

Supplementary Information for

“Abnormal condensation of nanoscale water drops at ambient temperature”

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1. Dielectric ordering at the surface of water droplets

Using the TIP4P/2005 water model, we divided the droplet into several shells along the radial direction and calculated the radial and tangential components of the overall dipole moment in each shell, respectively.^[1] The total dipole moment is defined as

$$M_n = \sum_{j \in n} \mu_j,$$

where μ_j is the dipole moment of molecule j in the n th shell. Figure S1 shows the dipole moment profiles in the surface, intermediate, and central shells as a function of simulation time for the water droplet with $R =$

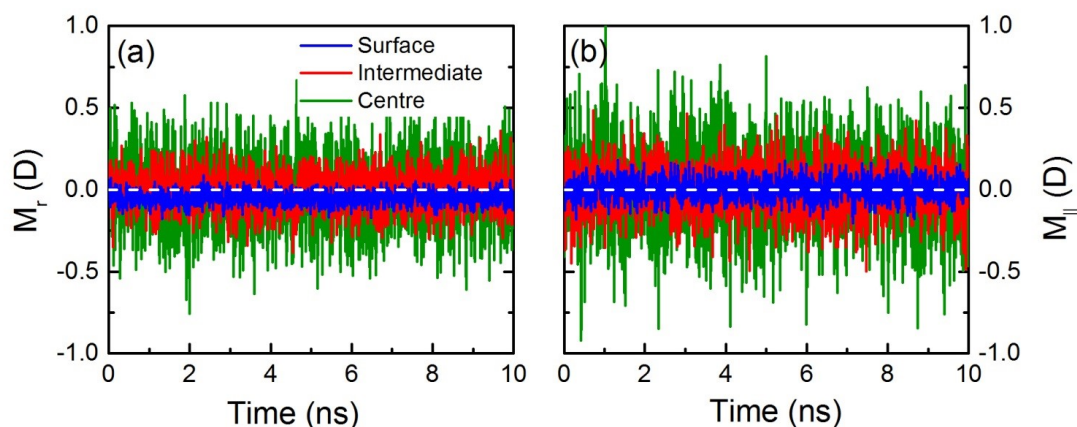


Fig. S1 Total dipole moments in surface, intermediate and central layers. (a) and (b) show the radial and the tangential components for the water droplet with $R = 5.2$ nm, respectively. ^[1]

5.2 nm at $T = 300$ K. Compared to the central water, the radial component of dipole moments at surface is significantly suppressed but the tangential components do not show visible changes. More importantly, we find that the mean radial component over time is negative in near-surface regions of droplets, which means that molecular dipoles tend to point toward the interior of droplets.

2. Molecular orientation at surface of water droplets

To quantitatively orientational characteristics of surface water molecules, we calculated the distributions of the angle between the molecular dipole and the direction normal to surface in the surface layer of droplets, as shown in Fig. S2a. The thickness of surface layers range from 2 to 2.4 Å, approximately the molecular monolayer. The angle distribution in bulk water is also provided in Fig. S2b. The most probability angle in the surface region of droplets is approximately 100°. For bulk water

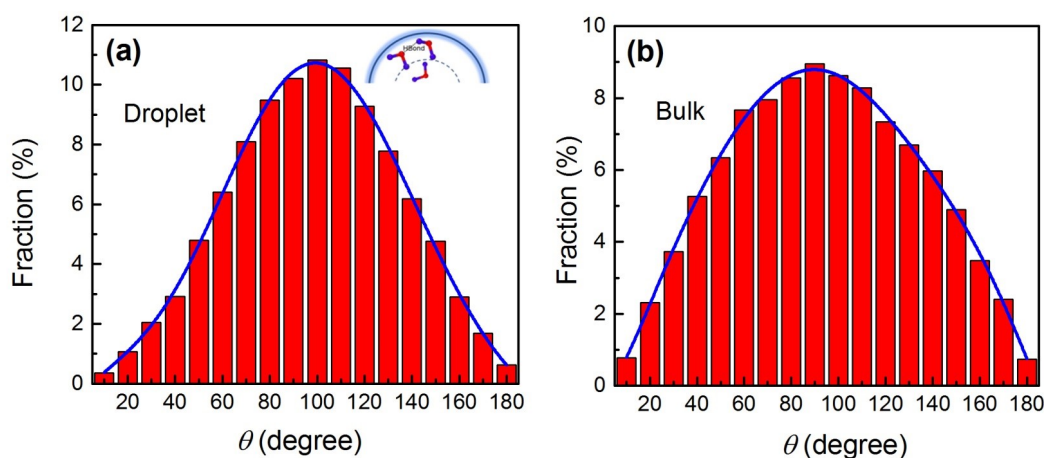


Fig. S2 Probability distributions of water molecular orientation in the surface layer at $T=300$ K. (a) Droplets. (b) Bulk water. The solid lines are fits to the Gaussian function. ^[1]

characterized by disordered molecular arrangements, it shows a Gaussian-type distribution around the value of 90°. It suggests that the dipoles in the droplet surface slightly point toward the interior, which is consistent with the conclusion drawn from Fig. S1.

3. Hydrogen-bond profile in water droplets

We used a geometric criterion to define hydrogen bond: the O-O distance less than the cutoff of 3.5 Å and the H-O-O bond angle less than 30° ^[2], and then calculated the number profile of hydrogen bonds along the radial direction, as shown in Fig. S3. The average in-plane hydrogen bond number per molecule in the surface layer rapidly increases in contrast to the drop of the hydrogen bonds bridging neighbouring layers. Therefore, the free surface of water droplets is characterized by the two-dimensional hydrogen-bonding network to some extent.

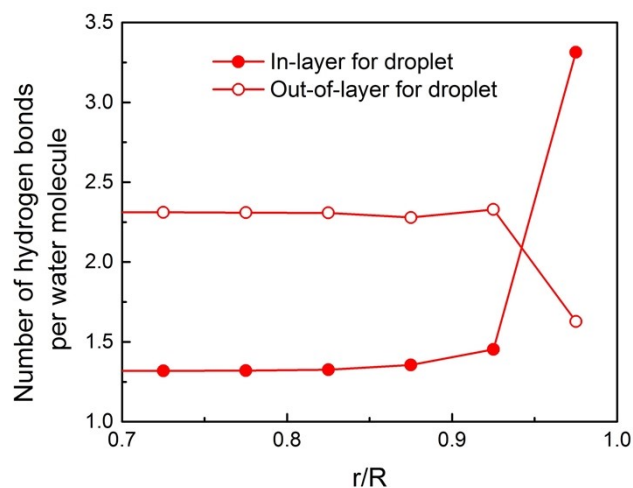


Fig. S3 Profiles of the average number of the in-layer and out-of-layer hydrogen bonds per water molecule along the direction perpendicular to the surface of water droplets. [1]

4. Dependence of the size distributions of water droplets on system size

Figure S4 shows the size distributions of water droplets after condensation in $N = 4000$, 5000 , and

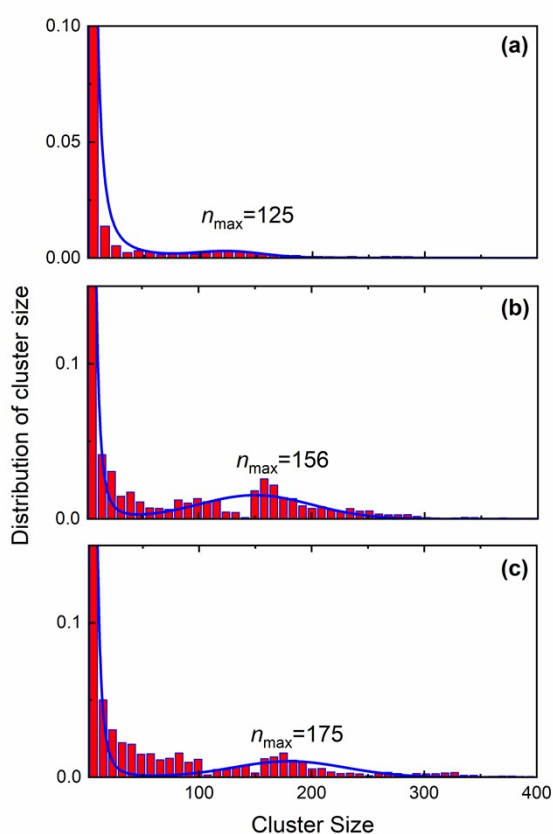


Fig. S4 Distributions of water droplet size for different system size at $T = 300$ K. (a) $N = 4000$; (b) $N = 5000$; (c) $N = 10000$.

10000 systems. The bimodal distributions are held in all simulation systems and the second-peak position

shifts to the large size region.

Reference

[1] Chenchen Guo, Hairong Qin, Yong Zhu, and Yongjun Lü, *Langmuir* 2021, 37, 13059–13066

[2] R. Kumar, J. R. Schmidt, J. L. Skinner. Hydrogen bonding definitions and dynamics in liquid water. *J. Chem. Phys.* 2007, 126(20), 204107.