Supporting Information

An unprecedented deoxygenation protocol of benzylic alcohols using *bis*(1-benzotriazolyl)methanethione

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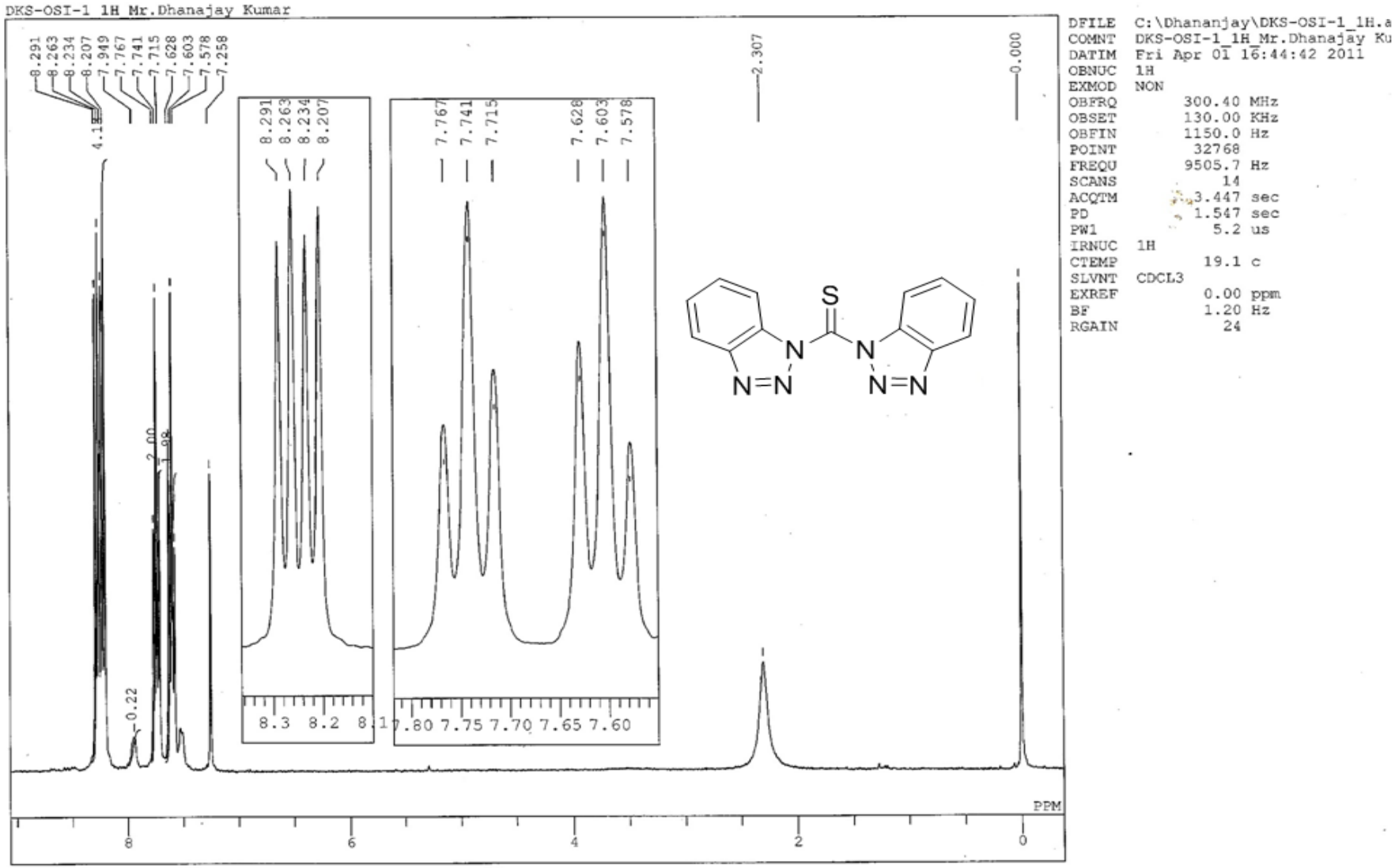
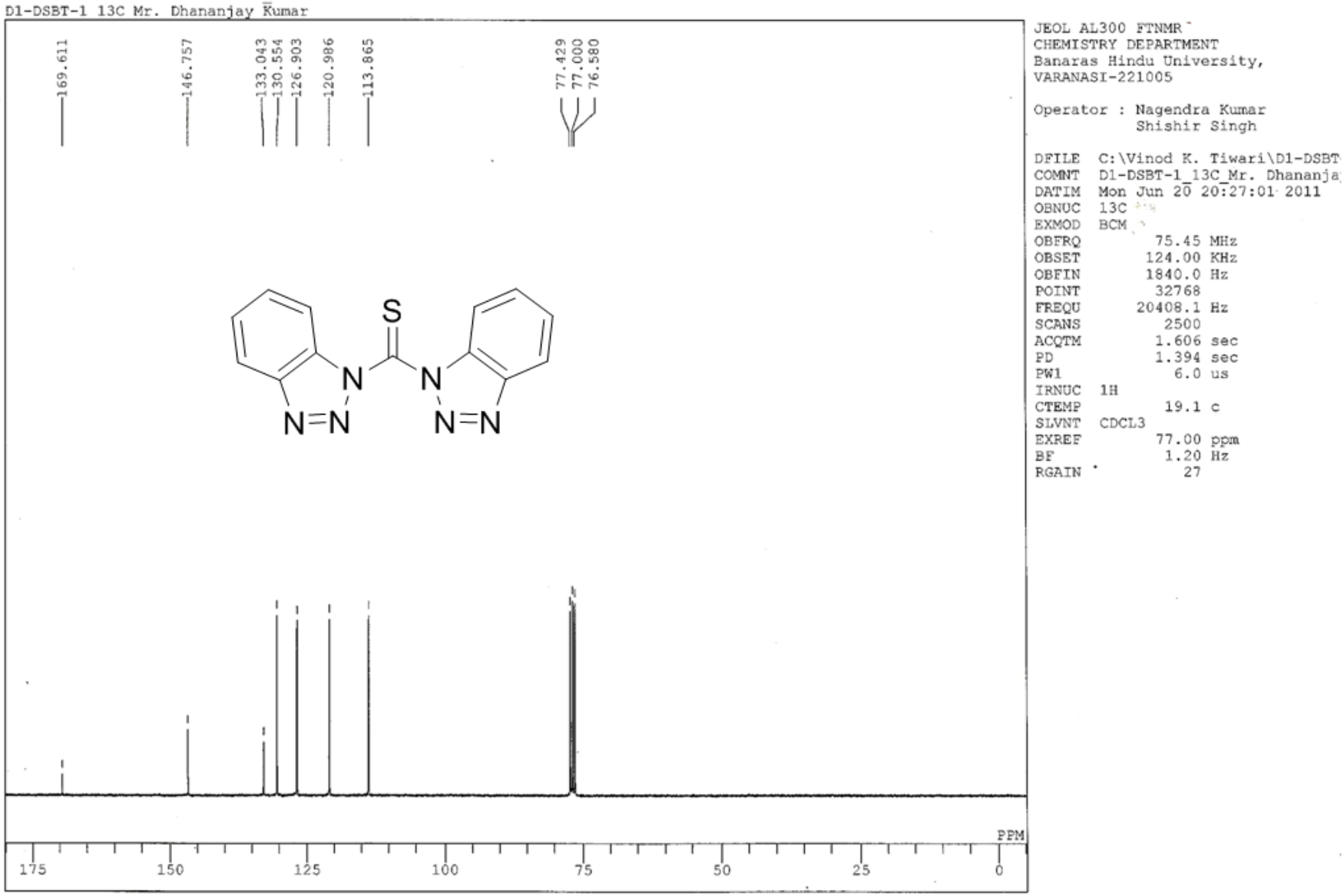


Spectrum 1: 300 MHz ^{1}H NMR of compound **2**



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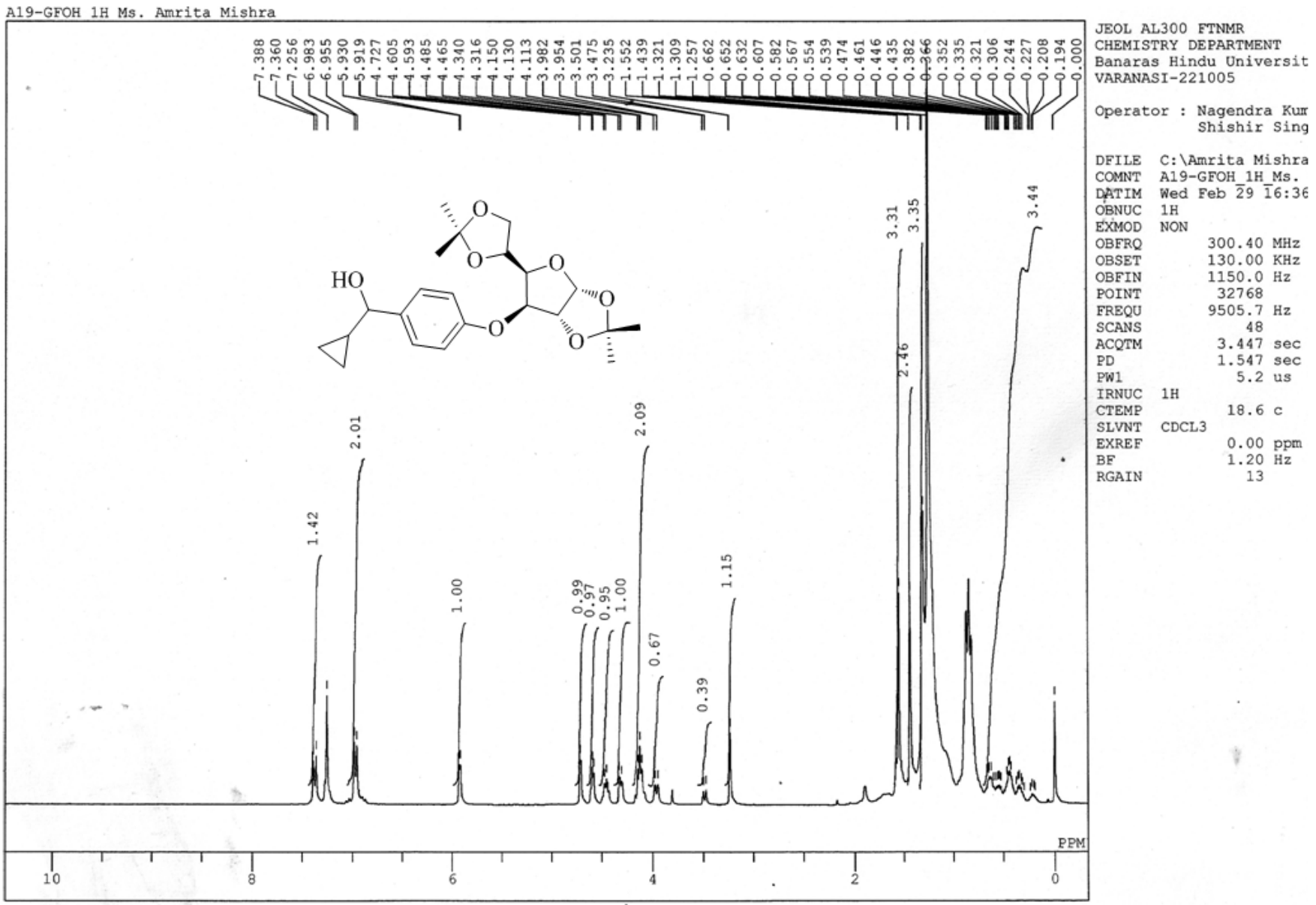

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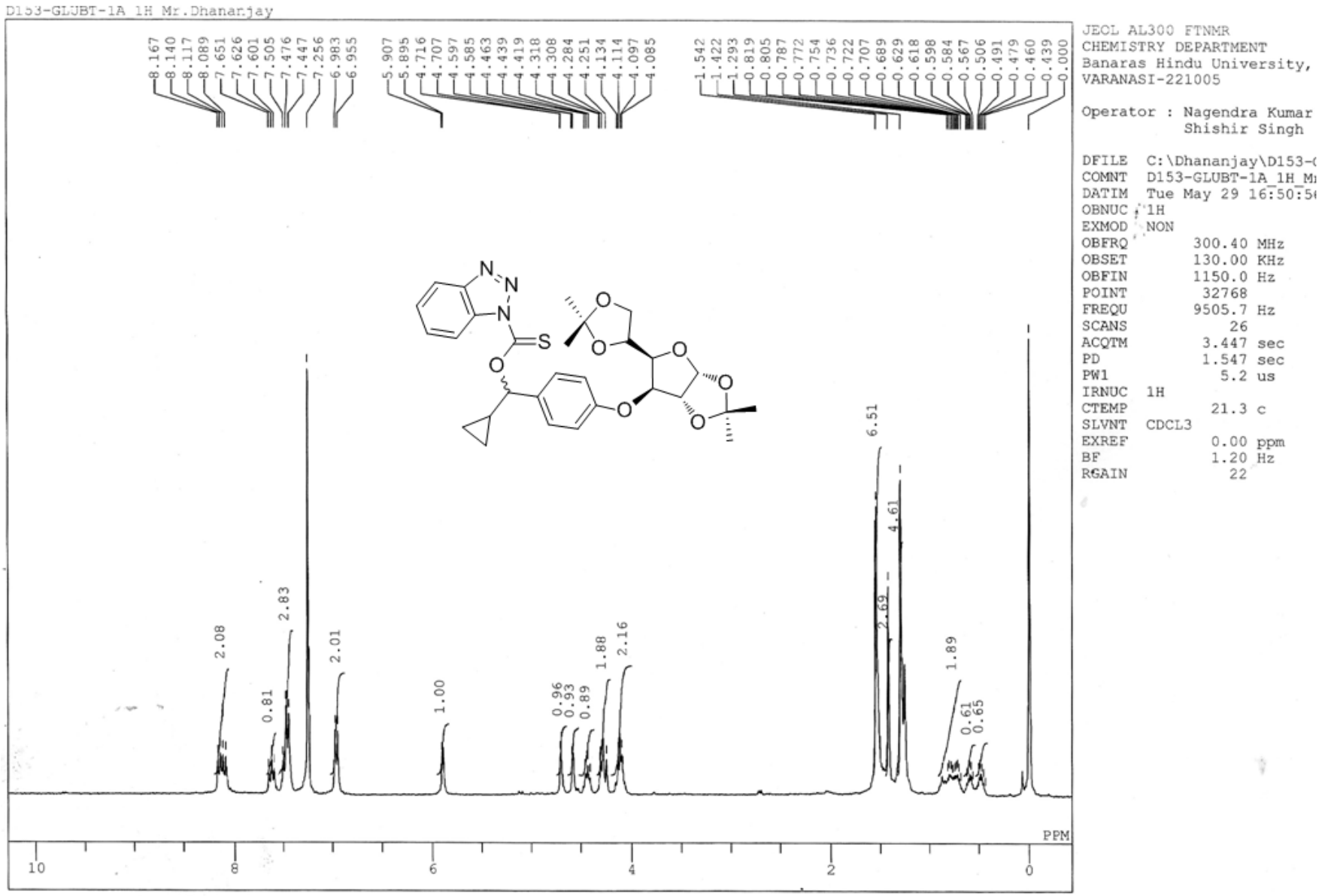

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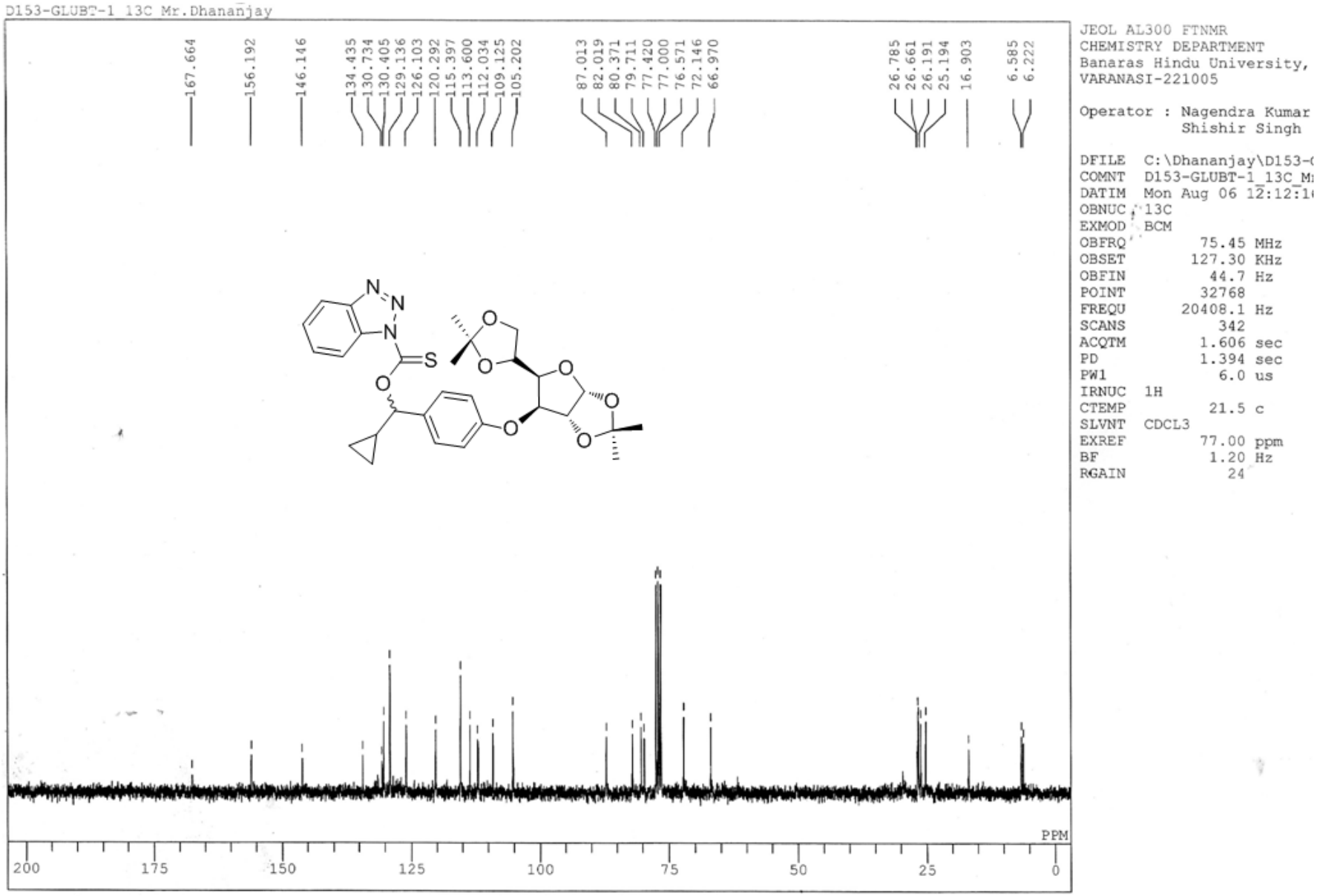

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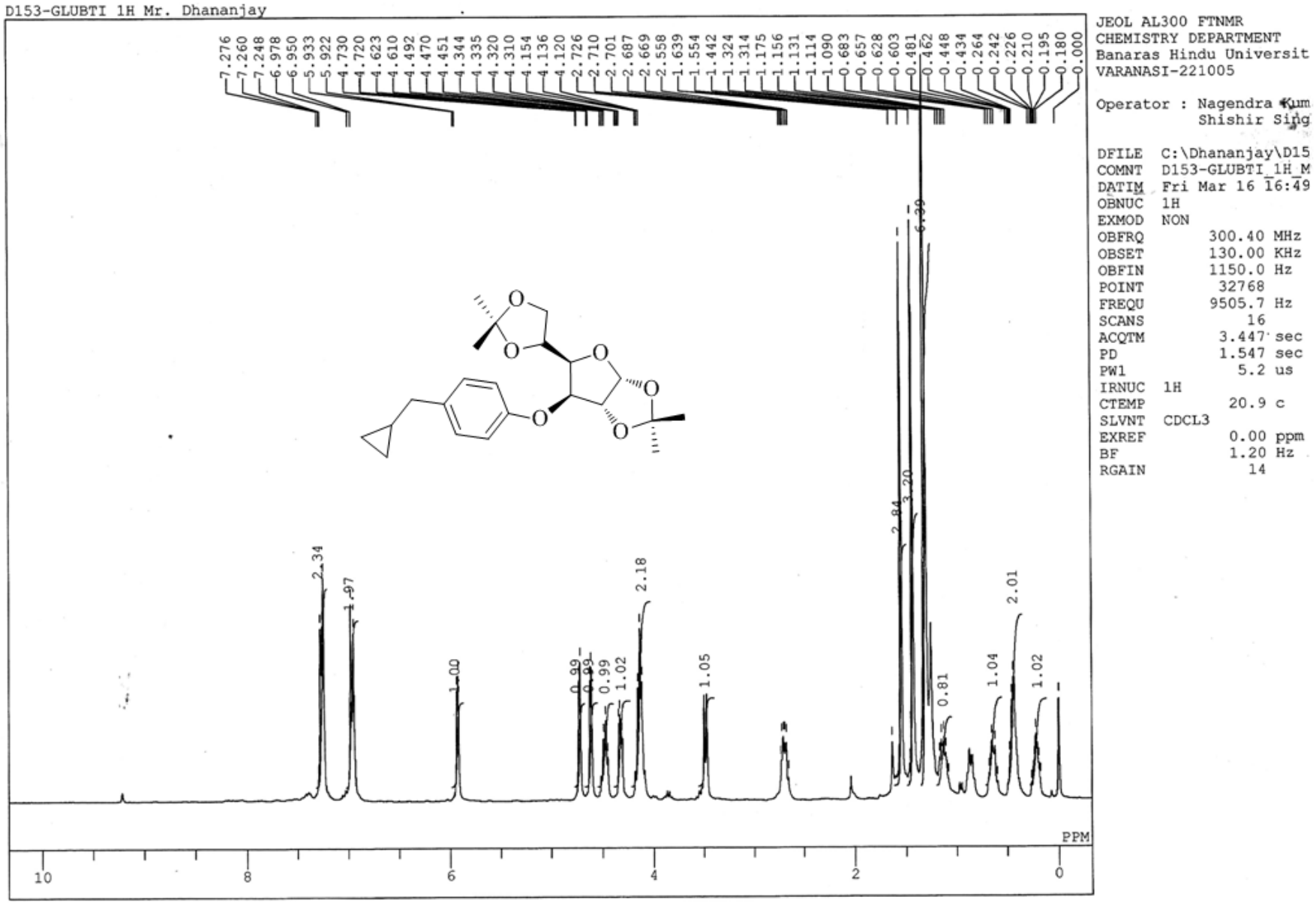

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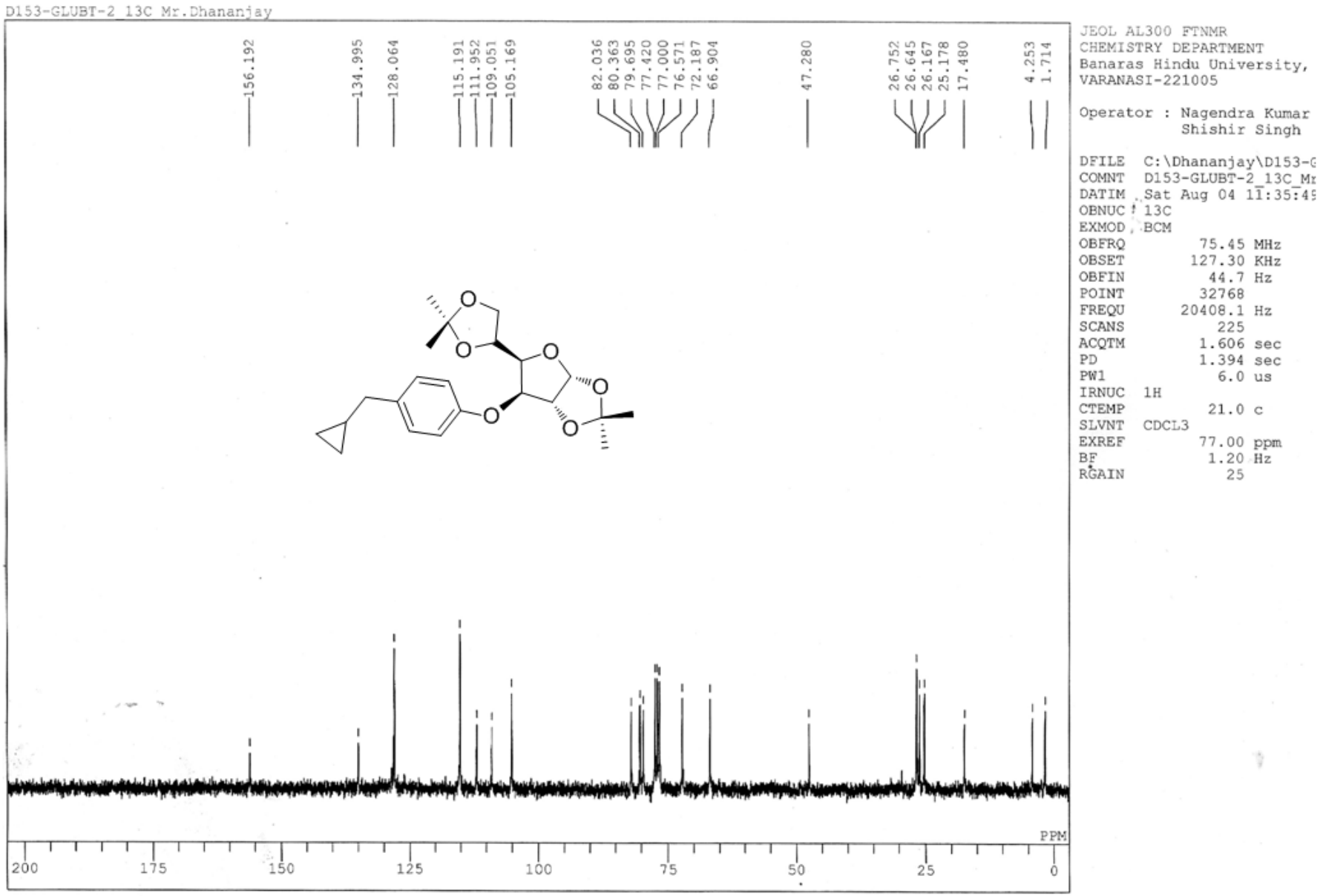

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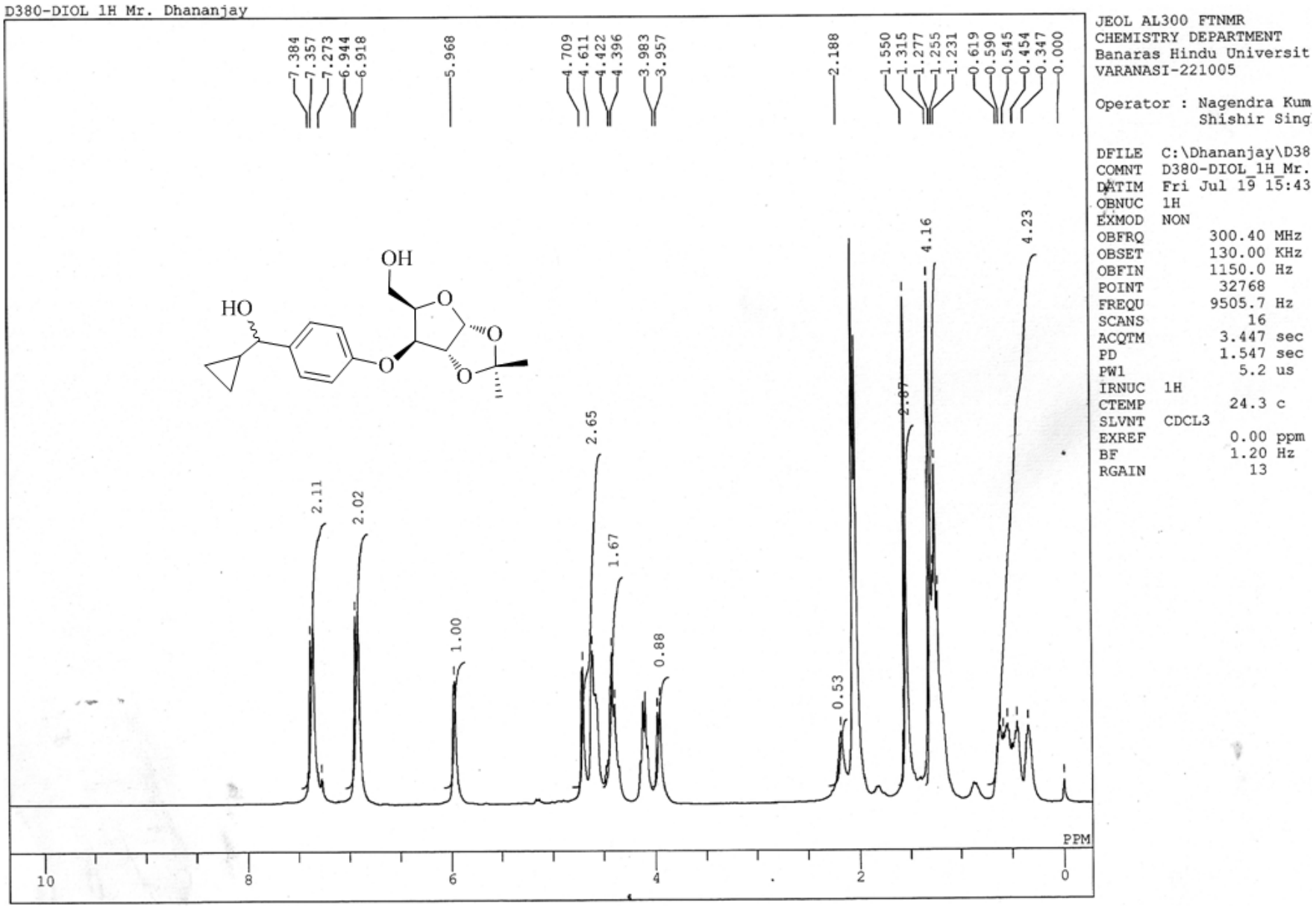

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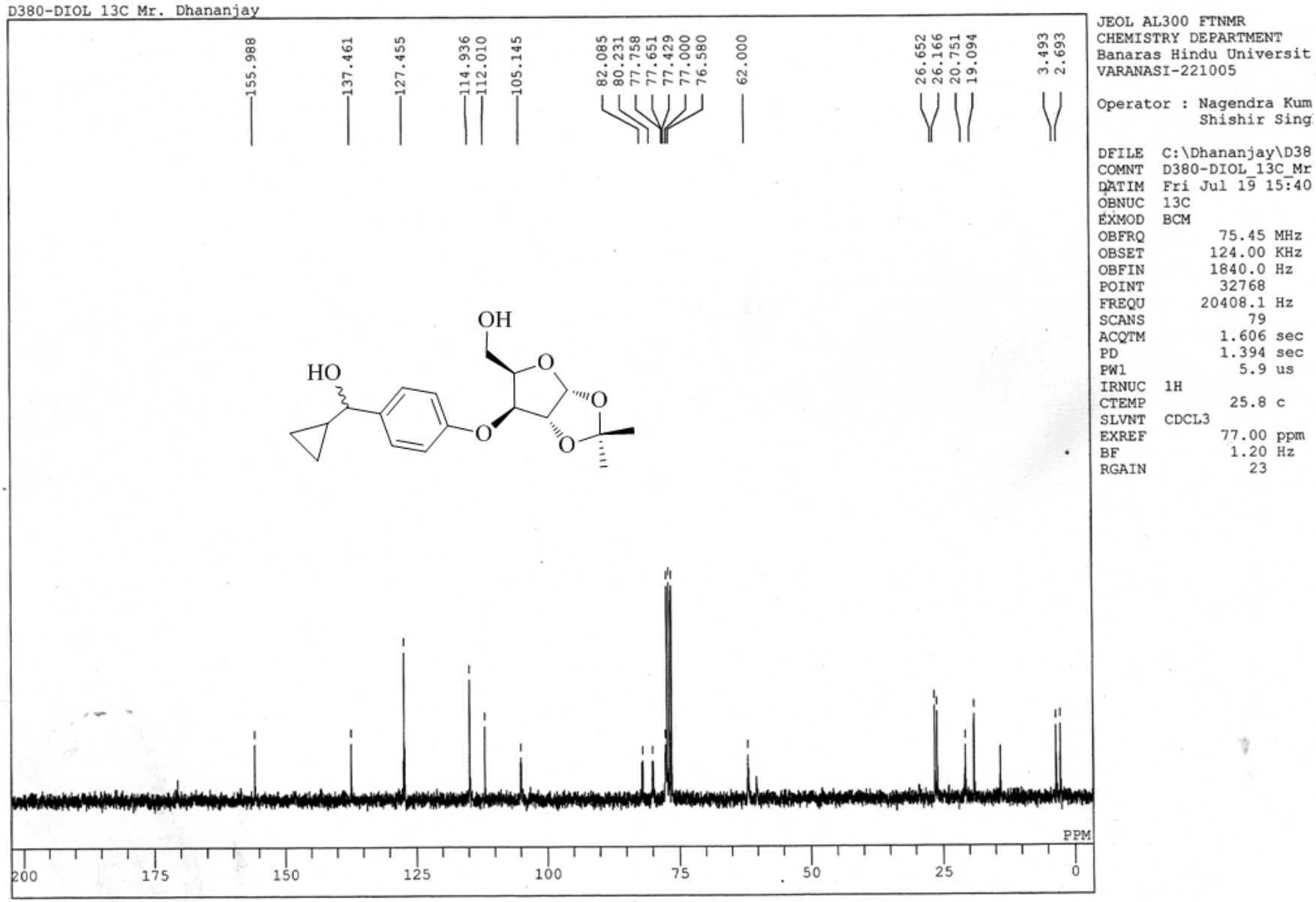

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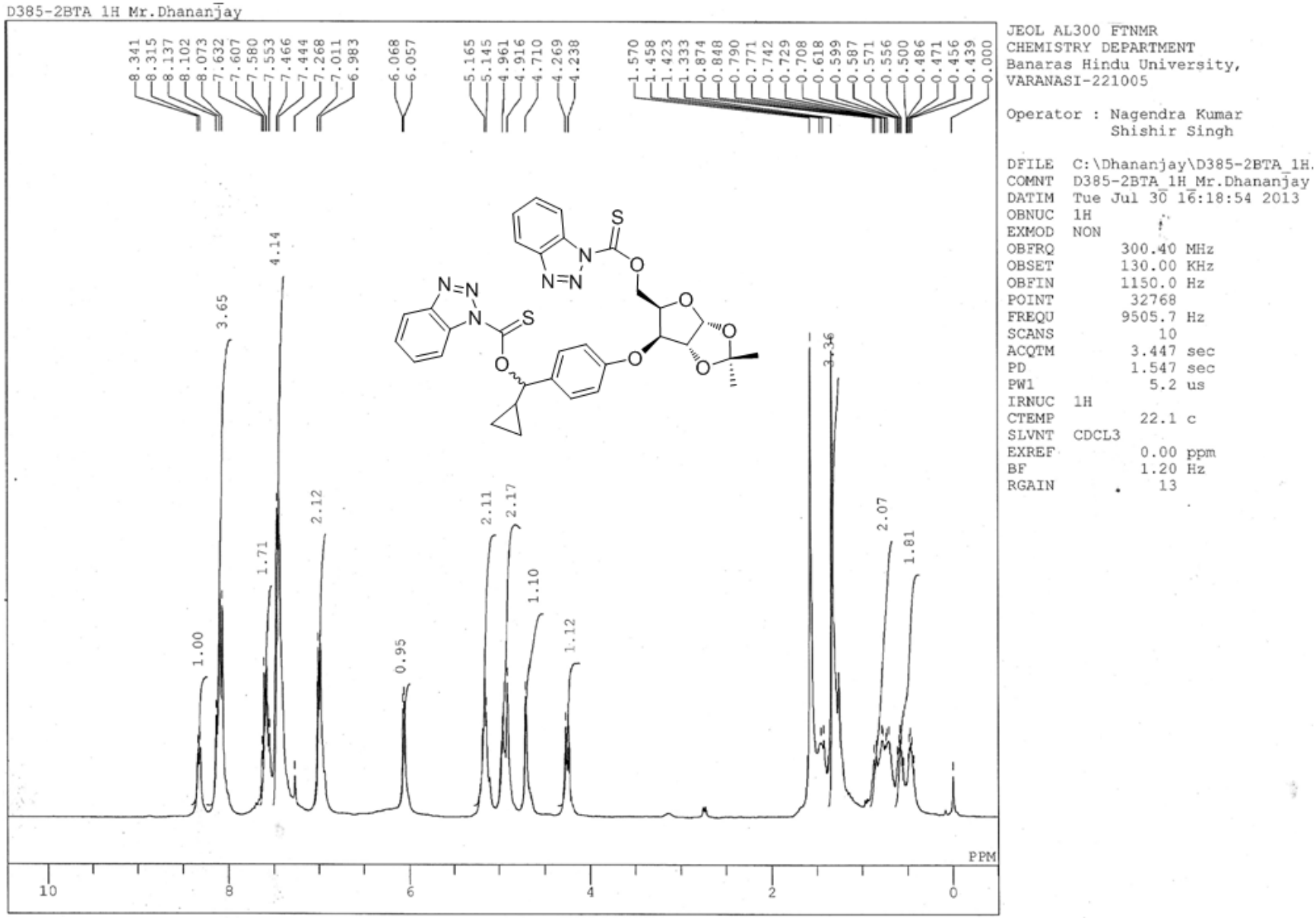

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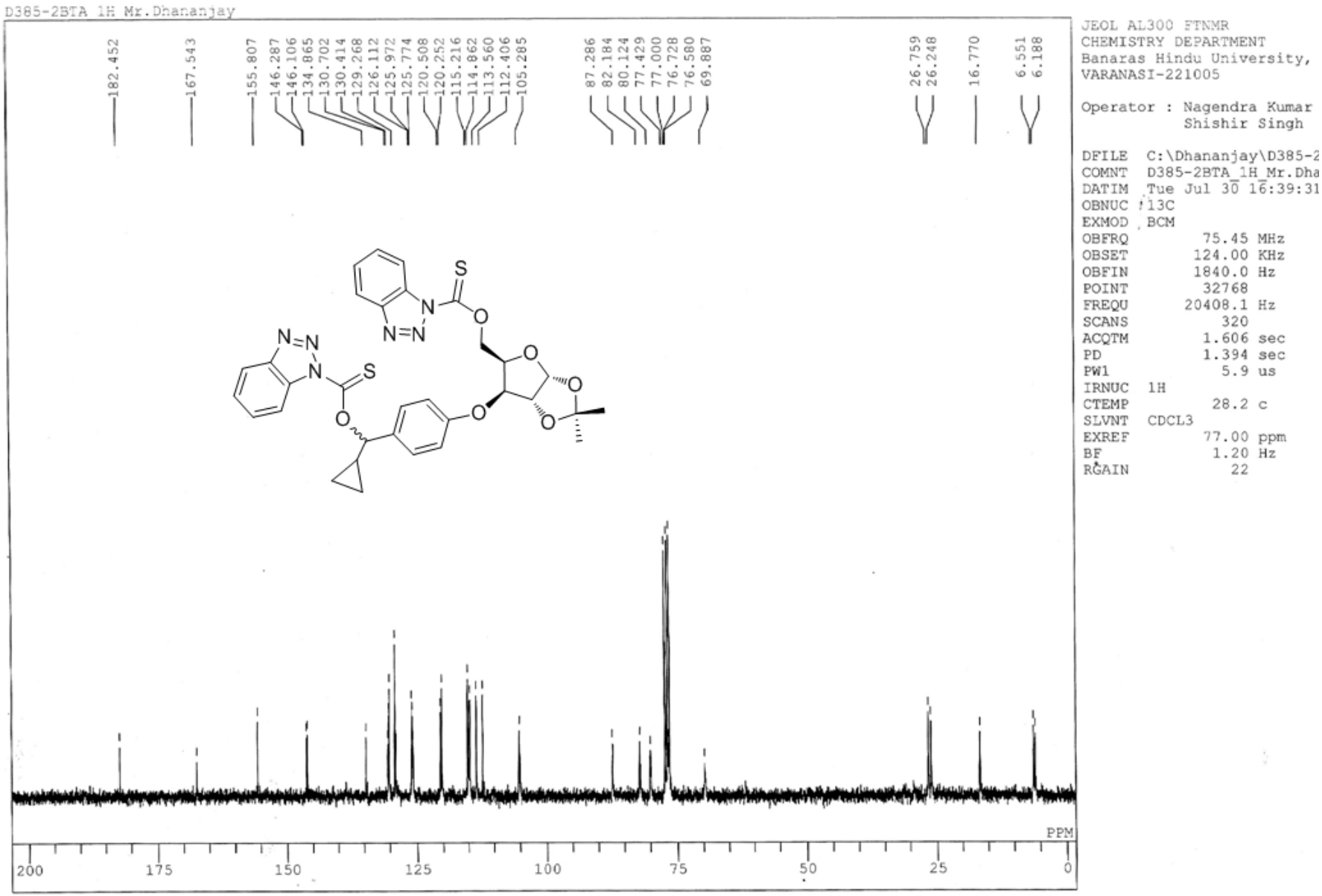

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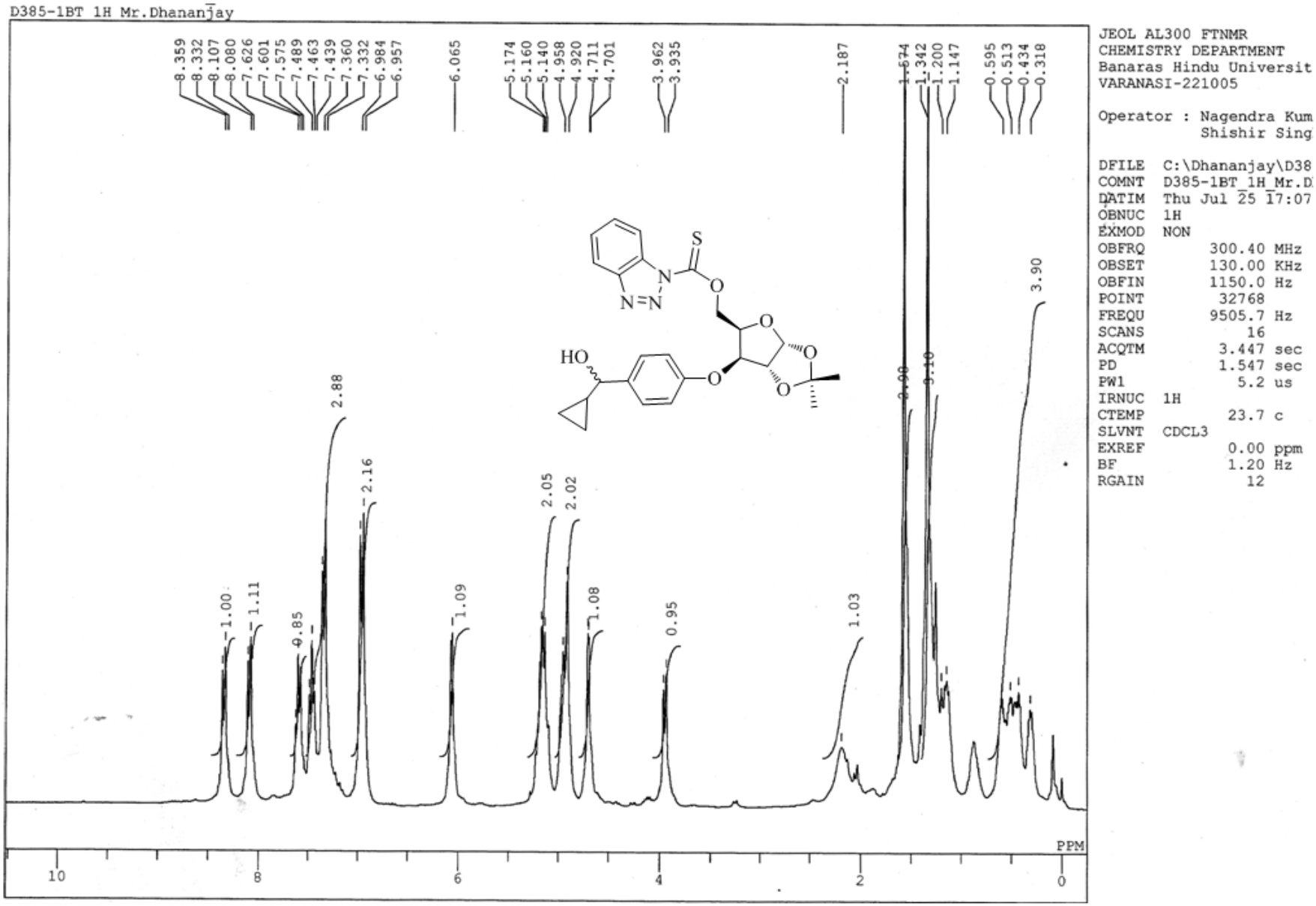

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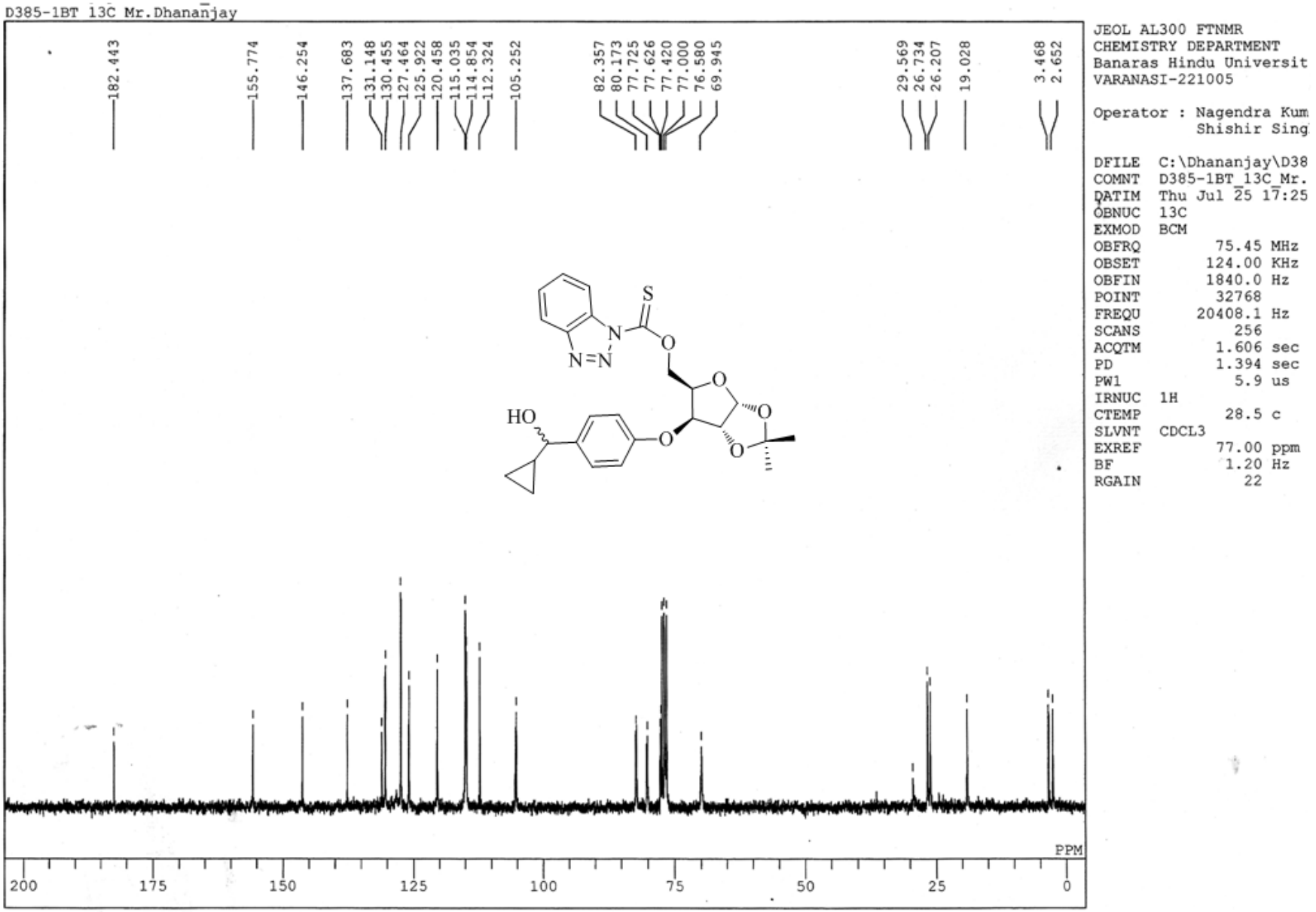
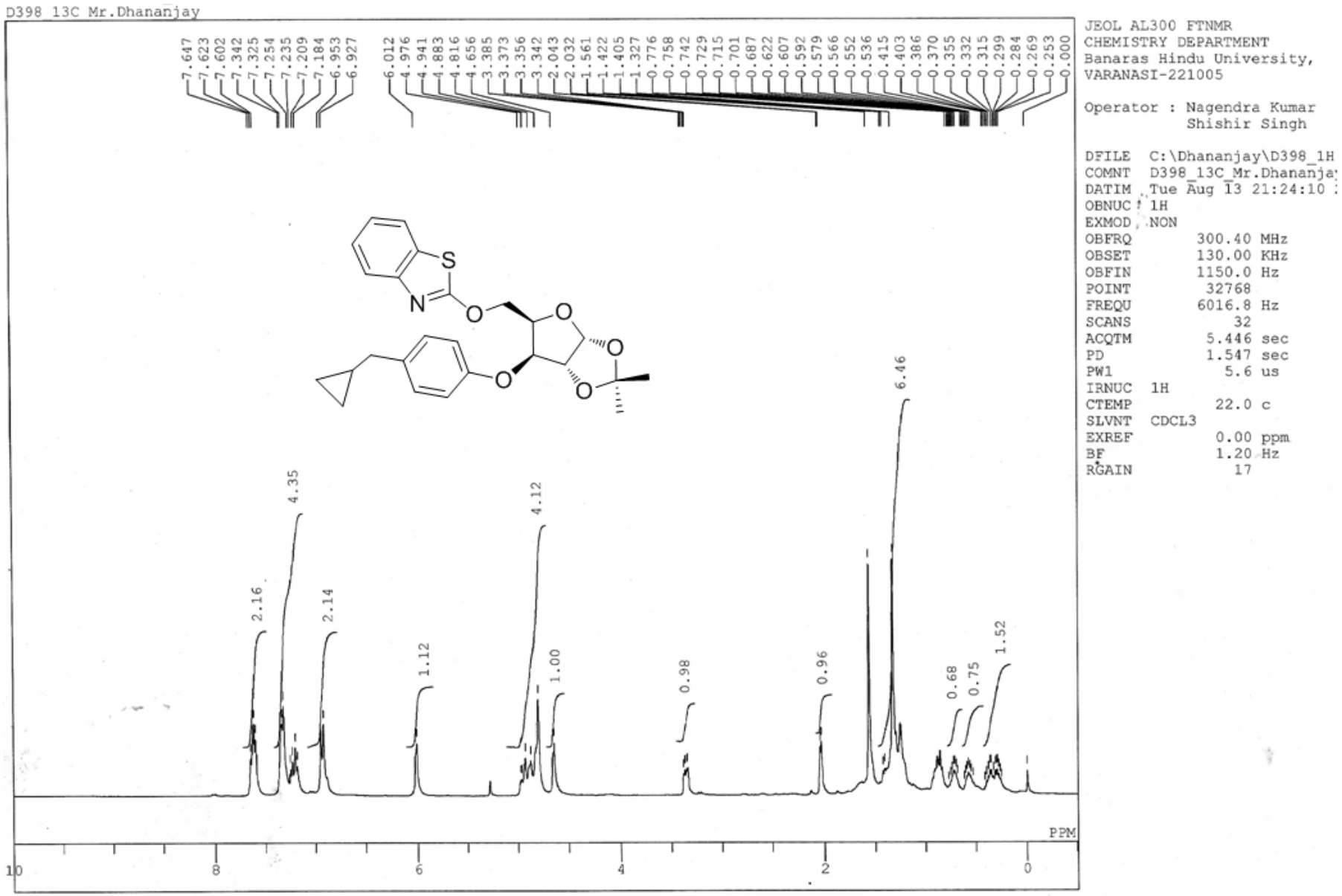


Spectrum 13: 75 MHz ^{13}C NMR of compound **18**



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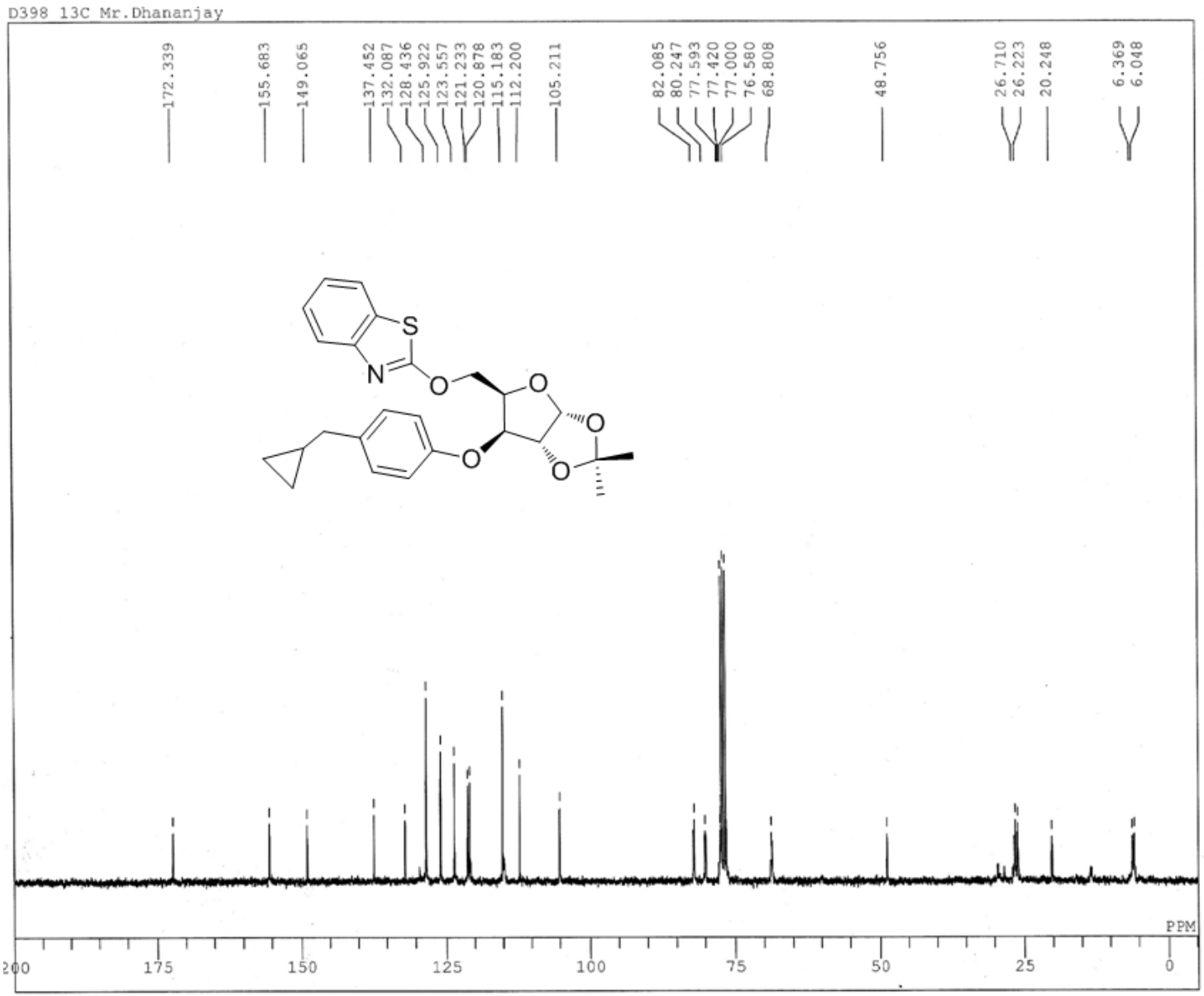

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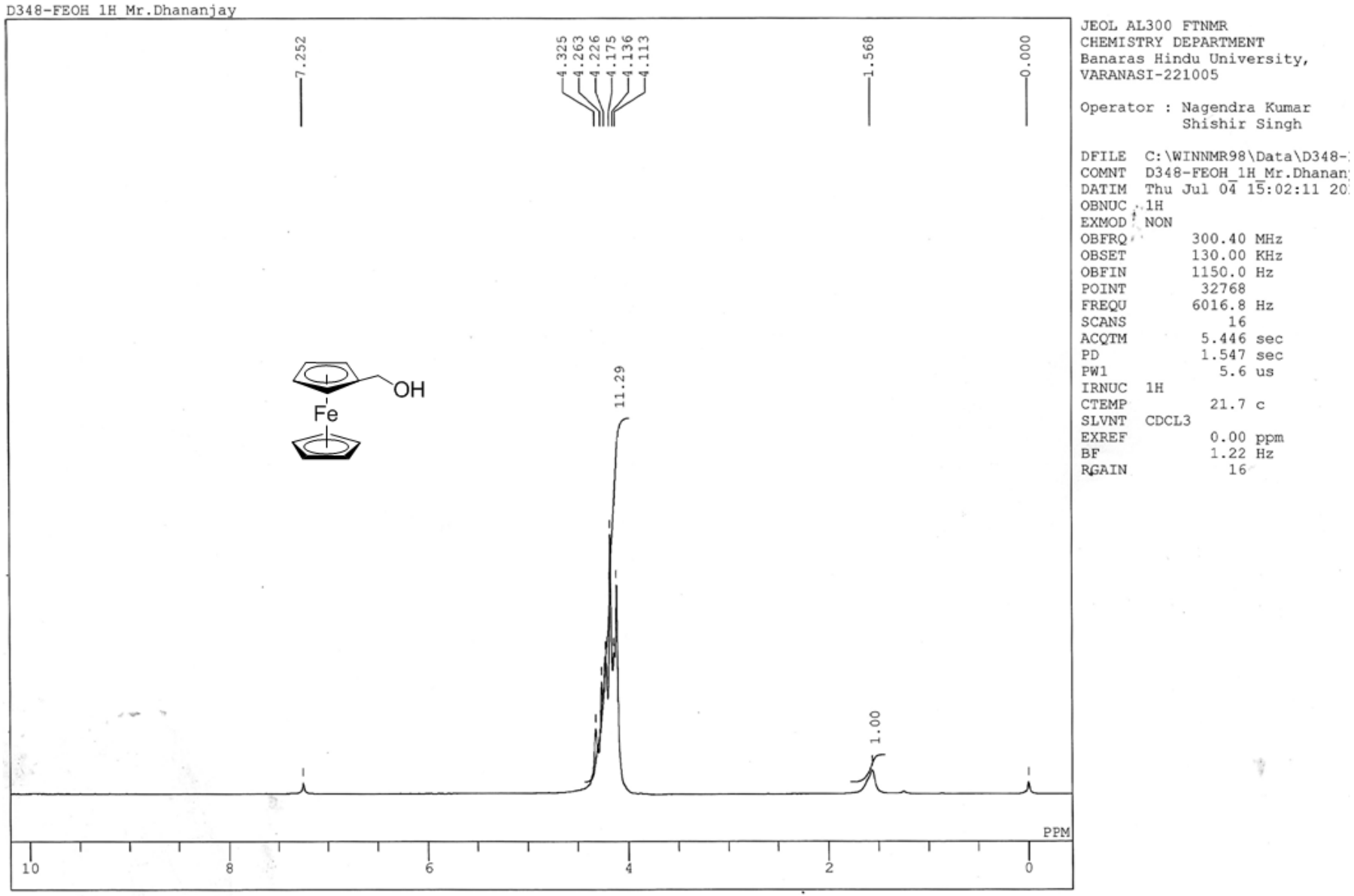

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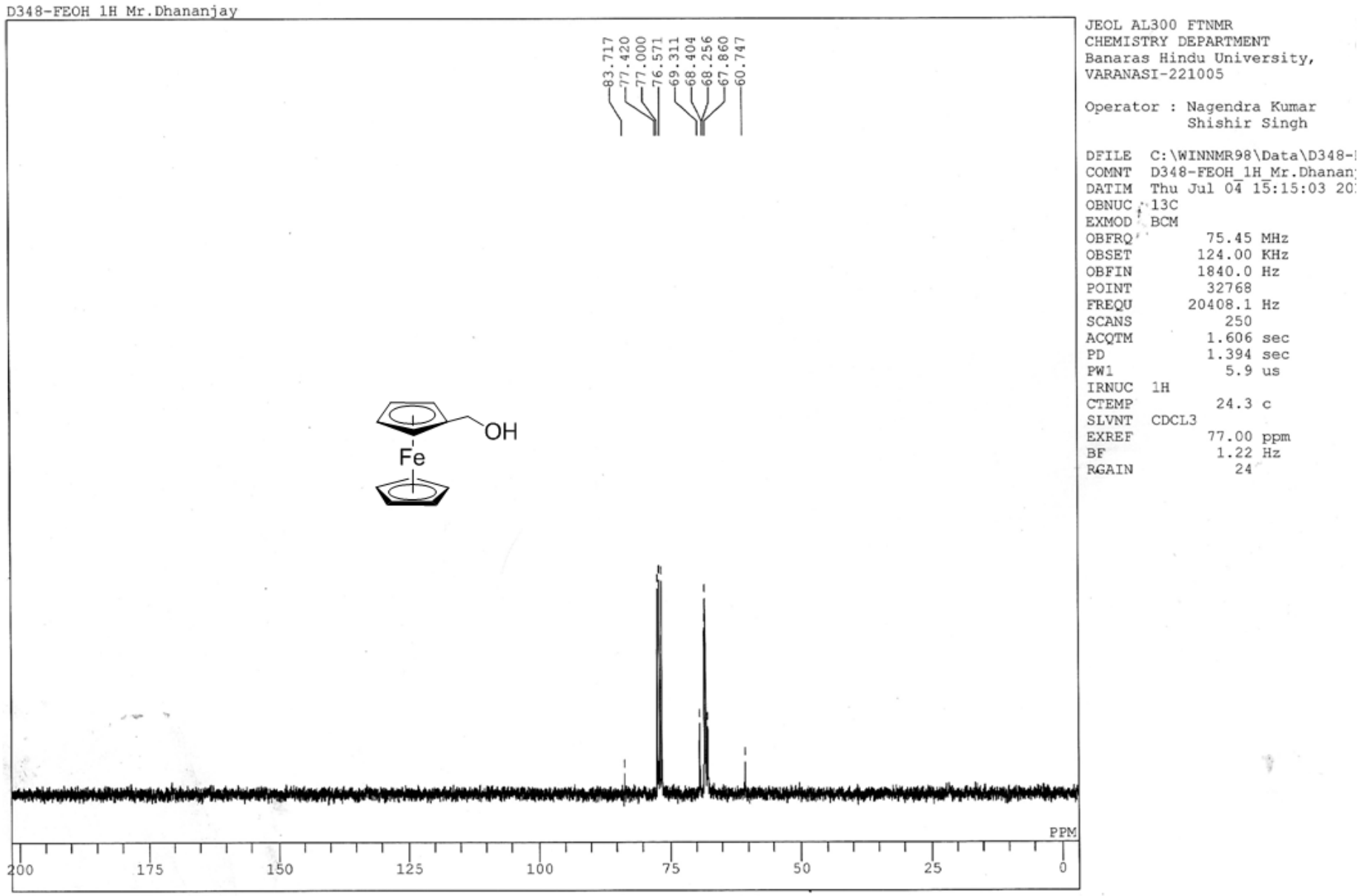

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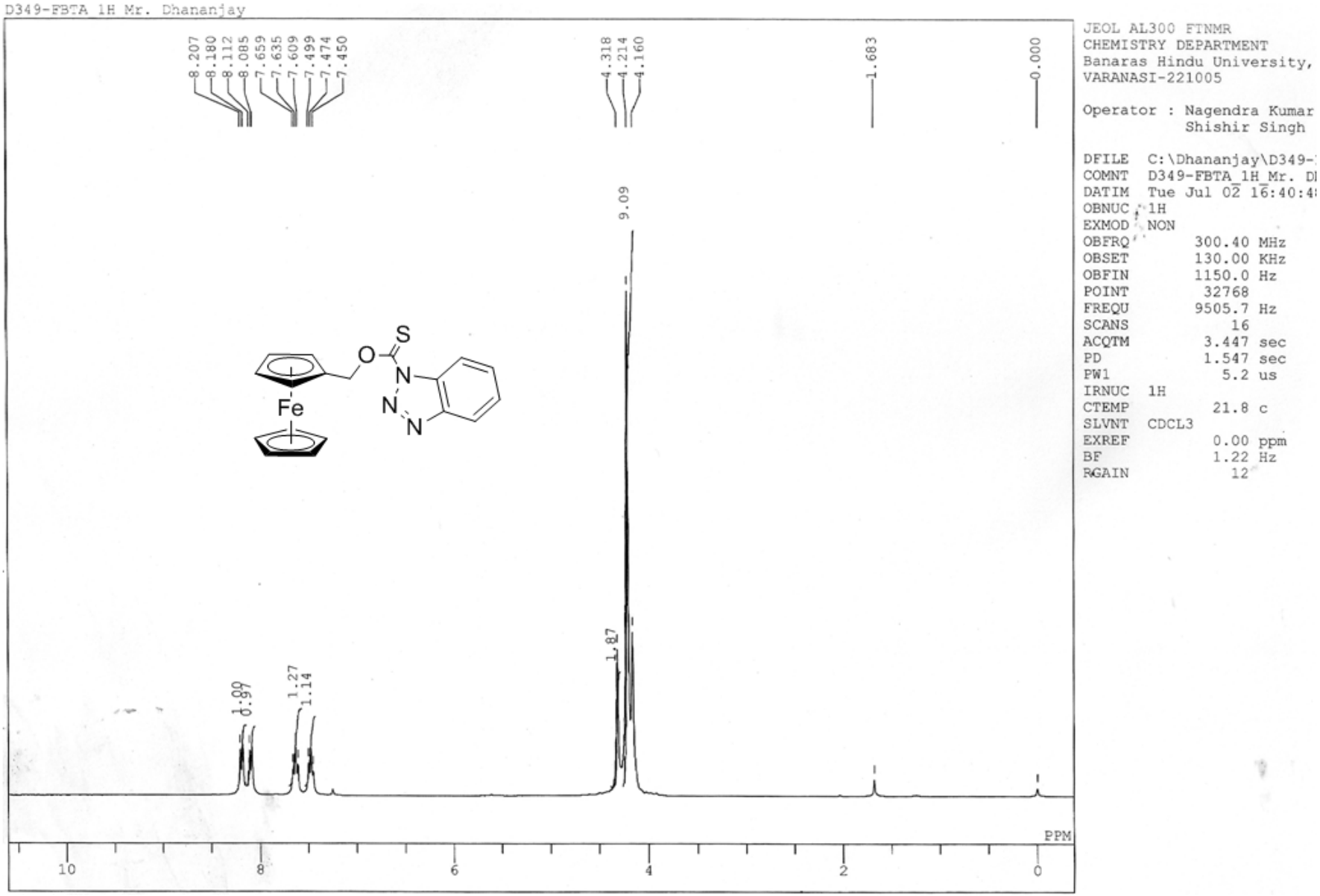

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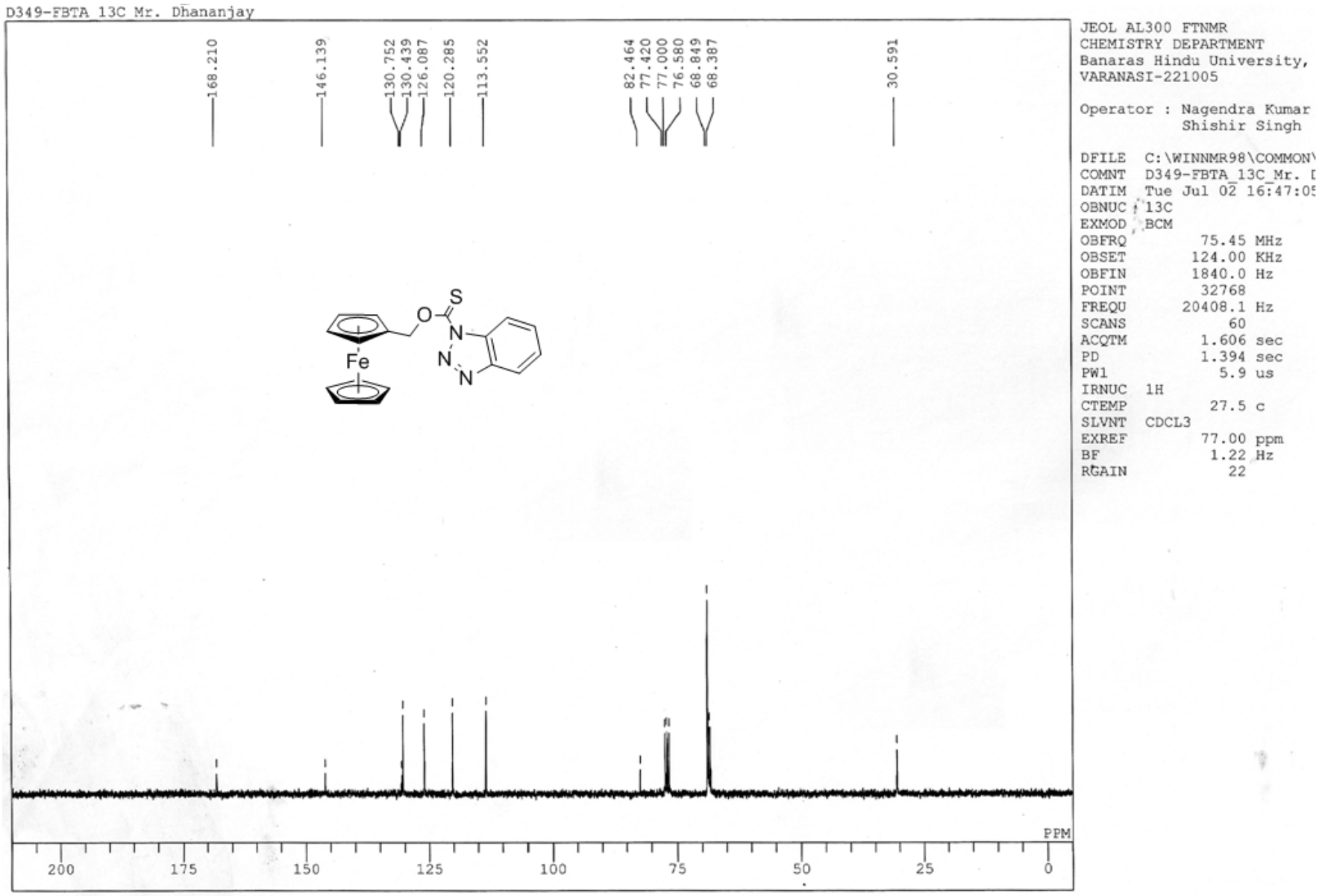

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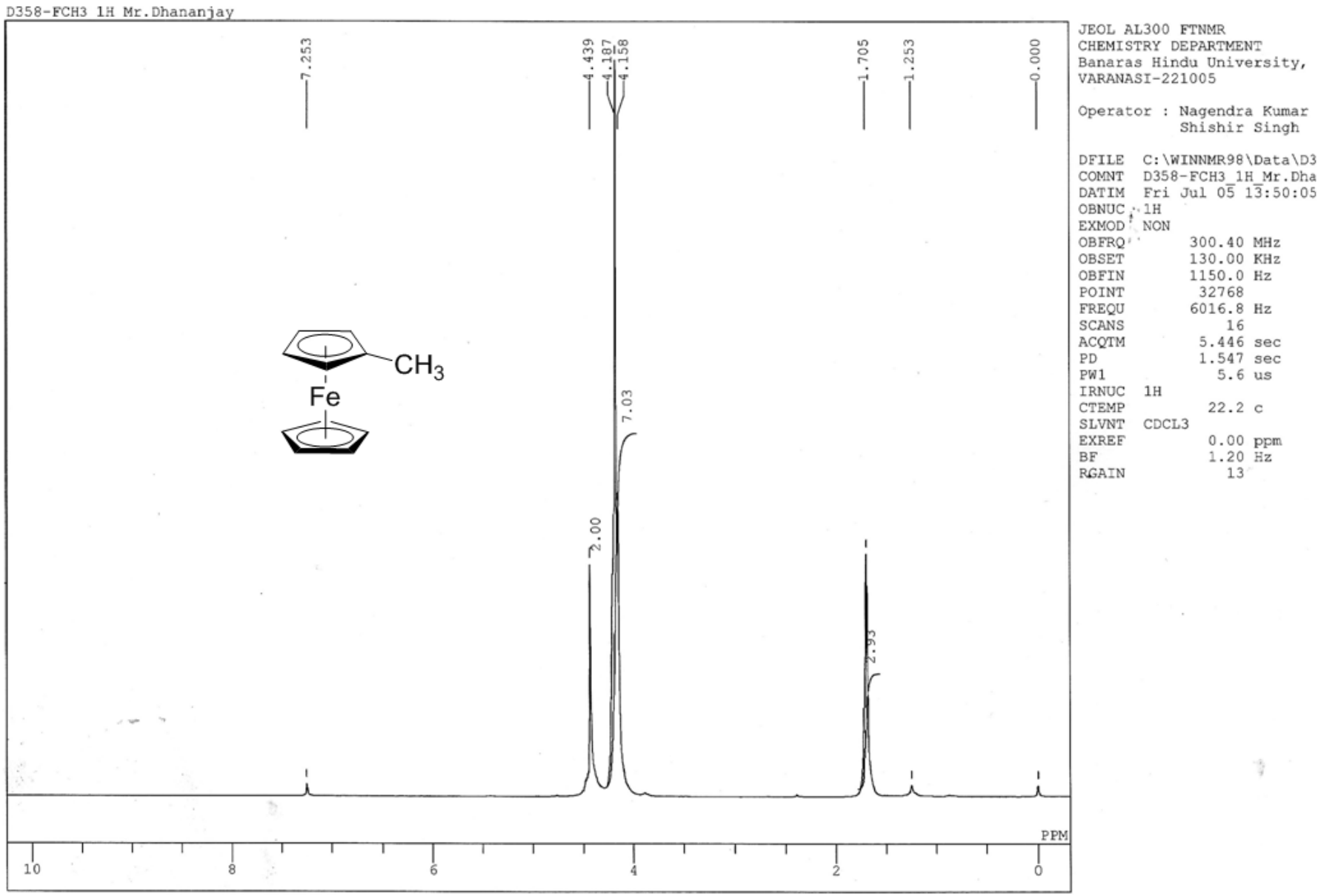

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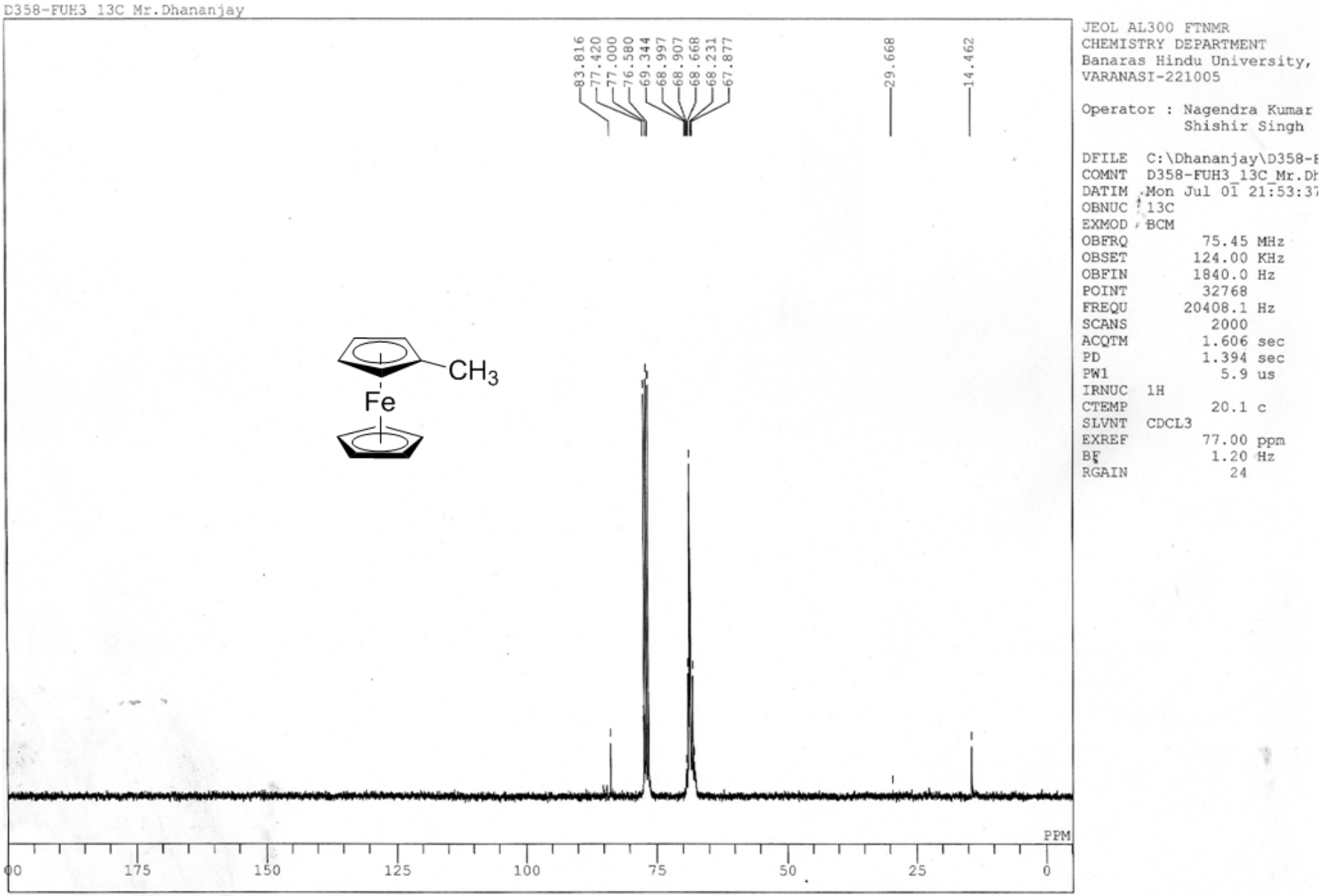

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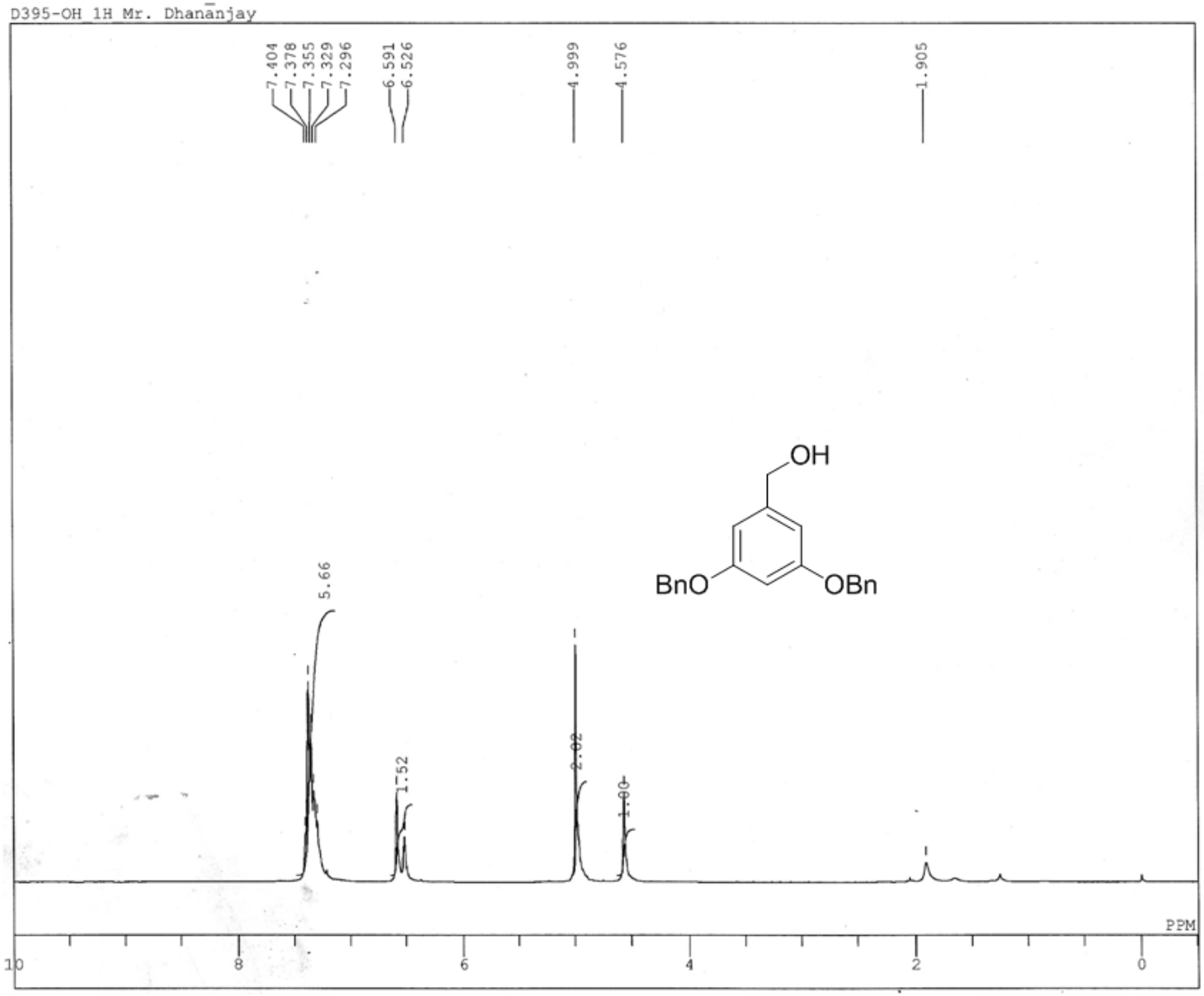
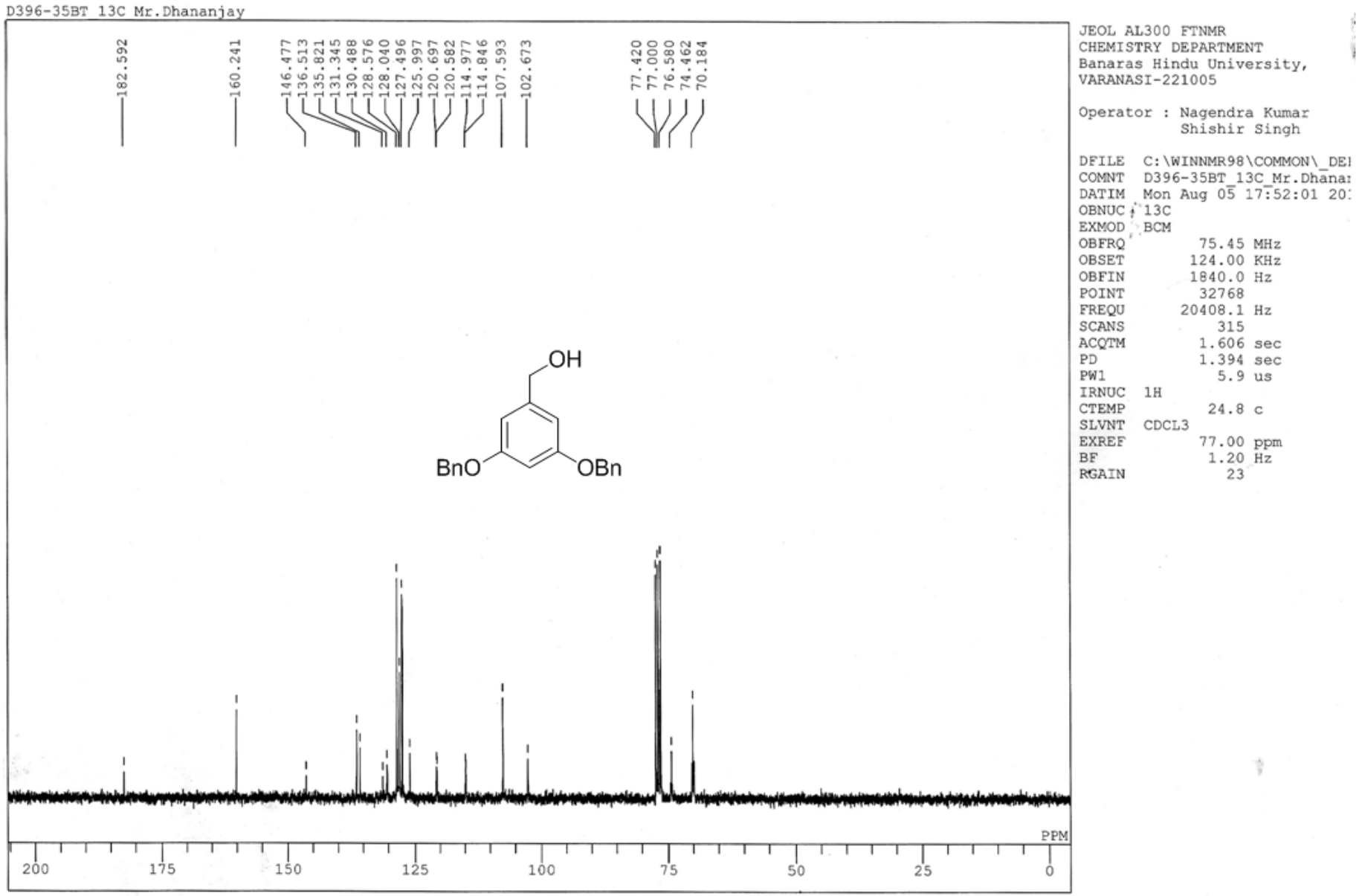
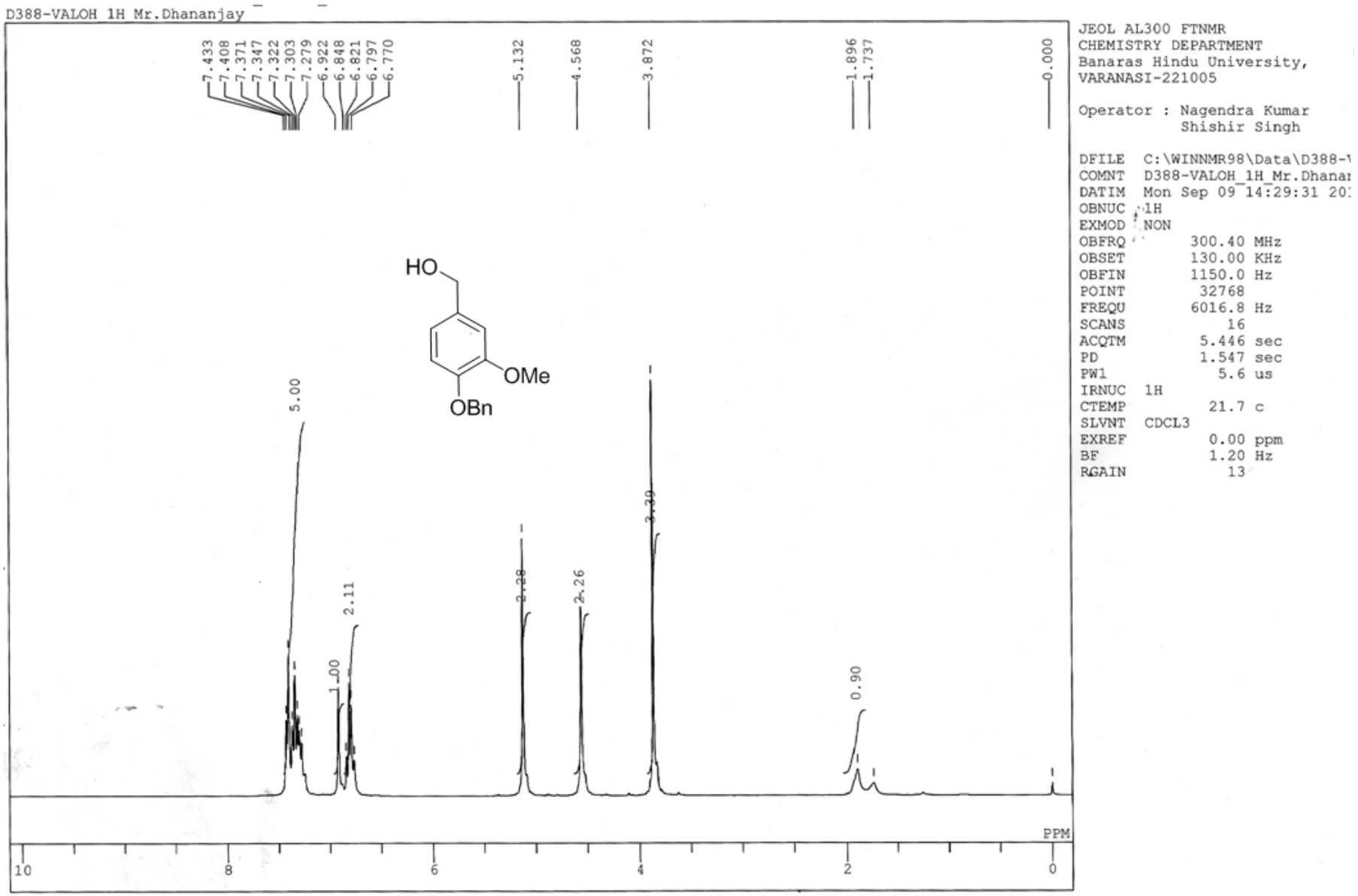


Spectrum 22: 300 MHz ^{1}H NMR of compound **24a**



Spectrum 23: 75 MHz ^{13}C NMR of compound **24a**



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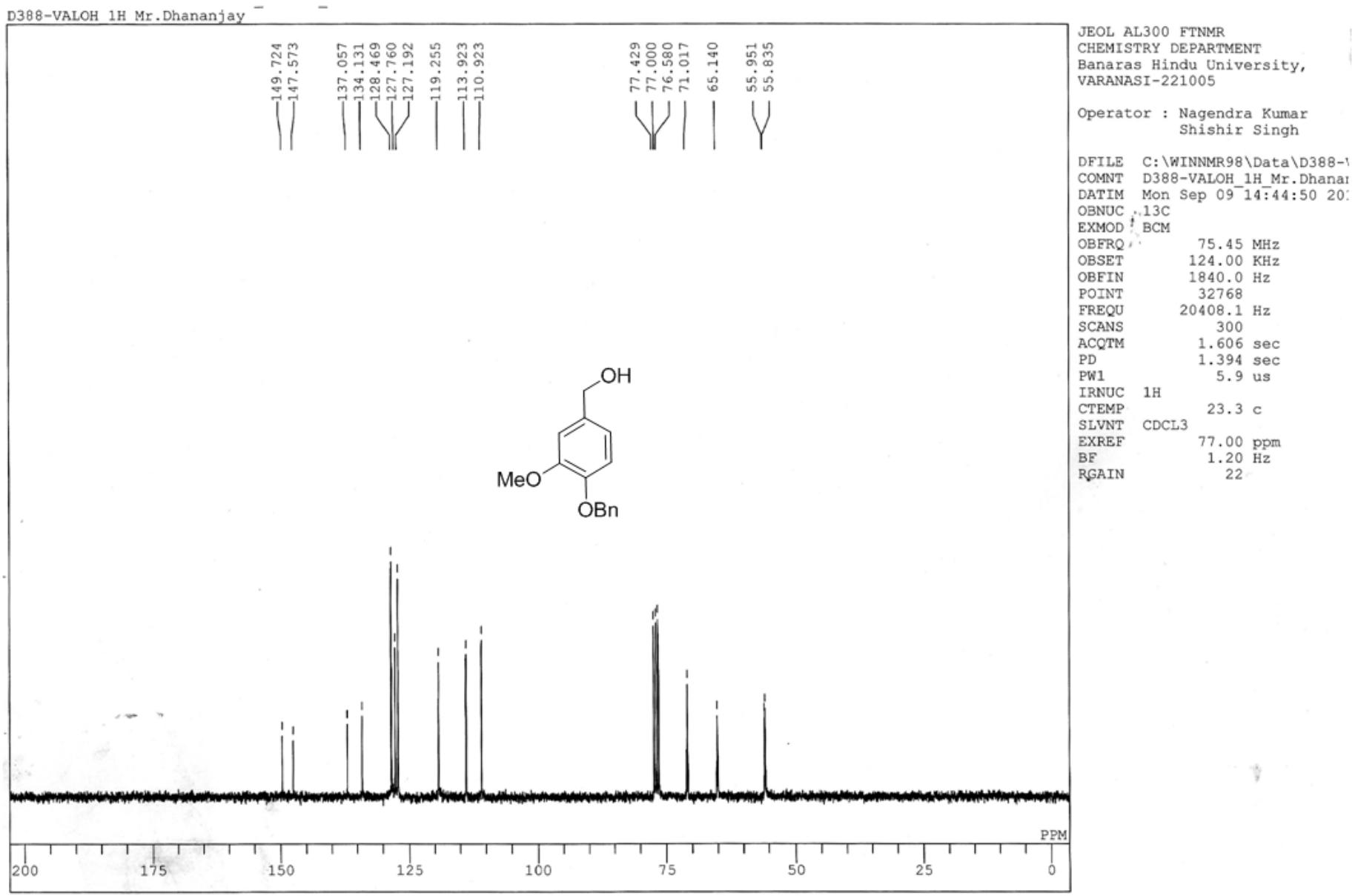

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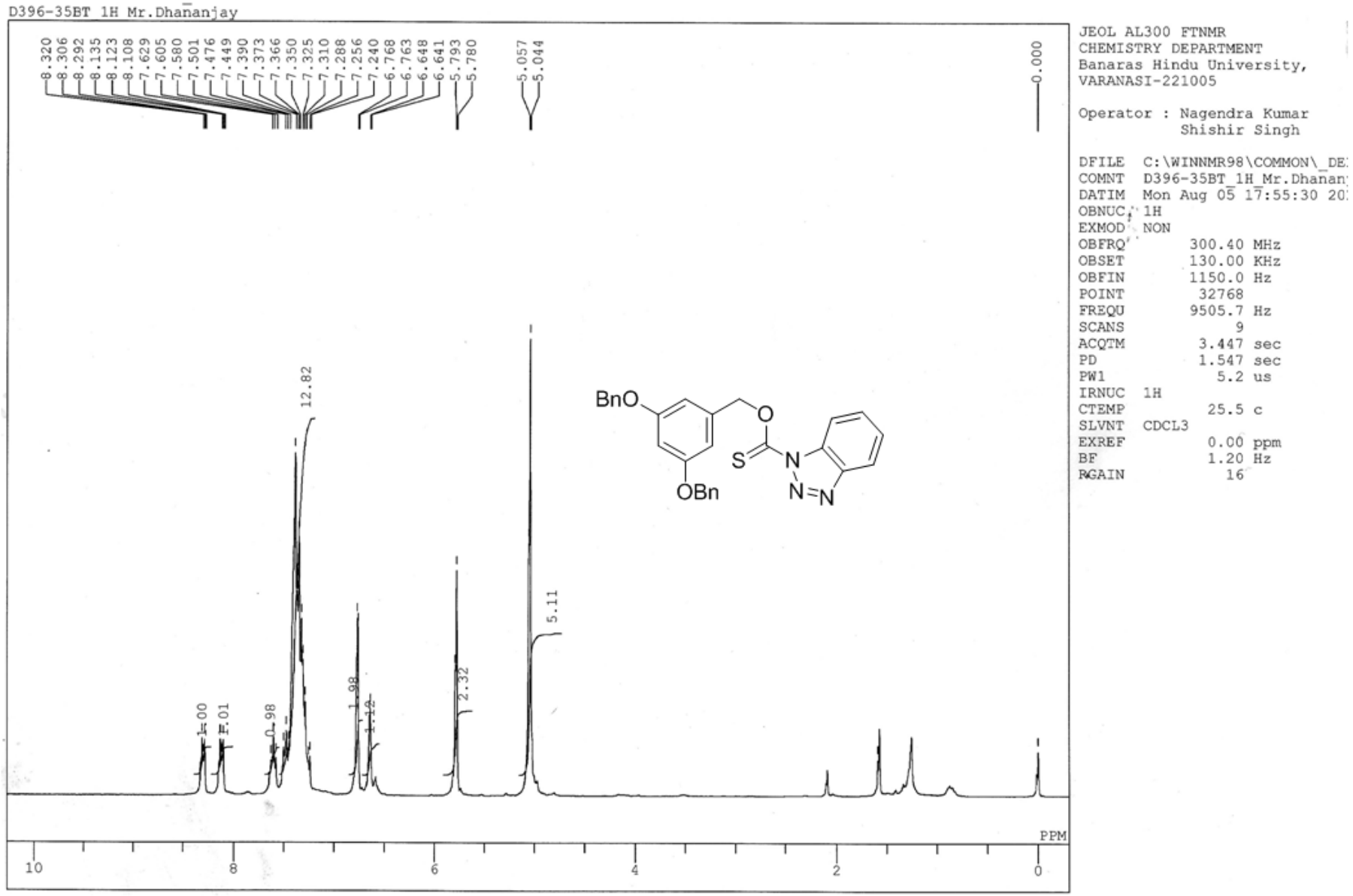

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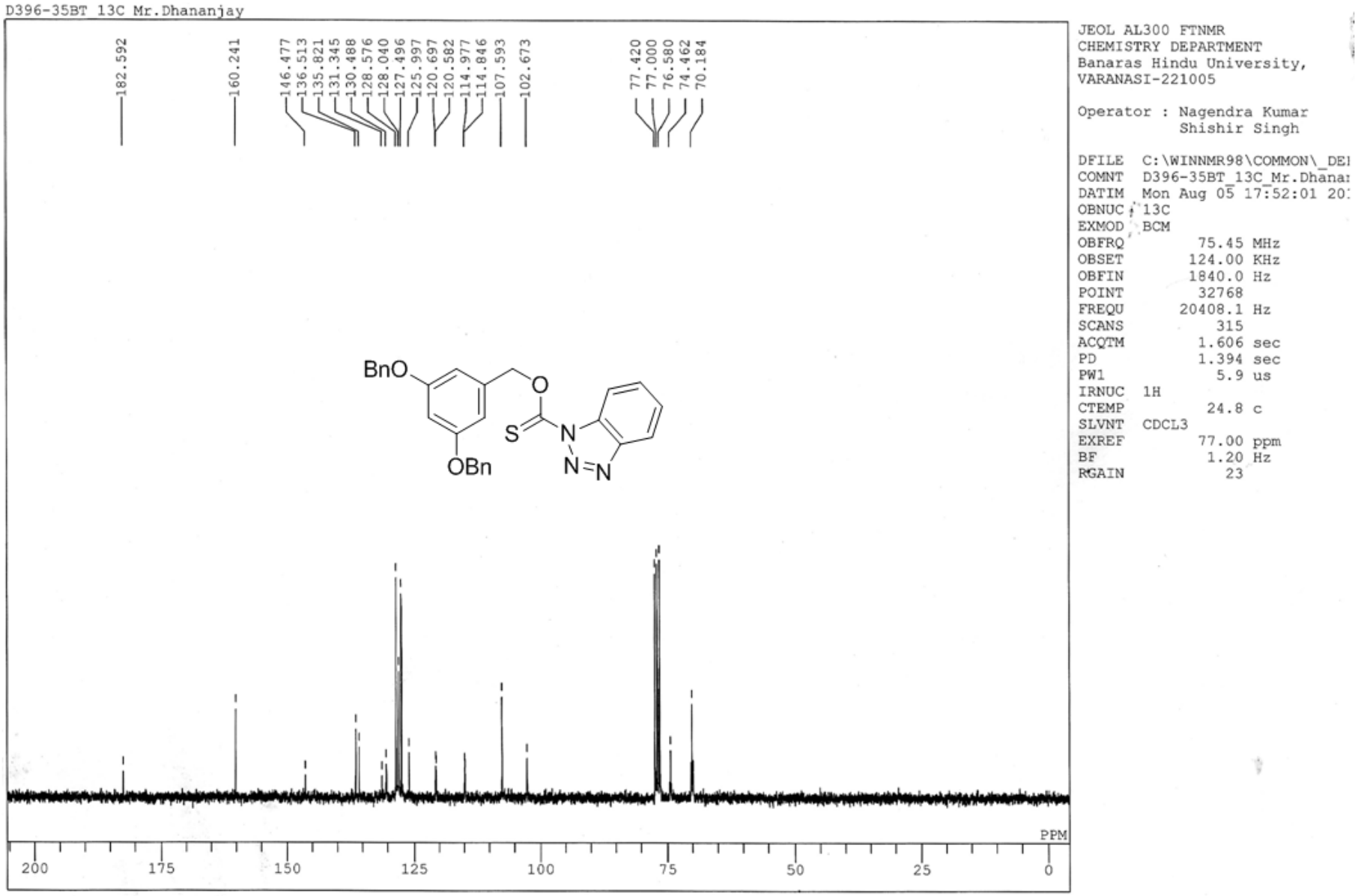
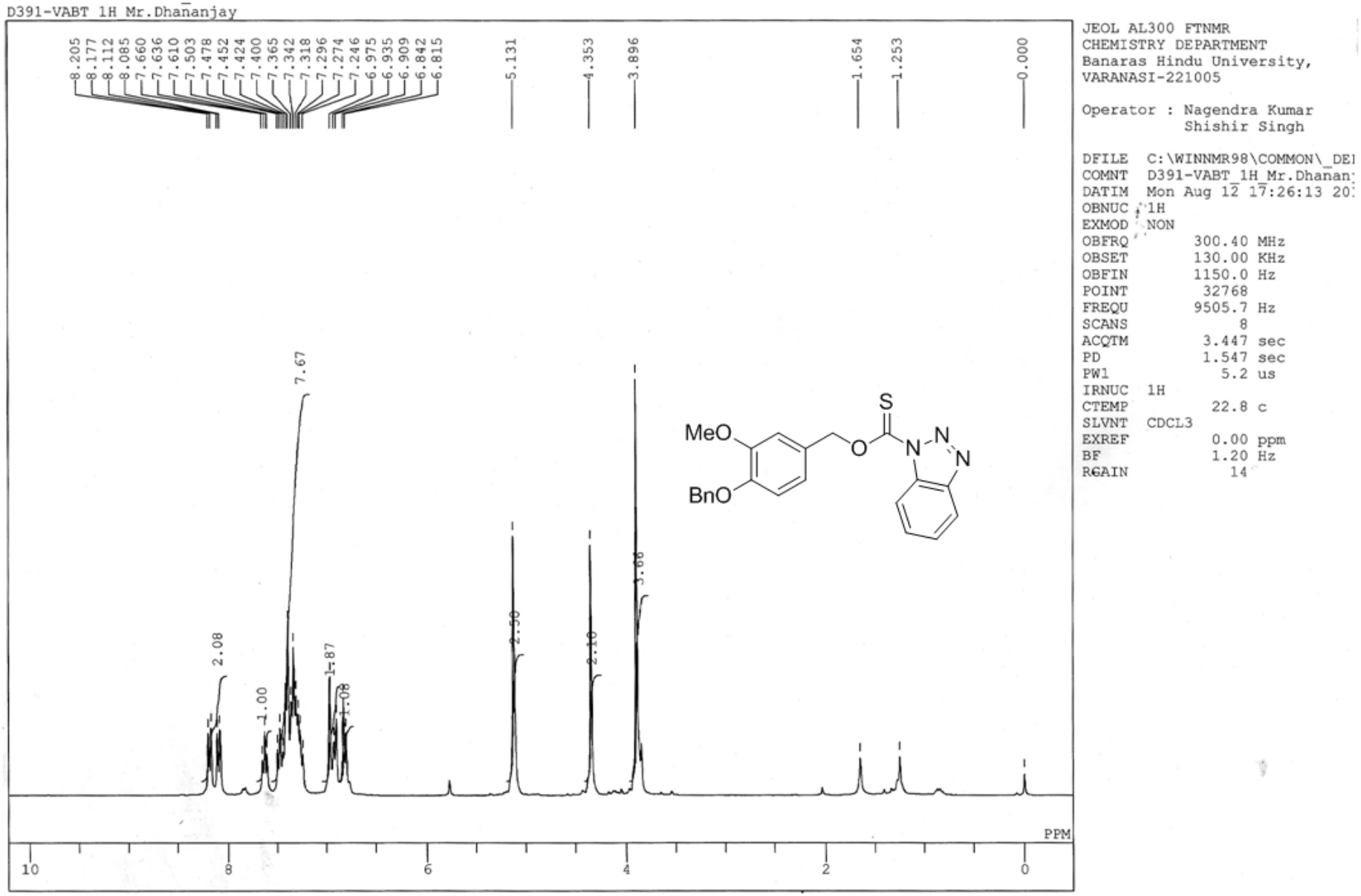
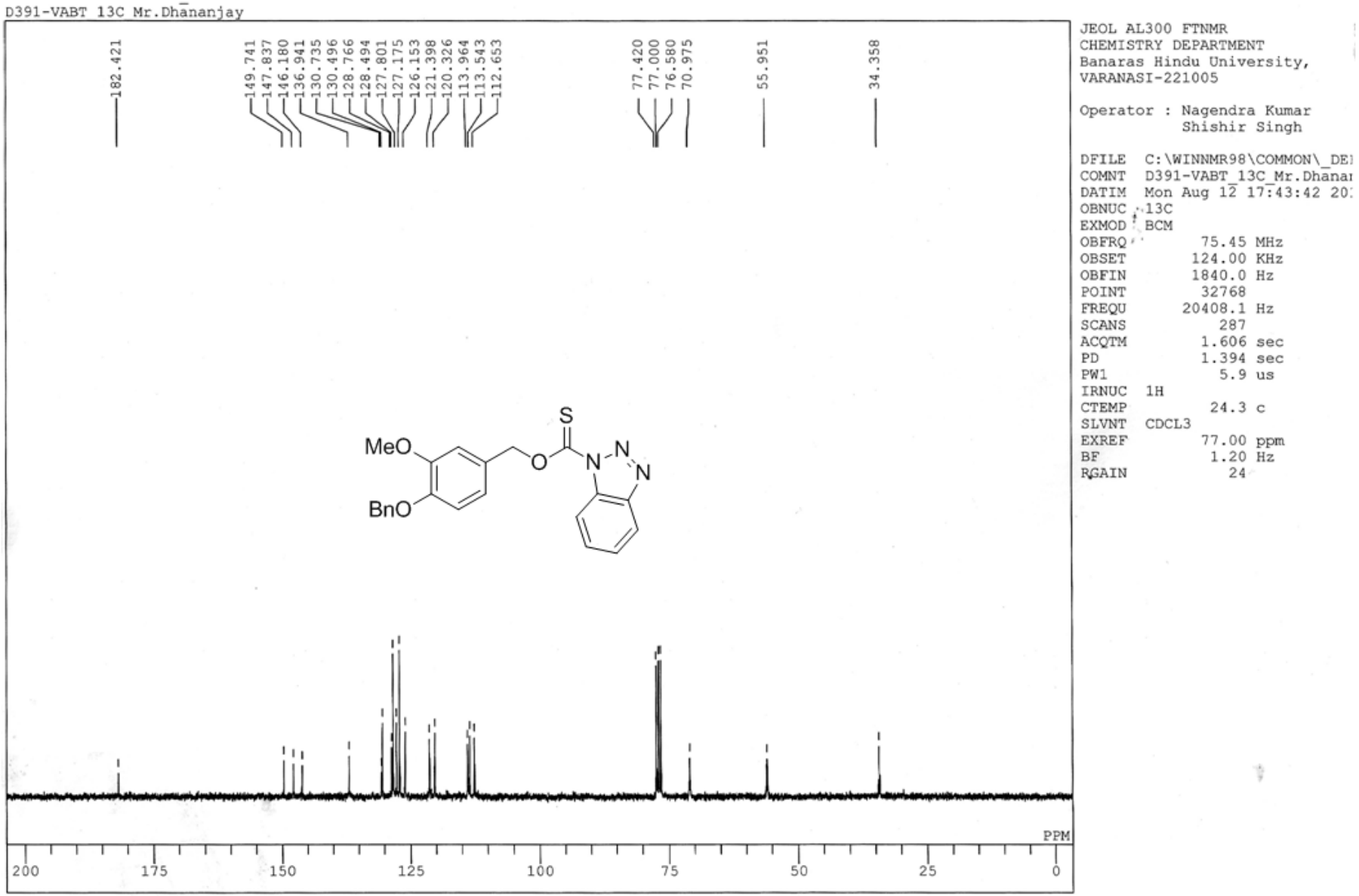


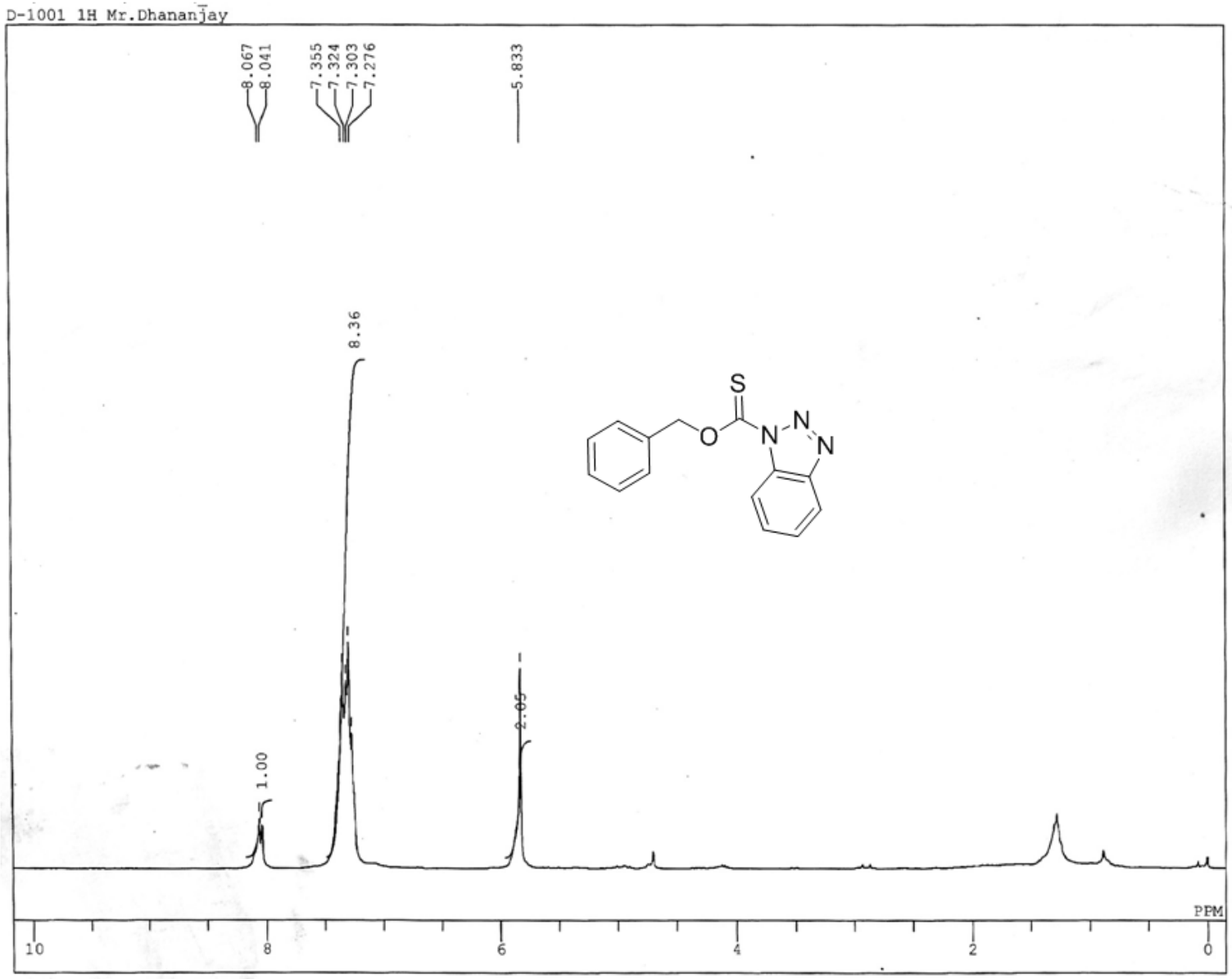
Spectrum 27: 75 MHz ^{13}C NMR of compound **25a**



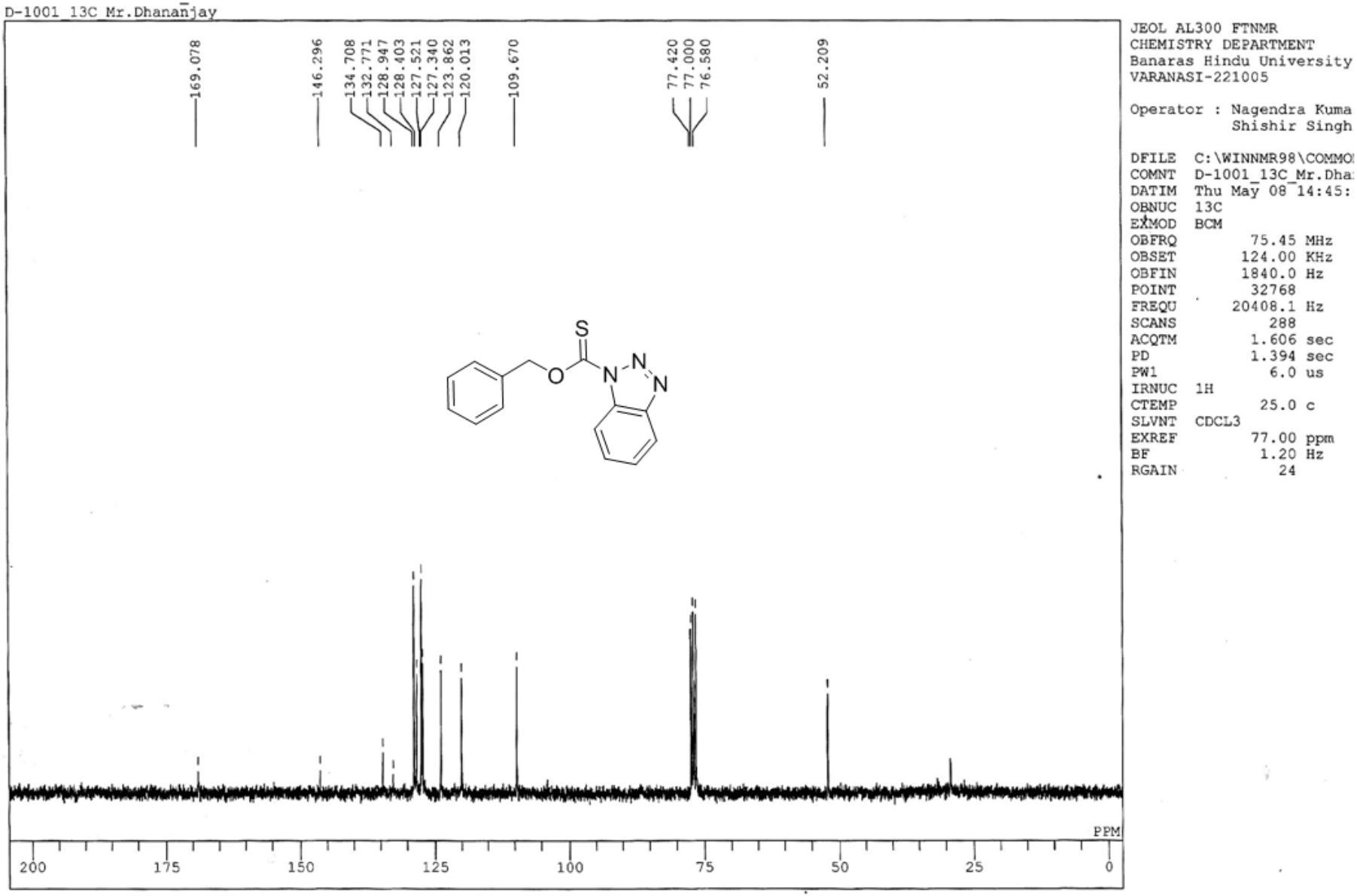
Spectrum 28: 300 MHz ^{1}H NMR of compound **25b**



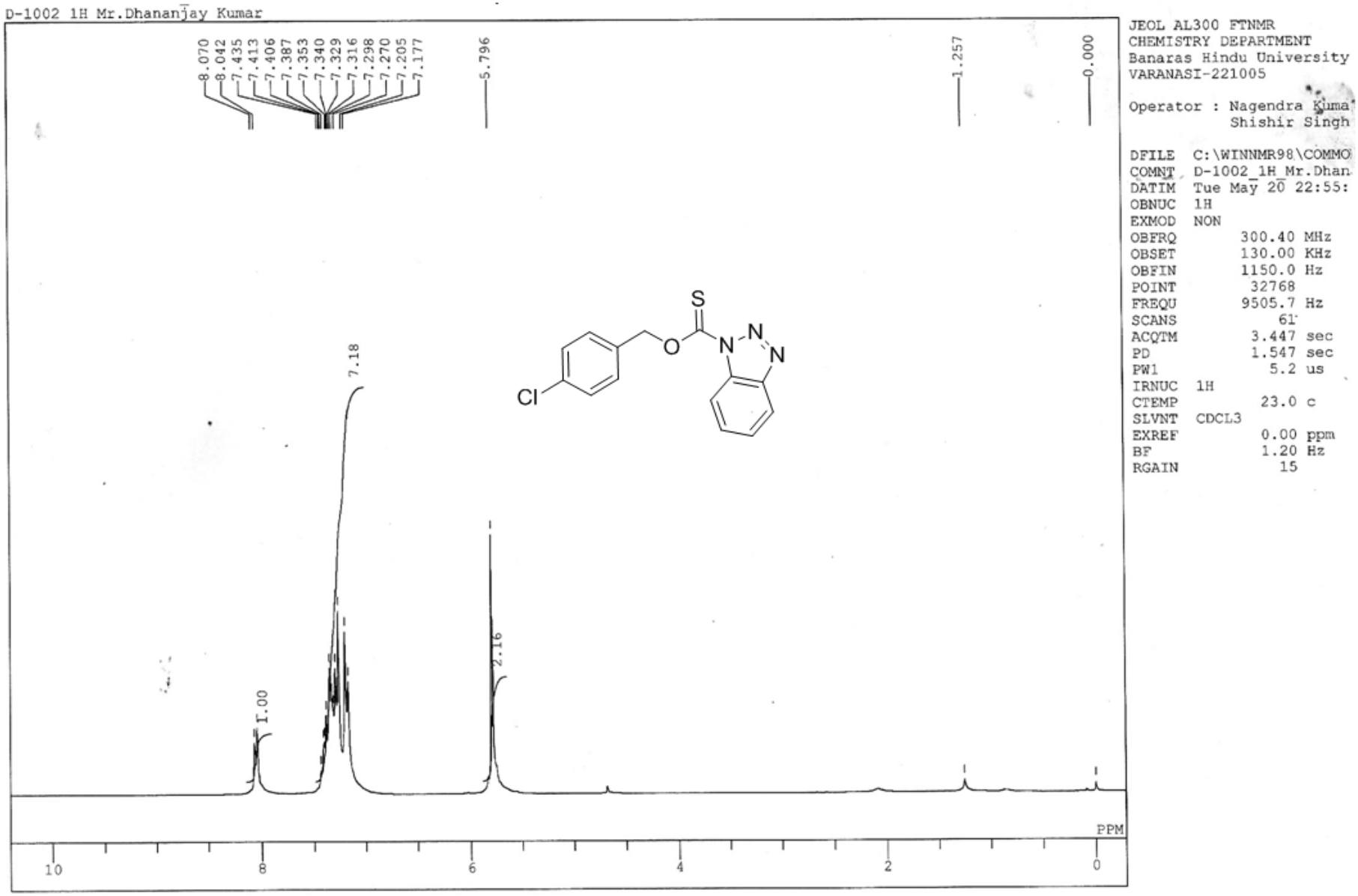
Spectrum 29: 75 MHz ^{13}C NMR of compound **25b**



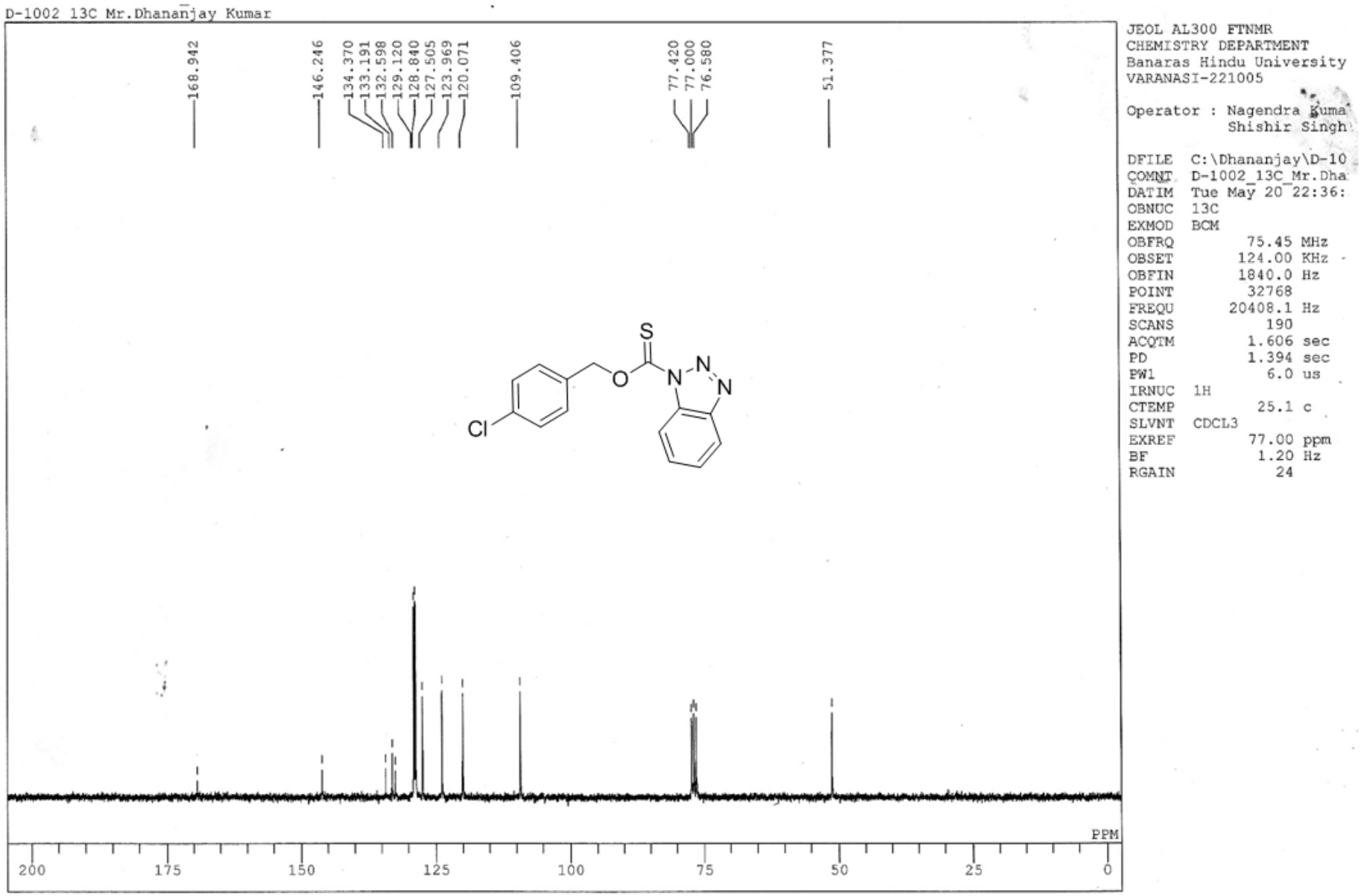
Spectrum 30: 300 MHz ^{1}H NMR of compound **25c**



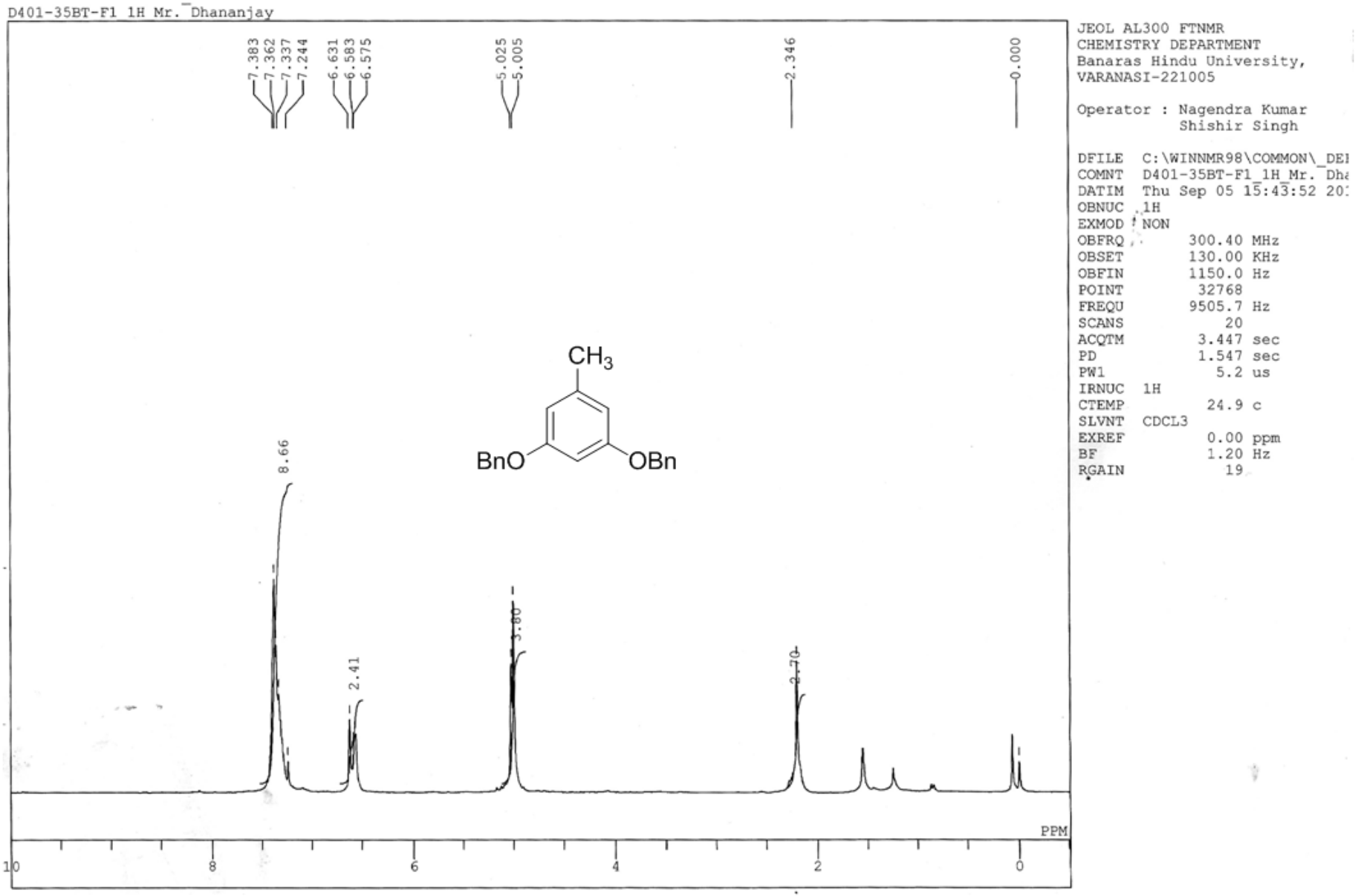
Spectrum 31: 75 MHz ^{13}C NMR of compound **25c**



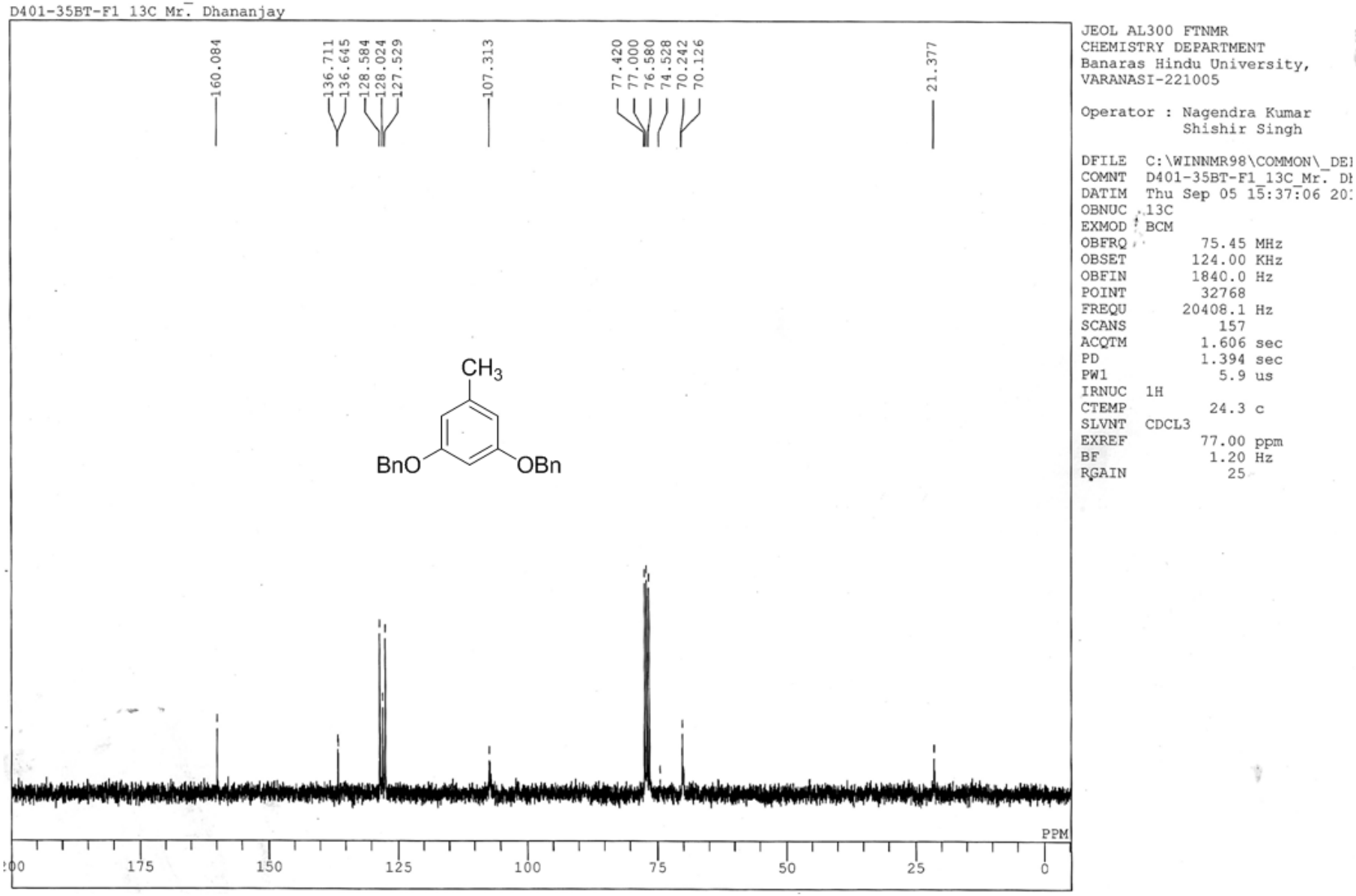
Spectrum 32: 300 MHz ^{1}H NMR of compound **25d**



Spectrum 33: 75 MHz ^{13}C NMR of compound **25d**



Spectrum 34: 300 MHz ^{1}H NMR of compound **26a**



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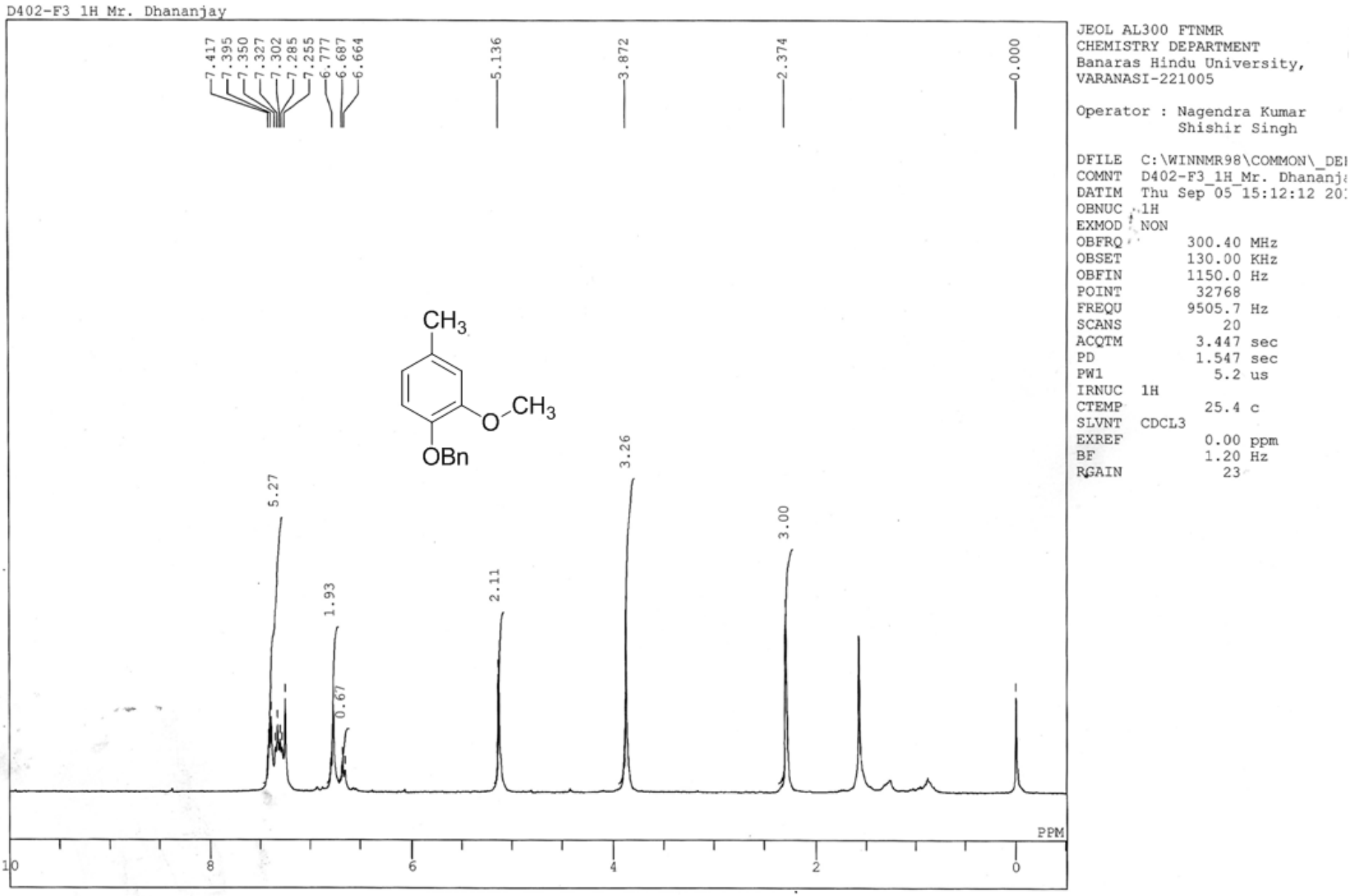

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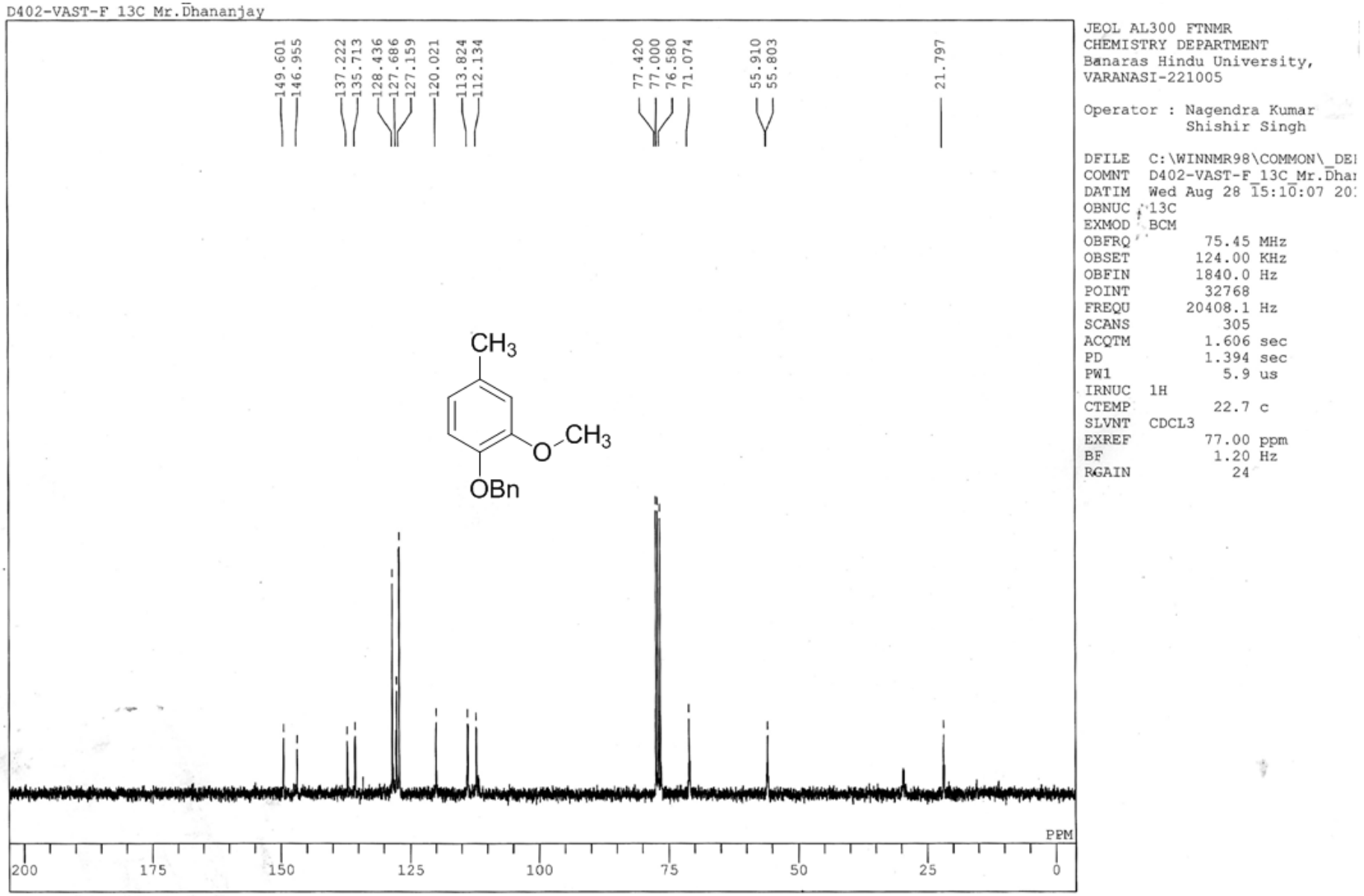

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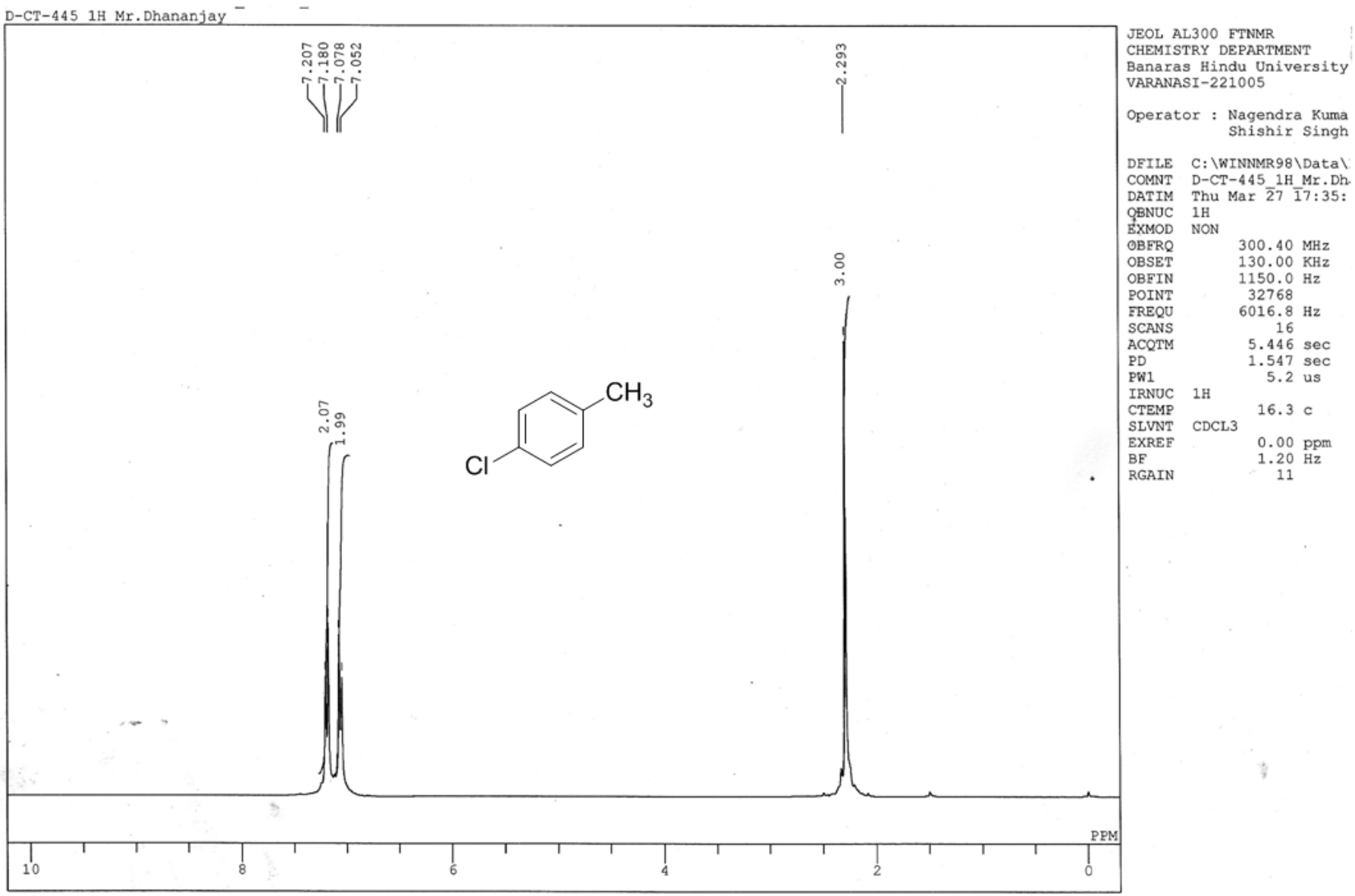

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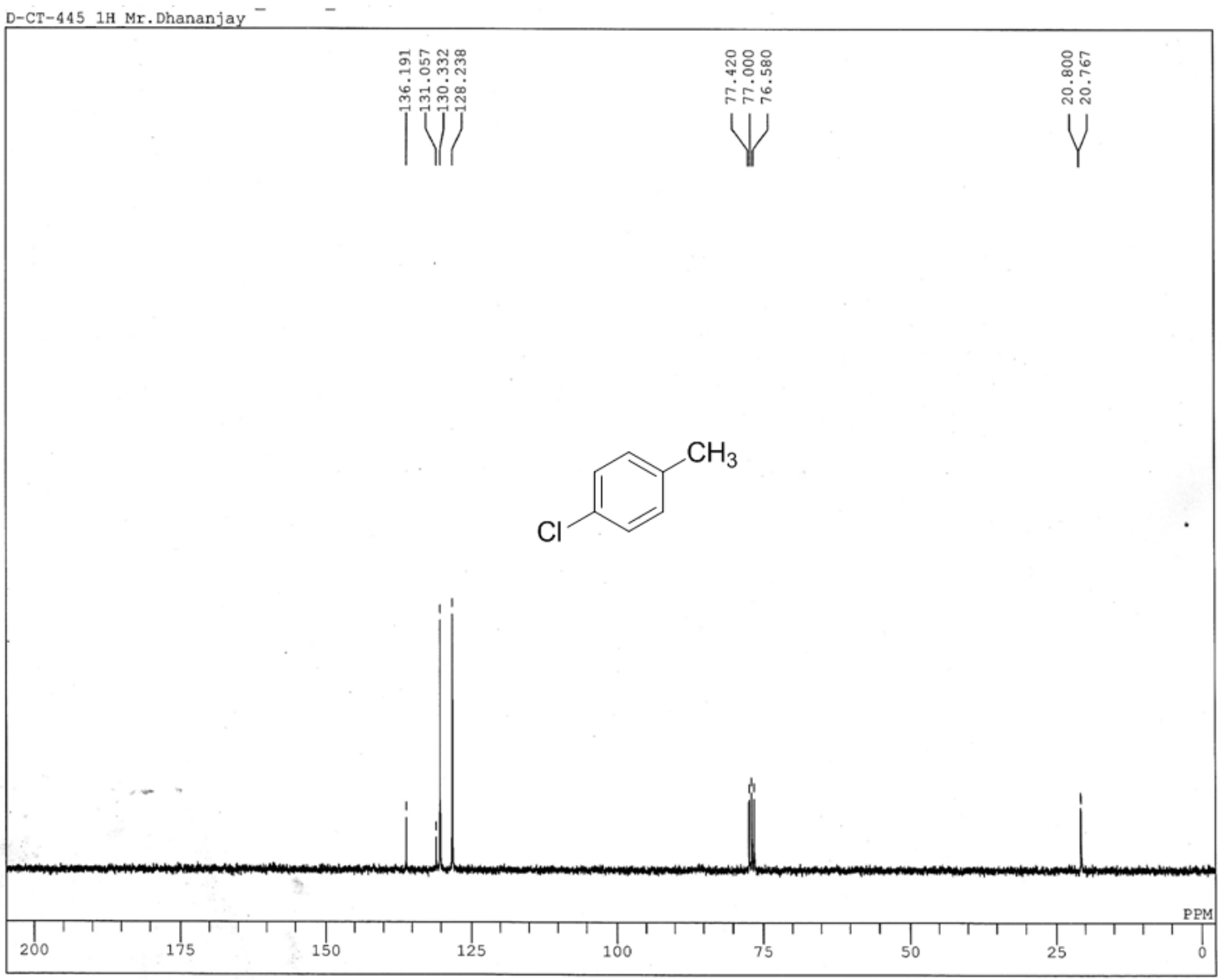
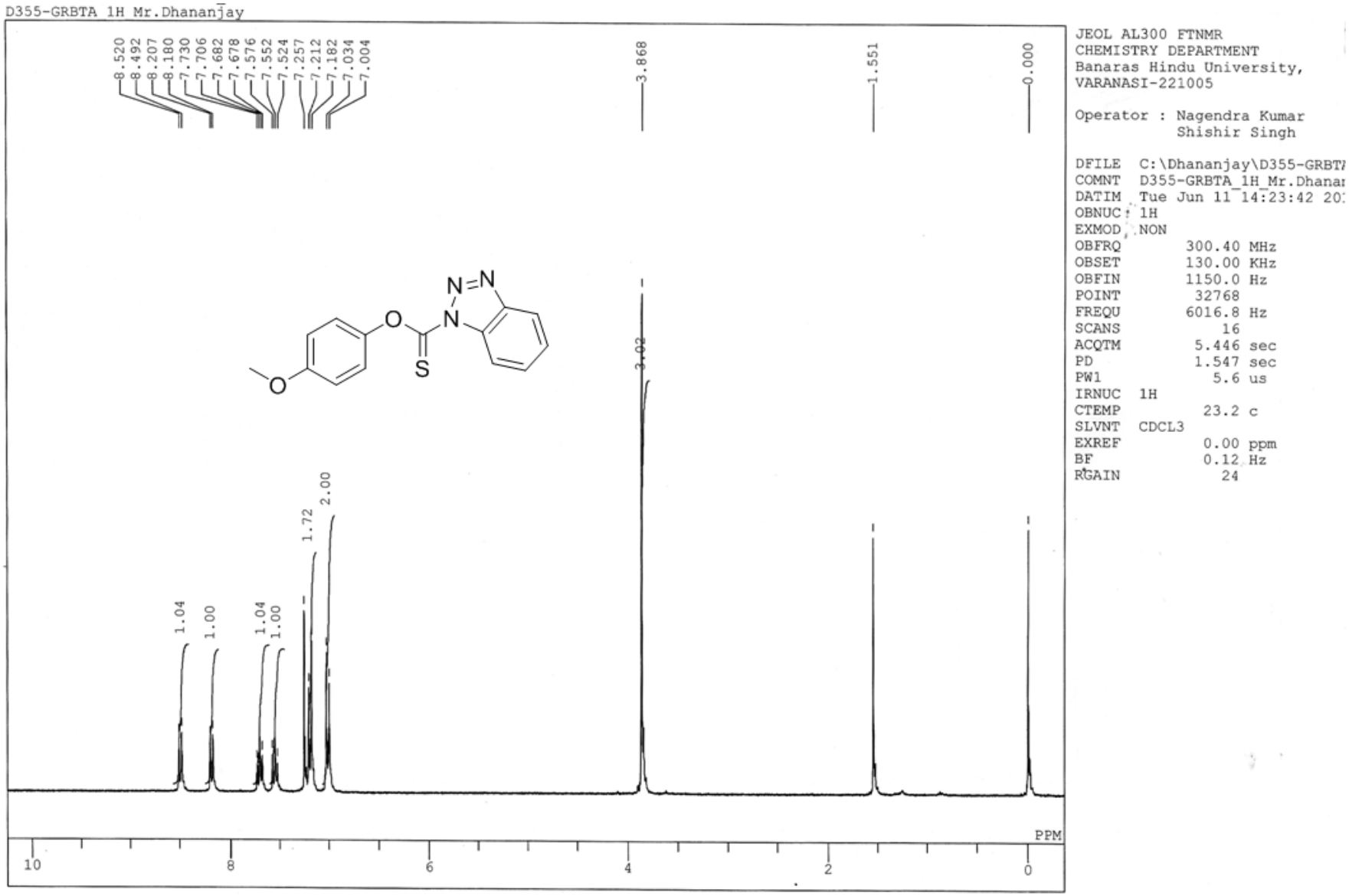
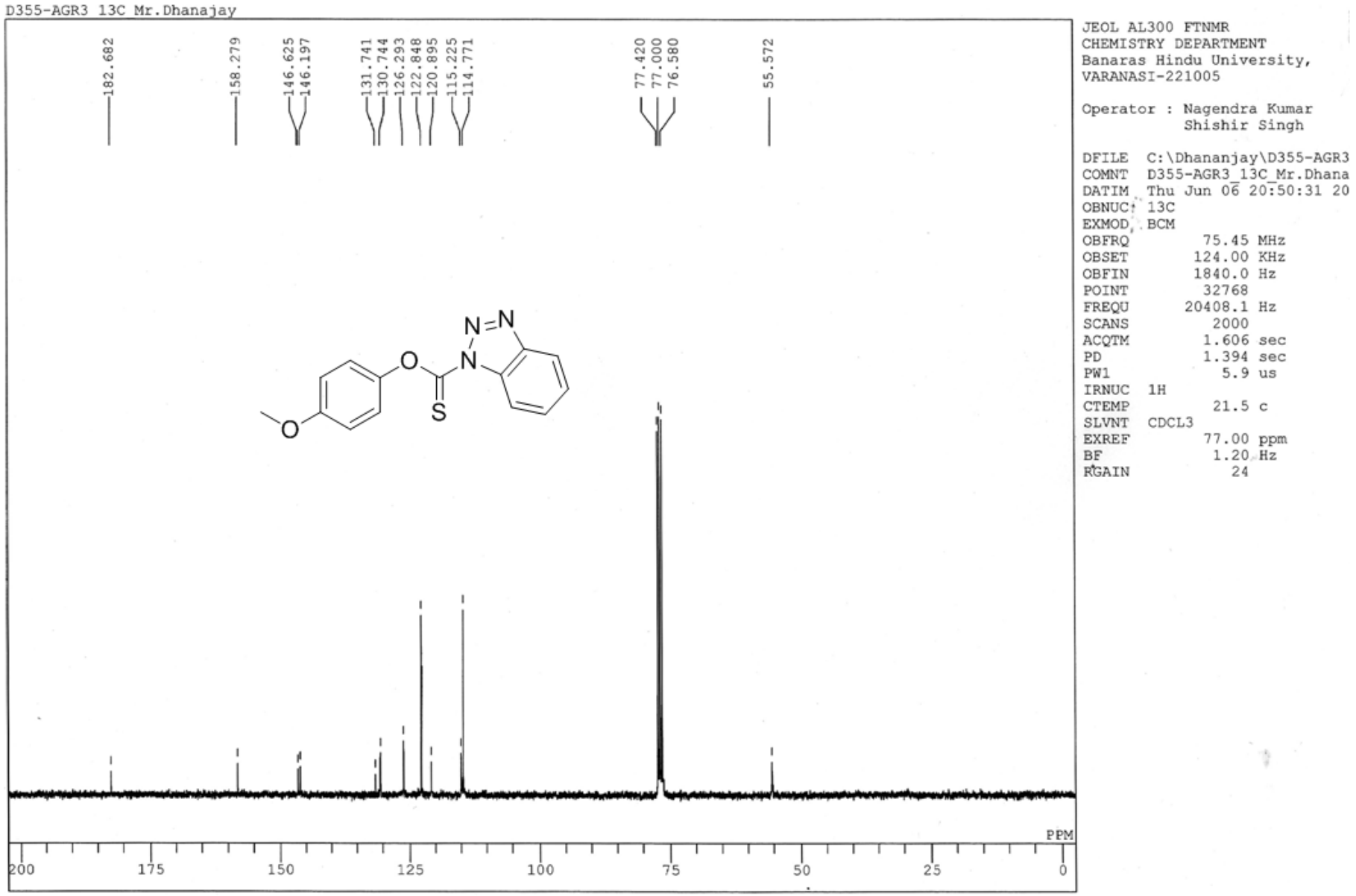


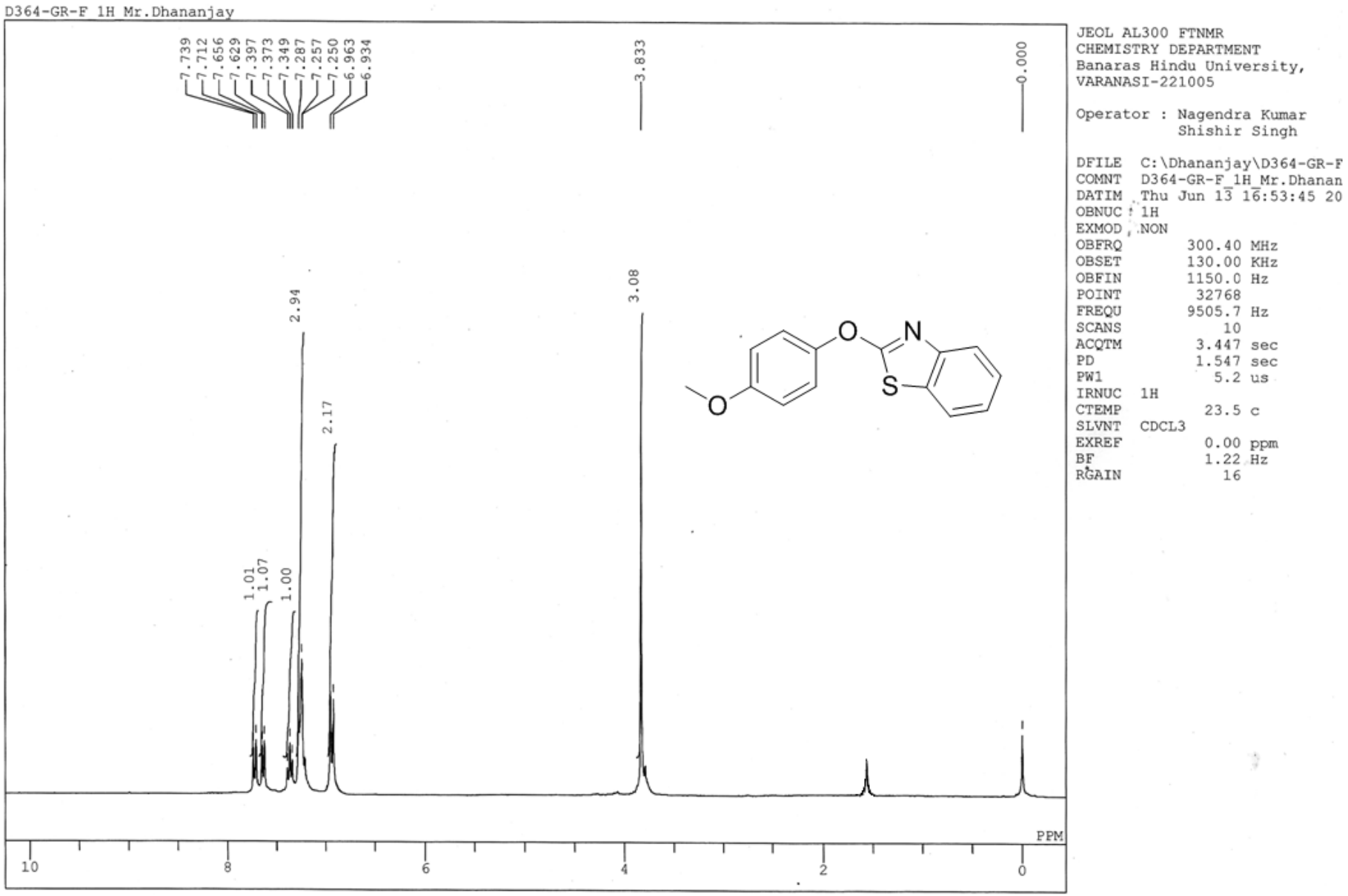
Spectrum 39: 75 MHz ^{13}C NMR of compound **26d**



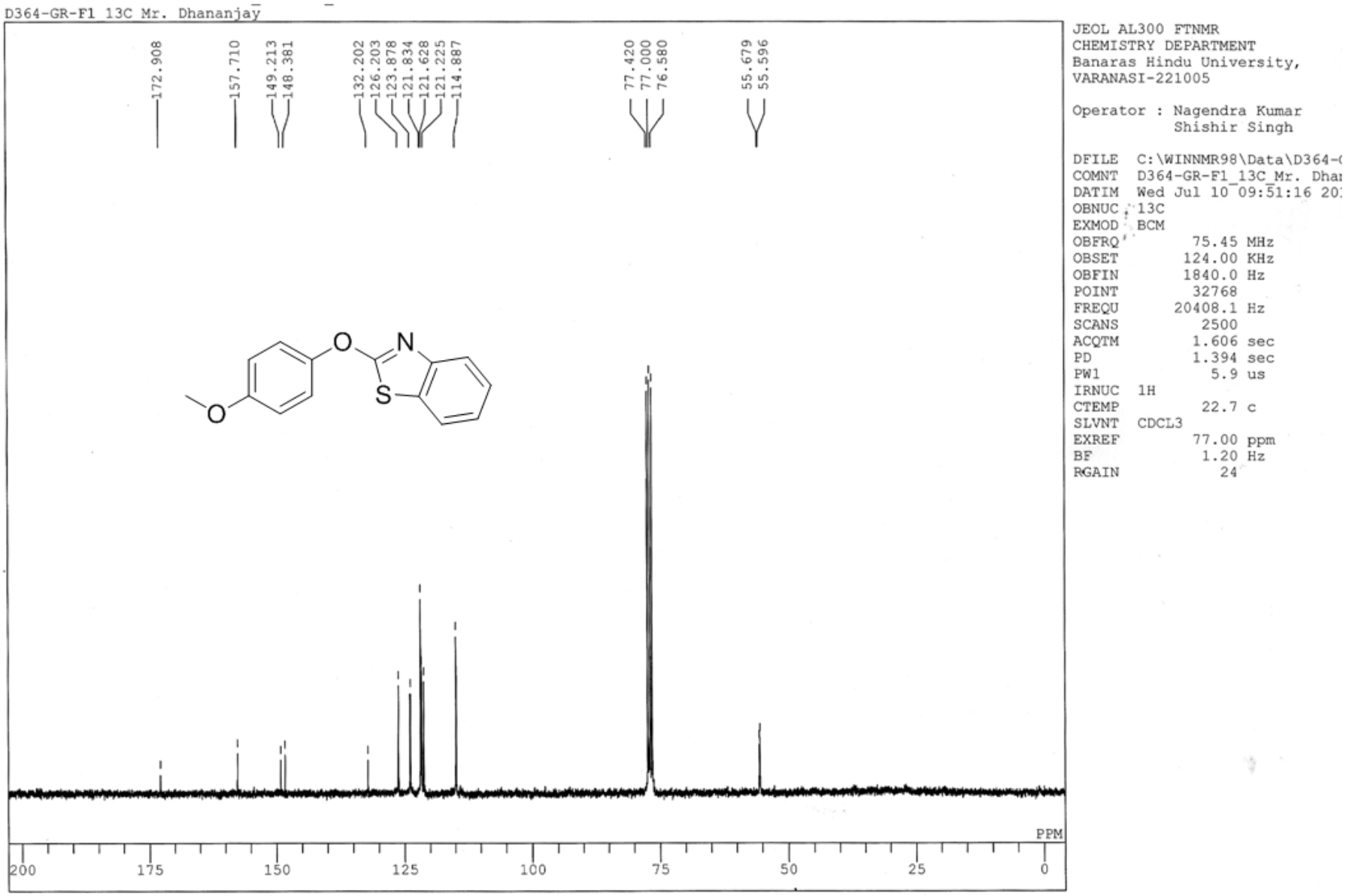
Spectrum 40: 300 MHz ^{1}H NMR of compound **28**



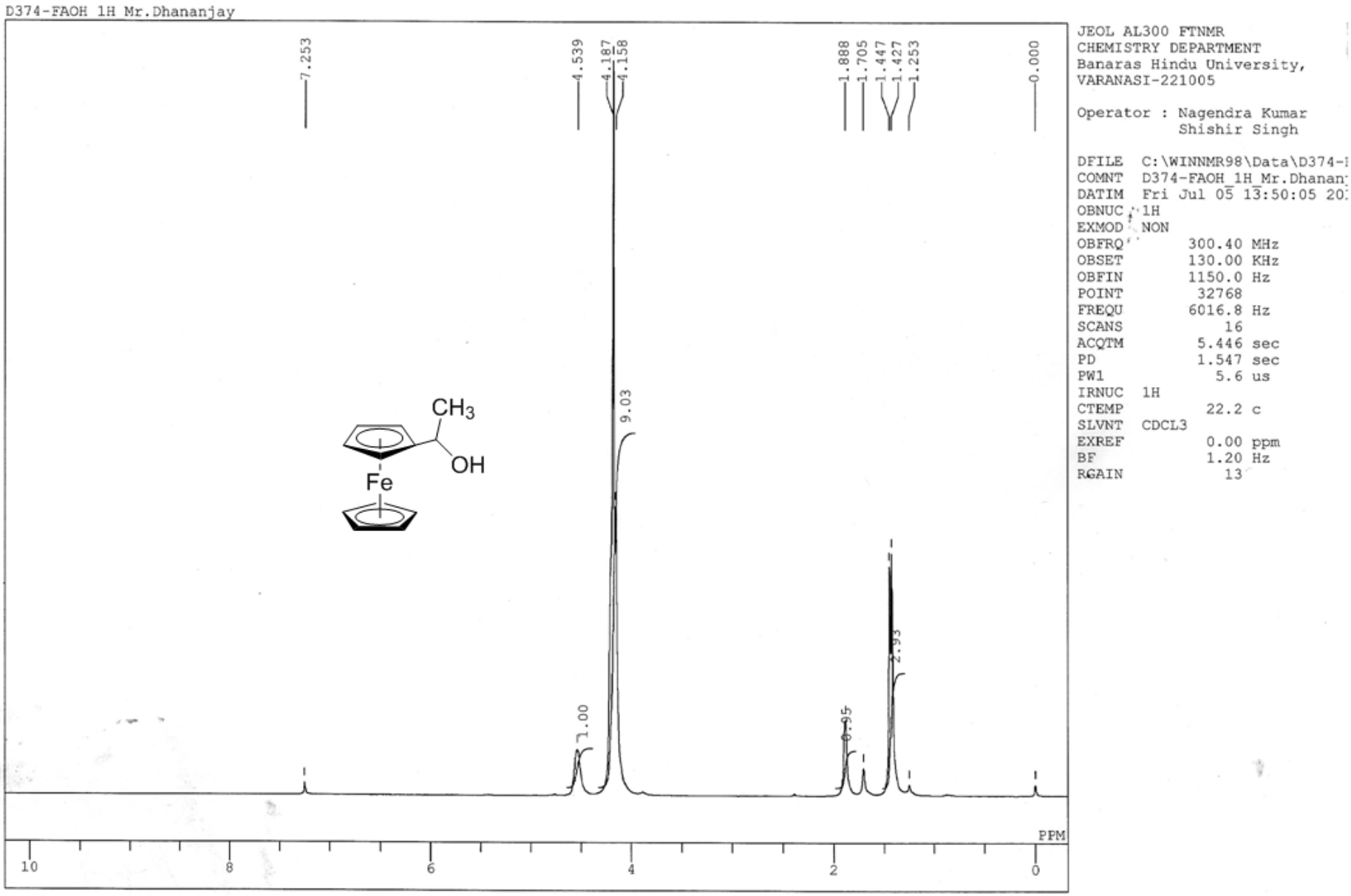
Spectrum 41: 75 MHz ^{13}C NMR of compound **28**



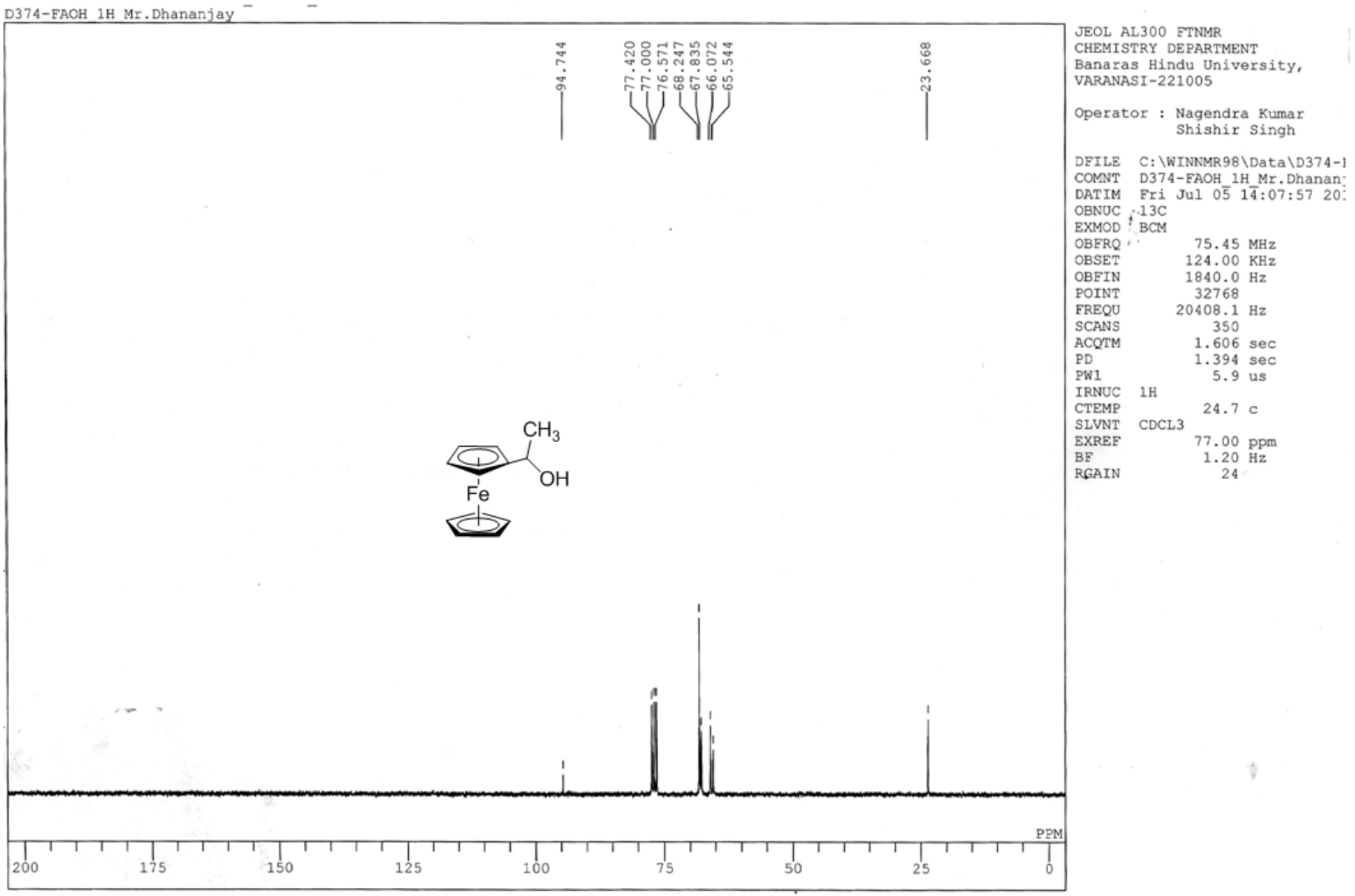
Spectrum 42: 300 MHz ^{1}H NMR of compound **29**



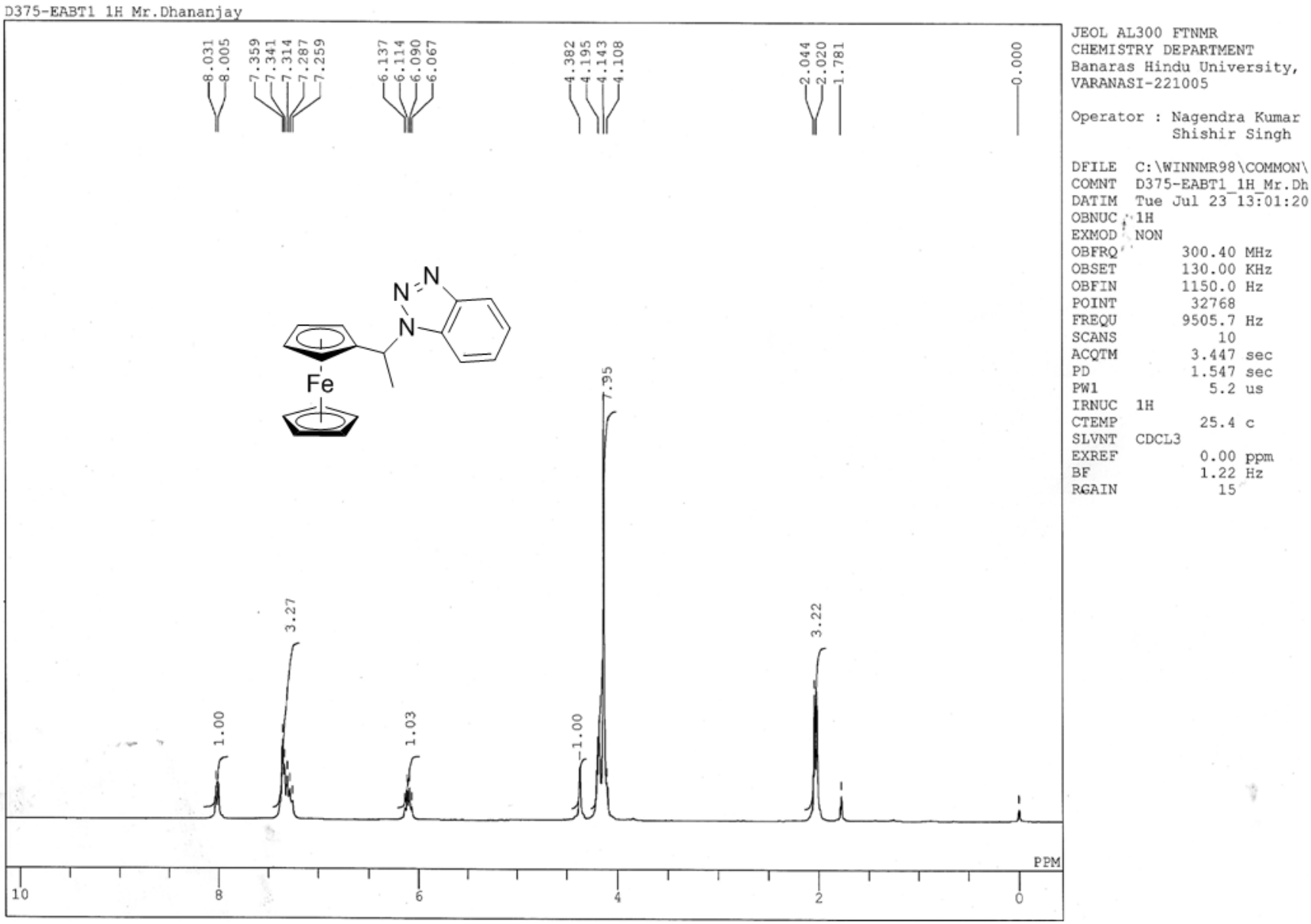
Spectrum 43: 75 MHz ^{13}C NMR of compound **29**



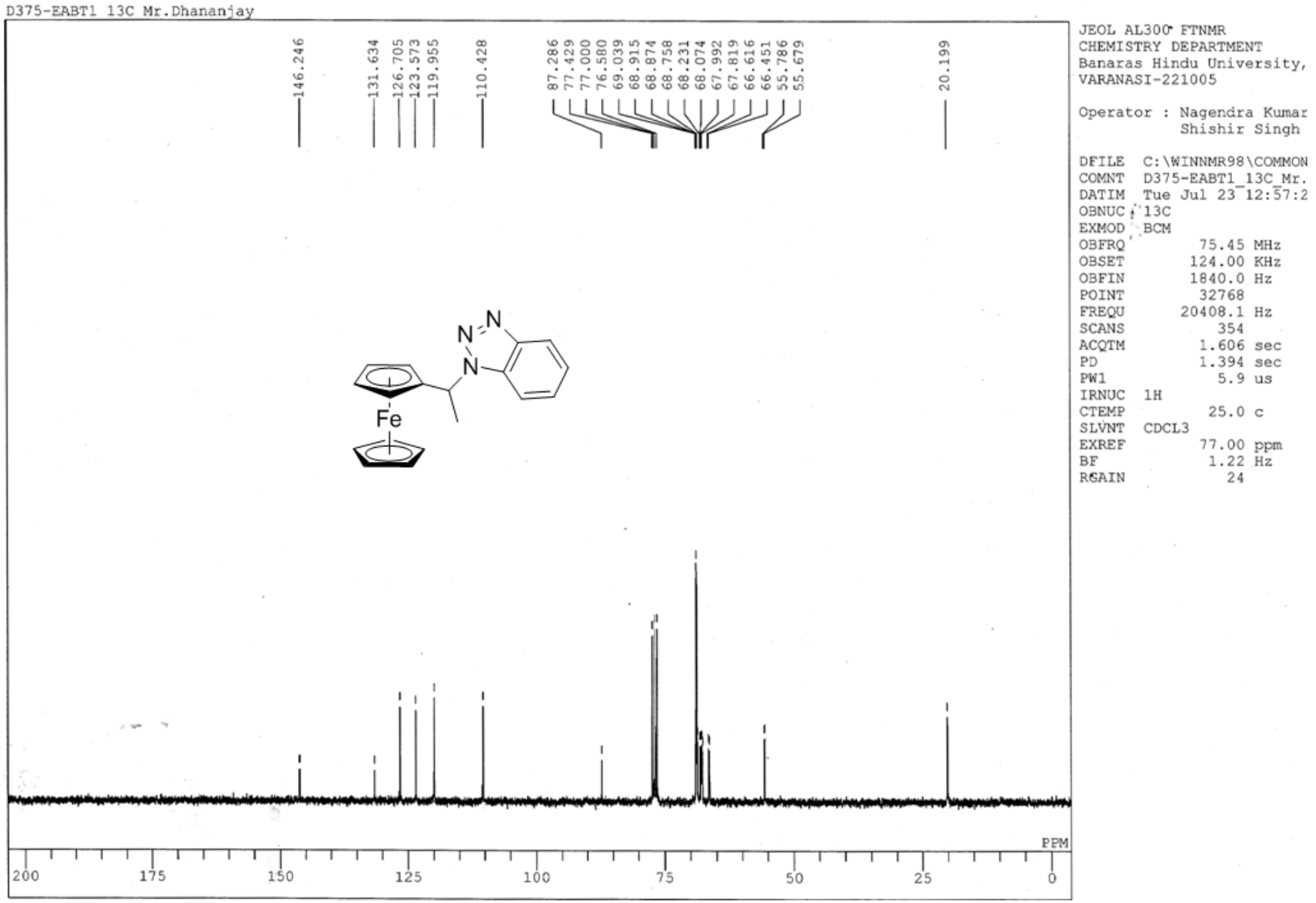
Spectrum 44: 300 MHz ^{1}H NMR of compound **30**



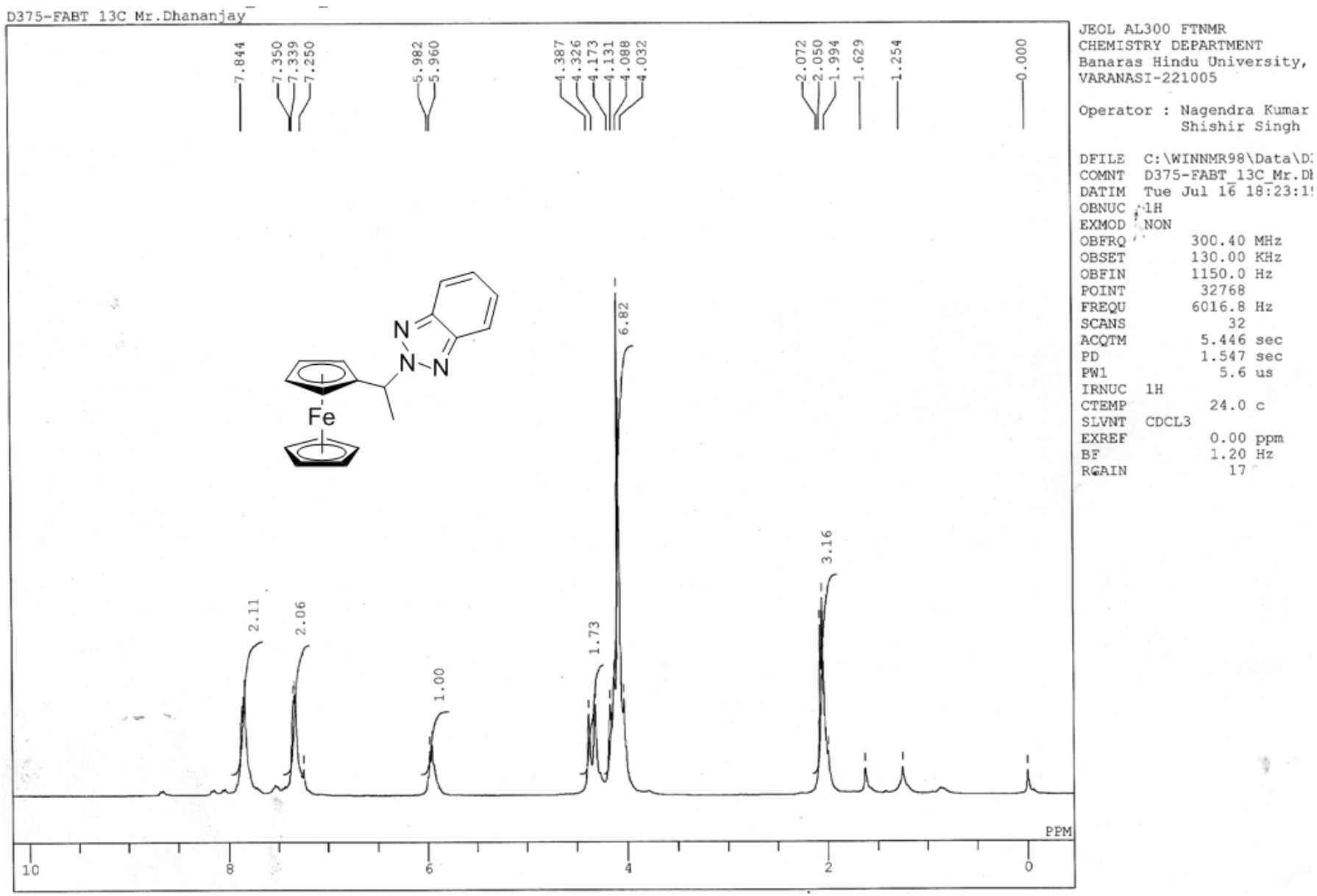
Spectrum 45: 75 MHz ^{13}C NMR of compound **30**



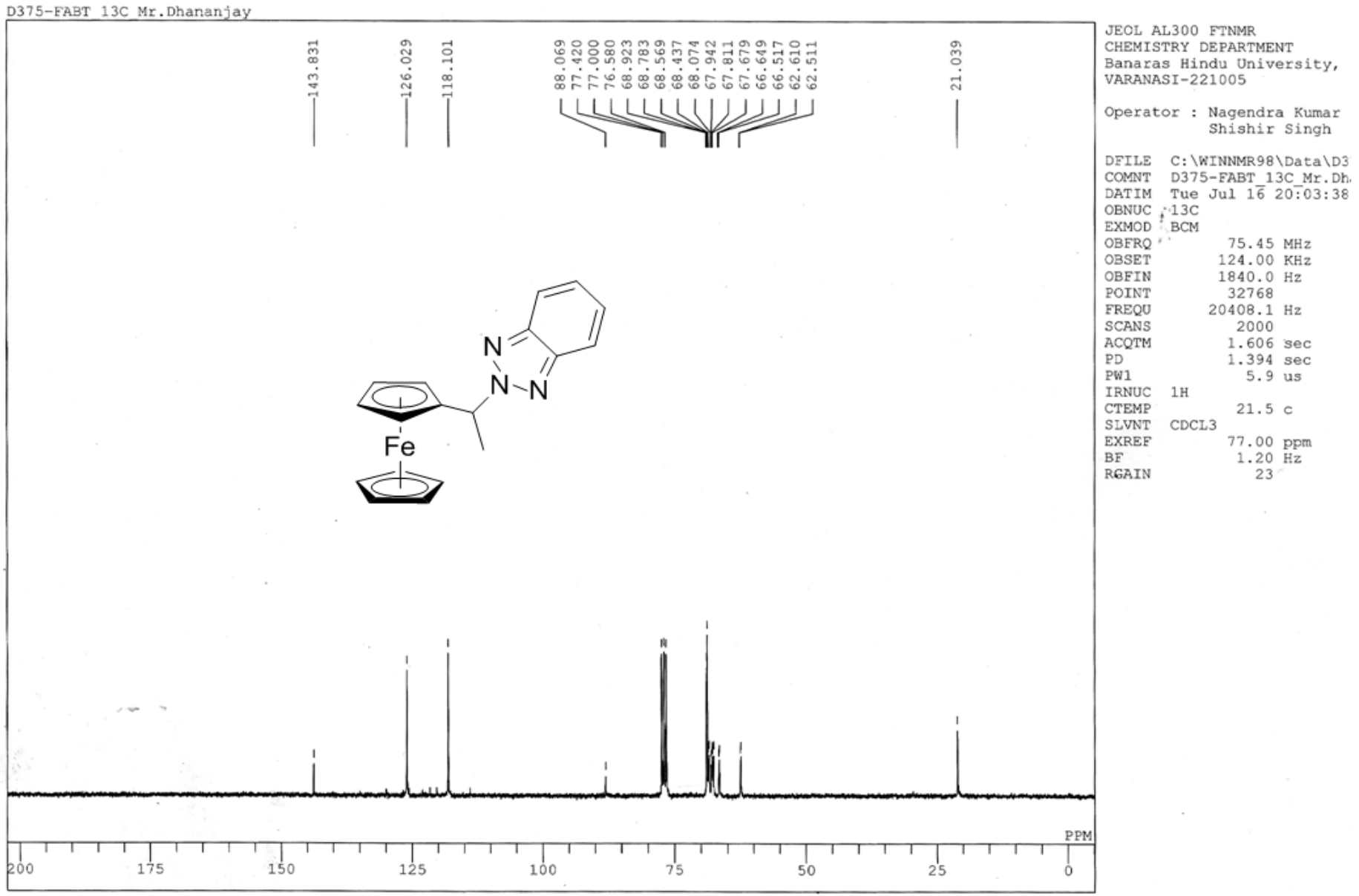
Spectrum 46: 300 MHz ^{1}H NMR of compound **31**



Spectrum 47: 75 MHz ^{13}C NMR of compound **31**



Spectrum 48: 300 MHz ^{1}H NMR of compound **32**



Spectrum 49: 75 MHz ^{13}C NMR of compound **32**

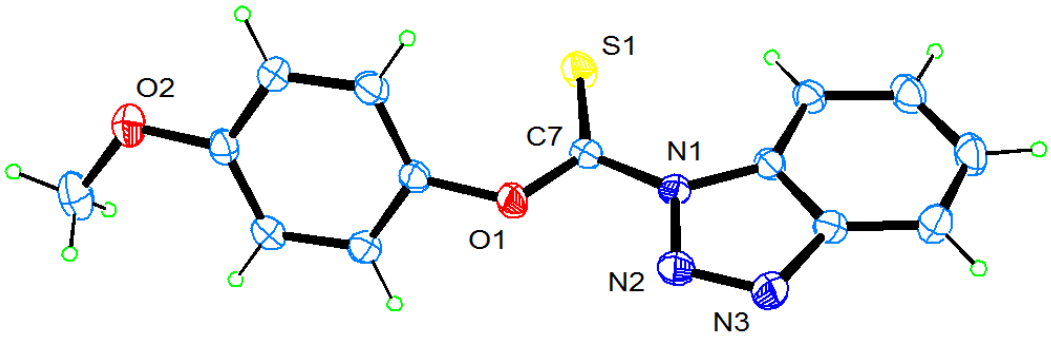
2. Single Crystal X-Ray Experimental details, refinement data and visualized molecular structure of compounds 28, 31 & 32

Data Collection and Refinement

Single-crystal X-ray data of compounds were collected on Xcalibur Eos (Oxford) CCD-Diffractometer using graphite monochromated Mo*K*α radiation (λ = 0.71073 Å). The data integration and reduction were processed with CrysAlis Pro software. The structures were solved by the direct method and then refined on F^2 by the full matrix least-squares technique with the SHELX-97 set of software using the WinGX (version 1.80.05) program package. All non-hydrogen atoms were refined anisotropically and hydrogen atoms were treated as riding atoms using SHELX default parameters. Molecular structures have drawn using ORTEP software given in Figures S1-3. Further information on the crystal structure determination (excluding structure factors) has been given as Table S1-3 and also deposited in the Cambridge Crystallographic Data Centre as supplementary publications nos. **978009, 978010** and **978011** for compounds **28**, **31** and **32** respectively. Copies of the data can be obtained free of charge upon application to CCDC, 12 Union Road, Cambridge CB2 1EZ, UK (fax: (+44) 1223-336-033. e-mail: deposit@ccdc.cam.ac.uk) or via internet.

Procedure for crystallization of compound 28, 31 & 32

For crystallization a 40:60 mixture of ethyl acetate and hexane has been used and kept in dark place at temperature 25 ^{o}C. The single crystal was isolated in its initial state of growth.

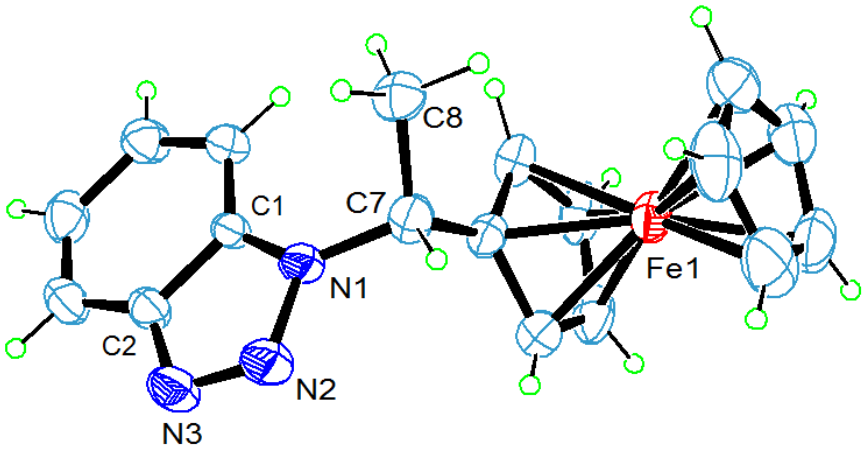


Selected bond length (Å): S1-C7 1.622, O1-C7 1.333, O1-C8 1.414, N1-N2 1.406, N1-C7 1.386, N1-C1 1.383, N2-N3 1.281, O2-C11 1.373, O2-C14 1.417; **Selected bond angles:** C7-O1-C8 120.2, N2-N1-C7 119.8, N2-N1-C1 109.0, C7-N1-C1 131.2, N1-N2-N3 108.4, S1-C7-O1 127.4, S1-C7-N1 123.9, O1-C7-N1 108.7, O1-C8-C13 117.1, O1-C8-C9 120.6, N1-C1-C6 134.6, N1-C1-C2 104.3, C1-C2-N3 108.8.

Figure S1. Molecular structure of compound **28.** Thermal ellipsoids of carbon, nitrogen, sulfur and oxygen are set at 40 % probability.

Table S1. Crystallographic refinement data for compound **28**

Compound 28	
Empirical Formula	**C_{14} H_{11} N_3 O_2 S**
Formula Weight	285.33
Crystal System	Triclinic
Space group	P -1
a (Å)	4.2311(7)
b (Å)	8.0019(16)
c (Å)	20.415(3)
β (°)	92.671(13)
V (Å^3)	676.2(2)
Z	2
Density (calc)	1.401
F(000)	280
μ (mm^{-1})	0.234
Crystal Size [mm]	0.16 x 0.17 x 0.24
Temperature (K)	293
Radiation	Mo*K*α 0.71073
θ Min-Max [°]	3.31, 28.87
h, k, l	-5:5; -8:10; -26:22
Tot.,UniqData, R(int)	5013, 3051, 0.0264
Obs. data [I > 2.0 σ(I)]	1550
Nref, Npar	3580, 181
R1, wR2, S	0.0590, 0.1104, 1.019
Min. - Max. resd. dens. [e/ Å^3]	-0.205, 0.167
CCDC	**978009**

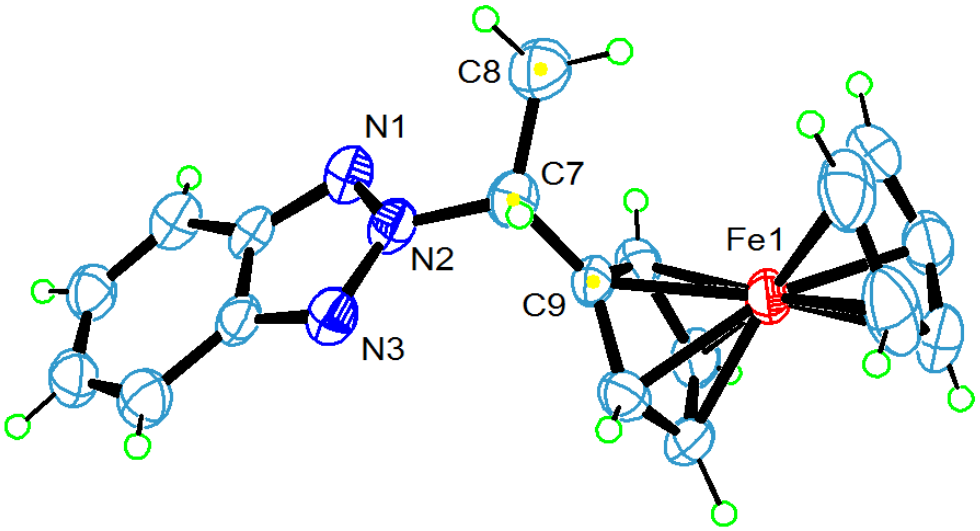


Selected bond length (Å): N1-N2 1.356, N1-C1 1.359, N1-C7 1.476, N2-N3 1.299, N3-C6 1.373; **Selected bond angles (º):** N2-N1-C1 109.9, N2-N1-C7 118.5, C1-N1-C7 131.4, N1-N2-N3 108.9, N2-N3-C6 109.0, N1-C1-C2 134.6, N1-C1-C6 104.6, N3-C6-C1 107.6.

Figure S2. Molecular structure of compound **31**. Thermal ellipsoids of carbon, nitrogen and iron are set at 40 % probability.

Table S2. Crystallographic refinement data for compound **31**

Compound 31 (*N*1)	
Empirical Formula	**C_{18} H_{17} Fe N_3**
Formula Weight	331.20
Crystal System	Monoclinic
Space group	P 1 21/n 1
a (Å)	6.6888(7)
b (Å)	28.847(2)
c (Å)	8.3300(10)
β (°)	104.396(11)
V (Å^3)	1556.8(3)
Z	4
Density (calc)	1.413
F(000)	688.0
μ (mm^{-1})	0.968
Crystal Size [mm]	0.13 x 0.15 x 0.23
Temperature (K)	293
Radiation	Mo*K*α 0.71073
θ Min-Max [°]	3.22, 29.13
h, k, l	-8:8; -18:39; -5:11
Tot.,UniqData, R(int)	6934, 3558, 0.0425
Obs. data [I > 2.0 σ(I)]	2076
Nref, Npar	4193, 259
R1, wR2, S	0.0694, 0.1545, 1.097
Min. - Max. resd. dens. [e/ Å^3]	-0.288, 0.370
CCDC	**978010**



Selected bond length (Å): N2-N3 1.33, N2-N1 1.32, N2-C7 1.48, N3-C6 1.36, N1-C1 1.35; **Selected bond angles (º):** N3-N2-N1 117.6, N3-N2-C7 120.1, N1-N2-C7 121.8, N2-N3-C6 102.7, N2-N1-C1 102.4, N1-C1-C2 130.0, N1-C1-C6 109.2, N3-C6-C5 129, N2-C7-C9 104.8, N2-C7-C8 111.3, N2-C7-H7 101, N3-C6-C5 131.1, N3-C6-C1 108.1.

Figure S3. Molecular structure of compound **32**. Thermal ellipsoids of carbon, nitrogen and iron are set at 40 % probability.

Table S3. Crystallographic refinement data for compound **32**

Compound 32 (*N*2)	
Empirical Formula	**C_{18} H_{17} Fe N_3**
Formula Weight	331.20
Crystal System	Monoclinic
Space group	P 1 21 1
a (Å)	6.1853(6)
b (Å)	30.023(3)
c (Å)	8.4797(9)
β (°)	104.478(10)
V (Å^3)	1524.7(3)
Z	4
Density (calc)	1.443
F(000)	688
μ (mm^{-1})	0.988
Crystal Size [mm]	0.12 x 0.18 x 0.20
Temperature (K)	293
Radiation	Mo*K*α 0.71073
θ Min-Max [°]	3.40, 29.08
h, k, l	-8:7; -38:39; -9:11
Tot.,UniqData, R(int)	6476, 4835, 0.0328
Obs. data [I > 2.0 σ(I)]	3163
Nref, Npar	4835, 433
R1, wR2, S	0.0611, 0.1212, 1.046
Min. - Max. resd. dens. [e/ Å^3]	-0.349, 0.416
CCDC	**978011**