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## **Supplementary Material**

## The Impact of Water on the Lateral Nanostructure of a Deep Eutectic Solvent–Solid Interface

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## Mica Crystal Structure



**Figure S1.** Side view and basal plane reconstructions of the crystal structure of the mica (muscovite) surface. The basal plane is shown without potassium ions, so that the hexagonal symmetry of the substrate is clear for the reader.



**Figure S2.** Example of amplitude data from the force spectroscopy curve conducted in this work. This specific curve was obtained from the 100% DES - mica interfaces. The zero on the x-axis is arbitrarily set, as the absolute tip-sample separation cannot be determined during an AFM experiment. Overall, the data is less instructive than the phase data shown in the main text.



Figure S3. Corresponding amplitude data for the phase force spectroscopy for the 0% DES (pure water) system shown in Figure 2.



**Figure S4.** Coordination numbers determined from partial radial distribution functions ( $N_{coord}$ ) for hydrogen bond interactions between the atom pairs of water oxygen (Ow) and hydrogen (Hw); chloride ion (Cl); choline oxygen (Oc) and hydrogen (Hc); and glycerol oxygen (Og) and hydrogen (Hg).