Electronic Supplementary Material (ESI) for Faraday Discussions.

Supplementary Material for:

Modifying surface forces through control of surface potentials

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Surface force measurements

Normal and shear forces between gold and mica across no added salt water and electrolyte solutions were measured using the surface force balance (SFB), shown schematically in figure 1A (main paper) and described in detail elsewhere[1]. The surfaces, gold and mica, are glued on opposing fused silica lenses in a crossed-cylinder configuration, equivalent to the geometry of a sphere over a flat surface and before each experiment their separation is calibrated in water with no added salt. The lower lens, onto which mica is glued, is mounted on a horizontal leaf spring and the top lens, onto which gold is glued, is mounted on a sectored piezoelectric tube (PZT), which can move either vertically or laterally with respect to the lower surface. This enables a direct measurement of both normal and shear forces between the surfaces. The separation D between the surfaces (mica and gold) in the SFB is calculated using the multilayer matrix method[2, 3], as previously described in detail[4].

Normal force measurement. Normal forces between the surfaces are measured as follows. The top lens (gold in this study) is moved to a distance D between the surfaces by a 3-stage mechanism, using two dc motors and a sensitive sectored piezoelectric tube (PZT). The normal force F_n between the surfaces is monitored through the bending of the horizontal leaf spring

(spring constant K_n = 81.5±2.7 N/m or K_n = 128±15 N/m) using multiple beam optical interferometry (suitably modified for a 2-layer interferometer[4]). Optimally, the SFB is capable of measuring normal forces with a sensitivity of ~50 -100 nN and surface separation resolution of ±2-3Å. In principle, normal forces can be measured either via a quasi-static step-wise approach or via a dynamic approach as described in detail elsewhere[1, 5]. Briefly, in a step-wise approach the upper surface is moved by a dc motor and a PZT towards the lower surface in known distance steps (ΔD_{PZT}) while measuring the actual change in distance (ΔD) so the force can be calculated through $\Delta F_n(D) = K(\Delta D_{PZT} - \Delta D)$. In the dynamic approach the lower surface is continuously moved towards the upper surface at a known velocity until contact is reached. During its approach, the location of the fringe is monitored via a fast video recording camera (60 frames/second) and the normal force, which reflected through the bending of the leaf spring, is calculated from knowing the applied motion of the lower surface and taking into account the hydrodynamic force acting on the surfaces[6].

Friction force measurement. Application of shear motion is achieved by a piezoelectric tube (PZT) whose outside silvered surface is divided into four equal sectors A–D. By applying equal and opposite voltages to two of the opposing outside sectors (say B & D) one expands while the other contracts providing a lateral motion Δx_0 to the top surface as indicated in the left of figure 1C (main paper). Any shear (or friction) forces F_s acting between the surfaces are transmitted to the vertical leaf springs (K_s=300N/m) via the rigid stainless steel frame on which the top surface is mounted. The resultant bending Δx of the shear springs in response to the shear forces is monitored by an air gap capacitor probe which can detect distance changes in the gap as low as 3Å. This provides a sensitivity of ~0.1 μN in friction detection and a lateral motion range of ca. 1.5μm.

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