Site-Specific Stabilization of DNA by a Tethered Major Groove Amine, 7-Aminomethyl-7-Deaza-2'-Deoxyguanosine

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Table S1. ^{1}H chemical shifts of the oligodeoxynucleotide non-exchangeable protons for the OL-1b duplex.

Nucleotide	H6/ H8	H2/ H5/ CH_3	H1'	H2'	H2''	H3'	H4'	H5'	H5''	H101	H102
G^{1}	7.80		5.49	2.39	2.59	4.76	4.18	4.09	3.61		
A^{2}	8.13	7.71	5.89	2.70	2.81	4.99	4.12	4.00	4.35		
G^{3}	7.64		5.45	2.52	2.64	4.94		4.14	4.10		
A^{4}	8.02	7.73	6.14	2.67	2.88	4.96	4.14	4.4	4.32		
X^{5}	7.72		5.49	2.23	2.21	4.82		4.14		3.64	3.47
C^{6}	7.25	5.15	5.78	1.86	2.36	4.77	4.15				
G^{7}	7.79		5.86	2.59	2.79	4.91	4.30	3.97	4.07		
C^{8}	7.35	5.34	5.84	1.99	2.46	4.65	4.14	4.26	4.06		
T^{9}	7.44	1.51	6.06	2.12	2.25	4.85	4.06	4.21			
C^{10}	7.59	5.64	5.97	2.49	2.49	4.74		4.12			
T^{11}	7.45	1.70	6.07	2.15	2.24	4.84		4.13			
C^{12}	7.65	5.84	6.24	2.70	2.48	4.53	4.13	3.99	4.17		

Table S2. Summary of NOE-based distance restraints used in the rMD calculations for the OL-1b duplex.

Nucleotide I		Atom I	Nucleotide II		Atom II	Lower bound	Upper bound
1	DG5	H2''	1	DG5	H1'	1.89	2.82
1	DG5	H2''	1	DG5	H1'	2.07	3.23
1	DG5	H3'	1	DG5	H1'	2.06	3.04
1	DG5	H3'	1	DG5	H2''	2.36	3.94
1	DG5	H3'	1	DG5	H2'	1.74	2.58
1	DG5	H8	1	DG5	H1'	2.56	4.32
1	DG5	H8	1	DG5	H2'	1.70	3.10
1	DG5	H8	1	DG5	H3'	3.33	5.62
2	DA	H2''	2	DA	H1'	2.10	2.40
2	DA	H2'	2	DA	H1'	2.23	2.93
2	DA	H3'	2	DA	H1'	2.76	3.98
2	DA	H3'	2	DA	H2''	2.23	2.90
2	DA	H3'	2	DA	H2'	2.66	3.73
2	DA	H8	1	DG5	H1'	3.13	3.81
2	DA	H8	1	DG5	H2''	2.47	3.14
2	DA	H8	1	DG5	H2'	2.66	3.89
2	DA	H8	1	DG5	H3'	3.52	6.6
2	DA	H8	1	DG5	H8	4.62	6.29
2	DA	H8	2	DA	H1'	2.54	3.47
2	DA	H8	2	DA	H2''	2.21	2.93
2	DA	H8	2	DA	H2'	2.56	3.52
2	DA	H8	2	DA	H3'	3.08	3.81
2	DA	H2	2	DA	H1'	3.58	4.69

3	DG	H1'	2	DA	H2	3.78	4.92
3	DG	H2''	3	DG	H1'	3.16	5.34
3	DG	H2'	3	DG	H1'	2.04	3.10
3	DG	H3'	2	DA	H2''	2.91	5.43
3	DG	H3'	3	DG	H1'	2.54	3.54
3	DG	H3'	3	DG	H2''	2.10	2.69
3	DG	H3'	3	DG	H2'	2.16	2.64
3	DG	H8	2	DA	H1'	1.99	2.67
3	DG	H8	2	DA	H2''	2.78	5.84
3	DG	H8	2	DA	H2'	1.88	2.63
3	DG	H8	2	DA	H8	3.98	6.42
3	DG	H8	3	DG	H1'	2.84	4.75
3	DG	H8	3	DG	H2''	2.84	5.09
3	DG	H8	3	DG	H2'	1.63	2.37
3	DG	H8	3	DG	H3'	2.08	2.95
4	DA	H2''	4	DA	H1'	1.70	2.38
4	DA	H2'	4	DA	H1'	2.29	3.17
4	DA	H3'	4	DA	H1'	3.57	5.77
4	DA	H3'	4	DA	H2''	2.03	2.75
4	DA	H3'	4	DA	H2'	2.40	3.31
4	DA	H8	3	DG	H1'	2.72	4.04
4	DA	H8	3	DG	H2''	2.93	4.11
4	DA	H8	3	DG	H2'	1.64	2.23
4	DA	H8	3	DG	H8	2.22	4.27
4	DA	H8	4	DA	H1'	2.71	4.21
4	DA	H8	4	DA	H2''	2.02	3.04
4	DA	H8	4	DA	H2'	2.54	3.56

4	DA	H8	4	DA	H3'	2.75	3.47
4	DA	H2	4	DA	H1'	3.66	5.04
5	AMG	H101	4	DA	H2''	1.83	2.48
5	AMG	H101	4	DA	H2'	2.11	2.68
5	AMG	H101	4	DA	H3'	2.92	5.40
5	AMG	H101	4	DA	H8	2.29	4.08
5	AMG	H102	4	DA	H2'	2.19	2.42
5	AMG	H102	4	DA	H2''	1.58	2.98
5	AMG	H102	4	DA	H3'	3.07	6.23
5	AMG	H102	4	DA	H8	2.49	4.48
5	AMG	H1'	4	DA	H2	3.70	5.21
5	AMG	H2'	4	DA	H2''	2.10	5.82
5	AMG	H2'	5	AMG	H1'	2.13	3.03
5	AMG	H3'	4	DA	H1'	3.43	5.16
5	AMG	H3'	4	DA	H2''	2.24	3.03
5	AMG	H3'	5	AMG	H1'	2.11	2.87
5	AMG	H3'	5	AMG	H2'	1.77	2.26
5	AMG	H8	4	DA	H1'	3.58	5.80
5	AMG	H8	4	DA	H2''	2.19	4.64
5	AMG	H8	4	DA	H2'	2.83	4.00
5	AMG	H8	4	DA	H3'	3.89	6.12
5	AMG	H8	4	DA	H8	3.50	5.84
5	AMG	H8	5	AMG	H101	2.99	5.63
5	AMG	H8	5	AMG	H102	2.61	5.45
5	AMG	H8	5	AMG	H1'	2.90	4.58
5	AMG	H8	5	AMG	H2'	2.10	3.34
5	AMG	H8	5	AMG	H3'	2.56	3.87

6	DC	H2''	6	DC	H1'	1.96	2.65
6	DC	H2'	5	AMG	H1'	3.47	5.23
6	DC	H2'	6	DC	H1'	2.51	3.77
6	DC	H3'	6	DC	H1'	3.48	5.53
6	DC	H3'	6	DC	H2'	2.36	2.99
6	DC	H6	5	AMG	H1'	3.06	4.56
6	DC	H6	5	AMG	H2'	2.66	3.40
6	DC	H6	5	AMG	H8	4.23	5.91
6	DC	H6	6	DC	H1'	3.06	4.16
6	DC	H6	6	DC	H2''	2.76	4.20
6	DC	H6	6	DC	H2'	2.20	2.63
6	DC	H6	6	DC	H3'	2.91	4.17
6	DC	H5	5	AMG	H1'	2.98	4.06
6	DC	H5	5	AMG	H2'	2.48	3.31
6	DC	H5	5	AMG	H3'	3.63	5.44
6	DC	H5	5	AMG	H8	3.57	4.56
6	DC	H5	6	DC	H2''	3.66	6.16
6	DC	H5	6	DC	H2'	3.18	4.40
7	DG	H2''	7	DG	H1'	1.90	2.82
7	DG	H2'	7	DG	H1'	1.96	2.79
7	DG	H3'	6	DC	H2''	2.22	4.47
7	DG	H3'	7	DG	H1'	1.62	2.10
7	DG	H3'	7	DG	H2''	2.09	3.94
7	DG	H3'	7	DG	H2'	1.96	3.78
7	DG	H8	6	DC	H1'	2.27	3.13
7	DG	H8	6	DC	H2''	1.69	2.38
7	DG	H8	6	DC	H2'	2.42	5.27

7	DG	H8	6	DC	H6	2.86	4.35
7	DG	H8	7	DG	H1'	2.03	3.54
7	DG	H8	7	DG	H2''	2.07	4.05
7	DG	H8	7	DG	H2'	2.28	4.75
7	DG	H8	7	DG	H3'	2.03	3.15
8	DC	H2''	8	DC	H1'	1.68	2.37
8	DC	H2'	8	DC	H1'	2.28	4.06
8	DC	H3'	8	DC	H1'	2.95	5.04
8	DC	H3'	8	DC	H2''	2.48	4.64
8	DC	H3'	8	DC	H2'	2.11	2.87
8	DC	H6	7	DG	H1'	2.32	3.14
8	DC	H6	7	DG	H2''	2.43	4.92
8	DC	H6	7	DG	H2'	2.10	4.11
8	DC	H6	7	DG	H3'	2.68	4.30
8	DC	H6	7	DG	H8	2.78	4.32
8	DC	H6	8	DC	H2''	2.75	4.37
8	DC	H6	8	DC	H2'	2.17	2.78
8	DC	H6	8	DC	H3'	3.10	4.02
8	DC	H5	7	DG	H1'	2.85	3.70
8	DC	H5	7	DG	H2''	2.60	5.47
8	DC	H5	7	DG	H2'	2.50	4.59
8	DC	H5	7	DG	H3'	2.87	4.01
8	DC	H5	7	DG	H8	3.08	3.84
8	DC	H5	8	DC	H2''	3.81	5.91
8	DC	H5	8	DC	H2'	3.36	4.46
8	DC	H5	8	DC	H3'	4.79	6.91
9	DT	H1'	8	DC	H1'	3.00	4.99

9	DT	H2''	9	DT	H1'	1.73	2.30
9	DT	H2'	9	DT	H1'	2.42	4.87
9	DT	H3'	9	DT	H1'	1.80	2.40
9	DT	H3'	9	DT	H2''	2.16	3.31
9	DT	H3'	9	DT	H2'	2.03	2.57
9	DT	H6	8	DC	H1'	2.97	6.01
9	DT	H6	8	DC	H2'	2.53	4.13
9	DT	H6	8	DC	H3'	3.51	5.01
9	DT	H6	8	DC	H6	3.17	5.10
9	DT	H6	9	DT	H2'	2.09	3.13
9	DT	ME	8	DC	H1'	2.84	5.00
9	DT	ME	8	DC	H2''	2.98	4.11
9	DT	ME	8	DC	H2'	2.96	4.04
9	DT	ME	8	DC	H3'	3.39	4.29
9	DT	ME	8	DC	H6	2.85	3.74
9	DT	ME	8	DC	H5	2.97	3.68
9	DT	ME	9	DT	H1'	3.33	5.85
9	DT	ME	9	DT	H2''	3.16	6.01
9	DT	ME	9	DT	H2'	3.30	5.08
9	DT	ME	9	DT	H3'	3.70	5.88
9	DT	ME	9	DT	H6	2.70	3.34
10	DC	H2''	10	DC	H1'	1.63	2.19
10	DC	H2'	10	DC	H1'	2.28	3.86
10	DC	H3'	9	DT	H1'	2.88	5.40
10	DC	H3'	10	DC	H1'	3.33	5.52
10	DC	H3'	10	DC	H2''	2.16	3.72
10	DC	H3'	10	DC	H2'	2.07	2.63

10	DC	H6	9	DT	H1'	2.29	4.09
10	DC	H6	9	DT	H2''	1.90	2.89
10	DC	H6	9	DT	H2'	3.10	5.10
10	DC	H6	9	DT	H3'	2.70	5.89
10	DC	H6	9	DT	H6	2.45	3.61
10	DC	H6	10	DC	H1'	2.86	4.39
10	DC	H6	10	DC	H2'	1.90	2.38
10	DC	H6	10	DC	H3'	2.97	5.84
10	DC	H5	9	DT	H1'	3.02	5.31
10	DC	H5	9	DT	H2''	2.57	3.66
10	DC	H5	9	DT	H2'	2.81	4.45
10	DC	H5	9	DT	H3'	3.10	4.06
10	DC	H5	9	DT	H6	3.07	3.65
10	DC	H5	9	DT	ME	3.27	3.99
10	DC	H5	10	DC	H1'	3.36	5.49
10	DC	H5	10	DC	H2'	2.92	4.98
10	DC	H5	10	DC	H3'	3.47	4.55
11	DT	H2''	11	DT	H1'	1.79	2.48
11	DT	H2'	11	DT	H1'	2.46	4.19
11	DT	H3'	10	DC	H1'	2.97	5.45
11	DT	H3'	11	DT	H1'	2.27	4.22
11	DT	H3'	11	DT	H2''	2.15	2.68
11	DT	H3'	11	DT	H2'	1.94	2.46
11	DT	H6	10	DC	H1'	2.44	3.46
11	DT	H6	10	DC	H3'	3.06	3.96
11	DT	H6	11	DT	H1'	2.01	3.21
11	DT	H6	11	DT	H2''	1.87	2.50

11	DT	H6	11	DT	H2'	2.27	3.12
11	DT	H6	11	DT	H3'	2.00	2.69
11	DT	ME	10	DC	H1'	3.34	4.68
11	DT	ME	10	DC	H3'	3.74	5.51
11	DT	ME	10	DC	H6	3.05	3.52
11	DT	ME	10	DC	H5	2.82	3.53
11	DT	ME	11	DT	H1'	2.98	5.15
11	DT	ME	11	DT	H2''	2.65	4.92
11	DT	ME	11	DT	H2'	3.40	6.27
11	DT	ME	11	DT	H3'	3.12	4.92
11	DT	ME	11	DT	H6	2.44	2.87
12	DC3	H2'	12	DC3	H1'	1.74	2.35
12	DC3	H3'	12	DC3	H2'	1.87	2.41
12	DC3	H6	11	DT	H1'	2.8	5.35
12	DC3	H6	11	DT	H2'	2.19	3.35
12	DC3	H6	11	DT	H3'	1.96	2.46
12	DC3	H6	11	DT	H6	2.21	3.34
12	DC3	H6	12	DC3	H1'	3.16	4.91
12	DC3	H6	12	DC3	H2'	1.78	2.40
12	DC3	H6	12	DC3	H3'	2.48	3.08
12	DC3	H5	11	DT	H2'	2.48	3.24
12	DC3	H5	11	DT	H3'	3.21	5.59
12	DC3	H5	11	DT	ME	3.52	5.06
12	DC3	H5	12	DC3	H1'	3.18	6.10
12	DC3	H5	12	DC3	H2'	2.85	5.14
12	DC3	H5	12	DC3	H3'	3.63	5.75
13	DG5	H2''	13	DG5	H1'	1.84	2.65

13	DG5	H2'	13	DG5	H1'	2.12	3.46
13	DG5	H3'	13	DG5	H1'	2.13	3.41
13	DG5	H3'	13	DG5	H2''	2.35	4.26
13	DG5	H3'	13	DG5	H2'	1.72	2.41
13	DG5	H8	13	DG5	H1'	2.69	5.12
13	DG5	H8	13	DG5	H2'	1.72	3.14
13	DG5	H8	13	DG5	H3'	3.29	4.85
14	DA	H2''	14	DA	H1'	2.09	2.40
14	DA	H2'	14	DA	H1'	2.20	2.91
14	DA	H3'	14	DA	H1'	2.79	3.97
14	DA	H3'	14	DA	H2''	2.21	2.91
14	DA	H3'	14	DA	H2'	2.74	3.69
14	DA	H8	13	DG5	H1'	3.07	4.67
14	DA	H8	13	DG5	H2''	2.48	3.45
14	DA	H8	13	DG5	H2'	2.62	3.77
14	DA	H8	13	DG5	H3'	3.49	6.67
14	DA	H8	13	DG5	H8	4.24	6.60
14	DA	H8	14	DA	H1'	2.57	3.50
14	DA	H8	14	DA	H2''	2.22	2.99
14	DA	H8	14	DA	H2'	2.53	3.51
14	DA	H8	14	DA	H3'	3.10	3.99
14	DA	H2	11	DT	H1'	3.81	5.56
14	DA	H2	12	DC3	H1'	3.84	5.40
14	DA	H2	14	DA	H1'	3.43	4.82
15	DG	H1'	14	DA	H2	3.74	5.06
15	DG	H2''	15	DG	H1'	3.11	5.18
15	DG	H2'	15	DG	H1'	2.07	2.85

15	DG	H3'	14	DA	H2''	3.10	5.54
15	DG	H3'	15	DG	H1'	2.49	3.50
15	DG	H3'	15	DG	H2''	2.08	2.68
15	DG	H3'	15	DG	H2'	2.17	2.73
15	DG	H8	14	DA	H1'	1.95	2.61
15	DG	H8	14	DA	H2''	2.70	5.85
15	DG	H8	14	DA	H2'	1.95	2.59
15	DG	H8	14	DA	H8	3.86	5.60
15	DG	H8	15	DG	H1'	2.61	4.50
15	DG	H8	15	DG	H2''	2.73	4.76
15	DG	H8	15	DG	H2'	1.72	2.31
15	DG	H8	15	DG	H3'	2.02	3.04
16	DA	H2''	16	DA	H1'	1.70	2.35
16	DA	H2'	16	DA	H1'	2.21	3.13
16	DA	H3'	16	DA	H1'	3.73	5.59
16	DA	H3'	16	DA	H2''	2.07	2.71
16	DA	H3'	16	DA	H2'	2.32	3.22
16	DA	H8	15	DG	H1'	2.69	4.49
16	DA	H8	15	DG	H2''	2.90	4.06
16	DA	H8	15	DG	H2'	1.75	2.29
16	DA	H8	15	DG	H8	2.23	4.19
16	DA	H8	16	DA	H1'	2.78	5.10
16	DA	H8	16	DA	H2''	2.05	2.89
16	DA	H8	16	DA	H2'	2.45	3.51
16	DA	H8	16	DA	H3'	2.77	3.57
16	DA	H2	9	DT	H1'	3.73	5.46
16	DA	H2	10	DC	H1'	3.07	4.01

16	DA	H2	16	DA	H1'	3.59	5.10
17	AMG	H101	16	DA	H2''	1.84	2.49
17	AMG	H101	16	DA	H2'	2.08	2.67
17	AMG	H101	16	DA	H3'	2.89	5.29
17	AMG	H101	16	DA	H8	2.46	3.79
17	AMG	H102	16	DA	H2'	2.18	2.41
17	AMG	H102	16	DA	H2''	1.58	2.98
17	AMG	H102	16	DA	H3'	3.18	6.08
17	AMG	H102	16	DA	H8	2.45	4.04
17	AMG	H1'	16	DA	H2	3.69	4.88
17	AMG	H2'	16	DA	H2''	2.04	5.47
17	AMG	H2'	17	AMG	H1'	2.14	2.90
17	AMG	H3'	16	DA	H1'	3.57	5.40
17	AMG	H3'	16	DA	H2''	2.32	3.09
17	AMG	H3'	17	AMG	H1'	2.09	2.72
17	AMG	H3'	17	AMG	H2'	1.76	2.30
17	AMG	H8	16	DA	H1'	3.46	5.16
17	AMG	H8	16	DA	H2''	2.16	4.39
17	AMG	H8	16	DA	H2'	2.75	4.15
17	AMG	H8	16	DA	H3'	3.77	6.15
17	AMG	H8	16	DA	H8	3.55	6.12
17	AMG	H8	17	AMG	H101	2.88	5.64
17	AMG	H8	17	AMG	H102	2.61	5.50
17	AMG	H8	17	AMG	H1'	3.03	4.73
17	AMG	H8	17	AMG	H2'	2.12	3.31
17	AMG	H8	17	AMG	H3'	2.55	3.86
18	DC	H2''	18	DC	H1'	1.98	2.71

18	DC	H2'	17	AMG	H1'	2.97	5.25
18	DC	H2'	18	DC	H1'	2.48	3.45
18	DC	H3'	18	DC	H1'	3.47	5.20
18	DC	H3'	18	DC	H2'	2.34	3.02
18	DC	H6	17	AMG	H1'	3.05	4.22
18	DC	H6	17	AMG	H2'	2.60	3.41
18	DC	H6	17	AMG	H8	3.96	6.16
18	DC	H6	18	DC	H1'	3.16	3.97
18	DC	H6	18	DC	H2''	2.80	5.15
18	DC	H6	18	DC	H2'	2.21	2.61
18	DC	H6	18	DC	H3'	2.91	4.17
18	DC	H5	17	AMG	H1'	3.04	3.99
18	DC	H5	17	AMG	H2'	2.38	3.44
18	DC	H5	17	AMG	H3'	3.57	5.56
18	DC	H5	17	AMG	H8	3.50	4.65
18	DC	H5	18	DC	H2''	3.58	6.46
18	DC	H5	18	DC	H2'	3.09	4.25
19	DG	H2''	19	DG	H1'	1.97	3.38
19	DG	H2'	19	DG	H1'	1.96	2.85
19	DG	H3'	18	DC	H2''	2.30	4.22
19	DG	H3'	19	DG	H1'	1.63	2.09
19	DG	H3'	19	DG	H2''	2.14	3.44
19	DG	H3'	19	DG	H2'	1.95	3.99
19	DG	H8	18	DC	H1'	2.26	3.12
19	DG	H8	18	DC	H2''	1.61	2.33
19	DG	H8	18	DC	H2'	2.55	5.23
19	DG	H8	18	DC	H6	2.89	4.41

19	DG	H8	19	DG	H1'	1.96	3.55
19	DG	H8	19	DG	H2''	2.06	4.33
19	DG	H8	19	DG	H2'	2.48	5.17
19	DG	H8	19	DG	H3'	2.00	3.02
20	DC	H2''	20	DC	H1'	1.65	2.33
20	DC	H2'	20	DC	H1'	2.42	4.06
20	DC	H3'	20	DC	H1'	3.03	4.71
20	DC	H3'	20	DC	H2''	2.47	4.86
20	DC	H3'	20	DC	H2'	2.23	2.78
20	DC	H6	19	DG	H1'	2.33	3.13
20	DC	H6	19	DG	H2''	2.44	4.61
20	DC	H6	19	DG	H2'	2.15	3.85
20	DC	H6	19	DG	H3'	2.66	4.62
20	DC	H6	19	DG	H8	2.77	4.55
20	DC	H6	20	DC	H2''	2.84	4.77
20	DC	H6	20	DC	H2'	2.24	2.81
20	DC	H6	20	DC	H3'	3.08	3.85
20	DC	H5	19	DG	H1'	2.80	3.70
20	DC	H5	19	DG	H2''	2.68	5.63
20	DC	H5	19	DG	H2'	2.57	4.25
20	DC	H5	19	DG	H3'	2.89	3.94
20	DC	H5	19	DG	H8	3.02	3.84
20	DC	H5	20	DC	H2''	3.29	5.70
20	DC	H5	20	DC	H2'	3.37	5.06
20	DC	H5	20	DC	H3'	4.59	6.70
21	DT	H1'	4	DA	H2	3.88	5.49
21	DT	H1'	20	DC	H1'	2.97	5.64

21	DT	H2''	21	DT	H1'	1.78	2.33
21	DT	H2'	21	DT	H1'	2.69	4.12
21	DT	H3'	21	DT	H1'	1.84	2.53
21	DT	H3'	21	DT	H2''	2.12	3.37
21	DT	H3'	21	DT	H2'	2.04	2.56
21	DT	H6	20	DC	H1'	2.75	5.28
21	DT	H6	20	DC	H2'	2.60	4.42
21	DT	H6	20	DC	H3'	3.54	5.14
21	DT	H6	20	DC	H6	3.21	5.00
21	DT	H6	21	DT	H2'	2.05	3.06
21	DT	ME	20	DC	H1'	2.83	5.36
21	DT	ME	20	DC	H2''	3.33	4.54
21	DT	ME	20	DC	H2'	2.95	4.77
21	DT	ME	20	DC	H3'	3.41	4.03
21	DT	ME	20	DC	H6	3.16	6.16
21	DT	ME	20	DC	H5	3.06	4.51
21	DT	ME	21	DT	H1'	3.46	6.04
21	DT	ME	21	DT	H2''	3.21	6.23
21	DT	ME	21	DT	H2'	3.18	4.88
21	DT	ME	21	DT	H3'	4.81	6.28
21	DT	ME	21	DT	H6	2.78	3.47
22	DC	H1'	4	DA	H2	3.12	3.93
22	DC	H2''	22	DC	H1'	1.68	2.15
22	DC	H2'	22	DC	H1'	2.40	4.24
22	DC	H3'	21	DT	H1'	3.12	5.54
22	DC	H3'	22	DC	H1'	3.38	5.15
22	DC	H3'	22	DC	H2''	2.28	3.51

22	DC	H3'	22	DC	H2'	2.21	2.84
22	DC	H6	21	DT	H1'	2.35	3.71
22	DC	H6	21	DT	H2''	1.99	3.18
22	DC	H6	21	DT	H2'	3.12	4.51
22	DC	H6	21	DT	H3'	2.89	6.19
22	DC	H6	21	DT	H6	2.57	3.70
22	DC	H6	22	DC	H1'	2.99	5.16
22	DC	H6	22	DC	H2'	1.93	2.42
22	DC	H6	22	DC	H3'	2.98	4.12
22	DC	H5	21	DT	H1'	3.32	5.00
22	DC	H5	21	DT	H2''	2.69	3.63
22	DC	H5	21	DT	H2'	2.98	4.48
22	DC	H5	21	DT	H3'	3.09	4.05
22	DC	H5	21	DT	H6	3.06	3.64
22	DC	H5	21	DT	ME	3.30	4.10
22	DC	H5	22	DC	H1'	3.53	6.13
22	DC	H5	22	DC	H2'	2.92	4.55
22	DC	H5	22	DC	H3'	3.48	4.82
23	DT	H1'	2	DA	H2	3.81	5.41
23	DT	H2''	23	DT	H1'	1.8	2.72
23	DT	H2'	23	DT	H1'	2.52	3.95
23	DT	H3'	22	DC	H1'	3.02	5.40
23	DT	H3'	23	DT	H1'	2.26	4.10
23	DT	H3'	23	DT	H2''	2.16	2.74
23	DT	H3'	23	DT	H2'	1.93	2.44
23	DT	H6	22	DC	H1'	2.49	3.25
23	DT	H6	22	DC	H3'	3.09	3.98

23	DT	H6	23	DT	H1'	2.00	2.97
23	DT	H6	23	DT	H2''	1.81	2.50
23	DT	H6	23	DT	H2'	2.25	3.50
23	DT	H6	23	DT	H3'	2.01	2.81
23	DT	ME	22	DC	H1'	3.27	5.05
23	DT	ME	22	DC	H3'	3.72	5.40
23	DT	ME	22	DC	H6	3.29	6.31
23	DT	ME	22	DC	H5	2.89	3.77
23	DT	ME	23	DT	H1'	2.75	5.05
23	DT	ME	23	DT	H2''	2.80	4.82
23	DT	ME	23	DT	H2'	3.25	5.48
23	DT	ME	23	DT	H3'	3.15	5.18
23	DT	ME	23	DT	H6	2.56	2.91
24	DC3	H1'	2	DA	H2	3.88	5.45
24	DC3	H2'	24	DC3	H1'	1.69	2.37
24	DC3	H3'	24	DC3	H1'	3.88	5.03
24	DC3	H3'	24	DC3	H2'	1.82	2.41
24	DC3	H6	23	DT	H1'	2.81	5.23
24	DC3	H6	23	DT	H2'	2.12	3.92
24	DC3	H6	23	DT	H3'	2.00	2.45
24	DC3	H6	23	DT	H6	2.23	3.75
24	DC3	H6	24	DC3	H1'	3.52	5.20
24	DC3	H6	24	DC3	H2'	1.72	2.47
24	DC3	H6	24	DC3	H3'	2.55	3.11
24	DC3	H5	23	DT	H2'	2.39	3.20
24	DC3	H5	23	DT	H3'	3.34	6.34
24	DC3	H5	23	DT	ME	3.66	5.40

24	DC3	H5	24	DC3	H1'	3.24	5.82
24	DC3	H5	24	DC3	H2'	2.85	4.98
24	DC3	H5	24	DC3	H3'	3.34	6.59

Table S3. Backbone restraints generated for OL-1b duplex used in rMD caluations.

2	DA	ALPHA	-90	-30
3	DG	ALPHA	-90	-30
4	DA	ALPHA	-90	-30
5	AMG	ALPHA	-120	0
6	DC	ALPHA	-90	-30
7	DG	ALPHA	-90	-30
8	DC	ALPHA	-90	-30
9	DT	ALPHA	-90	-30
10	DC	ALPHA	-90	-30
11	DT	ALPHA	-90	-30
14	DA	ALPHA	-90	-30
15	DG	ALPHA	-90	-30
16	DA	ALPHA	-90	-30
17	AMG	ALPHA	-120	0
18	DC	ALPHA	-90	-30
19	DG	ALPHA	-90	-30
20	DC	ALPHA	-90	-30
21	DT	ALPHA	-90	-30
22	DC A	LPHA	-90	-30
23	DT A	LPHA	-90	-30
2	DA	BETA	150	210
3	DG	BETA	150	210
4	DA	BETA	150	210
5	AMG	BETA	120	240
6	DC	BETA	150	210

7	DG	BETA	150	210
8	DC	BETA	150	210
9	DT	BETA	150	210
10	DC	BETA	150	210
11	DT	BETA	150	210
14	DA	BETA	150	210
15	DG	BETA	150	210
16	DA	BETA	150	210
17	AMG	BETA	120	240
18	DC	BETA	150	210
19	DG	BETA	150	210
20	DC	BETA	150	210
21	DT	BETA	150	210
22	DC	BETA	150	210
23	DT	BETA	150	210
2	DA	GAMMA	30	90
3	DG	GAMMA	30	90
4	DA	GAMMA	30	90
5	AMG	GAMMA	0	120
6	DC	GAMMA	30	90
7	DG	GAMMA	30	90
8	DC	GAMMA	30	90
9	DT	GAMMA	30	90
10	DC	GAMMA	30	90
11	DT	GAMMA	30	90
14	DA	GAMMA	30	90
15	DG	GAMMA	30	90

16	DA	GAMMA	30	90
17	AMG	GAMMA	0	120
18	DC	GAMMA	30	90
19	DG	GAMMA	30	90
20	DC	GAMMA	30	90
21	DT	GAMMA	30	90
22	DC	GAMMA	30	90
23	DT	GAMMA	30	90
2	DA	EPSILN	165	225
3	DG	EPSILN	165	225
4	DA	EPSILN	165	225
5	AMG	EPSILN	135	225
6	DC	EPSILN	165	225
7	DG	EPSILN	165	225
8	DC	EPSILN	165	225
9	DT	EPSILN	165	225
10	DC	EPSILN	165	225
11	DT	EPSILN	165	225
14	DA	EPSILN	165	225
15	DG	EPSILN	165	225
16	DA	EPSILN	165	225
17	AMG	EPSILN	135	225
18	DC	EPSILN	165	225
19	DG	EPSILN	165	225
20	DC	EPSILN	165	225
21	DT	EPSILN	165	225
22	DC	EPSILN	165	225

23	DT	EPSILN	165	225
2	DA	ZETA	-135	-75
3	DG	ZETA	-135	-75
4	DA	ZETA	-135	-75
5	AMG	ZETA	-165	-45
6	DC	ZETA	-135	-75
7	DG	ZETA	-135	-75
8	DC	ZETA	-135	-75
9	DT	ZETA	-135	-75
10	DC	ZETA	-135	-75
11	DT	ZETA	-135	-75
14	DA	ZETA	-135	-75
15	DG	ZETA	-135	-75
16	DA	ZETA	-135	-75
17	AMG	ZETA	-165	-45
18	DC	ZETA	-135	-75
19	DG	ZETA	-135	-75
20	DC	ZETA	-135	-75
21	DT	ZETA	-135	-75
22	DC	ZETA	-135	-75
23	DT	ZETA	-135	-75

Table S4. Deoxyribose torsion angles restraints generated for OL-1b used in rMD calculation.

2	DA	PPA	100	165
3	DG	PPA	125	165
4	DA	PPA	125	165
5	DG	PPA	125	165
6	DC	PPA	90	130
7	DG	PPA	125	165
8	DC	PPA	90	130
9	DT	PPA	90	130
10	DC	PPA	90	130
11	DT	PPA	100	165
14	DA	PPA	100	165
15	DG	PPA	125	165
16	DA	PPA	125	165
17	DG	PPA	125	165
18	DC	PPA	90	130
19	DG	PPA	125	165
20	DC	PPA	90	130
21	DT	PPA	90	130
22	DC	PPA	90	130
23	DT	PPA	100	165

Table S5. Base pair restraints generated for OL-1b used in rMD calculations.

1	DG5	H1	24	DC3	N3	1.84	2.04
1	DG5	H22	24	DC3	O2	1.75	1.95
1	DG5	N1	24	DC3	N3	2.85	3.05
1	DG5	O6	24	DC3	H42	1.8	2
1	DG5	O6	24	DC3	N4	2.81	3.01
2	DA	H61	23	DT	O4	1.84	2.04
2	DA	N1	23	DT	H3	1.71	1.91
2	DA	N1	23	DT	N3	2.72	2.92
3	DG	H1	22	DC	N3	1.84	2.04
3	DG	H22	22	DC	O2	1.75	1.95
3	DG	N1	22	DC	N3	2.85	3.05
3	DG	O6	22	DC	H42	1.8	2
3	DG	O6	22	DC	N4	2.81	3.01
4	DA	H61	21	DT	O4	1.84	2.04
4	DA	N1	21	DT	H3	1.71	1.91
4	DA	N1	21	DT	N3	2.72	2.92
5	AMG	H1	20	DC	N3	1.84	2.04
5	AMG	H22	20	DC	O2	1.75	1.95
5	AMG	N1	20	DC	N3	2.85	3.05
5	AMG	O6	20	DC	H42	1.8	2
5	AMG	O6	20	DC	N4	2.81	3.01
6	DC	H42	19	DG	O6	1.8	2
6	DC	N3	19	DG	H1	1.84	2.04
6	DC	N3	19	DG	N1	2.85	3.05
6	DC	N4	19	DG	O6	2.81	3.01

6	DC	O2	19	DG	H22	1.75	1.95
7	DG	H1	18	DC	N3	1.84	2.04
7	DG	H22	18	DC	O2	1.75	1.95
7	DG	N1	18	DC	N3	2.85	3.05
7	DG	O6	18	DC	H42	1.8	2
7	DG	O6	18	DC	N4	2.81	3.01
8	DC	H42	17	AMG	O6	1.8	2
8	DC	N3	17	AMG	H1	1.84	2.04
8	DC	N3	17	AMG	N1	2.85	3.05
8	DC	N4	17	AMG	O6	2.81	3.01
8	DC	O2	17	AMG	H22	1.75	1.95
9	DT	H3	16	DA	N1	1.71	1.91
9	DT	N3	16	DA	N1	2.72	2.92
9	DT	O4	16	DA	H61	1.84	2.04
10	DC	H42	15	DG	O6	1.8	2
10	DC	N3	15	DG	H1	1.84	2.04
10	DC	N3	15	DG	N1	2.85	3.05
10	DC	N4	15	DG	O6	2.81	3.01
10	DC	O2	15	DG	H22	1.75	1.95
11	DT	H3	14	DA	N1	1.71	1.91
11	DT	N3	14	DA	N1	2.72	2.92
11	DT	O4	14	DA	H61	1.84	2.04
12	DC3	H42	13	DG5	O6	1.8	2
12	DC3	N3	13	DG5	H1	1.84	2.04
12	DC3	N3	13	DG5	N1	2.85	3.05
12	DC3	N4	13	DG5	O6	2.81	3.01
12	DC3	O2	13	DG5	H22	1.75	1.95

Table S6. Helicoidal analysis for the OL-1b duplex carried out with the program Curves+ (*43*).

Base pair	Shift (Å)	Slide (Å)	Rise (Å)	Tilt (°)	Roll (°)	Twist (°)	Buckle (°)	Propel (°)	Opening (°)
G^1:C^{12}	0.26	-0.60	3.14	-3.7	-0.1	30.0	-0.9	-9.1	2.4
A^2:T^{11}	-0.98	-0.76	3.32	-0.8	6.8	31.1	9.0	-17.1	4.5
G^3:C^{10}	-0.78	-0.22	3.41	0.8	12.6	35.2	4.5	-7.8	-0.8
A^4:T^9	0.05	-0.53	3.31	-5.7	1.4	31.3	-0.5	-16	-0.5
X^5:C^8	0.09	-0.36	3.19	1.6	1.2	37.1	-6.3	-14.2	1.4
C^6:G^7	0.11	-0.49	3.16	0.3	6.8	32.0	-0.7	-8.6	0.4
$G^7 \cdot C^6$	-0.09	-0.41	3.23	-0.9	1.2	37.0	1.3	-9.5	0.3
$C^8 \cdot X^5$	-0.02	-0.53	3.34	5.3	1.9	31.6	5.9	-15	1.2
T^9:A^4	0.74	-0.24	3.37	-0.9	12.2	34.8	-0.5	-16.2	-0.2
C^{10}:G^3	0.99	-0.81	3.32	1.0	6.3	31.1	-4.1	-7.3	-1.0
T^{11}:A^2	-0.21	-0.60	3.12	3.8	-0.2	30.2	-8.6	-16.9	4.0
C^{12}:G^1							1.6	-10.1	2.4

Table S7. Backbone torsion angles of the refined OL-1b duplex, calculated from the program Curves+ (*44*). Values are averaged from two symmetry-related strands.

Base pair	Alpha (°)	Beta (°)	Gamma (°)	Delta (°)	Epsilon (°)	Zeta (°)
G^{1}:C^{12}			51.0	142.9	-174.3	-81.7
A^{2}:T^{11}	-67.8	170.7	56.7	108.3	-173.3	-97.3
G^{3}:C^{10}	-67.1	178.7	48.9	135.3	-174.6	-106.5
A^{4}:T^{9}	-70.0	-168.9	44.9	137.7	176.5	-98.5
X^{5}:C^{8}	-65.7	170.7	58.3	113.9	-172.3	-87.8
C^{6}:G^{7}	-65.7	174.4	55.2	119.6	-175.2	-95.2
G^{7}·C^{6}	-65.8	175.6	53.3	121.8	-175.2	-95.4
C^{8}·X^{5}	-68.0	170.5	58.4	104.1	-176.9	-84.6
T^{9}:A^{4}	-63.5	169.1	55.4	115.9	-170.0	-79.8
C^{10}:G^{3}	-66.2	173.8	47.7	117.5	-174.2	-86.5
T^{11}:A^{2}	-65.2	166.6	46.1	84.5	-175.0	-82.9
C^{12}:G^{1}	-68.5	166.9	54.9	83.8		

Figure S1. Backbone torsion angles of the refined OL-1b duplex calculated from the program Curves+ (*44*).

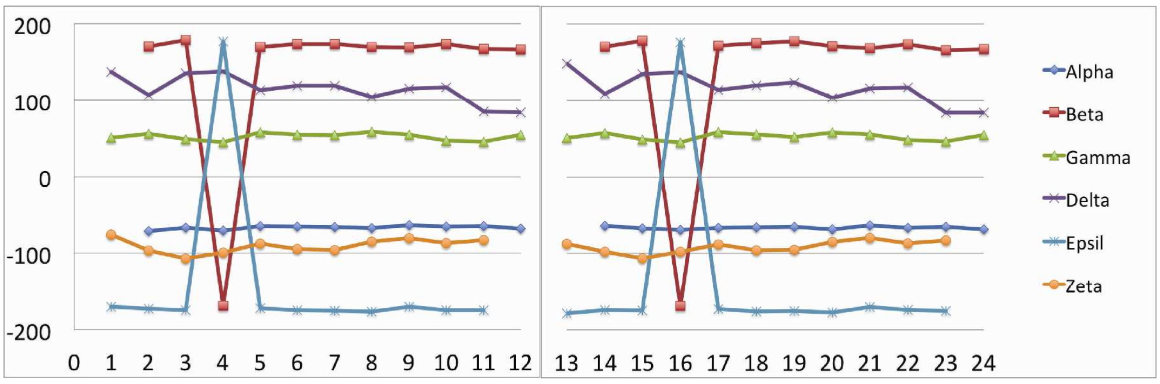


Figure S2. Linewidths of the guanine N1H and thymine N3H imino protons as a function of temperature. **A.** Unmodified OL-1a duplex. **B.** Modified OL-1b duplex.

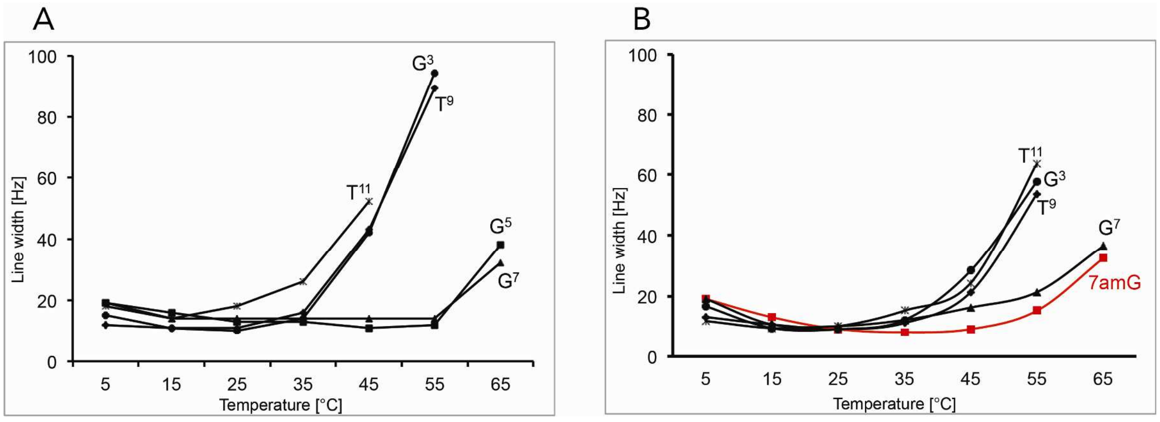
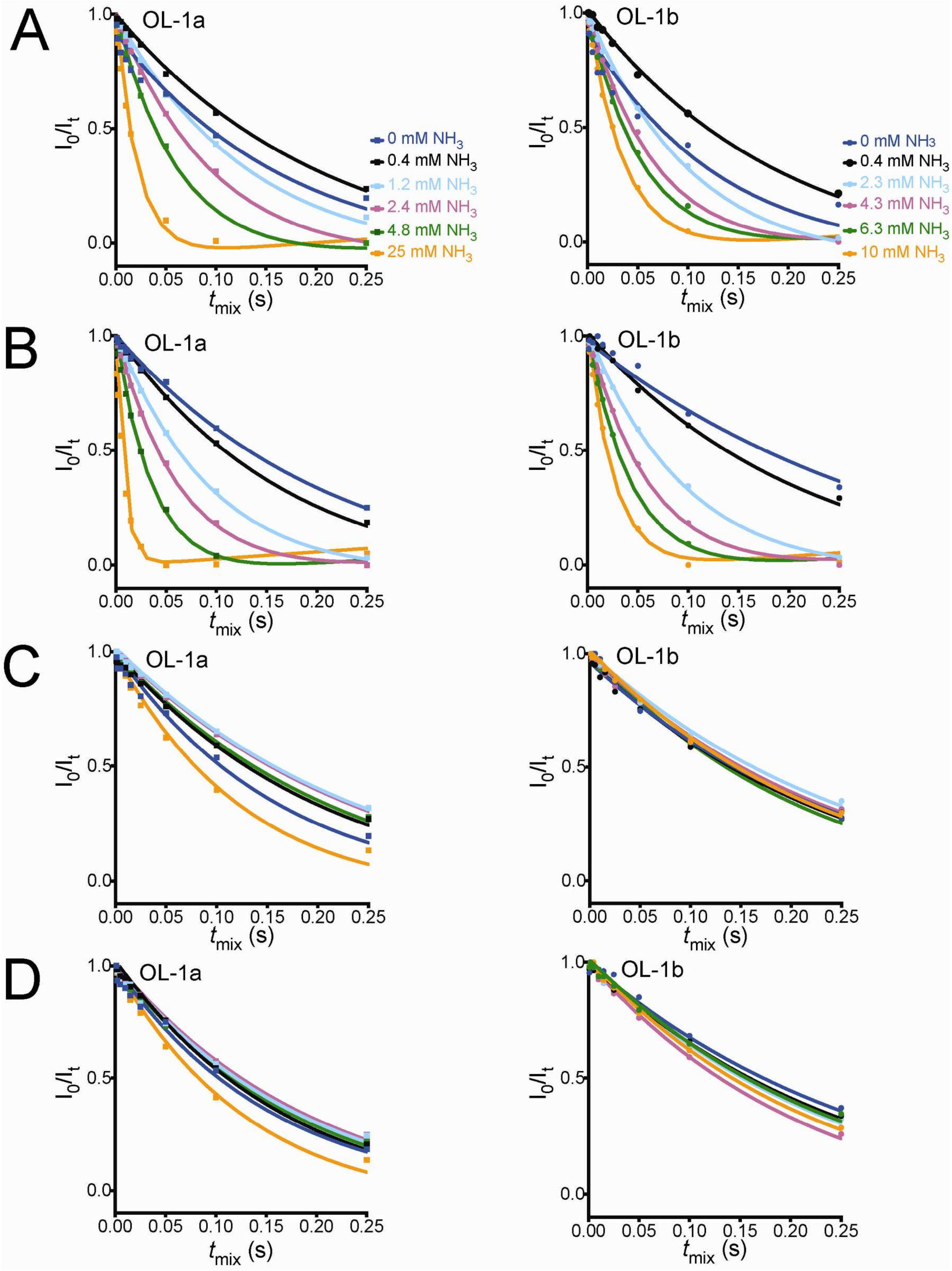


Figure S3. Representative time courses of transfer of solvent magnetization to the imino protons. **A.** The left panel shows base pair G^3:C^{10} in the OL-1a duplex and the right panel shows base pair G^3:C^{10} in the OL-1b duplex. **B.** The left panel shows base pair A^4:T^9 in the OL-1a duplex and the right panel shows base pair A^4:T^9 in the OL-1b duplex. **C.** The left panel shows base pair G^5:C^8 in the OL-1a duplex and the right panel shows base pair X^5:C^8 in the OL-1b duplex. **D.** The left panel shows base pair C^6:G^7 in the OL-1a duplex and the right panel shows base pair C^6:G^7 in the OL-1b duplex.


A
OL-1a
OL-1b
I_0/I_t
1.0
0.5
0.0
0.00 0.05 0.10 0.15 0.20 0.25
t_{mix} (s)
0 mM NH_3
0.4 mM NH_3
1.2 mM NH_3
2.4 mM NH_3
4.8 mM NH_3
25 mM NH_3
0 mM NH_3
0.4 mM NH_3
2.3 mM NH_3
4.3 mM NH_3
6.3 mM NH_3
10 mM NH_3
B
OL-1a
OL-1b
C
OL-1a
OL-1b
D
OL-1a
OL-1b