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Probing the nonlinear response of soft materials by active microrheology

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In passive microrheology, the linear viscoelastic properties of complex fluids are inferred from the Brownian motion of colloidal tracer particles. Active (but gentle) forcing may also be used to obtain such linear-response information. More significant forcing may drive the material significantly out of equilibrium, thus potentially providing a window into the nonlinear response properties of the material. In leaving the linear-response regime, however, the theoretical underpinning for passive microrheology is lost, and a variety of issues arise. Most generally, what exactly can be measured, and how can such measurements be interpreted? Using a model system (a large colloidal probe pulled through a dilute suspension of small bath particles), we examine the different sources of stress upon the probe particle (e.g. direct probe-bath collisions vs. microstructural deformations within the bulk suspension) and discuss their analog in the corresponding macro- rheological measurement (or lack thereof). Several crucial issues emerge for the interpretation of nonlinear microrheology: 1) how to interpret the inhomogeneous and non-viscometric nature of the deformation field around the probe, 2) the distinction between of direct and bulk stresses and their deconvolution, and 3) the (Lagrangian) time-dependent nature of the stress histories experienced by material elements as they advect past the probe. Having identified these issues, we discuss several adaptations of the basic technique/interpretation, both to more faithfully recover bulk rheology as well as to measure properties inaccessible to macro- rheology. While we specifically discuss a model colloidal suspension, we ultimately envision a technique capable of measuring the nonlinear rheology of general materials.