Supporting information:
Rieko Ishima, Aneta, P. Petkova, John M. Louis, and Dennis A. Torchia
"Comparison of methyl rotation axis order parameters determined by..."

Supplementary Table 1. Differences of S_{axis}^2 values, $(\chi_s^2)^a$, derived from model-free analyses of ^2H and ^{13}C relaxation data.

Amino acid	^2H S_{axis}^2 value	^2H S_{axis}^2 error	^{13}C S_{axis}^2 value	^{13}C S_{axis}^2 error	$(\chi_s^2)^a$
I3 γ2[b]	0.4852	0.0332	0.7642	0.0416	27.4785
L5 δ1[b]			0.8567	0.0788	
L5 δ2	0.8721	0.0185	0.8417	0.0181	1.3796
L10 δ1	0.4600	0.0065	0.5402	0.0136	28.3088
L10δ2	0.5365	0.0081	0.5490	0.0340	0.1279
V11 γ1	0.8418	0.0240	0.8082	0.0239	0.9841
V11γ2	0.6955	0.0134	0.7734	0.0268	6.7592
I13 γ2	1.0165	0.0234	0.9708	0.0302	1.4309
I15 γ2	0.8961	0.0167	0.9302	0.0151	2.2940
L19 δ1	0.2273	0.0043	0.2813	0.0096	26.3534
L19 δ2	0.2621	0.0046	0.3031	0.0168	5.5405
A22 β	0.9053	0.0578	1.1622	0.0323	15.0538
L23 δ1	0.4158	0.0162	0.5324	0.0174	24.0544
L23 δ2	0.4449	0.0130	0.5171	0.0219	8.0369
L24 δ1	0.8621	0.0192	0.9184	0.0208	3.9558
L24 δ2	0.9869	0.0201	0.9403	0.0333	1.4354
A28 β	0.8834	0.1403	1.0399	0.0246	1.2072
V32 γ1	0.7298	0.0142	0.7968	0.0140	11.2891
V32 γ2	0.7667	0.0176	0.8210	0.0165	5.0660
I33 γ2	0.6905	0.0166	0.7676	0.0155	11.5244
L38 δ1	0.5577	0.0177	0.7080	0.0264	22.3609
L38 δ2	0.5133	0.0080	0.5354	0.0194	1.1091
I47 γ2	0.5348	0.0087	0.5631	0.0238	1.2472
I50 γ2	0.7979	0.0665	0.8190	0.0303	0.0834
I54 γ2	0.8474	0.0336	0.9215	0.0211	3.4881
V56 γ1	1.0203	0.0228	1.0079	0.0115	0.2358
V56 γ2	0.8765	0.0200	0.8623	0.0144	0.3320
I62 δ2	0.7874	0.0162	0.7944	0.0207	0.0709
I63 δ2	0.6307	0.0109	0.6701	0.0267	1.8665
I64 δ2	0.3717	0.0101	0.4226	0.0181	6.0305
I66 δ2	0.9497	0.0204	0.9720	0.0153	0.7648

A67 β	1.1422	0.0307	1.1049	0.0135	1.2370
A71 β	1.1126	0.0316	1.1546	0.0146	1.4558
I72 γ2	0.4729	0.0064	0.5016	0.0117	4.6314
V75 γ1	0.4864	0.0086	0.5335	0.0183	5.4260
V75 γ2	0.4995	0.0099	0.5551	0.0193	6.5704
L76 δ1	0.9043	0.0246	0.7059	0.0344	22.0085
L76 δ2	0.8531	0.0188	0.8561	0.0217	0.0109
V77 γ1	0.7110	0.0227	0.7751	0.0316	2.7141
V77 γ2	0.7938	0.0172	0.8330	0.0316	1.1871
V82 γ1	0.1548	0.0046	0.2616	0.0141	51.8536
V82 γ2	0.1627	0.0067	0.2120	0.0243	3.8253
I84 γ1	0.8287	0.0158	0.8711	0.0170	3.3376
I85 γ2	0.8903	0.0378	0.9883	0.0359	3.5339
L89 δ1	0.7626	0.0178	0.8597	0.0222	11.6446
L89 δ2	0.7324	0.0134	0.7198	0.0174	0.3292
L90 δ1	0.9749	0.0447	1.1579	0.0253	12.6940
L90 δ2	0.8334	0.0360	0.9182	0.0259	3.6562
I93 γ2	0.9029	0.0190	0.9187	0.0189	0.3476
A95 β	0.9179	0.0198	0.9585	0.0297	1.2937
L97 δ1[b]	0.1883	0.0465	0.4350	0.0334	18.5675
L97 δ2[b]	0.1883	0.0500	0.4350	0.0369	15.7605

[a]Defined as, $\chi_s^2 = ({}^2H\ S_{axis}{}^2 - {}^{13}C\ S_{axis}{}^2)^2/\{({}^2H\ \Delta S_{axis}{}^2)^2+({}^{13}C\ \Delta S_{axis}{}^2)^2\}$ where $\Delta S_{axis}{}^2$ is the error in $S_{axis}{}^2$. [b]Chemical exchange contributes significantly to the $^{13}C\ R_2$.

Supplementary Table 2. $^{13}C\ T_1(R_1^{-1})$ and $T_2(R_2^{-1})$[a] data.

A.A.	T_1 (s)	T_{1error} (s)	T_2 (s)	T_{2error} (s)
I3 γ2	1.4091	0.0712	0.1653	0.0089
L5 δ1	0.3993	0.0269	0.1146	0.0074
L5 δ2	1.1300	0.0238	0.1476	0.0029
L10 δ1	1.1284	0.0163	0.2131	0.0052
L10 δ2	1.0871	0.0690	0.2088	0.0101
V11 γ1	0.5337	0.0158	0.1308	0.0029
V11 γ2	0.7956	0.0238	0.1490	0.0041
I13 γ2	1.7154	0.0581	0.1362	0.0039
I15 γ2	1.8722	0.0630	0.1427	0.0023
L19 δ1	1.2062	0.0333	0.3520	0.0085
L19 δ2	1.4083	0.0238	0.3495	0.0132
A22 β	0.3788	0.0111	0.0908	0.0018

L23 δ1	1.1154	0.0204	0.2151	0.0057
L23 δ2	0.9858	0.0358	0.2140	0.0065
L24 δ1	1.0392	0.0382	0.1353	0.0026
L24 δ2	1.7241	0.0803	0.1403	0.0045
A28 β	0.3461	0.0144	0.0954	0.0009
V32 γ1	0.8591	0.0266	0.1477	0.0021
V32 γ2	0.8844	0.0107	0.1449	0.0026
I33 γ2	0.6599	0.0151	0.1437	0.0023
L38 δ1	0.8742	0.0177	0.1630	0.0050
L38 δ2	0.9488	0.0295	0.2065	0.0064
I47 γ2	1.0397	0.0167	0.2027	0.0074
I50 γ2	0.4516	0.0058	0.1232	0.0036
I54 γ2	1.0874	0.0456	0.1358	0.0027
V56 γ1	2.0238	0.0655	0.1333	0.0014
V56 γ2	0.9834	0.0159	0.1416	0.0022
I62 γ2	1.0003	0.0198	0.1521	0.0035
I63 γ2	1.3506	0.0170	0.1841	0.0067
I64 γ2	1.0404	0.0219	0.2525	0.0091
I66 γ2	1.8639	0.0559	0.1370	0.0019
A67 β	1.6887	0.0209	0.1208	0.0014
A71 β	0.8781	0.0221	0.1085	0.0011
I72 γ2	1.3395	0.0284	0.2340	0.0048
V75 γ1	1.0086	0.0133	0.2101	0.0061
V75 γ2	0.6237	0.0181	0.1790	0.0043
L76 δ1	0.6086	0.0109	0.1498	0.0054
L76 δ2	1.0448	0.0381	0.1438	0.0033
V77 γ1	0.4107	0.0185	0.1238	0.0029
V77 γ2	0.8486	0.0282	0.1421	0.0043
V82 γ1	0.7733	0.0242	0.3113	0.0093
V82 γ2	0.6338	0.0261	0.3139	0.0164
I84 γ2	3.4306	0.0907	0.1578	0.0032
I85 γ2	1.7148	0.1073	0.1340	0.0048
L89 δ1	0.6794	0.0292	0.1325	0.0025
L89 δ2	1.5223	0.0388	0.1759	0.0040
L90 δ1	1.0947	0.0339	0.1112	0.0022
L90 δ2	1.6154	0.0436	0.1425	0.0041
I93 γ2	0.8388	0.0285	0.1308	0.0024
A95 β	1.3195	0.0334	0.1343	0.0039
L97 δ1	1.2690	0.0181	0.2592	0.0180
L97 δ2	1.2690	0.0181	0.2592	0.0180

[a]Recorded in a 2kHz spin lock field.

Supplementary Table 3. ^{2}H $T_1(R_1^{-1})$ and $T_{1\rho}(R_2^{-1})$[a] data.

A.A.	T_1 (s)	T_{1error} (s)	$T_{1\rho}$ (s)	$T_{1\rho error}$ (s)
I3 γ2	0.0587	0.0040	0.0089	0.0005
L5 δ1[b]				
L5 δ2	0.0476	0.0009	0.0052	0.0001
L10 δ1	0.0482	0.0008	0.0090	0.0001
L10 δ2	0.0603	0.0010	0.0082	0.0001
V11 γ1	0.0212	0.0006	0.0047	0.0001
V11 γ2	0.0333	0.0005	0.0060	0.0001
I13 γ2	0.0763	0.0035	0.0047	0.0001
I15 γ2	0.0786	0.0031	0.0053	0.0001
L19 δ1	0.0525	0.0005	0.0157	0.0002
L19 δ2	0.0605	0.0009	0.0147	0.0002
A22 β	0.0184	0.0018	0.0043	0.0002
L23 δ1	0.0405	0.0007	0.0094	0.0003
L23 δ2	0.0401	0.0005	0.0089	0.0002
L24 δ1	0.0436	0.0010	0.0052	0.0001
L24 δ2	0.0690	0.0032	0.0048	0.0001
A28 β	0.0133	0.0009	0.0040	0.0004
V32 γ1	0.0383	0.0006	0.0059	0.0001
V32 γ2	0.0358	0.0015	0.0056	0.0001
I33 γ2	0.0296	0.0010	0.0059	0.0001
L38 δ1	0.0365	0.0006	0.0073	0.0002
L38 δ2	0.0366	0.0004	0.0078	0.0001
I47 γ2	0.0403	0.0009	0.0077	0.0001
I50 γ2	0.0134	0.0010	0.0043	0.0002
I54 γ2	0.0533	0.0019	0.0054	0.0002
V56 γ1	0.0806	0.0031	0.0047	0.0001
V56 γ2	0.0418	0.0008	0.0051	0.0001
I62 γ2	0.0418	0.0012	0.0056	0.0001
I63 γ2	0.0527	0.0012	0.0070	0.0001
I64 γ2	0.0385	0.0007	0.0101	0.0002
I66 γ2	0.0743	0.0042	0.0050	0.0001
A67 β	0.0726	0.0023	0.0042	0.0001
A71 β	0.0394	0.0012	0.0041	0.0001
I72 γ2	0.0482	0.0009	0.0088	0.0001
V75 γ1	0.0379	0.0006	0.0082	0.0001
V75 γ2	0.0271	0.0003	0.0074	0.0001
L76 δ1	0.0285	0.0014	0.0047	0.0001
L76 δ2	0.0474	0.0016	0.0053	0.0001
V77 γ1	0.0157	0.0003	0.0049	0.0001

V77 γ2	0.0342	0.0008	0.0054	0.0001
V82 γ1	0.0294	0.0003	0.0155	0.0002
V82 γ2	0.0248	0.0004	0.0138	0.0002
I84 γ2	0.1038	0.0043	0.0058	0.0001
I85 γ2	0.0722	0.0025	0.0053	0.0002
L89 δ1	0.0722	0.0025	0.0053	0.0002
L89 δ2	0.0573	0.0020	0.0062	0.0001
L90 δ1	0.0573	0.0020	0.0062	0.0001
L90 δ2	0.0670	0.0031	0.0056	0.0002
I93 γ2	0.0374	0.0010	0.0049	0.0001
A95 β	0.0633	0.0026	0.0051	0.0001
L97 δ1	0.0488	0.0015	0.0174	0.0030
L97 δ2	0.0488	0.0015	0.0174	0.0030

[a]Recorded in a 1 kHz spin lock field.
[b]L5 δ1 has T_1 and T_2 have large errors due to the large ^{13}C linewidth, caused by chemical exchange.

Supplementary Table 4. Model-free parameters derived from ^{13}C R_1 and R_2 data.

A.A.	S^2_{axis} value	S^2_{axis} error	τ_e(ns) value	τ_e(ns) error	χ^2
I3 γ2	0.7642	0.0416	0.0366	0.0020	0.0000
L5 δ1	0.8567	0.0788	0.1736	0.0168	0.0000
L5 δ2	0.8417	0.0181	0.0471	0.0011	0.0000
L10 δ1	0.5402	0.0136	0.0473	0.0008	0.0000
L10 δ2	0.5490	0.0340	0.0493	0.0037	0.0000
V11 γ1	0.8082	0.0239	0.1150	0.0044	0.0000
V11 γ2	0.7734	0.0268	0.0706	0.0025	0.0000
I13 γ2	0.9708	0.0302	0.0285	0.0011	0.0000
I15 γ2	0.9302	0.0151	0.0257	0.0010	0.0000
L19 δ1	0.2813	0.0096	0.0442	0.0015	0.0001
L19 δ2	0.3031	0.0168	0.0374	0.0006	0.0000
A22 β	1.1622	0.0323	0.1970	0.0102	0.0000
L23 δ1	0.5324	0.0174	0.0479	0.0011	0.0000
L23 δ2	0.5171	0.0219	0.0550	0.0023	0.0000
L24 δ1	0.9184	0.0208	0.0519	0.0023	0.0000
L24 δ2	0.9403	0.0333	0.0284	0.0017	0.0000
A28	1.0399	0.0246	0.2271	0.0171	0.0000
V32 γ1	0.7968	0.0140	0.0646	0.0021	0.0000
V32 γ2	0.8210	0.0165	0.0626	0.0009	0.0000
I33 γ2	0.7676	0.0155	0.0881	0.0024	0.0000

L38 δ1	0.7080	0.0264	0.0632	0.0014	0.0000
L38 δ2	0.5354	0.0194	0.0574	0.0021	0.0002
I47 γ2	0.5631	0.0238	0.0518	0.0009	0.0000
I50 γ2	0.8190	0.0303	0.1441	0.0027	0.0000
I54 γ2	0.9215	0.0211	0.0492	0.0024	0.0001
V56 γ1	1.0079	0.0115	0.0230	0.0010	0.0000
V56 γ2	0.8623	0.0144	0.0553	0.0010	0.0001
I62 γ2	0.7944	0.0207	0.0542	0.0012	0.0001
I63 γ2	0.6701	0.0267	0.0386	0.0005	0.0000
I64 γ2	0.4226	0.0181	0.0518	0.0012	0.0000
I66 γ2	0.9720	0.0153	0.0257	0.0010	0.0000
A67 β	1.1049	0.0135	0.0287	0.0005	0.0000
A71β	1.1546	0.0146	0.0636	0.0019	0.0000
I72 γ2	0.5016	0.0117	0.0392	0.0009	0.0001
V75 γ1	0.5335	0.0183	0.0536	0.0008	0.0002
V75 γ2	0.5551	0.0193	0.0931	0.0031	0.0001
L76 δ1	0.7059	0.0344	0.0968	0.0021	0.0000
L76 δ2	0.8561	0.0217	0.0516	0.0022	0.0000
V77 γ1	0.7751	0.0316	0.1646	0.0113	0.0001
V77 γ2	0.8330	0.0316	0.0656	0.0028	0.0000
V82 γ1	0.2616	0.0141	0.0716	0.0026	0.0000
V82 γ2	0.2120	0.0243	0.0896	0.0043	0.0000
I84 γ2	0.8711	0.0170	0.0116	0.0005	0.0000
I85 γ2	0.9883	0.0359	0.0285	0.0022	0.0000
L89 δ1	0.8597	0.0222	0.0855	0.0048	0.0001
L89 δ2	0.7198	0.0174	0.0335	0.0010	0.0000
L90 δ1	1.1579	0.0253	0.0488	0.0017	0.0000
L90 δ2	0.9182	0.0259	0.0308	0.0011	0.0000
I93 γ2	0.9187	0.0189	0.0667	0.0024	0.0000
A95 β	0.9585	0.0297	0.0392	0.0012	0.0000
L97 δ1	0.4350	0.0334	0.0416	0.0007	0.0000
L97 δ2	0.4350	0.0369	0.0416	0.0006	0.0000

Supplementary Table 5. Model-free parameters derived from ^{2}H R_1 and R_2 data.

A.A.	S^2_{axis}		τ_e(ns)		χ^2
	value	error	value	error	
I3 γ2	0.4852	0.0332	0.0369	0.0031	0.0000
L5 δ1[a]					
L5 δ2	0.8721	0.0185	0.0436	0.0010	0.0000
L10 δ1	0.4600	0.0065	0.0467	0.0010	0.0000

L10 δ2	0.5365	0.0081	0.0352	0.0007	0.0001
V11 γ1	0.8418	0.0240	0.1152	0.0036	0.0000
V11 γ2	0.6955	0.0134	0.0692	0.0013	0.0000
I13 γ2	1.0165	0.0234	0.0206	0.0016	0.0000
I15 γ2	0.8961	0.0167	0.0210	0.0014	0.0001
L19 δ1	0.2273	0.0043	0.0444	0.0005	0.0001
L19 δ2	0.2621	0.0046	0.0378	0.0006	0.0000
A22 β	0.9053	0.0578	0.1352	0.0143	0.0000
L23 δ1	0.4158	0.0162	0.0572	0.0012	0.0000
L23 δ2	0.4449	0.0130	0.0576	0.0008	0.0000
L24 δ1	0.8621	0.0192	0.0490	0.0015	0.0001
L24 δ2	0.9869	0.0201	0.0247	0.0020	0.0001
A28 β	0.8834	0.1403	0.1950	0.0140	0.0000
V32 γ1	0.7298	0.0142	0.0585	0.0012	0.0001
V32 γ2	0.7667	0.0176	0.0631	0.0034	0.0000
I33 γ2	0.6905	0.0166	0.0792	0.0031	0.0000
L38 δ1	0.5577	0.0177	0.0632	0.0012	0.0000
L38 δ2	0.5133	0.0080	0.0633	0.0008	0.0001
I47 γ2	0.5348	0.0087	0.0566	0.0016	0.0000
I50 γ2	0.7979	0.0665	0.1926	0.0155	0.0000
I54 γ2	0.8474	0.0336	0.0378	0.0018	0.0000
V56 γ1	1.0203	0.0228	0.0186	0.0014	0.0000
V56 γ2	0.8765	0.0200	0.0515	0.0013	0.0000
I62 γ2	0.7874	0.0162	0.0522	0.0017	0.0000
I63 γ2	0.6307	0.0109	0.0405	0.0011	0.0000
I64 γ2	0.3717	0.0101	0.0608	0.0012	0.0001
I66 γ2	0.9497	0.0204	0.0224	0.0020	0.0000
A67	1.1422	0.0307	0.0208	0.0012	0.0000
A71	1.1126	0.0316	0.0535	0.0021	0.0001
I72 γ2	0.4729	0.0064	0.0466	0.0010	0.0000
V75 γ1	0.4864	0.0086	0.0610	0.0012	0.0000
V75 γ2	0.4995	0.0099	0.0883	0.0011	0.0000
L76 δ1	0.9043	0.0246	0.0817	0.0047	0.0000
L76 δ2	0.8531	0.0188	0.0441	0.0020	0.0000
V77 γ1	0.7110	0.0227	0.1608	0.0033	0.0001
V77 γ2	0.7938	0.0172	0.0665	0.0018	0.0000
V82 γ1	0.1548	0.0046	0.0824	0.0008	0.0000
V82 γ2	0.1627	0.0067	0.0982	0.0016	0.0000
I84 γ2	0.8287	0.0158	0.0136	0.0011	0.0002
I85 γ2	0.8903	0.0378	0.0241	0.0013	0.0000
L89 δ1	0.7626	0.0178	0.0830	0.0022	0.0000
L89 δ2	0.7324	0.0134	0.0355	0.0016	0.0000
L90 δ1	0.9749	0.0447	0.0434	0.0028	0.0000
L90 δ2	0.8334	0.0360	0.0277	0.0020	0.0000

I93 γ2	0.9029	0.0190	0.0589	0.0020	0.0000
A95	0.9179	0.0198	0.0291	0.0017	0.0000
L97 δ1	0.1883	0.0465	0.0483	0.0018	0.0000
L97 δ2	0.1883	0.0500	0.0483	0.0017	0.0000

[a]L5 δ1 was not fit due to large errors in relaxation data.

Supplementary Table 6. Values of the methyl ^{13}C principal shielding components, $\sigma_{11},\sigma_{22},\sigma_{33}$[a], and the isotropic shift, σ_{iso}, of three model peptides.

model peptide	σ_{11}	σ_{22}	σ_{33}	σ_{iso}
Gly-Val.HCl	-21.16±0.07	2.87±1.04	18.29±0.97	19.9±0.1
	-15.00±0.16	5.54±2.62	9.47±2.79	16.5±0.1
Gly-Leu-Gly.HCl	-15.52±0.59	2.16±1.83	13.36±1.39	25.7±0.1
	-20.08±0.16	7.53±1.79	12.54±1.72	24.3±0.1
Gly-Ile	-7.09±0.62	-2.61±0.81	9.70±0.31	16.6±0.1
	-9.34±0.33	2.21±1.23	7.13±0.91	12.4±0.1

[a]In ppm with $\sigma_{11} < \sigma_{22} < \sigma_{33}$ and $\sigma_{11} + \sigma_{22} + \sigma_{33} = 0$.
[b]The values of the isotropic chemical shifts are relative to TMS.