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“Strong or weak, quick or slow: interpreting nonlinear viscoelasticity from large amplitude oscillatory flow”

Summary

The way in which materials deform when stressed is of both scientific and technological importance. This is reflected by viscoelasticity, a physical property relating the stress to deformation history. From polymers to proteins to paints, nanostructured media exhibit a host of viscoelastic characteristics that can vary with both the strength – strong or weak – and speed – quick or slow – of the deformation. The rheological response to large amplitude, oscillatory deformation is thought to characterize both facets – strength and speed. A microstructural understanding of this response is an on-going challenge. I have conducted a detailed study of the response to large amplitude oscillatory flows (LAOF) from a micromechanical perspective via the micro-rheology of colloidal dispersions. Considerable insight into the physical processes underlying large amplitude flow behavior may be gained through examination of the motion of a colloidal probe through a dispersion of other colloids. In particular, the relative importance of hydrodynamic and thermal forces determines the amount of energy stored and lost during the process. Interestingly, while hydrodynamic interactions are largely responsible for the behavior of strongly driven materials, I show that hydrodynamics do not contribute to net energy storage. A comparison of the response for driven suspensions with and without hydrodynamic interactions reveals that a detailed understanding of the material microstructure is necessary for meaningful interpretation of large amplitude oscillatory flows. This very simple model provides such details and recovers the rich behavior observed experimentally in a broad range of viscoelastic materials – ranging from colloidal dispersions to polymer melts and even slug mucus.