Supplementary Information

Hydrogen Bond Dynamics of Water in Presence of an Amphiphile, Tetramethylurea: Signature of Confinement-induced Effects

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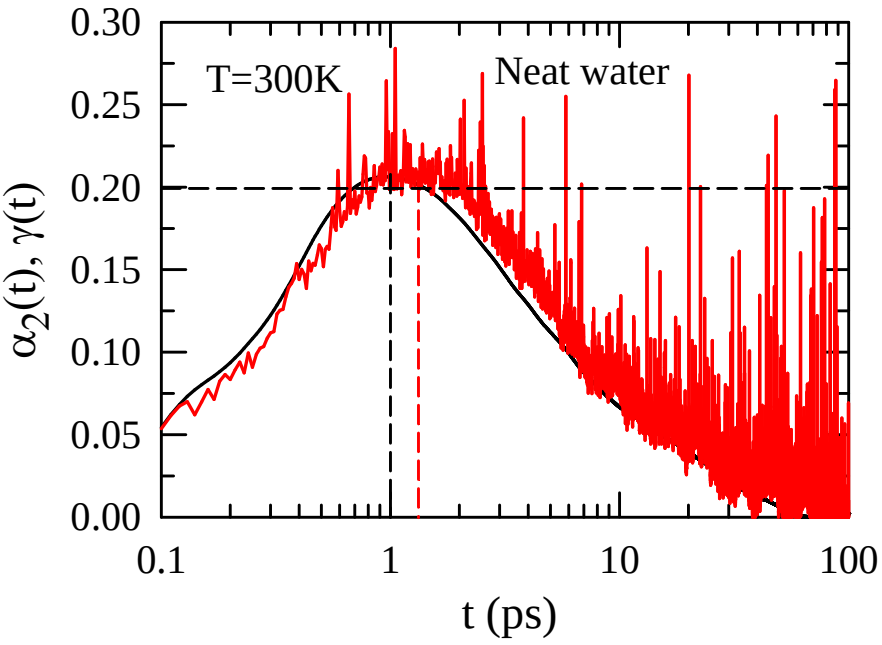

Fig. S1: Simulated $\alpha_2(t)$ and $\gamma(t)$ for neat water at 300 K. Black curve denotes $\alpha_2(t)$ and the red curve $\gamma(t)$. Vertical broken lines indicate the peak-times, τ_{NG} and τ_{NNG}, for $\alpha_2(t)$ and $\gamma(t)$, respectively. Horizontal line indicates the value of 0.2.

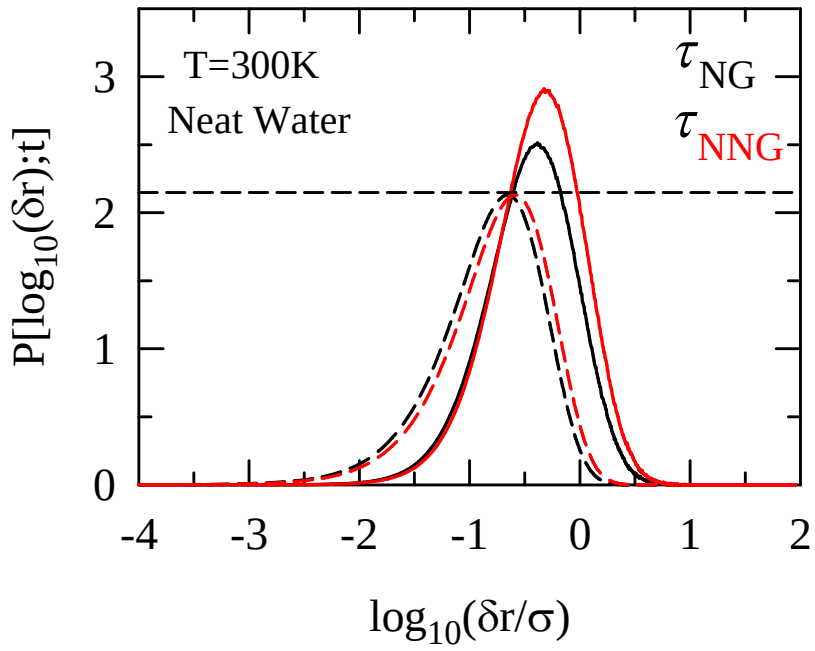

Fig. S2: Simulated single particle displacement distributions, $P[\log_{10}(\delta r);t)]$, for neat water at 300 K. Distributions both at τ_{NG} and τ_{NNG} are shown. $P[\log_{10}(\delta r);t)]$ corresponding to a Gaussian $G_s(\delta r,t)$ at these times are also shown for a comparison. Horizontal broken lines in these panels indicate a value of 2.13 expected for $P[\log_{10}(\delta r);t)]$ possessing a Gaussian $G_s(\delta r,t)$.

Table 1: Bi-exponential fit parameters for water-water H-bond lifetime correlation ($S_{HB}^{w-w}(t)$) decays at different TMU mole fractions and 300 K. Corresponding $\langle h \rangle$ values are listed in the last column.

X_{TMU}	a_1	τ_1/ps	a_2	τ_2/ps	τ_{av}/ps	$\langle h \rangle$
0.00	0.77	0.711	0.23	0.131	0.578	0.87
0.01	0.77	0.734	0.23	0.131	0.595	0.87
0.03	0.75	0.789	0.25	0.149	0.629	0.86
0.05	0.72	0.831	0.28	0.167	0.645	0.84
0.07	0.67	0.948	0.33	0.204	0.702	0.83
0.10	0.65	1.016	0.35	0.219	0.737	0.82
0.20	0.60	1.222	0.40	0.261	0.838	0.76
0.30	0.57	1.549	0.43	0.312	1.017	0.68
0.50	0.59	2.174	0.41	0.402	1.447	0.54
0.75	0.65	2.671	0.35	0.476	1.903	0.40

Table 2: Bi-exponential fit parameters for water-TMU H-bond lifetime correlation ($S_{HB}^{w-TMU}(t)$)decays at different TMU mole fractions and 300 K. Corresponding $\langle h \rangle$ values are listed in the last column.

X_{TMU}	a_1	τ_1/ps	a_2	τ_2/ps	τ_{av}/ps	$\langle h \rangle$
0.01	0.65	0.969	0.35	0.264	0.722	0.13
0.03	0.72	1.194	0.28	0.260	0.932	0.14
0.05	0.65	1.399	0.35	0.301	1.015	0.14
0.07	0.66	1.320	0.34	0.325	0.982	0.17
0.10	0.64	1.466	0.36	0.355	1.066	0.18
0.20	0.58	1.912	0.42	0.421	1.286	0.27
0.30	0.59	2.218	0.41	0.485	1.507	0.36
0.50	0.65	2.840	0.35	0.548	2.038	0.54
0.75	0.67	3.264	0.33	0.562	2.372	0.73

Table 3: Tri-exponential fit parameters for water-water H-bond structural relaxations ($C_{HB}^{w-w}(t)$) at different TMU mole fractions and 300 K. Corresponding $\langle h \rangle$ values are listed in the last column.

X_{TMU}	a_1	τ_1/ps	a_2	τ_2/ps	a_3	τ_3/ps	τ_{av}/ps	$\langle h \rangle$
0.00	0.05	22.233	0.79	3.766	0.16	0.173	4.114	0.87
0.01	0.08	19.036	0.76	3.784	0.16	0.173	4.426	0.87
0.03	0.10	21.853	0.74	4.608	0.16	0.217	5.630	0.86
0.05	0.11	23.790	0.71	5.188	0.18	0.269	6.349	0.84
0.07	0.14	27.318	0.67	6.398	0.19	0.348	8.177	0.83
0.10	0.15	31.730	0.66	7.025	0.19	0.368	9.466	0.82
0.20	0.16	55.025	0.63	11.603	0.21	0.675	16.256	0.76
0.30	0.20	88.626	0.59	18.829	0.21	1.015	29.047	0.68
0.50	0.18	184.439	0.64	36.734	0.18	1.967	57.063	0.54
0.75	0.37	170.567	0.54	36.240	0.09	0.650	82.738	0.40

Table 4: Tri-exponential fitting parameters for Wat-TMU H-bond structural relaxations ($C_{HB}^{w-TMU}(t)$) at different TMU mole fractions and 300 K. Corresponding $\langle h \rangle$ values are listed in the last column.

X_{TMU}	a_1	τ_1/ps	a_2	τ_2/ps	a_3	τ_3/ps	τ_{av}/ps	$\langle h \rangle$
0.01	0.27	20.611	0.63	6.576	0.10	0.141	9.722	0.13
0.03	0.18	34.700	0.68	10.435	0.14	0.550	13.419	0.14
0.05	0.19	43.383	0.67	11.226	0.14	0.535	15.839	0.14
0.07	0.15	66.284	0.68	15.150	0.17	1.064	20.425	0.17
0.10	0.31	41.225	0.57	12.858	0.12	0.463	20.164	0.18
0.20	0.30	87.040	0.58	22.048	0.12	0.714	38.985	0.27
0.30	0.38	121.042	0.49	30.768	0.13	1.332	61.245	0.36
0.50	0.62	143.735	0.28	39.289	0.10	1.748	100.291	0.54
0.75	0.66	166.667	0.28	63.878	0.06	0.784	127.933	0.73