Supplementary Information

Understanding Dielectric Loss in Water via Distance-Dependent Dipole Correlation Functions

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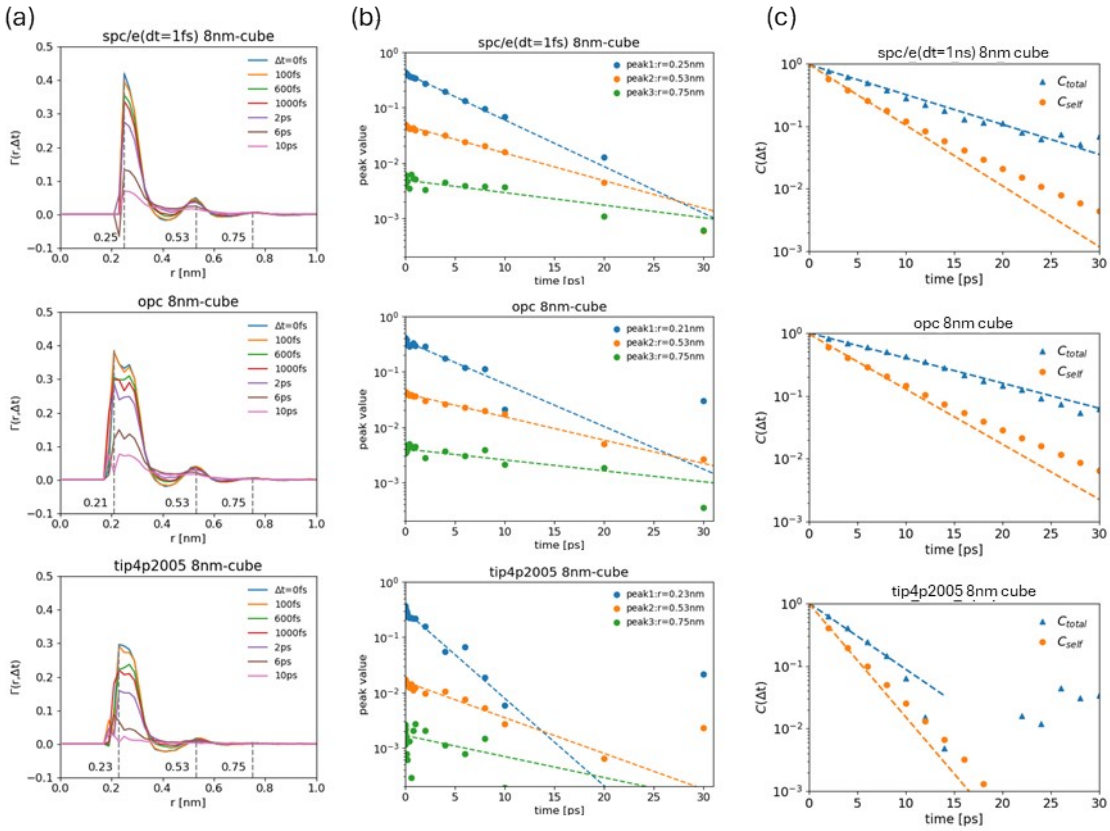

Fig. S1 (a) Distance-dependent dipole cross-correlation functions $\Gamma(r, \Delta t)$, (b) time evolution of the peak heights corresponding to the first (r = 0.25 nm), second (r = 0.53 nm), and third (r = 0.75 nm) correlation peaks, and (c) dipole moment autocorrelation functions C_{total} and C_{self} for SPC/E, OPC, and TIP4P/2005 water models. Fitting curves were obtained using data for which the correlation remains positive. All simulations were performed using an 8-nm cubic simulation box. Simulation time steps were 2 fs for TIP4P/2005 and OPC, while a reduced time step of 1 fs was applied for SPC/E. Dashed lines indicate single-exponential fits used to extract decay times.

Table S1 Cross correlation decay times, $\tau_{cross,1}$, $\tau_{cross,2}$ and $\tau_{cross,3}$, corresponding to the first, second and third peaks of $\Gamma(r, \Delta t)$ for SPC/E, OPC, and TIP4P/2005 water models. All simulations were performed using 8 nm cubic boxes, and simulation time steps were 2 fs for TIP4P/2005 and OPC, and 1 fs for SPC/E.

model	$\tau_{cross,1}$ [ps] (0.25 nm)	$\tau_{cross,2}$ [ps] (0.53 nm)	$\tau_{cross,3}$ [ps] (0.75 nm)
SPC/E (dt=1fs)	5.15 ± 0.16	8.71 ± 0.45	19.14 ± 4.05
OPC	5.62 ± 0.66	10.31 ± 0.44	22.01 ± 4.64
TIP4P/2005	2.75 ± 0.45	6.71 ± 0.82	11.4 ± 0.75

Table S2 Characteristic frequencies f=ω/2π (GHz) corresponding to the inverse decay times of distance-resolved dipole-dipole correlations (ωτ=1) for SPC/E, OPC, and TIP4P/2005 water models, shown as order-of-magnitude timescale or frequency indicators. All simulations were performed using 8 nm cubic boxes, and simulation time steps were 2 fs for TIP4P/2005 and OPC, and 1 fs for SPC/E.

model	$f_{cross,1}$ (0.25nm)	$f_{cross,2}$ (0.53nm)	$f_{cross,3}$ (0.75nm)
SPC/E (dt=1fs)	30.9	18.3	8.3
OPC	28.3	15.4	7.2
TIP4P/2005	57.9	23.7	14.0

Table S3 Total and self correlation decay times, τ_{total} and τ_{self} for SPC/E, OPC, and TIP4P/2005 water models. All simulations were performed using 8 nm cubic boxes, and simulation time steps were 2 fs for TIP4P/2005 and OPC, and 1 fs for SPC/E.

model	τ_{total} [ps]	τ_{self} [ps]
SPC/E (dt=1fs)	9.09 ± 0.80	4.47 ± 0.11
OPC	10.87 ± 0.20	4.95 ± 0.12
TIP4P/2005	4.11 ± 0.17	2.40 ± 0.05

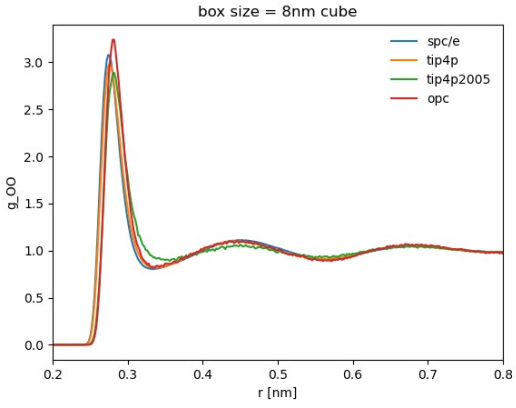

Fig. S2 Oxygen–oxygen radial distribution functions g_{OO}(r) for SPC/E, TIP4P, TIP4P/2005, and OPC water models. All simulations were performed using 8-nm cubic simulation boxes with a time step of 2 fs.