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Supporting Information, Table 1. ^1H - ^{15}N dipolar couplings in ubiquitin at 304 °K, using 5% w/v bicelles (1:30:10 CTAB:DMPC:DHPC) in 93% H₂O, 7% D₂O.

RES#	D ^{NH} (Hz) ^a	RES#	D ^{NH} (Hz) ^a
2	13.20	39	-14.97
3	-8.78	40	0.42
4	-6.81	41	-24.12
5	-3.19	42	-9.53
6	-0.92	43	-10.97
7	4.62	44	-1.69
8	-13.62	45	4.15
10	-5.57	46	0.97
11	14.87	47	-1.79
12	-0.59	48	20.19
13	1.42	49	3.89
14	-3.66	50	-1.84
15	-11.84	51	13.56
16	0.30	52	22.73
17	-2.18	54	4.17
18	17.18	55	-0.12
20	11.28	56	-16.10
21	-3.94	57	-22.51
22	-20.53	58	-10.29
23	-1.11	59	-9.23
25	3.23	60	-22.90
26	-0.39	61	-16.95
27	-2.61	62	-15.85
28	3.01	63	12.39
29	-0.82	64	-19.87
30	-1.01	65	-18.07
31	-1.83	66	-13.20
32	3.81	67	-5.26
33	-0.72	68	-1.75
34	-2.31	69	-4.42
35	10.14	70	-12.02
36	20.59	71	-19.48

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Supporting Information, Table 2. ^1H , ^{15}N and ^{13}C shifts^a in human ubiquitin at 290 °K, in the presence of 5% w/v bicelles (1:30:10 CTAB:DMPC:DHPC) in 93% H₂O, 7% D₂O.

RES#	^1H (ppm) ^b	^{15}N (ppm) ^c	$^{13}\text{C}'$ (ppm) ^d
1	-----	-----	170.382
2	8.986	122.614	175.955
3	8.301	114.969	172.275
4	8.629	118.384	175.092
5	9.307	121.027	174.754
6	8.957	127.725	177.050
7	8.772	115.416	176.924
8	9.203	121.227	178.920
9	7.648	119.008	175.482
10	7.834	109.207	173.897
11	7.288	121.988	175.691
12	8.689	120.564	174.303
13	9.576	127.558	175.091
14	8.778	121.282	173.662
15	8.769	124.890	174.475
16	8.140	122.316	175.695
17	8.963	117.471	173.958
18	8.646	119.044	-----
19	-----	-----	175.245
20	7.043	103.283	174.573
21	8.078	123.833	176.227
22	7.927	108.983	176.703
23	8.529	120.996	-----
24	-----	-----	179.016
25	7.952	121.362	178.276
26	8.132	122.089	177.892
27	8.570	118.784	180.493
28	7.971	123.324	180.217
29	7.864	120.193	180.339
30	8.308	121.375	178.166
31	8.555	123.398	178.838
32	8.043	119.646	177.265
33	7.431	115.352	177.791
34	8.732	114.169	177.881
35	8.508	108.895	173.856
36	6.150	120.324	-----
37	-----	-----	-----
38	-----	-----	178.299
39	8.551	113.589	176.995

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40	7.838	116.785	175.403
41	7.515	118.092	176.031
42	8.501	123.087	173.763
43	8.846	124.224	175.223
44	9.135	122.038	175.755
45	8.856	124.639	174.628
46	9.025	119.864	177.311
47	8.142	102.426	173.613
48	7.971	122.026	174.552
49	8.681	122.946	175.517
50	8.576	125.522	176.602
51	8.442	122.952	175.425
52	8.200	120.159	-----
53	-----	-----	174.698
54	7.487	119.384	175.249
55	8.841	108.732	176.468
56	8.159	117.769	180.803
57	8.527	113.478	178.309
58	7.957	124.415	177.403
59	7.262	115.686	174.624
60	8.163	116.006	174.211
61	7.276	118.921	174.452
62	7.647	124.725	175.685
63	8.528	120.444	175.782
64	9.338	114.538	175.200
65	7.697	115.019	171.906
66	8.750	117.310	173.610
67	9.435	127.506	175.146
68	9.231	119.252	173.837
69	8.279	123.626	175.412
70	9.183	126.116	174.105
71	8.172	122.853	177.856
72	8.607	123.614	175.342
73	8.414	124.578	177.442
74	8.498	122.056	176.921
75	8.534	111.116	173.604
76	7.988	115.051	-----

^a¹H and ¹³C shifts are relative to TSP and ¹⁵N shifts are indirectly referenced relative to liquid ammonia.

^bMeasurement error of ¹H chemical shifts, estimated using the differences in the ¹H chemical shift of the doublet components, is 0.05 ppb.

^cMeasurement error of ¹⁵N chemical shifts, estimated using the differences in the ¹H chemical shift of the doublet components (see text), is 0.4 ppb.

^dMeasurement error of ¹³C' chemical shifts, estimated using the differences in the ¹³C chemical shifts of the doublet components, is 0.16 ppb.

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Supporting Information, Table 3. ^1H , ^{15}N and ^{13}C chemical shift differences between aligned and isotropic states (corrected for the temperature dependence) in human ubiquitin at 290, 297 and 304 °K, in the presence of 5% w/v bicelles (1:30:10 CTAB:DMPC:DHPC) in 93% H_2O , 7% D_2O .

Res#	$\Delta\delta_{\text{H}}^{\text{a}}$ (ppb)	$\Delta\delta_{\text{N}}^{\text{b}}$ (ppb)	$\Delta\delta_{\text{C}}^{\text{c}}$ (ppb)
1	---	---	9.1
2	-4.7	112.1	63.8
3	-0.1	-87.6	-15.0
4	2.6	15.9	123.3
5	-4.7	2.0	-53.9
6	1.6	3.7	104.4
7	-8.2	42.1	19.2
8	3.0	-114.0	---
9	---	---	76.9
10	-4.1	-4.4	-14.6
11	-2.0	111.9	96.2
12	-2.2	-31.7	-107.2
13	6.1	-16.5	75.5
14	-0.8	-13.7	-98.3
15	8.0	-96.2	-4.2
16	1.6	59.3	-16.4
17	0.4	-22.6	45.4
18	-8.2	110.7	---
19	---	---	-70.2
20	0.6	68.0	56.6
21	-2.6	38.4	49.2
22	-0.1	-77.1	-4.7
23	-0.1	-11.3	---
24	---	---	100.2
25	-3.1	-20.0	-122.4
26	3.9	-4.9	126.2
27	-4.6	-11.2	-96.1
28	2.7	23.3	63.8
29	-3.3	-25.4	-40.3
30	1.0	-0.3	-2.0
31	1.4	12.0	60.3
32	-2.5	-1.8	-100.0
33	1.4	-2.8	63.6
34	-0.7	3.6	7.2
35	-0.3	111.2	39.6
36	-5.2	118.6	---

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37	---	---	---
38	---	---	62.7
39	0.9	-55.4	-75.0
40	1.6	-48.4	45.0
41	3.0	-110.8	-103.7
42	6.4	18.0	88.5
43	1.0	-33.1	-82.8
44	4.8	-16.8	103.9
45	-4.2	26.5	-100.9
46	4.1	-15.4	118.6
47	-2.0	-1.4	-41.4
48	-3.9	68.0	8.6
49	0.2	-41.4	-50.5
50	3.5	-18.7	-40.6
51	-1.1	62.6	-32.9
52	-6.0	59.8	---
53	---	---	1.0
54	-4.2	85.8	112.2
55	-6.2	-15.3	-69.8
56	6.4	-103.7	-11.4
57	2.8	-95.0	31.8
58	2.4	-69.1	-97.5
59	4.0	-85.0	58.4
60	5.1	-109.9	-43.0
61	6.9	-49.0	31.8
62	3.1	-16.8	69.5
63	-5.1	21.7	-56.8
64	8.1	-115.2	6.5
65	2.0	-64.6	51.3
66	2.5	-23.3	32.0
67	-1.2	11.3	104.1
68	-4.2	-33.9	16.0
69	-1.5	-20.9	77.9
70	1.9	-115.3	30.5
71	6.5	-114.4	---

^a propagated measurement error in $\Delta\delta_{\text{H}}$ is 0.14 ppb.

^b propagated measurement error in $\Delta\delta_{\text{N}}$ is 1.4 ppb.

^c propagated measurement error in $\Delta\delta_{\text{C}}$ is 0.3 ppb.

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Supporting Information, Table 4. ^1H , ^{15}N and ^{13}C temperature factors in human ubiquitin, in the presence of 5% w/v bicelles (1:30:10 CTAB:DMPC:DHPC) in 93% H_2O , 7% D_2O .

RES#	$k^T(^{13}\text{C})^a$ (ppb/°C)	$k^T(^{15}\text{N})^b$ (ppb/°C)	$k^T(^1\text{H})^c$ (ppb/°C)	$\beta(^1\text{H})^d$ (ppb/°C ²)
1	16.83	----	----	----
2	6.53	21.34	4.43	-0.026
3	15.09	27.79	10.77	0.002
4	9.51	14.25	7.51	0.014
5	11.90	18.88	8.21	-0.009
6	8.62	12.20	10.34	-0.020
7	7.04	-3.18	6.28	0.023
8	0.34	-2.34	0.59	0.019
9	8.19	4.83	7.83	0.007
10	12.76	0.28	7.82	0.017
11	12.89	-1.62	8.20	0.017
12	6.67	-2.50	3.26	-0.007
13	13.22	-3.08	7.42	-0.010
14	10.69	7.48	3.19	-0.018
15	11.69	13.53	7.33	-0.002
16	15.30	11.32	7.17	-0.026
17	14.12	11.41	6.96	-0.014
18	----	28.75	11.84	0.000
19	13.96	----	----	----
20	9.15	20.42	8.24	0.017
21	13.67	-1.53	7.15	0.006
22	11.84	0.32	5.46	0.021
23	----	14.80	7.85	0.001
24	7.69	----	----	----
25	9.36	-0.29	7.27	0.000
26	12.28	1.99	7.26	0.007
27	9.07	2.78	7.60	0.001
28	11.81	1.50	9.32	0.003
29	3.27	1.63	8.78	0.014
30	12.25	-4.41	7.11	0.013
31	6.72	7.69	8.58	-0.012
32	9.88	3.66	5.65	0.022
33	10.44	4.77	10.24	0.018
34	10.38	15.89	8.21	-0.020
35	12.01	-0.66	6.85	-0.004
36	----	-5.75	9.93	0.021
37	----	----	----	----
38	6.13	----	----	----
39	11.06	-2.75	6.48	0.024

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40	5.12	8.06	7.58	0.008
41	15.52	-6.99	5.93	0.025
42	12.35	-6.38	9.50	-0.031
43	12.25	3.82	5.87	-0.005
44	7.08	26.42	9.29	-0.009
45	7.20	21.81	9.00	-0.011
46	7.58	-17.33	4.68	-0.015
47	11.58	2.01	3.30	-0.009
48	14.09	0.02	9.64	0.014
49	10.18	2.96	4.51	-0.018
50	9.90	8.87	7.89	-0.033
51	11.96	8.07	4.68	-0.005
52	----	3.28	4.52	0.002
53	10.23	----	----	----
54	13.70	-4.24	7.96	0.019
55	11.43	12.01	8.77	0.008
56	6.59	14.88	8.98	0.011
57	4.94	2.89	5.43	0.007
58	7.25	3.12	7.52	-0.004
59	10.30	1.59	8.98	0.013
60	10.11	3.98	8.16	0.000
61	13.38	1.48	7.18	-0.011
62	15.40	6.22	5.78	-0.003
63	3.79	6.59	4.00	-0.018
64	11.65	7.54	5.70	-0.010
65	13.59	-2.51	7.60	0.009
66	12.49	7.23	4.86	-0.016
67	11.26	9.09	7.50	-0.009
68	11.66	26.11	9.02	-0.031
69	1.12	14.82	9.66	0.017
70	4.67	30.74	7.97	-0.054
71	5.39	9.32	2.03	-0.017
72	4.30	3.72	7.45	-0.002
73	5.38	-14.33	1.82	-0.017
74	1.70	-13.35	2.26	-0.008
75	7.79	-10.42	2.42	0.003
76	----	5.79	3.59	0.004

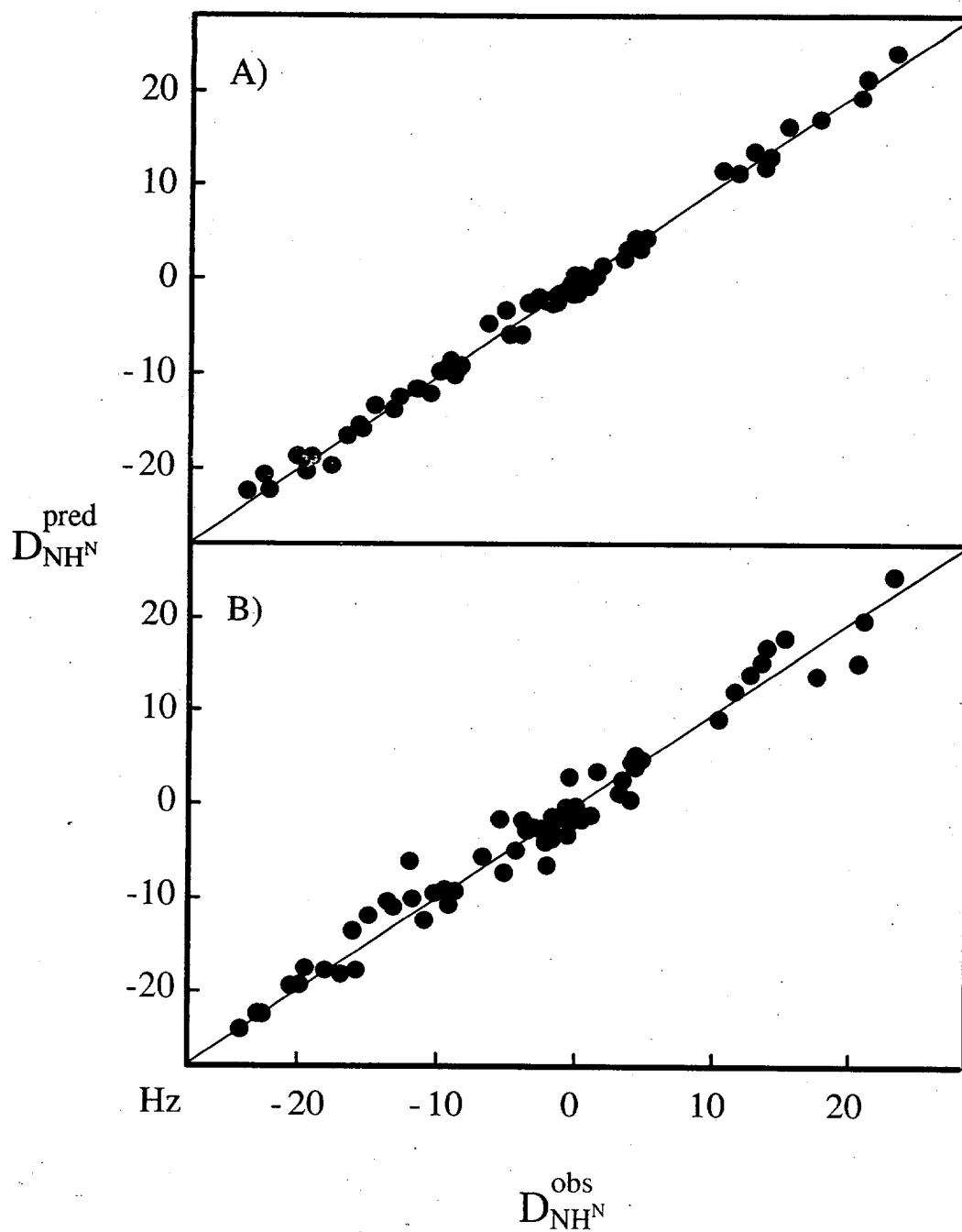
^a propagated measurement error in k^T (^{13}C) is 0.03 ppb/°C.

^b propagated measurement error in k^T (^{15}N) is 0.08 ppb/°C.

^c propagated measurement error in k^T (^1H) is 0.01 ppb/°C.

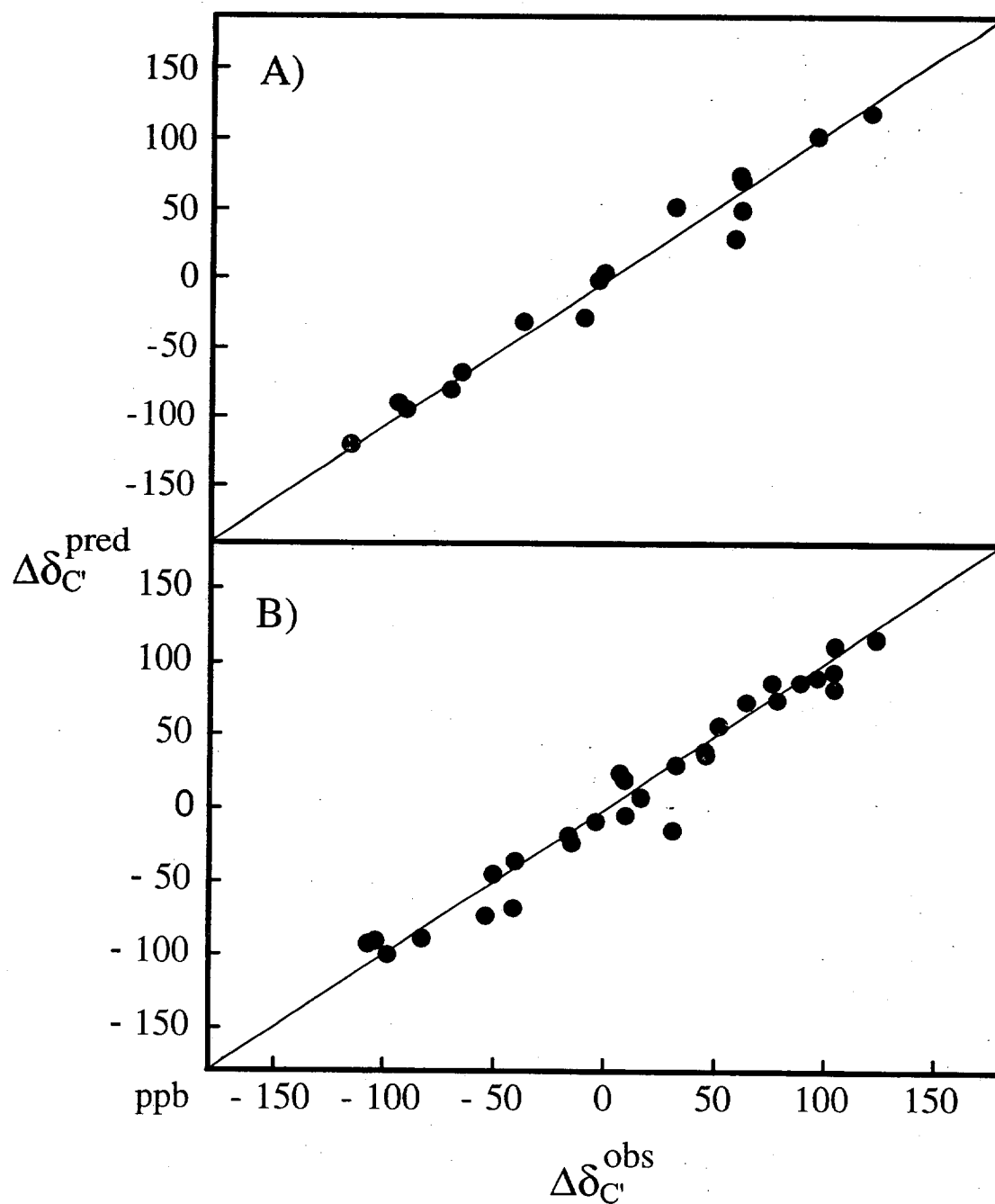
^d from linear system [4] (see text).

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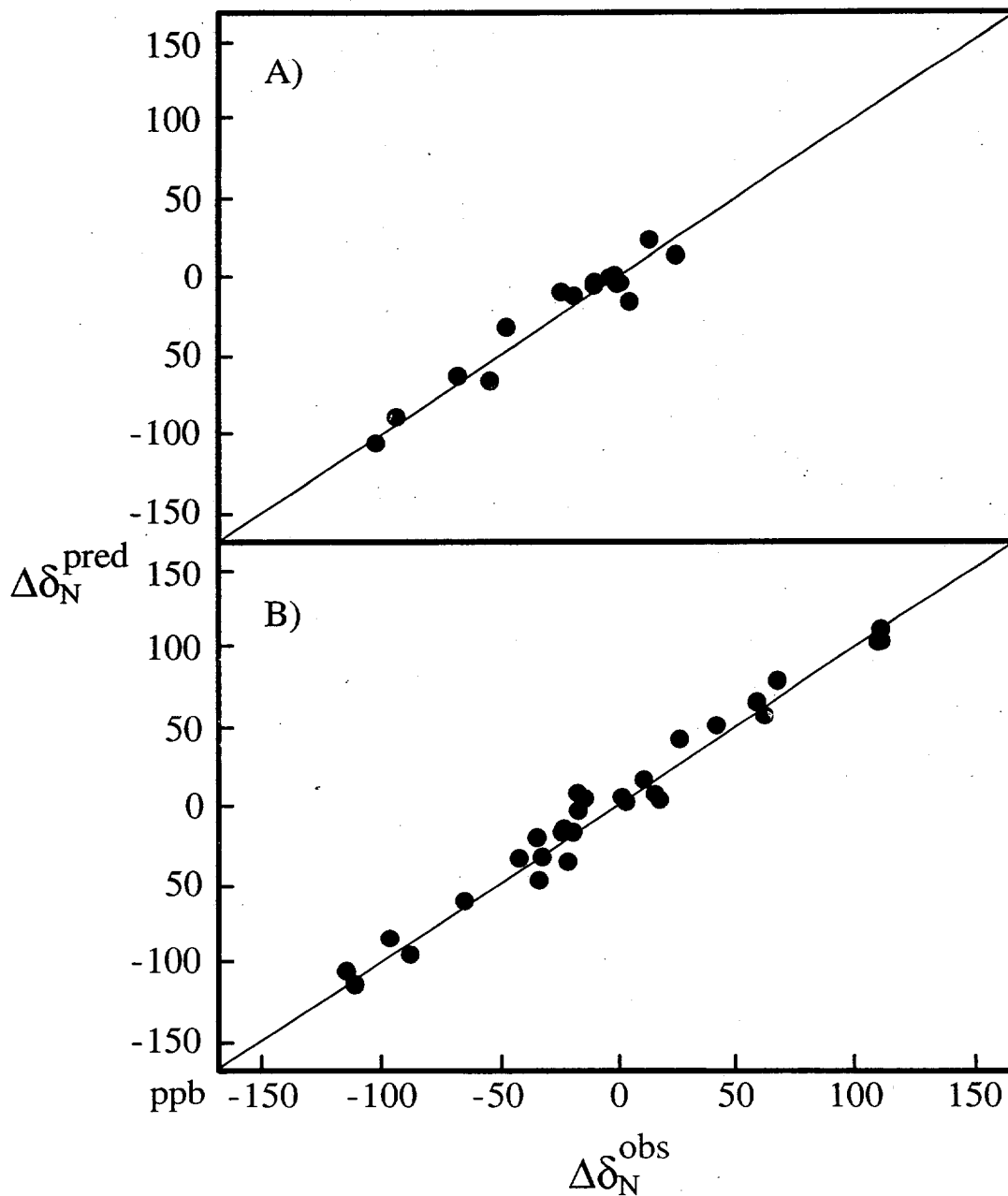
Supporting Information, Figure 1. Correlations between calculated and experimentally obtained values of $^1\text{H}^{\text{N}}\text{-}^{15}\text{N}$ dipolar couplings for (A) the NMR structure, and (B) the crystal structure of ubiquitin.

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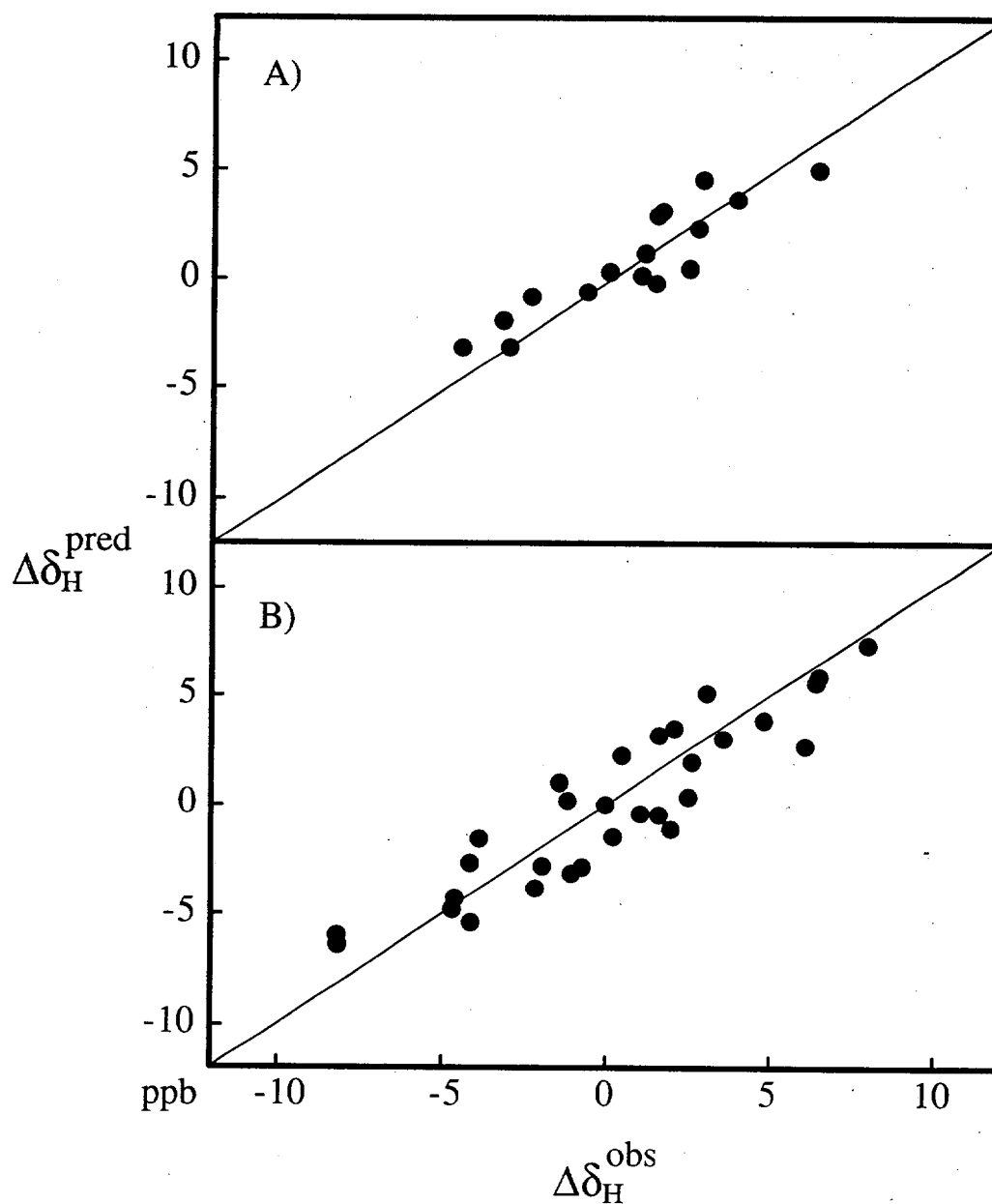
Supporting Information, Figure 2. Correlations between best-fitted and experimentally observed $\Delta\delta_{C'}$ values for (A) all helical and (B) all β -sheet residues in the crystal structure of ubiquitin.

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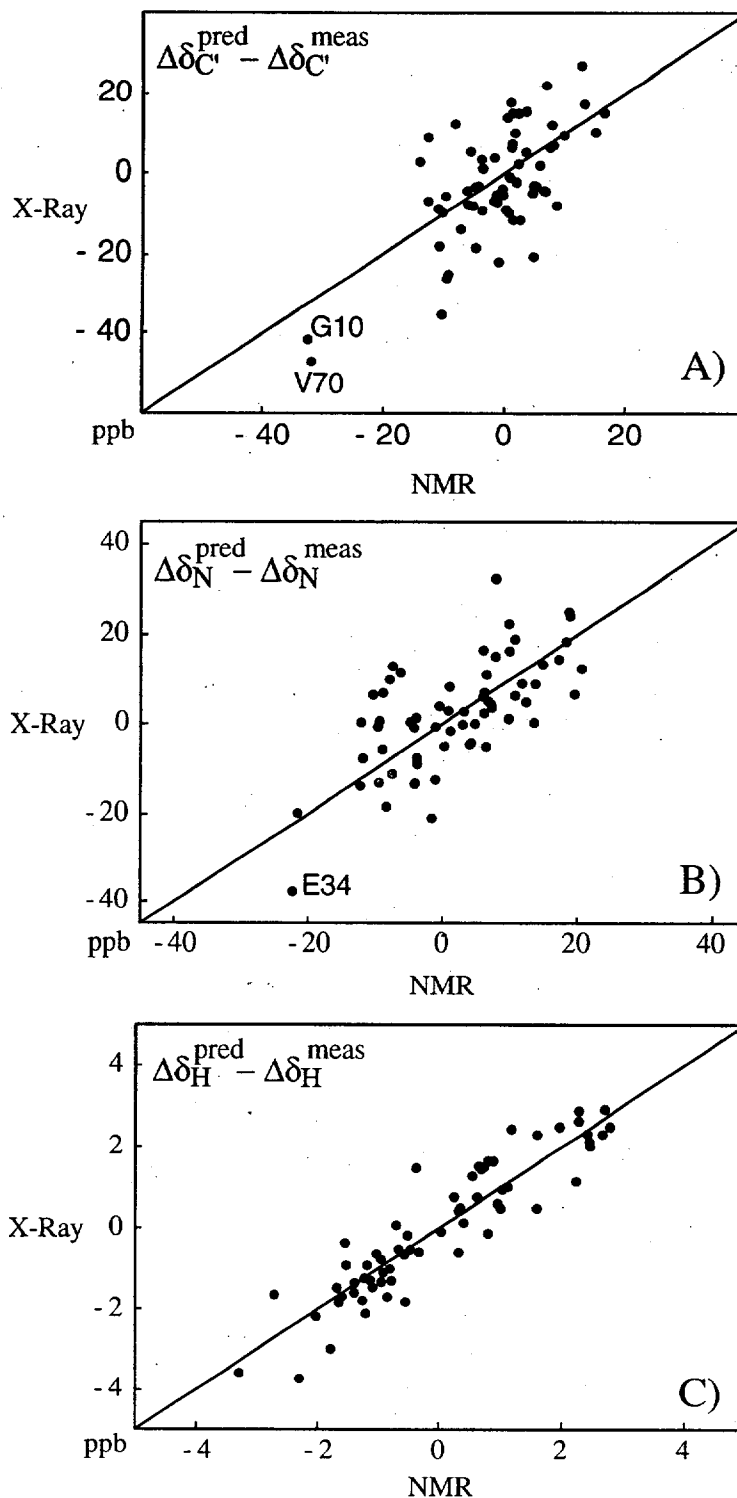
Supporting Information, Figure 3. Correlations between best-fitted $\Delta\delta_N$ and experimentally observed values for (A) helical and (B) β -sheet residues in the NMR structure of ubiquitin.

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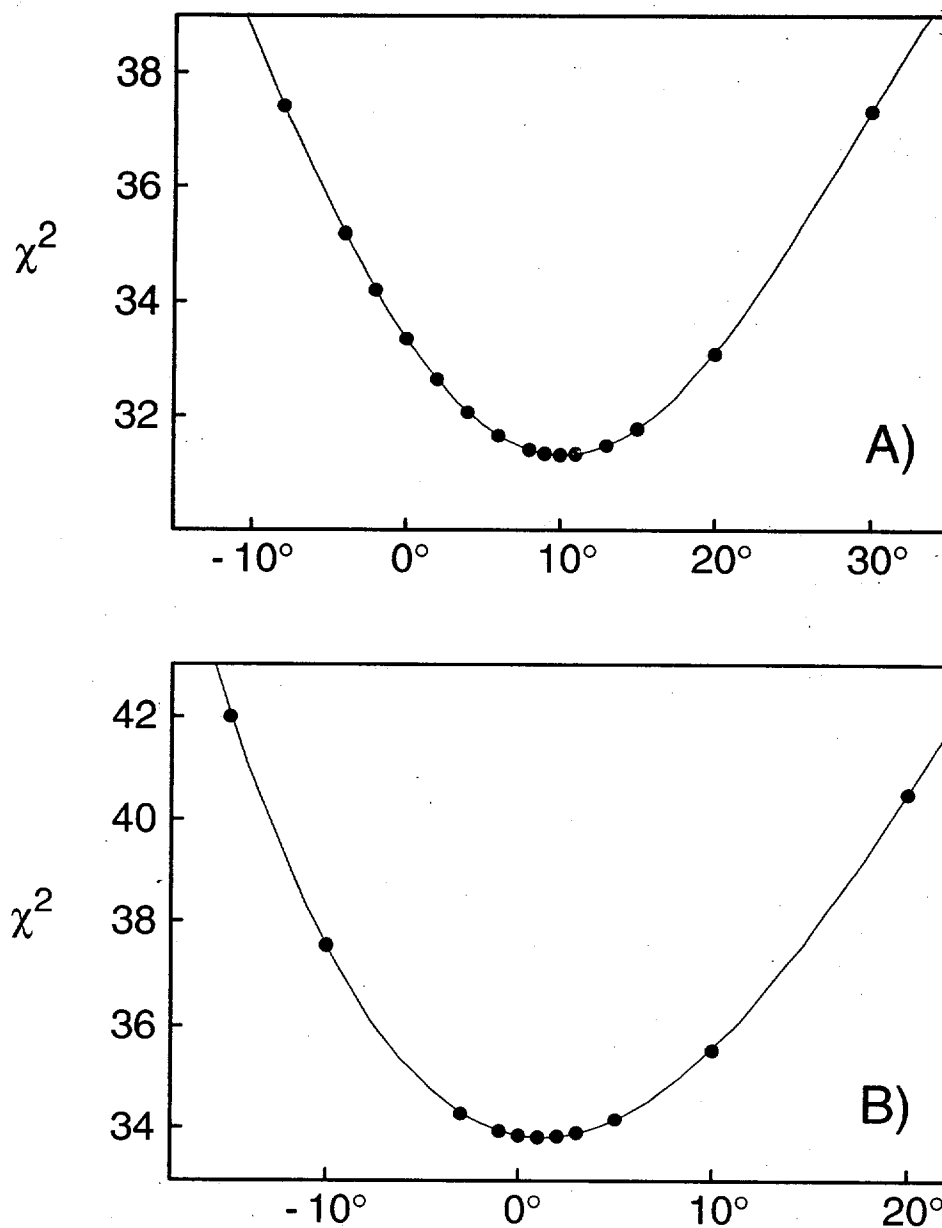
Supporting Information, Figure 4. Correlations between best-fitted and experimentally observed $\Delta\delta_H$ values in (A) helical and (B) β -sheet for the ubiquitin NMR structure. Linear regression yields correlation coefficients, R_p , of 93.2% (A) and 93.8%(B).

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Supporting Information, Figure 5. Correlations between $(\Delta\delta_i^{\text{meas}} - \Delta\delta_i^{\text{pred, NMR}})$ and $(\Delta\delta_i^{\text{meas}} - \Delta\delta_i^{\text{pred, Xray}})$ for (A) $^{13}\text{C}'$ (rmsd = 10.9 ppb), (B) ^{15}N (rmsd = 9.2 ppb), and (C) ^1H (rmsd = 0.7 ppb) in human ubiquitin.

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Supporting Information, Figure 6. Plots of the χ^2 dependence of the fit between experimental and best-fitted $\Delta\delta(^{15}\text{N})$ values on the clockwise rotation angle of σ_{22} and σ_{33} around the σ_{11} vector (Figure 7, main text) for (A) all, and (B) β -sheet residues.