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Soft particles at fluid interfaces: wetting, structure, and rheology.¹

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Most of our current knowledge concerning the behavior of colloidal particles at fluid interfaces is limited to model spherical, hard and uniform objects. Introducing additional complexity, in terms of shape, composition or surface chemistry or by introducing particle softness, opens up a vast range of possibilities to address new fundamental and applied questions in soft matter systems at fluid interfaces. In this talk I will focus on the role of particle softness, taking the case of core-shell microgels as a paradigmatic example. Microgels are highly swollen and cross-linked hydrogel particles that, in parallel with their practical applications, e.g. for emulsion stabilization and surface patterning, are increasingly used as model systems to capture fundamental properties of bulk materials. Most microgel particles develop a core-shell morphology during synthesis, with a more cross-linked core surrounded by a corona of loosely linked and dangling polymer chains. I will first discuss the difference between the wetting of a hard spherical colloid and a core-shell microgel at an oil-water interface, pinpointing the interplay between adsorption at the interface and particle deformation. I will then move on to discuss the interplay between particle morphology and the microstructure and rheological properties of the interface. In particular, I will demonstrate that synchronizing the compression of a core-shell microgel-laden fluid interface with the deposition of the interfacial monolayer makes it possible to transfer the 2D phase diagram of the particles onto a solid substrate, where different positions correspond to different values of the surface pressure and the specific area. Using atomic force microscopy, we analyzed the microstructure of the monolayer and discovered a phase transition between two crystalline phases with the same hexagonal symmetry, but with two different lattice constants. The two phases correspond to shell-shell or core-core inter-particle contacts, respectively, where with increasing surface pressure the former mechanically fail enabling the particle cores to come into contact. In the phase-transition region, clusters of particles in core-core contacts nucleate, melting the surrounding shell-shell crystal, until the whole monolayer moves into the second phase. We furthermore extended our analysis to measure the interfacial rheology of the monolayers as a function of the surface pressure using an interfacial microdisk rheometer; the interfaces always show a strong elastic response, with a dip in the elastic modulus in correspondence of the melting of the shell-shell phase, followed by a steep increase upon formation of a percolating network of the core-core contacts. The presented results highlight the complex interplay between the wetting and deformation of individual soft particles at fluid interfaces and the overall interface microstructure and mechanics. They show strong connections to fundamental studies on phase transitions in two-dimensional systems and pave the way for novel nanoscale surface patterning routes.

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