Supplementary Information

Diastereodivergent synthesis of dispiroheterocyclic structures comprising pyrrolidinyloxindole and imidazothiazolotriazine moieties

Alexei N. Izmest'ev, Galina A. Gazieva, Valentina A. Karnoukhova, Angelina N. Kravchenko

Table of Contents

General Methods

Commercially available solvents and reagents were used as purchased.

NMR spectroscopy was performed at 298 K using an Bruker AM300 (300.13 MHz, ^{1}H; 75.47 MHz, ^{13}C) or an Bruker DRX500 (500.13 MHz, ^{1}H; 125.76 MHz, ^{13}C). Data is expressed in parts per million (ppm) downfield shift from tetramethylsilane with residual solvent as an internal reference (δ 2.50 ppm for dimethyl sulfoxide) and is reported as position (δ in ppm), multiplicity (s = singlet, brs = broad singlet, d = doublet, t = triplet, q = quartet, m = multiplet, dd = doublet of doublets), coupling constant (*J* in Hz) and integration (number of protons). ^{13}C NMR spectra were recorded at 298 K with complete proton decoupling. Data is expressed in parts per million (ppm) downfield shift relative to the internal reference (δ 39.51 ppm for dimethyl sulfoxide).

Infrared spectra were obtained on a BrukerALPHA spectrometer using KBr pellets and are reported in wavenumbers (cm^{-1}).

HRMS were performed at the Bruker micrOTOF II using electrospray ionization (ESI). The measurements were done in a positive ion mode (interface capillary voltage: 4500 V); mass range from m/z 50 to 3000 Da; external or internal calibration was done with Electrospray Calibrant Solution (Fluka). A syringe injection was used for solutions in MeCN or MeOH (flow rate 3 μL/min). N_2 was applied as a dry gas.

Dipolarophiles **4** and **5** were prepared according to a literature procedure.[1,2] Dispiroheterocycles **3f,g,n-p** were synthesized earlier by the reaction of benzylidene derivatives **4e,k,l**, sarcosine **8a** and isatins **9a-c** in the mixture of acetonitrile and chloroform (3 : 1) under reflux for 72 h.[3]

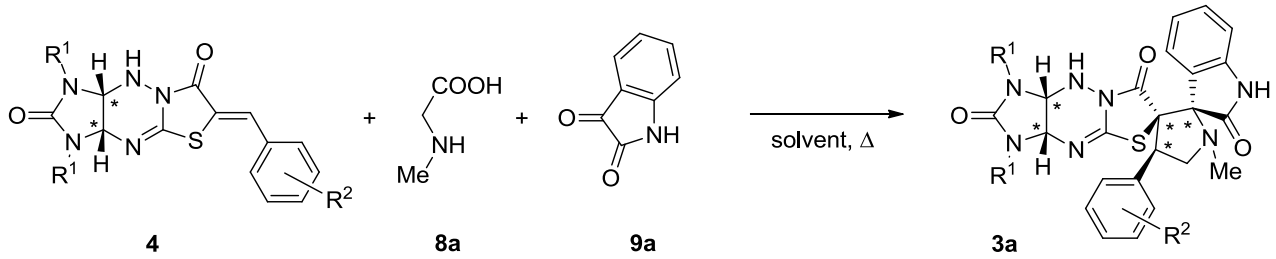
[1] A. N. Izmest'ev, D. A. Vasileva, E. K. Melnikova, N. G. Kolotyrkina, I. A. Borisova, A. N. Kravchenko, G. A. Gazieva, *New J. Chem*. **2019**, *43*, 1038–1052.

[2] A. N. Izmest'ev, N. A. Kim, V. A. Karnoukhova, N. G. Kolotyrkina, A. N. Kravchenko, G. A. Gazieva, *ChemistrySelect* **2019**, *4*, 10483–10487.

[3] A. N. Izmest'ev, G. A. Gazieva, N. V. Sigay, S. A. Serkov, V. A. Karnoukhova, V. V. Kachala, A. S. Shashkov, I. E. Zanin, A. N. Kravchenko, N. N. Makhova, *Beilstein J. Org. Chem*. **2016**, *12*, 2240-2249.

The interaction of compounds **4b, 8a** and **9a** was chosen as model reaction for the further optimization of the reaction parameters.

Table S1. Optimization of the 1,3-dipolar cycloaddition reaction conditions.[a]



4 **8a** **9a** **3a**

Entry	R^1	R^2	Solvent	t [h]	Equiv. of **8a** и **9a**	Yield of **3** [%][b,c]
1	Me	2-F	MeOH/$CHCl_3$ 1:3	36	1.0	**3b**, 35
2	Me	2-F	MeOH/$CHCl_3$ 1:3	72	1.5	**3b**, 49
3	Me	2-F	MeOH/$CHCl_3$ 1:3	120	1.5	**3b**, 51
4	Me	2-F	EtOH/$CHCl_3$ 1:3	36	1.5	**3b**, 55
5	Me	2-F	EtOH/$CHCl_3$ 1:3	72	1.5	**3b**, 91
6	Et	4-Br	MeOH/$CHCl_3$ 1:3	72	1.5	**3l**, 36
7	Et	4-Br	EtOH/$CHCl_3$ 1:3	72	1.5	**3l**, 82
8	Me	2,4-Cl_2	MeOH/$CHCl_3$ 1:8	72	1.5	**3h**, 61
9	Me	2,4-Cl_2	EtOH/$CHCl_3$ 1:3	72	1.5	**3h**, 74
10	Et	2,4-Cl_2	MeOH/$CHCl_3$ 1:3	72	1.5	**3p**, 63
11	Et	2,4-Cl_2	EtOH/$CHCl_3$ 1:3	72	1.5	**3p**, 74
12	Et	2,4-Cl_2	MeCN/$CHCl_3$ 3:1	72	1.5	**3p**, 74

[a]Reaction conditions: heating the solution of compounds **4** (1 mmol), isatin **9a** (1-1.5 mmol), and sarcosine (1-1.5 mmol) in the mixture of solvents (80 mL) for 72 h. [b]Yield of isolated product. [c]Only one diastereomer was observed.

General procedure for the reaction of dipolarophiles 4 with azomethyne ylides

A mixture of corresponding compound **4a-n** (1 mmol), isatin **9a-f** (1.5 mmol), and *N*-alkylglycine **8a,b** (1.5 mmol) in a mixture of ethanol (20 mL) and chloroform (60 ml) for the synthesis of compounds **3a-d,h-l,p-r,** (or in a mixture of acetonitrile (60 mL) and chloroform (20 ml) for the synthesis of compounds **3e-g,n,o,s-w,** or in acetonitrile (80 mL) for the synthesis of **3m**) was refluxed with stirring for 72 h. Then the reaction mixture was concentrated on a rotary evaporator to dryness. The residue was triturated with acetonitrile, formed precipitate of compounds **3a-w** was filtered off, washed with acetonitrile and dried in air.

General procedure for the reaction of dipolarophiles 5 with azomethyne ylides

A mixture of corresponding compound **5a-n** (1.0 mmol), isatin **9a-f** (1.5 mmol), and *N*-alkylglycine **8a,b** (1.5 mmol) in a mixture of ethanol (20 mL) and chloroform (60 ml) for the synthesis of compounds **6a-c,i-k,q,r** (or in a mixture of methanol (20 mL) and chloroform (60 ml) for the synthesis of compounds **6d,l**, or in a mixture of methanol (60 mL) and chloroform (20 ml) for the synthesis of compounds **6e-g,m-o,s-w**, or in methanol (80 mL) for the synthesis of compounds **6h,p**) was refluxed with stirring for 72 h. Then the reaction mixture was concentrated on a rotary evaporator to dryness. The residue was triturated with acetonitrile, formed precipitate of compounds **6a-w** was filtered off, washed with acetonitrile and dried in air.

General procedure for the rearrangement of compounds 3

To a stirred suspension of compounds **3a-w** (1.0 mmol) in refluxing methanol (20 ml), 0.10 ml of 40% aqueous KOH (1.0 mmol) was added. The resulting mixture was refluxed with stirring for 1 h. After cooling, the precipitate of compounds **7a-w** was filtered off, washed with methanol and dried.

Characterization data of products

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-1,1′,3-Trimethyl-4′-phenyl-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-2,2″,7(1*H*,1″*H*)-trione (3a).** Yield 247 mg (49%) as a light beige powder. Mp: 205-207 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.07 (s, 3H, 1′-NCH_3), 2.54 (s, 3H, NCH_3), 2.60 (s, 3H, NCH_3), 3.42 (t, *J* = 8.1 Hz, 1H, 5′-H), 3.92 (t, *J* = 9.3 Hz, 1H, 5′-H), 4.34 (t, *J* = 8.7, 1H, 4′-H), 4.43 (d, *J* = 5.9 Hz, 1H, 9a-H), 4.52 (d, *J* = 5.7 Hz, 1H, 3a-H), 6.61 (s, 1H, 9-NH), 6.82 (d, *J* = 7.5 Hz, 1H, 7″-H), 7.02 (t, *J* = 5.7 Hz, 1H, 5″-H), 7.21-7.35 (m, 7H, Ph, 4″-H, 6″-H), 10.74 (br.s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 26.80, 27.49 (1,3-NCH_3), 34.81 (1′-NCH_3), 52.55 (C-4′), 56.36 (C-5′), 65.17, 65.95 (C-3a, C-9a), 67.41 (C-3′), 79.20 (C-2′), 109.90 (C-7″), 122.41, 123.68, 127.20, 127.60, 128.59, 129.34, 130.44, 137.59 (C-3a″, C-4″, C-5″, C-6″, Ph), 143.76 (C-7a″), 149.01 (4a-C=N), 158.57 (2-C=O), 169.16 (7-C=O), 176.25 (2″-C=O); IR (KBr), ν 3400, 3301 (NH), 3090, 3030 (Ar), 2940, 2870 (Alk), 1709, 1637 (C=O, C=N); HRMS (ESI): Exact mass calcd for $C_{25}H_{25}N_7O_3S$ [*M* + H]$^+$: 504.1818, Found: 504.1812.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-4′-(2-Fluorophenyl)-1,1′,3-trimethyl-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-2,2″,7(1*H*,1″*H*)-trione (3b).** Yield 474 mg (91%) as a light beige powder. Mp: 214-216 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.06 (s, 3H, 1′-NCH_3), 2.58 (s, 6H, 1,3-NCH_3), 3.40 (t, *J* = 8.3 Hz, 1H, 5′-H), 4.01 (t, *J* = 9.4 Hz, 1H, 5′-H), 4.43 (d, *J* = 4.3 Hz, 1H, 9a-H), 4.52 (d, *J* = 5.6 Hz, 1H, 3a-H), 4.65 (t, *J* = 8.8 Hz, 1H, 4′-H), 6.60 (s, 1H, 9-NH), 6.83 (d, *J* = 7.6 Hz, 1H, 7″-H), 7.00-7.07 (m, 2H, 5″-H, 6″-H), 7.21-7.33 (m, 4H, 4′-Ar, 4″-H), 7.63 (t, *J* = 7.4 Hz, 1H, 4′-Ar), 10.71 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 26.78, 27.18 (1,3-NCH_3), 34.94 (1′-NCH_3), 43.16 (C-4′), 54.38 (C-5′), 65.31, 65.73 (C-3a, C-9a), 66.13 (C-3′), 79.32 (C-2′), 109.97 (C-7″), 114.87 (d, *J* = 21.9 Hz, 4′-Ar-3), 122.53, 123.20, 124.41, 126.67, 129.71, 130.52 (C-3a″, C-4″, C-5″, C-6″, 4′-Ar), 124.66 (d, *J* = 14.1 Hz, 4′-Ar-1), 129.46 (d, *J* = 8.6 Hz, 4′-Ar), 143.83 (C-7a″), 148.69 (4a-C=N), 158.39 (2-C=O), 161.24 (d, *J* = 245.0 Hz, C-F), 168.76 (7-C=O), 176.08 (2″-C=O); IR (KBr), ν 3413, 3251 (NH), 3088, 3052, 3016 (Ar), 2948, 2879 (Alk), 1712, 1638 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{25}H_{24}FN_7O_3S$ [*M* + Na]$^+$: 544.1532, Found: 544.1538.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-4′-(2-Fluorophenyl)-1,1′,3-trimethyl-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-2,2″,7(1*H*,1″*H*)-trione (3c).** Yield 281 mg (54%) as a beige powder. Mp: 225-227 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.06 (s, 3H, 1′-NCH_3), 2.54 (s, 3H, NCH_3), 2.60 (s, 3H, NCH_3), 3.42 (t, *J* = 8.1 Hz, 1H, 5′-H), 3.86 (t, *J* = 9.3 Hz, 1H, 5′-H), 4.33 (t, *J* = 8.4 Hz, 1H, 4′-H), 4.43 (d, *J* = 5.6 Hz, 1H, 9a-H), 4.52 (d, *J* = 6.0 Hz, 1H, 3a-H), 6.60 (s, 1H, 9-NH), 6.82 (d, *J* = 7.9 Hz, 1H, 7″-H), 7.63 (t, *J* = 7.6 Hz, 1H, 4′-Ar), 7.09-7.15 (m, 2H, 4″-H, 5″-H), 7.26-7.35 (m, 4H, 6″-H, 4′-Ar), 10.75 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 26.76, 27.49 (1,3-NCH_3), 34.75 (1′-NCH_3), 51.86 (C-4′), 56.58 (C-5′), 65.11, 65.94 (C-3a, C-9a), 67.34 (C-3′), 79.06 (C-2′), 109.89 (C-7″), 114.87 (d, *J* = 21.2 Hz, 4′-Ar-3,5), 122.42, 123.57, 127.21, 130.42, 133.82 (Ar-1, C-3a″, C-4″, C-5″, C-6″), 131.31 (d, *J* = 7.7 Hz, 4′-Ar-2,6), 143.72 (C-7a″), 148.83 (4a-C=N), 158.56 (2-C=O), 161.47 (d, *J* = 244.4 Hz, C-F), 169.01 (7-C=O), 176.23 (2″-C=O); IR (KBr), ν 3430 (NH), 3211, 3084 (Ar), 2946, 2841, 2799 (Alk), 1701, 1637 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{25}H_{24}FN_7O_3S$ [*M* + Na]$^+$: 544.1538, Found: 544.1538.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-4′-(4-Bromophenyl)-1,1′,3-trimethyl-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-2,2″,7(1*H*,1″*H*)-trione (3d).** Yield 355 mg (61%) as a light beige powder. Mp: 193-195 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.07 (s, 3H, 1′-NCH_3), 2.56 (s, 3H, NCH_3), 2.61 (s, 3H, NCH_3), 3.43 (t, *J* = 8.4 Hz, 1H, 5′-H), 3.87 (t, *J* = 9.3 Hz, 1H, 5′-H), 4.31 (t, *J* = 8.4 Hz, 1H, 4′-H), 4.46 (d, *J* = 5.6 Hz, 1H, 9a-H), 4.53 (d, *J* = 5.5 Hz, 1H, 3a-H), 6.63 (s, 1H, 9-NH), 6.83 (d, *J* = 7.7 Hz, 1H, 7″-H), 7.02 (t, *J* = 7.6 Hz, 1H, 5″-H), 7.18 (d, *J* = 7.9 Hz, 2H, 4′-Ar-2,6), 7.27-7.36 (m, 2H, 4″-H, 6″-H), 7.50 (d, *J* = 8.0 Hz, 2H, 4′-Ar-3,5), 10.78 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 26.74, 27.47 (1,3-NCH_3), 34.74 (1′-NCH_3), 52.03 (C-4′), 56.19 (C-5′), 65.13, 65.99 (C-3a, C-9a), 67.12 (C-3′), 79.03 (C-2′), 109.89 (C-7″), 120.82, 122.39, 123.50, 127.25, 130.40, 131.42, 131.47 (C-3a″, C-4″, C-5″, C-6″, 4′-Ar-2-6), 137.00 (4′-Ar-1), 143.73 (C-7a″), 148.79 (4a-C=N), 158.53 (2-C=O), 168.90 (7-C=O), 176.16 (2″-C=O); IR (KBr), ν 3423, 3211 (NH), 2944, 2872 (Alk), 1717, 1637 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{25}H_{24}BrN_7O_3S$ [*M* + H]$^{+}$: 582.0904, Found: 582.0917.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-1,1′,3-Trimethyl-4′-(4-nitrophenyl)-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-2,2″,7(1*H*,1″*H*)-trione (3e).** Yield 510 mg (93%) as a light gray powder. Mp: 243-244 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.08 (s, 3H, 1′-NCH_3), 2.53 (s, 3H, NCH_3), 2.63 (s, 3H, NCH_3), 3.50 (t, *J* = 8.2 Hz, 1H, 5′-H), 3.93 (t, *J* = 9.5 Hz, 1H, 5′-H), 4.46-4.55 (m, 3H, 3a-H, 4′-H, 9a-H), 6.67 (s, 1H, 9-NH), 6.84 (d, *J* = 7.7 Hz, 1H, 7″-H), 7.04 (t, *J* = 7.6 Hz, 1H, 5″-H), 7.29-7.36 (m, 2H, 4″-H, 6″-H), 7.53 (d, *J* = 8.6 Hz, 2H, 4′-Ar-2,6), 8.19 (d, *J* = 8.5 Hz, 2H, 4′-Ar-3,5), 10.83 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 26.73, 27.56 (1,3-NCH_3), 34.76 (1′-NCH_3), 52.14 (C-4′), 56.23 (C-5′), 65.13, 66.03 (C-3a, C-9a), 66.73 (C-3′), 79.10 (C-2′), 110.02 (C-7″), 122.53, 123.28, 123.57, 127.26, 130.55, 130.77 (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″), 143.75, 145.36 (4′-Ar-1, C-7a″), 146.87, 148.52 (4a-C=N, 4′-Ar-4), 158.59 (2-C=O), 168.67 (7-C=O), 176.17 (2″-C=O); IR (KBr), ν 3436, 3402, 3284 (NH), 3089 (Ar), 2947, 2920, 2870 (Alk), 1744, 1706, 1689, 1640 (C=O, C=N), 1519, 1348 (NO_2); HRMS (ESI): Exact mass calcd for $C_{25}H_{24}N_8O_5S$ [*M* + H]$^{+}$: 549.1641, Found: 549.1663.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-1″-Allyl-1,1′,3-trimethyl-4′-(4-nitrophenyl)-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-2,2′,7(1*H*,1″*H*)-trione (3f).**[3] Yield 383 mg (65%) as a light brown crystals. Mp: 238-240 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.06 (s, 3H, 1′-NCH_3), 2.52 (s, 3H, NCH_3), 2.63 (s, 3H, NCH_3), 3.54 (t, *J* = 8.2 Hz, 1H, 5′-H), 3.94 (t, *J* = 9.5 Hz, 1H, 5′-H), 4.22-4.43 (m, 2H, 1″-NCH_2), 4.46-4.56 (m, 3H, 3a-H, 4′-H, 9a-H), 5.15-5.22 (m, 2H, =CH_2), 5.75-5.86 (m, 1H, =CH), 6.67 (d, *J* = 1.8 Hz, 1H, 9-NH), 6.99 (d, *J* = 7.8 Hz, 1H, 7″-H), 7.13 (t, *J* = 7.6 Hz, 1H, 5″-H), 7.37-7.44 (m, 2H, 4″-H, 6″-H), 7.55 (d, *J* = 8.6 Hz, 2H, 4′-Ar-2,6), 8.20 (d, *J* = 8.5 Hz, 2H, 4′-Ar-3,5); ^{13}C NMR (75 MHz, DMSO-d_6): δ 26.70, 27.53 (1,3-NCH_3), 34.73 (1′-NCH_3), 41.25 (1″-NCH_2), 52.07 (C-4′), 56.40 (C-5′), 65.12, 65.98 (C-3a, C-9a), 66.77 (C-3′), 78.83 (C-2′), 109.61 (C-7″), 117.15, 122.49, 123.26, 123.59, 126.99, 130.64, 130.82, 131.38 (CH=CH_2, 4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″), 144.08, 145.25, 146.91, 148.21 (4′-Ar-1,4, C-7a″, 4a-C=N), 158.53 (2-C=O), 168.52 (7-C=O), 174.08 (2″-C=O); IR (KBr), ν 3464, 3289 (NH), 3080, 3059, 3033 (Ar), 2982, 2963, 2951 (Alk), 1714, 1699, 1644, 1610 (C=O, C=N), 1511, 1344 (NO_2); HRMS (ESI): Exact mass calcd for $C_{28}H_{28}N_8O_5S$ [*M* + H]$^{+}$: 589.1974, Found: 589.1976.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-5′′-Bromo-1′′-ethyl-1,1′,3-trimethyl-4′-(4-nitrophenyl)-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3′′-indole]-2,2′,7(1*H*,1′′*H*)-trione (3g).**[3] Yield 413 mg (63%) as a light yellow crystals. Mp: 250-252 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 1.13 (t, *J* = 7.0 Hz, 3H, CH_3), 2.08 (s, 3H, 1′-NCH_3), 2.55 (s, 3H, NCH_3), 2.67 (s, 3H, NCH_3), 3.55 (t, *J* = 8.2 Hz, 1H, 5′-H), 3.64-3.83 (m, 2H, 1′′-NCH_2), 3.95 (t, *J* = 9.4 Hz, 1H, 5′-H), 4.48-4.62 (m, 3H, 3a-H, 9a-H, 4′-H), 6.86 (s, 1H, 9-NH), 7.14 (d, *J* = 8.3 Hz, 1H, 7′′-H), 7.51-7.58 (m, 3H, 4′′-H, 4′-Ar-2,6), 7.64 (d, *J* = 8.3 Hz, 1H, 6′′-H), 8.21 (d, *J* = 8.4 Hz, 2H, 4′-Ar-3,5); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.40 (CH_3), 26.72, 27.55 (1,3-NCH_3), 34.15, 34.66 (1′-NCH_3, 1′′-NCH_2), 52.58 (C-4′), 56.17 (C-5′), 65.18, 66.15, 67.00 (C-3a, C-9a, C-3′), 78.26 (C-2′), 111.12 (C-7′′), 114.99, 123.57, 125.19, 130.20, 130.68, 133.52 (4′-Ar-2,3,5,6, C-3a′′, C-4′′, C-5′′, C-6′′), 143.33, 144.88 (4′-Ar-1, C-7a′′), 146.93, 148.10 (4′-Ar-4, 4a-C=N), 158.55 (2-C=O), 168.39 (7-C=O), 173.61 (2′′-C=O); IR (KBr), ν 3421, 3185 (NH), 3075 (Ar), 2978, 2949, 2900, 2868, 2853 (Alk), 1728, 1715, 1686, 1642, 1606 (C=O, C=N), 1518, 1345 (NO_2); HRMS (ESI): Exact mass calcd for $C_{27}H_{27}BrN_8O_5S$ [*M* + H]$^+$: 655.1083, Found: 655.1081.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-4′-(2,4-Diclorophenyl)-1,1′,3-trimethyl-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3′′-indole]-2,2′′,7(1*H*,1′′*H*)-trione (3h).** Yield 423 mg (74%) as a light gray powder. Mp: 217-219 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.06 (s, 3H, 1′-NCH_3), 2.53 (s, 3H, NCH_3), 2.60 (s, 3H, NCH_3), 3.47 (t, *J* = 8.1 Hz, 1H, 5′-H), 3.95 (t, 1H, *J* = 8.8 Hz, 5′-H), 4.42 (d, *J* = 5.3 Hz, 1H, 9a-H), 4.54 (d, *J* = 5.2 Hz, 1H, 3a-H), 4.69 (t, *J* = 8.6 Hz, 1H, 4′-H), 6.61 (s, 1H, 9-NH), 6.83 (d, *J* = 7.8 Hz, 1H, 7′′-H), 7.02 (t, *J* = 7.8 Hz, 1H, 5′′-H), 7.20 (d, *J* = 7.1 Hz, 1H, 4′′-H), 7.31 (t, *J* = 7.6 Hz, 1H, 6′′-H), 7.51 (d, *J* = 9.2 Hz, 1H, 4′-Ar-5), 7.60 (s, 1H, 4′-Ar-3), 7.83 (d, *J* = 8.5 Hz, 1H, 4′-Ar-6), 10.76 (s, 1H, 1′′-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 26.77, 27.41 (1,3-NCH_3), 34.99 (1′-NCH_3), 45.77 (C-4′), 55.05 (C-5′), 65.13 (C-3′), 65.23, 65.79 (C-3a, C-9a), 79.47 (C-2′), 110.14 (C-7′′), 122.73, 126.16, 127.41, 128.61, 130.64, 131.81, 132.85, 134.79, 136.74 (C-3a′′, C-4′′, C-5′′, C-6′′, 4′-Ar-2-6), 138.40 (4′-Ar-1), 143.83 (C-7a′′), 148.68 (4a-C=N), 158.39 (2-C=O), 168.64 (7-C=O), 175.98 (2′′-C=O); IR (KBr), ν 3449, 3233 (NH), 3098 (Ar), 2975, 2947 (Alk), 1710, 1687, 1636, 1620 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{25}H_{23}Cl_2N_7O_3S$ [*M* + H]$^+$: 572.1022, Found: 572.1033.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-1,3-Diethyl-1′-methyl-4′-phenyl-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3′′-indole]-2,2′′,7(1*H*,1′′*H*)-trione (3i).** Yield 431 mg (81%) as a light gray powder. Mp: 188-189 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.96-0.97 (m, 6H, 2CH_3), 2.07 (s, 3H, 1′-NCH_3), 2.81-2.88 (m, 1H, NCH_2), 3.01-3.43 (m, 5H, 1-CH_2, 3-CH_2, 5′-H), 4.33 (t, *J* = 7.4 Hz, 1H, 4′-H), 4.53-4.59 (m, 2H, 3a-H, 9a-H), 6.53 (s, 1H, 9-NH), 6.83 (d, *J* = 7.5 Hz, 1H, 7′′-H), 7.01 (t, *J* = 7.2 Hz, 1H, 5′′-H), 7.25-7.30 (m, 7H, 4′′-H, 6′′-H, Ph), 10.72 (s, 1H, 1′′-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 13.20, 13.59 (2CH_3), 34.08, 34.77, 35.22 (1,3-CH_2, 1′-NCH_3), 52.37 (4′-C), 56.82 (5′-C), 62.96, 64.66 (C-3a, C-9a), 67.23 (C-3′), 79.32 (C-2′), 109.88 (7′′-H), 122.39, 123.56, 127.00, 128.52, 129.58, 130.42 (Ph-2-6, C-3a′′, C-4′′, C-5′′, C-6′′), 137.64 (Ph-1), 143.76 (C-7a′′), 148.68 (4a-C=N), 157.72 (2-C=O), 169.09 (7-C=O), 176.22 (2′′-C=O); IR (KBr), ν 3400, 3301 (NH), 3090, 3030 (Ar), 2940, 2870 (Alk), 1709, 1637 (C=O, C=N); HRMS (ESI): Exact mass calcd for $C_{27}H_{29}N_7O_3S$ [*M* + H]$^+$: 532.2120, Found: 532.2125.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-4′-(2-Fluorophenyl)-1,3-diethyl-1′-methyl-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3′′-indole]-2,2′′,7(1*H*,1′′*H*)-trione (3j).** Yield 253 mg (46%) as a light beige powder. Mp: 194-196 °C; ^{1}H NMR (300

MHz, DMSO-d_6): δ 0.93-1.00 (m, 6H, $2CH_3$), 2.07 (s, 3H, 1′-NCH_3), 2.81-2.90 (m, 1H, NCH_2), 3.01-3.34 (m, 3H, NCH_2), 3.42 (t, *J* = 8.3 Hz, 3H, NCH_2), 3.97 (t, *J* = 9.3, 1H, 5′-H), 4.53-4.59 (m, 2H, 3a-H, 9a-H), 4.65 (t, *J* = 8.8, 1H, 4′-H), 6.55 (s, 1H, 9-NH), 6.83 (d, *J* = 7.6 Hz, 1H, 7″-H), 6.99-7.10 (m, 2H, 5″-H, 6″-H), 7.23-7.33 (m, 4H, 4″-H, 4′-Ar), 7.69 (t, *J* = 7.3 Hz, 1H, 4′-Ar), 10.73 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 13.00, 13.24 ($2CH_3$), 34.06, 34.95 (1,3-CH_2), 34.95 (1′-NCH_3), 54.94 (C-5′), 62.94, 64.29 (C-3a, C-9a), 66.06 (C-3′), 79.48 (C-2′), 110.02 (C-7″), 114.93 (d, *J* = 22.5 Hz, 4′-Ar-3), 122.59, 123.13, 124.43, 126.45, 130.56, 130.03 (4′-Ar, C-3a″, C-4″, C-5″, C-6″), 124.76 (d, *J* = 13.9 Hz, 4′-Ar-1), 129.50 (d, *J* = 8.1 Hz, 4′-Ar), 143.80 (C-7a″), 148.50 (4a-C=N), 157.66 (2-C=O), 161.16 (d, *J* = 244.7 Hz, C-F), 168.75 (7-C=O), 176.09 (2″-C=O); IR (KBr), ν 3422, 3214 (NH), 3089 (Ar), 2975, 2939, 2876, 2796 (Alk), 1719, 1702, 1638 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{27}H_{28}FN_7O_3S$ [*M* + Na]$^+$: 572.1852, Found: 572.1851.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-4′-(4-Fluorophenyl)-1,3-diethyl-1′-methyl-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-2,2″,7(1*H*,1″*H*)-trione (3k).** Yield 329 mg (60%) as a white powder. Mp: 204-206 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.94-0.99 (m, 6H, $2NCH_3$), 2.06 (s, 3H, 1′-NCH_3), 2.83-2.90 (m, 1H, NCH_2), 3.02-3.20 (m, 3H, NCH_2), 3.45 (t, *J* = 8.0 Hz, 1H, 5′-H), 3.84 (t, *J* = 9.4 Hz, 1H, 5′-H), 4.33 (t, *J* = 8.5 Hz, 1H, 4′-H), 4.54-4.60 (m, 2H, 3a-H, 9a-H), 6.63 (d, *J* = 2.1, 1H, 9-NH), 6.83 (d, *J* = 7.6 Hz, 1H, 7″-H), 7.02 (t, *J* = 7.4 Hz, 1H, 4′-Ar), 7.11-7.17 (m, 2H, 5″-H, 6″-H), 7.28-7.33 (m, 4H, 4′-Ar, 4″-H), 10.77 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 13.22, 13.50 ($2CH_3$), 34.20, 35.22 (1,3-CH_2), 34.76 (1′-NCH_3), 51.80 (C-4′), 57.04 (C-5′), 63.06, 64.64 (C-3a, C-9a), 67.23 (C-3′), 79.30 (C-2′), 109.95 (C-7″), 115.35 (d, *J* = 21.3 Hz, 4′-Ar-3, 4′-Ar-5), 122.47, 123.52, 127.11, 130.47 (C-3a″, C-4″, C-5″, C-6″), 131.54 (d, *J* = 8.2 Hz, 4′-Ar-2,6), 131.89 (d, *J* = 2.9 Hz, 4′-Ar-1), 143.77 (C-7a″), 148.62 (4a-C=N), 157.76 (2-C=O), 161.54 (d, *J* = 244.2 Hz, C-F), 169.03 (7-C=O), 178.13 (2″-C=O); IR (KBr), ν 3368 (NH), 3248, 3158, 3096 (Ar), 2974, 2935, 2868, 2798 (Alk), 1712, 1692, 1635 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{27}H_{28}FN_7O_3S$ [*M* + Na]$^+$: 572.1854, Found: 572.1851.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-4′-(4-Bromophenyl)-1,3-diethyl-1′-Methyl-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-2,2″,7(1*H*,1″*H*)-trione (3l).** Yield 500 mg (80%) as a white powder. Mp: 198-200 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.97 (t, 6H, $2CH_3$, *J* = 6.8 Hz), 2.06 (s, 3H, 1′-NCH_3), 2.85-2.92 (m, 1H, NCH_2), 3.05-3.21 (m, 3H, NCH_2), 3.44 (t, *J* = 8.8 Hz, 1H, 5′-H), 3.85 (t, *J* = 9.5 Hz, 1H, 5′-H), 4.30 (t, *J* = 8.5 Hz, 1H, 4′-H), 4.55 (d, *J* = 5.8, 2.2 Hz, 1H, 9a-H), 4.60 (d, *J* = 5.7 Hz, 1H, 3a-H), 6.62 (d, *J* = 2.4 Hz, 1H, 9-NH), 6.83 (d, *J* = 7.4 Hz, 1H, 7″-H), 7.02 (t, *J* = 8.1 Hz, 1H, 5″-H), 7.21 (d, *J* = 8.4 Hz, 2H, 4′-Ar-2,6), 7.27-7.34 (m, 2H, 4″-H, 6″-H), 7.51 (d, *J* = 8.3 Hz, 2H, 4′-Ar-3,5), 10.75 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.43, 12.57 ($2CH_3$), 34.62, 34.78, 36.88 (1,3-CH_2, 1′-NCH_3), 52.78 (4′-C), 57.09 (5′-C), 61.05, 63.03 (C-3a, C-9a), 68.99 (C-3′), 79.53 (C-2′), 109.88 (7″-H), 120.82, 122.30, 123.45, 127.29, 130.32, 131.22, 132.33, 134.17 (4′-Ar-2-6, C-3a″, C-4″, C-5″, C-6″), 137.26 (4′-Ar-1), 143.58 (C-7a″), 148.10 (4a-C=N), 157.73 (2-C=O), 173.08 (7-C=O), 176.56 (2″-C=O); IR (KBr), ν 3463, 3260 (NH), 3040 (Ar), 2975, 2944, 2872 (Alk), 1715, 1672, 1659 (C=O, C=N); HRMS (ESI): Exact mass calcd for $C_{27}H_{28}BrN_7O_3S$ [*M* + H]$^+$: 610.1218, Found: 610.1230.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-1′-Methyl-4′-(4-nitrophenyl)-1,3-diethyl-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-2,2″,7(1*H*,1″*H*)-trione (3m).** Yield 432 mg (75%) as a white powder. Mp: 252-254 °C; ^{1}H NMR (300 MHz,

DMSO-d_6): δ 0.91-1.01 (m, 6H, $2CH_3$), 2.08 (s, 3H, 1′-NCH_3), 2.79-2.89 (m, 1H, NCH_2), 3.05-3.20 (m, 3H, NCH_2), 3.51 (t, *J* = 8.1 Hz, 1H, 5′-H), 3.91 (t, *J* = 9.4 Hz, 1H, 5′-H), 4.48 (t, *J* = 8.5 Hz, 1H 4′-H), 4.56-4.62 (m, 2H, 3a-H, 9a-H), 6.68 (s, 1H, 9-NH), 6.84 (d, *J* = 7.6 Hz, 1H, 7″-H), 7.04 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.29-7.35 (m, 2H, 4″-H, 6″-H), 7.54 (d, *J* = 8.3 Hz, 2H, 4′-Ar-2,6), 8.20 (d, *J* = 8.3 Hz, 2H, 4′-Ar-3,5), 10.83 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 13.15, 13.38 ($2CH_3$), 34.23, 34.74, 35.23, (1,3-CH_2, 1′-NCH_3), 52.11 (C-4′), 56.56 (C-5′), 65.12, 64.60 (C-3a, C-9a), 66.60 (C-3′), 79.20 (C-2′), 110.02 (C-7″), 122.54, 123.20, 123.58, 127.16, 130.56, 130.92 (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″), 143.75, 145.39, 146.90, 148.26 (4′-Ar-1,4, C-7a″, 4a-C=N), 157.72 (2-C=O), 168.65 (7-C=O), 176.18 (2″-C=O); IR (KBr), ν 3436, 3413, 3280, 3184 (NH), 3071, 3057 (Ar), 2970, 2939 (Alk), 1716, 1693, 1635, 1616 (C=O, C=N), 1517, 1349 (NO_2); HRMS (ESI): Exact mass calcd for $C_{27}H_{28}N_8O_5S$ [*M* + H]$^+$: 577.1971, Found: 577.1976.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-1″-Allyl-1,3-diethyl-1′-methyl-4′-(4-nitrophenyl)-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-2,2′,7(1*H*,1″*H*)-trione (3n).**[3] Yield 438 mg (71%) as a light yellow crystals. Mp: 165-167 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.92-1.04 (m, 6H, $2CH_3$), 2.08 (s, 3H, 1′-NCH_3), 2.80-2.91 (m, 1H, NCH_2), 3.07-3.23 (m, 3H, NCH_2), 3.57 (t, *J* = 8.3 Hz, 1H, 5′-H), 3.95 (t, *J* = 9.4 Hz, 1H, 5′-H), 4.25-4.43 (m, 2H, 1″-NCH_2), 4.52 (t, *J* = 8.6, 1H, 4′-H), 4.59-4.65 (m, 2H, 3a-H, 9a-H), 5.18-5.24 (m, 2H, =CH_2), 5.78-5.90 (m, 1H, =CH), 6.73 (s, 1H, 9-NH), 7.02 (d, *J* = 7.8 Hz, 1H, 7″-H), 7.15 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.40-7.45 (m, 2H, 4″-H, 6″-H), 7.58 (d, *J* = 8.5 Hz, 2H, 4′-Ar-2,6), 8.22 (d, *J* = 8.3 Hz, 2H, 4′-Ar-3,5); ^{13}C NMR (75 MHz, DMSO-d_6): δ 13.18, 13.41 ($2CH_3$), 34.22, 34.74, 35.24, (1,3-CH_2, 1′-NCH_3), 41.28 (1″-NCH_2), 52.17 (C-4′), 56.71 (C-5′), 63.14, 64.63 (C-3a, C-9a), 66.67 (C-3′), 78.93 (C-2′), 109.65 (C-7″), 117.19, 122.45, 123.31, 123.62, 126.98, 130.68, 130.96, 131.41 (4′-Ar-2,3,5,6, CH=CH_2, C-3a″, C-4″, C-5″, C-6″), 144.12, 145.28, 146.94, 148.01 (4′-Ar-1,4, C-7a″, 4a-C=N), 157.70 (2-C=O), 168.56 (7-C=O), 174.10 (2″-C=O); IR (KBr), ν 3448, 3376, 3237 (NH), 3059, 3042, 3020 (Ar), 2977, 2962, 2942 (Alk), 1733, 1697, 1680, 1608 (C=O, C=N), 1524, 1350 (NO_2); HRMS (ESI): Exact mass calcd for $C_{30}H_{32}N_8O_5S$ [*M* + H]$^+$: 617.2283, Found: 617.2289.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-5″-Bromo-1,1″,3-triethyl-1′-methyl-4′-(4-nitrophenyl)-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-2,2″,7(1*H*,1″*H*)-trione (3o).**[3] Yield 431 mg (63%) as a light yellow crystals. Mp: 130-132 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.93 (t, *J* = 7.1 Hz, 3H, CH_3), 1.01 (t, *J* = 7.0 Hz, 3H, CH_3), 1.12 (t, *J* = 7.0 Hz, 3H, CH_3), 2.05 (s, 3H, 1′-NCH_3), 2.80-2.87 (m, 1H, NCH_2), 3.07-3.20 (m, 3H, NCH_2), 3.54 (t, *J* = 8.3 Hz, 1H, 5′-H), 3.62-3.81 (m, 2H, NCH_2), 3.91 (t, *J* = 8.6 Hz, 1H, 5′-H), 4.46 (t, *J* = 8.7 Hz, 1H, 4′-H), 4.61-4.66 (m, 2H, 3a-H, 9a-H), 6.85 (d, *J* = 1.8 Hz, 1H, 9-NH), 7.13 (d, *J* = 8.4 Hz, 1H, 7″-H), 7.50-7.54 (m, 3H, 4′-Ar-2,6, 4″-H), 7.62 (dd, *J* = 8.3, 2.0 Hz, 1H, 6″-H), 8.20 (d, *J* = 8.6 Hz, 2H, 4′-Ar-3,5) ; ^{13}C NMR (125 MHz, DMSO-d_6): δ 12.69, 13.41, 13.57 ($3CH_3$), 34.46, 34.62, 34.92, 35.50 ($4CH_2$), 52.92 (4′-C), 56.71 (5′-C), 63.52, 64.97, 67.19 (C-3′, C-3a, C-9a), 78.64 (C-2′), 111.43 (7″-H), 115.33 (C-5″), 123.87 (4′-Ar-3,5), 125.39, 130.43, 131.10, 133.86 (4′-Ar-2,6, C-3a″, C-4″, C-6″), 143.62, 145.17, 147.22, 148.29 (4′-Ar-1,4, C-7a″, 4a-C=N), 158.05 (2-C=O), 168.73 (7-C=O), 173.92 (2″-C=O); IR (KBr), ν 3441, 3282 (NH), 3107, 3076 (Ar), 2976, 2936, 2866 (Alk), 1698, 1649, 1605 (C=O, C=N), 1514, 1345 (NO_2); HRMS (ESI): Exact mass calcd for $C_{29}H_{31}BrN_8O_5S$ [*M* + H]$^+$: 683.1371, Found: 683.1394.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-4′-(2,4-Diclorophenyl)-1,3-diethyl-1′-methyl3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-**

2,2″,7(1*H*,1″*H*)-trione (3p).[3] Yield 445 mg (74%) as a white powder. Mp: 214-216 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.93-1.01 (m, 6H, 2CH_3), 2.06 (s, 3H, 1′-NCH_3), 2.86-2.93 (m, 1H, NCH_2), 3.00-3.10 (m, 2H, NCH_2), 3.17-3.28 (m, 1H, NCH_2), 3.48 (t, *J* = 8.3 Hz, 1H, 5′-H), 3.94 (t, *J* = 8.3 Hz, 1H, 5′-H), 4.54 (d, *J* = 6.0 Hz, 1H, 9a-H), 4.61 (d, *J* = 5.8 Hz, 1H, 3a-H), 4.70 (t, *J* = 8.5 Hz, 1H, 4′-H), 6.49 (s, 1H, 9-NH), 6.83 (d, *J* = 7.7 Hz, 1H, 7″-H), 7.02 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.18 (d, *J* = 7.5 Hz, 1H, 4″-H), 7.31 (t, *J* = 7.6 Hz, 1H, 6″-H), 7.51 (d, *J* = 8.4 Hz, 1H, 4′-Ar-5), 7.59 (s, 1H, 4′-Ar-3), 7.85 (d, *J* = 8.6 Hz, 1H, 4′-Ar-6), 10.75 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.45, 13.23 (2CH_3), 34.03, 34.89, 35.00 (1,3-CH_2, 1′-NCH_3), 45.42 (4′-C), 55.22 (5′-C), 62.72, 63.76 (C-3a, C-9a), 65.12 (C-3′), 79.56 (C-2′), 110.17 (7″-H), 122.70, 122.76, 126.04, 127.40, 128.55, 130.66, 131.89, 132.86, 134.84 (4′-Ar-2-6, C-3a″, C-4″, C-5″, C-6″), 136.73 (4′-Ar-1), 143.79 (C-7a″), 148.64 (4a-C=N), 157.56 (2-C=O), 168.63 (7-C=O), 175.97 (2″-C=O); IR (KBr), ν 3412, 3271 (NH), 3096 (Ar), 2974, 2934, 2876 (Alk), 1718, 1689, 1640 (C=O, C=N); HRMS (ESI): Exact mass calcd for $C_{27}H_{27}Cl_2N_7O_3S$ [*M* + Na]$^+$: 622.1151, Found: 622.1165.

(2′*R,3a*R**,3′*R**,4′*R**,9a*R**)-4′-(2-Fluorophenyl)-1,1′,3-trimethyl-2-thioxo-1,2,3,3a,9,9a-hexahydroimidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pirrolidine-2′,3″-indole)-2″,7(1″*H*)-dione (3q).** Yield 344 mg (64%) as a white powder. Mp: 201-203 °C, ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.06 (s, 3H, 1′-NCH_3), 2.90 (s, 3H, NCH_3), 2.94 (s, 3H, NCH_3), 3.43 (t, *J* = 8.2 Hz, 1H, 5′-H), 3.88 (t, *J* = 9.4 Hz, 1H, 5′-H), 4.32 (t, *J* = 8.7 Hz, 1H, 4′-H), 4.76-4.83 (m, 2H, 3a-H, 9a-H), 6.81-6.85 (m, 2H, 7″-H, 9-NH), 7.03 (t, *J* = 7.5 Hz, 1H, Ar), 7.13-7.37 (m, 6H, Ar, 4″-H, 5″-H, 6″-H), 10.77 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 30.65, 31.10 (1,3-NCH_3), 34.74 (1′-NCH_3), 52.11 (C-4′), 56.10 (C-5′), 67.02, 67.28, 68.37 (C-3′, C-3a, C-9a), 78.91 (C-2′), 109.90 (C-7″), 115.53 (d, *J* = 21.3 Hz, Ar-3, Ar-5), 122.44, 123.48, 127.37, 130.43 (C-3a″, C-4″, C-5″, C-6″), 131.04 (d, *J* = 8.2 Hz, Ar-2,6), 133.65 (d, *J* = 2.9 Hz, Ar-1), 143.74 (C-7a″), 150.12 (C-4a), 161.34 (d, *J* = 243.8 Hz, C-F), 168.91 (7-C=O), 176.14 (2″-C=O), 182.75 (2-C=S); IR (KBr), ν 3433, 3307, 3257, 3220 (NH), 3080, 3033 (Ar), 2973, 2946, 2913, 2868, 2797 (Alk), 1715, 1637 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{25}H_{24}FN_7O_2S_2$ [M + Na]$^+$: 560.1303, Found: 560.1309.

(2′*R,3a*R**,3′*R**,4′*R**,9a*R**)-1,3-Diethyl-4′-(2-fluorophenyl)-1′-methyl-2-thioxo-1,2,3,3a,9,9a-hexahydroimidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pirrolidine-2′,3″-indole)-2″,7(1″*H*)-dione (3r).** Yield 334 mg (62%) as a white powder. Mp: 238-240 °C, ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.96-1.08 (m, 6H, 2CH_3), 2.06 (s, 3H, 1′-NCH_3), 3.09-3.21 (m, 1H, NCH_2), 3.42-3.56 (m, 3H, NCH_2, 5′-H), 3.64-3.74 (m, 1H, NCH_2), 3.85 (t, *J* = 9.5 Hz, 1H, 5′-H), 4.31 (t, *J* = 8.8 Hz, 1H, 4′-H), 4.54 (d, *J* = 6.5 Hz, 1H, 3a-H), 4.89 (dd, *J* = 6.6, 2.5 Hz, 1H, 9a-H), 6.82-6,88 (m, 2H, 7″-H, 9-NH), 7.02 (t, *J* = 7.5 Hz, 1H, ArH), 7.11-7.17 (m, 2H, 5″-H, 6″-H), 7.14-7.35 (m, 6H, ArH, 4″-H), 10.80 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.50, 12.89 (2CH_3), 34.76, 38.30 (2NCH_2), 38.20 (1′-NCH_3), 52.03 (C-4′), 56.59 (C-5′), 64.89, 67.14 (C-3a), C-9a), 67.17 (C-3′), 79.08 (C-2′), 109.96 (C-7″), 115.53 (d, *J* = 21.3 Hz, Ar-3,5), 122.52, 123.43, 127.29, 130.50 (C-3a″, C-4″-6″), 131.31 (d, *J* = 8.2 Hz, Ar-2,6), 133.73 (d, *J* = 2.9 Hz, Ar-1), 143.77 (C-7a″), 149.93 (4a-C=N, 164.44 (d, *J* = 244.0 Hz, C-F), 168.95 (7-C=O), 176.20 (2″-C=O), 181.17 (2-C=S); **ИК (KBr), ν (см$^{-1}$)**: 3430, 3266 (NH), 3150, 3113, 3096, 3065,3035 (Ar), 2980, 2934, 2875, 2801 (Alk), 1721, 1630 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{27}H_{28}FN_7O_2S_2$ [M + Na]$^+$: 588.1612, Found: 588.1622.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-1′-Ethyl-1,3-dimethyl-4′-(4-nitrophenyl)-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-2,2″,7(1*H*,1″*H*)-trione (3s).** Yield 428 mg (76%) as a white powder. Mp: 259-260 °C; ^{1}H NMR (300 MHz,

DMSO-d_6): δ 0.95 (t, *J* = 7.0 Hz, 3H, CH_3), 2.07-2.19 (m, 1H, NCH_2), 2.29-2.35 (m, 1H, NCH_2), 2.52 (s, 3H, NCH_3), 2.62 (s, 3H, NCH_3), 3.65 (t, *J* = 8.1 Hz, 1H, 5′-H), 3.81 (t, *J* = 9.4 Hz, 1H, 5′-H), 4.47-4.55 (m, 3H, 3a-H, 4′-H, 9a-H), 6.63 (s, 1H, 9-NH), 6.82 (d, *J* = 7.7 Hz, 1H, 7″-H), 7.03 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.27-7.38 (m, 2H, 4″-H, 6″-H), 7.52 (d, *J* = 8.4 Hz, 2H, 4′-Ar-2,6), 8.18 (d, *J* = 8.3 Hz, 2H, 4′-Ar-3,5), 10.80 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 13.64 (CH_3), 26.71, 27.53 ($2NCH_3$), 42.53 (1′-NCH_2), 51.98, 53.13 (C-4′, C-5′), 65.08, 66.01 (C-3a, C-9a), 66.64 (C-3′), 79.05 (C-2′), 109.92 (C-7″), 122.49, 123.55, 123.83, 127.28, 130.48, 130.78 (Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″), 143.73, 145.49, 146.85, 148.52 (4′-Ar-1,4, C-7a″, 4a-C=N), 158.56 (2-C=O), 168.61 (7-C=O), 176.61 (2″-C=O); IR (KBr), ν 3435, 3382, 3283, 3201 (NH), 3066, 3055 (Ar), 2944, 2934, 2922, 2883 (Alk), 1738, 1722, 1698, 1640, 1618 (C=O, C=N), 1518, 1349 (NO_2); HRMS (ESI): Exact mass calcd for $C_{26}H_{26}N_8O_5S$ [*M* + H]$^+$: 563.1815, Found: 563.1820.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-1,1′,3-Triethyl-4′-(4-nitrophenyl)-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-2,2″,7(1*H*,1″*H*)-trione (3t).** Yield 402 mg (68%) as a white powder. Mp: 232-234 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.91-1.01 (m, 9H, $3CH_3$), 2.12-2.18 (m, 1H, NCH_2), 2.31-2.34 (m, 1H, NCH_2), 2.80-2.89 (m, 1H, NCH_2), 3.04-3.20 (m, 3H, NCH_2), 3.66 (t, *J* = 7.8 Hz, 1H, 5′-H), 3.79 (t, *J* = 9.2 Hz, 1H, 5′-H), 4.48 (t, *J* = 8.4 Hz, 1H, 4′-H), 4.57-4.61 (m, 2H, 3a-H, 9a-H), 6.65 (s, 1H, 9-NH), 6.83 (d, *J* = 7.6 Hz, 1H, 7″-H), 7.03 (t, *J* = 7.4 Hz, 1H, 5″-H), 7.28-7.36 (m, 2H, 4″-H, 6″-H), 7.53 (d, *J* = 8.2 Hz, 2H, 4′-Ar-2,6), 8.19 (d, *J* = 8.1 Hz, 2H, 4′-Ar-3,5), 10.82 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 13.21, 13.45, 13.67 ($3CH_3$), 34.29, 35.31 (1,3-NCH_2), 42.58 (1′-NCH_2), 52.00, 53.56 (C-4′, C-5′), 63.14, 64.69 (C-3a, C-9a), 66.56 (C-3′), 79.24 (C-2′), 110.02 (C-7″), 122.58, 123.61, 123.79, 127.22, 130.58, 131.00 (Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″), 143.79, 145.59, 146.94, 148.36 (4′-Ar-1,4, C-7a″, 4a-C=N), 157.80 (2-C=O), 168.69 (7-C=O), 176.71 (2″-C=O); IR (KBr), ν 3434, 3291, 3186 (NH), 2977, 2941, 2875 (Alk), 1716, 1697, 1636, 1618 (C=O, C=N), 1519, 1349 (NO_2); HRMS (ESI): Exact mass calcd for $C_{28}H_{30}N_8O_5S$ [*M* + H]$^+$: 591.2131, Found: 591.2133.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-7″-Chloro-1,1′,3-trimethyl-4′-(4-nitrophenyl)-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-2,2″,7(1*H*,1″*H*)-trione (3u).** Yield 442 mg (76%) as a beige powder. Mp: 241-243 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.07 (s, 3H, 1′-NCH_3), 2.53 (s, 3H, NCH_3), 2.62 (s, 3H, NCH_3), 3.52 (t, *J* = 8.1 Hz, 1H, 5′-H), 3.93 (t, *J* = 9.5 Hz, 1H, 5′-H), 4.49-4.58 (m, 3H, 3a-H, 9a-H, 4′-H), 6.64 (s, 1H, 9-NH), 7.09 (t, *J* = 7.9 Hz, 1H, 5″-H), 7.32 (d, *J* = 7.5 Hz, 1H), 7.41 (d, *J* = 8.2 Hz, 1H), 7.51 (d, *J* = 8.4 Hz, 2H, 4′-Ar-2,6), 8.19 (d, *J* = 8.4 Hz, 2H, 4′-Ar-3,5), 11.31 (s, 1H, 1″-NH); ^{13}C NMR (125 MHz, DMSO-d_6): δ 26.78, 27.64 (1-NCH_3, 3-NCH_3), 34.89 (1′-NCH_3), 52.15, 56.23 (C-4′, C-5′), 65.10, 66.07 (C-3a, C-9a), 66.92 (C-3′), 79.73 (C-2′), 114.22 (C-7″), 123.70 (4′-Ar-3,5), 123.91, 125.34, 126.04, 130.70 (C-3a″, C-4″, C-5″, C-6″), 130.79 (4′-Ar-2,6), 141.36, 145.14, 146.97, 148.31 (4′-Ar-1,4, C-7a″, 4a-C=N), 158.63 (2-C=O), 168.46 (7-C=O), 176.41 (2″-C=O); IR (KBr), ν 3288 (NH), 29.47, 2869 (Alk), 1708, 1642 (C=O, C=N), 1520, 1349 (NO_2); HRMS (ESI): Exact mass calcd for $C_{25}H_{23}ClN_8O_5S$ [*M* + H]$^+$: 583.1273, Found: 583.1272.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-1,1′,3,6″-Tetramethyl-4′-(4-nitrophenyl)-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-2,2″,7(1*H*,1″*H*)-trione (3v).** Yield 461 mg (82%) as a beige powder. Mp: 237-239 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.07 (s, 3H, 1′-NCH_3), 2.24 (s, 3H, CH_3), 2.27 (s, 3H, CH_3), 2.54 (s, 3H, CH_3), 3.52 (t, *J* = 8.1 Hz, 1H, 5′-H), 3.86 (t, *J* = 9.3 Hz, 1H, 5′-H), 4.43-4.49 (m, 2H, 3a-H, 9a-H), 4.56 (t, *J* = 8.6 Hz, 1H, 4′-H), 6.61 (s,

1H, 9-NH), 6.77-6.81 (m, 2H, 5″-H, 7″-H), 7.03 (d, *J* = 7.6 Hz, 1H, 6″-H), 7.69 (d, *J* = 8.4 Hz, 2H, 4′-Ar-2,6), 8.24 (d, *J* = 8.3 Hz, 2H, 4′-Ar-3,5), 10.69 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 21.28 (6″-CH_3), 26.65, 27.55 (1-NCH_3, 3-NCH_3), 34.74 (1′-NCH_3), 51.10, 57.47 (C-4′, C-5′), 64.74, 65.61 (C-3a, C-9a), 66.64 (C-3′), 79.31 (C-2′), 110.68 (C-7″), 120.16, 123.11, 123.50, 125.83 (4′-Ar-3,5, C-3a″, C-4″, C-5″), 131.62 (4′-Ar-2,6), 140.18 (C-6″), 143.42, 146.18, 146.90, 148.25 (4′-Ar-1,4, C-7a″, 4a-C=N), 158.30 (2-C=O), 169.20 (7-C=O), 176.72 (2″-C=O); IR (KBr), ν 3434 (NH), 3071 (Ar), 2934, 2869 (Alk), 1711, 1635 (C=O, C=N), 1520, 1347 (NO_2); HRMS (ESI): Exact mass calcd for $C_{26}H_{26}N_8O_5S$ [*M* + H]$^+$: 563.1820, Found: 563.1811.

(2′*R,3a*S**,3′*R**,4′*R**,9a*R**)-1,1′,3,7″-Tetramethyl-4′-(4-nitrophenyl)-3,3a,9,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[3,2-*b*][1,2,4]triazine-6,3′-pyrrolidine-2′,3″-indole]-2,2″,7(1*H*,1″*H*)-trione (3w).** Yield 364 mg (65%) as a light gray powder. Mp: 254-256 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.07 (s, 3H, 1′-NCH_3), 2.19 (s, 3H, 7″-CH_3), 2.53 (s, 3H, NCH_3), 2.63 (s, 3H, NCH_3), 3.48 (t, *J* = 8.2 Hz, 1H, 5′-H), 3.95 (t, *J* = 9.4 Hz, 1H, 5′-H), 4.46-4.57 (m, 3H, 3a-H, 9a-H, 4′-H), 6.64 (d, *J* = 1.8 Hz, 1H, 9-NH), 6.94 (t, *J* = 7.6 Hz, 1H, 5″-H), 7.13 (d, *J* = 7.6 Hz, 1H), 7.19 (d, *J* = 7.6 Hz, 1H), 7.51 (d, *J* = 8.6 Hz, 2H, 4′-Ar-2,6), 8.19 (d, *J* = 8.5 Hz, 2H, 4′-Ar-3,5), 10.85 (s, 1H, 1″-NH); ^{13}C NMR (150 MHz, DMSO-d_6): δ 16.32 (7″-CH_3), 26.76, 27.59 (1-NCH_3, 3-NCH_3), 34.85 (1′-NCH_3), 52.26, 56.13 (C-4′, C-5′), 65.11, 66.08 (C-3a, C-9a), 66.81 (C-3′), 79.22 (C-2′), 119.26, 122.48, 123.03, 123.64, 124.64, 130.77, 131.91 (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″, C-7″), 142.27, 145.44, 146.89, 148.63 (4′-Ar-1,4, C-7a″, 4a-C=N), 158.64 (2-C=O), 168.70 (7-C=O), 176.77 (2″-C=O); IR (KBr), ν 3434, 3297 (NH), 3073 (Ar), 2865, 2843, 2789 (Alk), 1703, 1642 (C=O, C=N), 1519, 1345 (NO_2); HRMS (ESI): Exact mass calcd for $C_{26}H_{26}N_8O_5S$ [*M* + H]$^+$: 563.1820, Found: 563.1816.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-1,1′,3-Trimethyl-4′-phenyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6a).** Yield 267 mg (53%) as a light gray powder. Mp: 234-236 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.07 (s, 3H, 1′-NCH_3), 2.42 (s, 3H, NCH_3), 2.92 (s, 3H, NCH_3), 3.46 (t, *J* = 7.5 Hz, 1H, 5′-H), 3.88 (t, *J* = 9.3 Hz, 1H, 5′-H), 4.37-4.42 (m, 2H, 3a-H, 4′-H), 5.26 (d, *J* = 6.6 Hz, 1H, 9a-H), 6.82 (d, *J* = 7.2 Hz, 1H, 7″-H), 7.01 (t, *J* = 7.8 Hz, 1H, 5″-H), 7.19 (d, *J* = 7.3 Hz, 1H, 6″-H), 7.26-7.36 (m, 7H, 4-NH, 4″-H, Ph), 10.70 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 27.44, 31.34 (1,3-NCH_3), 34.81 (1′-NCH_3), 51.46 (C-4′), 57.84 (C-5′), 65.07, 66.58 (C-3a, C-9a), 70.36 (C-3′), 79.67 (C-2′), 109.90 (7″-H), 122.07, 123.79, 125.96, 127.60, 128.64, 129.79, 130.34, 136.21 (Ph, C-3a″, C-4″, C-5″, C-6″), 138.39 (5a-C=N), 143.76 (C-7a″), 158.63 (2-C=O), 173.18 (8-C=O), 176.47 (2″-C=O); IR (KBr), ν 3298, 3183 (NH), 3063, 3028 (Ar), 2942, 2873, 2846 (Alk), 1718, 1694, 1672, 1645 (C=O, C=N); HRMS (ESI): Exact mass calcd for $C_{25}H_{25}N_7O_3S$ [*M* + H]$^+$: 504.1820, Found: 504.1812.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-4′-(2-Fluorophenyl)-1,1′,3-trimethyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6b).** Yield 406 mg (78%) as a light beige powder. Mp: 220-222 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.01 (s, 3H, 1′-NCH_3), 2.46 (s, 3H, NCH_3), 2.89 (s, 3H, NCH_3), 3.46 (t, *J* = 8.2 Hz, 1H, 5′-H), 4.04 (t, *J* = 9.2 Hz, 1H, 5′-H), 4.40 (d, *J* = 6.6 Hz, 1H, 3a-H), 4.73 (t, *J* = 8.6 Hz, 1H, 4′-H), 5.30 (d, *J* = 6.6 Hz, 1H, 9a-H), 6.81 (d, *J* = 7.7 Hz, 1H, 7″-H), 7.02 (t, *J* = 7.6 Hz, 1H, 5″-H), 7.14-7.37 (m, 6H, 4′-Ar, 4″-H, 6″-H, 4-NH), 7.73 (t, *J* = 7.3 Hz, 1H, 4′-Ar), 10.67 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 27.63, 30.87 (1,3-NCH_3), 35.00 (1′-NCH_3), 41.63 (C-4′), 54.94 (C-5′), 64.96, 66.26 (C-3a, C-9a), 69.46 (C-3′), 79.67 (C-2′), 109.99 (C-7″), 115.01 (d, *J* = 22.2 Hz, 4′-Ar-3), 122.17, 123.29, 124.55, 125.58, 129.42, 129.53, 130.44 (4′-

Ar-4-6, C-3a″, C-4″, C-5″, C-6″), 125.24 (d, *J* = 13.8 Hz, 4′-Ar-1), 135.85 (5a-C=N), 143.82 (C-7a″), 158.60 (2-C=O), 161.28 (d, *J* = 243.9 Hz, C-F), 172.82 (8-C=O), 176.26 (2″-C=O); IR (KBr), ν 3342, 3307 (NH), 2948, 2874, 2838 (Alk), 1717, 1696, 1659 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{25}H_{24}FN_7O_3S$ [*M* + Na]$^+$: 544.1536, Found: 544.1538.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-4′-(4-Fluorophenyl)-1,1′,3-trimethyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6c).** Yield 297 mg (57%) as a white powder. Mp: 225-227 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.07 (s, 3H, 1′-NCH$_3$), 2.42 (s, 3H, NCH$_3$), 2.91 (s, 3H, NCH$_3$), 3.48 (t, *J* = 8.3 Hz, 1H, 5′-H), 3.83 (t, *J* = 9.2 Hz, 1H, 5′-H), 4.39-4.44 (m, 2H, 3a-H, 4′-H), 5.26 (d, *J* = 6.5 Hz, 1H, 9a-H), 6.82 (d, *J* = 7.7 Hz, 1H, 7″-H), 7.01 (t, *J* = 7.4 Hz, 1H, 5″-H), 7.15-7.29 (m, 4H, 4″-H, 6″-H, 4-NH, 4′-Ar), 7.42 (t, *J* = 6.4 Hz, 2H, 4′-Ar), 10.72 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 27.45, 31.33 (1,3-NCH$_3$), 34.78 (1′-NCH$_3$), 50.57 (C-4′), 58.07 (C-5′), 65.02, 66.57 (C-3a, C-9a), 70.24 (C-3′), 79.63 (C-2′), 109.95 (C-7″), 115.39 (d, *J* = 21.2 Hz, 4′-Ar-3), 122.12, 123.68, 125.91, 130.38, 134.67 (4′-Ar-5, C-3a″, C-4″, C-5″, C-6″), 131.81 (d, *J* = 7.7 Hz, 4′-Ar), 135.91 (5a-C=N), 143.74 (C-7a″), 158.64 (2-C=O), 161.55 (d, *J* = 244.1 Hz, C-F), 173.01 (8-C=O), 176.50 (2″-C=O); IR (KBr), ν 3353, 3287 (NH), 3070 (Ar), 2972, 2939, 2876 (Alk), 1717, 1655, 1613 (C=O, C=N); HRMS (ESI): Exact mass calcd for $C_{25}H_{24}FN_7O_3S$ [*M* + Na]$^+$: 544.1531, Found: 544.1538.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-4′-(4-Bromophenyl)-1,1′,3-trimethyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6d).** Yield 175 mg (30%) as a light beige powder. Mp: 204-206 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.07 (s, 3H, 1′-NCH$_3$), 2.43 (s, 3H, NCH$_3$), 2.92 (s, 3H, NCH$_3$), 3.48 (t, *J* = 8.3 Hz, 1H, 5′-H), 3.82 (t, *J* = 9.3 Hz, 1H, 5′-H), 4.35-4.42 (m, 2H, 3a-H, 4′-H), 5.26 (d, *J* = 6.7 Hz, 1H, 9a-H), 6.82 (d, *J* = 7.7 Hz, 1H, 7″-H), 7.02 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.19 (d, *J* = 7.4 Hz, 1H, 4″-H), 7.26-7.35 (m, 4H, 4-NH, 4′-Ar-2,6, 6″-H), 7.55 (d, *J* = 8.2 Hz, 2H, 4′-Ar-3,5), 10.72 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 27.42, 31.36 (1,3-NCH$_3$), 34.76 (1′-NCH$_3$), 50.80 (C-4′), 57.73 (C-5′), 65.04, 66.63 (C-3a, C-9a), 69.94 (C-3′), 79.58 (C-2′), 109.95 (C-7″), 120.93, 122.12, 123.58, 125.93, 130.39, 131.50, 132.02 (4′-Ar-2-6, C-3a″, C-4″, C-5″, C-6″), 135.82 (5a-C=N), 137.81 (4′-Ar-1), 143.71 (C-7a″), 158.62 (2-C=O), 172.91 (8-C=O), 176.44 (2″-C=O); IR (KBr), ν 3449, 3247 (NH), 3023 (Ar), 2948, 2872 (Alk), 1707, 1648, 1620 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{25}H_{24}BrN_7O_3S$ [*M* + H]$^+$: 582.0920, Found: 582.0917.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-1,1′,3-Trimethyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6e).** Yield 515 mg (94%) as a light beige powder. Mp: 228-230 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.08 (s, 3H, 1′-NCH$_3$), 2.42 (s, 3H, NCH$_3$), 2.94 (s, 3H, NCH$_3$), 3.54 (t, *J* = 8.2 Hz, 1H, 5′-H), 3.89 (t, *J* = 9.2 Hz, 1H, 5′-H), 4.42 (d, *J* = 6.3 Hz, 1H, 3a-H), 4.57 (t, *J* = 8.6 Hz, 1H, 4′-H), 5.29 (d, *J* = 6.5 Hz, 1H, 9a-H), 6.83 (d, *J* = 7.6 Hz, 1H, 7″-H), 7.03 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.20-7.36 (m, 3H, 4-NH, 4″-H, 6″-H), 7.66 (d, *J* = 8.3 Hz, 2H, 4′-Ar-2,6), 8.22 (d, *J* = 8.2 Hz, 2H, 4′-Ar-3,5), 10.79 (s, 1 H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 27.43, 31.45 (1,3-NCH$_3$), 34.79 (1′-NCH$_3$), 51.00 (C-4′), 57.63 (C-5′), 65.05, 66.82 (C-3a, C-9a), 69.55 (C-3′), 79.13 (C-2′), 110.08 (C-7″), 122.25, 123.42, 123.64, 1276.02, 130.53, 131.27 (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″), 135.53, 143.06, 146.13, 146.97 (4′-Ar-1,4, C-7a″, 5a-C=N), 158.70 (2-C=O), 172.70 (8-C=O), 176.50 (2″-C=O); IR (KBr), ν 3337, 3228 (NH), 3083, 3059, 3011 (Ar), 2953 (Alk),

1707, 1665, 1649 (C=O, C=N), 1522, 1347 (NO_2); HRMS (ESI): Exact mass calcd for $C_{25}H_{24}N_8O_5S$ [*M* + H]$^+$: 549.1649, Found: 549.1663.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-1″-Allyl-1,1′,3-trimethyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2′,8(1″*H*,3*H*)-trione (6f).** Yield 454 mg (77%) as a light brown crystals. Mp: 162-164 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.06 (s, 3H, 1′-NCH_3), 2.41 (s, 3H, NCH_3), 2.94 (s, 3H, NCH_3), 3.59 (t, *J* = 8.3 Hz, 1H, 5′-H), 3.91 (t, *J* = 9.4 Hz, 1H, 5′-H), 4.21-4.43 (m, 3H, 1″-NCH_2, 3a-H), 4.61 (t, *J* = 8.5 Hz, 1H, 4′-H), 5.12-5.30 (m, 3H, =CH_2, 9a-H), 5.78-5.87 (m, 1H, =CH), 6.95 (d, *J* = 7.8 Hz, 1H, 7″-H), 7.12 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.27 (d, *J* = 7.5 Hz, 1H, 4″-H), 7.34-7.41 (m, 2H, 6″-H, 4-NH), 7.70 (d, *J* = 8.9 Hz, 2H, 4′-Ar-2,6), 8.24 (d, *J* = 8.5 Hz, 2H, 4′-Ar-3,5); ^{13}C NMR (75 MHz, DMSO-d_6): δ 27.38, 31.31 (1,3-NCH_3), 34.76 (1′-NCH_3), 41.26 (1″-NCH_2), 50.60 (C-4′), 57.82 (C-5′), 65.01, 66.75, 69,67 (C-3a, C-3′, C-9a), 79.54 (C-2′), 109.66 (C-7″), 116.84, 122.64, 122.95, 123.69, 125.59, 130.62, 131.26, 131.40, 135.10 (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″, 5a-C=N, $CH=CH_2$), 144.14 (C-7a″), 146.01, 146.98 (4′-Ar-1,4), 158.58 (2-C=O), 172.41 (8-C=O), 174.43 (2″-C=O); IR (KBr), ν 3435, 3380 (NH), 3080, 3055 (Ar), 2946, 2920 (Alk), 1727, 1707, 1648, 1609 (C=O, C=N), 1517, 1343 (NO_2); HRMS (ESI): Exact mass calcd for $C_{28}H_{28}N_8O_5S$ [*M* + H]$^+$: 589.1972, Found: 589.1976.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-5″-Bromo-1,1′,3-trimethyl-4′-(4-nitrophenyl)-1″-ethyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2′,8(1″*H*,3*H*)-trione (6g).** Yield 426 mg (63%) as a light yellow crystals. Mp: 220-222 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 1.15 (t, *J* = 7.0 Hz, 3H, CH_3), 2.09 (s, 3H, 1′-NCH_3), 2.44 (s, 3H, NCH_3), 2.97 (s, 3H, NCH_3), 3.57-3.95 (m, 4H, NCH_2, 5′-H), 4.57-4.63 (m, 2H, 3a-H, 4′-H), 5.24 (d, *J* = 7.9 Hz, 1H, 9a-H), 7.14 (d, *J* = 8.3 Hz, 1H, 7″-H), 7.36 (d, *J* = 2.1 Hz, 1H, 4″-H), 7.42 (d, *J* = 1.8 Hz, 1H, 4-NH), 7.63 (dd, *J* = 8.3, 2.1 Hz, 1H, 6″-H), 7.68 (d, *J* = 9.0 Hz, 2H, 4′-Ar-2,6), 8.24 (d, *J* = 8.7 Hz, 2H, 4′-Ar-3,5); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.37 (CH_3), 27.39, 31.35, 34.17, 34.64 (3NCH_3, NCH_2), 50.81 (C-4′), 57.83 (C-5′), 65.35, 67.05 (C-3a, C-9a), 70.10 (C-3′), 79.01 (C-2′), 111.26 (C-7″), 114.46, 123.70, 125.39, 128.49, 131.22, 133.46, 135.15 (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″, 5a-C=N), 143.33, 145.66, 147.01 (4′-Ar-1,4, C-7a″), 158.49 (2-C=O), 172.29 (8-C=O), 173.96 (2″-C=O); IR (KBr), ν 3436, 3345 (NH), 3067, 3056, 3041 (Ar), 2981, 2945 (Alk), 1717, 1689, 1657, 1606 (C=O, C=N), 3436, 3345 (NO_2); HRMS (ESI): Exact mass calcd for $C_{27}H_{27}BrN_8O_5S$ [*M* + H]$^+$: 655.1080, Found: 655.1081.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-4′-(2,4-Diclorophenyl)-1,1′,3-trimethyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6h).** Yield 549 mg (96%) as a white powder. Mp: 254-255 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.07 (s, 3H, 1′-NCH_3), 2.44 (s, 3H, NCH_3), 2.96 (s, 3H, NCH_3), 3.48 (t, *J* = 8.3 Hz, 1H, 5′-H), 3.98 (t, *J* = 9.0 Hz, 1H, 5′-H), 4.48 (d, *J* = 6.4 Hz, 1H, 3a-H), 4.75 (t, *J* = 8.5 Hz, 1H, 4′-H), 5.31 (d, *J* = 6.5 Hz, 1H, 9a-H), 6.82 (d, *J* = 7.7 Hz, 1H, 7″-H), 7.02 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.17 (d, *J* = 7.4 Hz, 1H, 4″-H), 7.26-7.31 (m, 2H, 4-NH, 6″-H), 7.52 (d, *J* = 8.4 Hz, 1H, 4′-Ar-5), 7.65 (s, 1H, 4′-Ar-3), 7.87 (d, *J* = 8.6 Hz, 1H, 4′-Ar-6), 10.69 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 27.61, 31.74 (1,3-NCH_3), 34.93 (1′-NCH_3), 45.22 (C-4′), 55.34 (C-5′), 65.68, 66.89 (C-3a, C-9a), 68.48 (C-3′), 79.62 (C-2′), 110.06 (C-7″), 122.24, 122.95, 125.59, 127.41, 128.61, 130.47, 131.58 (4′-Ar-3,5,6, C-3a″, C-4″, C-5″, C-6″), 132.84, 135.14, 136.21, 136.64 (4′-Ar-1,2,4, 5a-C=N), 143.82 (C-7a″), 158.82 (2-C=O), 172.76 (8-C=O), 176.19 (2″-C=O); IR

(KBr), ν 3436, 3327 (NH), 3098, 3068 (Ar), 2945, 2888, 2841 (Alk), 1713, 1698, 1679, 1654 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{25}H_{23}Cl_2N_7O_3S$ [*M* + H]$^+$: 572.1026, Found: 572.1033.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-1,3-Diethyl-1′-methyl-4′-phenyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6i).** Yield 367 mg (69%) as a light beige powder. Mp: 215-217 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.84 (t, *J* = 6.9 Hz, 3H, CH_3), 1.12 (t, *J* = 6.6 Hz, 3H, CH_3), 2.07 (s, 3H, 1′-NCH_3), 2.80-2.87 (m, 1H, NCH_2), 3.01-3.08 (m, 1H, NCH_2), 3.23-3.53 (m, 3H, NCH_2 , 5′-H), 3.89 (t, *J* = 9.2 Hz, 1H, 5′-H), 4.37-4.45 (m, 2H, 3a-H, 4′-H), 5.27 (d, *J* = 6.6 Hz, 1H, 9a-H), 6.81 (d, *J* = 7.5 Hz, 1H, 7″-H), 7.02 (t, *J* = 7.2 Hz, 1H, 5″-H), 7.18-7.36 (m, 8H, 4″-H, 6″-H, 4-NH, Ph), 10.70 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.26, 13.72 ($2CH_3$), 34.37, 34.80 (1,3-NCH_2), 38.83 (1′-NCH_3), 51.16 (4′-C), 57.94 (5′-C), 62.67, 64.65 (C-3a, C-9a), 70.30 (C-3′), 79.71 (C-2′), 109.91 (7″-H), 122.06, 123.78, 125.91, 127.58, 128.63, 129.82, 130.31, 135.81, 138.40 (Ph, C-3a″, C-4″, C-5″, C-6″, 5a-C=N), 143.80 (C-7a″), 157.72 (2-C=O), 172.99 (8-C=O), 176.47 (2″-C=O); IR (KBr), ν 3222, 3134 (NH), 3065, 3038 (Ar), 2912, 2893, 2843 (Alk), 1708, 1693, 1652, 1655 (C=O, C=N); HRMS (ESI): Exact mass calcd for $C_{27}H_{29}N_7O_3S$ [*M* + H]$^+$: 532.2110, Found: 532.2125.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-1,3-Diethyl-4′-(2-fluorophenyl)-1′-methyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6j).** Yield 390 mg (71 %) as a white powder. Mp: 166-169 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.88 (t, *J* = 6.4 Hz, 3H, CH_3), 1.13 (t, *J* = 6.6 Hz, 3H, CH_3), 2.09 (s, 3H, 1′-NCH_3), 2.82-2.88 (m, 1H, NCH_2), 3.06-3.24 (m, 2H, NCH_2), 3.42-3.52 (m, 2H, NCH_2, 5′-H), 4.01 (t, *J* = 8.5 Hz, 1H, 5′-H), 4.36 (d, *J* = 5.4, 1H, 3a-H), 4.73 (t, *J* = 8.0 Hz, 1H, 4′-H), 5.28 (d, *J* = 5.9 Hz, 1H, 9a-H), 6.81 (d, *J* = 7.3 Hz, 1H, 7″-H), 7.03 (t, *J* = 6.7 Hz, 1H, 5″-H), 7.14-7.33 (m, 6H, 4′-Ar, 4″-H, 6″-H, 4-NH), 7.74 (t, *J* = 7.1 Hz, 1H, 4′-Ar), 10.69 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.57, 13.52 ($2CH_3$), 34.74, 35.24 (1,3-NCH_2), 37.85 (1′-NCH_3), 41.44 (C-4′), 55.50 (C-5′), 62.28, 64.36 (C-3a, C-9a), 69.57 (C-3′), 80.00 (C-2′), 110.26 (C-7″), 115.24 (d, *J* = 22.1 Hz, 4′-Ar-3), 122.41, 123.53, 124.78 (d, *J* = 3.3 Hz), 125.51 (d, *J* = 13.7 Hz), 125.75, 129.53, 130.69 (4′-Ar-1,4-6, C-3a″, C-4″, C-5″, C-6″), 135.35 (5a-C=N), 144.11 (C-7a″), 157.96 (2-C=O), 161.47 (d, *J* = 244.7 Hz, C-F), 172.96 (8-C=O), 176.51 (2″-C=O); IR (KBr), ν 3296 (NH), 3092 (Ar), 2974, 2936, 2875, 2796 (Alk), 1721, 1650, 1619, 1599 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{27}H_{28}FN_7O_3S$ [*M* + Na]$^+$: 572.1851, Found: 572.1852.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-1′-Methyl-4′-(4-fluorophenyl)-1,3-diethyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6k).** Yield 500 mg (91%) as a light beige powder. Mp: 240-242 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.84 (t, , *J* = 6.4 Hz 3H, CH_3), 1.12 (t, *J* = 6.6 Hz, 3H, NCH_3), 2.07 (s, 3H, 1′-NCH_3), 2.81-2.88 (m, 1H, NCH_2), 3.01-3.08 (m, 1H, NCH_2), 3.23-3.34 (m, 2H, NCH_2), 3.49 (t, *J* = 9.1 Hz, 1H, 5′-H), 3.83 (t, *J* = 9.0 Hz, 1H, 5′-H), 4.38-4.46 (m, 2H, 3a-H, 4′-H), 5.28 (d, *J* = 6.3 Hz, 1H, 9a-H), 6.82 (d, *J* = 7.2 Hz, 1H, 7″-H), 7.02 (t, *J* = 7.2 Hz, 1H, 5″-H), 7.15-7.31 (m, 6H, 4′-Ar, 4″-H, 6″-H, 4-NH), 7.39-7,34 (m, 2H, 4′-Ar), 10.72 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.26, 13.67 ($2CH_3$), 34.37, 34.75 (1,3-NCH_2), 38.11 (1′-NCH_3), 50.37 (C-4′), 58.12 (C-5′), 62.45, 64.63 (C-3a, C-9a), 70.18 (C-3′), 79.64 (C-2′), 109.94 (C-7″), 115.37 (d, *J* = 21.2 Hz, 4′-Ar-3,5), 122.08, 123.68, 125.89, 130.32, 134.63 (Ar-2,6, C-3a″, C-4″, C-5″, C-6″), 131.81 (d, *J* = 7.7 Hz, 4′-Ar-1), 135.56 (5a-C=N), 143.77 (C-7a″), 157.70 (2-C=O), 161.53 (d, *J* = 244.2 Hz, C-F), 172.85 (8-C=O), 176.50 (2″-C=O); IR (KBr), ν 3331, 3204 (NH), 3101, 3072, 3042, 3016 (Ar), 2979,

2965, 2946, 2880, 2847, 2795 (Alk), 1717, 1704, 1684, 1644, 1622, 1603 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{27}H_{28}FN_7O_3S$ [*M* + Na]$^+$: 572.1860, Found: 572.1851.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-4′-(4-Bromophenyl)-1,3-diethyl-1′-methyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6l).** Yield 335 mg (55%) as a light beige powder. Mp: 168-170 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.84 (t, *J* = 6.9 Hz, 3H, CH_3), 1.12 (t, *J* = 6.8 Hz, 3H, CH_3), 2.06 (s, 3H, 1′-NCH_3), 2.81-2.88 (m, 1H, NCH_2), 3.00-3.08 (m, 1H, NCH_2), 3.23-3.30 (m, 1H, NCH_2), 3.44-3.53 (m, 2H, NCH_2, 5′-H), 3.82 (t, *J* = 9.2 Hz, 1H, 5′-H), 4.38 (t, *J* = 8.6 Hz, 1H, 4′-H), 4.47 (dd, *J* = 6.5, 1.8 Hz, 1H, 3a-H), 5.30 (d, *J* = 6.6 Hz, 1H, 9a-H), 6.82 (d, *J* = 7.5 Hz, 1H, 7″-H), 7.02 (t, *J* = 7.8 Hz, 1H, 5″-H), 7.19 (d, *J* = 7.1 Hz, 1H, 4″-H), 7.26-7.33 (m, 4H, 4-NH, 4′-Ar-2,6, 6″-H), 7.55 (d, *J* = 8.4 Hz, 2H, 4′-Ar-3,5), 10.75 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.33, 13.76 ($2CH_3$), 34.43, 34.80 (1,3-NCH_2), 38.21 (1′-NCH_3), 50.72 (C-4′), 57.81 (C-5′), 62.79, 64.74 (C-3a, C-9a), 69.95 (C-3′), 79.64 (C-2′), 110.04 (C-7″), 120.97, 122.16, 123.64, 126.00, 130.41, 131.57, 132.08 (Ar-2-6, C-3a″, C-4″, C-5″, C-6″), 135.57 (5a-C=N), 143.82 (C-7a″), 157.76 (2-C=O), 172.81 (8-C=O), 176.50 (2″-C=O); IR (KBr), ν 3435, 3333, 3257 (NH), 3095 (Ar), 2976, 2943, 2872 (Alk), 1708, 1649, 1620 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{27}H_{28}BrN_7O_3S$ [*M* + H]$^+$: 610.1225, Found: 610.1230.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-1,3-Diethyl-1′-methyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6m).** Yield 403 mg (70%) as a light yellow powder. Mp: 237-239 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.83 (t, *J* = 6.7 Hz, 3H, CH_3), 1.14 (t, *J* = 6.4 Hz, 3H, CH_3), 2.11 (s, 3H, 1′-NCH_3), 2.81-2.88 (m, 1H, NCH_2), 3.00-3.07 (m, 1H, NCH_2), 3.26-3.34 (m, 1H, NCH_2), 3.49-3.54 (m, 2H, NH_2, 5′-H), 3.90 (t, *J* = 8.8 Hz, 1H, 5′-H), 4.47-4.59 (m, 2H, 3a-H, 4′-H), 5.33 (d, *J* = 6.5 Hz, 1H, 9a-H), 6.83 (d, *J* = 6.5 Hz, 1H, 7″-H), 7.04 (t, *J* = 7.4 Hz, 1H, 5″-H), 7.21-7.32 (m, 3H, 4″-H, 6″-H, 4-NH), 7.65 (d, *J* = 8.2 Hz, 2H, 4′-Ar-2,6), 8.22 (d, *J* = 8.5 Hz, 2H, 4′-Ar-3,5); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.29, 13.70 (1-CH_2CH_3, 3-CH_2CH_3), 34.43, 34.75, 38.23, (1-CH_2, 3-CH_2, 1′-NCH_3), 50.95 (C-4′), 57.63 (C-5′), 62.80, 64.83 (C-3a, C-9a), 69.50 (C-3′), 79.65 (C-2′), 110.06 (C-7″), 122.20, 123.43, 123.63, 126.07, 130.45, 131.23 (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″), 135.24, 143.79, 146.06, 146.90 (4′-Ar-1,4, C-7a″, 5a-C=N), 157.73 (2-C=O), 172.54 (8-C=O), 176.49 (2″-C=O); IR (KBr), ν 3378, 3209 (NH), 3098, 3079 (Ar), 2974, 2944 (Alk), 1717, 1704, 1682, 1640 (C=O, C=N), 1525, 1351 (NO_2); HRMS (ESI): Exact mass calcd for $C_{27}H_{28}N_8O_5S$ [*M* + H]$^+$: 577.1969, Found: 577.1976.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-1″-Allyl-1,3-diethyl-1′-methyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2′,8(1″*H*,3*H*)-trione (6n).** Yield 426 mg (62%) as a light yellow powder. Mp: 206-208 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.83 (t, *J* = 7.0 Hz, 3H, CH_3), 1.14 (t, *J* = 6.9 Hz, 3H, CH_3), 2.06 (s, 3H, 1′-NCH_3), 2.81-2.88 (m, 1H, NCH_2), 3.00-3.07 (m, 1H, NCH_2), 3.26-3.33 (m, 1H, NCH_2), 3.49-3.61 (m, 2H, NCH_2, 5′-H), 3.92 (t, *J* = 8.2 Hz, 1H, 5′-H), 4.19-4.26 (m, 1H, 1″-NCH_2), 4.40-4.49 (m, 2H, 1″-NCH_2, 3a-H), 4.61 (t, *J* = 8.6 Hz, 1H, 4′-H), 5.12-5.16 (m, 2H, $=CH_2$), 5.35 (d, *J* = 6.6 Hz, 1H, 9a-H), 5.78-5.87 (m, 1H, =CH), 6.95 (d, *J* = 7.8 Hz, 1H, 7″-H), 7.12 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.28 (d, *J* = 7.5 Hz, 1H, 4″-H), 7.33-7.39 (m, 2H, 4-NH, 6″-H), 7.69 (d, *J* = 8.5 Hz, 2H, 4′-Ar-2,6), 8.23 (d, *J* = 8.4 Hz, 2H, 4′-Ar-3,5); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.27, 13.72 ($2CH_3$), 34.41, 34.75, 38.16 (1,3-CH_2, 1′-NCH_3), 41.30 (1″-NCH_2), 50.62 (C-4′), 57.86 (C-5′), 62.85, 64.85 (C-3a, C-9a), 69.64 (C-3′), 79.54 (C-2′), 109.67 (C-7″), 116.86, 122.65, 122.98, 123.74, 125.68,

130.62, 131.29, 134.92 (4′-Ar-2,3,5,6, CH=CH_2, C-3a″, C-4″, C-5″, C-6″, 5a-C=N), 144.20, 146.00, 146.96 (4′-Ar-1,4, C-7a″), 157.68 (2-C=O), 172.33 (8-C=O), 174.46 (2″-C=O); IR (KBr), ν 3346 (NH), 3083, 3060 (Ar), 2979, 2940, 2867 (Alk), 1705, 1693, 1658, 1610 (C=O, C=N), 1522, 1347 (NO_2); HRMS (ESI): Exact mass calcd for $C_{30}H_{32}N_8O_5S$ [*M* + H]$^+$: 617.2290, Found: 617.2289.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-5″-Bromo-1,1″,3-triethyl-1′-methyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6o).** Yield 424 mg (62%) as a light yellow powder. Mp: 204-205 °C; ^{1}H NMR (500 MHz, DMSO-d_6): δ 0.84 (t, *J* = 7.1 Hz, 3H, CH_3), 1.12-1.17 (m, 6H, $2CH_3$), 2.08 (s, 3H, 1′-CH_3), 2.84-2.89 (m, 1H, NCH_2), 3.00-3.06 (m, 1H, NCH_2), 3.26-3.31 (m, 1H, NCH_2), 3.50-3.68 (m, 3H, 5′-H, NCH_2), 3.77-3.83 (m, 1H, NCH_2), 3.90 (t, *J* = 9.3 Hz, 1H, 5′-H), 4.55-4.60 (m, 2H, 3a-H, 4′-H), 5.21 (d, *J* = 6.6 Hz, 1H, 9a-H), 7.12 (d, *J* = 8.4 Hz, 1H, 7″-H), 7.33 (d, *J* = 1.7 Hz, 1H, 4″-H), 7.41 (s, 1H, 4-NH), 7.62 (dd, *J* = 8.3, 1.8 Hz, 1H, 6″-H), 7.67 (d, *J* = 8.5 Hz, 2H, 4′-Ar-2,6), 8.23 (d, *J* = 8.6 Hz, 2H, 4′-Ar-3,5); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.33, 12.44, 13.53 ($3CH_3$), 34.25, 34.45, 34.82 (1,3-NCH_2, 1′-NCH_3), 38.14 (1″-NCH_2), 50.44 (4′-C), 58.02 (5′-C), 62.88, 64.91 (C-3a, C-9a), 70.09 (C-3′), 79.13 (C-2′), 111.36 (7″-H), 114.47 (C-5″), 123.80 (4′-Ar-3,5), 125.42, 128.43, 131.30, 133.51, 134.69 (4′-Ar-2,6, C-3a″, C-4″, C-6″, 5a-C=N), 143.41, 145.76, 147.04 (4′-Ar-1,4, C-7a″), 157.66 (2-C=O), 172.24 (8-C=O), 174.01 (2″-C=O); IR (KBr), ν 3400, 3357 (NH), 3111, 3064 (Ar), 2981, 2962, 2934, 2859 (Alk), 1713, 1695, 1656 (C=O, C=N), 1522, 1347 (NO_2); HRMS (ESI): Exact mass calcd for $C_{29}H_{31}BrN_8O_5S$ [*M* + H]$^+$: 683.1410, Found: 683.1394.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-4′-(2,4-Diclorophenyl)-1,3-diethyl-1′-methyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6p).** Yield 475 mg (79%) as a light beige powder. Mp: 209-211 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.87 (t, *J* = 6.9 Hz, 3H, CH_3), 1.14 (t, *J* = 6.9 Hz, 3H, CH_3), 2.07 (s, 3H, 1′-NCH_3), 2.78-2.90 (m, 1H, NCH_2), 3.03-3.14 (m, 1H, NCH_2), 3.27-3.37 (m, 1H, NCH_2), 3.45-3.56 (m, 2H, NCH_2, 5′-H), 3.97 (t, *J* = 9.0 Hz, 1H, 5′-H), 4.50 (dd, *J* = 6.2, 1.6 Hz, 1H, 3a-H), 4.74 (t, *J* = 8.8 Hz, 1H, 4′-H), 5.32 (d, *J* = 6.4 Hz, 1H, 9a-H), 6.82 (d, *J* = 7.7 Hz, 1H, 7″-H), 7.04 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.16 (d, *J* = 7.3 Hz, 1H, 4″-H), 7.26-7.32 (m, 2H, 4-NH, 6″-H), 7.51 (d, *J* = 8.4, 2.3 Hz, 1H, 4′-Ar-5), 7.63 (d, *J* = 2.2 Hz, 1H, 4′-Ar-3), 7.87 (d, *J* = 8.6 Hz, 1H, 4′-Ar-6), 10.72 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.11, 13.58 ($2CH_3$), 34.37, 34.99 (1,3-NCH_2), 38.22 (1′-NCH_3), 45.09 (C-4′), 55.43 (C-5′), 63.01, 64.52 (C-3a, C-9a), 68.40 (C-3′), 79.67 (C-2′), 110.01 (C-7″), 122.31, 122.97, 125.58, 127.42, 128.59, 130.49, 132.89, 135.16, 135.87, 136.68 (Ar-1-6, C-3a″, C-4″, C-5″, C-6″, 5a-C=N), 143.89 (C-7a″), 157.79 (2-C=O), 172.69 (8-C=O), 176.23 (2″-C=O); IR (KBr), ν 3333, 3291, 3275 (NH), 3066, 3019 (Ar), 2976, 2946, 2877 (Alk), 1714, 1693, 1652, 1620 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{27}H_{27}Cl_2N_7O_3S$ [*M* + H]$^+$: 600.1330, Found: 600.1346.

(2′*S,3a*R**,3′*S**,4′*S**,9a*R**)-4′-(4-Fluorophenyl)-1,1′,3-trimethyl-2-thioxo-1,2,3,3a,4,9a-hexahydrodispiro[imidazo[4,5-e]thiazolo[2,3-c][1,2,4]triazin-7,3′-pirrolidine-2′,3″-indole]-2″,8(1″*H*)-dione (6q).** Yield 393 mg (73%) as a light beige powder. Mp: 235-237 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.06 (s, 3H, 1′-NCH_3), 2.72 (s, 3H, NCH_3), 3.25 (s, 3H, NCH_3), 3.47 (t, *J* = 8.4 Hz, 1H, 5′-H), 3.85 (t, *J* = 9.3 Hz, 1H, 5′-H), 4.39 (t, *J* = 8.7 Hz, 1H, 3a-H), 4.88 (t, *J* = 6.9 Hz, 1H, 4′-H), 5.61 (d, *J* = 7.0 Hz, 1H, 9a-H), 6.83 (d, *J* = 7.6 Hz, 1H, 7″-H), 7.02 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.14-7.40 (m, 6H, 4″-H, 6″-H, 4-NH, 4′-Ar), 7.54 (s, 1H, 4′-Ar), 10.74 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 30.82, 34.82 (1,3-NCH_3), 35.66 (1′-NCH_3), 51.33 (C-4′), 57.60 (C-5′), 68.30, 68.77 (C-3a, C-9a), 70.31 (C-3′), 79.48 (C-2′), 110.05 (C-7″), 115.51 (d, *J* =

21.2 Hz, 4′-Ar-3,5), 122.30, 123.69, 126.37, 130.49 (C-3a″, C-4″, C-5″, C-6″), 131.80 (d, *J* = 8.1 Hz, 4′-Ar-2,6), 134.46 (4′-Ar-1), 137.47 (5a-C=N), 143.83 (C-7a″), 161.58 (d, *J* = 244.1 Hz, C-F), 172.89 (8-C=O), 176.55 (2″-C=O), 183.27 (2-C=S); IR (KBr), ν 3405, 3370 (NH), 3153, 3084, 3042, 3015 (Ar), 2976, 2962, 2933, 2897, 2871,2838, 2807, 2783 (Alk), 1735, 1713, 1641, 1620, 1604 (C=O, C=N); HRMS (ESI): Exact mass calcd for $C_{25}H_{24}FN_7O_2S_2$ $[M + Na]^+$: 560.1303, Found: 560.1309.

(2′*S,3a*R**,3′*S**,4′*S**,9a*R**)-1,3-Diethyl-4′-(4-fluorophenyl)-1′-methyl-2-thioxo-1,2,3,3a,4,9a-hexahydrodispiro[imidazo[4,5-e]thiazolo[2,3-c][1,2,4]triazin-7,3′-pirrolidine-2′,3″-indole]-2″,8(1″*H*)-dione (6r).** Yield 280 mg (52%) as a white powder. Mp: 245-247 °C; ^{1}H NMR (500 MHz, DMSO-d_6): δ 0.89 (t, *J* = 7.1 Hz, 3H, CH_3), 1.19 (t, *J* = 7.0 Hz, 3H,CH_3), 2.07 (s, 3H, 1′-NCH_3), 3.13-3.19 (m, 1H, NCH_2), 3.44-3.50 (m, 2H, NCH_2), 3.57-3.64 (m, 1H, NCH_2), 3.84 (t, *J* = 9.4 Hz, 1H, 5′-H), 3.99-4.06 (m, 1H, 5′-H), 4.38 (t, *J* = 8.8 Hz, 1H, 4′-H), 4.87 (dd, *J* = 7.2, 1.8 Hz, 1H, 3a-H), 5.61 (d, *J* = 7.2 Hz, 1H, 9a-H), 6.83 (d, *J* = 7.7 Hz, 1H, 7″-H), 7.03 (t, *J* = 7.6 Hz, 1H, 5″-H), 7.15-7.40 (m, 6H, 4′-Ar, 4″-H, 6″-H, 4-NH), 7.53 (d, *J* = 1.8 Hz, 1H, 4′-Ar), 10.75 (s, 1H, 1″-NH); ^{13}C NMR (125 MHz, DMSO-d_6): δ 11.47, 13.63 ($2CH_3$), 34.80, 37.71 (1,3-NCH_2), 41.63 (1′-NCH_3), 50.99 (C-4′), 57.72 (C-5′), 66.15, 66.81 (C-3a, C-9a), 70.29 (C-3′), 79.53 (C-2′), 110.04 (C-7″), 115.51 (d, *J* = 21.3 Hz, 4′-Ar-3,5), 122.29, 123.67, 126.29, 130.46 (C-3a″, C-4″, C-5″, C-6″), 131.83 (d, *J* = 8.1 Hz, 4′-Ar-2,6), 134.45 (d, *J* = 2.8 Hz, 4′-Ar-1), 136.93 (5a-C=N), 143.83 (C-7a″), 161.56 (d, *J* = 244.1 Hz, C-F), 172.68 (8-C=O), 176.55 (2″-C=O), 181.81 (2-C=S); IR (KBr), ν 3422, 3356, 3266 (NH), 3127, 3070, 3054 (Ar), 3002, 2977, 2955, 2931, 2871, 2802 (Alk), 1732, 1715, 1646, 1615, 1599 (C=O, C=N); HRMS (ESI): Exact mass calcd for $C_{27}H_{28}FN_7O_2S_2$ $[M + Na]^+$: 588.1616, Found: 588.1622.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-1′-Ethyl-1,3-dimethyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6s).** Yield 484 mg (86%) as a white powder. Mp: 226-228 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.94 (t, *J* = 7.0 Hz, 3H, CH_3), 2.13-2.19 (m, 1H, NCH_2), 2.30-2.36 (m, 1H, NCH_2), 2.41 (s, 3H, NCH_3), 2.94 (s, 3H, NCH_3), 3.66-3.80 (m, 2H, 5-CH_2), 4.41 (d, *J* = 6.6 Hz, 1H, 3a-H), 4.57 (t, *J* = 8.7 Hz, 1H, 4′-H), 5.26 (d, *J* = 6.6 Hz, 1H, 9a-H), 6.82 (d, *J* = 7.6 Hz, 1H, 7″-H), 7.02 (t, *J* = 7.6 Hz, 1H, 5″-H), 7.20-7.31 (m, 3H, 4″-H, 6″-H, 4-NH), 7.67 (d, *J* = 8.5 Hz, 2H, Ar-2,6), 8.22 (d, *J* = 8.4 Hz, 2H, Ar-3,5), 10.76 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 13.70 (CH_3), 27.47, 31.61 (1,3-NCH_3), 42.63 (1′-NCH_2), 50.81 (C-4′), 54.61 (C-5′), 65.09, 66.84 (C-3a, C-9a), 69.38 (C-3′), 79.73 (C-2′), 110.05 (C-7″), 122.27, 123.66, 123.97, 126.03, 130.52, 131.34 (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″), 135.59 (5a-C=N), 143.77, 146.34, 146.97 (4′-Ar-1,4, C-7a″), 158.75 (2-C=O), 172.70 (8-C=O), 176.98 (2″-C=O); IR (KBr), ν 3430, 3355, 3238 (NH), 3097 (Ar), 2944, 2875, 2825 (Alk), 1710, 1655, 1648 (C=O, C=N), 1523, 1348 (NO_2); HRMS (ESI): Exact mass calcd for $C_{26}H_{26}N_8O_5S$ $[M + H]^+$: 563.1816, Found: 563.1820.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-1,1′,3-Triethyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6t).** Yield 532 mg (90%) as a white powder. Mp: 214-215 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.83 (t, *J* = 7.0 Hz, 3H, CH_3), 0.94 (t, *J* = 7.0 Hz, 3H, CH_3), 1.13 (t, *J* = 6.9 Hz, 3H, CH_3), 2.13-2.20 (m, 1H, 1′-NCH_2), 2.30-2.36 (m, 1H, 1′-NCH_2), 2.80-2.93 (m, 1H, NCH_2), 2.99-3.06 (m, 1H, NCH_2), 3.23-3.37 (m, 1H, NCH_2), 3.46-3.56 (m, 1H, NCH_2), 3.67-3.80 (m, 2H, CH_2), 4.46 (d, *J* = 6.3 Hz, 1H, 3a-H), 4.56 (t, *J* = 8.5 Hz, 1H, 4′-H), 5.30 (d, *J* = 6.5 Hz, 1H, 9a-H), 6.82 (d, *J* = 7.7 Hz, 1H, 7″-H), 7.03 (t, *J* = 7.6 Hz, 1H, 5″-H), 7.21-7.29 (m, 3H, 4″-H, 6″-H, 4-NH), 7.65 (d, *J* = 8.4 Hz, 2H, Ar-2,6), 8.22 (d, *J* = 8.4 Hz, 2H, Ar-3,5), 10.76 (s, 1H, 1″-NH); ^{13}C NMR (75

MHz, DMSO-d_6): δ 12.30, 13.69 (3CH_3), 34.42, 38.18 (1,3-NCH_2), 42.60 (1′-NCH_2), 50.71 (C-4′), 54.60 (C-5′), 62.77, 64.76 (C-3a, C-9a), 69.33 (C-3′), 79.71 (C-2′), 110.02 (C-7″), 122.23, 123.66, 123.95, 126.03, 130.46, 131.30 (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″), 135.24 (5a-C=N), 143.78, 146.28, 146.91 (4′-Ar-1,4, C-7a″), 157.76 (2-C=O), 172.54 (8-C=O), 176.96 (2″-C=O); IR (KBr), ν 3346, 3248 (NH), 3095 (Ar), 2978, 296, 2870, 2848 (Alk), 1712, 1653 (C=O, C=N), 1523, 1347 (NO_2); HRMS (ESI): Exact mass calcd for $C_{28}H_{30}N_8O_5S$ [*M* + H]$^+$: 591.2135, Found: 591.2133.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-7″-Chloro-1,1′,3-trimethyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6u).** Yield 423 mg (73%) as a light beige powder. Mp: 247-249 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.11 (s, 3H, 1′-NCH_3), 2.42 (s, 3H, NCH_3), 2.94 (s, 3H, NCH_3), 3.57 (t, *J* = 8.4 Hz, 1H, 5′-H), 3.91 (t, *J* = 9.3 Hz, 1H, 5′-H), 4.48 (dd, *J* = 6.7, 1.8 Hz, 1H, 3a-H), 4.60 (t, *J* = 8.7 Hz, 1H, 4′-H), 5.32 (d, *J* = 6.7 Hz, 1H, 9a-H), 7.08 (t, *J* = 7.8 Hz, 1H, 5″-H), 7.18 (dd, *J* = 7.6, 1.1 Hz, 1H, 4″-H), 7.39-7.42 (m, 2H, 4-NH, 6″-H), 7.67 (d, *J* = 8.8 Hz, 2H, 4′-Ar-2,6), 8.24 (d, *J* = 8.8 Hz, 2H, 4′-Ar-3,5), 11.16 (s, 1 H, 1″-NH); ^{13}C NMR (125 MHz, DMSO-d_6): δ 27.47, 31.44 (1,3-NCH_3), 34.91 (1′-NCH_3), 50.78 (C-4′), 57.64 (C-5′), 65.29, 66.93 (C-3a, C-9a), 69.83 (C-3′), 80.33 (C-2′), 114.23 (C-7″), 123.63, 123.76, 124.68, 125.48, 130.68, 131.29, 135.34 (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″, C-7a″), 141.43, 145.93, 147.06 (4′-Ar-1,4, 5a-C=N), 158.68 (2-C=O), 172.36 (8-C=O), 176.64 (2″-C=O); IR (KBr), ν 3430, 3238 (NH), 3097, 3018 (Ar), 2974, 2944, 2875, 2825 (Alk), 1710, 1655, 1648 (C=O, C=N), 1523, 1348 (NO_2); HRMS (ESI): Exact mass calcd for $C_{25}H_{23}ClN_8O_5S$ [*M* + H]$^+$: 583.1273, Found: 583.1266.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-1,1′,3,6″-Tetramethyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6v).** Yield 345 mg (63%) as a light beige powder. Mp: 230-231 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.07 (s, 3H, 1′-NCH_3), 2.28 (s, 3H, 6″-CH_3), 2.42 (s, 3H, NCH_3), 2.95 (s, 3H, NCH_3), 3.51 (t, *J* = 8.2 Hz, 1H, 5′-H), 3.87 (t, *J* = 9.2 Hz, 1H, 5′-H), 4.47 (d, *J* = 6.5 Hz, 1H, 3a-H), 4.54 (t, *J* = 8.5 Hz, 1H, 4′-H), 5.33 (d, *J* = 6.6 Hz, 1H, 9a-H), 6.65 (s, 1H, 7″-H), 6.83 (d, *J* = 7.6 Hz, 1H, 5″-H), 7.10 (d, *J* = 7.7 Hz, 1H, 4″-H), 7.34 (s, 1H, 4-NH), 7.65 (d, *J* = 8.3 Hz, 2H, 4′-Ar-2,6), 8.21 (d, *J* = 8.2 Hz, 2H, 4′-Ar-3,5), 10.73 (s, 1 H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 21.12 (C-6″), 27.24, 31.34 (1,3-NCH_3), 34.52 (1′-NCH_3), 51.13 (C-4′), 57.26 (C-5′), 64.97, 66.68 (C-3a, C-9a), 69.18 (C-3′), 79.33 (C-2′), 110.57 (C-7″), 120.18, 122.69, 123.69, 125.80 (4′-Ar-3,5, C-3a″, C-4″, C-5″), 131.06 (4′-Ar-2,6), 135.53 (5a-C=N), 140.10 (C-6″), 143.68, 145.95, 146.73 (4′-Ar-1,4, C-7a″), 158.51 (2-C=O), 172.66 (8-C=O), 176.52 (2″-C=O); IR (KBr), ν 3373, 3185 (NH), 3078, 3040 (Ar), 2948, 2922, 2872 (Alk), 1710, 1638, 1597 (C=O, C=N), 1521, 1345 (NO_2); HRMS (ESI): Exact mass calcd for $C_{26}H_{26}N_8O_5S$ [*M* + H]$^+$: 563.1820, Found: 563.1824.

(2′*S,3a*R**,3′*S**,4′*S**,9a*S**)-1,1′,3,7″-Tetramethyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (6w).** Yield 399 mg (71%) as a light yellow powder. Mp: 231-233 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.07 (s, 3H, 1′-NCH_3), 2.18 (s, 3H, 7″-CH_3), 2.41 (s, 3H, NCH_3), 2.94 (s, 3H, NCH_3), 3.52 (t, *J* = 8.4 Hz, 1H, 5′-H), 3.91 (t, *J* = 9.2 Hz, 1H, 5′-H), 4.42 (d, *J* = 6.6 Hz, 1H, 3a-H), 4.56 (t, *J* = 8.5 Hz, 1H, 4′-H), 5.29 (d, *J* = 6.6 Hz, 1H, 9a-H), 6.94 (t, *J* = 7.6 Hz, 1H, 5″-H), 7.05 (d, *J* = 7.5 Hz, 1H), 7.11 (d, *J* = 7.5 Hz, 1H), 7.34 (s, 1H, 4-NH), 7.65 (d, *J* = 8.4 Hz, 2H, 4′-Ar-2,6), 8.22 (d, *J* = 8.5 Hz, 2H, 4′-Ar-3,5), 10.87 (s, 1 H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 16.38 (C-7″), 27.45, 31.53 (1,3-NCH_3), 34.83 (1′-NCH_3), 51.21

(C-4′), 57.49 (C-5′), 65.07, 66.86 (C-3a, C-9a), 69.56 (C-3′), 79.77 (C-2′), 119.25, 122.19, 123.14, 123.43, 123.66, 131.27, 131.85 (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″, C-7″), 135.76 (5a-C=N), 142.30, 146.15, 146.96 (4′-Ar-1,4, C-7a″), 158.74 (2-C=O), 172.77 (8-C=O), 177.04 (2″-C=O); IR (KBr), ν 3372 (NH), 3080 (Ar), 2946, 2865, 2845 (Alk), 1702, 1641 (C=O, C=N), 1520, 1349 (NO_2); HRMS (ESI): Exact mass calcd for $C_{26}H_{26}N_8O_5S$ [*M* + H]$^+$: 563.1820, Found: 563.1821.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-1,1′,3-Trimethyl-4′-phenyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7a).** Yield 179 mg (71%) as a light beige powder. Mp: 241-243 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.06 (s, 3H, 1′-NCH_3), 2.41 (s, 3H, NCH_3), 2.78 (s, 3H, NCH_3), 3.42 (t, *J* = 8.4 Hz, 1H, 5′-H), 3.87 (t, *J* = 9.6 Hz, 1H, 5′-H), 4.33 (t, *J* = 8.7, 1H, 4′-H) Hz, 4.46 (d, *J* = 4.8 Hz, 1H, 3a-H), 5.38 (d, *J* = 5.7 Hz, 1H, 9a-H), 6.80 (d, *J* = 7.5 Hz, 1H, 7″-H), 6.97 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.22-7.36 (m, 8H, 4″-H, 6″-H, 4-NH, Ph), 10.72 (br.s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 27.85, 31.33 (1,3-NCH_3), 34.76 (1′-NCH_3), 53.38 (C-4′), 57.19 (C-5′), 64.05, 65.87 (C-3a, C-9a), 69.14 (C-3′), 79.59 (C-2′), 109.81 (7″-H), 122.00, 123.57, 127.29, 127.40, 128.27, 130.03, 130.13, 134.76 (Ph, C-3a″, C-4″, C-5″, C-6″), 137.78 (5a-C=N), 143.84 (C-7a″), 159.06 (2-C=O), 173.26 (8-C=O), 176.62 (2″-C=O); IR (KBr), ν 3161, 3106 (NH), 3065, 3027 (Ar), 2952, 2863, 2793 (Alk), 1705, 1649, 1618 (C=O, C=N); HRMS (ESI): Exact mass calcd for $C_{25}H_{25}N_7O_3S$ [*M* + H]$^+$: 504.1814, Found: 504.1812.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-4′-(2-Fluorophenyl)-1,1′,3-trimethyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7b).** Yield 201 mg (77%) as a white powder. Mp: 278-280 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.08 (s, 3H, 1′-NCH_3), 2.44 (s, 3H, NCH_3), 2.80 (s, 3H, NCH_3), 3.38 (t, *J* = 8.2 Hz, 1H, 5′-H), 4.03 (t, *J* = 9.5 Hz, 1H, 5′-H), 4.46 (dd, *J* = 5.7, 1.9 Hz, 1H, 3a-H), 4.60 (t, *J* = 8.8 Hz, 1H, 4′-H), 5.34 (d, *J* = 5.7 Hz, 1H, 9a-H), 6.80 (d, *J* = 7.7 Hz, 1H, 7″-H), 6.98 (t, *J* = 7.6 Hz, 1H, 5″-H), 7.14 (t, *J* = 9.2 Hz, 6″-H), 7.24-7.37 (m, 6H, 4′-Ar, 4″-H, 4-NH), 7.59 (t, *J* = 7.3 Hz, 1H, 4′-Ar), 10.72 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 27.97, 31.33 (1,3-NCH_3), 34.98 (1′-NCH_3), 44.99 (C-4′), 54.28 (C-5′), 63.93, 65.98 (C-3a, C-9a), 68.25 (C-3′), 79.52 (C-2′), 109.99 (C-7″), 114.83 (d, *J* = 22.1 Hz, 4′-Ar-3), 122.23, 123.23, 124.55, 127.24, 129.85, 130.40 (4′-Ar-5,6, C-3a″, C-4″, C-5″, C-6″), 124.81 (d, *J* = 14.4 Hz, 4′-Ar-1), 129.42 (d, *J* = 8.6 Hz, 4′-Ar-4), 134.83 (5a-C=N), 143.88 (C-7a″), 159.18 (2-C=O), 161.32 (d, *J* = 242.9 Hz, C-F), 173.30 (8-C=O), 176.51 (2″-C=O); IR (KBr), ν 3369 (NH), 3161, 3092, 3015 (Ar), 2947, 2867, 2842, 2794 (Alk), 1721, 1698, 1651, 1618, 1599 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{25}H_{24}FN_7O_3S$ [*M* + Na]$^+$: 544.1534, Found: 544.1538.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-4′-(4-Fluorophenyl)-1,1′,3-trimethyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7c).** Yield 201 mg (77%) as a light gray powder. Mp: 270-272 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.07 (s, 3H, 1′-NCH_3), 2.44 (s, 3H, NCH_3), 2.76 (s, 3H, NCH_3), 3.40 (t, *J* = 9.0 Hz, 1H, 5′-H), 3.84 (t, *J* = 9.2 Hz, 1H, 5′-H), 4.33 (t, *J* = 8.4 Hz, 1H, 4′-H), 4.47 (d, *J* = 4.5 Hz, 1H, 3a-H), 5.36 (d, *J* = 5.6 Hz, 1H, 9a-H), 6.81 (d, *J* = 7.5 Hz, 1H, 7″-H), 6.96 (t, *J* = 7.3 Hz, 1H, 5″-H), 7.14-7.41 (m, 7H, 4-NH, 4″-H, 6″-H, 4′-Ar); ^{13}C NMR (75 MHz, DMSO-d_6): δ 27.96, 31.46 (1,3-CH_3), 34.82 (1′-NCH_3), 52.64 (C-4′), 57.56 (C-5′), 64.23, 65.99 (C-3a, C-9a), 69.13 (C-3′), 79.58 (C-2′), 109.97 (C-7″), 115.12 (d, *J* = 21.2 Hz, 4′-Ar-3,5), 122.21, 123.50, 127.36, 130.31 (C-3a″, C-4″, C-5″, C-6″), 132.18 (d, *J* = 8.1 Hz, 4′-Ar-2,6), 134.14 (d, *J* = 2.8 Hz, 4′-Ar-1), 134.68 (5a-C=N), 143.76 (C-7a″), 159.19 (2-C=O), 161.57 (d, *J* = 243.8 Hz, C-F), 173.18 (8-

C=O), 176.69 (2″-C=O); IR (KBr), ν 3436, 3400, 3334 (NH), 3146, 3090, 3018 (Ar), 2980, 2947, 2901, 2874, 2851, 2816, 2797 (Alk), 1728, 1703, 1691, 1683, 1649, 1619, 1606 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{25}H_{24}FN_7O_3S$ [*M* + Na]$^+$: 544.1531, Found: 544.1538.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-4′-(4-Bromophenyl)-1,1′,3-trimethyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7d).** Yield 262 mg (90%) as a white powder. Mp: 242-244 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.08 (s, 3H, 1′-NCH_3), 2.45 (s, 3H, NCH_3), 2.76 (s, 3H, NCH_3), 3.44 (t, *J* = 8.1 Hz, 1H, 5′-H), 3.85 (t, *J* = 9.4 Hz, 1H, 5′-H), 4.34 (t, *J* = 9.7 Hz, 1H, 4′-H), 4.48 (dd, *J* = 5.9, 2.1 Hz, 1H, 3a-H), 5.36 (d, *J* = 5.8 Hz, 1H, 9a-H), 6.81 (d, *J* = 7.7 Hz, 1H, 7″-H), 6.97 (t, *J* = 7.6 Hz, 1H, 5″-H), 7.22-7.35 (m, 5H, 4-NH, 4′-Ar-2,6, 4″-H, 6″-H), 7.53 (d, *J* = 8.4 Hz, 2H, 4′-Ar-3,5), 10.57 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 27.84, 31.30 (1,3-NCH_3), 34.71 (1′-NCH_3), 52.66 (C-4′), 57.11 (C-5′), 64.09, 65.89 (C-3a, C-9a), 68.82 (C-3′), 79.46 (C-2′), 109.82 (C-7″), 120.74, 122.13, 123.31, 127.29, 130.21, 131.13, 132.29, 134.36, 137.24 (4′-Ar-1-6, C-3a″, C-4″, C-5″, C-6″, 5a-C=N), 143.50 (C-7a″), 159.01 (2-C=O), 172.91 (8-C=O), 176.45 (2″-C=O); IR (KBr), ν 3297, 3212 (NH), 3099, 3058, 3037 (Ar), 2944, 2913, 2860 (Alk), 1713, 1688, 1660, 1621 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{25}H_{24}BrN_7O_3S$ [*M* + H]$^+$: 582.0900, Found: 582.0917.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-1,1′,3-Trimethyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7e).** Yield 258 mg (94%) as a light yellow powder. Mp: 269-272 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.07 (s, 3H, 1′-NCH_3), 2.41 (s, 3H, NCH_3), 2.80 (s, 3H, NCH_3), 3.50 (t, *J* = 8.2 Hz, 1H, 5′-H), 3.88 (t, *J* = 9.3 Hz, 1H, 5′-H), 4.48 (t, *J* = 9.1 Hz, 2H, 4′-H, 7″-H), 5.41 (d, *J* = 5.7 Hz, 1H, 3a-H), 6.82 (d, *J* = 7.6 Hz, 1H, 9a-H), 6.98 (t, *J* = 7.5, 1H, 5″-H), 7.24-7.39 (m, 3H, 4-NH, 4″-H, 6″-H), 7.69 (d, *J* = 8.5 Hz, 2H, 4′-Ar-2,6), 8.22 (d, *J* = 8.4 Hz, 2H, 4′-Ar-3,5), 10.81 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 27.89, 31.37 (1,3-NCH_3), 34.76 (1′-NCH_3), 52.74 (C-4′), 57.04 (C-5′), 64.10, 66.01 (C-3a, C-9a), 68.46 (C-3′), 79.54 (C-2′), 109.96 (C-7″), 122.27, 123.10, 123.27, 127.32, 130.37, 131.56 (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″), 134,01, 143.54, 145.74, 146.81 (4′-Ar-1,4, C-7a″, 5a-C=N), 159.06 (2-C=O), 172.64 (8-C=O), 176.48 (2″-C=O); IR (KBr), ν 3367, 3295 (NH), 3107, 3074, 3061 (Ar), 2957 (Alk), 1731, 1701, 1687 (C=O, C=N), 1515, 1345 (NO_2); HRMS (ESI): Exact mass calcd for $C_{25}H_{24}N_8O_5S$ [*M* + H]$^+$: 549.1657, Found: 549.1663.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-1″-Allyl-1,1′,3-trimethyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2′,8(1″*H*,3*H*)-trione (7f).** Yield 259 mg (88%) as a light yellow powder. Mp: 162-164 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.05 (s, 3H, 1′-NCH_3), 2.42 (s, 3H, NCH_3), 2.80 (s, 3H, NCH_3), 3.55 (t, *J* = 8.0 Hz, 1H, 5′-H), 3.89 (t, *J* = 9.2 Hz, 1H, 5′-H), 4.27-4.54 (m, 4H, 1″-NCH_2, 3a-H, 4′-H), 5.14-5.21 (m, 2H, =CH_2), 5.39 (d, *J* = 5.7 Hz, 1H, 9a-H), 5.76-5.88 (m, 1H, =CH), 6.97 (d, *J* = 7.7 Hz, 1H, 7″-H), 7.07 (t, *J* = 7.3 Hz, 1H, 5″-H), 7.32-7.38 (m, 3H, 4″-H,6″-H, 4-NH), 7.72 (d, *J* = 8.3 Hz, 2H, 4′-Ar-2,6), 8.23 (d, *J* = 8.3 Hz, 2H, 4′-Ar-3,5); ^{13}C NMR (75 MHz, DMSO-d_6): δ 27.95, 31.46 (1,3-NCH_3), 34.80 (1′-NCH_3), 41.38 (1″-NCH_2), 52.62 (C-4′), 57.23 (C-5′), 63.96, 66.13, 68,55 (C-3a, C-9a, C-3′), 79.33 (C-2′), 109.61 (C-7″), 117.23, 122.36, 123.04, 123.33, 127.06, 130.53, 131.51, 131.65, 133.70 (4′-Ar-2,3,5,6, CH=CH_2, C-3a″, C-4″, C-5″, C-6″, 5a-C=N), 143.95, 145.70, 146.87 (4′-Ar-1,4, C-7a″), 159.13 (2-C=O), 172.59 (8-C=O), 174.45 (2″-C=O); IR (KBr), ν 3413, 3402 (NH), 3084, 3059, 3051 (Ar), 2945, 2929 (Alk), 1701, 1655, 1609 (C=O, C=N), 1517, 1348 (NO_2); HRMS (ESI): Exact mass calcd for $C_{28}H_{28}N_8O_5S$ [*M* + H]$^+$: 589.1975, Found: 589.1976.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-5″-Bromo-1″-ethyl-1,1′,3-trimethyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2′,8(1″*H*,3*H*)-trione (7g).** Yield 210 mg (64%) as a light yellow powder. Mp: 227-229 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 1.15 (t, *J* = 6.4 Hz, 3H, 1″-CH_3), 2.07 (s, 3H, 1′-NCH_3), 2.52 (s, 3H, NCH_3), 2.79 (s, 3H, NCH_3), 3.54-3.93 (m, 4H, 5′-H, 1″-NCH_2), 4.46-4.52 (m, 2H, 3a-H, 4′-H), 5.47 (d, *J* = 5.2 Hz, 1H, 9a-H), 7.14 (d, *J* = 8.7 Hz, 1H, 7″-H), 7.36 (s, 1H, 4″-H), 7.42 (s, 1H, 4-NH), 7.64 (d, *J* = 8.3 Hz, 1H, 6″-H), 7.71 (d, *J* = 8.7 Hz, 2H, 4′-Ar-2,6), 8.26 (d, *J* = 8.7 Hz, 2H, 4′-Ar-3,5); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.53 (1″-CH_3), 27.86, 31.09, 34.31, 34.68 (1, 1′,3-NCH_3, 1″-NCH_2), 53.23 (C-4′), 57.12, 57.83, 63.50 (C-3a, C-5′, C-9a), 66.19 (C-3′), 69.22 (C-2′), 78.77 (C-7″), 111.33, 114.90, 123.31, 125.35, 130.38, 131.58, 133.58 (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″, 5a-C=N), 143.43, 145.23, 146.92 (4′-Ar-1,4, C-7a″), 158.91 (2-C=O), 172.38 (8-C=O), 174.00 (2″-C=O); IR (KBr), ν 3436, 3401, 3340 (NH), 3077, 3065 (Ar), 2975, 2947 (Alk), 1730, 1706, 1659, 1605 (C=O, C=N), 1521, 1346 (NO_2); HRMS (ESI): Exact mass calcd for $C_{27}H_{27}BrN_8O_5S$ [*M* + H]$^+$: 655.1075, Found: 655.1081.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-4′-(2,4-Diclorophenyl)-1,1′,3-trimethyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7h).** Yield 212 mg (74%) as a white powder. Mp: 274-276 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.07 (s, 3H, 1′-NCH_3), 2.47 (s, 3H, NCH_3), 2.83 (s, 3H, NCH_3), 3.46 (t, *J* = 8.0 Hz, 1H, 5′-H), 3.97 (t, *J* = 9.2 Hz, 1H, 5′-H), 4.40 (d, *J* = 4.9 Hz, 1H, 3a-H), 4.69 (t, *J* = 8.7 Hz, 1H, 4′-H), 5.34 (d, *J* = 5.5 Hz, 1H, 9a-H), 6.82 (d, *J* = 7.6 Hz, 1H, 7″-H), 6.99 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.25-7.31 (m, 2H, 4″-H, 6″-H), 7.37 (d, *J* = 1.9 Hz, 1H, 4-NH), 7.54 (d, *J* = 8.5 Hz, 1H, 4′-Ar-5), 7.62 (d, *J* = 2.1 Hz, 1H, 4′-Ar-3), 7.78 (d, *J* = 8.6 Hz, 1H, 4′-Ar-6), 10.78 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 27.93, 31.21 (1,3-NCH_3), 34.21 (1′-NCH_3), 48.06 (C-4′), 55.16 (C-5′), 63.34, 66.20 (C-3a, C-9a), 67.80 (C-3′), 79.37 (C-2′), 110.05 (C-7″), 122.28, 122.84, 127.13, 127.44, 128.52, 130.48, 132.09, 132.76, 134.54, 134.87, 136.25 (4′-Ar-1-6, C-3a″, C-4″, C-5″, C-6″, 5a-C=N), 143.74 (C-7a″), 159.12 (2-C=O), 173.16 (8-C=O), 176.32 (2″-C=O); IR (KBr), ν 3442, 3330 (NH), 3090, 3020 (Ar), 2979, 2946, 2899, 2874 (Alk), 1730, 1702, 1690, 1650 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{25}H_{23}Cl_2N_7O_3S$ [*M* + H]$^+$: 572.1029, Found: 572.1033.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-1,3-Diethyl-1′-methyl-4′-phenyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7i).** Yield 200 mg (63%) as a white powder. Mp: 206-208 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.87 (t, *J* = 6.9 Hz, 3H, CH_3), 1.03 (, *J* = 6.6 Hz t, 3H, CH_3), 2.06 (s, 3H, 1′-NCH_3), 2.79-2.94 (m, 2H, NCH_2), 3.05-3.12 (m, 1H, NCH_2), 3.26-3.33 (m, 1H, NCH_2), 3.42 (t, *J* = 8.1 Hz, 1H, 5′-H), 3.88 (t, *J* = 9.3 Hz, 1H, 5′-H), 4.35 (t, *J* = 8.7, 1H, 4′-H), 4.49 (d, *J* = 3.9 Hz, 1H, 3a-H), 5.48 (d, *J* = 4.8 Hz, 1H, 9a-H), 6.79 (d, *J* = 7.8 Hz, 1H, 7″-H), 6.99 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.25-7.36 (m, 8H, 4″-H, 6″-H, 4-NH, Ph), 10.71 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 14.37, 14.53 (2CH_3), 36.56, 36.76 (1,3-NCH_2), 38.83 (1′-NCH_3), 55.37 (C-4′), 59.11 (C-3′), 62.97 (C-2′), 64.91 (C-5′), 71.21, 81.57 (C-3a, C-9a), 111.74 (7″-H), 124.20, 125.56, 129.23, 129.42, 130.31, 132.01, 132.31, 136.41 (Ph, C-3a″, C-4″, C-5″, C-6″), 139.74 (5a-C=N), 145.55 (C-7a″), 159.68 (2-C=O), 175.30 (8-C=O), 178.53 (2″-C=O); IR (KBr), ν 3255 (NH), 3064, 3029 (Ar), 2942, 2868, 2742 (Alk), 1719, 1650, 1670 (C=O, C=N); HRMS (ESI): Exact mass calcd for $C_{27}H_{29}N_7O_3S$ [*M* + H]$^+$: 532.2116, Found: 532.2125.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-1,3-Diethyl-4′-(2-fluorophenyl)-1′-methyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7j).** Yield 187 mg (54%) as a white powder. Mp: 244-246 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.91 (t, *J* = 7.0 Hz, 3H, CH_3), 1.05 (t, *J* = 6.9 Hz, 3H, CH_3), 2.06 (s, 3H, 1′-NCH_3), 2.81-2.96 (m, 2H, NCH_2), 3.07-3.16 (m, 1H, NCH_2), 3.26-3.35 (m, 3H, NCH_2), 4.09 (t, *J* =9.3 Hz, 1H, 5′-H), 4.44 (d, *J* = 4.5 Hz, 1H, 3a-H), 4.57 (t, *J* = 8.8 Hz, 1H, 4′-H), 4.90 (d, *J* = 5.4 Hz, 1H, 9a-H), 6.73 (d, *J* = 7.6 Hz, 1H, 7″-H), 6.83 (t, *J* = 7.6 Hz, 1H, 5″-H), 7.09-7.37 (m, 7H, 4′-Ar, 4″-H, 6″-H, 4-NH), 7.64 (t, *J* = 7.3, 1H, 4′-Ar); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.48, 12.55 ($2CH_3$), 34.67, 36.89 (1,3-NCH_2), 34.93 (1′-NCH_3), 45.03 (C-4′), 54.24 (C-5′), 61.02, 63.09 (C-3a, C-9a), 68.25 (C-3′), 79.49 (C-2′), 109.91 (C-7″), 114.79 (d, *J* = 22.2 Hz, 4′-Ar-3), 122.18, 123.27, 124.54, 127.07, 129.81, 130.30 (Ar-5,6, C-3a″, C-4″, C-5″, C-6″), 124.77 (d, *J* = 14.5 Hz, 4′-Ar-1), 129.35 (d, *J* = 8.1 Hz, 4′-Ar-4), 134.53 (5a-C=N), 144.04 (C-7a″), 157.69 (2-C=O), 161.28 (d, *J* = 242.8 Hz, C-F), 173.34 (8-C=O), 176.54 (2″-C=O); IR (KBr), ν 3422, 3301, 3250 (NH), 2974, 2950, 2873 (Alk), 1712, 1689, 1654 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{27}H_{28}FN_7O_3S$ [*M* + H]$^+$: 550.2028, Found: 550.2031.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-1,3-Diethyl-4′-(4-fluorophenyl)-1′-methyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7k).** Yield 198 mg (72%) as a white powder. Mp: 177-179 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.87 (t, *J* = 6.9 Hz, 3H, CH_3), 1.03 (t, *J* = 6.8 Hz, 3H, CH_3), 2.06 (s, 3H, 1′-NCH_3), 2.79-2.95 (m, 2H, NCH_2), 3.05-3.12 (m, 1H, NCH_2), 3.27-3.33 (m, 1H, NCH_2), 3.43 (t, *J* = 9.0 Hz, 1H, 5′-H), 3.82 (t, *J* = 9.5 Hz, 1H, 5′-H), 4.35 (t, *J* = 8.7 Hz, 1H, 4′-H), 4.50 (d, *J* = 4.8 Hz, 1H, 3a-H), 5.47 (d, *J* = 5.5 Hz, 1H, 9a-H), 6.80 (d, *J* = 7.6 Hz, 1H, 7″-H), 6.98 (t, *J* = 7.6 Hz, 1H, 5″-H), 7.15-7.30 (m, 5H, 4′-Ar, 4″-H, 6″-H, 4-NH), 7.41 (t, *J* = 6.8 Hz, 2H, 4′-Ar), 10.73 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.43, 12.57 ($2CH_3$), 34.62, 34.77 (1,3-NCH_2), 36.90 (1′-NCH_3), 52.64 (C-4′), 57.39 (C-5′), 61.11, 63.02 (C-3a, C-9a), 69.22 (C-3′), 79.54 (C-2′), 109.85 (C-7″), 115.06 (d, *J* = 21.2 Hz, 4′-Ar-3,5), 122.28, 123.53, 127.28, 130.28 (C-3a″, C-4″, C-5″, C-6″), 132.08 (d, *J* = 8.2 Hz, 4′-Ar-2,6), 134.02 (Ar-1), 134.27 (5a-C=N), 143.57 (C-7a″), 157.73 (2-C=O), 161.53 (d, *J* = 243.9 Hz, C-F), 173.18 (8-C=O), 176.61 (2″-C=O); IR (KBr), ν 3458, 3321, 3262 (NH), 3064, 3048, 3011 (Ar), 2977, 2944, 2876 (Alk), 1716, 1671, 1661, 1622 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{27}H_{28}FN_7O_3S$ [*M* + Na]$^+$: 572.1848, Found: 572.1851.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-4′-(4-Bromophenyl)-1,3-diethyl-1′-methyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7l).** Yield 244 mg (80%) as a white powder. Mp: 198-201 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.87 (t, *J* = 6.6 Hz, 3H, CH_3), 1.03 (t, *J* = 6.5 Hz, 3H, CH_3), 2.05 (s, 3H, 1′-NCH_3), 2.77-2.96 (m, 2H, NCH_2), 3.02-3.12 (m, 1H, NCH_2), 3.26-3.35 (m, 1H, NCH_2), 3.43 (t, *J* = 7.5 Hz, 1H, 5′-H), 3.81 (t, *J* = 9.2 Hz, 1H, 5′-H), 4.32 (t, *J* = 8.4 Hz, 1H, 4′-H), 4.50 (d, *J* = 5.2 Hz, 1H, 3a-H), 5.47 (d, *J* = 5.1 Hz, 1H, 9a-H), 6.80 (d, *J* = 7.4 Hz, 1H, 7″-H), 6.98 (t, *J* = 7.3 Hz, 1H, 5″-H), 7.23-7.34 (m, 5H, 4-NH, 4′-Ar-2,6, 4″-H, 6″-H), 7.55 (d, *J* = 7.9 Hz, 2H, 4′-Ar-3,5), 10.76 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.43, 12.57 ($2CH_3$), 34.62, 34.78 (1,3-NCH_2), 36.88 (1′-NCH_3), 52.78 (C-4′), 57.09 (C-5′), 61.05, 63.03 (C-3a, C-9a), 68.99 (C-3′), 79.53 (C-2′), 109.88 (C-7″), 120.82, 122.30, 123.45, 127.29, 130.32, 131.22, 132.33, 134.17, 137.26 (Ar-2-6, C-3a″, C-4″, C-5″, C-6″, 5a-C=N), 143.58 (C-7a″), 157.73 (2-C=O), 173.08 (8-C=O), 176.56 (2″-C=O); IR (KBr), ν 3418, 3189 (NH), 3080 (Ar), 2926, 2878 (Alk), 1711, 1652, 1628 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{27}H_{28}BrN_7O_3S$ [*M* + H]$^+$: 610.1228, Found: 610.1230.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-1,3-Diethyl-1′-methyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7m).** Yield 265 mg (92%) as a light yellow powder. Mp: 233-234 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.87 (t, *J* = 7.0 Hz, 3H, CH_3), 1.04 (t, *J* = 6.9 Hz, 3H, CH_3), 2.07 (s, 3H, 1′-NCH_3), 2.79-2.96 (m, 1H, NCH_2), 3.04-3.11 (m, 1H, NCH_2), 3.26-3.35 (m, 1H, NCH_2), 3.47-3.52 (m, 2H, NCH_2, 5′-H), 3.89 (t, *J* = 9.5 Hz, 1H, 5′-H), 4.46-4.53 (m, 2H, 3a-H, 4′-H), 5.48 (d, *J* = 5.5 Hz, 1H, 9a-H), 6.81 (d, *J* = 7.6 Hz, 1H, 7″-H), 6.99 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.24-7.33 (m, 3H, 4″-H, 6″-H, 4-NH), 7.68 (d, *J* = 8.5 Hz, 2H, 4′-Ar-2,6), 8.22 (d, *J* = 7.6 Hz, 2H, 4′-Ar-3,5); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.42, 12.57 ($2CH_3$), 34.62, 34.79, 36.89, (1,3-NCH_2, 1′-NCH_3), 52.82 (C-4′), 56.94 (C-5′), 61.04, 63.11 (C-3a, C-9a), 68.61 (C-3′), 79.56 (C-2′), 109.96 (C-7″), 122.38, 123.21, 123.31, 127.28, 130.42, 131.55 (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″), 135.75, 143.59, 145.73, 146.80 (4′-Ar-1,4, C-7a″, 5a-C=N), 157.70 (2-C=O), 172.76 (8-C=O), 176.55 (2″-C=O); IR (KBr), ν 3436, 3324, 3257 (NH), 3109, 3050 (Ar), 2974, 2944 (Alk), 1713, 1666, 1622, 1599 (C=O, C=N), 1518, 1341 (NO_2); HRMS (ESI): Exact mass calcd for $C_{27}H_{28}N_8O_5S$ [*M* + H]$^+$: 577.1972, Found: 577.1976.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-1″-Allyl-1,3-diethyl-1′-methyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2′,8(1″*H*,3*H*)-trione (7n).** Yield 280 mg (91%) as light brown crystals. Mp: 164-166 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.85 (t, *J* = 7.1 Hz, 3H, CH_3), 1.05 (t, *J* = 7.1 Hz, 3H, CH_3), 2.06 (s, 3H, 1′-NCH_3), 2.80-2.87 (m, 1H, NCH_2), 2.93-3.09 (m, 2H, NCH_2), 3.33-3.39 (m, 1H, NCH_2), 3.59 (t, *J* = 8.3 Hz, 1H, 5′-H), 3.91 (t, *J* = 9.2 Hz, 1H, 5′-H), 4.25 (dd, *J* = 17.6, 4.5 Hz, 1H, 1″-NCH_2), 4.38 (dd, *J* = 17.2, 4.7 Hz, 1H, 1″-NCH_2), 4.51-4.56 (m, 2H, 3a-H, 4′-H), 5.12-5.21 (m, 2H, $=CH_2$), 5.78 (d, *J* = 5.4 Hz, 1H, 9a-H), 5.77-5.86 (m, 1H, =CH), 6.95 (d, *J* = 8.0 Hz, 1H, 7″-H), 7.08 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.32-7.36 (m, 3H, 4-NH, 4″-H, 6″-H), 7.71 (d, *J* = 8.6 Hz, 2H, 4′-Ar-2,6), 8.23 (d, *J* = 8.5 Hz, 2H, 4′-Ar-3,5); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.40, 12.66 ($2CH_3$), 34.57, 34.73, 36.97 (1,3-NCH_2, 1′-NCH_3), 41.26 (1″-NCH_2), 52.57 (C-4′), 57.11 (C-5′), 61.20, 63.17 (C-3a, C-9a), 68.56 (C-3′), 79.35 (C-2′), 109.50 (C-7″), 117.04, 122.37, 123.10, 123.27, 126.88, 130.44, 131.40, 131.52, 133.34 (4′-Ar-2,3,5,6, $CH=CH_2$, C-3a″, C-4″, C-5″, C-6″, 5a-C=N), 143.89, 145.63, 146.83 (4′-Ar-1,4, C-7a″), 157.62 (2-C=O), 172.60 (8-C=O), 174.49 (2″-C=O); IR (KBr), ν 3437, 3266 (NH), 3084, 3063, 3051 (Ar), 2978, 2931, 2899, 2872 (Alk), 1720, 1706, 1655 (C=O, C=N), 1515, 1352 (NO_2); HRMS (ESI): Exact mass calcd for $C_{30}H_{32}N_8O_5S$ [*M* + H]$^+$: 617.2284, Found: 617.2289.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-5″-Bromo-1,1″,3-triethyl-1′-methyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7o).** Yield 243 mg (71%) as a light beige powder. Mp: 199-201 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.92 (t, *J* = 7.1 Hz, 3H, CH_3), 1.08-1.15 (m, 6H, $2CH_3$), 2.05 (s, 3H, 1′-NCH_3), 2.76-2.91 (m, 2H, NCH_2), 3.12-3.19 (m, 1H, NCH_2), 3.32-3.41 (m, 1H, NCH_2), 3.54 (t, *J* = 8.1 Hz, 1H, 5′-H), 3.62-3.81 (m, 2H, NCH_2), 3.89 (t, *J* = 9.4 Hz, 1H, 5′-H), 4.45-4.54 (m, 2H, 4′-H, 3a-H), 5.53 (d, *J* = 5.4 Hz, 1H, 9a-H), 7.12 (d, *J* = 8.2 Hz, 1H, 7″-H), 7.33 (s, 1H, 4″-H), 7.51 (s, 1H, 4-NH), 7.62 (d, *J* = 8.4 Hz, 1H, 6″-H), 7.68 (d, *J* = 8.5 Hz, 2H, 4′-Ar-2,6), 8.22 (d, *J* = 8.5 Hz, 2H, 4′-Ar-3,5); ^{13}C NMR (75 MHz, DMSO-d_6): δ 11.92, 12.46, 12.73 ($3CH_3$), 34.25, 34.66, 36.69 (1,3-NCH_2 1′-NCH_3), 40.06 (1″-NCH_2), 53.27 (4′-C), 57.01 (5′-C), 60.40, 63.27 (C-3a, C-9a), 69.33 (C-3′), 78.74 (C-2′), 111.29 (7″-H), 114.99 (C-5″), 123.29 (4′-Ar-3,5), 125.42, 130.28, 131.50, 133.39, 133.59 (4′-Ar-2,6, C-3a″, C-4″, C-6″, 5a-C=N), 143.43, 145.16, 146.90 (4′-Ar-1,4,

C-7a″), 157.57 (2-C=O), 172.56 (8-C=O), 174.05 (2″-C=O); IR (KBr), ν 3436, 3280 (NH), 3107, 3078, 3052 (Ar), 2972, 2936, 2874, 2860 (Alk), 1702, 1656, 1605 (C=O, C=N), 1521, 1425 (NO_2); HRMS (ESI): Exact mass calcd for $C_{29}H_{31}BrN_8O_5S$ [*M* + H]$^+$: 683.1387, Found: 683.1394.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-1,3-Diethyl-4′-(2,4-diclorophenyl)-1′-methyl-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7p).** Yield 150 mg (50%) as a white powder. Mp: 229-231 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.91 (t, *J* = 6.9 Hz, 3H, CH_3), 1.08 (t, *J* = 6.9 Hz, 3H, CH_3), 2.08 (s, 3H, 1′-NCH_3), 2.83-2.92 (m, 1H, NCH_2), 2.95-3.04 (m, 1H, NCH_2), 3.06-3.16 (m, 1H, NCH_2), 3.32-3.39 (m, 1H, NCH_2), 3.46 (t, *J* = 9.0 Hz, 1H, 5′-H), 3.98 (t, *J* = 9.0 Hz, 1H, 5′-H), 4.43 (d, *J* = 6.2 Hz, 1H, 3a-H), 4.69 (t, *J* = 8.8 Hz, 1H, 4′-H), 5.44 (d, *J* = 6.4 Hz, 1H, 9a-H), 6.81 (d, *J* = 7.7 Hz, 1H, 7″-H), 6.99 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.25-7.30 (m, 3H, 4-NH, 4″-H, 6″-H), 7.54 (d, *J* = 8.4 Hz, 1H, 4′-Ar-5), 7.62 (s, 1H, 4′-Ar-3), 7.78 (d, *J* = 8.6 Hz, 1H, 4′-Ar-6), 10.76 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.29, 12.62 ($2CH_3$), 34.72, 34.84 (1,3-NCH_2), 36.80 (1′-NCH_3), 48.10 (C-4′), 55.13 (C-5′), 60.52, 63.31 (C-3a, C-9a), 67.83 (C-3′), 79.34 (C-2′), 109.95 (C-7″), 122.32, 122.86, 127.00, 127.35, 128.45, 130.40, 132.04, 132.70, 134.24, 134.84, 136.70 (Ar-1-6, C-3a″, C-4″, C-5″, C-6″, 5a-C=N), 143.70 (C-7a″), 157.70 (2-C=O), 173.18 (8-C=O), 176.30 (2″-C=O); IR (KBr), ν 3222 (NH), 2974, 2942, 2873 (Alk), 1717, 1699, 1653, 1620 (C=N, C=O); HRMS (ESI): Exact mass calcd for $C_{27}H_{27}Cl_2N_7O_3S$ [*M* + H]$^+$: 600.1327, Found: 600.1346.

(2′*R,3a*R**,3′*R**,4′*R**,9a*R**)-4′-(4-Fluorophenyl)-1,1′,3-trimethyl-2-thioxo-1,2,3,3a,4,9a-hexahydroimidazo[4,5-e]thiazolo[2,3-c][1,2,4]triazin-7,3′-pyrrolidine-2′,3″-indole)-2″,8(1″*H*)-dione (7q).** Yield 215 mg (80%) as a white powder. Mp: 242-244 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.06 (s, 3H, 1′-NCH_3), 2.74 (s, 3H, NCH_3), 3.00 (s, 3H, NCH_3), 3.43 (t, *J* = 8.1 Hz, 1H, 5′-H), 3.81 (t, *J* = 9.4 Hz, 1H, 5′-H), 4.32 (t, *J* = 8.4 Hz, 1H, 3a-H), 4.80 (d, *J* = 4.8 Hz, 1H, 4′-H), 5.64 (d, *J* = 6.0 Hz, 1H, 9a-H), 6.80 (d, *J* = 7.5 Hz, 1H, 7″-H), 7.02 (d, *J* = 7.4 Hz, 1H, 5″-H), 7.15-7.40 (m, 6H, 4′-Ar, 4″-H, 6″-H, 4-NH), 7.53 (s, 1H, 4′-Ar), 10.76 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 31.43, 34.73 (1,3-NCH_3), 35.01 (1′-NCH_3), 52.98 (C-4′), 57.29 (C-5′), 66.96, 67.21 (C-3a, C-9a), 69.30 (C-3′), 79.39 (C-2′), 109.93 (C-7″), 115.02 (d, *J* = 21.1 Hz, 4′-Ar-3,5), 122.51, 123.40, 127.51, 130.33 (C-3a″, C-4″, C-5″, C-6″), 132.06 (d, *J* = 8.2 Hz, 4′-Ar-2,6), 133.79 (d, *J* = 3.0 Hz, 4′-Ar-1), 135.09 (5a-C=N), 143.52 (C-7a″), 161.52 (d, *J* = 243.8 Hz, C-F), 172.71 (8-C=O), 176.50 (2″-C=O), 184.07 (2-C=S); IR (KBr), ν 3327, 3219 (NH), 3096 (ArH), 2959, 2923, 2866, 2795 (Alk), 1709, 1657, 1616 (C=O, C=N); HRMS (ESI): Exact mass calcd for $C_{25}H_{24}FN_7O_2S_2$ [M + Na]$^+$: 560.1303, Found: 560.1309.

(2′*R,3a*R**,3′*R**,4′*R**,9a*R**)-1,3-Diethyl-4′-(4-fluorophenyl)-1′-methyl-2-thioxo-1,2,3,3a,4,9a-hexahydroimidazo[4,5-e]thiazolo[2,3-c][1,2,4]triazin-7,3′-pyrrolidine-2′,3″-indole)-2″,8(1″*H*)-dione (7r).** Yield 248 mg (92%) as a white powder. Mp: 255-257 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.95 (t, *J* = 7.0 Hz, 3H, CH_3), 1.07 (t, *J* = 6.8 Hz, 3H, CH_3), 2.06 (s, 3H, 1′-NCH_3), 2.92-3.02 (m, 1H, NCH_2), 3.06-3.17 (m, 1H, NCH_2), 3.44 (t, *J* = 8.3 Hz, 1H, 5′-H), 3.58-3.85 (m, 3H, NCH_2, 5′-H), 4.34 (t, *J* = 8.8 Hz, 1H, 4′-H), 4.74 (d, *J* = 4.1 Hz, 1H, 3a-H), 5.68 (d, *J* = 5.8 Hz, 1H, 9a-H), 6.81 (d, *J* = 7.7 Hz, 1H, 7″-H), 7.04 (t, *J* = 7.6 Hz, 1H, 5″-H), 7.18 (t, *J* = 8.7 Hz, 2H, 4′-Ar), 7.25-7.44 (m, 5H, 4′-Ar, 4″-H, 6″-H, 4-NH), 7.41 (t, *J* = 6.8 Hz, 2H, 4′-Ar), 10.79 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 11.44, 12.08 ($2CH_3$), 34.76, 38.26 (1,3-NCH_2), 39.67 (1′-NCH_3), 53.03 (C-4′), 57.26 (C-5′), 63.39, 64.77 (C-3a, C-9a), 69.47 (C-3′), 79.50 (C-2′), 109.98 (C-7″), 115.08 (d, *J* = 21.0 Hz, 4′-Ar-3,5), 122.53, 123.55, 127.42, 130.46 (C-3a″, C-4″, C-5″, C-6″), 132.07 (d, *J*

= 8.0 Hz, 4′-Ar-2,6), 133.80 (d, *J* = 2.9 Hz, Ar-1), 134.47 (5a-C=N), 143.62 (C-7a″), 161.55 (d, *J* = 243.7 Hz, C-F), 172.94 (8-C=O), 176.60 (2″-C=O), 181.86 (2-C=S); IR (KBr), ν 3408, 3327 (NH), 3173, 3094 (Ar), 2974, 2945, 2874, 2846, 2796 (Alk), 1713, 1649, 1616 (C=O, C=N); HRMS (ESI): Exact mass calcd for $C_{27}H_{28}FN_7O_2S_2$ $[M + Na]^+$: 588.1611, Found: 588.1622.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-1′-Ethyl-1,3-dimethyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7s).** Yield 270 mg (96%) as a white powder. Mp: 247-248 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.95 (t, *J* = 7.0 Hz, 3H, CH_3), 2.13-2.19 (m, 1H, NCH_2), 2.28-2.34 (m, 1H, NCH_2), 2.42 (s, 3H, NCH_3), 2.78 (s, 3H, NCH_3), 3.66 (t, *J* = 8.2 Hz, 1H, 5′-H), 3.76 (t, *J* = 9.1 Hz, 1H, 5′-H), 4.46-4.51 (m, 2H, 3a-H, 4′-H), 5.38 (d, *J* = 5.8 Hz, 1H, 9a-H), 6.80 (d, *J* = 7.7 Hz, 1H, 7″-H), 6.98 (t, *J* = 7.5 Hz, 1H, 5″-H), 7.23-7.36 (m, 3H, 4″-H, 6″-H, 4-NH), 7.68 (d, *J* = 8.4 Hz, 2H, Ar-2,6), 8.22 (d, *J* = 8.4 Hz, 2H, Ar-3,5), 10.77 (s, 1H, 1″-NH); ^{13}C NMR (125 MHz, DMSO-d_6): δ 13.70 (CH_3), 27.96, 31.41 (1,3-NCH_3), 42.64 (1′-NCH_2), 52.68 (C-4′), 54.11 (C-5′), 64.15, 66.08 (C-3a, C-9a), 68.46 (C-3′), 79.64 (C-2′), 109.95 (C-7″), 122.33, 123.31, 123.75, 127.42, 130.38, 131.65, (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″), 134.11 (5a-C=N), 143.62, 145.96, 146.88 (4′-Ar-1,4, C-7a″), 159.11 (2-C=O), 172.71 (8-C=O), 177.03 (2″-C=O); IR (KBr), ν 3376, 3206 (NH), 3068, 3014 (Ar), 2972, 2943, 2860, 2823 (Alk), 1718, 1702, 1656 (C=O, C=N), 1522, 1348 (NO_2); HRMS (ESI): Exact mass calcd for $C_{26}H_{26}N_8O_5S$ $[M + H]^+$: 563.1817, Found: 563.1820.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-1,1′,3-Triethyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7t).** Yield 242 mg (82%) as a white powder. Mp: 214-216 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ ^{1}H NMR (300 MHz, DMSO-d_6) δ 0.87 (t, *J* = 7.0 Hz, 3H, CH_3), 0.95 (t, *J* = 7.0 Hz, 3H, CH_3), 1.04 (t, *J* = 6.7 Hz, 3H, CH_3), 2.12-2.18 (m, 1H, NCH_2), 2.28-2.34 (m, 1H, NCH_2), 2.79-2.92 (m, 2H, NCH_2), 3.03-3.12 (m, 1H, NCH_2), 3.27-3.45 (m, 1H, NCH_2), 3.65 (t, *J* = 8.2 Hz, 1H, 5′-H), 3.77 (t, *J* = 9.2 Hz, 1H, 5′-H), 4.46-4.52 (m, 2H, 3a-H, 4′-H), 5.47 (d, *J* = 5.4 Hz, 1H, 9a-H), 6.80 (d, *J* = 7.7 Hz, 1H, 7″-H), 6.99 (t, *J* = 7.6 Hz, 1H, 5″-H), 7.23-7.30 (m, 3H, 4″-H, 6″-H, 4-NH), 7.68 (d, *J* = 8.4 Hz, 2H, Ar-2,6), 8.22 (d, *J* = 8.4 Hz, 2H, Ar-3,5), 10.77 (s, 1H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.315, 12.59, 13.65 ($3CH_3$), 34.60, 6.83 (1,3-NCH_2), 42.58 (1′-NCH_2), 52.68 (C-4′), 53.89 (C-5′), 60.95, 63.08 (C-3a, C-9a), 68.51 (C-3′), 79.58 (C-2′), 109.87 (C-7″), 122.35, 123.26, 123.78, 127.28, 130.35, 131.54, (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″), 133.76 (5a-C=N), 143.58, 145.85, 146.81 (4′-Ar-1,4, C-7a″), 157.67 (2-C=O), 172.75 (8-C=O), 176.99 (2″-C=O); IR (KBr), ν 3376, 3240, 3229 (NH), 3014, 3065 (Ar), 29.71, 2936, 2873, 2823 (Alk), 1720, 1701, 1655 (C=O, C=N), 1521, 1350 (NO_2); HRMS (ESI): Exact mass calcd for $C_{28}H_{30}N_8O_5S$ $[M + H]^+$: 591.2132, Found: 591.2133.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-7″-Chloro-1,1′,3-trimethyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7u).** Yield 259 mg (89%) as a light yellow powder. Mp: 193-195 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.09 (s, 3H, 1′-NCH_3), 2.41 (s, 3H, NCH_3), 2.81 (s, 3H, NCH_3), 3.53 (t, *J* = 8.3 Hz, 1H, 5′-H), 3.88 (t, *J* = 9.4 Hz, 1H, 5′-H), 4.47-4.53 (m, 2H, 3a-H, 4′-H), 5.40 (d, *J* = 5.9 Hz, 1H, 9a-H), 7.02 (t, *J* = 7.9 Hz, 1H, 5″-H), 7.27 (d, *J* = 7.5 Hz, 1H), 7.35 (d, *J* = 7.8 Hz, 1H), 7.43 (s, 1H, 4-NH), 7.68 (d, *J* = 8.5 Hz, 2H, 4′-Ar-2,6), 8.23 (d, *J* = 8.5 Hz, 2H, 4′-Ar-3,5), 11.27 (s, 1 H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 27.92, 31.53 (1,3-NCH_3), 34.83 (1′-NCH_3), 52.54 (C-4′), 57.02 (C-5′), 64.39, 66.10 (C-3a, C-9a), 68.50 (C-3′), 80.15 (C-2′), 113.98 (C-7″), 123.32, 123.60, 125.07, 125.91, 130.45, 131.54 (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-

6″), 133.63 (5a-C=N), 141.15, 145.52, 146.86 (4′-Ar-1,4, C-7a″), 159.10 (2-C=O), 172.32 (8-C=O), 176.63 (2″-C=O); IR (KBr), ν 3391 (NH), 2966, 2934, 2869, 2846 (Alk), 1712, 1654 (C=O, C=N), 1521, 1345 (NO_2); HRMS (ESI): Exact mass calcd for $C_{25}H_{23}ClN_8O_5S$ [*M* + H]$^+$: 583.1273, Found: 583.1259.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-1,1′,3,6″-Tetramethyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7v).** Yield 236 mg (84%) as a yellow powder. Mp: 238-240 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.07 (s, 3H, 1′-NCH_3), 2.26 (s, 3H, 6″-CH_3), 2.43 (s, 3H, NCH_3), 2.80 (s, 3H, NCH_3), 3.48 (t, *J* = 8.2 Hz, 1H, 5′-H), 3.86 (t, *J* = 9.3 Hz, 1H, 5′-H), 4.43-4.49 (m, 2H, 3a-H, 4′-H), 5.38 (d, *J* = 5.8 Hz, 1H, 9a-H), 6.63 (s, 1H, 7″-H), 6.79 (d, *J* = 7.8 Hz, 1H, 5″-H), 7.17 (d, *J* = 7.7 Hz, 1H, 4″-H), 7.36 (d, *J* = 1.6 Hz, 1H, 4-NH), 7.68 (d, *J* = 8.7 Hz, 2H, 4′-Ar-2,6), 8.22 (d, *J* = 8.6 Hz, 2H, 4′-Ar-3,5), 10.74 (s, 1 H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 21.33 (C-6″), 27.96, 31.45 (1,3-NCH_3), 34.79 (1′-NCH_3), 52.67 (C-4′), 57.10 (C-5′), 63.98, 66.12 (C-3a, C-9a), 68.48 (C-3′), 79.54 (C-2′), 110.71 (C-7″), 120.09, 122.97, 123.32, 127.08 (4′-Ar-3,5, C-3a″, C-4″, C-5″), 131.62 (4′-Ar-2,6), 134.17 (5a-C=N), 140.33 (C-6″), 143.64, 145.92, 146.85 (4′-Ar-1,4, C-7a″), 159.20 (2-C=O), 172.84 (8-C=O), 176.82 (2″-C=O); IR (KBr), ν 3371 (NH), 2959, 2933, 2876 (Alk), 1721, 1696, 1650, 1630 (C=O, C=N), 1524, 1361 (NO_2); HRMS (ESI): Exact mass calcd for $C_{26}H_{26}N_8O_5S$ [*M* + H]$^+$: 563.1820, Found: 563.1817.

(2′*R,3a*R**,3′*R**,4′*R**,9a*S**)-1,1′,3,7″-Tetramethyl-4′-(4-nitrophenyl)-1,3a,4,9a-tetrahydrodispiro[imidazo[4,5-*e*]thiazolo[2,3-*c*][1,2,4]triazine-7,3′-pyrrolidine-2′,3″-indole]-2,2″,8(1″*H*,3*H*)-trione (7w).** Yield 258 mg (92%) as a light yellow powder. Mp: 218-220 °C; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.07 (s, 3H, 1′-NCH_3), 2.19 (s, 3H, 7″-CH_3), 2.44 (s, 3H, NCH_3), 2.76 (s, 3H, NCH_3), 3.49 (t, *J* = 8.2 Hz, 1H, 5′-H), 3.90 (t, *J* = 9.3 Hz, 1H, 5′-H), 4.45-4.48 (m, 2H, 3a-H, 4′-H), 5.40 (d, *J* = 5.8 Hz, 1H, 9a-H), 6.90 (t, *J* = 7.6 Hz, 1H, 5″-H), 7.09 (d, *J* = 7.6 Hz, 1H), 7.17 (d, *J* = 7.3 Hz, 1H), 7.36 (s, 1H, 4-NH), 7.69 (d, *J* = 8.5 Hz, 2H, 4′-Ar-2,6), 8.23 (d, *J* = 8.5 Hz, 2H, 4′-Ar-3,5), 10.84 (s, 1 H, 1″-NH); ^{13}C NMR (75 MHz, DMSO-d_6): δ 16.37 (7″-CH_3), 27.96, 31.29 (1,3-NCH_3), 34.86 (1′-NCH_3), 52.96 (C-4′), 56.94 (C-5′), 63.83, 66.08 (C-3a, C-9a), 68.73 (C-3′), 79.64 (C-2′), 119.16, 122.25, 122.97, 123.39, 124.84, 131.63, 131.86 (4′-Ar-2,3,5,6, C-3a″, C-4″, C-5″, C-6″, C-7″), 134.20 (5a-C=N), 142.20, 145.81, 146.88 (4′-Ar-1,4, C-7a″), 159.14 (2-C=O), 172.74 (8-C=O), 177.04 (2″-C=O); IR (KBr), ν 3380 (NH), 3087 (Ar), 2946, 2865 (Alk), 1712, 1708, 1645 (C=O, C=N), 1528, 1341 (NO_2); HRMS (ESI): Exact mass calcd for $C_{26}H_{26}N_8O_5S$ [*M* + H]$^+$: 563.1820, Found: 563.1809.

X-ray Crystallography: Data collection for samples **6b**, **6g**, **3j** and **7a** were performed on a Bruker APEX DUO diffractometer equipped with Apex II CCD detector and graphite monochromator for MoKα radiation (λ = 0.71073 Å). Frames were integrated using the Bruker SAINT software package[4] by a narrow-frame algorithm. A semiempirical absorption correction was applied with the SADABS[5] program using the intensity data of equivalent reflections. The structures were solved with a dual-space method with SHELXT program[6] and refined by the full-matrix least-squares technique against F^2_{hkl} in anisotropic approximation for non-hydrogen atoms with SHELXL program.[7] Hydrogen atoms connected to heteroatoms were found from difference Fourier synthesis and refined isotropically. Other hydrogen atoms were placed in calculated positions and refined in the riding model with $U_{iso}(H) = 1.5U_{eq}(C_m)$ and $1.2U_{eq}(C_i)$, where $U_{eq}(C_m)$ and $U_{eq}(C_i)$ are respectively the equivalent thermal parameters of the methyl carbon and all other carbon atoms to which corresponding H atoms are bonded. For structures **6b** and **7a** a SQUEEZE method implemented in the PLATON program[8] was applied to model disordered solvent molecules. Fragments of molecules **6b** and **7a** were found to be disordered by two positions with refined relative occupancies 0.40:0.60 and 0.49:0.51, respectively.

Detailed crystallographic information is given in **Table S2**. CCDC 1981082-1981085 contain supplementary crystallographic data for this paper. The data can be obtained free of charge from The Cambridge Crystallographic Data Centre *via* www.ccdc.cam.ac.uk/structures.

[4] Bruker, *SAINT, v8.34A*, **2013**.

[5] L. Krause, R. Herbst-Irmer, G. M. Sheldrick, D. Stalke, *J. Appl. Cryst.* **2015**, *48*, 3–10.

[6] G. M. Sheldrick, *Acta Crystallogr., Sect. A: Found. Crystallogr.* 2015, *71*, 3–8.

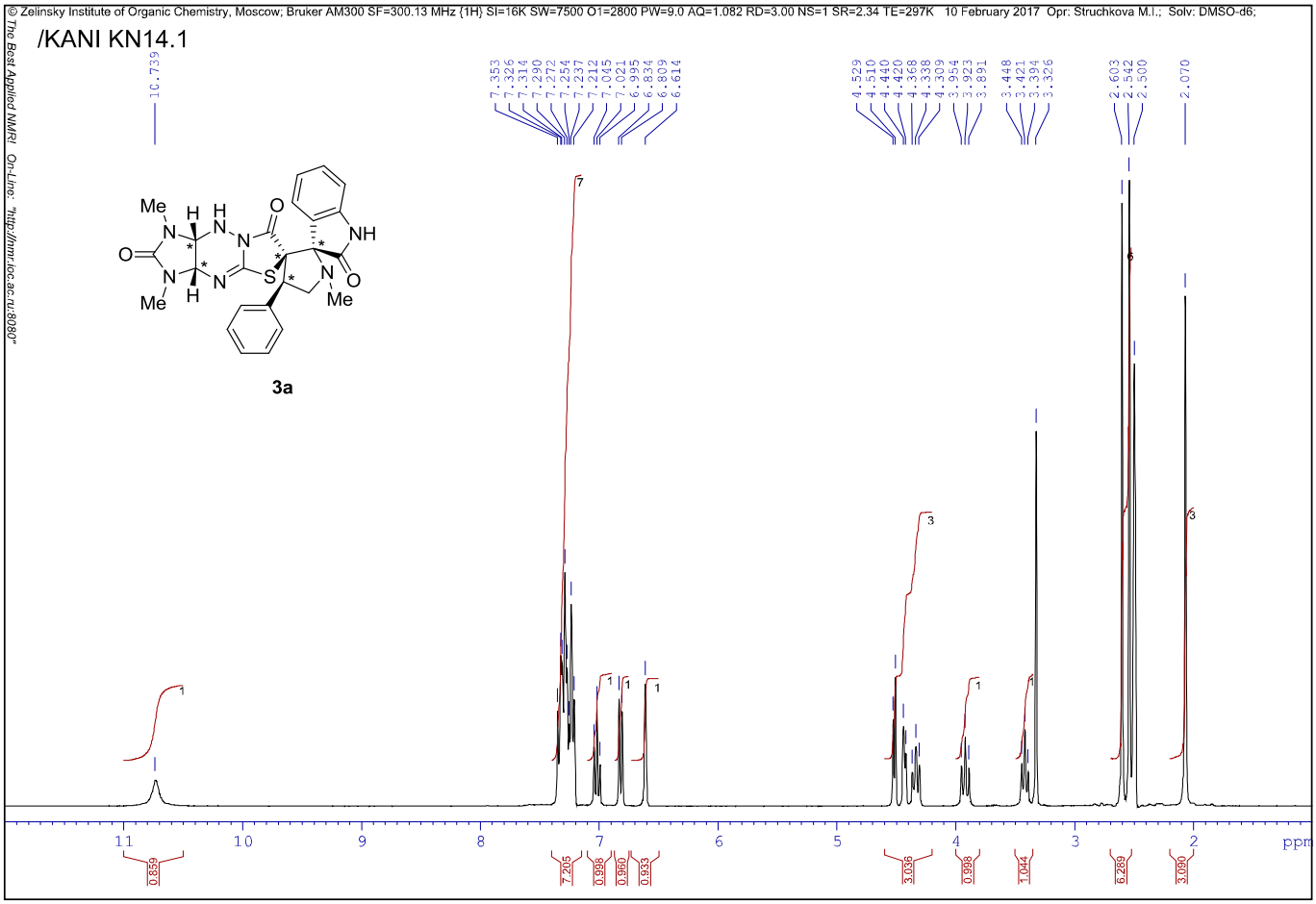
[7] G. M. Sheldrick, *Acta Crystallogr., Sect. C: Struct. Chem.* **2015**, *71*, 3–8.

[8] A. L. Spek, *Acta Crystallogr., Sect. C: Struct. Chem.* **2015**, *71*, 9–18.

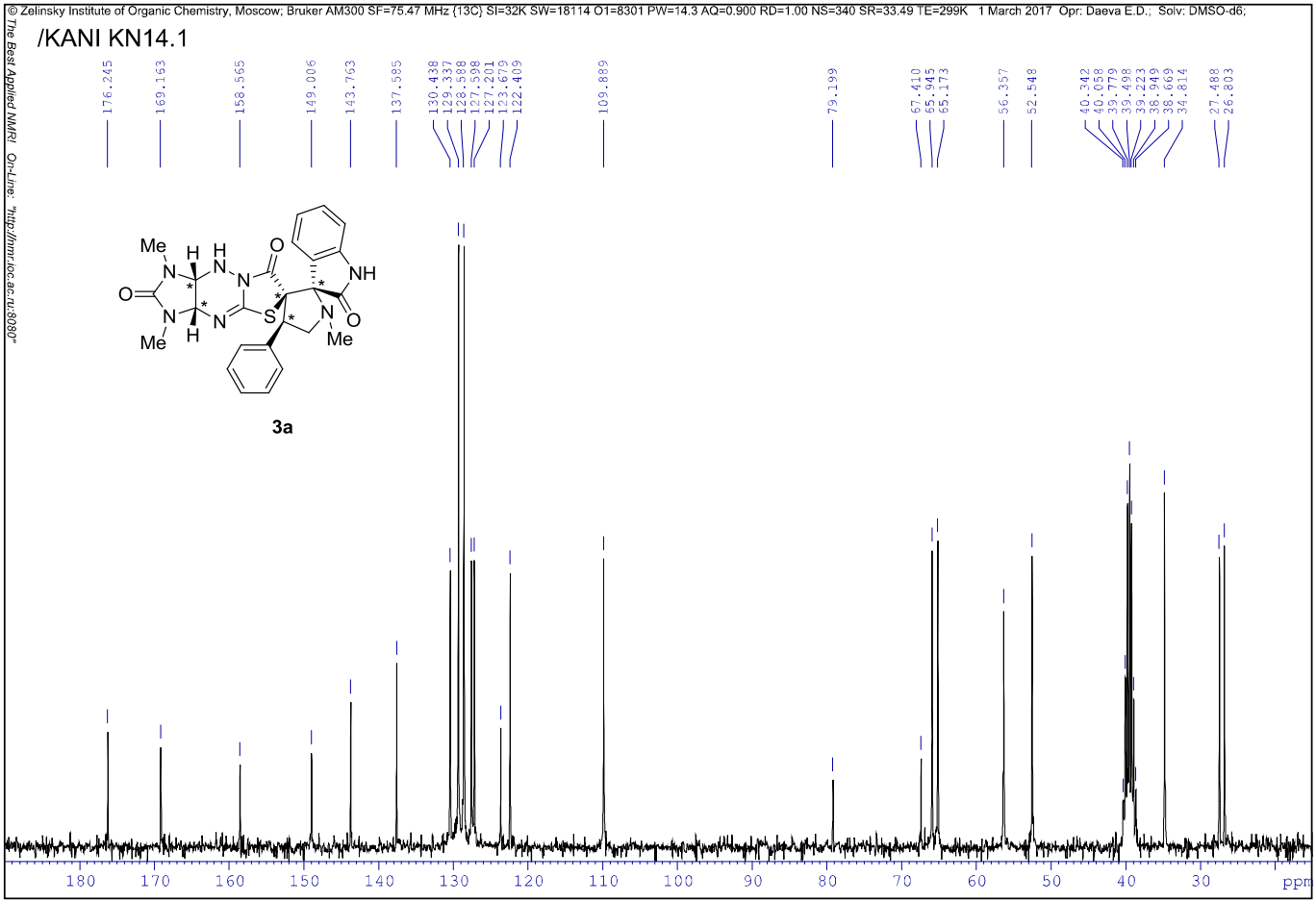
Table S2. Crystallographic data for **6b**, **6g**, **3j** and **7a**.

	6b	**6g**	**3j**	**7a**
CCDC	1981085	1981082	1981084	1981083
Empirical formula	$C_{27}H_{32}FN_7O_5S$	$C_{27}H_{27}BrN_8O_5S$	$C_{27.5}H_{31}FN_7O_4S$	$C_{25}H_{25}N_7O_3S$
Formula weight	585.65	655.53	574.65	503.58
T (K)	120	120	120	120
Crystal system	Monoclinic	Monoclinic	Triclinic	Orthorhombic
Space group	$P2_1/c$	$P2_1/n$	P-1	Pbca
Z / Z'	8 / 2	4 / 1	4 / 2	8 / 1
a, Å	17.9516(12)	9.7202(14)	13.4741(4)	9.7574(4)
b, Å	16.7643(11)	11.6843(16)	13.4934(4)	16.3229(7)
c, Å	19.6754(13)	24.726(4)	15.7758(5)	33.1283(14)
α, °	90	90	84.2195(7)	90
β, °	112.9590(10)	93.705(3)	85.3686(7)	90
γ, °	90	90	76.9008(7)	90
V, Å^3	5452.2(6)	2802.3(7)	2774.41(15)	5276.3(4)
d_{calc} (g cm^{-1})	1.427	1.554	1.376	1.268
μ, cm^{-1}	1.78	15.96	1.71	1.62
F(000)	2464	1344	1208	2112
$2\theta_{max}$, °	61.4	60.0	61.1	54.0
Refls collected	124818	48478	38708	53117
Independent refls [Rint]	16896 [0.0464]	8165 [0.0525]	16974 [0.0285]	5767 [0.0339]
Observed refls [$I > 2\sigma(I)$]	12892	6265	13265	5437
Parameters	799	387	765	355
*R*1	0.0504	0.0482	0.0423	0.0693
w*R*2	0.1372	0.1344	0.1124	0.2006
GOF	1.026	1.026	1.018	1.054
Residual density, $\Delta\rho_{max}$/ $\Delta\rho_{min}$ (e A^{-3})	0.578/-0.704	0.918/-0.534	0.454/-0.512	0.445/-0.534

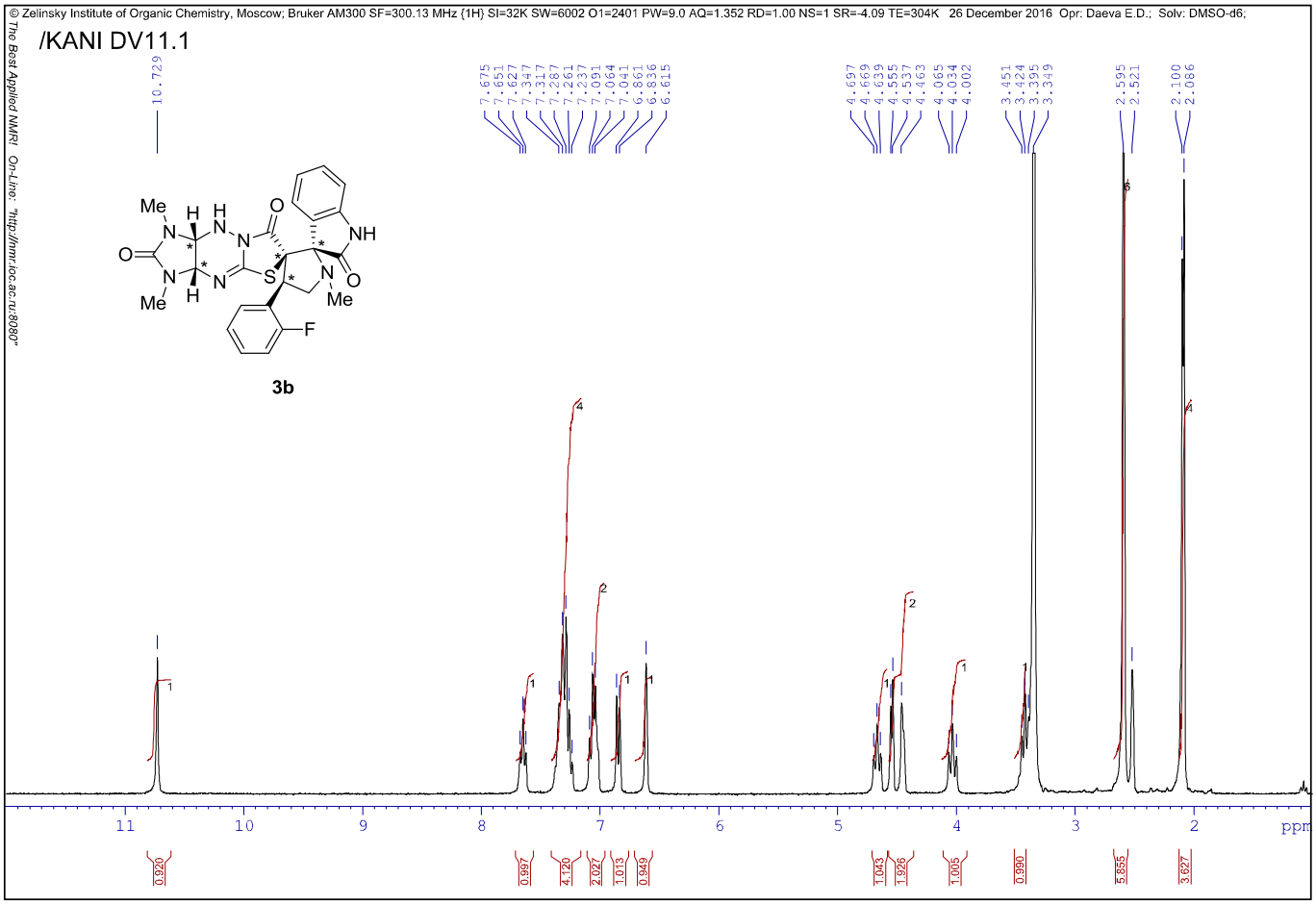
^{1}H NMR spectrum of **3a**



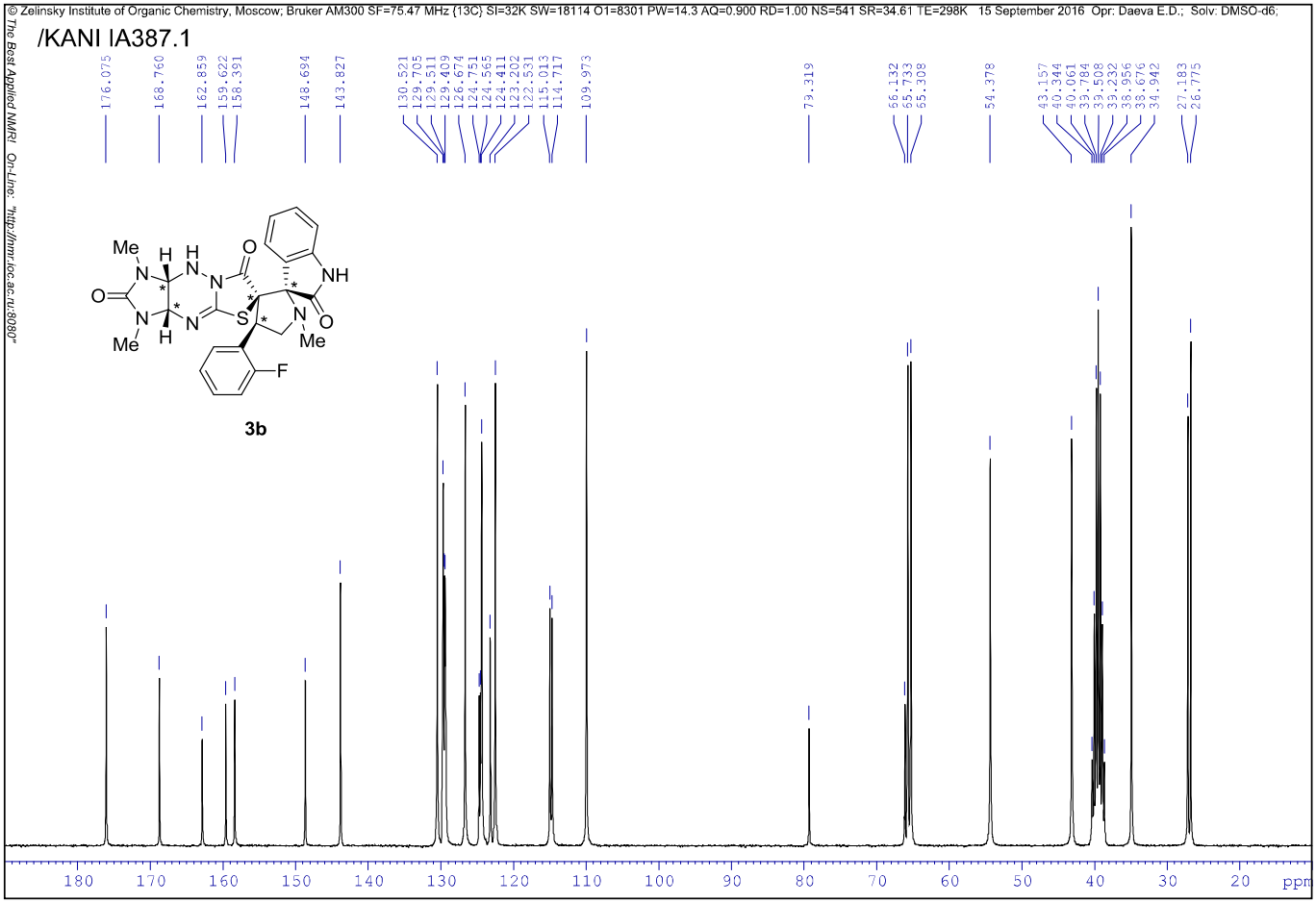
^{13}C NMR spectrum of **3a**



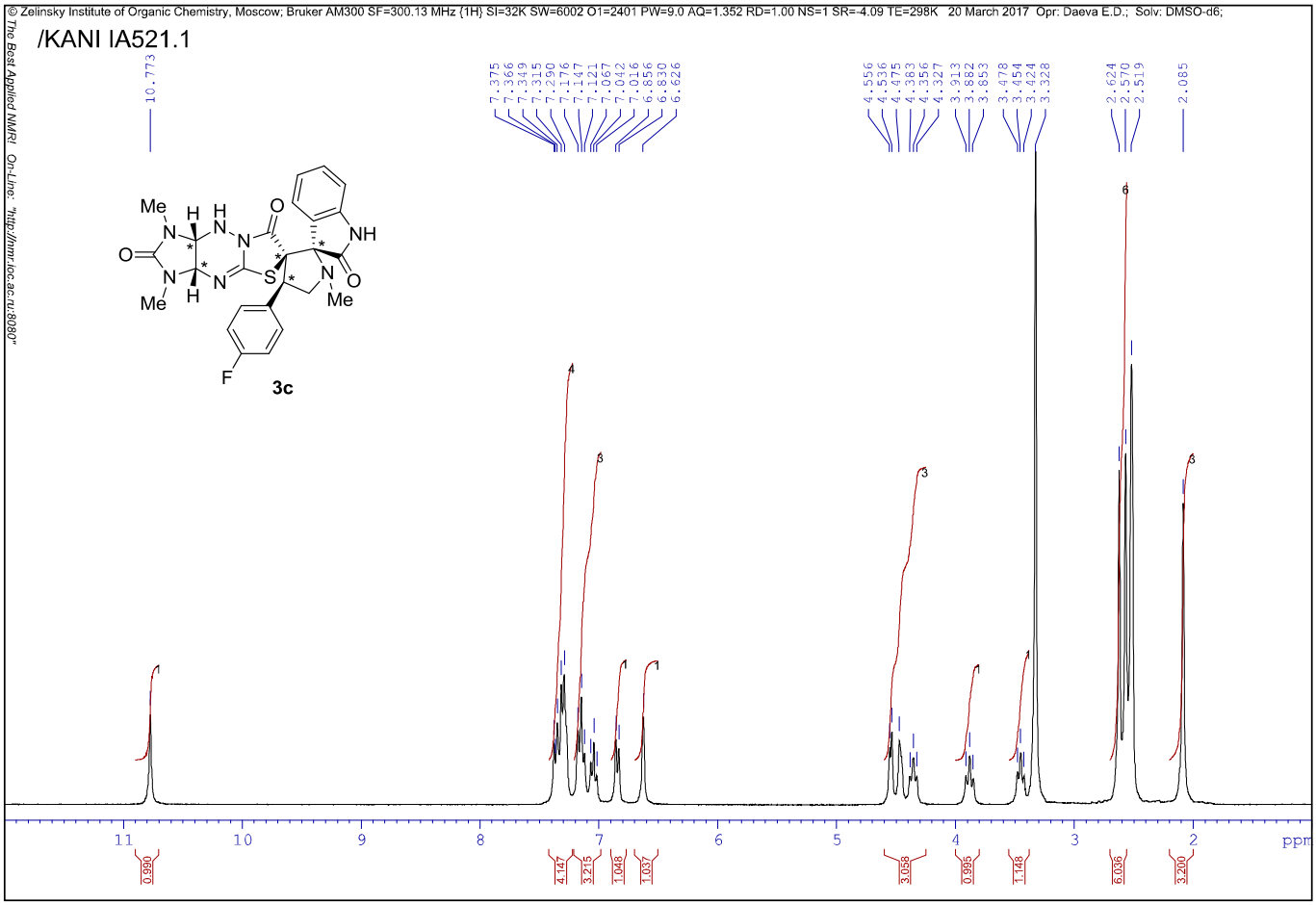
^{1}H NMR spectrum of **3b**



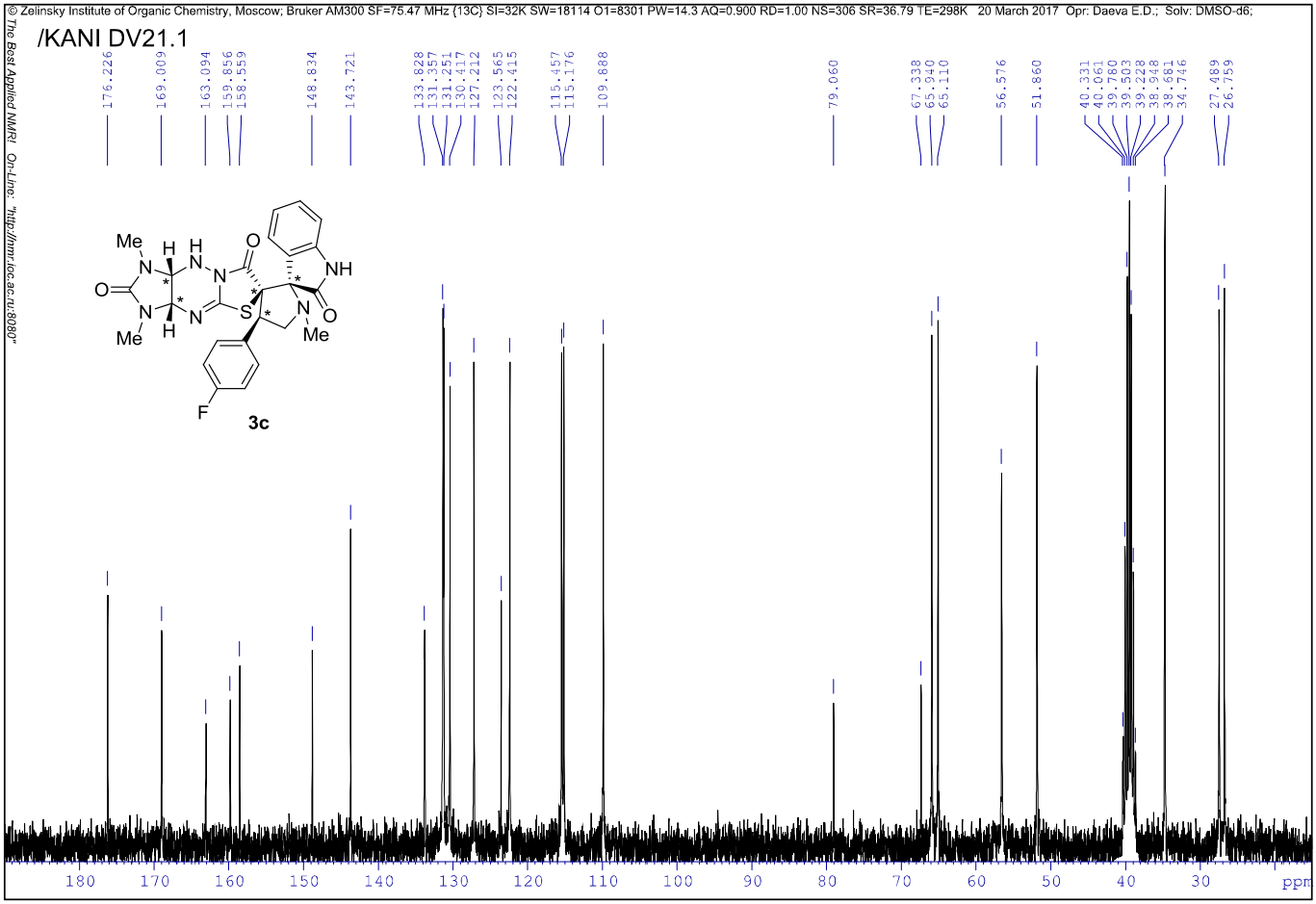
^{13}C NMR spectrum of **3b**



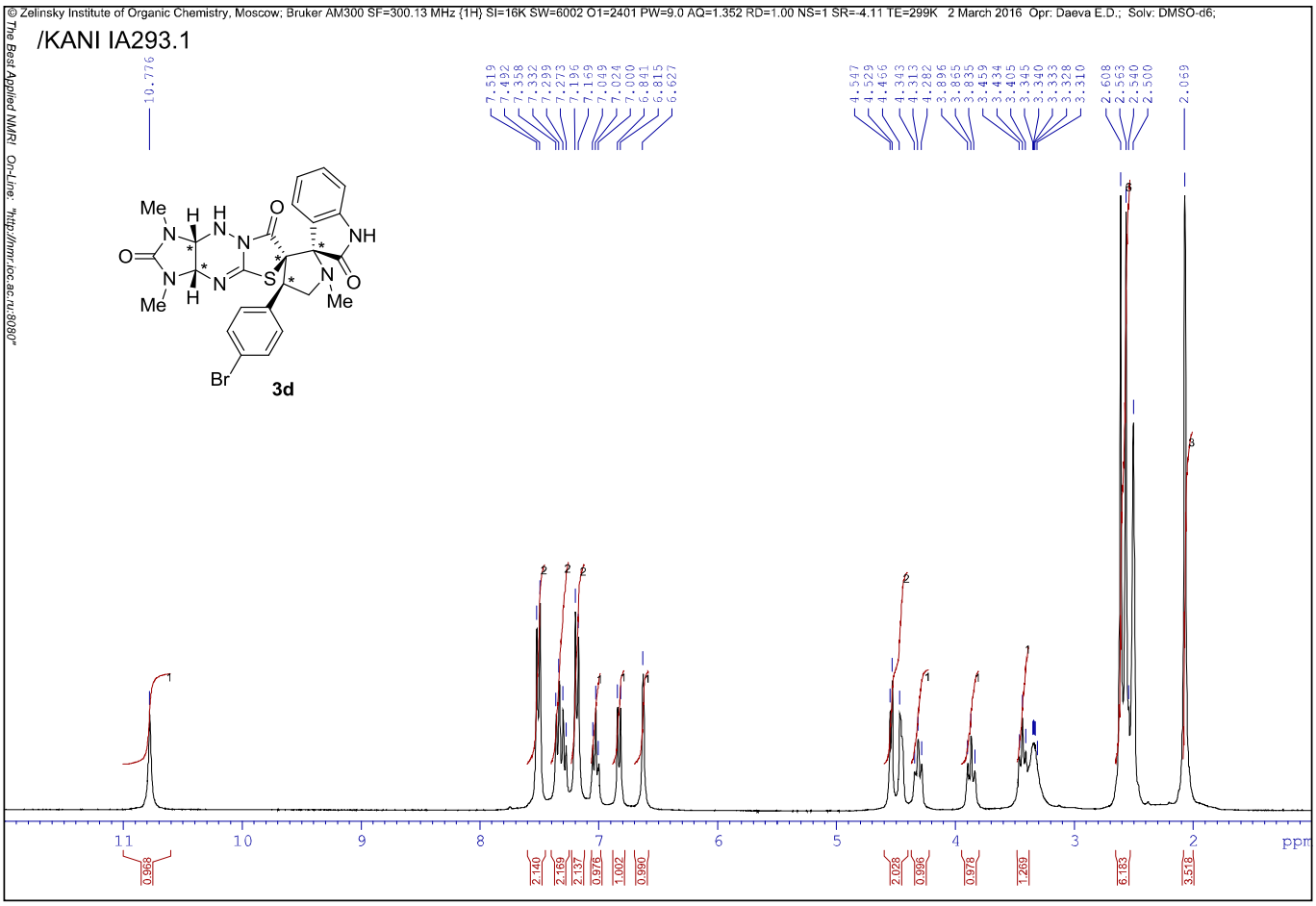
^{1}H NMR spectrum of **3c**



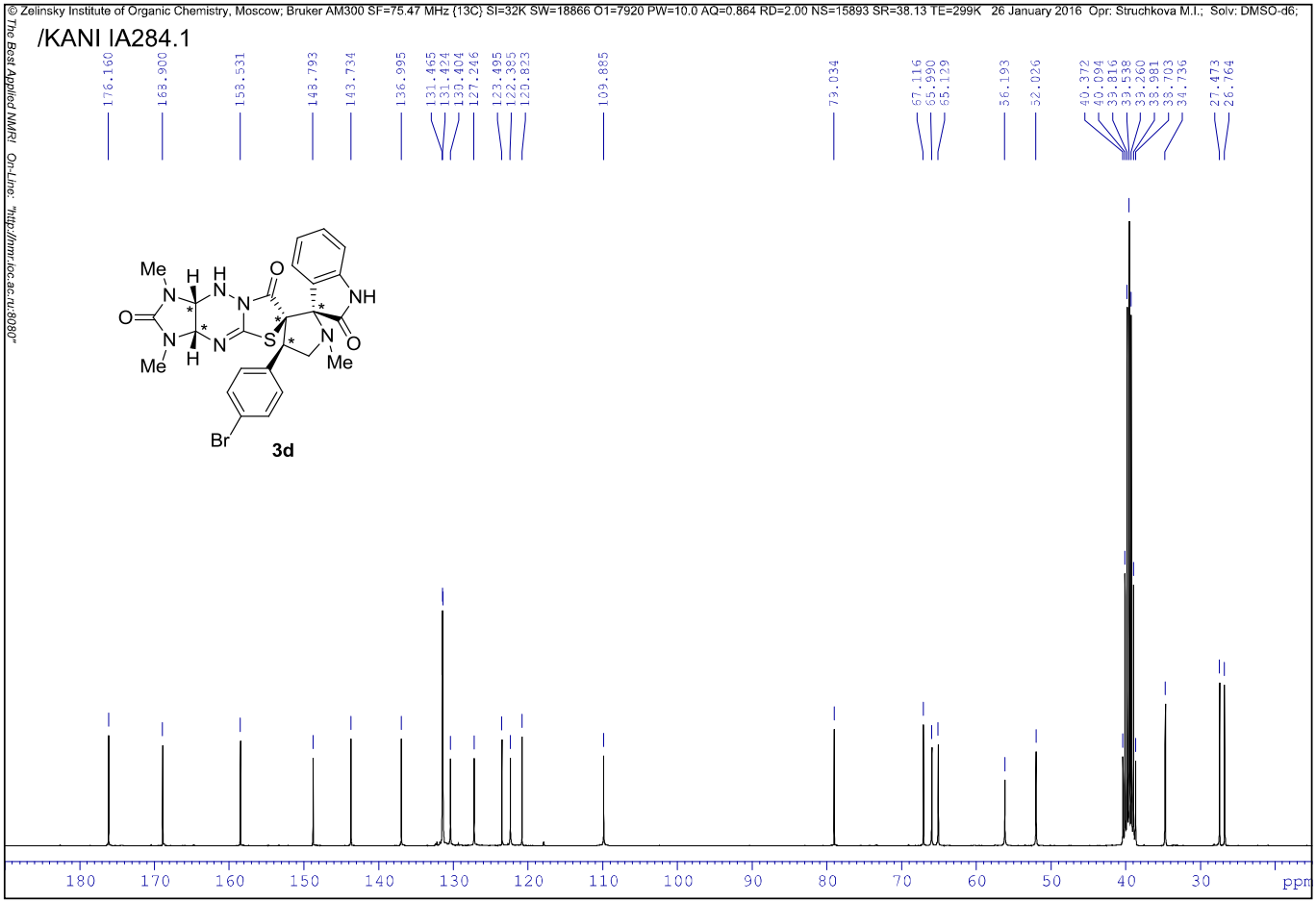
^{13}C NMR spectrum of **3c**



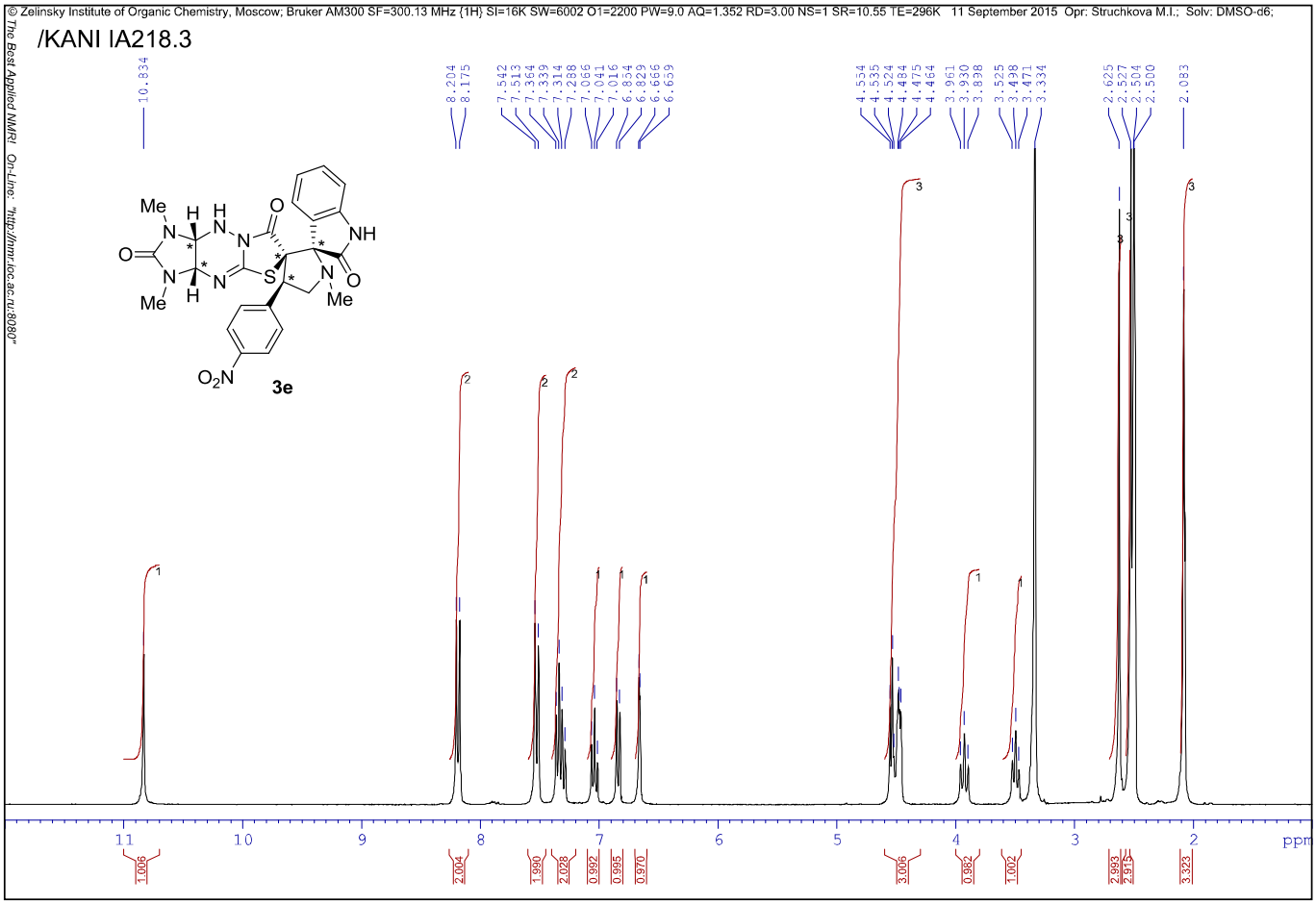
^{1}H NMR spectrum of **3d**



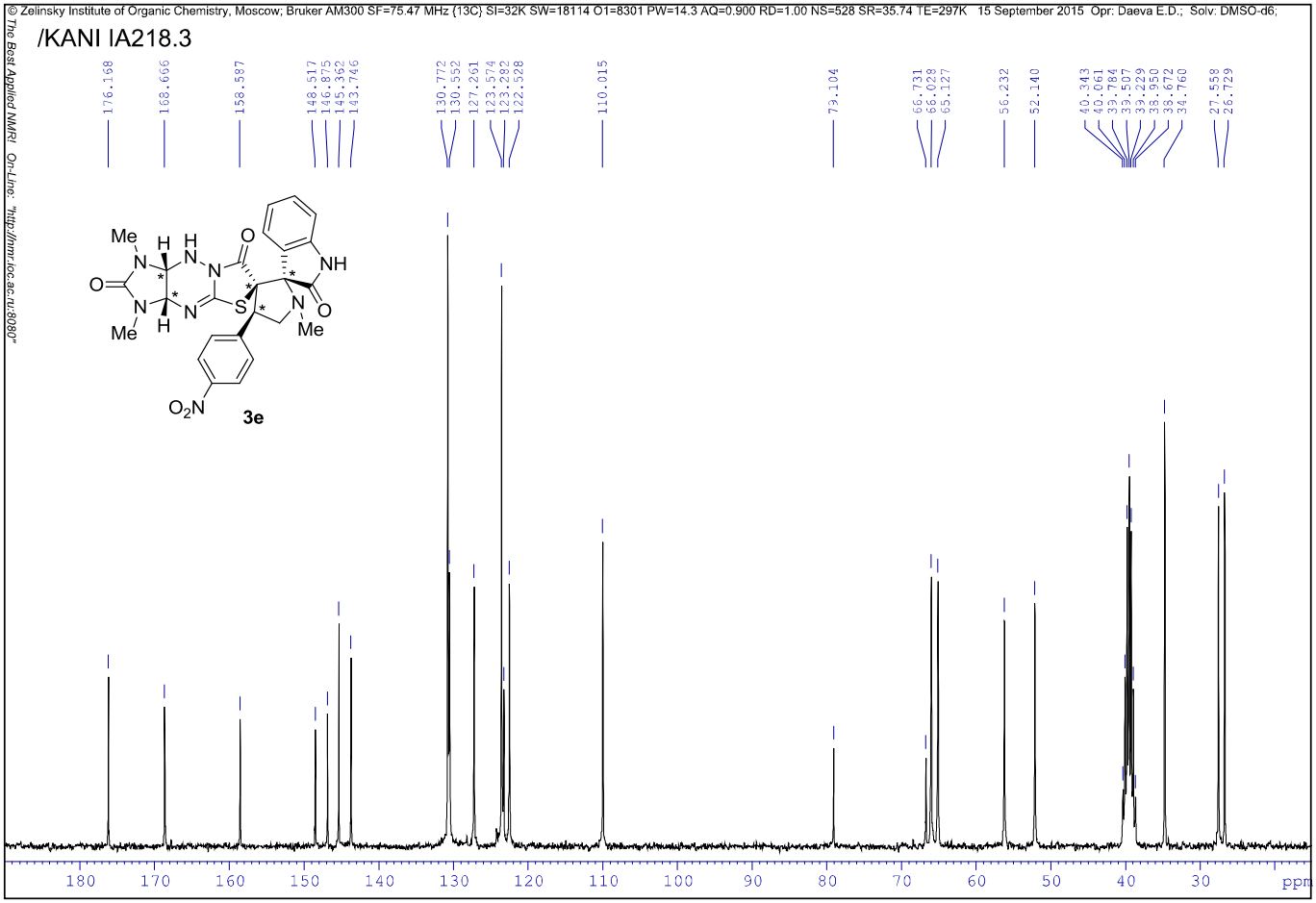
^{13}C NMR spectrum of **3d**



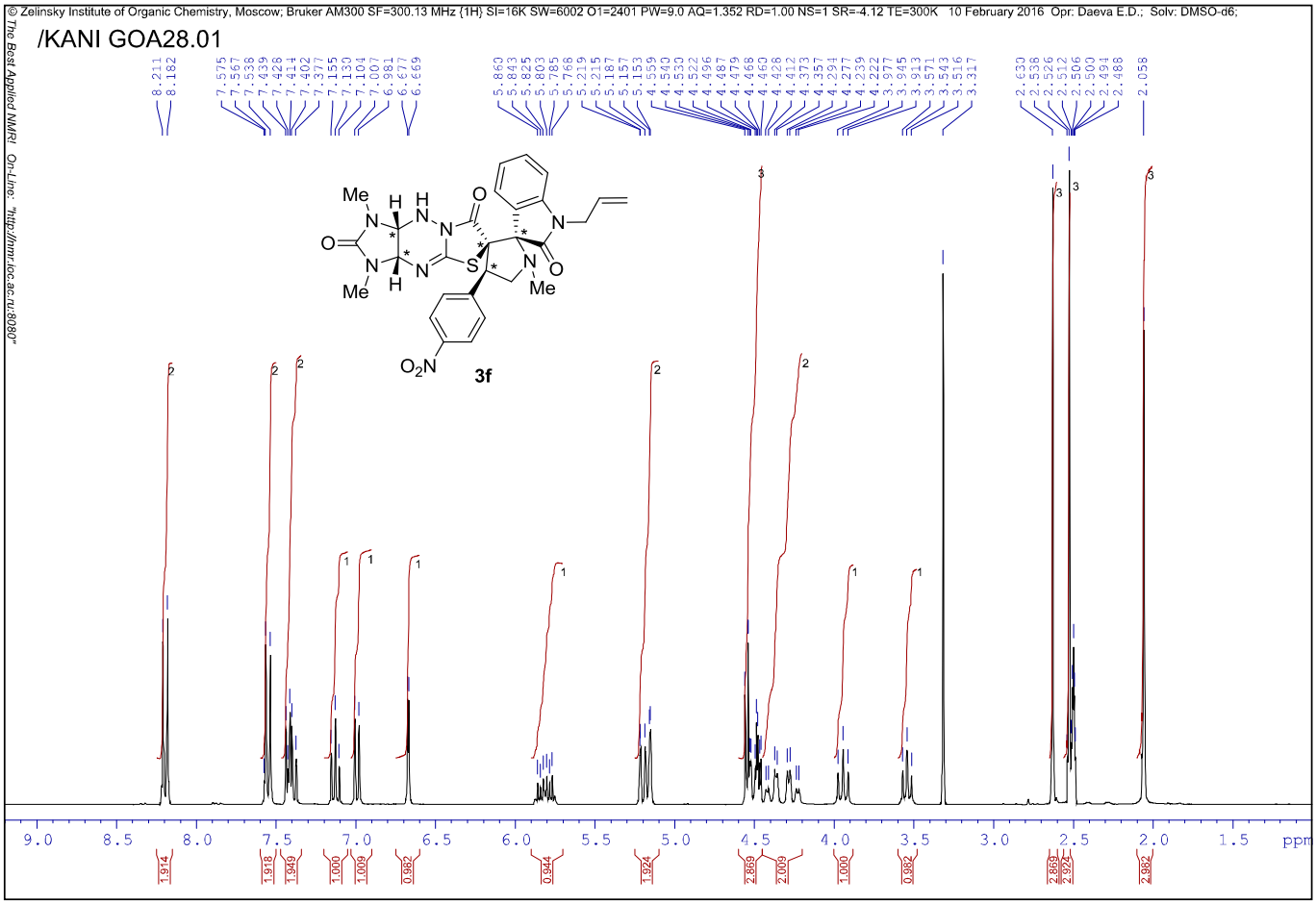
^{1}H NMR spectrum of **3e**



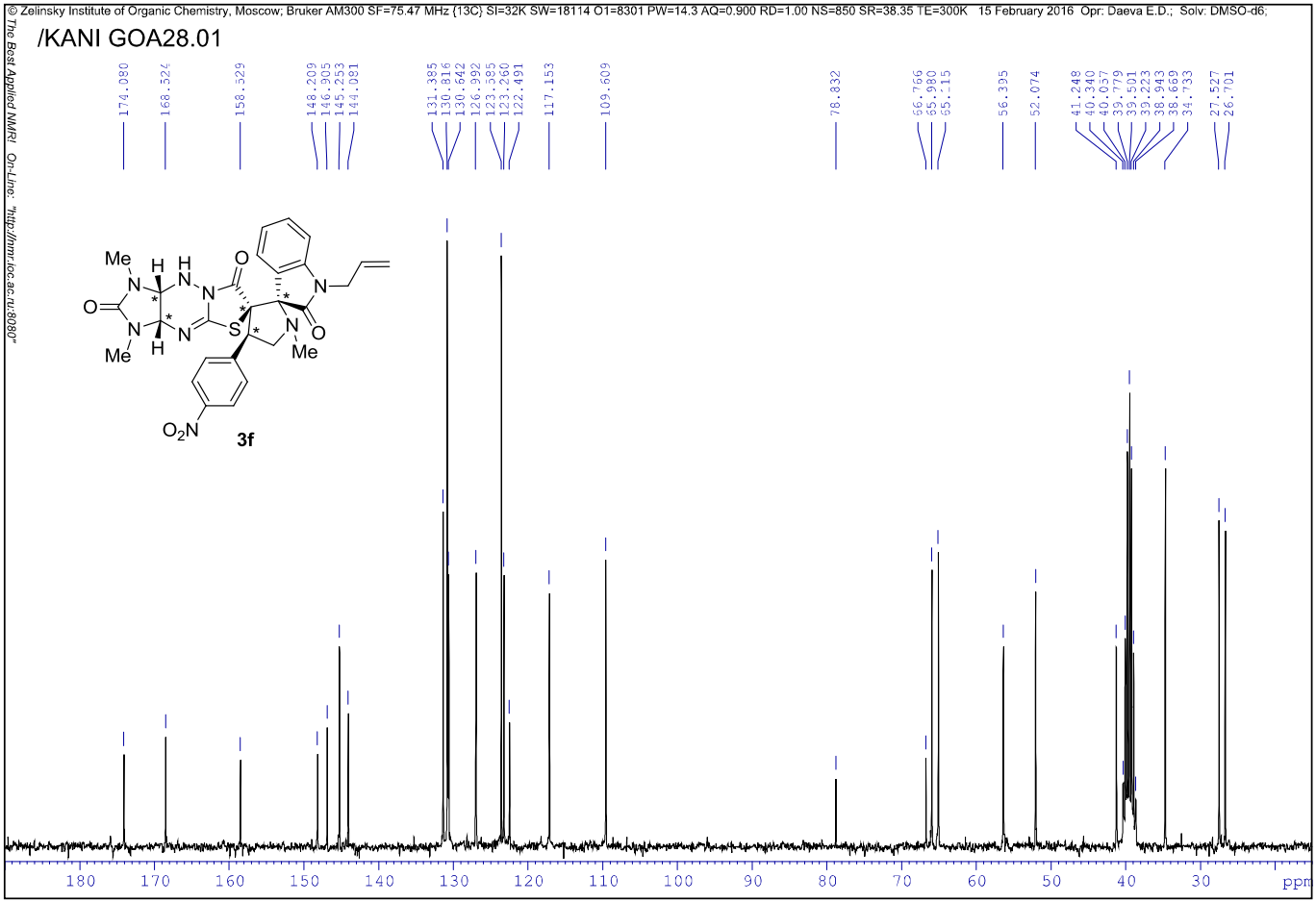
^{13}C NMR spectrum of **3e**



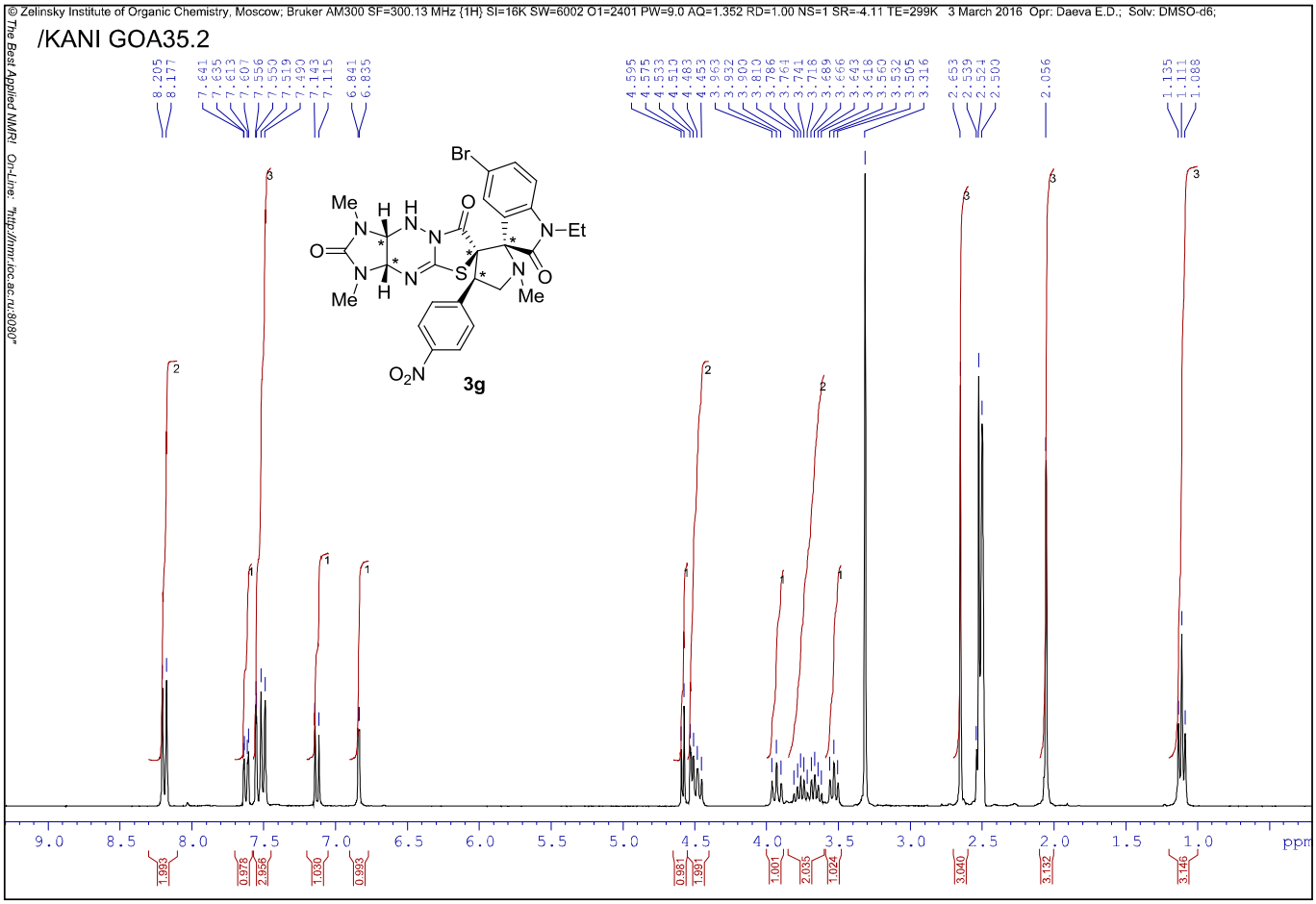
^{1}H NMR spectrum of **3f**



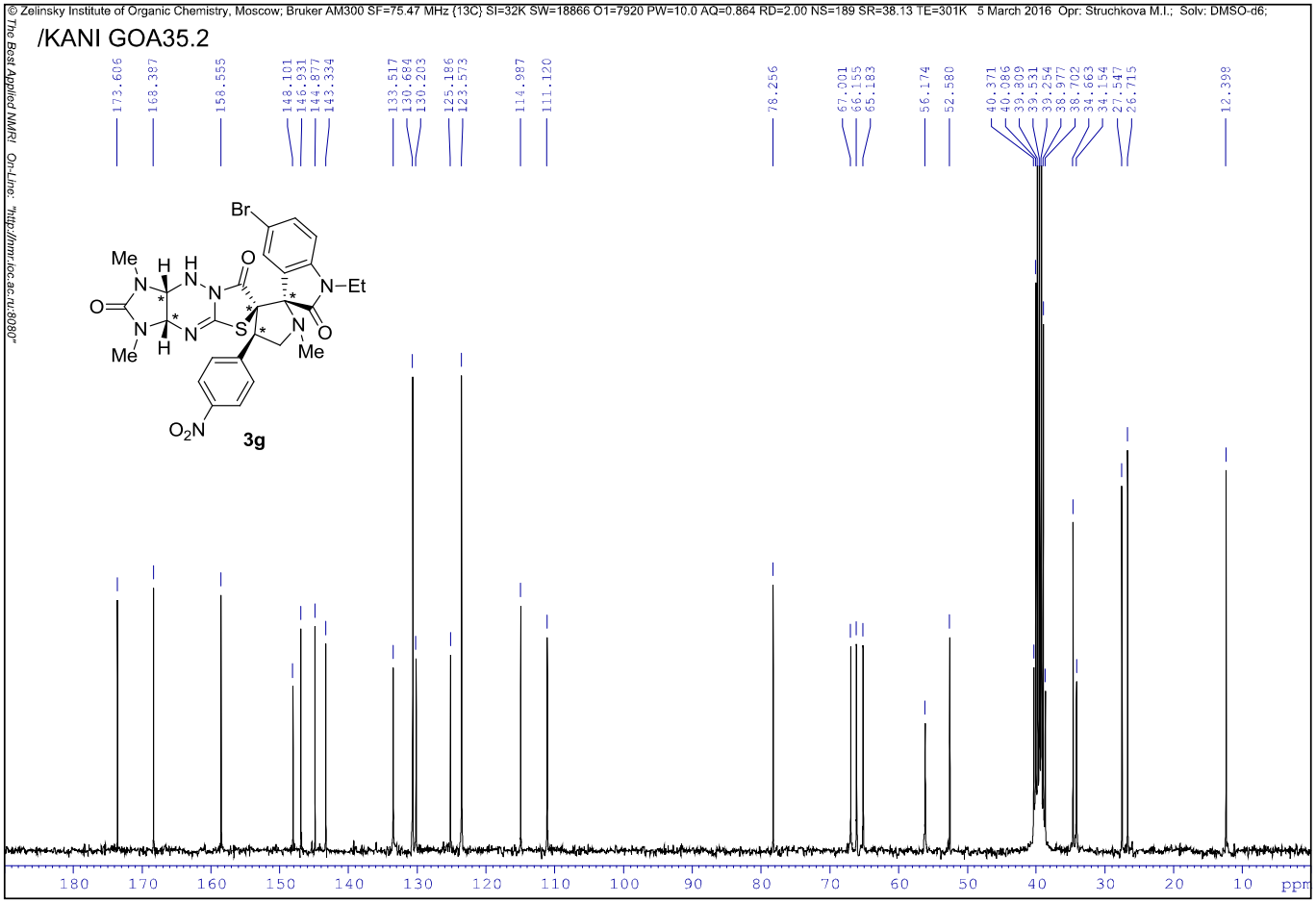
^{13}C NMR spectrum of **3f**



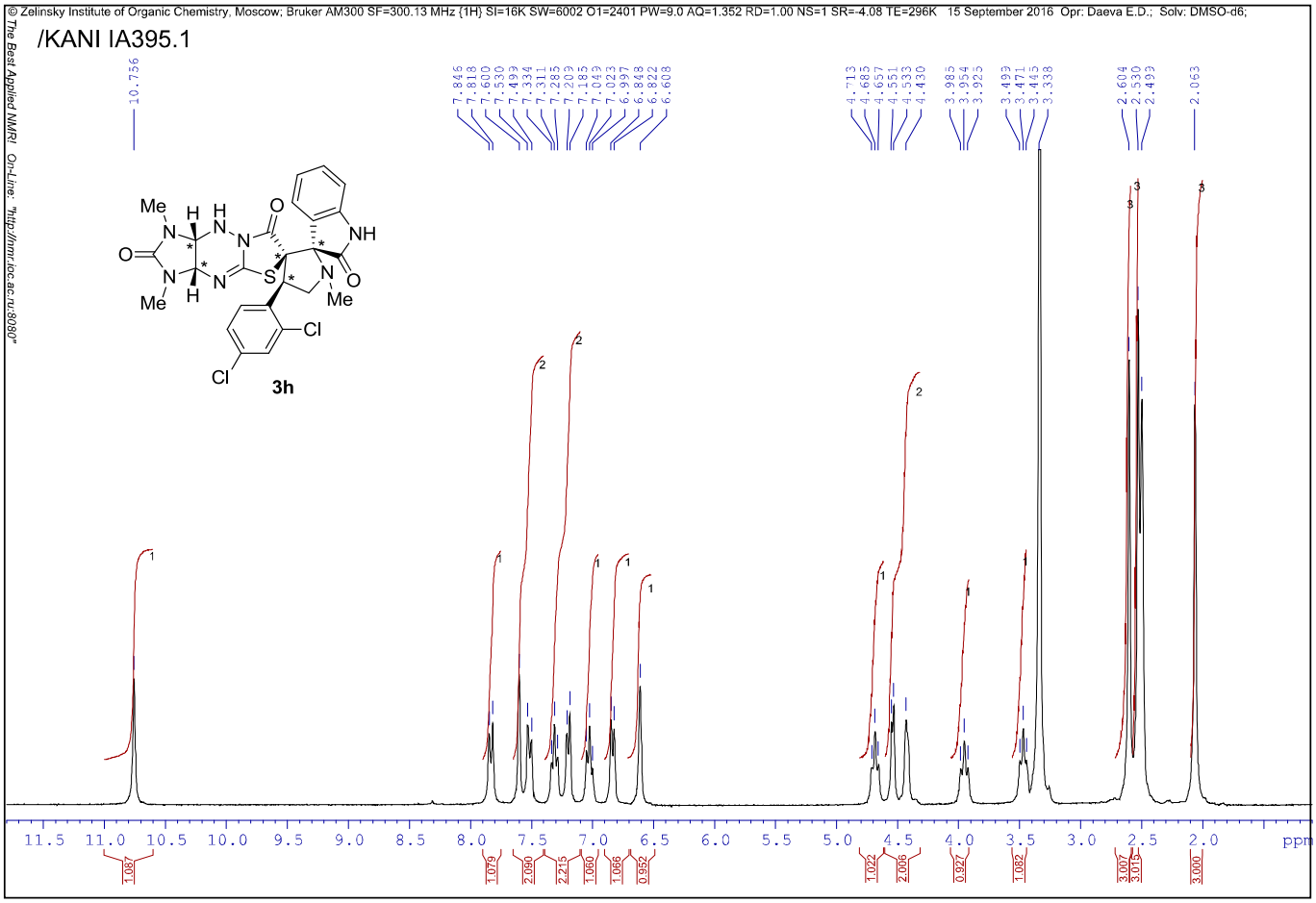
^{1}H NMR spectrum of **3g**



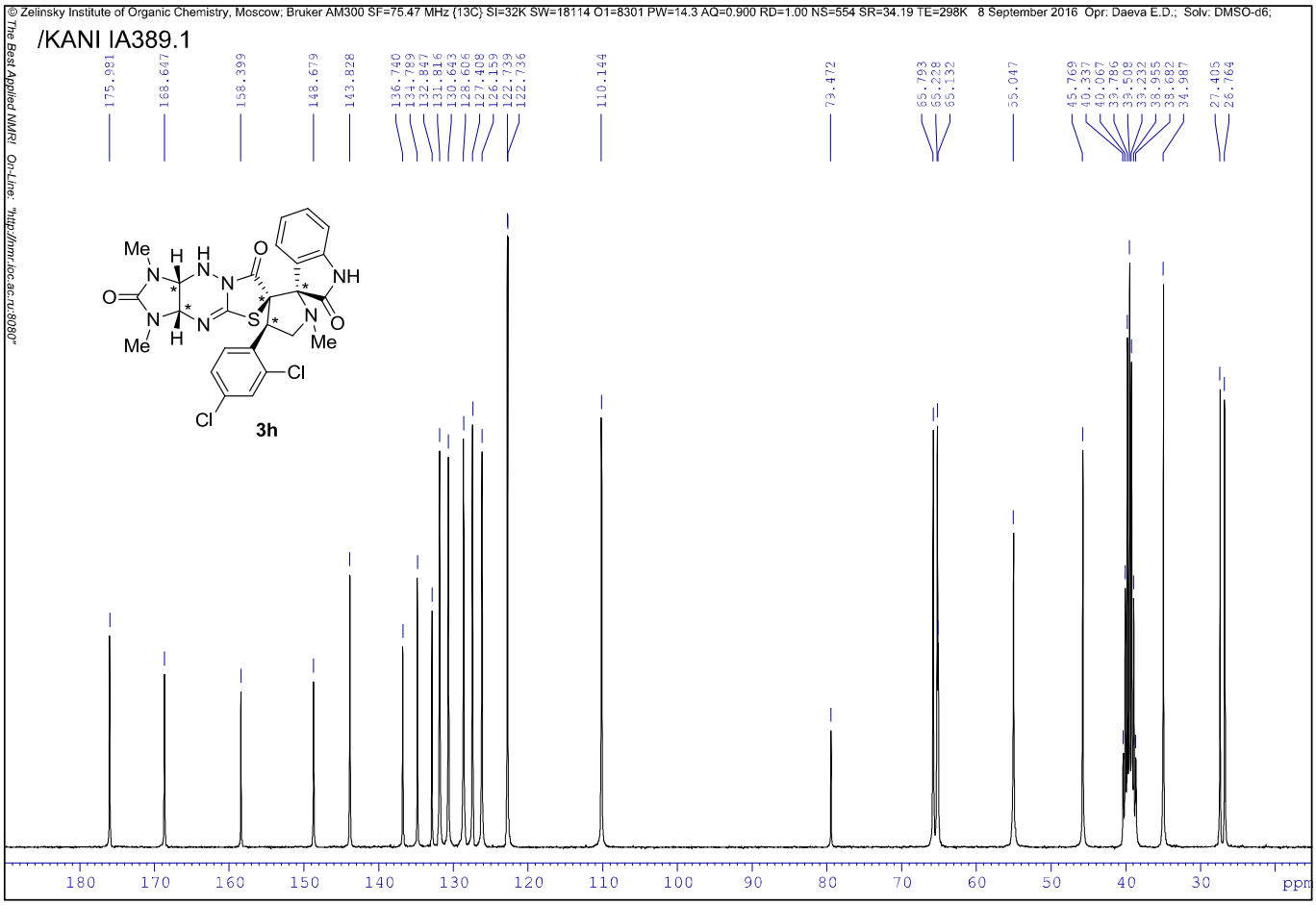
^{13}C NMR spectrum of **3g**



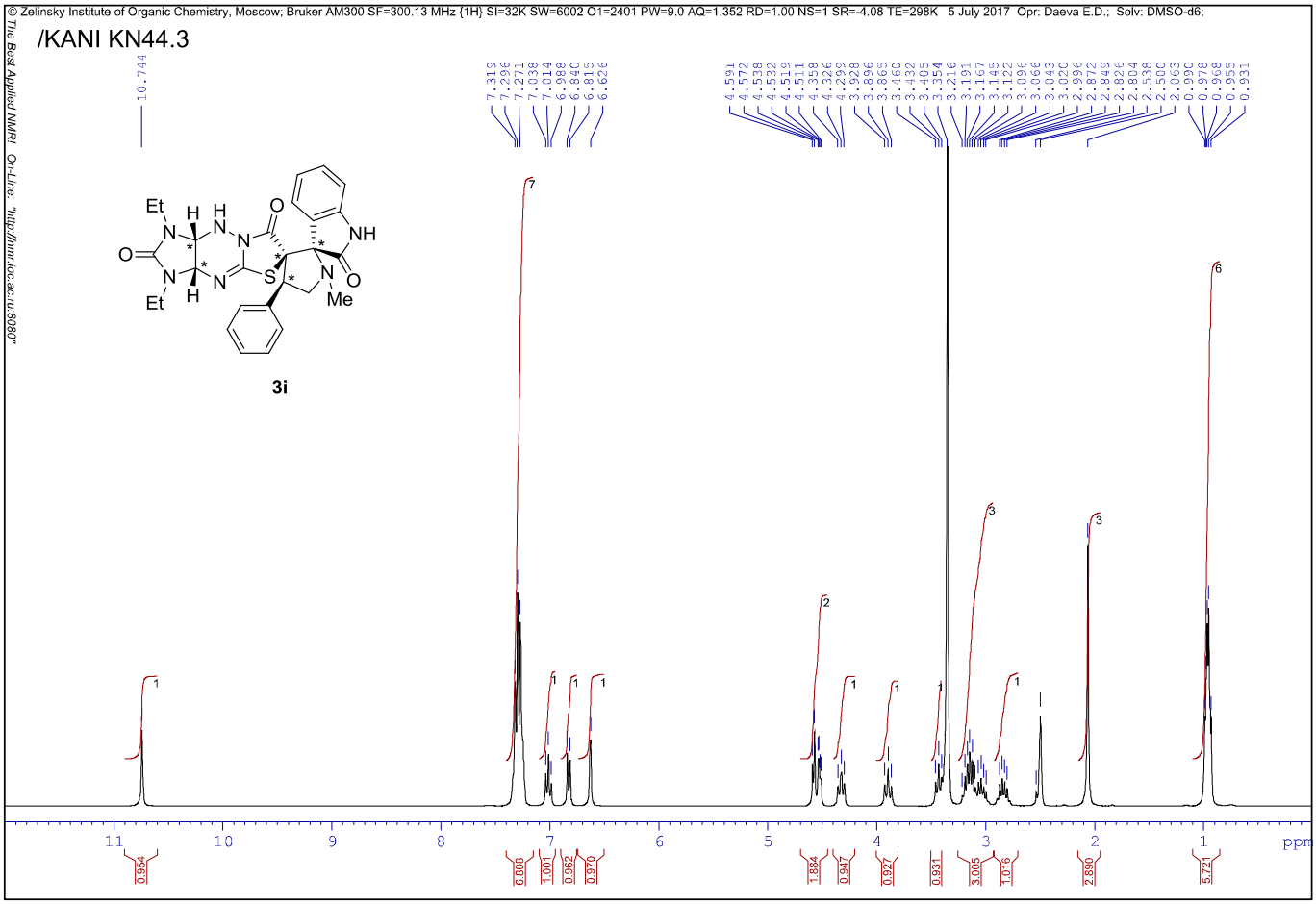
^{1}H NMR spectrum of **3h**



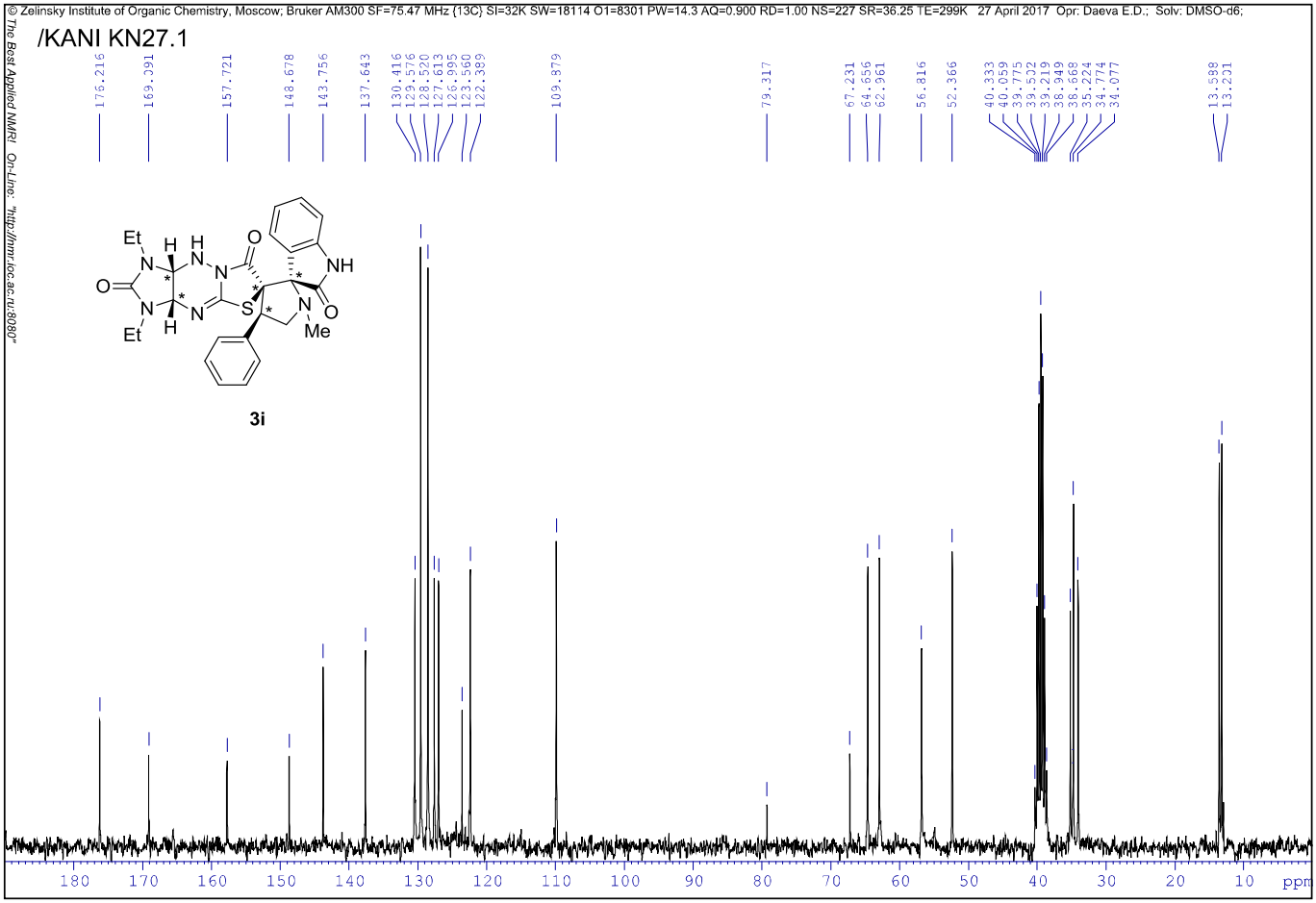
^{13}C NMR spectrum of **3h**



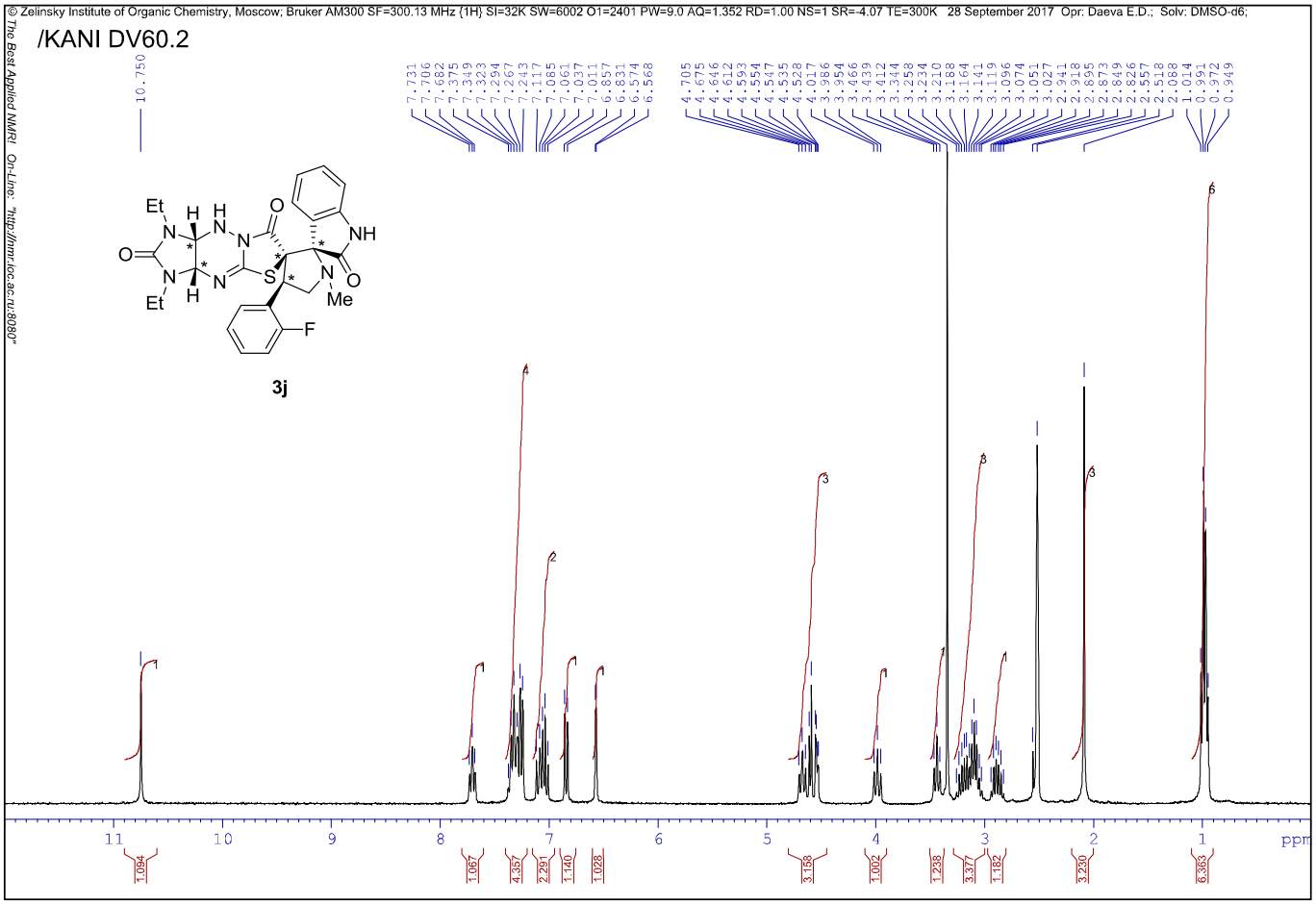
^{1}H NMR spectrum of **3i**



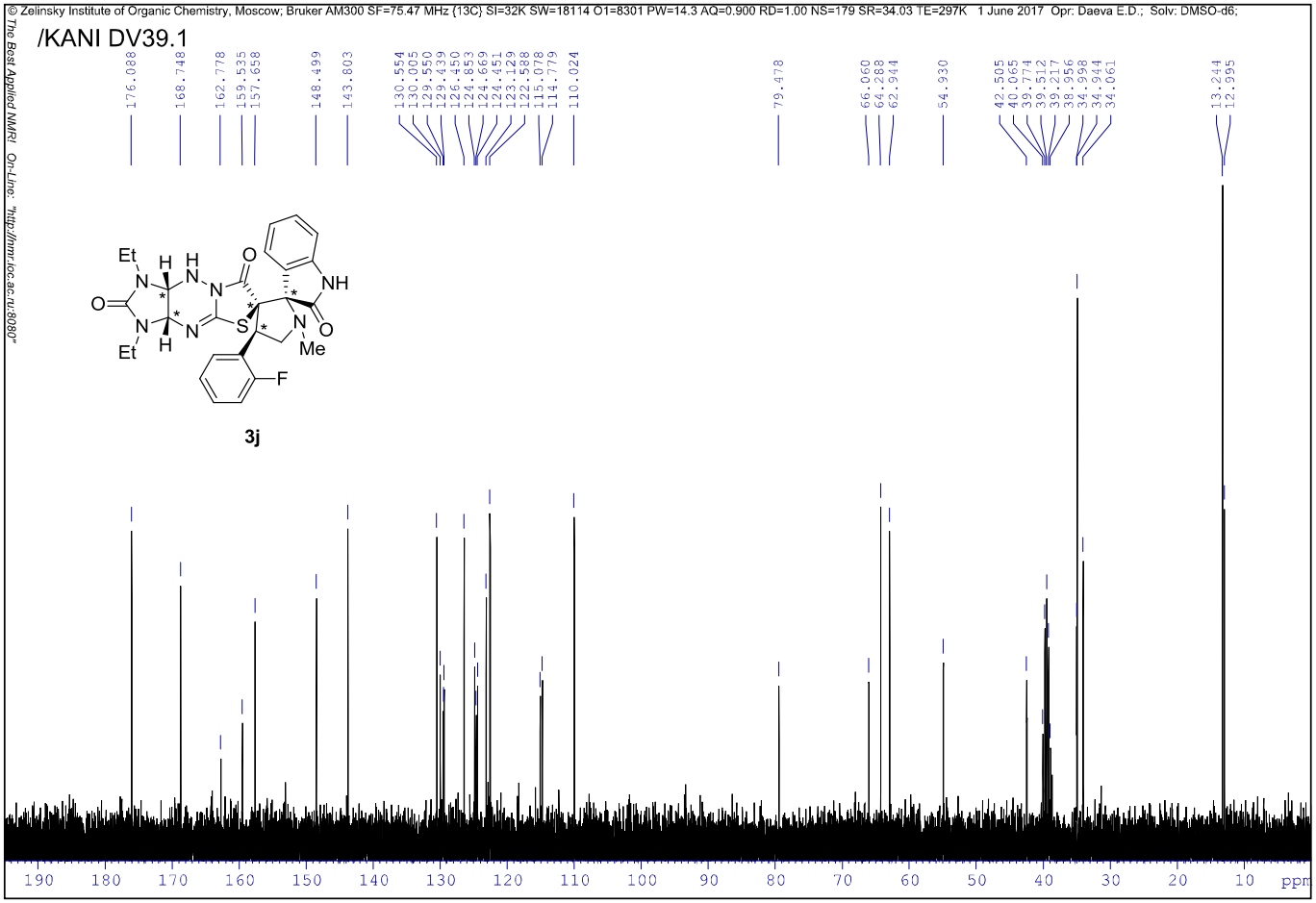
^{13}C NMR spectrum of **3i**



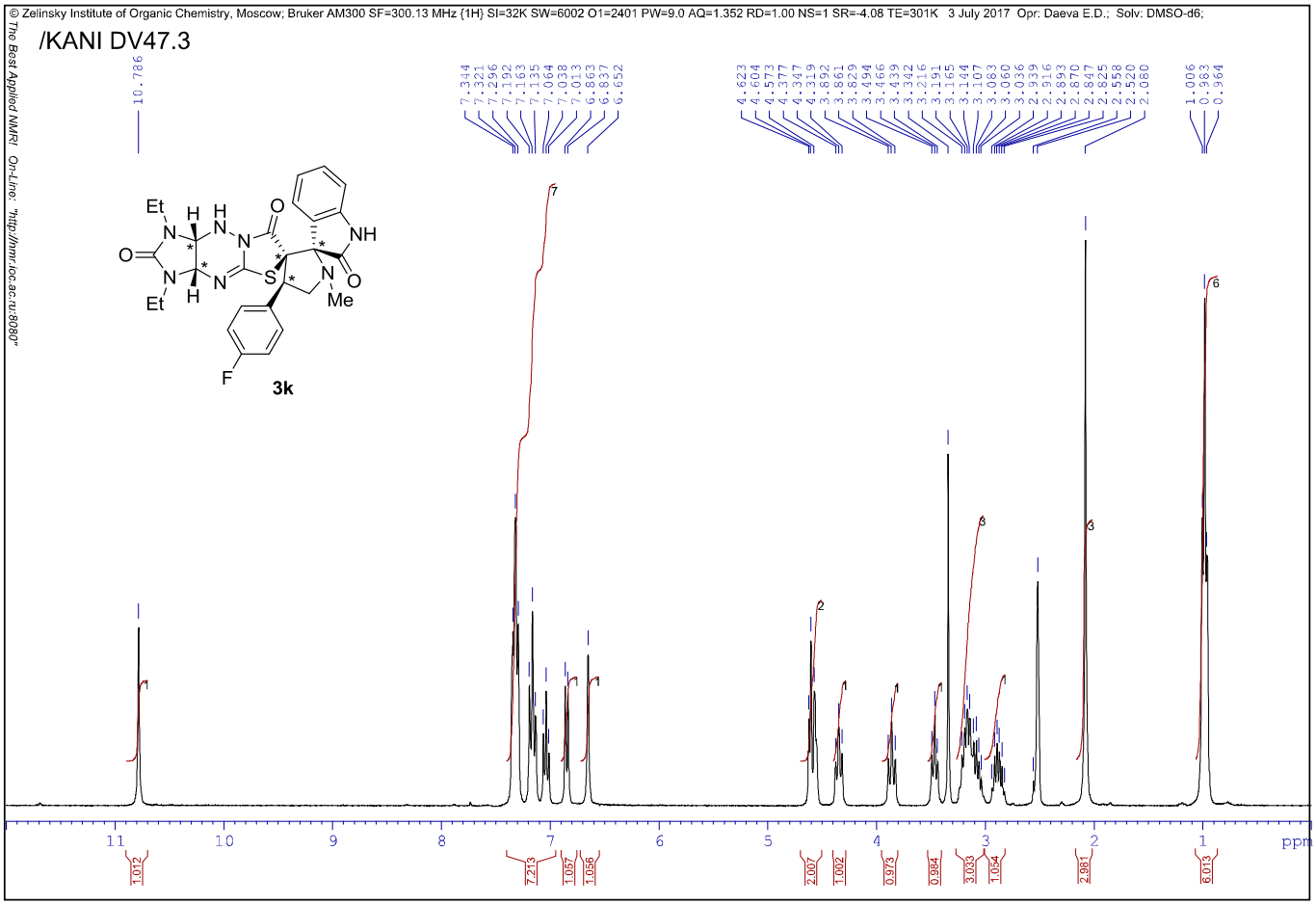
^{1}H NMR spectrum of **3j**



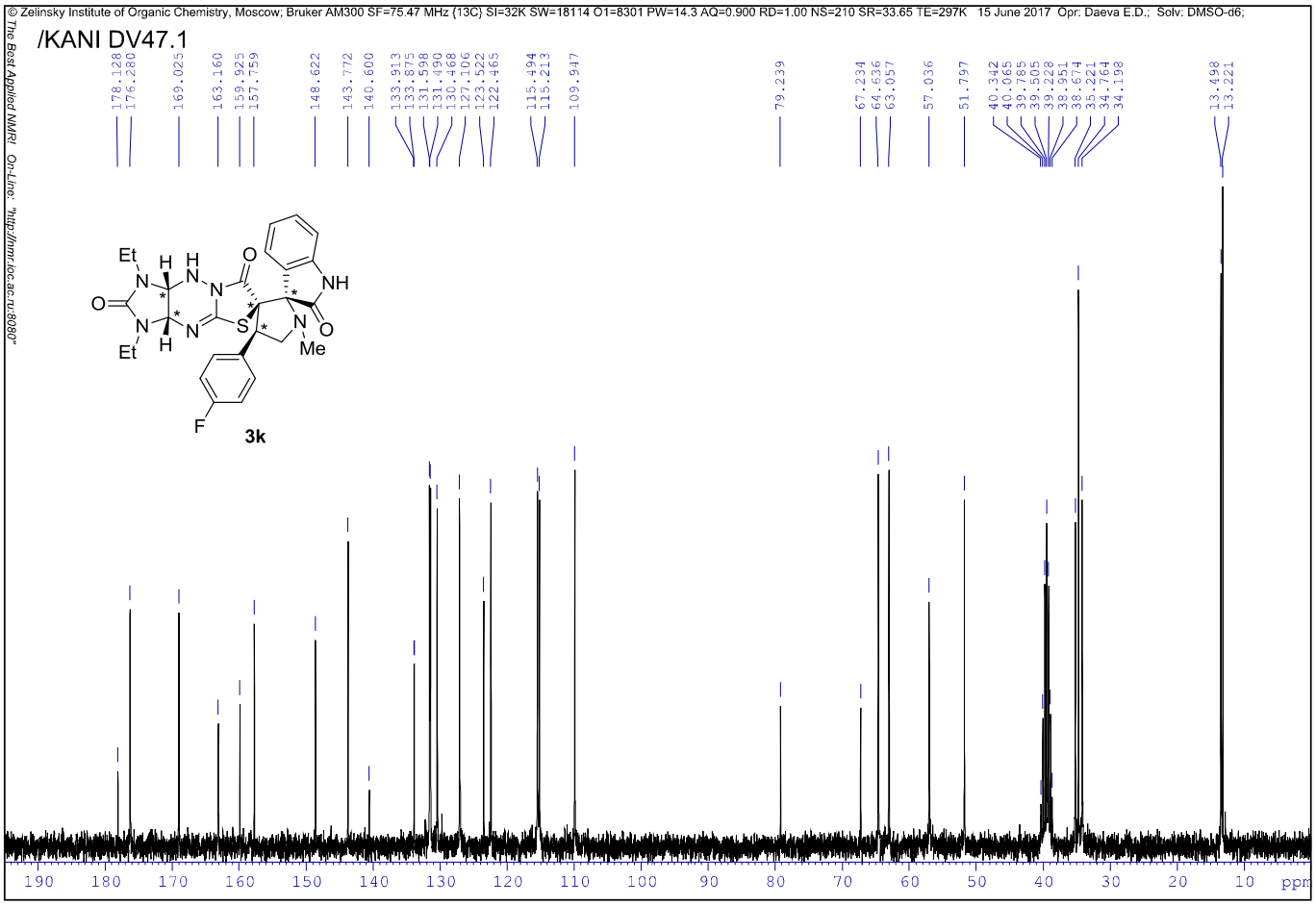
^{13}C NMR spectrum of **3j**



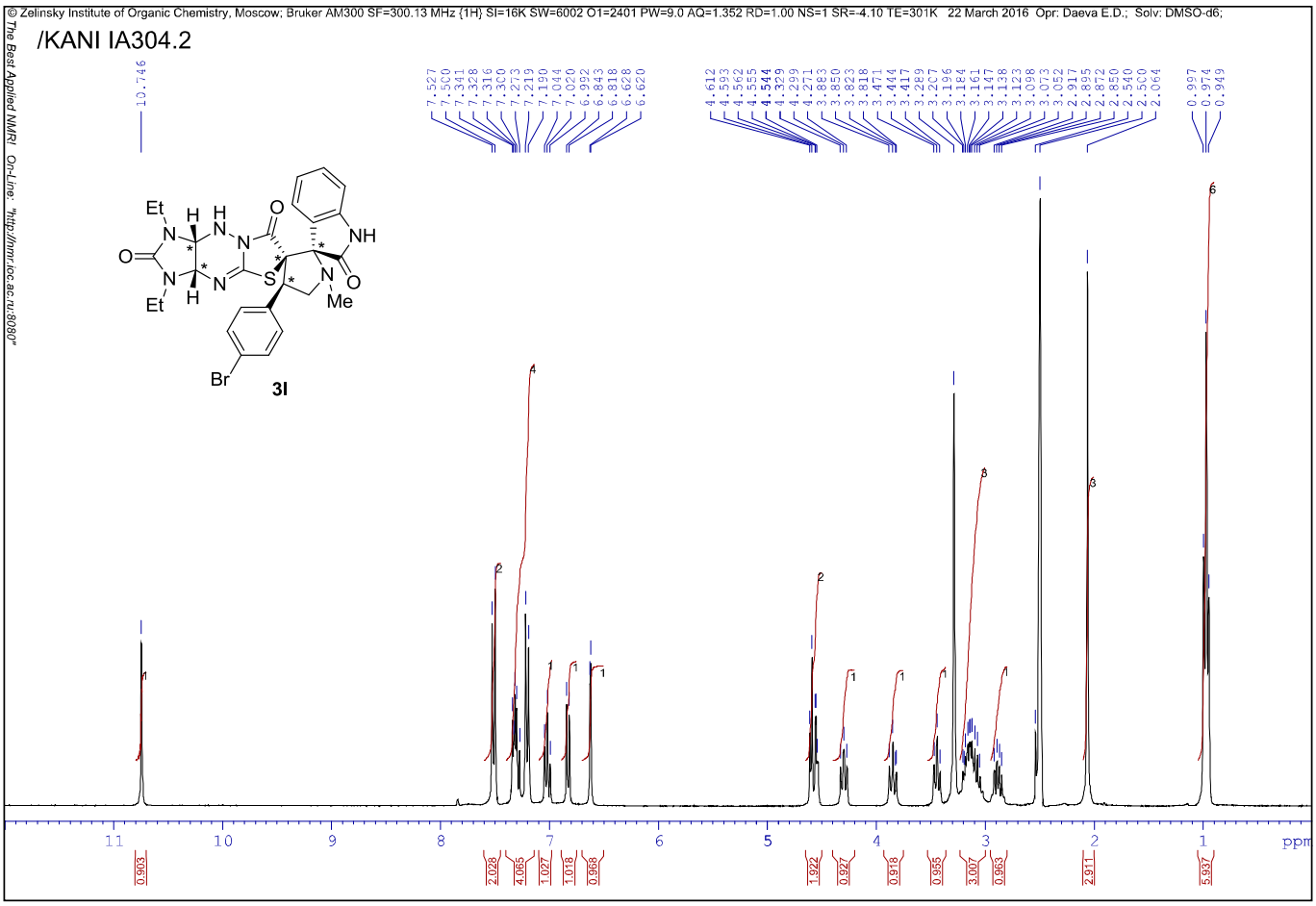
^{1}H NMR spectrum of **3k**



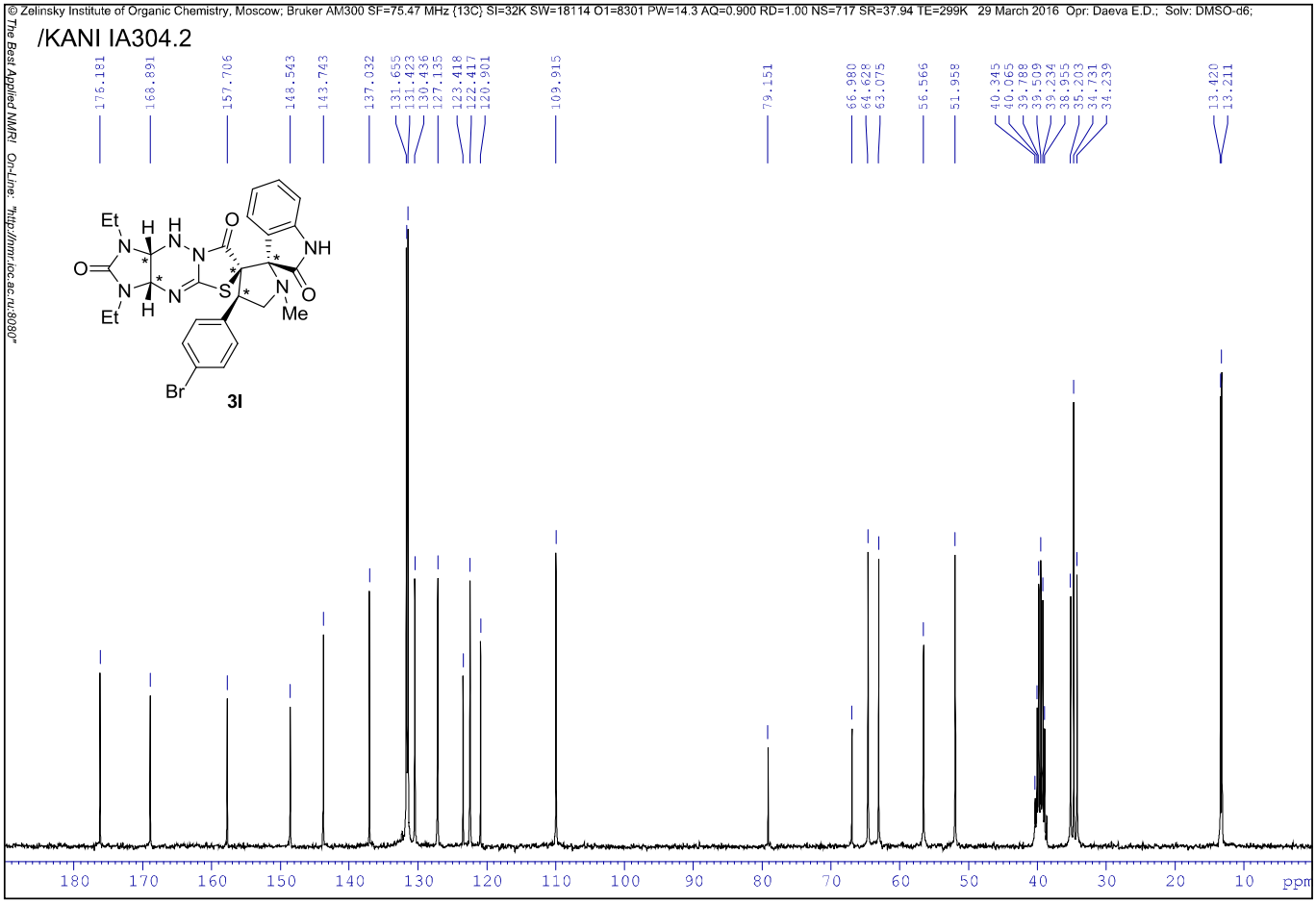
^{13}C NMR spectrum of **3k**



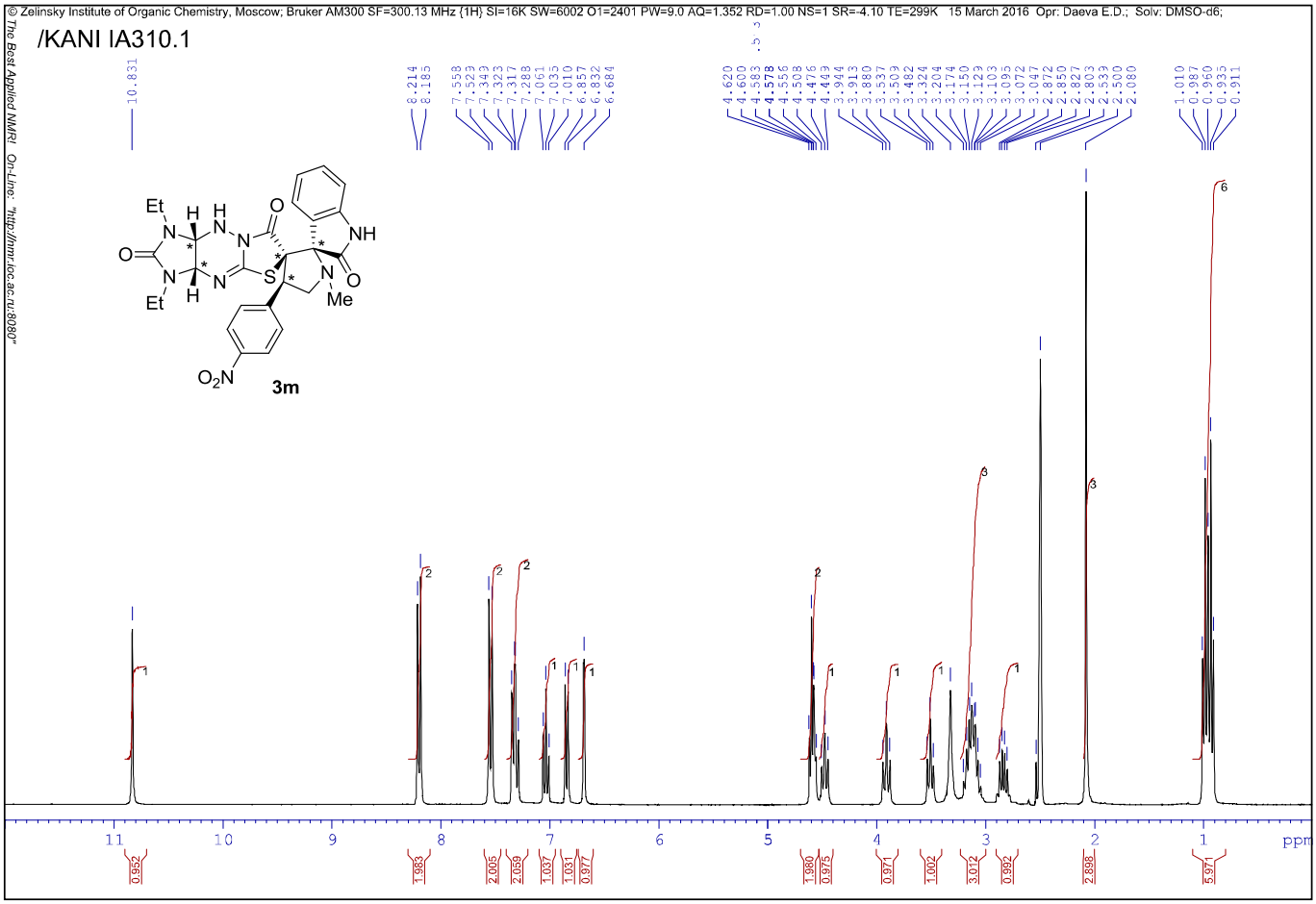
^{1}H NMR spectrum of **3l**



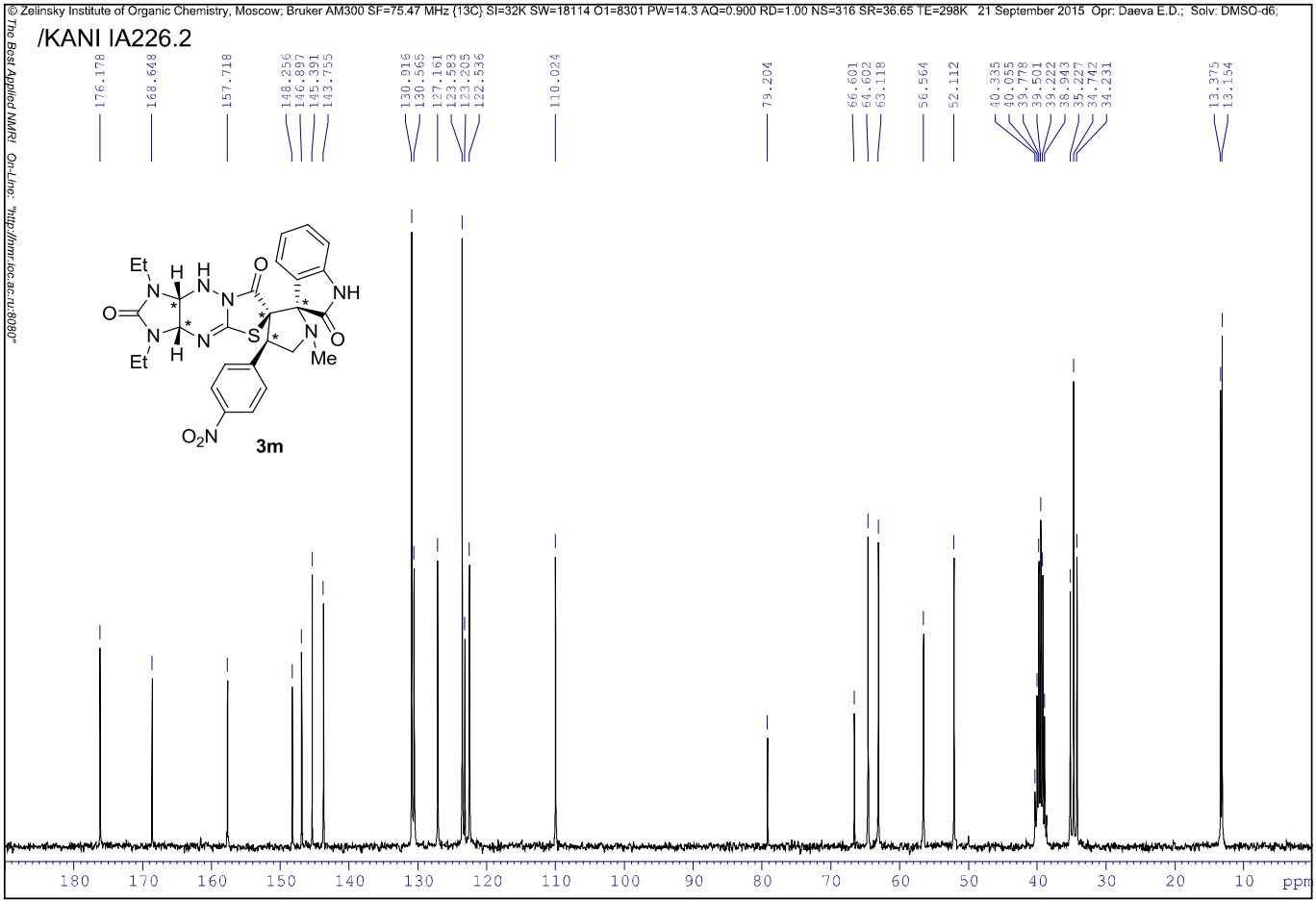
^{13}C NMR spectrum of **3l**



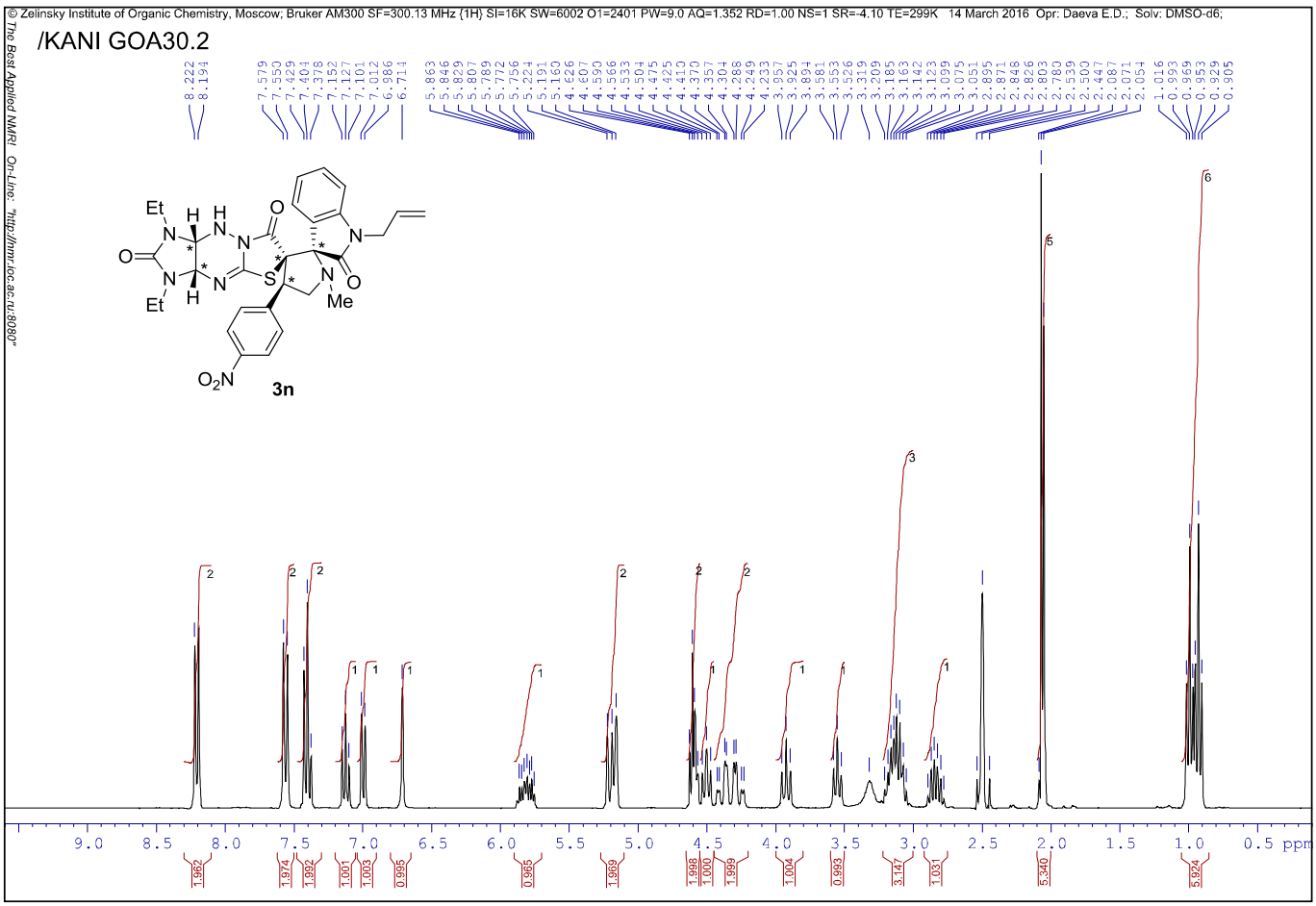
^{1}H NMR spectrum of **3m**



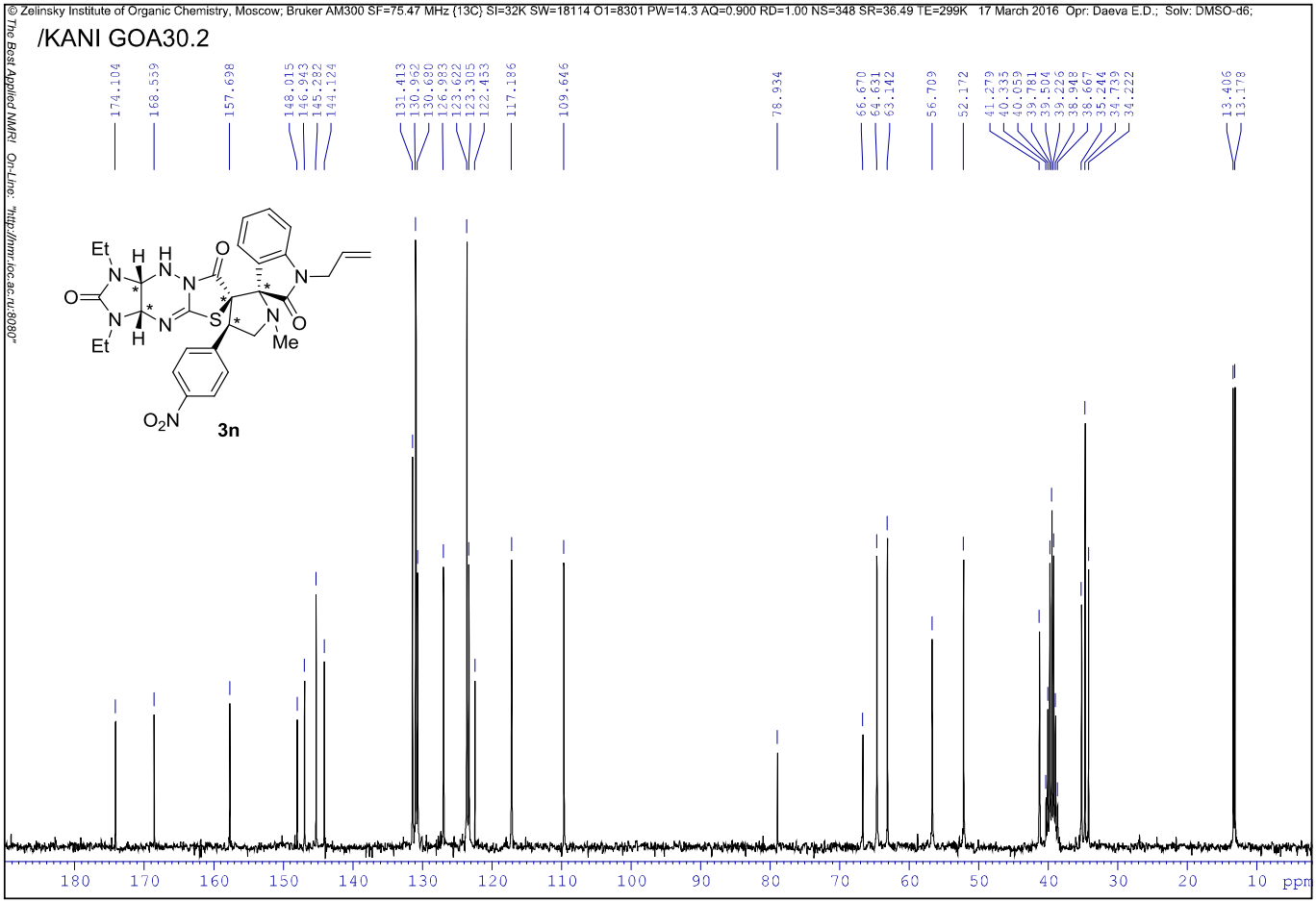
^{13}C NMR spectrum of **3m**



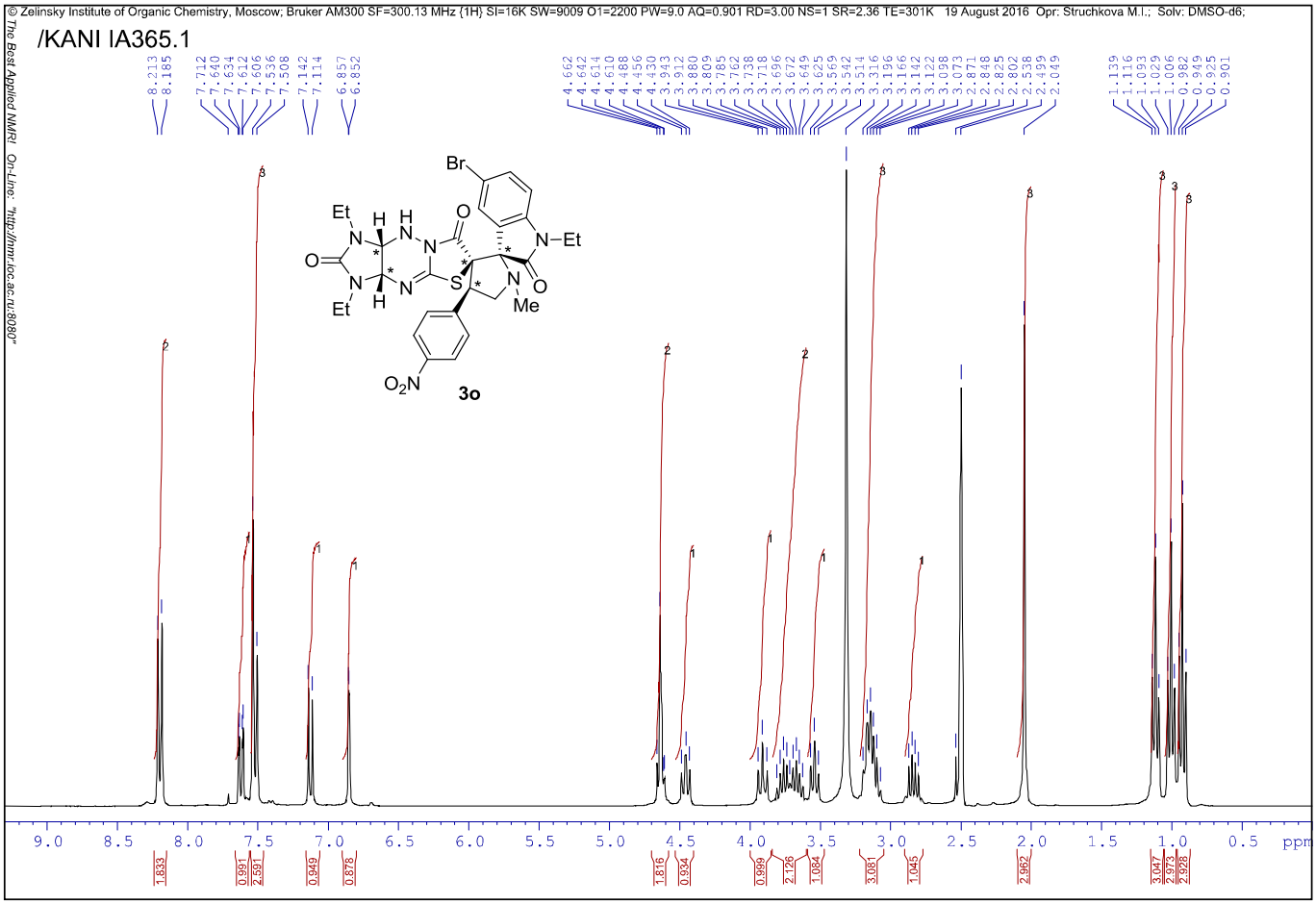
^{1}H NMR spectrum of **3n**



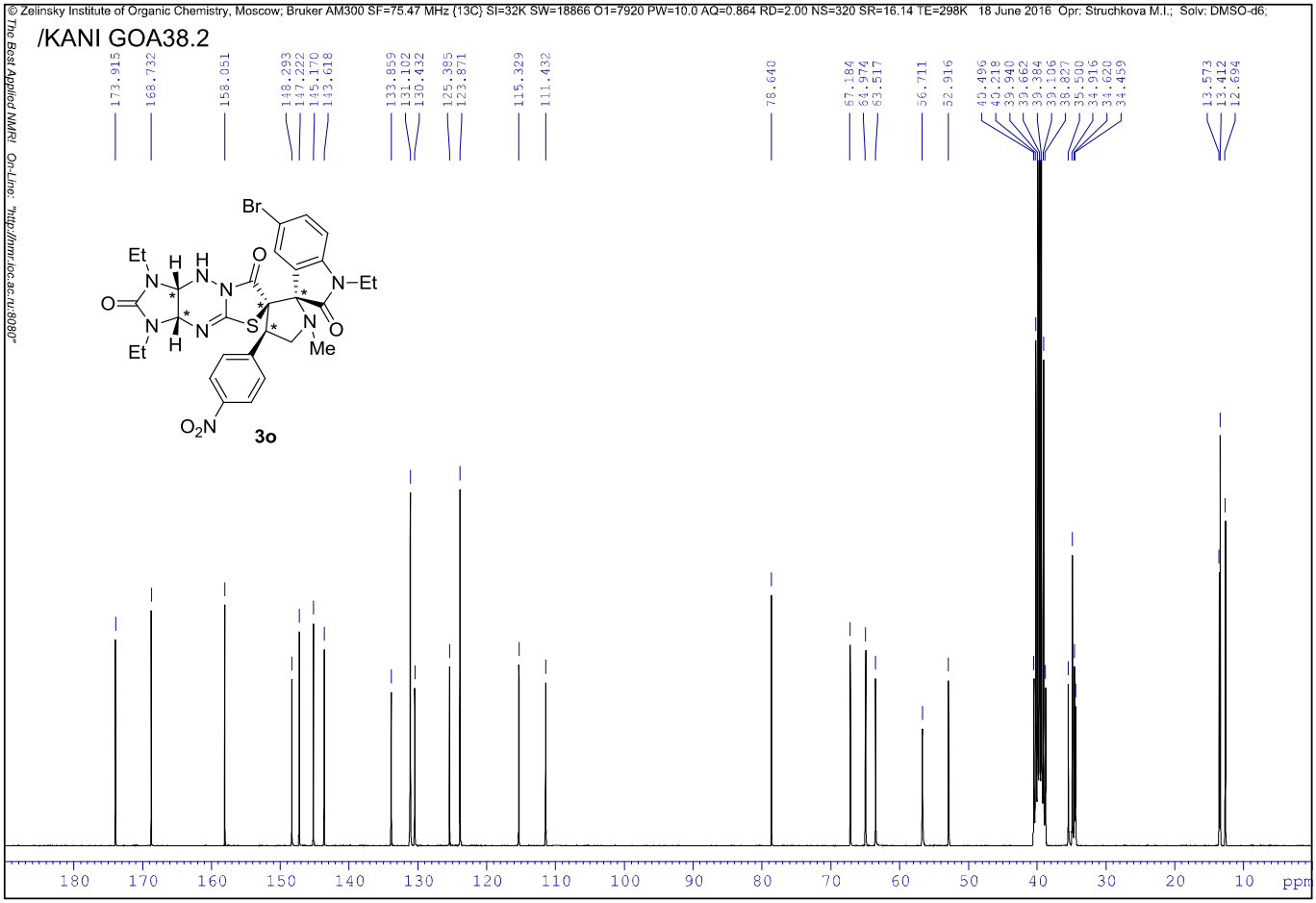
^{13}C NMR spectrum of **3n**



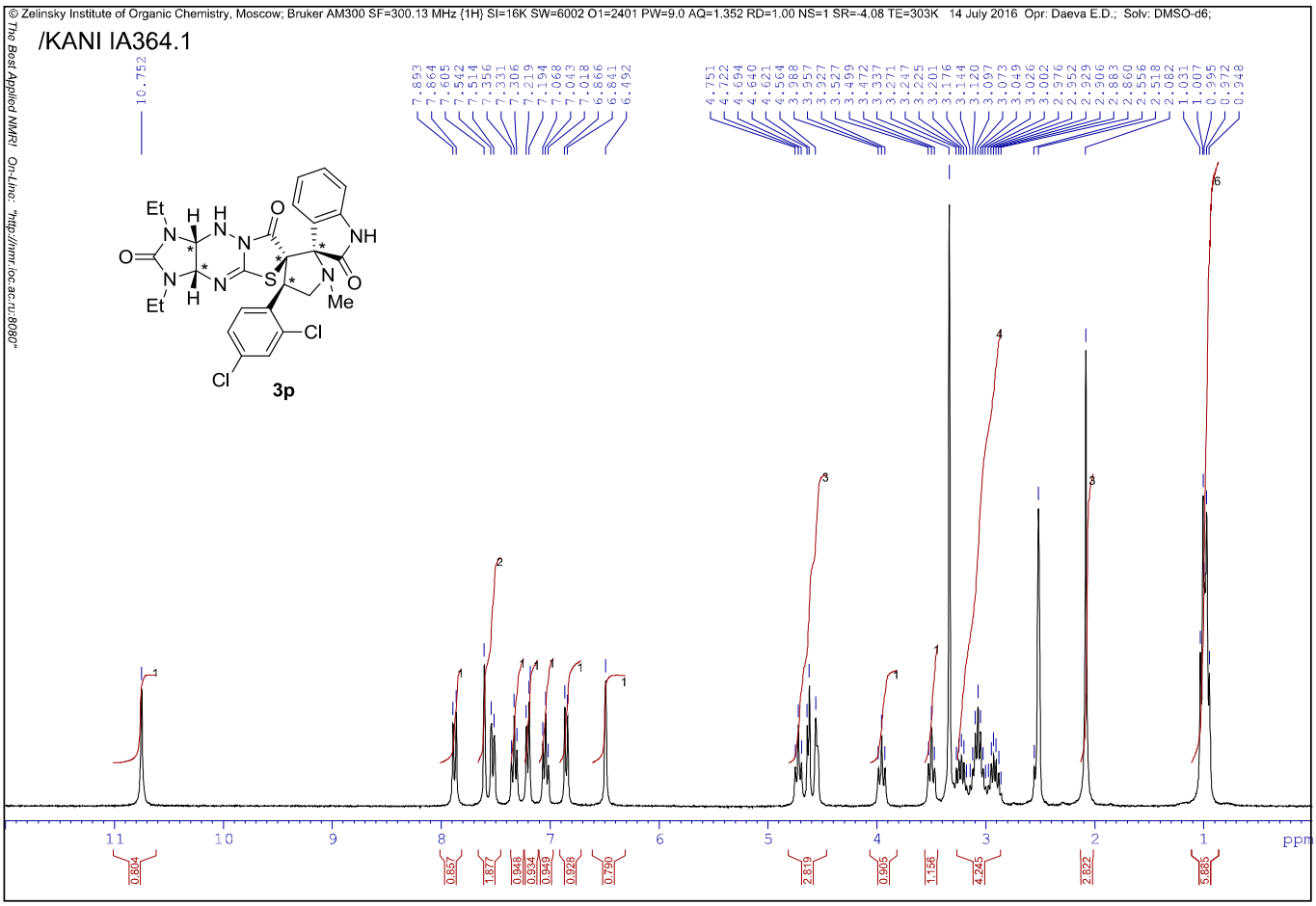
^{1}H NMR spectrum of **3o**



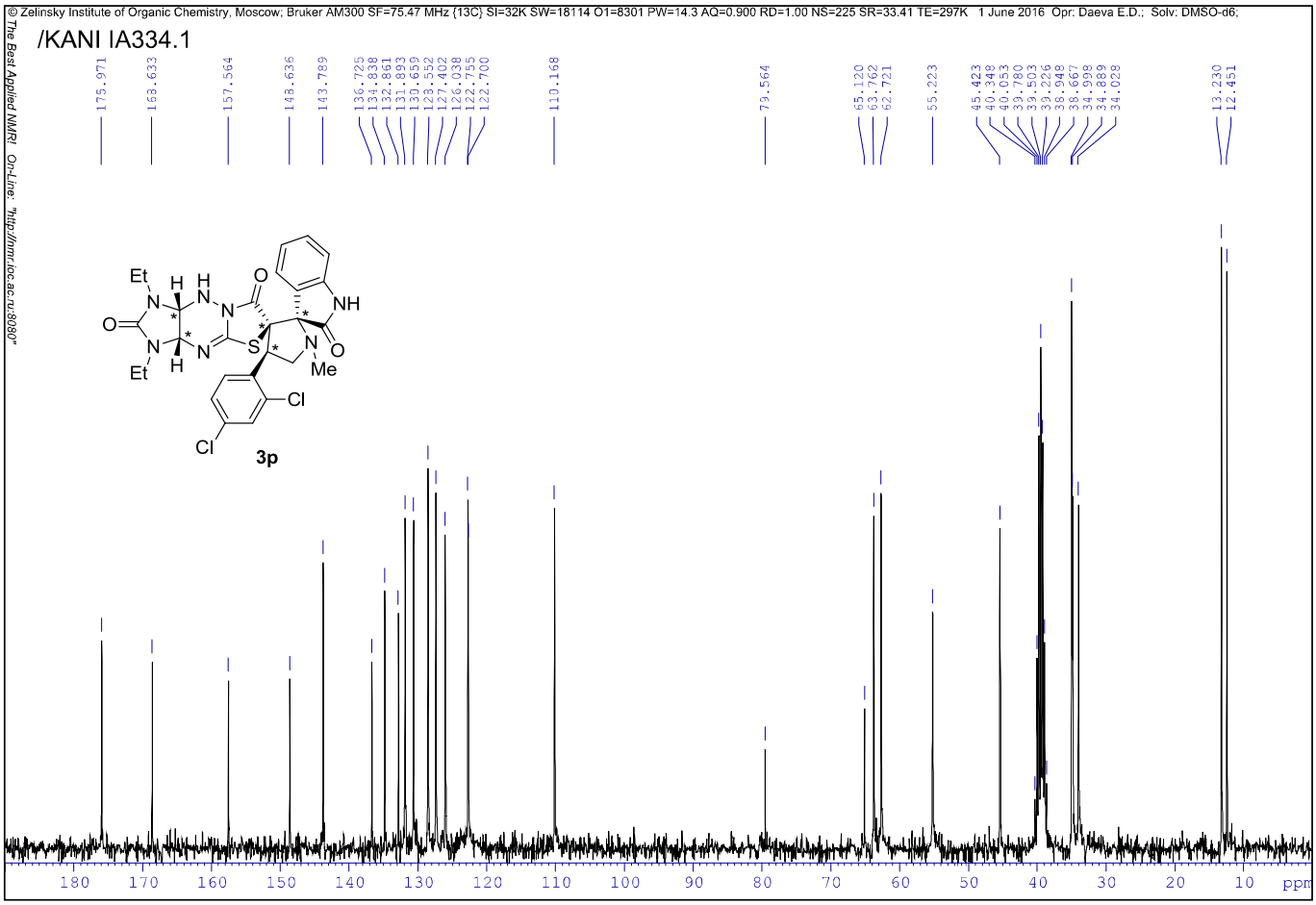
^{13}C NMR spectrum of **3o**



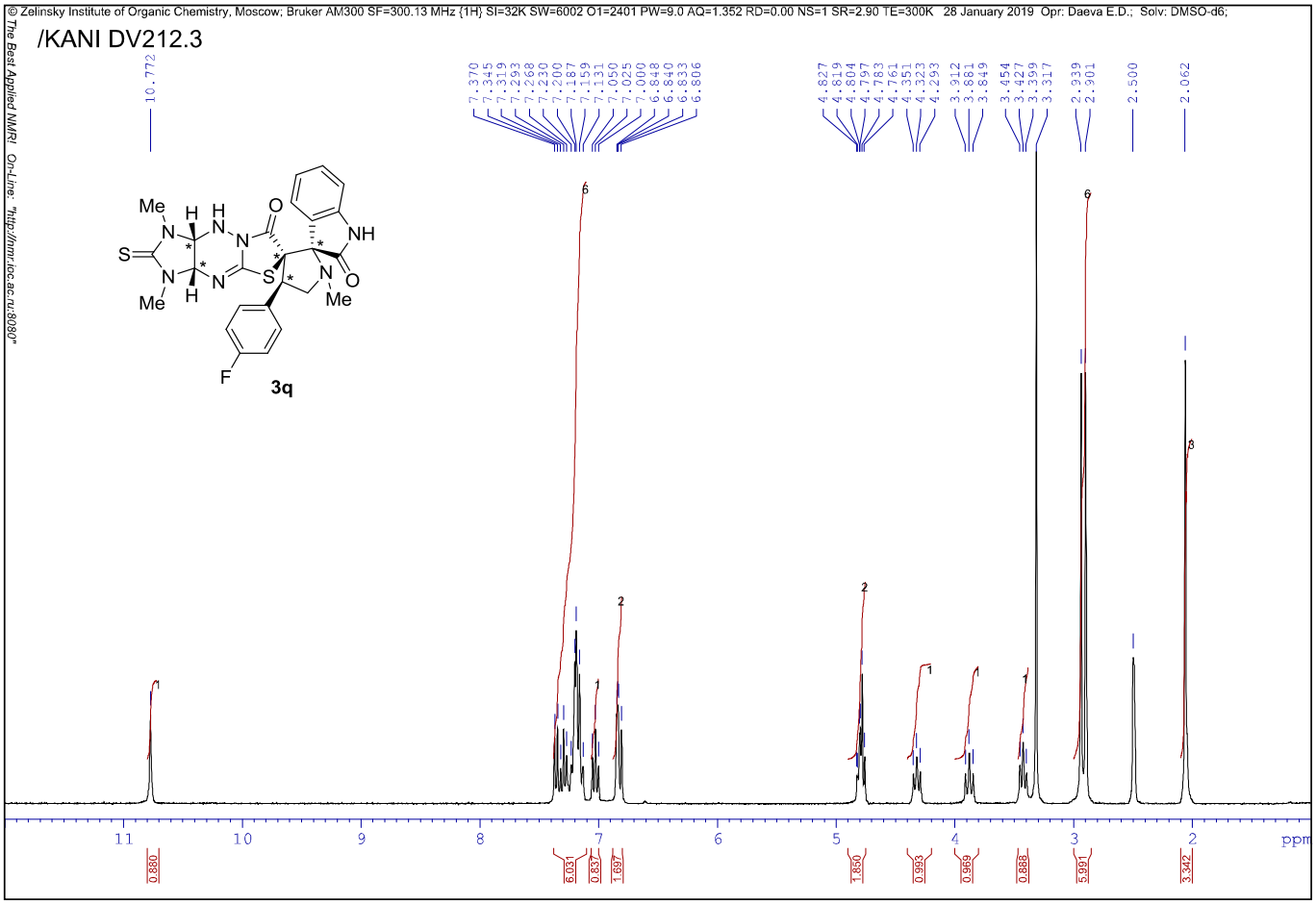
^{1}H NMR spectrum of **3p**



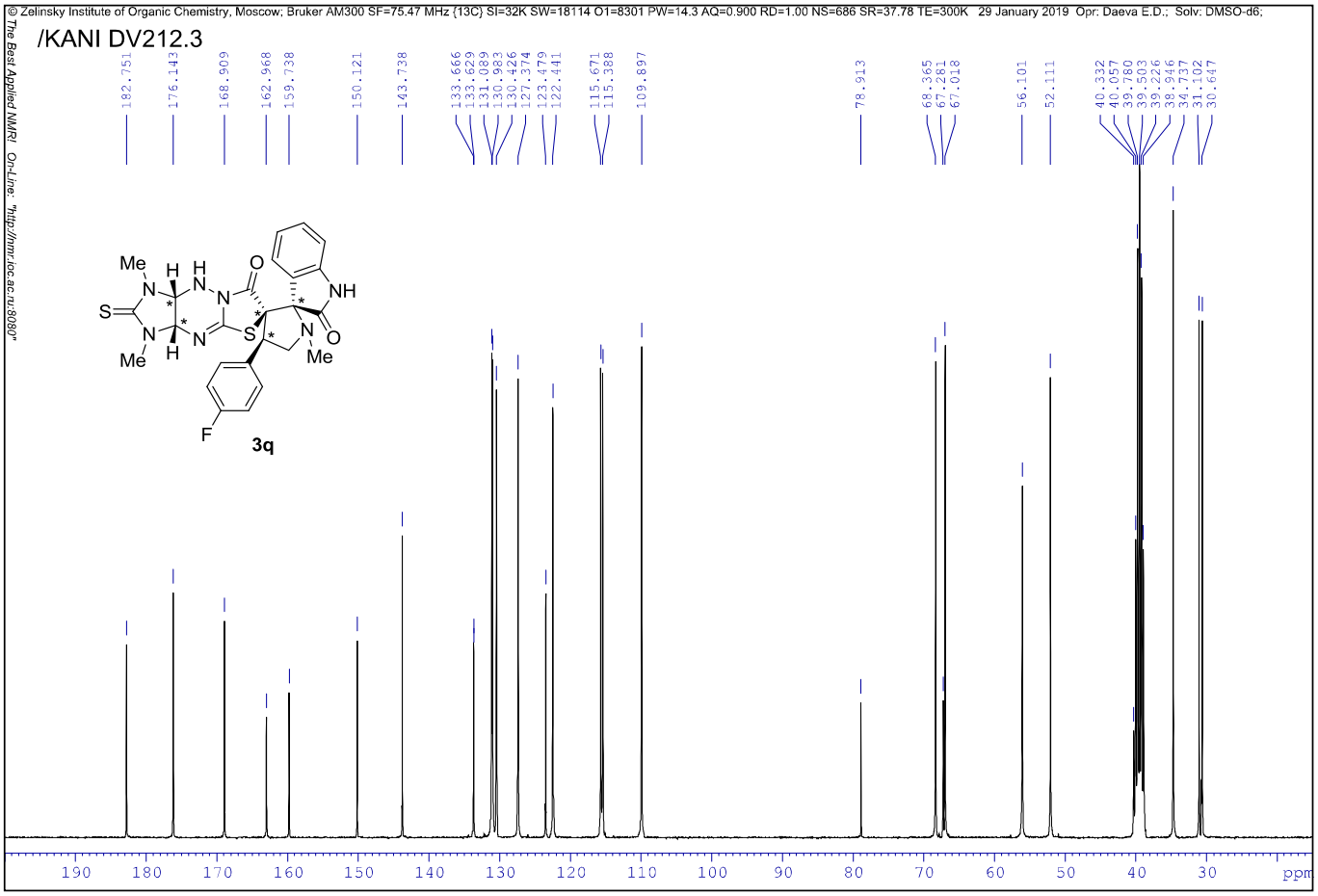
^{13}C NMR spectrum of **3p**



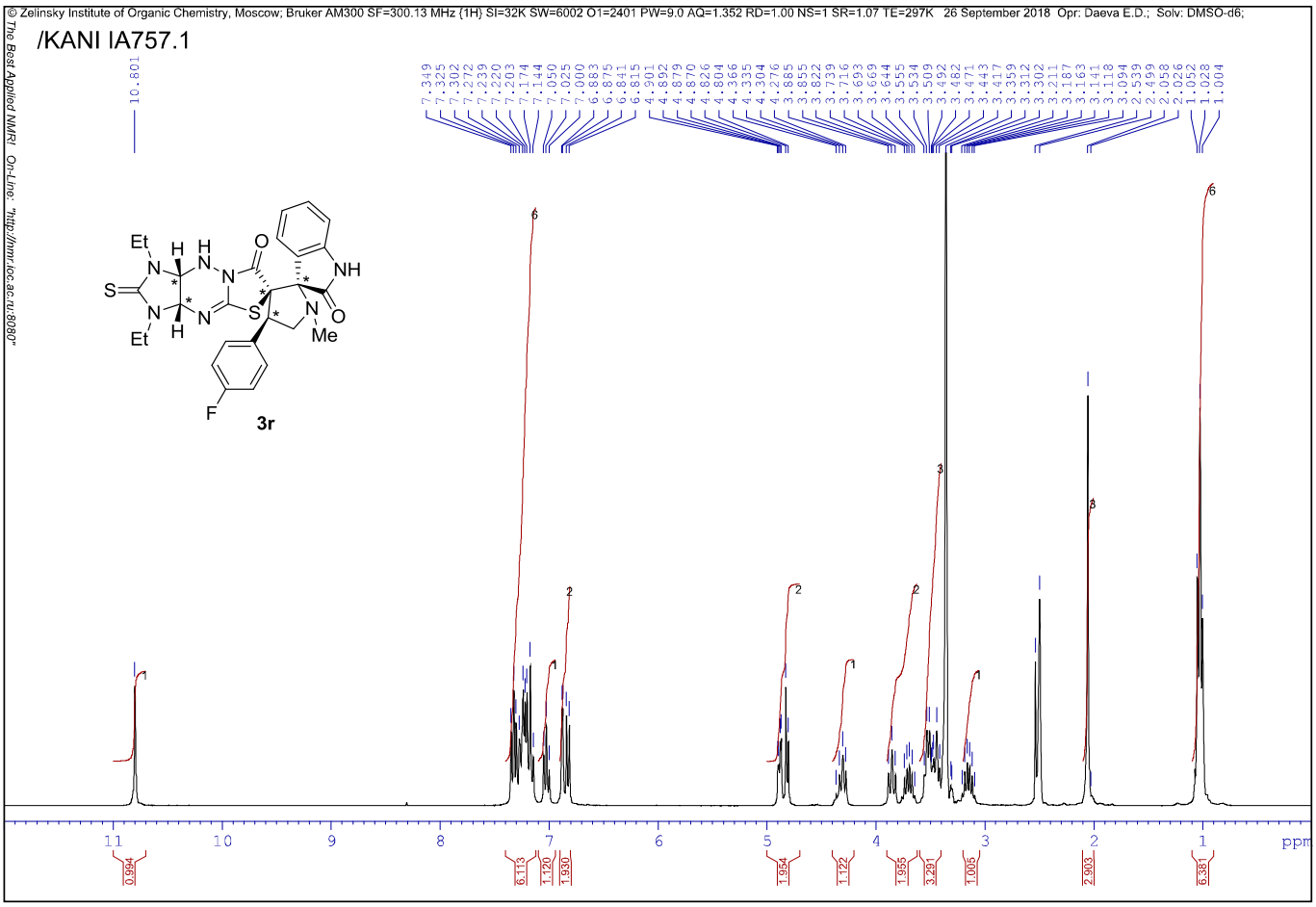
^{1}H NMR spectrum of **3q**



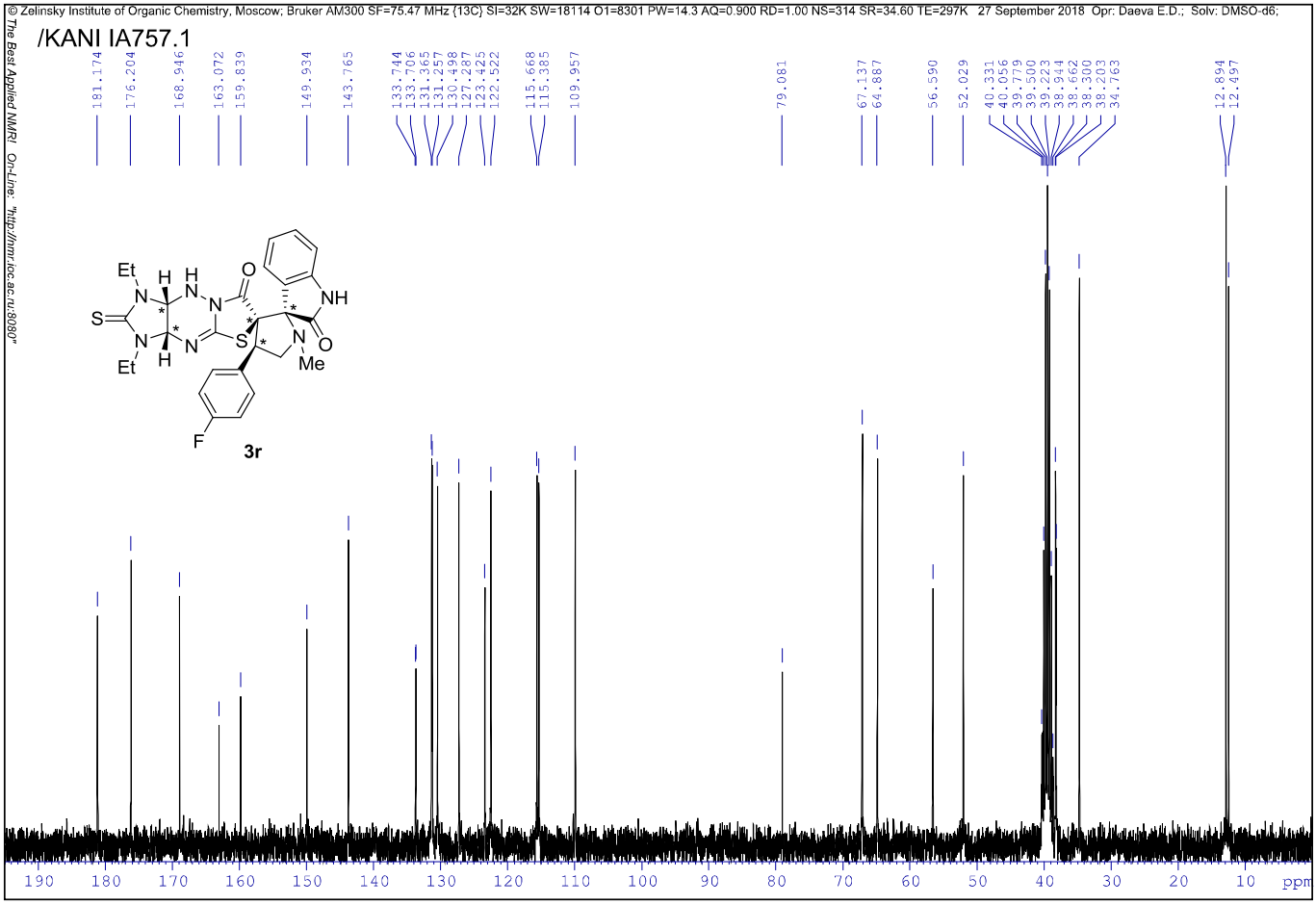
^{13}C NMR spectrum of **3q**



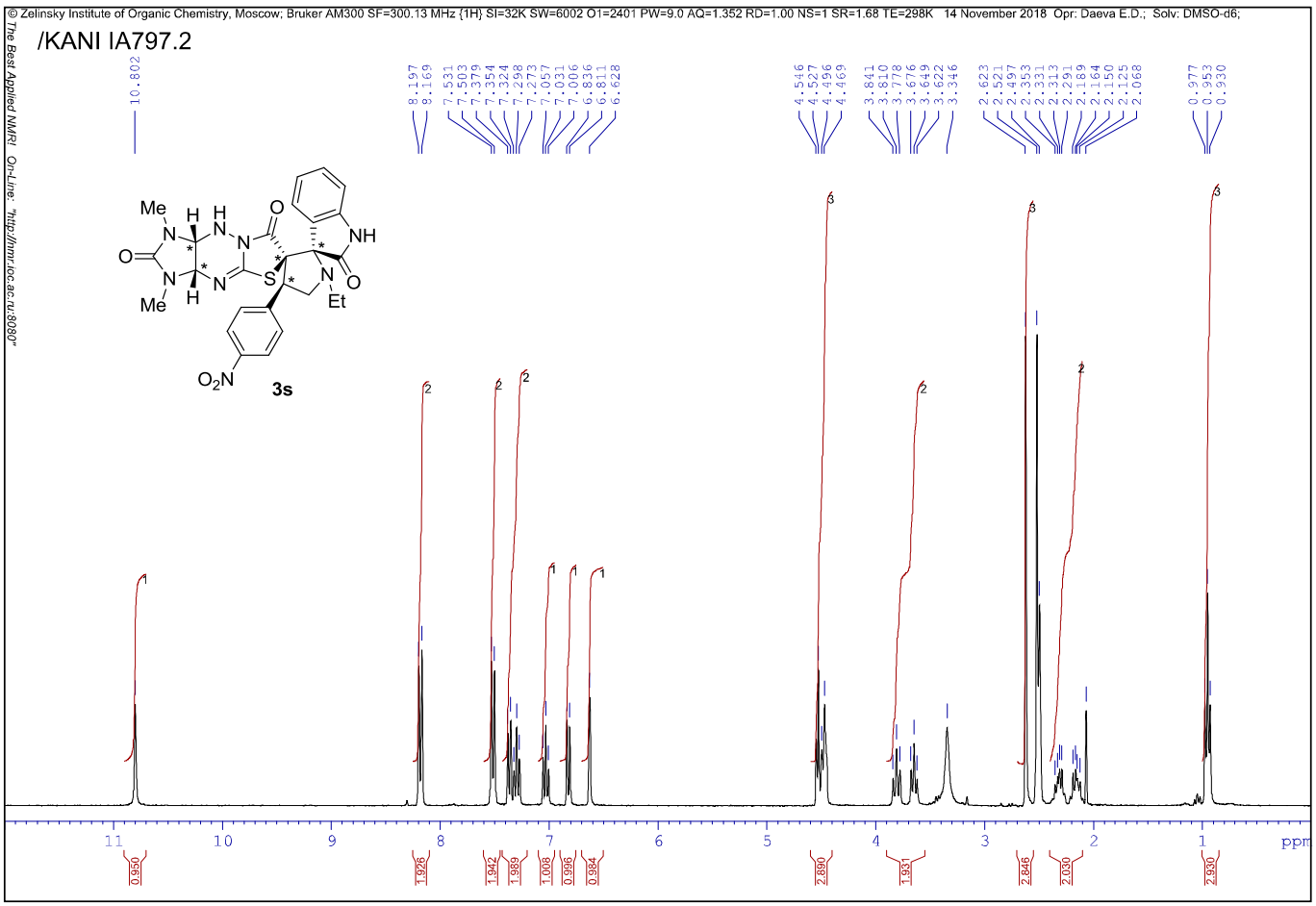
^{1}H NMR spectrum of **3r**



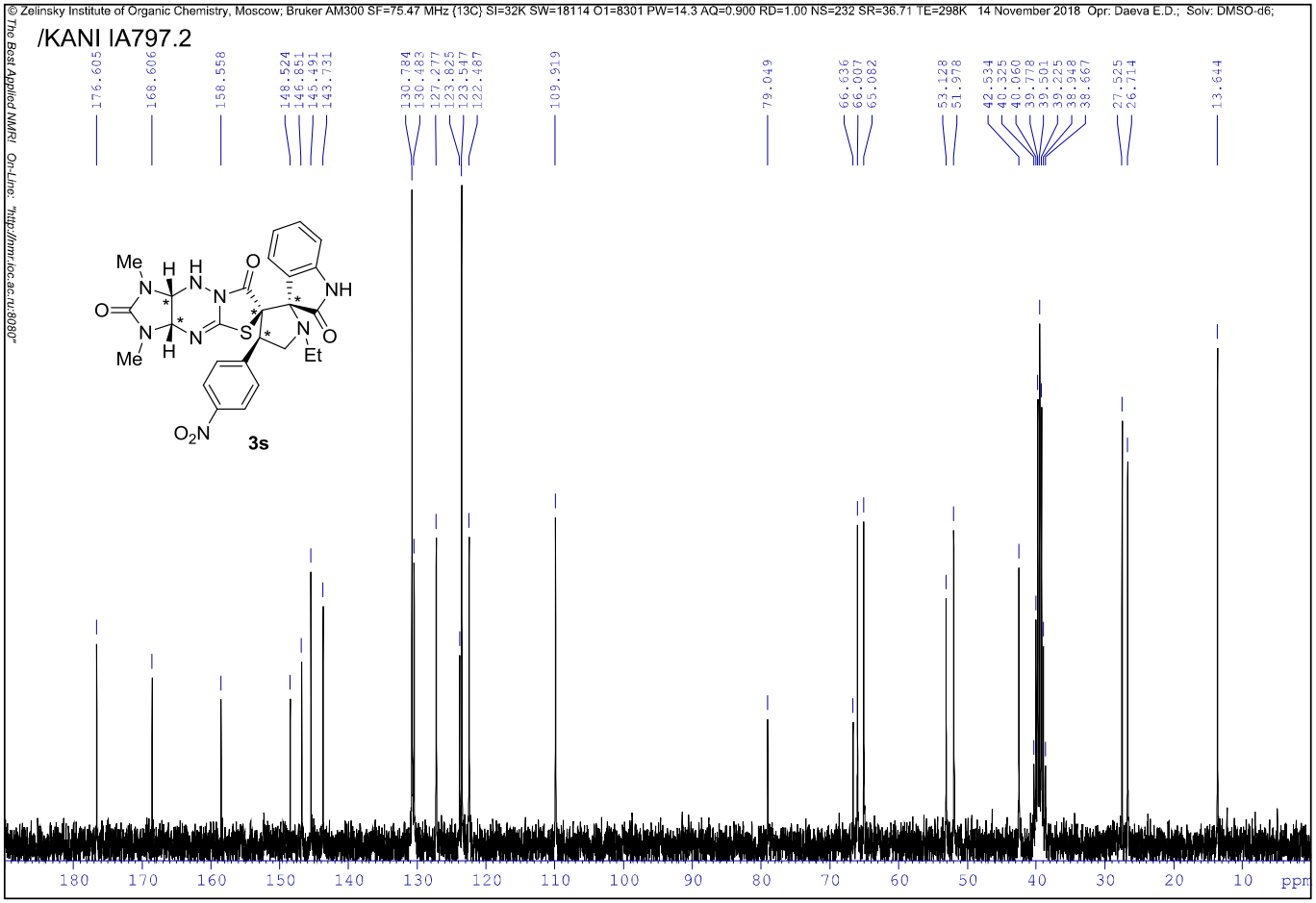
^{13}C NMR spectrum of **3r**



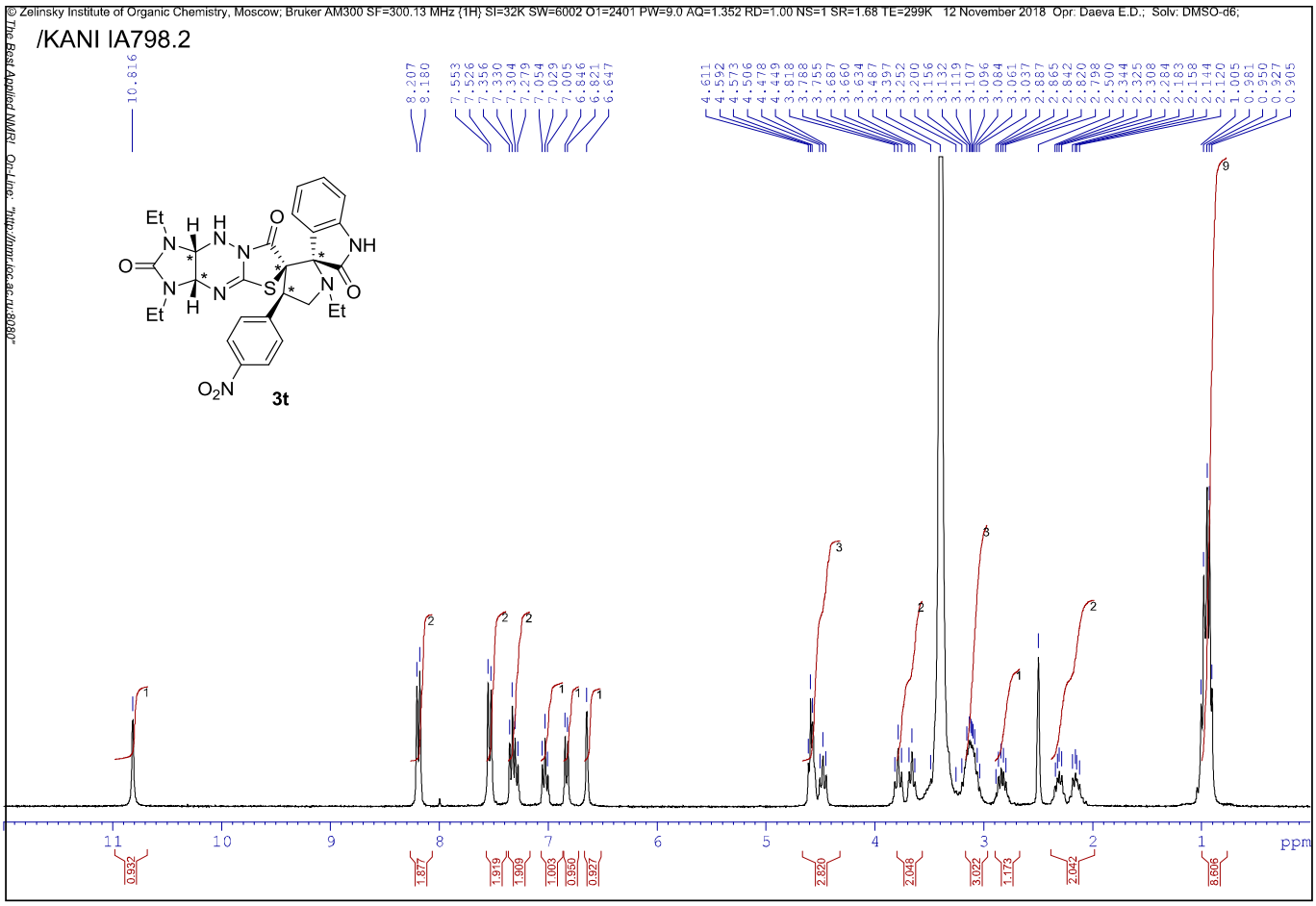
^{1}H NMR spectrum of **3s**



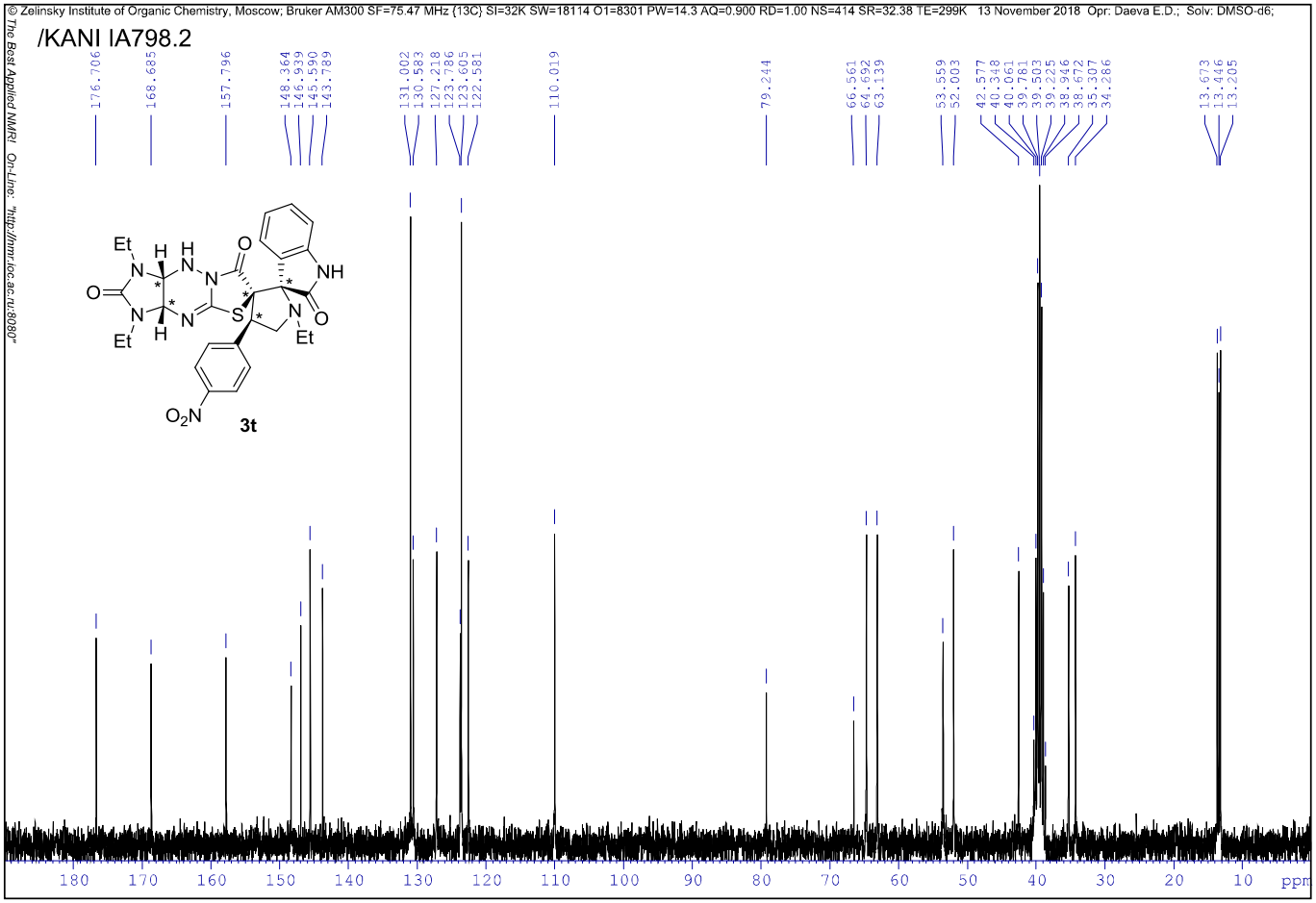
^{13}C NMR spectrum of **3s**



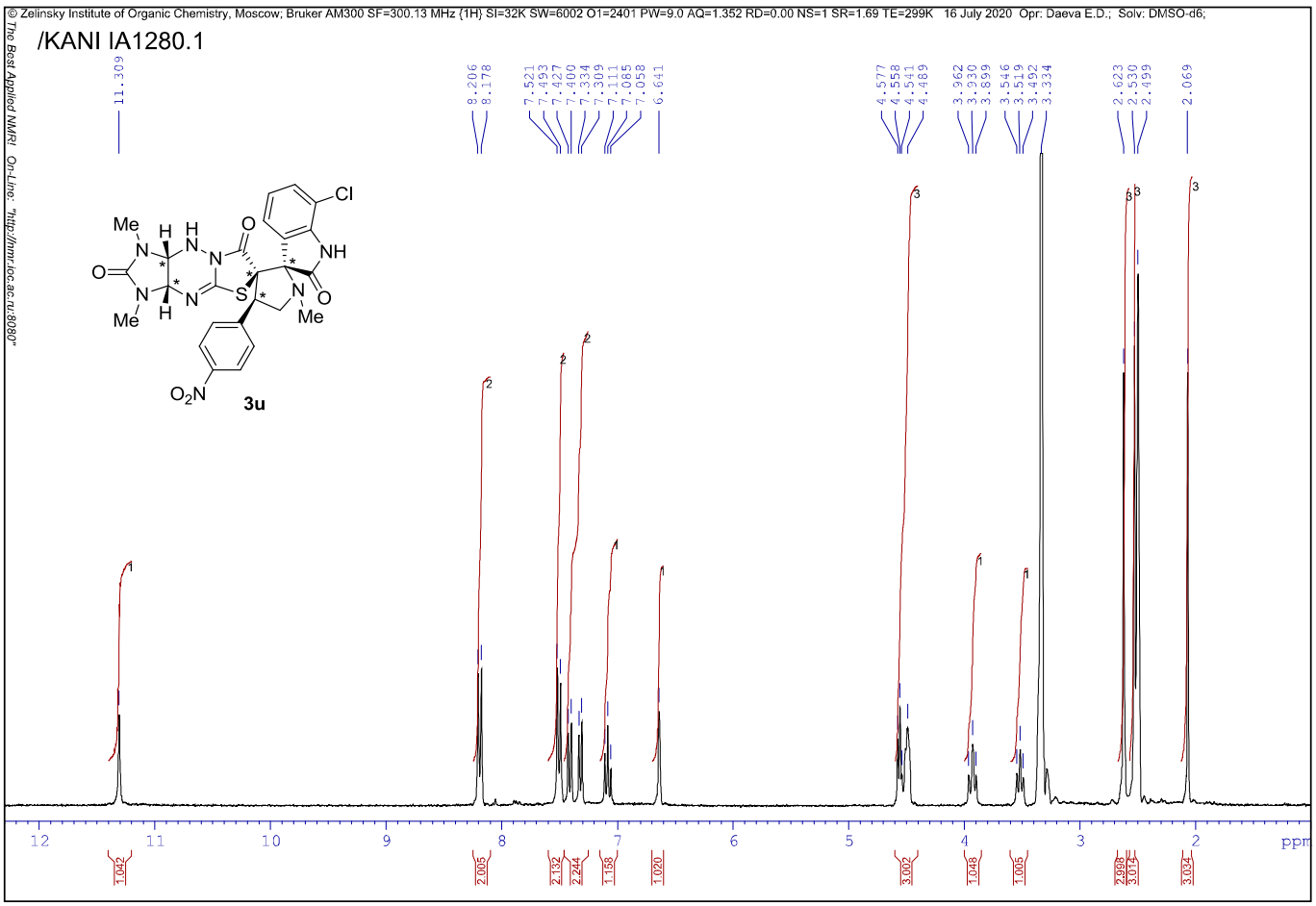
^{1}H NMR spectrum of **3t**



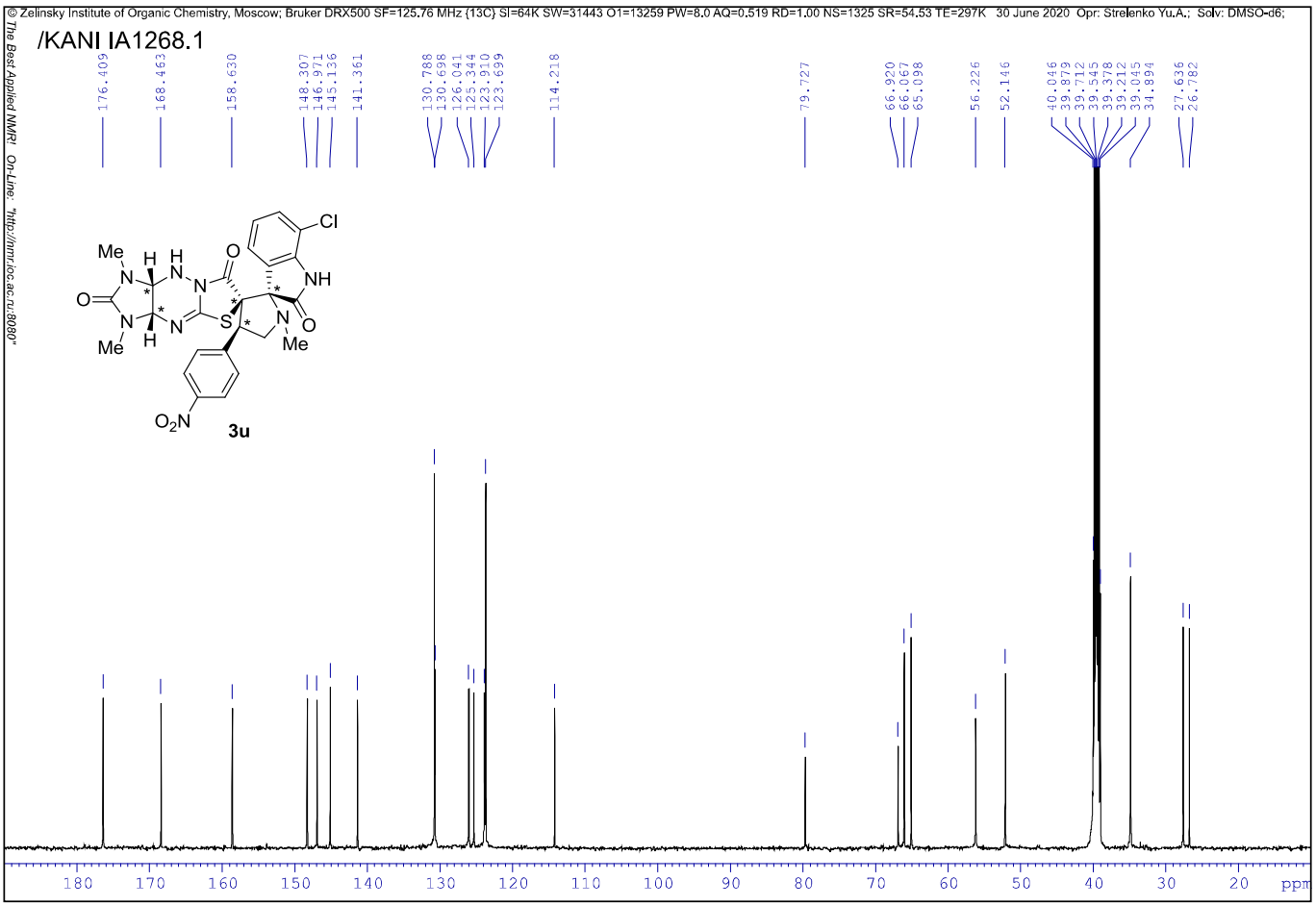
^{13}C NMR spectrum of **3t**



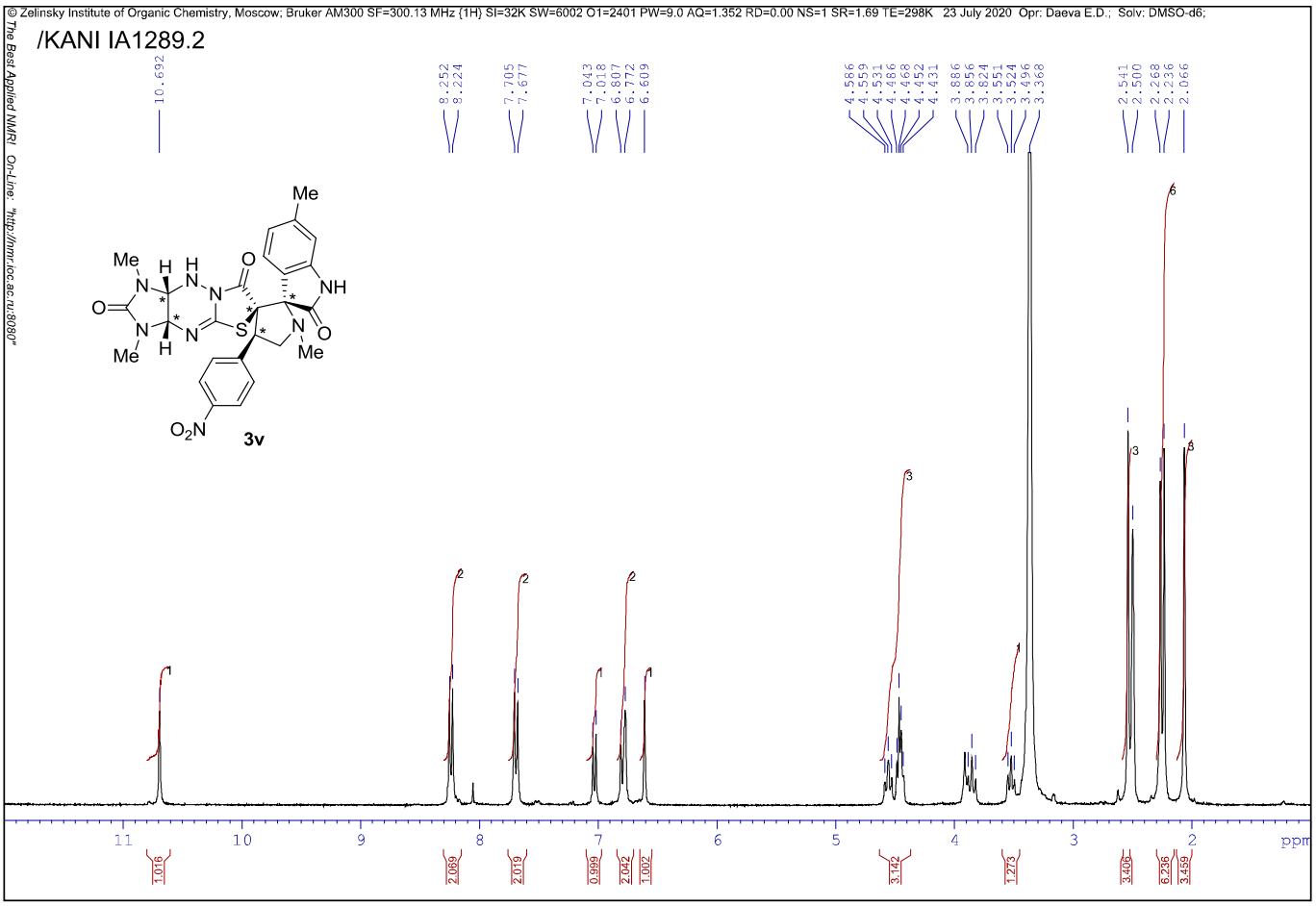
^{1}H NMR spectrum of **3u**



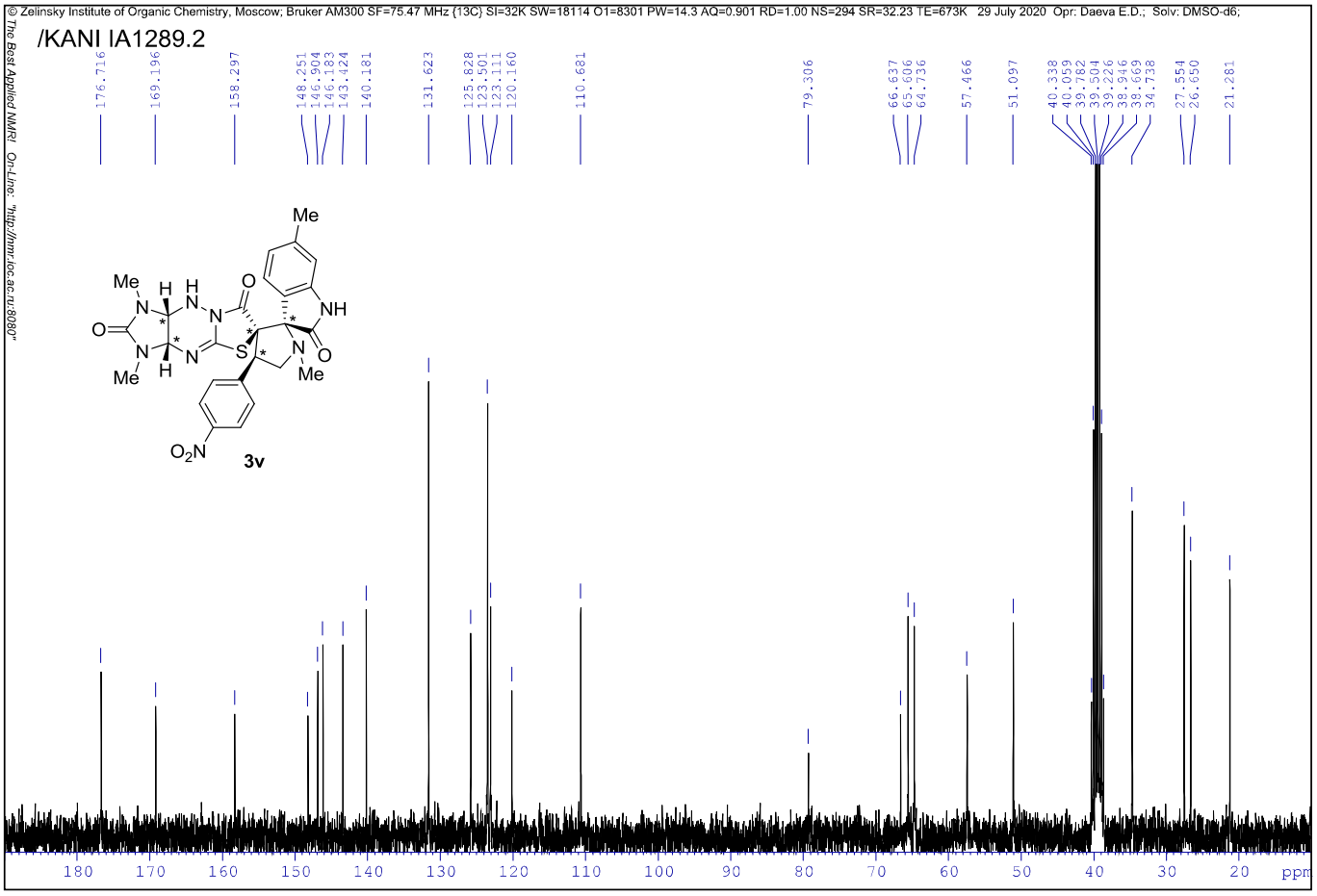
^{13}C NMR spectrum of **3u**



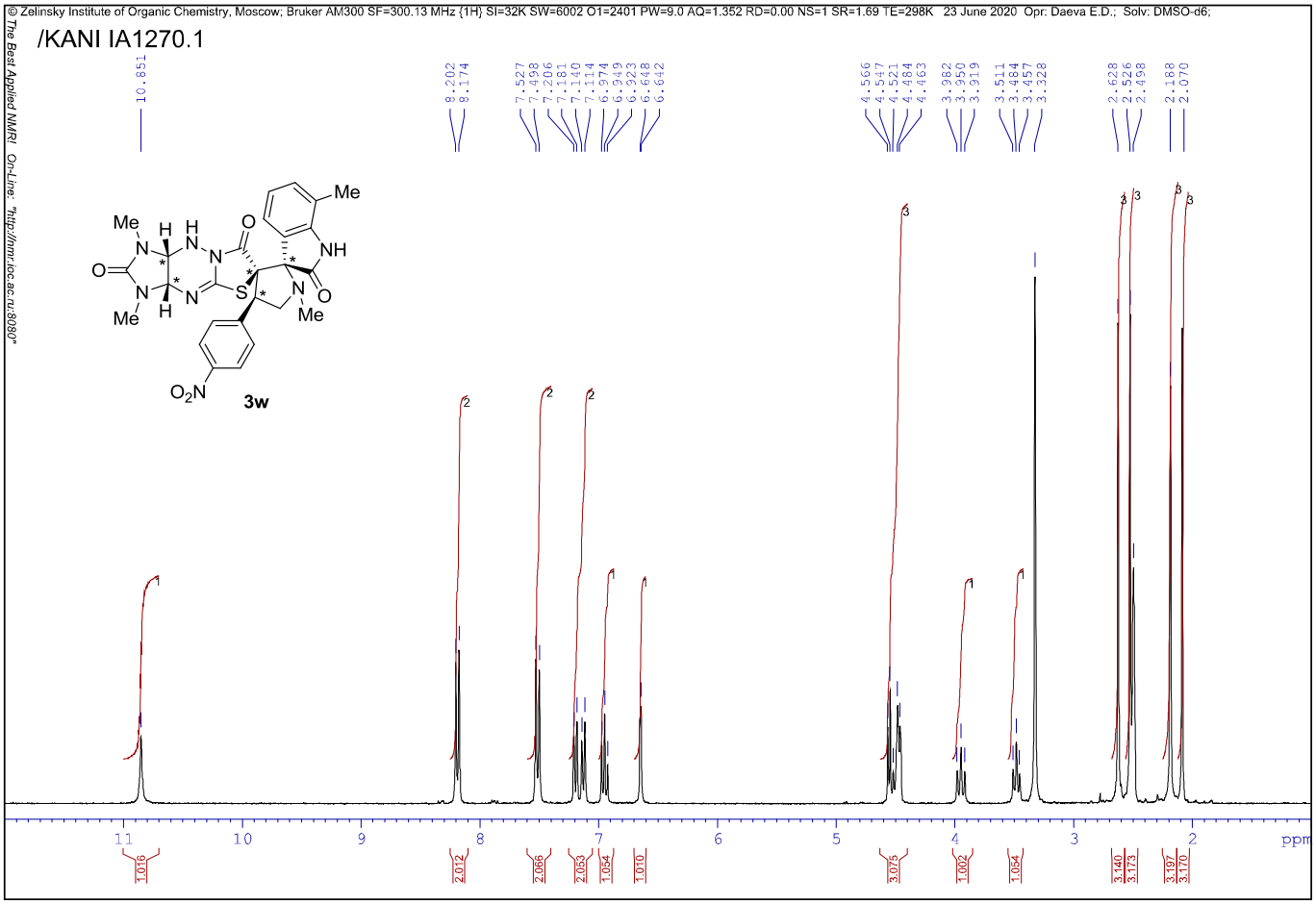
^{1}H NMR spectrum of **3v**



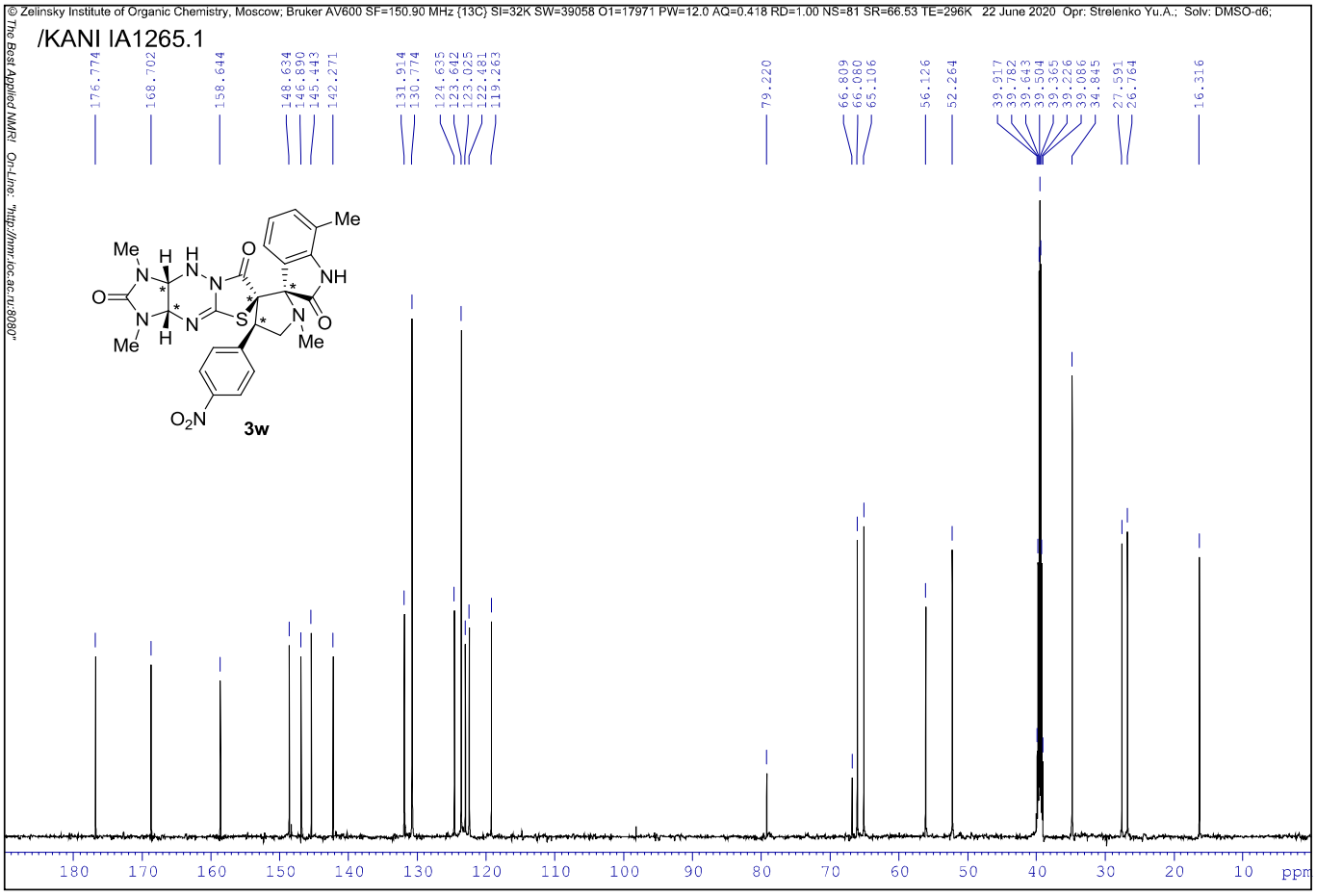
^{13}C NMR spectrum of **3v**



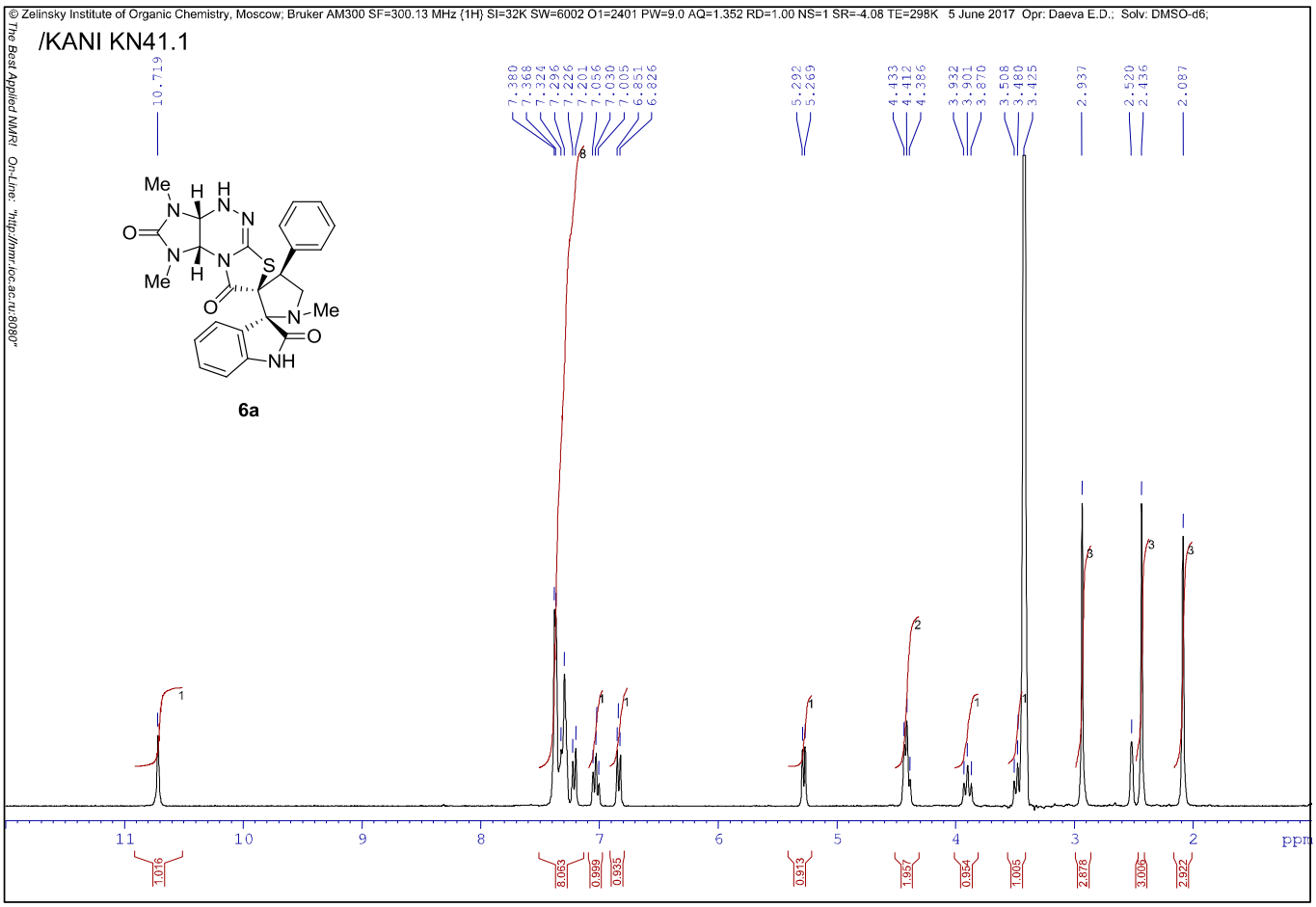
^{1}H NMR spectrum of **3w**



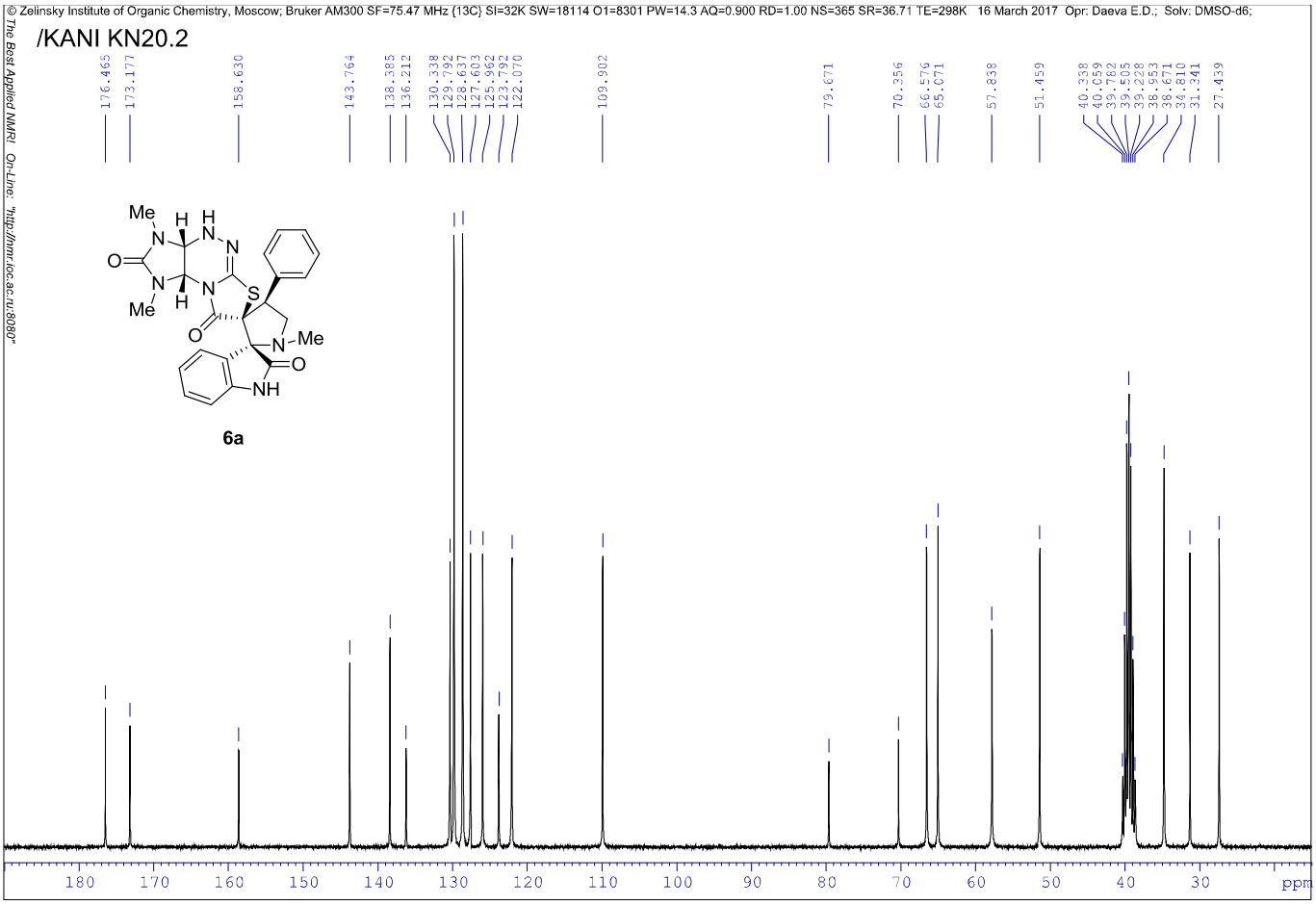
^{13}C NMR spectrum of **3w**



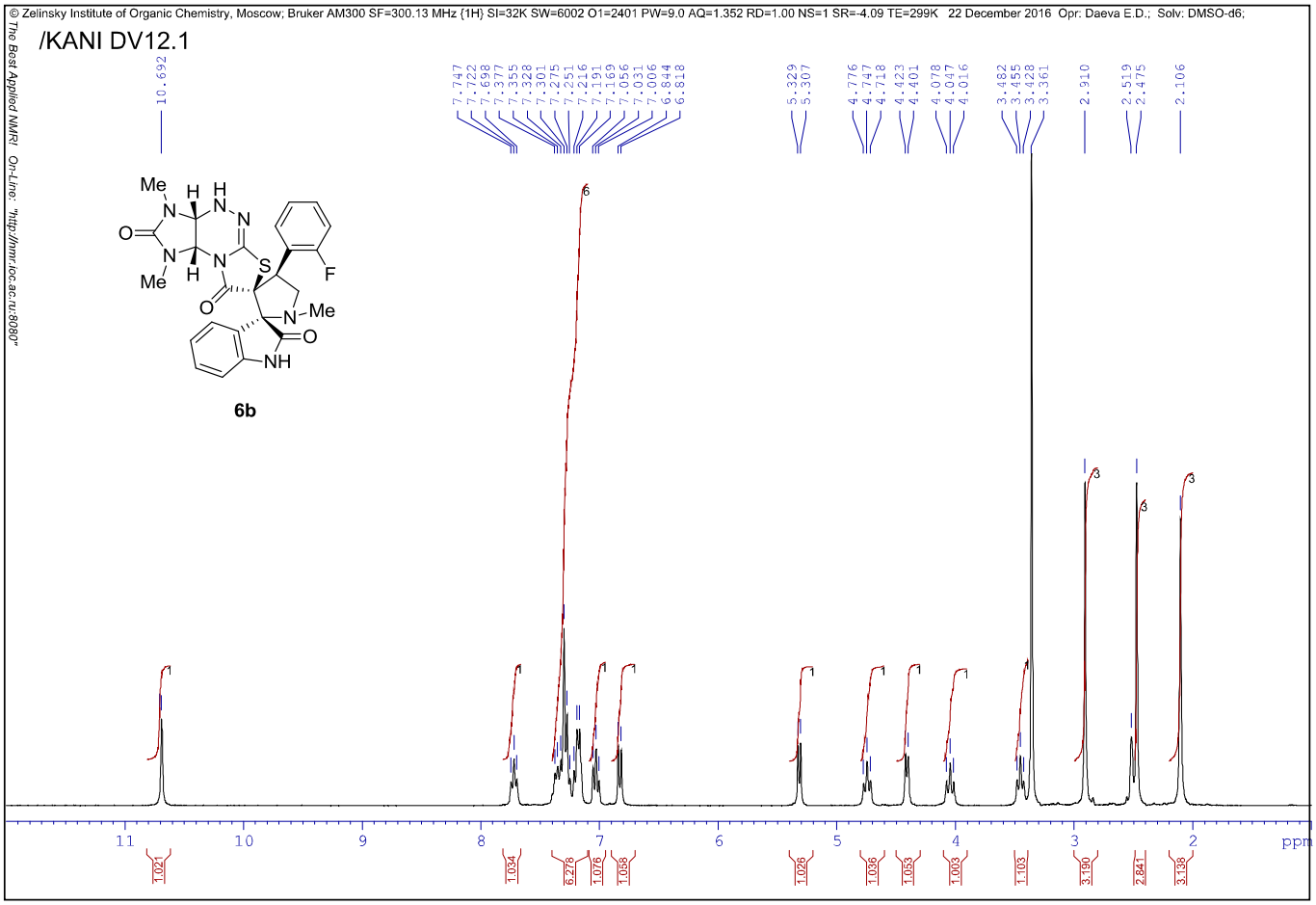
^{1}H NMR spectrum of **6a**



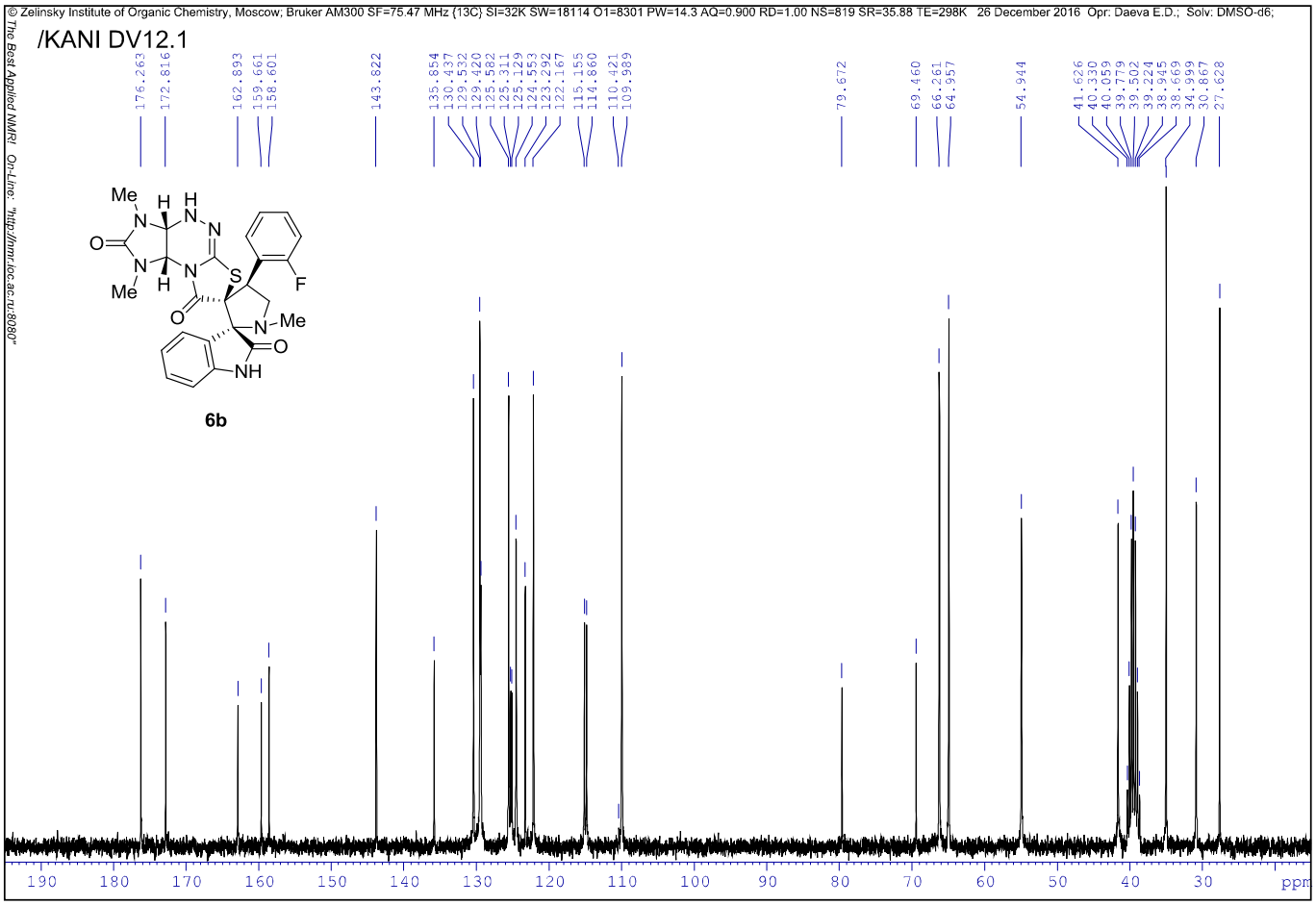
^{13}C NMR spectrum of **6a**



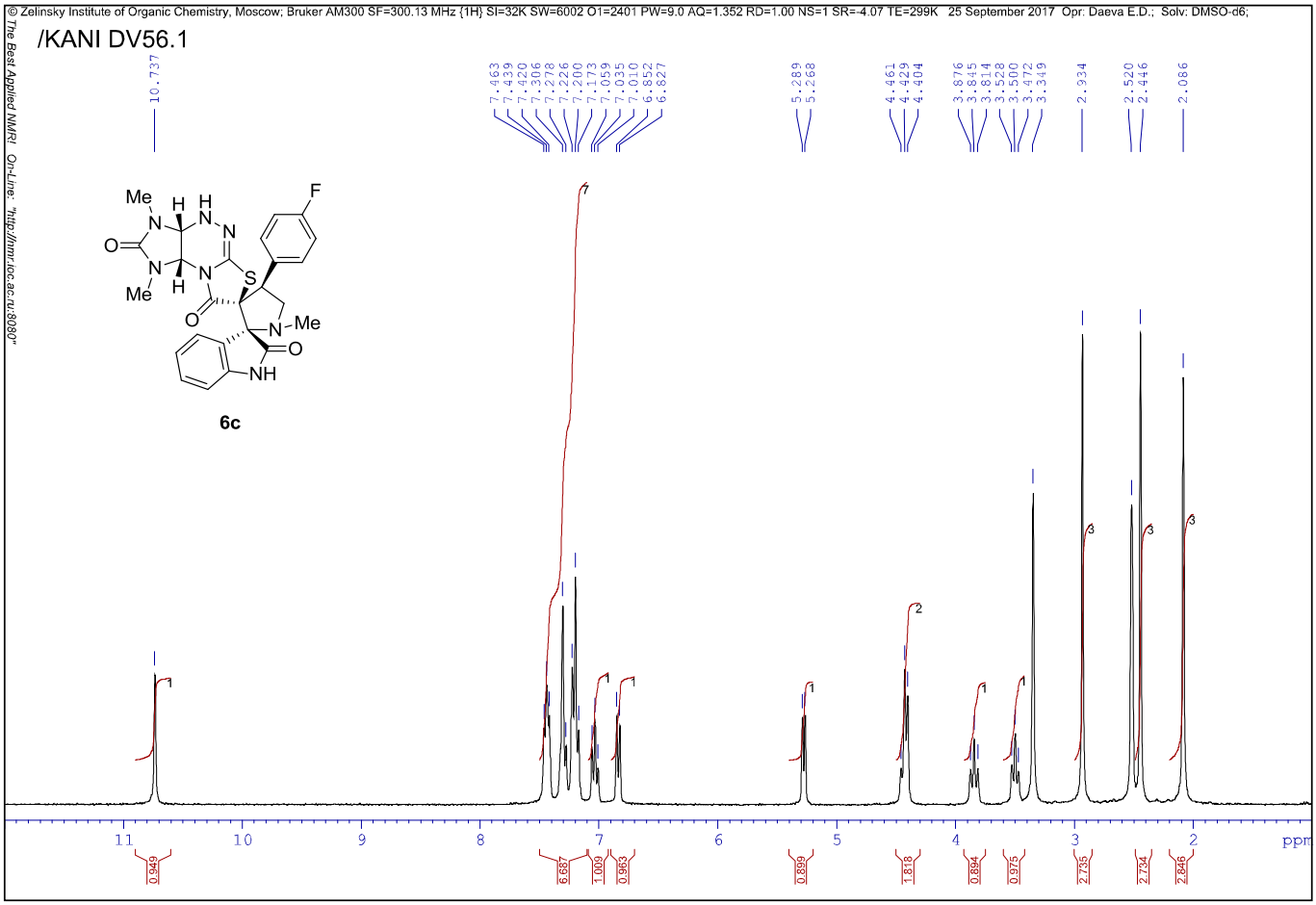
^{1}H NMR spectrum of **6b**



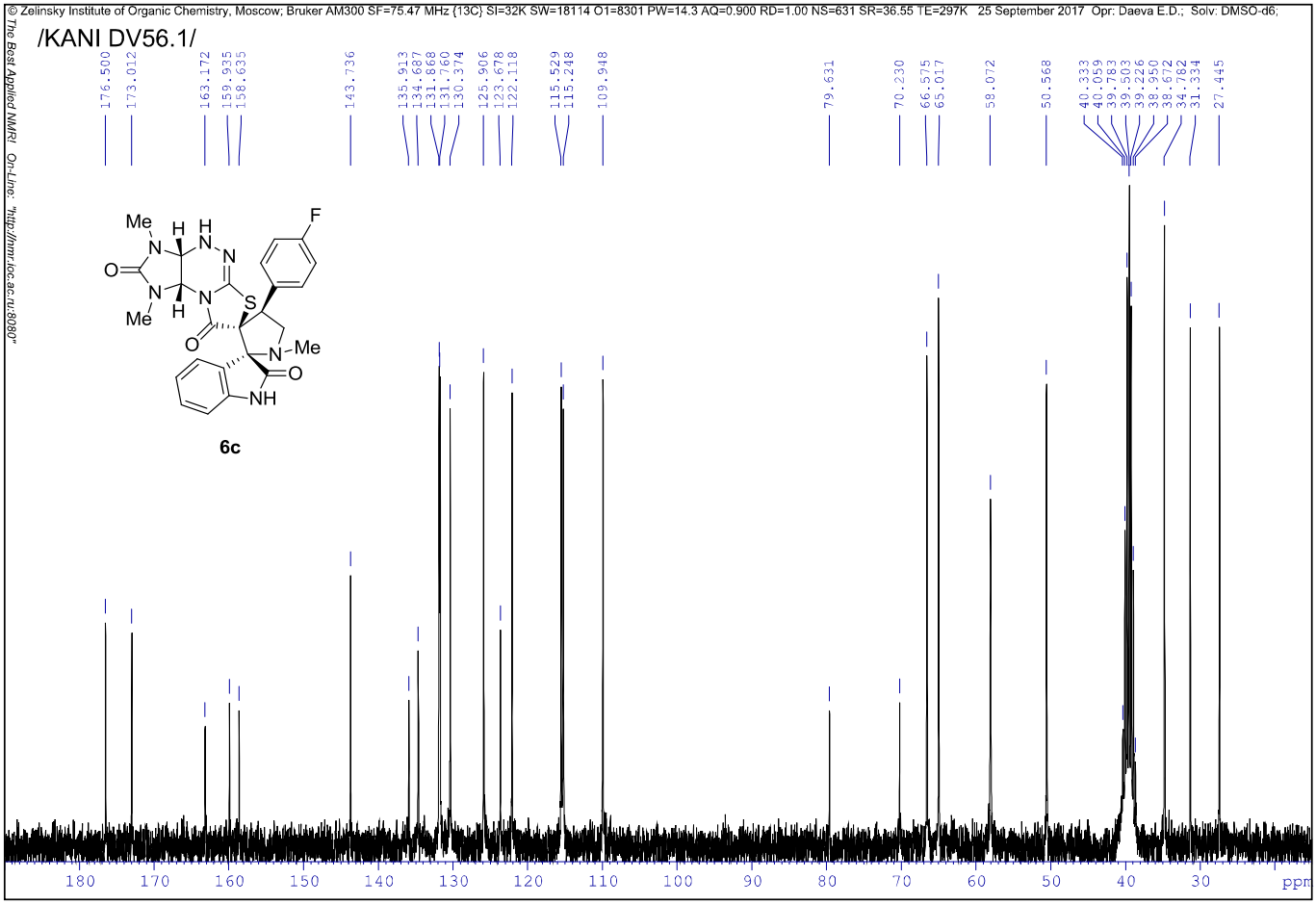
^{13}C NMR spectrum of **6b**



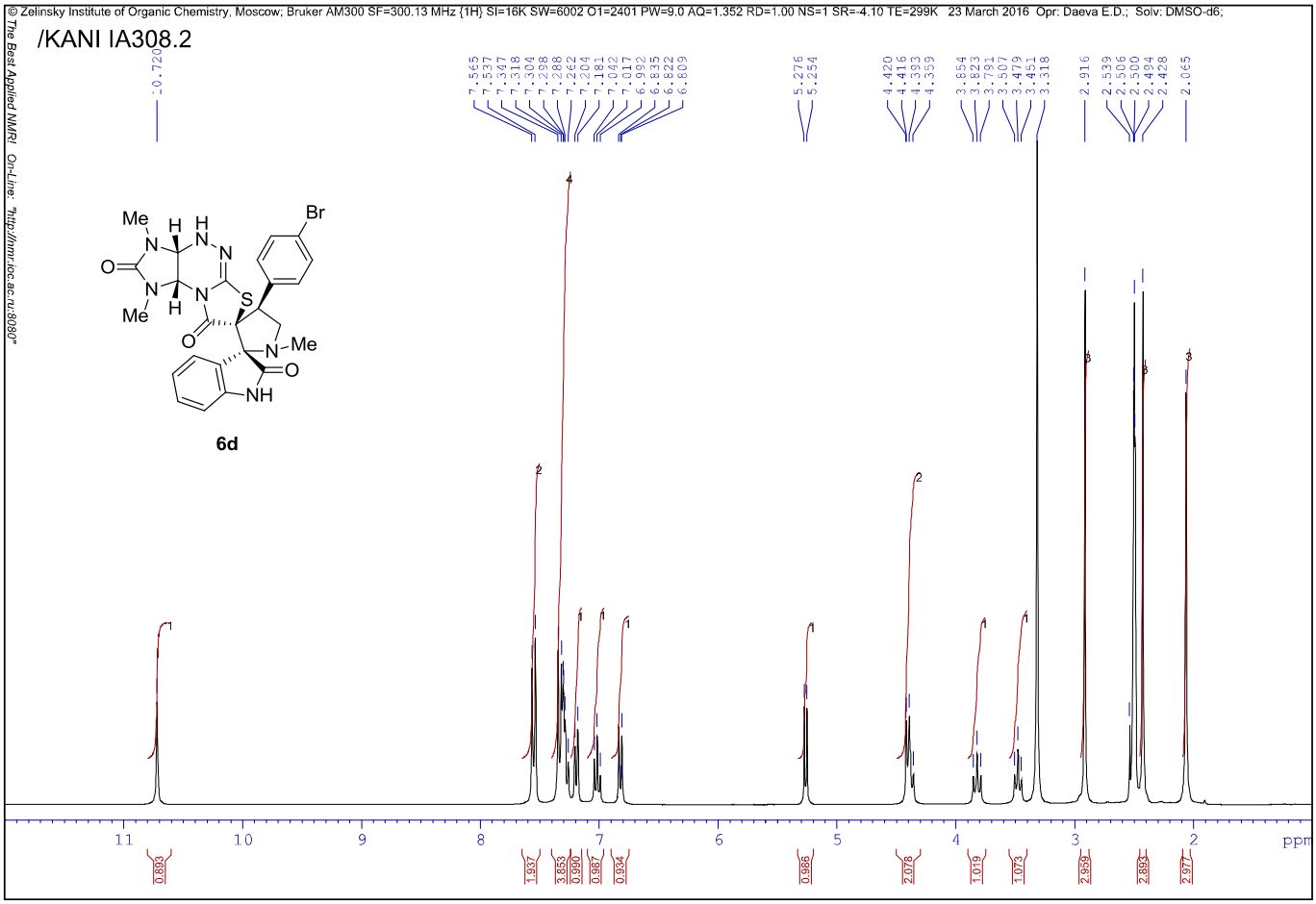
^{1}H NMR spectrum of **6c**



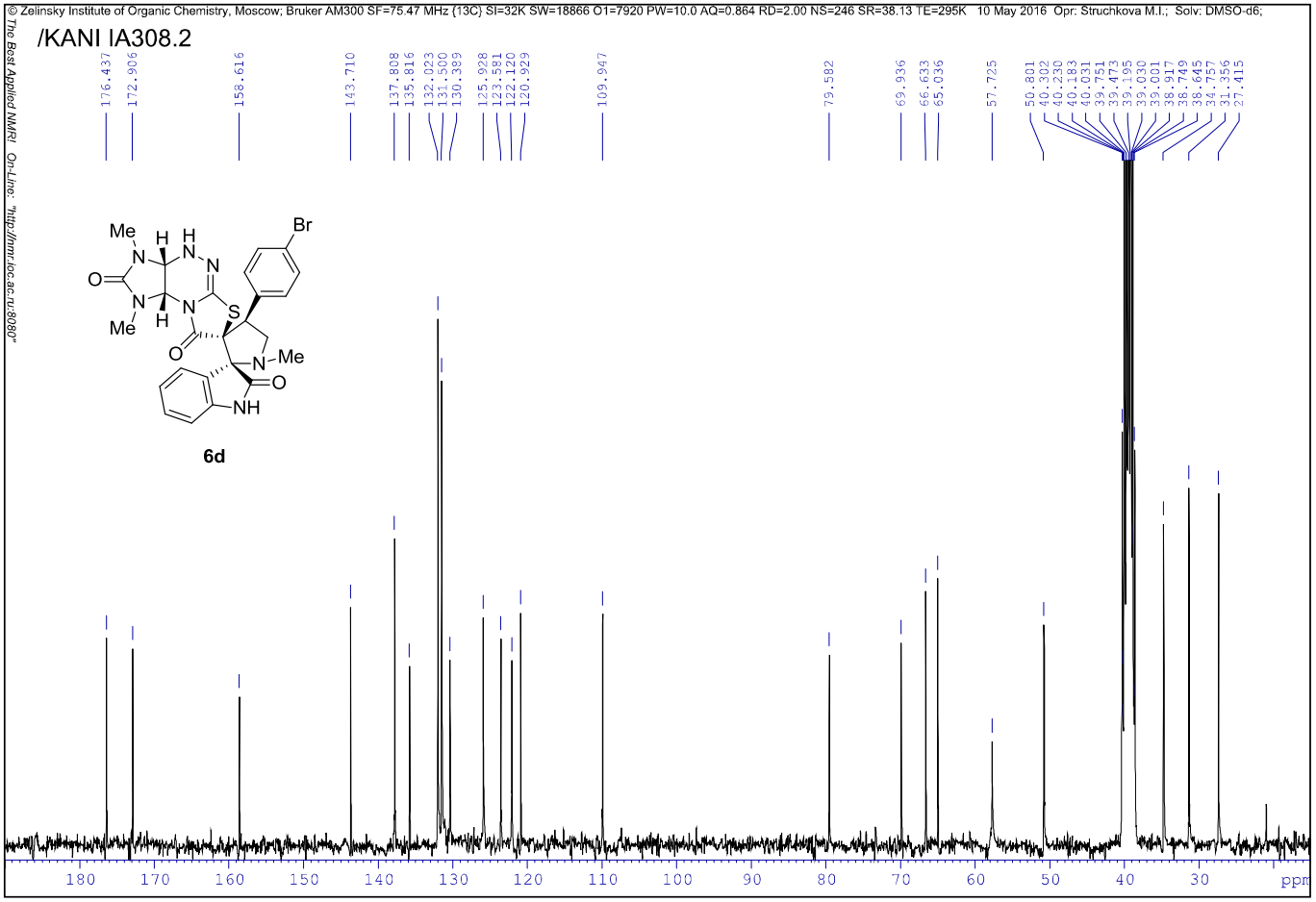
^{13}C NMR spectrum of **6c**



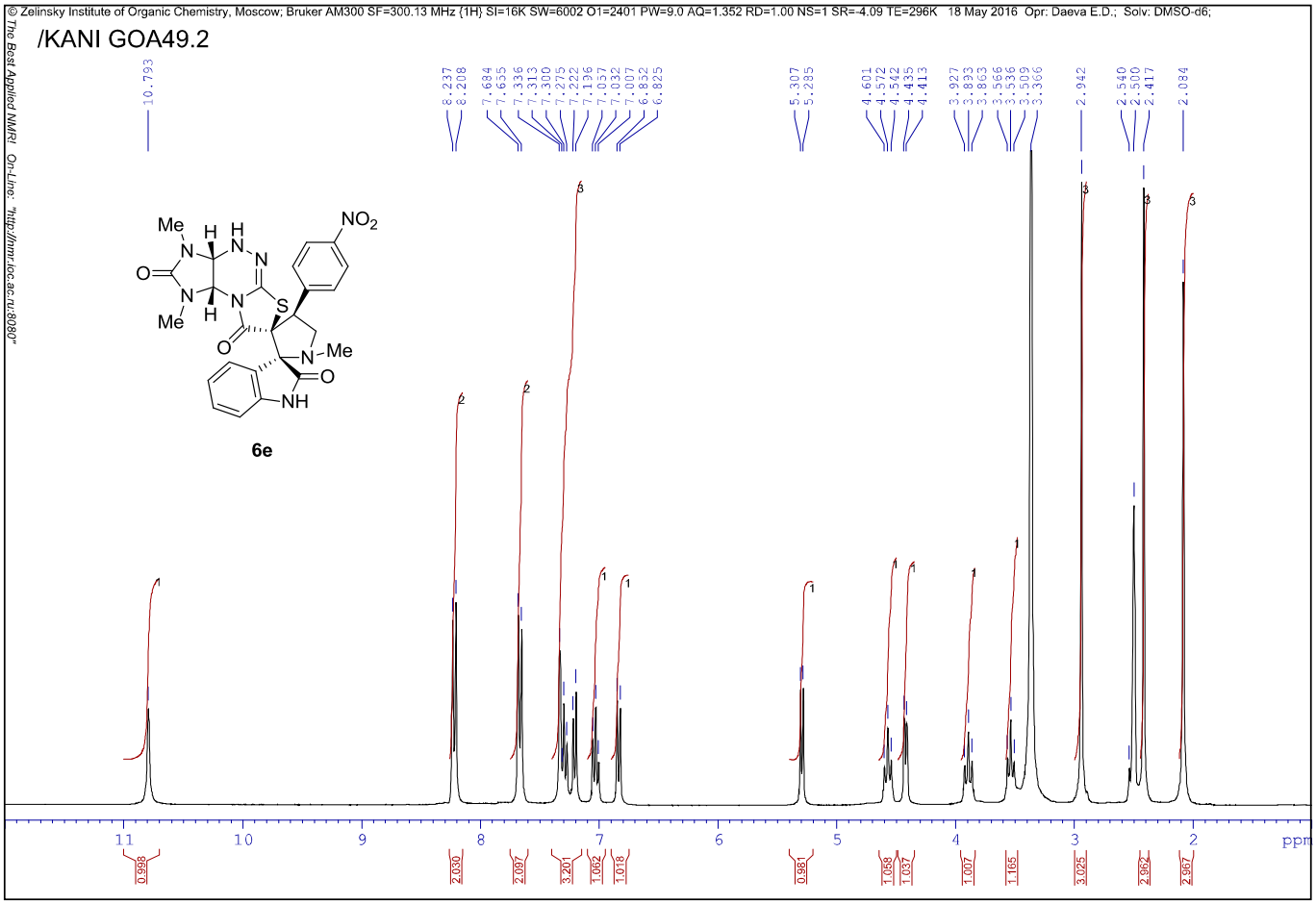
^{1}H NMR spectrum of **6d**



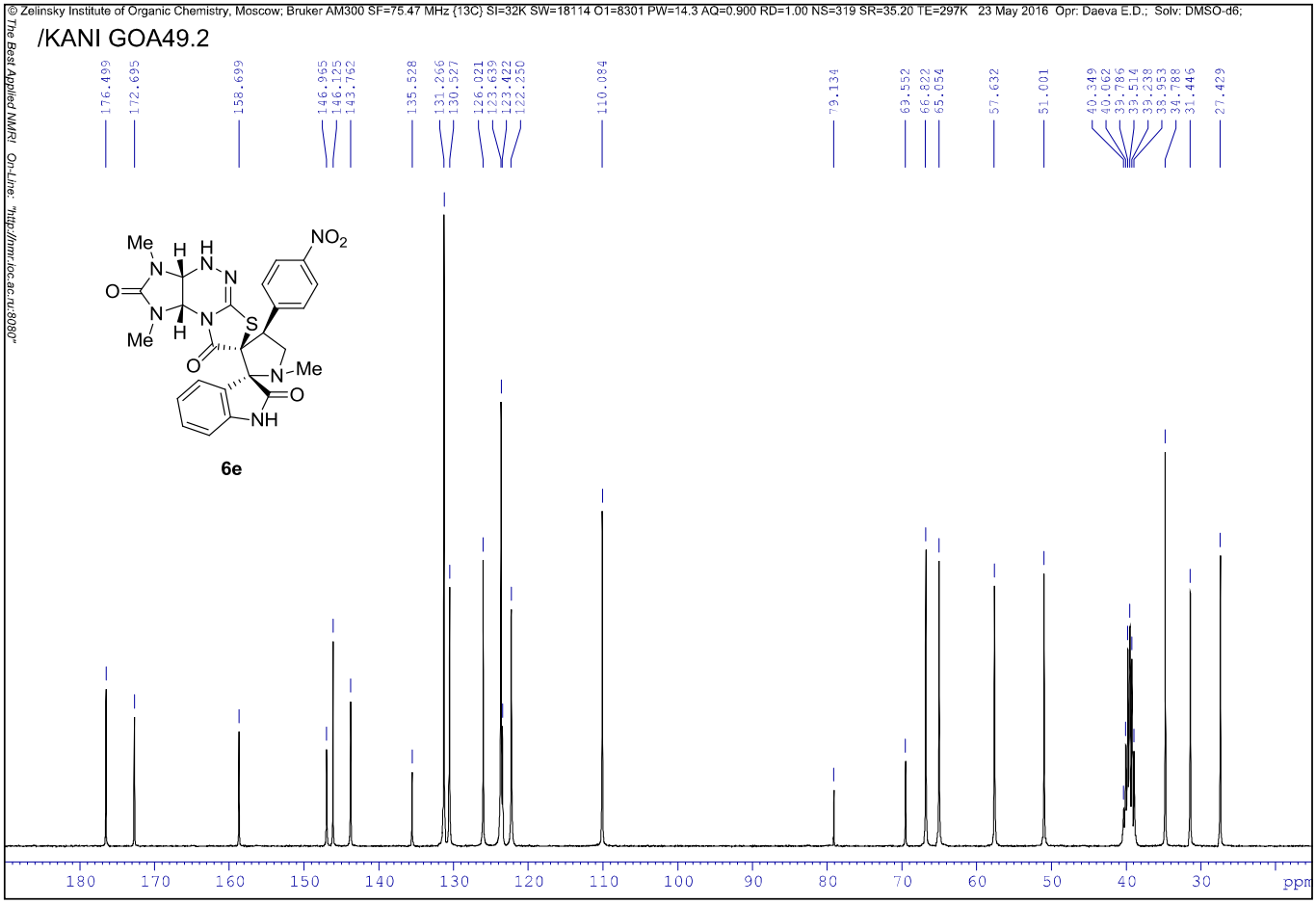
^{13}C NMR spectrum of **6d**



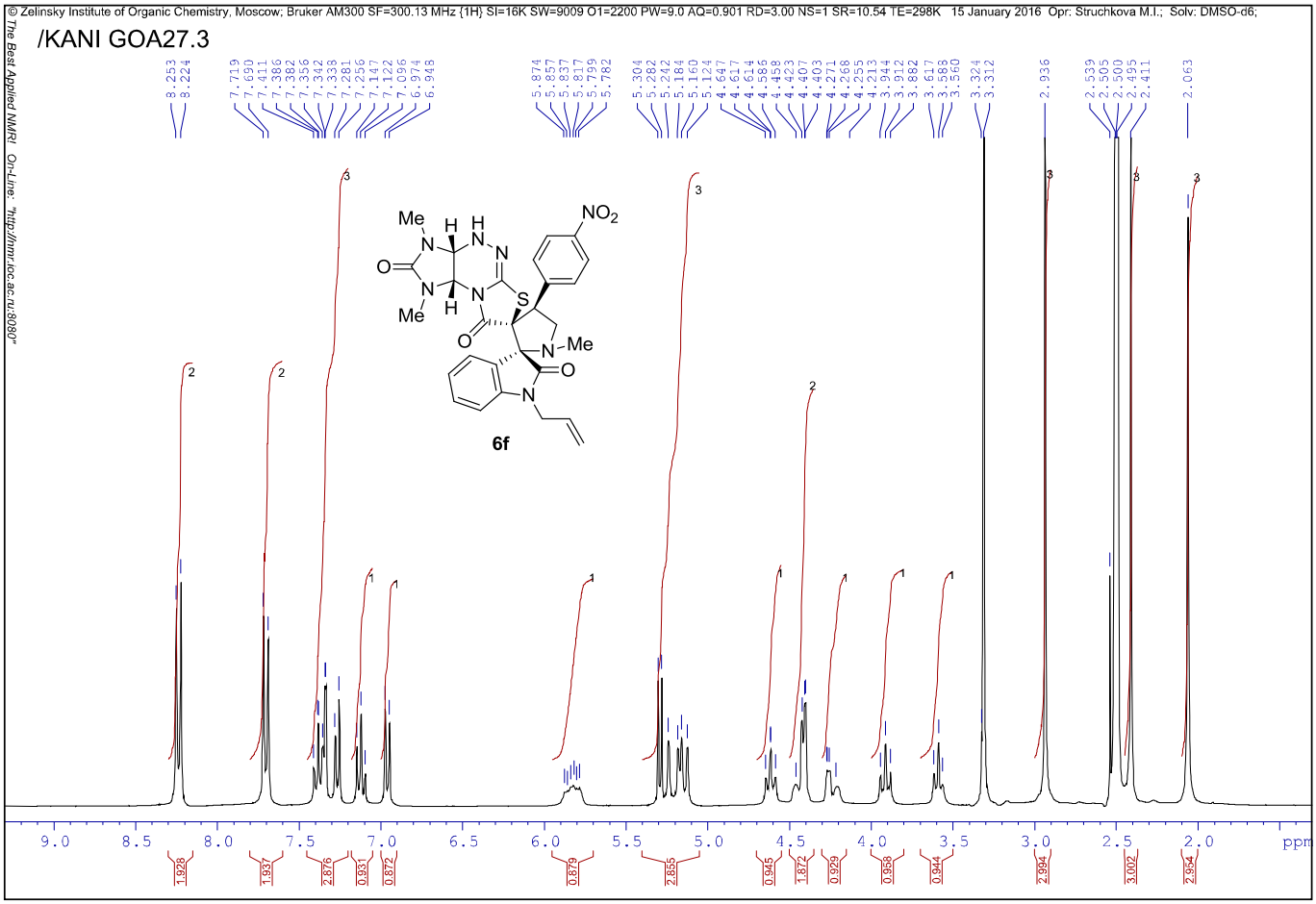
^{1}H NMR spectrum of **6e**



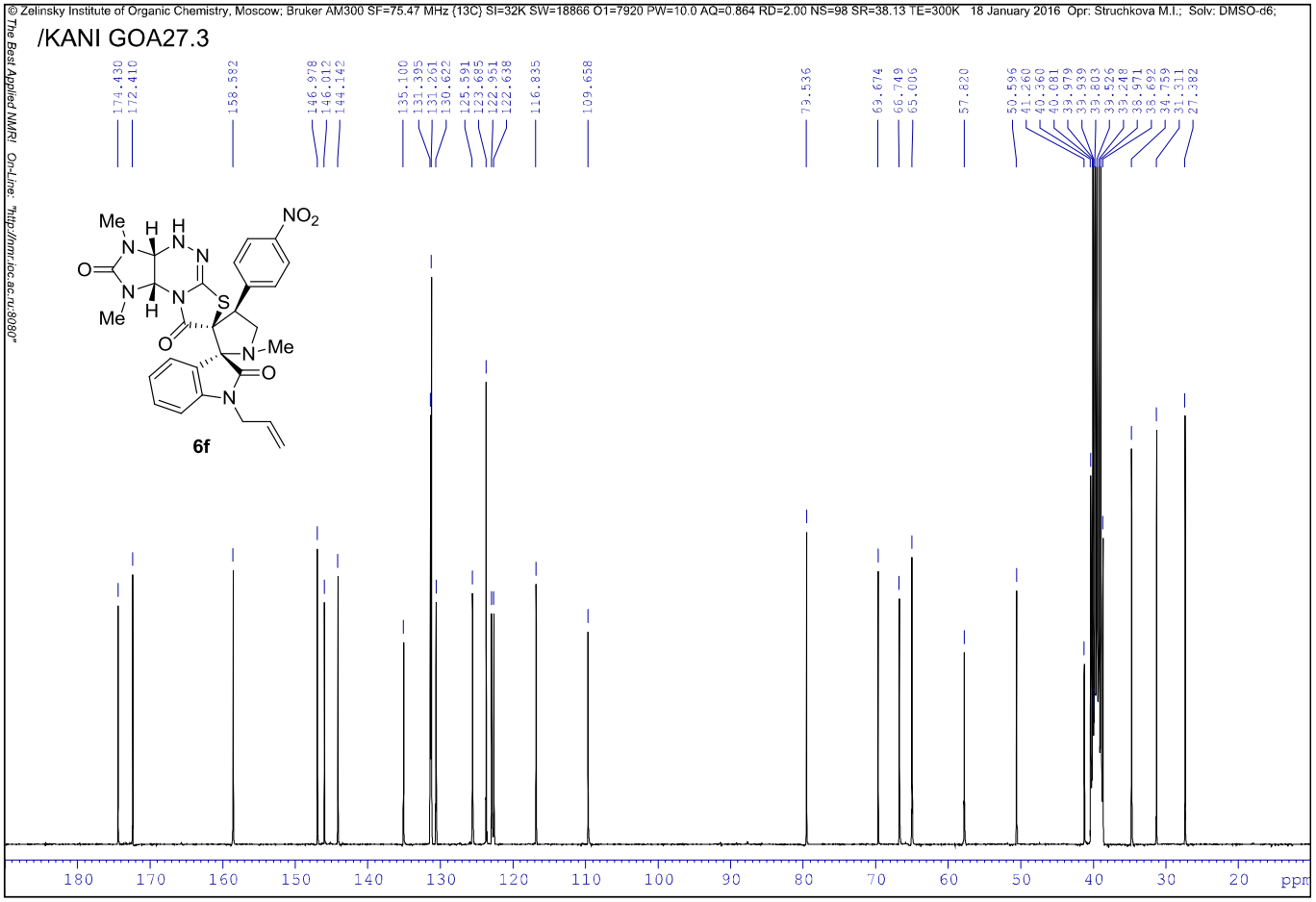
^{13}C NMR spectrum of **6e**



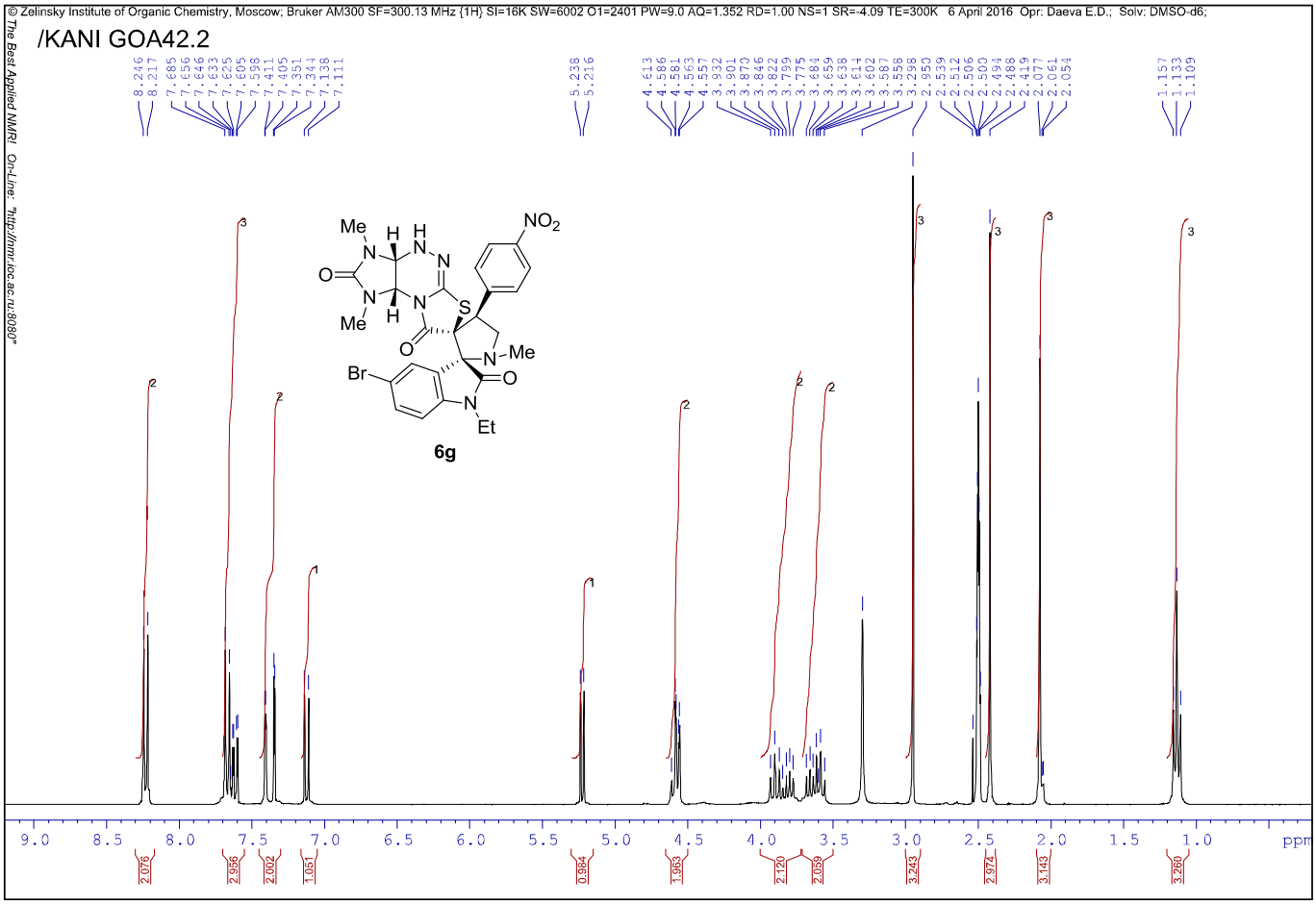
^{1}H NMR spectrum of **6f**



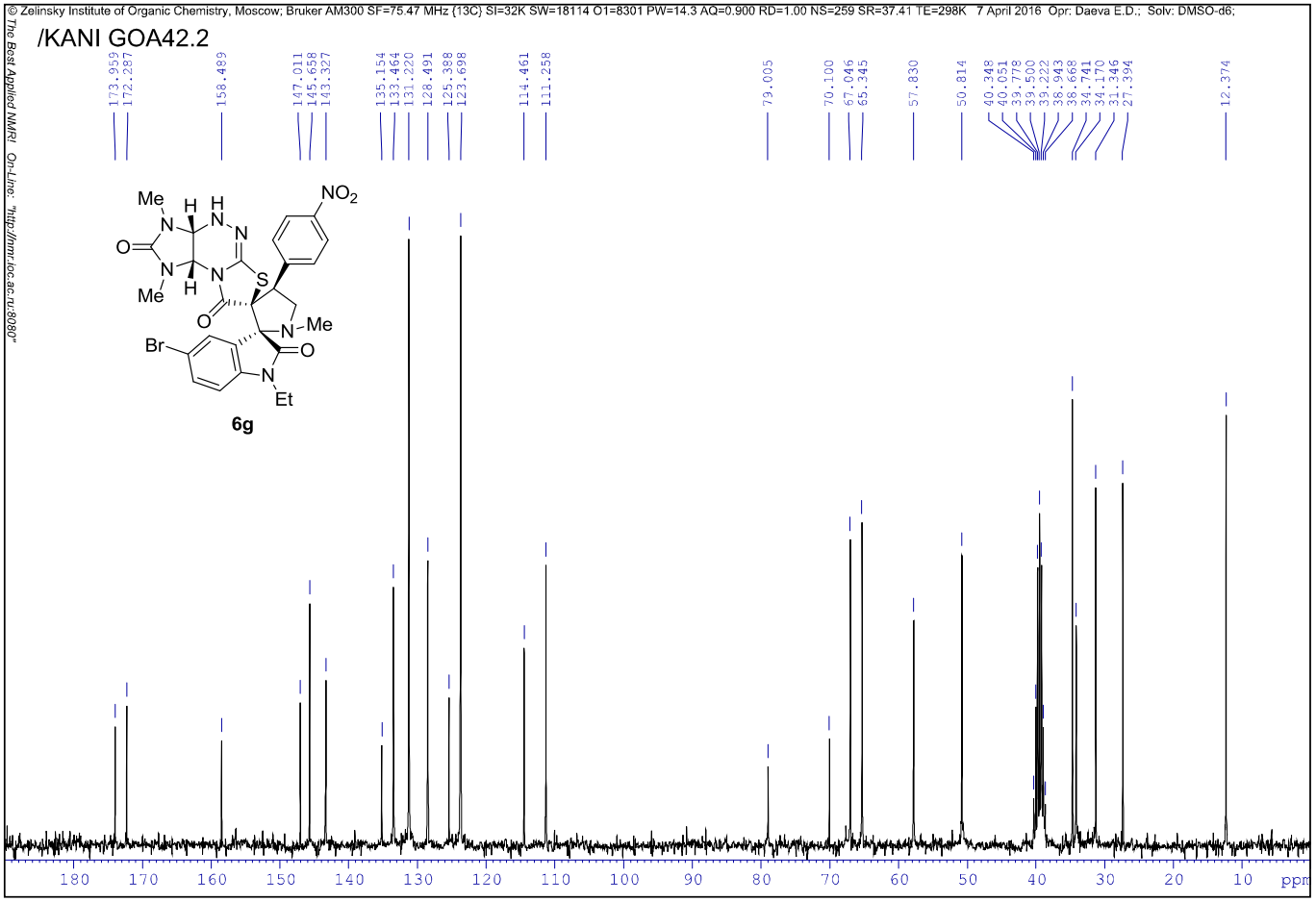
^{13}C NMR spectrum of **6f**



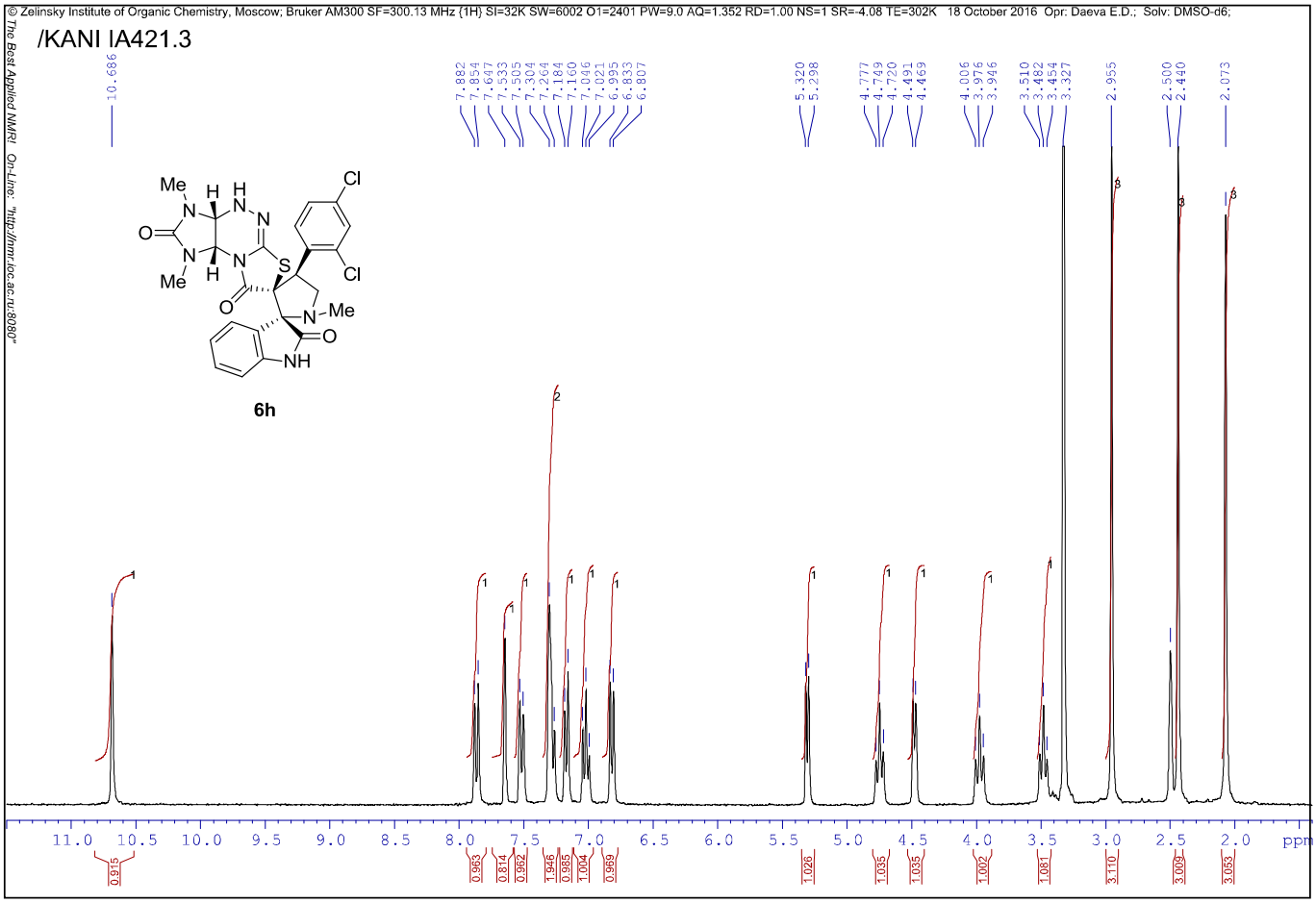
^{1}H NMR spectrum of **6g**



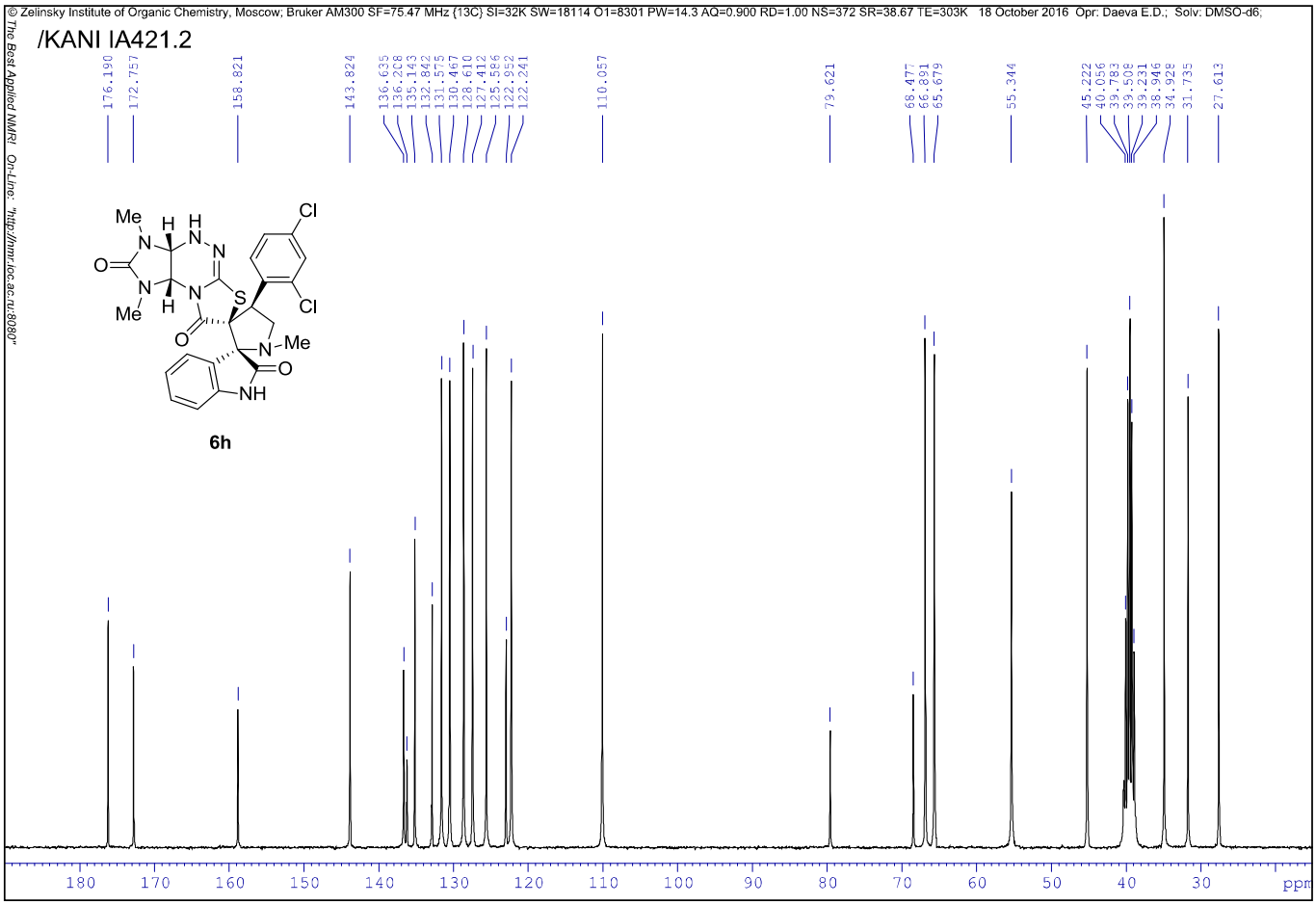
^{13}C NMR spectrum of **6g**



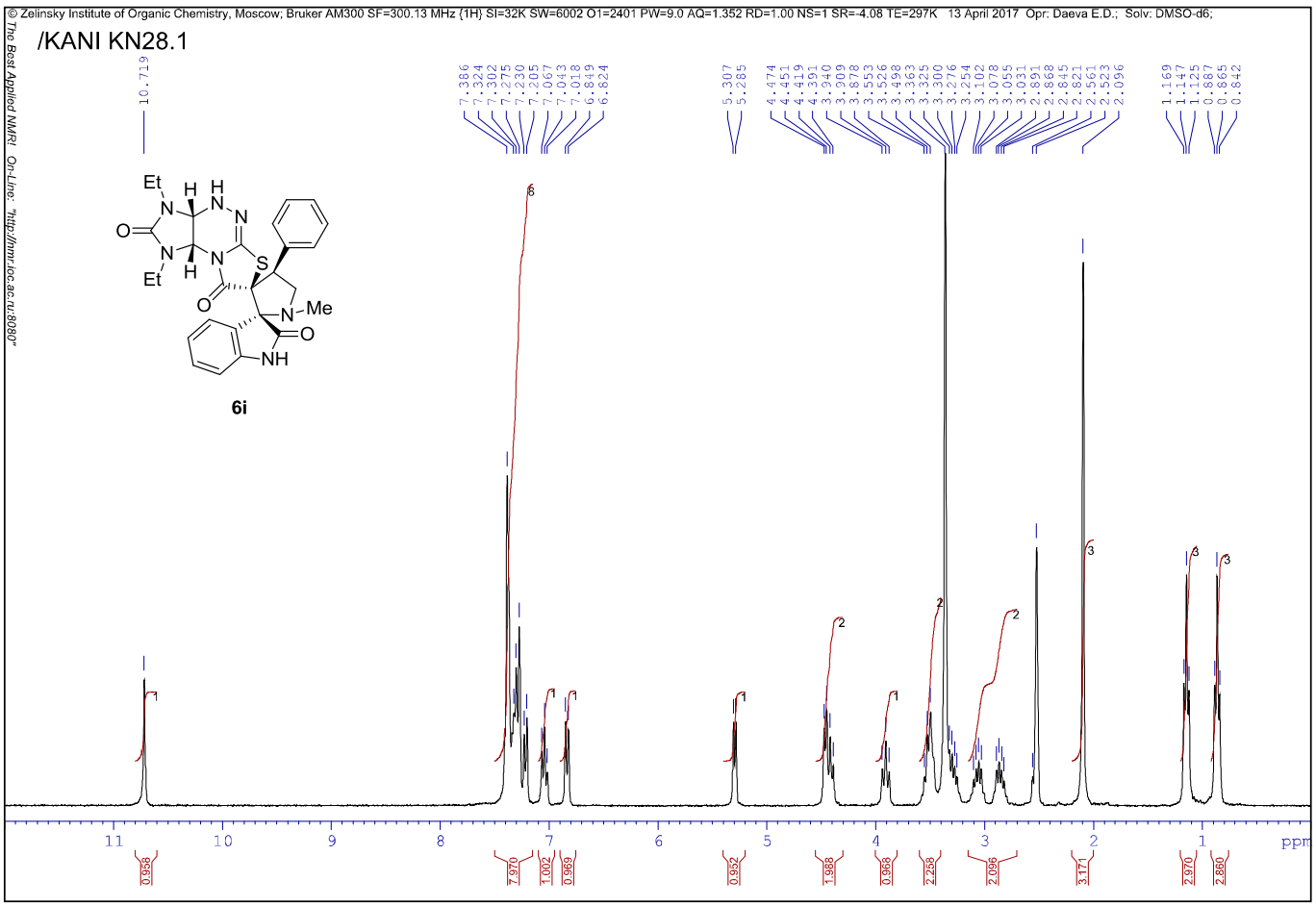
^{1}H NMR spectrum of **6h**



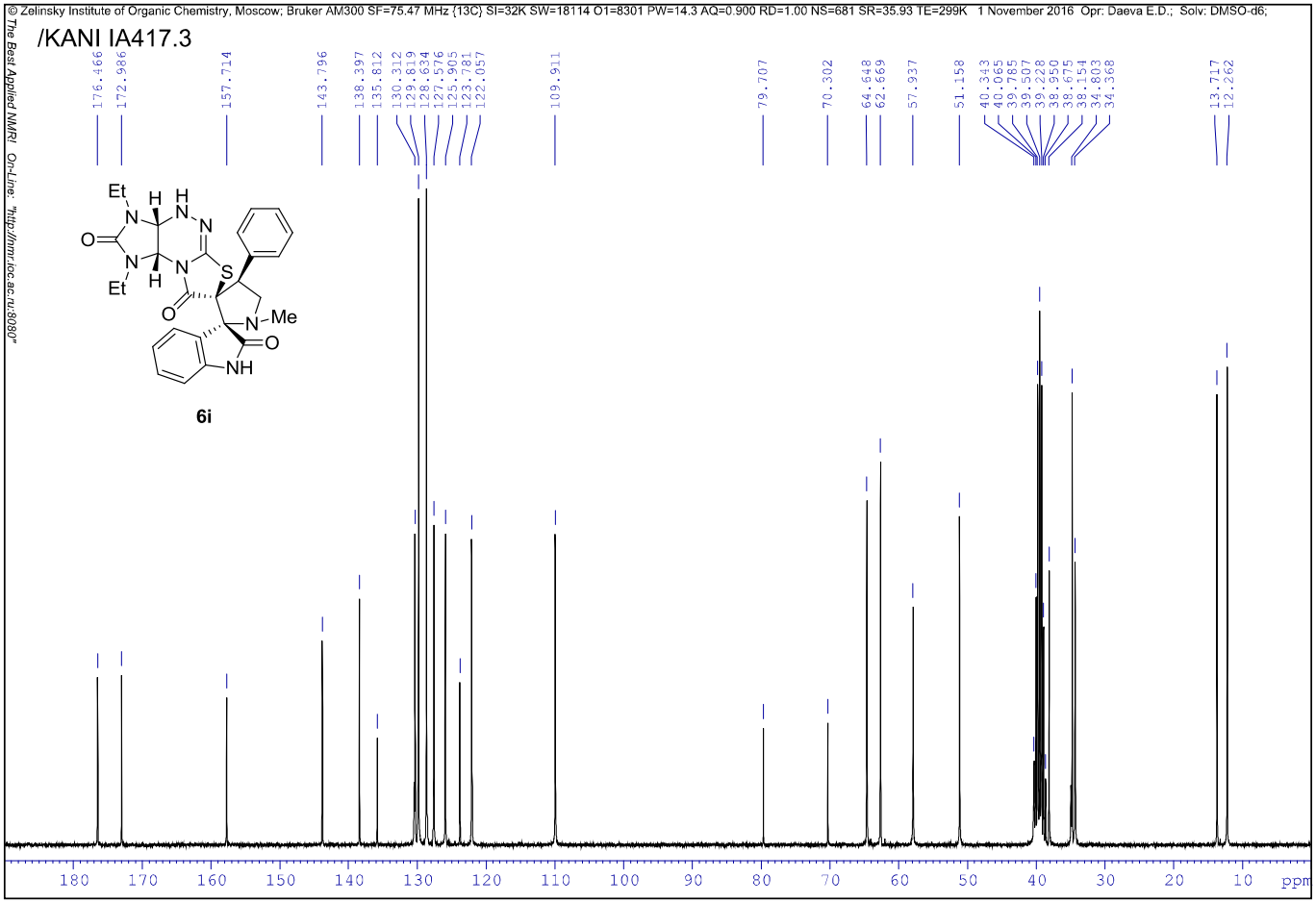
^{13}C NMR spectrum of **6h**



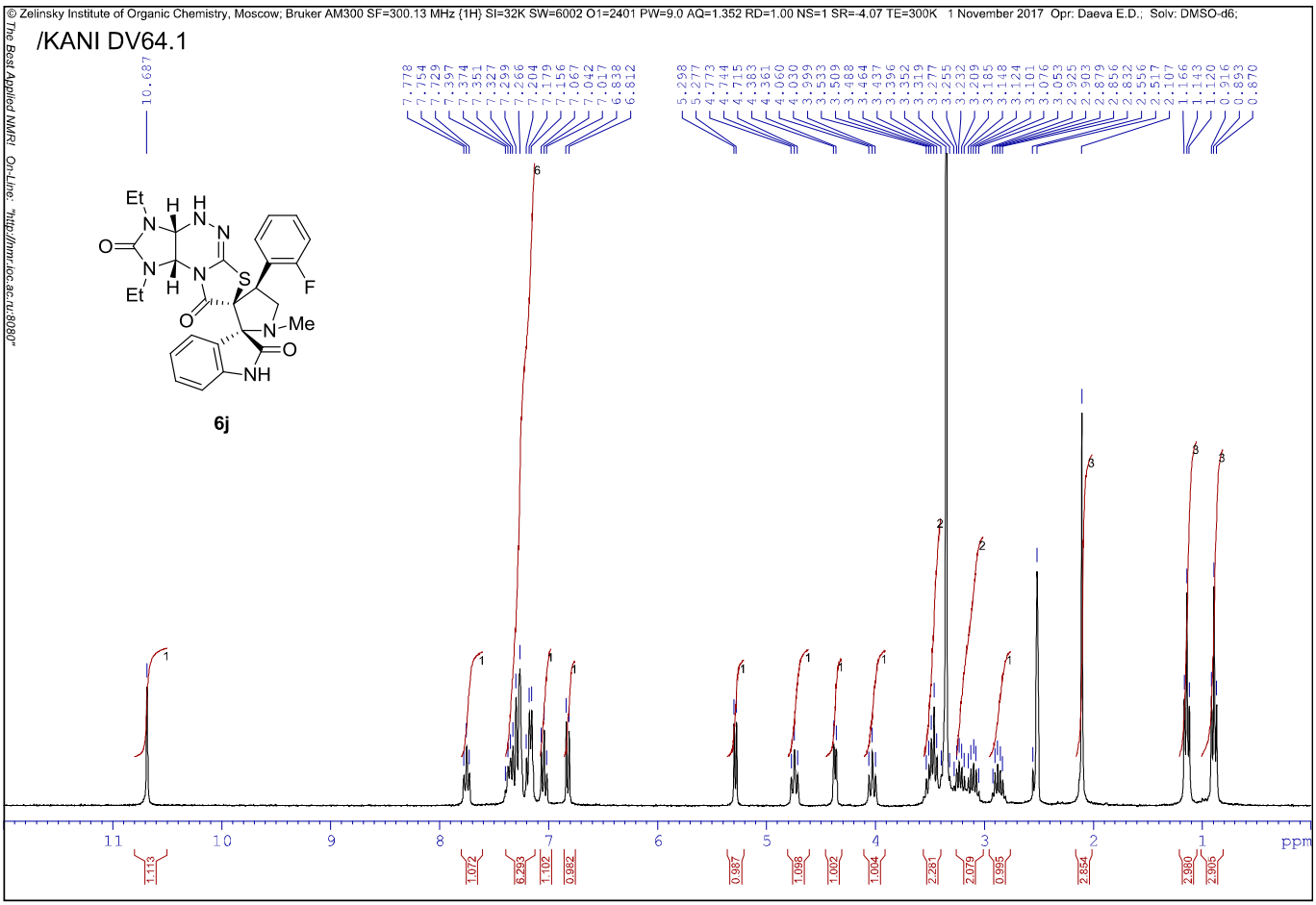
^{1}H NMR spectrum of **6i**



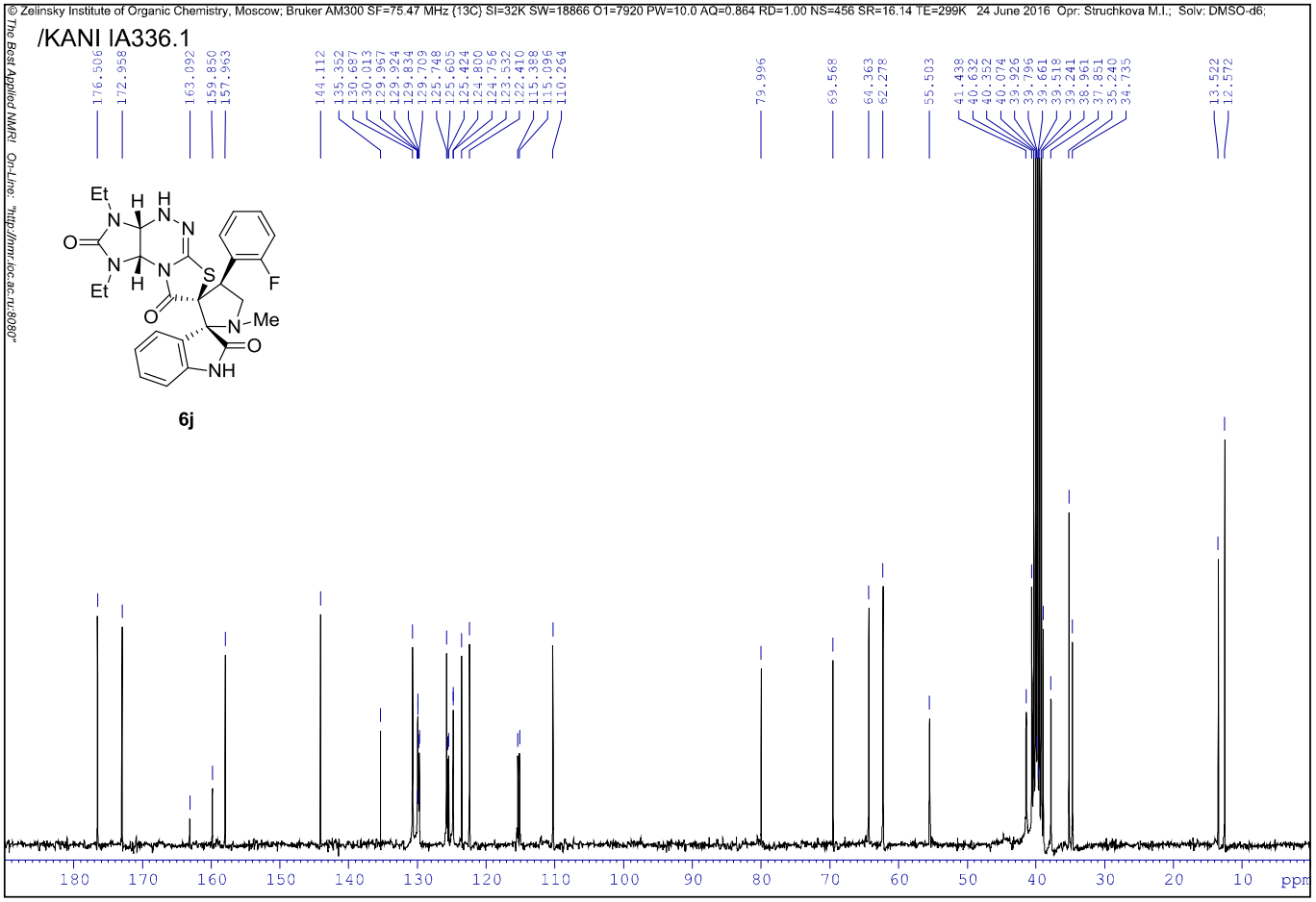
^{13}C NMR spectrum of **6i**



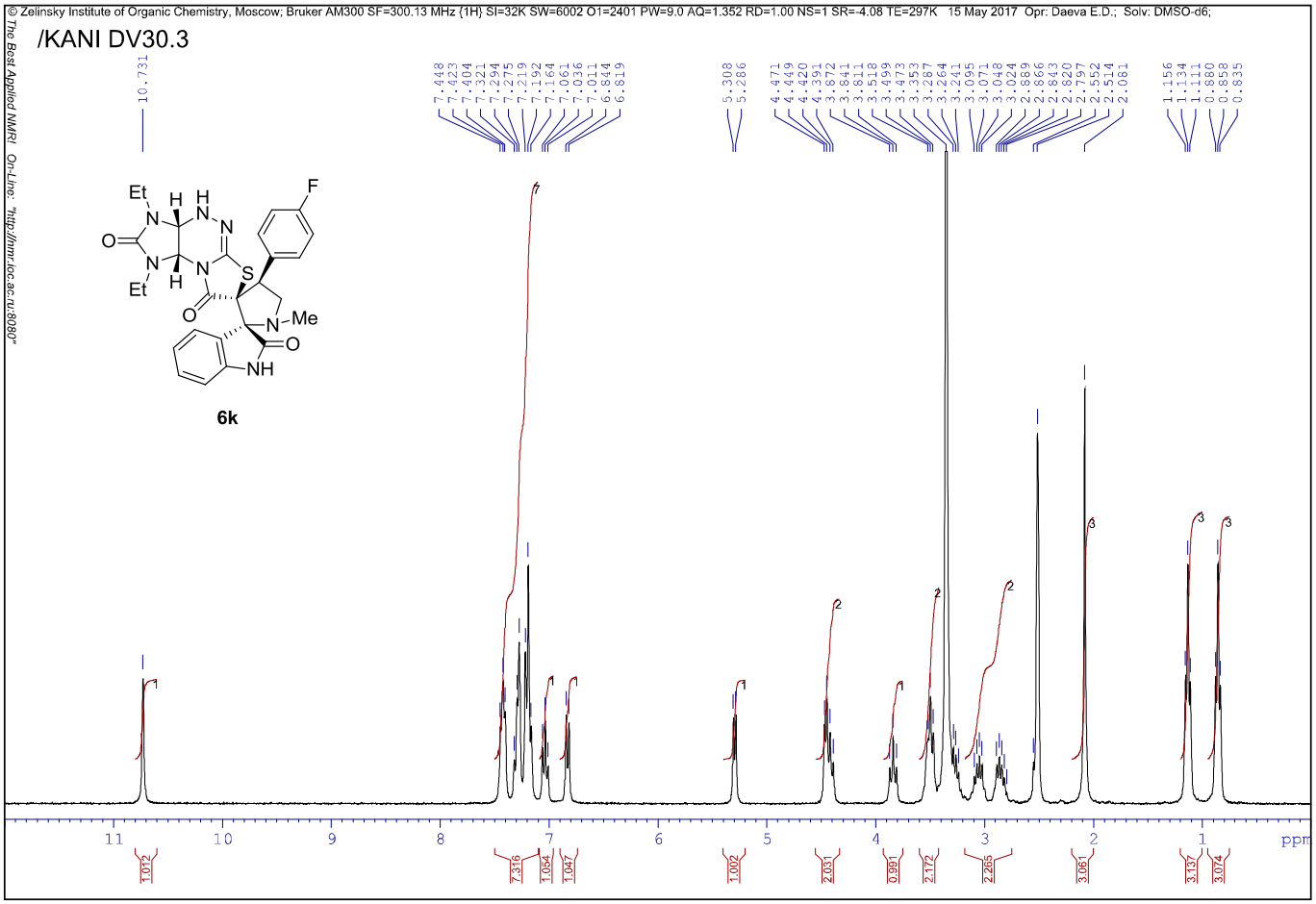
^{1}H NMR spectrum of **6j**



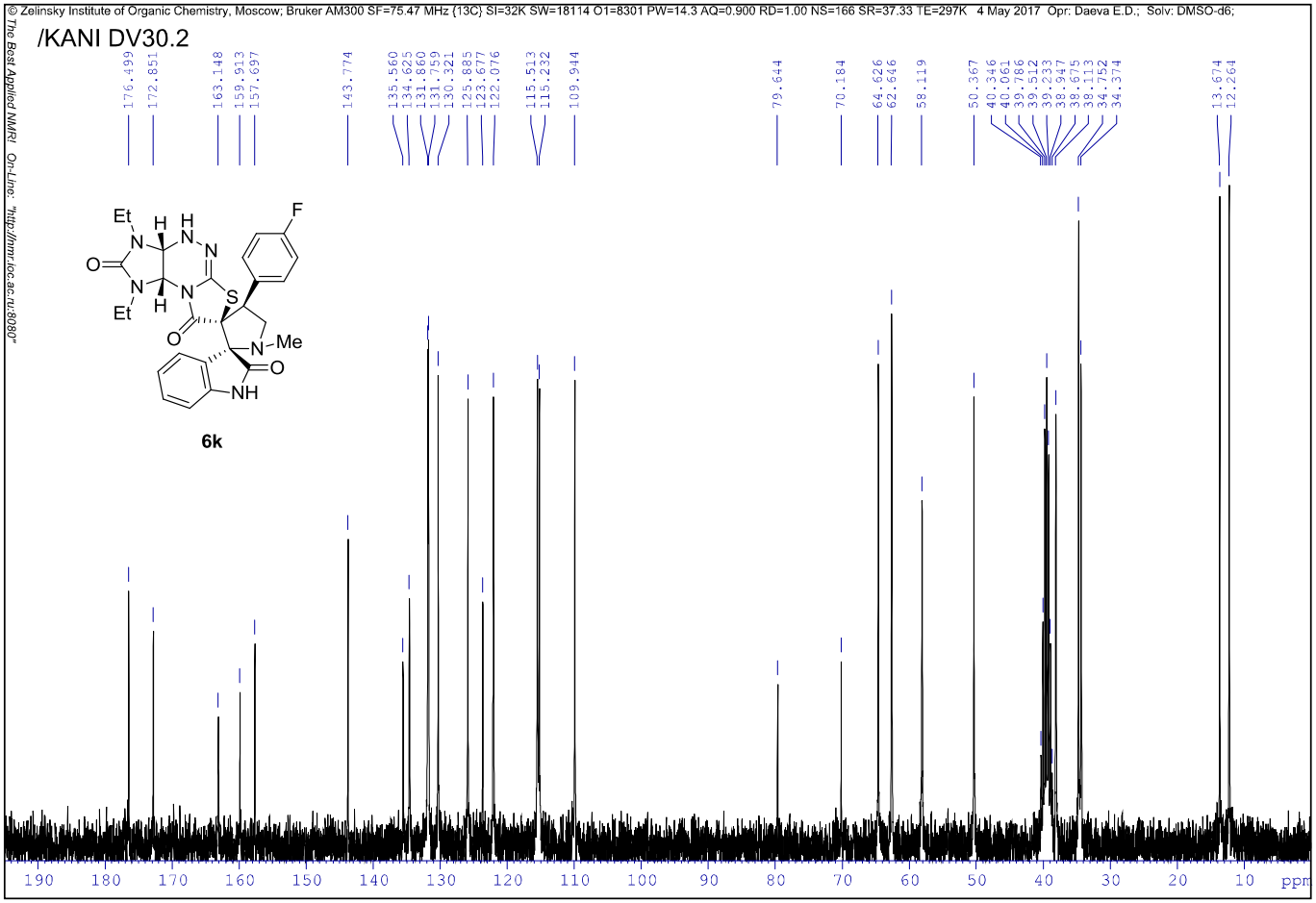
^{13}C NMR spectrum of **6j**



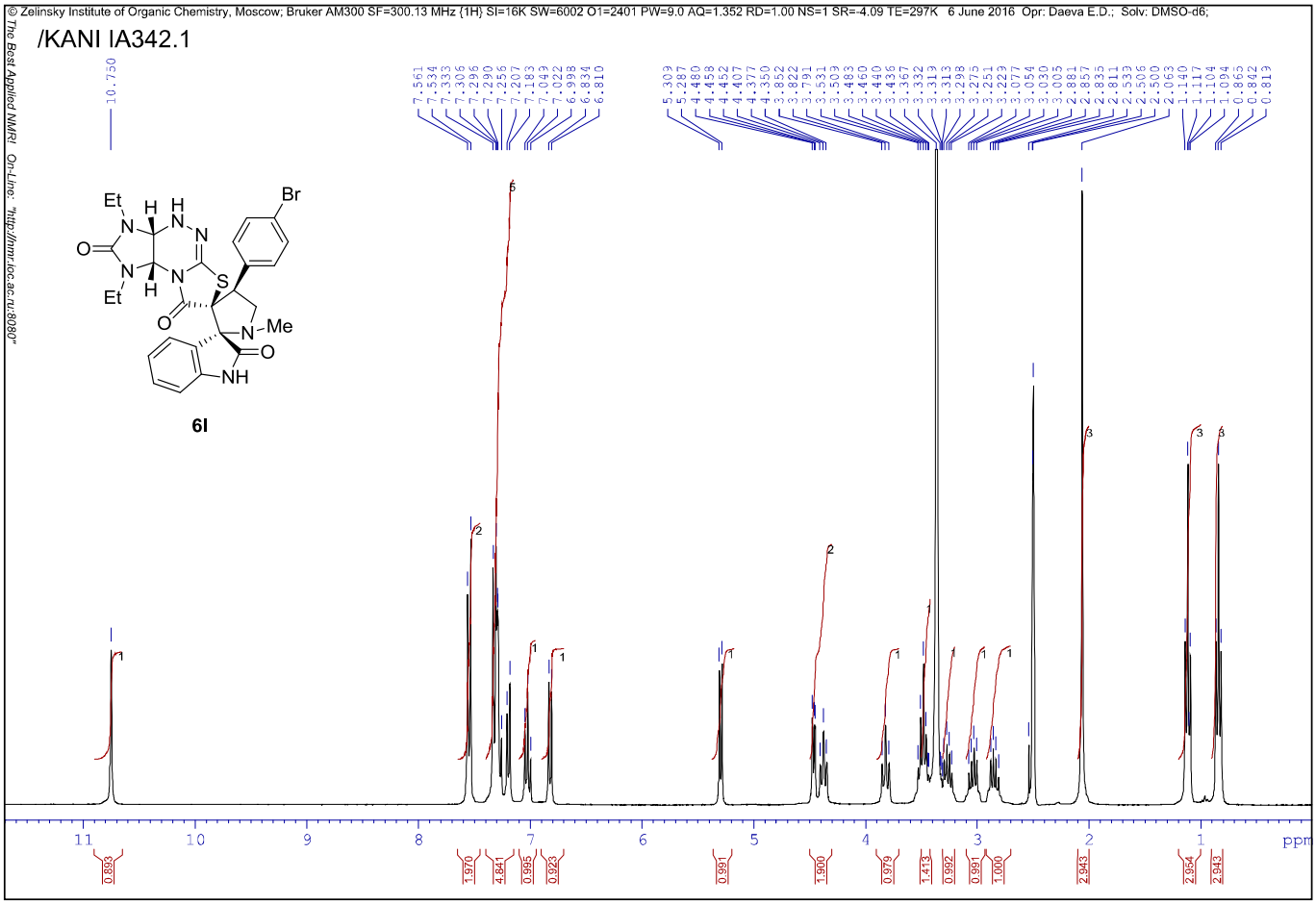
^{1}H NMR spectrum of **6k**



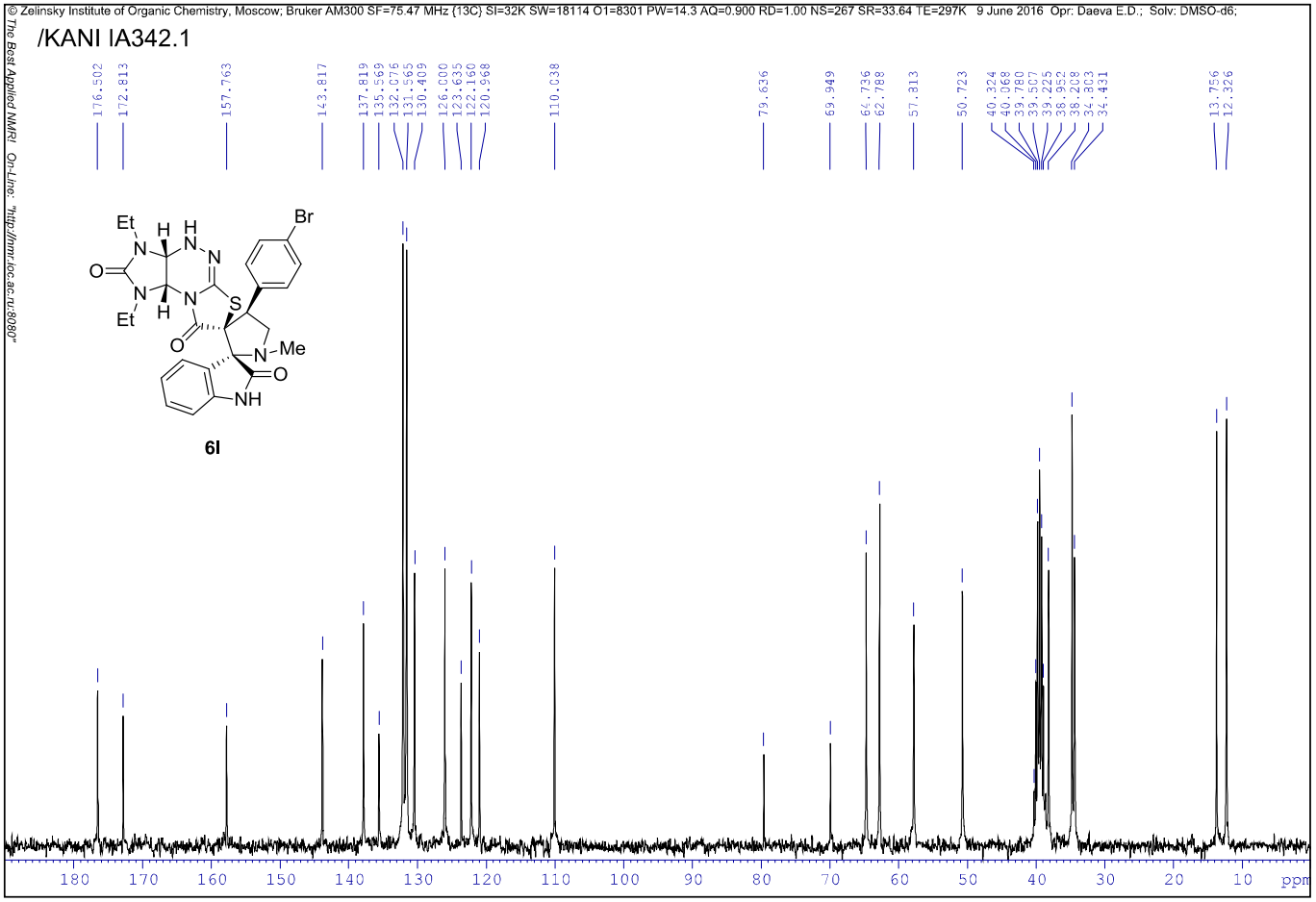
^{13}C NMR spectrum of **6k**



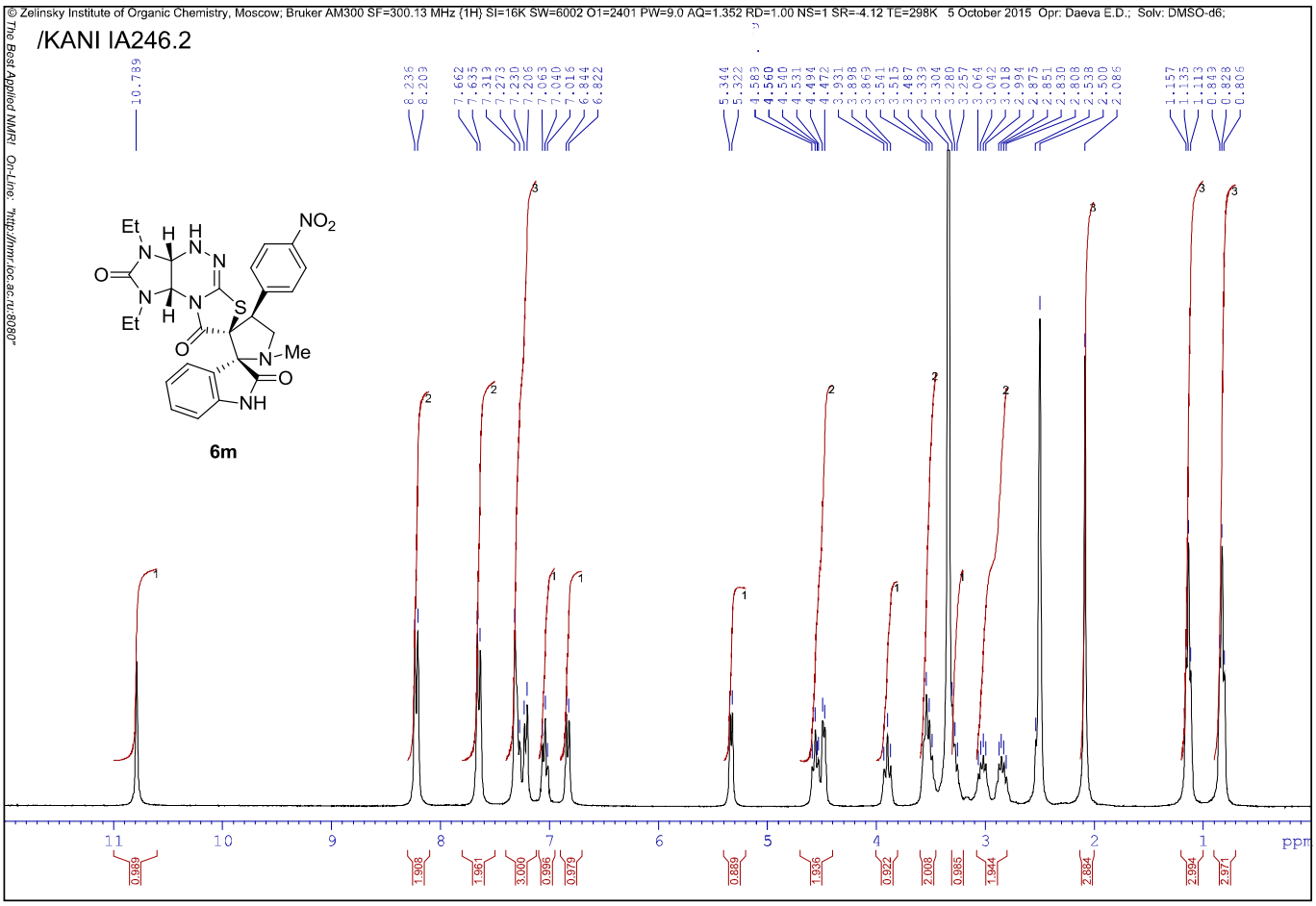
^{1}H NMR spectrum of **6l**



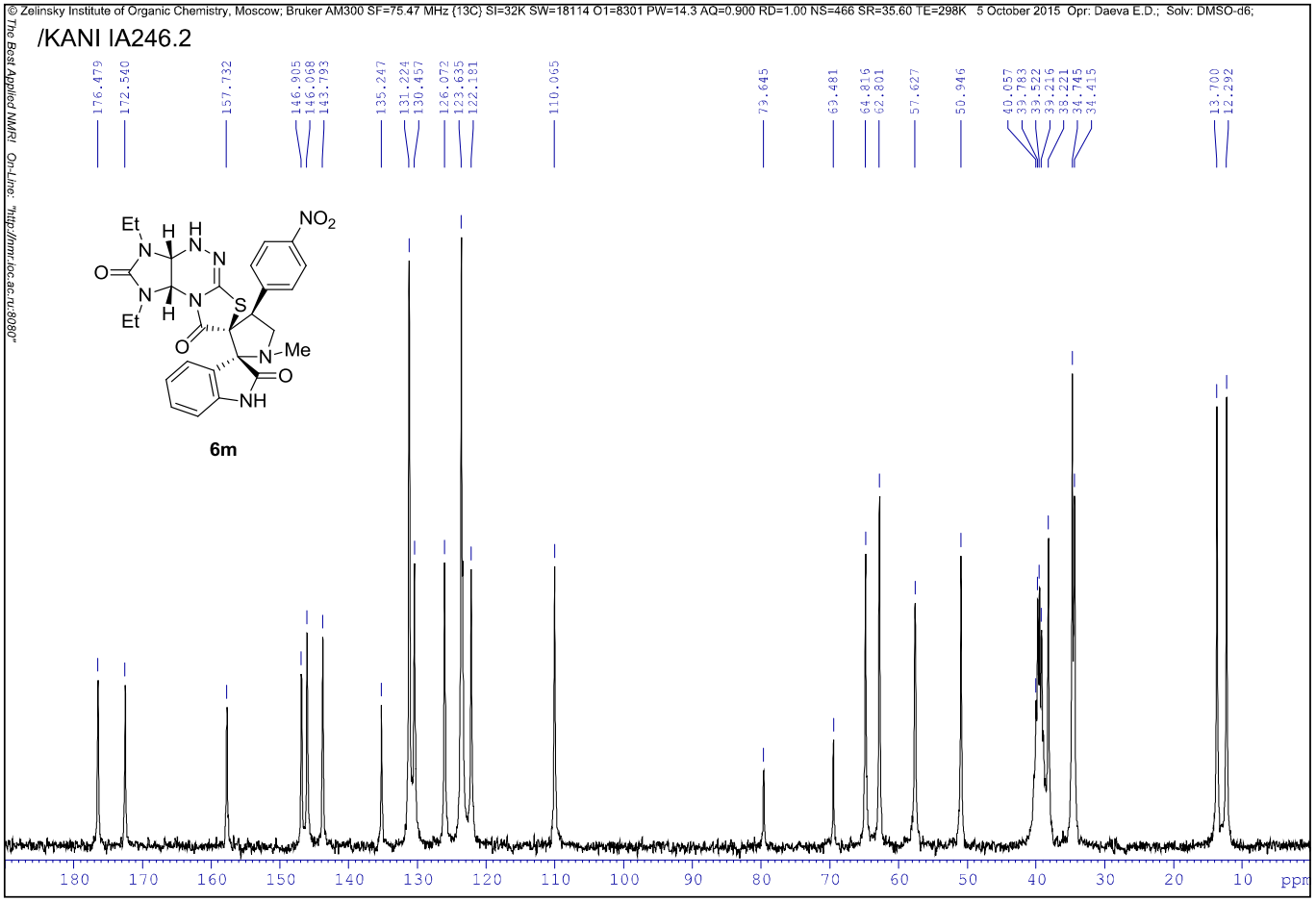
^{13}C NMR spectrum of **6l**



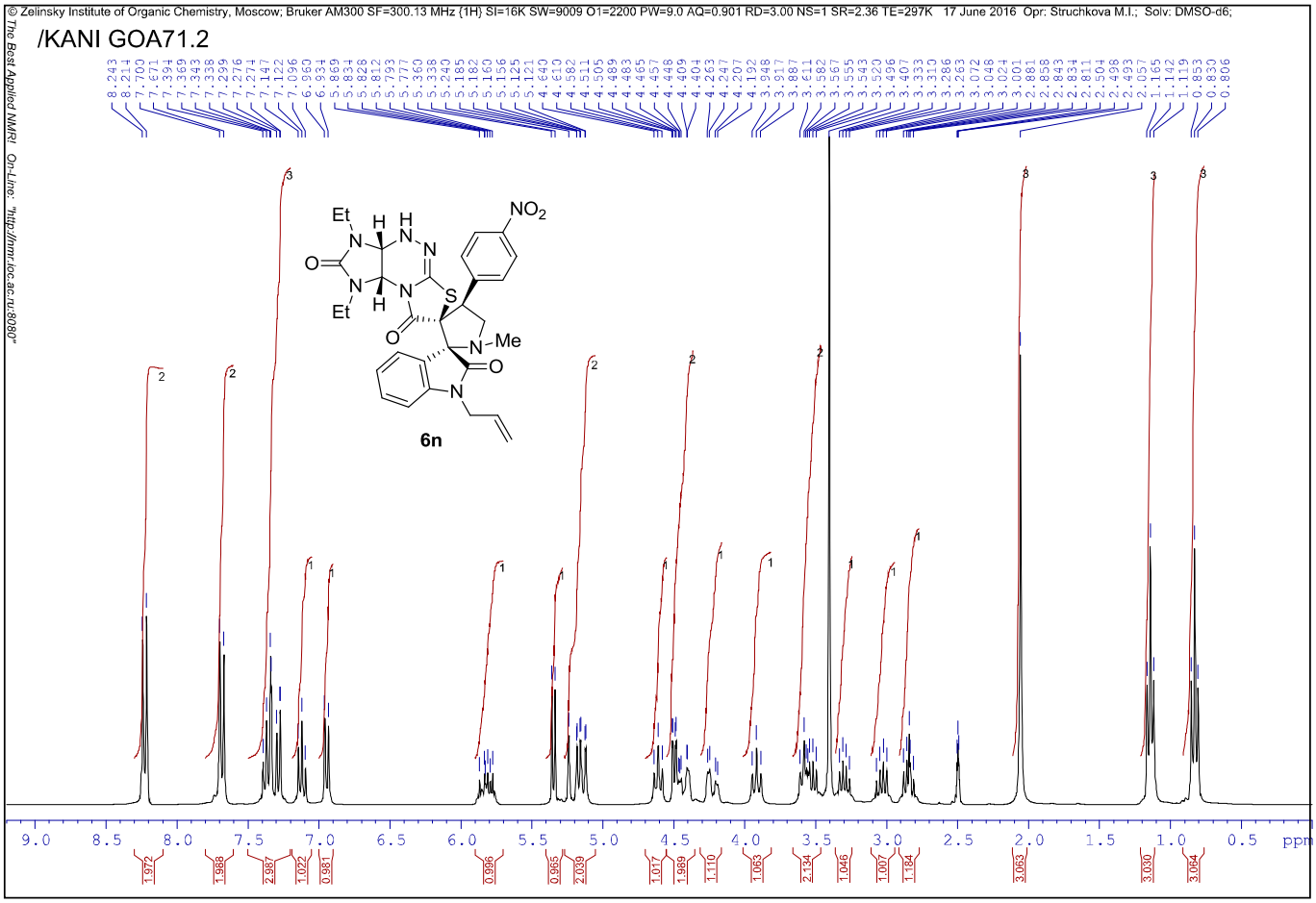
^{1}H NMR spectrum of **6m**



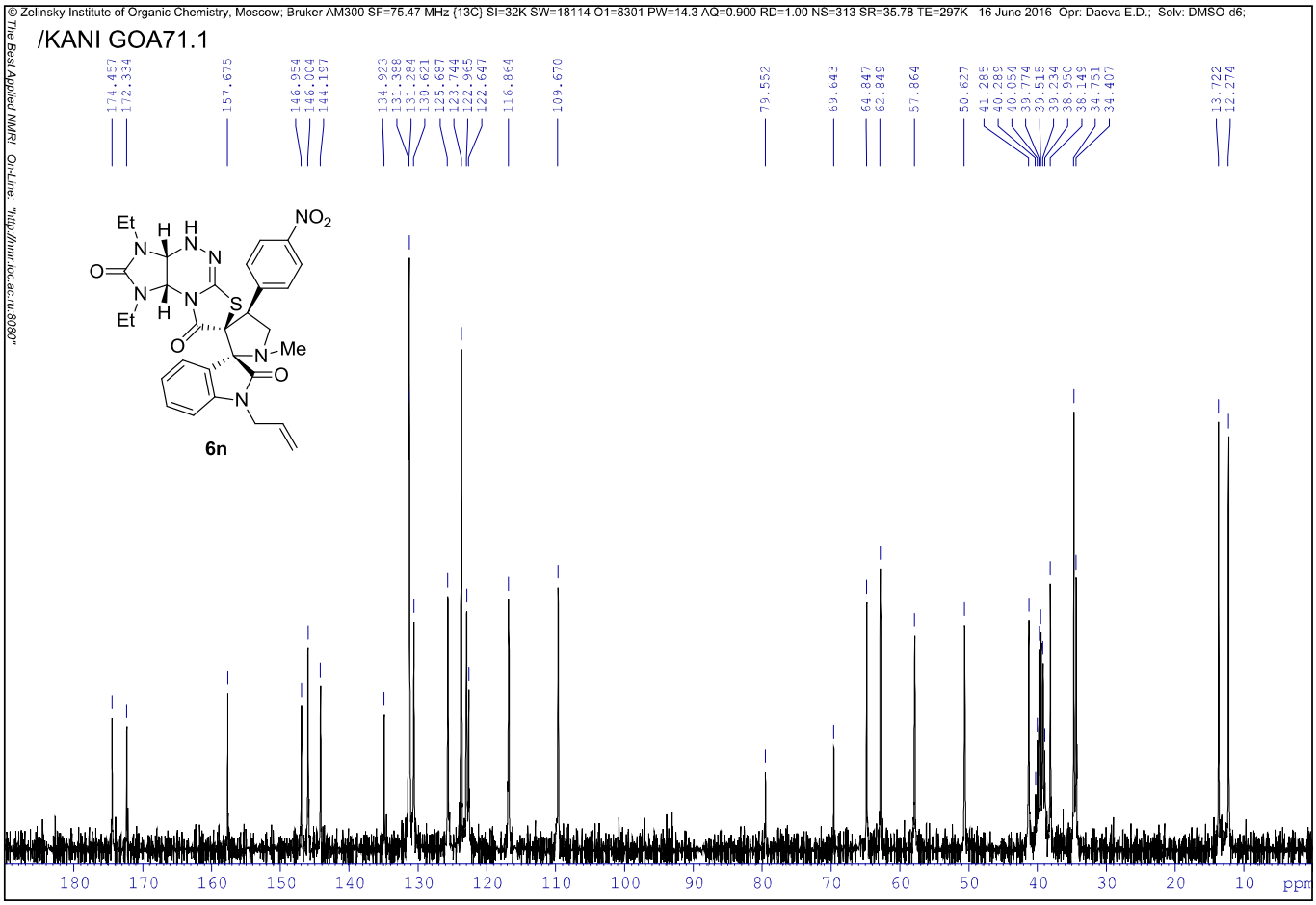
^{13}C NMR spectrum of **6m**



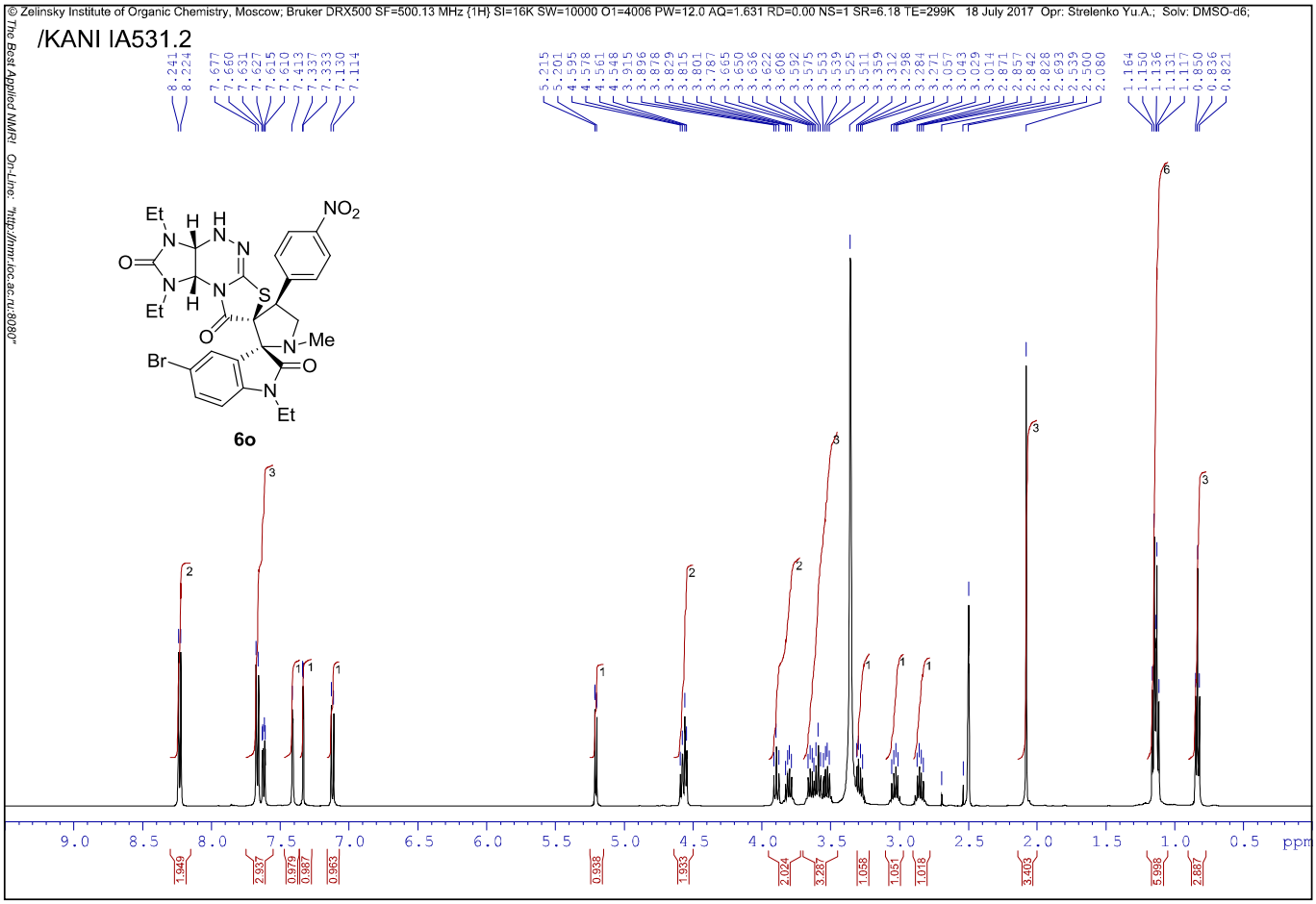
^{1}H NMR spectrum of **6n**



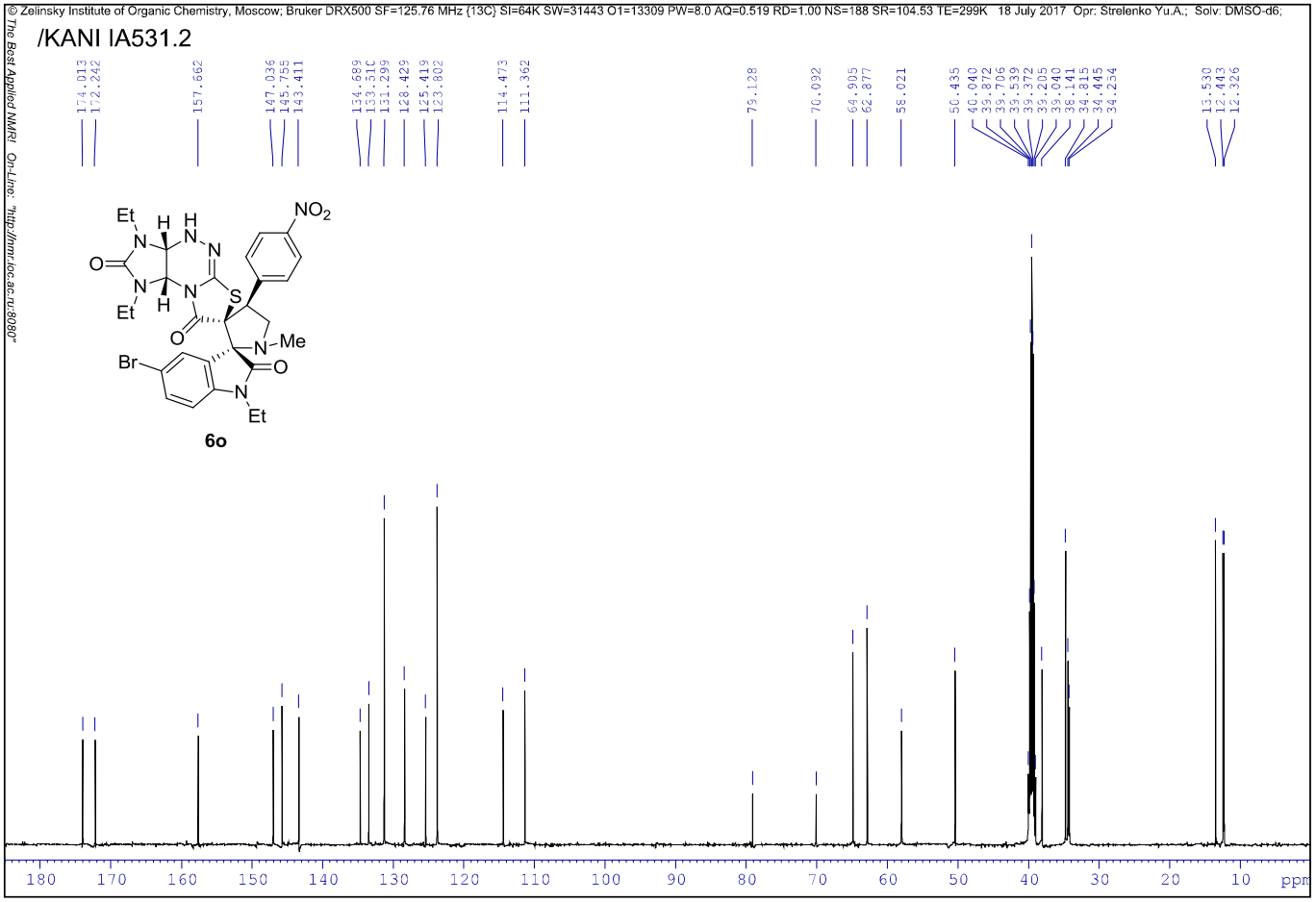
^{13}C NMR spectrum of **6n**



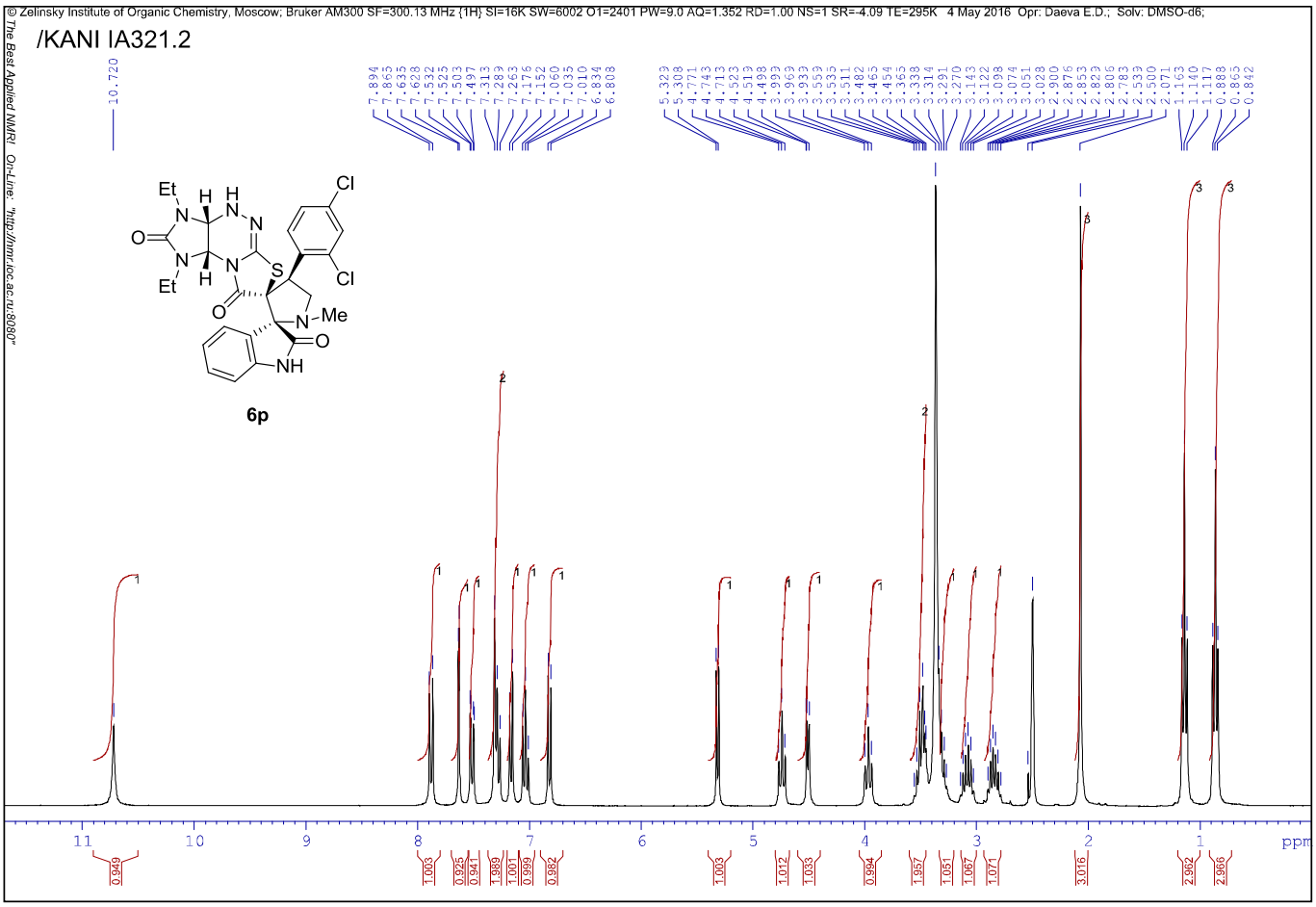
^{1}H NMR spectrum of **6o**



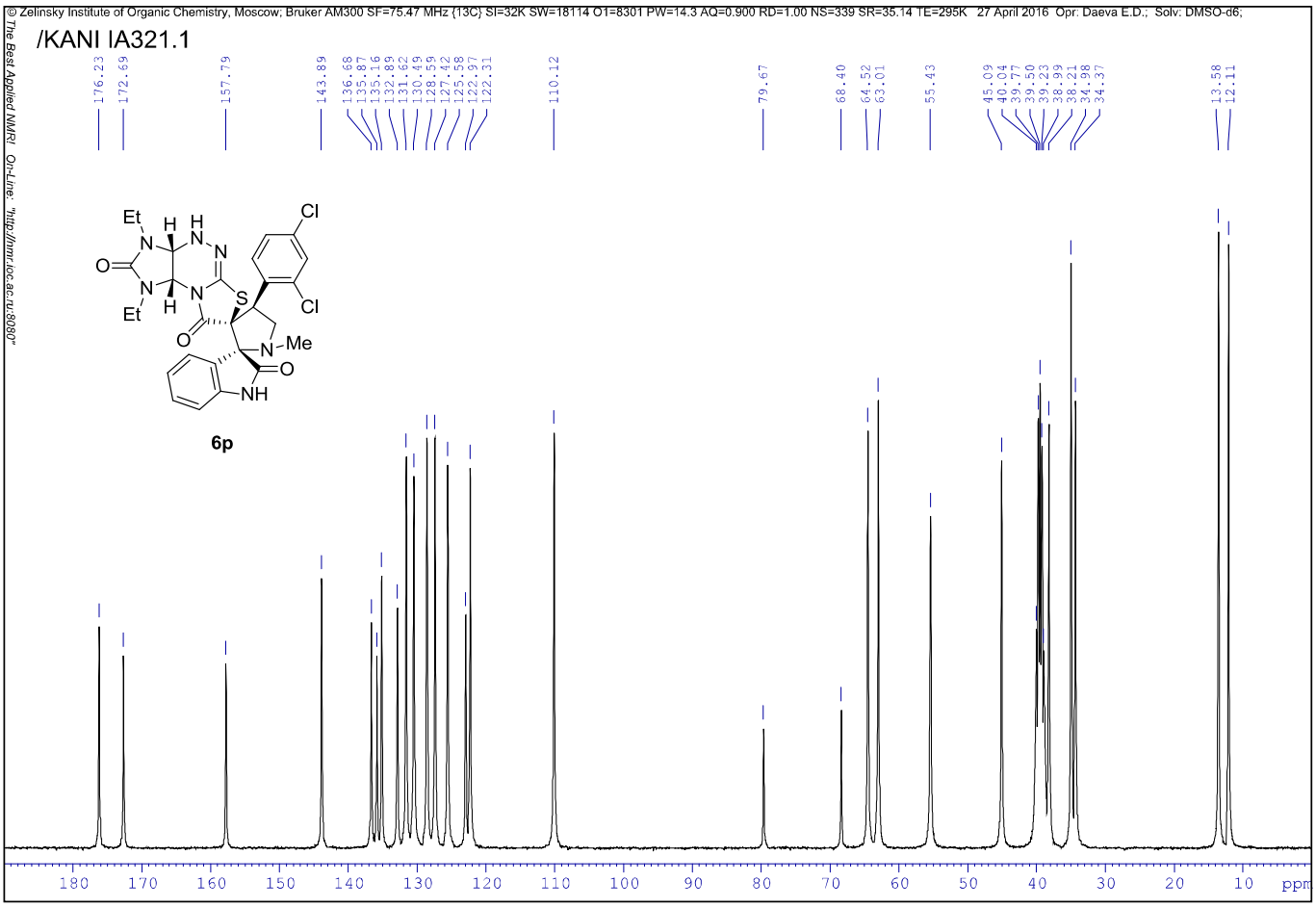
^{13}C NMR spectrum of **6o**



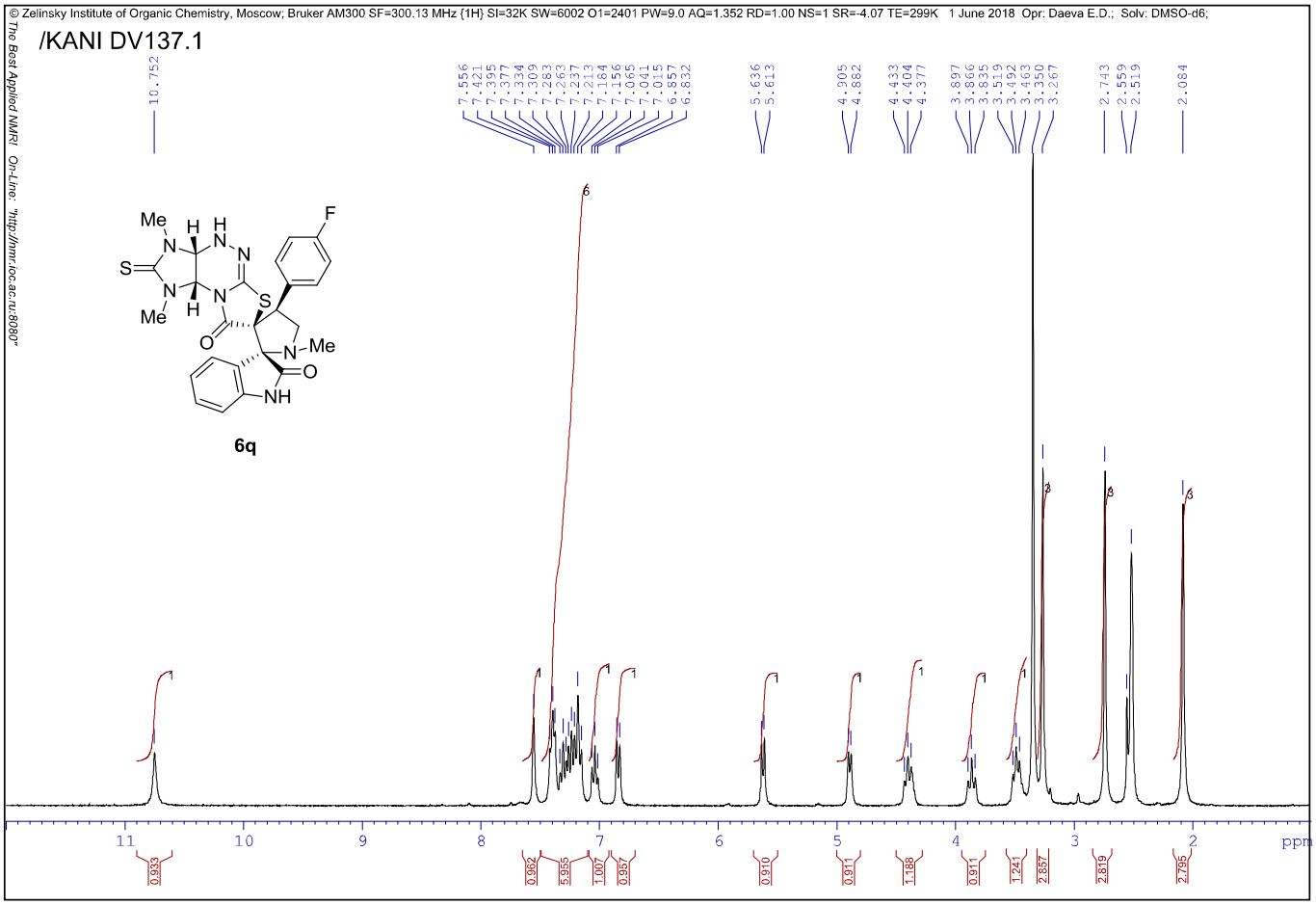
^{1}H NMR spectrum of **6p**



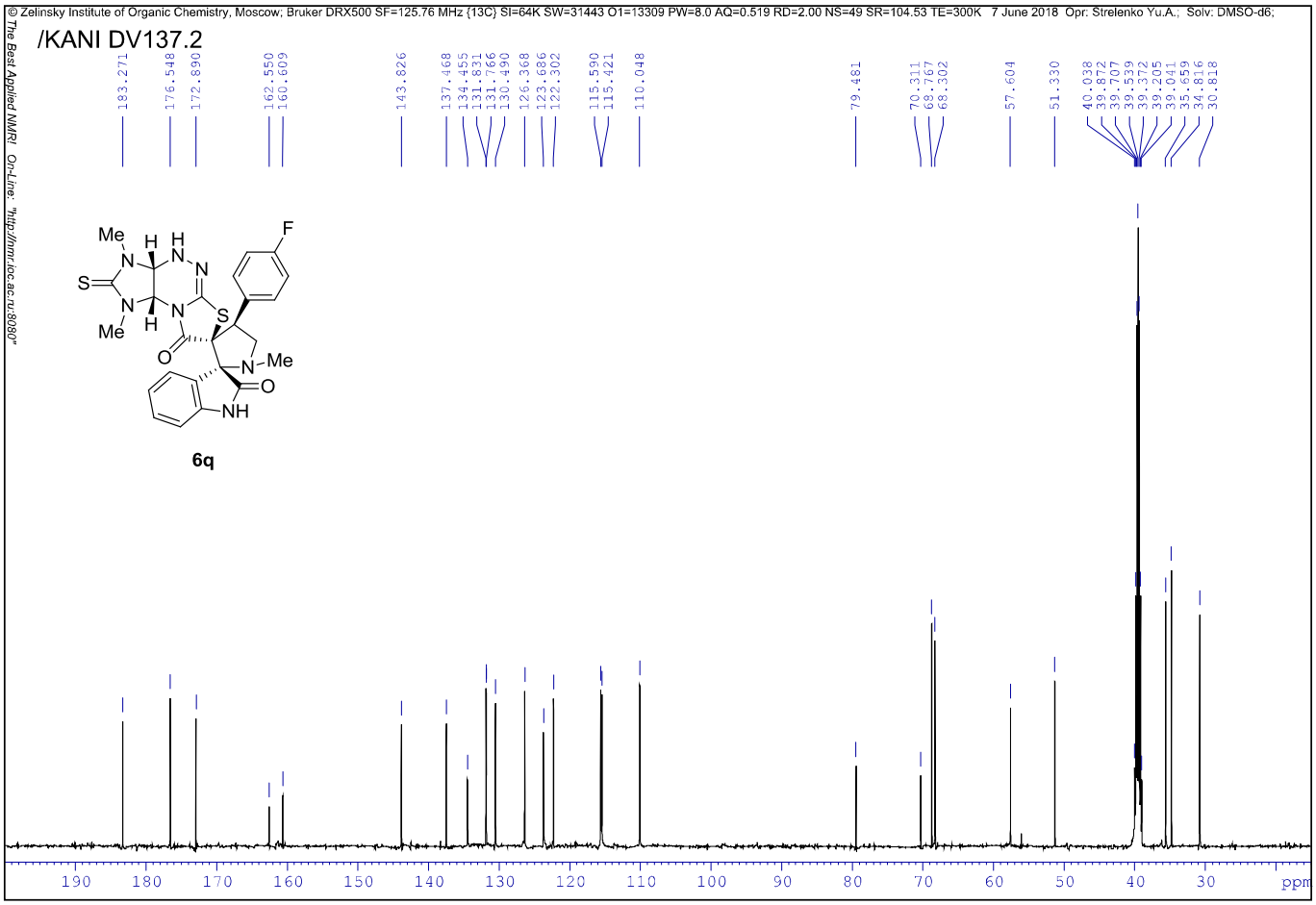
^{13}C NMR spectrum of **6p**



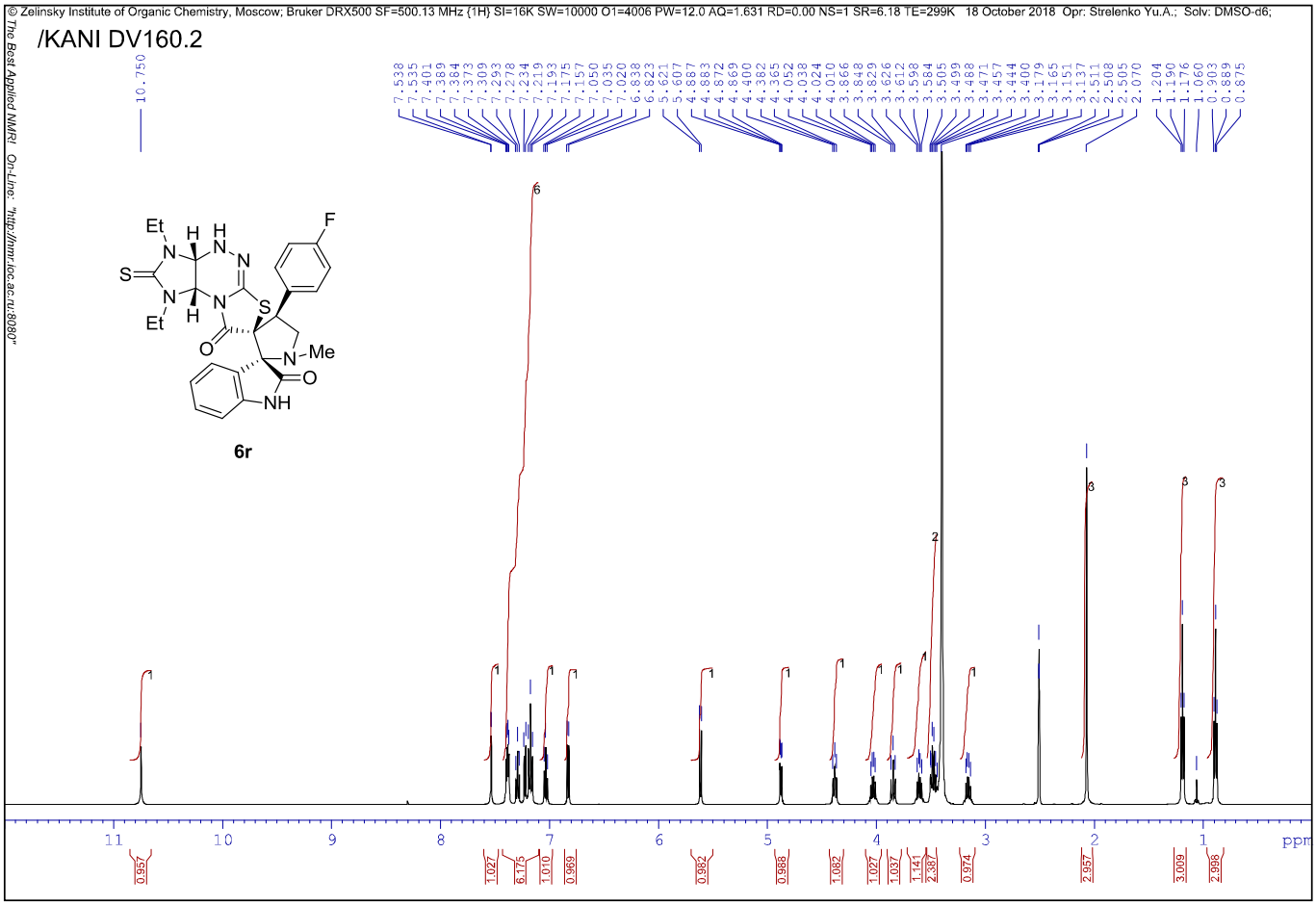
^{1}H NMR spectrum of **6q**



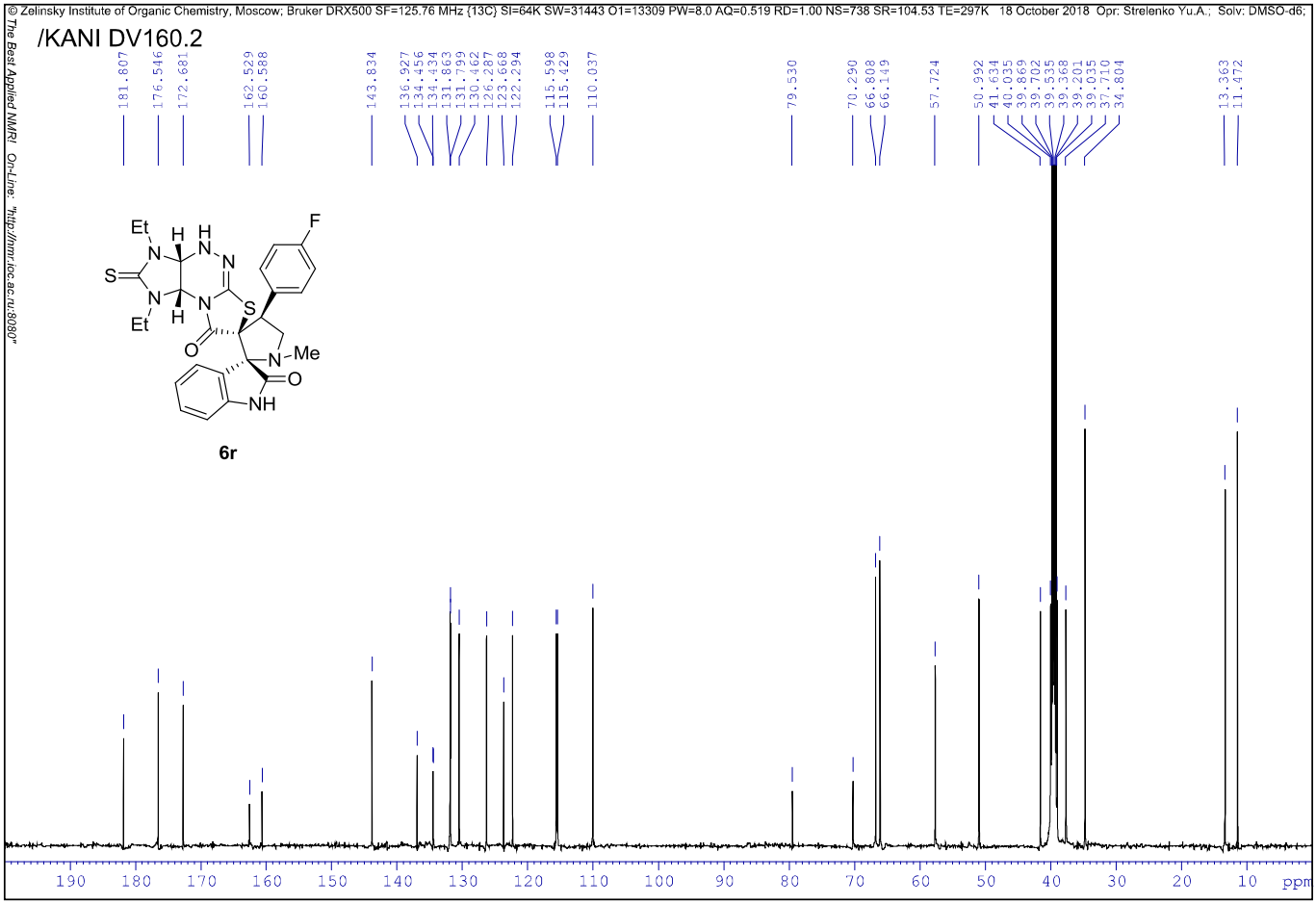
^{13}C NMR spectrum of **6q**



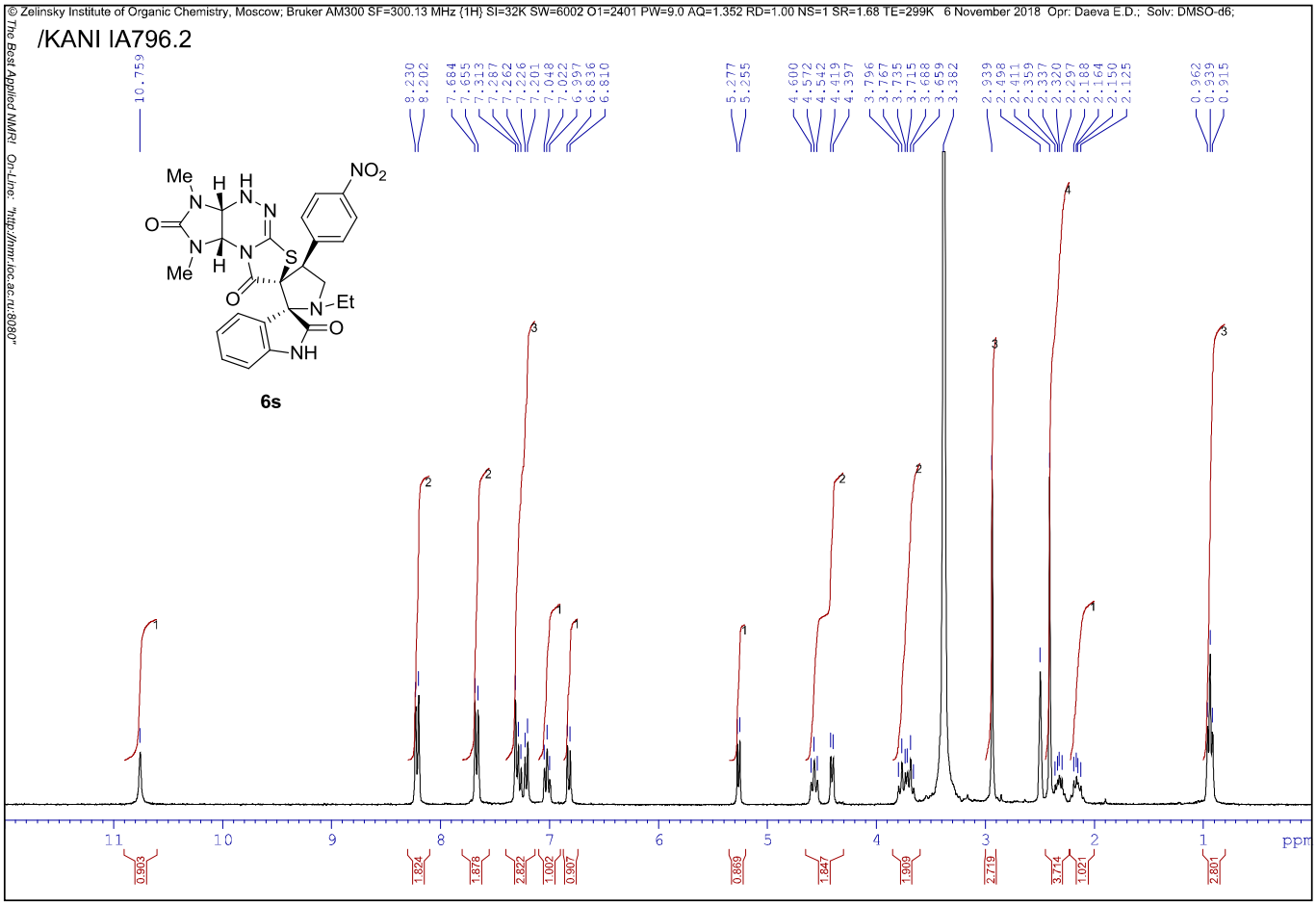
^{1}H NMR spectrum of **6r**



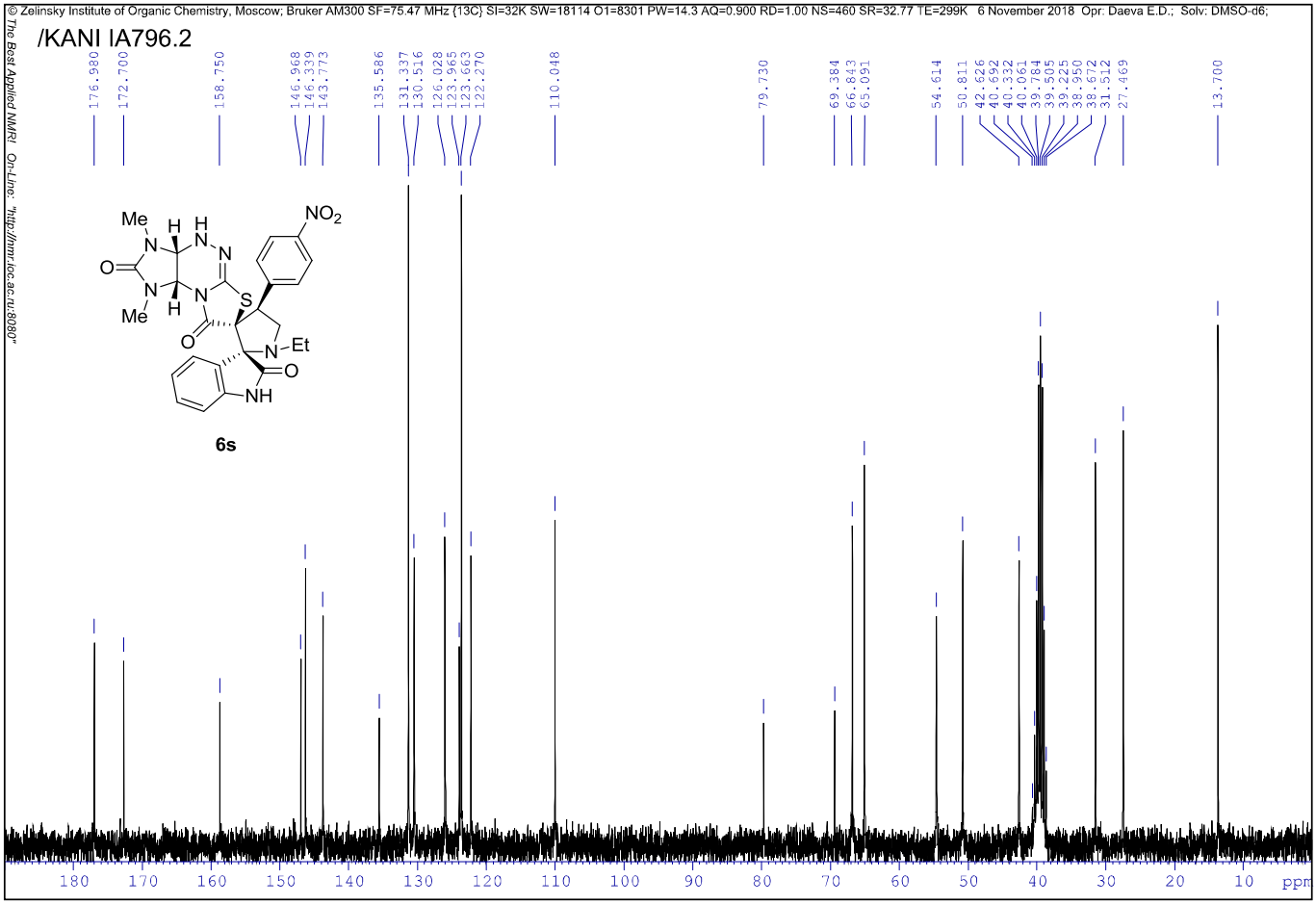
^{13}C NMR spectrum of **6r**



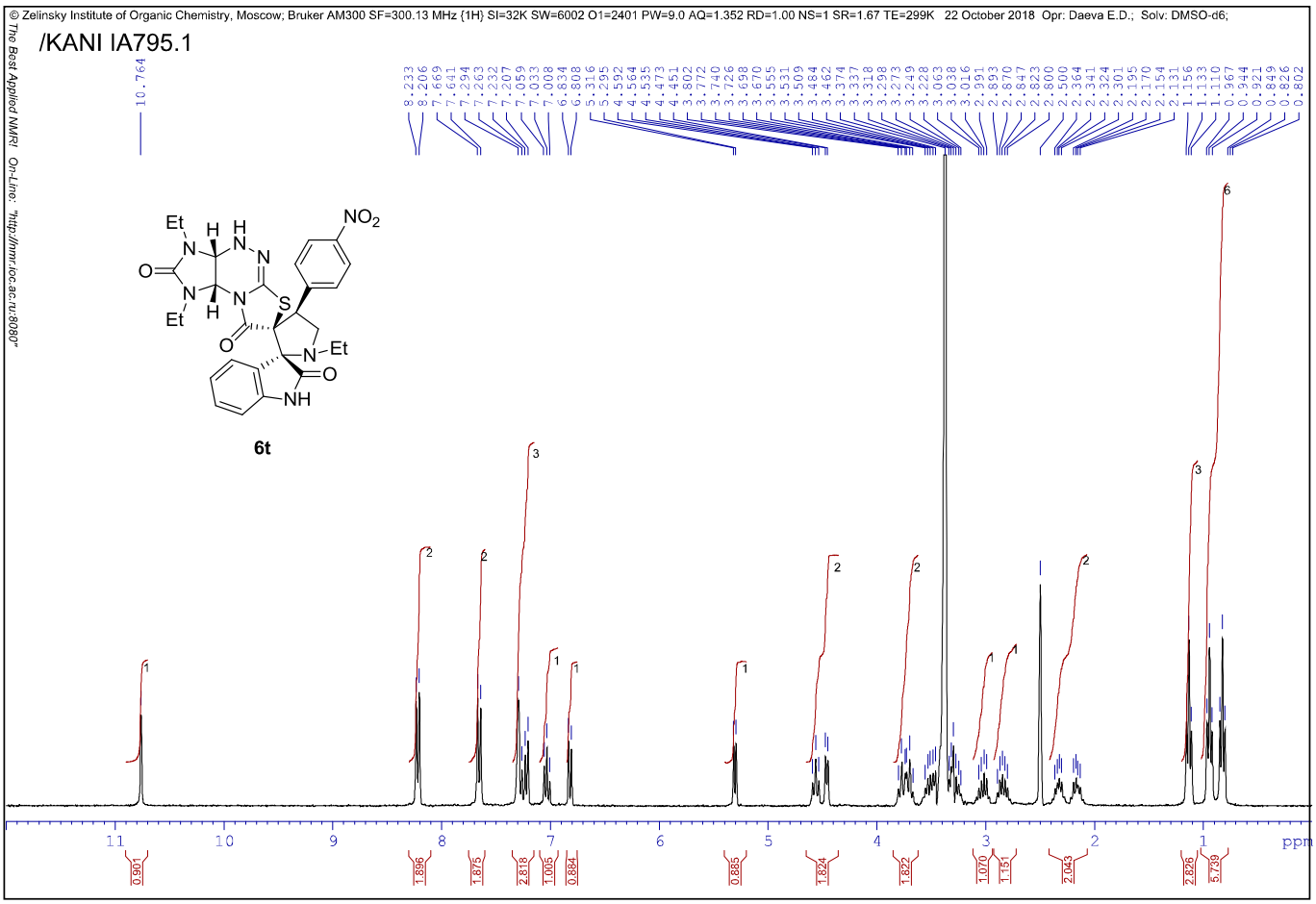
^{1}H NMR spectrum of **6s**



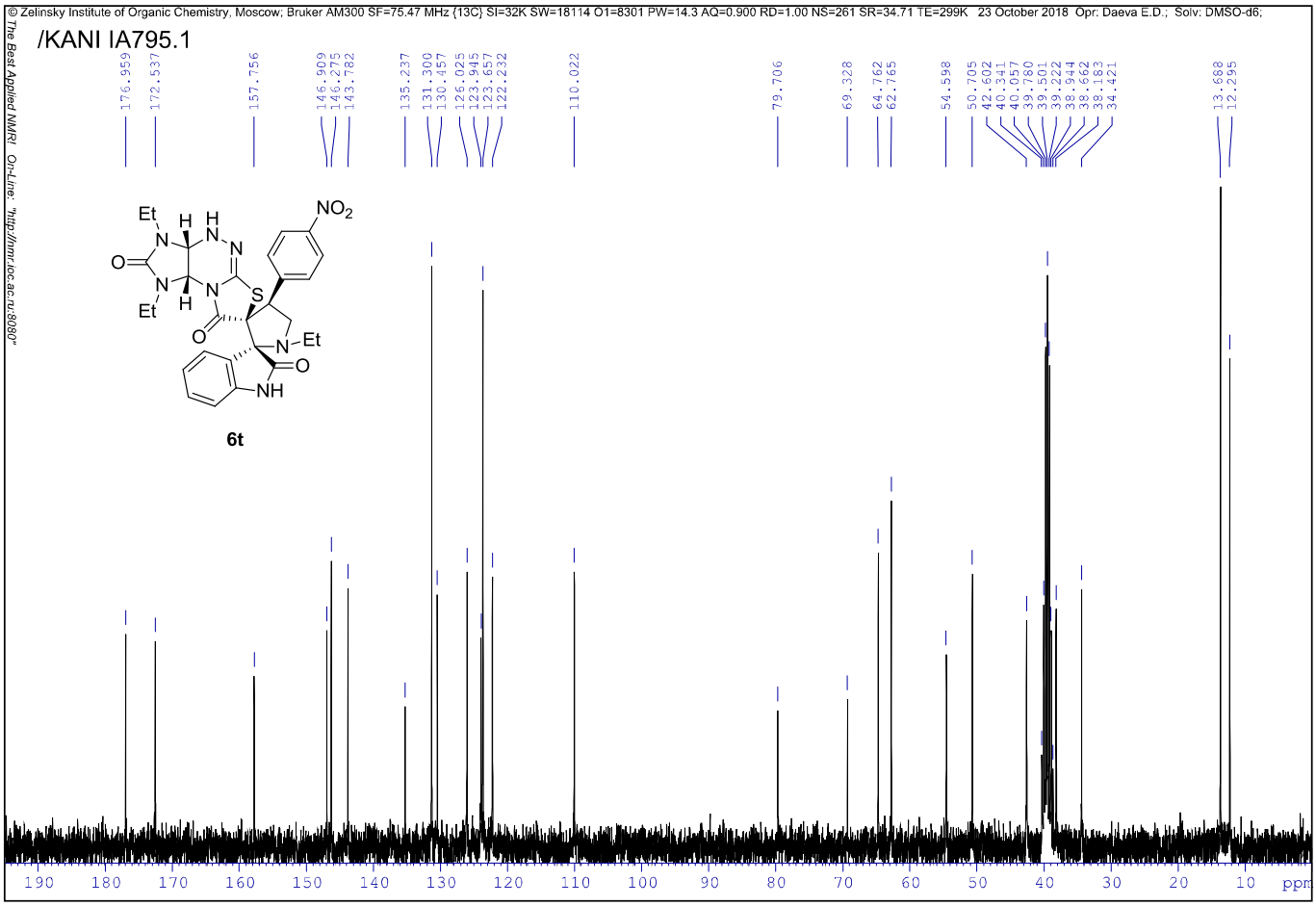
^{13}C NMR spectrum of **6s**



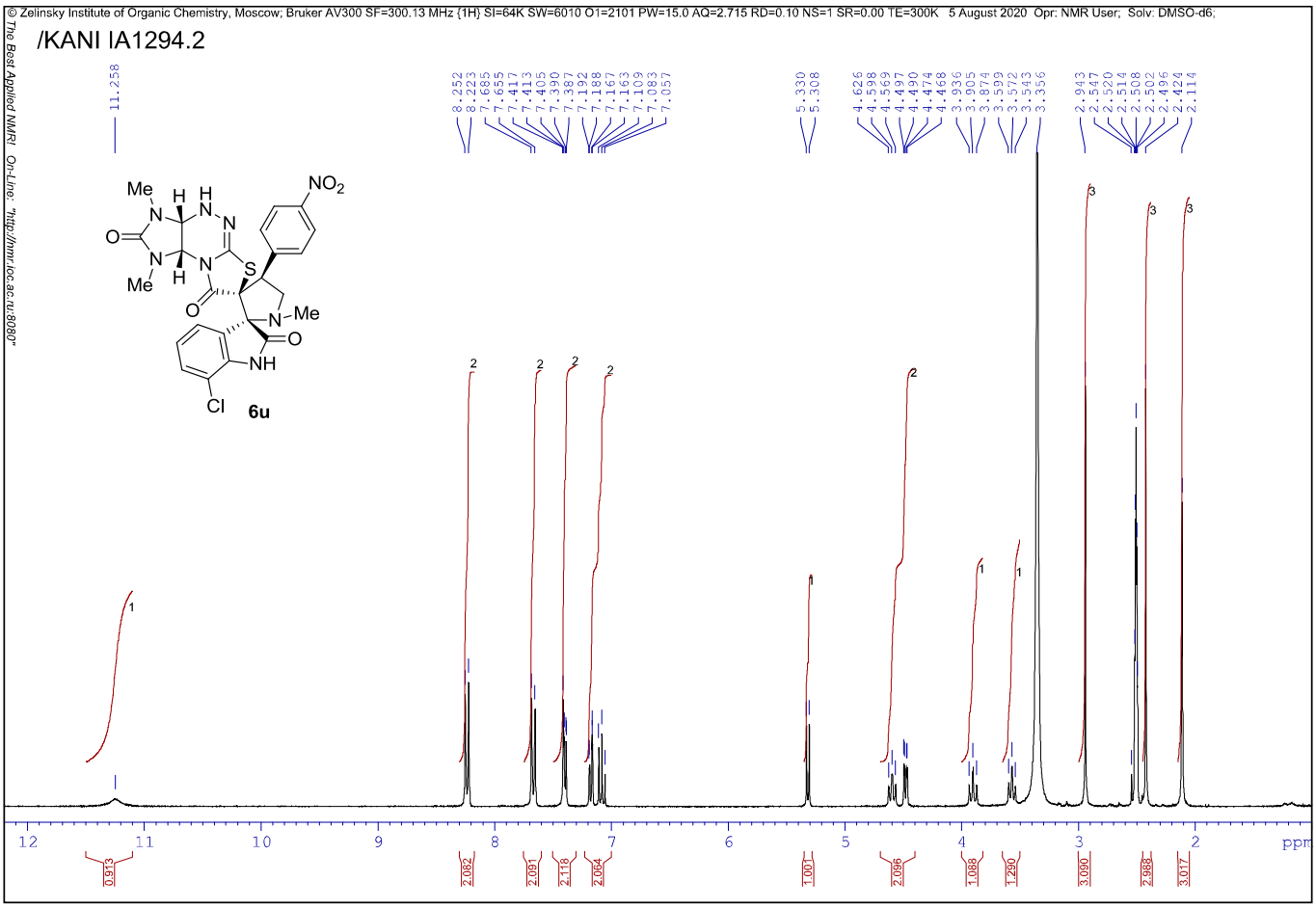
^{1}H NMR spectrum of **6t**



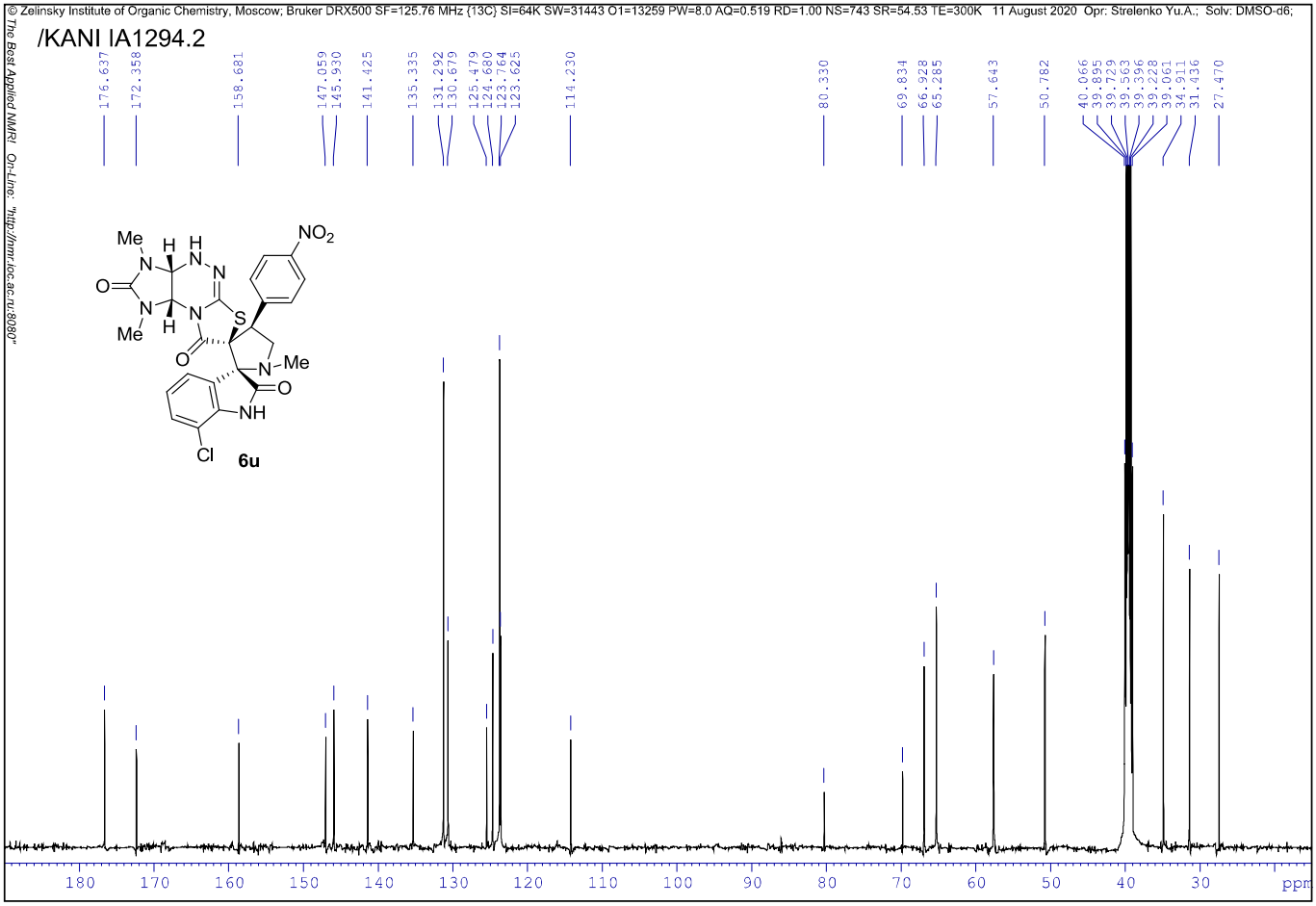
^{13}C NMR spectrum of **6t**



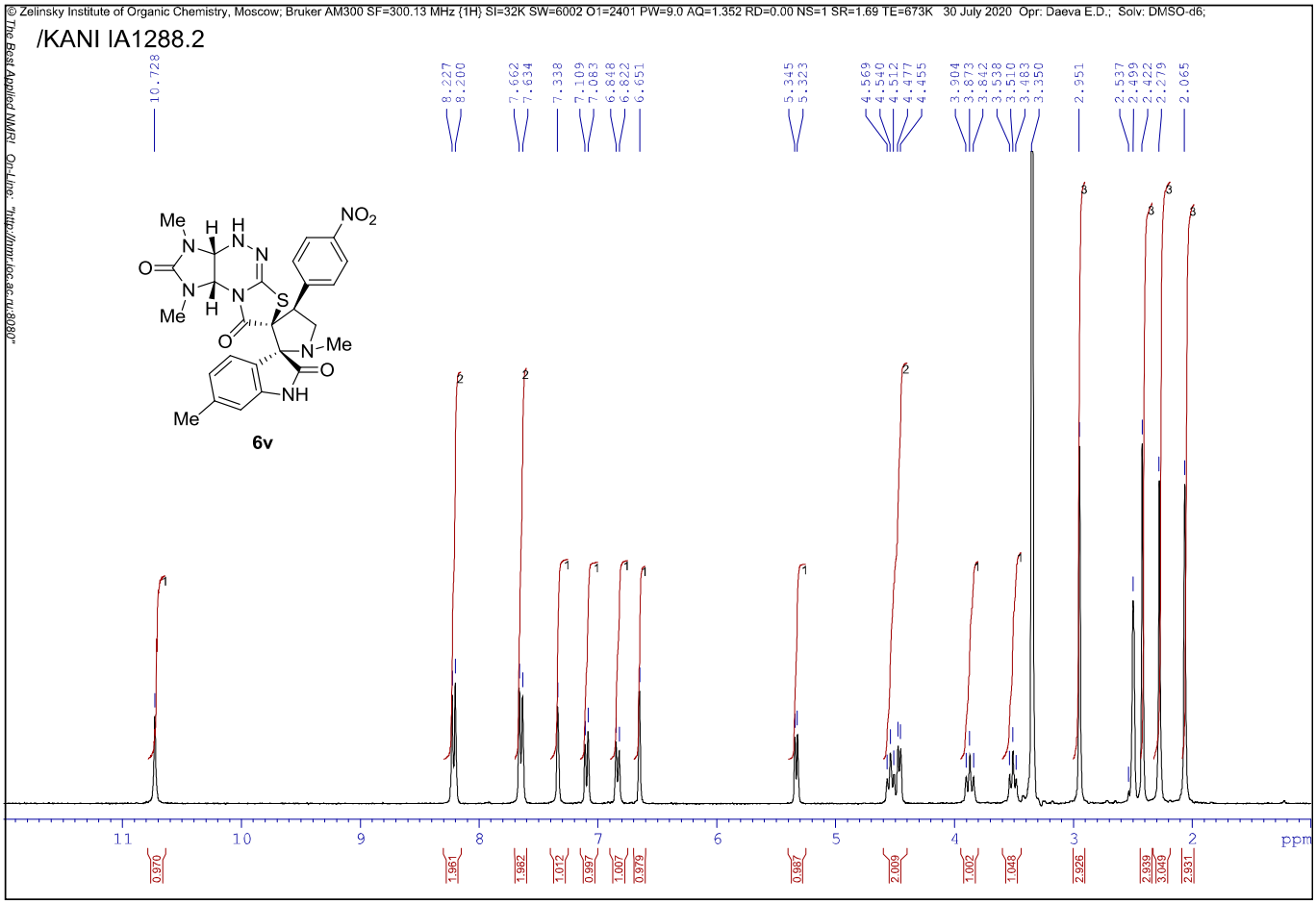
^{1}H NMR spectrum of **6u**



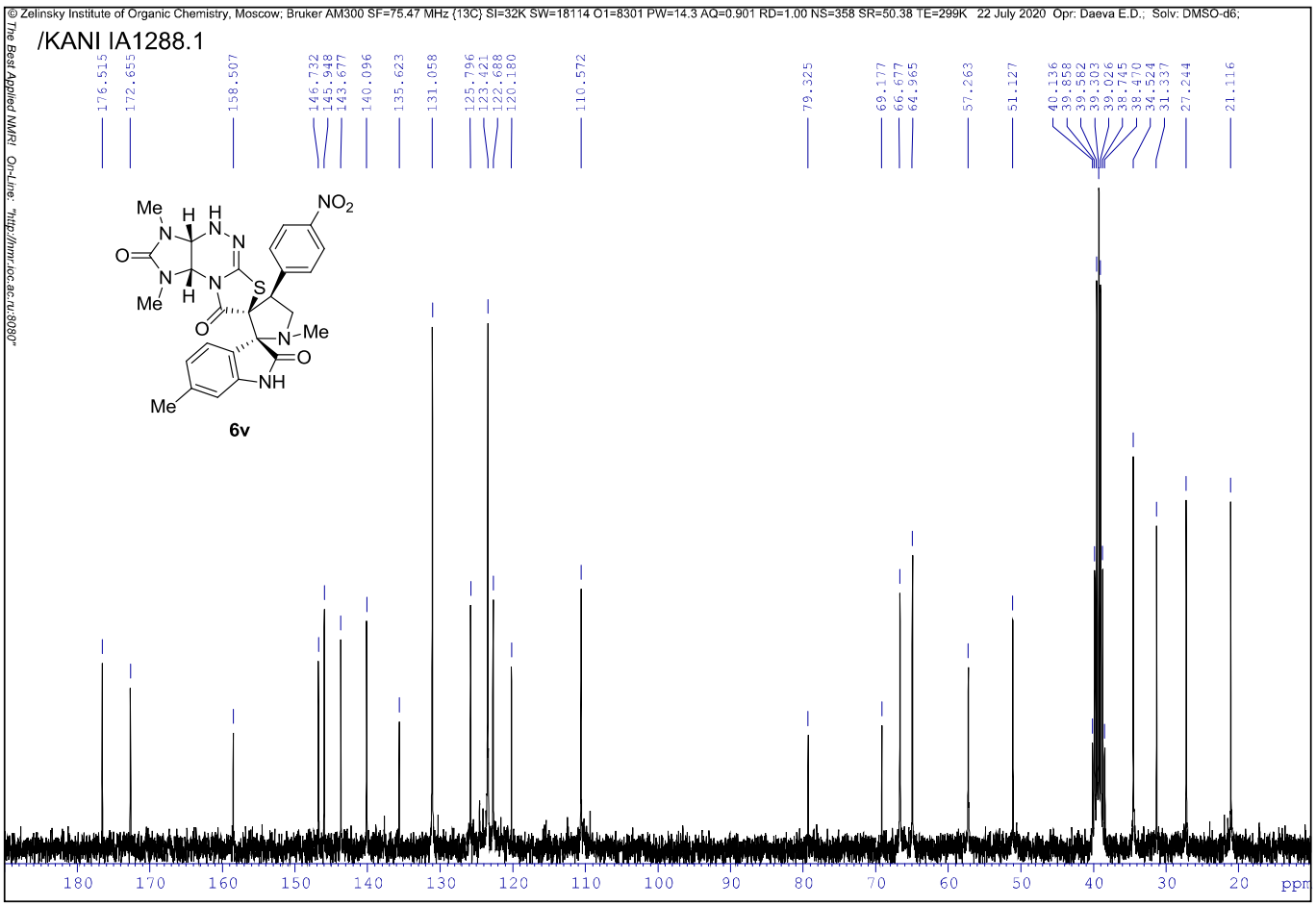
^{13}C NMR spectrum of **6u**



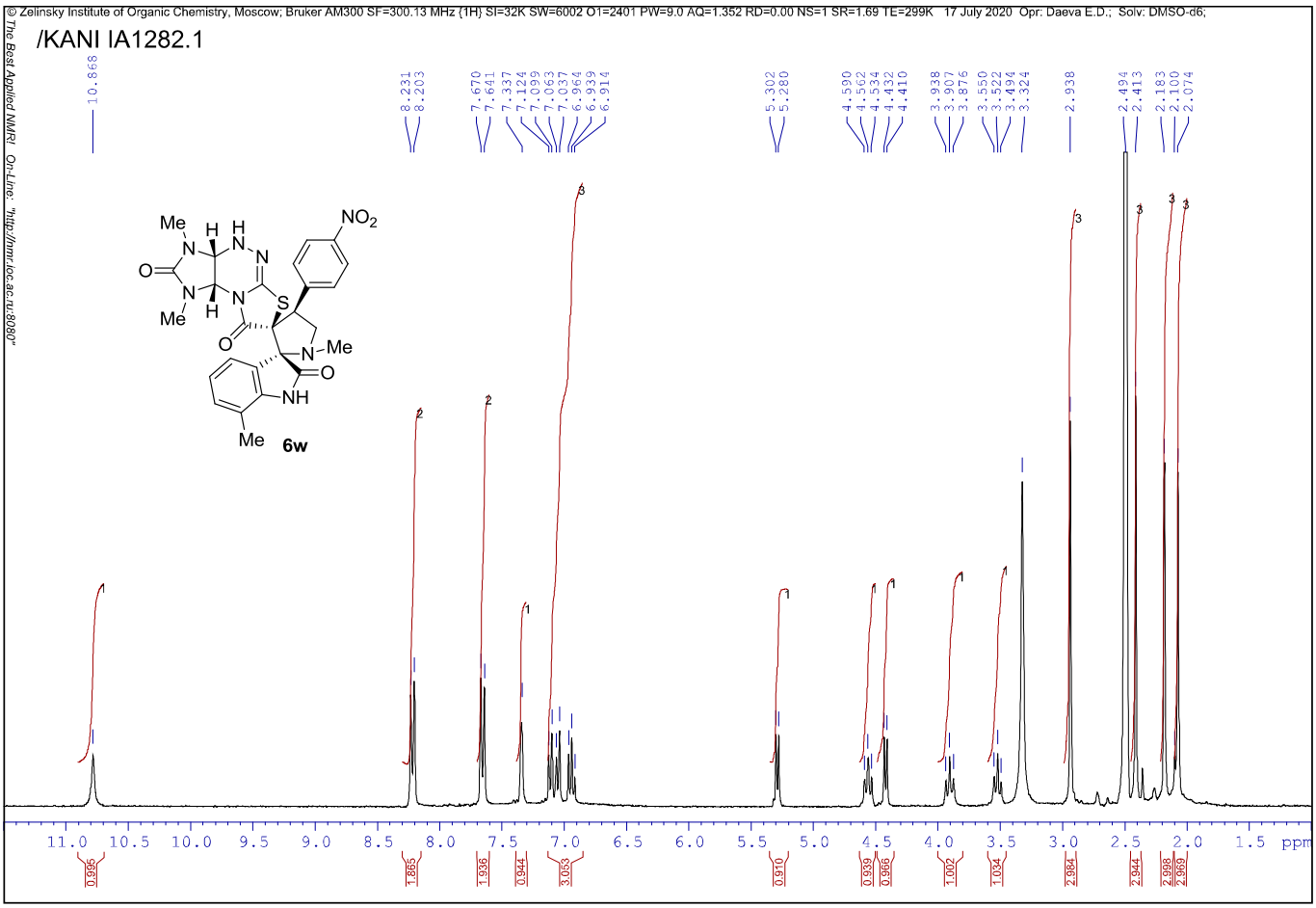
^{1}H NMR spectrum of **6v**



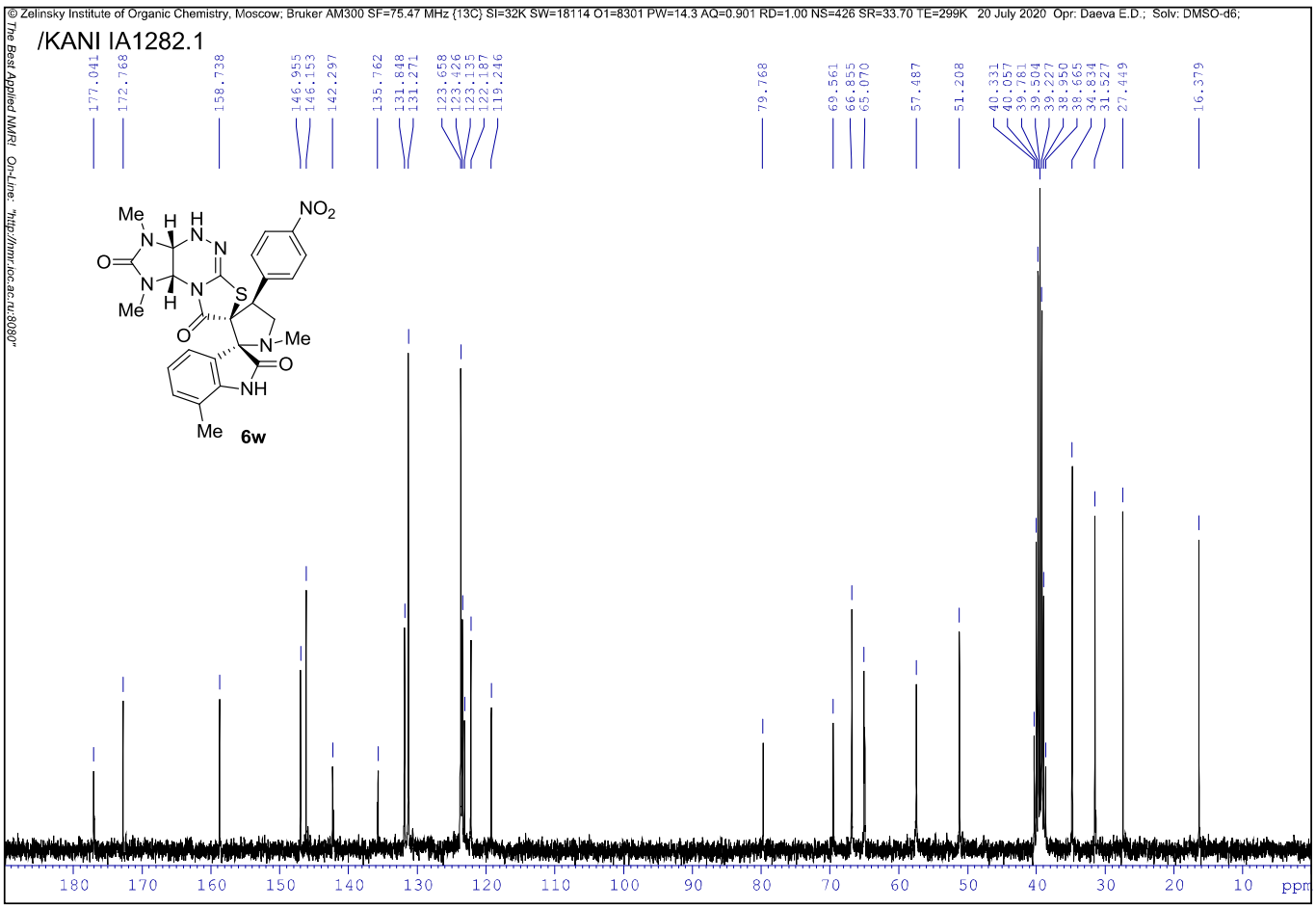
^{13}C NMR spectrum of **6v**



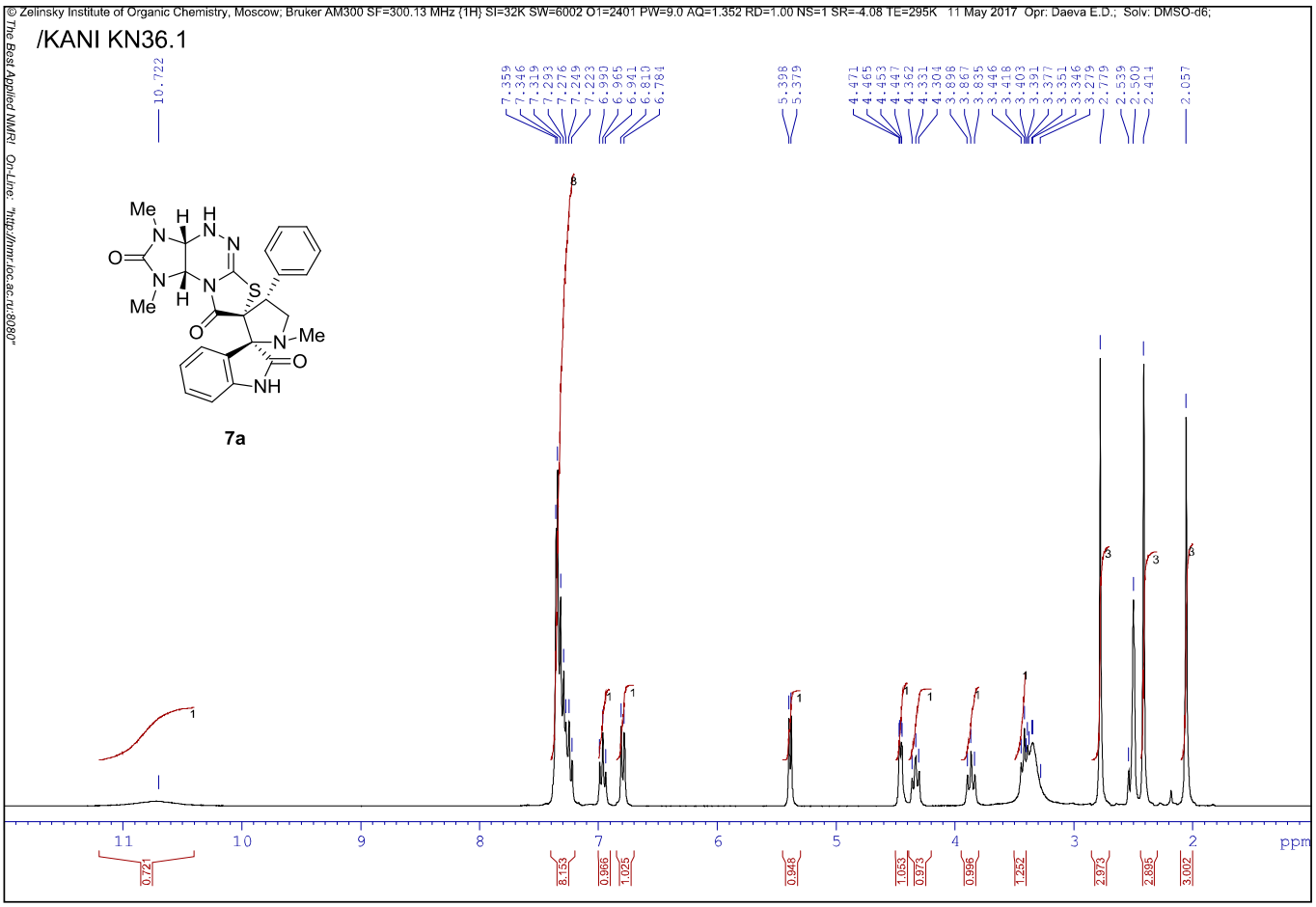
^{1}H NMR spectrum of **6w**



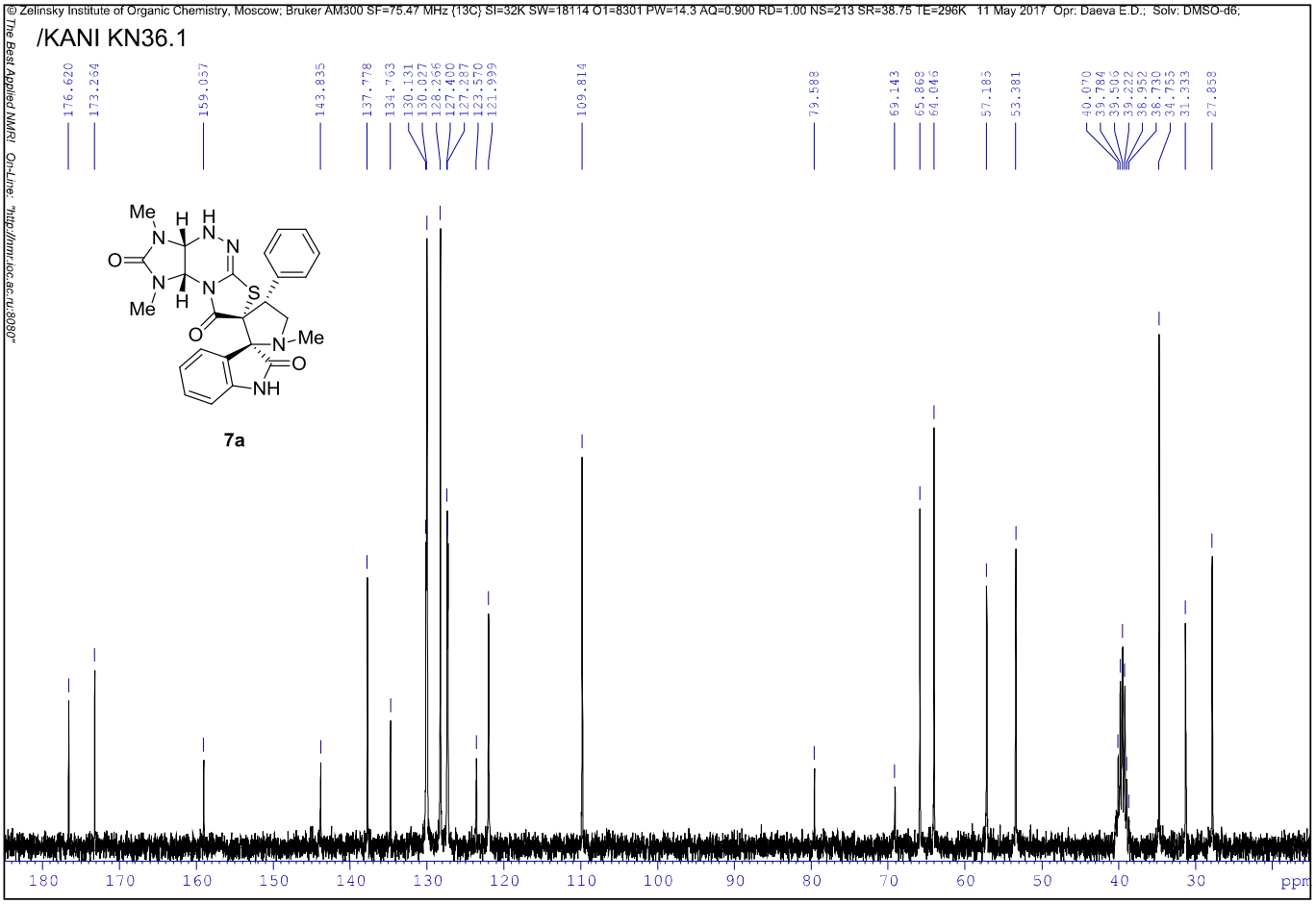
^{13}C NMR spectrum of **6w**



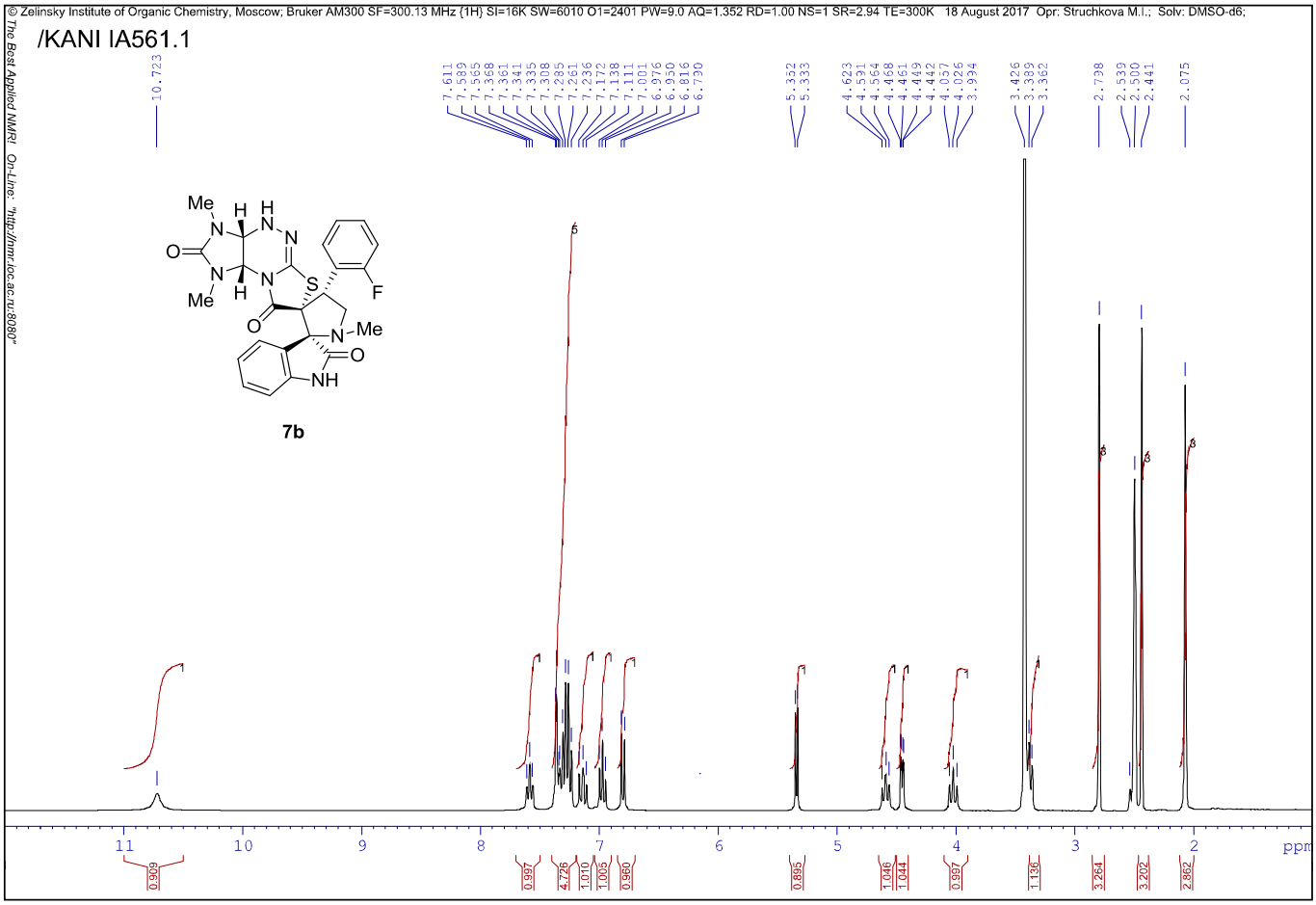
^{1}H NMR spectrum of **7a**



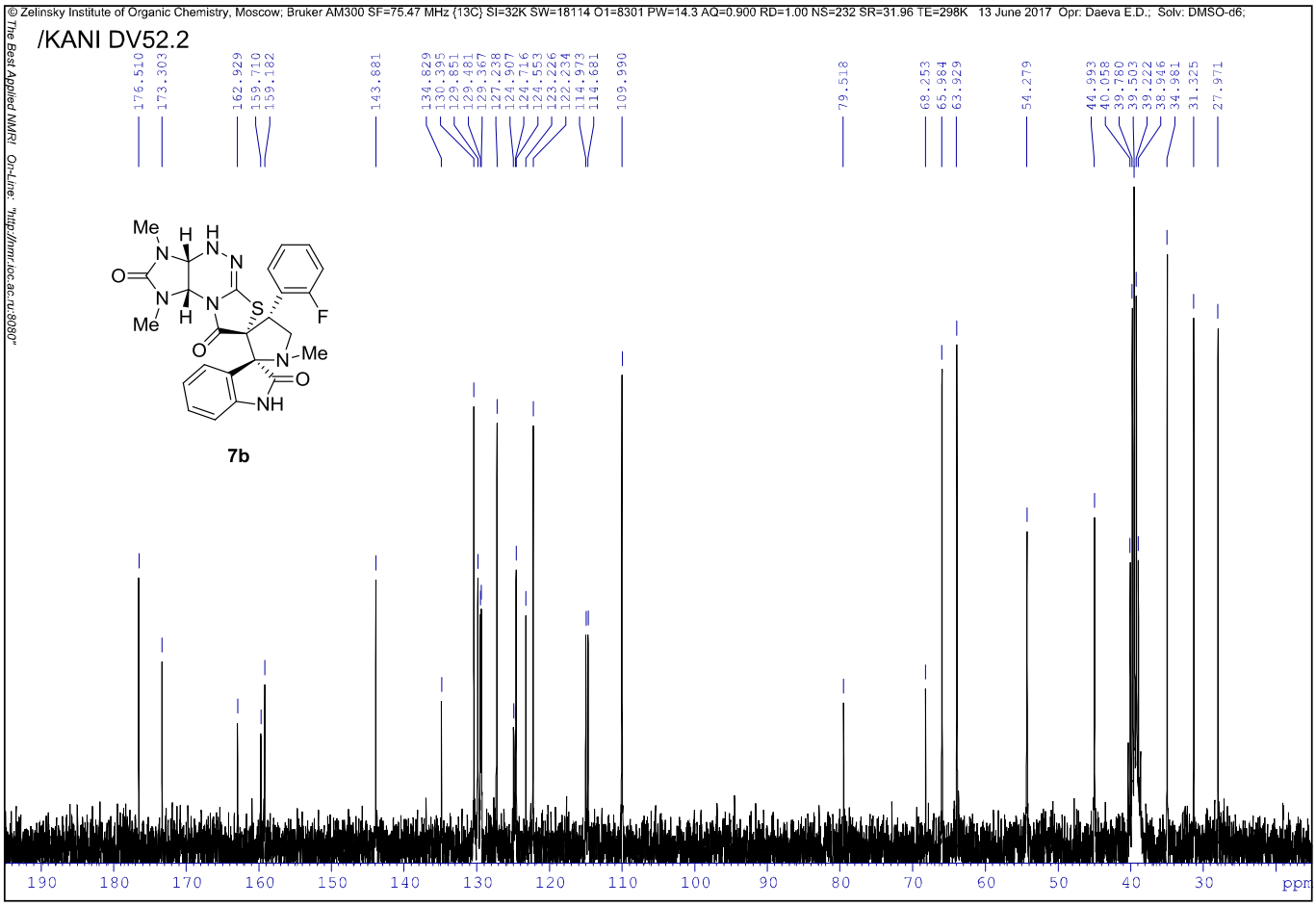
^{13}C NMR spectrum of **7a**



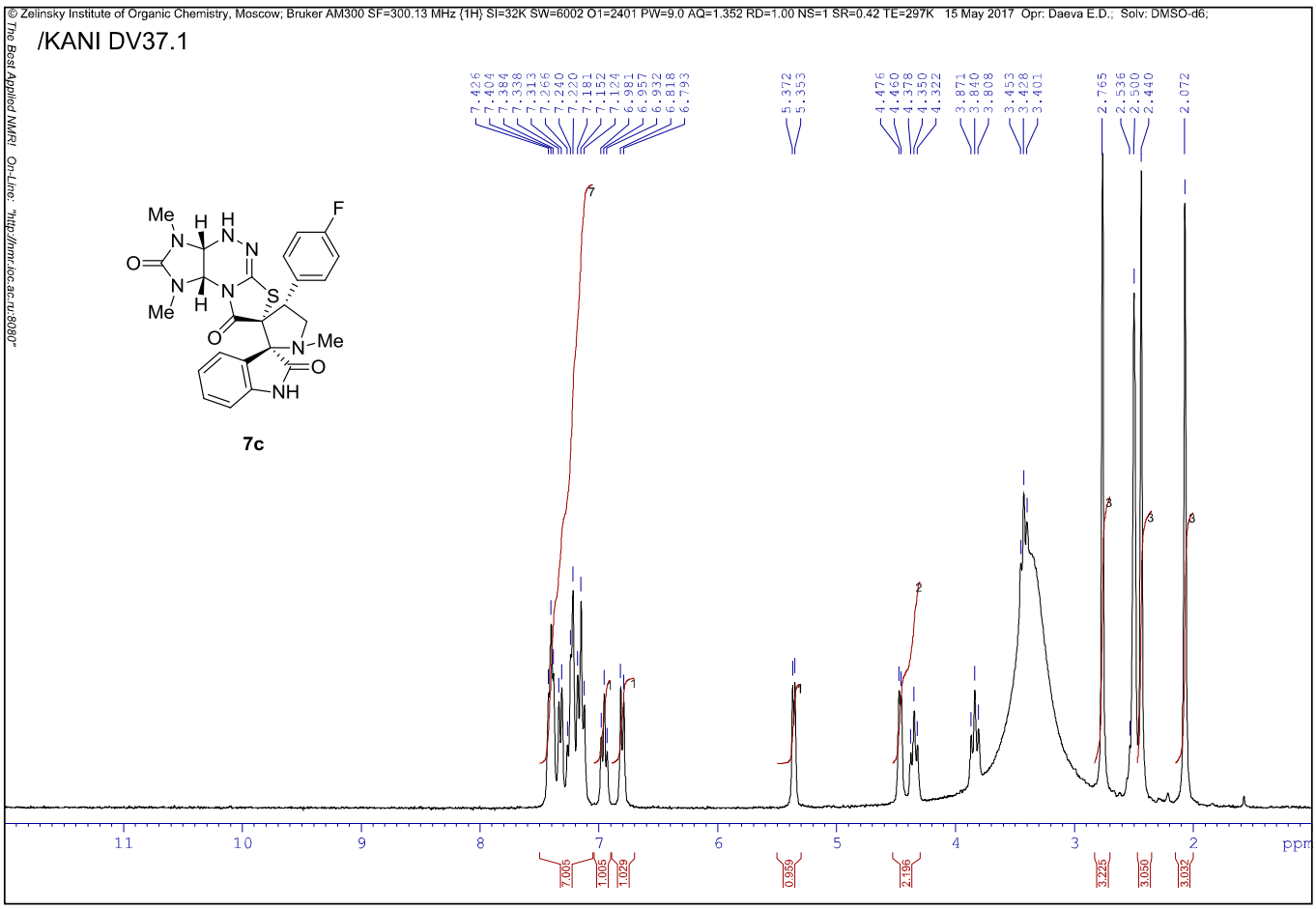
^{1}H NMR spectrum of **7b**



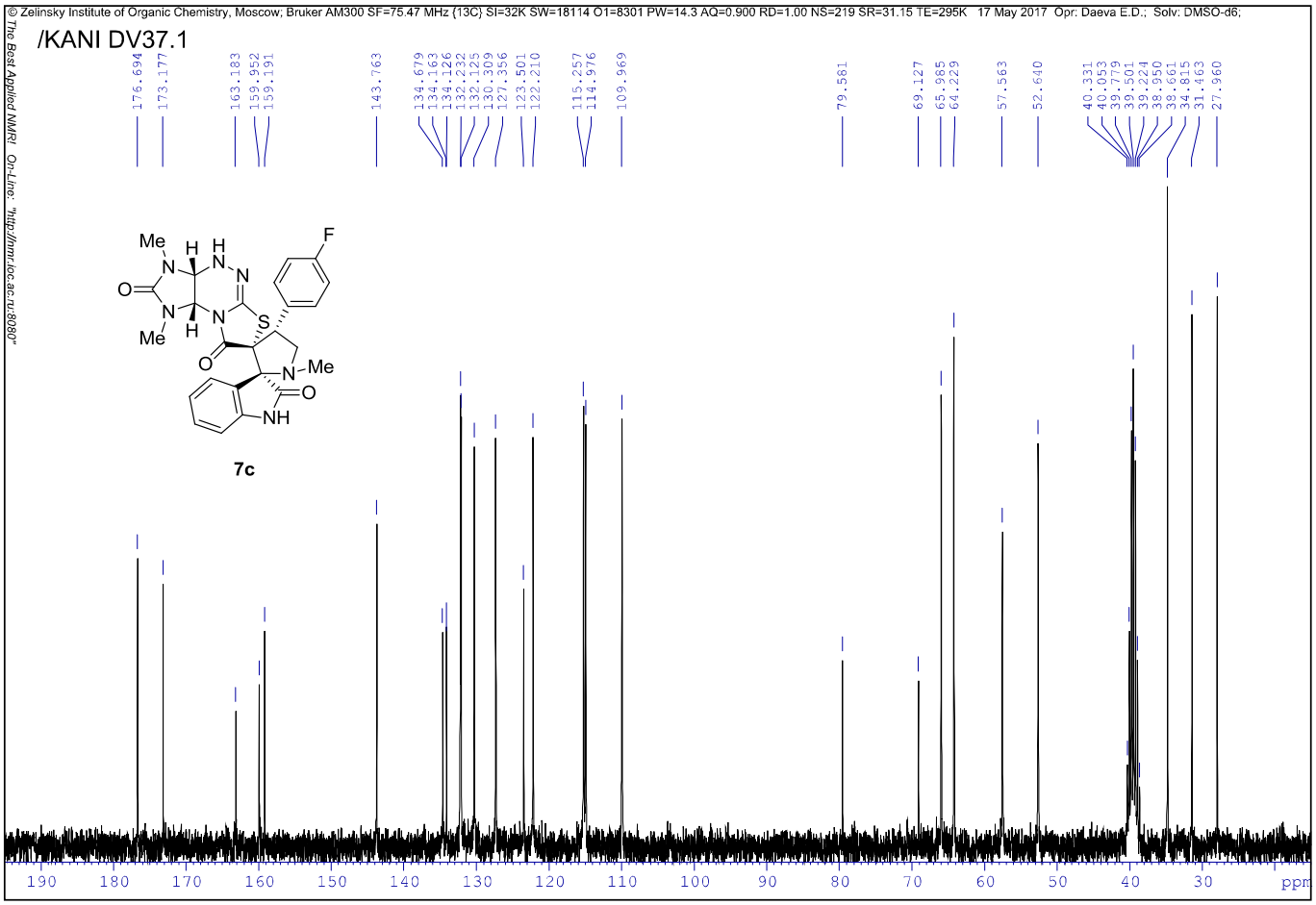
^{13}C NMR spectrum of **7b**



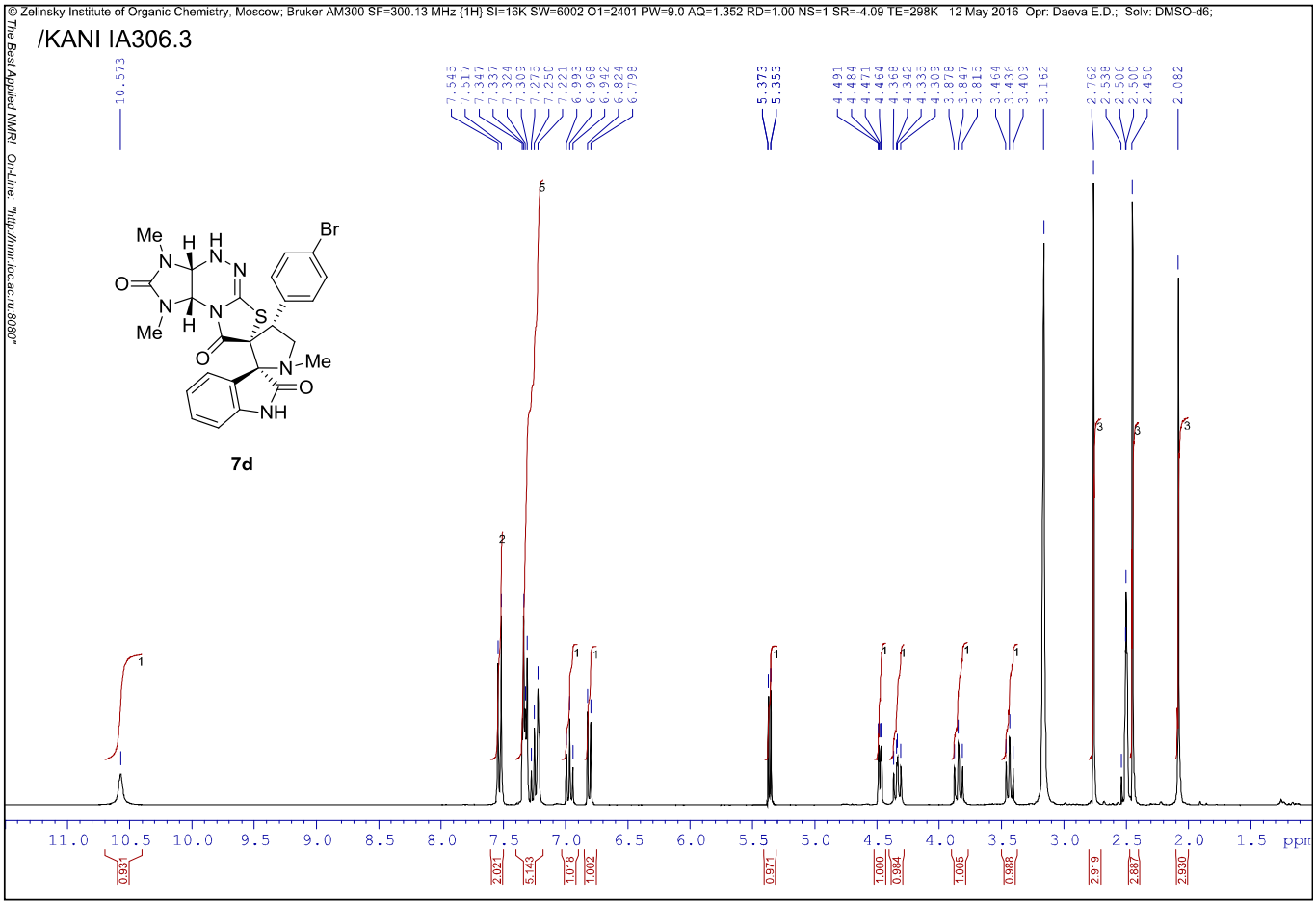
^{1}H NMR spectrum of **7c**



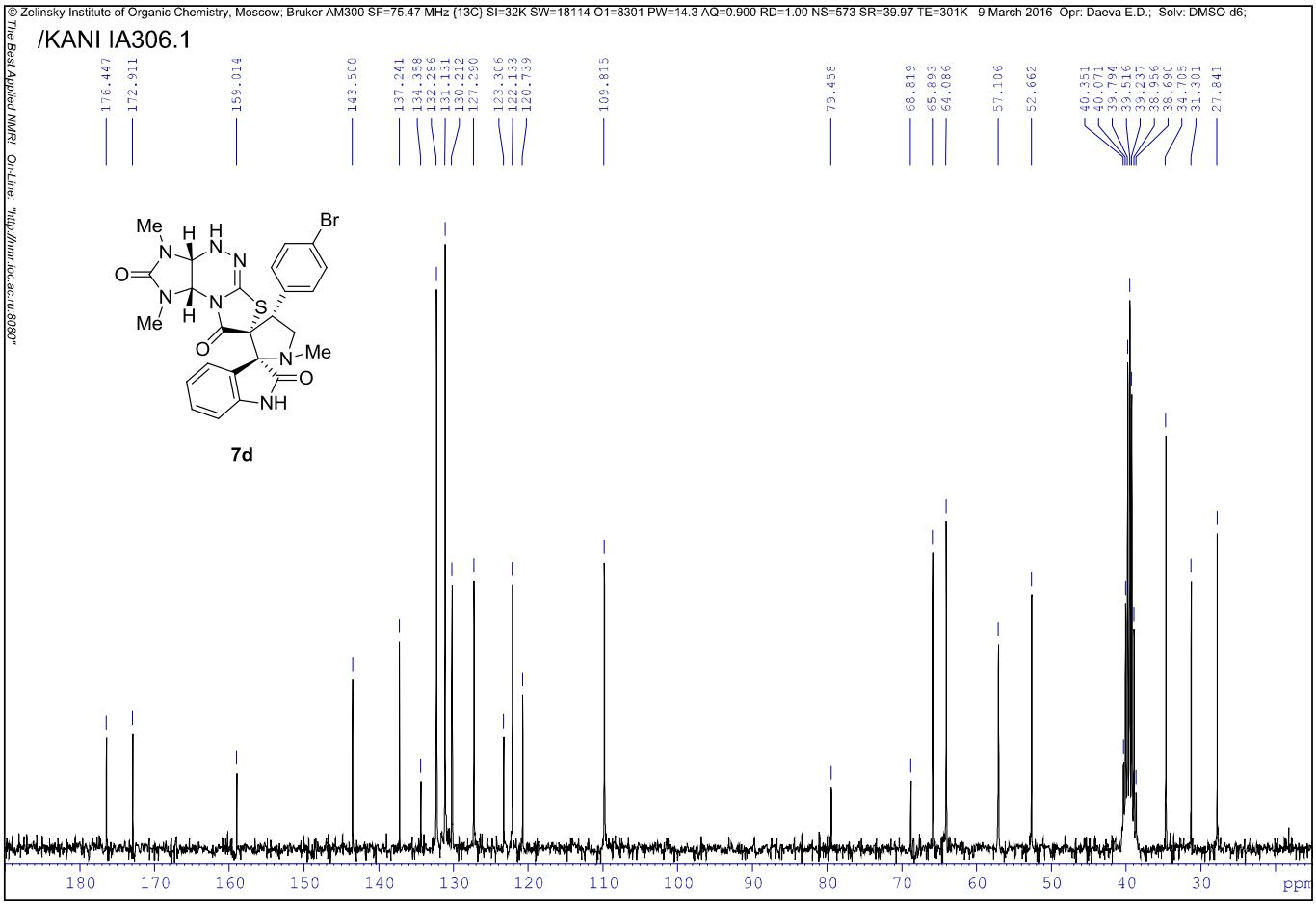
^{13}C NMR spectrum of **7c**



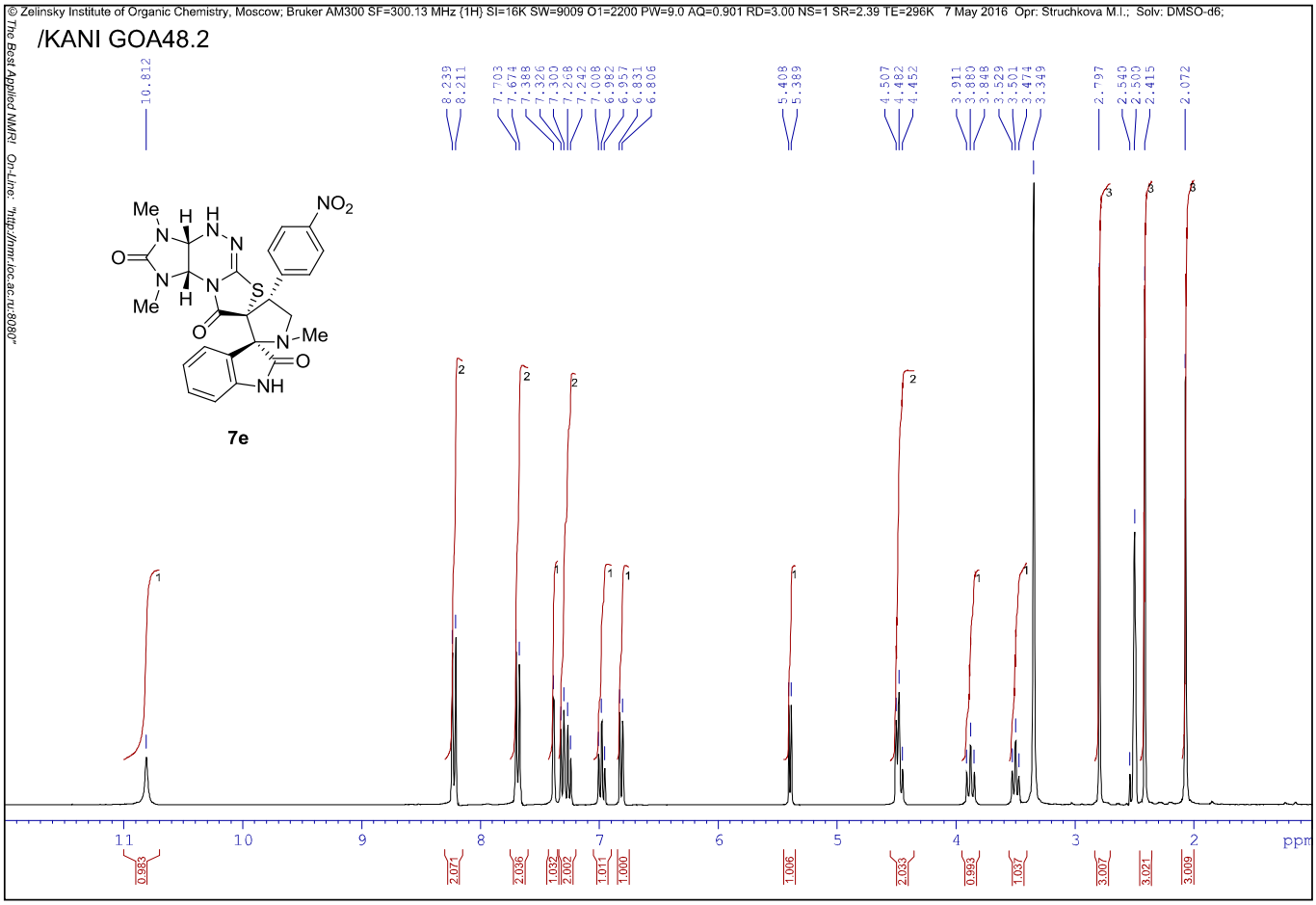
^{1}H NMR spectrum of **7d**



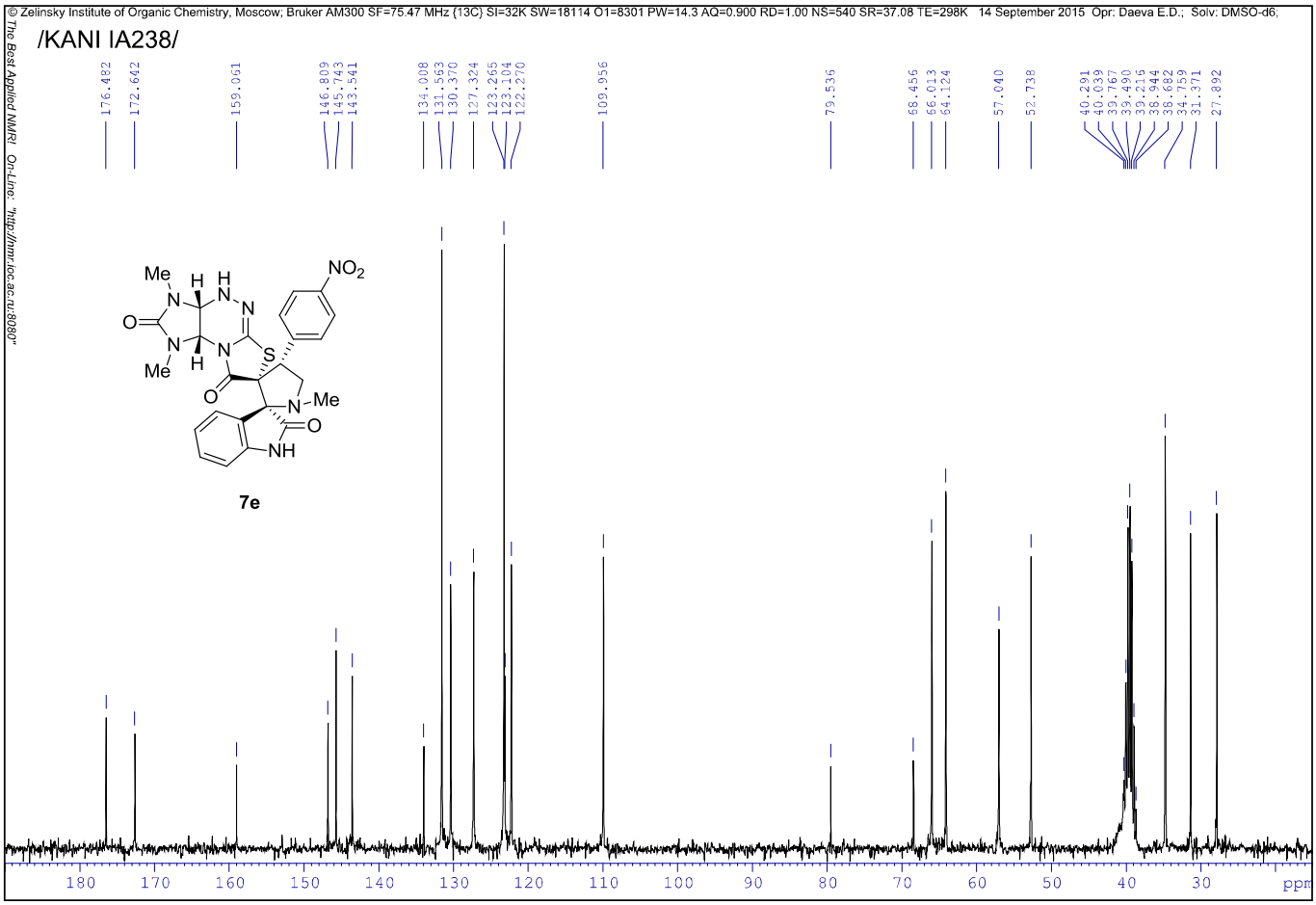
^{13}C NMR spectrum of **7d**



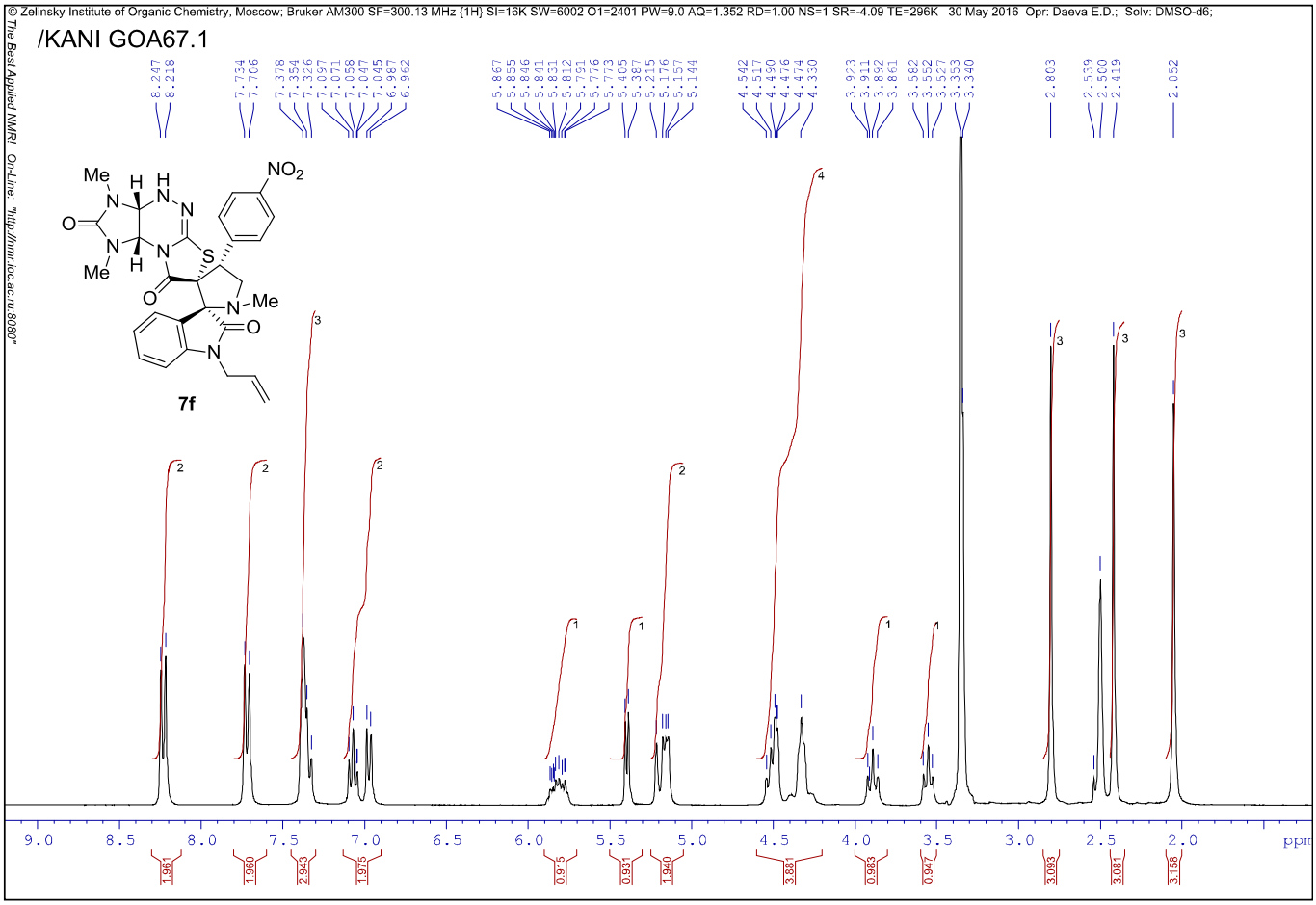
^{1}H NMR spectrum of **7e**



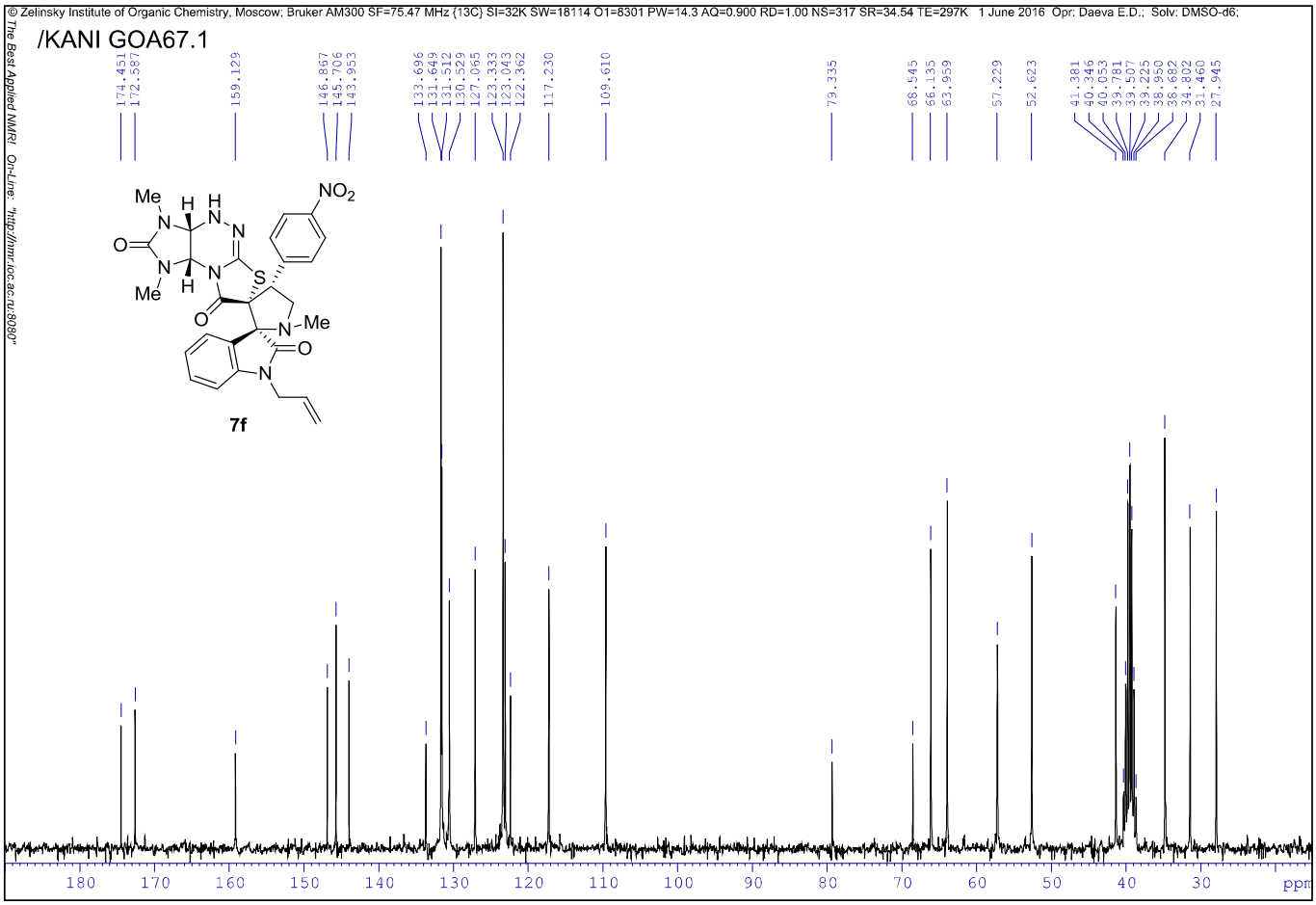
^{13}C NMR spectrum of **7e**



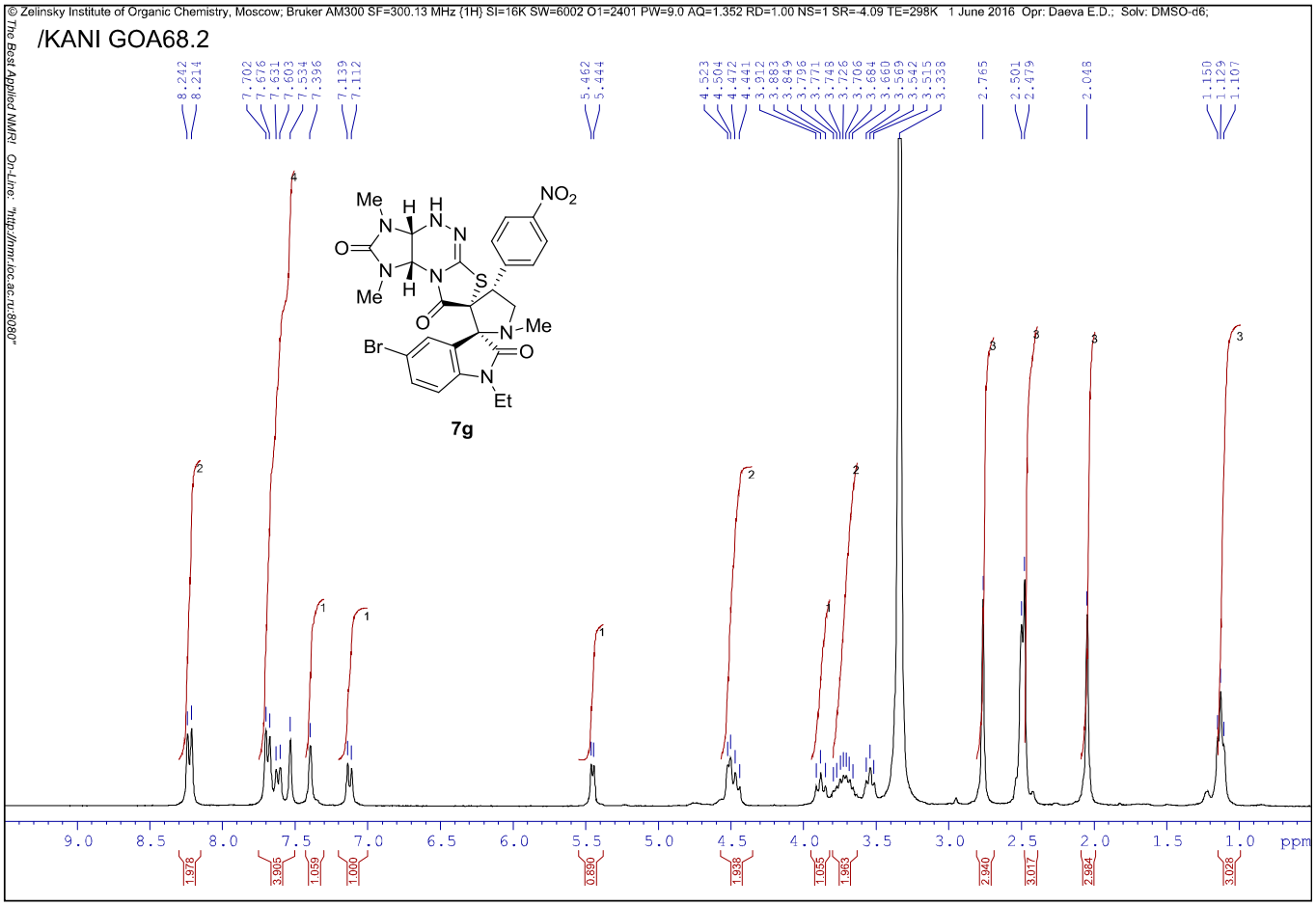
^{1}H NMR spectrum of **7f**



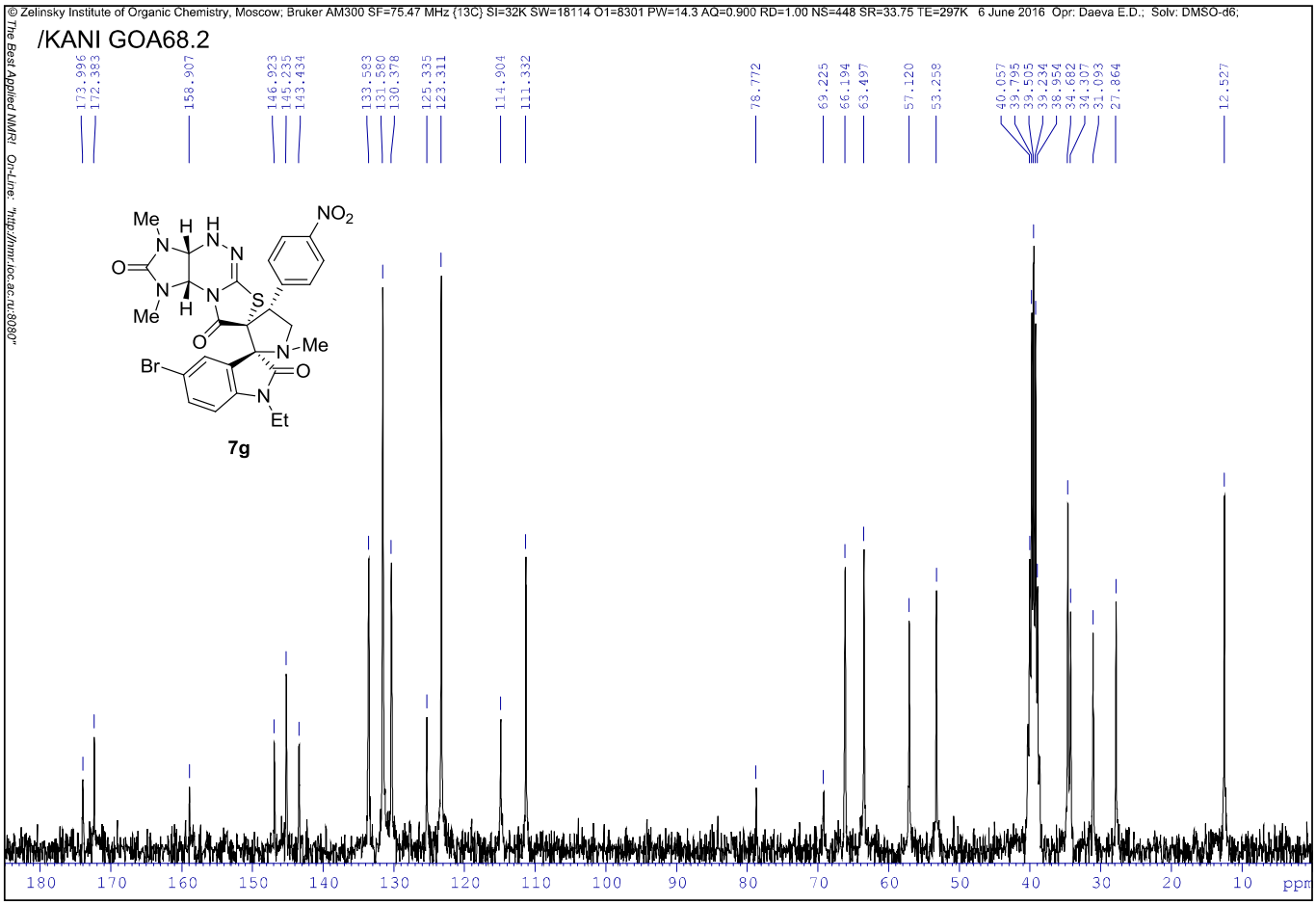
^{13}C NMR spectrum of **7f**



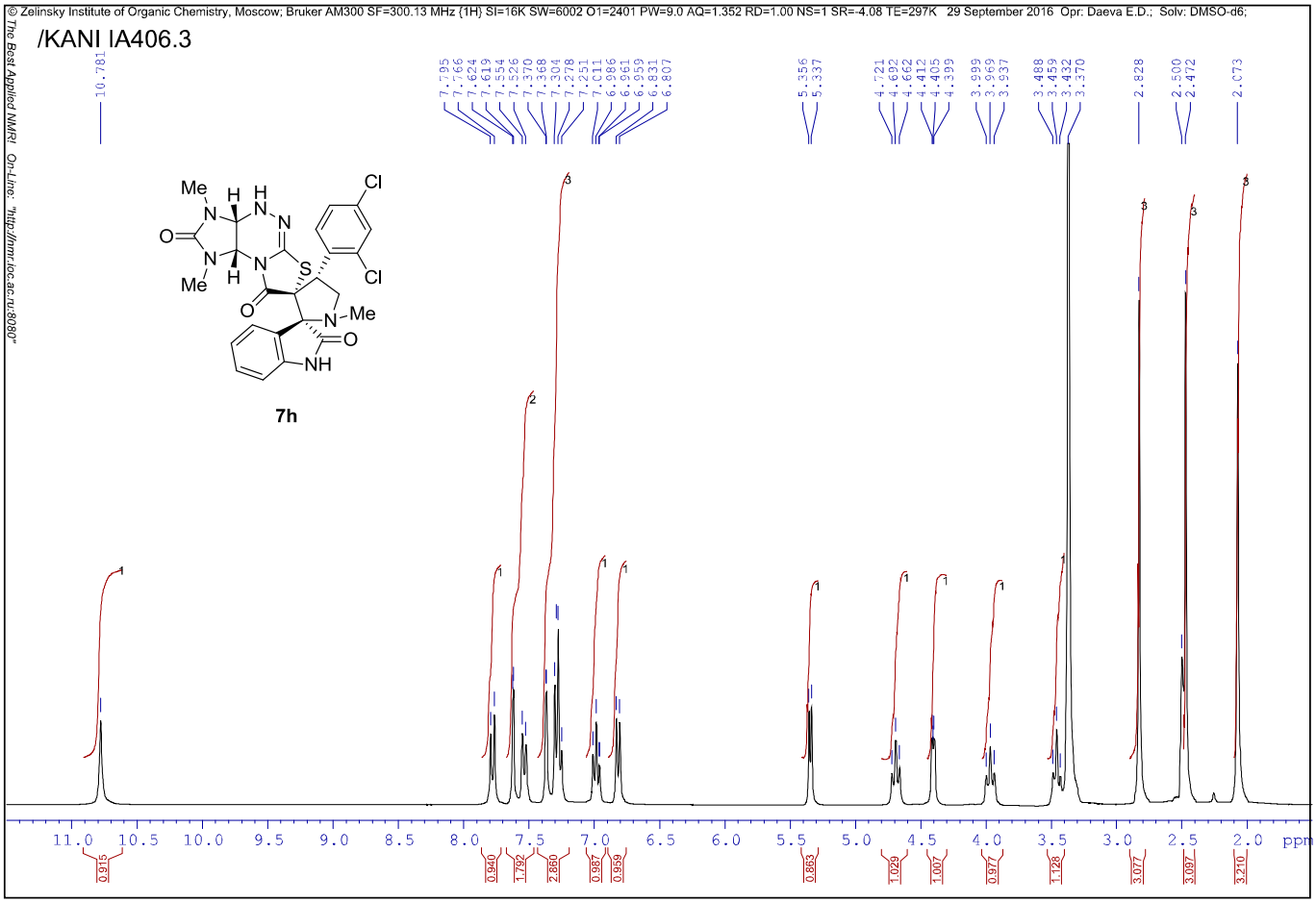
^{1}H NMR spectrum of **7g**



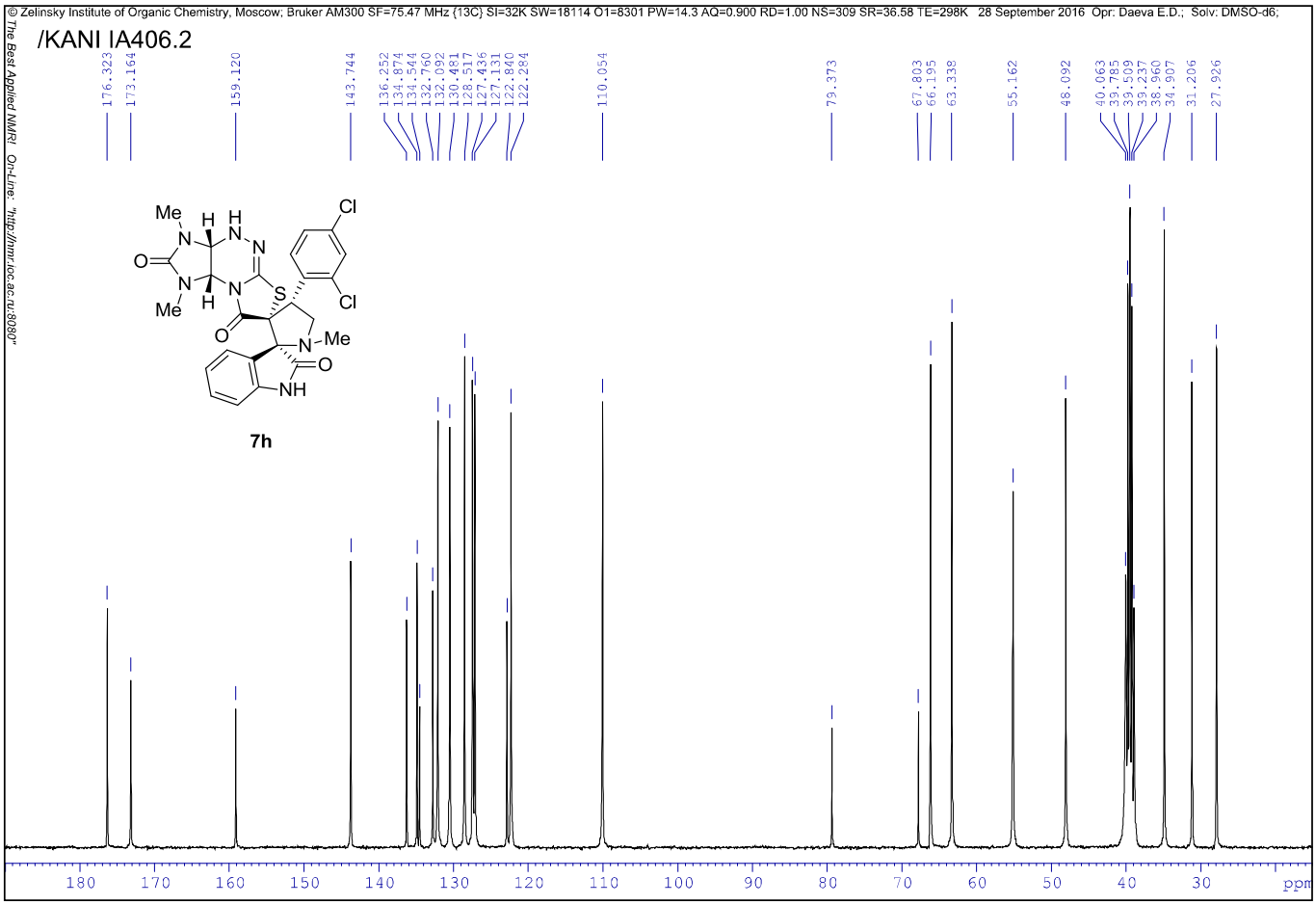
^{13}C NMR spectrum of **7g**



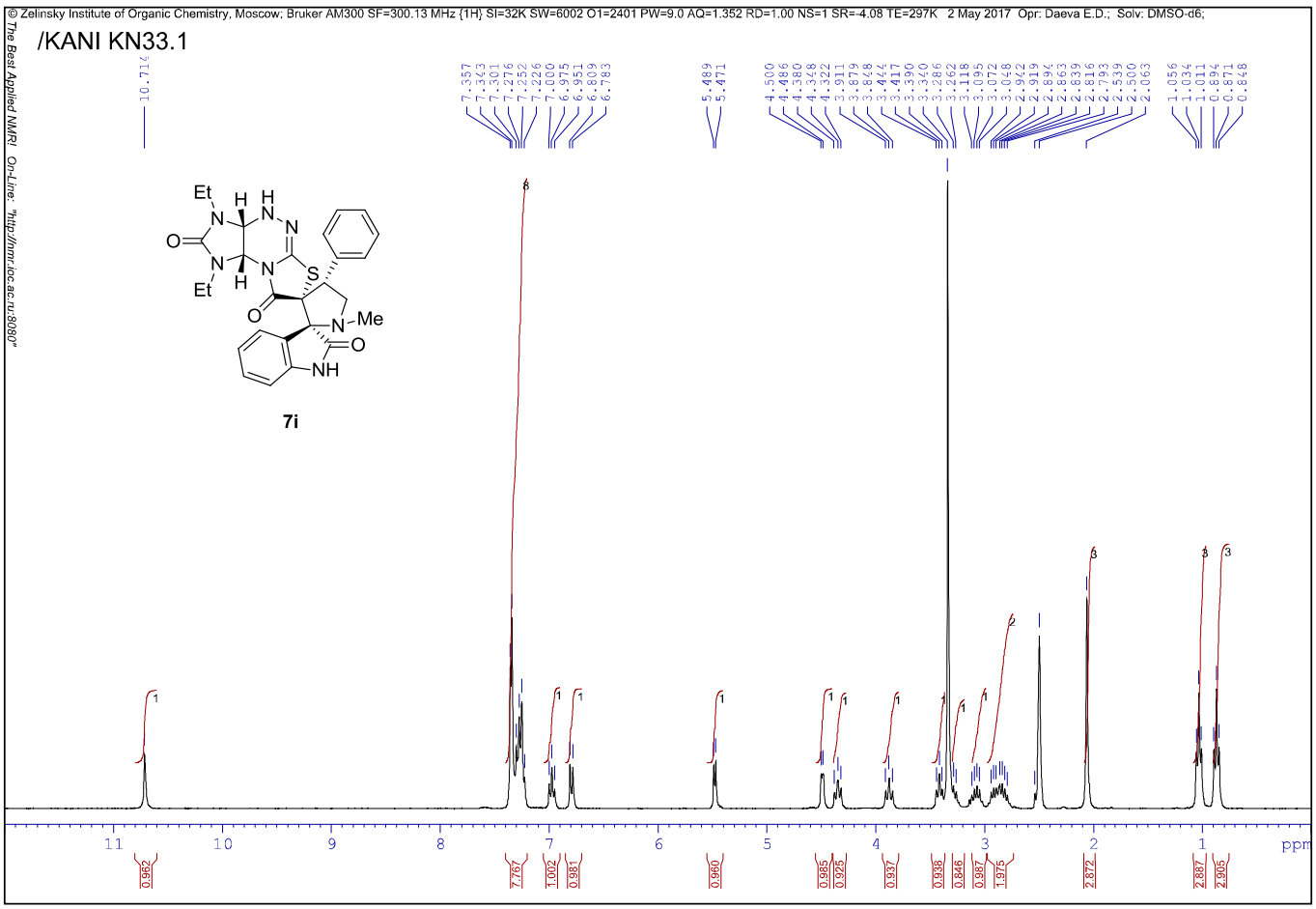
^{1}H NMR spectrum of **7h**



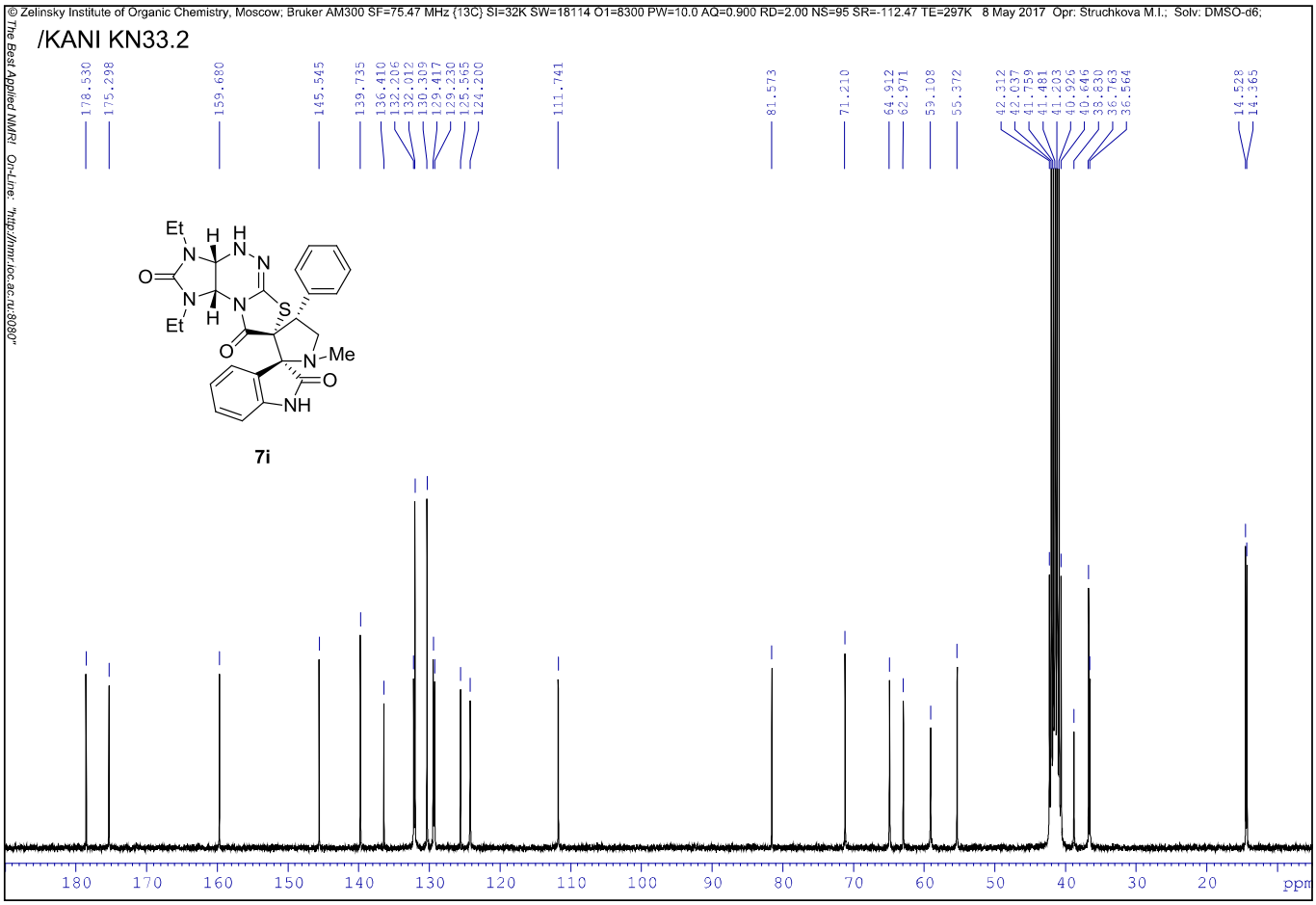
^{13}C NMR spectrum of **7h**



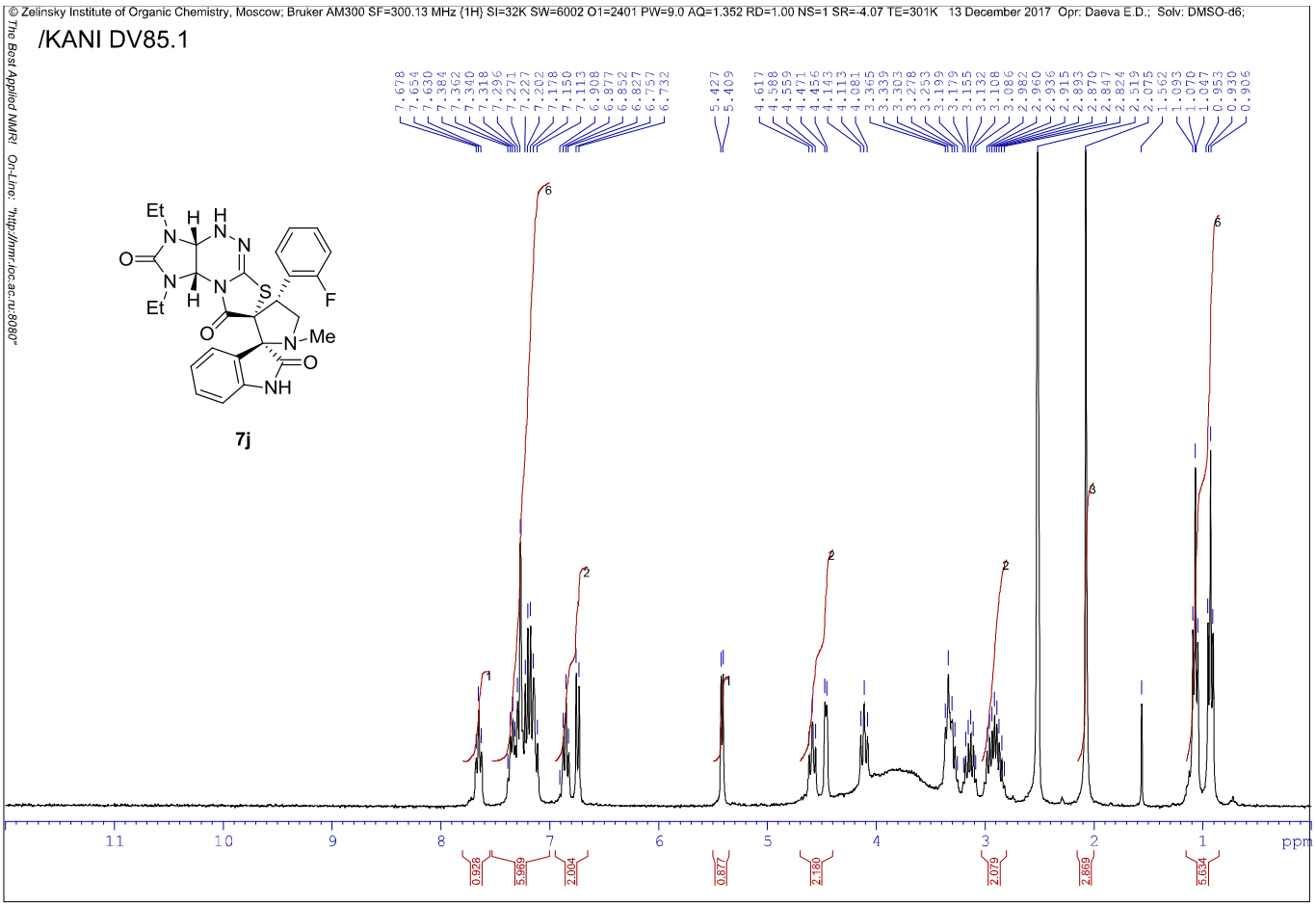
^{1}H NMR spectrum of **7i**



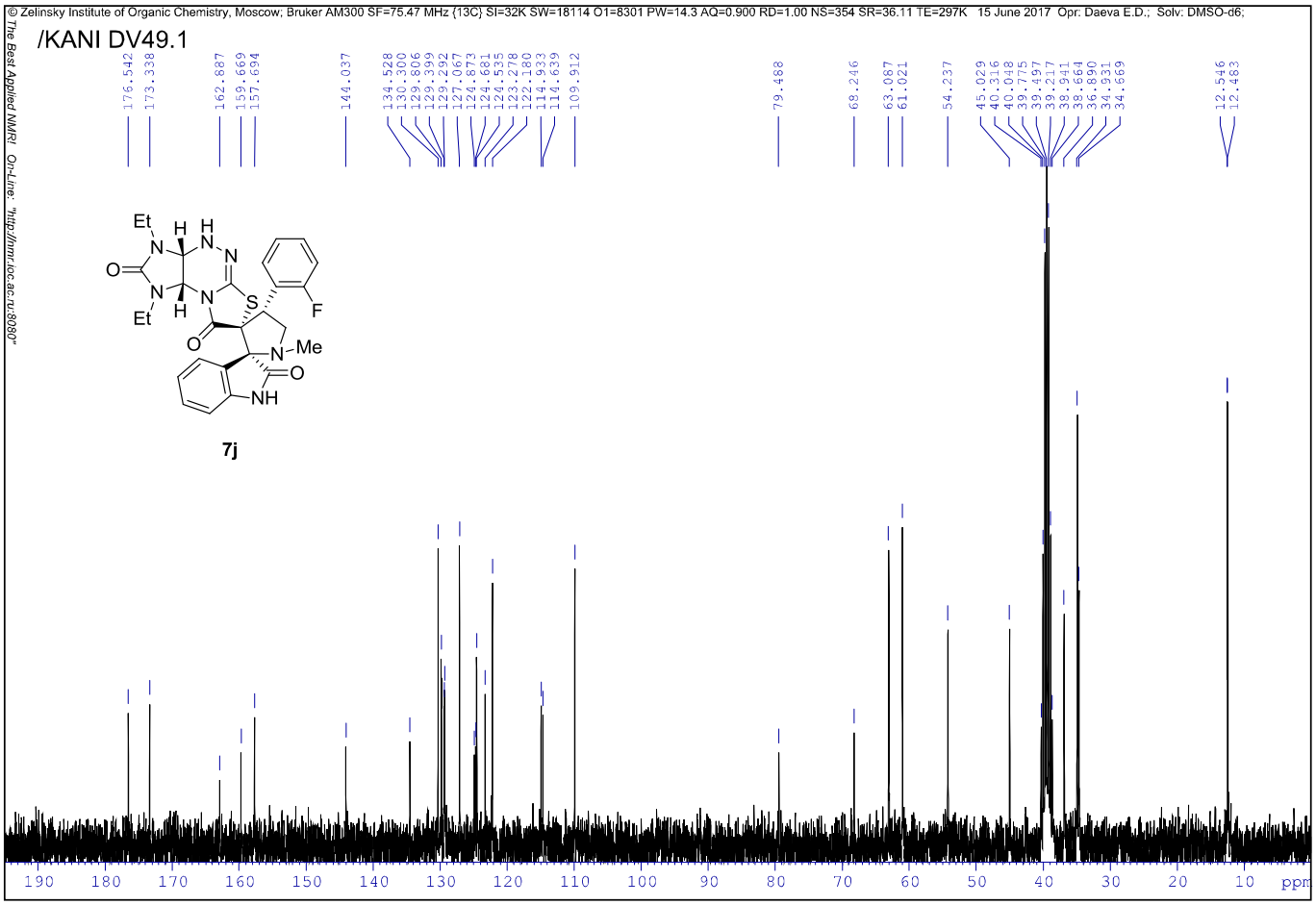
^{13}C NMR spectrum of **7i**



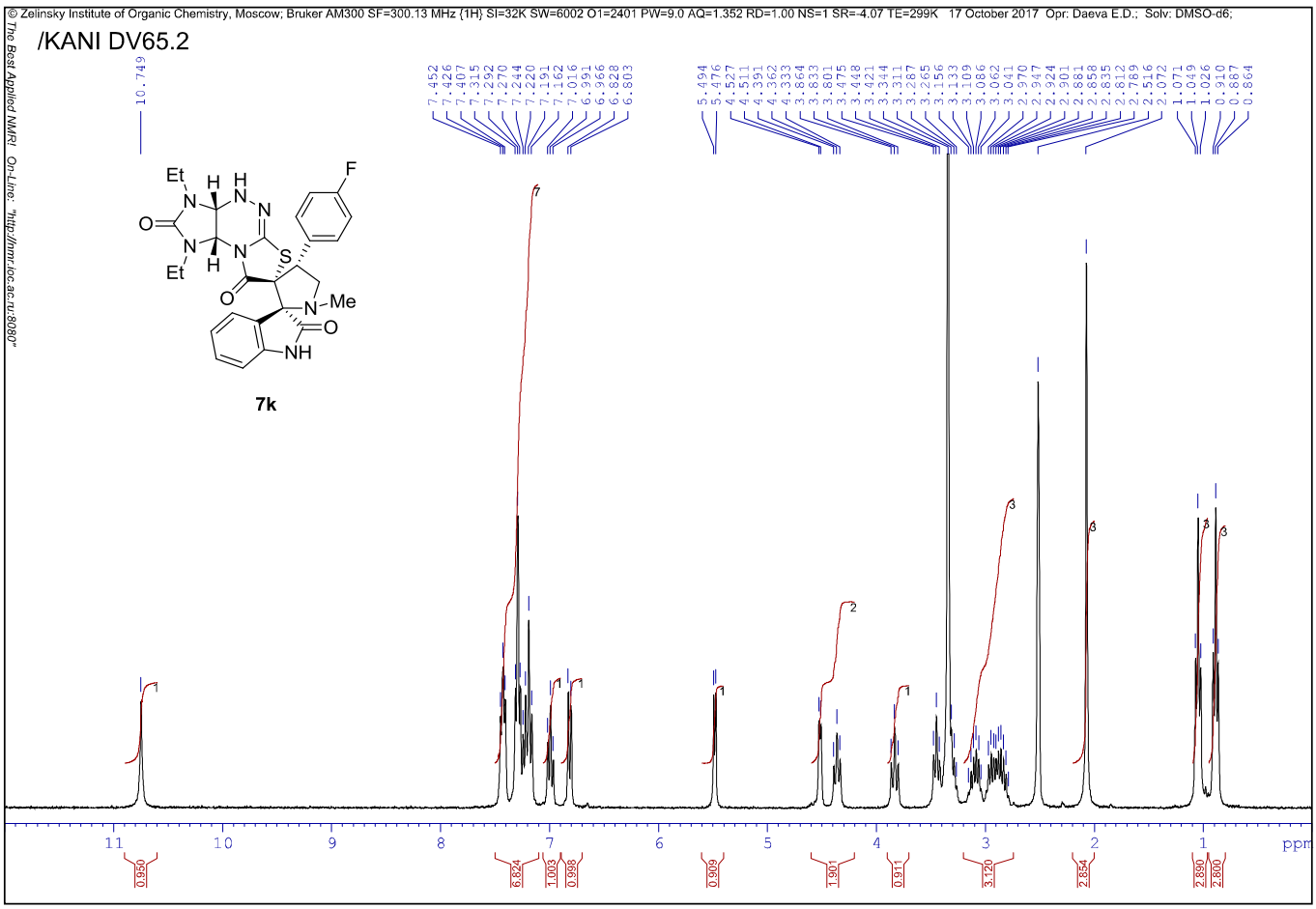
^{1}H NMR spectrum of **7j**



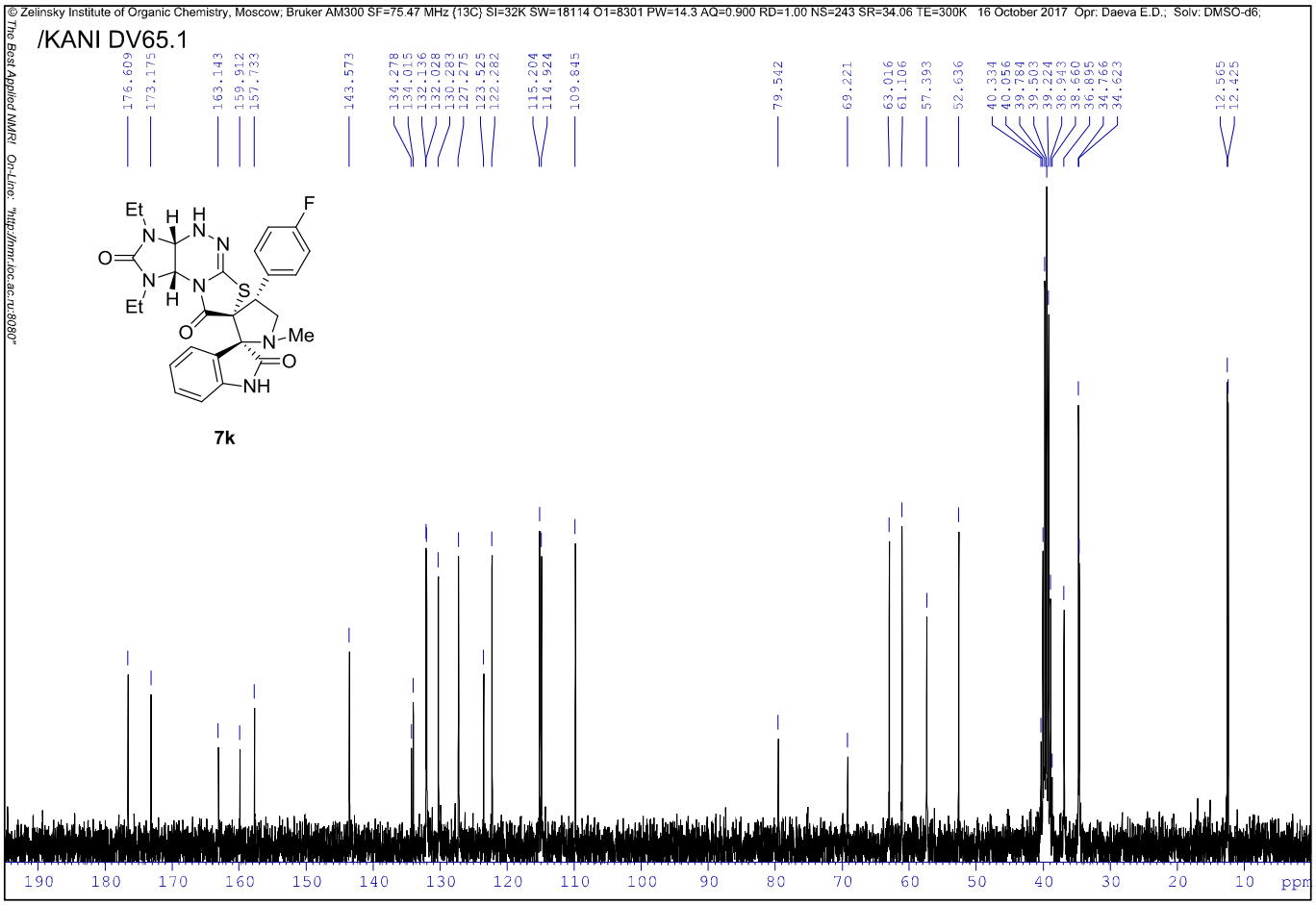
^{13}C NMR spectrum of **7j**



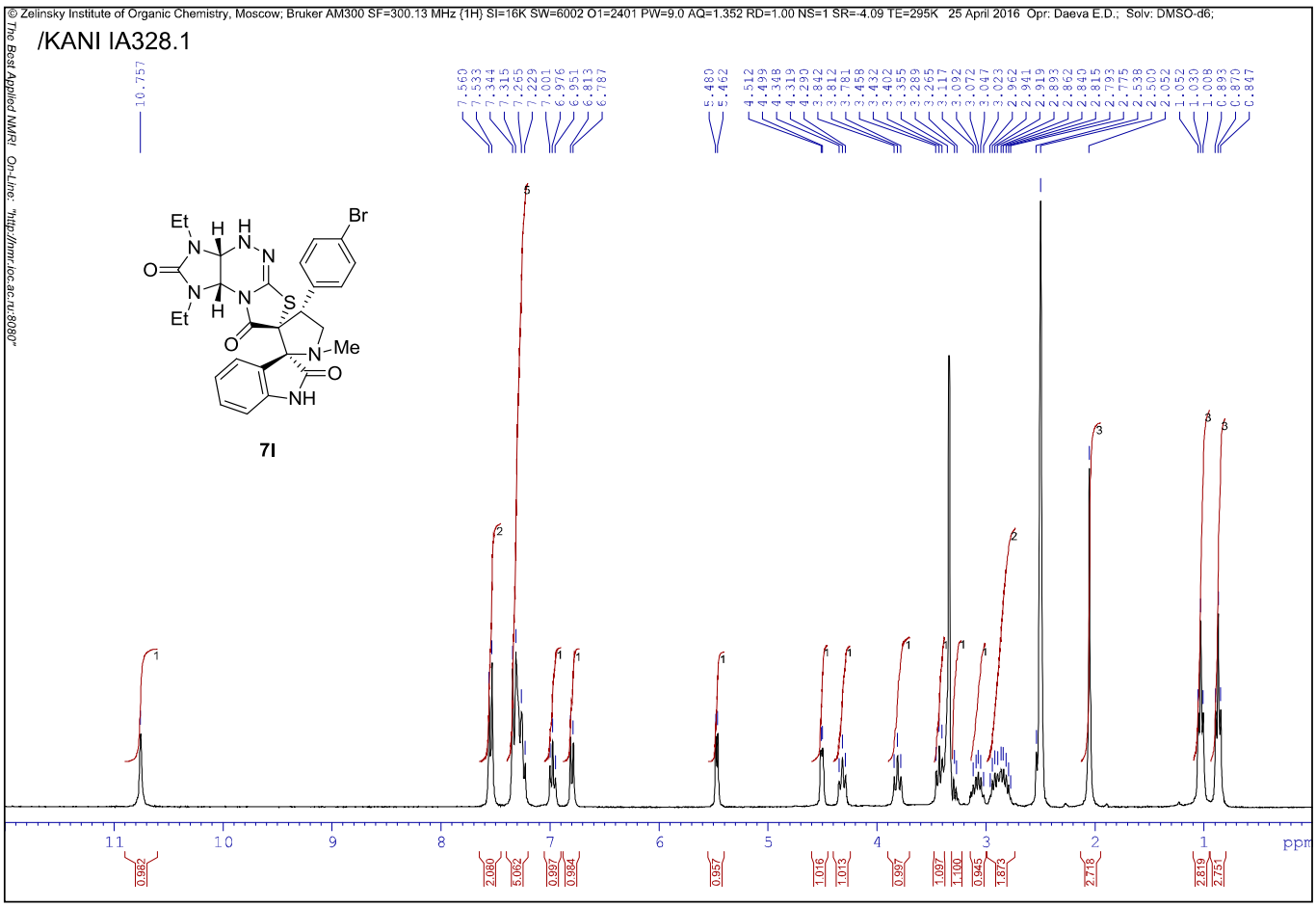
^{1}H NMR spectrum of **7k**



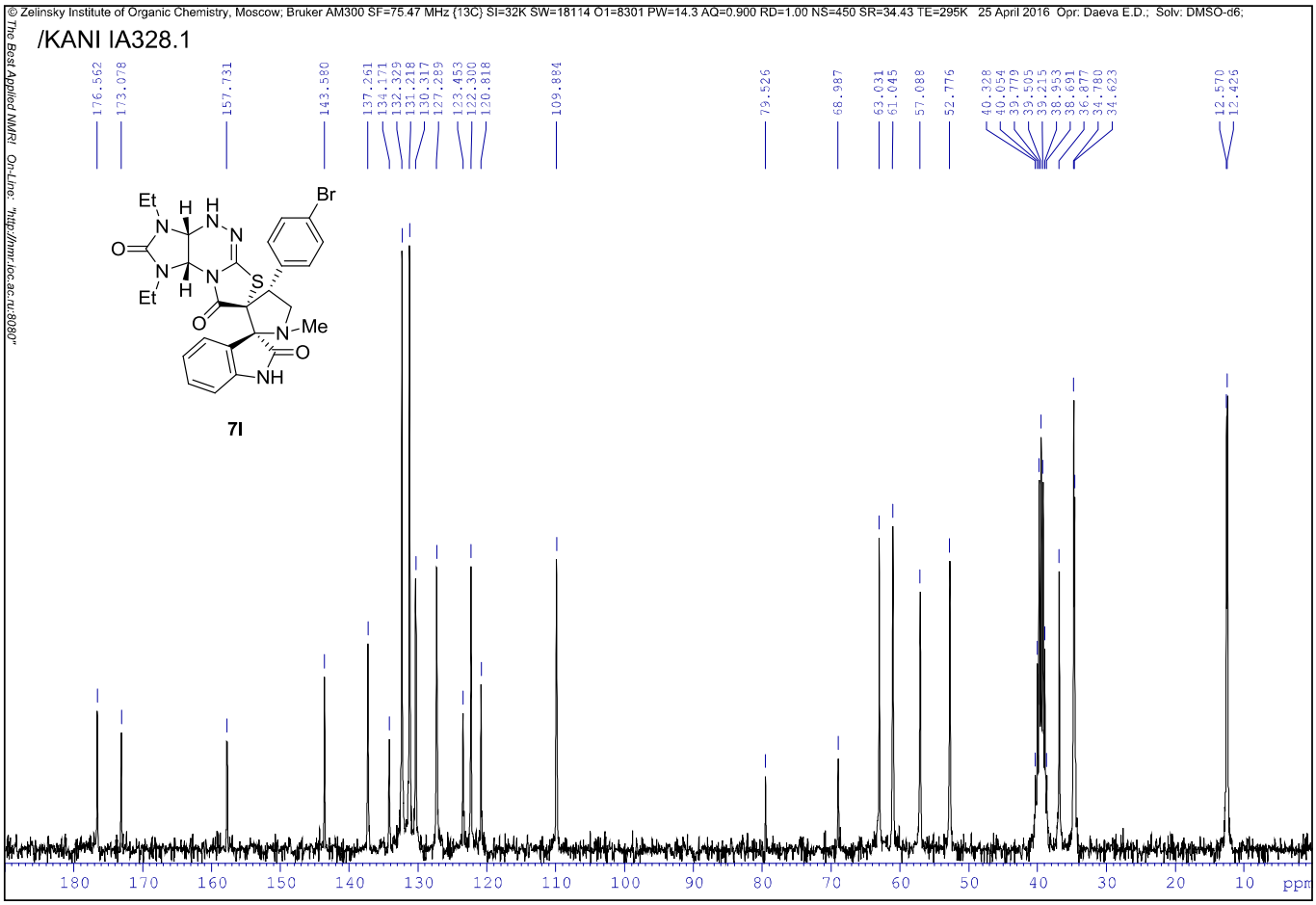
^{13}C NMR spectrum of **7k**



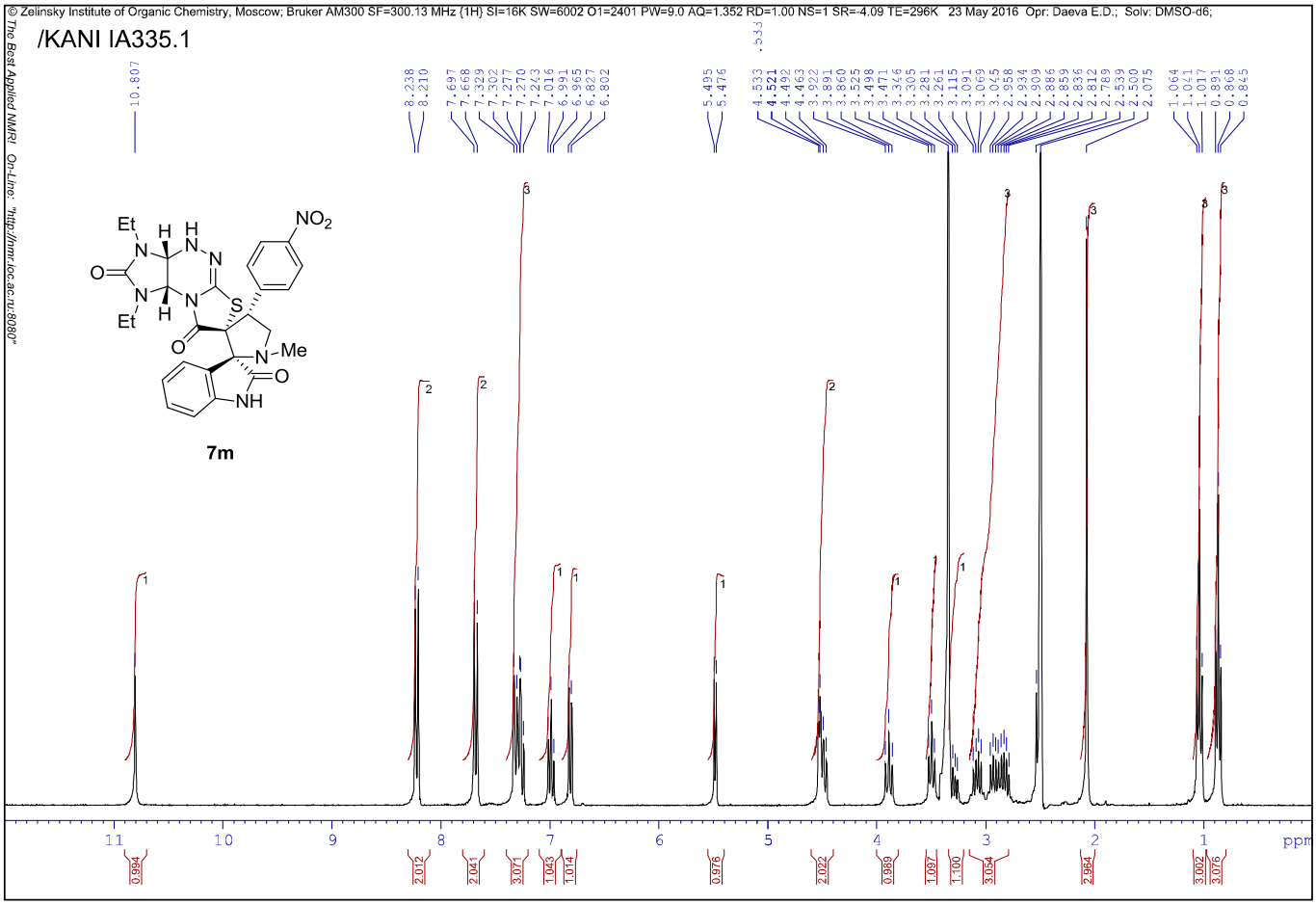
^{1}H NMR spectrum of **7l**



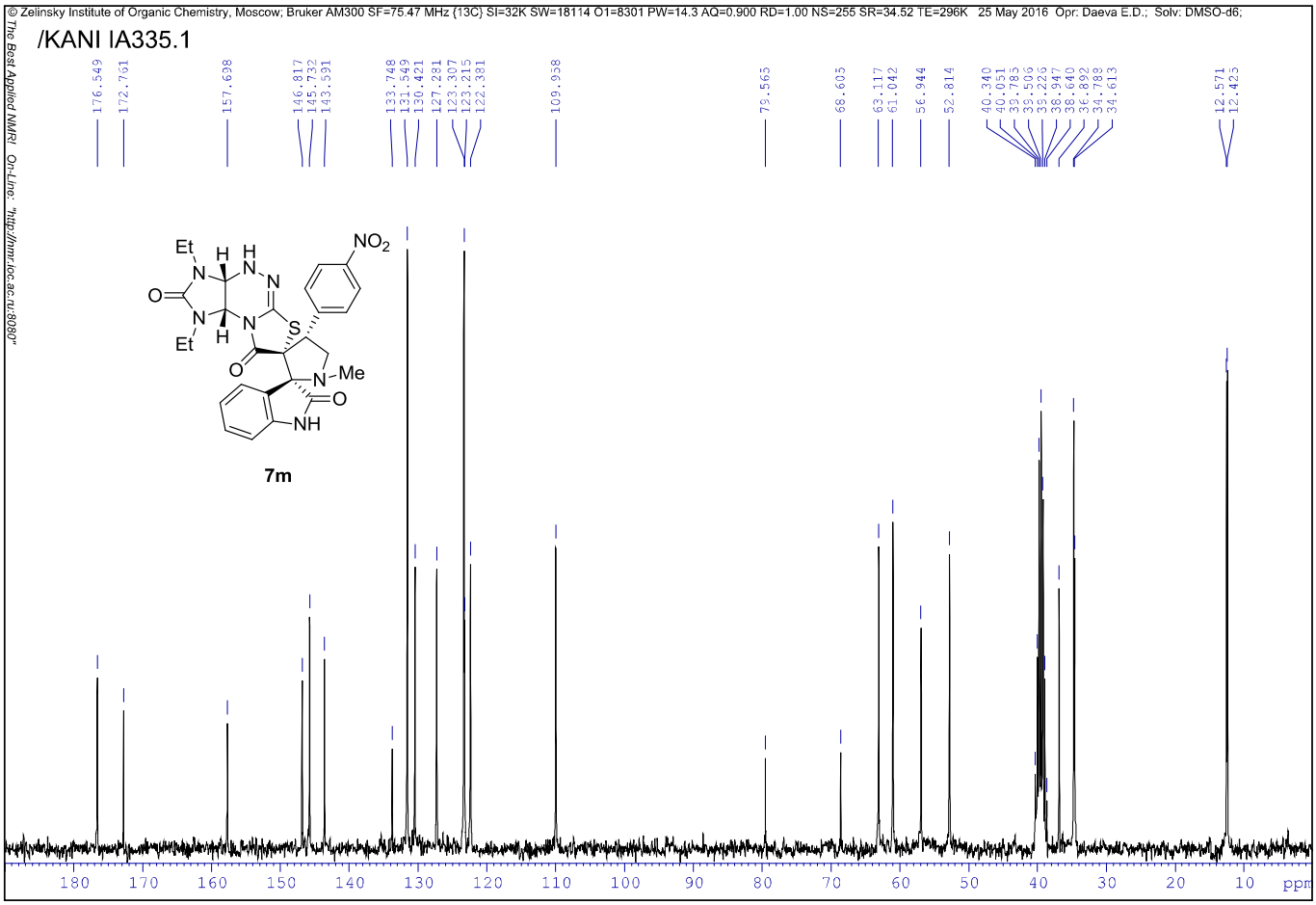
^{13}C NMR spectrum of **7l**



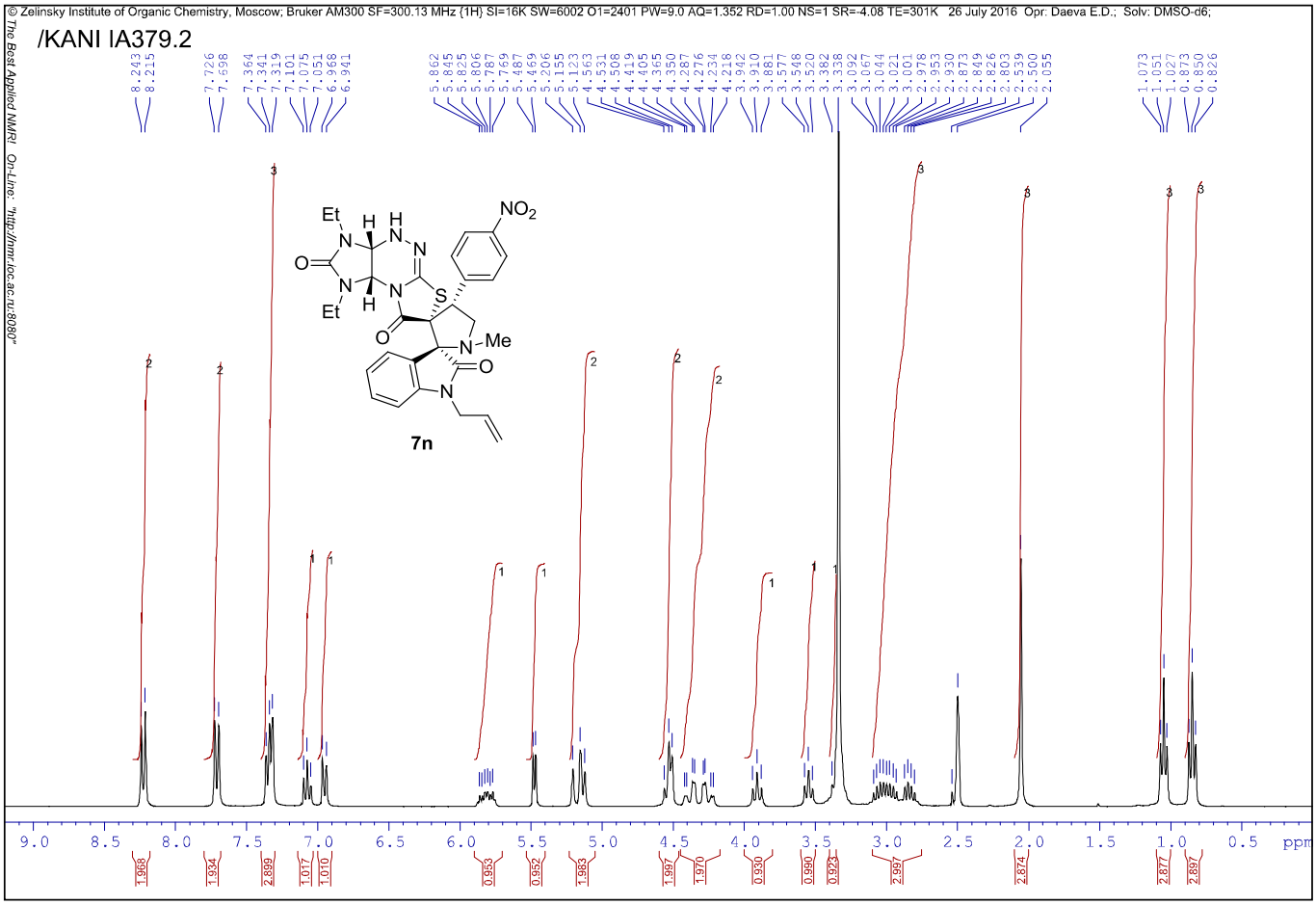
^{1}H NMR spectrum of **7m**



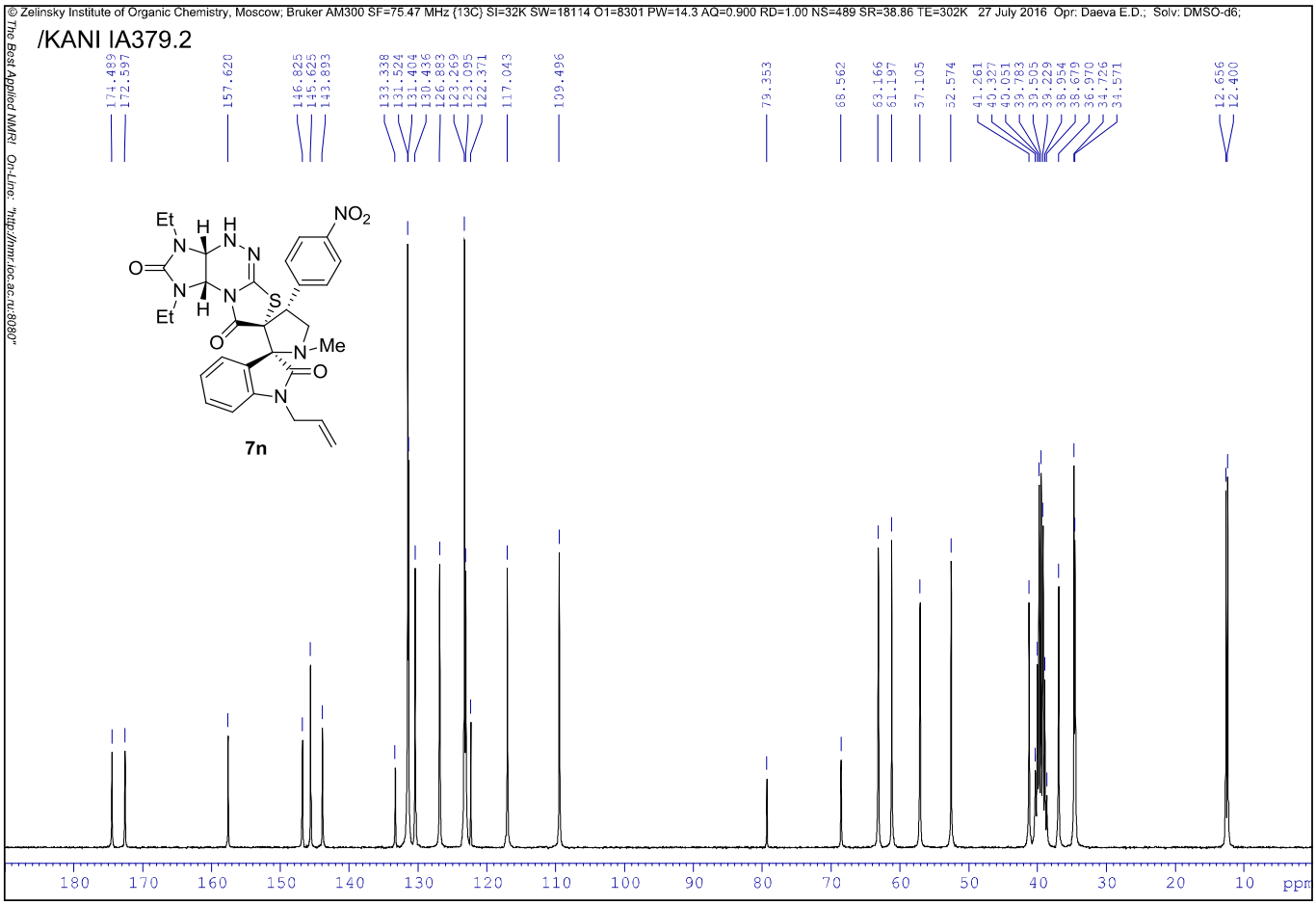
^{13}C NMR spectrum of **7m**



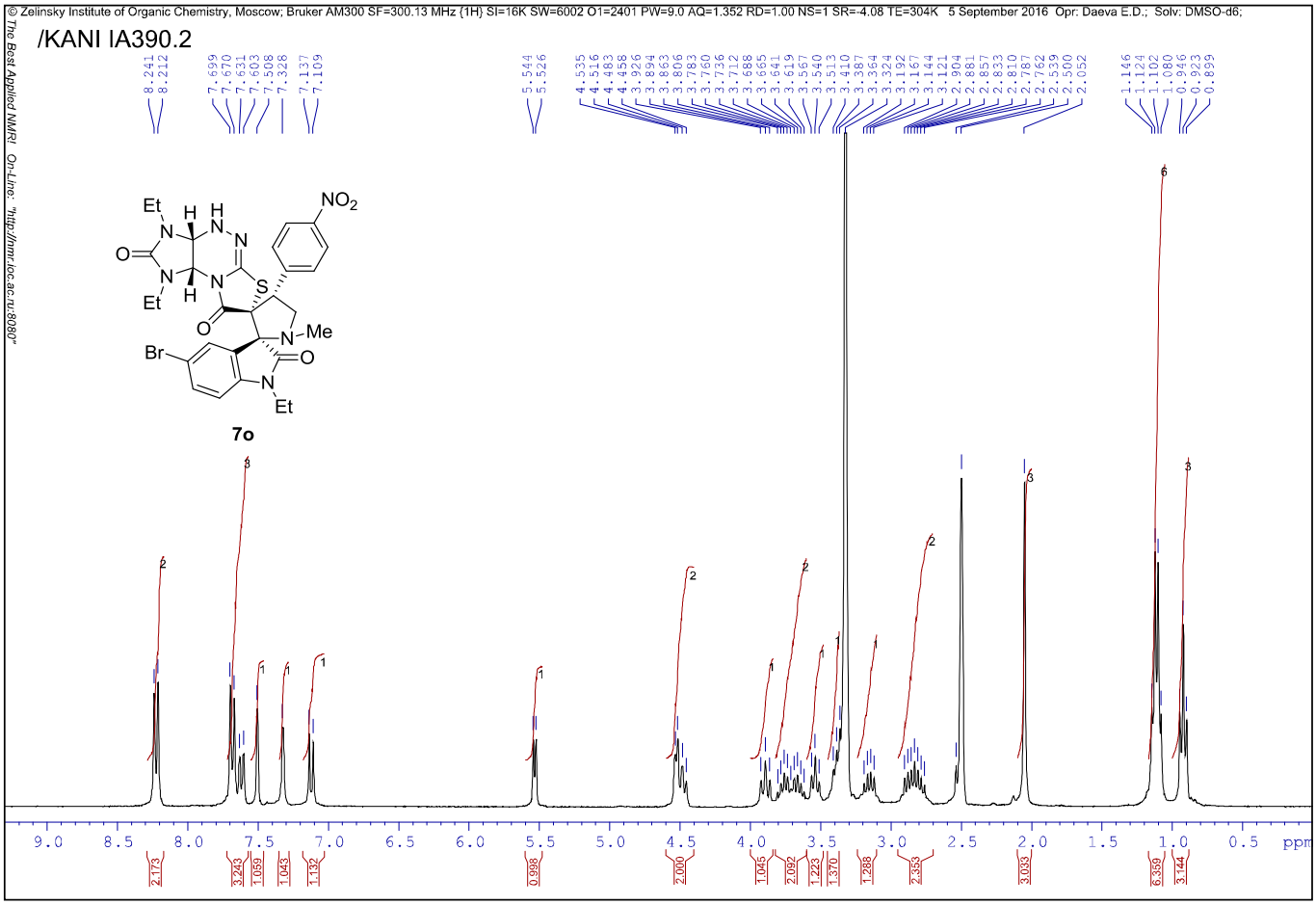
^{1}H NMR spectrum of **7n**



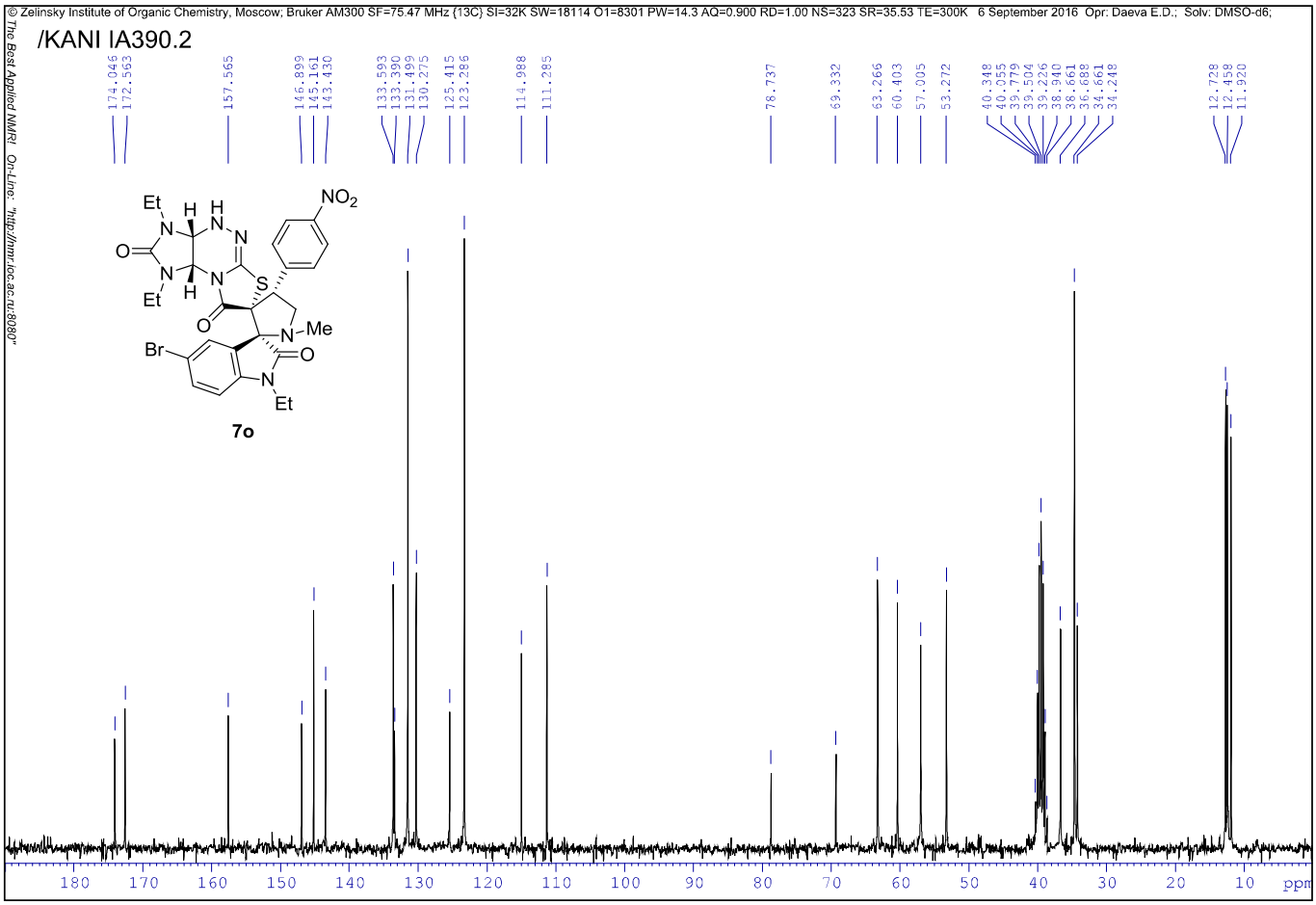
^{13}C NMR spectrum of **7n**



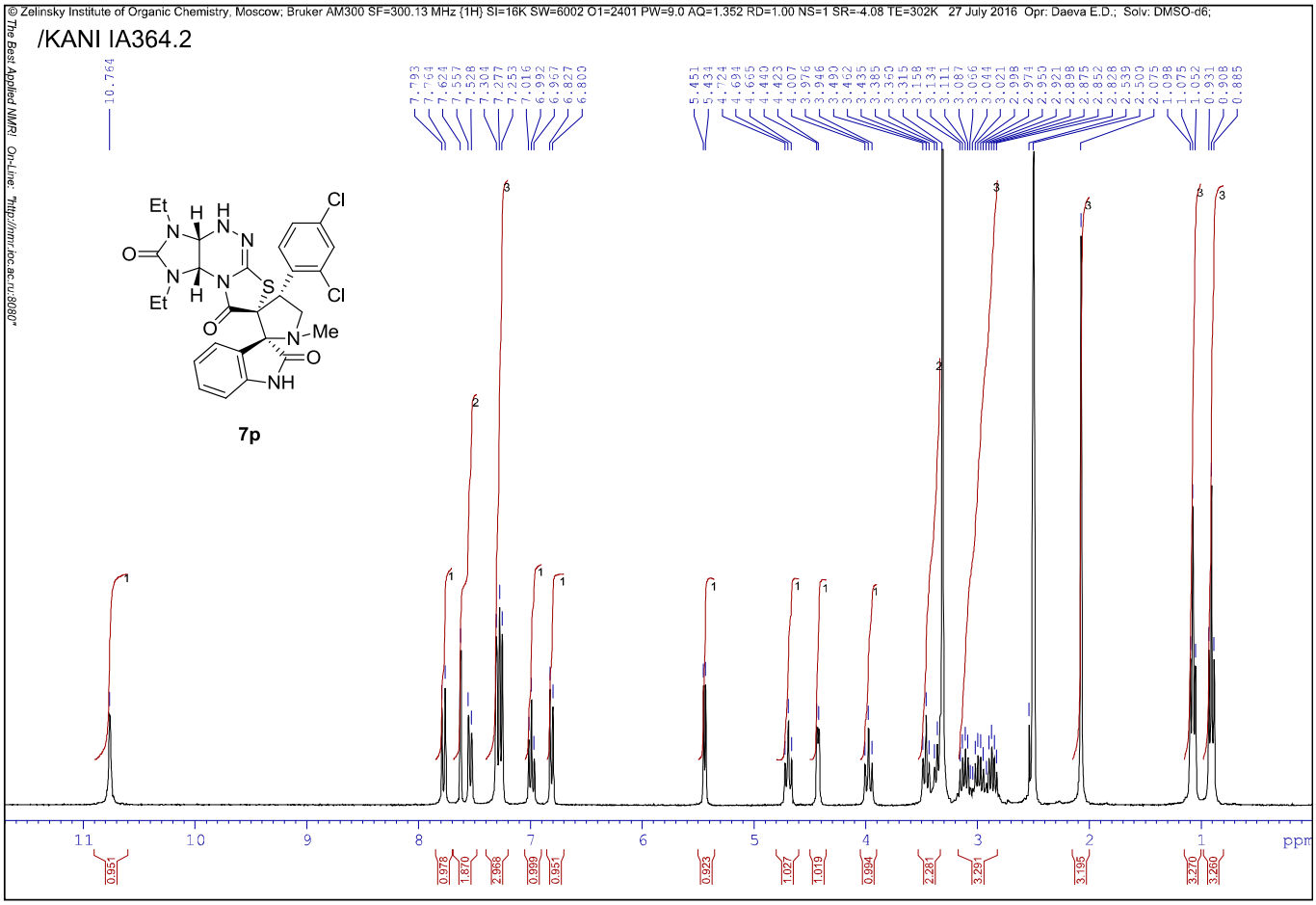
^{1}H NMR spectrum of **7o**



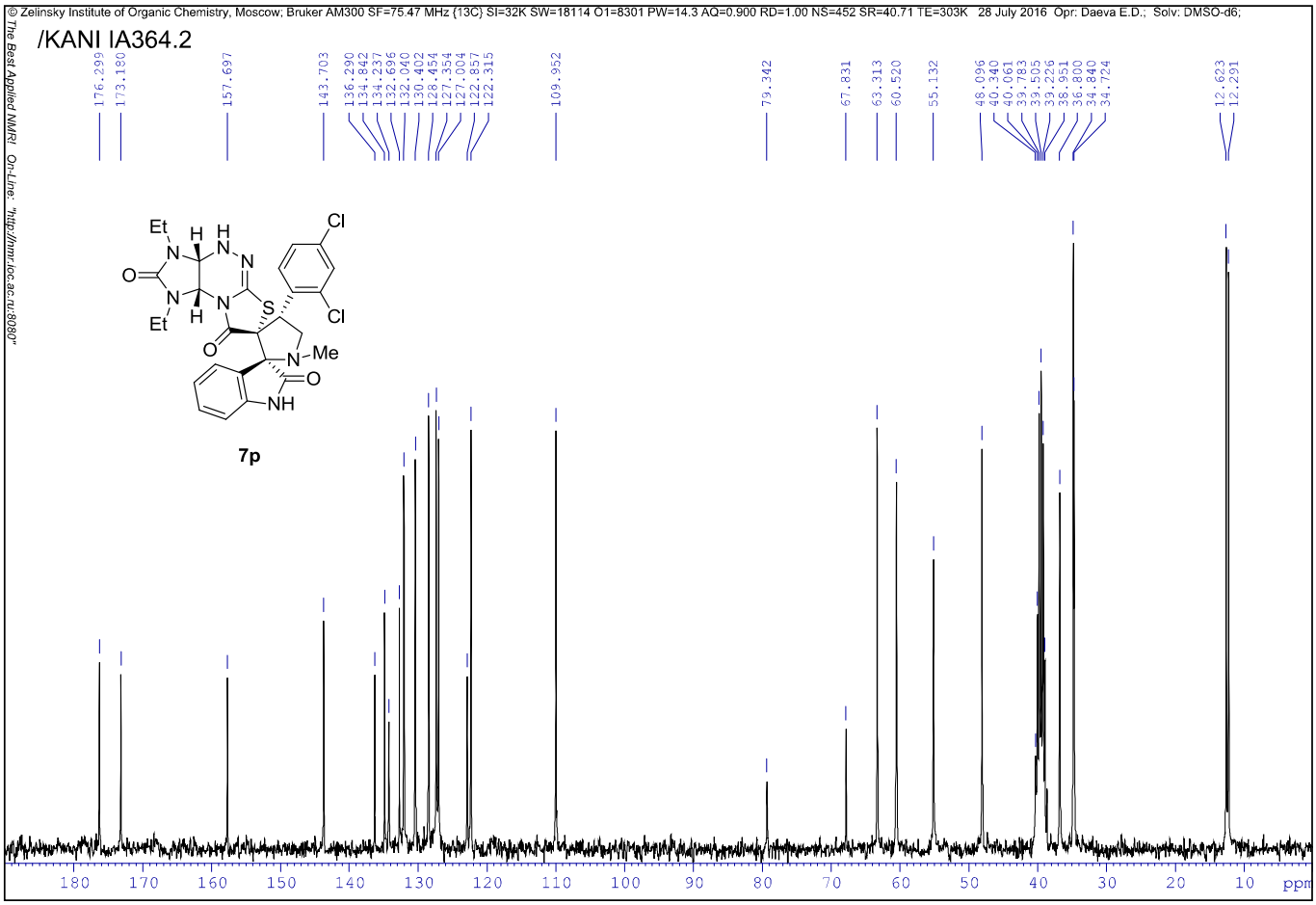
^{13}C NMR spectrum of **7o**



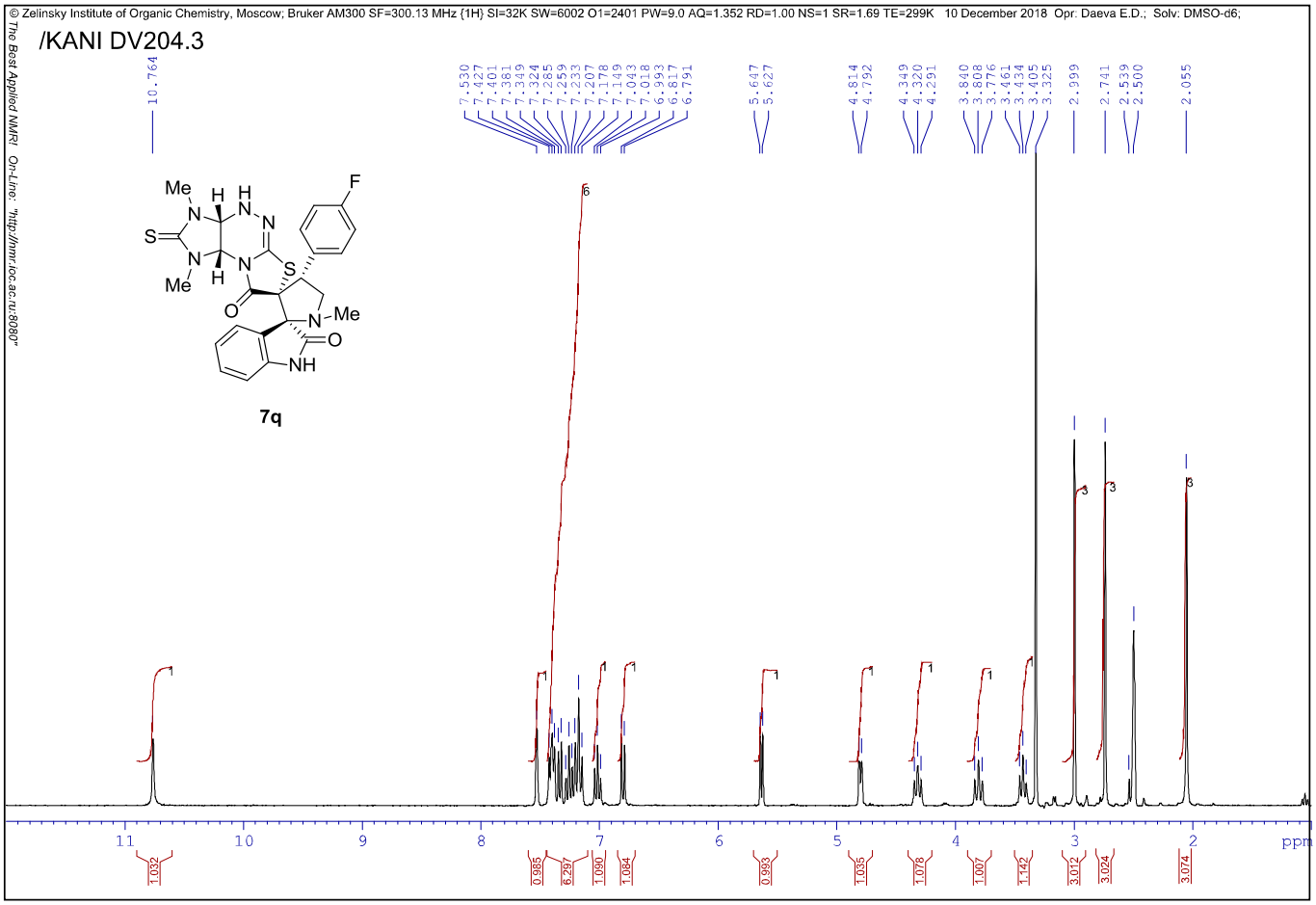
^{1}H NMR spectrum of **7p**



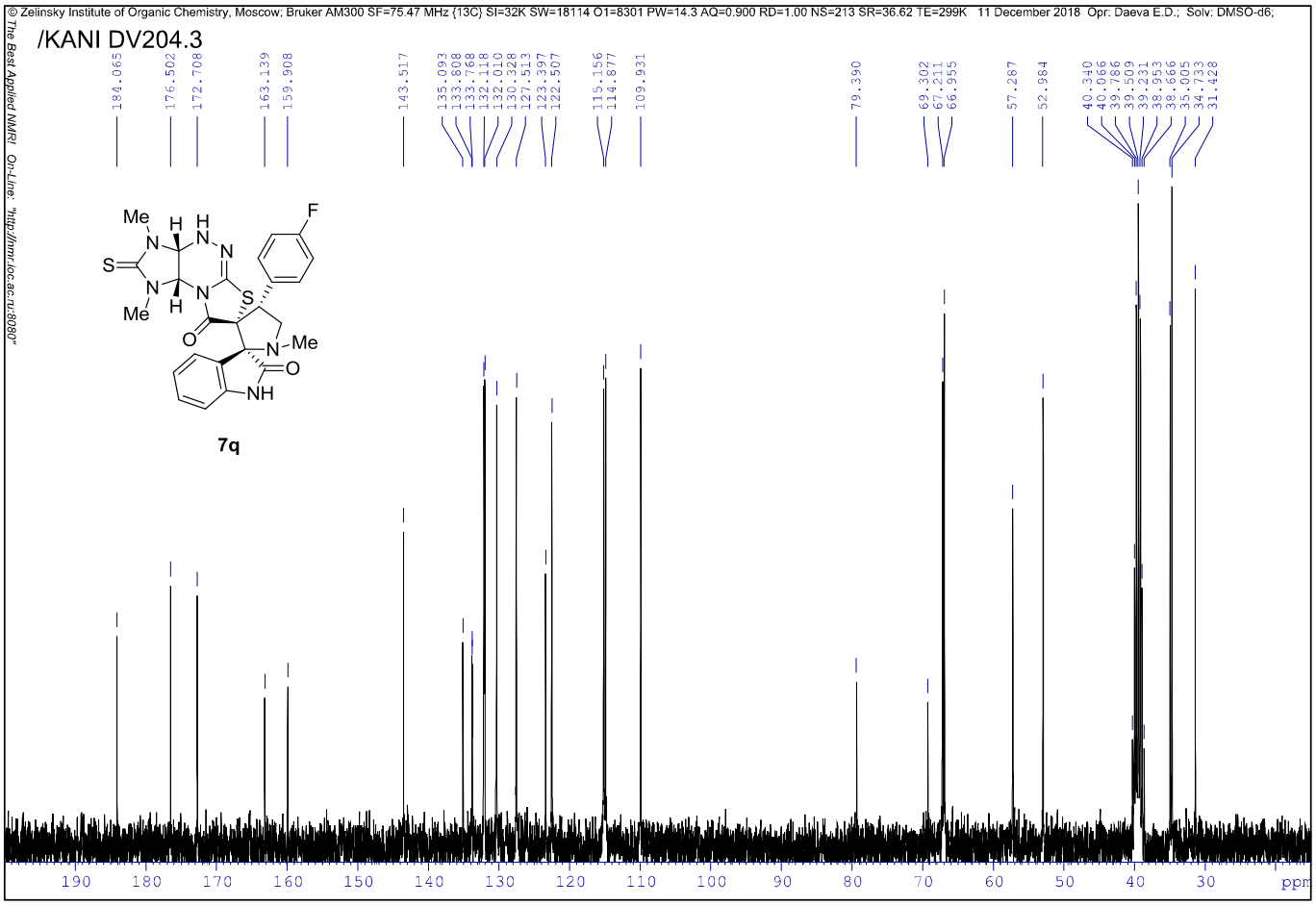
^{13}C NMR spectrum of **7p**



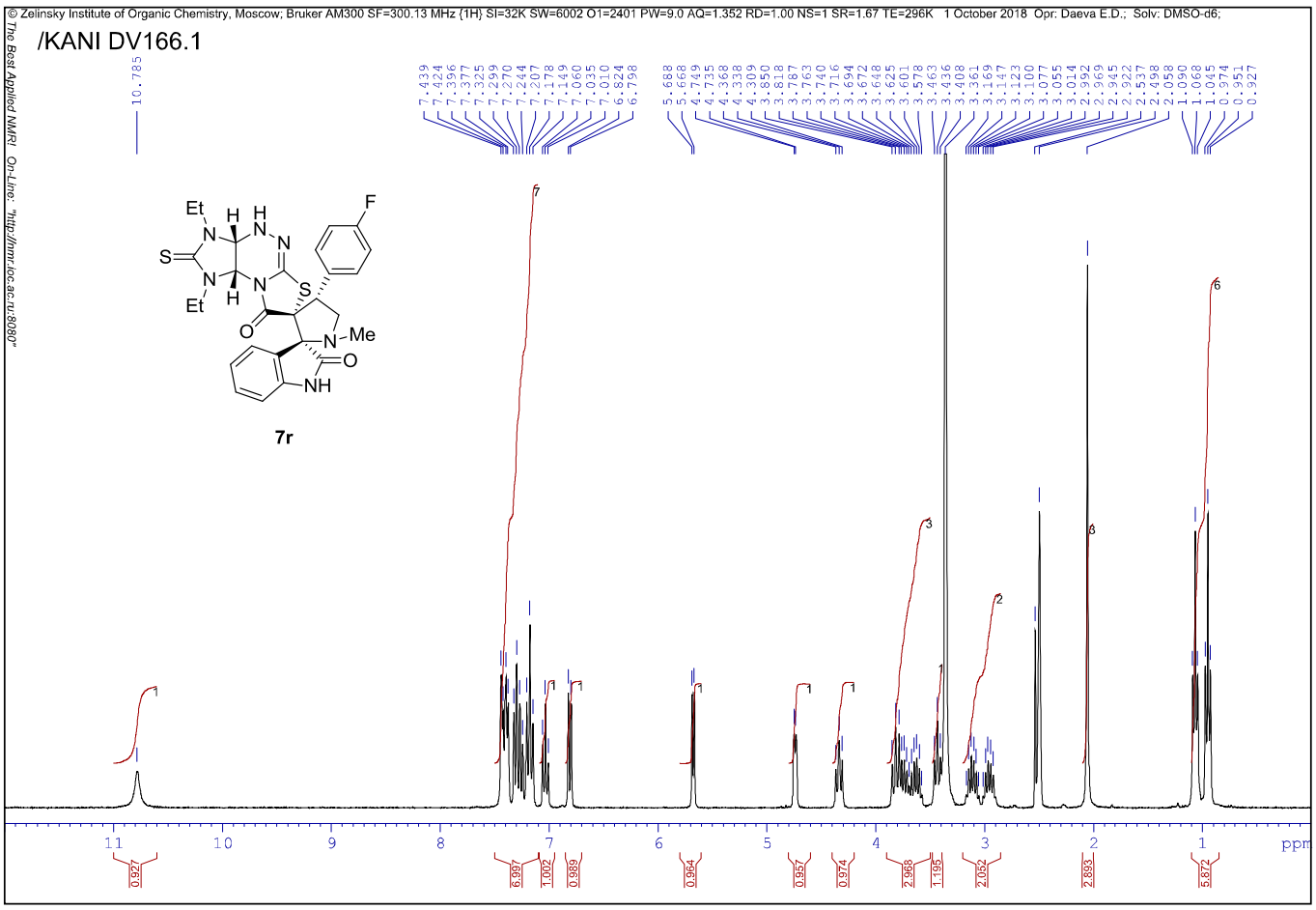
^{1}H NMR spectrum of **7q**



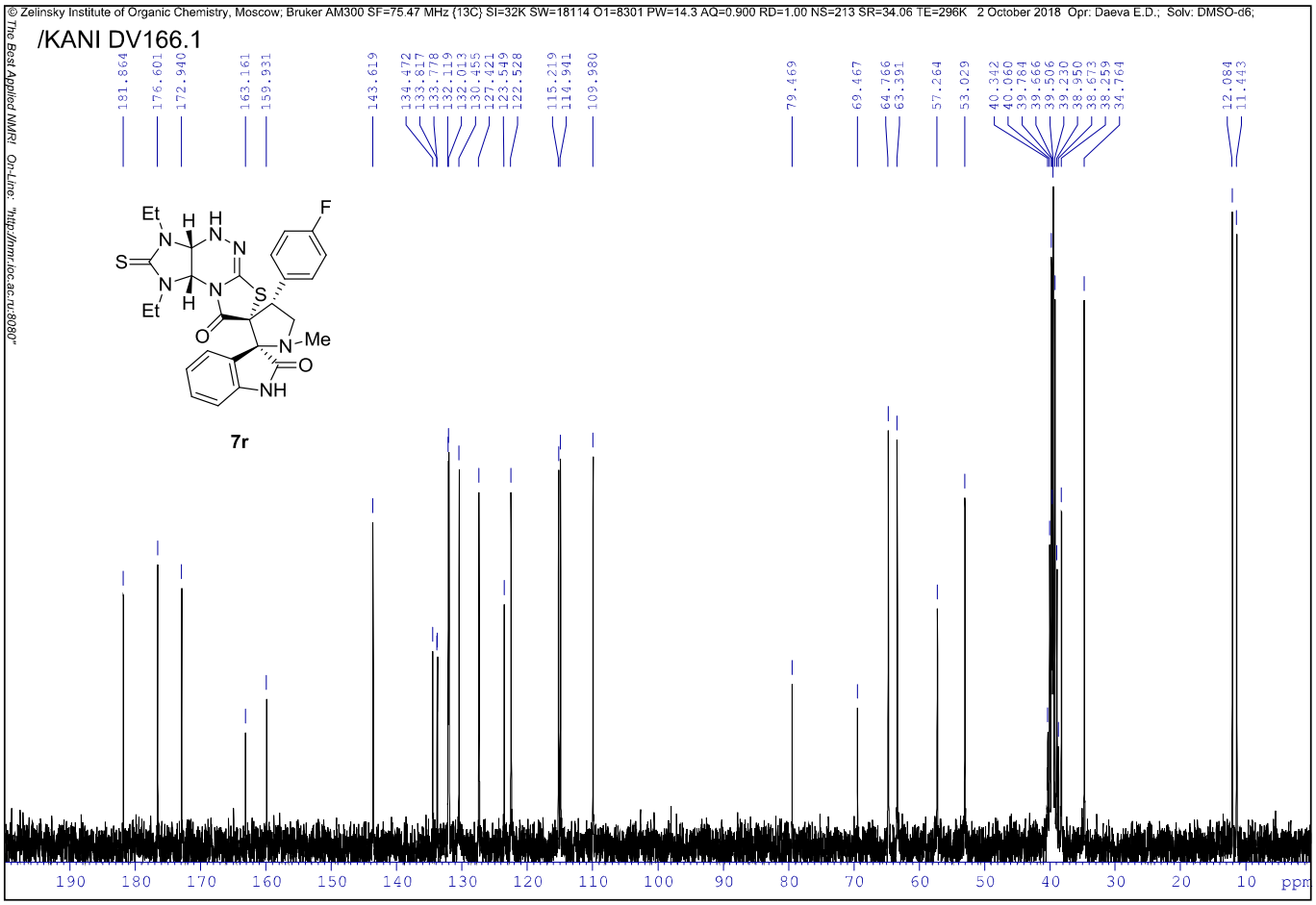
^{13}C NMR spectrum of **7q**



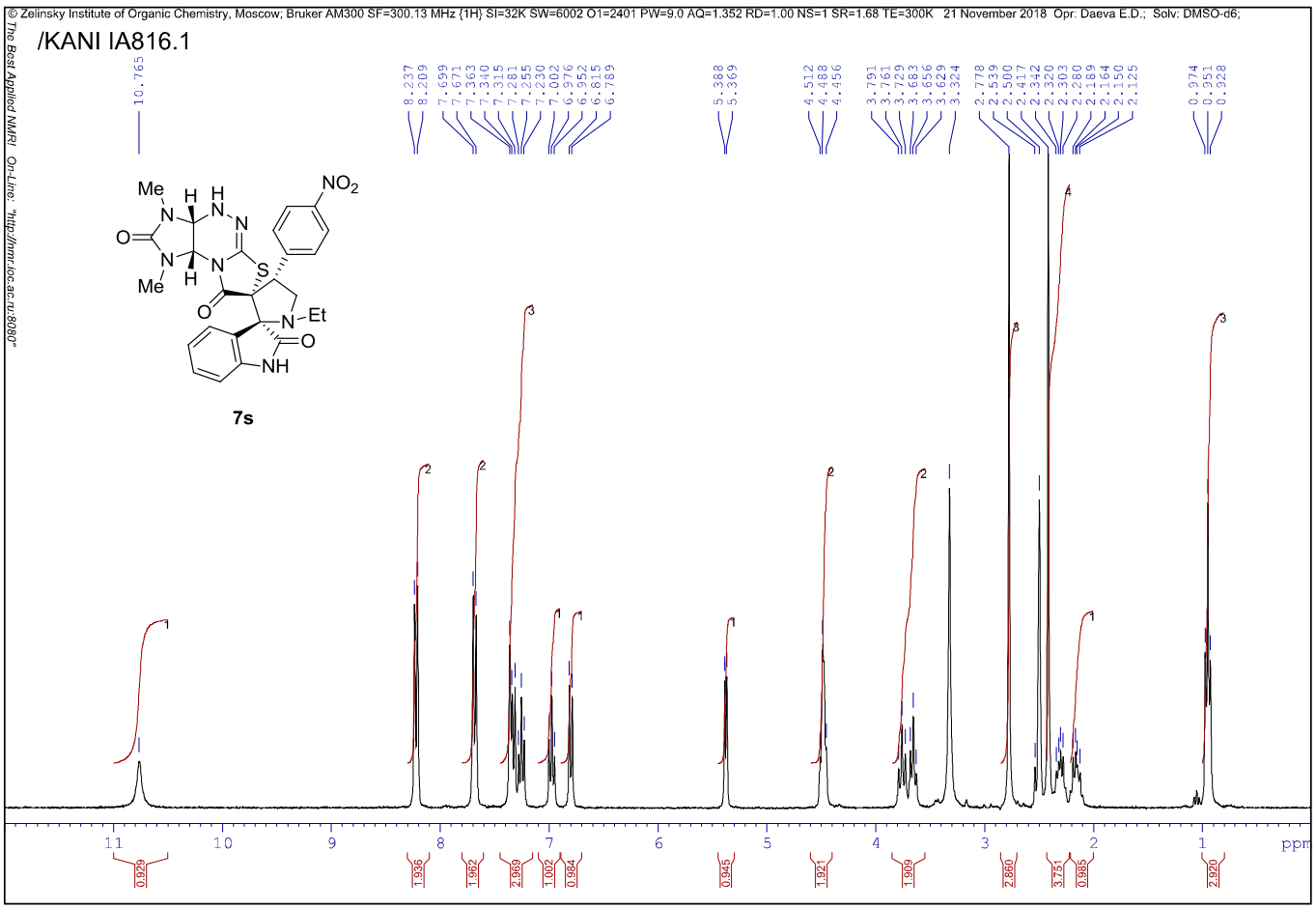
^{1}H NMR spectrum of **7r**



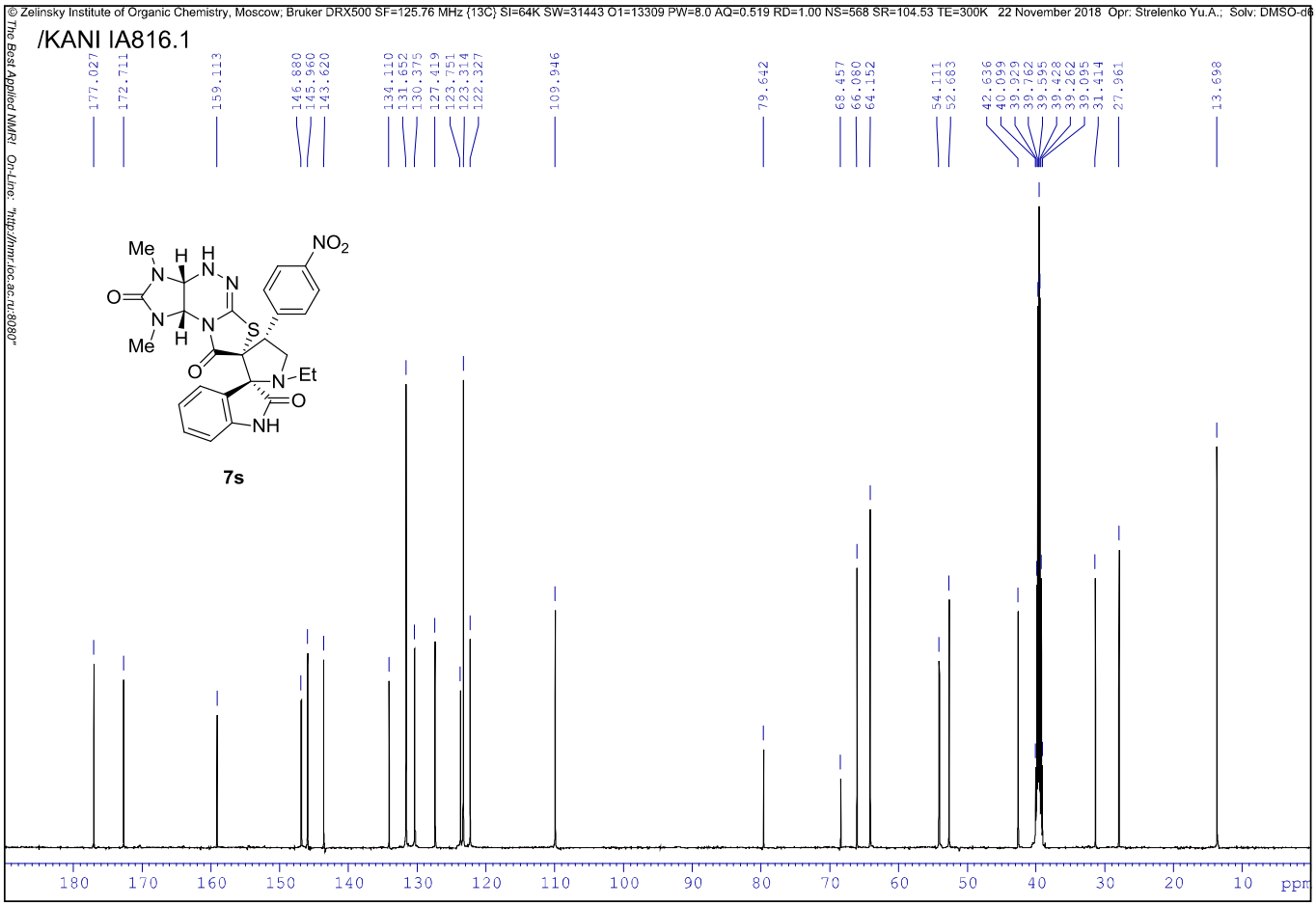
^{13}C NMR spectrum of **7r**



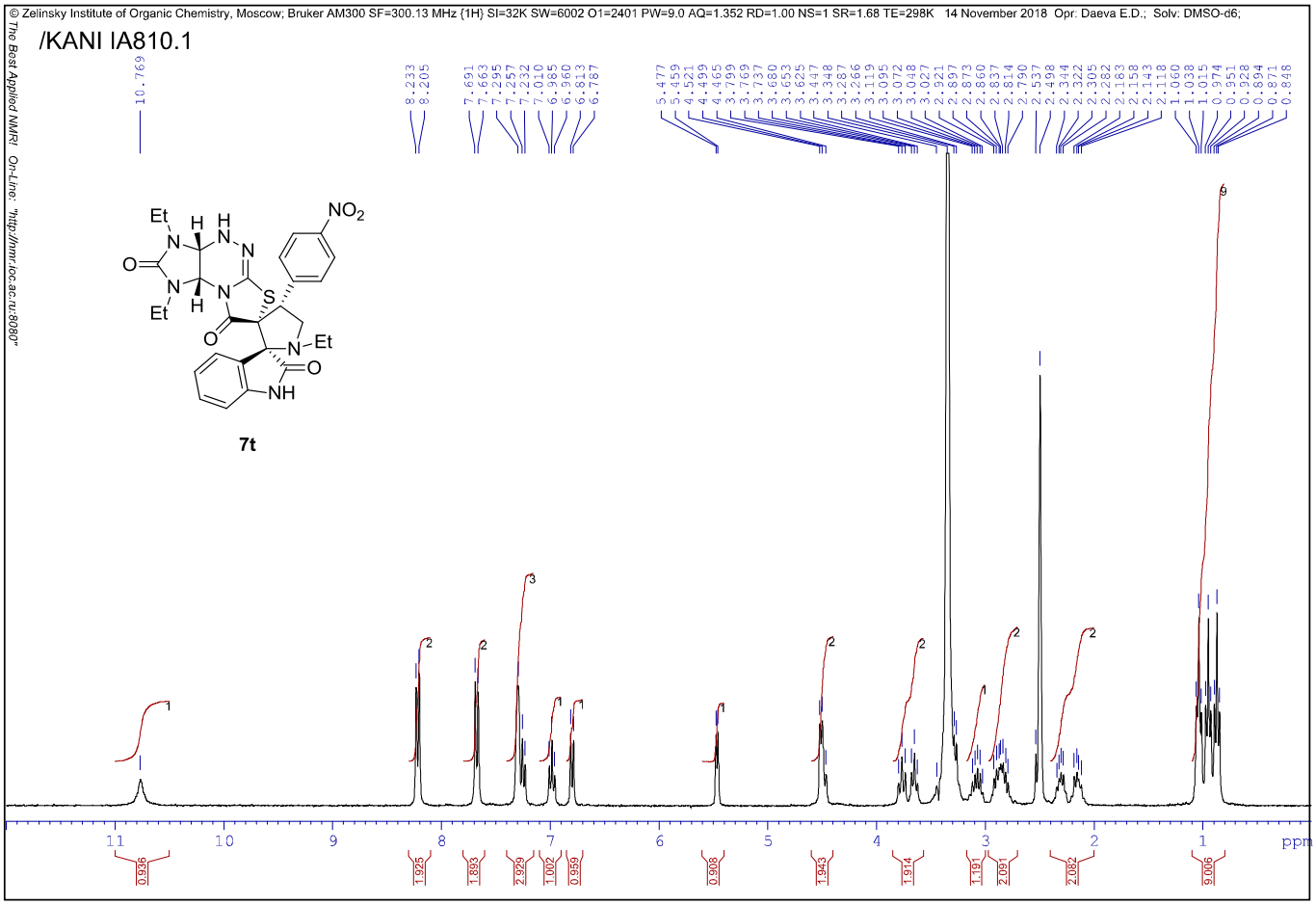
^{1}H NMR spectrum of **7s**



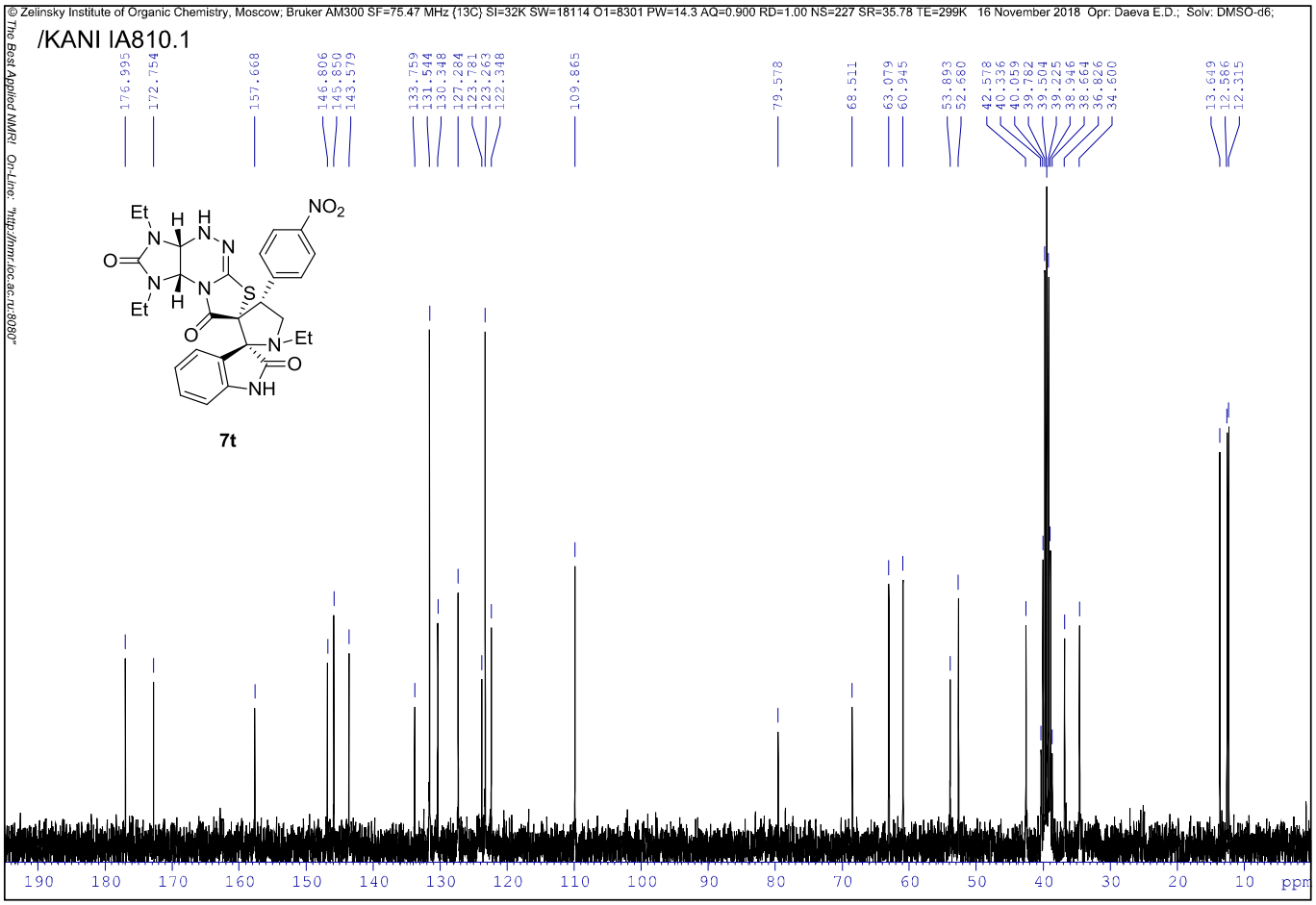
^{13}C NMR spectrum of **7s**



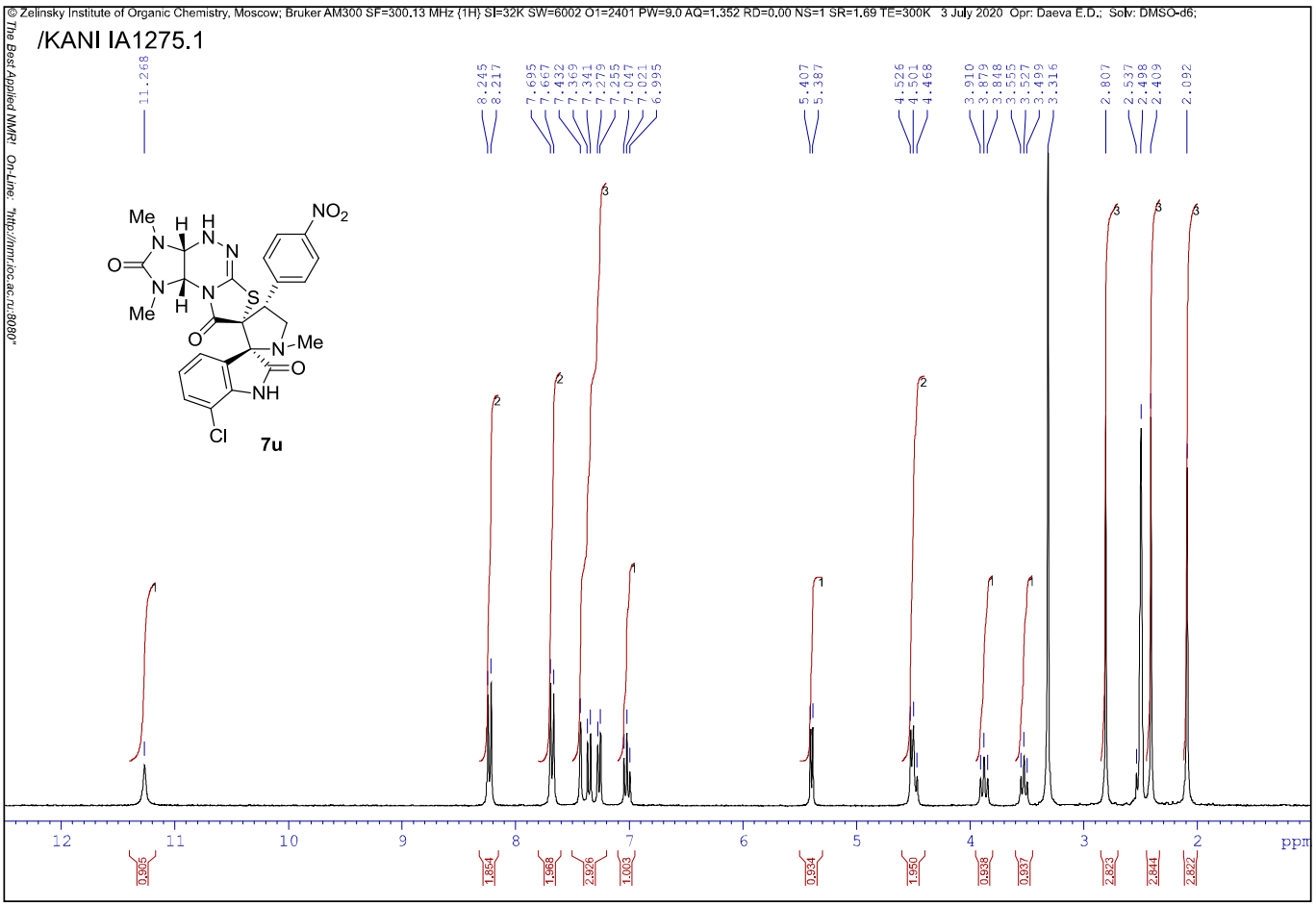
^{1}H NMR spectrum of **7t**



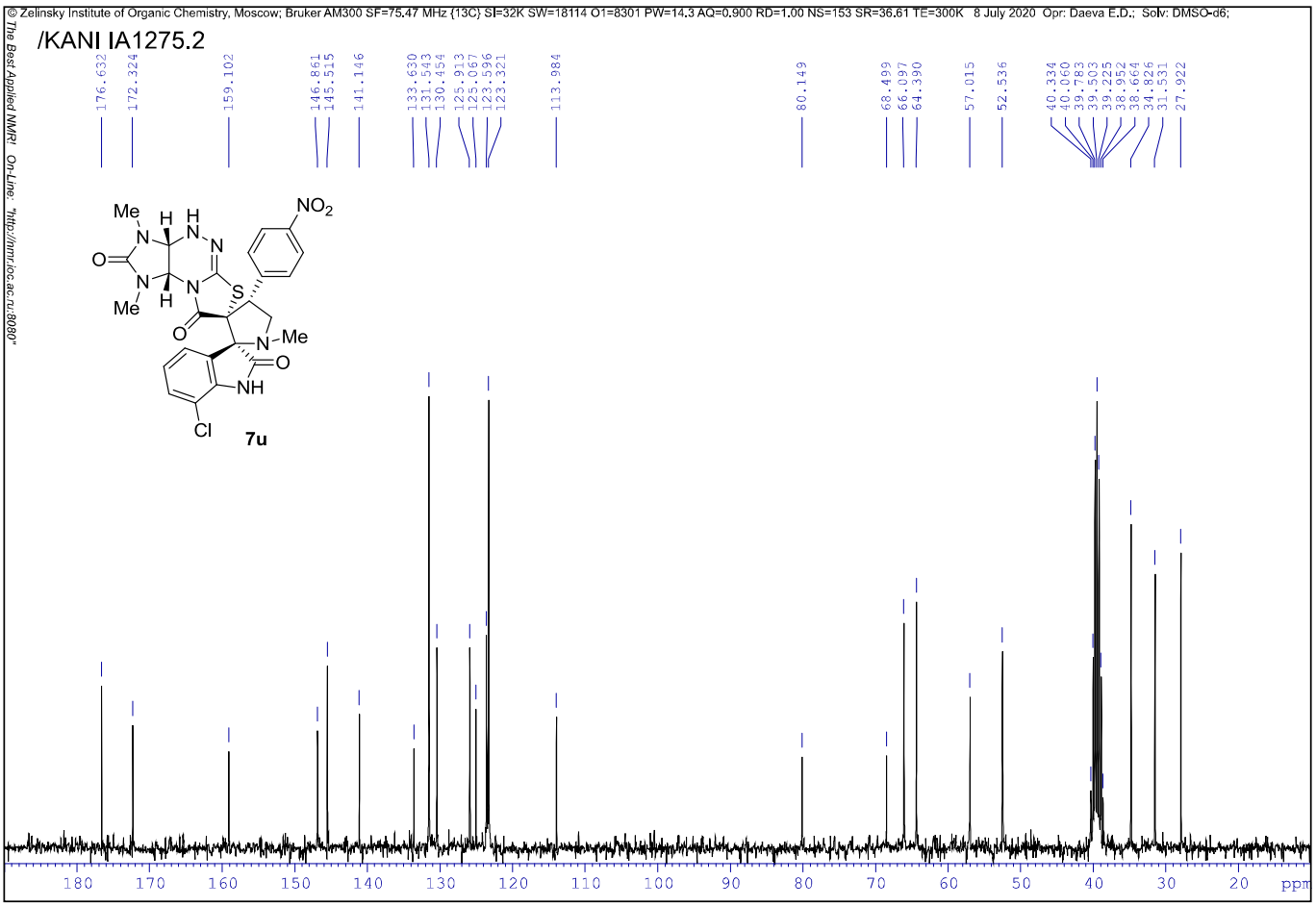
^{13}C NMR spectrum of **7t**



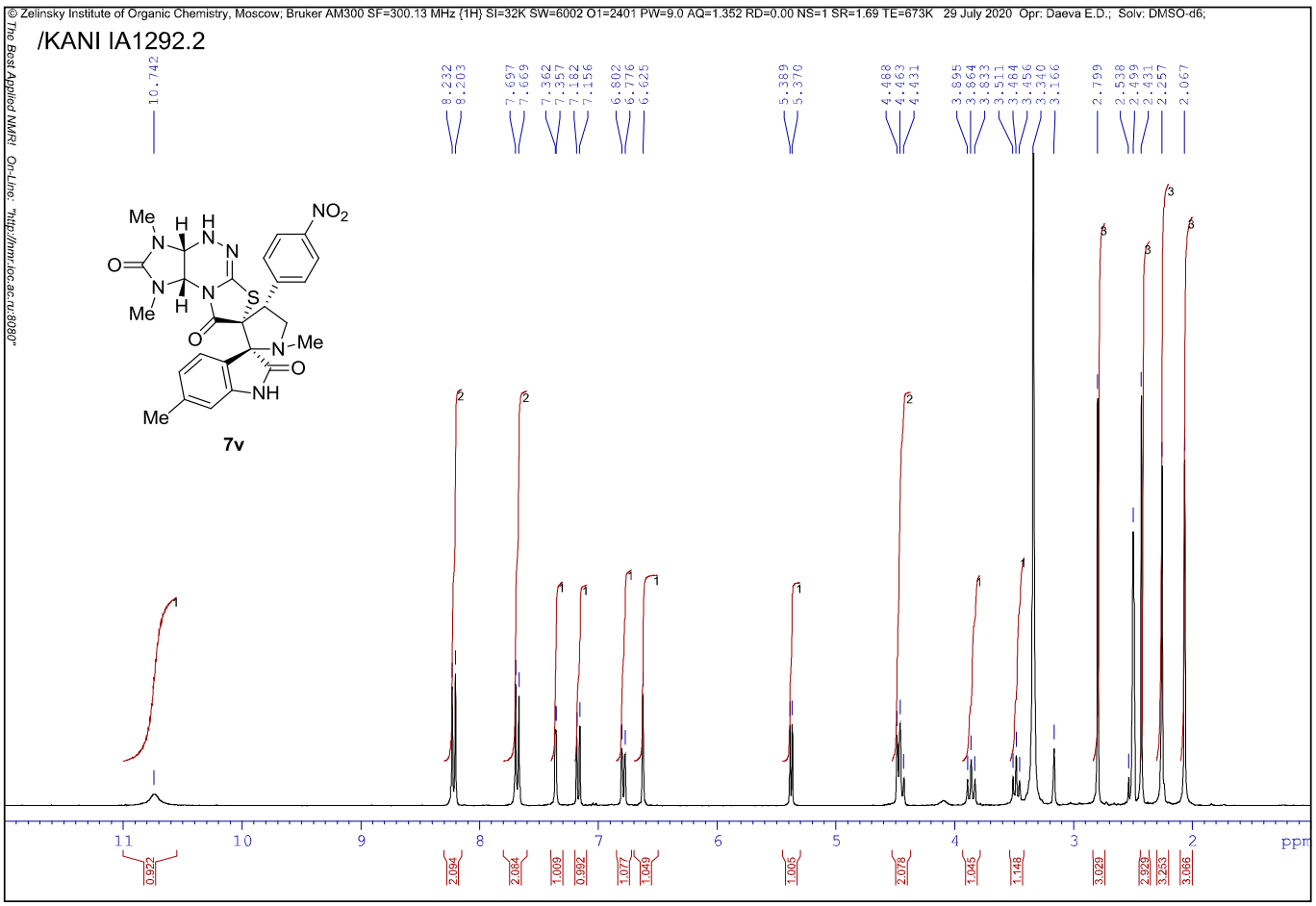
^{1}H NMR spectrum of **7u**



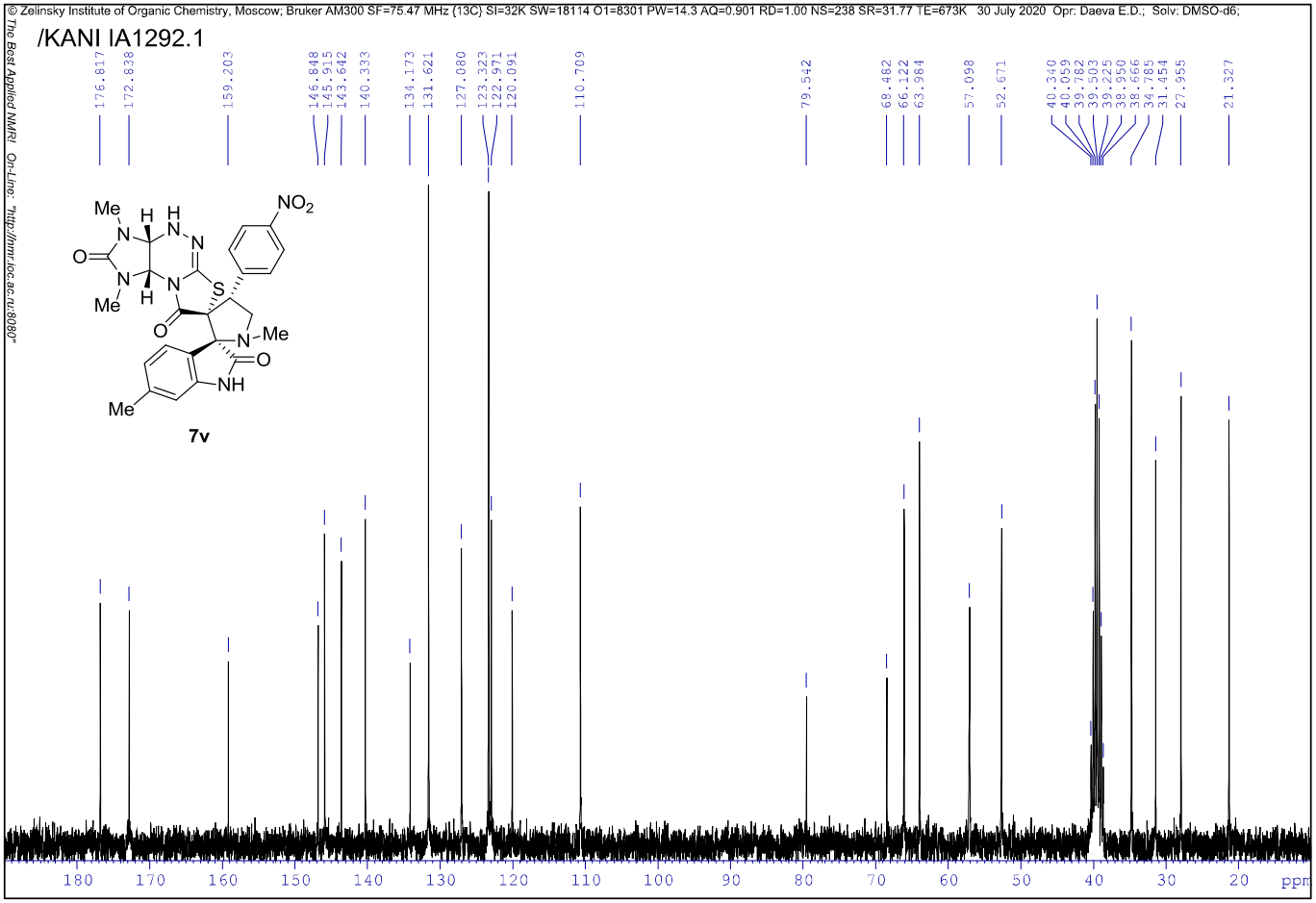
^{13}C NMR spectrum of **7u**



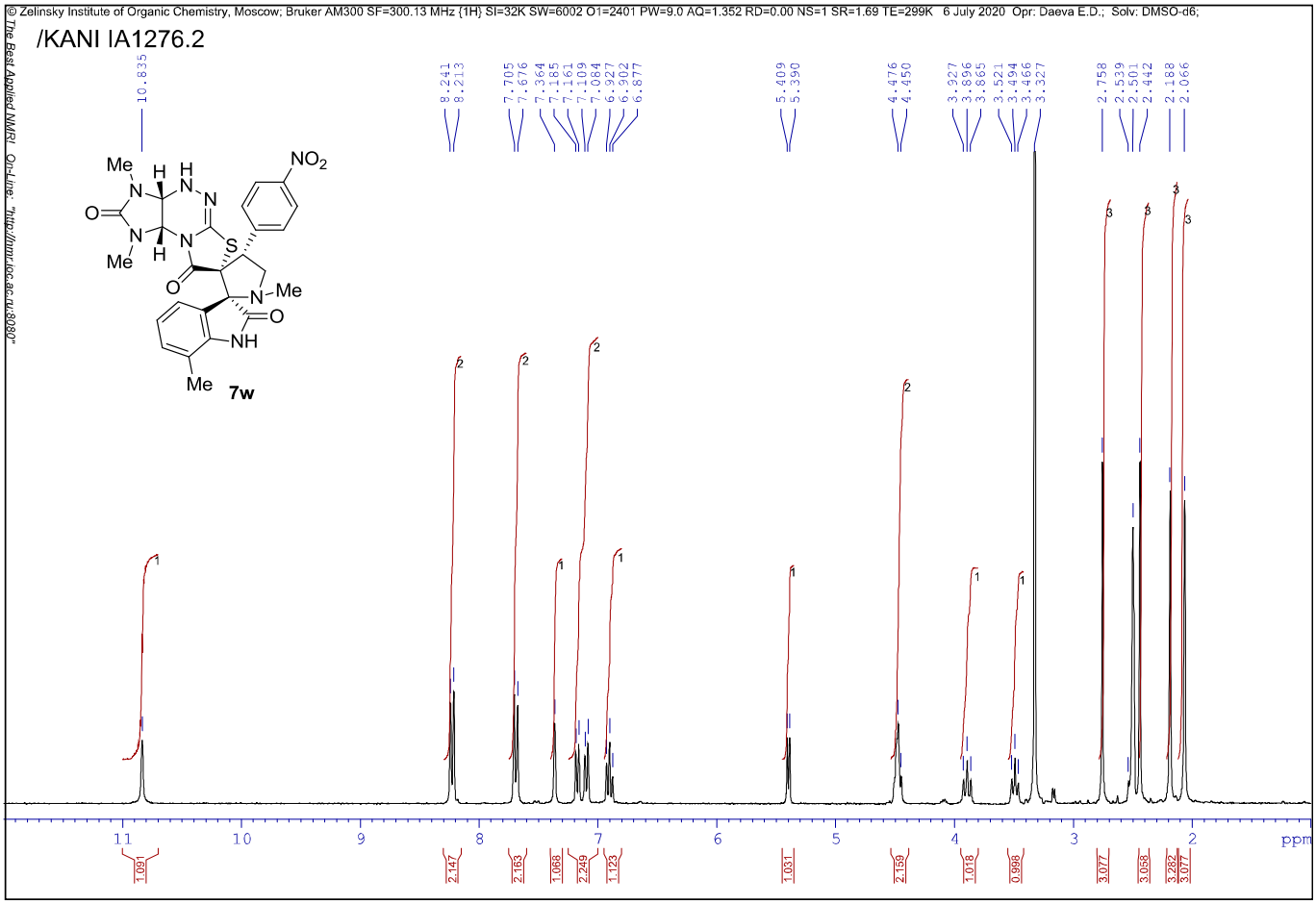
^{1}H NMR spectrum of **7v**



^{13}C NMR spectrum of **7v**



^{1}H NMR spectrum of **7w**



^{13}C NMR spectrum of **7w**