Supplementary Information

"Buffering Agents Modify the Hydration Landscape at Charged Interfaces"



Supplementary fig. S1: Low-magnification (100 nm) height (left) and phase (right) scans of mica in 10 mM Tris buffer highlighting horizontal discontinuities that were observed (arrows) between regions similar to that in fig. 2e and less well-resolved areas in the top and the bottom. These points were also visible in the phase. Scale bars are 30 nm, height colour scale is 150 pm, phase colour scale is 10°.



Supplementary fig. S2: high magnification (20 nm) height scan of mica in 10 mM SSC buffer. Scan illustrates heterogeneity of scanning conditions, with atomic-level resolution in the upper-right portion and much less structure observable in the lower half. Scale bar is 5 nm and height colour scale is 150 pm.



Supplementary fig. S3. Consecutive AM-AFM high-magnification height images of freshly-cleaved mica surface in 10 mM MES buffer. The initial image (left) shows horizontal discontinuities that persist for a few lines. These were interpreted to be a result of buffer interference with the tip-sample interaction, leading to loss of resolution. This was confirmed in the following scan, as the same area demonstrated similar lack of atomic-level resolution on the mica. Scale bars are 5 nm, height colour scale is 150 pm.



Supplementary fig. S4. Low-magnification AM-AFM image of freshly-cleaved mica surface in a 10 mM PBS buffer. The presence of stripes across the entire frame confirms the stability of imaging in this medium. The stripes have a periodicity of 2.67 nm, which, as in mentioned in the Results and Discussion section, is too large to be a direct representation of the mica lattice.



Supplementary fig. S5. AM-AFM image of bare silicon/silicon-dioxide surface in ultrapure water; height (left), phase (right). Clear features are visible that correlate to some extent with those in fig. 6a-d. However the similarities are related to the larger scale features (i.e. those associated with the bilayer following the silica surface); there are no high-frequency ripples as in fig. 6b or mesh-like networks visible as in the phases of fig. 6c,d. The roughness, R_q , was measured to be 0.137 nm; greater than that observed in fig. 6a, implying that the SLB produced 'smoothed over' surface features. Scale bars=30 nm, height colour scale = 500 pm, phase colour scale = 10° .