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Nanoparticles at fluid interfaces: how capping ligands control adsorption, stability and dynamics

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The spontaneous assembly of nanoparticles at fluid-fluid interfaces is exploited in microfluidic encapsulation, fabrication of nanomaterials, oil recovery, and catalysis. Control over the microstructure formed by interfacial nanoparticles is an important goal in these contexts: the ability to *reversibly* tune the packing fraction enables for nanomaterials with tunable properties, while control over nanoparticle removal and recycling is desirable for green processes. I will discuss how capping ligands can promote interfacial self-assembly by tuning the interfacial energies of the nanoparticles with the fluids. Ligand-mediated particle interactions at the interface then affect the formation of equilibrium and non-equilibrium two-dimensional phases. Important differences with colloidal interactions in a bulk suspension arise due to the discontinuity in solvent properties at the interface, which cause the ligand brushes to rearrange in asymmetric configurations. I will present experimental results for gold nanoparticles capped with short amphiphilic ligands, which spontaneously adsorb at an oil-water interface. Using pendant drop tensiometry, we measured the surface pressure of the nanoparticle monolayer during adsorption and subsequent compression. In contrast to the commonly observed buckling of solid-like films of interfacial particles, upon compression these nanoparticles are mechanically forced out of the interface and into suspension. Area density measurements by a newly developed optical method reveal that ligand-mediated short-range interparticle repulsion enables desorption upon compression. Brownian dynamics simulations corroborate this picture. Therefore, ligand-mediated interactions also determine the fate of nanoparticle monolayers upon out-of-plane deformation.