SUPPLEMENTAL MATERIAL

Contribution of capillary adhesion to friction at macroscopic solid-solid interfaces

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I. SINGLE NANOASPERITY ADHESION CALCULATION

We calculate the maximal adhesive force that results from the capillary bridge at the tip-on-substrate interface. This adhesive force consists of three contributions; the capillary force (F_c), the tension force (F_t) and the van der Waals force (F_v), $\overrightarrow{F_{ad}} = \overrightarrow{F_c} + \overrightarrow{F_t} + \overrightarrow{F_v}$. The capillary force (Eq. (S1)) results from the pressure difference inside and outside the meniscus known as the Laplace pressure ($P_{Laplace}$), which then acts on the circular projected area within which the meniscus wets the tip ($A = \pi R^2$), as illustrated in inset of Fig. 1(b):

$$|F_c| = P_{Laplace}A = \gamma\left(\frac{1}{r_1}+\frac{1}{r_2}\right)\pi R^2 \tag{S1}$$

where γ is the liquid-vapor surface tension, r_1 is the positive meniscus neck radius and r_2 (= $d_c/2$, d_c is critical distance of nucleation) is the negative curvature of the meniscus in the normal direction. The parameters for the calculation of the capillary force are listed in Table SI. The second contribution to the adhesive force, the tension force (Eq. (S2)), acts along the circular contact line on the AFM tip and is described by:

$$|F_t| = 2\pi R\gamma\cos(\alpha)\sin(\beta) \tag{S2}$$

where α is the meniscus-tip contact angle, and β is the angle between the normal axis of the tip and the vector that connects the center of the (spherical) tip to the meniscus-tip contact line (see inset of Fig. 1(b)). The relation between R and r can be approximated by: $R = 2(R_{tip}r+R_{tip}h)^{0.5}-r$ where R_{tip} is the tip apex radius and h is the condensate thickness. [1] We estimated the apex radius of the AFM tip to be R_{tip} = 26 nm and 30 nm through the tip qualification method (see Supplemental material II). The meniscus-tip contact angle and the liquid-vapor surface tension are set to α = 40° [2] and γ = 72.8 mN/m [1] for water. The last contribution to the adhesive force is the van der Waals force. The van der Waals interactions between SiO_2-air-SiO_2 and SiO_2-water-SiO_2 can be described by Eq. (S3): [2]

$$|F_v| = (H_{air}(1\text{-}A_{Rel})+H_{water}A_{Rel})\frac{R_{tip}}{6d_a^{\,2}} \tag{S3}$$

where $H_{air} = 10.38\times10^{-20}$ J and $H_{water} = 1.9\times10^{-20}$ J are the Hamaker constants for the interfaces of SiO_2-air-SiO_2 and SiO_2-water-SiO_2, respectively, and d_a (0.2 nm) is the average distance between atoms. [2] The adhesive force that is then calculated using the model is 54±4 nN.

II. AFM TIP RADIUS DETERMINATION

To compare theoretical predictions of the adhesive force to AFM measurements, we estimated the AFM tip radii (R_{tip}) using a tip reconstruction method. [3] Within this method, a titanium sample with sharp edges (RS-12M, Bruker) is scanned by the AFM tip over an area of 5 μm × 2.5 μm using Bruker PeakForce QNM mode. The tip geometry can then be extracted from the measured topography through deconvolution (NanoScope Analysis, Bruker) (Fig. S1). By fitting a circle to the (2D) height profile of the reconstructed tip, we can estimate the tip radius.

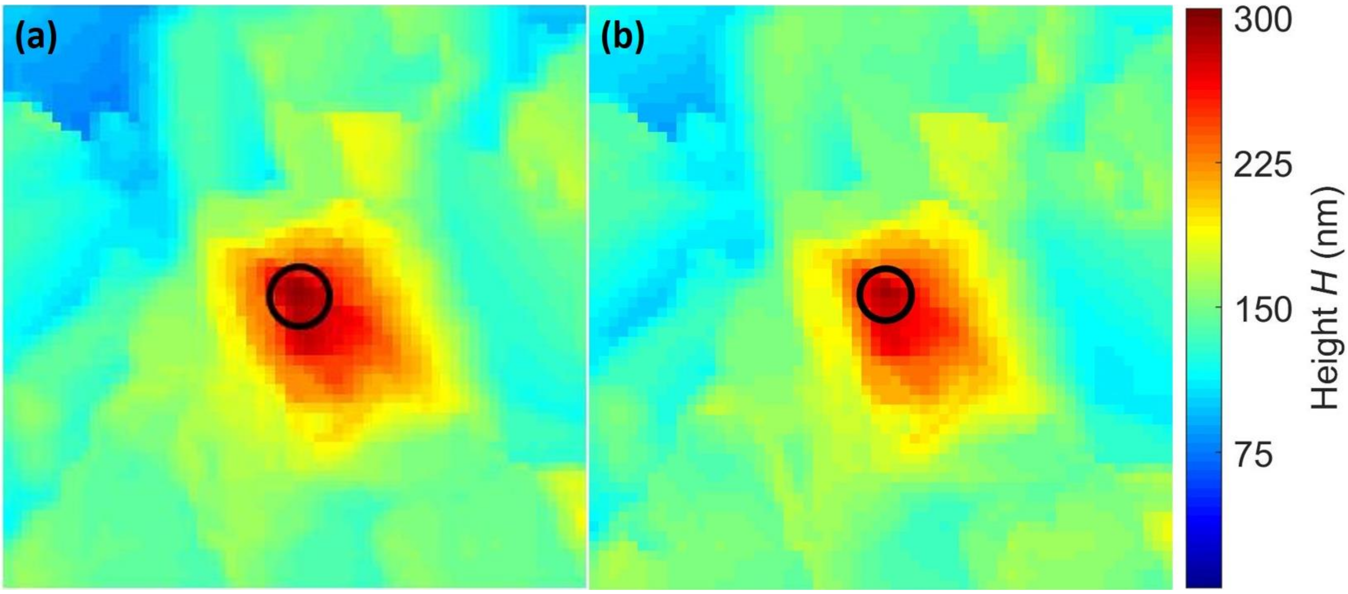

FIG. S1. AFM tip radius characterization. The reconstructed geometrical height map of the tip apex. The black circle is fitted to the spherical shape of the tip apex where the radius of the circle or the tip radius (R_{tip}) is given as 30 nm (a) and 26 nm (b). For the fitting process, we estimated the tip radius to be the smallest circumference of the tip apex determined.

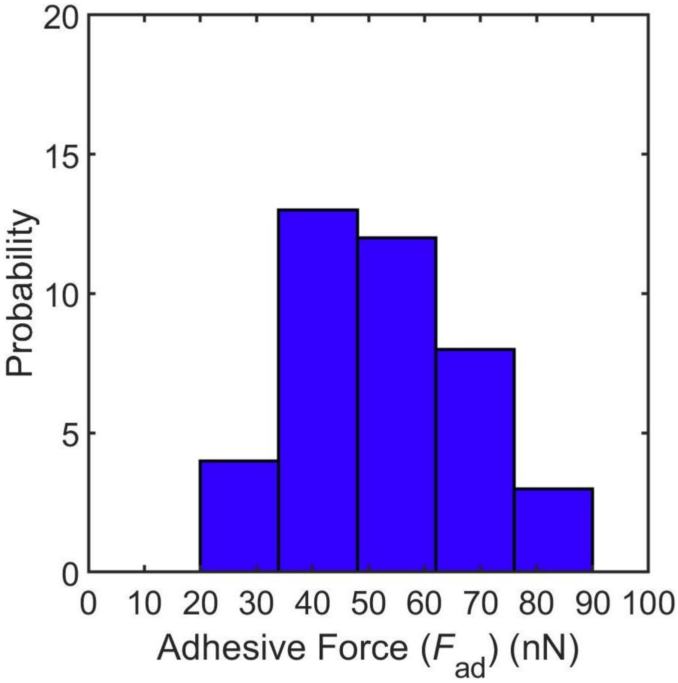

FIG. S2. Probability distribution of the adhesive force (F_{ad}) measured in water vapor environment (58±0.7% RH). The distribution of the adhesive force obtained through 40 AFM pull-off force measurements at different locations on the Si wafer (see Appendix A). The mean adhesive force is 52 nN with a standard deviation of 15 nN.

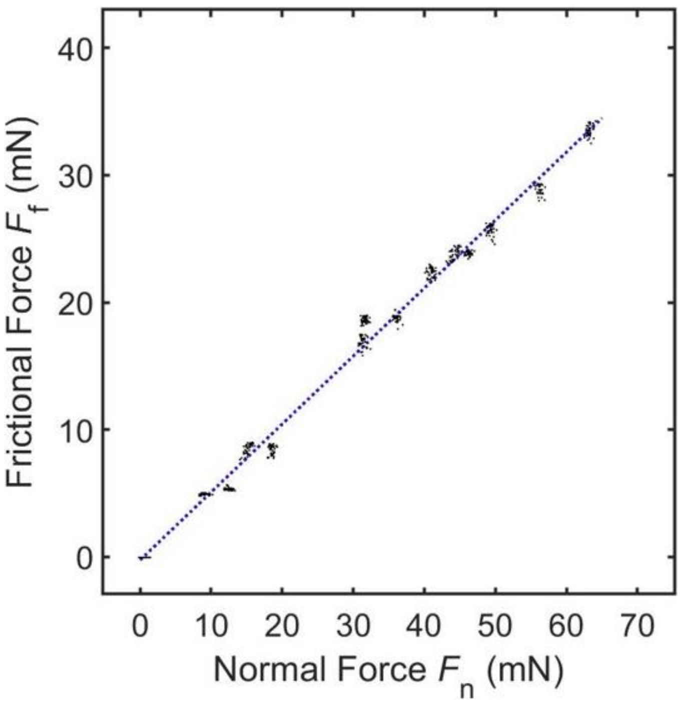

FIG. S3. Frictional force as a function of normal force for the contact between a Si ball and a Si wafer. The measured frictional force (black dots) linearly increases with the applied normal force at 1 μm/s sliding speed in ambient environment (36% RH; 20.3 °C). The linear fitting line is extended to the origin of the graph, indicating that the coefficient of friction (CoF) is well defined: CoF = F_f/F_n, with F_f the frictional force and F_n the normal force.

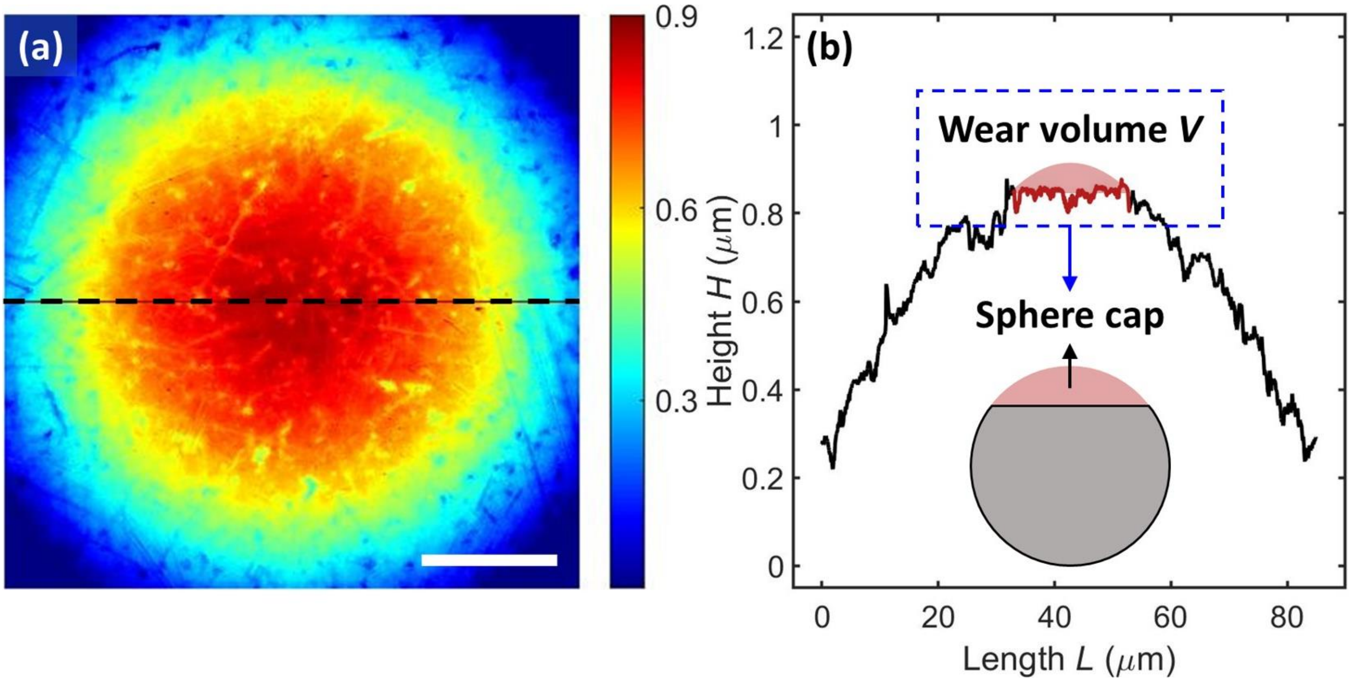

FIG. S4. Wear volume calculation. (a) The AFM surface topography of a Si ball after sliding. (b) The cross-sectional height profile through the wear scar (dashed line in (a)). The wear volume (V) is calculated by assuming that the lost material forms a spherical cap the base diameter of which is equal to the diameter of the wear scar. Scale bar, 20 μm.

TABLE SI. Condensate thickness (h) and critical distance (d_c) of water and isopropanol (IPA) at different relative humidity (RH) and partial pressure (P/P_{sat}).

Vapors	RH (%)	Thickness h (nm) [4]	Critical distance d_c (nm) [5]
Water	58	1.18	3.0
Water	50	1.11	2.5
Water	40	0.98	2.1
Vapors	P/P_{sat} (%)	Thickness h (nm) [6]	Critical distance d_c (nm) [7]
IPA	70	0.60	4.0*
IPA	45	0.51	2.8
IPA	30	0.46	2.3
IPA	10	0.29	1.6

*The critical distance at P/P_{sat} = 70% is extrapolated by the value of the linear correlation between critical distance and IPA partial pressure at 10–45%.

TABLE SII. Mechanical properties of the Si ball and the Si wafer.

Young's modulus (GPa)	Poisson's ratio	Hardness (GPa)
130	0.2	10

TABLE SIII. Total sliding distance and Wear volume of Si ball samples.

Samples	Total sliding distance D_{Tot} (μm)	Wear volume $\times10^9$ (nm^3)
1	190	4.75
2	180	4.09
3	180	2.77
4	270	5.76

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