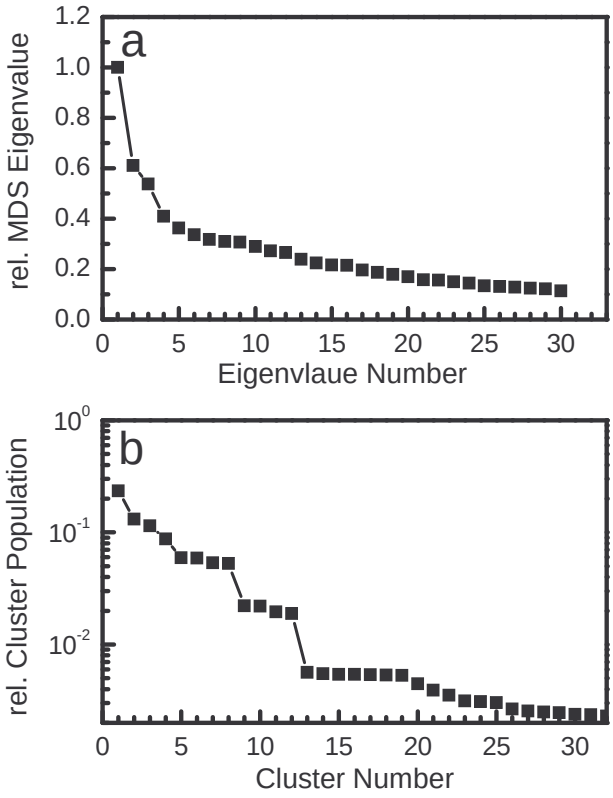
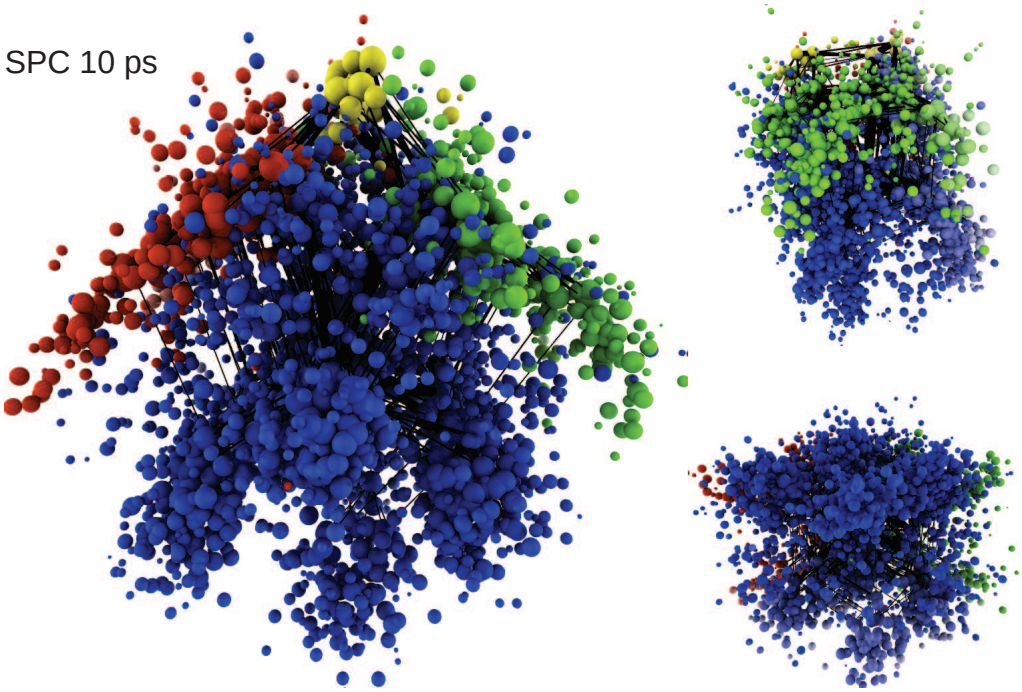
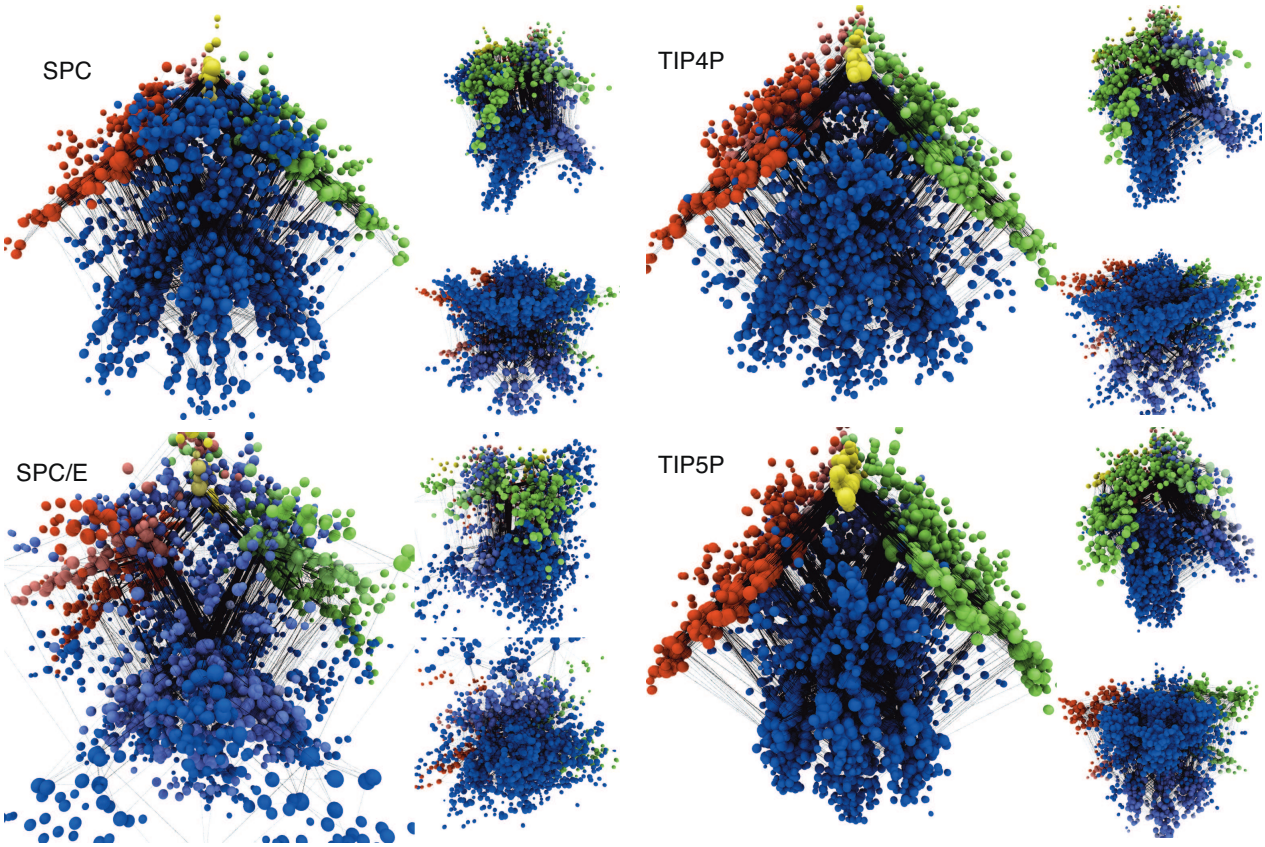
1 Supplementary Information



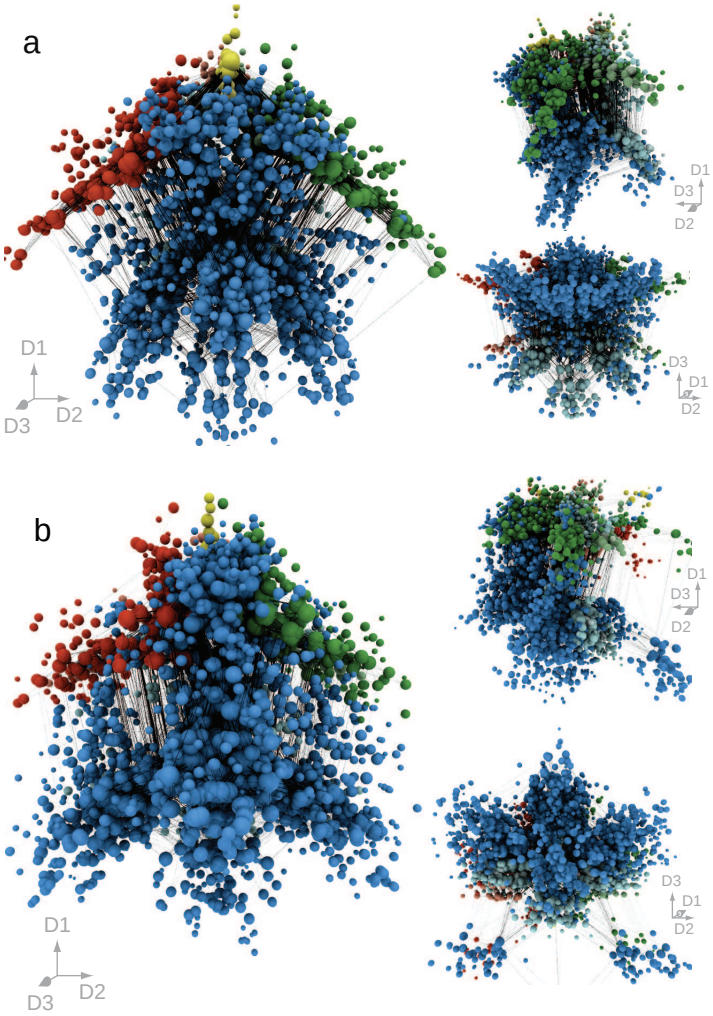
Supplemental Fig. 1: (a) MDS eigenvalues, sorted by size, and (b) gradient cluster, sorted by population.



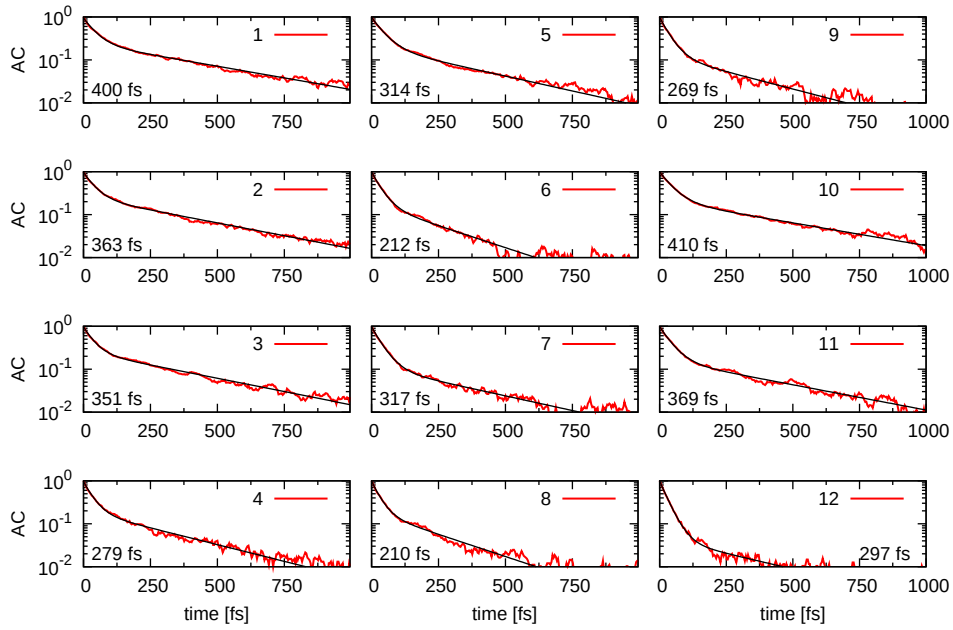
Supplemental Fig. 2: MDS conformation-space-network for short (10 ps) trajectory shows the same features as the long (100 ps) trajectory described in the main text, indicating that the sampling of the conformation-space has converged.



Supplemental Fig. 3: MDS networks for SPC, SPC/E, Tip4p, and Tip5p at 300 K show the same essential structures.



Supplemental Fig. 4: MDS networks using (a) a hydrogen-bond criterion which is based on orbital-mixing point of view, and which has been introduced recently by Skinner *et al.* [S1, S2], and (b) a criterion which uses simple geometry thresholds (O$\cdots$O distance smaller than 3.3 Å, angle between O–H and O$\cdots$O rays smaller than 30°). Both show the essential same structure.



Supplemental Fig. 5: Cluster lifetime autocorrelation function $\langle c_n(t)c_n(0)\rangle$, where $c_n(t) = 1$ when the trajectory populates a gradient-cluster n, and $c_n(t) = 0$ otherwise (normalized and average subtracted). The decay times of the slow component in a biexponential is indicated.

References

[S1] Kumar, R., Schmidt, J. R., and Skinner, J. L. Hydrogen bond definitions and dynamics in liquid water. *J. Chem. Phys.* **126**, 204107 (2007).

[S2] Auer, B., Schmidt, J. R., and Skinner, J. L. Hydrogen bonding and Raman, IR, and 2D-IR spectroscopy of dilute HOD in liquid D_2O. *Proc. Natl. Acad. Sci. USA* **104**, 14215–14220 (2007).