Modeling of Townsend discharges at high E/N and low pressure

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Abstract.
This report presents the results of an investigation of complex behavior of electrons and ions in Townsend discharge by a Monte Carlo simulation (MCS) technique. Although recognized as a relatively simple for implementation and at the same time difficult to test and verify, this technique is revealed as a right choice for modelling Townsend discharges. This work begins with selection of input data and benchmarking of codes. We have included description of effects of ions and fast neutrals to kinetics of excitation, description of nonhydrodynamic effects in the vicinity of electrodes and at the high E/N the runaway of electrons and ions. We also present description of impulse discharges and time resolved current impulses under similar conditions. Main presented results were electron energy distribution functions (EEDF), spatial excitation coefficients and arrival time spectra (ATS) obtained for the conditions of Townsend discharges in nitrogen and argon at low pressures and very high E/N. In addition we have compiled cross section sets that may serve for discharge modelling in argon and nitrogen in broad range of conditions and discussed numerous approximations that were recently used in modelling of plasmas.

1. Introduction
Nonequilibrium manifestations in electron and heavy particle transport in gaseous discharges [1] are standard part of all devices that use low pressure discharge. Nonequilibrium effects are related to behavior of these particles in gas that deviates from the hydrodynamic regime. In that sense phenomena presented in this work are part of the swarm physics for which it is characteristic that particles do not interact with particles of the same kind but only with the unperturbed background gas particles. Only partial information is available on elementary collisional processes that are important for modelling of gas discharges. As a result all the research on gas discharges is to a large degree connected to the fundamental studies in atomic and molecular physics.

Phelps and co-workers [2, 3] have performed a series of experiments at moderate to very high electric field to gas number density ratios E/N at low currents and in parallel plate geometry which were the first useful primary experimental data for comparisons with models for high E/N. So far only one dimensional beam models have been developed for such conditions [4]. Such models and the very high E/N experimental data are needed for understanding the nonlocal transport of electrons in the sheaths of the high current discharges [5, 6], microwave breakdown [7] and pseudospark discharges [8] and therefore an independent verification of the models of kinetics at high E/N is justified. In addition, a very interesting feature of the high E/N
transport is the importance of heavy particles in excitation and ionization \[2, 4, 9\], requiring
coupling of electron transport with ion and fast neutral transport models with enough energy for
the excitation of electronic transitions. High mean energies, especially in the case of electrons,
require reflection of electrons from the anode to be taken into account and low pressures imply
that equilibrium is not established throughout the gap.

While some attempts of developing the beam-like models or Boltzmann equation solutions
exist, the best method to describe processes that include such complex boundary conditions,
spatially dependent transport coefficients of electrons, ion and fast neutral transport, and
distribution functions that may include sharp peaks is MCS. We have developed the Monte
Carlo (MC) code for electron transport and a more detailed description of that code, together
with a comparison with experimental results for the processes dominated by electron excitation,
was given in \[10\].

Basic motivation for this research was found in the fact that existing models of discharges
could not explain measurements of Phelps and coworkers (assumes the emission of radiation
both spatially and time resolved) as well as our measurements in these conditions, that refer to
the left side of the Paschen curve.

According to knowledge of many authors, in these conditions nonhydrodynamic behavior
both of electron transport and of ions and fast neutral particles appears and also other effect
such as effect of runaway electrons and ions and fast neutral particles, effect of back-scattered
electrons, excitation by heavy particles.

Modelling in this work is based on experimental conditions in drift tubes of the Laboratory
of Gaseous Electronics in Belgrade or in Phelps’s experiments. Discharges are with currents
of the order of few \(\mu\)A, voltages of the order of \(k\)V and gas pressures of the order of mTorr to few
Torr.

2. Monte Carlo technique
MC code was built to include both boundary effects and nonequilibrium effects, and includes
also particle scattering anisotropy, nonconservative collisions and electron back-scattering on
surfaces.

2.1. Basic principles
MCS comprised running of four MC codes each based on null collision technique \[11\]: code
for electrons \[10\], codes for ions and fast neutrals \[12\] while separate code was used for back-scattered
electrons.

At first we followed primary(direct) electrons that were released at the cathode. Grand total
scattering cross section was used to determine time of the collision. Scattering was anisotropic
and differential cross section (DCS) was used to determine scattering angle after collision.
Angular distributions were selected in a way to keep agreement with momentum transfer cross
section which was determined from total cross section and original source of effective cross
sections.

According to the literature data \[13\] secondary electron share energy with primary electron
after the ionizing collision.

MCS for heavy particles transport was based on the codes for ions and fast neutrals which
were similar except by electric field value.

In all codes we have assumed stationary target, velocities in LAB system after collision were
obtained from the velocities in CMS after the collision, fast neutrals were followed only up to
the point where their energy is under the excitation threshold \(\varepsilon_L\).
2.2. Electron backscattering
Phelps and coworkers [2] have noticed the peak in the spatial profile of the emission closest to the cathode which was attributed to the electrons back scattered from the anode and latter was confirmed by MCS [14]. MCS were also used to obtain effect of back-scattered electrons on multiplication on argon [15].

Backscattered electrons were divided in two groups: reflected and secondary electrons. For both, from the literature data [16, 17, 18] we were able to select initial number, initial energy and direction from electrode surfaces of graphite (gr) and stainless steel (ss).

2.3. Selection of input data and benchmarks
Input data required to run MCS in both Ar and N\textsubscript{2} were based on best compilation we found in publications of Phelps and Hayashi. These include both total cross sections and differential scattering cross sections.

For the purpose of running the code for electrons we have prepared complete sets of cross sections of Phelps [19, 20] for molecular nitrogen and of Hayashi [21] for argon atom. Both were completed with differential cross sections for energies below 1 eV and up to 10 keV for argon [12] and molecular nitrogen [19, 10].

The cross sections for heavy particles were not found in great detail as for the electrons. Nevertheless, well validated cross sections of Phelps [22] were used as a base while additional modifications and verifications were performed to properly take into account the ratio of resonant charge transfer collisions to elastic collisions [23, 10].

Data used in plasma modelling are almost exclusively based on the swarm theory and experiments [24]. It was a common practice to apply benchmark calculation to any new theoretical model and numerical code before the code was accepted as accurate. Benchmark calculations were done for all available scattering models relevant for DC electric fields [25, 11, 26, 27] for all our transport codes as well as for common sampling procedures and conditions [28, 29].

3. Modelling of emission in dark Townsend discharges at high \( E/N \)
In drift tube experiments the spatially resolved emission obtained side on in large part of the discharge usually exhibits exponential trend or nearly exponential trend in the most of the interelectrode space. Exponential trend is a consequence of the equilibrium electron transport for these electron groups [19]. Thus appears "expected" that perfect agreement of Townsend ionization coefficient (TIC) from the MCS slope of the spatially resolved emission and experiments of Haydon and Williams [30] can be obtained. So if there were no other effects TIC was possible to obtain from the slope of the spatially resolved measurement of emission as was experimentally done for many gases [2, 31, 32, 33, 34, 35, 36, 37].

By increasing reduced electric field spatial emission dependencies with 391.4 nm in large part of the space stay nearly exponential while with some emission bands like 2\textsuperscript{+} band (see Fig. 1), drastically alters shape of the spatial dependency with respect to the exponential.

Similar behavior has also appeared in argon where "expected" behavior is observed for 750 nm line [38] while "unexpected" for 811 nm line [32]. It was found in Scott-Phelps experiments [38] that the cause for such spatial excitation dependency is actually contribution of heavy particles to excitation, precisely speaking fast N\textsubscript{2} in the case of nitrogen discharge.

Modelling results of digitized experimental data [2] for the spatial excitation coefficient for the 337.1 nm emission line of N\textsubscript{2} at high \( E/N \) are shown in Fig. 1. Clearly three components of spatial dependence of emission are observed obtained by electron excitation by direct (d) and secondary electrons (s) produced in the gas phase, heavy particle excitation (fn) and excitation by backscattered electrons (b) both reflected and secondary. Results of MCS are normalized to the electron excitation coefficient at the anode that corresponds to the normalization of
experimental excitation coefficients to the anode current [33]. Emission at the anode was used since the electron current was known at that point. It was actually the spatial profile of emission normalized to the excitation coefficient at the anode. Effect of low electron reflection (graphite) and high electron reflection (stainless steel) surface was successfully modelled by electron backscattering and present data.

Figure 1. Spatial excitation coefficient for high $E/N$ at pressure $p = 61$ mTorr ($E = 250$ V cm$^{-1}$, $d_{AC} = 3.61$ cm). The results from [2] are shown with EXP.

Excitation coefficients measured in drift tubes provide the basis to normalize the cross sections for specific channels of excitation and thus to avoid the use of the fictitious effective cross sections.

A combination of measurements of the excitation coefficients and the standard sets of the cross section data were also used in order to renormalize cross sections for specific channels [39].

4. Modelling energy distribution functions at the electrodes

One of the fundamental quantities determining the kinetics of the discharge is EEDF, since electrons are responsible for excitation, ionization and dissociation of heavy particles in gas discharges. At low pressure and correspondingly at high discharge voltage and $E/N$, electrons may not be in local equilibrium with the electric field. Thus, their EEDFs manifest a departure from the hydrodynamic distribution. This departure can be quite substantial, and groups of runaway electrons with beam-like behavior may be formed in the EEDF.

The anode boundary at very high $E/N$ represents significant electron nonequilibrium region due to the very high electron energies and its link to a metal surface.

First our modelling results of the EEDF at the absorbing anode will be presented. They show development of runaway electron groups as a function of increasing electric field i.e. available energy ($\varepsilon_{max}$). EEDF evolution at the anode as a function of $E/N$, for the Paschen curve [2] conditions at high and very high $E/N$ obtained by MCS is shown in Fig. 2. Main characteristic of this dependence is showing up the peaks at very high energies which characterize beams of runaway electrons. Beam component appeared at moderate $E/N$ has developed over the intensity, as a function of $E/N$, in such a way that pronounced components of the beam
were distinguished at inelastic threshold energy distance. By increasing electron energy beam resolution is less important since largest number of inelastic events happens close to the cathode and largest number of electrons accelerates almost up to the maximum energy. At very high $E/N$ the number of low energy electrons is very low because of their origins in ionization events close to the surface. The anode boundary is characterized by the high energy electrons which may do multiplication in gas or in surface layers of the electrode. In the first case high energy electrons produce reflected electrons and in the second case secondary electrons. Depletion of low energy electrons due to their absorption at the anode is not a dominant characteristic of these EEDFs.

Figure 2. EEDF at the anode in nitrogen obtained by MCS for series of high and very high values of reduced electric field. All EEDF are obtained for distance $d = 3.61$ cm with respect to the cathode, for the conditions of selfsustained discharge [2] between electrodes (sintered graphite). EEDF normalization is with respect to initial number of electrons from the cathode.

As a separate task we did modelling of EEDF measured at the anode by Vrhovac et al. [40] separated from the cathode by $d_{AC} = 1.85$ cm. Electrons with the energies $\varepsilon$ arriving at the anode were left to penetrate through the micrometer holes to the other side of the anode where they were collected by the retarding potential analyzer system. By MC modelling we were able to discuss both equilibrium and nonequilibrium parts of the EEDF (Fig. 3). In the modelling, in order to successfully describe measured EEDF it was necessary to include both secondary and reflected electrons.
5. Modelling impulse discharge

The studies of impulse discharges showed up as very promising since they may give useful information about the processes relevant to breakdown formation.

In this work our goal was to model Gylys-Phelps experiment (GPE) [3], an impulse discharge operating in prebreakdown conditions which exploits pulsed photoemission technique to initialize discharge. In GPE basic setup consisted of two planparallel electrodes to which high voltage was applied. One of the electrodes was semitransparent (cathode) and was illuminated by the pulsed laser beam (10 Hz). The laser energy was sufficient to induce low energy electron emission at the other side of the cathode. By detecting emission and/or determining the charge passing through the circuit one may figure out about breakdown of such a system.

The analysis of time dependence of accumulated charge at the capacitor connected in series to the electrode system was complicated by displacement currents which were the result of particles transport in discharge. If one wants to determine electron multiplication from the time dependence of the accumulated charge than time interval between the end of the electron ATS and beginning of heavy particles ATS should be larger than time resolution of recording device. Because of that the knowledge about the kind of particles we follow in certain time interval is of crucial importance in determination of the electron multiplication in discharge.

The MC technique we applied to model such a system was similar to the ones described previously, except that now we followed arrival time of the particles to the electrodes.

ATS’s of the avalanches reaching the electrodes are shown in Fig. 4. Isolated avalanche at the leading edge of the pulse train was a consequence of direct electrons, having the maximum pulse corresponding to runaway electron group. The continuing shape shows arrival times of secondary electrons until slowest and nearest electrons close to the anode have arrived. The direct electron and ion contribution were separated from the secondary contributions due to ionization by ions.
and fast neutrals. Excellent agreement was obtained for electron multiplication obtained from MCS and from GPE results for nitrogen.

Generally ATS in Fig. 4 a) are slightly wider than in Fig. 4 b) due to the higher gas density.

![Figure 4](image-url)

**Figure 4.** ATS for electrons, \( N_2^+ \) ions and fast neutrals obtained by MCS for the conditions of non-selfsustained discharge in GPE: a) \( E/N = 10 \text{kTd}, d_{AC} = 4 \text{ cm}, N d_{AC} = 8 \times 10^{15} \text{ cm}^{-2}, \varepsilon_L < 15 \text{ eV}, \) b) \( E/N = 52 \text{kTd}, d_{AC} = 4 \text{ cm}, N d_{AC} = 2 \times 10^{15} \text{ cm}^{-2}, \varepsilon_L < 10 \text{ eV}. \) All ATS are normalized to the initial number of electrons. ATS of electrons arriving at the anode \( e^{-}(o) \) - direct and secondary produced in gas, \( e^{-}(i^+) \) - formed by ion impact ionization of \( i^+(e^-) \) avalanche; \( e^{-}(fn) \) - formed by fn transport formed by ion avalanches \( i^+(e^-) \). ATS of \( N_2^+ \) ions arriving at the cathode: \( i^+(e^-) \) - formed in ionizing collisions of \( e^{-}(o) \) electron avalanche with \( N_2 \). ATS of fast neutrals to the cathode: \( fn(i^+) \) - formed by \( i^+(e^-) \), \( fn_{(2)} \) - formed by \( i^+ \) (not shown) following \( e^{-}(i^+) \) avalanche (not shown) formed by \( i^+(e^-) \) ion impact ionization, \( fn_{(3)} \) - formed by \( i^+ \) (not shown) following \( fn(i^+) \) avalanche (not shown) ionizations.

### 6. Conclusion

We built and tested Monte Carlo codes in order to follow transport of electrons, ions and fast neutrals in homogenous electric field between two plan-parallel electrodes.

We formed database for scattering of electrons, ions and heavy particles in nitrogen and argon, as well as electron scattering on metal surfaces (graphite, stainless steel).

We precisely determined contributions of electrons, ions and fast neutrals to spatially resolved emission in argon and nitrogen.

By using the precise models we calculated the spatial excitation coefficients at very high \( E/N \) and proved that excitations in the cathode boundary were produced by neutral particles.

By MCS we determined EEDF at the graphite anode and obtained good agreement with experimental results in the whole energy range.

The analysis of series of transient measurements of Phelps in nitrogen was performed. We showed the precise time frame in which Gylys-Phelps experiment proceeds from moderate to
very high $E/N$.

Acknowledgments

The work is supported by the MNRS under grants 141025. The author acknowledges all colleagues from the Institute of physics, Belgrade for numerous discussions. Special thanks go to Prof. Z. Lj. Petrović and Dr. Ž. Nikitović for their continuous encouragement and support and to Prof. A. V Phelps for his numerous suggestions and comments.

References

[1] Petrović Z Lj, Sušaković M, Nikitović Ž, Dujko S, Šašić O, Jovanović J, Malović G and Stojanović V 2007 Plasmas Sources Sci. Technol. 16 S1

[2] Jelenković B M and Phelps A V 1987 Phys. Rev. A 36 5310

[3] Gylys V T, Jelenković B M and Phelps A V, 1989 J. Appl. Phys. 65 3369

[4] Phelps A V, Jelenković B M and Pitchford L C 1987 Phys. Rev. 36 5327

[5] Vrhovac S B, Stojanović V D, Jelenković B M and Petrović Z Lj 2001 J. Vac. Sci. Technol. B 12 461; Date A, Kitamori K, Sakai Y and Tagashira H 1992 ibid. 25 442

[6] Den Hartog E A, Dougherty D A and Lawler J E 1988 Phys. Rev. A 38 2471

[7] Hays G N, Pitchford L C, Gerardo J B, Verdeyen J T and Li Y M 1987 Phys. Rev. A 36 2031

[8] Bouch J P and Pitchford L C 1991 IEEE Trans. Plasma Sci. 19 286

[9] Petrović Z Lj, Jelenković B M and Phelps A V 1992 Phys. Rev. Lett. 68, 325

[10] Stojanović V D and Petrović Z Lj 1998 J. Phys. D: Appl.Phys. 31 834

[11] Reid I D 1979 Aust. J. Phys. 32 231; ibid. 1982 35 473

[12] Petrović Z Lj and Stojanović V D 1998 J. Vac. Sci. Technol. A 16 329

[13] Opal C B, Peterson W K, and Beatty E C, J. Chem. Phys. 55 4100 1971

[14] Petrović Z Lj, Stojanović and Jelenković B M J. Appl. Phys. 81 1601

[15] Petrović Z Lj, Stojanović V and Radmilović M 2000 SPIG Book of contributed papers, Zlatibor Yugoslavia

[16] Woods M E, Hopkins B J, Matthews G F, McCracken G M, Sewell P M, and Fuhrang H 1987 J. Phys. D 20 1136

[17] Thomas E W 1985 in Particle Interaction with Surfaces Oak Ridge National Laboratory, Oak Ridge, Vol. III.

[18] Gergely G, Gruzza B, and Meynard M 1980 Acta Phys. Acad. Sci. Hung. 48 337

[19] Phelps A V and Pitchford L C 1985 Phys. Rev. A 31 2932; 1985 JILA Report No.25

[20] Phelps A V 2004 ftp://jila.colorado.edu/collision data/electronneutral/electron.txt

[21] Hayashi M 1992 personal communication

[22] Phelps A V 1991 J. Phys. Chem. Ref. Data 20 557; ibid. 1992 21 883

[23] Phelps A V 1994 J. Appl. Phys. 76 747

[24] Huxley L G H and Crompton R W 1973 The diffusion and drift of electrons in gases New York: Willey Interscience

[25] Lucas J and Saelee H T 1975 J. Phys. D: Appl. Phys. 8 640

[26] Nolan A M, Brennan M J, Ness K F and Weddeng A B, 1997 J.Phys. D: Appl.Phys. 30 2865

[27] Skullerud H R 1973 J. Phys. B: Atom. Molec. Phys. 6 728

[28] White R D 1997 J. Phys. D: Appl. Phys. 30 810

[29] Sakadžić S and Petrović Z Lj 2000 20th SPIG, Zlatibor, Yugoslavia

[30] Haydon S and Williams O 1976 J. Phys. D 9 523

[31] Stojanović V, Božin J, Petrović Z Lj and Jelenković B M 1999 Phys. Rev. 42 4983

[32] Phelps A V and Jelenković B M 1988 Phys. Rev. A 38 2975

[33] Stokić Z, Fraga M M F R, Božin J, Stojanović V, Petrović Z Lj and Jelenković B M 1992 Phys Rev A 45 7463

[34] Šašić O, Malović G, Strinić A, Nikitović Ž and Petrović Z Lj 2004 New Journal of Physics 6 74

[35] Malović G N, Strinić A I, Petrović Z Lj and Sadeghi 2004 Spectrochimica Acta part B 61 951

[36] Malović G N, Strinić A I, Petrović Z Lj, Božin J V and Manola SS 2000 Eur. Phys. J. D 10 147

[37] Nikitović Ž, Strinić A, Malović G, Stojanović V and Petrović Z Lj 2008 Acta Chim. Slov. 55 219

[38] Scott D A and Phelps A V 1991 Phys. Rev. A 43 3043

[39] Strinić A I, Malović G N, Petrović Z Lj and Sadeghi N 2004 Plasma Sources Sci. Technol. 13 333

[40] Vrhovac S B, Stojanović V D, Jelenković B M and Petrović Z Lj 2001 J. Appl. Phys. 90 5871

[41] Kelly L J and Blevin H 1989 Proc. 19th ICPIG Belgrade Editor J.M. Labat vol. IV 892 Donko Z and Rozsa 1995 Acta Physica Universitatis Comenianae 36 35