Final report

High-performance modeling and simulation of low-temperature and strongly coupled plasmas

OTKA K 77653

Principal investigator: Dr Zoltán Donkó, DSc.

Introduction

The focus of the research conducted within the frame of the OTKA K 77653 grant has been the modeling and computational simulation of low-temperature gas discharges and strongly coupled plasmas. Our work has principally been carried out in Budapest, however, our team has been strongly linked with foreign research groups; the collaboration with these groups has significantly been aided by the International Supplementary Grant OTKA IN 85261 that has provided support for mobility.

It is to mention that the simulation studies of dusty plasma systems, planned and accomplished within the frame of the project, have been conducted in close cooperation with the OTKA PD-75113 and NN 103150 projects, focusing on experiments (PI: Dr Peter Hartmann). Credit for the experimental results (some of which being mentioned in this report for the sake of completeness) is to be given to these grants.

Computational modeling has become an invaluable tool for the understanding of the behavior of complex systems in all areas of physics. Numerical modeling and simulations in basic research represent today a complementary approach to theoretical calculations and/or experimental investigations, and provide an independent source of knowledge. While the major part of our work has been based on numerical studies, we have also developed experimental setups in our own laboratory in Budapest and have exploited as well the possibility to carry out experiments in the laboratories of our collaborating partners abroad. This way, several of our publications report combined theoretical / numerical / experimental studies.

Our work has been focused on two areas of (non-fusion) plasma physics: (i) Low-temperature non-equilibrium plasmas and (ii) Strongly coupled plasmas.

Low-temperature non-equilibrium plasmas appear in light sources, gas lasers, technological processing of semiconductor devices, plasma-based surface modification (etching and deposition) techniques, surface analysis, and many more important laboratory, as well as industrial applications. One of the most relevant characteristics of these non-thermal plasmas is the high electron temperature, compared to the gas temperature - these systems are far from thermodynamical equilibrium. At low pressures electrons and ions have long free paths in the plasma volume and their motion can only be traced accurately at the kinetic level. Particle-based approaches, like Monte Carlo and Particle-in-Cell simulation methods, play a key role in the self-consistent description of such systems.

Strongly coupled plasmas, in which the average potential energy per particle dominates over the average kinetic energy, appear in a wide variety of physical systems: dusty plasmas, charged particles in cryogenic traps, condensed matter systems such as molten salts and liquid metals, electrons trapped on the surface of liquid helium, astrophysical systems, such as the ion liquids in white dwarf interiors, neutron star crusts, supernova cores, and giant planetary interiors, as well as in degenerate electron or hole liquids in two-dimensional or layered semiconductor nanostructures. The main techniques applicable for the description of these systems are Monte Carlo and Molecular Dynamics simulations, which allow one to determine both the static properties (equation of state, transport coefficients, etc.) and dynamical characteristics (relaxation effects, collective excitations, etc.).

Our work in both of the above areas has been based on simulation codes that have been developed, maintained and extended for the specific topics proposed, by our group.

Results

The main topics of our work, as listed in the project proposal, have been:

A. Transport phenomena in strongly-coupled many-particle systems
B. Thermodynamical properties and collective excitations in strongly coupled many-particle systems
C. Semiclassical many-particle simulations
D. Simulation of radio-frequency gas discharges by Particle-in-Cell (PIC) method
E. Investigation of the role of primary electrons in radio-frequency discharges

In the following we give details of our accomplishments in these fields, with references to the relevant publications.

A. Transport phenomena in strongly-coupled many-particle systems

Transport phenomena in low-dimensional systems are of high current interest. Due to the reduced dimensionality the correlation functions associated with the transport coefficients exhibit a slow decay for some settings, making it impossible to calculate the transport coefficients via the Green-Kubo relations. For such conditions, although simulations may give values for the transport coefficients; these do not exist rigorously, and consequently, the simulation results may depend on the system size. The existence of coefficients for diffusion, viscosity, and thermal conductivity has been examined in collaboration with the University of Iowa, for two-dimensional charged liquids via the analysis of time correlation functions of particle velocity, shear stress, and energy flux, respectively.[1] Equilibrium molecular dynamics (MD) simulations were performed using a Yukawa potential and the long-time behavior of autocorrelation functions was tested. We have found that transport coefficients exist for diffusion at high temperature and viscosity at low temperature, but not in the opposite limits. The thermal conductivity coefficient does not appear to exist at high temperature. While most of our simulations are carried out on systems of about 10,000 particles, we have explored an alternative way of computing, based on Graphics Processing Units. An equilibrium molecular dynamics code has been implemented under CUDA environment and was successfully used for the determination of the dependence of the shear viscosity on the plasma coupling parameter, in one-million-particle simulations.[2]

As a next step beyond investigating static properties, we have as well studied the complex viscosity of strongly coupled many-particle system to uncover their viscoelastic properties. This was first approached by simulations: we have used both (i) an equilibrium MD method (that yielded the complex viscosity via the generalized Green-Kubo relation, that includes Fourier transform of the velocity autocorrelation function) and a (ii) nonequilibrium method that assumes periodically oscillating sliding boundary conditions of the simulation cell to impose a harmonically varied shear on the system. The results obtained by the two methods showed an excellent agreement, and indicated that with increasing frequency the real part of the viscosity (connected with dissipation) decays, while the imaginary part (related to the elastic response) increases, and becomes dominant at high frequencies.[3] Later on we have confirmed this behavior experimentally by imposing a periodically varying shear (by means of laser light) in a novel optical setup, on a two-dimensional dusty plasma.[4,5]

[1] Z. Donkó, J. Goree, P. Hartmann and Bin Liu: "Time-correlation functions and transport coefficients of two-dimensional Yukawa liquids", Physical Review E 79, 026401 (2009).

[2] Á. Budea, A. Derzsi, P. Hartmann and Z. Donkó: "Shear Viscosity of Liquid-Phase Yukawa Plasmas from Molecular Dynamics Simulations on Graphics Processing Units", Contrib. Plasma Phys. 52, 194 (2012).

[3] Z. Donkó, J. Goree, and P. Hartmann: "Viscoelastic response of Yukawa liquids", Phys. Rev. E 81, 056404 (2010).

[4] P. Hartmann, M. Cs. Sándor, A. Kovács, Z. Donkó: "Static and dynamic shear viscosity of a single-layer complex plasma", Phys. Rev. E, 84, 016404 (2011).

[5] A. Zs. Kovács, P. Hartmann and Z. Donkó: "Dynamic Shear Viscosity in a 2D Yukawa System", Contrib. Plasma Phys. 52, 199 (2012).

B. Thermodynamical properties and collective excitations in strongly coupled many-particle systems

We have developed new simulation codes and extended our earlier codes to investigate a variety of strongly-coupled many-particle systems and the different prominent phenomena characterizing them. Besides having identified gapped collective excitations (with nonzero frequency at k=0 wave number) in earlier simulations of bilayer liquids, we have confirmed the existence of such modes experimentally.[6] The bilayer configuration has been realized by dispersing two types of microparticles into the ionized gas environment, which, due to the balance of gravity and electrostatic forces, have settled into two layers, depending of the particle mass and particle charge acquired from the plasma. The experimental data showed very good agreement with the molecular dynamics simulation results. In a system of monodisperse particles, that settle in a single layer, the crystallization dynamics has been studied experimentally and by means of simulations.[7]

Going beyond the conventionally used Coulomb / Yukawa interaction potentials we have studied the collective excitation spectrum of a two-dimensional setting of point dipoles[8], and investigated the effect of an attractive term of the interaction potential in addition to the Debye-Hückel potential, conventionally used to model dusty plasmas, on the static and dynamical characteristics of many-body dust–plasma systems.[9]

The magnetic field is known to shift the frequencies of the collective modes. In a collaboration with the Christian-Albrechts University of Kiel, analyzing the dynamical structure function and current-current fluctuation spectra of magnetized strongly coupled plasmas, we have discovered the appearance of nonlinear magnetoplasmons, which bear similarities to the Bernstein modes in weakly coupled magnetized plasmas.[10,11]The investigation of the collective modes has been extended, in a joint work with University of California at San Diego, to strongly coupled liquids consisting of superparamagnetic grains.[12]

We have analyzed via theoretical approaches and molecular dynamics simulations the collective mode structure of strongly coupled two-dimensional *binary* Yukawa systems, in collaboration with Boston College and the University of Vermont, for selected density, mass, and charge ratios, both in the liquid and crystalline solid phases.[13] Theoretically, the liquid phase is described through the quasilocalized charge approximation (QLCA) approach, while in the crystalline phase we study the centered honeycomb and the staggered rectangular crystal structures through the standard harmonic phonon approximation. We identified “longitudinal” and “transverse” acoustic and optic modes and have found that the longitudinal acoustic mode evolves from its weakly coupled counterpart in a discontinuous nonperturbative fashion. The low-frequency acoustic excitations are governed by the oscillation frequency of the average atom, while the high-frequency optic excitation frequencies are related to the Einstein frequencies of the systems.[14] As systems of reduced dimensionality, bilayer liquids have also been considered.[15,16]

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[6] P Hartmann, Z. Donkó, G J Kalman, S Kyrkos, K I Golden, M Rosenberg: “Collective Dynamics of Complex Plasma Bilayers”, Phys. Rev. Lett. 103, 245002 (2009).

[7] P. Hartmann, A. Douglas, J. C. Reyes, L. S. Matthews, T. W. Hyde, A. Kovács and Z. Donkó: “Crystallization Dynamics of a Single Layer Complex Plasma”, Phys. Rev. Lett. 105, 115004 (2010).

[8] K. I. Golden, G. J. Kalman, P. Hartmann and Z. Donkó: “Dynamics of two-dimensional dipole systems”, Phys. Rev. E 82, 036402 (2010).

[9] Z. Donkó, P. Hartmann and P. K. Shukla: "Consequences of an attractive force on collective modes and dust structures in a strongly coupled dusty plasma", Phys. Lett. A 376, 3199–3203 (2012).

[10] M. Bonitz, Z. Donkó, T. Ott, H. Kählert and P. Hartmann: “Nonlinear Magnetoplasmons in Strongly Coupled Yukawa Plasmas”, Phys. Rev. Lett. 105, 055002 (2010).

[11] T. Ott, M. Bonitz, P. Hartmann, and Z. Donkó: “Higher harmonics of the magnetoplasmon in strongly coupled Coulomb and Yukawa systems”, Phys. Rev. E 83, 046403 (2011).

[12] P. Hartmann, M. Rosenberg, G. J. Kalman, Z. Donkó: “Ground-state structures of superparamagnetic two-dimensional dusty plasma crystals”, Phys. Rev. E, 84, 016409 (2011).

[13] G. J. Kalman, Z. Donkó, P. Hartmann, K. I. Golden: “Strong Coupling Effects in Binary Yukawa Systems”, Phys. Rev. Lett., 107, 175003 (2011).

[14] 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, 043103 (2013).

[15] K. I. Golden, G. J. Kalman, P. Hartmann, Z. Donkó: "Collective Modes in Classical Mass-Asymmetric Bilayers", Contrib. Plasma Phys. 52, 130 (2012).

[16] G. J. Kalman, Z. Donkó, P. Hartmann, K. I. Golden and S. Kyrkos: "Collective Modes in Strongly Coupled Binary Liquids", Contrib. Plasma Phys. 52, 234 (2012).

The ability of a medium to sustain a shear wave requires an elastic response to a disturbance, where a particle tends to be restored to its equilibrium position after being disturbed. A liquid, unlike a solid, exhibits elastic responses only for a limited time and over a limited distance. This is so because the potential landscape surrounding a particle in a liquid does not remain unchanged indefinitely as in a solid. When neighboring particles rearrange their positions irreversibly, there will also be an irreversible change in the confining potential landscape, and energy will be dissipated. Until a rearrangement occurs, a disturbance in the position of one particle can be restored elastically. We have determined the minimum wave number, above which shear waves can propagate in strongly coupled liquids, via the analysis of the transverse current autocorrelation function. Additionally, the crossover frequency for the real and imaginary terms of the complex viscosity of a dusty plasma has been obtained; this crossover frequency corresponds to the Maxwell relaxation time.[17]

C. Semiclassical many-particle simulations

The dynamic ion structure factor (DISF), $S(k,\omega)$, of dense plasmas and warm dense matter (WDM) is of great importance as it contains the complete information on the ions in these strongly interacting systems and is also influenced by the electron properties. It is closely related to the density response function and determines many relaxation and transport properties, e.g. stopping power and electron-ion temperature equilibration, and the equation of state. The DISF may also be used for the diagnostics of extreme states of matter like WDM by means of x-ray Thomson scattering. We have studied the dynamics of the ion structure in WDM by molecular dynamics simulations[18] using an effective ion-ion potential, that has been obtained from ab-initio simulations (of our partners at the University of Warwick, UK) and has a strong short-range repulsion added to a screened Coulomb potential.

D. Simulation of radio-frequency gas discharges by Particle-in-Cell (PIC) method

Capacitively coupled radio frequency (CCRF) discharges have become widely used efficient tools for a variety of applications, ranging from plasma based etching and deposition procedures in the semiconductor industry to plasma assisted surface treatment of medical interest. Extensive study of CCRF discharges has been, and still is, strongly motivated by the practical goal of clarifying the effect of external control parameters on process relevant plasma characteristics. Especially, the feasibility of the separate control of the ion flux and energy distribution at the substrate, which is crucial for applications, has been attracting particular attention. The realization of such control necessitates a detailed understanding of the complex physics of CCRF plasmas: underlying the electron heating and ionization dynamics. Besides the important applications mentioned above, these systems exhibit a rich physics, making them attractive subjects of fundamental research. Their complexity mainly arises from the nonlocality of the electron transport, in temporally and spatially rapidly varying electric field. To describe correctly the discharges operated under such conditions mostly particle-based approaches have been used. Our group has developed several simulation codes based on the Particle-in-Cell approach, complemented with stochastic (Monte Carlo) handling of collision processes - known as PIC/MCC technique. In the frame of the grant we have studied several properties of radio-frequency (RF) discharges, by applying and further developing our codes. A central topic has been the investigation of the possibilities of the independent control of ion properties: the *ion mean energy* (or energy distribution) and the *ion flux* at the electrodes. The two main approaches that have been applied for this purpose, both using a superposition of two or more frequencies to drive the discharges, can be categorized as follows:

▸ Classical dual-frequency (DF) discharges are most frequently used in industry and are typically driven by a voltage waveform, $V(t)=V_{HF}\cos(2\pi f_{HF}t) + V_{LF}\cos(2\pi f_{LF}t)$. The idea to obtain separate control of ion properties in these discharges is the functional separation of both frequencies due to the substantial frequency difference $f_{HF} >> f_{LF}$. The high frequency f_{HF} voltage amplitude, V_{HF}, is assumed to sustain the plasma and, consequently, to control the charged particle density and the ion flux. The low frequency f_{LF} voltage amplitude, V_{LF}, is assumed to control the acceleration of the ions determining the mean ion energy without affecting the ion flux.

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[17] J. Goree, Z. Donkó and P. Hartmann: "Cutoff wave number for shear waves and Maxwell relaxation time in Yukawa liquids", Phys. Rev. E 85, 066401 (2012).

[18] J. Vorberger, Z. Donkó, I. M. Tkachenko and D. O. Gericke: "Dynamic Ion Structure Factor of Warm Dense Matter", Phys. Rev. Lett. 109, 225001 (2012).

▸ A more recently invented approach, called the “Electrical Asymmetry Effect” (EAE)[19], uses a fundamental frequency and its second harmonic, i.e. the following driving voltage waveform: $V(t)=V_{LF}\cos(2\pi f+\theta)+V_{HF}\cos(4\pi ft)$. Here, θ is the fixed, but adjustable, phase shift between the driving harmonics. Typically, f = 13.56 MHz is used. The concept to control the mean ion energies at the electrodes separately from the ion flux in these discharges is fundamentally different from the concept used in classical DF discharges. Here, the voltage amplitudes are kept constant and only the relative phase between the driving frequencies, θ, is changed to control the ion energy. In this way any frequency coupling is avoided. Tuning θ results in the electrical generation of a variable dc self-bias, that depends almost linearly on θ due to the EAE even in geometrically symmetric discharges. Thus, the mean ion energies at the electrodes can be controlled by tuning θ.

We have intensively studied operation of discharges under both of these driving modes, in collaboration with the Institute for Plasma and Atomic Physics, at Ruhr University, Bochum, where experimental facilities have been available to complement the simulation studies. The main achievements of these studies in the specific topics can be summarized as follows:

Plasma series resonance oscillations: At low pressures, nonlinear self-excited plasma series resonance (PSR) oscillations are known to drastically enhance the electron heating in geometrically asymmetric capacitively coupled radio frequency discharges by nonlinear electron resonance heating. We have demonstrated via Particle-in-Cell simulations that high-frequency PSR oscillations can also be excited in geometrically symmetric discharges if the driving voltage waveform makes the discharge electrically asymmetric. This can be achieved by a dual-frequency f+2f excitation, when PSR oscillations are turned on and off depending on the electrical discharge asymmetry, controlled by the phase difference of the driving frequencies.[20]

Optimization of the electrical asymmetry effect: An electrical asymmetry in capacitive rf discharges with a symmetrical electrode configuration can be induced by driving the discharge with a fundamental frequency and its second harmonic. For equal amplitudes of the applied voltage waveforms, it has been demonstrated by modeling, simulation, and experiments that this electrical asymmetry effect leads to the generation of a variable dc self-bias that depends almost linearly on the phase angle between the driving voltage signals. We have investigated the dependence of the dc self-bias generated by the EAE on the choice of the voltage amplitudes, i.e., the ratio of high to low frequency amplitude, experimentally as well as by using an analytical model andParticle-in-Cell simulations.[21]

Charge and excitation dynamics in electrically asymmetric discharges: An analytical model to describe the charge distribution dynamics in CCRF discharges has been developed. In contrast to the assumption of a temporally constant total uncompensated charge in the discharge made in several models of CCRF discharges, it predicted a modulation of the total charge by about 10% around its time average value, within one radio-frequency period.[22] Simulations carried out in conjunction with “Phase resolved optical emission spectroscopy” measurements[23] revealed information about the spatio-temporal distribution of the excitation and ionization events in the discharge.[24]

Power absorption in electrically asymmetric discharges: We have investigated the power absorption by electrons and ions in electrically asymmetric dual frequency capacitively coupled radio frequency discharges

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[19] U. Czarnetzki, J. Schulze , E. Schüngel and Z. Donkó: "The electrical asymmetry effect in capacitively coupled radio-frequency discharges", Plasma Sources Sci. Technol. 20, 024010 (2011).

[20] Z. Donkó, J. Schulze, U. Czarnetzki and D. Luggenhölscher: "Self-excited nonlinear plasma series resonance oscillations in geometrically symmetric capacitively coupled radio frequency discharges", Appl. Phys. Lett. 94, 131501 (2009).

[21] J. Schulze, E. Schüngel, U. Czarnetzki and Z. Donkó: "Optimization of the electrical asymmetry effect in dual-frequency capacitively coupled radio frequency discharges: Experiment, simulation, and model", J. Appl. Phys. 106, 063307 (2009).

[22] J. Schulze, E. Schüngel, Z. Donkó, and U. Czarnetzki: "Charge dynamics in capacitively coupled radio frequency discharges", J. Phys. D: Appl. Phys. 43 225201 (2010).

[23] . Schulze, E. Schüngel, Z. Donkó, D. Luggenhölscher, and U. Czarnetzki: "Phase resolved optical emission spectroscopy: a non-intrusive diagnostic to study electron dynamics in capacitive radio frequency discharges", J. Phys. D: Appl. Phys. 43 124016 (2010).

[24] J. Schulze, E. Schüngel, Z. Donkó, and U. Czarnetzki: "Excitation dynamics in electrically asymmetric capacitively coupled radio frequency discharges: experiment, simulation, and model", Plasma Sources Sci. Technol. 19 045028 (2010).

experimentally, by simulations, and by an analytical model. Based on the model of the EAE, we have shown that the power absorbed by the electrons is nearly constant as a function of the phase shift θ between the driving harmonics both at high and low pressures. These theoretical results have been verified experimentally and by PIC simulations. Based on the energy balance for the electrons, we have demonstrated that a constant electron power absorption causes the ion flux at the electrodes to remain constant as a function of θ. In this way, the questions why the ion flux is approximately constant as a function of θ and why the ion energy can be controlled separately from the ion flux in an almost ideal way in electrically asymmetric dual frequency discharges have been answered.[25]

Electronegative radio-frequency discharges: The electron heating mechanisms and the Electrical Asymmetry Effect in dual-frequency capacitive CF_4 discharges have been studied.[26] We have identified a novel operation mode of electronegative capacitively coupled radio-frequency discharges characterized by ionization in the bulk plasma and at the sheath edges at distinct times within the RF period.[27] An analytical model clarified the physical origin of this Drift-Ambipolar (DA) mode to be (i) a strong drift electric field in the discharge center due to the low dc conductivity caused by the low electron density and (ii) an ambipolar field at the sheath edges due to local maxima of the electron density in the electropositive edge region of the discharge. By increasing the voltage at fixed pressure a transition from the DA- into the sheath expansion heating (alpha) mode was induced, while increasing the pressure at fixed voltage induced a transition from the alpha into the DA mode. These transitions were observed both in simulations and experiments (carried out at our collaborating partners at the University of Greifswald, Germany). Our model predicted a high drift field in the bulk to be also caused by a high electron-neutral collision frequency, i.e., at high pressures, such as observed in atmospheric pressure microplasmas. We have developed a model that revealed the physical origin of this "Ω"-mode by identifying the high electric fields at the discharge center to be predominantly drift fields caused by a low conductivity due to the high electron–neutral collision frequency at atmospheric pressures.[28]

Dust particle manipulation in electrically asymmetric discharges: Based on the Electrical Asymmetry Effect, we have demonstrated a novel scheme for dust transport in radio-frequency discharges. A sudden change of the phase angle between the driving harmonics induces a rapid change in the time-averaged potential distribution in the plasma that in turn induces a transport of dust particles, originally residing near the sheath/bulk boundary of one of the discharge electrodes. The contribution of the Budapest team has been the determination of the time and space dependent discharge characteristics via particle-based simulations.[29,30]

Effect of driving frequencies and number of harmonics on the separate control of ion properties in electrically asymmetric discharges: We have investigated the effect of the fundamental driving frequency on the EAE, i.e. the electrical generation of a dc self-bias, the control range of the mean ion energy, and the ion flux at both electrodes, by PIC/MCC simulations and analytical modelling in dual-frequency capacitively coupled radio frequency discharges driven by two consecutive phase-locked harmonics with adjustable phase shift, θ. A strong decrease in the control range of the electrically induced dc self-bias and the mean ion energies at both electrodes as a function of θ was found at low compared with high fundamental driving frequencies. The control factor of the mean ion energy was found to be decreased from about 1.75 at 27.12 MHz to about 1.2 at 1 MHz. Although the ion flux to both electrodes remains fairly constant as a function of θ at all frequencies investigated, this result means that the EAE cannot be used effectively to control the mean ion energy at low driving

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[25] E. Schüngel, J. Schulze, Z. Donkó, and U. Czarnetzki: "Power absorption in electrically asymmetric dual frequency capacitive radio frequency discharges", Phys. Plasmas 18, 013503 (2011).

[26] J. Schulze, A. Derzsi and Z Donkó: "Electron heating and the electrical asymmetry effect in dual-frequency capacitive CF_4 discharges", Plasma Sources Sci. Technol. 20, 024001 (2011).

[27] J. Schulze, A. Derzsi, K. Dittmann, T. Hemke, J. Meichsner, and Z. Donkó: "Ionization by Drift and Ambipolar Electric Fields in Electronegative Capacitive Radio Frequency Plasmas", Phys. Rev. Lett. 107, 275001 (2011).

[28] T. Hemke, D. Eremin, T. Mussenbrock, A. Derzsi, Z. Donkó, K. Dittmann, J. Meichsner and J. Schulze: "Ionization by bulk heating of electrons in capacitive radio frequency atmospheric pressure microplasmas", Plasma Sources Sci. Technol. 22, 015012 (2013).

[29] S. Iwashita, G. Uchida, J. Schulze, E. Schüngel, P. Hartmann, M. Shiratani, Z. Donkó: "Sheath-to-sheath transport of dust particles in a capacitively coupled discharge", Plasma Sources Sci. Technol. 21, 032001 (2012).

[30] S. Iwashita, E. Schüngel, 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).

frequencies in such discharges.[31] Additionally, we have explored the effect of using more than two harmonics for the excitation of the discharge - we have found that such a driving waveform provides a wider control range for the ion energy, while maintaining a nearly constant ion flux at the electrodes.[32]

A comprehensive summary about these topics have been given in an invited keynote lecture at the 2012 Conference of the Plasma Physics of the European Physical Society.[33]

We note that all the above results have been obtained using spatially one-dimensional codes (which, however, provide a three-dimensional description in velocity space). The development of our two-dimensional PIC/MCC simulation code is in progress.

E. Investigation of the role of primary electrons in radio-frequency discharges

The self-sustainment mechanism of direct current glow discharges is based on two mechanisms. Ionization in the gas phase by fast electrons (traditionally called as the alpha mechanism) as well as emission of primary electrons from the cathode of the discharge due to the impact of different species: positive ions, fast neutrals, and ultraviolet photons (traditionally termed as the gamma mechanism). Radio-frequency excited discharges, on the other hand, may exclusively be sustained by gas-phase ionization, and indeed in most of the modeling studies of RF discharges the gamma mechanism has been neglected. It has not been recognized until recently that electron emission from the electrodes may contribute significantly to the ionization balance. This effect begins to receive much attention nowadays due to the fact that RF plasma sources have important applications in many high-tech fields of industry. Particle-in-Cell simulations can describe both the alpha and gamma mechanisms and thus provide a good basis for such studies.

Our simulation studies, carried out for dual-frequency discharges operated at 27.12 MHz & 1.937 MHz frequencies, have indeed confirmed a completely different behavior of the discharges, depending on the value of the secondary yield. At zero, or low yields a frequency coupling mechanism result in a decrease of the ion flux with increasing value of the low frequency voltage amplitude (V_{LF}). This is explained by the fact that at times when the low-frequency sheath is expanded the superimposed high-frequency sheath oscillations take place with smaller amplitude and velocity (due to the higher ion density further from the electrodes), resulting in a less efficient power deposition to electrons. At high secondary yields, on the other hand, the electrons emitted from the electrodes are accelerated to progressively higher energies when V_{LF} is increased, increasing the power deposition into the plasma.[34] We have also analyzed the effect of secondary electrons for discharges operated under conditions of the EAE.[35]

In a series of experiments we have explored the contemplated appearance of electrical asymmetry when electrodes made of different materials (that have different secondary electron yields) are used in a radiofrequency discharge. Further measurements and accompanying simulation studies will be conducted.

[31] I. Korolov, Z. Donkó, U. Czarnetzki, J. Schulze: "The effect of the driving frequencies on the electrical asymmetry of dual-frequency capacitively coupled plasmas", J. Phys. D: Appl. Phys. 45, 465205 (2012).

[32] J. Schulze, E Schüngel, Z. Donkó and U. Czarnetzki: "The electrical asymmetry effect in multi-frequency capacitively coupled radio frequency discharges", Plasma Sources Sci. Technol. 20, 015017 (2011).

[33] Z. Donkó, J. Schulze, U. Czarnetzki, A. Derzsi, P. Hartmann, I. Korolov, E. Schüngel: "Fundamental investigations of capacitive radio frequency plasmas: simulations and experiments", Plasma Phys. Control. Fusion 54, 124003 (2012).

[34] Z. Donkó, J. Schulze, P. Hartmann, I. Korolov, U. Czarnetzki, and E. Schüngel: "The effect of secondary electrons on the separate control of ion energy and flux in dual-frequency capacitively coupled radio frequency discharges", Appl. Phys. Lett. 97 081501 (2010).

[35] J. Schulze, Z. Donkó, E. Schüngel, U. Czarnetzki: "Secondary electrons in dual-frequency capacitive radio frequency discharges", Plasma Sources Sci. Technol. 20, 045007/1-13, 2011

Summary

The K77653 project has targeted a series of studies on low-temperature and strongly coupled plasmas with the aim of understanding their characteristic phenomena, like elementary reactions, formation of distribution functions, transport processes, collective excitations - based on numerical simulations that are built up from the level of individual particles, thereby complying with the kinetic level. The results obtained with the simulation codes, using different (Monte Carlo, Molecular Dynamics, Particle-in-Cell) approaches have in many cases been presented along, and in strict comparison with the outcomes of experiments and/or theoretical models and calculations. In many topics listed above we have had the pleasure of collaborating with groups in Europe and in the USA, their contributions to the accomplishments presented in this report and the publications have been essential and are gratefully acknowledged.

Besides the planned activities, inspired by emerging collaborations and new ideas we have as well developed new ways of discharge and dusty plasma diagnostics, and also adopted and applied our particle-based simulation codes to investigate the electron kinetics in gas mixtures[36], in the Franck-Hertz experiment[37], in high-pressure pulsed discharges[38], and under the conditions found in the Earth's atmosphere.[39]

Within the duration of the project several invited presentations were given in international conferences, including the 2012 EPS Plasma Physics Conference (keynote lecture), 2009 and 2013 Workshops on Radio Frequency Discharges, 2010 European Conference on the Atomic and Molecular Physics of Ionized Gases, 2010 Gaseous Electronics Conference, and 2013 Central European Symposium on Plasma Chemistry.

We have published 49 research articles in international journals, with a total impact factor of 142.5 (6 of the papers have appeared in Physical Review Letters). A BSc thesis (P. Magyar, ELTE) and an MSc thesis (Á. Budea, ELTE) have been accomplished.

The group of participants have been extended with two young members of our research group (Aranka Derzsi, Ihor Korolov), a PhD student of University of Pécs (Anikó Zsuzsa Kovács) and we had the privilege to host and to have as an official participant of the project, Dr Julian Schulze from the University of Bochum, during the duration of his one-year Humboldt postdoctoral fellowship that he spent in Hungary.

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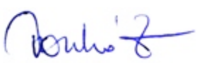

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[36] M. Stano, N. Pinhao, D. Loffhagen, M. Kucera, Z. Donkó and S. Matejcik: "Effect of small admixtures of N_2, H_2 or O_2 on the electron drift velocity in argon: experimental measurements and calculations", Eur. Phys. J. D, 65, 489 (2011).

[37] P. Magyar, I. Korolov and Z. Donkó: "Photoelectric Franck-Hertz experiment and its kinetic analysis by Monte Carlo simulation", Phys. Rev. E 85, 056409 (2012).

[38] Z. Donkó, J. Schulze, S. Mueller and U. Czarnetzki:"Kinetic simulation of a nanosecond-pulsed hydrogen microdischarge", Applied Physics Letters 98, 251502 (2011)

[39] F. J. Gordillo-Vazquez and Z. Donkó: "Electron energy distribution functions and transport coefficients relevant for air plasmas in the troposphere: impact of humidity and gas temperature", Plasma Sources Sci. Technol. 18, 034021 (2009).