Space-time resolved kinetics of low-pressure breakdown

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Abstract. A review of diagnostics of low-current low-pressure discharges is given with an aim to illustrate how such discharges are used to determine swarm parameters and also how such data may be applied to model and understand the discharges. We have revised how comprehensive modelling of breakdown has led to agreement between binary collision data and the data that may be inferred from the breakdown (Paschen) curves by including processes such as space charge (current) effect on the local field in front of the cathode, photoemission, heavy particle gas phase ionization and backdiffusion. It is also discussed how modelling of Volt-Ampere characteristics in addition to Paschen curves is necessary to establish models of secondary electron emission and how these models may be applied in high current discharges. Finally we show how space time resolved anatomy of the breakdown can lead to understanding of the physics of the initial stages of gas breakdown and formation of Townsend regime, glow and abnormal glow discharges.

1. Introduction
Conditions for definition of electron swarms are that electrons should not perturb the external field, should not be affected by Coulomb forces due to either other electrons or ions or should not have collisions that affect their properties with molecules excited by other swarm particles. Such conditions are found in non-selfsustained and also in Townsend dark discharges. In addition, in other regimes of discharges, conditions may be found where swarm parameters may be applied directly and locally. Townsend regime discharges serve both as a source of swarm data and also may be directly described by swarm parameters through application of Townsend’s theory of breakdown. With significant reduction in number of swarm experiments transport data are more often obtained from gas discharges. In addition to Townsend’s discharges, early stages of gas breakdown are often associated with swarm physics and described by direct application of swarm parameters. In this paper we apply novel diagnostics of Intensified CCD cameras (ICCD) to study the development of low pressure discharges through breakdown and different regimes.

Phelps and coworkers [1-5] have recently started a systematic revision of Townsend’s theory of low pressure breakdown and low current dark discharges (Townsend discharges), as it could not explain some phenomena that were observed in experiments [1-5], such as negative differential resistance, oscillations and discharge constriction. Most of the elements in their recent work have been presented previously by other authors; however, this has been a comprehensive study including all important processes. This study is based on quantitative comparisons with experimental data. By including space charge effects [1-3] and feedback mechanisms other than ion induced production of electrons at the cathode [5], it became possible to develop corrections to the standard Townsend’s breakdown theory in order to explain negative differential resistance and its relation to oscillations in
low current diffuse regime [1-4]. Consequently, the transition to the constricted regime became subject of further studies [4,6,7].

Systematic, well defined measurements in the dark Townsend regime [1-4,7] led to reanalysis of the role of different elementary processes in electron/ion production in gas breakdown and in the maintenance of self-sustained discharges [5]. In particular, it has been established that the secondary electron yield at the surface of the cathode depends on the mean energy of ions hitting the surface. The ion energy is affected by the external voltage, by the local reduced electric field $E/N$ (where $E$ is the electric field and $N$ is the gas number density) and also by the current which leads to a perturbation of the field mostly in front of the cathode where ion concentration is the highest. It has also been shown that this weak dependence of the secondary yield on the space charge perturbation to the field, associated with a strong dependence of the ionization coefficient on $E/N$ near the operating point, causes negative differential resistance that has been observed in low-current Townsend discharges [1-4,7]. Extension of Townsend’s theory developed on the basis of the local-field approximation by Phelps and coworkers [1-3] was shown to be quite successful and it gave a good agreement with the experimental results for field distributions [7].

Extending these studies to somewhat higher currents and transition to constricted (normal glow) and diffuse (abnormal) glow discharges one may utilize the Volt-Ampere characteristics or the spatial profiles of emission to test the models of non-local electron transport and modelling of secondary electron yield that is to be applied for higher current discharges [8,9].

Our most recent studies in this field include time and space resolved development of different regimes of low-pressure discharges. Time resolved development of gas discharge structure is related to the kinetics of basic processes that participate to low-pressure breakdown and discharge maintenance. Similar studies have been recently reported – studies of spatiotemporal development of cathode-fall dominated DC discharges in argon [10,11] and of ignition of high current glow discharge in complex (parabolic) geometry [12]. Our aim was to extend the investigation to different modes of low-pressure discharges and to extend the knowledge of kinetics of formation and maintenance of these discharges in a simple geometry. We have followed the transition from steady-state Townsend to glow discharge and indications of the development of the space charge effects. This approach enables us observe very directly processes leading to development of the constriction, cathode fall and other features of glow discharges. The discharge in glow regime does not satisfy the conditions for swarm physics but many models are based on implementation of basic transport coefficients and our measurements may reveal why and how such approximations may fail or work.

2. Experimental setup
In our experiment, the discharge is established in a simple plane-parallel geometry. The cathode is made of copper, while the anode is made of quartz with transparent yet conductive thin film of platinum deposited on its surface. Thus it is possible to record both radial and axial profiles of emission. The diameter of the electrodes is 5.4 cm while the electrode separation can be set at three different values – 1.1 cm, 2.1 cm and 3.1 cm.

We run a continuous very low current (1-2 µA) discharge by applying a DC voltage to resistors connected in series with the electrodes. Triggering part of the circuit produces short current pulses superimposed on the DC voltages and currents. Pulses of higher current usually last only long enough ($\Delta t < 2$ ms) to make a reliable recording of voltage and current transients. The voltage is measured by two probes, one at the cathode and the other in the anode circuit. The second probe is used only when a relatively high monitoring resistor is connected into the low-voltage anode circuit to determine the current. The emission profiles recorded by the ICCD camera correspond to the conditions of the pulse in the observed time interval. Keeping the pulses short enough enables us to avoid heating and significant conditioning of the cathode surface during the measurements.
The delay generator built into the ICCD camera (Andor, iStar DH720-18U-03) enables us to synchronize recording of the light emission with pulse development and voltage-current measurements.

The details of experimental setup are presented in our previous papers (e.g. [8,9]).

3. Recent developments in description of Townsend’s discharges
In this section we will review and discuss some of the new results in low-pressure gas discharge studies. Basic and, at the same time, the most questionable assumptions of the standard Townsend’s theory (at least the way it is presented in the standard textbooks) are that secondary electron emission is induced only by ions hitting the cathode surface, electric field is homogeneous and that hydrodynamic conditions are satisfied in the discharge (local field approximation applies everywhere). On top of that several processes occurring at surfaces may affect the discharge very strongly.

3.1. Secondary electron yields in homogeneous field
For a long time, there was a great deal of confusion in the literature about the use of secondary yield data in breakdown modelling and, generally about the meaning of the data entering the breakdown condition. The secondary electron yield data obtained from swarm experiments have always failed to match the direct measurements in the binary beam-surface experiments. Phelps and Petrović [5] have shown that in the case of argon, the well known, almost constant, secondary yield of around 8% for argon ions that has been obtained by ion beams on surfaces cannot be directly applied to model even the basic low pressure breakdown. A large number of processes participate in secondary electron production at the cathode (see [5] and references therein). As a matter of fact, ions dominate only in a very narrow range of moderately high $E/N$. Secondary electrons are produced mainly by photons at low $E/N$ and heavy-particle ionization of gas atoms at high $E/N$. Also, metastables have observable contribution at all $E/N$. Including all these processes with appropriate experimental collision data came close to actually reconciling the results for secondary yields obtained in beam experiments and by analysis of the Paschen curve or other gas discharge techniques [5].

Reliable secondary electron yield data obtained from gas breakdown (i.e. by application of Paschen’s law) are required to complete the understanding for other gases. We have analysed Paschen curves and spatial emission profiles from our measurements for a number of gases [13,14]. One has to be aware of two main problems when applying breakdown data. Spatial emission profiles may be used to take into account the real ionization rate [13]. However, hydrodynamic conditions are not completely satisfied in discharge, as it is assumed in Townsend’s theory. There is a distinct non-hydrodynamic region close to the cathode (figure 1). Not taking into account this region can lead to significant errors. In figure 2, we show that discrepancy in secondary electron yields in argon can be up to an order of magnitude if $d_0$ is not taken into account. At high $E/N$, the multiplication becomes smaller, close to 1 because of the low pressure, so the two curves converge as inclusion of the non-equilibrium cannot make a significant change in the multiplication. At high pressures the curves converge because $d_0$ is small and thus its inclusion makes little change in the overall multiplication. A very good agreement is achieved between the measured values of $d_0$ and those determined by the Monte Carlo code with realistic cross sections [14], at least for atomic gases. The semiempirical formula has also been proposed in [5]. Thus, for experiments that do not allow measurement of the spatial emission profiles, calculated data may be used.

The second problem is that the ionization rate taken from the literature may give a quite different multiplication (slope) as compared to the actual experiment and even small errors in ionization coefficient result in large discrepancies of the secondary electron yield [13,14]. This is in general a part of treatment of non-hydrodynamic swarms and one needs to be aware that most of the charged particle kinetics close to the electrodes will proceed in a non-local fashion, which affects not only the discharge itself but also many of the boundary processes such as back diffusion [15] and reflection [16,17].
3.2. Space charge effects: negative differential resistance and oscillations

A study of the low current diffuse discharges revealed that the origin of the negative differential resistance, and consequently oscillations, is the perturbation of the electric field and its effect on the effective ionization coefficient and secondary electron yield [1-3,7]. The non-uniformity of the field is due to the build-up of the space charge increasing the field close to the cathode and thus changing the value of ion induced electron yield. Moreover, it has been shown that even at very low currents these effects are observable. As the matter a fact, phenomenology of dark Townsend discharge and region that is usually labelled as “subnormal” is the same the difference being in the degree of space charge effects. In discussing the values of frequency and the damping of the oscillations we should distinguish between the different types of ions in the discharge and the main mechanisms of secondary electron yield on the cathode.

It has been shown that secondary electron yields obtained for homogeneous field conditions can not be used in modelling of higher current discharges where space charge effects become notable [18]. Nikolić et al. [18] have shown that secondary electron yield dependence on electric field (discharge current) has to be taken into account in order to get physical solution of the model (negative slope of voltage-current characteristics in low-current regime).

Theoretical descriptions of the transition from the low current diffuse to the constricted (normal) glow include those trying to predict the shape of the channel in imposed or self-consistently calculated field distributions by adding the radial fields and diffusion losses. However, the onset of transition is determined by processes at low currents. Clearly, Townsend’s theory in its basic form is not sufficient to achieve a totally adequate description, but some corrected forms of this theory may prove to be sufficient. Theories such as that of Phelps and coworkers [1-4] may be extended to predict the onset of transition to constrictions and its principal causes even without attempting to predict the exact properties of the constricted discharge itself.

3.3. Glow discharges

Significant advances in discharge modelling have been made in the past years, especially with the development of hybrid codes (e. g. [8,9,11,19]). Hybrid models are able to deal with the spatial non-locality of the electron transport in the cathode sheath and have provided important information about the phenomena taking place in a wide variety of discharge.
Hybrid codes, usually assuming a constant value for $\gamma$, have been successfully applied in studies of glow discharges [19]. In order to further improve the accuracy of these models possibility of use of secondary electron yields obtained for low-current discharge conditions (i.e. homogeneous field) [5] would yield satisfactory results in the region of the normal (constricted) glow and abnormal (high current diffuse) glow was investigated [20]. This test, however, resulted in a non-physical behaviour of the electrical characteristics (negative differential resistance in the abnormal glow mode), mainly caused by different combinations and properties of feedback mechanisms resulting from significantly different field and space charge distributions.

As an alternative approach, secondary electron yield has been used as a fitting parameter in some of the recent studies, it has been adjusted to fit different discharge conditions and to match the calculated electrical characteristics with the experimental results [8,9]. The validity of the model has been tested on the basis of our experimental results. The values obtained in this study have been found to be significantly lower compared to the case of the homogeneous field [5] and to change remarkably with changing discharge conditions. This implies that it is generally difficult to prescribe a certain value of $\gamma$ which describes the discharge correctly for a wide range of operating conditions. Using energy-dependent secondary yields, for argon ions and fast atoms and following the procedure proposed by Phelps and Petrović [5], a good fit of the normal and abnormal glow discharges has been achieved [8,9].

Application of the prescription of Phelps and Petrović [5] in higher current discharges and in RF discharges (where phenomenology of gas breakdown may be quite different) appears to be difficult as it is derived on the basis of the breakdown and low current limit where linearity between electron flux and other fluxes holds. This may not be the case in higher current discharges with complex field profiles. Nevertheless the fact that it worked well for DC glow discharges means that modelling of secondary electron yields need not be more complex than that of Phelps and Petrović [5].

4. Voltage-current characteristics and spatial profiles of emission – steady-state measurements

Paschen curve in the range of conditions relevant for our study is shown in figure 3. We have performed measurements in argon at pressure ($p$) $\times$ electrode gap ($d$) products 250 Pa cm, 150 Pa cm and 45 Pa cm and $d = 1.1$ cm, 2.1 cm and 3.1 cm. $pd$ products that are close to Paschen curve minimum ($pd = 150$ Pa cm) were used as a test case. Higher $pd$ values were interesting because of the constricted modes of a discharge, and lower $pd$ values to study contribution of heavy particles to discharge operation.

![Figure 3. Breakdown voltages as a function of gas pressure times the gap length (pd) (Paschen curve) for argon.](image-url)
Our study covered formation and maintenance of different modes of discharges – low current diffuse (Townsend) discharge, normal glow and abnormal glow discharge. Figure 4 illustrates the relation between voltage-current characteristics and the anatomy of the discharge for typical modes of low-pressure discharges. In low-current Townsend regime, axial profiles of emission exhibit exponential increase of emission from the cathode towards the anode. At the same time, radial profiles show that discharge is diffuse. Increase of current will lead to gradual change in exponential axial profile due to space charge formation. Eventually, at the transition from low-current regime to the normal glow, space charge effects will result in formation of cathode fall, which is revealed through the development of the peak of emission. As the current increases, the peak moves towards the cathode. Radially, the discharge structure can be significantly constricted. Constriction broadens up with the increase of current, until it occupies the whole electrode area at the beginning of abnormal glow regime of discharge.

**Figure 4.** Volt-Ampere characteristics of low pressure discharge with spatial structure of the most characteristic discharge regimes indicated. Upper insets show radial contour plots of Townsend discharge, constricted glow discharge and abnormal glow discharge. Lower insets show corresponding axial structure of the discharge (cathode is positioned at \( z = 0 \), while anode is situated at \( z = 1.1 \) cm).

5. **Time-resolved measurements of low-pressure discharges development**

The basis of these results are temporally resolved images of discharges made by a fast ICCD camera, supported by voltage-current measurements. Discharge is established in a plane-parallel system of electrodes and it operates in a pulsed DC regime with variable peak currents. We have performed measurements that covered formation and maintenance of different modes of discharge – low current diffuse (Townsend) discharge, constricted normal glow and abnormal glow discharge. Special care was taken in recording of the spatiotemporal development of discharge oscillations and constrictions. The development of glow and abnormal glow discharges was recorded in side-on and end-on view, revealing radial dependence of the discharge and showing development of constrictions. We selected
to present here one of the most interesting cases – formation of constricted (normal) glow discharge. Temporal development of other modes of low-pressure discharges will be presented elsewhere [21].

Development of constricted form of discharge was recorded at \( pd = 250 \) Pa cm. Under these conditions radial constriction is well developed, while negative differential resistance in low-current limit is low enough to achieve stable and reproducible results. A voltage-current characteristic for 250 Pa cm is shown in figure 5. Figure 6 shows current and voltage waveforms that correspond to development of constricted regime of discharge. Steady-state values of discharge voltage and current are indicated by an arrow in figure 6. 2D images of the discharge are presented in figure 7.

![Voltage-current characteristic](image1)

**Figure 5.** Voltage-current characteristics for \( pd = 250 \) Pa cm and \( d = 1.1 \) cm. Temporal development of discharge is described for selected steady-state conditions, indicated by the arrow [21].

![Current and voltage waveforms](image2)

**Figure 6.** Current and voltage waveforms during formation of discharge constriction. Final steady-state values of voltage and current correspond to conditions indicated by an arrow in figure 5 [21].

Starting from Townsend’s diffuse regime and typical exponential increase of intensity towards the anode, the discharge gradually exhibits space charge effects as the current increases. Gradual formation of the peak of emission can be observed, which is consistent with the formation of the cathode fall [7-11]. The peak of emission rapidly moves towards the cathode. At this point, the discharge is centred and curved towards the cathode (label 3 in figure 7), which is typical for rapid cathode fall development (rapid decrease of voltage, followed by the current growth). Detailed analysis of this kind of discharge development will be analyzed further on in text.

As the discharge current reaches maximum, the radial profile of the discharge broadens and the peak intensity moves further towards the cathode (label 4). Following the decrease of the current, the intensity of emission decreases and the peak of emission moves away from the cathode (label 5), until Townsend-like profile develops (label 6). As the discharge approaches the steady state, formation of constriction can be observed. During this phase of discharge development, the peak of emission gradually moves axially towards the cathode and radially towards the dielectric wall of the discharge chamber.

As it has been observed, throughout the fast cathode fall formation a radial deformation in emission spatial structure occurs (figure 7 1-8). Our initial assumption was that this kind of behaviour is due to the space charge effects. The basic idea was that the discharge can be considered as a group of independent parallel channels. In order to check this assumption, we followed development of spatial structure of the discharge in three different channels of the discharge. We have selected channel of the discharge that corresponds to the maximum intensity at the maximal current (axis of the discharge...
chamber) and channels that correspond to 90% and 70% of the emission intensity at the axis. Emission transients in different channels have been analyzed by establishing relation between position of the peak of emission $d_{\text{max}}$ and peak intensity (figure 8).

Figure 8 shows $d_{\text{max}}(\text{max. intensity})$ for selected channels of the discharge throughout the time interval indicated by the thick line in the current waveform (upper right inset). It can be observed that at specific times $d_{\text{max}}$ are different for different discharge channels. However, all the channels generally follow the same trend. Development of channels closer to the wall is delayed in comparison to the central channel. At the same time $d_{\text{max}}(\text{max. intensity})$ dependency exhibits hysteresis. This indicates non-equilibrium processes that occur during fast cathode fall development.

Presented results confirm our initial assumption that during the cathode fall development the discharge can be observed as a group of parallel channels that operate independently (or in weak dependence on diffusion coefficient).

Figure 7. 2D scans of temporal development of discharge constriction. Labels 1-8 correspond to labels at figure 6. White dashed lines indicate positions of the cathode (left) and the anode (right) [21].

Figure 8. Position of peak of emission in dependence on peak intensity during cathode fall formation [21].

Figure 9. Position of peak of emission in dependence on peak intensity during formation of constriction [21].
In order to investigate further creation of constriction we will continue with the analysis of the development of the spatial structure of the discharge along the same current-voltage waveform. Figure 9 shows $d_{\text{max}}$ for three selected channels of the discharge, for time interval indicated in upper right inset. We kept data obtained for cathode fall formation for comparison. During establishment of stationary state, in the channel that corresponds to highest emission intensity (solid squares) the discharge gradually develops from Townsend-like regime to normal glow regime of discharge operation. Peak of emission moves closer to the cathode, while intensity of emission increases. On the other hand, the remaining discharge channels retain Townsend-like behaviour with an exponential decrease of emission intensity.

Throughout formation of the constricted regime of the discharge, different discharge cannels become dependent. The current growth in one of the channels leads to turning off of the remaining channels due to the decreased operating voltage. Under the given operating conditions, electric field is too low for the discharge to operate in Townsend regime. This section of the discharge clearly operates in the non-selfsustained mode thanks to diffusion of charged particles from the constricted channel. Having this in mind, broadening of constrictions with an increase of the discharge current in the normal glow, can be explained. As the discharge current and charge particles density increase in the conducting channel, growing density of charged particles that diffuse to lateral channels will lead to an increase of the current in those channels and as a result to redistribution of the electric field and formation of the cathode fall. Eventually, the discharge will occupy the whole electrode diameter.

6. Summary
In this review we have covered some of the recent advances in description of the Townsend regime discharges and gas breakdown that is subject to Townsend’s phenomenology and theory. These advances include a new comprehensive modelling of secondary electron emission due to ion, fast neutral, metastable, and photo emission as well as gas phase processes, inclusion of surface processes such as back diffusion and reflection, non-hydrodynamic transport of electrons (and other particles) close to surfaces, and many more processes.

In particular it was found that fitting of not only of the Paschen breakdown curve, but also of the Volt-Ampere characteristics is necessary to represent properly the kinetics of secondary electron emission.

At the same time measurements of properties of low current discharges which include both volt-ampere characteristics and spatial profiles of emission proved to be a fertile basis for modelling and our systematic measurements in different regimes of operation were analyzed by using a hybrid code and appropriate models of collisions, surface processes and in particular by using some aspects of secondary electron production as a fitting parameter.

In this paper we connect those studies with our recent measurements where space and time resolved recordings were made revealing some new phenomena such as curved front of emission and multi regime operation of the same discharge at the same time.

Analysis of normal glow spatial structure development in time enabled us to follow kinetics of cathode fall formation. A constricted form of discharge is established, as the discharge approaches the steady state. In particular it is important to note that coexistence of several modes of operation from Townsend to glow was observed in the same discharge at the same moment, depending on the local current density.

Our goal was partly to provide the experimental data that may be modelled by 2D models, which will provide further information on basic kinetic processes in the discharge. Nevertheless, based merely on inspection of the images one can reveal some new phenomena and settle some issues in the formation of gas discharges upon the breakdown of low pressure gases.

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