Interdisciplinary research with complex plasmas

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Abstract. In this topical review we combine results of research from the field of complex plasmas and colloidal dispersions. This research has opened the way to study strong coupling phenomena in real space and time at the most fundamental kinetic level. The physics of complex plasmas is dominated by the dynamics of slow moving and individually visible microparticles. In contrast to colloidal suspensions,where the fluid background medium results in huge overdamping, the neutral gas background medium in complex plasmas introduces only very little damping so that processes at all relevant time scales can be studied. This is of particular importance for some of the most outstanding questions in the self-organization of matter and critical phenomena.

INTRODUCTION

Many fundamental issues in condensed matter physics such as crystallization, liquid ordering, phase separation, glass formation, etc. can be addressed experimentally using model systems of individually visible particles interacting via a certain potential determined by the properties of the background medium and/or by external "tuning". Clearly, the best known examples of natural model systems for (classical) condensed matter are indeed colloidal suspensions [1, 2, 3, 4, 5, 6, 7] and complex plasmas [8, 9, 10, 11, 12, 13, 14].

"Dusty" or "complex" plasmas are composed of a weakly ionized gas and charged microparticles. Dust and dusty plasmas are ubiquitous in space – they are present in planetary rings, cometary tails, interplanetary and interstellar clouds, the mesosphere, thunderclouds, they are found in the vicinity of artificial satellites and space stations, etc. [15, 16]. Dusty plasmas formed by micron-size particles are actively investigated in many laboratories [17]. Despite an almost a century-long history – the first observations of dust in discharges [18] have been reported by Langmuir in 1924 – the current enormous interest in complex plasmas started in the mid 1990's, triggered by the laboratory discovery of plasma crystals [19, 20]. Today, the physics of complex plasmas is a rapidly growing field of research. Current experimental technologies allow us to create fairly homogeneous clouds containing up to $\sim 10^9$ monodisperse microparticles. The term "complex plasmas" is widely used in the literature to distinguish dusty plasmas specially "designed" for such investigations, from naturally occurring systems.

Colloidal dispersions are solutions or suspensions of mesoscopic solid particles with a stable core and typical sizes ranging from ~ 1 nm to $\sim 1\mu$m, which are embedded in a molecular fluid solvent [1, 2, 3, 4]. These dispersions belong to the material class of soft matter and are therefore susceptible to external perturbations such that they can be

brought out of equilibrium in a controlled way. Since the trajectories of the individual particles can be followed in real space, the particle-resolved information of collective many-body effects is available. This property makes colloids ideal for investigating non-equilibrium phenomena. In the last two decades there is an enormous growth of publications in the realm of colloidal soft matter.

In complex plasmas, the overall dynamical time scales associated with microparticles (e.g., the inverse Einstein frequency) are in the range of tens of milliseconds, yet the microparticles themselves are large enough to be visualized. Thus, the individual trajectories can be obtained by recording with usual CCD cameras and, therefore, fully resolved kinetics can be easily reconstructed. Furthermore, the complex plasma systems are optically thin up to 1000's of particle layers (or more, depending on the plasma parameters that determine the interaction length). This allows full 3D measurements to be made. The two most important aspects are that the pair interaction potential can be tuned and that the rate of momentum/energy exchange through mutual interactions between the charged microparticles can substantially exceed the damping rate due to neutral gas friction. Therefore, the dynamics of individual particles in strongly coupled complex plasmas is *virtually undamped*, which provides a direct analogy to "regular" liquids and solids in terms of the internal atomistic dynamics.

In colloidal suspensions, the dynamics of microparticles is fully damped due to the presence of the viscous solvent. Hence the embedding host fluid thermalizes the system perfectly leading to Brownian motion of the individual particles. Therefore, colloidal dispersions can be brought into *equilibrium in a very controlled way*, complementing the complex plasma approach. Otherwise, the colloidal dispersions have the same advantages as complex plasmas: Fully resolved particle trajectories, both in 2D and 3D, can be easily visualized, the interactions are tuneable, and particles can be manipulated individually.

Thus, by combining research from both fields we are able to investigate a full range of complex phenomena occurring in strongly coupled systems at the most elementary individual particle level – a major breakthrough in classical physics.

In this review we focus on investigations of generic mechanisms operating in strongly coupled media at the "atomistic" (individual particle) level and give some brief examples of current work. In this sense the "review" is only topical, not comprehensive.

ATOMISTIC DYNAMICS OF SUPERCOOLED FLUIDS

Depending on the particle density and/or kinetic temperature, the dynamics of fluids at the atomistic level varies dramatically. As the freezing point approaches, particles become less mobile. The "caging effect" sets in, when at shorter timescales particles exhibit prolonged oscillatory motion in potential wells created by their neighbors. In the supercooled fluid regime, the dynamical heterogeneity becomes the dominant feature: Particles move in increasingly cooperative manner creating dynamically correlated mesoscopic domains [6, 21, 22, 23, 24]. An example of this process in colloids and complex plasmas is illustrated in Fig. 1. Decoupling of "slow" and "rapid" relaxation processes occurs, with the former being related to the mesoscopic cooperative restructuring and the latter to the local in-cage motion [25, 26, 27]. Eventually, below the glass

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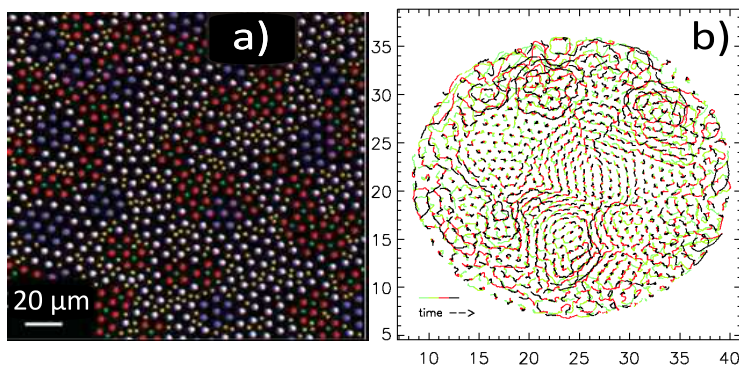
FIGURE 1. Dynamical heterogeneity in 2D strongly coupled (a) colloidal dispersions [21] and (b) complex plasma [22]. In (a) slow particles are coloured in blue and red. In (b), a superposition of consecutive video frames is color-coded from green to black, showing individual particle dynamics (during 10 s, distance is in mm).

transition point, complete arrest of structural relaxations occurs and only local in-cage motions remain.

The physics of supercooled liquids, especially in the vicinity of the glass transition, is one of the most controversial issues in contemporary fluid physics [28, 29]. There are a number of mutually exclusive interpretations of various aspects of the complex supercooled fluid behavior. For instance, there are at least two different scenarios of the dynamical heterogeneity resulting in the stretched-exponential relaxation at long times [24, 30]: One theory relates it to the spatial heterogeneity, and another to fluctuations in the stochastic activation processes. Another major issue is the temperature dependence of the alpha-relaxation timescale (and related transport coefficients, e.g., viscosity). Yet another very important question: What is the dependence of the structural glass transition on the spatial dimensionality. In particular, what is the role of geometrical frustration that – as many believe – is essential for the glass transition (see, e.g., [31]).

Most of the characteristic features peculiar to supercooled fluids (especially, dynamical heterogeneity) are quite general. The particular form of the binary interaction potential plays only a minor role – the described properties have been reported for many model atomic systems, e.g., viscous silica, network-forming liquids [29]. This gives us grounds to expect the behavior of supercooled fluids to be fairly universal, thus making strongly coupled complex plasmas and colloids ideally suited to investigate this topic at the most fundamental atomistic level. Observation of the virtually undamped individual particle dynamics may help us to shed light on what "elementary processes" determine the rich variety of unusual properties of regular supercooled liquids. Weak overall damping would play a constructive role here: It would allow us to control the cooling rate and therefore to bring the fluid to a desirable degree of undercooling (and, hence, vary the glass transition temperature).

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Kinetic investigations of "supercooled" fluids with complex plasmas and colloids may help us to get a deeper insight into various major problems: Which elementary mechanisms determine the stability of supercooled fluids against crystallization [28]? What is the kinetics of the glass transition and how do the relevant processes, e.g., arrest of the structural relaxation and loss of ergodicity, evolve [23, 24]? What microscopically determines the variation of the transport properties (especially, self-diffusion) in the supercooled state [30]? etc. Liquid complex plasmas (where the atomistic dynamics is practically undamped) are apparently the only available model system where the rapid relaxation can be studied at the kinetic level. The question is whether the long-term dynamics is influenced by the short-time dynamics which is different for complex plasmas and colloids.

PHASE TRANSITIONS IN SOLIDS
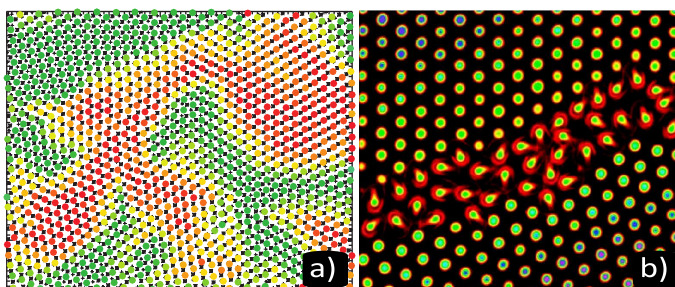
FIGURE 2. (a) Domain structure of a 3D plasma crystal [38]. Three consecutive lattice planes are shown, each particle in the middle plane is color-coded in accordance with the local order (red corresponds to the fcc lattice cell and green to hcp), particles in two adjacent planes are indicated by crosses and stars. (b) Domain interface in a crystalline colloid. [7] Near such interfaces (grain boundaries) the crystal is premelted – the particles move rapidly and show liquid-like diffusion (red represents the most movements, violet is for the least)

Although it is relatively straightforward to measure the atomic structure of (steady-state) 3D crystals, the detailed dynamics of crystal growth (including the evolution of self-organization, structure formation, and the associated kinetic and thermodynamic development) remains one of the most important topics of solid state physics [32, 33]. Very little is known, for instance, about the evolutionary paths of crystal structure development, and the structural complexity of the surface down to atomic dimensions and its change with time. Understanding all of these features is not only a matter of fundamental science – it is also exceptionally important for materials design. In particular, it has become clear that nanoscale structures and properties can have a profound effect on the optical, electrical and mechanical properties of the meso- or macroscopic material.

It is no surprise, therefore, that colloidal suspensions have been widely studied in the past in order to learn more about the generic properties of solids and liquids, melting, freezing and glass transitions [1, 2, 5, 6, 7, 34]. Colloidal crystals can also be used for, e.g., photonics, optical sensors, waveguides, chemical sensing and lithography

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[35, 36, 37]. The fact that their phase behavior is similar to that of some simple atomic or molecular systems make them invaluable tools for studies at the individual particle level – with the associated transfer of new insights into natural atomic (molecular) systems that cannot be resolved in such detail. The only essential limitation of colloids for this purpose is the damping by the suspension fluid, which makes it practically impossible to investigate particle dynamics at the atomistic timescales.

It is clear that fully resolved kinetic measurements of the crystallization process and its dynamics studied with complex plasmas will complement considerably our understanding of the elementary processes. This would open up the possibility to directly compare (and rescale) measurements made at the interesting dynamical frequencies using plasma crystals and colloidal systems. We illustrate this with characteristic examples from complex plasmas and colloids: Figure 2a shows a slice of a 3D plasma crystal where colors indicate different lattice structures found locally, in a single cell around each particle. We see the coexistence of the (presumable) ground state (fcc) and a metastable state (hcp), which seems to mark the domain borders [38]. Such borders are also seen in crystalline colloids (Fig. 2b) between domains of the same structure but different lattice orientation.

As regards the crystallization kinetics, one can observe both "uniform" (homogeneous) nucleation and "crystallization fronts" (sort of heterogeneous nucleation), and which pathway is realized in the experiment depends heavily on the boundary conditions: In the bulk region, where boundaries play no role, the system usually develops towards the uniform nucleation. In this case, one normally observes coexistence of mesoscopic crystalline domains of different structure and orientation (similar to nanostructured regular solids [39, 40], typical examples of such domains in complex plasmas and colloids are shown in Fig. 2). However, closer to the boundaries the crystallization often develops in the form of a front propagating from boundaries inwards. This process is illustrated in Fig. 3.

The distinct peculiarity of the crystallization fronts is the emergence of mesophases or *phaselets* – a distribution of small "droplets" in the crystal phase and small "crystallites" in the fluid phase [9], that are seen in Fig. 3b. The development of the crystallites can presumably be explained in terms of the thermodynamics: In agreement with the classical picture of the nucleation and growth, if the seed crystallite is large enough, the bulk contribution overcomes the surface part and it can grow further. As for the droplets, the mechanism responsible for their formation should be quite different, because thermodynamically, both the bulk and the surface contributions cause the free energy to increase. It is possible that after the initial solidification, a gradual relaxation from a metastable to a ground state (for example, from hcp to bcc or fcc structure, as one can see in Fig. 2a) occurs downstream from the crystallization front. This is naturally accompanied by a release of latent heat, so that the droplets could be a local manifestation of this relaxation.

Another feature, which is peculiar to both homogeneous and heterogeneous nucleation, is the *interfacial melting* – a narrow (few lattice distance extent) premelted region in the crystalline regime where particles exhibit enhanced mobility [9]. Regular solids usually reveal domains of locally ordered regimes (grains), which are separated by domain (grain) boundaries [40]. Thermodynamically, these grain boundaries are different (both in energy and entropy) from the homogeneous crystal regimes within. When such

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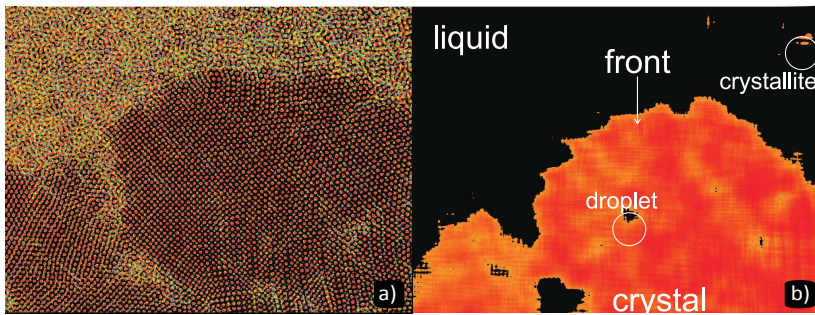
FIGURE 3. (Crystallization front in a 3D complex plasma [9]. (a) Superposition of 10 consecutive video frames (about 0.7s), particle positions are color-coded from green to red, i.e., "caged" particles appear redder, "fluid" are multicolored. (b) The local order, where red implies high crystalline order, black denotes the fluid phase, and yellow indicates transitional regions. Along with the crystallization front, droplets and crystallites are seen that may grow and then dissolve again.

a grainy crystal is heated and approaches its melting point, the grain boundaries may play a special role – they can act as "seeds" of pre-melting regions. The phenomenon of grain boundary melting can be studied in great detail both with colloids [1, 7] and complex plasmas [9], as illustrated in Figs 2b and 3a, respectively. This suggests that the less perfect crystal structure and the associated interfacial free energy is the parameter that determines grain boundary melting.

These examples show that a ubiquitous and still poorly understood process – like crystallization and melting – needs different inputs, different constraints, generalization from different sources and new approaches, so that the principal mechanisms can be identified and combined to a fundamental kinetic theory.

NON-EQUILIBRIUM PHASE TRANSITIONS IN DRIVEN SYSTEMS

The so-called "open" systems are systems that may exchange energy and matter. A remarkable property of nonlinear open systems is the *self-organization* [41, 42] – a spontaneous emergence of stable spatial (or temporal) structures, which are often referred to as "dissipative structures", since dissipation plays a constructive role in their formation. Dissipative structures are the manifestation of non-equilibrium phase transitions, with well-known examples being, e.g., formation of convection (Benard) or turbulent (Taylor) vortices [43]. In order for such transitions to occur, three basic requirements have to be satisfied: (i) Dissipation is necessary, to balance the external influx of energy. (ii) The structures may emerge only in systems described by nonlinear equations. (iii) There must be a relevant control parameter which ensures breaking of symmetry (viz., transition) above a certain threshold.

A remarkable example of a nonequilibrium phase transition is the formation of lanes – a phenomenon occurring in nature when two species of particles are driven
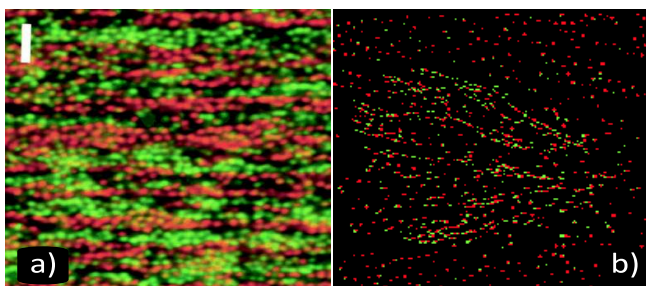
FIGURE 4. (a) Lane formation in experiments with driven colloidal suspensions [45]. Steady-state self-organized flow of positively (green) and negatively (red) charged microparticles is created by an external electric field in the horizontal direction. (b) Lane formation in complex plasmas [50]. A short burst of small (3.4 μm) particles injected into a cloud of large (9.2 μm) background particles are driven from left to right. Each figure is a superposition of two consecutive color-coded images (1/50th s apart, green to red)

against each other. When the driving forces are strong enough, like-driven particles form "stream lines" and move collectively in lanes. Typically, the lanes exhibit a considerable anisotropic structural order accompanied by an enhancement of their (unidirectional) mobility. The phenomenon is most commonly known from pedestrian dynamics in highly populated pedestrian zones [44], but also occurs in different systems of driven particles, such as colloidal dispersions (see Fig. 4a) [45, 46], lattice gases [47] and molecular ions [48]. In other words, this is a ubiquitous generic process of considerable interest in different branches of physics.

Complex plasmas represent a broad class of thermodynamically open systems where numerous types of self-organization can be observed. Figure 4b shows an experiment on lane formation performed with particles of two different size [49, 50]. The net force acting on the particles in a discharge plasma (a combination of the electric and ion drag forces) depends on their sizes and plays the role of an effective gravity pointed to the right (the force is relatively strong at the left edge and almost vanishing at the right edge of the figure). Initially, the large particles form a "background" fluid in hydrostatic equilibrium. When a small fraction of individual small particles entered the system from the left, their sedimentation towards the right edge of the figure is accompanied by a remarkable self-organization sequence: First, the particles form strings flowing along the force field; then, as the field decreases, strings organize themselves into larger mesoscopic streams. At a later stage, when the field almost vanishes, streams merge to form a spheroidal droplet with a well-defined surface, indicating the transition to the regime when the effective surface tension plays the primary role (see next Section).

As we already pointed out, complex plasmas provide a very important intermediate dynamical regime that is between classic undamped fluids and fully damped colloidal suspensions: In complex plasmas, the "internal" dynamics associated with the interparticle interaction is undamped whereas the large-scale hydrodynamics can be strongly affected by friction. Nevertheless, Fig. 4 shows that the mesoscopic appearance of the lane formation in colloids and in complex plasmas is quite similar.

Kinetic investigations of non-equilibrium phase transitions with complex plasmas

and colloids may help us to get a deeper insight into the following major problems: What is the order of this non-equilibrium phase transition? Is it hysteretic (first order) or continuous? If the external drive is oscillatory, lane formation can shift to band formation perpendicular to the drive – this has been seen for colloids [51] and for granulates [52].

PHASE SEPARATION IN BINARY SYSTEMS.
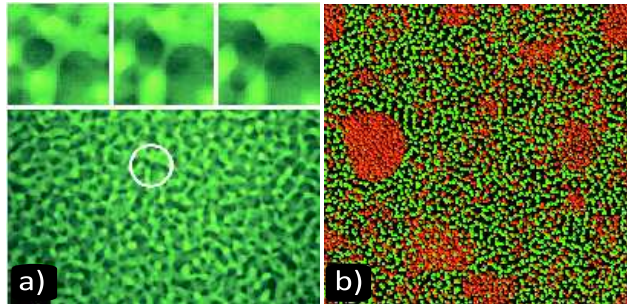
FIGURE 5. Spinodal decomposition in (a) colloid-polymer mixtures [56] and in (b) binary complex plasmas (MD simulations) [60].
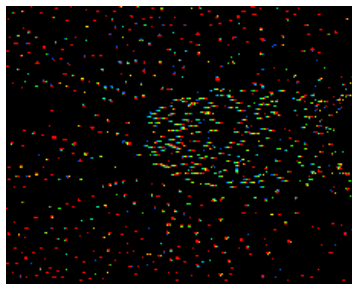
FIGURE 6. Phase separation and formation of a droplet in experiments with binary complex plasmas [59] (later stage of the experiment shown in Fig. 4b).

Phase separation in multicomponent mixtures is a ubiquitous phenomenon [53] occurring in very different systems, ranging from molecular fluids [54] to colloidal suspensions [55, 56]. This phenomenon, despite its long research history, remains of fundamental importance. In particular, the behavior of binary mixtures in the vicinity of the critical point belongs to the same universality class as that of, e.g., conventional liquid-vapor phase transition (provided interparticle interactions are sufficiently short-range), ferromagnetic transition in uniaxial magnets or the 3D Ising model [57].

The tendency for particles of different types to mix or demix is basically determined by the relative strengths (nonadditivity) of their interactions [58]. It is noteworthy that the phase separation in such multicomponent systems does not require an attraction in the interparticle interactions – the necessary condition for the fluid phase transition in

single-species systems. A classical example of the phase separation in binary systems is the colloid-polymer demixing illustrated in Fig. 5a.

The phase separation in binary complex plasmas was recently observed in experiments performed under microgravity conditions onboard the ISS [59, 60]. The initial stage of this experiment is shown in Fig. 4b, where the formation of interpenetrating small- and big-particle lanes was observed. The later stage is illustrated in Fig. 6: When the small particles approached the center of the chamber and thus the driving field practically vanished, an apparent phase separation was observed accompanied by the formation of a small-particle droplet with a well-defined ellipsoidal shape.

The tendency to phase separate does not depend on a particular shape of the interaction potential. The interaction nonadditivity in complex plasmas is solely determined by a nonlinear relation between the actual charge carried by a particle and an effective charge that characterizes the interaction potential at large distances. One can derive the spinodal line for isotropic binary complex plasmas and show that for typical experimental conditions the regime of the spinodal decomposition is easily achievable [59].

Numerical MD simulations performed with the nonadditive interactions typical to complex plasmas fully support the theoretical considerations [60]. As soon as the driving force in the simulations is set below a certain threshold (which mimics the conditions corresponding to Fig. 6) the demixing sets in. It evolves at the timescales of several seconds, similar to what was observed in the experiments, indicating a good quantitative correspondence to the experiment. Figure 5b shows a snapshot from "isotropic" simulations (i.e., without a background driving force). Typical particle sizes in complex plasmas are $\sim 10^2 - 10^3$ times larger than those in colloid-polymer mixtures, so that one can observe the onset of the spinodal decomposition, the coursing and crossover from the (initial) diffusive to the viscous growth regimes, etc. in greatest details at the fully resolved kinetic level.

These results provide us with strong grounds to believe that binary complex plasmas can effectively complement colloid-polymer mixtures and be an ideal model system to study atomistic dynamics of fluid phase transitions and the associated phenomena, such as surface tension, critical behavior, etc.

HYDRODYNAMICS AT THE DISCRETENESS LIMIT

Whilst the macroscopic (hydrodynamic) behavior of fluids has been well studied, the kinetic approach has clearly suffered from the lack of experimental possibilities (or options). Of particular interest could be kinetic investigations of the onset and nonlinear development of hydrodynamic instabilities. Individual particle observations can provide crucial new insights – e.g., whether the coarse-grained concept of basic hydrodynamical instabilities is still adequate at interparticle distance scales [61], is there any microscopic origin of instabilities [62] (in particular, what are the trajectories that trigger instabilities), etc. Apparently, the answer should depend on a particular problem and the similarity variables that play the major role (for instance, for a planar shear flow this is, primarily, the Rayleigh and Mach numbers, for a flow past an obstacle or a droplet breakup this can be the Weber number, etc.).
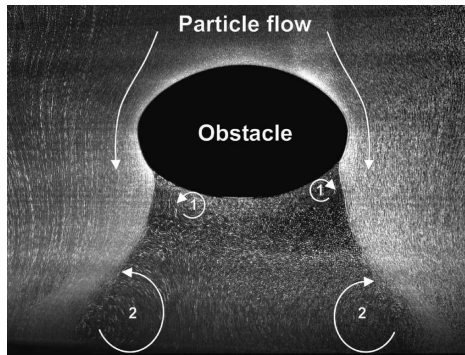
FIGURE 7. Topology of a flow past an obstacle in a fluid complex plasma [62]. The steady vortex flow patterns in the wake are illustrated (exposure time 1 s). The boundary between laminar flow and wake becomes unstable, a mixing layer is formed which grows in width with distance downstream. The system is axially symmetric, shown is a "slice" through the centre.

Complex plasmas can serve as a powerful new tool for fluid investigations on (effectively) nanoscales, including the all-important mesoscopic transition from collective hydrodynamic behavior to the dynamics of individual particles (see Fig. 7). Recent experiments with liquid complex plasmas suggested that the effective mechanism to trigger a local instability of shear flows could be large-angle particle scattering in localized structures and/or inhomogeneities of scales comparable to the particle correlation length [62]. This fruitful hypothesis, however, requires further thorough investigations. Also, the mathematical techniques to quantify the kinetic behavior and to transfer this to macroscopic scales still need to be developed.

In particular, a process to be studied is the development of self-organized nonlinear motion in different flow patterns [43]. Recent complex plasma experiments clearly demonstrated that such measurements, coupled with the appropriate theoretical and numerical simulation support, can provide new insights into both temperature-driven and shear-driven flow phenomena [63].

TUNABLE INTERACTIONS

It is very important to note that there are a number of subtle but very interesting new effects that are believed to occur when the interaction becomes multiscale (e.g., short-range repulsion and long-range attraction): For instance, new regimes of long-time relaxation in supercooled liquids close to the glass transition [27], changes in the surface tension [64], appearance of the liquid-vapor phase transition and the critical point [65, 66], etc. To investigate these phenomena in (canonical) phase space $(\mathbf{r}, \mathbf{v}, t)$ is a very important fundamental step towards a better understanding of self-organization, universality, and scaling in physical systems.

Therefore, to design attractive interactions is a problem of broad interest. This fundamental issue can be resolved by employing the so-called "electrorheological effect"

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which allows us to tune the interparticle interaction to practically any shape – in particular to make it attractive at large distances and change it from isotropic to anisotropic.
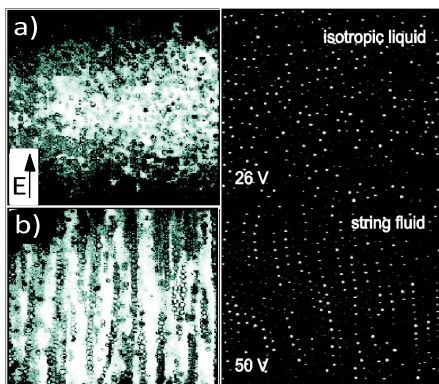
FIGURE 8. Formation of particle "strings" in electrorheological colloids (left column) [70] and plasmas (right column) [73]. Microparticles are illuminated by a thin laser sheet parallel to the vertical ac electric field. Examples of "low" (upper row) and "high" (lower row) fields are shown (the ac voltage is indicated for ER plasmas).

So far, colloidal suspensions have been the major focus for studies of electrorheological (ER) fluids, providing a wealth of information [67, 68, 69]. The interparticle interaction, and hence the rheology of such fluids is determined by an external electric field, which polarizes microspheres and thus induces additional dipole-dipole coupling. When the applied field is sufficiently large, particles arrange themselves into strongly coupled chains ("strings", or even "sheets") along the field [70], as illustrated in Fig. 8. This naturally changes the rheology – e.g., at low shear stresses ER fluids can behave like elastic solids, whilst at stresses greater than a certain yield stress they are viscous liquids again. ER fluids have a significant industrial application potential – they can be used in hydraulics, photonics, display production, etc. [71, 72].

The discovery that complex plasmas also have electrorheological properties adds a new dimension to such research – in terms of time/space scales and for studying new phenomena [73]. Figure 8 highlights the clear similarity in the appearance of ER colloids and plasmas. At the same time, there are important differences in the underlying physics: In contrast to ER colloids where the induced dipoles are due to polarization of the microparticles themselves, in complex plasmas the primary role is played by clouds of compensating plasma charges (mostly, excess ions) surrounding negatively charged microparticles.

The effective interaction in ER complex plasmas consists of two principal contributions [73]: The first "core" term represents the spherically-symmetric Debye-Hückel (Yukawa) part, whereas the second term is due to the interaction between the charge of one particle and the quadrupole part of the wake produced by another particle. The charge-quadrupole interaction is identical to the interaction between two equal and parallel dipoles. This implies that the interactions in ER plasmas are *equivalent* to dipolar interactions in conventional ER fluids.

The principle of unipolar ac field used to create dipole-dipole interactions in ER

colloids and plasmas can be directly generalized to the method of "tuning" the inter-action between particles [74, 75]: One can design new interaction classes of various isotropic/anisotropic and repulsive/attractive forms, by applying external ac fields with bi- and tripolar polarization.

CONCLUSIONS

The research with complex plasmas and colloidal dispersions gives us a unique opportunity to go beyond the limits of continuous media down to the smallest length scale available – the interparticle distance – and thus to study all relevant dynamical and structural processes using the fully resolved motion of individual particles, from the onset of cooperative phenomena to large strongly coupled systems. Individual particles in both systems can be easily manipulated in different ways, so that one can perform active controllable experiments to investigate a particular generic process occurring in liquids or solids at the most fundamental level. This cannot be achieved in any other way known to us.

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