



Structural characteristics of low-density environments in liquid water

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Supplemental Material for

Structural Characteristics of Low Density Environments in Liquid Water

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S1 Distribution of all O-O-O angles

As discussed in the main article, the distribution of O-O-O angles in liquid can be used for characterising tetrahedral environments in liquid water, and estimating the fraction of locally favoured tetrahedral structures (LFTS) in water. However, in the study, we used the average angle a water molecule forms with its neighbors where most studies reported in the literature uses all angles (θ_{all}) formed by water molecules with their neighbors. This section provides a brief description of why we used the average angle (θ_{avg}), and not all the angles (θ_{all}). The distribution of all O-O-O angles shows a bi-modal behavior with one peak at around 105° , close to the internal angle of a regular tetrahedron, and another peak at smaller angles. The second peak at smaller angles indicate another environment with more tighter orientation between the neighbors. With an increase in temperature, more hydrogen bonds get broken leading to a decline in the population of molecules in tetrahedral environments in liquid water. The behaviour is reflected by a reduction in the peak at θ_{all} close to the internal angle of a regular tetrahedron (Fig. S1).

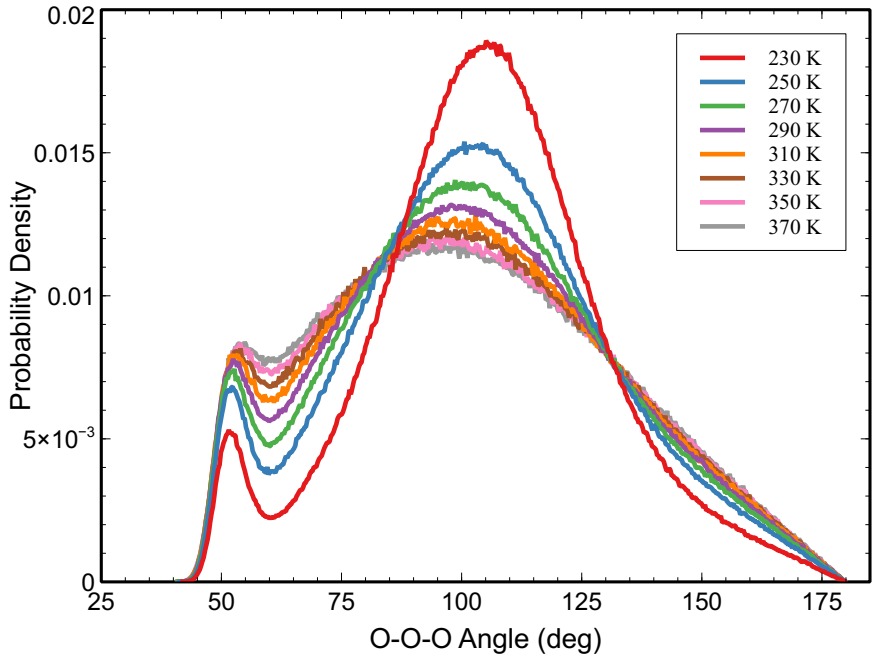


Figure S1: Distribution of all O-O-O angles formed by water molecules with their neighbors. The results are from simulations of the iAMOEBA water model at 1 bar.

The peak corresponding to the tetrahedral angles progressively moves to lower values as the temperature is increased. This could lead to an ambiguity in characterising the structure of local tetrahedral environments using the distribution of *all angles* (θ_{all}) an oxygen atom makes with its neighbors. For instance, the shift of the tetrahedral peak to lower angles may be wrongly interpreted as the tetrahedral structures formed at higher temperatures being slightly more skewed. On the other hand, the distribution of average angle

formed between neighbors (θ_{avg}) shows a consistent peak at $\sim 109.5^\circ$, as shown in Fig. 1b in the main article. This clearly indicates that the mean structure of tetrahedral environments is not significantly impacted by temperature and pressure. However, as will be shown later, the standard deviation of the distributions increases with temperature, indicating higher extent of fluctuations between structural forms. Another advantage of using θ_{avg} is that it assigns one single order parameter to any given water molecules, independent of its number of neighbors. This is more convenient than using all the $\binom{n}{2}$ (number of combinations possible when selecting 2 out of n objects) it makes with its n neighbors.

S2 Distribution of θ_{avg} estimated from the TIP4P/2005 model

Like described in the main article. the proposal to use O-O-O angles for characterising tetrahedral environments is dependant only on geometry of regular tetrahedra. Therefore, the method may be extended to other water models to study the two structural forms. In this section, we demonstrate the distribution of θ_{avg} in the TIP4P/2005 [1] water model. TIP4P/2005 is capable of reproducing properties of water with good accuracy [2,3]. This water model has also been extensively explored to study the two-state theory of water [4–8]. The widom line originating from the critical point [4], and the location of the critical point has both been reported for this water model [5,8]. We demonstrate the applicability of the method proposed in the manuscript to the TIP4P/2005 water model here.

S2.1 Simulation details

NPT Simulations of TIP4P/2005 model were performed with a box of 256 water molecules. The initial size of the box was maintained according to the density of water. Simulations as long as 50 ns were performed. The length of simulation trajectory was longer at lower temperatures. The leapfrog algorithm [9] was used to generate molecular trajectories with a time step of 1 fs. The temperature and pressure in the system were maintained using Nosé–Hoover thermostat [10–12] (coupling time 1 ps) and Parrinello Rahman barostat [13,14] (coupling time 2 ps). Simulations using this water model was done using Gromacs 2020 [15]. All the results reported here are at 1 bar.

S2.2 Characterization of tetrahedral structure in TIP4P/2005

The distribution of θ_{avg} in water at different temperatures and 1 bar are shown in Fig. S2. The distribution is similar to the results from the iAMOEBA water model - with two peaks. One peak corresponding to the internal angle of a regular tetrahedron and the other at smaller angles.

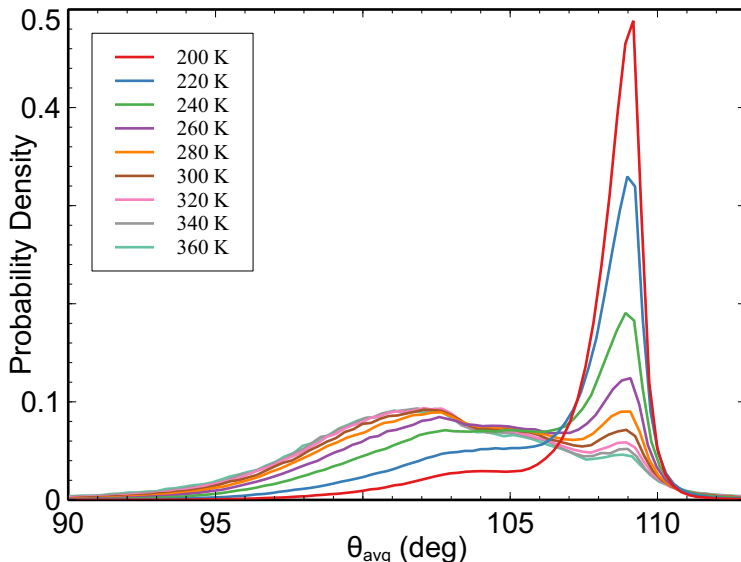


Figure S2: The distribution of θ_{avg} using TIP4P/2005 water model. The results shown are at different temperatures and 1 bar.

The distribution can be decomposed into two components as demonstrated in Fig. S3a. Subsequently, the fraction of LFTS molecules were estimated and compared with the values reported in the main article (using iAMOEBA water model). The results are shown in Fig. S3b. It is observed that iAMOEBA and TIP4P/2005 exhibit similar behaviour in Fig. S3b. The fraction of LFTS molecules increase as the temperature is reduced. The rate of increase also gradually increases as the temperature is lowered. A sharper change is observed between temperatures 210-230 K. The differences between the values, could be attributed to the inherent differences between the polarizable and flexible iAMOEBA water model and the rigid, non-polarizable TIP4P/2005 water model.

The fractions of LFTS liquid reported in this work are compared with the results reported by Russo and Tanaka [16] calculated using the ζ parameter. ζ is defined by equation 1.

$$\zeta(i) = d_{j'i} - d_{j''i} \quad (1)$$

where $d_{j'i}$ is the distance to the closest neighbour (j') which is not hydrogen bonded to the molecule i and $d_{j''i}$ is the distance to the farthest neighbour (j'') which is hydrogen bonded to the molecule i . The comparison of LFTS fractions estimated from TIP4P/2005 using our method (red line in Fig. S3b) and the values reported by Russo and Tanaka using ζ (TIP4P/2005 water model- Green line in S3b). We note that the fraction of LFTS estimated using the two methods exhibit similar trends, but with some deviation from each other. The deviation is observed to be smaller at higher temperatures.

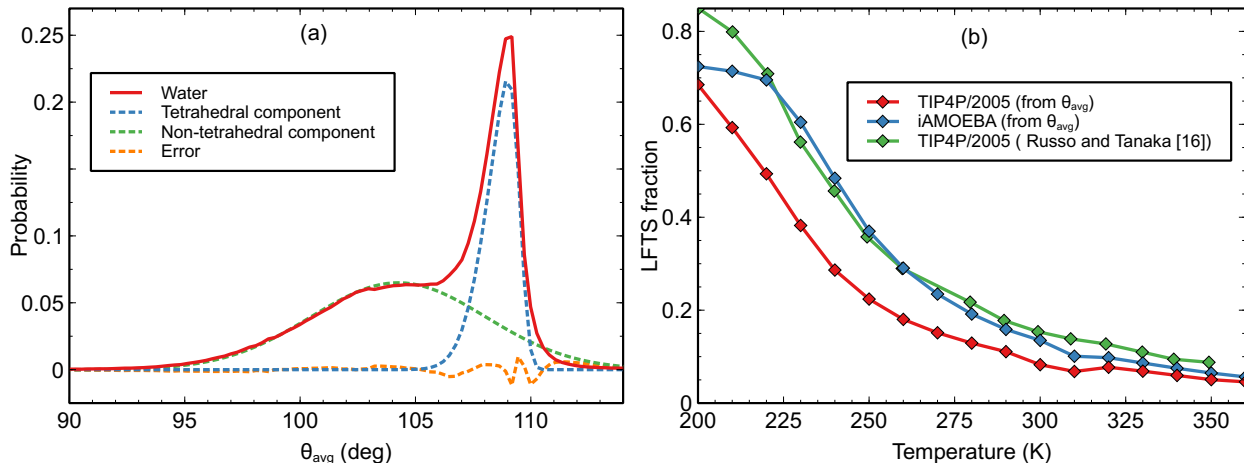


Figure S3: (a) Demonstration of Gaussian decomposition of the distribution of θ_{avg} in TIP4P/2005 water model. The decomposition is shown at 230 K and 1 bar. (b) The fraction of tetrahedral molecules estimated from decomposition of the distribution of θ_{avg} . The fractions estimated from TIP4P/2005 (red) are compared to the results reported in main manuscript (iAMOEBa - blue) and the values reported by Russo and Tanaka [16] using TIP4P/2005 model and the ζ parameter. All the results shown are at 1 bar.

S3 Parameters estimated by Gaussian Decomposition

The parameters of equation 1 in the main article were determined by minimising the least square error between the actual distribution and the sum of skewed Gaussian components. The range of search of each parameter is shown in Table S1. The change in the values of these parameters with temperature and pressure is shown in Figure S4a-S4f.

The mean of the tetrahedral distribution (μ_s shown in Fig. S4a) is observed to be more or less constant, lying in the range 109°-110°. This supports the argument in section S1 that the structure of the tetrahedral environment is not significantly impacted by temperature and pressure. On the other hand, the mean of the non tetrahedral distribution (μ_ρ shown in Fig. S4d) is observed to gradually decrease with temperature. The standard deviations of both tetrahedral and non-tetrahedral components (Fig. S4b and S4e) are observed to grow with temperature, which is consistent with the fact that structural fluctuations increase with temperature. The *skew-ness* of both distributions (α_s and α_ρ) are shown in Fig. S4c and Fig. S4f, respectively. All the values of the estimated parameters at different temperatures and pressures are shown in Table S2- S10.

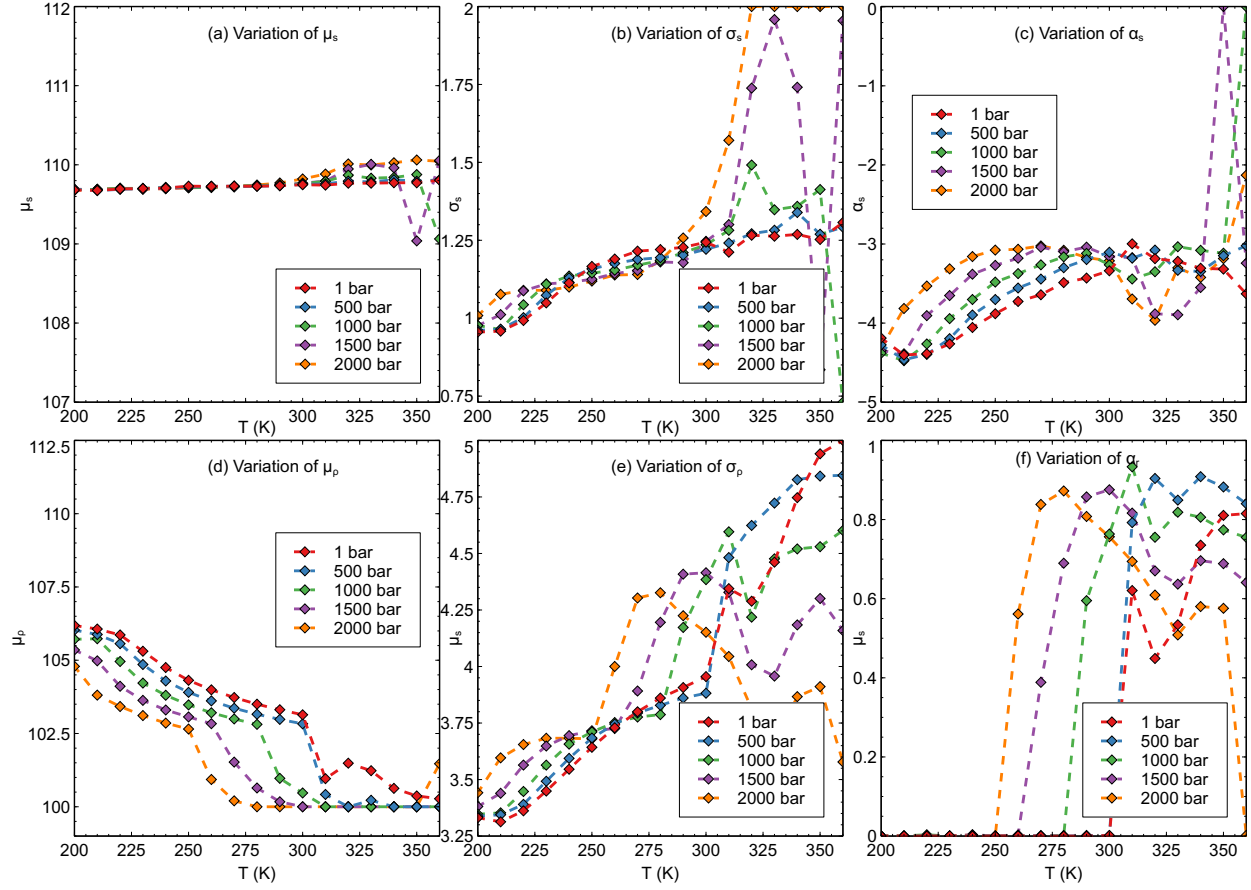


Figure S4: Visual representation of variation of parameters in equation 1 in the main article, with temperature and pressure.

Table S1: The search range of parameters of equation 1 in the main article.

Parameter	Range of search	
	Minimum	Maximum
s	0	1
μ_s	108	112
σ_s	0	2
α_s	-10	10
μ_ρ	100	107
σ_ρ	0	5
α_ρ	0	10

Table S2: Parameters of the Gaussian decomposition equation (equation 1 in the main article), estimated by minimising least square errors, for iAMOEBA and TIP4P/2005 water models at 1 bar.

T (K)	P (bar)	iAMOEBA							TIP4P/2005						
		s	μ_s	σ_s	α_s	μ_ρ	σ_ρ	α_ρ	s	μ_s	σ_s	α_s	μ_ρ	σ_ρ	α_ρ
200	1	0.724	109.68	0.96	-4.19	106.19	3.33	7.37e-05	0.685	109.49	1.06	-4.18	105.90	3.41	1.62e-06
210	1	0.714	109.68	0.96	-4.40	106.06	3.31	2.96e-04	0.593	109.52	1.09	-4.13	105.31	3.59	1.11e-05
220	1	0.695	109.69	0.99	-4.39	105.86	3.36	2.06e-06	0.493	109.52	1.16	-4.16	104.79	3.69	1.72e-05
230	1	0.605	109.70	1.05	-4.26	105.31	3.45	6.81e-04	0.382	109.52	1.22	-3.95	104.26	3.79	8.67e-07
240	1	0.484	109.71	1.11	-4.06	104.75	3.55	1.62e-05	0.286	109.52	1.25	-3.93	103.80	3.88	2.49e-07
250	1	0.370	109.73	1.17	-3.88	104.31	3.64	1.13e-04	0.224	109.53	1.27	-3.78	103.43	3.94	1.09e-03
260	1	0.291	109.73	1.19	-3.73	103.99	3.73	7.38e-04	0.180	109.54	1.31	-3.80	103.15	3.97	5.03e-04
270	1	0.235	109.73	1.22	-3.64	103.73	3.80	5.92e-05	0.151	109.55	1.31	-3.66	102.94	4.00	1.50e-03
280	1	0.192	109.73	1.22	-3.49	103.50	3.86	2.29e-05	0.129	109.56	1.38	-3.84	100.76	4.47	6.61e-01
290	1	0.159	109.74	1.23	-3.43	103.31	3.91	1.99e-07	0.111	109.59	1.49	-4.10	100.11	4.73	8.76e-01
300	1	0.135	109.75	1.24	-3.34	103.14	3.96	1.50e-03	0.100	109.62	1.53	-4.12	100.00	4.74	8.64e-01
310	1	0.101	109.74	1.21	-3.00	100.96	4.34	6.21e-01	0.082	109.62	1.45	-4.09	100.00	4.70	8.14e-01
320	1	0.098	109.77	1.27	-3.19	101.49	4.29	4.49e-01	0.077	109.67	1.56	-3.93	100.00	4.64	7.61e-01
330	1	0.086	109.77	1.26	-3.22	101.23	4.46	5.34e-01	0.069	109.64	1.50	-3.48	100.00	4.64	7.20e-01
340	1	0.075	109.77	1.27	-3.31	100.63	4.75	7.35e-01	0.060	109.65	1.49	-3.54	100.00	4.66	6.98e-01
350	1	0.065	109.77	1.25	-3.32	100.37	4.94	8.11e-01	0.051	109.64	1.40	-3.27	100.00	4.70	6.66e-01
360	1	0.056	109.81	1.31	-3.63	100.27	5.00	8.15e-01	0.037	108.88	0.68	-0.00	100.00	4.81	6.65e-01

Table S3: Parameters of the Gaussian decomposition equation (equation 1 in the main article), estimated by minimizing least square errors, for the iAMOEBA water model at 250 bar.

T (K)	P (bar)	s	μ_s	σ_s	α_s	μ_ρ	σ_ρ	α_ρ
200	250	0.716	109.68	0.95	-4.38	106.01	3.34	3.06e-04
210	250	0.717	109.67	0.96	-4.39	106.05	3.32	7.90e-06
220	250	0.672	109.69	1.00	-4.41	105.66	3.38	8.75e-06
230	250	0.575	109.70	1.06	-4.23	105.11	3.47	7.29e-06
240	250	0.434	109.73	1.13	-3.96	104.53	3.57	4.88e-05
250	250	0.332	109.72	1.16	-3.77	104.12	3.66	4.55e-04
260	250	0.260	109.73	1.18	-3.66	103.81	3.74	1.53e-07
270	250	0.209	109.72	1.20	-3.54	103.55	3.80	8.03e-08
280	250	0.174	109.74	1.21	-3.42	103.34	3.85	6.80e-06
290	250	0.146	109.74	1.22	-3.34	103.13	3.88	2.45e-03
300	250	0.125	109.75	1.22	-3.15	103.00	3.92	4.70e-04
310	250	0.099	109.77	1.27	-3.33	100.87	4.34	6.35e-01
320	250	0.087	109.77	1.26	-3.21	100.36	4.64	8.25e-01
330	250	0.083	109.79	1.31	-3.43	100.50	4.67	7.75e-01
340	250	0.070	109.78	1.26	-3.25	100.23	4.85	8.59e-01
350	250	0.061	109.78	1.25	-3.20	100.04	4.98	9.06e-01
360	250	0.054	109.77	1.23	-3.18	100.04	5.00	8.69e-01

Table S4: Parameters of the Gaussian decomposition equation (equation 1 in the main article), estimated by minimizing least square errors, for the iAMOEBA water model at 500 bar.

T (K)	P (bar)	s	μ_s	σ_s	α_s	μ_ρ	σ_ρ	α_ρ
200	500	0.714	109.69	0.96	-4.28	106.03	3.34	5.03e-06
210	500	0.701	109.67	0.96	-4.46	105.88	3.34	1.29e-05
220	500	0.659	109.69	1.00	-4.40	105.57	3.39	2.96e-04
230	500	0.527	109.70	1.07	-4.20	104.85	3.49	6.14e-06
240	500	0.388	109.71	1.13	-3.90	104.29	3.59	3.87e-04
250	500	0.292	109.72	1.16	-3.70	103.90	3.68	6.87e-06
260	500	0.232	109.73	1.18	-3.56	103.61	3.75	8.18e-08
270	500	0.190	109.73	1.19	-3.44	103.36	3.79	1.36e-03
280	500	0.158	109.73	1.19	-3.30	103.16	3.83	2.01e-07
290	500	0.134	109.74	1.20	-3.20	102.98	3.86	1.61e-03
300	500	0.116	109.75	1.22	-3.10	102.83	3.88	1.02e-03
310	500	0.094	109.76	1.24	-3.18	100.42	4.48	7.92e-01
320	500	0.077	109.79	1.27	-3.08	100.00	4.62	9.04e-01
330	500	0.075	109.78	1.28	-3.33	100.22	4.72	8.49e-01
340	500	0.069	109.81	1.34	-3.35	100.00	4.83	9.08e-01
350	500	0.060	109.79	1.27	-3.15	100.00	4.84	8.82e-01
360	500	0.053	109.81	1.29	-3.02	100.00	4.85	8.40e-01

Table S5: Parameters of the Gaussian decomposition equation (equation 1 in the main article), estimated by minimizing least square errors, for the iAMOEBA water model at 750 bar.

T (K)	P (bar)	s	μ_s	σ_s	α_s	μ_ρ	σ_ρ	α_ρ
200	750	0.692	109.68	0.97	-4.37	105.84	3.33	6.02e-06
210	750	0.685	109.68	0.97	-4.41	105.75	3.35	4.95e-04
220	750	0.607	109.70	1.03	-4.24	105.20	3.42	9.89e-07
230	750	0.462	109.71	1.09	-4.02	104.54	3.52	1.97e-07
240	750	0.337	109.72	1.13	-3.78	104.06	3.62	3.52e-07
250	750	0.259	109.72	1.16	-3.61	103.71	3.70	2.47e-06
260	750	0.206	109.72	1.16	-3.40	103.42	3.75	6.62e-04
270	750	0.169	109.73	1.17	-3.28	103.19	3.79	1.69e-04
280	750	0.143	109.73	1.18	-3.23	102.98	3.81	4.15e-03
290	750	0.124	109.74	1.20	-3.08	102.78	3.82	7.19e-03
300	750	0.107	109.75	1.24	-3.25	100.87	4.27	6.39e-01
310	750	0.090	109.77	1.25	-3.35	100.20	4.56	8.69e-01
320	750	0.077	109.83	1.37	-3.30	100.00	4.38	8.05e-01
330	750	0.072	109.79	1.26	-3.08	100.00	4.70	9.00e-01
340	750	0.068	109.82	1.35	-3.18	100.00	4.67	8.50e-01
350	750	0.059	109.82	1.30	-3.00	100.00	4.70	8.25e-01
360	750	0.056	109.87	1.36	-3.10	100.00	4.68	7.88e-01

Table S6: Parameters of the Gaussian decomposition equation (equation 1 in the main article), estimated by minimizing least square errors, for the iAMOEBA water model at 1000 bar.

T (K)	P (bar)	s	μ_s	σ_s	α_s	μ_ρ	σ_ρ	α_ρ
200	1000	0.688	109.69	0.98	-4.38	105.71	3.35	6.65e-06
210	1000	0.686	109.69	0.96	-4.48	105.73	3.35	4.81e-04
220	1000	0.566	109.70	1.04	-4.26	104.96	3.45	3.36e-03
230	1000	0.392	109.70	1.11	-3.94	104.22	3.56	2.60e-07
240	1000	0.289	109.71	1.14	-3.70	103.80	3.66	1.54e-04
250	1000	0.225	109.71	1.14	-3.48	103.48	3.71	1.66e-07
260	1000	0.182	109.71	1.15	-3.37	103.21	3.75	4.87e-07
270	1000	0.154	109.73	1.17	-3.26	103.00	3.78	8.40e-07
280	1000	0.131	109.74	1.18	-3.16	102.81	3.79	1.42e-03
290	1000	0.113	109.75	1.21	-3.12	100.97	4.17	5.95e-01
300	1000	0.099	109.76	1.24	-3.26	100.47	4.38	7.63e-01
310	1000	0.086	109.79	1.28	-3.44	100.00	4.60	9.33e-01
320	1000	0.079	109.87	1.49	-3.35	100.00	4.22	7.55e-01
330	1000	0.072	109.83	1.35	-3.03	100.00	4.48	8.18e-01
340	1000	0.066	109.84	1.36	-3.08	100.00	4.52	8.06e-01
350	1000	0.062	109.88	1.41	-3.12	100.00	4.53	7.74e-01
360	1000	0.048	109.06	0.73	0.00	100.00	4.60	7.55e-01

Table S7: Parameters of the Gaussian decomposition equation (equation 1 in the main article), estimated by minimizing least square errors, for the iAMOEBA water model at 1250 bar.

T (K)	P (bar)	s	μ_s	σ_s	α_s	μ_ρ	σ_ρ	α_ρ
200	1250	0.663	109.69	0.97	-4.33	105.51	3.39	1.27e-06
210	1250	0.642	109.68	1.00	-4.39	105.32	3.39	8.15e-06
220	1250	0.502	109.69	1.06	-4.20	104.63	3.49	8.68e-06
230	1250	0.330	109.71	1.11	-3.76	103.95	3.61	3.67e-07
240	1250	0.247	109.71	1.13	-3.55	103.54	3.68	1.17e-04
250	1250	0.197	109.71	1.14	-3.39	103.26	3.72	1.27e-07
260	1250	0.164	109.72	1.15	-3.29	103.02	3.74	1.30e-03
270	1250	0.139	109.73	1.16	-3.11	102.81	3.75	1.62e-03
280	1250	0.120	109.73	1.18	-3.12	101.48	3.95	4.05e-01
290	1250	0.104	109.74	1.19	-3.06	100.57	4.28	7.21e-01
300	1250	0.090	109.76	1.22	-3.16	100.07	4.51	9.06e-01
310	1250	0.083	109.79	1.28	-3.20	100.00	4.46	8.71e-01
320	1250	0.079	109.91	1.61	-3.68	100.00	4.08	6.94e-01
330	1250	0.076	109.90	1.59	-3.44	100.00	4.23	7.24e-01
340	1250	0.073	109.93	1.62	-3.47	100.00	4.33	7.36e-01
350	1250	0.054	109.05	0.81	0.00	100.00	4.45	7.40e-01
360	1250	0.049	109.08	0.80	0.00	100.00	4.46	7.16e-01

Table S8: Parameters of the Gaussian decomposition equation (equation 1 in the main article), estimated by minimizing least square errors, for the iAMOEBA water model at 1500 bar.

T (K)	P (bar)	s	μ_s	σ_s	α_s	μ_ρ	σ_ρ	α_ρ
200	1500	0.640	109.69	0.98	-4.35	105.35	3.38	1.24e-06
210	1500	0.586	109.69	1.01	-4.39	104.98	3.44	4.54e-06
220	1500	0.380	109.70	1.09	-3.91	104.11	3.56	3.86e-07
230	1500	0.270	109.70	1.11	-3.65	103.63	3.65	3.30e-07
240	1500	0.213	109.70	1.11	-3.38	103.30	3.69	3.35e-03
250	1500	0.174	109.71	1.12	-3.27	103.07	3.72	4.02e-08
260	1500	0.148	109.73	1.14	-3.18	102.84	3.73	2.83e-03
270	1500	0.127	109.72	1.15	-3.04	101.52	3.89	3.89e-01
280	1500	0.110	109.74	1.18	-3.10	100.64	4.20	6.90e-01
290	1500	0.094	109.75	1.18	-3.04	100.17	4.41	8.57e-01
300	1500	0.085	109.77	1.25	-3.16	100.00	4.41	8.75e-01
310	1500	0.080	109.79	1.30	-3.17	100.00	4.33	8.16e-01
320	1500	0.083	109.95	1.74	-3.88	100.00	4.01	6.70e-01
330	1500	0.085	110.01	1.96	-3.89	100.00	3.96	6.37e-01
340	1500	0.076	109.96	1.74	-3.55	100.00	4.18	6.96e-01
350	1500	0.055	109.04	0.83	0.00	100.00	4.30	6.89e-01
360	1500	0.073	110.06	1.95	-3.24	100.00	4.16	6.41e-01

Table S9: Parameters of the Gaussian decomposition equation (equation 1 in the main article), estimated by minimizing least square errors, for the iAMOEBA water model at 1750 bar.

T (K)	P (bar)	s	μ_s	σ_s	α_s	μ_ρ	σ_ρ	α_ρ
200	1750	0.588	109.69	1.01	-4.31	104.95	3.43	4.34e-04
210	1750	0.464	109.70	1.06	-4.09	104.41	3.50	6.12e-04
220	1750	0.301	109.70	1.10	-3.73	103.73	3.62	3.47e-07
230	1750	0.227	109.70	1.10	-3.47	103.36	3.67	7.53e-07
240	1750	0.184	109.72	1.11	-3.29	103.09	3.70	3.95e-08
250	1750	0.154	109.70	1.12	-3.16	102.85	3.70	4.29e-04
260	1750	0.133	109.72	1.14	-3.09	102.63	3.70	5.73e-03
270	1750	0.115	109.72	1.15	-3.00	100.80	4.09	6.21e-01
280	1750	0.099	109.74	1.17	-3.07	100.15	4.36	8.59e-01
290	1750	0.088	109.76	1.20	-3.11	100.00	4.37	8.78e-01
300	1750	0.083	109.78	1.29	-3.21	100.00	4.28	8.15e-01
310	1750	0.082	109.85	1.45	-3.58	100.00	4.16	7.41e-01
320	1750	0.086	109.97	1.84	-3.88	100.00	3.93	6.50e-01
330	1750	0.083	110.02	2.00	-3.42	100.00	3.77	5.62e-01
340	1750	0.082	110.03	1.98	-3.66	100.00	4.00	6.35e-01
350	1750	0.083	110.05	2.00	-3.10	101.62	3.62	8.61e-07
360	1750	0.082	110.06	2.00	-2.48	101.58	3.62	1.57e-05

Table S10: Parameters of the Gaussian decomposition equation (equation 1 in the main article), estimated by minimizing least square errors, for the iAMOEBA water model at 2000 bar.

T (K)	P (bar)	s	μ_s	σ_s	α_s	μ_ρ	σ_ρ	α_ρ
200	2000	0.559	109.68	1.01	-4.31	104.79	3.44	6.11e-04
210	2000	0.328	109.69	1.08	-3.82	103.81	3.60	2.10e-07
220	2000	0.244	109.70	1.09	-3.53	103.42	3.65	6.72e-06
230	2000	0.195	109.69	1.09	-3.31	103.11	3.68	6.25e-07
240	2000	0.163	109.70	1.10	-3.16	102.85	3.68	3.05e-03
250	2000	0.139	109.71	1.12	-3.08	102.65	3.68	8.91e-09
260	2000	0.120	109.72	1.14	-3.07	100.93	4.00	5.61e-01
270	2000	0.103	109.73	1.14	-3.02	100.20	4.30	8.38e-01
280	2000	0.092	109.74	1.19	-3.08	100.00	4.33	8.72e-01
290	2000	0.085	109.77	1.26	-3.17	100.00	4.22	8.08e-01
300	2000	0.081	109.82	1.34	-3.21	100.00	4.15	7.56e-01
310	2000	0.082	109.88	1.57	-3.69	100.00	4.04	6.94e-01
320	2000	0.090	110.01	2.00	-3.96	100.00	3.81	6.09e-01
330	2000	0.080	110.00	2.00	-3.30	100.00	3.66	5.08e-01
340	2000	0.081	110.03	2.00	-3.42	100.00	3.87	5.80e-01
350	2000	0.078	110.06	2.00	-3.18	100.00	3.91	5.76e-01
360	2000	0.081	110.05	2.00	-2.13	101.47	3.58	1.78e-03

SI References

- [1] Jose LF Abascal and Carlos Vega. A general purpose model for the condensed phases of water: Tip4p/2005. *J. Chem. Phys.*, 123(23):234505, 2005.
- [2] Harshad Pathak, JC Palmer, Daniel Schlesinger, Kjartan Thor Wikfeldt, Jonas A Sellberg, Lars GM Pettersson, and Anders Nilsson. The structural validity of various thermodynamical models of supercooled water. *J. Chem. Phys.*, 145(13):134507, 2016.
- [3] Carlos Vega and Jose LF Abascal. Simulating water with rigid non-polarizable models: A general perspective. *Phys. Chem. Chem. Phys.*, 13(44):19663–19688, 2011.
- [4] José LF Abascal and Carlos Vega. Widom line and the liquid–liquid critical point for the tip4p/2005 water model. *J. Chem. Phys.*, 133(23):234502, 2010.
- [5] Pablo G Debenedetti, Francesco Sciortino, and Gül H Zerze. Second critical point in two realistic models of water. *Science*, 369(6501):289–292, 2020.
- [6] Lars GM Pettersson. A two-state picture of water and the funnel of life. In *International Conference Physics of Liquid Matter: Modern Problems*, pages 3–39. Springer, 2018.
- [7] Alexander Späh, Harshad Pathak, Kyung Hwan Kim, Fivos Perakis, Daniel Mariedahl, Katrin Amann-Winkel, Jonas A Sellberg, Jae Hyuk Lee, Sangsoo Kim, Jaehyun Park, et al. Apparent power-law behavior of water’s isothermal compressibility and correlation length upon supercooling. *Phys. Chem. Chem. Phys.*, 21(1):26–31, 2019.
- [8] Paola Gallo, Katrin Amann-Winkel, Charles Austen Angell, Mikhail Alexeevich Anisimov, Frederic Caupin, Charusita Chakravarty, Erik Lascaris, Thomas Loerting, Athanassios Zois Panagiotopoulos, John Russo, and et al. Water: A tale of two liquids. *Chem. Rev.*, 116(13):7463–7500, 2016.
- [9] Roger Williams Hockney, SP Goel, and JW Eastwood. Quiet high-resolution computer models of a plasma. *J. Comput. Phys.*, 14(2):148–158, 1974.
- [10] Shūichi Nosé. A molecular dynamics method for simulations in the canonical ensemble. *Mol. Phys.*, 52(2):255–268, 1984.
- [11] William G Hoover. Canonical dynamics: Equilibrium phase-space distributions. *Phys. Rev. A*, 31(3):1695, 1985.

- [12] Shuichi Nosé. A unified formulation of the constant temperature molecular dynamics methods. *J. Chem. Phys.*, 81(1):511–519, 1984.
- [13] Michele Parrinello and Aneesur Rahman. Crystal structure and pair potentials: A molecular-dynamics study. *Phys. Rev. Lett.*, 45(14):1196, 1980.
- [14] Michele Parrinello and Aneesur Rahman. Polymorphic transitions in single crystals: New molecular dynamics method. *J. Appl. Phys.*, 52(12):7182–7190, 1981.
- [15] David Van Der Spoel, Erik Lindahl, Berk Hess, Gerrit Groenhof, Alan E Mark, and Herman JC Berendsen. Gromacs: Fast, flexible, and free. *J. Comput. Chem.*, 26(16):1701–1718, 2005.
- [16] John Russo and Hajime Tanaka. Understanding water’s anomalies with locally favoured structures. *Nat. Commun.*, 5(1):1–11, 2014.