2-Amino-2-methylpropionitrile (1) ^{1}H NMR spectrum

```
Current Data Parameters
NAME             06222007-2-rossM
EXPNO                          10
PROCNO                          1

F2 - Acquisition Parameters
Date_                    20070622
Time                        12.37
INSTRUM                  AVII400
PROBHD            5 mm PABBO BB-
PULPROG                     zg30
TD                         65536
SOLVENT                      D2O
NS                             8
DS                             2
SWH                     5597.015 Hz
FIDRES                  0.085404 Hz
AQ                     5.8545995 sec
RG                           287
DW                        89.333 usec
DE                          6.00 usec
TE                         300.0 K
D1                    1.00000000 sec
TD0                            1

======== CHANNEL f1 ========
NUC1                          1H
P1                         12.00 usec
PL1                        -2.00 dB
SFO1                 400.1324008 MHz

F2 - Processing parameters
SI                         32768
SF                   400.1300000 MHz
WDW                           EM
SSB                            0
LB                          0.30 Hz
GB                             0
PC                          1.00
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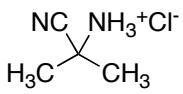
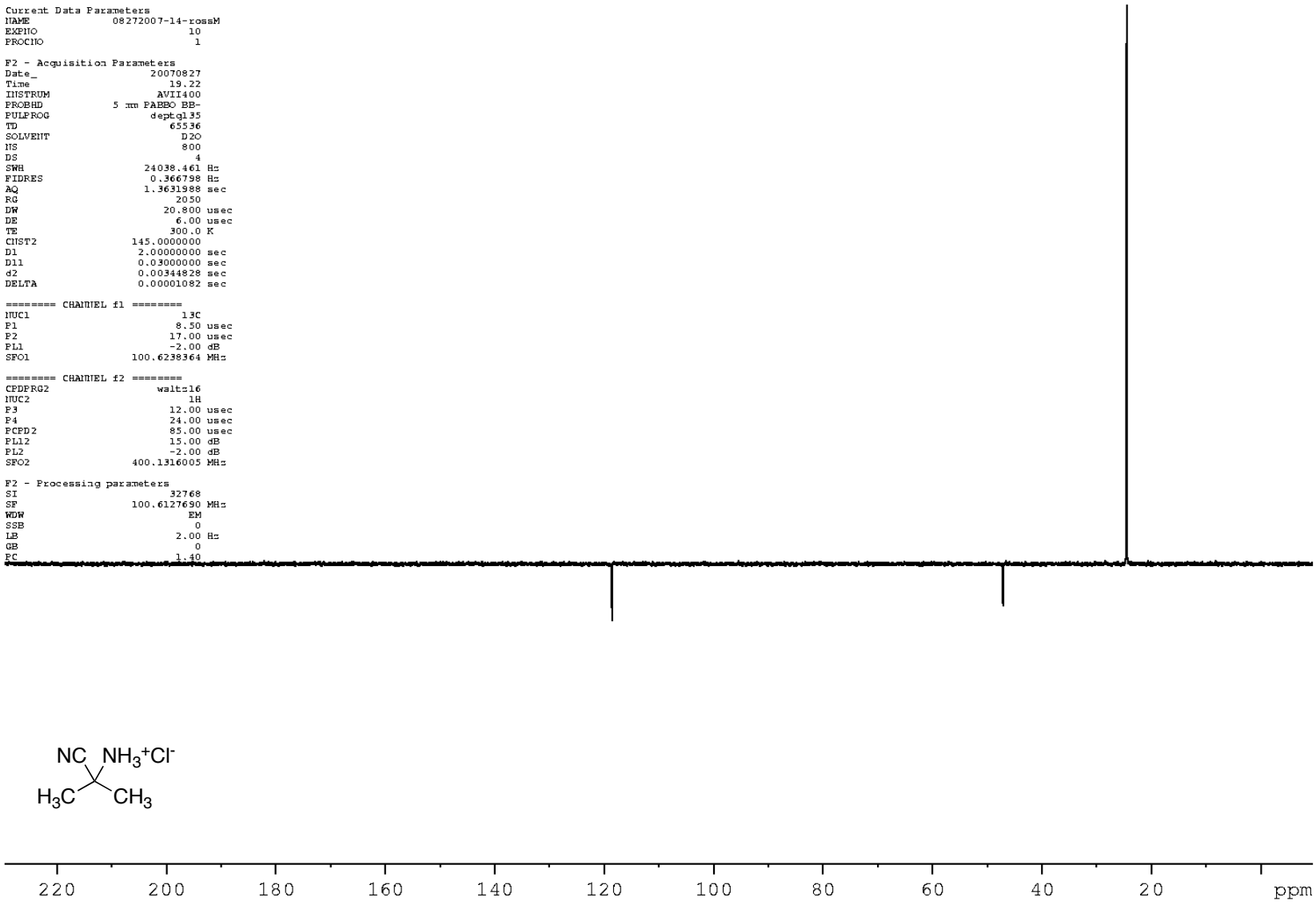


2-Amino-2-methylpropionitrile (1) ^{13}C NMR spectrum



2-Amino-2-methylheptanonitrile hydrochloride (5) ^{1}H NMR spectrum

```
Current Data Parameters
NAME             05162007-6-rossM
EXPNO                          10
PROCNO                          1

F2 - Acquisition Parameters
Date_                    20070516
Time                       16.10
INSTRUM                  AVII400
PROBHD            5 mm PABBO BB-
PULPROG                     zg30
TD                         65536
SOLVENT                      D2O
NS                             8
DS                             2
SWH                     5597.015 Hz
FIDRES                  0.085404 Hz
AQ                     5.8545995 sec
RG                           287
DW                        89.333 usec
DE                          6.00 usec
TE                         300.0 K
D1                   1.00000000 sec
TD0                            1

======== CHANNEL f1 ========
NUC1                          1H
P1                         12.00 usec
PL1                        -2.00 dB
SFO1                  400.1324008 MHz

F2 - Processing parameters
SI                         32768
SF                    400.1300000 MHz
WDW                           EM
SSB                            0
LB                          0.30 Hz
GB                             0
PC                          1.00
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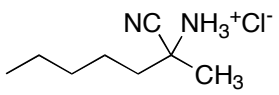
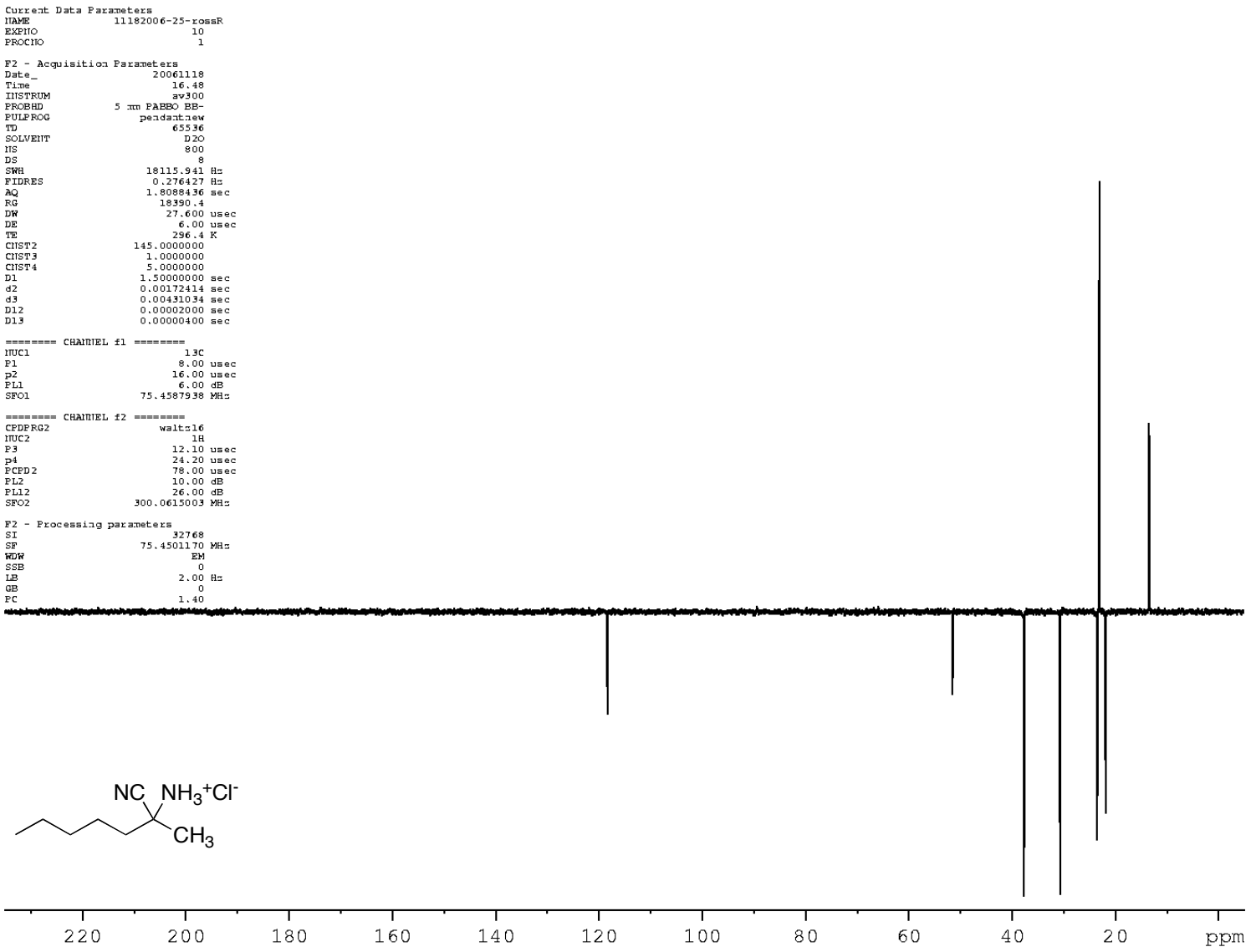


2-Amino-2-methylheptanonitrile hydrochloride (5) ^{13}C NMR spectrum



2-Amino-2,3,3-trimethylbutyronitrile hydrochloride (6) ^{1}H NMR spectrum

```
Current Data Parameters
NAME             11172006-6-ross
EXPNO                         10
PROCNO                         1

F2 - Acquisition Parameters
Date_                   20061117
Time                       11.29
INSTRUM                    av300
PROBHD           5 mm BBO BB-1H
PULPROG                     zg30
TD                         32768
SOLVENT                      D2O
NS                            16
DS                             2
SWH                     4194.631 Hz
FIDRES                  0.128010 Hz
AQ                     3.9059956 sec
RG                         645.1
DW                       119.200 usec
DE                         6.00 usec
TE                        294.5 K
D1                   1.00000000 sec
MCREST               0.00000000 sec
MCWRK                0.01500000 sec

======== CHANNEL f1 ========
NUC1                          1H
P1                          7.10 usec
PL1                         0.00 dB
SFO1                300.1318008 MHz

F2 - Processing parameters
SI                         32768
SF                  300.1300000 MHz
WDW                           EM
SSB                            0
LB                          0.30 Hz
GB                             0
PC                          1.00
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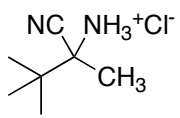
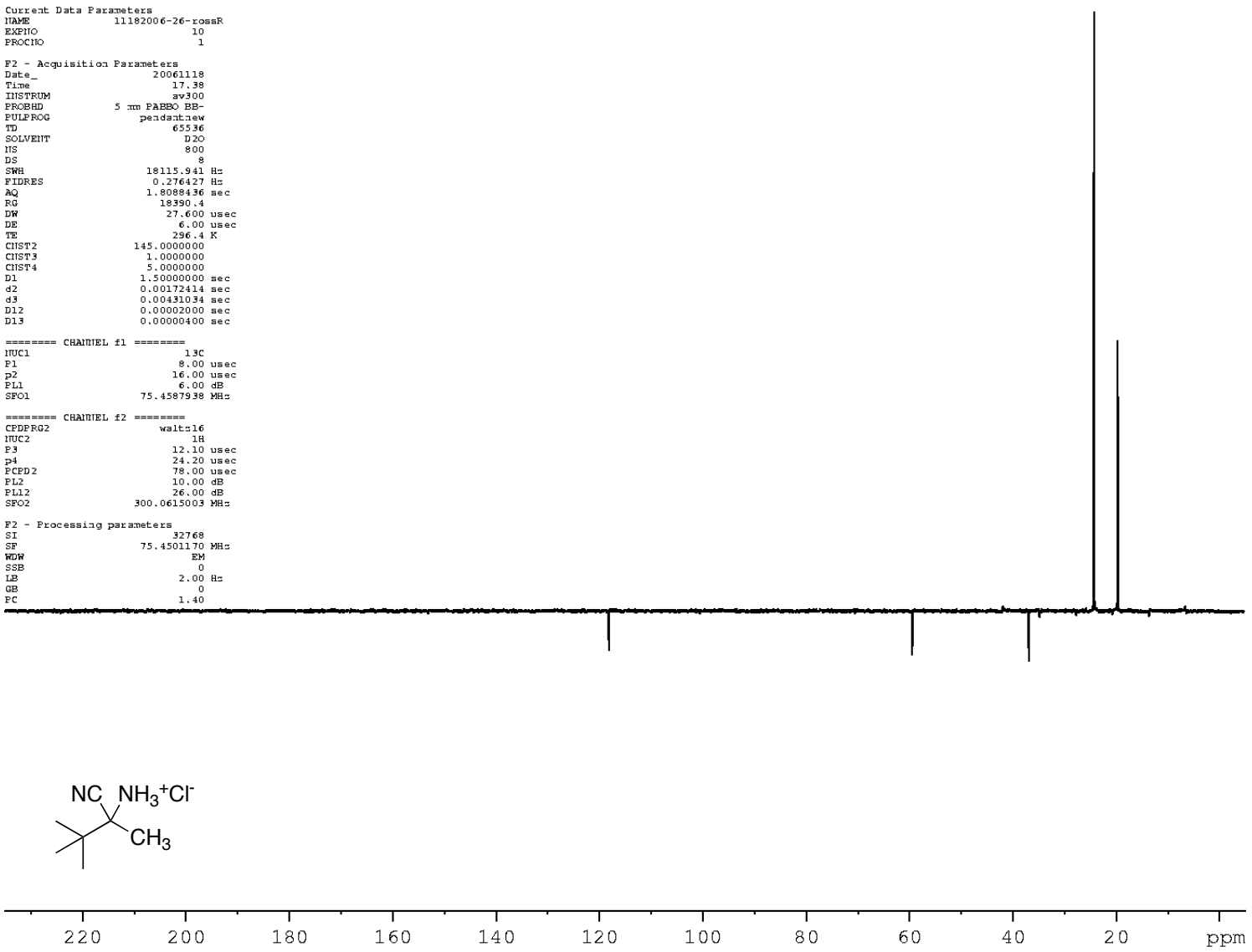


2-Amino-2,3,3-trimethylbutyronitrile hydrochloride (6) ^{13}C NMR spectrum



2-Amino-2-cyclopropylpropionitrile (2a) ^{1}H NMR spectrum

```
Current Data Parameters
NAME             06162007-25-rossM
EXPNO                           10
PROCNO                           1

F2 - Acquisition Parameters
Date_                     20070616
Time                         13.42
INSTRUM                   AVII400
PROBHD            5 mm PABBO BB-
PULPROG                       zg30
TD                           65536
SOLVENT                      CDCl3
NS                               8
DS                               2
SWH                       5597.015 Hz
FIDRES                    0.085404 Hz
AQ                        5.8545995 sec
RG                             287
DW                          89.333 usec
DE                            6.00 usec
TE                           300.0 K
D1                     1.00000000 sec
TD0                              1

======== CHANNEL f1 ========
NUC1                            1H
P1                           12.00 usec
PL1                          -2.00 dB
SFO1                  400.1324008 MHz

F2 - Processing parameters
SI                           32768
SF                    400.1300000 MHz
WDW                             EM
SSB                              0
LB                            0.30 Hz
GB                               0
PC                            1.00
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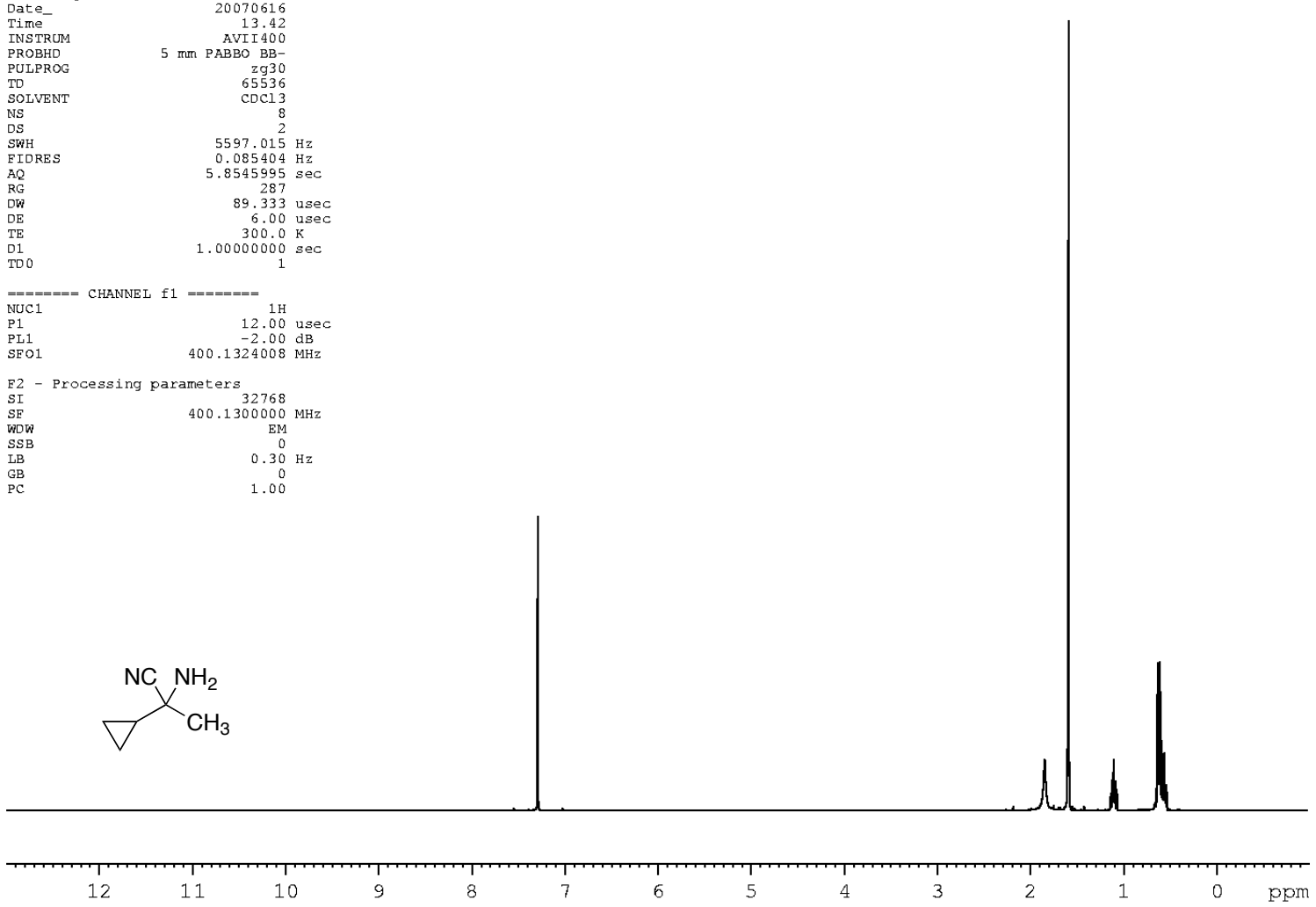
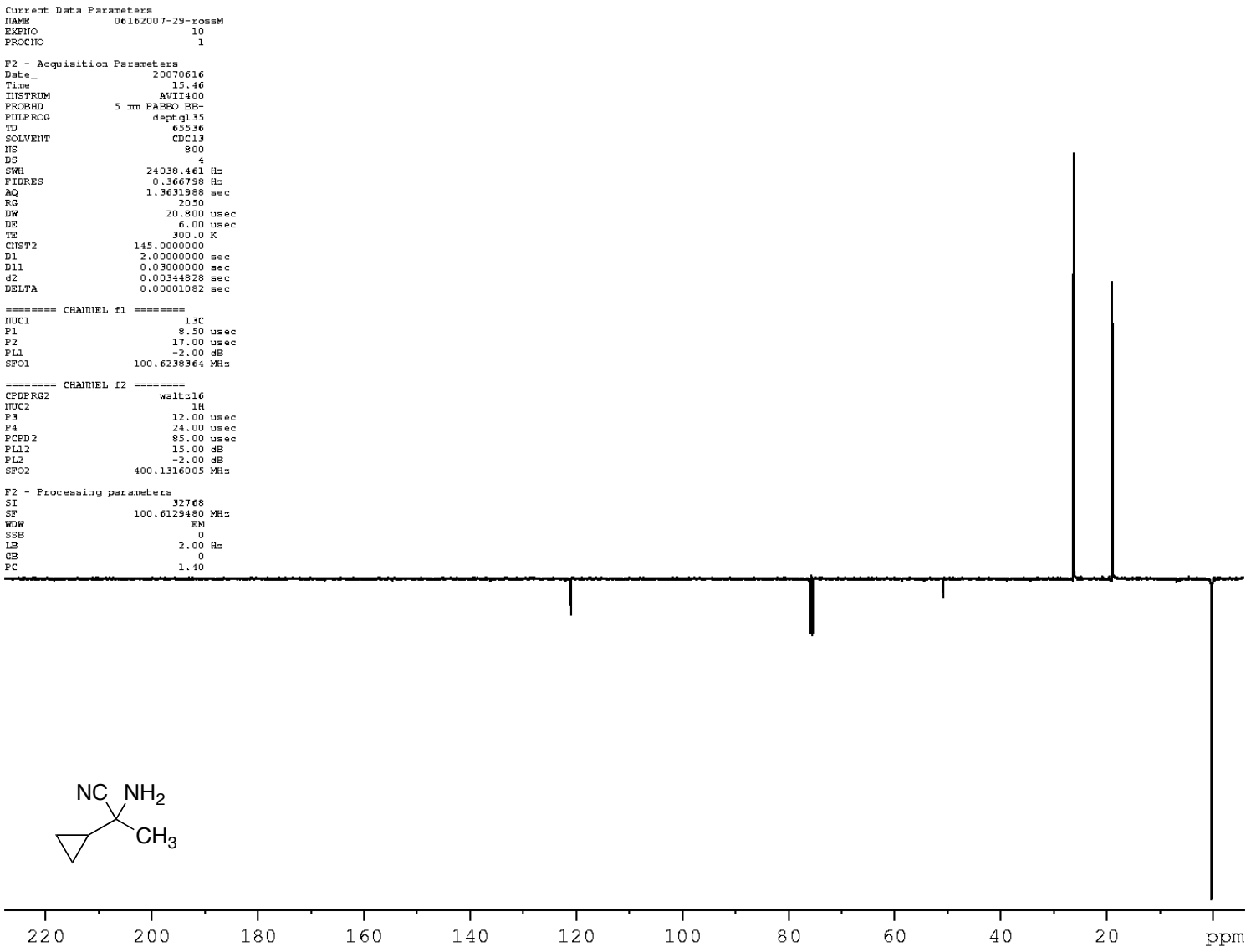


2-Amino-2-cyclopropylpropionitrile (2a) ^{13}C NMR spectrum



2-Amino-2-cyclopropylpropionitrile (2b) ^{1}H NMR spectrum

```
Current Data Parameters
NAME             09242007-31-rossM
EXPNO                          10
PROCNO                          1

F2 - Acquisition Parameters
Date_                    20070924
Time                        15.46
INSTRUM                  AVII400
PROBHD            5 mm PABBO BB-
PULPROG                     zg30
TD                         65536
SOLVENT                      D2O
NS                             8
DS                             2
SWH                     5597.015 Hz
FIDRES                  0.085404 Hz
AQ                     5.8545995 sec
RG                           287
DW                        89.333 usec
DE                         6.00 usec
TE                        300.0 K
D1                   1.00000000 sec
TD0                            1

======== CHANNEL f1 ========
NUC1                           1H
P1                         12.00 usec
PL1                        -2.00 dB
SFO1                400.1324008 MHz

F2 - Processing parameters
SI                         32768
SF                  400.1300000 MHz
WDW                           EM
SSB                            0
LB                          0.30 Hz
GB                             0
PC                          1.00
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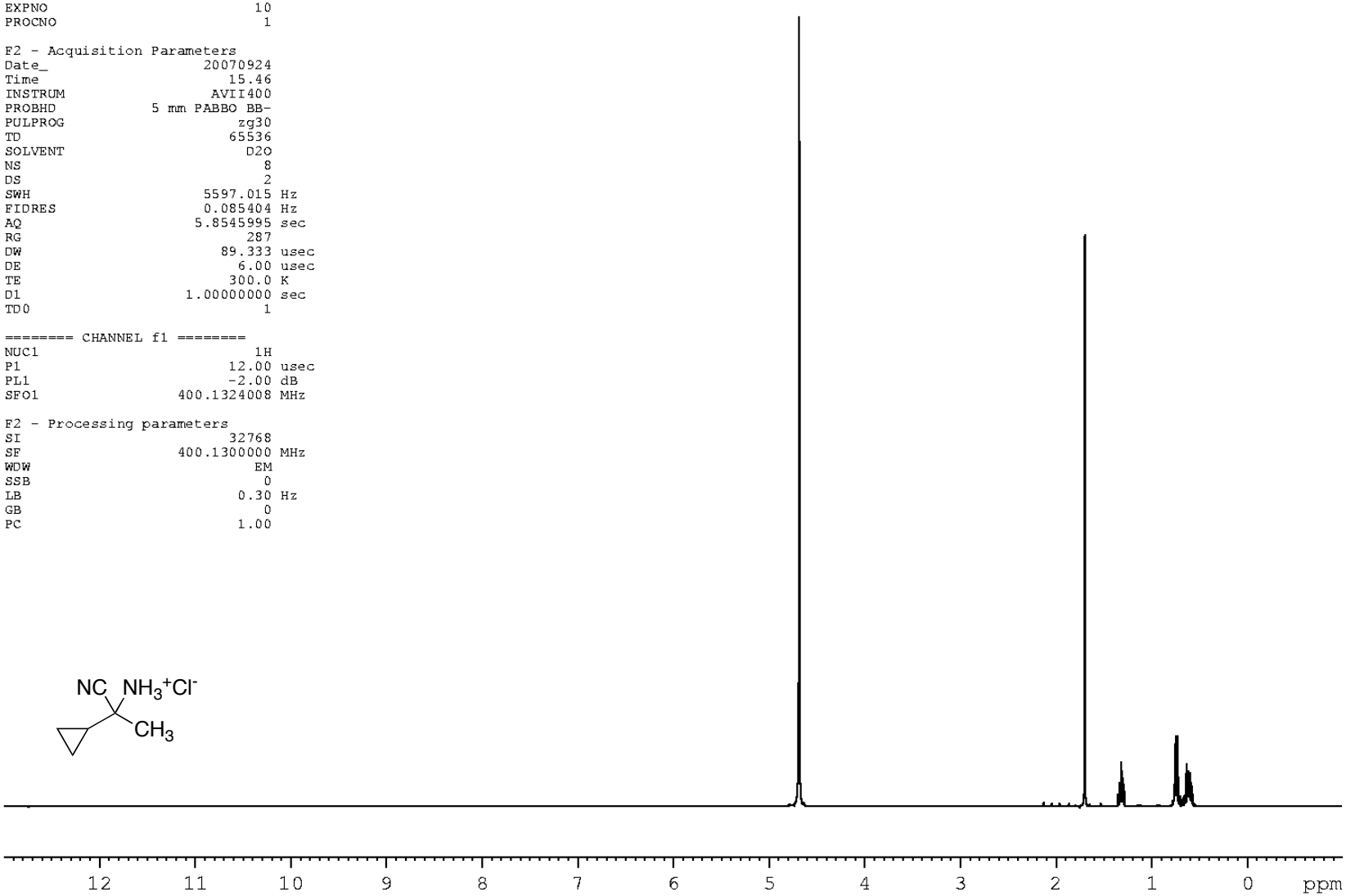
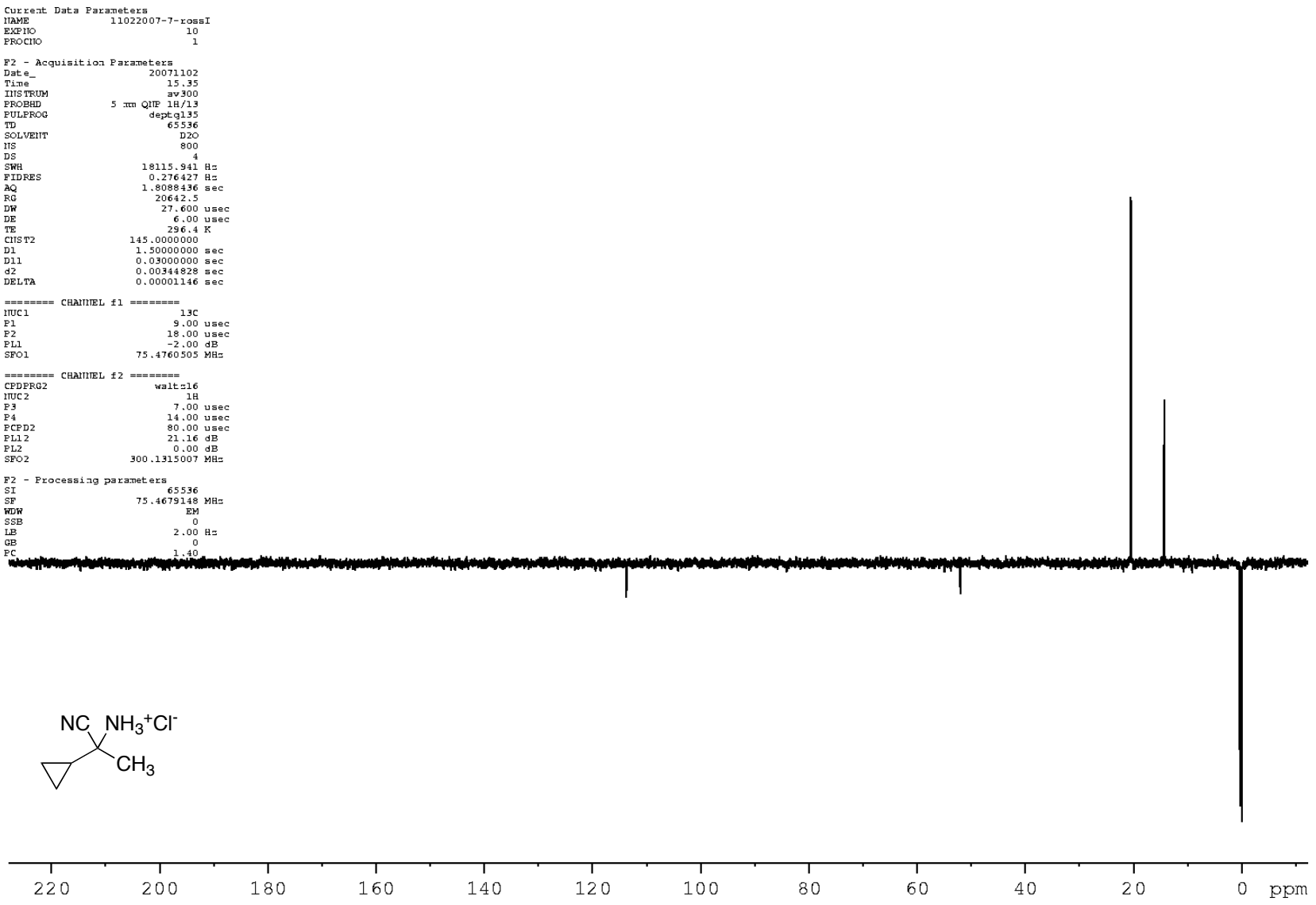


2-Amino-2-cyclopropylpropionitrile (2b) ^{13}C NMR spectrum



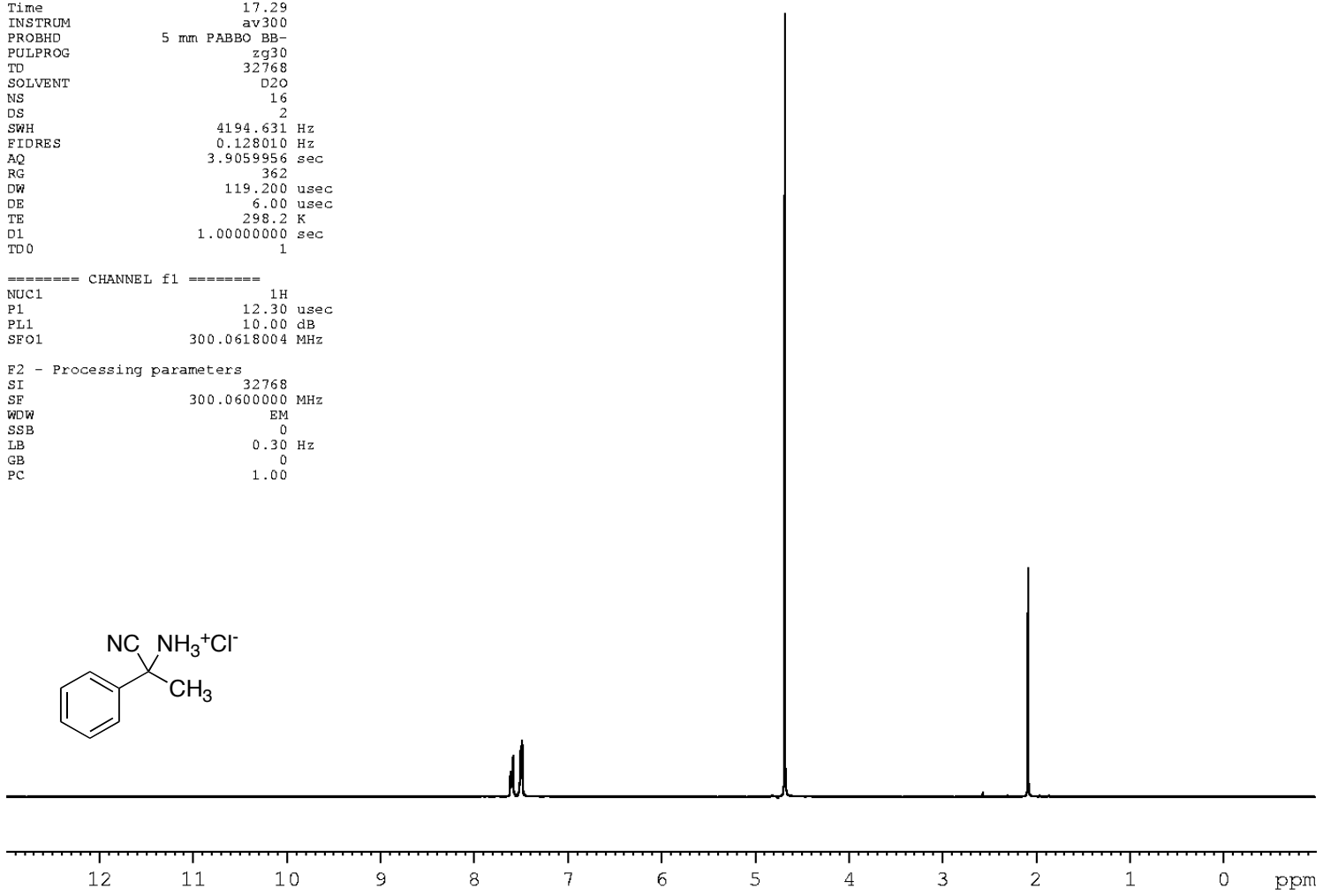
2-Amino-2-phenylpropionitrile hydrochloride (7) ^{1}H NMR spectrum

```
Current Data Parameters
NAME             08212006-2-ross
EXPNO                         10
PROCNO                         1

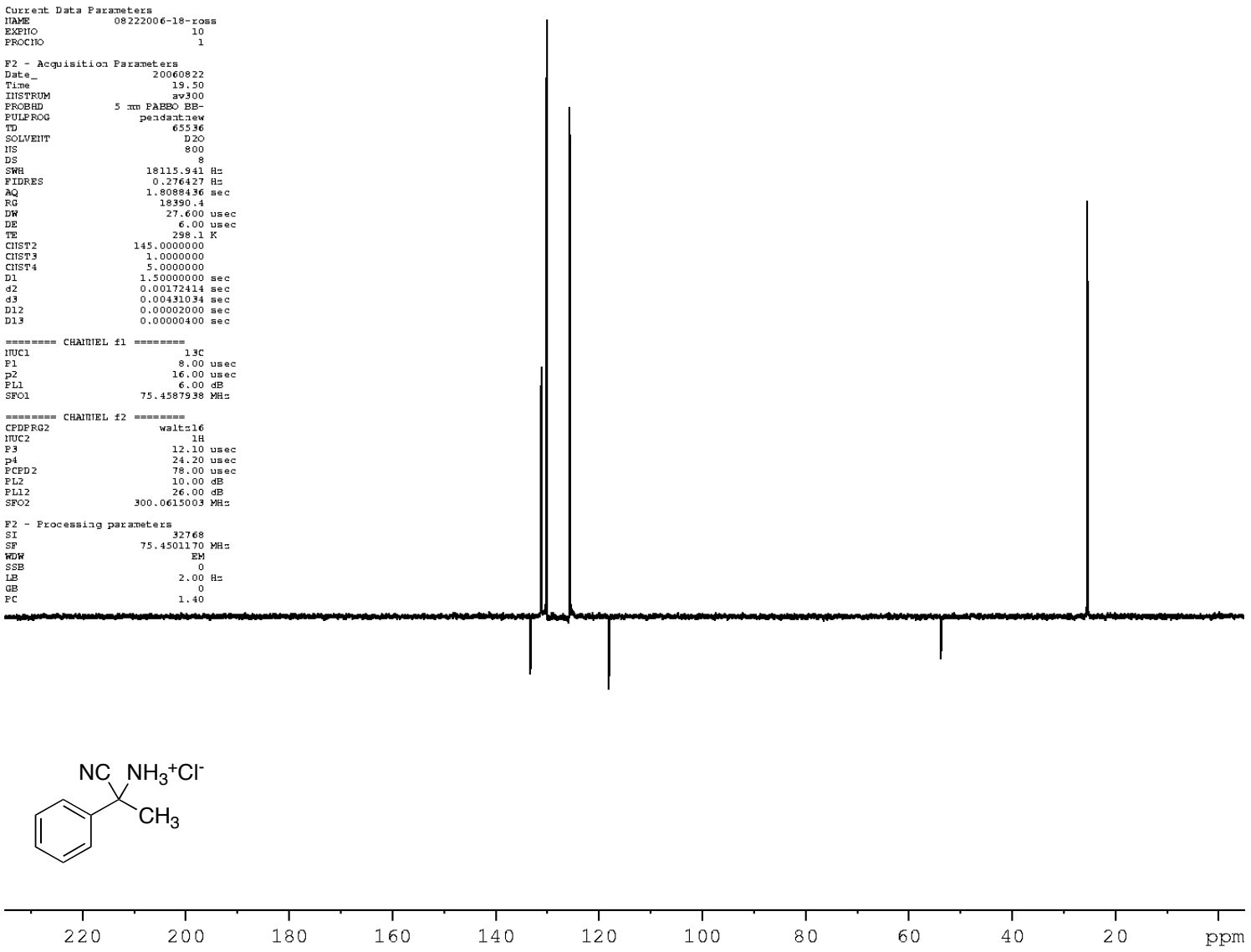
F2 - Acquisition Parameters
Date_                   20060821
Time                       17.29
INSTRUM                    av300
PROBHD           5 mm PABBO BB-
PULPROG                    zg30
TD                        32768
SOLVENT                     D2O
NS                           16
DS                            2
SWH                    4194.631 Hz
FIDRES                 0.128010 Hz
AQ                    3.9059956 sec
RG                          362
DW                      119.200 usec
DE                        6.00 usec
TE                       298.2 K
D1                  1.00000000 sec
TD0                           1

======== CHANNEL f1 ========
NUC1                         1H
P1                        12.30 usec
PL1                       10.00 dB
SFO1                 300.0618004 MHz

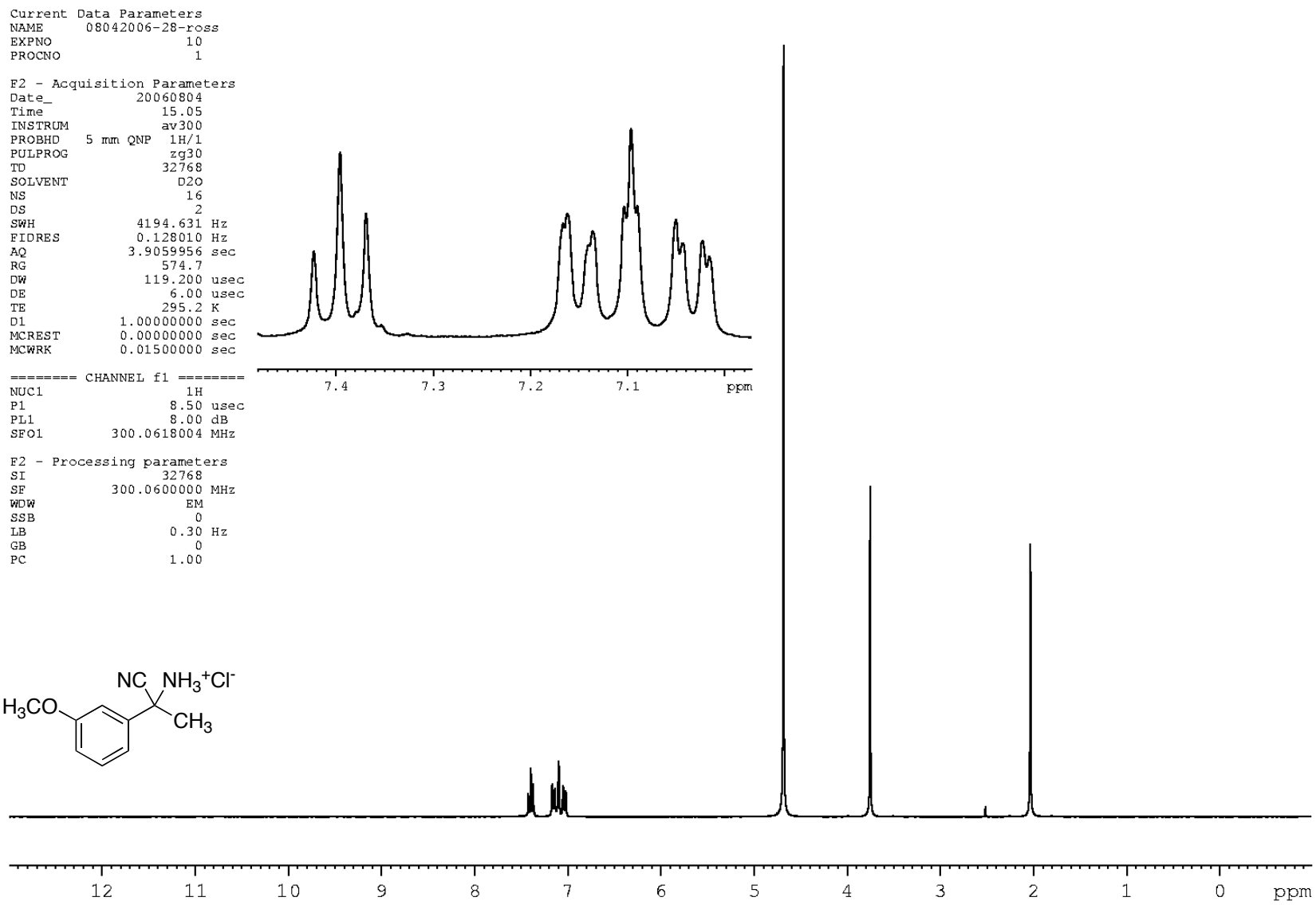
F2 - Processing parameters
SI                        32768
SF                   300.0600000 MHz
WDW                          EM
SSB                           0
LB                         0.30 Hz
GB                            0
PC                         1.00
```



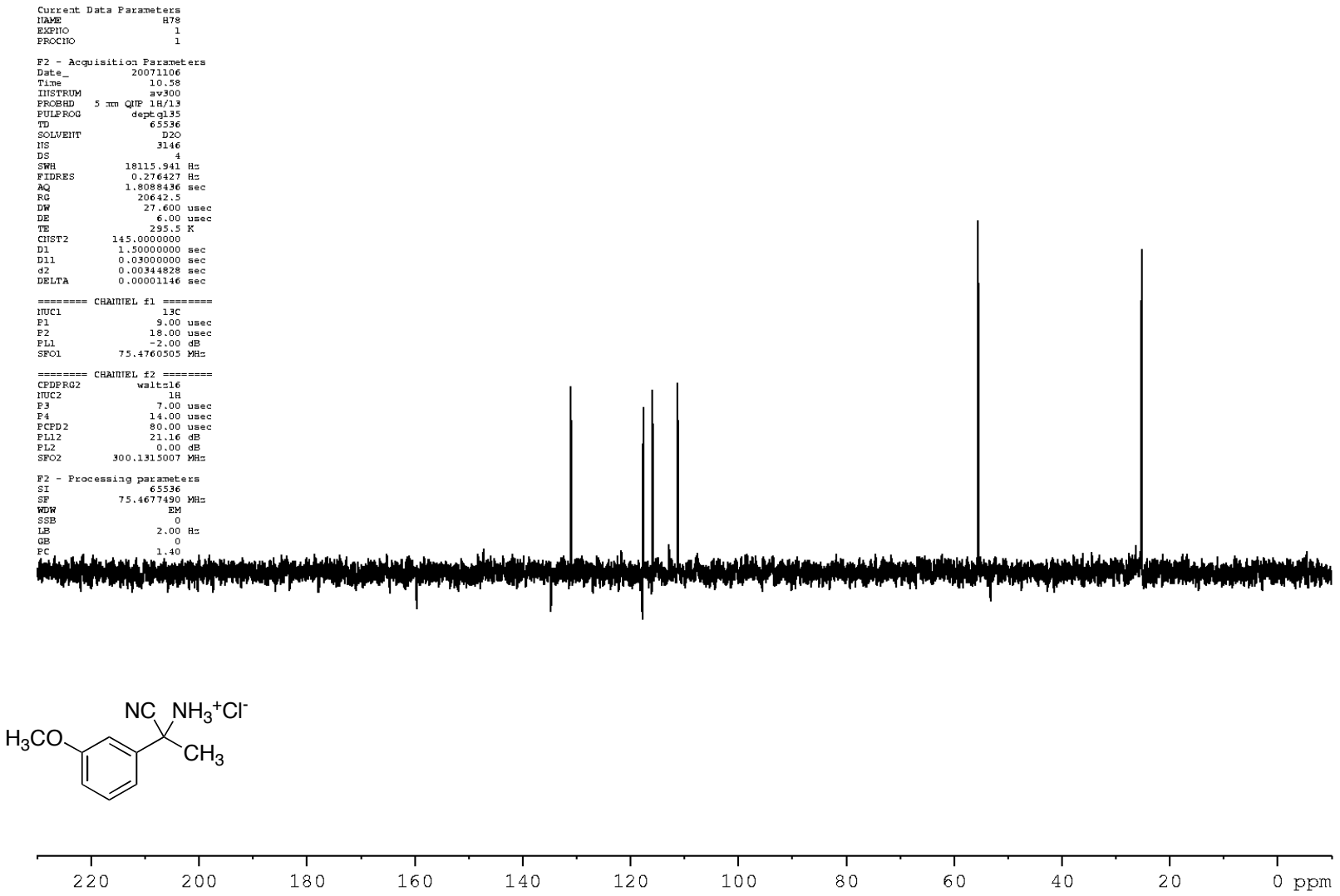
2-Amino-2-phenylpropionitrile hydrochloride (7) ^{13}C NMR spectrum



2-Amino-2-(3'-methoxyphenyl)propionitrile hydrochloride (8) ^{1}H NMR spectrum

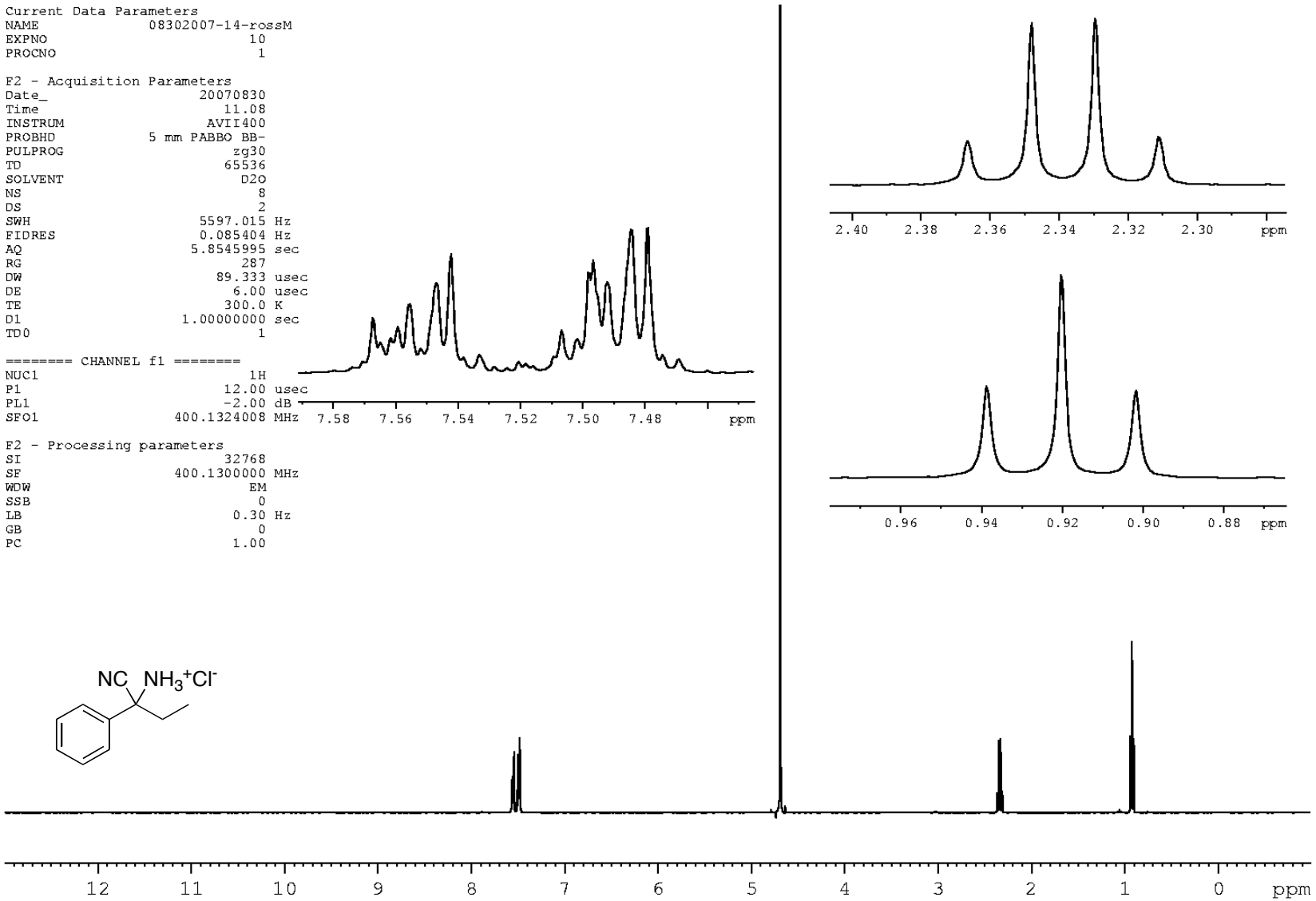


Crude 2-amino-2-(3'-methoxyphenyl)propionitrile hydrochloride (8) ^{13}C NMR spectrum

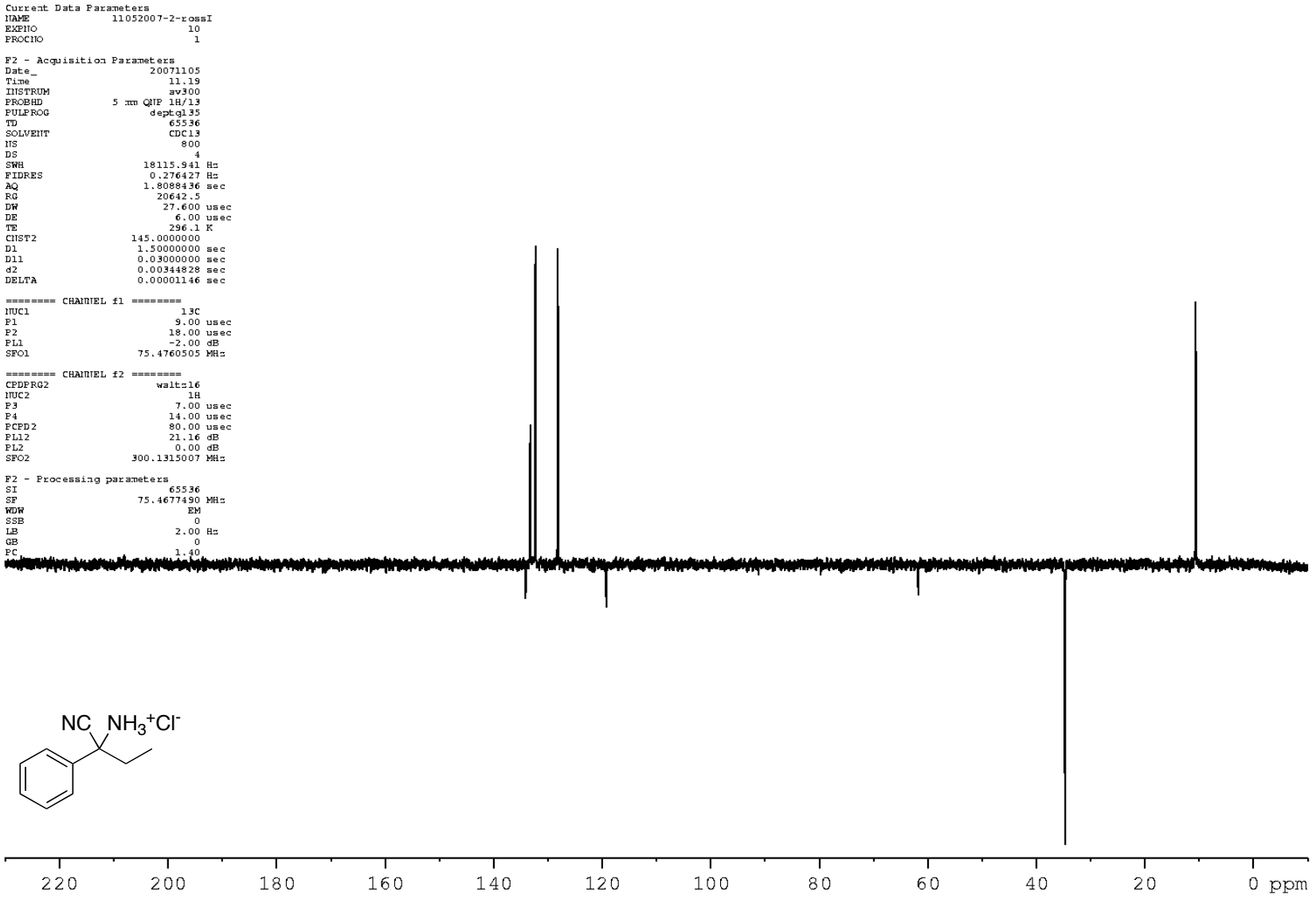


It proved difficult to obtain a clean ^{13}C spectrum of 2-amino-2-(3'-methoxyphenyl)propionitrile hydrochloride (**8**), most likely due to degradation of the compound while the spectrum was being obtained.

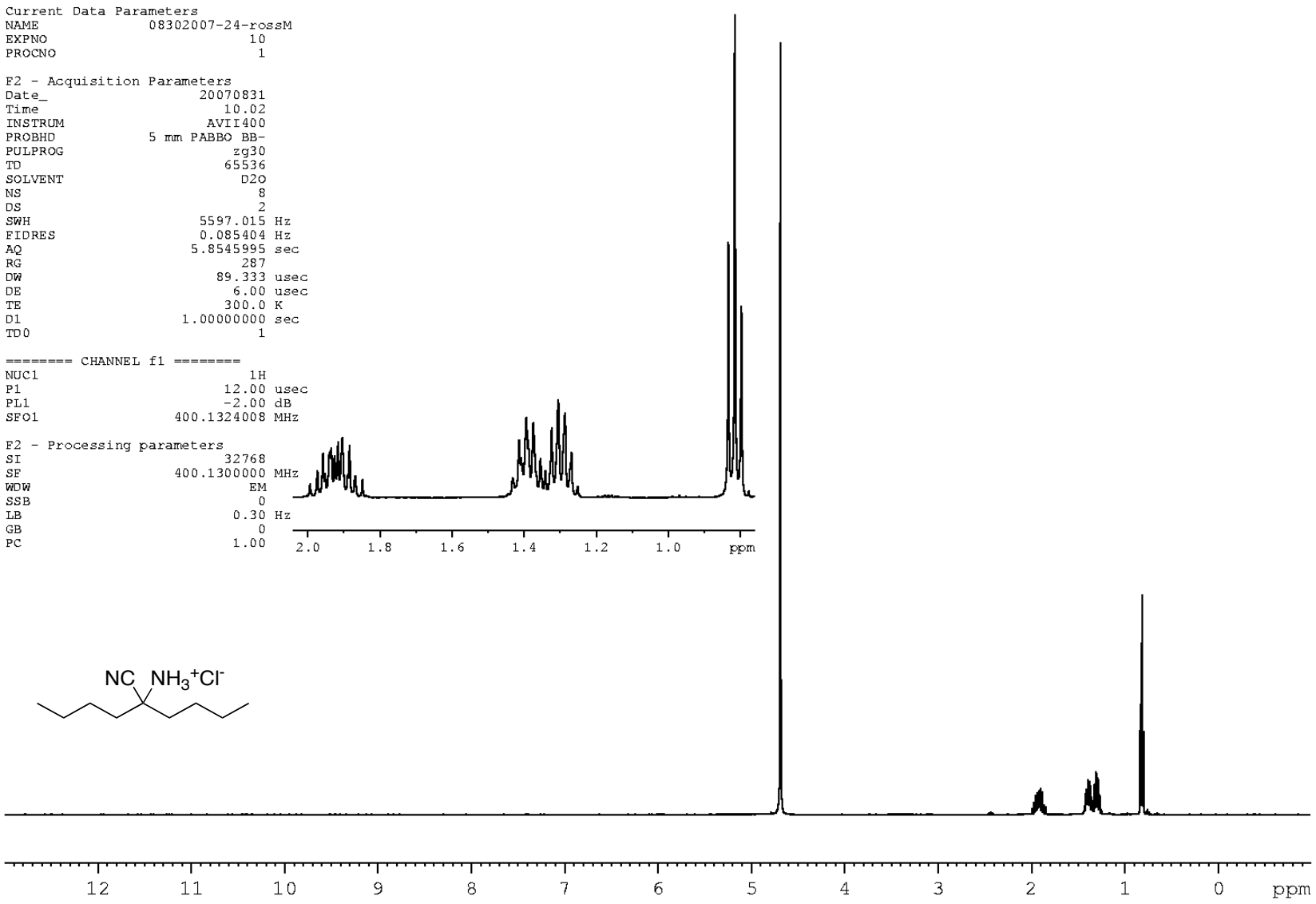
2-Amino-2-phenylbutyronitrile hydrochloride (9) ^{1}H NMR spectrum



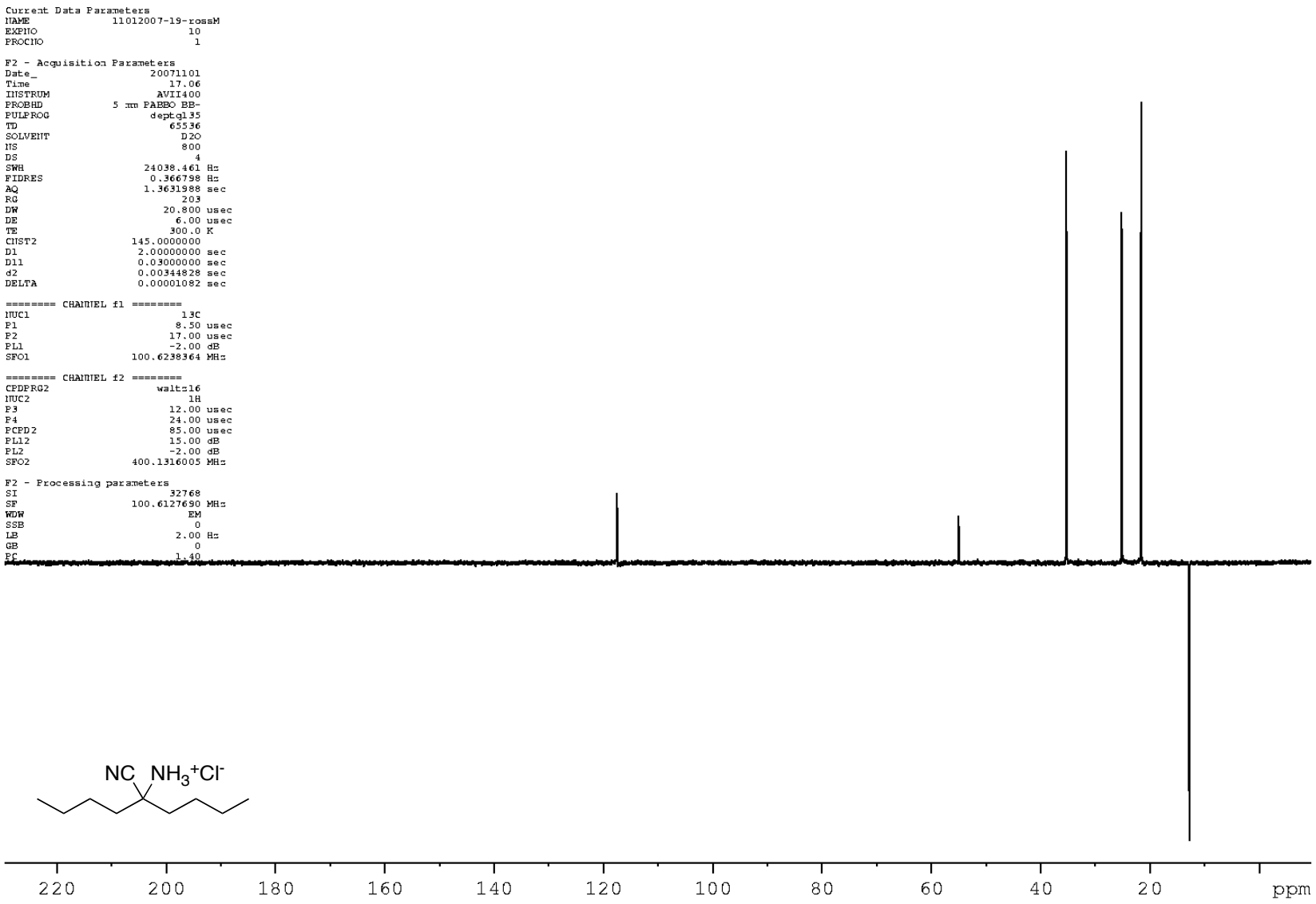
2-Amino-2-phenylbutyronitrile hydrochloride (9) ^{13}C NMR spectrum



2-Amino-2-*n*butylhexanenitrile hydrochloride (10) ^{1}H NMR spectrum



2-Amino-2-*n*butylhexanenitrile hydrochloride (10) ^{13}C NMR spectrum



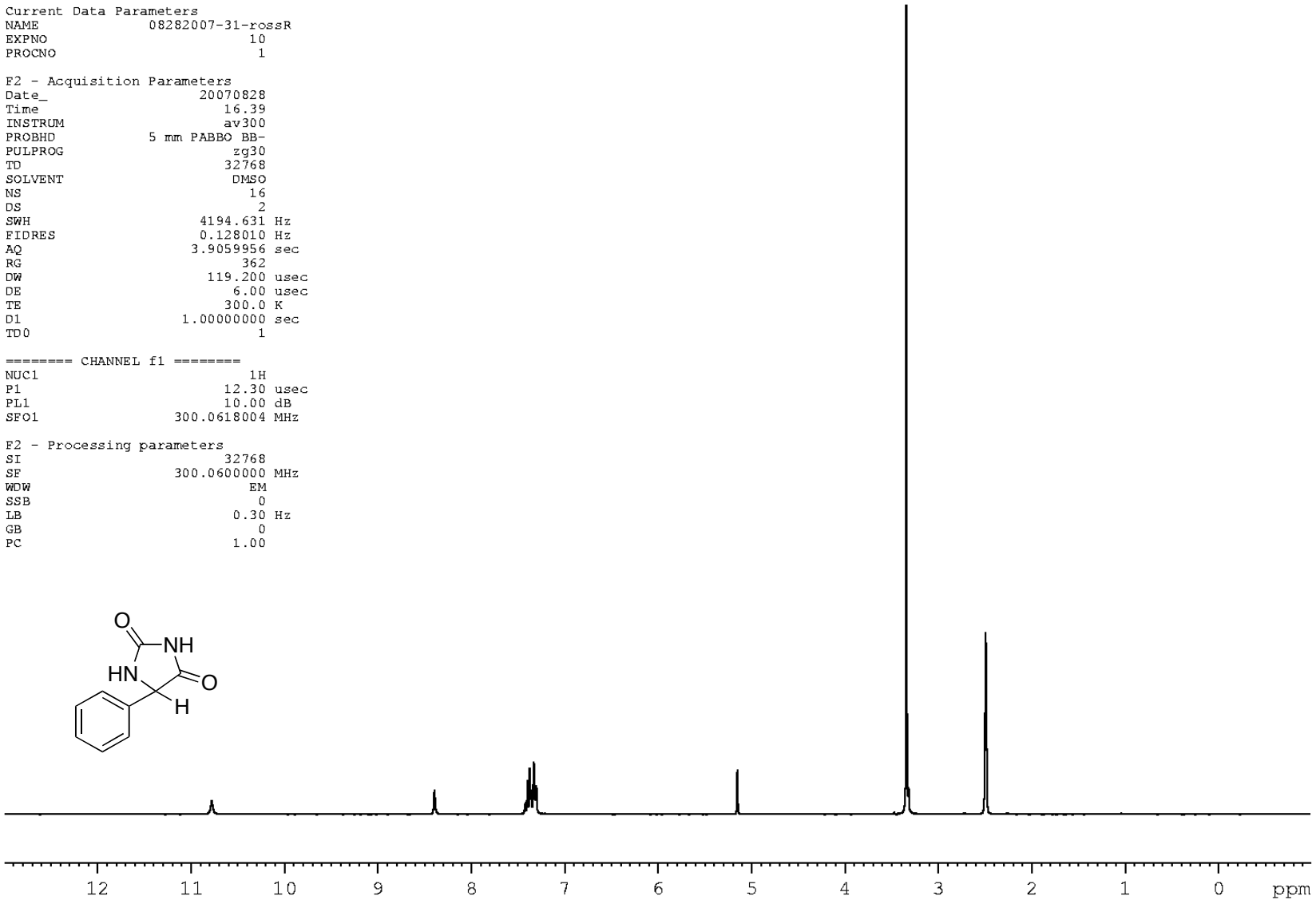
5-Phenylimidazolidine-2,4-dione (10) ^{1}H NMR spectrum

```
Current Data Parameters
NAME             08282007-31-rossR
EXPNO                           10
PROCNO                           1

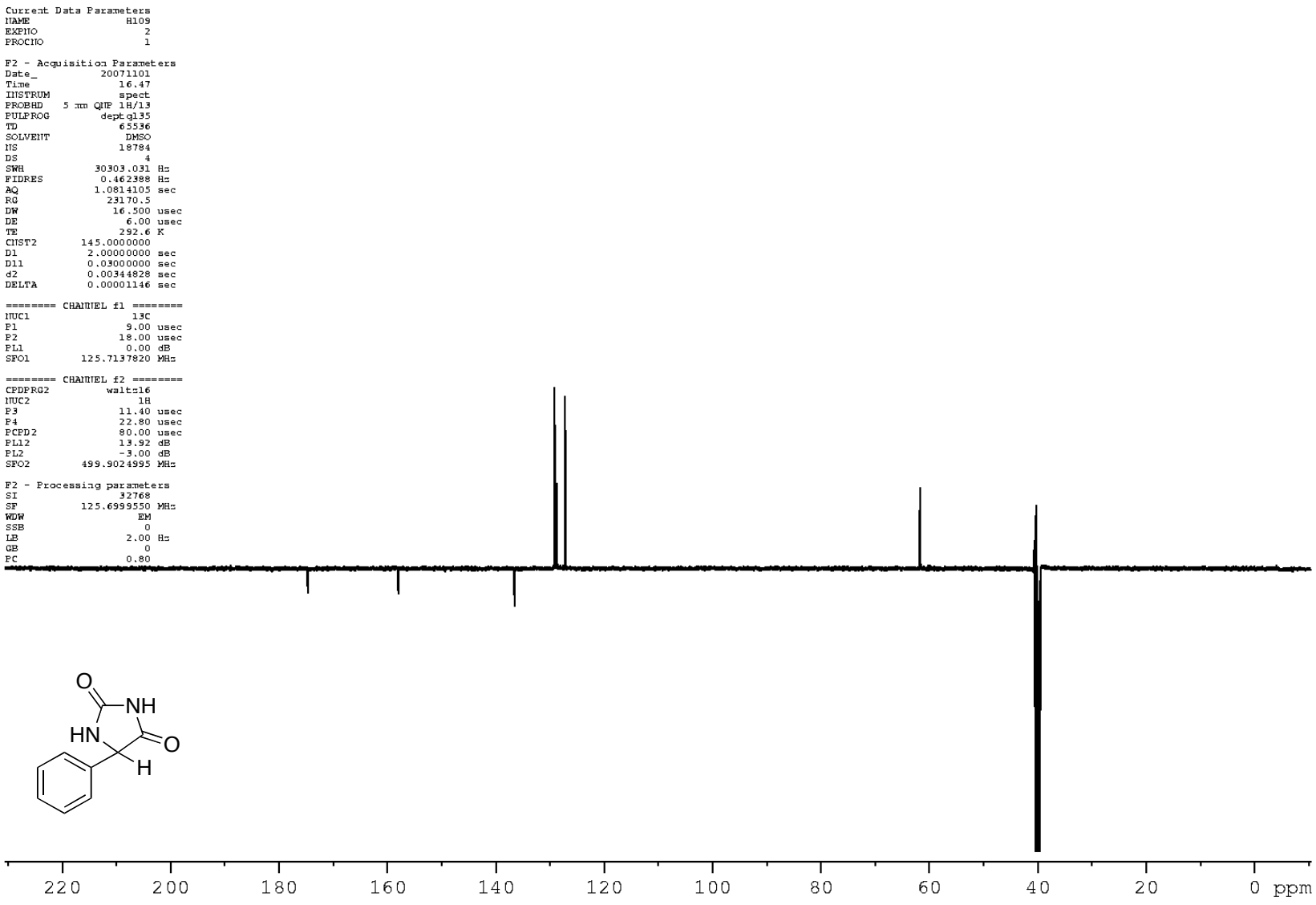
F2 - Acquisition Parameters
Date_                     20070828
Time                         16.39
INSTRUM                      av300
PROBHD             5 mm PABBO BB-
PULPROG                       zg30
TD                           32768
SOLVENT                       DMSO
NS                              16
DS                               2
SWH                       4194.631 Hz
FIDRES                    0.128010 Hz
AQ                       3.9059956 sec
RG                             362
DW                         119.200 usec
DE                           6.00 usec
TE                           300.0 K
D1                      1.00000000 sec
TD0                              1

======== CHANNEL f1 ========
NUC1                            1H
P1                           12.30 usec
PL1                          10.00 dB
SFO1                  300.0618004 MHz

F2 - Processing parameters
SI                           32768
SF                    300.0600000 MHz
WDW                             EM
SSB                              0
LB                            0.30 Hz
GB                               0
PC                            1.00
```



5-Phenylimidazolidine-2,4-dione (10) ^{13}C NMR spectrum



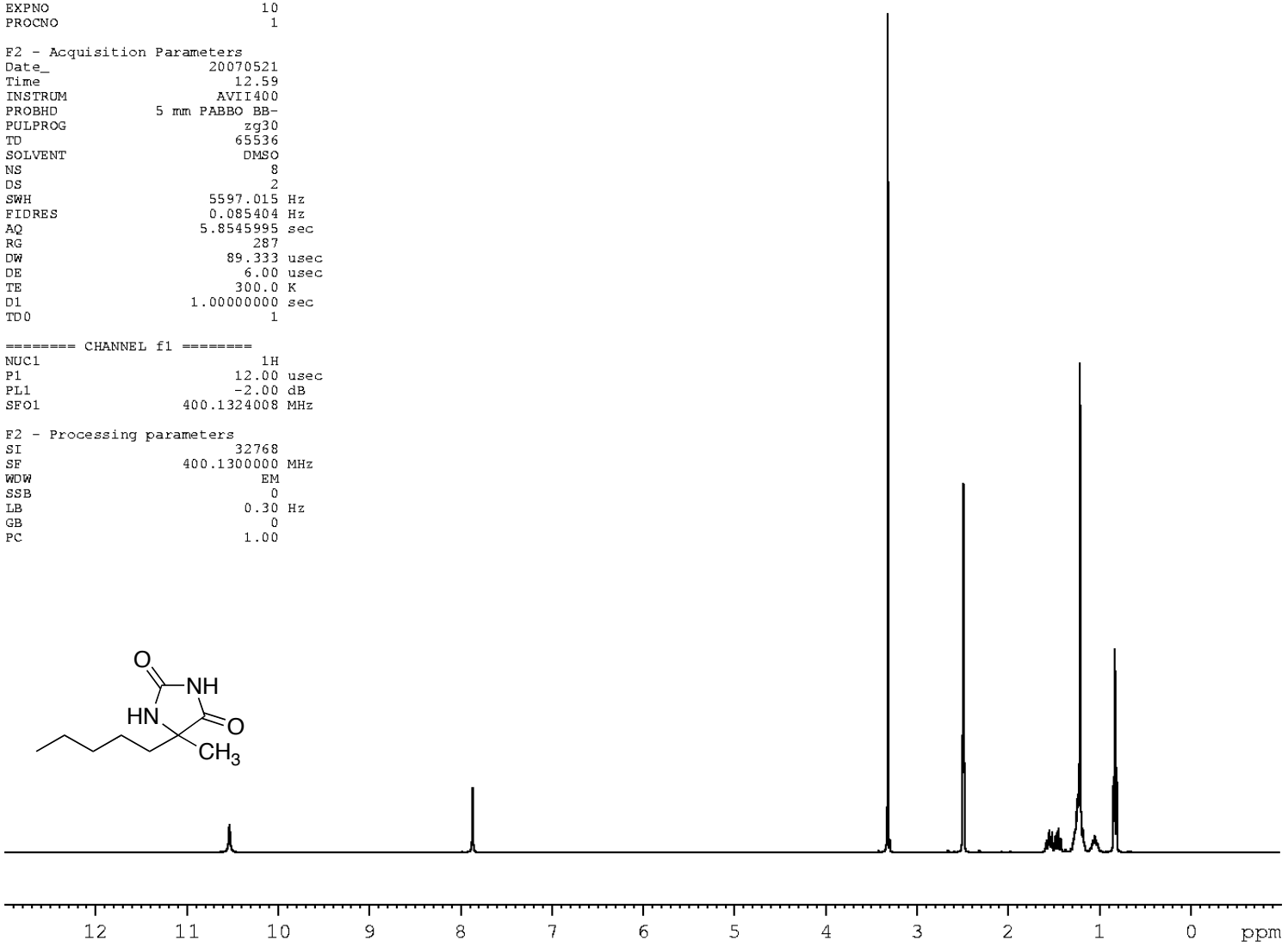
5-Methyl-5-pentylimidazolidine-2,4-dione (12) ^{1}H NMR spectrum

```
Current Data Parameters
NAME             05212007-17-rossM
EXPNO                           10
PROCNO                           1

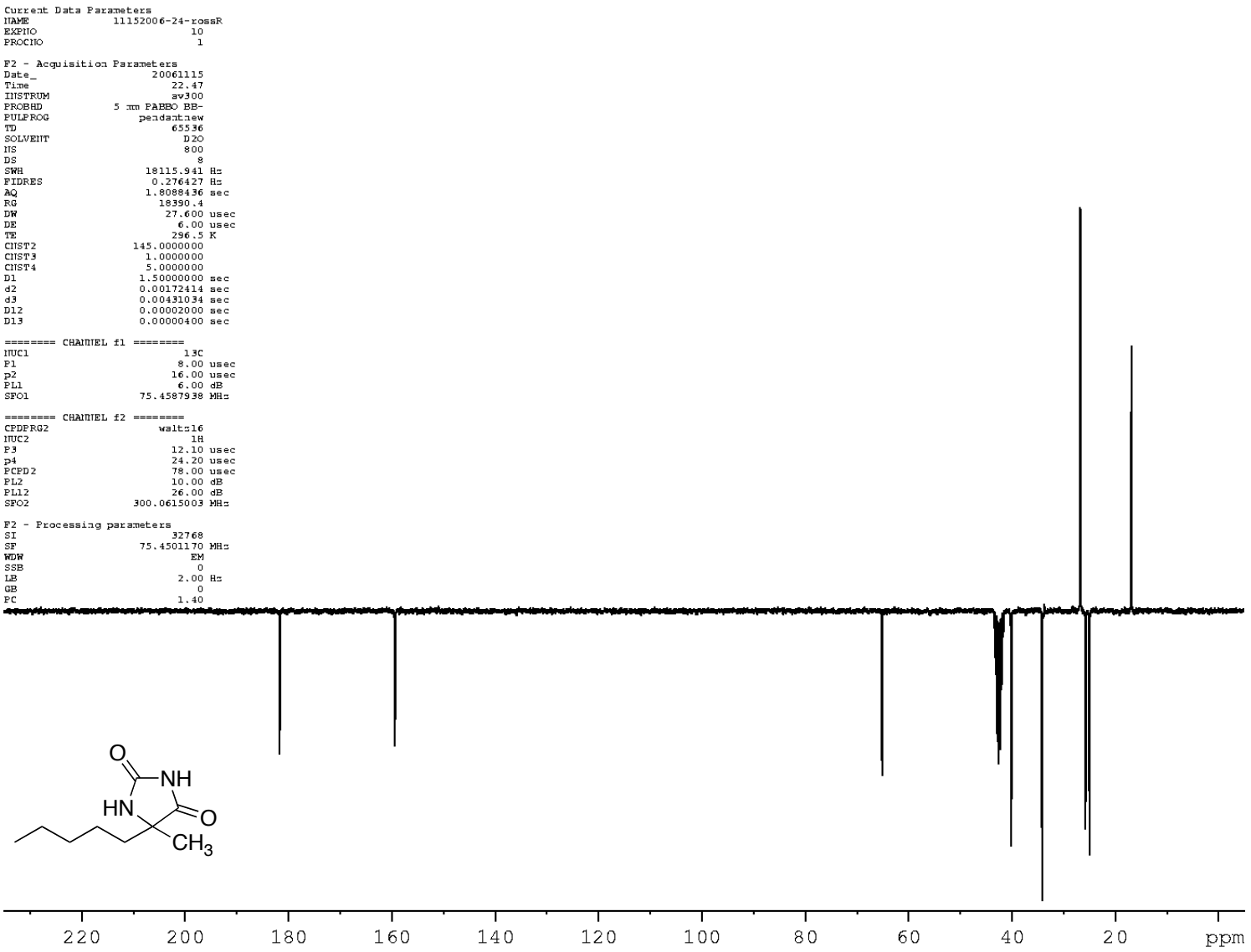
F2 - Acquisition Parameters
Date_                     20070521
Time                         12.59
INSTRUM                   AVII400
PROBHD            5 mm PABBO BB-
PULPROG                       zg30
TD                           65536
SOLVENT                       DMSO
NS                               8
DS                               2
SWH                       5597.015 Hz
FIDRES                    0.085404 Hz
AQ                        5.8545995 sec
RG                             287
DW                          89.333 usec
DE                            6.00 usec
TE                           300.0 K
D1                     1.00000000 sec
TD0                              1

======== CHANNEL f1 ========
NUC1                            1H
P1                           12.00 usec
PL1                          -2.00 dB
SFO1                  400.1324008 MHz

F2 - Processing parameters
SI                           32768
SF                    400.1300000 MHz
WDW                             EM
SSB                              0
LB                            0.30 Hz
GB                               0
PC                            1.00
```



5-Methyl-5-pentylimidazolidine-2,4-dione (12) ^{13}C NMR spectrum



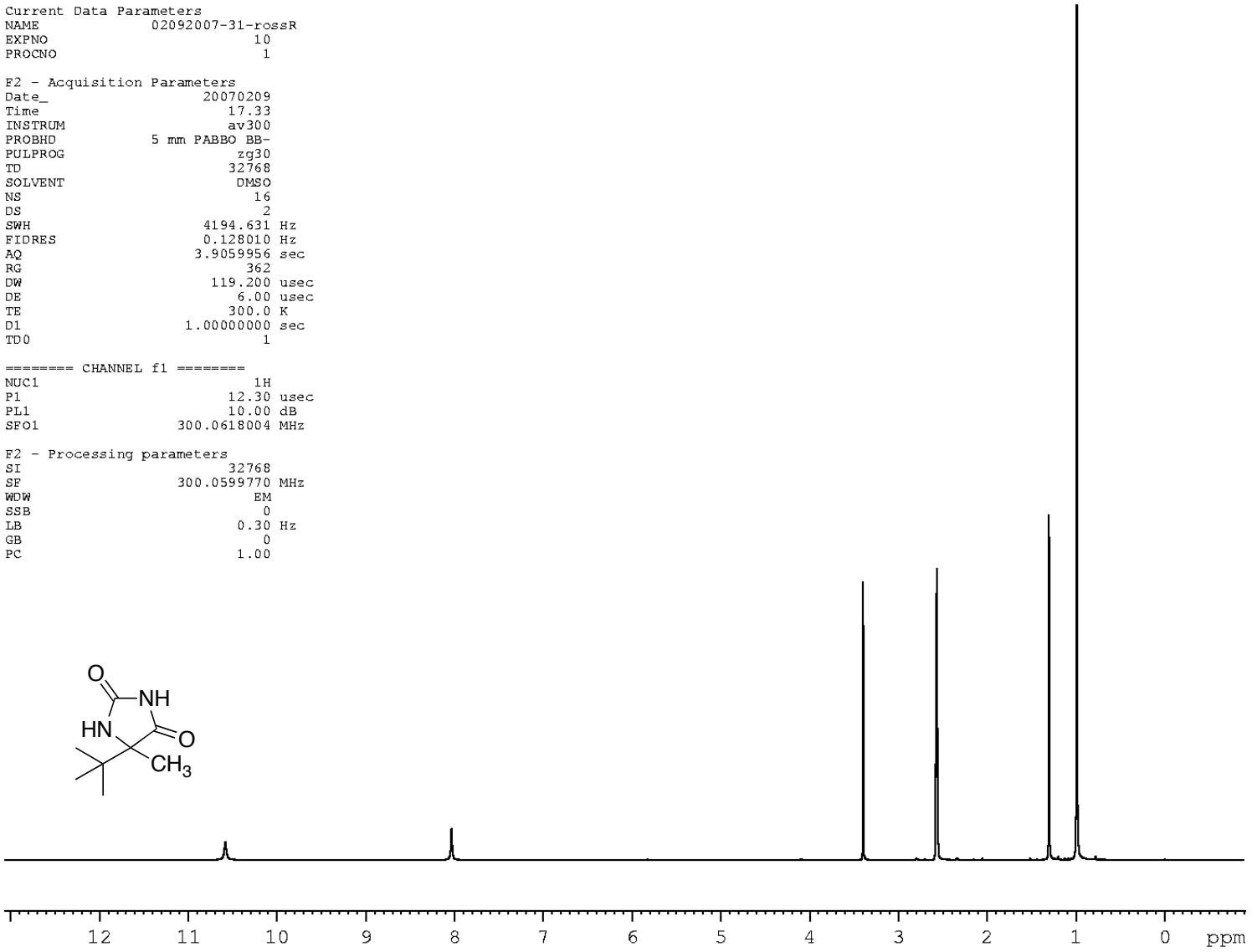
5-^{t}Butyl-5-methylimidazolidine-2,4-dione (13) ^{1}H NMR spectrum

```
Current Data Parameters
NAME             02092007-31-rossR
EXPNO                           10
PROCNO                           1

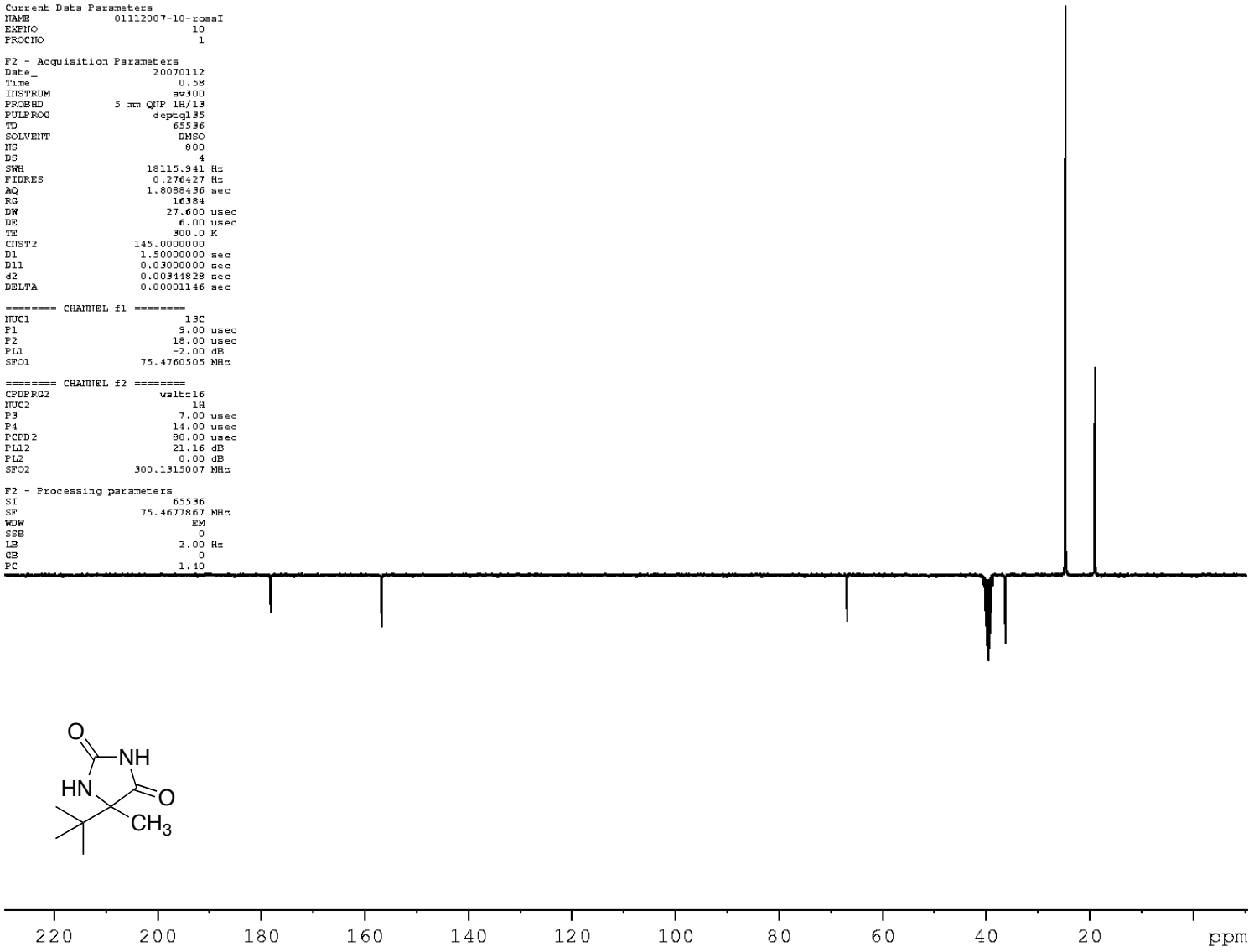
F2 - Acquisition Parameters
Date_                     20070209
Time                         17.33
INSTRUM                      av300
PROBHD             5 mm PABBO BB-
PULPROG                      zg30
TD                          32768
SOLVENT                      DMSO
NS                              16
DS                               2
SWH                       4194.631 Hz
FIDRES                    0.128010 Hz
AQ                       3.9059956 sec
RG                             362
DW                         119.200 usec
DE                           6.00 usec
TE                          300.0 K
D1                     1.00000000 sec
TD0                              1

======== CHANNEL f1 ========
NUC1                            1H
P1                           12.30 usec
PL1                          10.00 dB
SFO1                  300.0618004 MHz

F2 - Processing parameters
SI                          32768
SF                    300.0599770 MHz
WDW                            EM
SSB                             0
LB                           0.30 Hz
GB                              0
PC                           1.00
```



5-*t*Butyl-5-methylimidazolidine-2,4-dione (13) ^{13}C NMR spectrum



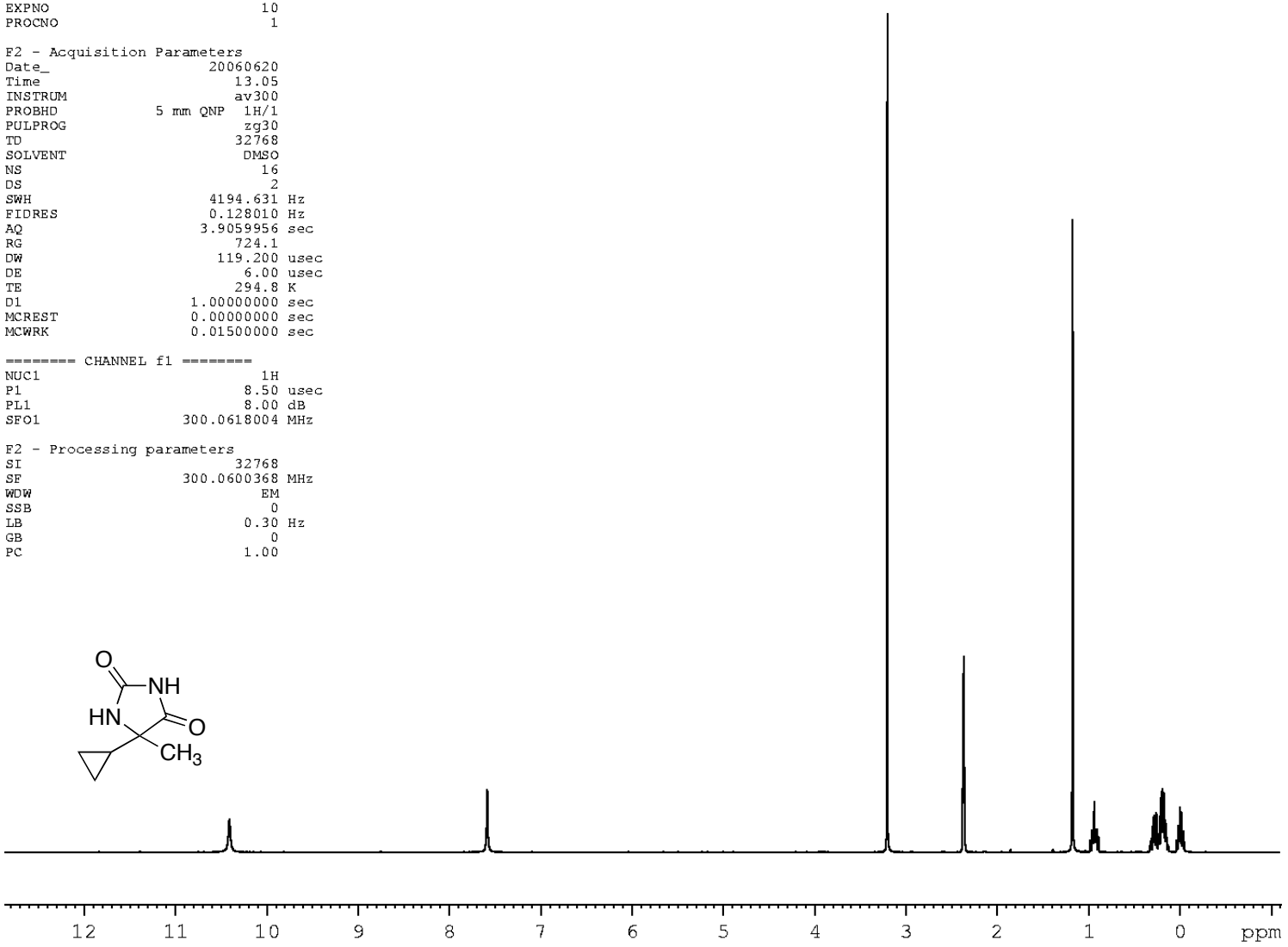
5-Cyclopropyl-5-methylimidazolidine-2,4-dione (4) ^{1}H NMR spectrum

```
Current Data Parameters
NAME             06202006-19-ross
EXPNO                          10
PROCNO                          1

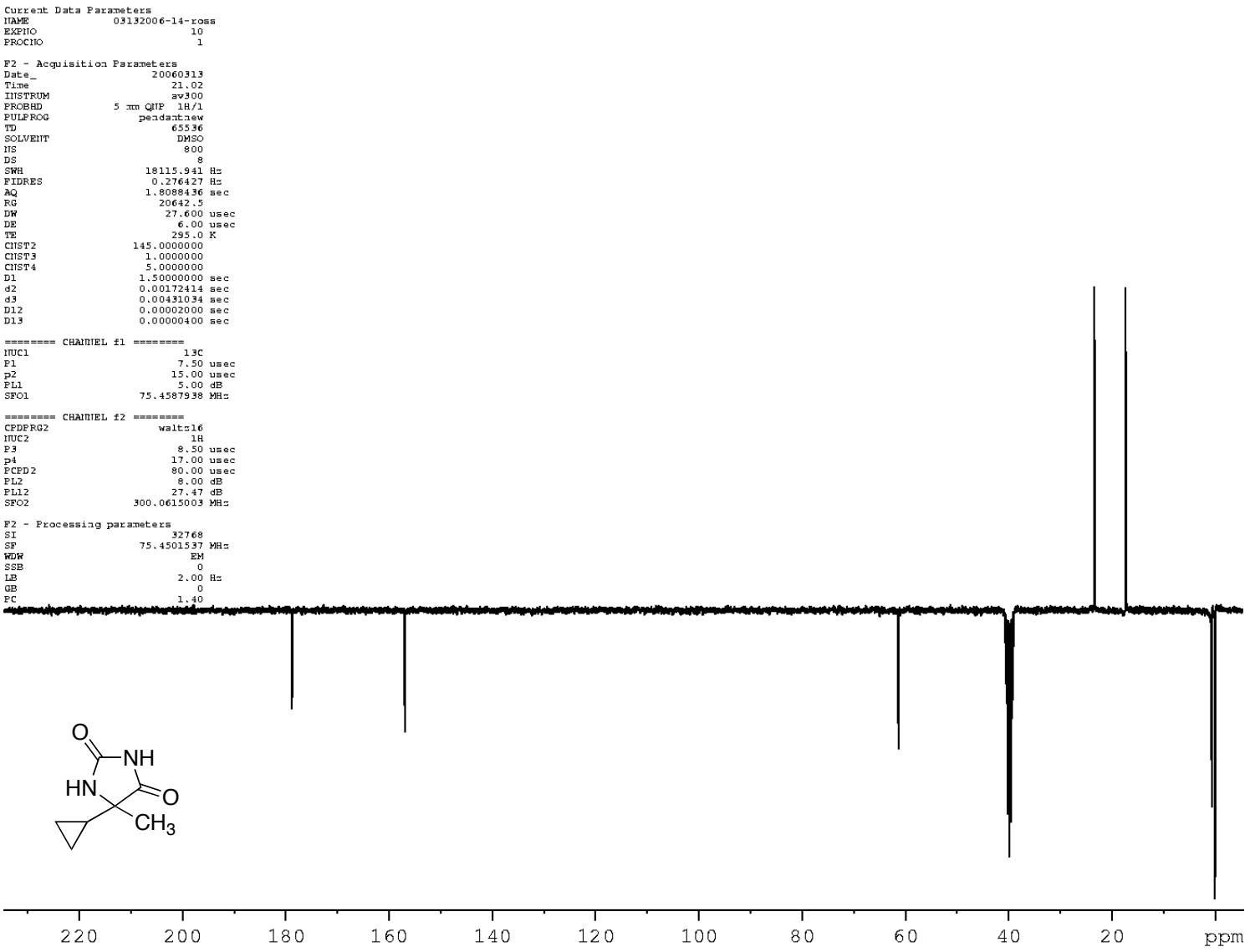
F2 - Acquisition Parameters
Date_                    20060620
Time                        13.05
INSTRUM                     av300
PROBHD           5 mm QNP  1H/1
PULPROG                      zg30
TD                          32768
SOLVENT                      DMSO
NS                             16
DS                              2
SWH                      4194.631 Hz
FIDRES                   0.128010 Hz
AQ                       3.9059956 sec
RG                           724.1
DW                         119.200 usec
DE                           6.00 usec
TE                          294.8 K
D1                    1.00000000 sec
MCREST                0.00000000 sec
MCWRK                 0.01500000 sec

======== CHANNEL f1 ========
NUC1                           1H
P1                           8.50 usec
PL1                          8.00 dB
SFO1                  300.0618004 MHz

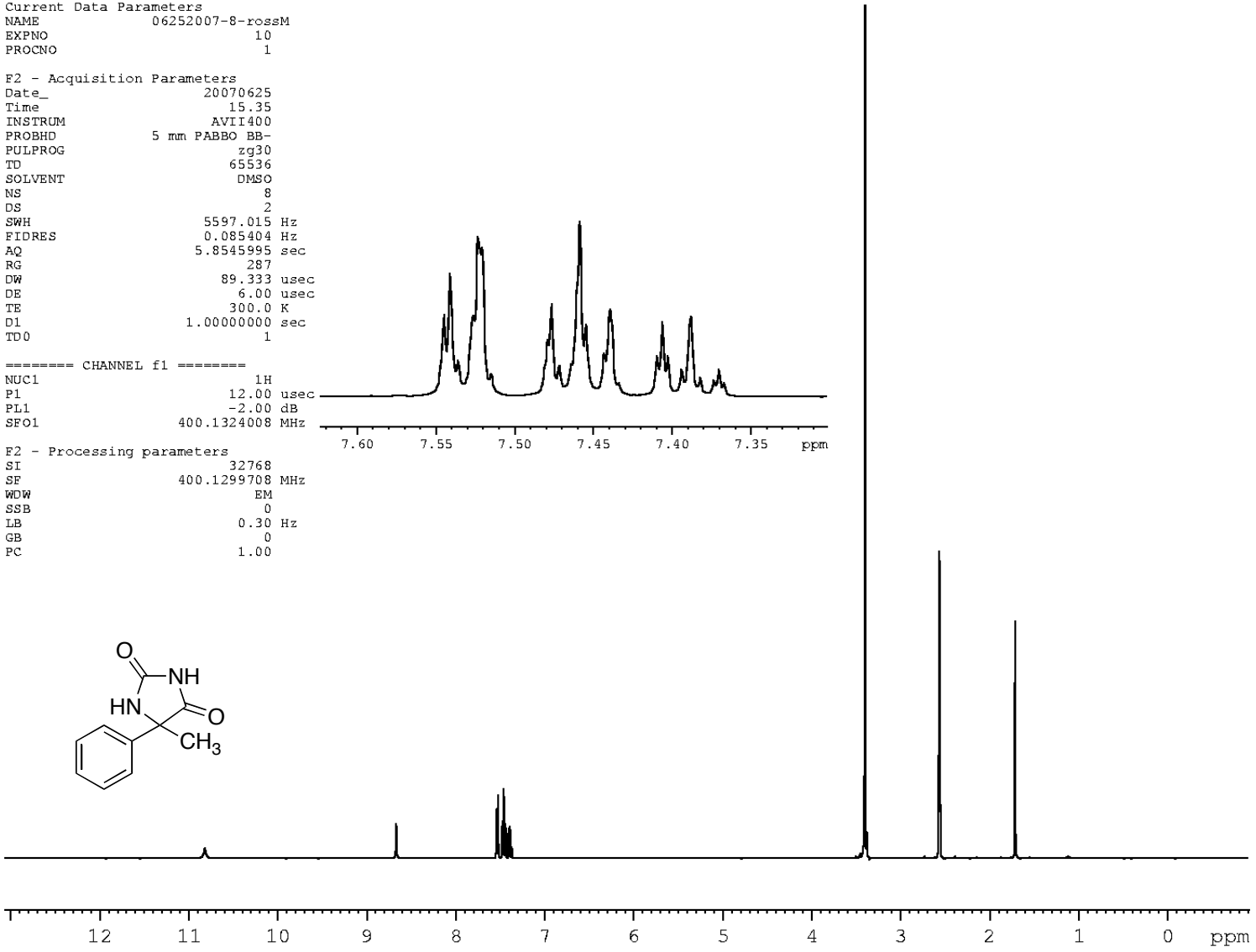
F2 - Processing parameters
SI                          32768
SF                    300.0600368 MHz
WDW                            EM
SSB                             0
LB                           0.30 Hz
GB                              0
PC                           1.00
```



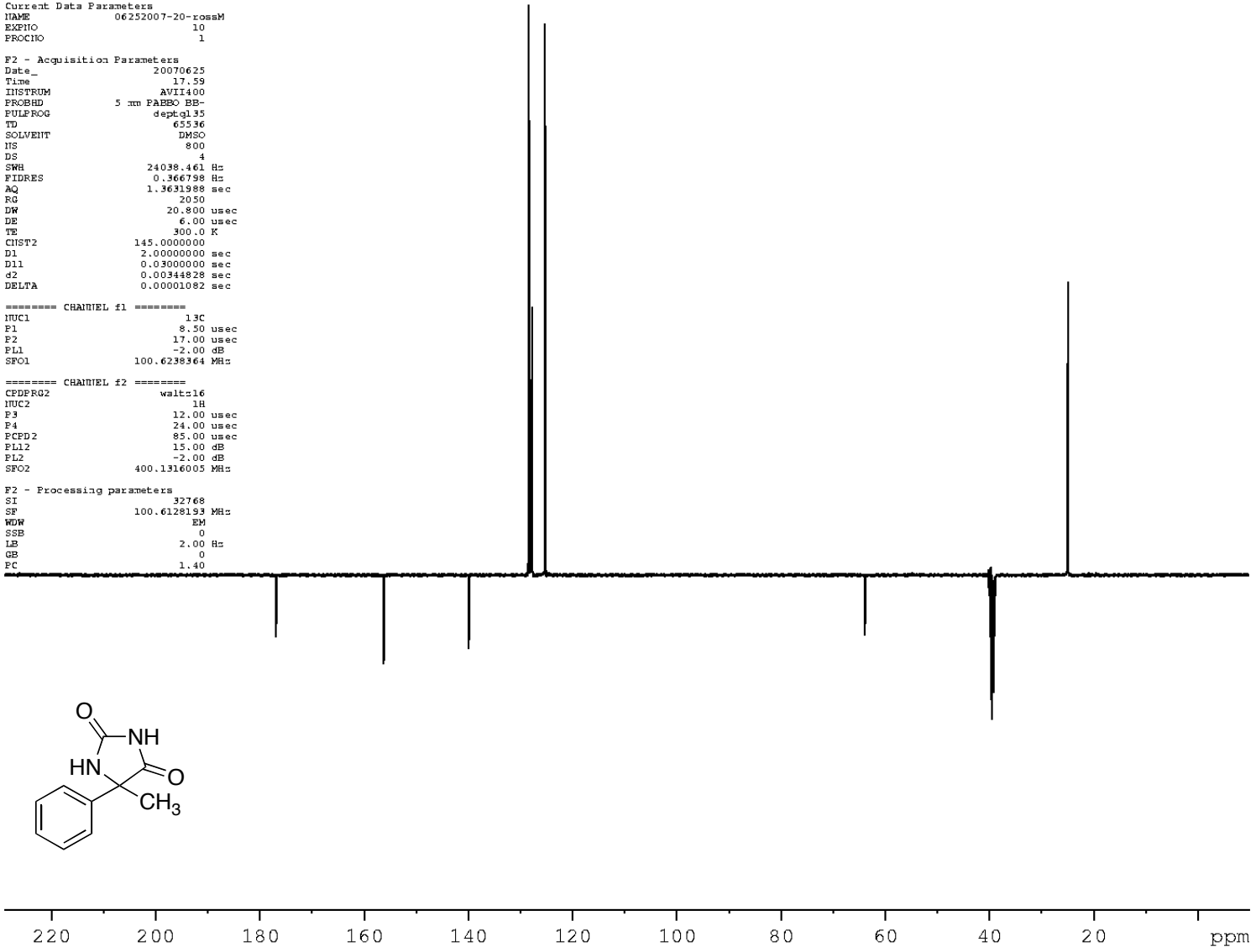
5-Cyclopropyl-5-methylimidazolidine-2,4-dione (4) ^{13}C NMR spectrum



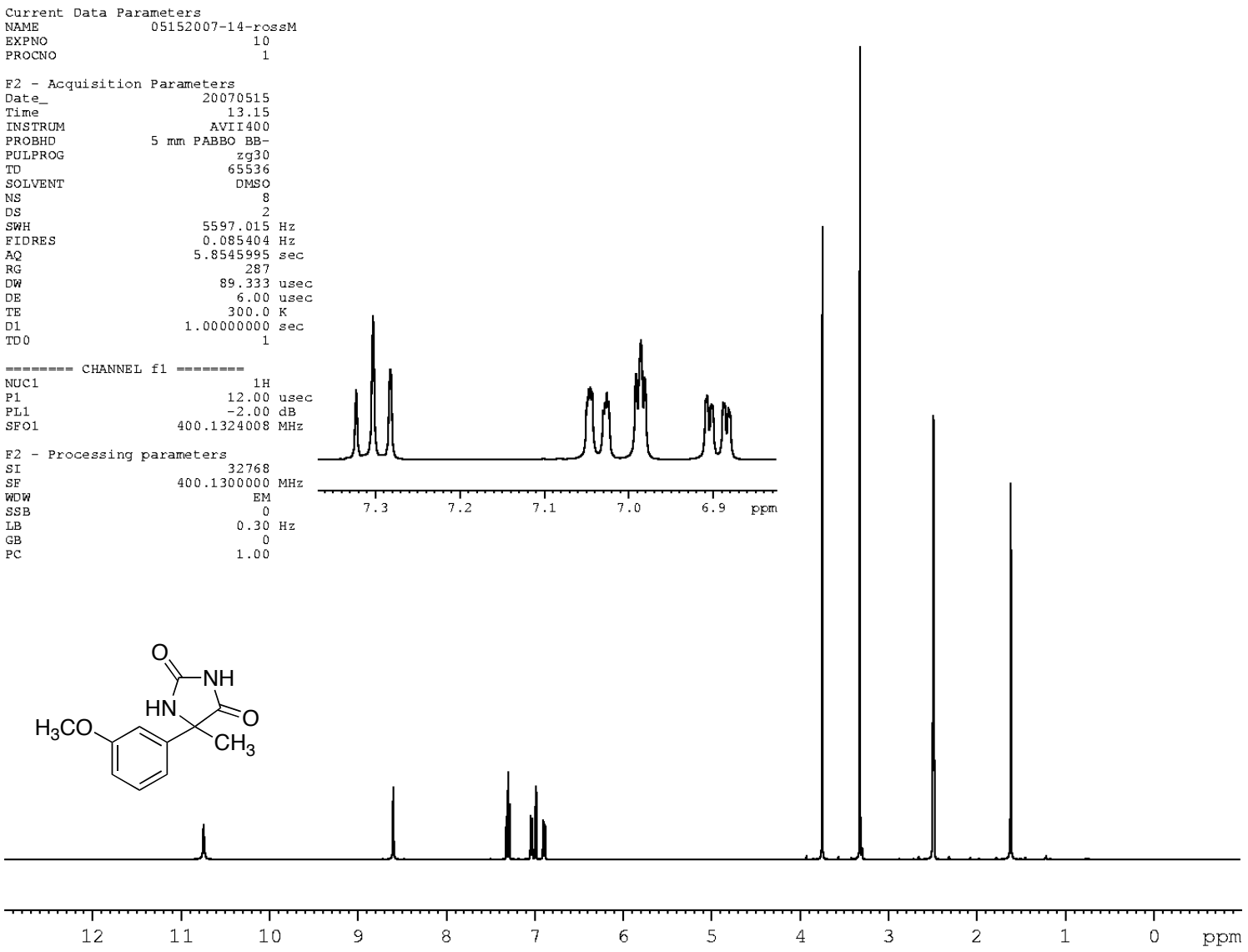
5-Methyl-5-phenylimidazolidine-2,4-dione (14) ^{1}H NMR spectrum



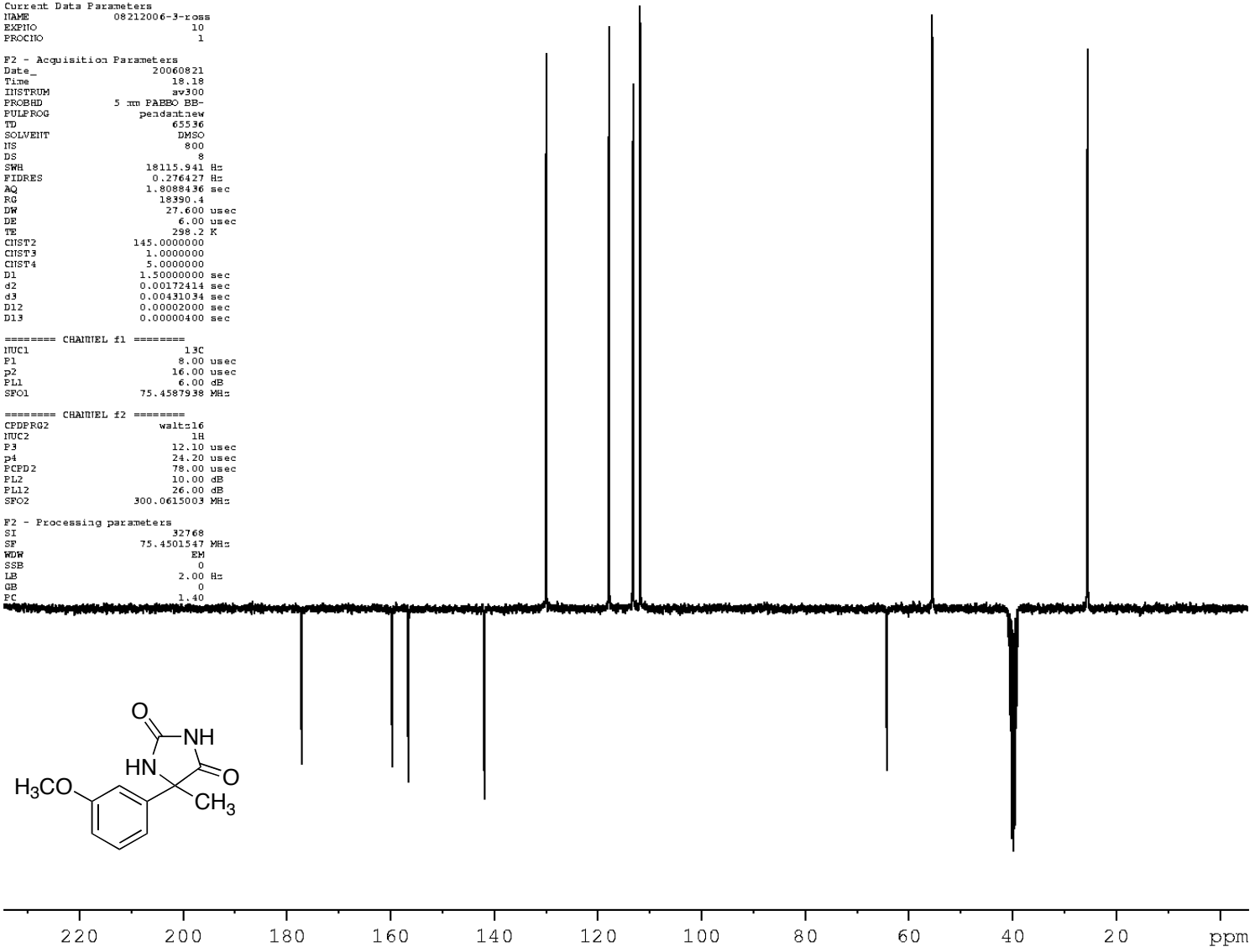
5-Methyl-5-phenylimidazolidine-2,4-dione (14) ^{13}C NMR spectrum



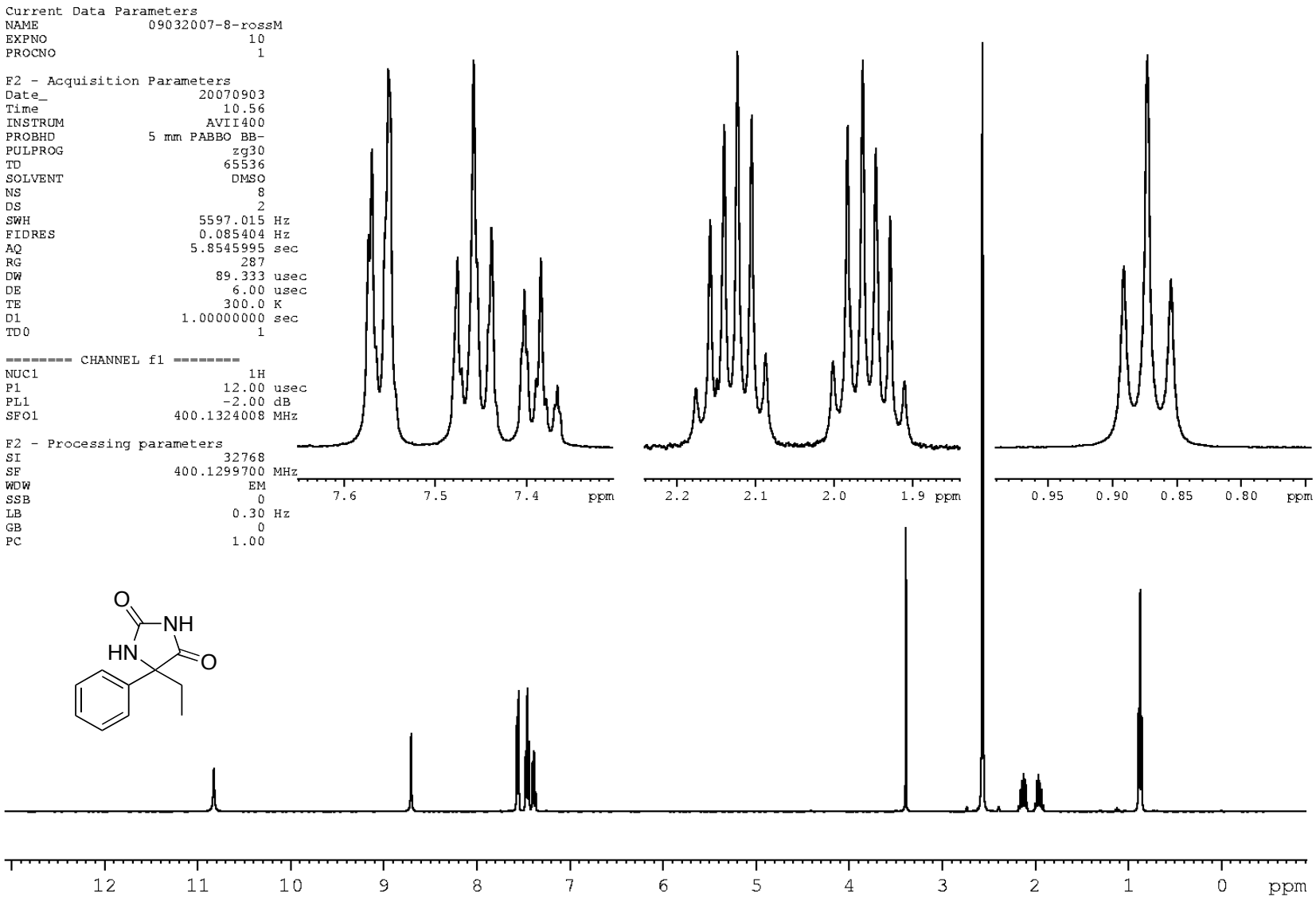
5-(3'Methoxyphenyl)-5-methylimidazolidine-2,4-dione (15) ^{1}H NMR spectrum



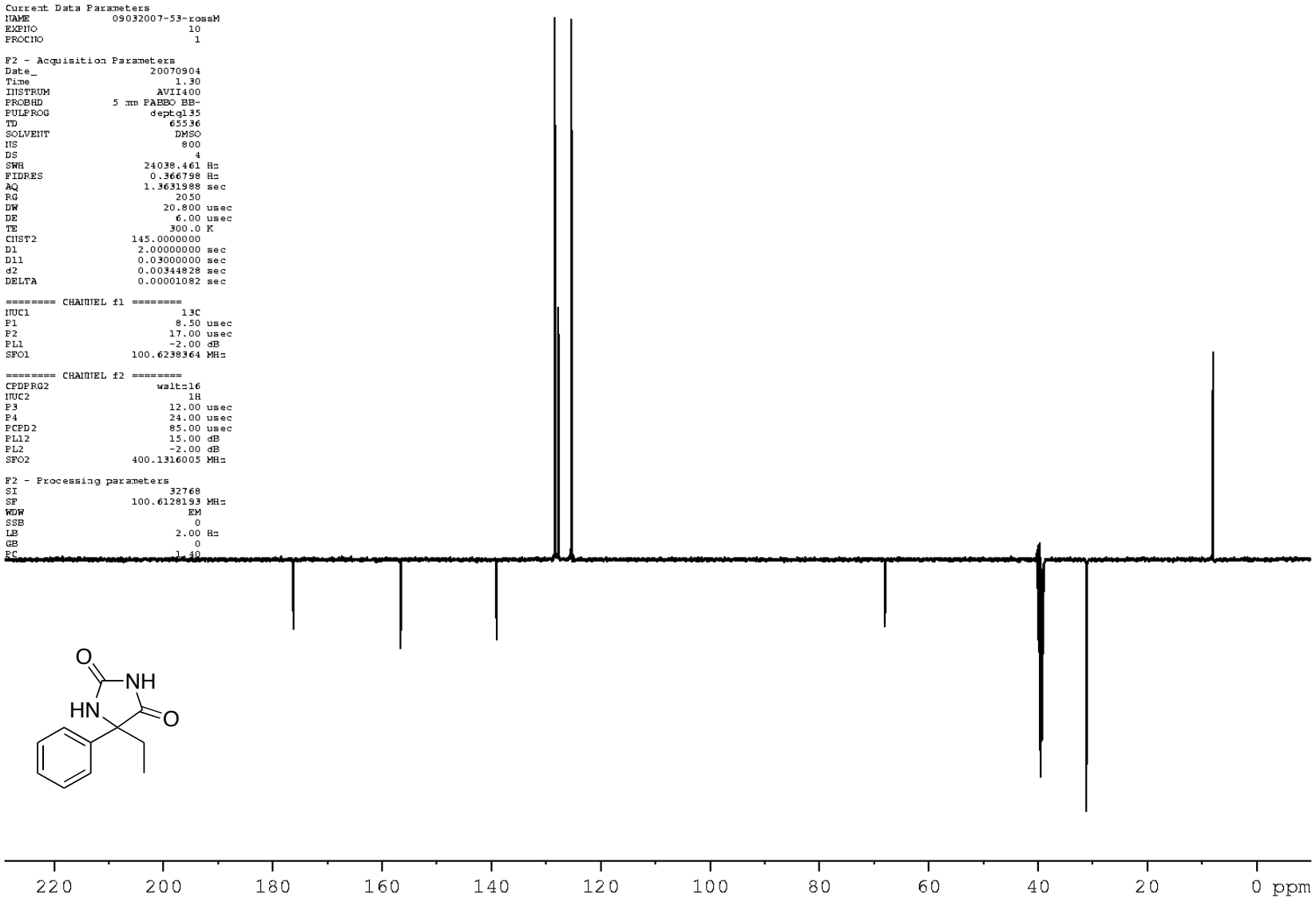
5-(3'Methoxyphenyl)-5-methylimidazolidine-2,4-dione (15) ^{13}C NMR spectrum



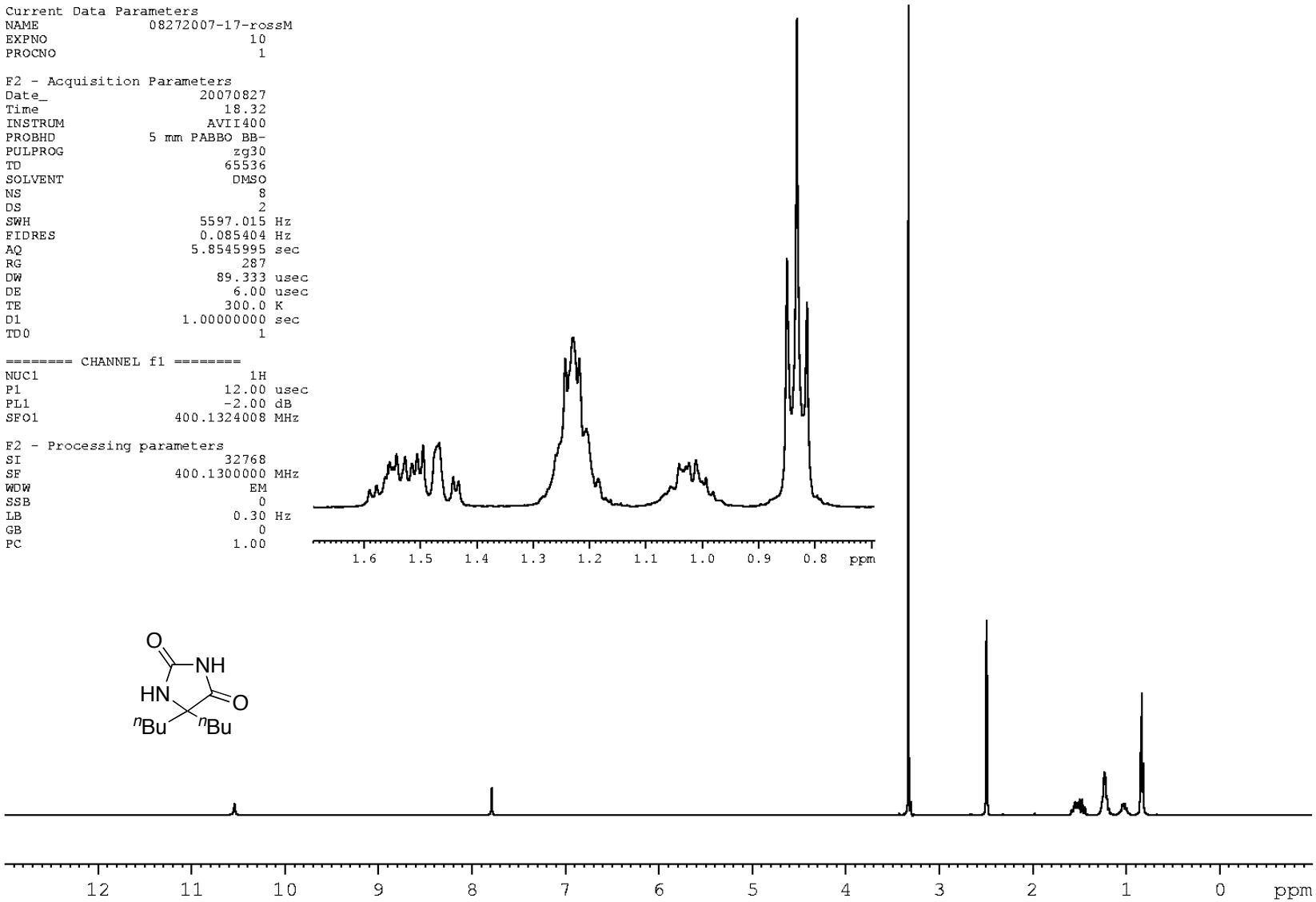
5-Ethyl-5-phenylimidazolidine-2,4-dione (16) ^{1}H NMR spectrum



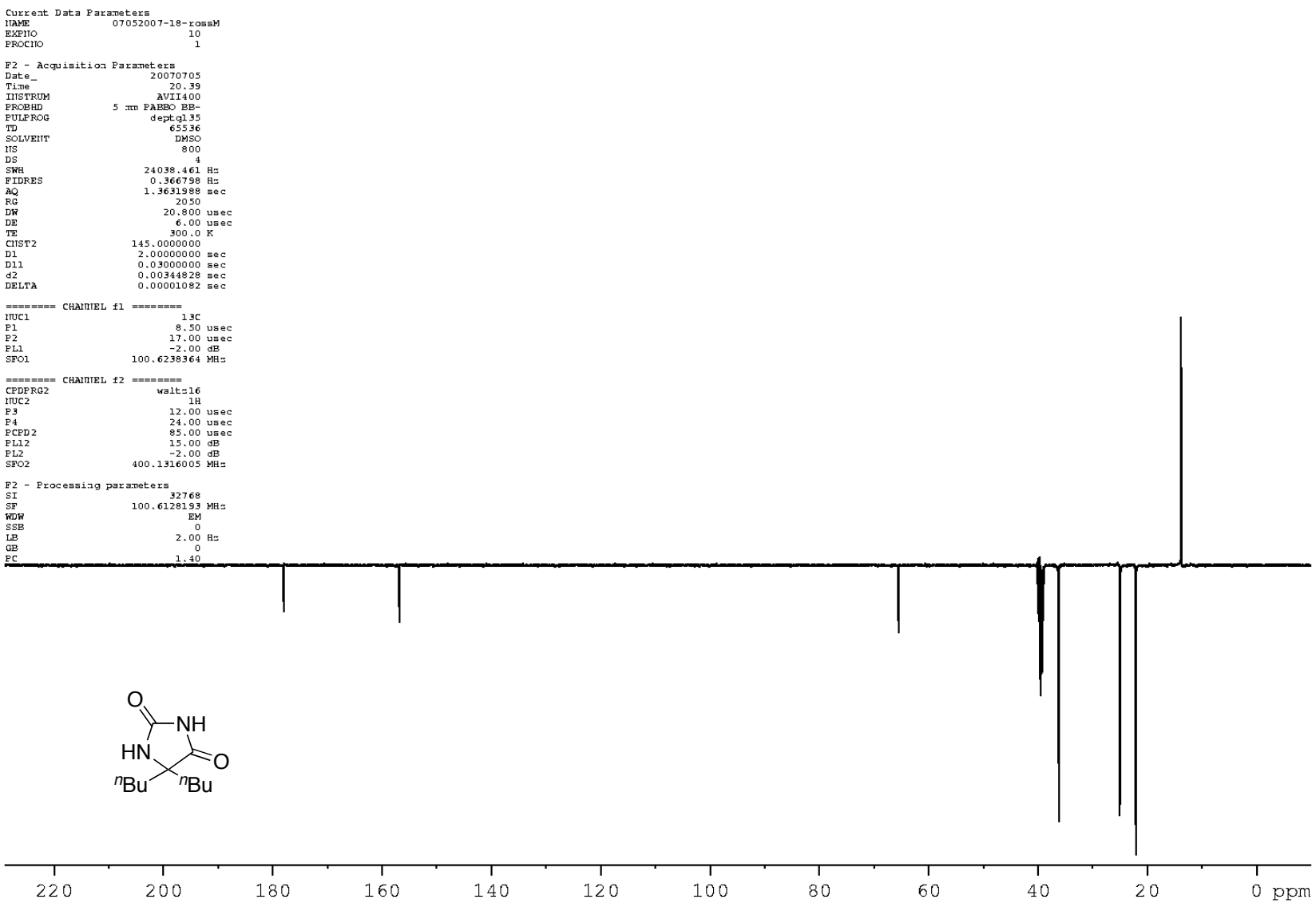
5-Ethyl-5-phenylimidazolidine-2,4-dione (16) ^{13}C NMR spectrum



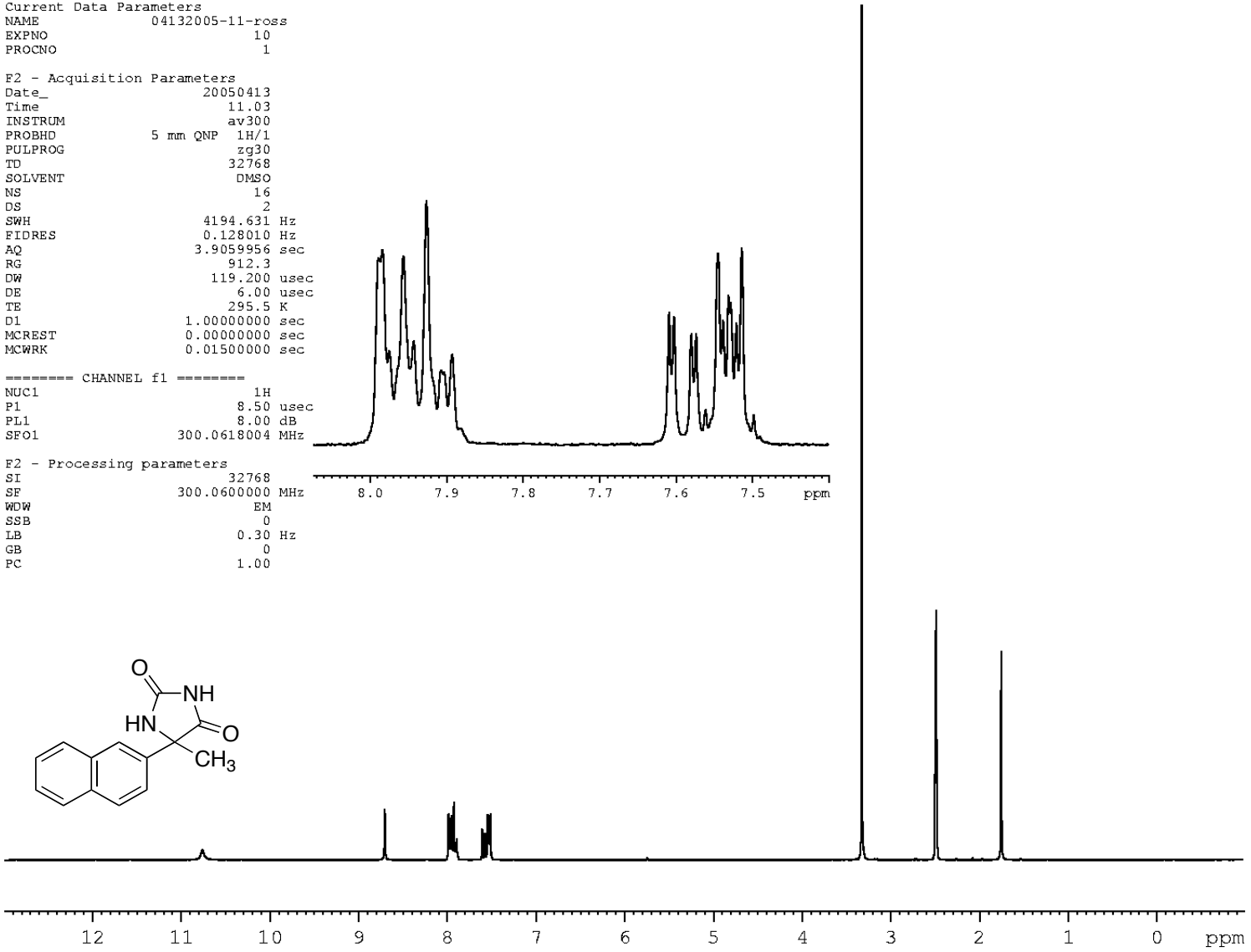
5,5-Di-(*n*butyl)imidazolidine-2,4-dione (17) ^{1}H NMR spectrum



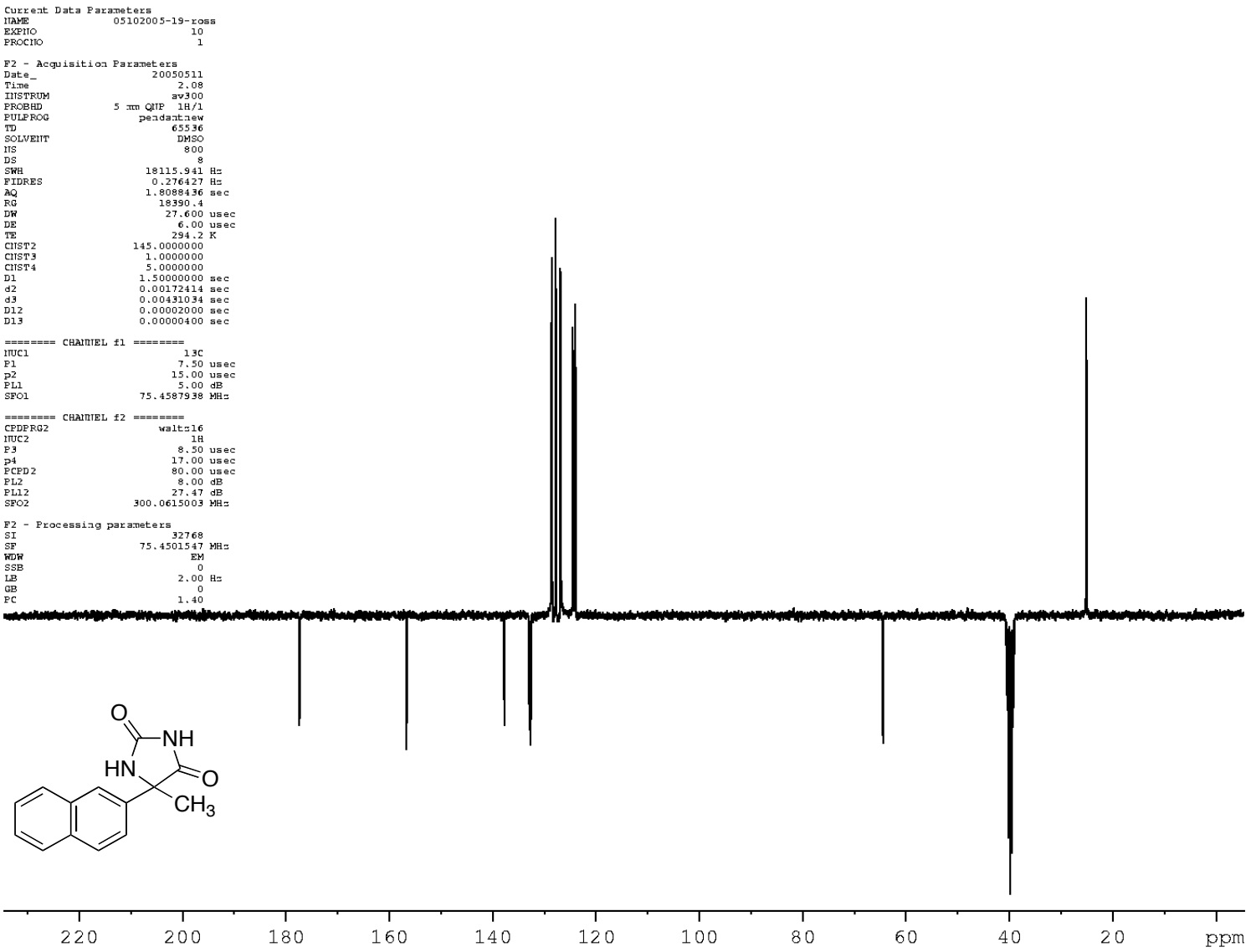
5,5-Di-(*n*butyl)imidazolidine-2,4-dione (17) ^{13}C NMR spectrum



5-Methyl-5-(naphthyl)imidazolidine-2,4-dione (19) ^{1}H NMR spectrum

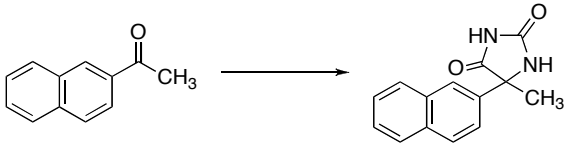


5-Methyl-5-(naphthyl)imidazolidine-2,4-dione (19) ^{13}C NMR spectrum



Optimisation of the one-pot reaction

The reaction was conducted by condensing liquid ammonia into a solution of the ketone and gallium (III) triflate in dichloromethane at -78 °C, followed by stirring either at room temperature or -78 °C for the time shown. A solution of hydrogen cyanide was preformed by stirring TMS cyanide in methanol for 2 h. This solution was then added to the reaction mixture, either with ammonia present (reactions at -78 °C) or after the ammonia had evaporated (room temperature reactions). The Hünig's base was then added and carbon dioxide bubbled through the reaction for the time shown.



Entry	Imine formation conditions	Amino nitrile formation conditions	Hydantoin formation conditions	Recovered ketone	Yield
1	$Ga(OTf)_3$, CH_2Cl_2, $NH_{3\ (l)}$, -78 °C, 3 h	HCN solution, RT 14 h	Hünig's base, CO_2 (g), 8 h, RT	60 %	18 %
2	$Ga(OTf)_3$, CH_2Cl_2, $NH_{3\ (l)}$, -78 °C, 24 h	HCN solution, RT 24 h	Hünig's base, CO_2 (g), 3 h, RT	39 %	27 %
3	$Ga(OTf)_3$, CH_2Cl_2, $NH_{3\ (l)}$, -78 °C then warmed to RT, 24 h	HCN solution, RT 24 h	Hünig's base, CO_2 (g), 8 h, RT	88 %	3 %
4	$Ga(OTf)_3$, CH_2Cl_2, $NH_{3\ (l)}$, -78 °C, 24 h	HCN solution, -78 °C, 30 min then RT 24 h	Hünig's base, CO_2 (g), 6 h, RT	38 %	53 %
5	$Ga(OTf)_3$, CH_2Cl_2, $NH_{3\ (l)}$, -78 °C, 3 h	HCN solution, -78 °C, 30 min then RT 24 h	Hünig's base, CO_2 (g), 6 h, RT	-	47-50 % (n = 2)
6	$Ga(OTf)_3$, CH_2Cl_2, $NH_{3\ (l)}$, -78 °C, 24 h	HCN solution, -78 °C, 24 h	Hünig's base, CO_2 (g), 6 h, RT	70 %	5 %
7	CH_2Cl_2, $NH_{3\ (l)}$, -78 °C, 24 h	HCN solution, -78 °C, 24 h	Hünig's base, CO_2 (g), 8 h, RT	94 %	0 %
8	CH_2Cl_2, $NH_{3\ (l)}$, -78 °C, TMSCN, MeOH, 10 h		Hünig's base, CO_2 (g), 8 h, RT	87 %	9 %