Electronic Supporting Material

Internal protein dynamics on ps to μs timescales as studied by multi-frequency ^{15}N solid-state NMR relaxation.

Tatjana Zinkevich,[1] VeniaminChevelkov,[2] Bernd Reif,[3] Kay Saalwächter,[1] Alexey Krushelnitsky[1,4]

[1] Martin-Luther-Universität Halle-Wittenberg, Institut für Physik, Halle (Saale), Germany;

[2] Max-Planck-Institut für Biophysikalische Chemie, Göttingen, Germany;

[3] Technische Universität München, Department Chemie, Garching, Germany;

[4] Kazan Federal University, Microbiology Department, Kazan, Russia.

Table S1. Experimentally measured (yellow rows) and simulated residue-resolved relaxation rates and dipolar coupling order parameters. The simulated rates were calculated according to the dynamic parameters presented in Fig. S2 and the models providing the best fit results. Signs "I", "II", "IIa" and "III" (second column) denote the model: "one motion", "two motions", "two motions with distribution" and "three motions", respectively.
T_1 (A) - ^{15}N T_1 at 400 MHz, MAS rate 13 kHz, t=12 °C;
T_1 (B) - ^{15}N T_1 at 600 MHz, MAS rate 13 kHz, t=12 °C,
T_1 (C) - ^{15}N T_1 at 900 MHz, MAS rate 13 kHz, t=12 °C;
T_1 (D) - ^{15}N T_1 at 600 MHz, MAS rate 10 kHz, t=14 °C;
T_1 (E) - ^{15}N T_1 at 600 MHz, MAS rate 10 kHz, t=24 °C;
$T_{1\rho}$ (A) - ^{15}N $T_{1\rho}$ at 400 MHz, MAS rate 20 kHz, on-resonance spin-lock 8 kHz (^{15}N resonance frequency in the rotating frame), t=10 °C;
$T_{1\rho}$ (B) - ^{15}N $T_{1\rho}$ at 400 MHz, MAS rate 20 kHz, on-resonance spin-lock 13 kHz, t=27 °C;
$T_{1\rho}$ (C) - ^{15}N $T_{1\rho}$ at 400 MHz, MAS rate 20 kHz, on-resonance spin-lock 8 kHz, t=27 °C;
$T_{1\rho}$ (D)- ^{15}N $T_{1\rho}$ at 600 MHz, MAS rate 10 kHz, off-resonance spin-lock 35 kHz (effective ^{15}N resonance frequency in the tilted rotating frame), the off-resonance angle between B_0 and B_{1e} fields 24°, t=14 °C;
$T_{1\rho}$ (E) - ^{15}N $T_{1\rho}$ at 600 MHz, MAS rate 10 kHz, off-resonance spin-lock 35 kHz, the off-resonance angle between B_0 and B_{1e} fields 24°, t=24 °C;
$T_{1\rho}$ (F) - ^{15}N $T_{1\rho}$ at 600 MHz, MAS rate 10 kHz, off-resonance spin-lock 46.3 kHz, the off-resonance angle between B_0 and B_{1e} fields 24°, t=14 °C.

Residue №		Dip. Coupl. order parameters	T_1 (A)	T_1 (B)	T_1 (C)	T_1 (D)	T_1 (E)	$T_{1\rho}$ (A)	$T_{1\rho}$ (B)	$T_{1\rho}$ (C)	$T_{1\rho}$ (D)	$T_{1\rho}$ (E)	$T_{1\rho}$ (F)
L8	e	0.635±0.108	4.88±2.2	4.65±0.55	5.36±0.75	4.07±0.2		0.046±0.004	0.081±0.005	0.105±0.008	0.27±0.01	0.413±0.024	0.34±0.03
	III	0.634	3.44	4.45	5.36	4.09		0.0435	0.0889	0.0889	0.273	0.383	0.319
V9	e	0.832±0.06	25.71±5.92	34.5±4.4	49.35±3.39		36.1±4.2	0.5±0.05	0.56±0.09	0.962±0.268	2.78±0.78		
	II	0.832	24.5	38.5	49.4		34.8	0.5	0.638	0.638	2.8		
L10	e	0.848±0.049	13.29±2.13	21.95±1.97	25.58±2.42		15.3±1.4	0.415±0.038	0.48±0.08				
	III	0.844	13.4	20.1	27.1		15.4	0.41	0.48				
A11	e	0.846±0.036	17.84±2.57	28.53±6.06	18.47±3.94			0.654±0.14	0.65±0.09	0.66±0.09			
	II	0.847	18.6	21	20.4			0.52	0.67	0.67			
L12	e	0.836±0.036	20.86±8.06	19.79±4.09	43.53±13.23			0.769±0.136	0.76±0.21	0.76±0.1			
	III	0.837	11.7	22	33.5			0.553	0.819	0.819			
Y13	e	0.79±0.035	40.51±8.34	49.32±4.26	41.4±5.8	36.9±6.7	42.7±7.8	0.5±0.05	0.5±0.07	0.67±0.12	4.5±1.3		3.57±1.07
	II	0.792	37.2	44	42.6	44.7	47	0.472	0.607	0.607	2.6		2.6
D14	e	0.766±0.038	13.85±2	15.57±3.75	19.35±1.96			0.46±0.03	0.49±0.05	0.66±0.03	3.85±1.41	3.13±0.5	
	II	0.768	14.7	18.7	18.9			0.446	0.62	0.62	2.39	2.78	

Residue №		Dip. Coupl. order parameters	T_1 (A)	T_1 (B)	T_1 (C)	T_1 (D)	T_1 (E)	$T_{1\rho}$ (A)	$T_{1\rho}$ (B)	$T_{1\rho}$ (C)	$T_{1\rho}$ (D)	$T_{1\rho}$ (E)	$T_{1\rho}$ (F)
Y15	e	0.8±0.047	23.86±3.93	29.9±1.4	37.56±2.53	19.8±1.9	26.4±2.5	0.53±0.12	0.61±0.07	0.645±0.046			
	I	0.933	13.8	26	40.5	23.9	20.5	0.461	0.61	0.61			
Q16	e	0.754±0.046	10.83±1.72	15.36±1.7	19.73±1.95	12.4±0.8	9±0.57	0.38±0.04	0.614±0.075	0.62±0.06	2.22±0.7		2.44±0.4
	III	0.752	8.65	14.8	20.5	12.4	9.22	0.42	0.78	0.78	2.38		2.69
E17	e	0.772±0.046	16.99±3.28	17.86±2.32	32.2±4			0.38±0.04	0.326±0.033	0.4±0.04	1.22±0.2		1.28±0.27
	I	0.9	10.3	19.5	30.8			0.272	0.356	0.356	1.5		1.5
K18	e	0.816±0.023	15.02±3.35	34.65±2	38.94±4.17	21±1.28	29.7±1.8	0.38±0.04	0.48±0.1	0.465±0.121	2.38±0.5		
	I	0.927	15.6	29.6	45.5	26.9	23.2	0.33	0.44	0.44	1.85		
S19	e	0.71±0.067	10.72±2.28	11.02±0.86	12.74±0.57	8.3±0.5	12.7±0.8	0.19±0.02	0.238±0.018	0.32±0.018	1.18±0.19	1.47±0.15	2.22±0.6
	III	0.682	8.68	10.7	12.9	9.77	12.5	0.182	0.262	0.262	1.2	1.28	1.9
V23	e	0.779±0.07	8.45±2.33	18.72±0.28	14.61±2.59		12.7±0.7	0.27±0.02	0.446±0.05	0.614±0.07			
	II	0.774	11.8	18.1	20.6		12.8	0.259	0.568	0.568			
T24	e	0.637±0.13	7.51±0.92	10.3±0.4	17.2±1.24	8.3±0.34	8.8±0.36	0.13±0.01	0.215±0.008	0.29±0.027	1.67±0.56		2.5±1
	III	0.606	8.29	11.9	15.2	9.87	9.87	0.13	0.218	0.218	0.866		1.34
M25	e	0.843±0.036	37.67±12.83	30.42±3.06	32.9±2.5	23 ±1.2	31.4±1.7	0.93±0.32	0.71±0.16	1.67±0.6			
	III	0.842	22.5	25	28.2	24.9	27.5	0.567	0.9	0.9			
K26	e	0.815±0.036	22.33±3.85	30.68±2.56	33.6±3.13	17±2.4	19.2±2.7	0.47±0.04	0.468±0.053	0.59±0.05			
	I	0.932	12.8	24.1	37.4	22	19.1	0.453	0.602	0.602			
K27	e	0.806±0.047	20.25±5.74	16.2±3.4	22.6±2.5		10.6±0.5	0.52±0.07	0.694±0.058		2.27±0.66		2.6±0.5
	II	0.81	8.03	15.1	23.4		10.5	0.47	0.7		2.44		2.44
G28	e	0.837±0.036	22.47±7.67	27.7±1.13	34.5±2.9	16.8±1.2	18.8±1.3	0.71±0.08	0.752±0.107	0.89±0.09	4.17±1.46		2.86±0.63
	I	0.939	12.4	23.4	36.4	21	16.6	0.589	0.895	0.895	3.18		3.18
D29	e	0.847±0.048	10.87±0.95	18.6±3.3	25.9±3.16			0.58±0.07	0.866±0.21	0.8±0.14			
	II	0.847	10.7	18.7	25.9			0.58	0.818	0.818			
I30	e	0.812±0.047	26.38±9.98	17.27±3.17	34.2±8.3		19.1±2.7	0.46±0.04	0.404±0.075	0.625±0.105			
	I	0.921	11.1	20.9	33.1		16.7	0.376	0.502	0.502			
L31	e	0.866±0.074	22.25±3.9	24.9±3.4	47.74±11.12		44±12	1.43±0.5	1.67±1.08	1.01±0.29			
	III	0.879	22.7	26.8	34.2		36.4	1.39	0.966	1.02			
T32	e	0.725±0.139	5.85±1.14	5.84±1.09	9.86±1.3		4.8±0.21	0.12±0.008	0.227±0.027	0.214±0.016		0.82±0.068	
	I	0.753	3.74	7.04	11.2		4.74	0.124	0.201	0.201		0.868	
L33	e	0.853±0.061	23.63±4.96	39.8±.8	47.67±7.48	20.5±2.4	31.5±3.7	0.21±0.02	0.319±0.051	0.377±0.03	0.763±0.074		0.9±0.15

Residue №		Dip. Coupl. order parameters	T_1 (A)	T_1 (B)	T_1 (C)	T_1 (D)	T_1 (E)	$T_{1\rho}$ (A)	$T_{1\rho}$ (B)	$T_{1\rho}$ (C)	$T_{1\rho}$ (D)	$T_{1\rho}$ (E)	$T_{1\rho}$ (F)
	I	0.897	15.1	31	47.67	24.3	16.5	0.153	0.317	0.317	0.953		0.953
L34	e	0.825±0.072	24.17±6.46	23.97±0.25	23.9±0.66		15.2±1.7	0.28±0.03	0.392±0.046	0.4±0.06			
	II	0.81	18	24.7	24.7		19.6	0.21	0.45	0.45			
N35	e	0.726±0.058	21.36±2.85	36.7±2.1	54.63±10.3	23.6±2.3	22.2±1.8	0.472±0.05	0.234±0.07	0.455±0.05		3.85±1.14	2.38±0.63
	I	0.929	16.6	31.9	50.5	29.1	24	0.296	0.42	0.42		2.07	1.71
K39	e	0.756±0.069	7.36±1.71	7.85±0.56	12.88±0.77			0.38±0.06	0.446±0.038	0.76±0.057	2±0.6		
	III	0.759	4.17	8.07	12.7			0.393	0.467	0.725	2.1		
D40	e	0.815±0.06	22.68±7.94	17.72±8.08	12.54±4.02		21±2.4	0.17±0.008	0.226±0.014	0.28±0.02		0.65±0.05	
	I	0.831	7.26	13.8	21.9		10.6	0.157	0.214	0.214		1.04	
W41	e	0.799±0.059	12.82±3.08	16.48±2.53	20.76±4.55			0.18±0.013	0.25±0.019	0.392±0.043	0.74±0.12		1.05±0.25
	I	0.858	7.9	15.8	24.5			0.173	0.271	0.271	0.983		0.983
W42	e	0.838±0.036	14.78±5.27	20.25±7.64	23.9±5.1			0.47±0.05	0.34±0.05	0.465±0.065			
	III	0.847	14.4	18.9	24.1			0.48	0.35	0.427			
V44	e	0.867±0.049	29.63±7.7	14.16±3.28	23.7±4.4			0.64±0.11	0.47±0.049	0.543±0.038			
	I	0.909	8.28	15.2	23.8			0.406	0.538	0.538			
E45	e	0.814±0.048	9.12±2.19	18.78±1.31	28.05±2.06		14±1.3	0.27±0.055	0.31±0.02	0.358±0.042			
	III	0.821	9.87	18.6	28.7		13.7	0.27	0.309	0.355			
V46	e	0.782±0.07	13.5±3.8	7.4±0.8	11.09±0.14			0.11±0.03	0.09±0.006	0.097±0.01	0.588±0.112	0.444±0.094	
	I	0.673	3.7	7.1	11.09			0.07	0.095	0.095	0.402	0.463	
R49	e	0.696±0.044	6.56±3.07	5.18±0.43	7.57±0.94			0.058±0.004	0.087±0.005	0.091±0.007			
	II	0.698	3.34	5.34	7.6			0.058	0.0861	0.0861			
Q50	e	0.686±0.044	4.71±0.72	6.07±0.17	9.8±0.3	4.9±0.1	4±0.1	0.096±0.007	0.133±0.007	0.144±0.005	0.41±0.026	0.56±0.02	0.53±0.04
	I	0.666	3.13	5.98	9.6	5.16	3.78	0.077	0.136	0.136	0.46	0.586	0.46
G51	e	0.775±0.035	14.47±2.62	17.12±1.72	29.71±3.12	19.6±1.2	16.8±1	0.175±0.011	0.274±0.022	0.3±0.03	0.83±0.15	1.33±0.23	
	III	0.777	13.3	19.5	26.9	18.4	16.6	0.168	0.28	0.28	0.964	1.27	
F52	e	0.848±0.036	26.4±7.04	26.64±1.7	39.15±4.27	23 ±2.2	17.2±1.6	0.298±0.023	0.332±0.035	0.35±0.02			1.43±0.15
	I	0.91	13.3	25.5	39.8	23.1	19.3	0.258	0.361	0.361			1.47
V53	e	0.836±0.048	32.15±7.83	45.35±2.09	60.2±18.02			0.56±0.06	0.662±0.15	0.72±0.12	2.5±0.65		
	II	0.836	30.3	45.3	54.8			0.54	0.696	0.696	2.93		
A55	e	0.818±0.024	27.85±2.28	27.53±2.65	41.02±2.12	20.5±1.4	18.6±1.3	0.402±0.073	0.458±0.099	0.74±0.1		3.03±0.53	
	I	0.926	14	26.3	41.7	23.9	19.6	0.336	0.577	0.577		2.62	
A56	e	0.797±0.035	12.96±2	15.44±6.45	20.5±3.37			0.138±0.01	0.289±0.032	0.42±0.04			

	II	0.797	12.9	18.2	20.1			0.138	0.342	0.342			
Residue №		Dip. Coupl. order parameters	T_1 (A)	T_1 (B)	T_1 (C)	T_1 (D)	T_1 (E)	$T_{1\rho}$ (A)	$T_{1\rho}$ (B)	$T_{1\rho}$ (C)	$T_{1\rho}$ (D)	$T_{1\rho}$ (E)	$T_{1\rho}$ (F)
Y57	e	0.797±0.051	19.91±6.49	11.73±4.94	27.7±4.67			0.36±0.05	0.505±0.21	0.56±0.1			
	I	0.902	8.4	15.9	25.7			0.33	0.525	0.525			
V58	e	0.82±0.048	21.32±7.07	37.33±5.34	53.34±8.46		19.9±1.5	0.6±0.15	0.74±0.16	1.15±0.3			
	III	0.82	18.8	37.3	56.3		19.9	0.6	0.582	0.9			
K60	e	0.77±0.035	5.28±0.74	5.37±0.48	9.3±0.5				0.39±0.03	0.42±0.04	0.65±0.05		0.72±0.03
	III	0.776	4.1	6.2	8.78				0.391	0.391	0.674		0.674
L61	e	0.731±0.034	3.24±0.37	6.35±0.35	7.62±0.87	3.7±0.12	4.9±0.16	0.113±0.007	0.243±0.018	0.28±0.007	0.78±0.12	0.752±0.072	1.11±0.17
	III	0.68	3.43	5.1	6.86	4.17	4.34	0.115	0.255	0.255	0.715	0.966	0.941
D62	e	0.3±0.08	1.12±0.07	1.33±0.11	1.9±0.12	1.18±0.05	1.12±0.05	0.031±0.005	0.058±0.005	0.052±0.002	0.16±0.008	0.22±0.004	0.21±0.004
	III	0.35	1.04	1.38	1.81	1.24	1.15	0.029	0.0546	0.0546	0.168	0.217	0.215
Side chains													
Q16a	e	0.088±0.01				0.82±0.05	0.766±0.04	0.13±0.02	0.235±0.017	0.235±0.022	0.625±0.084	0.568±0.05	0.645±0.09
	III	0.089				0.851	0.776	0.16	0.237	0.237	0.523	0.523	0.606
Q16b	e	0.088±0.013				0.94±0.07	1.2±0.1	0.14±0.03	0.228±0.031	0.27±0.034	0.787±0.098	0.75±0.11	0.741±0.116
	II	0.089				0.956	1.2	0.185	0.255	0.255	0.64	0.73	0.74
N35s	e	0.735±0.09		19.86±0.53	24.66±1.28	20±2.3	20±2.2	0.56±0.18	0.261±0.048	0.435±0.057	2.13±0.7		
	III	0.73		19.6	24	19	18.5	0.396	0.27	0.331	2.82		
N38s	e	0.756±0.11		22.41±7.22	33.2±5.1	21±3	27±4	0.27±0.07	0.365±0.053	0.435±0.04			
	I	0.915		23.2	36.6	22	18.6	0.301	0.402	0.402			
We41	e	0.768±0.12	5.11±0.27	7.3±0.4	13.3±0.56	5±0.23	2.9±0.13	0.039±0.003	0.021±0.002	0.027±0.002	0.135±0.003	0.114±0.002	0.227±0.008
	III	0.323	4.32	7.78	10.9	5.65	2.98	0.02	0.0207	0.0207	0.135	0.117	0.234
We42	e	0.832±0.012				17.7±2.6	47±7	0.535±0.054	0.366±0.04	0.555±0.096			
	III	0.824				19.5	43.6	0.44	0.309	0.327			
Q50a	e	0.391±0.005		1.81±0.03	1.88±0.04	1.44±0.14	1.12±0.2	0.225±0.01	0.275±0.027	0.397±0.031	1.23±0.17	0.752±0.05	1.05±0.14
	II	0.391		1.73	1.91	1.71	1.64	0.221	0.315	0.315	0.821	0.912	0.821
Q50b	e	0.394±0.006	1.26±0.12	1.89±0.03	1.88±0.04	1.56±0.07	1.2±0.07	0.248±0.06	0.267±0.024	0.333±0.013	0.82±0.07	0.61±0.06	0.787±0.083
	II	0.396	1.3	1.83	1.98	1.7	1.31	0.121	0.309	0.309	0.61	0.78	0.61

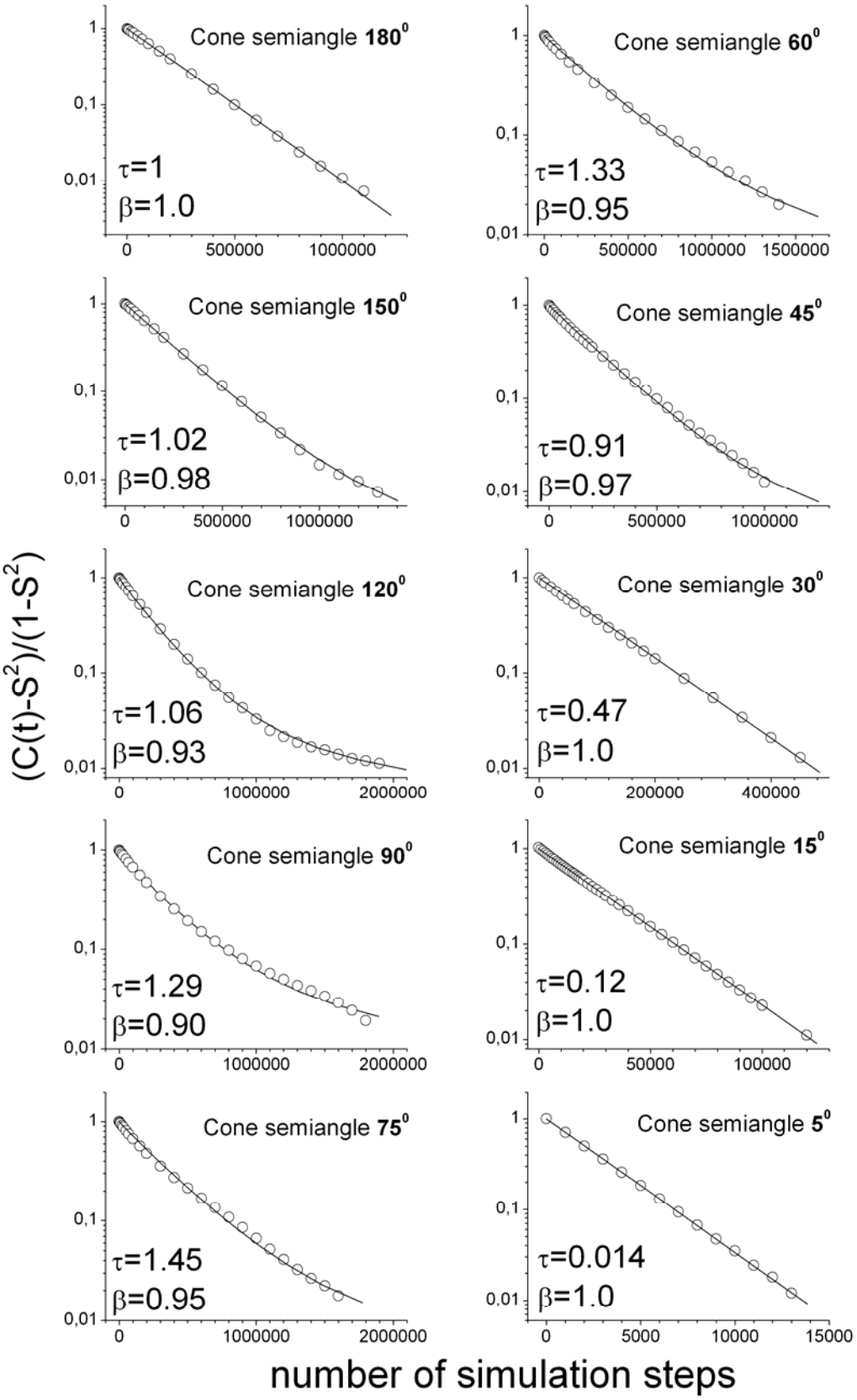

Figure S1. The comparison of the correlation functions for the "wobbling-in-a-cone" model obtained from the computer simulation of the internuclear vector diffusion within a cone (performed according to the protocol described in Kurbanov et al., *J Chem Phys*, 135, 184104, 2011) and numerical integration of the correlation function using the Fuoss-Kirkwood distribution function. The 180º-cone was taken as a reference for the correlation time (τ=1).

Table S2. Limitations for the activation energies during the Monte-Carlo simulations.

Fitting model	Activation energy limitations, kJ/mol
One component	24±12
Two components with distribution	32±19
Two components	fast 24±15 slow 31±21
Three components	intermediate 39±23 slow 42±22

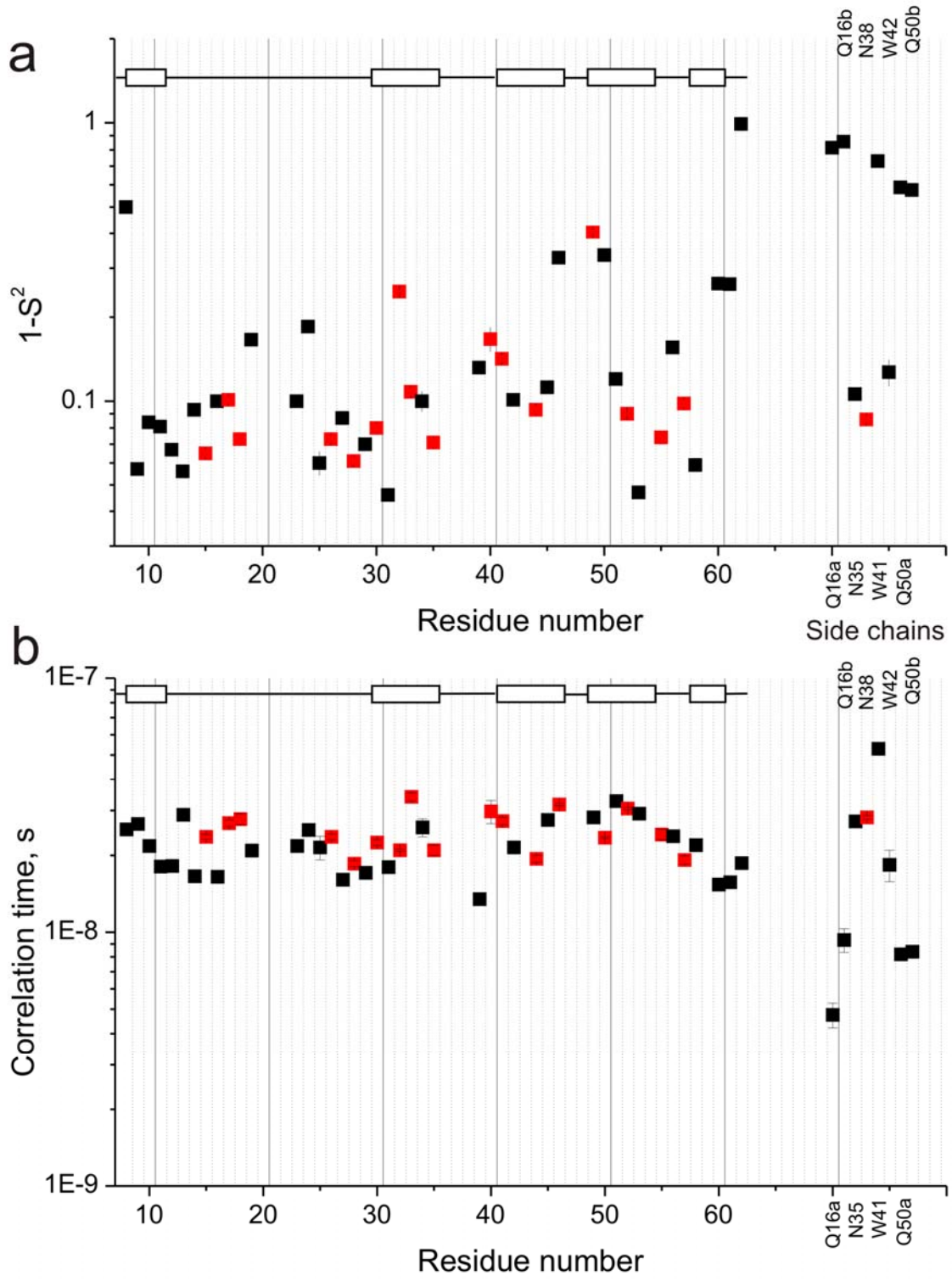

Figure S2. Fitting results for "one component" model. The order parameter (a) and the correlation time (b) as a function of a residue number. The residues having minimum *AIC* values among three discrete models are marked by red colour.

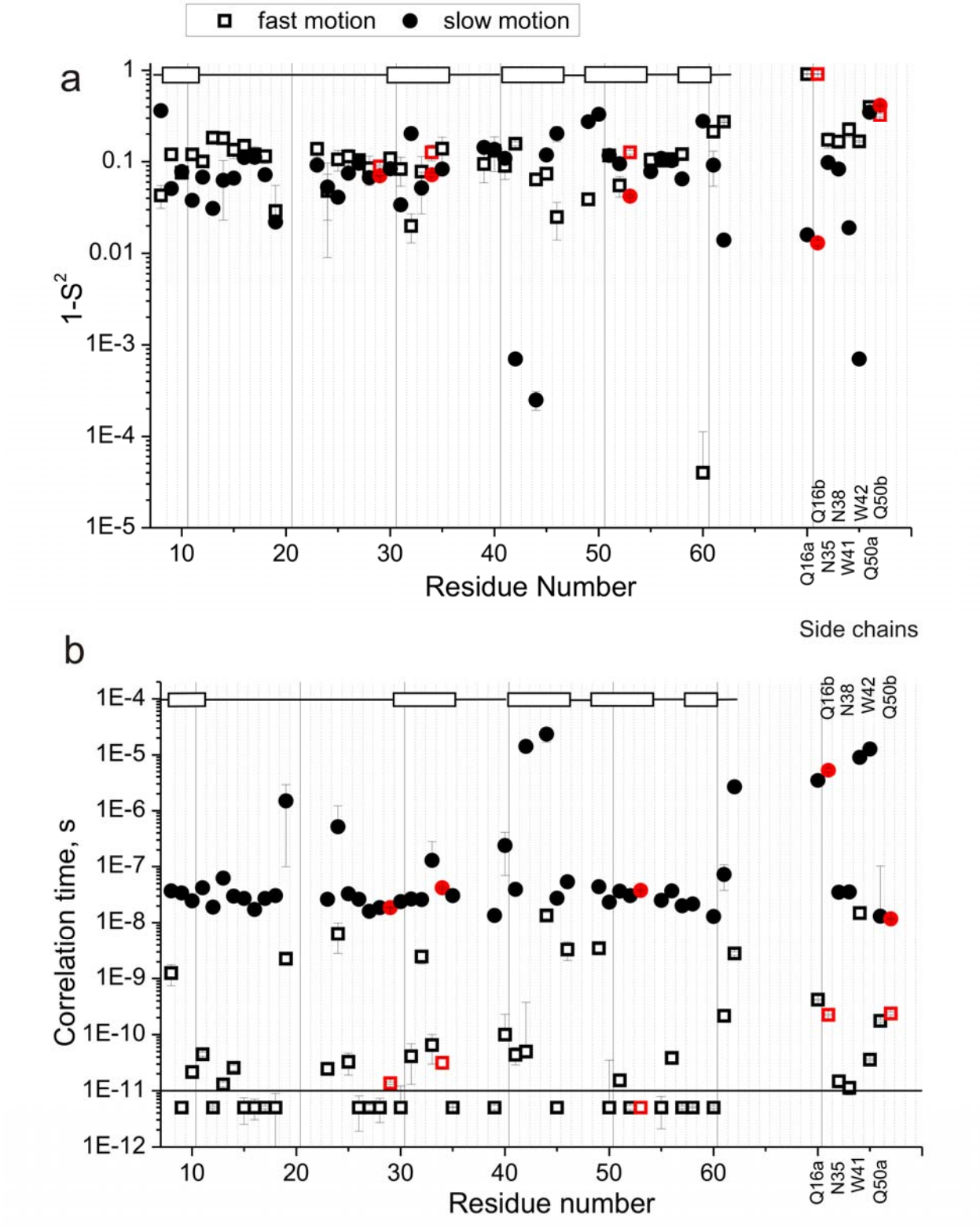

Figure S3. Fitting results for "two component" model. The order parameters (a) and the correlation times (b) for the fast and slow motions as a function of a residue number. The residues having minimum *AIC* values among three discrete models are marked by red colour.

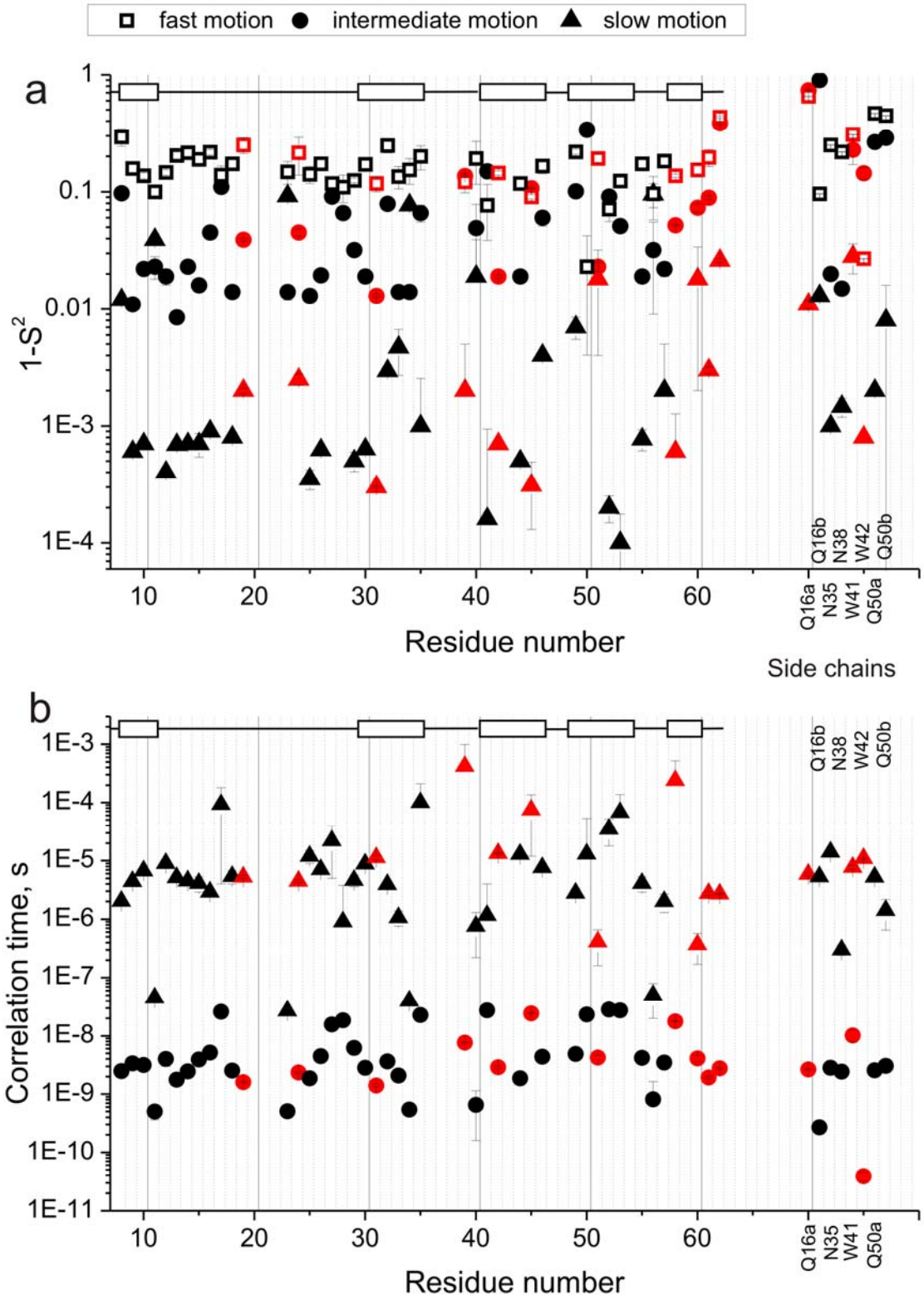

Figure S4. Fitting results for "three component" model. The order parameters (a) and the correlation times (b) for the fast, intermediate and slow motions as a function of a residue number. The residues having minimum *AIC* values among three discrete models are marked by red colour.

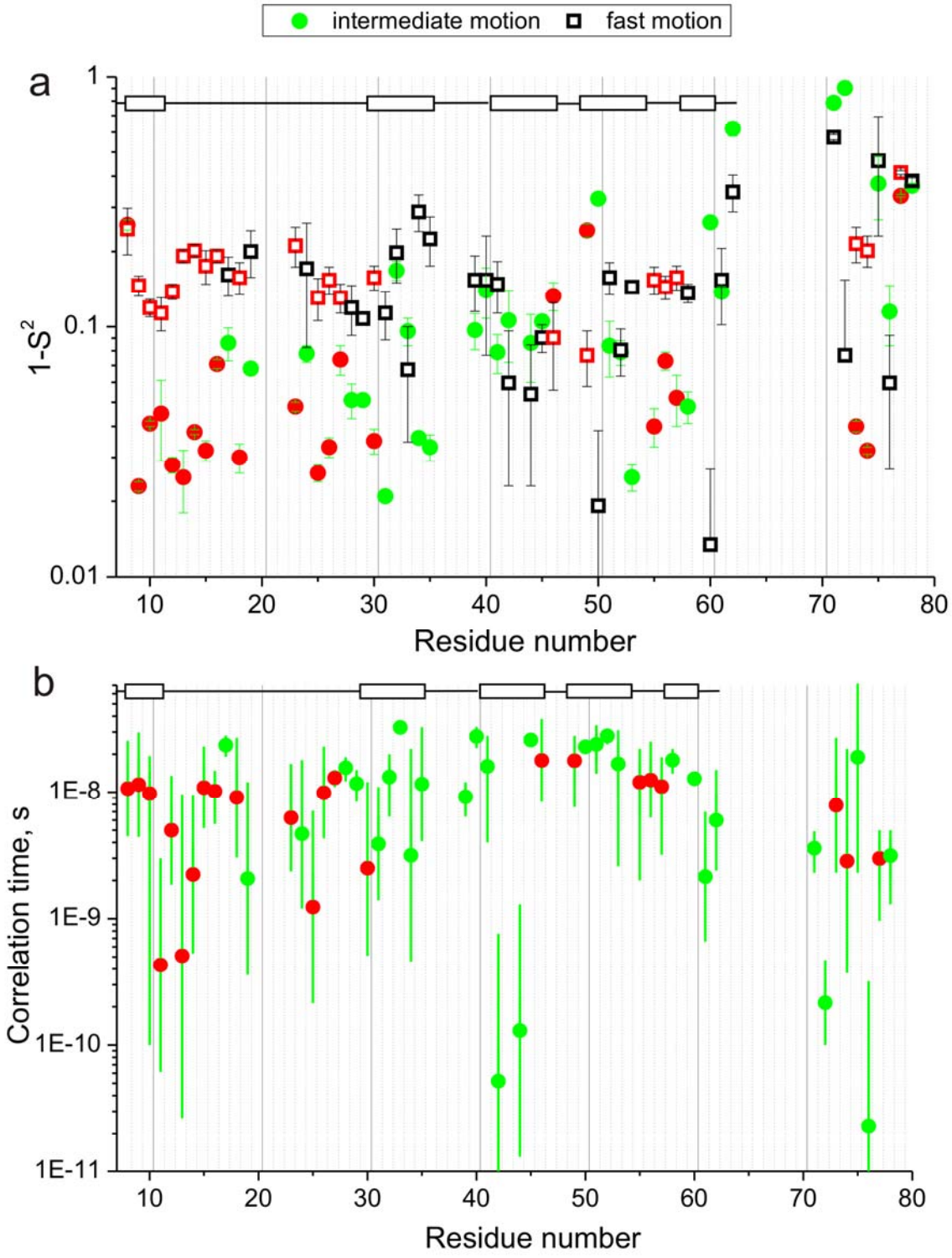

Figure S5. Fitting results for "two components with distribution" model. The order parameters (a) and the correlation times (b) as a function of a residue number. Green bars in the bottom figure define the width of the distribution (50% of the maximal intensity). The residues having minimum *AIC* values among all four fitting models are marked by red colour.

Table S3. χ's and *AIC* values for four fitting models. "1", "2a", "2" and "3" define "one component", "two components with distribution", "two components" and "three components" models, respectively. RL is the residue label, NP is the number of experimental data (relaxation times and dipolar order parameter). Yellow and green colours define the minimal *AIC* values among all four models and among three discrete models, respectively. χ is defined according to Eq. (16).

RL	NP	χ1	*AIC*1	χ2a	*AIC*2a	χ2	*AIC*2	χ3	*AIC*3
L8	11	1.5	14.92	0.975	9.441	1.18	15.64	1.02	14.44
V9	9	1.25	10.02	0.583	0.29	0.612	3.16	0.631	5.71
L10	8	1.22	9.18	0.557	0.64	0.733	7.03	0.5	2.91
A11	7	1.94	15.28	0.615	3.19	0.653	6.03	0.816	11.15
L12	7	1.33	10	0.701	5.03	0.706	7.13	0.538	5.32
Y13	11	2.22	23.55	0.682	1.58	0.963	11.17	0.893	11.51
D14	9	2.06	19.0	0.743	4.65	1.11	13.88	1.07	15.22
Y15	9	1.99	18.39	1.45	16.69	1.54	19.78	1.39	19.93
Q16	11	1.28	11.43	0.604	-1.09	0.758	5.90	0.534	0.2
E17	9	1.69	15.45	1.41	16.18	1.39	17.93	1.41	20.18
K18	10	2.72	26.01	2.14	25.22	2.11	26.93	1.97	27.56
S19	12	2.71	29.93	1.76	23.57	2.13	30.15	1.3	20.3
V23	8	2.28	19.19	1.4	15.38	1.29	16.07	1.2	16.92
T24	11	2.02	21.47	1.89	24.00	1.85	25.53	1.4	21.4
M25	9	2.82	24.67	1.71	19.66	1.89	23.46	1.49	21.18
K26	9	1.98	18.3	1.46	16.81	1.62	20.68	1.4	20.06
K27	9	1.14	8.36	0.789	5.73	0.804	8.07	0.776	9.44
G28	11	2.03	21.58	1.83	23.29	1.86	25.65	1.84	27.42
D29	9	0.874	3.58	0.143	-25	0.105	-28.57	0.135	-22.04
I30	8	1.42	11.61	0.889	8.12	1.08	13.23	0.862	11.62
L31	8	1.17	8.51	0.908	8.46	0.958	11.31	0.633	6.68
T32	9	0.994	5.89	0.869	7.47	0.756	6.97	0.659	6.49
L33	11	2	21.25	2	25.25	1.88	25.89	1.65	25.02
L34	8	6.48	35.9	1.97	20.85	1.52	18.7	1.39	19.27
N35	11	2.32	24.5	2.015	25.41	2.17	29.04	1.69	25.54
K39	8	1.87	16.02	1.79	19.32	1.79	21.32	0.841	11.23
D40	9	3.24	27.16	3.32	31.6	2.86	30.91	3.12	34.48
W41	9	1.48	13.06	1.42	16.31	1.34	17.27	1.44	20.56
W42	7	1.55	12.14	0.758	6.12	0.743	7.84	0.298	-2.95
V44	7	1.48	11.49	1.12	11.59	1.21	14.67	0.871	12.07
E45	8	0.747	1.33	0.487	-1.51	0.497	0.81	0.193	-12.32
V46	9	1.3	10.72	1.04	10.71	1.1	13.72	0.9	12.10
R49	7	1.1	7.33	0.459	-0.9	0.462	1.19	0.477	3.64
Q50	12	1.79	19.97	1.82	24.37	1.79	25.97	1.81	28.24
G51	11	1.47	14.48	1.11	12.3	0.91	9.93	0.78	8.53
F52	10	1.14	8.62	1.02	10.4	1	12	0.908	12.07
V53	8	1.04	6.63	0.445	-2.96	0.36	-4.35	0.544	4.26
A55	10	2.67	25.64	2	23.86	2.19	27.68	1.9	26.84
A56	7	1.6	12.58	1.14	11.83	1	12	1	14
Y57	7	1.13	7.71	0.774	6.41	0.793	8.75	0.726	9.52
V58	8	1.11	7.67	0.625	2.48	0.637	4.78	0.243	-8.64
K60	8	2.37	19.81	1.81	19.49	1.73	20.77	1.11	15.67

L61	12	4.2	40.44	3.15	37.54	3.11	39.23	2.2	32.92
D62	12	2.7	29.84	1.54	20.36	1.8	26.11	0.88	10.932
Side chains									
Q16a	9	3.66	29.35	0.975	9.54	0.954	11.15	0.755	8.94
Q16b	9	3.8	30.03	0.915	8.4	0.812	8.25	0.808	10.16
N35	9	2.05	18.92	0.9	8.1	1.04	12.71	0.752	8.87
N38	8	1.07	7.08	0.673	3.66	0.765	7.71	0.669	7.57
W41	12	7.66	54.86	7.55	58.52	3.8	44.04	3.1	41.15
W42	6	3.51	21.07	1.86	17.45	1.09	13.03	0.801	11.34
Q50a	11	6.82	48.24	1.98	25.03	2.05	27.79	2	29.25
Q50b	12	6.7	51.65	2.65	33.39	1.98	28.39	2.92	39.72

Table S4. χ_{abs}'s and *AIC* values for four fitting models, denotations are the same as in Table S3. χ_{abs} is defined according to Eq. (19).

RL	NP	$\chi_{abs}1$	*AIC*1	$\chi_{abs}2a$	*AIC*2a	$\chi_{abs}2$	*AIC*2	$\chi_{abs}3$	*AIC*3
L8	11	0.208	-28.54	0.135	-34.05	0.138	-31.57	0.113	-33.97
V9	9	0.179	-24.97	0.129	-26.86	0.133	-24.31	0.14	-21.39
L10	8	0.13	-26.64	0.058	-35.56	0.0878	-26.92	0.0438	-36.05
A11	7	0.159	-19.7	0.117	-20.04	0.131	-16.46	0.119	-15.8
L12	7	0.233	-14.4	0.187	-13.47	0.198	-10.67	0.199	-8.6
Y13	11	0.294	-20.93	0.131	-34.72	0.188	-24.77	0.158	-26.59
D14	9	0.244	-19.39	0.129	-26.86	0.164	-20.54	0.161	-18.87
Y15	9	0.19	-23.89	0.147	-24.51	0.165	-20.43	0.129	-22.86
Q16	11	0.129	-39.05	0.0815	-45.16	0.11	-36.56	0.0742	-43.22
E17	9	0.204	-22.6	0.198	-19.15	0.204	-16.61	0.2	-14.97
K18	10	0.15	-31.94	0.146	-28.48	0.136	-27.9	0.159	-22.78
S19	12	0.267	-25.69	0.16	-33.98	0.207	-25.8	0.113	-38.33
V23	8	0.363	-10.2	0.355	-6.57	0.229	-11.58	0.217	-10.45
T24	11	0.292	-21.08	0.256	-19.98	0.301	-14.41	0.224	-18.91
M25	9	0.316	-14.74	0.262	-14.11	0.276	-11.17	0.332	-5.85
K26	9	0.212	-21.92	0.181	-20.77	0.2	-16.97	0.167	-18.22
K27	9	0.216	-21.58	0.201	-18.88	0.213	-15.84	0.21	-14.09
G28	11	0.201	-29.3	0.19	-26.54	0.202	-23.19	0.197	-21.74
D29	9	0.0699	-41.89	0.0284	-54.1	0.0229	-55.98	0.0234	-53.59
I30	8	0.257	-15.74	0.211	-14.89	0.246	-10.44	0.201	-11.67
L31	8	0.282	-14.25	0.26	-11.55	0.255	-9.86	0.189	-12.66
T32	9	0.159	-27.1	0.129	-26.86	0.122	-25.87	0.108	-26.06
L33	11	0.24	-25.4	0.249	-20.59	0.246	-18.85	0.198	-21.63
L34	8	0.247	-16.37	0.18	-17.44	0.177	-15.71	0.176	-13.8
N35	11	0.381	-15.23	0.328	-14.52	0.38	-9.29	0.203	-21.08
K39	8	0.201	-19.67	0.178	-17.62	0.194	-14.24	0.166	-14.73
D40	9	0.421	-9.57	0.423	-5.49	0.235	-14.07	0.232	-12.3
W41	9	0.213	-21.84	0.196	-19.33	0.166	-20.32	0.209	-14.18
W42	7	0.246	-13.63	0.145	-17.03	0.197	-10.74	0.0425	-30.22
V44	7	0.303	-10.72	0.239	-10.04	0.203	-10.32	0.203	-8.32
E45	8	0.0691	-36.76	0.0701	-35.53	0.0611	-32.72	0.0319	-41.12
V46	9	0.288	-16.41	0.242	-15.54	0.263	-12.04	0.222	-13.09
R49	7	0.226	-14.82	0.188	-13.4	0.188	-11.4	0.182	-9.85

Q50	12	0.132	-42.6	0.131	-38.78	0.131	-36.78	0.13	-34.97
G51	11	0.144	-36.63	0.134	-34.22	0.113	-35.97	0.102	-36.22
F52	10	0.172	-29.21	0.162	-26.4	0.171	-23.32	0.167	-21.8
V53	8	0.137	-25.8	0.106	-25.91	0.0844	-27.56	0.126	-19.14
A55	10	0.211	-25.12	0.18	-24.3	0.215	-18.74	0.177	-20.63
A56	7	0.201	-16.46	0.134	-18.14	0.12	-17.68	0.124	-15.22
Y57	7	0.27	-12.33	0.269	-8.38	0.271	-6.29	0.257	-5.02
V58	8	0.163	-23.02	0.145	-20.9	0.152	-18.14	0.0601	-30.99
K60	8	0.214	-18.67	0.188	-16.74	0.188	-14.74	0.11	-21.32
L61	12	0.2	-32.63	0.2	-28.63	0.246	-21.66	0.152	-31.21
D62	12	0.171	-36.39	0.0924	-47.16	0.139	-35.36	0.0568	-54.84
Side chains									
Q16a	9	0.326	-14.18	0.131	-26.59	0.11	-27.73	0.104	-26.74
Q16b	9	0.47	-7.59	0.125	-27.43	0.114	-27.09	0.117	-24.62
N35	9	0.234	-20.14	0.201	-18.88	0.232	-14.3	0.18	-16.87
N38	8	0.152	-24,14	0.111	-25.17	0.117	-22.33	0.107	-21.76
W41	12	0.358	-18.65	0.358	-14.65	0.226	-23.69	0.215	-22.89
W42	6	0.43	-4.13	0.256	-6.35	0.153	-10.53	0.123	-11.15
Q50a	11	0.374	-15.64	0.233	-22.05	0.216	-21.71	0.212	-20.13
Q50b	12	0.344	-19.61	0.194	-29.36	0.199	-26.75	0.175	-27.83

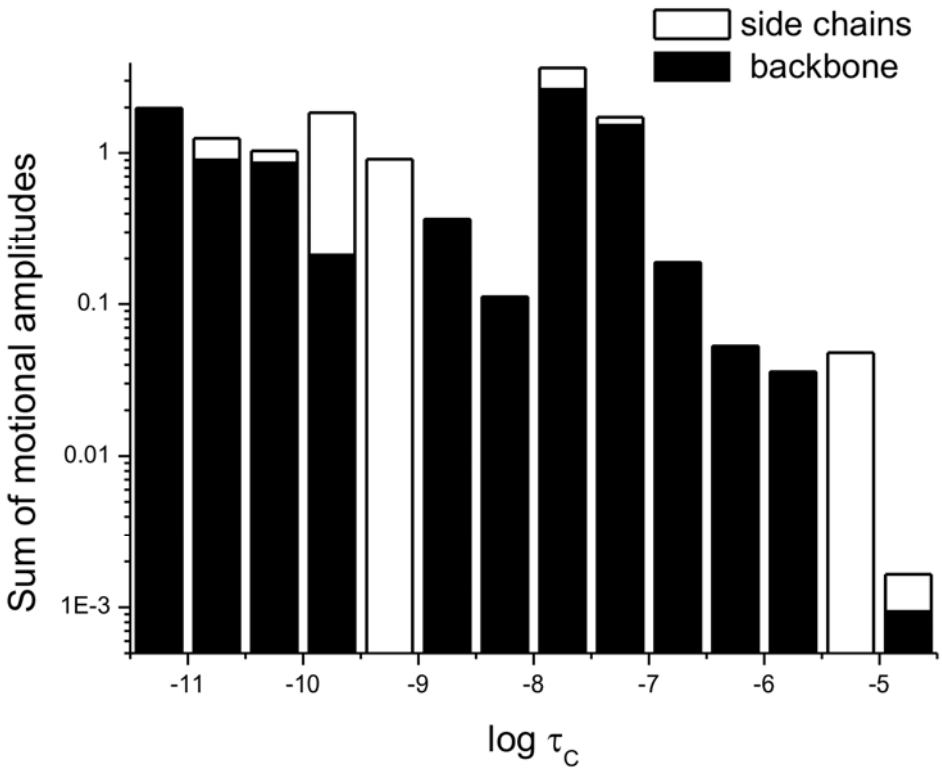

Figure S6. Histogram of summarized amplitudes ($1\text{-}S^2$) as a function of the time scale of motion for the "two components" model.

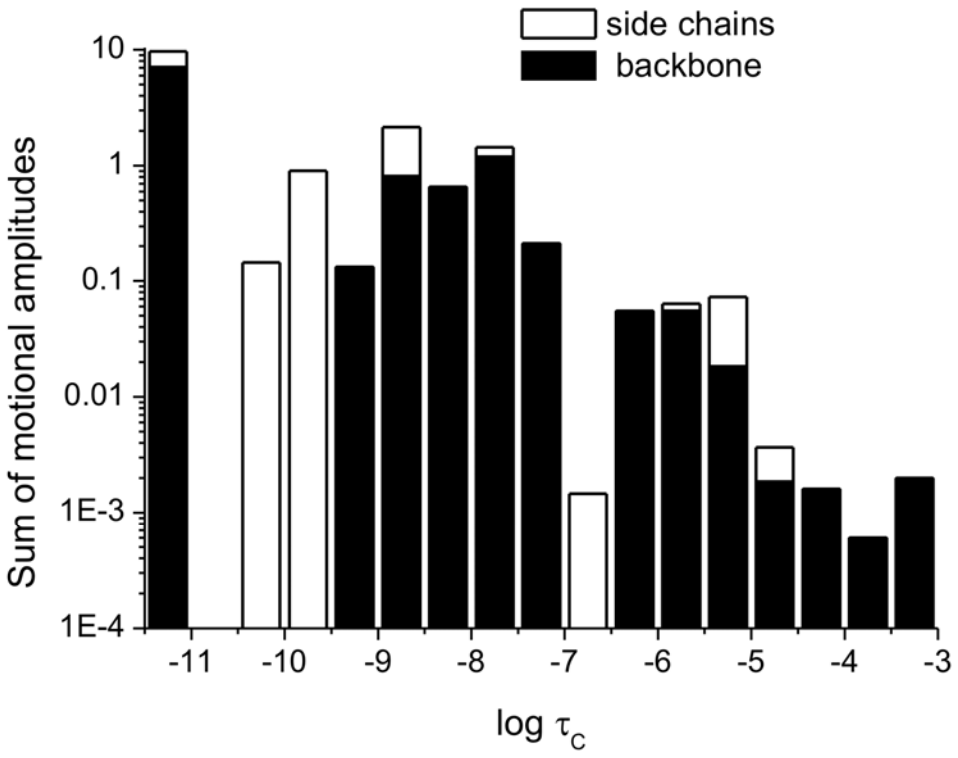

Figure S7. Histogram of summarized amplitudes ($1\text{-}S^2$) as a function of the time scale of motion for the "three components" model.

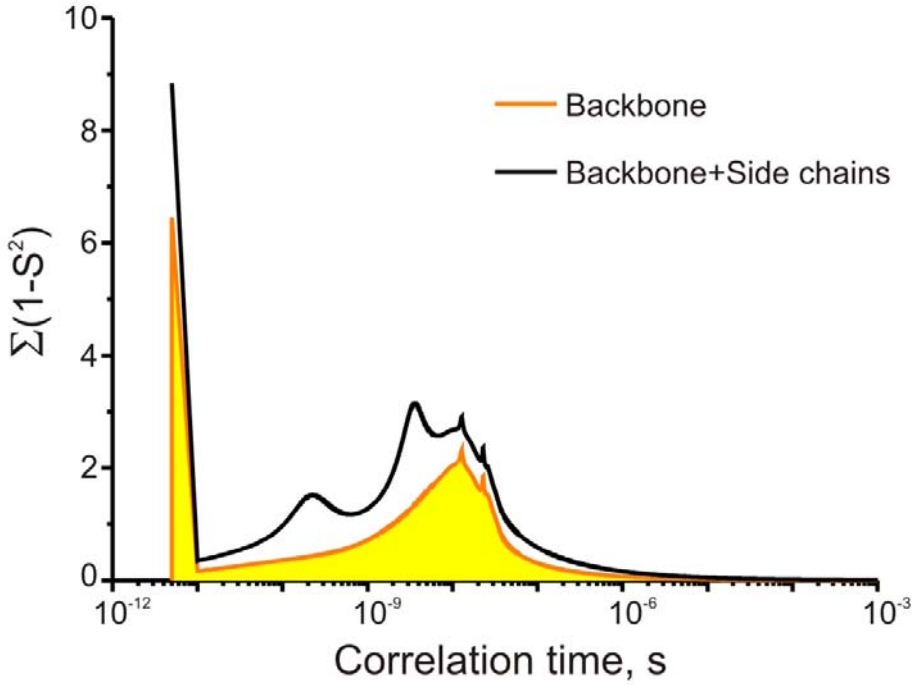

Figure S8. Smooth distribution of the internal mobility over the time scale axis (analogously to Figs. S6 and S7) for the "two components with distribution" fitting model.

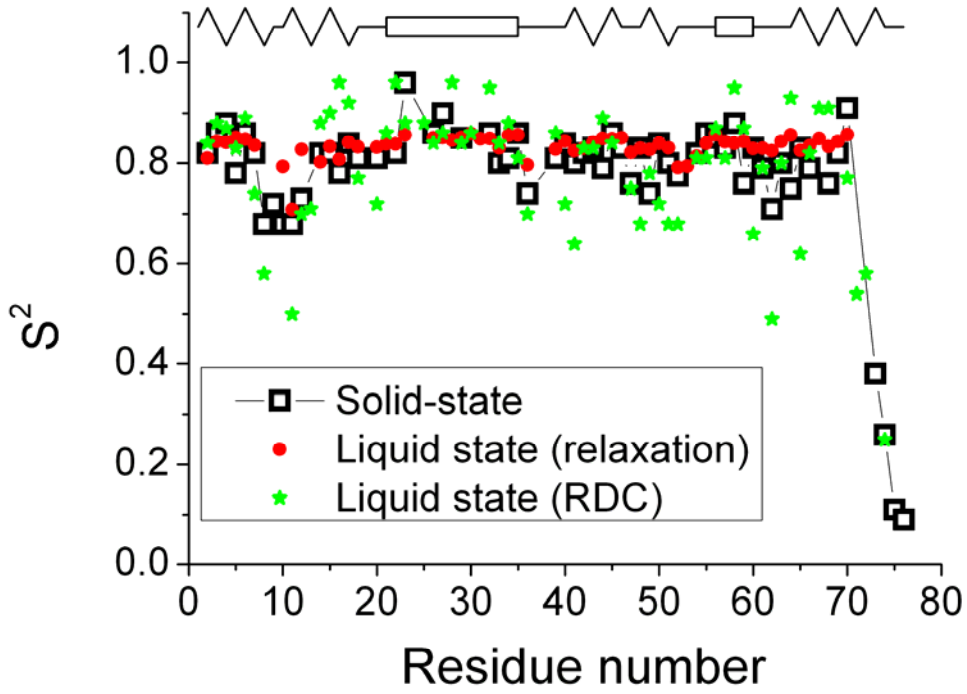

Figure S9: Solid- and liquid-state (determined from relaxation times and RDC) order parameters in ubiquitin.