“Tunable colloids”: Experimental complex for studying generic phenomena in classical condensed matter

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Abstract. The experimental complex “Tunable colloids” is developed for particle-resolved studies of generic processes in classical condensed matter. The complex consists of an experimental setup, generating a rotating electric field for a controlled self-assembly of colloidal suspensions (with tunable interactions induced by external fields), and a developed software for phase identification in two-dimensional condensable systems. The complex can be applied to solve a broad range of fundamental and applied scientific problems in condensed matter, chemical physics, physical chemistry, and materials science.

1. Introduction
Colloidal suspensions and complex (dusty) plasma – two domains in soft matter – represent two broad classes of systems, mimicking molecular-like many-body behavior at the macroscopic level that result, in particular, in ability to observe self-assembly and phase transitions [1–6].

Due to interaction with solvent playing a role of Brownian thermostat, the thermal energy of particles in colloidal suspensions is determined by to ambient temperature. Owing to diffusion of the particles in the solvent, the system relaxes to its statistical equilibrium state and this feature strictly distinguishes colloidal suspensions from granular media. Characteristic times of collective processes in such systems are much larger than diffusion time for individual particles. Therefore, generic phenomena in strongly coupled media can be directly observed with resolution of individual particles in real-time regime using video microscopy [7, 8]. Particle-resolved studies allow to analyze spinodal decomposition [9, 10], glass formation [11, 12], crystallization [13, 14], martensitic transformations [15, 16], two-dimensional (2D) melting [17, 18], coalescence [19], phenomena in vicinity of critical point [20], domain dynamics in crystals [21, 22] etc. Apart from particle-resolved studies of fundamental phenomena in condensed matter, there are a lot of applications of colloidal suspensions with induced tunable interactions, for instance, for production of photonic crystalline structures [23, 24], or for manipulation of biological cells [25, 26], their separation [26–30], transportation [31–34], and sorting biological objects [35].

Development of novel technologies for tunable self-assembly in colloidal suspensions is an important problem of fundamental and applied physics, chemical and material sciences, biology
and medicine. Technologies for assembling the colloidal particles employ sedimentation in gravitation field [36, 37], assembly by inclined deposition [38, 39] and depletion forces [40, 41], assembly in flows [42, 43], with interparticle attraction induced by external magnetic [14, 45] or electric fields [6, 46, 47].

In this paper, we present an experimental complex “Tunable colloids” for particle-resolved studies of the generic phenomena in classical condensed matter. The complex realizes tunable interactions between colloidal particles in external rotating electric fields. It consists of an experimental setup for generating an external electric rotating field [6, 47] and a special software for recognition and tracking of colloidal particles, followed by identification of condensed and gaseous phases using analysis of Voronoi cells [48].

2. Experimental Setup

In this section, we briefly describe composition of the experimental setup for generation of external rotating electric field. The setup is described in detail in Ref. [6]. Also in Ref. [6], it was shown that the homogeneity of the electric field in the experimental cell is very high and provide enhanced characteristics comparing to analogs.

Figure 1 sketchily shows a cuvette for samples, microscopic and experimental cell part of installation for tunable self-assembly of colloidal particles in rotating external electric fields. Camera with microscopic objectives enable to resolve dynamics of particles in experiment using video microscopy. To build the optical part of setup were used 8 Megapixel Color Scientific CCD camera with microscopic objectives (magnification 20x, 40x 60x or 100x). The accurate movement of the experimental cell relatively to the optical axis was achieved using a stage with
microactuators. Broadband halogen fiber optic is used for illumination of the sample. The experimental cuvette consists of two standard coverslips glasses (20x20x0.15 mm), separated by a 10-µm-layer of polymer spacer between them applied along the perimeter of the coverslips glasses. Electrical field generating on eight metal (silver, chromium or titan) electrodes, having the shape of arrows, with thickness of 100 nm, were made lithographically at the transparent silica substrate.

The electric field is controlled by four sinusoidal signals produced by voltage signal generation unit and shifted consistently by the phase of $\phi = \pi/4$. The amplitude of signals is varied in the range 0 – 2500V, and it frequency varies in the range 40Hz – 30kHz. All glass surfaces of the cuvette were treated with deionized water, ethyl alcohol, and then dried in a muffle furnace at 150 °C for half an hour. After cleaning the glass, working surfaces were coated by a hydrophobic layer. A sample of the colloidal suspension was placed in cuvette in the gap between the coverslips glasses. The voltage signal generation unit with an experimental cell allow to generate rotating electric fields with a specified frequency and electrical strength.

As a sample, we use colloidal particles of silica, polymethyl methacrylate (PMMA) or polystyrene in deionised water (resistivity – 18.0 MΩ × cm). Diameters of particles, which we used for controlled self-assembly induced by rotating electric field, are from 1 µm to 10 µm.

Colloidal particles become polarized by external electric field, that leads to dipole-dipole anisotropic interactions between particles. Due to high frequency of field rotation in the plane of self-assembly (comparing to the inverse diffusion time for the colloidal particle), the interaction between particles becomes averaged angularly, and attains dipole-like long-range isotropic attraction. The magnitude of the attraction can be tuned by change the field magnitude.

3. Post-Processing

With reduce in temperature, single-component systems undergo spinodal decomposition: An initially homogeneous phase decays into high- and low-density ones, known as condensate and gas, respectively [49]. Such processes are inherent not only in molecular systems but also in complex matter, like colloidal suspensions, complex fluids and plasmas [9]. For example, colloidal suspensions with different kind of attraction between particles, provided by magnetic forces [13, 15], electric fields [6, 46, 47], and other mechanisms, demonstrate spinodal decomposition. Modeling techniques (molecular dynamic and Monte-Carlo), as well as particle-resolved experimental studies, together with post-processing techniques, are a powerful tool for studying such complex processes in classical solids and liquids. With the use of particle-resolved methods, it is possible to study generic phenomena in such systems, identifying generic laws governing evolution of many-body systems. In this work, we showing previously developed method for phase identification in 2D systems, described in details in Ref. [48].

As shown earlier, in 2D system of individual particles there are two phases: condensed clusters and a gas. Phase recognition of such systems has been a difficult problem. In [50, 51], this problem was solved using data on particle velocities and the interaction potential, which were known with high accuracy. These data are extremely difficult to reconstruct with high accuracy in experimental conditions. We propose a method based on the particles tracking, which can be performed with high accuracy in the experiments. Using the coordinates of all particles, we identify particles belonging to condensed clusters, gas, and interface (“surface”) between them.

Phase identification in 2D condensable systems is based on the analysis of Voronoi cells characteristics. The developed algorithm for determining the particles belonging to condensate, gas state, and interface between them (the “surface”) is described in detail in Ref. [48]. Examples of phase identification is shown Fig. 2. Figures 2a-2c present photos of experimental colloidal cluster of 2.12-µm-diameter colloidal silica particles assembled in a rotating external electric field at different regimes of interaction, while Figs. 2d-2f present corresponding results of phase
Figure 2. **Phase identification in 2D colloidal system**: Photos of colloidal system and result phase recognition: a,b,c are photos of colloidal system at different regimes of interaction between particles; d,e,f correspond to the identification results. Particles colored by blue are the condensate, yellow particles are in the gaseous state and the white particles belong to interface. Identification. Particles colored by blue belong to the condensed phase, while yellow particles are in the gaseous state, and the white particles are the interface between them. The stability and efficiency of the method were studied in Ref. [48] using molecular dynamics simulations in 2D systems with short- and long-range attraction. This method of post-processing is an inalienable part of the complex for studying nonequilibrium processes in soft matter.

4. Conclusions

In conclusions, we developed novel experimental complex for fundamental and applied particle-resolved studies. The complex employs self-assembly of colloidal suspensions due to attraction induced by external rotating electric field and phase identification method. The results can be applied for studies of non-equilibrium processes and different states in soft matter, e.g., spinodal decomposition, phase transitions solid-solid, 2D melting and crystallization, coalescence, domain dynamics, gelation, and critical phenomena. Also, it is possible to use the developed system for assembly of monolayers of particles or photonic crystals, and for development of new medical
diagnostic methods based on the controlled assembly of biological cells.

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