Additional graphene layer height information

Figure S1(a) shows a zoomed-out image of the scan presented in Figure 4 in the main text. The scanned area in the main text is indicated by a blue dashed square. The layered structure of the few layer graphene area can be clearly observed. Figure S1(b) shows the height profile of the red line in Figure S1(a). The stepwise increase in height can clearly be observed and corresponds to \(~0.34\) nm, which is the height of single layer graphene.

Additional fractal shape information

Figure S2 shows an additional image where the influence of the layer thickness on the alcohol island shape can be observed. The alcohol molecules form small islands under single layer graphene. Under thicker graphene blankets, in this case 3 layers graphene, the fractal shape is
Figure S1: (a) AFM topography image. (b) Height profile of the red line in (a).
better preserved. This is attributed to different line tensions for different graphene thickness.

Figure S2: Topography image of intercalated 2-propanol fractals.

Lateral force calibration

The friction force $F_L$ can be determined by using the improved wedge calibration method of Varenberg et al. This method is based on the wedge calibration method of Ogletree et al. In this method, the half-width of the experimentally observed friction loop $W_{exp}$ is converted to the friction force with a lateral calibration constant $\alpha$, see equation (1):

$$F_L = \alpha W_{exp}$$  \hspace{1cm} (1)

This method is a direct calibration method, which means that no knowledge of the lateral spring constant $k_L$ is required.
Lateral calibration constant

The lateral calibration constant $\alpha$ can be obtained by determining several frictional characteristics of a calibration sample. We used a TGF11 silicon calibration grating (MikroMasch), which has trapezoidal steps under an angle $\theta$ of 54.74°. The following frictional characteristics of the calibration sample have been determined:

- $W_{\text{slope}}$, the half-width of the friction loop on the sloped surface
- $W_{\text{flat}}$, the half-width of the friction loop on the flat surface
- $\Delta$, the friction loop offset of the sloped surface compared to the flat surface
- $F_N$, the normal force
- $F_A$, the adhesion force

From these parameters the friction coefficient of the sloped surface $\mu$ can be determined using equation (2):

$$\sin \theta (F_N \cos \theta + F_A) \mu^2 - \frac{\Delta}{W_{\text{slope}}} (F_N + F_A \cos \theta) \mu + F_N \sin \theta \cos \theta = 0 \quad (2)$$

This second order polynomial equation can be solved with e.g. Matlab and has two solutions. The real solution must be smaller than $1 / \tan(\theta)$. If both values of $\mu$ satisfy this condition, both solutions have to be inserted in equation (3) to obtain $\alpha$:

$$\alpha = \frac{1}{W_{\text{slope}}} \frac{\mu (F_N + F_A \cos \theta)}{(\cos^2 \theta - \mu^2 \sin^2 \theta)} \quad (3)$$

When two values for $\alpha$ are obtained, these values have to be inserted in equation (4):

$$\mu_{\text{flat}} = \frac{\alpha W_{\text{flat}}}{F_N + F_A} \quad (4)$$

where $\mu_{\text{flat}}$ is the friction coefficient of the flat surface. The $\alpha$ value which results in the smallest difference between $\mu_{\text{flat}}$ and $\mu$ should be taken as the lateral calibration constant.
Normal force

The normal force $F_N$ to the sample during the measurement is determined from equation (5):

$$F_N = k_N S_N U_N,$$

(5)

where $k_N$ is the normal spring constant of the cantilever, $S_N$ is the normal deflection sensitivity and $U_N$ is the vertical deflection voltage. $k_N$ is given by the manufacturer of the AFM tip, $S_N$ is obtained by taking the inverse slope of the force-distance curve in the contact regime and $U_N$ is the voltage difference between the contact setpoint and the scanning setpoint.

Adhesion force

The adhesion force $F_A$ of the cantilever to the sample during the calibration is determined from equation (6):

$$F_A = k_N S_N U_A,$$

(6)

where $U_A$ is the voltage difference between the contact setpoint and the voltage point when the cantilever jumps out of contact.

Example calculation

Below results for the calculation for the friction on the monolayer graphene on top of the alcohol island in Figure 4.

Calculating the normal force $F_N$ during the measurement:

Input: $k_N = 0.2$ N/m
Input: $S_N$ measurement = 16.002 nm/V
Input: $U_N$ measurement = 20.74 V
Output: $F_N$ measurement = 66.38 nN

Calculating the normal force $F_N$ during the calibration:
Input: $k_N = 0.2$ N/m
Input: $S_N$ calibration = 15.887 nm/V
Input: $U_N$ calibration = 5.57 V
Output: $F_N$ calibration = 17.70 nN

Calculating the adhesion force during the calibration:
Input: $k_N = 0.2$ N/m
Input: $S_N$ calibration = 15.887 nm/V
Input: $U_A$ calibration = 0.88 V
Output: $F_A = 2.80$ nN

Calculating the lateral calibration constant $\alpha$:
Input: $\theta = 54.74^\circ$
Input: $W_{slope} = 0.364$ V
Input: $W_{flat} = 0.1335$ V
Input: $\Delta = 1.0395$
Input: $F_N$ calibration = 17.70 nN
Input: $F_A = 2.80$ nN
Intermediate output: $\mu_1 = 5.0344 > 1/\tan(\theta) = 0.707$, invalid solution
Intermediate output: $\mu_2 = 0.156$
Output: $\alpha = 26.10$ nN/V
Calculating the lateral force $F_L$:

Input: $W_{exp} = 45.52$ mV

Input: $\alpha = 26.10$ nN/V

Output: $F_L = 1.19$ nN

**Associated Content**

The Supporting Information is available free of charge on the ACS Publication website.

**References**

(S1) Varenberg, M.; Etsion, I.; Halperin, G. An Improved Wedge Calibration Method for Lateral Force in Atomic Force Microscopy. *Rev. Sci. Instrum.* **2003**, *74*, 3362–3367.

(S2) Ogletree, D. F.; Carpick, R. W.; Salmeron, M. Calibration of Frictional Forces in Atomic Force Microscopy. *Rev. Sci. Instrum.* **1996**, *67*, 3298–3306.