# **NKFIH K-115805**

"Complex Plasmas in Action"



final report 2015. Dec. 01. — 2019. Nov. 30.

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# **Complex plasmas in Action**

final report

# GENERAL REMARKS

In our original research proposal we have taken on the challenge to conduct complex plasma research aiming to advance this field in three thematic groups by means of experiments and numerical simulations:

STRONG COUPLING PHENOMENA: using the dust component to model collective, strong coupling effects;

DUST IN PLASMA: interaction between gas discharge and dust grains, using dust as discharge probes, controlled transport of dust in discharges;

NANO-PARTICLE SYNTHESIS: fundamental plasma processes during synthesis of nanometersized dust in reactive plasmas for optimization of deposition processes for future applications.

During the supported research period all of the three listed topics were addressed, although not with equal intensities, resulting in scientific achievements disseminated in the form of conference presentations and peer-reviewed journal articles. In relation to these topics we've found it important to place some more emphasis on the understanding of the plasma environment and therefore extended towards the FUNDAMENTALS OF RADIO-FREQUENCY DISCHARGES operating at atmospheric conditions, molecular environment, and elementary charged particle transport to the topics.

In the following sections I will introduce the details of the scientific achievements grouped according to the topical list above and based on the journal articles that were published during the supported period. Some of the recent studies are still in the pre-publication phase, those will be listed in a separate section called "Ongoing Research"

# STRONG COUPLING PHENOMENA

The influence of an external homogeneous magnetic field on the quasilocalization of the particles (characterized quantitatively by cage correlation functions) in strongly coupled three-dimensional Yukawa systems was investigated via molecular dynamics computer simulations over a wide domain of the system parameters. The caging time was found to be enhanced by the magnetic field B. The anisotropic migration of the particles in the presence of magnetic field was quantified via computing directional correlation functions, which indicate a more significant increase of localization in the direction perpendicular to B, while a moderate increase is also found along the B field lines. Associating the particles' escapes from the cages with jumps of a characteristic length, a connection was found with the diffusion process: the diffusion coefficients derived from the decay time of the directional correlation functions in both the directions perpendicular to and parallel with B are in very good agreement with respective diffusion coefficients values obtained from their usual computation based on the mean-squared displacement of the particles. [Dzhumagulova K N, Masheyeva R U, Ott T, Hartmann P, Ramazanov T S, Bonitz M, Donkó Z; "Cage correlation and diffusion in strongly coupled three-dimensional Yukawa systems in magnetic fields"; Physical Review E. 93, 063209 (2016)]

In a two-dimensional dusty plasma composed of superparamagnetic, charged dust grains and immersed in an external magnetic field B, the grains interact via both Yukawa and magnetic dipoledipole potentials. Because the grains' magnetic dipole moments are induced by B, the dipole moments all lie along B. When B is tilted with respect to the normal to the dust layer, the interaction between the grains becomes anisotropic. We have considered a realistic quasi-2D system where the grains are confined by an external potential and can undergo small oscillations perpendicular to the layer. We have analyzed the effect of the strength of the confining potential on the in-plane correlations and on the wave propagation. In addition to the in-plane compressional and transverse waves, there now appears an out-of-plane transverse wave generated by the oscillation of the grains in the confining potential. The theoretical approach uses the Quasi-Localized Charge approximation paralleled by molecular dynamics simulations. [Rosenberg M, Kalman G J, Hartmann P, Donkó Z; "Waves in a quasi-two-dimensional superparamagnetic dusty plasma liquid in a trap"; Physical Review E. 94, 033203 (2016)]

The self-diffusion phenomenon in a two-dimensional dusty plasma at extremely strong (effective) magnetic fields was studied experimentally and by means of molecular dynamics simulations. In the experiment the high magnetic field was introduced by rotating the particle cloud and observing the particle trajectories in a co-rotating frame, which allows reaching effective magnetic fields up to 3000 T. The experimental results confirm the predictions of the simulations: (i) super-diffusive behavior was found at intermediate timescales and (ii) the dependence of the self-diffusion coefficient on the magnetic field was well reproduced. [P. Hartmann, J. C. Reyes, E. G. Kostadinova, L. S. Matthews, T. W. Hyde, R. U. Masheyeva, K. N. Dzhumagulova, T. S. Ramazanov, T. Ott, H. Kählert, M. Bonitz, I. Korolov, and Z. Donkó, "Self-diffusion in two-dimensional quasimagnetized rotating dusty plasmas", Phys. Rev. E 99, 013203 (2019)]

A magnetic field was shown earlier to enhance field-parallel heat conduction in a strongly correlated plasma whereas cross-field conduction is reduced. We have shown that in such plasmas, the magnetic field has the additional effect of inhibiting the isotropization process between field-parallel and cross-field temperature components, thus leading to the emergence of strong and long-lived temperature anisotropies when the plasma is locally perturbed. An extended heat equation was derived and shown to describe this process accurately. [T. Ott, M. Bonitz, P. Hartmann, and Z. Donkó, "Spontaneous generation of temperature anisotropy in a strongly coupled magnetized plasma", Phys. Rev. E 95, 013209 (2017)]

In many-body systems the convolution approximation states that the 3-point static structure function,  $S^{(3)}(k_1,k_2)$ , can approximately be "factorized" in terms of the 2-point counterpart,  $S^{(2)}(k_1)$ . We have investigated the validity of this approximation in 3-dimensional strongly-coupled Yukawa liquids: the factorization was tested for specific arrangements of the wave vectors  $k_1$  and  $k_2$ , with molecular dynamics simulations. With the increase of the coupling parameter we've found a breakdown of factorization, of which a notable example is the appearance of negative values of  $S^{(3)}(k_1,k_2)$ , whereas the approximate factorized form was restricted to positive values. These negative values – based on the quadratic Fluctuation-Dissipation Theorem – imply that the quadratic part of the density response of the system changes sign with wave number. Our simulations that incorporate an external potential energy perturbation clearly confirm this behavior. [Magyar P, Hartmann P, Kalman G J, Golden K I, Donkó  $Z_j$ : "Factorization of 3-Point Static Structure Functions in 3D Yukawa Liquids"; Contrib. Plasma Phys. 56, 816 (2016)]

The direct experimental determination of the 3-point static structure function  $S^{(3)}(k_1,k_2,k_0)$  of a 2dimensional dusty plasma liquid was presented. The measurements were complemented by molecular dynamics simulations of the system, using parameters (dust charge, plasma frequency, coupling and screening coefficients), which are derived from the experimentally obtained 2-point static structure function  $S^{(2)}$ , as well as the dynamic structure function and current-current fluctuation spectra. The experimental results of  $S^{(3)}$  are in good agreement with those of the simulations, including the (low wavenumber) domain, where  $S^{(3)}$  acquires negative values. The "Convolution Approximation" (giving  $S^{(3)}$  in a factorized form of  $S^{(2)}$  functions) clearly breaks down in this domain; however, it was found to be a useful aid for explaining the main features of the  $S^{(3)}(k_1,k_2,k_0)$  functions, for which (experimental and simulation) maps are presented at selected values of one of its arguments. [Z. Donkó, P. Hartmann, P. Magyar, G. J. Kalman, and K. I. Golden, "Higher order structure in a complex plasma", Physics of Plasmas 24, 103701 (2017)]

Dynamic characteristics of strongly coupled classical one-component Coulomb and Yukawa plasmas were obtained within the non-perturbative model-free moment approach without any data input from simulations so that the dynamic structure factor (DSF) satisfies the first three non-vanishing sum rules automatically. The DSF, dispersion, decay, sound speed, and other characteristics of the collective modes are determined using exclusively the static structure factor calculated from various theoretical approaches including the hypernetted chain approximation. A good quantitative agreement with molecular dynamics simulation data was achieved. [Yu. V. Arkhipov, A. Askaruly, A. E. Davletov, D. Yu. Dubovtsev, Z. Donkó, P. Hartmann, I. Korolov, L. Conde, and I. M. Tkachenko, "Direct Determination of Dynamic Properties of Coulomb and Yukawa Classical One-Component Plasmas", Phys. Rev. Lett. 119, 045001 (2017)]

Yukawa one-component plasmas (YOCP) are completely characterized by two parameters: the screening parameter,  $\kappa$ , and the nominal coupling strength,  $\Gamma$ . It is well known that the collective spectrum of the YOCP is governed by a longitudinal acoustic mode, both in the weakly and strongly coupled regimes. In the long-wavelength limit, the linear term in the dispersion (i.e.,  $\omega = sk$ ) defines the sound speed s. We have studied the evolution of this latter quantity from the weak through the strong-coupling regimes by analyzing the dynamic structure function  $S(k, \omega)$  in the low-frequency domain. Depending on the values of and  $\kappa$  and  $w = s/v_{th}$  (i.e., the ratio between the phase velocity of the wave and the thermal speed of the particles), we have identified five domains in the  $(\kappa, \Gamma)$ parameter space in which the physical behavior of the YOCP exhibits different features. The competing physical processes are the collective Coulomb like versus binary collision dominated behavior and the individual particle motion versus quasilocalization. Our principal tool of investigation was molecular dynamics (MD) computer simulation from which we obtain  $S(k, \omega)$ . Recent improvements in the simulation technique have allowed us to obtain a large body of highquality data in the range  $\Gamma = \{0.1-10\ 000\}$  and  $\kappa = \{0.5-5\}$ . The theoretical results based on various models were compared in order to see which one provides the most cogent physical description and the best agreement with MD data in the different domains. [Silvestri L G, Kalman G J, Donkó Z, Hartmann P, Rosenberg M, Golden K I, Kyrkos S, "Sound speed in Yukawa one-component plasmas across coupling regimes", Phys. Rev. E 100, 063206 (2019)]

# DUST IN PLASMA

We have developed a very simple and sensitive method to measure the sputtering rate of solid materials in stationary low-pressure gas discharges. The method is based on the balance of the centrifugal force and the confinement electric force acting on a single electrically charged dust particle in a rotating environment. We have demonstrated the use and sensitivity of this method in a capacitively coupled radio frequency argon discharge. We were able to detect a reduction of 10 nm in the diameter of a single dust particle. [P. Hartmann, J. C. Reyes, I. Korolov, L. S. Matthews, and T. W. Hyde, "Simple experiment on the sputtering rate of solids in gas discharges", Physics of Plasmas 24, 060701 (2017)]

### NANAO-PARTICLE AND THIN FILM SYNTHESIS

Nanocrystalline diamond films were grown from a CH4/H2 gas mixture by the MWCVD technique at 1200 W microwave power and a frequency of 2.45 GHz. Silicon wafers served as substrate material; bias enhanced nucleation on the substrate surface was performed at 4% methane and 200 V bias voltage for 30 min to form diamond nuclei. The chamber pressure was kept at 40 mbar for nucleation and growth likewise. The total gas flow rate in each case was 100 sccm. The deposition of SiV containing nanocrystalline diamond film was performed at methane concentrations of 0.2%, 0.5%, 1.0%, 1.5%, 2% and 3% of the feed gas, while the substrate temperature was kept at 650, 700, 750, 800 and 850 °C. The growth process was performed for 2 h for each sample. The layer thickness was determined by in situ laser reflection interferometry; it ranges around 200 nm. It was found that the deposition parameters have a significant effect on the structure of ND films containing SiV centers mainly in form of residual stress of different magnitude. Experimental results show that an increase of residual compressive stress causes a decrease of line broadening and a blue shift of the SiV ZPL. So the control of the residual stress by appropriate growth conditions allows to tailor the ZPL parameters of the SiV centers. [Himics, M. Veres, S. Tóth, I. Rigó, and M. Koós, Experimental Study of Spectral Parameters of Silicon-Vacancy Centers in MWCVD Nanodiamond Films Important for Sensing Applications, In: Petkov P., Tsiulyanu D., Popov C., Kulisch W. (eds) Advanced Nanotechnologies for Detection and Defence against CBRN Agents. NATO Science for Peace and Security Series B: Physics and Biophysics. Springer, Dordrecht]

The spectral line shape of the 1.68 eV (738 nm) emission line – usually associated with the negatively charged silicon-vacancy (SiV) center in diamond and promoted as a strong candidate for many quantum technology and nanobiology related applications - was studied by luminescence spectroscopy in different nanocrystalline diamond films prepared by MW CVD technique. An asymmetric line profile, expressed as a long tail on the lowenergy side of the narrow luminescence band located around 1.68 eV, have been observed in all samples. By applying a multi-wavelength laser excitation, it was found that the asymmetric shape of the 1.68 eV emission line varies by changing the excitation energy and becomes more pronounced under red (1.95 eV) excitation, especially for nanodiamond film characterized by larger average grain size. Based on the observed excitation dependent line shape behavior and on the analysis of the fine-structured emission lines, recorded by photoluminescence mapping experiments, the asymmetry of the ZPL was assigned to another optically active defect, simultaneously presented in diamond nanocrystals, namely to the GR1 center. The common presence of the GR1 and SiV centers within the CVD diamond structures even in the case of high-quality microcrystals was supported by photoluminescence excitation measurements also. [L. Himics, M. Veres, S. Tóth, I. Rigó, M. Koós, "Origin of the asymmetric zero-phonon line shape of the siliconvacancy center in nanocrystalline diamond films", Journal of Luminescence 215 (2019) 116681]

### FUNDAMENTALS OF RADIO-FREQUENCY DISCHARGES

Low pressure capacitive radio frequency (RF) plasmas are often described by equivalent circuit models based on continuum approaches that predict the self-excitation of resonances, e.g., high frequency oscillations of the total current in asymmetric discharges, but do not provide a kinetic interpretation of these effects. In fact, they leave important questions open: How is current continuity ensured in the presence of energetic electron beams generated by the expanding sheaths that lead to a local enhancement of the conduction current propagating through the bulk? How do the beam electrons interact with cold bulk electrons? What is the kinetic origin of resonance phenomena? Based on kinetic simulations, we have found that the energetic beam electrons interact with cold bulk electrons (modulated on a timescale of the inverse local electron plasma frequency) via a time dependent electric field outside the sheaths. This electric field is caused by the electron beam itself, which leaves behind a positive space charge, that attracts cold bulk electrons towards the expanding sheath. The resulting displacement current ensures current continuity by locally compensating the enhancement of the conduction current. The back-flow of cold electron beams during one phase of sheath expansion and contribute to a strongly non-sinusoidal RF current. These kinetic mechanisms are the basis for a fundamental understanding of the electron power absorption dynamics and resonance phenomena in such plasmas, which were found to occur in discharges of different symmetries including perfectly symmetric plasmas. [Wilczek S, Trieschmann J, Eremin D, Brinkmann R P, Schulze J, Schüngel E, Derzsi A, Korolov I, Hartmann P, Donkó Z, Mussenbrock T; "Kinetic interpretation of resonance phenomena in low pressure capacitively coupled radio frequency plasmas"; Physics of Plasmas 23, 063514 (2016)]

The electron power absorption dynamics in radio frequency driven micro atmospheric pressure capacitive plasma jets were studied based on experimental phase resolved optical emission spectroscopy and the computational particle in cell simulations with Monte Carlo treatment of collisions. The jet was operated at 13.56 MHz in He with different admixture concentrations of N2 and at several driving voltage amplitudes. We have found the spatiotemporal dynamics of the light emission of the plasma at various wavelengths to be markedly different. This was understood by revealing the population dynamics of the upper levels of selected emission lines/bands based on comparisons between experimental and simulation results. The populations of these excited states are sensitive to different parts of the electron energy distribution function and to contributions from other excited states. Mode transitions of the electron power absorption dynamics from the  $\Omega$ - to the Penning-mode were found to be induced by changing the N2 admixture concentration and the driving voltage amplitude. Our numerical simulations have revealed details of this mode transition and provide novel insights into the operation details of the Penning-mode. The characteristic excitation/ emission maximum at the time of maximum sheath voltage at each electrode was found to be based on two mechanisms: (i) a direct channel, i.e. excitation/emission caused by electrons generated by Penning ionization inside the sheaths and (ii) an indirect channel, i.e. secondary electrons emitted from the electrode due to the impact of positive ions generated by Penning ionization at the electrodes. [L. Bischoff, G. Hübner, I. Korolov, Z. Donkó, P. Hartmann, T. Gans, J. Held, V. Schulz-von der Gathen, Y. Liu, T. Mussenbrock, J. Schulze, "Experimental and computational investigations of electron dynamics in micro atmospheric pressure radio-frequency plasma jets operated in He/N2 mixtures", Plasma Sources Sci. Technol. 27, 125009 (2018)]

We have investigated the origin of uncertainties in the results of numerical simulations of lowtemperature plasma sources, focusing on capacitively coupled plasmas. These sources can be operated in various gases/gas mixtures, over a wide domain of excitation frequency, voltage, and gas pressure. At low pressures, the non-equilibrium character of the charged particle transport prevails and particlebased simulations become the primary tools for their numerical description. The particle-in-cell method, complemented with Monte Carlo type description of collision processes, is a well-established approach for this purpose. Codes based on this technique have been developed by several authors/ groups, and have been benchmarked with each other in some cases. Such benchmarking demonstrates the correctness of the codes, but the underlying physical model remains unvalidated. This is a key point, as this model should ideally account for all important plasma chemical reactions as well as for the plasma-surface interaction via including specific surface reaction coefficients (electron yields, sticking coefficients, etc). In order to test the models rigorously, comparison with experimental 'benchmark data' is necessary. Examples were given regarding the studies of electron power absorption modes in  $O_2$ , and  $CF_4$ -Ar discharges, as well as on the effect of modifications of the parameters of certain elementary processes on the computed discharge characteristics in  $O_2$ capacitively coupled plasmas. [Z. Donkó, A. Derzsi, I. Korolov, P. Hartmann, S. Brandt, J. Schulze, B. Berger, M. Koepke, B. Bruneau, E. Johnson, T. Lafleur, J-P. Booth, A. R. Gibson, D. O'Connell and T Gans, "Experimental benchmark of kinetic simulations of capacitively coupled plasmas in molecular gases", Plasma Phys. Control. Fusion 60, 014010 (2018)]

We have investigated the spatially and temporally resolved electron kinetics in a homogeneous electric field in argon gas, in the vicinity of an emitting boundary. This (transient) region, where the electron swarm exhibits non-equilibrium character with energy gain and loss processes taking place at separate positions (in space and time), was monitored experimentally in a scanning drift tube apparatus. Depending on the strength of the reduced electric field we observe the equilibration of the swarm over different length scales, beyond which the energy gain and loss mechanism becomes locally balanced and transport properties become spatially invariant. The evolution of the electron swarm in the experimental apparatus was also described by Monte Carlo simulations, of which the results are in good agreement with the experimental observations, over the domains of the reduced electric field and the gas pressure covered. [Donkó Z, Hartmann P, Korolov I, Jeges V, Bošnjaković D, and Dujko S, "Experimental observation and simulation of the equilibration of electron swarms in a scanning drift tube", Plasma Sources Sci. Technol. 28, 095007 (2019)]

Single frequency, geometrically symmetric Radio-Frequency (RF) driven atmospheric pressure plasmas exhibit temporally and spatially symmetric patterns of electron heating, and consequently, charged particle densities and fluxes. Using a combination of phase-resolved optical emission spectroscopy and kinetic plasma simulations, we have demonstrated that tailored voltage waveforms consisting of multiple RF harmonics induce targeted disruption of these symmetries. This confines the electron heating to small regions of time and space and enables the electron energy distribution function to be tailored. [A. R. Gibson, Z. Donkó, L. Alelyani, L. Bischoff, G. Hübner, J. Bredin, S. Doyle, I. Korolov, K. Niemi, T. Mussenbrock, P. Hartmann, J. P. Dedrick, J. Schulze, T. Gans, D. O'Connell, "Disrupting the spatio-temporal symmetry of the electron dynamics in atmospheric pressure plasmas by voltage waveform tailoring", Plasma Sources Sci. Technol. 29, 01LT01 (2019)]

Atmospheric pressure capacitively coupled radio frequency discharges operated in He/N2 mixtures and driven by tailored voltage waveforms were investigated experimentally using a COST microplasma reference jet and by means of kinetic simulations as a function of the reactive gas admixture and the number of consecutive harmonics used to drive the plasma. Pulse-type 'peaks'waveforms, that consist of up to four consecutive harmonics of the fundamental frequency (f = 13.56MHz), were used at a fixed peak-to-peak voltage of 400 V. Based on an excellent agreement between experimental and simulation results with respect to the DC self-bias and the spatiotemporal electron impact excitation dynamics, we have demonstrated that Voltage Waveform Tailoring allows for the control of the dynamics of energetic electrons, the electron energy distribution function in distinct spatiotemporal regions of interest, and, thus, the generation of atomic nitrogen as well as helium metastables, which are highly relevant for a variety of technological and biomedical applications. By tuning the number of driving frequencies and the reactive gas admixture, the generation of these important species can be optimized. The behavior of the DC self-bias, which is different compared to that in low pressure capacitive radio frequency plasmas, was understood based on an analytical model. [Korolov I, Donkó Z, Hübner G, Bischoff L, Hartmann P, Gans T, Liu Y, Mussenbrock T, and Schulze J, "Control of electron dynamics, radical and metastable species generation in atmospheric pressure RF plasma jets by Voltage Waveform Tailoring", Plasma Sources Sci. Technol. 28, 094001 (2019)]

An atmospheric pressure surface-wave microwave discharge and a kHz plasma jet are used to activate purified water. It is shown, that by varying the treatment distance and the initial  $Ar/N_2/O_2$  mixture composition of the surface-wave microwave discharge the concentration ratio of  $NO_3$  and  $H_2O_2$ radicals created in the plasma activated water (PAW) can be varied over three orders of magnitude, which can be preserved during months of storage at room temperature. At the same time, with the 5 min treatment of the 32 ml water the absolute radical concentrations are varied in the range of 0.5 - 85 mg/l for H<sub>2</sub>O<sub>2</sub>, 20 - 180 mg/l for NO<sup>-3</sup> and 0.5 - 14 mg/l for NO<sup>-2</sup>. In the case of the N<sub>2</sub> kHz plasma jet this concentration ratio can be tuned within one order of magnitude by varying the treatment distance. By treating different volumes very similar concentration ratios are obtained, which evolve differently during storage, as the aging dynamics is determined by the absolute concentration of radicals. In general, the radical most affected by aging is  $NO_2^{-1}$ , whose recombination is found to be determined by the  $H_2O_2$  radical. In order to control the  $H_2O_2$  concentration and thus the NO radicals recombination, the application of a Fenton type reaction is suggested, which is implied by inserting a copper surface into PAW during or after plasma treatment. [Kutasi K, Popović D, Krstulović N, Milošević S, "Tuning the composition of plasma-activated water by a surface-wave microwave discharge and a kHz plasma jet", Plasma Sources Sci. Technol. 28 (2019) 095010]

#### ONGOING RESEARCH

The research activities that have made significant progress with the support of the research grant K-115805 but could not be finished by the end of the supported time period and are currently in the pre-publication phase are introduced in this section. The activities match the topics of the expired research proposal and are to be viewed as trivial continuation of it.

We investigate via Molecular Dynamics simulations the propagation of solitons in a two-dimensional many-body system characterized by Yukawa interaction potential. The solitons are created in an equilibrated system by the application of electric field pulses. Such pulses generate pairs of solitons, which are characterized by a positive and negative density peak, respectively, and which propagate into opposite directions. At small perturbation, the features propagate with the longitudinal sound speed, form which an increasing deviation is found at higher density perturbations. An external magnetic field is found to block the propagation of the solitons, which can, however, be released upon the termination of the magnetic field and can propagate further into directions that depend on the time of trapping and the magnetic field strength. [Donkó Z, Hartmann P, Masheyeva R U, Dzhumagulova K N, "Molecular dynamics investigation of soliton propagation in a two-dimensional Yukawa liquid", submitted to Contributions to Plasma Physics]

This work presents the transport coefficients of electrons (bulk drift velocity, bulk longitudinal diffusion coefficient, and effective ionization frequency) in  $C_2H_n$ , with n = 2, 4 and 6, measured in a scanning drift tube apparatus under time-of-flight conditions over a wide range of the reduced electric field, 1  $Td \leq E/N \leq 1790 Td$  (1  $Td = 10^{-21} Vm^2$ ). The data obtained in the experiments are also applied to determine the effective steady-state Townsend ionization coefficient. These parameters are compared to the results of previous experimental studies, as well as to the results of various kinetic computations: solutions of the electron Boltzmann equation under different approximations (multiterm and density gradient expansions) and Monte Carlo simulations. The experimental data are consistent with most of the transport parameters obtained in earlier studies. The kinetic computation results show that, in case of  $C_2H_2$ , the thermally excited vibrational population should not be neglected, in particularly, in the fitting of cross sections to swarm results. [*N R Pinhao, D Loffhagen, M*]

Vass, P Hartmann, I Korolov, S Dujko, D Bosnjakovic, Z Donkó, "Electron swarms parameters in  $C_2H_2$ ,  $C_2H_4$  and  $C_2H_6$ : measurements and kinetic computations", submitted to Plasma Sources Sci. Technol.]

The equilibrium structure and the dispersion relations of collective mode excitations in trilayer Yukawa systems in the strongly coupled liquid regime are examined. The equilibrium correlations reveal a rich variety of structures in the liquid phase, reminiscent of the corresponding structures in the solid phase. At small layer separation substitutional disorder becomes the governing feature. Theoretical dispersion relations are obtained by applying the Quasi-Localised Charge Approximation (QLCA) formalism, while numerical date are obtained from micro-canonical molecular dynamics (MD) method. The dispersion relations obtained both ways are compared and discussed in detail. We discover the existence of a novel spectral feature in the dynamical current-current correlation functions. Moreover, we provide new insight into the long-standing problem of gap-frequency discrepancy observed in layered strongly coupled systems in some earlier studies. [manuscript in preparation]

The PK-4 system is a micro-gravity dusty plasma experiment currently in operation on-board the International Space Station. The experiment utilizes a long DC discharge in neon or argon gases. We apply our 2D particle-in-cell with Monte Carlo collisions (PIC/MCC) discharge simulation to compute local plasma parameters that serve as input data for future dust dynamics models. The simulation includes electrons, Ne<sup>+</sup> ions, and Ne<sup>m</sup> metastable atoms in neon gas and their collisions at solid surfaces including secondary electron emission and glass wall charging. On the time-scale of the on-board optical imaging, the positive column appears stable and homogeneous. On the other hand, our simulations show that on microsecond time-scales the positive column is highly inhomogeneous, ionization waves with phase velocities in the range between 500 m/s and 1200 m/s dominate the structure. In these waves, the electric field and charged particle densities can reach amplitudes up to 10 times of their average value. Our experiments on a ground-based PK-4 replica system fully support the numerical findings. In the experiment, the direction of the DC current can be alternated. We show, that during the polarity switching an ionization tsunami swipes along the whole discharge towards the new cathode. [manuscript in preparation, preliminary results: P Hartmann, M Rosenberg, L S Matthews, T W Hyde, "Ionization waves in the PK-4 neon DC discharge", book of abstracts, page 69, 15th Dusty Plasma Workshop, May 29 – June 1, 2018, Baltimore, Maryland, USA

Since the early years of research on strongly coupled dusty plasmas, hydrodynamics has always appeared to be one of the topics that might be impacted by this exciting field to allow rapid progress. However, analyses of early experimental data have taught us that a more detailed understanding of dust inter-particle interactions and the fundamental system parameters involved is necessary before quantitatively meaningful results can be obtained. At the same time, single layer dusty plasma crystals produced in plane-parallel RF discharges became the main subject of many laboratory dusty plasma experiments in part because these configurations provide best controllability and reproducibility. As a result, they have become fairly well understood over time. We investigate hydrodynamic flows in a complex plasma by utilizing the control provided by a plane-parallel RF discharge. Two metallic disks are placed on the powered electrode of the new large area (16" diameter) dusty plasma setup at the CASPER Lab, forming an electrostatic potential channel. Dust particle flow through the channel is induced by indirect laser manipulation. By adjusting the argon gas pressure and RF power, the channel could be tuned to allow the formation of single (see image on title page illustrating the timeaveraged density distribution) or multiple lanes of transiting dust particles. We use this system to address fundamental details of hydrodynamic flows like the acceleration and stopping of particles, lane formation and ordering in the channel. [manuscript in preparation, preliminary results: P Hartmann, J C Reyes, L S Matthews, T W Hyde, "Microfluidic flow in single-layer dusty plasmas", book of abstracts, page 67, 15th Dusty Plasma Workshop, May 29 – June 1, 2018, Baltimore, Maryland, USA]

First experimental findings obtained with the new Magnetized Dusty Plasma Experiment (MDPX at Auburn University) include the filamentation of the gas discharge and the formation of ordered structures in high vertical magnetic fields (>1 T) imposed by the wire-grid upper horizontal electrode and visualized by micrometer dust particles levitating on top of the lower flat electrode sheath of a capacitively coupled RF discharge [E. Thomas Jr., Physics of Plasmas 23, 055701 (2016)]. Simple arguments based on charged particles being confined to helical trajectories along the magnetic field lines are not sufficient because all relevant collision processes have mean free paths well shorter than the distance between the structured electrode and the lower electrode sheath. We apply our newly implemented two-dimensional GPU accelerated particle in cell (PIC) discharge simulation both for cylindrical geometry to compute global discharge parameters and with cartesian symmetry for the determination of local plasma parameters. At the current stage the PIC simulations do reproduce both the filamentation and the imposed structure formation as shown in the figure. Detailed analysis of the magnetic field parallel and perpendicular diffusion shows strong connection between the magnetically altered particle transport and the ordering process. [manuscript in preparation, preliminary results: P Hartmann, E Thomas, 7r., M Rosenberg, "Filamentation and imposed ordered dust structure in magnetized discharge", book of abstracts, page 68, 15th Dusty Plasma Workshop, May 29 – June 1, 2018, Baltimore, Maryland, USA]

#### SUMMARY

The specific research topics of the current grant were grouped around Complex Plasmas, which are charged particle systems, where besides the elementary constituents (electrons and ions) the interaction with a more complex objects (solid/liquid surfaces, molecules, etc.) is of significant importance. Investigations were performed on several levels of complexity, including the fundamental operation of the gas discharges themselves, to the interaction (erosion, growth, composition modification) of plasmas with condensed materials, as well as the physical properties of strongly coupled dust particle ensembles inside of gas discharges. Our activities resulted in **18 peer-reviewed journal articles** and **one book chapter** that got published during the supported period, as well as **2 submitted and several in-preparation manuscripts**. Being a research project of purely fundamental nature, these, and the **numerous conference contributions** represent our primary output. We truly believe that some of our results (e.g. voltage waveform tailoring) will find direct application on short time scale in some high-tech industrial applications related to microelectronic and thin film fabrication.

I wish to express my gratitude to the funding agency National Research, Development and Innovation Office – NKFIH for making the above listed research activities possible through the grant K-115805 "Complex Plasmas in Action" by providing the necessary financial background absolutely essential to perform fundamental research in Hungary and the participation in international collaborations.

Budapest, 2019. Dec. 26.

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K-115805 final report