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Counterion liquids between biological polyelectrolytes

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Electrostatic interactions between charged surfaces in water are dominated by counterion behavior. For example, repulsion between like-charged objects are due to the osmotic pressure of compressed counterions, and attraction between oppositely-charged objects are due to the entropy gain of counterion release. In systems with strong electrostatic interactions, mean field theories break down and counterion correlations and dynamics become important. To explain counterintuitive phenomena such as like-charge attraction between polyelectrolytes, exotic models such as counterion Wigner lattices were proposed, but no experimental comparisons at sufficient spatial resolution exist. Using inelastic x-ray scattering, the spatiotemporal correlations of counterions that mediate binding between charged polymers have been measured in aqueous solution down to molecular length-scales. We find that these ions are hierarchically organized into a dense, strongly correlated liquid that exhibits an acoustic-like phonon mode. The excitation spectra at high wave-vector Q reveal unexpected dynamics due to ions interacting with their 'cages' of nearest neighbors. The measured speed of sound and collective relaxation rates in this liquid agree well with simple model calculations.