Supplementary Information

A folded ice monolayer

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This file includes the details of calculating the N (Table S1); the structures of the C_b, C_T, C_c and C_{bh} ice models (Fig. S1-2); the thermal stability analysis of the C_f ice model (Fig. S3-4); Energies due to the oscillation entropy for several typical ice models (Table S2); Free energy of the C_f, C_T, C_{sp} and C_b at ρ = 0.1 Å^{-2} at different temperatures (Fig. S5); E_f as a function of water density for the C_p, C_{dw}, C_{ps1}, C_{ps2} and C_{psh} (Fig. S6) and the crystallographic information of the six new ice models.

Table S1. Detailed $|S_i\rangle$, N_{S_i} and $P(|S_i\rangle)$ for the six ice models. a and b are used to describe the oxygen atoms in the top and bottom planes. The a* and b* in the following table are of mirror-symmetry with the a and b, respectively.

	$\|S_i\rangle$	N_{S_i}	$P(\|S_i\rangle)$	N
C_f	a	0	0.25	0
	a*	0	0.25	
	b	0	0.25	
	b*	0	0.25	
C_{dw}	aa	1	0.5	1
	bb	1	0.5	
C_p	aa	2	0.5	2
	bb	2	0.5	
C_{ps1}	aaaa	2	0.5	2
	bbbb	2	0.5	
C_{ps2}	aaaa	2	0.5	2
	bbbb	2	0.5	
C_{psh}	aa	1	0.125	0.25
	bb	1	0.125	
	a	0	0.125	
	b	0	0.125	

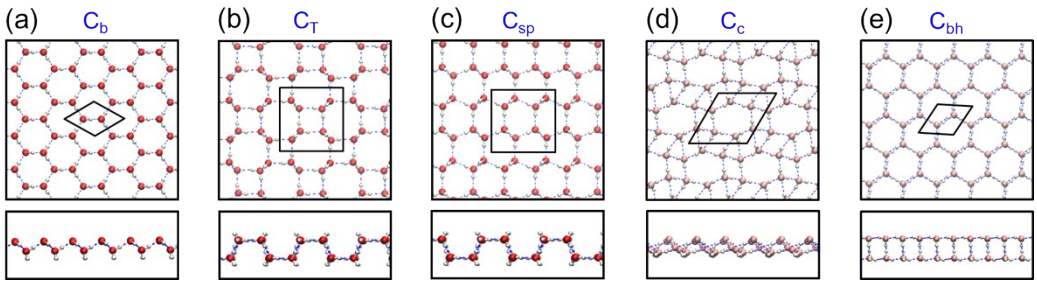

Fig. S1 Front and side views of the (a) buckled honeycomb (C_b), (b) truncated-square (C_T), (c) secondary prism, (c) corrugated (C_c) and (e) bilayer hexagonal (C_{bh}) ice models.

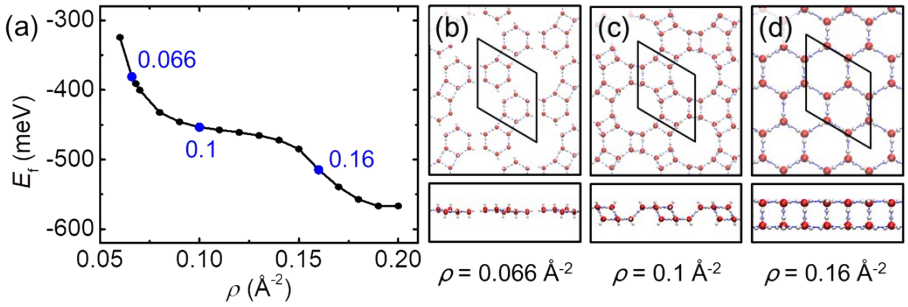

Fig. S2 (a) Formation energy (E_f) as a function of water density for the C_f. (b-d) Detailed structures of C_f with water densities of 0.066, 0.1 and 0.16 Å^{-2}, respectively.

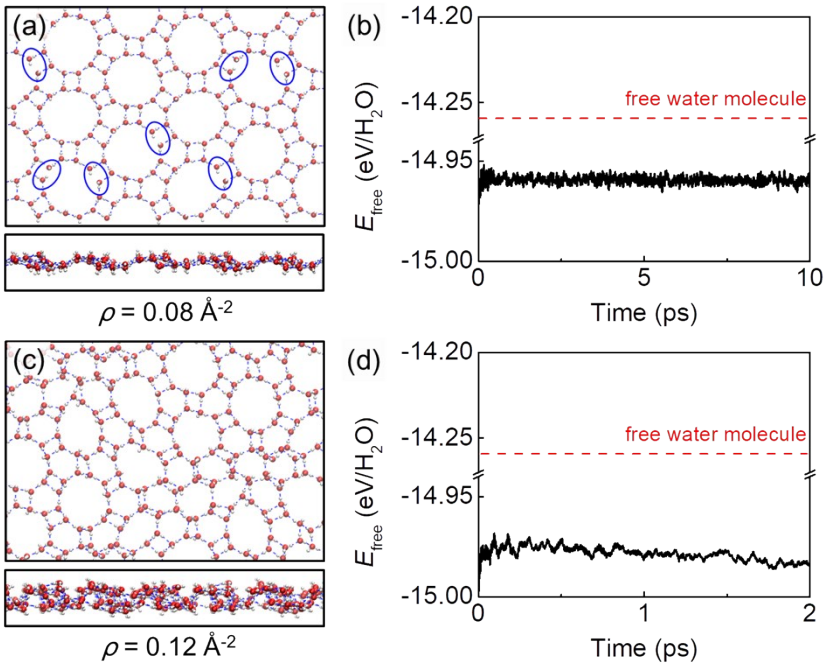

Fig. S3 (a) Front and side views of snapshots of the C_f at the end of 10 ps AIMD simulation at 78 K at the water density of 0.08 Å^{-2} and (b) its evolution of total energy throughout the AIMD simulation. (c) Front and side views of snapshots of the C_f at the end of 2 ps AIMD simulations at 78 K at the water density of 0.12 Å^{-2} and (d) its evolution of total free energy throughout the AIMD simulation.

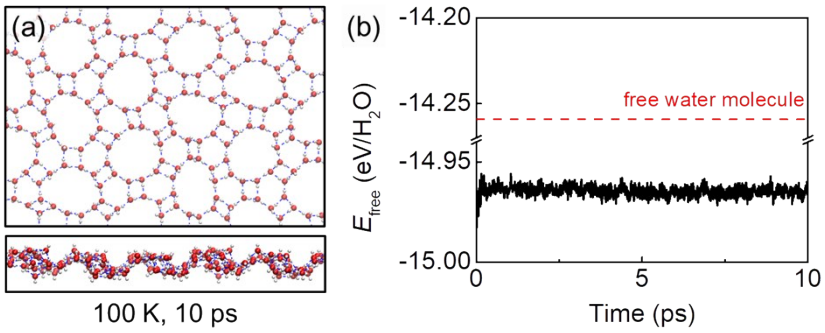

Fig. S4 (a) Front and side views of a snapshot of the C_f at the end of 10 ps AIMD simulation at 100 K. (b) The evolution of total free energy throughout the AIMD simulations.

Table S2. Free energies at 0 K (E_{free}) and energies due to the oscillation entropy for several typical ice models at a given water density ρ = 0.1 Å^{-2}. The water densities for C_{bh}, C_{dw}, C_p and C_{ps2} are chosen by the E_f at which these ice models have the lowest E_f.

	density (Å^{-2})	E_{free} (eV)	TS (eV)	
		0 K	78 K	100 K
C_f	0.1	-180.794	0.117	0.199
C_T	0.1	-120.493	0.061	0.108
C_{sp}	0.1	-120.447	0.066	0.116
C_b	0.1	-30.091	0.004	0.009
C_{bh}	0.185	-60.711	0.020	0.036
C_{dw}	0.11	-120.517	0.061	0.106
C_p	0.19	-60.430	0.023	0.042
C_{ps2}	0.18	-120.718	0.073	0.123
C_{psh}	0.18	-150.782	0.087	0.150

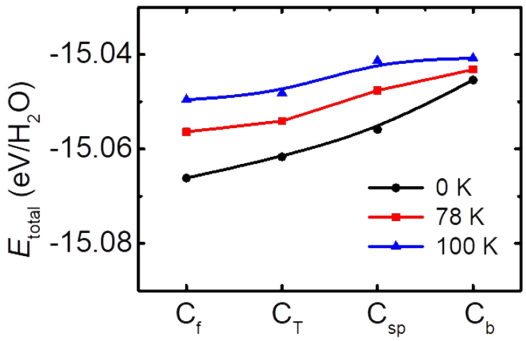

Fig. S5 Free energy of the C_f, C_T, C_{sp} and C_b at ρ = 0.1 Å^{-2} at different temperatures, including the contribution from the oscillation entropy.

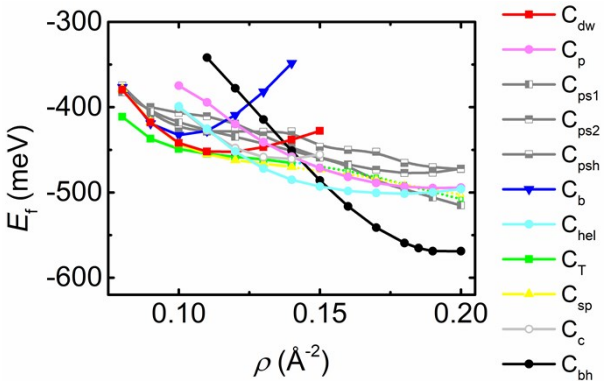

Fig. S6 Formation energy (E_f) as a function of water density for the C_{dw} (red solid squares), C_p (light magenta solid circles), C_{ps1} (grey half left squares), C_{ps2} (grey half up squares), C_{psh} (grey half down squares), C_b (blue solid down triangles), C_{hel} (cyan solid circles), C_T (green solid squares), C_{sp} (yellow solid up triangles), C_c (light grey open circles) and C_{bh} (black solid circles) models. The C_T and C_{sp} become thick double-layer ices in the region of ρ > 0.14 Å^{-2}, as shown by the dotted line parts.

Crystallographic Information of the C_f

cell_length_a	11.772		
cell_length_b	11.772		
cell_length_c	33.000		
cell_angle_alpha	90.000		
cell_angle_beta	90.000		
cell_angle_gamma	60.000		
O1	8.637	3.398	13.945
O2	7.178	5.887	13.816
O3	4.473	5.762	13.942
O4	3.032	3.259	13.828
O5	7.389	0.996	13.811
O6	4.500	0.983	13.938
O7	13.109	9.194	15.899
O8	14.589	6.924	16.000
O9	8.988	6.748	15.905
O10	10.460	4.265	16.010
O11	13.165	4.408	15.914
O12	10.219	9.161	15.997
H1	9.768	8.272	15.910
H2	8.310	6.538	15.225
H3	9.548	5.944	15.978
H4	11.456	4.321	15.921
H5	13.684	3.922	15.235
H6	13.584	5.295	15.981
H7	14.043	7.758	15.910
H8	12.132	9.111	15.970
H9	13.267	9.884	15.218
H10	6.183	5.839	13.915
H11	4.045	4.880	13.866
H12	3.572	2.422	13.922
H13	5.477	1.057	13.858
H14	7.848	1.879	13.914
H15	8.084	4.207	13.866
H16	10.301	3.791	16.843
H17	9.889	9.538	16.829
H18	15.082	7.025	16.831
H19	4.341	0.280	14.607
H20	3.950	6.255	14.611
H21	7.720	0.626	12.976
H22	9.321	3.608	14.618
H23	7.334	6.361	12.983
H24	2.537	3.157	12.998

Crystallographic Information of the C_p

cell_length_a	5.432		
cell_length_b	5.799		
cell_length_c	33.000		
cell_angle_alpha	90.000		
cell_angle_beta	90.000		
cell_angle_gamma	90.000		
O1	0.275	5.525	13.546
O2	0.258	1.277	15.797
O3	2.989	4.172	13.702
O4	2.973	2.628	15.957
H1	0.270	0.314	14.354
H2	0.276	0.339	12.791
H3	4.890	1.836	15.900
H4	1.055	1.840	15.901
H5	2.192	4.735	13.599
H6	3.788	4.732	13.599
H7	2.978	3.213	15.147
H8	2.972	3.244	16.709

Crystallographic Information of the C_{dh}

cell_length_a	7.936		
cell_length_b	7.936		
cell_length_c	33.000		
cell_angle_alpha	90.000		
cell_angle_beta	90.000		
cell_angle_gamma	90.000		
O1	1.100	5.354	17.017
O2	5.084	5.377	18.577
O3	3.275	6.888	17.021
O4	7.245	6.930	18.528
O5	0.965	2.871	18.464
O6	3.362	1.366	18.527
O7	4.978	2.924	17.073
O8	7.327	1.349	16.921
H1	0.446	5.892	17.521
H2	1.131	4.486	17.480
H3	4.422	5.900	18.068
H4	5.133	4.511	18.112
H5	2.458	6.299	16.978
H6	3.535	7.069	16.103
H7	6.432	6.339	18.595
H8	7.511	7.142	19.438
H9	0.346	2.309	17.900
H10	0.492	3.048	19.294
H11	2.492	1.825	18.592
H12	3.207	0.515	18.060
H13	4.523	3.148	16.244
H14	4.337	2.348	17.598
H15	6.467	1.828	16.880
H16	7.166	0.510	17.408

Crystallographic Information of the C_{ps1}

cell_length_a	7.322		
cell_length_b	8.603		
cell_length_c	33.000		
cell_angle_alpha	90.000		
cell_angle_beta	90.000		
cell_angle_gamma	90.000		
O1	7.089	7.414	18.654
O2	0.059	3.263	16.375
O3	6.347	4.676	18.671
O4	4.137	8.018	18.625
O5	0.504	0.477	16.380
O6	3.455	1.097	16.353
O7	2.686	3.825	16.268
O8	3.721	5.230	18.553
H1	0.453	7.578	19.321
H2	6.947	6.433	18.647
H3	6.963	3.653	17.174
H4	0.055	2.280	16.476
H5	6.535	4.218	19.507
H6	5.347	4.814	18.644
H7	3.929	0.063	17.917
H8	5.112	8.022	18.721
H9	0.285	8.443	17.095
H10	1.479	0.469	16.293
H11	3.304	2.075	16.339
H12	4.160	0.928	15.705
H13	2.880	4.266	15.424
H14	1.687	3.684	16.288
H15	3.703	6.215	18.475
H16	3.299	4.850	17.751

Crystallographic Information of the C_{ps2}

cell_length_a	8.662		
cell_length_b	7.272		
cell_length_c	33.000		
cell_angle_alpha	90.000		
cell_angle_beta	90.000		
cell_angle_gamma	90.000		
O1	7.223	3.496	18.357
O2	1.908	6.977	16.661
O3	8.584	0.960	18.634
O4	4.814	1.873	18.845
O5	0.488	4.553	16.367
O6	2.911	2.946	16.841
O7	4.262	5.488	16.579
O8	6.239	6.721	18.555
H1	7.281	4.137	19.089
H2	7.790	2.721	18.590
H3	1.442	0.254	17.325
H4	1.304	6.205	16.514
H5	0.290	0.742	19.508
H6	7.812	0.341	18.513
H7	4.070	2.133	18.262
H8	5.522	2.525	18.652
H9	8.404	4.290	16.946
H10	1.197	3.902	16.559
H11	3.475	3.728	16.620
H12	2.978	2.316	16.101
H13	4.629	5.708	15.706
H14	3.485	6.100	16.699
H15	5.634	0.219	18.707
H16	5.773	6.178	17.887

Crystallographic Information of the C_{psh}

cell_length_a	7.700		
cell_length_b	11.400		
cell_length_c	33.000		
cell_angle_alpha	90.000		
cell_angle_beta	90.000		
cell_angle_gamma	90.000		
O1	5.230	10.441	13.952
O2	3.351	10.026	15.919
O3	6.901	8.060	13.605
O4	1.329	8.115	15.578
O5	2.835	5.946	14.451
O6	3.045	3.181	15.455
O7	5.637	3.480	14.449
O8	5.567	6.072	15.066
O9	6.579	0.902	15.333
O10	1.619	0.751	14.908
H1	2.047	8.768	15.766
H2	5.759	11.189	14.375
H3	4.866	10.785	13.120
H4	4.022	10.100	15.184
H5	3.858	10.108	16.743
H6	0.804	8.029	16.390
H7	6.380	8.889	13.665
H8	0.019	8.224	14.116
H9	2.591	5.054	14.767
H10	3.158	3.275	16.416
H11	3.963	3.220	15.077
H12	5.749	4.462	14.671
H13	5.720	3.429	13.482
H14	6.041	6.748	14.518
H15	4.604	6.186	14.868
H16	2.201	6.580	14.848
H17	2.101	1.593	15.059
H18	2.161	0.058	15.340
H19	6.349	1.836	15.141
H20	7.563	0.837	15.221