K 105476

ADVANCED NUMERICAL METHODS FOR THE PARTICLE-BASED SIMULATIONS OF COMPLEX PLASMAS

FINAL REPORT - APRIL 2017

The main focus of the project has been the investigation of various types of low-temperature (non-thermal) plasmas by different *numerical particle-based simulation approaches, their combinations,* and by *experimental methods*. Most of the studies have been accomplished within an international collaboration network. Part of the experimental work has been conducted in Budapest and the other part has been conducted in the specialized laboratories of our collaborators. In particular, two unique experimental systems has been designed and built up in the Wigner Research Centre within the frame of the project: (1) a dedicated system has been constructed to measure the breakdown characteristics of gases under radio-frequency excitation and (2) a scanning drift tube apparatus has been developed that allows measurements of electron transport parameters via mapping of electron swarms. More details about these activities will be given later.

Below, the main achievements of the project are summarized, in the same structure as it was described in the original research plan of the project. For each of the highlighted topics *a few representative publications* are cited, for the full list of journal articles, please see the main report. The activities have largely followed the plans, although their sequence was changed in a few cases. An extension of the deadline was requested (and granted) by three months to allow the successful accomplishment of planned experimental activities related to point "a)" as described below.

(a) Experimental studies of particle transport parameters. The principal devices for the investigations of particle transport in homogeneous electric fields have been the so-called *drift tubes*, of which the basic design contains two plane-parallel electrodes separated by a fixed distance. Electrons are emitted from one of the electrodes either in a pulsed, or continuous manner. The pulsed method is the choice for the measurements of drift velocity, the longitudinal diffusion coefficient, and the ionization frequency, whereas the continuous operation allows the determination of the Townsend-alpha ionization coefficient. The latter, however may also be derived from the quantities obtained in pulsed measurements by utilizing the connection between the Pulsed-Townsend and Steady-State Townsend transport coefficients. We have developed an experimental system that generates electron swarms on the basis of photoemission – by applying a forth-harmonic Nd:YAG laser with ns pulses. The system constructed in our laboratory in Budapest is unique in the sense that, in contrast with conventional systems, takes measurements at a high number of electrode separations to construct a map of the electron density, i.e. allows *scanning of the space and time-dependent development of the electron density* in the system. The capabilities of the system have been explored using argon gas,[1] subsequently measurements have been taken on methane, synthetic air, and deuterium.[2] *For carbon dioxide the experimental investigations have been complemented with theoretical studies based on Monte Carlo simulations as well as on solutions of the Boltzmann transport equation* (the latter being carried out by our collaborators in Germany and

[1] I. Korolov, M. Vass, N. Kh. Bastykova and Z. Donkó: "A scanning drift tube apparatus for spatiotemporal mapping of electron swarms", Rev. Sci. Instrum. 87, 63102 (2016).
[2] I. Korolov, M. Vass and Z. Donkó: "Scanning drift tube measurements of electron transport parameters in different gases: argon, synthetic air, methane and deuterium", J. Phys. D Appl. Phys. 49, 415203 (2016).

Portugal).[3] It is a unique feature of the system that – thanks to the visualization of the spatio-temporal development of the electron swarms – it allows identifying *deviations from the hydrodynamic transport conditions*. To our best knowledge such direct observations of *equilibration effects* were made possible first time with this apparatus.

(b) The phenomenon of gas breakdown in radio-frequency electric fields. To investigate the effect of gas breakdown in radio-frequency electric fields another unique apparatus has been constructed in our laboratory, which differs in two important aspects from the settings where this phenomenon is usually studied. (i) *We have built a high-voltage radio-frequency oscillator that allows precise control of high-frequency (1 MHz - 20MHz) voltages up to 1000 V peak-to-peak value (with a precision of 1V). The use of such a source allows more precise measurements of gas breakdown voltages as compared to the use of high-power radio-frequency generators that have less control on the output voltage. (ii) A special, geometrically symmetrical cell was built for the studies and care was taken to minimize stray capacitances, unlike in most experiments when the breakdown cell is enclosed within a grounded vacuum chamber, of which the presence can significantly affect the measured breakdown voltages in the MHz range*. The experimental system was first tested using argon gas, and subsequently measurements were taken in helium, synthetic air,[4] hydrogen and deuterium[5] gas as well. *In case of synthetic air comprehensive numerical simulations have also be conducted using a Monte Carlo type particle simulation code developed for this purpose, which uncovered the details of the breakdown mechanisms.*

(c) – (d) Studies of the mechanisms shaping the electron distribution function (EDF) in a gas under the effect of an external electric field and the phenomenon of electron energy relaxation in non-ideal plasmas. In a weakly ionized medium the EDF is shaped by the balance between energy gain (acceleration in the external electric field) and energy loss channels (elastic and inelastic energy losses in electron-atom collisions). Under these conditions Monte Carlo simulations provide an accurate description of the electron transport. In a fully-ionized medium electron-electron collisions lead to Maxwellization of the EDF, which can be described by Molecular Dynamics simulation. In the intermediate regime of the ionization degree both electron-atom and electron-electron collisions play a (comparable) role. This situation, to date, could only be described in an approximate manner as theoretical calculations made use of certain assumptions to account for the interactions between charged particles. *By incorporating the Monte Carlo collision approach into our Molecular Dynamics code in a novel way, we created an approximation-free computational tool[6] that due to its first principles approach accurately accounts for all these phenomena at the level of kinetic theory*. The particle simulations have been accompanied by the solutions of the Boltzmann equation (carried out by one of our collaborator in Russia). In these studies the effect of *the "bistability" of the EDF (i.e. the possibility of finding two stable solutions for the same set of conditions)* and its relaxation from different initial states has been addressed[7], as well as the peculiar effects of the *Negative Differential Conductivity and Transient Negative Mobility*[8] effects that appear in heavy noble gases.

[3] M. Vass, I. Korolov, D. Loffhagen, N. Pinhão and Z. Donkó, "Electron transport parameters in CO2: scanning drift tube measurements and kinetic computations", Plasma Sources Sci. Technol. Accepted for publication. (2017).
[4] I. Korolov, A. Derzsi and Z. Donkó: "Experimental and kinetic simulation studies of radio-frequency and direct-current breakdown in synthetic air", J. Phys. D Appl. Phys. 47, 475202 (2014).
[5] I. Korolov and Z. Donkó: "Breakdown in hydrogen and deuterium gases in static and radio-frequency fields", Phys. Plasmas 22, 93501 (2015).
[6] Z. Donkó: "First principles calculation of the effect of Coulomb collisions in partially ionized gases", Physics of Plasmas. 21, 43504 (2014).
[7] N. Dyatko and Z. Donkó: "Bistable solutions for the electron energy distribution function in electron swarms in xenon a comparison between the results of first-principles particle simulations and conventional Boltzmann equation analysis", Plasma Sources Sci. Technol. 24, 45002 (2015).
[8] Z. Donkó and N. Dyatko: "First-principles particle simulation and Boltzmann equation analysis of negative differential conductivity and transient negative mobility effects in xenon", Eur. Phys. J. D 70, 135 (2016).

(e) <u>Formation of waves and resonance effects in radio-frequency discharges.</u> According to our plans *we have implemented diagnostics methods routinely used in Molecular Dynamics simulations for the computation of dynamical structure functions (based on spatial and temporal Fourier transforms) into our Particle-in-Cell electrical discharge simulation code, to investigate the spatial distribution of the frequency spectra of several discharge characteristics, like the electron density, electric field and potential.* The studies have revealed *the spatial location of the generation of the higher harmonics of the radio-frequency excitation, and allowed studies of the non-linear Plasma Series Resonance oscillations that can be self-excited in radiofrequency discharges with asymmetric electrode configuration and/or under dual-frequency excitation.*[9] *A kinetic interpretation of the resonance effects was give*n based on particle-based simulations in which, as a key mechanism, opposite fluxes of electron groups with high / low energies were observed, which is clearly a kinetic effect that cannot be captured at the hydrodynamic level.[10]

(f) <u>Studies of the charge-voltage relation of radiofrequency (RF) electrode sheaths</u>. Recent theoretical predictions of *the deviations of the charge-voltage relation of RF sheaths from the quadratic behavior* (which has been assumed in the theory of RF discharges for decades) *have been confirmed by computing the relation in Particle-in-Cell simulations.*[11] These studies have considered the conditions when the ion transit time through the electrode sheaths is comparable to the period of the excitation radiofrequency voltage, which creates a considerable ion dynamics within the electrode sheaths. More detailed studies of the charge-voltage relation have *identified (and quantified for the conditions investigated) a cubic correction to the "basic" quadratic functional form*.[8]

g) <u>Investigations of heavy-particle effects in radio-frequency discharges.</u> It is well known that in direct-current (DC) and pulsed discharges besides positive ions fast neutral heavy particles, originating from charge exchange collisions, may contribute significantly to the electron emission from the electrodes and to the sputtering of the cathode, as well as to the light emission originating from the gas discharges. To uncover these effects *we have incorporated into our Particle-in-Cell simulation code a heavy-particle module that included ion and fast atom impact excitation and ionization processes.* In our studies of radio-frequency plasma sources we have found that, predominantly at low pressures (< few Pa) and at high driving voltage amplitudes *heavy particle ionization may play an important role in the ionization balance*. This is the case because *these ionization processes take place in the vicinity of the electrode surfaces (where ion and consequently fast neutral energies are high) and the electrons that are created in these processes can be accelerated to high energies in the high sheath electric field*. Our studies have also concluded that fast neutrals may give a significant contribution to the emission of secondary electrons at the electrodes of the discharge.[12]

h) <u>Studies of the effects of dust particles growing in radiofrequency discharges.</u> We have addressed two effects related to the presence of dust in electrical discharges. The first study has addressed the *possibility of transporting dust particles within the discharge plasma* due to the

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[9] E. Schuengel, S. Brandt, I. Korolov, A. Derzsi, Z. Donkó and J. Schulze: "On the self-excitation mechanisms of plasma series resonance oscillations in single- and multi-frequency capacitive discharges", Phys. Plasmas 22, 43512 (2015).

[10] S. Wilczek, J. Trieschmann, D. Eremin, R. P. Brinkmann, J. Schulze, E. Schuengel, A. Derzsi, I. Korolov, P. Hartmann, Z. Donkó and T. Mussenbrock: "Kinetic interpretation of resonance phenomena in low pressure capacitively coupled radio frequency plasmas", Phys. Plasmas 23, 63514 (2016).

[11] M. Shihab, A. T. Elgendy, I. Korolov, A. Derzsi, J. Schulze, D. Eremin, T. Mussenbrock, Z. Donkó, R. P. Brinkmann: "Kinetic simulation of the sheath dynamics in the intermediate radio frequency regime", Plasma Sources Sci. Technol. 22, 55013 (2013).

[12] A. Derzsi, I. Korolov, E. Schuengel, Z. Donkó and J. Schulze: "Effects of fast atoms and energy-dependent secondary electron emission yields in PIC/MCC simulations of capacitively coupled plasmas", Plasma Sources Sci. Technol. 24, 34002 (2015).

change of the excitation waveform.[13] Dust particles in plasmas typically form a layer located at the sheath edge adjacent to the bottom electrode. We proposed *a method of manipulating this distribution by the application of a specific excitation waveform*, i.e. two consecutive harmonics, is discussed. Tuning the phase angle between the two harmonics allows one to adjust the discharge symmetry via the electrical asymmetry effect. We have found that *by applying an abrupt phase shift the dust particles were transported between both sheaths through the plasma bulk and partially reside at an equilibrium position close to the upper sheath edge*. The motion was understood by an analytical model, showing both the limitations and possible ways of optimizing this *sheath-to-sheath transport*. In another study we investigated the possibility of the *control of the equilibrium position of micrometer sized dust particles* in radio-frequency discharges both experimentally and via Particle-in-Cell simulations using an RF + DC driving voltage signal for the plasma source.[14]

i) Binary mixtures of strongly correlated systems of charged particles. In *binary systems* two types of charged particles, which in general have *different charges and different masses*, co-exist. For such systems profound differences between the properties of the collective modes exist between the weakly and strongly correlated limits: at weak coupling the two components interact via the mean field only and the oscillation frequency is governed by the light component. In the strongly correlated limit the mode frequency is governed by the combined mass, where the heavy component dominates. In the project we have addressed the collective mode structure of two-dimensional binary Yukawa systems,[15] and gave an analysis of the creation of a second plasmon mode in three-dimensional systems:[16] *at strong coupling the doublet of high-frequency (first) and low-frequency (second) plasmons was found to replace the single-plasmon excitation that prevailed at weak coupling. We observed the formation of the second plasmon from the acoustic Goldstone-type mode associated with a short-range interaction as the range was extended to infinity* (i.e. when the Coulomb potential replaced the Yukawa interaction potential due the disappearance of screening effects). Our molecular dynamics simulations of a strongly coupled binary ionic mixture have revealed *the appearance of sharp minima in the species-resolved dynamical density fluctuation spectra, being reminiscent of the well-known Fano anti-resonance, occurring in various physical processes*. We gave a theoretical analysis using the quasi-localized charge approximation, and demonstrated that *the observed phenomenon in the equilibrium spectrum is a novel manifestation of the Fano mechanism, that occurs at characteristic frequencies of the system different from the conventional classical Fano frequencies*.[17]

j) Various model potentials used in the description of strongly-interacting many-body systems. During these studies we have devoted special attention to *the dipole-dipole interaction that arises in many body systems when besides the electric charge the particles also acquire a magnetic moment in the presence of an external electric field*. This particular interaction of superparamagnetic grains can be tuned by the direction of the magnetic field with respect to the particle layer where the particles are situated. We have determined the *limits of stability of the system*, (which is determined by the competition between the repulsive Coulomb (or Yukawa) interaction and the dipole-dipole interaction that can turn to be attractive at certain directions of the

[13] S. Iwashita, E. Schuengel, J. Schulze, P. Hartmann, Z. Donkó, G. Uchida, K. Koga, M. Shiratani, U. Czarnetzki: "Transport control of dust particles via the electrical asymmetry effect experiment, simulation and modelling", J. Phys. D Appl. Phys. 46, 245202 (2013).

[14] N. Kh. Bastykova, A. Zs. Kovács, I. Korolov, S. K. Kodanova, T. S. Ramazanov, P. Hartmann and Z. Donkó: "Controlled Levitation of Dust Particles in RF+DC Discharges", Contrib. Plasma Phys. 55, 671 (2015).

[15] G. J. Kalman, P. Hartmann, Z. Donkó, K. I. Golden and S. Kyrkos: "Collective modes in two-dimensional binary Yukawa systems", Phys. Rev. E 87, 43103 (2013).

[16] G. J. Kalman, Z. Donkó, P. Hartmann, K. I. Golden: "Second plasmon and collective modes in binary Coulomb systems", Europhys. Lett. 107, 35001 (2014).

[17] L. Silvestri, G. J. Kalman, Z. Donkó, P. Hartmann and H. Kahlert: "Fano-like anti-resonances in strongly coupled binary Coulomb systems", Europhys. Lett. 109, 15003 (2015).

external B field) and the properties of the collective excitations in the system.[18] Whenever the magnetic field is not inclined in a perpendicular direction with respect the particle layer the *pair correlations and the collective mode properties become anisotropic depending on their direction of the propagation*. As an extension we have also investigated a setting where the *particles are confined by a parabolic potential to a single layer with finite width*; this trapping gives rise to an additional *optic collective mode*.[19]

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During the course of the research planned originally, several *new interesting questions* came up and efforts have been made to extend the investigations into these *additional directions*, as well. A few of these directions included the following: (1) we have successfully *benchmarked* our Particle-in-Cell simulation code with codes of other groups[20], (2) we have demonstrated the applicability of *light field photography* for the reconstruction of three-dimensional configuration of dust particles in discharge plasmas from single-exposure recording[21], (3) we have worked on the *optimization of the flux-energy distribution function of the ions reaching the electrodes* in radio-frequency plasma sources[22], (4) we have carried out combined experimental and simulation studies – using different electronegative gases like carbon-tetrafluoride[23] and oxygen[24] – of multi-frequency-driven capacitively-coupled RF plasma sources, which are currently entering the focus of interest in the field of low-pressure plasma discharges, (5) we have investigated the effects of magnetic fields on strongly coupled plasmas.[25]

Our *international collaboration network* has considerably been extended as driven by mutual benefits of the availability of the different experimental infrastructures, simulation codes, and know-how at different institutions. In particular, links have been developed with the leading European institutions of, (i) Ecole Polytechnique, Palaiseau, and the (ii) York Plasma Institute, Department of Physics, University of York, as well as with the al-Farabi Kazakh National University and the State Research Center of Russian Federation Troitsk Institute for Innovation and Fusion Research.

Students have successfully been participating in the research activities:

- Péter Magyar (ELTE) submitted his MSc thesis about the quadratic static structure functions of Coulomb and Yukawa liquids in 2014, since that time his PhD research has been conducted in the field of higher-order structure and response functions of strongly coupled plasmas. His work on this topic received a *poster prize* at the Physics of Non-ideal Plasmas (PNP-11)

[18] P. Hartmann, Z. Donkó, M. Rosenberg and G. J. Kalman: "Waves in two-dimensional superparamagnetic dusty plasma liquids", Phys. Rev. E 89, 43102 (2014).
[19] M. Rosenberg, G. J. Kalman, P. Hartmann and Z. Donkó: "Waves in a quasi-two-dimensional superparamagnetic dusty plasma liquid in a trap", Phys. Rev. E 94, 33203 (2016).
[20] M. M. Turner, A. Derzsi, Z. Donkó, D. Eremin, S. J. Kelly, T. Lafleur, T. Mussenbrock: "Simulation benchmarks for low-pressure plasmas Capacitive discharges", Phys. Plasmas 20, 13507 (2013).
[21] P. Hartmann, I. Donkó and Z. Donkó: "Single exposure three-dimensional imaging of dusty plasma clusters", Rev. Sci. Instrum. 84, 23501 (2013).
[22] E. Schuengel, Z. Donkó, P. Hartmann, A. Derzsi, I. Korolov and J. Schulze: "Customized ion flux-energy distribution functions in capacitively coupled plasmas by voltage waveform tailoring", Plasma Sources Sci. Technol. 24, 45013 (2015).
[23] S. Brandt, B. Berger, E. Schuengel, I. Korolov, A. Derzsi, B. Bruneau, E. Johnson, T. Lafeur, D. O'Connell, M. Koepke, T. Gans, J.-P. Booth, Z. Donkó and J. Schulze: "Electron power absorption dynamics in capacitive radio frequency discharges driven by tailored voltage waveforms in CF4", Plasma Sources Sci. Technol. 25, 45015 (2016).
[24] A. Derzsi, T. Lafleur, J.-P. Booth, I. Korolov and Z. Donkó: "Experimental and simulation study of a capacitively coupled oxygen discharge driven by tailored voltage waveforms", Plasma Sources Sci. Technol. 25, 15004 (2016).
[25] K. N. Dzhumagulova, R. U. Masheyeva, T. Ott, P. Hartmann, T. S. Ramazanov, M. Bonitz and Z. Donkó: "Cage correlation and diffusion in strongly coupled three-dimensional Yukawa systems in magnetic fields", Phys. Rev. E 93, 63209 (2016); T. Ott, Z. Donkó and M. Bonitz: "The Energy-Autocorrelation Function in Magnetized and Unmagnetized Strongly Coupled Plasmas" Contrib. Plasma Phys. 56, 246 (2016); P. Hartmann, Z. Donkó, T. Ott, H. Kahlert and M. Bonitz: "Magnetoplasmons in Rotating Dusty Plasmas", Phys. Rev. Lett. 111, 155002 (2013).

Conference in 2015, and one of his related articles[26] has been chosen as *Editor's choice* in the journal Contributions to Plasma Physics.

- Máté Vass (ELTE) has been participating in the activities related to the electron transport parameters, both via experimental data analysis and kinetic computations of electrons swarm transport parameters. His BSc thesis will be submitted in the first semester of 2017 about his work related to investigations on CO_2 gas. This work was given the privilege of a *hot-topic presentation* at the ESCAMPIG 2016 conference. He received as well a *poster prize* at the Symposium on the Application of Plasma Processes (SAPP) 2017 Conference, *first prize* at the ELTE TDK Conference in 2016 and *first prize* in 2017 in the National TDK (OTDK) Conference in Debrecen.

The results of the work have been published in *45 research articles* in leading scientific journals, with a *total impact factor* of 104.7. The participants have given *several invited lectures* at international conferences that resulted in a good visibility of the project at the international scene.

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Zoltán Donkó, PI

[26] P. Magyar, P. Hartmann, G.J. Kalman, K. I. Golden and Z. Donkó: "Factorization of 3-Point Static Structure Functions in 3D Yukawa Liquids", Contrib. Plasma Phys. 56, 816 (2016).