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# Colloidal dispersions in external fields: recent developments

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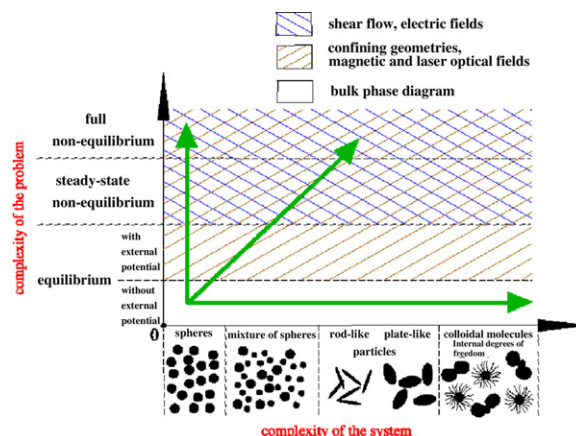
## Abstract

The past five years have witnessed considerable progress in the field of colloid physics, in particular when colloidal dispersions are exposed to external fields such as shear flow, electric, magnetic and laser-optical fields as well as confinement. This progress is summarized and brought into relation to the subsequent articles of this special issue.

## 1. Introduction

Colloidal dispersions are solutions of mesoscopic solid particles with typical sizes ranging from 1 nm to 1  $\mu\text{m}$  with a stable (i.e., non-fluctuating) core embedded in a fluid solvent. Monodisperse solutions of strongly interacting particles are excellent model systems that play a pivotal role for many-body statistical mechanics. In the past two decades this has been exploited to understand the principles of equilibrium many-body transitions like fluid–fluid phase separation and freezing. More recently, colloidal dispersion have also played a key role to unravel the underlying physics of *nonequilibrium* situations such as glass and gel formation, homogeneous and heterogeneous crystal nucleation and genuine nonequilibrium phase transitions like shear-banding and lane formation. The big advantage of colloids lies in the fact that they can be studied in real-space, that their interactions can be tuned, and that they can be brought into excellent equilibrium via the surrounding molecular solvent. This sets them apart from other driven systems like granular matter [1] or complex plasmas [2] which are typically kept in a nonequilibrium steady state [3].

An external field provides a natural way to control a suspension [4], this control is either to steer the model interaction parameters into equilibrium or to bring the suspension into nonequilibrium in a controlled way [5]. Thus the response of colloidal suspensions to external fields brings about new insights, in particular into nonequilibrium phenomena. Different kinds of problems and systems are schematically shown in a road map of complexity in figure 1. First, different particle shapes can by now be prepared including spheres, mixtures, rods, platelets and even colloidal molecules with dumbbells and other non-convex particle shapes [6]. This increasing complexity is indicated on the  $x$ -axis of figure 1. The  $y$ -axis, on the other hand,



**Figure 1.** Road map of complexity for colloidal dispersions in external fields: the  $x$ -axis shows the complexity of the systems as embodied in the statistical degrees of freedom necessary to describe a configuration, the  $y$ -axis sketches the complexity of the problem ranging from equilibrium bulk problems to full time-dependent nonequilibrium. The regions associated with different kinds of external fields are indicated. The arrows indicate typical progress of recent research.

(This figure is in colour only in the electronic version)

describes the different situations ranging from bulk to full nonequilibrium. As already mentioned, external fields can be employed to control the interactions in equilibrium and to bring the suspensions into nonequilibrium in a controlled way. The arrows indicate progress obtained within the last decades.

The aim of this introductory article is twofold: first progress in the field of colloidal dispersions achieved within the last five years is summarized and highlighted. Second, it brings into interrelation the subsequent papers by classifying the problems according to the type of the external field applied.

## 2. Colloidal dispersions in the bulk

As already mentioned—thanks to the variety in colloidal shape—the physics of the *gel transition* has been explored using colloids. Patchy colloids with well-defined sticky spots [6] of low functionality form low volume fraction gels [8] with novel properties. The liquid part of the bulk phase diagram is shifted to a very small volume fraction (so-called empty liquids) [7] which opens a wide stability range for the fluid such that an underlying gel transition is not masked by phase separation. Colloidal particles with well-defined crosslinkers have been prepared and an entropically driven gelation transition was discovered [9] as opposed to the traditional picture where gelation arises from short-range attraction.

The mode coupling theory of the bulk *glass transition* has been recently extended to predict dynamical heterogeneities [10]. Underlying and hidden (dynamical) correlation length scales accompanying the glass transition are debated [11]. From simulation and real-space experiments an insight was gained into the interplay between crystallization and glass formation [12], and the occurrence of crystalline clusters deep inside the glass has been reported [13].

*Bulk crystals* are still a fascinating research topic: recent elasticity measurements have revealed non-central forces in an fcc lattice of a charged suspension [14]. Colloidal crystals are excellent model systems to study phonon spectra. Sound propagation in binary colloidal mixtures can be tailored by the individual species and their composition [15]. The displacement fields of defects in two-dimensional solids have been explored by computer simulation and elasticity theory [16]; again this can be tested directly in two-dimensional colloidal crystals. The competition between glass and crystal formation can be steered by sedimentation [12]. Crystal nucleation from the melt, both homogeneous and heterogeneous, as for example discussed in [17], is another very active research field where colloids play a prominent role as model systems.

A higher level of complexity arises when the colloidal particles are made ‘active’, i.e. if they are *self-propelled* by an internal motor that dissipates energy in the quiescent solvent at low Reynolds number. It has been shown [18] that magnetic beads attached to colloidal particles can be steered by an external field in order to let the colloidal particles propagate. Also a local chemical reaction at the particle surface can be exploited as a propagation mechanism [19]. Such self-propelled particles are excellent model micro-swimmers which are numerous realized in nature by sperm and flagella driven microorganisms. Due to their internal degrees of freedom, active particles are in nonequilibrium. Even a single self-propelled particle exhibits new behaviour in the bulk [20] and in confinement [21]. Many particles show fascinating collective effects in nonequilibrium-like swarming with giant density fluctuations [22], swirling [23] and transient clustering in confinement [24]. It is expected that a wealth of new nonequilibrium effects will be explored in the future if these active suspensions are exposed to further external fields.

Colloids with *fluctuating shapes* do share the behaviour known from flexible polymers. The details of the similarities

and differences are still being explored [25]. Finally there are many cross-correlations from the interdisciplinary colloidal research domains to other scientific areas. For instance, the basic theory of colloidal interactions can even be applied to understand the sticking of geckos [26, 27].

## 3. Colloidal dispersions in shear fields

The *crystallization* behaviour under shear has been analysed by real-space experiments and been shown to be qualitatively different from the bulk. Recent activities have been focused on nucleation and growth of charged suspensions [28]. It was shown that the crystal growth rate and the induction time for a fixed concentration exhibits a maximum as a function of the shear rate. Three-dimensional imaging in shear cells is possible with confocal microscopy to reveal the underlying particle trajectories, and different kinds of shear cells have been developed for such *in situ* real-space measurements [29].

Another breakthrough was achieved in understanding the formation and ageing of colloidal *glasses* and *gels* under shear, see e.g. [30]. The yielding behaviour in repulsive and attractive glasses, as realized by hard-sphere suspensions and colloid–polymer mixtures, exhibits a single and two-step scenario, respectively [31]. Colloid–polymer mixtures may also serve as models for gels. Subjected to oscillatory shear, a full ‘shear state diagram’ was experimentally obtained [32]. On the theoretical side, mode coupling theory was generalized to steady-state shear [33] and to time-dependent simple shear [34] providing a suitable framework to classify, discuss and analyse experimental and simulation data (see e.g. [35]) for glasses and gels under shear. If shear is suddenly turned on, the start up of the flow has been analysed by using mode coupling theory, computer simulations and experiments [36]. In the transient colloidal dynamics a superdiffusive motion was detected.

*Colloid–polymer mixtures* are also ideal model systems for a fluid–fluid interface separating the colloid-rich fluid from the colloid-poor fluid. Thermally activated capillary waves have been made visible in these systems [37]. Upon shearing the interface, it was found that capillary excitations are strongly suppressed [38]. This phenomenon can be understood by introducing an effective interfacial tension that increases with the shear rate. Phase separation under shear apparently stops once a maximal length scale is achieved, in striking contrast to the bulk case [39].

*Rod-like particles* are much more affected by shear flow due to their orientational degree of freedom that aligns in shear flow and results in a so-called paranematic phase even at small concentrations. Vorticity banding in rod-like virus suspensions under steady shear flow was observed [40] within a part of the paranematic–nematic biphasic region. The underlying mechanism was shown to be related to the normal stresses generated by inhomogeneities formed due to the underlying paranematic–nematic phase transition. Furthermore, the nematic–isotropic spinodal decomposition kinetics of rod-like viruses was explored and experimental evidence was provided that spinodal decomposition is driven by orientational diffusion [41]. The nematic phase of rods in steady shear was simulated exhibiting a collective rotational motion in the

nematic phase [42]. A ‘living’ system like worm-like micelles under shear can merge into each other or get cut into pieces under shear leading to shear-thinning. It was shown, however, that the slowing-down of the orientational diffusion close to the isotropic–nematic spinodal is the main reason for shear-thinning [43].

In *computer simulations* of shear flow, the proper inclusion of hydrodynamic interactions between the particles is important. Recent progress was obtained by using the stochastic-rotation-dynamics scheme. The hydrodynamic modes of star polymers in linear shear flow was simulated, and anisotropies [44] and hydrodynamic screening [45] were observed. The shear-induced elongation of the star polymers was recently confirmed experimentally using *in situ* rheology and small angle neutron scattering [46]. Another alternative to simulate shear flow is to consider in strongly asymmetric binary mixtures the smaller species mimicking the solvent; then shear-thinning and shear-thickening can be studied by molecular dynamics simulations [47] as well as migration of colloids and polymers in shear flow [48].

#### 4. Colloidal dispersions in electric fields

If a charged colloid is placed into an electric field, the *electrophoretic mobility* is one of the key quantities governing the colloidal response. It has already been measured under controlled conditions and various circumstances in terms of salt concentration and particle density [49, 50]. In comparison with extensive computer simulations, which take Coulomb effects [51] and hydrodynamic interactions into account, good agreement was found in the low-salt regime using an effective particle charge and a reduced screening parameter. Two different volume fraction-dependent regimes for the electrophoretic mobility were identified [52] by scaling analysis [53]. Last but not least, a theoretical framework was proposed based on the mode coupling scheme which includes electrokinetic and hydrodynamic effects and was applied to long-time diffusion of charged suspensions [54, 55].

Another important development lies in the preparation of *oppositely charged colloids* [56]. In the absence of an electric field, the mixtures form binary crystals with a wealth of different structures [57]. When exposed to an electric field, these suspensions are excellent model systems for oppositely driven mixtures which were predicted to exhibit a nonequilibrium phase transition towards lane formation [58]. Each lane consists of particles driven in the same way. By Brownian dynamics computer simulations the full nonequilibrium phase diagram was mapped out and crystallization of lanes was found [59]. Hydrodynamic interactions are screened in this case and do not significantly alter the phase behaviour as revealed by computer simulations [60] based on the Ajdari–Long screened mobility tensor [61]. A full quantitative comparison between experiment [56] and simulation [60] has still to be done.

A difference in the dielectric constant of the colloidal material and the solvent makes colloids susceptible to electric field gradients. The migration of colloids in an ac electric field exhibits a *Rayleigh–Taylor-like instability* [62]. It is

intriguing to watch the onset of this instability on a ‘molecular’ (i.e. particle-resolved) scale there.

#### 5. Colloidal dispersions in laser-optical and magnetic fields

Laser-optical fields frequently serve as *tweezers* to fix and guide colloidal particles in the solution combined with a confocal microscope to detect the particle in real-space [63]. Capped silica colloids, in which half of the surface is covered with metal layers, were investigated. Due to their asymmetry, the capped colloids rotate when exposed to intense laser fields. Hence they act as colloidal light-mills with many fascinating applications [64]. By combining different laser-optical fields an oscillating trap can be realized. The breathing mode of colloidal particles contained in an oscillating trap was explored by dynamical density functional theory [65] including hydrodynamic interactions. The theory was tested against Brownian dynamics and good agreement was found between theory and simulation.

*Periodically-modulated static light fields* are readily realized by crossing laser beams. When colloids are placed into these external potentials at high density the new crystalline lattice is expected to be stable [66]. An experimental realization is proposed in this special issue [67].

An external magnetic field can be used to steer the interactions between the colloidal particles. A relevant set-up that has already been studied for decades is that of *superparamagnetic particles* at an air–water interface. An applied magnetic field induces dipole moments in the particles such that the effective interaction is well described by a dipole–dipole interaction. These suspensions serve as excellent models for strictly two-dimensional systems. The freezing scenario in two spatial dimensions is expected to differ qualitatively from that in three dimensions. In fact, a Kosterlitz–Thouless–Nelson–Halperin–Young (KTNHY) scenario with an intervening hexatic phase has been confirmed in these suspensions. In recent progress Frank’s constant was measured in the hexatic phase and good agreement with the KTNHY theory was found [68]. The gross features of freezing can be calculated within a microscopic classical density functional theory [69]. The superparamagnetic particles were further subjected to microchannels and their transport and layering properties were studied [70, 19].

Another study concerns *binary suspensions* of low and high effective magnetic susceptibility which are described as dipolar mixtures. In the fluid phase, a partial clustering of the small particles was found [71], as revealed in a pre-peak of the small–small partial structure factor. This effect is described and explained by theory and computer simulation [72]. At higher magnetic fields the particles undergo a glass transition. The dynamical signature is in line with one-component mode coupling theory in two dimensions [73]. A detailed study of the structure of the two-dimensional glass formed in the binary systems reveals [13] finite crystallites which possess the structure of thermodynamically stable crystals in equilibrium [74, 75]. When two of these crystallites fuse they

merge into one due to a ripening process [76]. This provides a new insight into the interplay between glass formation and crystallization. A mutual attraction between the particles can be turned on by tilting the field which yields anisotropic correlations [77] and much more complicated phase behaviour. Another way of increasing the complexity in the mixture is to expose it to an external periodic potential [78] corresponding to a modulated light field. Then laser-induced demixing occurs [79].

In three dimensions, a magnetic field can likewise be used to induce *orientational order*. This was exemplified in the isotropic phase of colloidal gibbsite in [80]. Board-like particles such as goethite show fascinating liquid-crystalline behaviour which can be steered via a magnetic field, in particular when their magnetic properties are modified due to an addition of Chromium [81]. Furthermore the Faraday effect was used to characterize magnetic colloids according to their magneto-optical signals [82].

## 6. Colloidal dispersions in confining geometry

In appropriate thermodynamic conditions, a colloid–polymer mixture exhibits a fluid–fluid phase separation with a coexisting colloid-rich and colloid-poor phase. How this fluid–fluid transition is affected by a *slit-pore confinement* was investigated by density functional theory [83] and by computer simulation [84]. Furthermore the critical behaviour of a colloid–polymer mixture in a slit-like confinement was obtained [85]. The exponents were found to be close to the two-dimensional Ising model as opposed to the bulk behaviour, which falls into the three-dimensional Ising class [86]. Finally, between asymmetric walls, an interface localization transition was predicted [87].

A longstanding conjecture of de Gennes [88], namely that the critical behaviour of liquids in porous media falls into the *universality class of the random-field Ising model*, was confirmed by computer simulation for a colloid–polymer mixture [89]. The corresponding susceptibilities have also been obtained and analysed close to criticality [90]. An extensive systematic study of the fluid–fluid binodals of colloid–polymer mixtures in random matrices has been performed [91]. In [91], the performance of replica liquid integral theories and replica density functional theories [92] was tested against simulation data for various matrices. The replica density functional method was also applied to soft-core particles [93].

Hard colloidal platelets were studied using density functional theory. *Capillary condensation* of platelets occurs when they are confined between parallel hard walls [94]. The isotropic–nematic interface tension of colloidal platelets is extremely small and the nematic phase wets the wall completely [95].

Experiments on colloidal *sedimentation* when a slit-cell is turned quickly upside-down were theoretically tackled by dynamical density functional theory and quantitative agreement was found at low Peclet numbers in the time-dependent laterally averaged density profiles [96]. Still the dynamical density functional theory lacks the prediction of

lateral inhomogeneities during sedimentation and neglects hydrodynamic interactions.

Elasticity is important when confining *rod-like particles* to cavities. In fact, for increasing density, it was shown [97] that first a uniaxial surface film forms and then a biaxial surface film, which eventually fills the entire cavity. Combining hard rods with polymers leads to depletion-induced network-formation [98] and isotropic–isotropic phase separation, which was studied in confinement [99]. In an orientationally ordered layer close to the walls the critical point of demixing is shifted to higher rod concentrations [100]. The response of nematic layers was studied with respect to another particle dragged through the suspension at constant speed and a rhythmic clustering in front of the dragged particle was observed [101].

The crystallization behaviour and dynamics of charged colloidal particles in a *charged wedge* of very low opening angles was examined by experiments, theory and simulations. A wedge geometry has the advantage that in one single set-up many different plate separations are realized simultaneously such that one can scan through the plate distance by moving along the wedge plate [102]. Even for a few layers exotic crystalline structures were found [103–106]. Furthermore the motion of particles towards the wedge cusp was explained by an attractive trapping force close to the wedge [107] without invoking a like-charge attraction picture. When sheared, these crystals exhibit an interesting relaxation dynamics back to equilibrium and can be aligned by shear [108].

Colloidal particles can be trapped at *fluid interfaces* and their effective interactions have been debated recently [109]. In particular it is important to understand the electrostatic, magnetic, or elastic building blocks in the interactions, both in terms of theory [110] and measurements [111]. The importance of line tensions and the effects of capillary waves are discussed in this issue [112]. Tuning these interactions leads to new crystalline superstructures which can be exploited as nano-sieves and filtering devices.

## 7. Conclusions

In conclusion, soft matter (like colloidal dispersion) is very susceptible to an external field. This can be exploited to steer the interactions between the colloidal particles but also to bring the suspension into nonequilibrium in a controlled way. Future activities will concentrate on full nonequilibrium and typical problems are as follows: ageing and rejuvenations in colloidal glasses and gels, a particle-resolved understanding of hydrodynamic instabilities, dynamics of cluster formation, and crystal nucleation out of dynamically arrested host phases (like glasses and gels). The dynamics of defects will be of future importance, both the annealing of defects in colloidal crystals and the motion of topological defects found in confined colloidal dispersions in liquid-crystalline solvents [113]. These questions could first be explored for one-component systems and then extended towards binary and multicomponent mixtures where a more complex scenario is expected. Another research direction where significant progress is expected during the next five years is the formation and construction of colloidal molecules with sticky-spot

particles. One can play with the whole periodic table (and much more) by self-aggregation of various individual colloidal building blocks. If these particles are offered to tailored substrates, new interface behaviour—both in statics and dynamics—is expected. One example on the kinetics of protein adsorption on tailored substrates is provided in this special issue [27]. Last, but not least, the collective behaviour of self-propelled colloidal particles is expected to become a research area with a wealth of genuinely new nonequilibrium effects, in particular in external fields.

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