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Solid-Liquid Interfaces: Atomic-scale Structure and Dynamics

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Solid-liquid interfaces present an unparalleled opportunity to manipulate charge at the atomic scale and drive chemical transformations.^{1,2} It is critical to quantify the atomic-scale structure and dynamics of these interfaces to understand and exploit novel interfacial phenomena that emerge from the significant complexity of these interfaces. Recent developments in fields ranging from *ab initio* molecular dynamics (AIMD)³ accelerated with machine-learned potentials⁴ to spatially-resolved *in situ* interfacial characterization techniques⁵ bridge the scales accessible by computation and experiment, providing an increasingly detailed atomic picture of solid-liquid interfaces.⁶ This special topic in the Journal of Applied Physics, “Solid-Liquid Interfaces: Atomic-Scale Structure and Dynamics” brings together many of these recent developments.

The grain structure of crystals grown from liquids depends on the initial nucleation mechanisms, which therefore affect the achievable quality of materials ranging from structural alloys to semiconductor substrates. Computational advances now allow direct simulation of the solidification process in *ab initio* molecular dynamics simulations of solid-liquid interfaces to reveal ultrafast processes that determine the formation of initial nuclei. For example, AIMD simulations of the solidification of aluminum on intermetallic surfaces find that the solid-phase structure is driven by liquid-phase ordering during the growth, and that enhanced interfacial bonding and reduction of misfit strain drive heteroepitaxial growth of the solidifying metal.⁷ Direct simulation of free solidification can be challenging however, on account of the long time scales involved in nucleation. New simulation techniques, such as capillary wave analysis, have been shown to accurately reproduce the predictions of more expensive solidification simulations of metal alloys,⁸ making *ab initio* investigation of crystal growth more accessible. Combinations of such simulations with experimental growth provide new insights into the development of high-quality sapphire crystals for optoelectronic applications, with simulations elucidating the difficulty of *c*-axis growth and highlighting the possibility of solid-solid transitions *after* solidification.⁹ While *ab initio* simulations based on density functional theory (DFT) provide a useful starting point to investigate solid-liquid interfaces, self-interaction errors in DFT limit the accuracy of energy level alignment across interfaces. New techniques to combine many-body *GW* perturbation theory with solvation models show that electric screening by the liquid environment strongly impacts the energy levels of molecules in solution and must be accounted for to correctly describe interfacial states in solid-liquid interfaces.¹⁰

Interactions between biomolecules and cells in solution and solid surfaces are useful for probing their structure, separating molecules and controlling their synthesis. These interactions typically arise from a combination of electrostatic and specific chemical forces, and are therefore expected to change with the solution's ionic strength. Using total internal reflection aqueous fluorescence, measurements of the equilibrium distance between algae and silica surfaces align with expectations based on double-layer theories, the cell potential and surface charge at low ionic concentrations.¹¹ However, substantial increases in the equilibrium distance at high ionic concentrations reveal possible effects of changes in hydrogen-bonding interactions in the solvent or of the surface charges of the cells due to thinning or disruption of cell walls. The interaction of electric fields at the interface with solutes also determines the prospect for trapping and sorting solutes at solid surfaces. While electrophoresis of charged particles is widely used, dielectrophoresis of neutral polarizable particles has been expected to require impractically large electric fields. Remarkably, despite this, neutral proteins can be trapped with experimentally-accessible fields, which models explain could be due to orientation of permanent dipoles in proteins with the applied field, an effect that could be enhanced by applying fields on micrometer-sized pores.¹² Interfacial effects more generally provide opportunities to substantially expand the design space for applications, compared to bulk materials alone. For example, microwave absorber materials for electromagnetic shielding applications can be engineered to exploit the interfaces between magnetic and non-magnetic materials in ferrite composites, leveraging recent methods to control the morphology of ferrite particles and their interfaces to tune absorption.¹³

Many fundamental questions remain outstanding about the range and importance of interface interactions at solid-liquid interfaces. The articles in this Special Topic on "Solid-Liquid Interfaces: Atomic-Scale Structure and Dynamics" highlight recent developments of new computational and experimental techniques that provide further atomistic detail and quantitative accuracy for the prediction of the dynamics at solid-liquid interfaces, such as for crystal growth during solidification. The control of electric fields at these interfaces allows trapping, probing and manipulating biomolecules and cells, while appropriately engineered interfaces in composite materials can strongly modify electromagnetic field profiles for microwave absorption. Further development of computational and experimental techniques to bridge between the atomic and macroscopic scales will be vital to complete our understanding of solid-liquid interfaces and realize their full potential for technological applications.

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