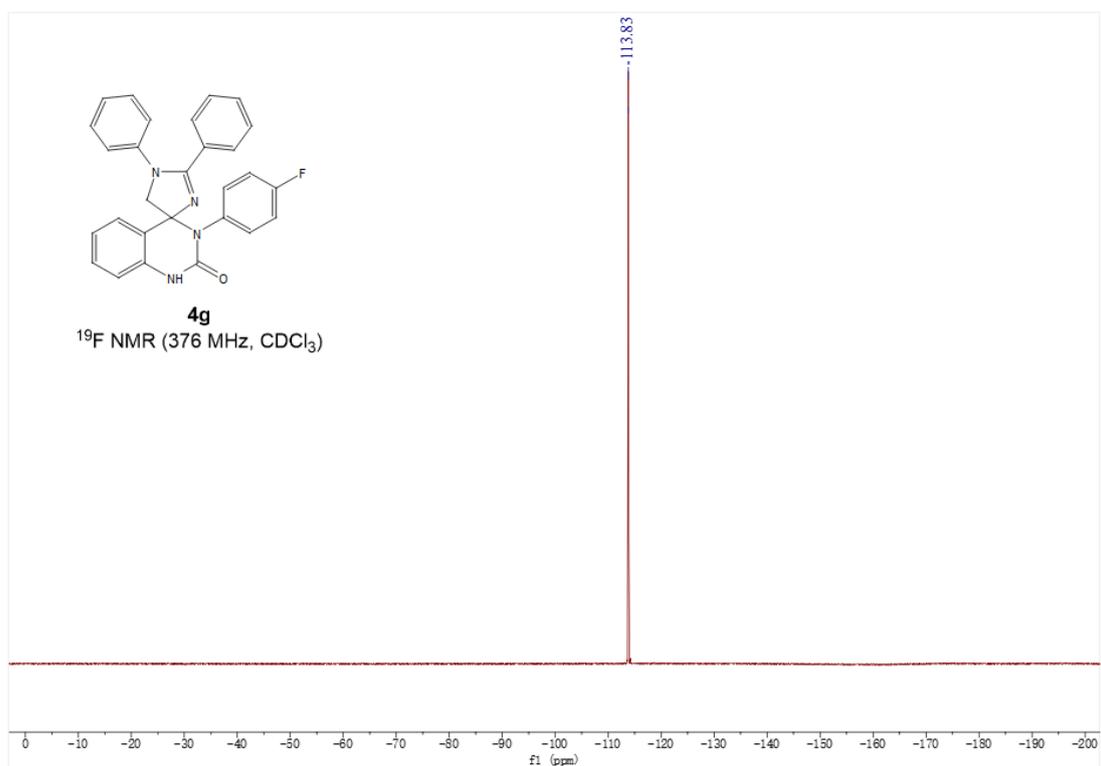
^1H NMR (400 MHz, CDCl_3) spectrum of **4g**



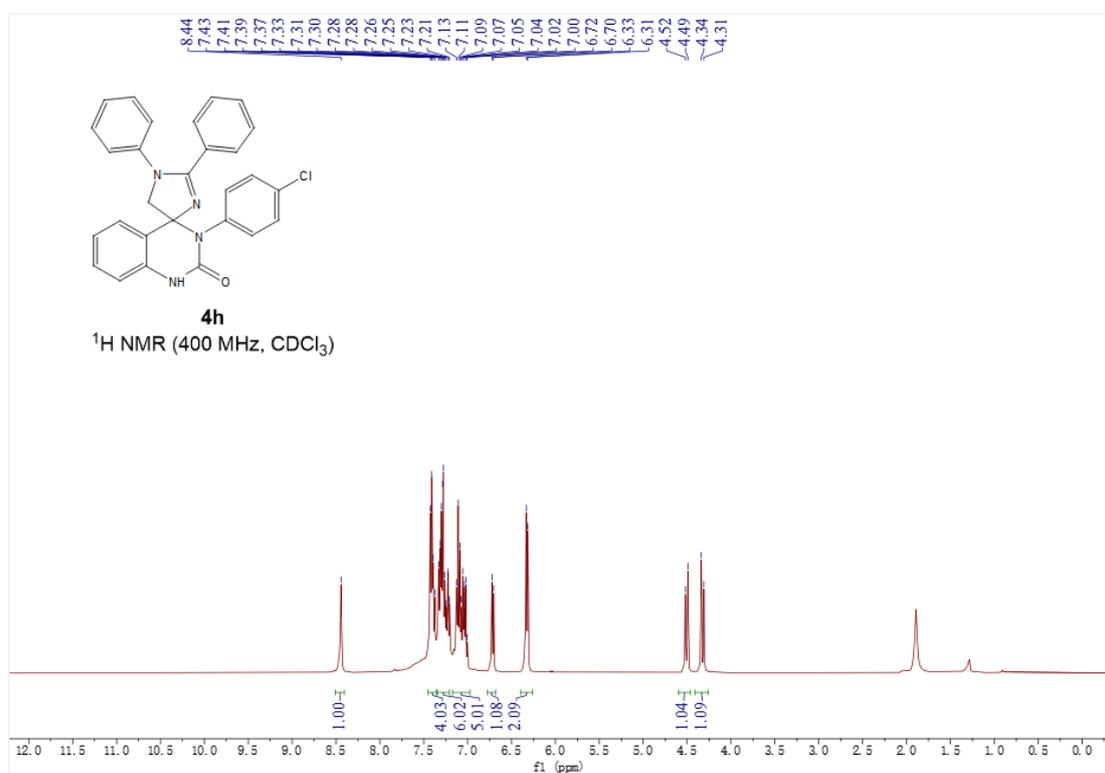
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4g**



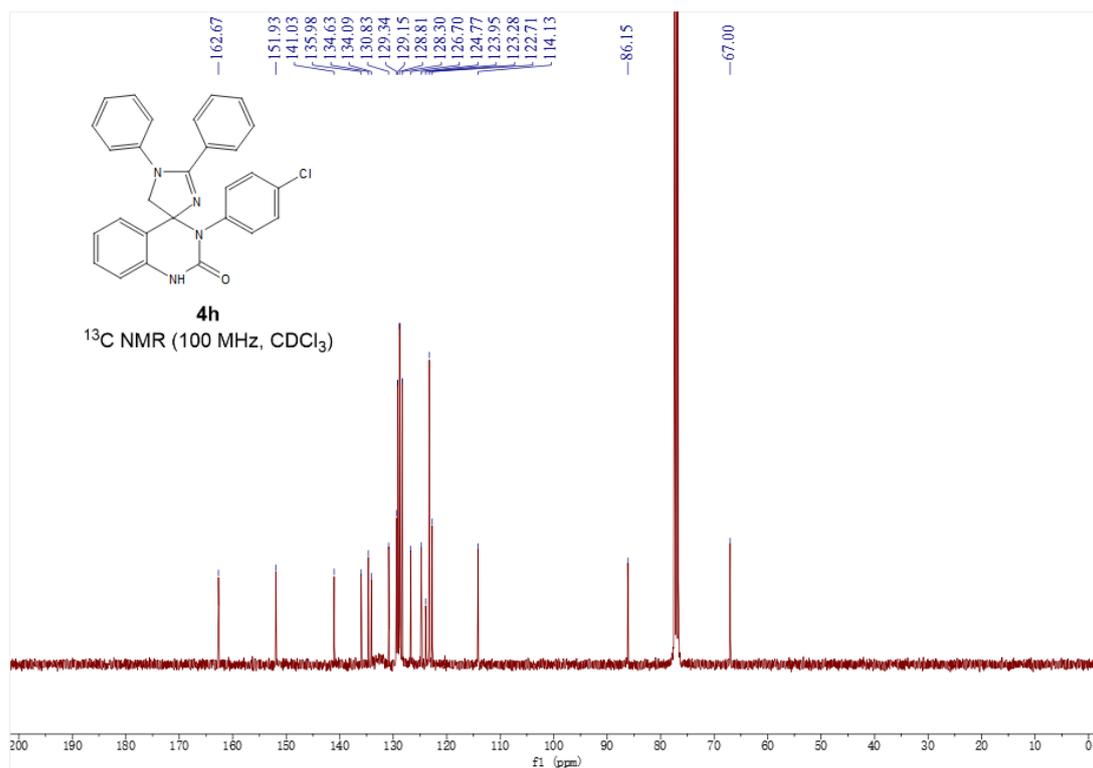
$^{19}\text{F}\{^1\text{H}\}$ NMR (376 MHz, CDCl_3) spectrum of **4g**



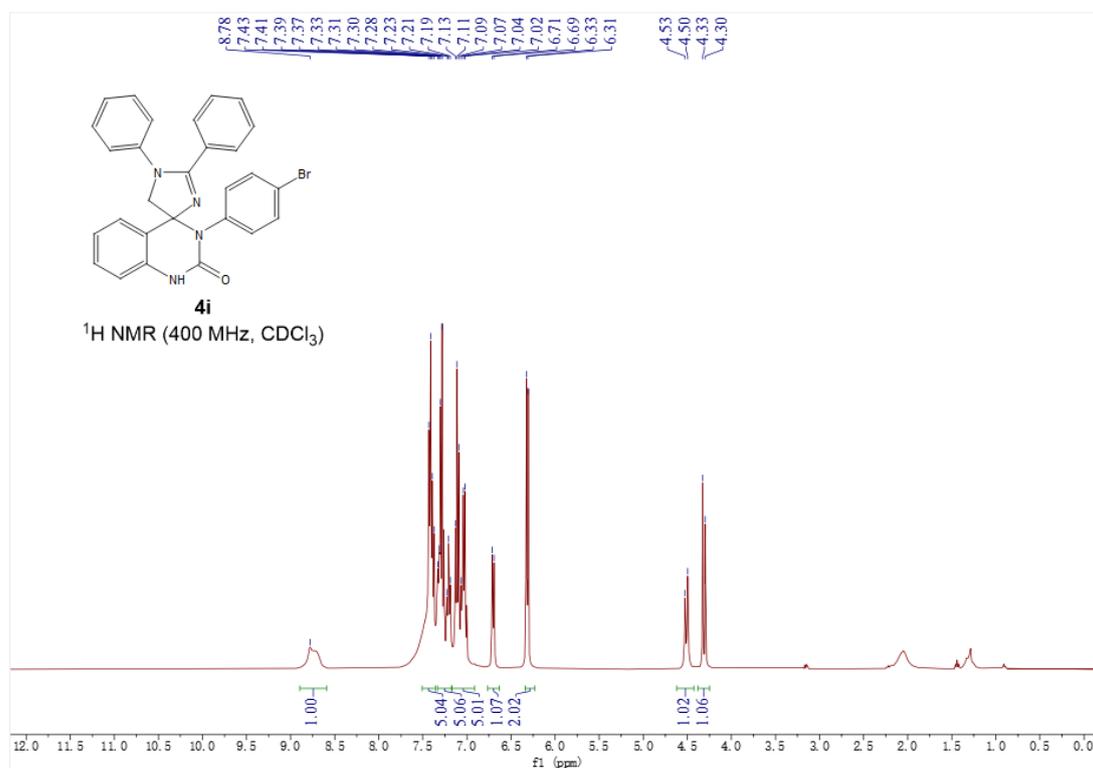
^1H NMR (400 MHz, CDCl_3) spectrum of **4h**



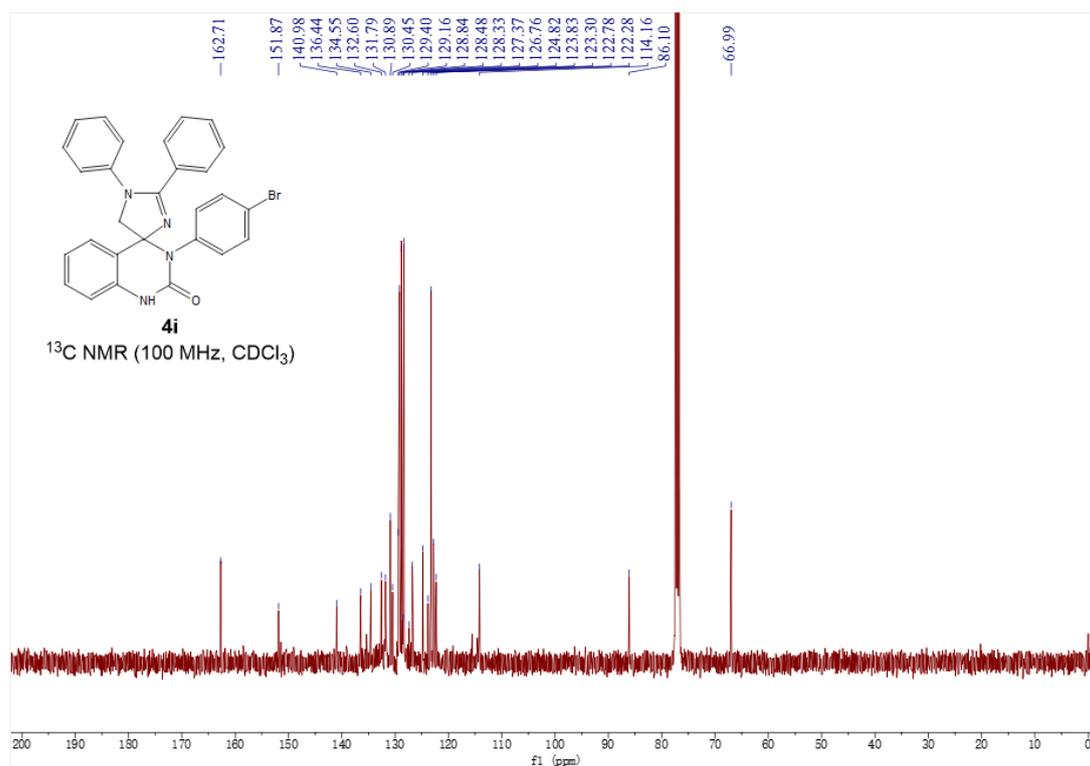
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4h**



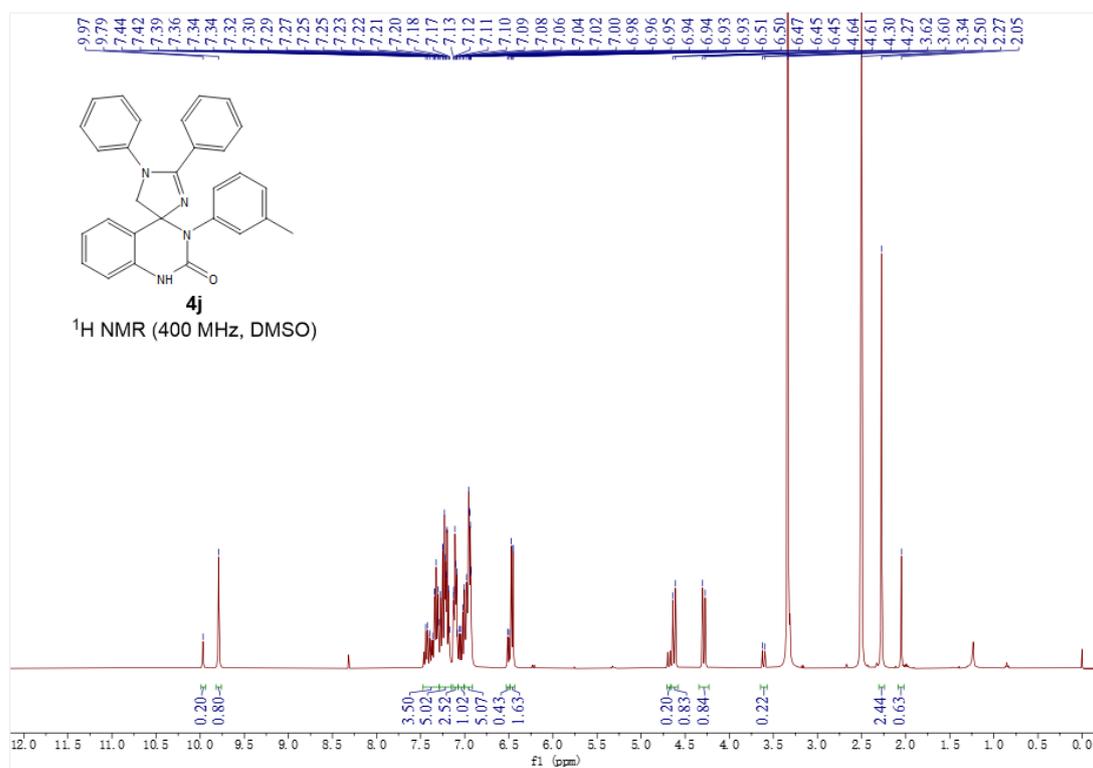
^1H NMR (400 MHz, CDCl_3) spectrum of **4i**



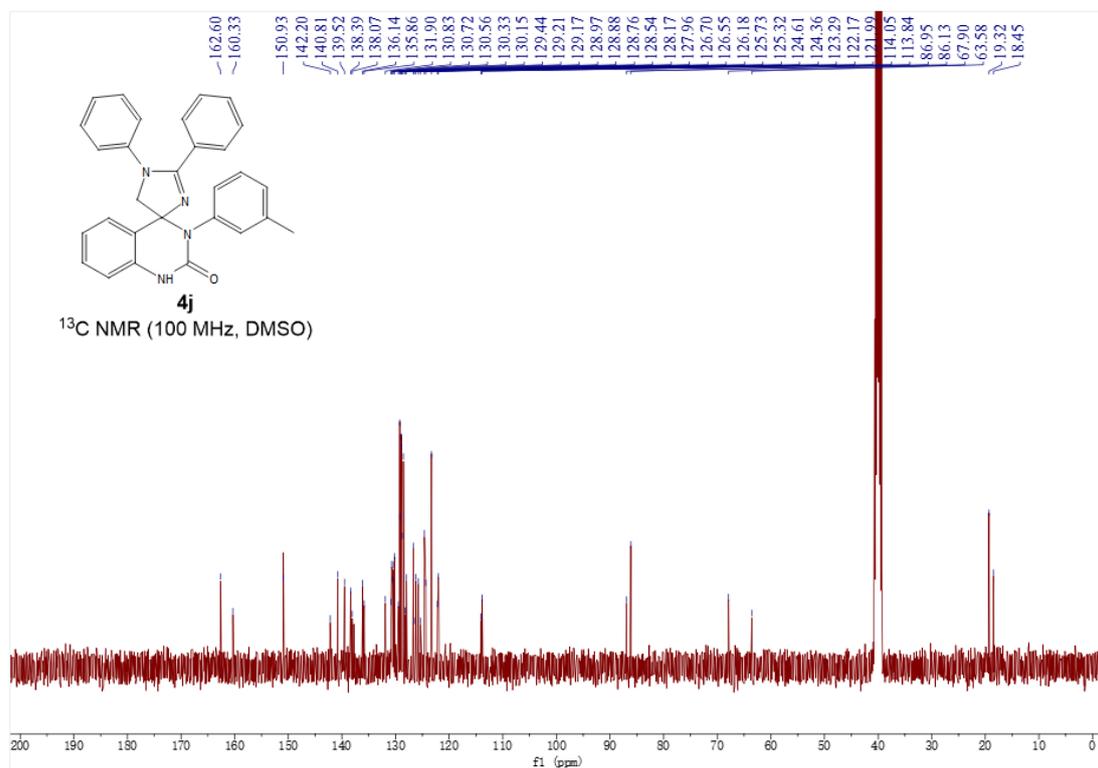
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4i**



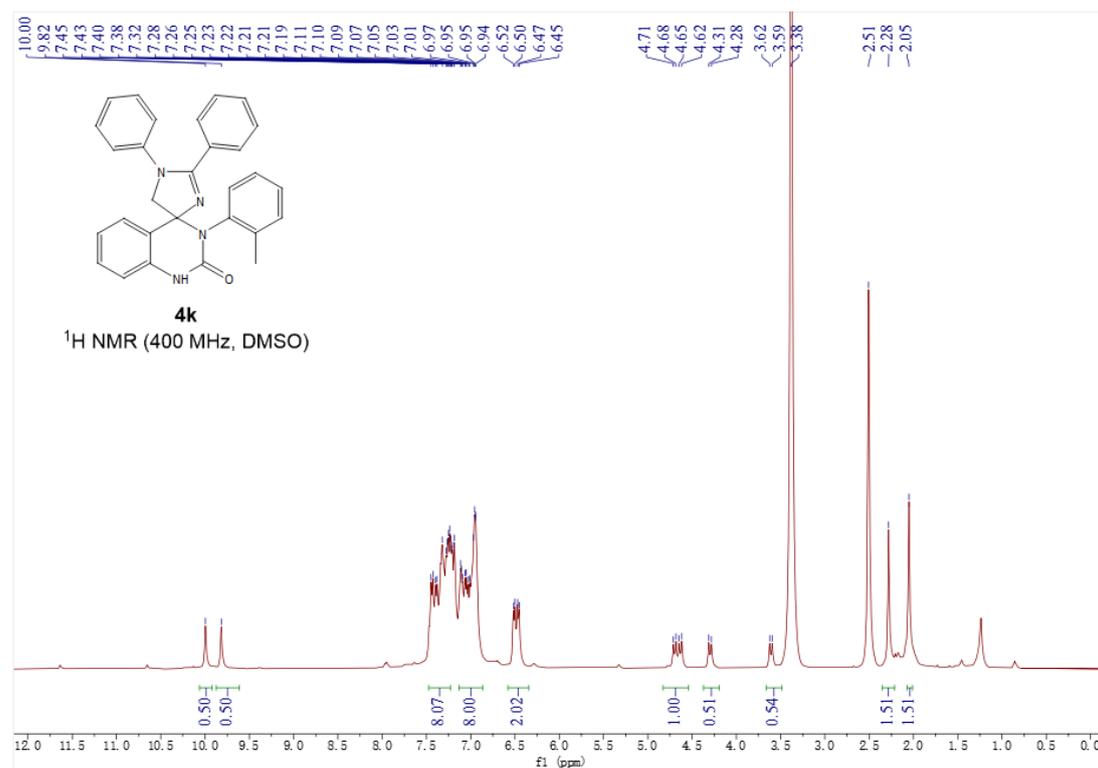
^1H NMR (400 MHz, DMSO) spectrum of **4j**



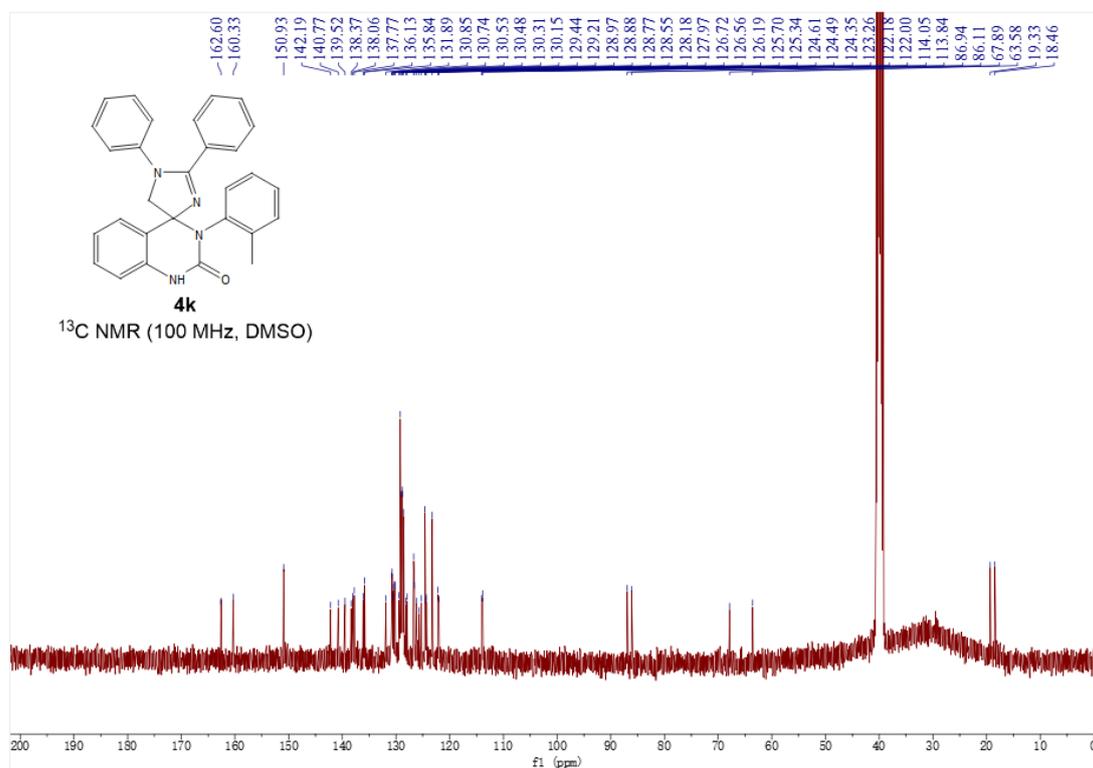
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO) spectrum of **4j**



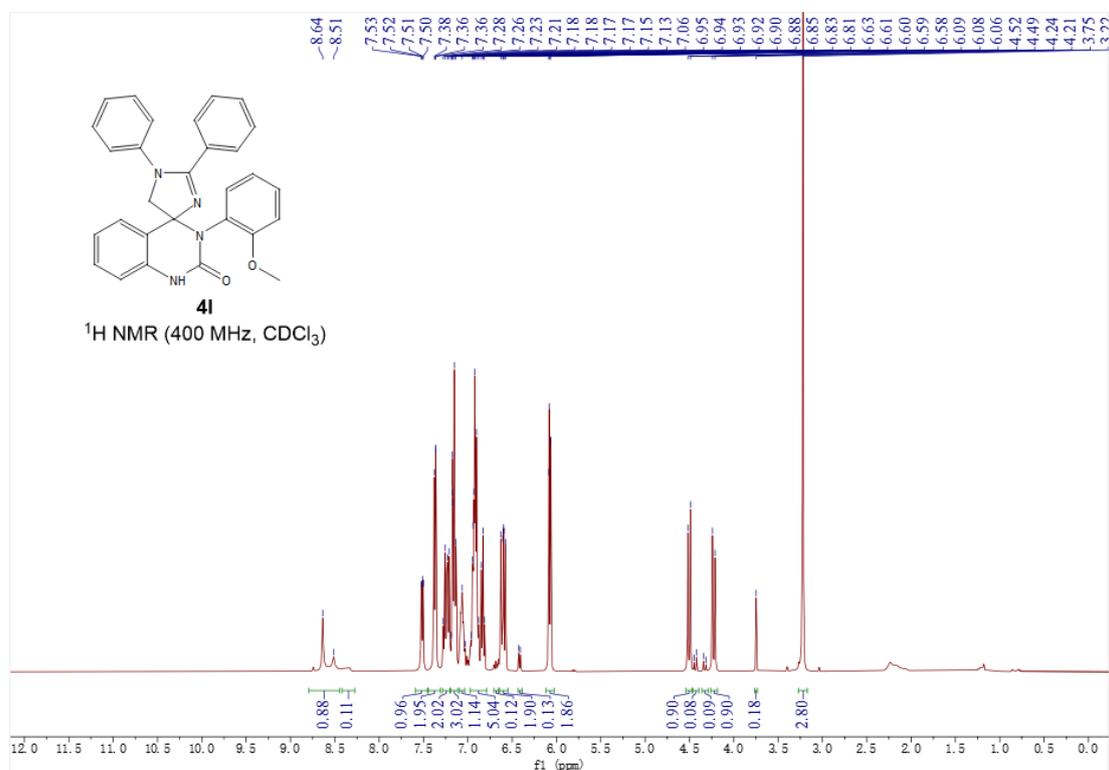
^1H NMR (400 MHz, DMSO) spectrum of **4k**



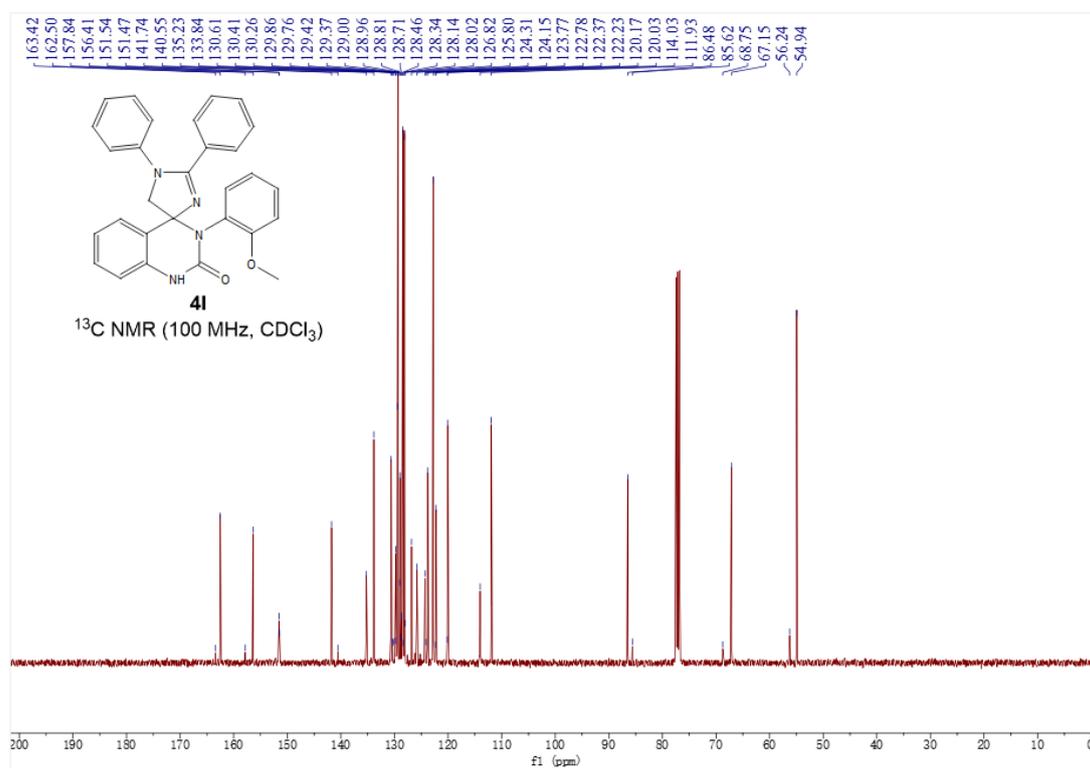
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO) spectrum of **4k**



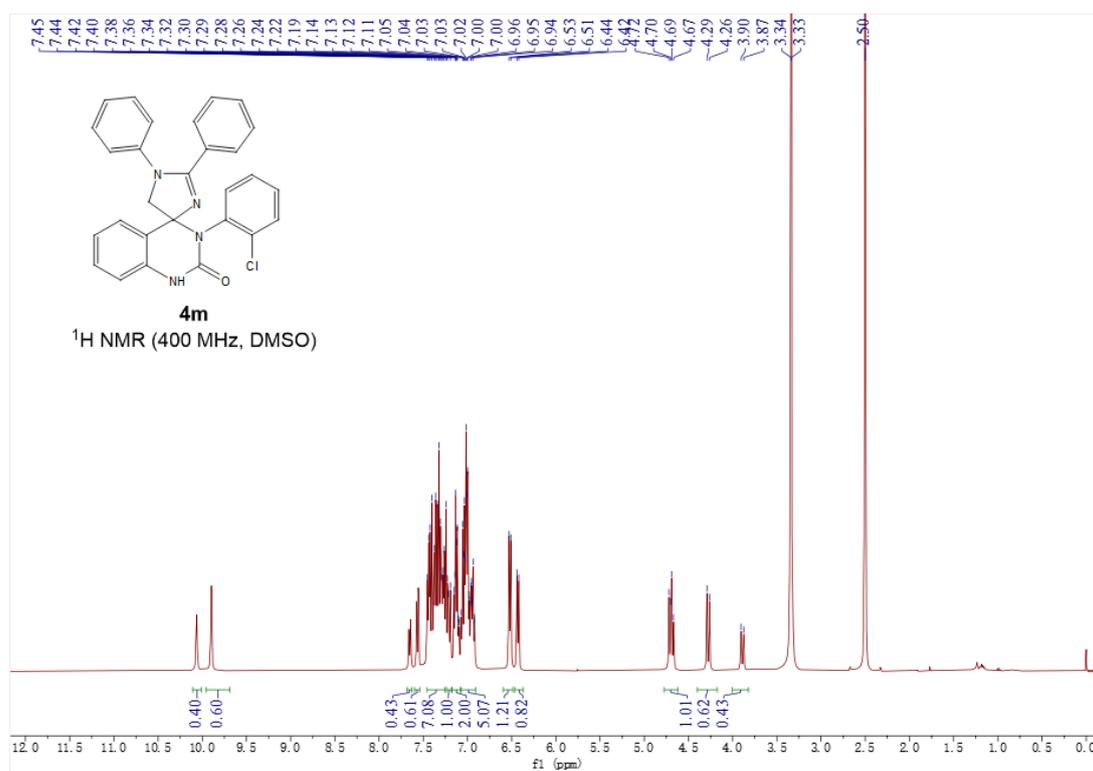
^1H NMR (400 MHz, CDCl_3) spectrum of **4l**



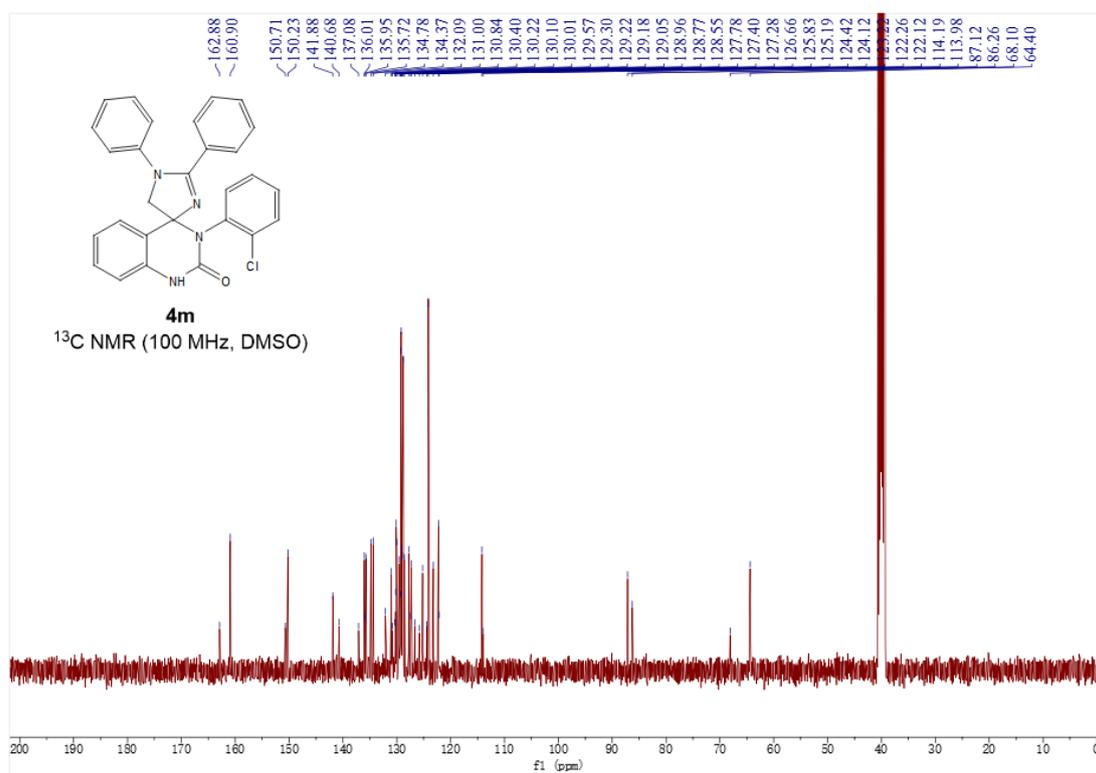
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4l**



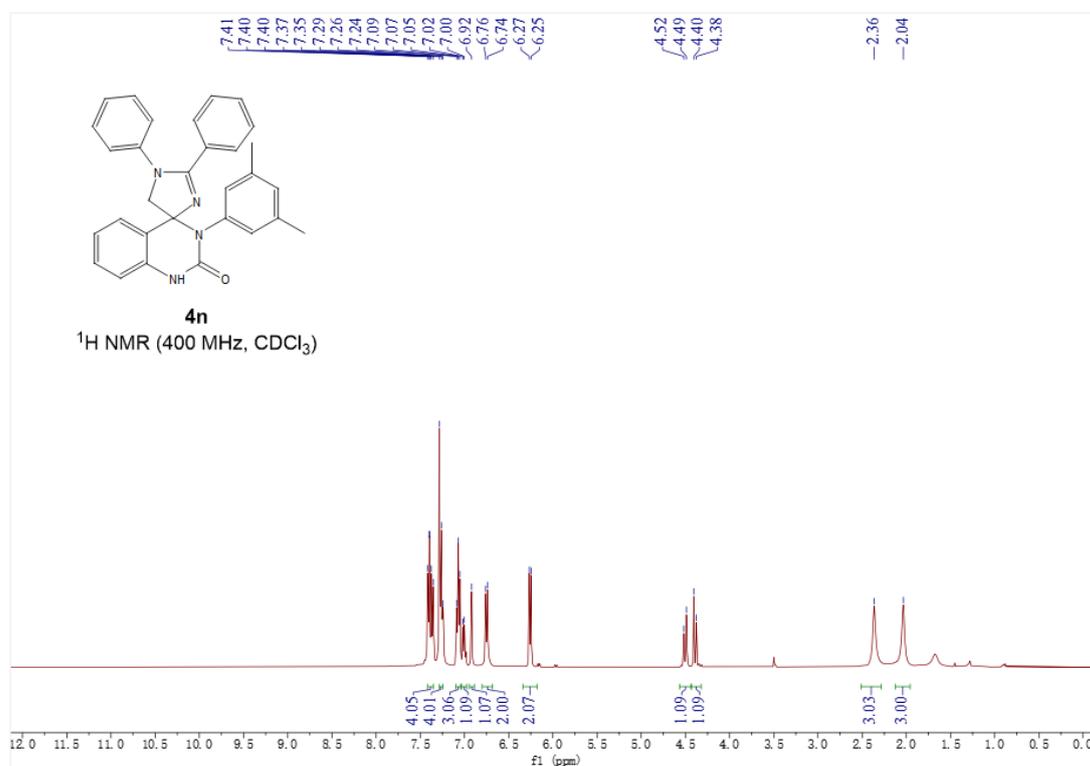
^1H NMR (400 MHz, DMSO) spectrum of **4m**



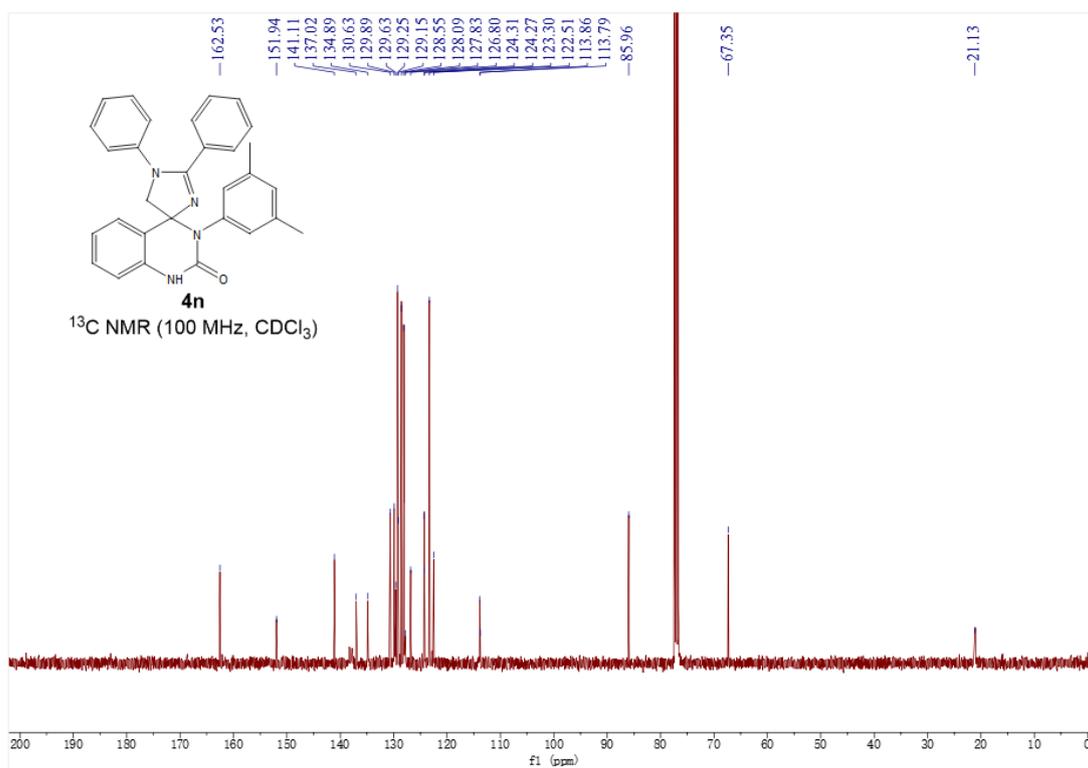
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO) spectrum of **4m**



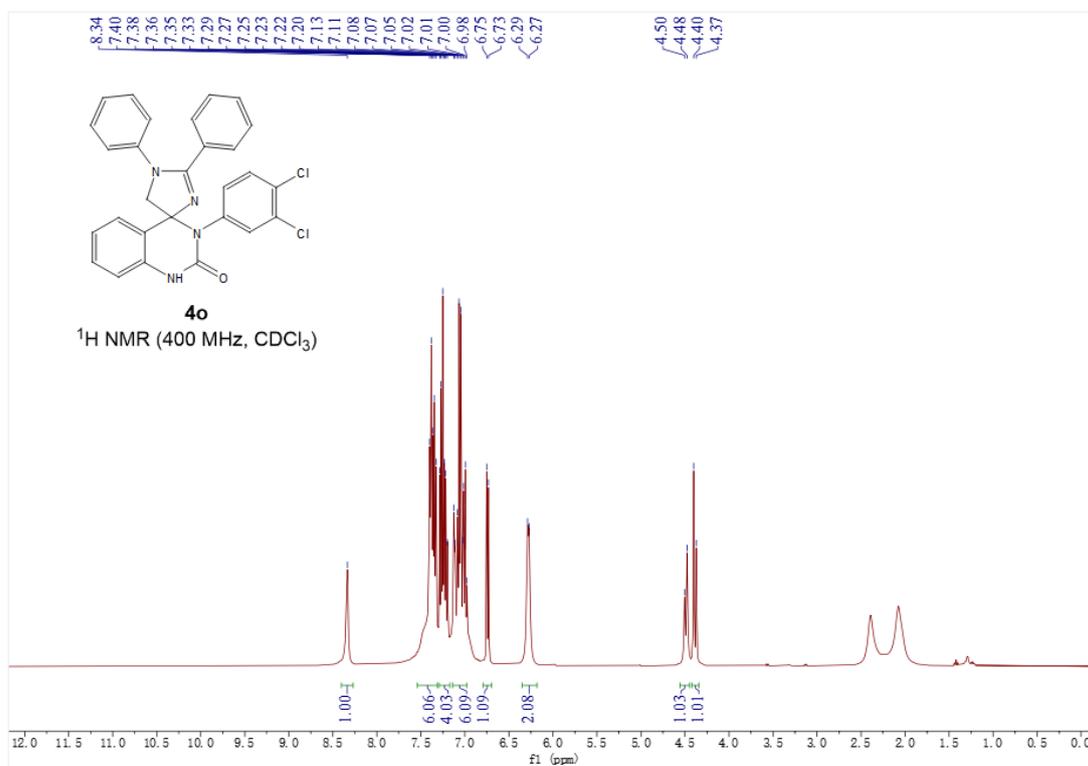
^1H NMR (400 MHz, CDCl_3) spectrum of **4n**



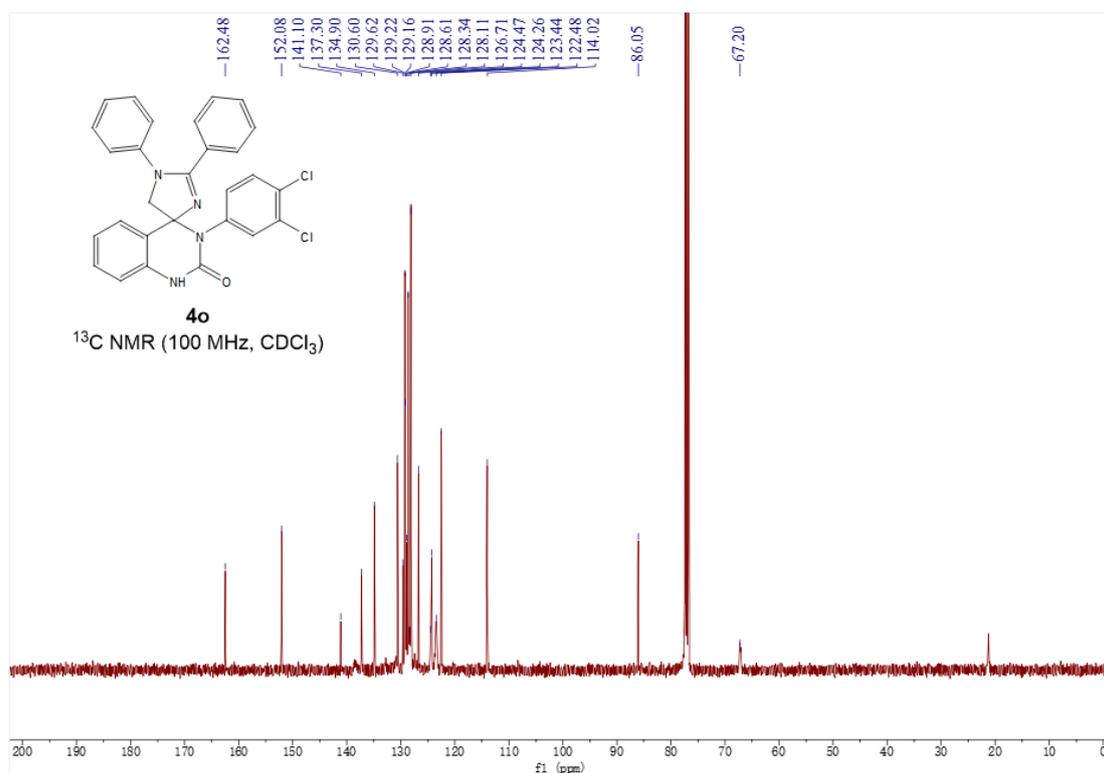
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4n**



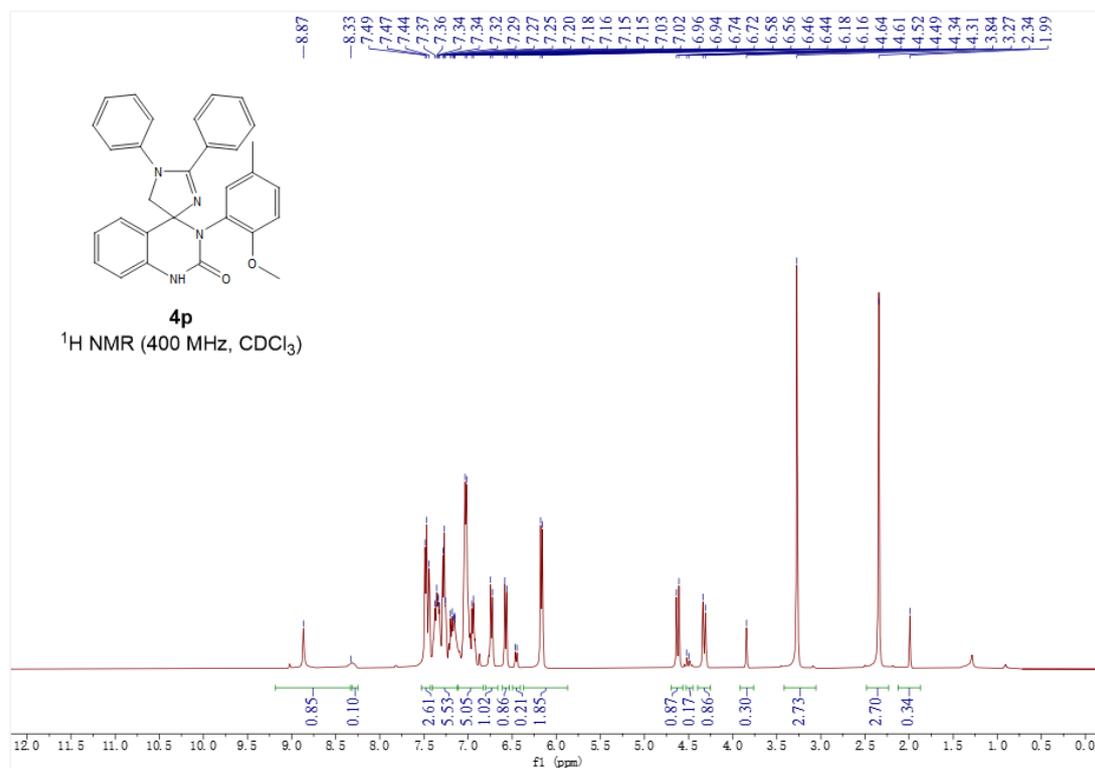
^1H NMR (400 MHz, CDCl_3) spectrum of **4o**



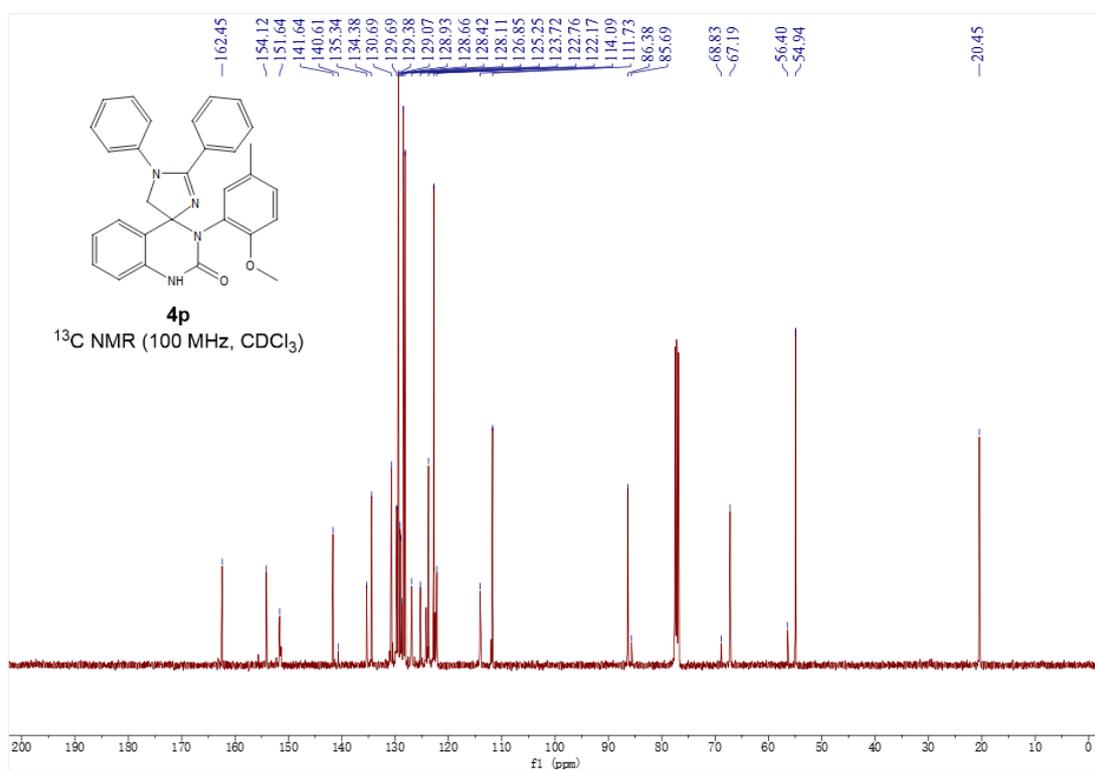
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4o**



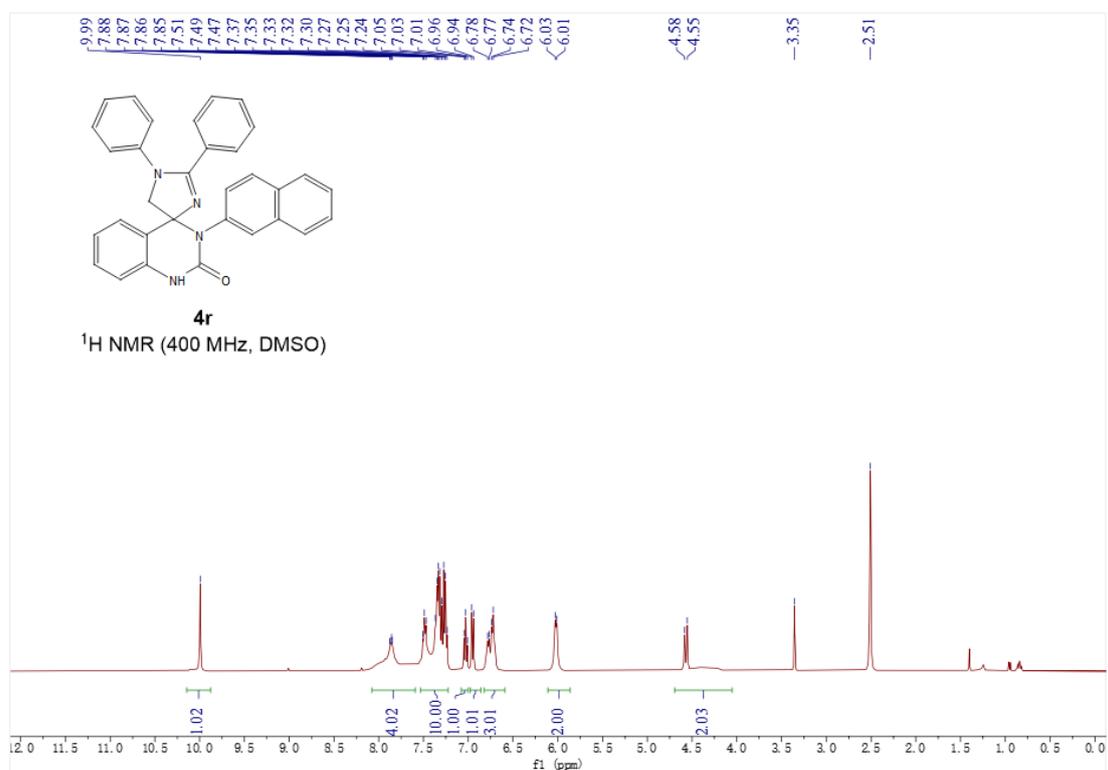
^1H NMR (400 MHz, CDCl_3) spectrum of **4p**



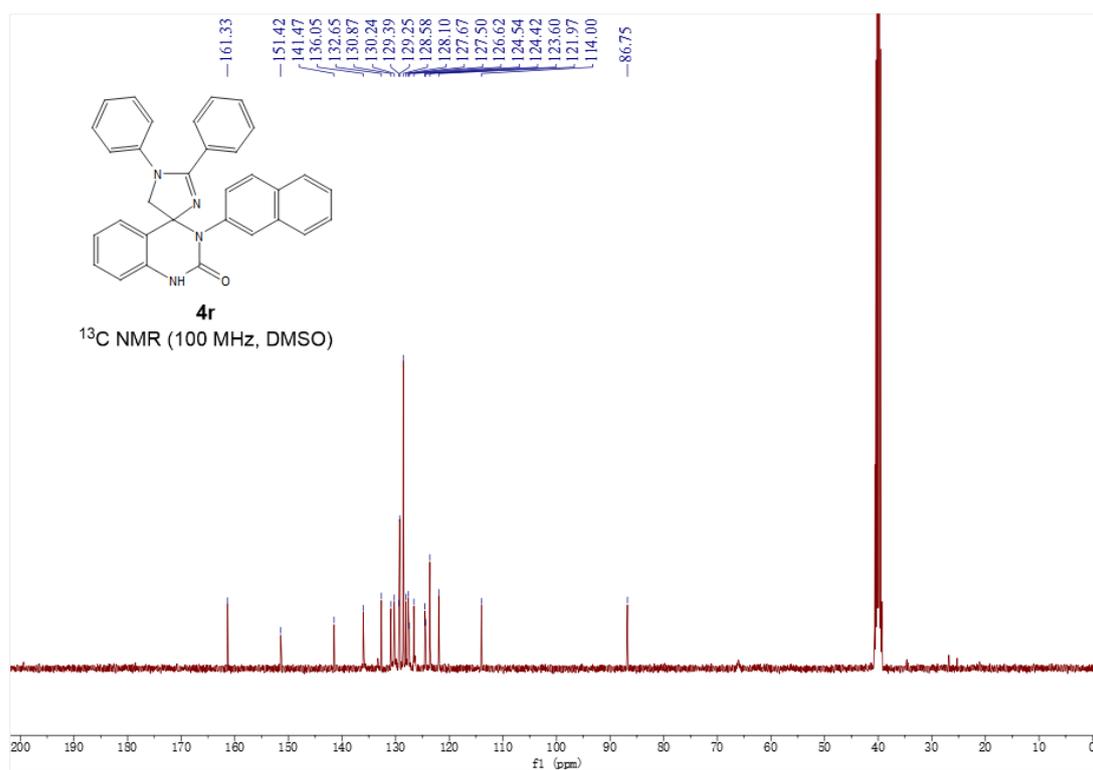
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4p**



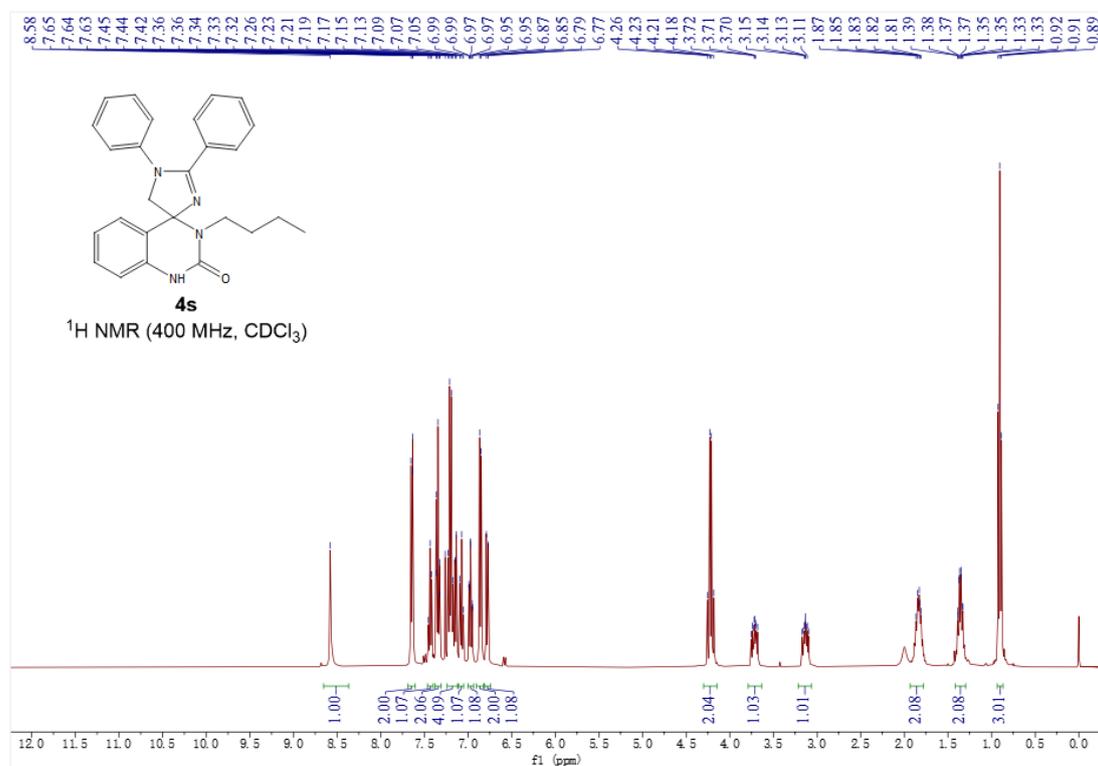
^1H NMR (400 MHz, DMSO) spectrum of **4r**



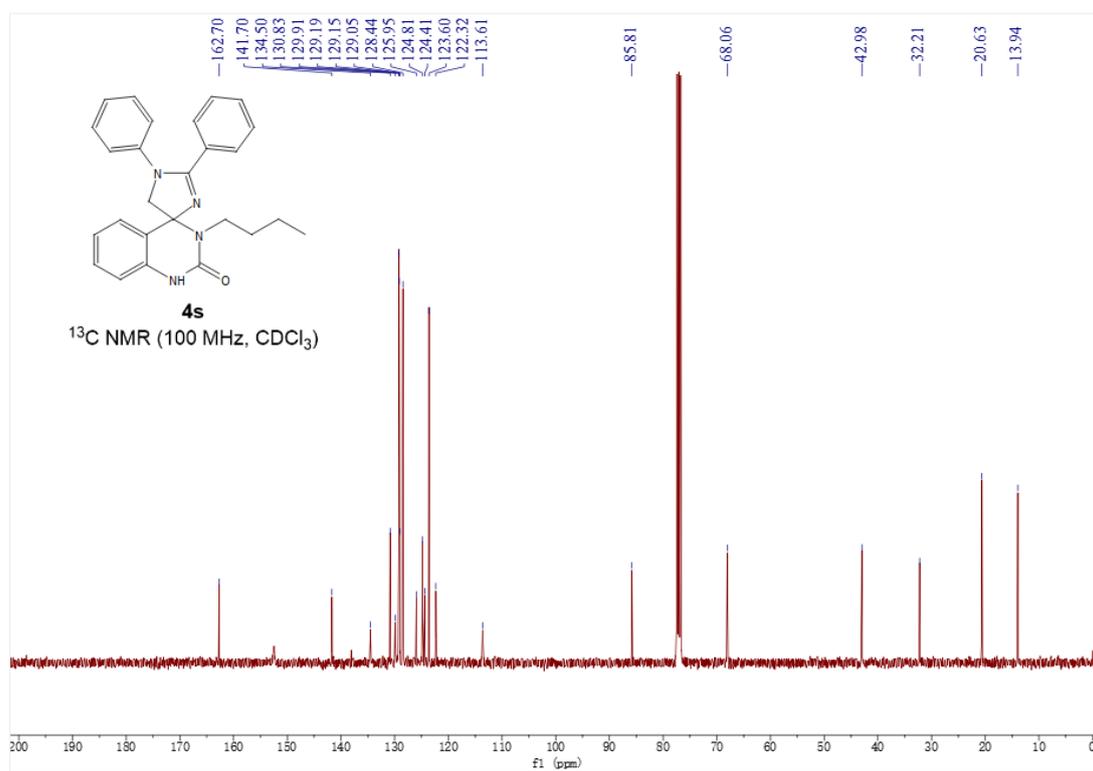
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO) spectrum of **4r**



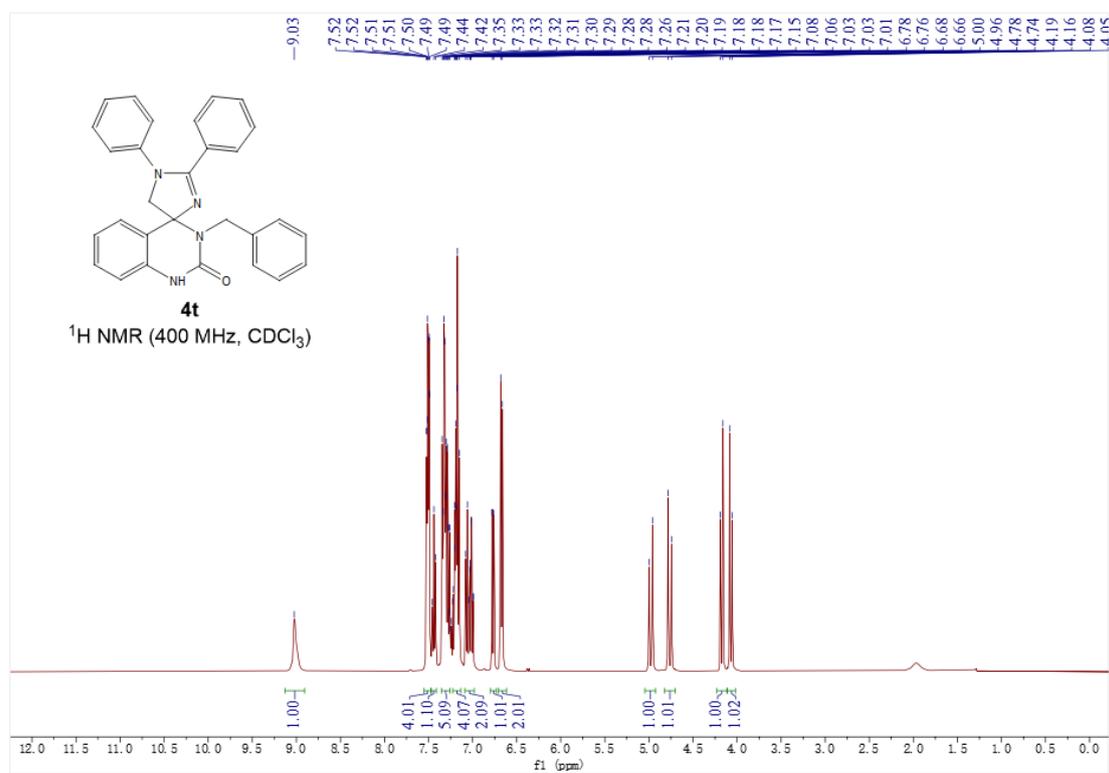
^1H NMR (400 MHz, CDCl_3) spectrum of **4s**



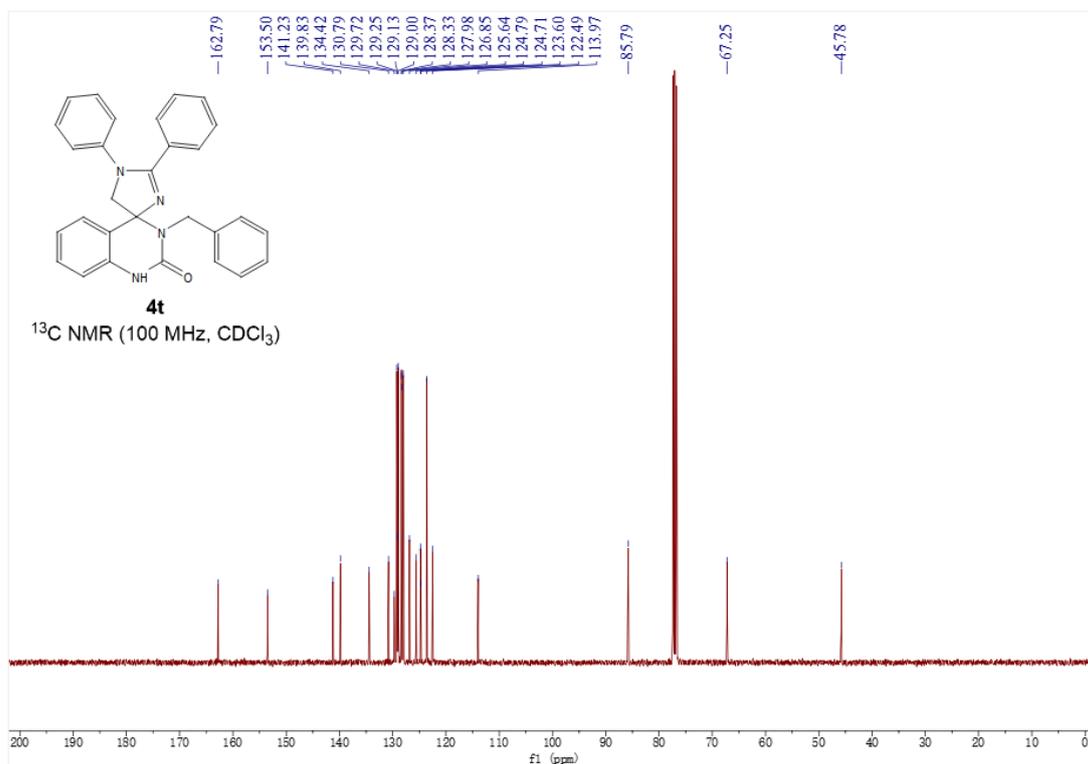
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4s**



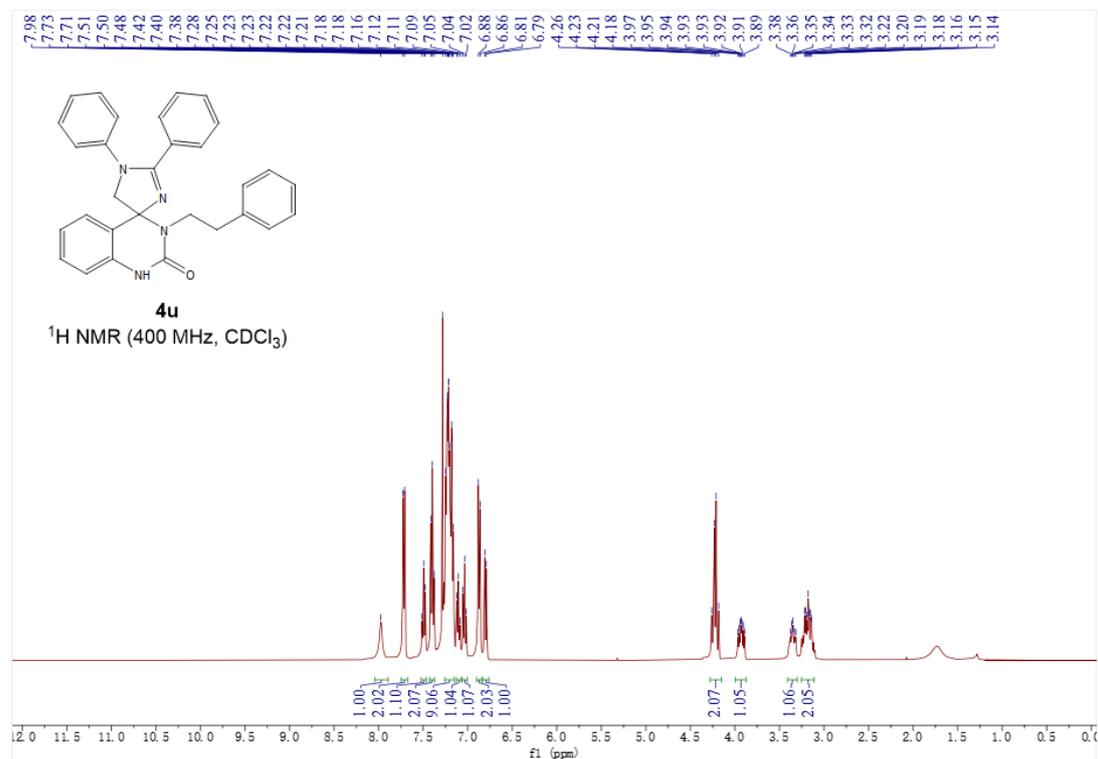
^1H NMR (400 MHz, CDCl_3) spectrum of **4t**



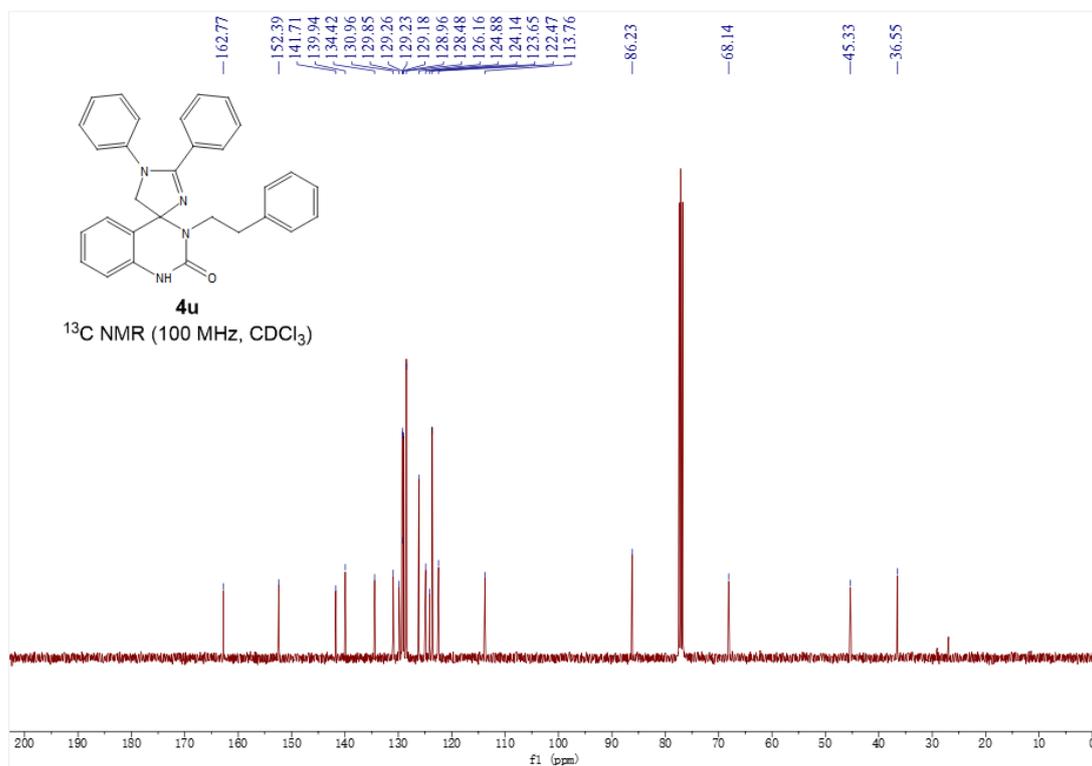
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4t**



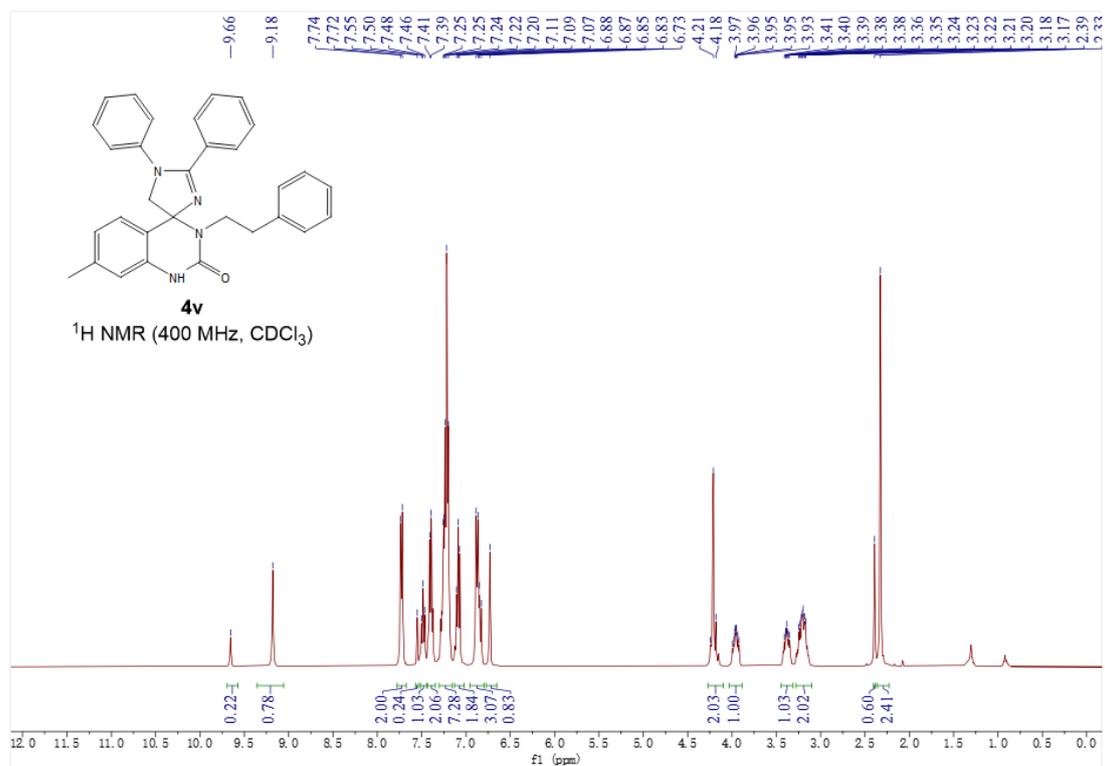
^1H NMR (400 MHz, CDCl_3) spectrum of **4u**



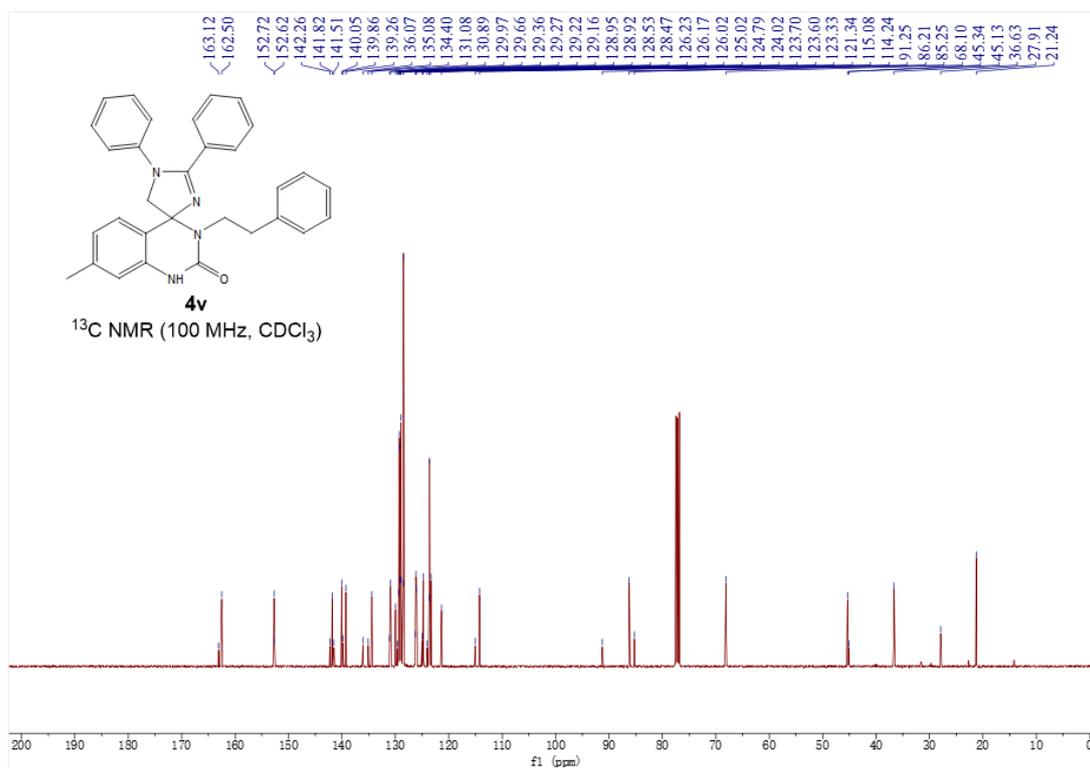
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4u**



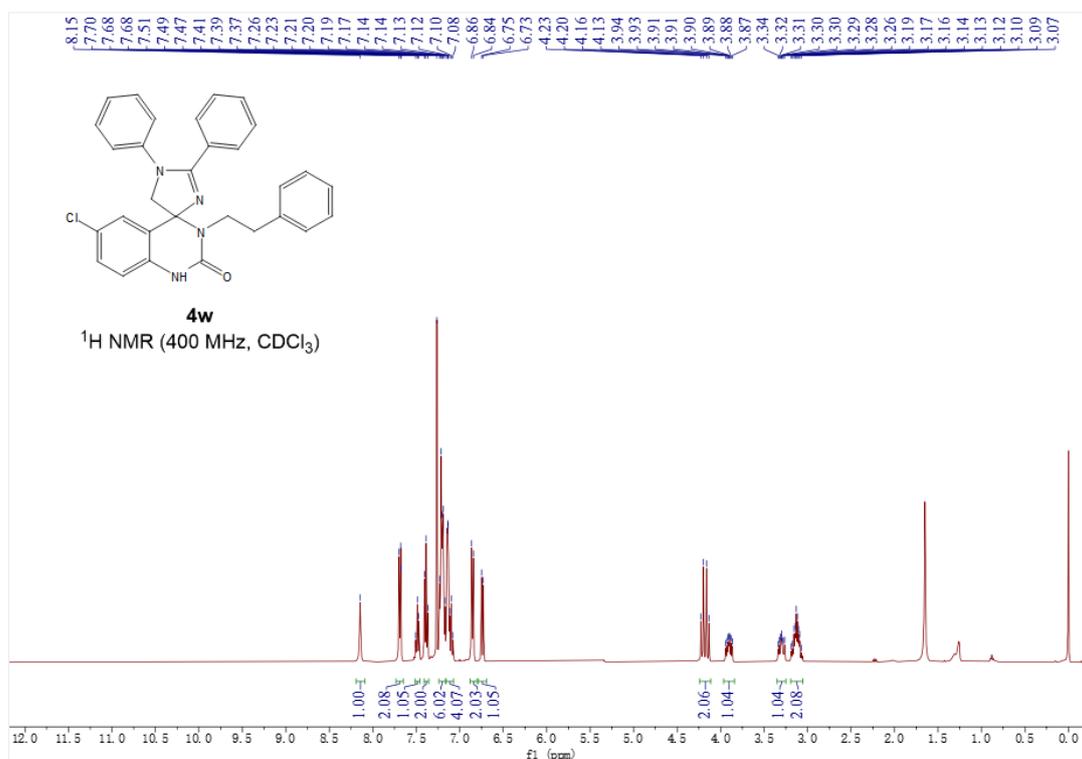
^1H NMR (400 MHz, CDCl_3) spectrum of **4v**



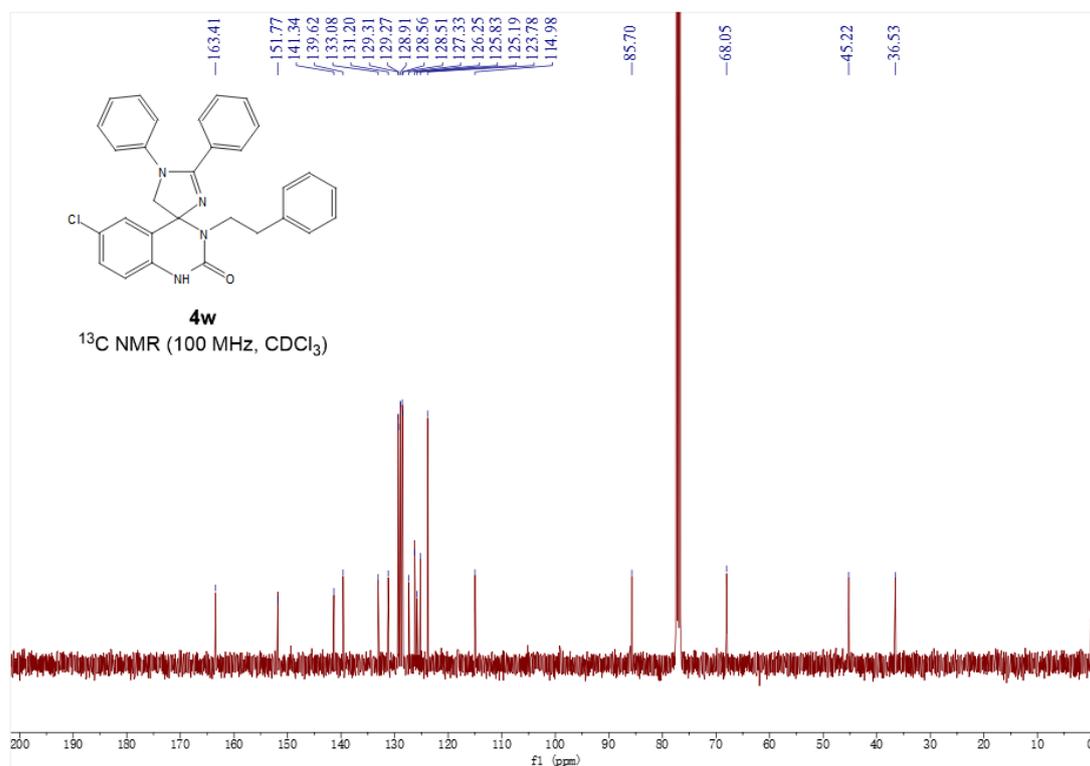
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4v**



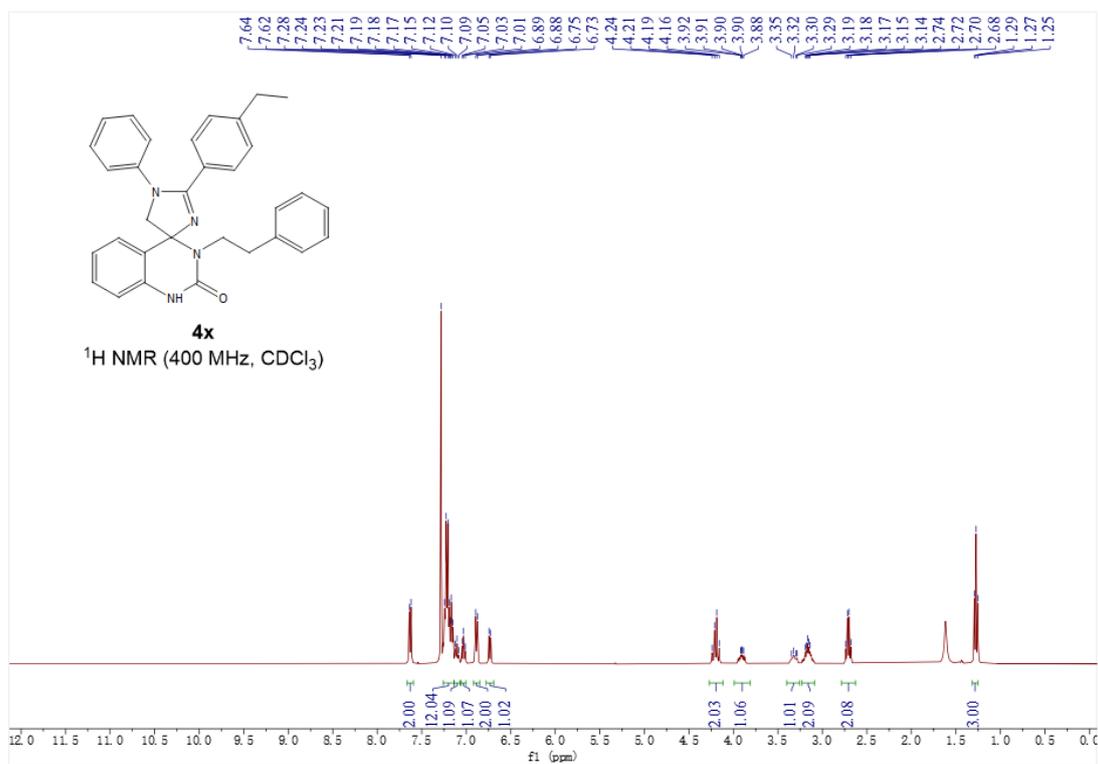
^1H NMR (400 MHz, CDCl_3) spectrum of **4w**



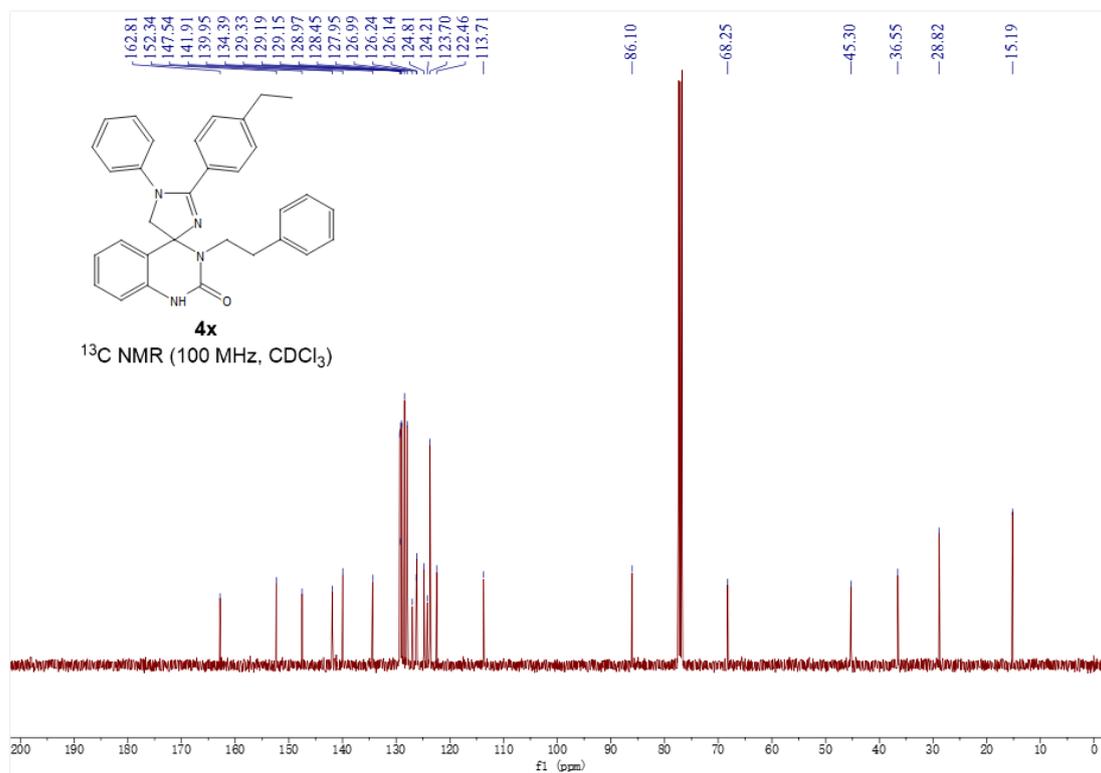
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4w**



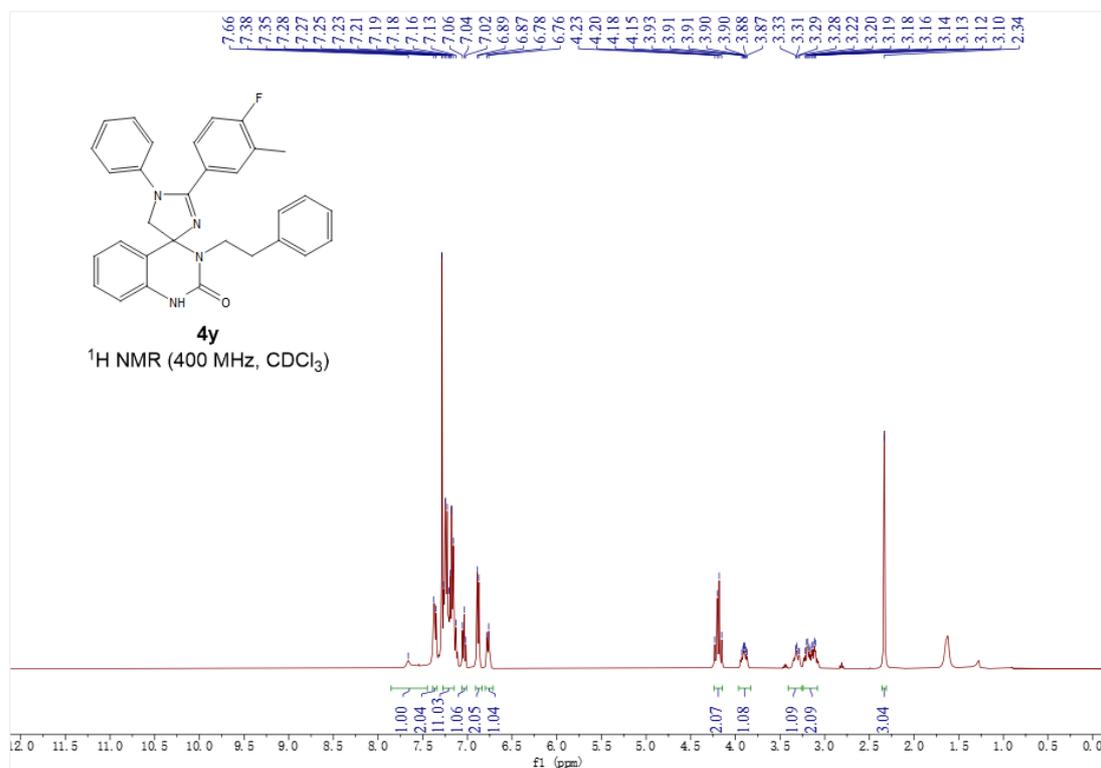
^1H NMR (400 MHz, CDCl_3) spectrum of **4x**



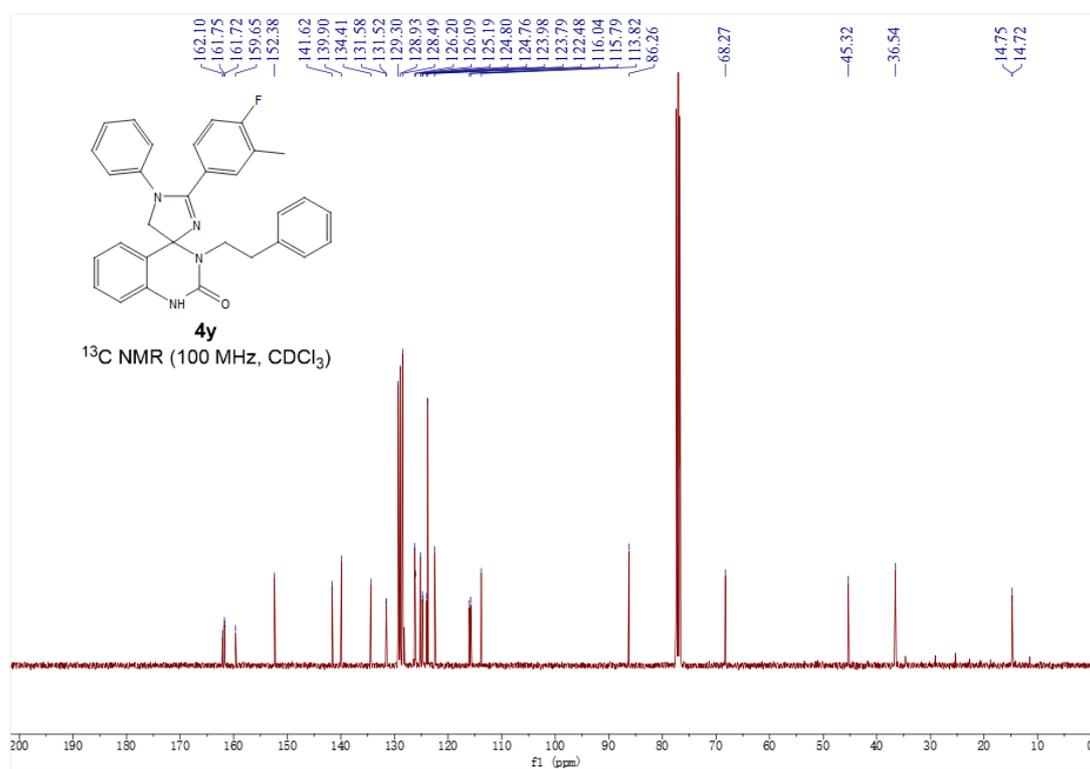
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4x**



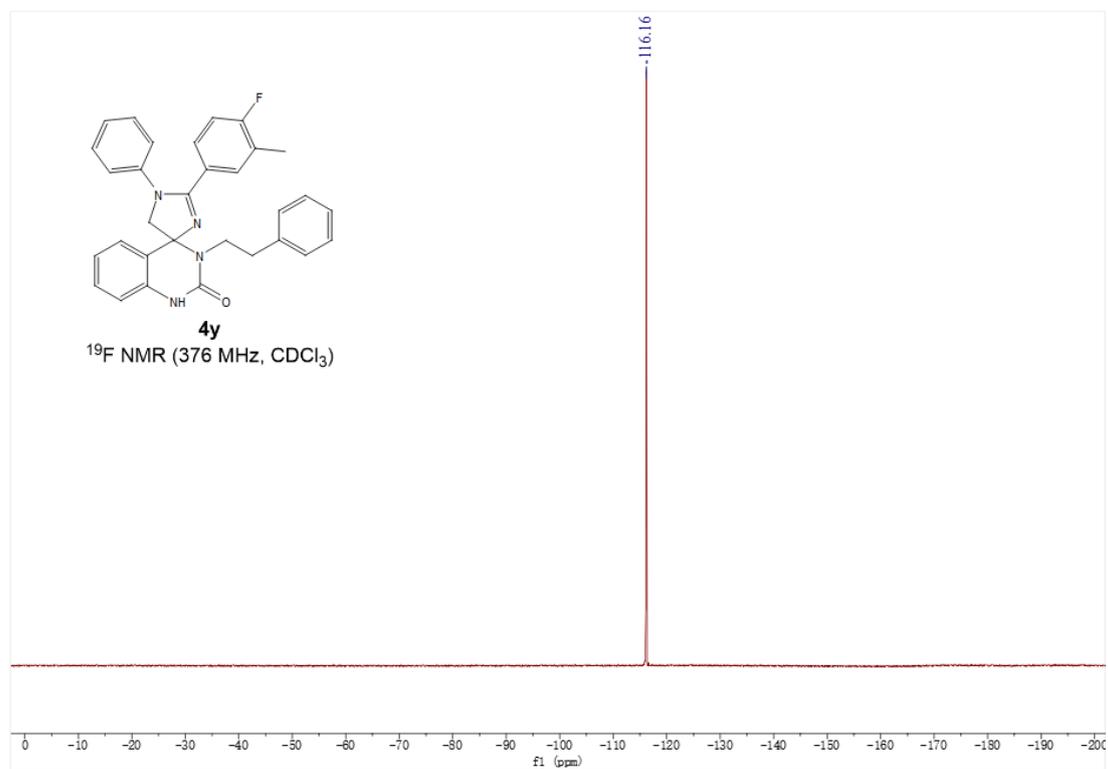
^1H NMR (400 MHz, CDCl_3) spectrum of **4y**



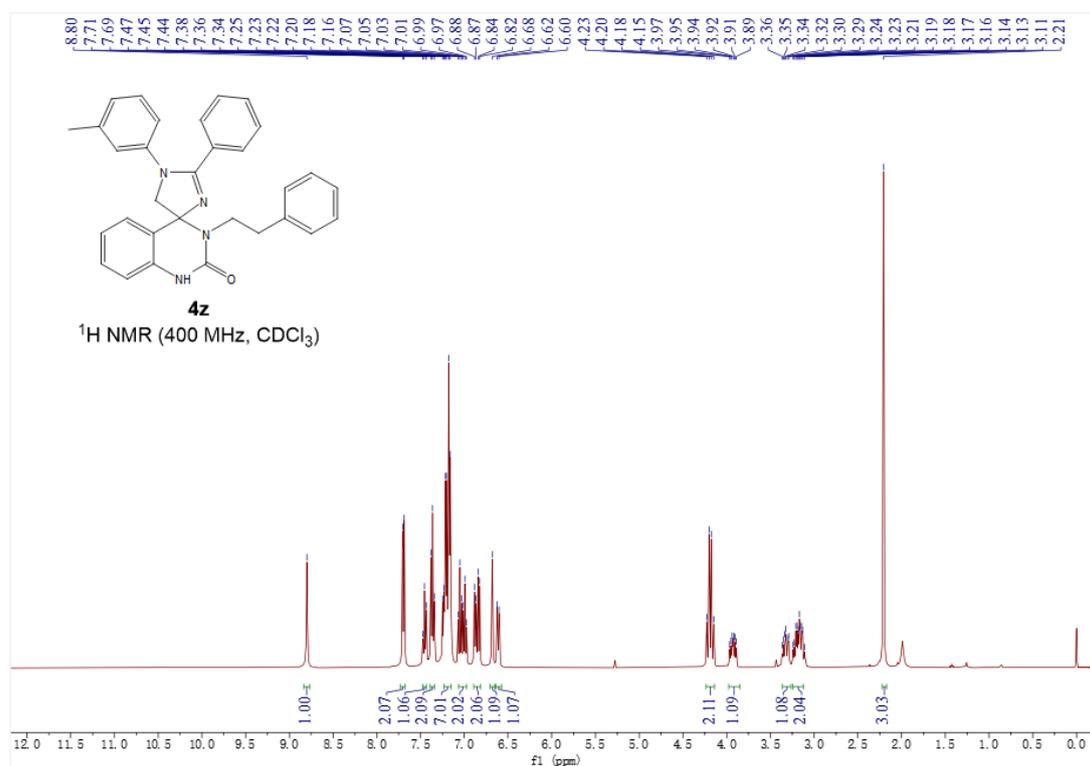
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4y**



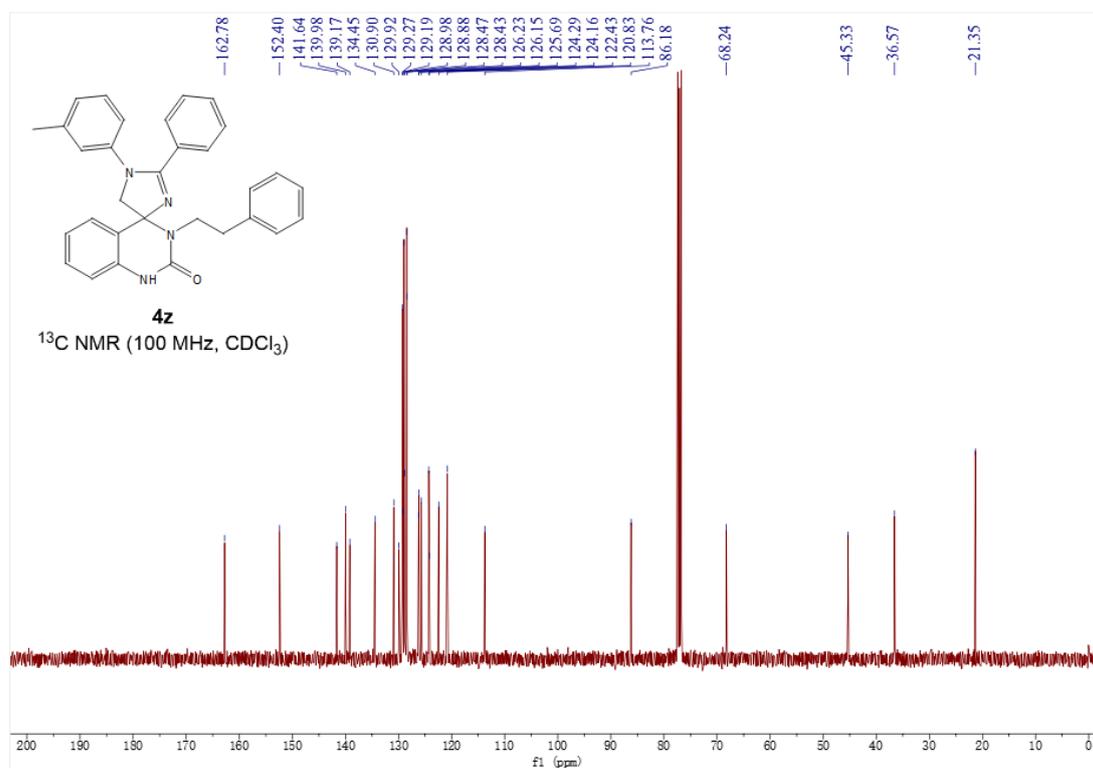
$^{19}\text{F}\{^1\text{H}\}$ NMR (376 MHz, CDCl_3) spectrum of **4y**



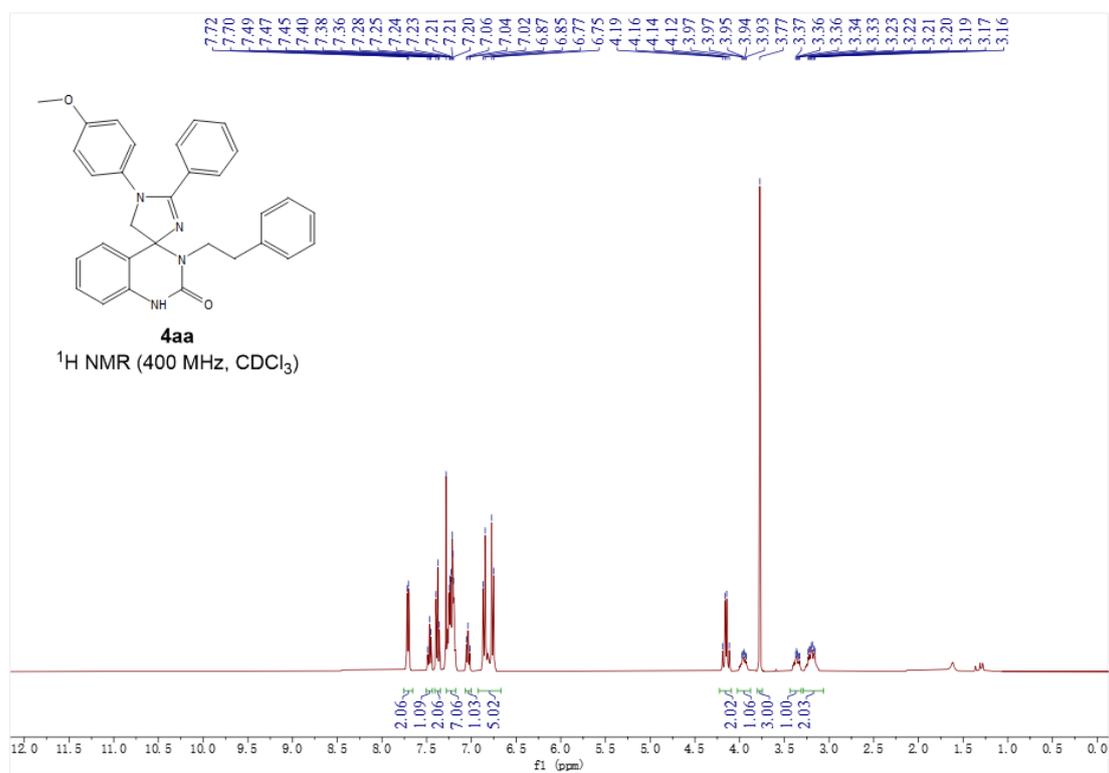
^1H NMR (400 MHz, CDCl_3) spectrum of **4z**



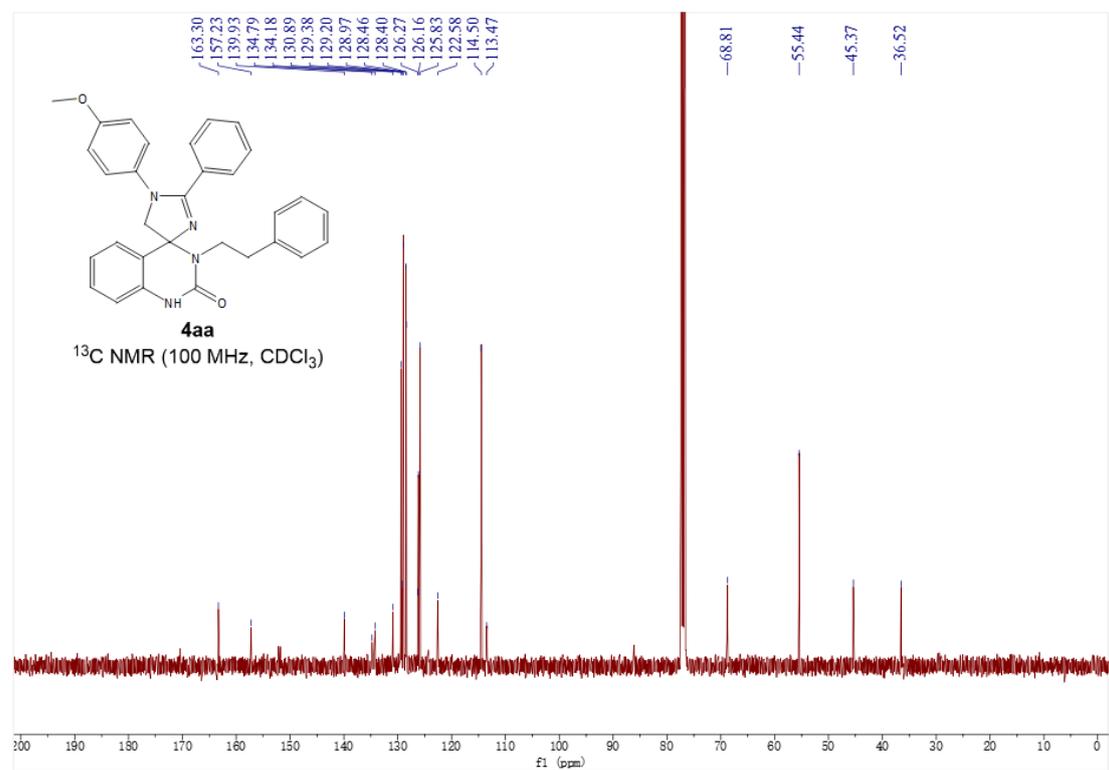
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4z**



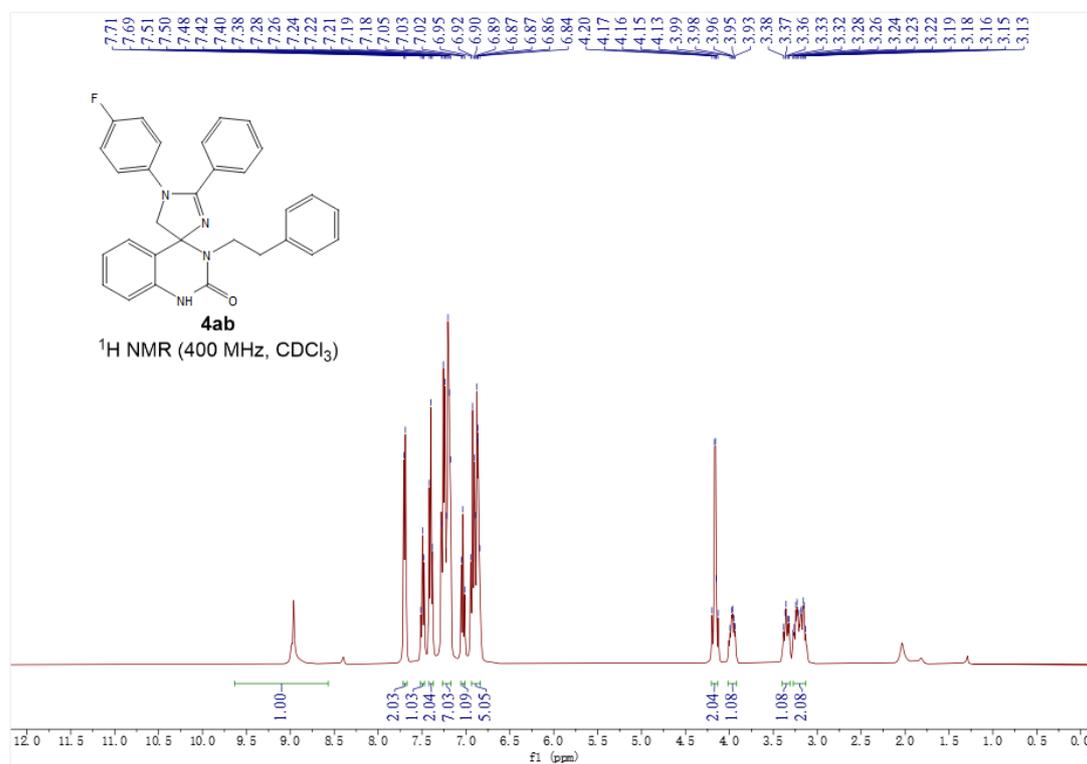
^1H NMR (400 MHz, CDCl_3) spectrum of **4aa**



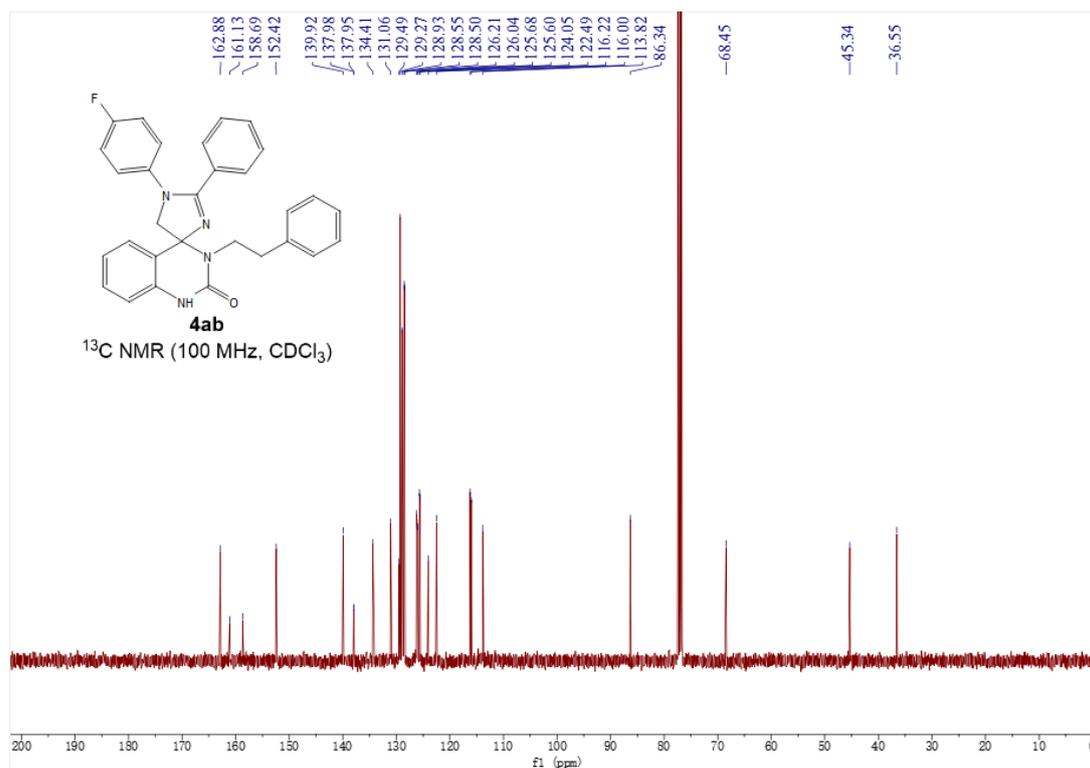
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4aa**



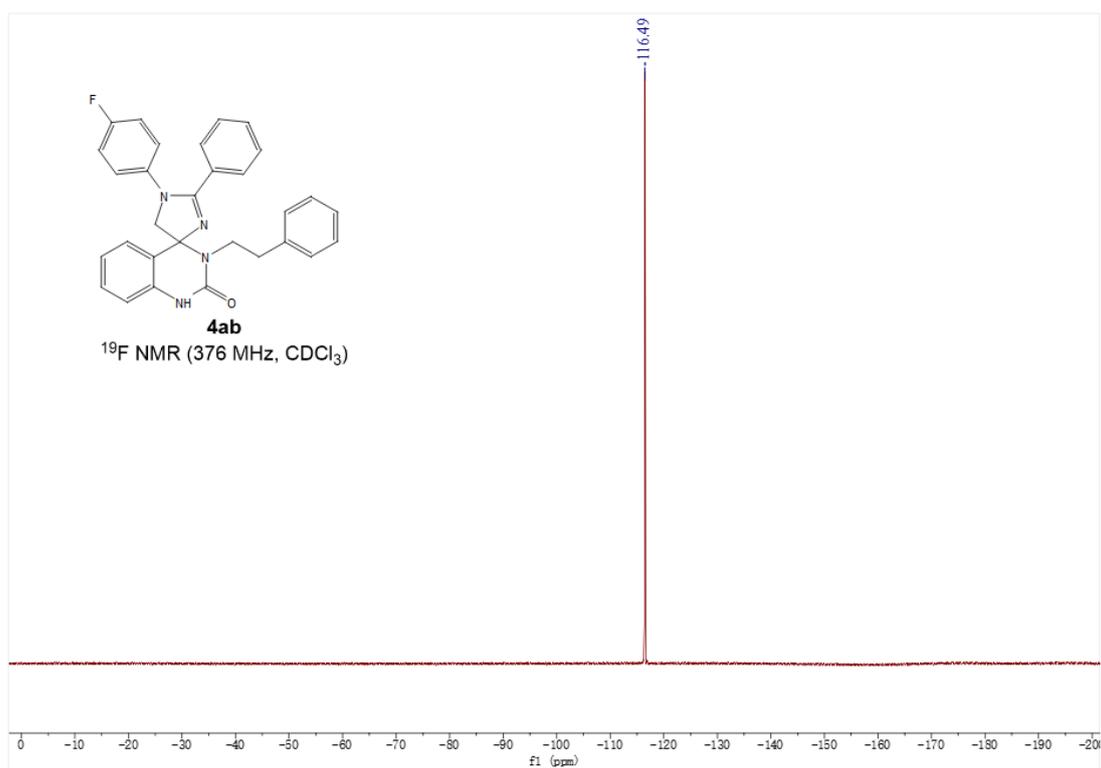
^1H NMR (400 MHz, CDCl_3) spectrum of **4ab**



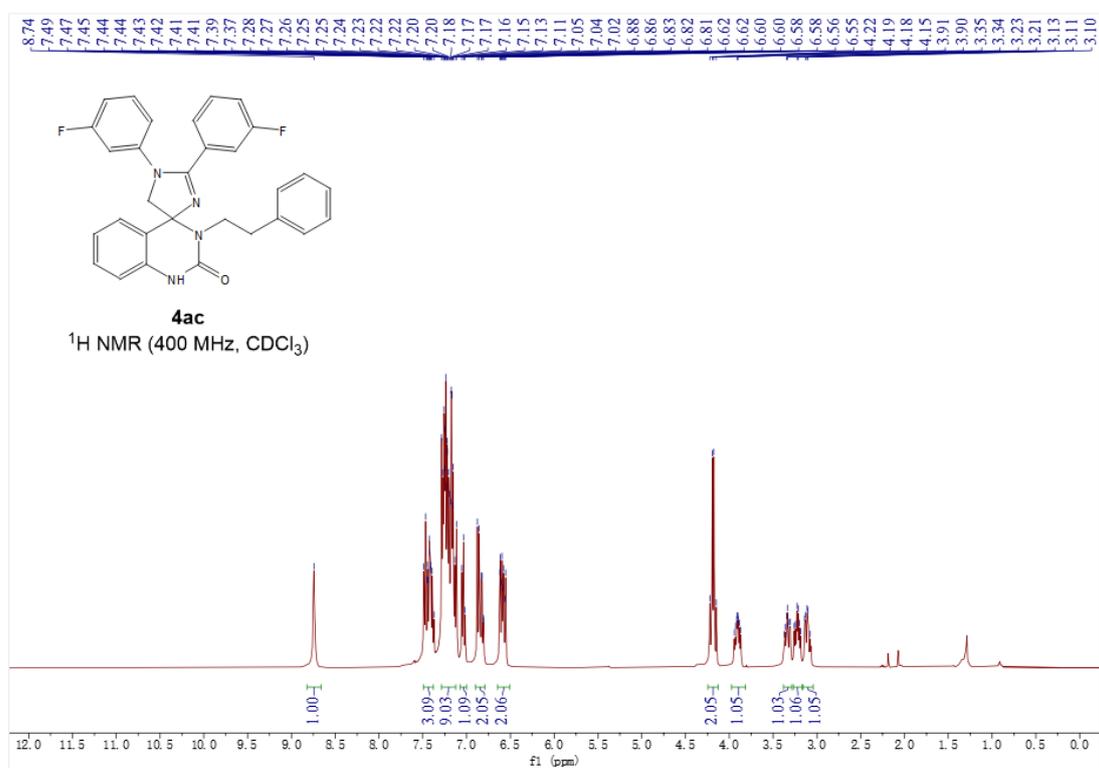
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4ab**



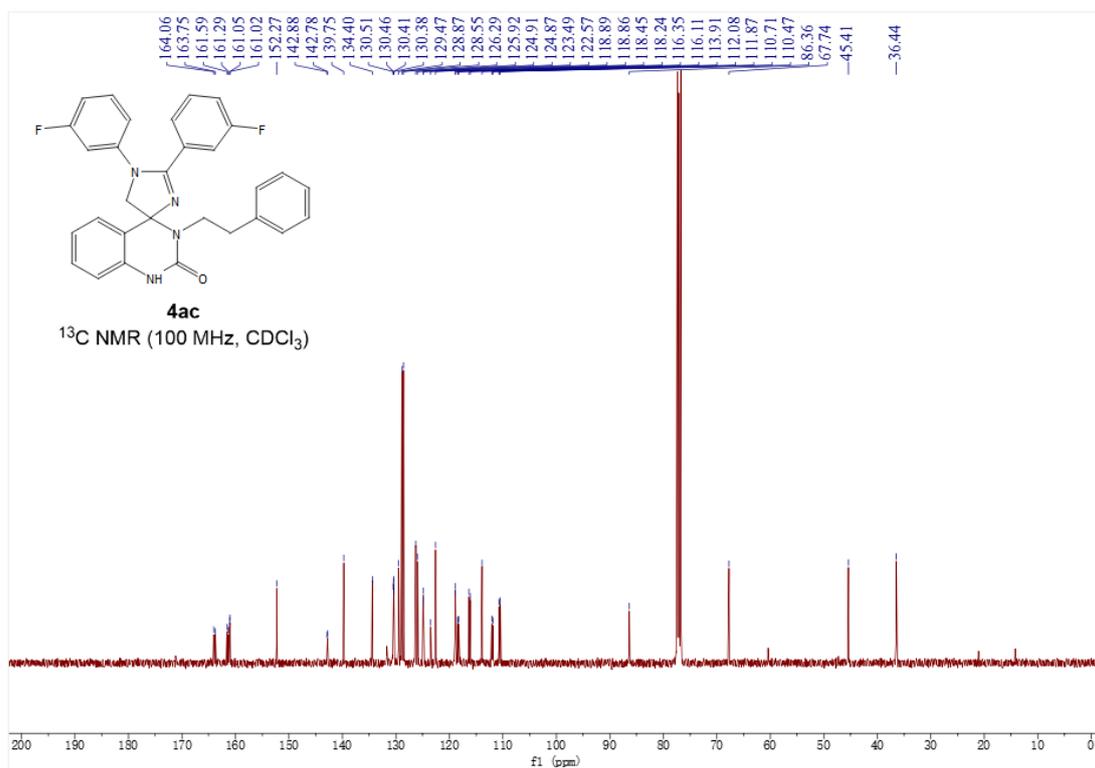
^{19}F $\{^1\text{H}\}$ NMR (376 MHz, CDCl_3) spectrum of **4ab**



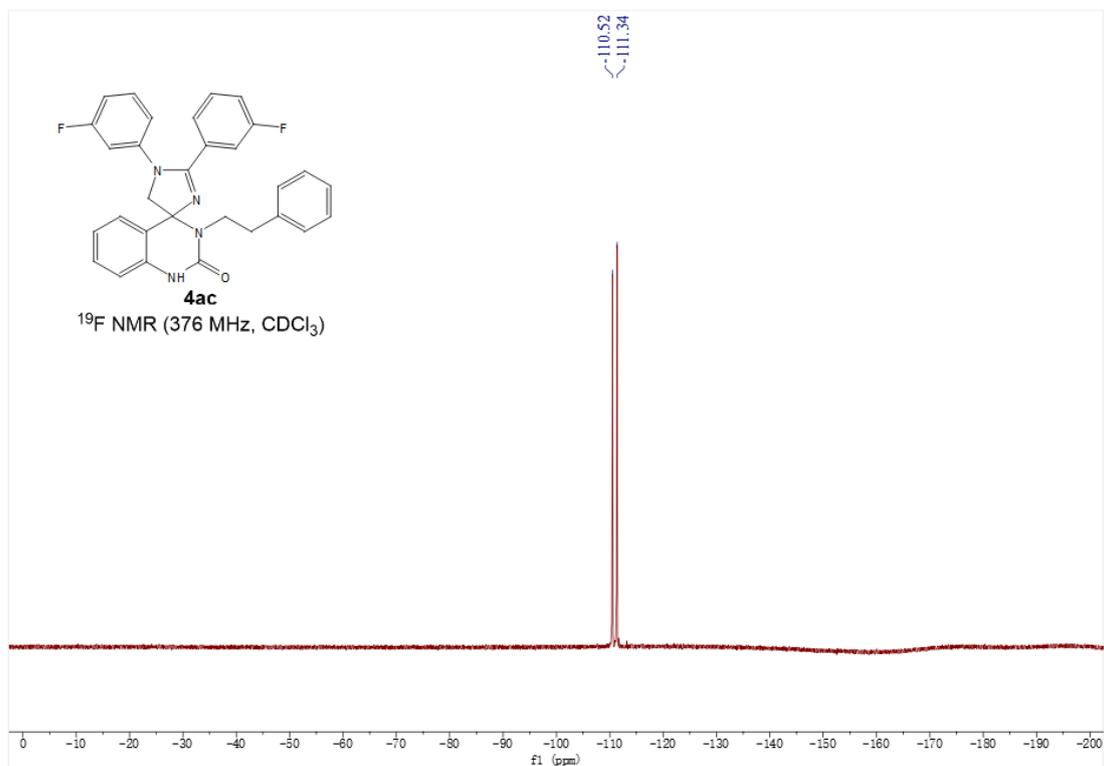
^1H NMR (400 MHz, CDCl_3) spectrum of **4ac**



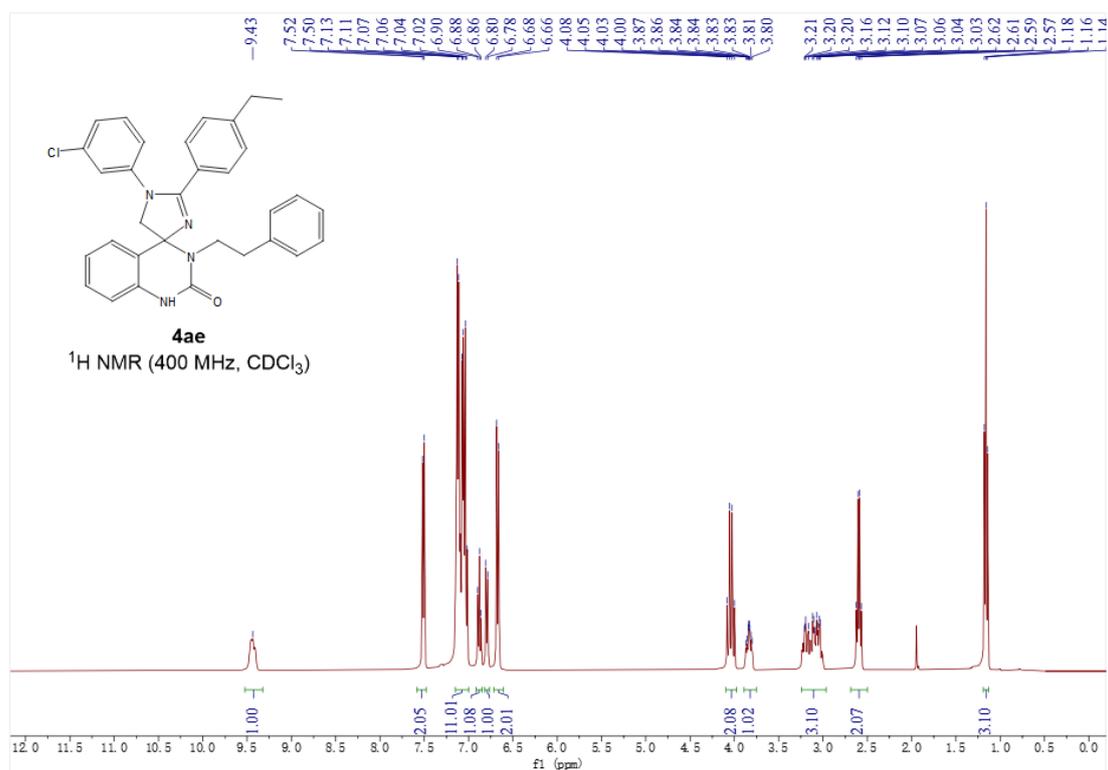
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4ac**



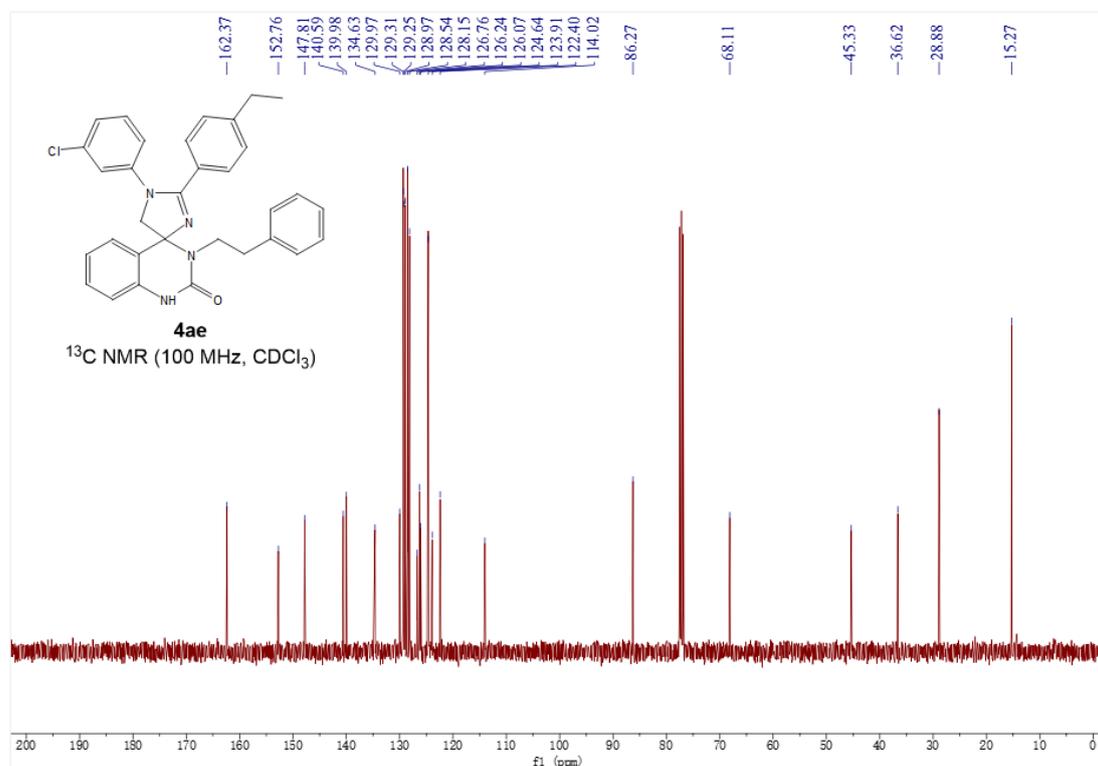
$^{19}\text{F}\{^1\text{H}\}$ NMR (376 MHz, CDCl_3) spectrum of **4ac**



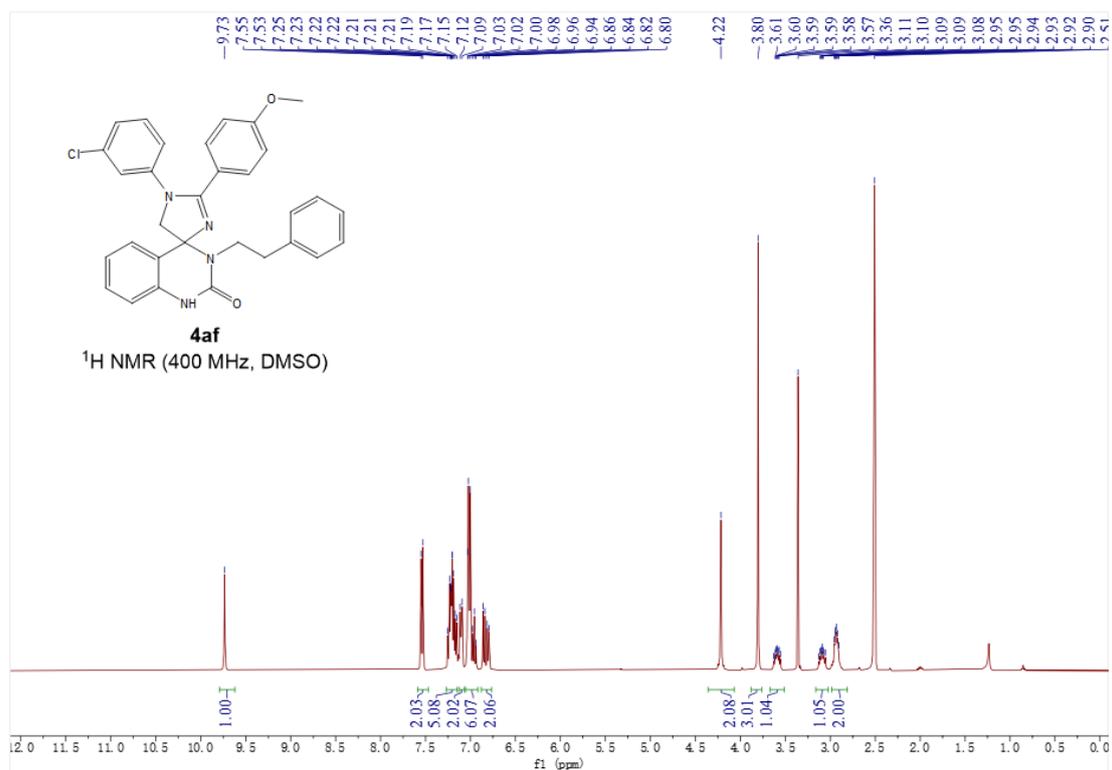
^1H NMR (400 MHz, CDCl_3) spectrum of **4ae**



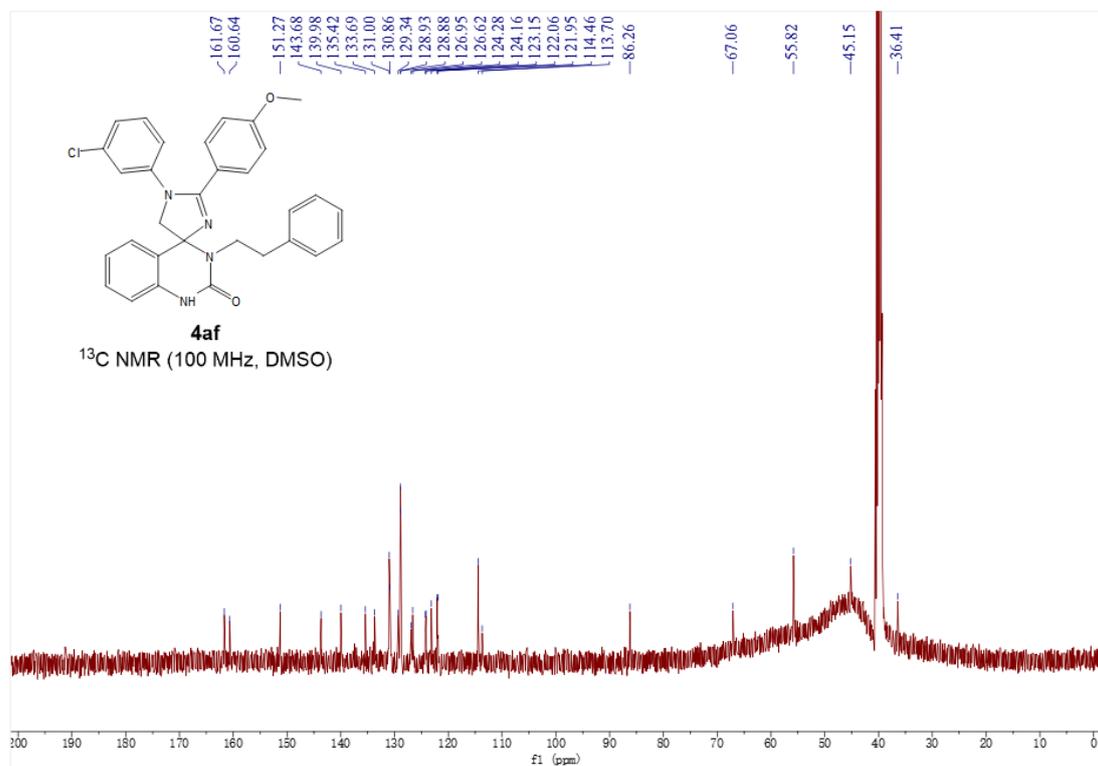
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4ae**



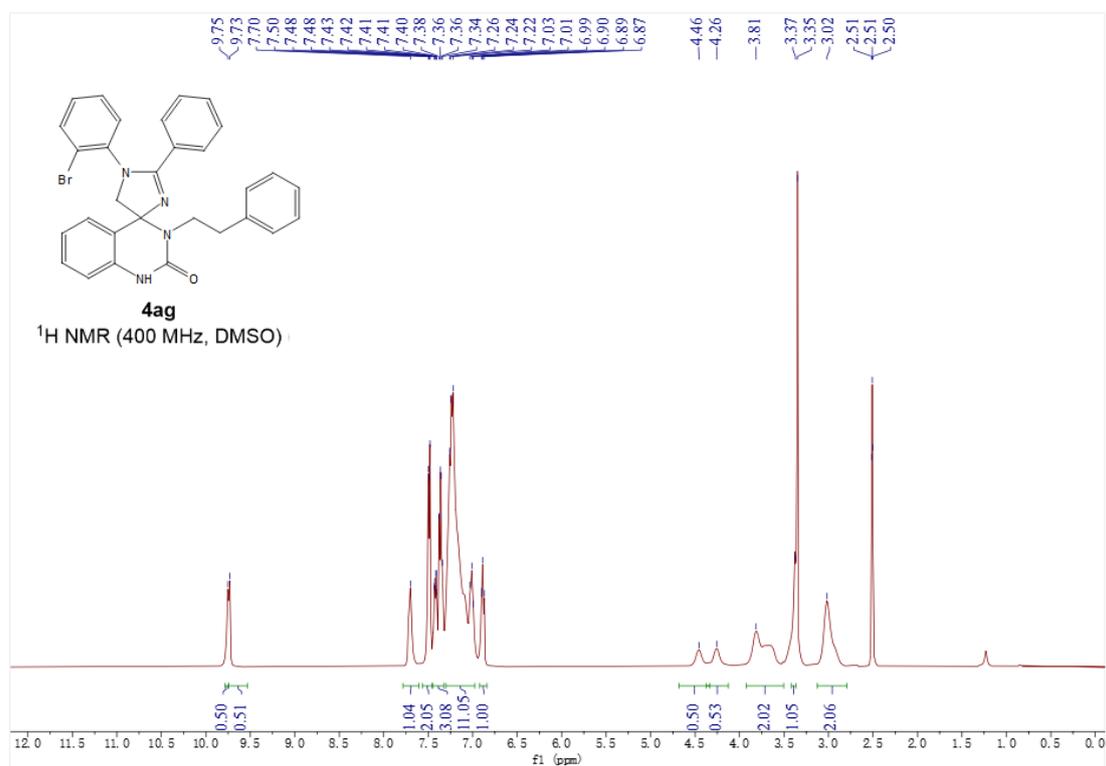
^1H NMR (400 MHz, DMSO) spectrum of **4af**



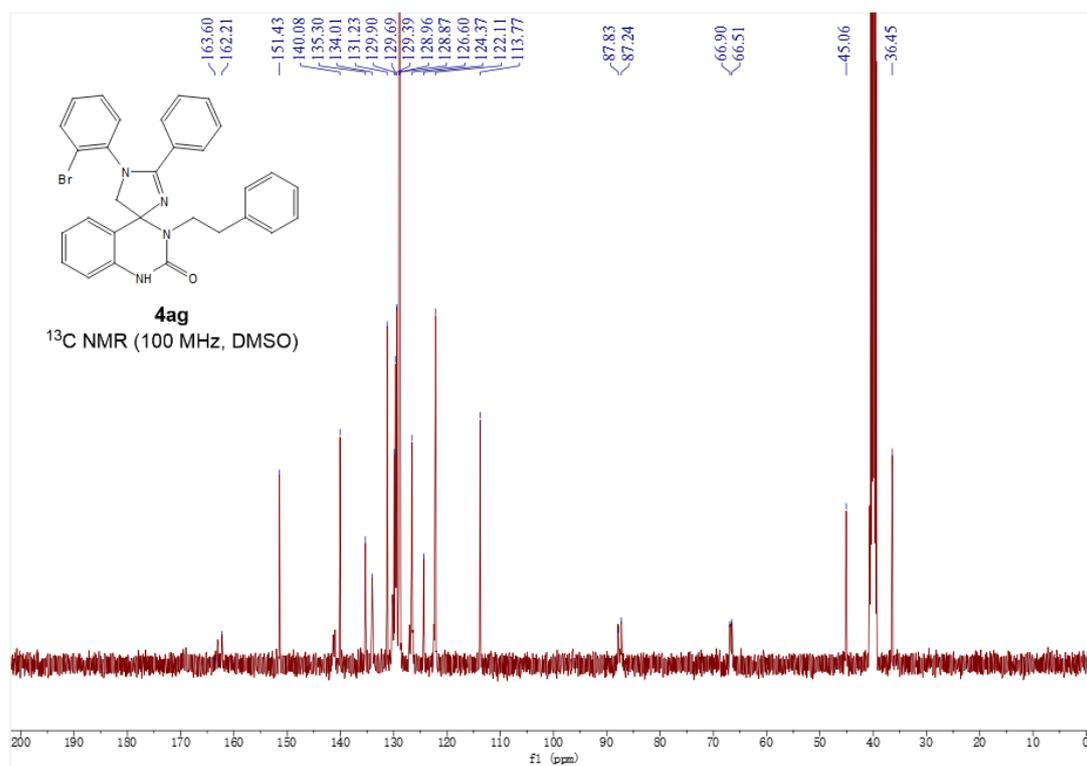
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO) spectrum of **4af**



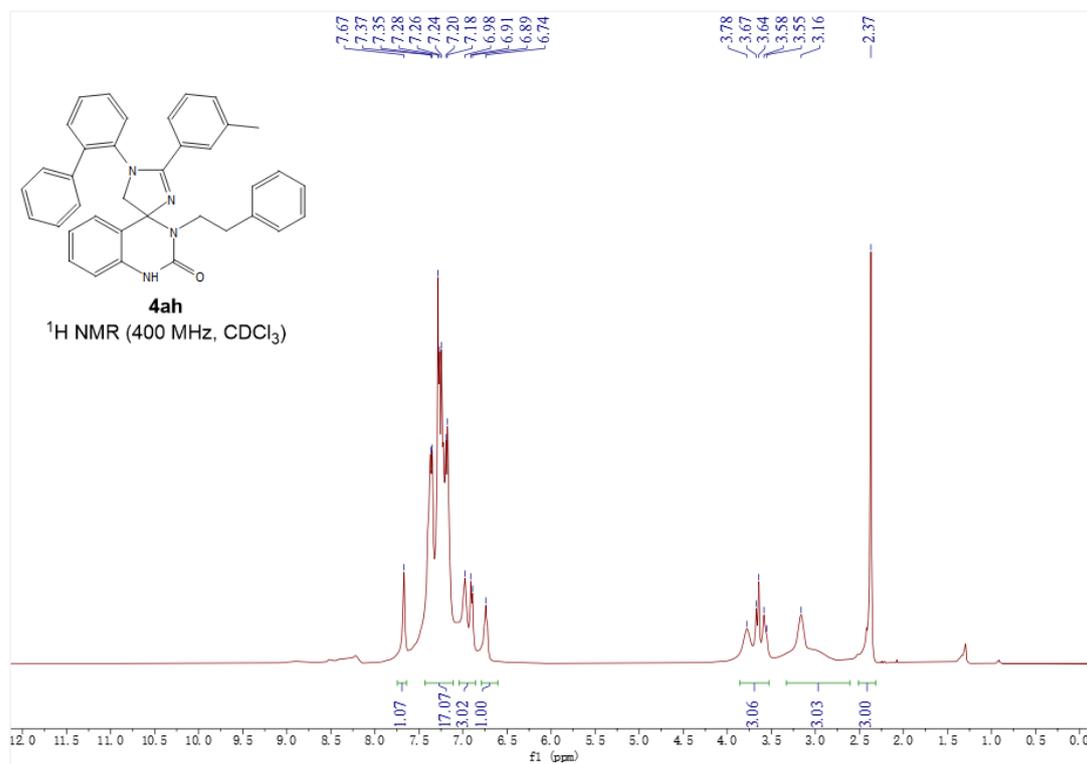
^1H NMR (400 MHz, DMSO) spectrum of **4ag**



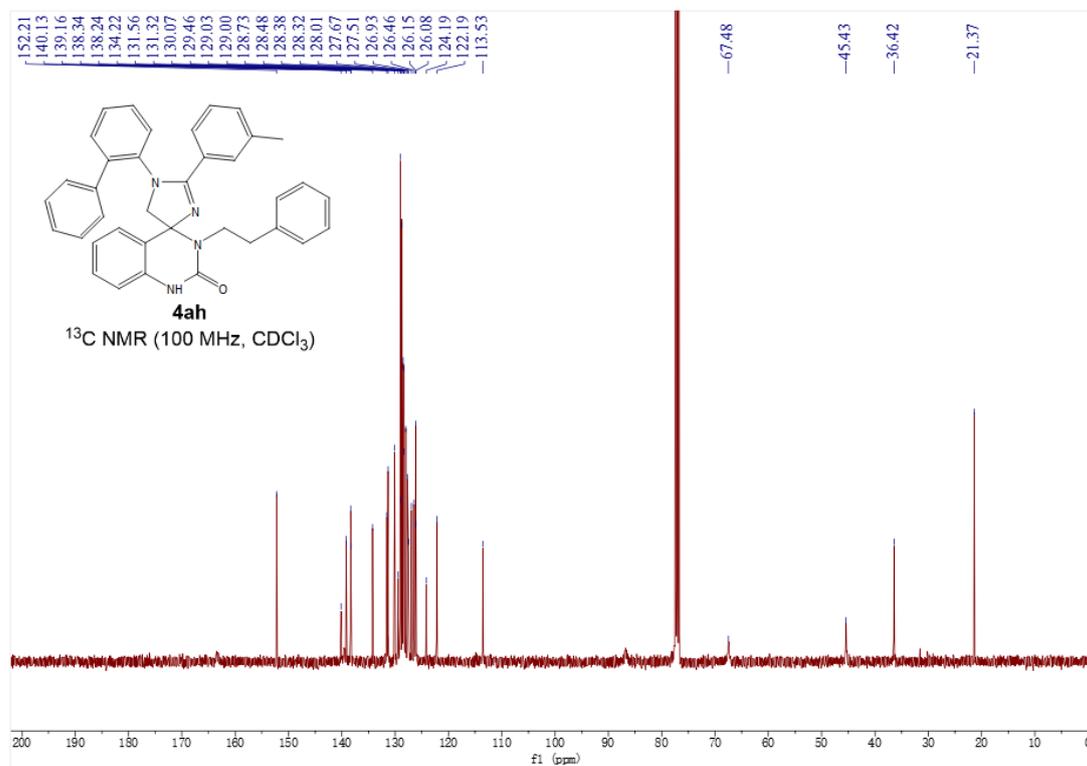
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO) spectrum of **4ag**



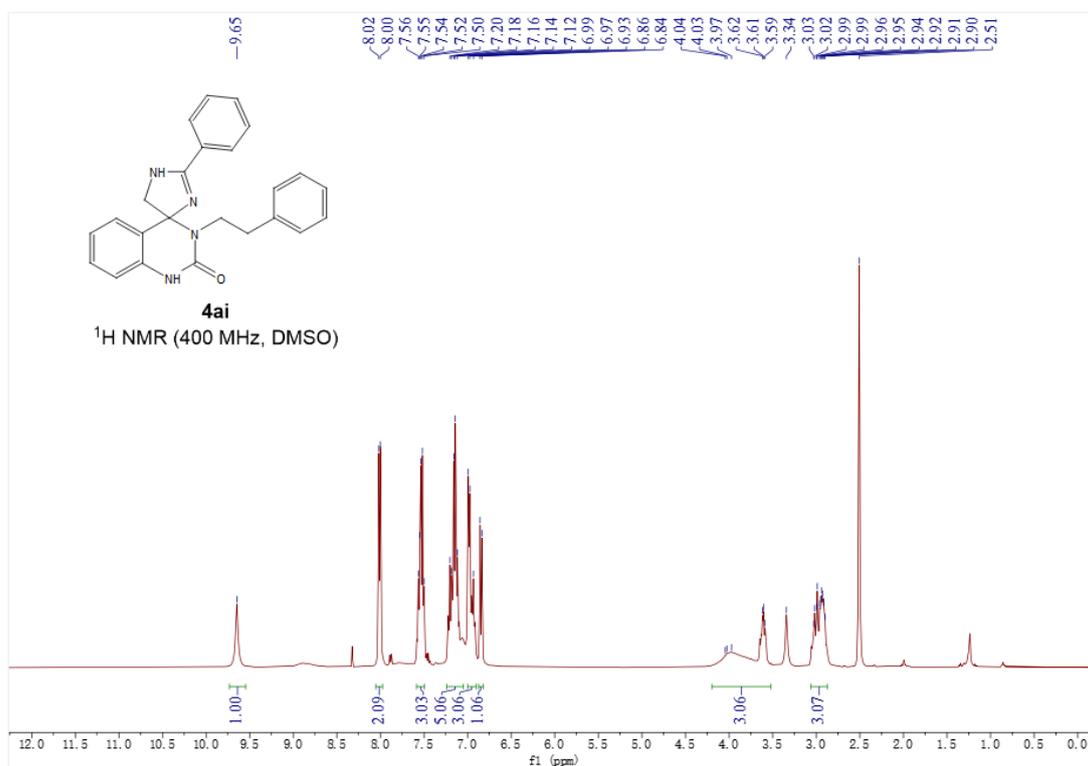
^1H NMR (400 MHz, CDCl_3) spectrum of **4ah**



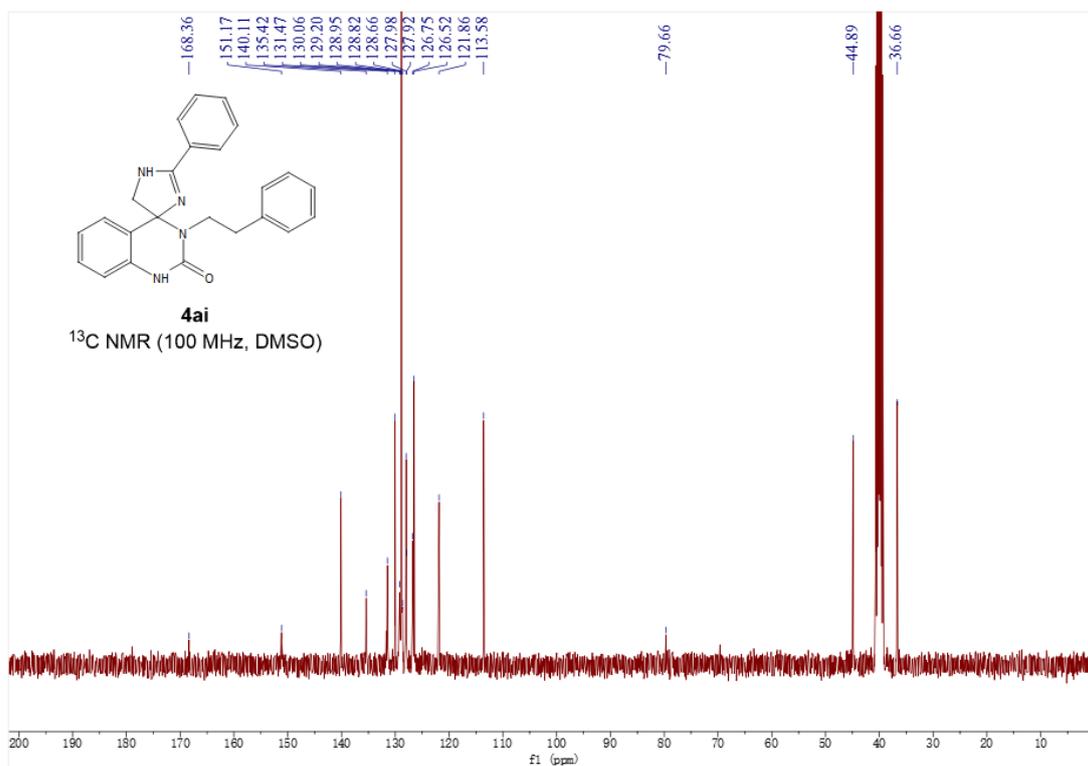
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3) spectrum of **4ah**



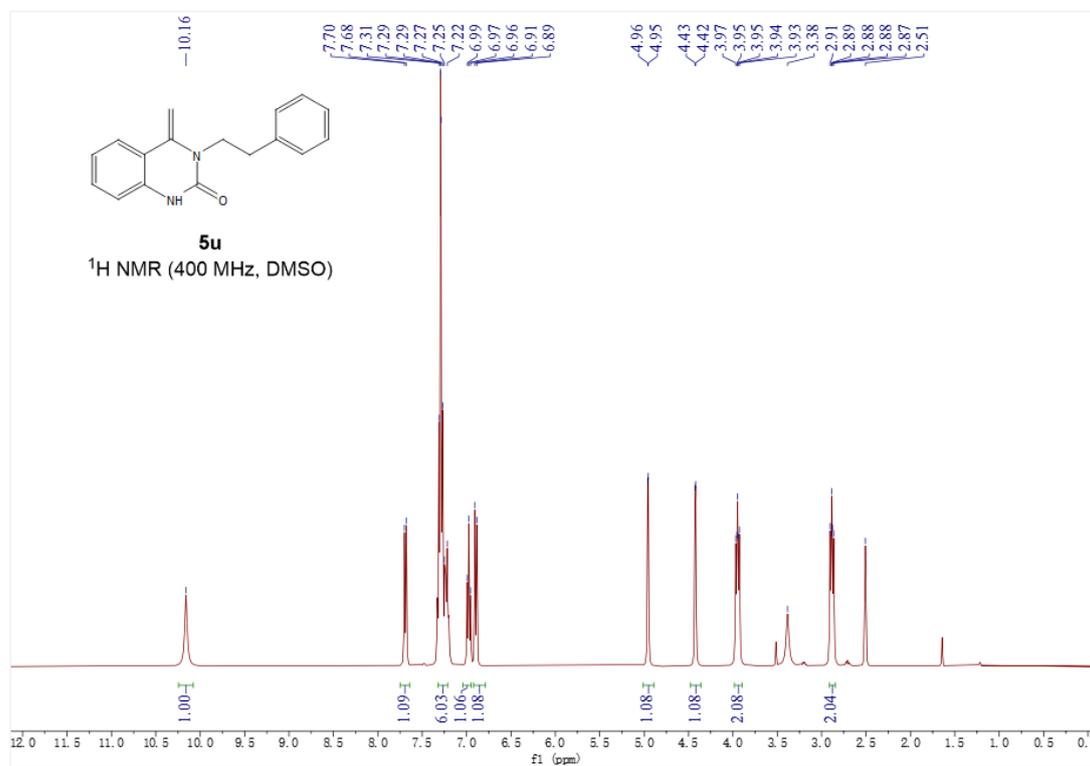
^1H NMR (400 MHz, DMSO) spectrum of **4ai**



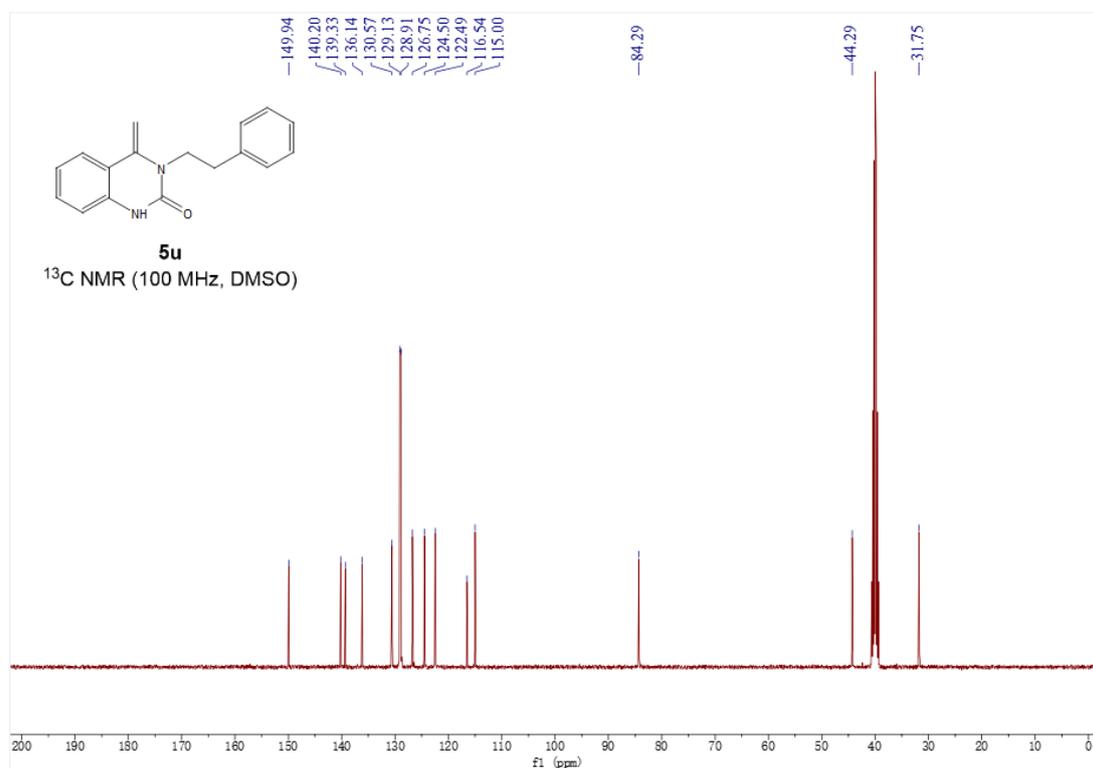
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO) spectrum of **4ai**



^1H NMR (400 MHz, DMSO) spectrum of **5u**



$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, DMSO) spectrum of **5u**

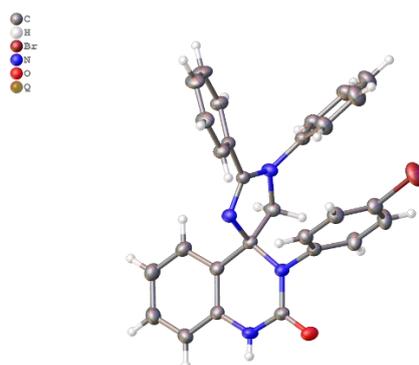
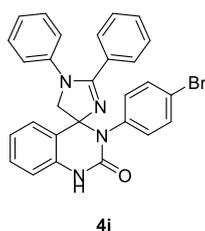


6. Crystal Data of 4i

The crystals suitable for single crystal X-ray diffraction were obtained by slow evaporating the chloroform and methanol solution of **4i** at room temperature in open air for about five days.

For X-ray structure analyses, the crystals were mounted onto the tip of glass fibers, and diffraction data were collected on a Bruker Smart Apex II CCD diffractometer with graphite-monochromated Mo K α ($\lambda = 0.71073 \text{ \AA}$). An empirical (multi-scan) absorption correction was applied with the program SADABS. The structures were solved by direct method and subsequently refined on F2 by using full-matrix least-squares techniques (SHELXT). If not noted otherwise, all non-hydrogen atoms were refined anisotropically, and hydrogen atoms were located at calculated positions. Figures of the solid-state molecular structures were generated using XP as implemented in SHELXTL program.

A summary of the crystallographic data and selected experimental information are given in Table 1. Crystallographic data for the structures of compounds reported in this paper have been deposited with the Cambridge Crystallographic Data Center as supplementary publication nos. CCDC 2489970 (**4i**).



CCDC 2489970

Table 1 Crystal data and structure refinement for CCDC 2489970.

Identification code	CCDC 2489970
Empirical formula	C ₂₉ H ₂₂ BrCl ₃ N ₄ O

Formula weight	628.76
Temperature/K	298.0
Crystal system	triclinic
Space group	P-1
a/Å	9.3480(8)
b/Å	10.0431(8)
c/Å	15.9468(13)
α /°	80.587(3)
β /°	76.524(3)
γ /°	87.501(3)
Volume/Å ³	1436.3(2)
Z	2
$\rho_{\text{calc}}/\text{cm}^3$	1.454
μ/mm^{-1}	1.740
F(000)	636.0
Crystal size/mm ³	0.15 × 0.15 × 0.1
Radiation	MoK α (λ = 0.71073)
2 Θ range for data collection/°	4.11 to 57.706
Index ranges	-12 ≤ h ≤ 12, -13 ≤ k ≤ 13, -21 ≤ l ≤ 21
Reflections collected	43296
Independent reflections	7449 [R _{int} = 0.0986, R _{sigma} = 0.0655]
Data/restraints/parameters	7449/6/384
Goodness-of-fit on F ²	1.013
Final R indexes [I ≥ 2 σ (I)]	R ₁ = 0.0487, wR ₂ = 0.1138
Final R indexes [all data]	R ₁ = 0.1068, wR ₂ = 0.1360
Largest diff. peak/hole / e Å ⁻³	0.37/-0.64