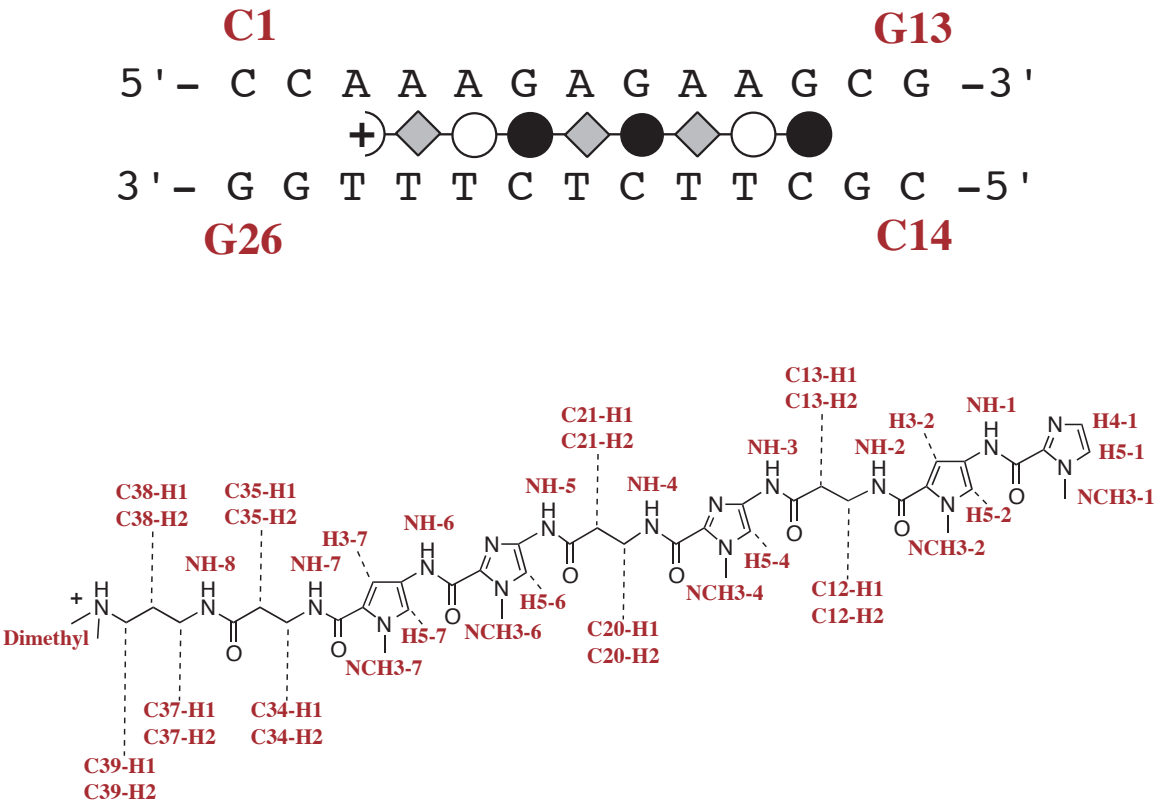
Appendix A

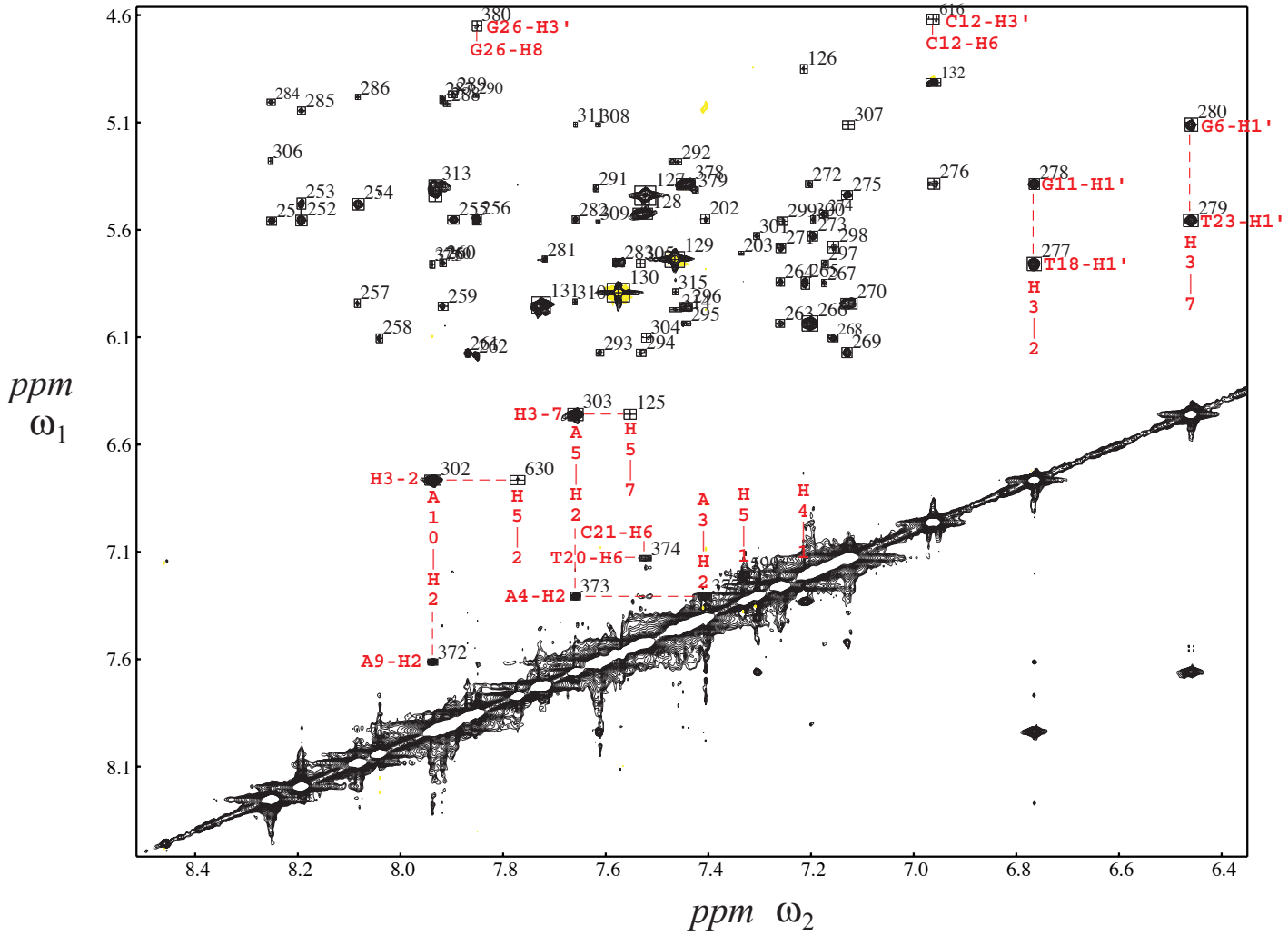
NMR Spectra



Expanded and annotated NOESY spectra in D2O and H2O (90%) are provided here for the 1:1 polyamide:DNA complex illustrated above. The binding model shown at top gives DNA base numbers used in the assignments, as well as the orientation and binding site of the ligand. The chemical structure drawn below is annotated with all proton nomenclature. Due to the similarity of the correlations assigned in each plot, assignments are typically annotated with the DNA residue number, and the correlation type is given in the title of the plot. Special legends are provided for complicated regions. Experimental conditions and NOESY mixing time is given above each plot. Eleven regions of the D2O NOESY spectrum are given prior to nine regions of the H2O NOESY spectrum. A 75 ms mixing time was used in both experiments.

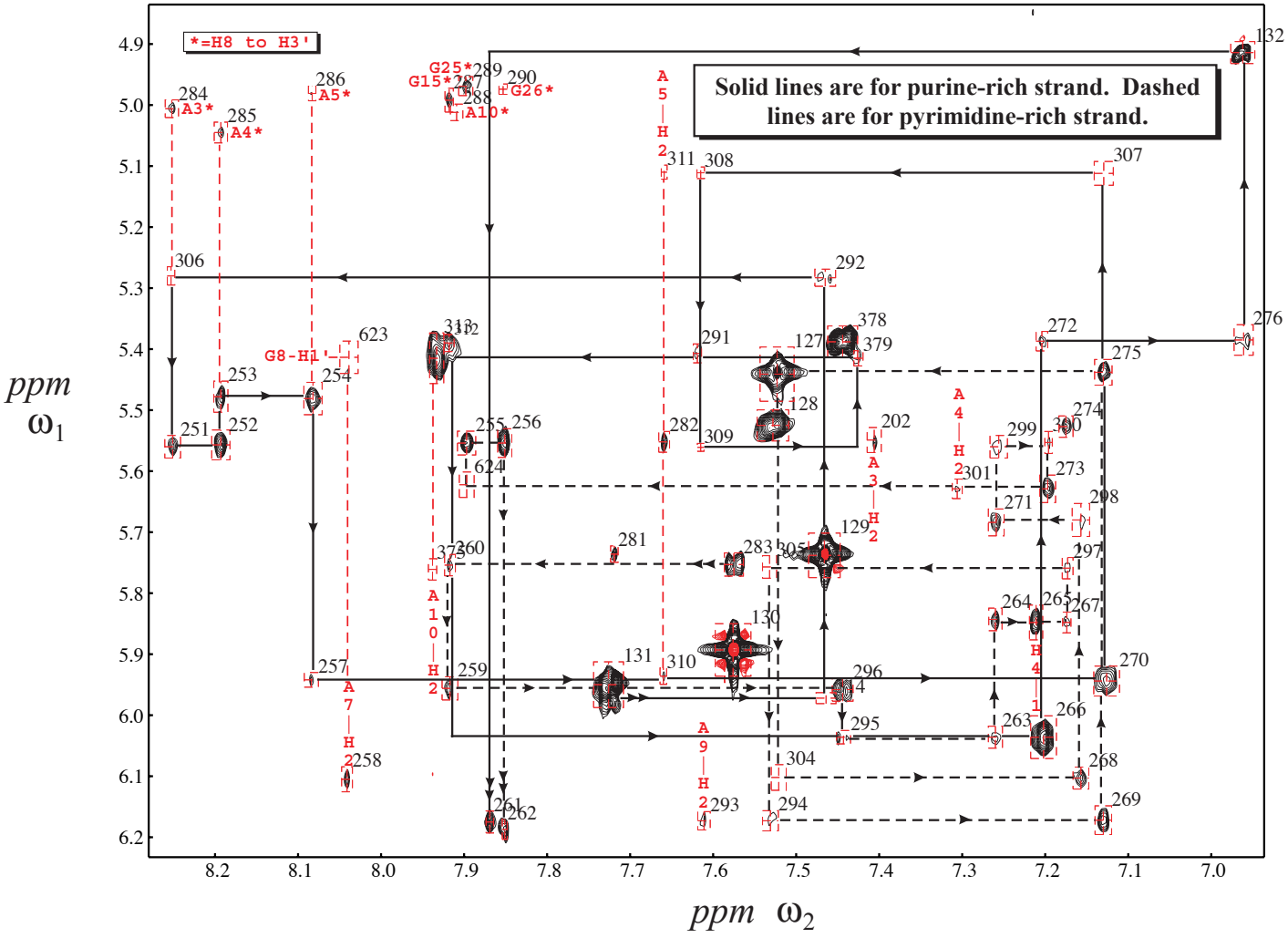
Aromatic Region of NOESY spectrum in D_2O

(t_{mix} = 75 ms., 600 MHz, 25℃)



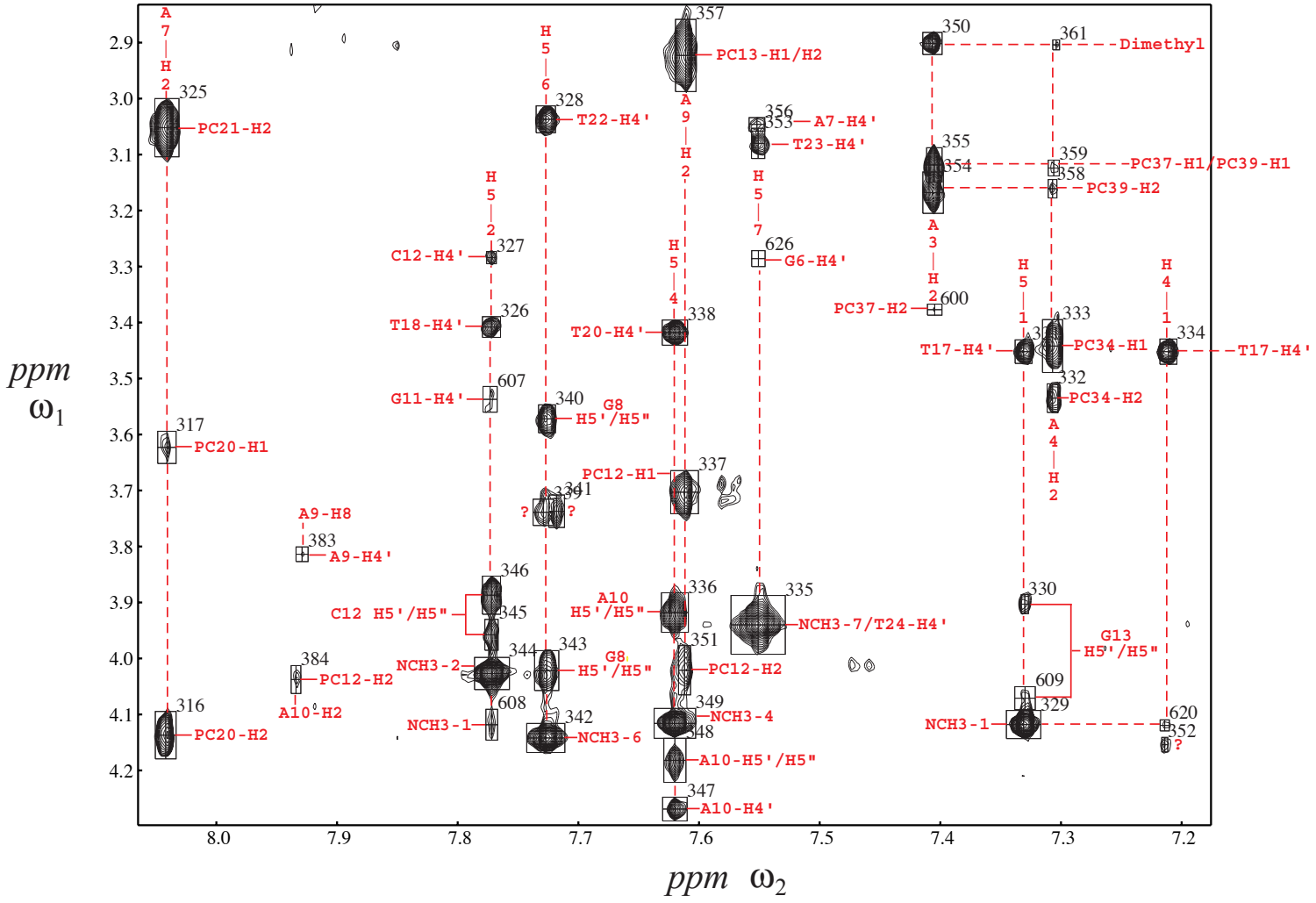
H1' to Aromatic Region of NOESY spectrum in D_2O

(t_{mix} = 75 ms., 600 MHz, 25°C)



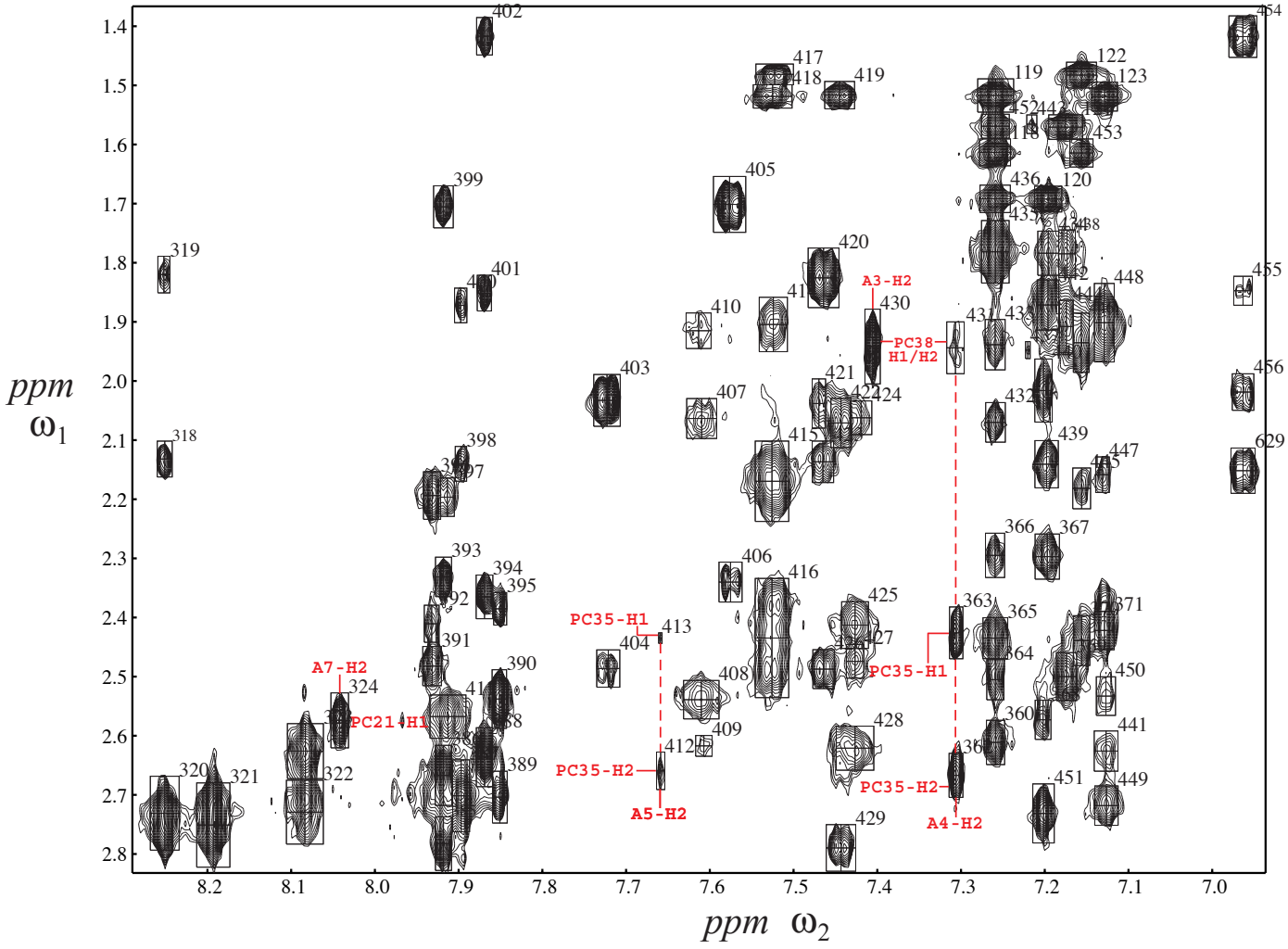
Aromatic to H4' and NCH_3 Region of NOESY spectrum in D_2O

(t_{mix}= 75 ms., 600 MHz, 25°C)



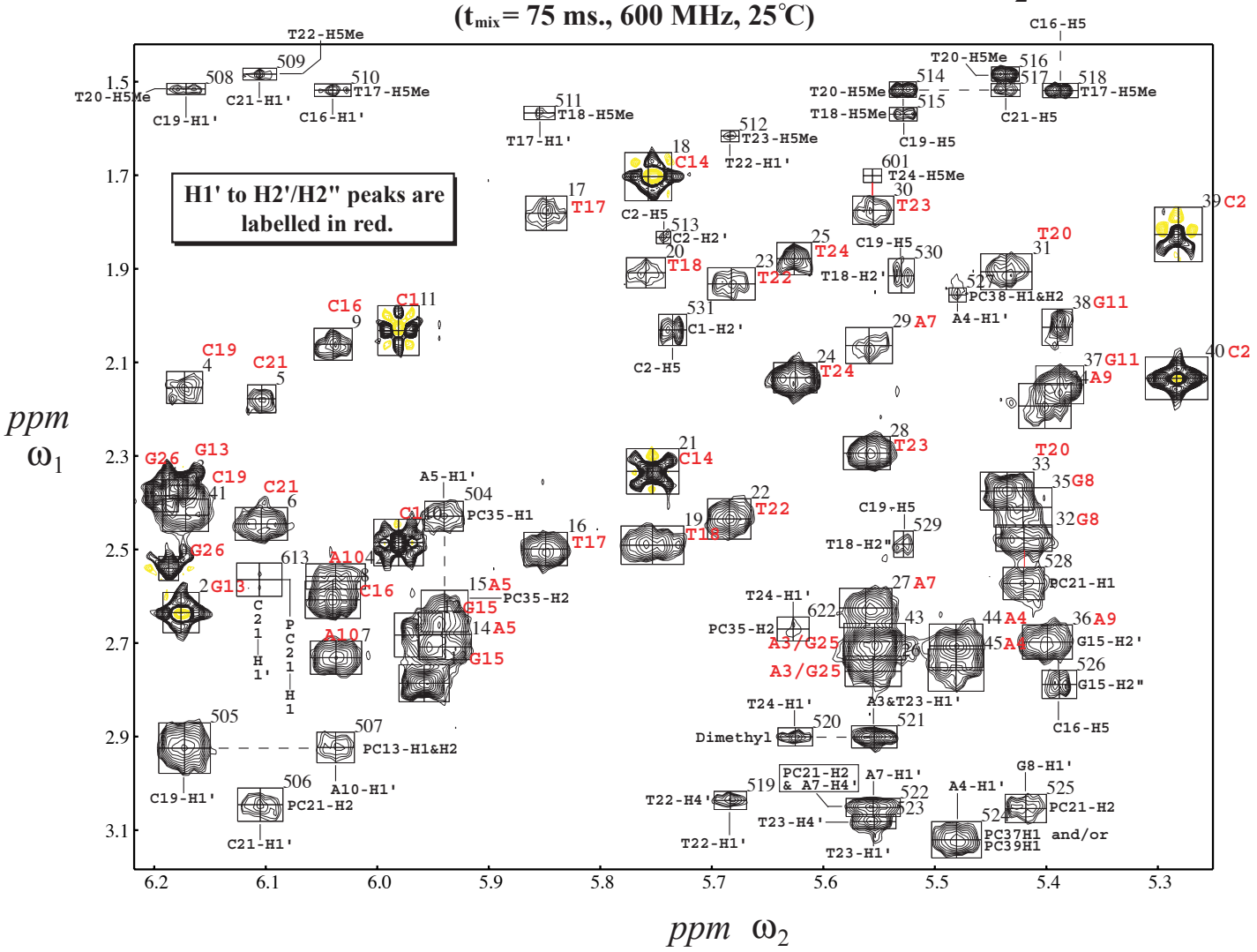
Aromatic to H2'/H2" Region of NOESY spectrum in D_2O

(t_{mix}= 75 ms., 600 MHz, 25°C)



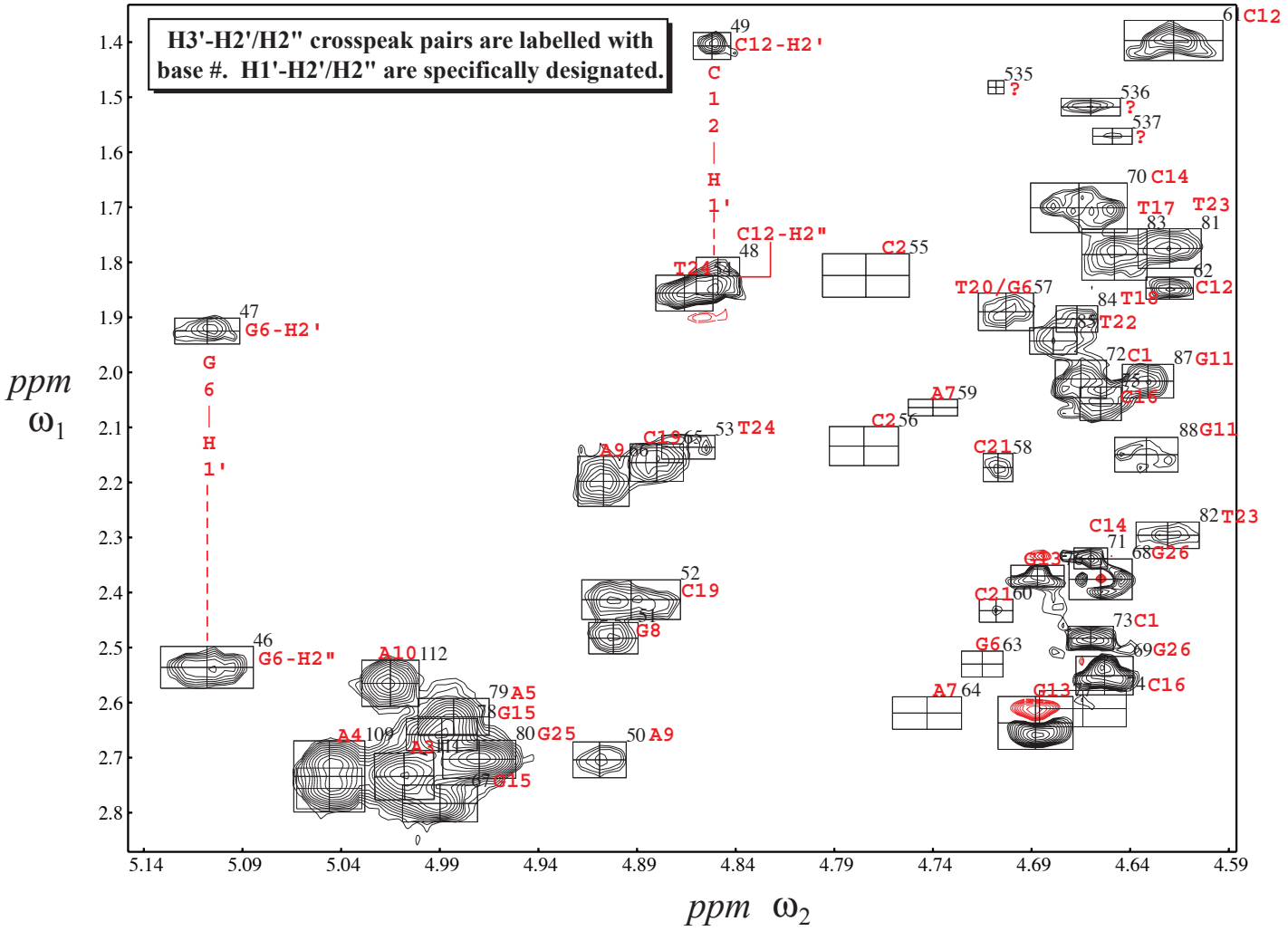
H1' to H2'/H2" Region of NOESY spectrum in D_2O

(t_{mix} = 75 ms., 600 MHz, 25°C)



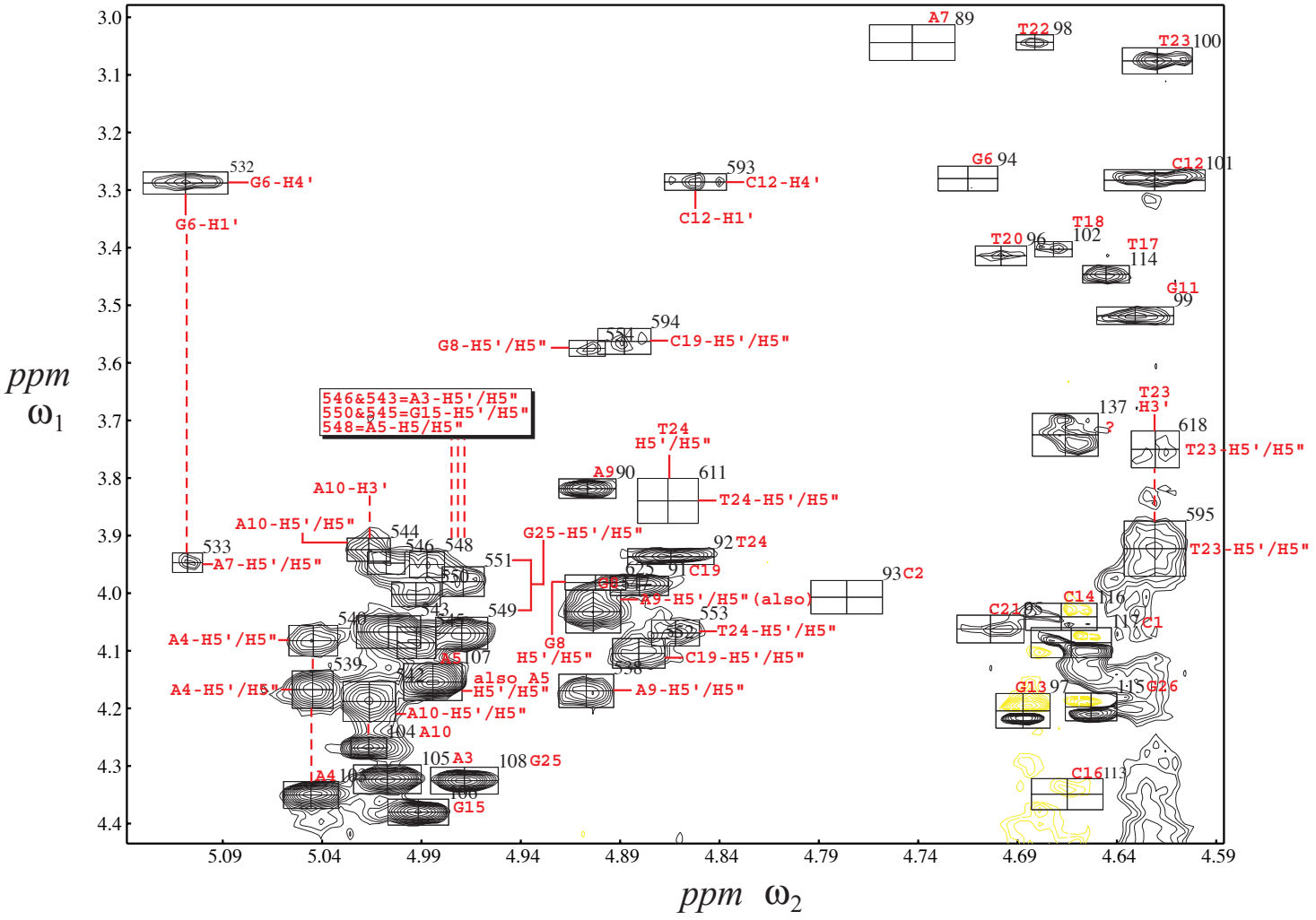
H3' to H2'/H2" Region of NOESY spectrum in D_2O

(t_{mix} = 75 ms., 600 MHz, 25°C)



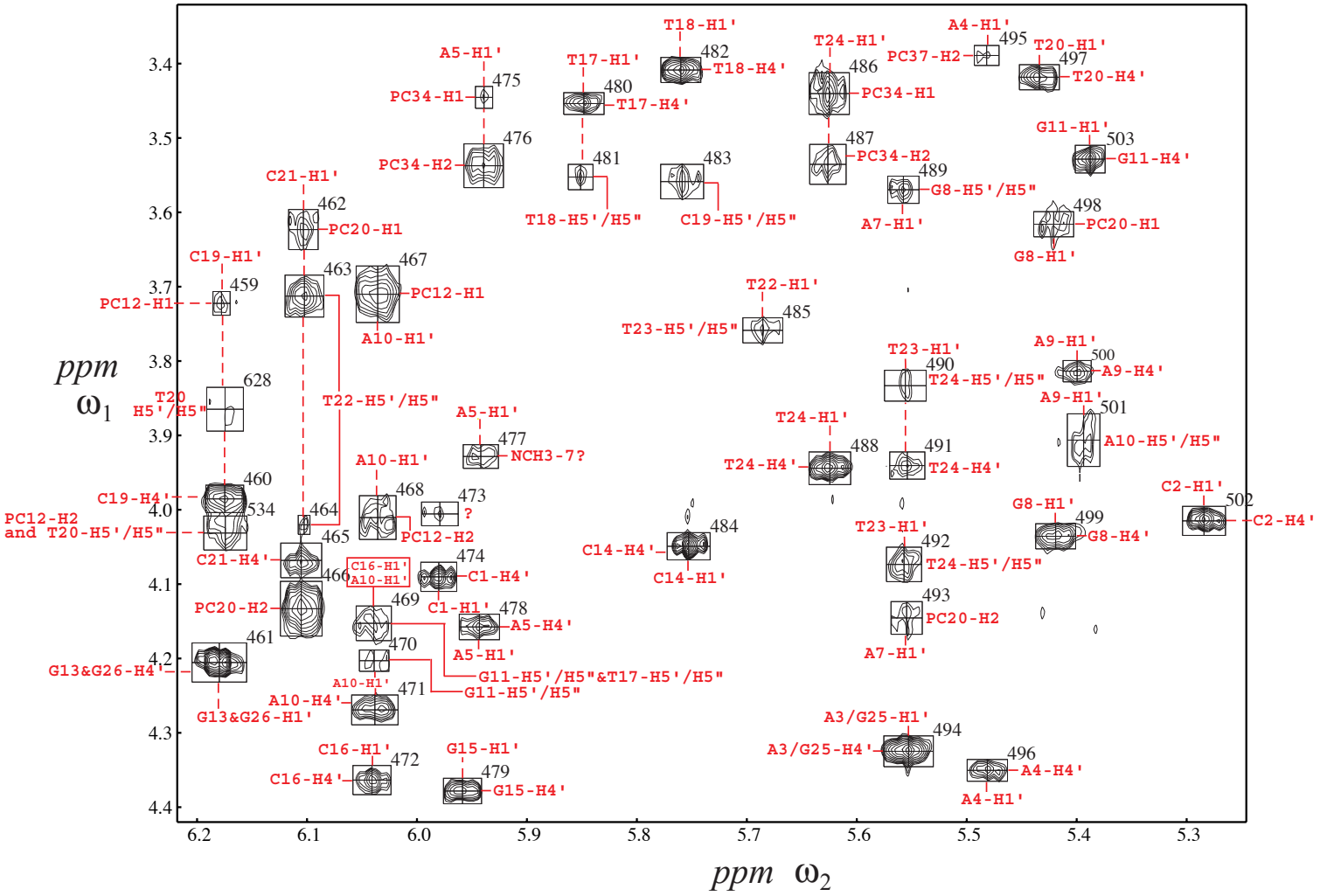
H3' to H4'/H5'/H5" Region of NOESY spectrum in D_2O

(t_{mix}= 75 ms., 600 MHz, 25℃)



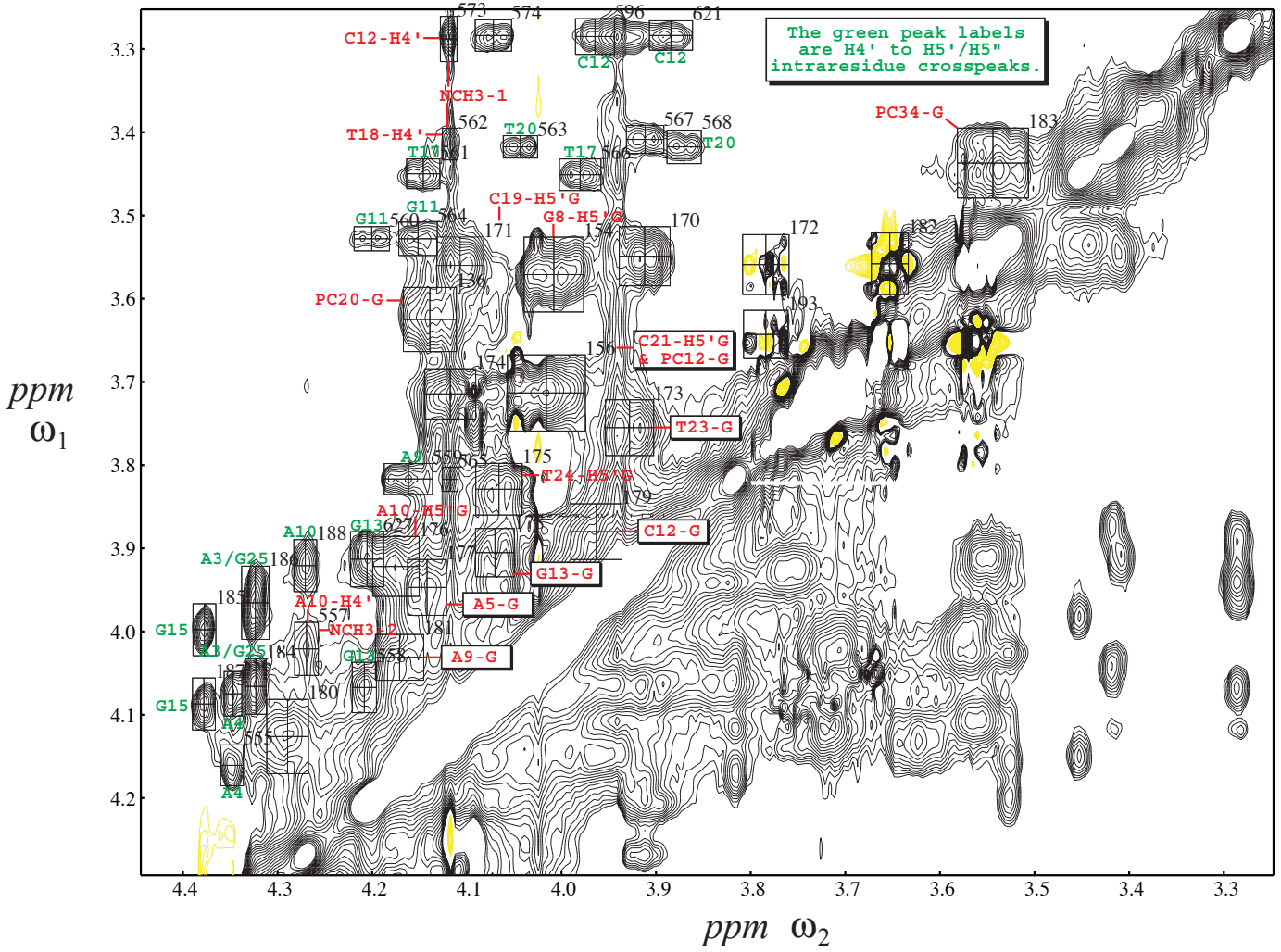
H1' to H4' Region of NOESY spectrum in D_2O

(t_{mix} = 75 ms., 600 MHz, 25°C)



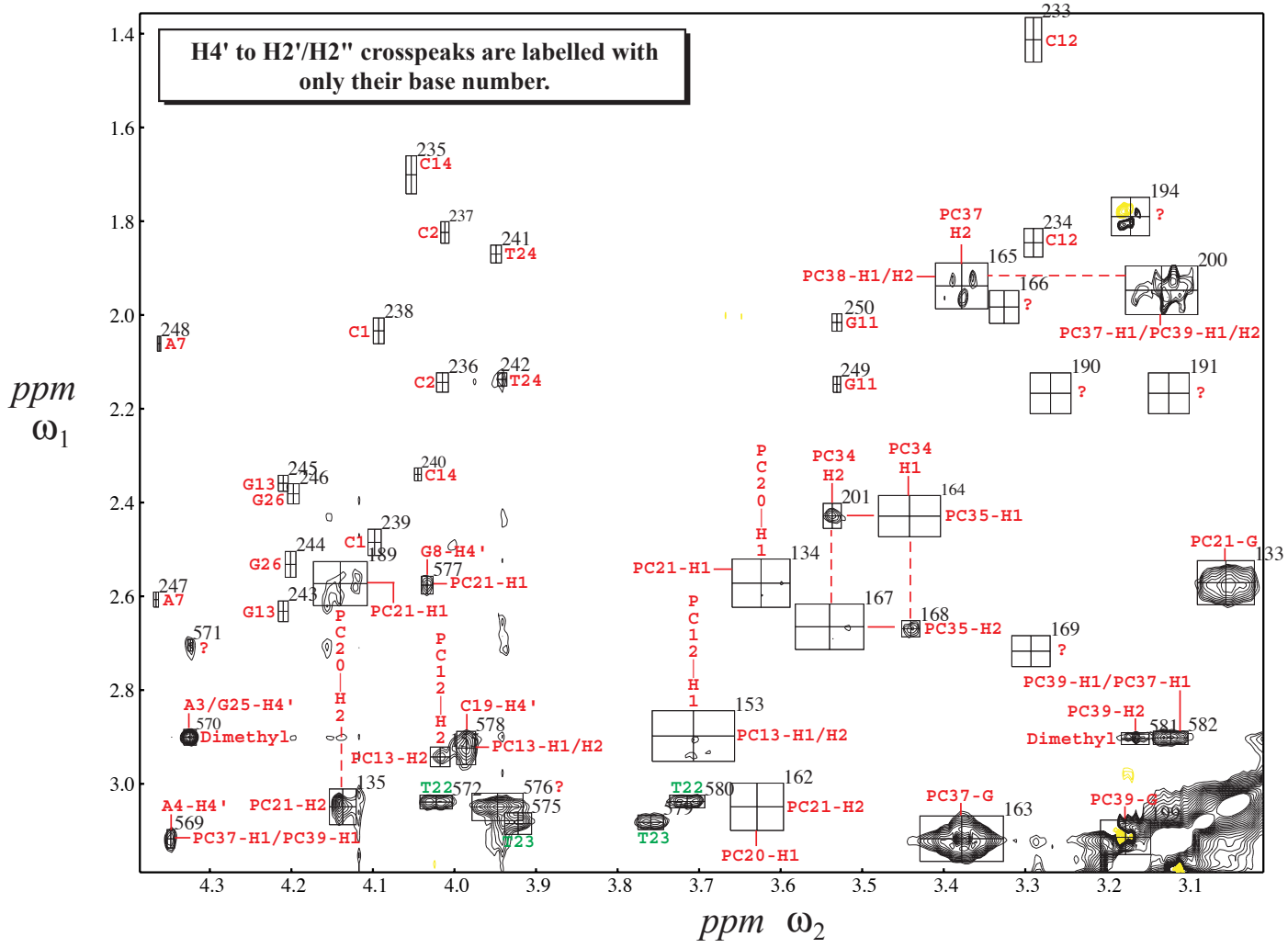
H5' to H5" Region of NOESY spectrum in D_2O

(t_{mix} = 75 ms., 600 MHz, 25°C)



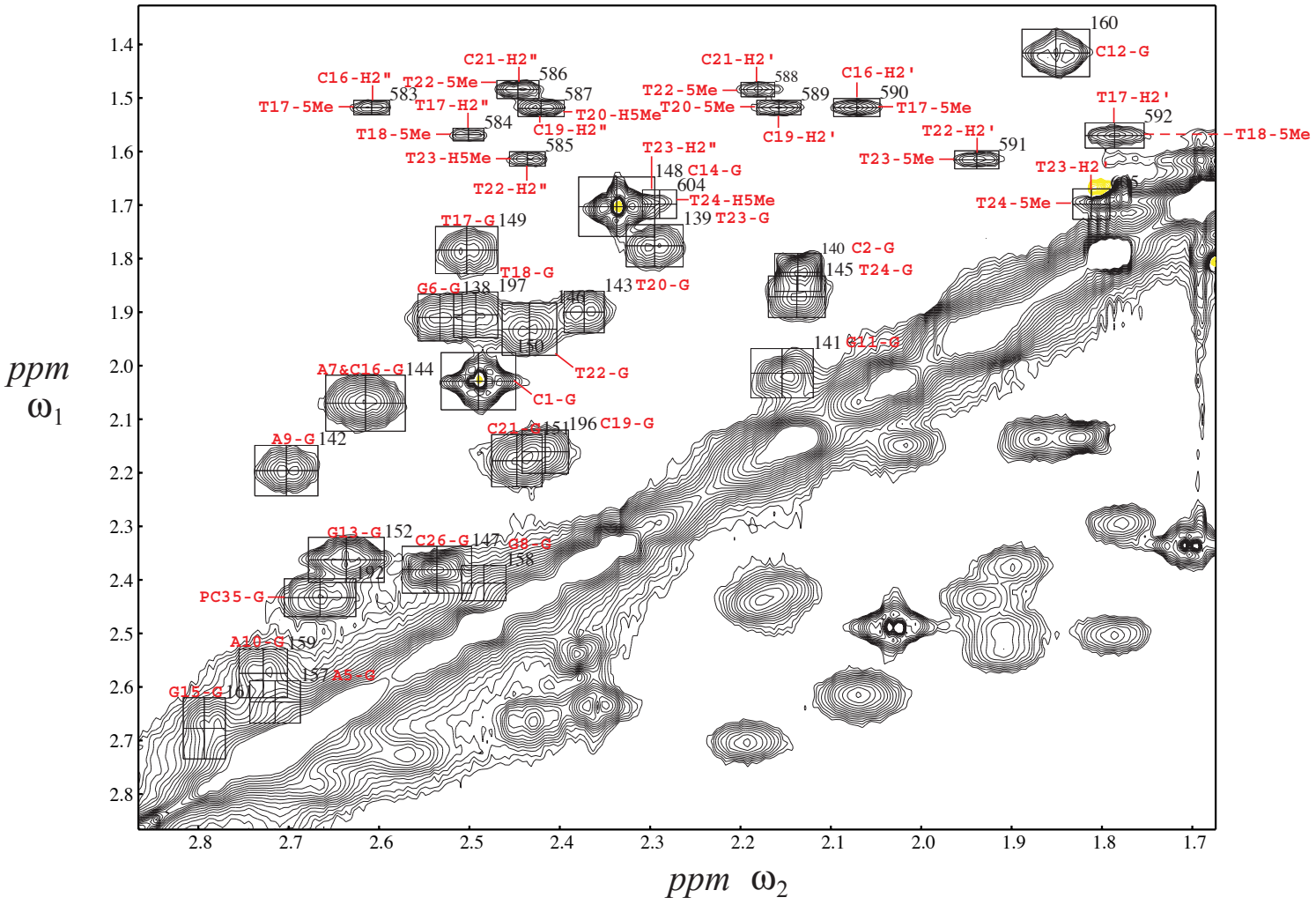
Methylene Region of NOESY spectrum in D_2O

(t_{mix} = 75 ms., 600 MHz, 25°C)



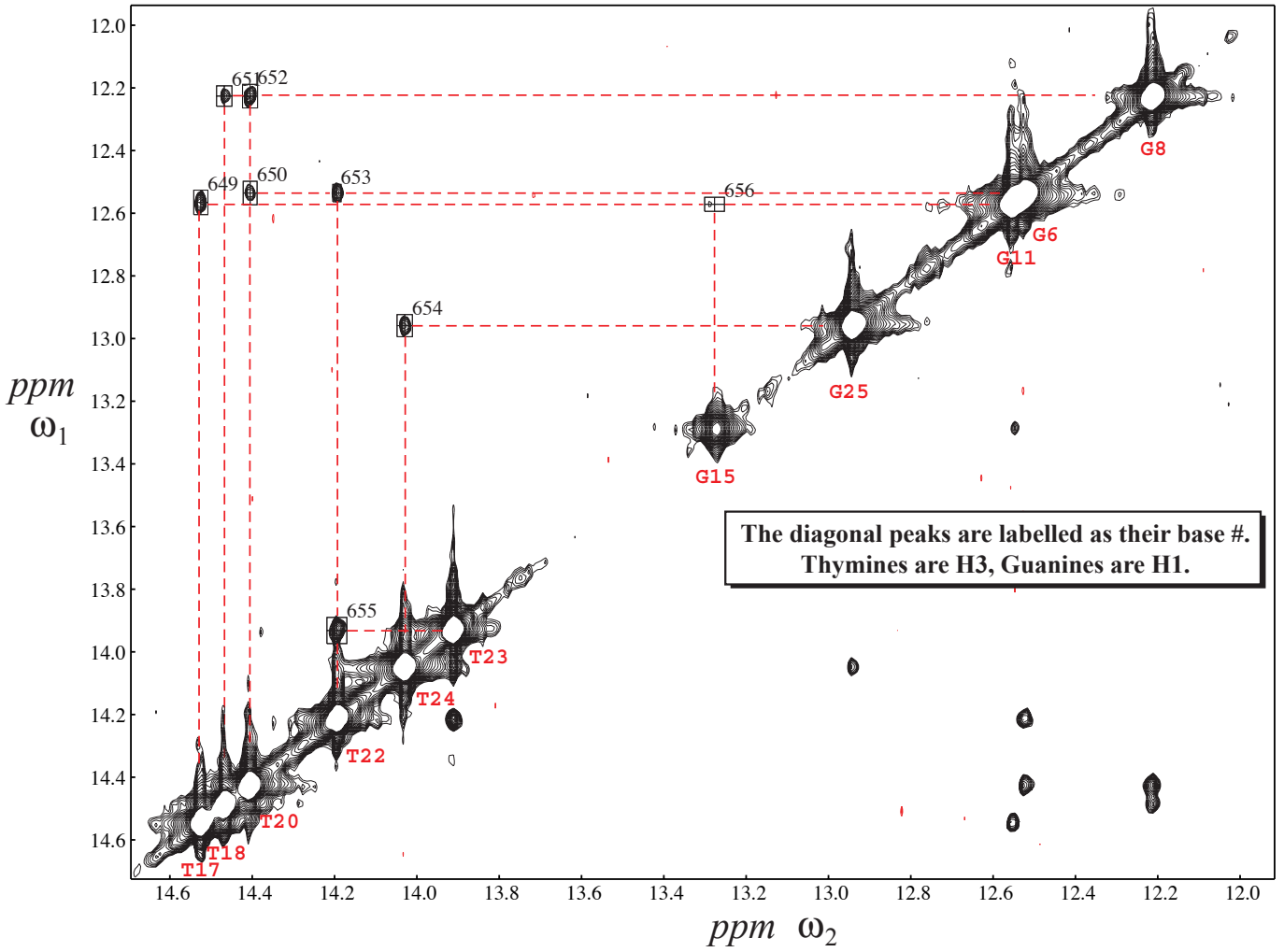
H2' to H2" Region of NOESY spectrum in D_2O

(t_{mix} = 75 ms., 600 MHz, 25℃)



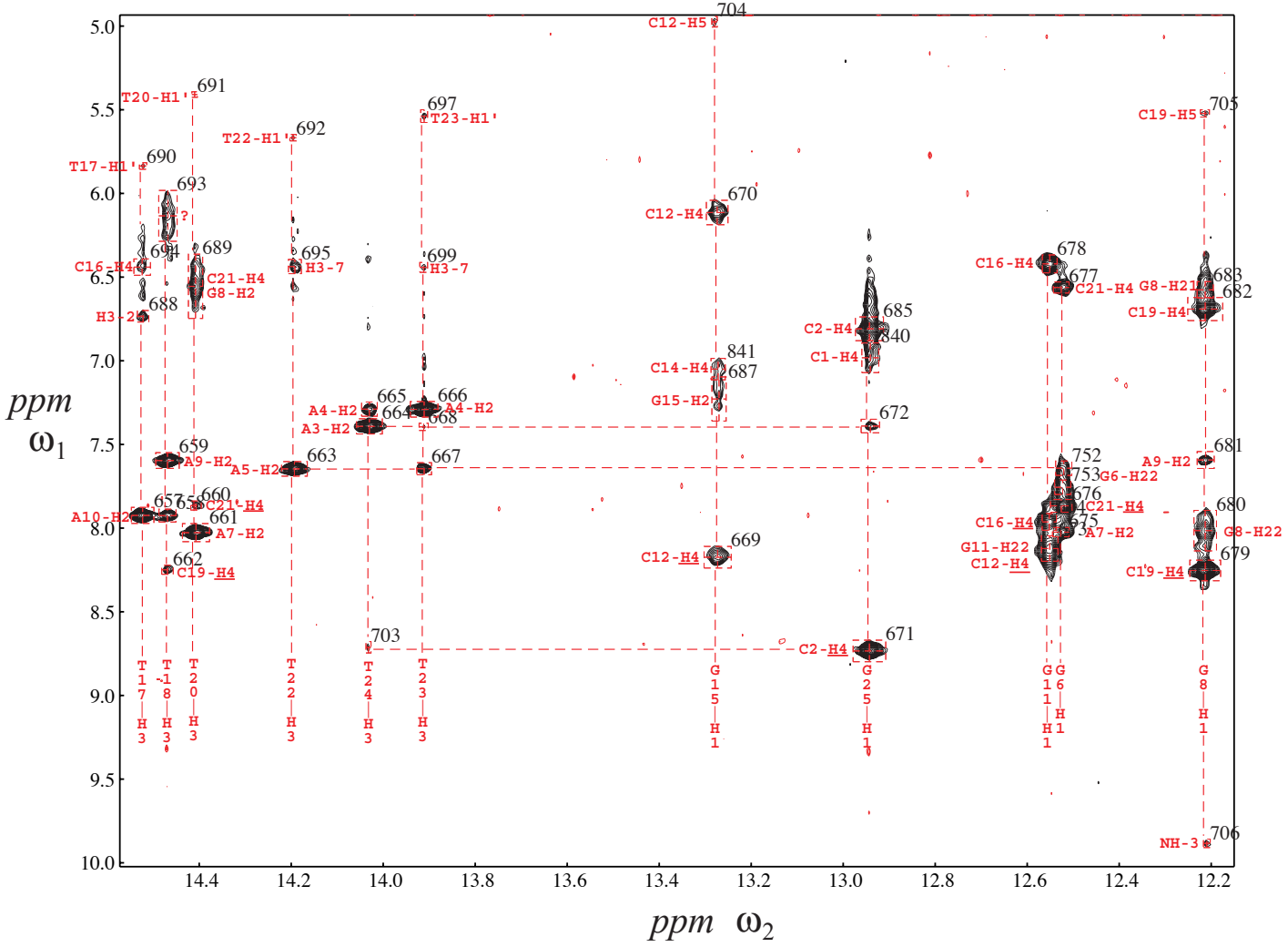
Sequential Imino Region of NOESY spectrum in H_2O

(t_{mix}= 75 ms., 600 MHz, 25°C)



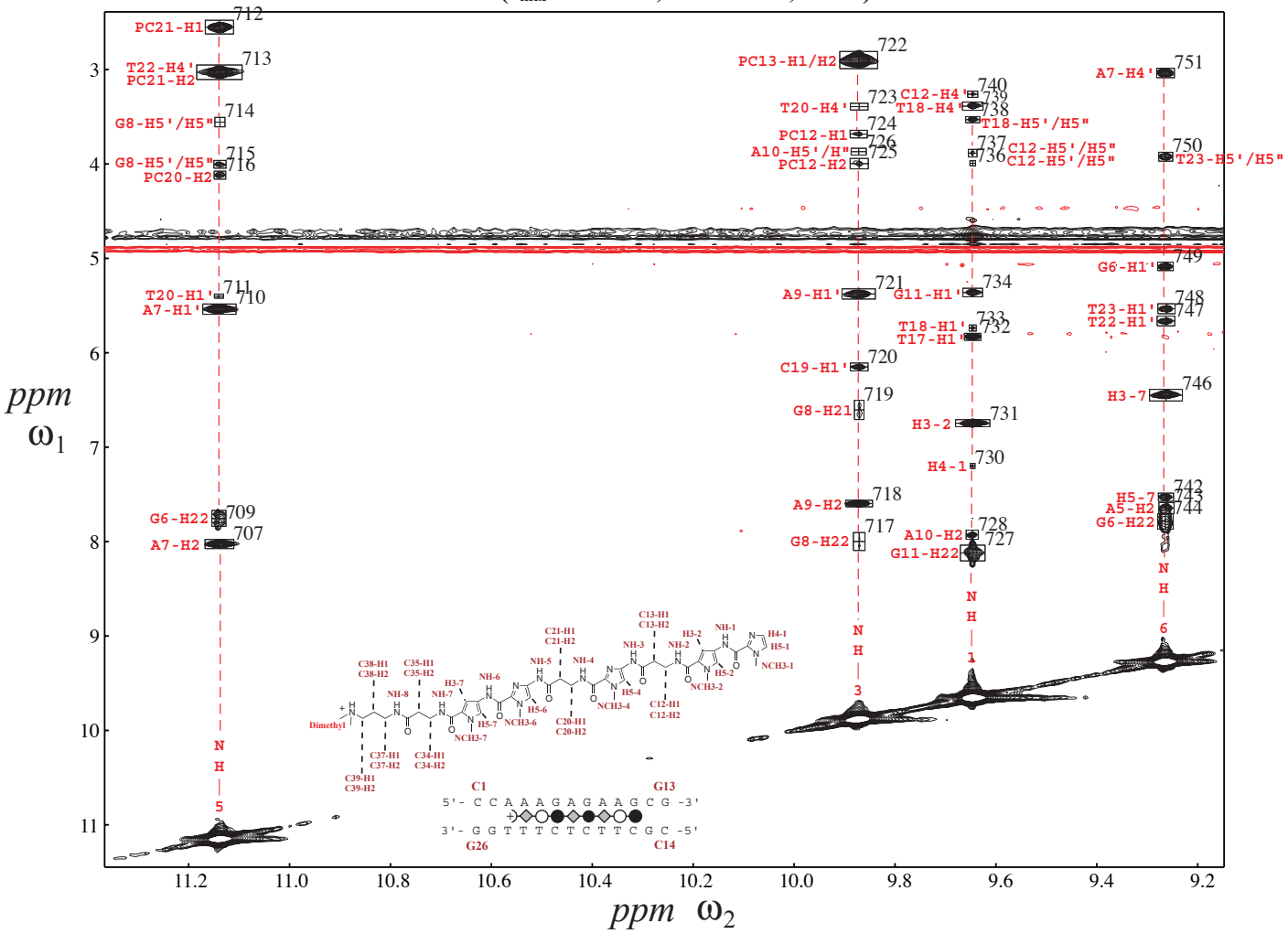
Imino to Amino and AH2 Region of NOESY spectrum in H_2O

(t_{mix}= 75 ms., 600 MHz, 25°C)



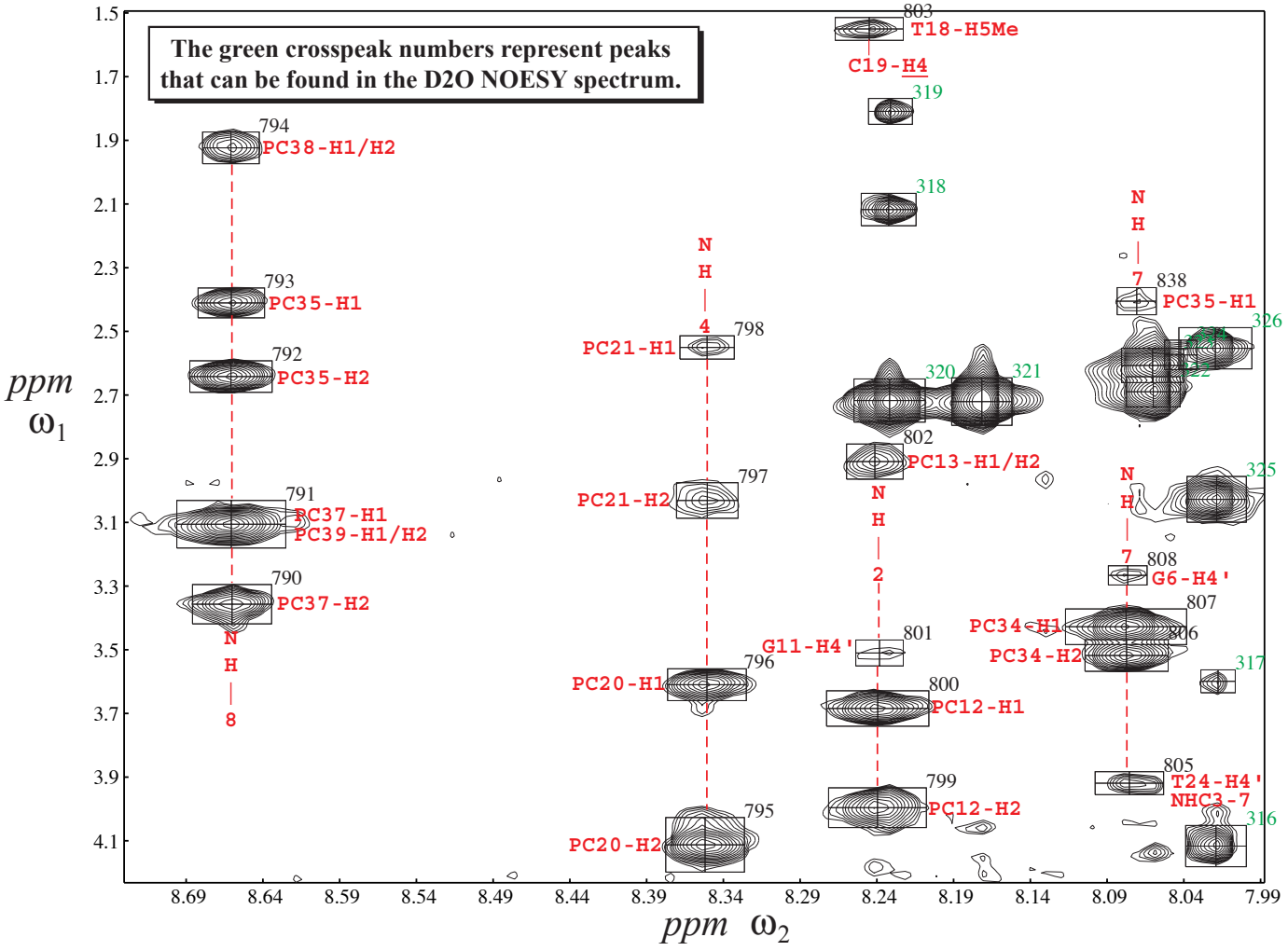
Aromatic Amide Region of NOESY spectrum in H_2O

(t_{mix} = 75 ms., 600 MHz, 25°C)



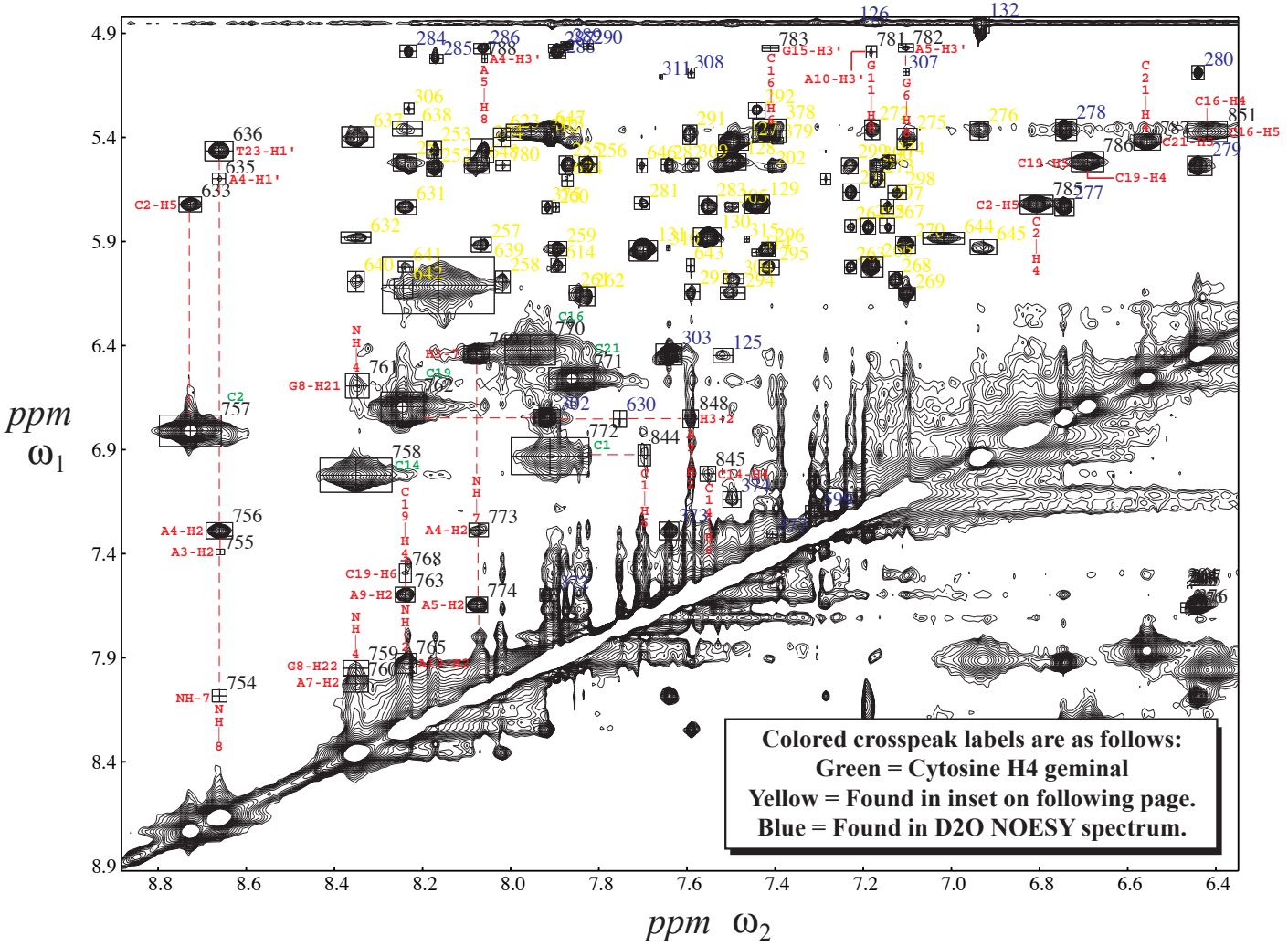
Aliphatic Amide Region of NOESY spectrum in H_2O

(t_{mix}= 75 ms., 600 MHz, 25°C)



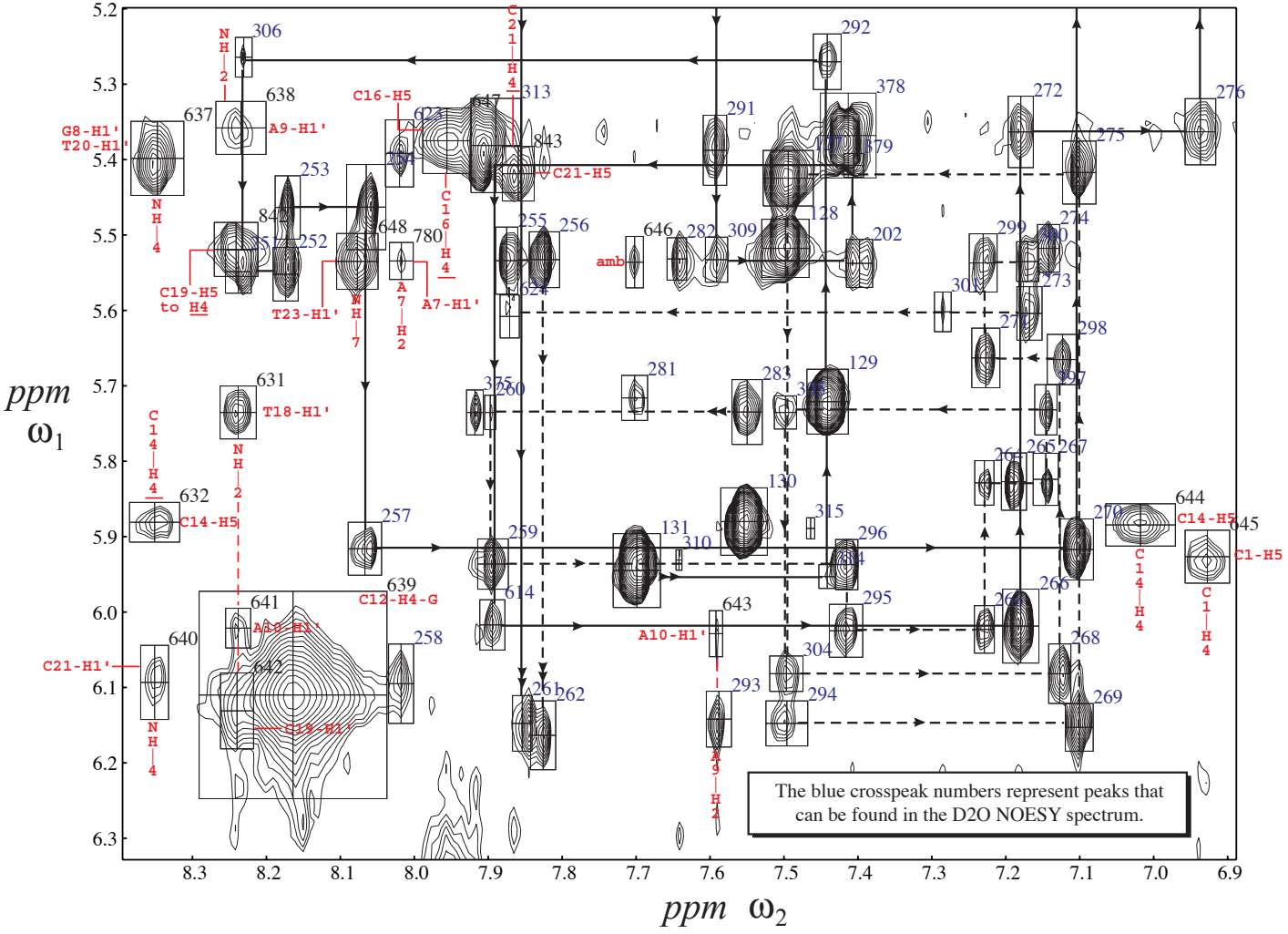
Aromatic Region of NOESY spectrum in H_2O

(t_{mix}= 75 ms., 600 MHz, 25°C)



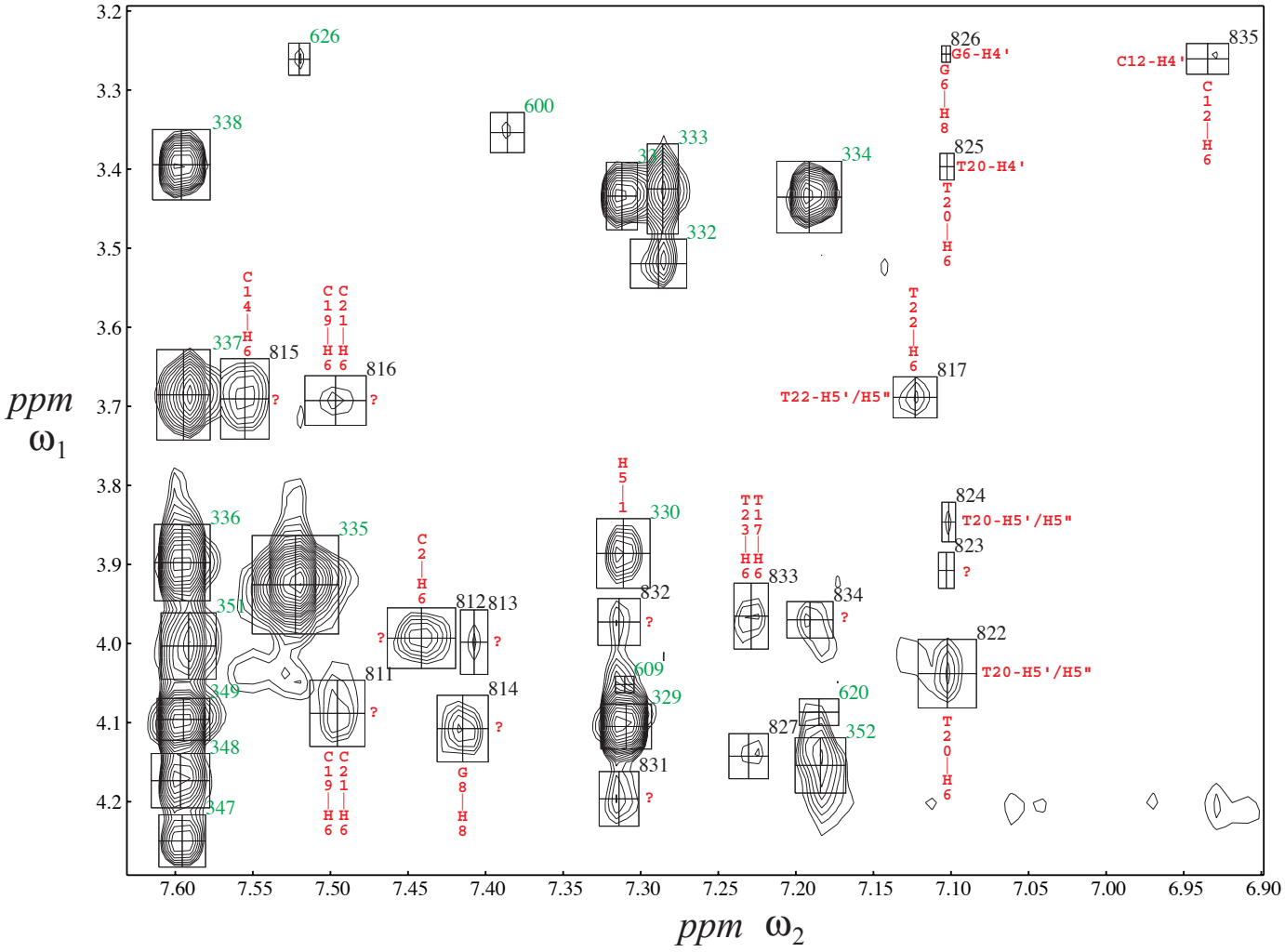
Aromatic to H1' Region of NOESY spectrum in H_2O

(t_{mix}= 75 ms., 600 MHz, 25°C)



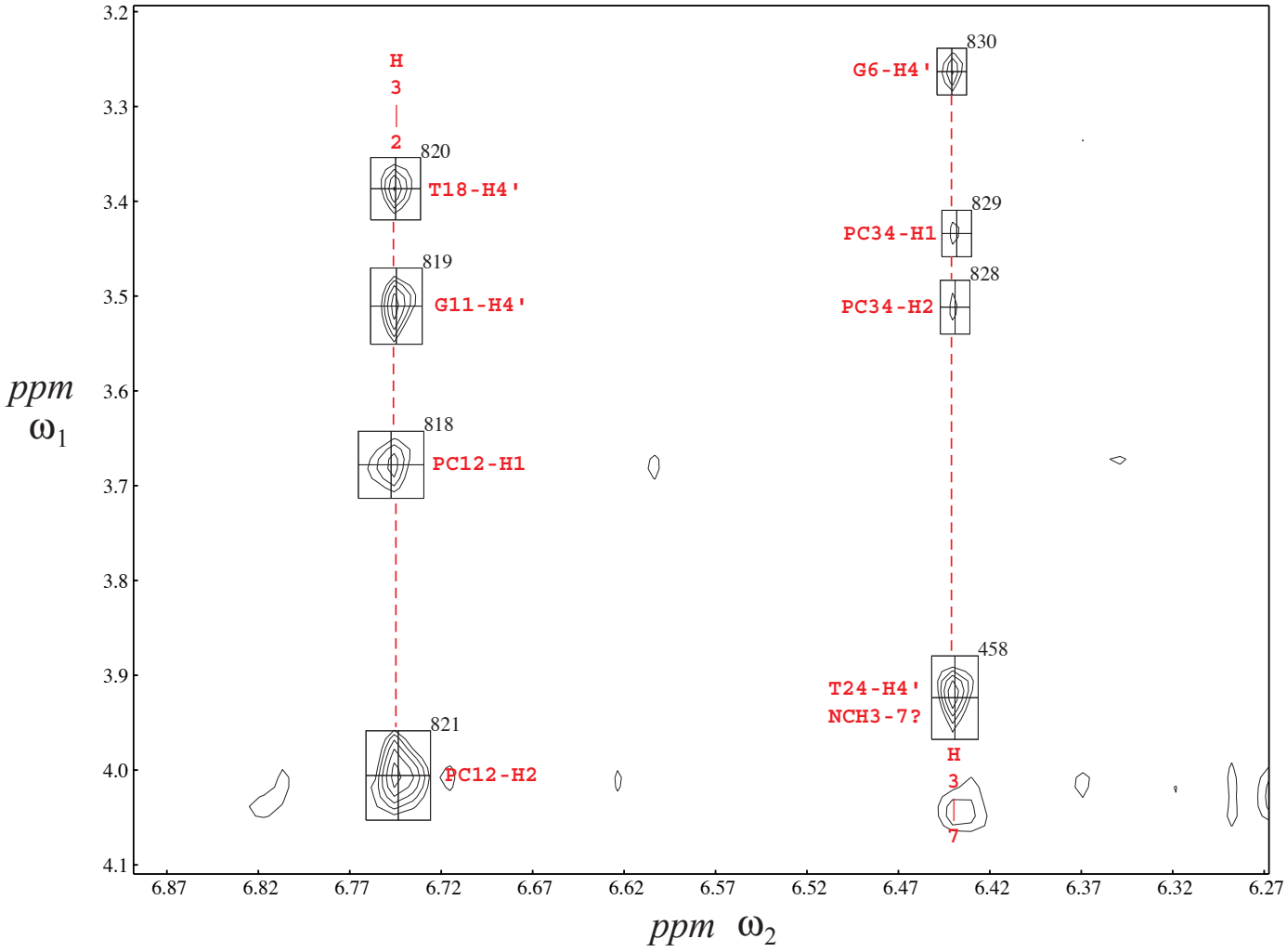
Aromatic to H4'/NCH3 Region of NOESY spectrum in H_2O

(t_{mix} = 75 ms., 600 MHz, 25°C)



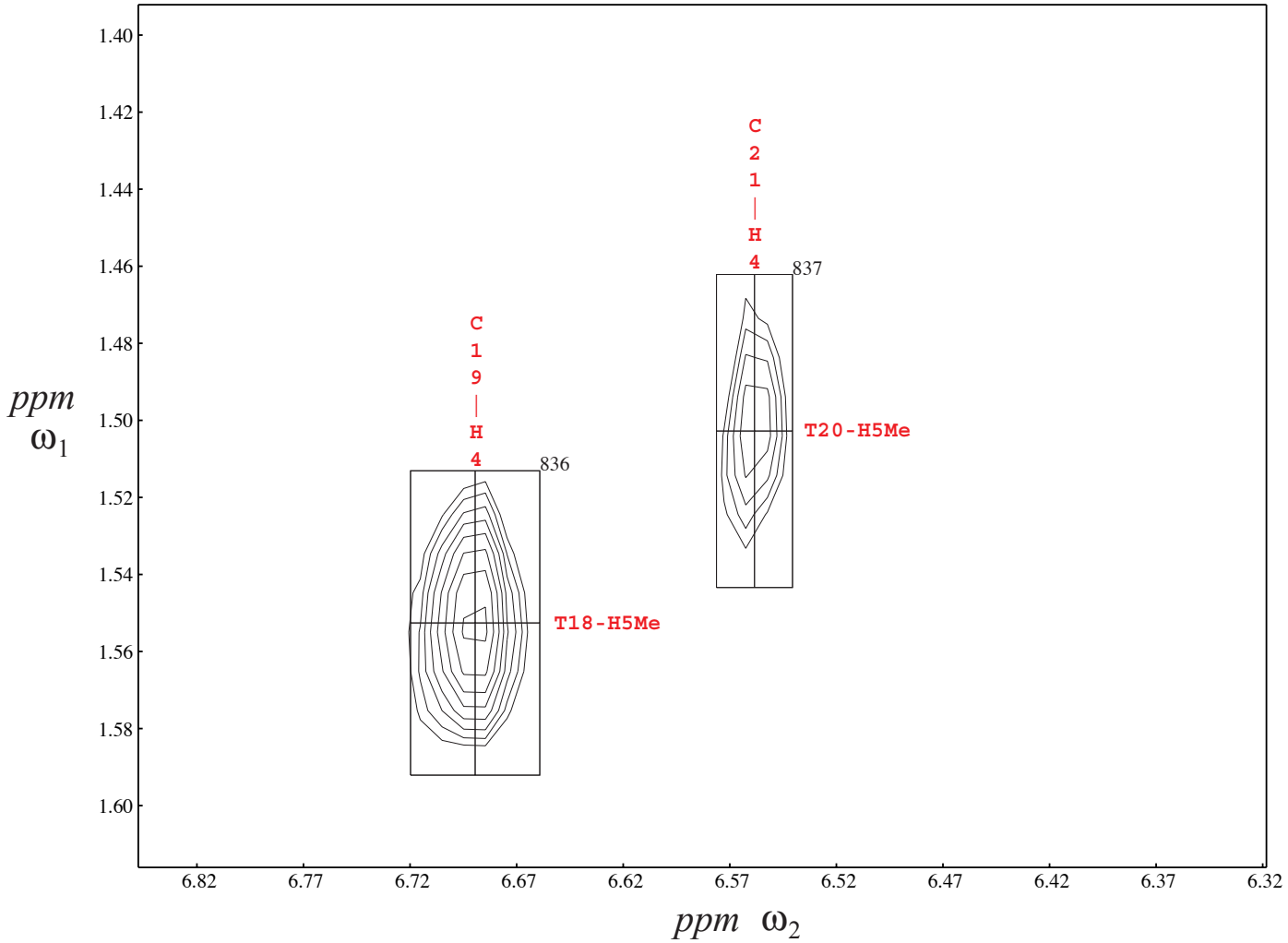
Pyrrole H3 Region of NOESY spectrum in H_2O

(t_{mix}= 75 ms., 600 MHz, 25°C)



CH4 to TH5Me Region of NOESY spectrum in H_2O

(t_{mix}= 75 ms., 600 MHz, 25°C)



Appendix B

Distance Constraints

A complete list of all constraints used in the structure calculations of the 1:1 polyamide-DNA complex is provided here. Methods for obtaining these restraints, based on the NOESY spectra in Appendix A, and are described in the Experimental.

1) Intraresidue DNA to DNA Constraints

residue number	residue name	atom name	residue number	residue name	atom name	upper bound
1	CYT	H2'1	1	CYT	H1'	4.25
1	CYT	H2'2	1	CYT	H1'	2.8
1	CYT	H4'	1	CYT	H1'	3.58
1	CYT	H4'	1	CYT	H2'1	4.38
1	CYT	H4'	1	CYT	H2'2	5.5
1	CYT	H6	1	CYT	H2'1	2.8
1	CYT	H6	1	CYT	H2'2	4.78
2	CYT	H2'1	2	CYT	H6	2.75
2	CYT	H2'2	2	CYT	H1'	2.87
2	CYT	H4'	2	CYT	H2'1	4.36
2	CYT	H4'	2	CYT	H2'2	5.5
2	CYT	H6	2	CYT	H1'	4.48
3	ADE	H1'	3	ADE	H3'	5.57
3	ADE	H1'	3	ADE	H4'	5.5
3	ADE	H1'	3	ADE	H8	4.29
3	ADE	H3'	3	ADE	H5'1	5.5
3	ADE	H3'	3	ADE	H5'2	5.5
3	ADE	H3'	3	ADE	H8	4.96
3	ADE	H4'	3	ADE	H3'	3.3
3	ADE	H8	3	ADE	H2'1	5.5
3	ADE	H8	3	ADE	H2'2	5.5
4	ADE	H3'	4	ADE	H1'	4.88
4	ADE	H3'	4	ADE	H8	5.04
4	ADE	H4'	4	ADE	H1'	4.18
4	ADE	H4'	4	ADE	H3'	3.22
4	ADE	H8	4	ADE	H1'	4.23
4	ADE	H8	4	ADE	H2'1	5.5
4	ADE	H8	4	ADE	H2'2	5.5
4	ADE	Q5'	4	ADE	H3'	4.27
5	ADE	H2	5	ADE	H1'	4.83
5	ADE	H3'	5	ADE	H5'1	5.5
5	ADE	H3'	5	ADE	H5'2	5.5
5	ADE	H4'	5	ADE	H1'	3.65
5	ADE	H8	5	ADE	H1'	4.35
5	ADE	H8	5	ADE	H2'1	5.5
5	ADE	H8	5	ADE	H2'2	5.5
6	GUA	H2'1	6	GUA	H1'	4.2
6	GUA	H2'2	6	GUA	H1'	3.06
6	GUA	H4'	6	GUA	H1'	4.18
6	GUA	H8	6	GUA	H1'	5.76
6	GUA	H8	6	GUA	H2'1	5.5
7	ADE	H1'	7	ADE	H2'1	3.53

7	ADE	H1'	7	ADE	H4'	5.5
7	ADE	H1'	7	ADE	H8	5.67
7	ADE	H2'1	7	ADE	H8	3.38
7	ADE	H2'2	7	ADE	H8	4.57
7	ADE	H4'	7	ADE	H2'1	5.85
7	ADE	H4'	7	ADE	H2'2	4.39
8	GUA	H3'	8	GUA	H2'2	4.1
8	GUA	H3'	8	GUA	Q5'	5.5
8	GUA	H8	8	GUA	H1'	5.83
8	GUA	H8	8	GUA	H2'1	5.5
8	GUA	H8	8	GUA	H2'2	5.5
9	ADE	H1'	9	ADE	H4'	4.17
9	ADE	H2'2	9	ADE	H3'	3.6
9	ADE	H3'	9	ADE	H1'	4.22
9	ADE	H3'	9	ADE	H4'	4.13
9	ADE	H8	9	ADE	H1'	5.5
9	ADE	H8	9	ADE	H2'1	5.5
9	ADE	Q5'	9	ADE	H4'	3.37
10	ADE	H1'	10	ADE	H2'1	5.5
10	ADE	H1'	10	ADE	H4'	3.62
10	ADE	H2	10	ADE	H1'	4.89
10	ADE	H2'1	10	ADE	H3'	3.05
10	ADE	H2'2	10	ADE	H1'	2.95
10	ADE	H3'	10	ADE	H1'	5
10	ADE	H3'	10	ADE	H4'	3.42
10	ADE	H3'	10	ADE	H5'1	5.5
10	ADE	H3'	10	ADE	H5'2	5.5
10	ADE	H4'	10	ADE	Q5'	3.38
10	ADE	H8	10	ADE	H2'1	2.93
11	GUA	H1'	11	GUA	H4'	4.19
11	GUA	H2'1	11	GUA	H1'	3.57
11	GUA	H2'1	11	GUA	H4'	5.82
11	GUA	H2'1	11	GUA	H8	3.18
11	GUA	H2'2	11	GUA	H3'	4.54
11	GUA	H2'2	11	GUA	H4'	4.39
11	GUA	H3'	11	GUA	H4'	4.31
11	GUA	H4'	11	GUA	Q5'	3.47
11	GUA	H8	11	GUA	H1'	4.68
11	GUA	H8	11	GUA	H2'2	5.5
12	CYT	H2'1	12	CYT	H3'	3.32
12	CYT	H2'1	12	CYT	H6	3.12
12	CYT	H2'2	12	CYT	H3'	3.67
12	CYT	H4'	12	CYT	H1'	4.53
12	CYT	H4'	12	CYT	H2'1	4.4
12	CYT	H4'	12	CYT	H3'	4.16
13	GUA	H1'	13	GUA	H2'2	3.12
13	GUA	H1'	13	GUA	H4'	5.5
13	GUA	H1'	13	GUA	H8	4.76
13	GUA	H2'2	13	GUA	H8	5.5
13	GUA	H4'	13	GUA	H2'1	5.5
13	GUA	H4'	13	GUA	H2'2	4.67
14	CYT	H2'1	14	CYT	H1'	4.38
14	CYT	H2'1	14	CYT	H6	2.86
14	CYT	H2'2	14	CYT	H1'	2.83
14	CYT	H3'	14	CYT	H2'1	3.25
14	CYT	H4'	14	CYT	H1'	3.5
14	CYT	H4'	14	CYT	H2'2	4.26

14	CYT	H6	14	CYT	H1'	3.64
15	GUA	H1'	15	GUA	H4'	3.64
15	GUA	H2'2	15	GUA	H1'	2.89
15	GUA	H3'	15	GUA	H4'	3.35
15	GUA	H3'	15	GUA	H5'1	5.5
15	GUA	H3'	15	GUA	H5'2	5.5
15	GUA	H3'	15	GUA	H8	4.46
15	GUA	H8	15	GUA	H1'	4.43
15	GUA	H8	15	GUA	H2'1	5.5
15	GUA	H8	15	GUA	H2'2	5.5
15	GUA	Q5'	15	GUA	H4'	3.35
16	CYT	H2'1	16	CYT	H1'	3.47
16	CYT	H3'	16	CYT	H1'	5.91
16	CYT	H4'	16	CYT	H1'	4.11
16	CYT	H6	16	CYT	H1'	4.47
16	CYT	H6	16	CYT	H2'1	5.5
17	THY	H1'	17	THY	H4'	4.17
17	THY	H2'1	17	THY	H1'	3.64
17	THY	H2'2	17	THY	H1'	2.96
17	THY	H2'2	17	THY	H6	3.58
17	THY	H3	17	THY	H1'	6
17	THY	H3'	17	THY	H4'	4.46
17	THY	H6	17	THY	H1'	4.56
17	THY	H6	17	THY	H2'1	5.5
17	THY	Q5'	17	THY	H4'	3.45
18	THY	H2'1	18	THY	H1'	4.19
18	THY	H4'	18	THY	H3'	5.09
18	THY	H6	18	THY	H1'	4.57
18	THY	H6	18	THY	H2'1	5.5
18	THY	H6	18	THY	H2'2	5.5
19	CYT	H1'	19	CYT	H4'	5.5
19	CYT	H2'1	19	CYT	H1'	3.51
19	CYT	H3'	19	CYT	Q5'	4.68
19	CYT	H3'	19	CYT	Q5'	5.5
19	CYT	H6	19	CYT	H1'	4.31
19	CYT	H6	19	CYT	H2'1	5.5
19	CYT	H6	19	CYT	H2'2	5.5
20	THY	H1'	20	THY	H4'	3.67
20	THY	H2'1	20	THY	H1'	3.27
20	THY	H3	20	THY	H1'	6
20	THY	H3'	20	THY	H4'	5.83
20	THY	H6	20	THY	H1'	5.5
20	THY	H6	20	THY	H2'1	5.5
21	CYT	H2'1	21	CYT	H1'	4.1
21	CYT	H2'2	21	CYT	H1'	3.03
21	CYT	H4'	21	CYT	H1'	3.54
21	CYT	H6	21	CYT	H1'	4.33
21	CYT	H6	21	CYT	H2'1	5.5
21	CYT	H6	21	CYT	H2'2	5.5
22	THY	H1'	22	THY	H4'	4.21
22	THY	H2'1	22	THY	H1'	3.48
22	THY	H2'2	22	THY	H1'	3.04
22	THY	H3	22	THY	H1'	5.87
22	THY	H3'	22	THY	H4'	5.05
22	THY	H6	22	THY	H1'	4.61
22	THY	H6	22	THY	H2'1	5.5
22	THY	H6	22	THY	H2'2	5.5

22	THY	Q5'	22	THY	H4'	3.45
23	THY	H1'	23	THY	H4'	5.5
23	THY	H2'1	23	THY	H1'	3.54
23	THY	H2'2	23	THY	H1'	2.94
23	THY	H2'2	23	THY	H3'	4.16
23	THY	H3	23	THY	H1'	5.56
23	THY	H3'	23	THY	H4'	4.38
23	THY	H3'	23	THY	H5'1	5.5
23	THY	H3'	23	THY	H5'2	5.5
23	THY	H3'	23	THY	Q5'	4.87
23	THY	H4'	23	THY	Q5'	3.4
23	THY	H6	23	THY	H1'	4.5
23	THY	H6	23	THY	H2'1	5.5
24	THY	H2'1	24	THY	H1'	4.16
24	THY	H2'2	24	THY	H1'	2.87
24	THY	H4'	24	THY	H1'	3.46
24	THY	H4'	24	THY	H2'1	5.5
24	THY	H4'	24	THY	H2'2	5.5
24	THY	H4'	24	THY	H3'	4.11
24	THY	H6	24	THY	H1'	4.24
24	THY	H6	24	THY	H2'1	5.5
24	THY	H6	24	THY	H2'2	5.5
24	THY	Q5'	24	THY	H3'	4.95
25	GUA	H1'	25	GUA	H3'	4.36
25	GUA	H1'	25	GUA	H4'	5.5
25	GUA	H1'	25	GUA	H8	4.12
25	GUA	H3'	25	GUA	H5'1	5.5
25	GUA	H3'	25	GUA	H5'2	5.5
25	GUA	H3'	25	GUA	H8	4.76
25	GUA	H4'	25	GUA	H3'	3.33
25	GUA	H8	25	GUA	H2'1	5.5
25	GUA	H8	25	GUA	H2'2	5.5
26	GUA	H1'	26	GUA	H2'2	3.14
26	GUA	H1'	26	GUA	H3'	5.6
26	GUA	H1'	26	GUA	H4'	5.5
26	GUA	H1'	26	GUA	H8	4.28
26	GUA	H2'1	26	GUA	H8	3.28
26	GUA	H4'	26	GUA	H2'1	5.5
26	GUA	H4'	26	GUA	H2'2	5.5

2) Intramolecular Ligand to Ligand Constraints

residue number	residue name	atom name	residue number	residue name	atom name	upper bound
27	NIM	H	27	NIM	H4	6
27	NIM	H	28	PYL	H3	3.55
28	PYL	H	29	BAL	H11	3.95
28	PYL	H	29	BAL	H12	3.95
28	PYL	H	29	BAL	2Q	6
29	BAL	H	29	BAL	H11	5.3
29	BAL	H	29	BAL	H12	5.3
29	BAL	H	29	BAL	2Q	6
29	BAL	H11	29	BAL	H21	5.5
29	BAL	H11	29	BAL	H22	5.5
29	BAL	H12	29	BAL	H21	5.5
29	BAL	H12	29	BAL	H22	5.5
30	IMI	H	31	BAL	H11	4.59
30	IMI	H	31	BAL	H12	4.59
30	IMI	H	31	BAL	H21	4.7
30	IMI	H	31	BAL	H22	4.7
31	BAL	1Q	31	BAL	2Q	5.5
31	BAL	1Q	31	BAL	2Q	5.5
31	BAL	2Q	31	BAL	1Q	4.29
31	BAL	H	31	BAL	1Q	4.72
31	BAL	H	31	BAL	H21	6
31	BAL	H	31	BAL	H22	6
32	IMI	H	33	PYL	H3	3.53
32	IMI	H	33	PYL	H5	4.53
33	PYL	H	34	BAL	H11	3.55
33	PYL	H	34	BAL	H12	3.55
33	PYL	H	34	BAL	2Q	4.79
33	PYL	H	33	PYL	H3	3.12
33	PYL	H5	33	PYL	NM	5.5
34	BAL	2Q	34	BAL	1Q	4.17
34	BAL	H	34	BAL	H21	4.31
34	BAL	H	34	BAL	H22	4.31
34	BAL	H	35	DMP	2Q	6
34	BAL	H	35	DMP	3Q	6
34	BAL	H	35	DMP	H11	6
34	BAL	H	35	DMP	H12	6
34	BAL	H	33	PYL	H	4.96
35	DMP	3Q	35	DMP	DM	5.5

3) Interresidue DNA to DNA Constraints

residue number	residue name	atom name	residue number	residue name	atom name	upper bound
2	CYT	H5	1	CYT	H6	5.5
2	CYT	H6	1	CYT	H2'2	5.5
2	CYT	H6	1	CYT	H1'	4.13
2	CYT	H6	1	CYT	H2'1	4.87
3	ADE	H8	2	CYT	H2'1	5.5
3	ADE	H8	2	CYT	H2'2	5.5
3	ADE	H8	2	CYT	H1'	5.5
3	ADE	H2	4	ADE	H2	5.5
3	ADE	H2	25	GUA	H1'	6
4	ADE	H8	3	ADE	H1'	4.25
4	ADE	H8	3	ADE	H2'1	5.5
4	ADE	H8	3	ADE	H2'2	5.5
4	ADE	H2	5	ADE	H2	3.62
4	ADE	H2	24	THY	H1'	5.5
5	ADE	H8	4	ADE	H1'	4.13
5	ADE	H8	4	ADE	H2'1	5.5
5	ADE	H8	4	ADE	H2'2	5.5
5	ADE	H2	23	THY	H1'	4.24
5	ADE	H2	23	THY	H3	4.88
6	GUA	H1	5	ADE	H2	4.66
6	GUA	H8	5	ADE	H2'1	4.05
6	GUA	H8	5	ADE	H2'2	6
6	GUA	H8	5	ADE	H1'	5.5
6	GUA	H1	7	ADE	H2	4.68
6	GUA	H1	21	CYT	H41	5.5
6	GUA	H1	21	CYT	H42	6
7	ADE	H8	6	GUA	H2'2	6
7	ADE	H8	6	GUA	H2'1	3.44
7	ADE	H8	6	GUA	H1'	3.64
7	ADE	Q5'	6	GUA	H1'	3.15
7	ADE	H1'	8	GUA	Q5'	5.5
7	ADE	H2	8	GUA	H1'	4.88
7	ADE	H2	21	CYT	H1'	4.88
8	GUA	H8	7	ADE	H2'1	4.44
8	GUA	H8	7	ADE	H2'2	5.5
8	GUA	H1	19	CYT	H42	3.84
8	GUA	H1	19	CYT	H5	6
8	GUA	H1	19	CYT	H41	3.18
8	GUA	H1	9	ADE	H2	5.5
9	ADE	H8	8	GUA	H2'1	5.5
9	ADE	H8	8	GUA	H2'2	4.9
9	ADE	H8	8	GUA	H1'	5.53
9	ADE	H2	10	ADE	H2	3.5
9	ADE	H2	10	ADE	H1'	5.54
9	ADE	H2	19	CYT	H1'	4.24
10	ADE	H8	9	ADE	H1'	5.5
10	ADE	H8	9	ADE	H2'1	5.5
10	ADE	Q5'	9	ADE	H1'	4.92
10	ADE	H1'	11	GUA	Q5'	3.6
10	ADE	H2	11	GUA	H1'	4.53
11	GUA	H8	10	ADE	H2'2	5.5

11	GUA	H8	10	ADE	H2'1	5.5
11	GUA	H8	10	ADE	H1'	3.53
11	GUA	Q5'	10	ADE	H1'	3.09
11	GUA	H1	12	CYT	H42	3.43
11	GUA	H1	16	CYT	H41	4.67
11	GUA	H1	16	CYT	H42	3.24
12	CYT	H6	11	GUA	H2'2	3.32
12	CYT	H6	11	GUA	H2'1	5.66
12	CYT	H6	11	GUA	H1'	6
12	CYT	H41	15	GUA	H1	5.53
12	CYT	H5	15	GUA	H1	5.4
13	GUA	H8	12	CYT	H2'1	3.25
13	GUA	H8	12	CYT	H2'2	3.49
15	GUA	H1	11	GUA	H1	4.68
15	GUA	H1	12	CYT	H42	5.78
15	GUA	H1	14	CYT	H41	6
15	GUA	H8	14	CYT	H2'2	4.12
15	GUA	H8	14	CYT	H2'1	4.15
15	GUA	H8	14	CYT	H1'	3.26
16	CYT	H5	15	GUA	H8	5.5
16	CYT	H5	15	GUA	H2'2	3.59
16	CYT	H6	15	GUA	H1'	4.25
16	CYT	H1'	17	THY	Q5'	5.5
16	CYT	H5	17	THY	M7	4.8
16	CYT	H6	17	THY	M7	4.9
17	THY	H3	10	ADE	H2	3.58
17	THY	H3	11	GUA	H1	4.66
17	THY	H6	16	CYT	H2'2	3.25
17	THY	H6	16	CYT	H2'1	3.42
17	THY	H6	16	CYT	H1'	4.44
17	THY	M7	16	CYT	H2'1	3.6
17	THY	M7	16	CYT	H1'	4.95
17	THY	H6	18	THY	M7	4.24
18	THY	H3	8	GUA	H1	5.17
18	THY	H3	9	ADE	H2	4.61
18	THY	H1'	10	ADE	H2	6.22
18	THY	H3	10	ADE	H2	4.4
18	THY	H6	17	THY	H1'	4.58
18	THY	H6	17	THY	H2'1	5.5
18	THY	H6	17	THY	H2'2	5.5
18	THY	M7	17	THY	H2'1	5.5
18	THY	M7	17	THY	H1'	5.74
18	THY	Q5'	17	THY	H1'	5.88
18	THY	H3	19	CYT	H42	5.28
19	CYT	H5	18	THY	H2'2	3.62
19	CYT	H41	18	THY	M7	4.45
19	CYT	H42	18	THY	M7	4.4
19	CYT	H5	18	THY	H6	4.57
19	CYT	H5	18	THY	M7	4.95
19	CYT	H5	18	THY	H2'1	5.5
19	CYT	H6	18	THY	H1'	4.5
19	CYT	H6	18	THY	H2'1	5.5
19	CYT	H6	18	THY	H2'2	5.5
19	CYT	Q5'	18	THY	H1'	4.96
19	CYT	H2'2	20	THY	M7	3.68
19	CYT	H6	20	THY	M7	5.5
20	THY	H3	6	GUA	H1	5.16

20	THY	H3	7	ADE	H2	3.5
20	THY	H3	8	GUA	H1	4.79
20	THY	H6	19	CYT	H1'	4.2
20	THY	H6	19	CYT	H2'1	4.25
20	THY	H6	19	CYT	H2'2	5.5
20	THY	M7	19	CYT	H2'1	3.6
20	THY	M7	19	CYT	H5	4.7
20	THY	M7	19	CYT	H1'	4.9
20	THY	Q5'	19	CYT	H1'	4.89
21	CYT	H42	20	THY	H3	5.47
21	CYT	H5	20	THY	H6	5.5
21	CYT	H41	20	THY	M7	4.5
21	CYT	H5	20	THY	M7	5.5
21	CYT	H6	20	THY	H2'1	5.5
21	CYT	H6	20	THY	H2'2	5.5
21	CYT	H5	22	THY	M7	5.5
21	CYT	H6	22	THY	M7	5.5
22	THY	H3	5	ADE	H2	4.25
22	THY	H3	6	GUA	H1	4.94
22	THY	H6	21	CYT	H2'1	3.62
22	THY	H6	21	CYT	H1'	4.4
22	THY	H6	21	CYT	H2'2	5.5
22	THY	M7	21	CYT	H2'1	4.26
22	THY	M7	21	CYT	H1'	5.62
22	THY	H3	23	THY	H3	4.16
23	THY	H3	3	ADE	H2	6
23	THY	H3	4	ADE	H2	3.44
23	THY	H6	22	THY	H2'2	3.09
23	THY	H6	22	THY	H2'1	3.44
23	THY	H6	22	THY	H1'	4.37
23	THY	M7	22	THY	H2'1	3.64
23	THY	M7	22	THY	H6	4.38
23	THY	Q5'	22	THY	H1'	5.05
24	THY	M7	23	THY	H2'1	5.5
24	THY	M7	23	THY	H2'2	5.5
24	THY	H3	3	ADE	H2	3.5
24	THY	H3	4	ADE	H2	4.73
24	THY	H6	23	THY	H2'2	3.18
24	THY	H6	23	THY	H1'	4.8
24	THY	H6	23	THY	H2'1	5.5
24	THY	M7	23	THY	H6	4.25
24	THY	M7	23	THY	H1'	5.56
25	GUA	H1	24	THY	H3	4.79
25	GUA	H1	1	CYT	H41	6
25	GUA	H1	2	CYT	H42	3.44
25	GUA	H1	2	CYT	H41	6
25	GUA	H1	3	ADE	H2	4.95
25	GUA	H8	24	THY	H2'1	4.12
25	GUA	H8	24	THY	H2'2	4.23
25	GUA	H8	24	THY	H1'	4.93
26	GUA	H8	25	GUA	H2'1	5.5
26	GUA	H8	25	GUA	H2'2	5.5
26	GUA	H8	25	GUA	H1'	4.18

4) Intermolecular Ligand to DNA Constraints

residue number	residue name	atom name	residue number	residue name	atom name	upper bound
27	NIM	H	10	ADE	H2	4.77
27	NIM	H	11	GUA	H22	3.78
27	NIM	H	11	GUA	H1'	4.54
27	NIM	H	12	CYT	H4'	5.39
27	NIM	H	12	CYT	Q5'	6
27	NIM	H	17	THY	H1'	4.74
27	NIM	H	18	THY	H4'	4.85
27	NIM	H	18	THY	H1'	5.64
27	NIM	H	18	THY	Q5'	6
27	NIM	H4	16	CYT	H1'	5.5
27	NIM	H4	17	THY	H4'	4.15
27	NIM	H4	17	THY	H1'	4.19
27	NIM	H5	13	GUA	Q5'	5.53
27	NIM	H5	13	GUA	Q5'	5.5
27	NIM	H5	17	THY	H4'	4.15
27	NIM	NM	12	CYT	H4'	5.5
27	NIM	NM	18	THY	H4'	5.5
28	PYL	H	9	ADE	H2	4.13
28	PYL	H	10	ADE	H1'	4.88
28	PYL	H	10	ADE	H2	6
28	PYL	H	11	GUA	H4'	4.7
28	PYL	H	18	THY	H1'	4.36
28	PYL	H	19	CYT	H1'	6
28	PYL	H3	9	ADE	H2	4.68
28	PYL	H3	10	ADE	H2	2.07
28	PYL	H3	11	GUA	H1'	3.26
28	PYL	H3	11	GUA	H4'	4.69
28	PYL	H3	17	THY	H3	5.07
28	PYL	H3	18	THY	H1'	3.08
28	PYL	H5	12	CYT	H4'	3.24
28	PYL	H5	12	CYT	Q5'	3.25
28	PYL	H5	12	CYT	H4'	3.24
28	PYL	H5	12	CYT	Q5'	3.25
28	PYL	H5	18	THY	H4'	3.05
29	BAL	1Q	9	ADE	H2	4.44
29	BAL	1Q	9	ADE	H2	5.5
29	BAL	1Q	10	ADE	H1'	4.38
29	BAL	1Q	10	ADE	H2	4.83
29	BAL	1Q	19	CYT	H1'	4.83
29	BAL	2Q	9	ADE	H2	5.5
29	BAL	2Q	10	ADE	H1'	5.5
29	BAL	2Q	19	CYT	H1'	5.5
29	BAL	2Q	19	CYT	H4'	5.5
29	BAL	2Q	19	CYT	H1'	5.5
29	BAL	2Q	19	CYT	H4'	5.5
29	BAL	H	8	GUA	H22	4.48
29	BAL	H	9	ADE	H1'	3.93
29	BAL	H	9	ADE	H2	4.94
29	BAL	H	10	ADE	Q5'	6
29	BAL	H	19	CYT	H1'	4.67
29	BAL	H	20	THY	H4'	5.22
30	IMI	H	7	ADE	H2	6

30	IMI	H	8	GUA	H1'	6
30	IMI	H	8	GUA	H22	6
30	IMI	H	20	THY	H1'	6
30	IMI	H	21	CYT	H1'	4.34
30	IMI	H5	10	ADE	Q5'	4.34
30	IMI	H5	10	ADE	H4'	4.74
30	IMI	H5	20	THY	H4'	3.66
31	BAL	1Q	7	ADE	H2	4.54
31	BAL	1Q	8	GUA	H1'	4.65
31	BAL	1Q	21	CYT	H1'	3.6
31	BAL	2Q	7	ADE	H2	3.6
31	BAL	2Q	7	ADE	H1'	5.5
31	BAL	2Q	8	GUA	H4'	4.1
31	BAL	2Q	21	CYT	H1'	4.48
31	BAL	H	6	GUA	H22	4.49
31	BAL	H	7	ADE	H1'	3.41
31	BAL	H	7	ADE	H2	3.56
31	BAL	H	8	GUA	Q5'	6
32	IMI	H	5	ADE	H2	6
32	IMI	H	6	GUA	H1'	4.36
32	IMI	H	6	GUA	H22	6
32	IMI	H	7	ADE	H4'	4.18
32	IMI	H	22	THY	H1'	4.5
32	IMI	H	23	THY	H1'	4.44
32	IMI	H	23	THY	Q5'	6
32	IMI	H5	8	GUA	Q5'	4.38
32	IMI	H5	22	THY	H4'	4.19
33	PYL	H	4	ADE	H2	4.38
33	PYL	H	5	ADE	H2	3.99
33	PYL	H	6	GUA	H4'	4.96
33	PYL	H	23	THY	H1'	3.97
33	PYL	H	24	THY	H4'	6
33	PYL	H3	5	ADE	H2	2.08
33	PYL	H3	6	GUA	H1'	3.14
33	PYL	H3	6	GUA	H4'	5.21
33	PYL	H3	22	THY	H3	4.73
33	PYL	H3	23	THY	H3	5.53
33	PYL	H5	6	GUA	H4'	3.44
33	PYL	H5	7	ADE	H4'	5.5
33	PYL	H5	23	THY	H4'	5.5
34	BAL	1Q	4	ADE	H2	4.47
34	BAL	1Q	5	ADE	H1'	4.63
34	BAL	1Q	24	THY	H1'	4.56
34	BAL	2Q	4	ADE	H2	4.24
34	BAL	2Q	5	ADE	H1'	4.14
34	BAL	2Q	5	ADE	H2	4.82
34	BAL	2Q	24	THY	H1'	4.65
34	BAL	H	3	ADE	H2	5.79
34	BAL	H	4	ADE	H2	4.77
34	BAL	H	4	ADE	H1'	5.12
35	DMP	1Q	4	ADE	H1'	5.5
35	DMP	2Q	3	ADE	H2	5.5
35	DMP	2Q	4	ADE	H1'	5.5
35	DMP	2Q	4	ADE	H2	5.5
35	DMP	3Q	3	ADE	H2	5.5
35	DMP	3Q	4	ADE	H1'	5.5
35	DMP	3Q	4	ADE	H2	5.5

```
35      DMP      DM       3       ADE       H2      5.5
35      DMP      DM       3       ADE      H4'      5.5
35      DMP      DM       4       ADE       H2      5.5
35      DMP      DM      24       THY      H1'      5.5
35      DMP      DM      25       GUA      H4'      5.5
```

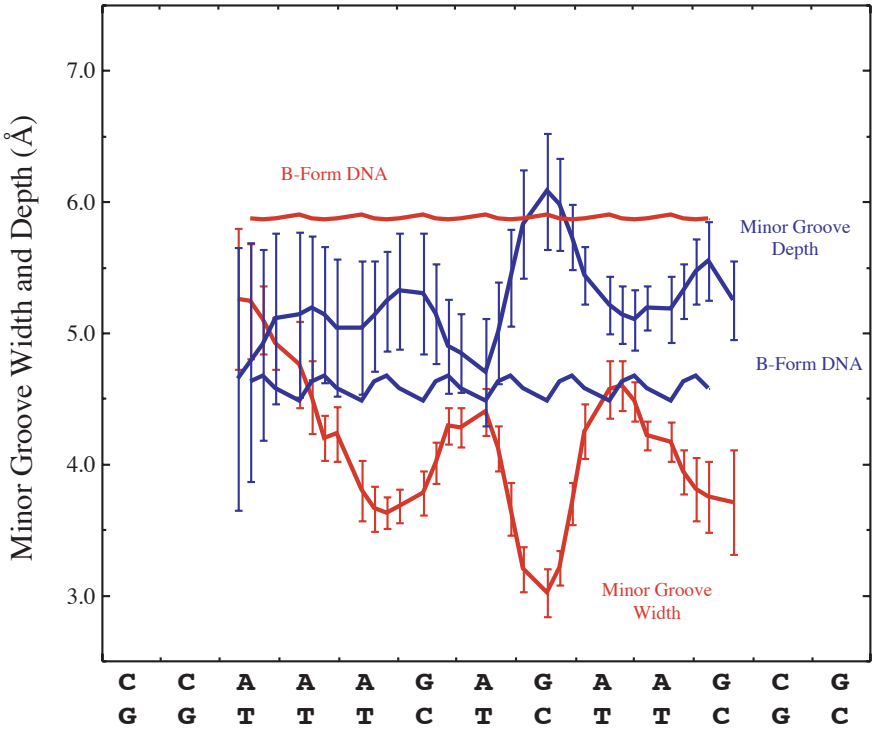
5) Watson-Crick Hydrogen Bonding Constraints

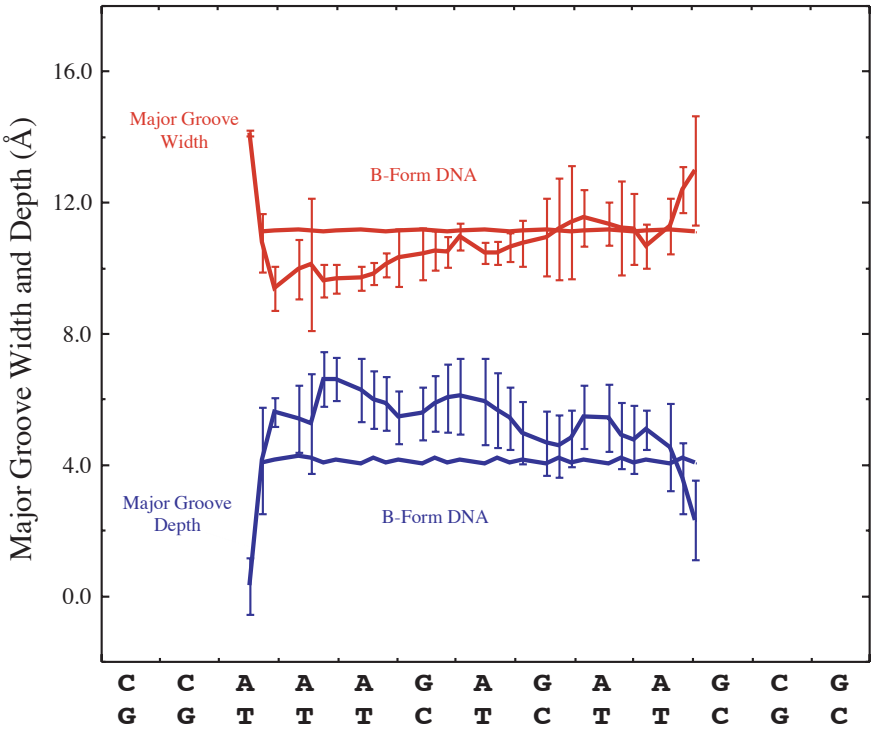
residue number	residue name	atom name	residue number	residue name	atom name	upper bound	lower bound
1	CYT	H42	26	GUA	O6	1.76	2.16
1	CYT	N3	26	GUA	H1	1.8	2.2
1	CYT	N3	26	GUA	N1	2.85	3.05
1	CYT	N4	26	GUA	O6	2.81	3.01
2	CYT	H42	25	GUA	O6	1.76	2.16
2	CYT	N3	25	GUA	H1	1.8	2.2
2	CYT	N3	25	GUA	N1	2.85	3.05
2	CYT	N4	25	GUA	O6	2.81	3.01
3	ADE	N1	24	THY	H3	1.67	2.07
3	ADE	N1	24	THY	N3	2.72	2.92
4	ADE	N1	23	THY	H3	1.67	2.07
4	ADE	N1	23	THY	N3	2.72	2.92
5	ADE	N1	22	THY	H3	1.67	2.07
5	ADE	N1	22	THY	N3	2.72	2.92
6	GUA	H1	21	CYT	N3	1.8	2.2
6	GUA	N1	21	CYT	N3	2.85	3.05
6	GUA	O6	21	CYT	H42	1.76	2.16
6	GUA	O6	21	CYT	N4	2.81	3.01
7	ADE	N1	20	THY	H3	1.67	2.07
7	ADE	N1	20	THY	N3	2.72	2.92
8	GUA	H1	19	CYT	N3	1.8	2.2
8	GUA	N1	19	CYT	N3	2.85	3.05
8	GUA	O6	19	CYT	H42	1.76	2.16
8	GUA	O6	19	CYT	N4	2.81	3.01
9	ADE	N1	18	THY	H3	1.67	2.07
9	ADE	N1	18	THY	N3	2.72	2.92
10	ADE	N1	17	THY	H3	1.67	2.07
10	ADE	N1	17	THY	N3	2.72	2.92
11	GUA	H1	16	CYT	N3	1.8	2.2
11	GUA	N1	16	CYT	N3	2.85	3.05
11	GUA	O6	16	CYT	H42	1.76	2.16
11	GUA	O6	16	CYT	N4	2.81	3.01
12	CYT	H42	15	GUA	O6	1.76	2.16
12	CYT	N3	15	GUA	H1	1.8	2.2
12	CYT	N3	15	GUA	N1	2.85	3.05
12	CYT	N4	15	GUA	O6	2.81	3.01
13	GUA	H1	14	CYT	N3	1.8	2.2
13	GUA	N1	14	CYT	N3	2.85	3.05
13	GUA	O6	14	CYT	H42	1.76	2.16
13	GUA	O6	14	CYT	N4	2.81	3.01

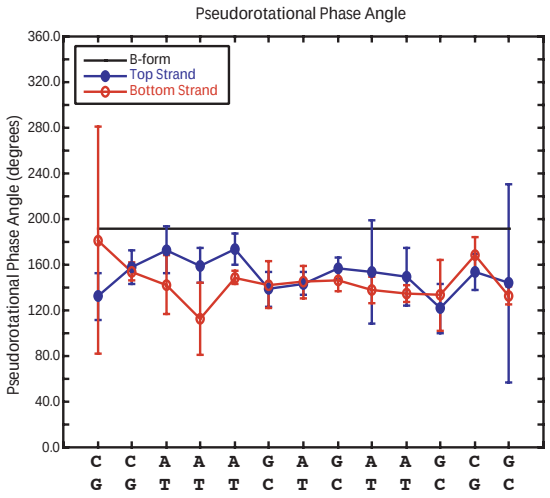
Appendix C

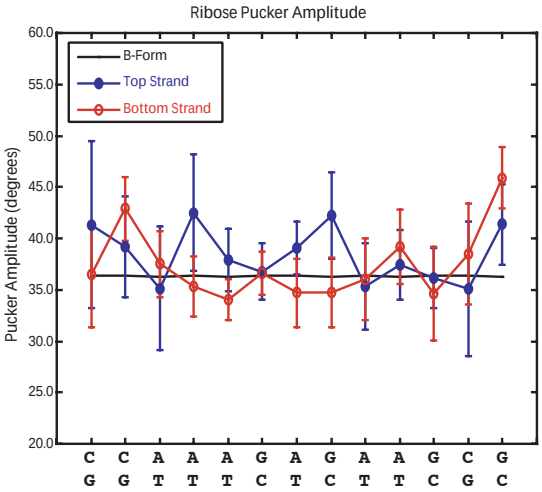
DNA Helical Parameters

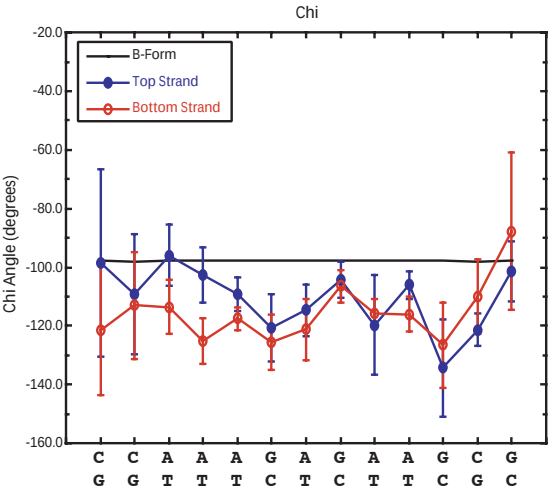
DNA helical parameters for the 1:1 polyamide-DNA complex determined by NMR methods are provided here. The parameters are plotted along the ordinate for each DNA base step along the abscissa. Average values over the final ensemble of 12 structures are connected by solid lines, and the y-axis error bars indicates one standard deviation from the average. Horizontal lines without error bars indicate the average values for standard B-form DNA.

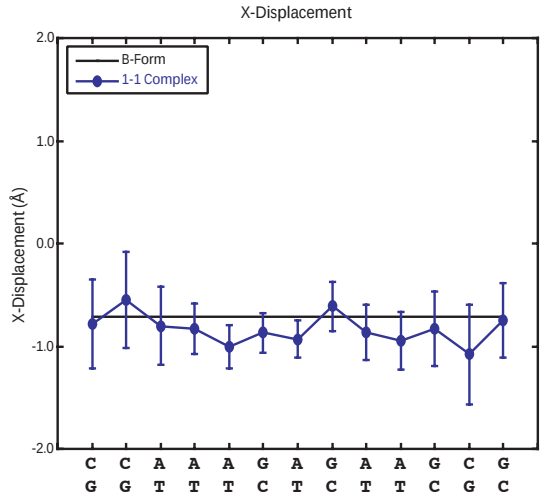

Minor Groove Width and Depth (Å)
7.0
6.0
5.0
4.0
3.0
B-Form DNA
Minor Groove Depth
B-Form DNA
Minor Groove Width
C C A A A G A G A A G C G
G G T T T C T C T T C G C

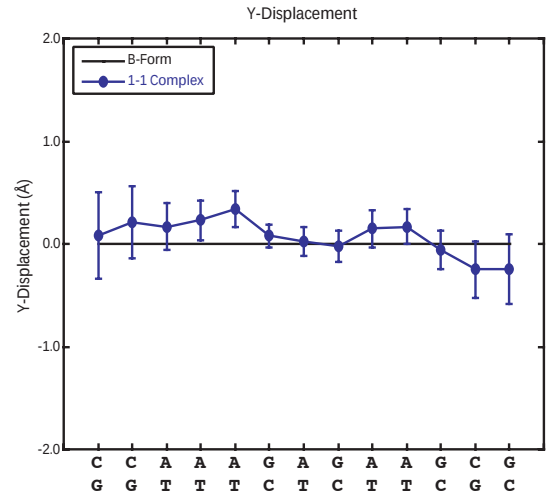

Major Groove Width and Depth (Å)
16.0
12.0
8.0
4.0
0.0
Major Groove Width
B-Form DNA
Major Groove Depth
B-Form DNA
C C A A A G A G A A G C G
G G T T T C T C T T C G C

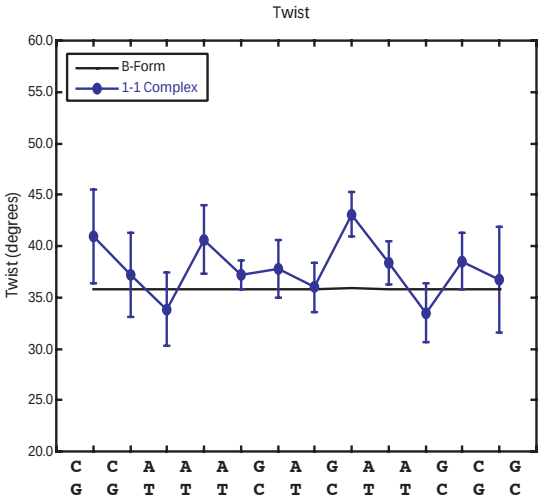

Pseudorotational Phase Angle
Pseudorotational Phase Angle (degrees)
B-form
Top Strand
Bottom Strand
C C A A A G A G A A G C G
G G T T T C T C T T C G C

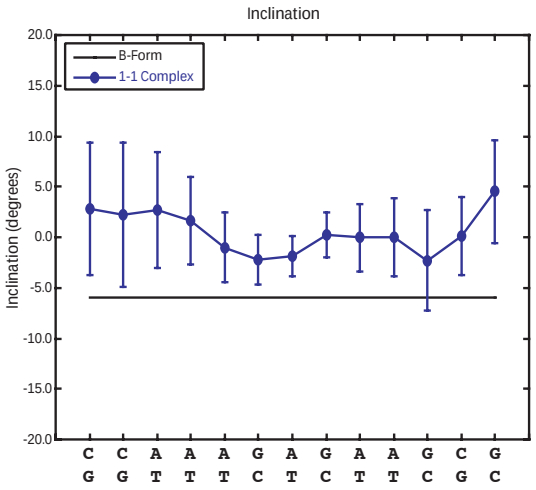

Ribose Pucker Amplitude
Pucker Amplitude (degrees)
B-Form
Top Strand
Bottom Strand
C C A A A G A G A A G C G
G G T T T C T C T T C G C

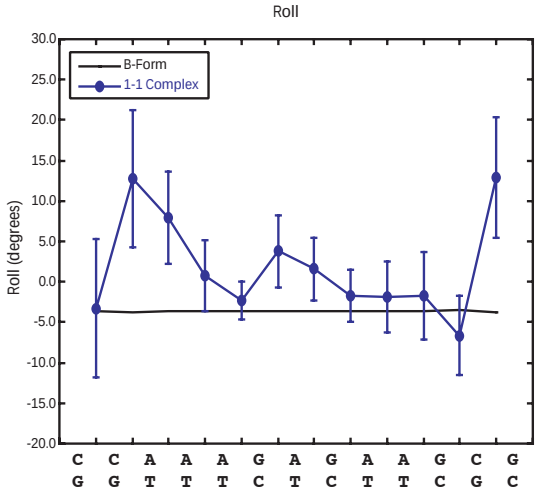

Chi
Chi Angle (degrees)
B-Form
Top Strand
Bottom Strand
C C A A A G A G A A G C G
G G T T T C T C T T C G C

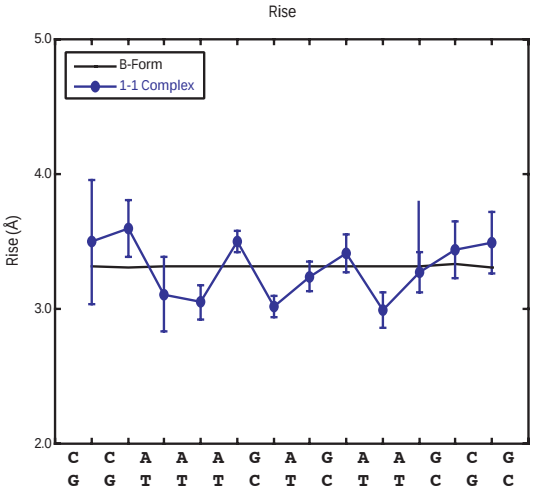

X-Displacement
X-Displacement (Å)
B-Form
1-1 Complex
C C A A A G A G A A G C G
G G T T T C T C T T C G C

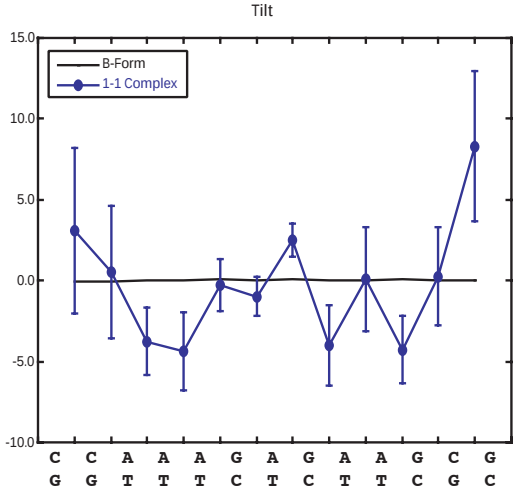

Y-Displacement
Y-Displacement (Å)
B-Form
1-1 Complex
C C A A A G A G A A G C G
G G T T T C T C T T C G C

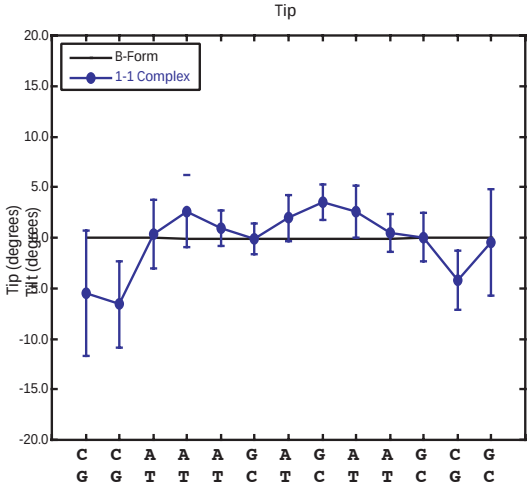

Twist
Twist (degrees)
B-Form
1-1 Complex
C C A A A G A G A A G C G
G G T T T C T C T T C G C

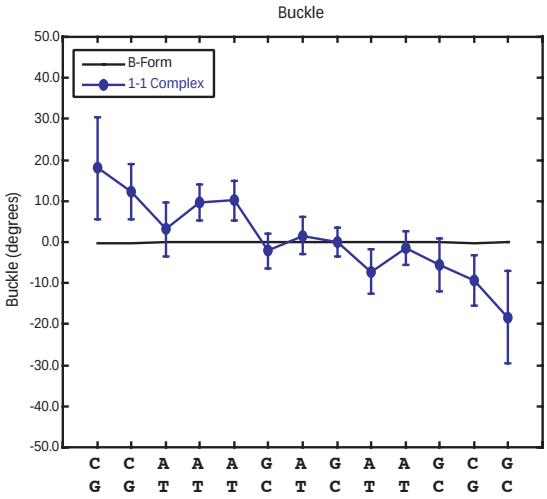

Inclination
B-Form
1-1 Complex
Inclination (degrees)
20.0
15.0
10.0
5.0
0.0
-5.0
-10.0
-15.0
-20.0
C C A A A G A G A A G C G
G G T T T C T C T T C G C

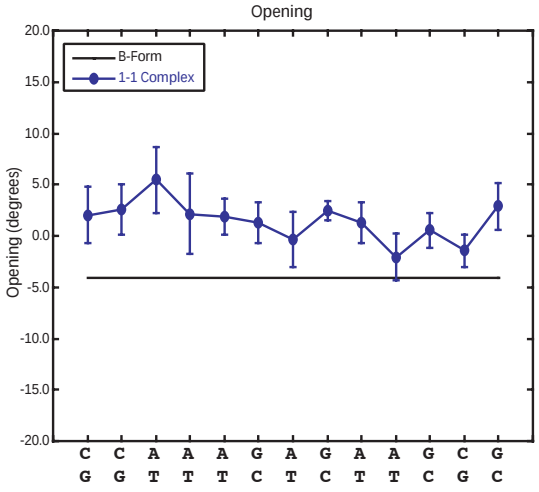

Roll
B-Form
1-1 Complex
Roll (degrees)
30.0
25.0
20.0
15.0
10.0
5.0
0.0
-5.0
-10.0
-15.0
-20.0
C C A A A G A G A A G C G
G G T T T C T C T T C G C

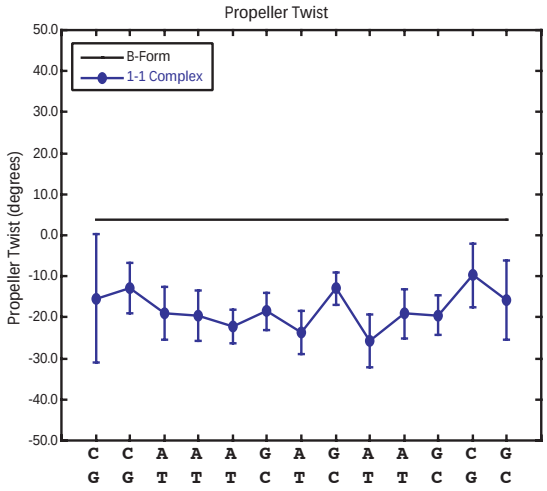

Rise
B-Form
1-1 Complex
Rise (Å)
5.0
4.0
3.0
2.0
C C A A A G A G A A G C G
G G T T T C T C T T C G C

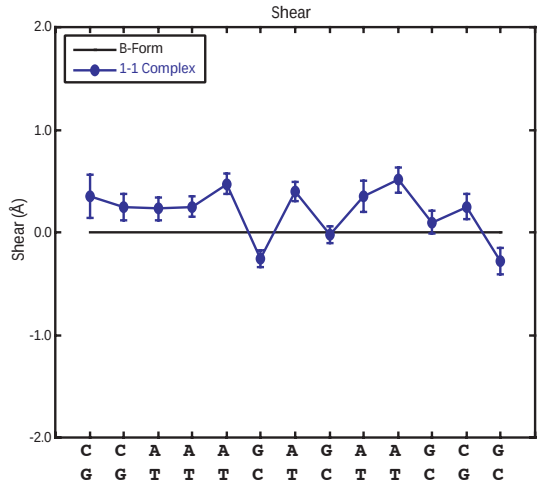

Tilt
B-Form
1-1 Complex
15.0
10.0
5.0
0.0
-5.0
-10.0
C C A A A G A G A A G C G
G G T T T C T C T T C G C

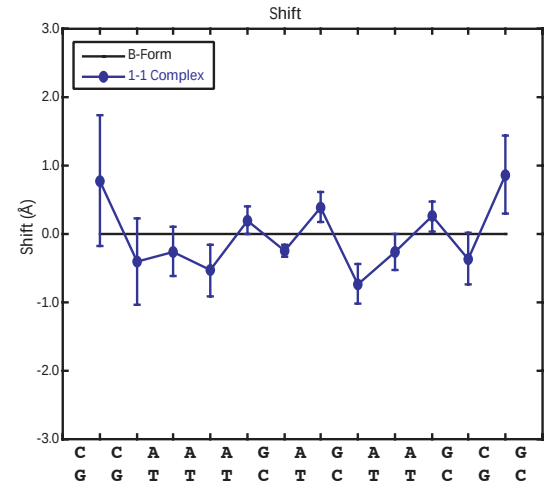

Tip
B-Form
1-1 Complex
Tip (degrees)
Tilt (degrees)
20.0
15.0
10.0
5.0
0.0
-5.0
-10.0
-15.0
-20.0
C C A A A G A G A A G C G
G G T T T C T C T T C G C

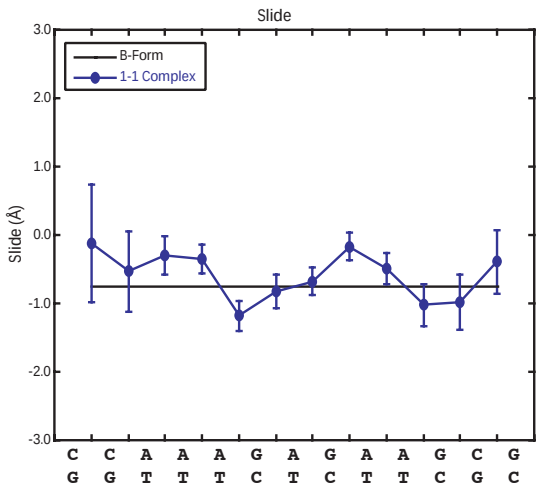

Buckle
Buckle (degrees)
B-Form
1-1 Complex
50.0
40.0
30.0
20.0
10.0
0.0
-10.0
-20.0
-30.0
-40.0
-50.0
C C A A A G A G A A G C G
G G T T T C T C T T C G C

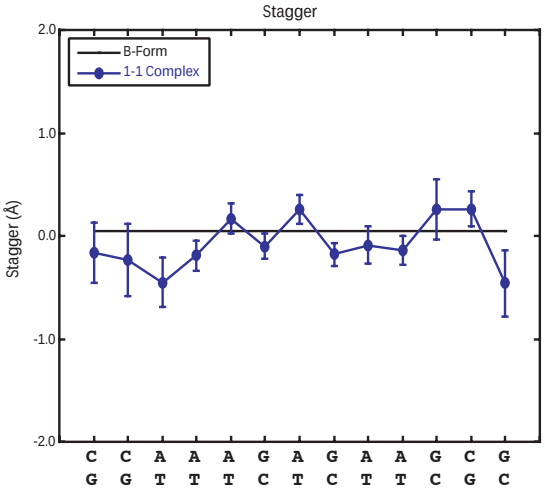

Opening
Opening (degrees)
B-Form
1-1 Complex
20.0
15.0
10.0
5.0
0.0
-5.0
-10.0
-15.0
-20.0
C C A A A G A G A A G C G
G G T T T C T C T T C G C

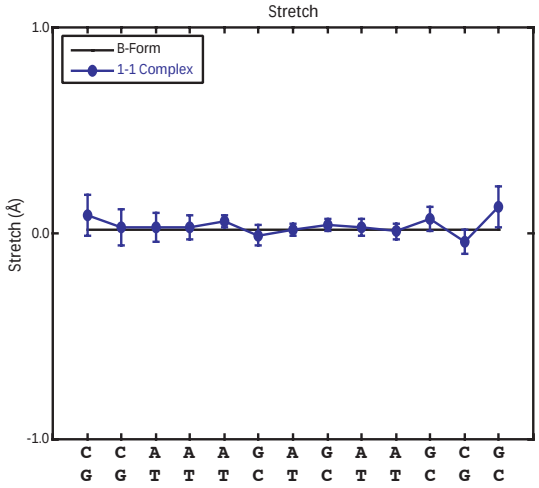

Propeller Twist
Propeller Twist (degrees)
B-Form
1-1 Complex
50.0
40.0
30.0
20.0
10.0
0.0
-10.0
-20.0
-30.0
-40.0
-50.0
C C A A A G A G A A G C G
G G T T T C T C T T C G C


Shear
Shear (Å)
B-Form
1-1 Complex
2.0
1.0
0.0
-1.0
-2.0
C C A A A G A G A A G C G
G G T T T C T C T T C G C


Shift
Shift (Å)
B-Form
1-1 Complex
3.0
2.0
1.0
0.0
-1.0
-2.0
-3.0
C C A A A G A G A A G C G
G G T T T C T C T T C G C


Slide
Slide (Å)
B-Form
1-1 Complex
3.0
2.0
1.0
0.0
-1.0
-2.0
-3.0
C C A A A G A G A A G C G
G G T T T C T C T T C G C


Stagger
B-Form
1-1 Complex
Stagger (Å)
2.0
1.0
0.0
-1.0
-2.0
C C A A A G A G A A G C G
G G T T T C T C T T C G C


Stretch
B-Form
1-1 Complex
Stretch (Å)
1.0
0.0
-1.0
C C A A A G A G A A G C G
G G T T T C T C T T C G C