Supplementary information

Interfacial Icelike Water Local Doping Graphene

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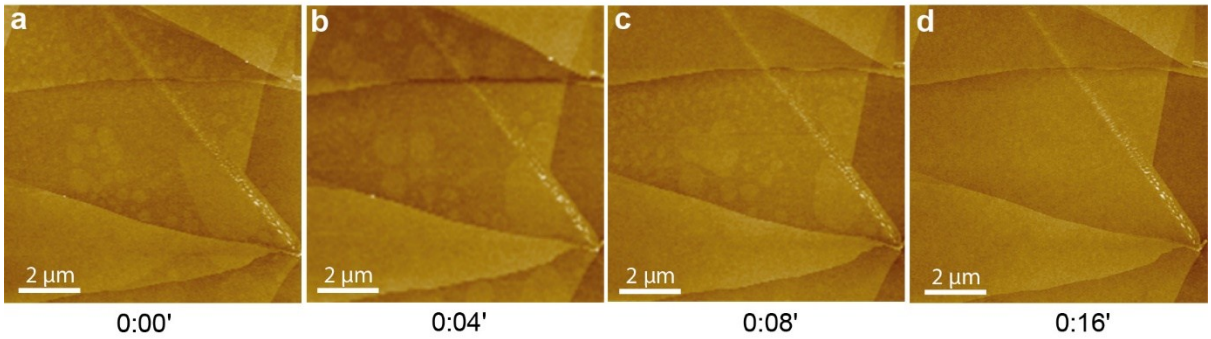

Figure S1. (a-d) *In situ* AFM height images of the interfacial water layer growth during the scanning. It can be clearly seen that the isolated interfacial water layers coalesced into a continuous water layer takes ~16 mins.

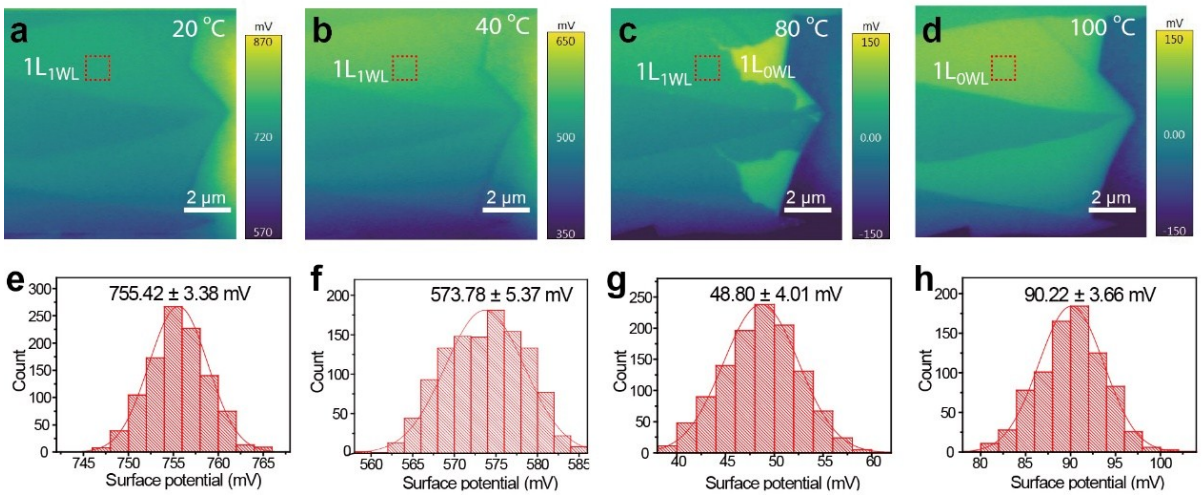

Figure S2. The surface potential of graphene supported on SiO_2/Si at different temperature. (a-d) *In situ* surface potential mapping corresponding to Figure 2 measured by SKPM. (e-h) The statistical distribution of surface potential of red dashed box area in (a-d).

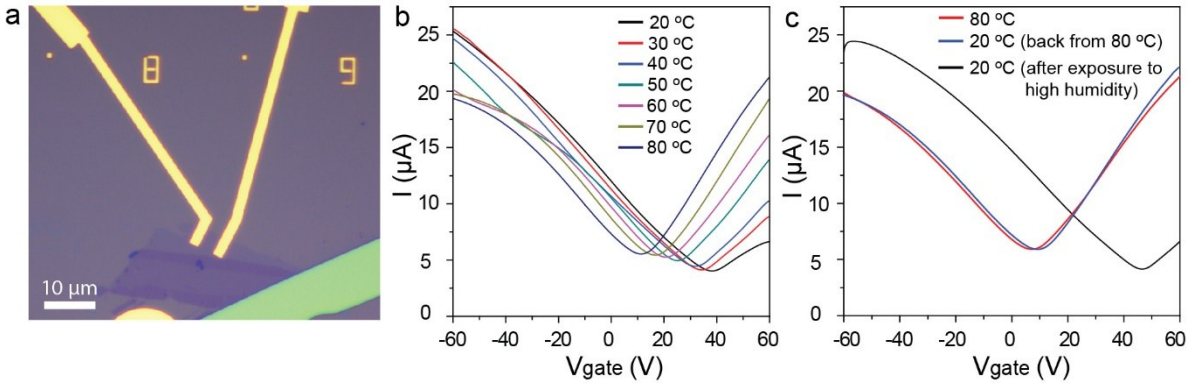

Figure S3. Graphene FET transport measurement under different temperatures. (a) Optical image of the as-fabricated graphene FET devices. (b) Source-drain current (I) as a function of back gate voltage (V_{gate}) of monolayer graphene supported on SiO_2/Si substrate measured at different temperatures under vacuum. (c) I- V_{gate} curves of monolayer graphene measured at 80 ℃, 20℃ (back from 80 ℃) and 20 ℃ (after exposure to high humidity).

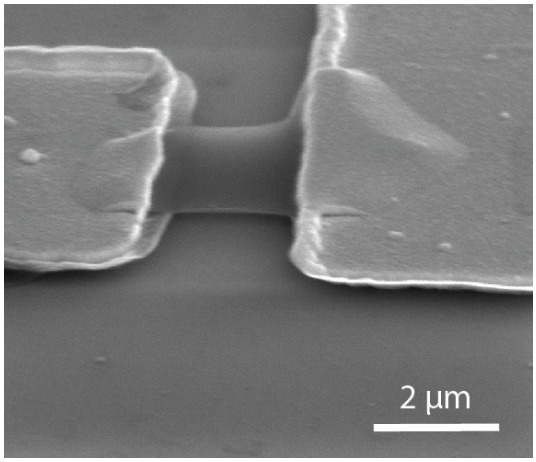

Figure S4. SEM image of suspended monolayer graphene device.

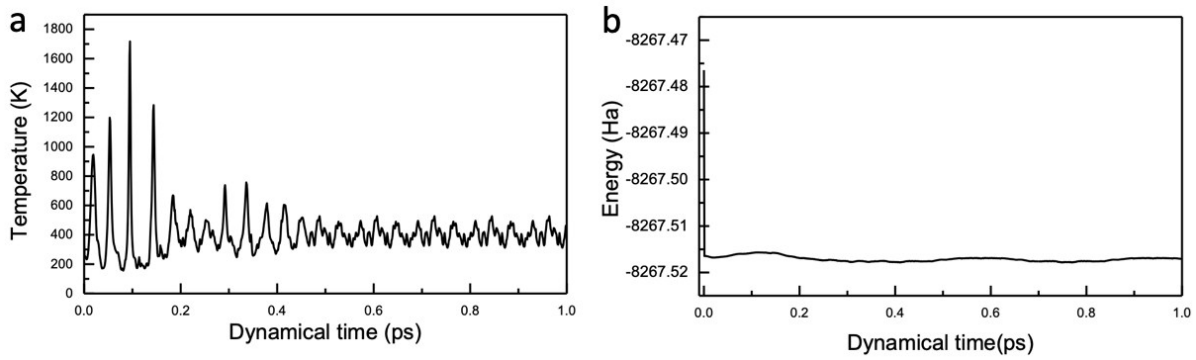

Figure S5. Ab initio molecular dynamics of $1L_{0WL}$ by Dmol^3, in the NVT ensemble and Massive GGM thermostat. The step is 0.5fs and 2000 steps totally. The temperature (a) and energy (b) reach to stability after 1ps relaxation. At the end of calculation, we found the work function at 400K is close to 4.56eV as measured by SKPM at 80 ℃.

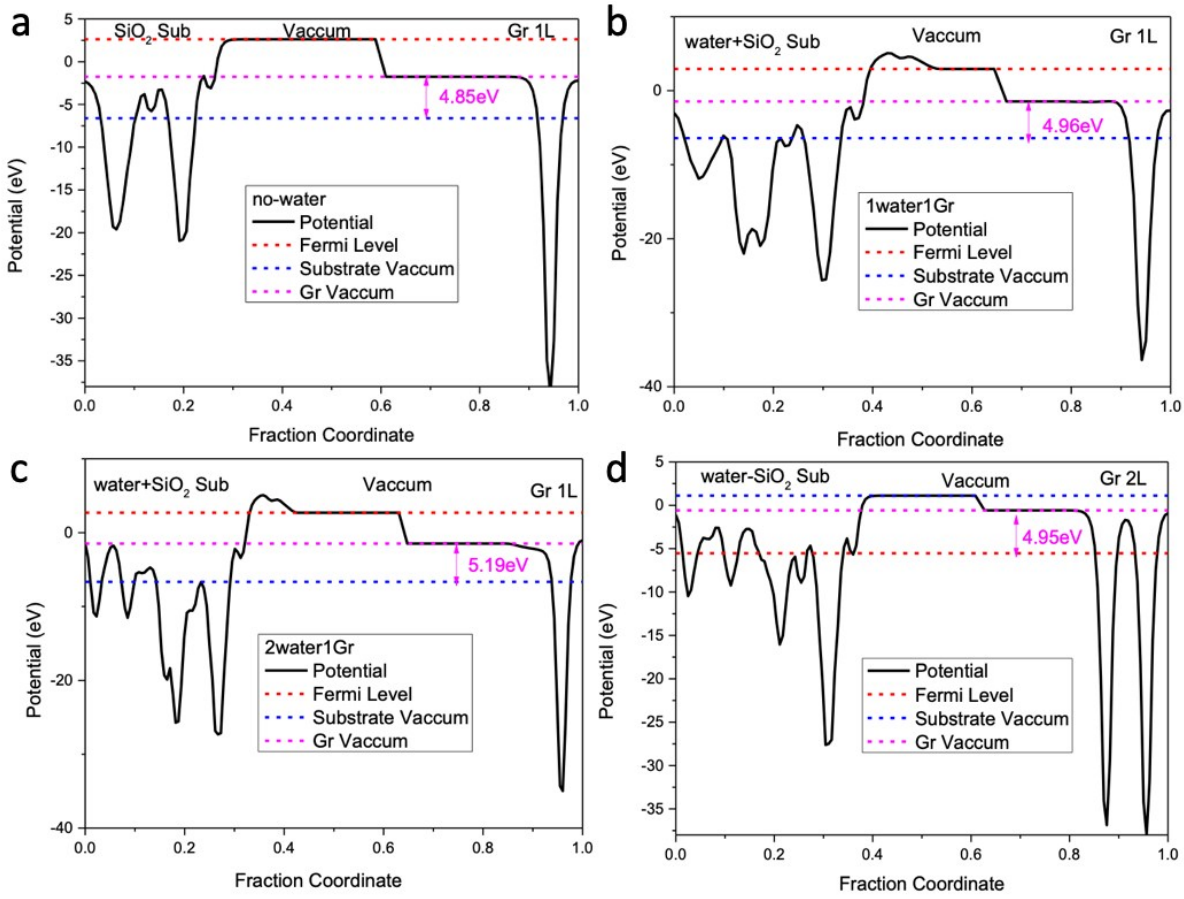

Figure S6. The electrostatic potential profile along the z-direction of four structures: (a) 1G + 0W + SiO_2, (b) 1G + 1W + SiO_2, (c) 1G + 2W + SiO_2, (d) 2G + 1W + SiO_2. The potential of water + SiO_2 substrate and the graphene layers were plot on the two sides of the profile. The potential of vacuum slab was plot in the middle. There are two potential energies for vacuum slab - one is for the substrate and the other for the graphene. The work function of graphene is the energy difference between the vacuum energy (Gr) and the Fermi level.

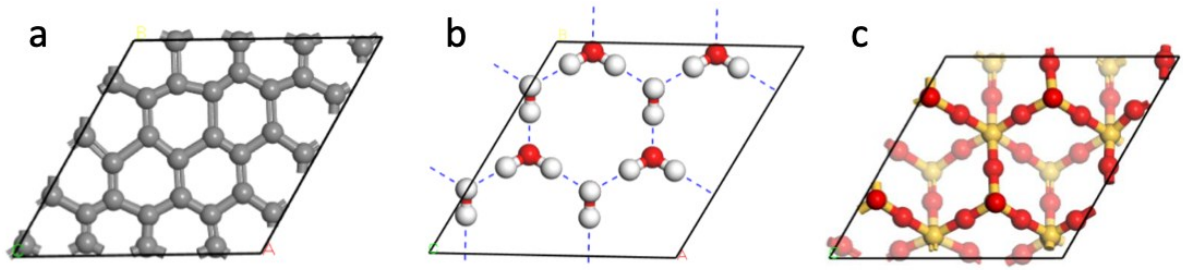

Figure S7. Top-view of the supercells of (a) monolayer graphene (4×4), (b) monolayer water (2×2), and (c) three-layer SiO_2 substrate (2×2).

Table S1. The lattice parameters of the combined unit cell

a	b	α	β	γ
9.032 Å	9.032 Å	90^{o}	90^{o}	60^{o}