Project C2: Influence of shear prehistory and particle attractions on the relaxation dynamics from non-equilibrium states to metastable colloidal glassy states

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Current state of research. Many products of the daily life (gels, cosmetics, paints, etc.) are metastable colloidal systems whose rheological behaviour and performance can strongly depend on processing conditions and history. Processing inevitably involves some kind of shear. In recent years, much progress has been made in correlating microstructure, microscopic dynamics and macroscopic rheological behaviour of glass-forming colloidal suspensions. This has been due to a combination of well-defined model systems (e.g. the PMMA hard sphere (HS) colloidal system), new instrumental techniques (e.g. confocal microscopy or light echo dynamic light scattering) and the advent of mesoscopic and *ab initio* theories for rheological properties. Especially the mode coupling theory has led to a quantitative description of the linear and nonlinear rheology as well as of transient shear effects close to the HS glass transition. This has also paved the way for a qualitative understanding of the effect of particle softness on the rheological behaviour. However, systematic work on the rheology of attractive colloidal glasses is scarce, and the understanding of effects of the (shear) prehistory is significantly lacking behind. This present project aims at providing a data base which could serve as a starting point for a development of a theory on the influence of processing conditions and particle attraction on the properties of colloidal glasses.

Contributions of the principal investigators. The Freiburg group has expertise in the synthesis and characterization (light-scattering) of colloidal suspensions, in particular for systems where a competition between repulsive and attractive glass formation exists. The phase diagram of such systems has been mapped out, the generalized Stokes-Einstein relation has been tested, and dynamic light-scattering suggests the formation of (transient) cluster states near (but below) the attractive glass line. The Strasbourg group has expertise in the characterization of the viscoelastic behaviour of colloidal suspensions. Relevant to this project is for instance the finding that oscillatory pre-shear histories of small amplitudes can lead to partial rejuvenation of glassy colloidal suspensions (of laponite) and also that the formation of contacts in attractive colloidal suspension can increase the yield stress.

Research project and collaborations. We plan to study the influence of the preshear history on the dynamics of colloidal glasses as probed by linear and nonlinear rheology (Strasbourg) and by dynamic light scattering (DLS, Freiburg). Samples will be conditioned by well-defined shear protocols and the response of the system to identical pretreament as seen by linear rheology and DLS will be compared. For this purpose storage and loss moduli, $G'(\omega)$ and $G''(\omega)$, will be derived from DLS data by the generalized Stokes-Einstein relation. The comparison of $G'(\omega)$ and $G''(\omega)$ from DLS and rheology and their evolution with time as well as the time evolution of the yield stress will give insight into the influence of the preshear history on the behaviour of attractive and repulsive colloidal glasses.
Work plan. This PhD project involves the following tasks. *1st year:* first 6 months in Freiburg (synthesis and characterization colloidal dispersions; determination of glass transition line; decision on state points for rheological experiments), next 6 months in Strasbourg with short visits to Freiburg (nonlinear and linear rheology; variation of the preshear protocol). *2nd year in Freiburg*: Rebuilding of the DLS setup (introduction of shear cell for preshear treatment); control experiments on quiescent samples. First exploratory measurements on attractive colloid glasses. Short visits to Strasbourg. *3rd year:* Freiburg with several short stays in Strasbourg; systematic study of the preshear dependence of the relaxations dynamics with DLS; comparison of the results from optical tracer microrheology and bulk rheology.