Final Report for Award DE-FG02-99ER54554
Kinetics of Electron Fluxes in Low-Pressure Nonthermal Plasmas.

Report period: 8/15/1999 – 8/14/2004

Principal Investigator:
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Post-doctoral Associates:
Dr. Vitaly Schweigert was an expert on computational plasma physics. He was concerned with the modeling activities under this grant. Unfortunately, after being employed 4 weeks on the grant, Dr. Schweigert was diagnosed with a brain tumor and died 6 months after that.
Dr. Konstantin Orlov is an expert on radiofrequency discharges and worked on experimental problems under this project from Oct. 2001-Oct. 2002.

Graduate Research Assistants:
Mike Hebert worked on time-resolved probe measurements of electron fluxes in afterglow plasmas.
He received his M.S. degree in Nov. 2004.
Antonio Maresca worked on probe measurements of electron fluxes.
He received his M.S. degree in Sep. 2001.
Chengbin He worked on Monte Carlo modeling of the positive column problem.
He left for a job at Microsoft before finishing his M.S. degree.

Research Activities:

This grant has focused on the study of several aspects of electron kinetics in low pressure plasmas. Entirely new effects arise from the fact that the electron kinetics is governed by non-local effects, in which the electron distribution function is not equilibrium with the local electric field but is governed by spatial transport effects. In this grant, we were able to demonstrate several previously unstudied effects which are a direct result of the nonlocal transport. These are:

1) The existence of a "convective cell" in electron phase space. The phenomenon was observed and studied in CW plasma conditions.
2) The occurrence of non-collisional cooling of electrons through an effect known as "diffusive cooling."

1) Observation of the "convective cell" in experiments

Based on the results of the convection cell in coordinate-energy space found in the positive column simulations [Kortshagen & Lawler, J. Phys. D: Appl. Phys. **32**, 2737 (1999).], we performed extensive two-dimensionally resolved Langmuir probe measurements of the electron distribution function (EDF) in an inductively coupled plasma (ICP) in order to verify the existence of such a convection cell [J1]. Measurements were performed in Argon for various aspect ratios of the discharge, various RF powers, and different coil configurations. Measurements at constant total energy are

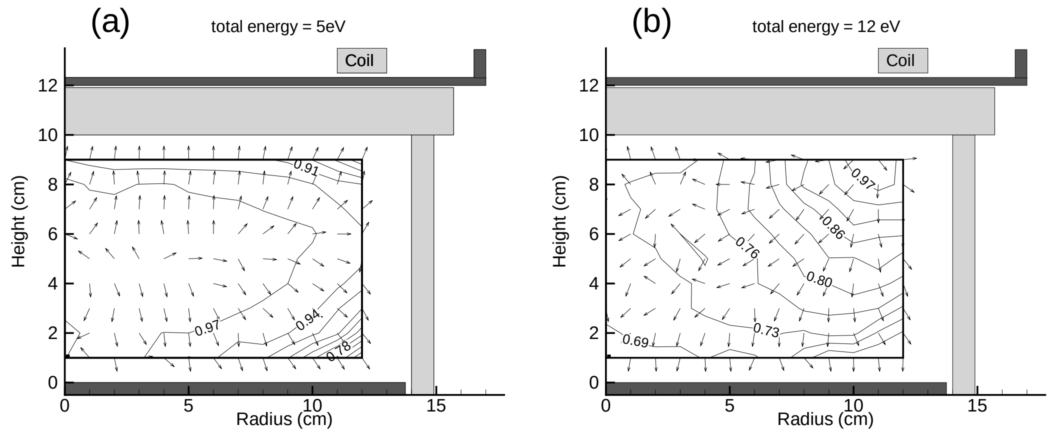

Figure 1: Electron flux density in an Argon ICP at (a) 5 eV and (b) 12 eV total energy.

particularly simple with Langmuir probes, since the negative probe voltage scale is equal to the total energy up to an additive constant. From two-dimensional measurements of the EDF at constant probe voltage and computations of the gradients of the measured profiles, we were able to derive the electron flux patterns in coordinate space at constant total energy.

Figure 1 shows two electron flux density patterns which were measured for the same plasma conditions at different total energies. Again, the vector arrows only represent the direction of the electron flux. It can be seen that at the lower total energy of 5 eV the electron flux density is directed from the central region of the discharge, which corresponds to the maximum plasma potential or the “bottom” of the potential well, towards the periphery. At the higher total energy of 12 eV, the maximum of the EDF has shifted closely towards the coil position inducing an electron flux towards the discharge center. This flux pattern bears close similarity to the “convection cell” in configuration-total energy space observed in the positive column simulation.

The electrons experience the strongest heating in the region close to the induction coil, where the RF electric field is strongest. The region close to the induction coil can thus be considered as a source of energetic electrons. For the above experiment, in which the plasma potential is maximum in the discharge center, the space charge electric field preferentially draws the electrons towards the center. Hence for given total energy, the maximum of kinetic energy is at the center. Due to the monotonous increase of most excitation and ionization cross sections with kinetic energy close to their thresholds, the discharge center also is the location of the maximum of ionization and excitation processes. This region thus corresponds to a sink for energetic electrons and a source for low energy electrons. The spatial separation of the maximum of inelastic processes and the maximum of the electric field results in the observed flux pattern of electrons: Low-energy electrons are produced in the central region of the discharge and diffuse towards the periphery. Close to the coil, they experience strong heating and are “lifted” to higher energies. For these high energy electrons the sink region in the discharge center induces the inward directed electron flux. There, the “convection cell” is closed by inelastic collisions that transform high-energy into low-energy electrons.

With our two-dimensional model based on the solution of the nonaveraged Boltzmann equation [J2] the exact same flux pattern could be found in the simulations. The model indicates that for our experimental conditions, a typical electron goes through the "convection loop" more than ten times before reaching the discharge wall. This can be concluded from the electronic excitation frequency being more than ten times higher than the ionization frequency, which equals the electron loss frequency in a steady-state plasma.

2) Observation of the "diffusive cooling" in experiments

A second focus of our studies was the investigation of the time-evolution of the electron distribution function in the afterglow of a pulsed plasma [J2,J3]. For this purpose, we performed a systematic study of time-resolved probe measurements in the afterglow of an inductively coupled Argon plasma. Our discharge system used a Pyrex chamber with 28 cm diameter, and a height of 10 cm. Pressures were between 5 and 70 Pa. Probe measurements were performed with a 5 mm long, 0.254 mm diameter Tungsten cylindrical probe. Our measurement procedure consisted of taking time-resolved current-time samples $I_p(t)$ at constant probe voltage, repeating this procedure for various probe voltages, and then cross-converting the data set into a complete time series of current-voltage characteristics $I_p(V_p)$ for the entire afterglow. From the probe characteristics, the EDFs were determined using the well-known Druyvesteyn method and numerical double differentiation.

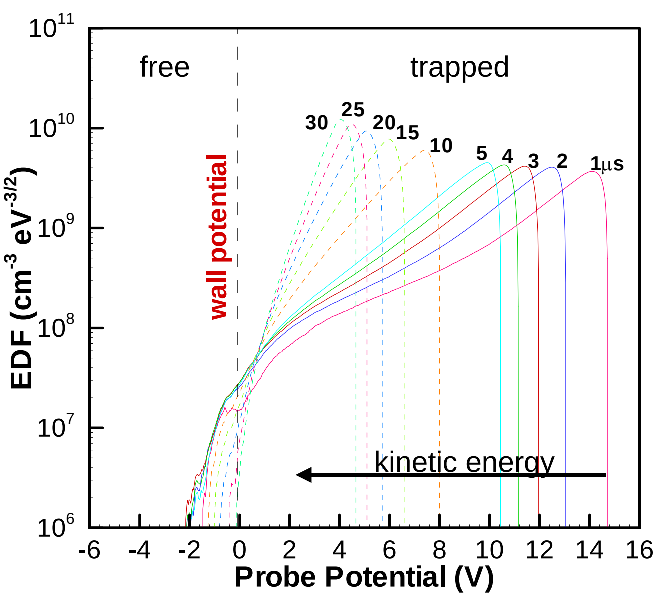

Figure 2: Measured EDFs at different times in the afterglow of a 15 mTorr Argon plasma.

Figure 2 shows a set of measured EDFs at different times in the afterglow. For physical clarity, we displayed the EDF as functions of the probe voltage rather than the more commonly used form of kinetic energy. The reason for this choice is that the wall potential, which plays the role of dividing the EDF into trapped and free electrons, remains fixed in this representation. Since we referenced our probe to a grounded metallic wall, the wall potential for all times in the afterglow is at 0 V. The zero of kinetic energy of electrons is given by the right-hand zero crossing of the EDF, i.e. at 14.8 V for the $1\mu s$ EDF. From this voltage, the kinetic energy is increasing towards the left—the more negative voltages—with the range between the zero crossing and 0V representing the trapped electrons, and the range of voltages less than 0V representing the free electrons. It is clearly seen that the EDFs at all times in the afterglow show a significant change in slope at the boundary between free and trapped electrons. In the free electron range, the EDF drops much faster than in the trapped electron range, indicating that the wall loss of electrons leads to a rapid depletion of the EDF at higher electron energies. To our knowledge, these measurements are the first measurements that clearly identify the wall potential as the EDF "cut-off" threshold [J3]. However, it should be noted that a similar behavior can be seen in the probe measurements of Singh and Graves [J. Appl. Phys., 2000. **88**(7): p. 3889-3898] that were performed in a low-pressure ICP steady state plasma in various molecular gases. The authors observed a clear sharp drop in the EDF at energies much higher than the collisional thresholds which is likely caused by electron wall loss.

The collisionless heating of electrons leads to a peculiar behavior of the electron cooling timescale (or energy relaxation time). While one would expect that for a collisional plasma the energy relaxation time decreases with increasing pressure, exactly the opposite was observed in our case of a low pressure plasma [J3]. The reason for this behavior is that the cooling of electrons is based on a collision-less wall loss mechanism, which is actually slowed down by an increased collisionality of the plasma, since the ambipolar electron and ion loss is slowed down by an increase in pressure.

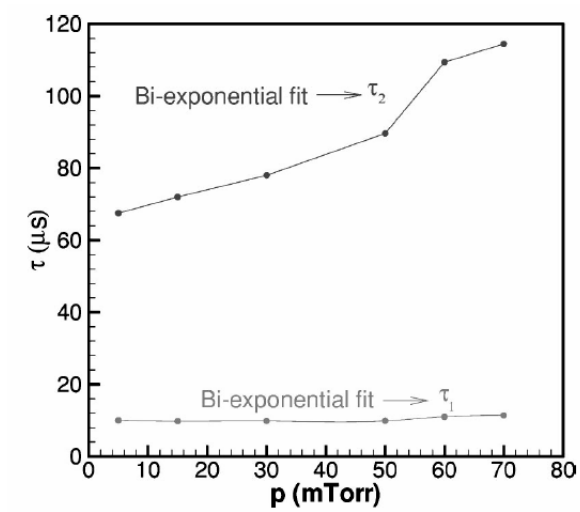

Figure 3: Electron energy decay time as function of the pressure: τ_1 denotes the initial fast decay of the temperature, τ_2 the decay in the later afterglow.

Finally, we report results of our attempts to confirm the existence of subcooled electrons using Langmuir probe measurements in a pulsed Argon ICP [J4]. In principle, when the wall loss of electrons is a very fast process and the thermal contact of the electrons to the neutral gas is poor, it should be possible to achieve electron temperatures lower than the gas temperature. For instance, electron temperatures as low as 30 K were claimed by Biondi, based on rather indirect measurements of the density decay in a microwave resonator [Phys. Rev., 1954. **93**: p. 1136.]. We performed time resolved probe measurements and evaluated those in two different ways: 1) The electron temperature was obtained from a semilog plot of the electron current in the electron retardation regime after subtracting the ion current contribution, and 2) by fitting a simulated probe characteristic using the Orbital Motion Limited (OML) theory after Laframboise [*4th International Symposium on Rarefied Gas Dynamics*. 1964. Toronto: Academic Press, New York] to the measured characteristics. Figure 4 shows the results that were obtained with these two methods. It is obvious that deviations are significant in that method 1) yields electron temperatures lower than the gas temperature in the late afterglow, indicating

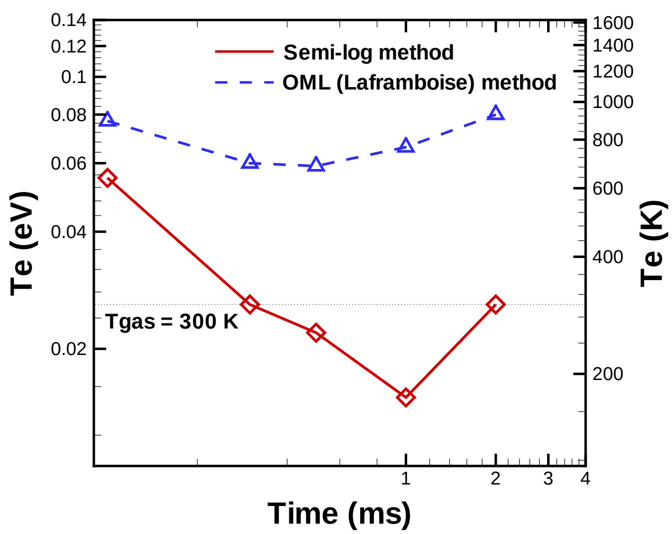

Figure 4: Comparison of Langmuir probe measurements of the electron temperature in a pulsed Argon ICP at 20 mTorr. Semi-log method refers to T_e being computed from a linear regression of a semi-logarithmic plot of the electron retardation current.

subcooled electrons, while method 2) does not. It is likely that the OML theory is not strictly applicable in the late afterglow since it only applies to situations where the probe sheath is much thicker than the probe radius. However, the very low electron temperatures in the late afterglow likely lead to a thin sheath. Unfortunately, our experience shows that the temperatures obtained from the semi-log method also depend sensitively on how the ion current is subtracted from the probe current. Figure 4 thus demonstrates the significant difficulty involved in studying subcooled electrons with Langmuir probes.

Publications and Presentations that resulted from this grant:

Journals:

J1. "Exerperimental Observation of a ``Convective Cell" in Electron Phase Space in an Inductively Coupled RF Plasma," U. Kortshagen and B. Heil, Appl. Phys. Lett. **77**, 1265 (2000).

J2. "Recent progress in the understanding of electron kinetics in low-pressure inductive plasmas," U. Kortshagen, A. Maresca, K. Orlov, and B. Heil, Appl. Surf. Sci. **192**, 240 (2002).

J3. "Experimental study of diffusive cooling of electrons in pulsed inductively coupled plasma," Antonio Maresca, Konstantin Orlov, and Uwe Kortshagen, Phys. Rev. E 65, 056405 (2002).

J4. "Experimental study of diffusive cooling and the question of "subcooled electrons" in pulsed inductively coupled Argon Plasmas," M. Hebert and U. Kortshagen, Plasma Sources Science and Technol., in preparation.

Book Articles:

B1. "Recent progress in the understanding of electron kinetics in low-pressure inductive plasmas," U. Kortshagen, A. Maresca, K. Orlov, and B. Heil, in "Advances in low temperature RF Plasmas: Basis for Process Design," ed. T. Makabe, North-Holland, Amsterdam, NL, p. 244-256, 2002.

Invited conference lectures:

L1. "Electron Kinetics in Low-Pressure Discharges," Cordon Conference on Plasma Science, Tilton, NH, Aug. 13-18, 2000.

L2. "Electron Kinetics in Low-Pressure Discharges," International Workshop of Electron Kinetics, Greifswald, Germany, Sep. 14, 2000.

L3. "Low Temperature ICP Modeling on the Basis of the Boltzmann Equation," International Workshop on the Basis for Low Temperature Plasma Applications, Hakone, Japan, July 23-25, 2001.

Conference Contributions:

C1. "Particle in Cell Monte Carlo Collision Modeling of Inductively Coupled Plasmas," V. A. Schweigert, U. R. Kortshagen, 2000 IEEE International Conference on Plasma Science, New Orleans, Louisiana, June 4-7, 2000, p. 165.

C2. "Computation and Measurement of Electron Circulation Patterns in Phase Space," U. R. Kortshagen, B. Heil, 2000 IEEE International Conference on Plasma Science, New Orleans, Louisiana, June 4-7, 2000, p. 166.

C3. "Electron Kinetics and Self-Consistent Modeling of High Current Positive Column Plasmas," U. Kortshagen, J. D. Michael, and J. H. Ingold, Bulletin Amer. Phys. Soc. **45**, No. 6, p. 62 (2000).

C4. "Comparison of Experiments and Self-Consistent Monte Carlo Simulations of Helium Positive Column Discharges," J. E. Lawler and U. Kortshagen, Bulletin Amer. Phys. Soc. **45**, No. 6, p. 62 (2000)

C5. "Particle Nucleation in Silane Plasmas," U. Bhandarkar, U. Kortshagen, S. L. Girshick, and U. Kortshagen, Bulletin Amer. Phys. Soc. **45**, No. 6, p. 64 (2000).

C6. "Two-dimensional Particle-In-Cell Monte Carlo Simulation of High Density Plasma Sources," V. A. Schweigert and U. Kortshagen, Bulletin Amer. Phys. Soc. **45**, No. 6, p. 42 (2000).

C7. "Study of the evolution of the electron energy distribution function in the afterglow of low pressure inductively coupled plasmas," Antonio Maresca, Konstantin Orlov and Uwe Kortshagen, 15th International Symposium on Plasma Chemsitry, Orléans, France, July 9-13, 2001, ed. A. Bouchoule *et al.*, Gremi, CNRS-Université Orléans, VIII-3207

C8. "Evaporative cooling"of electrons in the afterglow of low pressure plasmas, Antonio Maresca, Konstantin Orlov, Uwe Kortshagen, Bull. American Physical Soc. **46** (6), 56 (2001).

C9. "Experimental study of diffusive and collisional cooling of electrons in a pulsed inductively coupled plasma," Michael Hebert, Uwe Kortshagen, Bull. American Physical Soc. **47** (7), 45 (2002).

C10. "Measurement of cold electrons in a pulsed inductively coupled plasma," Michael Hebert, Uwe Kortshagen, Dirk Urlandt, Bulletin of the American Physical Society **48** (6), 2003, p. 18.