Supporting Information: Temperature-Dependence of the Dielectric Relaxation of Water using Non-Polarizable Water Models

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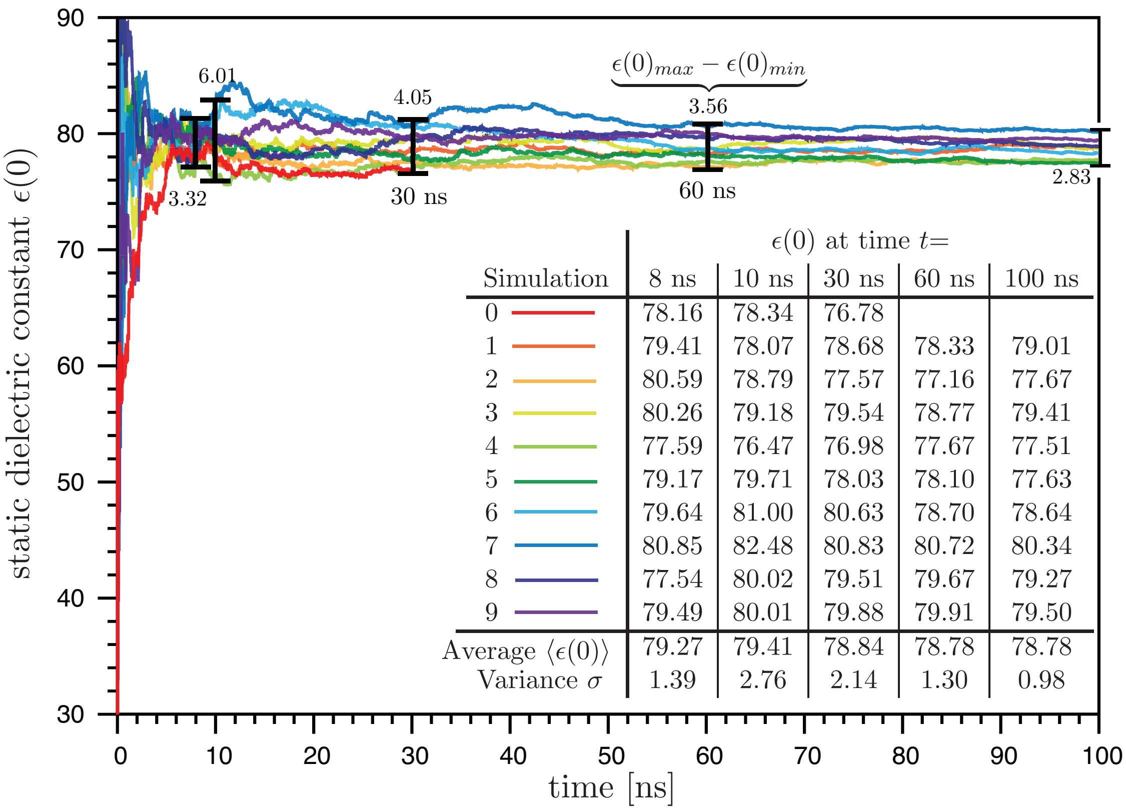
Table S1: Calculated Kirkwood factors, rate constant for the hydrogen-bond breaking, and HB-bond energetics of water at 298K (19 non-polarizable water models).

Water Model	Kirkwood factors g_K	G_K	Hydrogen-bond network dynamics τ_{HB}[ps]	k_b[ps^{-1}]	$n_b^{cycle}(\nu_D)$	ΔG[kcal/mol]
			three-point models			
SPC	2.515	3.744	6.003	0.167	4.94	2.144
SPC/E	2.499	3.722	6.888	0.145	5.17	2.226
SPC/E$_\mathrm{B}$	2.468	3.676	8.742	0.114	5.36	2.367
SPC/FW	2.794	4.166	7.417	0.135	5.66	2.270
SPC-DC	2.693	4.015	6.306	0.159	4.97	2.173
TIP3P	3.605	5.381	3.658	0.273	5.88	1.851
TIP3PF	3.278	4.891	4.959	0.202	6.17	2.031
TIP3P-FB	2.678	3.993	7.670	0.130	5.93	2.289
H_2O-DC	2.670	3.980	7.340	0.136	5.37	2.263
OPC3	2.655	3.958	7.539	0.133	4.99	2.279
			four-point models			
OPC	2.519	3.755	7.354	0.136	4.87	2.264
TIP4P	2.127	3.160	5.353	0.187	3.91	2.076
TIP4PEW	2.345	3.491	7.160	0.140	4.76	2.249
TIP4P-FB	2.545	3.793	8.493	0.118	4.59	2.350
TIP4P2005	2.121	3.154	7.966	0.126	4.31	2.312
TIP4Q	2.657	3.961	7.630	0.131	5.58	2.286
TIP4P/ϵ	2.618	3.902	7.986	0.125	5.32	2.313
			five-point models			
TIP5P	3.485	5.199	6.490	0.154	6.93	2.190
TIP5PEW	3.646	5.441	6.173	0.162	6.76	2.161



	$\epsilon(0)$ at time t=				
Simulation	8 ns	10 ns	30 ns	60 ns	100 ns
0 —	78.16	78.34	76.78		
1 —	79.41	78.07	78.68	78.33	79.01
2 —	80.59	78.79	77.57	77.16	77.67
3 —	80.26	79.18	79.54	78.77	79.41
4 —	77.59	76.47	76.98	77.67	77.51
5 —	79.17	79.71	78.03	78.10	77.63
6 —	79.64	81.00	80.63	78.70	78.64
7 —	80.85	82.48	80.83	80.72	80.34
8 —	77.54	80.02	79.51	79.67	79.27
9 —	79.49	80.01	79.88	79.91	79.50
Average $\langle\epsilon(0)\rangle$	79.27	79.41	78.84	78.78	78.78
Variance σ	1.39	2.76	2.14	1.30	0.98

Figure S1: Slow convergence of the static dielectric constant (illustrated for the TIP4P-FB water model at 298 K). The results presented in tables are averages of the dielectric properties over last nanosecond of 30 ns simulations.

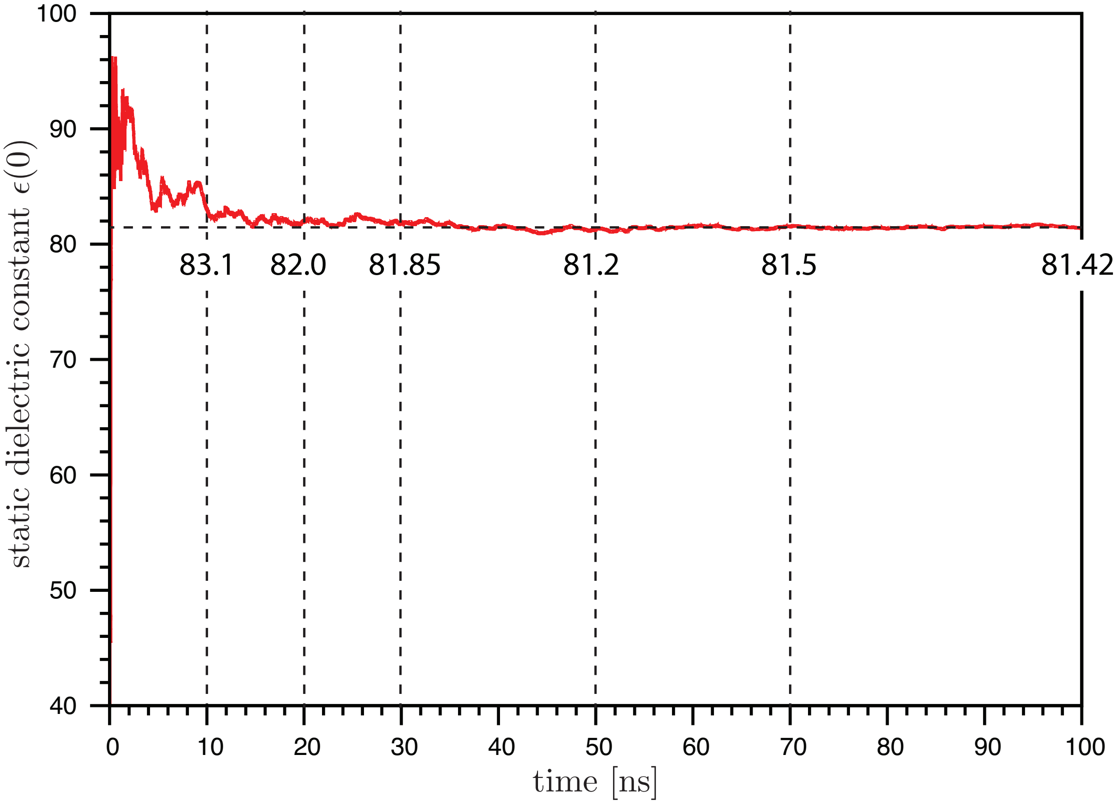
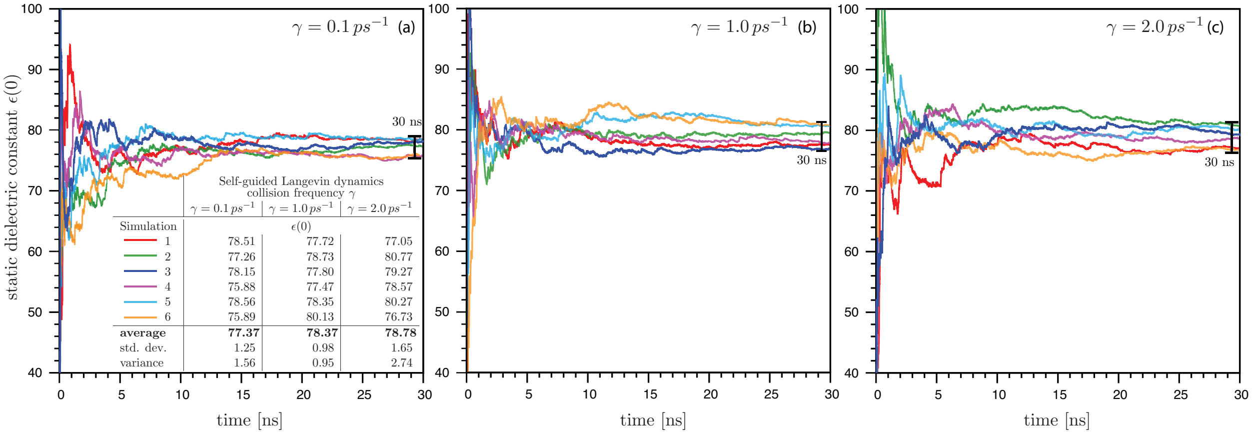


Figure S2: Slow convergence of the static dielectric constant (illustrated for the TIP3P-FB water model at 298 K).



	Self-guided Langevin dynamics collision frequency γ		
	$\gamma = 0.1\,ps^{-1}$	$\gamma = 1.0\,ps^{-1}$	$\gamma = 2.0\,ps^{-1}$
Simulation	$\epsilon(0)$		
🟥 1	78.51	77.72	77.05
🟩 2	77.26	78.73	80.77
🟦 3	78.15	77.80	79.27
🟪 4	75.88	77.47	78.57
🟦 5	78.56	78.35	80.27
🟧 6	75.89	80.13	76.73
average	**77.37**	**78.37**	**78.78**
std. dev.	1.25	0.98	1.65
variance	1.56	0.95	2.74

Figure S3: Stochastic variation in ϵ in Langevin dynamics for varying collision frequency γ in Langevin thermostat (TIP4P-FB water model): (a) 0.1 ps^{-1}, (b) 1.0 ps^{-1}, (c) 2.0 ps^{-1}.

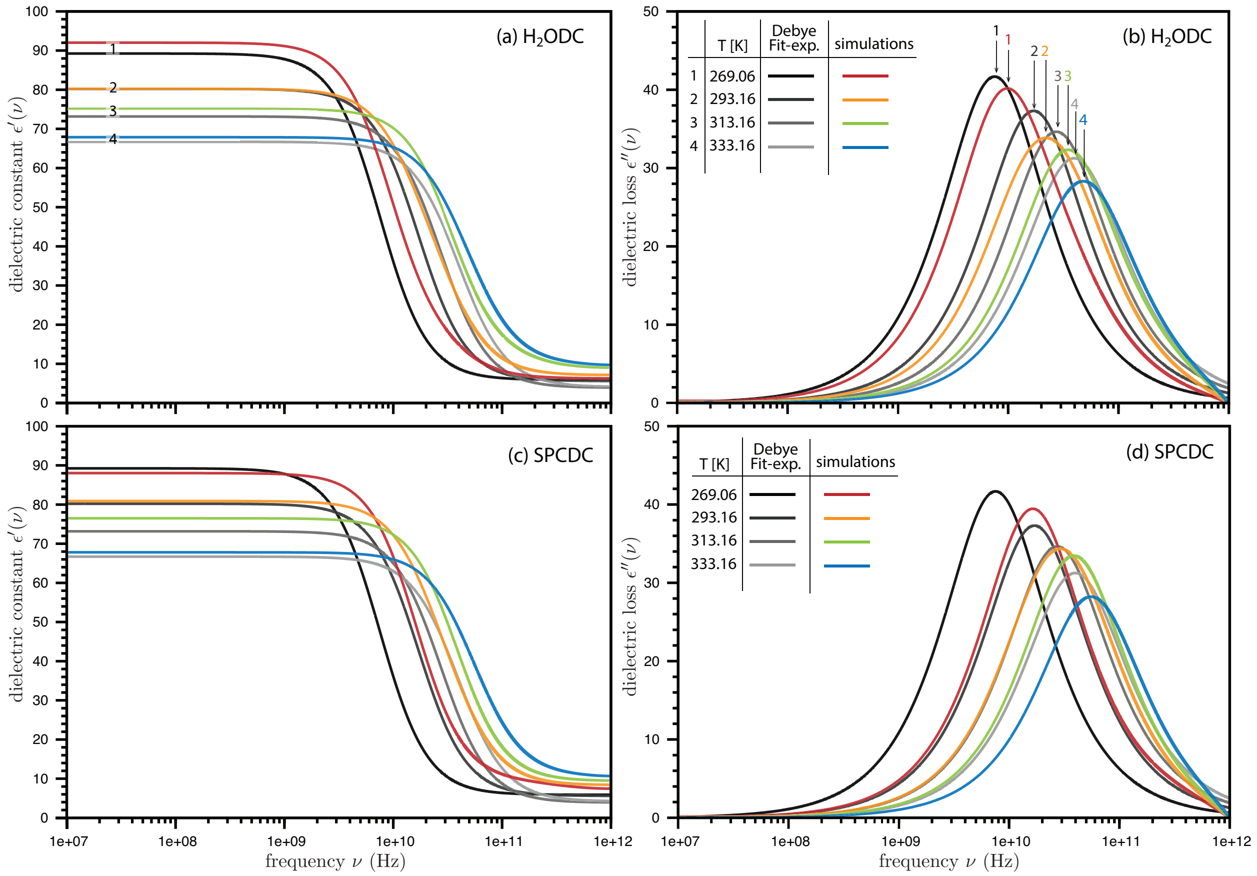


Figure S4: Comparison of the Debye function fit to the experimental data and the simulated dielectric spectra obtained for H$_2$ODC and SPCDC water models.[S1] The difference between experimental and simulated static dielectric constants are within the simulation error (2-3%). The simulates predict the water relaxation is simulated at higher frequencies and shorter relaxation times (τ_D) than the Debye-function fitted to the experimental data.

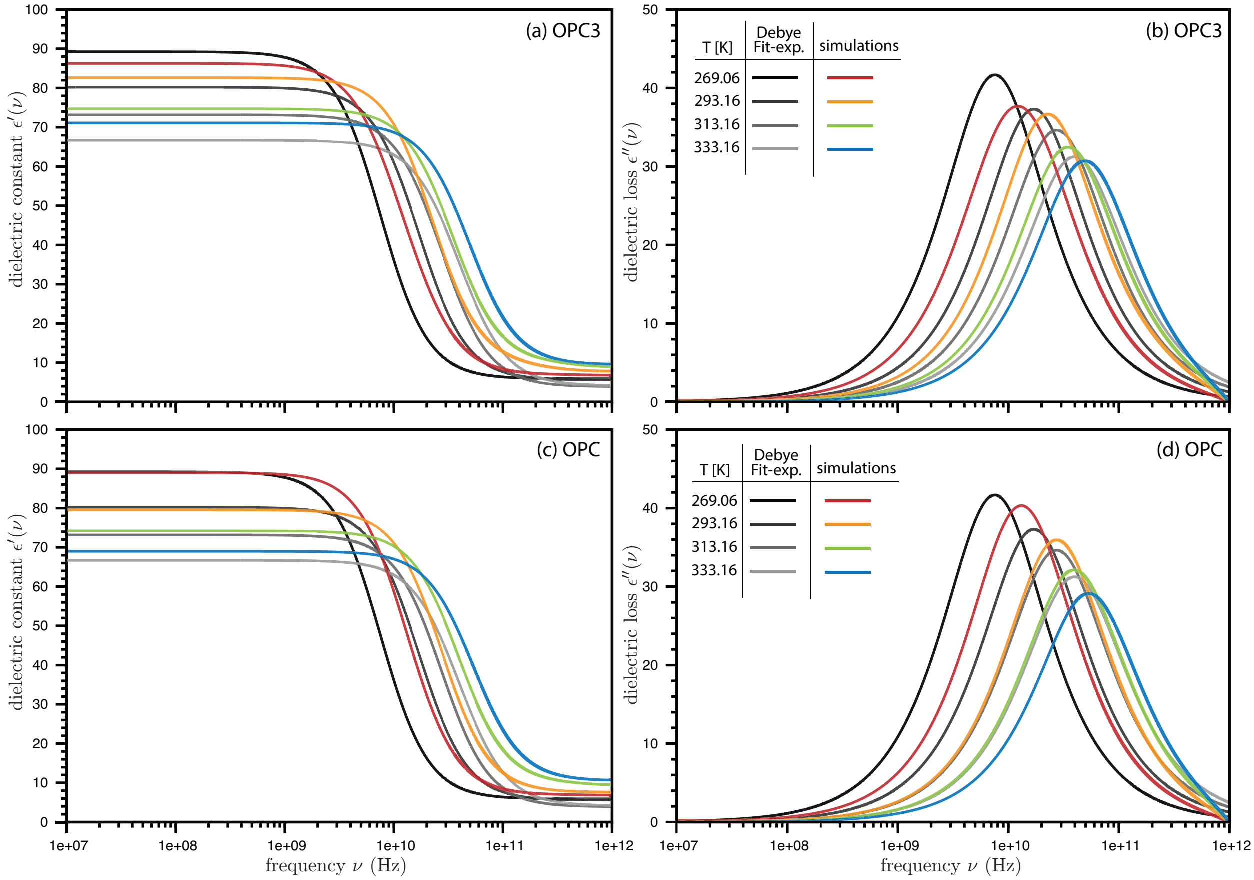


Figure S5: Comparison of the Debye function fit to the experimental data and the simulated dielectric spectra obtained for OPC3 and OPC water models.[S2,S3] The difference between experimental and simulated static dielectric constants are within the simulation error (2-3%). The simulates predict the water relaxation is simulated at higher frequencies and shorter relaxation times (τ_D) than the Debye-function fitted to the experimental data.

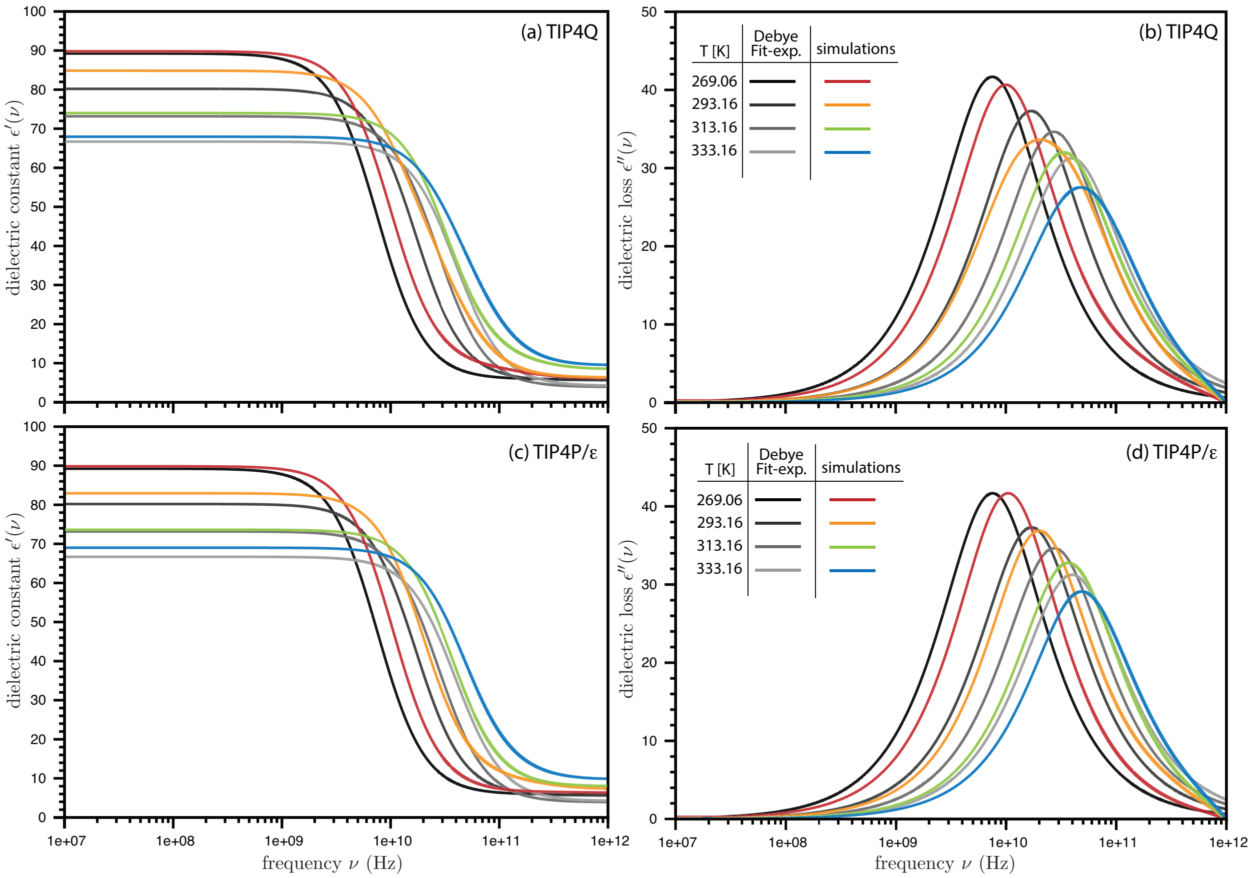


Figure S6: Comparison of the Debye function fit to the experimental data and the simulated dielectric spectra obtained for TIP3P-FB and TIP4P-FB water models.[S4] The difference between experimental and simulated static dielectric constants are within the simulation error (2-3%). The simulates predict the water relaxation is simulated at higher frequencies and shorter relaxation times (τ_D) than the Debye-function fitted to the experimental data.

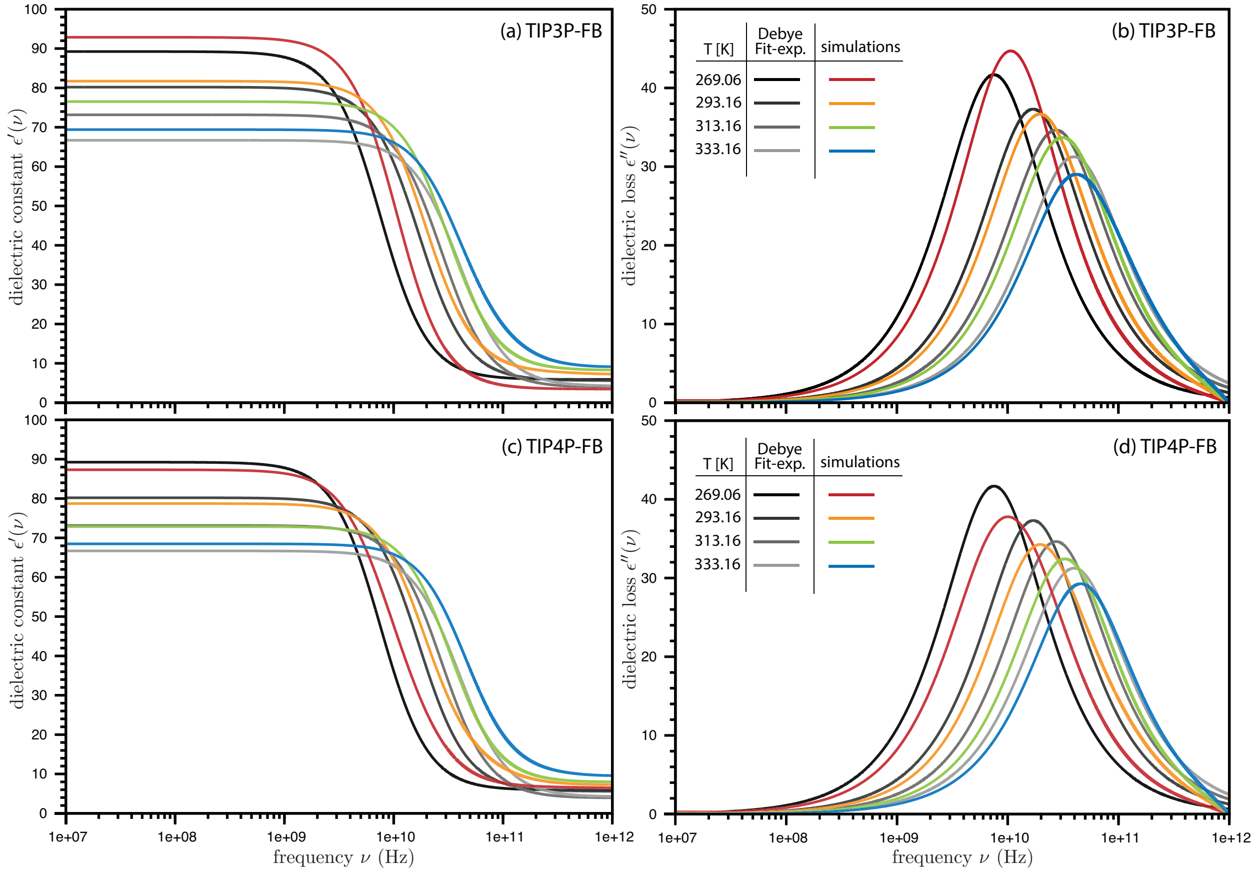


Figure S7: Comparison of the Debye function fit to the experimental data and the simulated dielectric spectra obtained for TIP4Q and TIP4P/ϵ water models.[S5,S6] The difference between experimental and simulated static dielectric constants are within the simulation error (2-3%). The simulates predict the water relaxation is simulated at higher frequencies and shorter relaxation times (τ_D) than the Debye-function fitted to the experimental data.

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