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Obtaining precise electron swarm parameters from a pulsed Townsend setup

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Abstract. A swarm parameter experiment is introduced, which implements the pulsed Townsend electrical method with a high degree of automatization. The experimental setup and measurement procedures are described in detail, and a comprehensive definition of the swarm model is given and used for signal analysis. The intrinsic parameters of electron drift currents in the pulsed Townsend method are identified, and novel regression methods are presented for obtaining electron swarm parameters from pulsed Townsend measurements. The setup and methods are verified with measurements in Ar, N₂ and CO₂, which are focused on the (E/N) -range between dominating electron attachment and weakly dominating ionization. The present data are compared to experimental reference data, and to electron transport coefficients calculated by a Boltzmann solver and simulated by a Monte Carlo method. Excellent agreement was found between the present data and the Monte Carlo results, but there are significant discrepancies to widely used recommended swarm parameters of N₂ and CO₂. Finally it is proposed to revise some hitherto recommended values of electron transport coefficients.

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1. Introduction

Measurements of electron swarm parameters in gases are required for basic studies of electron kinetics and electron-atom or electron-molecule interactions [1], and are essential for simulations of plasma devices [2, 3, 4]. The pulsed Townsend (PT) method is a traditional experiment for measuring electron swarm parameters [1, 5]. Its main advantages are that it permits direct observation of the spatiotemporal evolution of an electron swarm yielding ionization and electron drift and diffusion data at the same time. The operation of a PT experiment involves several repetitive tasks in between measurements, which consume more time than the measurement itself. We have built the SParX (swarm parameter experiment) PT setup using dedicated control software for running measurements in automated processes. These techniques permit quick

coverage of multi-dimensional parameter spaces, such as different mixing ratios of dual and ternary gas mixtures.

The PT setup (figure 1) consists basically of two parallel-plane electrodes at spacing d inside a vacuum chamber which is filled with a gas sample. By a short laser pulse an ensemble of electrons is generated at the cathode. It drifts through the sample gas at constant average velocity in a homogeneous electric field E_z . The collective motion of these electrons under the influence of their interactions with the gas molecules is modelled as a swarm. This motion gives rise to a displacement current which is recorded and analysed.

The analysis is based on the time dependent Gaussian spatial distribution of swarm electrons [5]

$$\frac{dn_e(z, t)}{dz} = \frac{n_0 \exp(\alpha_{\text{eff}} z)}{\sqrt{4\pi D_L t}} \exp\left(-\frac{(z - wt)^2}{4D_L t}\right), \quad (1)$$

where α_{eff} is the effective ionization coefficient, D_L is the diffusion coefficient in the z -direction, w is the swarm velocity in the z -direction, and n_0 is the electron number at $t = 0$ and $z = 0$. However, in the PT method the parameters α_{eff} and D_L can not be determined directly without precise knowledge of z . Moreover, there is pertinent discussion about the interpretation of PT measurements, perhaps because the relation between the PT method and theoretical quantities of electron transport was considered for special cases only [6, 7], but not universally.

A different evaluation procedure is applied here. This paper begins with a transformation of (1). It yields an expression $n_e(z, t)$, where the parameters are time intervals and frequencies. Waveforms of PT measurements can then be analyzed without a priori assuming values z and w , and it is possible to develop regression analyses for obtaining the electron swarm parameters. Subsequent sections present details of the experimental setup and measurement procedures. Briefly we review the available reference data of Ar, N₂ and CO₂, then our results for these gases are presented and compared to those of the references.

2. Theory

The kinetic theory of swarms is found e.g. in reviews by Kumar [1] or Petrović [2]. On the basis of kinetic theory, the PT method was described by a temporal electron growth [6, 7, 8]. It is necessary to transform the spatial swarm model (1) to the temporal domain using the relations

$$\alpha_{\text{eff}} dz = \nu_{\text{eff}} dt \quad (2)$$

$$2D_L t = w^2 \tau_D t, \quad (3)$$

which introduce the effective reaction rate ν_{eff} and the diffusion time constant τ_D . Inserting these relations in (1) yields

$$\frac{dn_e(z, t)}{dz} = \frac{n_0 \exp(\nu_{\text{eff}} t)}{w \sqrt{2\pi \tau_D t}} \exp\left(-\frac{(t - T)^2}{2\tau_D t}\right). \quad (4)$$

With

$$w = \frac{dz}{dt} \quad (5)$$

one obtains a temporal electron number distribution dn_e/dt with its peak at the instant $T = d/w$, which implicitly is given by the electrode spacing.

2.1. Model of electron swarms

At an arbitrary position $z = d_0$ an initial electron distribution is assumed,

$$\frac{dn_e(d_0, t)}{dt} = \frac{n_0}{\sqrt{2\pi\sigma_0^2}} \exp\left(-\frac{(t-t_0)^2}{2\sigma_0^2}\right), \quad (6)$$

with initial electron number n_0 and initial temporal broadening σ_0 . At the instant t_0 is the center of the Gaussian laser pulse. This swarm moves at constant average bulk velocity w , and, after the transit time T , passes an observer at position $d = d_0 + wT$. There the electron distribution is

$$\frac{dn_e(d, t)}{dt} = \frac{n_0 \exp(\nu_{\text{eff}}(t-t_0))}{\sqrt{2\pi\sigma^2(t)}} \exp\left(-\frac{(t-t_0-T)^2}{2\sigma^2(t)}\right). \quad (7)$$

The swarm temporal broadening $\sigma(t)$ increases with the diffusion time constant τ_D .

$$\sigma^2(t) = \sigma_0^2 + \tau_D(t-t_0) \quad (8)$$

The total number of electrons changes at an effective rate ν_{eff} , which accounts for electron attachment and ionization. The model collapses with electron detachment, cathodic feedback or photoionization by secondary photons. However, ν_{eff} is not affected by ionic or metastable species *accumulated* in the gas, because their residence time is shorter than the interval between two subsequent measurements.

2.2. Electron currents

An analytical expression of the swarm drift current is deduced and serves for comparison to measured currents. Similar expressions for electron currents were given by de Urquijo [9] and Ridenti [10] and have elsewhere been used for analysing PT measurements.

The electron current $I_e(t)$ of a swarm that moves across an electrode gap d and has a transit time T is proportional to the number of electrons in the gap [11].

$$I_e(t) = \frac{q_0}{T} \int_0^d n_e(z, t) dz, \quad (9)$$

where q_0 is the electron charge. $I_e(t)$ can be expressed as the product of a constant amplitude and three time dependent amplitude factors [9, 10].

$$I_e(t) = \frac{n_0 q_0}{T} \frac{1 + A_L(t)}{2} \frac{1 - A_D(t)}{2} A_\nu(t) \quad (10)$$

$A_L(t)$ accounts for the swarm initiation in front of the cathode, $A_D(t)$ accounts for the absorption of the swarm at the anode, and

$$A_\nu(t) = \exp(\nu_{\text{eff}}(t-t_0)). \quad (11)$$

An ensemble of photoelectrons is initiated at position d_0 in front of the cathode by a laser pulse of Gaussian temporal profile, and it is accelerated in the electric field. We assume that the laser pulse profile determines the electron swarm for the times $t \leq t_0$, and that the effect of diffusion begins at the instant t_0 . Then the amplitude factor $A_L(t)$ can be defined using the canonical error function erf.

$$A_L(t) = \begin{cases} \operatorname{erf}\left(\frac{t-t_0}{\sqrt{2\sigma_0^2}}\right) & \text{for } t \leq t_0 \\ \operatorname{erf}\left(\frac{t-t_0}{\sqrt{2\sigma_0^2+2\tau_D(t-t_0)}}\right) & \text{for } t > t_0 \end{cases} \quad (12)$$

For an electrode gap $d = d_0 + wT$ the amplitude factor $A_D(t)$ is

$$A_D(t) = \operatorname{erf}\left(\frac{t-t_0-T}{\sqrt{2\sigma_0^2+2\tau_D(t-t_0)}}\right). \quad (13)$$

The experimental conditions can be chosen such that T is much larger than τ_D . Then $A_L(t)$ can be evaluated when $A_D = -1$, and $A_D(t)$ can be evaluated when $A_L = 1$. When $A_D = -1$ and $A_L = 1$, then $A_\nu(t)$ can be evaluated independently of initial broadening and diffusion.

2.3. Ion currents

In the present investigation only electron currents $I_e(t)$ are analysed, but in many cases substantial ion currents are observed after the electron transit. Then ions are also present during the electron transit, and it is essential to separate the ion current $I_{\text{ion}}(t)$ from $I_e(t)$ in order to obtain ν_{eff} .

$$I_{\text{measured}}(t) = I_e(t) + I_{\text{ion}}(t) \quad (14)$$

A simple expression of $I_{\text{ion}}(t)$ is used, which is for the times $t_0 \leq t \leq T$

$$I_{\text{ion}}(t) = \begin{cases} A_{\text{ion}} \frac{t-t_0}{T} & \text{for } \nu_{\text{eff}} \leq 10^3/\text{s} \\ A_{\text{ion}} \frac{\exp(\nu_{\text{eff}}(t-t_0))-1}{\exp(\nu_{\text{eff}}T)-1} & \text{for } \nu_{\text{eff}} > 10^3/\text{s} , \end{cases} \quad (15)$$

and for $t > T$ it is

$$I_{\text{ion}}(t) = A_{\text{ion}} , \quad (16)$$

where A_{ion} is the ion current amplitude at the end of the electron transit.

3. Methods

3.1. Pulsed Townsend setup and operation

The experiment is installed in a cylindrical stainless steel vacuum ($< 10^{-5}$ Pa) chamber. After the inflow of sample gases is completed, the gas temperature is measured by a Pt100 sensor, and the pressure is measured by a capacitive sensor for the range 50 Pa to 11 kPa. In the chamber two polished stainless steel electrodes with radius

Figure 1. Optical setup and electric circuit diagram. The two electrodes with spacing d are within a vacuum chamber. The laser beam (---) illuminates the photocathode (.....) via a beam expander (BE). Arrows indicate the swarm drift direction. High voltage elements are the DC source U , damping capacitance C_h and resistance R_h . The anode is connected to the oscilloscope via a transimpedance amplifier with variable gain R_a .

$r_0 = 56$ mm represent a $\pi/2$ -Rogowski geometry for their ideal spacing $z_0 = 15$ mm. The parametrization of their profile (r, z) is

$$r = [0 \dots r_0] \quad (17)$$

$$z(r) = \frac{z_0}{\pi} \exp \left(2.35 - \frac{r_0 - r}{z_0/\pi} \right). \quad (18)$$

This electrode geometry produces an electric field of cylindrical symmetry that is almost homogeneous for $r < 20$ mm with $d \leq 18$ mm, and avoids critical field strengths at the electrode edges. The z -position of the anode, and thus the electrode spacing d , can be varied by a motor driven micrometer screw in steps of $6 \mu\text{m}$.

Centrally in the cathode a plane photocathode with diameter 25 mm is installed, which consists of a transmissive palladium film on quartz. It will be described elsewhere in more detail. The film is back-illuminated, as indicated in figure 1, by 266 nm laser pulses of 1.5 ns FWHM and about 155 μJ pulse energy at 20 Hz repetition rate, producing about $3 \cdot 10^7$ initial swarm electrons. The laser beam is expanded to cover the complete photocathode. We were able to choose this simple and economic laser system (CryLas FQSS 266-200) because the photocathode is so very efficient.

Figure 1 shows the electric circuit that contains the electrodes. We use damping elements $R_h = 150 \Omega$ and $C_h = 2.7 \mu\text{F}$ between the DC high voltage source and the cathode. The low voltage circuit was optimised for high signal bandwidth (200 MHz) and high fidelity. The anode is connected to a transimpedance amplifier (FEMTO DHPCA-100) with gain resistor R_a . For recording electron currents we set $R_a = 10^2$ or 10^3 V/A , and ion currents can be recorded with $R_a = 10^5$ or 10^6 V/A . The oscilloscope (R&S RTO) is equipped with a 10 GS/s 8 bit AD-converter and a 16 bit digital signal processor, which is configured for data compression and averaging. It is triggered by a photodiode built into the laser system.

Figure 2. Electron drift currents in Ar at 30 Td and in CO₂ at 65 Td, the latter current was divided by 10 and time shifted by 0.2 μ s. Averages of measured waveforms (—) and the fit (·····) of (10) coincide well. T marks the electron transit time in Ar, and the markers T_1 to T_4 are explained in section 3.2.

The operation of all elements of the setup is remotely controlled and has been integrated into a single Matlab environment on a control computer.

3.1.1. Range and uncertainty of experiment parameters Currently the experiment operates at gas temperatures 293 to 300 K, which are measured but not controlled. (E/N) is controlled by the experiment parameters d , p , U . For the present study their range of values is:

- d from 9 to 18 mm.
- p from 1 to 11 kPa (N from 0.27 to $3 \cdot 10^{24} \text{ m}^{-3}$).
- U up to 6 kV.

The uncertainty of the absolute electrode spacing is $\pm 200 \mu\text{m}$, but care was taken that the relative electrode spacing can be controlled reproducibly to $\pm 6 \mu\text{m}$. Due to the regression methods described below, reproducibility of setting U and p is not important. The uncertainty of absolute (E/N) -values depends on the *relative* error of the U -, p - and temperature-measurements, and on the uncertainty of electrode spacing. In the present setup the uncertainty of (E/N) is typically $\pm 1.0\%$ to $\pm 1.5\%$.

3.2. Analysing electron currents

Two examples of current waveforms are shown in figure 2. The initial broadening of the swarm σ_0 and its starting time t_0 are obtained by fitting (6) to the time derivative of the rising edge of the waveform. A tentative fit of (10) with four free parameters to the whole waveform produces preliminary values n_0 , T , ν_{eff} , τ_D . Using these preliminary values the waveform is partitioned into intervals which are analysed individually. The ion current

amplitude A_{ion} is determined at T_4 , and the ion current $I_{\text{ion}}(t)$ is subtracted from the waveform using (15). The electron current $I_e(t)$ is analysed by fitting expressions of only two free parameters. From the interval T_1 to T_2 , as shown in figure 2, the definitive values n_0 and ν_{eff} are determined. This allows the injected electrons to settle to an equilibrium state (see also sections 3.4.1 and 6.1) and eliminates the influence of minor field inhomogeneities at the electrodes. Another fit to $I_e(t)$ between T_2 and T_3 produces T and τ_D .

This analysis relates one set of electron current parameters $(T, \nu_{\text{eff}}, \tau_D)$ to one set of experiment parameters (N, d, U) .

3.3. Algorithm of measurement sequence

A particular gas sample is examined systematically in a sequence of measurements. At different settings of the experiment parameters (N, d, U) current waveforms are sampled, and typically a certain value (E/N) is implemented for several different combinations of (N, d, U) . For the presently adopted procedures the admissible range of (E/N) values is limited. A minimum value $(E/N)_{\text{min}}$ is required for obtaining a measurable current amplitude, and an upper limit $(E/N)_{\text{max}}$ is reached when ionization is strong and substantial ion currents are observable. The value $(E/N)_{\text{min}}$ is determined by the experimenter prior to starting the measurement sequence, but $(E/N)_{\text{max}}$ is a priori unknown, and it depends on Nd and on the sample gas. However, the algorithm of the measurement sequence requires a termination criterion, which can be defined as a maximum number of electrons $n_{\text{max}} \approx 10^8$ at the end of electron transit. Whenever n_{max} is observed the algorithm has reached the upper limit $(E/N)_{\text{max}}$ and terminates the sequence.

In principle the algorithm consists of three nested loops. The outer loop iterates over a list of gas concentrations $N_1..N_{\text{end}}$, the second loop iterates over a list of electrode distances $d_1..d_{\text{end}}$. In the inner loop the voltage U is set, then a current waveform is recorded and analysed for the number of electrons, which arrive at the anode. Initially the inner loop implements $(E/N)_{\text{min}}$, and iteratively U is increased. The inner loop terminates when n_{max} is exceeded.

Typically one sequence consists of measurements at four d -values and four N -values, and one obtains about two hundred current waveforms, which are analysed individually.

3.4. Regression and normalization of swarm parameters

Paschen's relation states that all swarms are similar under the condition $Nd = \text{const.}$ Then ν_{eff} and τ_D can be normalized by the gas density N for obtaining the temporal swarm parameters (ν_{eff}/N) and $N\tau_D$, and for one particular gas mixture these swarm parameters are a function of (E/N) only.

When swarm parameter data are produced by normalizing and averaging measurements at similar (E/N) , possible offsets or systematic disturbances of measured quantities will be manifested in the results. In order to overcome these drawbacks,

Figure 3. Individual values of effective ionization rates ν_{eff} of one measurement sequence for N_2 . The box near 150 Td marks the values that appear in the inset. The inset shows the measured ($\circ$) and fitted (+) values in the regression of $\nu_{\text{eff}}d$ over Nd for obtaining (ν_{eff}/N) at 152.5 Td.

regression methods have been developed for obtaining w and D_L from time-of-flight measurements [12]. The present swarm model permits regression analyses also for PT measurements, and we generalize the approach to include measurements at different pressures and slightly different (E/N) -values.

For this section we introduce the abbreviations $R = (\nu_{\text{eff}}/N)$ and $D = N\tau_D$, and the subscript is omitted from ν_{eff} in order to improve the readability of equations.

Be $(E/N)_0$ an arbitrary value within the (E/N) -range of the measurement sequence, and be $d(E/N)$ much smaller than the (E/N) -range. Then, between $(E/N)_0 - d(E/N)$ and $(E/N)_0 + d(E/N)$ are $j = 1, \dots, J$ sets of measured quantities $(N_j, (E/N)_j, d_j, T_j, \nu_j, \tau_{Dj})$ from the sequence. These J sets are treated in three independent analyses for obtaining the values w_0 , R_0 , D_0 for $(E/N)_0$. An example of this procedure is given in figure 3, where the individual ν_{eff} from one sequence are shown. For $(E/N)_0 = 152.5$ Td the J selected values are marked by a box. The inset of figure 3 shows the regression analysis for R (see section 3.4.2) on the basis of these J values.

3.4.1. Swarm velocity w We assume that w depends linearly on (E/N) in the selected very narrow (E/N) -range.

$$w_j = w_0 + dw [(E/N)_j - (E/N)_0] \quad (19)$$

This dependence is inserted into (5) for obtaining a linear regression of d_j on T_j , which is a generalization of the usual difference method of time-of-flight experiments.

$$d_j = T_j \{w_0 + dw [(E/N)_j - (E/N)_0]\} + d_0, \quad (20)$$

From (20) the value of the swarm velocity w_0 and its tangent $\frac{dw_0}{d(E/N)}$ at $(E/N)_0$ can be obtained. The constant term d_0 in (20) accounts for the measurement uncertainty of the

gap distance and for the fact, that the swarm is only initiated immediately in front of the cathode and equilibrates during the initial drift, as it was pointed out by Townsend [13].

3.4.2. Effective reaction rate (ν_{eff}/N) For all the distances d_j one can write

$$\nu_j d_j = R N_j d_j . \quad (21)$$

Equation (21) states that the validity of Paschen's similarity relation also implies that $\nu_{\text{eff}} d = \text{const}$ is a similarity relation of electron swarms.

When R is a linear function of (E/N) in the selected small interval, then the regression equation for R_0 at $(E/N)_0$ is

$$\nu_j d_j = N_j d_j \{ R_0 + dR_0 [(E/N)_j - (E/N)_0] \} + (\nu d)_0 , \quad (22)$$

from which the tangent $\frac{dR_0}{d(E/N)}$ at $(E/N)_0$ is also obtained. As in (20) a constant term $(\nu d)_0$ is included.

3.4.3. Diffusion time $N\tau_D$ The considerations, which lead to (20) and (22) also apply for D .

$$\frac{\tau_{Dj}}{d_j} = \frac{D}{N_j d_j} \quad (23)$$

Expression (23) implies the similarity relation $\tau_D/d = \text{const}$, and it defines the regression equation for D_0

$$\frac{\tau_{Dj}}{d_j} = \frac{D_0 + dD_0 [(E/N)_j - (E/N)_0]}{N_j d_j} + (\tau_D/d)_0 , \quad (24)$$

and the tangent $\frac{dD_0}{d(E/N)}$. The constant term $(\tau_D/d)_0$ is due to the initial broadening of the electron swarm, for which the laser pulse duration is one reason. As pointed out by Wetzer [14], the deviation of the electrodes from parallel alignment gives also rise to an offset in the measurement of τ_D .

4. Benchmark data

Ar, N₂ and CO₂ were chosen for benchmarking our methods, because for these gases the electron drift velocity and ND_L have been measured with highest precision using time-of-flight methods [12, 15, 16], and because (α_{eff}/N) is well known from steady-state Townsend measurements [17, 18].

On the basis of these measurements, and including results from beam methods [19, 20], cross section sets were established for the benchmark gases [15, 21, 22], and their electron transport parameters can be calculated or simulated. The calculations of benchmark data were done with a Boltzmann solver using the two-term approximation (Bolsig+ [8] version 11.2011, settings: Precision 10^{-13} , Convergence 10^{-5} , Iterations 100, default SIGLO cross sections). Simulations were done with a Monte Carlo method (Magboltz [23] version 9.0.1, setting $4 \cdot 10^8$ real collisions, Biagi's cross sections). It has to

Figure 4. Measured ($\circ$) and fitted (+) values in the regression analyses for CO_2 , which include measurements at 80 and 85 Td. (a) regression of $\nu_{\text{eff}} d$ over Nd used for obtaining (ν_{eff}/N) at 82.5 Td. (b) regression of τ_D/d over $(Nd)^{-1}$ used for obtaining $N\tau_D$ at 82.5 Td.

be noted that Bolsig+ and Magboltz use different cross section sets. Although Magboltz can produce dedicated PT parameters for higher (E/N) -values, in the (E/N) -range of the present study the general transport coefficients must be used.

For N_2 and CO_2 additionally experimental reference data was considered, but we restricted our options to publications containing tabulated numerical data. We selected the PT measurements by de Urquijo in N_2 [24]. Raju's compilation of swarm parameters [25], which was the most recent available to us, gives Haydon's (α_{eff}/N) for N_2 [17], and Bhalla's (α_{eff}/N) for CO_2 [18].

5. Results

Measurement sequences were made in Ar (purity 6.0) at pressures between 2.1 and 10.5 kPa, in N_2 (5.0) between 1.5 and 10.5 kPa. In CO_2 (5.0) we made an overview measurement with step-size 5 Td at pressures between 2.5 and 10.5 kPa. A second

Table 1. Swarm parameters of Ar. Column units are Td, 10^3 m/s, 10^{-19} m³/s and 10^{16} s/m³.

(E/N)	w	(ν_{eff}/N)	$N\tau_D$
18	17.16	$-0.48 \pm .02$	2.41
20	18.91	$-0.30 \pm .02$	2.07
22	20.65	$0.16 \pm .02$	1.78
24	22.43	$0.64 \pm .02$	1.55
26	24.11	$1.68 \pm .03$	1.35
28	25.84	$2.96 \pm .04$	1.19
30	27.84	$4.85 \pm .06$	1.03
32	29.81	$7.39 \pm .09$	0.92
34	31.53	$9.07 \pm .09$	0.85
36	32.24	$11.39 \pm .04$	0.62
38	34.10	$17.26 \pm .03$	0.51

investigation of the (E/N) -range 20 to 45 Td was done with step-size 1 Td at pressures between 5.0 and 9.1 kPa. The gas pressure and temperature (293 to 300 K) were simultaneously measured for every recorded waveform.

The measurements were carried out as described in section 3.3 and analysed individually according to the procedure given in section 3.2. For example, the sequence data of ν_{eff} in N₂ are given in figure 3. The sequence data were then sampled using the regression methods of section 3.4 for obtaining w , (ν_{eff}/N) and $N\tau_D$. Both sequences of CO₂ have been merged and treated together by the regression analyses. Examples of regression analyses are presented in figures 3 and 4. Figure 4 includes measurements at 80 Td, where attachment dominates ($(\nu_{\text{eff}}/N) < 0$), and at 85 Td, where ionization dominates. Thus, two distinct branches are apparent in panel (a), one with positive and one with negative slope. In panel (b) two branches coincide, because electron diffusion is practically the same at 80 and 85 Td. The inset of figure 3 includes measurements at 150 and 155 Td, and two branches with slightly different slopes are discernible in the analysis used for obtaining (ν_{eff}/N) at 152.5 Td.

The evaluated swarm parameters of the three gases are listed in tables 1, 2 and 3. The 99% confidence intervals of the least-squares fit parameters were interpreted as statistical errors of the regression analyses. The tables include the statistical error of (ν_{eff}/N) , where it is larger than the rounding error. The statistical errors of w are about 0.2%, and less than 2% for $N\tau_D$. The uncertainty of the (E/N) values is less than 1.5%.

In order to compare our results to benchmark data, the electron flux velocity v_d and mobility μN was estimated from our w -data according to Tagashira's definition for time-of-flight experiments [6].

$$\mu N = \frac{v_d}{(E/N)} = \frac{w}{(E/N)} \sqrt{1 - (\nu_{\text{eff}}/N) (N\tau_D)} \quad (25)$$

Over the range of parameters considered here (tables 1-3) the difference between v_d and

Table 2. Swarm parameters of N_2 . Column units are Td, 10^3 m/s, 10^{-18} m³/s and 10^{14} s/m³.

(E/N)	w	(ν_{eff}/N)	$N\tau_D$
45.0	56.03	−0.04	6.09
55.0	65.11	−0.04	4.73
65.0	74.21	−0.07 ± .01	4.50
75.0	83.58	−0.06 ± .01	4.47
85.0	93.16	0.10 ± .01	4.25
95.0	103.0	0.53 ± .01	3.96
102.5	110.7	1.09 ± .01	3.76
107.5	115.9	1.60 ± .02	3.57
112.5	121.0	2.44 ± .02	3.45
117.5	126.0	3.29 ± .04	3.29
122.5	131.2	4.60 ± .04	3.17
127.5	136.4	5.84 ± .07	2.95
132.5	141.6	7.85 ± .07	2.82
137.5	147.1	10.52 ± .13	2.65
142.5	152.4	13.25 ± .12	2.56
147.5	156.9	16.48 ± .17	2.48
152.5	162.1	19.99 ± .18	2.40
157.5	167.0	22.66 ± .12	2.31
162.5	172.3	27.09 ± .14	2.23
167.5	177.3	29.72 ± .16	2.23
172.5	182.3	34.96 ± .21	2.10
177.5	186.6	38.34 ± .22	2.06

w is generally below 0.5%. Using v_d the effective ionization coefficient (α_{eff}/N) was estimated by

$$(\alpha_{\text{eff}}/N) = \frac{(\nu_{\text{eff}}/N)}{v_d} . \quad (26)$$

Our electron mobilities μN and effective ionization coefficients (α_{eff}/N) are presented in figure 5, together with the reference data introduced in section 4.

The present $N\tau_D$ -results are shown in figure 6 as cubic spline curves, which were constructed from the $N\tau_D$ -data of tables 1, 2 and 3 and from the tangents $d(N\tau_D)/d(E/N)$ as defined in (24). The ND_L -results of Bolsig+ and Magboltz were included in figure 6 using

$$N\tau_D = \frac{2}{v_d^2} ND_L . \quad (27)$$

In CO_2 the critical field strength $(E/N)_{\text{crit}} = 81.6 \pm .9$ Td was determined from our (ν_{eff}/N) -data, and $(E/N)_{\text{crit}} = 82.1 \pm .3$ Td follows from Magboltz results.

Table 3. Swarm parameters of CO₂. Column units are Td, 10³ m/s, 10⁻¹⁸ m³/s and 10¹⁴ s/m³.

(E/N)	w	(ν_{eff}/N)	$N\tau_D$
21.0	66.88	-0.09	13.1
23.0	74.32	-0.07	9.29
25.0	80.37	-0.09	7.16
27.0	84.96	-0.08	5.68
29.0	88.70	-0.07	4.93
32.0	93.12	-0.12	4.67
35.0	96.72	-0.15	4.79
38.0	99.65	-0.22	4.45
42.5	103.5	$-0.40 \pm .01$	4.29
47.5	107.1	$-0.65 \pm .01$	3.68
52.5	110.6	$-0.90 \pm .01$	3.58
57.5	114.1	$-1.14 \pm .01$	3.51
62.5	117.5	$-1.31 \pm .01$	3.51
67.5	120.9	$-1.34 \pm .01$	3.53
72.5	124.3	$-1.13 \pm .01$	3.52
77.5	127.8	$-0.65 \pm .02$	3.49
82.5	131.2	$0.17 \pm .03$	3.48
87.5	134.8	$1.36 \pm .04$	3.48
92.5	138.5	$2.92 \pm .06$	3.51
97.5	142.4	$5.00 \pm .07$	3.53
102.5	146.3	$7.37 \pm .10$	3.53
107.5	150.2	$10.17 \pm .14$	3.53
112.5	154.2	$14.03 \pm .20$	3.61
117.5	157.8	$18.74 \pm .29$	3.68
122.5	161.8	$23.47 \pm .32$	3.64

6. Discussion

6.1. Waveforms

Figure 2 demonstrates in the current measurements strong effects of processes in the immediate vicinity of the cathode. These effects depend on the sample gas. In Ar the current shows a sharp initial peak. This is probably so because the initial electrons are accelerated by the electric field without many collisions, due to the Ramsauer minimum of the Ar momentum transfer cross section, thus giving rise to a high current. As soon as electrons have reached substantial kinetic energy (> 0.5 eV), elastic collisions become important and electrons are also backscattered towards the cathode. In contrast, in CO₂ the momentum transfer cross section is highest for slow electrons and the threshold of inelastic processes is 80 meV, and therefore it takes longer to accelerate the initial electrons. However, in both gases the swarm equilibrates after 20 to 30 ns, which is consistent with the relaxation of transport parameters found in [7].

Generally, after swarm equilibration there is excellent agreement between measured

currents and the ones derived from our swarm model, as can be seen from figure 2.

6.2. Advantage of regression methods

(ν_{eff}/N) is supposed to approach zero for $(E/N) < 85$ Td in N_2 , and for $(E/N) < 20$ Td in Ar. In these parameter ranges our methods determined small negative values of (ν_{eff}/N) , as can be seen from tables 1 and 2. In the present setup the detection limit for (ν_{eff}/N) should be $\pm 5 \cdot 10^{-20}$ m³/s, and the regression permits measurements of relatively small (ν_{eff}/N) .

Another advantage of using regression methods can be seen from the lower ν_{eff} -branch in figure 4 (a). The observed ν_{eff} are positive for unknown reasons. If (ν_{eff}/N) was determined by averaging the values of the lower ν_{eff} -branch, then (ν_{eff}/N) would be positive for CO_2 at 80 Td, and one would conclude $(E/N)_{\text{crit}}$ of CO_2 was below 80 Td. However, our method determines (ν_{eff}/N) from the slope of the ν_{eff} -branches, and $(E/N)_{\text{crit}} = 81.6 \pm .9$ Td is obtained for CO_2 . The regression method is especially well suited for determining $(E/N)_{\text{crit}}$ of the sample gas.

Diffusion time constants $N\tau_{\text{D}}$ are determined by the regression with statistical errors of 1% to 2%, whereas averaging methods produce statistical errors in the order of 10% for ND_{L} [24].

The regression method permits plotting swarm parameters as spline curves, as it was done in figure 6, and the plot can be analyzed for data consistency. For example, in our $N\tau_{\text{D}}$ -results of CO_2 between 30 and 40 Td the tangents deviate from the slope of the global curve. An inconsistency of $N\tau_{\text{D}}$ appeared where the two measurement sequences have been merged.

6.3. Comparison to reference data and simulations (refer to figures 5 and 6)

6.3.1. Ar Our results of μN and (α_{eff}/N) lie in between the values from Magboltz and Bolsig+, as shown in figure 5. As presented in figure 6 (a), the $N\tau_{\text{D}}$ -results of SPaRX agree to within 10% to the longitudinal values from Magboltz.

6.3.2. N_2 For μN , the difference between our results and Magboltz is $\leq 1\%$. Also Bolsig+ produced values within $\pm 1\%$ for $(E/N) < 100$ Td, but for $(E/N) > 100$ Td the present results are up to 10% higher than the values from Bolsig+. The SPaRX results are in excellent qualitative agreement with Urquijo's data, but our μN are 5% higher. Perhaps this difference appears because our approach allows for the time required to accelerate and equilibrate the swarm.

The (α_{eff}/N) -values from SPaRX are nearly the same as the Magboltz values for $(E/N) < 150$ Td. Above 150 Td our results are slightly below the Magboltz results, but reproduce Raju's recommended value at 165 Td. The SPaRX data in the range 110 to 180 Td are significantly higher than the values from Bolsig+ and de Urquijo. Between 100 and 150 Td our (α_{eff}/N) are also higher than Raju's recommendations.

Figure 5. Present results (—) of electron mobility (a) and effective ionization coefficient (b) of Ar, N₂ and CO₂. Reference data for selected (E/N) -values are Bolsig+ (---), Urquijo's (.....) for N₂, Magboltz (×), and Raju's compilations [17, 18, 25] (□). The inset shows (α_{eff}/N) of Ar on a magnified scale.

Our $N\tau_D$ -results above 80 Td agree to the *transversal* values from Magboltz. Below 80 Td the SParX-results show a tendency towards halfway the transversal and the longitudinal values from Magboltz, but our measured $N\tau_D$ are always higher than the Magboltz results of longitudinal diffusion in the (E/N) -range of the present study.

6.3.3. CO₂ There is a prominent peak of μN at 25 Td, which was well resolved by our measurements. Our μN reproduce Raju's recommendations and provide complementary data between 20 and 50 Td. Please note that in this (E/N) -range tabulated μN -values were previously given by Elford [16], which are well reproduced by our measurements. Again there is excellent agreement with Magboltz results, only between 50 and 80 Td our values are 3% smaller.

The small negative values (α_{eff}/N) for $(E/N) \leq 50$ Td from SParX match to Magboltz results. The two methods also agree on $(E/N)_{\text{crit}}$. However, the minimum

Figure 6. Present results (—) for the diffusion time constant of (a) Ar, N_2 and (b) CO_2 . Reference data are Bolsig+ (---), Magboltz longitudinal ($\times$) and transversal ($+$) values for selected values of (E/N) .

of (α_{eff}/N) from SParX around 65 Td is slightly above the Magboltz value, and for $(E/N) > 100$ Td our results are 5% lower. Raju’s recommended values appear much too high between 80 and 100 Td, and Raju does not allow for negative values of (α_{eff}/N) and thus $(E/N)_{\text{crit}}$ can not be interpolated.

The SParX data for $N\tau_D$ and the longitudinal values from Magboltz both show a prominent fall of $N\tau_D$ between 20 and 50 Td, which is not present in the values derived from Bolsig+. However, on the whole (E/N) -range our $N\tau_D$ -data are significantly higher than the longitudinal diffusion obtained from Magboltz, and also deviate from data based on transverse diffusion, except around 120 Td, where they become similar.

6.3.4. General The (α_{eff}/N) -data for N_2 and CO_2 shown in figure 5 (b) seem to make up two groups in each gas. On the one hand there seems to be agreement between Urquijo’s measurements, Bolsig+ and Raju’s recommendations, on the other there is very good agreement between SParX and Magboltz. Moreover, there is no uniform

tendency for the difference between the two groups; for N_2 the results of Magboltz and SParX are higher than the other data, but lower for CO_2 . Considering the measurements it is obvious that the numerical results and the cross sections on which they are based should be subject of further critical assessment.

It should be remembered that Bolsig+ produces isotropic flux diffusion, that Magboltz produces longitudinal and transversal flux diffusion, and that only longitudinal bulk diffusion is obtained from SParX. From figure 6 (a) the highly anisotropic diffusion in Ar becomes apparent, with only small differences between longitudinal bulk and flux values, and noticeably higher transversal values by Magboltz and $N\tau_D$ produced by Bolsig+. These results indicate that anisotropic diffusion is a successful concept for Ar in the (E/N) -range below 50 Td. In this (E/N) -range elastic processes dominate [15]. Whereas for N_2 and CO_2 longitudinal bulk diffusion (SParX data) is substantially larger than the longitudinal flux values produced by Magboltz. From figure 6 (b) it becomes apparent that for CO_2 diffusion could be reproduced neither by the Boltzmann solver nor by the Monte Carlo method. Future studies on N_2 and CO_2 could also consider effects of inelastic electron-molecule interactions on bulk diffusion.

7. Conclusions

We presented the SParX pulsed Townsend (PT) setup, the operation of which is entirely computer controlled. As its components and the processes are robust and reliable, trustworthy experiments can run in an autonomous mode.

7.1. Model and analysis method

The resulting quantities of PT measurements are the electron transit time T , the ionization rate ν_{eff} , and the diffusion time constant τ_D . For any future work on the basis of the PT method we recommend using these temporal parameters, because the previously used spatial parameters required estimating the electron velocity prior to the analysis of measured waveforms. For each of the parameters T , ν_{eff} , τ_D one regression method was presented. These regression methods were applied to our measurements for obtaining the swarm velocity w and the density normalized swarm parameters (ν_{eff}/N) and $N\tau_D$. Our swarm model and regression methods explicitly take into account non-idealized conditions for electron swarms within the experimental setup, namely effects of electrode misalignment, of laser pulse duration, ion currents, sensor offsets, and the initial lack of swarm equilibrium. It was demonstrated that these regression methods are superior to averaging methods and should be preferred for analyses of PT measurements.

7.2. Recommendations on Ar, N_2 and CO_2

Electron swarm parameters have been measured in Ar, N_2 and CO_2 . Tabulated data of our measurements are provided, which might later be compared to the output of dedicated simulations. The electron mobilities obtained by our method precisely

reproduce literature data. It was shown that our electron transport parameters are in excellent agreement with electron transport parameters from Monte Carlo simulations, except for noticeable differences for diffusion in N_2 and CO_2 . In applications, where electron diffusion is important, anisotropic diffusion should be used for Ar, but isotropic diffusion should be preferred for N_2 above 80 Td. For CO_2 experimental data of longitudinal diffusion should be preferred for applications in the parameter range of the present study. Because of evident discrepancies between hitherto recommended and new experimental values one would advise critical assessment and revision of recommendations for ionization of N_2 below 170 Td, and for attachment and ionization of CO_2 below 120 Td, in order to improve the input data for plasma models.

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