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Topical: Critical Point Investigations using Complex Plasmas in a Microgravity Environment

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Introduction: Phase Transitions and the Critical Point

Phase transitions are one of the most interesting phenomena in physics. Kenneth Wilson received in 1982 the Nobel Prize for his seminal discoveries on the universality of phase transitions based on the renormalization group, playing an important role also in high-energy particle physics. Phase transitions exhibit different orders characterized by the continuity behavior of the order parameter. In the case of water, for example, the solid-liquid and solid-gas transitions are of first order. However, the liquid-gas phase transition starts in the phase diagram (pressure vs. temperature) as a first order transition but ends at a critical point. Beyond this point a supercritical system exists which shows no clear separation between the two phases. At the critical point the phase transition is of second order, where fluctuations on all scale are present leading to critical opalescence allowing the experimental identification of the critical point. However, since under gravity there is a pressure gradient in the experiment chamber, the critical point, e.g. in water or SF_6, can be seen only in a plane. Under microgravity conditions, e.g. in the DECLIC facility on the International Space Station (ISS), on the other hand, critical phenomena such as critical opalescence can be observed in the entire chamber [1].

A critical point in a phase transition exists only in case of an attractive interaction between the particles. In molecular gases this is realized by the Lennard-Jones potential. If there is only a repulsive interaction, only a supercritical system showing no phase transition is possible. *Complex or dusty plasmas are ideal model systems for studying fundamental aspects of many-body systems in physics on the microscopic level [2]*. The behavior of the micro-particles, embedded in a low-temperature discharge, can be observed and recorded easily by laser illumination and cameras, *revealing the time evolution of the full 6-dimensional phase space of the system*. Owing to the string interaction between the highly-charged micro-particles, gas, liquid, and solid phases can be present within the micro-particle many-body system and the transitions between them can be studied [3]. The interaction between the micro-particles is assumed to be in most cases purely repulsive, resembling the form of a Yukawa potential while being confined as a whole by the plasma electric fields. As such no critical point in the liquid-gas transitions is expected under this circumstance. However, *with the introduction of an attractive component to the particle interaction by the here proposed user-controlled experiment manipulations, the investigation of critical point phenomena on the quasi atomic scale can be performed by utilizing complex plasma in a microgravity environment.*

A proposal to utilize upcoming ISS based space experiments and supporting a space based complex plasma facility development that enables critical point studies aboard the ISS

To perform critical point phenomena studies on the dynamical and spatial scales of the individual „atoms“, we propose to use complex plasma as a model system. To allow the study of a full 3D system in a user defined plasma background, experiments must be performed under reduced gravity conditions. Utilizing existing (PK-4) and the proposed upcoming setup (COMPACT), operated aboard the ISS, should be considered. Due to the limitations of the already planned experiments in controlling the form of an attractive particle interaction, a *dedicated facility needs to be build and considered as follow-up experiment for operation aboard the ISS to explore the full potential of complex plasmas as model system for the study of critical point phenomena.*

Complex plasma alterations to allow the study of critical point phenomena

Since an attractive potential between the "atoms" in the substance undergoing a phase transition with critical point is mandatory, a modification to the standard screened interaction in a complex plasma experiment is necessary to allow the utilization of this unique model system to study the critical point on the quasi "atomic/molecular" level. An attractive interaction between micro-particles in a complex plasma is not new. It can be introduced by applying external fields. In the following we discuss different basic approaches that will lead initially to anisotropic, partly attractive interactions. The further proposed complex modulation of the applied fields is then expected to lead to time-averaged, quasi-isotropic, attractive interactions. While the first, anisotropic interactions might lead to some insight into critical phenomena of a new, anisotropic kind, the isotropic interaction will support studies of critical point phenomena in the classical sense with a major, important difference that, via the utilization of a complex plasma model system, phenomena can be studied experimentally down to the full 6-dimensional phase-space information for the particles.

Anisotropic, attractive forces

To add an anisotropic interaction to a complex plasma, that is otherwise governed by a screened-Coulomb interaction (Yukawa-type potentials), a static or low frequency modulated external electric or magnetic field can be added to the experiment. The well know ion focusing effect can result from this manipulation and thus be used to generate the required attractive interaction. The ion focusing effect is based on an electrical manipulation of the plasma, where a current is driven through the system. The imposed ion flow is deflected in the vicinity of the highly charged particles and can (in situations where the ion neutral mean free path is larger than the screening length) lead to positive space charges in the wake of each micro-particle in the direction downstream of the ion flow. Particles are attracted as a result by the positive space charge regions behind other particles. In case of a forced alternating current (ac) through the system *(polarity switching of the applied electric field)* at frequencies clearly below the ion plasma frequency but above the typical dynamic frequency of the dust particles, the asymmetric wake field leads to a time-averaged symmetric attractive interaction between micro-particles. The frequency of modulation has to be much higher than the dust plasma frequency so that the micro-particles are not directly affected by the varying, externally imposed electric field, but only via the established, time-averaged ion wake-fields. Using this manipulation techniques leads to the formation of string-like configurations of micro-particles known as electrorheological plasma as for example observed in the International Space Station (ISS) based complex plasma experiments PK-3 Plus (2006 – 2013) and PK-4 (since 2014) [4-6].

An alternative manipulation technique can be realized via imposed weak magnetic fields on the order of 10 Gauss in combination with the utilization of ferro- or superparamagnetic particles as complex plasma component. Due to the magnetic field the particles will develop a magnetic dipole moment that contributes to the total dust-dust particle interaction. This form of interaction adds an attractive component to the Yukawa interaction between particles that are aligned within a limiting angular constraint with respect to the magnetic field direction. However, the interaction will be more repulsive across the magnetic field direction. The study of magnetic dipole systems as an alternative approach to establish a particle interaction will not further be elaborated here as these systems are subject to a different white paper “Superparamagnetic Dusty Plasma Experiments in Microgravity”.

In contrast to an intermolecular potential, the described complex plasma interactions are anisotropic. To the best of our knowledge, the existence of a critical point in the presence of an anisotropic inter-particle interaction has not been studied. Electrorheological plasmas in future microgravity experiments could enable the search for a critical point from a string-like to an isotropic system. In PK-4, so far, only a supercritical transition (cross-over) has been found [7].

Natural isotropic and user controlled quasi-isotropic forces

No experimental observation of isotropic, attractive forces between micro-particles in complex plasmas has so far been reported. However, multiple theoretical works [8,10,11] have predicted that, both natural as well as user modulation-based effects, can lead to isotropic attraction in complex plasma. The i*on shadowing effects* is the most known source for a natural occurring attractive force [11]. Isotropically incoming ion fluxes to the surface of the individual particles are shadowed in specific directions by the neighboring particles leading to an effective attraction. To represent a relevant contribution to the interaction, it is mandatory that the ion mean free path is significantly larger than the inter-particle spacing. This requires neutral gas pressures below 10 Pa for typical experiment scenarios [9]. Ongoing and former microgravity based (ISS) experiment facilities such as PKE-Nefedov, PK-3 Plus and the ongoing PK-4 do not support the operation at these conditions. The newly proposed COMPACT facility for the ISS is specifically designed to be able to operate at this low-pressure regime [9] and thus might allow to use this natural effect to study critical point phenomena.

Beside the natural occurrence of isotropic particle attraction in a complex plasma, dust-dust attraction can also be realized by directional-temporal modulation of the aforementioned anisotropic attractive forces such as the ion-wake field and the magnetic dipole force. The initially directional manipulation has to be modulated at a sufficiently high rate over all possible directions. Due to the slow response times of the micro-particle system (order of 10 - 100 ms), the micro-particles can only react to the time-averaged force field. Depending on the exact, user defined modulation pattern, the resulting interaction can be either isotropic or user-defined anisotropic. Kompaneets et al. [10] predicted especially that the modulation of the ion wake field effect can lead to a time averaged interaction that shows similarities to the Lennard-Jones potentials. We can expect that the modulation of a weak magnetic field and its impact on ferro- or superparamagnetic particles will show a similar behavior.

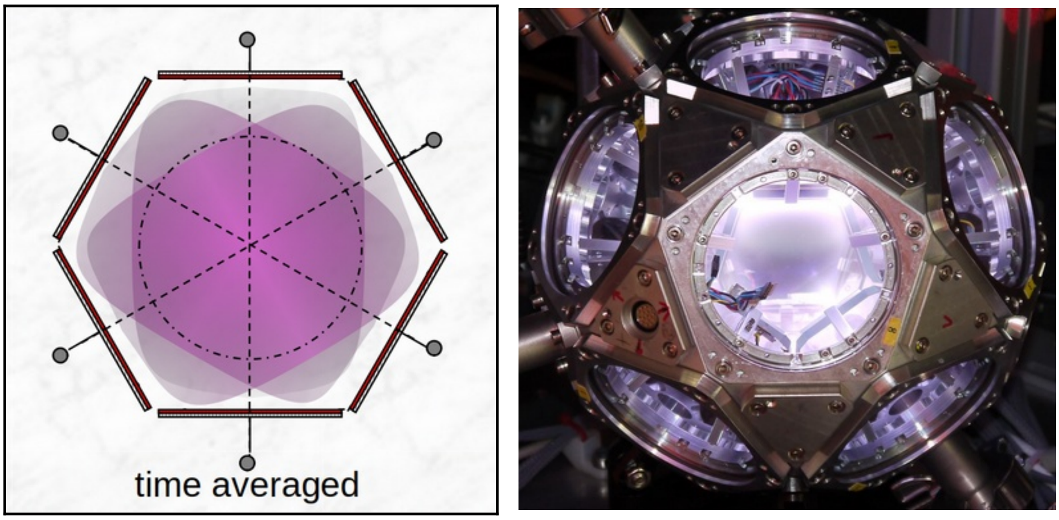

Figure 1: Left: The sketch illustrates a plasma system consisting of six individual electrodes. The plasma can be generated by applying rf/dc voltages on either or multiple electrodes at the same time. A modulation of the driving pattern allows to manipulate the overall plasma homogeneity and isotropy. Right: The first realization of the sketched multi electrode system concept, however utilizing twelve electrodes arranged in a dodecahedron geometry. This chamber was designed with the intend to support critical point phenomena investigations with complex plasmas.

Review of experimental setups “suitable” for complex plasma critical point studies

To study complex plasma that support attractive particle interaction, a wide range of dedicated techniques and experimental vessels have been developed in recent years. Some techniques, such as the magnetic field induced manipulation of ferromagnetic or superparamagnetic particles, are still in their early design phase. On the contrary, techniques to modulate the rf system, amplitude and phase, and the ac/dc manipulation of currents through the discharge, are well developed to an extend that has allowed breadboard setups of different complexity to be build (see Figure 1). A low complexity, test setup utilizing 6 electrodes (cubic electrode geometry enclosing a spherical glass chamber) has been tested already during parabolic flights about a decade ago. The latter experiments could demonstrate the general capability to shape a time-average force fields by high frequency plasma modulations. The observed dust cloud in the parabolic flight experiments did not show the typical imposed cylindrical geometry of a parallel plate discharge, but instead resembled the spherical geometry of the vacuum vessel. Unfortunately, the setup did not support the ac/dc current mode at that time and thus could not be used to study the particle attraction based on imposed ion currents. More recently, a twelve-electrode breadboard chamber has been built. It is specifically designed for critical point studies. This “dodecahedron” chamber supports twelve individual electrodes, each of which is connected to its own rf/ac/dc generator. The plasma generation is therefore very flexible, ranging from driving a single electrode to multiple electrodes at the same time in a fully user-defined fashion. For example, driving two opposing electrodes establishes the typical cylindrical discharge configuration. If the discharge mode (direction of the cylindrical discharge mode) is changing at a high rate, a quasi-isotropic plasma can be sustained. Similarly, driving ac fields under a user-defined modulation is expected to allow the particle interaction to be altered to create a time-averaged, isotropic attractive interaction. The breadboard system has not yet been fully built and was never operated under microgravity condition. However, since it is especially, since designed for the critical point studies, it is presently the most advanced experimental setup for studying critical points with complex plasmas.

As a shorter-term alternative, initial experiments might be performed with the COMPLEX PLASMA FACILITY (COMPACT). COMPACT, the proposed next generation ISS based complex plasma space experiment is based on an advanced parallel plate discharge vessel, similar to the so-called Zyflex chamber [12] (COMPACT breadboard chamber). COMPACT is expected to support two electrode systems that each consist of a ring and a center electrode. With this chamber a wide range of electric (rf, ac, dc) manipulations will be possible allowing the generation of user-manipulated attractive forces between the particles. Due to the already fixed, cylindrical vessel geometry and the parallel plate discharge mode, the introduced, current driven attractive forces will have a preferred direction though. The natural expected isotropic ion shadow force might be utilized with this experiment at low discharge pressures. With some addition to the setup such as a triple Helmholtz coil pair to support magnetic particle dipole interaction, another crucial manipulation technique could be implemented. (The latter is unlikely at this point of development as COMPACT reached already a mature state, but is a viable option for a future microgravity experiment).

The need for long duration microgravity and a potentially dedicated facility

Beside utilizing the planned and ongoing experiment setups aboard the ISS (PK-4 and probably COMPACT) to perform initial studies on critical point phenomena based on potential anisotropic attractive interactions, it is necessary to consider the development of a dedicated, space based (ISS) facility to study critical point related topics to their full extend. This facility could be based on the breadboard setup – the twelve-electrode discharge chamber – shown in figure 1 and described in more detail in the previous section. It would be a follow up experiment of the COMPACT facility aboard the ISS. *It has to be stressed here that complex plasma studies of three-dimensional systems, especially, if the homogeneity and isotropy of the system matters, can practically only be performed under reduced gravitational influence.* For systems that contain millions of particles, such as those required to study critical point phenomena, also microgravity durations on order of minutes or longer have to be realized. Short duration microgravity periods only allow to explore the dust cloud *distribution* in the vacuum vessel. Recent experiments on ground using the COMPACT breadboard setup indicate that, for example, the crystallization of a micro-particle cloud in this specific setup can take several minutes [12]. The relaxation time for larger systems would scale accordingly. Therefore, places, where such experiments can be performed are ultimately restricted to either the ISS, sounding rockets or suborbital flights. In case crew intervention might be necessary, free-flying satellite mission would be excluded, but could be considered if the experiment could be automatized sufficiently and enriched with an artifical intelligence (AI) based pre-analysis system that allows pre-selection of data depending on its content before sending it to ground. Otherwise, satellite missions have the disadvantage of limited data transfer capabilities (complex plasma experiments with state-of-the-art diagnostic system will literally produce multiple terabyte of data for an experiment campaign).

References:

[1] R. Marcout et al., *DECLIC: A facility to investigate fluids and transparent materials in microgravity conditions in ISS*, https://doi.org/10.2514/6.IAC-06-A2.5.02

[2] A. Ivlev, H. Löwen, G. Morfill and C.P. Royall, *Complex Plasmas and Colloidal Dispersions: Particle-Resolved Studies of Classical Liquids and Solids*, World Scientific Publishing Company (2012)

[3] H. M. Thomas and G. E. Morfill, *Melting dynamics of a plasma crystal*, Nature **379**, pages 806–809 (1996). https://doi.org/10.1038/379806a0

[4] A.V. Ivlev et al., *First Observation of Electrorheological Plasmas*, Phys. Rev. Lett. 100, 095003 (2008). https://doi.org/10.1103/physrevlett.100.095003

[5] A. V. Ivlev, M. H. Thoma, C. Räth, G. Joyce, and G. E. Morfill, *Complex Plasmas in External Fields: The Role of Non-Hamiltonian Interactions*, Phys. Rev. Lett. 106, 155001 (2011). https://doi.org/10.1103/PhysRevLett.106.155001

[6] M. Pustylnik, B. Klumov, M. Rubin-Zuzic, A. Lipaev, V. Nosenko, D. Erdle, A. Usachev, A. Zobnin, V. Molotkov, G. Joyce, H.M. Thomas, M. Thoma, O. Petrov, V. Fortov, O. Kononenko, *Three-dimensional structure of a string-fluid complex plasma*, Physical Review Research 2, 033314 (2020). https://dx.doi.org/10.1103/PhysRevResearch.2.033314

[7] C. Dietz, J. Budak, T. Kamprich, M. Kretschmer, M. H. Thoma, *Phase transition in electrorheological plasmas*, Contrib. Plasma Phys. (2021), https://doi.org/10.1002/ctpp.202100079

[8] S. A. Khrapak et al., *Critical Point in Complex Plasmas*, Phys. Rev. Lett. 96, 015001 (2006). https://doi.org/10.1103/PhysRevLett.96.015001

[9] COMPACT, Science Envelope Requirements Document, in preparation.

[10] Kompaneets R., Morfill, G.E. and A.V. Ivlev, *Design of new binary interaction classes in complex plasmas*, Physics of Plasmas, **16**, 043705 (2009). https://doi.org/10.1063/1.3112703

[11] Morfill, G.E., Tsytovich V.N. and H. Thomas, *Complex Plasmas: II Elementary processes in complex plasmas*, Plas. Phys. Rep. **29**, 1-30, 2003. https://doi.org/10.1134/1.1538499

[12] Knapek, C.A., Konopka, U., Mohr, D.P., Huber, P., Lipaev, A.M. and H.M. Thomas, *„Zyflex“: Next generation plasma chamber for complex plasma research in space*, Rev.Sci.Instrum., **92**, 103505, 2021. https://doi.org/10.1063/5.0062165