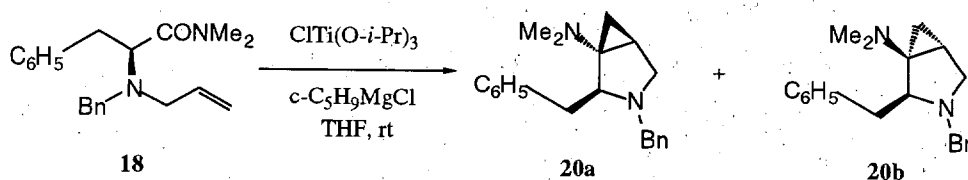


SUPPORTING INFORMATION

General

All solvents were reagent grade and were distilled before use. Tetrahydrofuran (THF) was distilled over sodium and benzophenone. Reagent cyclopentyl magnesium chloride was purchased from Aldrich. Proton magnetic resonance spectra (^1H NMR) and carbon magnetic resonance spectra (^{13}C NMR) were recorded on a Bruker AMX-500 spectrometer. Chemical shifts are in parts per million (ppm) relative to the solvent as the internal reference. Infrared spectra were obtained on a Perkin-Elmer Model 281-B spectrometer. Absorptions were reported in wavenumber (cm^{-1}). Optical rotations (in degrees) were measured with a Perkin-Elmer Model 241 polarimeter. Flash column chromatography was carried out on E. Merck silica gel 60 (240-400 mesh) using the solvent systems listed under individual experiments.

Representative Procedure for the Intramolecular Aminocyclopropanation



To a solution of 2-(allylbenzylamino)-*N,N*-dimethyl-3-phenylpropionamide (**18**, 50mg, 0.155 mmol) in 1.55 mL anhydrous THF was added $\text{ClTi}(\text{O-}i\text{-Pr})_3$ (0.155 mL of 1.0 M solution in hexanes, 1.0 equiv), cyclopentyl magnesium chloride (0.349 mL of 2.0 M solution in diethyl ether, 4.5 equiv) was added dropwise at room temperature over a period of 1 h. The reaction mixture was stirred for an additional 2 h, poured into ice water (5 mL), and stirred for 30 min. THF was evaporated under reduced pressure and 10 mL Et_2O was added to the residue. The organic layer was separated, and the aqueous layer was extracted with ether (3x10 mL). The combined extracts were washed with aqueous NaHCO_3 (10 mL), followed by brine (10 mL), and dried over Na_2SO_4 . Evaporation of the solvent, followed by silica gel column chromatography (acetone-hexanes, 10:90) of the residue afforded 28 mg (60% yield) of **20a** as a light yellow solid, and 11mg (23%) of **20b** as a light yellow oil.

(1*R*,2*S*,6*S*)-2,3-Dibenzyl-3-aza-bicyclo[3.1.0]hex-1-yl-dimethyl amine (**20a**)

- Mp: 32.8-33.8 °C; R_f 0.2 (ethyl acetate:petroleum ether, 20:80); $[\alpha]_D^{20} +18.4$ ($c=1.0$, CHCl_3); ^1H NMR (500MHz, CDCl_3) δ 7.14-7.32 (m, 10H), 3.81 (d, 1H, $J = 13.3$ Hz), 3.18 (m, 1H), 3.14-3.19 (dd, 1H, $J = 4.2$ Hz, 13.7 Hz), 3.02 (d, 1H, $J = 13.3$ Hz), 2.86 (dd, 1H, $J = 13.7$ Hz, 4.8 Hz), 2.72 (d, 1H, $J = 9.0$ Hz), 2.38 (s, 6H), 2.30 (dd, 1H, $J = 3.9$ Hz, 9.0Hz), 1.24 (m, 1H), 0.89 (dd, 1H, $J = 4.5$ Hz), 0.59 (dd, 1H, $J = 4.5$ Hz, 8.7Hz); ^{13}C NMR (500MHz, CDCl_3) δ 140.4, 139.8, 129.6, 128.4, 128.0, 128.0, 126.6, 125.8, 62.9, 58.3, 55.4, 54.6, 42.2, 38.7, 21.7, 14.5; IR (neat): 3026, 2926.7, 2817.4, 2776.9, 1602.1, 1494.0, 1452.2, 1376.8, 1351.6, 737.2, 697.1 cm^{-1} ; HRMS calcd for $\text{MH}^+ \text{C}_{21}\text{H}_{27}\text{N}_2$, 307.2174; found, 307.2160.

(1S,2S,6R)-2,3-Dibenzyl-3-aza-bicyclo[3.1.0]hex-1-yl-dimethyl amine (**20b**)

- R_f 0.2 (ethyl acetate:petroleum ether, 5:95); $[\alpha]_D^{20}$ -48.0 (c=1.0, CHCl₃); ¹H NMR (500MHz, CDCl₃) δ 7.07-7.23 (m, 10H), 3.68 (d, 1H, J = 13.8 Hz), 3.55 (d, 1H, J = 13.8 Hz), 3.42 (dd, 1H, J = 4.7 Hz, 6.4 Hz), 3.10 (dd, 1H, J = 13.8 Hz, 4.5 Hz), 2.79 (dd, 1H, J = 3.2 Hz, 9.3 Hz), 2.73 (dd, 1H, J = 6.4 Hz, 13.8Hz), 2.51 (d, 1H, J = 9.3 Hz), 2.12 (s, 6H), 1.55-1.58 (m, 1H), 0.72 (dd, 1H, J = 4.2 Hz, 8.5 Hz), 0.63 (dd, 1H, J = 4.0 Hz); ¹³C NMR (500MHz, CDCl₃) δ 142.5, 140.5, 129.6, 128.2, 127.9, 127.8, 126.4, 125.0, 68.0, 57.0, 55.8, 51.9, 42.1, 34.5, 22.8, 8.45; IR (neat): 3025.2, 2936.1, 2817.4, 2774.8; 1602.1, 1493.4, 1452.7, 1348.6, 730.7, 697.3 cm⁻¹; HRMS calcd for MH⁺ C₂₁H₂₇N₂, 307.2174; found, 307.2160.

(1R,2S,6S)-3-Benzyl-2-(4-hydroxybenzyl)-3-aza-bicyclo[3.1.0]hex-1-yl-dimethyl amine (**22a**)

- Mp: 143-144 °C; R_f 0.35 (acetone:hexanes, 30:70); $[\alpha]_D^{20}$ +14.4 (c=1.0, CHCl₃); ¹H NMR (500MHz, CDCl₃) δ 7.15-7.29 (m, 7H), 6.70 (d, 2H, J = 8.1 Hz), 5.88 (br, s, 1H), 3.84 (d, 1H, J = 13.3 Hz), 3.16 (m, 1H), 3.02-3.10 (m, 2H), 2.81 (d, 1H, J = 10.5 Hz), 2.72 (d, 1H, J = 8.4 Hz), 2.38 (s, 6H), 2.31 (m, 1H), 1.28 (m, 1H), 0.89 (m, 1H), 0.62 (d, 1H, J = 3.0 Hz); ¹³C NMR (500MHz, CDCl₃) δ 154.0, 139.5, 132.1, 130.6, 128.5, 128.1, 126.6, 115.2, 62.8, 58.3, 55.4, 54.5, 42.3, 37.7, 21.9, 14.5; IR (neat): 3300 (br), 2930.5, 2785.1, 1612.6; 1514.3, 1453.5, 1240.1, 1170.0, 827.1, 751.7, 699.2 cm⁻¹; HRMS calcd for MH⁺ C₂₁H₂₇N₂O, 323.2123; found, 323.2111.

(1S,2S,6R)-3-Benzyl-2-(4-hydroxybenzyl)-3-aza-bicyclo[3.1.0]hex-1-yl-dimethyl amine (**22b**)

- R_f 0.55 (acetone:hexanes, 30:70); $[\alpha]_D^{20}$ -34.2 (c=1.0, CHCl₃); ¹H NMR (500MHz, CDCl₃) δ 7.08-7.24 (m, 7H), 6.67 (d, 2H, J = 8.4 Hz), 4.91 (br, s, 1H), 3.68 (d, 1H, J = 13.8 Hz), 3.56 (d, 1H, J = 13.8 Hz), 3.35 (m, 1H), 3.01 (dd, 1H, J = 13.7 Hz, 3.6 Hz), 2.79 (dd, 1H, J = 6.9 Hz, 8.7 Hz), 2.68 (dd, 1H, J = 6.1 Hz, 13.6 Hz), 2.49 (d, 1H, J = 9.4 Hz), 2.13 (s, 6H), 1.56 (m, 1H), 0.74 (dd, 1H, J = 3.6 Hz, 7.7Hz), 0.59 (m, 1H); ¹³C NMR (500MHz, CDCl₃) δ 153.2, 140.5, 134.4, 130.7, 128.2, 128.0, 126.4, 114.7, 68.2, 57.3, 55.8, 52.0, 42.0, 33.8, 22.9, 8.73; IR (neat): 3318.8 (br), 2934.8, 2817.8, 2776.1, 1612.3, 1513.5, 1453.3, 1352.2, 1243.2, 1170.4, 828.5, 740.0, 698.4 cm⁻¹; HRMS calcd for MH⁺ C₂₁H₂₇N₂O, 323.2123; found, 323.2118.

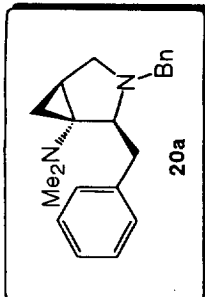
(1R,2S,6S)-3-Benzyl-2-(1H-indol-3-ylmethyl)-3-aza-bicyclo[3.1.0]hex-1-yl-dimethyl amine (**27a**)

- R_f 0.35 (acetone:hexanes, 30:70); $[\alpha]_D^{20}$ -9.8 (c=1.0, CHCl₃); ¹H NMR (500MHz, CDCl₃) δ 7.97 (br, s, 1H), 7.72 (d, 1H, J = 7.7 Hz), 7.30 (d, 1H, J = 7.8 Hz), 7.10-7.23 (m, 7H), 7.05 (d, 1H, J = 1.9 Hz), 3.80 (d, 1H, J = 13.3 Hz), 3.36 (dd, 1H, J = 4.1 Hz, 6.1 Hz), 3.30 (dd, 1H, J = 4.0 Hz, 15.3 Hz), 2.98-3.03 (m, 2H), 2.77 (d, 1H, J = 9.0 Hz), 2.42 (s, 6H), 2.33 (dd, 1H, J = 3.9 Hz, 9.0Hz), 1.30 (m, 1H), 1.09 (dd, 1H, J = 4.3 Hz), 0.65 (dd, 1H, J = 4.4 Hz, 8.6Hz); ¹³C NMR (500MHz, CDCl₃) δ 140.0, 136.3, 128.4, 127.9, 127.9, 126.5, 122.1, 121.8, 119.3, 119.1, 114.8, 111.0, 61.2, 58.8, 56.0, 54.9, 42.2, 28.9, 22.4, 14.5; IR (neat): 3420.3, 3058.2, 2922.9, 2783.6, 1619.7, 1493.8, 1477.5, 1453.8, 1353.2, 738.7, 699.2 cm⁻¹; HRMS calcd for M⁺ C₂₃H₂₇N₃, 345.2205; found, 345.2203.

(1S,2S,6R)-3-Benzyl-2-(1H-indol-3-ylmethyl)-3-aza-bicyclo[3.1.0]hex-1-yl-dimethyl amine (**27b**)

- R_f 0.55 (acetone:hexanes, 30:70); $[\alpha]_D^{20}$ -30.3 ($c=1.0$, CHCl_3); ^1H NMR (500MHz, CDCl_3) δ 7.81 (br, s, 1H), 7.60 (d, 1H, $J = 7.8$ Hz), 7.28 (d, 1H, $J = 8.0$ Hz), 7.06-7.21 (m, 7H), 6.92 (d, 1H, $J = 1.7$ Hz), 3.73 (d, 1H, $J = 14.0$ Hz), 3.56 (d, 1H, $J = 14.0$ Hz), 3.51 (dd, 1H, $J = 4.0$ Hz, 6.9 Hz), 3.29 (dd, 1H, $J = 3.9$ Hz, 14.8 Hz), 2.91 (dd, 1H, $J = 3.2$ Hz, 9.3 Hz), 2.84 (dd, 1H, $J = 7.0$ Hz, 14.8 Hz), 2.50 (d, 1H, $J = 9.3$ Hz), 2.21 (s, 6H), 1.61 (m, 1H), 0.77 (dd, 1H, $J = 4.2$ Hz, 8.5 Hz), 0.62 (dd, 1H, $J = 4.0$ Hz); ^{13}C NMR (500MHz, CDCl_3) δ 140.9, 136.1, 128.1, 128.0, 127.9, 126.3, 122.5, 121.4, 119.2, 118.8, 115.9, 110.9, 67.2, 57.2, 55.9, 52.2, 42.4, 23.8, 23.0, 9.34; IR (neat): 3421.1, 3057.1, 2935.8, 2817.0, 2774.6, 1617.9, 1492.5, 1454.1, 1351.1, 738.1, 698.0 cm^{-1} ; HRMS calcd for MH^+ $\text{C}_{23}\text{H}_{28}\text{N}_3$, 346.2283; found, 346.2273.

HS



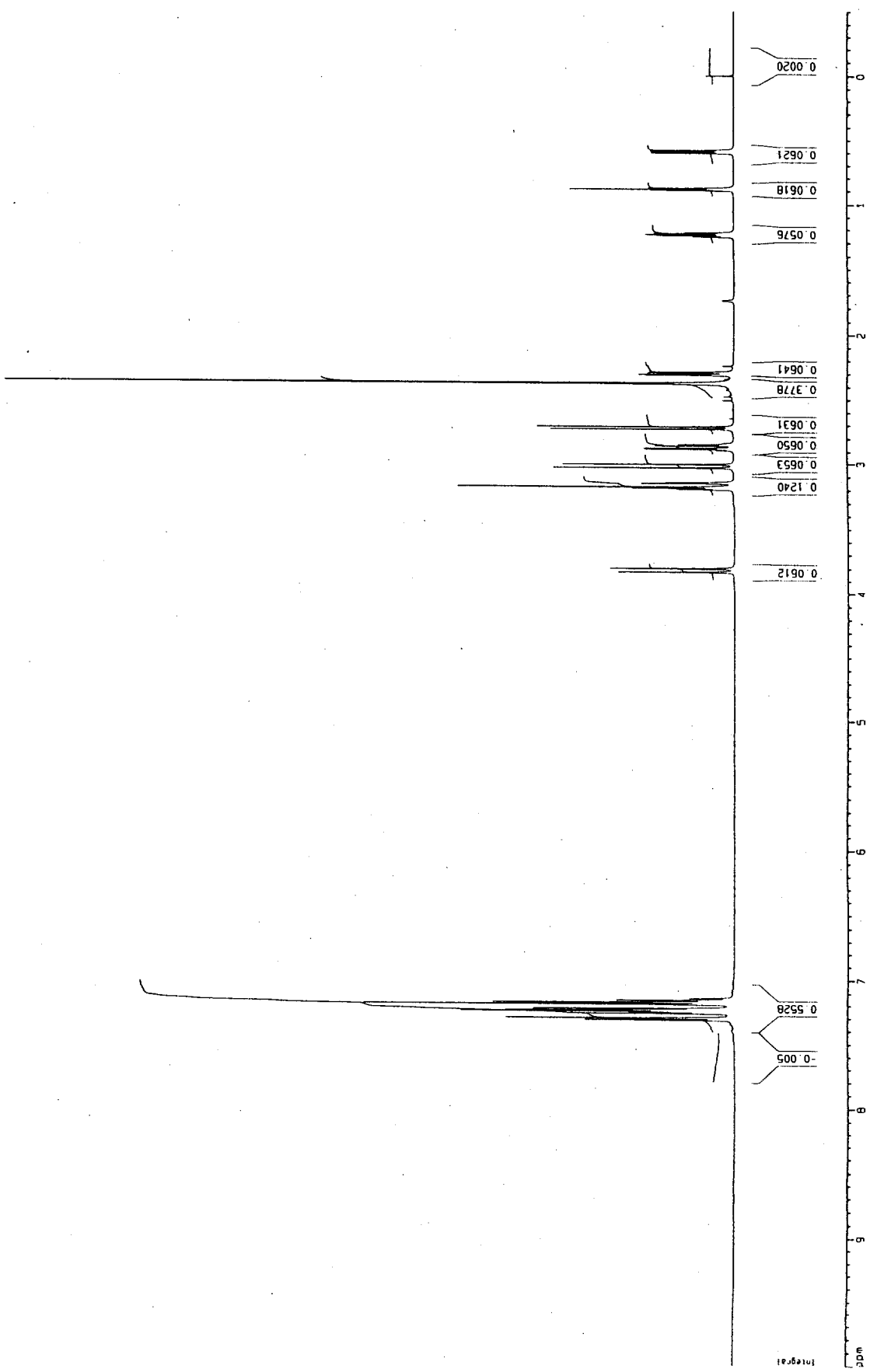
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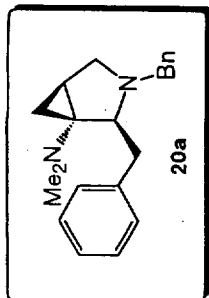
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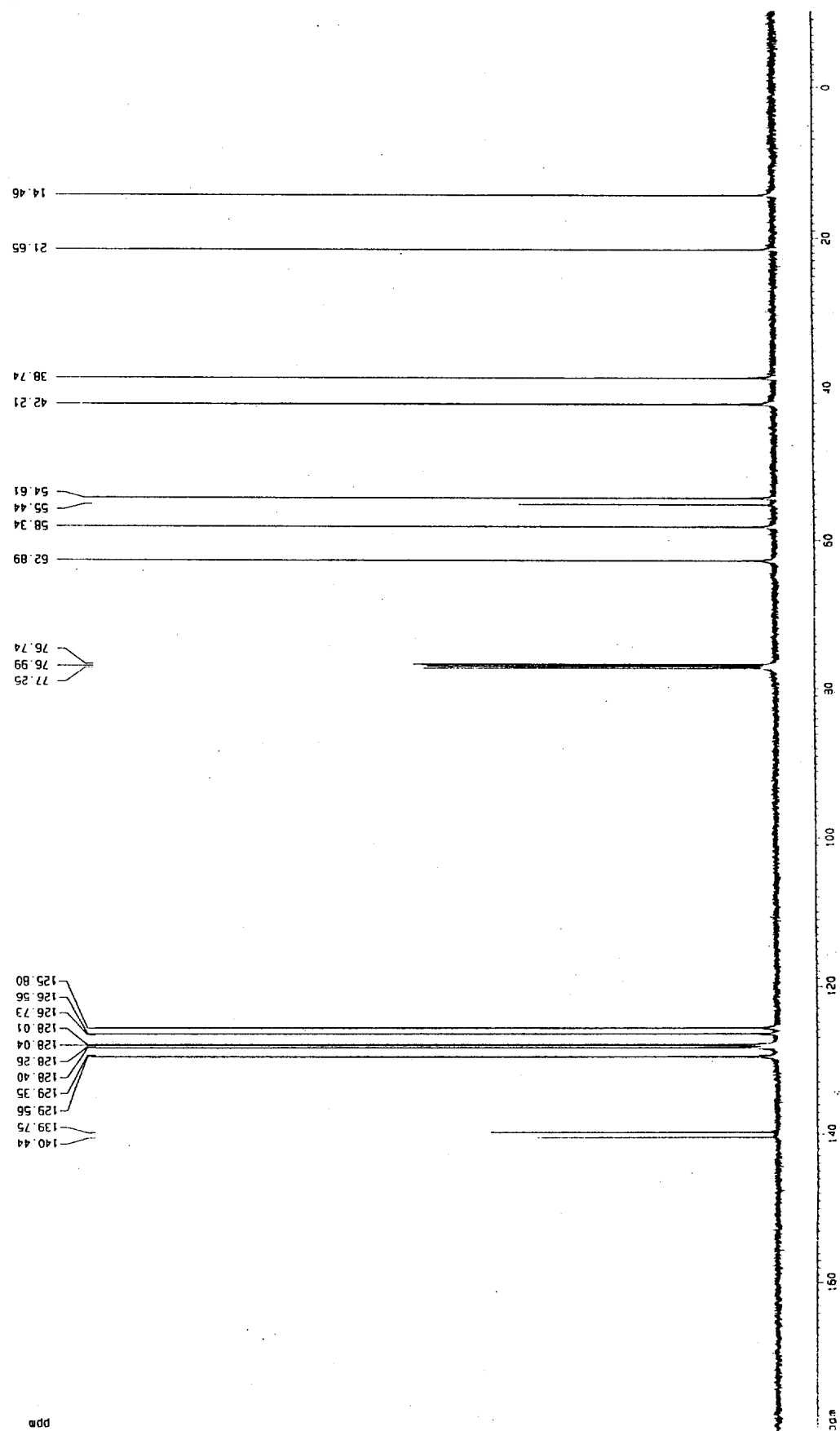
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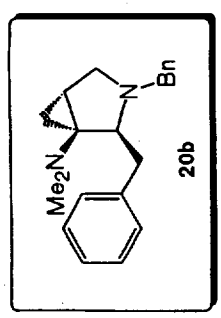


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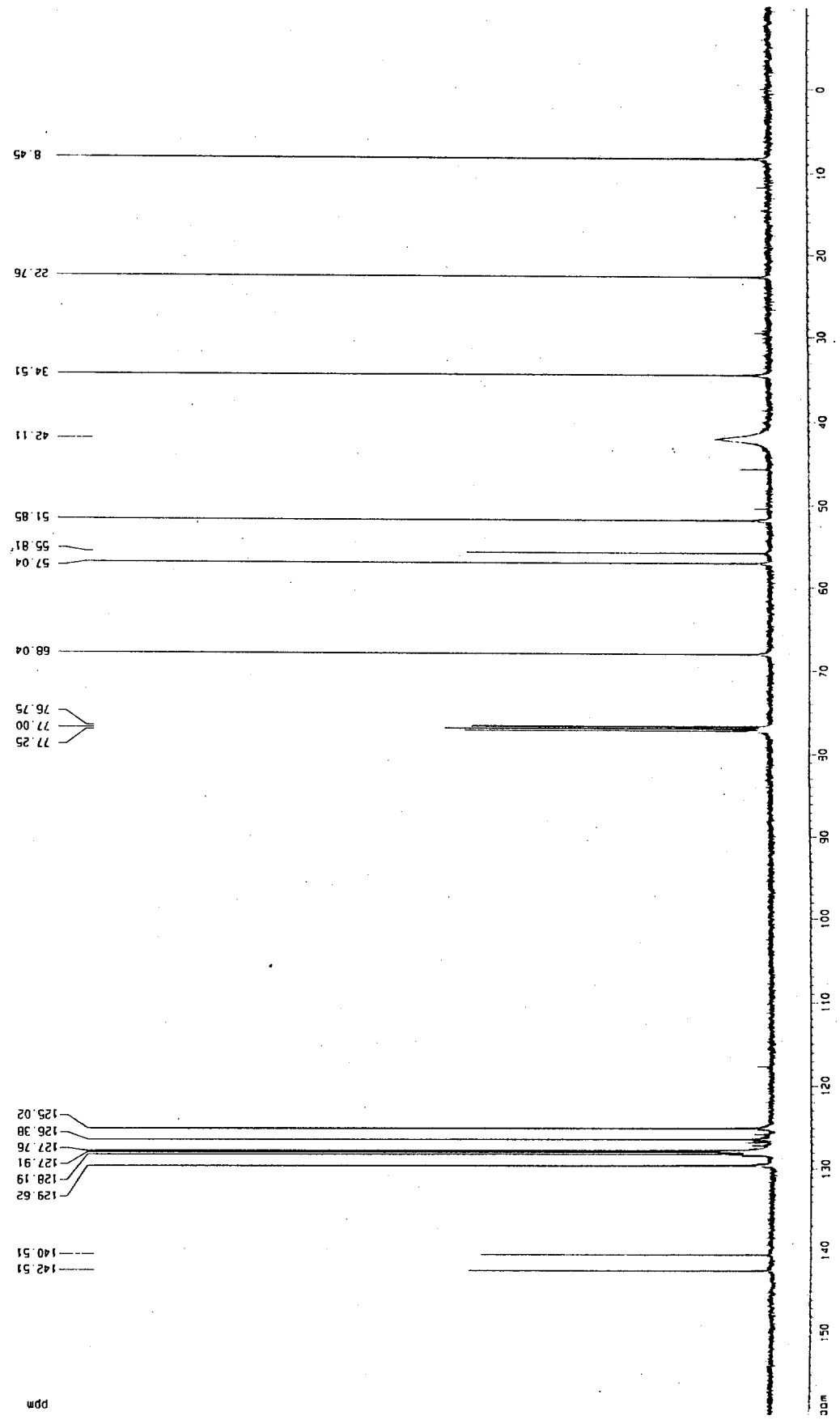
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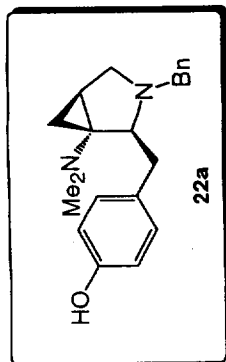


¹³C-NMR



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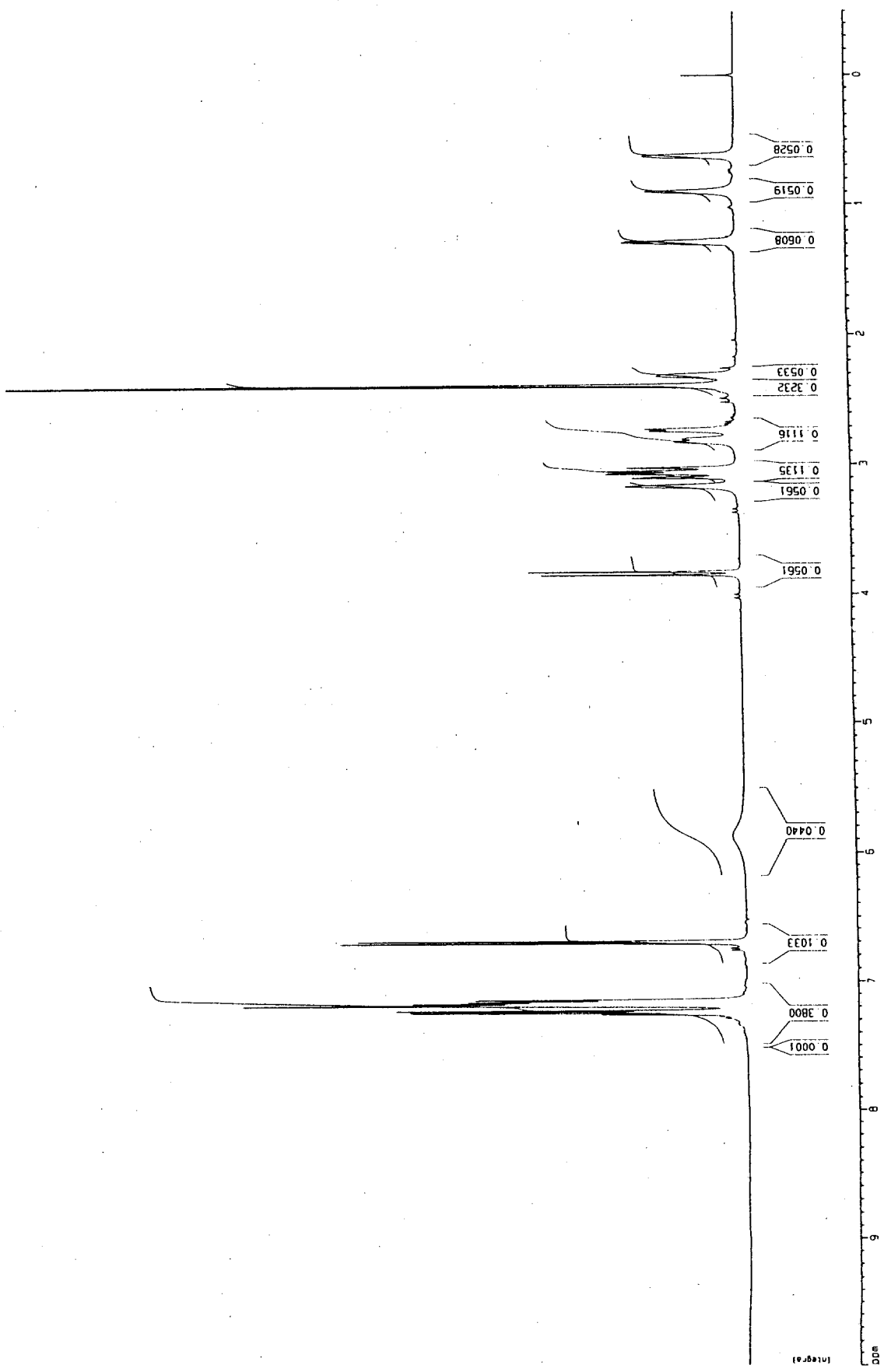


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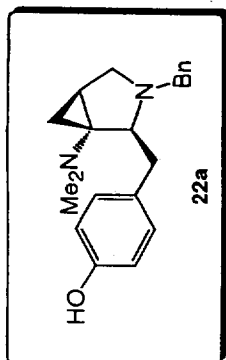
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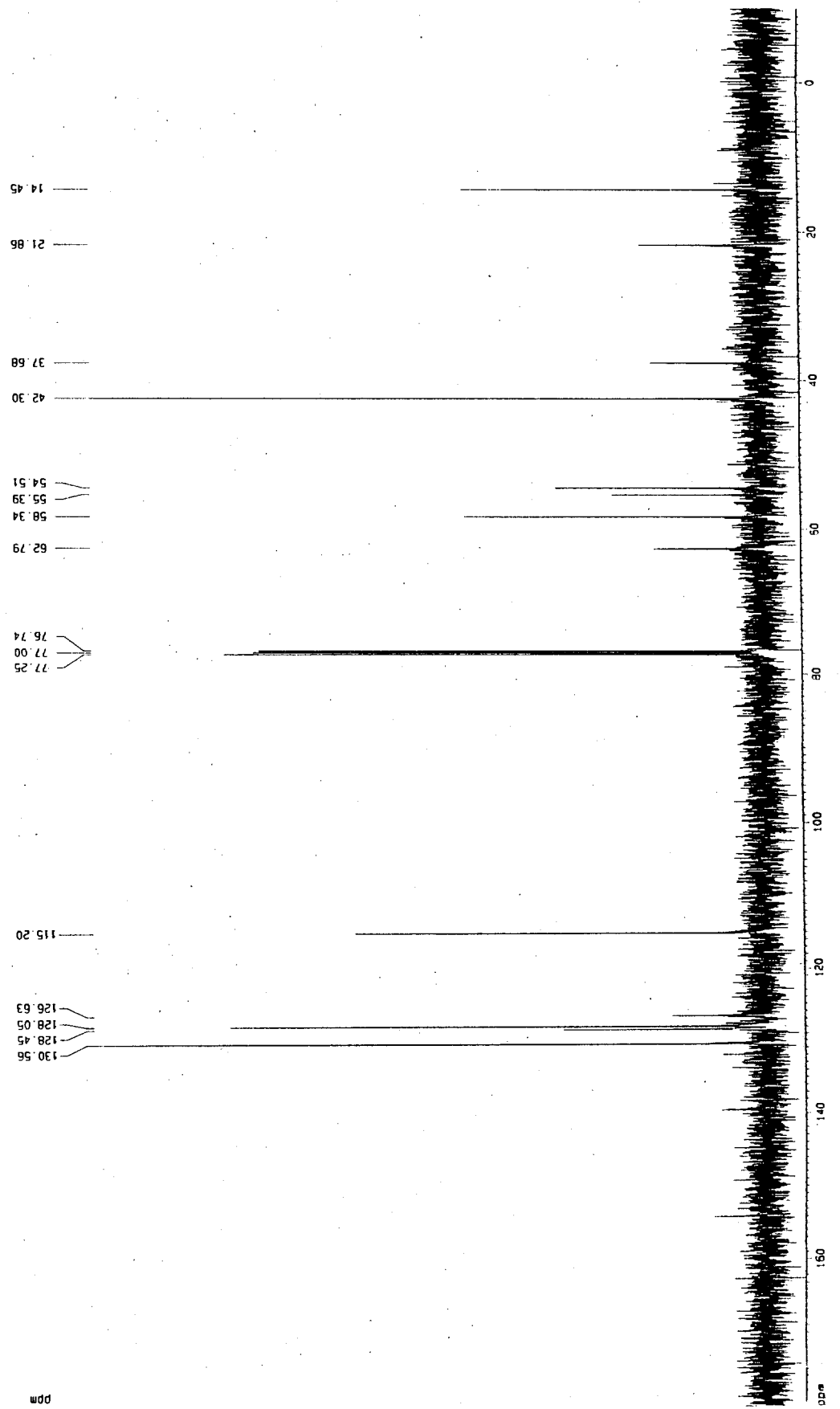


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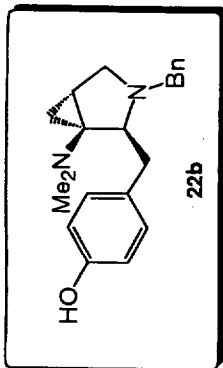
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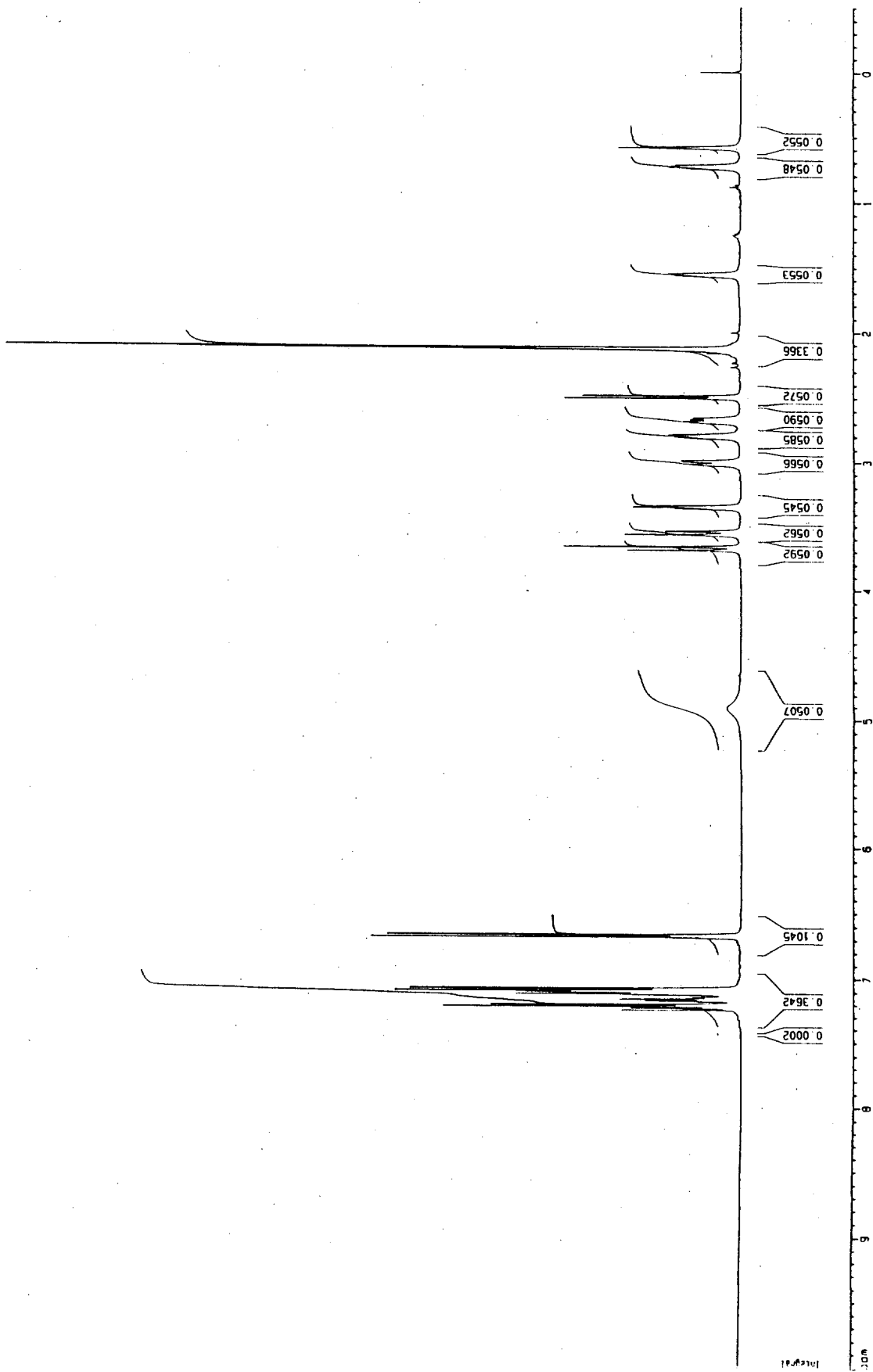


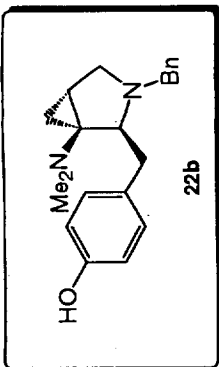
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 P65 118.37
 P66 118.37
 P67 118.37
 P68 118.37
 P69 118.37
 P70 118.37
 P71 118.37
 P72 118.37
 P73 118.37
 P74 118.37
 P75 118.37
 P76 118.37
 P77 118.37
 P78 118.37
 P79 118.37
 P80 118.37
 P81 118.37
 P82 118.37
 P83 118.37
 P84 118.37
 P85 118.37
 P86 118.37
 P87 118.37
 P88 118.37
 P89 118.37
 P90 118.37
 P91 118.37
 P92 118.37
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 P94 118.37
 P95 118.37
 P96 118.37
 P97 118.37
 P98 118.37
 P99 118.37
 P100 118.37

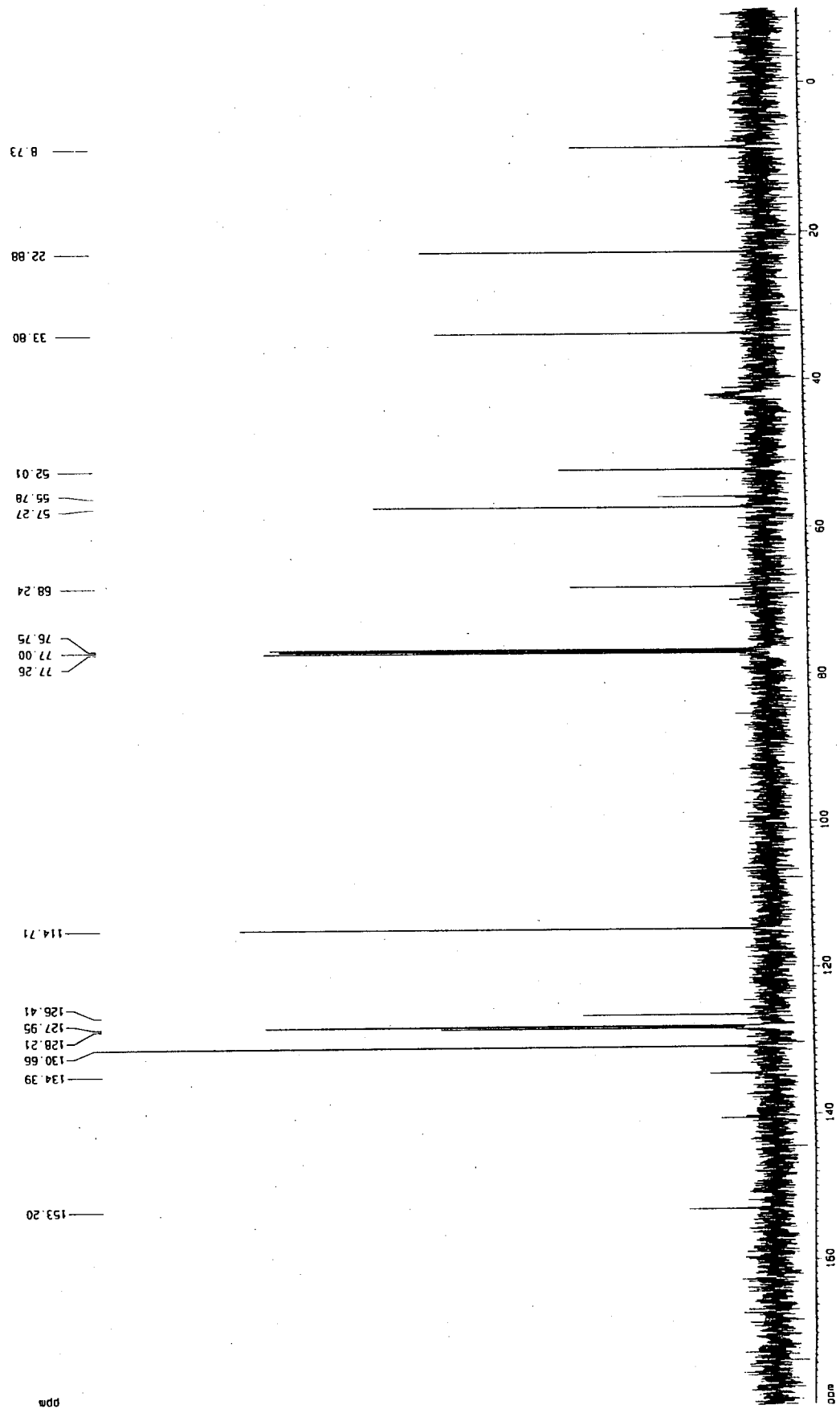
F2 - Processing Parameters
 SI 16384
 SF 500.350000000 MHz
 SW 500.350000000 MHz
 FWHM 0.10 Hz
 LB 0.10 Hz
 GB 0.00 Hz
 PC 1.40

1D NMR plot parameters
 CX 30.00 cm
 FIP 10.000 ppm
 FI 5003.90 Hz
 F2P -1.500 ppm
 F2 118.37 MHz
 F3 118.37 MHz
 F4 118.37 MHz
 F5 118.37 MHz
 F6 118.37 MHz
 F7 118.37 MHz
 F8 118.37 MHz
 F9 118.37 MHz
 F10 118.37 MHz
 F11 118.37 MHz
 F12 118.37 MHz
 F13 118.37 MHz
 F14 118.37 MHz
 F15 118.37 MHz
 F16 118.37 MHz
 F17 118.37 MHz
 F18 118.37 MHz
 F19 118.37 MHz
 F20 118.37 MHz
 F21 118.37 MHz
 F22 118.37 MHz
 F23 118.37 MHz
 F24 118.37 MHz
 F25 118.37 MHz
 F26 118.37 MHz
 F27 118.37 MHz
 F28 118.37 MHz
 F29 118.37 MHz
 F30 118.37 MHz
 F31 118.37 MHz
 F32 118.37 MHz
 F33 118.37 MHz
 F34 118.37 MHz
 F35 118.37 MHz
 F36 118.37 MHz
 F37 118.37 MHz
 F38 118.37 MHz
 F39 118.37 MHz
 F40 118.37 MHz
 F41 118.37 MHz
 F42 118.37 MHz
 F43 118.37 MHz
 F44 118.37 MHz
 F45 118.37 MHz
 F46 118.37 MHz
 F47 118.37 MHz
 F48 118.37 MHz
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 F66 118.37 MHz
 F67 118.37 MHz
 F68 118.37 MHz
 F69 118.37 MHz
 F70 118.37 MHz
 F71 118.37 MHz
 F72 118.37 MHz
 F73 118.37 MHz
 F74 118.37 MHz
 F75 118.37 MHz
 F76 118.37 MHz
 F77 118.37 MHz
 F78 118.37 MHz
 F79 118.37 MHz
 F80 118.37 MHz
 F81 118.37 MHz
 F82 118.37 MHz
 F83 118.37 MHz
 F84 118.37 MHz
 F85 118.37 MHz
 F86 118.37 MHz
 F87 118.37 MHz
 F88 118.37 MHz
 F89 118.37 MHz
 F90 118.37 MHz
 F91 118.37 MHz
 F92 118.37 MHz
 F93 118.37 MHz
 F94 118.37 MHz
 F95 118.37 MHz
 F96 118.37 MHz
 F97 118.37 MHz
 F98 118.37 MHz
 F99 118.37 MHz
 F100 118.37 MHz

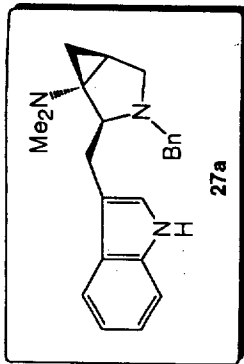




Current Data Parameters
 NAME [-888C13]
 EXPNO 1
 PROCNO 1
 F2 - Acquisition Parameters
 Date_ 991224
 Time 15:11
 INSTRUM spect
 PROBNM 5 ac Dat1 13
 PULPROG zgpg30
 ID 85378
 SOLVENT CDCl3
 NS 28
 DS 2
 SWH 3333.33 Hz
 FIDRES 0.000264 Hz
 AQ 0.983000 sec
 RG 65384
 DB 15.000 usec
 DE 18.75 usec
 TE 300.2 K
 FL1 40 dB
 D11 0.03000000 sec
 D1 2.00000000 sec
 D2 2.00000000 sec
 CHPROG waltz16
 P31 107.00 usec
 S3 18 dB
 P1 5.00 usec
 DE 18.75 usec
 SFO1 125.830448 MHz
 NUC1 13C
 F2 - Processing parameters
 SI 32788
 SF 125.8231705 MHz
 WHW 0
 SSB 0
 LB 1.30 Hz
 GB 0
 PC 1.40
 10 MHz plot parameters
 CX 20.00 cm
 FIP 180.000 ppm
 FI 22648.37 Hz
 F2 -1258.23 Hz
 PCQA 5.42857 cm²/s
 K1C4 683.04010 Hz/s



S12

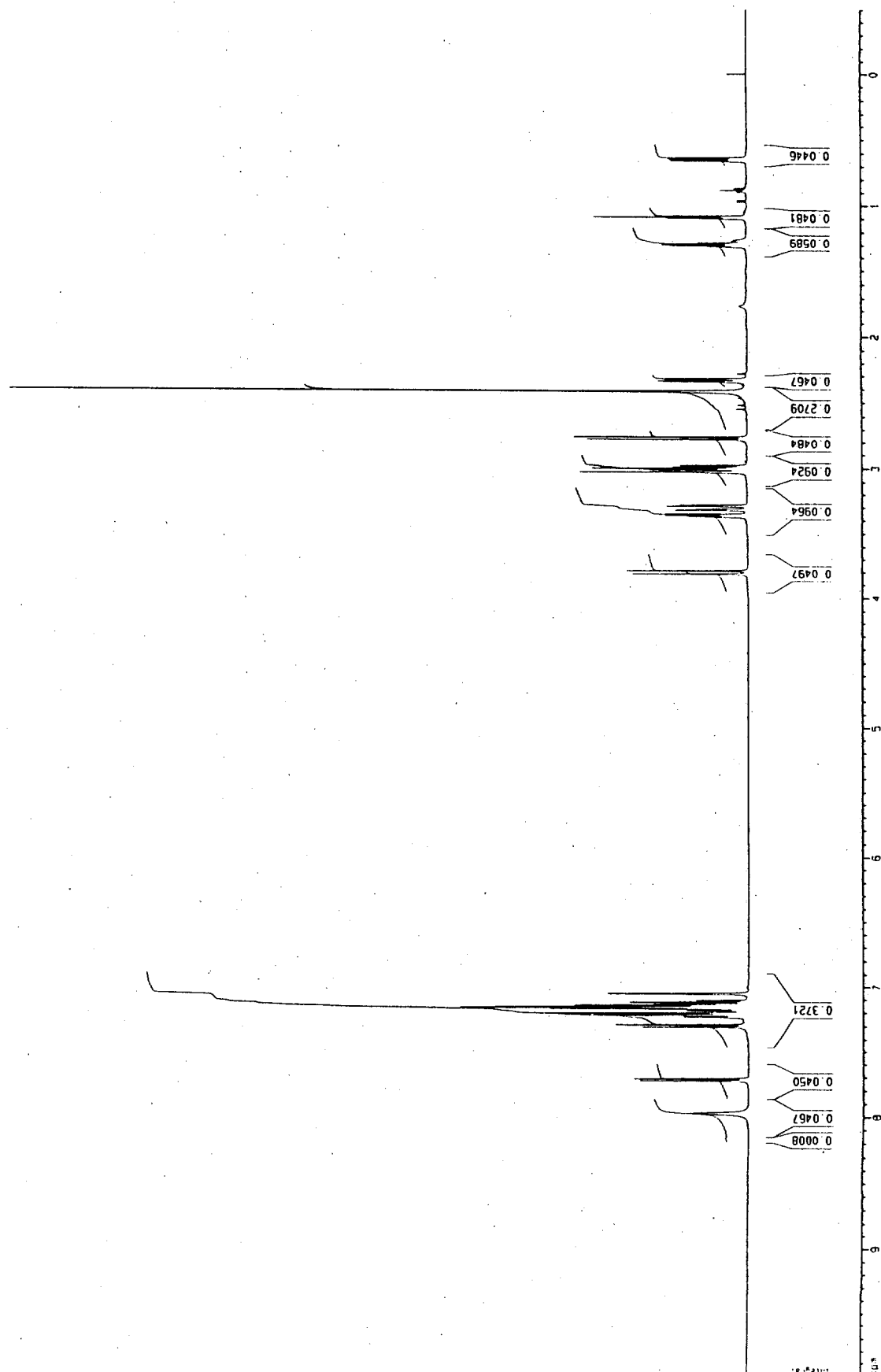


Current Data Parameters
 Name: [REDACTED]
 Sample: 1
 P0000: 1

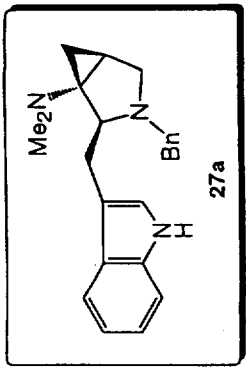
F2 - Acquisition Parameters
 Date_: 990121
 Time: 14.23
 INSTRUM: AHS500
 P0000: 5 mm Dual 13
 PULPROG: zg
 ID: 32765
 SOLVENT: CDCl3
 NS: 1000
 DS: 2
 SFO1: 500.362518 MHz
 NUC1EUS: 1H

F2 - Processing parameters
 SI: 6554
 SF: 500.362518 MHz
 WDW: EM
 SSB: 0
 LB: 0.10 Hz
 GB: 0
 PC: 1.40

1D NMR plot parameters
 SI: 6554
 SF: 500.362518 MHz
 P1: 10.000000
 F1: 500.362518 MHz
 F2: -250.20 MHz
 SFO1: 500.362518 MHz
 HZCN: 130.11702 MHz



S13

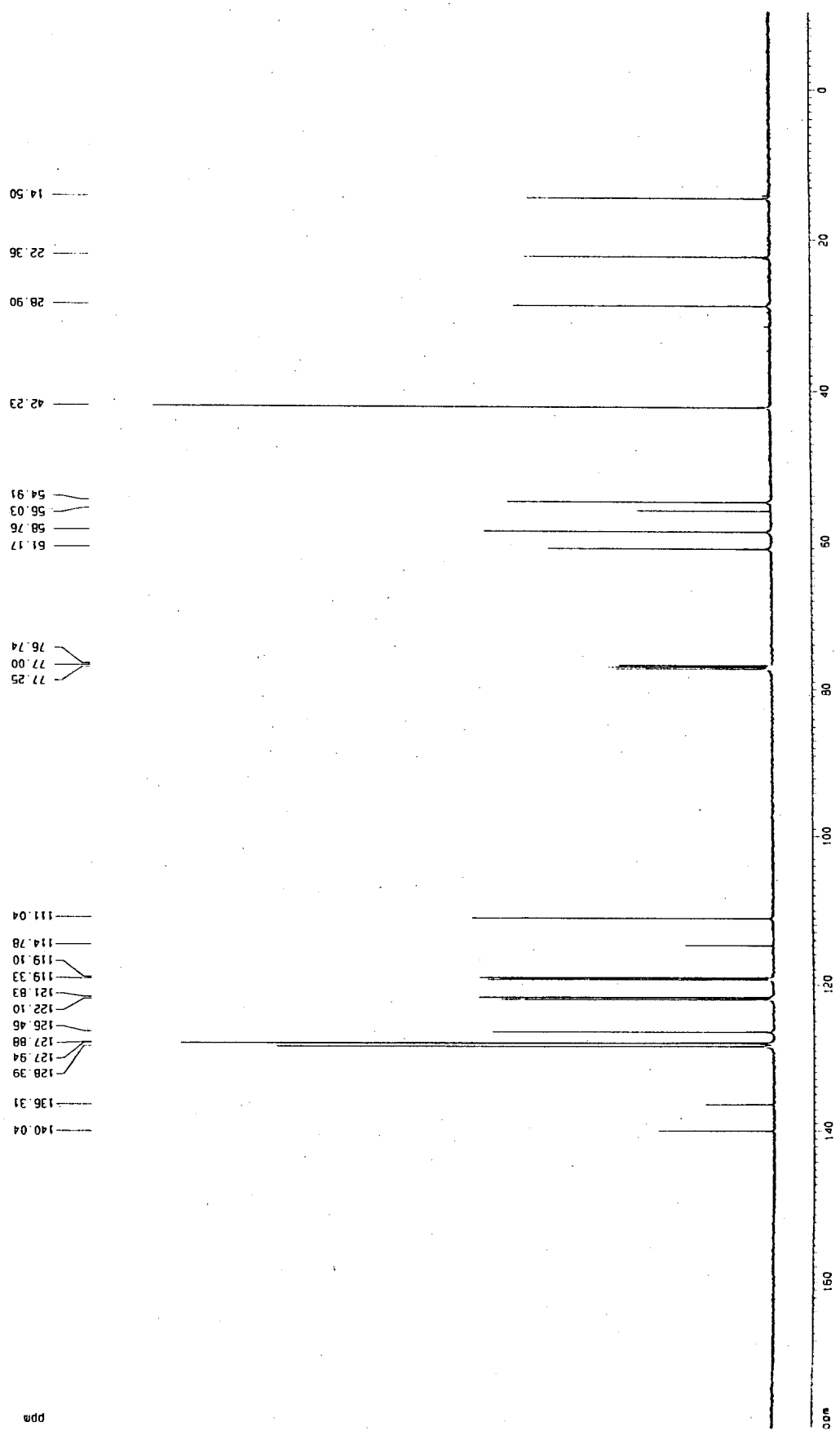


Current Data Parameters
 NAME 1-686C13
 EXPNO 1
 PROCNO 1

F2 - Acquisition Parameters
 Date_ 990121
 Time 14.37
 INSTRUM spect
 PULPROG zgpg30
 ID 685336
 SOLVENT CDCl3
 NS 633
 DS 2
 SWH 3333.332 Hz
 FIDRES 0.300000 Hz
 AQ 0.999999 SEC
 RG 63.004
 DM 15.000 USEC
 DE 18.75 USEC
 TE 300.0 K
 K1 40 dB
 D11 0.000000 SEC
 S4 24 dB
 CDPRG2 waltz16
 CPDPRG2 waltz16 SEC
 P31 107.00 USEC
 S2 18 dB
 P1 5.00 USEC
 DE 18.75 USEC
 SF01 125.638148 MHz
 NUC16S 13C

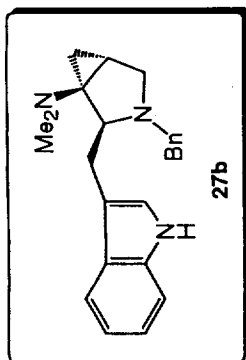
F2 - Processing parameters
 SI 32768
 SF 125.631748 MHz
 WDW EM
 SSB 0
 LB 1.00 Hz
 GB 0
 PC 1.49

ID MS plot parameters
 CX 35.00 cm
 FIP 180.000 Da
 FI 28048.17 Hz
 FPR -10.000 Da
 FZ 12.000 Hz
 PC 1.000 Hz
 MZCM 683.0410 Hz/C1



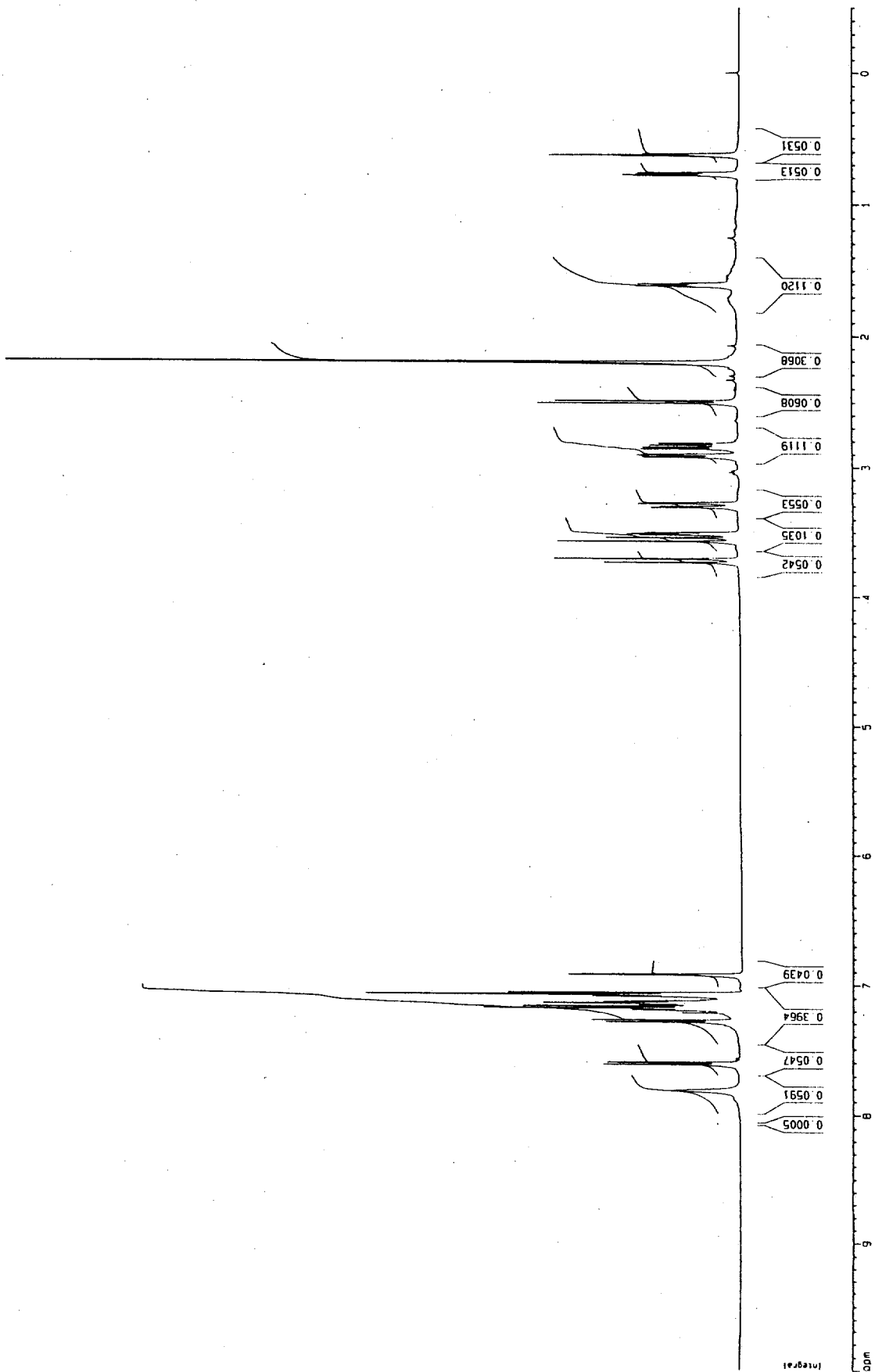
ppm

HS

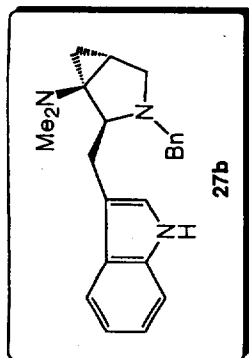


Current Data Parameters
 Name 1-504
 EXPNO 1
 PROCNO 1

F2 - Acquisition Parameters
 Date_ 980122
 Time 15 41
 INSTRUM spect
 AMPL 500
 PULPROG zgpg30
 5 mm QNP 13
 TD 32768
 SOLVENT CDCl₃
 NS 17
 DS 2
 SWH 6024.096 Hz
 FIDRES 0.183641 Hz
 AQ 2.719760 sec
 RG 128
 DE 83.460 VPP
 TE 300.0 K
 K1 3.00
 K2 1.00
 K3 1.00
 K4 1.00
 K5 1.00
 K6 1.00
 K7 1.00
 K8 1.00
 K9 1.00
 K10 1.00
 K11 1.00
 K12 1.00
 K13 1.00
 K14 1.00
 K15 1.00
 K16 1.00
 K17 1.00
 K18 1.00
 K19 1.00
 K20 1.00
 K21 1.00
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 K27 1.00
 K28 1.00
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 K31 1.00
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 K82 1.00
 K83 1.00
 K84 1.00
 K85 1.00
 K86 1.00
 K87 1.00
 K88 1.00
 K89 1.00
 K90 1.00
 K91 1.00
 K92 1.00
 K93 1.00
 K94 1.00
 K95 1.00
 K96 1.00
 K97 1.00
 K98 1.00
 K99 1.00
 K100 1.00



S15



Current Data Parameters
 NAME I-80413
 EXPNO 1
 PROCNO 1

F2 - Acquisition Parameters
 DATE_ 991222
 TIME 15.59
 INSTRUM sm500
 PROBHD 5 mm QNP 13
 PULPROG zgpg30
 ID 8336
 SOLVENT CDCl3
 NS 2
 DS 2
 SWH 3333.132 Hz
 FIDRES 0.508236 Hz
 AQ 0.9820900 sec
 RG 16384
 DM 15.000 usec
 DE 18.75 usec
 TE 300.2 K
 HL 1.40 cm
 SI 0.02000000 sec
 SFO1 2.00000000 sec
 D1 2.00000000 sec
 D2 107.00 usec
 D3 18.00 usec
 P1 5.00 usec
 P2 18.75 usec
 DE 18.75 usec
 SFO1 125.0052148 MHz
 NUC1EUS 13C

F2 - Processing parameters
 SI 32768
 SF 125.0052148 MHz
 WDW EM
 SSB 0
 LB 1.50 Hz
 GB 0
 PC 1.40

ID Web plot parameters
 CO 30.00 usec
 FIP 100.000 usec
 F1 22848.17 Hz
 F2P -10.000 usec
 F2 -1254.23 Hz
 PPH2 5.42857 usec
 N2CN 503.0410 usec

