

# NON-EQUILIBRIUM CHARGED PARTICLE KINETICS IN IONIZED GASES

NKFIH - 119357

Final Report

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In the frame of the project, we investigated various phenomena that belong to the *forefront of basic research in low-temperature plasma science*. Most of the studies have been based on *joint computational approaches and experimental investigations*, with a few exceptions that have only been approached from the *theory/simulation* side. All the simulation studies have been conducted with *kinetic codes developed by our group*. *Experimental studies have been conducted in both our Laboratory and at collaborating partners of foreign institutions*. The *focus topics* of the project – as laid out in the original research plan – have been:

- A) Tailored voltage waveform radiofrequency (RF) discharges in reactive gases and gas mixtures
- B) Electron power absorption modes and mode transitions in RF plasmas
- C) Resonances, pattern formation and instabilities
- D) Plasma-surface interactions
- E) Plasma response and transport processes

## I. Summary of the achievements

### **I. A) Tailored voltage waveform radiofrequency (RF) discharges in reactive gases and gas mixtures**

*(a) Low-pressure discharges.* Tailored voltage waveforms (TVW) represent a class of driving waveforms of radiofrequency (RF) capacitively coupled plasmas (CCPs), which contain *multiple harmonics of a base RF voltage*. Low-pressure oxygen CCPs driven by such waveforms (specifically, pulse-type (peaks- / valleys-) and sawtooth-type waveforms) were investigated experimentally and by means of kinetic simulations, using principally the Particle-in-Cell/Monte Carlo Collisions (PIC/MCC) technique. During these studies we also analysed the *energy and angular distributions of ions reaching the electrodes* in low-pressure oxygen CCPs. These distributions, as well as the possibility of the independent control of the ion flux and the ion energy have been studied for different types of excitation: *single- and classical dual-frequency, as well as valleys- and sawtooth-type waveforms*. The studies covered conditions from weakly collisional to highly collisional domains of ion transport via the electrode sheaths. Analytical models have also been applied to understand the features of the energy and angular distribution functions.<sup>1</sup> In a separate investigation, the effect of the electrode gap on the discharge properties was addressed. We have obtained a good agreement regarding the *dependence of the electron density and light emission intensity on the electrode separation*, at a fixed pressure. This comparison also served as an *experimental benchmark of our PIC/MCC code for oxygen*.<sup>2</sup> We have discussed the *origin of uncertainties in the results of numerical simulations of low-temperature plasma sources operated in electronegative molecular gases/gas mixtures*, over a wide domain of excitation frequency, voltage, and gas pressure.<sup>3</sup> A *comparison of experimental and simulation results was also accomplished for CF<sub>4</sub>/Ar discharges driven by TVWs*.<sup>4</sup> In a modelling study, we made an attempt to realise pre-defined ion energy distributions (dominated at a single peak at a given energy) by composing the driving waveform from a high number of harmonics.<sup>5</sup>

<sup>1</sup> Z. Donkó, A. Derzsi, M. Vass, J. Schulze, E. Schuengel, S. Hamaguchi: *Ion energy and angular distributions in low-pressure capacitive oxygen RF discharges driven by tailored voltage waveforms*, Plasma Sources Sci. Technol. 27, 104008, 2018

<sup>2</sup> K. H. You, J. Schulze, A. Derzsi, Z. Donkó, H. J. Yeom, J. H. Kim, D. J. Seong, Hyo-Chang Lee: *Experimental and computational investigations of the effect of the electrode gap on capacitively coupled radio frequency oxygen discharges*, Phys. Plasmas 26, 13503, 2019

<sup>3</sup> 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

<sup>4</sup> S. Brandt, B. Berger, Z. Donkó, A. Derzsi, E. Schuengel, M. Koepke, J. Schulze: *Control of charged particle dynamics in capacitively coupled plasmas driven by tailored voltage waveforms in mixtures of Ar and CF<sub>4</sub>*, Plasma Sources Sci. Technol. 28, 095021, 2019

<sup>5</sup> E. Schuengel, Z. Donko, J. Schulze: *A Simple Model for Ion Flux-Energy Distribution Functions in Capacitively Coupled Radio-Frequency Plasmas Driven by Arbitrary Voltage Waveforms*, Plasma Proc. Polymers 14, 1600117, 2017

**(b) High-pressure discharges.** Atmospheric plasma jets are advanced high-pressure plasma sources with a wide range of emerging applications (e.g., plasma biomedicine). The characteristics of such a plasma source operated with RF excitation in a flowing gas mixture of He and N<sub>2</sub> was studied experimentally and via PIC/MCC simulations based on the code developed recently by our group.<sup>6</sup> The *spatially resolved density of He metastable atoms* in this plasma source driven by 'peaks'- and 'valleys'-type tailored voltage waveforms was obtained from our simulations and the results were compared with those determined via *tunable diode-laser absorption spectroscopy (TDLAS) measurements* at the Ruhr University of Bochum. The comparison showed a good quantitative agreement: the density of helium metastables was found to be significantly enhanced by increasing the number of consecutive driving harmonics. It is foreseen that their generation can be further optimized by tuning the peak-to-peak voltage amplitude and the concentration of the reactive gas admixture. A high degree of correlation between the metastable creation rate and the electron impact excitation rate from the helium ground state into the He-I (3s)<sup>3</sup>S<sub>1</sub> level was observed for some conditions, which may open a *possibility for an estimation of the metastable densities based on phase resolved optical emission spectroscopy (PROES) measurements* on the 706.5 nm He-I line originating from this level and known metastable density values at proper reference conditions.<sup>7</sup>

## I. B) Electron power absorption modes and mode transitions in RF plasmas

**(a) Low-pressure discharges.** We studied the *amplitude asymmetry effect as well as the slope asymmetry effect in low-pressure oxygen CCPs* at different fundamental frequencies (5, 10, and 15 MHz) and at different pressures (50–700 mTorr). The spatio-temporal excitation rates derived from *phase resolved optical emission spectroscopy measurements* were compared with the results of our PIC/MCC simulations. *Transitions of the discharge electron heating (electron power absorption) mode from the drift-ambipolar mode to the alpha-mode were found to be induced by changing the number of consecutive harmonics* included in the driving voltage waveform or by changing the gas pressure. Changing the number of harmonics in the waveform was observed to have a *strong effect on the electronegativity of the discharge*, on the generation of the direct current (DC) self-bias and on the control of ion properties at the electrodes, both for pulse-type, as well as sawtooth-type driving voltage waveforms.<sup>8</sup>

*The electron power absorption dynamics has been studied in Ne in a combined experimental (PROES) + simulations (PIC/MCC) study.* We have critically examined the applicability of PROES (which provides information about the spatio-temporal distribution of the electron-impact excitation dynamics) to probe the discharge operation mode in neon (determined by the ionisation dynamics). *The spatio-temporal excitation rates measured by PROES were in a good agreement with the excitation rates obtained from the simulations*, for all the discharge conditions. However, *the ionization dynamics was found to be significantly different from the excitation dynamics for most discharge conditions*, especially at higher driving frequency and lower pressure, when energetic secondary electrons are more likely to ionise than to excite the atoms of the gas.<sup>9</sup>

We have developed a *novel method for the analysis of electron power absorption in low pressure RF plasmas based on the momentum balance equation derived from the Boltzmann equation.*<sup>10</sup> In contrast to conventional theoretical models, which predict "stochastic/collisionless" heating to be important in *low-pressure Ar discharges*, we observed the *dominance of Ohmic power absorption.*<sup>11</sup> A further study in O<sub>2</sub> plasmas indicated an *attenuation of the ambipolar power absorption at low pressures due to the strong electronegativity, and the presence of electropositive edge regions in the discharge*, which cause a high degree

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<sup>6</sup> 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/N<sub>2</sub> mixtures*, Plasma Sources Sci. Technol. 27, 125009, 2018

<sup>7</sup> I. Korolov, M. Leimkühler, M. Böke, Z. Donkó, V. Schulz-von der Gathen, L. Bischoff, G. Hübner, P. Hartmann, T. Gans, Y. Liu, T. Mussenbrock, J. Schulze: *Helium metastable species generation in atmospheric pressure RF plasma jets driven by tailored voltage waveforms in mixtures of He and N<sub>2</sub>*, J. Phys. D Appl. Phys. 53, 185201, 2020

<sup>8</sup> A Derzsi, B Bruneau, A R Gibson, E Johnson, D O'Connell, T Gans, J-P Booth and Z Donkó: *Power coupling mode transitions induced by tailored voltage waveforms in capacitive oxygen discharges*, Plasma Sources Sci. Technol. 26, 034002, 2017; P. Hartmann, L. Wang, K. Nösches, B. Berger, S. Wilczek, R. P. Brinkmann, T. Mussenbrock, Z. Juhasz, Z. Donkó, A. Derzsi, E. Lee, J. Schulze: *Charged particle dynamics and distribution functions in low pressure dual-frequency capacitively coupled plasmas operated at low frequencies and high voltages*, Plasma Sources Sci. Technol. 29, 075014, 2020

<sup>9</sup> B. Horváth, A. Derzsi, J. Schulze, I. Korolov, P. Hartmann, Z. Donkó: *Experimental and kinetic simulation study of electron power absorption mode transitions in capacitive radiofrequency discharges in neon*, Plasma Sources Sci. Technol. 29, 055002, 2020

<sup>10</sup> J. Schulze, Z. Donko, T. Lafleur, S. Wilczek, R. P. Brinkmann: *Spatio-temporal analysis of the electron power absorption in electropositive capacitive RF plasmas based on moments of the Boltzmann equation*, Plasma Sources Sci. Technol. 27, 055010, 2018

<sup>11</sup> M. Vass, S. Wilczek, T. Lafleur, R. P. Brinkmann, Z. Donkó, J. Schulze: *Observation of dominant Ohmic electron power absorption in capacitively coupled radio frequency argon discharges at low pressure*, Plasma Sources Sci. Technol. 29, 085814, 2020

of temporal symmetry of the electron temperature within the RF period.<sup>12</sup> The *effect of an external magnetic field on the operation characteristics of oxygen discharges* has also been addressed in a simulation study. We have found a *significant reduction of the electronegativity of these plasmas due to the enhanced confinement of the electrons in the plasma bulk*. At the same time, *as another consequence of the magnetic field, a strong electric field reversal was found to be generated near each electrode during the local sheath collapse, which locally enhances the electron power absorption*. A model of the electric field generation revealed that the reversed electric field is caused by the reduction of the electron flux to the electrodes due to their trapping by the magnetic field.<sup>13</sup>

**(b) High-pressure (atmospheric) discharges.** A new discharge model and its computational implementation were developed for *atmospheric pressure nanosecond pulsed discharges*<sup>14</sup> and *atmospheric pressure microjet plasma sources*<sup>6</sup>, for the case of operation in helium gas with small admixtures of nitrogen gas. For both systems our studies have revealed *strong kinetic effects, in contrast with the common "belief" that the high collisionality in these systems (consequence of the high pressure) results in hydrodynamic transport of the charged particles*. We have demonstrated the *possibility of controlling the electron energy distribution function, the spatiotemporal distribution of the excitation/ionisation dynamics, and the electron power absorption mode in these high-pressure plasma sources by tailoring of the driving voltage waveform*. In the case of transient discharges, *we have also developed a simulation approach for the description of the transport of the vacuum-ultraviolet resonance radiation of helium and studied the effect of the excitation waveform*.<sup>15</sup>

Using a combination of PROES and kinetic plasma simulations, we have demonstrated that *tailored voltage waveforms consisting of multiple RF harmonics induce targeted disruption of the plasma symmetry*. We have found a confinement of the electron heating to small regions of time and space and an emerging possibility for tailoring the electron energy distribution function in such plasmas.<sup>16</sup> *Tailoring of the excitation waveform was shown to allow for the control of the generation of atomic nitrogen as well as helium metastables*, which are highly relevant for a variety of technological and biomedical applications.<sup>17</sup>

## I. C) Resonances, pattern formation and instabilities

*Self-organized spatial structures in the light emission from the ion-ion capacitive RF plasma of a strongly electronegative gas (CF<sub>4</sub>) were observed experimentally (at Dalian University, by PROES) for the first time*. The formation of this "striated" structure was analysed and understood with our PIC/MCC simulations. The striations were found to be generated by the *resonance between the driving radio frequency and the eigenfrequency of the positive ion – negative ion plasma* (that typically forms in electronegative gas discharges). The growth of the instability was also followed by the numerical simulations.<sup>18</sup> This study was followed by subsequent detailed investigations of the effects of the gas pressure, the radio-frequency driving voltage amplitude, the driving frequency, and the electrode gap, on the striated structure.<sup>19,20</sup> A transition between the striated and non-striated modes was observed by changing either the pressure or the RF voltage; for 13.56 and 18 MHz driving frequencies we computed a phase diagram of the operation mode as a function of the pressure and voltage amplitude parameters.<sup>21</sup>

<sup>12</sup> M. Vass, S. Wilczek, T. Laffleur, R. P. Brinkmann, Z. Donkó, J. Schulze: *Electron power absorption in low pressure capacitively coupled electronegative oxygen radio frequency plasmas*, Plasma Sources Sci. Technol. 29, 025019, 2020

<sup>13</sup> L. Wang, D.-Q. Wei, P. Hartmann, Z. Donkó, A. Derzsi, X.-F. Wang, Y.-H. Song, Y.-N. Wang, J. Schulze: *Electron power absorption dynamics in magnetized capacitively coupled radio frequency oxygen discharges*, Plasma Sources Sci. Technol. 29, 105004, 2020

<sup>14</sup> Z. Donkó, S. Hamaguchi, T. Gans: *The effect of photoemission on nanosecond helium microdischarges at atmospheric pressure*, Plasma Sources Sci. Technol. 27, 054001, 2018

<sup>15</sup> Z. Donkó, S. Hamaguchi, T. Gans: *Effects of excitation voltage pulse shape on the characteristics of atmospheric-pressure nanosecond discharges*, Plasma Sources Sci. Technol. 28, 75004, 2019

<sup>16</sup> A. R. Gibson, Z. Donko, L. Alelyani, L. Bischoff, G. Hubner, 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. 28, 01LT01, 2019

<sup>17</sup> I. Korolov, Z. Donkó, G. Hübner, L. Bischoff, P. Hartmann, T. Gans, Y. Liu, T. Mussenbrock, J. Schulze: *Control of electron dynamics, radical and metastable species generation in atmospheric pressure RF plasma jets by Voltage Waveform Tailoring*, Plasma Sources Sci. Technol. 28, 94001, 2019

<sup>18</sup> Y.-X. Liu, E. Schüngel, I. Korolov, Z. Donkó, Y.-N. Wang and J. Schulze: *Experimental Observation and Computational Analysis of Striations in Electronegative Capacitively Coupled Radio-Frequency Plasmas*, Physical Review Letters 116, 255002, 2016

<sup>19</sup> Y.-X. Liu, I. Korolov, E. Schuengel, Y.-N. Wang, Z. Donko and J. Schulze: *Striations in electronegative capacitively coupled radio-frequency plasmas: Effects of the pressure, voltage, and electrode gap*, Phys. Plasmas 24, 073512, 2017

<sup>20</sup> Y.-X. Liu, I. Korolov, E. Schuengel, Y.-N. Wang, Z. Donko and J. Schulze: *Striations in electronegative capacitively coupled radio-frequency plasmas: analysis of the pattern formation and the effect of the driving frequency*, Plasma Sources Sci. Technol. 26, 055024, 2017

<sup>21</sup> Y.-X. Liu, Z. Donkó, I. Korolov, E. Schuengel, Y.-N. Wang, J. Schulze: *Striations in dual-frequency capacitively coupled CF<sub>4</sub> plasmas: the role of the high-frequency voltage amplitude*, Plasma Sources Sci. Technol. 28, 75005, 2019

## I. D) Plasma-surface interactions

In order to achieve a more accurate description of the operation of CCPs, we have studied the effects of implementing *energy-dependent secondary electron emission coefficients (SEEC) for ions and fast neutrals*, and taking surface conditions of the electrodes into account in PIC/MCC simulations. Simulations were performed using self-consistently calculated effective SEECs, for "clean" (e.g., heavily sputtered) and "dirty" (e.g., oxidized) metal surfaces in single- and dual-frequency discharges in argon, and the results were compared to those obtained by assuming a constant secondary electron yield of 0.1 for ions. In single-frequency (13.56 MHz) discharges operated under conditions of low heavy particle energies at the electrodes, *the pressure and voltage at which the transition between the alpha- and gamma-mode electron power absorption occurs, were found to strongly depend on the surface conditions*. For "dirty" surfaces, the discharge was found operate in alpha-mode for all conditions investigated due to a low effective SEEC. In classical dual-frequency (driven by radio-frequency voltage sources with frequencies 1.937 MHz and 27.12 MHz) discharges the SEEC increased with increasing low-frequency voltage amplitude, for oxidized surfaces. This was identified to be caused by *the effect of the driving voltage on the heavy particle energies at the electrodes, which negatively influences the quality of the separate control of ion properties at the electrodes. The new results on the separate control of ion properties in such discharges indicated significant differences compared to previous results obtained with different constant values of the SEEC.*<sup>22</sup> A theoretical model to determine the SEEC was also developed based on the classical Hagstrum theory on Auger emission. Our ab-initio model was able to *accurately predict the SEEC for metal surfaces with a wide range of surface conditions and for a variety of ion species.*<sup>23</sup>

The effect of the *electron-induced secondary electrons on radio-frequency plasmas operated at high driving voltages* was studied by including a *complex implementation of plasma-surface interactions* in our kinetic simulation codes.<sup>24</sup> Electron-induced secondary electron emission ("delta-electrons") were found to have a strong effect on the ionization dynamics and the plasma density. Due to the high ion energies at the electrodes, very high effective yields of the ion-induced secondary electron emission ("gamma-electrons") were found. At the low-pressure conditions studied, these gamma-electrons did not cause significant ionization directly, since upon acceleration in the high voltage sheaths, these electrons are too energetic to ionize the neutral gas efficiently. *The gamma- and delta-electrons as well as electrons created in the plasma bulk and accelerated towards the electrodes to high energies by reversed electric fields during the local sheath collapse were found to induce the emission of most of the delta-electrons.*

## I. E) Plasma response and transport processes

**(a) Weakly-coupled systems.** We 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* developed in our Laboratory. Depending on the reduced electric field we observed the *equilibration of the swarm over different length scales, beyond which the energy gain and loss mechanism become locally balanced and transport properties spatially invariant*. The evolution of the electron swarm in the apparatus was also described by *Monte Carlo simulation*, of which the results were in good agreement with the experimental observations, over the domains of the reduced electric field and the gas pressure covered.<sup>25</sup> In a *combined theoretical / simulation study we have demonstrated that in partially ionised plasmas, Coulomb scattering can be significantly perturbed by electron collisions with neutral gas particles, and that this effect can be incorporated in the Coulomb collision terms of the Boltzmann equation by a modification of the classical Coulomb logarithm*. We have shown that Boltzmann transport calculations using this modified

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<sup>22</sup> A. Derzsi, B. Horváth, I. Korolov, Z. Donkó, J. Schulze: *Heavy-particle induced secondary electrons in capacitive radio frequency discharges driven by tailored voltage waveforms*, 2019 J. Appl. Phys. 126, 43303, 2019

<sup>23</sup> M. Daksha, A. Derzsi, Z. Mujahid, D. Schulenberg, B. Berger, Z. Donkó, J. Schulze: *Material dependent modeling of secondary electron emission coefficients and its effects on PIC/MCC simulation results of capacitive RF plasmas*, Plasma Sources Sci. Technol. 28, 34002, 2019; M Daksha, A Derzsi, S Wilczek, J Trieschmann, T Mussenbrock, P Awakowicz, Z Donkó and J Schulze: *The effect of realistic heavy particle induced secondary electron emission coefficients on the electron power absorption dynamics in single- and dual-frequency capacitive discharges*, Plasma Sources Sci. Technol. 26, 085006, 2017

<sup>24</sup> B. Horváth, J. Schulze, Z. Donko, A. Derzsi: *The effect of electron induced secondary electrons on the characteristics of low-pressure capacitively coupled radio frequency plasmas*, J. Phys. D Appl. Phys. 51, 355204, 2018

<sup>25</sup> Z. Donkó, P. Hartmann, I. Korolov, V. Jeges, D. Bosnjakovic, S. Dujko: *Experimental observation and simulation of the equilibration of electron swarms in a scanning drift tube*, Plasma Sources Sci. Technol. 28, 95007, 2019

Coulomb logarithm are in excellent agreement, for a sensitive model problem and a wide range of conditions, with particle simulations describing the many-body Coulomb interactions from first principles (see also Sec. II).<sup>26</sup>

We have carried out *measurements of the swarm parameters of electrons* (the bulk drift velocity, the bulk longitudinal component of the diffusion tensor, and the effective ionization frequency) in CO<sub>2</sub>, as well as acetylene, ethylene, and ethane gases in a scanning drift tube apparatus under time-of-flight conditions over a wide range of the reduced electric field. A kinetic simulation of the experimental drift cell allowed estimation of the uncertainties introduced in the data acquisition procedure and provided a correction factor to each of the measured swarm parameters. These parameters were compared to results of previous experimental studies, as well as to results of various kinetic swarm calculations: solutions of the electron Boltzmann equation under different approximations (multiterm and density gradient expansions) and Monte Carlo simulations.<sup>27</sup> In a comparative study we investigated the complementary character of two different experimental setups for electron transport coefficient measurements (Budapest vs. ETH Zürich).<sup>28</sup>

**(b) Strongly coupled systems.** We have accomplished in our Laboratory the *first direct experimental determination of the 3-point static structure function* of a two-dimensional dusty plasma liquid in our dusty plasma apparatus. The measurements were complemented by Molecular Dynamics (MD) simulations of the system, using parameters (dust charge, plasma frequency, coupling and screening coefficients), which were derived from the experimentally obtained 2-point static structure function, as well as the dynamic structure function and current-current fluctuation spectra. *The experimental results of the 3-point structure function were in good agreement with those of the simulations, including the (low wavenumber) domain, where the function acquires negative values.*<sup>29</sup>

*Our MD simulations have verified a new, non-perturbative model-free theoretical moment approach developed for the calculation of dynamical characteristics of strongly coupled classical one-component Coulomb and Yukawa plasmas.* The method allows the derivation of the dynamical structure factor, dispersion, decay, sound speed, and other characteristics of the collective modes from the static structure factor. A good quantitative agreement with the MD simulation data has been demonstrated. Transport characteristics of these systems have as well been addressed: a magnetic field was recently shown to enhance field-parallel heat conduction in a strongly correlated plasma whereas cross-field conduction was shown to be reduced. Now, we have shown that *in strongly coupled 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.*<sup>30</sup>

We have addressed the *peculiarities of the sound speed in a many-body system of charged particles interacting via a pairwise Yukawa potential*, the so-called Yukawa one-component plasma (YOCP), which is a good approximation for a variety of physical systems. It is well known that the collective mode 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 relation defines the sound speed. We studied the evolution of this quantity from the weak- through the strong-coupling regimes by analysing the dynamic structure function in the low-frequency domain. We identified domains in the space of the system parameters in which the *physical behaviour of the YOCP exhibits different features due to the competing physical processes of collective Coulomb-like versus binary-collision-dominated behaviour and the individual particle motion versus quasi-localisation.*<sup>31</sup>

We investigated as well via MD simulations *the propagation of solitons* in a two-dimensional many-body system characterized by Yukawa interaction potential. The solitons have been created in an equilibrated system by the application of electric field pulses. Such pulses generate pairs of solitons, which were found to be characterised by a positive and negative density peak, which propagate into opposite directions. At small

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<sup>26</sup> G. J. M. Hagelaar, Z. Donkó, N. Dyatko: *Modification of the Coulomb Logarithm due to Electron-Neutral Collisions*, Phys. Rev. Lett. 123, 25004, 2019

<sup>27</sup> M. Vass, I. Korolov, D. Loffhagen, N. Pinhão and Z. Donkó: *Electron transport parameters in CO<sub>2</sub>: scanning drift tube measurements and kinetic computations*, Plasma Sources Sci. Technol. 26, 065007, 2017; N. R. Pinhão, D. Loffhagen, M. Vass, P. Hartmann, I. Korolov, S. Dujko, D. Bosnjakovic, Z. Donkó: *Electron swarm parameters in C<sub>2</sub>H<sub>2</sub>, C<sub>2</sub>H<sub>4</sub> and C<sub>2</sub>H<sub>6</sub>: measurements and kinetic calculations*, Plasma Sources Sci. Technol. 29, 045009, 2020

<sup>28</sup> M. Vass, E. Eguz, A. Chachereau, P. Hartmann, I. Korolov, A. Hosl, D. Bosnjakovic, S. Dujko, Z. Donkó, C. M. Franck: *Electron transport parameters in CO<sub>2</sub>: a comparison of two experimental systems and measured data*, J. Phys. D: Appl. Phys. 54, 035202, 2021

<sup>29</sup> Z. Donko, P. Hartmann, P. Magyar, G. J. Kalman and K. I. Golden: *Higher order structure in a complex plasma*, Phys. Plasmas 24, 103701, 2017

<sup>30</sup> T. Ott, M. Bonitz, P. Hartmann and Z. Donko: *Spontaneous generation of temperature anisotropy in a strongly coupled magnetized plasma*, Physical Review E 95, 013209, 2017

<sup>31</sup> L. G. Silvestri, G. J. Kalman, Z. Donkó, P. Hartmann, M. Rosenberg, K. I. Golden, S. Kyrkos: *Sound speed in Yukawa one-component plasmas across coupling regimes*, Phys. Rev. E 100, 063206 (2019), 2019

perturbation, the features propagate with the longitudinal sound speed, from which an increasing deviation was found at higher density perturbations. *An external magnetic field was 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.*<sup>32</sup>

Our MD simulation code has been adopted to the description of multi-layered strongly coupled many-body systems and using this the collective mode structure of a three-layers system has been uncovered. These results (the dispersions and polarizations of the collective excitations) have been compared to the theoretical dispersion relations obtained by applying the quasilocalised charge approximation (QLCA) formalism. By analysing the dynamical longitudinal and transverse current fluctuation spectra we discovered the existence of a structure not related to the collective mode spectra. This also provided insights into the *long-standing problem of the gap frequency discrepancy*, observed in strongly coupled layered systems in earlier studies.<sup>33</sup>

In a simulation study we have addressed the question how the *simultaneous presence of a magnetic field and gas-induced friction modifies the (quasi) localisation of particles* in strongly coupled many-body systems<sup>34</sup> and investigated the effect of the magnetic field on the self-diffusion of particles in an experimental setup with a rotating dusty plasma that emulates extremely strong magnetic fields based on the equivalence of the magnetic Lorentz force and the Coriolis force in the rotating system.<sup>35</sup>

## **II. Research highlights**

(1) We provided an explanation for the appearance of *self-organized spatial structures observed in the ion-ion capacitive rf plasma* of a strongly electronegative gas (CF<sub>4</sub>) using kinetic simulations. These “striations” were found to be generated by the resonance between the driving radio frequency and the eigenfrequency of the ion-ion plasma that was found to establish a modulation of the electric field, the ion densities, as well as the energy gain and loss processes of electrons in the plasma. The growth of the instability was followed by the numerical simulations [PRL 116, 255002 (2016)].<sup>18</sup>

(2) Using Molecular Dynamics simulations, we have verified a novel, *nonperturbative model-free moment approach that was developed for the derivation of the dynamic characteristics* (i.e., the dynamic structure factor (DSF)) of *strongly coupled classical one-component Coulomb and Yukawa plasmas*. The DSF, dispersion, decay, sound speed, and other characteristics of the collective modes were determined using exclusively the static structure factor calculated from various theoretical approaches including the hypernetted chain approximation [PRL 119, 045001 (2017)].<sup>36</sup>

(3) We demonstrated that *in partially ionized plasmas, Coulomb scattering can be significantly perturbed by electron collisions with neutral gas particles, and that this effect can be incorporated in the Coulomb collision terms of the Boltzmann equation by a modification of the classical Coulomb logarithm*. We have shown that Boltzmann transport calculations using this modified Coulomb logarithm are in excellent agreement, for a sensitive model problem and a wide range of conditions, with particle simulations describing the many-body Coulomb interactions from first principles [PRL 123, 025004 (2019)].<sup>26</sup>

## **III. Achievements by students supported by the grant**

During the past years, two students had part-time employment supported by the grant: (1) *Máté Vass* (ELTE Univ.) obtained his MSc degree in 2019 based on his work in our project “*Electron and ion kinetics in radio-frequency oxygen capacitively coupled plasmas*”; the results of his studies have been published in *Plasma Sources Science and Technology*.<sup>12</sup> (2) *Andrea Albert* (ELTE Univ.) conducted research for her BSc degree. She has won a 1st prize at the Students’ Scientific Competition (TDK Conference) at ELTE University in 2020 and a Special Prize at the national level conference (OTDK) in 2021. Her results on “*Monte Carlo simulation*

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<sup>33</sup> H. Pan, G. J. Kalman, P. Hartmann, Z. Donkó: *Strongly coupled Yukawa trilayer liquid: Structure and dynamics*, Phys. Rev. E 102, 043206, 2020

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of resonance effects of electron transport in a spatially modulated electric field in Ar, N<sub>2</sub> and their mixtures” have been published in an article in J. Phys. D: Appl. Phys.<sup>37</sup>; these results will also serve as the basis of her MSc thesis to be submitted in a May.

#### **IV. Public and educational outreach**

Before the coronavirus pandemic, the experiments related to the project have been presented to visitors during Wigner Open Days and International Girls’ Days in which Wigner RCP has been actively participating. Having been invited by the American Journal of Physics, we developed a freely accessible interactive tool<sup>38</sup> for the simulation of two-dimensional Yukawa liquids.<sup>39</sup>

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*The results obtained during the past four years have contributed to the fundamental understanding of low-ionization degree low-temperature plasmas (in terms of the control of distribution functions, power absorption modes and transitions between these, controlling the generation of active species, self-organization, etc.) and many body systems characterized by classical potentials (in terms of structural and transport properties, collective excitations, sound propagation, etc.). Our research has been conducted in a strong collaboration with several foreign institutions (some of which have been official collaborators of the project) in a number of countries (China, France, Germany, Japan, Kazakhstan, Russia, Serbia, Spain, Switzerland, UK, USA). The achievements have been published in 44 research articles in leading scientific journals, with a total impact factor of ~150. The participants have given several invited lectures at major international conferences that resulted in a good visibility of the project at the international scene.*

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Zoltán Donkó, PI

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