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Supporting Information

Catalyst- and solvent-free one-pot synthesis of some novel polyheterocycles from aryldiazenyl salicylaldehyde derivatives

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General information and experimental procedures

Solvents were dried by the standard procedures. ^{1}H and ^{13}C NMR spectra were determined in $CDCl_3$ or DMSO-d_6 on a BRUKER Avance (^{1}H: 400 MHz, ^{13}C: 100 MHz). IR spectra were obtained on a SHIMADZU FT-IR 8300 spectrophotometer using KBr disc. Mass spectra were obtained on SHIMADZU LCMS 2010 instrument. All the reactions were monitored by thin layer chromatography (TLC) using silica gel 60 F254 plates (Merck). The melting points were determined in open capillary tube on a TEMPO melting point apparatus and are uncorrected. The UV spectra were obtained on SHIMADZU UV-160 A.

General procedure

- **Synthesis of O-allylated or prenylated-5-aryldiazenyl salicylaldehydes (3a-d).**

To a stirred solution of 10 mmol diazenylsalicylaldehyde **2** in 25 ml DMF, in the presence of 15 mmol anhydrous potassium carbonate, was added dropwise a solution of 1.5 mmol of allyl bromide in 5 ml DMF. The resulted mass was then further stirred at room temperature to complete the reaction as monitored by TLC (10-12 h). It was then poured into ice with constant stirring. The solid precipitates were filtered, washed with cold water, and dried at room temperature. The products **3a-b** were received quantitatively. Similarly prenyl bromide was used to obtain O-prenylated salicylaldehyde derivatives **3c-d**.

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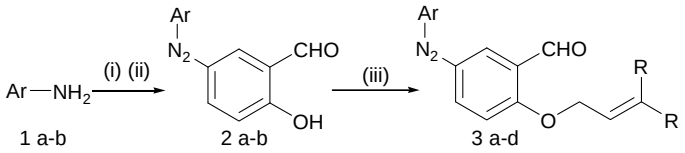


1 a-b

2 a-b

3 a-d

1a, Ar = Ph

1b, Ar = 4-ClPh

3a: Ar = Ph, R= H

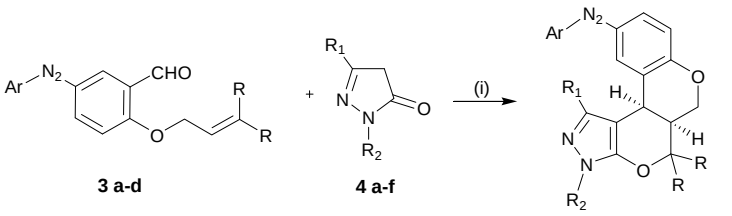
3b: Ar = 4-ClPh, R= H

3c: Ar = Ph, R= Me

3d: Ar = 4-ClPh, R= Me

- **Synthesis of benzopyran derivatives (7a–p) & (9a-d)**

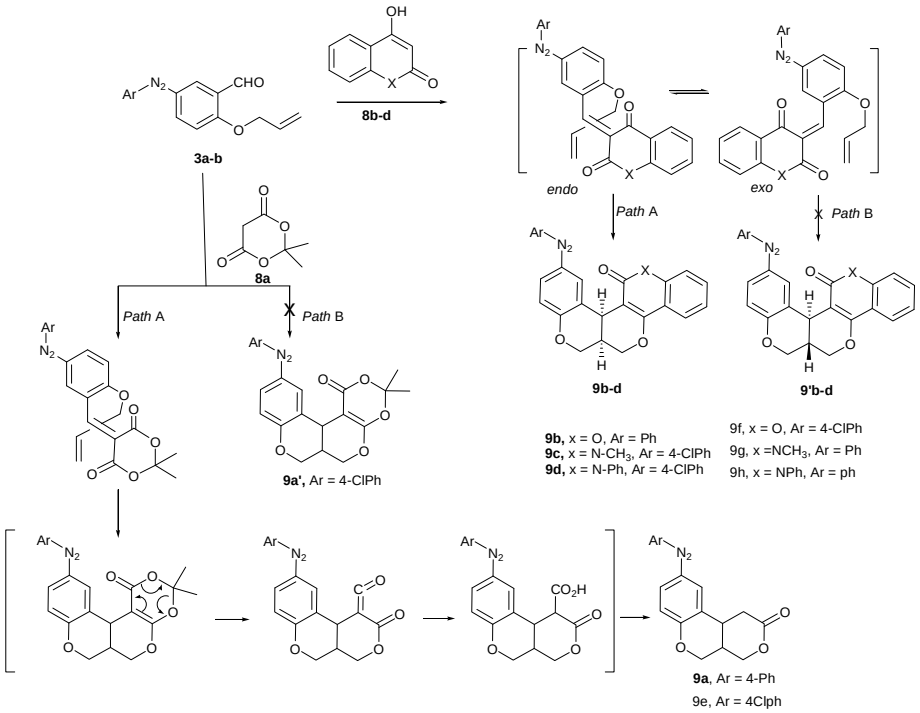
In a round-bottom flask, O-allylated/prenylated-5-aryldiazenyl salicylaldehydes **3** (3.7 mmol) and 5-pyrazolones **4** (3.7 mmol) or diketone **8a-d** (3.7 mmol) was heated at 180˚C until the substrate **3** disappeared as monitored by TLC. It gave products in good yields. Crude product left was then subjected to a column chromatography, employing an ethyl acetate/hexane (3:7) eluent, affording compounds **7a-p or 9a-h** in pure product with excellent yields.



3 a-d **4 a-f**

7a-l : Ar = C_6H_5-, 4-ClC_6H_4-, R = H, R_1 = Me

7m-p : Ar = C_6H_5-, 4-ClC_6H_4-, R = R_1 =Me



9b, x = O, Ar = Ph
9c, x = N-CH_3, Ar = 4-ClPh
9d, x = N-Ph, Ar = 4-ClPh

9f, x = O, Ar = 4-ClPh
9g, x =NCH_3, Ar = Ph
9h, x = NPh, Ar = ph

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Characterization of compounds

(5a*R*,11b*S*)-1-methyl-3-phenyl-10-[(*E*)-phenyldiazenyl]-3,5a,6,11b-tetrahydro-5*H*-chromeno [4',3':4,5]pyrano[2,3-c]pyrazole (7a): Isolated yield (1.23 g, 78%) as yellow crystals, mp 190-192˚C; λ_{max}(DMF)/nm 348 (ε/dm^3 mol^{-1} cm^{-1} 17721), 270 (12552); ν_{max}/cm^{-1} = 2957, 2927, 1490, 1467, 1241, 1092, 1026, 831, 758; δ_H(400 MHz; $CDCl_3$) 2.42 (s, 3H, Me), 2.60 (s, 1H, C(5_a)H), 4.16 (t, 1H, J_b= 10.4 Hz, C(6)H), 4.33 (d, 1H, J = 4.8 Hz, C(11_b)H), 4.43 (m, 2H, C(5)H), 4.65 (dd, 1H, J = 2.4 Hz, C(6)H), 6.99 (d, 1H, J = 10.0 Hz ArH), 7.22-7.87 (m, 12H ArH); δ_C(100 MHz; $CDCl_3$) 14.41 (CH_3), 29.51 (CH, benzylic methane), 30.12 (CH, benzylic methane), 66.20 (CH_2), 68.52 (CH_2), 118.10, 120.34, 122.73, 123.65, 125.66, 126.04, 129.56, 129.86, 131.36 (ArCH), 99.48, 125.15, 138.65, 145.11, 146.49, 146.77, 148.09, 149.24, 149.95, 152.40, 152.81 (ArC); m/z (ESI) 423.1 [M + H$^+$], $C_{26}H_{22}N_4O_2$: calcd. C 73.92, H 5.25, N 13.26; found C 73.80, H 5.20, N 13.30.

N. B.: To justify presence of chloroform appeared in single crystal X-Ray data of compound 7a, we have carried out GC-MS as well as CHN analysis. As expected, both the reports confirmed the presence of chloroform molecule in compound **7a.**

CHN analysis report of compound 7a

Cash paid

Central Salt & Marine Chemicals Research Institute

Gijubhai Badheka Marg,
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Date: 5/11/2011

To,
Mr. Rikin Patel,
V.V. Nagar,
Anand.
Ph. No. 9879594272

ID No.: 16358/114755

Sr. No.	Test	No. of sample	Rate	Discount (Educational 75%)	Total Amount in Rs.
1	C,H,N ananlysis	1	2000.00	1500.00	500.00

Analysis report
(C H N S / O Ananlysis)
Analytical Science Discipline

Instrument : Perkin Elmer-2400

No. of samples : 1

Sr.no.	Sample code	C	H	N	S	O
1	D1	61.01	4.80	10.56	-	-

Reference standard (Theoritical value)

Acetanilide	71.09	6.71	10.36	-	-
Cystine	29.99	5.03	11.67	-	-

Reference standard (Experimental value)

Acetanilide	71.03	6.52	10.46	-	-
Cystine	29.91	5.10	11.59	-	-

Signature of analyst :

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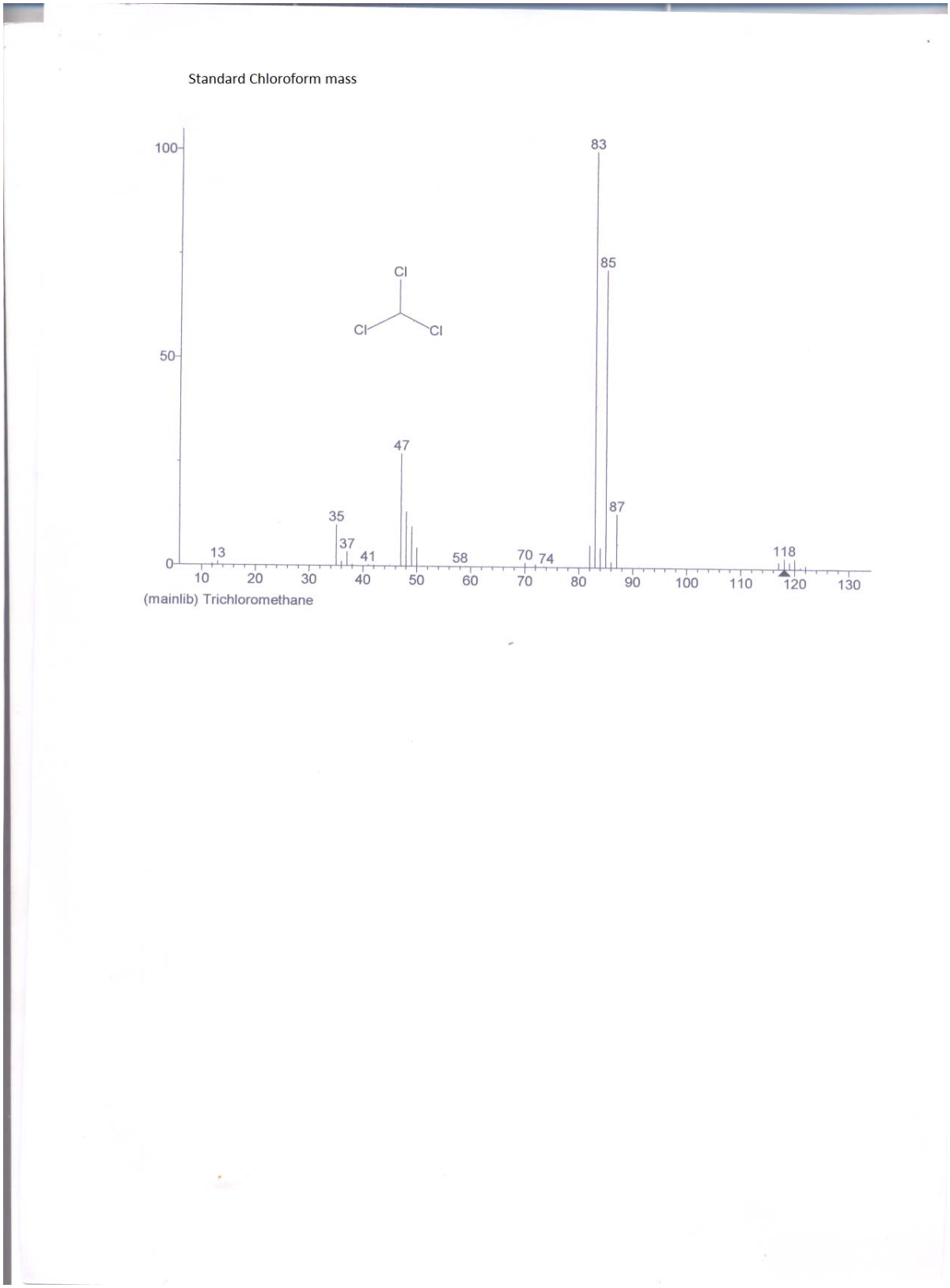
GC-MS trace of Compound 7a



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N.B.: Obviously, for others, elemental analysis without crystallizing from chloroform but chromatographically pure ones, it showed expected results. Below is an elemental analysis of compound 7a isolated by column chromatography.

Cash paid

Central Salt & Marine Chemicals Research Institute

Gijubhai Badheka Marg,
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E-Mail: salt@csmcri.org

To,
Mr. Rikin Patel,
V.V. Nagar,
Anand.

ID No.: 16342/114755

Sr. No.	Test	No. of sample	Rate	Discount (Educational 75%)	Total Amount in Rs.
1	C,H,N ananlysis	1	2000.00	1500.00	500.00

Analysis report
(C H N S / O Ananlysis)
Analytical Science Discipline

Instrument : Perkin Elmer-2400

No. of samples : 1

Sr.no.	Sample code	C	H	N	S	O
1	D1	73.82	5.15	13.37	-	-

Reference standard (Theoritical value)

Acetanilide	71.09	6.71	10.36	-	-
Cystine	29.99	5.03	11.67	-	-

Reference standard (Experimental value)

Acetanilide	71.05	6.57	10.26	-	-
Cystine	29.88	5.15	11.60	-	-

Signature of analyst :

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(5a*R*, 11b*S*)-3-(3-chlorophenyl)-1-methyl-10-[(*E*)-phenyldiazenyl]-3,5a,6,11b-tetrahydro-5*H*-chromeno[4',3':4,5]pyrano[2,3-*c*]pyrazole (7b): Isolated yield (1.29 g, 75%) as yellow crystals, mp 191-193˚C; λmax(DMF)/nm 348 (ε/dm3 mol-1 cm-1 17628), 271 (15803); νmax/cm-1 = 2950, 2930, 2890, 1490, 1481, 1235, 1092, 1089, 1030, 898, 822, 763; δH(400 MHz; CDCl3) 2.57 (m, 4H, Me, C(5a)H), 4.24 (d, 1H, J = 4.8 Hz, C(11b)H), 4.35 (t, 1H, Jb = 10.8 Hz, C(6)H), 4.47 (m, 2H, C(5)H), 4.57 (dd, 1H, J = 2.8 Hz, C(6)H), 6.95 (d, 1H, J = 8.4 Hz, ArH), 7.18–7.94 (m, 11H, ArH); δC(100 MHz; CDCl3) 14.12 (CH3), 30.11 (CH, benzylic methane), 30.77 (CH, benzylic methane), 66.12 (CH2), 68.49 (CH2), 117.72, 118.19, 120.29, 122.67, 123.37, 125.69, 125.84, 129.03, 130.00, 130.53 (ArCH), 99.42, 123.60, 134.69, 139.23, 147.21, 147.61, 149.41, 152.62, 154.88 (ArC); m/z (ESI) 457.1 [M + H+]; $C_{26}H_{21}N_4O_2Cl$: calcd. C 68.34, H 4.63, N 12.26; found C 68.25, H 4.58, N 12.06.

(5a*R*,11b*S*)-1,3-diphenyl-10-[(*E*)-phenyldiazenyl]-3,5a,6,11b-tetrahydro-5*H*-chromeno[4',3':4,5]pyrano[2,3-*c*]pyrazole (7c): Isolated yield (1.47 g, 80%) as yellow crystals, mp 202-204˚C; λ_{max}(DMF)/nm 348 (ε/dm^3 mol^{-1} cm^{-1} 15738), 275 (23353); ν_{max}/cm^{-1} 2957, 2920, 2890, 1491, 1471, 1238, 1094, 1028, 888, 817, 760; δ_H(400 MHz; $CDCl_3$) 2.12 (m, 1H, C(5a)H), 4.42-4.62 (m, 5H), 6.92–8.06 (m, 18H, ArH); δ_C(100 MHz; $CDCl_3$) 30.75 (CH, benzylic methane), 31.09 (CH, benzylic methane), 66.64 (CH_2), 68.00 (CH_2), 117.38, 121.14, 122.58, 123.75, 125.64, 126.21, 127.33, 128.34, 128.84, 128.93, 128.96, 130.31 (ArCH), 98.44, 123.78, 134.13, 138.31, 147.42, 148.85, 149.81, 152.54, 154.55 (ArC); m/z (ESI) 485.0 [M + H$^+$]; $C_{31}H_{24}N_4O_2$: calcd. C 76.84, H 4.99, N 11.56; found C 76.70, H 4.87, N 11.48.

(5a*R*,11b*S*)-3-(2,5-dichlorophenyl)-1-methyl-10-[(*E*)-phenyldiazenyl]-3,5a,6,11b-tetrahydro-5*H*-chromeno[4',3':4,5]pyrano[2,3-*c*]pyrazole (7d): Isolated yield (1.55 g, 83%) as brown crystals, mp 96-98˚C; λ_{max}(DMF)/nm 348 (ε/dm^3 mol^{-1} cm^{-1} 18306), 270 (8969); ν_{max}/cm^{-1} 2957, 2927, 1490, 1467, 1241, 1092, 1026, 831, 758; δ_H(400 MHz; $CDCl_3$) 2.55 (s, 4H, Me, C(5a)H), 4.28 (m, 2H, C(6)H), 4.42 (m, 3H, c(5)H, C(11b)H), 6.96 (d, 1H, *J* = 8.4 Hz ArH), 7.33– 7.99 (m, 10H, ArH); δ_C(100 MHz; $CDCl_3$) 14.24 (CH_3), 30.33 (CH, benzylic methane), 30.97 (CH, benzylic methane), 65.95 (CH_2), 68.66 (CH_2), 117.77, 122.70, 123.14, 126.29, 129.05, 129.57, 130.02, 130.56, 131.06 (ArCH), 97.99, 135.47, 147.13, 148.22, 150.27, 152.63, 155.06 (ArC) $C_{26}H_{20}Cl_2N_4O_2$: calcd. C 63.55, H 4.10, N 11.40; found C 63.49, H 4.18, N 11.49.

(5a*R*,11b*S*)-1-methyl-3-(4-methylphenyl)-10-[(*E*)-phenyldiazenyl]-3,5a,6,11b-tetrahydro-5*H*-chromeno[4',3':4,5]pyrano[2,3-*c*]pyrazole (7e): Isolated yield(1.41 g, 85%) as yellow crystals, mp 179-181˚C; λ_{max}(DMF)/nm 348 (ε/dm^3 mol^{-1} cm^{-1} 19138), 270 (14154); ν_{max}/cm^{-1} 2960, 2923, 2892, 1487, 1444, 1222, 1094, 1032, 818, 767; δ_H(400 MHz; $CDCl_3$) 2.37 (s, 3H, p-Me), 2.60 (m, 4H, Me, C(5a)H), 4.25 (d, 1H, *J* = 4.8 Hz, C(11_b)H), 4.33 (t, 1H, J_b = 3.2 Hz, C(6)H), 4.47 (m, 2H, C(5)H), 4.54 (dd, 1H, *J* = 3.2 Hz, C(6)H), 6.95 (d, 1H, *J* = 8.8 Hz, ArH), 7.21–7.95 (m, 11H, ArH),; δ_C(100 MHz; $CDCl_3$) 13.95 (CH_3), 20.97 (CH_3), 30.11 (CH, benzylic methane), 30.88 (CH, benzylic methane), 66.16 (CH_2), 68.41 (CH_2), 117.72, 120.92, 122.68, 123.25, 125.99, 129.04, 129.58, 130.49, (ArCH), 122.32, 123.70, 129.77, 146.57, 147.22, 149.11, 152.64, 154.90, 155.36 (ArC) $C_{27}H_{24}N_4O_2$: calcd. C 74.29, H 5.54, N 12.84; found C 74.33, H 5.51, N 12.78.

(5a*R*,11b*S*)-3-(2-chlorophenyl)-1-methyl-10-[(*E*)-phenyldiazenyl]-3,5a,6,11b-tetrahydro-5*H*-chromeno[4',3':4,5]pyrano[2,3-*c*]pyrazole (7f): Isolated yield (1.42 g, 82%) as yellow crystals, mp 114-115˚C; λ_{max}(DMF)/nm 348 (ε/dm^3 mol^{-1} cm^{-1} 18711), 271 (16305); ν_{max}/cm^{-1} 2924, 1489, 1446, 1249, 1094, 1031, 877, 759; δ_H(400 MHz; $CDCl_3$) 2.57 (m, 4H, Me, C(5a)H), 4.23 (d, 1H, *J* = 4.8 Hz, C(11_b)H), 4.34 (t, 1H, J_b = 10.8 Hz, C(6)H), 4.47 (m, 2H, C(5)H), 4.57 (dd, 1H, *J* = 2.8 Hz, C(6)H), 6.95 (d, 1H, *J* = 8.8 Hz, ArH), 7.18–7.94 (m, 11H, ArH); δ_C(100 MHz; $CDCl_3$) 14.15 (CH_3), 30.12 (CH, benzylic methane), 30.77 (CH, benzylic methane), 66.12 (CH_2), 68.48 (CH_2), 117.71, 118.15, 120.26, 122.67, 123.35, 125.63, 125.85, 129.03, 129.99, 130.52, (ArCH),99.40, 123.63, 125.64, 134.68, 139.29, 147.20, 147.61, 149.39, 152.62, 154.88 (ArC) $C_{26}H_{21}ClN_4O_2$: calcd. C 68.34, H 4.63, N 12.26; found C 68.28, H 4.55, N 12.38.

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(5a*R*,11b*S*)-10-[(*E*)-(4-chlorophenyl)diazenyl]-1-methyl-3-phenyl-3,5a,6,11b-tetrahydro-5*H*-chromeno[4',3':4,5]pyrano[2,3-*c*]pyrazole (7g): Isolated yield (1.38 g, 80%) as yellow crystals, mp 219-221°C; λ_{max}(DMF)/nm 361 (ε/dm^3 mol^{-1} cm^{-1} 17274), 270 (13614); ν_{max}/cm^{-1} 2926, 1490, 1440, 1249, 1000, 1030, 878, 760; δ_H(400 MHz; $CDCl_3$) 2.55 (s, 3H, Me), 2.58 (m, 1H, C(5a)H), 4.24 (d, 1H, J = 4.8 Hz, C(11_b)H), 4.32 (t, 1H, J_b = 10.4 Hz, C(6)H), 4.47 (m, 2H, C(5)H), 4.54 (dd, 1H, J = 2.0 Hz, C(6)H), 6.94 (d, 1H, J = 8.8, ArH), 7.21–7.97 (m, 11H, ArH); δ_C(100 MHz; $CDCl_3$) 14.25 (CH_3), 30.22 (CH, benzylic methane), 30.87 (CH, benzylic methane), 66.27 (CH_2), 68.23 (CH_2), 117.71, 120.49, 123.26,123.91, 125.71, 126.26, 128.93, 129.25 (ArCH), 98.79, 124.06, 136.28, 138.48, 146.97, 147.01, 149.08, 151.02, 155.20 (ArC) $C_{26}H_{21}ClN_4O_2$: calcd. C 68.34, H 4.63, N 12.26; found C 68.25, H 4.50, N 12.32.

(5a*R*,11b*S*)-3-(3-chlorophenyl)-10-[(*E*)-(4-chlorophenyl)diazenyl]-1-methyl-3,5a,6,11b-tetrahydro-5*H*-chromeno[4',3':4,5]pyrano[2,3-*c*]pyrazole (7h): Isolated yield (0.76 g, 84%) as yellow crystals, mp 180-181°C; ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 2.55 (s, 3H, Me), 2.58 (m, 1H, C(5a)H) 2.57 (m, 1H, C(11b)H), 4.34 (t, J = 10.4 Hz, 1H, C(6)H), 4.48 (m, 2H), 4.59 (dd, J = 3.2 Hz, 1H, C(6)H), 6.95–7.94 (m, 11H, ArH); ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 14.23, 30.14, 30.77, 66.19, 68.38, 99.23, 117.76, 118.00, 120.16, 123.38, 123.85, 123.91, 125.50, 126.08, 129.26, 129.95, 134.67, 136.32, 139.55, 147.02, 147.57, 149.31, 150.99, 155.15; λ_{max}(DMF)/nm 360 (ε/dm^3 mol^{-1} cm^{-1} 17644), 271 (17128); MS: calcd. for $C_{26}H_{20}Cl_2N_4O_2$ [M + H] 491.10; found 491.29; $C_{26}H_{20}Cl_2N_4O_2$ (490.10 g/mol): calcd. C 63.55, H 4.10, N 11.40; found C 63.48, H 4.18, N 11.55 $C_{26}H_{20}Cl_2N_4O_2$: calcd. C 63.55, H 4.10, N 11.40; found C 63.48, H 4.18, N 11.55.

(5aR,11bS)-10-[(E)-(4-chlorophenyl)diazenyl]-1,3-diphenyl-3,5a,6,11b-tetrahydro-5H-chromeno[4',3':4,5]pyrano[2,3-c]pyrazole (7i): Isolated yield (0.70 g, 86%) as yellow crystals, mp 209-210°C; IR (KBr): ν = 2915, 1480, 1444, 1250, 1094, 1035, 875, 755; ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 2.74 (m, C(5a)H), 4.32 (t, J = 11.2 Hz, 1H, C(6)H), 4.54 (m, 2H, C(5)H), 4.59 (dd, J = 3.6 Hz, 1H, C(6)H), 4.74 (d, J = 4.8 Hz, 1H, C(11_b)H), 6.90 (d, J = 8.4 Hz, 1H, ArH), 7.25–8.02 (m, 16H, ArH); ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 30.69, 30.98, 66.62, 67.89, 98.28, 117.43, 120.94, 123.80, 123.90, 123.98, 125.66, 126.10, 127.18, 128.20, 128.87, 128.93, 129.09, 134.40, 136.10, 138.48, 147.22, 148.79, 149.72, 150.87, 154.75; λ_{max}(DMF)/nm 360 (ε/dm^3 mol^{-1} cm^{-1} 16943), 274 (25725); MS: calcd. for $C_{31}H_{23}ClN_4O_2$ [M + H] 519.15; found 519.43; $C_{31}H_{23}ClN_4O_2$ (518.15 g/mol): calcd. C 71.74, H 4.47, N 10.80; found C 71.60, H 4.55, N 11.05.

(5aR,11bS)-10-[(E)-(4-chlorophenyl)diazenyl]-3-(2,5-dichlorophenyl)-1-methyl-3,5a,6,11b-tetrahydro-5H-chromeno[4',3':4,5]pyrano[2,3-c]pyrazole (7j): Isolated yield(1.40 g, 70%) as yellow crystals, mp 222-224°C; λ_{max}(DMF)/nm 361 (ε/dm^3 mol^{-1} cm^{-1} 17260), 269 (9829); ν_{max}/cm^{-1} 2935, 1485, 1433, 1251, 1087, 1034, 865, 761; δ_H(400 MHz; $CDCl_3$) 2.51 (s, 3H, Me), 2.56 (m, C(5a)H), 4.27 (m, 2H, C(6)H), 4.46 (m, 3H, C(11b)H), 6.95–7.99 (m, 10H, ArH); δ_C(100 MHz; $CDCl_3$) 14.46 (Me), 30.42 (CH, benzylic methane), 31.01 (CH, benzylic methane), 66.05 (CH_2), 68.47 (CH_2), 117.78, 123.09, 123.93, 126.61, 129.27, 129.47, 129.79, 131.00 (ArCH), 97.61, 123.72, 129.91, 132.93, 136.19, 136.31, 146.93, 148.29, 150.01, 151.03, 155.37 (ArC); $C_{26}H_{19}Cl_3N_4O_2$: calcd. C 59.39, H 3.64, N 10.66; found C 59.48, H 3.58, N 10.54.

1-Methyl-3-(4-methylphenyl)-10-(4-chlorophenylazo)-3, 5a, 6, 11b-tetrahydro - 5*H*-benzopyrano [4', 3':4, 5] pyrano[2,3-*c*] pyrazole (7k) : Isolated yield (1.25 g, 70%) as yellow crystals, mp 175-176°C ; λ_{max}(DMF)/nm 360 (ε/dm3 mol-1 cm-1 17673), 270 (14935); ν_{max}/cm^{-1} 2922, 1480, 1443, 1240, 1080, 1022, 754; δ_H(400 MHz; $CDCl_3$) 2.36 (s, 3H, p-tolyl), 2.54 (m, 4H, Me, C(5a)H), 4.23 (d, 1H, J = 4.8 Hz, C(11_b)H), 4.31 (t, 1H, J_b = 10.0 Hz, C(6)H), 4.47 (m, 3H, C(5)H, C(6)H), 6.94 (d, 1H, ArH), 6.96–7.97 (m, 10H, ArH); δ_C(100 MHz; $CDCl_3$) 14.24 (Me), 20.94 (Me), 30.25 (CH, benzylic methane), 30.91 (CH, benzylic methane), 66.30 (CH_2), 68.19 (CH_2), 117.69, 120.61, 123.21, 123.90, 126.32, 129.25, 129.47 (ArCH), 98.55, 124.13, 135.52, 136.02, 136.26, 146.63, 147.00, 148.91, 151.02, 155.21 (ArC); $C_{27}H_{23}ClN_4O_2$: calcd. C 68.86, H 4.92, N 11.90; found C 68.70, H 5.01, N 11.84.

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1,5,5-trimethyl-3-(2-chlorophenyl)--10-phenylazo-3, 5a, 6, 11b-tetrahydro-5*H*-chromeno [4',3':4,5] pyrano[2,3-*c*]pyrazole (7m): Yellow crystals (1.25 g, 80%), mp 212-214˚C; ν_{max}/cm^{-1} 3030, 3000, 1630, 1245, 825, 750, 680; δ_H(400 MHz; $CDCl_3$) 1.55 (s, 3H, Me), 1.60 (s, 3H, Me), 2.23 (s, 3H, Me), 2.25 (m, 1H, C(5a)H), 4.12 (t, 1H, *J* = 11.0 Hz, C(6)H), 4.25 (d, 1H, *J* = 4.0 Hz, C(11$_b$)H), 4.51 (dd, 1H, *J* = 3.2 Hz, C(6)H), 6.92 (d, *J* = 8.4 Hz, 1H, Ar-H), 7.21-8.09 (m, 11H, Ar-H); δ_C(100 MHz; $CDCl_3$) 15.25, 25.48, 26.10, 30.55, 38.20, 63.70, 80.80, 95.90, 117.23, 120.60, 121.58, 122.60, 123.50, 125.67, 127.20, 129.07, 129.56, 130.45, 131.11, 135.68, 146.11, 147.04, 147.80, 148.50, 152.61, 156.86 ; $C_{28}H_{25}ClN_4O_2$: calcd. C 69.34, H 5.20, N 11.55; found C 69.29, H 5.17, N 11.64.

1,5,5-trimethyl-3-(2,5-dichlorophenyl)-10-phenylazo-3, 5a, 6, 11b-tetrahydro-*5H*- chromeno [4',3':4,5] pyrano [2,3-*c*]pyrazole (7n): Yellow crystals (1.42 g, 84%), mp 244-246˚C; ν_{max}/cm^{-1} 2957, 2926, 1490, 1245, 1022, 754; δ_H(400 MHz; $CDCl_3$) 1.50 (s, 3H, Me), 1.53 (s, 3H, Me), 2.20 (m, 4H, Me & C(5a)H), 4.13 (t, 1H, *J* = 11.4 Hz C(6)H), 4.32 (d, 1H, *J* = 4.4 Hz, C(11$_b$)H), 4.53 (dd, 1H, *J* = 4.0 Hz, C(6)H), 6.99 (d, 1H, *J* = 8.4 Hz, Ar-H), 7.28-8.01 (m, 10H, Ar-H); δ_C(100 MHz; $CDCl_3$) 15.10, 25.20, 25.98, 30.00, 38.90, 63.55, 81.10, 96.83, 117.30, 120.76, 121.45, 123.60, 123.90, 126.80, 127.94, 128.59, 129.10, 129.13, 129.94, 130.30, 133.71, 135.20, 145.52, 148.98, 152.62, 156.78 ; $C_{28}H_{24}Cl_2N_4O_2$: calcd. C 64.74, H 4.66, N 10.79; found C 64.89, H 4.50, N 10.60.

1,5,5-trimethyl-3-(2-chlorophenyl)-10-(4-chlorophenylazo)-3,5a,6,11b-tetrahydro-5*H*-chromeno [4',3':4,5] pyrano [2,3-*c*] pyrazole (7o): Yellow crystals (1.39 g, 79%), mp 118-120˚C; ν_{max}/cm^{-1} 3030, 2980, 1640, 1250, 850, 770, 690; δ_H(400 MHz; $CDCl_3$) 1.50 (s, 3H, Me), 1.55 (s, 3H, Me), 2.22 (m, 4H, Me & 5a-H), 4.10 (t, 1H, *J* = 11.0 Hz, C(6a)H), 4.32 (d, 1H, *J* = 4.0 Hz, C(11b)H), 4.50 (dd, 1H, *J* = 3.2 Hz, C(6)H), 6.96 (d, 1H, *J* = 8.4 Hz, Ar-H), 7.28-8.09 (m, 10H, Ar-H); δ_C(100 MHz; $CDCl_3$) 15.23, 25.43, 25.98, 29.70, 30.52, 38.33, 63.62, 80.67, 94.71, 117.37, 123.69, 123.89, 127.35, 127.48, 129.25, 129.62, 129.86, 130.20, 131.75, 136.18, 145.85, 147.89, 148.98, 151.17 ; $C_{28}H_{25}ClN_4O_2$: calcd. C 69.34, H 5.20, N 11.55; found C 69.20, H 5.28, N 11.67.

1,5,5-trimethyl-3-(2,5-dichlorophenyl)-10-(4-chlorophenylazo)-3,5a,6,11b-tetrahydro-5*H*-chromeno [4',3':4,5]pyrano[2,3-*c*]pyrazole (7p): Yellow crystals (1.45 g, 75%), mp 276-278˚C; ν_{max}/cm^{-1} 3025, 3000, 1635, 1220, 875, 780, 690; δ_H(400 MHz; $CDCl_3$) 1.52 (s, 3H, Me), 1.55 (s, 3H, Me), 2.23 (m, 4H, Me & C(5a)H), 4.07 (t, 1H, *J* = 11.2 Hz, C(6a)H), 4.31(d, 1H, *J* = 4.4 Hz, C(11b)H), 4.50 (dd, 1H, *J* = 4.0 Hz, C(6)H), 6.97 (d, 1H, *J* = 8.8 Hz, Ar-H), 7.35-8.08 (m, 9H, Ar-H); δ_C(100 MHz; $CDCl_3$) 15.19, 25.38, 25.97, 30.44, 38.27, 63.56, 81.22, 95.01, 117.41, 121.28, 123.78, 123.90, 127.24, 128.80, 129.28, 129.65, 129.94, 130.96, 132.98, 135.89, 136.23, 145.88, 148.52, 149.04, 151.11, 156.79 ; $C_{28}H_{23}Cl_3N_4O_2$: calcd. C 60.72, H 4.19, N 10.12; found C 60.50, H 4.10, N 10.30.

(4a*R*,10b*S*)-9-[(E)-phenyldiazenyl]-1,4a,5,10b-tetrahydro-2*H*,4*H*-pyrano[3,4-*c*]chromen-2-one(9a): Yellow crystals (0.91 g, 78%), mp 195-196˚C; Yellow crystals (0.77 g, 73%), mp 212-214˚C; IR (KBr): ν = 3050, 2990, 1670, 1600, 1380, 1250, 1000, 840, 650; ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 2.58 (m, 2H, C(6)H), 2.98 (dd, *J* = 7.2 Hz, 1H, C(1)H), 3.62 (q, 1H, C(10b)H), 4.37 (m, 2H, C(4)H), 4.12 (m, 2H, C(5)H), 6.91 (d, *J* = 8.4 Hz, 1H, Ar-H), 7.51-7.90 (m, 7H, Ar-H); ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 29.93, 31.55, 35.71, 65.24, 67.87, 118.31, 123.29, 123.93, 124.10, 129.34; MS: calcd. for $C_{18}H_{16}N_2O_3$ [M + H] 309.1; found 309.20; $C_{18}H_{16}N_2O_3$ (309.1 g/mol): calcd. C 70.12, H 5.23, N 9.09; found C 70.20, H 5.20, N 9.12.

5-[(*E*)-phenyldiazenyl]-1,6b,14,14a-tetrahydro-7*H*,14*H*-cromeno[3',4':5,6]pyrano[3,4-*c*]-chromen-7-one (9b): Yellow crystals (0.91 g, 70%), mp 210-212˚C; IR (KBr): ν = 3050, 2970, 1620, 1465, 1380, 1250, 1130, 750; ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 2.52 (m, 1H, C(6)H), 4.38 (t, 1H, C(14a)H), 4.56 (m, 4H, Me & C(14)H), 6.92-8.15 (m, 12H, Ar-H); ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 29.22, 29.70, 66.07, 103.31, 115.29, 116.73, 117.81, 120.53, 121.49, 122.72, 122.99, 124.03, 127.39, 128.51, 128.94, 130.38, 132.21, 147.73, 152.70, 154.38, 160.21, 164.14; MS: calcd. for $C_{25}H_{18}N_2O_4$ [M + H] 411.1; found 411.30; $C_{25}H_{18}N_2O_4$ (410.1 g/mol): calcd. C 73.16, H 4.42, N 6.83; found C 73.25, H 4.34, N 6.70.

(6a*R*,12b*S*)-14-methyl-11-[(*E*)-(4-chlorophenyl)phenyldiazenyl]-6a,7,12b,14-tetrahydro-6*H*,13*H*-chromeno[4',3':4,5]pyrano[3,2-*c*]quinolin-13-one(9c): Yellow crystals (1.30 g, 71%), mp 240-241˚C;

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δ_H(400 MHz; $CDCl_3$) 2.51 (m, 1H, C(14b)H), 3.87 (s, 3H, N-CH3), 4.32 (m, 1H, C(6b)H), 4.56 (m, 4H, 1 & 14-H), 6.89-8.10 (m, 12H, Ar-H); δ_C(100 MHz; $CDCl_3$) 29.31, 29.70, 29.92, 65.36, 66.45, 108.70, 113.99, 115.74, 117.66, 119.74, 121.83, 123.32, 123.88, 124.00, 129.11, 129.81, 130.99, 135.94, 139.09, 147.42, 151.12, 154.89, 156.40, 164.01**;** $C_{25}H_{18}N_2O_4$: calcd. C 68.20, H 4.40, N 9.18; found C 68.48, H 4.36, N 9.10.

(6a*R*,12b*S*)-11-[(E)-(4-chlorophenyl)diazenyl]-14-phenyl-6a,7,12b,14-tetrahydro-6*H*,13*H*-chromeno[4',3':4,5]pyrano[3,2-*c*]quinolin-13-one (9d): Yellow crystals (0.67 g, 76%), mp 220-223˚C; IR (KBr): ν = 3050, 2980, 1730, 1490, 1240, 1100, 990, 790, 680; ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 2.54 (m, 1H, C(14b)H), 4.40 (m, 1H, C(6b)H), 4.49 (m, 4H, Me & C(14)H), 6.89-8.10 (m, 16H, Ar-H); ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 29.33, 29.74, 29.92, 64.36, 66.45, 108.54, 113.80, 115.68, 117.24, 119.56, 121.90, 123.48, 123.62, 124.48, 129.24, 129.61, 130.67, 135.36, 139.38, 147.42, 151.46, 154.17, 156.12, 164.51; MS: calcd. for $C_{31}H_{22}ClN_3O_3$ [M + H] 520.1; found 520.37; $C_{31}H_{22}ClN_3O_3$ (520.1 g/mol): calcd. C 71.61, H 4.26, N 8.08; found C 71.48, H 4.36, N 8.10.

(4aR,10bS)-9-[(E)-(4-chlorophenyl)-phenyldiazenyl]-1,4a,5,10b-tetrahydro-2H,4H-pyrano[3,4-c]chromen-2-one (9e): Yellow crystals (0.77 g, 73%), mp 212-214˚C; IR (KBr): ν = 3050, 2975, 1760, 1450, 1270, 1080, 790, 630, 550; ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 2.66 (m, 2H, C(6)H), 3.12 (dd, J = 7.2 Hz, 1H, C(1)H), 3.55 (q, 1H, C(10b)H), 4.13 (m, 2H, C(4)H), 4.54 (m, 2H, C(5)H), 7.01 (d, J = 8.4 Hz, 1H, Ar-H), 7.48-7.86 (m, 6H, Ar-H); ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 29.93, 31.55, 35.71, 65.24, 67.87, 118.31, 123.29, 123.93, 124.10, 129.34; MS: calcd. for $C_{25}H_{17}ClN_2O_4$ [M + H] 445.2; found 445.4; $C_{25}H_{17}ClN_2O_4$ (444.2 g/mol): calcd. C 67.50, H 3.85, N 6.30; found C 67.29, H 3.88, N 6.37.

5-[(*E*)-(4-chlorophenyl)phenyldiazenyl]-1,6b,14,14a-tetrahydro-7*H*,14*H*-cromeno[3',4':5,6]pyrano[3,4-*c*]-chromen-7-one (9f): Yellow crystals (0.91 g, 78%), mp 210-212˚C; IR (KBr): ν = 3050, 2975, 1760, 1450, 1270, 1080,790, 630, 550; ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 2.55 (m, 1H, C(6)H), 4.30 (t, 1H, C(14a)H), 4.52 (m, 4H, Me & C(14)H), 6.90-8.20 (m, 11H, Ar-H); ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 29.20, 29.74, 66.37, 103.20, 115.11, 116.65, 117.58, 120.37, 121.65, 122.88, 123.00, 124.14 127.42, 128.48, 128.86, 130.41, 132.44, 147.64, 152.55, 154.61, 160.43, 164.45; MS: calcd. for $C_{25}H_{17}ClN_2O_4$ [M + H] 445.1; found 445.37; $C_{25}H_{17}ClN_2O_4$ (444.1 g/mol): calcd. C 67.50, H 3.85, N 6.30; found C 67.29, H 3.70, N 6.50.

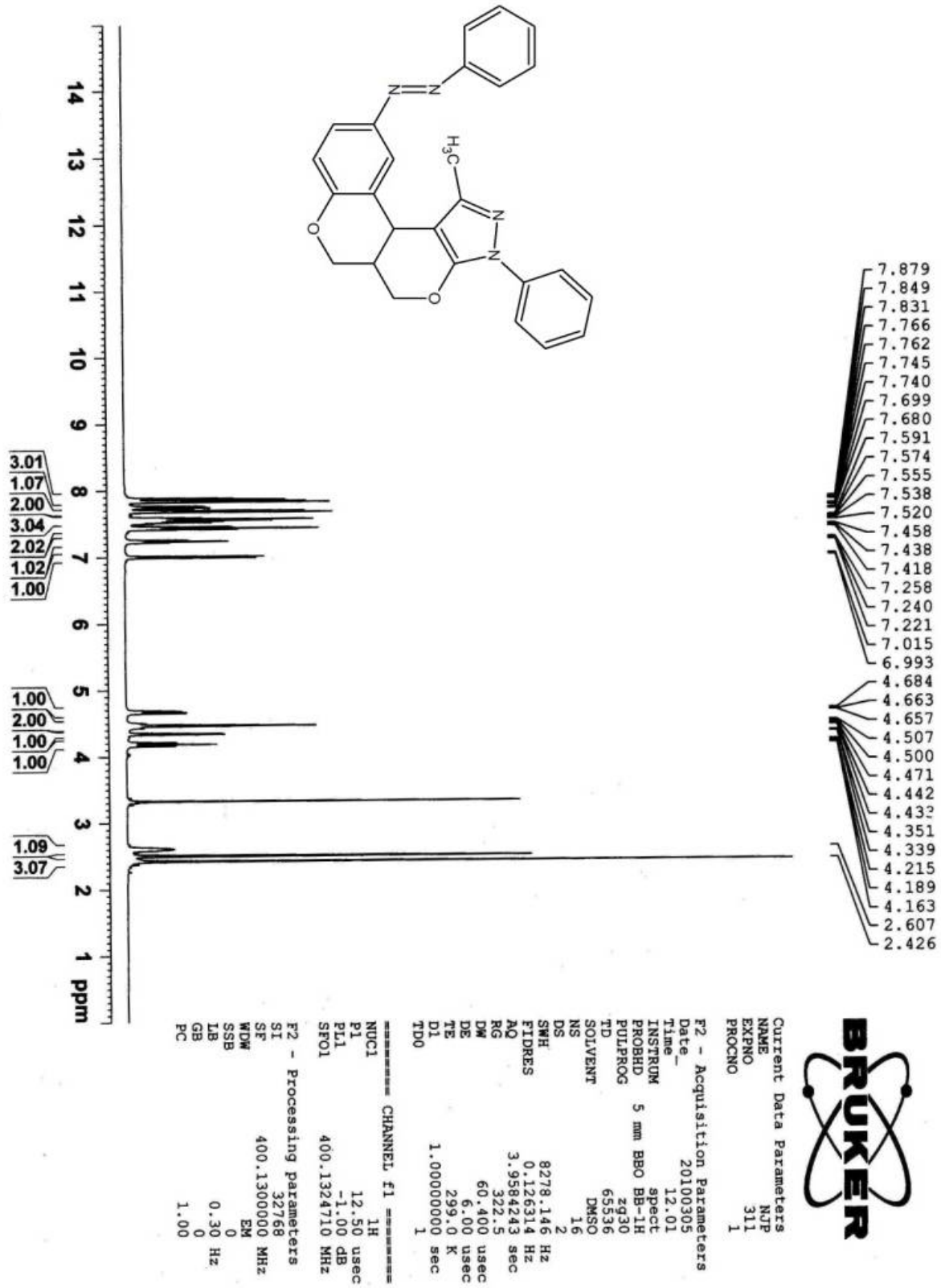
(6a*R*,12b*S*)-14-methyl-11-[(*E*)-phenyldiazenyl]-6a,7,12b,14-tetrahydro-6*H*,13*H*-chromeno[4',3':4,5]pyrano[3,2-*c*]quinolin-13-one(9g): Yellow crystals (0.75 g, 77%), mp 220-222˚C; IR (KBr): ν = 3350, 2980, 1620, 1490, 1365, 1270, 1080, 770, 570; ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 2.55 (m, 1H, C(14b)H), 3.86 (s, 3H, N-CH_3), 4.29 (m, 1H, C(6b)H), 4.60 (m, 4H, Me & C(14)H), 6.91-8.20 (m, 11H, Ar-H); ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 29.40, 30.00, 30.10, 65.40, 66.05, 108.75, 114.09, 115.34, 118.46, 118.14, 121.90, 123.12, 123.80, 124.10, 129.20, 129.01, 130.80, 136.00, 139.29, 147.60, 151.34, 154.93, 156.56, 164.35; MS: calcd. for $C_{26}H_{21}N_3O_3$ [M + H] 424.2; found 424.50; $C_{25}H_{18}N_2O_4$ (423.2 g/mol): calcd. C 73.74, H 5.00, N 9.92; found C 73.48, H 4.96, N 9.70.

(6aR,12bS)-14-phenyl-11-[(E)-phenyldiazenyl]-6a,7,12b,14-tetrahydro-6H,13H-chromeno[4',3':4,5]pyrano[3,2-c]quinolin-13-one (9h): Yellow crystals (0.69 g, 72%), mp 230-231˚C; IR (KBr): ν = 3025, 2930, 1650, 1430, 1240, 1090, 830, 750; ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 2.80 (m, 1H, C(14b)H), 4.31 (m, 1H, C(6b)H), 4.33 (m, 4H, Me & C(14)H), 6.32-8.47 (m, 17H, Ar-H); ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 30.11, 30.70, 30.92, 66.36, 66.45, 109.70, 113.04, 116.74, 117.59, 119.27, 121.28, 123.83, 123.99, 124.34, 129.67, 130.01, 132.04, 135.94, 139.42, 147.41, 151.12, 154.94, 156.62, 164.38; MS: calcd. for $C_{31}H_{23}N_3O_3$ [M + H] 486.2; found 486.23; $C_{31}H_{23}N_3O_3$ (485.2 g/mol): calcd. C 76,69, H 4.77, N 8.65; found C 76.58, H 4.65, N 8.78.

(5a*R*,11b*S*)-1-methyl-3-phenyl-3,5a,6,11b-tetrahydro-5*H*-chromeno [4',3':4,5]pyrano[2,3-*c*]pyrazol-10-amine (10): Isolated yield (0.52 g, 45%) as white powder, mp 230-234˚C; ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 2.39 (s, 3H, Me), 2.60 (s, 1H, C(5a)H), 4.04 (m, 2H, C(6)H & C(11b)H), 4.22 (s, 2H, C(5)H), 4.58 (dd, J = 10.8 Hz, 1H, C(6)H), 4.72 (s, 2H, NH_2), 6.36 (d, J = 8.4 Hz, 1H, ArH), 6.46-7.69 (m, 7H ArH); ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 14.35, 29.57, 31.03, 65.55, 68.79, 100.24, 114.67, 115.46, 117.19, 120.16, 124.71,

125.80, 129.53, 138.79, 142.72, 143.60, 147.05, 149.33; MS: calcd. for $C_{20}H_{19}N_3O_2$ [M + H] 334.3; found 334.43; $C_{20}H_{19}N_3O_2$ (333.3 g/mol): calcd. C 72.05, H 5.74, N 12.60; found C 72.15, H 5.65, N 12.78.

Supplementary data NMR
Compound 7a ^{1}H NMR



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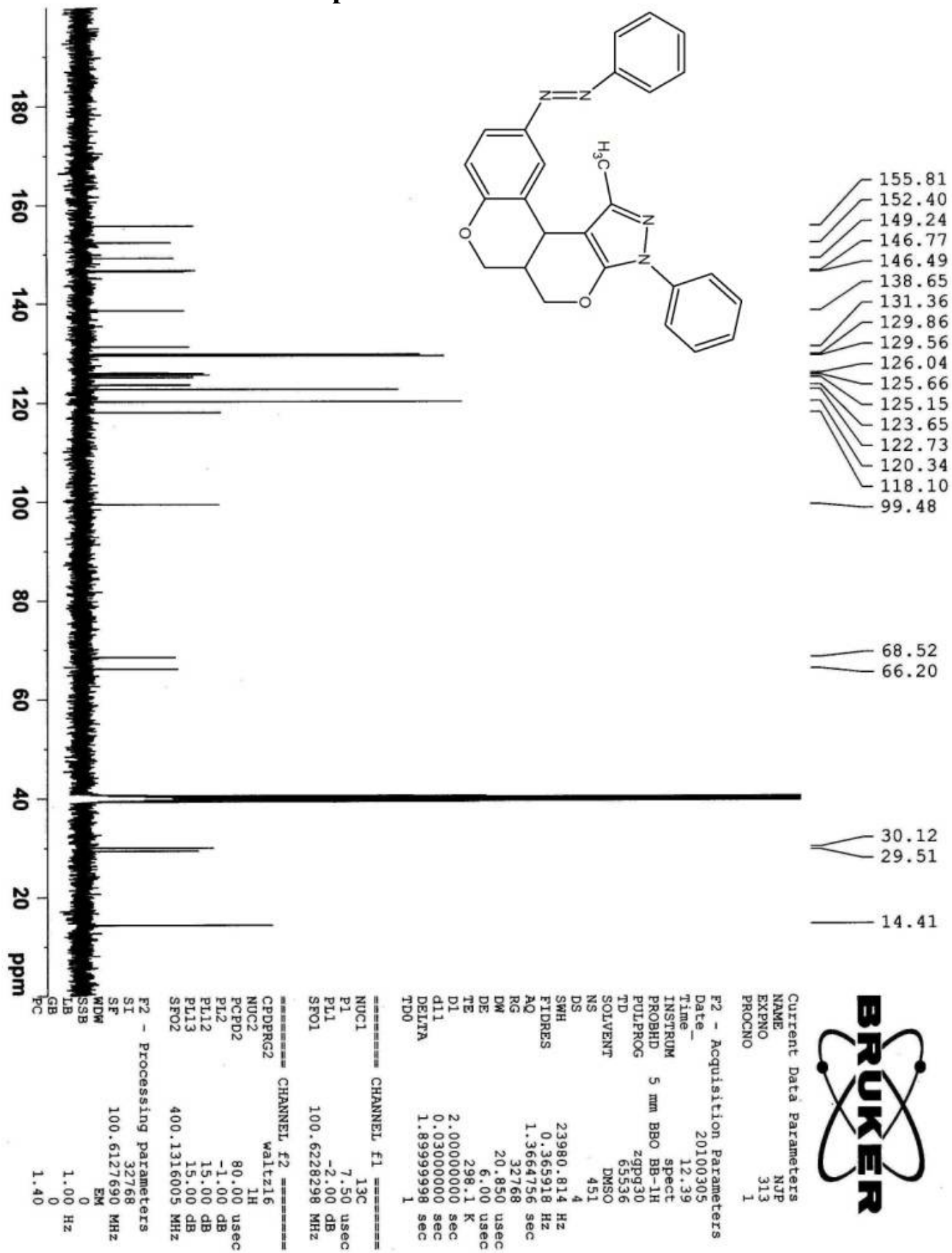
Compound 7a - ^{13}C NMR



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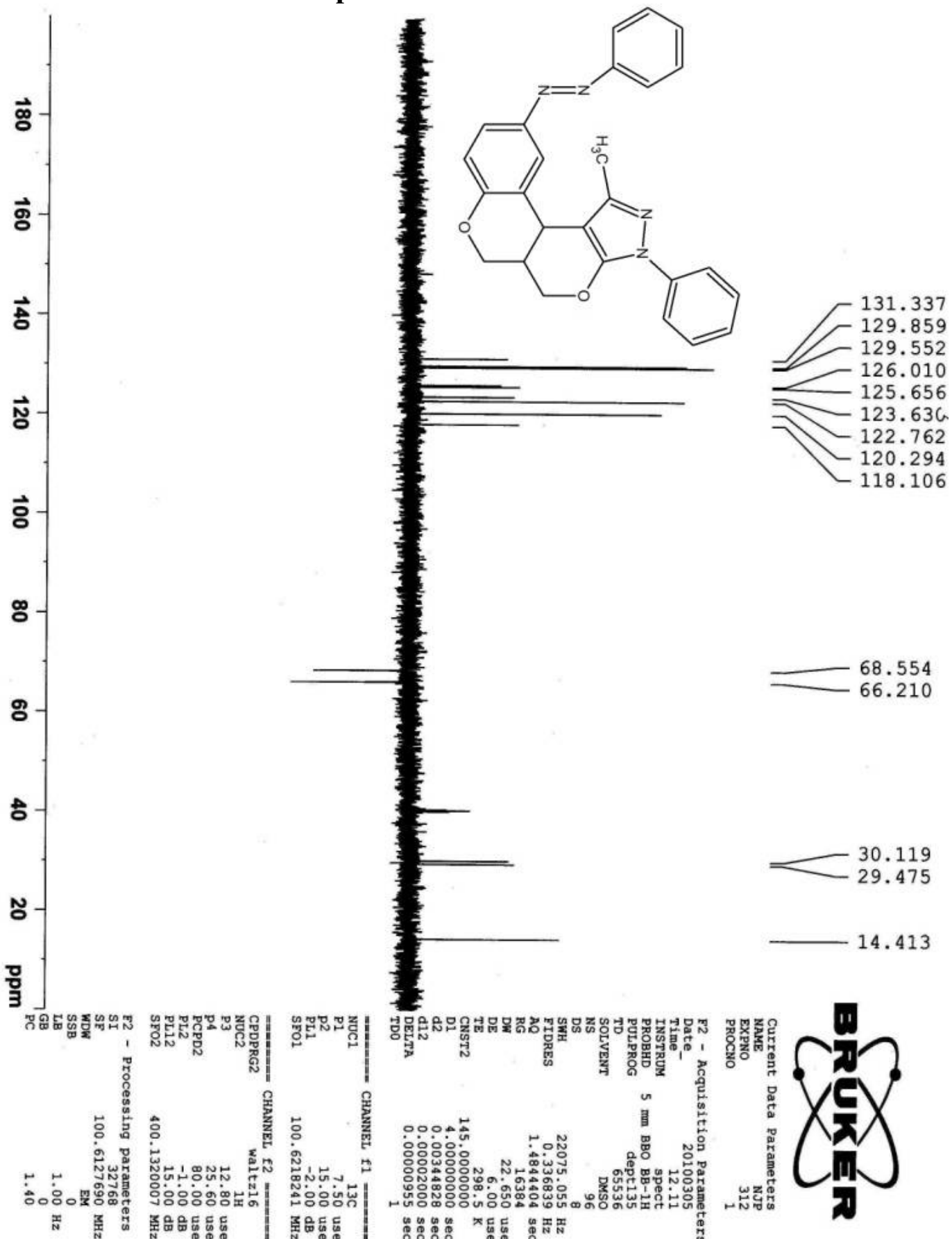
Compound 7a – DEPT 135



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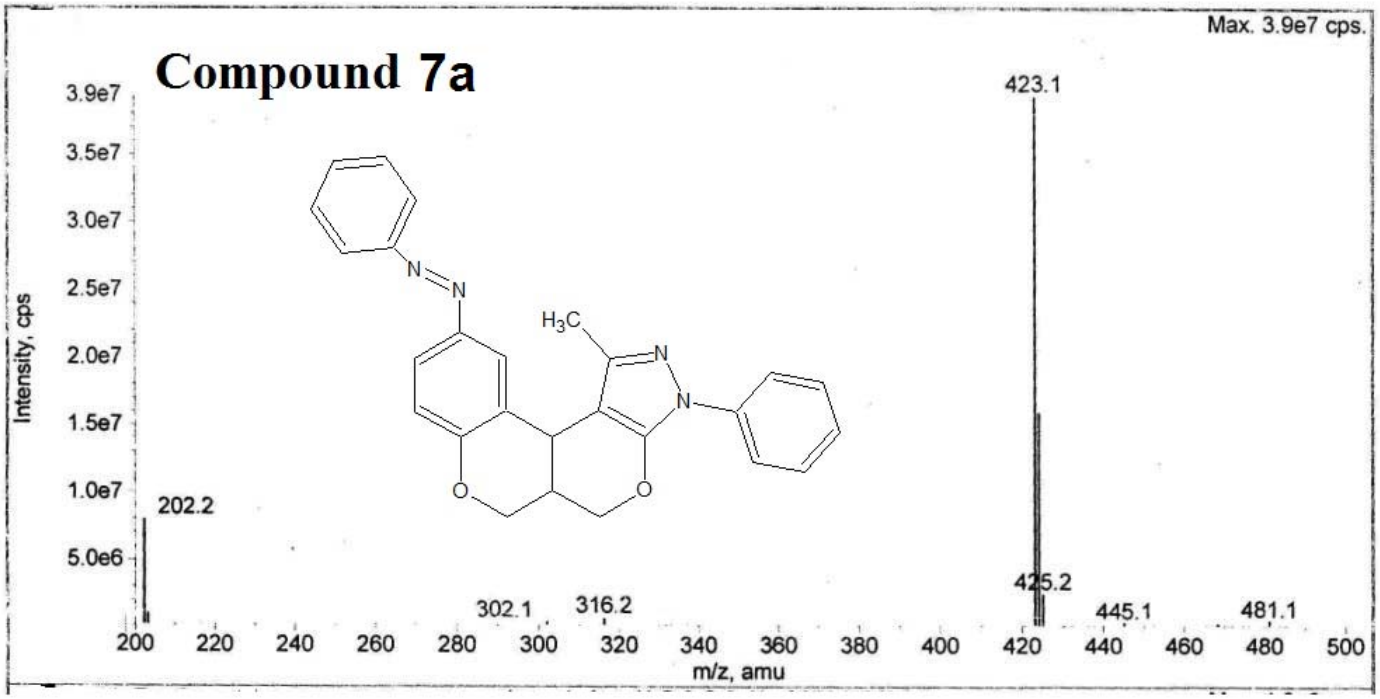
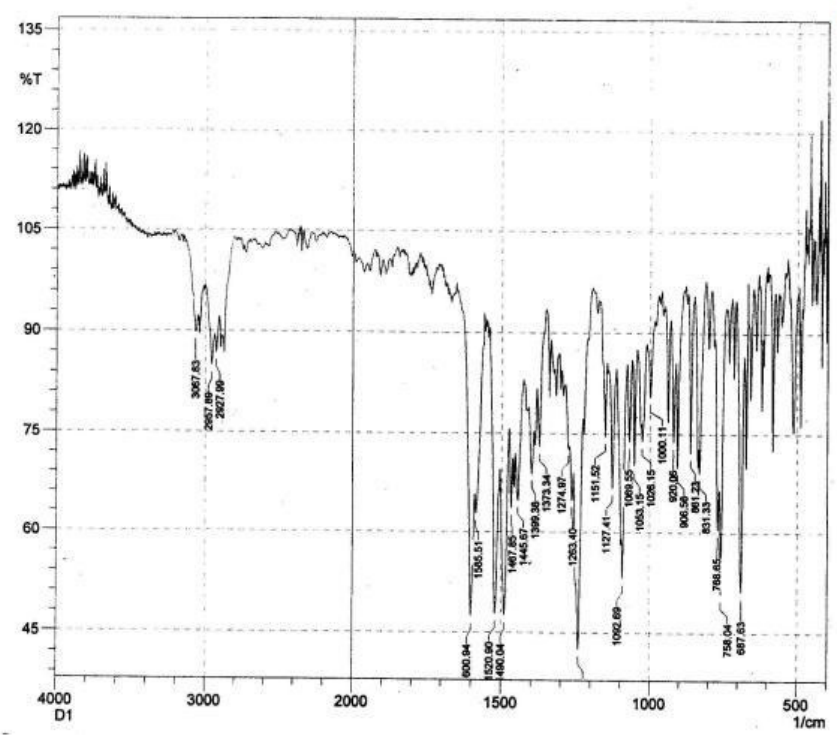
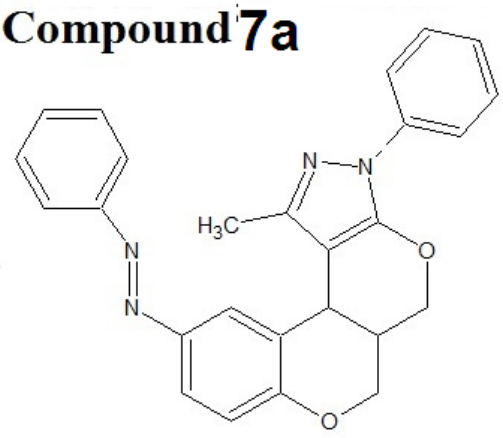
Compound 7a mass



Compound 7a IR





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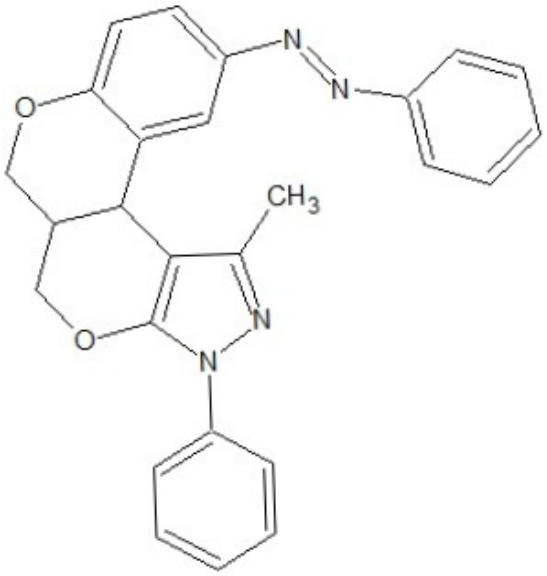
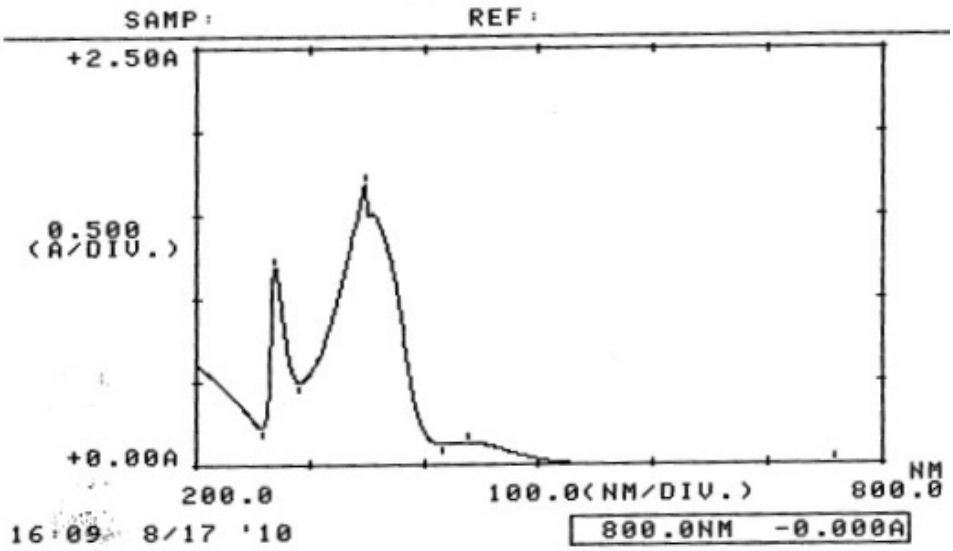
Compound 7a UV



λ	ABS	λ	ABS	λ	ABS
758.0	0.002	598.0	-0.003		
438.0	0.134	416.0	0.123		
348.0	1.680	290.0	0.494		
270.0	1.190	258.0	0.227		



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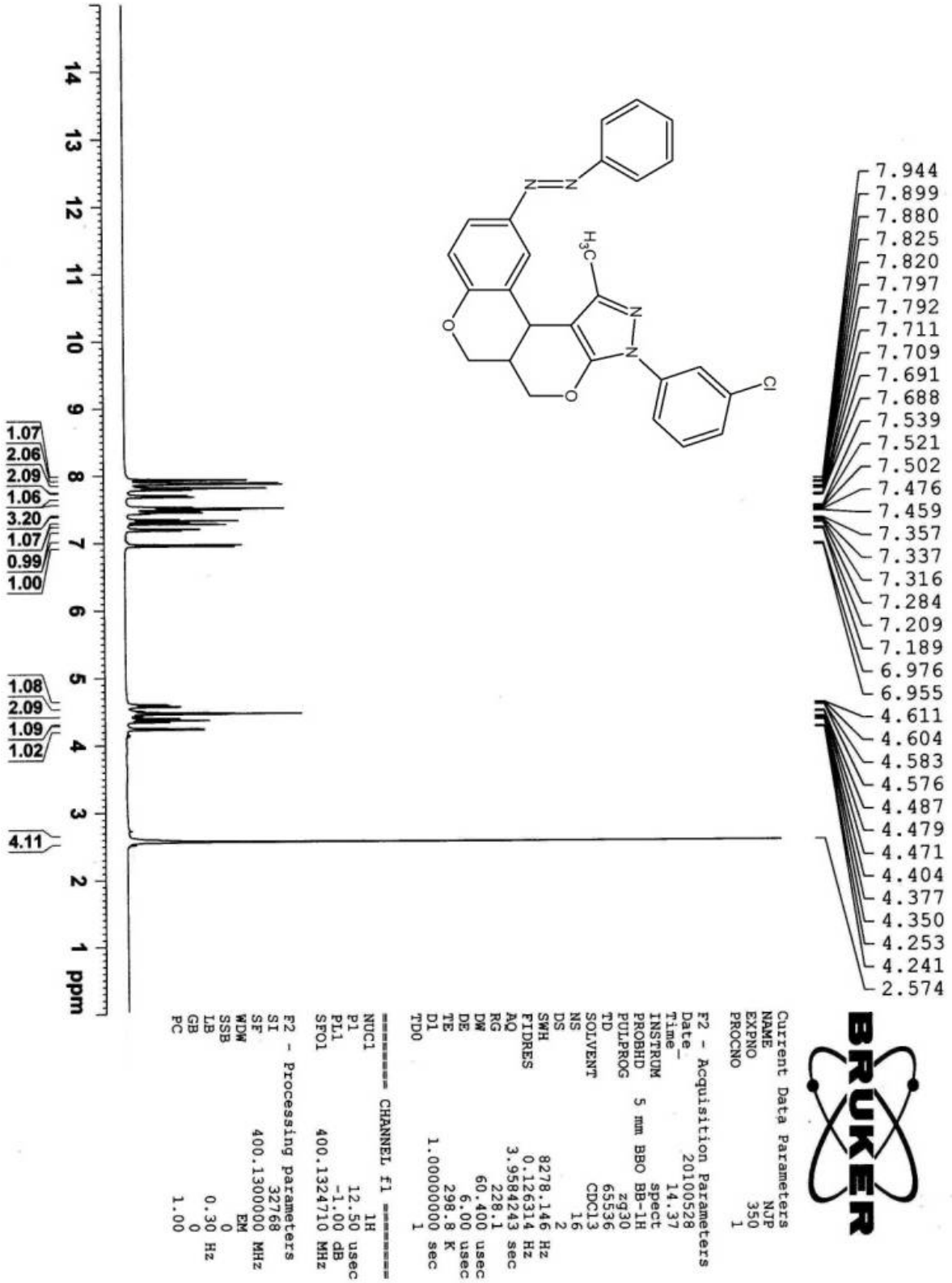
Compound 7b
Compound 7b ^{1}H NMR



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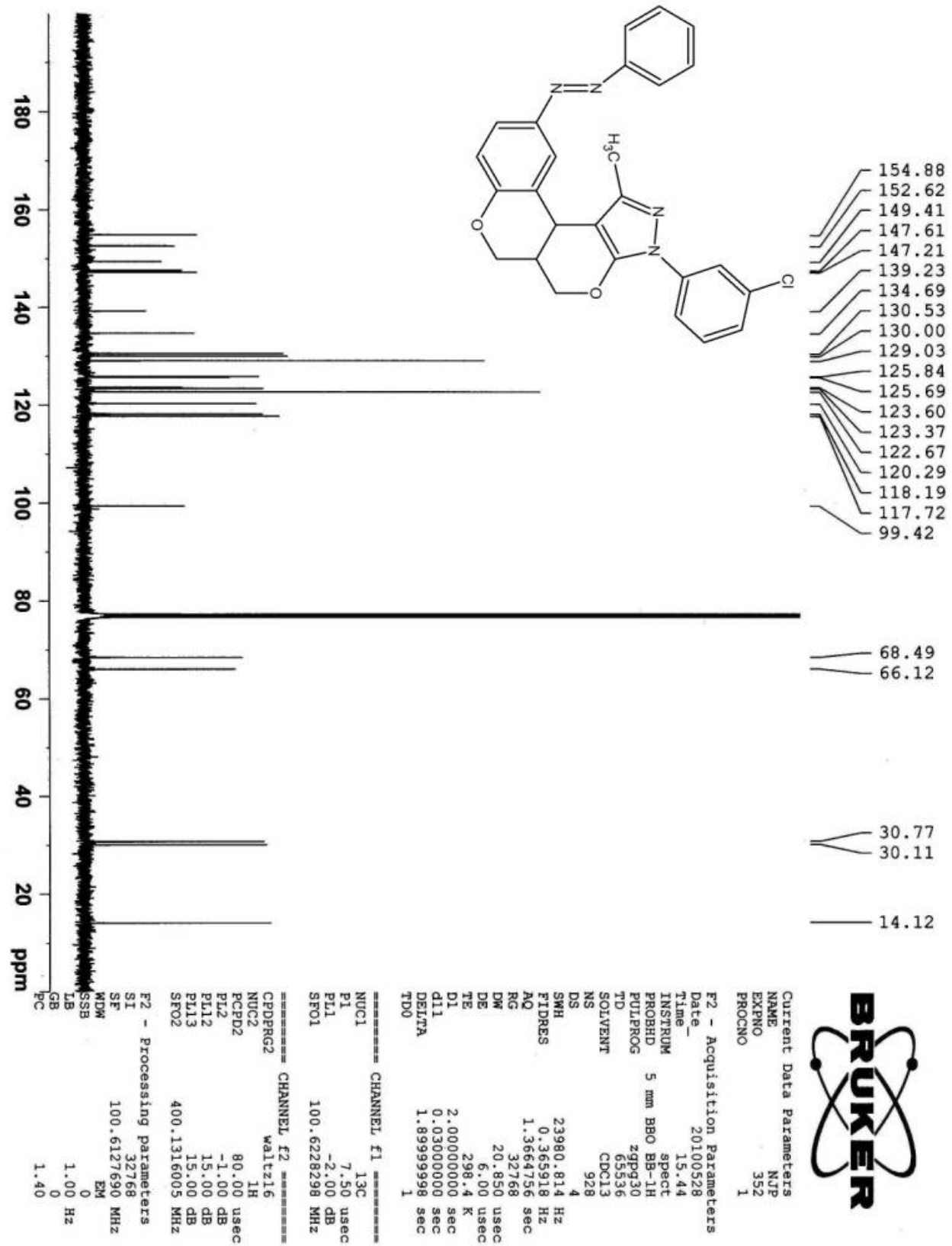
Compound 7b - ^{13}C NMR



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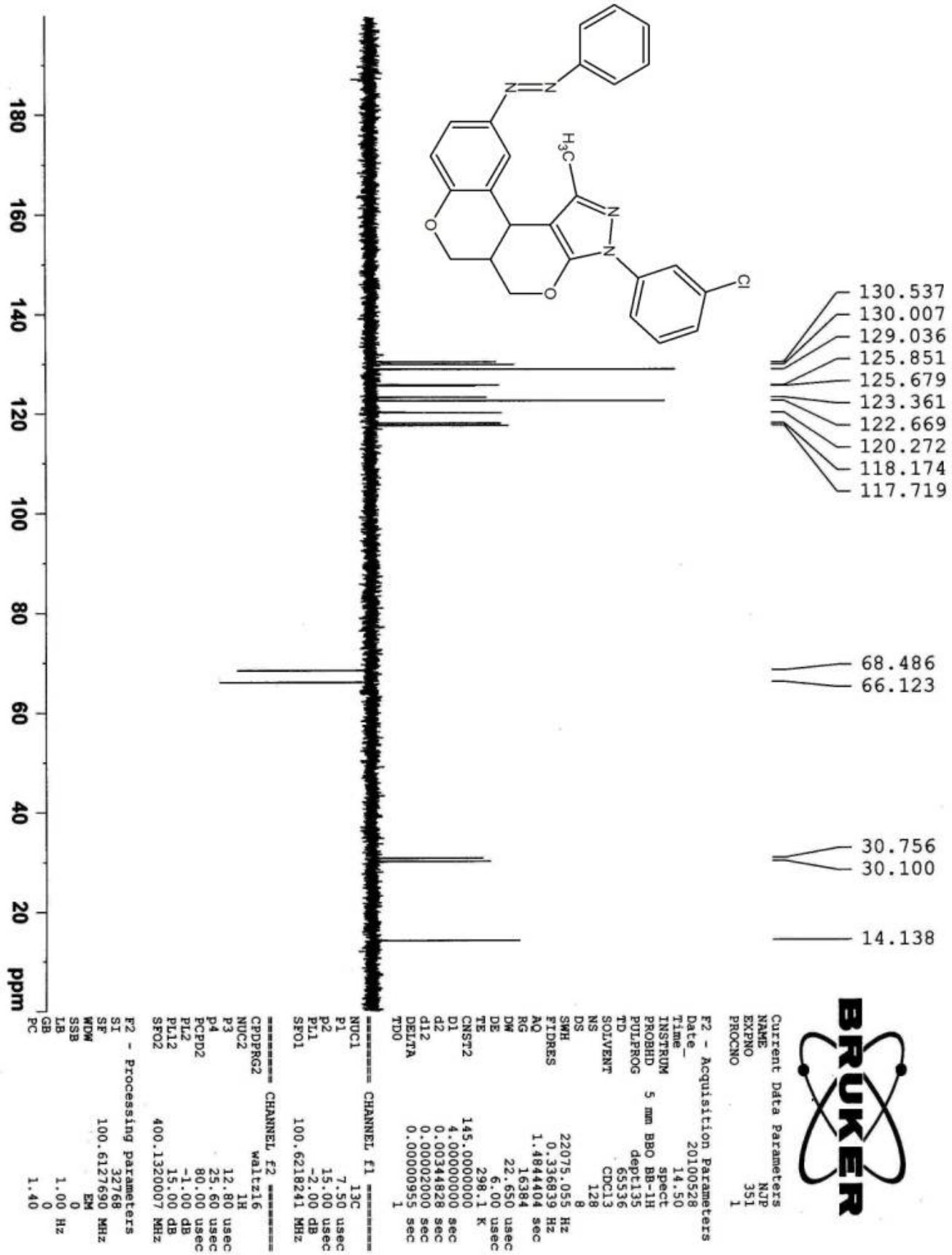
Compound 7b – DEPT 135



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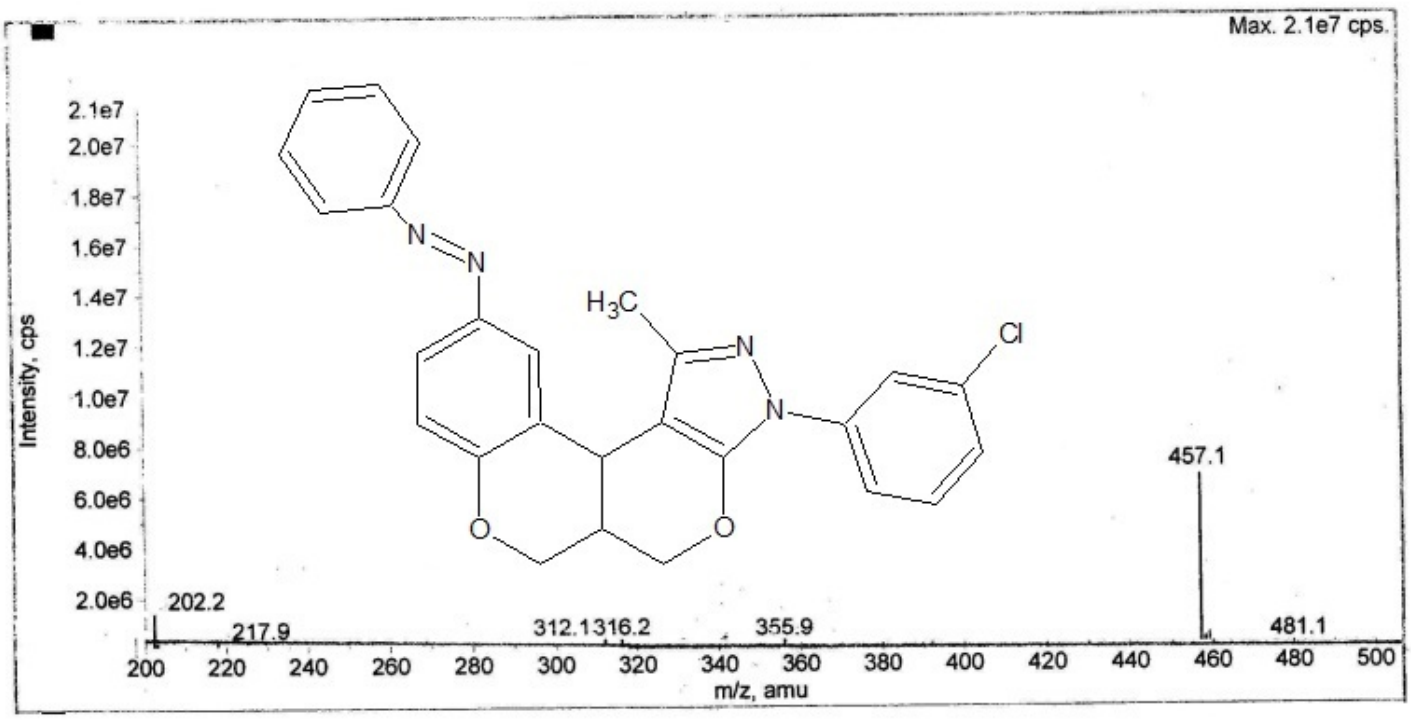
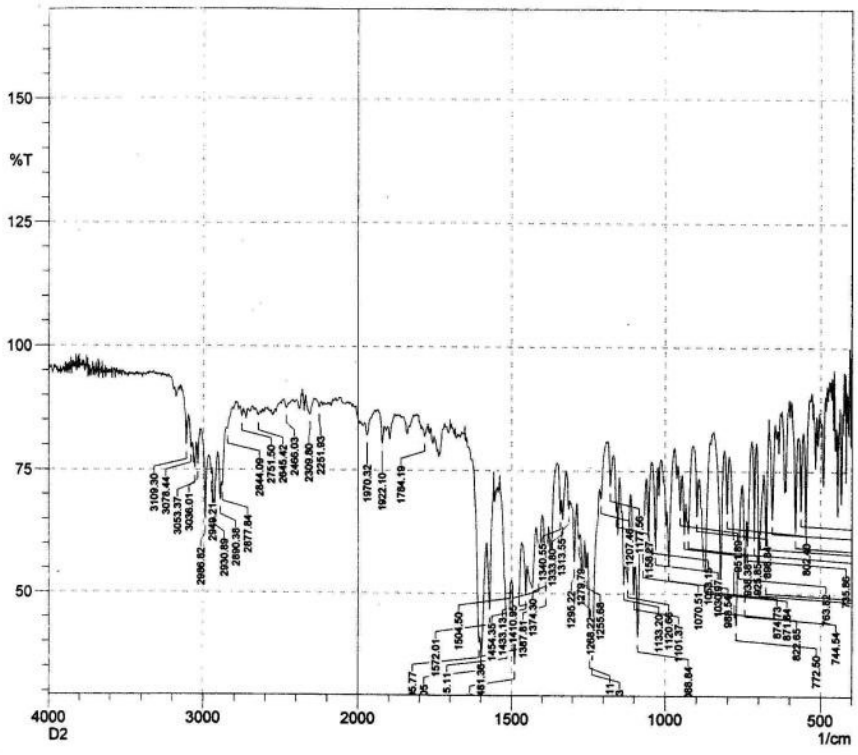
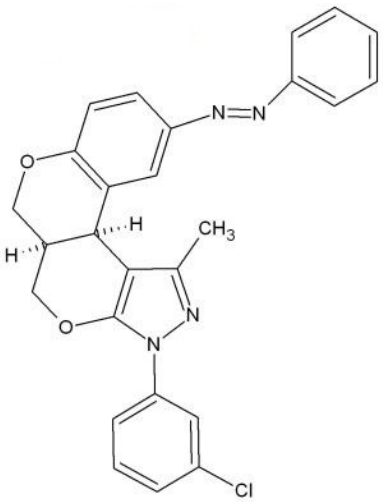
Compound 7b mass



Compound 7b IR





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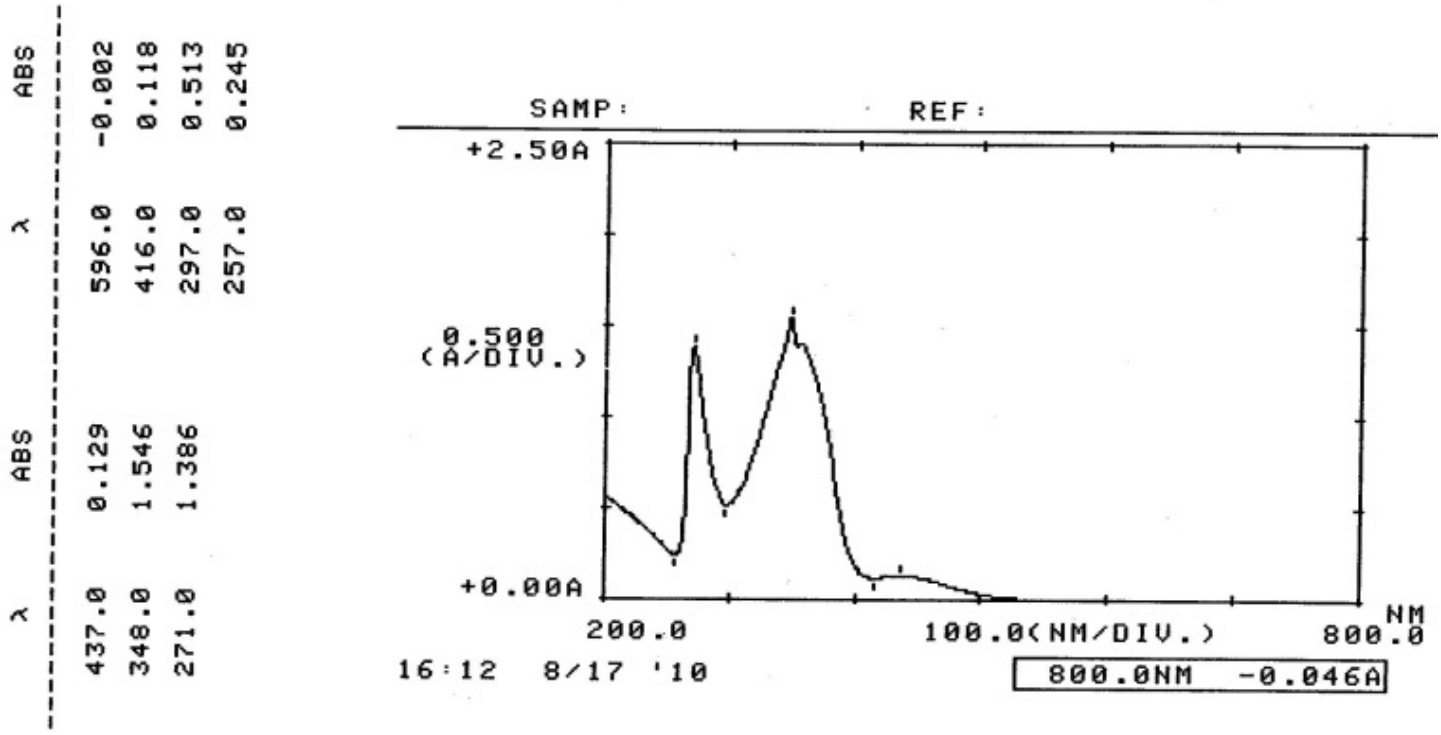
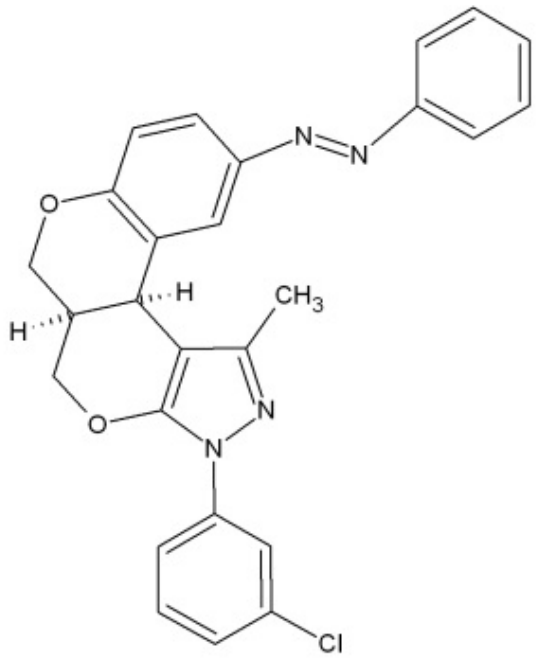
Compound 7b UV





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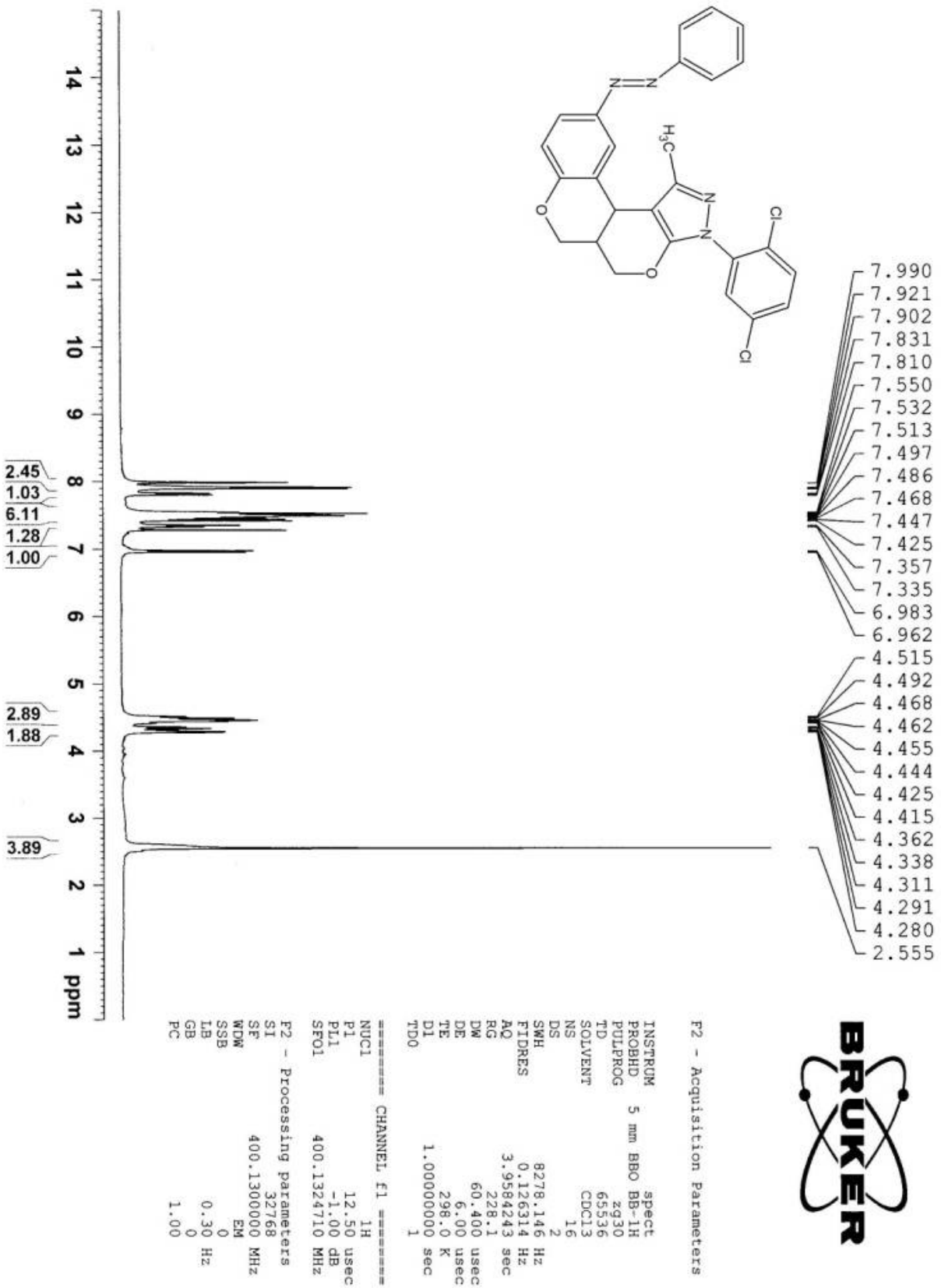
Compound 7d
Compound 7d ^{1}H NMR



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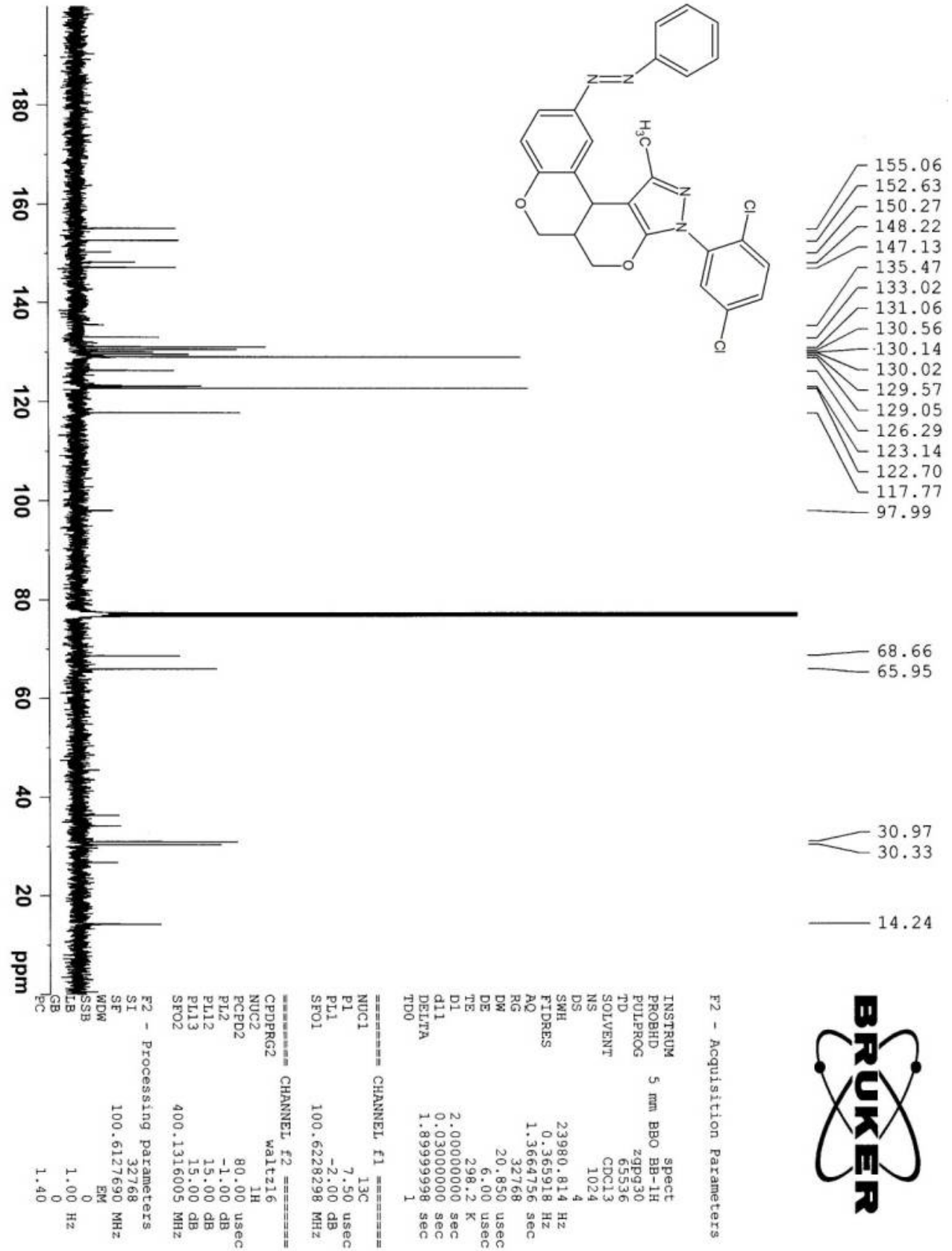
Compound 7d - ^{13}C NMR



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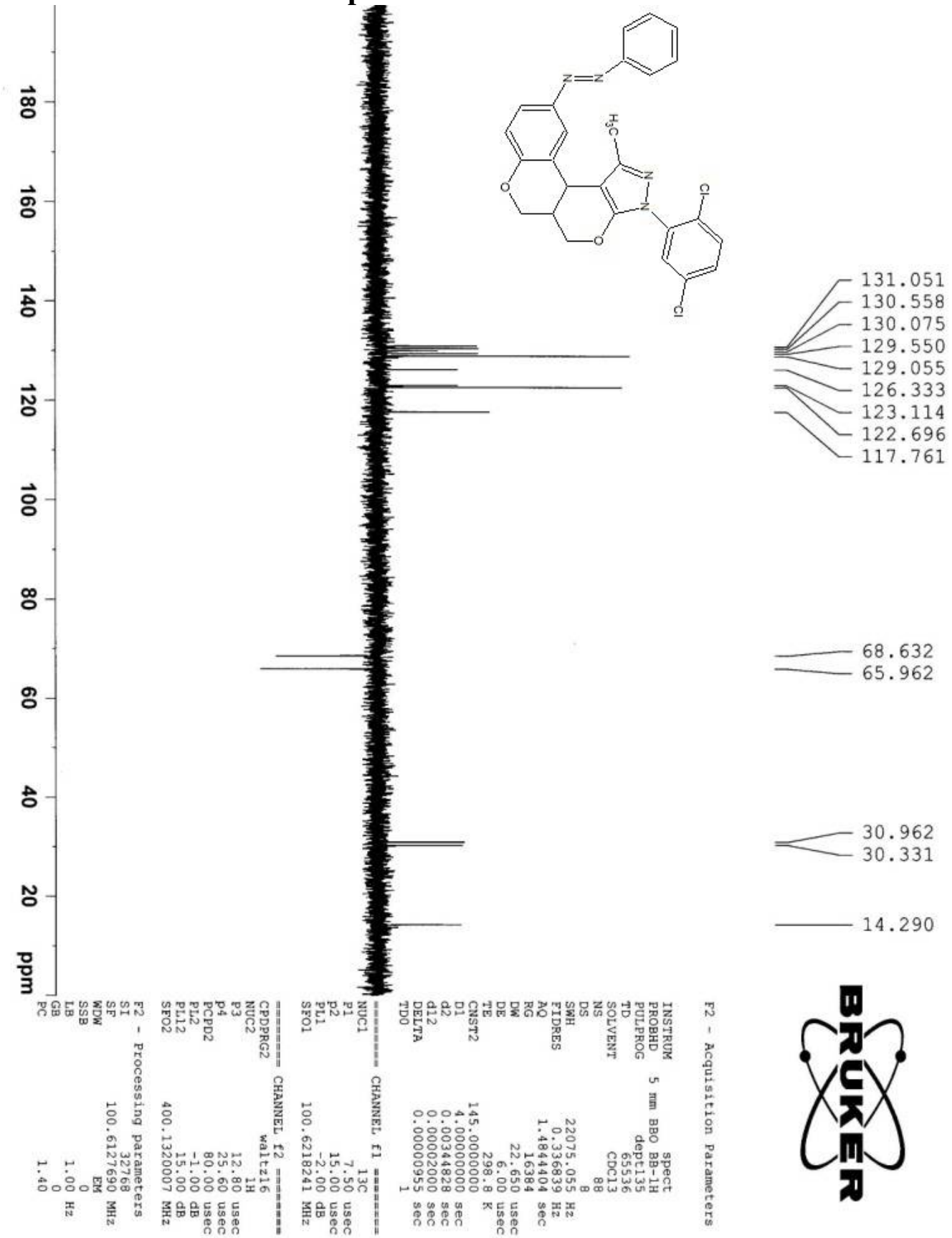
Compound 7d – DEPT 135



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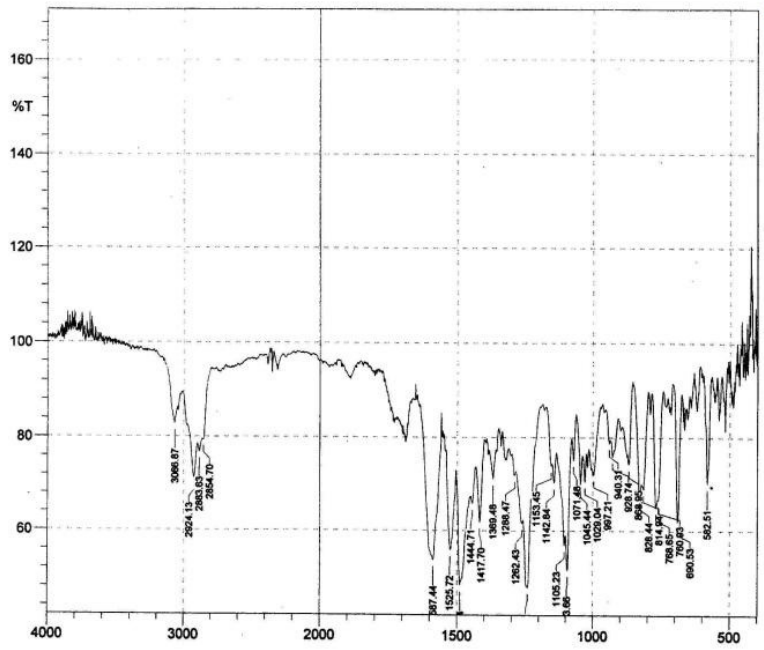
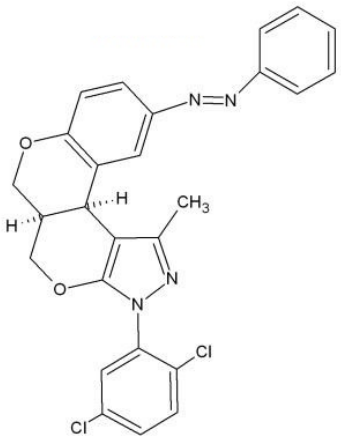
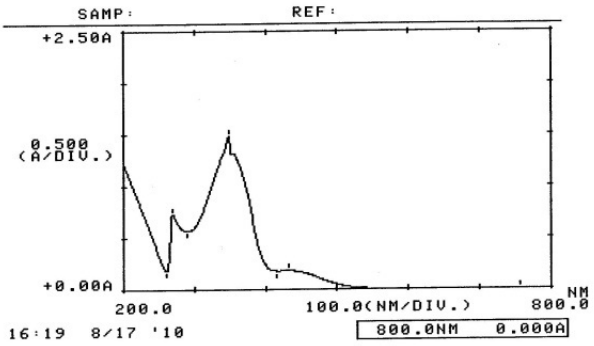
Compound 7d IR

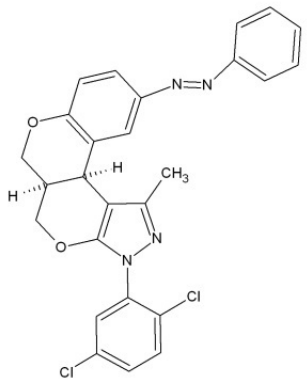




Compound 7d UV

	ABS	λ	ABS	λ
	-0.001	789.0	0.002	757.0
	-0.001	600.0	0.182	433.0
	0.178	416.0	1.492	348.0
	0.567	291.0	0.731	270.0
	0.181	261.0		





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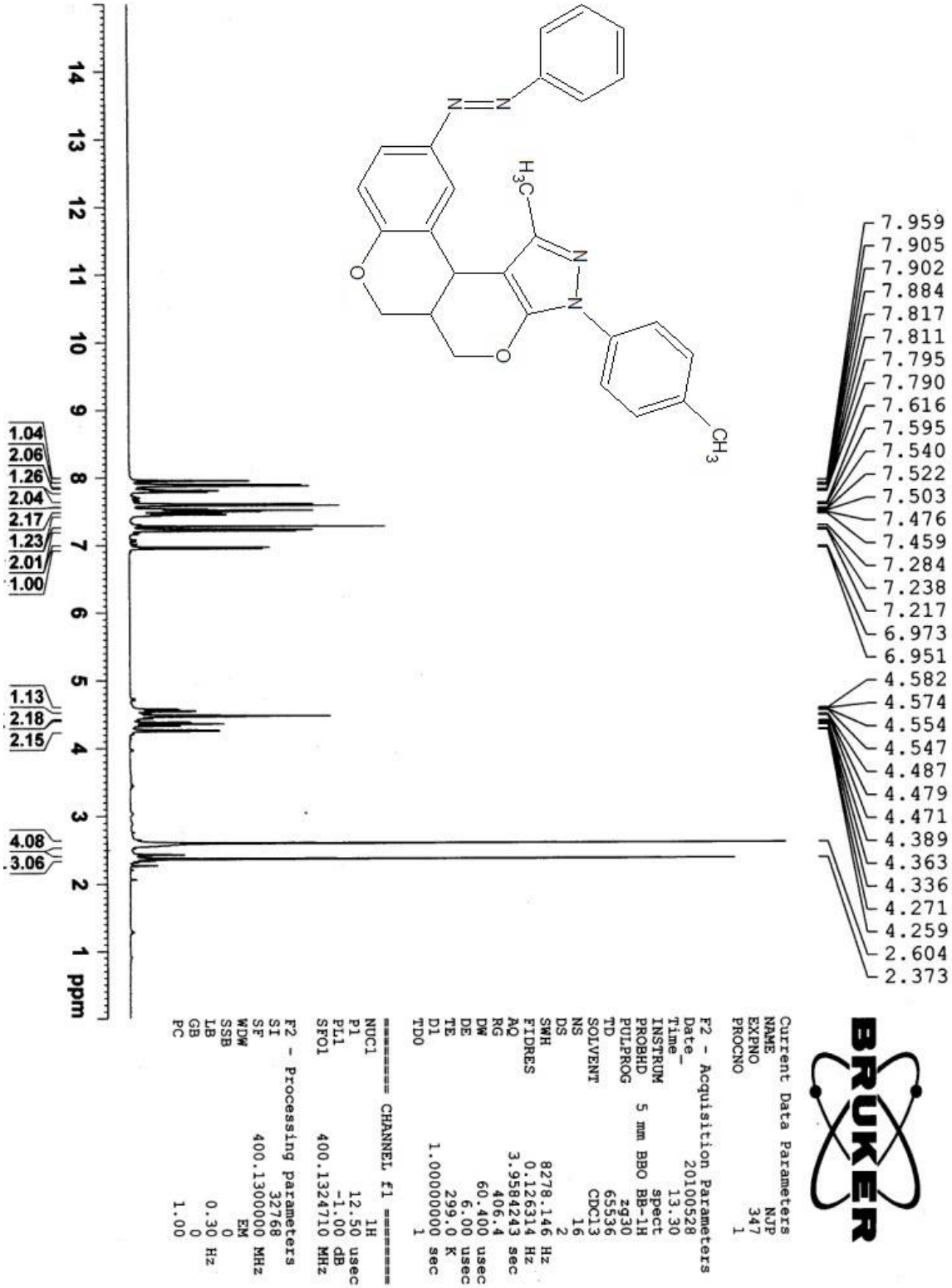
Compound 7e

Compound 7e ^{1}H NMR



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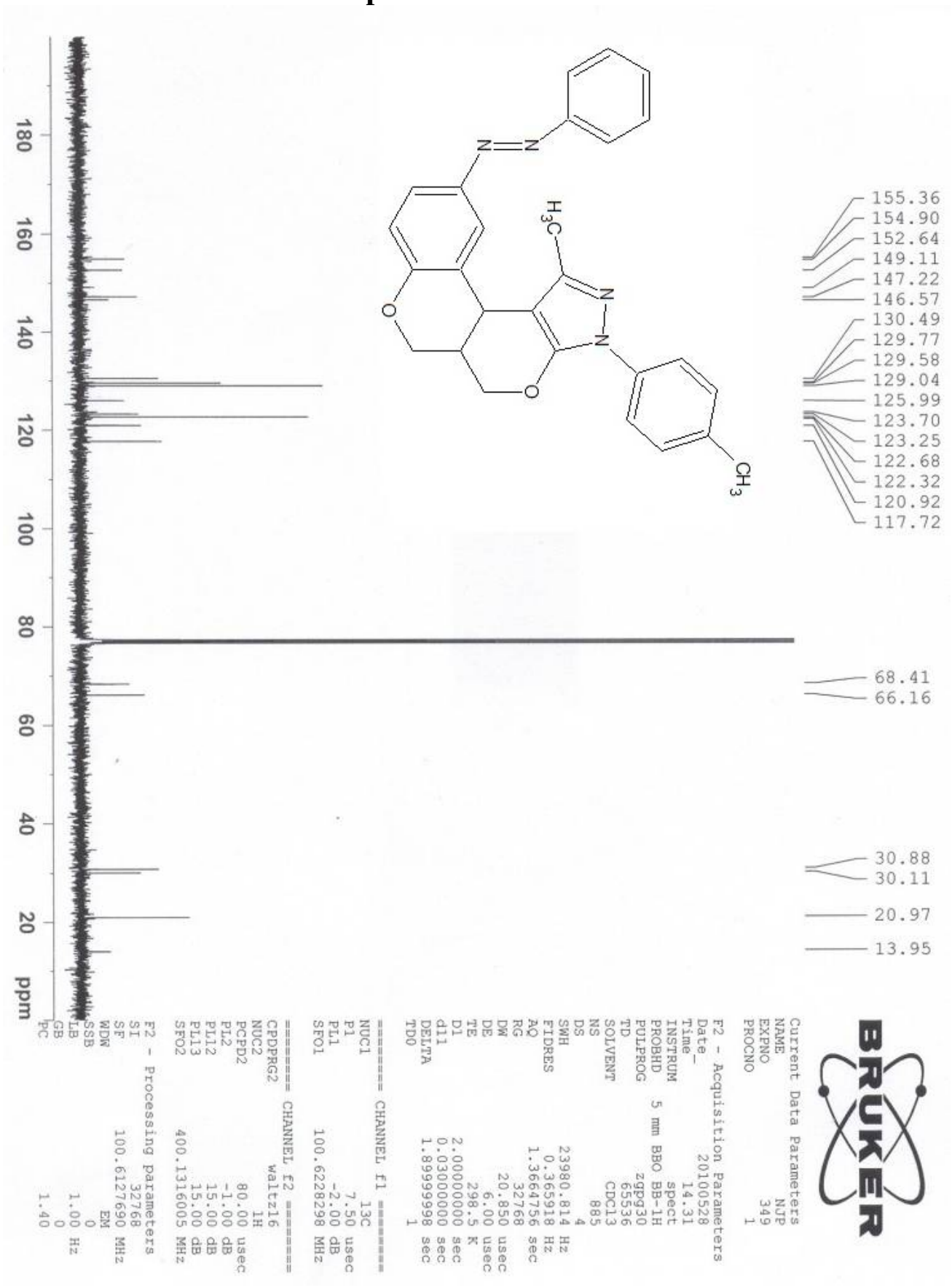
Compound 7e - ^{13}C NMR



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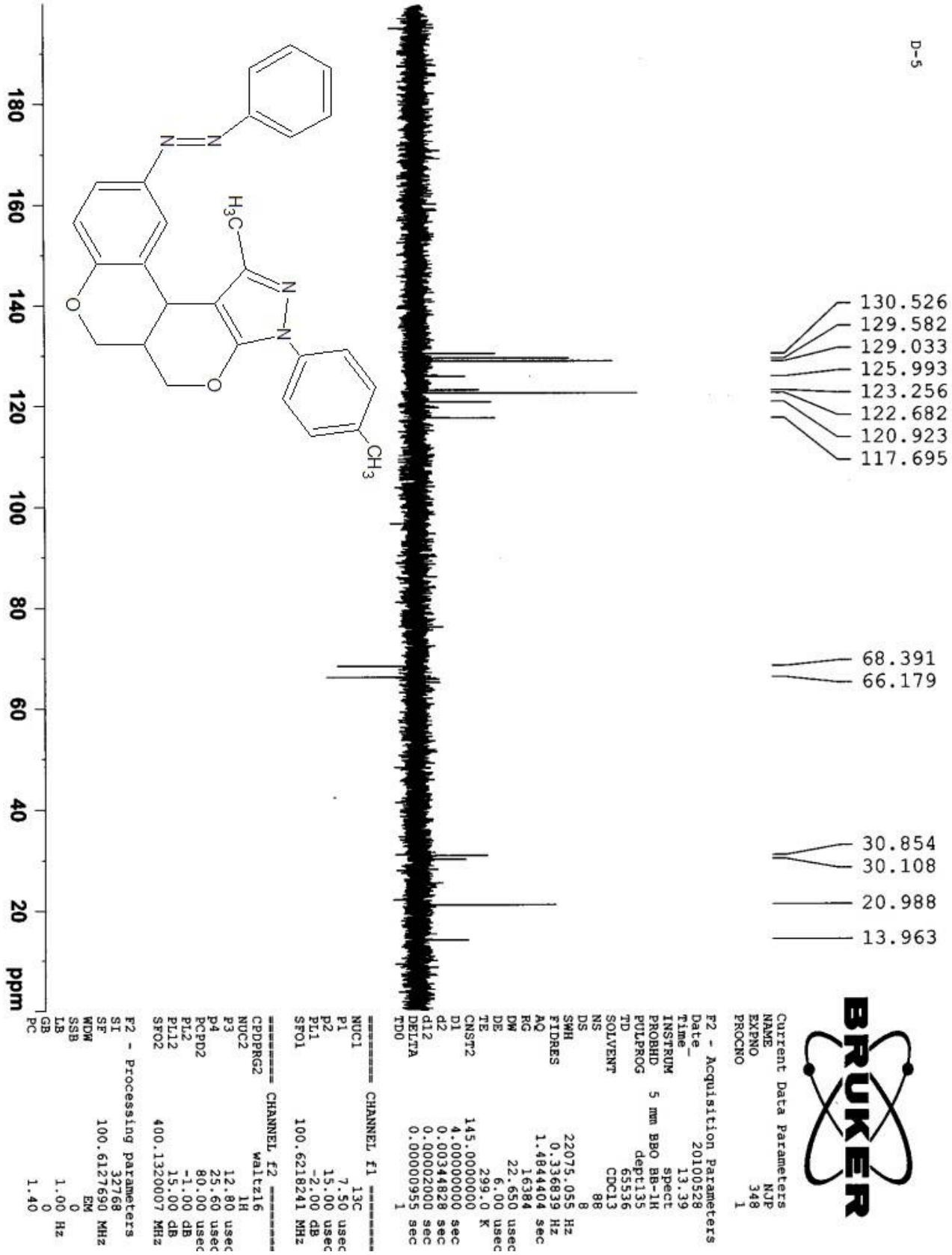
Compound 7e – DEPT 135



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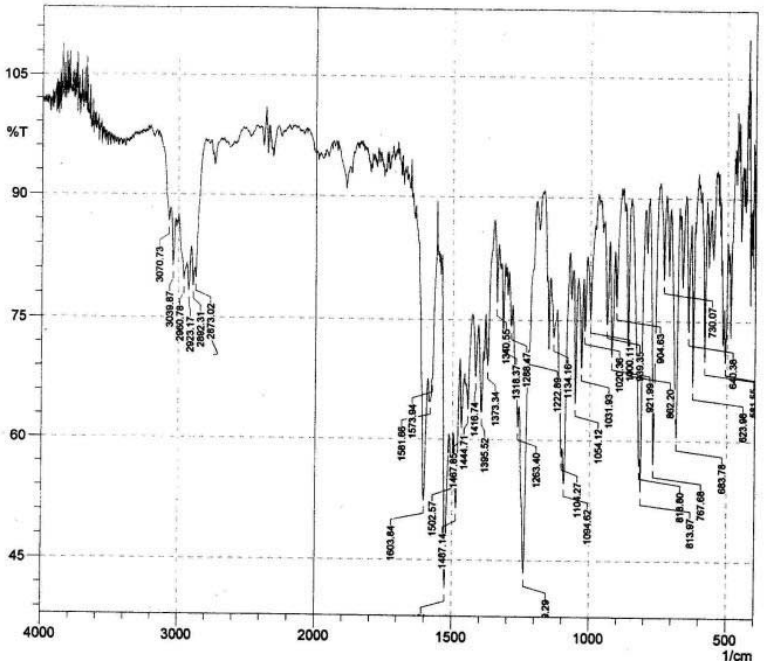
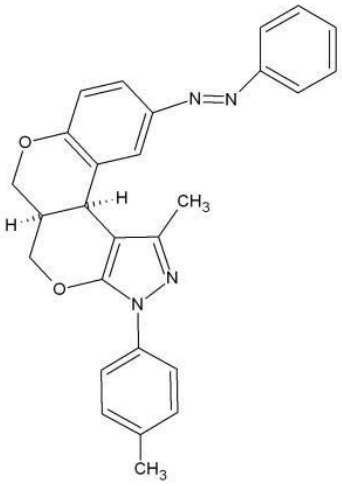
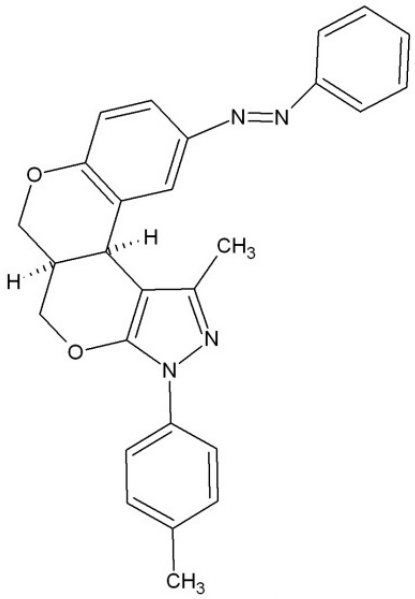
Compound 7e IR

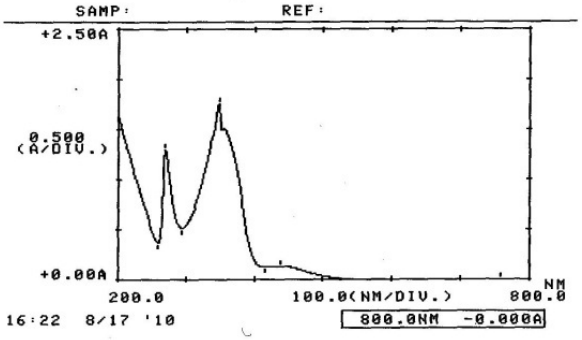




Compound 7e UV



λ	ABS	λ	ABS
759.0	0.002	600.0	-0.003
438.0	0.133	416.0	0.122
348.0	1.755	295.0	0.511
270.0	1.298	259.0	0.367



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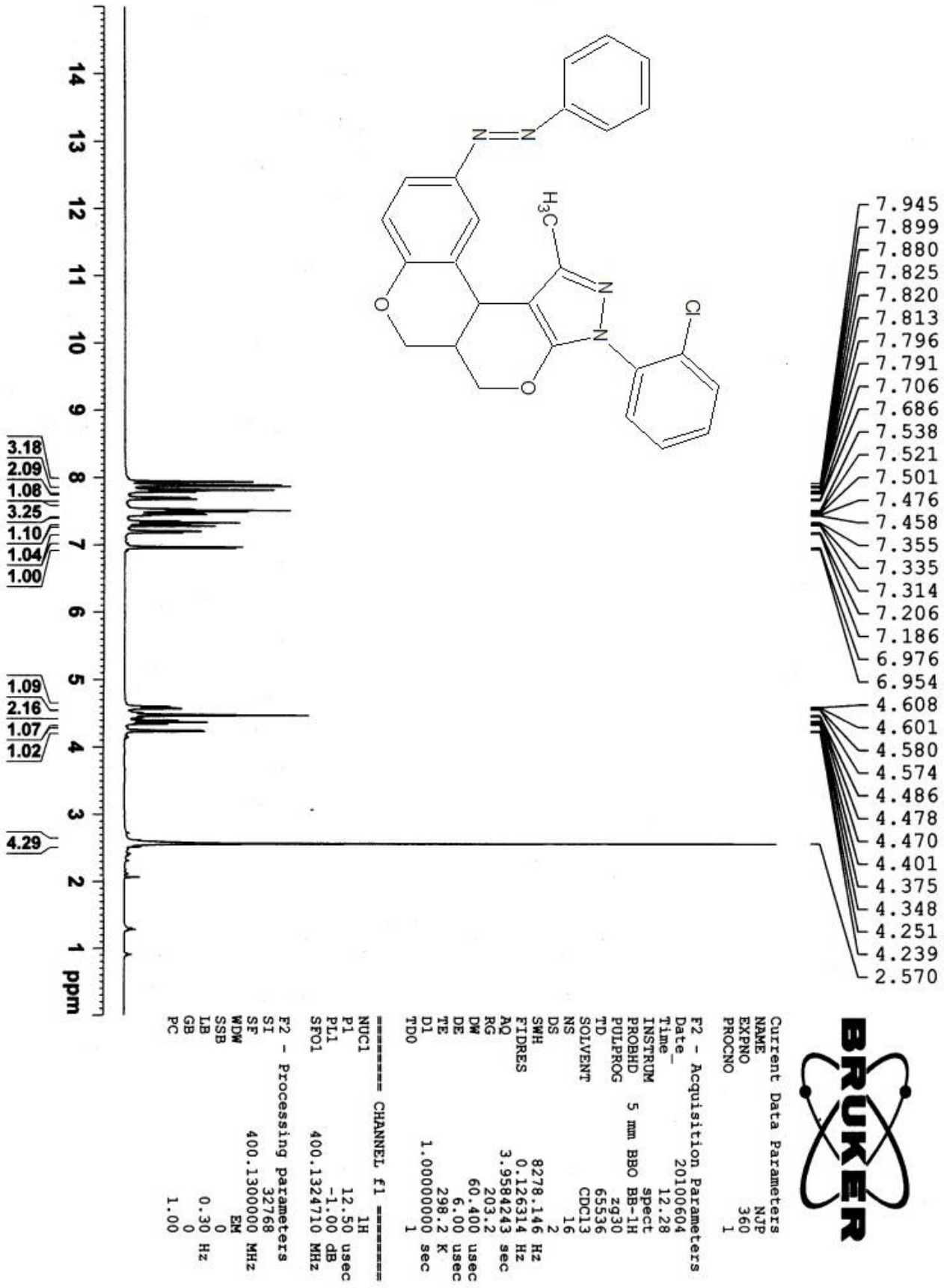
Compound 7f ^{1}H NMR



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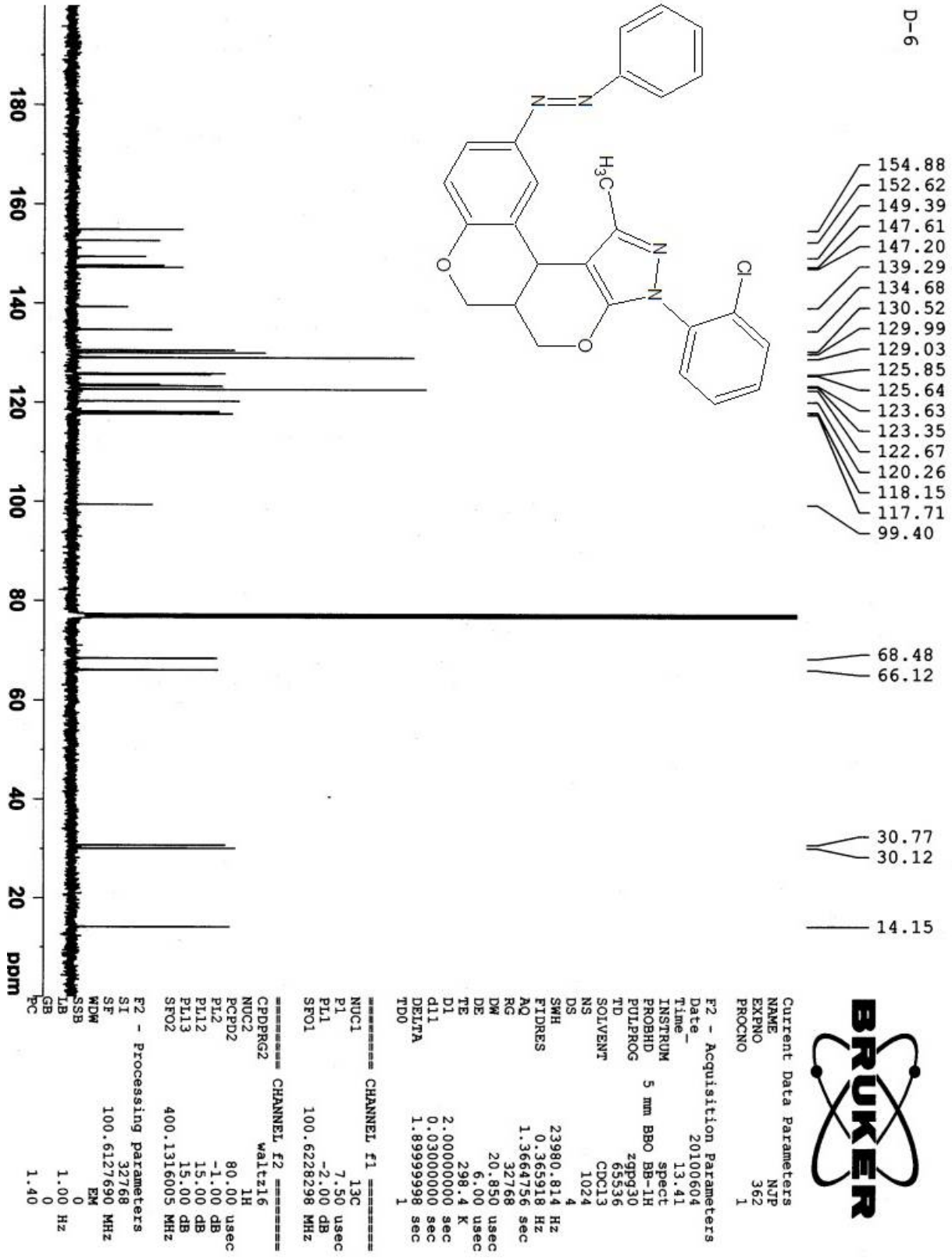
Compound 7f - ^{13}C NMR



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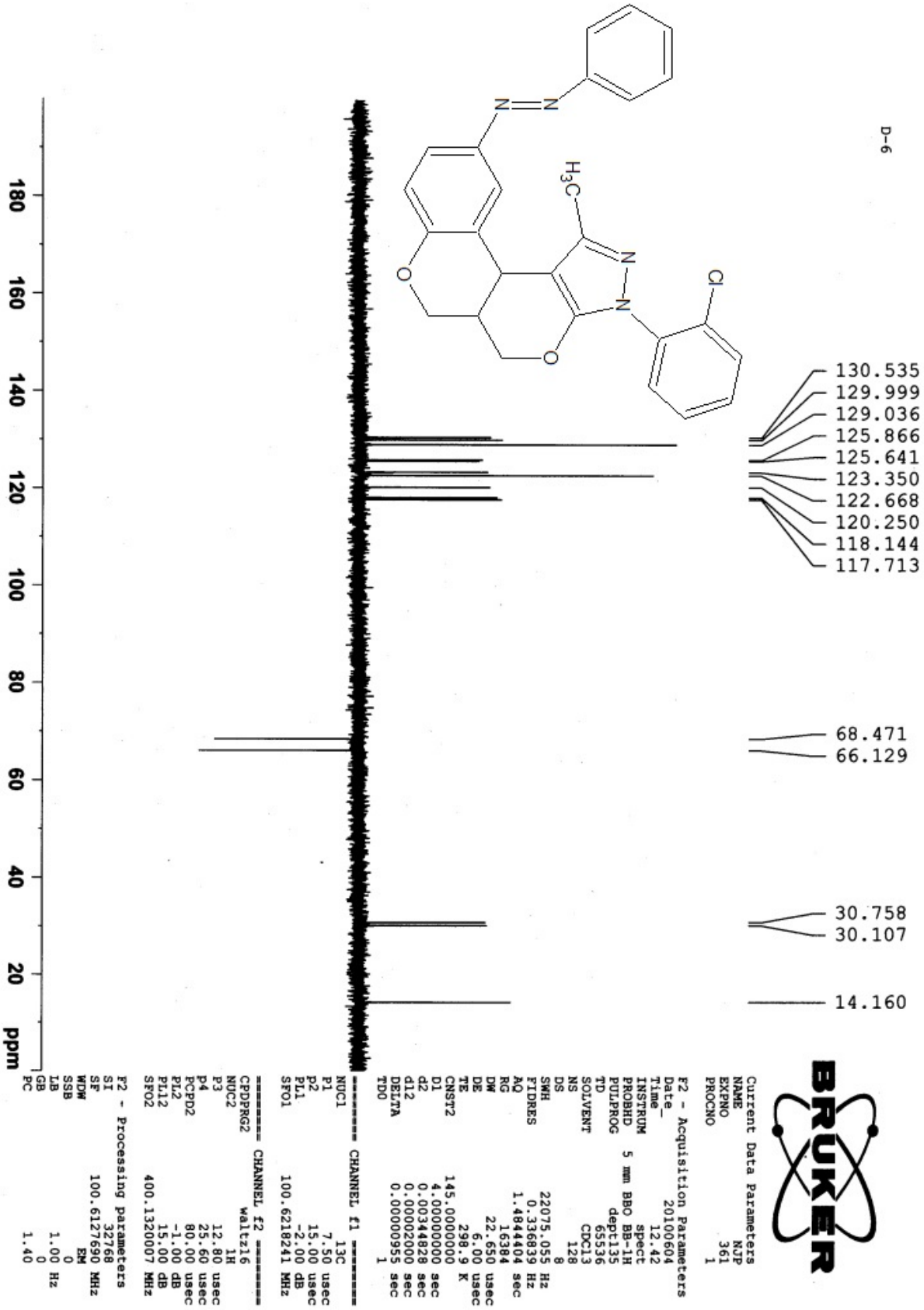
Compound 7f – DEPT 135



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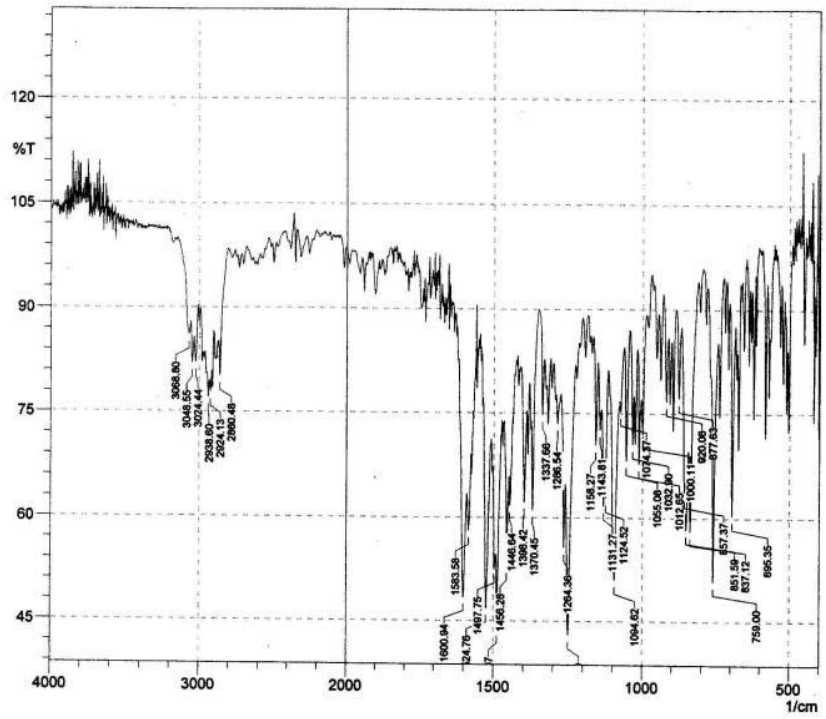
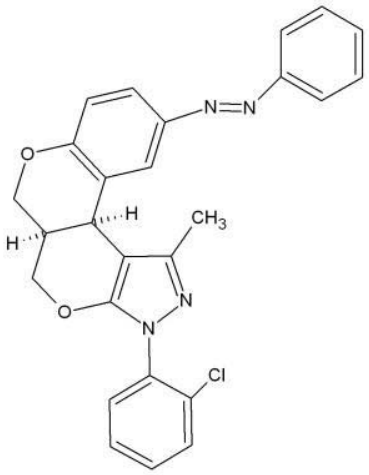
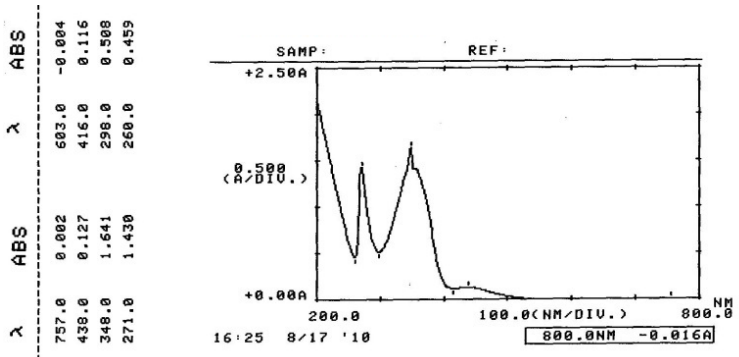
Compound 7f IR

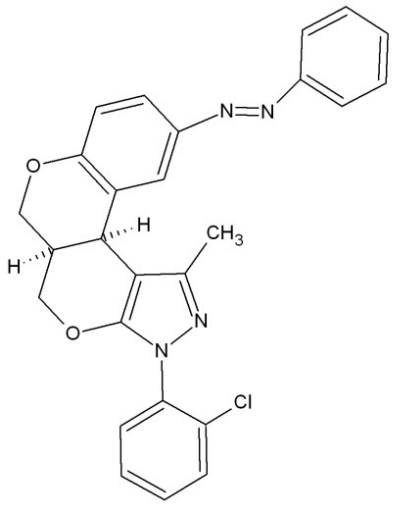




Compound 7f UV



λ	ABS	λ	ABS	λ	ABS
757.0	0.002	603.0	-0.004		
438.0	0.127	416.0	0.116		
348.0	1.641	298.0	0.508		
271.0	1.430	260.0	0.459		



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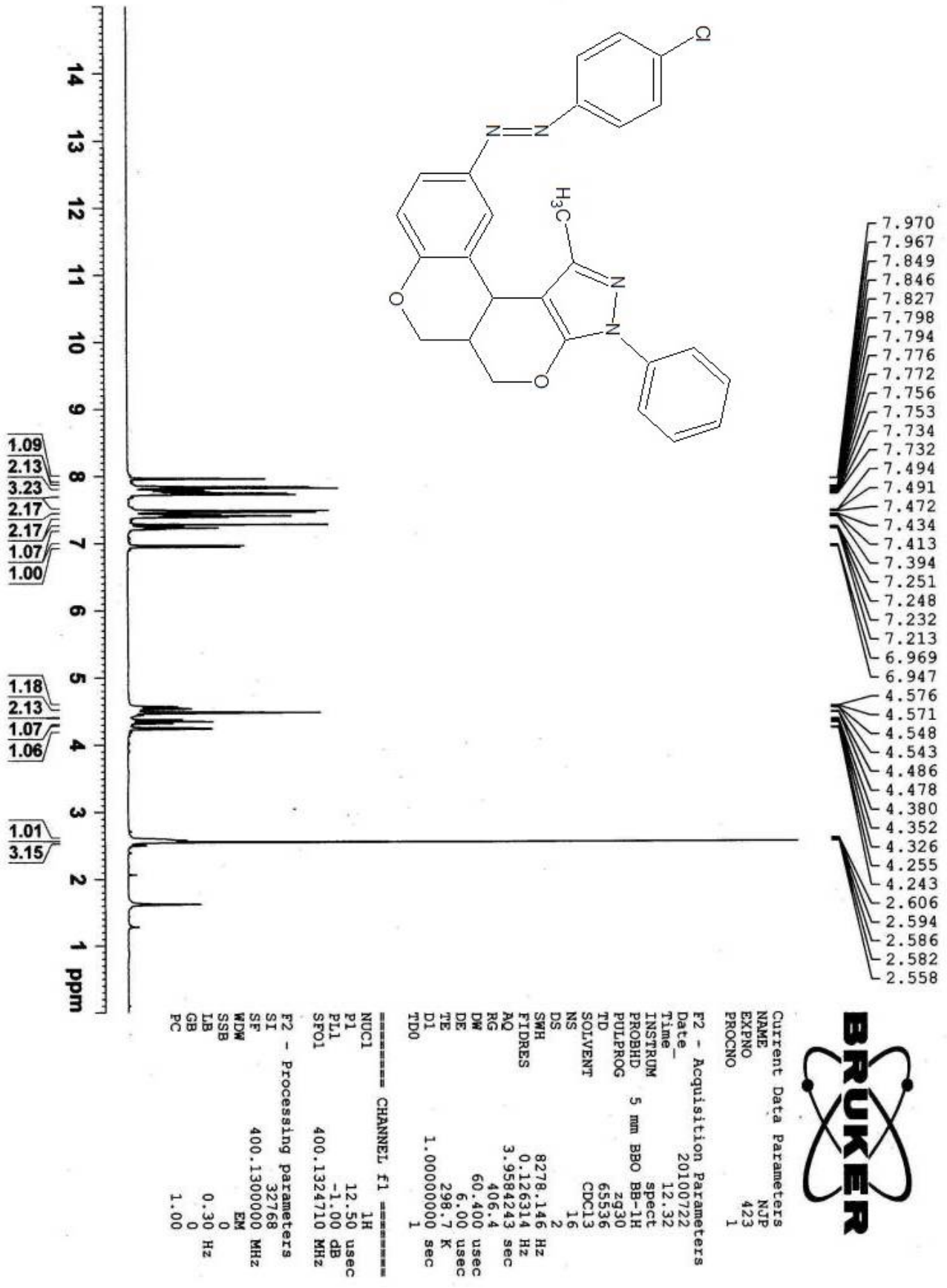
Compound 7g ^{1}H NMR



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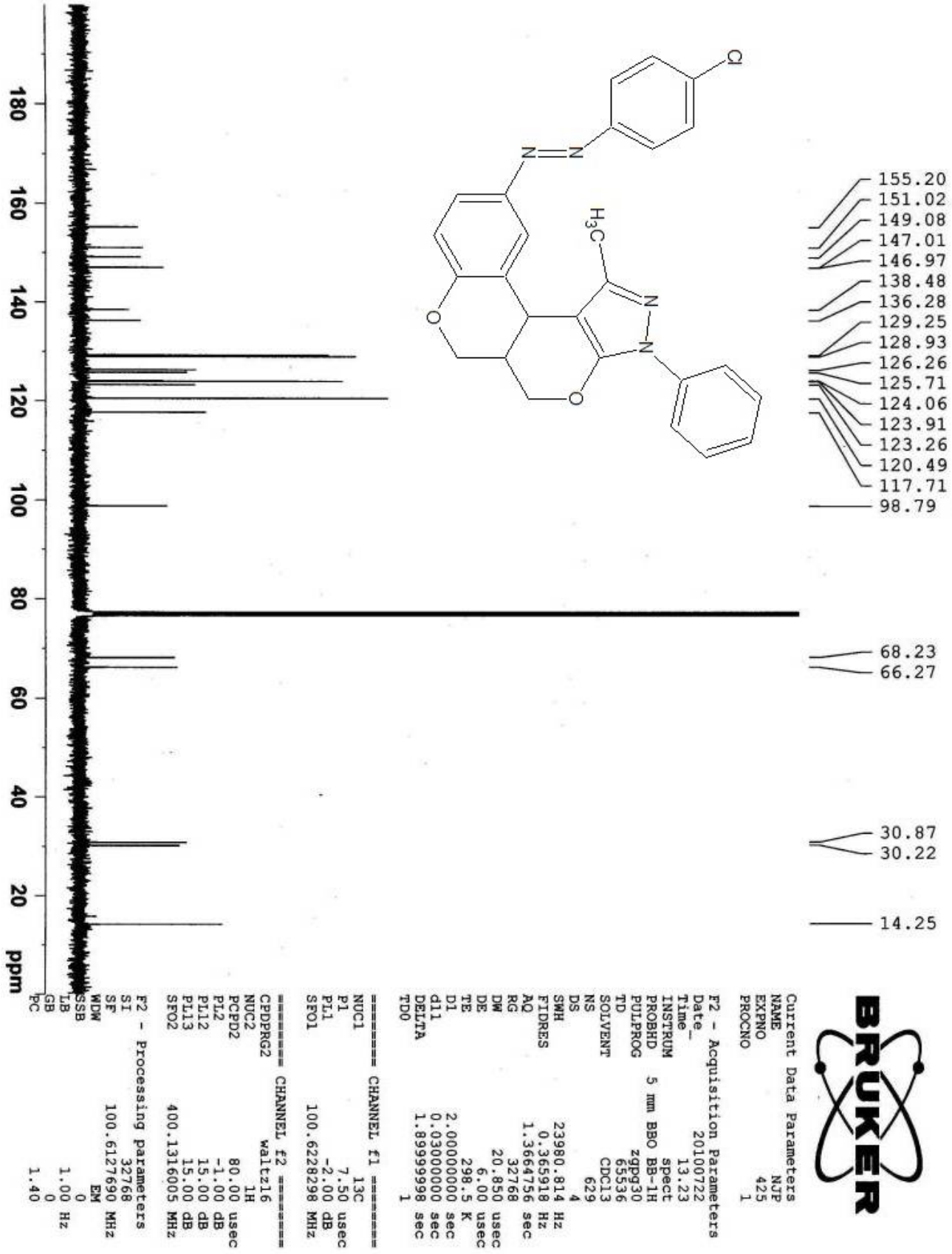
Compound 7g - ^{13}C NMR



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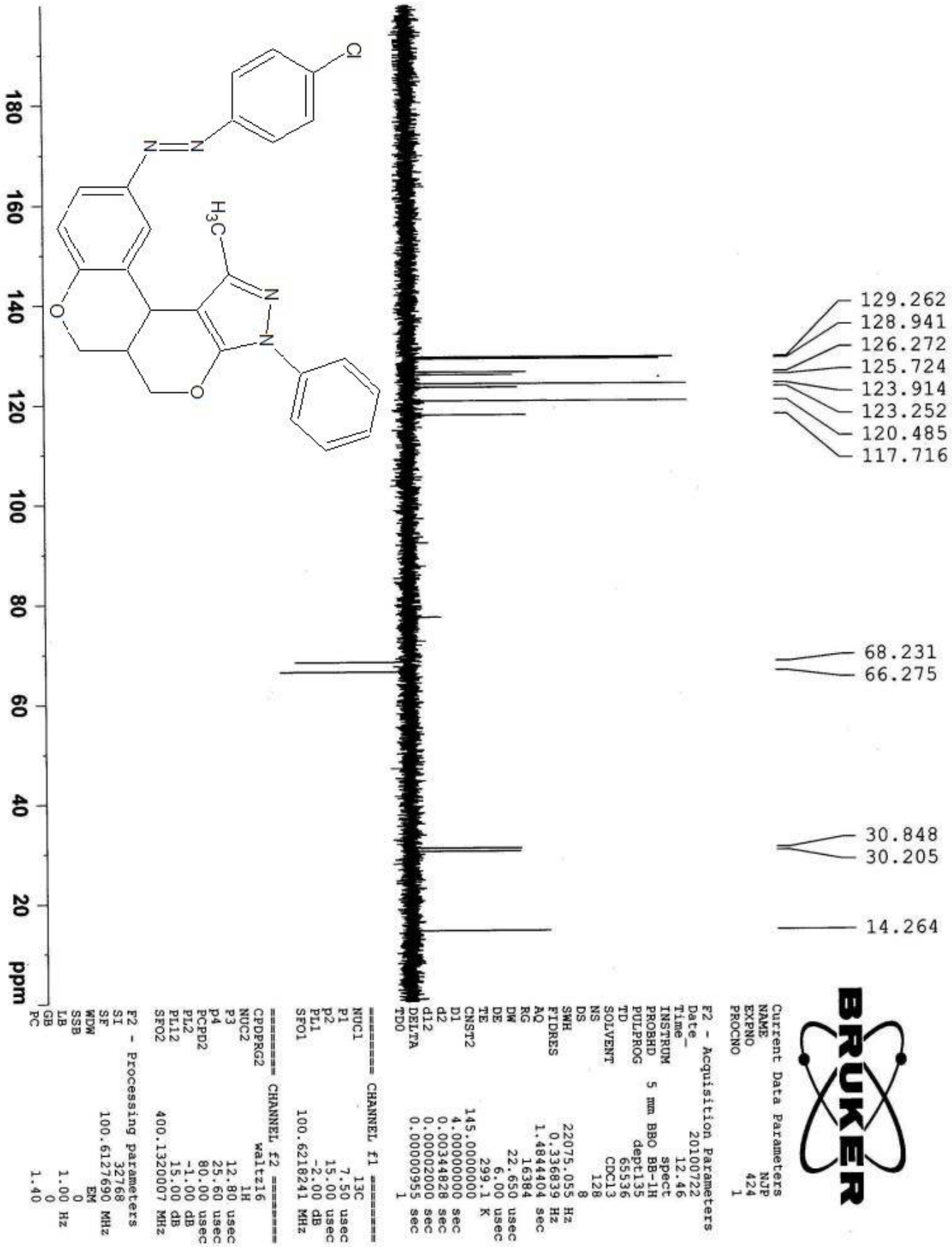
Compound 7g – DEPT 135



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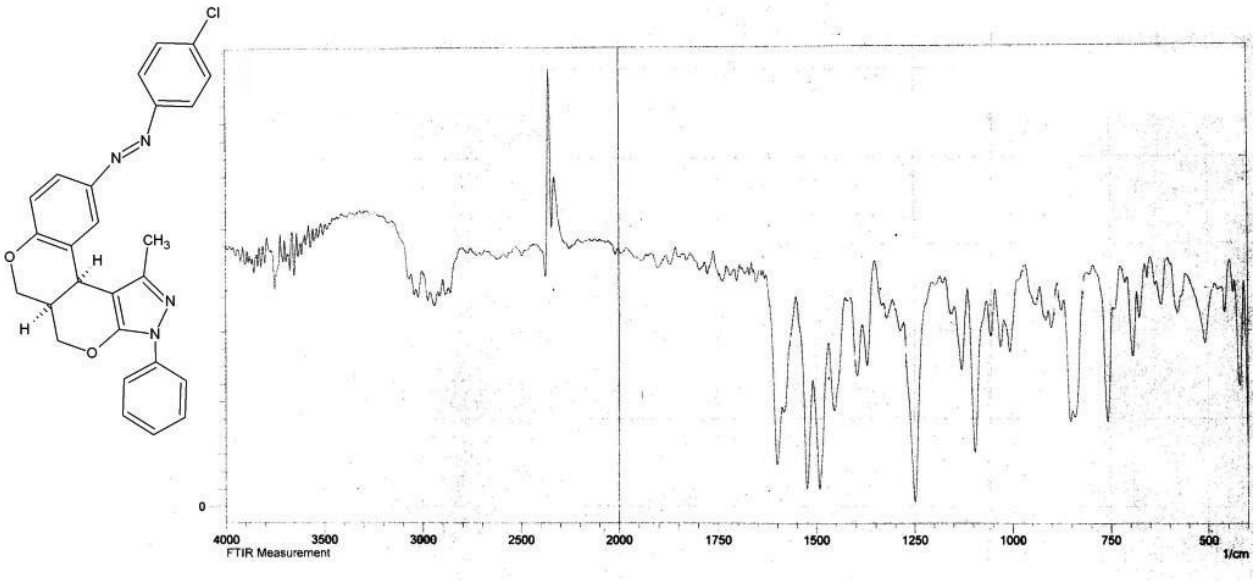
Compound 7g IR



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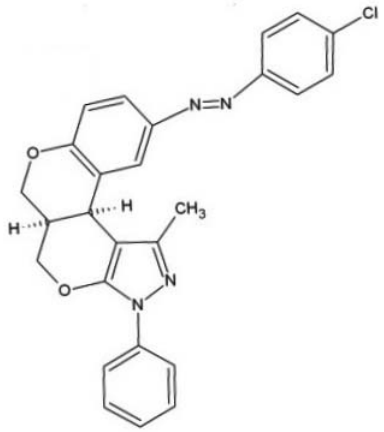

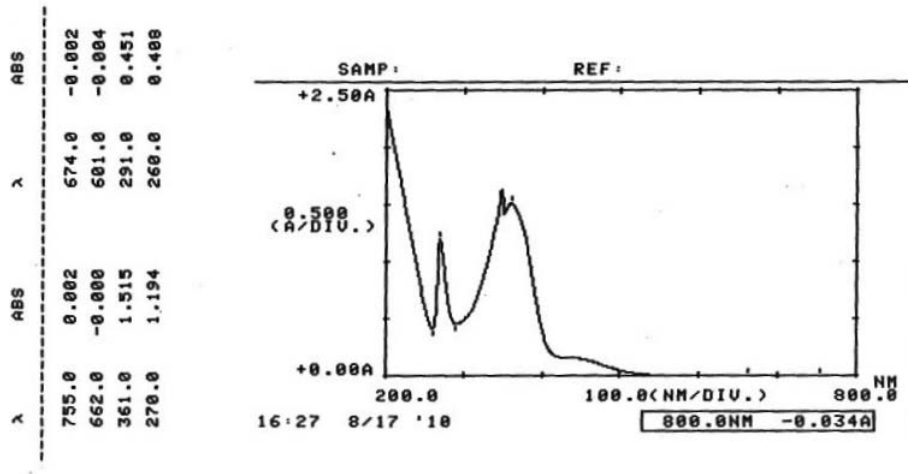

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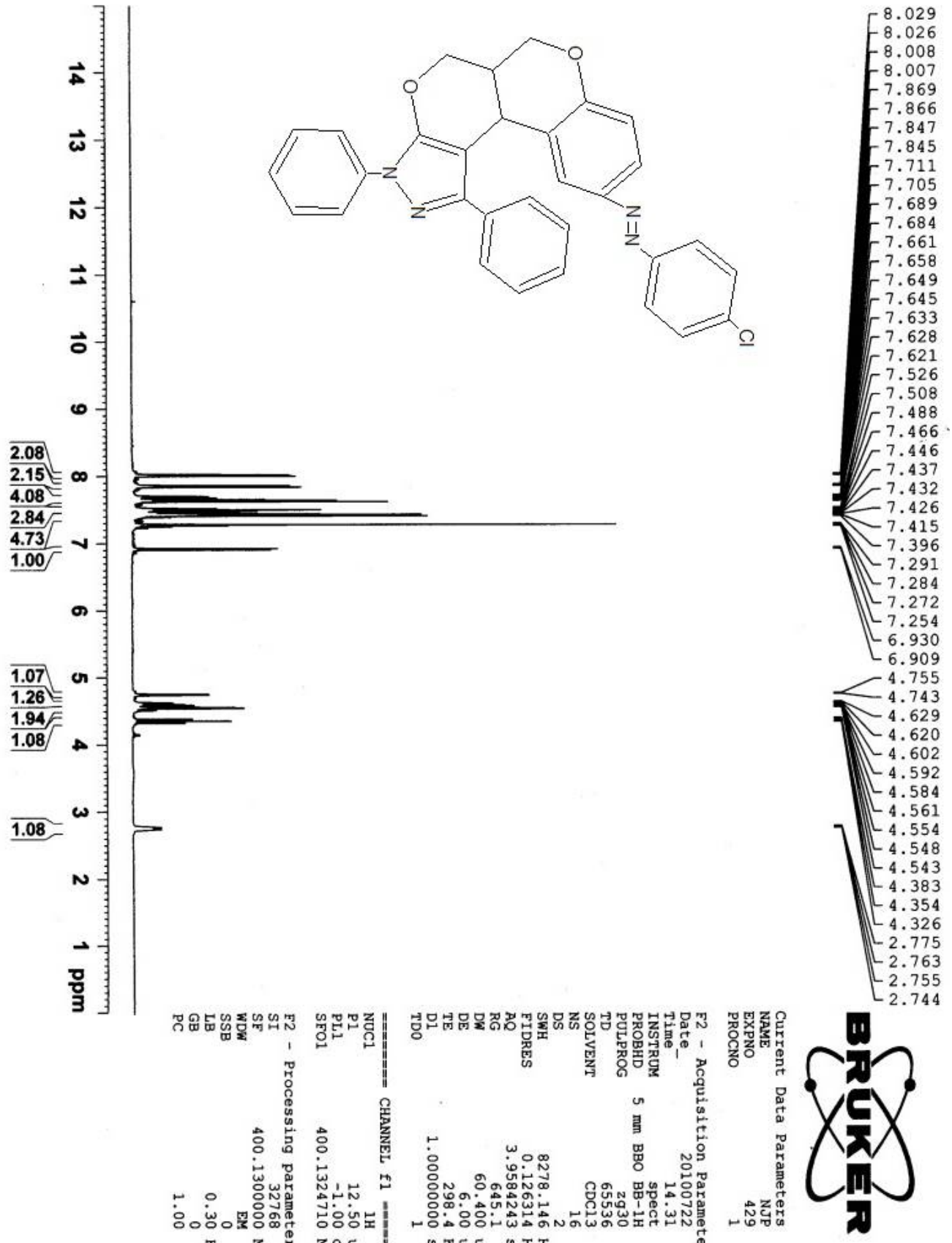
Compound 7i ^{1}H NMR



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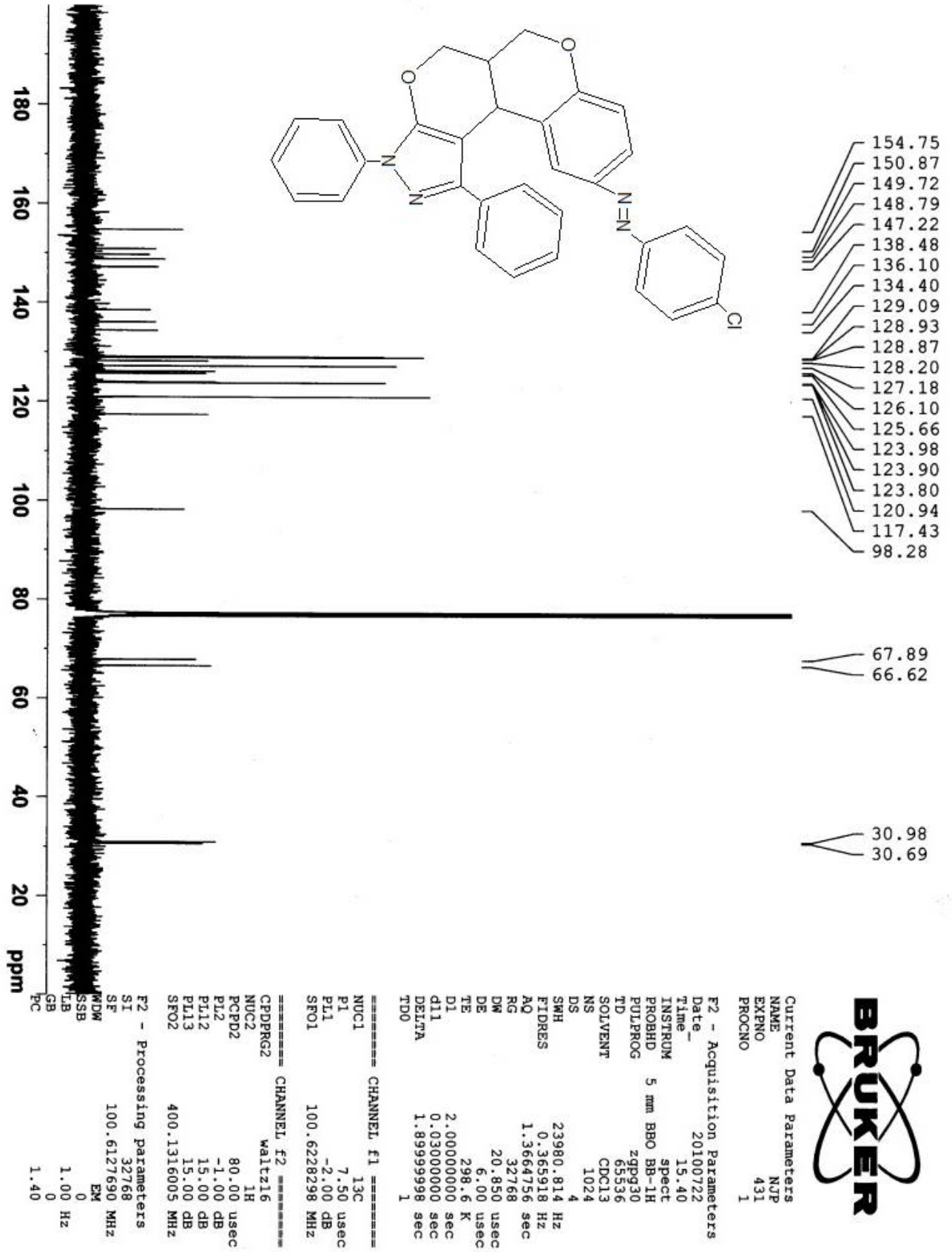
Compound 7i - ^{13}C NMR



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EXPNO 431
PROCNO 1

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Time_ 15.40
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FIDRES 0.365918 Hz
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RG 32768
DW 20.850 usec
DE 6.00 usec
TE 298.6 K
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PL1 -2.00 dB
SFO1 100.6228298 MHz

======== CHANNEL f2 ========
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SFO2 400.1316005 MHz

F2 - Processing parameters
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GB 0
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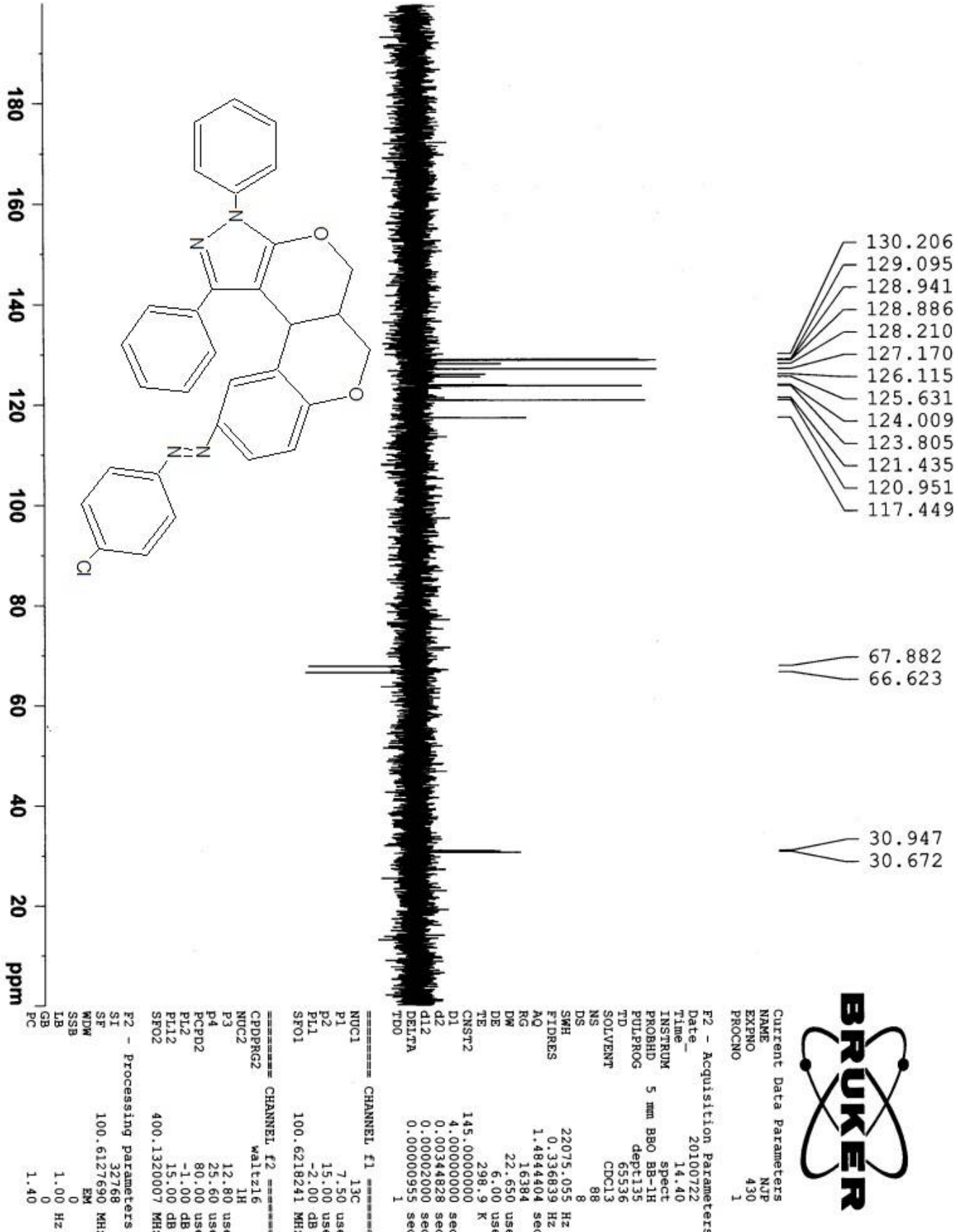
Compound 7i – DEPT 135



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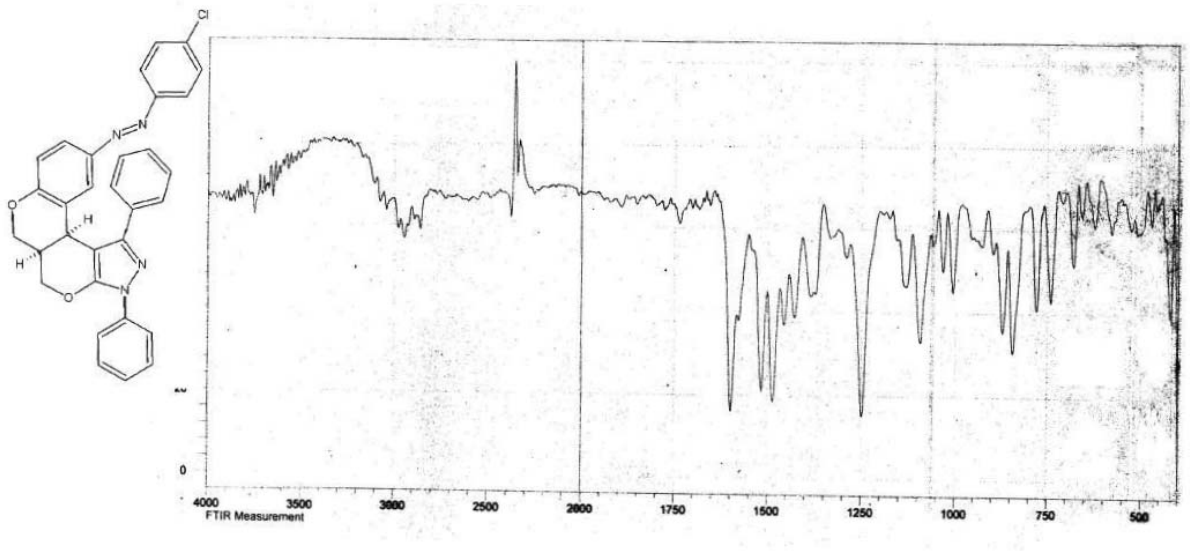
Compound 7i IR



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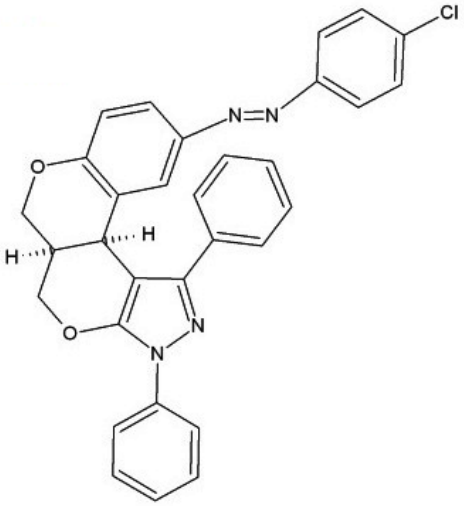
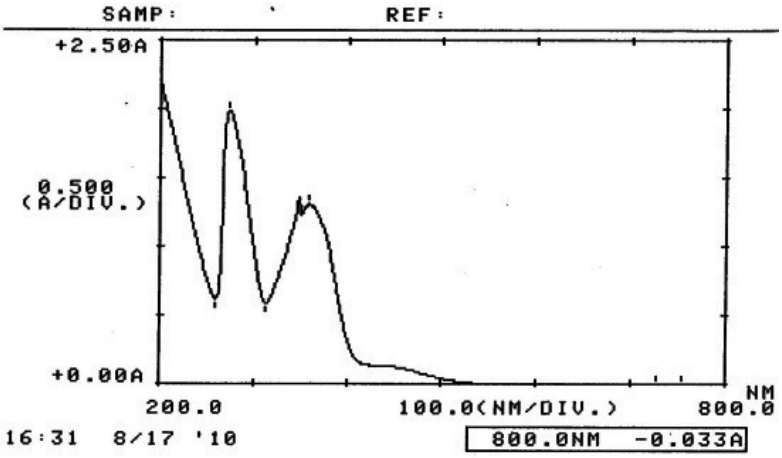


λ	ABS
736.0	-0.001
601.0	-0.004
313.0	0.580
259.0	0.619

λ	ABS
754.0	0.002
727.0	0.001
360.0	1.308
274.0	1.986



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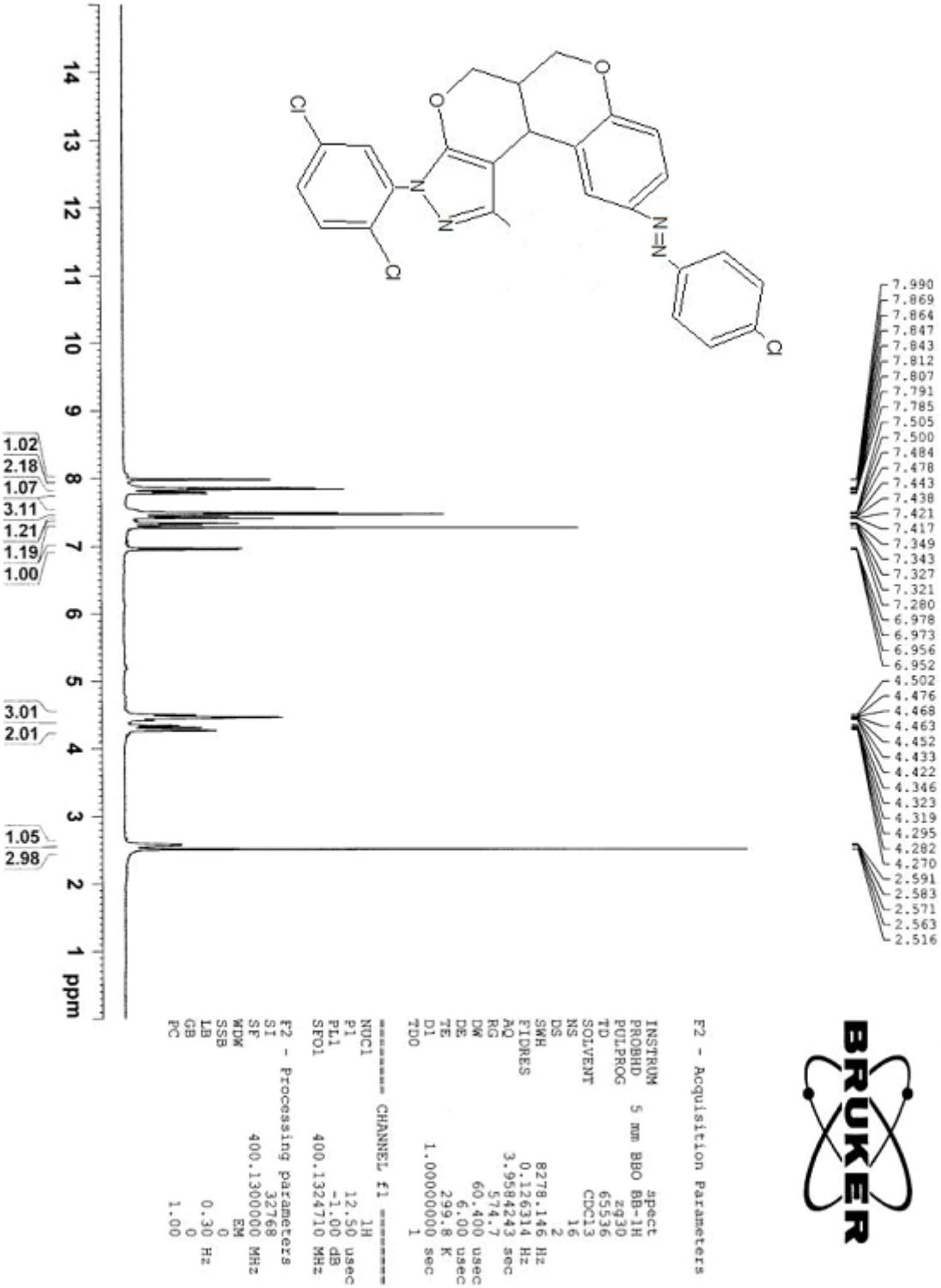
Compound 7j ^{1}H NMR



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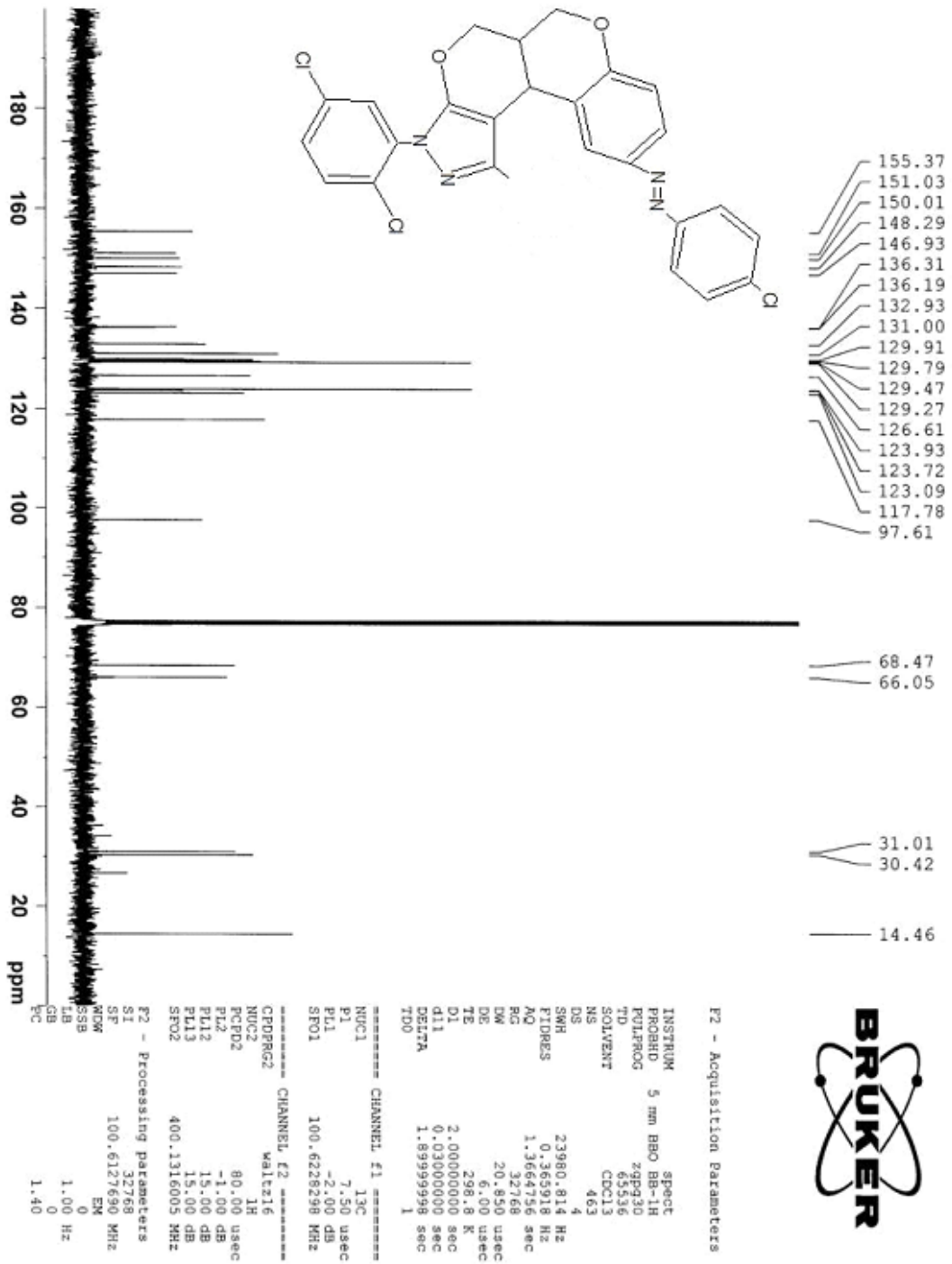
Compound 7j - ^{13}C NMR



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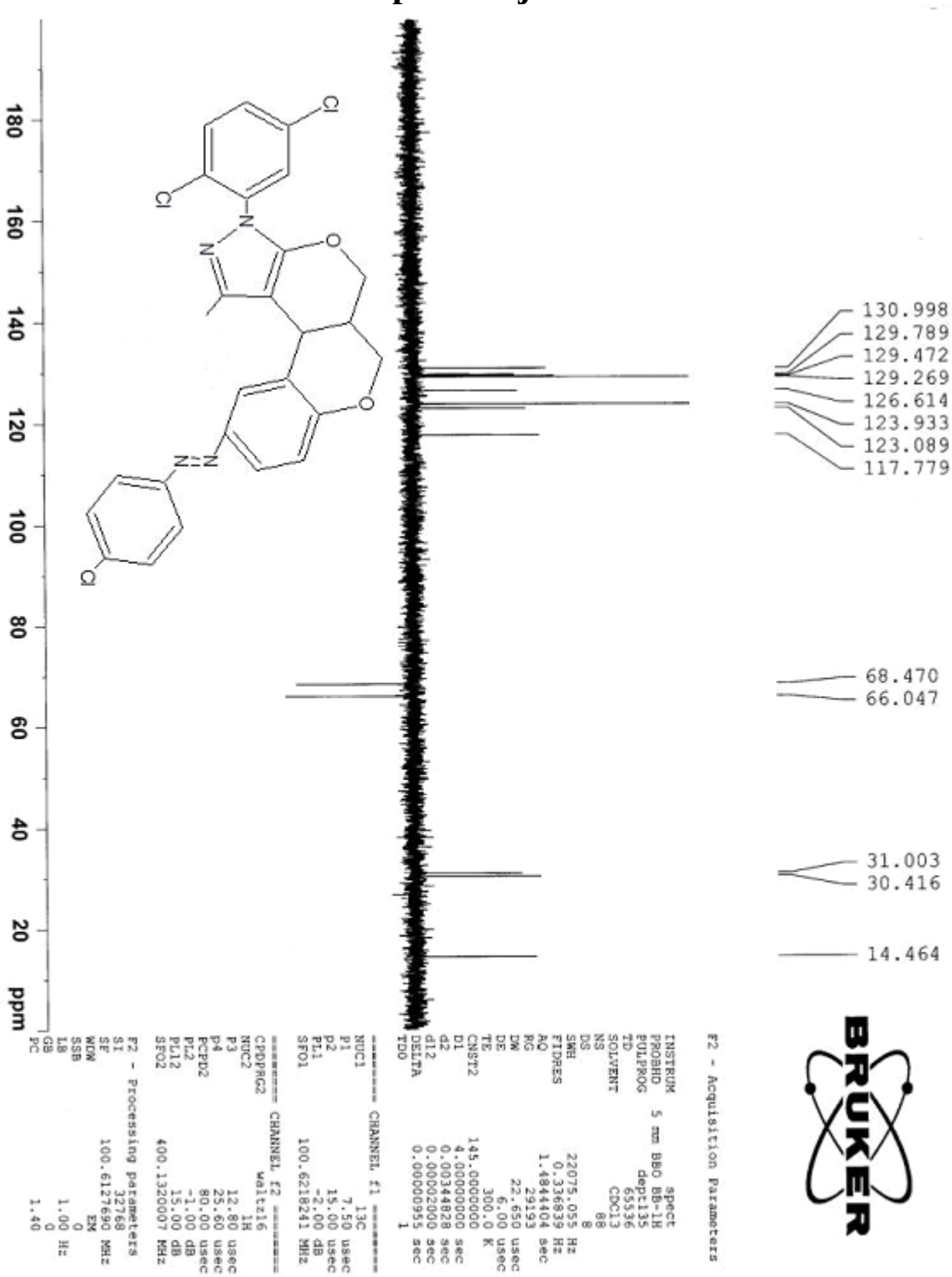
Compound 7j – DEPT 135



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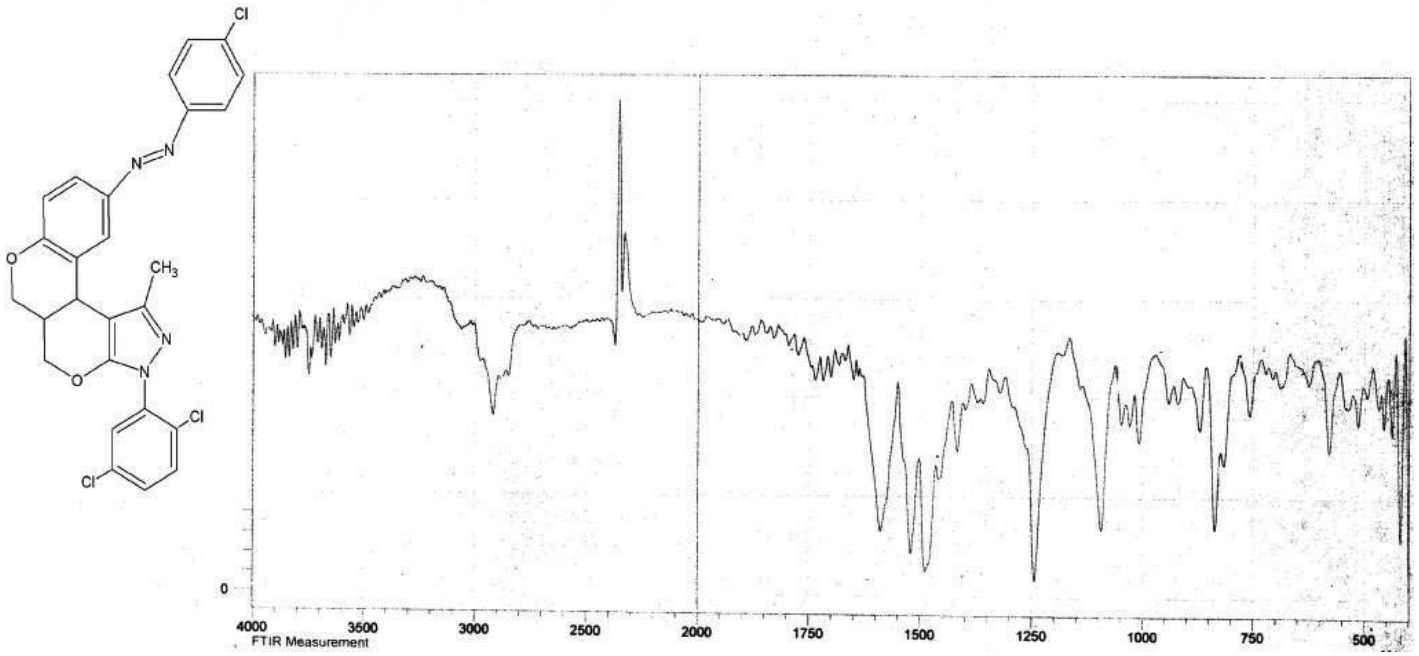
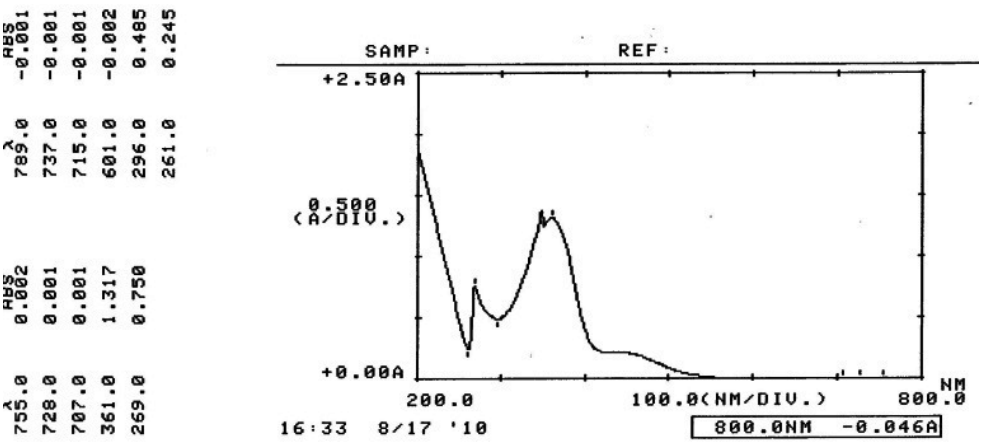
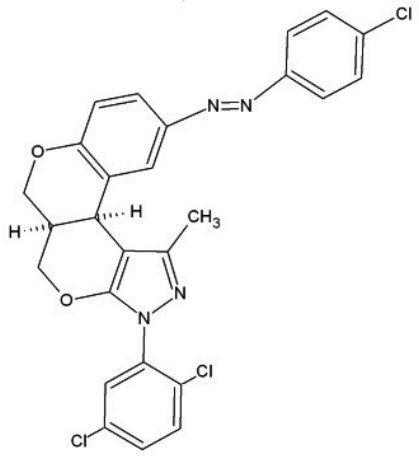
Compound 7j IR



Compound 7j UV





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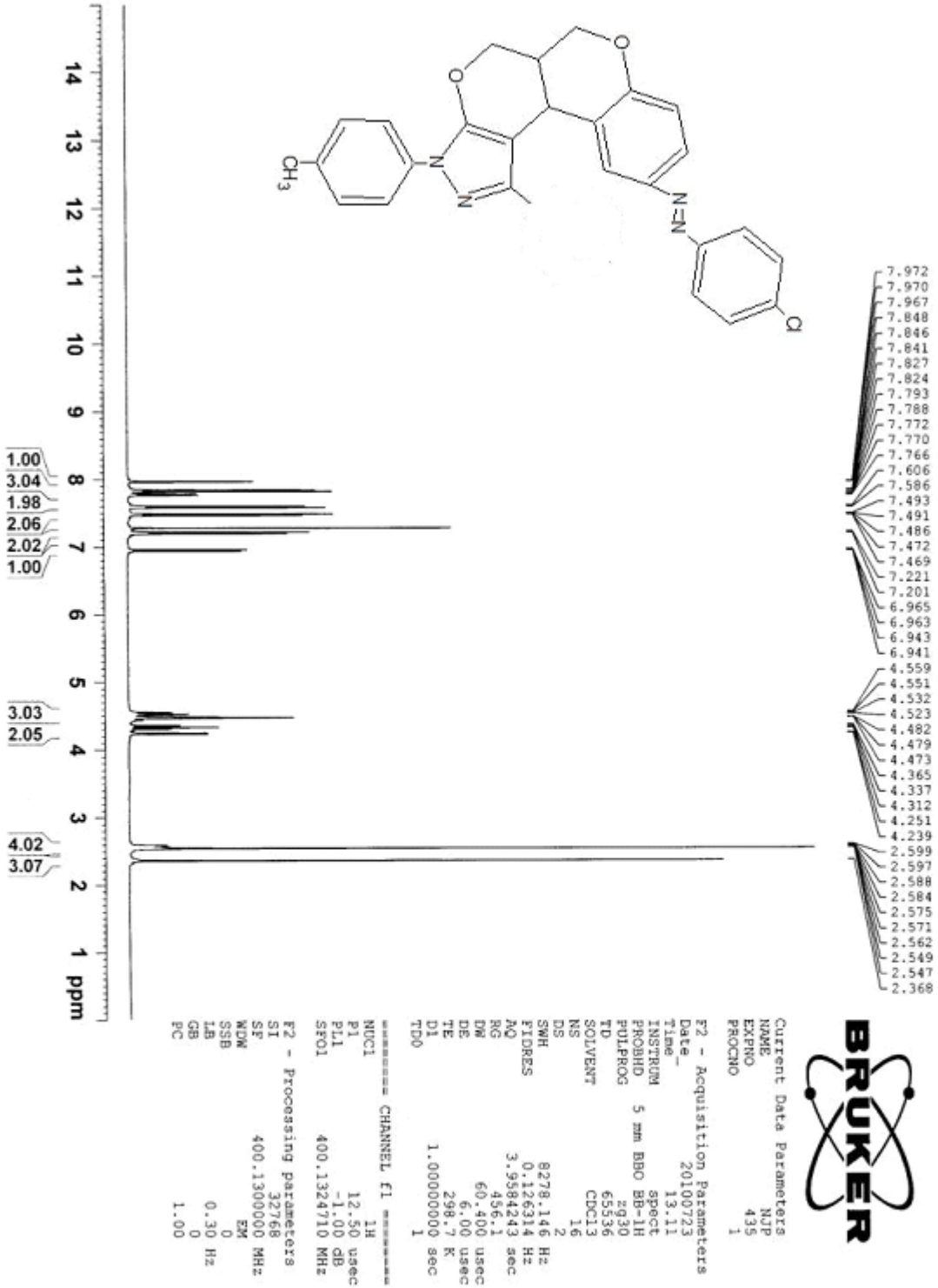
Compound 7k ^{1}H NMR



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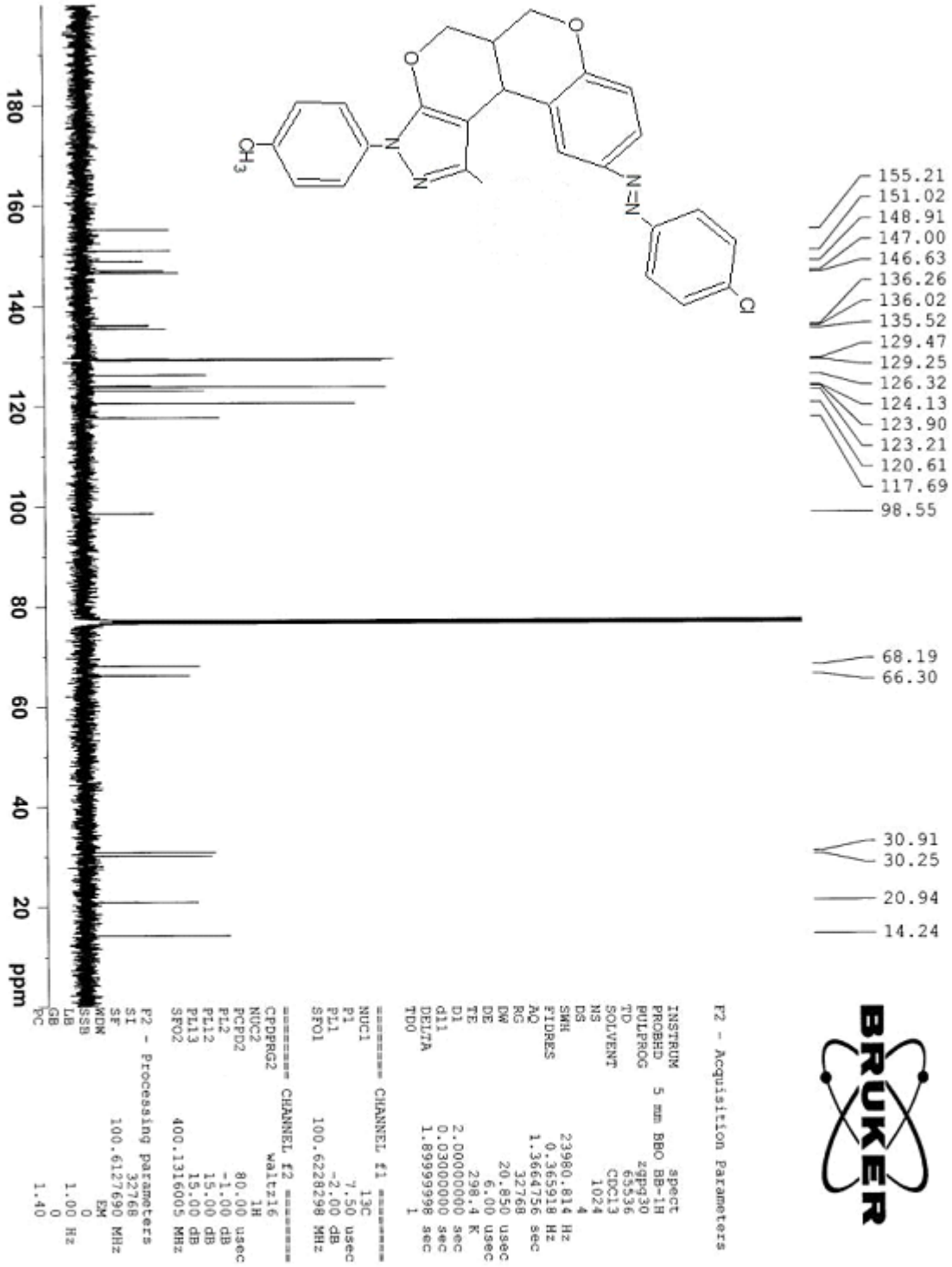
Compound 7k - ^{13}C NMR



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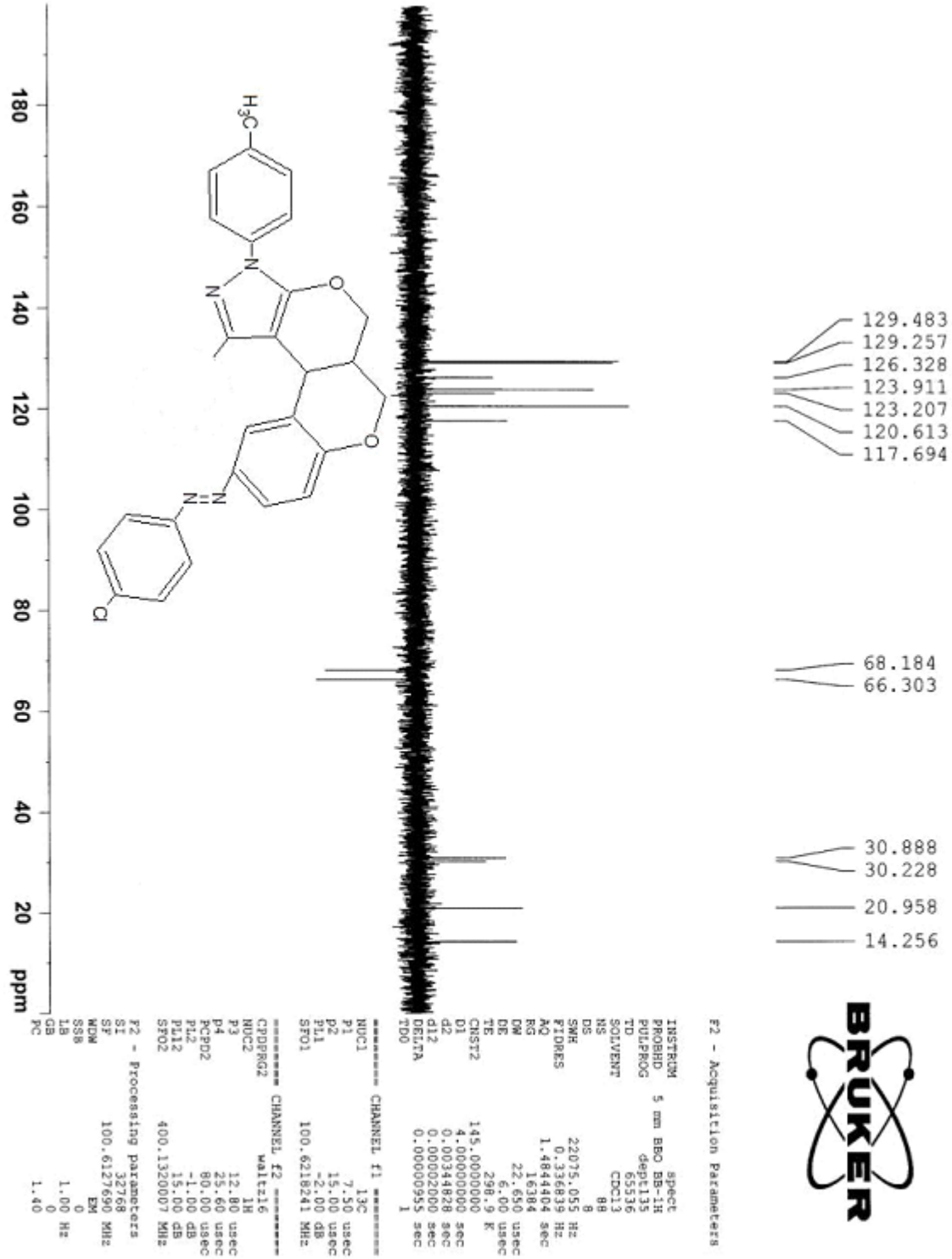
Compound 7k – DEPT 135



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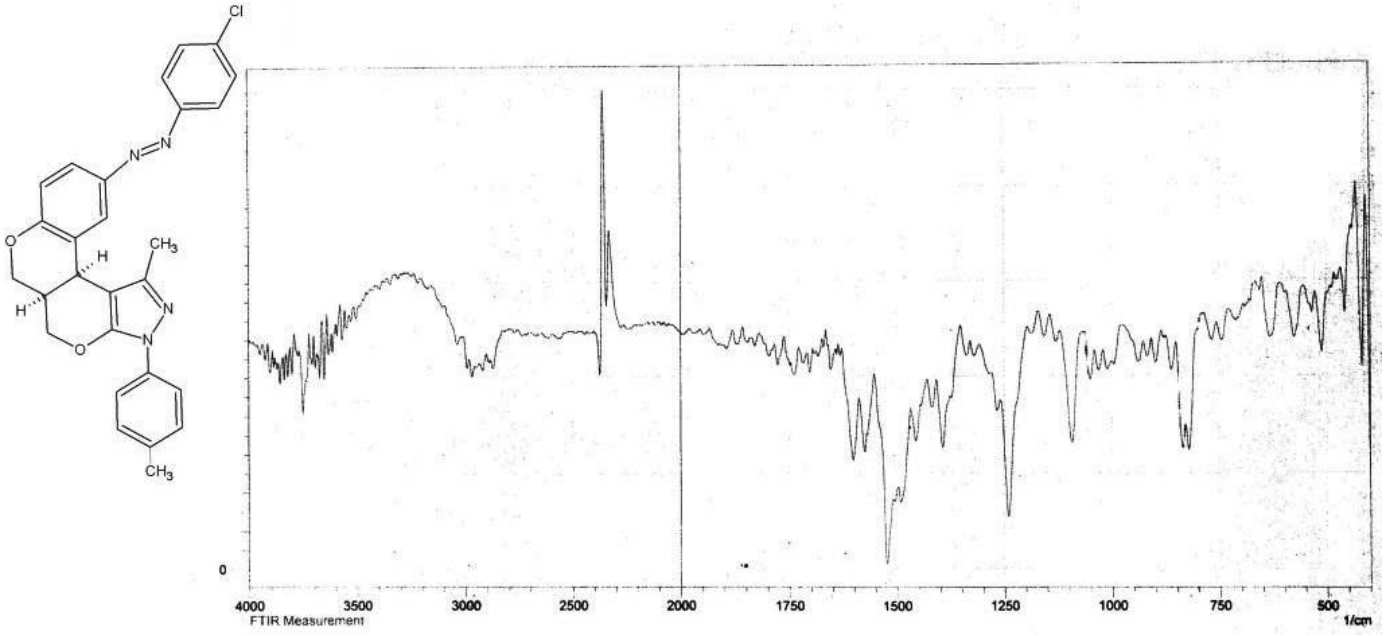
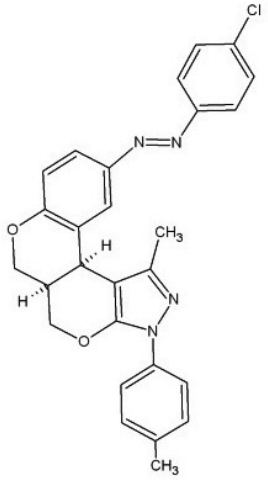
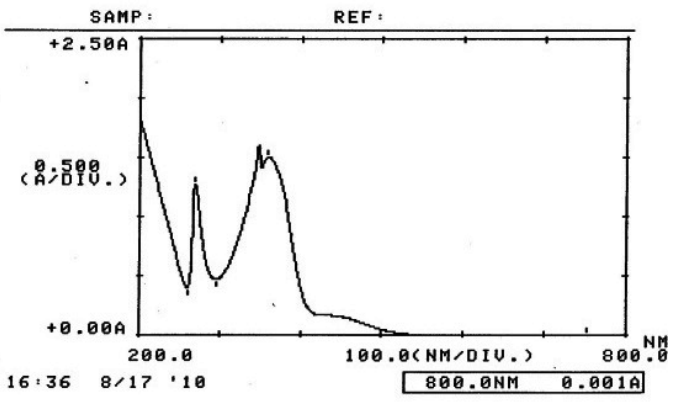
Compound 7k IR



Compound 7k UV



A	ABS	λ	ABS
752.0	0.001	764.0	-0.001
360.0	1.504	603.0	-0.005
270.0	1.271	296.0	0.464
		260.0	0.401



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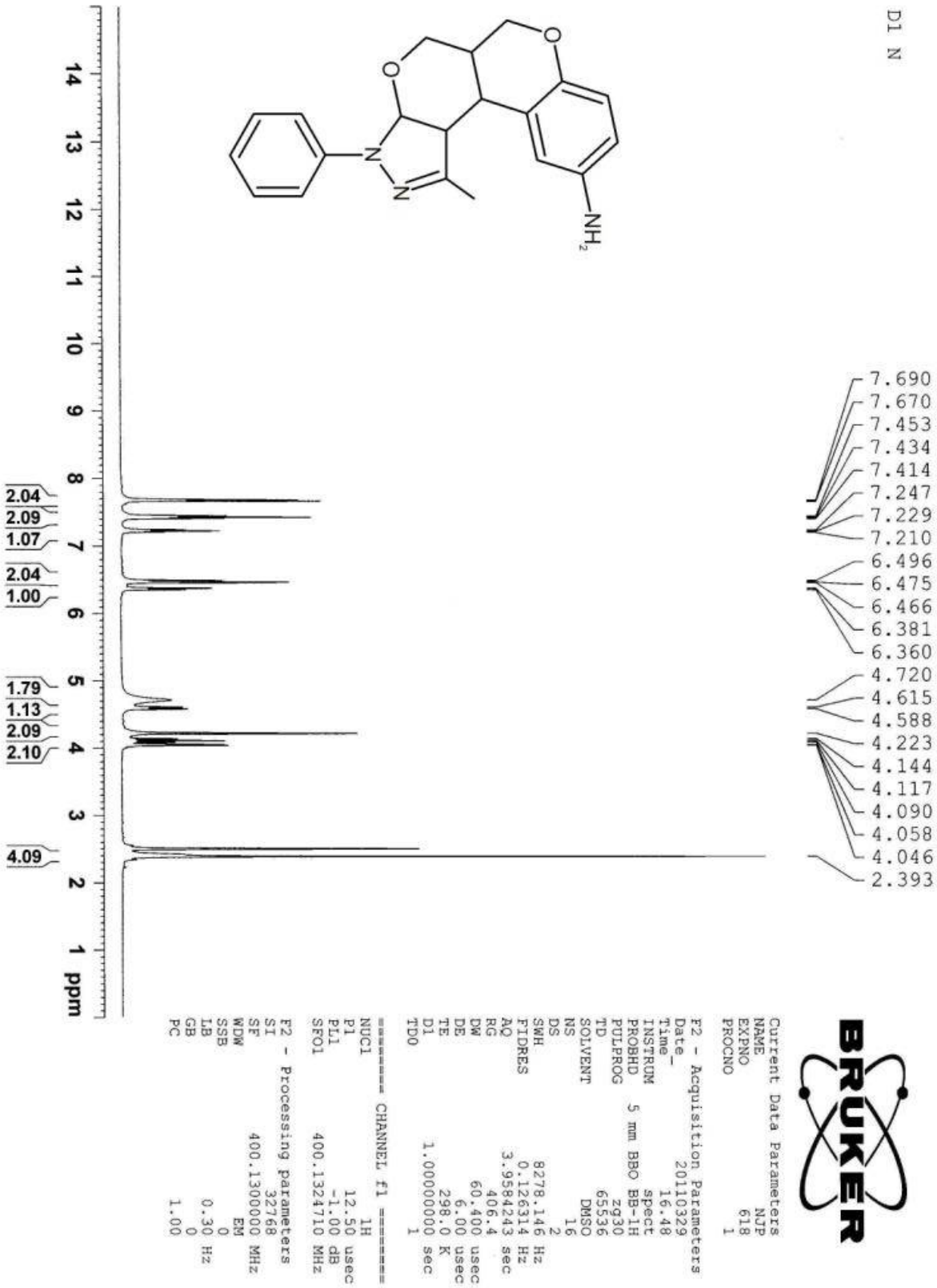
Compound 10 ^{1}H NMR



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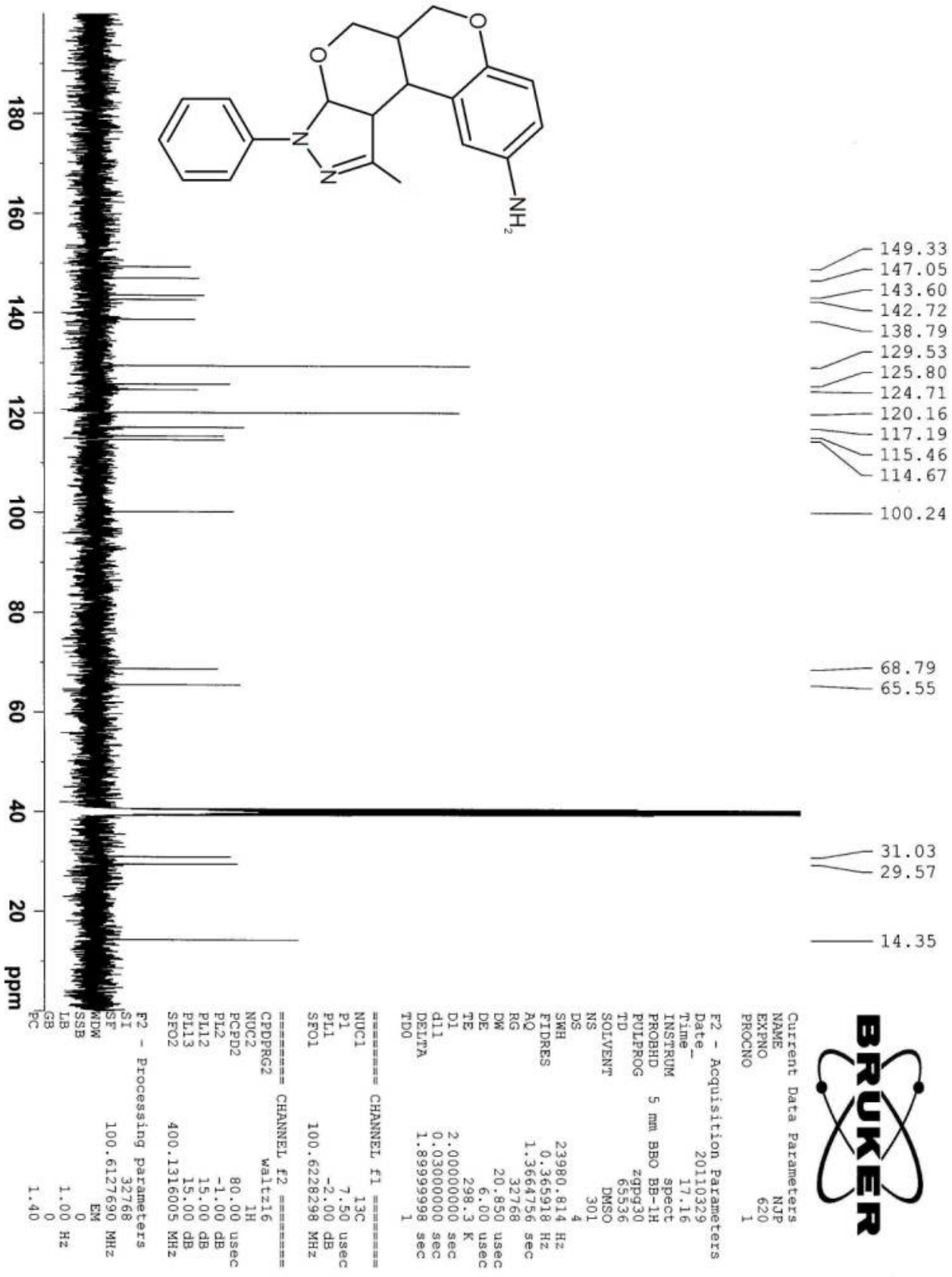
Compound 10 - ^{13}C NMR



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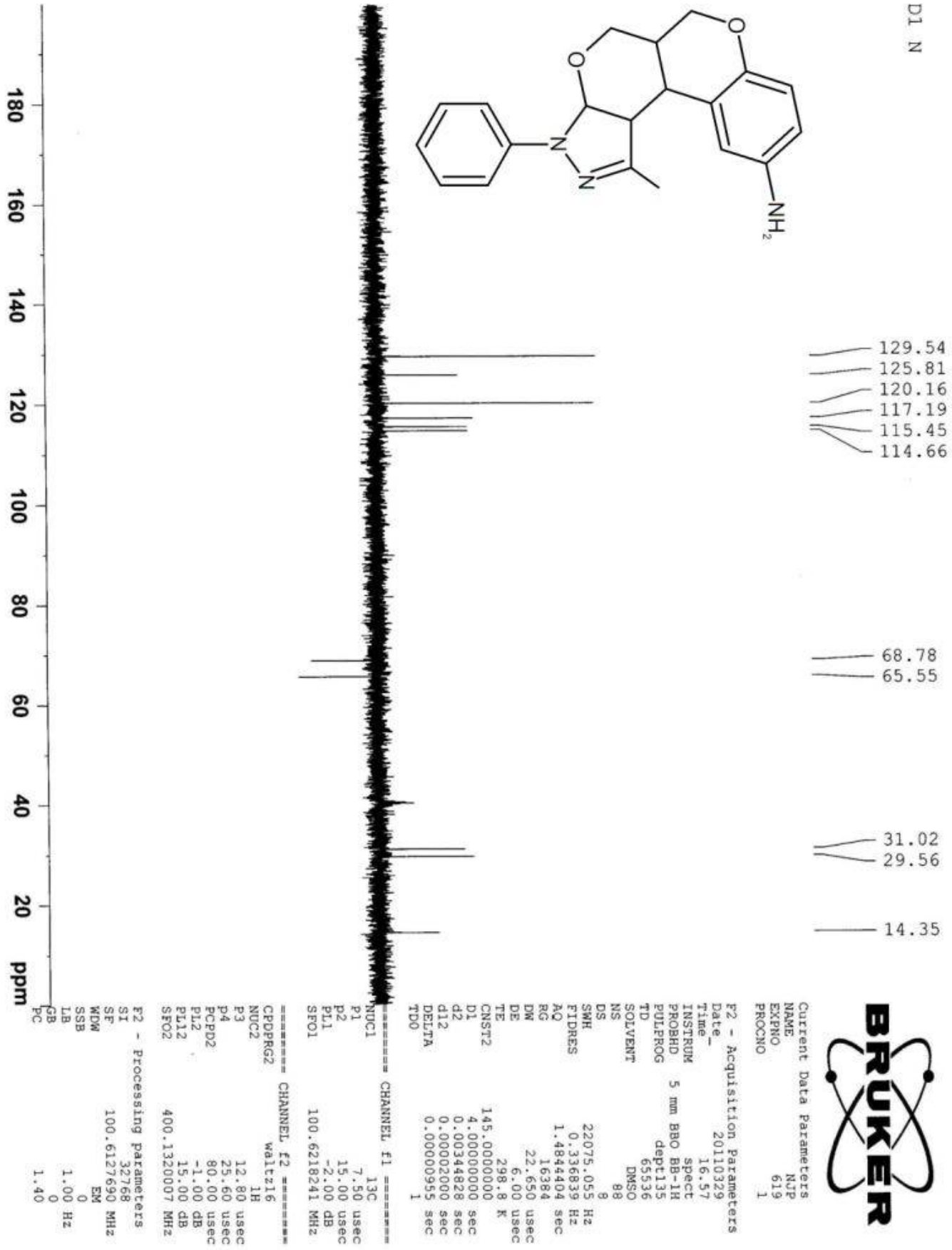
Compound 10 – DEPT 135



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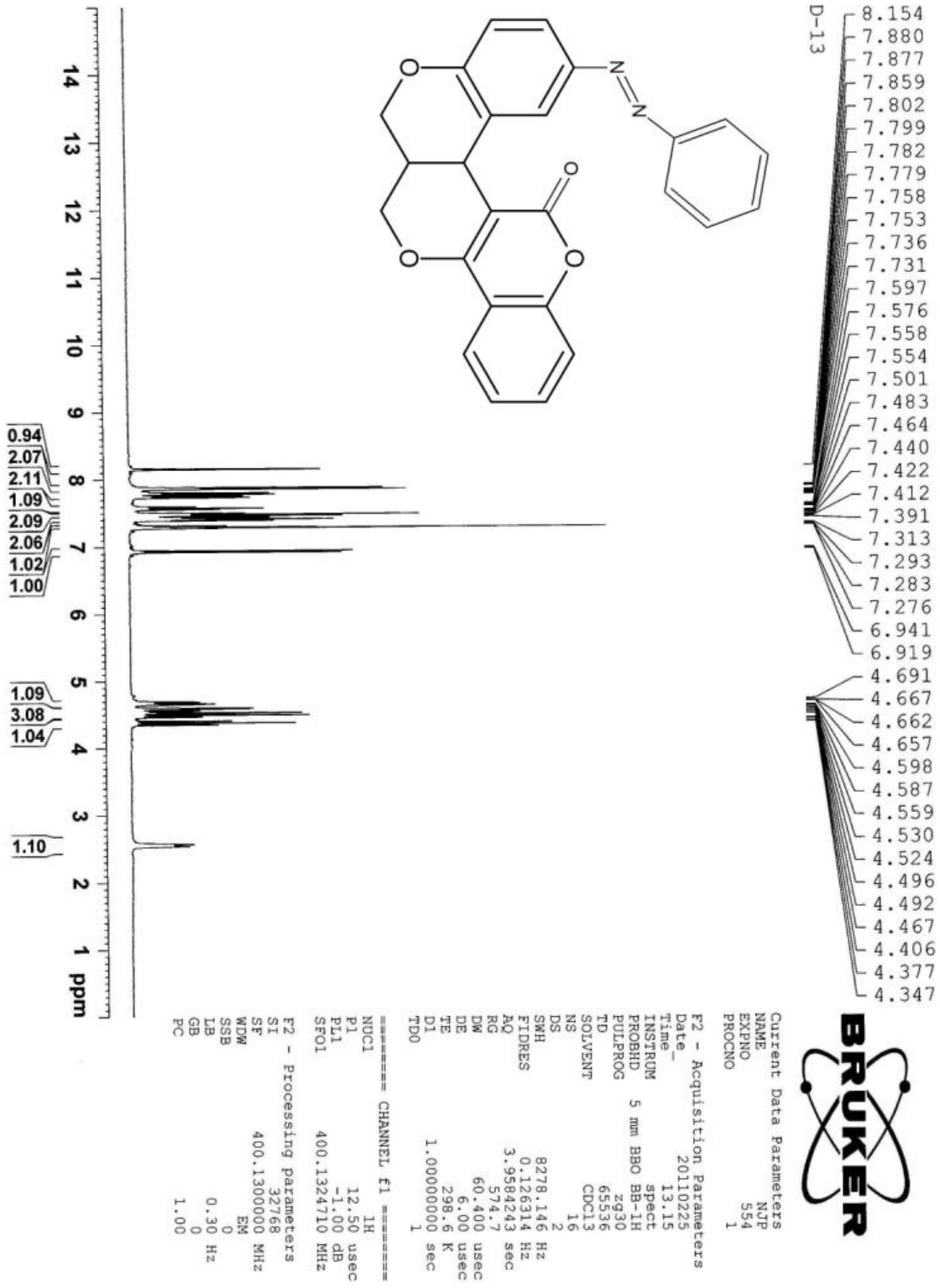
Compound 9b ^{1}H NMR



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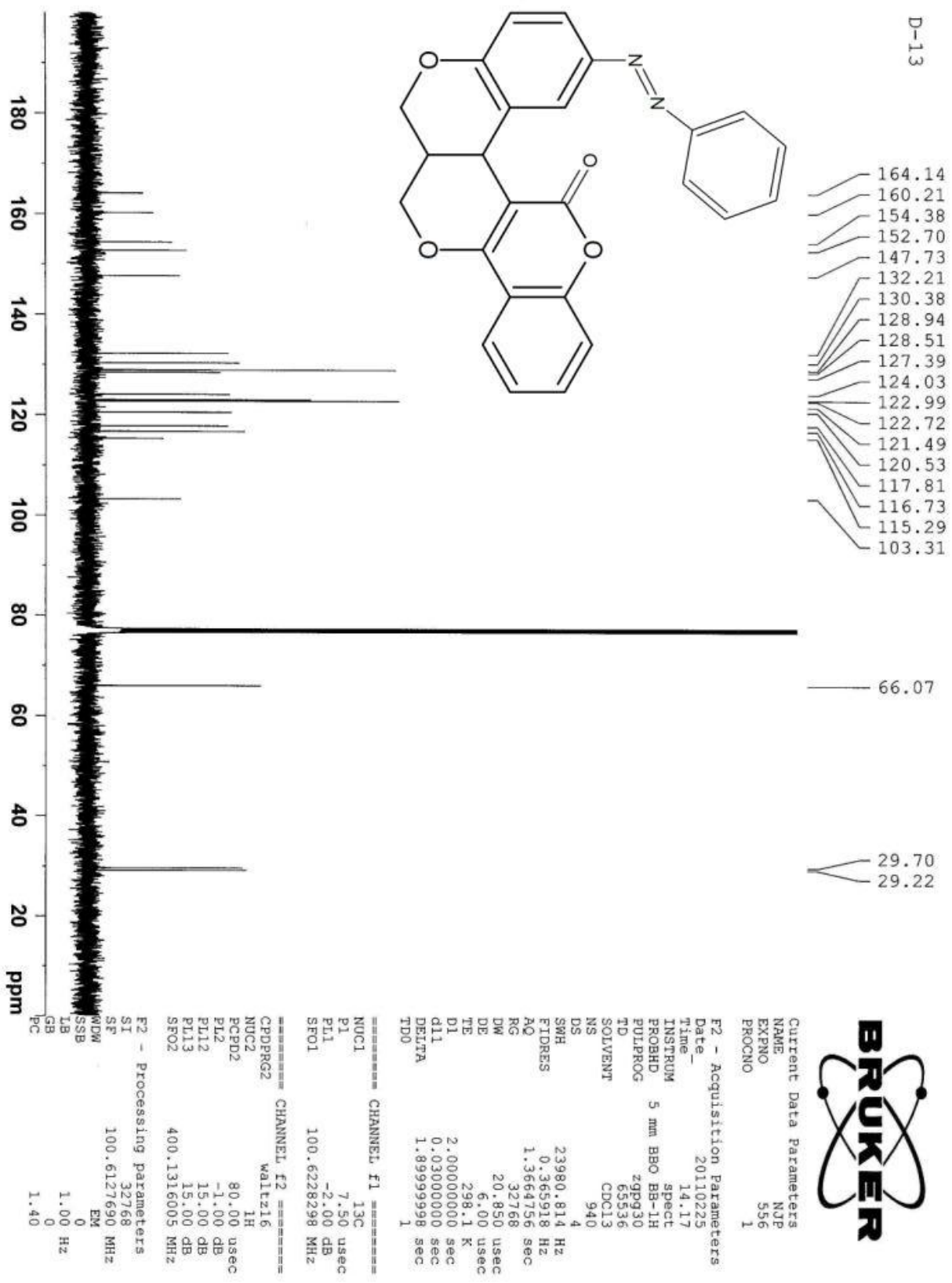
Compound 9b - ^{13}C NMR



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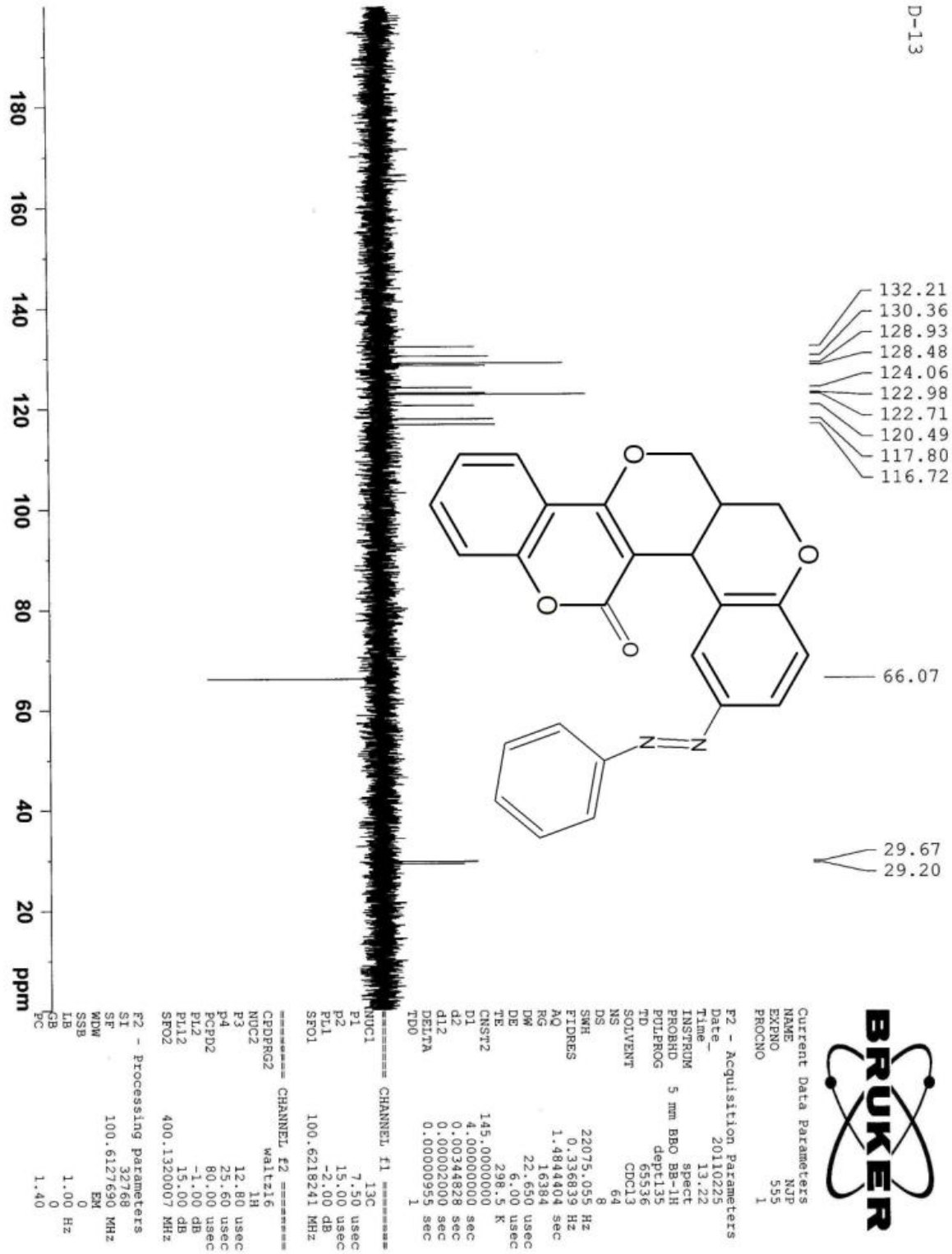
Compound 9b – DEPT 135



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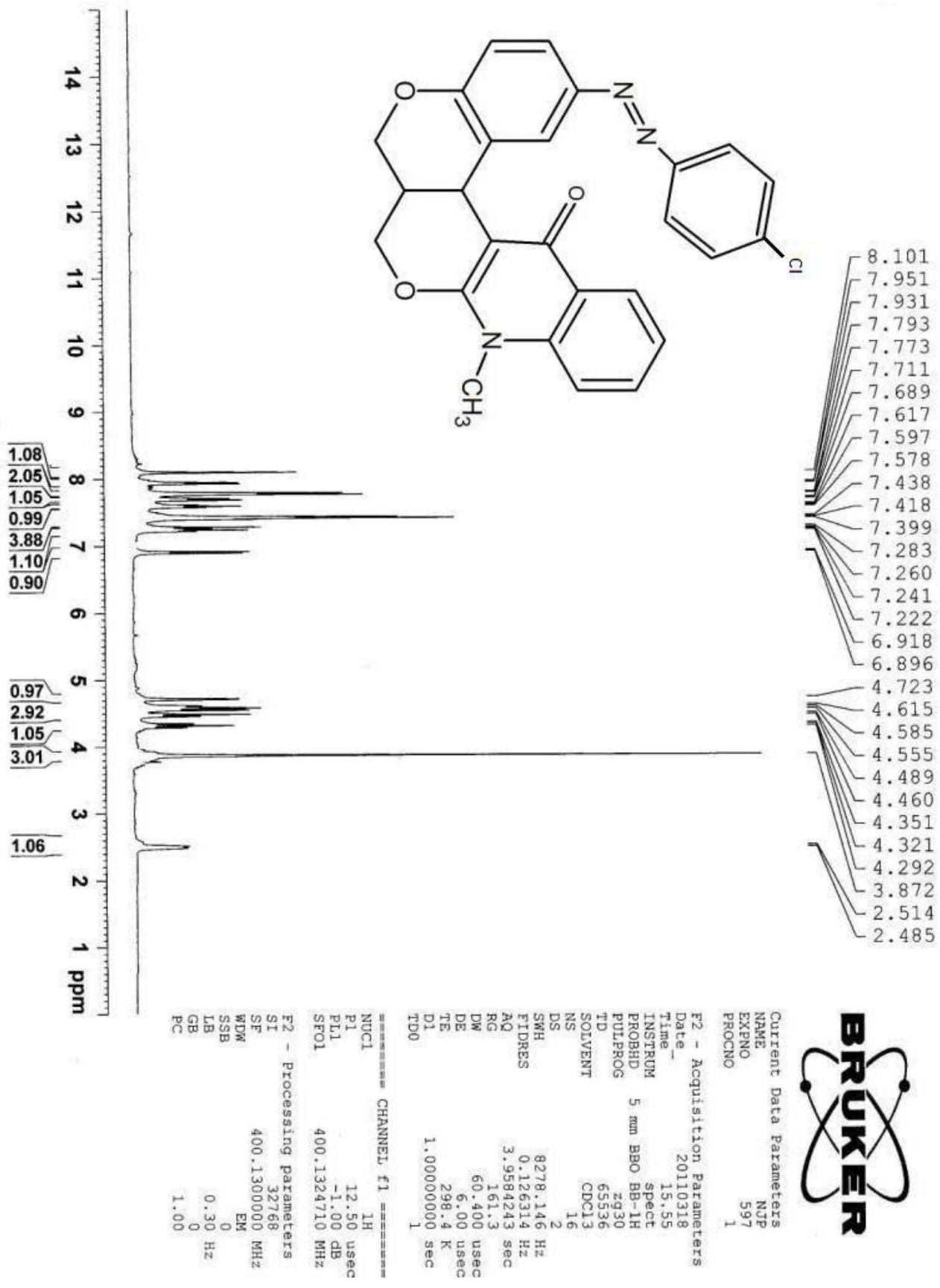
Compound 9c ^{1}H NMR



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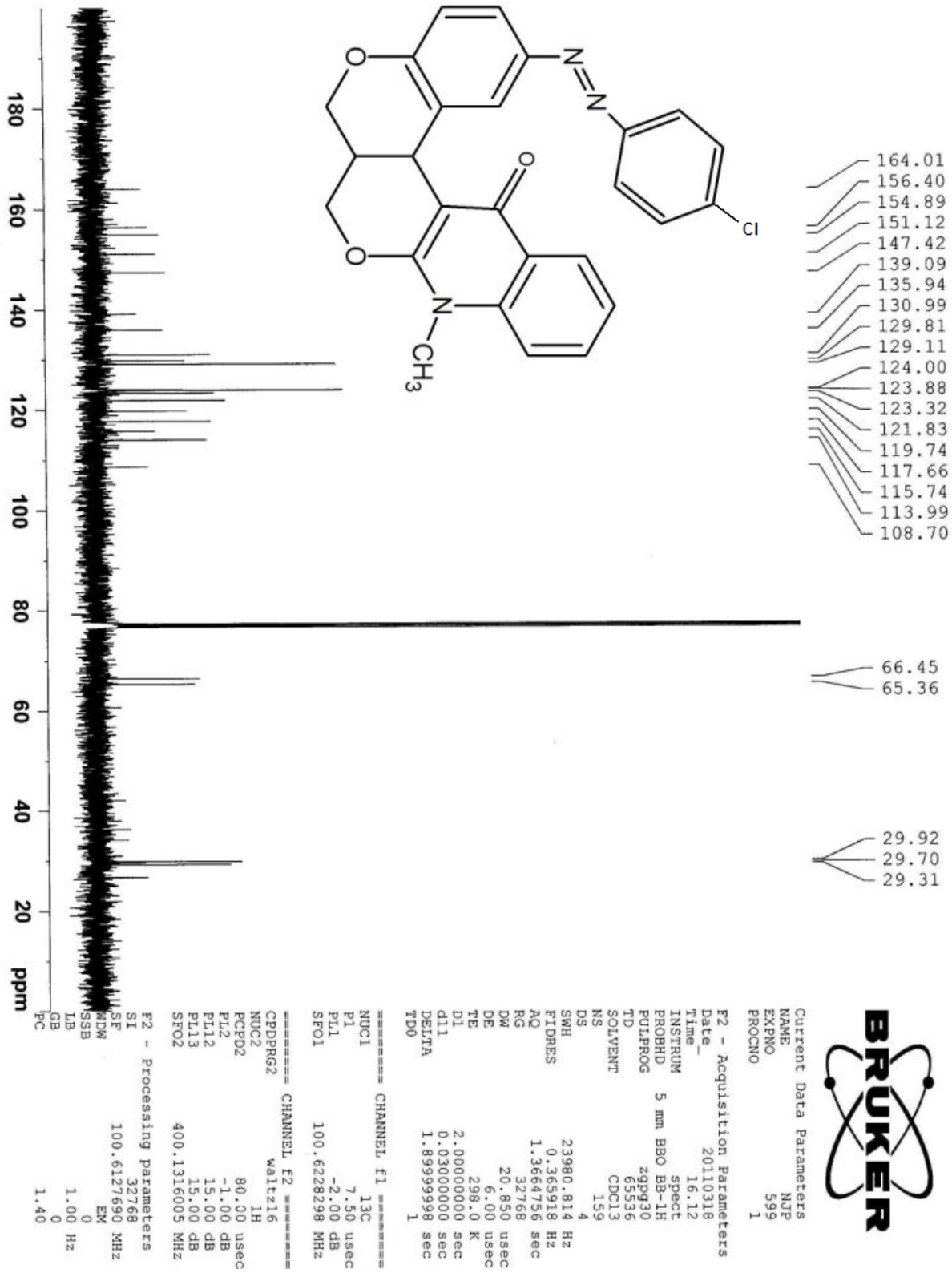
Compound 9c - ^{13}C NMR



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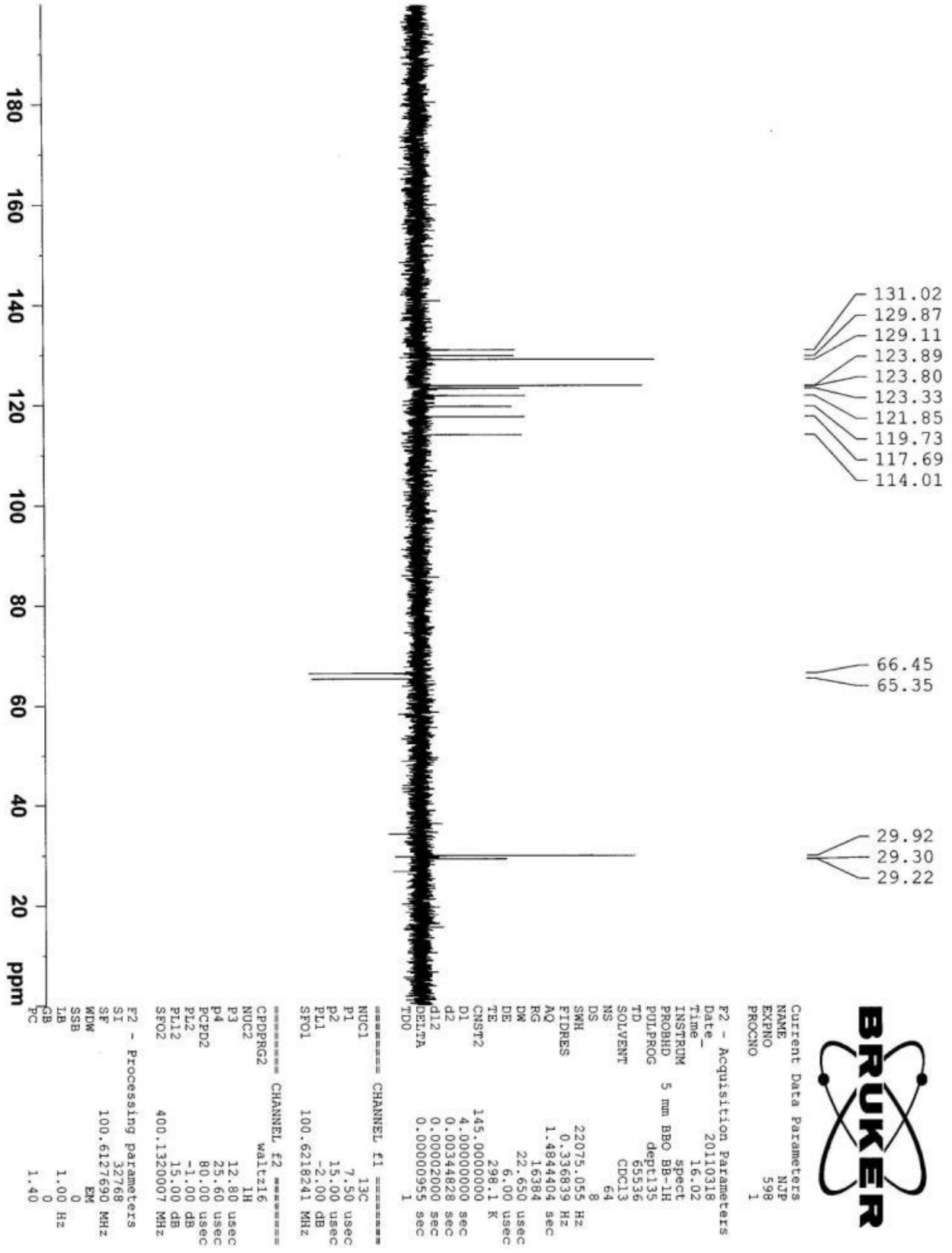
Compound 9c – DEPT 135



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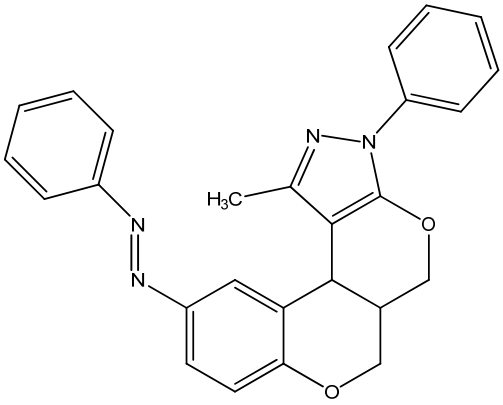
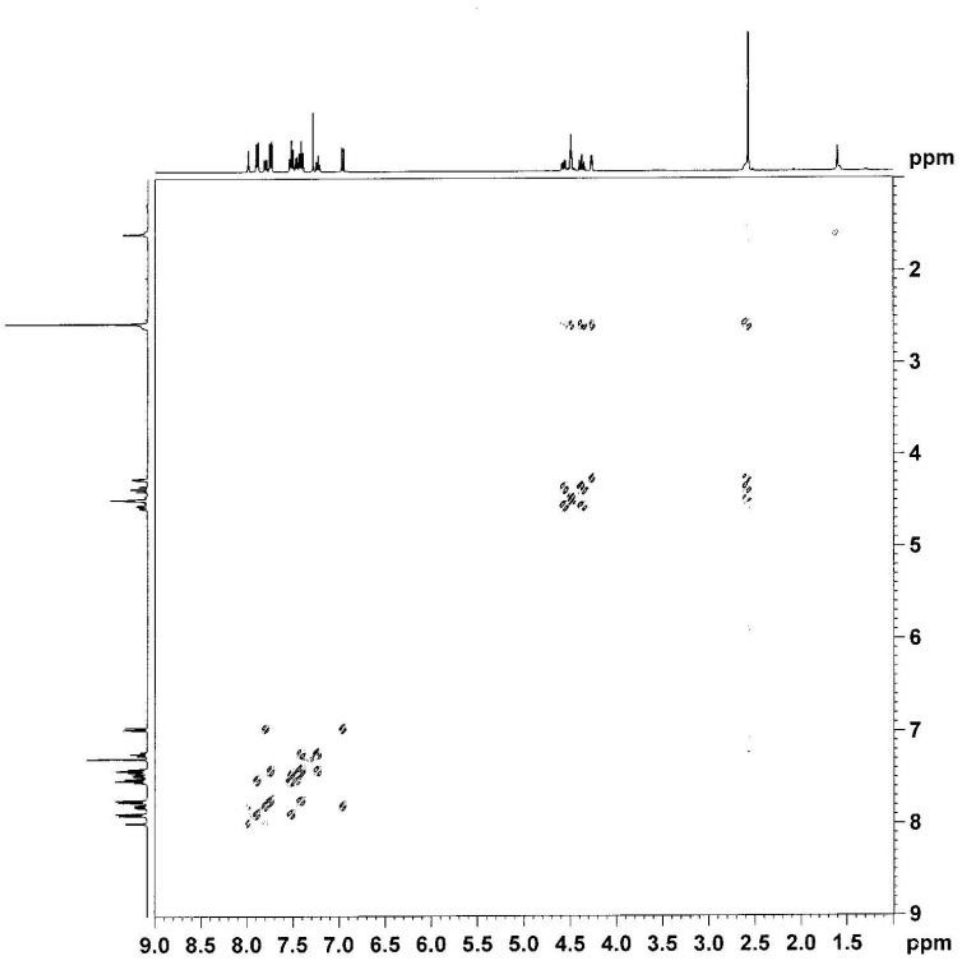
Compound 7a ^{1}H-^{1}H COSY





BRUKER

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PROCNO                   1

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TD                     2048
SOLVENT               CDCl3
NS                       16
DS                       16
SWH                3205.128 Hz
FIDRES             1.565004 Hz
AQ                 0.3195380 sec
RG                     574.7
DW                 156.000 usec
DE                   6.00 usec
TE                   298.2 K
d0              0.00014072 sec
D1              2.00000000 sec
d13             0.00000400 sec
IN0             0.00031200 sec
ST1CNT                  256

======== CHANNEL f1 ========
NUC1                    1H
P1                  12.00 usec
PL1                 -1.00 dB
SFO1          400.1319998 MHz

F1 - Acquisition parameters
ND0                      1
TD                     512
SFO1               400.132 MHz
FIDRES             6.260016 Hz
SW                 8.010 ppm
FnMODE         States-TPPI

F2 - Processing parameters
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SSB                       2
LB                   0.00 Hz
GB                        0
PC                     1.40

F1 - Processing parameters
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WDW                   QSINE
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GB                        0
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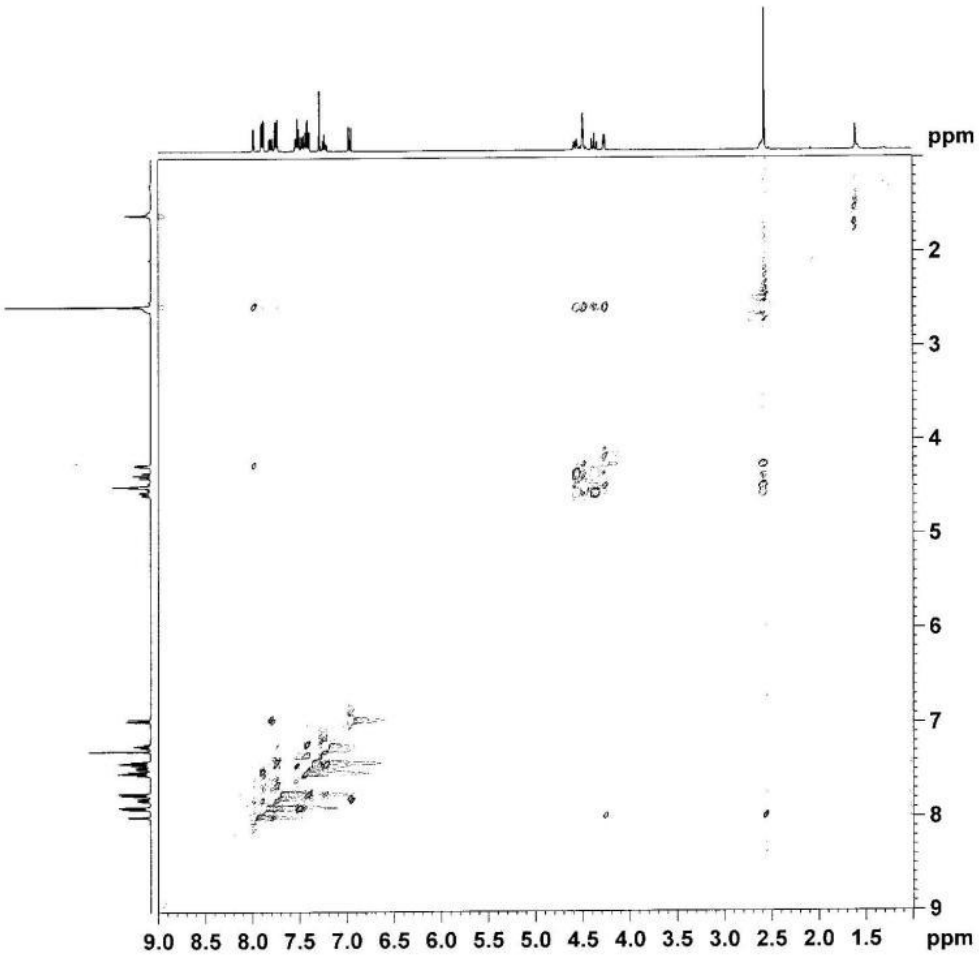
Compound 7a NOESY



BRUKER

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Current Data Parameters
NAME                  NJP
EXPNO                 588
PROCNO                  1

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TD                  2048
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NS                    32
DS                    16
SWH              3205.128 Hz
FIDRES           1.565004 Hz
AQ               0.3195380 sec
RG                 574.7
DW               156.000 usec
DE                 6.00 usec
TE                 298.2 K
d0               0.00014072 sec
D1               2.00000000 sec
D8               0.50000000 sec
IN0              0.00031200 sec
ST1CNT                256

======== CHANNEL f1 ========
NUC1                   1H
P1                 12.00 usec
PL1                -1.00 dB
SFO1         400.1319998 MHz

F1 - Acquisition parameters
ND0                    1
TD                   512
SFO1             400.132 MHz
FIDRES          6.260016 Hz
SW                 8.010 ppm
FnMODE       States-TPPI

F2 - Processing parameters
SI                  1024
SF           400.1300000 MHz
WDW                 QSINE
SSB                    2
LB                  0.00 Hz
GB                     0
PC                  1.00

F1 - Processing parameters
SI                  1024
MC2          States-TPPI
SF           400.1300000 MHz
WDW                 QSINE
SSB                    2
LB                  0.00 Hz
GB                     0
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