

Direct β -Acylation of 2-Arylidene-1,3-indandiones with Acyl Chlorides Catalyzed by Organophosphanes

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Supplementary Data

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I. Representative Experimental procedures:

TP1: Typical procedure for the preparation of compounds **1**

Preparation of **1aa** (entry 1, Table 2): A dry and nitrogen-flushed 10-mL Schlenk flask, equipped with a magnetic stirring bar and a septum, was sequentially charged with a solution of **6a** (93.9 mg, 0.3 mmol), MePPh₂ (5.7 µL, 10 mol%), **11a** (42.6 µL, 1.2 equiv) and Et₃N (54.4 µL, 1.3 equiv) in THF (1.5 mL). The reaction mixture was stirred for 2 hours at room temperature (27-30 °C). Thereafter, the solvent was removed by evaporation *in vacuo*. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1aa** as yellow solid (123.0 mg, 98%).

TP2: Typical procedure for the preparation of compounds **1xm**

Preparation of **1am** (Scheme 2): A dry and nitrogen-flushed 10-mL Schlenk flask, equipped with a magnetic stirring bar and a septum, was sequentially charged with a solution of **6a** (93.9 mg, 0.3 mmol), EtPPh₂ (9.4 µL, 15 mol%), **11m** (54.2 µL, 1.3equiv) and Et₃N (58.9 µL, 1.4 equiv) in THF (1.5 mL). The reaction mixture was stirred for 10 minutes at room temperature (27-30 °C). Thereafter, the solvent was removed by evaporation *in vacuo*. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1am** as yellow solid (120.3 mg, 98%).

TP3: Typical procedure for the preparation of compounds **2-5**

Preparation of **2a** (Scheme 6): A dry and nitrogen-flushed 10-mL Schlenk flask, equipped with a magnetic stirring bar and a septum, was sequentially charged with a solution of **7a** (111.1 mg, 0.3 mmol), Bu₃P (8.7 µL, 10 mol%), **11a** (38.3 µL, 1.1equiv) and Et₃N (50.2 µL, 1.2 equiv) in THF (1.5 mL). The reaction mixture was stirred for 50 minutes at room temperature (27-30 °C). Thereafter, the solvent was removed by evaporation *in vacuo*. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **2a** as yellow solid (93.4 mg, 66%).

TP4: Typical procedure for the preparation of compounds **16, 17, 18**

Preparation of **16a** (Scheme 7): A 10-mL Schlenk flask, equipped with a magnetic stirring bar and a septum, was sequentially charged with a solution of **1am** (122.7 mg, 0.3 mmol), hydrazine hydrate (22.0 µL, 1.5 equiv) in methanol (1.5 mL). The reaction mixture was stirred for 10 minutes at room temperature (27-30 °C). Thereafter, the solvent was removed by evaporation *in vacuo*. Purification by flash chromatography (hexanes/dichloromethane: 1/3) furnished **16a** as yellow solid (88.7 mg, 98%).

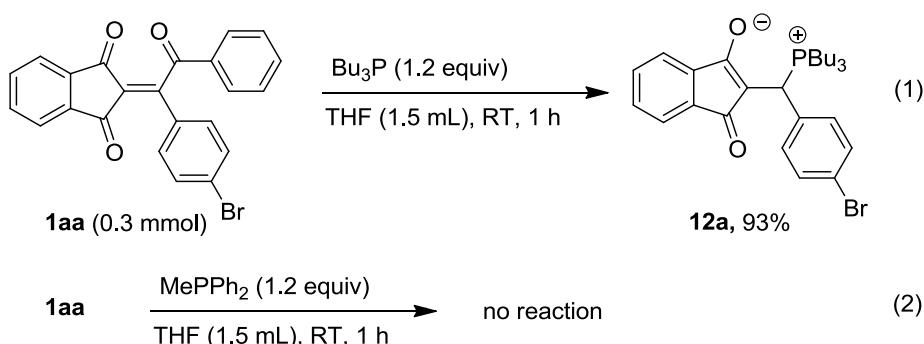
TP5: Typical procedure for the preparation of starting material **6**

Preparation of **6a**: A 50-mL Schlenk flask, equipped with a magnetic stirring bar, was sequentially charged with a solution of 1,3-indanedione (1506.5 mg, 10 mmol) *L*-proline

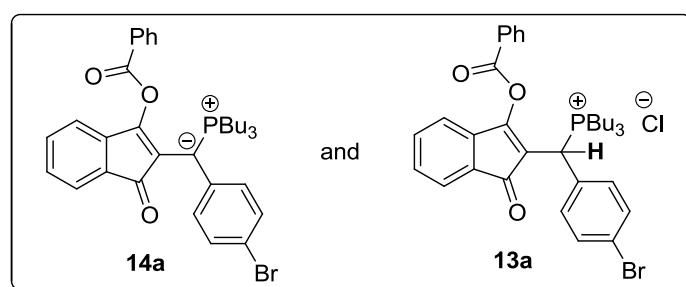
(349.0 mg, 0.3equiv) and benzaldehyde (2035.2 mg, 1.1 equiv) in methanol (20 mL). The reaction mixture was stirred for 12 hours at room temperature (27-30 °C). Thereafter, the resulting mixture was purified using vacuum-filtration and washed with methanol for several times. The resulted product yielded **6a** as solid (2974.0 mg, 95%).

II. Controlled experiment: formation of phosphorus zwitterion

In order to understand the mechanism of our developed β -acylation reactions, four interesting results were selected for discussion (Table 1, entries 4, 9, 11, and 12). We found that 1.2 equiv of Bu_3P did not catalyze the expected reaction of **6a** and **11a** towards **1aa**, which indicated that organophosphane may not only play an important role to direct our reaction pathway towards the final product (entry 4 vs 11). In addition, the same phenomena were not observed when $MePPh_2$ was used instead of Bu_3P (entries 11 and 12). Based on our hypothesis, we assumed that the formation of the intermediate **13a** as the final product was possible when a strong nucleophilic organophosphane, such as Bu_3P , was present (Schemes S1 and 5). Because of the labile ester functionality of **13a**, only the phosphorus zwitterion **12a** was possibly observed in $CDCl_3$ and purified (entry 11). One controlled experiment was carried out to support our assumption of possible reactive intermediates **13a** and **14a** coming from the reaction of **1aa** and Bu_3P (Scheme 5, equation 1). The phosphorus zwitterionic intermediate **12a** was given in 93% yield after the treatment of **1aa** with 1.2 equiv of Bu_3P in THF. However, the adduct **1aa** was quite stable in the presence of $MePPh_2$ (equation 2). And we found that $MePPh_2$, $EtPPh_2$ and PPh_3 do not have enough nucleophilicity to carry out this reaction (Schemes S2). On the other hand, PBu_3 is the better choice for these substrates.

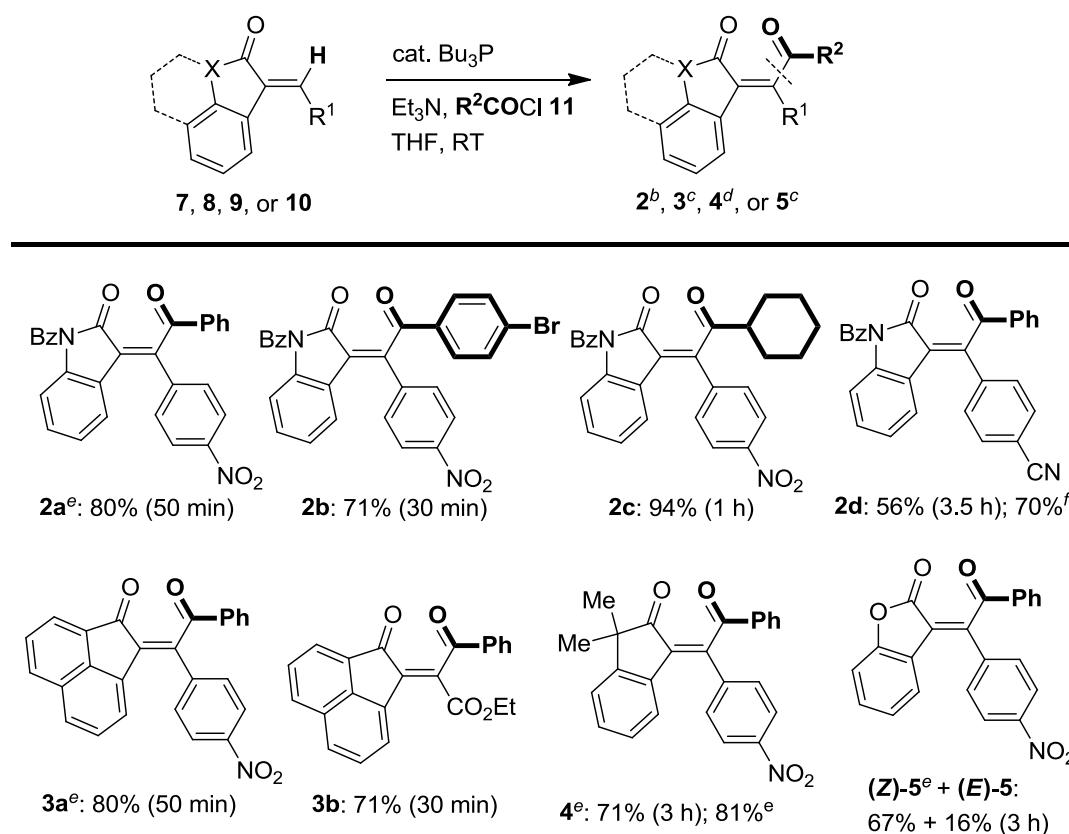


Presumable reactive intermediates:



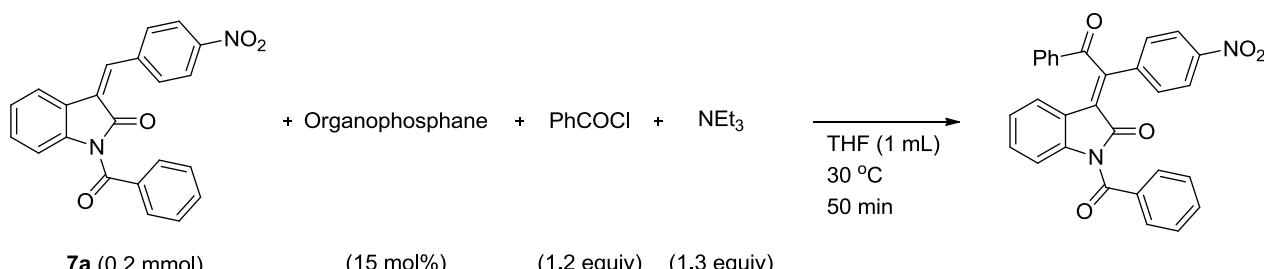
Scheme S1 Controlled experiment: formation of phosphorus zwitterion **12a**.

III. The reaction conditions for Scheme 6 (manuscript): organocatalytic synthesis of 2-5 via direct β -acylation.



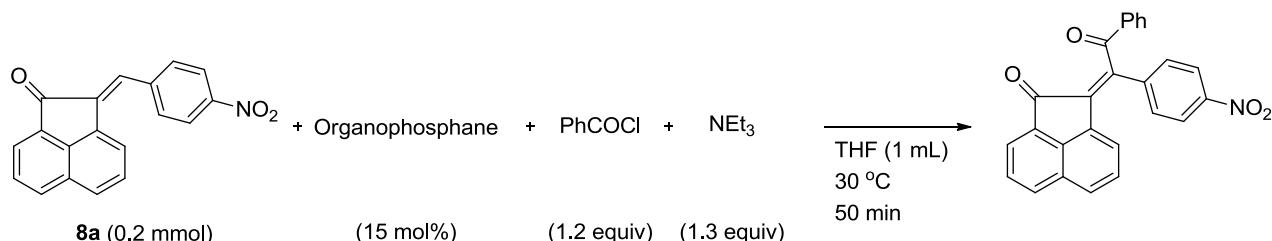
Scheme S2 Organocatalytic synthesis of **2-5^a** via direct β -acylation. ^aFor all reactions 0.3 mmol of **7-10** were used in anhydrous THF (1.5 mL) under N_2 . ^b1.1 Equiv of **11** and 1.2 equiv of Et_3N in the presence of Bu_3P (10 mol%) were used. ^c1.3 Equiv of **11a** and 1.5 equiv of Et_3N in the presence of Bu_3P (20 mol%) were used. ^d1.2 Equiv of **11a** and 1.3 equiv of Et_3N in the presence of Bu_3P (20 mol%) were used. ^eThe structures of **2a**, **3a**, **4**, and **5** were determined by X-ray analysis.¹¹ ^fRecovered yield.

Table S1 The optimization of various organophosphanes with compound **7a**.



entry	Organophosphane	yield
1	PBu ₃	80
2	PPh ₂ Et	2
3	PPh ₂ Me	5
4	PPh ₃	0

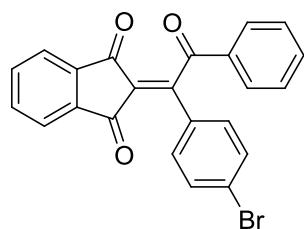
Table S2 The optimization of various organophosphanes with compound **8a**.



entry	Organophosphane	yield
1	PBu ₃	80
2	PPh ₂ Et	1
3	PPh ₂ Me	2
4	PPh ₃	0

IV. Spectra data of the substrates

Synthesis of 2-(1-(4-bromophenyl)-2-oxo-2-phenylethylidene)-1*H*-indene-1,3(2*H*)-dione (**1aa**):



Prepared according to **TP 1** from 2-(4-bromobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6a** (93.9 mg, 0.3 mmol), MePPh₂ (5.7 µL, 10 mol%), benzoyl chloride **11a** (42.6 µL, 1.2 equiv) and Et₃N (54.4 µL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 2 hours]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1aa** as yellow solid (123.0 mg, 98%). mp: 193.2–194.0 °C; R_f 0.33 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.00 (dd, 1H, J = 6.6, 1.4 Hz), 7.95 (d, 2H, J = 7.8 Hz), 7.91–7.79 (m, 3H), 7.63–7.55 (m, 5H), 7.47 (t, 2H, J = 7.7 Hz).

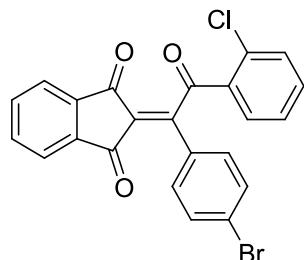
¹³C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 195.0, 188.7, 187.3, 157.3, 142.3, 140.1, 135.9, 135.8, 135.0, 134.0, 131.7, 131.4, 129.6, 128.9, 128.0, 126.6, 123.5, 123.4.

MS (70 eV, EI) m/z (%): 418 [M+2]⁺ (42), 416 [M]⁺ (50), 176 (49), 105 (100), 77 (46).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3076 (w), 2924 (w), 2855 (w), 1730 (m), 1691 (s), 1669 (s), 1585 (m), 1486 (m), 594 (m).

HRMS (FAB) for C₂₃H₁₄BrO₃ [M+H]⁺ (417.0126) found 417.0119.

Synthesis of 2-(1-(4-bromophenyl)-2-(2-chlorophenyl)-2-oxoethylidene)-1*H*-indene-1,3(2*H*)-dione (**1ab**):



Prepared according to **TP 1** from 2-(4-bromobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6a** (93.9 mg, 0.3 mmol), MePPh₂ (5.7 µL, 10 mol%), 2-chlorobenzoyl chloride **11b** (46.5 µL, 1.2 equiv) and Et₃N (54.4 µL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 1 hour]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1ab** as yellow solid (131.0 mg, 97%).

mp: 217.4–217.9 °C; R_f 0.41 (hexanes/dichloromethane: 1/1)

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 7.92–7.85 (m, 2H), 7.85–7.79 (m, 1H), 7.78–7.69 (m, 2H), 7.49 (s, 4H), 7.40–7.32 (m, 2H), 7.29–7.23 (m, 2H).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 193.0, 189.2, 187.4, 158.0, 142.4, 140.0, 135.9, 135.7, 134.3, 134.1, 133.1, 132.3, 132.1, 131.8, 131.5, 128.7, 128.0, 127.0, 126.7, 123.5, 123.4.

MS (70 eV, EI) m/z (%): 450 [M]⁺ (72), 414 (37), 176 (55), 139 (100), 111 (41), 75 (25).

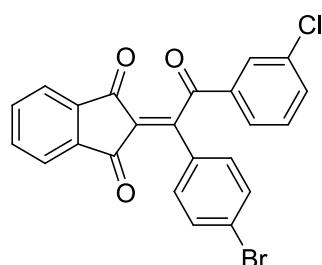
IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3092 (w), 3063 (w), 2923 (w), 1727 (m), 1694 (s), 1608 (m), 1484 (m), 1241 (s), 618 (m).

HRMS (EI) for C₂₃H₁₂BrClO₃ [M]⁺ (449.9658) found 449.9656.

Synthesis

of

2-(1-(4-bromophenyl)-2-(3-chlorophenyl)-2-oxoethylidene)-1*H*-indene-1,3(2*H*)-dione (**1ac**):



Prepared according to **TP 1** from 2-(4-bromobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6a** (93.9 mg, 0.3 mmol), MePPh₂ (5.7 μ L, 10 mol%), 3-chlorobenzoyl chloride **11c** (47.5 μ L, 1.2 equiv) and Et₃N (54.4 μ L, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27-30 °C for 0.5 hour]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1ac** as yellow solid (131.0 mg, 98%).

mp: 139.0-139.9 °C; R_f 0.46 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ /ppm: 7.99 (dd, 1H, J = 6.4, 1.3 Hz), 7.94 (t, 1H, J = 1.7 Hz), 7.90-7.78 (m, 4H, 7.60 (s, 4H), 7.54 (ddd, 1H, J = 8.0, 2.0, 0.9 Hz), 7.40 (t, 1H, J = 7.9 Hz).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ /ppm: 193.6, 188.6, 186.9, 155.9, 142.2, 139.9, 136.5, 135.9, 135.7, 135.1, 133.7, 131.7, 131.3, 130.2, 129.0, 128.4, 128.2, 127.0, 126.7, 123.5, 123.3.

MS (70 eV, EI) m/z (%): 452 [M+2]⁺ (19), 450 [M]⁺ (18), 176 (42), 139 (100), 111 (59), 75 (37).

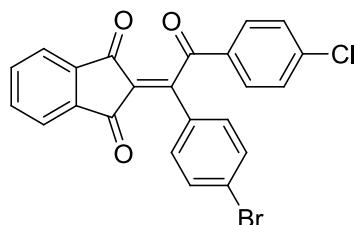
IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3063 (w), 2952 (w), 2923 (w), 1731 (m), 1694 (s), 1606 (m), 1580 (m), 1484 (m), 1241 (s), 594 (m).

HRMS (EI) for C₂₃H₁₂BrClO₃ [M]⁺ (449.9658) found 449.9659.

Synthesis

of

2-(1-(4-bromophenyl)-2-(4-chlorophenyl)-2-oxoethylidene)-1*H*-indene-1,3(2*H*)-dione (**1ad**):



Prepared according to **TP 1** from 2-(4-bromobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6a** (93.9 mg, 0.3 mmol), MePPh₂ (5.7 μ L, 10 mol%), 4-chlorobenzoyl chloride **11d** (45.8 μ L, 1.2 equiv) and Et₃N (54.4 μ L, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27-30 °C for 0.5 hour]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1ad** as yellow solid (126.6 mg, 93%).

mp: 207.6-208.4 °C; R_f 0.46 (hexanes/dichloromethane: 1/1).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 °C) δ/ppm: 7.96 (d, 1H, $J = 6.8$ Hz), 7.91-7.74 (m, 5H), 7.57 (s, 4H), 7.41 (d, 2H, $J = 8.5$ Hz).

$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 °C) δ/ppm: 193.8, 188.7, 187.1, 156.5, 142.3, 140.4, 140.1, 136.0, 135.8, 133.4, 131.8, 131.4, 130.2, 129.3, 128.1, 126.8, 123.6, 123.4.

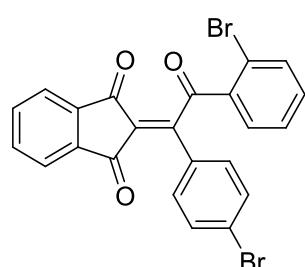
MS (70 eV, EI) m/z (%): 450 [M]⁺, 176 (33), 139 (100), 111 (19), 75 (3).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3092 (w), 2924 (w), 1726 (m), 1695 (s), 1669 (s), 1585 (s), 1486 (m), 1250 (s), 594 (m).

HRMS (EI) for $\text{C}_{23}\text{H}_{12}\text{BrClO}_3$ [M]⁺ (449.9658) found 449.9659.

Synthesis of

2-(2-(2-bromophenyl)-1-(4-bromophenyl)-2-oxoethylidene)-1*H*-indene-1,3(2*H*)-dione (**1ae**):



Prepared according to **TP 1** from 2-(4-bromobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6a** (93.9 mg, 0.3 mmol), MePPh₂ (5.7 μL, 10 mol%), 2-bromobenzoyl chloride **11e** (48.0 μL, 1.2 equiv) and Et₃N (54.4 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27-30 °C for 0.5 hour]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1ae** as yellow solid (144.0 mg, 97%).

mp: 198.7-199.7 °C; R_f 0.40 (hexanes/dichloromethane: 1/1).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 °C) δ/ppm: 7.95 (d, 1H, $J = 6.8$ Hz), 7.90 (d, 1H, $J = 6.6$ Hz), 7.85-7.76 (m, 3H), 7.73-7.65 (m, 1H), 7.62-7.53 (m, 4H), 7.37-7.29 (m, 2H).

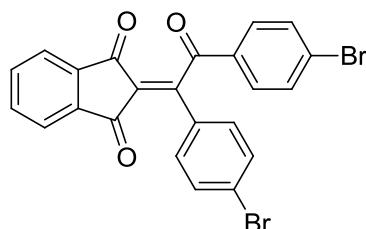
$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 °C) δ/ppm: 193.4, 189.0, 187.3, 157.2, 142.4, 140.0, 135.9, 135.7, 135.6, 133.8, 132.8, 131.9, 131.6, 129.1, 128.2, 127.3, 126.7, 123.5, 122.4.

MS (70 eV, EI) m/z (%): 496 [M+2]⁺ (46), 415 (24), 311 (14), 183 (100), 176 (55), 155 (43), 76 (51).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3053 (w), 2924 (w), 1726 (m), 1691 (s), 1585 (s), 1482 (m), 594 (m).

HRMS (FAB) for $\text{C}_{23}\text{H}_{12}\text{Br}_2\text{O}_3$ [M+H]⁺ (494.9231) found 494.9236.

Synthesis of 2-(1,2-bis(4-bromophenyl)-2-oxoethylidene)-1*H*-indene-1,3(2*H*)-dione (1af**):**



Prepared according to **TP 1** from 2-(4-bromobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6a** (93.9 mg, 0.3 mmol), MePPh₂ (5.7 μL, 10 mol%), 4-bromobenzoyl chloride **11f** (80.6 mg, 1.2 equiv) and Et₃N (54.4 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 0.5 hour]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1af** as yellow solid (148.9 mg, 97%).

mp: 225.4–226.2 °C; R_f 0.46 (hexanes/dichloromethane: 1/1).

¹H-NMR (500 MHz, CDCl₃, 25 °C) δ/ppm: 7.98 (d, 1H, J = 7.4 Hz), 7.89 (d, 1H, J = 7.4 Hz), 7.86–7.77 (m, 4H), 7.61–7.56 (m, 6H).

¹³C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 194.1, 188.8, 187.1, 156.5, 142.4, 140.1, 136.0, 135.9, 133.8, 132.3, 131.8, 131.4, 130.3, 129.3, 129.2, 127.1, 126.8, 123.6, 123.5.

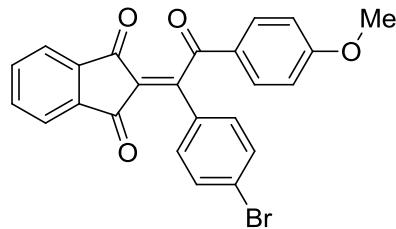
MS (70 eV, EI) m/z (%): 496 [M+2]⁺ (30), 494 [M]⁺ (27), 185 (100), 176 (41), 155 (15), 104 (9), 76 (6).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3085 (w), 2923 (w), 1727 (m), 1694 (s), 1583 (m), 1488 (m), 592 (m).

HRMS (EI) for C₂₃H₁₂Br₂O₃ [M]⁺ (493.9153) found 493.9149.

Synthesis of

2-(1-(4-bromophenyl)-2-(4-methoxyphenyl)-2-oxoethylidene)-1*H*-indene-1,3(2*H*)-dione (1ag**):**



Prepared according to **TP 1** from 2-(4-bromobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6a** (93.9 mg, 0.3 mmol), MePPh₂ (8.7 μL, 15 mol%), 4-methoxybenzoyl chloride **11g** (49.7 μL, 1.2 equiv) and Et₃N (54.4 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 7.5 hours]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1ag** as yellow solid (130.5 mg, 97%).

mp: 190.6–191.5 °C; R_f 0.2 (hexanes/dichloromethane: 1/1).

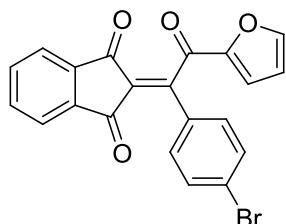
¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 7.95 (d, 1H, J = 7.6 Hz), 7.92–7.74 (m, 5H), 7.60–7.51 (m, 4H), 6.90 (d, 2H, J = 8.8 Hz), 3.82 (s, 3H).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 193.4, 188.7, 187.5, 164.2, 157.6, 142.2, 140.2, 135.8, 135.7, 131.6, 131.3, 131.3, 130.0, 128.2, 127.6, 126.4, 123.4, 123.4, 114.2, 55.5.

MS (70 eV, EI) m/z (%): 448 [M+2]⁺ (62), 446 [M]⁺ (70), 176 (18), 135 (100), 92 (5).

IR (KBr) $\tilde{\nu}$ (cm^{-1}): 2938 (w), 1726 (m), 1691 (s), 1661 (s), 1592 (s), 1486 (m), 1170 (s), 590 (m).
HRMS (EI) for $\text{C}_{24}\text{H}_{15}\text{BrO}_4 [\text{M}]^+$ (446.0154) found 446.0159.

Synthesis of 2-(1-(4-bromophenyl)-2-(furan-2-yl)-2-oxoethylidene)-1*H*-indene-1,3(2*H*)-dione (1ah):



Prepared according to **TP 1** from 2-(4-bromobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6a** (93.9 mg, 0.3 mmol), MePPh₂ (8.7 μL , 15 mol%), 2-furoyl chloride **11h** (37.0 μL , 1.2 equiv) and Et₃N (54.4 μL , 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 3 hours]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1ah** as yellow solid (86.7 mg, 71%). mp: 226.7–227.5 °C; R_f 0.18 (hexanes/dichloromethane: 1/1).

¹H-NMR (500 MHz, CDCl₃, 25 °C) δ /ppm: 7.97 (dd, 1H, *J* = 6.3, 2.2 Hz), 7.91 (dd, 1H, *J* = 5.9, 1.9 Hz), 7.86–7.80 (m, 2H), 7.60–7.53 (m, 5H), 7.23 (d, 1H, *J* = 3.6 Hz), 6.55 (dd, 1H, *J* = 3.6, 1.6 Hz).

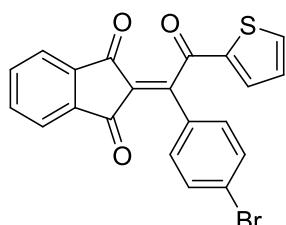
¹³C-NMR (125 MHz, CDCl₃, 25 °C) δ /ppm: 188.6, 187.4, 182.6, 154.9, 151.5, 147.5, 142.3, 140.3, 135.9, 135.8, 131.6, 131.3, 129.6, 128.4, 126.4, 123.5, 123.5, 123.4, 119.1, 112.8.

MS (70 eV, EI) m/z (%): 408 [M+2]⁺ (14), 406 [M]⁺ (13), 379 (50), 311 (9), 207 (5), 127 (5), 76 (35), 95 (100), 76 (14).

IR (KBr) $\tilde{\nu}$ (cm^{-1}): 3125 (w), 3093 (w), 1731 (m), 1692 (s), 1651 (s), 1586 (m), 1462 (m), 1219 (m), 575 (w).

HRMS (ESI) for $\text{C}_{21}\text{H}_{12}\text{BrO}_4 [\text{M}+\text{H}]^+$ (406.9919) found 406.9931.

Synthesis of 2-(1-(4-bromophenyl)-2-oxo-2-(thiophen-2-yl)ethylidene)-1*H*-indene-1,3(2*H*)-dione (1ai):



Prepared according to **TP 1** from 2-(4-bromobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6a** (93.9 mg, 0.3 mmol), MePPh₂ (8.7 μL , 15 mol%), 2-thiophene carbonyl chloride **11i** (39.3 μL , 1.2 equiv) and Et₃N (54.4 μL , 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 5 hours]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1ai** as yellow solid (110.4 mg, 87%).

mp: 223.1–224.0 °C; R_f 0.30 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ /ppm: 7.98 (dd, 1H, *J* = 6.0, 2.1 Hz), 7.91 (dd, 1H, *J* = 5.6, 1.9

Hz), 7.87-7.79 (m, 2H), 7.72 (d, 1H, $J = 4.8$ Hz), 7.63-7.56 (m, 4H), 7.54 (d, 1H, $J = 3.8$ Hz), 7.08 (t, 1H, $J = 4.4$ Hz).

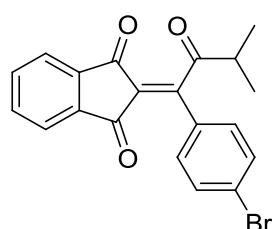
$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 °C) δ/ppm: 188.3, 187.4, 186.9, 155.8, 142.3, 142.2, 140.3, 135.9, 135.9, 135.2, 134.1, 131.7, 131.4, 129.8, 128.4, 127.9, 126.6, 123.5.

MS (70 eV, EI) m/z (%): 422 [M^+] (35), 343 (2), 311 (3), 176 (29), 111 (100), 83 (14).

IR (KBr) $\tilde{\nu}$ (cm^{-1}): 3111 (w), 3088 (w), 2923 (w), 1728 (m), 1690 (s), 1643 (s), 1585 (m), 1487 (m), 580 (m).

HRMS (ESI) for $\text{C}_{21}\text{H}_{12}\text{BrO}_3\text{S} [\text{M}+\text{H}]^+$ (422.9691) found 422.9704.

Synthesis of 2-(1-(4-bromophenyl)-3-methyl-2-oxobutylidene)-1*H*-indene-1,3(2*H*)-dione (1aj):



Prepared according to **TP 1** from 2-(4-bromobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6a** (93.9 mg, 0.3 mmol), MePPh₂ (28.5 μL, 50 mol%), isobutryl chloride **11j** (38.5 μL, 1.2 equiv) and Et₃N (54.4 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27-30 °C for 1 hour]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1af** as yellow solid (91.0 mg, 79%).

mp: 129.3-130.2 °C; R_f 0.68 (hexanes/dichloromethane: 1/3).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 °C) δ/ppm: 7.92-7.78 (m, 2H), 7.78-7.66 (m, 2H), 7.50 (d, 2H, $J = 8.5$ Hz), 7.38 (d, 2H, $J = 8.5$ Hz), 2.78 (sep, 1H, $J = 7.0$ Hz), 1.07 (d, 6H, $J = 7.0$ Hz).

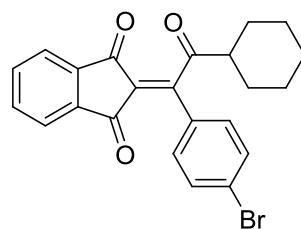
$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 °C) δ/ppm: 209.2, 189.2, 187.2, 159.4, 142.1, 140.0, 135.8, 135.7, 131.5, 130.9, 129.1, 126.6, 126.2, 123.4, 123.3, 41.0, 17.8.

MS (70 eV, EI) m/z (%): 384 [$\text{M}+2]^+$ (16), 382 [M^+] (14), 340 (75), 313 (100), 233 (36), 150 (12), 128 (7), 76 (9), 71 (76).

IR (KBr) $\tilde{\nu}$ (cm^{-1}): 3089 (w), 2969 (m), 1730 (m), 1691 (s), 1589 (m), 1482 (m), 590 (m).

HRMS (EI) for $\text{C}_{20}\text{H}_{15}\text{BrO}_3 [\text{M}]^+$ (382.0205) found 382.0208.

Synthesis of 2-(1-(4-bromophenyl)-2-oxoundecylidene)-1*H*-indene-1,3(2*H*)-dione (1ak):



Prepared according to **TP 1** from 2-(4-bromobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6a** (93.9 mg, 0.3 mmol), MePPh₂ (28.5 μL, 50 mol%), cyclohexanecarbonyl chloride **11k** (50.2 μL, 1.2 equiv) and Et₃N (54.4 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27-30 °C for 1 hour]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1ak** as yellow solid (100.4 mg, 79%).

mp: 151.9-152.8 °C; R_f 0.68 (hexanes/dichloromethane: 1/3).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 7.99-7.89 (m, 2H), 7.86-7.78 (m, 2H), 7.59 (d, 2H, *J* = 8.5 Hz), 7.47 (d, 2H, *J* = 8.5 Hz), 2.76-2.60 (m, 1H), 1.96 (d, 2H, *J* = 11.9 Hz), 1.72 (d, 2H, *J* = 12.0 Hz), 1.62 (d, 2H, *J* = 10.6 Hz), 1.44-1.05 (m, 5H).

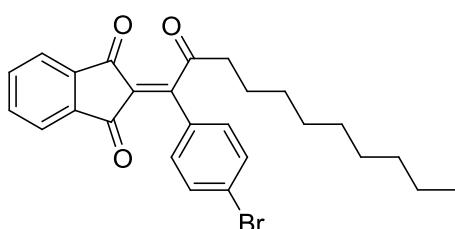
¹³C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 208.3, 189.2, 187.3, 159.5, 142.1, 140.0, 135.8, 135.6, 131.4, 130.9, 129.0, 126.4, 126.1, 123.4, 123.2, 28.2, 25.6, 25.6.

MS (70 eV, EI) m/z (%): 424 [M+2]⁺ (20), 422 [M]⁺ (19), 342 (14), 311 (14), 233 (14), 176 (45), 83 (72).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3090 (w), 3069 (w), 2932 (s), 1728 (s), 1693 (s), 1629 (m), 1587 (m), 1485 (m), 589 (m).

HRMS (EI) for C₂₃H₁₉BrO₃ [M]⁺ (422.0518) found 422.0516.

Synthesis of 2-(1-(4-bromophenyl)-2-oxoundecylidene)-1*H*-indene-1,3(2*H*)-dione (**1al**):



Prepared according to **TP 1** from 2-(4-bromobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6a** (93.9 mg, 0.3 mmol), MePPh₂ (28.5 μL, 50 mol%), decanoyl chloride **11l** (76.2 μL, 1.2 equiv) and Et₃N (54.4 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27-30 °C for 0.5 hour]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1al** as yellow solid (80.5 mg, 57%).

mp: 151.9-152.8 °C; R_f 0.80 (hexanes/dichloromethane: 1/3).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.00-7.90 (m, 2H), 7.86-7.80 (m, 2H), 7.60 (d, 2H, *J* = 8.6 Hz), 7.46 (d, 2H, *J* = 8.6 Hz), 2.67 (t, 2H, *J* = 7.5 Hz), 1.73 (quin, 2H, *J* = 7.3 Hz), 1.36-1.20 (m, 12H), 0.87 (t, 3H, *J* = 6.8 Hz).

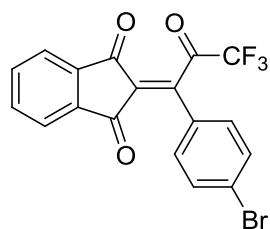
¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 205.3, 189.3, 187.5, 159.2, 142.2, 140.1, 135.9, 135.7, 131.6, 131.0, 128.7, 126.4, 125.9, 123.5, 123.4, 42.1, 31.8, 29.3, 29.3, 29.2, 28.9, 22.9, 22.6, 14.0.

MS (70 eV, EI) m/z (%): 468 [M+2]⁺ (17), 466[M]⁺ (19), 311 (31), 233 (48), 176 (100), 155 (27), 104 (43), 76 (53), 57 (95).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3063 (w), 2925 (s), 2851 (m), 1732 (m), 1695 (s), 1602 (m), 1580 (m), 144 (w), 743 (s).

HRMS (EI) for C₂₆H₂₇BrO₃ [M]⁺ (466.1144) found 466.1132.

Synthesis of 2-(1-(4-bromophenyl)-3,3,3-trifluoro-2-oxopropylidene)-1*H*-indene-1,3(2*H*)-dione (1am**):**



For table 2, prepared according to **TP 1** from 2-(4-bromobenzylidene)-*1H*-indene-1,3(2*H*)-dione **6a** (93.9 mg, 0.3 mmol), MePPh₂ (5.7 μL, 10 mol%), trifluoroacetic anhydride **11m** (50.0 μL, 1.2 equiv) and Et₃N (54.4 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 10 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1am** as yellow solid (98.5 mg, 80%).

For scheme 2, **1am** can also be prepared according to **TP 2**.

mp: 171.5–172.4 °C, R_f 0.51 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.07–7.99 (m, 2H), 7.95–7.87 (m, 2H), 7.66 (d, 2H, J = 8.6 Hz), 7.53 (d, 2H, J = 8.6 Hz).

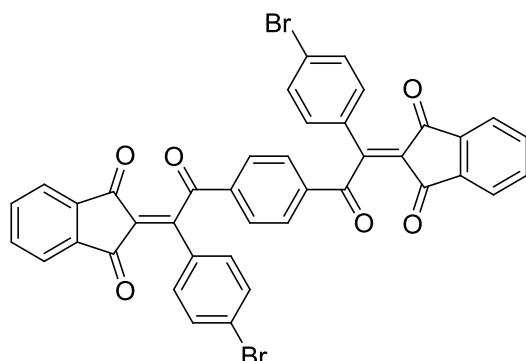
¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 189.2, 187.2 (q, J = 38.8 Hz), 185.6, 149.8, 143.2, 139.3, 136.7, 136.3, 132.1, 131.7, 130.8, 128.0, 126.2, 124.0, 115.1 (q, J = 291.5 Hz)

MS (70 eV, EI) m/z (%): 408 [M]⁺ (32), 341 (69), 313 (100), 284 (26), 176 (77), 127 (19), 75 (54), 69 (77).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3085 (w), 2923 (w), 1746 (m), 1727 (m), 1694 (s), 1617 (m), 1484 (w), 1218 (s), 595 (m).

HRMS (EI) for C₁₈H₈BrF₃O₃ [M]⁺ (407.9609) found 407.9607.

Synthesis of 2,2'-(1,4-phenylenebis(1-(4-bromophenyl)-2-oxoethan-2-yl-1-ylidene))bis(1*H*-indene-1,3(2*H*)-dione) (1an**):**



A dry and nitrogen-flushed 10-mL Schlenk tube, equipped with a magnetic stirring bar and a septum, was sequentially charged with a solution of terephthaloyl chloride **11n** (61.5 mg, 0.3 mmol), 2-(4-bromobenzylidene)-*1H*-indene-1,3(2*H*)-dione **6a** (194.2 mg, 2.1 equiv), MePPh₂ (30 mol%) and Et₃N (92.0 μL, 2.2 equiv) in dry THF (3.0 mL). The reaction mixture was stirred for 7 h at

room temperature (27-30 °C). Thereafter, the solvent was removed by evaporation *in vacuo*. Purification by flash chromatography (hexanes/dichloromethane 1/3; and then dichloromethane) furnished **1an** as yellow solid (167.0 mg, 74%).

mp: 351.4-352.3 °C; R_f 0.3 (hexanes/dichloromethane: 1/3).

1H-NMR (500 MHz, CDCl₃, 25 °C) δ/ppm: 8.02-7.96 (m, 6H), 7.88-7.78 (m, 6H), 7.62-7.53 (m, 8H).

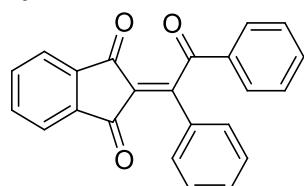
13C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 194.3, 188.9, 187.1, 156.5, 142.5, 140.0, 138.6, 136.1, 135.9, 131.9, 131.5, 129.3, 128.9, 128.3, 127.1, 123.7, 123.6.

MS (70 eV, EI) m/z (%): 758 [M+4]⁺ (75), 756 [M+2]⁺ (100), 443 (15), 311(16), 283 (17), 232 (14), 176 (100), 150 (8), 104 (73), 76 (27).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3098 (w), 3070 (w), 1737 (s), 1610 (m), 1601 (m), 1545 (m), 1474 (w).

HRMS (FAB) for C₄₀H₂₀Br₂O₆ [M+H]⁺ (754.9705) found 754.9713.

Synthesis of 2-(2-oxo-1,2-diphenylethylidene)-1*H*-indene-1,3(2*H*)-dione (**1ba**):



Prepared according to **TP 1** from 2-benzylidene-1*H*-indene-1,3(2*H*)-dione **6b** (70.3 mg, 0.3 mmol), MePPh₂ (5.7 μL, 10 mol%), benzoyl chloride **11a** (42.6 μL, 1.2 equiv) and Et₃N (54.4 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27-30 °C for 1 h]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1ba** as yellow solid (93.0 mg, 92%).

mp: 187.7-188.6 °C; R_f 0.25 (hexanes/dichloromethane: 1/1).

1H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 7.97 (d, 3H, *J* = 7.2 Hz), 7.87 (d, 1H, *J* = 7.4 Hz), 7.85-7.76 (m, 2H), 7.72 (d, 2H, *J* = 7.6 Hz), 7.55 (t, 1H, *J* = 7.3 Hz), 7.52-7.39 (m, 5H).

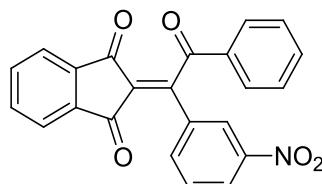
13C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 195.3, 189.0, 187.4, 159.0, 142.4, 140.2, 135.7, 135.6, 135.3, 133.8, 131.6, 130.9, 130.0, 129.0, 128.8, 128.4, 127.7, 123.5, 123.4.

MS (70 eV, EI) m/z (%): 338 [M]⁺ (29), 176 (8), 105 (100), 77 (17).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3055 (w), 1727 (m), 1694 (s), 1668 (m), 1594 (m), 1451 (w).

HRMS (EI) for C₂₃H₁₄O₃ [M]⁺ (338.0943) found 338.0953.

Synthesis of 2-(1-(3-nitrophenyl)-2-oxo-2-phenylethylidene)-1*H*-indene-1,3(2*H*)-dione (**1ca**):



Prepared according to **TP 1** from 2-(3-nitrobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6c** (83.7. mg, 0.3 mmol), MePPh₂ (5.7 μL, 10 mol%), benzoyl chloride **11a** (42.6 μL, 1.2 equiv) and Et₃N (54.4 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27-30 °C for 3 hours]. Purification by flash chromatography (hexanes/dichloromethane: 1/2) furnished **1ca** as yellow solid (86.1 mg, 75%).

mp: 228.8-229.7 °C; R_f 0.18 (hexanes/dichloromethane: 1/1).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 °C) δ/ppm: 8.59 (s, 1H), 8.32 (1H, $J = 8.5$ Hz), 8.03-7.94 (m, 4H), 7.94-7.80 (m, 3H), 7.67-7.56 (m, 2H), 7.48 (t, 2H, $J = 7.6$ Hz).

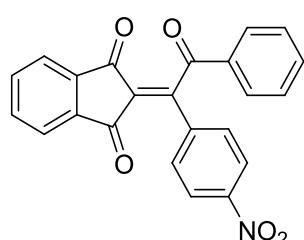
$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 °C) δ/ppm: 194.4, 188.2, 187.0, 154.9, 147.9, 142.3, 140.4, 136.2, 136.1, 135.0, 134.6, 134.3, 132.4, 129.5, 129.4, 129.1, 129.0, 125.5, 124.7, 123.8, 123.7.

MS (70 eV, EI) m/z (%): 383 [M]⁺ (58), 176 (6), 105 (100), 77 (30).

IR (KBr) $\tilde{\nu}$ (cm^{-1}): 3076 (w), 2924 (w), 1730 (m), 1691 (s), 1665 (s), 1596 (m), 1531 (s), 1451 (m), 1349 (s).

HRMS (EI) for $\text{C}_{23}\text{H}_{13}\text{NO}_5$ [M]⁺ (383.0794) found 383.0785.

Synthesis of 2-(1-(4-nitrophenyl)-2-oxo-2-phenylethylidene)-1*H*-indene-1,3(2*H*)-dione (1da):



Prepared according to **TP 1** from 2-(4-nitrobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6d** (83.8 mg, 0.3 mmol), MePPh₂ (5.7 μL, 10 mol%), benzoyl chloride **11a** (42.6 μL, 1.2 equiv) and Et₃N (54.4 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27-30 °C for 1 hour]. Purification by flash chromatography (hexanes/dichloromethane: 1/2) furnished **1da** as yellow solid (122.8 mg, 97%).

mp: 210.6-211.3 °C; R_f 0.15 (hexanes/dichloromethane: 1/1).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 °C) δ/ppm: 8.62-8.57 (m, 1H), 8.32 (d, 1H, $J = 8.0$ Hz), 8.02-7.95 (m, 4H), 7.93-7.81 (m, 3H), 7.66-7.57 (m, 2H), 7.49 (t, 2H, $J = 7.7$ Hz).

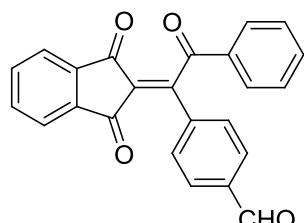
$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 °C) δ/ppm: 194.4, 188.2, 187.0, 154.9, 147.9, 142.3, 140.4, 136.2, 136.1, 135.0, 134.6, 134.3, 132.4, 129.5, 129.3, 129.1, 129.0, 125.5, 124.7, 123.8, 123.7.

MS (70 eV, EI) m/z (%): 383 [M]⁺ (12), 176 (9), 105 (100), 77 (67).

IR (KBr) $\tilde{\nu}$ (cm^{-1}): 3099 (w), 3061 (w), 2855 (w), 1733 (m), 1697 (s), 1664 (m), 1597 (m), 1516 (s), 1450 (w), 1347 (s).

HRMS (EI) for $\text{C}_{23}\text{H}_{13}\text{NO}_5$ [M]⁺ (383.0794) found 383.0792.

Synthesis of 4-(1-(1,3-dioxo-1*H*-inden-2(3*H*)-ylidene)-2-oxo-2-phenylethyl)benzaldehyde (1ea):



Prepared according to **TP 1** from 4-((1,3-dioxo-1*H*-inden-2(3*H*)-ylidene)methyl)benzaldehyde **6e** (79.6 mg, 0.3 mmol), MePPh₂ (5.7 μL, 10 mol%), benzoyl chloride **11a** (42.6 μL, 1.2 equiv) and Et₃N (54.4 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27-30 °C for 0.5 hour]. Purification

by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1ea** as yellow solid (106.0 mg, 97%).

mp: 169.2-170.1 °C; R_f 0.08 (hexanes/dichloromethane: 1/1).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 °C) δ/ppm : 10.05 (s, 1H), 8.12-7.88 (m, 6H), 7.88-7.74 (m, 4H), 7.58 (t, 1H, J = 7.3 Hz), 7.47 (t, 2H, J = 7.5 Hz).

$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 °C) δ/ppm : 194.6, 191.3, 188.4, 187.1, 156.7, 142.4, 140.4, 137.6, 136.7, 136.1, 136.0, 134.9, 134.2, 130.1, 129.4, 129.1, 129.0, 128.9, 123.7, 123.6.

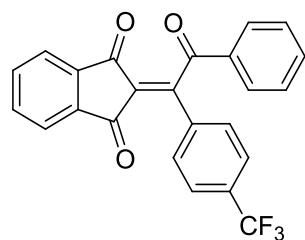
MS (70 eV, EI) m/z (%): 366 [M^+] (23), 261 (3), 105 (100), 76 (75).

IR (KBr) $\tilde{\nu}$ (cm^{-1}): 3055 (w), 2923 (w), 1731 (m), 1694 (s), 1668 (m), 1598 (m), 1447 (w).

HRMS (EI) for $\text{C}_{24}\text{H}_{14}\text{O}_4$ [M^+] (366.0892) found 366.0892.

Synthesis of

2-(2-oxo-2-phenyl-1-(4-(trifluoromethyl)phenyl)ethylidene)-1*H*-indene-1,3(2*H*)-dione (**1fa**):



Prepared according to **TP 1** from 2-(4-(trifluoromethyl)benzylidene)-*1H*-indene-1,3(2*H*)-dione **6f** (121.9 mg, 0.3 mmol), MePPh₂ (5.7 μL , 10 mol%), benzoyl chloride **11a** (42.6 μL , 1.2 equiv) and Et₃N (54.4 μL , 1.3 equiv) in THF (1.5 mL) [reaction condition: 27-30 °C for 1 hour]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1fa** as yellow solid (118.0 mg, 97%).

mp: 190.8-191.7 °C; R_f 0.43 (hexanes/dichloromethane: 1/1).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 °C) δ/ppm : 8.01-7.93 (m, 3H), 7.93-7.77 (m, 5H), 7.69 (d, 2H, J = 8.3 Hz), 7.58 (t, 1H, J = 7.4 Hz), 7.47 (t, 2H, J = 7.7 Hz).

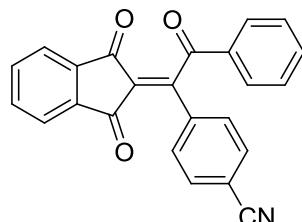
$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 °C) δ/ppm : 194.7, 188.4, 187.1, 156.5, 142.3, 140.3, 136.1, 136.0, 134.8, 134.4, 134.1, 132.6 (q, J = 32.8 Hz), 129.8, 129.0, 125.3 (q, J = 3.7 Hz), 123.6 (q, J = 272.6 Hz), 123.6, 123.6,

MS (70 eV, EI) m/z (%): 406 [M^+] (56), 301 (3), 176 (11), 105 (100), 77 (44)

IR (KBr) $\tilde{\nu}$ (cm^{-1}): 3076 (w), 2924 (w), 1730 (m), 1695 (s), 1669 (s), 1596 (m), 1451 (m), 1337 (s), 1326 (s).

HRMS (EI) for $\text{C}_{24}\text{H}_{13}\text{F}_3\text{O}_3$ [M^+] (406.0817) found 406.0818.

Synthesis of 2-(1-(4-cyanophenyl)-2-oxo-2-phenylethylidene)-1*H*-indene-1,3(2*H*)-dione (1ga**):**



Prepared according to **TP 1** from 2-(4-(cyano)benzylidene)-1*H*-indene-1,3(2*H*)-dione **6g** (77.8 mg, 0.3 mmol), MePPh₂ (5.7 μL, 10 mol%), benzoyl chloride **11a** (42.6 μL, 1.2 equiv) and Et₃N (54.4 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 0.5 hour]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1ga** as yellow solid (107.7 mg, 99%). mp: 221.9–222.9 °C; R_f 0.18 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.01–7.92 (m, 3H), 7.92–7.81 (m, 3H), 7.78 (d, 2H, J = 8.3 Hz), 7.71 (d, 2H, J = 8.1 Hz), 7.59 (t, 1H, J = 7.3 Hz), 7.47 (t, 2H, J = 7.7 Hz)

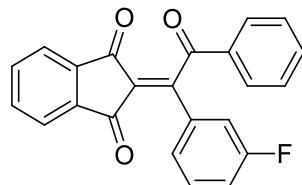
¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 194.4, 188.2, 187.0, 155.5, 142.3, 140.4, 136.2, 136.1, 135.3, 134.7, 134.3, 132.0, 130.0, 129.3, 129.1, 129.0, 128.9, 123.7, 123.7, 118.0, 114.4.

MS (70 eV, EI) m/z (%): 363 [M]⁺ (67), 105 (100), 77 (88).

IR (KBr) ν (cm⁻¹): 3044 (w), 2920 (w), 2233 (m), 1730 (m), 1694 (s), 1668 (s), 1596 (m), 1452 (w).

HRMS (EI) for C₂₄H₁₃NO₃ [M]⁺ (363.0895) found 363.0902.

Synthesis of 2-(1-(3-fluorophenyl)-2-oxo-2-phenylethylidene)-1*H*-indene-1,3(2*H*)-dione (1ha**):**



Prepared according to **TP 1** from 2-(3-fluorobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6h** (75.7 mg, 0.3 mmol), MePPh₂ (5.7 μL, 10 mol%), benzoyl chloride **11a** (42.6 μL, 1.2 equiv) and Et₃N (54.4 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 1 hour]. Purification by flash chromatography (hexanes/dichloromethane: 2/3) furnished **1ha** as yellow solid (97.0 mg, 91%). mp: 194.8–195.6 °C; R_f 0.35 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 7.90 (dd, 1H, J = 12.8, 7.8 Hz), 7.81 (d, 2H, J = 7.2 Hz), 7.80–7.71 (m, 2H), 7.50 (t, 1H, J = 7.0 Hz), 7.44–7.30 (m, 5H), 7.11 (t, 1H, J = 8.2 Hz).

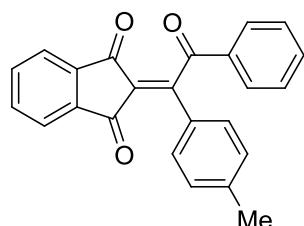
¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 194.8, 188.7, 187.1, 162.3 (d, J = 247.7 Hz), 157.0, 157.0, 124.4, 140.2, 136.0, 135.8, 135.0, 134.0, 132.8 (d, J = 8.1 Hz), 130.0 (d, J = 8.1 Hz), 129.0, 128.4, 125.6 (d, J = 3.1 Hz), 123.6, 123.5, 118.4 (d, J = 21.1 Hz), 116.7 (d, J = 23.4 Hz).

MS (70 eV, EI) m/z (%): 356 [M]⁺ (19), 105 (100), 77 (15).

IR (KBr) ν (cm⁻¹): 3070 (w), 2922 (w), 1731 (m), 1693 (s), 1667 (s), 1581 (m), 1480 (w), 1255 (s).

HRMS (EI) for C₂₃H₁₃FO₃ [M]⁺ (356.0849) found 356.0846.

Synthesis of 2-(2-oxo-2-phenyl-1-(p-tolyl)ethylidene)-1*H*-indene-1,3(2*H*)-dione (1ia**):**



Prepared according to **TP 1** from 2-(4-methylbenzylidene)-1*H*-indene-1,3(2*H*)-dione **6i** (74.4 mg, 0.3 mmol), MePPh₂ (8.7 µL, 15 mol%), benzoyl chloride **11a** (42.6 µL, 1.2 equiv) and Et₃N (54.4 µL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 2 hours]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1ia** as yellow solid (106.3 mg, 99%). mp: 166.6–167.4 °C; R_f 0.30 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 7.99–7.93 (m, 3H), 7.85 (d, 1H, J = 6.8 Hz), 7.83–7.73 (m, 2H), 7.65 (d, 2H, J = 8.2 Hz), 7.53 (t, 1H, J = 7.4 Hz), 7.43 (t, 2H, J = 7.7 Hz), 7.24 (d, 2H, J = 8.3 Hz), 2.38 (s, 3H).

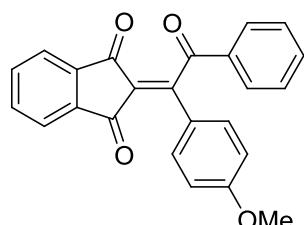
¹³C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 195.4, 189.1, 187.4, 159.1, 142.7, 142.2, 139.8, 135.5, 135.4, 135.2, 133.5, 130.2, 129.0, 128.8, 128.7, 127.9, 126.9, 123.2, 123.1, 21.5.

MS (70 eV, EI) m/z (%): 352 [M]⁺ (35), 105 (100), 77 (17).

IR (KBr) $\tilde{\nu}$ (cm^{−1}): 3061 (w), 2924 (w), 1726 (m), 1691 (s), 1672 (s), 1600 (m), 1451 (m).

HRMS (ESI) for C₂₄H₁₇O₃ [M+H]⁺ (353.1178) found 353.1176.

Synthesis of 2-(1-(4-methoxyphenyl)-2-oxo-2-phenylethylidene)-1*H*-indene-1,3(2*H*)-dione (1ja**):**



Prepared according to **TP 1** from 2-(4-methoxybenzylidene)-1*H*-indene-1,3(2*H*)-dione **6j** (79.3 mg, 0.3 mmol), MePPh₂ (8.7 µL, 15 mol%), benzoyl chloride **11a** (42.6 µL, 1.2 equiv) and Et₃N (54.4 µL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 3 hours]. Purification by flash chromatography (hexanes/dichloromethane: 1/2) furnished **1ja** as yellow solid (103.5 mg, 94%). mp: 165.4–166.4 °C; R_f 0.15 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.00–7.92 (m, 3H), 7.86–7.73 (m, 5H), 7.54 (t, 1H, J = 7.4 Hz), 7.43 (t, 2H, J = 7.6 Hz), 6.95 (d, 2H, J = 8.9 Hz), 3.86 (s, 3H).

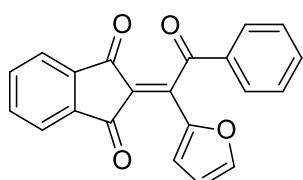
¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 195.9, 189.4, 187.8, 163.0, 159.0, 142.2, 139.8, 135.6, 135.4, 135.3, 133.5, 133.2, 128.9, 128.7, 125.8, 123.2, 123.1, 113.9, 55.4.

MS (70 eV, EI) m/z (%): 368 [M]⁺ (67), 263 (100), 235 (30), 105 (77), 77 (67).

IR (KBr) $\tilde{\nu}$ (cm^{−1}): 3061 (w), 3016 (w), 2935 (w), 1726 (m), 1687 (s), 1596 (m), 1452 (w), 1258 (s).

HRMS (FAB) for $\mathbf{C}_{24}\mathbf{H}_{17}\mathbf{O}_4 [\mathbf{M}+\mathbf{H}]^+$ (369.1127) found 369.1125.

Synthesis of 2-(1-(furan-2-yl)-2-oxo-2-phenylethylidene)-1*H*-indene-1,3(2*H*)-dione (1ka**):**



Prepared according to **TP 1** from 2-(furan-2-ylmethylene)-1*H*-indene-1,3(2*H*)-dione **6k** (67.3 mg, 0.3 mmol), MePPh₂ (8.7 μ L, 15 mol%), benzoyl chloride **11a** (42.6 μ L, 1.2 equiv) and Et₃N (54.4 μ L, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 7 hours]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1ka** as yellow solid (32.6 mg, 33%). mp: 288.1–289.0 °C; R_f 0.25 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.81 (d, 1H, J = 3.6 Hz), 8.03–7.93 (m, 3H), 7.97 (t, 2H, J = 6.5 Hz), 7.73 (t, 1H, J = 7.3 Hz), 7.65 (s, 1H), 7.58 (t, 1H, , J = 7.3 Hz), 7.47 (t, 2H, , J = 7.6 Hz), 6.73 (d, 1H, J = 2.5 Hz).

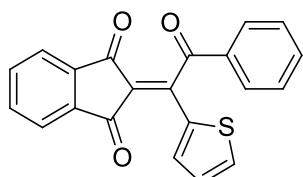
¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 193.8, 189.4, 188.5, 149.5, 148.6, 144.0, 142.4, 140.1, 136.2, 135.3, 135.2, 133.7, 128.8, 128.6, 126.4, 123.1, 123.1, 122.5, 114.3.

MS (70 eV, EI) m/z (%): 328 [M]⁺ (10), 230 (74), 223 (5), 105 (100), 77 (57).

IR (KBr) $\tilde{\nu}$ (cm^{−1}): 2924 (w), 1718 (m), 1680 (s), 1562 (s), 1448 (s), 1250 (s).

HRMS (FAB) for $\mathbf{C}_{21}\mathbf{H}_{13}\mathbf{O}_4 [\mathbf{M}+\mathbf{H}]^+$ (329.0814) found 329.0821.

Synthesis of 2-(2-oxo-2-phenyl-1-(thiophen-2-yl)ethylidene)-1*H*-indene-1,3(2*H*)-dione (1la**):**



Prepared according to **TP 1** from 2-(thiophen-2-ylmethylene)-1*H*-indene-1,3(2*H*)-dione **6l** (72.1 mg, 0.3 mmol), MePPh₂ (8.7 μ L, 15 mol%), benzoyl chloride **11a** (42.6 μ L, 1.2 equiv) and Et₃N (54.4 μ L, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 7 hours]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1la** as yellow solid (54.5 mg, 53%). mp: 235.9–236.5 °C; R_f 0.50 (hexanes/dichloromethane: 1/1).

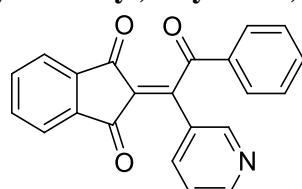
¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 7.97 (dd, 1H, J = 6.4, 1.3 Hz), 7.88–7.75 (m, 5H), 7.61–7.53 (m, 6H).

¹³C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 195.6, 189.4, 188.8, 150.2, 142.2, 140.0, 139.9, 138.0, 136.1, 135.4, 135.3, 135.1, 133.8, 128.9, 128.9, 128.4, 123.2, 122.4.

IR (KBr) $\tilde{\nu}$ (cm^{−1}): 3092 (w), 1718 (m), 1680 (s), 1581 (m), 1448 (m).

HRMS (EI) for $\mathbf{C}_{21}\mathbf{H}_{12}\mathbf{O}_3\mathbf{S} [\mathbf{M}]^+$ (344.0507) found 344.0510

Synthesis of 2-(2-oxo-2-phenyl-1-(pyridin-3-yl)ethylidene)-1*H*-indene-1,3(2*H*)-dione (1ma):



Prepared according to **TP 1** from 2-(pyridin-3-ylmethylene)-*1H*-indene-1,3(2*H*)-dione **6m** (70.6 mg, 0.3 mmol), MePPh₂ (5.7 μL, 10 mol%), benzoyl chloride **11a** (42.6 μL, 1.2 equiv) and Et₃N (54.4 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 1 hour]. Purification by flash chromatography (hexanes/dichloromethane: 1/5) furnished **1ma** as yellow solid (96.0 mg, 94%). mp: 203.8–204.7 °C; R_f 0.04 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.95 (s, 1H), 8.71 (d, 1H, J = 4.6 Hz), 8.09 (d, 1H, J = 8.0 Hz), 8.01 (d, 1H, J = 6.9 Hz), 7.96 (d, 2H, J = 7.8 Hz), 7.93–7.81 (m, 3H), 7.60 (t, 1H, J = 7.3 Hz), 7.52–7.44 (m, 3H).

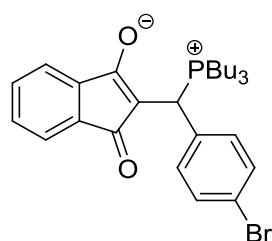
¹³C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 194.6, 188.3, 187.2, 154.6, 151.5, 149.9, 142.3, 140.3, 136.7, 136.0, 136.0, 134.8, 134.2, 129.0, 129.0, 127.3, 123.6, 123.0.

MS (70 eV, EI) m/z (%): 339 [M]⁺ (100), 105 (18), 77 (7).

IR (KBr) ν (cm⁻¹): 3048 (w), 1727 (m), 1687 (s), 1676 (s), 1613 (m), 1591 (m), 1447 (m), 1244 (s).

HRMS (FAB) for C₂₂H₁₄NO₃ [M+H]⁺ (340.0974) found 340.0969.

Synthesis of 2-((4-bromophenyl)(tributylphosphonio)methyl)-1-oxo-*1H*-inden-3-olate (12a):



A dry and nitrogen-flushed 10-mL Schlenk tube, equipped with a magnetic stirring bar and a septum, was sequentially charged with a solution of 2-(4-bromobenzylidene)-*1H*-indene-1,3(2*H*)-dione **6a** (93.9 mg, 0.3 mmol), Bu₃P (67.0 μL, 1.2 equiv) in dry THF (1.5 mL). The reaction mixture was stirred for less than 1 hour at room temperature (27–30 °C). Thereafter, the solvent was removed by evaporation *in vacuo*. Purification by flash chromatography (hexanes/ethyl acetate: 1/1) furnished **12a** as orange solid (143.8 mg, 93%).

mp: 187.7–190.2 °C; R_f 0.30 (hexanes/ ethyl acetate: 1/1).

¹H-NMR (500 MHz, CDCl₃, 25 °C) δ/ppm: 7.54 (dd, 2H, J = 8.5, 2.1 Hz), 7.43 (d, 2H, J = 8.3 Hz), 7.37–7.33 (m, 2H), 7.32–7.27 (m, 2H), 4.91 (d, 1H, J = 17.7 Hz), 2.36–2.13 (m, 6H), 1.46–1.29 (m, 12H), 0.91–0.85 (m, 9H).

¹³C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 189.9 (d, J = 4.6 Hz), 139.1, 134.9, 131.4, 131.3 (d, J = 4.5 Hz), 129.4, 121.6 (d, J = 3.7 Hz), 117.4, 97.9, 34.8 (d, J = 44.1 Hz), 23.6 (d, J = 14.8 Hz),

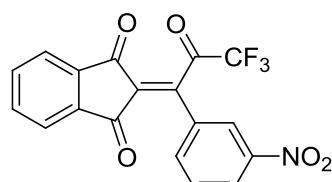
23.5 (d, $J = 5.0$ Hz), 19.3 (d, $J = 44.1$ Hz), 12.8.

$^{31}\text{P-NMR}$ (200 MHz, CDCl_3 , 25 °C) δ/ppm: 33.4.

IR (KBr) $\tilde{\nu}$ (cm^{-1}): 3055 (w), 3030 (w), 2929 (m), 1647 (w), 1615 (m), 1550 (s), 1487 (m).

HRMS (FAB) for $\text{C}_{28}\text{H}_{37}\text{O}_2\text{Br} [\text{M}+\text{H}]^+$ (515.1715) found 515.1719.

Synthesis of 2-(3,3,3-trifluoro-1-(3-nitrophenyl)-2-oxopropylidene)-1*H*-indene-1,3(2*H*)-dione (1cm):



Prepared according to **TP 2** from 2-(3-nitrobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6c** (83.8 mg, 0.3 mmol), EtPPh₂ (9.4 μL, 15 mol%), **11m** (54.2 μL, 1.3 equiv) and Et₃N (58.9 μL, 1.4 equiv) in THF (1.5 mL). [reaction condition: 27-30 °C for 10 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1cm** as yellow solid (104.3 mg, 93%).

mp: 153.2-154.1 °C; R_f 0.40 (hexanes/dichloromethane: 1/1).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 °C) δ/ppm: 8.56-8.52 (m, 1H), 8.44 (d, 1H, $J = 8.3$ Hz), 8.10-8.06 (m, 1H), 8.06-8.01 (m, 1H), 7.97-7.92 (m, 2H), 7.90 (d, 1H, $J = 7.8$ Hz), 7.72 (t, 1H, $J = 8.0$ Hz).

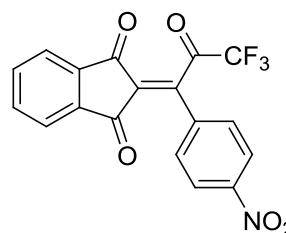
$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 °C) δ/ppm: 188.5, 186.7 (q, $J = 39.4$ Hz), 185.2, 148.2, 147.2, 143.3, 139.6, 137.1, 136.8, 135.1, 132.6, 129.9, 129.1, 126.6, 125.1, 124.3, 115.0 (q, $J = 291.3$ Hz).

MS (70 eV, EI) m/z (%): 375 [$\text{M}]^+$ (36), 307 (25), 278 (100), 251 (30), 176 (29), 104 (4), 69 (24).

IR (KBr) $\tilde{\nu}$ (cm^{-1}): 3094 (w), 3070 (w), 2876 (w), 1734 (s), 1608 (m), 1590 (m), 1538 (s), 1483 (w), 1353 (s), 1261 (s).

HRMS (EI) for $\text{C}_{18}\text{H}_{8}\text{F}_3\text{NO}_5 [\text{M}]^+$ (375.0355) found 375.0344.

Synthesis of 2-(3,3,3-trifluoro-1-(4-nitrophenyl)-2-oxopropylidene)-1*H*-indene-1,3(2*H*)-dione (1dm):



Prepared according to **TP 2** from 2-(4-nitrobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6d** (83.8 mg, 0.3 mmol), EtPPh₂ (9.4 μL, 15 mol%), **11m** (54.2 μL, 1.3 equiv) and Et₃N (58.9 μL, 1.4 equiv) in THF (1.5 mL). [reaction condition: 27-30 °C for 10 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1dm** as yellow solid (92.2 mg, 82%).

mp: 154.5-154.9 °C; R_f 0.34 (hexanes/dichloromethane: 1/1).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 °C) δ/ppm: 8.35 (d, 2H, $J = 8.8$ Hz), 8.11-8.00 (m, 2H), 7.99-7.92 (m, 2H), 7.78 (d, 2H, $J = 8.8$ Hz).

$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 °C) δ/ppm: 188.4, 186.7 (q, $J = 39.3$ Hz), 185.1, 149.5, 147.2,

143.3, 139.6, 137.2, 136.8, 133.6, 132.9, 130.8, 124.3, 124.2, 123.7, 115.0 (q, $J = 291.3$ Hz).

MS (70 eV, EI) m/z (%): 375 [M]⁺ (8), 307 (72), 279 (100), 250 (15), 176 (23), 76 (19), 69 (59).

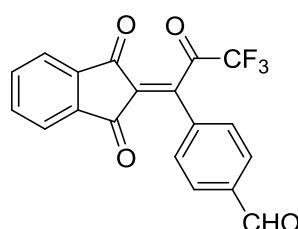
IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3092 (w), 3023 (w), 2924 (w), 1741 (m), 17023 (s), 1634 (m), 1600 (m), 1528 (s), 1349 (s), 1219 (s).

HRMS (EI) for **C₁₈H₈F₃NO₅** [M]⁺ (375.0355) found 375.0366.

Synthesis

of

4-(1-(1,3-dioxo-1*H*-inden-2(3*H*)-ylidene)-3,3,3-trifluoro-2-oxopropyl)benzaldehyde (**1em**):



Prepared according to **TP 2** from 4-((1,3-dioxo-1*H*-inden-2(3*H*)-ylidene)methyl)benzaldehyde **6e** (79.6 mg, 0.3 mmol), EtPPh₂ (9.4 μ L, 15 mol%), **11m** (54.2 μ L, 1.3 equiv) and Et₃N (58.9 μ L, 1.4 equiv) in THF (1.5 mL). [reaction condition: 27-30 °C for 10 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 1/2) furnished **1em** as yellow solid (67.6 mg, 63%).

mp: 132.8-134.2; R_f 0.21 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 10.13 (s, 1H), 8.11-8.00 (m, 4H), 7.98-7.91 (m, 2H), 7.79 (d, 2H, $J = 8.3$ Hz).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 191.2, 188.7, 186.9 (q, $J = 39.1$ Hz), 185.3, 148.9, 143.3, 139.6, 138.3, 137.0, 136.6, 133.0, 132.2, 130.5, 129.6, 124.2, 124.1, 115.0 (q, $J = 291.5$ Hz).

MS (70 eV, EI) m/z (%): 358 [M]⁺, 289 (55), 261 (100), 176 (39), 104 (9), 76 (14), 69 (77).

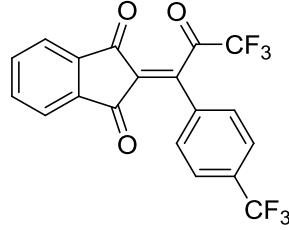
IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3061 (w), 2924 (w), 1745 (m), 1723 (s), 1611 (s), 1589 (m), 1215 (s).

HRMS (FAB) for **C₁₉H₁₀F₃O₄** [M+H]⁺ (359.0531) found 359.0527.

Synthesis

of

2-(3,3,3-trifluoro-2-oxo-1-(4-(trifluoromethyl)phenyl)propylidene)-1*H*-indene-1,3(2*H*)-dione (**1fm**):



Prepared according to **TP 2** from 2-(4-(trifluoromethyl)benzylidene)-1*H*-indene-1,3(2*H*)-dione **6f** (90.7 mg, 0.3 mmol), EtPPh₂ (9.4 μ L, 15 mol%), **11m** (54.2 μ L, 1.3 equiv) and Et₃N (58.9 μ L, 1.4 equiv) in THF (1.5 mL). [reaction condition: 27-30 °C for 10 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1fm** as yellow solid (102.3 mg, 86%).

mp: 127.1-128.0 °C; R_f 0.56 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.09-7.99 (m, 2H), 7.97-7.89 (m, 2H), 7.80-7.71 (m,

4H).

¹³C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 187.6, 164.1, 150.1 (q, *J* = 35.6 Hz), 140.2, 136.7, 135.1, 134.9, 133.4, 132.6, 131.9 (q, *J* = 32.9 Hz), 128.8, 127.8, 125.3, 125.2 (q, *J* = 3.7 Hz), 123.8 (q, *J* = 272.5 Hz), 123.2, 121.4 (q, *J* = 276.7 Hz).

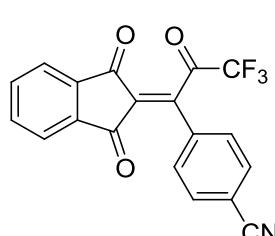
MS (70 eV, EI) m/z (%): 398 [M]⁺, 329 (73), 301 (100), 273 (34), 225 (31), 197 (32), 176 (27), 147 (8), 104 (6), 76 (14), 69 (24).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3092 (w), 2931 (w), 1745 (s), 1730 (s), 1695 (s), 1627 (m), 1589 (m), 1219 (s).

HRMS (EI) for C₁₉H₈F₆O₃ [M]⁺ (398.0378) found 398.0378.

Synthesis of 2-(3,3,3-cyano-2-oxo-1-(4-(trifluoromethyl)phenyl)propylidene)-1*H*-indene-1,3(2*H*)-dione

(**1gm**):



Prepared according to **TP 2** from 2-(4-(cyano)benzylidene)-1*H*-indene-1,3(2*H*)-dione **6g** (77.8 mg, 0.3 mmol), EtPPh₂ (9.4 μL, 15 mol%), **11m** (54.2 μL, 1.3 equiv) and Et₃N (58.9 μL, 1.4 equiv) in THF (1.5 mL). [reaction condition: 27-30 °C for 10 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **1gm** as yellow solid (108.8 mg, 97%).

mp: 191.4-192.1 °C; R_f 0.25 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.10-7.98 (m, 2H), 7.98-7.90 (m, 2H), 7.80 (d, 2H, *J* = 8.3 Hz), 7.72 (d, 2H, *J* = 8.3 Hz).

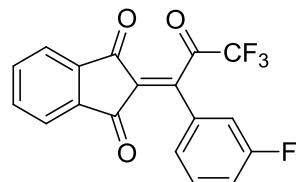
¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 188.5, 186.7 (q, *J* = 39.2 Hz), 185.1, 147.6, 143.2, 139.5, 137.1, 136.7, 132.5, 132.2, 131.8, 130.3, 130.3, 124.2, 124.1, 117.7, 114.9 (q, *J* = 291.4 Hz), 117.7.

MS (70 eV, EI) m/z (%): 355 [M]⁺ (11), 286 (100), 258 (90), 154 (47), 103 (14), 75 (44).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3085 (w), 3018 (w), 2930 (w), 2856 (w), 2229 (m), 1749 (m), 1731 (m), 1628 (m), 1587 (m), 1218 (s).

HRMS (EI) for C₁₉H₈F₃NO₃ [M]⁺ (355.0456) found 355.0460.

Synthesis of 2-(3,3,3-trifluoro-1-(3-fluorophenyl)-2-oxopropylidene)-1*H*-indene-1,3(2*H*)-dione (**1hm**):



Prepared according to **TP 2** from 2-(3-fluorobenzylidene)-1*H*-indene-1,3(2*H*)-dione **6h** (75.7 mg, 0.3 mmol), EtPPh₂ (9.4 μL, 15 mol%), **11m** (54.2 μL, 1.3 equiv) and Et₃N (58.9 μL, 1.4 equiv) in

THF (1.5 mL). [reaction condition: 27-30 °C for 10 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 2/1) furnished **1hm** as yellow solid (89.6 mg, 86%).
mp: 128.5-129.2 °C; R_f 0.55 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.08-7.99 (m, 2H), 7.95-7.88 (m, 2H), 7.54-7.46 (m, 1H), 7.44-7.37 (m, 2H), 7.33-7.27 (m, 1H).

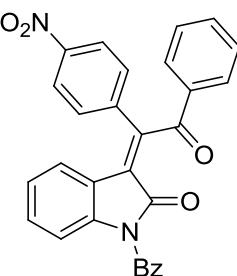
¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 189.0, 187.1 (q, J = 38.9 Hz), 185.3, 162.4 (d, J = 248.6 Hz), 149.2, 149.2, 143.3, 139.4, 136.6 (d, J = 44.0 Hz), 131.4, 130.4 (d, C, J = 8.1 Hz), 129.3 (d, J = 8.3 Hz), 125.9 (d, J = 3.0 Hz), 124.1, 124.0, 119.6 (d, J = 21.1 Hz), 117.2 (d, J = 23.8 Hz), 115.0 (q, J = 291.6 Hz).

MS (70 eV, EI) m/z (%): 348 [M]⁺ (37), 279 (75), 251 (100), 175 (29), 147 (55), 104 (15), 98 (30), 75 (52), 69 (84).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3077 (w), 2923 (w), 1746 (m), 1731 (m), 1694 (s), 1583 (m), 1222 (s).

HRMS (EI) for C₁₈H₈F₄O₃ [M]⁺ (348.0410) found 348.0406.

Synthesis of (*Z*)-1-benzoyl-3-(1-(4-nitrophenyl)-2-oxo-2-phenylethylidene)indolin-2-one (**2a**):



Prepared according to **TP 3** from (*Z*)-1-benzoyl-3-(4-nitrobenzylidene)indolin-2-one **7a** (111.1 mg, 0.3 mmol), Bu₃P (29.8 μL, 10 mol%), **11a** (38.3 μL, 1.1 equiv) and Et₃N (50.2 μL, 1.2 equiv) in THF (1.5 mL) [reaction condition: 27-30 °C for 50 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **2a** as yellow solid (118.9 mg, 80%).

mp: 232.5-233.1 °C; R_f 0.25 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.35 (d, 2H, J = 8.7 Hz), 7.96 (d, 2H, J = 7.8 Hz), 7.82 (t, 3H, J = 8.6 Hz), 7.69 (d, 2H, J = 7.8 Hz), 7.58-7.49 (m, 2H), 7.47-7.34 (m, 5H), 6.99 (t, 1H, J = 7.8 Hz), 6.86 (d, 1H, J = 7.9 Hz).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 194.4, 168.7, 165.2, 148.5, 147.4, 141.7, 140.0, 134.4, 134.0, 133.2, 131.5, 129.7, 129.2, 129.0, 128.9, 128.2, 126.9, 124.6, 124.5, 123.2, 120.7, 115.4.

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3116 (w), 3053 (w), 1736 (s), 1694 (s), 1671 (m), 1597 (m), 1519 (s), 1462 (m), 1341 (s).

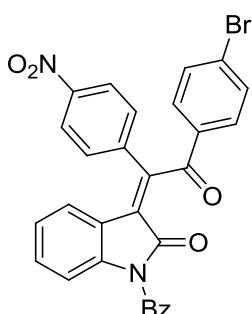
HRMS (EI) for C₂₉H₁₈N₂O₅ [M]⁺ (474.1216) found 474.1210.

CCDC: 935873.

Synthesis

of

(Z)-1-benzoyl-3-(2-(4-bromophenyl)-1-(4-nitrophenyl)-2-oxoethylidene)indolin-2-one (2b):



Prepared according to **TP 3** from (Z)-1-benzoyl-3-(4-nitrobenzylidene)indolin-2-one **7a** (111.1 mg, 0.3 mmol), Bu₃P (29.8 μL, 10 mol%), **11f** (73.9 μL, 1.1 equiv) and Et₃N (50.2 μL, 1.2 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 30 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **2b** as yellow solid (120.7 mg, 71%).

mp: 264.7–265.3 °C; R_f 0.32 (hexanes/dichloromethane: 1/1).

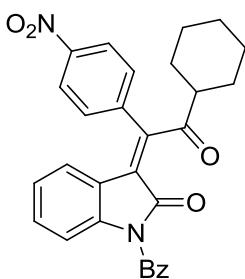
¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.35 (d, 2H, J = 9.7 Hz), 7.85–7.77 (m, 5H), 7.69 (d, 2H, J = 7.3 Hz), 7.60–7.51 (m, 3H), 7.44–7.35 (m, 3H), 7.00 (t, 1H, J = 7.7 Hz), 6.87 (d, 1H, J = 7.8 Hz).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 193.5, 168.7, 165.2, 148.2, 146.6, 141.9, 139.7, 133.4, 133.3, 133.2, 132.3, 131.7, 130.4, 129.7, 129.4, 129.2, 128.3, 127.2, 124.8, 124.6, 123.2, 120.6, 115.5.

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 1743 (s), 1690 (m), 1664 (m), 1600 (m), 1518 (s), 1464 (m), 1341 (s), 594 (m).

HRMS (EI) for C₂₉H₁₇BrN₂O₅ [M]⁺ (552.0321) found 552.0321.

Synthesis of (Z)-1-benzoyl-3-(2-cyclohexyl-1-(4-nitrophenyl)-2-oxoethylidene)indolin-2-one (2c):



Prepared according to **TP 3** from (Z)-1-benzoyl-3-(4-nitrobenzylidene)indolin-2-one **7a** (111.1 mg, 0.3 mmol), Bu₃P (29.8 μL, 10 mol%), **11k** (45.5 μL, 1.1 equiv) and Et₃N (50.2 μL, 1.2 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 1 hour]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **2c** as yellow solid (135.4 mg, 94%).

mp: 211.0–212.0 °C; R_f 0.30 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.38 (d, 2H, J = 8.5 Hz), 7.82–7.74 (m, 5H), 7.62 (t, 1H, J = 7.5 Hz), 7.48 (t, 2H, J = 7.7 Hz), 7.35 (t, 1H, J = 7.9 Hz), 6.93 (t, 1H, J = 7.7 Hz), 6.73 (d, 1H, J = 7.9 Hz), 2.63 (tt, 1H, J = 11.2, 3.2 Hz), 1.88 (d, 2H, J = 10.3 Hz), 1.67 (d, 2H, J = 12.0 Hz), 1.59 (d, 1H, J = 12.6 Hz), 1.29–1.02 (m, 5H).

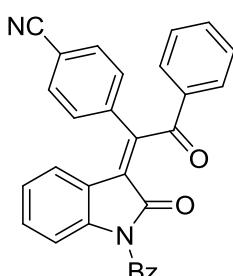
¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 207.8, 168.8, 165.6, 150.1, 148.6, 141.5, 140.3, 133.6,

133.3, 131.2, 129.8, 129.0, 128.3, 125.3, 124.6, 124.5, 123.0, 120.9, 115.3, 50.7, 28.4, 25.6.

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3107 (w), 2924 (m), 1729 (s), 1685 (s), 1601 (m), 1520 (m), 1461 (m), 1343 (s).

HRMS (EI) for $\mathbf{C}_{29}\mathbf{H}_{24}\mathbf{N}_2\mathbf{O}_5 [\mathbf{M}]^+$ (480.1685) found 480.1679.

Synthesis of (*Z*)-4-(1-(1-benzoyl-2-oxoindolin-3-ylidene)-2-oxo-2-phenylethyl)benzonitrile (2d**):**



Prepared according to **TP 3** from (*Z*)-4-((1-benzoyl-2-oxoindolin-3-ylidene)methyl)benzonitrile **7b** (105.12 mg, 0.3 mmol), Bu₃P (8.7 μ L, 10 mol%), **11a** (38.3 μ L, 1.1 equiv) and Et₃N (50.2 μ L, 1.2 equiv) in THF (1.5 mL) [reaction condition: 27-30 °C for 3.5 hours]. Purification by flash chromatography (hexanes/dichloromethane: 1/2) furnished **2a** as yellow solid (76.4 mg, 56%). mp: 208.1-208.9 °C; R_f 0.12 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ /ppm: 7.95 (d, 2H, J = 7.2 Hz), 7.83 (d, 1H, J = 8.2 Hz), 7.76 (dd, 4H, J = 19.6, 8.3 Hz), 7.68 (d, 2H, J = 7.2 Hz), 7.53 (m, 2H), 7.40 (m, 5H), 7.00 (t, 1H, J = 7.2 Hz), 6.85 (d, 1H, J = 7.8 Hz).

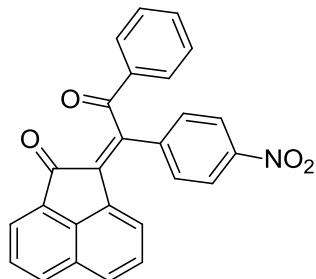
¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ /ppm: 194.5, 168.8, 165.3, 147.8, 141.8, 138.3, 134.5, 134.0, 133.3, 133.3, 133.2, 131.5, 129.8, 129.1, 129.0, 128.9, 128.2, 126.7, 124.6, 123.2, 120.8, 117.9, 115.5, 113.9.

MS (70 eV, EI) m/z (%): 454 (68), 104 (100), 79 (13).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3084 (w), 3053 (w), 2221 (m), 1749 (s), 1680 (s), 1669 (s), 1596 (m), 1459 (s).

HRMS (ESI) for $\mathbf{C}_{30}\mathbf{H}_{18}\mathbf{N}_2\mathbf{O}_3\mathbf{Na}, [\mathbf{M}+\mathbf{Na}]^+$ (477.1215) found: 477.1219.

Synthesis of (*E*)-2-(1-(4-nitrophenyl)-2-oxo-2-phenylethylidene)acenaphthylen-1(2*H*)-one (3a**):**



Prepared according to **TP 3** from (*E*)-2-(4-nitrobenzylidene)acenaphthylen-1(2*H*)-one **8a** (90.3 mg, 0.3 mmol), Bu₃P (17.4 μ L, 20 mol%), **11a** (42.6 μ L, 1.2 equiv) and Et₃N (54.7 μ L, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27-30 °C for 50 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **3a** as yellow solid (97.5 mg, 80%).

mp: 230.8-231.2 °C; R_f 0.23 (hexanes/dichloromethane: 1/1).

¹H-NMR (500 MHz, CDCl₃, 25 °C) δ/ppm: 8.28 (d, 2H, J = 8.7 Hz), 8.03 (m, 3H), 7.87 (d, 1H, J = 7.0 Hz), 7.83 (m, 3H), 7.66 (t, 1H, J = 7.5 Hz), 7.50 (t, 1H, J = 7.4 Hz), 7.40 (t, 3H, J = 7.4 Hz), 6.98 (d, 1H, J = 7.2 Hz).

¹³C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 196.1, 191.1, 148.4, 144.1, 141.5, 140.9, 135.3, 134.4, 133.9, 131.9, 131.4, 130.9, 130.8, 129.7, 129.1, 129.0, 129.0, 128.4, 128.1, 127.1, 124.5, 122.3, 120.8.

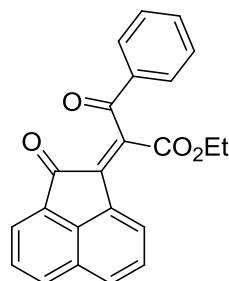
MS (70 eV, EI) m/z (%): 405 [M]⁺ (40), 226 (22), 105 (100), 77 (65).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3107 (w), 1707 (s), 1686 (w), 1601 (w), 1517 (m), 1491 (w), 1344 (s).

HRMS (FAB) for C₂₆H₁₆NO₄ [M+H]⁺ (406.1079) found: 406.1070.

CCDC: 935872.

Synthesis of (*E*)-ethyl 3-oxo-2-(2-oxoacenaphthylen-1(2*H*)-ylidene)-3-phenylpropanoate (3b):



Prepared according to **TP 3** from (*E*)-(2-oxoacenaphthylen-1(2*H*)-ylidene)methyl propionate **8b** (75.7 mg, 0.3 mmol), Bu₃P (17.4 μL, 20 mol%), **11a** (42.6 μL, 1.2 equiv) and Et₃N (54.7 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 50 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **3b** as yellow solid (76.0 mg, 71%). mp: 138.8–139.7 °C; R_f 0.30 (hexanes/dichloromethane: 1/1).

¹H-NMR (500 MHz, CDCl₃, 25 °C) δ/ppm: 8.70 (d, 1H, J = 7.3 Hz), 8.17 (d, 1H, J = 8.1 Hz), 8.07 (m, 3H), 7.93 (d, 1H, J = 6.9 Hz), 7.79 (t, 1H, J = 7.7 Hz), 7.74 (t, 1H, J = 7.3 Hz), 7.60 (m, 1H), 7.50 (t, 2H, J = 7.9 Hz), 4.38 (q, 2H, J = 7.1 Hz), 1.26 (t, 3H, J = 7.1 Hz).

¹³C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 193.9, 191.6, 164.2, 142.9, 140.5, 136.5, 135.2, 133.4, 132.0, 130.7, 130.6, 129.9, 128.7, 128.7, 128.6, 128.2, 128.0, 125.5, 122.2, 62.3, 13.9.

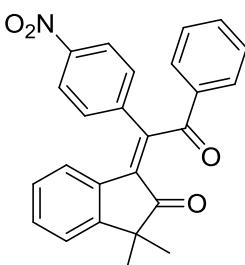
IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3062 (w), 2957 (w), 1706 (s), 1677 (s), 1596 (m), 1443 (w), 1241 (s).

HRMS (ESI) for C₂₃H₁₆O₄Na, [M+Na]⁺ (379.0947) found: 379.0961

Synthesis

of

(Z)-1,1-dimethyl-3-(1-(4-nitrophenyl)-2-oxo-2-phenylethylidene)-1*H*-inden-2(3*H*)-one (4):



Prepared according to **TP 3** from (*E*)-1,1-dimethyl-3-(4-nitrobenzylidene)-*1H*-inden-2(3*H*)-one **9** (87.9 mg, 0.3 mmol), Bu₃P (17.4 μL, 20 mol%), **11a** (42.6 μL, 1.2 equiv) and Et₃N (54.4 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 3 hours]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **4** as yellow solid (84.3 mg, 71%). mp: 174.3–175.0 °C; R_f 0.50 (hexanes/dichloromethane: 1/1).

¹H-NMR (500 MHz, CDCl₃, 25 °C) δ/ppm: 8.24 (d, 2H, J = 8.8 Hz), 7.93 (d, 2H, J = 7.2 Hz), 7.74 (d, 2H, J = 8.8 Hz), 7.49 (t, 1H, J = 7.5 Hz), 7.39 (t, 2H, J = 7.7 Hz), 7.32 (t, 1H, J = 7.5 Hz), 7.28 (t, 1H, J = 7.2 Hz), 7.01 (t, 1H, J = 7.7 Hz), 6.80 (d, 1H, J = 7.8 Hz), 1.26 (s, 6H).

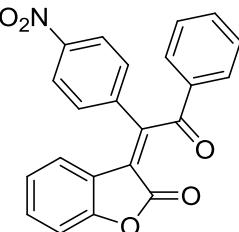
¹³C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 208.4, 196.1, 150.9, 148.2, 142.2, 141.0, 135.1, 133.7, 133.6, 133.3, 131.1, 129.3, 128.9, 127.5, 124.6, 123.8, 123.7, 47.1, 25.3.

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3091 (w), 3066 (w), 2961 (m), 1729 (s), 1667 (s), 1594 (m), 1520 (s), 1468 (w), 1346 (s).

HRMS (EI) for C₂₅H₁₉NO₄ [M]⁺ (397.1314) found 397.1306.

CCDC: 935873.

Synthesis of (Z)-3-(1-(4-nitrophenyl)-2-oxo-2-phenylethylidene)benzofuran-2(3*H*)-one ((Z)-5):



Prepared according to **TP 3** from (*E*)-3-(4-nitrobenzylidene)benzofuran-2(3*H*)-one **10** (80.2 mg, 0.3 mmol), Bu₃P (17.4 μL, 20 mol%), **11a** (42.6 μL, 1.2 equiv) and Et₃N (54.4 μL, 1.3 equiv) in THF (1.5 mL) [reaction condition: 27–30 °C for 3 hours]. Purification by flash chromatography (hexanes/dichloromethane/ethyl acetate: 20/5/1) furnished mixture of *E* and *Z*-form **5** as yellow solid (92.2 mg, 83%).

mp: 180.2–180.6 °C; R_f 0.48 (hexanes/dichloromethane: 1/1).

¹H-NMR (500 MHz, CDCl₃, 25 °C) δ/ppm: 8.35 (d, 2H, J = 9.7 Hz), 8.02 (d, 2H, J = 7.4 Hz), 7.85 (dt, 2H, J = 9.3, 2.2 Hz), 7.60 (t, 1H, J = 7.4 Hz), 7.48 (t, 2H, J = 7.8 Hz), 7.39 (t, 1H, J = 7.8 Hz), 7.13 (d, 1H, J = 8.2 Hz), 6.99 (t, 1H, J = 7.8 Hz), 6.93 (dd, 1H, J = 7.8, 1.2 Hz).

¹³C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 193.7, 165.5, 155.4, 149.0, 148.7, 139.4, 134.4, 134.4, 132.4, 129.1, 129.1, 124.6, 124.2, 123.2, 120.7, 111.7.

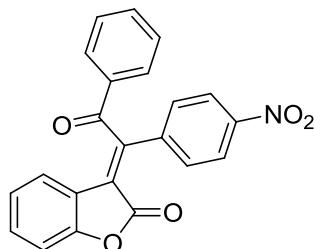
MS (70 eV, EI) m/z (%): 371 [M]⁺ (10), 266 (5), 163 (14), 105 (100), 77 (95).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3108 (w), 3068 (w), 1776 (s), 1670 (s), 1609 (m), 1513 (s), 1459 (s), 1345 (s).

HRMS (EI) for C₂₂H₁₃NO₅ [M]⁺ (371.0794) found 371.0787.

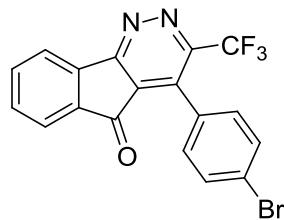
CCDC: 937197.

Synthesis of (*E*)-3-(1-(4-nitrophenyl)-2-oxo-2-phenylethylidene)benzofuran-2(3*H*)-one ((*E*)-5):



¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.26 (d, 2H, *J* = 8.7 Hz), 8.07 (d, 2H, *J* = 7.6 Hz), 7.74 (d, 2H, *J* = 8.7 Hz), 7.68 (t, 1H, *J* = 7.3 Hz), 7.54 (t, 2H, *J* = 7.7 Hz), 7.34 (t, 1H, *J* = 7.7 Hz), 7.12 (d, 1H, *J* = 8.1 Hz), 7.01-6.91 (m, 2H).

Synthesis of 4-(4-bromophenyl)-3-(trifluoromethyl)-5*H*-indeno[1,2-*c*]pyridazin-5-one (16a):



Prepared according to **TP4** from a solution of 2-(1-(4-bromophenyl)-3,3,3-trifluoro-2-oxopropylidene)-1*H*-indene-1,3(2*H*)-dione **1am** (122.7 mg, 0.3 mmol), hydraine monohydrate (22.0 μL, 1.5 equiv) in methanol (1.5 mL). [reaction condition: 27-30 °C for 10 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 1/2) furnished **16a** as yellow solid (94.5mg, 78%).

mp: 171.5-172.4 °C; R_f 0.68 (hexanes/dichloromethane: 1/3).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.27 (d, 1H, *J* = 7.5 Hz), 7.83-7.75 (m, 2H), 7.69-7.68 (m, 3H), 7.22 (d, 2H, *J* = 8.1 Hz).

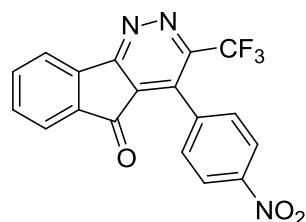
¹³C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 187.8, 164.1, 150.3 (q, *J* = 32.4 Hz), 140.2, 136.6, 135.7, 135.0, 133.3, 131.5, 129.8, 127.7, 125.3, 124.5, 123.1, 121.4 (q, *J* = 276.7 Hz).

MS (70 eV, EI) m/z (%): 406 [M+2]⁺ (94), 404 [M]⁺ (100), 334 (9), 325 (52), 200 (43), 69 (46).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3089 (w), 3052 (w), 1735 (s), 1609 (m), 1491 (m), 1177 (s), 1132 (s), 621 (w).

HRMS (EI) for C₁₈H₈BrF₃N₂O [M]⁺ (403.9772) found 403.9765.

Synthesis of 4-(4-nitrophenyl)-3-(trifluoromethyl)-5*H*-indeno[1,2-*c*]pyridazin-5-one (16b**):**



Prepared according to **TP4** from a solution of 2-(3,3,3-trifluoro-1-(4-nitrophenyl)-2-oxopropylidene)-1*H*-indene-1,3(2*H*)-dione **1dm** (112.6 mg, 0.3 mmol), hydrazine hydrate (22.0 μ L, 1.5 equiv) in methanol (1.5 mL). [reaction condition: 27-30 °C for 10 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **16b** as yellow solid (84.4 mg, 76%).

mp: 184.2-185.2 °C; R_f 0.51 (hexanes/dichloromethane: 1/3).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 °C) δ /ppm: 8.37 (d, 2H, J = 8.6 Hz), 8.25 (d, 1H, J = 7.5 Hz), 7.82 (t, 1H, J = 7.6 Hz), 7.78 (d, 1H, J = 7.4 Hz), 7.66 (t, 1H, J = 7.5 Hz), 7.56 (d, 2H, J = 8.5 Hz).

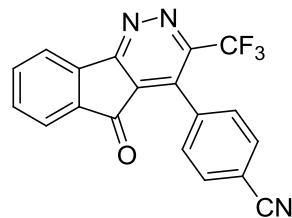
$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 °C) δ /ppm: 187.5, 164, 1, 149.6 (q, J = 32.8 Hz), 148.7, 140.1, 136.8, 135.4, 134.8, 134.1, 133.6, 129.5, 127.8, 123.4, 123.2, 123.1, 121.3 (q, J = 277.8 Hz).

MS (70 eV, EI) m/z (%): 371 [$\text{M}]^+$ (100), 325 (70), 302 (41), 274 (69), 229 (45), 200 (55).

IR (KBr) $\tilde{\nu}$ (cm^{-1}): 3076 (m), 2859 (w), 1726 (s), 1607 (m), 1521 (s), 1472 (w), 1348 (s), 1176 (s), 1150 (s).

HRMS (EI) for $\text{C}_{18}\text{H}_8\text{F}_3\text{N}_3\text{O}_3$ [$\text{M}]^+$ (371.0518) found 371.0523.

Synthesis of 4-(4-cyanophenyl)-3-(trifluoromethyl)-5*H*-indeno[1,2-*c*]pyridazin-5-one (16c**):**



Prepared according to **TP4** from a solution of 2-(3,3,3-trifluoro-1-(4-cyanophenyl)-2-oxopropylidene)-1*H*-indene-1,3(2*H*)-dione **1gm** (106.6 mg, 0.3 mmol), hydrazine monohydrate (22.0 μ L, 1.5 equiv) in methanol (1.5 mL). [reaction condition: 27-30 °C for 10 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **16c** as yellow solid (78.5 mg, 75%).

mp: 219.3-220.0 °C; R_f 0.38 (hexanes/dichloromethane: 1/3).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 °C) δ /ppm: 8.28 (d, 1H, J = 7.5 Hz), 7.87-7.75 (m, 4H), 7.65 (d, 1H, J = 7.5 Hz), 7.47 (d, 2H, J = 8.1 Hz).

$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 °C) δ /ppm: 187.6, 164.2, 149.8 (q, J = 32.8 Hz), 140.2, 136.9, 134.9, 134.4, 133.6, 132.0, 129.1, 127.7, 125.5, 123.3, 121.3 (q, J = 276.6 Hz), 114.0.

MS (70 eV, EI) m/z (%): 351 [$\text{M}]^+$ (100), 282 (29), 254 (39), 226 (23), 199 (13), 75 (11).

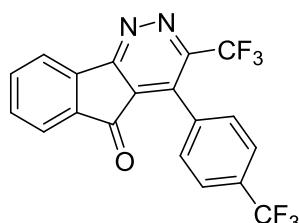
IR (KBr) $\tilde{\nu}$ (cm^{-1}): 3096 (m), 2924 (m), 2229 (s), 1731 (s), 1606 (m), 1583 (s), 1472 (s), 1258 (s).

HRMS (EI) for $\mathbf{C}_{19}\mathbf{H}_{8}\mathbf{F}_3\mathbf{N}_3\mathbf{O} [\mathbf{M}]^+$ (351.0619) found 351.0616.

Synthesis

of

3-(trifluoromethyl)-4-(4-(trifluoromethyl)phenyl)-5*H*-indeno[1,2-*c*]pyridazin-5-one (**16d**):



Prepared according to **TP4** from a solution of 2-(3,3,3-trifluoro-2-oxo-1-(4-(trifluoromethyl)phenyl)propylidene)-1*H*-indene-1,3(2*H*)-dione **1fm** (119.5 mg, 0.3 mmol), hydraine hydrate (22.0 μL , 1.5 equiv) in methanol (1.5 mL). [reaction condition: 27-30 $^\circ\text{C}$ for 10 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **16d** as yellow solid (96.0 mg, 81%).

mp: 158.4-159.2 $^\circ\text{C}$; R_f 0.68 (hexanes/dichloromethane: 1/3).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 $^\circ\text{C}$) δ/ppm : 8.26-8.19 (m, 1H), 7.84-7.76 (m, 3H), 7.74 (d, 1H, J = 7.4 Hz), 7.62 (t, 1H, J = 7.5 Hz), 7.49 (d, 2H, J = 8.1 Hz).

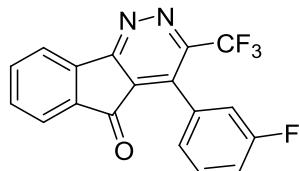
$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 $^\circ\text{C}$) δ/ppm : 187.5, 164.1, 150.0 (q, J = 32.7 Hz), 140.1, 140.1, 136.7, 135.1, 134.9, 133.4, 132.6, 131.8 (q, J = 32.8 Hz), 128.8, 125.3, 125.2, 125.1 (q, J = 3.3 Hz), 123.7 (q, J = 276.4 Hz), 123.3, 123.1, 121.3 (q, J = 276.6 Hz).

MS (70 eV, EI) m/z (%): 394 [$\mathbf{M}]^+$ (87), 325 (100), 298 (47), 269 (37), 250 (22), 200 (23), 69 (21).

IR (KBr) $\tilde{\nu}$ (cm^{-1}): 3098 (m), 3071 (m), 1738 (s), 1610 (s), 1602 (s), 1592 (s), 1475 (w), 1380 (s), 1322 (s), 1265 (s).

HRMS (EI) for $\mathbf{C}_{19}\mathbf{H}_{8}\mathbf{F}_6\mathbf{N}_2\mathbf{O} [\mathbf{M}]^+$ (394.0541) found 394.0549.

Synthesis of 4-(3-fluorophenyl)-3-(trifluoromethyl)-5*H*-indeno[1,2-*c*]pyridazin-5-one (**16e**):



Prepared according to **TP4** from a solution of 2-(3,3,3-trifluoro-1-(3-fluorophenyl)-2-oxopropylidene)-1*H*-indene-1,3(2*H*)-dione **1hm** (104.5 mg, 0.3 mmol), hydraine hydrate (22.0 μL , 1.5 equiv) in methanol (1.5 mL). [reaction condition: 27-30 $^\circ\text{C}$ for 10 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 1/1) furnished **16e** as yellow solid (69.2 mg, 67%).

mp: 133.0-133.8 $^\circ\text{C}$; R_f 0.65 (hexanes/dichloromethane: 1/3).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 $^\circ\text{C}$) δ/ppm : 8.26 (d, 1H, J = 7.5 Hz), 7.84-7.74 (m, 2H) 7.63 (t, 1H, J = 7.5 Hz), 7.49 (td, 1H, J = 7.9, 5.8 Hz), 7.26 (t, 1H, J = 8.5 Hz), 7.11 (d, 1H, J = 7.7 Hz), 7.06 (d,

1H, $J = 8.9$ Hz)

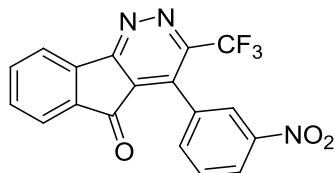
$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 °C) δ/ppm: 187.5, 164.1, 162.1 (q, $J = 247.9$ Hz), 150.2 (q, $J = 32.5$ Hz), 140.1, 136.6, 135.2, 135.1, 134.9, 133.3, 130.7 (q, $J = 8.2$ Hz), 130.0 (q, $J = 8.4$ Hz), 127.8, 125.2, 124.1, 124.1, 123.0, 121.4 (q, $J = 276.8$ Hz), 116.9 (q, $J = 21.0$ Hz), 115.6 (q, $J = 23.5$ Hz).

MS (70 eV, EI) m/z (%): 344 [M]⁺ (100), 325 (35), 275 (40), 251 (67), 248 (82), 218 (92), 109 (36), 75 (51), 69 (66).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3088 (s), 3061 (w), 1734 (s), 1609 (m), 1587 (m), 1488 (m), 1378 (m), 1264 (s), 1186 (s).

HRMS (EI) for $\text{C}_{18}\text{H}_8\text{F}_4\text{N}_2\text{O} [\text{M}]^+$ (344.0573) found 344.0576.

Synthesis of 4-(3-nitrophenyl)-3-(trifluoromethyl)-5*H*-indeno[1,2-*c*]pyridazin-5-one (**16f**):



Prepared according to **TP4** from a solution of 2-(3,3,3-trifluoro-1-(3-nitrophenyl)-2-oxopropylidene)-1*H*-indene-1,3(2*H*)-dione **1cm** (112.6 mg, 0.3 mmol), hydraine hydrate (22.0 μL, 1.5 equiv) in methanol (1.5 mL). [reaction condition: 27-30 °C for 10 minutes]. Purification by flash chromatography (hexanes/dichloromethane: 1/3) furnished **16f** as yellow solid (88.0 mg, 79%).

mp: 188.0-188.9 °C; R_f 0.55 (hexanes/dichloromethane: 1/3).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 °C) δ/ppm: 8.43 (d, 1H, $J = 8.0$ Hz), 8.34-8.20 (m, 2H), 7.79-7.59 (m, 5H).

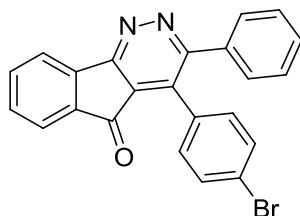
$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 °C) δ/ppm: 187.5, 164, 2, 149.9 (q, $J = 32.7$ Hz), 147.8, 140.1, 136.9, 134.9, 134.2, 133.8, 133.6, 130.4, 129.5, 128.0, 124.7, 123.6, 123.3, 121.3 (q, $J = 277.6$ Hz).

MS (70 eV, EI) m/z (%): 371 [M]⁺ (100), 325 (88), 69 (83).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3092 (w), 3069 (m), 2924 (w), 1733 (s), 1604 (m), 1589 (s), 1478 (w), 1352 (s), 1257 (s).

HRMS (EI) for $\text{C}_{18}\text{H}_8\text{F}_3\text{N}_3\text{O}_3 [\text{M}]^+$ (371.0518) found 371.0518.

Synthesis of 4-(4-bromophenyl)-3-phenyl-5*H*-indeno[1,2-*c*]pyridazin-5-one (17**):**



Prepared according to **TP4** from a solution of 2-(1-(4-bromophenyl)-2-oxo-2-phenylethylidene)-1*H*-indene-1,3(2*H*)-dione **1aa** (118.9 mg, 0.3 mmol), hydrazine hydrate (22.0 μ L, 1.5 equiv) in methanol (1.5 mL). [reaction condition: 27-30 °C for 1.5 hours]. Purification by flash chromatography furnished **17** as yellow solid (117.3 mg, 95%). mp: 194.8-195.4 °C; R_f 0.19 (hexanes/dichloromethane: 1/3).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ /ppm: 8.24 (d, 1H, *J* = 7.4 Hz), 7.8-7.71 (m, 2H), 7.56 (t, 1H, *J* = 7.5 Hz), 7.48 (d, 2H, *J* = 8.2 Hz), 7.39-7.27 (m, 5H), 7.11 (d, 2H, *J* = 8.3 Hz).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ /ppm: 189.5, 161.5, 161.0, 141.3, 136.3, 135.8, 135.4, 134.8, 132.2, 131.5, 131.3, 130.0, 129.9, 129.2, 128.2, 126.5, 124.9, 124.0, 122.4.

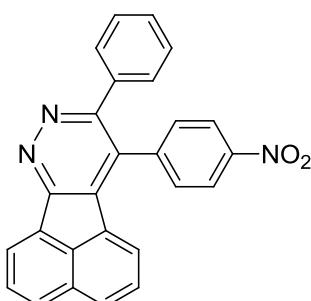
MS (70 eV, EI) m/z (%): 414 [M+2]⁺ (32), 412 [M]⁺ (35), 333 (100), 76 (21).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3061 (w), 2924 (w), 1726 (s), 1604 (m), 1589 (m), 1486 (s), 610 (m).

HRMS (ESI) for C₂₃H₁₄BrN₂O [M+H]⁺ (413.0290) found 413.0283.

CCDC: 935874

Synthesis of 10-(4-nitrophenyl)-9-phenylacenaphtho[1,2-*c*]pyridazine (18**):**



Prepared according to **TP4** from a solution of (*E*)-2-(1-(4-nitrophenyl)-2-oxo-2-phenylethylidene)acenaphthylen-1(2*H*)-one **3a** (121.6 mg, 0.3 mmol), hydrazine hydrate (22.0 μ L, 1.5 equiv) in methanol (1.5 mL). [reaction condition: 27-30 °C for 2 hours]. Purification by flash chromatography (dichloromethane/ethyl acetate: 5/1) furnished **18** as yellow solid (98.7 mg, 82%).

mp: 247.8-248.5; R_f 0.63 (hexanes/dichloromethane: 1/3).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ /ppm: 8.48 (d, 1H, *J* = 6.6 Hz), 8.32 (d, 2H, *J* = 8.5 Hz), 7.99 (d, 2H, *J* = 8.0 Hz), 7.76 (t, 1H, *J* = 7.5 Hz), 7.59 (d, 2H, *J* = 8.6 Hz), 7.48 (t, 1H, *J* = 7.7 Hz), 7.37 (d, 2H, *J* = 6.8 Hz), 7.34-7.21 (m, 3H), 7.16 (d, 1H, *J* = 7.0 Hz).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ /ppm: 158.5, 157.2, 147.9, 142.1, 136.4, 133.6, 133.3, 131.8, 131.3, 130.7, 130.5, 130.2, 130.1, 129.9, 129.5, 128.8, 128.7, 128.1, 125.6, 124.1, 123.3.

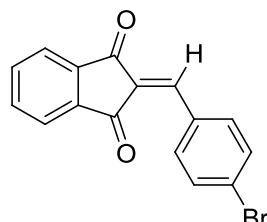
MS (70 eV, EI) m/z (%): 401 [M]⁺ (89), 400 [M-1]⁺ (100), 354 (92), 324 (42).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3103 (w), 3050 (w), 2840 (w), 1597 (m), 1510 (s), 1460 (w), 1341 (s).

HRMS (EI) for C₂₆H₁₅N₃O₂ [M]⁺ (401.1164) found 401.1165.

CCDC: 935875

Synthesis of 2-(4-bromobenzylidene)-1*H*-indene-1,3(2*H*)-dione (6a):



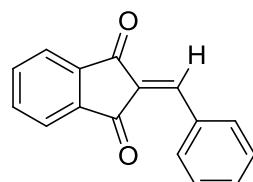
Prepared according to **TP 5**.

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.33 (d, 2H, *J* = 8.5 Hz), 8.06-7.97 (m, 2H), 7.87-7.77 (m, 3H), 7.64 (d, 2H, *J* = 8.5 Hz).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 189.7, 188.8, 145.1, 142.4, 140.0, 135.4, 135.3, 135.2, 132.0, 131.8, 129.5, 128.3, 123.3, 123.2.

MS (70 eV, EI) m/z (%): 314 [M+2]⁺ (41), 312 [M]⁺ (46), 233 (87), 104 (64), 76 (100).

Synthesis of 2-benzylidene-1*H*-indene-1,3(2*H*)-dione (6b):



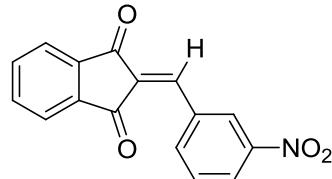
Prepared according to **TP 5** from 1,3-indanedione (1506.5 mg, 10 mmol), benzaldehyde (1.1181 mL, 1.1 equiv) and *L*-proline (349.0 mg, 0.3 equiv) in methanol (20 mL) furnished **6b** as dark green solid (2131.5 mg, 91%).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.47 (d, 2H, *J* = 8.0 Hz), 8.06-7.99 (m, 2H), 7.92 (s, 1H), 7.86-7.79 (m, 2H), 7.60 (m, 3H).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 190.3, 189.0, 147.0, 142.6, 140.1, 135.4, 135.2, 134.1, 133.2, 133.1, 129.2, 128.8, 123.4, 123.3.

MS (70 eV, EI) m/z (%): 234 [M]⁺ (62), 314 [M-1]⁺ (100), 76 (30).

Synthesis of 2-(3-nitrobenzylidene)-1*H*-indene-1,3(2*H*)-dione (6c):



Prepared according to **TP 5** from 1,3-indanedione (1506.5 mg, 10 mmol), 3-nitrobenzaldehyde (1511.2 mg, 1.1 equiv) and *L*-proline (349.0 mg, 0.3 equiv) in methanol (20 mL) furnished **6c** as dark green solid (2289.8 mg, 82%).

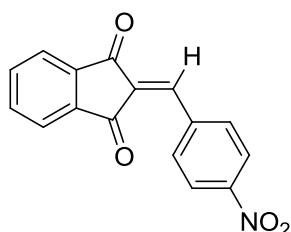
¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 9.40 (s, 1H), 8.68 (d, 1H, *J* = 7.7 Hz), 8.39 (d, 1H, *J* =

8.4 Hz), 8.12-8.03 (m, 2H), 7.91 (s, 1H), 7.88 (dd, 2H, $J = 5.6, 3.0$ Hz), 7.71 (t, 1H, $J = 8.0$ Hz).

$^{13}\text{C-NMR}$ (125 MHz, CDCl_3 , 25 °C) δ/ppm : 189.2, 188.6, 148.5, 143.0, 142.6, 140.3, 138.9, 135.9, 135.8, 134.3, 131.6, 129.7, 128.0, 126.7, 123.8, 123.7.

MS (70 eV, EI) m/z (%): 279 [M] $^+$ (54), 262 (73), 232 (76), 176 (83), 76 (100).

Synthesis of 2-(4-nitrobenzylidene)-1*H*-indene-1,3(2*H*)-dione (6d**):**



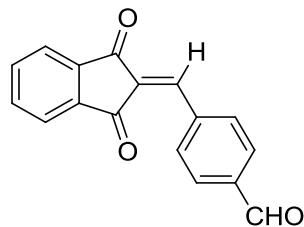
Prepared according to **TP 5** from 1,3-indanedione (1506.5 mg, 10 mmol), 4-nitrobenzaldehyde (1679.1 mg, 1.1 equiv) and *L*-proline (349.0 mg, 0.3 equiv) in methanol (20 mL) furnished **6d** as yellow solid (2454.0 mg, 94%).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 °C) δ/ppm : 8.55 (d, 2H, $J = 8.8$ Hz), 8.34 (d, 2H, $J = 8.8$ Hz), 8.07 (dd, 2H, $J = 5.6, 3.0$ Hz), 7.92-7.85 (m, 3H).

$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 °C) δ/ppm : 189.1, 188.4, 149.5, 142.6, 140.3, 138.4, 136.0, 135.9, 134.2, 132.3, 123.8, 123.7, 123.6.

MS (70 eV, EI) m/z (%): 279 [M] $^+$ (55), 262 (100), 232 (93), 176 (80), 104 (46), 76 (96).

Synthesis of 4-((1,3-dioxo-1*H*-inden-2(3*H*)-ylidene)methyl)benzaldehyde (6e**):**



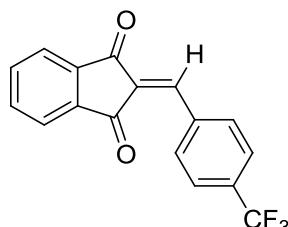
Prepared according to **TP 5** from 1,3-indanedione (1506.5 mg, 10 mmol), terephthalaldehyde (1475.5 mg, 1.1 equiv) and *L*-proline (349.0 mg, 0.3 equiv) in methanol (20 mL) furnished **6e** as yellow solid (2454.0 mg, 94%).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 °C) δ/ppm : 10.11 (s, 1H), 8.55 (d, 2H, $J = 8.0$ Hz), 8.10-8.03 (m, 2H), 8.01 (d, 2H, $J = 8.1$ Hz), 7.92 (s, 1H), 7.90-7.83 (m, 2H).

$^{13}\text{C-NMR}$ (125 MHz, CDCl_3 , 25 °C) δ/ppm : 191.5, 189.5, 188.6, 144.3, 142.6, 140.2, 138.4, 138.1, 135.8, 135.7, 134.1, 131.5, 129.7, 123.7, 123.6.

MS (70 eV, EI) m/z (%): 262 [M] $^+$ (100), 233 (69), 104 (23), 76 (39).

Synthesis of 2-(4-(trifluoromethyl)benzylidene)-1*H*-indene-1,3(2*H*)-dione (6f**):**



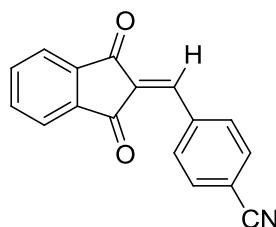
Prepared according to **TP 5** from 1,3-indanedione (753.3 mg, 5 mmol), aldehyde (819.4 mL, 1.1 equiv) and *L*-proline (174.5 mg, 0.3 equiv) in methanol (10 mL) furnished **6f** as light yellow solid (2454.0 mg, 94%).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.52 (d, 2H, *J* = 8.2 Hz), 8.08-8.02 (m, 2H), 7.90 (s, 1H), 7.89-7.83 (m, 2H), 7.76 (d, 2H, *J* = 8.3 Hz).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 189.5, 188.6, 144.2, 142.6, 140.2, 135.9, 135.7, 135.6, 133.8, 133.6 (q, *J* = 32.6 Hz), 131.1, 125.5 (q, *J* = 3.8 Hz), 123.6 (q, *J* = 272.7 Hz), 123.6, 123.6.

MS (70 eV, EI) m/z (%): 302 [M]⁺ (61), 301 [M-1]⁺ (79), 233 (100), 104 (50), 76 (77).

Synthesis of 2-(4-(cyano)benzylidene)-1*H*-indene-1,3(2*H*)-dione (6g**):**



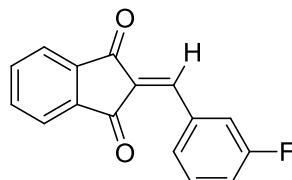
Prepared according to **TP 5** from 1,3-indanedione (1506.5 mg, 10 mmol), 4-formylbenzonitrile (1471.9 mg, 1.1 equiv) and *L*-proline (349.0 mg, 0.3 equiv) in methanol (20 mL) furnished **6g** as yellow solid (2444.0 mg, 94%).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.50 (d, 2H, *J* = 8.3 Hz), 8.09-8.03 (m, 2H), 7.91-7.84 (m, 3H), 7.79 (d, 2H, *J* = 8.3 Hz).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 189.2, 188.4, 143.3, 142.6, 140.2, 136.7, 135.9, 135.8, 133.7, 132.2, 131.8, 123.7, 123.7, 118.2, 115.4.

MS (70 eV, EI) m/z (%): 259 [M]⁺ (63), 258 [M-1]⁺ (100), 104 (44), 76 (84).

Synthesis of 2-(3-fluorobenzylidene)-1*H*-indene-1,3(2*H*)-dione (6h**):**



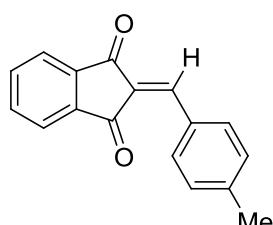
Prepared according to **TP 5** from 1,3-indanedione (1506.5 mg, 10 mmol), 3-fluorobenzaldehyde (1060.8 uL, 1.1 equiv) and *L*-proline (349.0 mg, 0.3 equiv) in methanol (20 mL) furnished **6h** as green solid (1841.4 mg, 73%).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.43 (dd, 1H, *J* = 10.2, 1.3 Hz), 8.09-7.99 (m, 3H), 7.89-7.80 (m, 3H), 7.48 (dt, 1H, *J* = 8.0, 7.0 Hz), 7.26 (td, 1H, *J* = 8.4, 2.8).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 189.7, 188.7, 162.5 (d, *J* = 246.7 Hz), 145.1, 145.0, 142.5, 140.1, 135.6, 135.4, 134.9 (d, *J* = 8.5 Hz), 130.2 (d, *J* = 3.4 Hz), 130.1 (d, *J* = 4.1 Hz), 123.5 (d, *J* = 6.2 Hz), 120.1, 119.9, 119.7.

MS (70 eV, EI) m/z (%): 252 [M]⁺ (65), 251 [M-1]⁺ (100), 233 (7), 104 (31), 76 (47).

Synthesis of 2-(4-methylbenzylidene)-1*H*-indene-1,3(2*H*)-dione (6i**):**



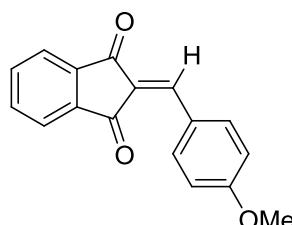
Prepared according to **TP 5** from 1,3-indanedione (753.3 mg, 5 mmol), 4-methylbenzaldehyde (729.3 mL, 1.1 equiv) and *L*-proline (174.5 mg, 0.3 equiv) in methanol (10 mL) furnished **6i** as yellow green solid (1039.0 mg, 84%).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.40 (d, 2H, *J* = 8.2 Hz), 8.04-7.98 (m, 2H), 7.89 (s, 1H), 7.84-7.78 (m, 2H), 7.33 (d, 2H, *J* = 8.1 Hz), 2.46 (s, 3H).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 190.1, 188.8, 146.6, 144.3, 142.2, 139.7, 134.9, 134.7, 134.3, 130.4, 129.4, 127.9, 122.9, 122.9, 21.8.

MS (70 eV, EI) m/z (%): 248 [M]⁺ (76), 247 [M-1]⁺ (54), 233, (100), 76 (19).

Synthesis of 2-(4-methoxybenzylidene)-1*H*-indene-1,3(2*H*)-dione (6j**):**



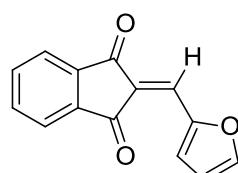
Prepared according to **TP 5** from 1,3-indanedione (753.35 mg, 5 mmol), 4-methoxybenzaldehyde (6.828 mL, 1.1 equiv) and *L*-proline (124.5 mg, 0.3 equiv) in methanol (20 mL) furnished **6j** as yellow solid (1107.0 mg, 84%).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.51 (d, 2H, *J* = 8.5 Hz), 8.05-7.88 (m, 2H), 7.79 (s, 1H), 7.78-7.68 (m, 2H), 6.98 (d, 2H, *J* = 8.5 Hz), 3.88 (s, 3H).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 190.6, 189.3, 163.9, 146.6, 142.3, 139.8, 137.1, 134.9, 134.7, 126.5, 126.4, 122.9, 122.8, 114.3, 55.5.

MS (70 eV, EI) m/z (%): 264 [M]⁺ (100), 263 [M-1]⁺ (78), 233 (27), 132 (20), 104 (34), 76 (52).

Synthesis of 2-(furan-2-ylmethylene)-1*H*-indene-1,3(2*H*)-dione (6k**):**



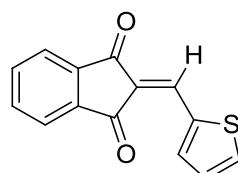
Prepared according to **TP 5** from 1,3-indanedione (1506.5 mg, 10 mmol), 2-furaldehyde (828.3 uL, 1.1 equiv) and *L*-proline (349.0 mg, 0.3 equiv) in methanol (20 mL) furnished **6k** as black green solid (1681.0 mg, 75%).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.58 (d, 1H, *J* = 3.7 Hz), 8.01-7.94 (m, 2H), 7.82-7.77 (m, 3H), 7.76 (s, 1H), 6.73 (d, 1H, *J* = 3.8 Hz).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 190.0, 188.9, 151.4, 149.0, 142.3, 140.4, 135.1, 134.8, 129.2, 124.8, 124.7, 123.1, 122.9, 114.6.

MS (70 eV, EI) m/z (%): 224 [M]⁺ (100), 168 (62), 119 (6), 104 (16), 76 (41).

Synthesis of 2-(thiophen-2-ylmethylene)-1*H*-indene-1,3(2*H*)-dione (6l**):**



Prepared according to **TP 5** from 1,3-indanedione (753.3 mg, 5 mmol), 2-thiophenecarbaldehyde (467.3 uL, 1.1 equiv) and *L*-proline (174.5 mg, 0.3 equiv) in methanol (10 mL) furnished **6l** as yellow solid (2454.0 mg, 94%).

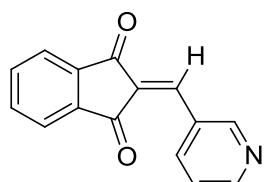
¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.07 (d, 1H, *J* = 3.4 Hz), 8.02 (s, 1H), 8.01-7.95 (m, 2H), 7.87 (d, 1H, *J* = 5.2 Hz), 7.92-7.76 (m, 2H), 7.25 (t, 1H, *J* = 4.4 Hz).

¹³C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 190.3, 189.4, 142.1, 141.6, 140.4, 138.2, 137.4, 136.2,

135.1, 134.9, 128.6, 124.8, 123.1, 123.0.

MS (70 eV, EI) m/z (%): 240 [M]⁺ (100), 239 [M-1]⁺ (83), 184 (48), 104 (27), 82 (8), 76 (55).

Synthesis of 2-(pyridin-3-ylmethylene)-1*H*-indene-1,3(2*H*)-dione (6m):



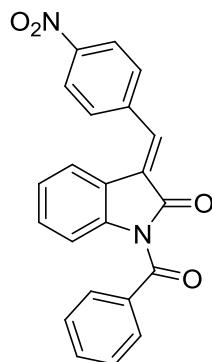
Prepared according to **TP 5** from 1,3-indanedione (1506.5 mg, 10 mmol), 3-pyridinecarboxaldehyde (1.1265 mL, 1.1 equiv) and *L*-proline (349.0 mg, 0.3equiv) in methanol (20 mL) furnished **6m** as yellow solid (1033.0 mg, 44%).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 9.18 (s, 1H), 9.15 (d, 1H, *J* = 8.2 Hz) 8.72 (d, 1H, *J* = 3.7 Hz), 8.01 (dd, 2H, *J* = 5.5, 3.1 Hz), 7.87-7.81 (m, 3H), 7.47 (dd, 1H, *J* = 7.8, 4.9 Hz).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 189.1, 188.6, 154.6, 152.7, 142.3, 142.2, 140.0, 139.6, 135.6, 135.5, 131.2, 128.9, 123.5, 123.4.

MS (70 eV, EI) m/z (%): 235 [M]⁺ (62), 234 [M-1]⁺ (100), 104 (26), 76 (58).

Synthesis of (Z)-1-benzoyl-3-(4-nitrobenzylidene)indolin-2-one (7a):



The synthesis is related to ref 1.

mp: 219.5-220.4 °C; R_f 0.24 (hexanes/dichloromethane: 1/1).

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.35 (d, 2H, *J* = 8.6 Hz), 7.91 (d, 1H, *J* = 8.2 Hz), 7.78 (m, 5H), 7.62 (t, 1H, *J* = 7.4 Hz), 7.51 (m, 3H), 7.41 (t, 1H, *J* = 7.7 Hz), 7.08 (t, 1H, *J* = 7.7 Hz).

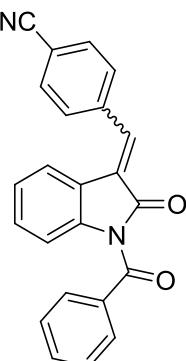
¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 169.1, 166.9, 148.2, 141.2, 141.1, 134.9, 134.2, 133.1, 131.4, 129.9, 129.5, 128.7, 128.3, 124.6, 124.1, 122.7, 121.2, 115.6.

MS (70 eV, EI) m/z (%): 370 (33), 190 (13), 105 (100), 77 (82).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 1734 (s), 1674 (m), 1601 (w), 1518 (m), 1462 (w), 1348 (s).

HRMS (FAB) for C₂₂H₁₅N₂O₄ [M+H]⁺ (371.1032) found: 371.1032.

Synthesis of 4-((1-benzoyl-2-oxoindolin-3-ylidene)methyl)benzonitrile (7b):



The synthesis is related to ref 1.

mp: 191.8-193.2; R_f 0.20 (hexanes/dichloromethane: 1/1).

$^1\text{H-NMR}$ (125 MHz, CDCl_3 , 25 °C) δ/ppm : 8.15 (d, 2H, $J = 8.3$ Hz), 7.91 (d, 2H, $J = 8.2$ Hz), 7.83 (d, 1H, $J = 8.1$ Hz), 7.81-7.75 (m, 10H), 7.75-7.72 (m, 6H), 7.76-7.57 (m, 7H), 7.56-7.46 (m, 8H), 7.45-7.39 (m, 3H), 7.29-7.26 (m, 1H), 7.08 (t, 2H, $J = 7.7$ Hz). [For E, Z-form mixture with 1:2 ratio.]

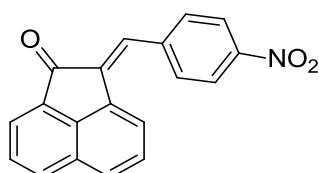
$^{13}\text{C-NMR}$ (500 MHz, CDCl_3 , 25 °C) δ/ppm : 169.4, 169.2, 167.0, 165.0, 141.1, 139.6, 139.2, 127.3, 135.6, 135.4, 134.3, 134.2, 133.0, 132.6, 132.0, 132.0, 131.2, 130.5, 129.6, 129.5, 129.4, 128.4, 128.3, 128.2, 127.7, 124.7, 124.5, 124.3, 122.7, 121.3, 119.6, 118.5, 118.2, 115.6, 115.1, 113.4, 113.3. [For E, Z-form mixture]

MS (70 eV, EI) m/z (%): 350 (37), 190 (14), 105 (100), 77 (100).

IR (KBr) $\tilde{\nu}$ (cm^{-1}): 3090 (w), 2929 (w), 2222 (m), 1729 (s), 1686 (s), 1598 (m), 1458 (m).

HRMS (FAB) for $\text{C}_{23}\text{H}_{15}\text{N}_2\text{O}_2$ [M+H^+] (351.1134) found: 351.1136.

Synthesis of (E)-2-(4-nitrobenzylidene)acenaphthylen-1(2H)-one (8a):



The synthesis is related to ref 2.

mp: 233.8-236.2; R_f 0.31 (hexanes/dichloromethane: 1/1).

$^1\text{H-NMR}$ (400 MHz, CDCl_3 , 25 °C) δ/ppm : 8.40 (d, 2H, $J = 8.6$ Hz), 8.15 (m, 2H), 7.95 (d, 1H, $J=8.3$ Hz), 7.91 (m, 3H), 7.82 (t, 1H, $J=7.3$ Hz), 7.74 (d, 1H, $J=7.1$ Hz), 7.56 (t, 1H, $J=7.4$ Hz).

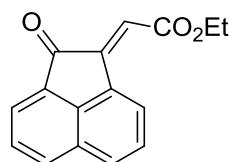
$^{13}\text{C-NMR}$ (100 MHz, CDCl_3 , 25 °C) δ/ppm : 129.0, 147.9, 142.3, 141.2, 136.4, 132.0, 131.7, 131.4, 131.1, 130.9, 130.2, 128.3, 128.0, 126.9, 124.0, 122.1, 120.4.

MS (70 eV, EI) m/z (%): 302 [M+1^+] (24), 301 [M^+] (98), 255 (54), 254 (98), 226 (100), 213 (12), 200 (10), 150 (22), 113 (28), 100(16).

IR (KBr) $\tilde{\nu}$ (cm^{-1}): 2829 (w), 1708 (s), 1687 (w), 1624 (m), 1517 (s), 1489 (w), 1344 (s).

HRMS (FAB) for $\text{C}_{19}\text{H}_{12}\text{NO}_3$, [M+H^+] (302.0817) found: 302.0821.

Synthesis of (*E*)-ethyl 2-(2-oxoacenaphthylen-1(2*H*)-ylidene)acetate (8b):



The synthesis is related to ref 2.

mp: 106.3-107.0 °C; R_f 0.30 (hexanes/dichloromethane: 1/1).

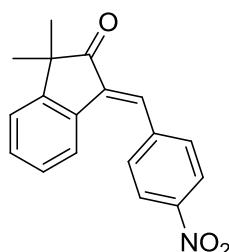
¹H-NMR (500 MHz, CDCl₃, 25 °C) δ/ppm: 8.60 (d, 1H, *J* = 7.3 Hz), 8.08 (d, 1H, *J* = 8.3 Hz), 7.97 (t, 3H, *J* = 8.6 Hz), 7.83 (d, 1H, *J* = 6.9 Hz), 7.69 (t, 1H, *J* = 7.9 Hz), 7.65 (t, 1H, *J* = 7.6 Hz), 7.51 (t, 1H, *J* = 7.3 Hz), 7.40 (t, 2H, *J* = 7.7 Hz), 4.29 (q, 2H, *J* = 7.1 Hz), 1.17 (t, 3H, *J* = 7.1 Hz).

¹³C-NMR (125 MHz, CDCl₃, 25 °C) δ/ppm: 193.9, 191.6, 164.2, 142.9, 140.5, 136.5, 135.2, 133.4, 132.0, 130.7, 130.6, 129.9, 128.7, 128.7, 128.6, 128.2, 128.0, 125.5, 122.2, 62.3, 13.9.

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 2985 (w), 1719 (s), 1684 (w), 1638 (m), 1601 (w), 1266 (s), 1189 (s).

MS (70 eV, EI) m/z (%): 252 (100), 207 (63), 180 (69), 151 (93), 75 (18), 74 (8).

Synthesis of (*E*)-1,1-dimethyl-3-(4-nitrobenzylidene)-1*H*-inden-2(3*H*)-one (9):



The synthesis is related to ref 3.

mp: 156.7-157.6 °C; R_f 0.48 (hexanes/dichloromethane: 1/1).

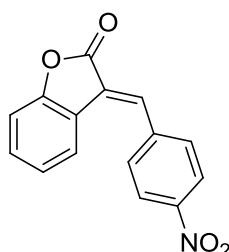
¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.31 (d, 2H, *J* = 8.7 Hz), 7.76 (d, 2H, *J* = 8.6 Hz), 7.52 (d, 1H, *J* = 7.9), 7.48 (s, 1H), 7.43-7.34 (m, 2H), 7.14 (td, 1H, *J* = 10.9, 1.8 Hz), 1.40 (s, 6H).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm: 209.3, 150.3, 147.7, 142.5, 135.4, 134.0, 130.8, 129.6, 129.5, 127.2, 124.0, 123.7, 123.2, 47.3, 25.5.

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3100 (w), 2952 (w), 1726 (s), 1618 (w), 1514 (s), 1344 (s).

HRMS (EI) for C₁₈H₁₅NO₃ [M]⁺ (293.1052) found 293.1059.

Synthesis of (*E*)-3-(4-nitrobenzylidene)benzofuran-2(3*H*)-one (10):



The synthesis is related to ref 4.

mp: 234.5-235.4 °C; R_f 0.50 (hexanes/dichloromethane: 1/1)

¹H-NMR (400 MHz, CDCl₃, 25 °C) δ/ppm: 8.31 (d, 2H, *J* = 8.9 Hz), 8.26 (d, 2H, *J* = 8.8 Hz), 7.63 (s, 1H), 7.59 (d, 1H, *J* = 7.6 Hz), 7.42 (t, 1H, *J* = 7.7 Hz), 7.23 (t, 1H, *J* = 7.6 Hz), 7.15 (d, 1H, *J* = 8.1 Hz).

¹³C-NMR (100 MHz, CDCl₃, 25 °C) δ/ppm:

MS (70 eV, EI) m/z (%): 267 [M]⁺ (100), 239 (5), 221 (6), 193 (15), 165 (86), 74 (15).

IR (KBr) $\tilde{\nu}$ (cm⁻¹): 3115 (w), 1753 (s), 1594 (m), 1513 (s), 1462 (m), 1344 (s).

HRMS (EI) for C₁₅H₉NO₄ [M]⁺ (267.0532) found 267.0530.

1. Mokrosz, M. J.; Charakchieva-Minol, S.; Kozioł, A.; Kłodzińska, A.; Chojnacka-Wójcik, E. *Bioorg. Med. Chem. Lett.* **2001**, *11*, 1229.
2. Tsuge, O.; Tashiro, M.; Shinkai, I. *Bull. Chem. Soc. Jpn.* **1969**, *42*, 181.
3. Martínez, A.; Fernández, M.; Estévez, J. C.; Estévez, R. J.; Castedo, L. *Tetrahedron* **2005**, *61*, 485.
4. Marelli, C.; Monti, C.; Galli, S.; Masciocchi, N.; Piarulli, U. *Tetrahedron* **2006**, *62*, 8943.

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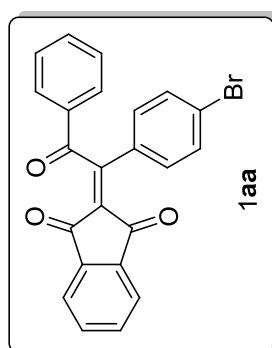
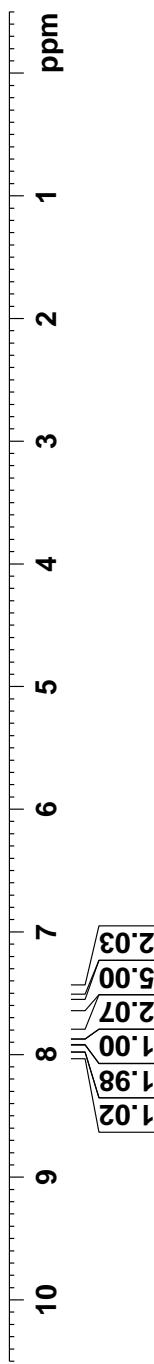
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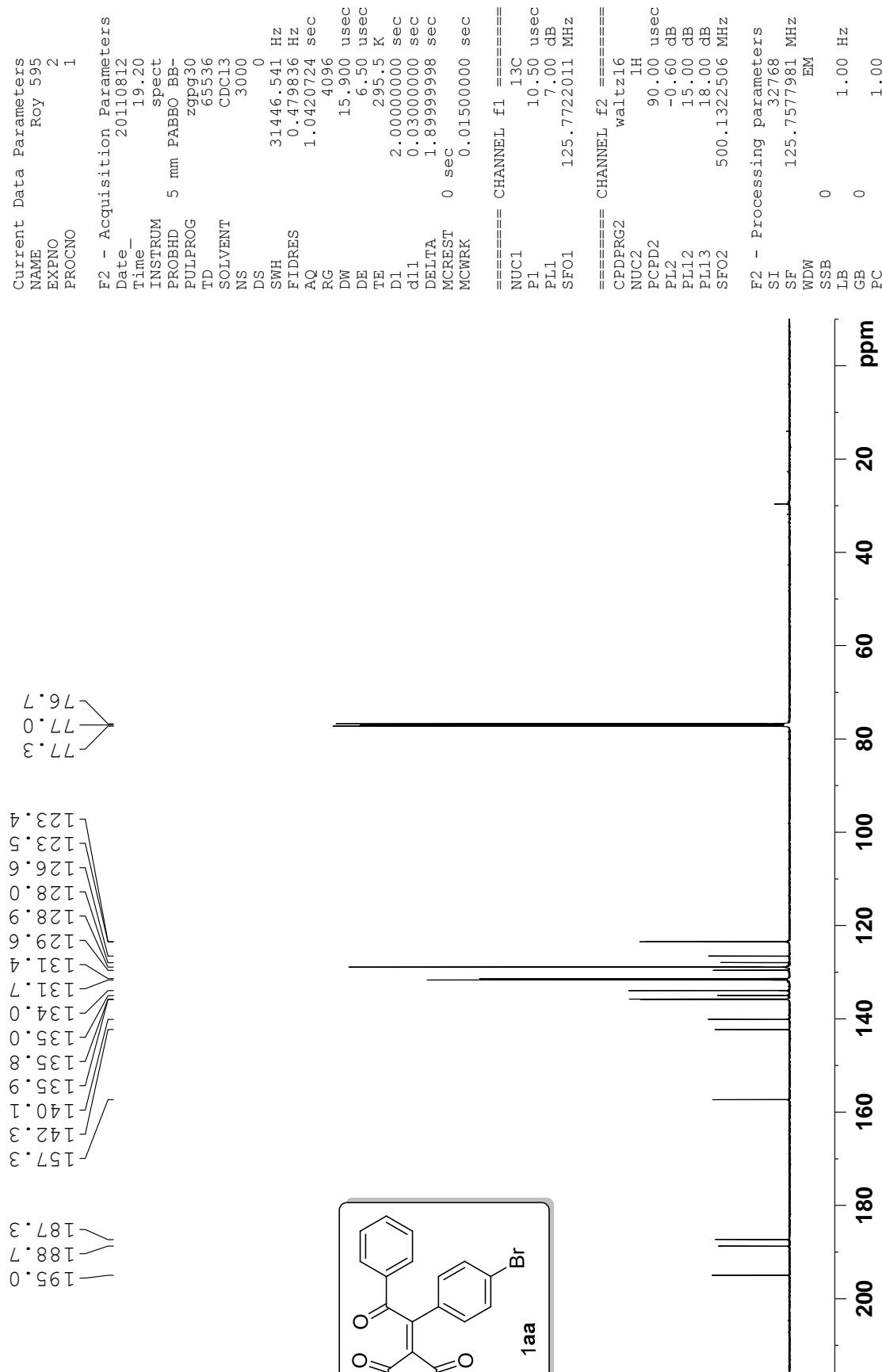
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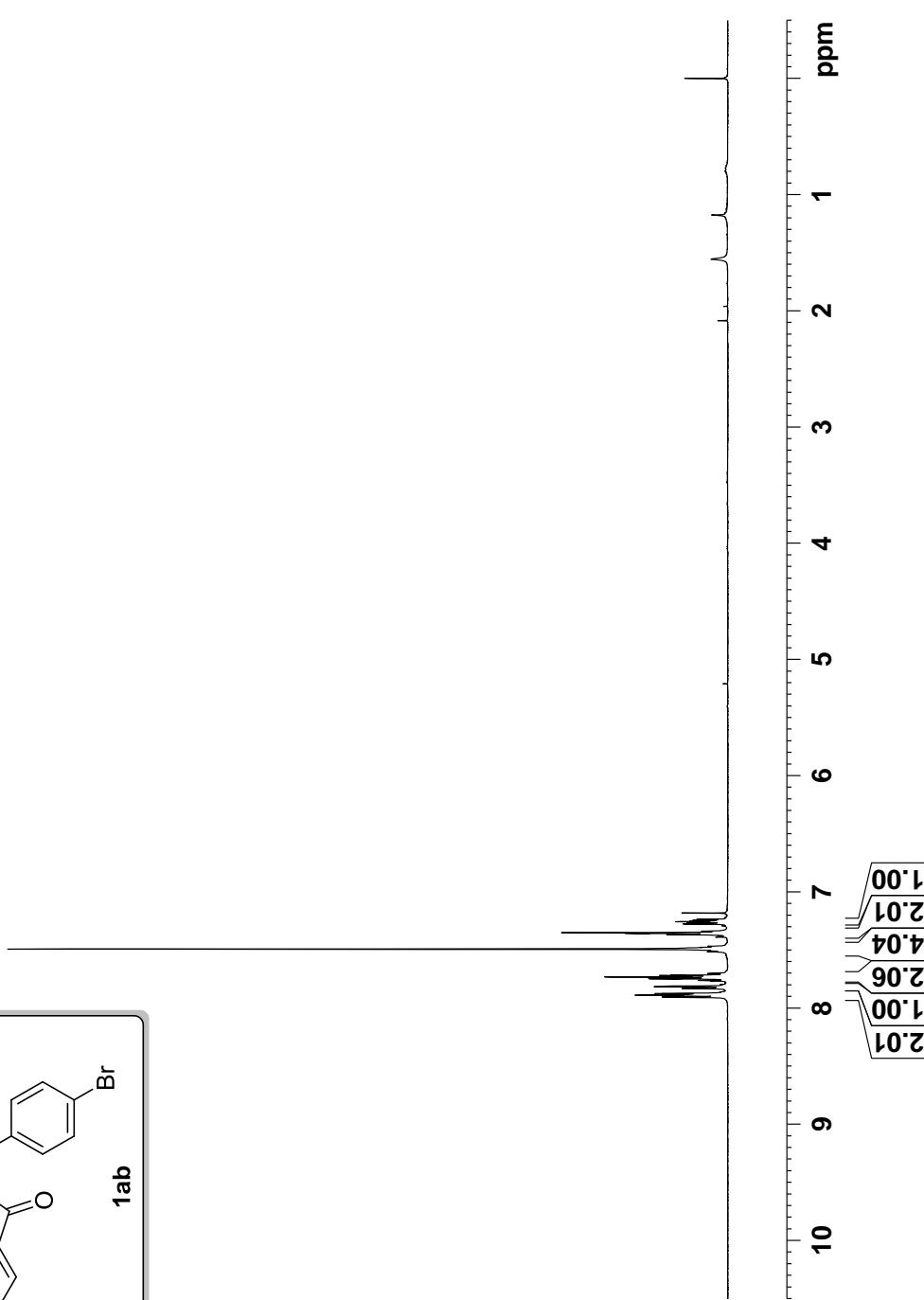
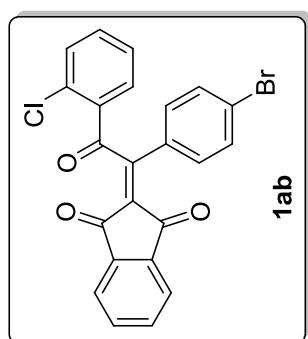
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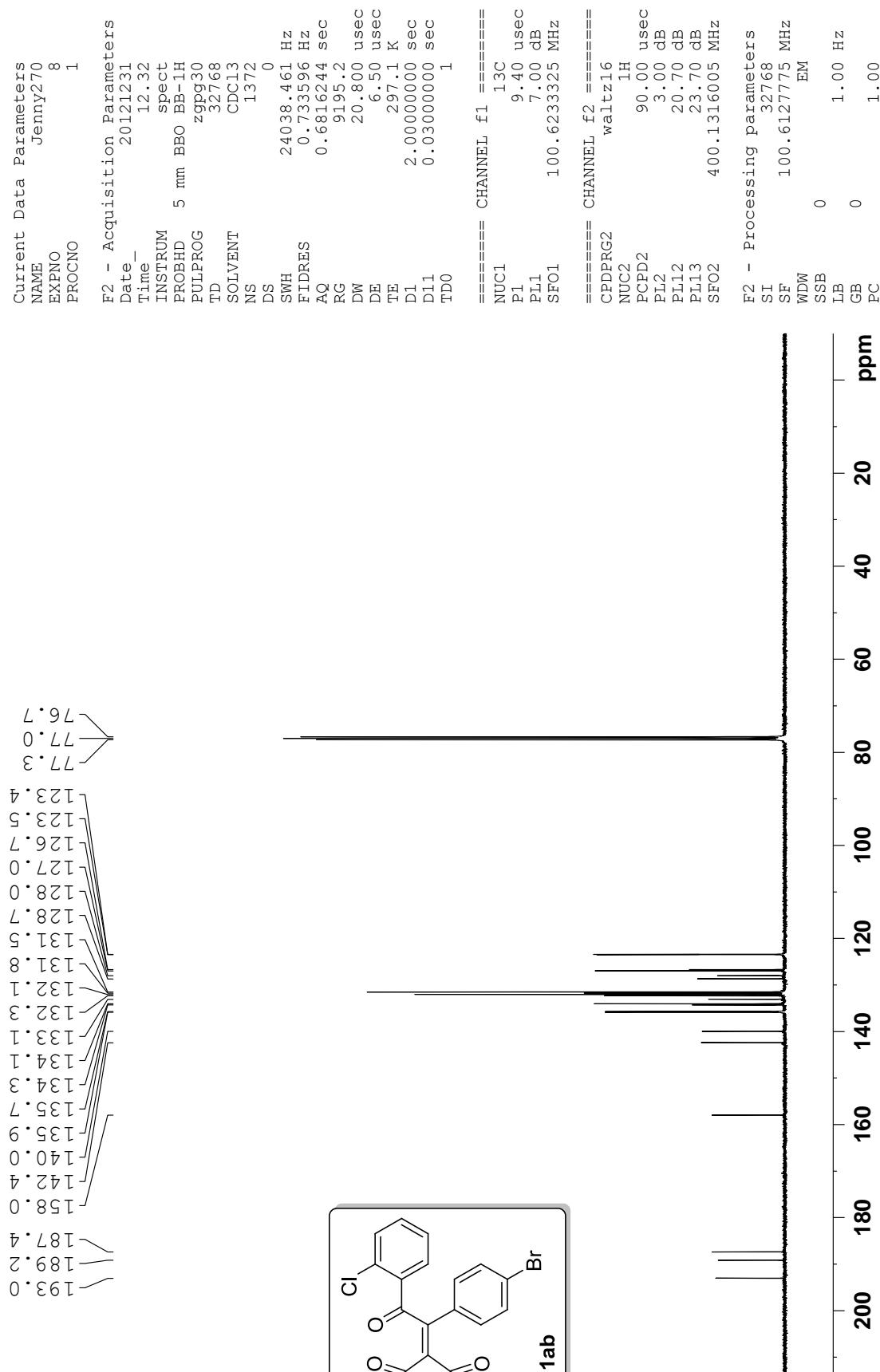
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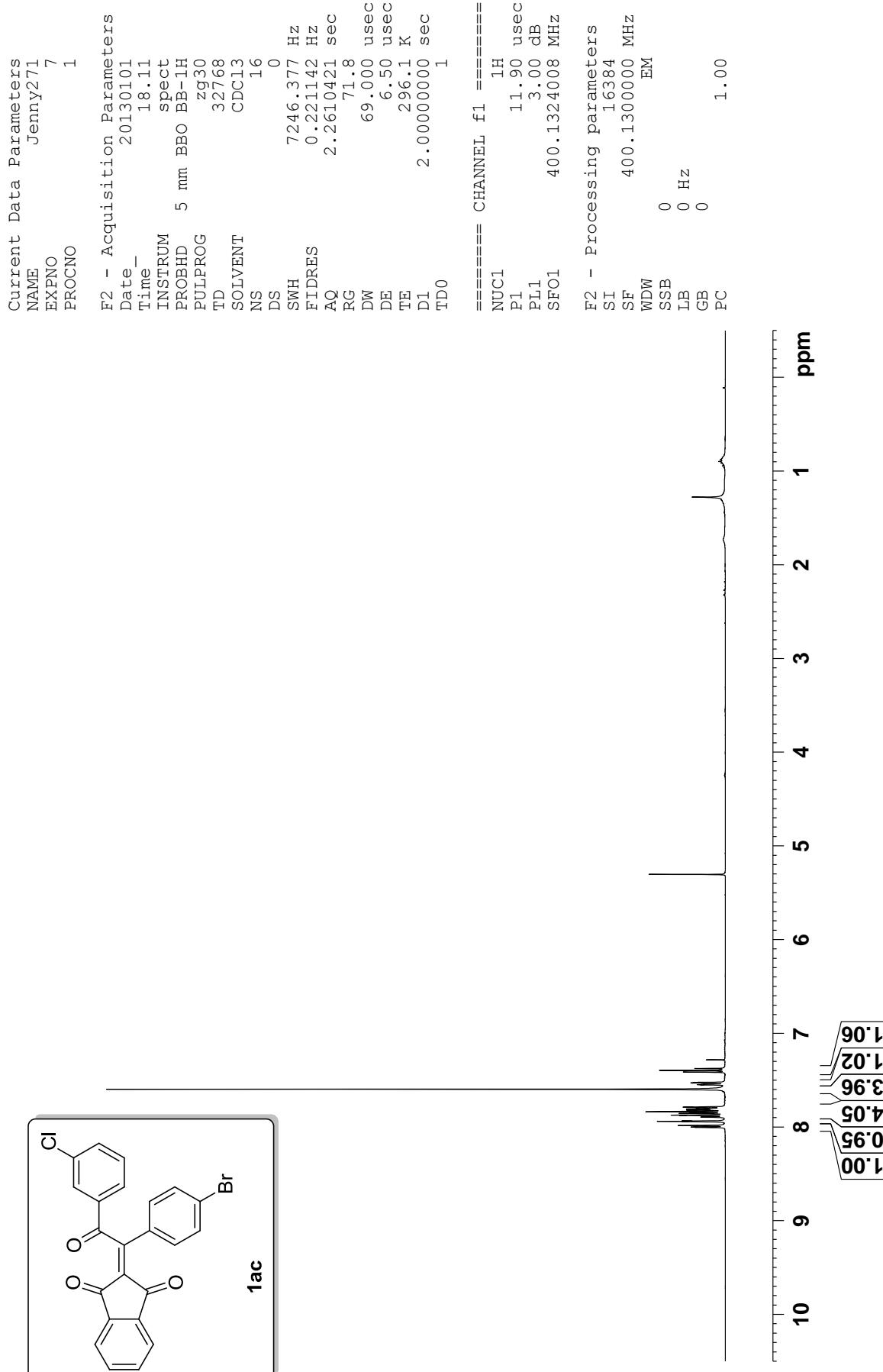
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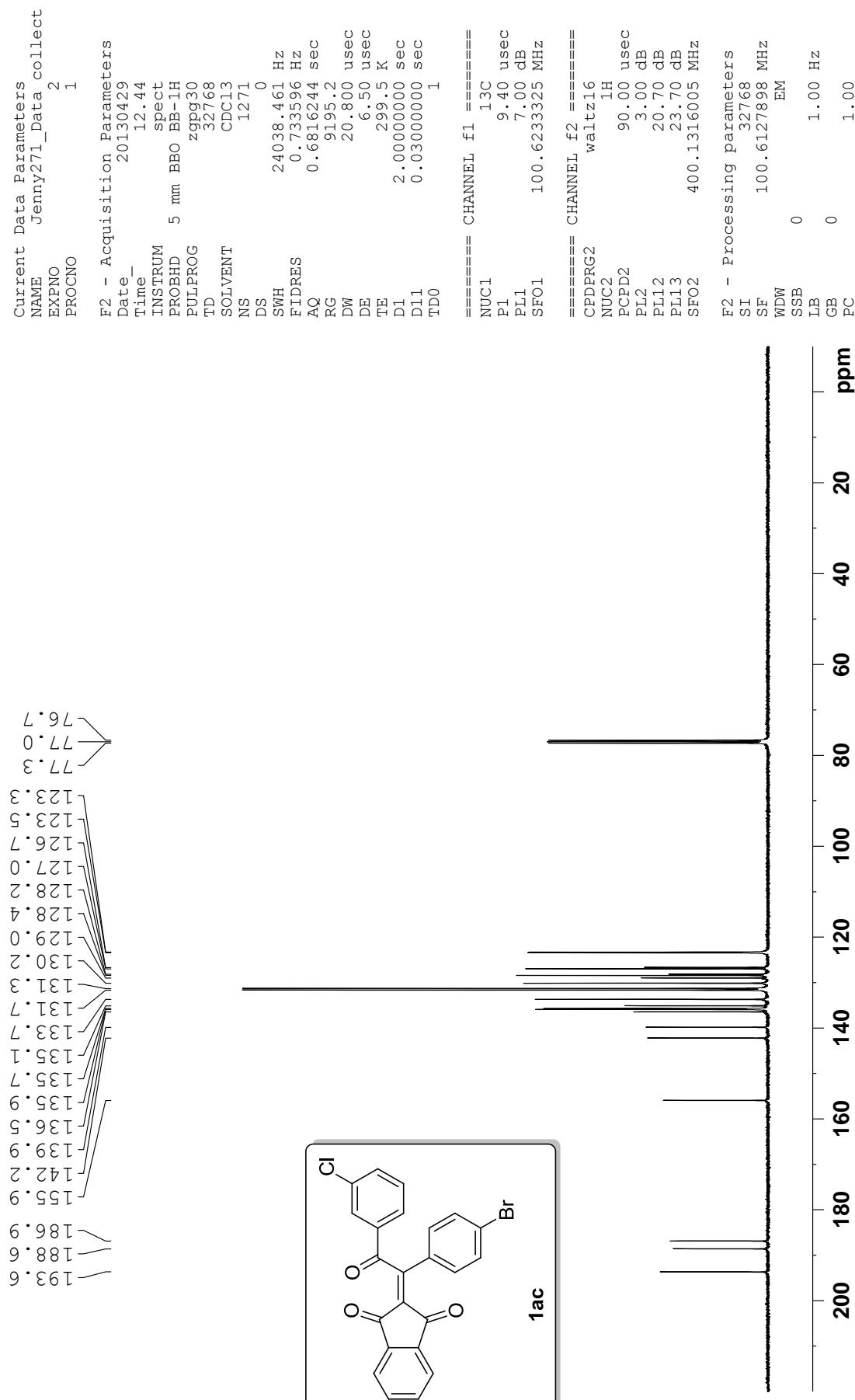
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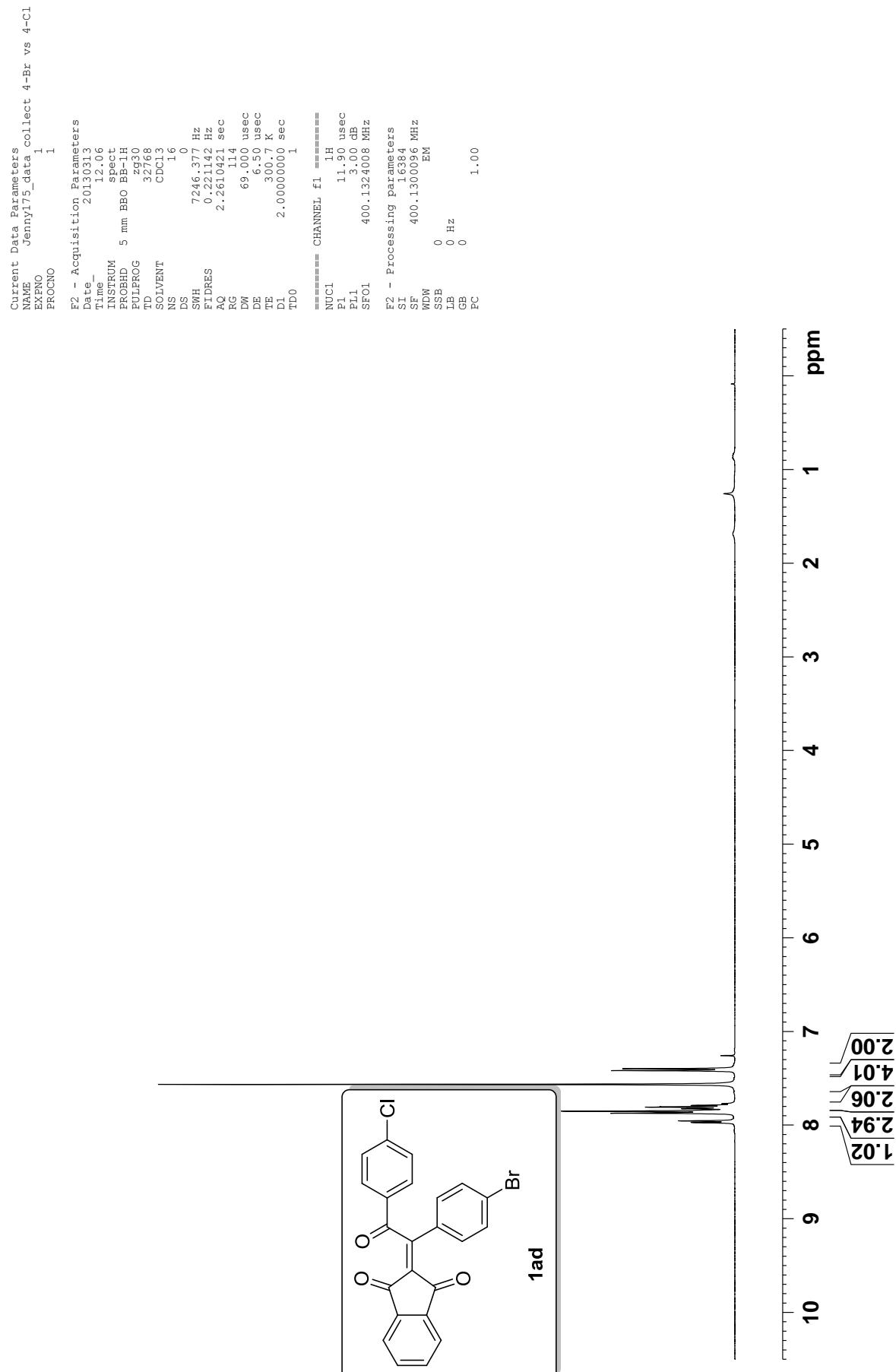
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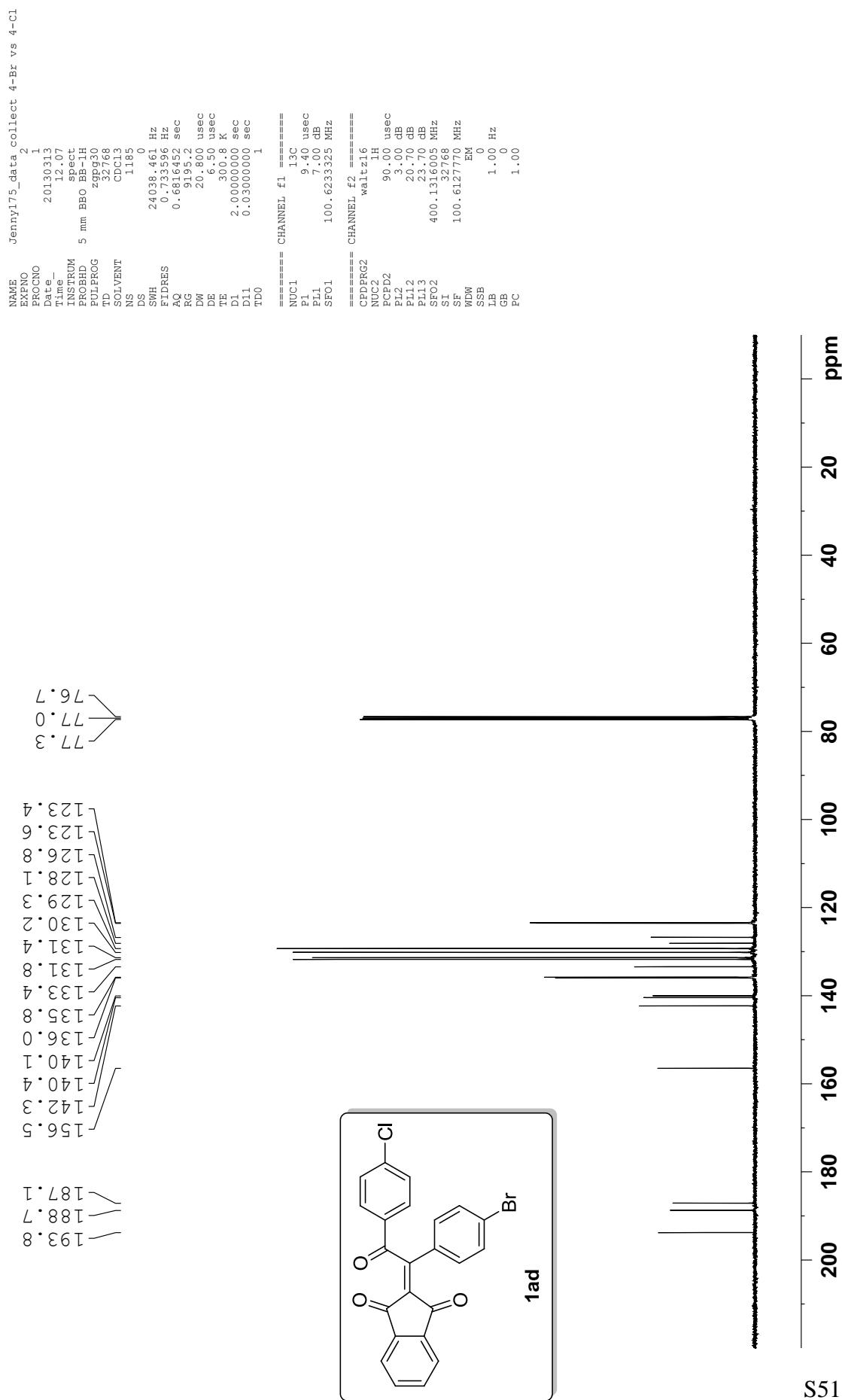


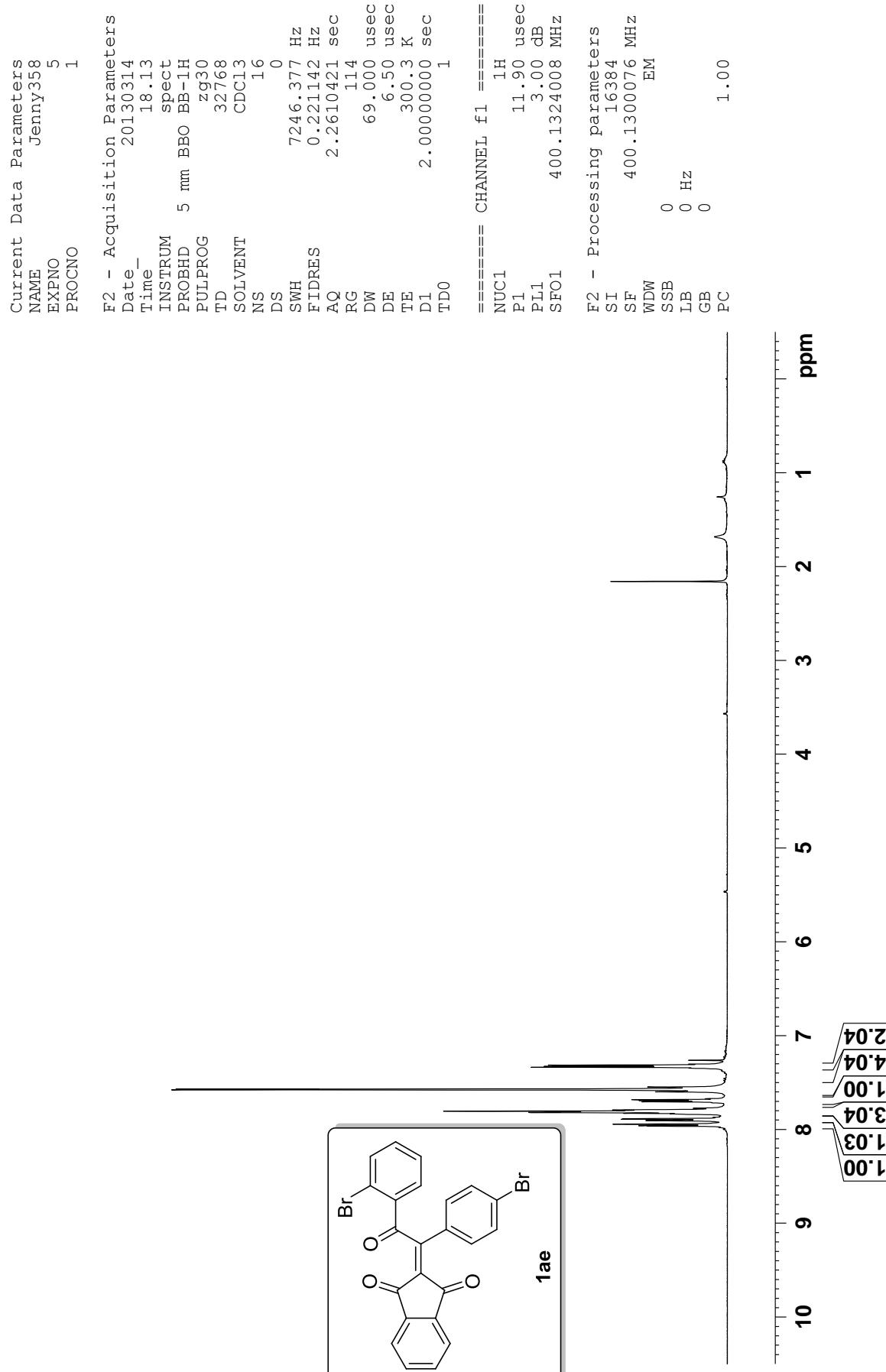


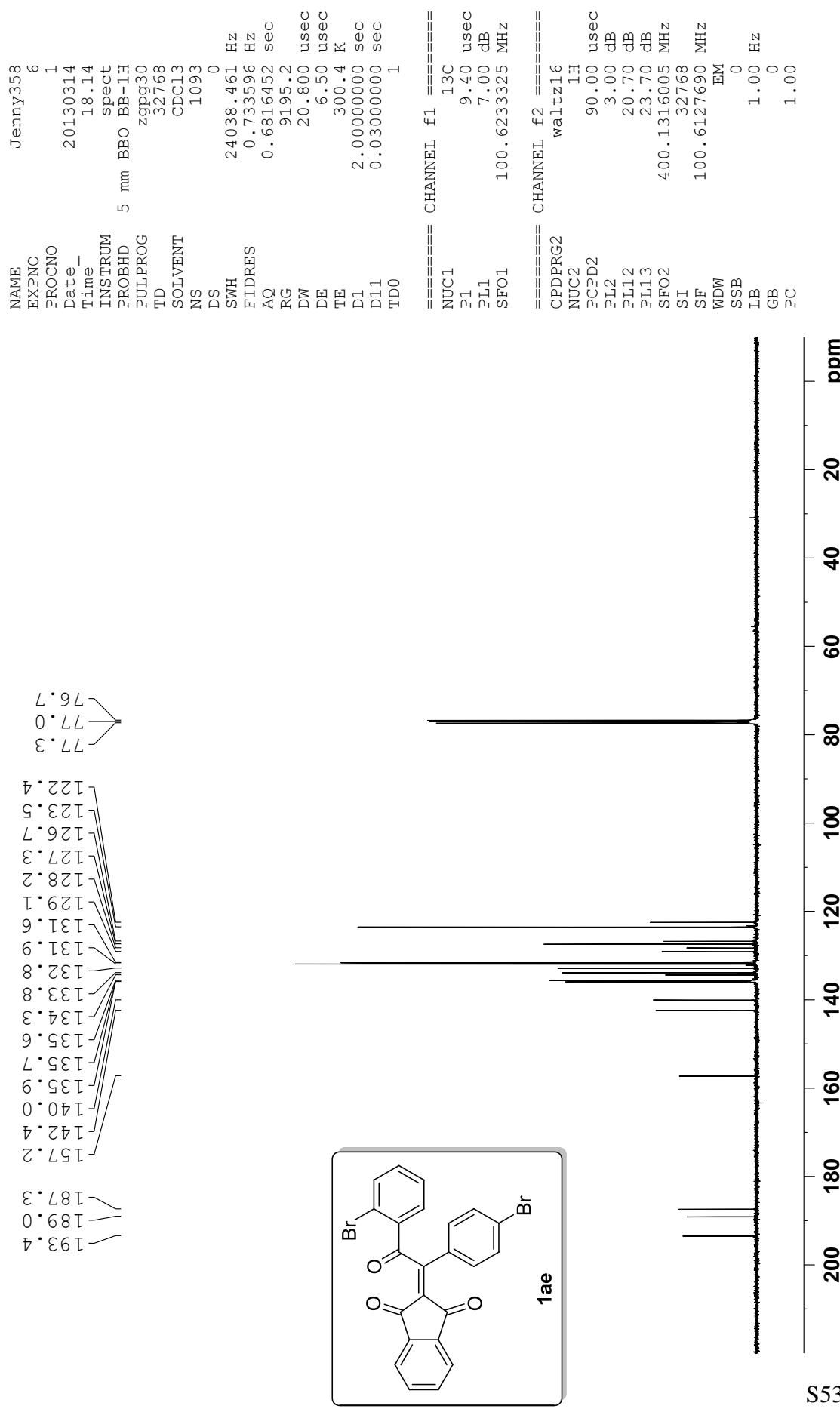


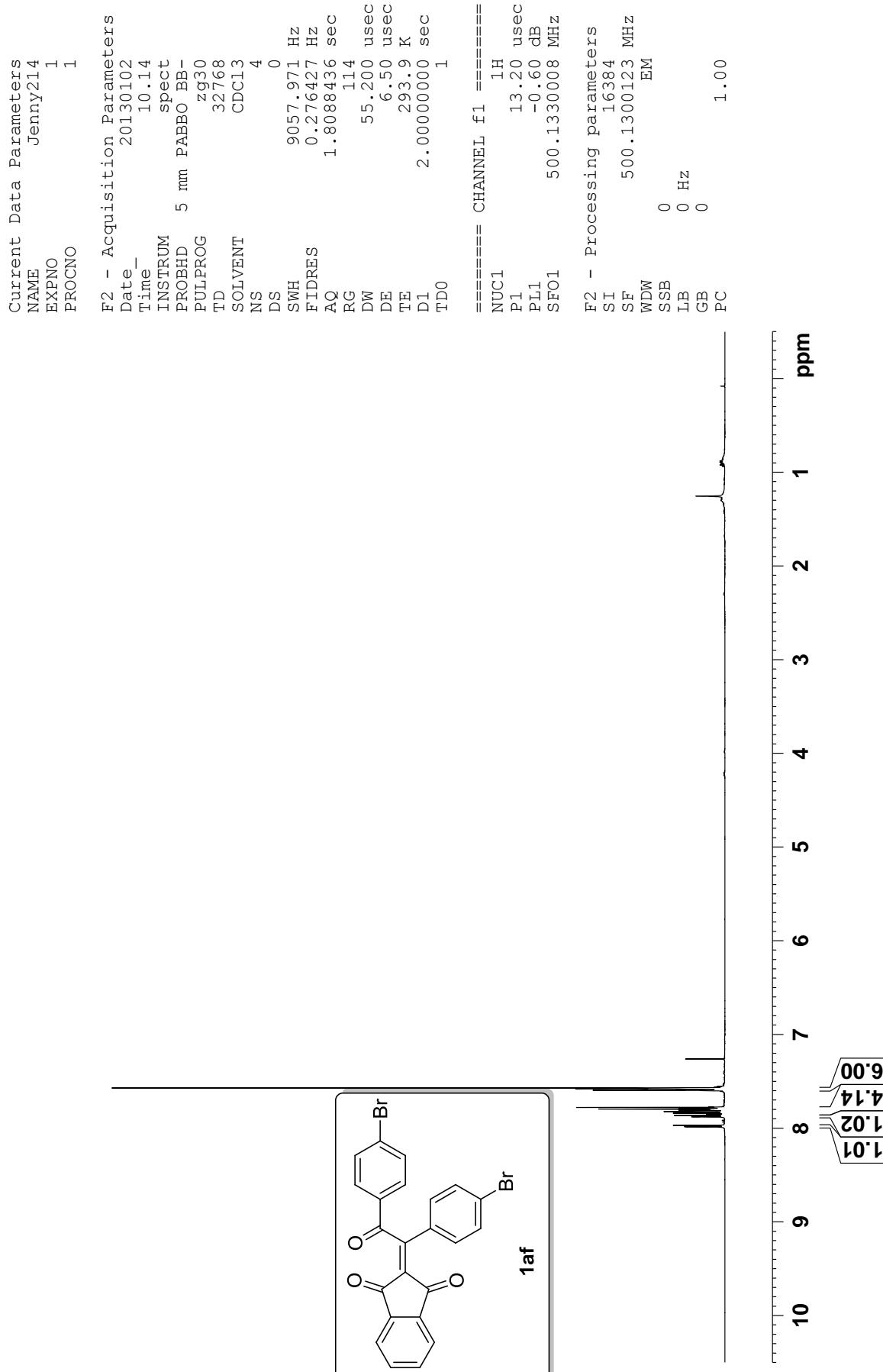


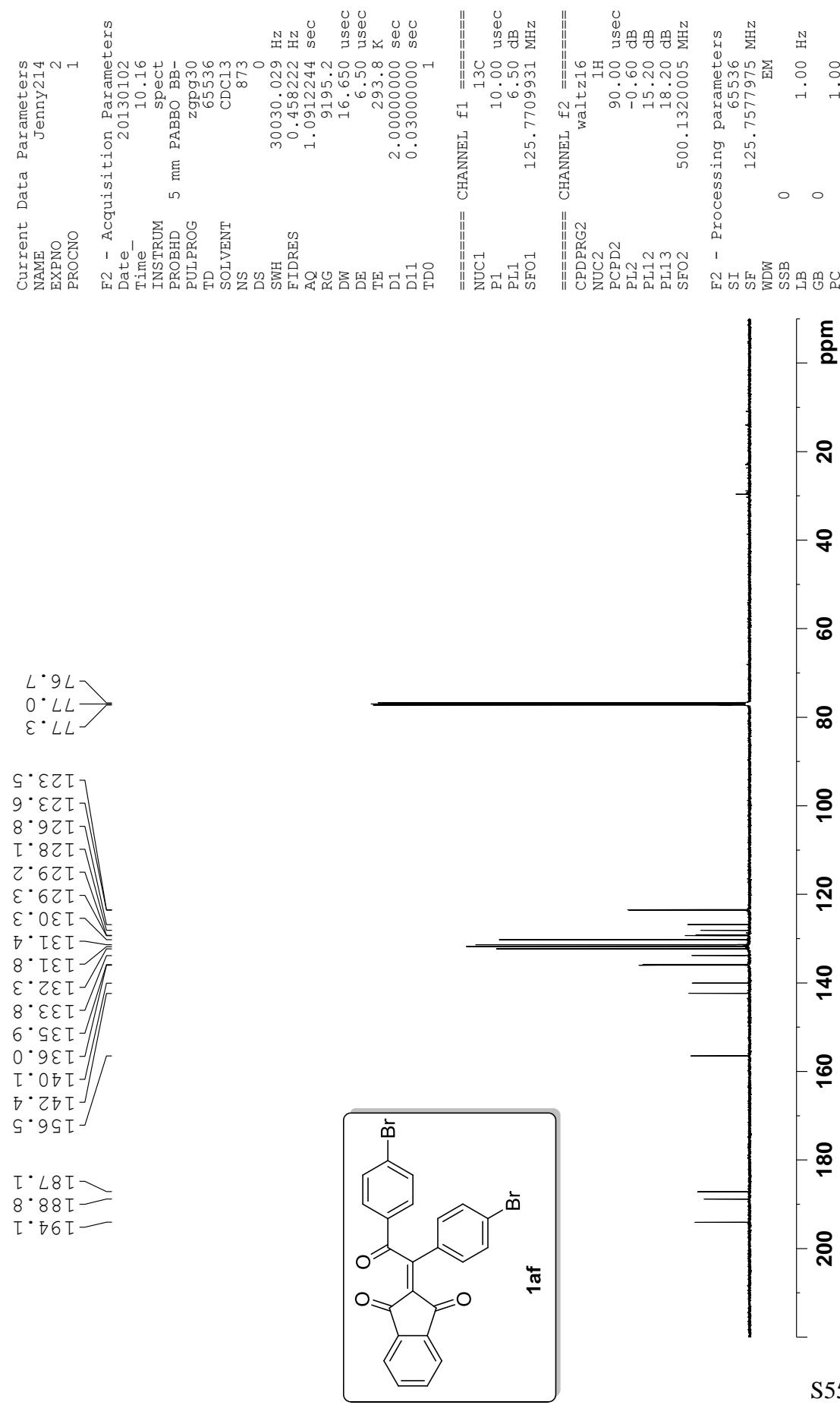


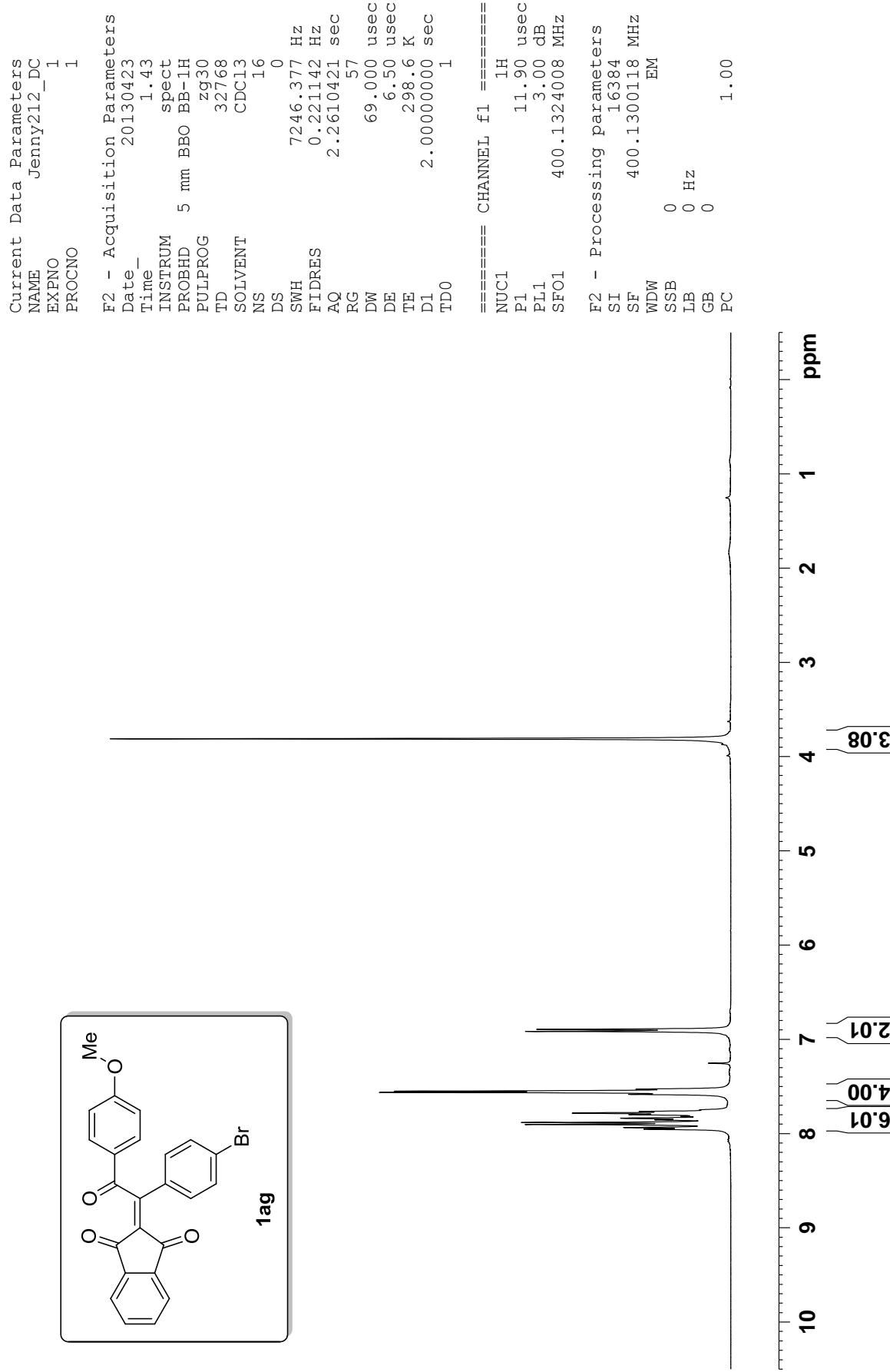












Current Data Parameters

NAME	Jenny212_DC
EXPNO	2
PROCNO	1

F2 - Acquisition Parameters

Date	20130423
Time	1.45
INSTRUM	spect
PROBHD	5 mm BBO BB-1H
PULPROG	zgpg30
TD	32768
SOLVENT	CDC13
NS	826
DS	0
SWH	24038.461 Hz
FIDRES	0.733596 Hz
AQ	0.6816244 sec
RG	9195.2
DW	20.800 usec
DE	6.50 usec
TE	29.87 K
D1	2.00000000 sec
D11	0.03000000 sec
TDO	

===== CHANNEL f1 =====

NUC1	13C
P1	9.40 usec
PL1	7.00 dB
SFO1	100.6233325 MHz

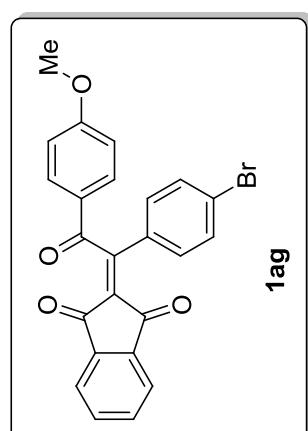
===== CHANNEL f2 =====

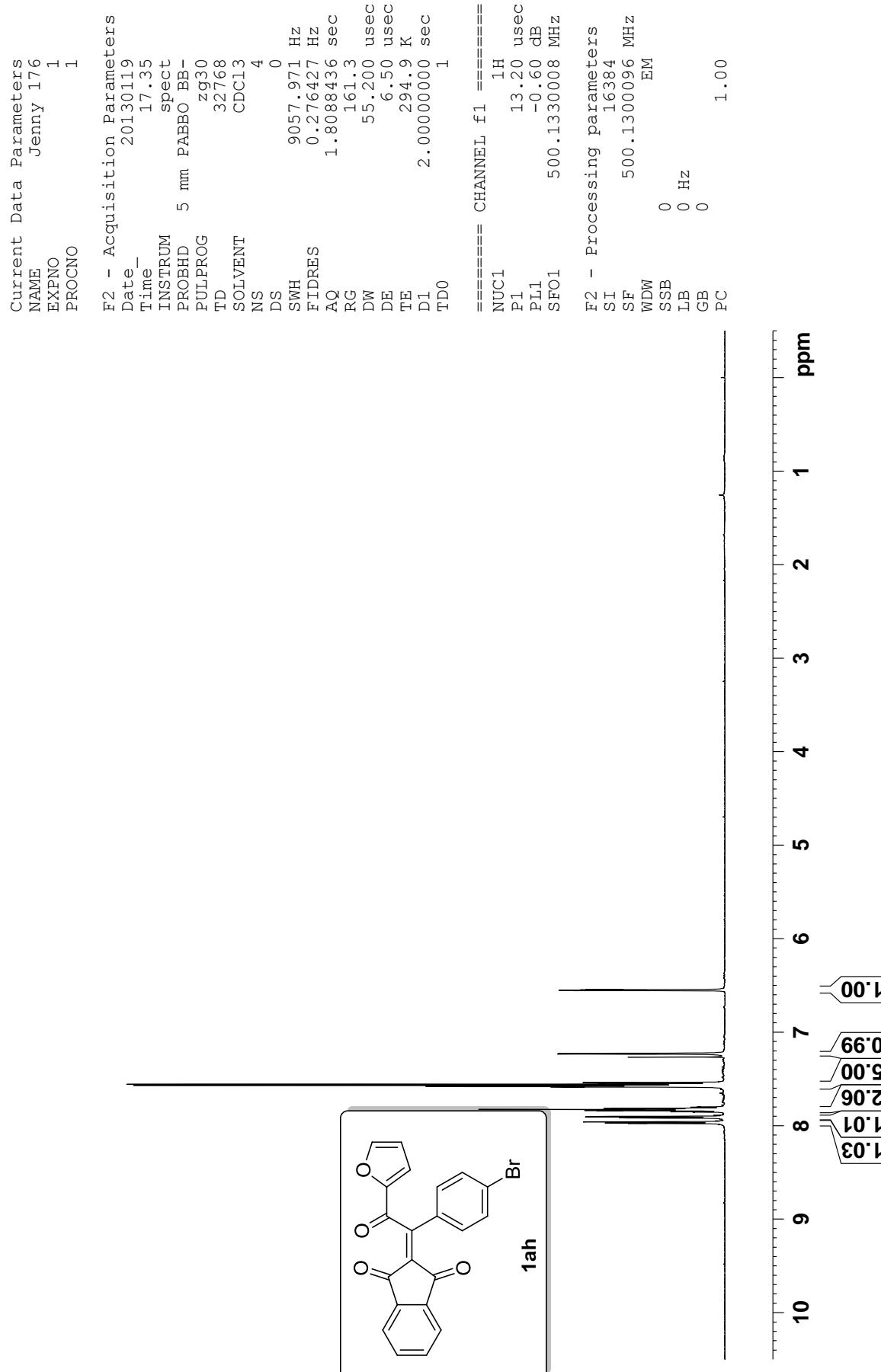
CPDPRG2	waltz16
NUC2	1H
PCPD2	90.00 usec
PL2	3.00 dB
PLL2	20.70 dB
PL13	23.70 dB
SFO2	400.1316005 MHz

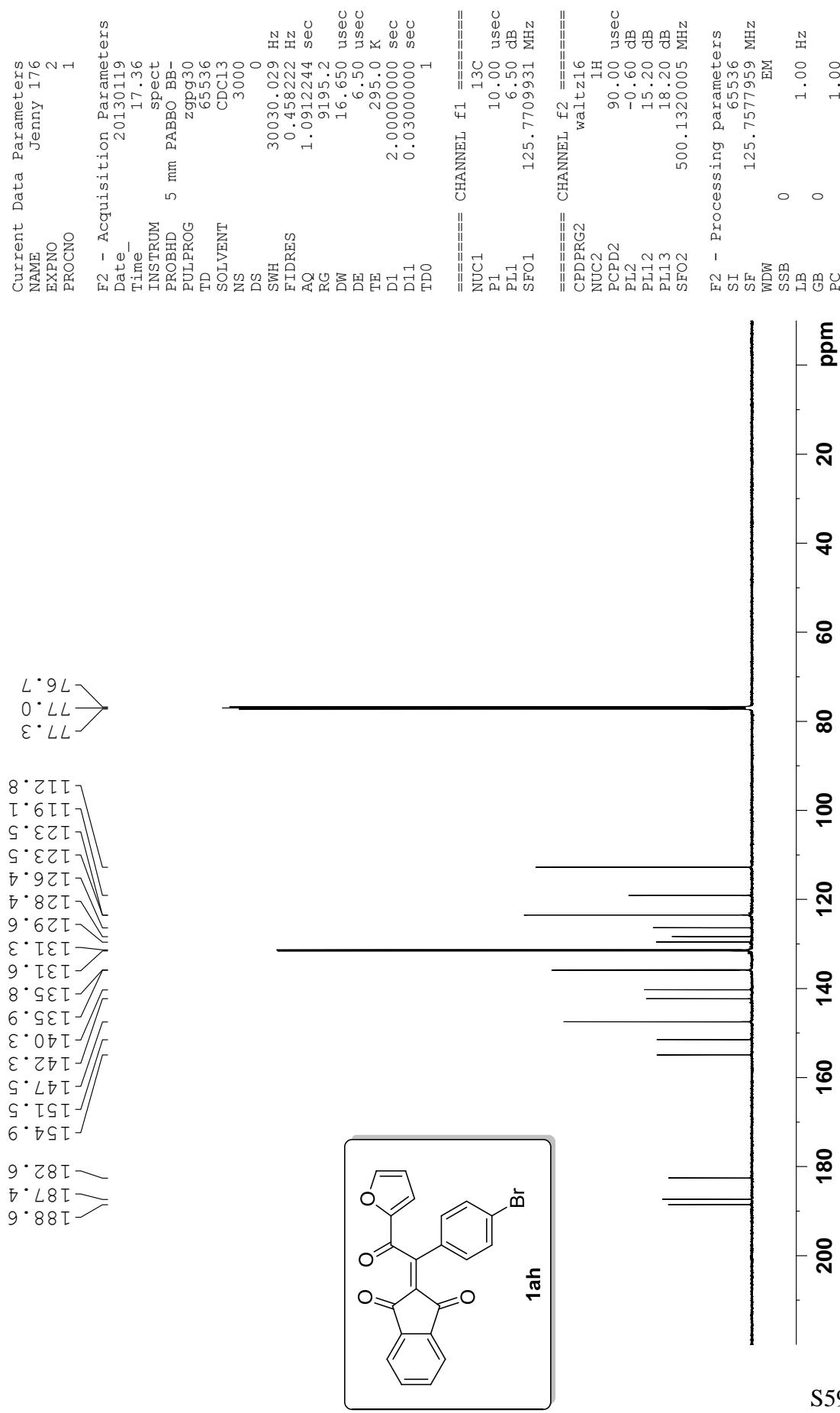
F2 - Processing parameters

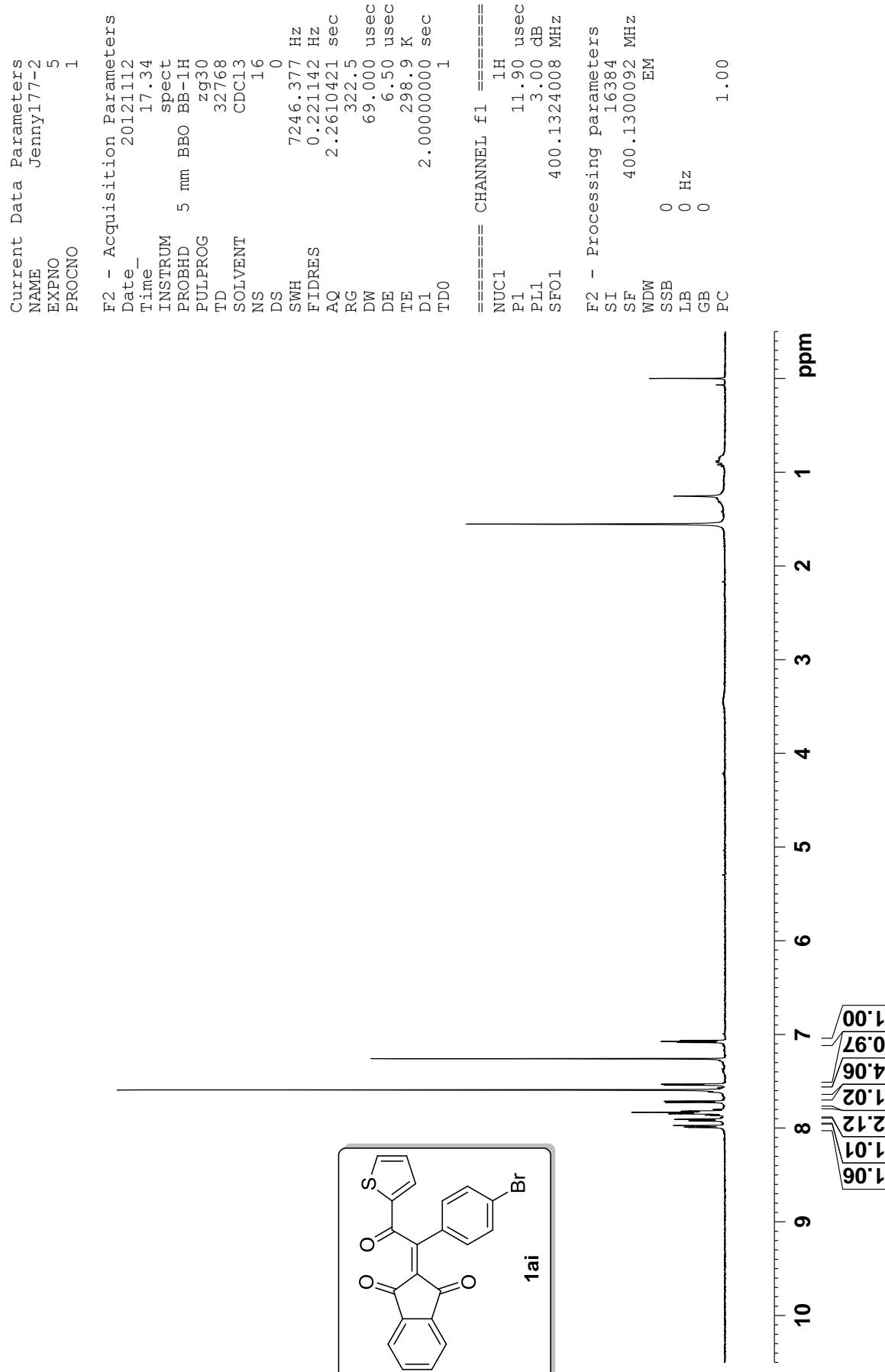
SI	32768
SF	100.6127778 MHz
WDW	EM
SSB	0
LB	1.00 Hz
GB	0
PC	1.00

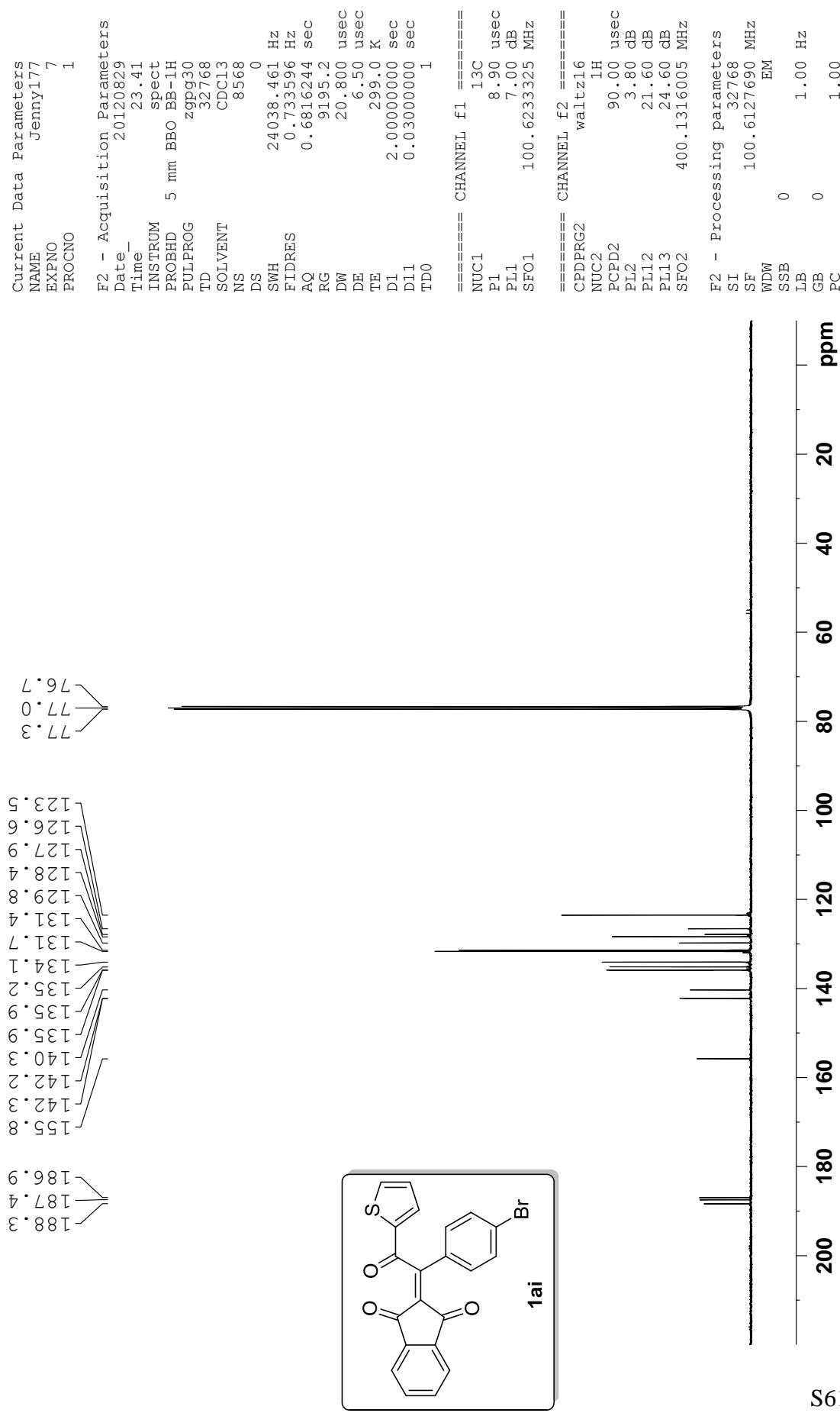
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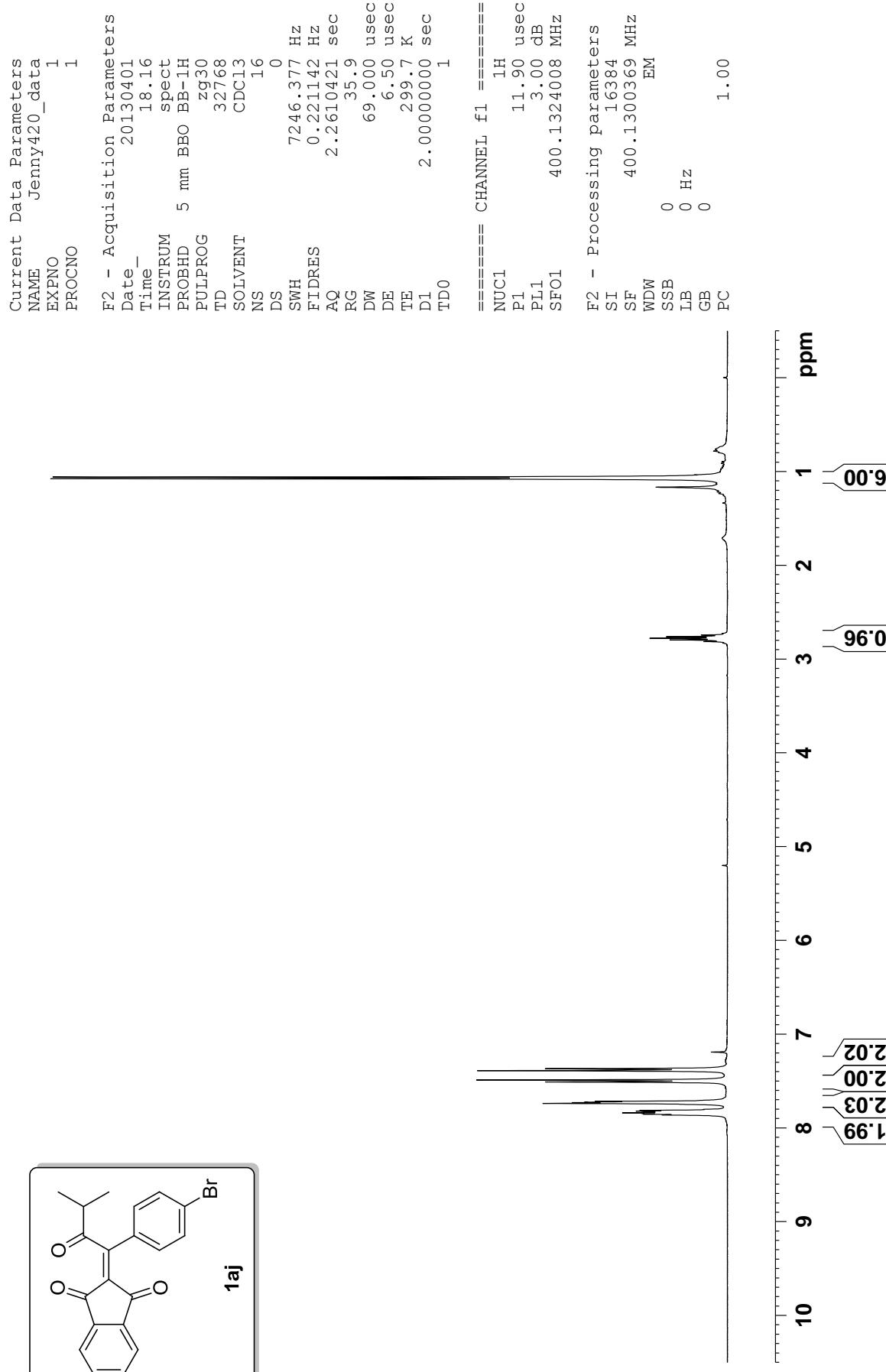


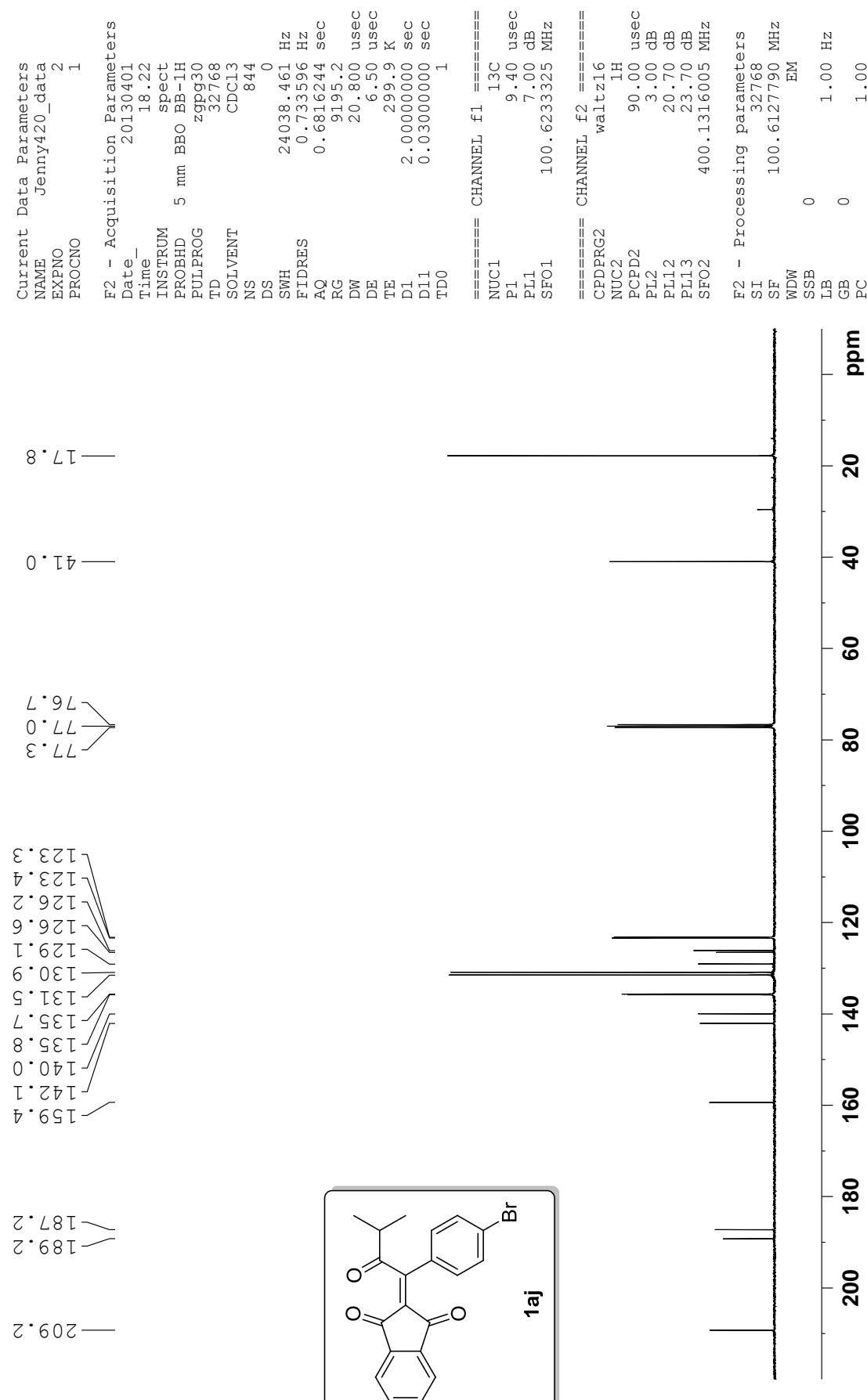


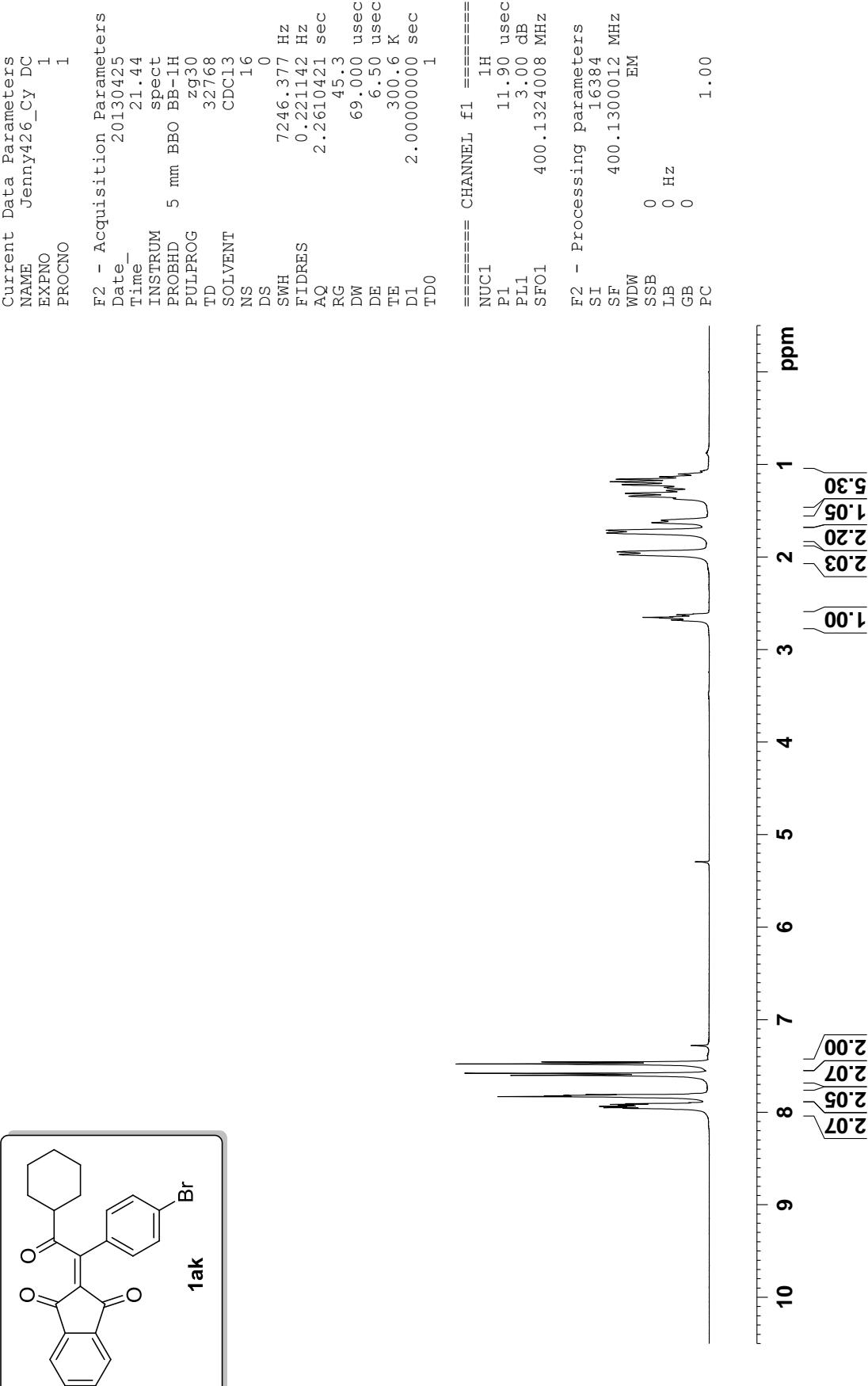


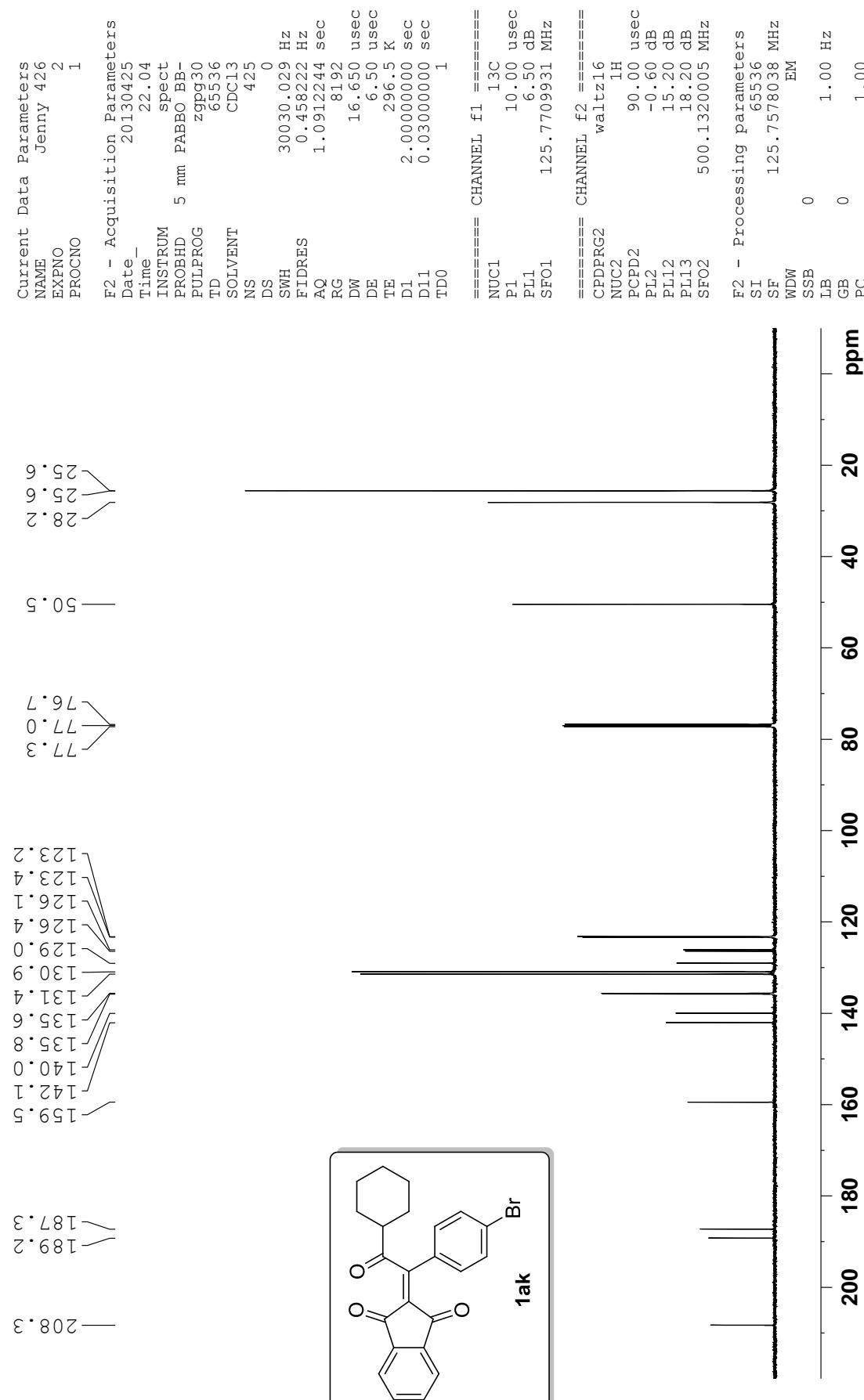












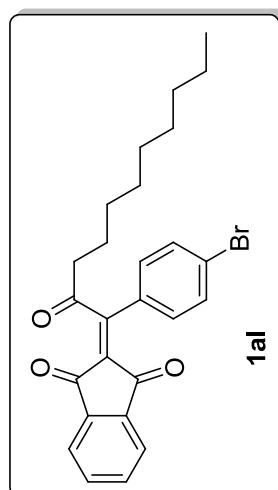
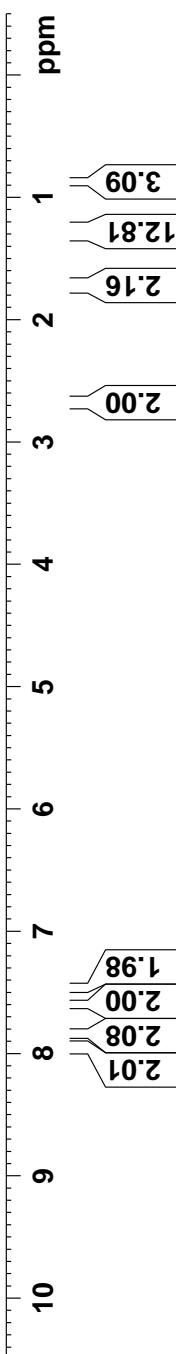
Current NAME	Data EXPNO	Parameters
Eric	267	decanoyl_DC
	1	
	1	
PROCNO		
EF2 - Acquisition Parameters		
Date	20130430	
Time	22.07	
INSTRUM	spect	
PROBHD	5 mm	BBO
PULPROG	BB-1H	
TD	zg30	
SOLVENT	32768	
NS	CDC13	
DS	16	
SWH	0	
FIDRES	7246.37 Hz	
AQ	0.221142 Hz	
RG	2.2610421 sec	
DW	57	
DE	69.000 usec	
TE	6.50 usec	
D1	300.8 K	
TD0	2.00000000 sec	

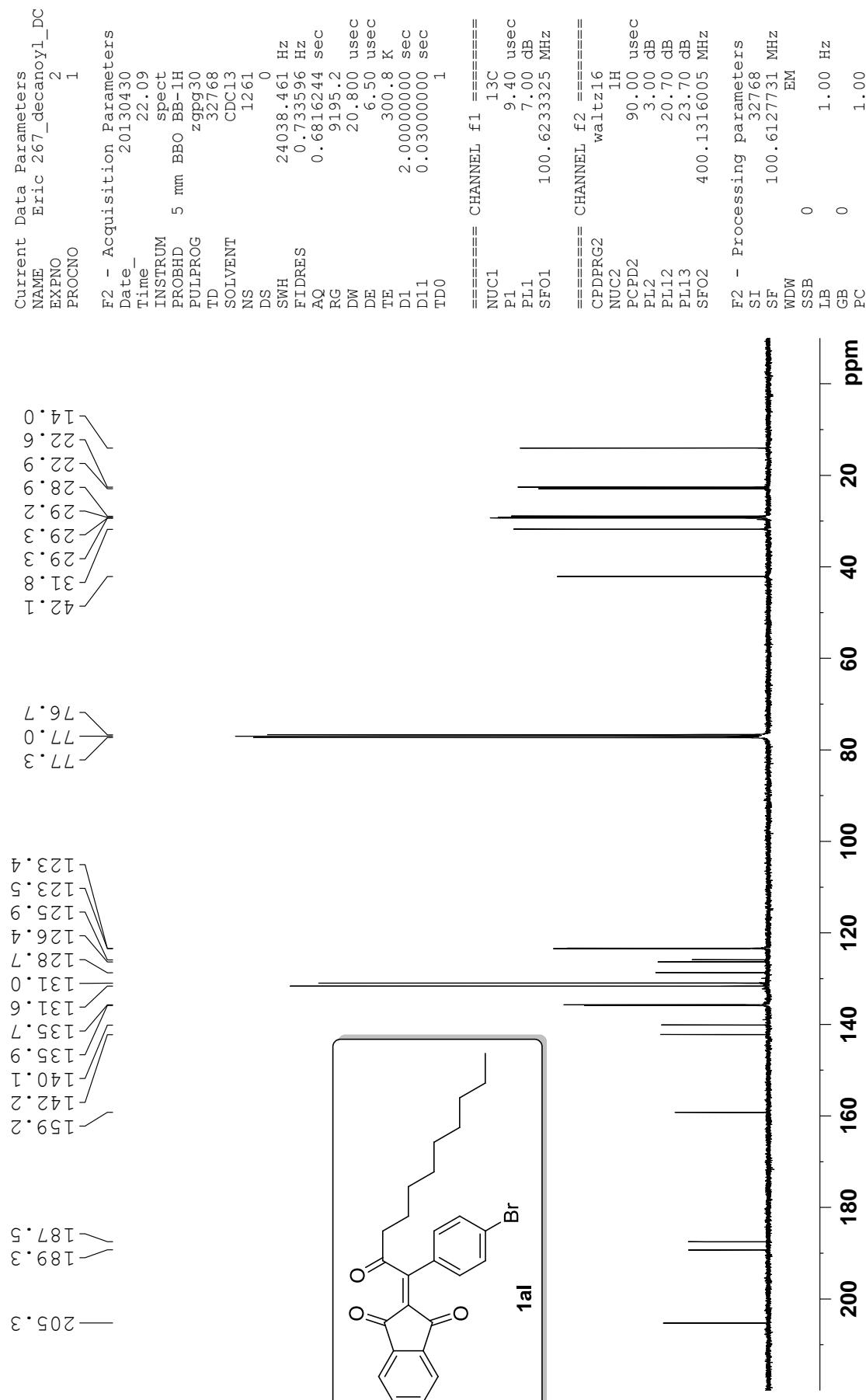
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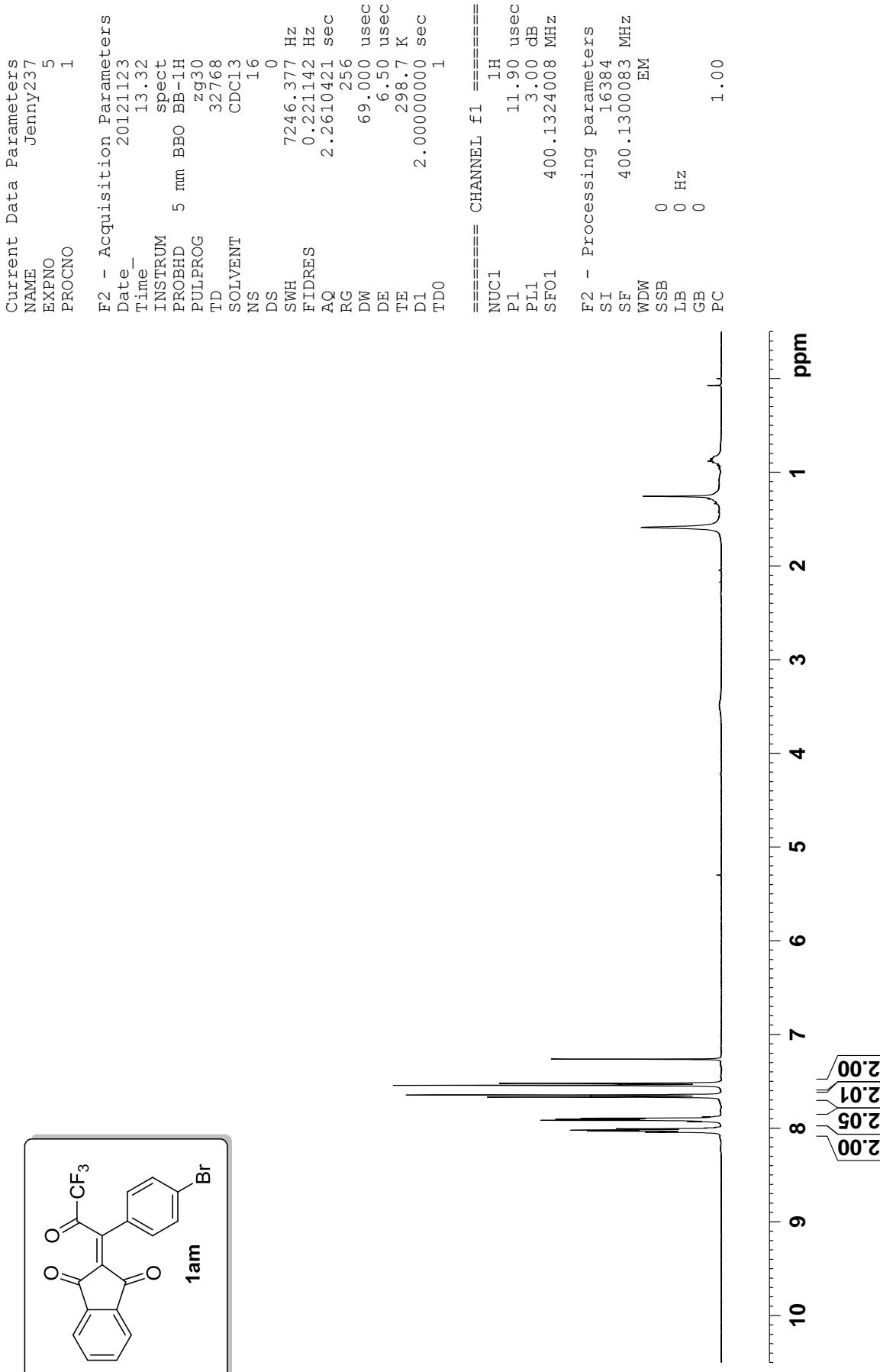
===== CHANNEL f1 =====
NUC1          1 H
P1           11.90 usec
PL1          3.00 dB
SFO1        400.1324008 MHz
                           EM

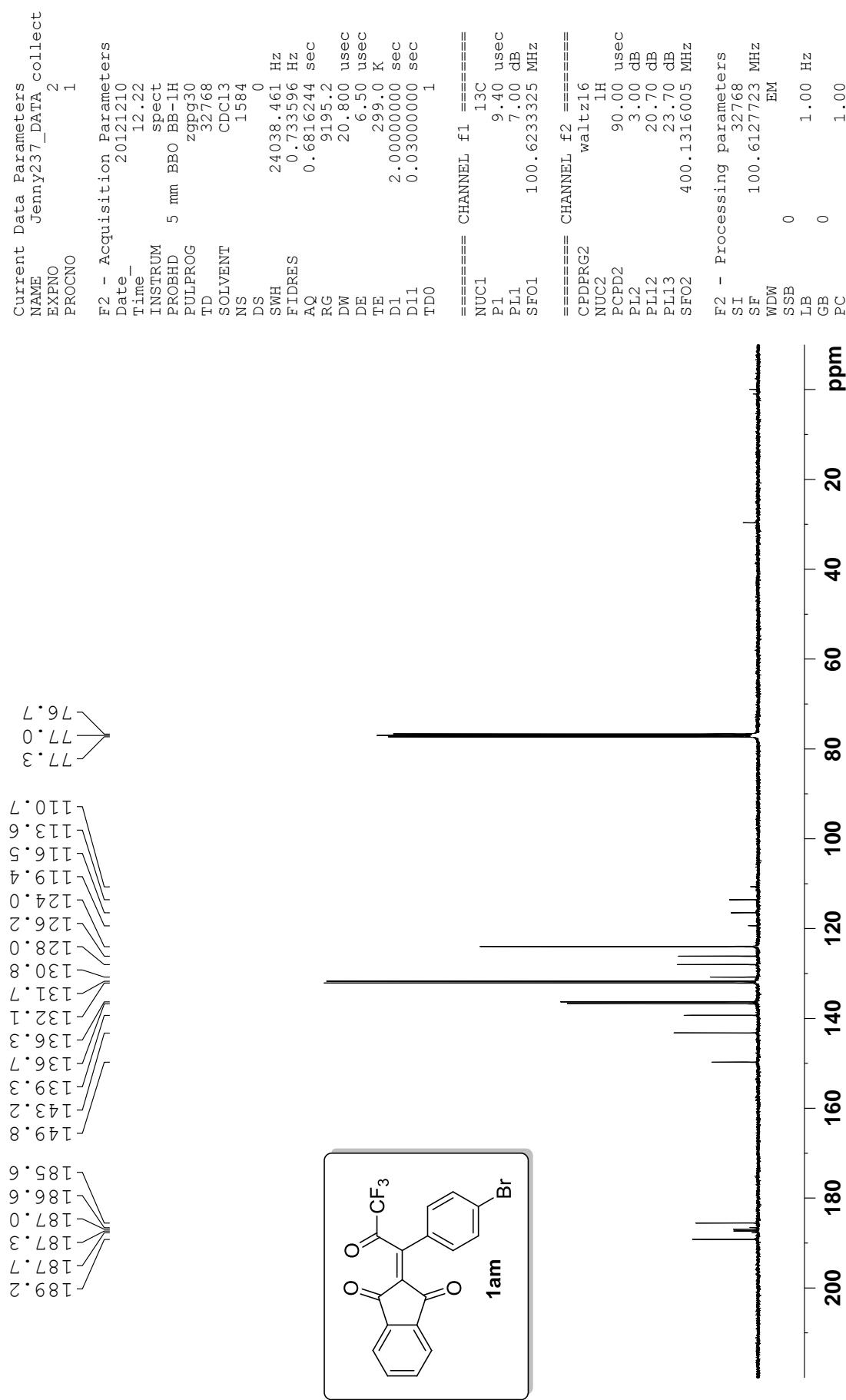
===== Processing parameters =====
SI           16384
SF          400.13000056 MHz
WDW          0 Hz
SSSB         0
LB          0
GB          0
PC          1.00

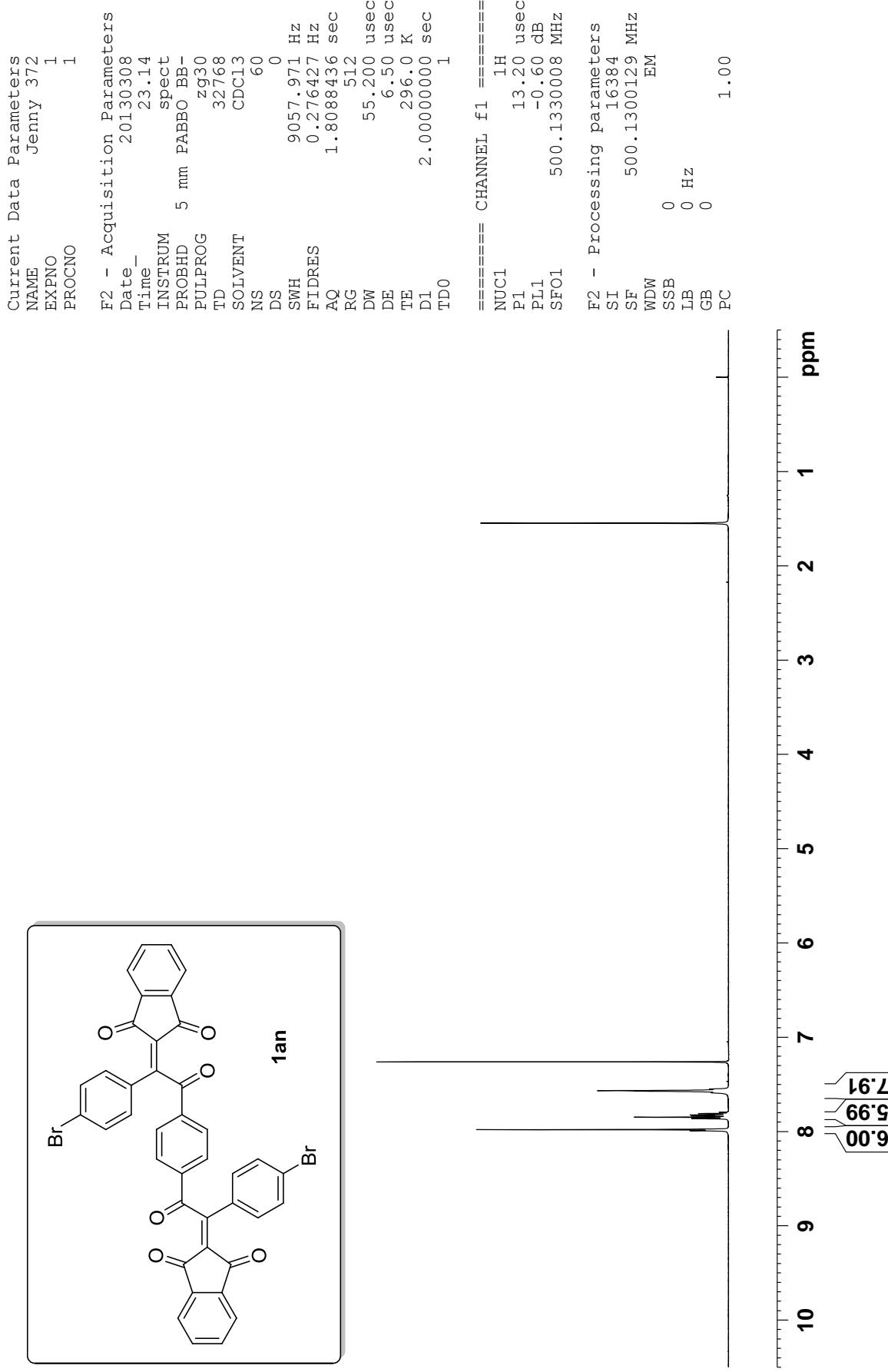
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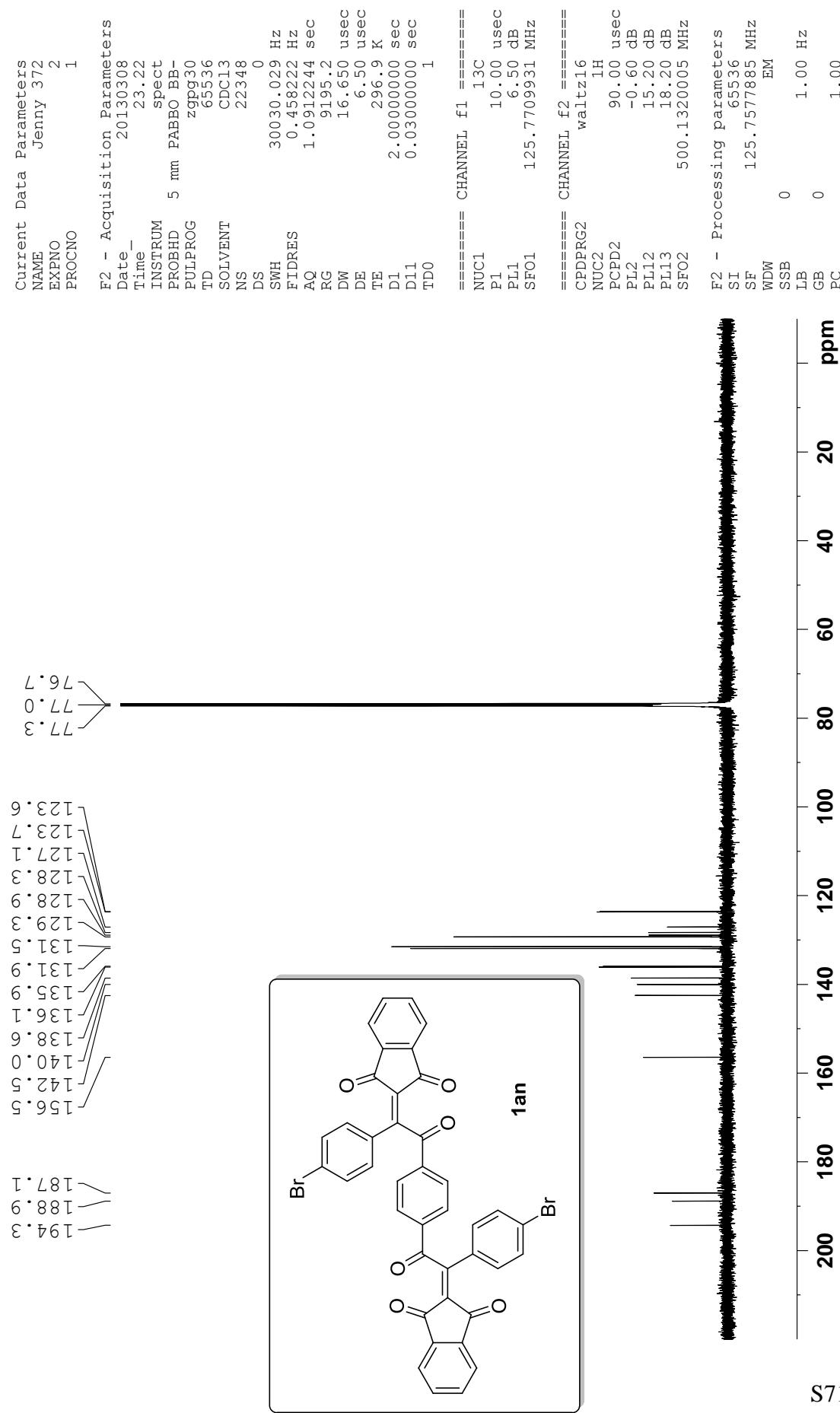


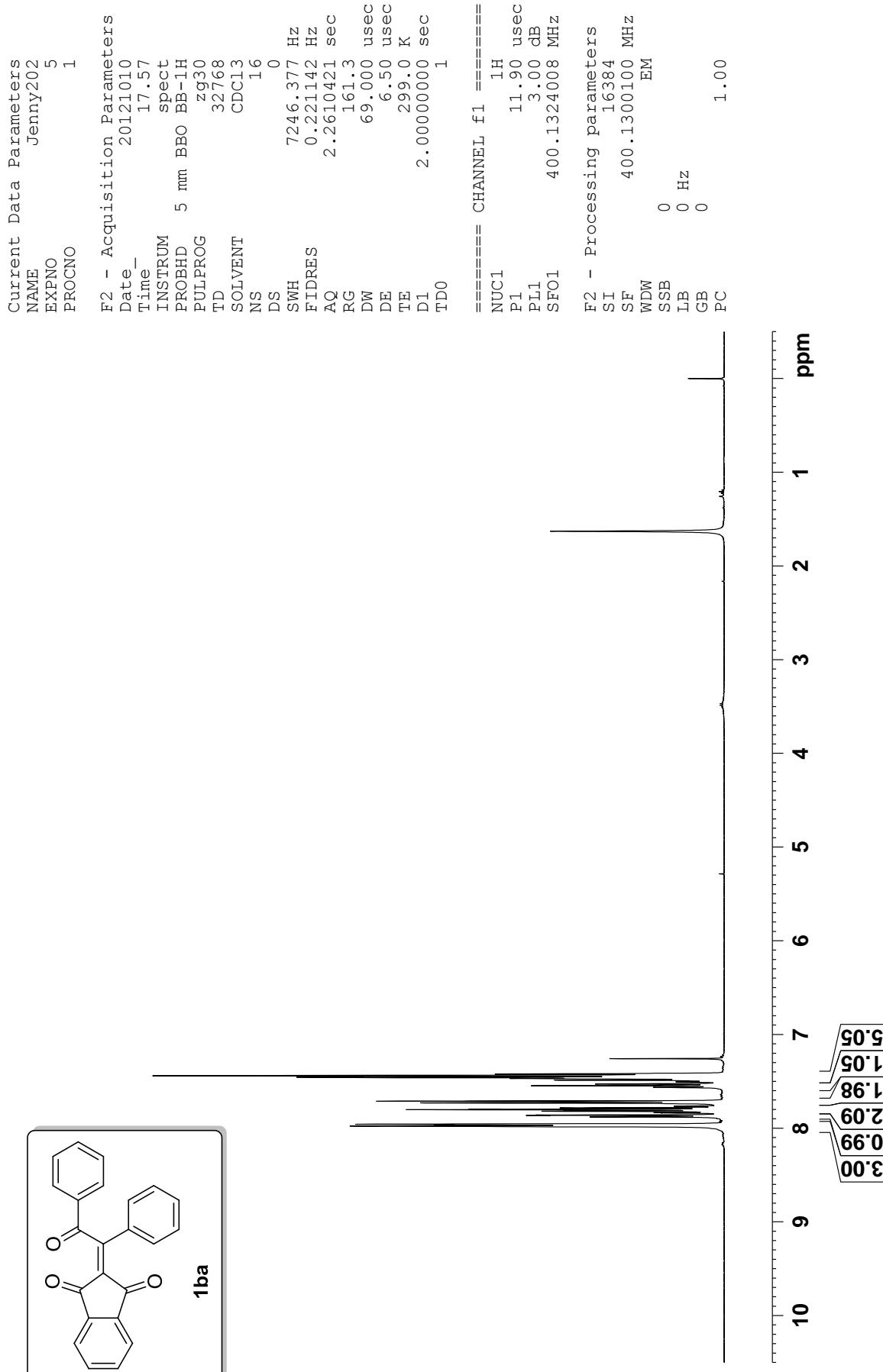


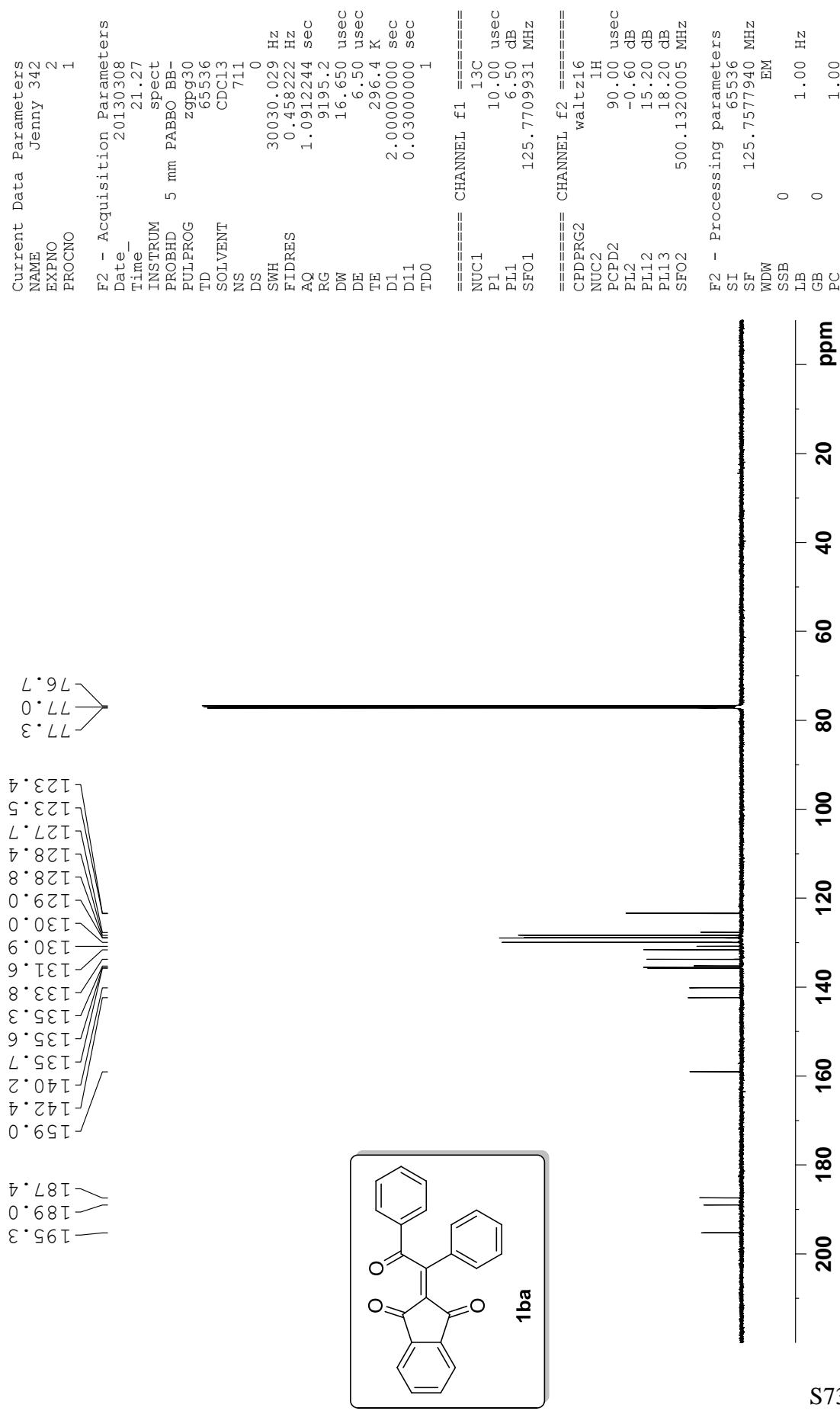


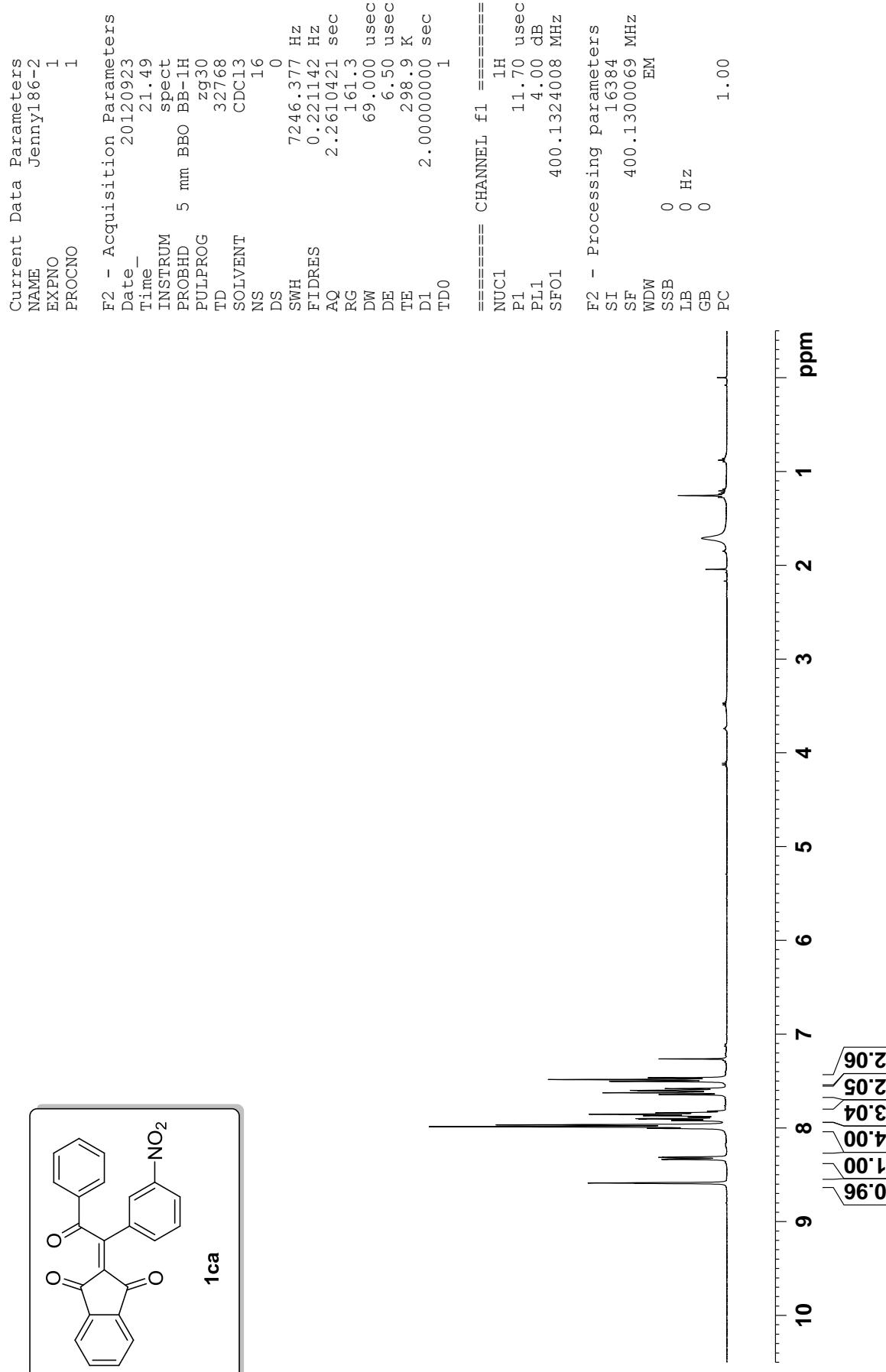


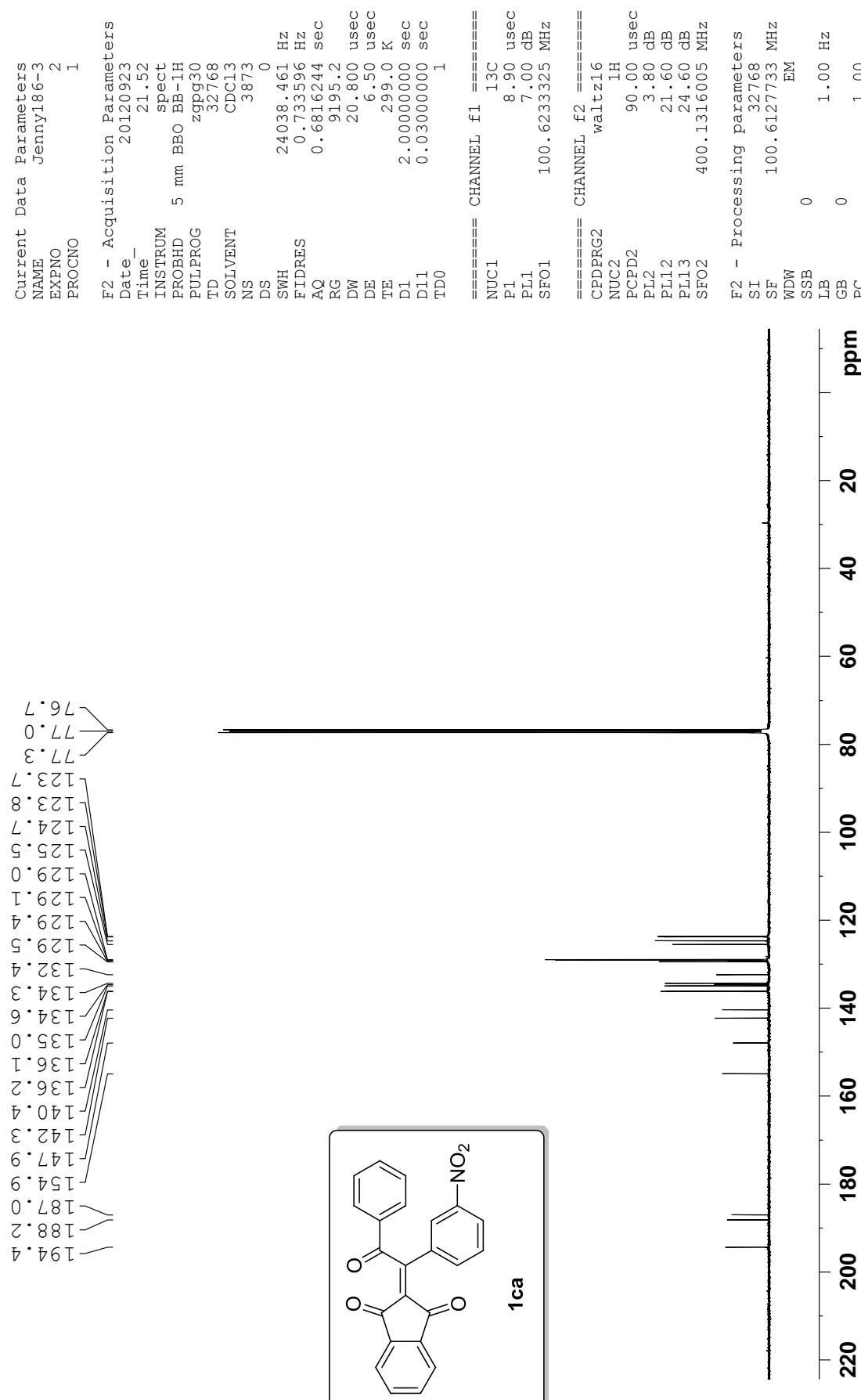


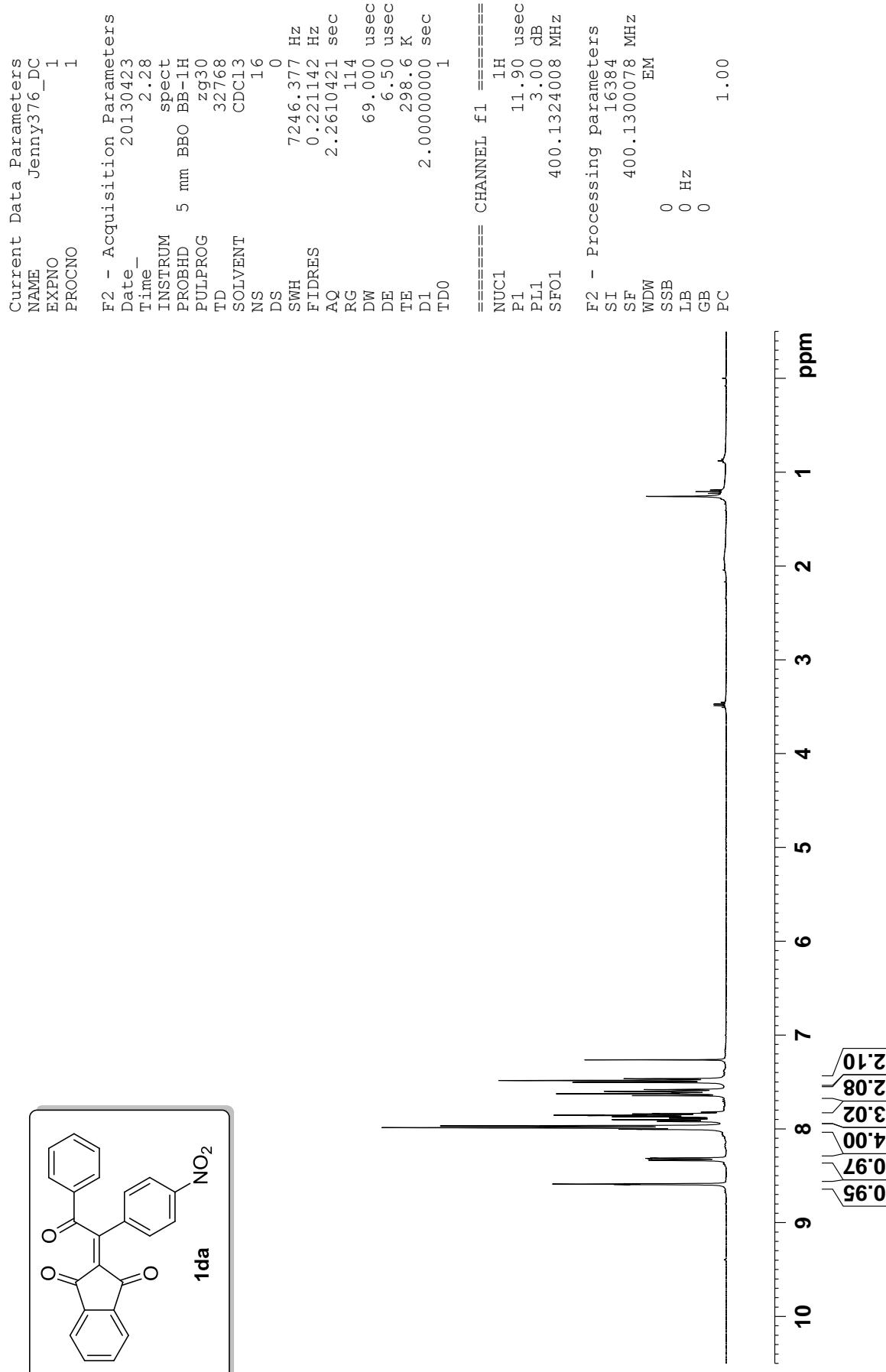


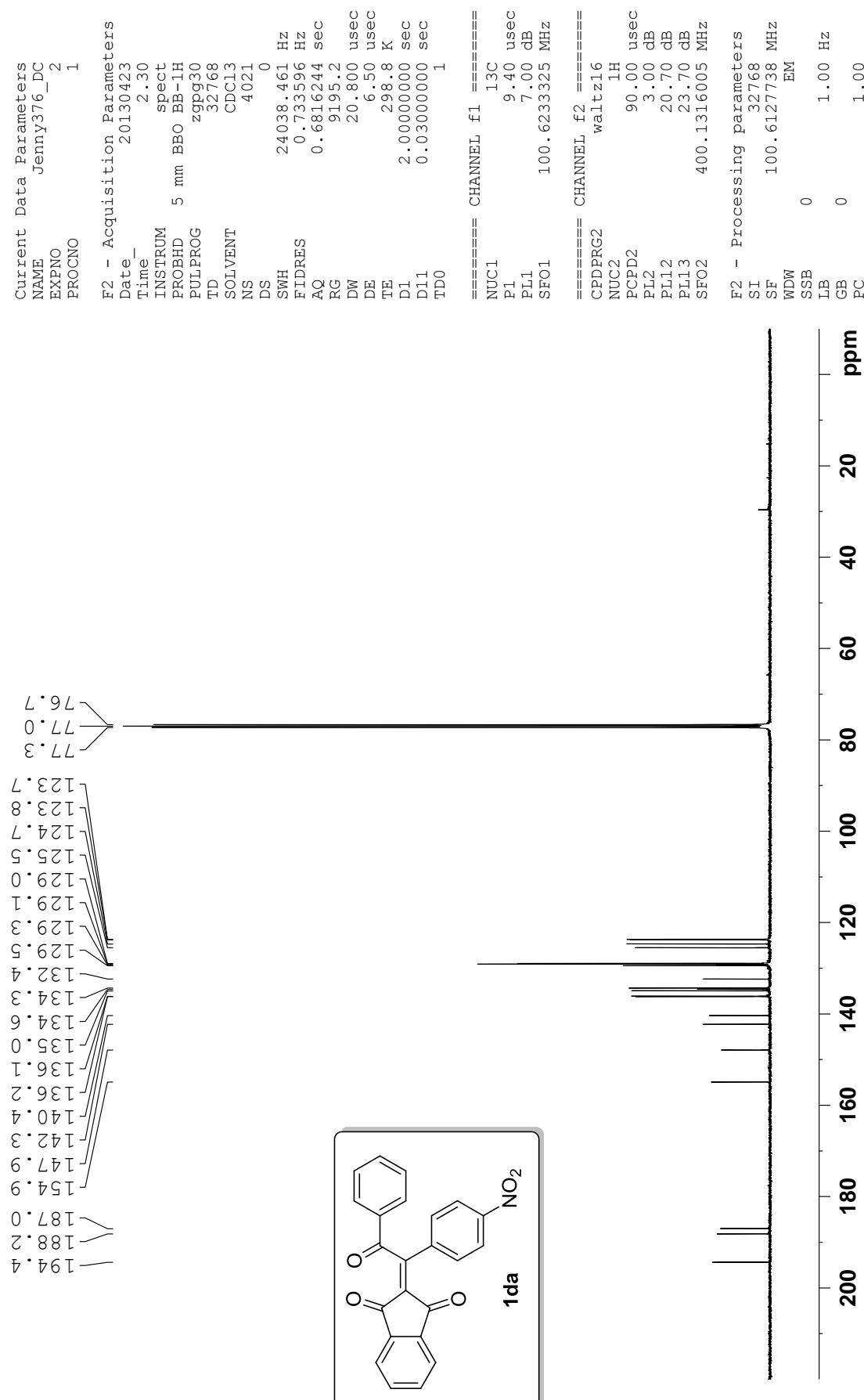


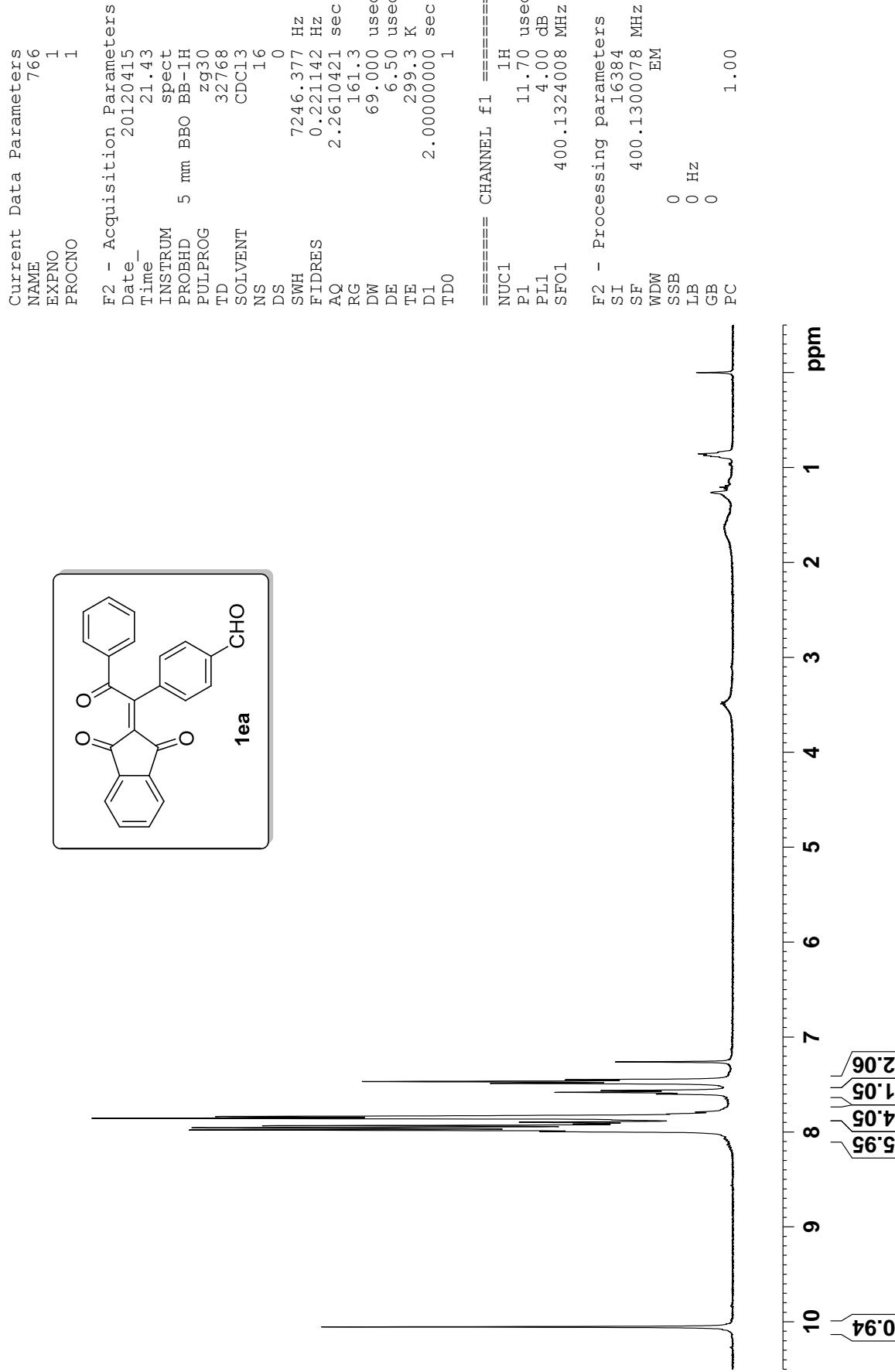


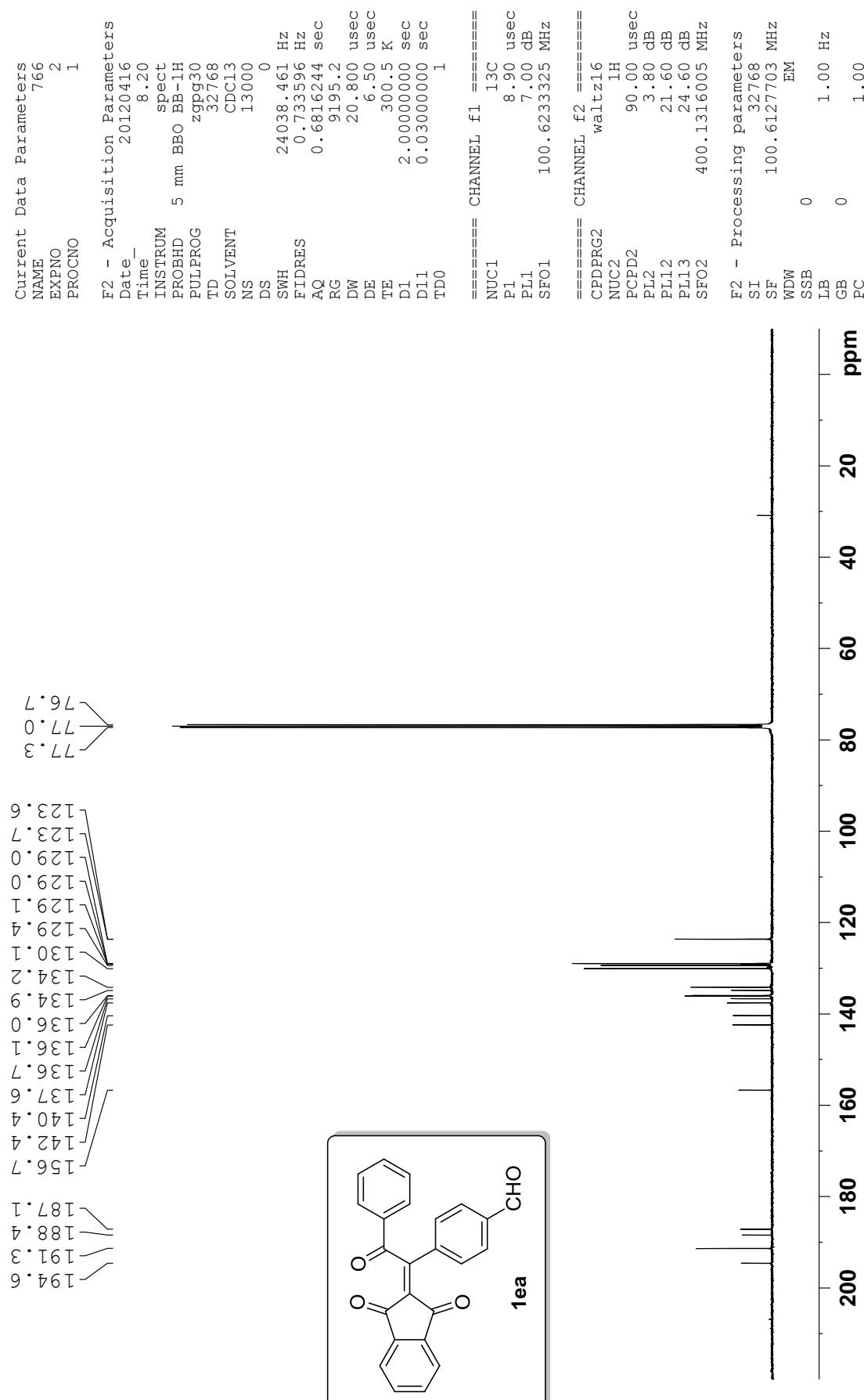


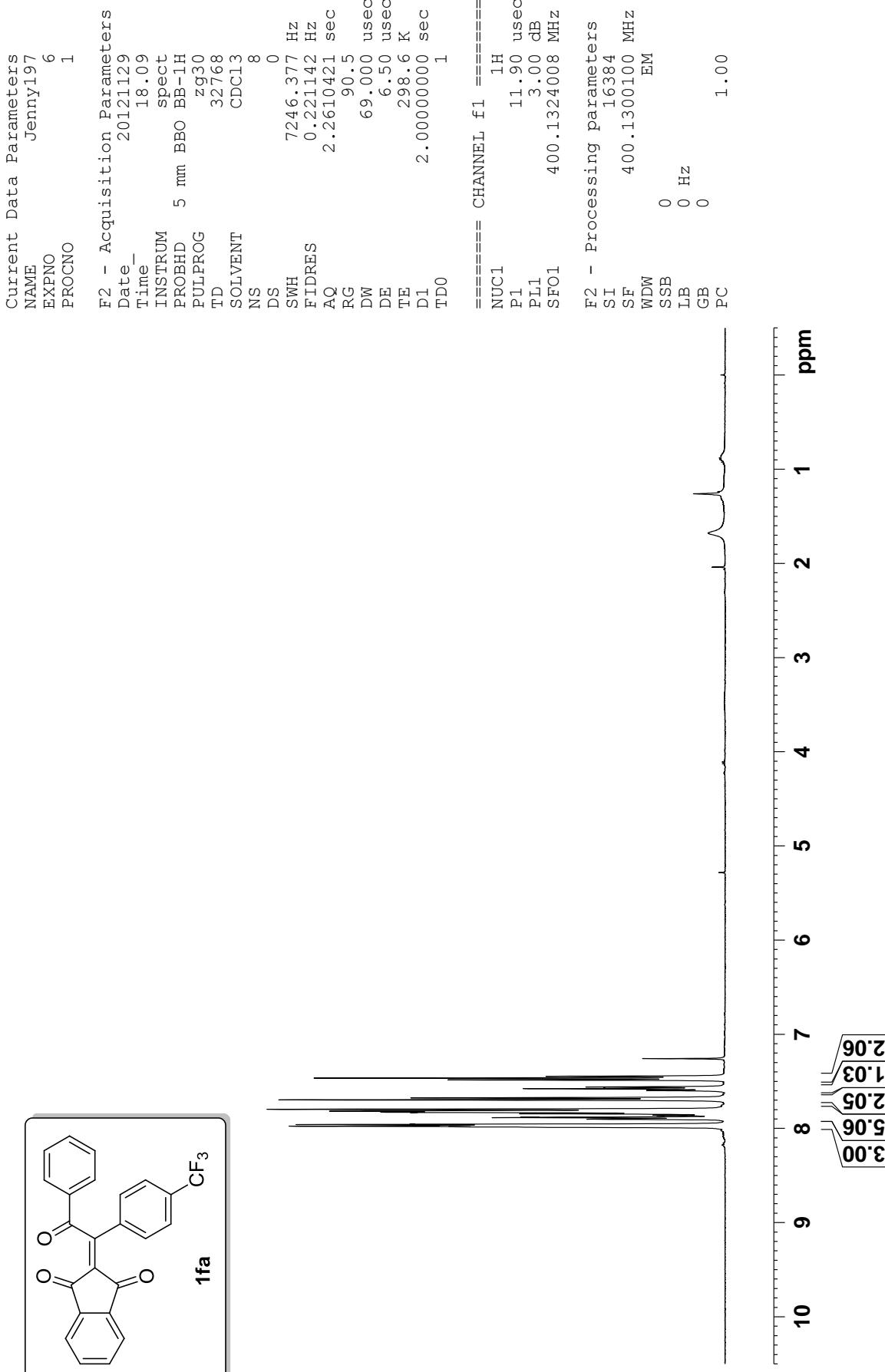


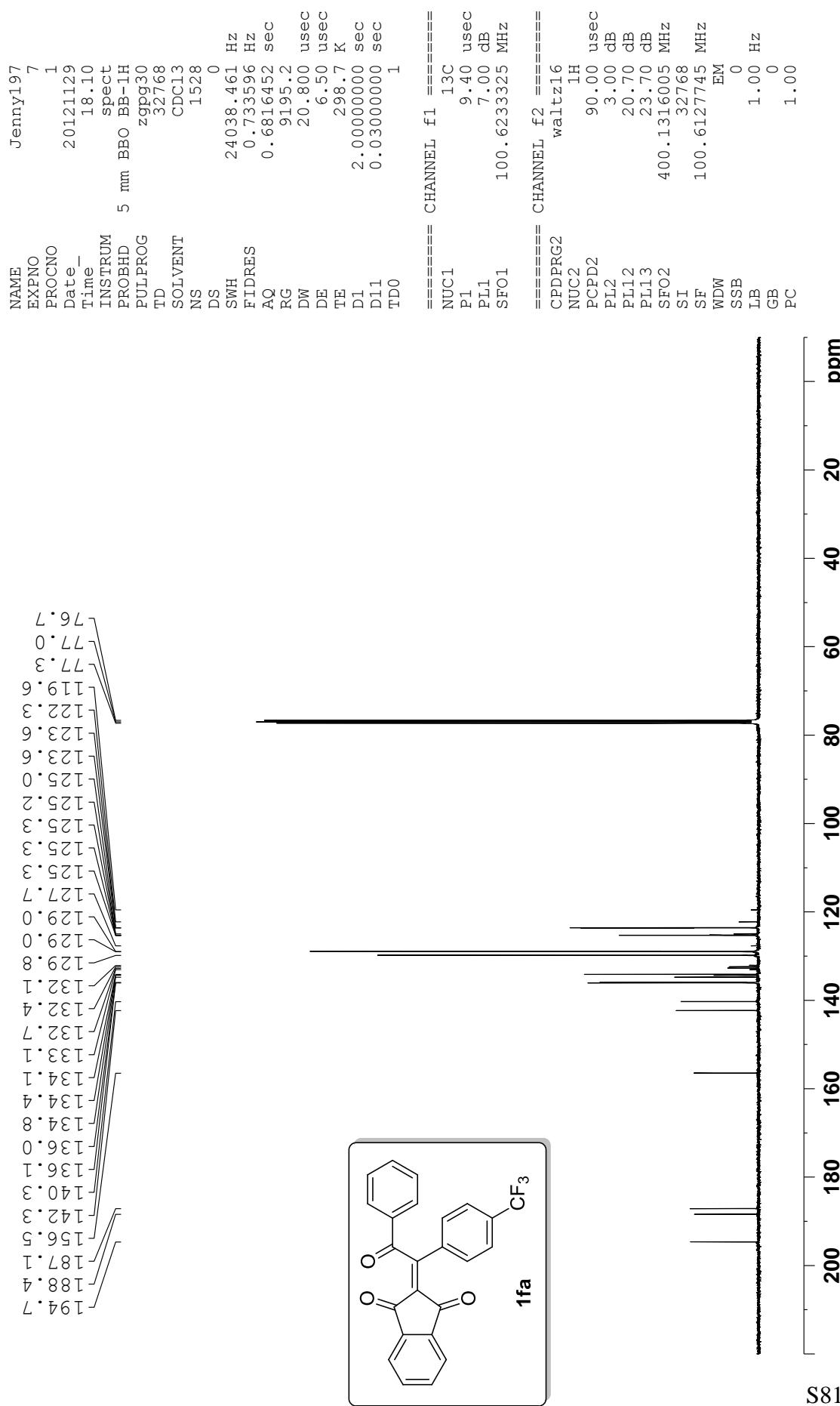


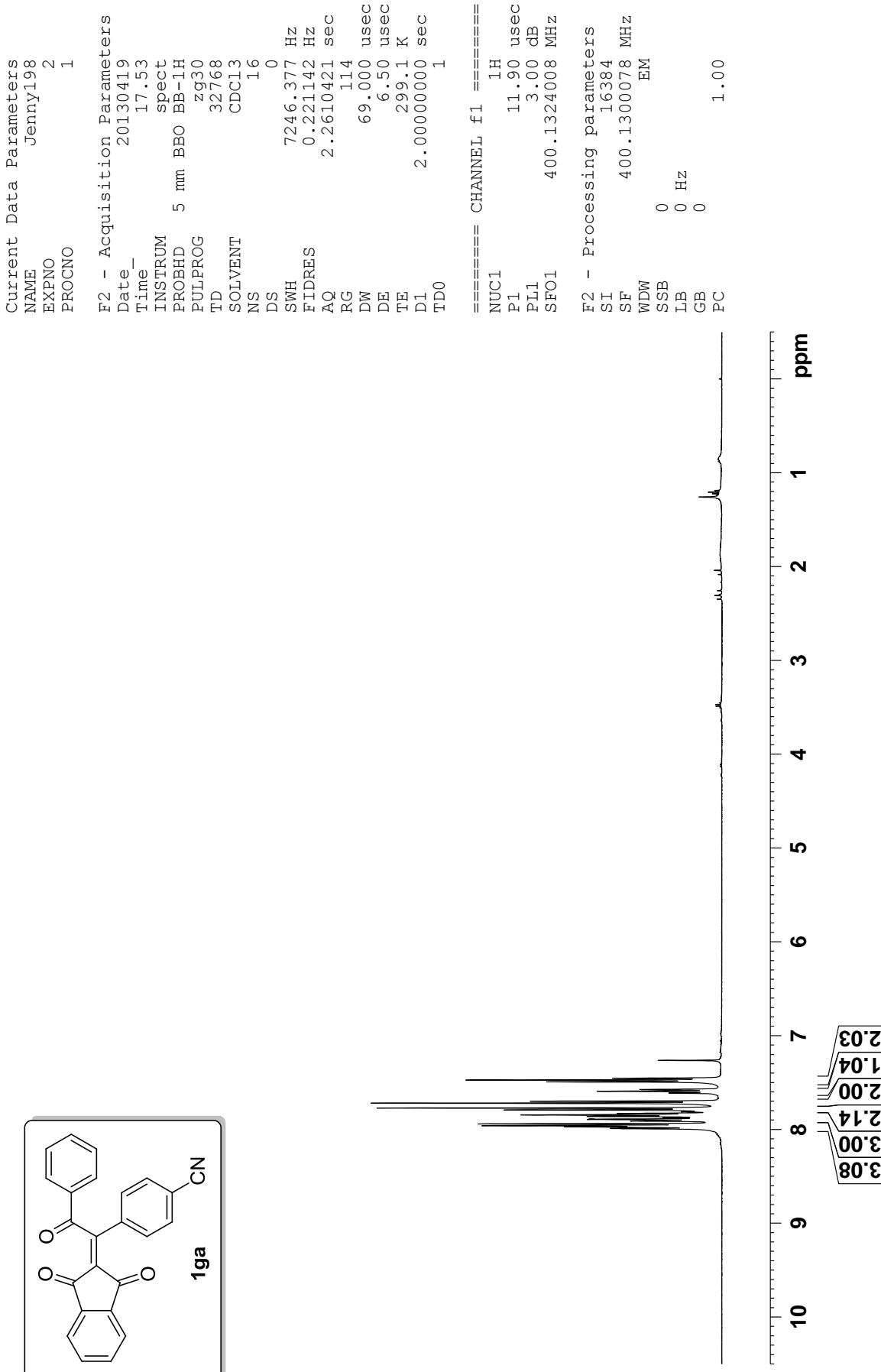


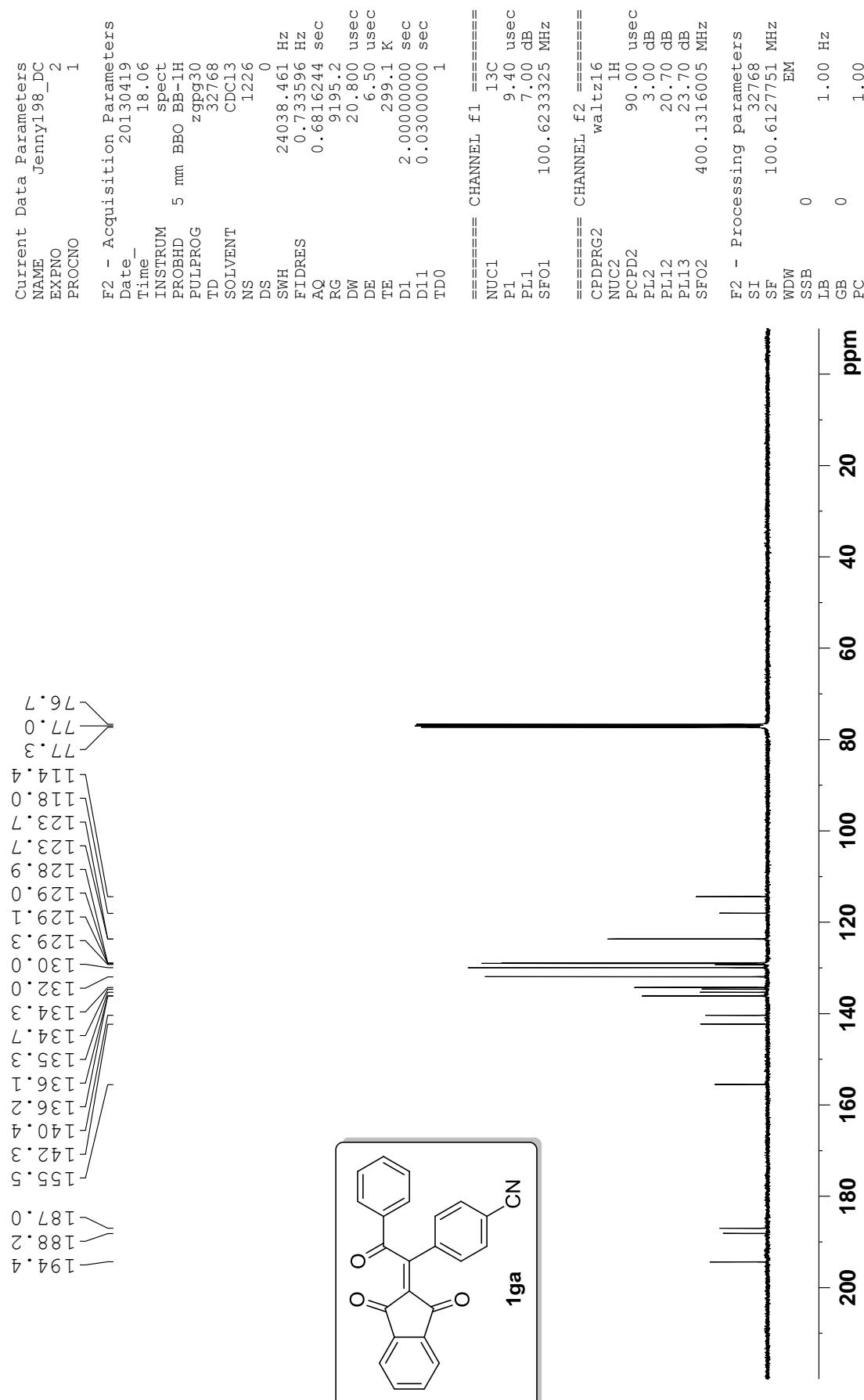


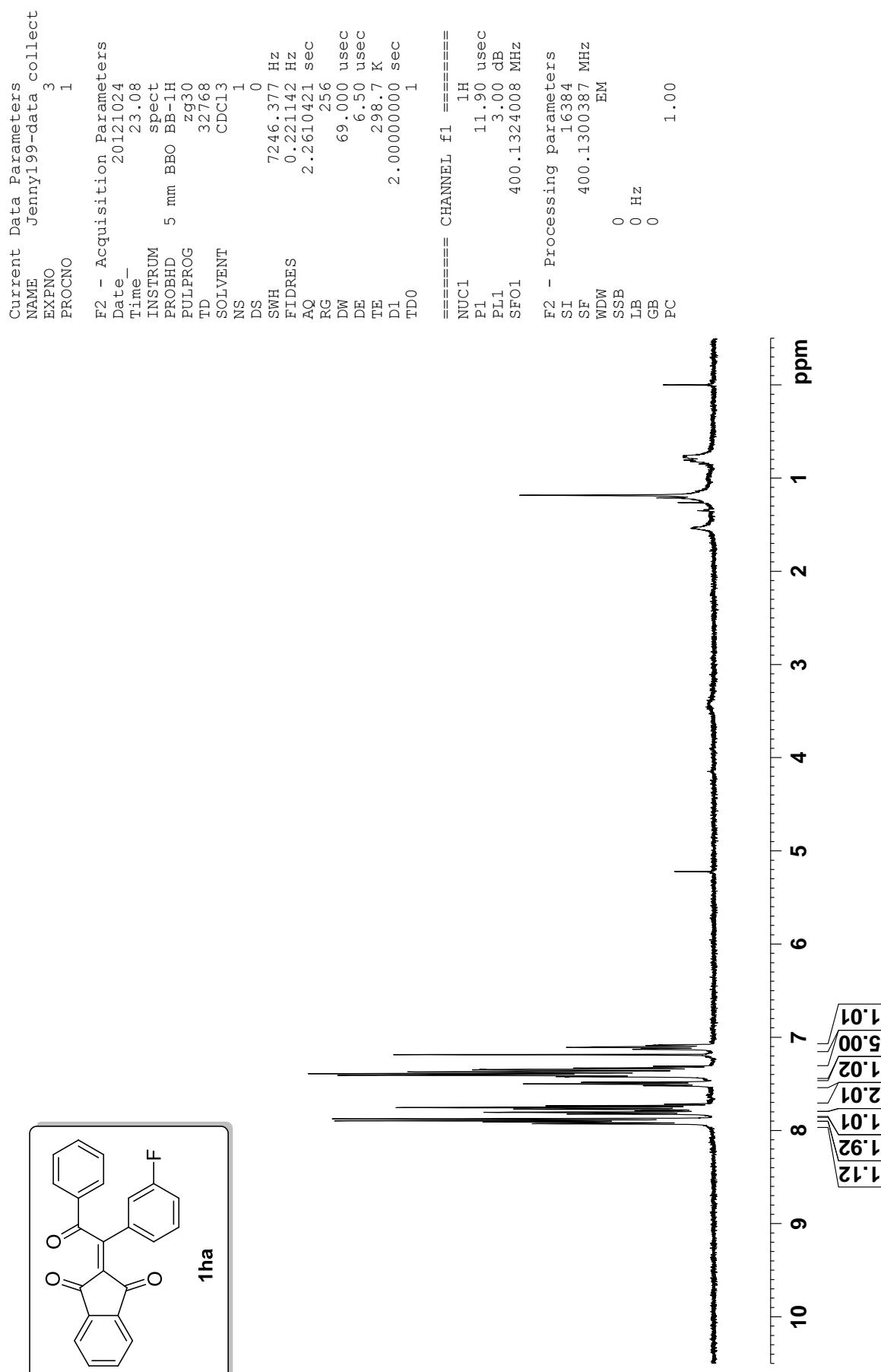


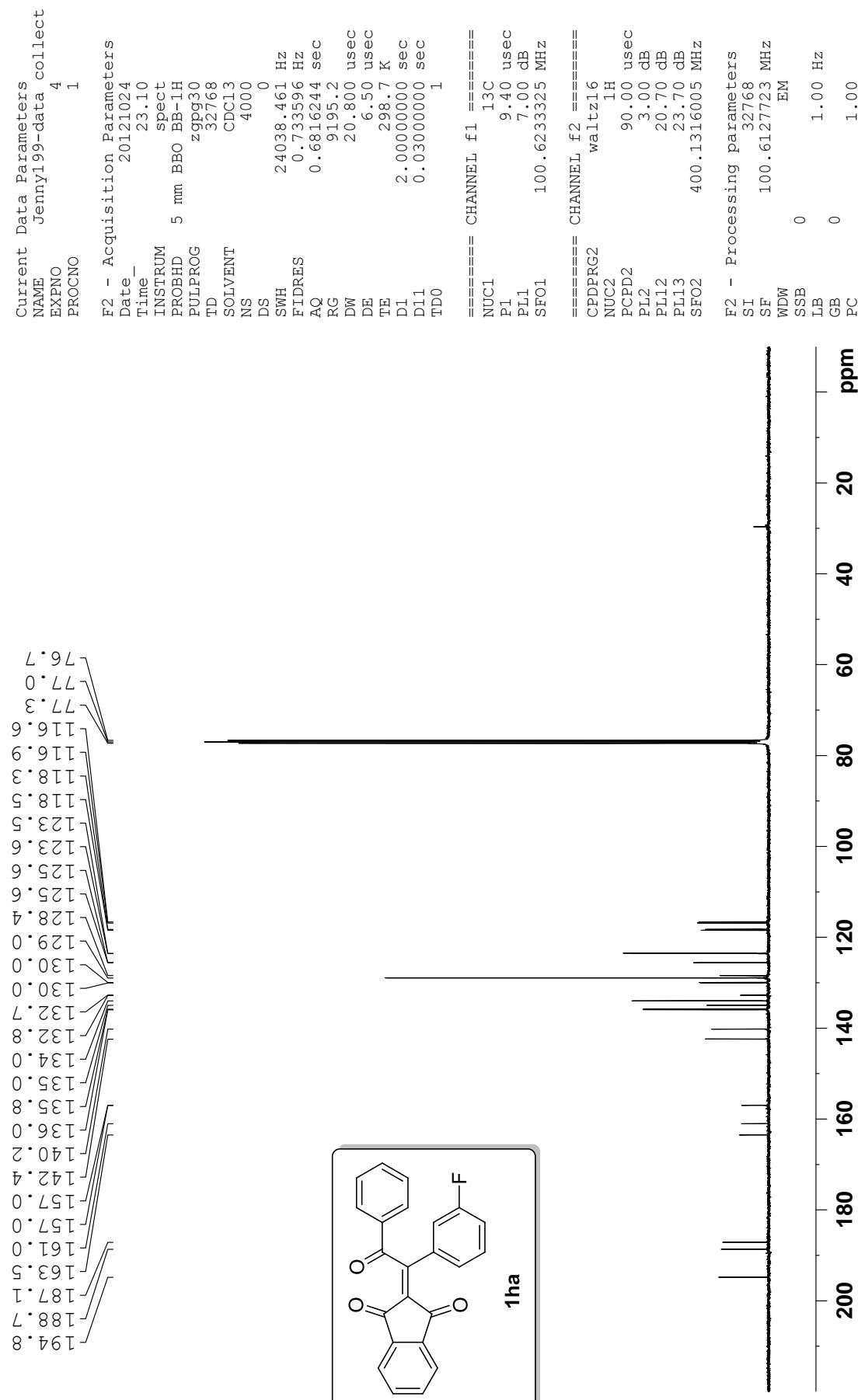


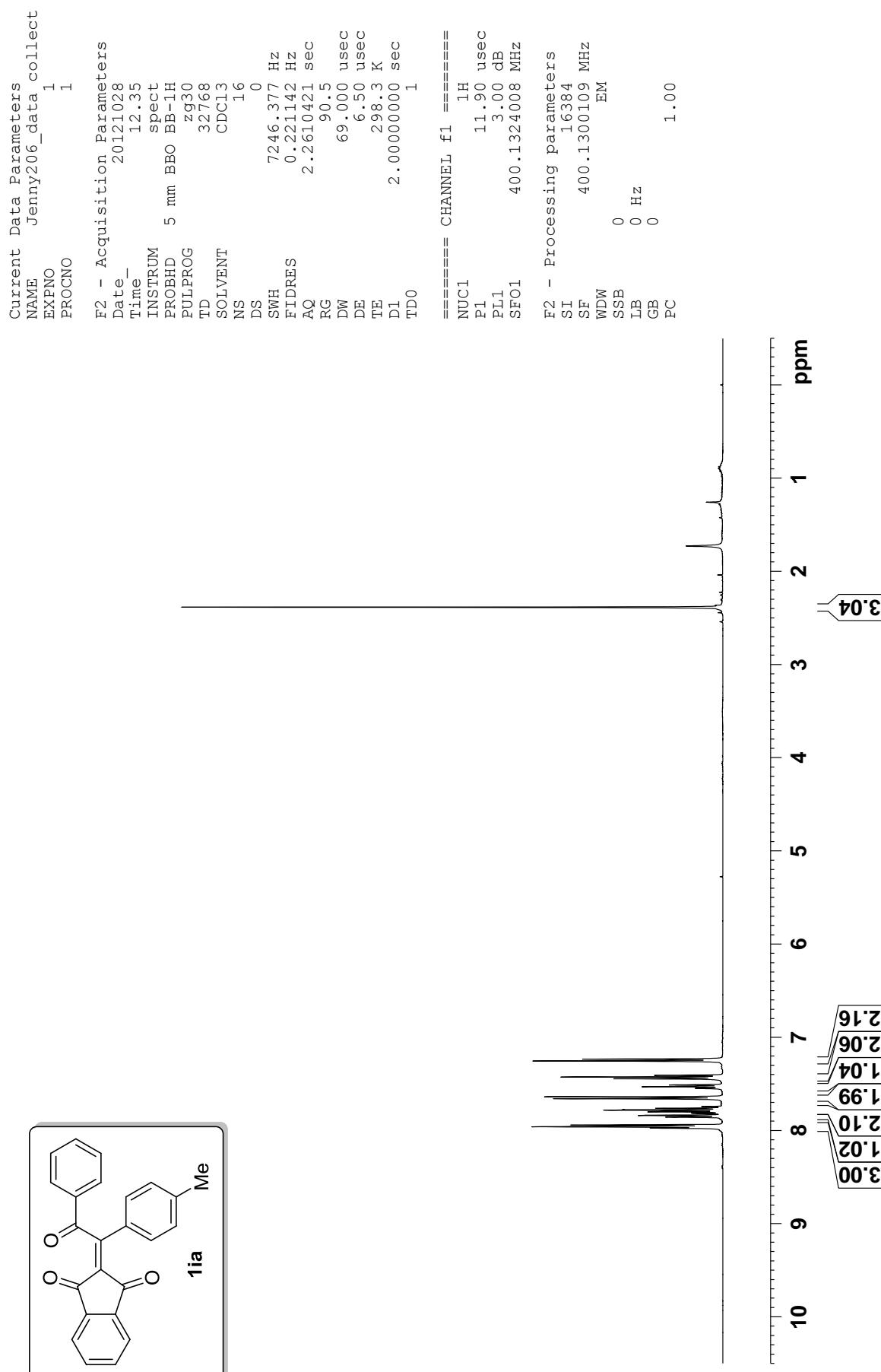


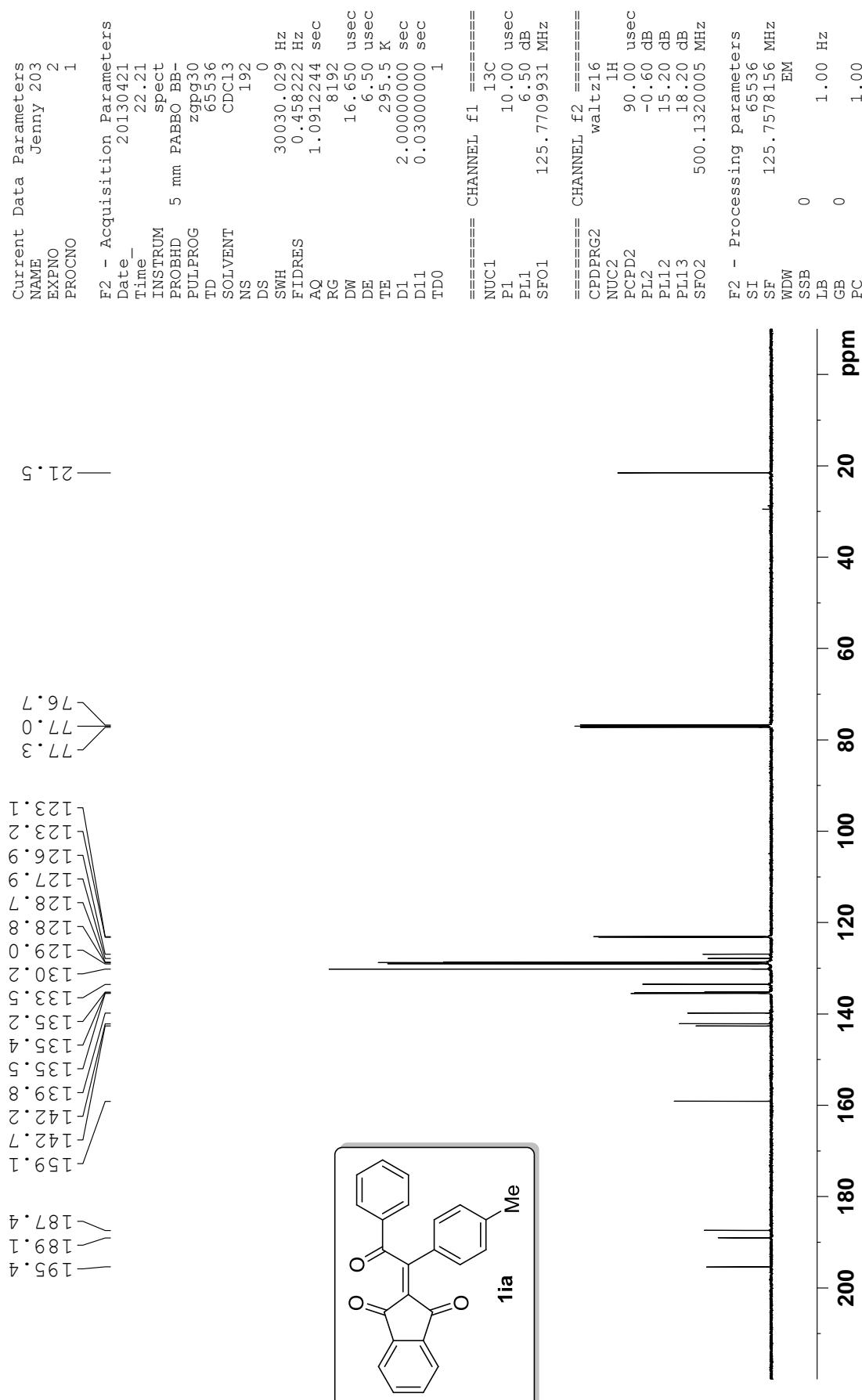


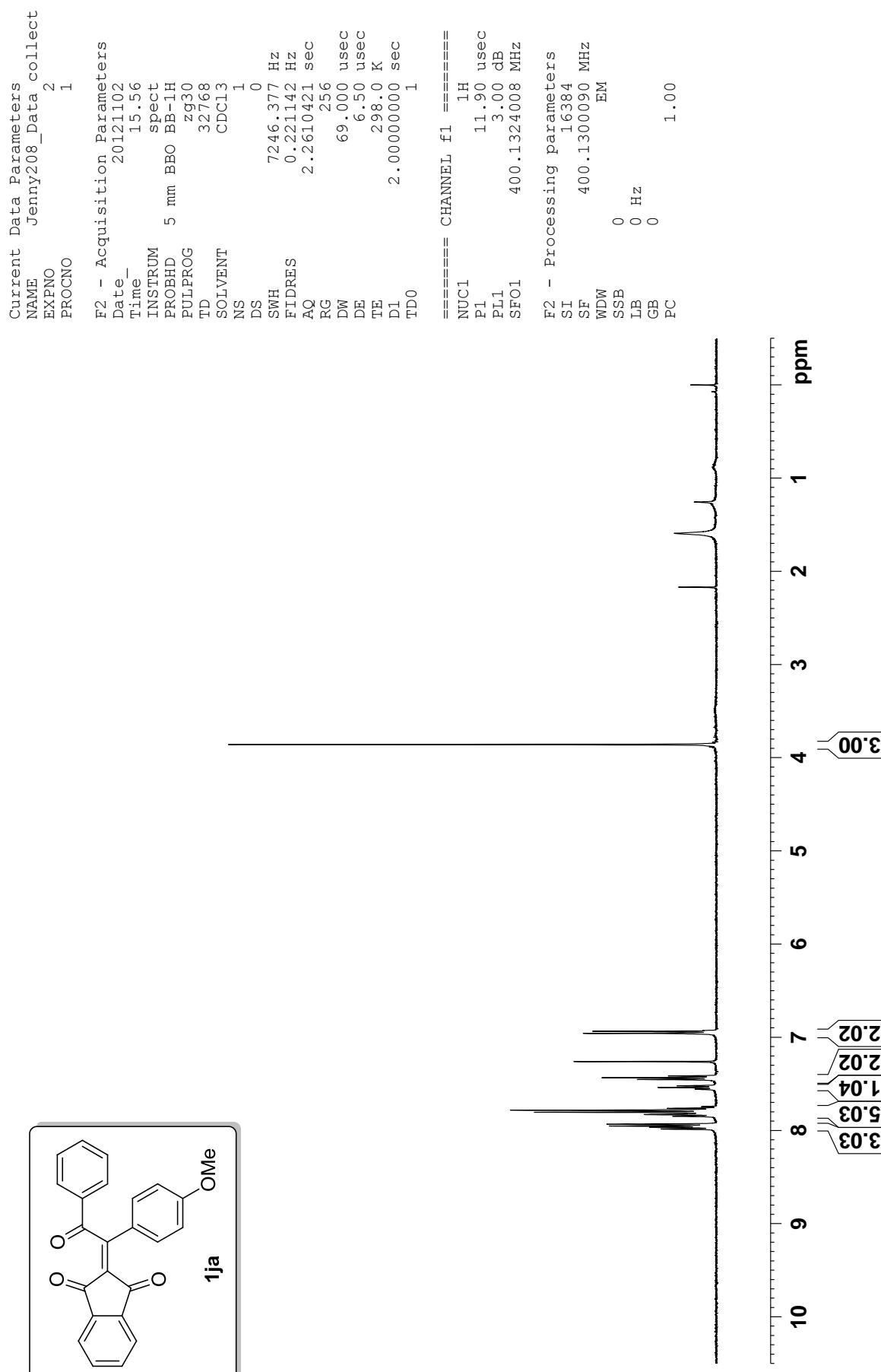


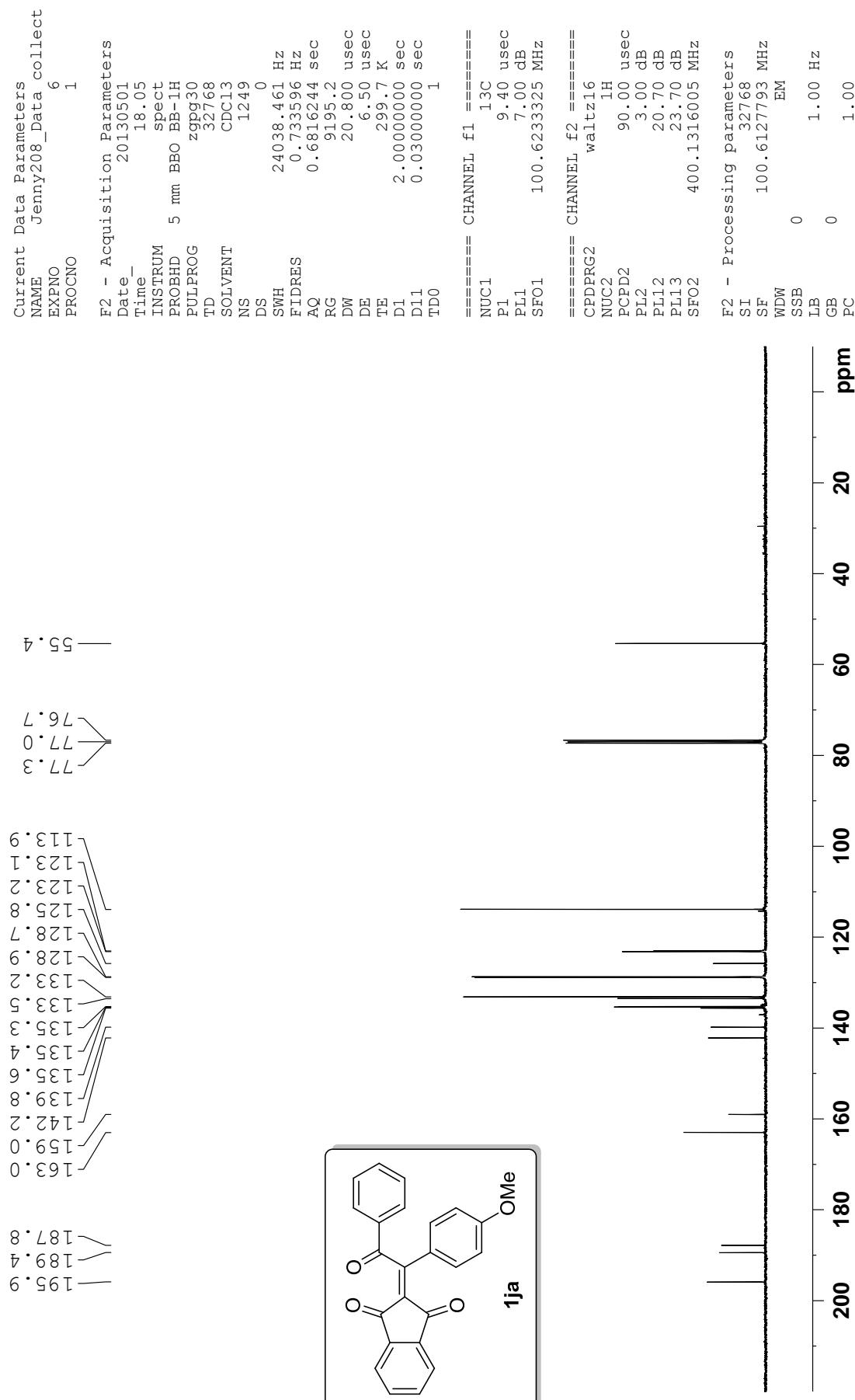


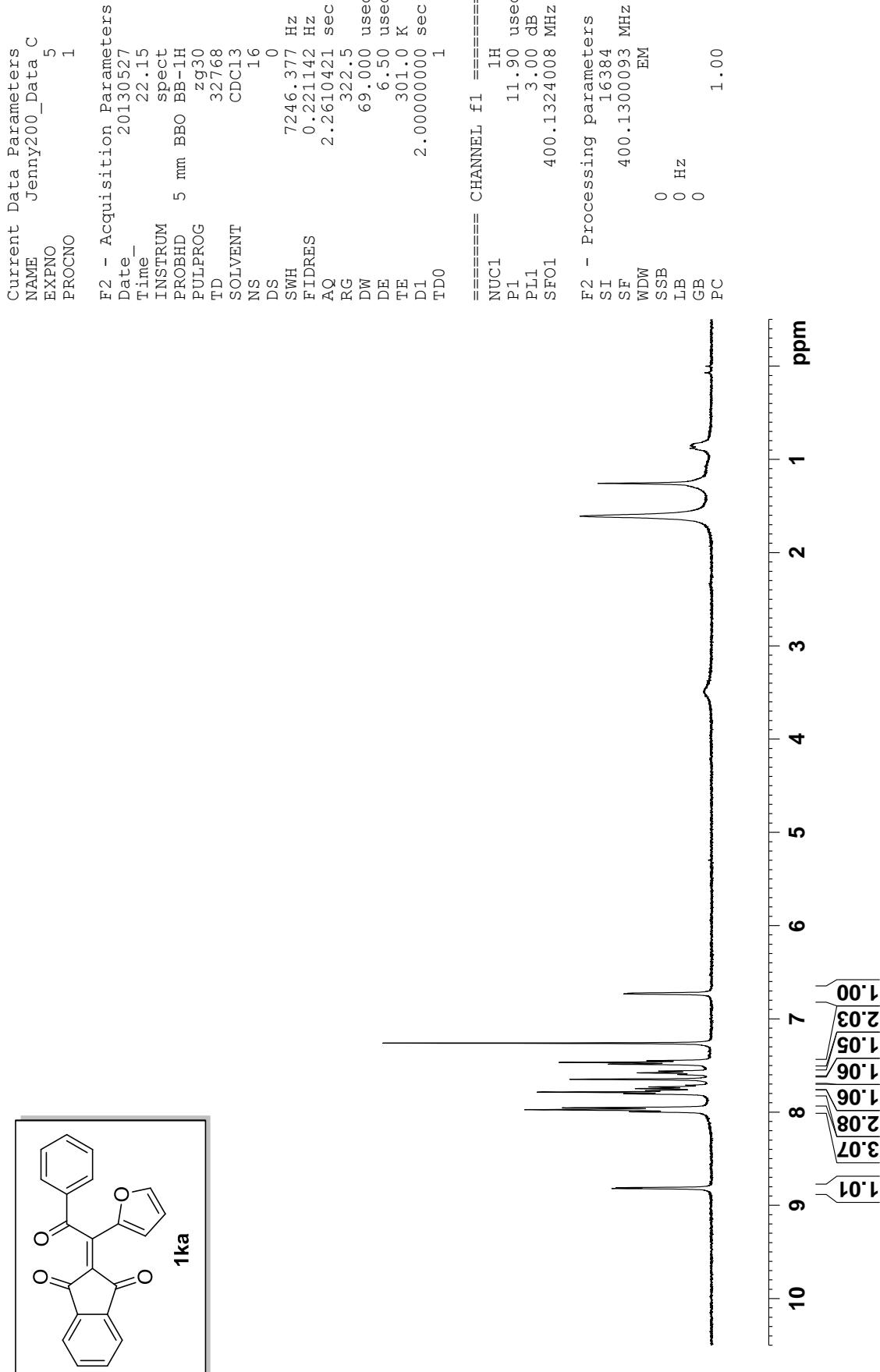


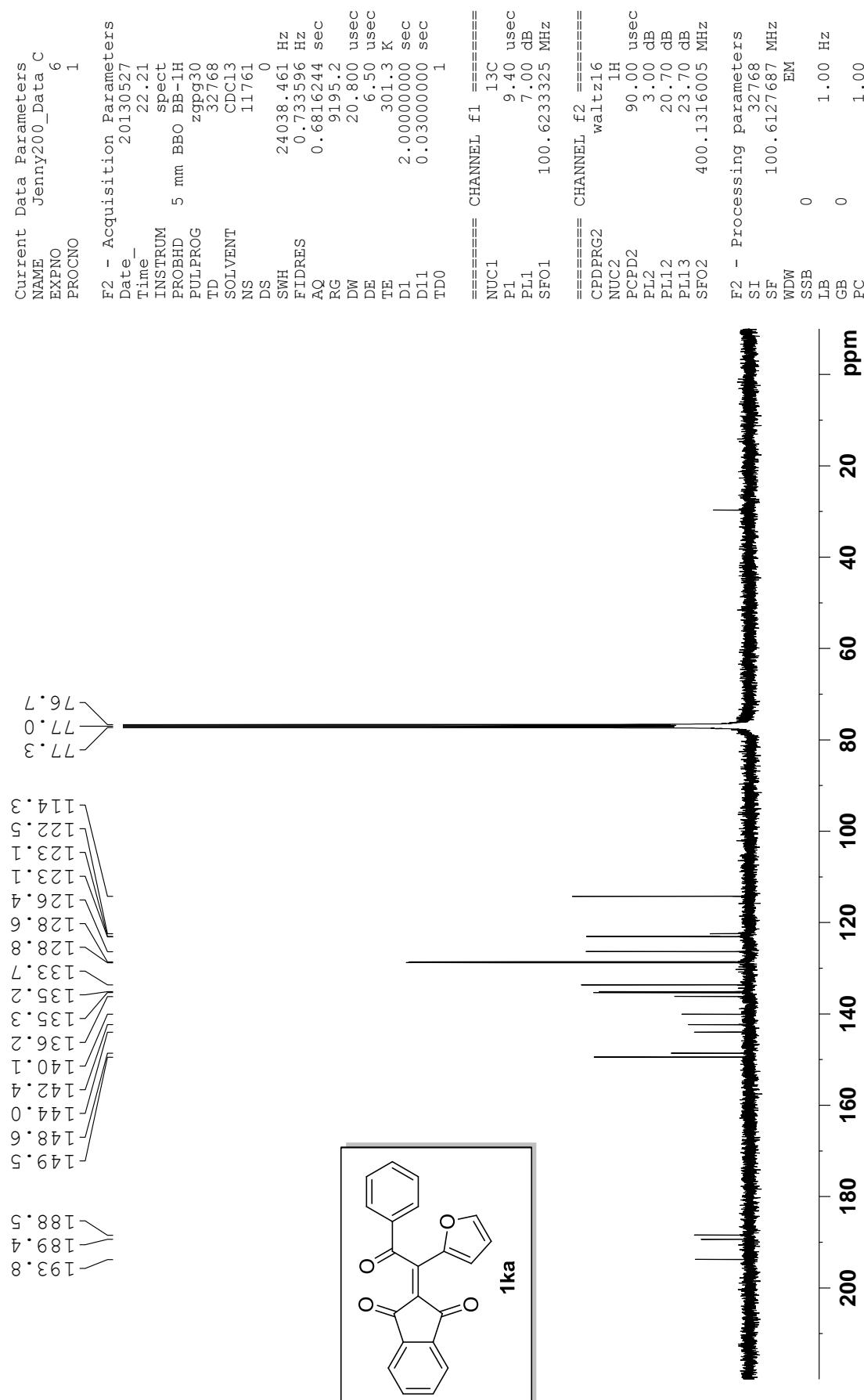


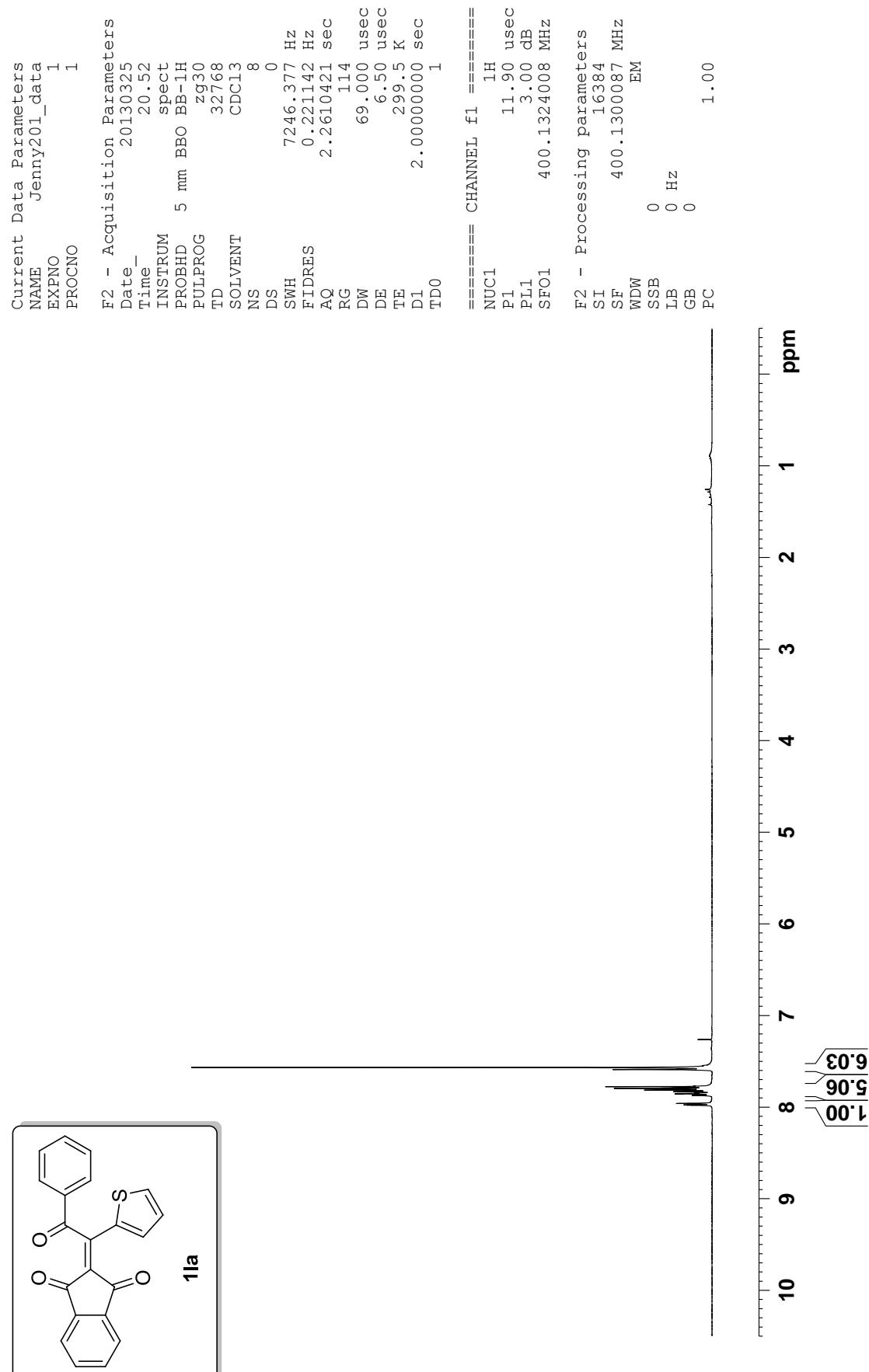


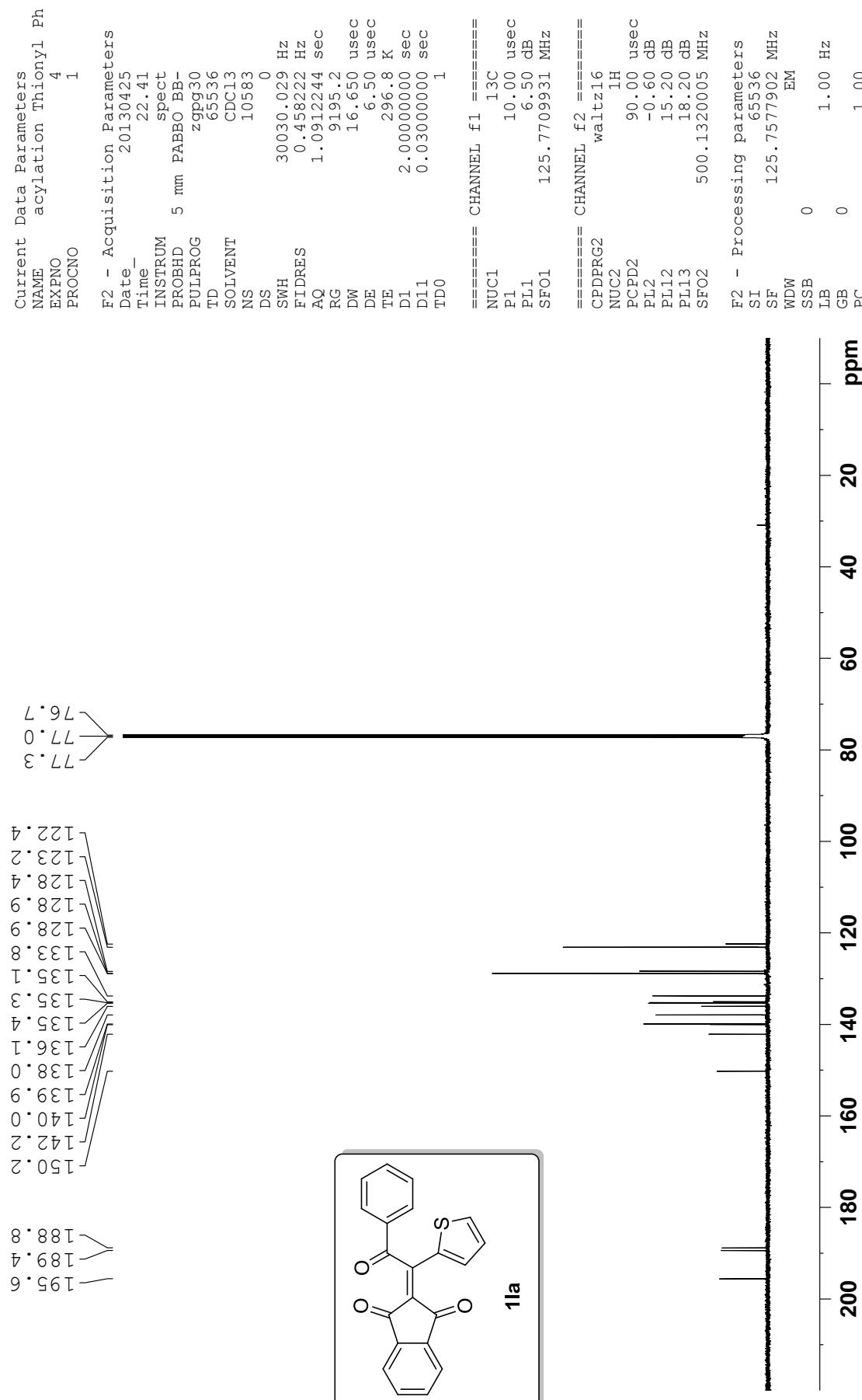












Current Data Parameters
NAME Jenny365_data collect
EXPNO 4
PROCNO 1

F2 - Acquisition Parameters

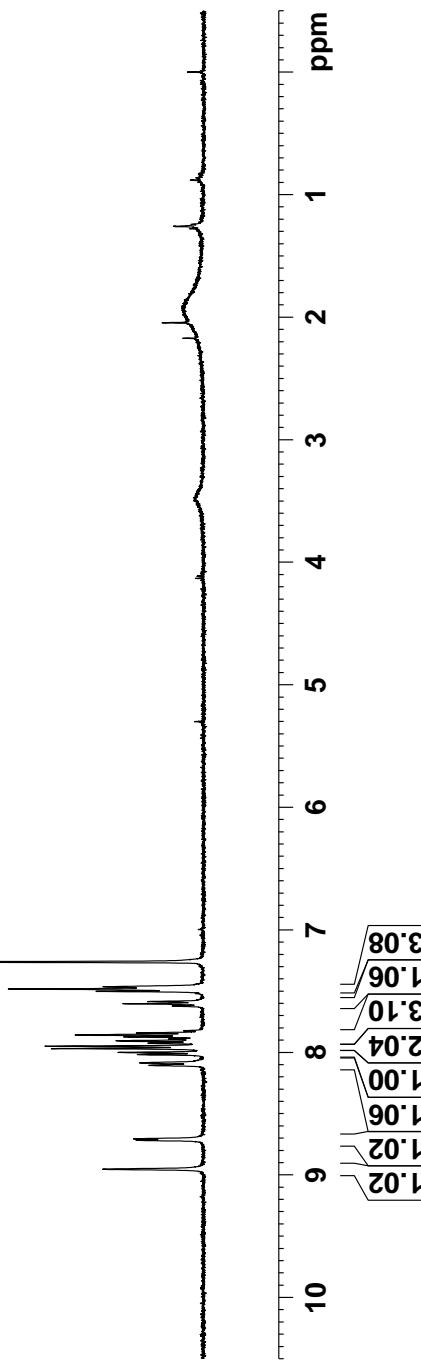
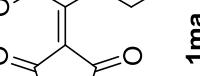
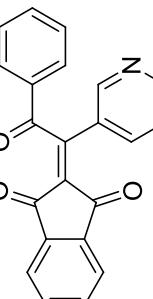
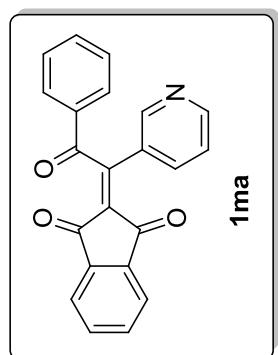
Date	20130222
Time	20.14
INSTRUM	BB-1H
PROBHD	5 mm BBO
PULPROG	Z930
TD	32768
SOLVENT	CDC13
NS	16
DS	0
SWH	7246.377 Hz
EIDRRES	0.2221142 sec
AQ	2.2610421 sec
RG	362
DW	69.0000 usec
DE	6.50 usec
TE	298.0 K
D1	2.00000000 sec
TDDO	1

```

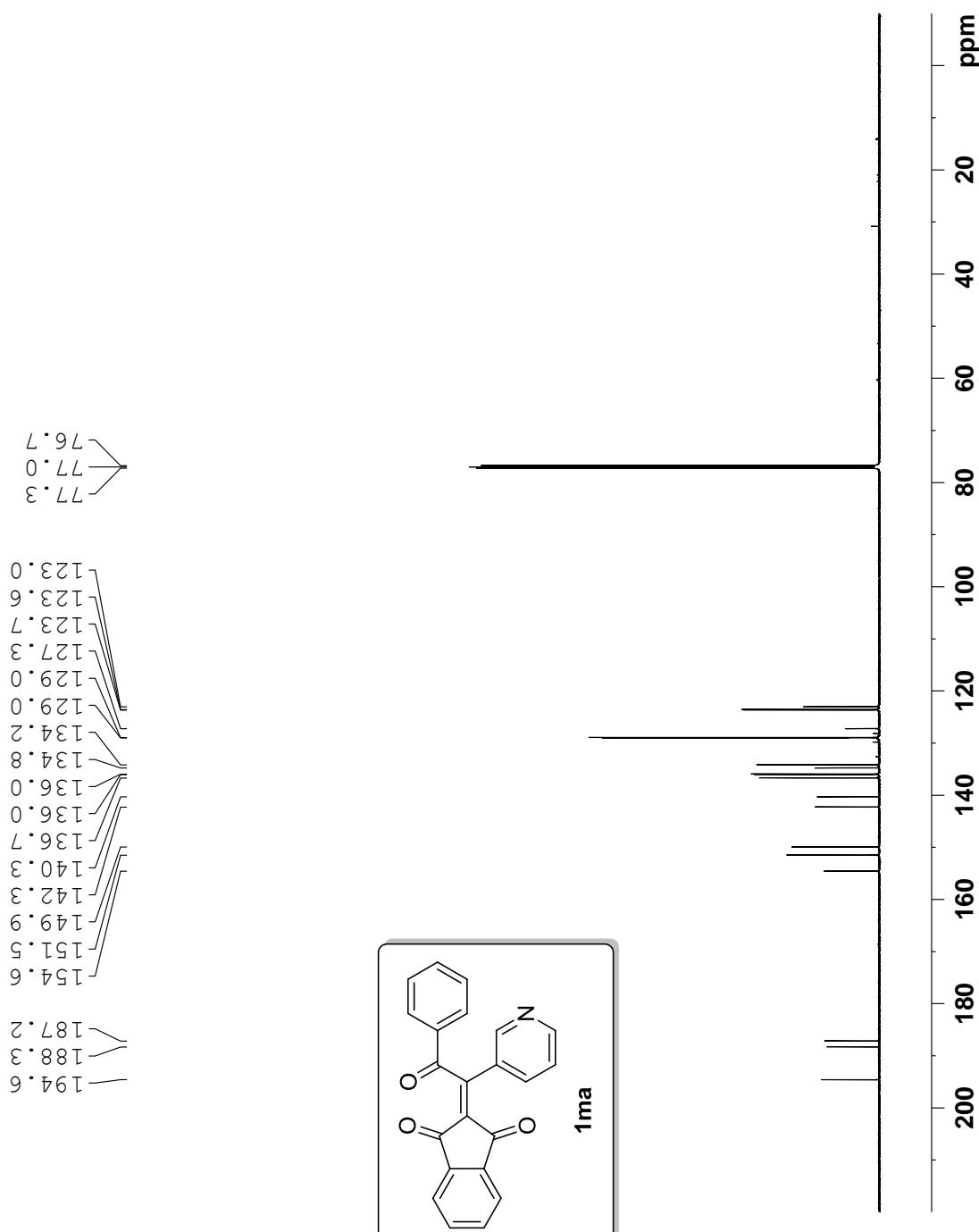
===== CHANNEL f1 =====
NUC1          1 H
P1            11.90 usec
PL1           3.00 dB
SF01          400.1324008 MHz
                           EM

F2 - Processing parameters
SI            16384
SF            400.1300091 MHz
WDW           0 Hz
SSB           0 GB
LB            0 PC
PC

```

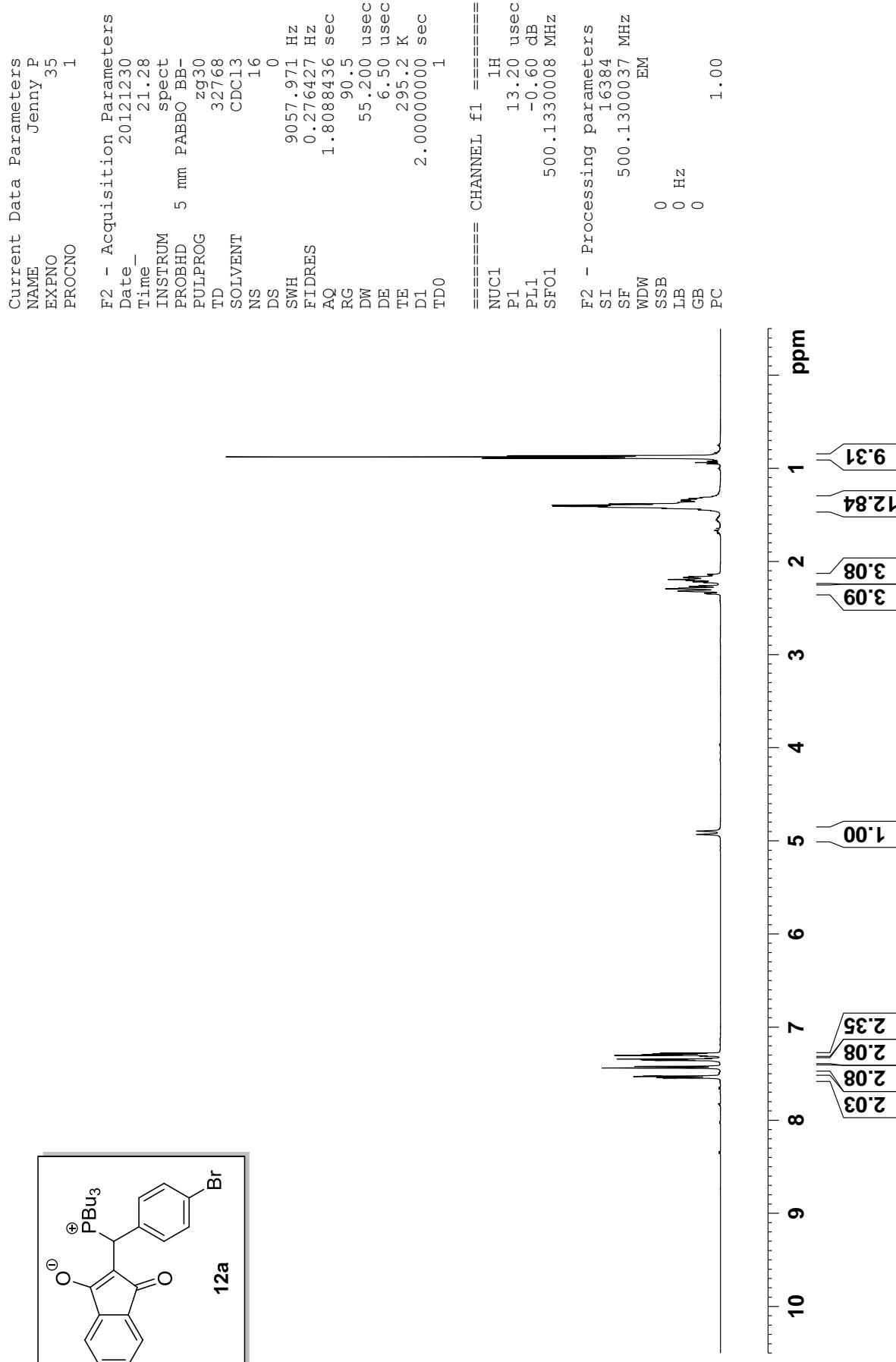


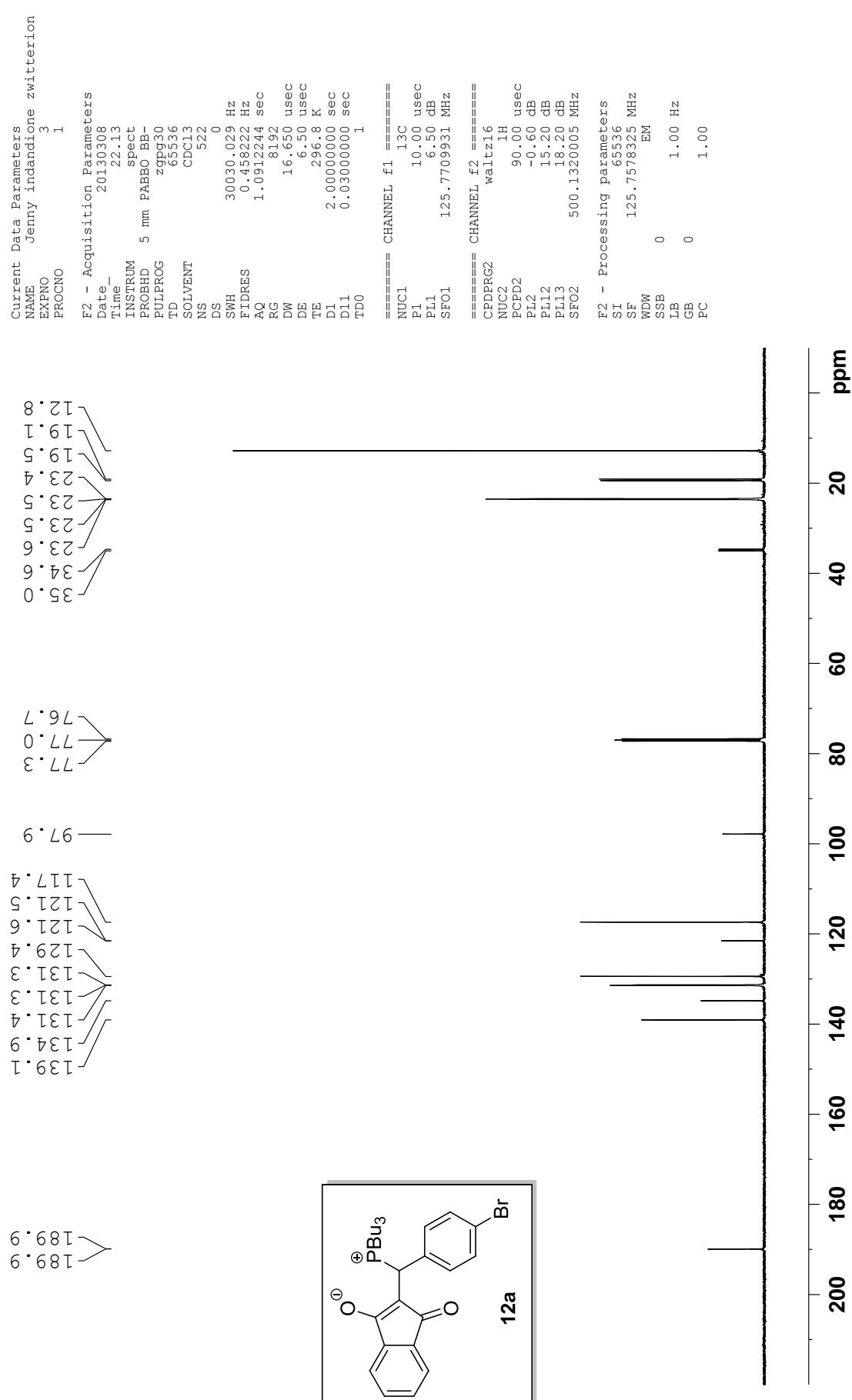
Current NAME	Data EXPNO	Parameters	Jenny
PROCNO			365
			2
F2 - Acquisition Parameters			
Date		20130501	
Time		18.15	
INSTRUM	5 mm PROBDH	PABB0 BB-spect	
PULPROG		zgpg30	
TD		65536	
SOLVENT	NS	CDC13	
DS		2000	
SWH		0	
FIDRES		0.029 Hz	
AQ		0.458222 Hz	
RG		1.091244 sec	
DW		9195.2	
DE		16.650 usec	
TE		6.50 usec	
D1		296.9 K	
D11		2.00000000 sec	
TDO		0.03000000 sec	
		1	
===== CHANNEL f1 =====			
NUC1		13C	
P1		10.00 usec	
PL1		6.50 dB	
SFO1		125.7709931 MHz	
===== CHANNEL f2 =====			
CPDPGR2		waltz16	
NUC2		1H	
PCPFD2		90.00 usec	
PL2		-0.60 dB	
PL12		15.20 dB	
PL13		18.20 dB	
SFO2		500.1320005 MHz	
===== Processing parameters =====			
SI		65536	
SF		125.7577968 MHz	
WDW		EM	
SSB	0	0	1.00 Hz
LB	0	0	1.00 Hz
PC			

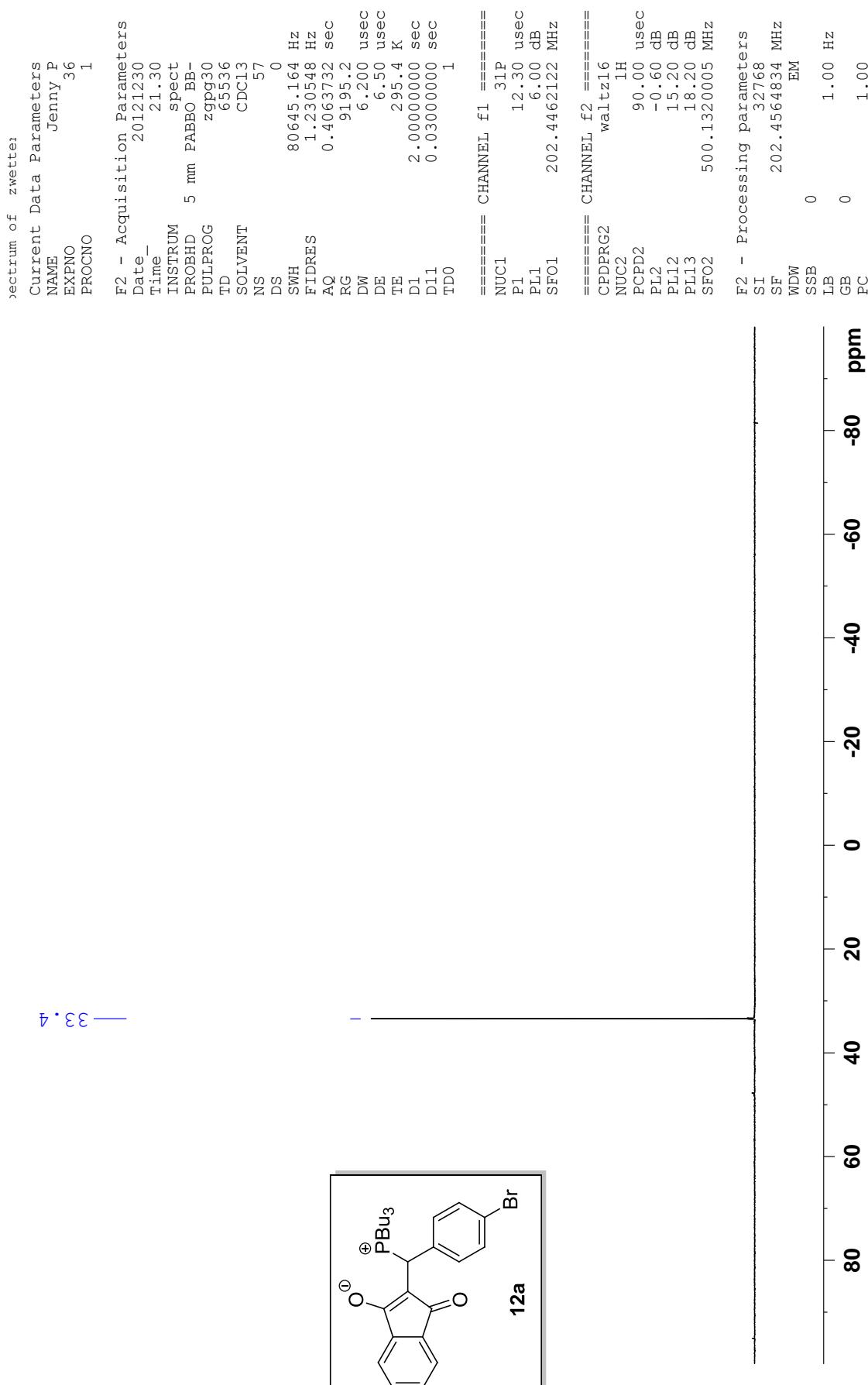


1ma

The chemical structure of compound 1ma is shown as a substituted chromene derivative. It features a central five-membered ring fused to a six-membered ring. The six-membered ring has carbonyl groups at positions 2 and 6, and a phenyl group attached to the 4-position. A pyridine ring is attached to the 6-position of the chromene ring.







Current Data Parameters
NAME Jenny245_Data collect 1a
EXPNO 1
PROCNO 1

F2 - Acquisition Parameters

Date 2013/5/07

Time 18:14

INSTRUM spect

PROBHD 5 mm BBO BB-1H

PULPROG z930

TD 32768

SOLVENT CDCl₃

NS 4

DS 0

SWH 7246.377 Hz

FIDRES 0.221142 Hz

AQ 2.2610421 sec

RG 256

DW 69.000 usec

DE 6.50 usec

DE 298.9 K

TE 2.0000000 sec

D1 1

TD0

===== CHANNEL f1 =====

NUC1 1H

P1 11.90 usec

PL1 3.00 dB

SF01 400.1324008 MHz

F2 - Processing parameters

SI 16384

SF 400.1300074 MHz

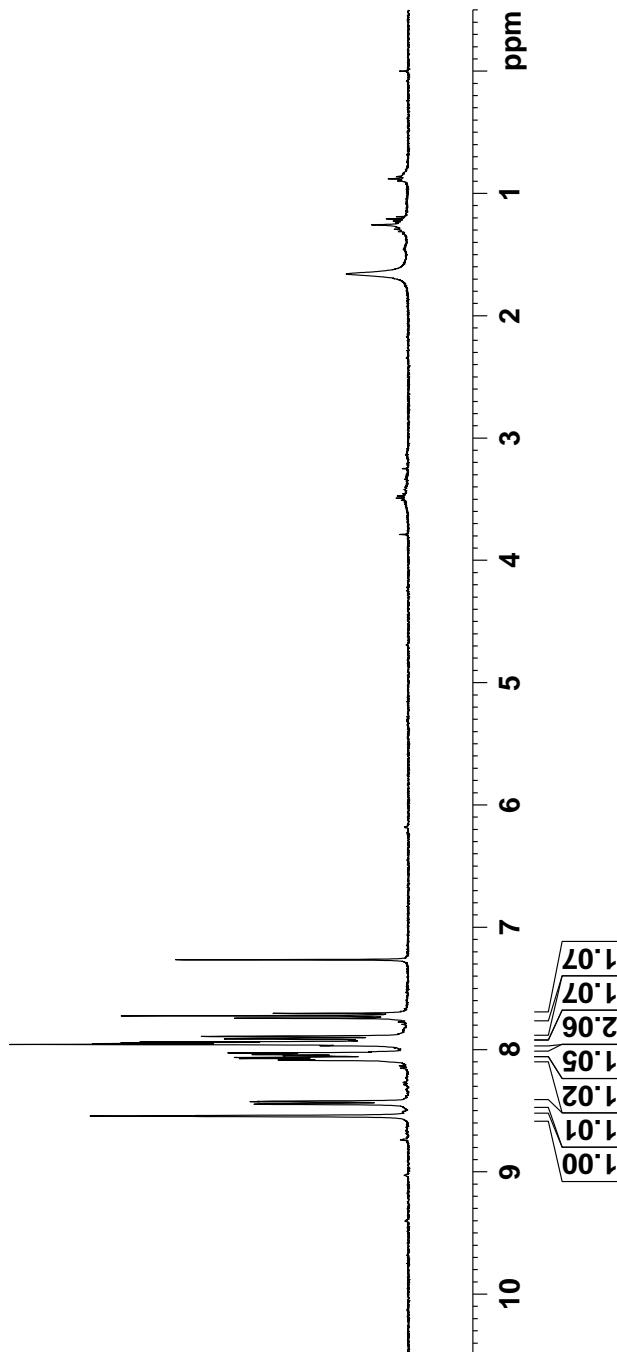
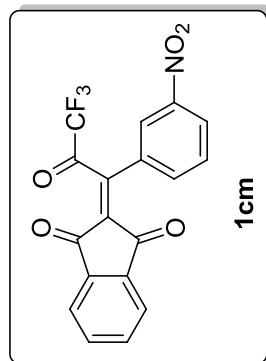
WDW EM

SSB 0

LB 0 Hz

GB 0

PC 1.00



```
Current Data Parameters
      NAME        Jenny245_Data_collect.la
      EXPNO       2
      PROCNO     1
```

FE2 - Acquisition Parameters
 Date 201507
 Time 18.16
 INSTRUM spect
 PROBHD 5 mm BBO BB-1H
 PULPROG zprg30
 TD 32768
 CPC13
 SOLVENT T211
 NS 0
 DS 24038.461 Hz
 SWH 0.73595 Hz
 FIDRES 0.6816244 sec
 AQ 9.15.2
 RG 20.800 usec
 DW 6.50 usec
 DE 239.0 K
 TE 2.0000000 sec
 D1 0.0300000 sec
 TDO 1

```

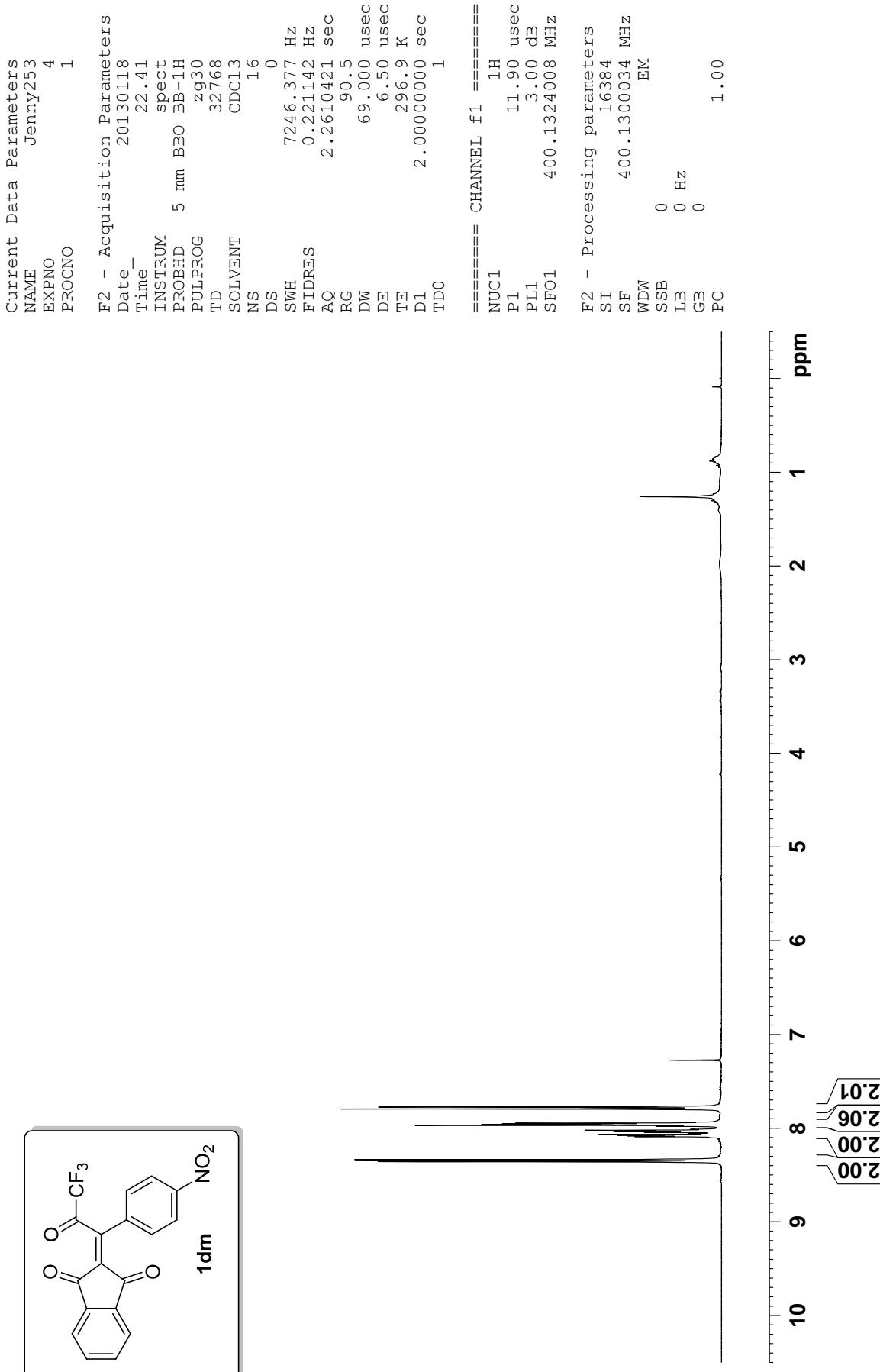
===== CHANNEL f1 =====
NUC1          13C
P1            9.40 usec
PL1           7.00 dB
SF01          100.6233325 MHz

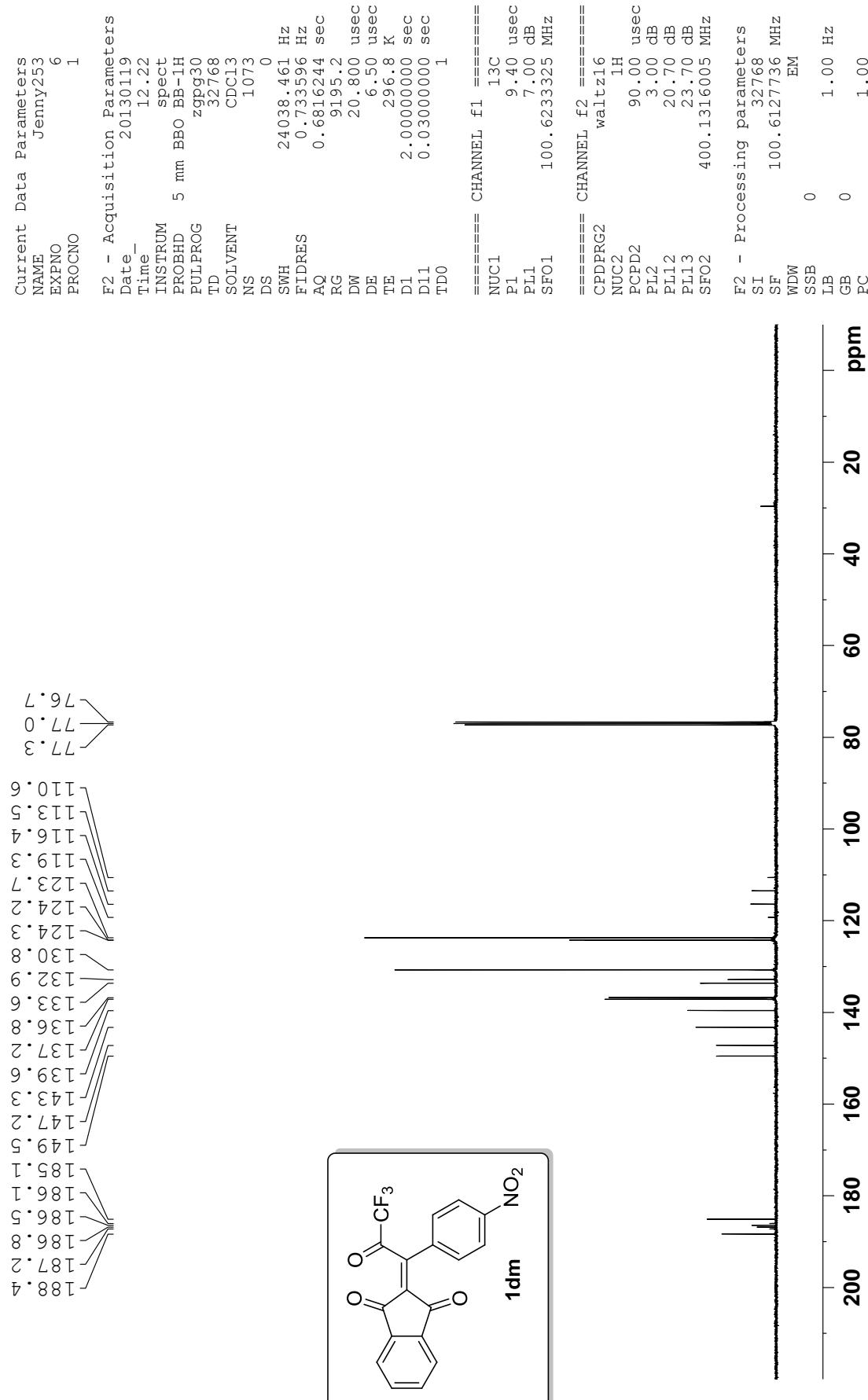
===== CHANNEL f2 =====
waltz16
NUC2          1H
PCPD2         90.00 usec
PL2           3.00 dB
PL112         20.70 dB
PL113         23.70 dB
SF02          400.1316005 MHz

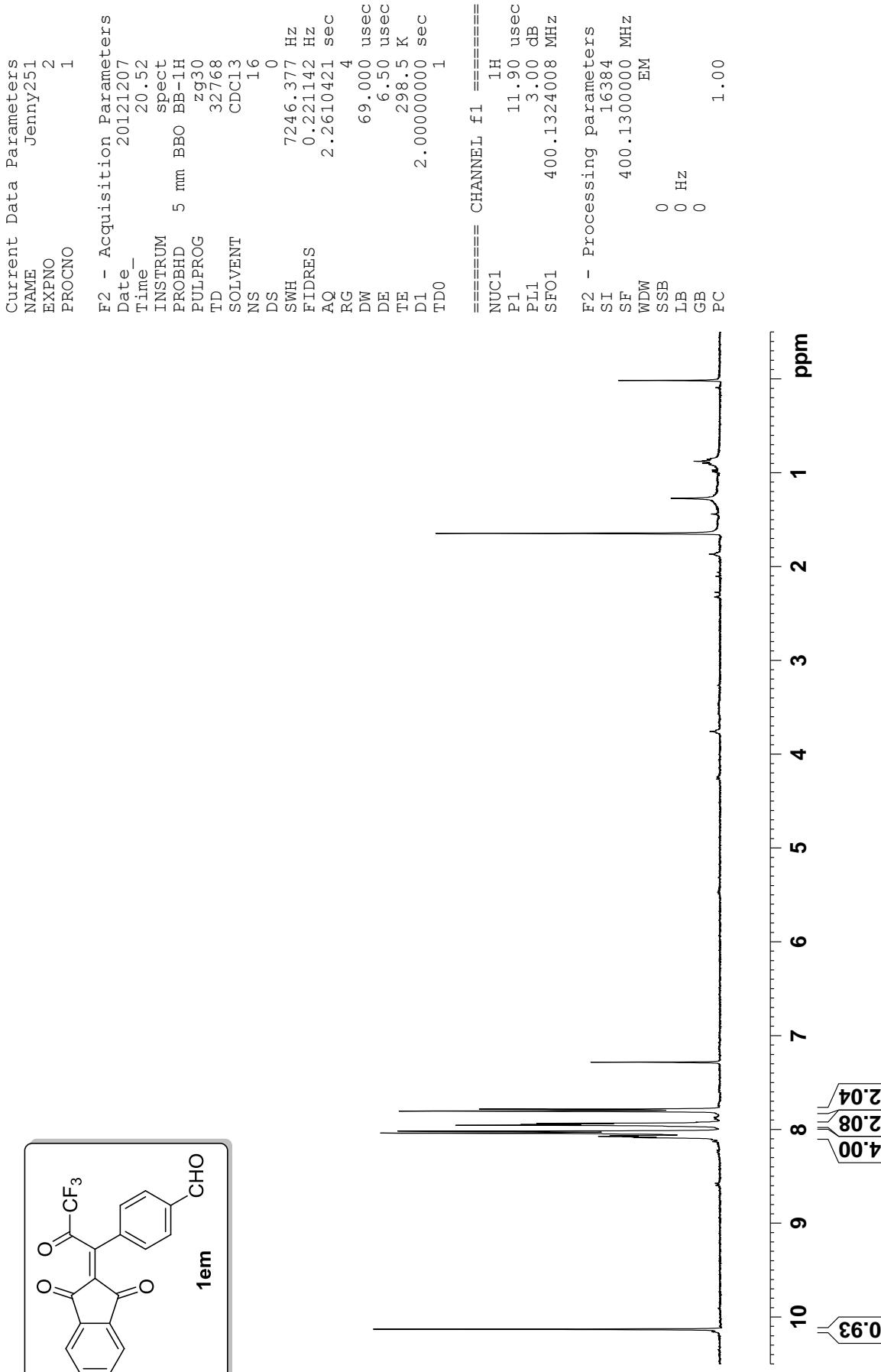
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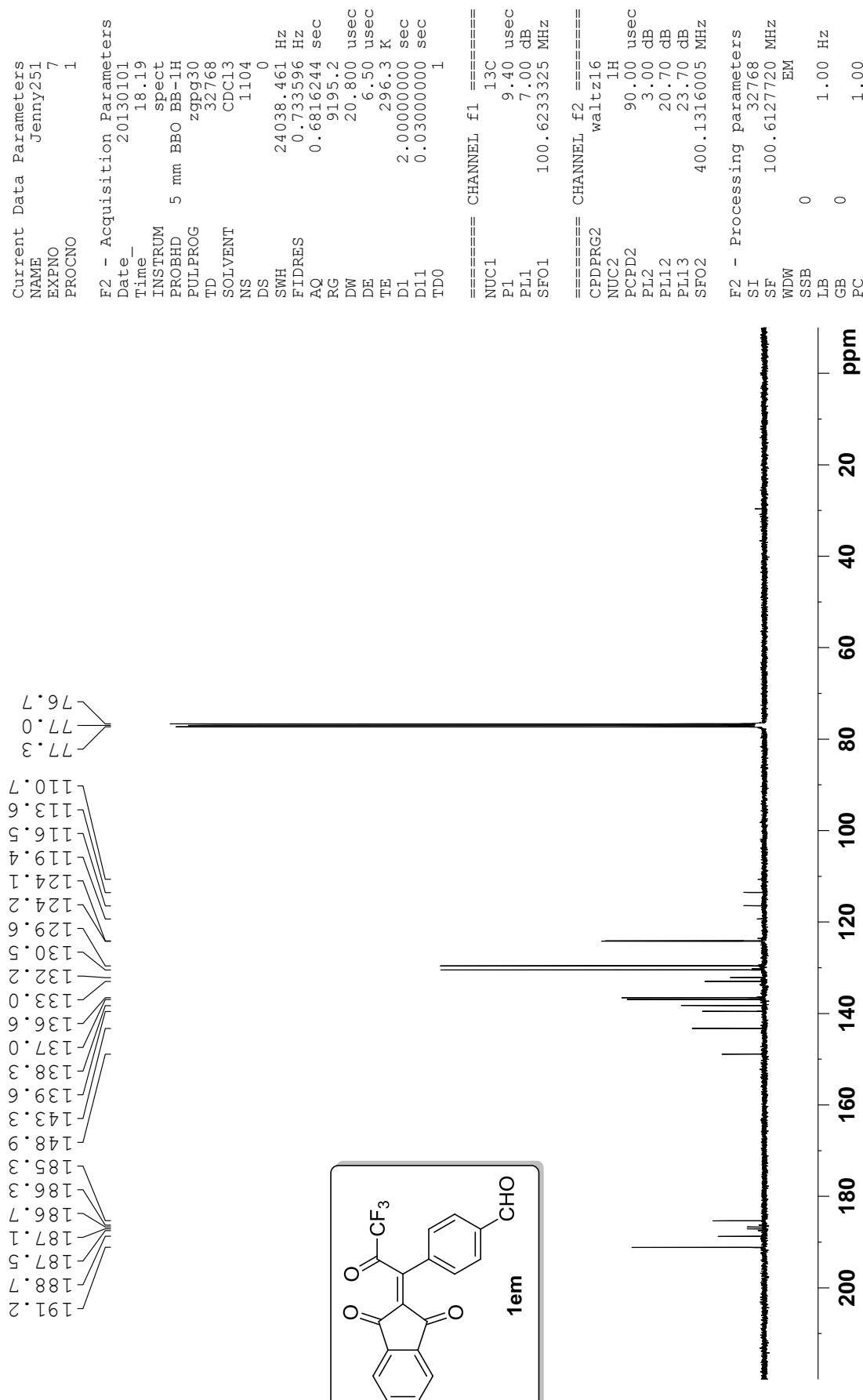
F2 - Processing parameters
 SI 32768
 SF 100.6127701 MHZ
 WDW EM
 SSB 0 1.00 Hz
 LB 0
 GB
 PC

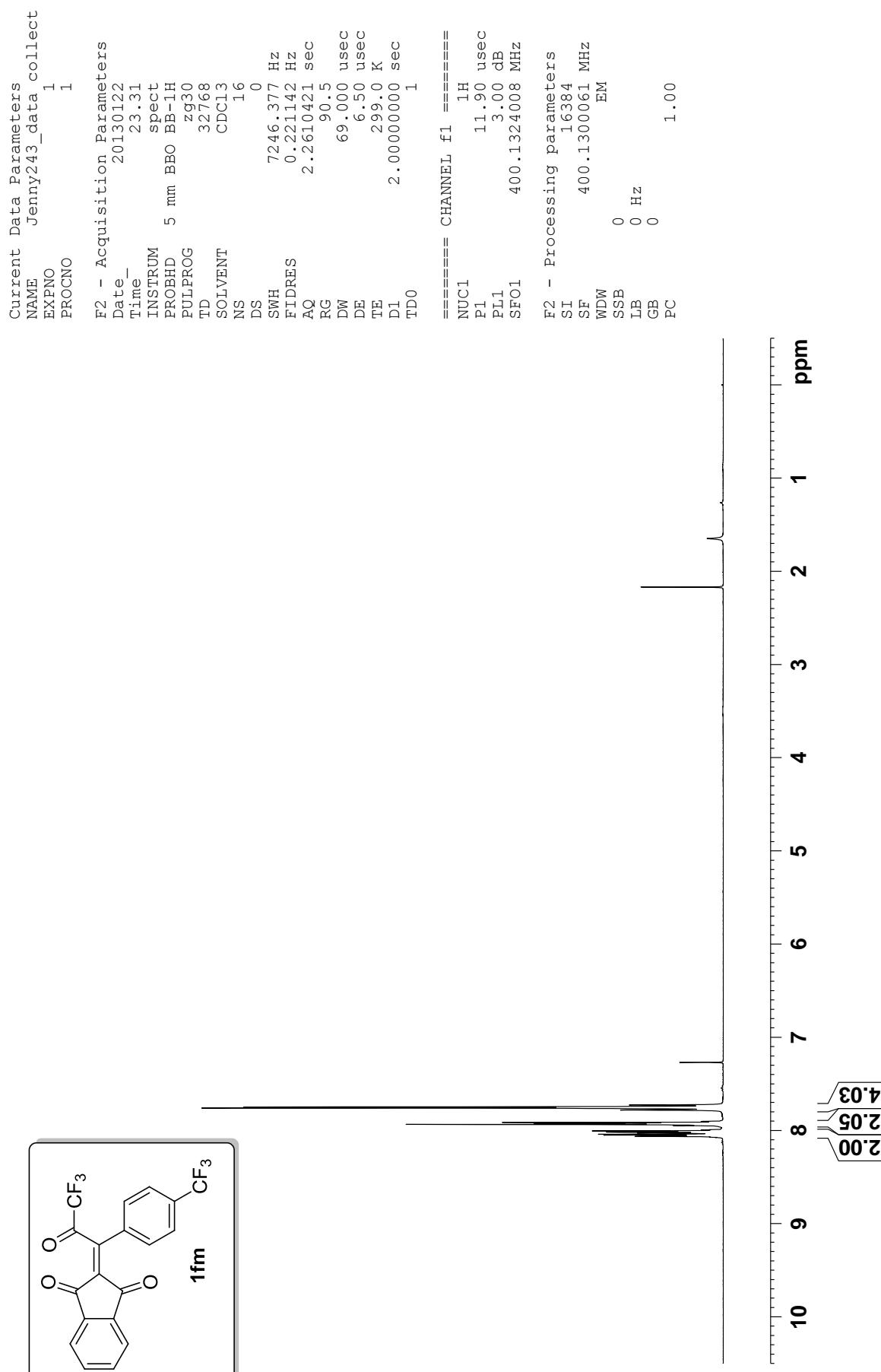












Current Data Parameters

NAME	Jenny
EXPNO	2
PROCNO	1

F2 - Acquisition Parameters

Date	20130420
Time	14.39
INSTRUM	spect
PROBHD	5 mm PABBO BB-
PULPROG	zgpg30
TD	65536
SOLVENT	CDC13
NS	1515
DS	0
SWH	300.30-0.029 Hz
FIDRES	0.458222 Hz
AQ	1.091244 sec
RG	9195.2
DW	16.650 usec
DE	6.50 usec
TE	29.6.3 K
D1	2.00000000 sec
D11	0.03000000 sec
TDO	

===== CHANNEL f1 =====

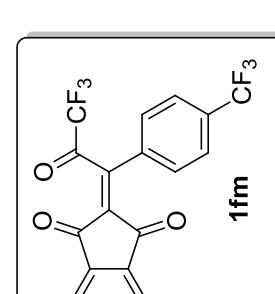
NUC1	13C
P1	10.00 usec
PL1	6.50 dB
SFO1	125.7709931 MHz

===== CHANNEL f2 =====

CPDPRG2	waltz16
NUC2	1H
PCPD2	90.00 usec
PL2	-0.60 dB
PL12	15.20 dB
PL13	18.20 dB
SFO2	500.1320005 MHz

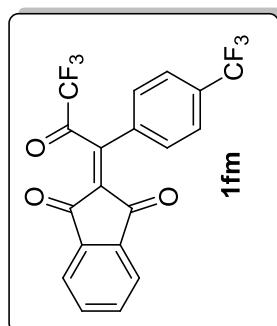
F2 - Processing parameters

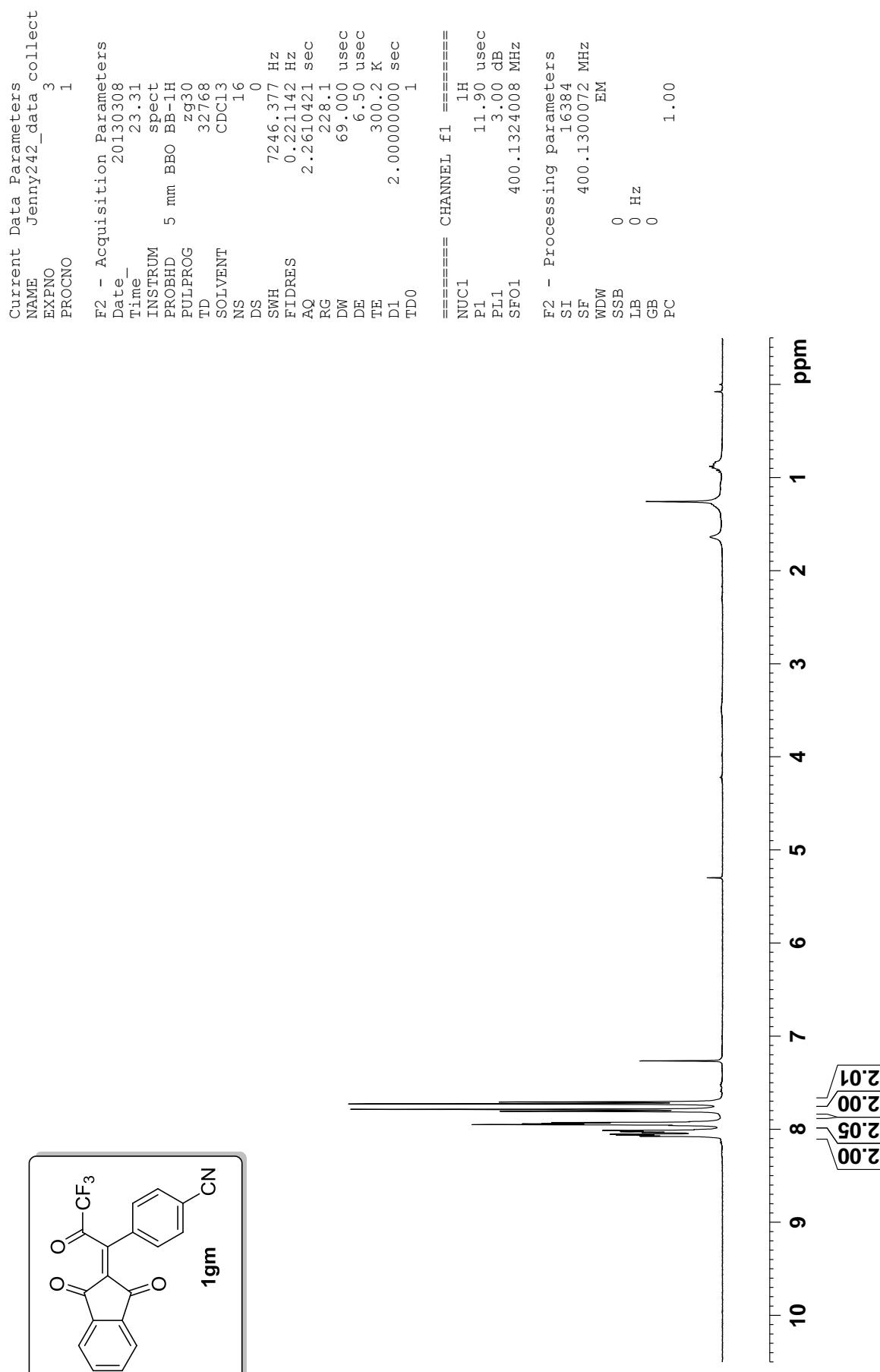
SI	65536
SF	125.7577930 MHz
WDW	SSB
SSB	0
LB	1.00 Hz
GB	0
PC	1.00

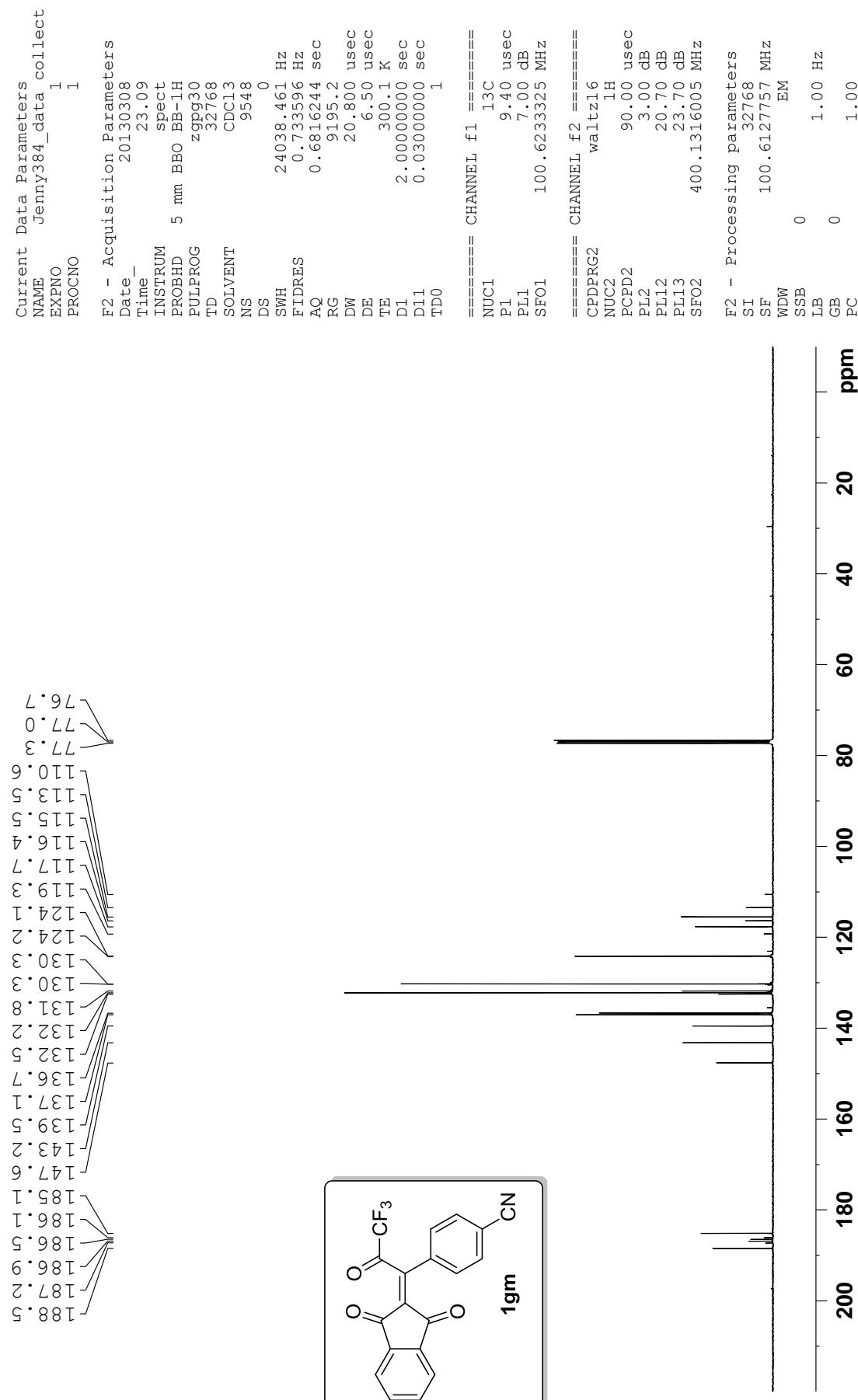


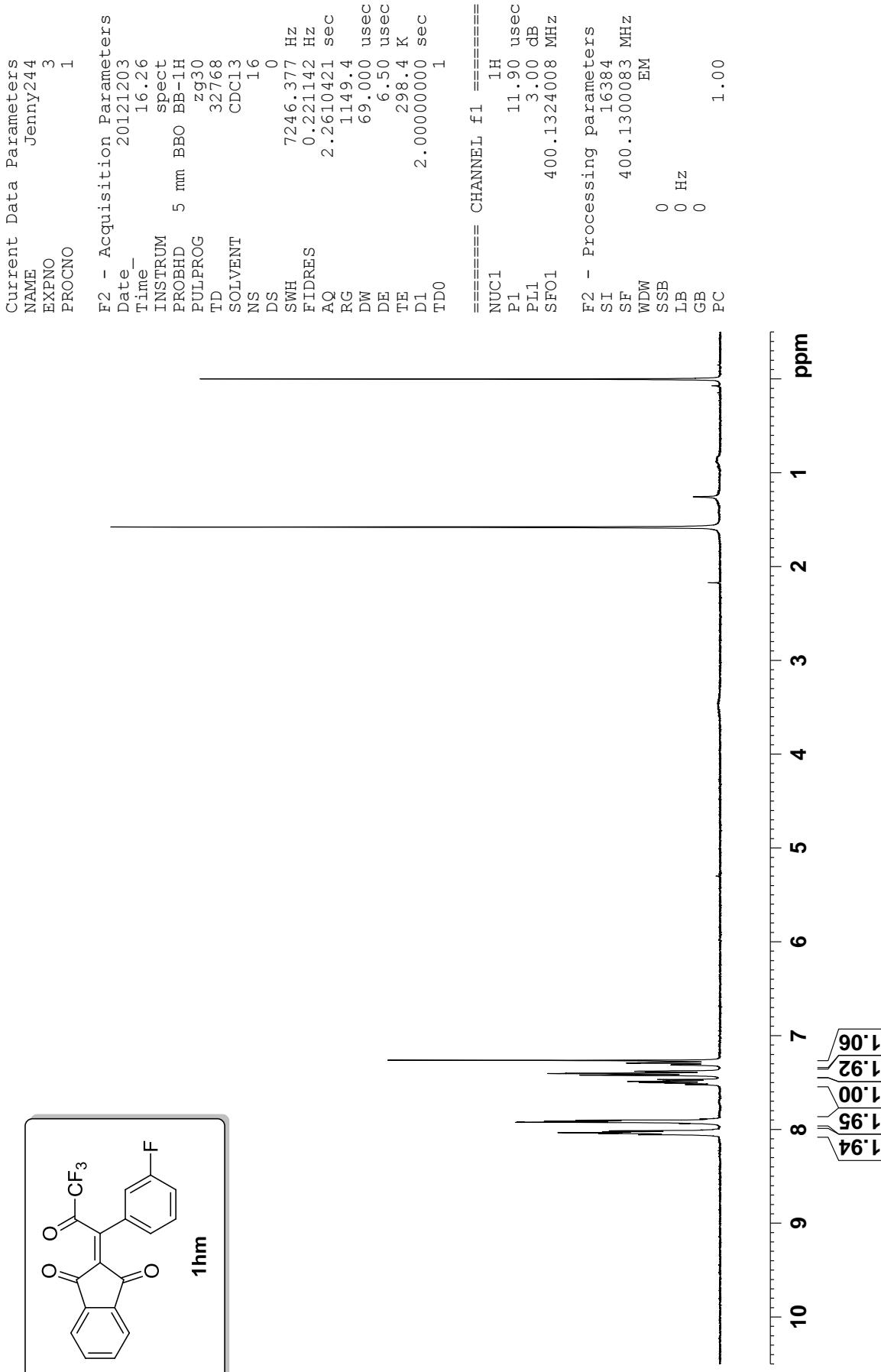
ppm

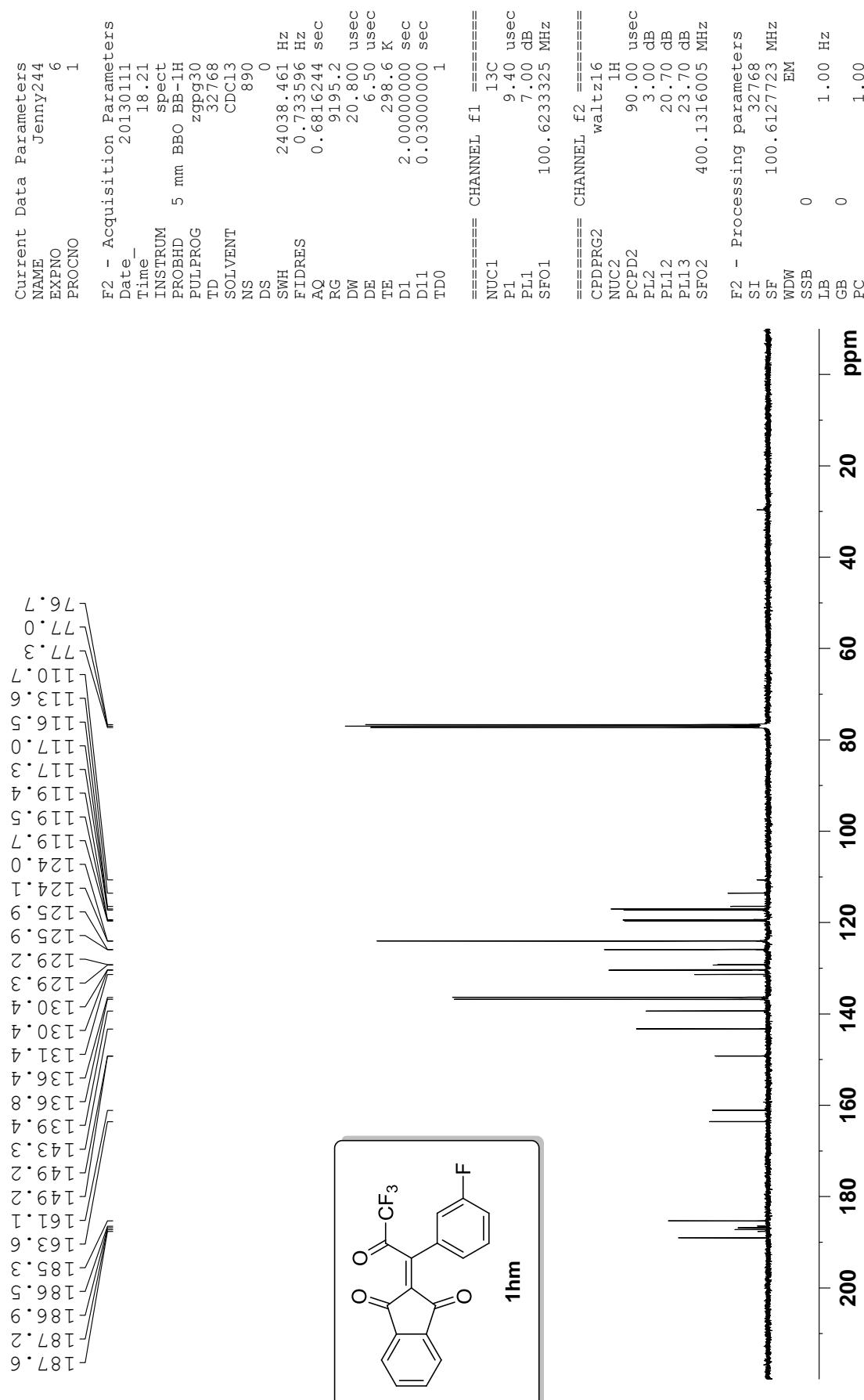
200 180 160 140 120 100 80 60 40 20

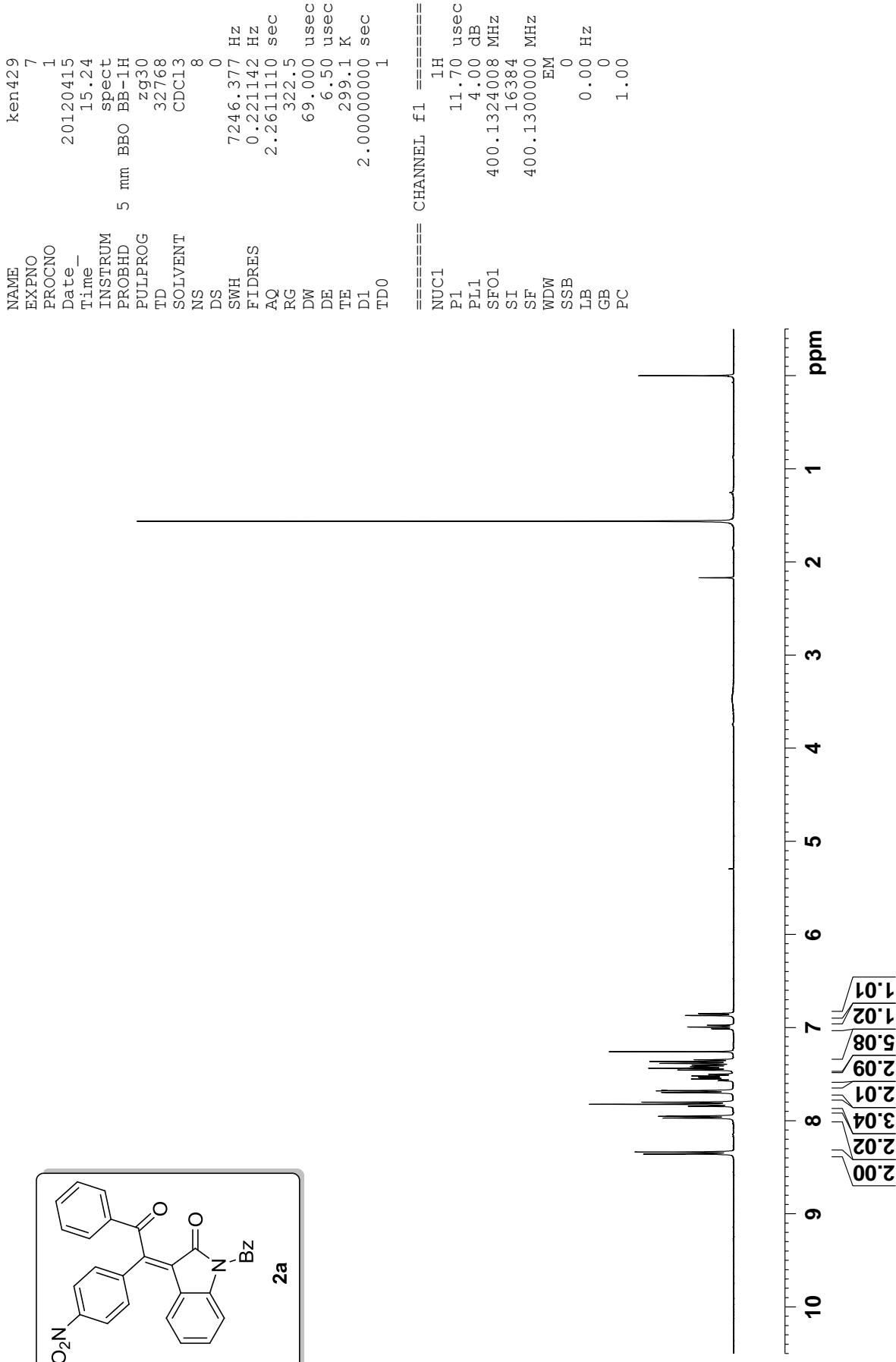


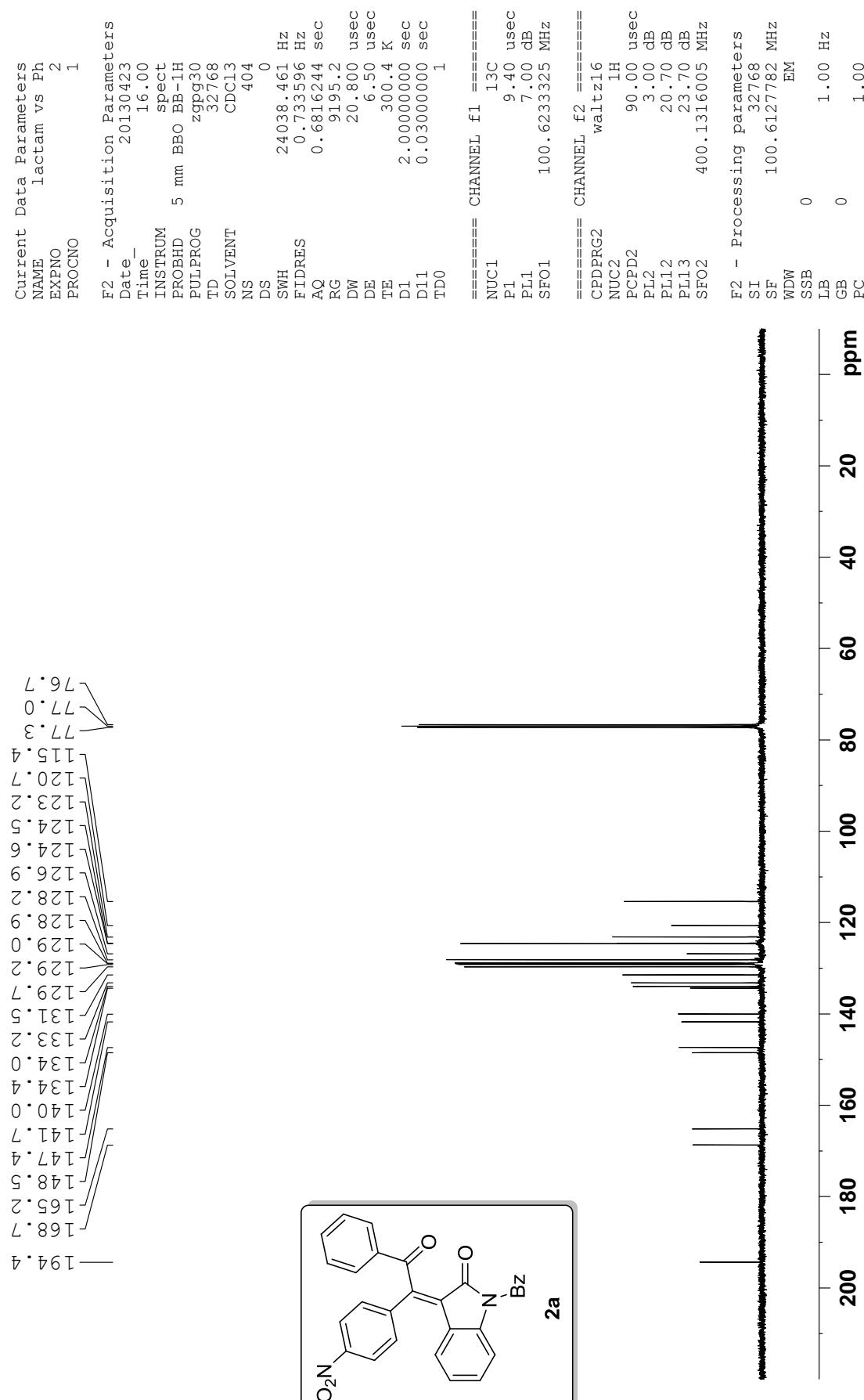


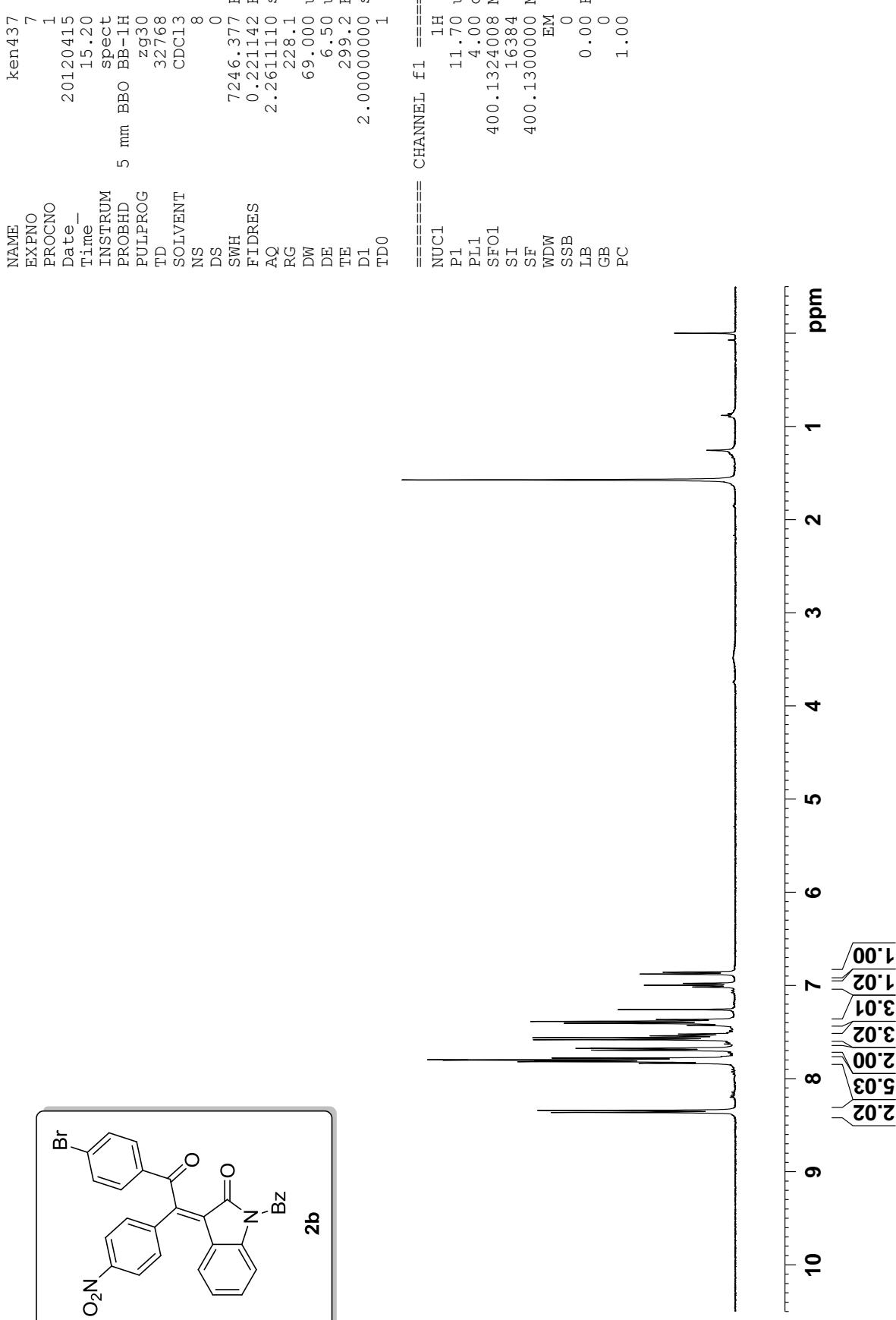


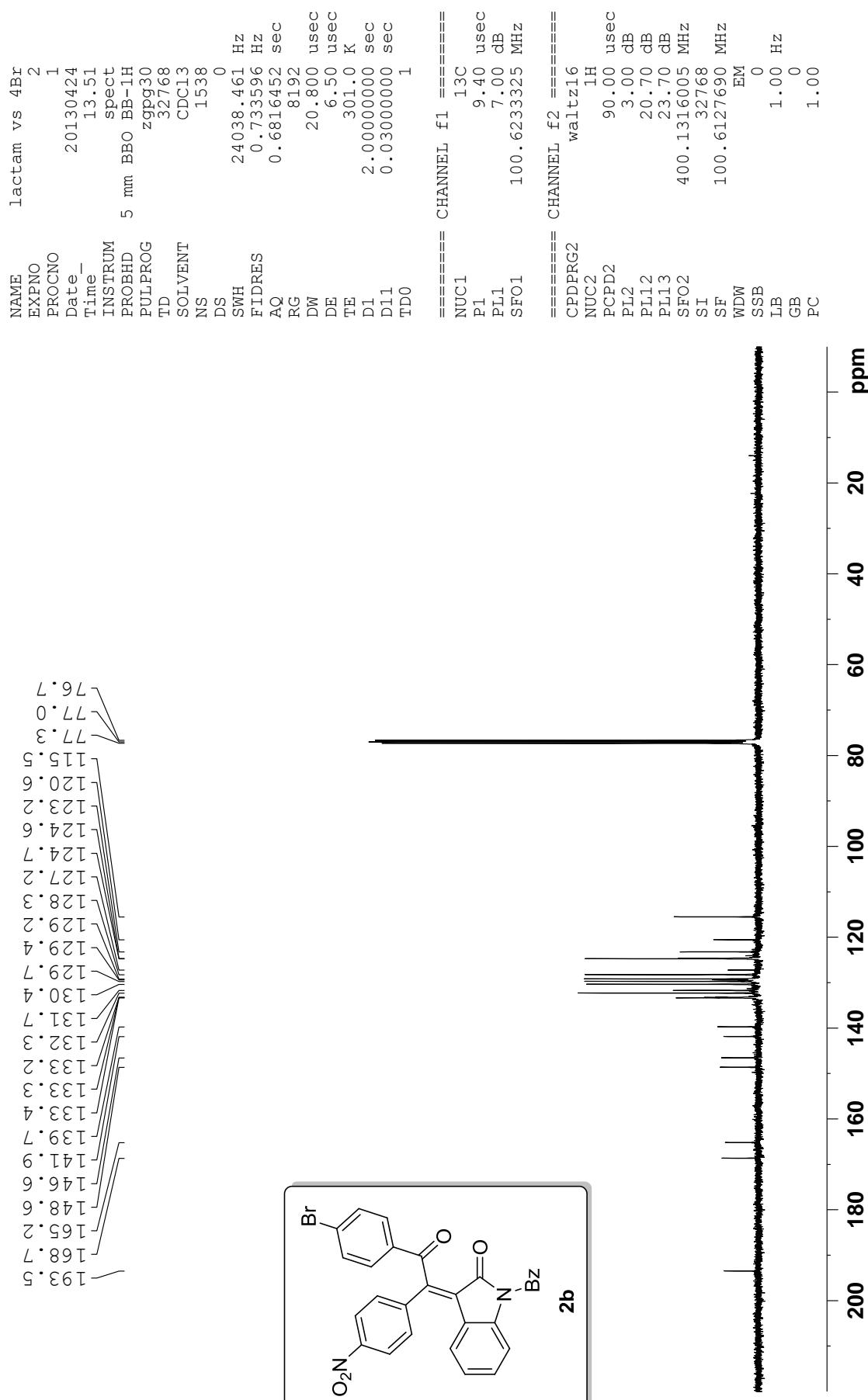


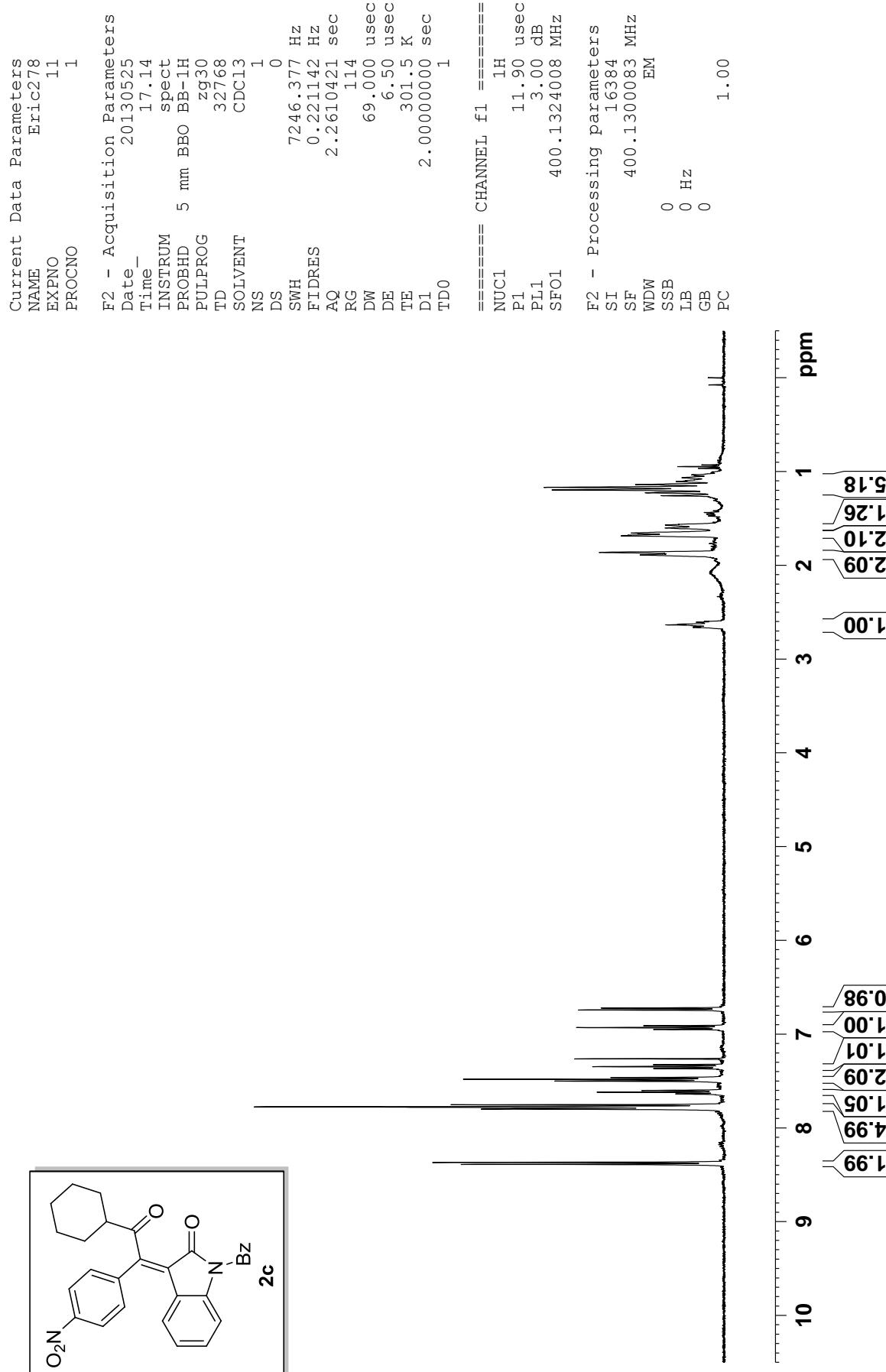


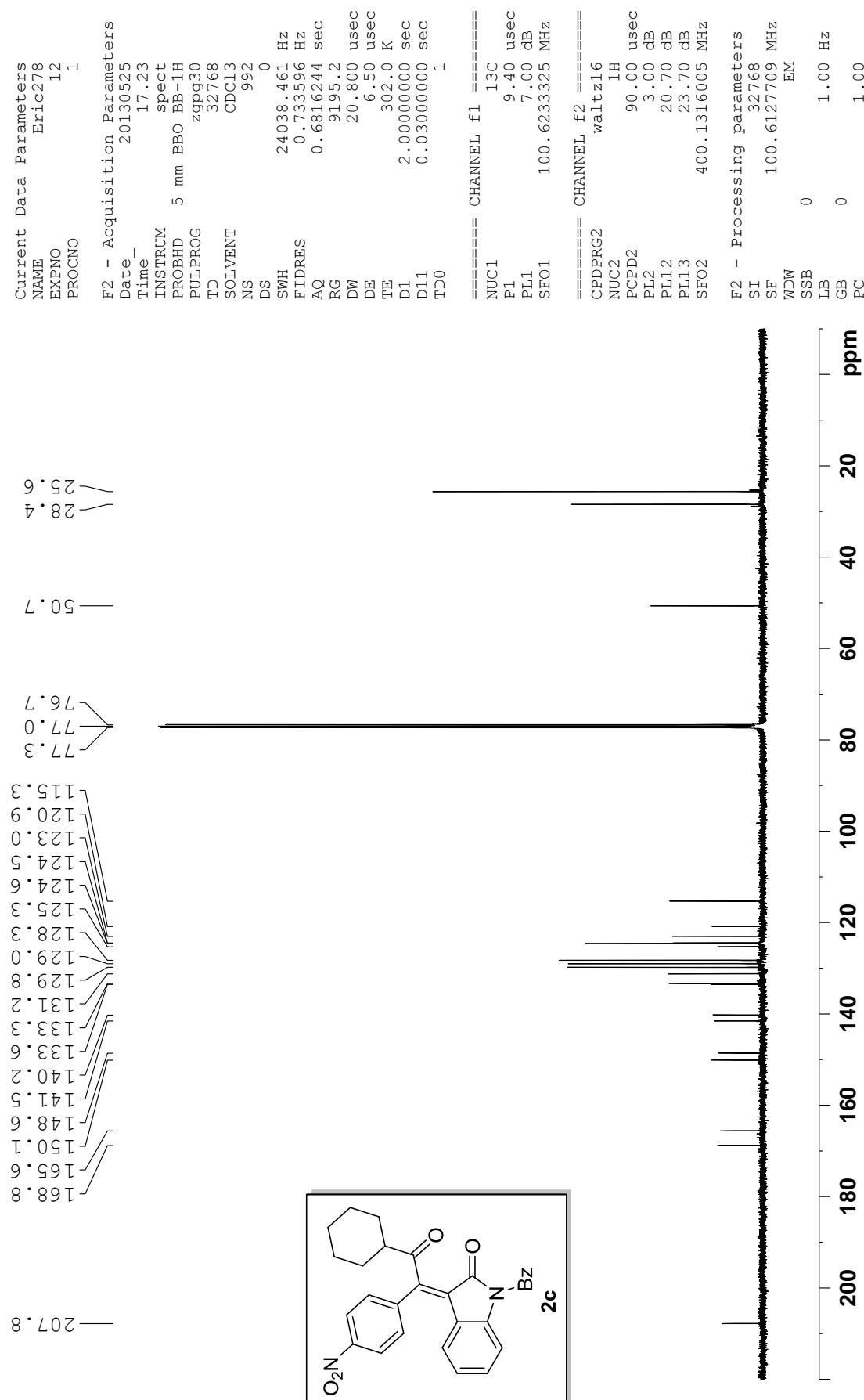


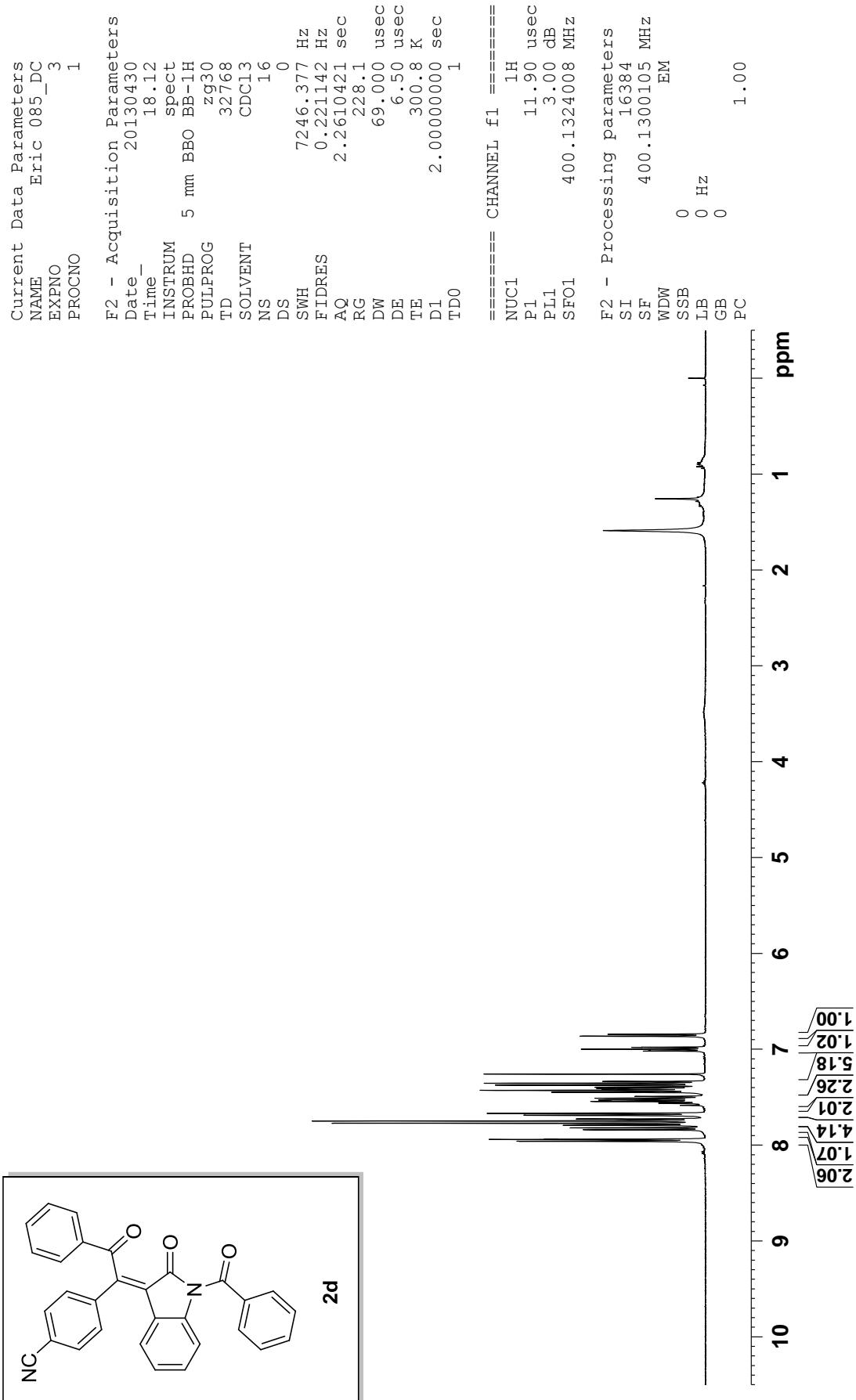


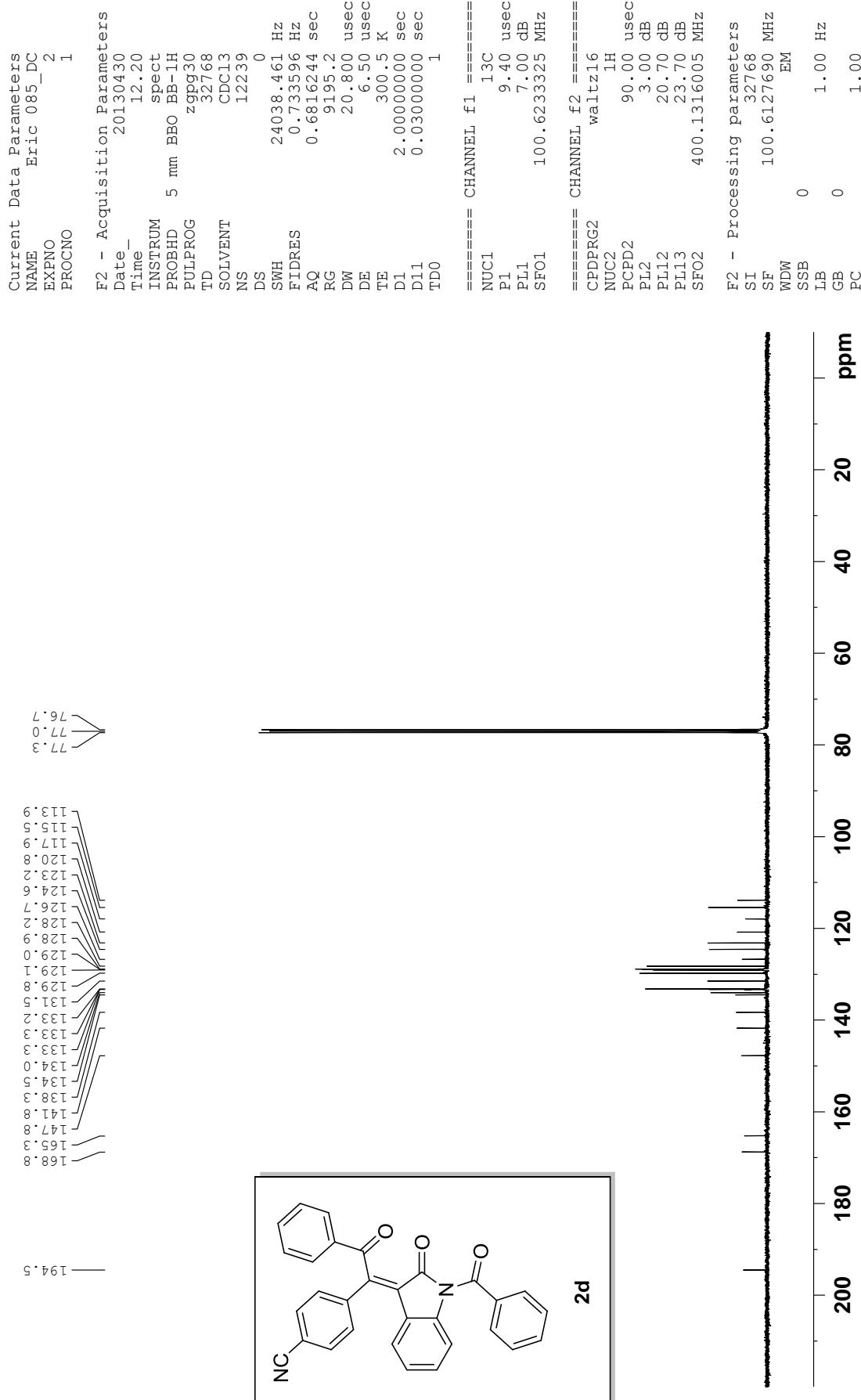


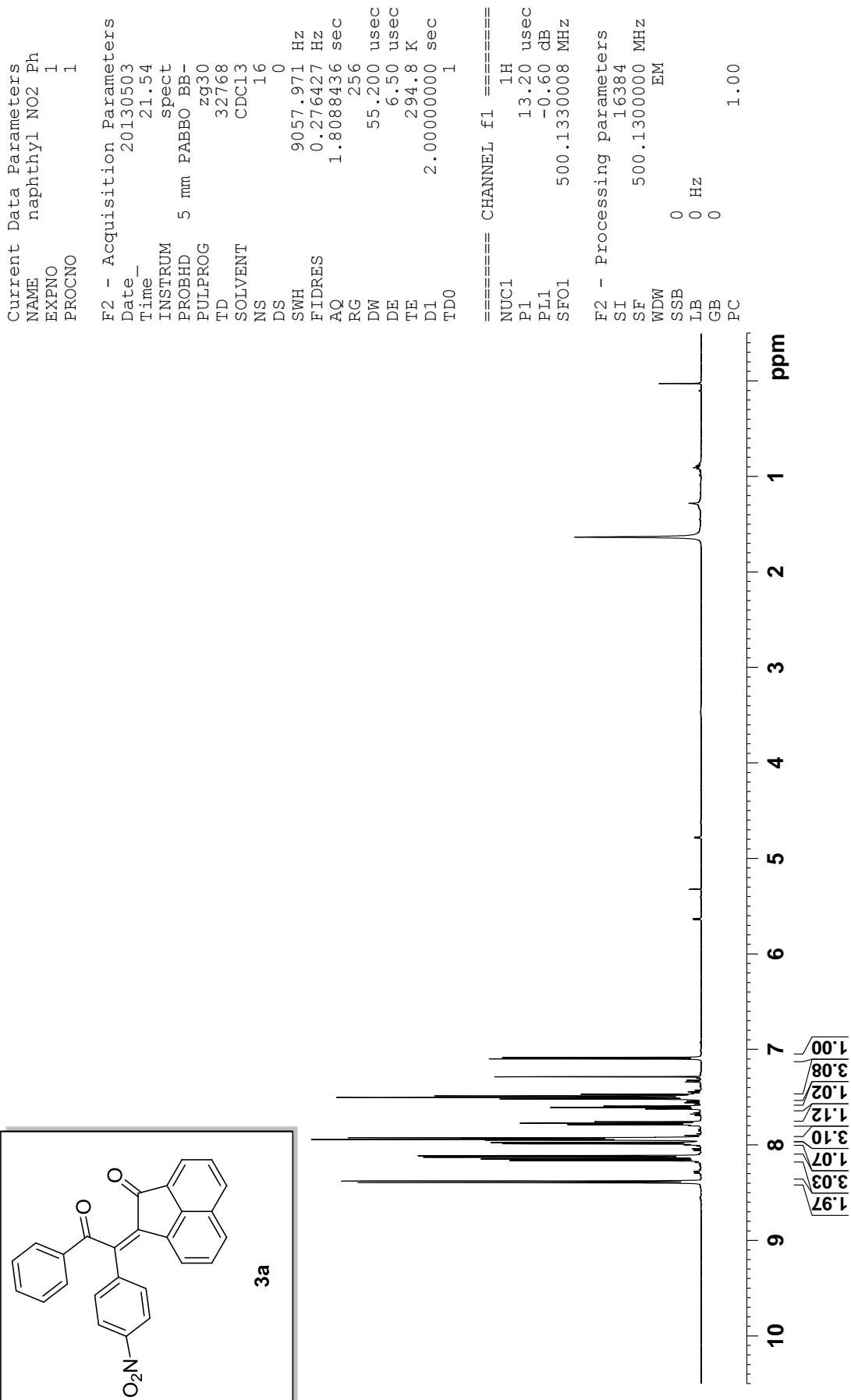


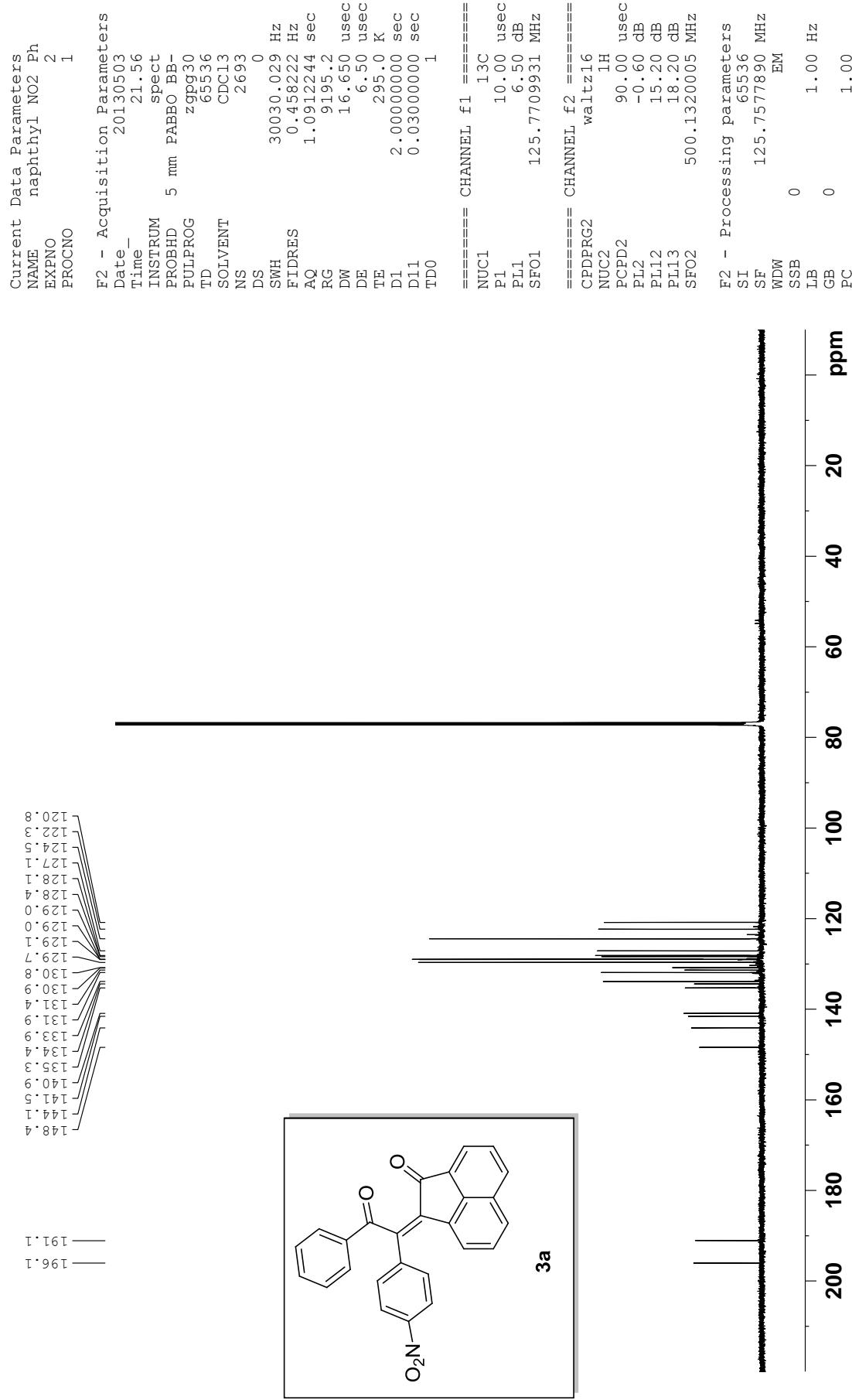


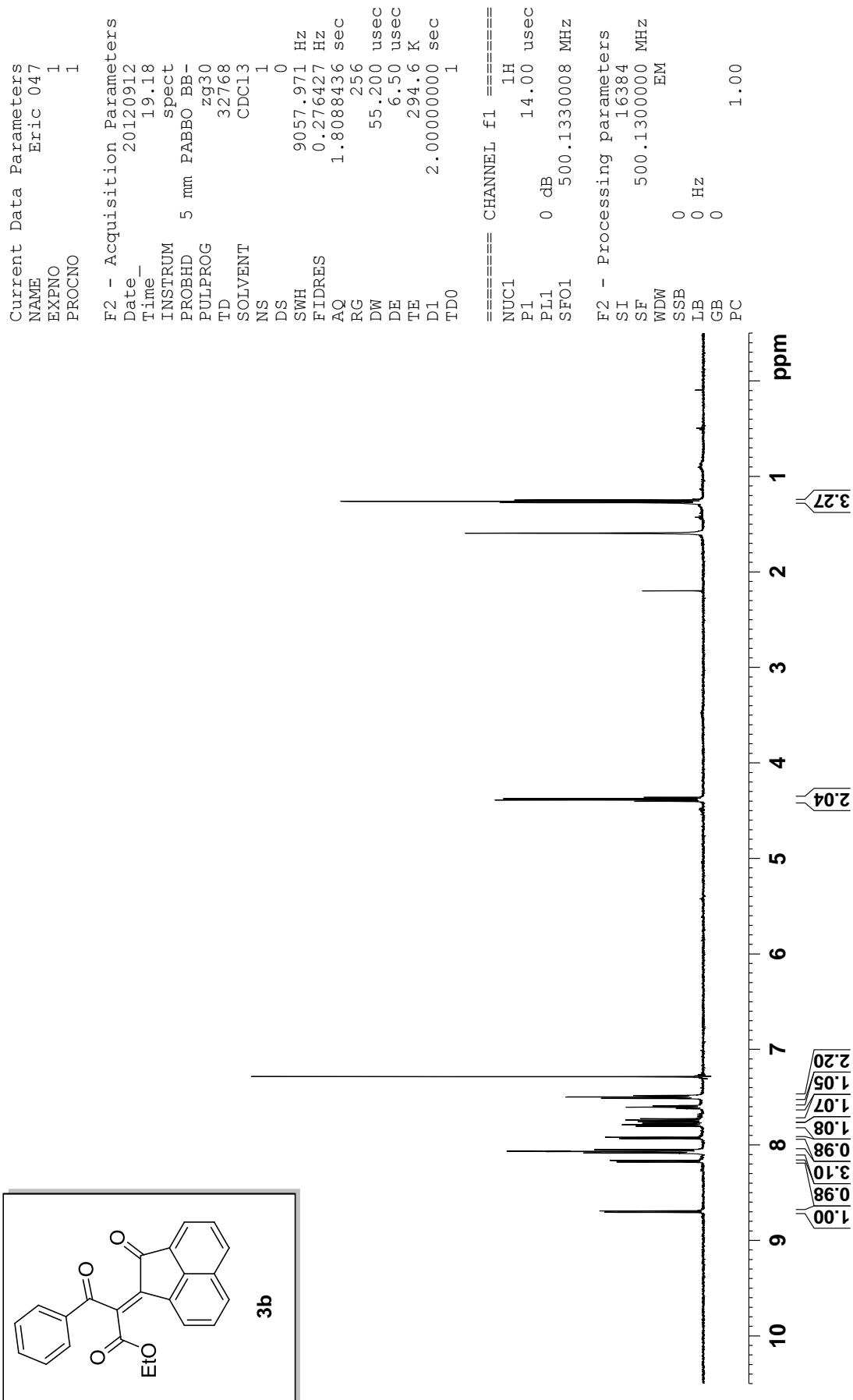


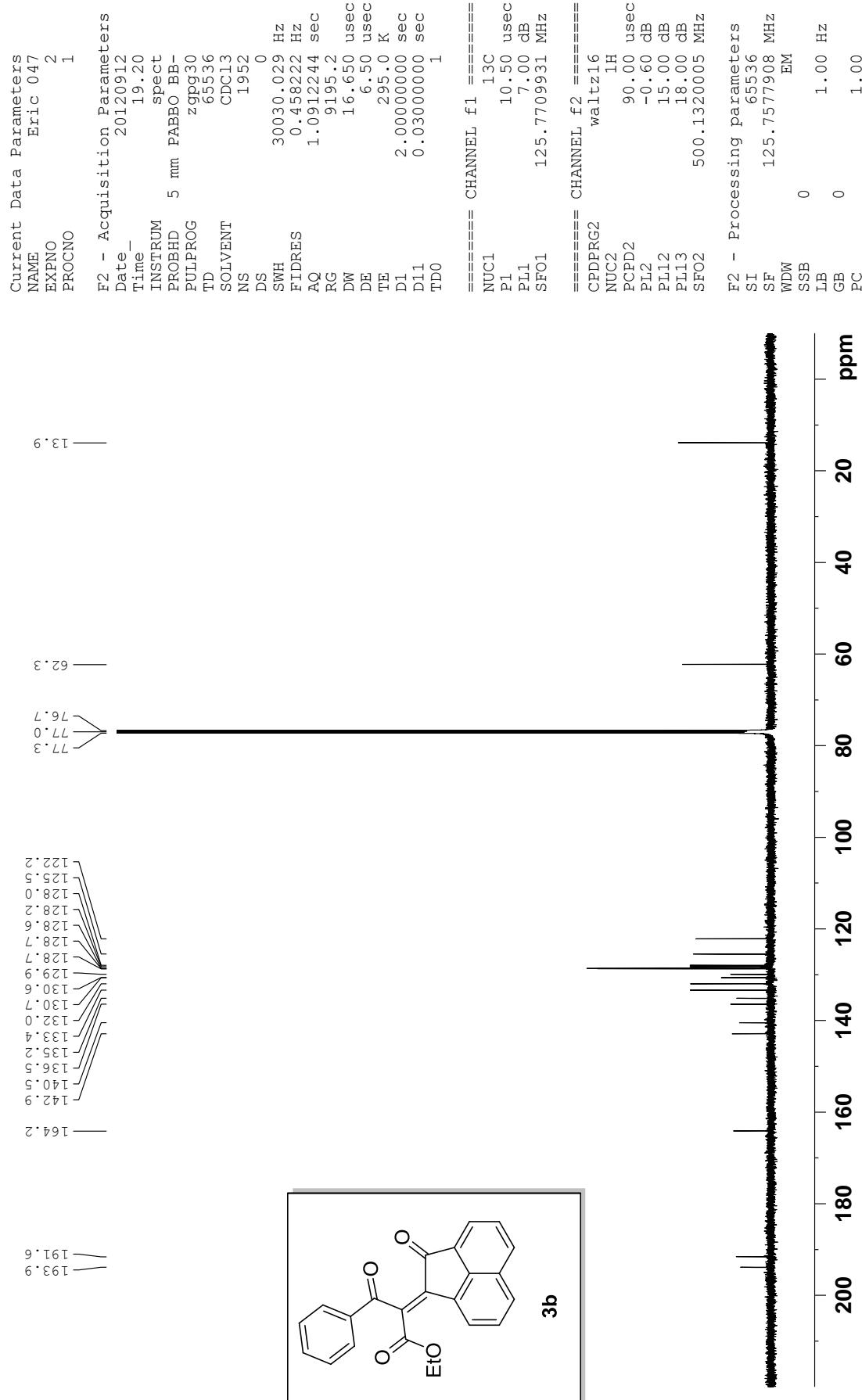


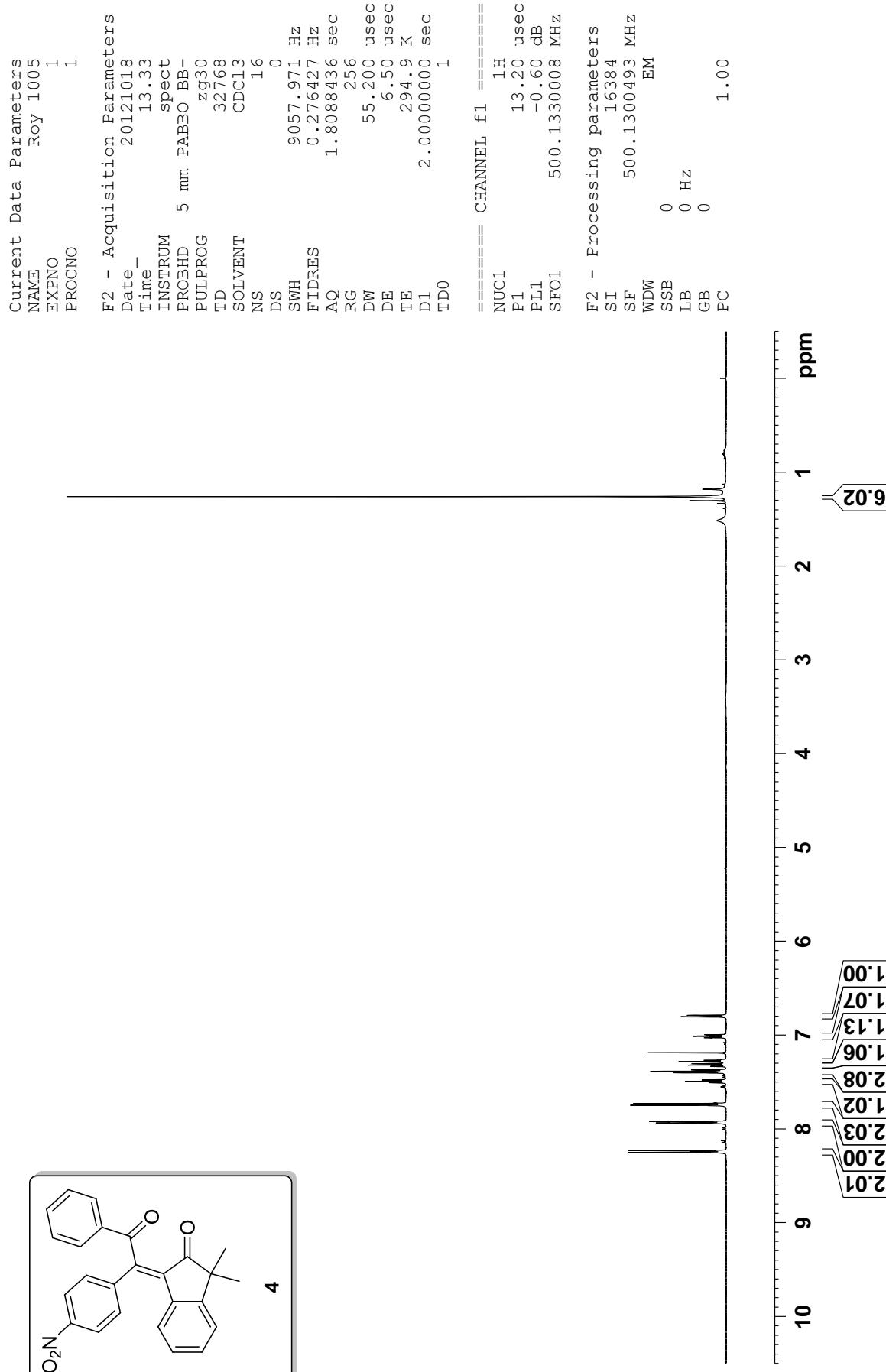


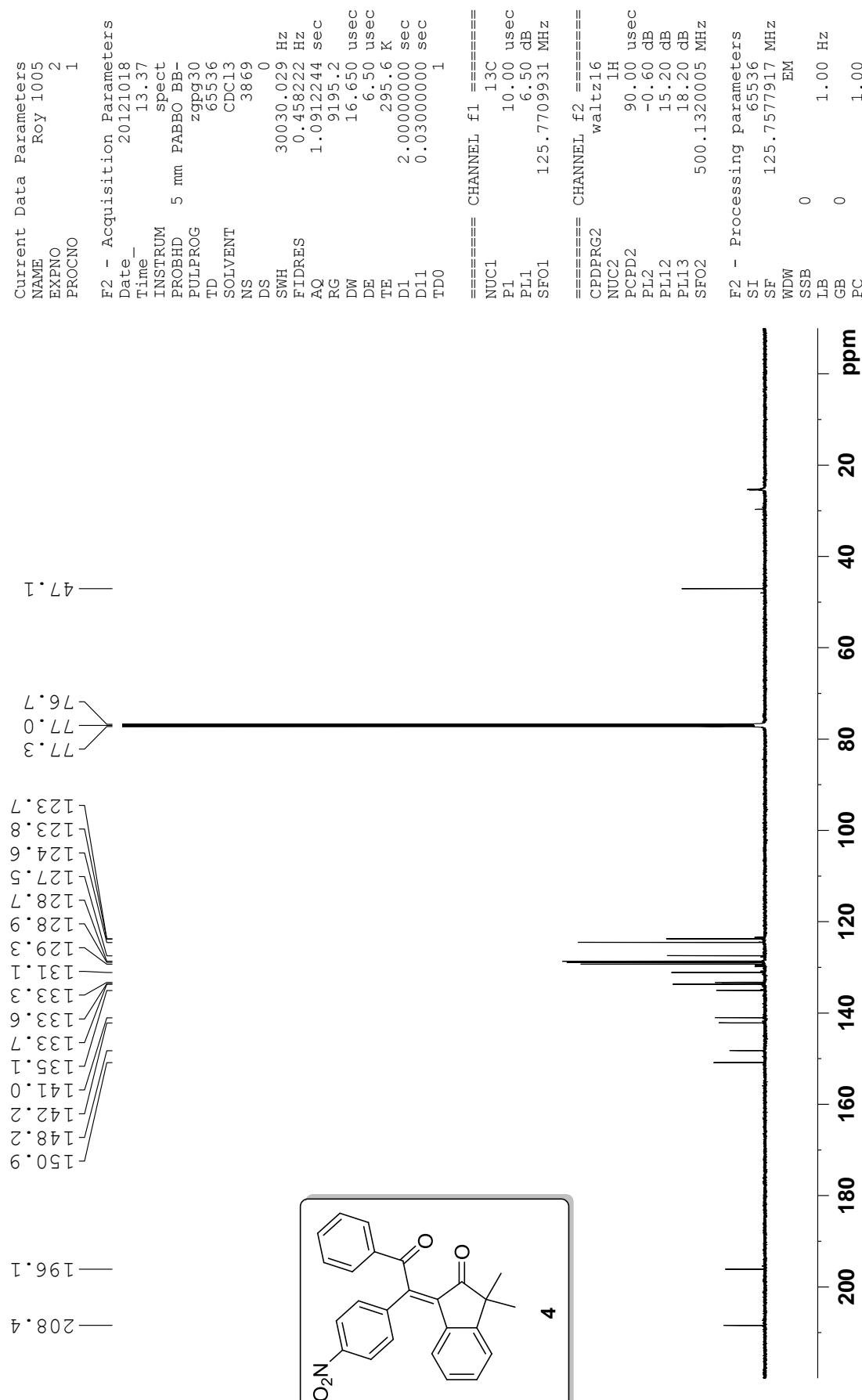










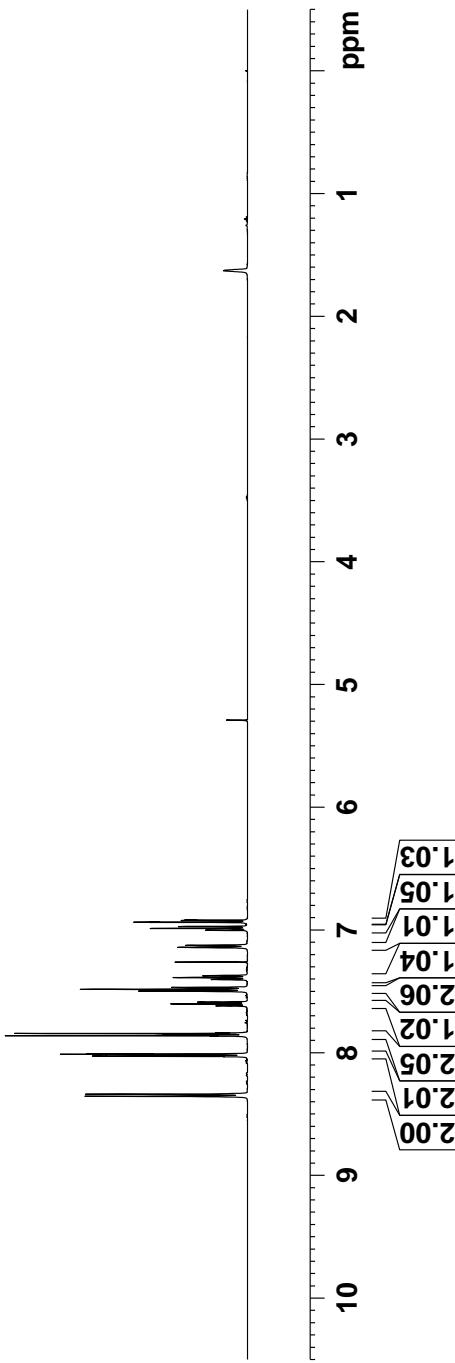
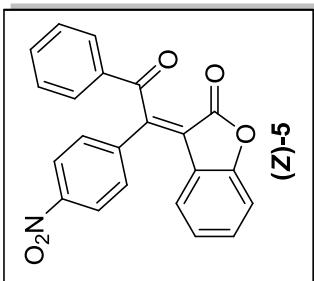


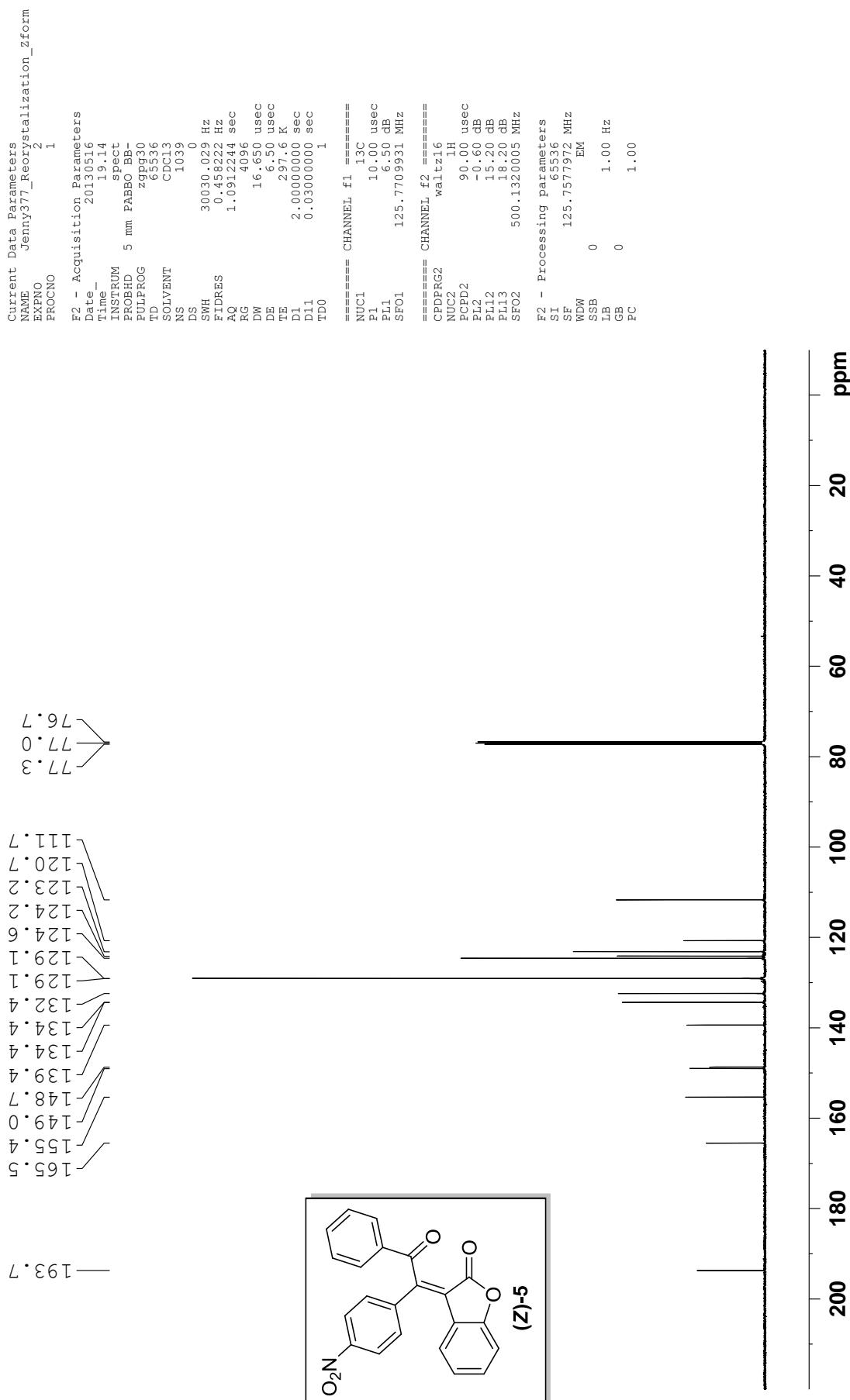
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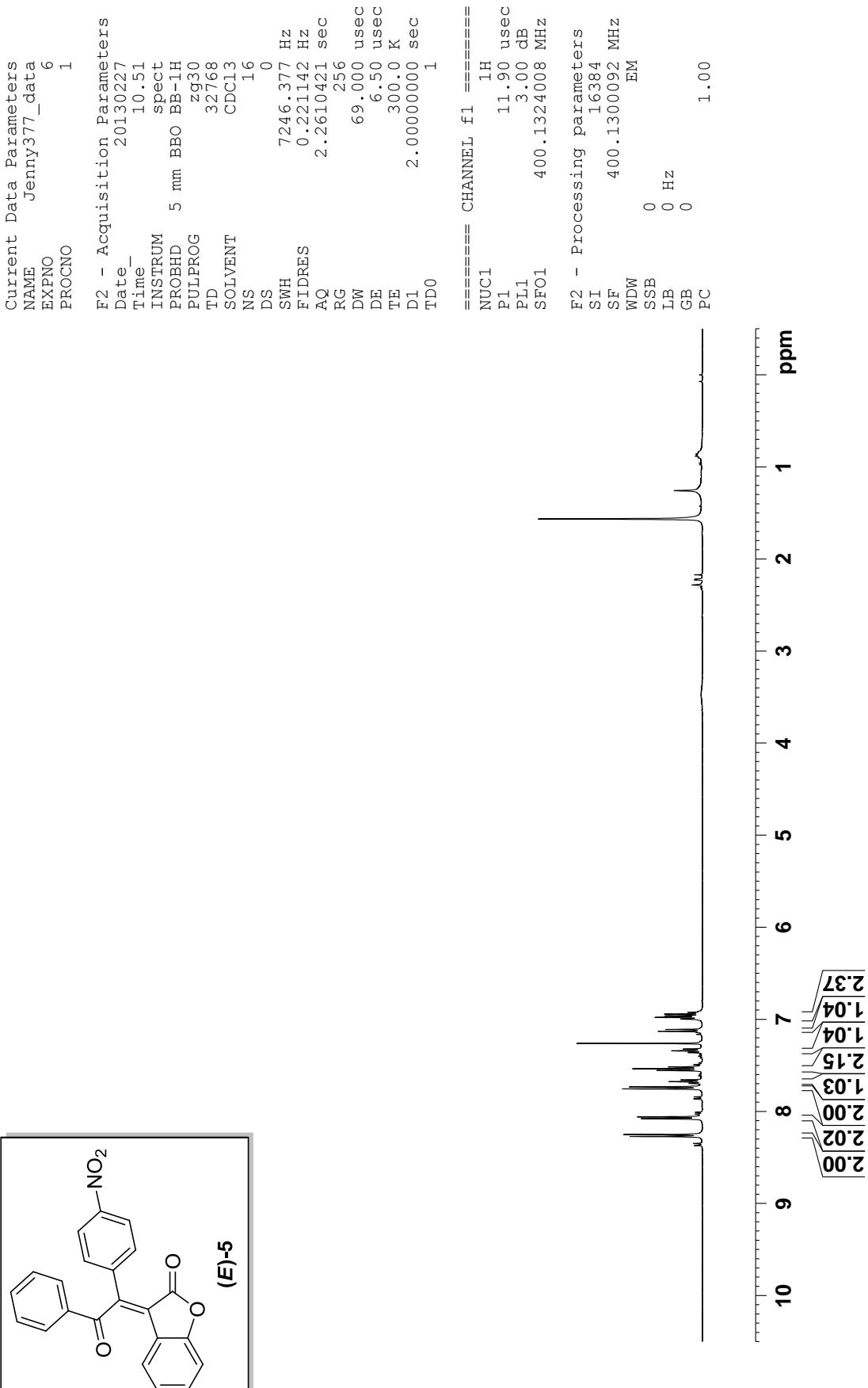
Current Data Parameters           F2 - Acquisition Parameters
NAME      Jenny377_ReCRYSTALLIZATION_Zform
EXPNO     1
PROCNO    1

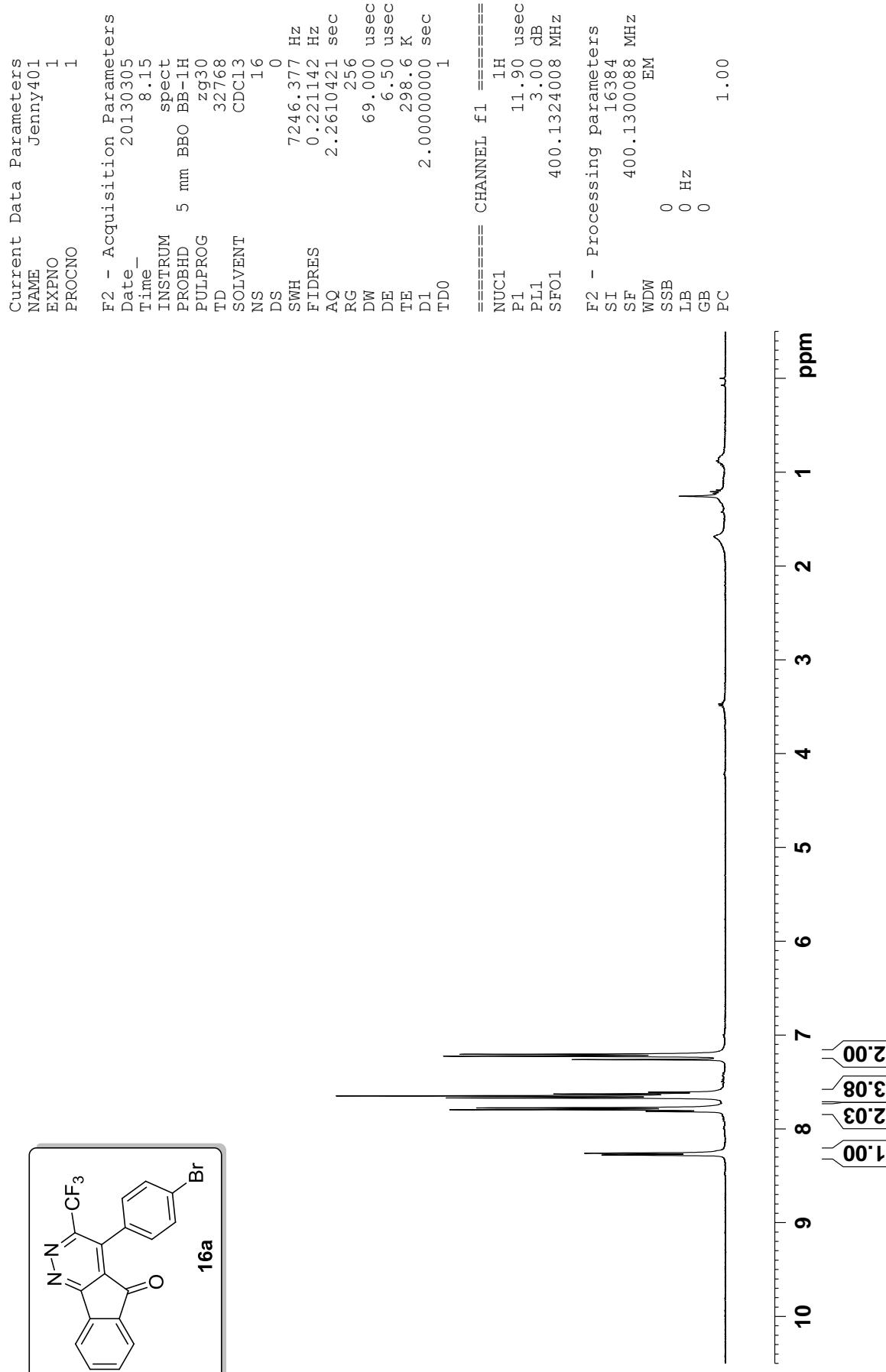
F2 - Processing parameters
SI          16384
SF          500.1300123 MHz
WDW        EM
SSB        0 Hz
LB          0 Hz
GB          0
PC          1

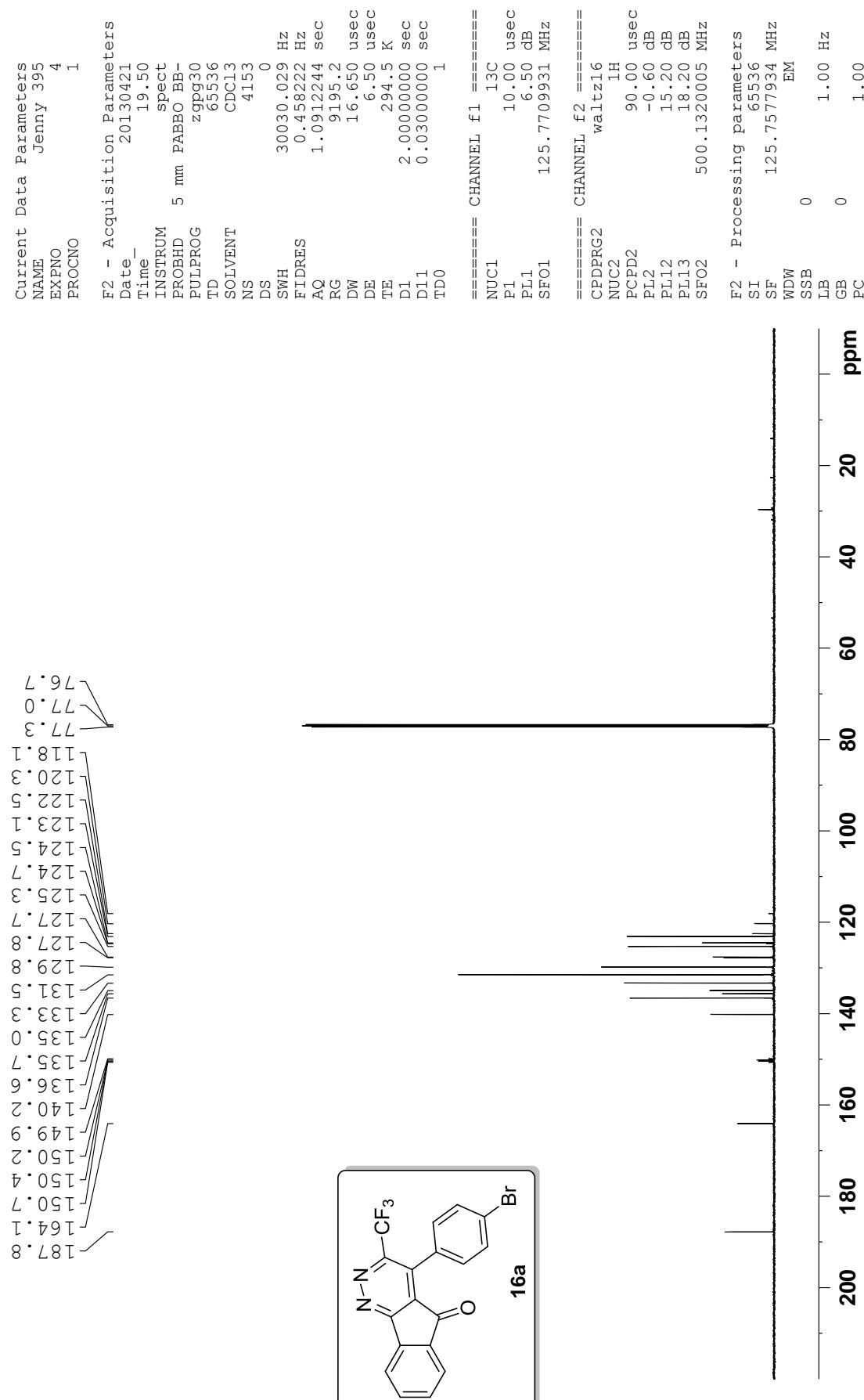
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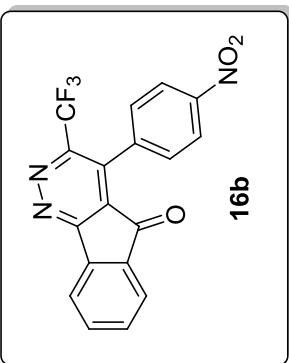












```

Current Data Parameters
NAME      Jenny394_Data
EXPNO    1
PROCNO  1

```

```

F2 - Acquisition Parameters
Date : 20130417
Time : 1.48

INSTRUM      BBO          spect
PROBHD      5 mm        BB-1H
PULPROG     zg30        zg30
TD           32768       sec
                CDC13      :
                CDC13      :
SOLVENT      NS          16
                DS          0
                SWH         7246.377 Hz
                FIDRES     0.221147 Hz
                AQ          2.2610421 sec
                RG          114
                DW          69.000 usec
                DE          6.50  usec
                TE          299.9 K
                D1          2.00000000 sec
                TDO

```

```

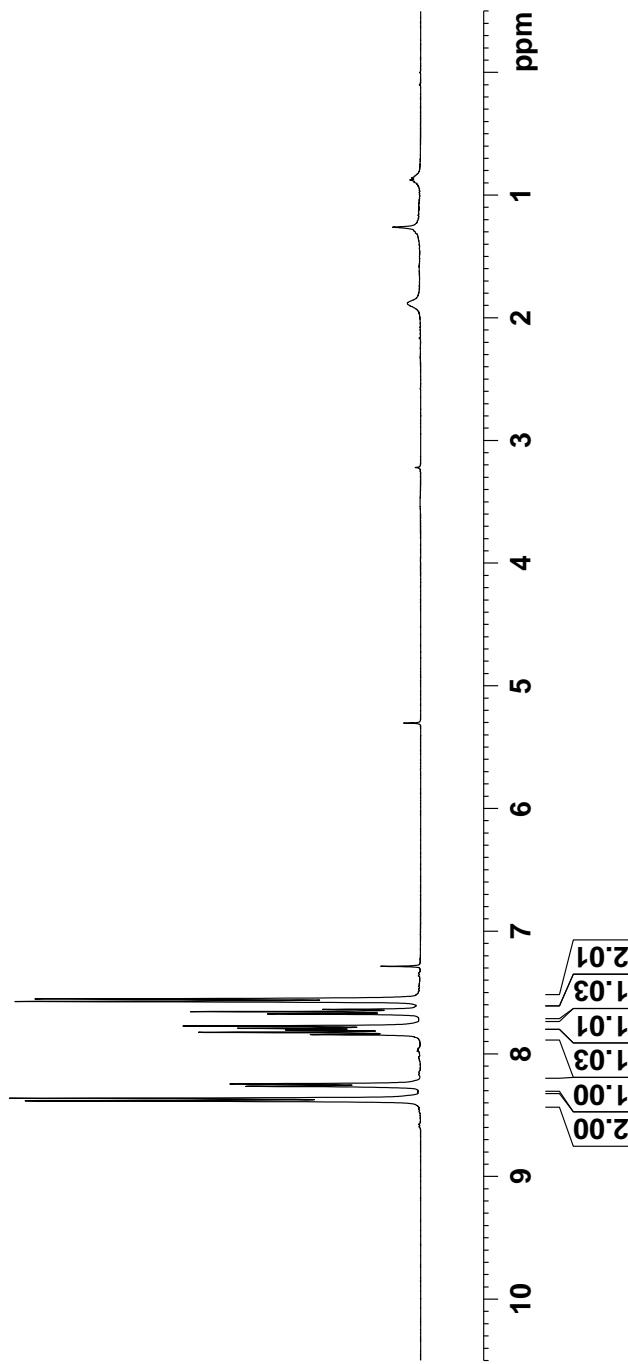
===== CHANNEL f1 =====
NUC1          1H
P1           11.90  usec
PL1          3.00   dB
SF01         400.1324008 MHz
                           EM

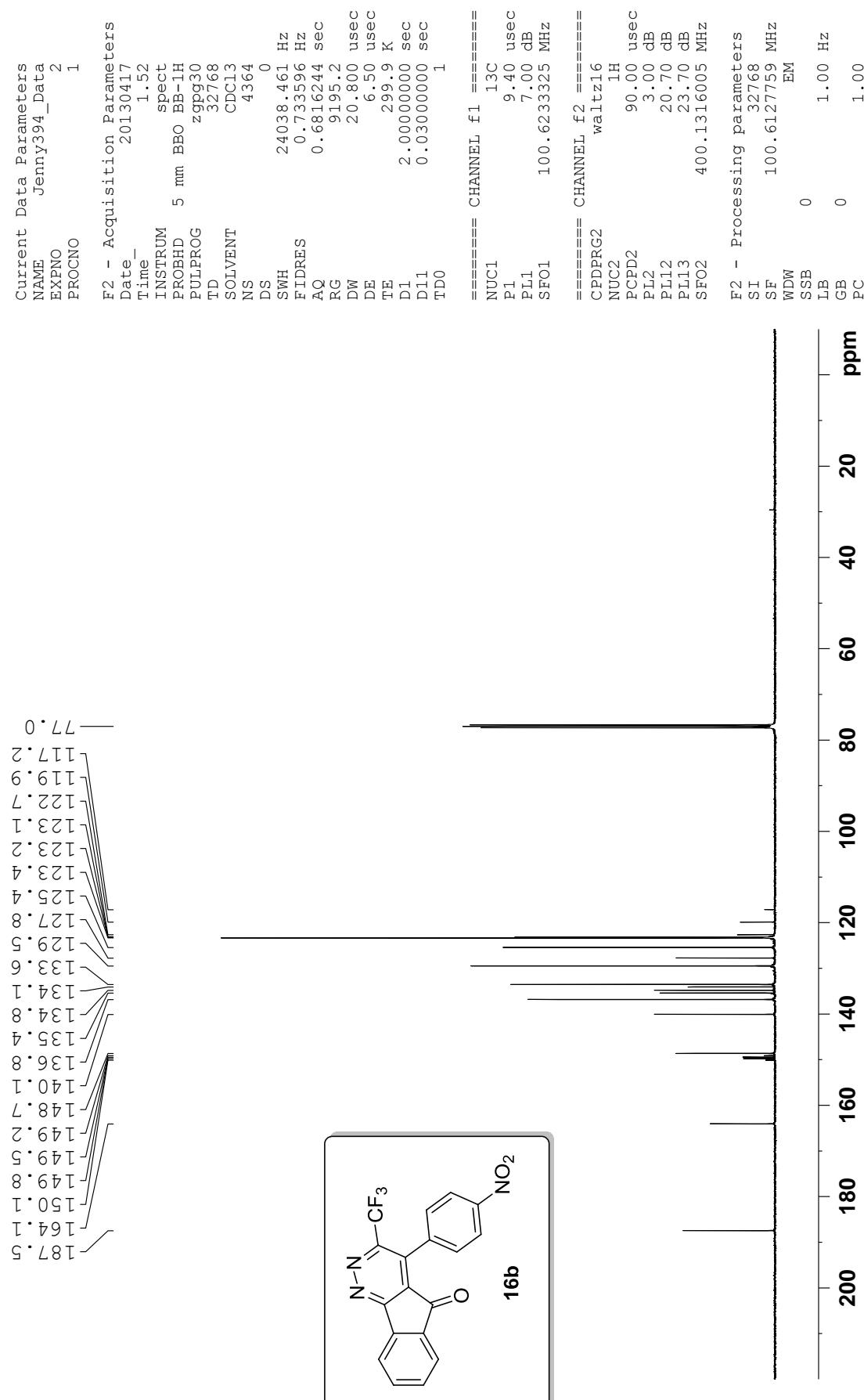
F2 - Processing parameters
SI            16384
SF           400.1299990 MHz
                           EM

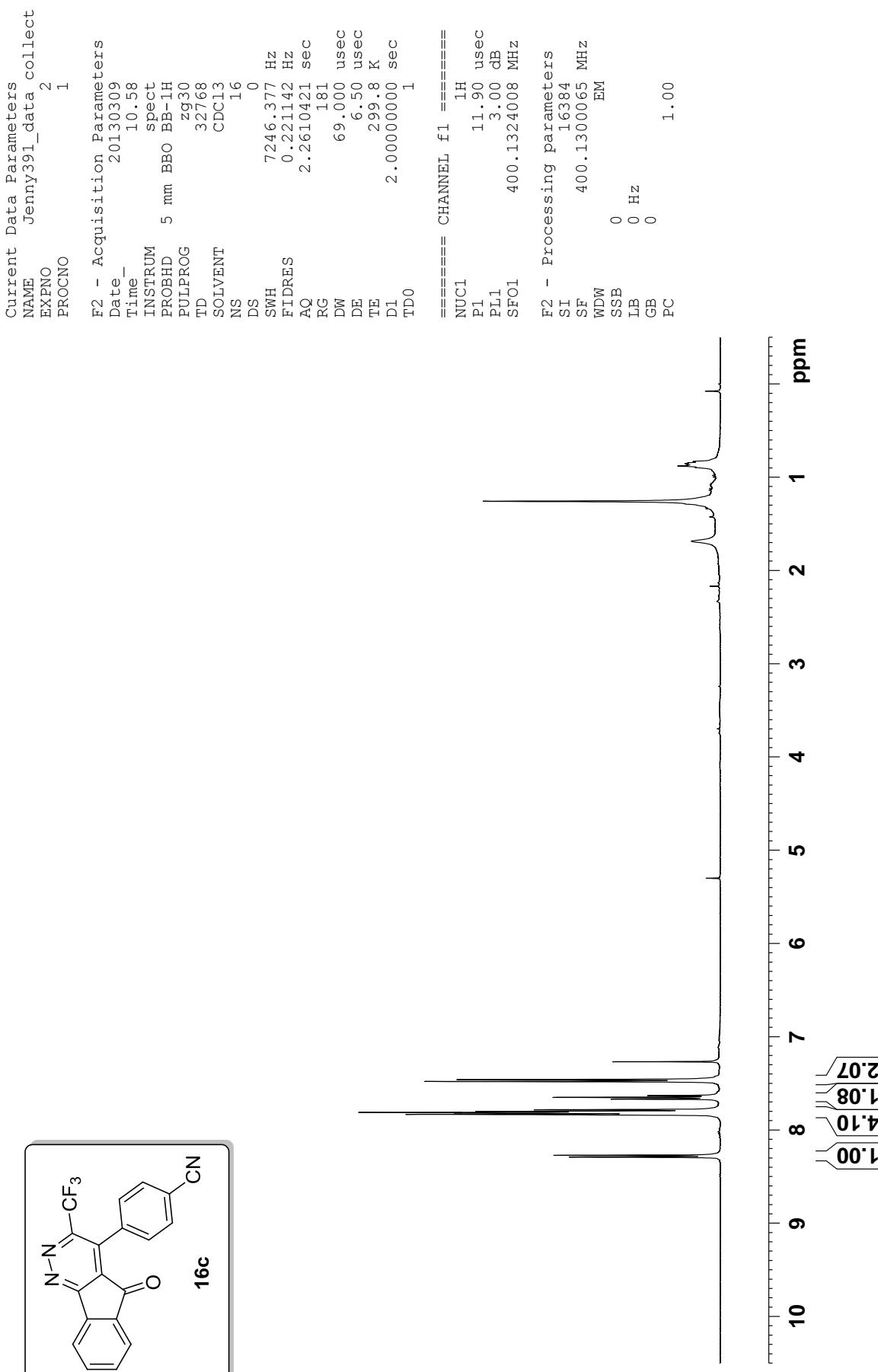
WDW
SSB          0
LB           0 Hz
GB           0
PC

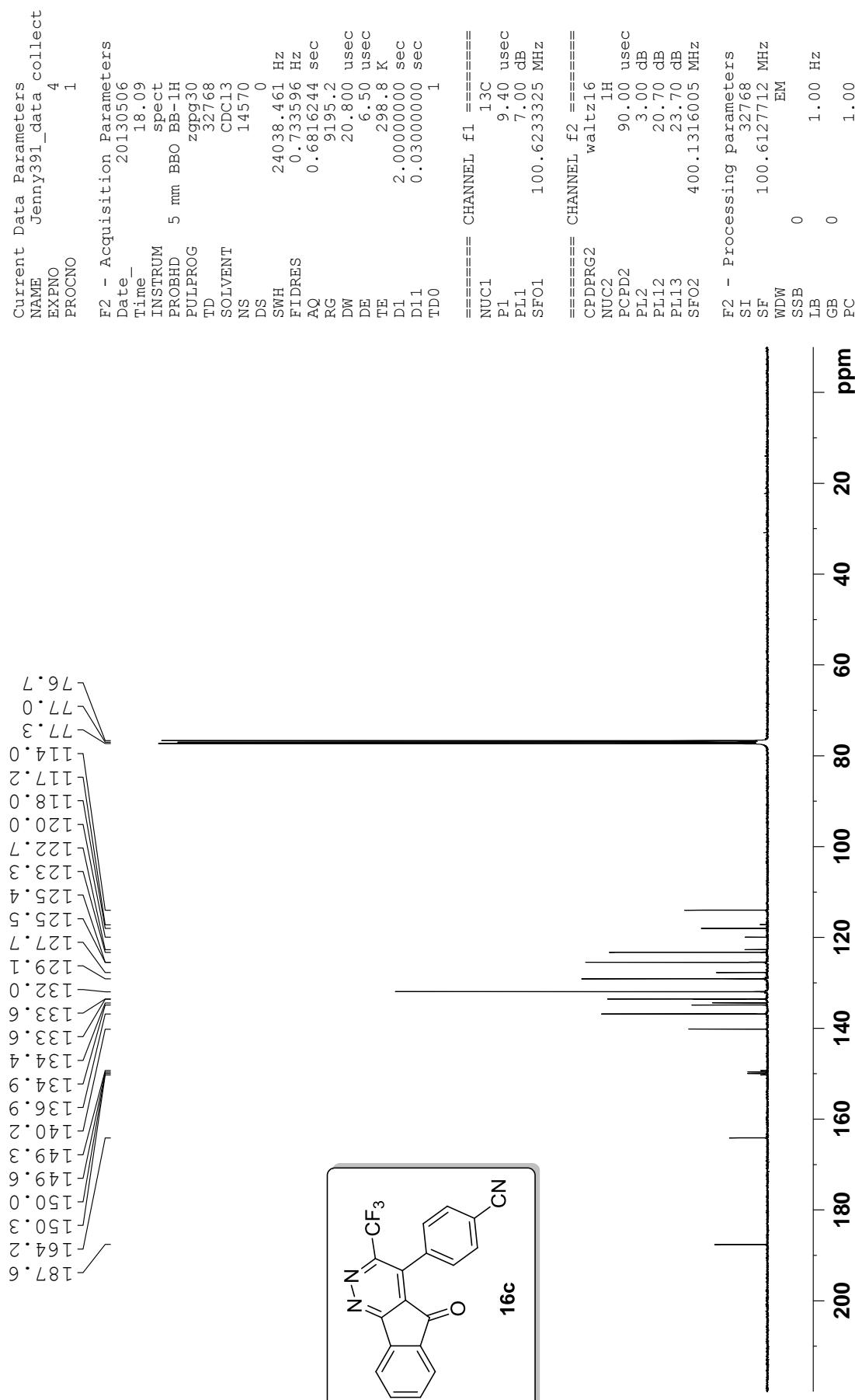
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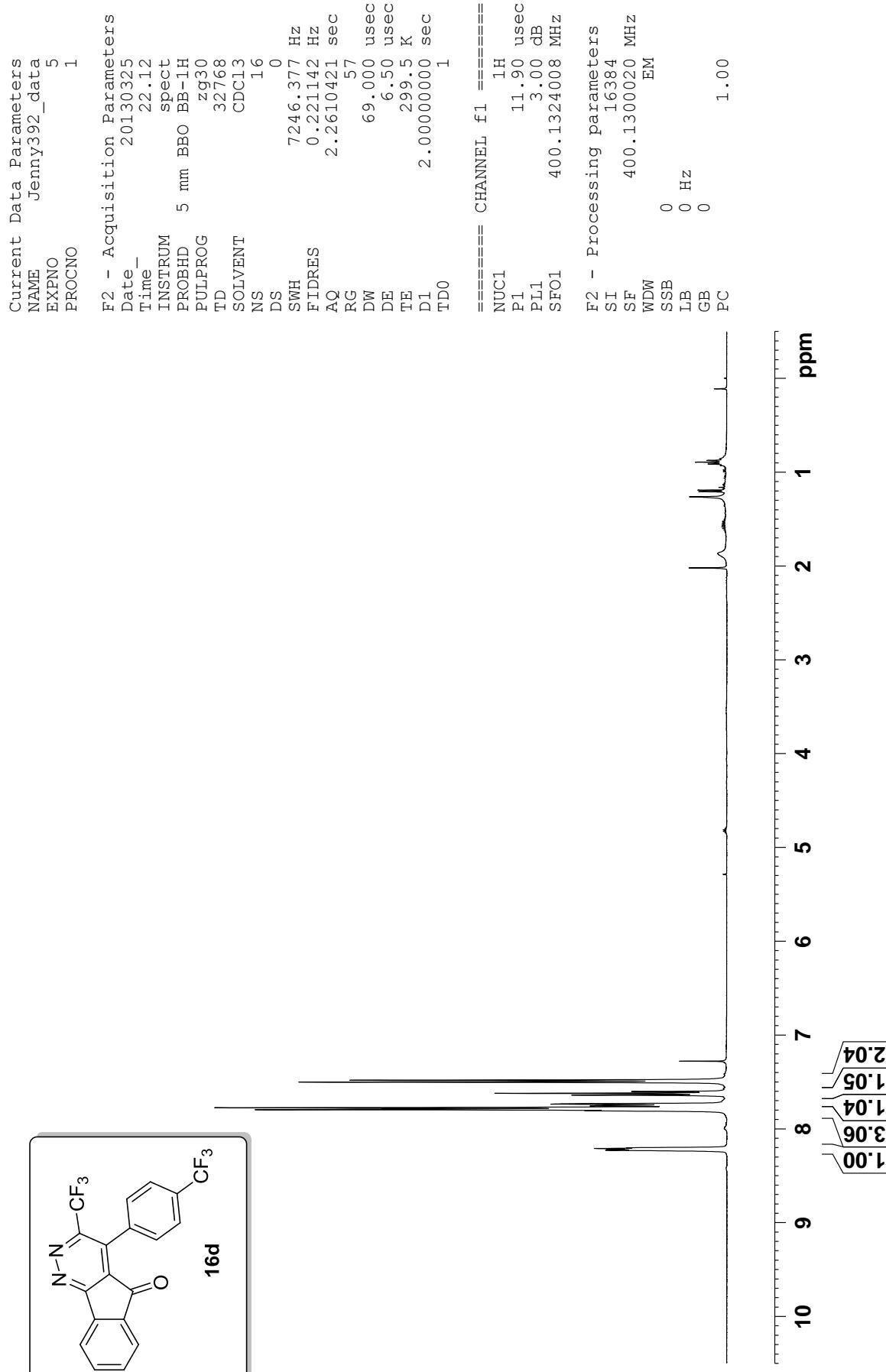
F2 – Processing parameters
 SI 16384
 SF 400 .129990 MHz
 WDM ^{FM}

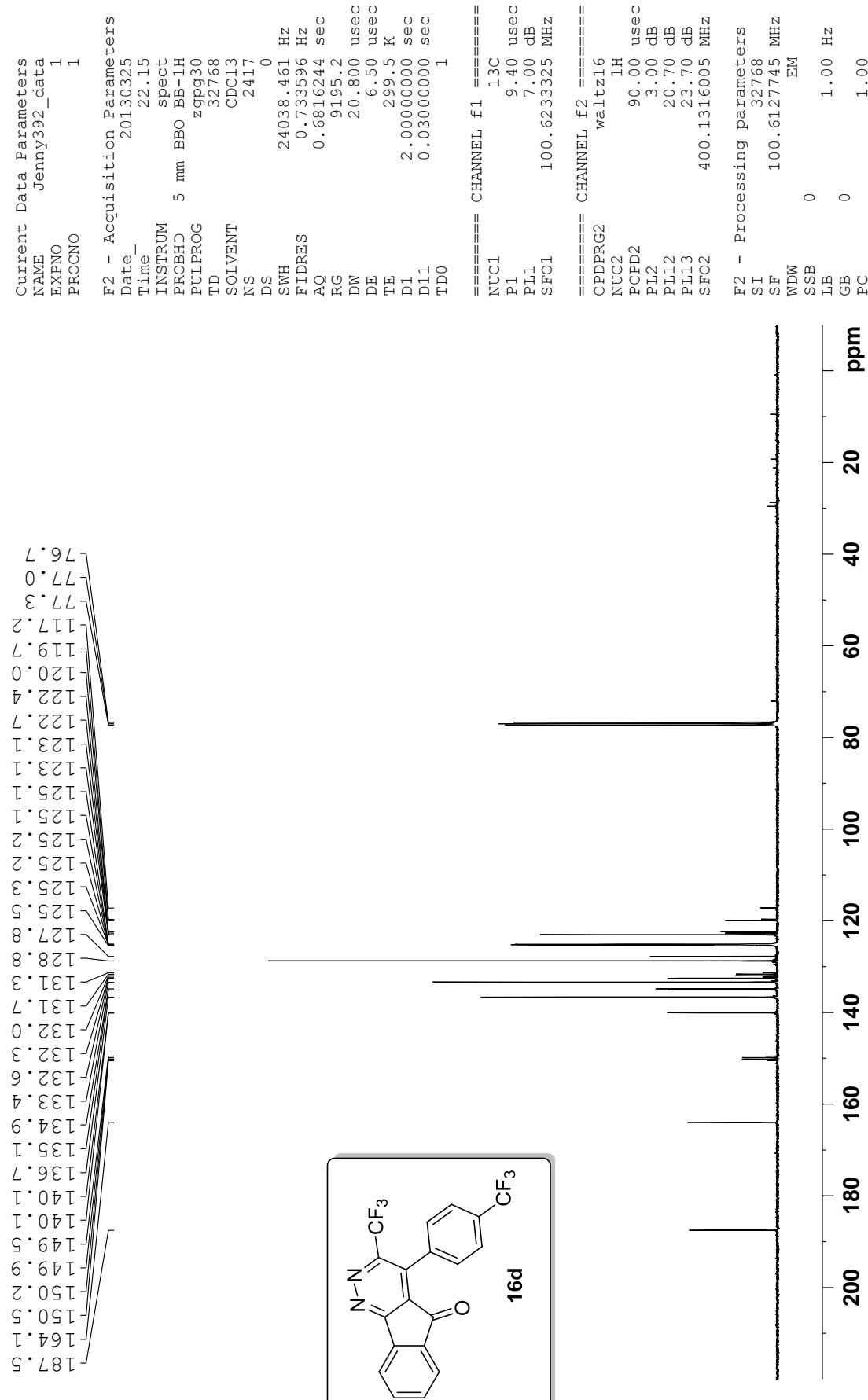




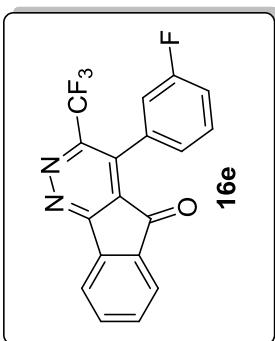
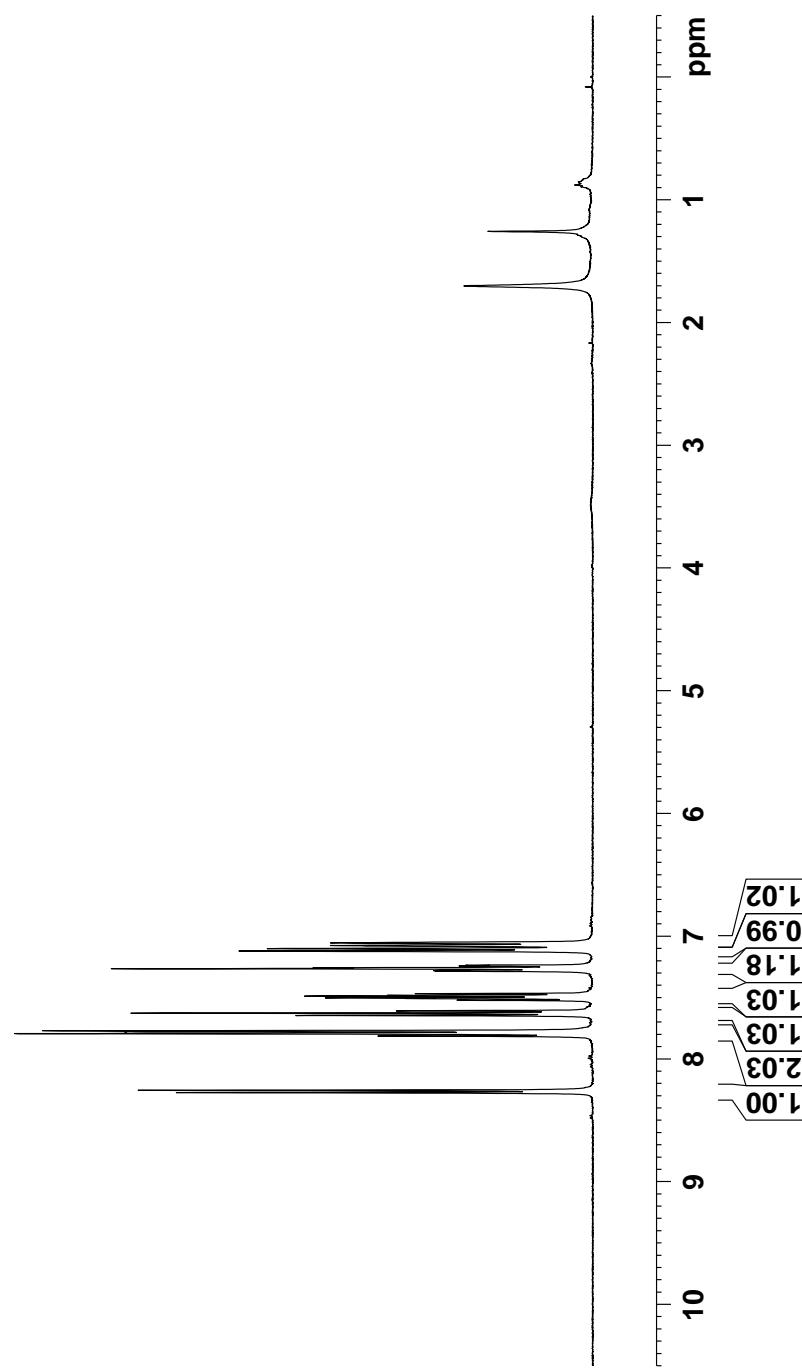


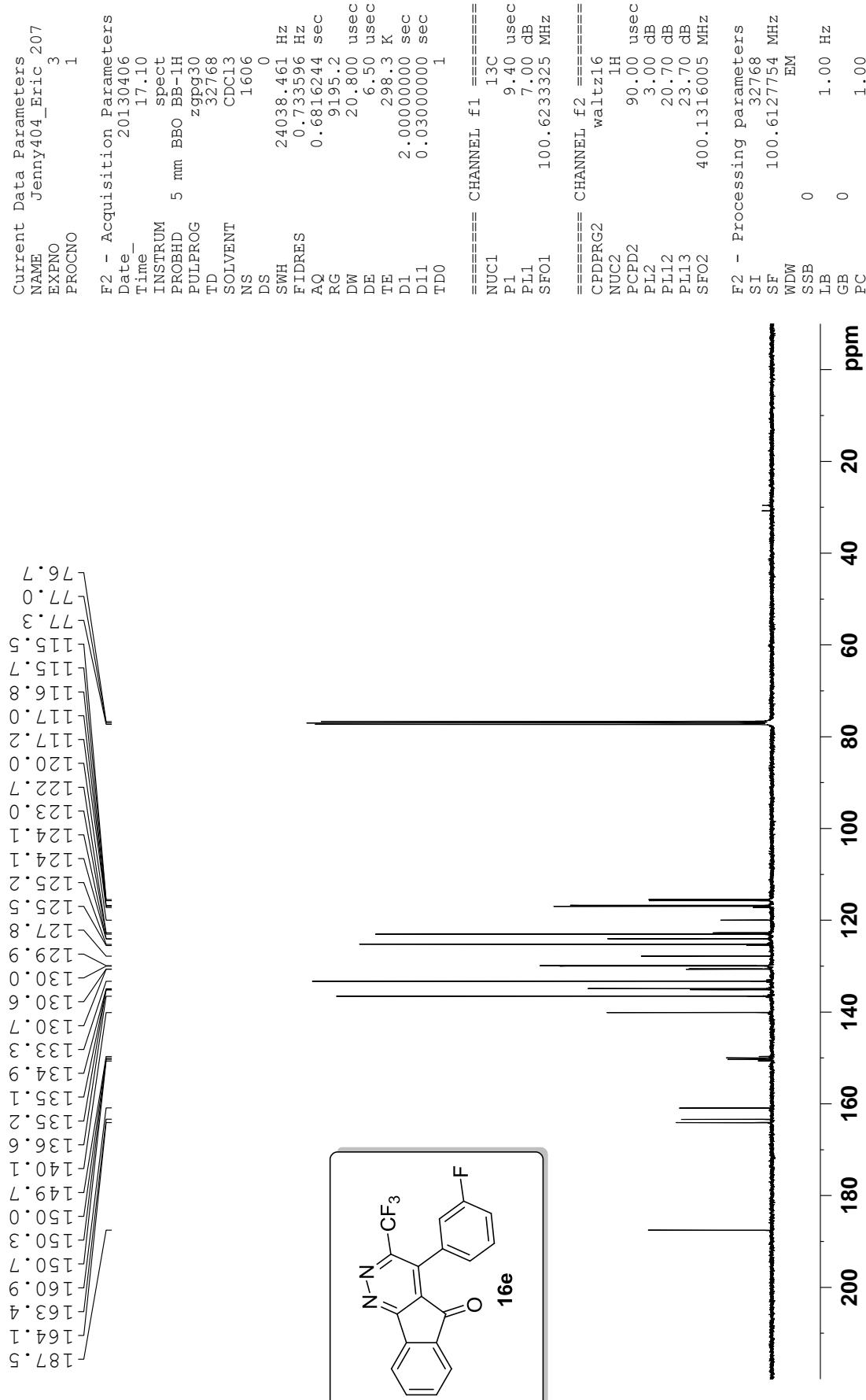






Current Data Parameters		F2 - Acquisition Parameters	
NAME	Jenny404_Eric 207	Date_	20130405
EXPNO	1	Time_	19.50
PROBHD	5 mm BBO BB-1H	INSTRUM	spect
PULPROG	zg30	TD	32768
SOLVENT	CDC13	NS	16
RG	0	DS	0
DW	7246.377 Hz	SWH	0.221142 Hz
DE	2.2610421 sec	FIDRES	181
TE		AQ	69.000 used
D1		RG	6.50 used
		DW	299.6 K
		DE	2.0000000 sec
		TE	TD0



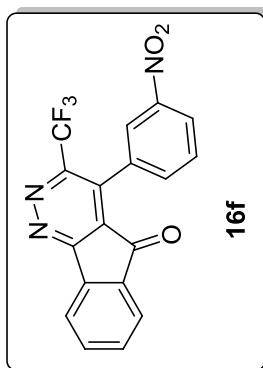
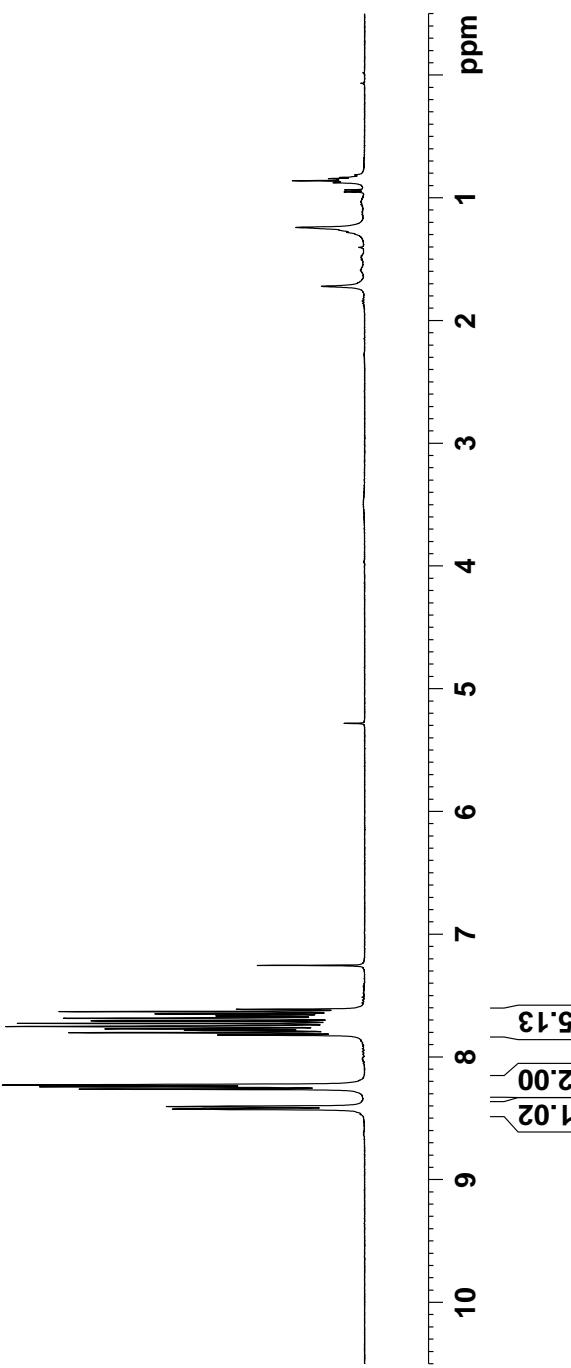


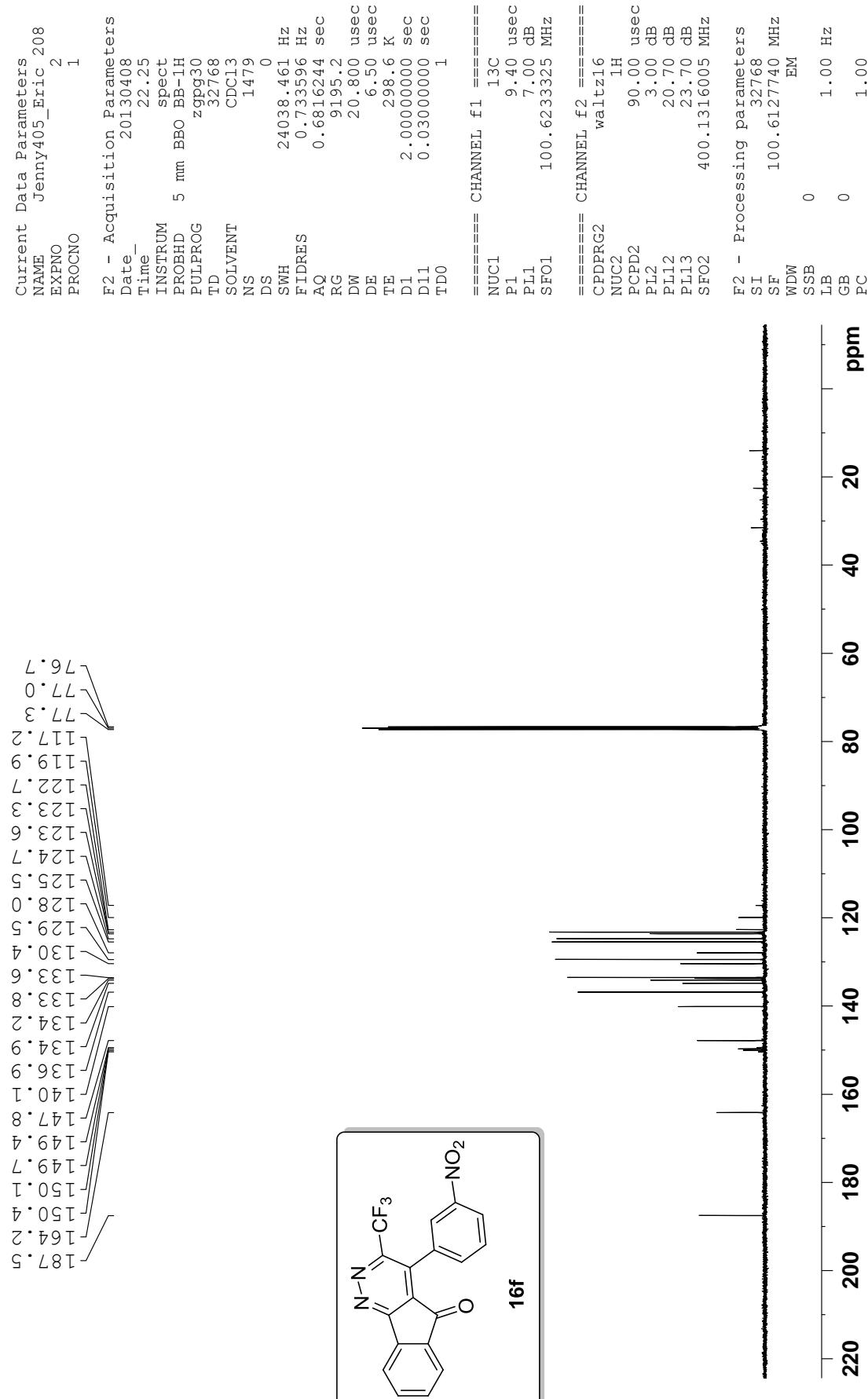
Current Data Parameters
NAME Jenry405_Eric 208
EXPNO 1
PROCNO 1

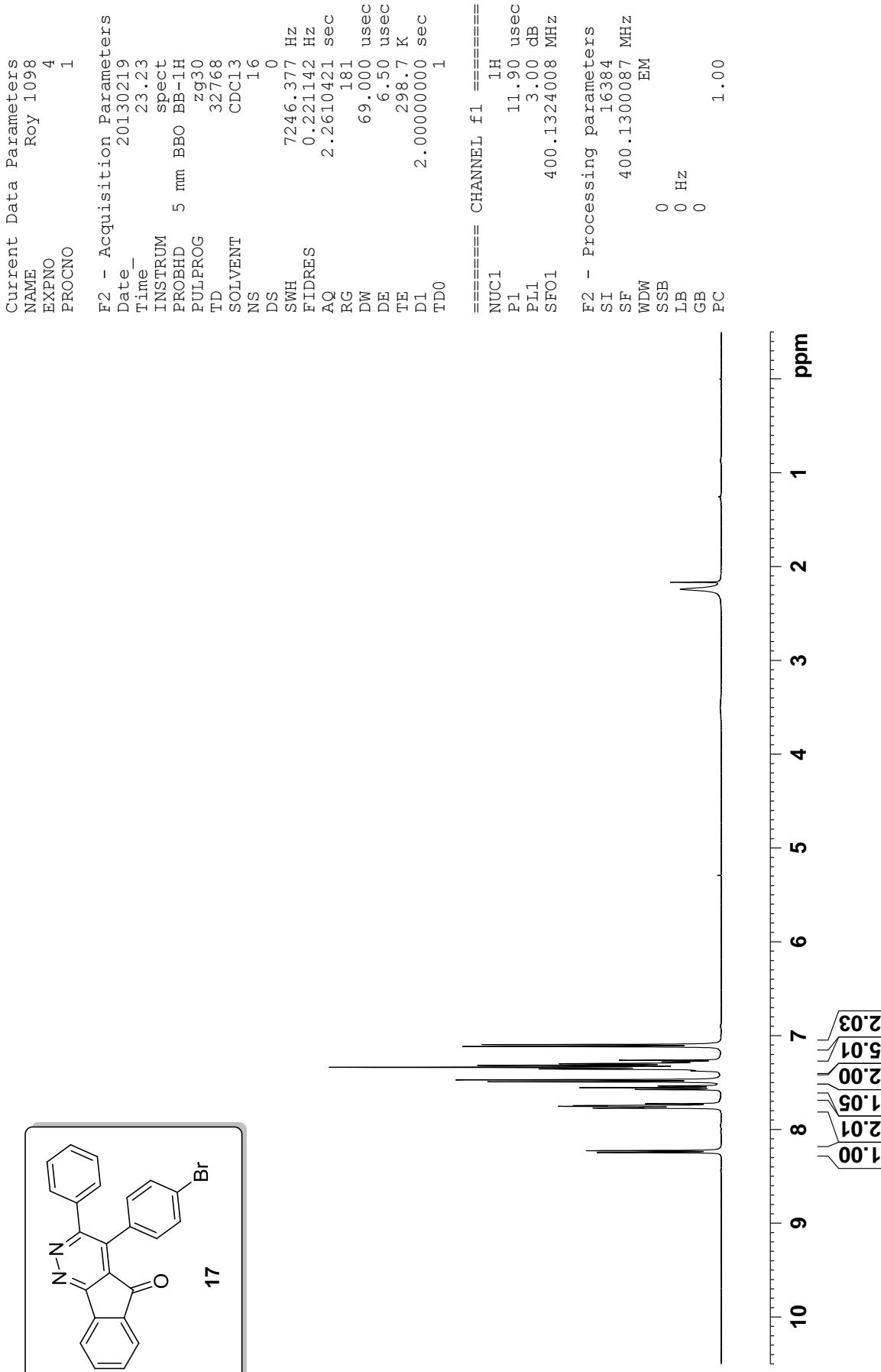
F2 - Acquisition Parameters
Date_ 20130408
Time_ 22.22
INSTRUM spect
PROBHD 5 mm BBO BB-1H
PULPROG zg30
TD 32768
SOLVENT CDCl3
NS 16
DS 0
SWH 7246.377 Hz
FIDRES 0.221142 Hz
AQ 2.2610421 sec
RG 114
DW 69.000 usec
DE 6.50 usec
TE 298.4 K
D1 2.0000000 sec
TDO 1

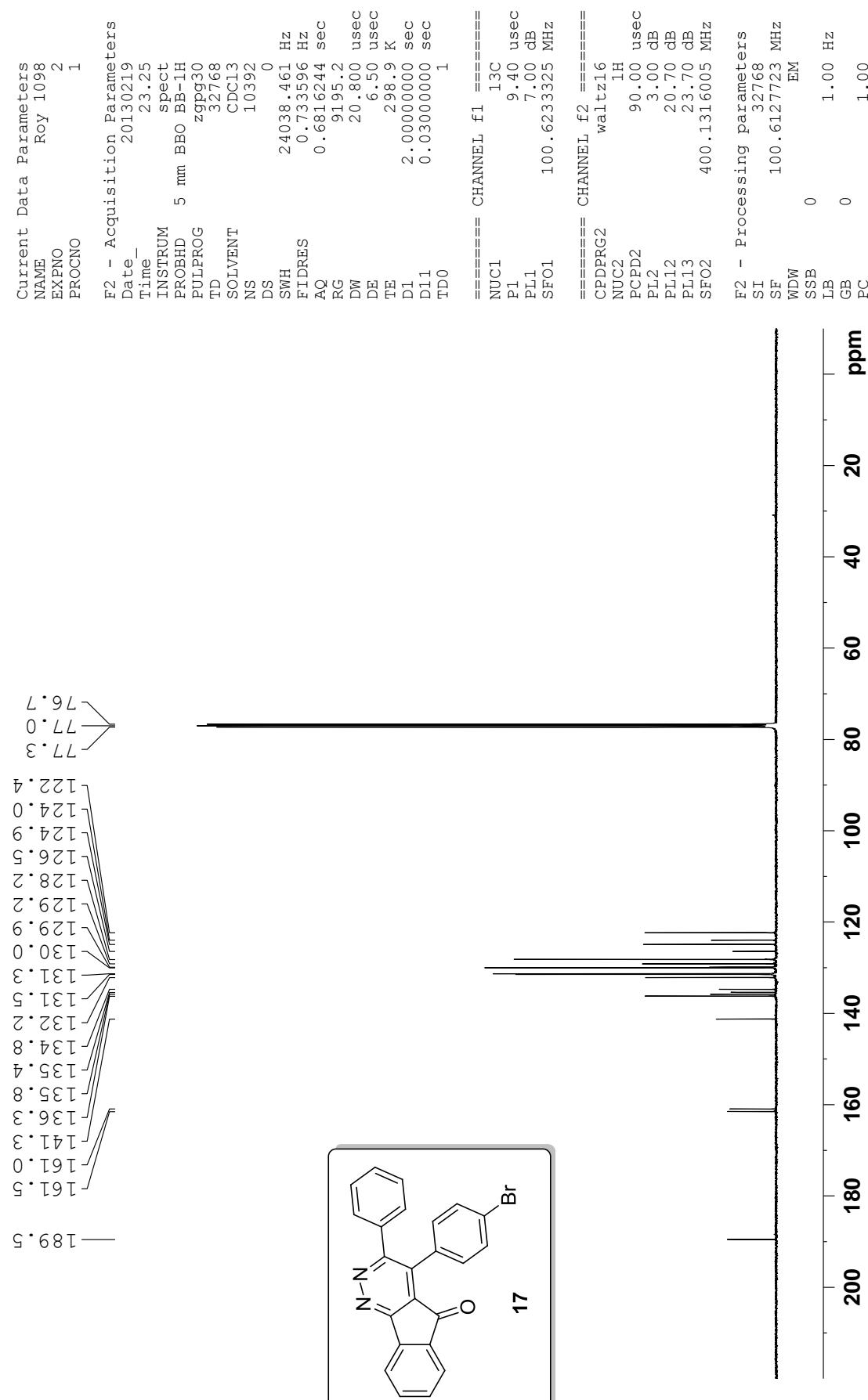
===== CHANNEL f1 =====
NUC1 1H
P1 11.90 usec
PL1 3.00 dB
SFO1 400.1324008 MHz

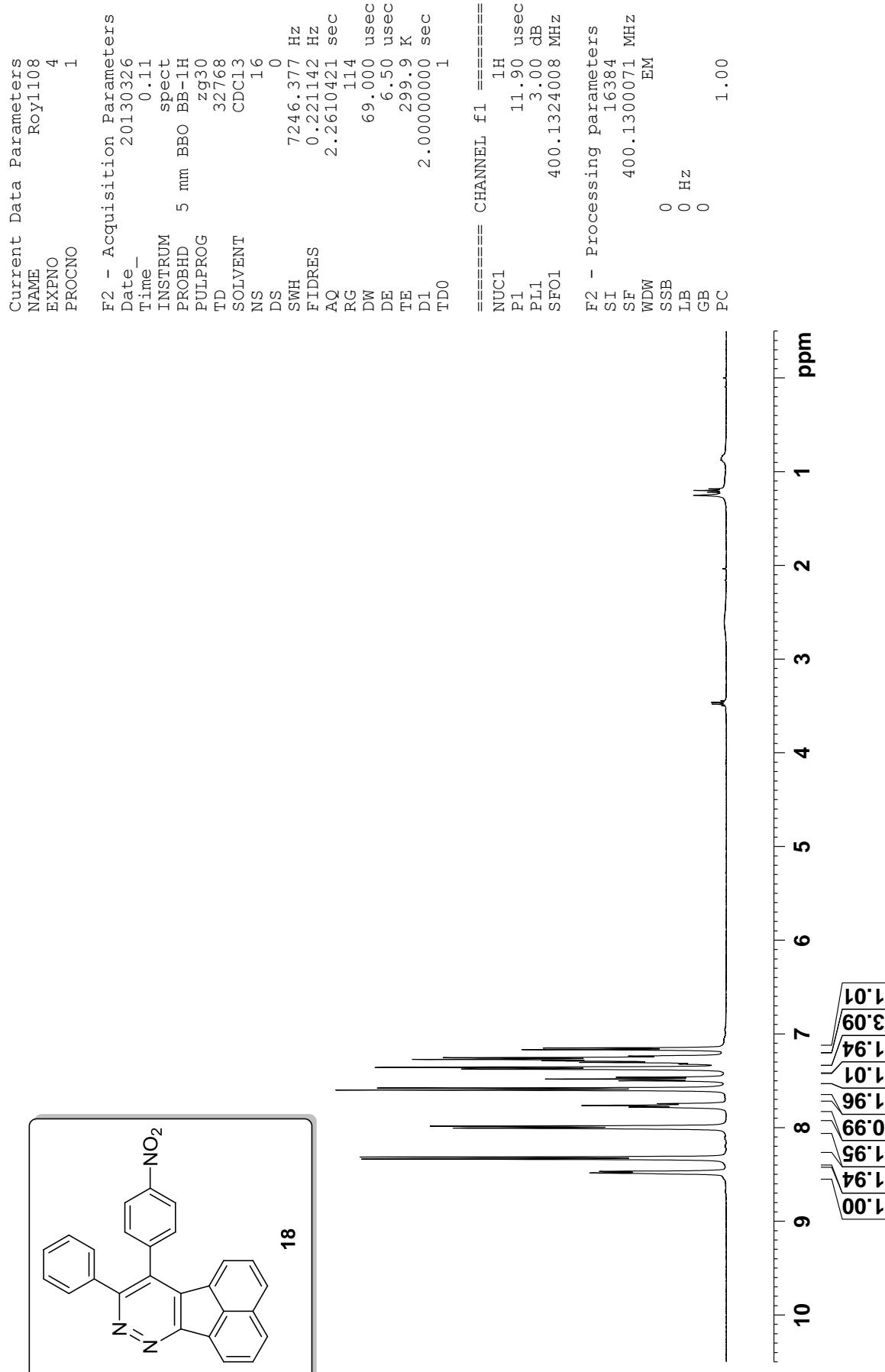
F2 - Processing parameters
SI 16384
SF 400.1300118 MHz
WDW EM
SSB 0
LB 0 Hz
GB 0
PC 1.00

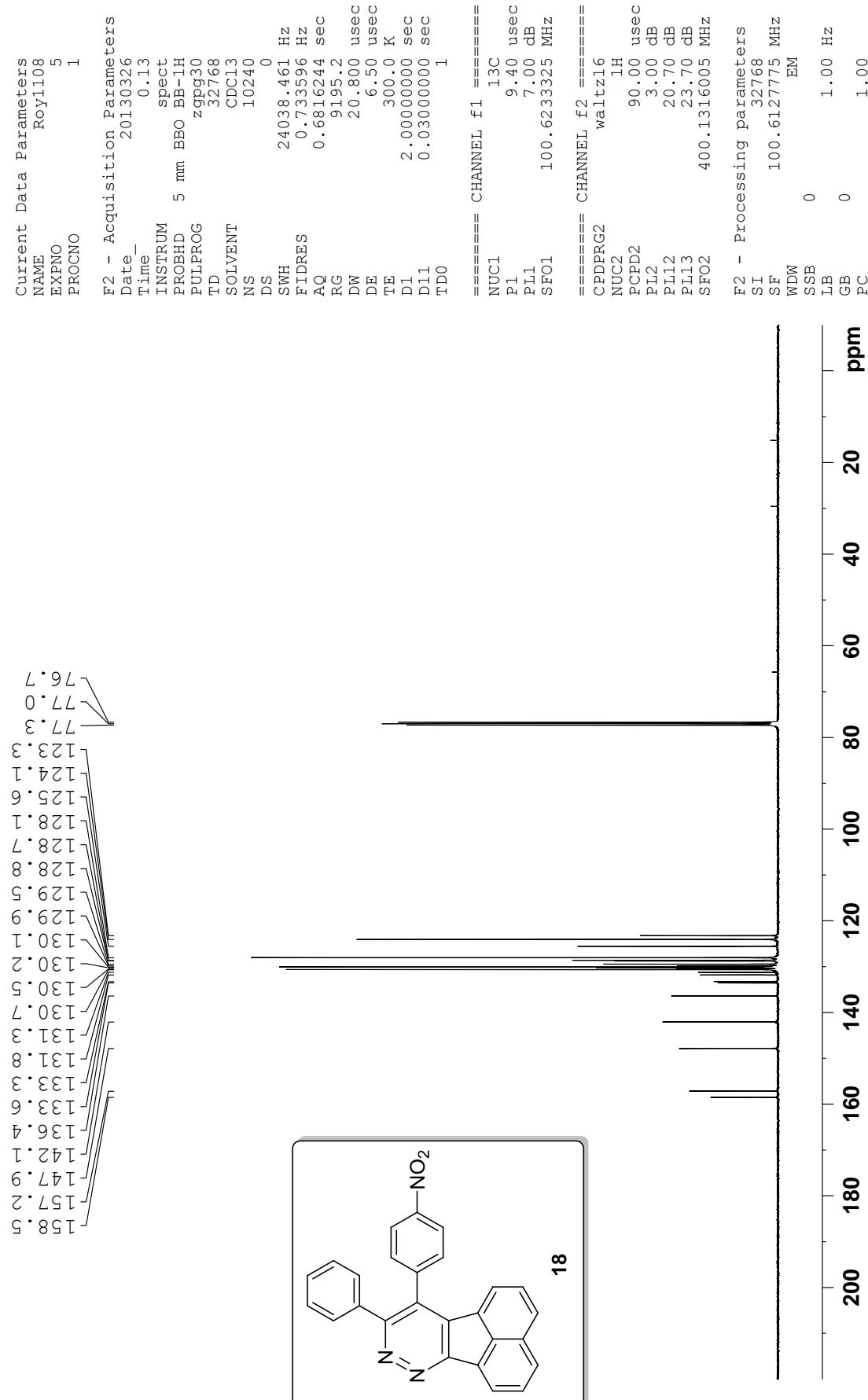


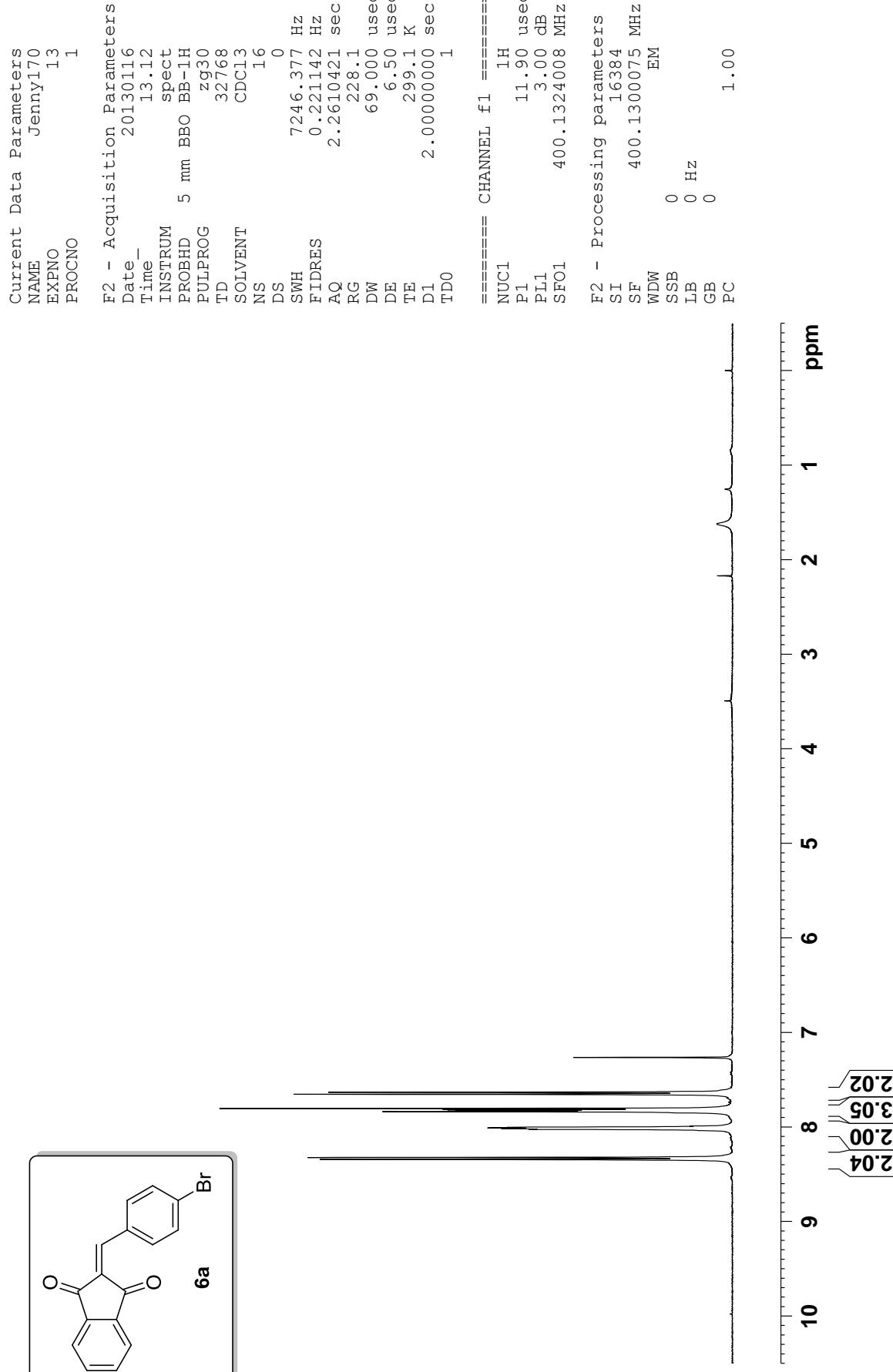


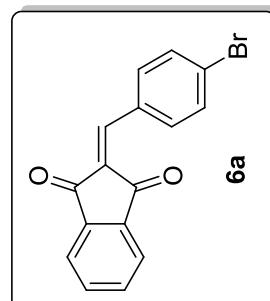
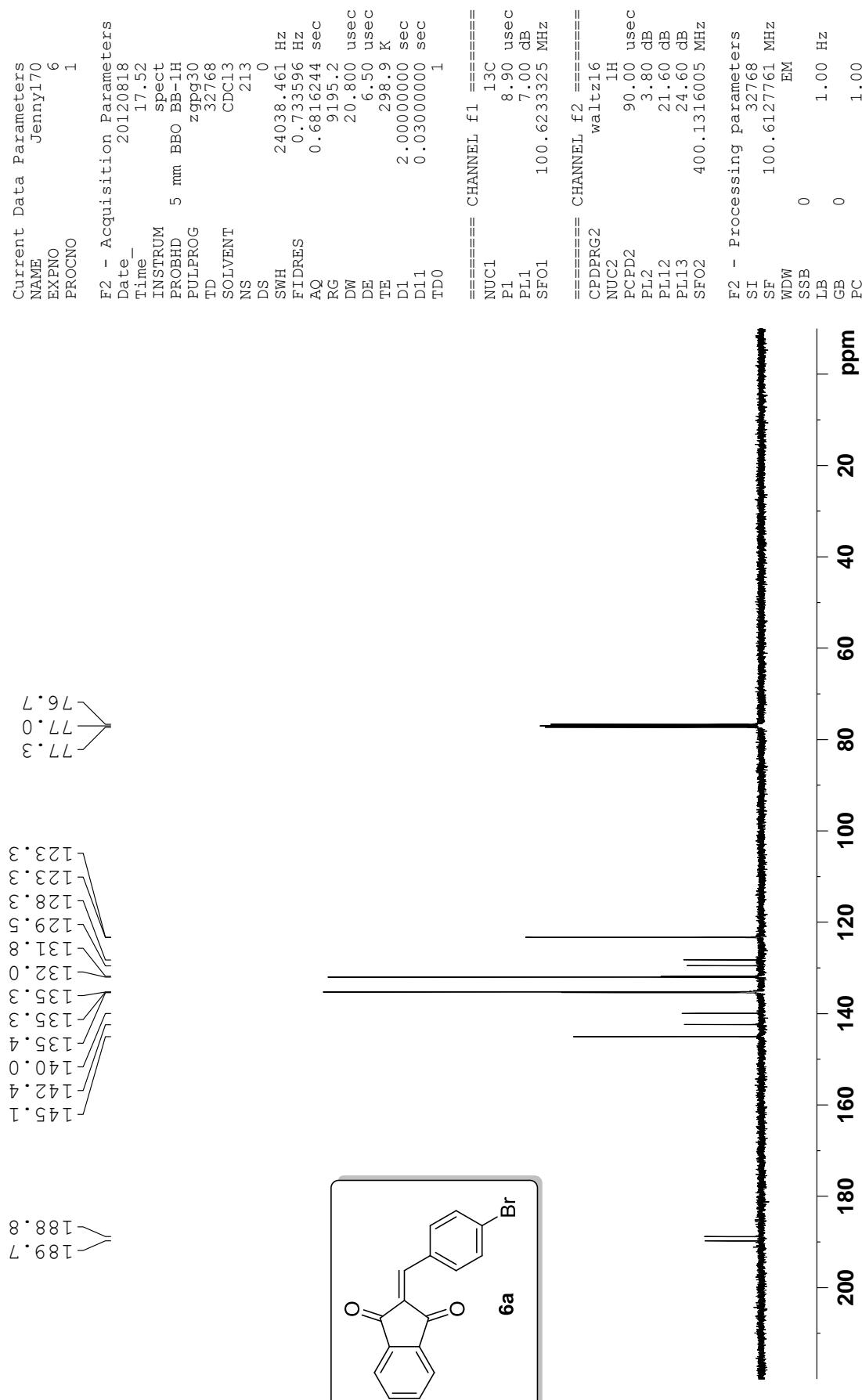












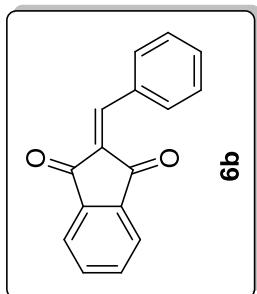
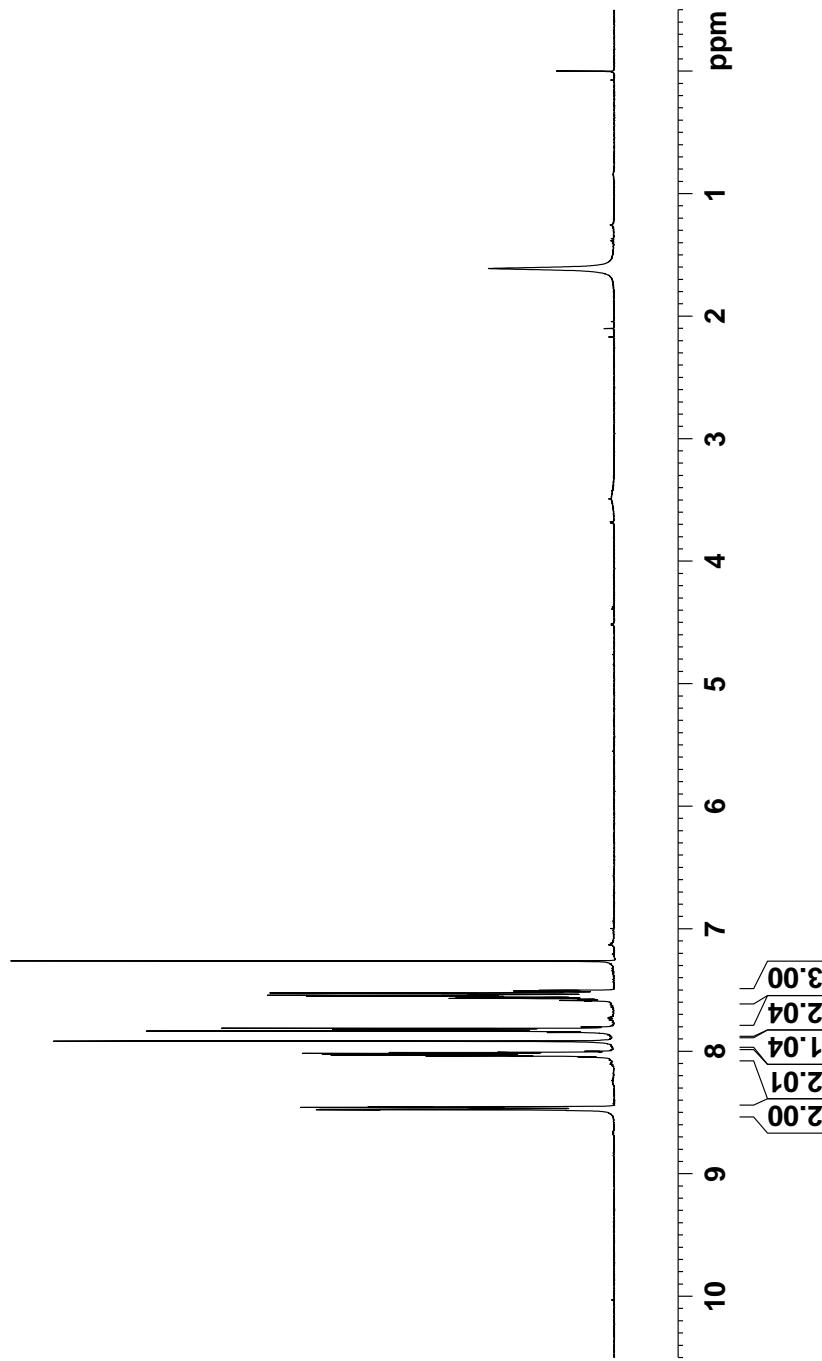
Current Data Parameters
NAME Jenny S.M of indandione
EXPNO 15
PROCNO 1

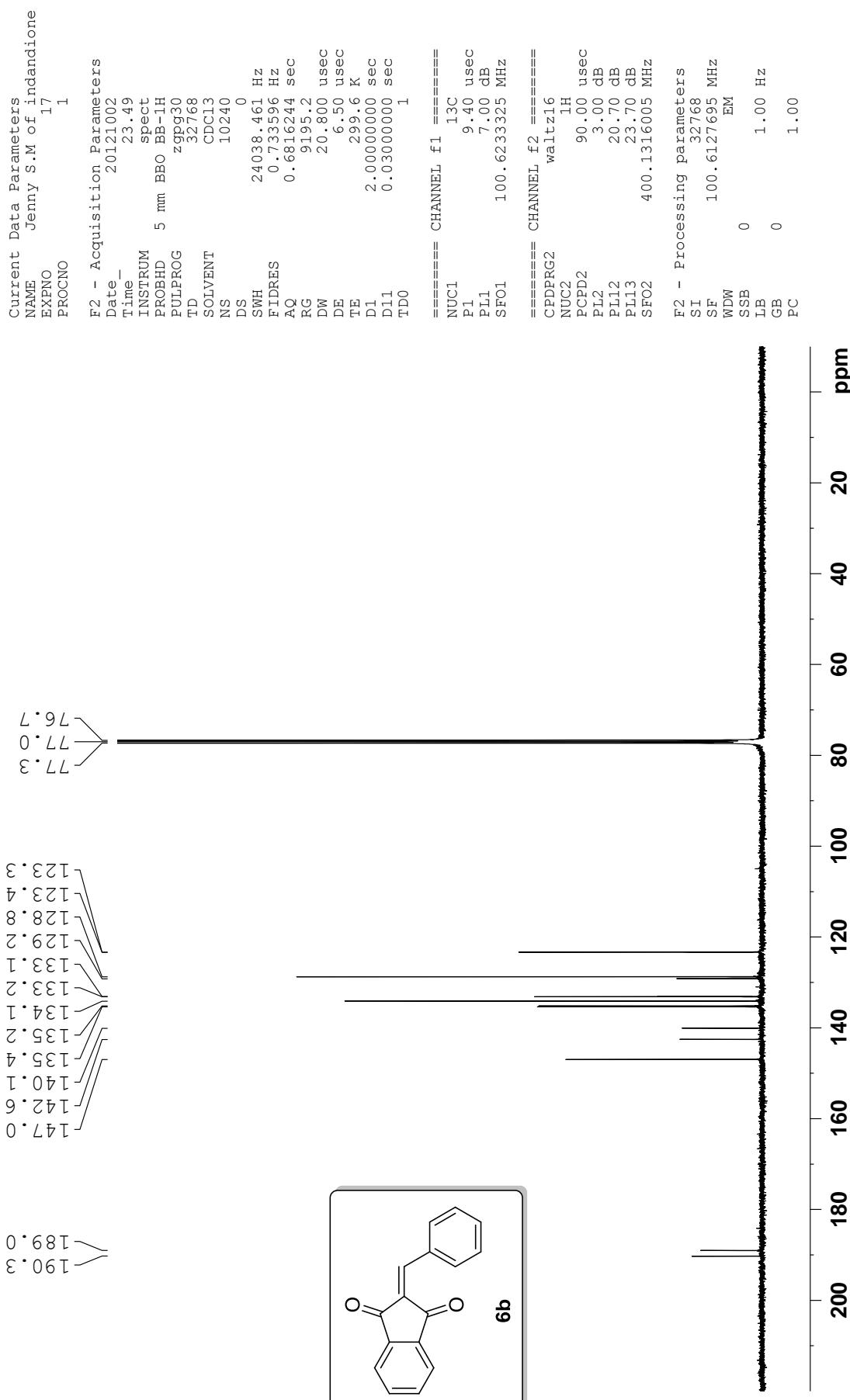
F2 - Acquisition Parameters

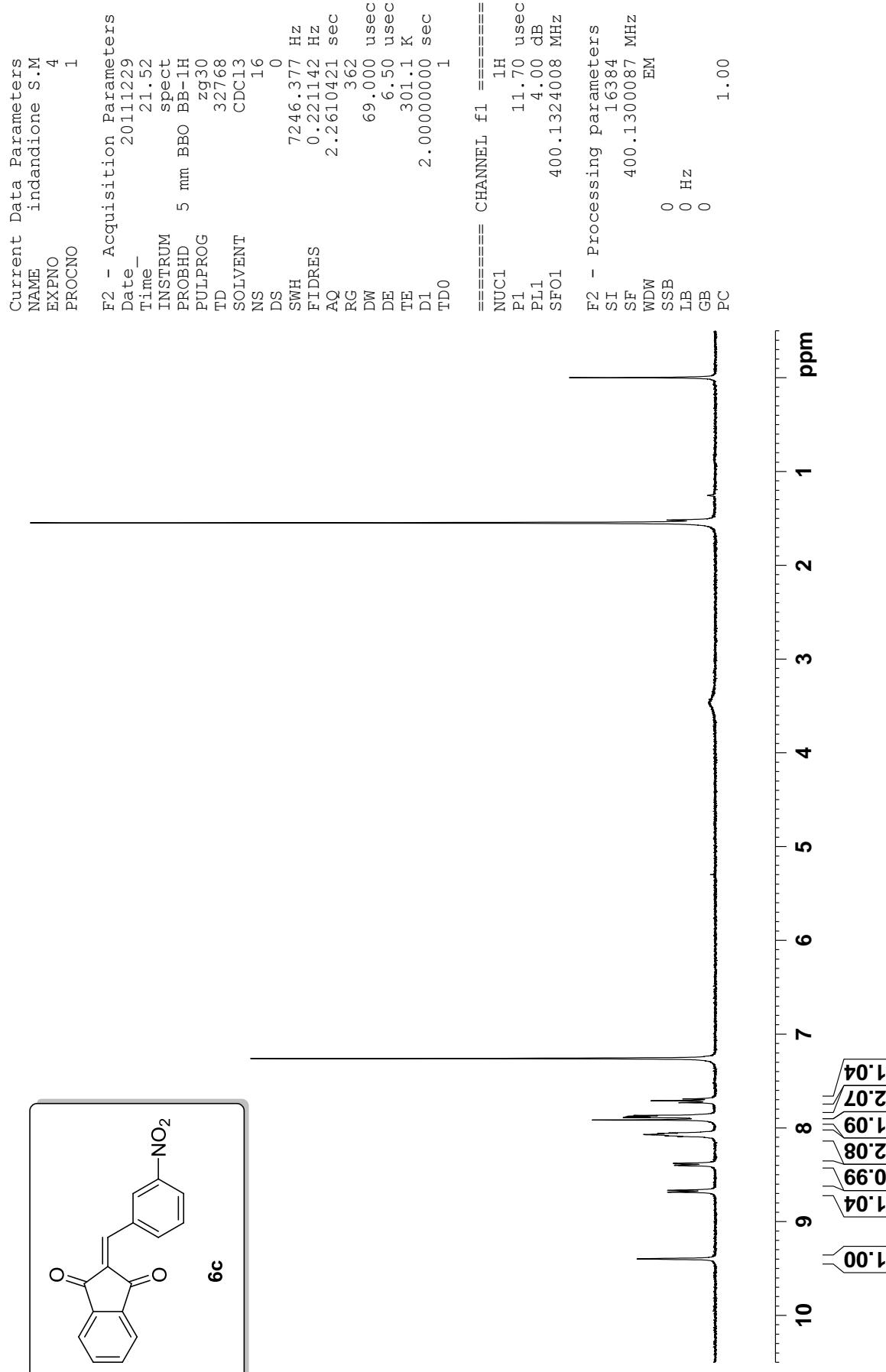
Date 2012/10/02
Time 23.46
INSTRUM spect
PROBHD 5 mm BBO BB-1H
PULPROG zg30
TD 32768
SOLVENT CDC13
NS 16
DS 0
SWH 7246.377 Hz
FIDRES 0.221142 Hz
AQ 2.2610421 sec
RG 256
DW 69.000 usec
DE 6.50 usec
TE 299.4 K
D1 2.00000000 sec
TDO 1

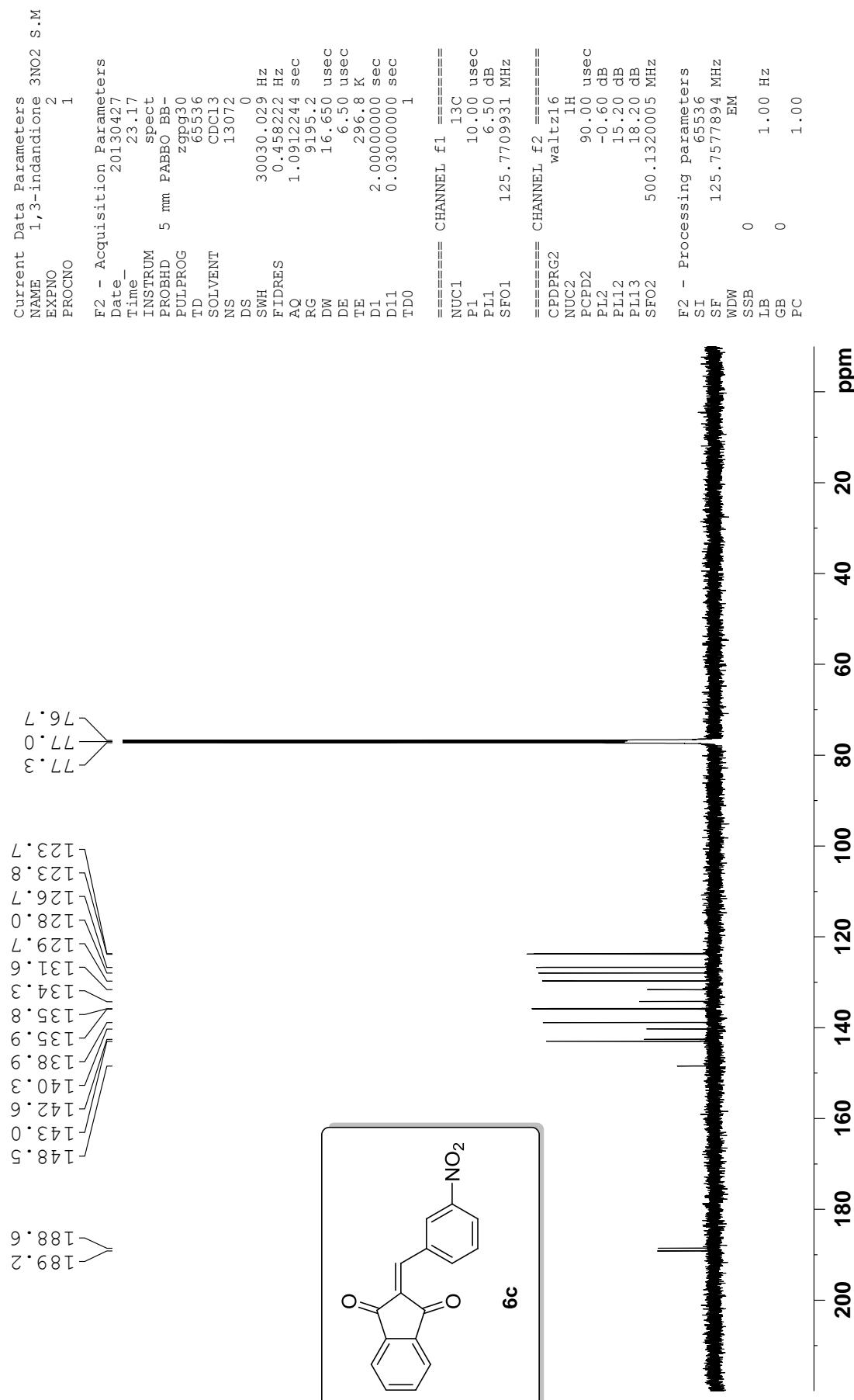
===== CHANNEL f1 =====

NUC1 1H
P1 11.90 usec
PL1 3.00 dB
SFO1 400.1324008 MHz
F2 - Processing parameters
SI 16384
SF 400.1300083 MHz
WDW EM
SSB 0
LB 0 Hz
GB 0
PC 1.00









Current Data Parameters
 NAME Jenny S.M._Indandione
 EXPNO 29
 PROCNO 1

F2 - Acquisition Parameters

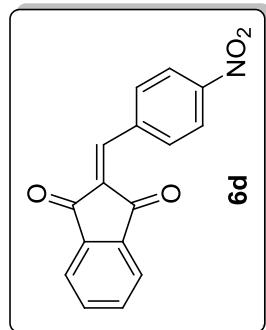
Date	20130417
Time	5.16
INSTRUM	5 mm BBO
PROBHD	spec
PULPROG	BB-1H
TD	zg30
SOLVENT	322768
NS	CDC13
DS	16
SWH	0
FIDRES	7246.377 Hz
AQ	0.221142 Hz
RG	2.2610421 sec
DW	322.5
DE	69.000 usec
TE	6.50
D1	299.7 K
TD0	2.00000000 sec

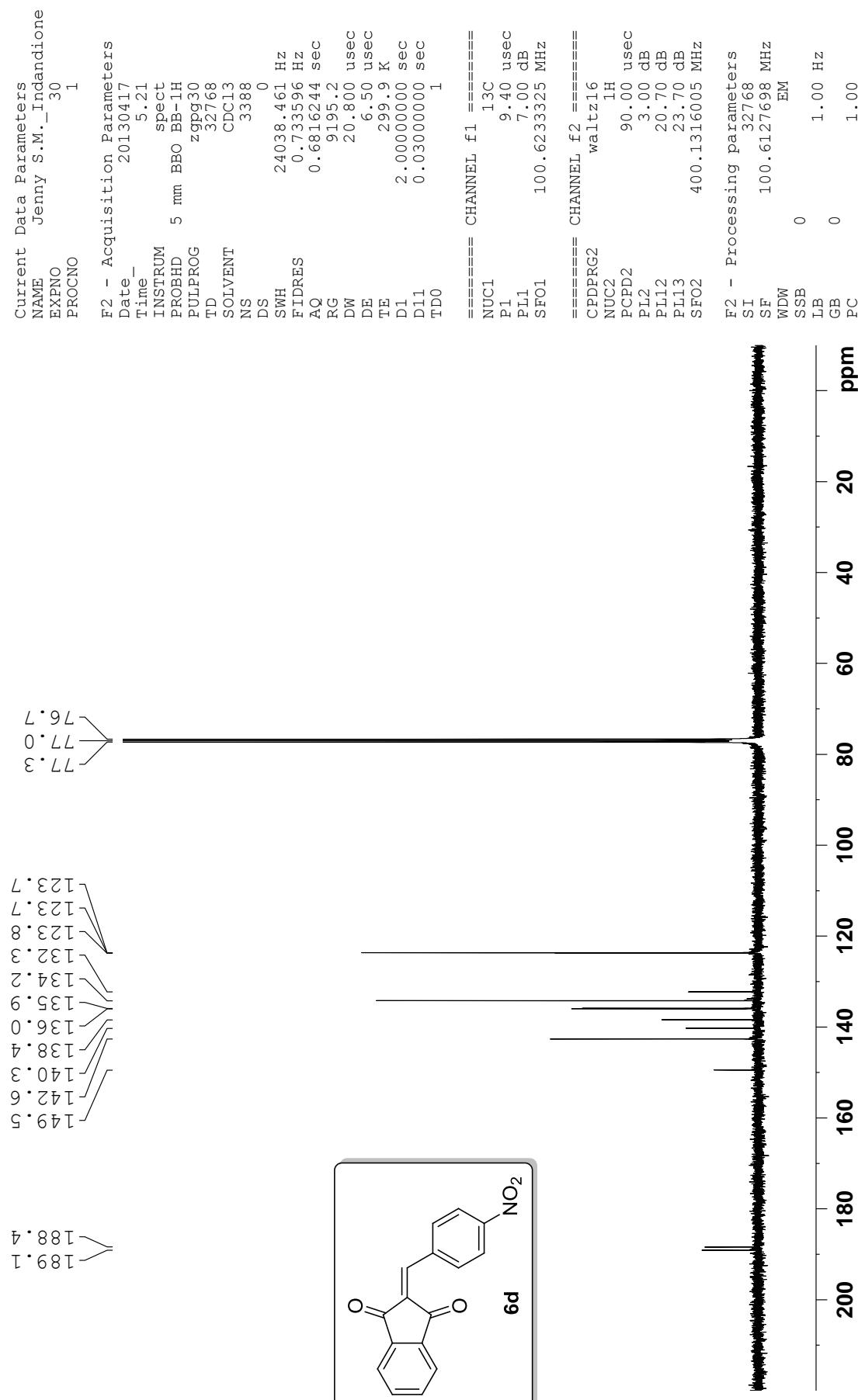
```

===== CHANNEL f1 =====
NDCL          1H
P1           11.90 usec
PLL          3.00 dB
SF01         400.1324008 MHz
                           EM

F2 - Processing parameters
SI           16384
SF          400.1300078 MHz
WDW
SSB          0 Hz
LB           0 Hz
GB          0
PC

```





Current Data Parameters
NAME Jenny S.M of indandione
EXPNO 3
PROCNO 1

F2 - Acquisition Parameters

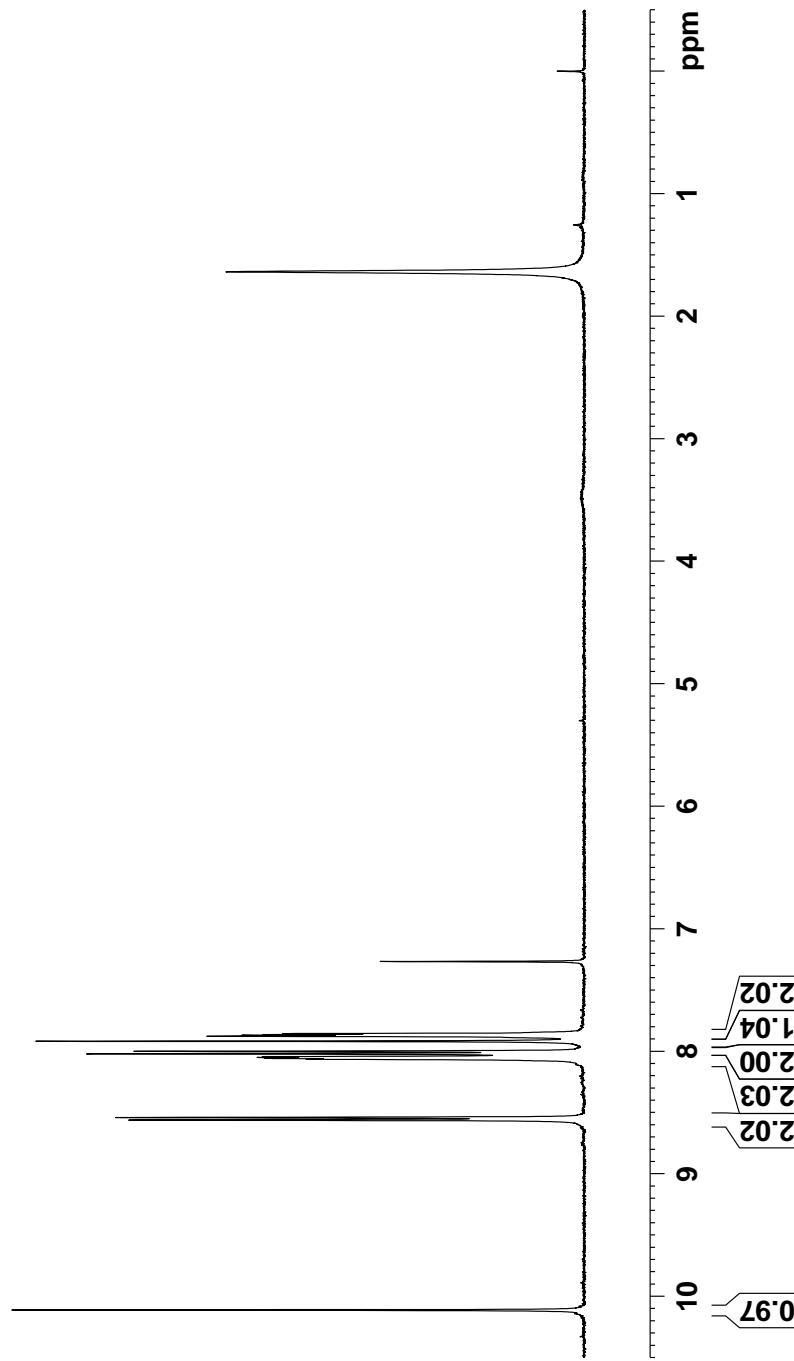
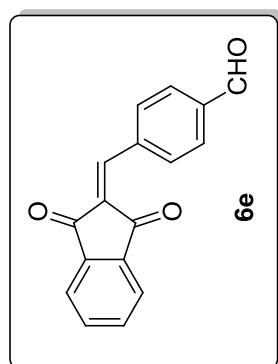
Date 20120907
Time 22.06
INSTRUM spect
PROBHD 5 mm BBO BB-1H
PULPROG zg30
TD 32768
SOLVENT CDCl₃
NS 8
DS 0
SWH 7246.377 Hz
FIDRES 0.221142 Hz
AQ 2.2610421 sec
RG 256
DW 69.000 usec
DE 6.50 usec
TE 298.9 K
D1 2.00000000 sec
TDO 1

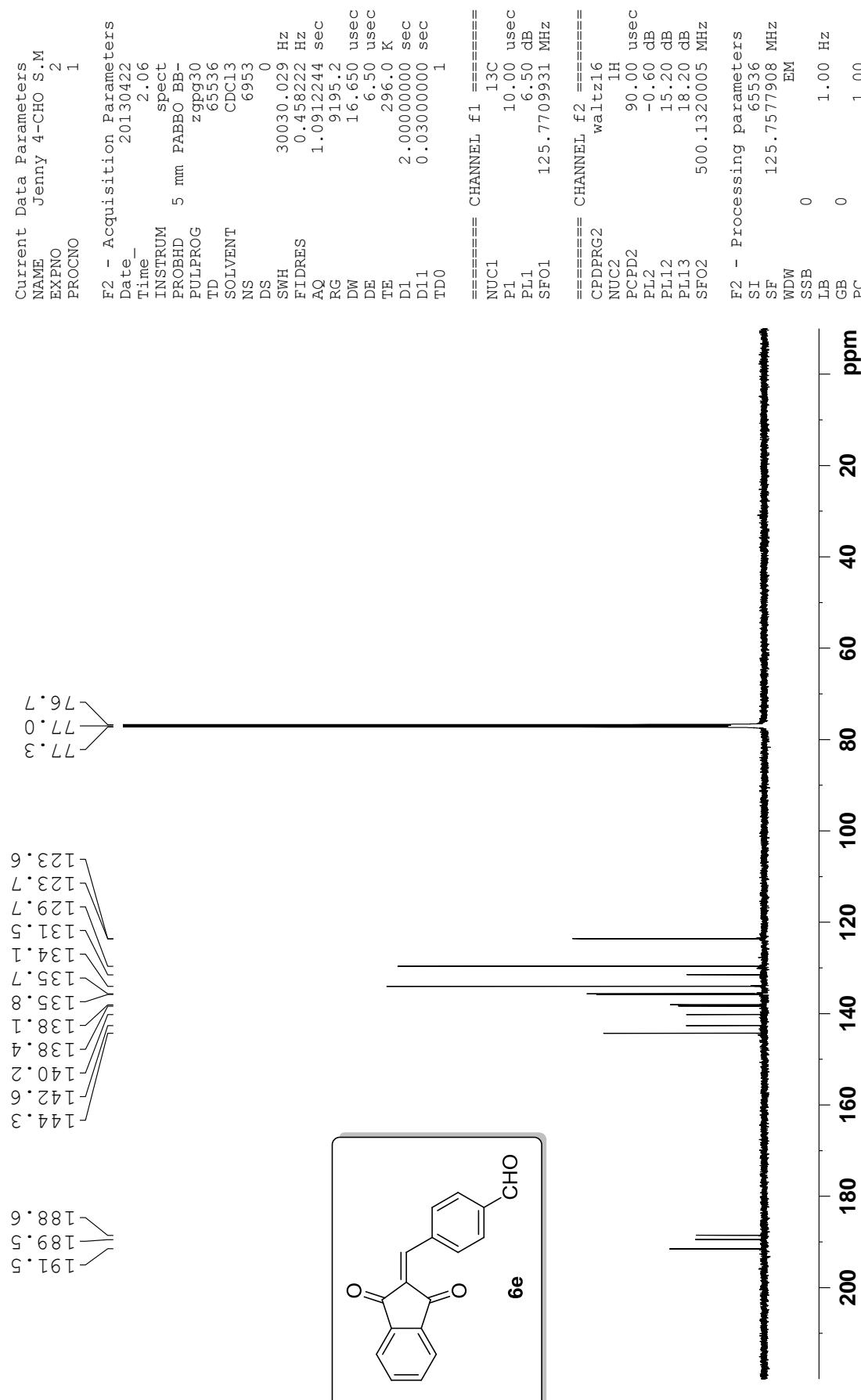
===== CHANNEL f1 =====

NUC1 1H
P1 11.70 usec
PL1 4.00 dB
SFO1 400.1324008 MHz

F2 - Processing parameters

SI 16384
SF 400.1300054 MHz
WDW EM
SSB 0
LB 0 Hz
GB 0
PC 1.00





Current Data Parameters
NAME Jenny S.M._Indandione
EXPNO 16
PROCNO 1

F2 - Acquisition Parameters

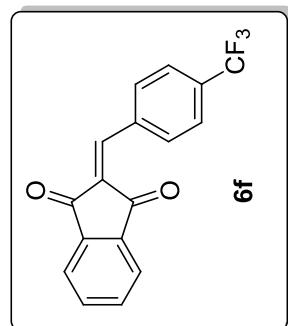
Date 20121108
Time 7.33
INSTRUM spect
PROBHD 5 mm BBO BB-1H
PULPROG zg30
TD 32768
SOLVENT CDCl₃
NS 16
DS 0
SWH 7246.377 Hz
FIDRES 0.221142 Hz
AQ 2.2610421 sec
RG 362
DW 69.000 usec
DE 6.50 usec
TE 299.1 K
D1 2.0000000 sec
TDO 1

===== CHANNEL f1 =====

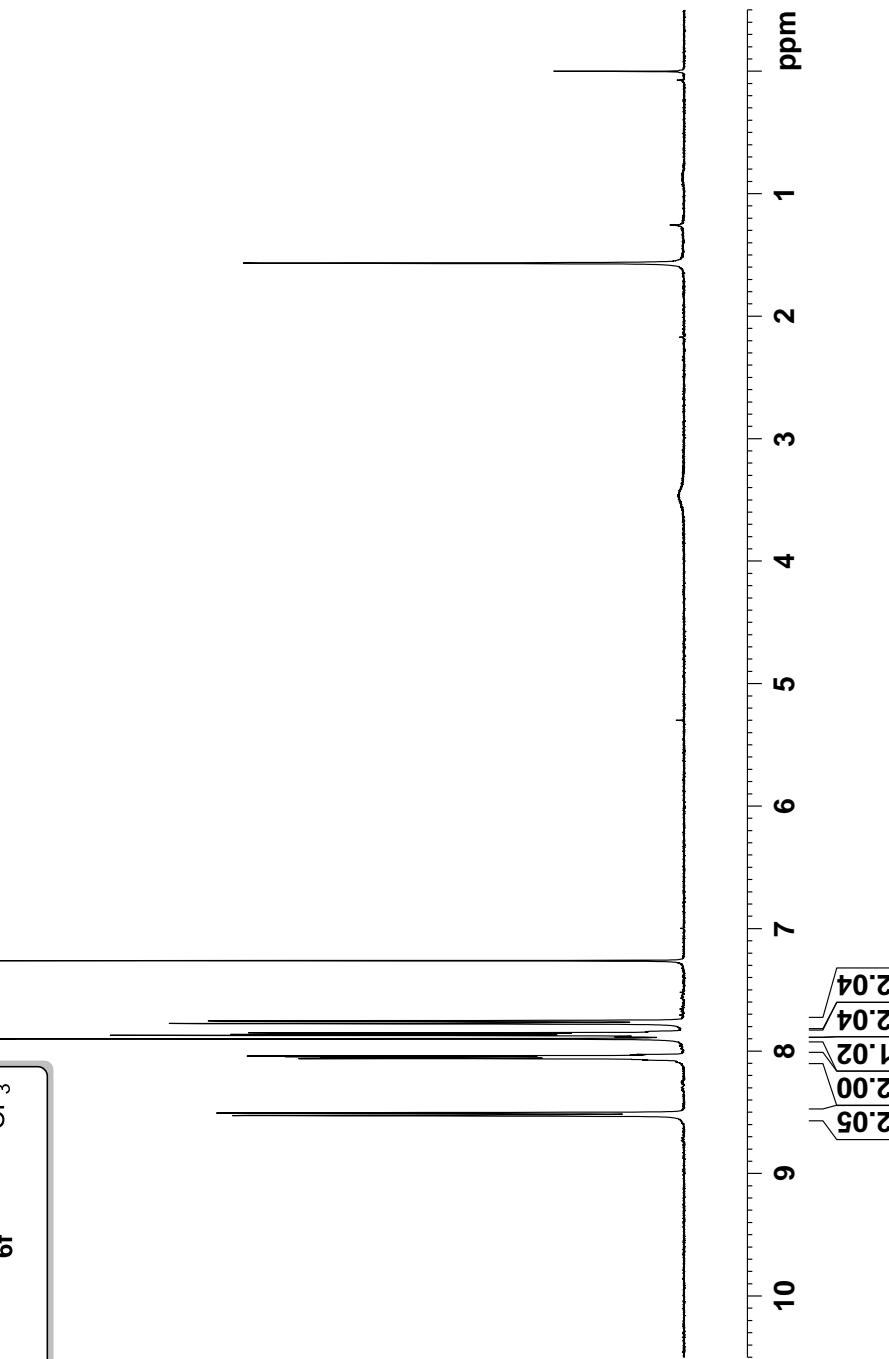
NUC1 1H
P1 11.90 usec
PL1 3.00 dB
SFO1 400.1324008 MHz
WDW EM
SSB 0 Hz
LB 0 Hz
GB 0
PC 1.00

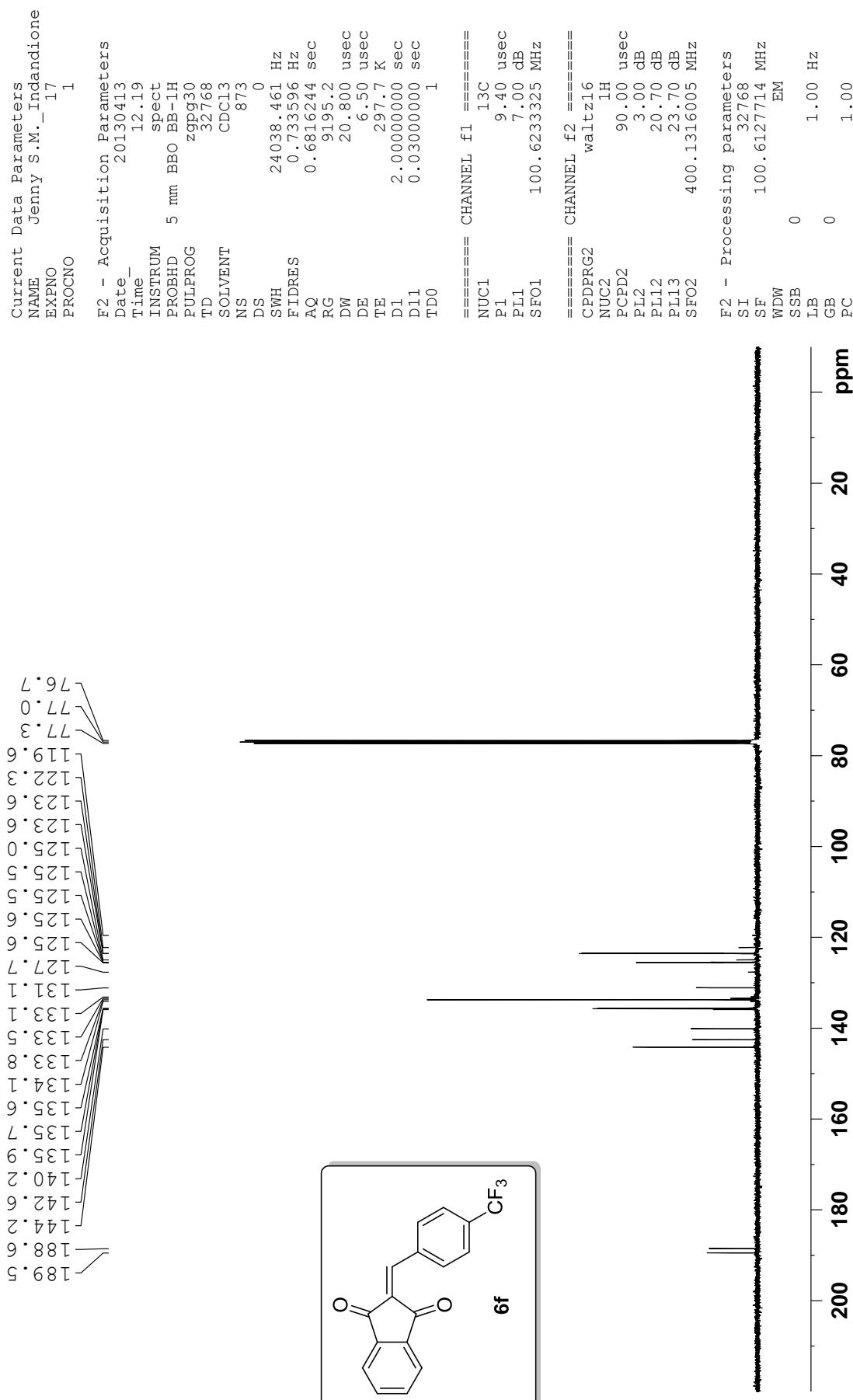
F2 - Processing parameters

SI 16384
SF 400.1300085 MHz
WDW EM
SSB 0 Hz
LB 0 Hz
GB 0
PC 1.00



6f





Current Data Parameters
 NAME Jenny S.M._Indandione
 EXPNO 13
 PROCNO 1

F2 - Acquisition Parameters

Date	20121105
Time	12.35
INSTRUM	5 mm BBO BB-1H
PROBHD	ZG30
PULPROG	32768
TD	CDC13
SOLVENT	16
NS	0
DS	7246.377 Hz
SWH	0.221142 sec
FIDRES	2.2610421 sec
AQ	322.5
RG	69.000 usec
DW	6.50 usec
DE	298.5 K
TE	2.00000000 sec
D1	
TDDO	1

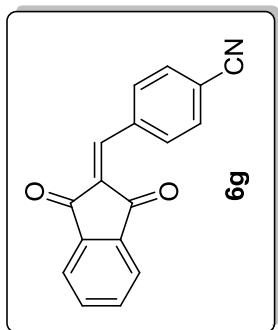
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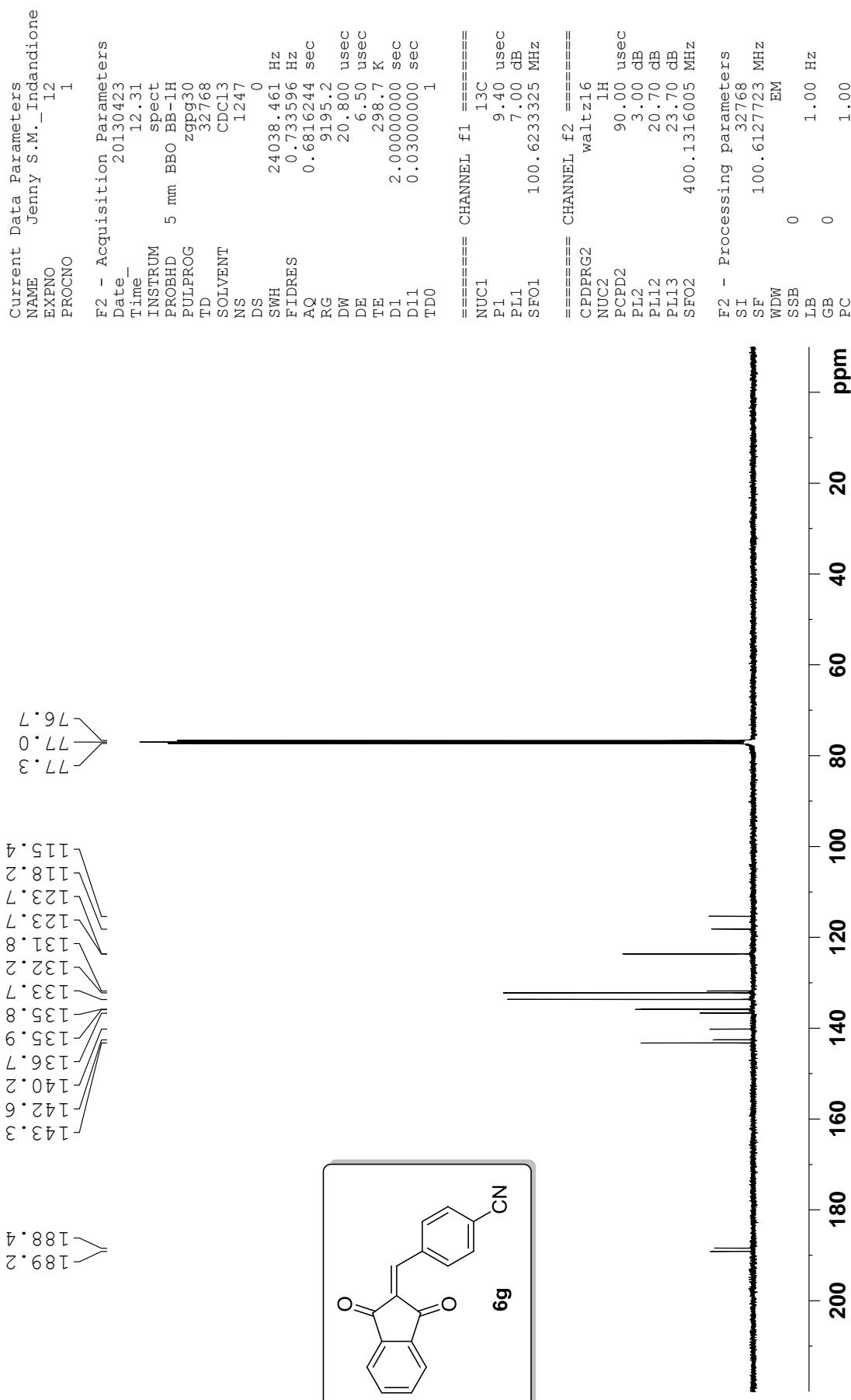
=====
 CHANNEL f1 =====
 NUC1.          1 H
 P1            11.90 usec
 PLL           3.00 dB
 SF01.         400.1324008 MHz
                           EM

F2 - Processing parameters
SI            16384
SF           400.1300078 MHz
WDW
SSB           0 Hz
LB            0 Hz
GB
PC

```

The figure displays a ^1H NMR spectrum. The x-axis is labeled "ppm" and ranges from 1 to 10. Key peaks are labeled with their chemical shifts: a sharp peak at 1.00 ppm, a broad peak at 2.00 ppm, a peak at 2.02 ppm, a peak at 3.01 ppm, and a sharp peak at 7.02 ppm.





Current Data Parameters
NAME Jenny S.M._Indandione
EXPNO 49
PROCNO 1

F2 - Acquisition Parameters

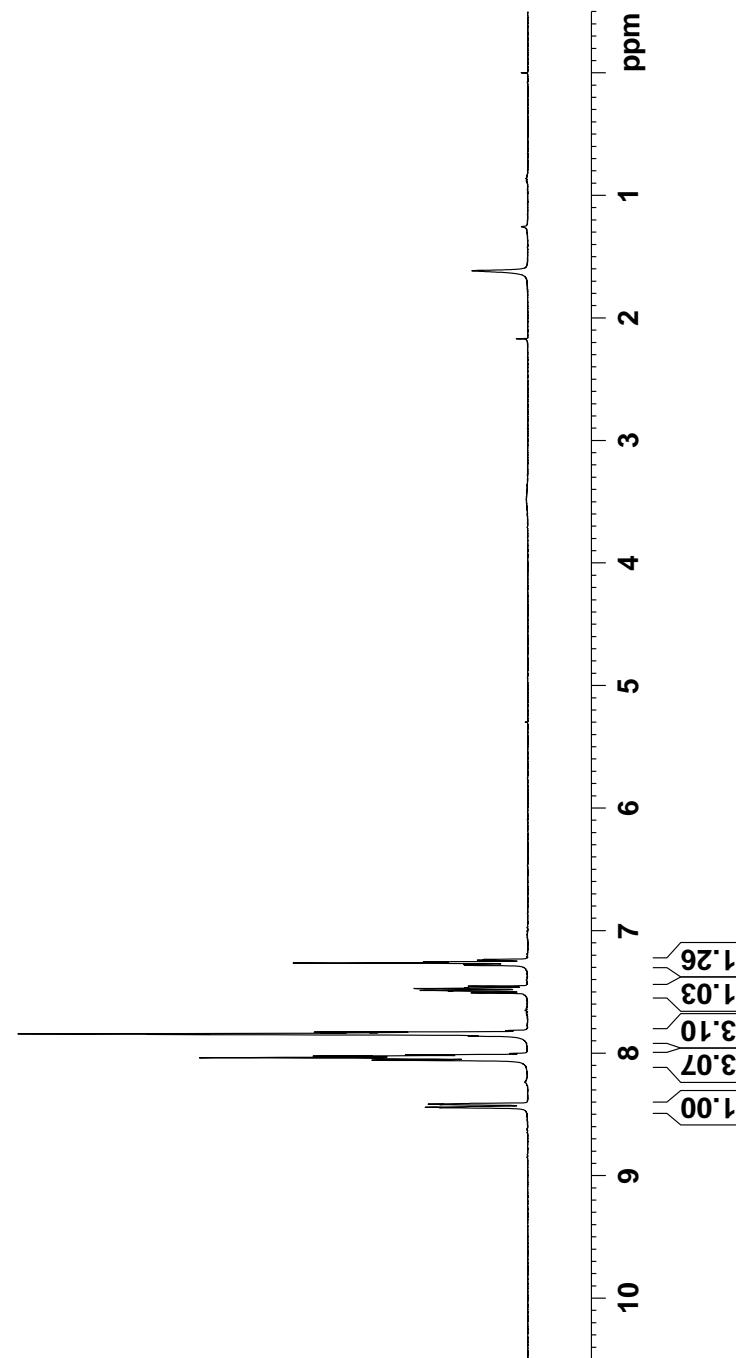
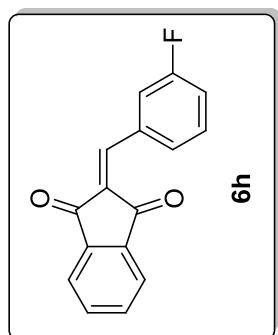
Date 20130430
Time 23.10
INSTRUM spect
PROBHD 5 mm BBO BB-1H
PULPROG zg30
TD 32768
SOLVENT CDCl3
NS 16
DS 0
SWH 7246.377 Hz
FIDRES 0.221142 Hz
AQ 2.2610421 sec
RG 256
DW 69.000 usec
DE 6.50 usec
TE 301.0 K
D1 2.0000000 sec
TDO 1

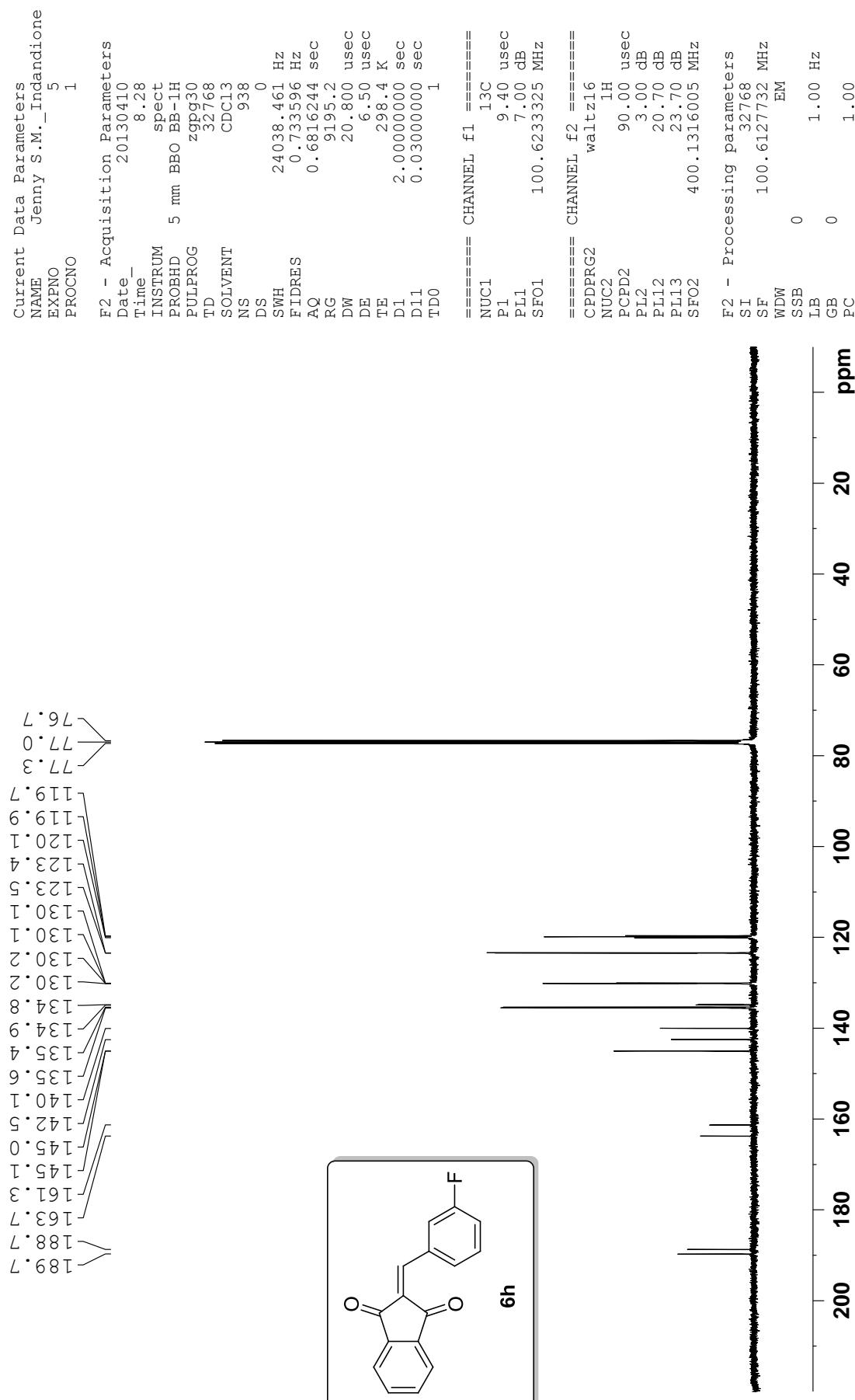
===== CHANNEL f1 =====

NUC1 1H
P1 11.90 usec
PL1 3.00 dB
SFO1 400.1324008 MHz
EM

F2 - Processing parameters

SI 16384
SF 400.1300074 MHz
WDW EM
SSB 0 Hz
LB 0 Hz
GB 0
PC 1.00





Current Data Parameters
NAME Jenny S.M of indandione
EXPNO 18
PROCNO 1

F2 - Acquisition Parameters

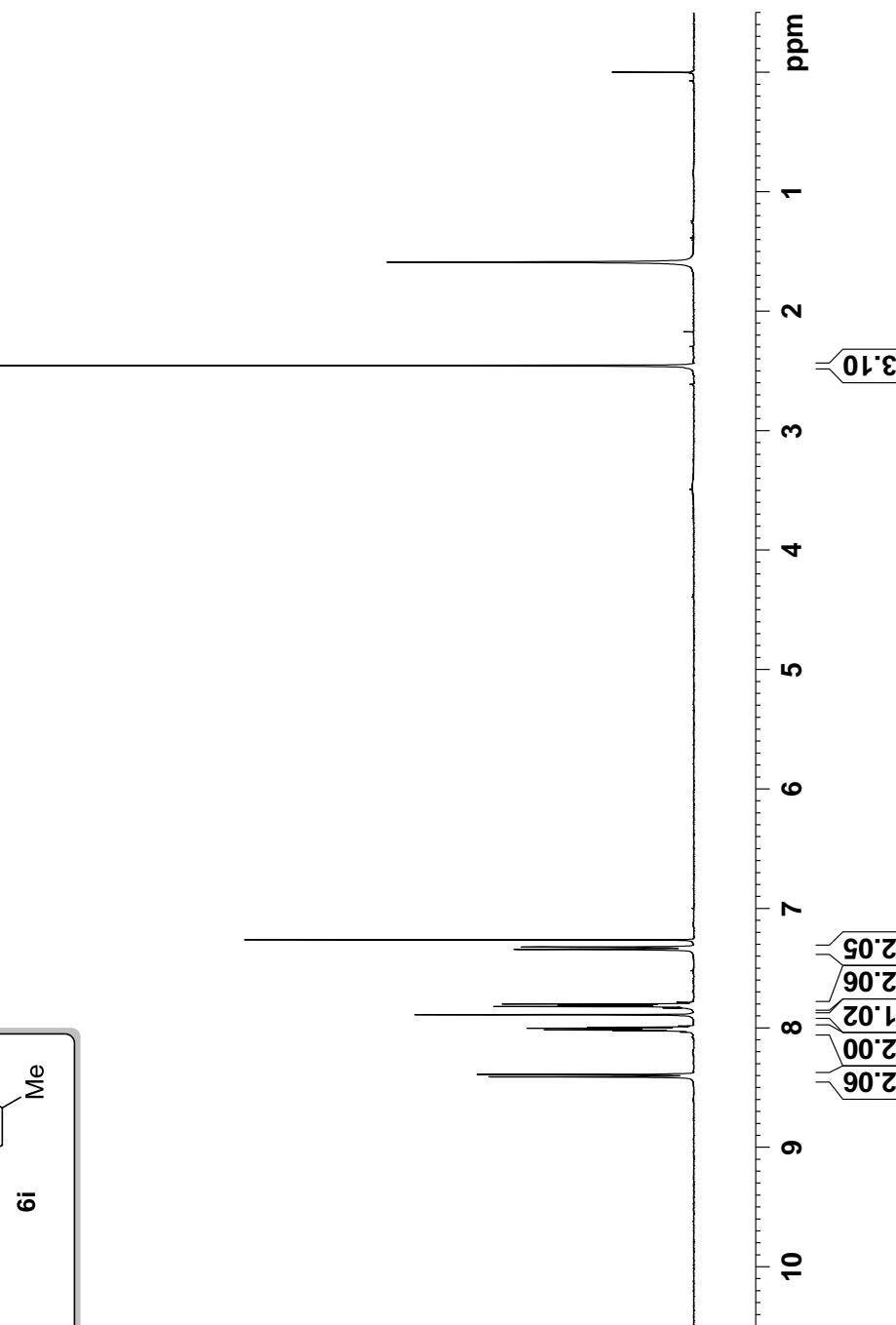
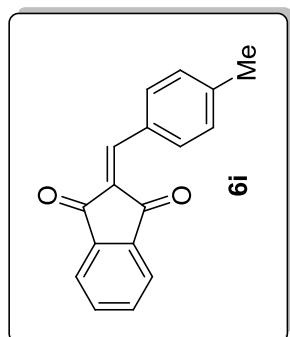
Date 20121004
Time 15.49
INSTRUM spect
PROBHD 5 mm BBO BB-1H
PULPROG zg30
TD 32768
SOLVENT CDC13
NS 16
DS 0
SWH 7246.377 Hz
FIDRES 0.221142 Hz
AQ 2.2610421 sec
RG 322.5
DW 69.000 usec
DE 6.50 usec
TE 297.4 K
D1 2.00000000 sec
TDO 1

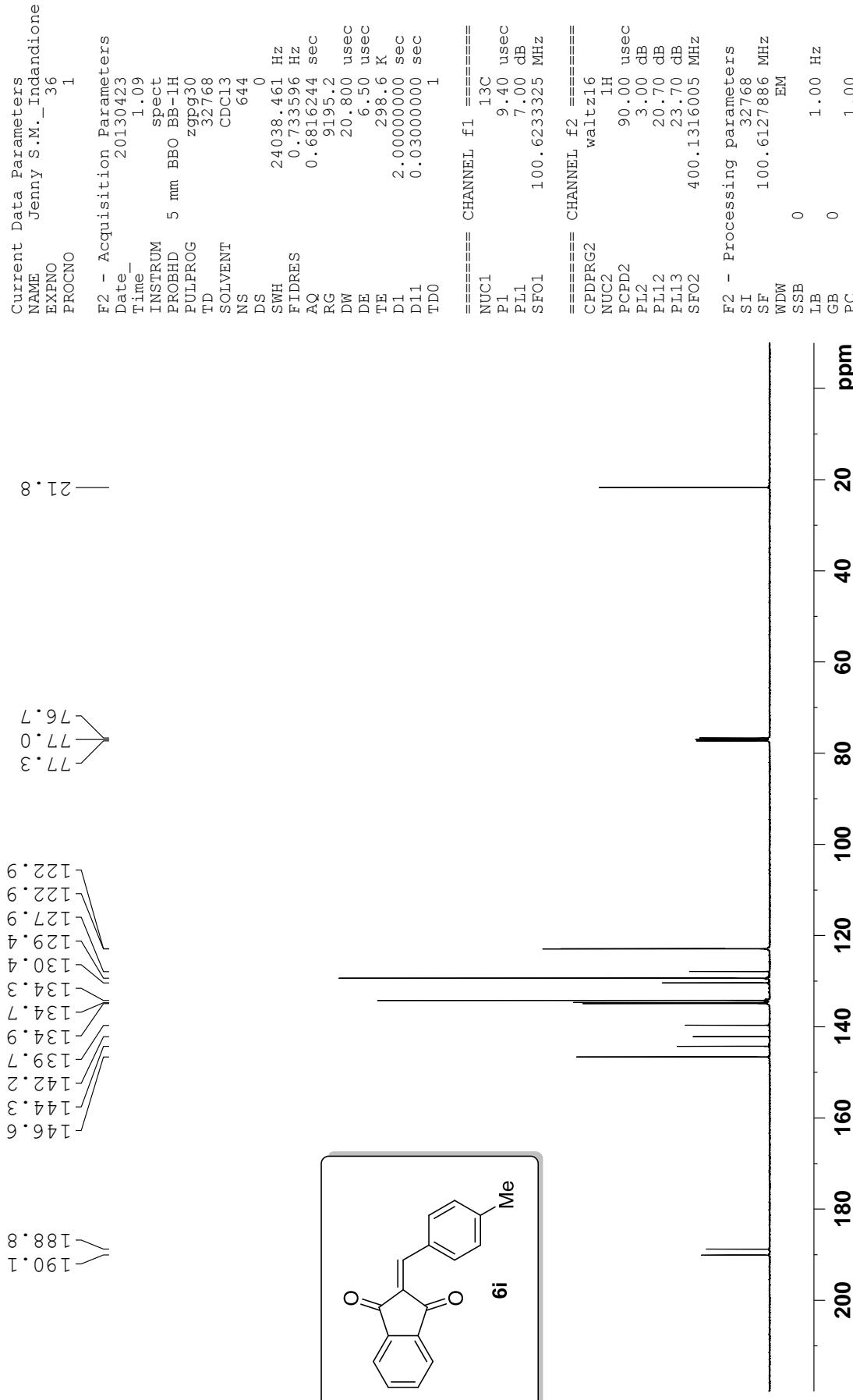
===== CHANNEL f1 =====

NUC1 1H
P1 11.90 usec
PL1 3.00 dB
SFO1 400.1324008 MHz

F2 - Processing parameters

SI 16384
SF 400.1300082 MHz
WDW EM
SSB 0
LB 0 Hz
GB 0
PC 1.00





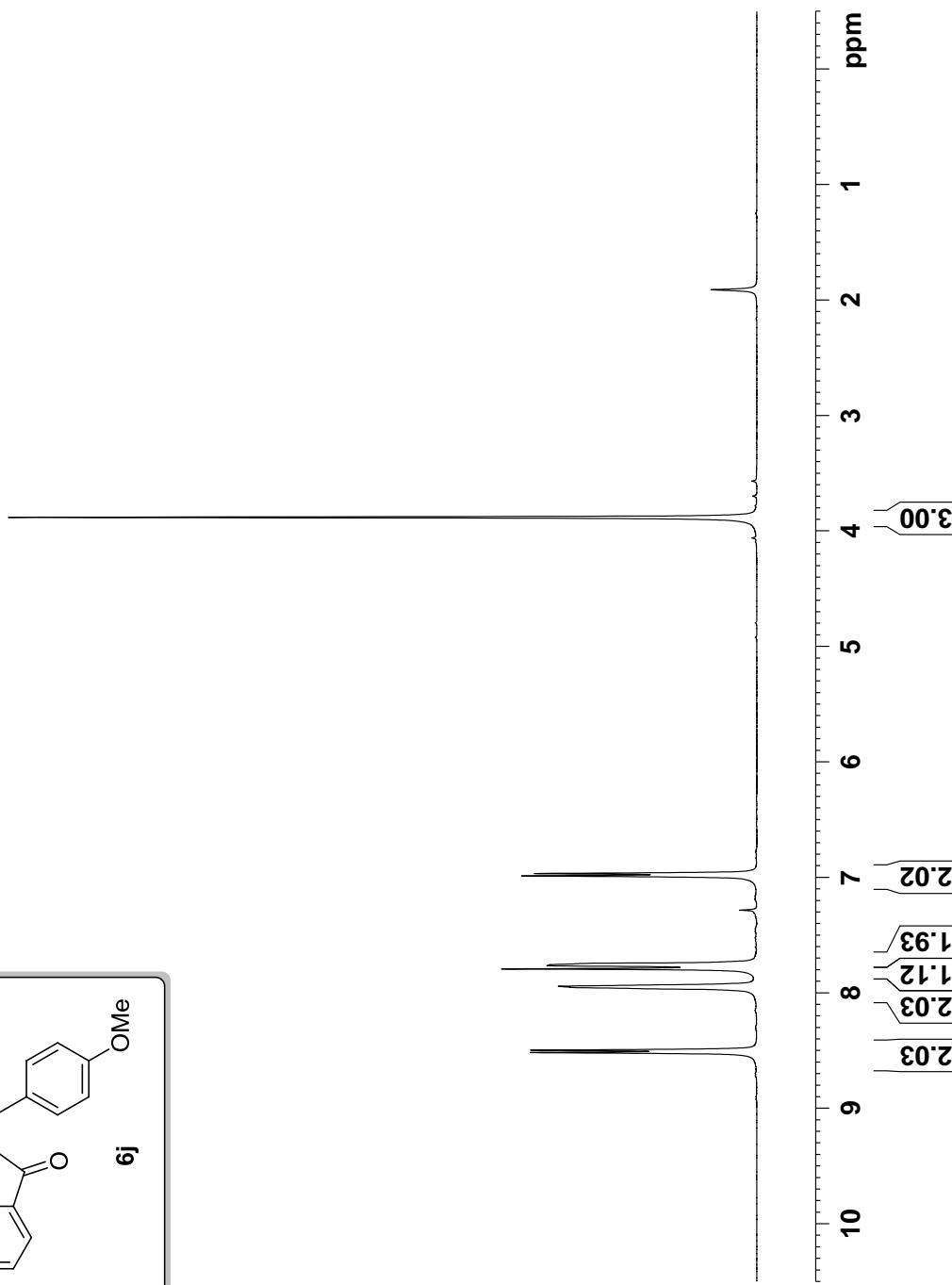
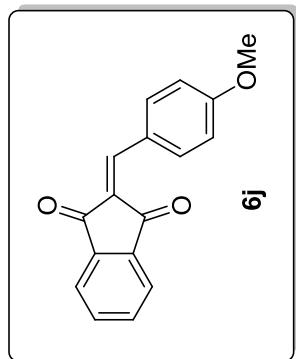
Current	Data Parameters
NAME	Jenny S.M._Indandione
EXPNO	27
PROCNO	1

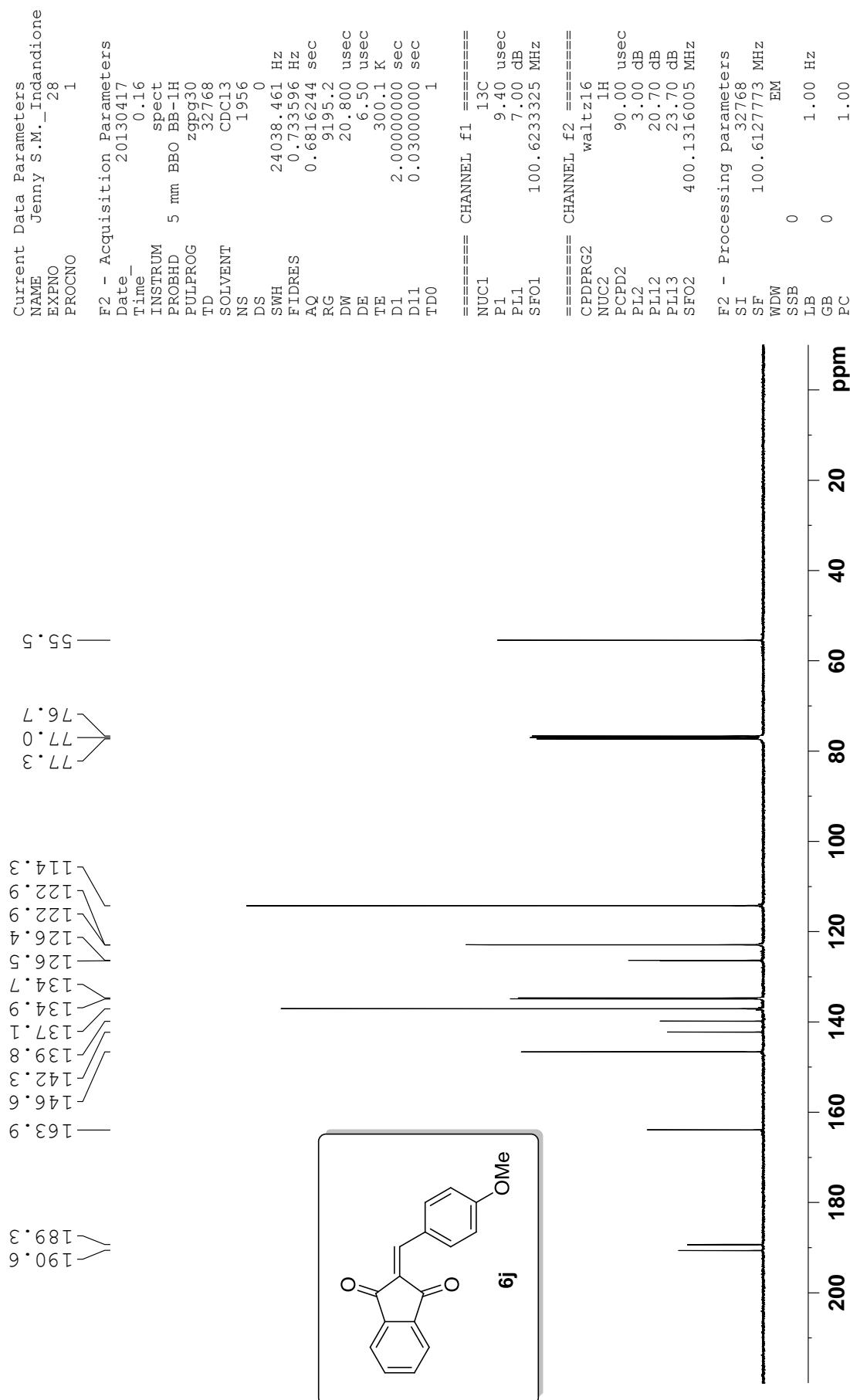
```

FF2 - Acquisition Parameters
Date_      20130417
Time_      0.12
INSTRUM   PROBHD
PROBTD    5 mm BBO BB-LH
PULPROG   Zg30
TD        32768
SOLVENT   CDC13
NS         16
DS         0
SWH       7246.377 Hz
FIDRES   0.221142 Hz
AQ        2.261042 sec
RG        64
DW        69.0000 usec
DE        6.50 usec
TE        299.9 K
D1        2.0000000 sec
TDTO     1

```

CHANNEL f1 = =====			
NUC1	1 H	11.90	usec
P1	3.00	dB	
PLL1	400.13244008	MHz	
SF01	400.12999999	MHz	EM
SF01 - Processing parameters			
SI	1.6384		
SF	400.12999999	MHz	
WDW	0	Hz	
SSB	0	Hz	
LB	0	Hz	
GB	0	Hz	
PC	1.00		





Current Data Parameters
NAME Jenny S.M of indandione
EXPNO 6
PROCNO 1

F2 - Acquisition Parameters

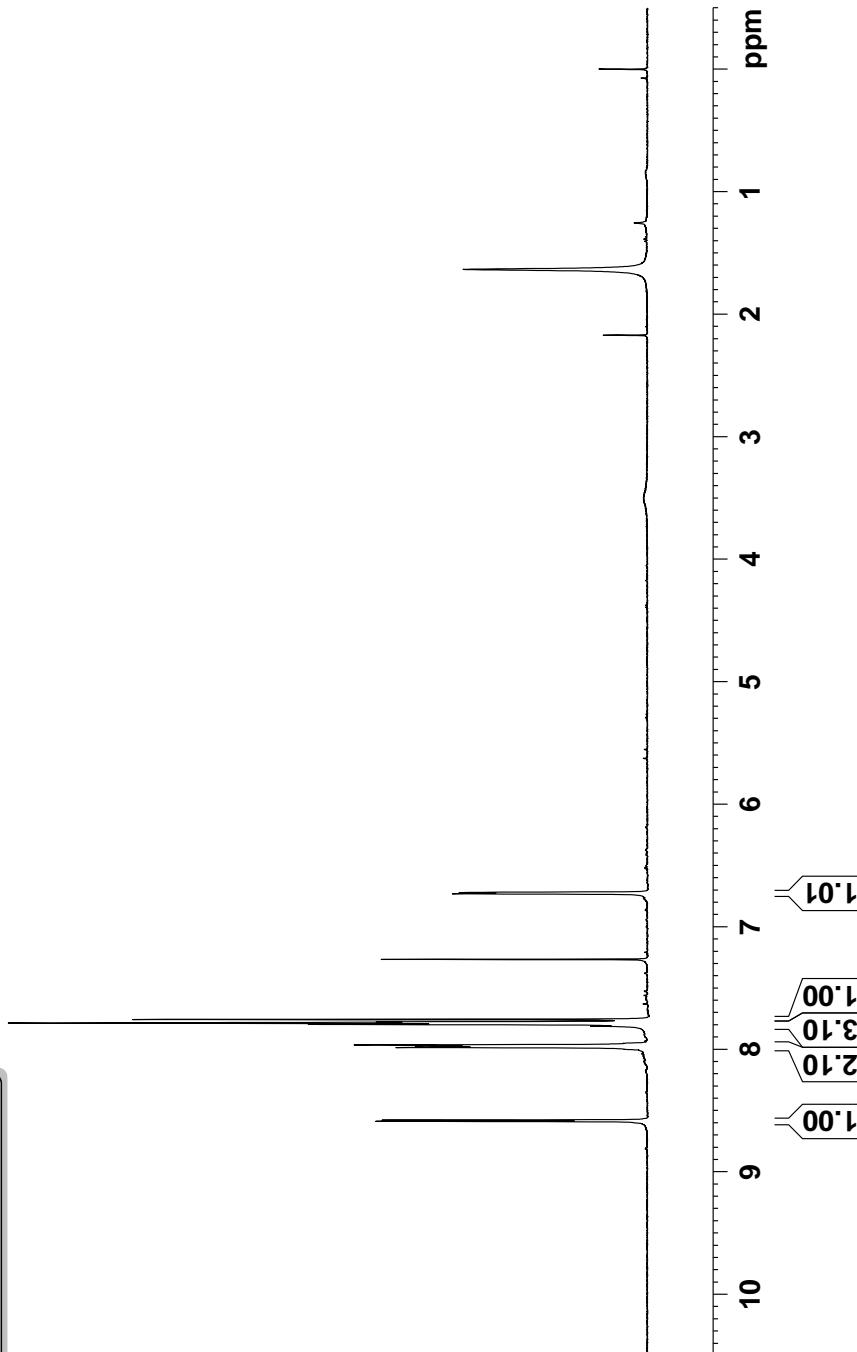
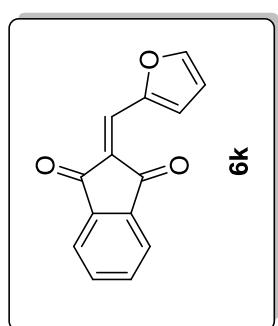
Date 20121012
Time 18.58
INSTRUM spect
PROBHD 5 mm BBO BB-1H
PULPROG zg30
TD 32768
SOLVENT CDC13
NS 16
DS 0
SWH 7246.377 Hz
FIDRES 0.221142 Hz
AQ 2.2610421 sec
RG 256
DW 69.000 usec
DE 6.50 usec
TE 299.5 K
D1 2.00000000 sec
TDO 1

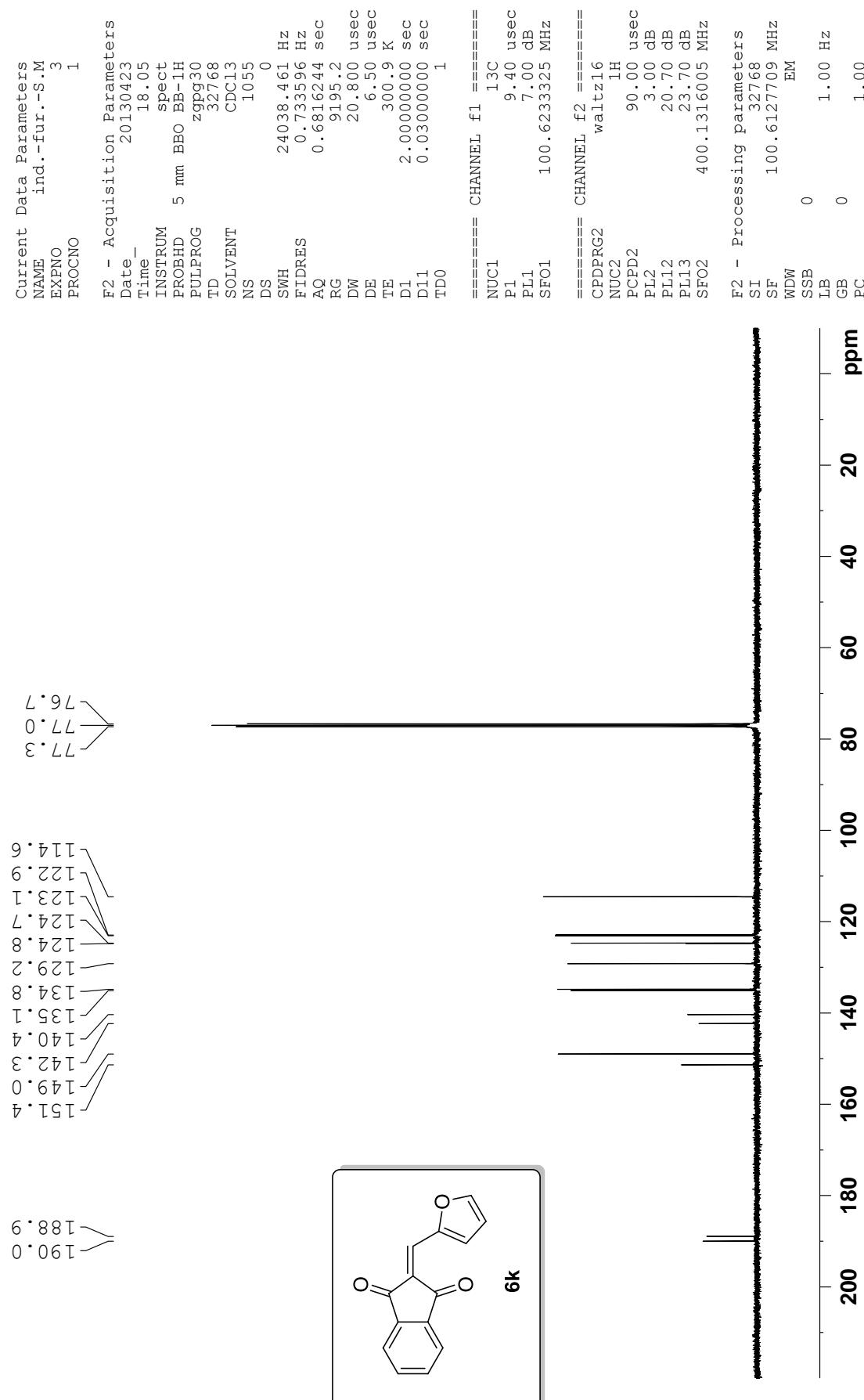
===== CHANNEL f1 =====

NUC1 1H
P1 11.90 usec
PL1 3.00 dB
SFO1 400.1324008 MHz

F2 - Processing parameters

SI 16384
SF 400.1300069 MHz
WDW EM
SSB 0
LB 0 Hz
GB 0
PC 1.00





Current Data Parameters
NAME Jenny S.M of indandione
EXPNO 9
PROCNO 1

F2 - Acquisition Parameters

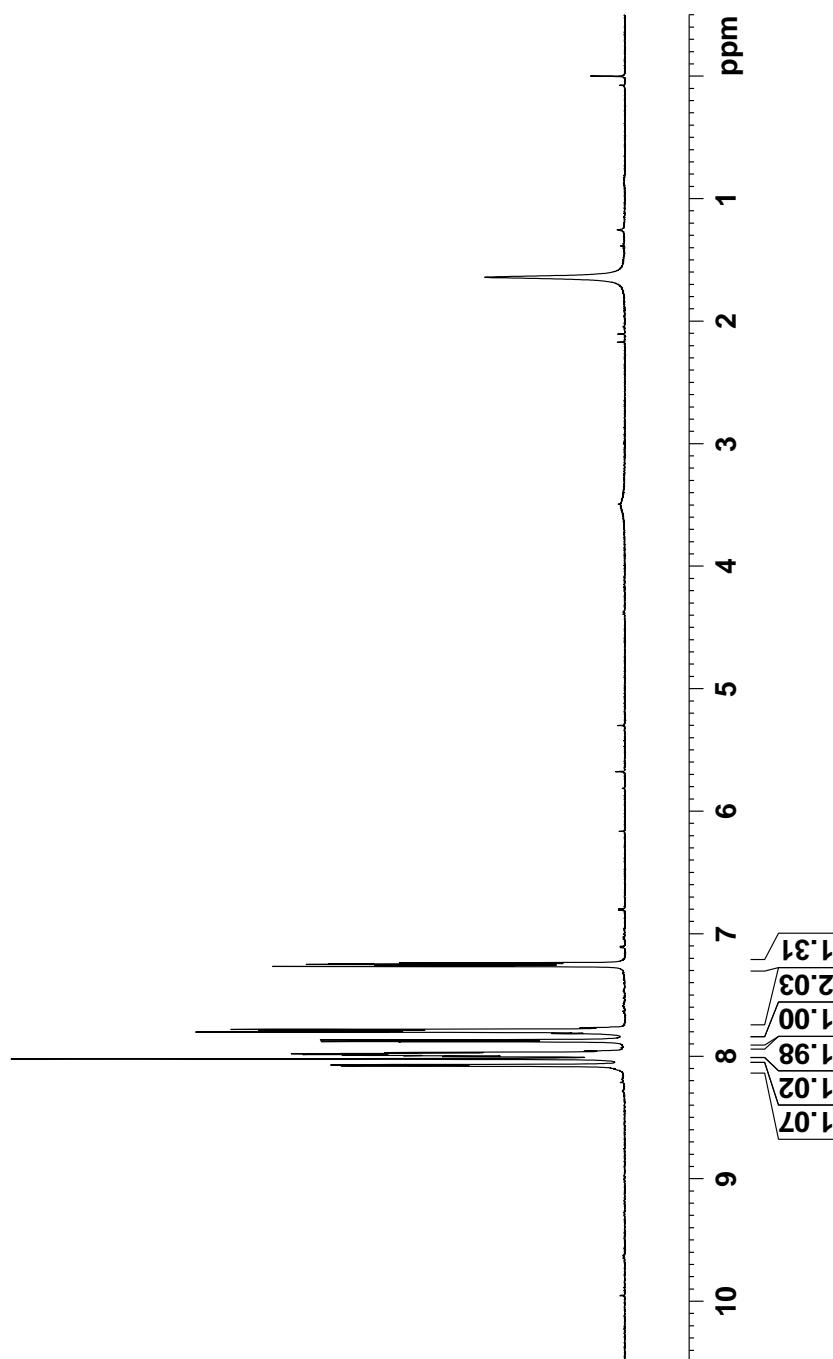
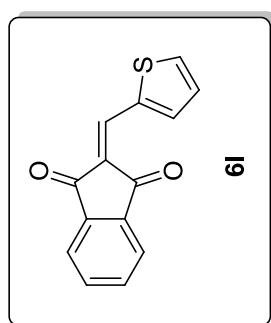
Date 20120930
Time 15.42
INSTRUM spect
PROBHD 5 mm BBO BB-1H
PULPROG zg30
TD 32768
SOLVENT CDC13
NS 16
DS 0
SWH 7246.377 Hz
FIDRES 0.221142 Hz
AQ 2.2610421 sec
RG 256
DW 69.000 usec
DE 6.50 usec
TE 299.4 K
D1 2.00000000 sec
TDO 1

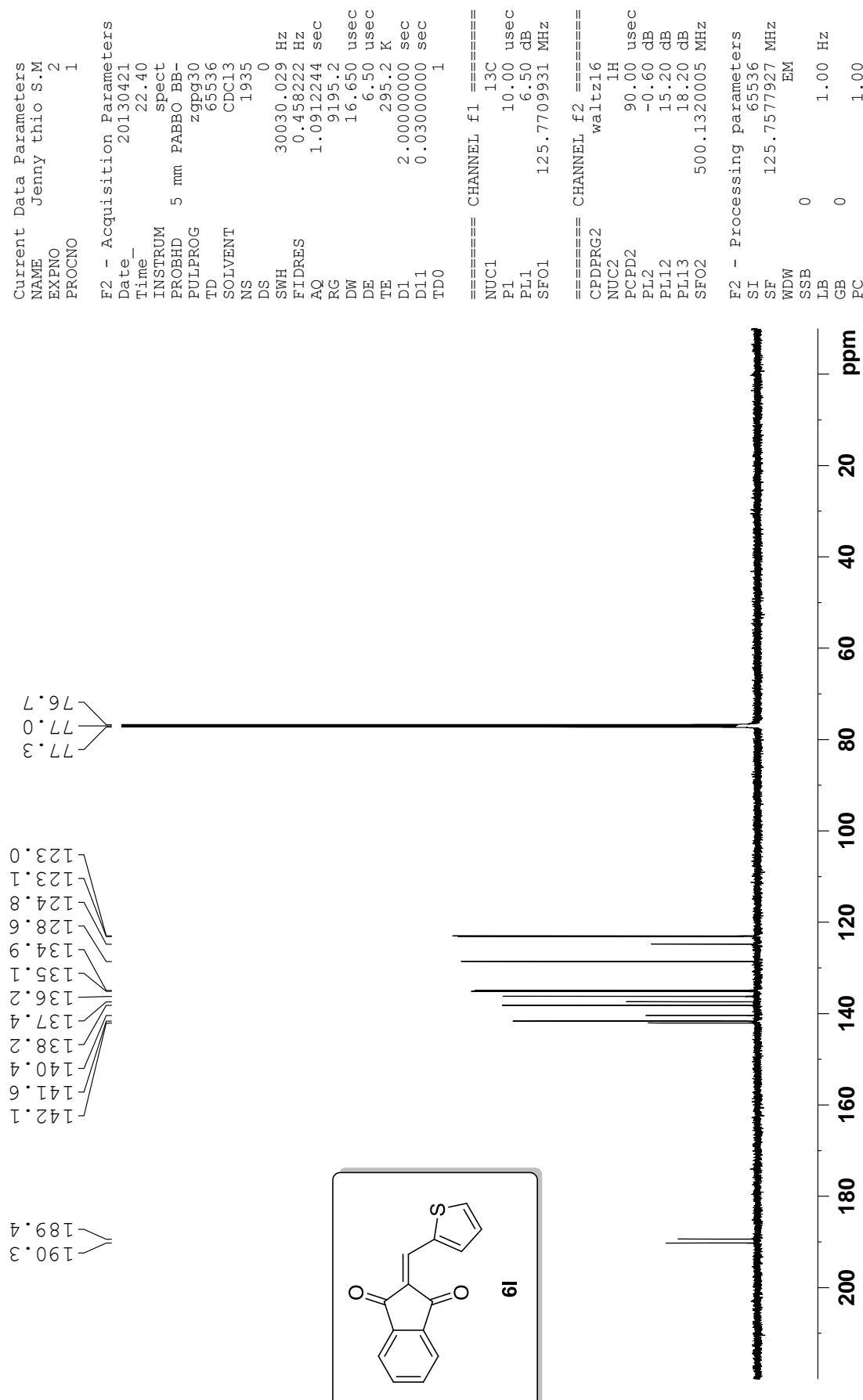
===== CHANNEL f1 =====

NUC1 1H
P1 11.70 usec
PL1 4.00 dB
SFO1 400.1324008 MHz
WDM EM
SSB 0
LB 0 Hz
GB 0
PC 1.00

F2 - Processing parameters

SI 16384
SF 400.1300061 MHz
WDW EM
SSB 0
LB 0 Hz
GB 0
PC 1.00





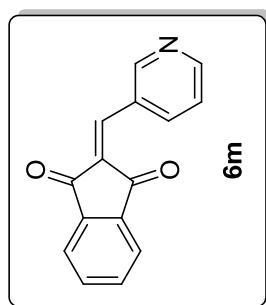
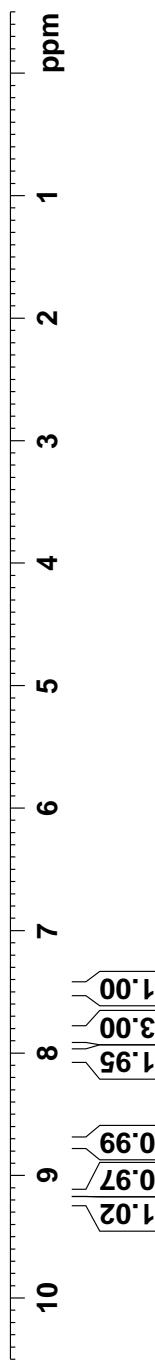
Current Data Parameters
NAME Jenny S.M._Indandione
EXPNO 25
PROCNO 1

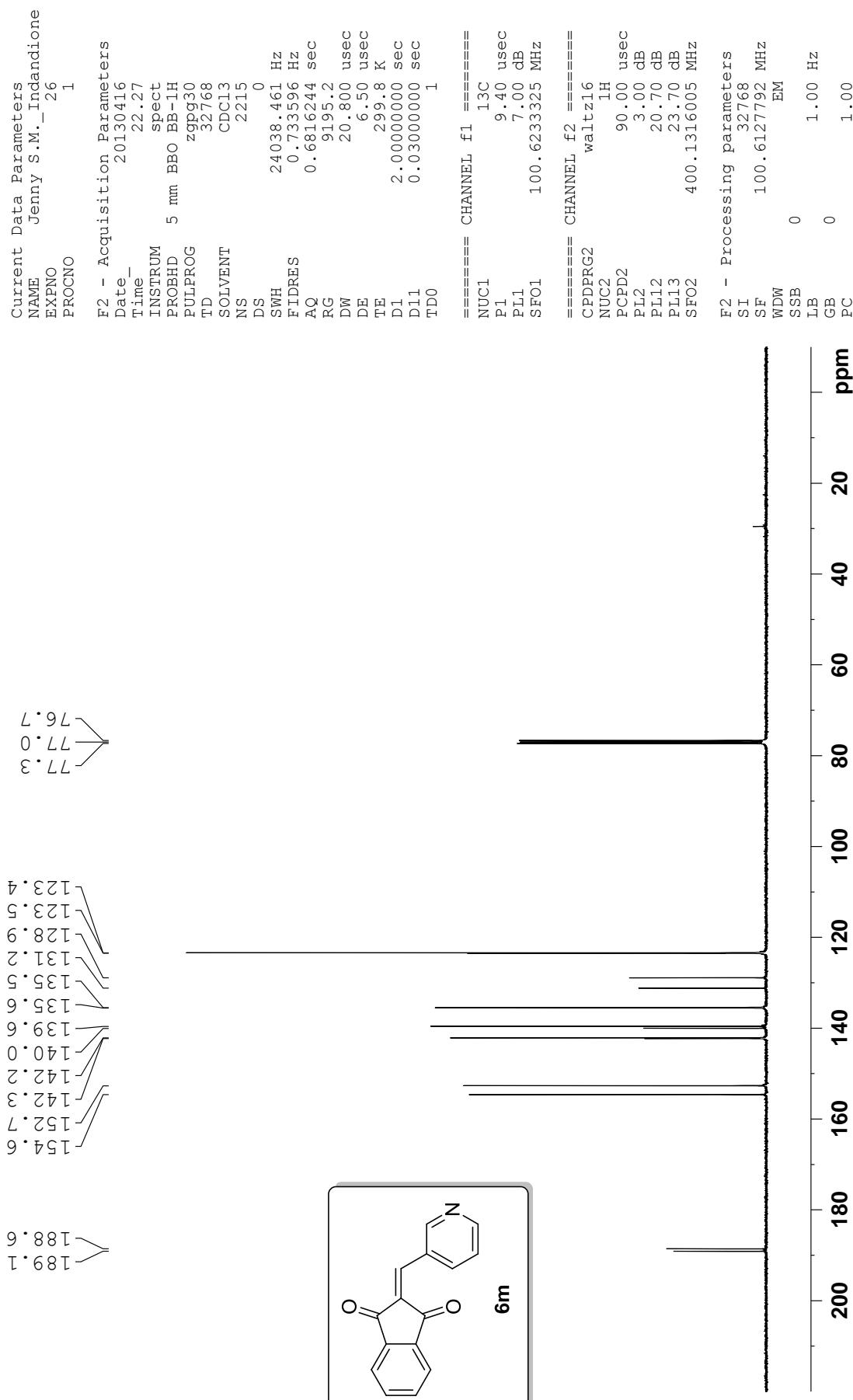
F2 - Acquisition Parameters

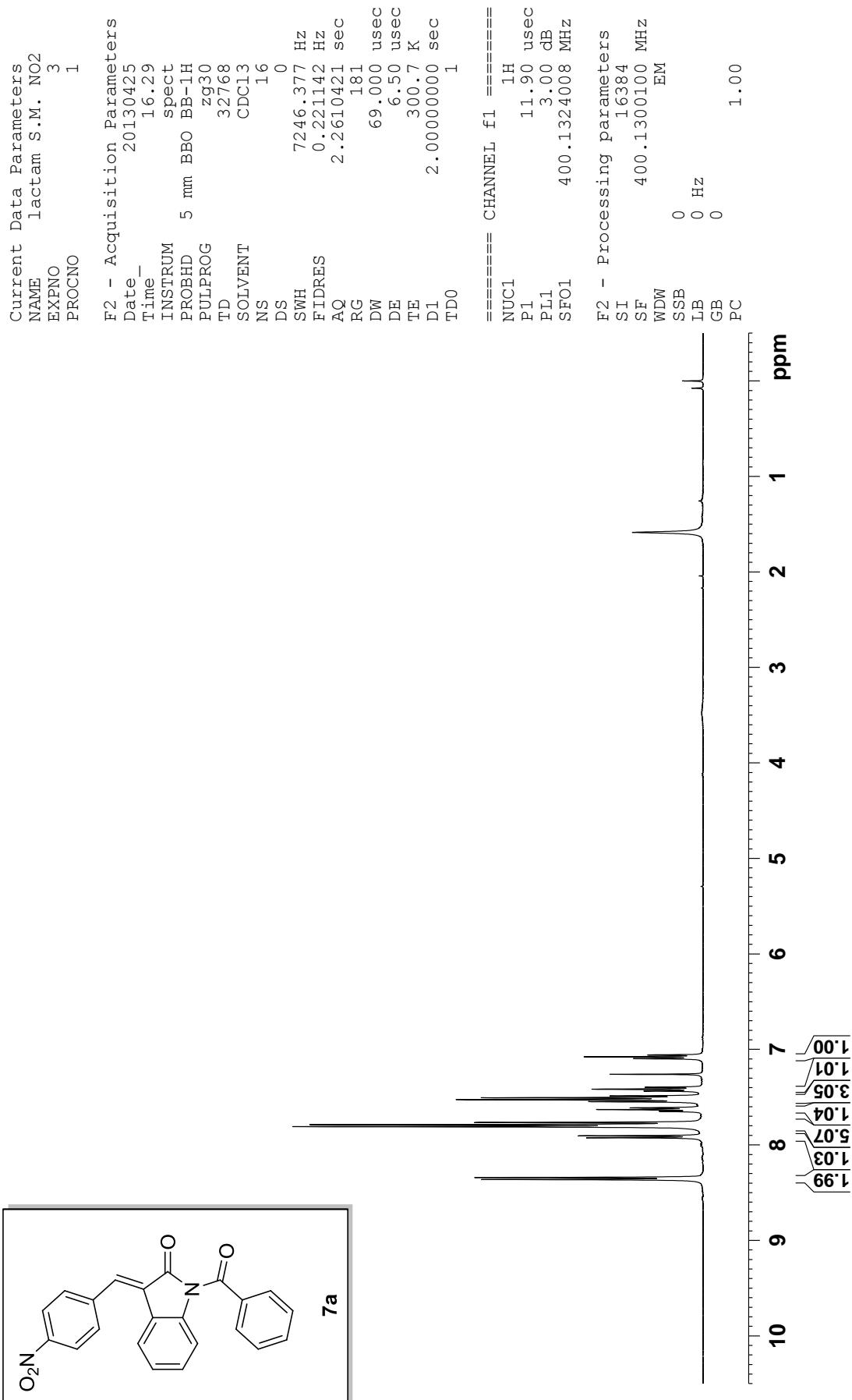
Date 20130416
Time 22.24
INSTRUM spect
PROBHD 5 mm BBO BB-1H
PULPROG zg30
TD 32768
SOLVENT CDC13
NS 16
DS 0
SWH 7246.377 Hz
FIDRES 0.221142 Hz
AQ 2.2610421 sec
RG 64
DW 69.000 usec
DE 6.50 usec
TE 299.6 K
D1 2.0000000 sec
TDO 1

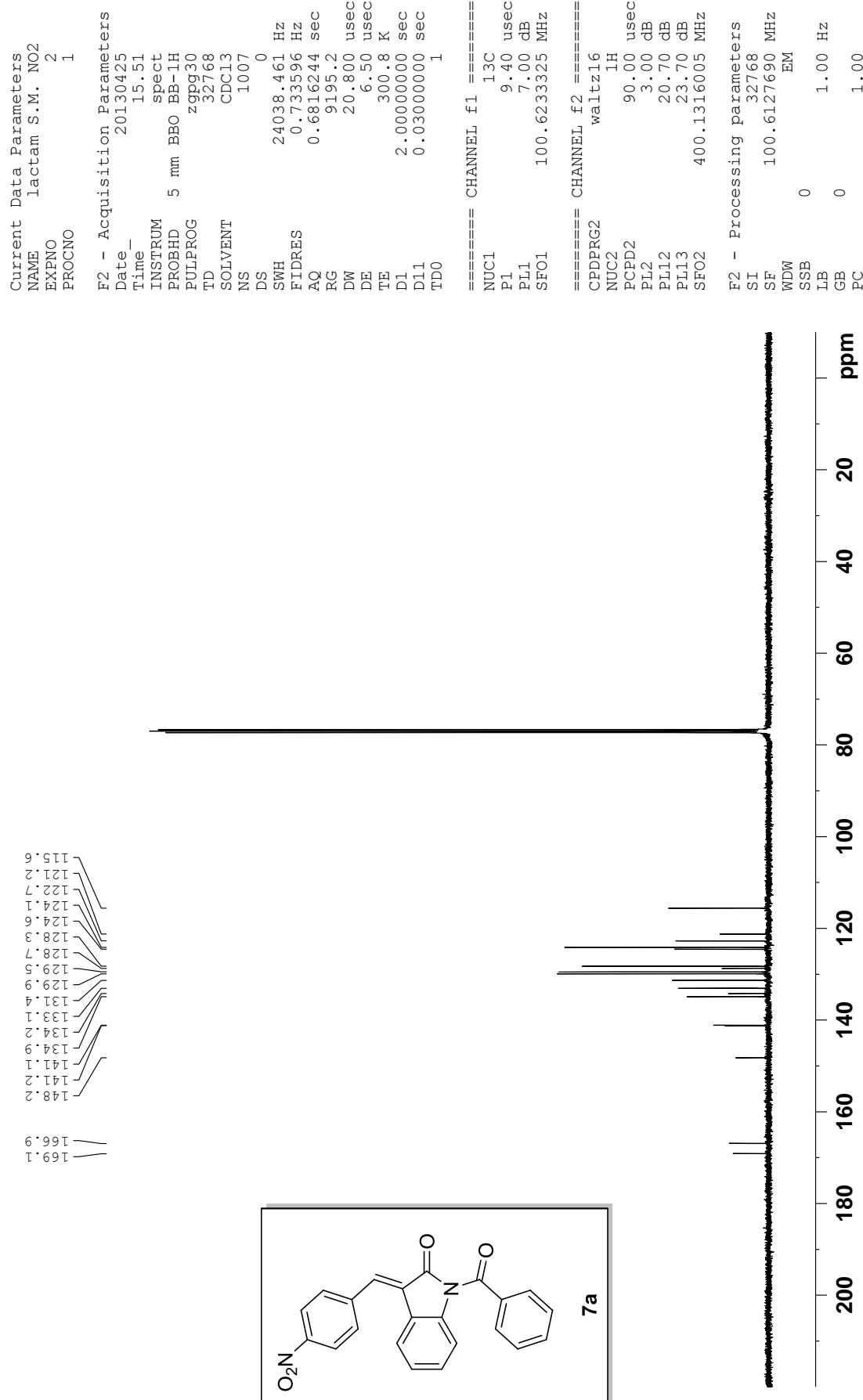
===== CHANNEL f1 =====

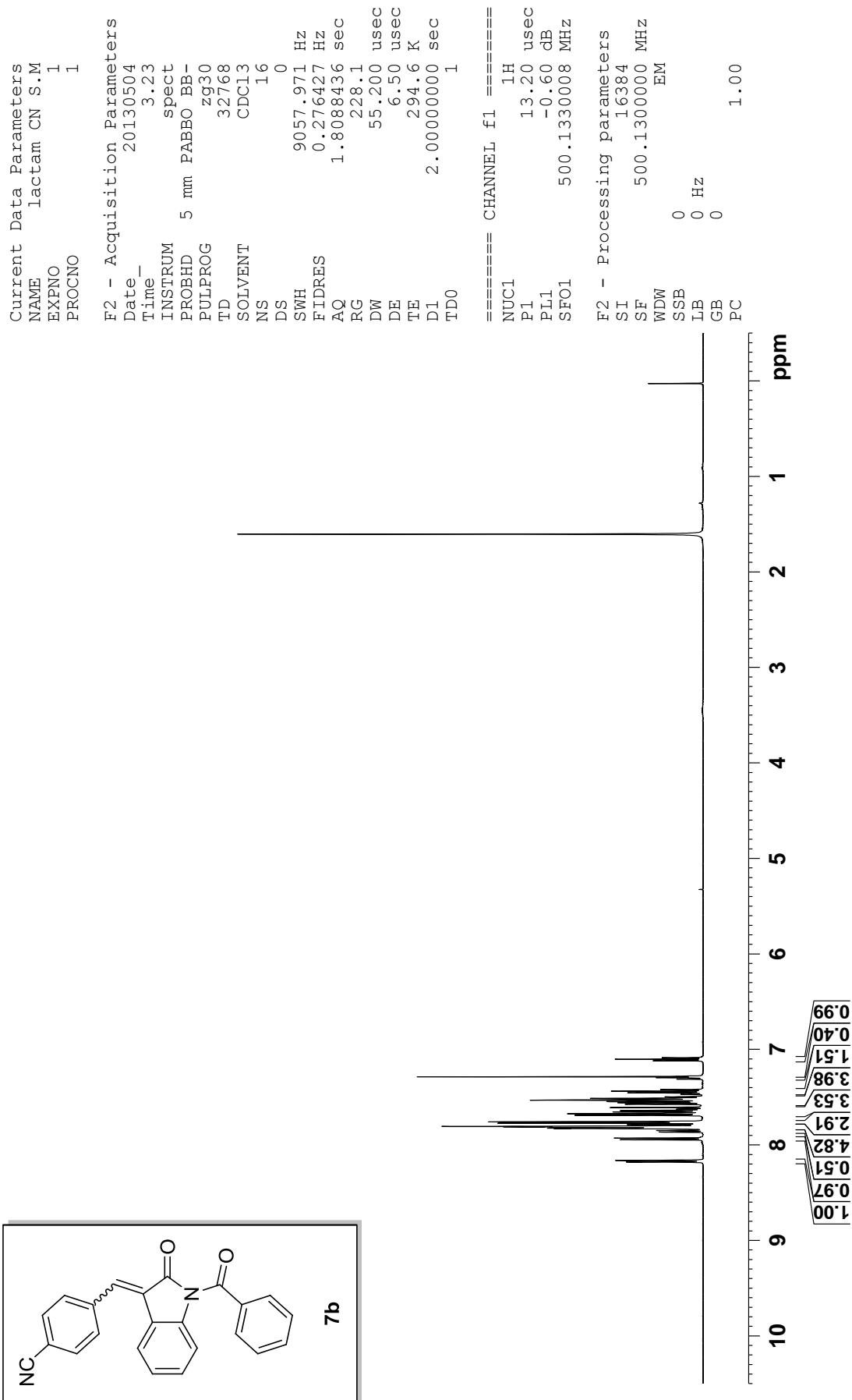
NUC1 1H
P1 11.90 usec
PL1 3.00 dB
SFO1 400.1324008 MHz
WDW EM
SSB 0 Hz
LB 0 Hz
GB 0
PC 1.00

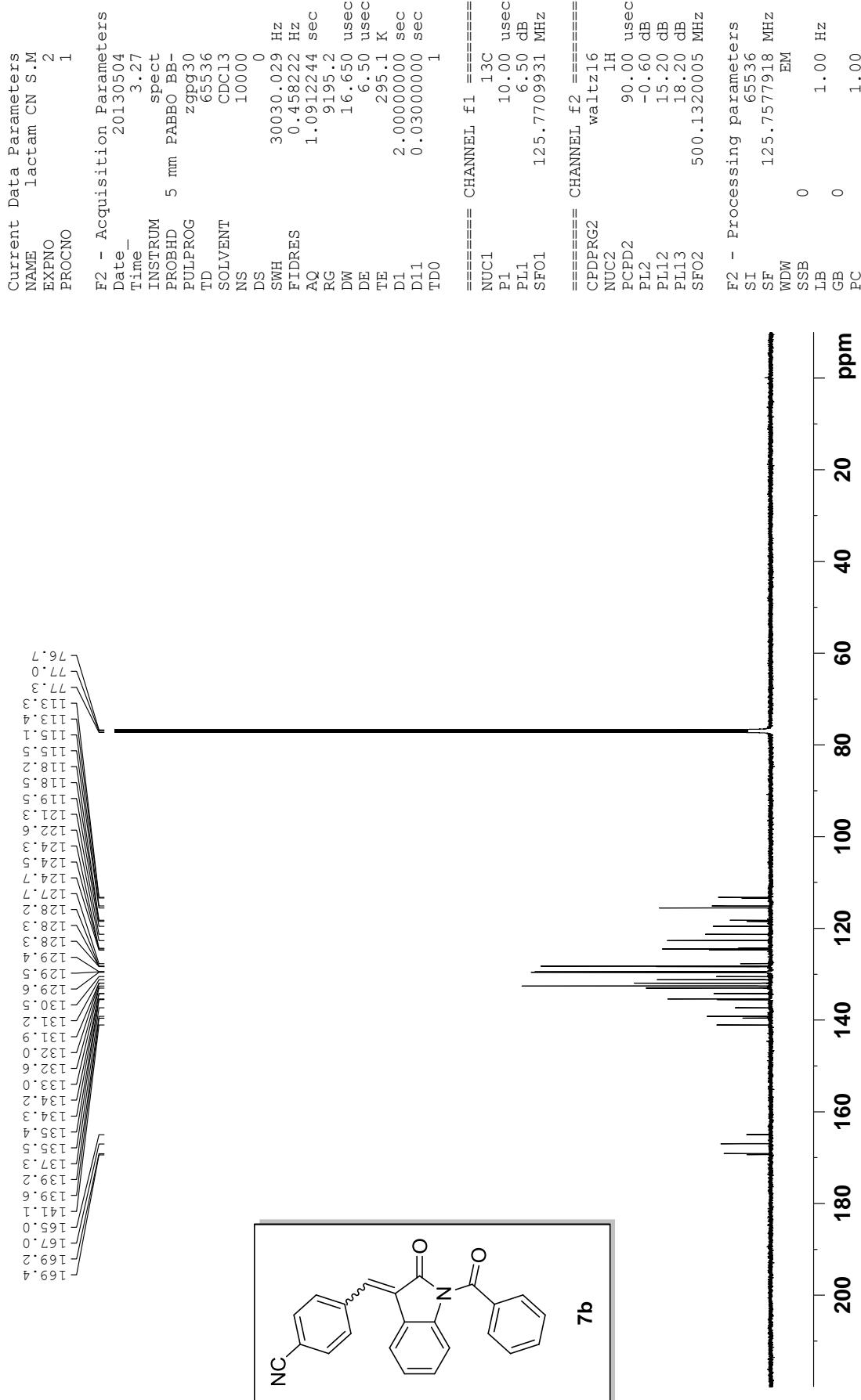


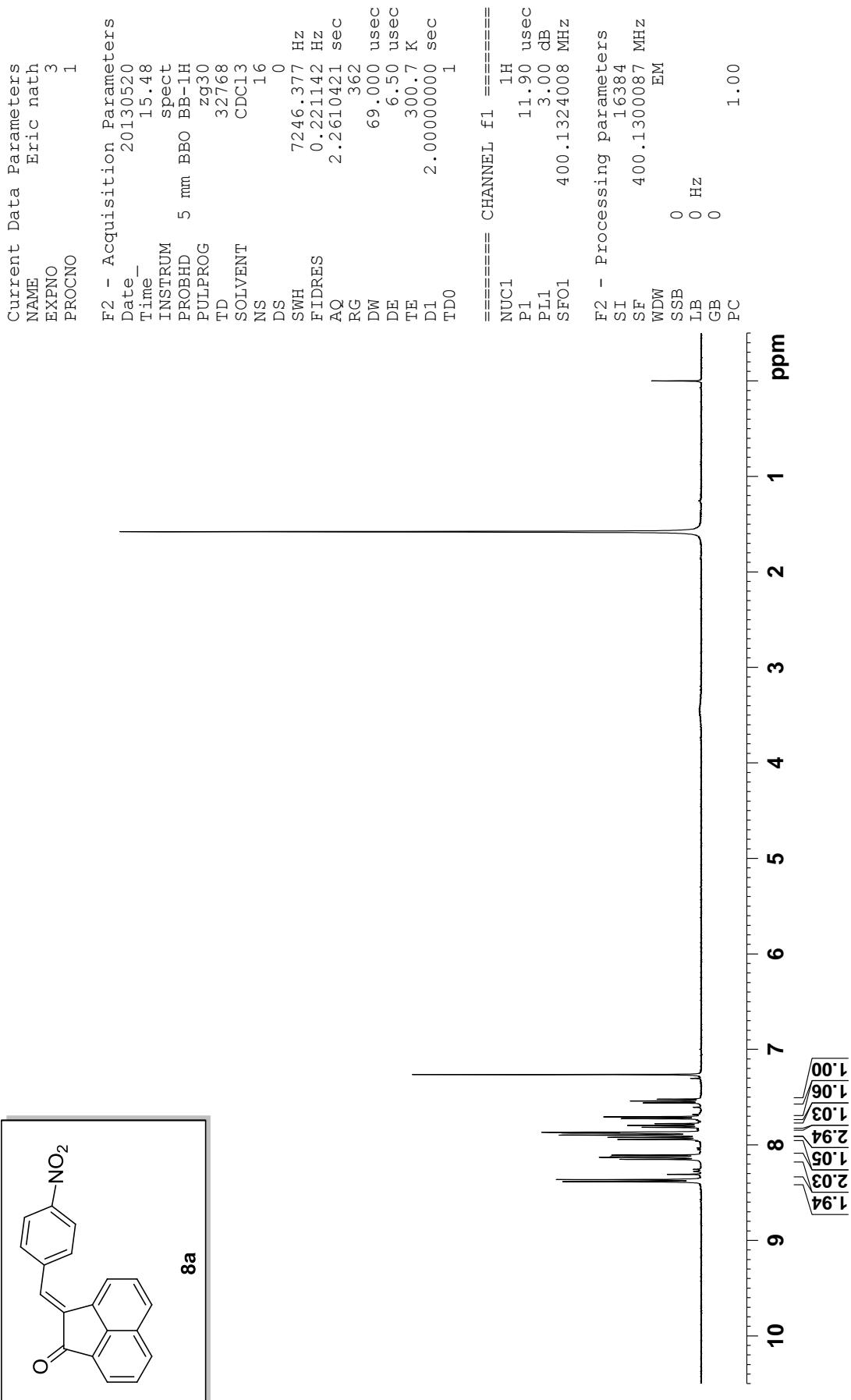


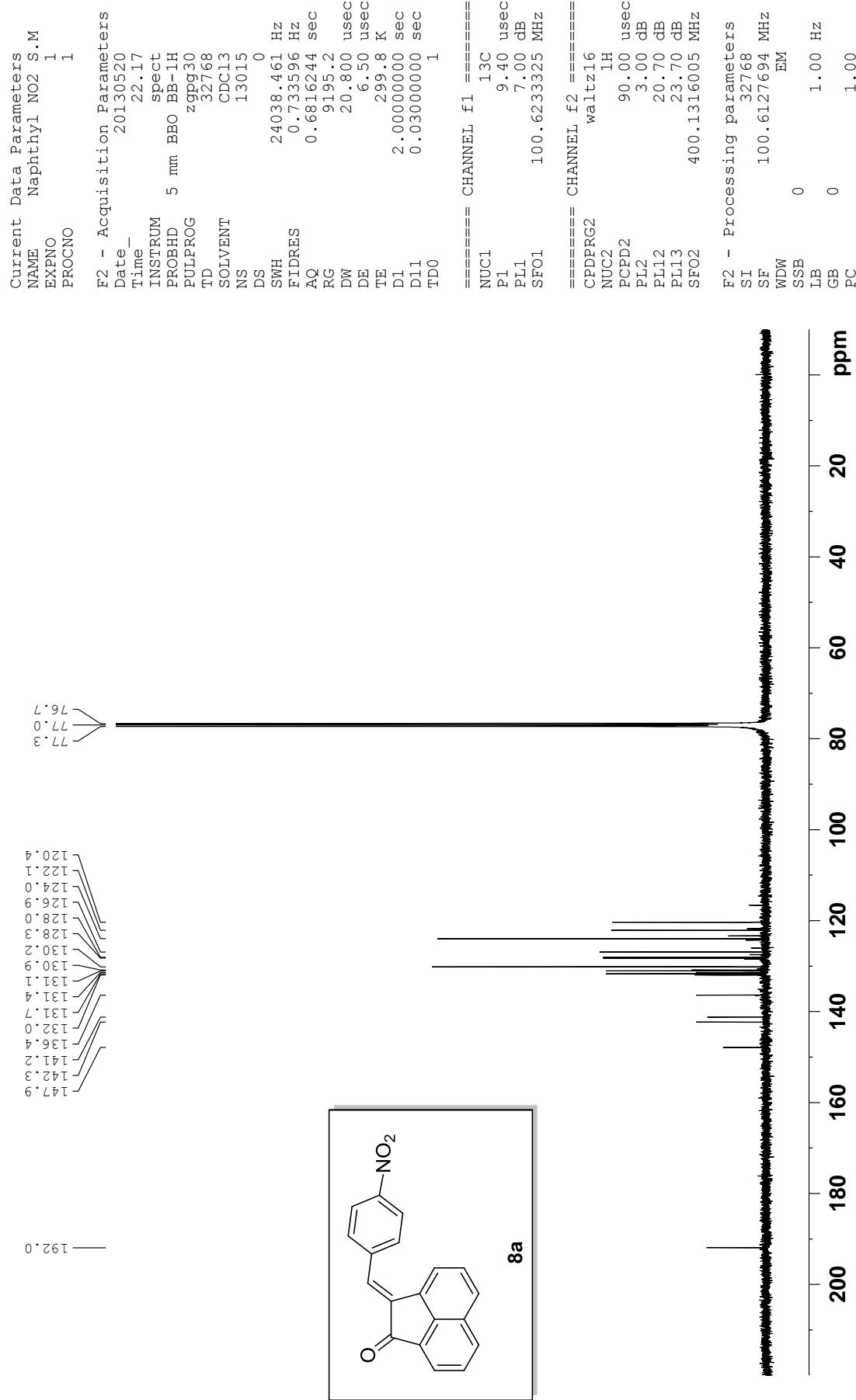


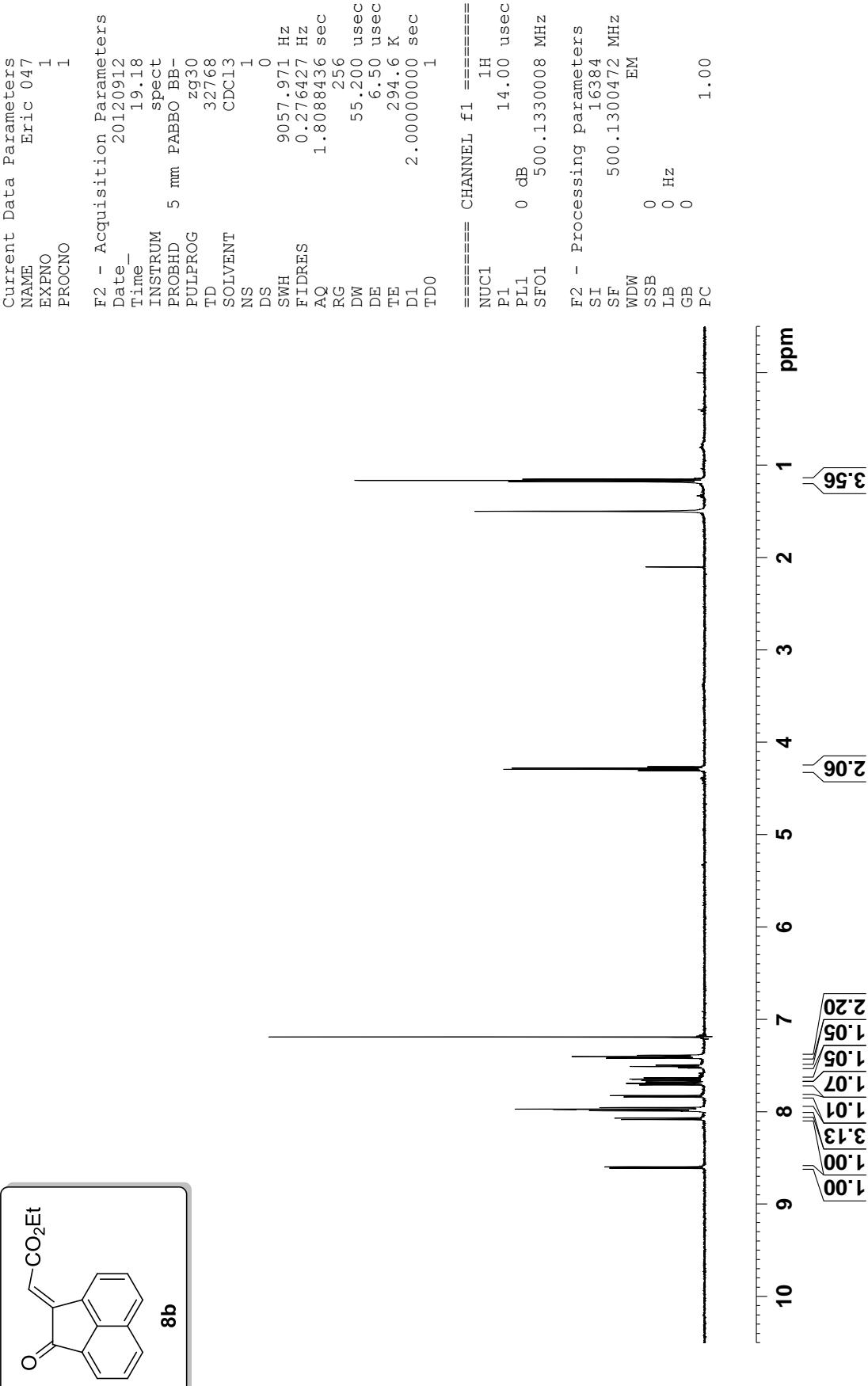


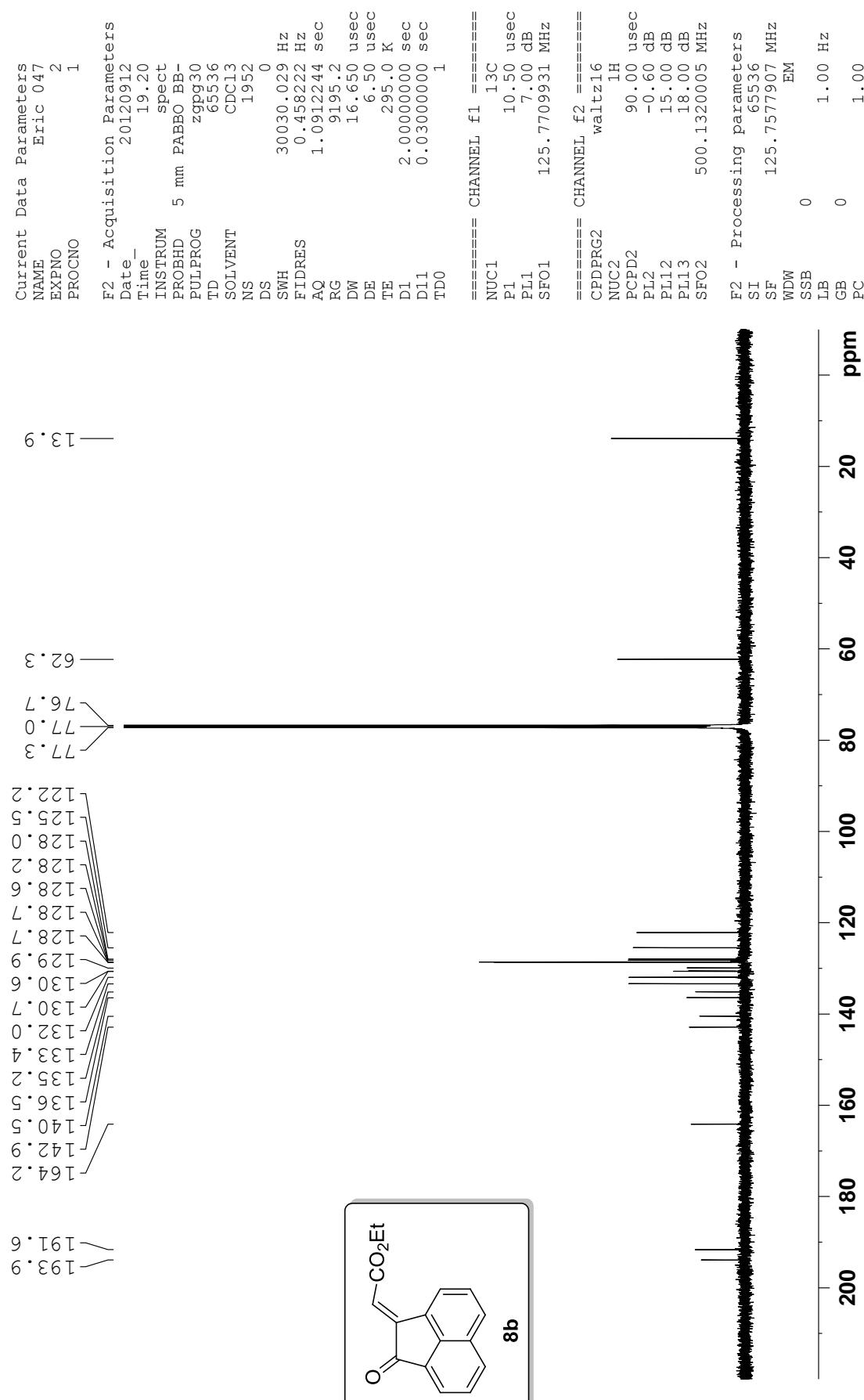


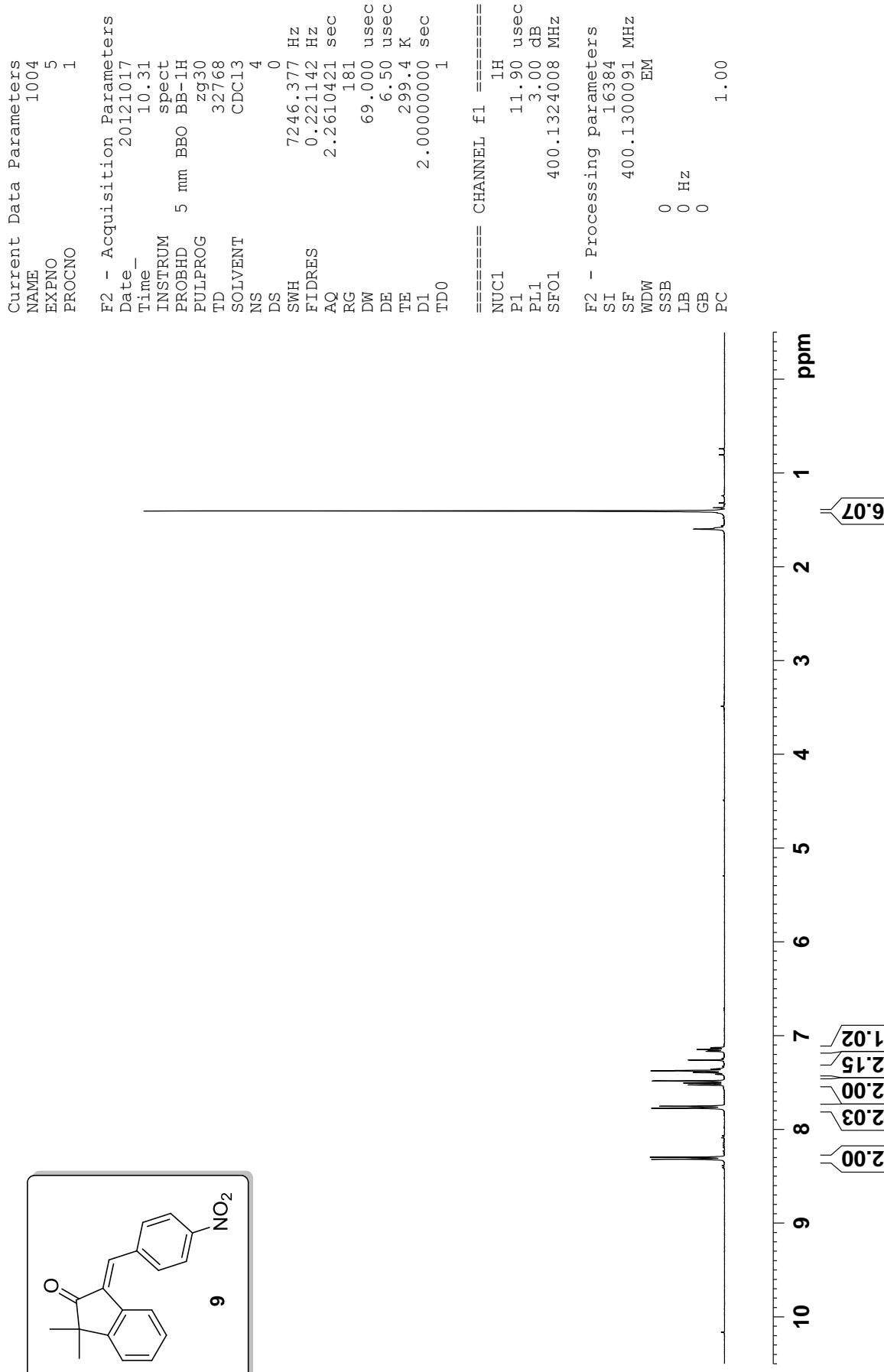


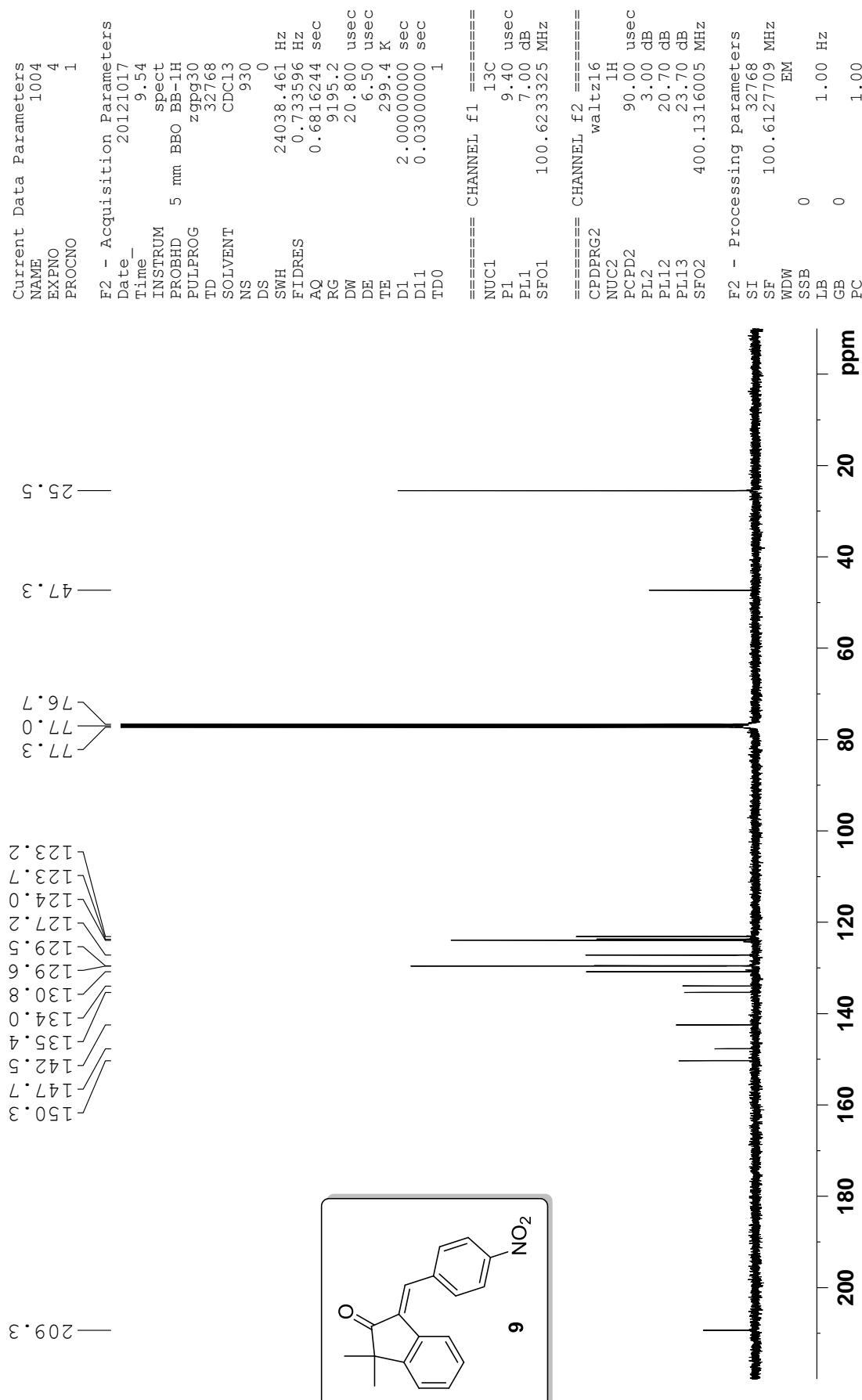










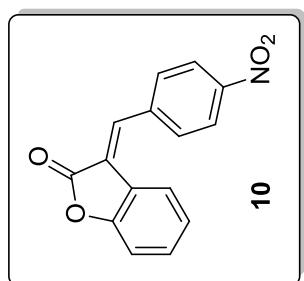
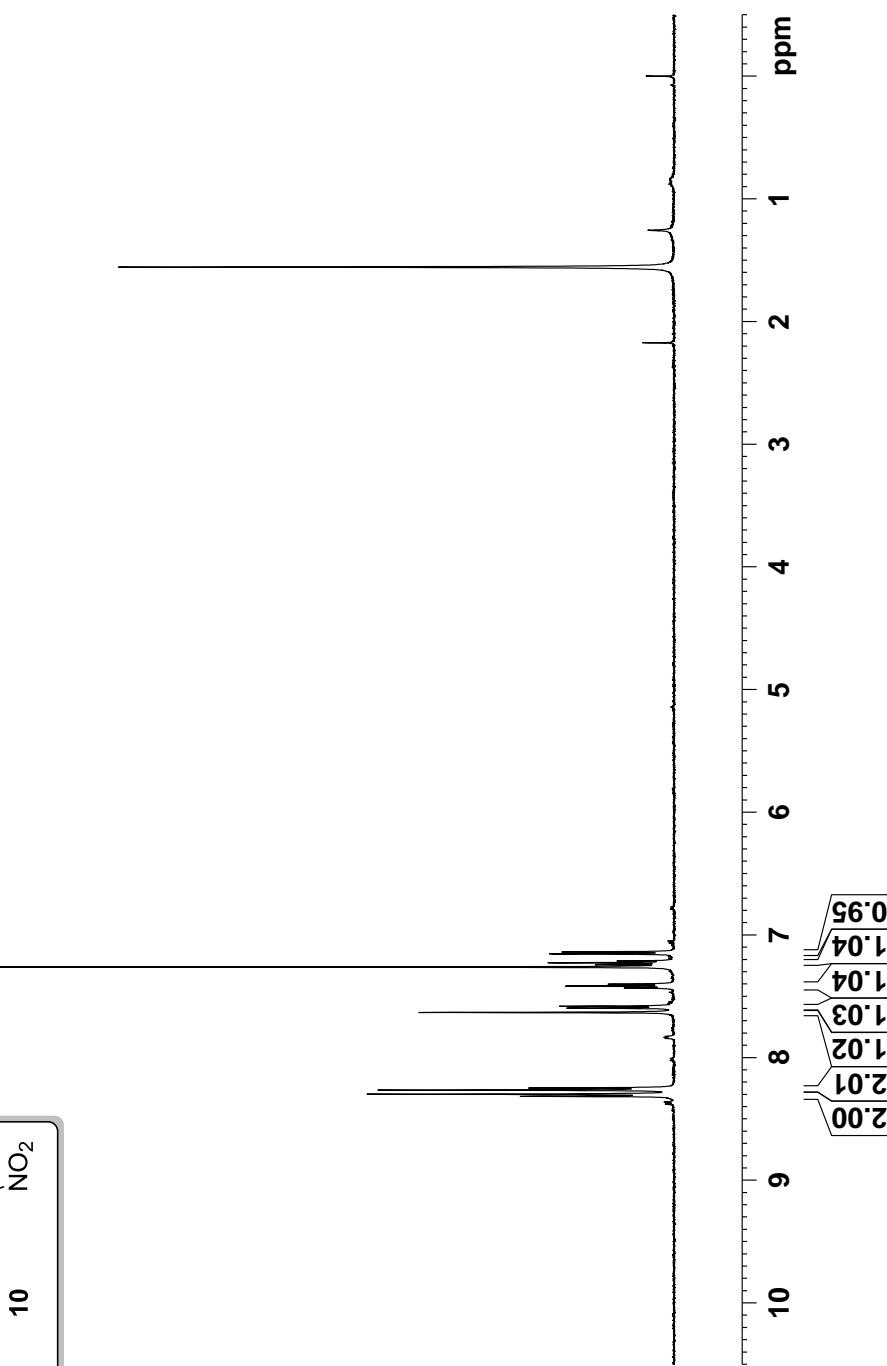


Current Data Parameters
NAME Jenny lactone S.M
EXPNO 1
PROCNO 1

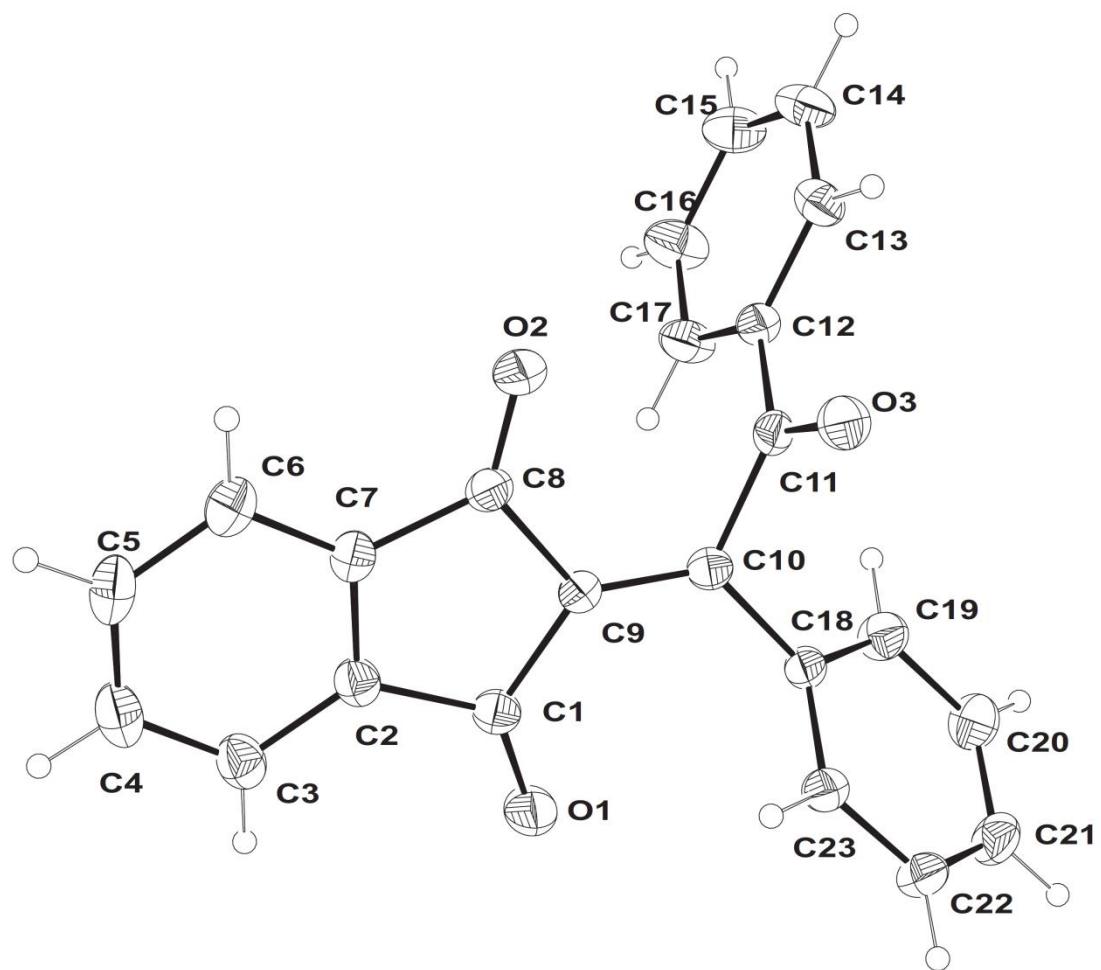
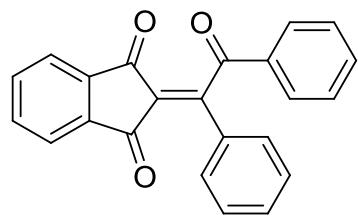
F2 - Acquisition Parameters
Date 20130420
Time 14.26
INSTRUM spect
PROBHD 5 mm PABBO BB-
PULPROG Zg30
TD 32768
SOLVENT CDCl3
NS 16
DS 0
SWH 9057.971 Hz
FIDRES 0.276427 Hz
AQ 1.8088436 sec
RG 574.7
DW 55.200 usec
DE 6.50 usec
TE 295.7 K
D1 2.0000000 sec
TD0 1

===== CHANNEL f1 =====
NUC1 1H
P1 13.20 usec
PL1 -0.60 dB
SFO1 500.1330008 MHz

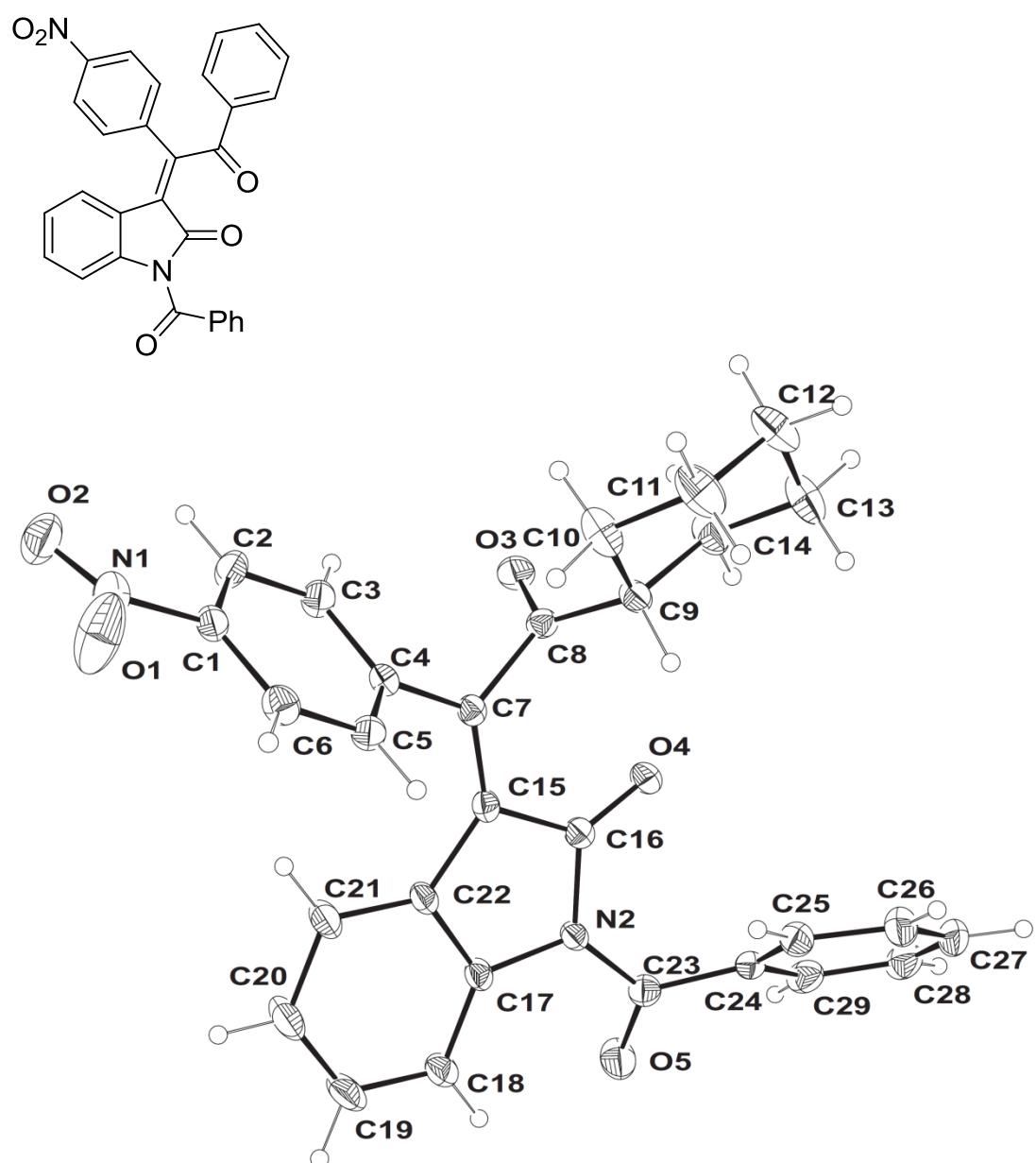
F2 - Processing parameters
SI 16384
SF 500.1300123 MHz
WDW EM
SSB 0
LB 0 Hz
GB 0
PC 1.00



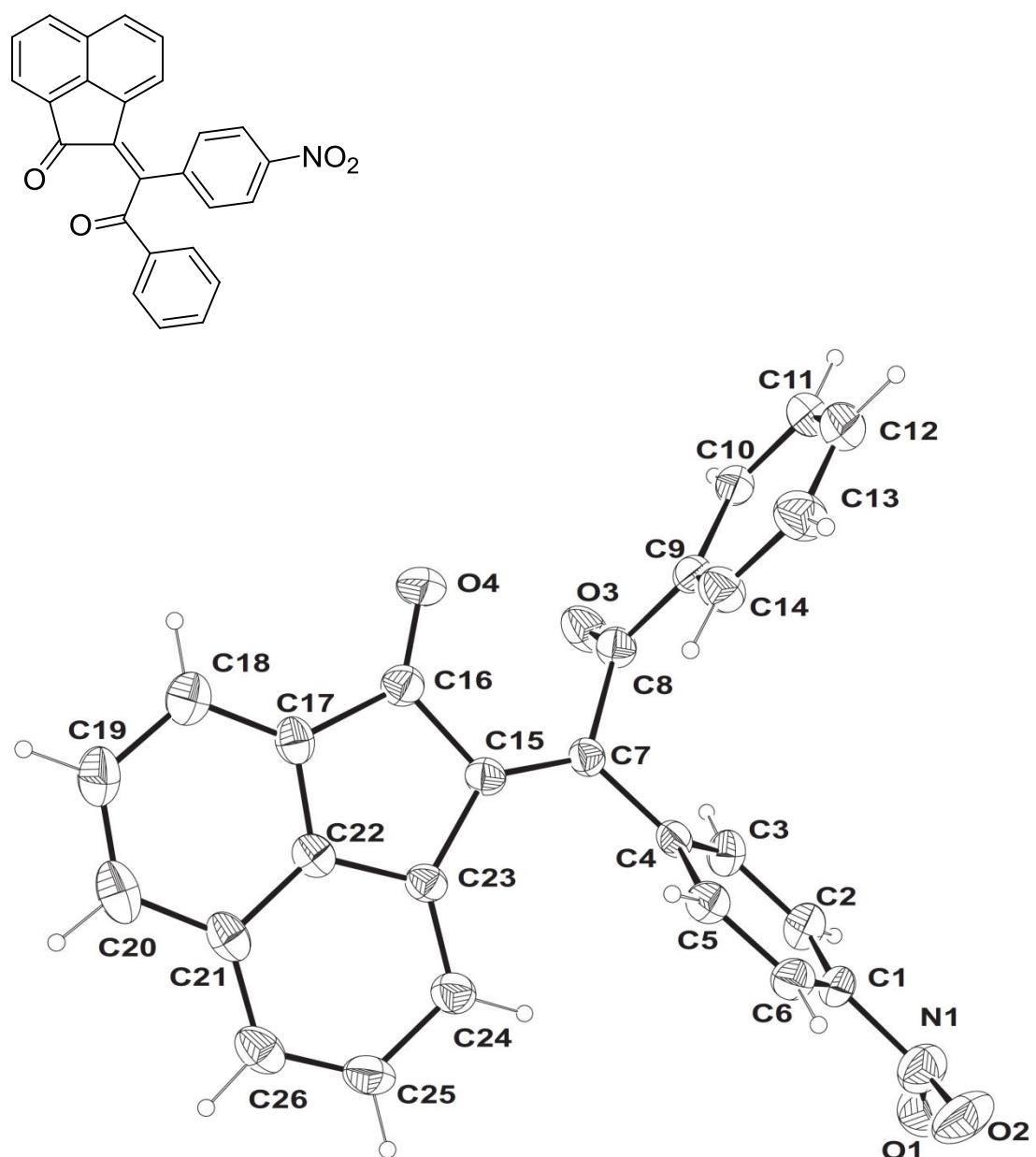
CCDC 935870 (1ab)



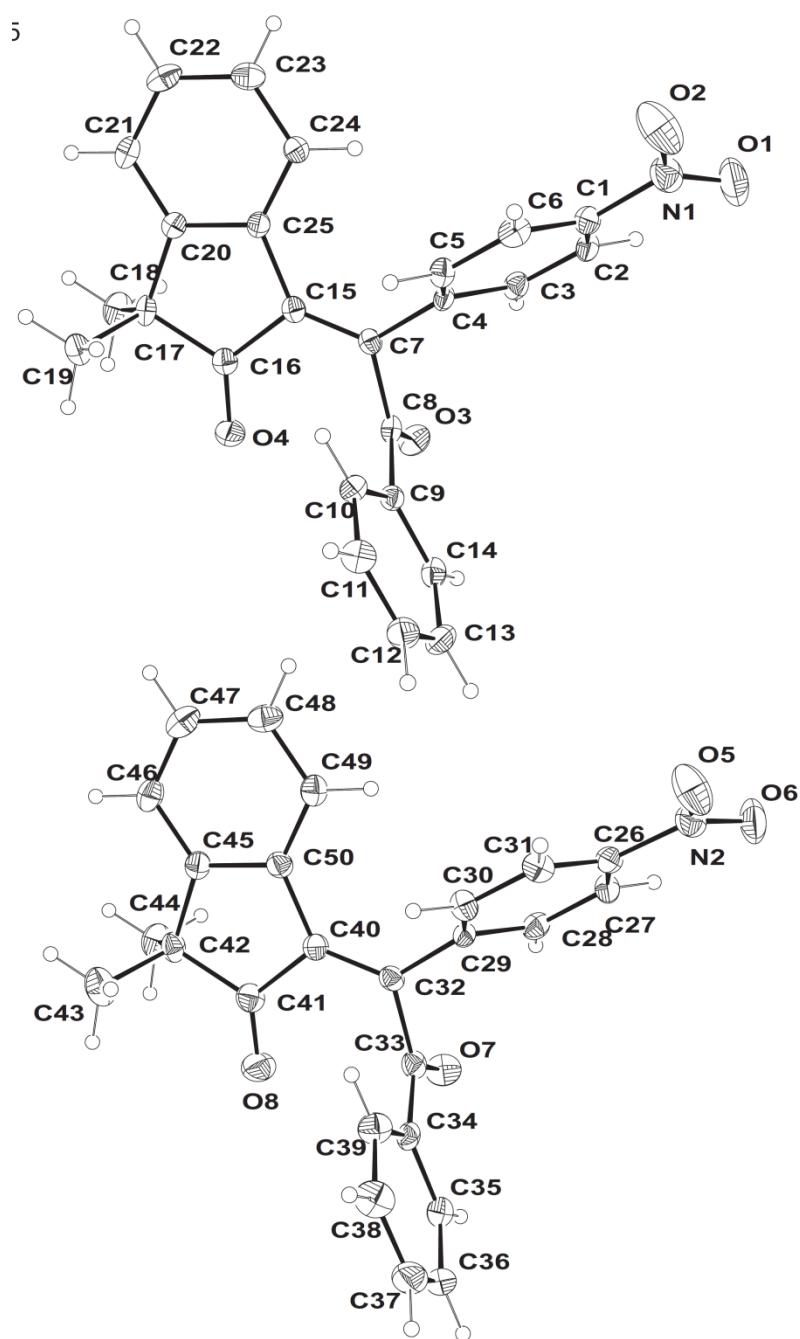
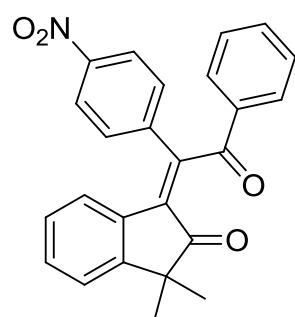
CCDC 935871 (2a)



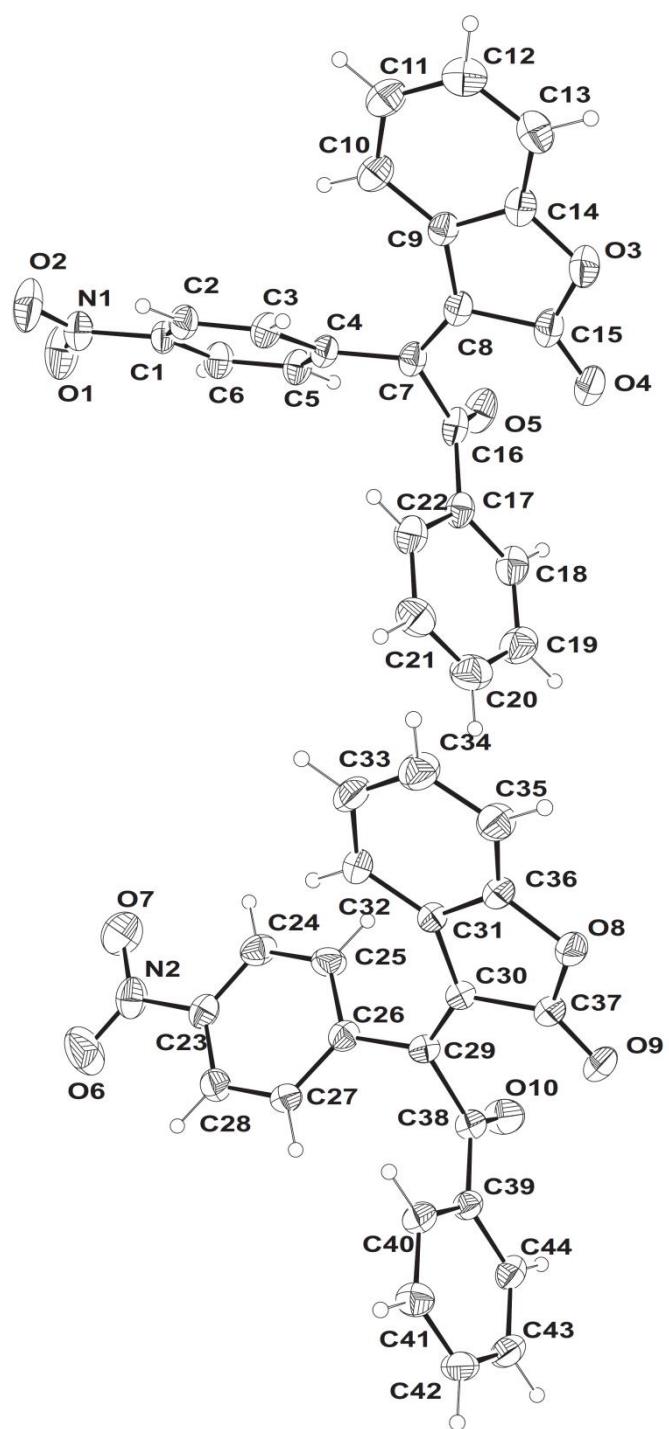
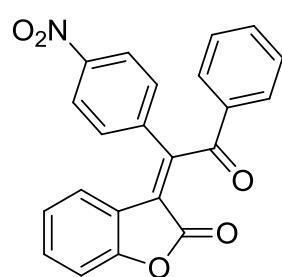
CCDC 935872 (3a)



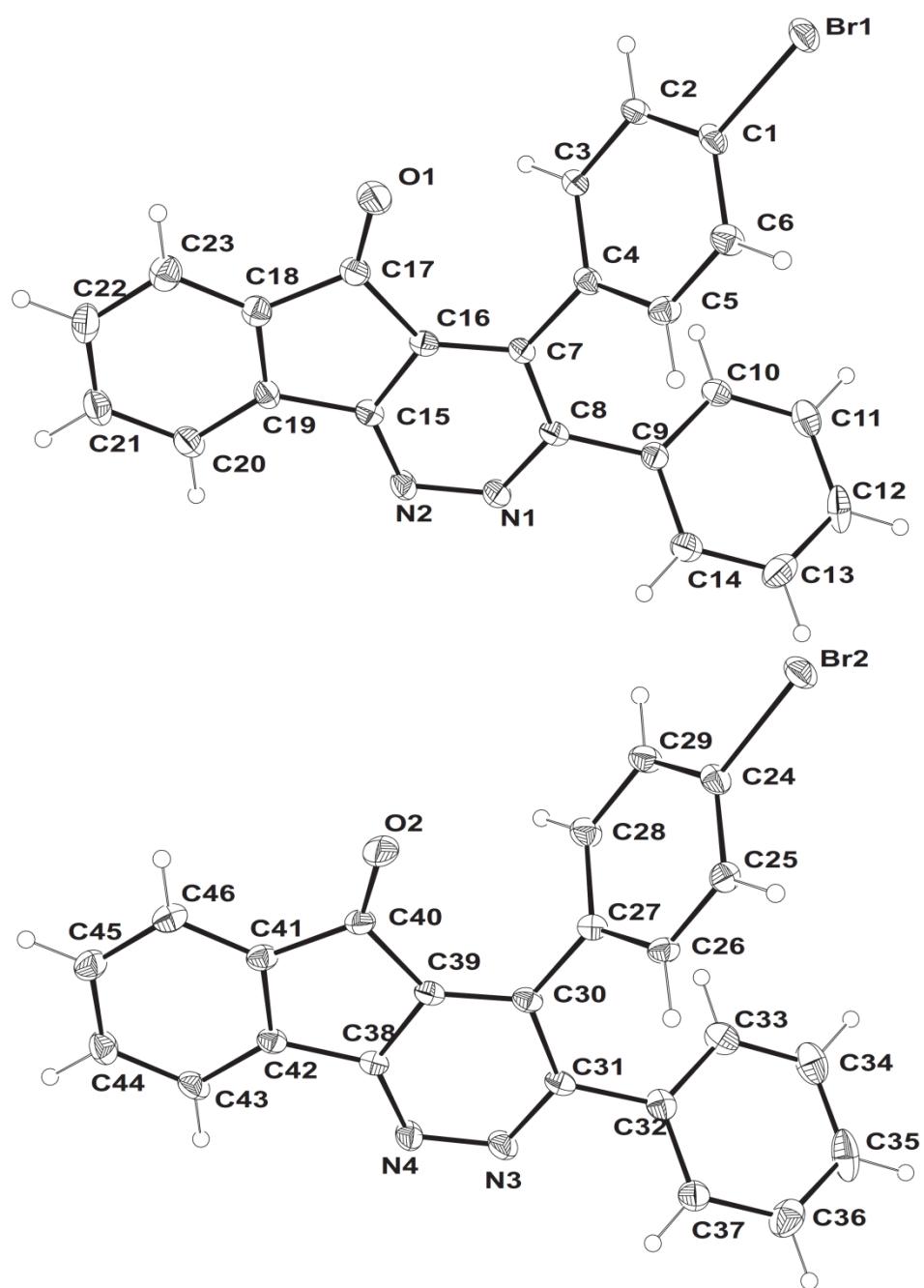
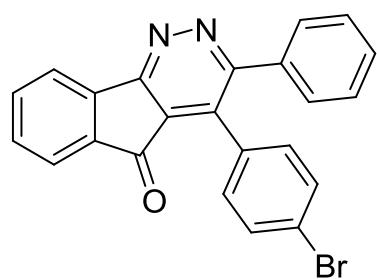
CCDC 935873 (4)



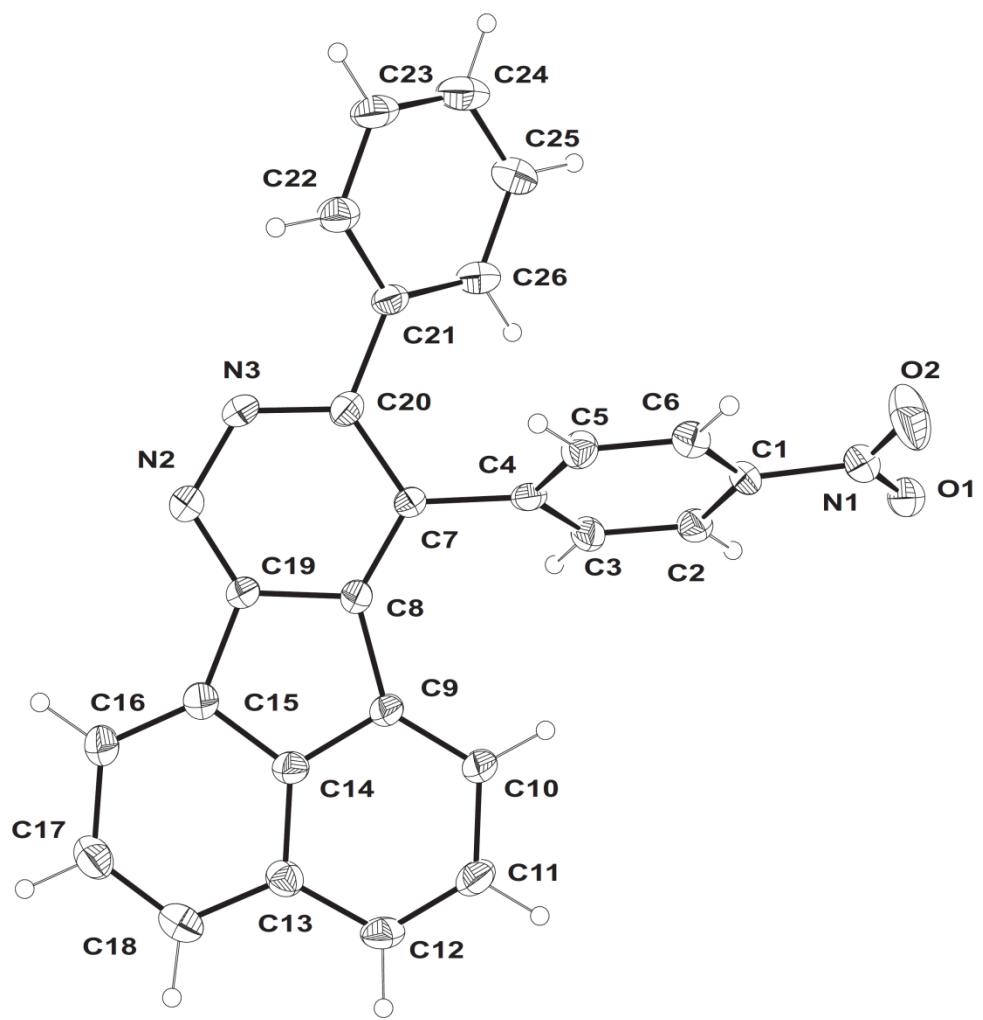
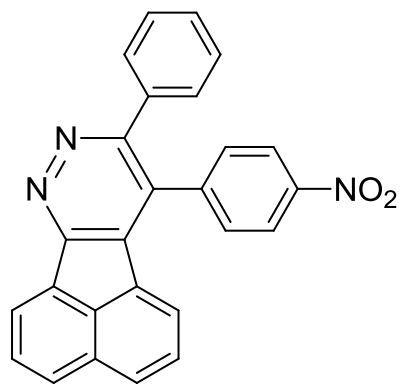
CCDC 937197 (5)



CCDC 935874 (17)



CCDC 935875 (18)



CCDC 935876 (9)

