Supplementary Material

CHARMM model	⊕ translation			⊖ translation			⊕ rotation			⊖ rotation		
	D	τ_0	t_{max}	D	τ_0	t_{max}	τ	β	τ_{avg}	τ	β	τ_{avg}
nonpol	2.16	318	215	0.72	1400	1240	478	0.68	625	55.8	0.43	153
no H, 0.1	2.26	299	250	0.86	955	1380	467	0.69	600	54.0	0.42	156
no H, 0.2	2.49	265	183	0.98	727	1360	457	0.69	585	50.9	0.43	144
no H, 0.3	2.76	200	218	1.05	854	1420	415	0.71	516	48.2	0.42	141
no H, 0.4	2.93	130	203	1.19	650	878	424	0.70	534	48.1	0.41	149
no H, 0.5	2.97	222	163	1.29	646	588	394	0.68	510	45.5	0.42	130
no H, 0.6	3.21	155	175	1.50	463	633	389	0.70	493	44.9	0.42	133
no H, 0.7	3.44	111	120	1.64	402	480	377	0.71	472	44.2	0.43	124
no H, 0.8	3.39	209	113	1.69	496	425	348	0.71	434	42.9	0.43	121
no H, 0.9	3.63	234	105	1.99	377	373	343	0.73	418	42.4	0.43	116
no H, 1.0	4.08	72.7	108	2.11	305	303	331	0.71	412	40.6	0.43	114
impl H, 0.1	2.40	231	158	1.01	684	1460	472	0.69	609	53.7	0.43	152
impl H, 0.2	2.52	259	275	0.94	1030	635	427	0.69	547	50.2	0.43	140
impl H, 0.3	2.77	204	190	1.24	599	605	408	0.71	510	45.8	0.42	133
impl H, 0.4	3.00	233	230	1.29	689	670	398	0.70	503	43.6	0.42	129
impl H, 0.5	3.51	140	150	1.69	354	900	361	0.71	448	40.8	0.42	117
impl H, 0.6	3.46	155	120	1.63	537	320	354	0.71	443	39.9	0.43	111
impl H, 0.7	3.88	131	128	1.98	352	800	328	0.71	410	37.6	0.42	107
impl H, 0.8	4.15	125	118	2.16	395	610	304	0.73	369	35.9	0.43	100
impl H, 0.9	4.23	172	113	2.20	452	168	285	0.74	345	33.6	0.43	92.4
impl H, 1.0	5.49	54.2	85.0	3.19	193	125	259	0.74	314	31.7	0.44	82.7

TABLE S1. MSD and $\langle \vec{\mu}(0) \cdot \vec{\mu}(t) \rangle / \langle \vec{\mu}(0)^2 \rangle$ fit parameters of all CHARMM simulations. D is given in units of 10^{-11} m^2 s^{-1}, τ_0, t_{max}, τ and τ_{avg} in units of ps. The first column lists the polarisability model and the respecitve strength of polarisabilities.

AMBER model	⊕ translation			⊖ translation			⊕ rotation			⊖ rotation		
	D	τ_0	t_{max}	D	τ_0	t_{max}	τ	β	τ_{avg}	τ	β	τ_{avg}
nonpol	2.24	282	185	0.76	1200	740	491	0.68	643	55.8	0.42	160
no H, 0.1	2.62	136	228	0.97	809	763	449	0.70	568	52.8	0.42	150
no H, 0.2	2.43	302	178	0.98	866	883	446	0.70	566	51.7	0.42	147
no H, 0.3	2.63	193	153	1.06	754	440	446	0.69	575	52.2	0.42	148
no H, 0.4	2.82	221	220	1.19	708	860	419	0.68	544	50.2	0.42	147
no H, 0.5	3.01	182	153	1.48	382	618	403	0.72	499	48.9	0.43	139
no H, 0.6	3.18	183	170	1.51	530	745	394	0.71	492	49.7	0.43	139
no H, 0.7	3.49	124	120	1.74	378	213	388	0.72	478	49.7	0.43	139
no H, 0.8	4.39	85.6	113	2.45	213	305	314	0.74	379	42.8	0.43	115
no H, 0.9	4.34	97.9	120	2.47	225	143	328	0.74	394	47.6	0.44	123
no H, 1.0	4.24	114	123	2.43	275	128	344	0.72	423	50.9	0.44	132
impl H, 0.1	2.55	232	253	1.02	671	1440	450	0.69	577	51.0	0.43	144
impl H, 0.2	2.56	255	173	1.02	865	1340	428	0.68	558	48.6	0.42	138
impl H, 0.3	2.80	221	160	1.26	550	628	419	0.70	527	47.8	0.43	135
impl H, 0.4	3.03	185	143	1.38	536	660	403	0.71	502	45.9	0.43	129
impl H, 0.5	3.22	153	175	1.51	514	550	383	0.73	466	44.3	0.42	130
impl H, 0.6	3.56	175	135	1.95	336	428	357	0.73	434	43.9	0.44	117
impl H, 0.7	4.19	121	133	2.35	261	328	322	0.73	393	40.8	0.43	111
impl H, 0.8	5.76	80.7	78.0	3.58	151	150	250	0.73	304	33.1	0.45	82.3
impl H, 0.9	5.62	145	75.0	3.55	213	100	252	0.75	299	35.2	0.45	86.1
impl H, 1.0	6.36	66.5	78.0	4.21	173	133	248	0.75	297	36.9	0.47	85.0
expl H, 0.1	2.57	205	201	1.05	743	1720	447	0.70	569	50.5	0.43	140
expl H, 0.2	2.76	213	205	1.20	634	910	409	0.69	522	45.7	0.43	127
expl H, 0.3	3.13	185	118	1.41	568	478	379	0.72	464	42.5	0.44	113
expl H, 0.4	3.58	125	140	1.85	326	355	348	0.72	431	39.9	0.44	104
expl H, 0.5	4.00	78.1	105	2.07	304	340	335	0.72	412	38.0	0.44	101
expl H, 0.6	4.26	116	100	2.38	301	253	314	0.73	380	36.0	0.44	94.0
expl H, 0.7	4.82	90.7	105	2.78	221	408	288	0.72	353	34.6	0.45	86.3
expl H, 0.8	5.97	69.3	95.0	3.66	161	163	237	0.75	283	29.0	0.46	68.7
expl H, 0.9	6.03	77.9	70.0	3.92	186	125	243	0.76	288	31.7	0.47	72.7
expl H, 1.0	6.29	92.5	80.0	4.11	199	68	242	0.76	284	33.1	0.47	76.0

TABLE S2. MSD and $\langle \vec{\mu}(0) \cdot \vec{\mu}(t) \rangle / \langle \vec{\mu}(0)^2 \rangle$ fit parameters of all AMBER simulations. D is given in units of 10^{-11} m^2 s^{-1}, τ_0, t_{max}, τ and τ_{avg} in units of ps. The first column lists the polarisability model and the respecitve strength of polarisabilities.

CHARMM model	$\oplus\oplus$			$\ominus\ominus$			$\oplus\ominus$		
	$D^{\oplus\oplus}$	$\tau_0^{\oplus\oplus}$	$D^\oplus + D^\oplus$	$D^{\ominus\ominus}$	$\tau_0^{\ominus\ominus}$	$D^\ominus + D^\ominus$	$D^{\oplus\ominus}$	$\tau_0^{\oplus\ominus}$	$D^\oplus + D^\ominus$
nonpol	4.48	267	4.33	1.55	1250	1.43	3.01	485	2.88
no H, 0.1	4.79	218	4.52	1.75	959	1.73	3.30	376	3.12
no H, 0.2	5.13	270	4.97	1.98	1010	1.96	3.60	429	3.47
no H, 0.3	5.42	189	5.52	2.08	867	2.11	3.74	356	3.81
no H, 0.4	5.77	189	5.86	2.32	841	2.38	4.05	349	4.12
no H, 0.5	5.78	201	5.94	2.46	623	2.57	4.12	318	4.25
no H, 0.6	6.54	173	6.41	2.94	502	3.00	4.73	269	4.71
no H, 0.7	6.77	117	6.88	3.35	307	3.28	5.07	174	5.08
no H, 0.8	6.95	189	6.79	3.53	397	3.38	5.24	256	5.08
no H, 0.9	7.38	220	7.26	3.90	442	3.98	5.63	296	5.62
no H, 1.0	7.92	96.2	8.17	4.16	284	4.23	6.05	155	6.20
impl H, 0.1	4.70	383	4.81	2.06	751	2.02	3.40	476	3.41
impl H, 0.2	5.00	278	5.03	1.89	1040	1.88	3.44	466	3.46
impl H, 0.3	5.57	202	5.54	2.49	590	2.48	4.04	309	4.01
impl H, 0.4	6.03	225	6.00	2.59	697	2.58	4.31	355	4.29
impl H, 0.5	6.99	150	7.01	3.33	329	3.39	5.15	207	5.20
impl H, 0.6	6.83	234	6.93	3.24	562	3.26	5.03	337	5.09
impl H, 0.7	7.69	152	7.77	3.97	362	3.95	5.83	221	5.86
impl H, 0.8	8.41	62.4	8.31	4.36	345	4.32	6.39	148	6.31
impl H, 0.9	8.60	90.4	8.45	4.49	334	4.39	6.55	168	6.42
impl H, 1.0	11.2	33.0	11.0	6.44	170	6.39	8.80	79.9	8.68

TABLE S3. Fit parameters for the pair displacement $\langle[\vec{R}_{ij}(t) - \vec{R}_{ij}(0)]^2\rangle$ for all CHARMM simulations. D is given in units of 10^{-11} m^2 s^{-1}, τ_0 in units of ps. The parameters show that each pair diffusion coefficient is roughly the sum of the respective single-particle diffusion coefficients of the two species.

AMBER	⊕⊕			⊖⊖			⊕⊖		
model	$D^{\oplus\oplus}$	$\tau_0^{\oplus\oplus}$	$D^\oplus + D^\oplus$	$D^{\ominus\ominus}$	$\tau_0^{\ominus\ominus}$	$D^\ominus + D^\ominus$	$D^{\oplus\ominus}$	$\tau_0\oplus\ominus$	$D^\oplus + D^\ominus$
nonpol	4.32	311	4.47	1.38	1450	1.52	2.85	2.99	545
no H, 0.1	5.07	155	5.25	1.83	1030	1.94	3.45	3.59	353
no H, 0.2	4.71	241	4.86	1.80	901	1.97	3.26	3.41	401
no H, 0.3	5.09	219	5.26	2.08	701	2.12	3.58	3.69	348
no H, 0.4	5.59	182	5.63	2.30	784	2.38	3.95	4.01	338
no H, 0.5	5.92	229	6.03	2.96	417	2.97	4.44	4.50	288
no H, 0.6	6.45	177	6.36	3.00	543	3.01	4.72	4.68	285
no H, 0.7	6.98	96.8	6.97	3.34	379	3.48	5.16	5.23	183
no H, 0.8	8.73	85.2	8.78	4.85	211	4.90	6.78	6.84	130
no H, 0.9	8.48	124	8.67	4.79	263	4.94	6.64	6.80	171
no H, 1.0	8.29	118	8.47	4.69	306	4.86	6.49	6.67	182
impl H, 0.1	4.87	287	5.11	1.84	928	2.03	3.35	3.57	446
impl H, 0.2	5.00	281	5.12	1.91	1070	2.04	3.45	3.58	476
impl H, 0.3	5.44	259	5.60	2.43	604	2.52	3.93	4.06	362
impl H, 0.4	5.71	185	6.05	2.60	563	2.77	4.16	4.41	295
impl H, 0.5	6.20	159	6.44	3.03	452	3.02	4.62	4.73	249
impl H, 0.6	7.01	164	7.13	3.88	272	3.89	5.44	5.51	203
impl H, 0.7	8.07	123	8.39	4.45	286	4.69	6.26	6.54	179
impl H, 0.8	11.3	97.5	11.5	6.75	199	7.16	9.00	9.34	134
impl H, 0.9	11.1	144	11.3	7.02	176	7.10	9.04	9.17	158
impl H, 1.0	12.9	65.0	12.7	8.56	169	8.42	10.7	10.6	106
expl H, 0.1	4.76	329	5.14	1.86	942	2.11	3.32	3.62	485
expl H, 0.2	5.63	198	5.53	2.41	651	2.39	4.02	3.96	324
expl H, 0.3	6.32	175	6.26	2.70	661	2.82	4.50	4.54	309
expl H, 0.4	7.10	172	7.15	3.60	351	3.70	5.34	5.43	230
expl H, 0.5	7.85	86.1	8.00	4.03	307	4.13	5.94	6.07	155
expl H, 0.6	8.30	120	8.53	4.64	330	4.75	6.46	6.64	192
expl H, 0.7	9.21	106	9.65	5.43	243	5.56	7.31	7.60	156
expl H, 0.8	11.9	76.1	12.0	7.18	170	7.31	9.50	9.63	111
expl H, 0.9	12.3	42.9	12.1	7.95	135	7.84	10.1	9.95	78.2
expl H, 1.0	12.3	113	12.6	8.22	187	8.22	10.3	10.4	141

TABLE S4. Fit parameters for the pair displacement $\langle[\vec{R}_{ij}(t) - \vec{R}_{ij}(0)]^2\rangle$ for all AMBER simulations. D is given in units of 10^{-11} m^2 s^{-1}, τ_0 in units of ps. The parameters show that each pair diffusion coefficient is roughly the sum of the respective single-particle diffusion coefficients of the two species.

CHARMM	⊕⊕					⊖⊖							⊕⊖				
model	$\langle CN\rangle$	τ	β	a_0	τ_{avg}	$\langle CN\rangle$	a_1	τ_1	τ_2	β	a_0	τ_{avg}	$\langle CN\rangle$	τ	β	a_0	τ_{avg}
nonpol	10.1	3350	0.57	0.00	5450	4.11	1.45	9.97	5760	0.47	0.00	8490	7.05	5410	0.77	0.45	6310
no H, 0.1	10.1	3290	0.57	0.00	5370	4.12	1.45	10.0	5240	0.49	0.00	7190	7.07	5330	0.78	0.40	6140
no H, 0.2	10.1	2930	0.57	0.01	4750	4.12	1.45	10.0	4680	0.50	0.00	6070	7.21	4960	0.77	0.32	5780
no H, 0.3	10.1	2660	0.57	0.04	4290	4.11	1.46	10.0	4260	0.49	0.00	5620	7.32	4570	0.76	0.24	5380
no H, 0.4	10.1	2480	0.57	0.10	3980	4.10	1.45	10.3	4090	0.51	0.00	5150	7.11	4290	0.79	0.40	4920
no H, 0.5	10.1	2320	0.57	0.13	3790	4.09	1.46	10.4	3740	0.51	0.00	4700	7.33	4160	0.75	0.27	4960
no H, 0.6	10.0	2160	0.57	0.15	3490	4.09	1.30	10.3	2780	0.48	0.00	4060	7.40	3850	0.74	0.28	4630
no H, 0.7	10.0	2070	0.58	0.11	3270	4.08	1.34	10.9	2600	0.50	0.03	3510	7.24	3560	0.79	0.32	4070
no H, 0.8	9.97	1930	0.56	0.12	3170	4.07	1.43	12.1	2810	0.52	0.00	3400	7.27	3330	0.77	0.34	3890
no H, 0.9	9.96	1900	0.56	0.09	3160	4.06	1.18	9.16	2000	0.48	0.01	3040	7.29	3090	0.75	0.38	3660
no H, 1.0	9.94	1730	0.57	0.15	2770	4.04	1.42	12.5	2390	0.53	0.00	2840	7.34	2980	0.76	0.32	3490
impl H, 0.1	10.1	3071	0.58	0.00	4836	4.12	1.37	10.3	4342	0.47	0.00	6446	7.40	5433	0.78	0.08	6288
impl H, 0.2	10.1	2888	0.59	0.00	4466	4.12	1.36	10.1	4452	0.45	0.00	7258	7.53	5507	0.75	0.00	6580
impl H, 0.3	10.1	2645	0.58	0.00	4202	4.12	1.39	10.3	3743	0.48	0.00	5452	6.99	4064	0.79	0.54	4659
impl H, 0.4	10.0	2469	0.56	0.00	4107	4.12	1.40	10.3	3391	0.50	0.00	4497	7.67	4444	0.71	0.00	5526
impl H, 0.5	10.0	2214	0.57	0.00	3577	4.12	1.37	10.4	3065	0.49	0.00	4329	7.64	3967	0.73	0.00	4812
impl H, 0.6	9.99	2132	0.56	0.00	3507	4.10	1.41	11.1	2749	0.51	0.00	3445	7.36	3414	0.75	0.30	4078
impl H, 0.7	9.96	1944	0.56	0.00	3202	4.11	1.41	11.3	2429	0.53	0.00	2934	7.14	2978	0.78	0.47	3437
impl H, 0.8	9.93	1790	0.56	0.00	2972	4.11	1.40	11.6	2191	0.53	0.00	2598	7.27	2779	0.77	0.37	3239
impl H, 0.9	9.90	1644	0.55	0.04	2774	4.09	1.41	11.7	2083	0.53	0.00	2489	7.38	2639	0.74	0.33	3172
impl H, 1.0	9.87	1250	0.57	0.20	2005	4.10	1.28	9.81	1377	0.53	0.03	1718	7.59	2196	0.74	0.21	2647

TABLE S5. Fit parameters for the cage relaxation function $\langle n(0)n(t)\rangle$ for all CHARMM simulations. $\langle CN\rangle$ is the average number of particles of particles in the first solvation shell and a_0 is the steady-state value this function is approaching in the asymptotic limit. τ and τ_{avg} are given in units of ps. τ_{avg} describes the mean residence time of a particle in the first solvation shell of another particle.

AMBER model	⊕⊕ ⟨CN⟩	τ	β	a₀	τ_avg	⊖⊖ ⟨CN⟩	a₁	τ₁	τ₂	β	a₀	τ_avg	⊕⊖ ⟨CN⟩	τ	β	a₀	τ_avg
nonpol	10.2	3140	0.57	0.14	5130	4.10	1.49	9.59	6190	0.49	a0	8200	7.28	5750	0.74	0.28	6920
no H, 0.1	10.1	3060	0.57	0.00	4890	4.08	1.45	10.1	4920	0.49	0.00	6710	7.30	5080	0.75	0.28	6060
no H, 0.2	10.1	2800	0.56	0.13	4680	4.08	1.46	10.2	4730	0.49	0.00	6260	7.16	5030	0.76	0.35	5910
no H, 0.3	10.1	2520	0.58	0.21	3990	4.06	1.48	10.6	4580	0.49	0.00	6030	7.23	4640	0.76	0.35	5460
no H, 0.4	10.1	2460	0.57	0.11	3930	4.05	1.46	11.0	3960	0.51	0.00	4890	7.33	4350	0.75	0.27	5150
no H, 0.5	10.0	2390	0.57	0.09	3880	4.04	1.33	11.6	3010	0.49	0.00	4200	7.43	4140	0.73	0.24	5030
no H, 0.6	10.0	2190	0.57	0.13	3560	4.02	1.45	12.2	3260	0.51	0.02	4020	7.45	3890	0.73	0.23	4720
no H, 0.7	9.99	2020	0.57	0.17	3260	4.00	1.32	13.8	2640	0.49	0.00	3660	7.33	3520	0.76	0.33	4160
no H, 0.8	9.95	1600	0.58	0.17	2530	4.00	1.24	11.2	1870	0.50	0.00	2580	7.48	2830	0.75	0.22	3360
no H, 0.9	9.92	1630	0.57	0.15	2640	3.98	1.26	10.0	1690	0.52	0.00	2150	7.34	2780	0.77	0.32	3230
no H, 1.0	9.90	1600	0.57	0.20	2610	3.96	1.09	8.31	1490	0.49	0.06	2270	7.54	2800	0.74	0.24	3380
impl H, 0.1	10.1	2900	0.56	0.04	4740	4.09	1.46	10.0	4770	0.48	0.04	6610	7.21	4890	0.76	0.33	5760
impl H, 0.2	10.1	2650	0.56	0.14	4370	4.09	1.46	9.99	4440	0.49	0.00	5840	7.11	4650	0.76	0.43	5460
impl H, 0.3	10.1	2660	0.55	0.02	4520	4.08	1.46	10.6	3850	0.50	0.00	4940	7.23	4360	0.76	0.34	5120
impl H, 0.4	10.1	2350	0.56	0.10	3930	4.07	1.44	10.7	3570	0.51	0.00	4400	7.30	4090	0.75	0.31	4880
impl H, 0.5	10.0	2240	0.57	0.11	3640	4.06	1.44	11.2	3260	0.52	0.00	3970	7.23	3850	0.78	0.32	4420
impl H, 0.6	9.98	1920	0.57	0.13	3110	4.06	1.34	10.7	2100	0.51	0.00	2710	7.38	3330	0.76	0.25	3900
impl H, 0.7	9.94	1680	0.56	0.12	2740	4.06	1.31	10.5	1820	0.51	0.06	2350	7.53	2910	0.73	0.21	3530
impl H, 0.8	9.90	1190	0.58	0.16	1890	4.05	1.20	8.88	1170	0.51	0.05	1610	7.60	2060	0.73	0.20	2500
impl H, 0.9	9.86	1150	0.56	0.19	1880	4.05	1.09	7.93	1080	0.49	0.04	1640	7.67	1970	0.71	0.21	2450
impl H, 1.0	9.81	1060	0.57	0.18	1700	4.04	0.93	3.00	850	0.48	0.03	1410	7.76	1790	0.71	0.17	2220
expl H, 0.1	10.1	2780	0.57	0.12	4490	4.10	1.47	10.0	4620	0.49	0.02	6280	7.18	4760	0.77	0.36	5560
expl H, 0.2	10.1	2720	0.56	0.01	4480	4.10	1.45	10.3	4070	0.51	0.00	5140	7.42	4640	0.74	0.19	5600
expl H, 0.3	10.0	2290	0.56	0.08	3740	4.11	1.46	10.5	3440	0.51	0.00	4260	7.32	3890	0.75	0.31	4630
expl H, 0.4	10.0	1960	0.57	0.16	3150	4.11	1.45	11.0	2860	0.52	0.00	3480	7.25	3460	0.78	0.33	4000
expl H, 0.5	9.97	1870	0.57	0.09	3020	4.11	1.29	10.7	2140	0.50	0.00	2990	7.45	3170	0.75	0.25	3790
expl H, 0.6	9.93	1620	0.57	0.16	2590	4.12	1.19	9.58	1820	0.49	0.02	2720	7.41	2820	0.76	0.29	3330
expl H, 0.7	9.88	1400	0.57	0.19	2260	4.12	1.14	9.07	1520	0.48	0.00	2360	7.51	2500	0.75	0.23	2980
expl H, 0.8	9.83	1150	0.57	0.14	1880	4.13	1.22	9.73	1180	0.52	0.01	1580	7.67	1970	0.72	0.20	2420
expl H, 0.9	9.79	1110	0.57	0.16	1800	4.13	1.09	8.01	1010	0.50	0.03	1490	7.71	1930	0.72	0.18	2380
expl H, 1.0	9.73	1040	0.56	0.16	1720	4.14	0.96	7.01	920	0.49	0.03	1490	7.80	1780	0.71	0.19	2240

TABLE S6. Fit parameters for the cage relaxation function $\langle n(0)n(t)\rangle$ for all AMBER simulations. $\langle CN\rangle$ is the average number of particles of particles in the first solvation shell and a_0 is the steady-state value this function is approaching in the asymptotic limit. τ and τ_{avg} are given in units of ps. τ_{avg} describes the mean residence time of a particle in the first solvation shell of another particle.