Supporting Information: Correlated Motions of C'─N and C_α─C_β pairs in Protonated and Per-deuterated GB3.

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SI1. Effect of fast motion order parameters.

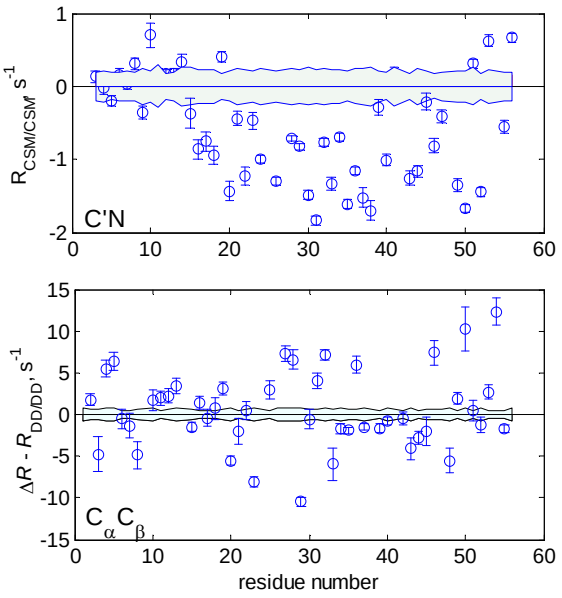

Figure S1. Top: $R_{CSM\,/\,CSM}$ *vs* residue number for the C'─N experiment for the protonated sample at 500 MHz. The zero line sets reference to $R_{CSA\,/\,CSA}$ and $R_{DD\,/\,DD}$ contributions with all order parameters of 0.8, as in Figure 4 of the main text. The aqua-colored bands represent *alternative* $R_{CSA\,/\,CSA}$ and $R_{DD\,/\,DD}$ contributions within the order parameter range of 0.7-0.9. **Bottom:** $\Delta R - R_{DD/DD}$ *vs* residue number for the C_α–C_β experiment for the protonated sample at 500 MHz. The zero line sets reference to $R_{DD\,/\,DD}$ contributions calculated as in Figure 4 of the main text: A value of 0.8 was used for all dipolar interactions except for those involving H_β or D_β, for which a value of 0.5 was used. The aqua-colored band represents *alternative* $R_{DD\,/\,DD}$ contributions within the following order parameter ranges: 0.7─0.9 for all dipolar interactions except for those involving H_β or D_β, for which a range of 0.3─0.7 is used.

SI2. Expressions for calculations of the $R_{CSA/CSA}$ term.

The following equation was used to calculate $R_{CSA/CSA}$ rates for two anisotropic σ^C and σ^N CSA tensors:

$$\begin{aligned} R_{CSA/CSA} = & \left(\frac{2}{18}\right) B_0^2 \gamma_N \gamma_C \frac{8}{5} \tau_c \\ & \times \left\{ \left[\sigma_{xx}{}^C - \sigma_{zz}{}^C\right]\left[\sigma_{xx}{}^N - \sigma_{zz}{}^N\right] P_2\left(\cos(\beta_C + 180^\circ - \alpha + \beta_N)\right) \right. \\ & + \left[\sigma_{xx}{}^C - \sigma_{zz}{}^C\right]\left[\sigma_{yy}{}^N - \sigma_{zz}{}^N\right] P_2\left(\cos 90^\circ\right) \\ & + \left[\sigma_{yy}{}^C - \sigma_{zz}{}^C\right]\left[\sigma_{xx}{}^N - \sigma_{zz}{}^N\right] P_2\left(\cos(180^\circ - \alpha + \beta_N - (90^\circ - \beta_C))\right) \\ & \left. + \left[\sigma_{yy}{}^C - \sigma_{zz}{}^C\right]\left[\sigma_{yy}{}^N - \sigma_{zz}{}^N\right] P_2\left(\cos 90^\circ\right) \right\} S^2_{CSA/CSA} \end{aligned}$$

where α is the $C'_{i-1}N_iH^N$ angle, β_N is the angle between $\sigma_{xx}{}^N$ (the longest component of the ^{15}N CSA tensor) and the N-H bond, β_C is the angle between $\sigma_{xx}{}^C$ and the C′-N bond, $\sigma_{zz}{}^C$ and $\sigma_{yy}{}^N$ are directed perpendicular to the peptide plane. The reference frame is defined in detail in Figure 1 of Loth *et al*.(Loth et al. 2005). The principal components are defined so that $\sigma_{xx} > \sigma_{yy} > \sigma_{zz}$.

In the case of $C_\alpha C_\beta$, the tensors were assumed to be axially symmetric with the components $\Delta\sigma_{C_\alpha}$ and $\Delta\sigma_{C_b}$:

$$R_{CSA/CSA} = \frac{8}{45}(B_0\gamma_C)^2 \Delta\sigma_{C\alpha}\Delta\sigma_{C\beta} P_2(\cos\theta_{C\alpha,C\beta}) S^2_{CSA/CSA}$$

where $\theta_{C\alpha,C\beta}$ is the angle between the symmetry axes of the two CSA tensors.

SI3. C' chemical shift assignments for GB3.

Performed on protonated GB3 using HNCO experiment at 500 MHz and 25 °C.

Residue #	C' Shift	Residue #	C' Shift
2	174.36	29	181.64
3	174.78	30	178.43
4	172.88	31	179.51
5	174.66	32	177.16
6	174.41	33	178.92
7	175.17	34	171.26
8	175.02	35	179.33
9	173.29	36	177.15
10	178.43	37	174.07
11	173.82	38	174.29
12	173.62	39	174.09
13	176.34	40	174.64
14	179.39	41	171.84

15	175.53	42	176.71
16	172.18	43	177.00
17	173.91	44	172.78
18	171.22	45	173.25
19	175.91	46	174.54
20	177.43	47	178.11
21	174.97	48	179.94
22	174.82	49	175.27
23	179.41	50	175.05
24	177.07	51	174.96
25	177.01	52	174.44
26	179.27	53	176.71
27	177.75	54	172.62
28	179.59	55	175.97

SI4. Contributions of $R_{DD/DD}$ and $R_{CSA/CSA}$ into ΔR values.

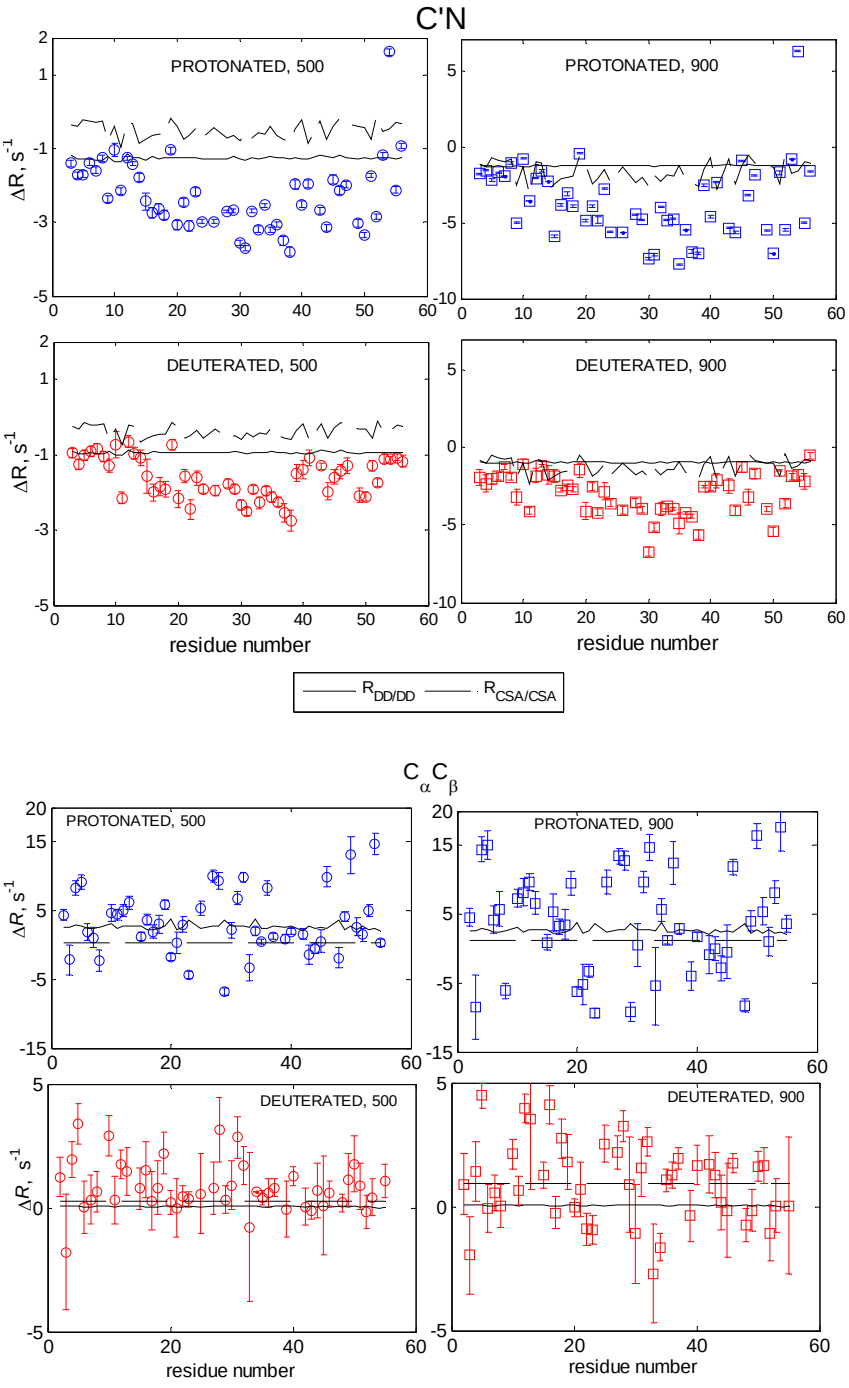

Figure SI2. Contributions of $R_{DD/DD}$ (solid lines) and $R_{CSA/CSA}$ (dotted lines) into ΔR values versus residue number. Top panels: C'N measurements. Bottom panels, $C_\alpha C_\beta$ measurements. $R_{CSA/CSA}$ values for $C_\alpha C_\beta$ measurements are shown as half the maximum values stated in main text.

SI5. Experimental values of Δ*R* rates and J-coupling constants

C'N 500 (numbering on amide)

Residue#	ΔR, s^{-1} protonated	error	$R_{DD/DD} + R_{CSA/CSA}$, s^{-1}	ΔR, s^{-1} deuterated	error	$R_{DD/DD} + R_{CSA/CSA}$, s^{-1}
3	-1.40	0.08	-1.54	-0.96	0.04	-1.22
4	-1.72	0.08	-1.71	-1.26	0.04	-1.33
5	-1.70	0.07	-1.50	-1.01	0.05	-1.12
6	-1.38	0.07	-1.54	-0.92	0.05	-1.15
7	-1.59	0.08	-1.62	-0.84	0.04	-1.22
8	-1.27	0.08	-1.58	-1.04	0.05	-1.20
9	-2.37	0.09	-2.02	-1.29	0.06	-1.64
10	-1.04	0.17	-1.74	-0.72	0.11	-1.35
11	-2.16	0.07	-2.33	-2.14	0.05	-1.88
12	-1.27	0.06	-1.43	-0.65	0.06	-1.11
13	-1.41	0.06	-1.59	-0.97	0.05	-1.22
14	-1.78	0.10	-2.12	-1.09	0.07	-1.75
15	-2.44	0.20	-2.08	-1.57	0.15	-1.67
16	-2.74	0.13	-1.88	-2.00	0.08	-1.52
17	-2.64	0.12	-1.89	-1.85	0.08	-1.50
18	-2.80	0.12	-1.86	-1.91	0.07	-1.49
19	-1.05	0.07	-1.45	-0.74	0.05	-1.11
20	-3.08	0.13	-1.64	-2.18	0.07	-1.29
21	-2.45	0.10	-2.01	-1.57	0.06	-1.61
22	-3.11	0.13	-1.88	-2.45	0.09	-1.54
23	-2.17	0.11	-1.70	-1.61	0.06	-1.34
24	-3.00	0.05	-2.00	-1.90	0.04	-1.63
26	-2.99	0.05	-1.70	-1.95	0.05	-1.32
28	-2.72	0.04	-2.00	-1.78	0.04	-1.62
29	-2.69	0.04	-1.86	-1.90	0.04	-1.51
30	-3.56	0.06	-2.07	-2.33	0.04	-1.70
31	-3.72	0.07	-1.88	-2.51	0.03	-1.49
32	-2.71	0.05	-1.94	-1.93	0.03	-1.59
33	-3.19	0.09	-1.86	-2.28	0.05	-1.48
34	-2.55	0.05	-1.85	-1.96	0.04	-1.49
35	-3.23	0.07	-1.61	-2.11	0.05	-1.28
36	-3.06	0.06	-1.90	-2.26	0.04	-1.56
37	-3.51	0.13	-1.98	-2.55	0.08	-1.64
38	-3.79	0.13	-2.09	-2.76	0.10	-1.72
39	-1.97	0.11	-1.69	-1.49	0.08	-1.36
40	-2.52	0.08	-1.51	-1.40	0.08	-1.16
41	-1.96	0.10	-2.13	-1.09	0.07	-1.75
43	-2.69	0.09	-1.44	-1.30	0.03	-1.11

44	-3.14	0.09	-1.98	-1.98	0.07	-1.63
45	-1.84	0.11	-1.63	-1.59	0.06	-1.29
46	-2.14	0.10	-1.33	-1.44	0.06	-1.04
47	-2.00	0.08	-1.58	-1.30	0.07	-1.25
49	-3.04	0.09	-1.68	-2.08	0.06	-1.32
50	-3.34	0.05	-1.67	-2.11	0.04	-1.32
51	-1.73	0.06	-2.04	-1.29	0.05	-1.67
52	-2.84	0.06	-1.39	-1.74	0.04	-1.05
53	-1.19	0.07	-1.82	-1.11	0.04	-1.45
54	1.61	0.09	-1.74	-1.13	0.03	-1.36
55	-2.13	0.09	-1.57	-1.07	0.03	-1.20
56	-0.92	0.06	-1.59	-1.17	0.05	-1.20

C'N 900 (numbering on amide)

Residue#	ΔR, s^{-1} protonated	error	$R_{DD/DD} + R_{CSA/CSA}$, s^{-1}	ΔR, s^{-1} deuterated	error	$R_{DD/DD} + R_{CSA/CSA}$, s^{-1}
3	-1.78	0.03	-2.32	-1.93	0.50	-1.56
4	-1.56	0.04	-2.64	-2.30	0.53	-1.76
5	-2.20	0.09	-1.97	-2.05	0.34	-1.24
6	-1.71	0.05	-2.14	-1.83	0.27	-1.36
7	-1.92	0.03	-2.31	-1.26	0.19	-1.48
8	-1.07	0.09	-2.15	-1.89	0.23	-1.39
9	-5.01	0.06	-3.78	-3.20	0.49	-2.62
10	-0.77	0.05	-2.64	-1.08	0.37	-1.74
11	-3.57	0.03	-4.49	-4.10	0.23	-3.11
12	-2.07	0.05	-2.09	-1.82	0.46	-1.38
13	-1.59	0.06	-2.33	-1.14	0.07	-1.53
14	-2.28	0.03	-4.08	-1.78	0.60	-2.86
15	-5.87	0.08	-5.32	-1.85	0.40	-2.60
16	-3.80	0.09	-3.32	-2.73	0.11	-2.29
17	-3.09	0.10	-3.26	-2.41	0.10	-2.23
18	-3.95	0.10	-3.20	-2.69	0.15	-2.18
19	-0.43	0.04	-1.90	-1.42	0.47	-1.21
20	-4.82	0.09	-2.53	-4.10	0.53	-1.69
21	-3.93	0.08	-3.68	-2.51	0.22	-2.53
22	-4.83	0.20	-3.31	-4.18	0.22	-2.30
23	-2.79	0.04	-2.72	-2.82	0.53	-1.82
24	-5.61	0.06	-3.63	-3.60	0.21	-2.51
26	-5.64	0.03	-2.64	-4.03	0.24	-1.76
28	-4.47	0.03	-3.64	-3.53	0.30	-2.51
29	-4.80	0.06	-3.27	-3.99	0.23	-2.26
30	-7.35	0.07	-3.86	-6.71	0.26	-2.69
31	-7.10	0.06	-3.13	-5.15	0.28	-2.12

32	-3.96	0.02	-3.52	-3.95	0.40	-2.45
33	-4.82	0.03	-3.15	-3.81	0.17	-2.15
34	-4.75	0.06	-3.18	-3.92	0.09	-2.18
35	-7.73	0.04	-4.03	-4.89	0.67	-1.66
36	-5.47	0.03	-3.36	-4.21	0.05	-2.33
37	-6.90	0.15	-3.63	-4.44	0.12	-2.54
38	-7.01	0.11	-3.91	-5.60	0.30	-2.73
39	-2.52	0.08	-2.70	-2.55	0.08	-1.84
40	-4.62	0.08	-2.03	-2.49	0.31	-1.31
41	-2.33	0.05	-4.03	-2.11	0.46	-2.81
43	-5.37	0.05	-2.00	-2.46	0.45	-1.30
44	-5.67	0.09	-3.66	-4.06	0.32	-2.56
45	-0.93	0.07	-2.50	-1.24	0.35	-1.67
46	-3.21	0.04	-1.71	-3.17	0.52	-1.11
47	-1.88	0.06	-2.32	-1.71	0.34	-1.55
49	-5.50	0.05	-2.58	-3.95	0.18	-1.72
50	-7.03	0.01	-2.61	-5.41	0.28	-1.75
51	-1.70	0.10	-3.76	-1.47	0.06	-2.61
52	-5.46	0.06	-1.73	-3.61	0.22	-1.08
53	-0.81	0.04	-3.04	-1.87	0.20	-2.07
54	6.30	0.07	-2.82	-1.77	0.38	-1.89
55	-5.01	0.03	-2.24	-2.17	0.50	-1.45
56	-1.62	0.05	-2.35	-0.52	0.21	-1.52

$C_\alpha C_\beta$ 500 (numbering on C_α)

Residue#	ΔR, s^{-1} protonated	error	$R_{DD/DD}$, s^{-1} protonated	ΔR, s^{-1} deuterated	error	$R_{DD/DD}$, s^{-1} deuterated
2	4.36	0.74	2.60	1.25	0.81	0.05
3	-2.14	2.13	2.65	-1.80	2.33	0.05
4	8.38	1.03	2.89	1.94	0.73	0.05
5	9.18	1.08	2.78	3.39	0.80	0.05
6	1.93	1.20	2.47	0.03	1.07	0.05
7	1.11	1.52	2.43	0.32	0.99	0.05
8	-2.28	1.60	2.57	0.65	0.80	0.05
10	4.71	1.24	3.04	2.92	0.82	0.06
11	4.41	0.83	2.46	0.31	0.95	0.05
12	5.01	0.87	2.76	1.75	0.56	0.05
13	6.15	0.94	2.69	1.48	0.96	0.05
15	1.27	0.55	2.77	0.77	0.84	0.05
16	3.67	0.82	2.26	1.53	1.12	0.04
17	1.90	0.95	2.30	0.28	1.18	0.04
18	3.06	1.26	2.35	0.81	1.08	0.04

19	5.84	0.74	2.74	2.19	0.84	0.05
20	-1.78	0.54	3.80	0.22	0.50	0.07
21	0.30	1.60	2.29	0.00	1.17	0.04
22	3.01	1.05	2.52	0.47	0.41	0.05
23	-4.37	0.55	3.71	0.34	0.31	0.07
25	5.39	1.08	2.42	0.57	1.60	0.05
27	10.00	0.90	2.70	0.78	1.05	0.05
28	9.34	1.22	2.75	3.16	1.29	0.05
29	-6.83	0.52	3.63	0.32	0.55	0.07
30	2.18	1.13	2.76	0.90	0.99	0.05
31	6.82	0.95	2.74	2.84	0.83	0.05
32	9.88	0.63	2.72	1.69	0.77	0.05
33	-3.33	1.88	2.65	-0.78	3.01	0.05
34	1.99	0.60	3.73	0.66	0.00	0.07
35	0.54	0.35	2.48	0.42	0.28	0.05
36	8.37	1.06	2.40	0.59	0.57	0.05
37	1.25	0.39	2.75	0.81	0.36	0.05
39	0.94	0.61	2.58	-0.06	1.12	0.05
40	1.96	0.47	2.77	1.28	0.40	0.05
42	1.63	0.79	2.13	0.03	0.74	0.04
43	-1.38	1.32	2.68	-0.11	0.37	0.05
44	-0.47	0.83	2.34	0.70	1.11	0.04
45	0.59	1.71	2.67	0.09	2.00	0.05
46	9.89	1.47	2.48	0.58	0.52	0.05
48	-1.87	1.51	3.67	0.20	0.37	0.07
49	4.12	0.68	2.22	1.13	1.07	0.04
50	13.09	2.65	2.84	1.76	1.13	0.05
51	2.65	1.26	2.16	0.88	0.93	0.04
52	1.51	0.93	2.79	-0.18	0.67	0.05
53	5.00	0.86	2.31	0.43	0.75	0.04
54	14.74	1.60	2.40			
55	0.28	0.49	1.98	1.08	0.68	0.04

$C_\alpha C_\beta$ 900 (numbering on C_α)

Residue#	ΔR, s^{-1} protonated	error	$R_{DD/DD}$, s^{-1} protonated	ΔR, s^{-1} deuterated	error	$R_{DD/DD}$, s^{-1} deuterated
2	4.56	1.29	2.60	0.91	1.22	0.05
3	-8.49	4.62	2.65	-1.97	1.58	0.05
4	14.45	1.81	2.89	1.42	1.20	0.05
5	15.05	2.09	2.78	4.47	0.52	0.05
6	4.16	1.99	2.47	-0.08	0.97	0.05
7	5.75	2.48	2.43	0.56	0.72	0.05

8	-6.13	1.15	2.57	0.01	0.86	0.05
10	7.34	1.23	3.04	2.13	0.60	0.06
11	8.18	2.00	2.46	0.67	0.58	0.05
12	9.75	1.19	2.76	3.96	0.57	0.05
13	6.59	1.64	2.69	3.52	2.84	0.05
15	0.94	1.18	2.77	1.28	0.54	0.05
16	5.28	2.75	2.26	4.12	0.77	0.04
17	3.21	1.09	2.30	-0.24	0.66	0.04
18	3.48	2.18	2.35	2.74	0.79	0.04
19	9.47	1.77	2.74	1.81	1.10	0.05
20	-6.21	0.73	3.80	-0.04	0.35	0.07
21	-5.15	2.93	2.29	0.70	1.09	0.04
22	-3.27	0.92	2.52	-0.91	0.66	0.05
23	-9.40	0.69	3.71	-0.94	0.58	0.07
25	9.66	1.69	2.42	2.55	0.75	0.05
27	13.42	1.09	2.70	2.19	0.67	0.05
28	12.78	1.41	2.75	3.24	0.62	0.05
29	-9.15	1.42	3.63	0.88	1.91	0.07
30	0.50	3.17	2.76	-1.09	2.00	0.05
31	9.70	1.60	2.74	1.57	1.15	0.05
32	14.70	1.90	2.72	2.62	0.57	0.05
33	-5.42	5.59	2.65	-2.70	1.99	0.05
34	5.66	1.63	3.73	-1.64	0.57	0.07
35	1.26	0.57	2.48	1.06	0.46	0.05
36	12.52	3.12	2.40	1.28	0.51	0.05
37	2.91	0.77	2.75	1.94	0.43	0.05
39	-4.05	2.09	2.58	-0.38	1.04	0.05
40	1.70	0.74	2.77	1.68	0.79	0.05
42	-0.87	2.77	2.13	1.70	1.15	0.04
43	0.01	1.68	2.68	1.27	0.92	0.05
44	-2.83	1.86	2.34	0.16	1.10	0.04
45	-0.48	3.93	2.67	-0.16	1.90	0.05
46	11.88	1.19	2.48	1.75	0.37	0.05
48	-8.25	0.93	3.67	-0.75	0.66	0.07
49	4.01	1.47	2.22	-0.16	0.84	0.04
50	16.43	1.81	2.84	1.63	0.60	0.05
51	5.44	2.06	2.16	1.67	0.73	0.04
52	1.02	2.01	2.79	-1.08	1.09	0.05
53	8.21	1.57	2.31	0.05	1.08	0.04
54	17.70	3.47	2.41			
55	3.63	1.17	1.98	0.03	2.76	0.04

J coupling constants (Hz) from $C_\alpha C_\beta$ data, 500MHz

	Protonated		Deuterated		Protonated		Deuterated	
Residue#	$^1J(C_\beta C_\gamma)$	error	$^1J(C_\beta C_\gamma)$	error	$^2J(C_a C_\gamma)$	error	$^2J(C_a C_\gamma)$	error
2	32.60	0.33	33.24	0.33	1.48	0.33	1.36	0.33
4	37.14	0.97	32.97	0.25	-3.52	0.97	0.96	0.25
5	32.54	0.35	33.08	0.32	0.61	0.35	0.33	0.32
6	34.85	0.44	35.55	0.36	0.62	0.44	0.35	0.36
7	34.90	0.45	34.82	0.47	0.39	0.45	-0.69	0.47
10	33.56	0.58	33.06	0.46	-0.38	0.58	0.32	0.46
11	39.11	0.39	38.57	0.48	0.28	0.39	1.41	0.48
12	33.88	0.37	34.08	0.25	-0.38	0.37	0.75	0.25
13	34.73	0.55	35.58	0.42	-2.23	0.55	-1.86	0.42
15	32.95	0.24	33.33	0.33	0.61	0.24	0.69	0.33
16	36.21	0.35	36.93	0.52	1.70	0.35	1.74	0.52
17	39.37	0.65	38.04	0.57	-1.78	0.65	0.97	0.57
18	37.72	0.63	39.11	0.55	-0.14	0.63	0.69	0.55
19	35.26	0.57	33.59	0.44	-2.15	0.57	-0.07	0.44
21	34.23	0.63	35.05	0.46	0.52	0.63	0.78	0.46
25	33.73	0.46	35.37	0.77	6.99	0.46	7.10	0.77
27	32.02	0.38	33.86	0.44	3.88	0.38	2.79	0.44
28	35.55	0.90	32.51	0.58	-1.77	0.90	1.69	0.58
31	31.98	0.31	33.23	0.43	1.76	0.31	0.38	0.43
32	29.55	0.26	32.75	0.30	4.83	0.26	2.62	0.30
39	35.21	0.20	35.07	0.28	0.46	0.20	0.08	0.28
42	33.21	0.51	35.60	0.60	1.89	0.51	-1.21	0.60
44	36.01	0.42	37.80	0.54	2.71	0.42	1.59	0.54
49	37.35	0.39	37.05	0.50	0.83	0.39	1.17	0.50
50	39.30	1.35	34.50	0.62	-5.64	1.35	-1.17	0.62
51	37.95	0.54	39.07	0.41	2.72	0.54	3.32	0.41
53	34.06	0.33	38.73	0.40	-1.21	0.33	3.36	0.40
54	34.87	0.79	38.10	0.40	0.63	0.79	4.70	0.40
55	35.05	0.40	35.46	0.44	1.51	0.40	0.39	0.44

J coupling constants (Hz) from $C_\alpha C_\beta$ data, 900MHz

	Protonated		Deuterated		Protonated		Deuterated	
Residue#	$^1J(C_\beta C_\gamma)$	error	$^1J(C_\beta C_\gamma)$	error	$^2J(C_a C_\gamma)$	error	$^2J(C_a C_\gamma)$	error
2	32.76	0.41	33.02	0.40	0.53	0.41	1.10	0.40
4	34.70	0.63	33.22	0.41	-0.36	0.63	0.98	0.41
5	32.25	0.75	33.82	0.12	0.76	0.75	0.78	0.12
6	35.14	0.66	35.38	0.36	0.28	0.66	0.79	0.36

7	35.05	0.74	35.17	0.47	0.79	0.74	0.86	0.47
10	32.73	0.52	32.68	0.12	0.70	0.52	0.03	0.12
11	37.75	0.58	38.76	0.18	1.76	0.58	2.00	0.18
12	33.10	0.37	33.77	0.16	0.68	0.37	1.58	0.16
13	33.11	0.65	39.46	1.31	-0.19	0.65	-0.50	1.31
15	33.08	0.36	33.62	0.11	0.01	0.36	-0.23	0.11
16	35.25	0.88	38.09	0.28	1.50	0.88	2.14	0.28
17	37.77	0.39	38.55	0.25	0.72	0.39	1.97	0.25
18	35.86	0.57	40.18	0.31	0.21	0.57	2.16	0.31
19	33.28	0.65	33.58	0.39	-0.14	0.65	0.33	0.39
21	34.64	0.94	34.67	0.31	-0.57	0.94	-0.69	0.31
25	37.16	0.51	37.36	0.15	4.71	0.51	4.51	0.15
27	32.48	0.48	33.93	0.13	3.41	0.48	1.88	0.13
28	33.45	0.53	32.08	0.18	0.35	0.53	0.71	0.18
31	31.74	0.72	31.86	0.39	1.06	0.72	0.39	0.39
32	29.18	0.56	33.79	0.11	5.47	0.56	1.43	0.11
39	35.45	0.66	35.19	0.29	0.20	0.66	0.55	0.29
42	35.07	0.82	35.10	0.30	-0.34	0.82	0.37	0.30
44	39.15	0.76	35.48	0.42	0.36	0.76	0.57	0.42
49	34.92	0.47	37.23	0.28	1.56	0.47	3.03	0.28
50	33.99	0.89	33.65	0.18	-0.92	0.89	-0.46	0.18
51	37.56	0.51	39.15	0.17	2.68	0.51	3.95	0.17
53	33.05	0.50	38.78	0.37	0.03	0.50	4.26	0.37
54	32.86	0.76			1.77	0.76		
55	31.71	0.40	43.00	3.06	0.87	0.40	1.90	3.06

SI6. Correlation plots of Δ*R* rates for the protonated versus deuterated proteins at 500 MHz .

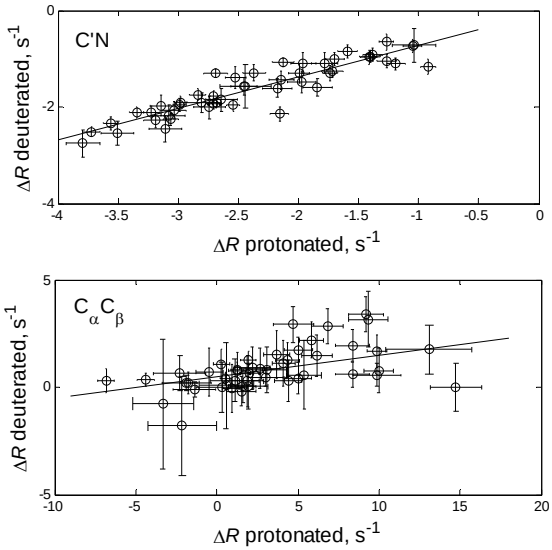

Figure SI3. Correlation plots of Δ*R* for protonated and deuterated GB3 at 500 MHz field strength and 25 ºC for the C'N (top) and $C_\alpha C_\beta$ experiment (bottom).

SI7. Correlation plot of differences in J-coupling constants between protonated and deuterated proteins at two fields.

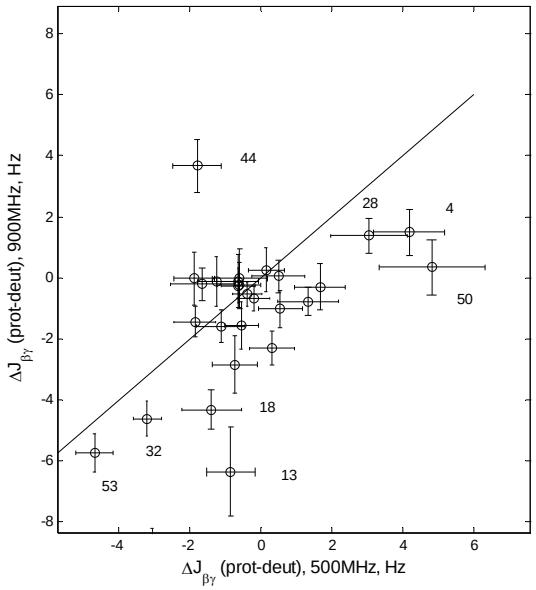

Figure SI4. Correlation plot of the difference in $J_{\beta\gamma}$ between protonated and deuterated proteins, $\Delta J_{\beta\gamma}$, at 500 and 900 MHz. The overall offset suggests residual systematic errors between the two spectrometers.

SI8. Fits for the $^{13}C_\alpha$—$^{13}C_\beta$ experiment at 500 MHz for the protonated data with J-coupling constants values averaged between protonated and deuterated proteins.

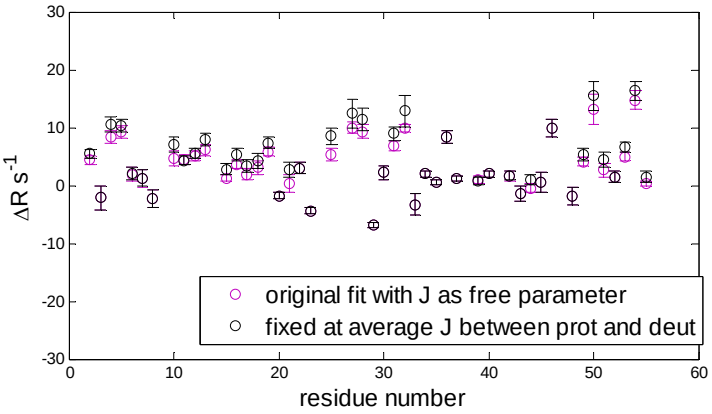

Figure SI5. Δ*R versus* residue number for 500 MHz data on protonated GB3 at 25 °C with $J_{\beta\gamma}$- and $J_{\alpha\gamma}$-coupling constants as free fitting parameters according to Eq.12 (purple) and with the values fixed at the weighted average values originating from the independent fits for the protonated and deuterated proteins (black). The average J-coupling constants were calculated using the values listed in SI5.

Loth K, Pelupessy P, Bodenhausen G (2005) Chemical shift anisotropy tensors of carbonyl, nitrogen, and amide proton nuclei in proteins through cross-correlated relaxation in NMR spectroscopy. J Am Chem Soc 127:6062-6068