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Recent Progress in Complex Plasmas

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An overview on recent developments in the field of complex plasmas is presented. The focus is directed on the activities of research groups participating in the Collaborative Research Centre 24 "Fundamentals of Complex Plasmas" at the Universities in Greifswald and Kiel as well as at the Leibniz-Institute for Plasma Research and Technology. This article summarizes results on strongly correlated dusty plasmas, reactive as well as electronegative plasmas and plasma-surface interaction, and gives an introduction to the following articles of this special issue. Important developments are highlighted and future research directions are outlined.

1 Introduction

This special issue "Progress in Complex Plasmas" presents highlighted papers arising from the recent research work in the Transregional Collaborative Research Centre 24 "Fundamentals of Complex Plasmas" (TRR24). The TRR24 is government-funded by the German Research Foundation (DFG) since 2005 and stands currently in the second funding period 2009–2013. In particular, the Ernst-Moritz-Arndt-Universität Greifswald, the Christian-Albrechts-Universität zu Kiel and the Leibniz-Institut für Plasmaforschung und Technologie Greifswald contribute with 14 scientific projects to the collaborative research on key topics in the field of complex plasmas.

A complex plasma is — in our terms — a multi-component plasma containing, besides electrons, positive ions and neutral atoms/molecules, additional components such as negative ions, charged nano- or micrometer-sized particles, metastables and reactive atoms or molecules strongly interacting with solid surfaces, see Fig. 1.

The importance of complex plasma research derives from the growing relevance of plasma physics in general. Plasma science has been identified as a key field of basic and applied research with proven impact on society and economy. This applies to energy research, ranging from controlled nuclear fusion to solar cells, to lighting and display technology, as well as to the field of nanotechnology including plasma-assisted deposition or etching.

The 2007 review on plasma research and its perspectives in the next decade [1] by the National Research Council, USA, had already stated both the necessity of further progress in the field of plasma science and identified essential fields of importance for fundamental research and applications. The identified plasma processes of high interest cover an extremely wide range of temperatures, densities and magnetic fields including relativistic, classical and highly correlated plasmas. Two of the hot topics defined in the above US report, namely "Multiphase Plasma Dynamics" and "Correlations in Plasmas", are key questions addressed by the research activities of the TRR24. Furthermore, in the recent review article "The 2012 Plasma Roadmap" [2] 16 key topics for plasma research are addressed. Among them, the importance of plasma diagnostics, of accurate atomic and molecular data for plasma physics, and of plasma modelling as a cross-road is stressed. Moreover, hot topics are highlighted

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concerning microplasmas and the field of plasma applications, e.g., plasma surface treatment for nano-scaled devices and deposition of functional films, nanoparticle formation in reactive plasmas, plasma catalysis, plasma medicine and plasma thrusters. Many of theses topics are part of the research of the TRR24.

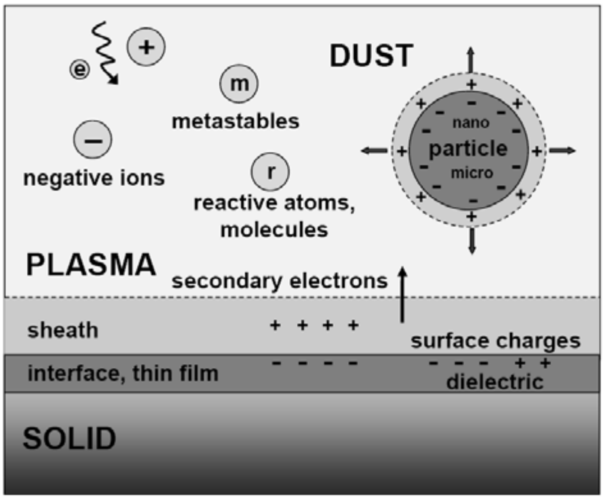

Fig. 1 Complex plasmas are characterized by additional plasma components such as negative ions, charged nano- or micrometer-sized particles, as well as metastables and reactive species interacting with condensed matter surfaces.

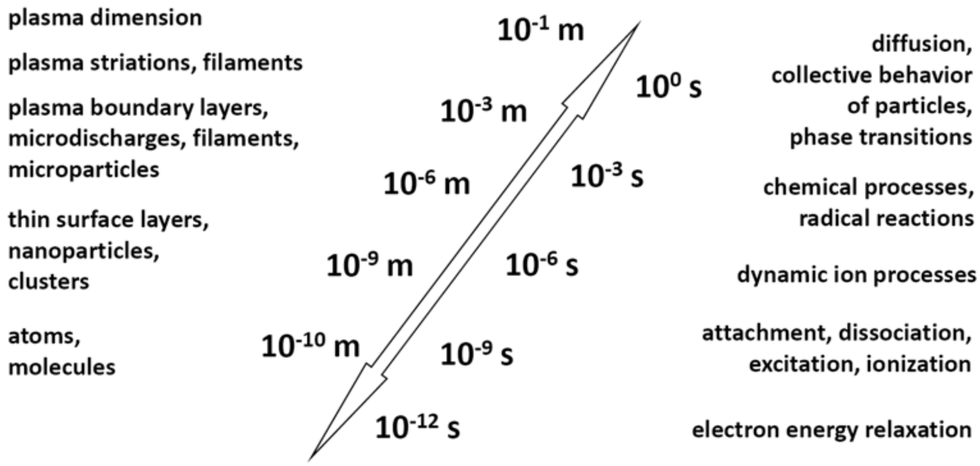

Fig. 2 Sketch of the huge range of space and time scales and the related physical and chemical processes in complex plasmas.

Complex plasmas span a huge range of parameters as shown, for the example of space and time scales, in Fig. 2. These scales involve enormously diverse and complicated physical and chemical processes which, nevertheless, are governed by common fundamental physical principles. Both, modern diagnostic techniques and advanced computer modelling have contributed to rapidly advancing knowledge of complex plasmas over the last years. The contributions of the TRR24 in the field of complex plasmas cover the following three fundamental areas of high interest in plasma science which were pursued by means of the joint research effort of the involved cooperating groups:

(i) Correlations and coupling effects in two-dimensional (2D) and 3D particle ensembles in plasma confinement, their ordering and dynamic behaviour in the presence of wake fields, shielding and magnetic fields.

(ii) Dynamics of multi-component plasmas and plasma-surface interaction involving the plasma sheath dynamics, the build-up of surface charges at internal and external boundaries, the emission of secondary species, and the influence of negative ions and metastables on discharge operation modes.

(iii) Reactive processes in molecular plasmas and at surfaces leading to synthesis of nano-sized particles, surface modification and deposition of functional thin films.

The articles in this special issue are arranged according to these three topics. In the following sections we discuss the progress in these three areas and conclude with an outlook in Sec. 5.

2 Strong coupling effects in complex plasmas

The first topical part is devoted to recent investigations of the TRR24 in the field of strongly coupled charged particle systems where the mutual electrostatic interaction among the particles by far exceeds their thermal energy. Particle-containing, dusty plasmas are a paradigm for the study of strongly coupled systems. The first paper by Melzer et al. [3] reviews different experimental techniques to drive finite systems of dust particles from ordered, solid-like arrangements to liquid states. These techniques include melting due to shear forces, instabilities or equilibrium laser-heating. The melting studies are complemented by dedicated theories and simulations to characterize these phase transitions.

Besides the direct particle-particle interaction, the highly charged dust particles necessarily alter the plasma environment. A striking example is the formation of ion wake fields in the presence of an ion flow that even leads to net attractive interactions between two dust particles. The ion wake effects are summarized in the second paper by Block et al. [4] where the wake formation is studied both, in linear response theory and by particle in cell (PIC) simulations. Experimentally the wake field strength is derived from precision charge measurements.

Magnetic fields add a new feature to the investigation of dusty plasma. As a specific example, in the paper by Reichstein et al. [5], a dust torus formed in the anodic plasma with magnetic fields is studied. There the toroidal dust flow is calculated in Langevin Molecular Dynamics simulations and compared to experimental results. The un-sheared and incompressible dust flow shows a nested shell structure due to the interplay of Coulomb interaction and friction. At reduced friction, Kelvin-Helmholtz-type instabilities arise.

A different aspect of strongly-coupled systems is realized in bilayers of spatially separated positive and negative charges. The paper by Schleede et al. [6] considers theoretically the example of electron-hole bilayers that add quantum correlation effects to the Coulomb coupling. Using path integral Monte Carlo simulations quantum and Coulomb correlation effects are treated from first principles, revealing a rich phase behaviour, including a gas of bound pairs (excitons), an exciton crystal, an electron-hole liquid and a hole crystal.

These investigations have addressed key questions in the field of strongly coupled systems in experiment, theory and simulations. Future investigations will open up novel research directions such as magnetic field effects in dusty plasmas, in particular the modification of interparticle forces by strong magnetic fields. Because of the large particle mass the magnetization of micrometer-sized dust in experiments is practically impossible, but the formal equivalence of the Coriolis force in rotating dusty plasmas with the Lorentz force opens exciting new prospects for dusty plasma research in the future. Further, the modification of dust-plasma interactions and the transition from finite systems to bulk behaviour in strongly-coupled dust systems will be studied. Wake fields and their role for structure and dynamics add a novel and important line of research.

3 Dynamics of plasmas and plasma sheaths. Plasma-surface interaction

The second topical part provides recent results on plasma sheath and plasma-surface interaction, especially about the investigation of a single microparticle trapped in the rf plasma sheath, the operation modes of electronegative low pressure radio-frequency (rf) plasmas and atmospheric pressure barrier discharges, as well as the microphysical description of surface charging and secondary species emission. In the first article, Schubert et al. [7] study the behaviour of microparticles trapped in the curved plasma sheath of a complex electrode geometry by experiment and PIC-MCC simulations. In particular, an additional cylinder was placed vertically in the center on the planar rf powered electrode. Thereby, the equilibrium position of a single particle embedded in the rf sheath was probed in dependence on the particle diameter and plasma processing parameters for different materials and surface potentials of the cylinder in order to achieve information about the forces on the particle, the sheath potential and electric field.

The following article by Küllig et al. [8] concerns the dynamics and electronegativity of a low-pressure capacitively coupled oxygen rf plasma investigated by experiments and simulations. Two operation modes of the oxygen rf plasma at low and high electronegativity, mainly controlled by the rf power, have been found by detailed analysis of the electron and negative ion density as well as the rf phase resolved optical emission intensity.

In particular, the important role of the metastable oxygen molecules in attachment and detachment reactions as well as the different electron heating mechanisms in the rf sheath have been pointed out. Further, the collisional detachment of negative ions in the sheath reveals the generation of pseudo-secondary electrons. Fluctuations in the plasma parameters due to the attachment induced ionization instability are discussed.

The operation modes and the structure formation (strongly localized discharge spots) of atmospheric-pressure barrier discharges in nitrogen and helium have been studied within the same discharge cell configuration by Bogaczyk et al. [9]. In particular, the absolute surface charge and the metastable N_2(A) density were measured together with the evaluation of the spatio-temporally resolved discharge development using the plasma-induced optical emission intensity at identical and well-defined experimental conditions. The different operation modes, namely the filamentary and the diffuse Townsend- or glow-like mode of the barrier discharge are mainly controlled by the gas composition, the cell geometry, and the signal shape of the applied voltage.

The final article in this topical part concerns the microscopic description of the charge transfer across a plasma wall, which is of great importance to the charging of floating walls, embedded micro-particles in a plasma environment and the generation of secondary electrons. With the ultimate goal of a microscopic description of grain charging and the build-up of surface charges in dielectric barrier discharges, Bronold et al. [10] investigated — as a first step — electron trapping and desorption at uncharged metallic and dielectric surfaces. In addition they studied the distribution of the quasi-stationary wall charge across the plasma-wall interface and how effciently electrons can be extracted from a dielectric surface by de-excitation of metastable molecules.

The contributions in this topical part have addressed key topics in the investigation of complex plasmas such as the particle trapping in shaped rf sheaths as well as different discharge operation modes and the temporal development of transient discharges influenced by surface charges, metastables, negative ions and secondary electrons. Novel lines in the further study of electronegative low-pressure rf plasmas are the impact of negative ions on electron heating mechanisms (e.g. bulk heating mode in strongly electronegative plasmas), the mode switching between the low and high electronegativity as well as the CCP and ICP operation, and the onset of plasma instabilities. In atmospheric pressure barrier discharges, the novel perspectives concern the influence of secondary species generation at dielectric surfaces, the role of helium metastables and the oxygen admixture to nitrogen or helium on the discharge development and the appearance of filamented or diffuse discharge modes. Progress will be achieved by close cooperation between experiments, microscopic description and molecular dynamics simulations of surface processes as well as kinetic and fluid model discharge simulations.

4 Reactive plasmas, synthesis of functional thin films and nanoparticles

The articles in the third topical part are focused on reactive processes in plasmas and at surfaces including the formation of nano-sized clusters and particles. The first contribution concerns the plasma chemical study of a low pressure rf discharge in a mixture of Ar or Ar/N_2 with the metal-organic precursor aluminum tri-isopropoxide by Lopatik et al. [11]. This precursor is applied in plasma-enhanced chemical vapor deposition for deposition of metal oxide thin films (Al_xO_y). In particular, the absolute concentrations of 6 stable gaseous reaction products have been measured in dependence on the precursor admixture for different rf power and total pressure by infrared absorption spectroscopy using an external-cavity quantum-cascade laser arrangement. The product concentrations of different hydrocarbons and H_2O molecules and, at nitrogen admixture, the additional formation of HCN and HNO_3 provide first information on the fragmentation behaviour of aluminum tri-isopropoxide and the hydrocarbon plasma chemistry in the rf discharge.

The following article by Schäfer et al. [12] is focused on the deposition of thin SiO_x films on flat polymer and glass samples using different organo-silicon precursors and an rf-excited plasmajet operating at atmospheric pressure. The plasma processing parameters, the discharge operation condition and the deposition rate were evaluated concerning the O/Si ratio of the deposited films. In particular, the influence of the gas flow regime was taken into account and discussed in relation to results of a two-dimensional axis-symmetric fluid model. For laminar flow regimes the O/Si ratio close to two and a low carbon content were found. The calculated flux of precursor fragments onto the substrate surface agrees qualitatively with measured profiles of the film thickness.

By use of a nano-cluster source, consisting of a planar DC magnetron discharge plasma in a high- pressure gas aggregation chamber, the formation and deposition of nano-sized silver clusters have been studied by Ganeva et al. [13]. Thereby, the mass spectrometric analysis of silver clusters in the gas phase (cluster size below 10 nm) was compared with the deposited silver nano-clusters by transmission electron microscopy (TEM). The effect of the helium to argon ratio in the aggregation chamber, the gas flow rate and the magnetron power on the cluster size distribution was evaluated. The comparison of the results about the cluster size analysis from the gas phase and the surface showed that the source predominantly produces clusters out of range of the mass spectrometer.

The last article in this topical part by Bonitz et al. [14] concerns the simulation of nano-composite formation in a magnetron discharge plasma. Besides an overview on the particle-based simulation in nano-composite formation and the discussion of the kinetic Monte Carlo simulation of the metal cluster growth, first simulation results are presented for the co-sputtering of metallic and polymer material. Furthermore, results for the time-resolved coalescence of two clusters, consisting of 565 atoms each are presented using molecular dynamics simulations.

The planned experimental investigation on complex reactive plasmas containing organic and metal organic precursors, respectively, or related sputtered species for the formation and growth of nanoparticles and (composite) thin films will be extended to the analysis of intermediate and transient species in the gas phase as well as the detailed evaluation of surface processes. Thereby, the important reaction channels in the gas phase and at the surface have to be addressed. The combination of experiments, theory and integrated simulations of processes in the plasma and at surfaces are the key for further progress in the plasma-based synthesis of nano-sized particles as well as of novel functional materials.

5 Conclusions and outlook

Complex plasmas are a genuine interdisciplinary research field at the interface between low-temperature plasma physics, atomic and molecular physics, condensed matter and surface physics as well as many-particle and quantum physics. The present introductory article has given a brief overview on recent developments in this field, with the focus on research activities in the Collaborative Research Centre 24 "Fundamentals of Complex Plasmas". We conclude by outlining the continuation of this research in the near future in the frame of the three overlapping fields correlations, dynamics and reactivity & surfaces, see Fig. 3.

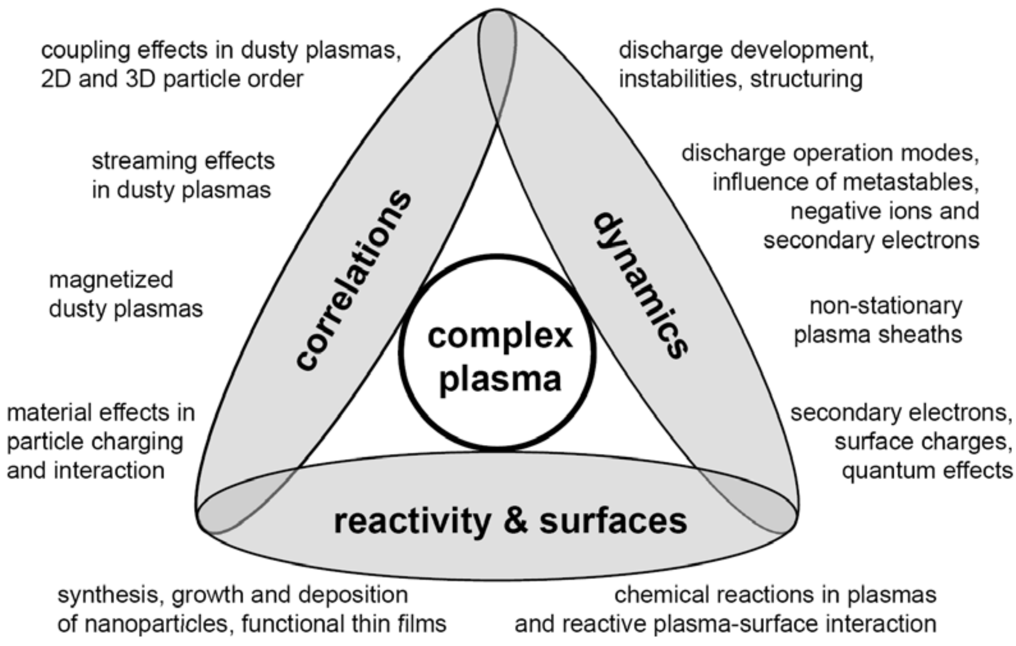

Fig. 3 Key areas and research topics of the collaborative research centre 24 "Fundamentals of Complex Plasmas".

For a future comprehensive study of complex plasmas, the following new questions have come into the focus: (i) correlations in 2D and 3D particle ensembles with nonequilibrium plasma effects such as streaming ions or neutrals, (ii) the dynamics of magnetized strongly coupled dusty plasmas, (iii) the specific impact of metastables, negative ions, surface charges and secondary electrons on the development of transient discharges and the different operation modes of discharges at atmospheric and low pressure, (iv) the microscopic description of the charge transfer across dielectric plasma walls, (v) the reaction kinetics of transient molecules and intermediate products in the bulk and at surfaces of discharges containing organic or metal-organic precursors for the deposition of functional films, and (vi) the generation and deposition of nanoparticles or composite films due to material sputtering or the nucleation and growth processes of nanoparticles in hydrocarbon plasmas.

Thereby, high-end experimental diagnostics, theoretical modelling, and advanced computer simulations will be applied in a combined manner to achieve fundamental knowledge on plasma and surface processes. In particular, the detailed knowledge about the particle-particle, particle-plasma and particle-surface interaction and the interplay of elementary surface and volume processes in nonthermal molecular plasmas will provide substantial contributions to the plasma science in general. Furthermore, the results of the fundamental research on complex plasmas in the TRR24 will have a significant impact on plasma technologies such as the design of tailored plasma sources and plasma process control, and the plasma applications in nano and materials science, environmental science, biology and medicine.

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