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Near-surface dynamics of 1-ethyl-3-methylimidazolium acetate above and below the glass transition

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Ionic liquids (ILs) are a class of molten salts which are liquid at room temperature. Their properties are determined by strong electrostatic forces and generally include low volatility, negligible vapour pressure, and a low melting point. This makes them attractive candidates for improving battery and capacitor technology. The device effectiveness, however, is often determined by the ion arrangement and dynamics at the interface between the IL electrolyte and the electrode.

A much simpler interface is that of the free surface (vacuum interface). Studies of IL surface chemistry reveal a number of surprising phenomena, such as orientational ordering and regions of surface crystallization. As the ionic constituents may differ greatly in size and shape, many ILs do not crystallize (other than at the surface, perhaps) and will instead vitrify. The unusual behaviour of ILs may help to reveal novel aspects of glassy dynamics.

Having demonstrated that β-NMR was a good probe of IL solvation dynamics and dynamic heterogeneity,[1] we now turn to the question of how the surface modifies these properties, presenting the first depth-resolved β-NMR measurements in 1-ethyl-3-methylimidazolium acetate. The surface clearly has a large dynamical effect in the glassy phase: there is enhancement in the relaxation rate near the surface, resembling a more liquid-like state, yet no significant change in the dynamical heterogeneity. Additionally, the relaxation grows faster as the material is cooled through the glass transition temperature. These two latter aspects are very surprising, and are an extreme departure from the behaviour of other glasses.[2]

[1] Fujimoto, D. et al. Chem. Mater. 31, 9346–9353 (2019).
[2] McKenzie, I. et al. J. Chem. Phys. 156, 084903 (2022).

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