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Non-equilibrium phase transitions in complex plasma

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Abstract

Complex plasma being the ‘plasma state of soft matter’ is especially suitable for investigations of non-equilibrium phase transitions. Non-equilibrium phase transitions can manifest in dissipative structures or self-organization. Two specific examples are lane formation and phase separation. Using the permanent microgravity laboratory PK-3 Plus, operating onboard the International Space Station, we performed unique experiments with binary mixtures of complex plasmas that showed both lane formation and phase separation. These observations have been augmented by comprehensive numerical and theoretical studies. In this paper we present an overview of our most important results. In addition we put our results in context with research of complex plasmas, binary systems and non-equilibrium phase transitions. Necessary and promising future complex plasma experiments on phase separation and lane formation are briefly discussed.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

A complex plasma [1, 2] is a self-consistent system of (highly) charged dust grains immersed in a plasma. The dust grains are “macroscopic” particles with respect to other plasma species (electrons, ions and neutrals). One grain consists of hundreds of thousands or millions of atoms and is at least nanometers in size. In most complex plasma experiments mono-disperse spherical grains with a diameter of 1–10 μm are used. Each grain collects several thousand elementary charges and acquires a substantial (usually negative) surface potential. The full kinetics of individual dust grains can be investigated, e.g., by recording scattered laser light with commercial off-the-shelf optics and cameras. The dust grain motion is damped mainly through

collisional drag associated with the neutral gas component. Typical laboratory conditions with pressures between a fraction of a pascal and hundreds of pascals allow us to investigate the full range from virtually undamped to fully damped dynamics.

Charging of dust and the shielding of the electrical potential by the surrounding plasma is a broad field of ongoing research in laboratories and theory [3–11]. When an isolated dust grain enters a plasma it is subjected to electron and ion currents, leading to a dynamic equilibrium surface potential. The resulting electric potential is shielded from the plasma, by a process very similar to the formation of a sheath. For low-temperature plasmas this potential is negative compared with the plasma potential, due to the higher mobility of the electrons. Charging time is of the order of 10^{-6} – 10^{-5} s for typical laboratory plasmas. As the surface potential of a dust grain depends strongly on local plasma conditions, the grain charge will adjust to changes in the surrounding plasma conditions. In addition, the charge on the dust is subject to statistical fluctuations, which cannot be neglected for sub-micrometer-sized dust, and the coupling to the local plasma environment can result in instabilities for dust grains of several micrometers in size [12, 13]. In the case of streaming ions, the presence of dust can result in ion focusing, creating wake regions of enhanced positive space charge behind the dust grain. By applying external fields, this ion focusing effect can be used to create specific ‘tuned’ spatial distributions of the potential around each dust grain [14].

The transition from ‘physics of dust in plasma’ to ‘physics of complex plasma’ happens when either the charged dust grains cannot be treated individually, or the plasma cannot be considered undisturbed by the dust. Complex plasmas are usually described in terms of Γ and κ parameters. The so-called coupling parameter Γ denotes the ratio of potential energy of the inter-grain interaction to the thermal energy of the grains. The screening (lattice) parameter $\kappa = \Delta/\lambda$ is the ratio of mean distance Δ between dust grains to the length scale of grain–grain interaction λ (plasma screening length). It is clear that these two parameters depend strongly on the exact nature of the grain–grain interaction mechanism. One of the main interaction mechanisms is clearly electrical repulsion between like charged grains. The shape of the electrical interaction potential is defined by grain charging and the shielding of their surface potential. The charging and shielding of many grains interact strongly with the embedding plasma, and interfere with each other. The inter-grain interaction potential can often be approximated by a Debye–Hückel (also known as Yukawa) form, even though investigations over the last ten years generally predict significant deviations (especially at long inter-grain distances) [3, 8–11, 15–22]. Strong inter-grain coupling allows complex plasmas to attain liquid and crystalline states enabling the investigation of flows, crystallization and melting. Complex plasmas are an interesting model for strongly coupled Coulomb systems (SCCS) (or Yukawa type systems), allow for the study of wave phenomena, e.g., dust-acoustic, dust-lattice and solitary waves, shocks, etc.

Complex plasmas have recently been perceived as a state of soft matter [23]. Other members of the soft-matter family are polymers, gels, foams, colloids and granular media. All these systems share fundamental properties, while each has special unique advantages, either in application, or in its suitability to do specific experiments. Complex plasmas have properties that make them well suited to the research of hydrodynamics at the discreteness limit, the transition from laminar to turbulent flow, dissipative structures (e.g., shocks, vortices, dissipative solitons), physics at the critical point and (the dynamics of) non-equilibrium phase transitions. All these problems can be studied at the most fundamental individual-particle level. Such research using complex plasmas as model systems can give important insights into fundamental physics that are of a wide interest outside the immediate field of complex plasmas: like melting of 2D crystals [24], reentrance effects in phase transitions [25, 26] and non-equilibrium phase transitions, as for instance shear induced melting [27].

In this paper we report on two specific non-equilibrium liquid–liquid phase transitions occurring in binary complex plasma: phase separation and lane formation. We were able to conduct laboratory experiments exhibiting lane formation and phase separation, and their transition. Lane formation is a ‘classical’ non-equilibrium phase transition (as known from open systems) that develops in driven dissipative systems. Unlike this, phase separation occurs for systems of binary mixtures with well-defined free energy that are forced into the unstable state of fully developed spinodal decomposition. These two manifestations of non-equilibrium phase transitions are in general independent. Yet we were able to observe a transition from lane formation to phase separation with decreasing Weber number. These unique experiments were enabled by the PK-3 Plus [28] setup, a permanent microgravity complex plasma laboratory onboard the International Space Station (ISS). The experimental setup and observations are described in section 2. Phase separation is well known from mixtures of macroscopic liquids that undergo a transition from a homogeneous to a heterogeneous mixture, when cooled below a certain temperature. In section 3 we will describe how phase separation can happen in systems with only repulsive interaction through the so-called non-additivity of binary interactions. We will recapitulate that the dynamics of phase separation depends strongly on the grain–grain interaction potential. Additionally, this non-equilibrium phase transition offers an opportunity to explore physics at the critical point. Once two phases of complex plasma liquids have been separated, non-additivity will give rise to an effective surface tension on the phase boundaries [17, 29]. If the two phases are driven against each other, a Rayleigh–Taylor instability develops across the boundary layer. Two of us (AW and HL) have shown [30] that a Rayleigh–Taylor instability at the discreteness limit coincides with lane formation—which is best known from research of pedestrian dynamics [31]. This transition from a Rayleigh–Taylor instability to lane formation is controlled by the ratio of surface tension to driving force. Lane formation in driven binary complex plasma is described in more detail in section 4. In the concluding section 5 we will give an outlook on current research topics.

2. Experimental setup and observations

The experiments leading to our current results were performed on board of the ISS using the PK-3 Plus [28] laboratory operated by Russian cosmonauts, a unique research opportunity for the complex plasma community operational in orbit since 2006. The main goal of PK-3 Plus is to study in detail all aspects of condensed complex plasma [32]. That is, the liquid and solid state of complex plasmas and the melting and crystallization dynamics [33, 34]. In addition PK-3 Plus is intended to investigate physics at the critical point and multi-component complex plasmas.

The PK-3 Plus laboratory (see figure 1) uses a capacitively coupled radio-frequency (13.56 MHz) parallel plate discharge chamber. The electrodes are disks with 6 cm diameter, surrounded by 1.5 cm wide grounded guard rings. Distance between the electrodes is 3 cm. The discharge chamber itself is a cuboid 10 cm × 10 cm × 5.4 cm in size. The outer walls are quartz glass windows, which allow visual inspection of the plasma glow and the dust species of the complex plasma. Six different mono-disperse spherical dust grains can be injected separately into the chamber: silica particles with 1.55 μm diameter, and melamine-formaldehyde particles with diameters of 2.55, 3.42, 6.81, 9.19 and 14.9 μm . The dust is illuminated with a thin ($\sim 100\text{--}300 \mu\text{m}$) sheet of laser light. The light scattered by the dust grains is recorded under 90° by a total of four PAL progressive scan cameras. The cameras have different magnifications and different but overlapping fields of view (see figure 1). In the PK-3 Plus laboratory we can use either argon or neon gases at pressures in the range 1–100 Pa. The gas is electron impact ionized mainly in the center of the PK-3 Plus discharge chamber.

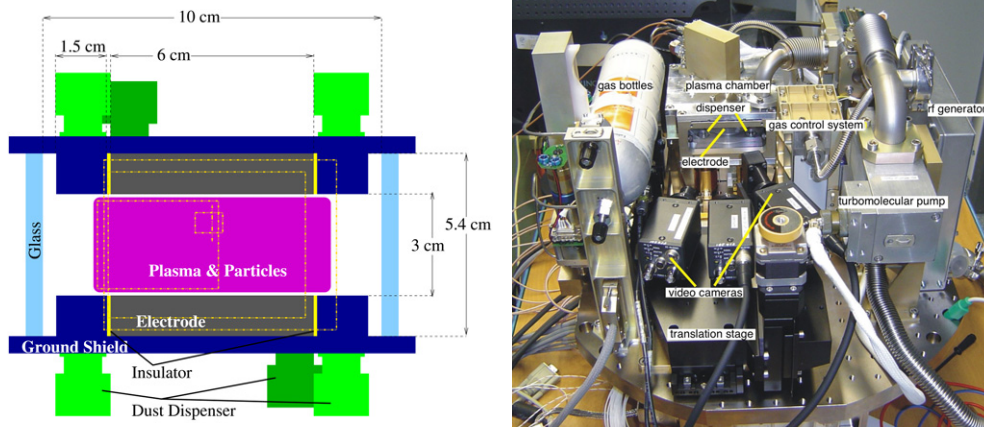


Figure 1. The PK-3 Plus laboratory. To the right is a photograph of the fully integrated laboratory as it is flying on the ISS. To the left is a schematic view in line of sight of the four PAL progressive scan cameras. The dashed-dotted rectangles depict the fields of view of the hires-, quadrant-, overview- and glow-camera (from smallest to largest field of view). The hires-camera can be moved up and down (indicated by small arrows). The glow-camera looks ‘from behind’. Six dust dispensers can inject mono-disperse dust grains of different sizes through the guard ring into the discharge chamber.

Under microgravity the strongest forces acting on individual dust grains in a complex plasma originate from the global confining (pre-)sheath/ambipolar electric field and the counteracting ion drag. The main plasma production takes place in the center of the chamber; plasma loss occurs mainly on the chamber walls and electrodes. As the electrons have a much higher mobility, they initially diffuse much quicker toward the walls of the setup. This instantly results in an electric field pointing outward, slowing down the electrons and accelerating the ions. Ultimately ambipolar diffusion of electrons and ions and a positive plasma potential establish. The negatively charged dust grains are pulled toward the center by the ambipolar field E but pushed away from the center by the drag of streaming ions [35, 36]. The ion drag force mainly depends on the grain diameter $2a$ and on the ion streaming velocity v_i , which itself is a function of E and pressure p . This allows us to change the force acting on the dust particles by changing the discharge conditions. For our experimental setup the net force F acting on an individual dust grain can be defined implicitly via its position r in the discharge chamber: $F = F(E(r), v_i(E(r), p), a) = F(r; a, p, \dots)$. This net force becomes zero at some distance $R = R(a)$ from the center of the chamber, closer to the center for smaller grains. Particles ‘inside’ (‘outside’) R are pushed away from (pulled toward) the center. This results in a dust free region at the center of the discharge chamber, the so-called ‘void’ [37–39].

In our current lane formation and phase separation experimental runs (see figure 2) we first fill most of the discharge volume with large dust grains to form a fairly homogeneous background complex plasma⁵. Then we inject smaller grains into the periphery outside this background. These smaller grains are pulled by the net force of confinement and ion drag through the background of larger grains toward the center and settle around the perimeter

⁵ An important results from PK-3 Plus research has been that inside such a homogeneous one-component complex plasma the net force F between ambipolar electric field force and ion drag force becomes very small in the symmetry plane of the chamber [40]. In the interpretation of our experiments we use this fact by assuming zero net force acting on large particles in the numerical simulations [41, 42].

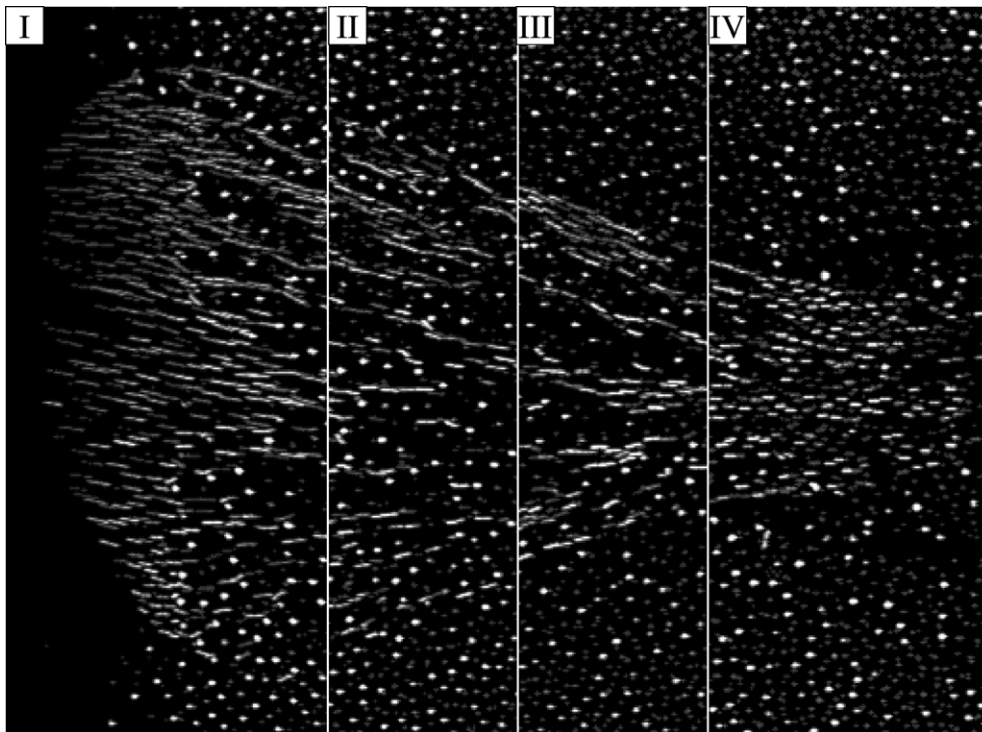


Figure 2. Experiment showing lane formation and phase separation in binary complex plasma. A complex plasma composed of large dust grains fills most of the discharge volume. Small grains are injected from the left. They are driven toward the center of the discharge chamber through the complex plasma of large grains. The dust grains are illuminated from the side by a thin ($\sim 100\text{--}300\ \mu\text{m}$) laser sheet, the scattered light is recorded under 90° . Only this narrow vertical layer out of the whole complex plasma can be seen in the recorded videos. The small grains move noticeably during one video frame (0.04 s) and can therefore be identified as streaks in the images. The penetration develops in four steps: (I, $t = 0\ \text{s}$) small grains clash against the background complex plasma, only a few grains penetrate into the cloud of large grains; (II, $t = 0.4\ \text{s}$) more small grains follow behind the forerunners, thereby forming lanes. The flow of small grains combs the large grains into lanes, too; (III, $t = 1\ \text{s}$) while the small grains approach the center, the net force diminishes and the lanes unite to form larger streams; (IV, $t = 2\ \text{s}$) at some characteristic distance from the center, the driving force becomes negligible, and the small grains form a single drop of complex plasma. This marks the transition from lane formation to phase separation, where effective surface tension is dominant.

of the void. This penetration shows some very interesting dynamics: at first the complex plasma of small grains clashes against the background of larger grains pushing the whole cloud collectively. Only very few small grains penetrate into the cloud of big grains. Then, more small grains follow behind these forerunners creating lanes of small grains instead of diffusing through the background individually. In short time the large grains are combed into lanes by the small grains flowing past them, too. Both particle species thus form an array of interpenetrating lanes. As the small grains approached the center, two more things happen. First, the individual lanes come closer to each other because of the central force pulling the particles, and individual lanes merge into larger streams. Second, the net force gets weaker and weaker. At some characteristic distance from the center there are no more individual lanes of small grains, but instead they form one big drop of complex plasma, which indicates the

onset of the phase separation regime. This drop deflects big grains at its bow, creating a flow of big grains around the object, and big grains captured inside the bubble of small grains are squeezed out.

Interestingly enough, the lanes combed into the big grains persisted for more than a second after the flow of small grains subsided. This is more than ten times longer than the characteristic self-diffusion time of the big grains.

3. Phase separation of binary complex plasmas

As mentioned in the introduction, phase separation is a non-equilibrium liquid–liquid phase transition, where a homogeneous mixture separates into a heterogeneous mixture via spinodal decomposition when cooled below a certain temperature [43]. The mixing-gap responsible for this phase transition in binary complex plasma derives from a positive non-additivity (see next paragraph) of the grain–grain interaction potential. We are especially interested in two features of phase separation. First, the dynamics of phase separation strongly depends on the specifics of particle interaction potentials. This allows us to test different theories for the interaction potential. Second, phase separation has a critical point, which belongs to the same universality class as the liquid–vapor critical point of regular simple fluids. Physics at the critical point is a major research topic of the PK-3 Plus laboratory.

For a mixture of point particles (of sort 1 and 2) with a repulsive interaction, phase equilibrium is determined by an asymmetry between the ‘self-interaction’ energies $W_{11}(r)$ and $W_{22}(r)$, and the ‘cross-interaction’ energy $W_{12}(r)$ (Lorentz–Berthelot mixing rules [44]). This asymmetry is characterized by the non-additivity parameter δ : $W_{12} = (1 + \delta)\sqrt{W_{11}W_{22}}$. For $\delta > 0$ particles 1 and 2 tend to ‘avoid’ each other, which can result in phase separation, effective surface tension⁶, etc. For $\delta < 0$ particles 1 and 2 tend to stay close to each other, resulting in, e.g., active mixing of separated species. The value of δ naturally derives from the particle interaction potential.

It is important to note that even though phase separation is a non-equilibrium phase transition, the system has a well-defined free energy and the concept of a non-additivity parameter is fully applicable. Unlike lane formation it does not derive from an external driving force, nor does it depend on some external energy input into the system as is common among non-equilibrium phase transitions. Phase separation in binary complex plasmas is a consequence of the non-linearity of the particle interaction potential. The exact interaction potential between the dust grains in a complex plasma is under active research [8–11, 14–16, 18–22]. For the topic of phase separation it is very important to note that binary mixtures of complex plasmas always have a positive non-additivity [45] irrespective of a particular form of the interaction potential. That means, phase separation can always be stimulated in these systems.

As a corollary, detailed time resolved phase separation experiments allow us to test different theories related to the grain–grain interaction potential in complex plasmas. Unfortunately, the conditions of our current phase separation experiments (cf figure 2) always lead to a separation of dust grains of different sizes under the action of the external confinement

⁶ In a binary mixture with purely repulsive particle interaction with positive non-additivity, phase separation of particles of sort 1 and 2 is attained because the systems tries to minimize the particle interaction energy, i.e., the ‘surface’ between unlike particles is minimized: this relates directly to the concept of ‘surface tension’ in classical liquids. Effective surface tension can also be understood from the point of view of an effective attractive interaction between like particles [17]: even though there is only repulsive forces between particles in our binary mixture, the forces between all particles are fully compensated. This means that forces between particles attain a relative magnitude, where less repulsion is equivalent to attraction.

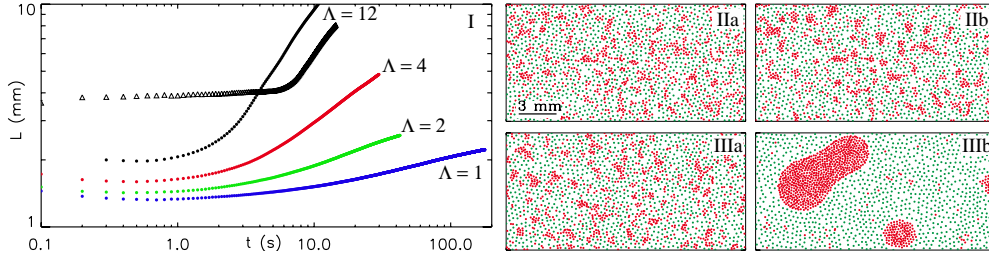


Figure 3. Numerical study of phase separation in binary complex plasma. The influence of long-range (LR) and short-range (SR) non-additive particle interaction potentials was investigated using a double-Yukawa potential with $\lambda_{LR} = \Lambda \lambda_{SR}$ [29]. Panel I displays $L(t)$, the characteristic length of growing domains, versus time, obtained for four different values of Λ (from bottom to top): $\Lambda = 1$ (blue online), $\Lambda = 2$ (green online), $\Lambda = 4$ (red online), $\Lambda = 12$ (black online). The black triangles show results for simulations where only the LR interactions are non-additive. Panels IIa–IIb display 0.3 mm thick slices from the simulation domain, for $t = 1$ s (a) and $t = 10$ s (b) respectively. Panels IIa and IIb show domain morphology for $\Lambda = 1$ (i.e., no LR potential), IIIa and IIIb for $\Lambda = 12$. The minority species (colored in red online) has a higher coexistence density than the majority species (green online).

and ion drag. This is certainly not what we mean by phase separation, and is more like phase segregation or sedimentation. Due to this fast phase segregation we were so far unable to investigate the temporal evolution of spinodal decomposition with laboratory experiments. Therefore, we made use of dust particle resolved numerical simulations at the Langevin level (see figure 3) to predict the dynamical behavior for different discharge conditions and different models of the inter-particle potential [29]. Our systematic investigations revealed the sensitivity of phase separation dynamics to the exact nature of the interaction potential. We specifically highlighted the critical role of the competition between long-range (LR) and short-range (SR) interactions at the initial stage of the spinodal decomposition. The numerical simulations also allowed us to estimate the effective surface tension resulting from different values of non-additivity and different models of the particle interaction potential.

Even though we were unable to resolve the dynamics of spinodal decomposition in laboratory experiments, our PK-3 Plus measurements allow us to determine an important self-adjusting property of non-additive binary mixtures: the average distance \bar{d}_{ij} between grains of species $i, j \in [s, l]$ (s and l denote small and large grains, respectively). For separated phases \bar{d}_{ss} and \bar{d}_{ll} correspond to the so-called coexistence densities, which are well defined by the value of non-additivity. We could show that the theoretical estimates for the coexistence densities match the values determined for the numerical simulations, and agree well with the values measured in the experiment. The cross term \bar{d}_{sl} which gives the average separation between grains of different species can be measured in the mixture, and along phase boundaries. As \bar{d}_{ij} is a direct result of the interaction energy, from $\bar{d}_{ss} < \bar{d}_{ll}$ follows $\bar{d}_{ss} < \bar{d}_{sl} < \bar{d}_{ll}$ for additive mixtures. In the case of non-additivity \bar{d}_{sl} will become larger for increasing δ . For our laboratory measurements $\bar{d}_{sl} \gtrsim 450 \mu\text{m}$ which was larger than $\bar{d}_{ll} \approx 420 \mu\text{m}$.

4. Lane formation in driven binary complex plasmas

Lane formation is a form of self-organization that occurs whenever two particle species are driven through each other with sufficient but not too strong force. It is a representative of dissipative structures, i.e., a manifestation of a non-equilibrium phase transition in open dissipative systems. Lane formation is ubiquitous and has been researched for,

e.g., lattice gases [46], pedestrians [31], colloidal suspension [47–49], and molecular ions [50]. The occurrence of lanes has been shown to be independent of cognitive abilities or communication—for instance an interaction potential between the particles—even driven hard spheres form lanes. Lane formation coincides with a Rayleigh–Taylor instability at the discreteness limit [30].

Previously lane formation has been studied mainly for its (temporal) asymptotic behavior and its dependence on external parameters (like driving force). Extended numerical and theoretical investigations on lane formation in colloids were performed by one of us (HL) and collaborators [51]. Due to the (tunable) low damping in complex plasmas and the possibility to record the full kinetics of single dust grains, binary complex plasmas offer a unique research opportunity: namely to resolve the dynamical onset of lane formation, and investigate the temporal evolution of lanes and its dependence on complex plasma parameters. The complex plasma dynamics can be considered virtually undamped at the time scale of dust particle interactions, allowing the temporal resolution of all relevant frequencies. Still, the system has substantial dissipation, enabling the occurrence of dissipative structures in the first place.

The onset of lane formation was studied in experiments with driven binary mixtures of complex plasmas described in section 2. The observed time-resolved lane formation process was compared with three-dimensional molecular dynamics simulations at the Langevin level. The theoretical model behind these assumed that grain–grain interaction can be described by a Debye–Hückel form, defined via the grain charge $Q(a)$, with grain radius a and plasma screening length λ , which depend on each other by a fixed coupling strength $\Gamma = Q^2 \exp(-a/\lambda)/\Delta k_B T$, where $k_B T$ is the kinetic dust temperature. The extent of lane formation was quantified in terms of the anisotropic scaling index, leading to a universal order parameter for driven systems.

The main results were as follows. (i) A universal order parameter to quantify the lane formation non-equilibrium phase transition. (ii) The dynamical onset of lane formation can be resolved in our laboratory experiments. (iii) The small grains form lanes practically immediately, within inter-particle distance of the background complex plasma. (iv) The much more inert large grains are arranged into lanes by the passing flow of smaller grains, too. (v) In the laboratory lanes in the background persist much longer than the self-diffusion time of large particles. (vi) The dynamics of lane formation depends strongly on the internal parameters of the complex plasma, and on the initial phase of the background complex plasma. (vii) Internal parameters of the complex plasma that are difficult to measure directly in the PK-3 Plus laboratory, e.g., screening length λ and particle charge Q , can be estimated by varying them in the MD simulations.

Details on our laboratory and numerical experiments as well as on the order parameter we developed were published in [41]. A study of the dependence of lane formation dynamics on complex plasma parameters, based on our numerical simulations, was published in [42].

5. Conclusion and outlook

Lane formation and phase separation are hot topics in soft-matter research for several reasons. They have been shown to be two sides of the same coin, i.e., a Rayleigh–Taylor instability at the discreteness limit [30], discriminated by the ratio of surface tension to driving force, or Weber number. Lane formation is a dissipative structure, like vortices, shocks or dissipative solitons [52]. Phase separation has a critical point which belongs to the same universality class as the liquid–vapor critical point of classical fluids. In addition, lane formation and phase separation stimulate fundamental complex plasma research.

Lane formation supplies us with interesting new insight into the complex plasma and new experimental venues of fundamental complex plasma research. Two examples are:

- The decay of lanes in the background plasma which showed some kind of hysteresis or memory-effect. This provides a strong hint on the existence of metastable states.
- Lane formation experiments in complex plasmas with streaming ions and ion focusing could allow us to probe the ion wake structures between large grains.

One of the immediate science goals for phase separation experiments is the exploration of physics at the critical point. For phase separation, which has been ‘proven’ to exist for all binary complex plasmas, the search for the critical point is straightforward, and in principle reduces to a binary search—at least for numerical experiments. For these experiments we need to attain very homogeneous conditions. Experiments on phase separation in binary complex plasmas are currently only possible using the PK-3 Plus setup onboard the ISS. This limits severely the diagnostic possibilities and the number of parameters that can be checked systematically. Additionally it is very difficult to suppress the strong net force segregating grains of different sizes (cf sections 2 and 3). An important goal for the near future is the adaptation and design of ground based or parabolic flight experiments, using the full possibilities of current diagnostics, especially on plasma parameters and recording of grain trajectories using high speed cameras, possibly in 3D.

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