# NONLINEAR VISCOELASTICITY AND YIELDING OF SOFT COLLOIDAL GLASSES PROBED BY LARGE AMPLITUDE OSCILLATORY SHEAR

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A colloidal glass at rest behaves like a soft solid, with the elastic modulus G' larger than the viscous modulus G" at all accessible frequencies. However, the same material will flow when subjected to a strain (or stress) that exceeds yielding. Beyond this shear-induced solid-to-liquid transition, the soft glass exhibits complex non-linear viscoelasticity that is still poorly understood.

Here we study the yielding and non-linear viscoelasticity of model concentrated colloidal suspensions and glasses, formed by diblock copolymer micelles [1]. Large amplitude oscillatory shear (LAOS) experiments are performed, where the amplitude of oscillation is progressively increased to beyond the linear response regime. The results are presented as Lissajous plots (stress vs strain) in a Pipkin diagram of amplitude of oscillation against angular frequency. As the amplitude of oscillation is increased, the stress response becomes progressively less sinusoidal. The complex stress waveforms are decomposed by Fourier analysis into a fundamental frequency and higher harmonics contributions, following Wilhelm et al [2], or equivalently into elastic and viscous parts and orthogonal sets of Chebyshev polynomials [3, 4]. Based on the third harmonic, such approaches enable us to map out the rich phenomenology of non-linear behaviour inside a period of oscillation. Thus, as the oscillation amplitude is increased, the colloidal glass progressively makes the transition from a linear viscoelastic response to a strain hardening intracycle response (intermediate amplitudes), to a shear thinning intracycle response (large amplitudes). Interestingly, the same qualitative non-linear behaviour is deduced from LAOS experiments on lower effective volume fraction liquid (i.e. non-glassy) micellar suspensions, but with the onset of non-linear response happening at higher amplitudes.

The LAOS results on soft micelles are compared to previous results on hard spheres and deformable soft particles with the purpose of contributing to the general understanding of the yielding process in soft matter systems. Moreover, some limitations of the current analysis methods of LAOS experiments are presented, based on current data. Finally, we propose alternative methods of looking at LAOS experimental results, which provide additional physical insight.

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