Supporting Information for "Two-dimensional Ice-like water adlayers on a mica surface with and without a graphene coating under ambient conditions"

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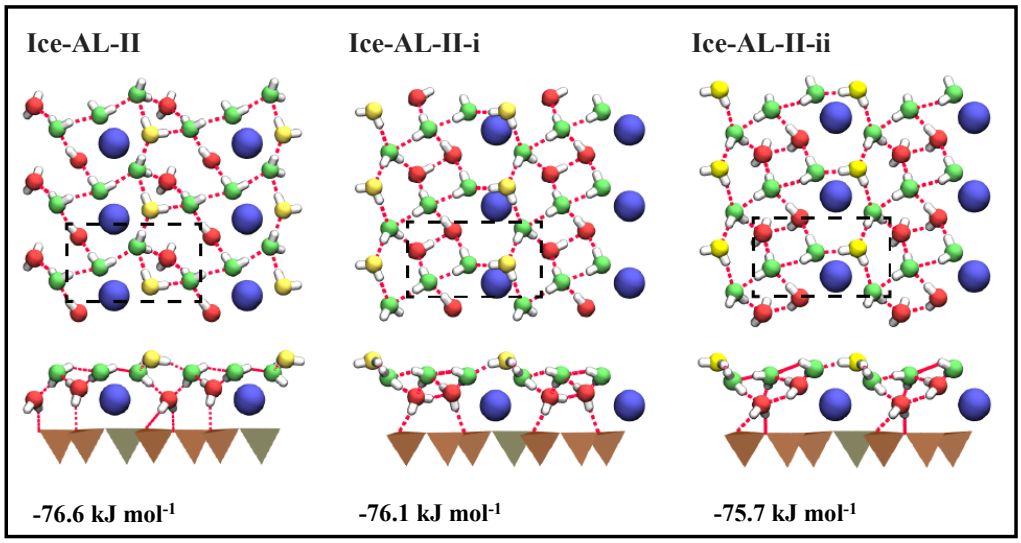
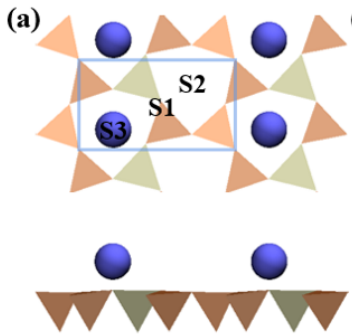


Fig. S1 Three configurations observed in the AIMD simulation of water on mica surface have the same lattice of oxygen atoms as Ice-AL-II, but have different H-bonds networks. The binding energy per water is shown below each configuration.



(b)

Adsorption site	S1	S2	S3
Adsorption energy (kJ mol^{-1})	-68.7	-58.1	-22.7

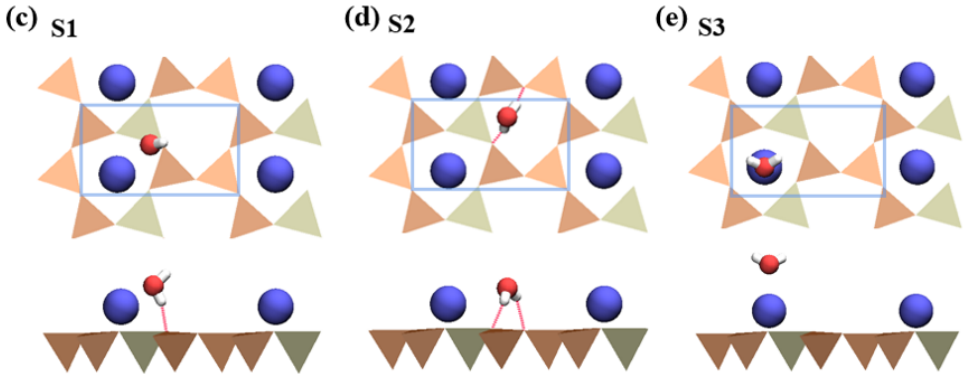


Fig. S2 (a) Schematic representation of the three adsorption sites on the mica surface, i.e. S1, S2, and S3. (b) Adsorption energies of a water molecule adsorbed in these three adsorption sites and the corresponding adsorption configurations (c-e). Unit cell are indicated by blue rectangles.

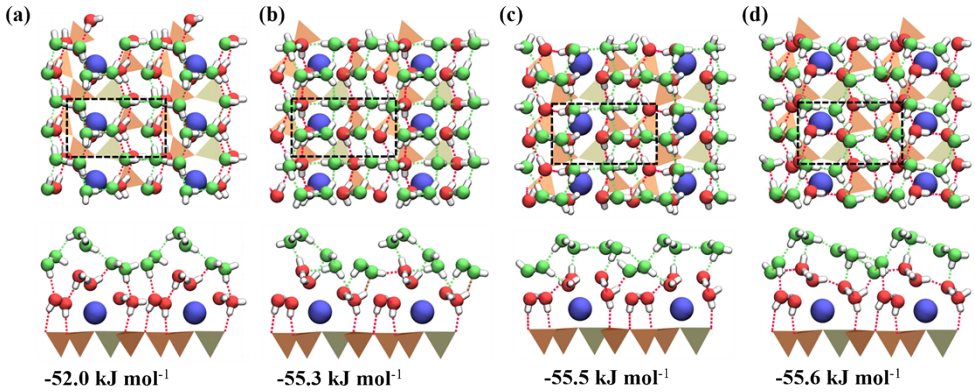


Fig. S3 Top and side views of the optimized AA-stacked (a) and AB-stacked (b-d) bilayer Ice-AL-I structures on mica surface.

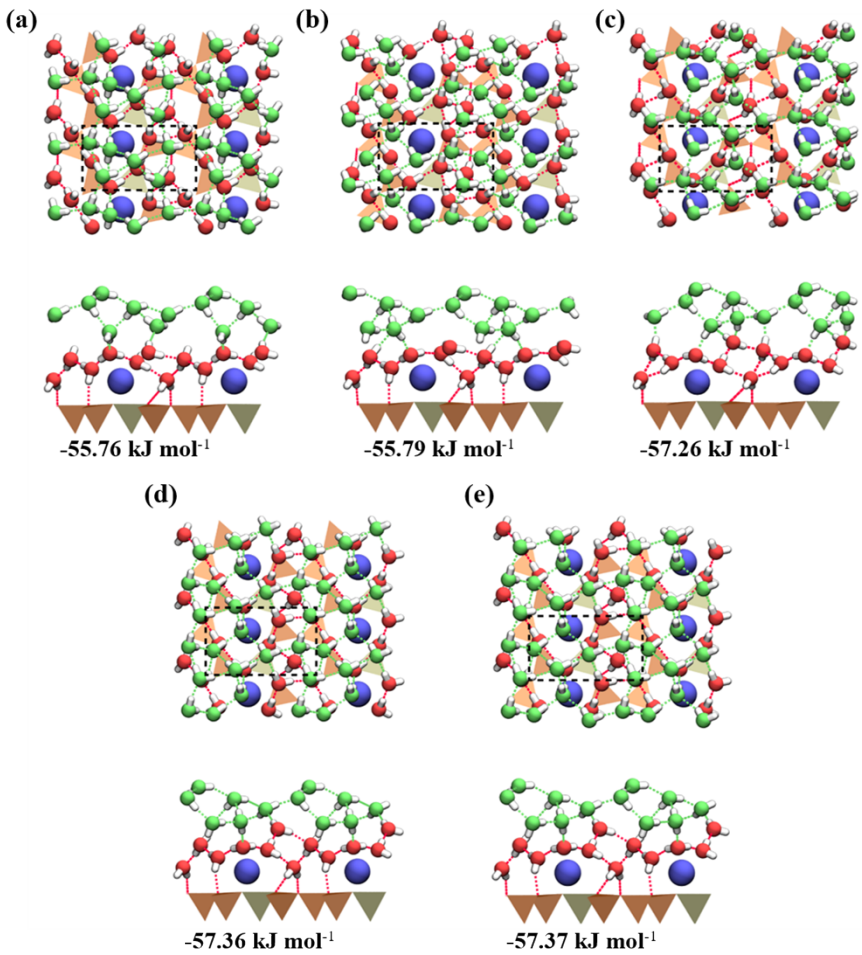
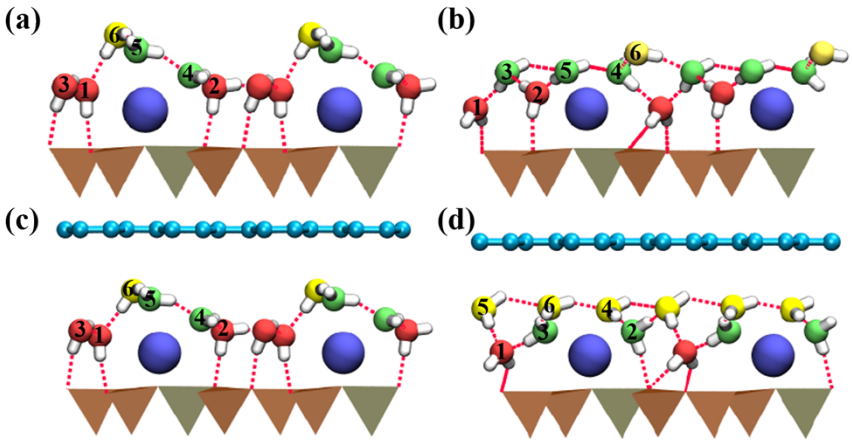


Fig. S4 Top and side views of the optimized AB-stacked bilayer Ice-AL-II structures on mica surface.



(e)

The monolayer ice				
ΔE_{int} (kJ mol^{-1})	Ice-AL-I	Ice-AL-II	Ice-AL-I	Ice-AL-II
water-1	-105.2	-142.3	-107.7	-123.7
water-2	-109.5	-106.9	-109.2	-106.2
water-3	-72.7	-103.9	-72.9	-104.7
water-4	-117.7	-120.6	-119.5	-105.6
water-5	-98.7	-82.0	-105.1	-107.0
water-6	-61.7	-78.5	-70.1	-101.3

Fig. S5 Calculated interaction energies (ΔE_{int}, in kJ mol^{-1}) of a water molecule in the unit cell with the remaining water molecules as well as the mica substrate (and graphene) for monolayer Ice-AL-I and monolayer Ice-AL-II. Structures of Ice-AL-I without (a) and with (c) a graphene coating. Structures of monolayer Ice-AL-II without (b) graphene coating and monolayer Ice-AL-II with (d) a graphene coating are shown, respectively. The water molecules in the unit cell are marked by numbers. (e) The calculated ΔE_{int} for all water molecules in the unit cell shown in (a-d).

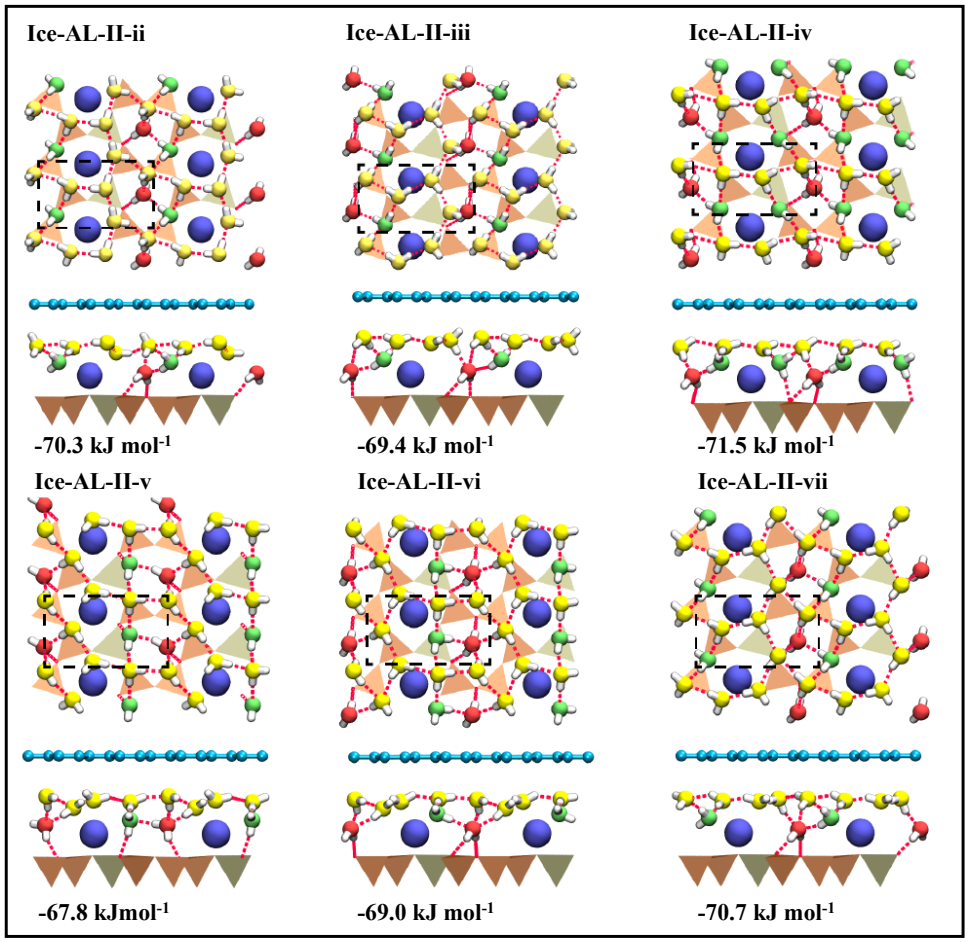


Fig. S6 Six configurations observed in the AIMD simulation of water on mica surface with a graphene coating have the same lattice of oxygen atoms as Ice-AL-II, but have different H-bonds networks. The binding energy per water is shown below each configuration. For the sake of clarity, graphene is not shown in the top view.

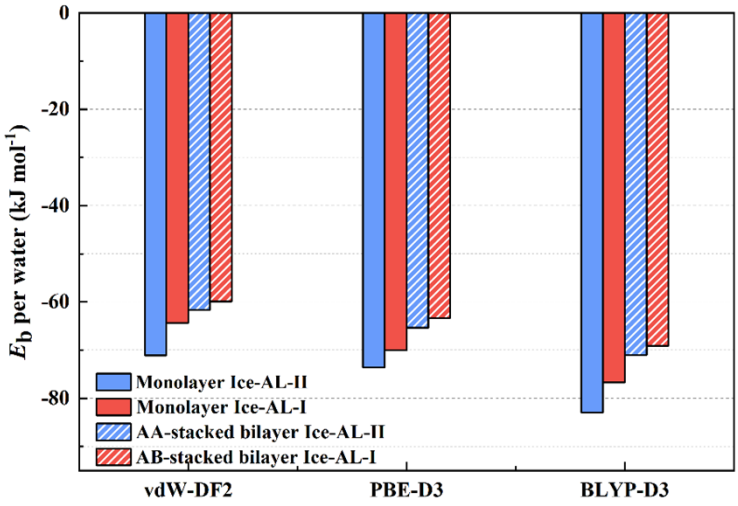
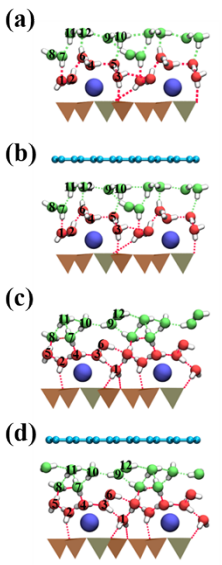


Fig. S7 Calculated binding energies per water of Ice-AL-II and Ice-AL-I on mica surface with a graphene coating.



(e)

The bilayer ice				
ΔE_{int} (kJ mol^{-1})	Ice-AL-I	Ice-AL-II	Ice-AL-I	Ice-AL-II
water-1	-111.6	-138.6	-113.0	-139.4
water-2	-109.5	-99.6	-109.7	-99.5
water-3	-132.1	-134.6	-128.0	-135.5
water-4	-110.3	-110.0	-113.8	-110.2
water-5	-120.0	-120.9	-115.8	-120.5
water-6	-113.1	-86.4	-114.9	-87.2
water-7	-107.9	-121.4	-111.8	-123.0
water-8	-92.9	-123.2	-92.4	-120.7
water-9	-96.8	-112.8	-106.2	-116.9
water-10	-104.1	-79.8	-107.3	-91.7
water-11	-94.9	-96.5	-107.8	-104.9
water-12	-102.4	-75.0	-112.5	-81.3

Fig. S8 Optimized Structures of the AB-stacked bilayer Ice-AL-I structure without (a) and with (b) a graphene coating and AA-stacked bilayer Ice-AL-II structure without (c) and with (d) a graphene coating. The water molecules in the unit cell are marked by numbers. (e) The calculated ΔE_{int} for all water molecules in the unit cell shown in (a-d).

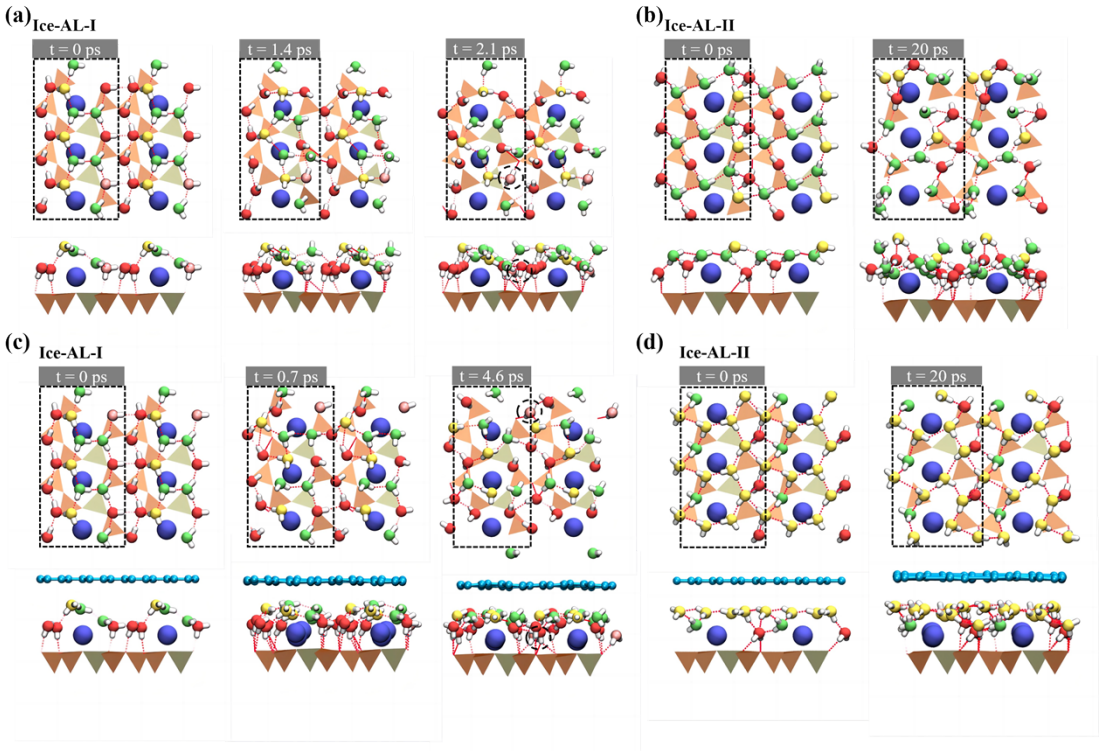


Fig. S9 Stability of Ice-AL-I and Ice-AL-II in a relatively large system. Top and side views of snapshot structures taken from the AIMD simulation of monolayer (a) Ice-AL-I structure and monolayer (b) Ice-AL-II structure on a mica surface at 300 K. Top and side views of snapshot structures taken from the AIMD simulation of the monolayer (c) Ice-AL-I structure and monolayer (d) Ice-AL-II structure on a mica surface with a graphene coating at 300 K. Graphene is not shown in the top view for the sake of clarity. The water molecule adsorbed at site S1 transfers to site S2 and is highlighted by a pink sphere and a dashed circle. Red dashed lines denote hydrogen bonds. The dashed boxes represent simulation cells. Color codes: K (purple), H (white), O (pink, red, green, or yellow), C (cyan).

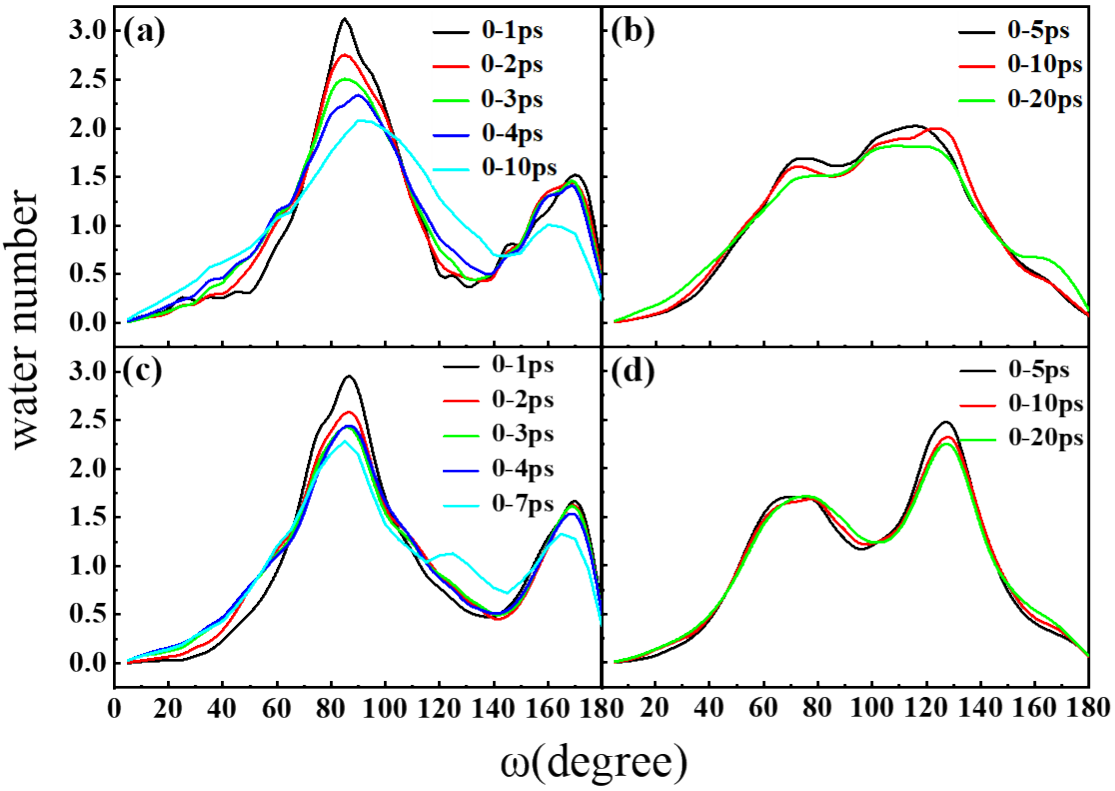


Fig. S10 Distributions of the angle (ω) between the z axis (normal to the mica surface) and the OH bonds of water molecules at 300 K for the (a) monolayer Ice-AL-I structure and (b) monolayer Ice-AL-II structure, as well as the (c) monolayer Ice-AL-I structure and (d) monolayer Ice-AL-II structure with a graphene coating.

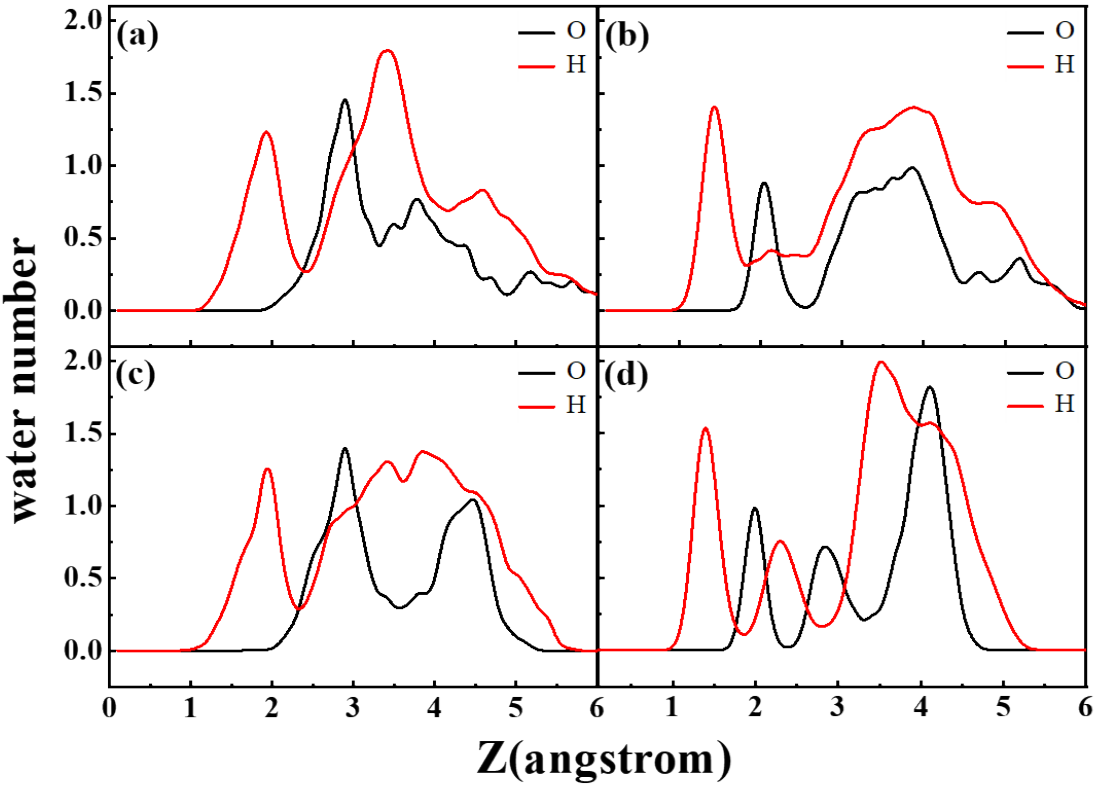


Fig. S11 Density profiles of oxygen and hydrogen of water along the z axis at 250 K for the (a) monolayer Ice-AL-I structure and (b) monolayer Ice-AL-II structure, as well as the (c) monolayer Ice-AL-I structure and (d) monolayer Ice-AL-II structure with a graphene coating.

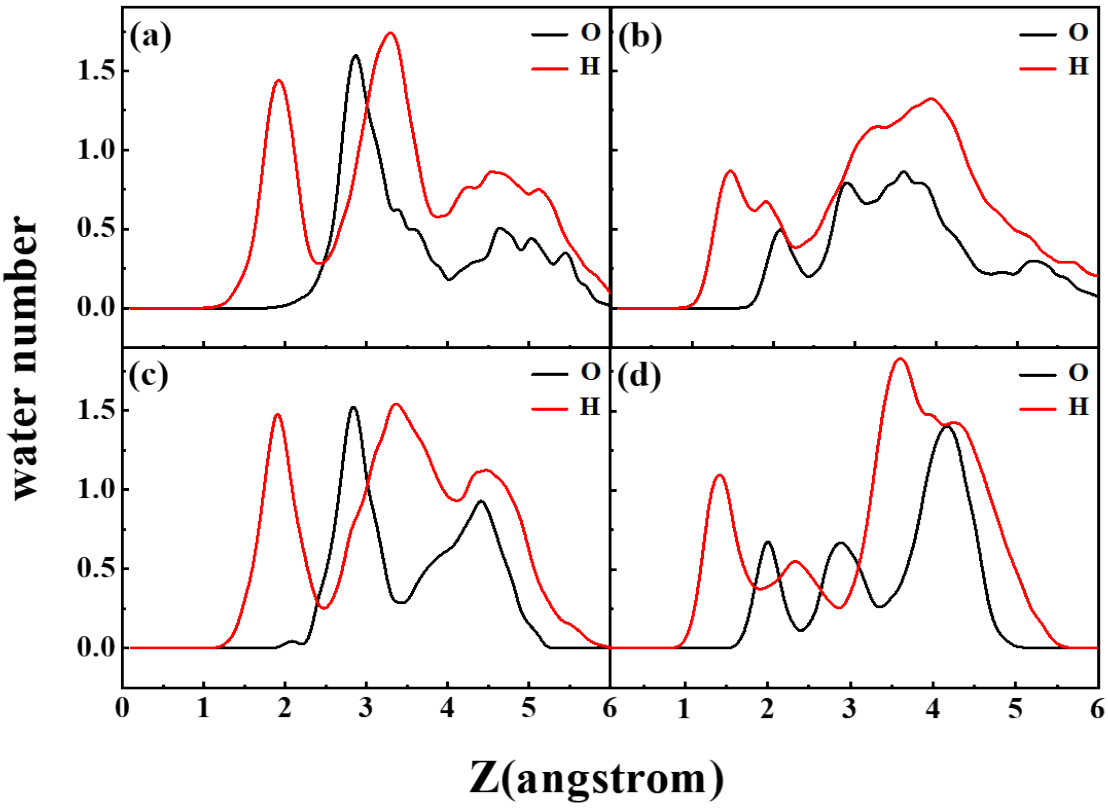


Fig. S12 Density profiles of oxygen and hydrogen of water along the z axis at 300 K for the (a) monolayer Ice-AL-I structure and (b) monolayer Ice-AL-II structure, as well as the (c) monolayer Ice-AL-I structure and (d) monolayer Ice-AL-II structure with a graphene coating.

Supporting Movies

Movie S1. Side view of spontaneous structural transition from Ice-AL-I to Ice-AL-II. The video was generated by ab initio molecular dynamics simulation of Ice-AL-I structure on mica surface at 300 K. Color codes: Si (Ocher), Al (Tan), K (purple), H (white), O (red, green or yellow).
Movie S2. Top view of spontaneous structural transition from Ice-AL-I to Ice-AL-II. The video was generated by ab initio molecular dynamics simulation of Ice-AL-I structure on mica surface at 300 K. Color codes: Si (Ocher), Al (Tan), K (purple), H (white), O (red, green or yellow).
Movie S3. Side view of spontaneous structural transition from Ice-AL-I to Ice-AL-II. The video was generated by ab initio molecular dynamics simulation of Ice-AL-I structure on mica surface with a graphene coating at 300 K. Color codes: Si (Ocher), Al (Tan), K (purple), H (white), O (red, green or yellow), C(cyan).
Movie S4. Top view of spontaneous structural transition from Ice-AL-I to Ice-AL-II. The video was generated by ab initio molecular dynamics simulation of Ice-AL-I structure on mica surface with a graphene coating at 300 K. Graphene is not shown for the sake of clarity. Color codes: Si (Ocher), Al (Tan), K (purple), H (white), O (red, green or yellow).
Movie S5. Instability of Ice-AL-I structure on mica surface (side view). The video was generated by ab initio molecular dynamics simulation of Ice-AL-I structure on mica surface at 250 K using a relatively large system. Color codes: Si (Ocher), Al (Tan), K (purple), H (white), O (red, green or yellow).
Movie S6. Instability of Ice-AL-I structure on mica surface (top view). The video was generated by ab initio molecular dynamics simulation of Ice-AL-I structure on mica surface at 250 K using a relatively large system. Color codes: Si (Ocher), Al (Tan), K (purple), H (white), O (red, green or yellow).
Movie S7. Stability of Ice-AL-II structure on mica surface (side view). The video was generated by ab initio molecular dynamics simulation of Ice-AL-II structure on mica surface at 250 K using a relatively large system. Color codes: Si (Ocher), Al (Tan), K (purple), H (white), O (red, green or yellow).
Movie S8. Stability of Ice-AL-II structure on mica surface (top view). The video was generated by ab initio molecular dynamics simulation of Ice-AL-II structure on mica surface at 250 K using a relatively large system. Color codes: Si (Ocher), Al (Tan), K (purple), H (white), O (red, green or yellow).
Movie S9. Instability of Ice-AL-I structure on mica surface with a graphene coating (side view). The video was generated by ab initio molecular dynamics simulation of Ice-AL-I structure on mica surface with a graphene coating at 250 K using a relatively large system. Color codes: Si (Ocher), Al (Tan), K (purple), H (white), O (red, green or yellow), C(cyan).
Movie S10. Instability of Ice-AL-I structure on mica surface with a graphene coating (top view). The video was generated by ab initio molecular dynamics simulation of Ice-AL-I structure on mica surface with a graphene coating at 250 K using a relatively large

system. Color codes: Si (Ocher), Al (Tan), K (purple), H (white), O (red, green or yellow).

Movie S11. Stability of Ice-AL-II structure on mica surface with a graphene coating (side view). The video was generated by ab initio molecular dynamics simulation of Ice-AL-II structure on mica surface with a graphene coating at 250 K using a relatively large system. Color codes: Si (Ocher), Al (Tan), K (purple), H (white), O (red, green or yellow), C(cyan).

Movie S12. Stability of Ice-AL-II structure on mica surface with a graphene coating (top view). The video was generated by ab initio molecular dynamics simulation of Ice-AL-II structure on mica surface with a graphene coating at 250 K using a relatively large system. Color codes: Si (Ocher), Al (Tan), K (purple), H (white), O (red, green or yellow).