Using Swarm Models as an Exact Representation of Ionized Gases

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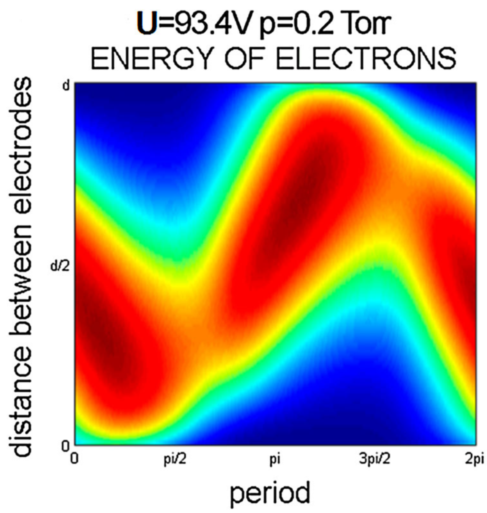
In this review, several examples of ionized gases are presented where swarm models may be employed to provide full description. Those situations include low space charge pre-breakdown, Townsend region breakdown where space charge effects may be calculated from the swarm model and used as the first order perturbation to describe oscillations and transient signal and afterglows. In addition, implications are considered for micro-discharges, discharges in and close to liquids, gas-filled particle traps, thermalization of particles in living tissue, and many more. In all those situations, swarm models provide full description of the discharge, while for most other collision dominated non-equilibrium plasmas swarm physics (transport-related phenomena) provides a part of the foundation of modeling.



1. Introduction

Swarm data and the basic transport equations have been the foundation of the modeling of low temperature (i.e., non-equilibrium) plasmas.[1–4] In doing so, it is often assumed that the transport data obtained under such conditions fit well the fluid or other equations used to model plasmas. Without going into discussion of whether that is the case or not, we need to stress that the use of swarm data or of the swarm derived cross-section sets[5–7] is a prerequisite in achieving proper energy, momentum, and number balances in plasma models and in having properly calculated non-equilibrium distribution functions. Even though in RF fields and in the presence of strong variations of the distribution function, the use of swarm parameters may become complex due to non-locality[8–10] these data have been used successfully and with little evidence of inadequacy. That is presumably due to a robust nature of plasma models (physics) dictated primarily by the space charge adjustment that provides a field distribution necessary to maintain the existence of the plasma itself.

In this paper, we shall, however, focus on the ionized gases where the swarm models are an exact representation of the system, as exact as the available data allow it and as exact as small perturbations of the external field do not constitute a major source of the relevant particles. Usually it is assumed that by cornering yourself into the low current limit and favoring situations where ionized gas does not use its ability to self adjust the field profile, will lead to very few, if any (with exception of swarm experiments of course), examples where such physical models is adequate. That, luckily, is not true and we have a number of examples where swarm models provide sufficient and even complete description. This paper attempts to provide a review of such examples and also to

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provide an insight on how swarm models may be used as plasma models. This work is naturally primarily focused on the results of our group.

1.1. What Are Swarms?

Swarms have been defined as ensembles of charged (although the paradigm may be extended beyond charge) ensembles of particles freely moving through the background gas, gaining energy from the external electric field, being under the influence of the external magnetic field, and dissipating energy and momentum in collisions with the gas molecules. It is assumed that all collisions are with the pristine, unperturbed gas and also that space charge effects and Coulomb coupling are both negligible. In other words, it is the zero ionization limit of plasmas where collisions reign supreme and where the field is "known," i.e., defined by the external voltage. The behavior of charged particles is defined by collisions and also by the energy gain and the field configuration. Thus, this is the end where atomic and molecular physics, integrated into the transport theory, and overall kinetic calculations dominate.

An additional condition is also often introduced that boundaries (such as metallic walls, sometimes grounded, sometimes at some potential) are not felt throughout most of the volume of the discharge. That, however, is not absolutely necessary, it is merely there to provide the basis for the so called hydrodynamic expansion that allows us to separate the distribution function into a velocity space distribution (f) multiplied by the real space particle density profile ($n(\boldsymbol{r},t)$) by using spatial gradients of the density:

$$f(\boldsymbol{r},\nu,t)=\sum_{k=0}^{\infty}f^{(k)}(\nu)\otimes(-\nabla)^{k}n(\boldsymbol{r},t) \tag{1}$$

and spherical harmonics:

$$f(\nu)=\sum_{l=0}^{\infty}f_{lm}(\nu)P_{l}^{|m|}(\cos\,\theta)e^{-im\varphi}, \tag{2}$$

where $P_l^m(\cos\,\theta)$ are Legendre polynomials and θ and φ are polar angles.

Very important aspect of this expansion, that allows us a much easier numerical solution to the Boltzmann equation, is the fact that if it is satisfied we effectively assume the distribution function to be uniform throughout the entire volume of the discharge. The standard swarm experiments, and swarm physics have always been strongly associated with well-defined experiments, that are able to achieve such conditions throughout most of the volume of the discharge. To do so, a combination of the use of high pressure and control of the current density is required. In any case, substituting Equation (1) and (2) into Boltzmann equation:

$$\frac{\partial f}{\partial t}+\mathbf{c}\cdot\frac{\partial f}{\partial \mathbf{r}}+\frac{q}{m}(\mathbf{E}+\mathbf{c}\times\mathbf{B})\cdot\frac{\partial f}{\partial \mathbf{c}}=-J(f,F_0) \tag{3}$$

where $J(f,F_0)$ is the collision operator, provides the means to define and calculate the transport (swarm) coefficients.

On the other hand, most low temperature plasmas operate under conditions where hydrodynamic approximation may not be appropriate. Those include low pressure discharges, where mean free path may be comparable to the size of the vessel, sheaths, and electrode regions and also high gradient areas, like those found in the front of the streamers, thermalization from the initial distribution, or the very high E/N conditions, when charged particles may achieve a runaway. Thus we shall apply swarm models in both sets of circumstances allowing for non-hydrodynamic conditions when required.

1.2. Swarm Models

A swarm model would be a model based on a Boltzmann equation (BE),[11–14] Monte Carlo simulation (MCS),[15–18] or some of the simplified equations such as the Momentum transfer theory (MTT).[19–21] More than actual modeling, one may use transport data to directly calculate properties in hydrodynamic region. If, however, it is not hydrodynamic then the ionized gas should perhaps be modeled by MCS.

An important feature of swarm models is that those are often approximate, like MTT in general, or BE if only two terms are maintained (in expansion given by Equation (2)) or some model collisional operator is employed. Verifying exact nature of the model is thus an important issue and for that purpose, swarm benchmarks are often employed.[15,18,22–25] We will not spend more time on this issue as it has been well covered by a number of papers.[26]

We will, however, define conditions where swarm models are expected to be appropriate and then proceed to illustrate some, such as:

- Low space charge density ionized gas in general like the charges in the atmospheric gas.
- Pre-breakdown avalanches requiring external field but not quite making it to the self-sustained regime.
- Breakdown where the initial phase and the transition to the self-sustained regime are in swarm regime while the final stage may be a fully developed plasma, and thus the conditions for the breakdown are defined by the swarm regime.
- Gaseous dielectrics are also defined by the operation in the swarm regime as their use is to prevent development of the plasma in the first place.
- Gas-filled traps such as Penning Malmberg Surko trap for positrons.

- Detectors of elementary particles starting from Willson's chamber and Geiger counter, through drift and avalanche chambers and finally including the most frequently used resistive plate chambers (RPC) detectors.
- Low current diffuse discharge (Townsend discharge) that operates in the low space charge limit, although even when space charge starts making the entrance it is usually as a perturbation to the swarm model.[27–29]
- Afterglows, after the collapse of the ambipolar field.
- Thermalization of elementary particles emitted from radioactive sources or of cosmic rays and of their secondary products, and many more.

2. Pre-Breakdown, Free Electrons in Ionized Gases

Pre-breakdown, or transport of charged particles in field free conditions or in fields too weak to achieve self-sustained operation have been studied for many years. For obvious reasons, the primary target of such studies has always been modeling of swarm experiments that have been designed to provide high accuracy without uncertainties in interpretation so that the measured transport coefficients may be used to normalize the sets of cross-sections. It is important to say that modeling may be done in the real space and thus provide the connection between observables and actual transport coefficients under the study. Modeling may also be in the velocity space where calculating data is easier and then one comes to the point when it is possible that due to differences between the real space (bulk) and velocity space (flux) transport coefficients one needs to actually model non-conservative aspects of the complete transport in the experiment in order to fit the measured observables and the resulting transport data.[8,9] In a basic swarm model in weakly ionized gas, would be the use of an equation:

$$j = e\ n\ v_{dr} \tag{4}$$

where j is the current density, v_{dr} the electron drift velocity, and n is the charged particle density together with the swarm data for the drift velocity. The spatial and temporal profiles of swarms are usually described in hydrodynamic approximation by using the so called continuity equation[30]:

$$-\frac{\partial n}{\partial t} + (\nu_i - \nu_{att})n - v_{dr}\frac{\partial n}{\partial z} + D_T\left(\frac{\partial^2 n}{\partial x^2} + \frac{\partial^2 n}{\partial y^2}\right) + D_L\left(\frac{\partial^2 n}{\partial z^2}\right) = 0 \tag{5}$$

where D is the diffusion coefficient (which may be either transverse (T) or longitudinal (L)) and non-conservative rate coefficients are ν (ionization − i and attachment − att). Equations such as 4 and 5 have been used successfully to model a number of experiments[30–32] provided that hydrodynamic approximation holds. Best examples of the validity of this approach and the use of continuity equation may be observed in photon emission profiles of time of flight experiments of Blevin and Fletcher.[31,33] In some way, these profiles are akin to the detected profiles of avalanches by Raether[34] in gaseous elementary particle detectors (see also ref.[35] and discussion of detectors later on in the paper).

When, however, assumptions going into the hydrodynamic approximation are not valid,[9] then a door is opened for kinetic effects such as diffusion or attachment heating or cooling,[36–40] transient negative mobility,[41–43] negative absolute mobility,[43–47] anomalous time varying diffusion,[48,49] negative transient diffusion,[50] Holst Oosterhuis (Frank Hertz) luminous layers,[51–53] negative differential conductivity (NDC),[20,54] and many more. A group of kinetic phenomena may occur even when hydrodynamic conditions are not met and transport is not local.

As an example of how we may use swarm physics to get an insight about the functioning of a device, we may use a display of NDC to discuss its role in gas-filled (diffuse discharge) switches. In Figure 1, we show drift velocity of electrons in pure CF_4 where the most prominent feature is a peak around 20 Td. The region beyond the peak where drift velocity counter intuitively decreases is the NDC.

One class of devices, the so called diffuse discharge switches, were developed to control inductive storage of energy. Apparently, the power density in inductive discharges is two orders of magnitude greater than the power density of capacitive storage, which proved to be essential for applications in space. Unlike capacitive storage switch, the switch for inductive storage requires a high conductivity at low E/N and low conductivity at high. Thus it has been possible just to use the calculated drift

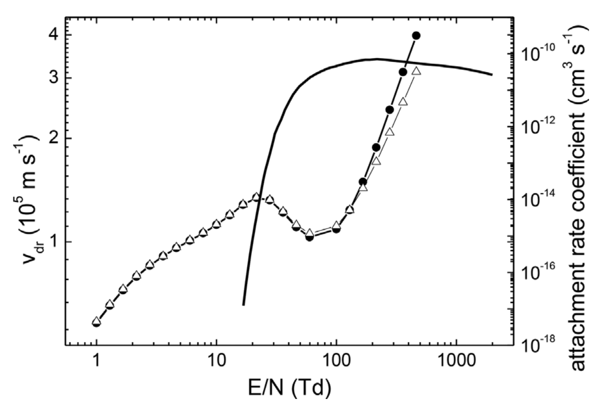

Figure 1. Drift velocity v_{dr} (points triangles flux, solid circles bulk) and attachment rate (line) for electrons in pure CF_4.[55] The drift velocity shows NDC from 20 to 60 Td while the attachment rate peaks at around 120 Td.

velocity versus E/N to select the best candidates for practical devices.[56,57]

2.1. DC Breakdown

In studies of the breakdown, the self-sustained discharge is achieved when production exceeds the losses. For electropositive gases, losses are difficult to calculate and depend strongly on the geometry of the discharge. In case of electronegative gases attachment dominates the losses and as it is a gas phase process, it is easily established in general terms. Thus, one could claim that the breakdown E/N (which defines the breakdown voltage for a specific geometry) is that where ionization rate becomes greater than the attachment rate; or when the effective multiplication coefficient $(\nu_i - \nu_{att})$ (or represented in spatial Townsend rate coefficients $(\alpha - \eta)$) becomes zero. Actually it has to be larger than zero to compensate other geometry dependent losses, but zero point is a good indicator of the dielectric strength of the gas (mixture). In Figure 2, we also show effective multiplication rate coefficient for tetrafluoroethane mixtures with argon.[58,59]

One can see in Figure 2 that for a low content of the electronegative gas (5%), the breakdown E/N is smaller, around 25 Td. For 50%, it is more than 70 Td and for pure tetrafluoroethane it is more than 110 Td. Thus, having in mind a cross-section set for a good gaseous dielectric, it is required that there is an attachment process (dissociative presumably) peaking at high energies just below the threshold for ionization in order to postpone to higher E/N the predominance of production over losses. Good dielectrics, however, also have an attachment (scavenging process) that would peak at the lowest energies and thus remove low energy electrons before they have a chance of accelerating to higher energies. SF_6 is such a gas with several non-dissociative and dissociative attachment processes covering continuously energies from zero to just

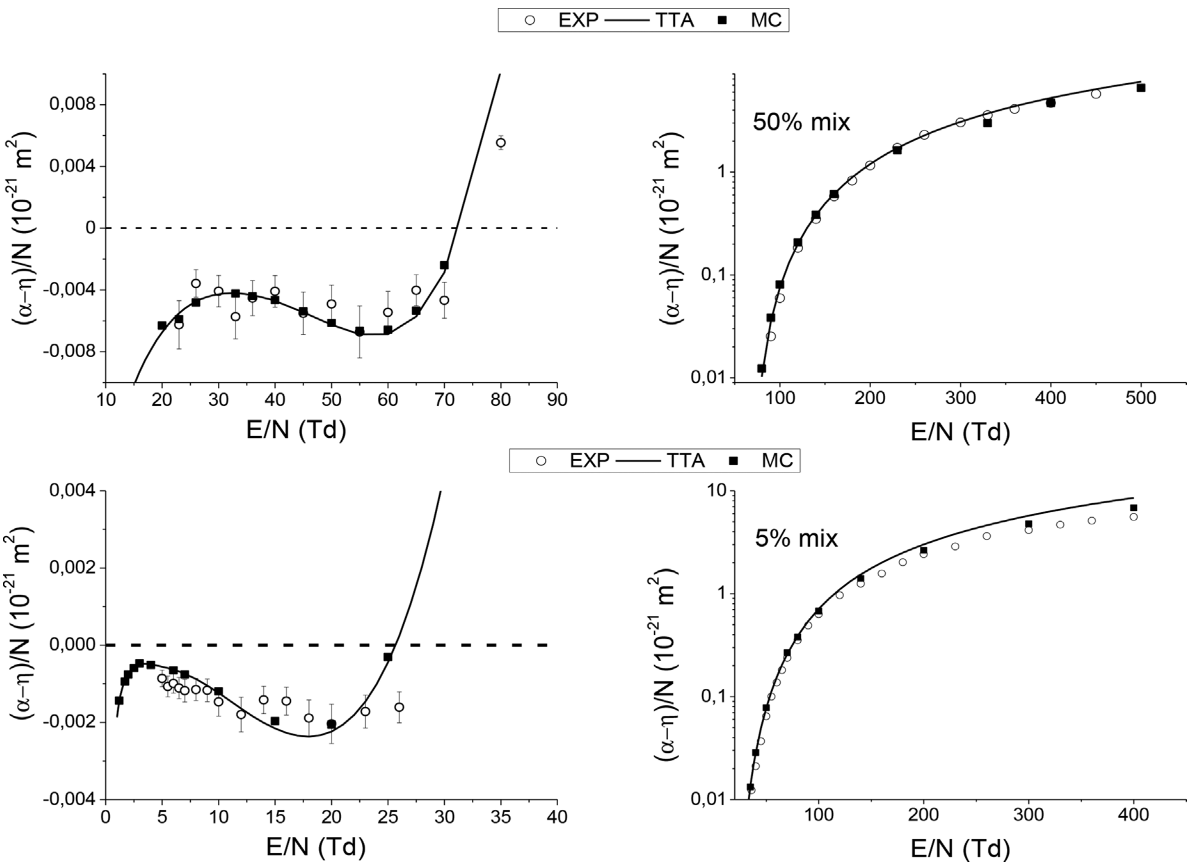

Figure 2. Effective multiplication rate for the mixture of tetrafluoroethane (5 and 50%) and argon. As we had to show the ionization rate (denoted by α) in a logarithmic plot, the negative part due to the attachment rate (denoted by η) is shown separately on a linear scale.[58,59] MC denotes results of a Monte Carlo simulation, while TTA is an abbreviation for the two term approximation theory. Both sets of calculated results are compared to experiment. The cross-sections were obtained by employing a standard swarm procedure to the available experimental transport data.[58,59] The principal thing that should be observed here is that for gases with large attachment, the breakdown E/N may be easily established from the calculated ionization and attachment rates as a crossover point. For many application, such analysis is sufficient for making engineering decisions.

below the ionization. Other issues in developing a good dielectric would certainly include plasma chemistry that ensues after a possible breakdown. It is preferable that the original molecule is re-formed to ascertain the longevity of the dielectric, and should the gas from the dielectric be released to the atmosphere, toxicity of products of a discharge may become an issue. Except for the plasma chemistry calculations of the dissociative effects, all other aspects of gaseous dielectrics may be modeled by a swarm model or related zero dimensional chemical kinetics models.

Similar issues need to be resolved in gas discharge switches except that the conducting phase involves high conductivity plasma that may (or may not) require a full plasma model. Yet the breakdown itself, a critical feature of the switch, is established easily through swarm modeling. Returning to the diffuse discharge opening switch, the basic principle involving NDC in gaseous mixture may be improved upon by adding a gas with a high threshold attachment that would reduce further the conductivity at the high (off phase) E/N, but in this case without the low energy attachment (as that would be a hindrance). CF_4 would be excellent for this purpose (as shown in Figure 1).

The issue of breakdown in general terms is also related purely to swarm modeling and it has been covered in some detail by Phelps and Petrović.[60] The standard Townsend model of breakdown consists of an electron-induced avalanche and ion feedback producing new electrons at the cathode by secondary emission. It turned out[60] that actually photo emission at low E/N, fast neutrals at high, and metastables at all E/N contribute significantly to the breakdown, sometimes even dominating. Adding to this the loss processes not accounted for in the Townsend model, such as back-diffusion, we may actually have an effective secondary electron yield obtained from the breakdown curves (according to the Paschen's theory) one order of magnitude lower or up to two orders of magnitude higher than the secondary electron yield measured in a binary beam experiment.[60] Townsend's theory worked basically because in the SWARM regime, all fluxes are proportional (linearly) and thus the effective production could be associated with the flux of only one particle. However, in real plasmas with non-linearities and especially temporal developments the basic Townsend theory may not be adequate and thus analysis akin to that of Phelps and Petrović (that is still a swarm-type modeling) may be in order.

2.2. RF Breakdown

Basic data for radio-frequency plasma applications can be acquired from simulations and experimental results, and from recorded breakdown voltage curves. DC breakdown voltages versus *pd* (*p*-pressure, *d*-gap between two parallel electrodes) are known as Paschen curves. For RF breakdown, *pd* scaling may be expected to work again but the curves, although similar in shape, are not determined by the Paschen law. Nevertheless, these curves are often called Paschen curves in the literature while it is better to call them RF breakdown curves or sometimes even Paschen-like RF breakdown curves.

One should never attempt to determine secondary electron yields from RF breakdown curves in a direct manner, at least for the following reason. A necessary condition for a self-sustained discharge is to have feedback between the electron growth toward instantaneous anode and their initialization at the cathode. In DC breakdown, it is the drift of ions toward the cathode coupled with a secondary electron production that provides the feedback. In RF fields, however, electrons go in both directions, depending on the phase, so a discharge may be supported purely by electrons. We have performed calculations with only electrons and also with added heavy particles, ions, and fast neutrals.

Initially, electrons were released from the middle of the gap. Cross-section sets have been compiled and tested (argon, oxygen – Itikawa[61]; synthetic air – Phelps[62]). In our previous paper, we have examined radio-frequency breakdown in argon under conditions when ion-induced secondary emission is negligible (electron-dominated regime).[63] In this paper, we move further by including ions and their contribution to secondary electrons emitted from electrodes surfaces. Breakdown points are determined by slowly increasing the voltage to approach the breakdown from below the curve (right hand side and lower branch of the curve) and by increasing pressure to approach higher breakdown voltage branch (left hand side). Breakdown point is established as the one where the number of electrons begins to increase over extended time of many periods (detailed discussion is given in ref.[63]).

At first, we examine MC simulation that includes only electrons with distance between electrodes of 1 cm. Figure 3 shows RF breakdown curves for synthetic air. Furthermore, we adjust two parameters to try to fit the experimental data, the first being the reflection coefficient for electrons on the surface of electrodes (R) and also the secondary electron yield γ (gamma) due to ion bombardment. One can observe deformation of Paschen-like curve pushing breakdown point toward lower voltages when reflection coefficient is increased arbitrarily. However the "second minimum," as Korolov et al.[65] obtained in their experiment (also shown in Figure 3), is only achieved through an addition of the effect of secondary electron emission due to ions. Multipacting effects are observed only at much lower pressures and higher voltages. A good agreement with Korolov et al. was achieved by an assumption that secondary electron yield for ions is 0.002.

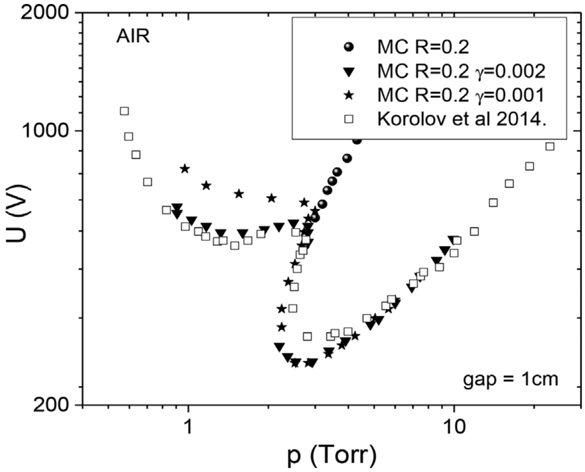

Figure 3. Paschen-like RF breakdown curves in synthetic air.[64] Gap is 1 cm, frequency 13.56 MHz. Open points are the available experimental results in the literature.[65] At first, only reflection of electrons (R) was adjusted but it failed to reproduce the shape of the experimental data. Furthermore, a secondary electron yield due to ion bombardment (γ) was added and a good fit of the left side branches has been achieved for $\gamma = 0.2\%$. It must be noted that Korolev et al. managed to fit their experimental points equally well by using a particle in the cell (PIC) code. While technically their code is a plasma code and thus more complex and perhaps less detailed than our Monte Carlo code. Their code has all the ingredients of a swarm model but it was not certain to what degree in their modeling the plasma-related features were necessary to fit and explain the experiments.

In Figure 4, we show space time resolved development of electron swarm properties in an RF breakdown in argon. Results are presented for both conditions where a self-sustained mode (lower (a) and higher (b) peak voltages) is operational (i.e., this is done in the region of the breakdown curves where for a fixed *pd* there are two values of the curve). For argon, we have used data from ref.[60] At the lower branch of the breakdown curve, majority of electrons does not make a translation from one electrode to another as assumed in simple models[66,67] and the discharge is maintained by a small group of higher energy particles some of which reach the electrodes. At higher branch, most electrons make the excursion along the entire gap. Remaining and newly produced electrons stay by the electrode until direction of the field changes and thus some phase shift between the positions and the field waveforms are observed together with a skewness for some properties. In both situations, the critical issue is in achieving high energies and consequently ionization to compensate the losses at the surface.

3. Low Current Discharges

In the low current limit of DC discharges, the space charge effects are either negligible or small enough to be treated as a perturbation to the external field.[27,28,68,69] In particular, the diffuse low current regime known as

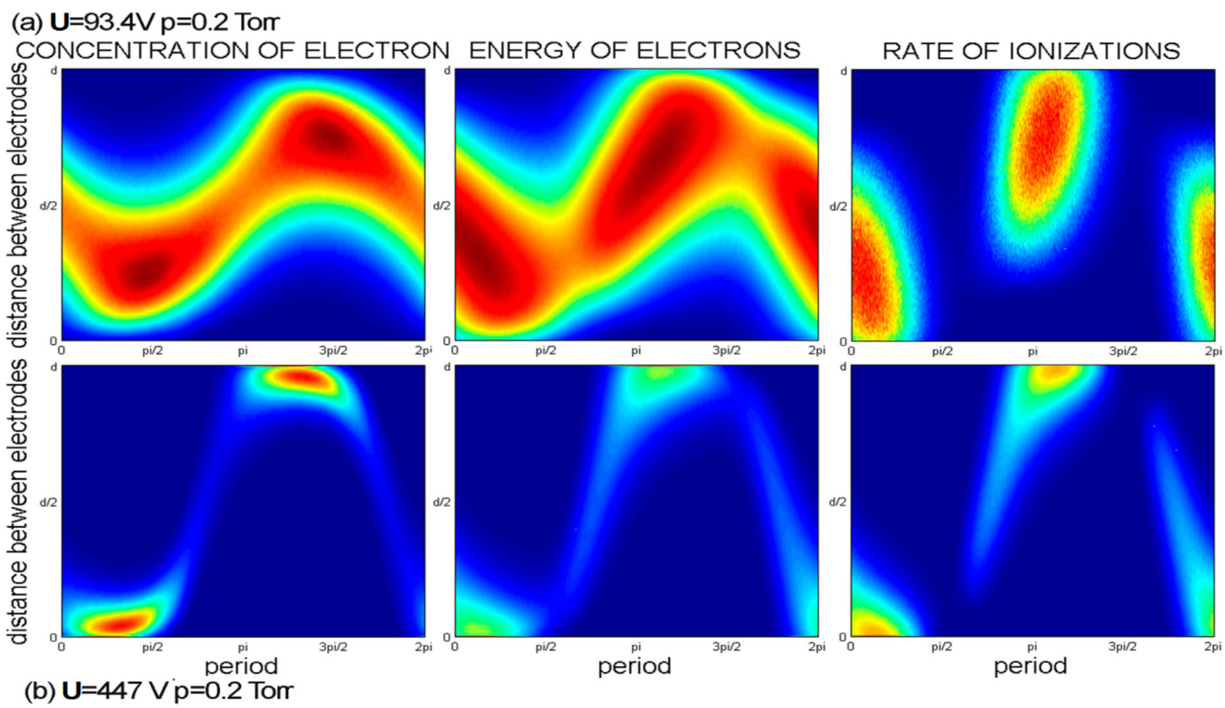

Figure 4. Spatial profiles of electron density, energy, and electron-induced ionization under conditions of an RF breakdown.[63]

Townsend regime of DC discharges is often taken as a representation of swarm conditions[70,71] (steady state Townsend experiment[32,72]).

3.1. Townsend Regime/DC Discharges

If one records axial and radial profiles of emission along the Volt–Ampere (*V–A*) characteristics (Figure 5) of the low current DC discharges, it is clear that for the lowest of currents the profile is diffusion defined and centered[73] while electron density exponentially increases all the way to the anode. That is the sign of the space charge free conditions and the swarm-based models will suffice or in other terms the regime is the so called Townsend discharge. The profiles in the two other regimes (Figure 5 – normal and abnormal glow) reveal a strong effect of the space charge, one in the radial the other in the axial direction. Nevertheless, even under those circumstances, charged particles may not be strongly coupled and for the purposes of theoretical description may behave like swarms, within the limitations of the modified local electric field, thus allowing the swarm physics and transport theory to be the foundation for the description of plasma.

In spite of being supposedly free of space charge effects and any strong coupling, the *V–A* characteristics shows negative slope even in the low current limit (according to the basic Townsend theory, it should be constant) and as a result of the overall *V–A* characteristics and external circuit the discharge may slip into oscillations of different kinds.[73,74]

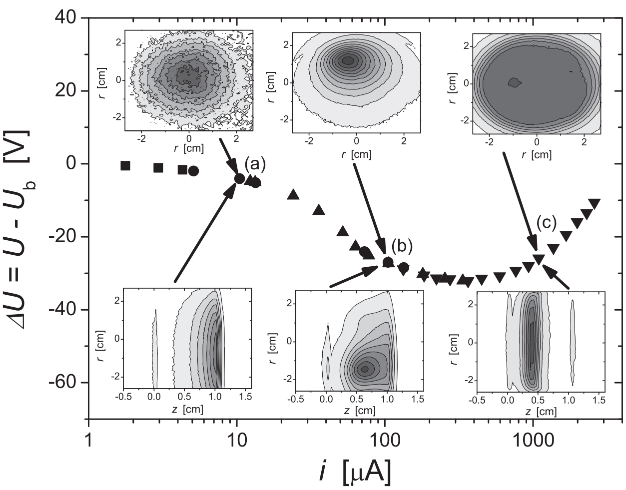

Figure 5. Spatial profiles of emission in low current DC discharges, covering the three regimes with three characteristic profiles: (a) Townsend regime or diffuse low current discharge-falling well under the realm of swarm models; (b) glow discharge-constricted low current discharge; and (c) abnormal glow-diffuse high current discharge[73] (© IOP Publishing. Reproduced with permission. All rights reserved – doi: 10.1088/0963-0252/18/3/034009). Anode is at 1.1 cm and cathode at 0 cm and some small amount of scattered light indicates the positions of electrodes.

The model developed to explain these oscillations due to the negative slope (and thus to extend Townsend's theory) is an example of swarm-based models where space charge effect is calculated based on swarm calculations and added as a perturbation. The basic assumption is that the field right next to the cathode is affected by the space charge and is affecting the energy of ions. The feedback coefficient – the secondary electron yield – cannot be assumed to be constant, as it has been well established that it is dependent on the energy of ions hitting the surface.[60] During oscillations, the voltage and the field will change and affect secondary electron yield (the development from Equation (6) to (11) follows the theory in Phelps et al.[27]):

$$\gamma - \gamma_p + k_U U + k_I I, \tag{6}$$

The term k_U describes the linear component of the voltage dependence. However, the critical assumption in the model is the term proportional to the current. It is actually representation of the space charge which is proportional to the current which then provides additional effect due to the slightly skewed electric field right next to the cathode. It is possible to use the density profiles of electrons and ions (ions being much slower have a much higher density and predominantly define the field perturbation) obtained by swarm physics considerations (drift and free diffusion). Using a Poisson equation we get:

$$E_{Sz}(z) - E_S^C = \frac{1}{\varepsilon_0}\frac{j_z}{W_{+z}}\frac{1}{\alpha_0}[\alpha_0 z - \exp(\alpha_0(z-d)) + \exp(-\alpha_0 d)], \tag{7}$$

where n_+ and n_e are concentrations of ions and electrons, j_z electrical current density in z-direction, W_{+z} drift velocity of ions, e elementary charge of electrons. The corresponding voltage drop is thus equal to:

$$\delta U_S = -\int_0^d \left(E_{Sz}(z) - E_S^C\right)dz = -\frac{1}{\varepsilon_0}\frac{j_z}{W_{+z}} f \quad (\alpha_0, d), \tag{8}$$

where f is:

$$f(\alpha_0, d) = d^2\left[\frac{1}{2} + \frac{\exp(-\alpha_0 d)}{\alpha_0 d} - \frac{(1-\exp(-\alpha_0 d))}{(\alpha_0 d)^2}\right], \tag{9}$$

with round the loop multiplication being

$$g(t) = \gamma[\exp(\alpha(t)d) - 1] = 1. \tag{10}$$

The effective normalized discharge resistance is

$$R_N = \frac{A}{d^2}R_D = \frac{A}{d^2}\frac{\delta U}{I} = -\frac{\hat{\gamma}}{\varepsilon_0 W_{+Z}\hat{g}}\frac{f(\alpha d)}{d^2}, \tag{11}$$

where A is the area of the electrode and $\hat{g}$ is the logarithmic derivative of the multiplication factor g ($\hat{g} = \hat{\gamma} + \gamma\alpha d\ \exp(\alpha d)\hat{\alpha}$).

Experimental tests have confirmed the theory both in the field distribution and in the scaling of the effective resistance.[75] R_N depends on variation with the field (and current) of the secondary electron yield but the theory also provides a foundation for the standard scaling of discharge parameters E/N, pd (Nd), and even jd^2. This bit of theory is added here as an example of how swarm-type considerations are weaved into the plasma theory as its fundamental aspect and also how swarm data enter such calculations as the basic input on atomic and molecular collisions.

3.2. High *E/N*-Runaway Swarms

If one extends the range of measurements to the left hand branch of the Paschen curve, two things become obvious. The first is that the voltage drop between the Townsend regime and the glow discharge diminishes (see Figure 2 in ref.[76]) and sometimes it is even a continuous transition. The reason is simply in the large mean free paths and then the effect of the space charge become less obvious (spread over a larger area). Another effect is observed if energy distribution function (EDF) is sampled, whereby the EDF has a strong peak at maximum available energy thus indicating a runaway.[77] This is a situation where the initial conditions (energy distribution) is maintained (augmented by the energy drop in during the travel through the discharge) and thus properties vary from one point to the next. The low energy tail of secondary electrons also develops, which varies according to the position. Thus a hydrodynamic model is not appropriate, but a MCS may produce excellent results. It may also allow for exact inclusion of the boundary conditions, such as energy-dependent yields due to ions and electrons, energy-dependent angular distribution of secondary or reflected particles, energy-dependent energy loss, and energy-dependent electron reflection. The procedure allows for exact, experimental data to be included for any other possible boundary effect related to electrons, ions, neutrals, reactive species, metastables, and photons. At high E/N that corresponds to the left hand side of the Paschen curve, electrons are not very efficient in ionization (requiring a rapid increase in the voltage necessary for the breakdown as pd is reduced) and even then the multiplication coefficients are only slightly higher than 1.

Due to the low pressure, mean free paths become large for both electrons and ions and they gain much larger energy then in standard discharges. This is particularly critical for ions, allowing them to extend much beyond the standard low energies (below 1 eV) all the way up to the maximum available energy. This opens the door for charge transfer collisions producing fast neutrals. It turns out that fast neutrals are at those conditions more efficient in excitation than either electrons or ions and this all leads to a peak of emission close to the cathode, that is the signature of fast neutral excitation.[78,79] Under those circumstances, momentum transfer in heavy particle collisions leads to a transfer of kinetic energy producing Doppler profiles with excessively high energy wings.[79–81]. As a result, one could predict a possible application for fast neutral etching that would reduce the charging problems[82,83] in treating dielectrics in nanoelectroncs and allow even higher aspect ratios of nanostructures with an increased spatial resolution.

While being mostly non-hydrodynamic, the high E/N discharges are best described by swarm physics. They also open the need for similar models of ions and fast neutrals.

3.3. Microdischarges, Atmospheric Pressure Discharges, Discharges in Liquids

By the virtue of the jd^2 scaling (that has been tested), the Townsend regime may be extended to higher currents at very small gaps. While being counterintuitive, this is a well established fact at least as long as the standard gas discharge physics operates (i.e., below the onset of field emission).[84] Extension of the Townsend regime into higher currents allows for applications of microdischarges that take advantage of the flexibility and ability to directly adjust and achieve a high efficiency of excitation and dissociation (by merely changing the E/N) while still having a high enough total output of photons and/or chemically active species[85] for possible applications.

Microdischarges are one way to produce non-equilibrium plasma at high pressures. They simply operate close to the Paschen minimum and thus allow for much higher pressure for the given reduction in the discharge gap. In the atmosphere, the kinetics of charges is defined by swarm physics, yet if a field is added, the increase of charge density is quite large and a quick transition to highly conductive thermal plasma ensues. Atmospheric pressure discharges often have a high charge density, but in general, the whole atmosphere is an ionized gas that may be described by the physics of swarms. Of the plasmas, the corona discharge consists of streamers and diffuse discharge, which falls under the swarms jurisdiction.[86] Calculation of transport of electrons, ions, and other particles may proceed mainly by using free electron and ion diffusion, but one needs to take into account clusters formed by water vapor molecules and other issues.

One of the lines of the fastest development and new applications are the discharges in water (liquids in general), at the interface between liquid and gas phase, and in gas phase of vapors.[87,88] In liquids one needs to re-establish both cross-sections, transport theory, and transport coefficients. More data are needed for low pressure transport (in gas or vapor).[89,90] In addition, we need to establish techniques to determine and apply data at high pressures and in liquid, i.e., and multiple collision conditions,[91] and under the influence of hydration and breakdown[92] of clusters. In addition, even the low pressure collision and transport data for the most important ions in water vapor are missing.[93] The liquid-related discharges[94] provide a number of challenges and in many circumstances swarm modeling (albeit adjusted to the needs of very high densities) is required. A comprehensive review on discharges associated with liquids may be found in a forthcoming article, containing most importantly list of open issues.[92]

3.4. Afterglow

Upon switching off of the discharge, the space charge may remain for a while as long as the charged particle densities are sufficient to produce a modified field profile. At some moment, the ambipolar field will collapse and free diffusion will ensue that is modeled by swarm physics. In that period, electrons are supposed to continuously lose energy and diminish in numbers. Under some circumstances (depending on the gas, impurities, initial energy, and distribution function and electron and ion densities), an increase of the mean energy occurs during the afterglow leading to a transient peak in the decay of the mean electron energy. Sometimes that peak may be even greater than the initial mean energy, again depending on the gas and on the initial conditions. This phenomenon has often been explained by evoking atomic and molecular physics, including processes such as Penning ionization, Rydberg states, superelastic collisions and recombination.[95,96]

However, a very important process is often overlooked. It is the above-mentioned diffusion heating or cooling[37] (also there is a possibility of an attachment cooling or heating[40]). While the aforementioned processes depend on the initial densities of excited states, this process is universal as it depends only on the ground state momentum transfer cross-section. The presence of the Ramsauer–Townsend minimum in some gases allows very large mean free paths and escape of electrons to the wall of the vessel, thus increasing losses (speeding up thermalization as this could be regarded as evaporative cooling).

Results of a MCS are shown in Figure 6 where we present the development of the mean energy in a limited size discharge vessels and also for an infinite plasma in argon. In an infinite case, thermalization is slow while in the parallel plate geometry decay is much faster (Figure 6a). We also show the decay of the number density of electrons (Figure 6b). In Figure 7, we show energy distribution functions in the parallel plate and in infinite cases. In the latter case, the high energy tail disappears quickly, while the rest of the distribution is close to the Maxwell Boltzmann (MB) distribution with the same energy. This graph also shows as a very general conclusion that, even when the bulk of the distribution function is a Maxwellian, the extrapolation to the high energy tail by a Maxwellian may lead to errors of many orders of magnitude. The reason is that the high energy loss processes such as ionization

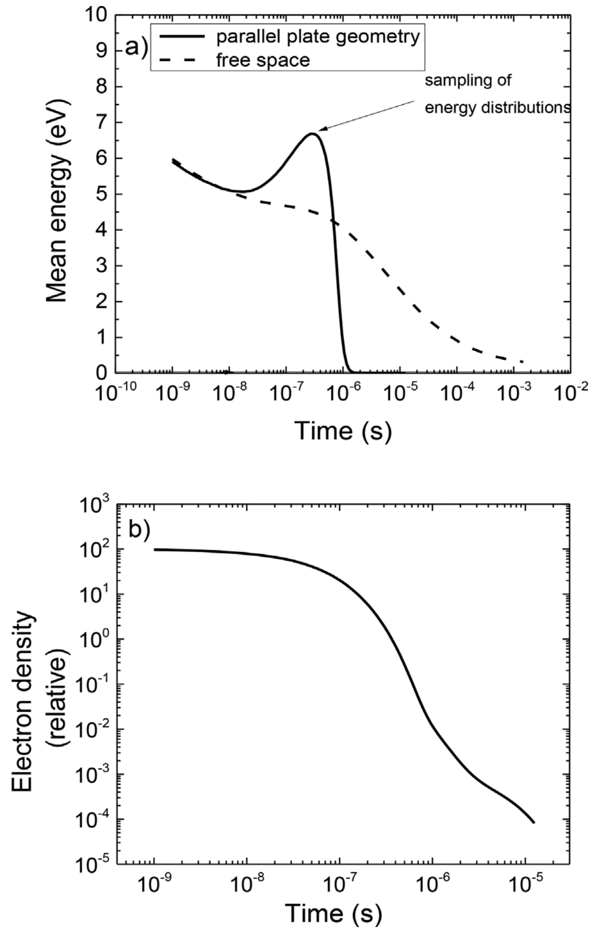

Figure 6. Thermalization (a) and decay of the number (b) of electrons in Ar from the initial MB distribution at 7 eV (argon at 1 Torr and a gap of 1 cm) in plane-parallel geometry and in free space. Arrow marks the point in time where the energy distributions that are shown in Figure 7 are sampled. Two Maxwell Boltzman distributions are shown for comparison, each at a mean energy equal to that in the decayed measured distribution (6.5 and 4.4 eV).

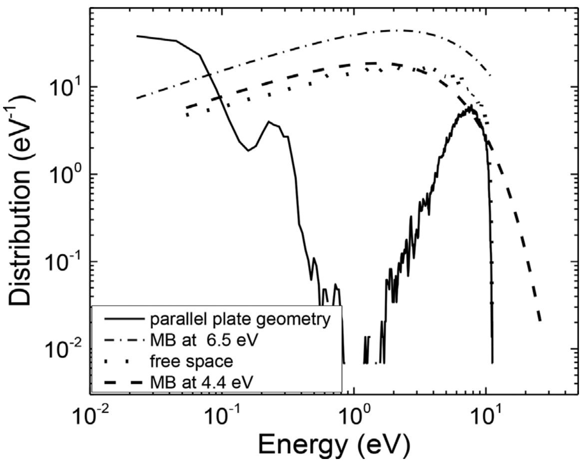

Figure 7. Energy distribution functions at the point marked in Figure 6 for the parallel plate geometry (solid) and infinite geometry (dashed). Two Maxwell Boltzmann distributions for the mean energies corresponding to the two distributions are shown.

deplete the high energy tail of the EDF especially in the low pressure discharges. The bulk of the distribution, and therefore the mean energy, slowly head toward the room temperature thermal equilibrium energy.

For the parallel plate system, due to the long mean free paths as compared to the size of the discharge (even for 1 Torr of argon and 1 cm dimension), a hole is rapidly burned in the Ramsauer–Townsend energy range. At higher energies, the distribution is similar to that of the corresponding Maxwellian, albeit with the high energy tails removed by high energy loss collisions. With the peak of the mean energy greater than the initial energy, the diffusion cooling should be taken into account by including a complete calculation of thermalization (with all the cross-sections and atomic and molecular processes). Diffusion cooling will invariably take place depending on the cross-section shapes; in case of resonant attachment a similar heating/cooling situation may arise and these processes may affect strongly non-local (non-hydrodynamic) systems such as plasma sheaths, Langmuir probe at low pressures[97] and distribution function at the beginning of thermalization or close to the walls.

4. Data Bases for Pristine Gases and Gases with a Large Density of Excited States and Radicals

It is often discussed that the main basis of the plasma modeling is in a set of fluid equations supplied by the swarm data. It is truly important to set up the data bases with a well-prescribed procedures for evaluation in order to be able to appraise different calculations with seemingly same gas mixtures. In addition, coherence between the cross-sections and transport data should be achieved and maintained. The fluid models that use swarm coefficients must be compared to the kinetic and hybrid codes that are based on the cross-sections. Consistency between the two sets should be achieved and it is the task of the atomic, molecular collision physics and necessarily also the swarm physics. A number of data bases has been developed for the purpose of providing low temperature plasma physics with data starting with the JILA data Center (Joint Institute for Laboratory Astrophysics of the Colorado University at Boulder and National Institute for Science and Technology),[98] the data base of Art Phelps at JILA,[62] NIST Data reviews,[99] the data base of Prof. Hayashi,[99] our data base,[100] and many more. The most focused and certainly the largest in number of participants and quantity of data is the currently active LxCAT data base.[101,102] One needs to include the data for ions[100,103] including the most comprehensive data base of Prof. L. Viehland as included in LxCAT[101] and fast neutrals.[104] If the system is a candidate for being described by swarm models sometime it suffices just to observe the available data for some of the transport coefficients to at least appreciate qualitatively the feasibility of the particular system. However, if some kinetic phenomena and in particular non-hydrodynamic kinetic phenomena are involved a full-fledged kinetic simulation is necessary. In that case, however, in order to have proper balances of number, momentum, and energy a swarm normalized cross-sections are necessary to obtain the quantitative comparisons with experiments.

A separate issue is the modeling of plasmas when the background gas may not be regarded as pristine (unperturbed). This issue may be introduced to plasma modeling by making a self-consistent (coupled) calculations of the excited state populations and then of the energy distribution functions and effective rates and other coefficients. While the very presence of the excited states breaks the principal definition of swarms as developed for swarm experiments[105] this, self-consistent modeling extends the applicability of swarm physics to the realm of higher currents/densities of electrons and elevated temperatures. Under these circumstances, a whole new realm of transport data opens, that involves the cross-sections for excited states (stepwise excitation, stepwise ionization, etc.). Not surprisingly first important applications involved modeling of CO_2 lasers,[106] but also hydrogen discharges[107,108] and nitrogen containing discharges.[109,110] Perhaps the widest and the simplest similar modeling is that involving properties of discharges in rare gases with a large abundance of metastables in particular in argon.[111–113] Interest in CO_2 has been reactivated recently due to the activity in energy conversion and storage.[114] In a similar fashion, swarm models may be applied to discharges where fragments of the molecules that may have certain properties abound. For example, the issue of attachment in CF_4 when a significant part of the molecules has been dissociated has been considered in ref.[115] In this section, we did not attempt to make a comprehensive review, we only show few examples and we apologize to those whose work was not included in the list. There are many better sources for detailed bibliography and many much more comprehensive reviews. We only wanted to stress the importance of data when swarm modeling is attempted and also that this approach may be used in the realm of strongly perturbed gases by a large degree of excitation or dissociation thus extending the domain of applicability of swarm physics but requesting a wide range of new data.

5. Conclusion

In addition to all the examples covered here, we have recently made swarm-type modeling in a number of cases involving ionized gases but outside the realm of standard low temperature plasmas. These applications will be the subject of a separate publication[116]:

1) Studies of swarms of positrons may be modeled in the same manner as electrons (without the comfort of production in collisions through ionization, although ionization produces a lot of secondary electrons).[117] With real swarm experiments lacking, the models cannot be used to get quantitative scaling of the cross-sections, but the results provide an insight into processes and new kinetic phenomena, such as Positronium (Ps) formation fueled negative differential conductivity of the bulk component of the drift velocity.[118]
2) In the absence of swarm data measurements, one may define averaged properties that may be used in the same fashion as the swarm data, for example thermalization times (or full thermalization development), ranges of particles, density of deposited energy, and more.[119]
3) Trajectories of particles have been used to describe the properties, although each individual particle does not have enough distinction to give a full insight on the pertinent processes. Still, overall image, obtained by a large set of particles or individual trajectories that are selected, provide sufficient information to make important conclusions.[120,121]
4) Thermalization of positronium in gases.[122,123]
5) Modeling of PET like environment and modeling of chemistry induced by the initial positrons in living tissue/liquid.[91]
6) Modeling of gas-filled positron (and electron) traps, such as Penning Malmberg Surko traps[124,125] or other gas-filled traps.
7) Avalanches and current pulses in gas-filled RPC frequently used detectors in elementary particle detection and the corresponding properties of the gases used in the mixture.[126]
8) Streamer breakdown conditions.[127,128]
9) Modeling of ionization fronts in streamers.[129,130]

The list does not end here and in all those cases swarm studies provide models or an insight into the most salient properties of the discharge. At the same time, one should pay more attention to understanding plasma modeling (from the global to the complex hybrid and PIC codes), the role of plasma models in crossed electric and magnetic fields (e.g., for propulsion studies) and the description of some atmospheric and astrophysical systems (elves, blue jays, clouds of electrons and positrons formed in the vicinity of neutron stars, etc.).

Swarm physics is one of the building blocks of the physics of non-equilibrium plasmas. Another important building block is the swarm data which originate from swarm studies. In addition to being sufficient for some systems, learning how to deal with those will improve our knowledge on applying swarm-based transport equations and data in plasma models. In the meantime, each of the problems mentioned here is interesting, even fun to pursue and a worthwhile contribution.

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