Using Ionic Liquids to Control Interface Properties over Macroscopic Distances

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We have found that it is possible to induce long range order (*ca*. 100 μm) in room temperature ionic liquids (IL) by means of applying a potential to the surface on which the IL resides. We have used 1-butyl-3-methylimidazolium tetrafluoroborate (BMIM+BF4-), supported on a silica surface to demonstrate the effect. Long-range ordering, over five orders of magnitude greater than predicted by the Guoy-Chapman or similar models, can be seen in the rotational diffusion dynamics three structurally similar dyes, where one is anionic, one is cationic and a third is neutral. The rotational diffusion time constants of the charged dyes vary in a regular manner as a function of distance from the (charged) support surface. We model these data in the context of an excess charge density gradient being induced in the IL by the charge present on the support. The effect depends directly on the surface charge and affords a novel means of establishing the potential of zero charge (pzc) for the support material. Rotational diffusion measurements of these same chromophores in ethylene glycol are spatially invariant, demonstrating that this effect is associated with the ionic liquid. Capping the silica support with Me2SiCl2 prevents the establishment of a charge density gradient also, demonstrating the need for surface charge to induce long-range order in ILs. The IL exhibits a spatially-damped piezoelectric response mediated by IL fluidity and disorder, and we consider the potential of this effect for controlling interface properties.