Supplementary Information

Solvent-free and Efficient Synthesis of Imidazo[1,2-*a*]pyridine Derivatives via a One-pot Three-component Reaction

Li-Rong Wen,* Zhao-Rui Li, Ming Li,* and Han Cao

State Key Laboratory Base of Eco-Chemical Engineering, College of Chemistry and Molecular Engineering, Qingdao University of Science and Technology, Qingdao 266042, P. R. China

Contents

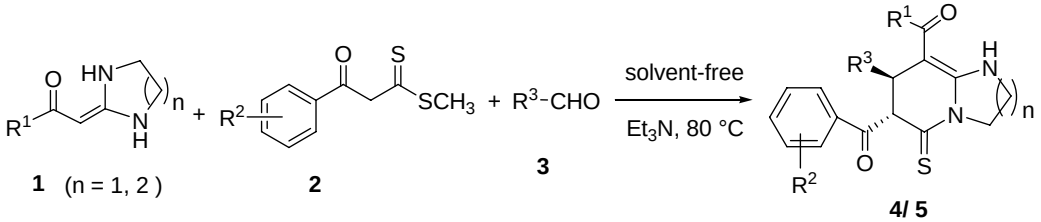
General Information

All reagents and solvents were obtained from commercial suppliers and used without further purification. All reagents were weighed and handled in air at room temperature. Melting points were recorded on a RY-1 microscopic melting apparatus and uncorrected. ^{1}H NMR spectra were recorded on 500 MHz and ^{13}C NMR spectra were recorded on 125 MHz by using a Bruker Avance 500M spectrometer. Chemical shifts were reported in parts per million (δ) relative to tetramethylsilane (TMS). IR spectra were recorded on a Nicolet iS10 FT-IR spectrometer and only major peaks are reported in cm^{-1}. Mass spectra were performed on an Ultima Global spectrometer with an ESI source. The X-ray single-crystal diffraction was performed on Saturn 724+ instrument.

The raw materials **1a-j** were synthesized according to the literatures.[1~5]

The raw materials **2a-f** were synthesized according to the literatures.[6~7]

General procedure for the preparation of hexahydroimidazo[1,2-*a*]pyridines 4 and hexahydropyrido[1,2-*a*]pyridines 5 via one-pot three-component reactions



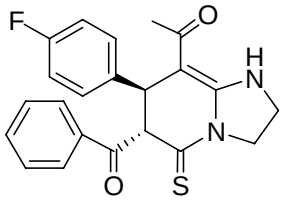
1 (n = 1, 2) **2** **3** **4/ 5**

General procedure for the preparation of hexahydroimidazo[1,2-*a*]pyridines **4** and hexahydropyrido[1,2-*a*] pyrimidins **5** via one-pot three-component reactions HKAs **1** (0.5 mmol), *β*-oxodithioesters **2** (0.5 mmol) and aldehydes **3** (0.5 mmol) were placed into a 25 mL flask, and

the mixture was stirred for the appropriate time at 80 °C in oil bath until the HKAs **1** were completely consumed. The mixture was washed with EtOH (10 mL) three times and vacuum filtration. The crude products so obtained were recrystallized from EtOH to afford the pure products **4/5**.

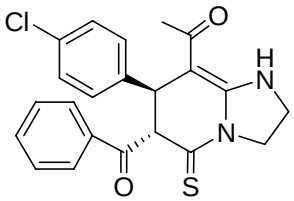
Characterization data

1-((6*S*,7*S*)-6-Benzoyl-7-(4-fluorophenyl)-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-8-yl)ethanone (4a)



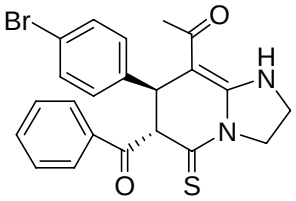
Primrose yellow solid; Mp 227–228 °C; **IR** (KBr): 1678, 1654, 1424, 1196 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.48 (s, 1H, NH), 7.01–8.04 (m, 9H, ArH), 5.29 (d, J = 1.0 Hz, 1H, CHCO), 4.49–4.54 (m, 1H, NCH_2), 4.22–4.28 (m, 1H, NCH_2), 4.12 (s, 1H, ArCH), 3.93–4.00 (m, 2H, $NHCH_2$), 1.75 (s, 3H, CH_3); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.5, 194.0, 193.2, 163.0, 161.1, 151.9, 138.3, 134.5, 133.9, 129.2, 128.8, 128.2, 128.1, 116.3, 116.1, 87.4, 66.2, 48.3, 42.0, 26.2; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{22}H_{20}N_2O_2FS$ [$(M + H)^+$], 395.1230; found, 395.1239.

1-((6*S*,7*S*)-6-Benzoyl-7-(4-chlorophenyl)-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*] pyridin-8-yl)ethanone (4b)



Brown yellow solid; Mp 234–235 °C; **IR** (KBr): 1679, 1653, 1424, 1197 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.49 (s, 1H, NH), 7.07–8.03 (m, 9H, ArH), 5.28 (d, J = 1.0 Hz, 1H, CHCO), 4.48–4.53 (m, 1H, NCH_2), 4.21–4.27 (m, 1H, NCH_2), 4.10 (s, 1H, ArCH), 3.90–3.99 (m, 2H, $NHCH_2$), 1.74 (s, 3H, CH_3); **^{13}C NMR** (125 MHz,$CDCl_3$): δ = 194.3, 193.9, 193.1, 152.0, 141.1, 134.6, 133.9, 133.4, 129.4, 129.2, 128.8, 128.0, 87.1, 65.9, 48.3, 42.1, 42.0, 26.2; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{22}H_{20}N_2O_2SCl$, [$(M + H)^+$], 411.0934; found, 411.0942.

1-((6*S*,7*S*)-6-Benzoyl-7-(4-bromophenyl)-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*] pyridin-8-yl)ethanone (4c)

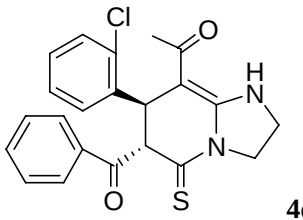


Yellow solid; Mp 229–230 °C; **IR** (KBr): 1679, 1654, 1424, 1197 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.49 (s, 1H, NH), 7.01–8.03 (m, 9H, ArH), 5.28 (d, J = 1.0 Hz, 1H, CHCO), 4.48–4.53 (m, 1H, NCH_2), 4.21–4.27 (m, 1H, NCH_2), 4.09 (s, 1H, ArCH), 3.92–4.00 (m, 2H,

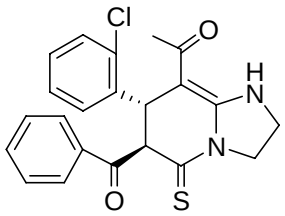
$NHCH_2$), 1.74 (s, 3H, CH_3); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.4, 193.9, 193.1, 152.0, 141.6, 134.5, 133.9, 132.4, 129.2, 128.8, 128.4, 121.4, 87.0, 65.8, 48.3, 42.2, 42.0, 26.2; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{22}H_{20}N_2O_2SBr$ [$(M + H)^+$], 455.0429; found, 455.0419.

1-((6*S*,7*R*)-6-Benzoyl-7-(2-chlorophenyl)-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*] pyridin-8-yl)ethanone (4d)

and 1-((6*R*,7*S*)-6-Benzoyl-7-(2-chlorophenyl)-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*] pyridin-8-yl)ethanone (4d')



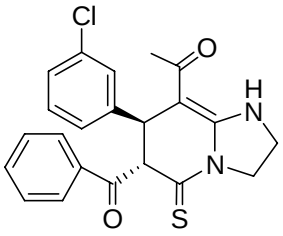
4d



4d'

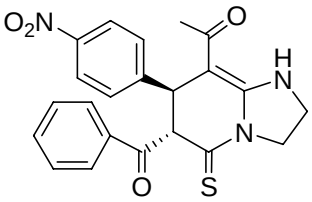
Brown yellow solid; Mp 200–201 °C; **IR** (KBr): 1677, 1654, 1425, 1201 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.51 (s, 1H, NH, **4d**), 9.42 (s, 1H, NH, **4d'**), 6.85–8.10 (m, 2ArH, **4d/4d'**), 5.26 (d, *J* = 1.0 Hz, 1H, CHCO, **4d**), 4.61 (d, *J* = 1.0 Hz, 1H, CHCO, **4d'**), 4.54–4.56 (m, 1H, NCH_2, **4d**), 4.44–4.50 (m, 1H, NCH_2, **4d'**), 4.34–4.39 (m, 1H, NCH_2, **4d**), 4.22–4.28 (m, 1H, NCH_2, **4d'**), 4.11–4.18 (m, 2ArCH, **4d/4d'**), 3.93–4.01 (m, 2H, $NHCH_2$, **4d**), 3.81–3.92 (m, 2H, $NHCH_2$, **4d'**), 1.90 (s, 3H, CH_3, **4d**), 1.75 (s, 3H, CH_3, **4d'**); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 197.5, 195.4, 194.7, 194.5, 193.0, 152.5, 152.0, 140.0, 139.5, 134.9, 133.8, 133.0, 132.3, 130.1, 129.4, 128.9, 128.7, 128.5, 128.4, 127.7, 127.6, 127.3, 89.2, 87.5, 63.7, 48.5, 47.9, 47.3, 42.1, 41.8, 38.6, 34.5, 26.2, 18.4; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{22}H_{20}N_2O_2ClS$ [$(M + H)^+$], 411.0934; found, 411.0938.

1-((6*S*,7*S*)-6-Benzoyl-7-(3-chlorophenyl)-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*] pyridin-8-yl)ethanone (**4e**)



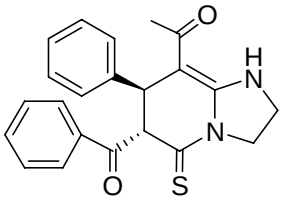
Primrose yellow solid; Mp 238–239 °C; **IR** (KBr): 1685, 1654, 1423, 1193 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.50 (s, 1H, NH), 7.03–8.04 (m, 9H, ArH), 5.30 (d, *J* = 1.0 Hz, 1H, CHCO), 4.48–4.54 (m, 1H, NCH_2), 4.22–4.28 (m, 1H, NCH_2), 4.11 (s, 1H, ArCH), 3.93–3.98 (m, 2H, $NHCH_2$), 1.75 (s, 3H, CH_3); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.3, 194.0, 193.0, 152.1, 144.6, 135.0, 134.5, 133.9, 130.6, 129.2, 128.8, 127.9, 126.8, 124.9, 86.7, 65.7, 48.3, 42.3, 42.0, 26.3; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{22}H_{20}N_2O_2SCl$ [$(M + H)^+$], 411.0934; found, 411.0942.

1-((6*S*,7*S*)-6-Benzoyl-7-(4-nitrophenyl)-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-8-yl)ethanone (4f)



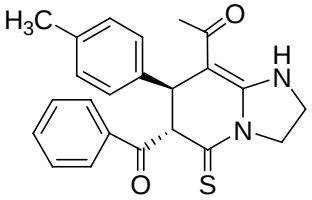
Brown yellow solid; Mp 210–211 °C; **IR** (KBr): 1683, 1654, 1428, 1199 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.55 (s, 1H, NH), 7.26–8.23 (m, 9H, ArH), 5.27 (d, J = 1.5 Hz, 1H, CHCO), 4.49–4.55 (m, 1H, NCH_2), 4.25–4.29 (m, 1H, NCH_2), 4.23 (s, 1H, ArCH), 3.97–4.01 (m, 2H, $NHCH_2$) 1.74 (s, 3H, CH_3); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 193.9, 193.6, 192.5, 152.3, 149.9, 147.5, 134.5, 134.1, 129.3, 128.8, 127.7, 124.6, 86.4, 65.3, 48.5, 42.6, 42.1, 26.2; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{22}H_{20}N_3O_4S$ [$(M + H)^+$], 422.1175; found, 422.1185.

1-((6*S*,7*S*)-6-Benzoyl-7-phenyl-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-8-yl) ethanone (4g)



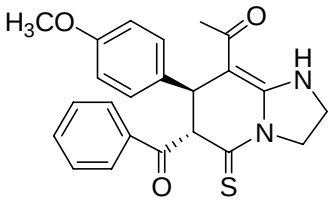
Yellow solid; Mp 237–238 °C; **IR** (KBr): 1686, 1652, 1421, 1199 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.48 (s, 1H, NH), 7.13–8.06 (m, 10H, ArH), 5.33 (d, J = 1.5 Hz, 1H, CHCO), 4.49–4.55 (m, 1H, NCH_2), 4.22–4.28 (m, 1H, NCH_2), 4.13 (s, 1H, ArCH), 3.93–3.98 (m, 2H, $NHCH_2$), 1.75 (s, 3H, CH_3); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.7, 194.2, 193.5, 151.9, 142.5, 134.5, 133.8, 129.3, 129.1, 128.8, 127.6, 126.6, 87.5, 66.0, 48.3, 42.7, 42.0, 26.3; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{22}H_{21}N_2O_2S$ [$(M + H)^+$], 377.1324; found, 377.1315.

1-((6*S*,7*S*)-6-Benzoyl-5-thioxo-7-p-tolyl-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-8-yl) ethanone (4h)



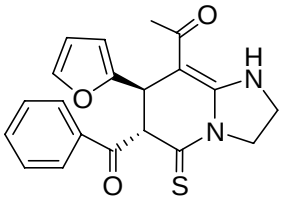
Brown yellow solid; Mp 235–236 °C; **IR** (KBr): 1678, 1653, 1421, 1198 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.47 (s, 1H, NH), 7.01–8.05 (m, 9H, ArH), 5.32 (d, J = 1.0 Hz, 1H, CHCO), 4.49–4.55 (m, 1H, NCH_2), 4.21–4.27 (m, 1H, NCH_2), 4.10 (s, 1H, ArCH), 3.92–3.99 (m, 2H, $NHCH_2$), 2.33 (s, 3H, $ArCH_3$) 1.76 (s, 3H, CH_3); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.6, 194.3, 193.6, 151.9, 139.6, 137.2, 134.7, 133.7, 129.9, 129.1, 128.8, 126.5, 87.7, 66.3, 48.3, 42.4, 42.0, 26.2, 21.0; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{23}H_{23}N_2O_2S$ [$(M + H)^+$], 391.1480; found, 391.1495.

1-((6*S*,7*S*)-6-Benzoyl-7-(4-methoxyphenyl)-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*] pyridin-8-yl)ethanone (4i)



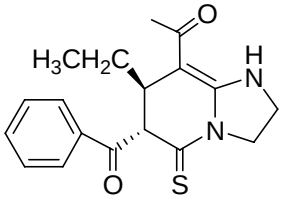
Primrose yellow solid; Mp 226–227 °C; **IR** (KBr): 1683, 1652, 1420, 1194 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.46 (s, 1H, NH), 6.85–8.05 (m, 9H, ArH), 5.32 (d, J = 1.5 Hz, 1H, CHCO), 4.49–4.53 (m, 1H, NCH_2), 4.21–4.27 (m, 1H, NCH_2), 4.09 (s, 1H, ArCH), 3.92–3.97 (m, 2H, $NHCH_2$), 3.79 (s, 3H, OCH_3), 1.76 (s, 3H, CH_3); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.6, 194.2, 193.6, 158.9, 151.9, 134.7, 134.6, 133.7, 129.1, 128.8, 127.7, 114.6, 87.8, 66.5, 55.3, 48.3, 42.0, 26.1; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{23}H_{23}N_2O_3S$ [$(M + H)^+$], 407.1429; found, 407.1438.

1-((6*S*,7*R*)-6-Benzoyl-7-(furan-2-yl)-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-8-yl)ethanone (4j)



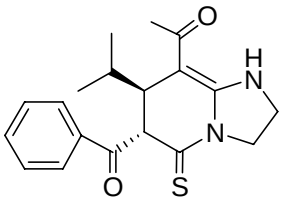
Brown yellow solid; Mp 219–220 °C; **IR** (KBr): 1682, 1652, 1425, 1195 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.46 (s, 1H, NH), 6.04–8.09 (m, 8H, ArH), 5.64 (d, J = 1.5 Hz, 1H, CHCO), 4.47–4.53 (m, 1H, NCH_2), 4.29 (s, 1H, ArCH), 4.12–4.18 (m, 1H, NCH_2), 3.85–3.94 (m, 2H, $NHCH_2$), 1.88 (s, 3H, CH_3); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.1, 194.0, 193.9, 154.5, 151.9, 142.7, 134.2, 133.9, 129.1, 128.9, 110.4, 106.4, 85.3, 62.7, 48.2, 42.0, 36.7, 25.8; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{20}H_{19}N_2O_3S$ [$(M + H)^+$], 367.1116; found, 367.1125.

1-((6*S*,7*R*)-6-Benzoyl-7-ethyl-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-8-yl) ethanone (4k)



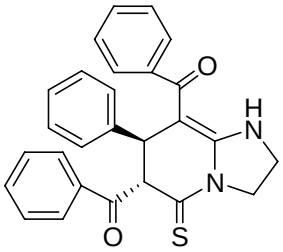
Brown yellow solid; Mp 192–193 °C; **IR** (KBr): 1686, 1652, 1424, 1194 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.34 (s, 1H, NH), 7.48–7.92 (m, 5H, ArH), 5.21 (d, J = 1.0 Hz, 1H, CHCO), 4.50–4.51 (m, 1H, NCH_2), 4.13–4.15 (m, 1H, NCH_2), 3.83–3.88 (m, 2H, $NHCH_2$), 2.87–2.90 (m, 1H, $CHCH_2CH_3$), 1.84 (s, 3H, CH_3), 1.55–1.62 (m, 2H, $CHCH_2CH_3$), 1.06 (t, J = 7.5 Hz, 3H, $CHCH_2CH_3$); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 195.4, 194.9, 193.7, 151.5, 134.9, 133.5, 129.0, 128.5, 89.1, 63.0, 48.1, 41.9, 38.9, 29.2, 25.6, 11.2; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{18}H_{21}N_2O_2S$ [$(M + H)^+$], 329.1324; found, 329.1329.

1-((6*S*,7*R*)-6-Benzoyl-7-isopropyl-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-8-yl) ethanone (4l)



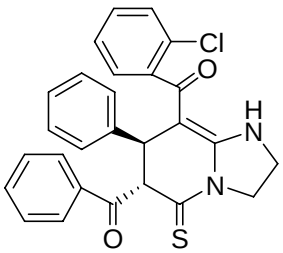
Brown yellow solid; Mp 185–186 °C; **IR** (KBr): 1682, 1655, 1429, 1196 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.46 (s, 1H, NH), 7.49–7.93 (m, 5H, ArH), 5.28 (s, 1H, CHCO), 4.52–4.57 (m, 1H, NCH_2), 4.07–4.13 (m, 1H, NCH_2), 3.84–3.88 (m, 2H, $NHCH_2$), 2.76 (d, J = 6.0 Hz, 1H, *CH*$CH(CH_3)_2$), 1.81 (s, 3H, CH_3), 1.74–1.79 (m, 1H, $CHCH(CH_3)_2$), 1.06 (d, J = 7.0 Hz, 3H, *CH*$_3$$CHCH_3$), 1.00 (d, J = 6.5 Hz, 3H, CH_3CHCH_3); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 196.2, 195.3, 194.2, 151.9, 134.9, 133.5, 128.9, 128.5, 88.2, 60.7, 48.1, 43.5, 41.9, 34.3, 25.7, 20.7, 18.6; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{19}H_{23}N_2O_2S$ [$(M + H)^+$], 343.1480; found, 343.1495.

((6*S*,7*S*)-7-Phenyl-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridine-6,8-diyl)bis (phenylmethanone) (4m)



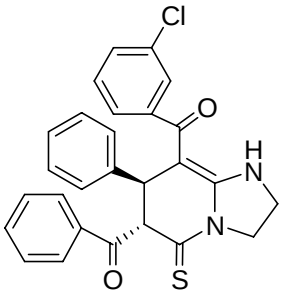
Yellow solid; Mp 243–244 °C; **IR** (KBr): 1684, 1640, 1419, 1204 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.75 (s, 1H, NH), 6.76–7.99 (m, 15H, ArH), 5.31 (d, J = 1.5 Hz, 1H, CHCO), 4.59–4.65 (m, 1H, NCH_2), 4.25–4.29 (m, 1H, NCH_2), 4.24 (s, 1H, ArCH), 3.98–4.02 (m, 2H, $NHCH_2$); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.7, 194.6, 193.2, 153.5, 142.8, 140.6, 134.8, 133.7, 129.0, 128.9, 128.7, 127.7, 127.2, 126.5, 126.0, 87.5, 66.0, 48.4, 42.3, 42.1; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{27}H_{23}N_2O_2S$ [$(M + H)^+$], 439.1480; found, 439.1466.

((6*S*,7*S*)-6-Benzoyl-7-phenyl-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-8-yl) (2-chlorophenyl)methanone (4n)



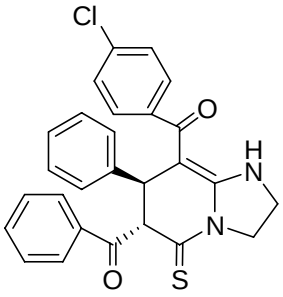
Brown yellow solid; Mp 225–226 °C; **IR** (KBr): 1681, 1648, 1417, 1208 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.72 (s, 1H, NH), 6.41–7.97 (m, 14H, ArH), 5.25 (s, 1H, CHCO), 4.58–4.63 (m, 1H, NCH_2), 4.30–4.36 (m, 1H, NCH_2), 3.99–4.06 (m, 2H, $NHCH_2$), 3.81 (s, 1H, ArCH); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.5, 190.7, 190.6, 153.4, 134.6, 133.7, 129.4, 129.0, 128.8, 128.7, 127.3, 126.5, 126.0, 88.3, 65.9, 48.4, 42.4, 42.3; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{27}H_{22}N_2O_2ClS$ [$(M + H)^+$], 473.1091; found, 473.1098.

((6*S*,7*S*)-6-Benzoyl-7-phenyl-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-8-yl)(3-chlorophenyl)methanone (4o)



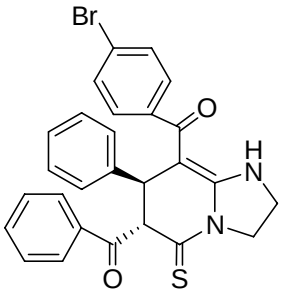
Yellow solid; Mp 257–258 °C; **IR** (KBr): 1684, 1638, 1421, 1205 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.78 (s, 1H, NH), 6.71–7.99 (m, 14H, ArH), 5.32 (d, J = 1.5 Hz, 1H, CHCO), 4.59–4.64 (m, 1H, NCH_2), 4.24–4.30 (m, 1H, NCH_2), 4.18 (d, J = 1.0 Hz, 1H, ArCH), 4.00–4.03 (m, 2H, $NHCH_2$); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.6, 194.5, 191.7, 153.8, 142.5, 138.9, 134.9, 134.6, 133.9, 129.2, 129.1, 128.6, 128.0, 127.6, 127.4, 126.4, 87.2, 66.0, 48.4, 42.4, 22.2; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{27}H_{22}N_2O_2ClS$ [$(M + H)^+$], 473.1091; found, 473.1086.

((6*S*,7*S*)-6-Benzoyl-7-phenyl-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-8-yl)(4-chlorophenyl)methanone (4p)



Yellow solid; Mp 256–257 °C; **IR** (KBr): 1684, 1638, 1422, 1205 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.78 (s, 1H, NH), 6.71–7.99 (m, 14H, ArH), 5.32 (s 1H, CHCO), 4.59–4.64 (m, 1H, NCH_2), 4.23–4.30 (m, 1H, NCH_2), 4.17 (s, 1H, ArCH), 3.99–4.05 (m, 2H, $NHCH_2$); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.6, 194.5, 191.7, 153.8, 142.5, 138.9, 134.9, 134.6, 133.9, 129.2, 129.1, 128.4, 128.0, 127.6, 127.4, 126.4, 87.2, 66.0, 48.4, 42.4, 42.2; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{27}H_{22}N_2O_2SCl$ [$(M + H)^+$], 473.1091; found, 473.1098.

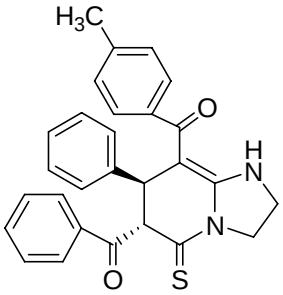
((6*S*,7*S*)-6-Benzoyl-7-phenyl-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-8-yl)(4-bromophenyl)methanone (4q)



Yellow solid; Mp 250–251 °C; **IR** (KBr): 1683, 1643, 1417, 1201 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.78 (s, 1H, NH), 6.64–7.98 (m, 14H, ArH), 5.32 (s, 1H, CHCO), 4.59–4.64 (m, 1H, NCH_2), 4.23–4.30 (m, 1H, NCH_2), 4.17 (s, 1H, ArCH), 4.00–4.05 (m, 2H, $NHCH_2$); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.6, 194.5, 191.7, 153.8, 142.5, 139.3, 134.6, 133.9, 131.0, 129.2, 129.1, 128.6, 127.8, 127.4, 126.4, 123.2, 87.2, 66.0, 48.4, 42.3, 42.2; **HRMS** (ESI-TOF$^+$): *m/z* calcd for

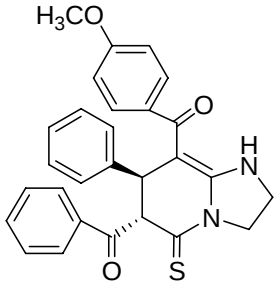
$C_{27}H_{22}N_2O_2BrS$ [$(M + H)^+$], 517.0585; found, 517.0596.

((6*S*,7*S*)-6-Benzoyl-7-phenyl-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-8-yl)(*p*-tolyl)methanone (4r)



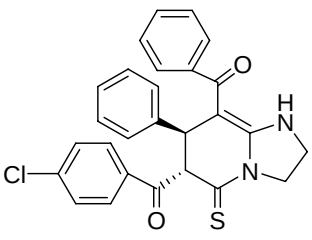
Yellow solid; Mp 259–260 °C; **IR** (KBr): 1684, 1637, 1421, 1204 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.76 (s, 1H, NH), 6.69–8.00 (m, 14H, ArH), 5.34 (d, J = 1.5 Hz, 1H, CHCO), 4.59–4.65 (m, 1H, NCH_2), 4.32 (s, 1H, ArCH), 4.22–4.26 (m, 1H, NCH_2), 3.98–4.01 (m, 2H, $NHCH_2$), 2.21 (s, 3H, $ArCH_3$); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.6, 194.5, 193.2, 153.4, 142.7, 139.0, 137.7, 134.8, 133.6, 129.0, 128.9, 128.7, 128.4, 127.1, 126.4, 126.2, 87.5, 65.8, 48.4, 42.2, 42.0, 21.2; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{28}H_{25}N_2O_2S$ [$(M + H)^+$], 453.1637; found, 453.1645.

((6*S*,7*S*)-6-Benzoyl-7-phenyl-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-8-yl)(4-methoxyphenyl)methanone (4s)



Yellow solid; Mp 251–252 °C; **IR** (KBr): 1685, 1641, 1419, 1202 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.75 (s, 1H, NH), 6.48–8.00 (m, 14H, ArH), 5.35 (d, J = 1.5 Hz, 1H, CHCO), 4.58–4.63 (m, 1H, NCH_2), 4.35 (d, J = 1.0 Hz, 1H, ArCH), 4.21–4.27 (m, 1H, NCH_2), 3.96–4.00 (m, 2H, $NHCH_2$), 3.70 (s, 3H, OCH_3); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.6, 194.5, 192.6, 160.2, 153.4, 142.8, 134.8, 133.7, 133.2, 129.1, 129.0, 128.7, 128.1, 127.2, 126.5, 113.1, 87.4, 66.0, 55.1, 48.4, 42.4, 42.1; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{28}H_{25}N_2O_3S$ [$(M + H)^+$], 469.1586; found, 469.1595.

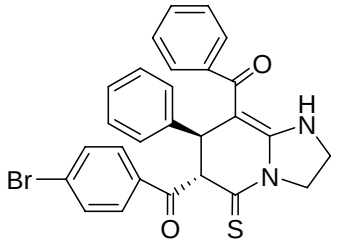
((6*S*,7*S*)-8-Benzoyl-7-phenyl-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-6-yl)(4-chlorophenyl)methanone (4t)



Yellow solid; Mp 265–266 °C; **IR** (KBr): 1686, 1639, 1417, 1204 cm^{-1}; **^{1}H NMR** (500 MHz,

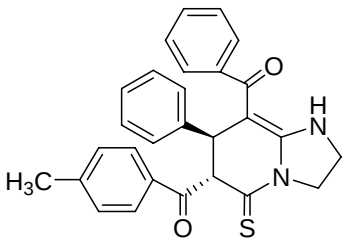
$CDCl_3$): δ = 9.75 (s, 1H, NH), 6.78–7.92 (m, 14H, ArH), 5.24 (d, J = 1.5 Hz, 1H, CHCO), 4.58–4.64 (m, 1H, NCH_2), 4.24–4.30 (m, 1H, NCH_2), 4.17 (d, J = 1.0 Hz, 1H, ArCH), 3.99–4.03 (m, 2H, $NHCH_2$); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.0, 193.4, 193.2, 153.4, 142.6, 140.5, 140.3, 133.0, 130.0, 129.3, 129.0, 127.8, 127.3, 126.3, 125.9, 87.3, 65.9, 48.4, 42.4, 42.1; **HRMS** (ESI-TOF$^+$): m/z calcd for $C_{27}H_{22}N_2O_2ClS$ [$(M + H)^+$], 473.1091; found, 473.1082.

((6*S*,7*S*)-8-Benzoyl-7-phenyl-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-6-yl)(4-bromophenyl)methanone (4u)



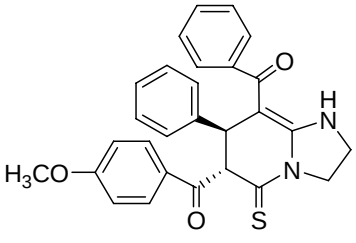
Brown yellow solid; Mp 249–250 °C; **IR** (KBr): 1684, 1637, 1416, 1202 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.75 (s, 1H, NH), 6.77–7.89 (m, 14H, ArH), 5.26 (d, J = 1.5 Hz, 1H, CHCO), 4.58–4.60 (m, 1H, NCH_2), 4.24–4.27 (m, 1H, NCH_2), 4.16 (d, J = 0.5 Hz, 1H, ArCH), 3.97–4.00 (m, 2H, $NHCH_2$); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.8, 194.1, 193.3, 153.5, 147.3, 142.9, 142.6, 140.6, 133.6, 132.4, 130.8, 130.2, 129.1, 129.0, 127.9, 127.4, 127.3, 126.5, 126.1, 126.0 125.3, 87.4, 65.7, 48.5, 42.6, 42.2; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{27}H_{22}N_2O_2BrS$ [$(M + H)^+$], 517.0585; found, 517.0575.

((6*S*,7*S*)-8-Benzoyl-7-phenyl-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-6-yl)(*p*-tolyl)methanone (4v)



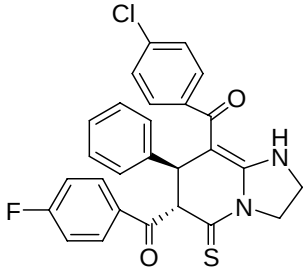
Yellow solid; Mp 253–254 °C; **IR** (KBr): 1683, 1642, 1420, 1196 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.76 (s, 1H, NH), 6.78–7.89 (m, 14H, ArH), 5.29 (d, J = 1.5 Hz, 1H, CHCO), 4.59–4.63 (m, 1H, NCH_2), 4.25–4.28 (m, 1H, NCH_2), 4.24 (d, J = 1.0 Hz, 1H, ArCH), 3.99–4.02 (m, 2H, $NHCH_2$); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.9, 194.2, 193.2, 153.5, 144.7, 142.8, 140.6, 132.6, 129.7, 128.9, 128.8, 127.7, 127.1, 126.5, 126.1, 87.5, 65.8, 48.3, 42.4, 42.1, 21.7; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{28}H_{25}N_2O_2S$ [$(M + H)^+$], 453.1637; found, 453.1629.

((6*S*,7*S*)-8-Benzoyl-7-phenyl-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-6-yl)(4-methoxyphenyl)methanone (4w)



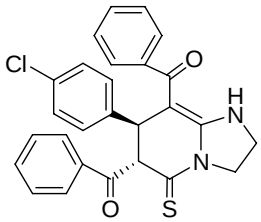
Brown yellow solid; Mp 243–244 °C; **IR** (KBr): 1677, 1645, 1420, 1205 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.75 (s, 1H, NH), 6.80–8.00 (m, 14H, ArH), 5.27 (d, J = 1.5 Hz, 1H, CHCO), 4.60–4.62 (m, 1H, NCH_2), 4.24–4.28 (m, 1H, NCH_2), 4.23 (s, 1H, ArCH), 3.99–4.02 (m, 2H, $NHCH_2$), 3.91 (s, 3H, OCH_3); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 195.1, 193.3, 193.1, 153.5, 143.0, 140.7, 131.2, 129.0, 127.9, 127.4, 127.2, 126.5, 126.2, 123.3, 114.3, 87.7, 65.6, 55.7, 48.6, 42.6, 42.2; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{28}H_{25}N_2O_3S$ [(M + H)$^+$], 469.1586; found, 469.1586.

((6*S*,7*S*)-8-(4-Chlorobenzoyl)-7-phenyl-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridin-6-yl)(4-fluorophenyl)methanone (4x)



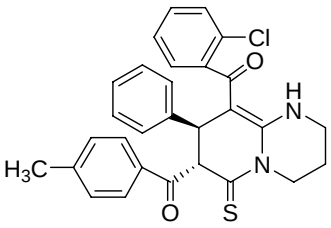
Yellow solid; Mp 248–249 °C; **IR** (KBr): 1686, 1637, 1417, 1202 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.80 (s, 1H, NH), 6.73–8.02 (m, 13H, ArH), 5.26 (d, J = 1.5 Hz, 1H, CHCO), 4.58–4.61 (m, 1H, NCH_2), 4.25–4.28 (m, 1H, NCH_2), 4.14 (d, J = 1.0 Hz, 1H, ArCH), 3.99–4.03 (m, 2H, $NHCH_2$); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.7, 194.1, 193.4, 167.2, 165.2, 153.8, 147.5, 142.6, 139.0, 134.9, 131.4, 131.1, 130.7, 129.1, 128.1, 127.4, 126.5, 125.4, 116.3, 87.4, 65.9, 48.5, 42.6, 42.2; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{27}H_{21}N_2O_2FClS$ [(M + H)$^+$], 491.0996; found, 491.0985.

((6*S*,7*S*)-7-(4-Chlorophenyl)-5-thioxo-1,2,3,5,6,7-hexahydroimidazo[1,2-*a*]pyridine-6,8-diyl) bis(phenylmethanone) (4y)

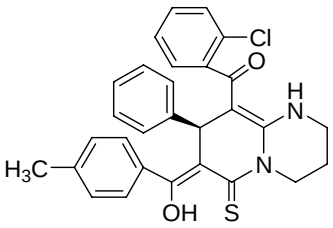


Yellow solid; Mp 255–256 °C; **IR** (KBr): 1683, 1643, 1419, 1204 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 9.74 (s, 1H, NH), 6.75–7.97 (m, 14H, ArH), 5.25 (d, J = 1.5 Hz, 1H, CHCO), 4.59–4.64 (m, 1H, NCH_2), 4.24–4.30 (m, 1H, NCH_2), 4.21 (d, J = 0.5 Hz, 1H, ArCH), 4.00–4.03 (m, 2H, $NHCH_2$); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 194.4, 194.3, 193.2, 153.4, 141.2, 140.4, 134.6, 133.8, 133.0, 129.1, 129.0, 128.6, 127.9, 125.9, 87.1, 65.8, 48.4, 42.2, 41.8; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{27}H_{22}N_2O_2SCl$ [(M + H)$^+$], 473.1091; found, 473.1075.

((7*S*,8*S*)-9-(2-Chlorobenzoyl)-8-phenyl-6-thioxo-2,3,4,6,7,8-hexahydro-1*H*-pyrido[1,2-*a*] pyrimidin-7-yl)(p-tolyl)methanone (5a)
and (*S*,*Z*)-(2-Chlorophenyl)(7-(hydroxy(p-tolyl)methylene)-8-phenyl-6-thioxo-2,3,4,6,7,8-hexahydro-1*H*-pyrido[1,2-*a*]pyrimidin-9-yl)methanone (5a')



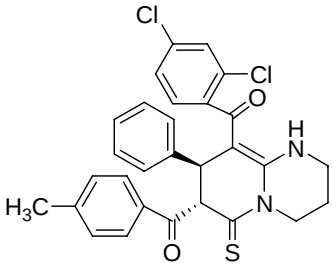
5a



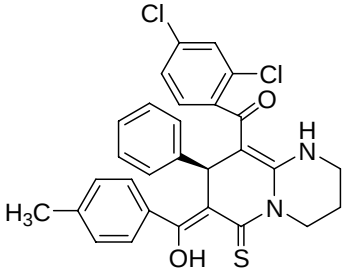
5a'

Yellow solid; Mp 203–204 °C; **IR** (KBr): 1679, 1620, 1391, 1158 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 12.79 (s, 1H, NH), 6.15–7.88 (m, 13H, ArH), 5.39 (s, 1H, CHCO), 4.79–4.81 (m, 1H, NCH_2), 4.11–4.16 (m, 1H, NCH_2), 4.00 (s, 0.4H, ArCH, **5a'**), 3.71 (s, 0.6H, ArCH, **5a**), 3.67 (s, 2H, $NHCH_2$), 2.44–2.50 (d, J = 29.0 Hz, 3H, $ArCH_3$), 2.34–2.37 (m, 1H, $CH_2CH_2CH_2$), 2.23–2.26 (m, 1H, $CH_2CH_2CH_2$), 1.64 (s, OH, **5a'**, exchanged by D_2O); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 199.7, 199.3, 194.3, 193.4, 188.4, 155.1, 144.5, 142.5, 141.2, 139.7, 132.1, 131.1, 129.7, 129.5, 128.9, 128.7, 128.4, 127.5, 127.1, 126.8, 125.8, 125.6, 89.9, 89.6, 67.7, 67.1, 46.8, 41.6, 41.4, 38.9, 21.7, 21.6; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{29}H_{26}N_2O_2SCl$ $[(M + H)^+]$, 501.1404; found, 501.1415.

((7*S*,8*S*)-9-(2,4-Dichlorobenzoyl)-8-phenyl-6-thioxo-2,3,4,6,7,8-hexahydro-1*H*-pyrido[1,2-*a*] pyrimidin-7-yl)(*p*-tolyl)methanone (5b)
and (*S*,*Z*)-(2,4-Dichlorophenyl)(7-(hydroxy(*p*-tolyl)methylene)-8-phenyl-6-thioxo-2,3,4,6,7,8-hexahydro-1*H*-pyrido[1,2-*a*]pyrimidin-9-yl)methanone (5b')



5b



5b'

Yellow solid; Mp 223–224 °C; **IR** (KBr): 1678, 1622, 1377, 1185 cm^{-1}; **^{1}H NMR** (500 MHz, $CDCl_3$): δ = 12.75 (s, 1H, NH), 6.09–7.86 (m, 12H, ArH), 5.40 (s, 1H, CHCO), 4.76–4.80 (m, 1H, NCH_2), 4.11–4.14 (m, 1H, NCH_2), 3.95 (s, 0.4H, ArCH, **5b'**), 3.66 (s, 0.6H, ArCH, **5b**), 3.66 (s, 2H, $NHCH_2$), 2.45–2.51 (d, J = 27.0 Hz, 3H, $ArCH_3$), 2.35 (m, 1H, $CH_2CH_2CH_2$), 2.23 (m, 1H, $CH_2CH_2CH_2$); **^{13}C NMR** (125 MHz, $CDCl_3$): δ = 199.6, 199.2, 194.2, 193.3, 155.2, 144.7, 142.3, 140.8, 138.2, 137.9, 134.0, 132.0, 130.7, 129.6, 128.8, 128.4, 127.7, 127.3, 126.7, 126.2, 89.8, 89.6, 67.7, 67.1, 46.8, 41.5, 39.0, 21.7, 21.5; **HRMS** (ESI-TOF$^+$): *m/z* calcd for $C_{29}H_{25}N_2O_2SCl_2$ $[(M + H)^+]$, 535.1014; found, 535.1024.

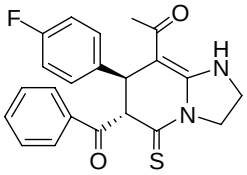
References

1. (a) L. S. Geoffroy, C. Alain, K. Gilbert, *Synthetic Communications*. **2005**, *35*, 693–699. (b) M. M. E. Ahmed, A. M. Abdussalam, A. A. Hatif, *Synthetic Communications*. **1997**, *27(14)*, 2433–2444.
2. (a) A. V. N. Reddy, S. N. Maiti, I. P. Singh, G. Micetich, *Synth. Commun*. **1989**, *19*, 3021−3025. (b) H. Foks, D. Pancechowska-Ksepko, M. Janowiec, Z. Zwolska, E. Augustynowicz-Kopec, *Phosphorus, Sulfur Silicon Relat. Elem*. **2005**, *180*, 2291−2297.
3. M.-X. Zhao, M.-X. Wang, Z.-T. Huang, *Tetrahedron* **2002**, *58*, 1309−1316.
4. (a) Z.-T. Huang, M.-X. Wang, *Synthesis* **1992**, *12*, 1273−1276. (b) Z.-J. Li, D. Charles, *Synth.*

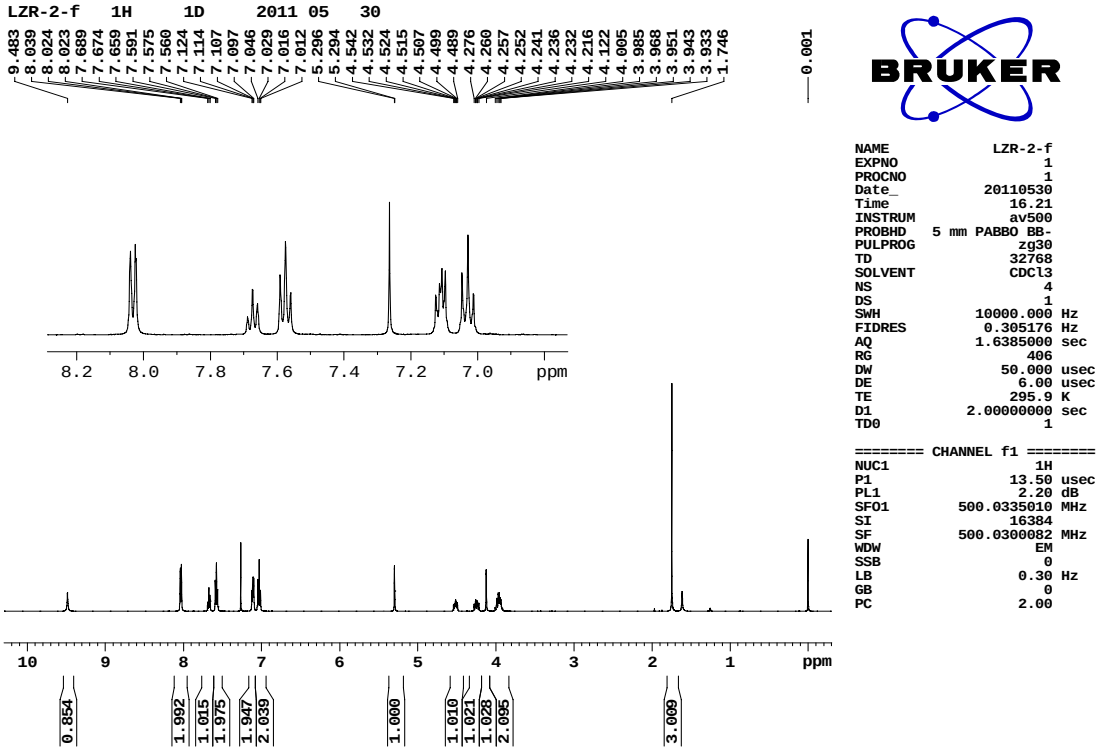
Commun. **2001**, *31*, 527−533.

5. (a) Z.-T. Huang, P.-C. Zhang, *Chem. Ber*. **1989**, *122*, 2011–2016. (b) Z.-T. Huang, X. Shi, *Synthesis* **1990**, *2*, 162–167.
6. (a) R. K. Ali, F. M. Mehdi, *Journal of the Chinese Chemical Society*, **2008**, *55*, 639–642. (b) T. Bahman, A. R. Kiasat, *J. Chem. Researchs*, **1998**, 455. (c) R. K. Ali, K. Foad, S. Ali, *Synthetic Communications*, **2008**, *38*, 1057–1063. (d) K. Riza, R. B. Nicholas, *J. Org. Chem*. **1981**, *46*, 197–201. (e) S. Mohammad, A. Zeinab, M. Barahman, *SYNTHESIS*, **2010**, *3*, 0392–0394.
7. (a) G. Singh, S. S. Bhattacharjee, H. Ila, H. Junjappa, *Communications Synthesis* **1982**, 693-694. (b) R. Samuel, C. V. Asokan, S. Suma, P. Chandran, S. Retnamma, E. R. Anabha, *Tetrahedron Letters* **2007**, *48*, 8376–8378. (c) K. N. Satheesh, C. V. Asokan, *Synthetic Communications*, **1999**, *29(5)*, 791–798.

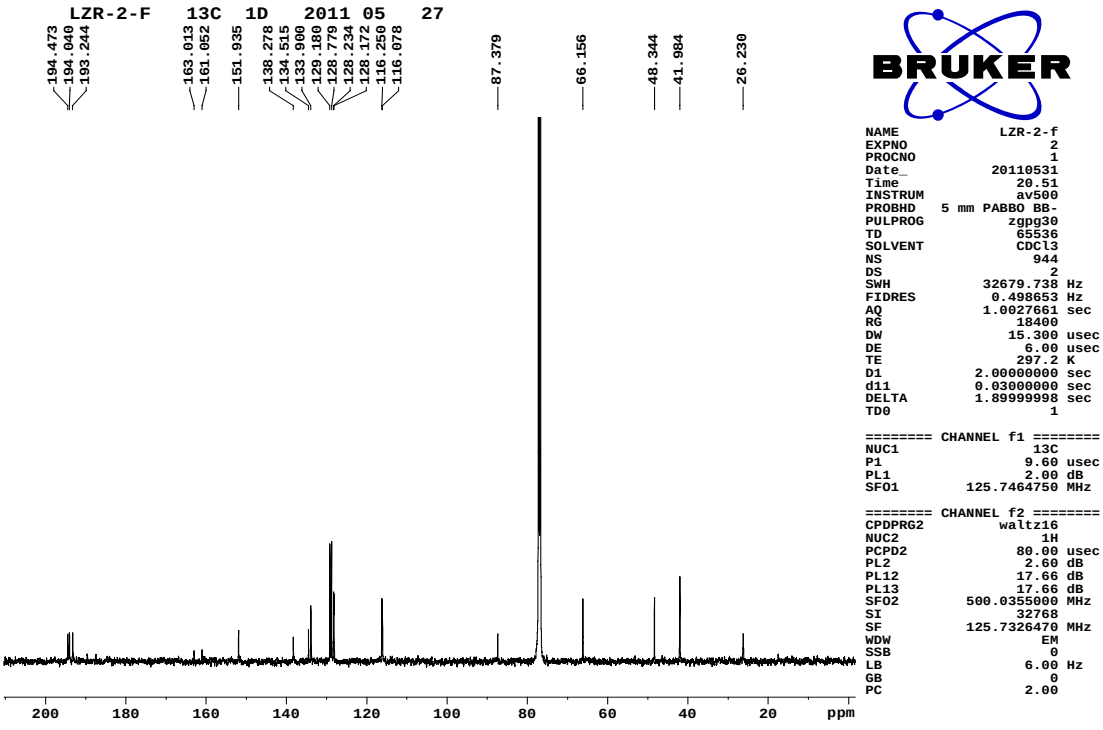
^{1}H and ^{13}C NMR spectra



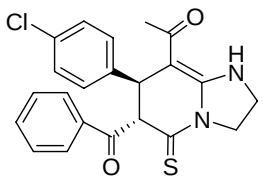
^{1}H and ^{13}C NMR spectra of **4a**



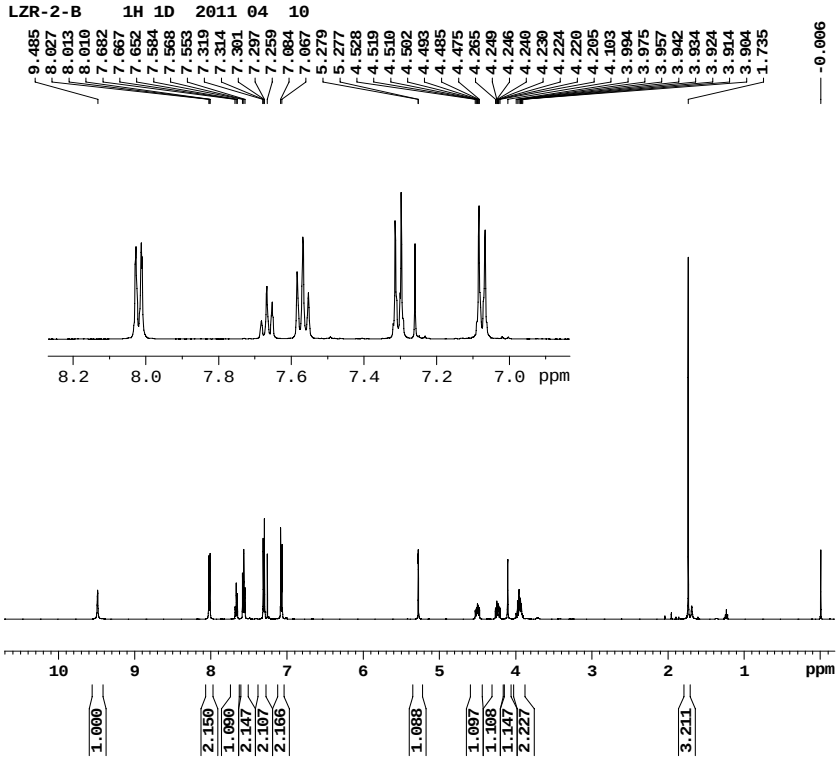

```
NAME              LZR-2-f
EXPNO                   1
PROCNO                  1
Date_            20110530
Time                16.21
INSTRUM             av500
PROBHD   5 mm PABBO BB-
PULPROG              zg30
TD                  32768
SOLVENT             CDCl3
NS                      4
DS                      1
SWH             10000.000 Hz
FIDRES              0.305176 Hz
AQ               1.6385000 sec
RG                    406
DW                 50.000 usec
DE                  6.00 usec
TE                 295.9 K
D1              2.00000000 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                   1H
P1                  13.50 usec
PL1                  2.20 dB
SFO1          500.0335010 MHz
SI                  16384
SF            500.0300082 MHz
WDW                    EM
SSB                     0
LB                   0.30 Hz
GB                      0
PC                   2.00
```



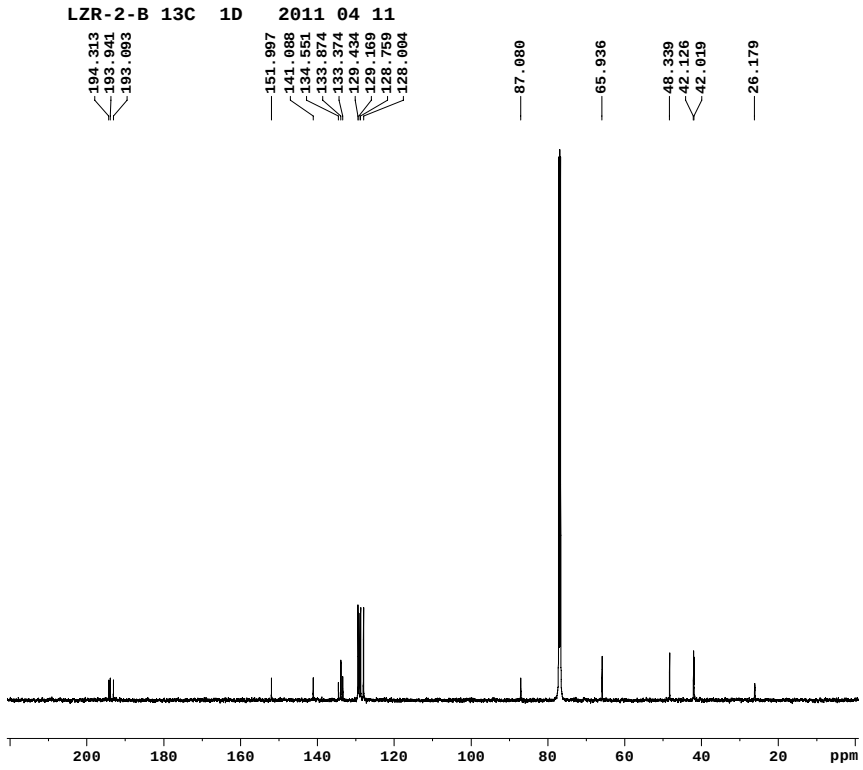

```
NAME              LZR-2-f
EXPNO                   2
PROCNO                  1
Date_            20110531
Time                20.51
INSTRUM             av500
PROBHD   5 mm PABBO BB-
PULPROG            zgpg30
TD                  65536
SOLVENT             CDCl3
NS                    944
DS                      2
SWH             32679.738 Hz
FIDRES              0.498653 Hz
AQ               1.0027661 sec
RG                    18400
DW                 15.300 usec
DE                  6.00 usec
TE                 297.2 K
D1              2.00000000 sec
d11             0.03000000 sec
DELTA           1.89999998 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                   13C
P1                  9.60 usec
PL1                 2.00 dB
SFO1          125.7464750 MHz

======== CHANNEL f2 ========
CPDPRG2             waltz16
NUC2                   1H
PCPD2              80.00 usec
PL2                 2.60 dB
PL12               17.66 dB
PL13               17.66 dB
SFO2          500.0355000 MHz
SI                  32768
SF            125.7326470 MHz
WDW                    EM
SSB                     0
LB                   6.00 Hz
GB                      0
PC                   2.00
```

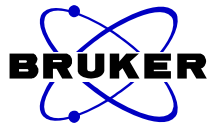


^{1}H and ^{13}C NMR spectra of **4b**

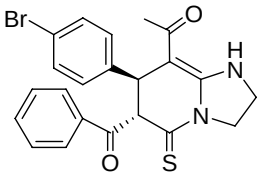



```
NAME                LZR-2-B
EXPNO                     1
PROCNO                    1
Date_              20110410
Time                  19.36
INSTRUM               av500
PROBHD   5 mm PABBO BB-
PULPROG                zg30
TD                    32768
SOLVENT               CDCl3
NS                        4
DS                        1
SWH               10000.000 Hz
FIDRES                0.305176 Hz
AQ                1.6385000 sec
RG                      256
DW                   50.000 usec
DE                    6.00 usec
TE                   293.6 K
D1               2.00000000 sec
TD0                       1

======== CHANNEL f1 ========
NUC1                    1H
P1                   13.50 usec
PL1                   2.20 dB
SFO1           500.0335010 MHz
SI                    16384
SF             500.0300106 MHz
WDW                     EM
SSB                      0
LB                    0.30 Hz
GB                       0
PC                    4.00
```

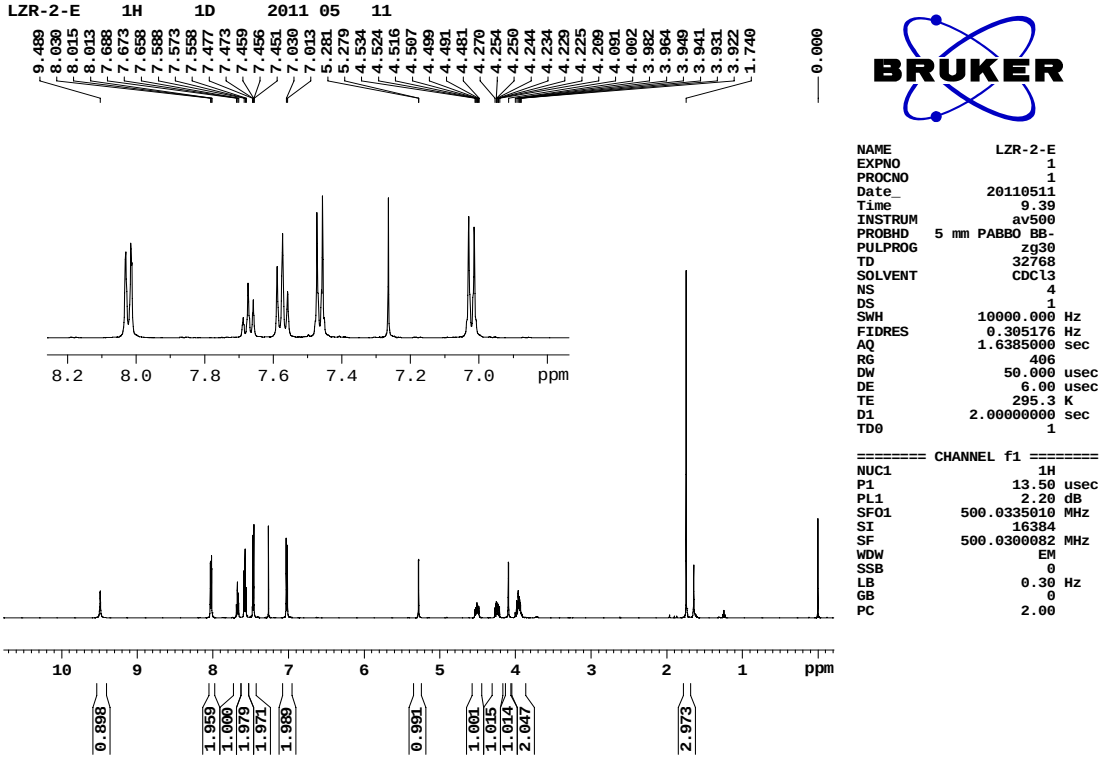




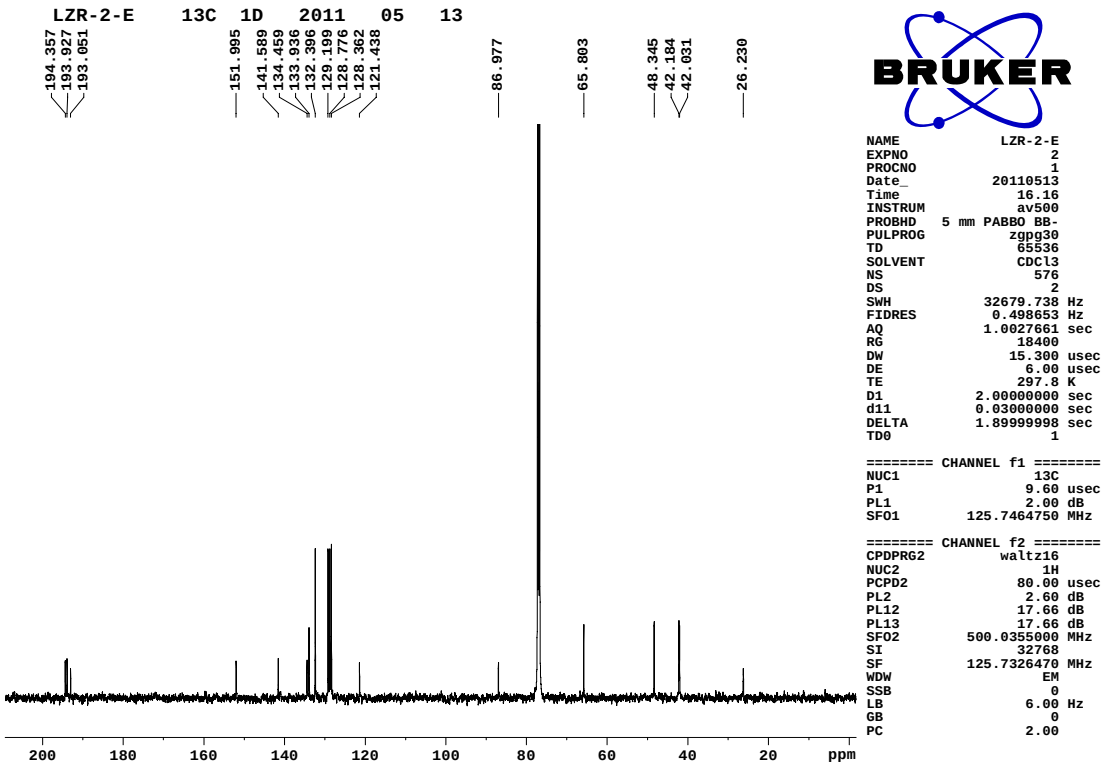

```
NAME                LZR-2-B
EXPNO                     2
PROCNO                    1
Date_              20110411
Time                  16.25
INSTRUM               av500
PROBHD   5 mm PABBO BB-
PULPROG             zgpg30
TD                    65536
SOLVENT               CDCl3
NS                      802
DS                        2
SWH               32679.738 Hz
FIDRES                0.498653 Hz
AQ                1.0027661 sec
RG                      18400
DW                   15.300 usec
DE                    6.00 usec
TE                   296.6 K
D1               2.00000000 sec
d11              0.03000000 sec
DELTA            1.89999998 sec
TD0                       1

======== CHANNEL f1 ========
NUC1                   13C
P1                    9.60 usec
PL1                   2.00 dB
SFO1          125.7464750 MHz

======== CHANNEL f2 ========
CPDPRG2             waltz16
NUC2                    1H
PCPD2                80.00 usec
PL2                   2.60 dB
PL12                 17.66 dB
PL13                 17.66 dB
SFO2           500.0355000 MHz
SI                    32768
SF            125.7326470 MHz
WDW                     EM
SSB                      0
LB                    5.00 Hz
GB                       0
PC                    2.00
```



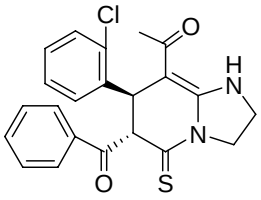
^{1}H and ^{13}C NMR spectra of **4c**



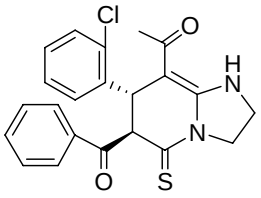

```
NAME              LZR-2-E
EXPNO                   1
PROCNO                  1
Date_            20110511
Time                 9.39
INSTRUM             av500
PROBHD   5 mm PABBO BB-
PULPROG              zg30
TD                  32768
SOLVENT             CDCl3
NS                      4
DS                      1
SWH          10000.000 Hz
FIDRES           0.305176 Hz
AQ           1.6385000 sec
RG                    406
DW               50.000 usec
DE                6.00 usec
TE                295.3 K
D1           2.00000000 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                  1H
P1                13.50 usec
PL1                2.20 dB
SFO1         500.0335010 MHz
SI                  16384
SF           500.0300082 MHz
WDW                   EM
SSB                    0
LB                0.30 Hz
GB                     0
PC                2.00
```




```
NAME              LZR-2-E
EXPNO                   2
PROCNO                  1
Date_            20110513
Time                16.16
INSTRUM             av500
PROBHD   5 mm PABBO BB-
PULPROG           zgpg30
TD                  65536
SOLVENT             CDCl3
NS                    576
DS                      2
SWH          32679.738 Hz
FIDRES           0.498653 Hz
AQ           1.0027661 sec
RG                  18400
DW               15.300 usec
DE                6.00 usec
TE                297.8 K
D1           2.00000000 sec
d11          0.03000000 sec
DELTA        1.89999998 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                 13C
P1                9.60 usec
PL1                2.00 dB
SFO1        125.7464750 MHz

======== CHANNEL f2 ========
CPDPRG2          waltz16
NUC2                  1H
PCPD2            80.00 usec
PL2                2.60 dB
PL12              17.66 dB
PL13              17.66 dB
SFO2        500.0355000 MHz
SI                  32768
SF          125.7326470 MHz
WDW                   EM
SSB                    0
LB                6.00 Hz
GB                     0
PC                2.00
```

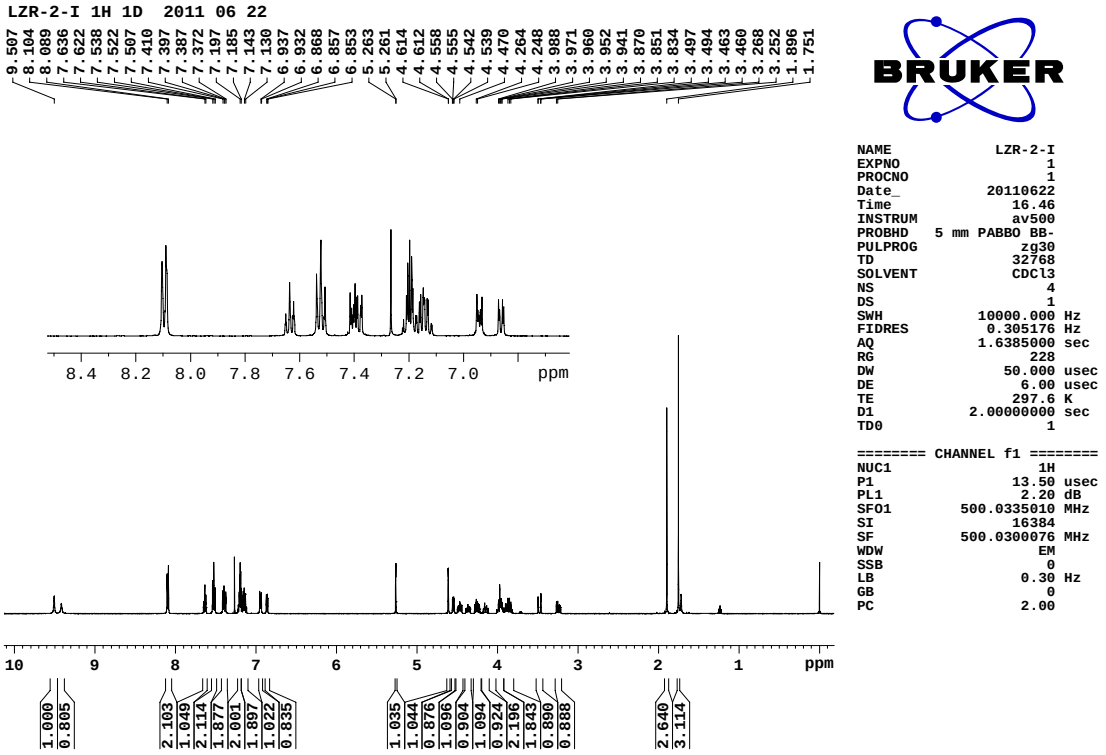


4d

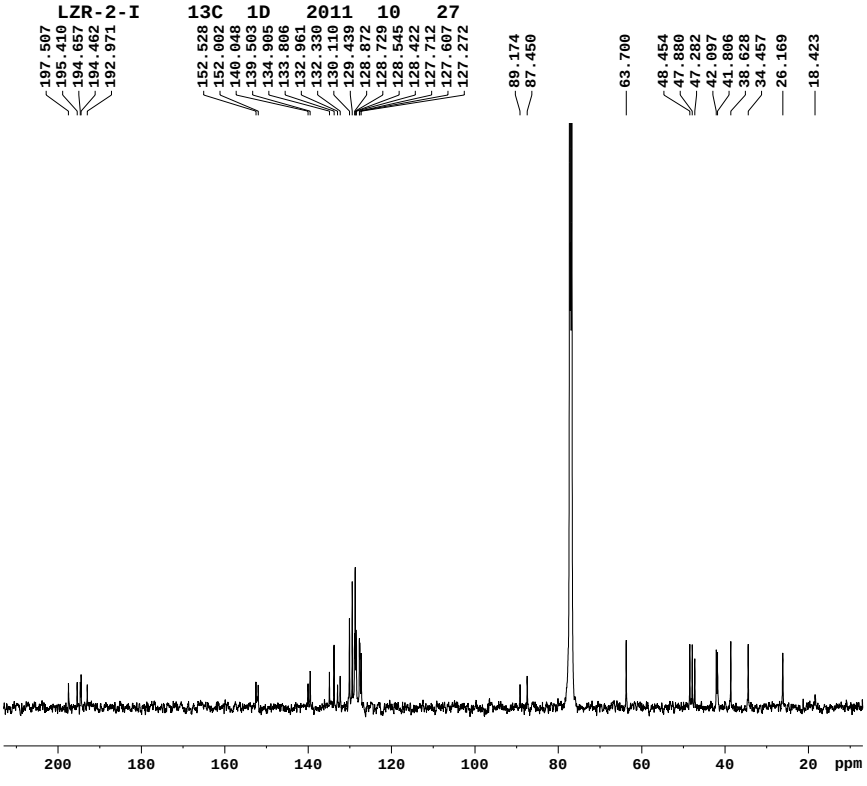


4d'

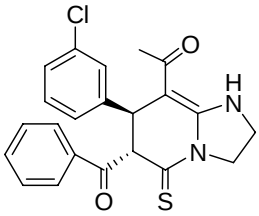
^{1}H and ^{13}C NMR spectra of **4d/4d'**



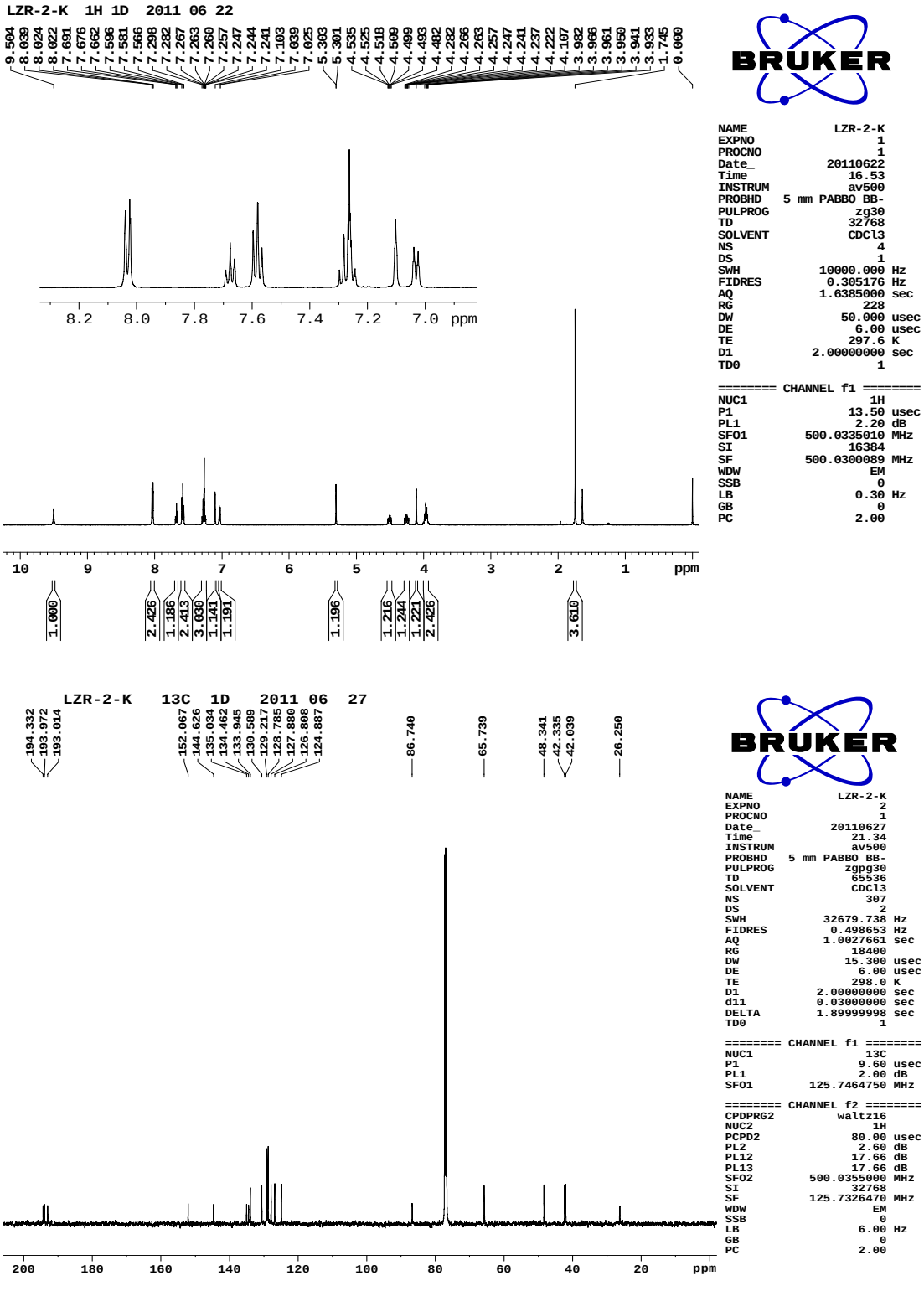

```
NAME              LZR-2-I
EXPNO                   1
PROCNO                  1
Date_            20110622
Time                16.46
INSTRUM             av500
PROBHD   5 mm PABBO BB-
PULPROG              zg30
TD                  32768
SOLVENT             CDCl3
NS                      4
DS                      1
SWH           10000.000 Hz
FIDRES         0.305176 Hz
AQ             1.6385000 sec
RG                    228
DW                 50.000 usec
DE                  6.00 usec
TE                297.6 K
D1            2.00000000 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                   1H
P1                  13.50 usec
PL1                  2.20 dB
SFO1          500.0335010 MHz
SI                  16384
SF            500.0300076 MHz
WDW                    EM
SSB                     0
LB                   0.30 Hz
GB                      0
PC                   2.00
```

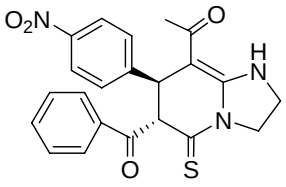



```
NAME              LZR-2-I
EXPNO                   2
PROCNO                  1
Date_            20111027
Time                16.07
INSTRUM             av500
PROBHD   5 mm PABBO BB-
PULPROG             zgpg30
TD                  65536
SOLVENT             CDCl3
NS                   1177
DS                      2
SWH           32679.738 Hz
FIDRES         0.498653 Hz
AQ             1.0027661 sec
RG                  18400
DW                 15.300 usec
DE                  6.00 usec
TE                296.3 K
D1            2.00000000 sec
d11           0.03000000 sec
DELTA         1.89999998 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                   13C
P1                   9.60 usec
PL1                  2.00 dB
SFO1          125.7464750 MHz

======== CHANNEL f2 ========
CPDPRG2             waltz16
NUC2                   1H
PCPD2               80.00 usec
PL2                  2.60 dB
PL12                17.66 dB
PL13                17.66 dB
SFO2          500.0355000 MHz
SI                  32768
SF            125.7326437 MHz
WDW                    EM
SSB                     0
LB                  10.00 Hz
GB                      0
PC                   1.00
```

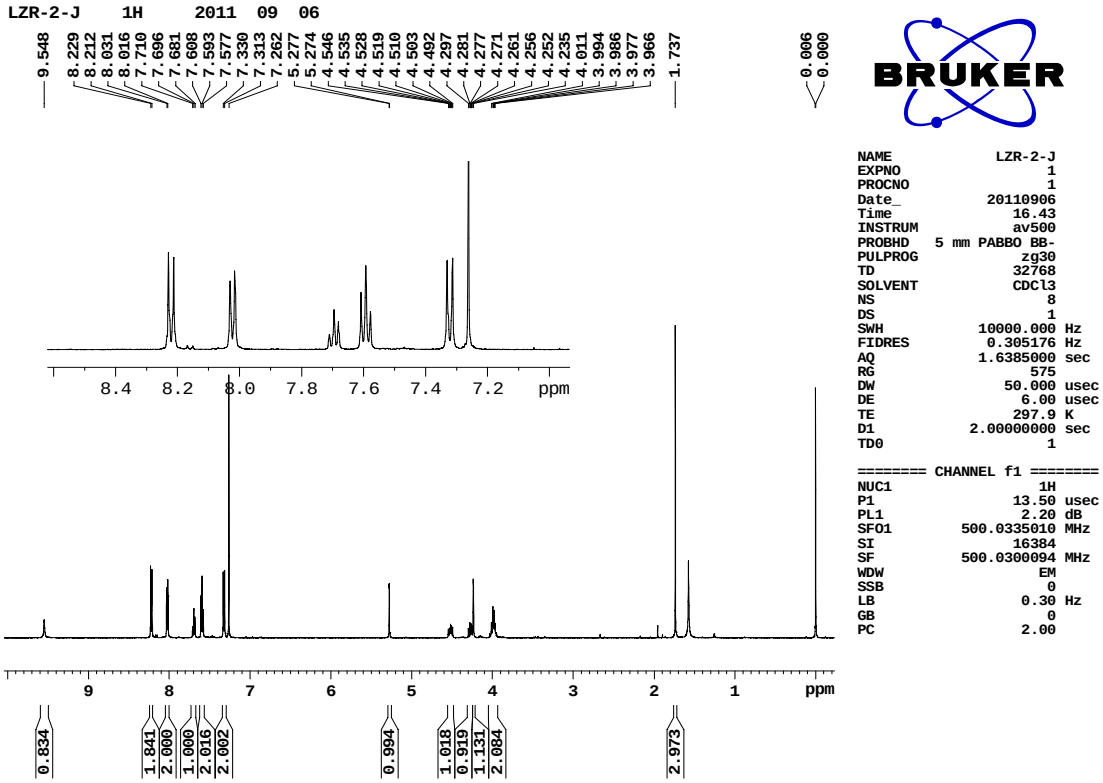


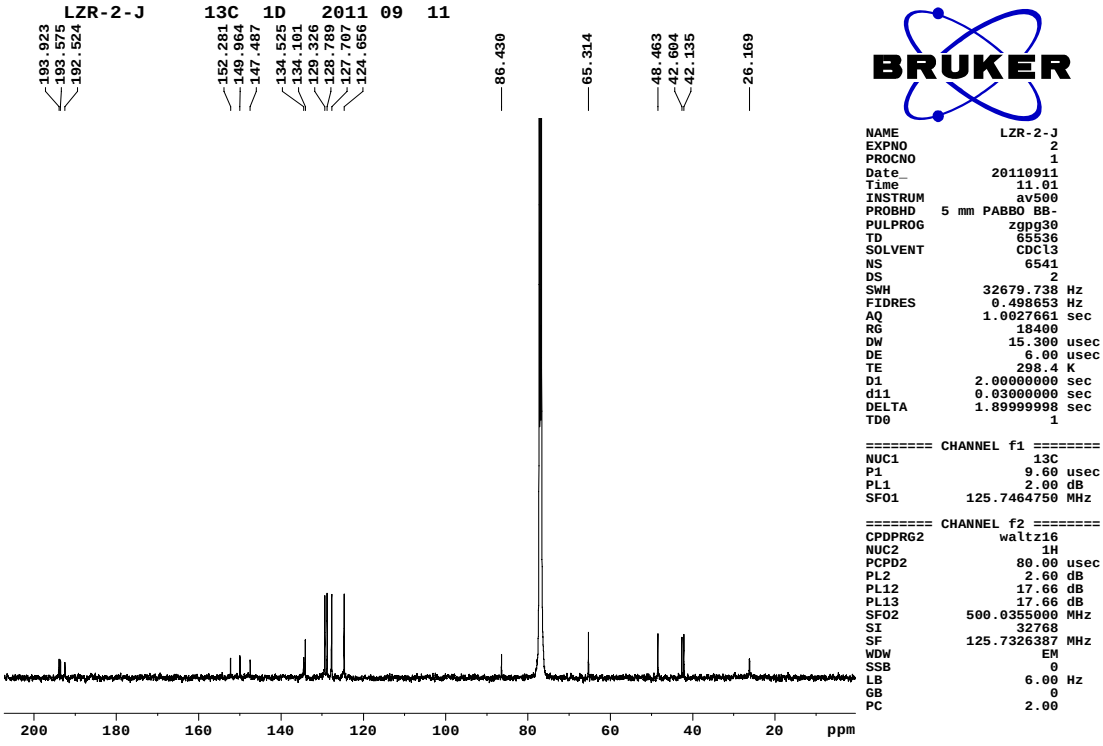
^{1}H and ^{13}C NMR spectra of **4e**

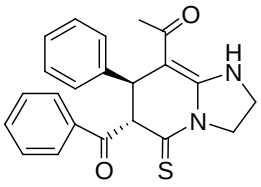




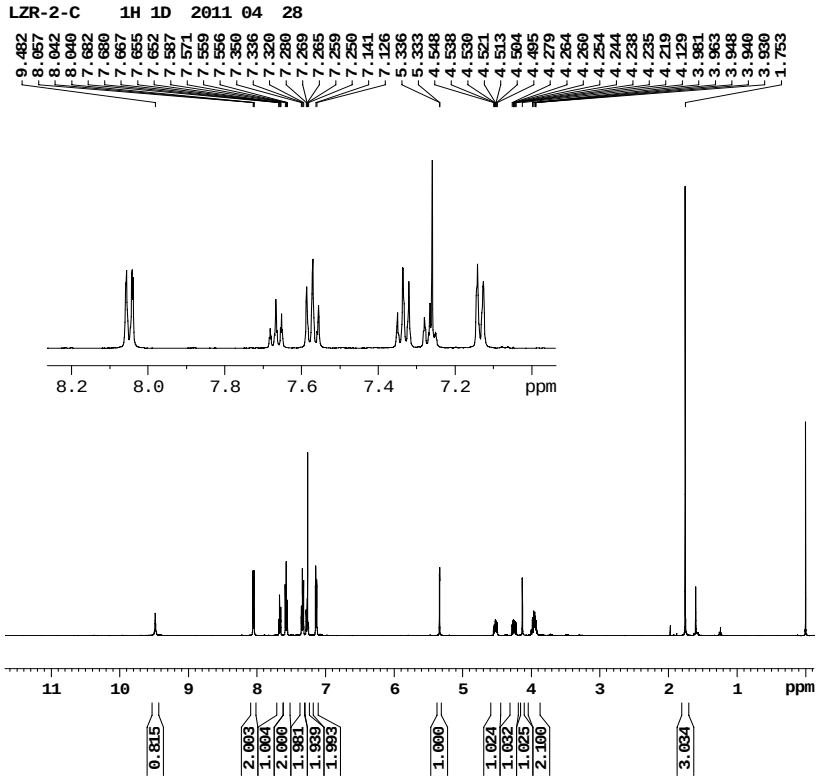
^{1}H and ^{13}C NMR spectra of **4f**





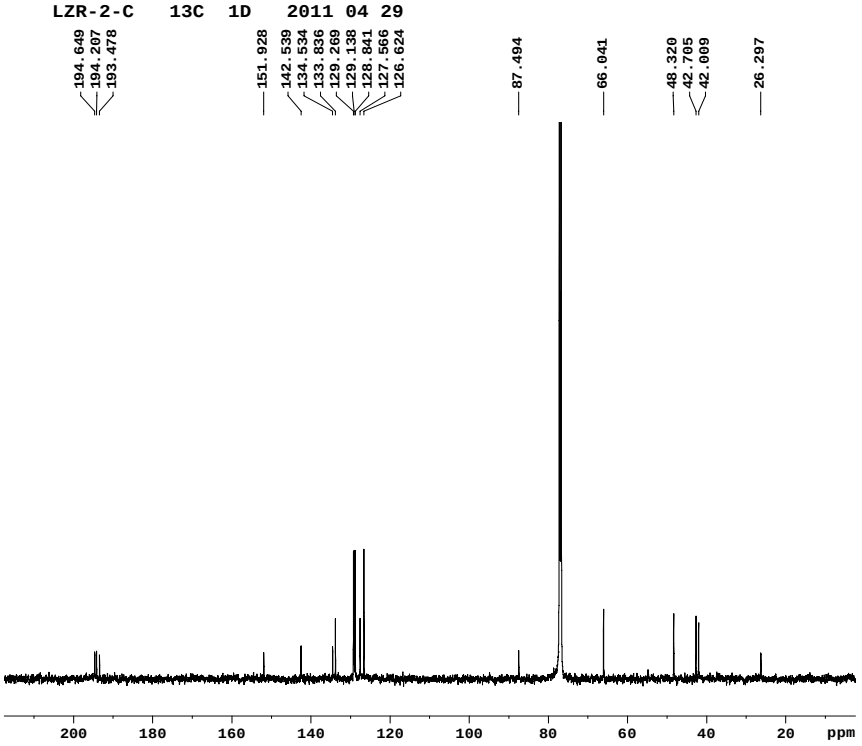


^{1}H and ^{13}C NMR spectra of **4g**



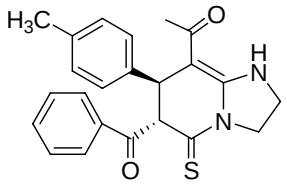
BRUKER

```
NAME              LZR-2-C
EXPNO                   1
PROCNO                  1
Date_            20110428
Time                20.27
INSTRUM             av500
PROBHD   5 mm PABBO BB-
PULPROG              zg30
TD                  32768
SOLVENT             CDCl3
NS                      4
DS                      1
SWH          10000.000 Hz
FIDRES        0.305176 Hz
AQ            1.6385000 sec
RG                    456
DW               50.000 usec
DE                6.00 usec
TE                295.4 K
D1           2.00000000 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                  1H
P1                13.50 usec
PL1                2.20 dB
SFO1        500.0335010 MHz
SI                  16384
SF          500.0300106 MHz
WDW                   EM
SSB                    0
LB                0.30 Hz
GB                     0
PC                10.00
```

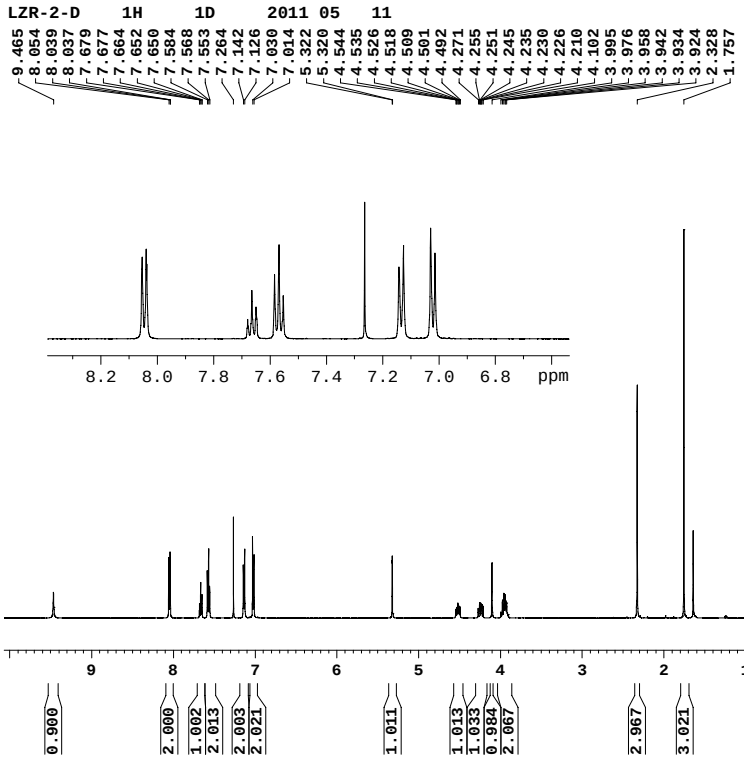


BRUKER

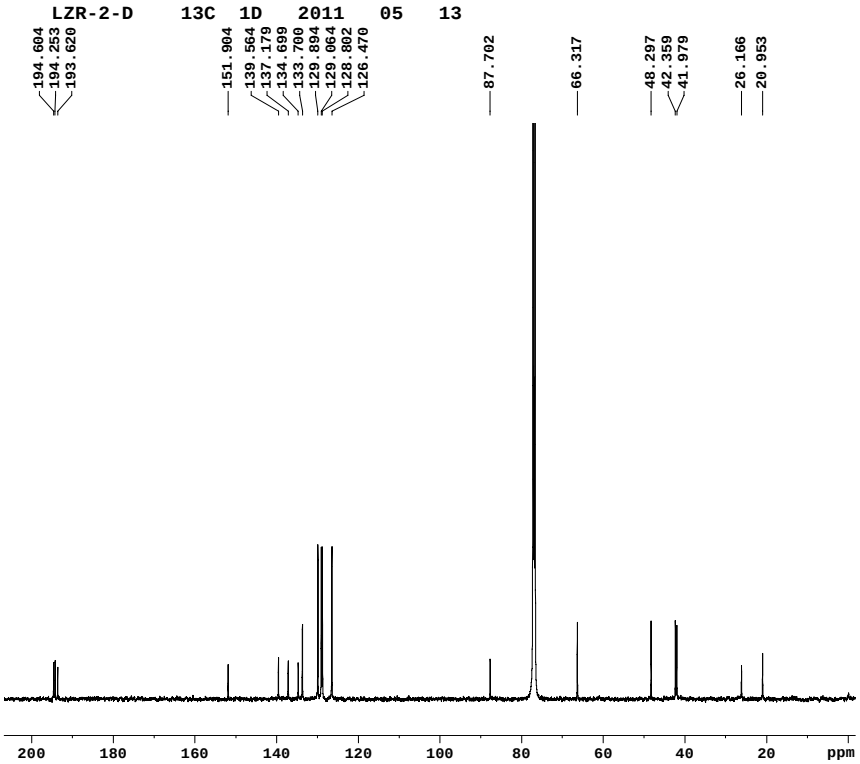
```
NAME              LZR-2-C
EXPNO                   2
PROCNO                  1
Date_            20110429
Time                16.48
INSTRUM             av500
PROBHD   5 mm PABBO BB-
PULPROG            zgpg30
TD                  65536
SOLVENT             CDCl3
NS                   2440
DS                      2
SWH          32679.738 Hz
FIDRES        0.498653 Hz
AQ            1.0027661 sec
RG                  18400
DW               15.300 usec
DE                6.00 usec
TE                297.9 K
D1           2.00000000 sec
d11          0.03000000 sec
DELTA        1.89999998 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                 13C
P1                9.60 usec
PL1               2.00 dB
SFO1        125.7464750 MHz

======== CHANNEL f2 ========
CPDPRG2          waltz16
NUC2                  1H
PCPD2            80.00 usec
PL2               2.60 dB
PL12             17.66 dB
PL13             17.66 dB
SFO2        500.0355000 MHz
SI                  32768
SF          125.7326470 MHz
WDW                   EM
SSB                    0
LB                5.00 Hz
GB                     0
PC                2.00
```

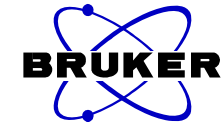


^{1}H and ^{13}C NMR spectra of **4h**

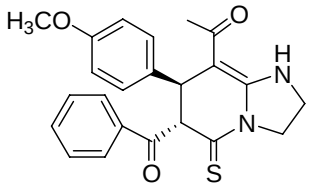


```
NAME              LZR-2-D
EXPNO                   1
PROCNO                  1
Date_            20110511
Time                9.46
INSTRUM            av500
PROBHD    5 mm PABBO BB-
PULPROG             zg30
TD                 32768
SOLVENT            CDCl3
NS                      4
DS                      1
SWH           10000.000 Hz
FIDRES            0.305176 Hz
AQ             1.6385000 sec
RG                    406
DW                 50.000 usec
DE                  6.00 usec
TE                295.3 K
D1             2.00000000 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                   1H
P1                  13.50 usec
PL1                  2.20 dB
SFO1          500.0335010 MHz
SI                  16384
SF            500.0300082 MHz
WDW                    EM
SSB                     0
LB                   0.30 Hz
GB                      0
PC                   2.00
```

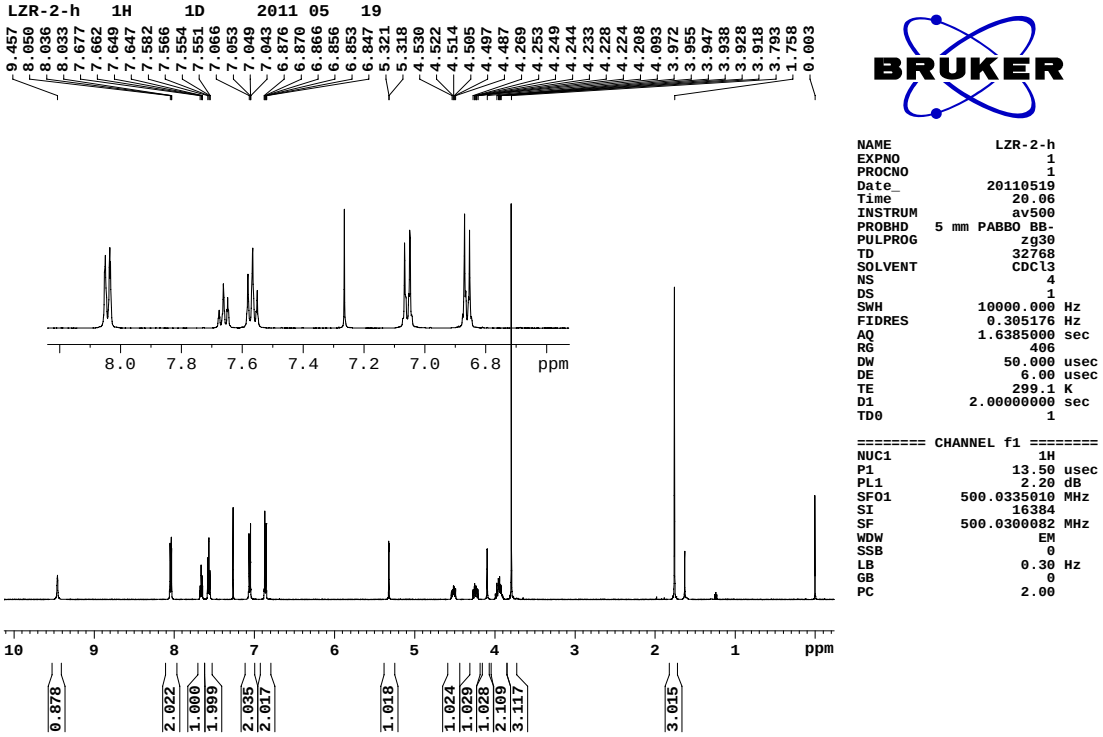


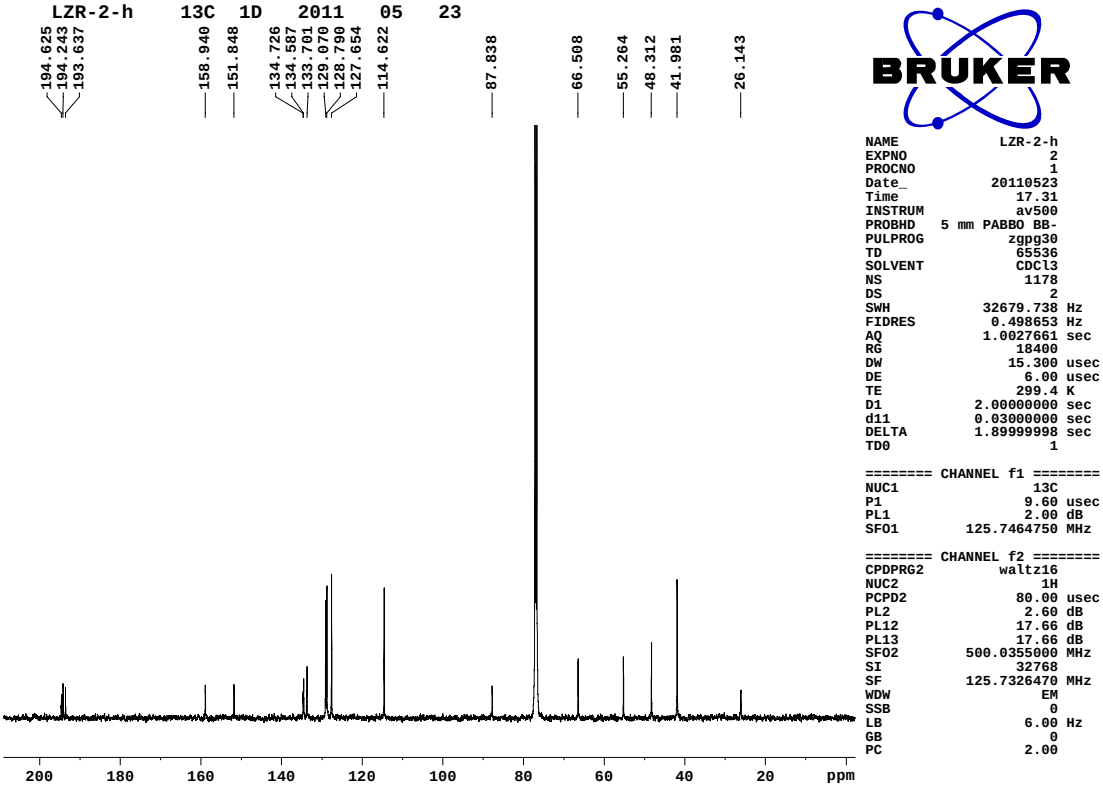


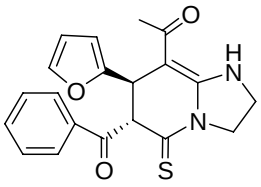
```
NAME              LZR-2-D
EXPNO                   2
PROCNO                  1
Date_            20110513
Time               14.29
INSTRUM            av500
PROBHD    5 mm PABBO BB-
PULPROG           zgpg30
TD                 65536
SOLVENT            CDCl3
NS                  3599
DS                     2
SWH           32679.738 Hz
FIDRES            0.498653 Hz
AQ             1.0027661 sec
RG                  18400
DW                 15.300 usec
DE                  6.00 usec
TE                300.2 K
D1             2.00000000 sec
d11            0.03000000 sec
DELTA          1.89999998 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                  13C
P1                  9.60 usec
PL1                  2.00 dB
SFO1         125.7464750 MHz

======== CHANNEL f2 ========
CPDPRG2          waltz16
NUC2                   1H
PCPD2               80.00 usec
PL2                  2.60 dB
PL12                17.66 dB
PL13                17.66 dB
SFO2          500.0355000 MHz
SI                  32768
SF            125.7326470 MHz
WDW                    EM
SSB                     0
LB                   6.00 Hz
GB                      0
PC                   2.00
```



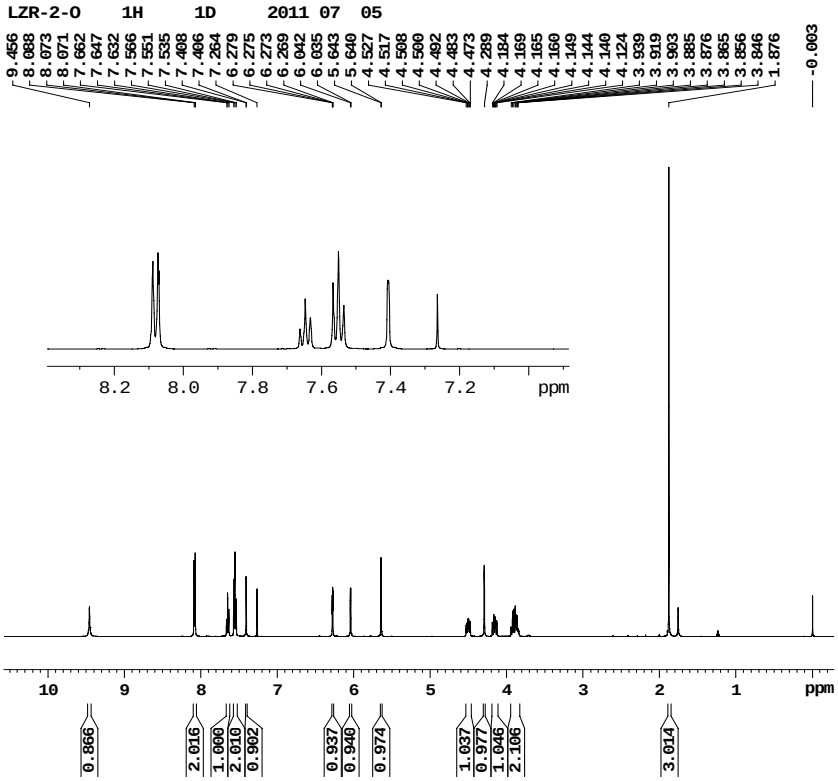
^{1}H and ^{13}C NMR spectra of **4i**



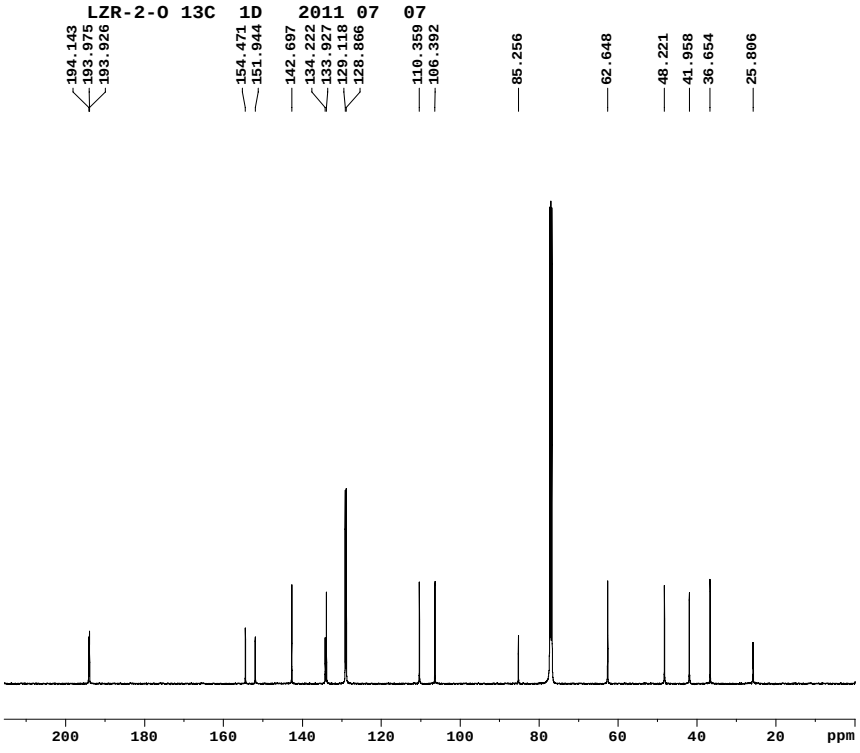




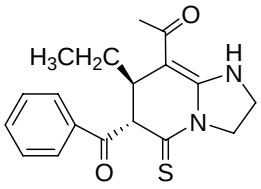
^{1}H and ^{13}C NMR spectra of **4j**



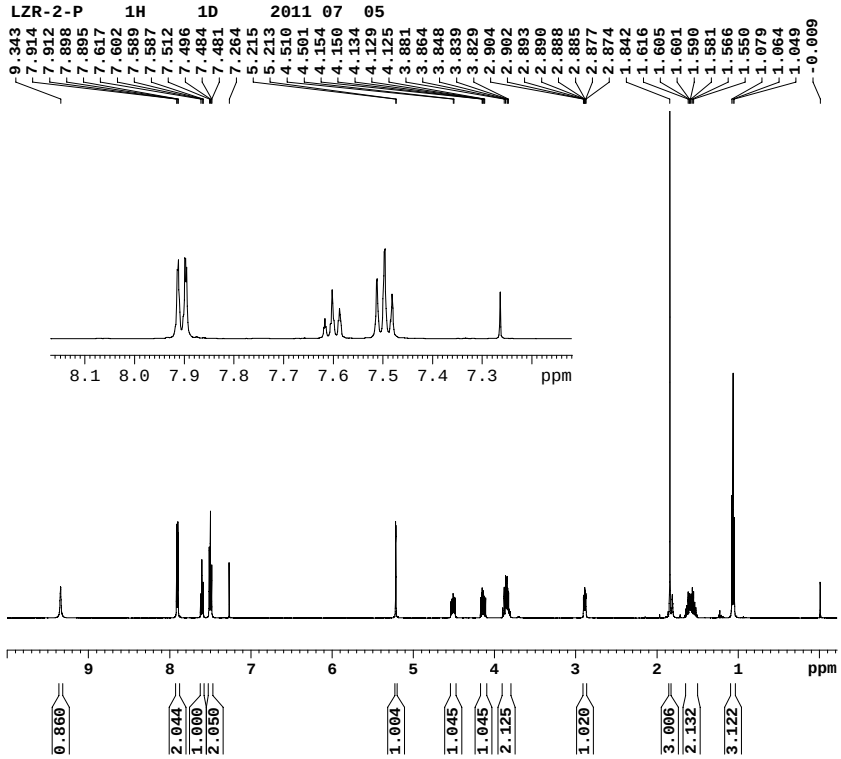
NAME	LZR-2-O
EXPNO	1
PROCNO	1
Date_	20110705
Time	19.39
INSTRUM	av500
PROBHD	5 mm PABBO BB-
PULPROG	zg30
TD	32768
SOLVENT	CDCl3
NS	4
DS	1
SWH	10000.000 Hz
FIDRES	0.305176 Hz
AQ	1.6385000 sec
RG	228
DW	50.000 usec
DE	6.00 usec
TE	295.7 K
D1	2.00000000 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	1H
P1	13.50 usec
PL1	2.20 dB
SFO1	500.0335010 MHz
SI	16384
SF	500.0300082 MHz
WDW	EM
SSB	0
LB	0.30 Hz
GB	0
PC	2.00



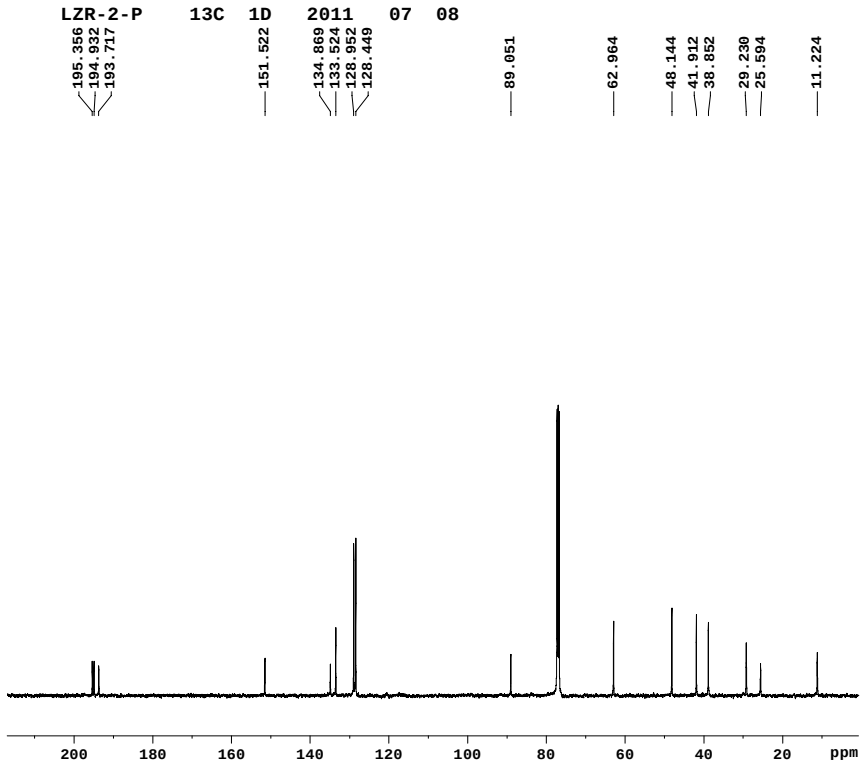
NAME	LZR-2-O
EXPNO	2
PROCNO	1
Date_	20110707
Time	16.12
INSTRUM	av500
PROBHD	5 mm PABBO BB-
PULPROG	zgpg30
TD	65536
SOLVENT	CDCl3
NS	8030
DS	2
SWH	32679.738 Hz
FIDRES	0.498653 Hz
AQ	1.0027661 sec
RG	18400
DW	15.300 usec
DE	6.00 usec
TE	298.8 K
D1	2.00000000 sec
d11	0.03000000 sec
DELTA	1.89999998 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	13C
P1	9.60 usec
PL1	2.00 dB
SFO1	125.7464750 MHz
======== CHANNEL f2 ========	
CPDPRG2	waltz16
NUC2	1H
PCPD2	80.00 usec
PL2	2.60 dB
PL12	17.66 dB
PL13	17.66 dB
SFO2	500.0355000 MHz
SI	32768
SF	125.7326470 MHz
WDW	EM
SSB	0
LB	5.00 Hz
GB	0
PC	2.00

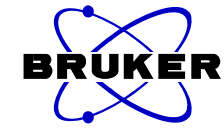


^{1}H and ^{13}C NMR spectra of **4k**

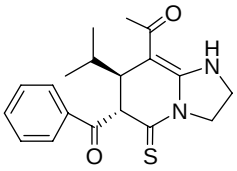



```
NAME              LZR-2-P
EXPNO                   1
PROCNO                  1
Date_            20110705
Time                19.46
INSTRUM             av500
PROBHD    5 mm PABBO BB-
PULPROG              zg30
TD                  32768
SOLVENT             CDCl3
NS                      4
DS                      1
SWH           10000.000 Hz
FIDRES            0.305176 Hz
AQ             1.6385000 sec
RG                    228
DW                 50.000 usec
DE                  6.00 usec
TE                 295.6 K
D1            2.00000000 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                   1H
P1                  13.50 usec
PL1                  2.20 dB
SFO1          500.0335010 MHz
SI                  16384
SF            500.0300082 MHz
WDW                    EM
SSB                     0
LB                   0.30 Hz
GB                      0
PC                   2.00
```

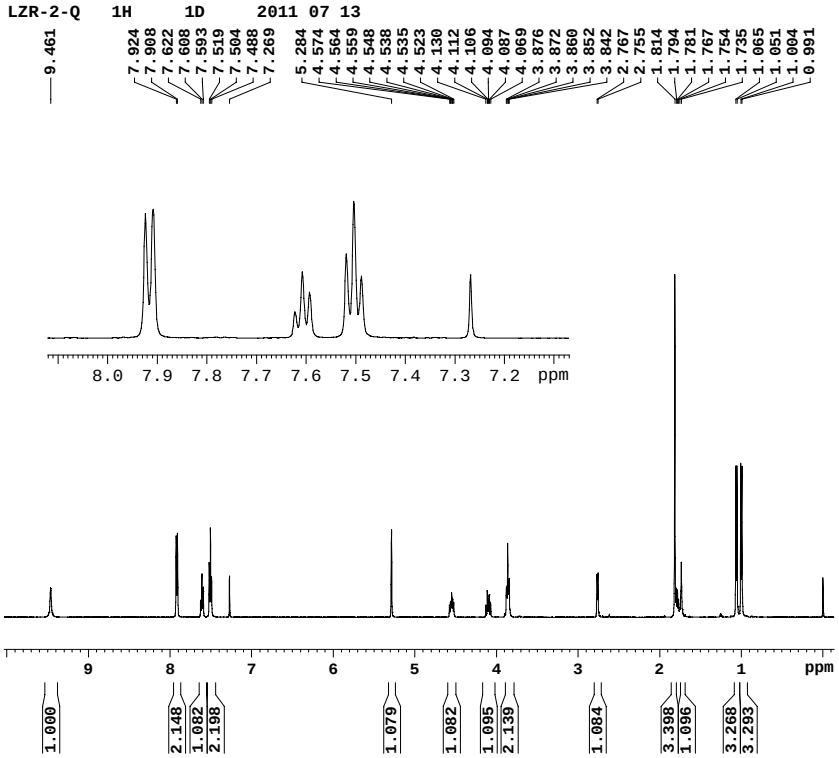




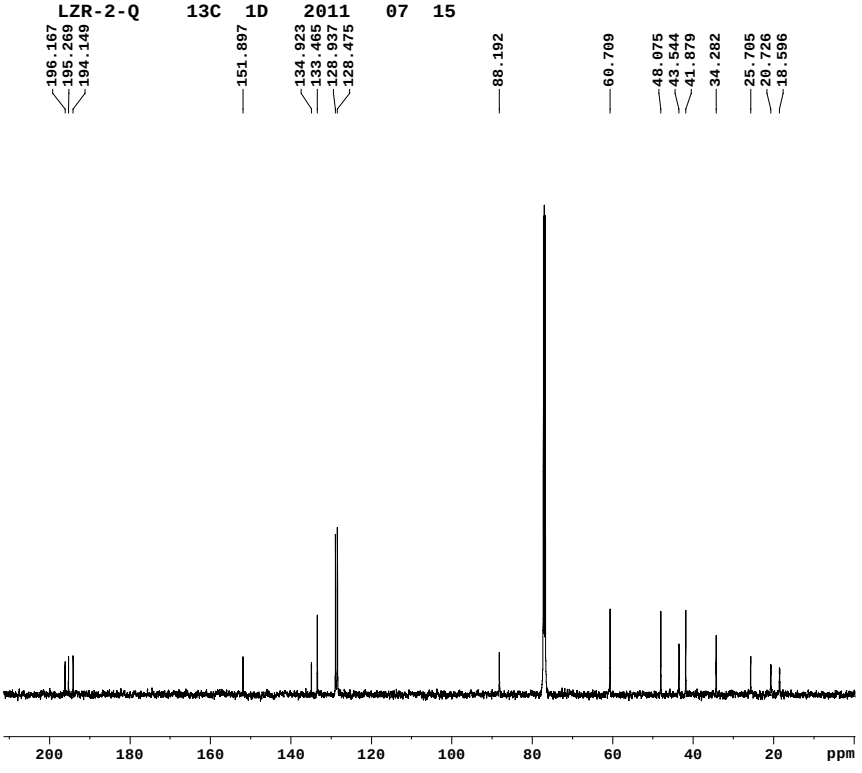

```
NAME              LZR-2-P
EXPNO                   2
PROCNO                  1
Date_            20110708
Time                10.16
INSTRUM             av500
PROBHD    5 mm PABBO BB-
PULPROG            zgpg30
TD                  65536
SOLVENT             CDCl3
NS                    552
DS                      2
SWH           32679.738 Hz
FIDRES            0.498653 Hz
AQ             1.0027661 sec
RG                  18400
DW                 15.300 usec
DE                  6.00 usec
TE                 299.7 K
D1            2.00000000 sec
d11           0.03000000 sec
DELTA         1.89999998 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                  13C
P1                  9.60 usec
PL1                  2.00 dB
SFO1         125.7464750 MHz

======== CHANNEL f2 ========
CPDPRG2           waltz16
NUC2                   1H
PCPD2               80.00 usec
PL2                  2.60 dB
PL12                17.66 dB
PL13                17.66 dB
SFO2          500.0355000 MHz
SI                  32768
SF            125.7326470 MHz
WDW                    EM
SSB                     0
LB                   6.00 Hz
GB                      0
PC                   2.00
```



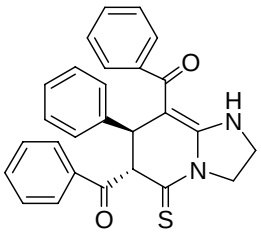
^{1}H and ^{13}C NMR spectra of **4l**



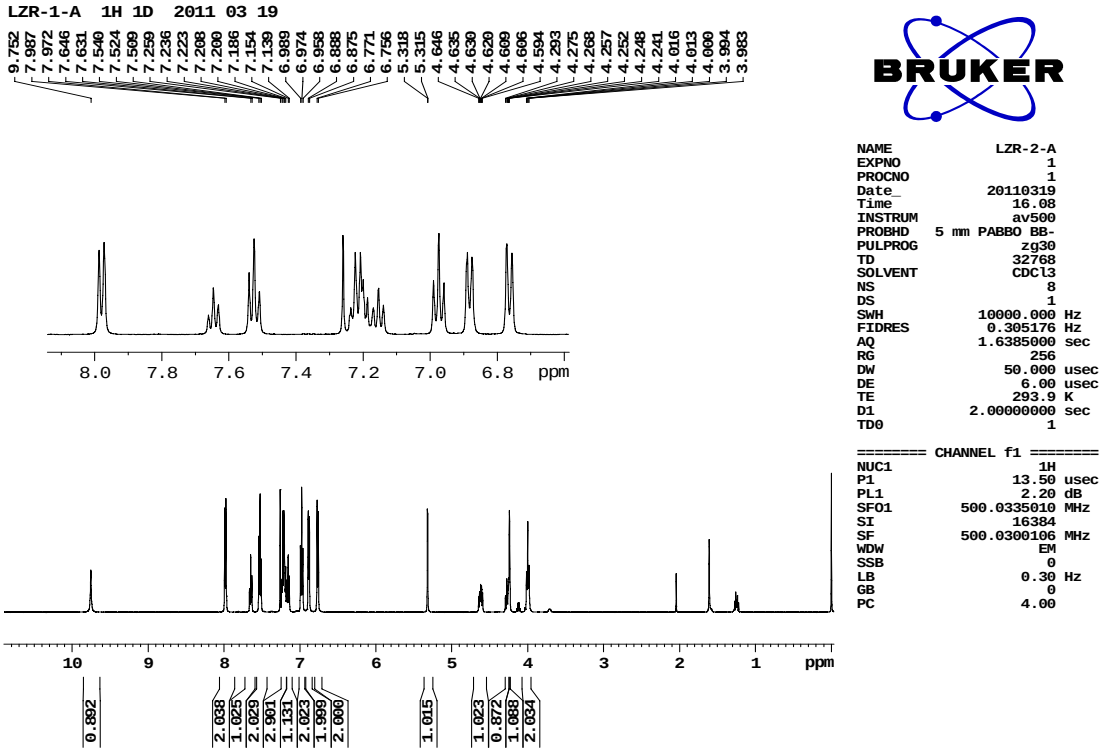

```
NAME            LZR-2-Q
EXPNO                 1
PROCNO                1
Date_          20110713
Time              19.33
INSTRUM           av500
PROBHD   5 mm PABBO BB-
PULPROG            zg30
TD                32768
SOLVENT           CDCl3
NS                    4
DS                    1
SWH           10000.000 Hz
FIDRES        0.305176 Hz
AQ            1.6385000 sec
RG                  287
DW               50.000 usec
DE                6.00 usec
TE               297.1 K
D1           2.00000000 sec
TD0                   1

======== CHANNEL f1 ========
NUC1                 1H
P1                13.50 usec
PL1                2.20 dB
SFO1         500.0335010 MHz
SI                16384
SF           500.0300060 MHz
WDW                  EM
SSB                   0
LB                0.30 Hz
GB                    0
PC                2.00
```




```
NAME            LZR-2-Q
EXPNO                 2
PROCNO                1
Date_          20110715
Time              9.53
INSTRUM           av500
PROBHD   5 mm PABBO BB-
PULPROG         zgpg30
TD                65536
SOLVENT           CDCl3
NS                  386
DS                    2
SWH           32679.738 Hz
FIDRES        0.498653 Hz
AQ            1.0027661 sec
RG                18400
DW               15.300 usec
DE                6.00 usec
TE               298.0 K
D1           2.00000000 sec
d11          0.03000000 sec
DELTA        1.89999998 sec
TD0                   1

======== CHANNEL f1 ========
NUC1                13C
P1                9.60 usec
PL1                2.00 dB
SFO1        125.7464750 MHz

======== CHANNEL f2 ========
CPDPRG2         waltz16
NUC2                 1H
PCPD2             80.00 usec
PL2                2.60 dB
PL12              17.66 dB
PL13              17.66 dB
SFO2         500.0355000 MHz
SI                32768
SF           125.7326470 MHz
WDW                  EM
SSB                   0
LB                6.00 Hz
GB                    0
PC                2.00
```

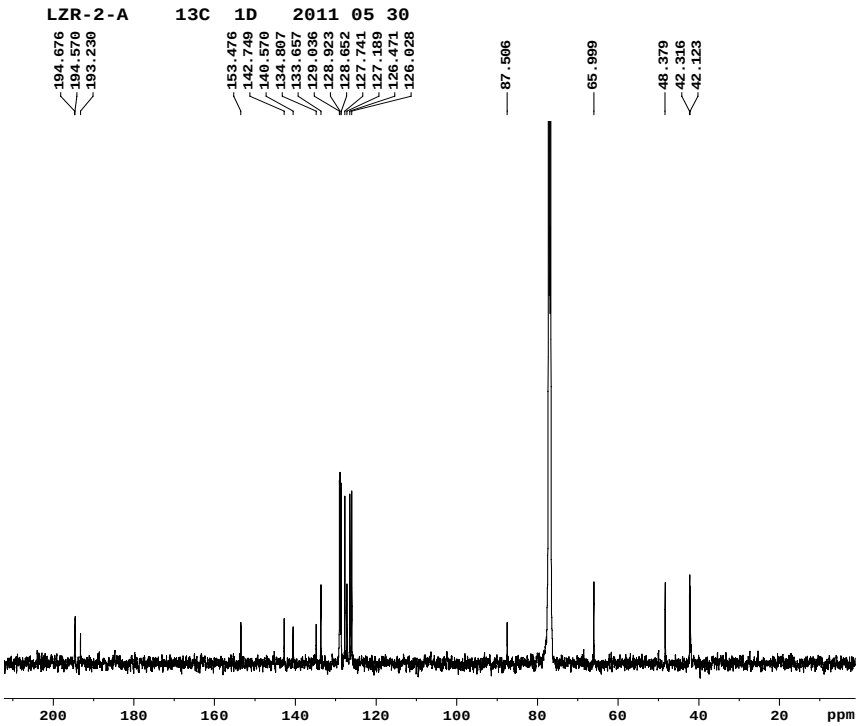


^{1}H and ^{13}C NMR spectra of **4m**



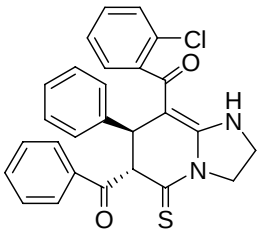
BRUKER

NAME	LZR-2-A
EXPNO	1
PROCNO	1
Date_	20110319
Time	16.08
INSTRUM	av500
PROBHD	5 mm PABBO BB-
PULPROG	zg30
TD	32768
SOLVENT	CDCl3
NS	8
DS	1
SWH	10000.000 Hz
FIDRES	0.305176 Hz
AQ	1.6385000 sec
RG	256
DW	50.000 usec
DE	6.00 usec
TE	293.9 K
D1	2.00000000 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	1H
P1	13.50 usec
PL1	2.20 dB
SFO1	500.0335010 MHz
SI	16384
SF	500.0300106 MHz
WDW	EM
SSB	0
LB	0.30 Hz
GB	0
PC	4.00

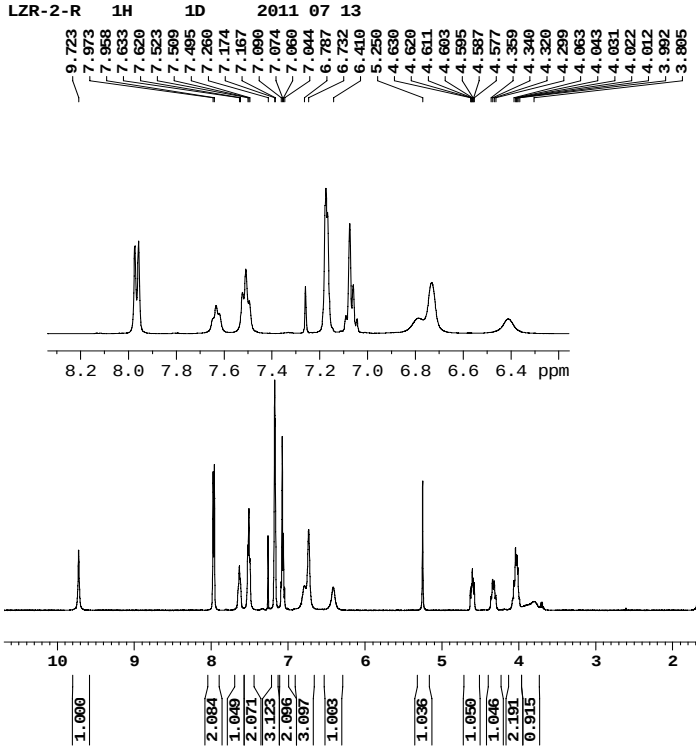


BRUKER

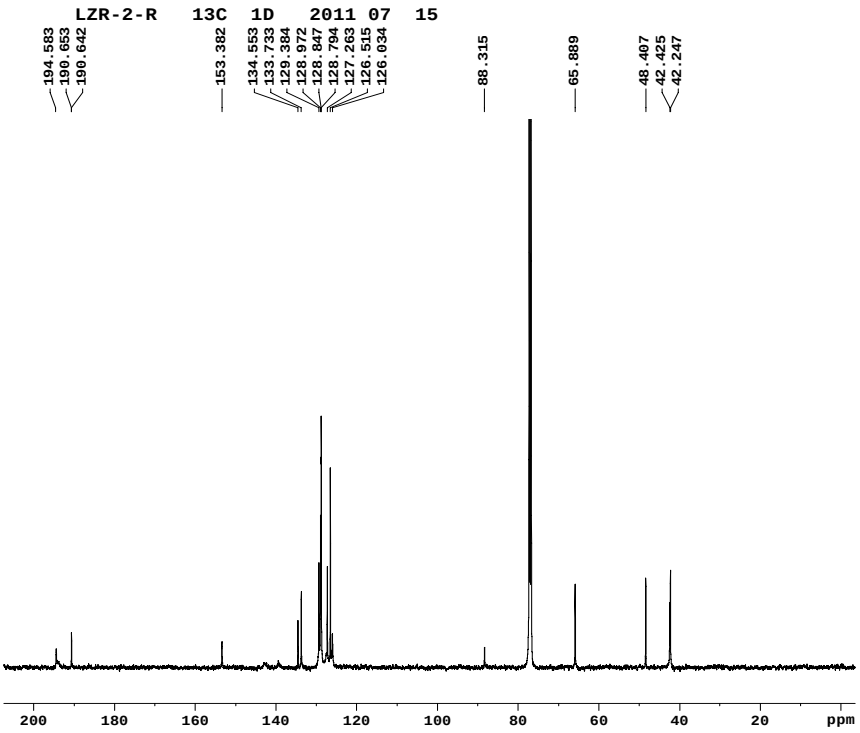
NAME	LZR-2-A
EXPNO	2
PROCNO	1
Date_	20110530
Time	17.49
INSTRUM	av500
PROBHD	5 mm PABBO BB-
PULPROG	zgpg30
TD	65536
SOLVENT	CDCl3
NS	1672
DS	2
SWH	32679.738 Hz
FIDRES	0.498653 Hz
AQ	1.0027661 sec
RG	16400
DW	15.300 usec
DE	6.00 usec
TE	298.6 K
D1	2.00000000 sec
d11	0.03000000 sec
DELTA	1.89999998 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	13C
P1	9.60 usec
PL1	2.00 dB
SFO1	125.7464750 MHz
======== CHANNEL f2 ========	
CPDPRG2	waltz16
NUC2	1H
PCPD2	80.00 usec
PL2	2.20 dB
PL12	17.66 dB
PL13	17.66 dB
SFO2	500.0355000 MHz
SI	32768
SF	125.7326470 MHz
WDW	EM
SSB	0
LB	6.00 Hz
GB	0
PC	2.00



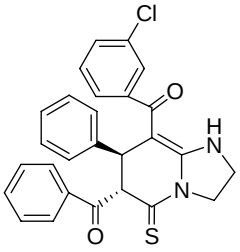
^{1}H and ^{13}C NMR spectra of **4n**



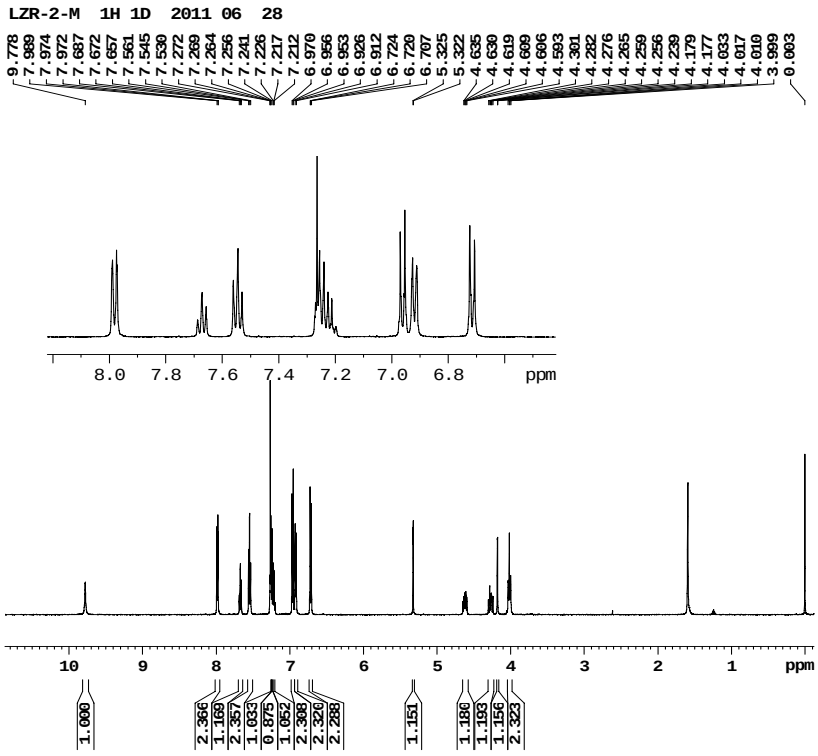
```
NAME            LZR-2-R
EXPNO                 1
PROCNO                1
Date_          20110713
Time              19.38
INSTRUM           av500
PROBHD   5 mm PABBO BB-
PULPROG            zg30
TD                32768
SOLVENT           CDCl3
NS                    4
DS                    1
SWH           10000.000 Hz
FIDRES        0.305176 Hz
AQ            1.6385000 sec
RG                  287
DW               50.000 usec
DE                6.00 usec
TE                297.1 K
D1           2.00000000 sec
TD0                   1

======== CHANNEL f1 ========
NUC1                 1H
P1                13.50 usec
PL1                2.20 dB
SFO1         500.0335010 MHz
SI                16384
SF           500.0300103 MHz
WDW                  EM
SSB                   0
LB                0.30 Hz
GB                    0
PC                2.00
```



```
NAME            LZR-2-R
EXPNO                 2
PROCNO                1
Date_          20110715
Time              12.21
INSTRUM           av500
PROBHD   5 mm PABBO BB-
PULPROG         zgpg30
TD                65536
SOLVENT           CDCl3
NS                 1210
DS                    2
SWH           32679.738 Hz
FIDRES        0.498653 Hz
AQ            1.0027661 sec
RG                18400
DW               15.300 usec
DE                6.00 usec
TE                298.9 K
D1           2.00000000 sec
d11          0.03000000 sec
DELTA        1.89999998 sec
TD0                   1

======== CHANNEL f1 ========
NUC1                13C
P1                9.60 usec
PL1               2.00 dB
SFO1         125.7464750 MHz

======== CHANNEL f2 ========
CPDPRG2         waltz16
NUC2                 1H
PCPD2            80.00 usec
PL2               2.60 dB
PL12             17.66 dB
PL13             17.66 dB
SFO2         500.0355000 MHz
SI                32768
SF           125.7326470 MHz
WDW                  EM
SSB                   0
LB                6.00 Hz
GB                    0
PC                2.00
```

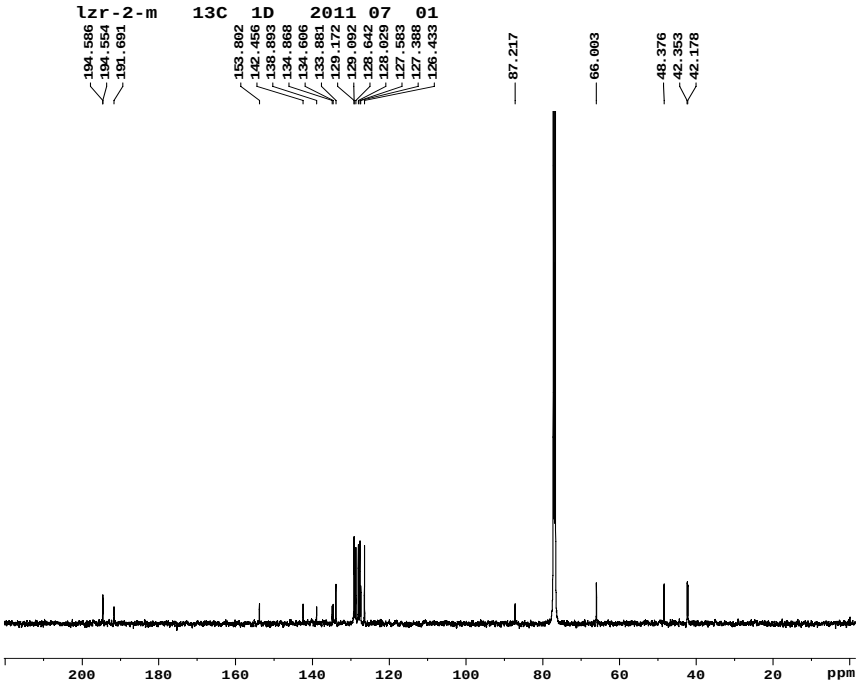


^{1}H and ^{13}C NMR spectra of **4o**



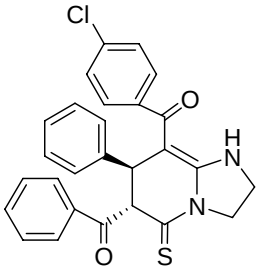
BRUKER

```
NAME              LZR-2-M
EXPNO                   1
PROCNO                  1
Date_            20110628
Time                19.13
INSTRUM             av500
PROBHD   5 mm PABBO BB-
PULPROG              zg30
TD                  32768
SOLVENT             CDCl3
NS                      4
DS                      1
SWH          10000.000 Hz
FIDRES        0.305176 Hz
AQ           1.6385000 sec
RG                    362
DW             50.000 usec
DE              6.00 usec
TE              296.2 K
D1           2.00000000 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                   1H
P1                 13.50 usec
PL1                 2.20 dB
SFO1         500.0335010 MHz
SI                  16384
SF           500.0300082 MHz
WDW                    EM
SSB                     0
LB                 0.30 Hz
GB                      0
PC                 2.00
```

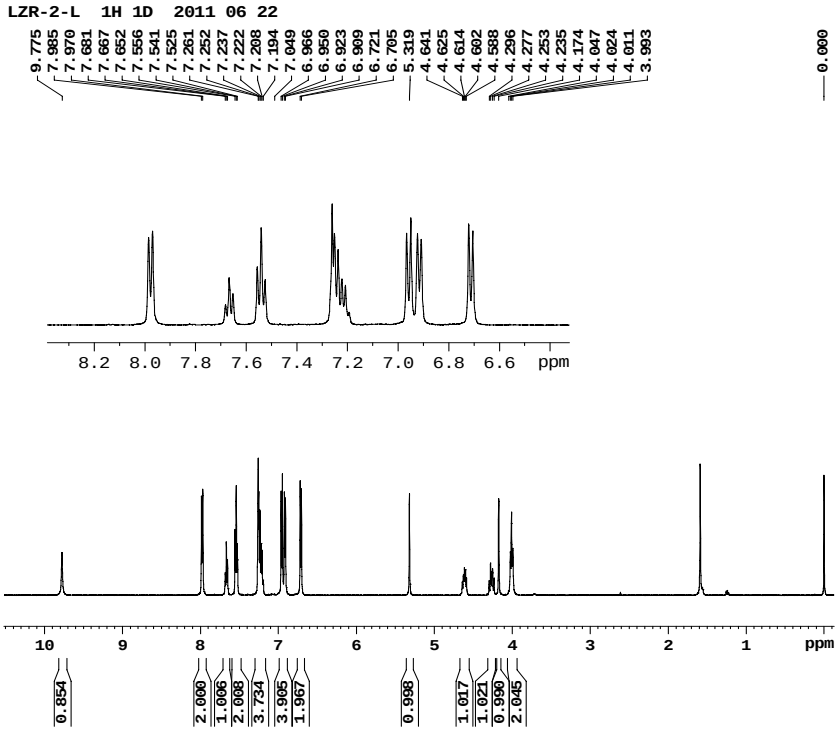


BRUKER

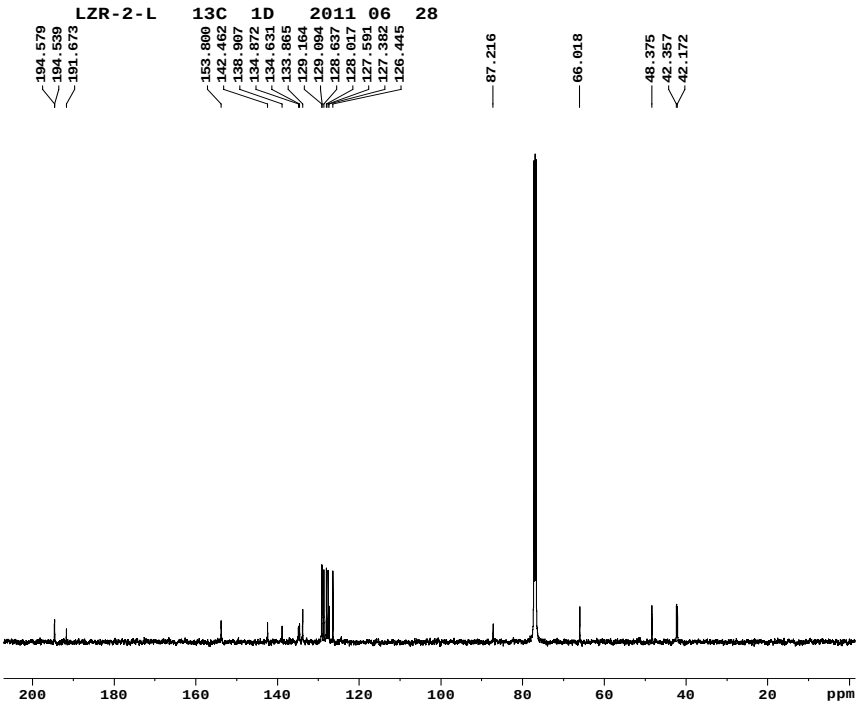
```
NAME              LZR-2-M
EXPNO                   2
PROCNO                  1
Date_            20110701
Time                19.39
INSTRUM             av500
PROBHD   5 mm PABBO BB-
PULPROG             zgpg30
TD                  65536
SOLVENT             CDCl3
NS                   1162
DS                      2
SWH          32679.738 Hz
FIDRES        0.498653 Hz
AQ           1.0027661 sec
RG                  10300
DW             15.300 usec
DE              6.00 usec
TE              296.9 K
D1           2.00000000 sec
d11          0.03000000 sec
DELTA        1.89999998 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                  13C
P1                 9.60 usec
PL1                 2.00 dB
SFO1         125.7464750 MHz

======== CHANNEL f2 ========
CPDPRG2          waltz16
NUC2                   1H
PCPD2              80.00 usec
PL2                 2.60 dB
PL12               17.66 dB
PL13               17.66 dB
SFO2         500.0355000 MHz
SI                  32768
SF           125.7326470 MHz
WDW                    EM
SSB                     0
LB                 6.00 Hz
GB                      0
PC                 1.00
```



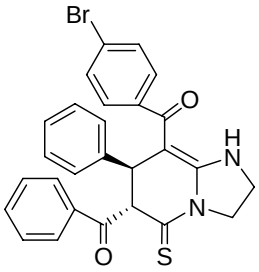
^{1}H and ^{13}C NMR spectra of **4p**



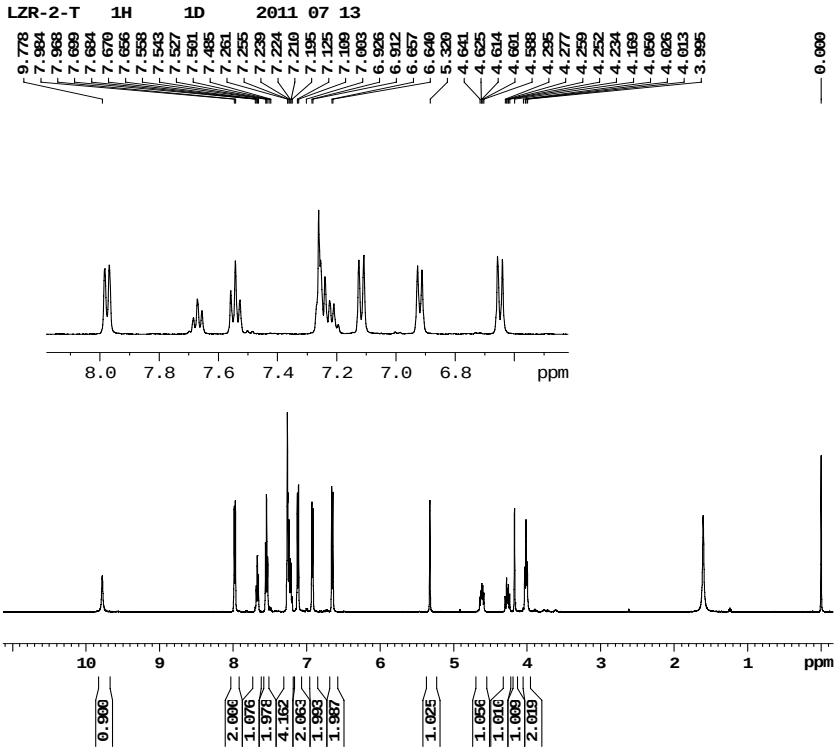
NAME	LZR-2-L
EXPNO	1
PROCNO	1
Date_	20110622
Time	16.58
INSTRUM	av500
PROBHD	5 mm PABBO BB-
PULPROG	zg30
TD	32768
SOLVENT	CDCl3
NS	4
DS	1
SWH	10000.000 Hz
FIDRES	0.305176 Hz
AQ	1.6385000 sec
RG	406
DW	50.000 usec
DE	6.00 usec
TE	297.6 K
D1	2.00000000 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	1H
P1	13.50 usec
PL1	2.20 dB
SFO1	500.0335010 MHz
SI	16384
SF	500.0300101 MHz
WDW	EM
SSB	0
LB	0.30 Hz
GB	0
PC	2.00



NAME	LZR-2-L
EXPNO	2
PROCNO	1
Date_	20110628
Time	15.18
INSTRUM	av500
PROBHD	5 mm PABBO BB-
PULPROG	zgpg30
TD	65536
SOLVENT	CDCl3
NS	453
DS	2
SWH	32679.738 Hz
FIDRES	0.498653 Hz
AQ	1.0027661 sec
RG	18400
DW	15.300 usec
DE	6.00 usec
TE	297.8 K
D1	2.00000000 sec
d11	0.03000000 sec
DELTA	1.89999998 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	13C
P1	9.60 usec
PL1	2.00 dB
SFO1	125.7464750 MHz
======== CHANNEL f2 ========	
CPDPRG2	waltz16
NUC2	1H
PCPD2	80.00 usec
PL2	2.60 dB
PL12	17.66 dB
PL13	17.66 dB
SFO2	500.0355000 MHz
SI	32768
SF	125.7326470 MHz
WDW	EM
SSB	0
LB	6.00 Hz
GB	0
PC	2.00

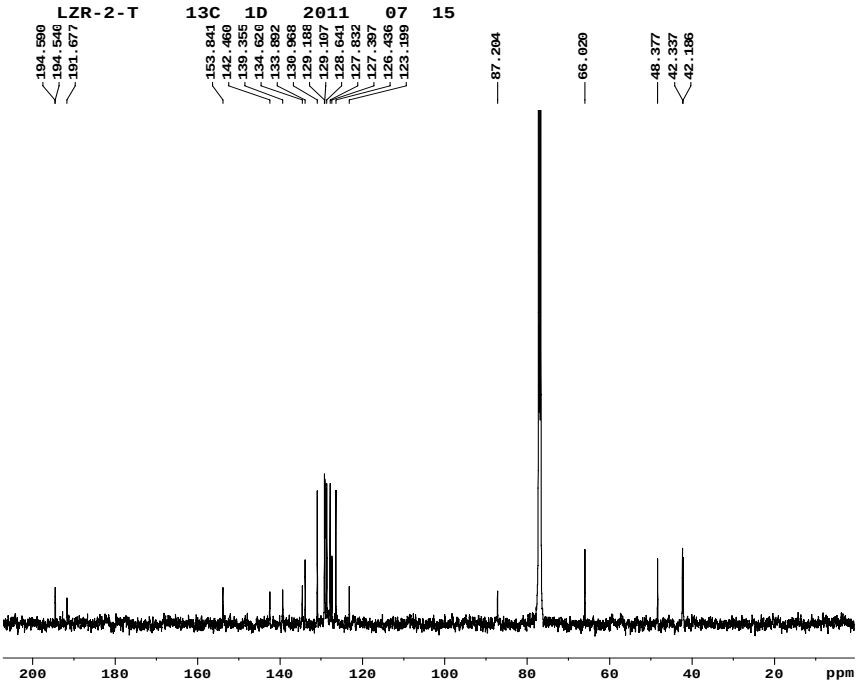


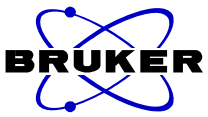
^{1}H and ^{13}C NMR spectra of **4q**



NAME LZR-2-T
EXPNO 1
PROCNO 1
Date_ 20110713
Time 19.42
INSTRUM av500
PROBHD 5 mm PABBO BB-
PULPROG zg30
TD 32768
SOLVENT CDCl3
NS 4
DS 1
SWH 10000.000 Hz
FIDRES 0.305176 Hz
AQ 1.6385000 sec
RG 645
DW 50.000 usec
DE 6.00 usec
TE 297.1 K
D1 2.00000000 sec
TD0 1

======== CHANNEL f1 ========
NUC1 1H
P1 13.50 usec
PL1 2.20 dB
SFO1 500.0335010 MHz
SI 16384
SF 500.0300098 MHz
WDW EM
SSB 0
LB 0.30 Hz
GB 0
PC 2.00

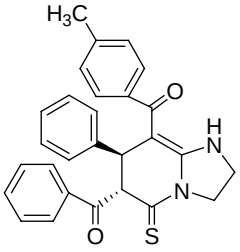




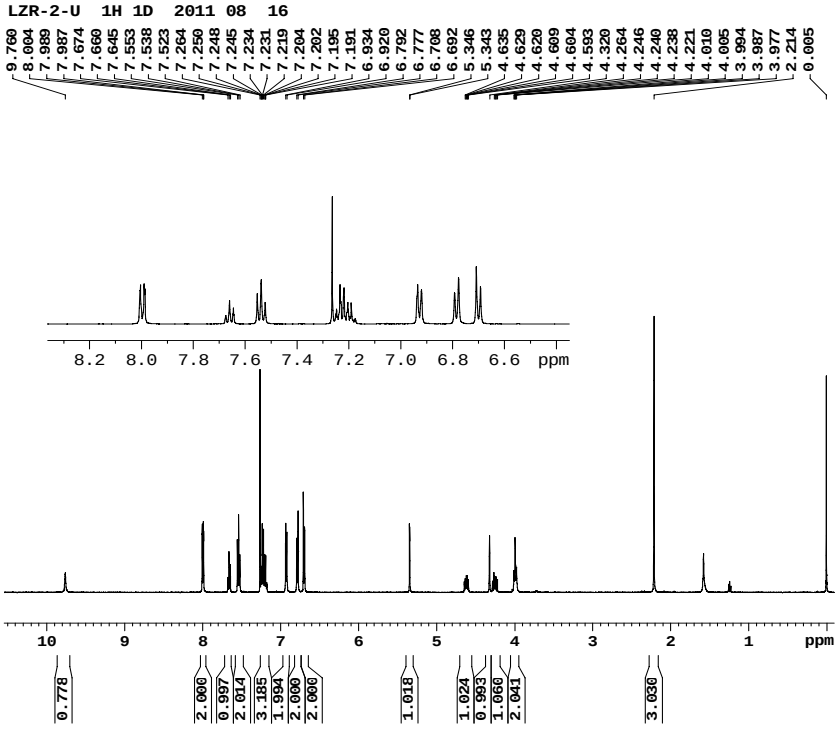
NAME LZR-2-T
EXPNO 2
PROCNO 1
Date_ 20110715
Time 10.28
INSTRUM av500
PROBHD 5 mm PABBO BB-
PULPROG zgpg30
TD 65536
SOLVENT CDCl3
NS 933
DS 2
SWH 32679.738 Hz
FIDRES 0.498653 Hz
AQ 1.0027661 sec
RG 18400
DW 15.300 usec
DE 6.00 usec
TE 298.6 K
D1 2.00000000 sec
d11 0.03000000 sec
DELTA 1.89999998 sec
TD0 1

======== CHANNEL f1 ========
NUC1 13C
P1 9.60 usec
PL1 2.00 dB
SFO1 125.7464750 MHz

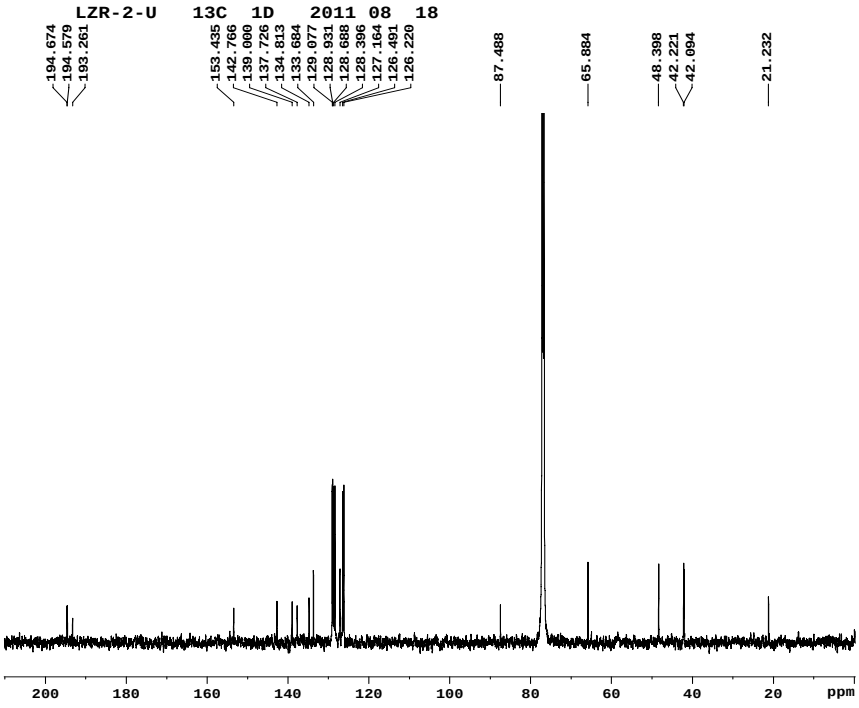
======== CHANNEL f2 ========
CPDPRG2 waltz16
NUC2 1H
PCPD2 80.00 usec
PL2 2.60 dB
PL12 17.66 dB
PL13 17.66 dB
SFO2 500.0355000 MHz
SI 32768
SF 125.7326470 MHz
WDW EM
SSB 0
LB 6.00 Hz
GB 0
PC 2.00

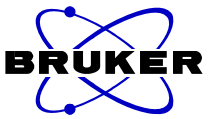


^{1}H and ^{13}C NMR spectra of **4r**

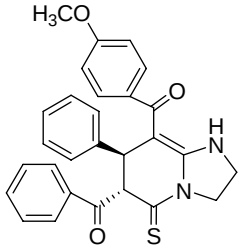


NAME	LZR-2-U
EXPNO	1
PROCNO	1
Date_	20110816
Time	16.57
INSTRUM	av500
PROBHD	5 mm PABBO BB-
PULPROG	zg30
TD	32768
SOLVENT	CDCl3
NS	4
DS	1
SWH	10000.000 Hz
FIDRES	0.305176 Hz
AQ	1.6385000 sec
RG	362
DW	50.000 usec
DE	6.00 usec
TE	297.7 K
D1	2.00000000 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	1H
P1	13.50 usec
PL1	2.20 dB
SFO1	500.0335010 MHz
SI	16384
SF	500.0300082 MHz
WDW	EM
SSB	0
LB	0.30 Hz
GB	0
PC	2.00

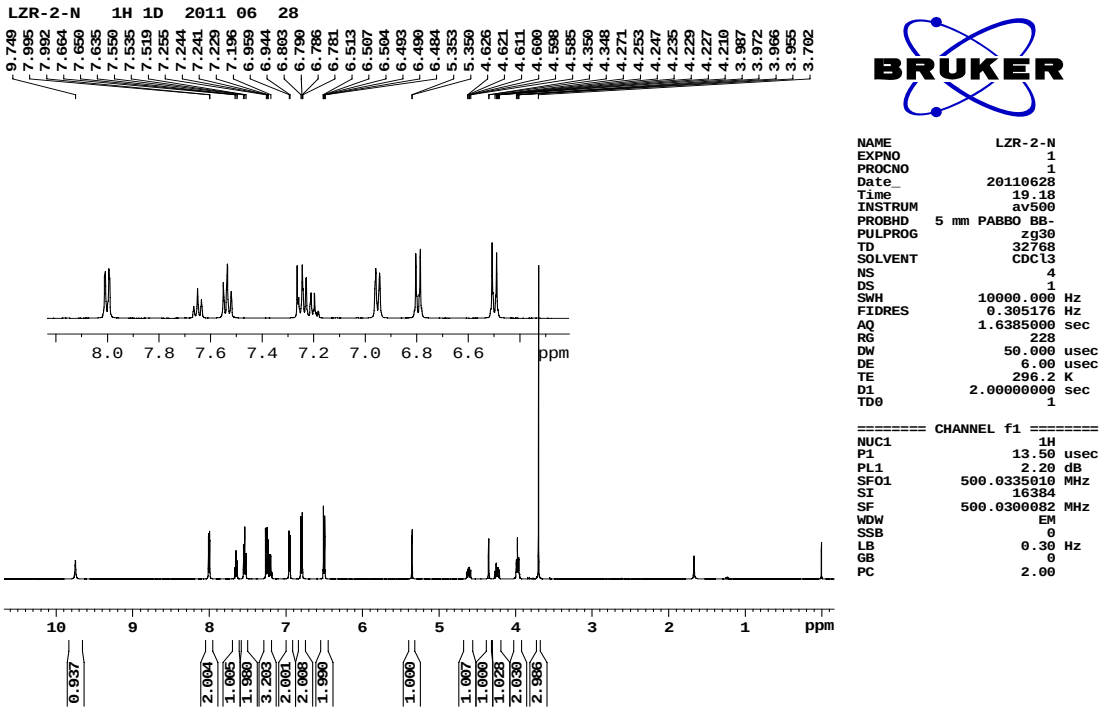




NAME	LZR-2-U
EXPNO	2
PROCNO	1
Date_	20110818
Time	10.23
INSTRUM	av500
PROBHD	5 mm PABBO BB-
PULPROG	zgpg30
TD	65536
SOLVENT	CDCl3
NS	1501
DS	2
SWH	32679.738 Hz
FIDRES	0.498653 Hz
AQ	1.0027661 sec
RG	18400
DW	15.300 usec
DE	6.00 usec
TE	299.1 K
D1	2.00000000 sec
d11	0.03000000 sec
DELTA	1.89999998 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	13C
P1	9.60 usec
PL1	2.00 dB
SFO1	125.7464750 MHz
======== CHANNEL f2 ========	
CPDPRG2	waltz16
NUC2	1H
PCPD2	80.00 usec
PL2	2.60 dB
PL12	17.66 dB
PL13	17.66 dB
SFO2	500.0355000 MHz
SI	32768
SF	125.7326470 MHz
WDW	EM
SSB	0
LB	6.00 Hz
GB	0
PC	2.00

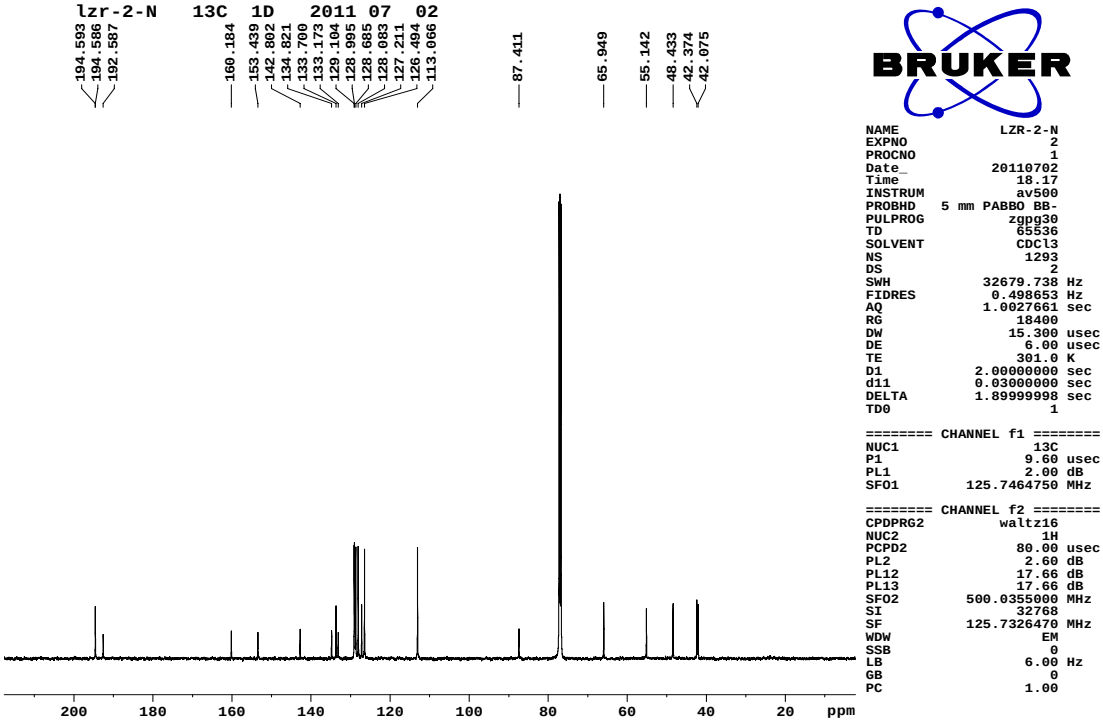


^{1}H and ^{13}C NMR spectra of **4s**



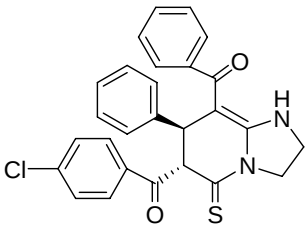
BRUKER

NAME	LZR-2-N
EXPNO	1
PROCNO	1
Date_	20110628
Time	19.18
INSTRUM	av500
PROBHD	5 mm PABBO BB-
PULPROG	zg30
TD	32768
SOLVENT	CDCl3
NS	4
DS	1
SWH	10000.000 Hz
FIDRES	0.305176 Hz
AQ	1.6385000 sec
RG	228
DW	50.000 usec
DE	6.00 usec
TE	296.2 K
D1	2.00000000 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	1H
P1	13.50 usec
PL1	2.20 dB
SFO1	500.0335010 MHz
SI	16384
SF	500.0300082 MHz
WDW	EM
SSB	0
LB	0.30 Hz
GB	0
PC	2.00

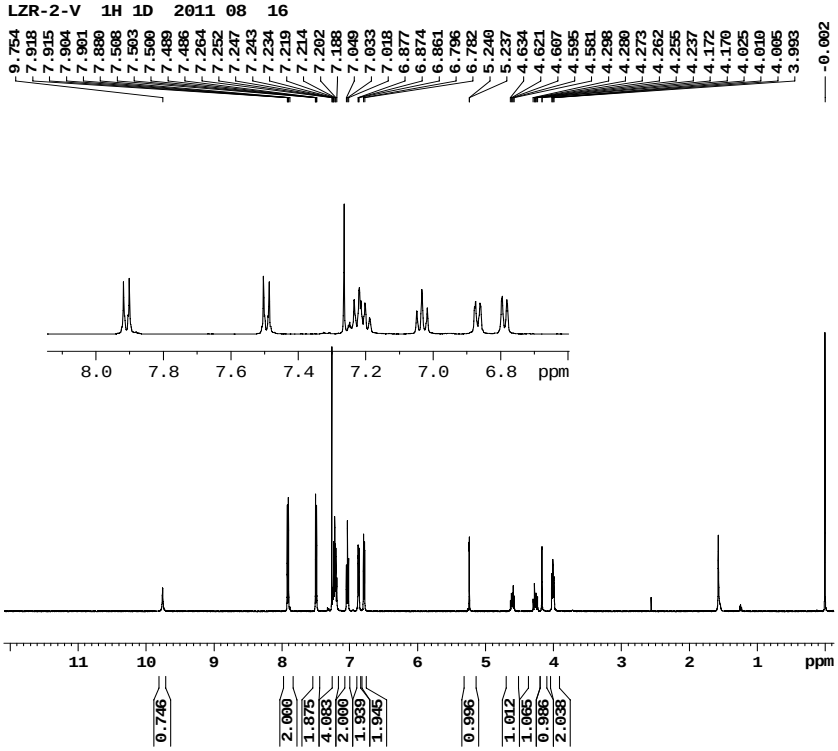


BRUKER

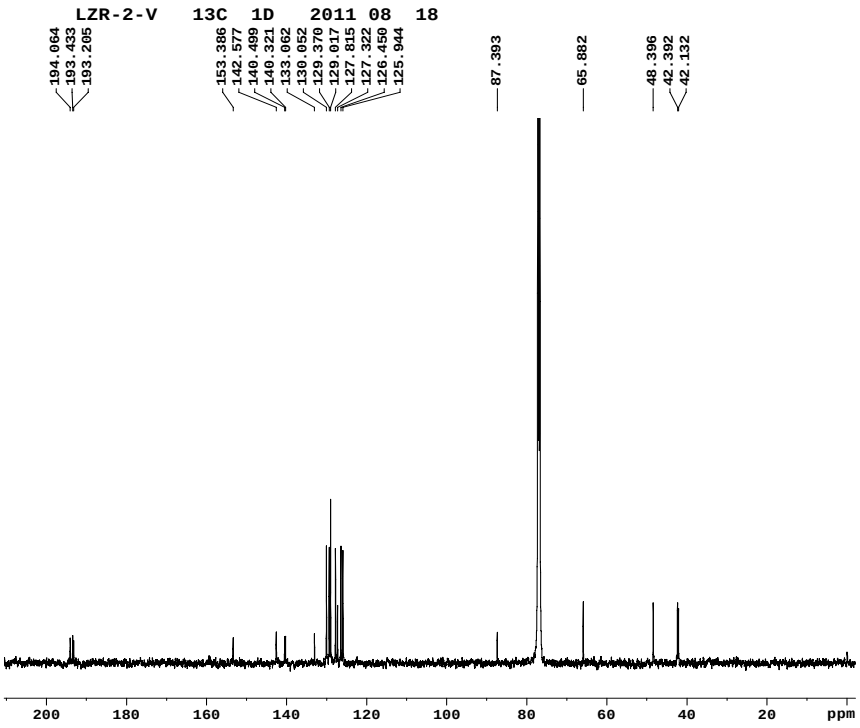
NAME	LZR-2-N
EXPNO	2
PROCNO	1
Date_	20110702
Time	18.17
INSTRUM	av500
PROBHD	5 mm PABBO BB-
PULPROG	zgpg30
TD	65536
SOLVENT	CDCl3
NS	1293
DS	2
SWH	32679.738 Hz
FIDRES	0.498653 Hz
AQ	1.0027661 sec
RG	18400
DW	15.300 usec
DE	6.00 usec
TE	301.0 K
D1	2.00000000 sec
d11	0.03000000 sec
DELTA	1.89999998 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	13C
P1	9.60 usec
PL1	2.00 dB
SFO1	125.7464750 MHz
======== CHANNEL f2 ========	
CPDPRG2	waltz16
NUC2	1H
PCPD2	80.00 usec
PL2	2.60 dB
PL12	17.66 dB
PL13	17.66 dB
SFO2	500.0355000 MHz
SI	32768
SF	125.7326470 MHz
WDW	EM
SSB	0
LB	6.00 Hz
GB	0
PC	1.00

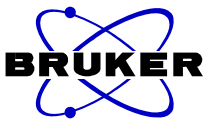


^{1}H and ^{13}C NMR spectra of **4t**

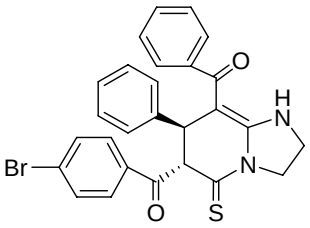


```
NAME              LZR-2-V
EXPNO                   1
PROCNO                  1
Date_            20110816
Time                17.01
INSTRUM             av500
PROBHD   5 mm PABBO BB-
PULPROG              zg30
TD                  32768
SOLVENT             CDCl3
NS                      4
DS                      1
SWH          10000.000 Hz
FIDRES           0.305176 Hz
AQ            1.6385000 sec
RG                    362
DW               50.000 usec
DE                6.00 usec
TE                297.6 K
D1           2.00000000 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                   1H
P1                 13.50 usec
PL1                 2.20 dB
SFO1         500.0335010 MHz
SI                  16384
SF           500.0300082 MHz
WDW                    EM
SSB                     0
LB                 0.30 Hz
GB                      0
PC                 2.00
```

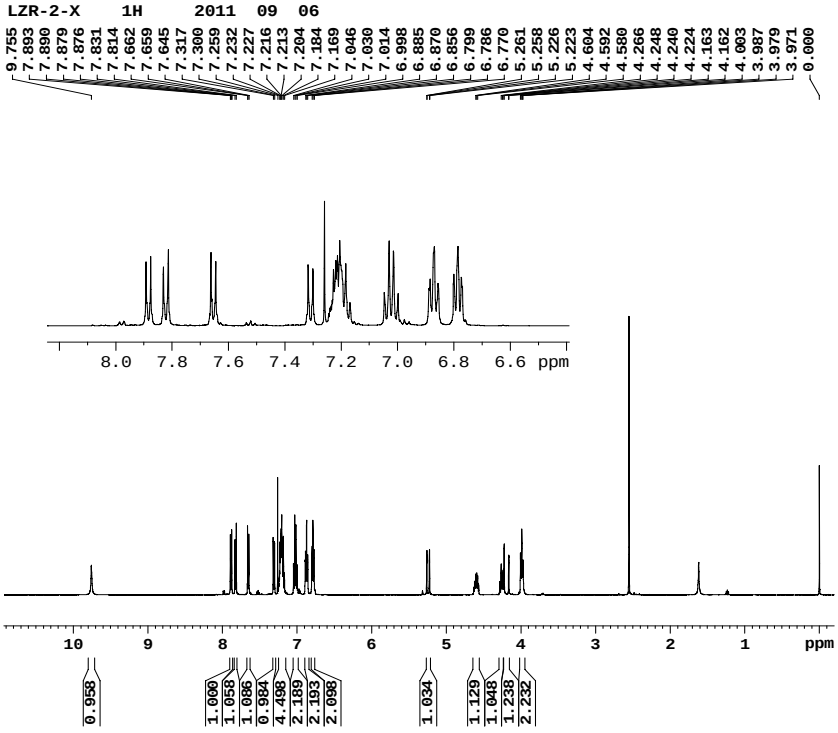




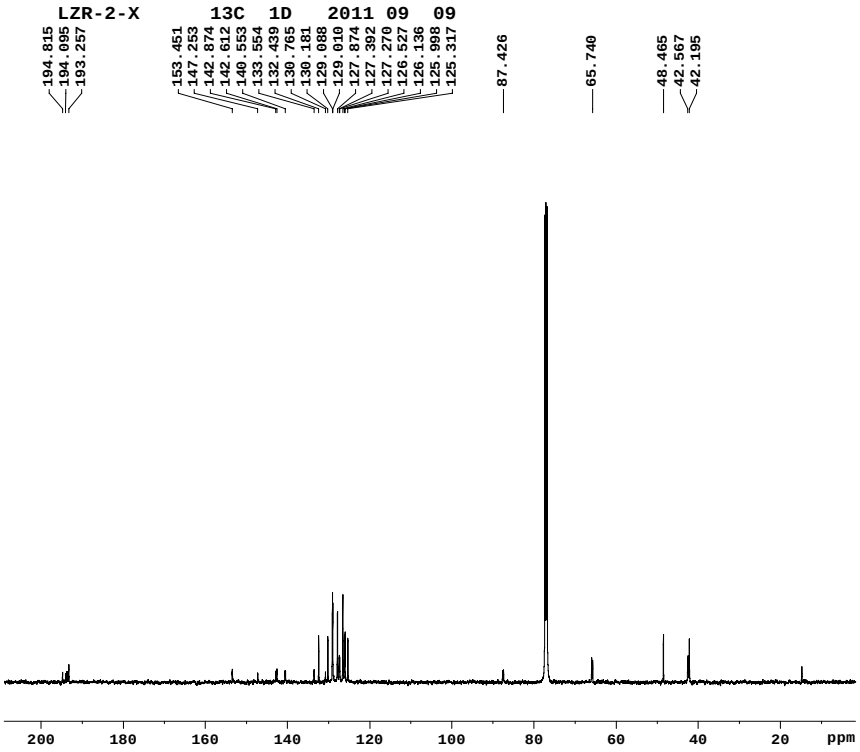
```
NAME              LZR-2-V
EXPNO                   2
PROCNO                  1
Date_            20110818
Time                15.22
INSTRUM             av500
PROBHD   5 mm PABBO BB-
PULPROG            zgpg30
TD                  65536
SOLVENT             CDCl3
NS                   2334
DS                      2
SWH          32679.738 Hz
FIDRES           0.498653 Hz
AQ            1.0027661 sec
RG                  18400
DW               15.300 usec
DE                6.00 usec
TE                299.5 K
D1           2.00000000 sec
d11          0.03000000 sec
DELTA        1.89999998 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                  13C
P1                 9.60 usec
PL1                 2.00 dB
SFO1         125.7464750 MHz

======== CHANNEL f2 ========
CPDPRG2           waltz16
NUC2                   1H
PCPD2              80.00 usec
PL2                 2.60 dB
PL12               17.66 dB
PL13               17.66 dB
SFO2         500.0355000 MHz
SI                  32768
SF           125.7326470 MHz
WDW                    EM
SSB                     0
LB                 6.00 Hz
GB                      0
PC                 2.00
```

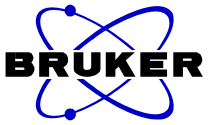


^{1}H and ^{13}C NMR spectra of **4u**

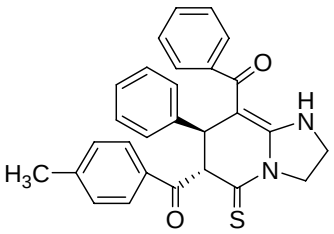


NAME	LZR-2-X
EXPNO	1
PROCNO	1
Date_	20110906
Time	16.50
INSTRUM	av500
PROBHD	5 mm PABBO BB-
PULPROG	zg30
TD	32768
SOLVENT	CDCl3
NS	8
DS	1
SWH	10000.000 Hz
FIDRES	0.305176 Hz
AQ	1.6385000 sec
RG	256
DW	50.000 usec
DE	6.00 usec
TE	297.9 K
D1	2.00000000 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	1H
P1	13.50 usec
PL1	2.20 dB
SFO1	500.0335010 MHz
SI	16384
SF	500.0300107 MHz
WDW	EM
SSB	0
LB	0.30 Hz
GB	0
PC	2.00

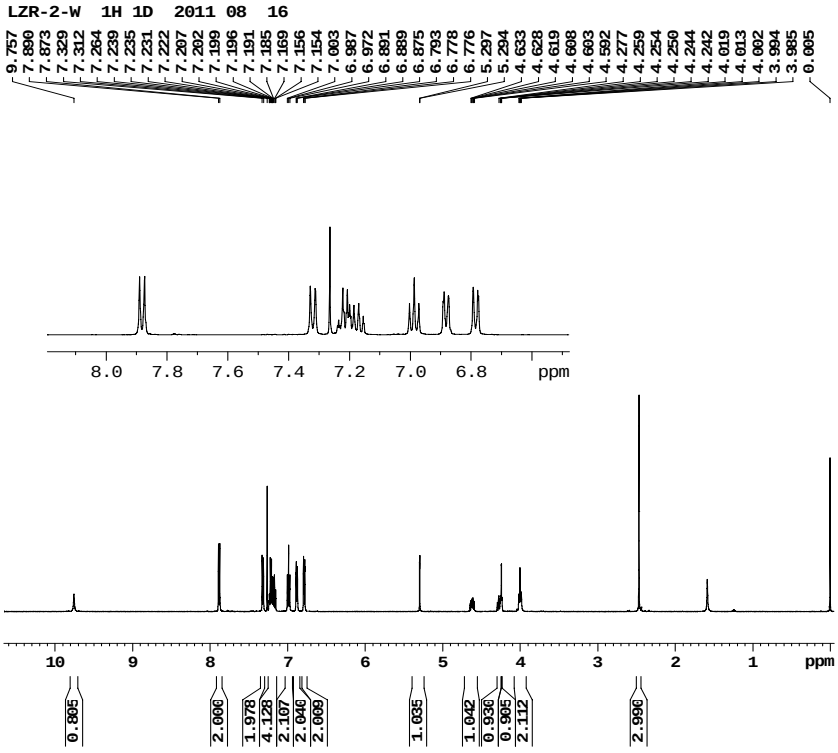




NAME	LZR-2-X
EXPNO	2
PROCNO	1
Date_	20110909
Time	18.05
INSTRUM	av500
PROBHD	5 mm PABBO BB-
PULPROG	zgpg30
TD	65536
SOLVENT	CDCl3
NS	1613
DS	2
SWH	32679.738 Hz
FIDRES	0.498653 Hz
AQ	1.0027661 sec
RG	18400
DW	15.300 usec
DE	6.00 usec
TE	299.1 K
D1	2.00000000 sec
d11	0.03000000 sec
DELTA	1.89999998 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	13C
P1	9.60 usec
PL1	2.00 dB
SFO1	125.7464750 MHz
======== CHANNEL f2 ========	
CPDPRG2	waltz16
NUC2	1H
PCPD2	80.00 usec
PL2	2.60 dB
PL12	17.66 dB
PL13	17.66 dB
SFO2	500.0355000 MHz
SI	32768
SF	125.7326387 MHz
WDW	EM
SSB	0
LB	6.00 Hz
GB	0
PC	2.00

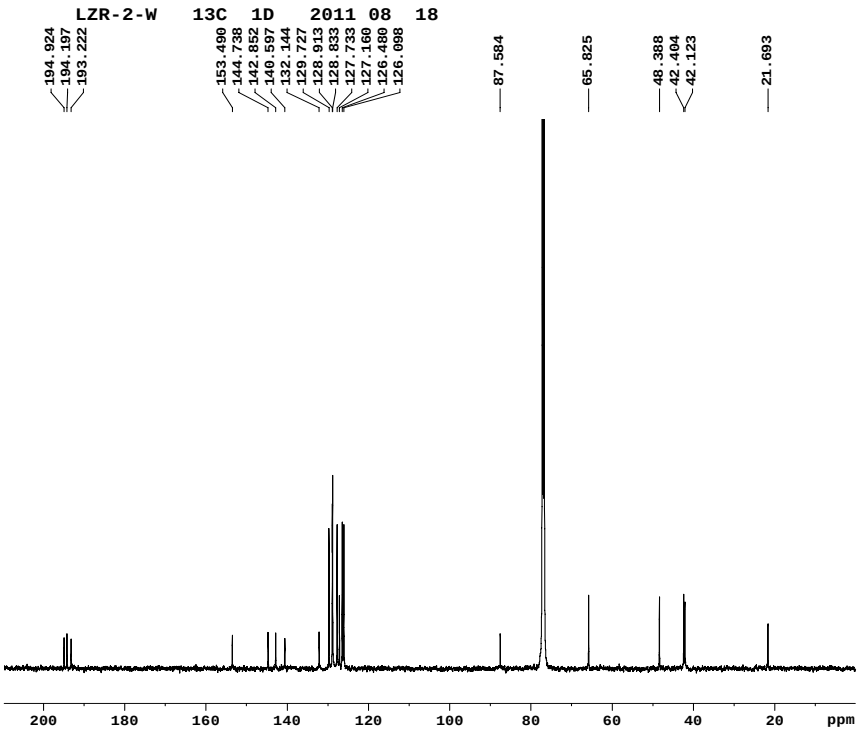


^{1}H and ^{13}C NMR spectra of **4v**



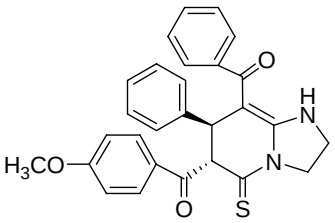
BRUKER

```
NAME              LZR-2-W
EXPNO                   1
PROCNO                  1
Date_            20110816
Time                17.05
INSTRUM             av500
PROBHD   5 mm PABBO BB-
PULPROG              zg30
TD                  32768
SOLVENT             CDCl3
NS                      4
DS                      1
SWH             10000.000 Hz
FIDRES              0.305176 Hz
AQ               1.6385000 sec
RG                    362
DW                 50.000 usec
DE                  6.00 usec
TE                 297.6 K
D1              2.00000000 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                   1H
P1                  13.50 usec
PL1                  2.20 dB
SFO1           500.0335010 MHz
SI                  16384
SF             500.0300082 MHz
WDW                    EM
SSB                     0
LB                   0.30 Hz
GB                      0
PC                   2.00
```

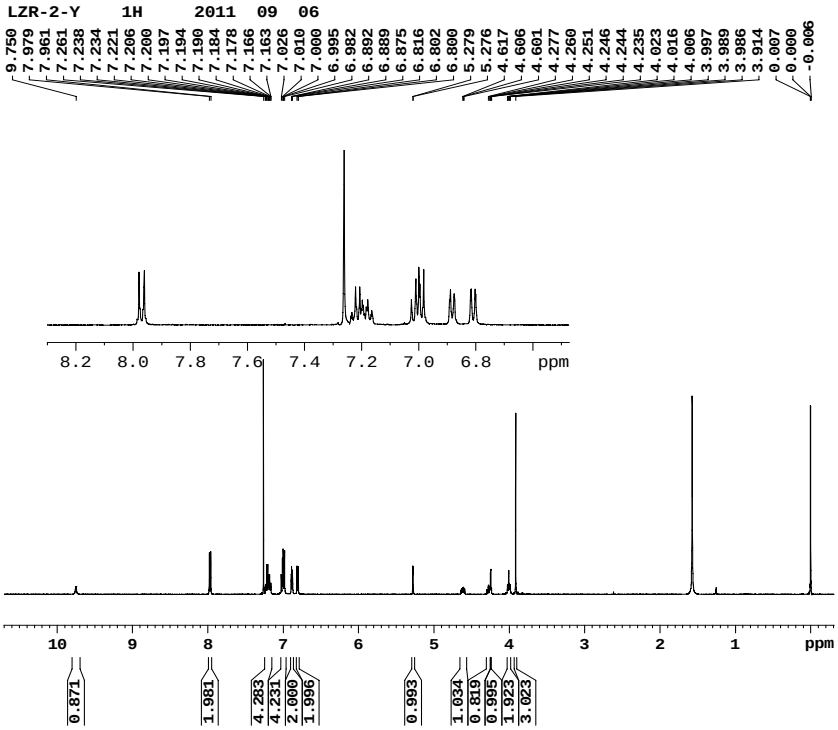


BRUKER

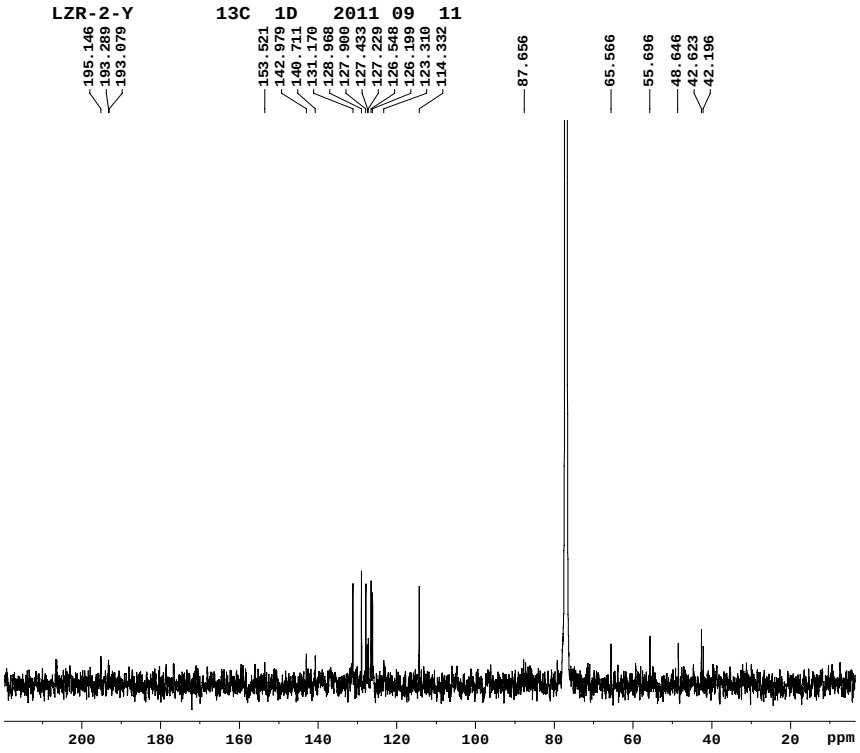
```
NAME              LZR-2-W
EXPNO                   2
PROCNO                  1
Date_            20110818
Time                15.11
INSTRUM             av500
PROBHD   5 mm PABBO BB-
PULPROG           zgpg30
TD                  65536
SOLVENT             CDCl3
NS                   4115
DS                      2
SWH             32679.738 Hz
FIDRES              0.498653 Hz
AQ               1.0027661 sec
RG                  18400
DW                 15.300 usec
DE                  6.00 usec
TE                 299.7 K
D1              2.00000000 sec
d11             0.03000000 sec
DELTA           1.89999998 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                   13C
P1                   9.60 usec
PL1                  2.00 dB
SFO1           125.7464750 MHz

======== CHANNEL f2 ========
CPDPRG2             waltz16
NUC2                   1H
PCPD2               80.00 usec
PL2                  2.60 dB
PL12                17.66 dB
PL13                17.66 dB
SFO2           500.0355000 MHz
SI                  32768
SF             125.7326470 MHz
WDW                    EM
SSB                     0
LB                   6.00 Hz
GB                      0
PC                   2.00
```

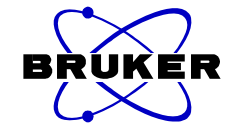


^{1}H and ^{13}C NMR spectra of **4w**

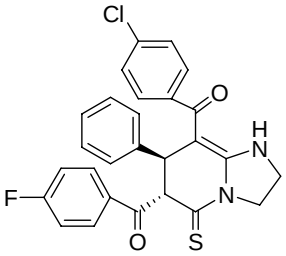


NAME	LZR-2-Y
EXPNO	1
PROCNO	1
Date_	20110906
Time	17.00
INSTRUM	av500
PROBHD	5 mm PABBO BB-
PULPROG	zg30
TD	32768
SOLVENT	CDCl3
NS	8
DS	1
SWH	10000.000 Hz
FIDRES	0.305176 Hz
AQ	1.6385000 sec
RG	575
DW	50.000 usec
DE	6.00 usec
TE	297.9 K
D1	2.00000000 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	1H
P1	13.50 usec
PL1	2.20 dB
SFO1	500.0335010 MHz
SI	16384
SF	500.0300097 MHz
WDW	EM
SSB	0
LB	0.30 Hz
GB	0
PC	2.00

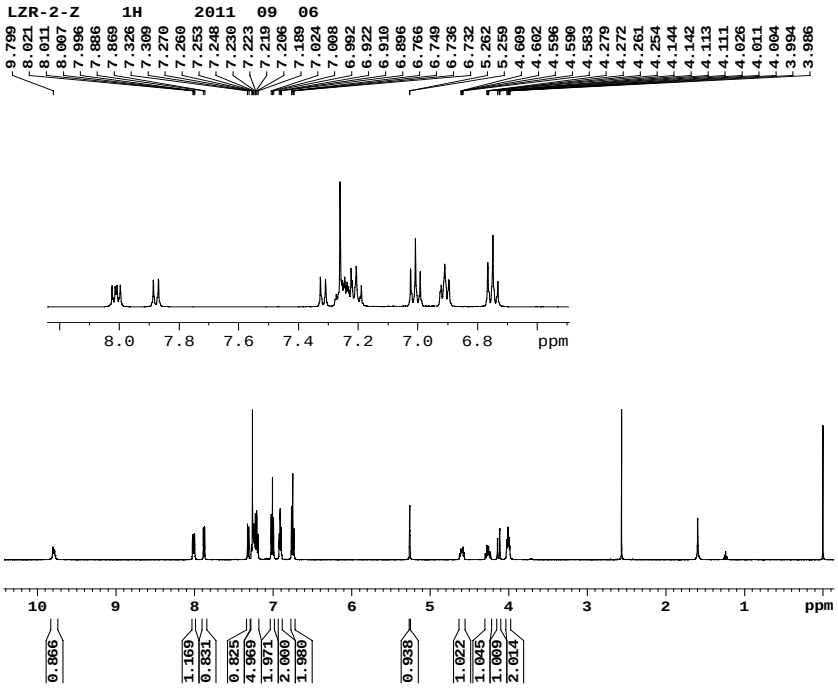




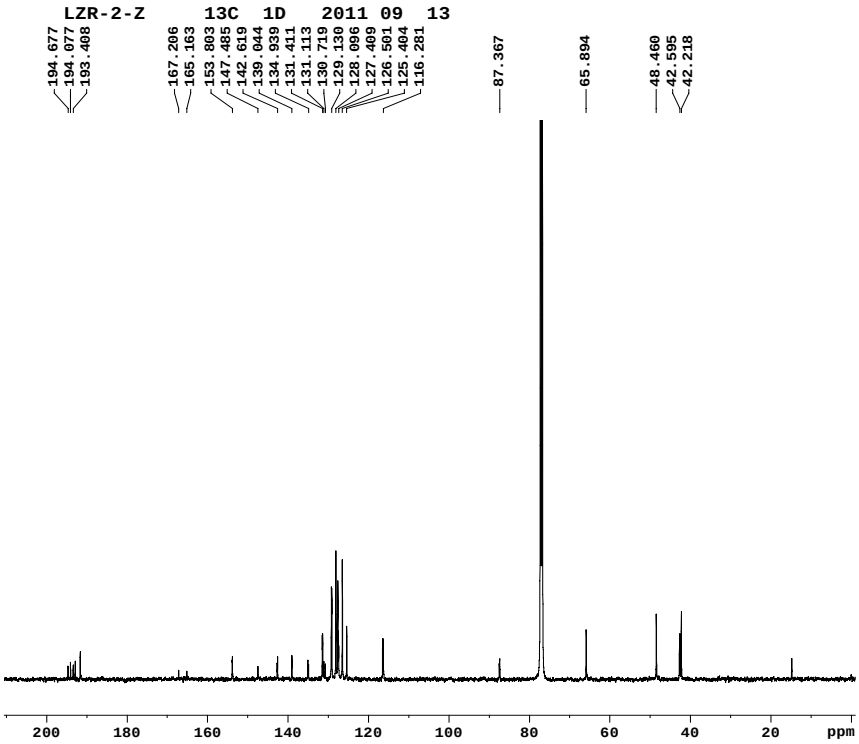
NAME	LZR-2-Y
EXPNO	2
PROCNO	1
Date_	20110911
Time	18.47
INSTRUM	av500
PROBHD	5 mm PABBO BB-
PULPROG	zgpg30
TD	65536
SOLVENT	CDCl3
NS	2739
DS	2
SWH	32679.738 Hz
FIDRES	0.498653 Hz
AQ	1.0027661 sec
RG	18400
DW	15.300 usec
DE	6.00 usec
TE	299.3 K
D1	2.00000000 sec
d11	0.03000000 sec
DELTA	1.89999998 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	13C
P1	9.60 usec
PL1	2.00 dB
SFO1	125.7464750 MHz
======== CHANNEL f2 ========	
CPDPRG2	waltz16
NUC2	1H
PCPD2	80.00 usec
PL2	2.60 dB
PL12	17.66 dB
PL13	17.66 dB
SFO2	500.0355000 MHz
SI	32768
SF	125.7326387 MHz
WDW	EM
SSB	0
LB	6.00 Hz
GB	0
PC	1.00



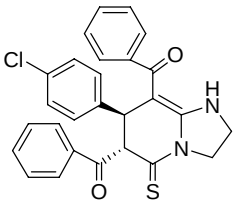
^{1}H and ^{13}C NMR spectra of **4x**



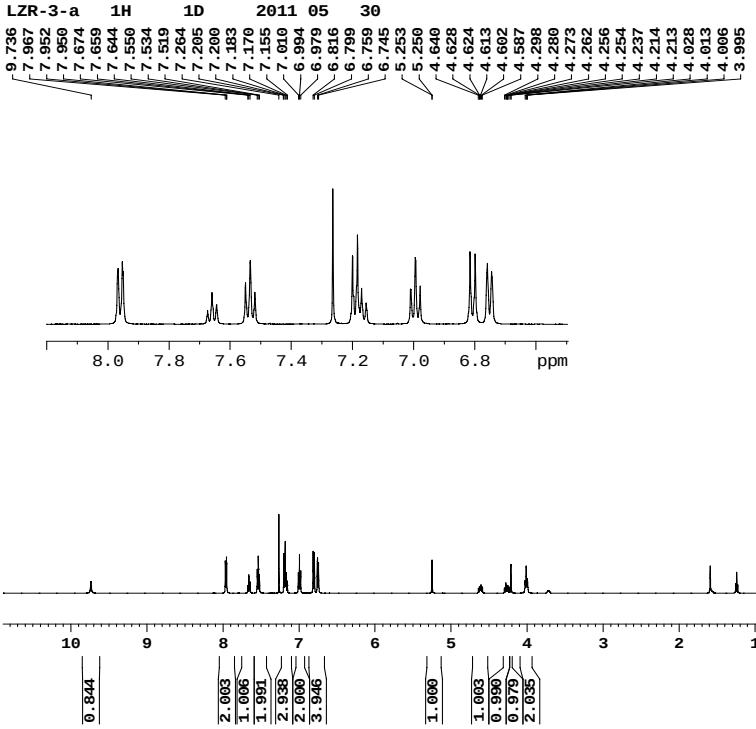
```
NAME                 LZR-2-Z
EXPNO                      1
PROCNO                     1
Date_               20110906
Time                   16.55
INSTRUM                av500
PROBHD    5 mm PABBO BB-
PULPROG                 zg30
TD                     32768
SOLVENT                CDCl3
NS                         8
DS                         1
SWH              10000.000 Hz
FIDRES               0.305176 Hz
AQ                1.6385000 sec
RG                       256
DW                    50.000 usec
DE                     6.00 usec
TE                    297.9 K
D1                2.00000000 sec
TD0                        1

======== CHANNEL f1 ========
NUC1                      1H
P1                     13.50 usec
PL1                     2.20 dB
SFO1            500.0335010 MHz
SI                       16384
SF              500.0300101 MHz
WDW                       EM
SSB                        0
LB                      0.30 Hz
GB                         0
PC                      2.00
```



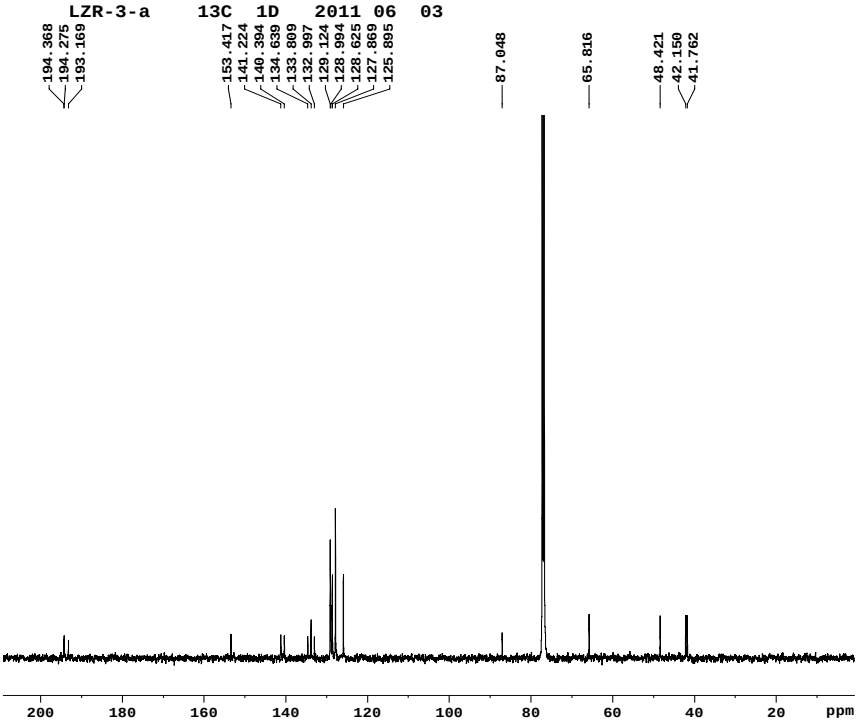
```
NAME                 LZR-2-Z
EXPNO                      2
PROCNO                     1
Date_               20110913
Time                   11.16
INSTRUM                av500
PROBHD    5 mm PABBO BB-
PULPROG              zgpg30
TD                     65536
SOLVENT                CDCl3
NS                      3861
DS                         2
SWH              32679.738 Hz
FIDRES               0.498653 Hz
AQ                1.0027661 sec
RG                      1620
DW                    15.300 usec
DE                     6.00 usec
TE                    300.5 K
D1                2.00000000 sec
d11               0.03000000 sec
DELTA             1.89999998 sec
TD0                        1

======== CHANNEL f1 ========
NUC1                     13C
P1                      9.60 usec
PL1                     2.00 dB
SFO1            125.7464750 MHz

======== CHANNEL f2 ========
CPDPRG2              waltz16
NUC2                      1H
PCPD2                  80.00 usec
PL2                     2.60 dB
PL12                   17.66 dB
PL13                   17.66 dB
SFO2            500.0355000 MHz
SI                       32768
SF              125.7326387 MHz
WDW                       EM
SSB                        0
LB                      6.00 Hz
GB                         0
PC                      2.00
```

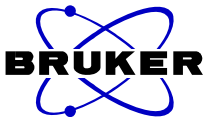


^{1}H and ^{13}C NMR spectra of **4y**

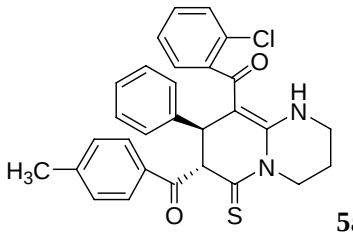



```
NAME              LZR-3-a
EXPNO                   1
PROCNO                  1
Date_            20110530
Time                16.26
INSTRUM             av500
PROBHD   5 mm PABBO BB-
PULPROG              zg30
TD                  32768
SOLVENT             CDCl3
NS                      4
DS                      1
SWH           10000.000 Hz
FIDRES            0.305176 Hz
AQ             1.6385000 sec
RG                    406
DW                 50.000 usec
DE                  6.00 usec
TE                 295.9 K
D1             2.00000000 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                   1H
P1                  13.50 usec
PL1                  2.20 dB
SFO1          500.0335010 MHz
SI                  16384
SF            500.0300082 MHz
WDW                    EM
SSB                     0
LB                   0.30 Hz
GB                      0
PC                   2.00
```

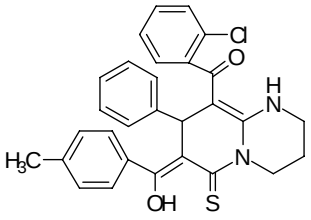





```
NAME              LZR-3-a
EXPNO                   2
PROCNO                  1
Date_            20110603
Time                16.40
INSTRUM             av500
PROBHD   5 mm PABBO BB-
PULPROG           zgpg30
TD                  65536
SOLVENT             CDCl3
NS                    628
DS                      2
SWH           32679.738 Hz
FIDRES            0.498653 Hz
AQ             1.0027661 sec
RG                  18400
DW                 15.300 usec
DE                  6.00 usec
TE                 297.5 K
D1             2.00000000 sec
d11            0.03000000 sec
DELTA          1.89999998 sec
TD0                     1

======== CHANNEL f1 ========
NUC1                  13C
P1                   9.60 usec
PL1                  2.00 dB
SFO1         125.7464750 MHz

======== CHANNEL f2 ========
CPDPRG2           waltz16
NUC2                   1H
PCPD2               80.00 usec
PL2                  2.60 dB
PL12                17.66 dB
PL13                17.66 dB
SFO2          500.0355000 MHz
SI                  32768
SF            125.7326470 MHz
WDW                    EM
SSB                     0
LB                   6.00 Hz
GB                      0
PC                   2.00
```

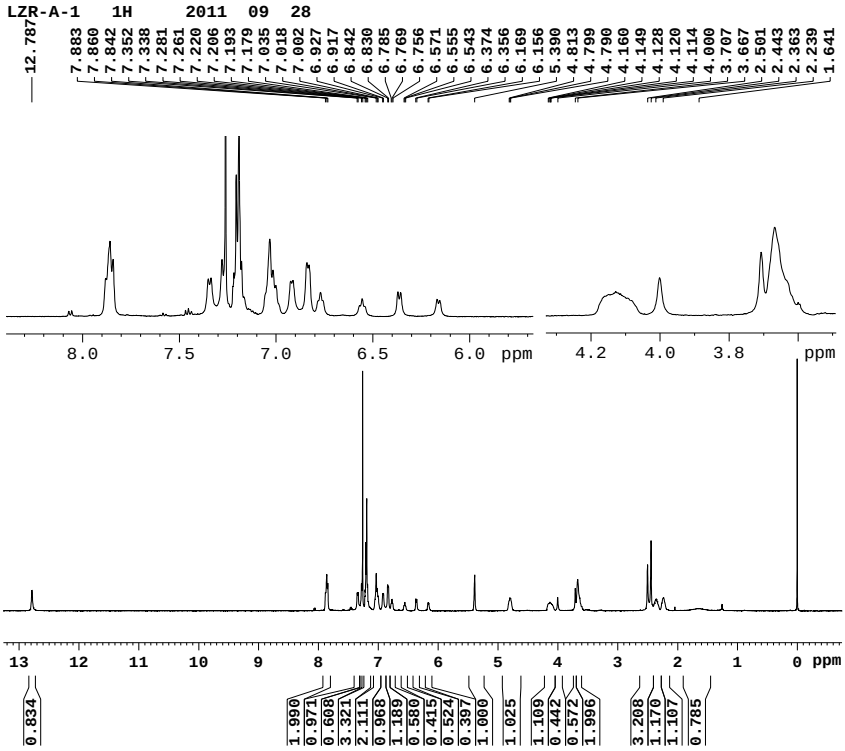


5a

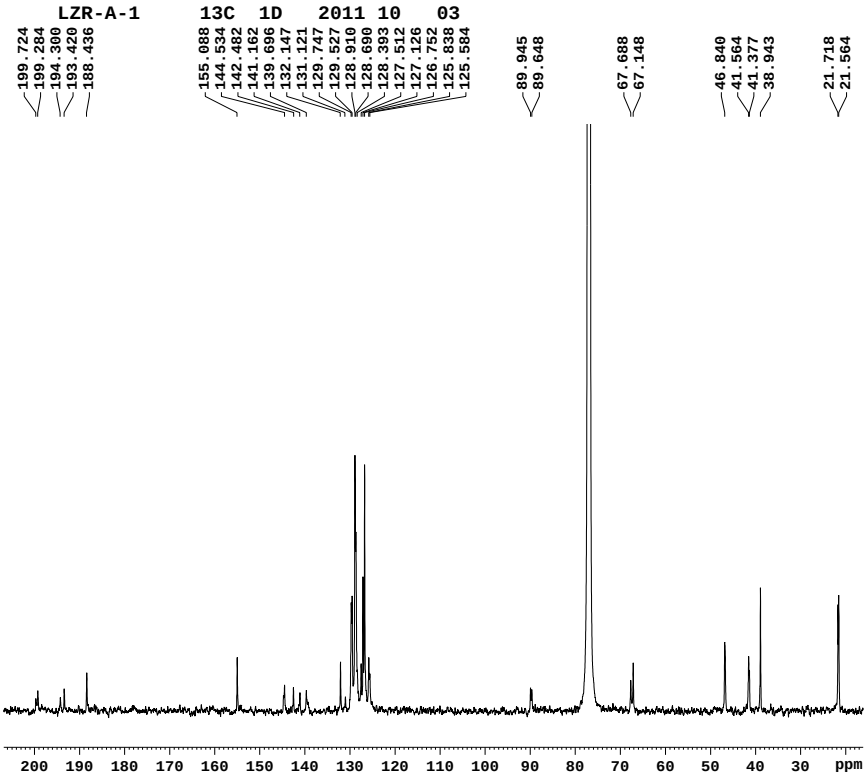


5a'

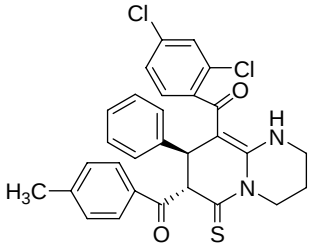
^{1}H and ^{13}C NMR spectra of **5a/5a'**



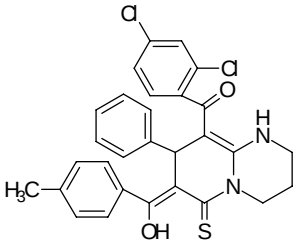

```
NAME                 LZR-A-1
EXPNO                      1
PROCNO                     1
Date_               20110928
Time                   15.37
INSTRUM                av500
PROBHD   5 mm PABBO BB-
PULPROG                 zg30
TD                     32768
SOLVENT                CDCl3
NS                         8
DS                         1
SWH              10000.000 Hz
FIDRES               0.305176 Hz
AQ                1.6385000 sec
RG                       406
DW                    50.000 usec
DE                     6.00 usec
TE                    296.9 K
D1               2.00000000 sec
TD0                        1

======== CHANNEL f1 ========
NUC1                      1H
P1                     13.50 usec
PL1                     2.20 dB
SFO1             500.0335010 MHz
SI                       16384
SF               500.0300101 MHz
WDW                       EM
SSB                        0
LB                      0.30 Hz
GB                         0
PC                      2.00
```




```
NAME                 LZR-A-1
EXPNO                      2
PROCNO                     1
Date_               20111003
Time                   11.09
INSTRUM                av500
PROBHD   5 mm PABBO BB-
PULPROG               zgpg30
TD                     65536
SOLVENT                CDCl3
NS                     11478
DS                         2
SWH             32679.738 Hz
FIDRES               0.498653 Hz
AQ                1.0027661 sec
RG                     18400
DW                    15.300 usec
DE                     6.00 usec
TE                    296.6 K
D1               2.00000000 sec
d11              0.03000000 sec
DELTA            1.89999998 sec
TD0                        1

======== CHANNEL f1 ========
NUC1                      13C
P1                      9.60 usec
PL1                     2.00 dB
SFO1            125.7464750 MHz

======== CHANNEL f2 ========
CPDPRG2               waltz16
NUC2                      1H
PCPD2                  80.00 usec
PL2                     2.60 dB
PL12                   17.66 dB
PL13                   17.66 dB
SFO2             500.0355000 MHz
SI                       32768
SF               125.7326437 MHz
WDW                       EM
SSB                        0
LB                     10.00 Hz
GB                         0
PC                      1.00
```

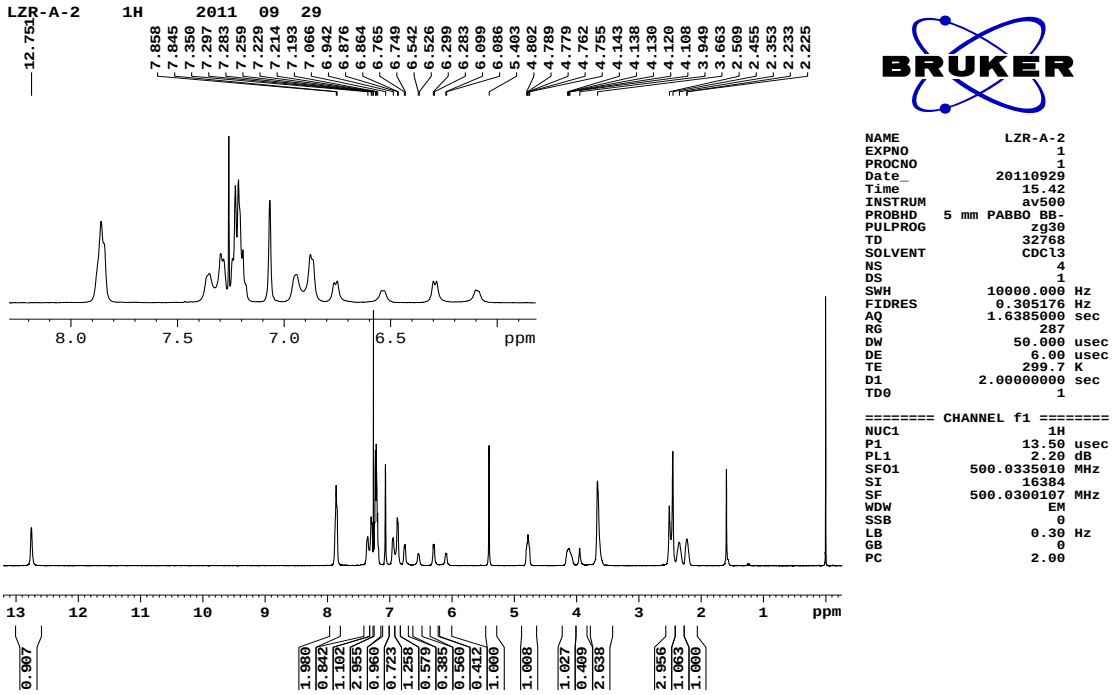


5b

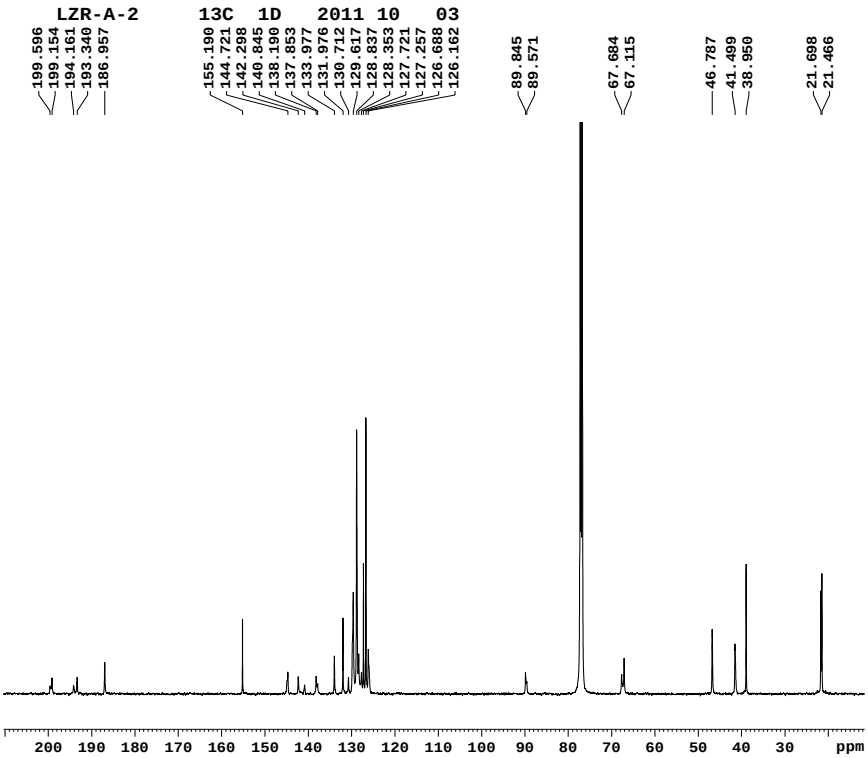


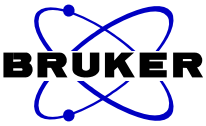
5b'

^{1}H and ^{13}C NMR spectra of **5b/5b'**



NAME	LZR-A-2
EXPNO	1
PROCNO	1
Date_	20110929
Time	15.42
INSTRUM	av500
PROBHD	5 mm PABBO BB-
PULPROG	zg30
TD	32768
SOLVENT	CDCl3
NS	4
DS	1
SWH	10000.000 Hz
FIDRES	0.305176 Hz
AQ	1.6385000 sec
RG	287
DW	50.000 usec
DE	6.00 usec
TE	299.7 K
D1	2.00000000 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	1H
P1	13.50 usec
PL1	2.20 dB
SFO1	500.0335010 MHz
SI	16384
SF	500.0300107 MHz
WDW	EM
SSB	0
LB	0.30 Hz
GB	0
PC	2.00





NAME	LZR-A-2
EXPNO	2
PROCNO	1
Date_	20111004
Time	9.51
INSTRUM	av500
PROBHD	5 mm PABBO BB-
PULPROG	zgpg30
TD	65536
SOLVENT	CDCl3
NS	14827
DS	2
SWH	32679.738 Hz
FIDRES	0.498653 Hz
AQ	1.0027661 sec
RG	18400
DW	15.300 usec
DE	6.00 usec
TE	299.7 K
D1	2.00000000 sec
d11	0.03000000 sec
DELTA	1.89999998 sec
TD0	1
======== CHANNEL f1 ========	
NUC1	13C
P1	9.60 usec
PL1	2.00 dB
SFO1	125.7464750 MHz
======== CHANNEL f2 ========	
CPDPRG2	waltz16
NUC2	1H
PCPD2	80.00 usec
PL2	2.60 dB
PL12	17.66 dB
PL13	17.66 dB
SFO2	500.0355000 MHz
SI	32768
SF	125.7326464 MHz
WDW	EM
SSB	0
LB	6.00 Hz
GB	0
PC	2.00