Supporting Information

Thick two-dimensional water film confined between atomically-thin mica nanosheet and hydrophilic substrate

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1. Height profiles of mica nanosheet and confined water film

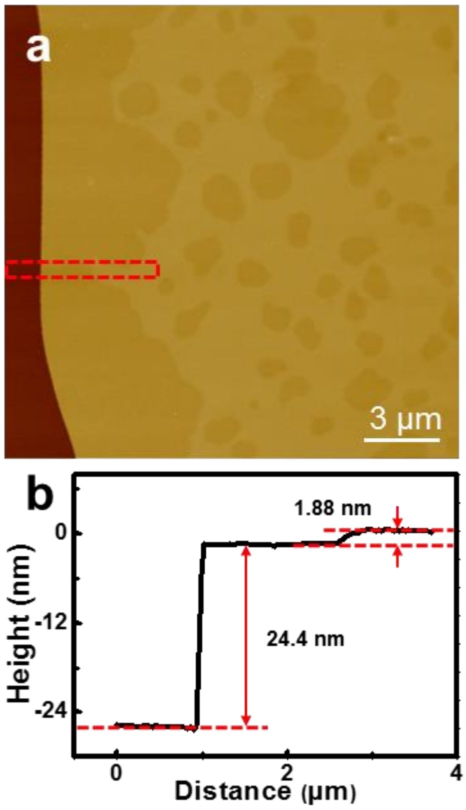


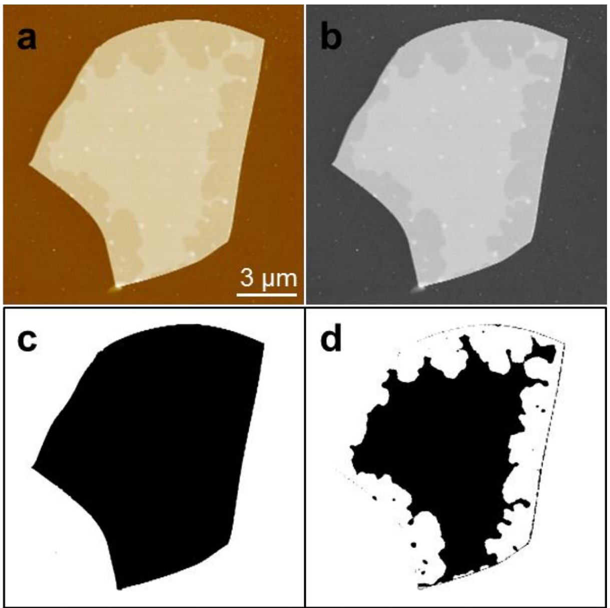
Figure S1. (a) AFM image of mica nanosheet and water film. (b) The height profiles of mica nanosheet and water film marked by red box in (a), respectively.

2. Measurement of the coverage of mica flake by confined water adlayer.

1) Open ImageJ software.
2) Click File → Open in ImageJ to open an AFM image of confined water film (Figure S2a).
3) Click Image → Color→Split Channels. Choose a suitable channel with the most obvious contrast (Figure S2b).
4) Click Image → Adjust → Threshold. Adjust suitable threshold value to mark all mica nanosheet on substrate in black (Figure S2c).
5) Click Analyze → Measure, the area coverage of SiO_2 substrate covered by mica nanosheet (Sm) is shown in %Area (Figure S2e).
6) Repeat steps 4-5 to measure the area coverage of SiO_2 substrate covered by confined water film (Sw), which is shown in Figures S2d.

The area coverage (AC) is defined as the area ratio of confined water film to the corresponding mica flake, which can be described by the following equation,

$$AC = \frac{S_w}{S_m} \times 100\%$$



e

	Area	Mean	Min	Max	%Area
1	6250000	130.745	53	252	46.287
2	6250000	130.745	53	252	27.272

Figure S2. Measurement of the area ratio of confined water film to the mica nanosheet. (a) AFM image of confined water film and mica nanosheet (RH = 57%). (b) Green channel of the AFM image shown in (a). (c-d) The area percentage of SiO_2 substrate covered by mica nanosheet and confined water film, respectively. (e) The measured %area of mica nanosheet and confined water film by ImageJ, respectively.

3. Calculating the volume of water droplets and films.

3.1 Calculating the volume of water droplets

As shown in Figure S3, the height of water droplet is ~ 7 nm. While the width of droplet is usually larger than 200 nm. In this case, the water droplet can be considered as spherical cap.[1-2] In order to compensate the overestimation induced by the tip broadening effect in AFM measurement,[3-4] the width at half-maximal height (w) was well accepted to represent the diameter of droplet. Therefore, the volume of spherical cap-shaped water droplet (V) can be calculated by geometric method, which is described by equation (1),[1-2]

$$V = \frac{\pi h}{6}\left(3\left(\frac{w}{2}\right)^2 + h^2\right) \tag{1}$$

Here, V is the volume of water droplet, w is the width at half-maximal height and h is the height of water droplet, respectively.

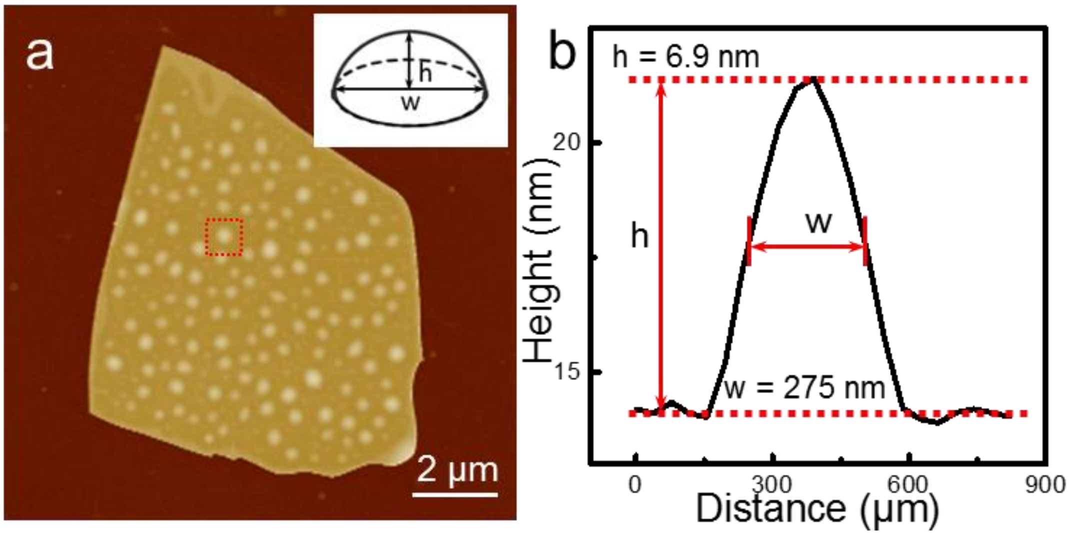


Figure S3. Calculating the volume of water droplet by the geometric method. (a) AFM image of water droplets confined under mica nanosheet. Inset shows the spherical cap model. (b) Height and width profiles of the water droplet marked by red box in (a).

Since there are too many water droplets in one AFM image, it is time-consuming and tedious to obtain the total volume of water droplets by summing the volume of each droplet one by one. In order to roughly estimate the total volume of water droplets, 20 water droplets with various height and size were randomly chosen to calculate their volume and then get the average volume of individual droplet (V_i) firstly, as shown in equation (2).

$$V_i = \frac{V_1 + V_2 + \cdots + V_{20}}{20} \tag{2}$$

After the total number of droplets in one AFM image was counted (N), the total volume of water droplets (V) in it can be calculated by equation (3) as follows,

$$V = V_i \times N \tag{3}$$

3.2 Calculation of water film volume

In terms of the volume of water films confined between mica and substrate, the situation is a bit easier due to the uniform height of water film. The area of water film (S) can be obtained according to the description shown in Figure S2. In this case, the volume of confined water film can be calculated by equation (4),

$$V = S \times h \tag{4}$$

Where V is the total volume of confined water film, and S and h are the area and height of confined water film, respectively.

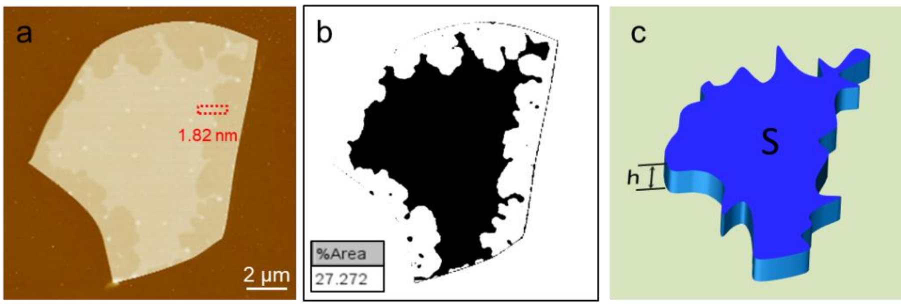


Figure S4. (a) AFM image of confined water film. (b) The area of water film calculated by Image J software. (c) Schematic volume illustration of water film.

4. **AFM images of mica nanosheets incubated at 11%, 33%, 75% and 98% RH for 1, 5, 10, 30 and 60 min**

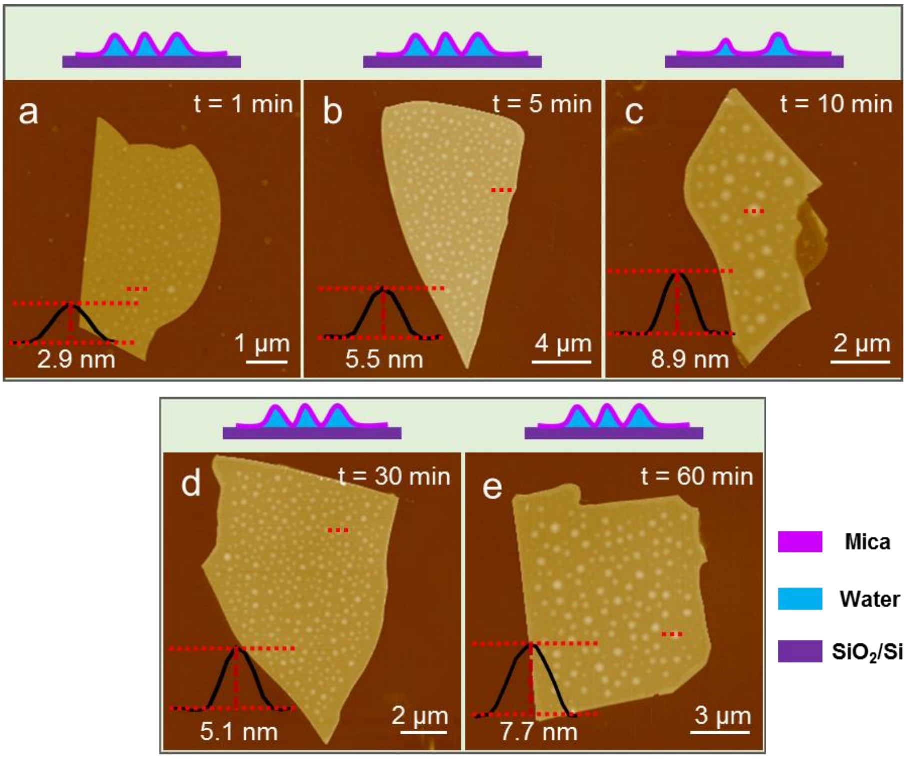


Figure S5. (a-e) AFM images of mica nanosheets deposited on SiO_2/Si substrate before being incubated at 11% RH for 1, 5, 10, 30 and 60 min, respectively. The schemes above each AFM image show the schematic illustration of confined water from front view.

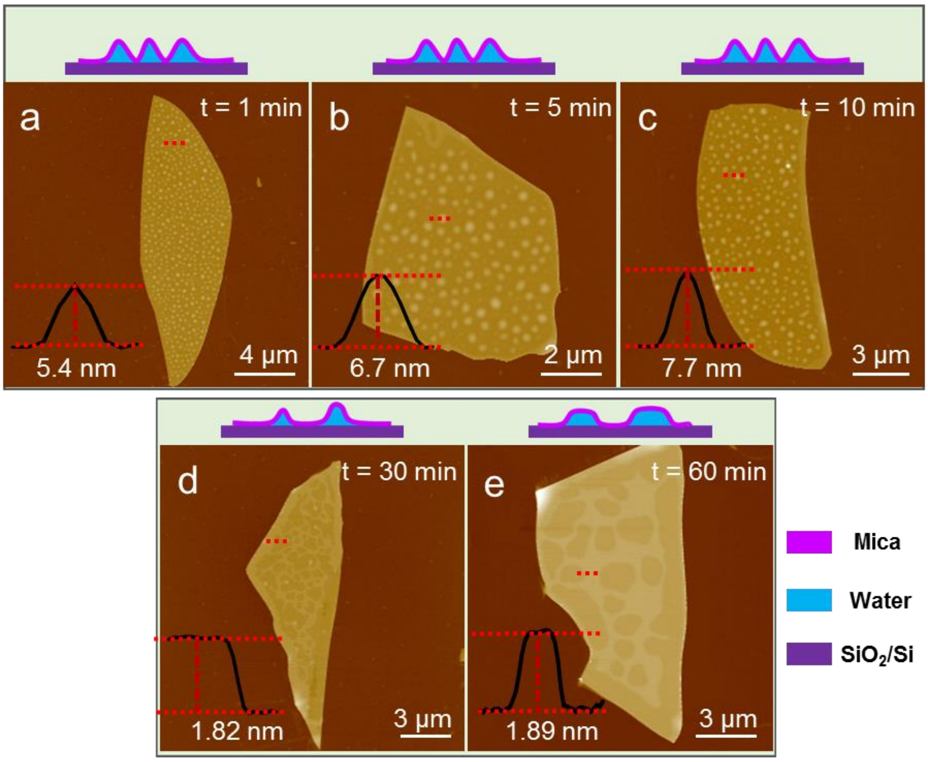


Figure S6. (a-e) AFM images of mica nanosheets deposited on SiO_2/Si substrate before being incubated at 33% RH for 1, 5, 10, 30 and 60 min, respectively. The schemes above each AFM image show the schematic illustration of confined water from front view.

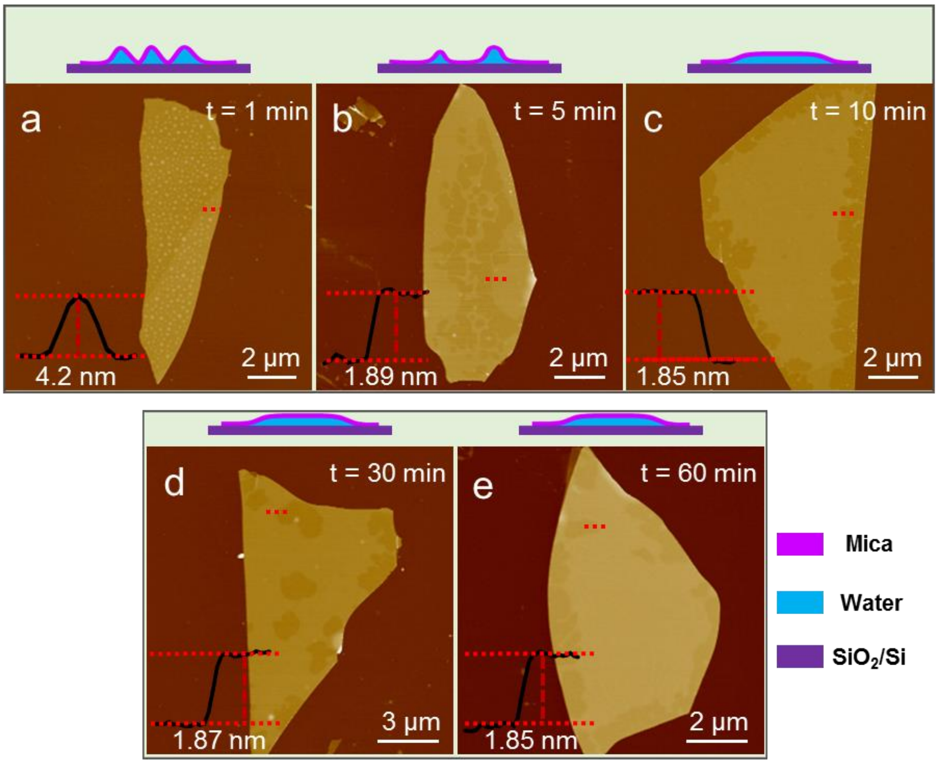


Figure S7. (a-e) AFM images of mica nanosheets deposited on SiO_2/Si substrate before being incubated at 75% RH for 1, 5, 10, 30 and 60 min, respectively. The schemes above each AFM image show the schematic illustration of confined water from front view.

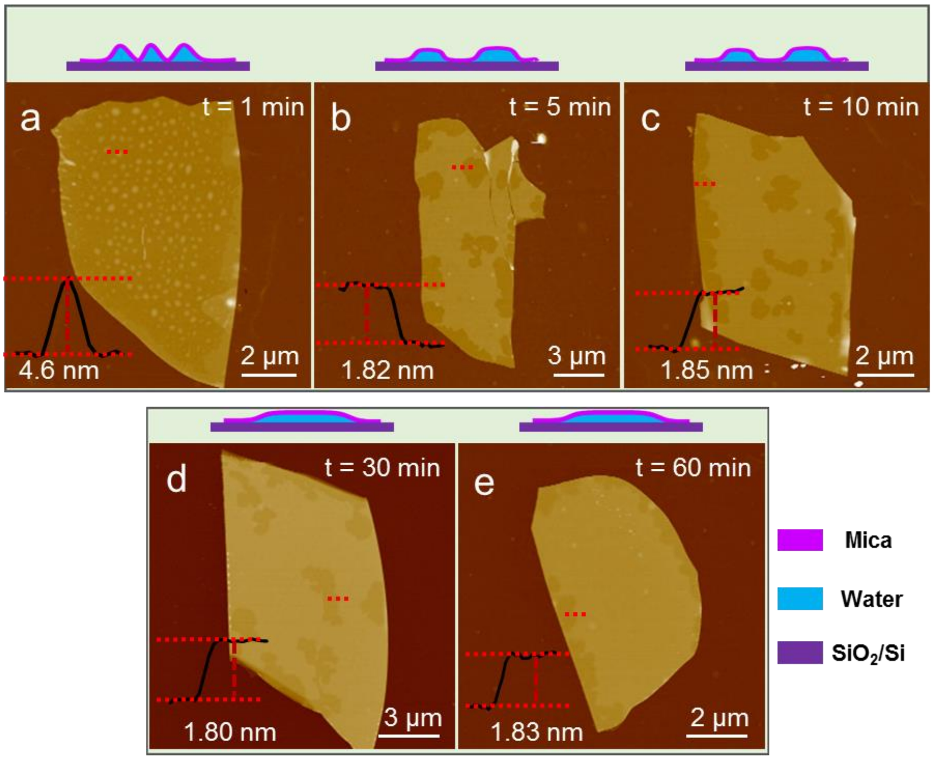


Figure S8. (a-e) AFM images of mica nanosheets deposited on SiO_2/Si substrate before being incubated at 98% RH for 1, 5, 10, 30 and 60 min, respectively. The schemes above each AFM image show the schematic illustration of confined water from front view.

5. AFM images of confined water droplets incubated at 11% RH

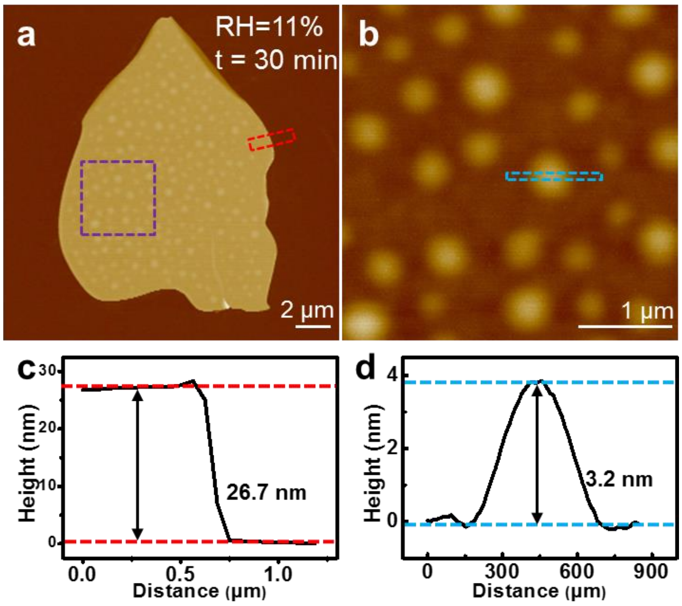


Figure S9. (a) AFM image of droplets between mica and SiO_2/Si substrate. (b) Magnified AFM image of (a). (c-d) Height profiles of the region marked by red and blue boxes shown in (a) and (b), respectively.

6. Stability of confined water film under ambient conditions

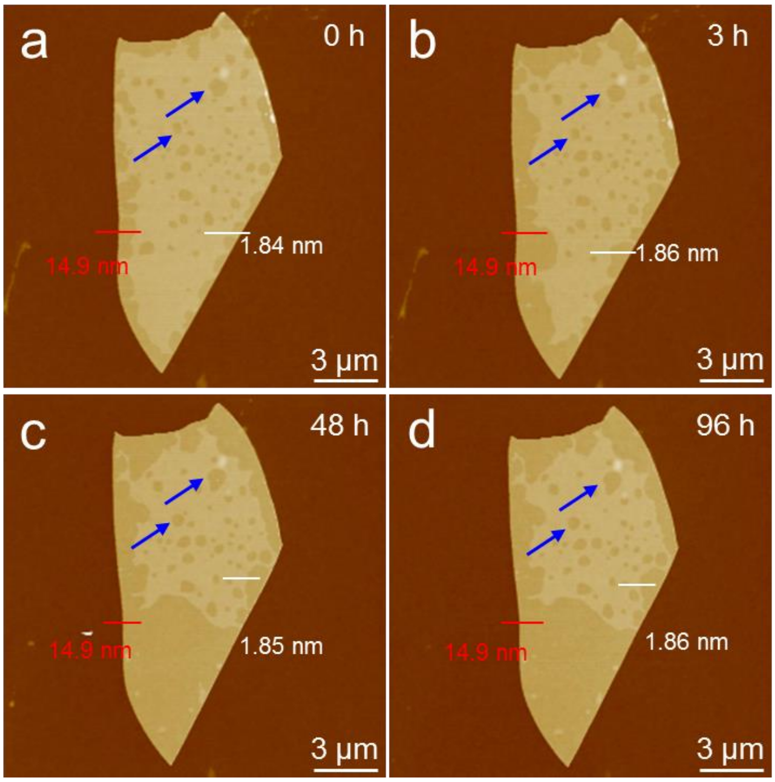


Figure S10. Stability of confined water film under ambient conditions. AFM images of confined water film with storage time of (a) 0, (b) 3, (c) 48 and (d) 96 h, respectively. The thicknesses of the mica nanosheet and water film were kept at 14.9 and 1.86 nm even after 96 h, respectively.

7. Stability of confined water film at high temperature

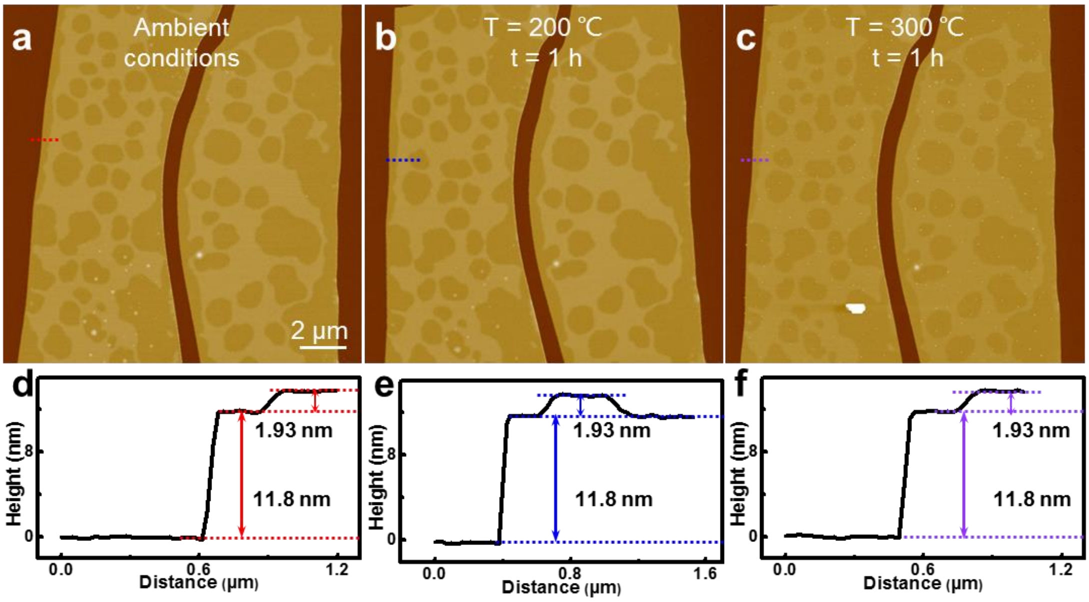


Figure S11. AFM images of confined water films (a) before and after being annealed at (b) 200℃ and (c) 300℃ for 1 h, respectively. (d-f) Corresponding height profiles of confined water film and mica nanosheet shown in (a), (b) and (c), respectively.

8. Influence of mica thickness on confined water film

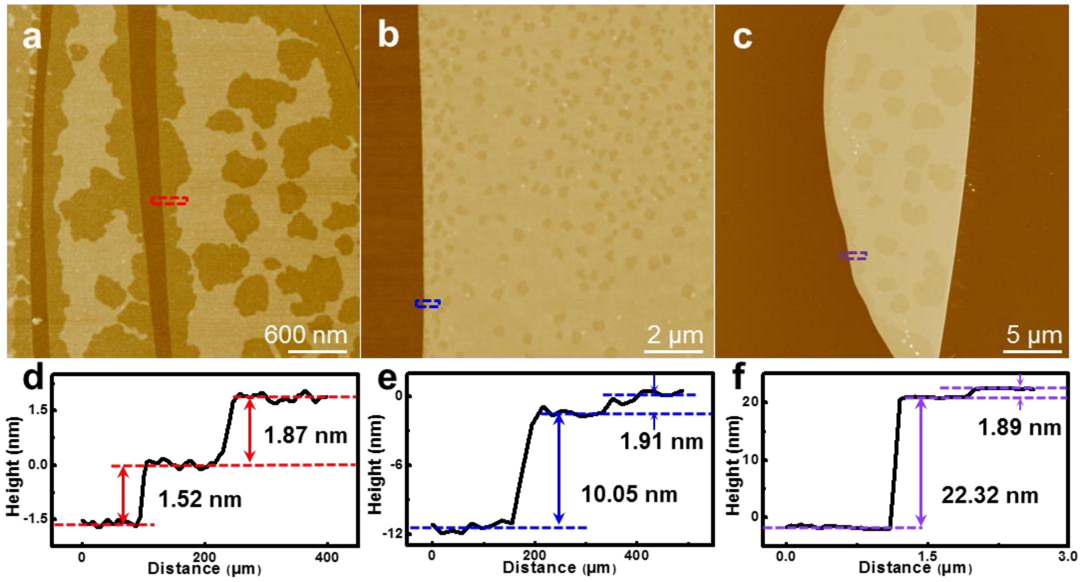


Figure S12. AFM images of confined water films between (a) thin (1.5 nm), (b) mid-thick (10 nm) and (c) thick (22 nm) mica nanosheets and SiO_2/Si substrate under ambient conditions. (d-f) Corresponding height profiles of the region marked by red, blue and purple boxes shown in (a), (b) and (c), respectively.

9. Contact angle measurement

Table S1 Contact angles of different substrates.

Substrates	Contact angle(°)
Hydrophilic-SiO_2	5.8
Mica	7.5
APS-SiO_2	49.2
MoS_2	74.5
HOPG	83.9
OTS-SiO_2	108.5

Reference:

(1) Fuentes-Perez, M. E.; Dillingham, M. S.; Moreno-Herrero, F. AFM Volumetric Methods for the Characterization of Proteins and Nucleic Acids. *Methods* **2013**, *60*, 113-121.

(2) Schneider, S. W.; Larmer, J.; Henderson, R. M.; Oberleithner, H. Molecular Weights of Individual Proteins Correlate with Molecular Volumes Measured by Atomic Force Microscopy. *Pflug Arch Eur J Phy* **1998**, *435*, 362-367.

(3) Ramirez-Aguilar, K. A.; Rowlen, K. L. Tip Characterization from AFM Images of Nanometric Spherical Particles. *Langmuir* **1998**, *14*, 2562-2566.

(4) Markiewicz, P.; Goh, M. C. Atomic Force Microscopy Probe Tip Visualization and Improvement of Images Using a Simple Deconvolution Procedure. *Langmuir* **1994**, *10*, 5-7.