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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 (PT) 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 PT method are identified, and novel regression methods are presented for obtaining electron swarm parameters from PT 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 with 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.

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 [2–4]. 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 the 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]

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

where \(\alpha_{\text{eff}}\) 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 \(\alpha_{\text{eff}}\) and \(D_L\) cannot be determined directly without the 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 analysed 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, N\(_2\) and CO\(_2\), then our results for these gases are presented and compared with 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 [6–8]. It is necessary to transform the spatial swarm model (1) to the temporal domain using the relations

\[
\alpha_{\text{eff}} \, dz = v_{\text{eff}} \, dt, \tag{2}
\]

\[
2 D_{\text{eff}} t = w^2 \tau_0 t, \tag{3}
\]

which introduce the effective reaction rate \( v_{\text{eff}} \) and the diffusion time constant \( \tau_0 \). Inserting these relations in (1) yields

\[
\frac{d n_e(z, t)}{dz} = \frac{n_0 \exp(v_{\text{eff}} t)}{w \sqrt{2\pi \tau_0}} \exp \left( -\frac{(t - T)^2}{2 \tau_0} \right). \tag{4}
\]

With

\[
w = \frac{dz}{dt} \tag{5}
\]

one obtains a temporal electron number distribution \( d n_e / dt \) 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,

\[
\frac{d n_e(d_0, t)}{dt} = \frac{n_0}{\sqrt{2\pi \sigma_0^2}} \exp \left( -\frac{(t - t_0)^2}{2 \sigma_0^2} \right), \tag{6}
\]

with initial electron number \( n_0 \) and initial temporal broadening \( \sigma_0 \). 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 \( d = d_0 + w T \). There the electron distribution is

\[
\frac{d n_e(d, t)}{dt} = \frac{n_0 \exp (v_{\text{eff}} (t - t_0))}{\sqrt{2\pi \sigma^2(t)}} \exp \left( -\frac{(t - t_0 - T)^2}{2 \sigma^2(t)} \right). \tag{7}
\]

The temporal broadening \( \sigma(t) \) increases with the diffusion time constant \( \tau_0 \).

\[
\sigma^2(t) = \sigma_0^2 + \tau_0 (t - t_0). \tag{8}
\]

The total number of electrons changes at an effective rate \( v_{\text{eff}} \), which accounts for electron attachment and ionization. The model collapses with electron detachment, cathodic feedback or photoionization by secondary photons. However, \( v_{\text{eff}} \) 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].

\[
I_e(t) = \frac{q_0}{T} \int_0^d n_e(z, t) \, dz, \tag{9}
\]

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].

\[
I_e(t) = \frac{n_0 q_0}{T} \frac{1 + A_L(t)}{2} \frac{1 - A_D(t)}{2} A_r(t) \tag{10}
\]

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

\[
A_r(t) = \exp \left( v_{\text{eff}} (t - t_0) \right). \tag{11}
\]

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:

\[
A_L(t) = \begin{cases} 
\text{erf} \left( \frac{t - t_0}{\sqrt{2 \sigma_0^2}} \right) & \text{for } t \leq t_0, \\
\text{erf} \left( \frac{t - t_0}{\sqrt{2 \sigma_0^2 + 2 \tau \tau (t - t_0)}} \right) & \text{for } t > t_0.
\end{cases} \tag{12}
\]

For an electrode gap \( d = d_0 + w T \) the amplitude factor \( A_D(t) \) is

\[
A_D(t) = \text{erf} \left( \frac{t - t_0 - T}{\sqrt{2 \sigma_0^2 + 2 \tau \tau (t - t_0)}} \right). \tag{13}
\]

The experimental conditions can be chosen such that \( T \) is much larger than \( \tau \). 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 \( A_r(t) \) 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 $v_{\text{eff}}$.

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

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

$$I_{\text{ion}}(t) = \begin{cases} 0 & \text{for } v_{\text{eff}} \leq 10^3/s, \\ A_{\text{ion}} \frac{t - t_0}{T} \exp(v_{\text{eff}}(t - t_0)) - 1 & \text{for } v_{\text{eff}} > 10^3/s, \end{cases}$$

and for $t > T$ it is

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

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

### 3. Methods

#### 3.1. PT 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–11 kPa. In the chamber two polished stainless-steel electrodes with radius $r_0 = 56$ mm represent a $\pi/2$-Rogowski geometry for their ideal spacing $z_0 = 15$ mm. The parametrization of their profile $(r, z)$ is

$$r = [0 ... r_0],$$

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

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 $\mu$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 $\mu$J pulse energy at 20 Hz repetition rate, producing about $3 \times 10^3$ 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 $R_\text{a}$ = 150 $\Omega$ and $C_\text{a} = 2.7 \mu$F between the dc high voltage source and the cathode. The low voltage circuit was optimized for high signal bandwidth (200 MHz) and high fidelity. The anode is connected to a transimpedance amplifier (FEMTO DHPACA-100) with gain resistor $R_a$. For recording electron currents we set $R_a = 10^2$ or $10^3 \text{V A}^{-1}$, and ion currents can be recorded with $R_a = 10^4$ or $10^5 \text{V A}^{-1}$. The oscilloscope (R&S RTO) is equipped with a 10 GS s$^{-1}$ 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.

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 293–300 K, which are measured but not controlled. $(E/N)$ is controlled by the experiment parameters $d$, $p$, $U$. For this study their range of values is:

- $d$ from 9 to 18 mm.
- $p$ from 1 to 11 kPa ($N$ from 0.27 to $3 \times 10^4$ m$^{-3}$).
- $U$ up to 6 kV.

The uncertainty of the absolute electrode spacing is $\pm 200 \mu$m, but care was taken that the relative electrode spacing can be controlled reproducibly to $\pm 6 \mu$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 $r_0$ 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 $n_0$, $T$, $v_{\text{eff}}$, $T_0$. Using these preliminary values the waveform is partitioned into intervals which are analysed individually. The ion current amplitude $A_{\text{ion}}$ is determined at

![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 $R_a$.](image-url)
The latter current was divided by 10 and time shifted by 0.2 μs. Averages of measured waveforms (—) and the fit (⋯⋯⋯) of (10) coincide well. T marks the electron transit time in Ar, and the markers T1 to T4 are explained in section 3.2.

\[ I_{\text{ion}}(t) \] 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 \( v_{\text{eff}} \) are determined. This allows the injected electrons to settle to an equilibrium state (see also sections 3.4.1 and 6.1) and 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 \( \tau_D \).

This analysis relates one set of electron current parameters \((T, v_{\text{eff}}, \tau_D)\) 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 experimentor 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 \(n_{\text{max}} \approx 10^5 \) at the end of electron transit. Whenever \(n_{\text{max}}\) 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 \(N_1, \ldots, N_{\text{end}}\), the second loop iterates over a list of electrode distances \(d_1, \ldots, d_{\text{end}}\). 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 \(n_{\text{max}}\) 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

Puschen’s relation states that all swarms are similar under the condition \(Nd = \text{const}\). Then \(v_{\text{eff}} \) and \(\tau_D\) can be normalized by the gas density \(N\) for obtaining the temporal swarm parameters \((v_{\text{eff}}/N)\) 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, 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 \(R = (v_{\text{eff}}/N)\) and \(D = N\tau_D\), and the subscript is omitted from \(v_{\text{eff}}\) 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 \(j = 1, \ldots, J\) sets of measured quantities \((N_j, (E/N)_j, d_j, T_j, v_j, \tau_D)\) 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 \(v_{\text{eff}}\) 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:

\[
w_j = w_0 + dw \left[ (E/N)_j - (E/N) \right] \tag{19}
\]

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

\[
d_j = T_j \left[ w_0 + dw \left( (E/N)_j - (E/N) \right) \right] + d_0. \tag{20}
\]

From (20) the value of the swarm velocity \( w_0 \) and its tangent \( \frac{dR_0}{d(E/N)} \) 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 \((\nu_{\text{eff}}/N)\). For all the distances \( d_j \) one can write

\[
v_j d_j = R N_j d_j. \tag{21}
\]

Equation (21) states that the validity of Paschen’s similarity relation also implies that \( \nu_{\text{eff}} = \text{const} \) 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

\[
v_j d_j = N_j d_j \left[ R_0 + dR_0 \left( (E/N)_j - (E/N) \right) \right] + (vd)_0,
\]

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

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

\[
\frac{\tau_{Dj}}{d_j} = \frac{D}{N_j d_j}. \tag{23}
\]

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

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

and the tangent \( \frac{dD_0}{d(E/N)} \). The constant term \( \left( \tau_D/d_j \right)_0 \) 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 also gives rise to an offset in the measurement of \( \tau_D \).

4. Benchmark data

\( \text{Ar, N}_2 \) and \( \text{CO}_2 \) were chosen for benchmarking our methods, because for these gases the electron drift velocity and \( ND_\text{eff} \) have been measured with highest precision using time-of-flight methods [12, 15, 16], and because \((\alpha_{\text{eff}}/N)\) 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 SIGLIO cross sections). Simulations were done with a Monte Carlo method (Magboltz [23] version 9.0.1, setting \(4 \times 10^8\) real collisions, Biagi’s cross sections). It has to 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 this study the general transport coefficients must be used.

For \( \text{N}_2 \) and \( \text{CO}_2 \) 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 \( \text{N}_2 \) [24]. Raju’s compilation of swarm parameters [25], which was the most recent available to us, gives Haydon’s \((\alpha_{\text{eff}}/N)\) for \( \text{N}_2 \) [17], and Bhalla’s \((\alpha_{\text{eff}}/N)\) for \( \text{CO}_2 \) [18].

5. Results

Measurement sequences were made in \( \text{Ar} \) (purity 6.0) at pressures between 2.1 and 10.5 kPa, in \( \text{N}_2 \) (5.0) between 1.5 and 10.5 kPa. In \( \text{CO}_2 \) (5.0) we made an overview measurement with step-size 5 Td at pressures between 2.5 and 10.5 kPa. A second investigation of the \((E/N)\)-range 20–45 Td was done with step-size 1 Td at pressures between 5.0 and 9.1 kPa. The gas pressure and temperature (293–300 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 \( \nu_{\text{eff}} \) in \( \text{N}_2 \) are shown in figure 3. The sequence data were then sampled using the regression methods of section 3.4 for obtaining \( \nu_0, (\nu_{\text{eff}}/N) \) and \( N \tau_D \). Both sequences of \( \text{CO}_2 \) 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 \((\nu_{\text{eff}}/N)\) at 152.5 Td.

The evaluated swarm parameters of the three gases are listed in tables 1–3. 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 \((\nu_{\text{eff}}/N)\), 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 \( \mu N \) were estimated...
of-flight experiments \[6\].

\[
\begin{align*}
\mu N &= \frac{v_d}{(E/N)} = \frac{w}{(E/N)} \sqrt{1 - (v_{ei}/N)} (N_{TD}).
\end{align*}
\]  

Over the range of parameters considered here (tables 1–3) the difference between \(v_d\) and \(w\) is generally below 0.5%. Using \(v_d\)

from our \(w\)-data according to Tagashira’s definition for time-of-flight experiments \[6\].

\[\frac{(E/N)}{(E/N)} w (v_{ei}/N) N_{TD}\]

\[\begin{array}{llll}
(10^4 \text{ m} \text{s}^{-1}) & (10^{-18} \text{ m}^3 \text{s}^{-1}) & (10^{14} \text{ m} \text{s}^{-1}) & \\
18 & 17.16 & -0.48 \pm 0.02 & 2.41 \\
20 & 18.91 & -0.30 \pm 0.02 & 2.07 \\
22 & 20.65 & 0.16 \pm 0.02 & 1.78 \\
24 & 22.43 & 0.64 \pm 0.02 & 1.55 \\
26 & 24.11 & 1.68 \pm 0.03 & 1.35 \\
28 & 25.84 & 2.96 \pm 0.04 & 1.19 \\
30 & 27.84 & 4.85 \pm 0.06 & 1.03 \\
32 & 29.81 & 7.39 \pm 0.09 & 0.92 \\
34 & 31.53 & 9.07 \pm 0.09 & 0.85 \\
36 & 32.24 & 11.39 \pm 0.04 & 0.62 \\
38 & 34.10 & 17.26 \pm 0.03 & 0.51 \\
\end{array}\]

Table 2. Swarm parameters of \(\text{N}_2\).

\[
\begin{array}{llll}
(E/N) & w & (v_{ei}/N) & N_{TD} \\
(\text{Td}) & (10^4 \text{ m} \text{s}^{-1}) & (10^{-18} \text{ m}^3 \text{s}^{-1}) & (10^{14} \text{ m} \text{s}^{-1}) \\
45.0 & 56.03 & -0.04 & 6.09 \\
55.0 & 65.11 & -0.04 & 4.73 \\
65.0 & 74.21 & -0.07 \pm 0.01 & 4.50 \\
75.0 & 83.58 & -0.06 \pm 0.01 & 4.47 \\
85.0 & 93.16 & 0.10 \pm 0.01 & 4.25 \\
95.0 & 103.0 & 0.53 \pm 0.01 & 3.96 \\
102.5 & 110.7 & 1.09 \pm 0.01 & 3.76 \\
107.5 & 115.9 & 1.60 \pm 0.02 & 3.57 \\
112.5 & 121.0 & 2.44 \pm 0.02 & 3.45 \\
117.5 & 126.0 & 3.29 \pm 0.04 & 3.29 \\
122.5 & 131.2 & 4.60 \pm 0.04 & 3.17 \\
127.5 & 136.4 & 5.84 \pm 0.07 & 2.95 \\
132.5 & 141.6 & 7.85 \pm 0.07 & 2.82 \\
137.5 & 147.1 & 10.52 \pm 0.13 & 2.65 \\
142.5 & 152.4 & 13.25 \pm 0.12 & 2.56 \\
147.5 & 156.9 & 16.48 \pm 0.17 & 2.48 \\
152.5 & 162.1 & 19.99 \pm 0.18 & 2.40 \\
157.5 & 167.0 & 22.66 \pm 0.12 & 2.31 \\
162.5 & 172.3 & 27.09 \pm 0.14 & 2.23 \\
167.5 & 177.3 & 29.72 \pm 0.16 & 2.23 \\
172.5 & 182.3 & 34.96 \pm 0.21 & 2.10 \\
177.5 & 186.6 & 38.34 \pm 0.22 & 2.06 \\
\end{array}\]

Table 3. Swarm parameters of \(\text{CO}_2\).

\[
\begin{array}{llll}
(E/N) & w & (v_{ei}/N) & N_{TD} \\
(\text{Td}) & (10^4 \text{ m} \text{s}^{-1}) & (10^{-18} \text{ m}^3 \text{s}^{-1}) & (10^{14} \text{ m} \text{s}^{-1}) \\
21.0 & 66.88 & -0.09 & 13.1 \\
23.0 & 74.32 & -0.07 & 9.29 \\
25.0 & 80.37 & -0.09 & 7.16 \\
27.0 & 84.96 & -0.08 & 5.68 \\
29.0 & 88.70 & -0.07 & 4.93 \\
32.0 & 93.12 & -0.12 & 4.67 \\
35.0 & 96.72 & -0.15 & 4.79 \\
38.0 & 99.65 & -0.22 & 4.45 \\
42.5 & 103.5 & -0.40 \pm 0.01 & 4.29 \\
47.5 & 107.1 & -0.65 \pm 0.01 & 3.68 \\
52.5 & 110.6 & -0.90 \pm 0.01 & 3.58 \\
57.5 & 114.1 & -1.14 \pm 0.01 & 3.51 \\
62.5 & 117.5 & -1.31 \pm 0.01 & 3.51 \\
67.5 & 120.9 & -1.34 \pm 0.01 & 3.53 \\
72.5 & 124.3 & -1.13 \pm 0.01 & 3.52 \\
77.5 & 127.8 & -0.65 \pm 0.02 & 3.49 \\
82.5 & 131.2 & 0.17 \pm 0.03 & 3.48 \\
87.5 & 134.8 & 1.36 \pm 0.04 & 3.48 \\
92.5 & 138.5 & 2.92 \pm 0.06 & 3.51 \\
97.5 & 142.4 & 5.00 \pm 0.07 & 3.53 \\
102.5 & 146.3 & 7.37 \pm 0.10 & 3.53 \\
107.5 & 150.2 & 10.17 \pm 0.14 & 3.53 \\
112.5 & 154.2 & 14.03 \pm 0.20 & 3.61 \\
117.5 & 157.8 & 18.74 \pm 0.29 & 3.68 \\
122.5 & 161.8 & 23.47 \pm 0.32 & 3.64 \\
\end{array}\]

the effective ionization coefficient \((v_{ei}/N)\) was estimated by

\[
(v_{ei}/N) = \frac{(v_{ei}/N)}{v_d}.  \tag{26}
\]

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

The present \(N_{TD}\)-results are shown in figure 6 as cubic spline curves, which were constructed from the \(N_{TD}\)-data of
As soon as electrons have reached substantial kinetic 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 20–30 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

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

Another advantage of using regression methods can be seen from the lower \( \nu_{\text{eff}} - \)branch in figure 4(a). The observed \( \nu_{\text{eff}} \) are positive for unknown reasons. If \( \nu_{\text{eff}} / N \) was determined by averaging the values of the lower \( \nu_{\text{eff}} - \)branch, then \( \nu_{\text{eff}} / N \) would be positive for \( \text{CO}_2 \) at 80 Td, and one would conclude \( (E/N)_{\text{crit}} \) of \( \text{CO}_2 \) was below 80 Td. However, our method determines \( \nu_{\text{eff}} / N \) from the slope of the \( \nu_{\text{eff}} - \)branches, and \( (E/N)_{\text{crit}} = 81.6 \pm 0.9 \text{Td} \) is obtained for \( \text{CO}_2 \). The regression method is especially well suited for determining \( (E/N)_{\text{crit}} \) of the sample gas. Diffusion time constants \( N \tau_0 \) are determined by the regression with statistical errors of 1–2%, whereas averaging methods produce statistical errors in the order of 10% for \( N \tau_0 \).

The regression method permits plotting swarm parameters as spline curves, as it was done in figure 6, and the plot can be analysed for data consistency. For example, in our \( N \tau_0 \)-results of \( \text{CO}_2 \) between 30 and 40 Td the tangents deviate from the slope of the global curve. An inconsistency of \( N \tau_0 \) 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 \( \mu N \) and \( \nu_{\text{eff}} / N \) lie in between the values from Magboltz and Bolsig+, as shown in figure 5. As presented in figure 6(a), the \( N \tau_0 \)-results of SPARX agree to within 10% to the longitudinal values from Magboltz.

6.3.2. \( N_2 \). For \( \mu N \), the difference between our results and Magboltz is \( \leq 1 \% \). Also Bolsig+ produced values within \( \pm 1 \% \) for \( (E/N) < 100 \text{Td} \), but for \( (E/N) > 100 \text{Td} \) 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 \( \mu N \) are 5% higher. Perhaps this difference appears because our approach allows for the time required to accelerate and equilibrate the swarm.

The \( \nu_{\text{eff}} / N \)-values from SPARX are nearly the same as the Magboltz values for \( (E/N) < 150 \text{Td} \). Above 150 Td our
values of Magboltz longitudinal $Raju's$ recommendations and provide complementary data was well resolved by our measurements. Our $\mu N$ tabulated and 80 Td our values are 3% smaller. There is excellent agreement with Magboltz results, only between 50 and 80 Td our values are 3% smaller.

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 transversal diffusion in the $(E/N)$-range of this study.

6.3.3. CO$_2$. There is a prominent peak of $\mu N$ at 25 Td, which was well resolved by our measurements. Our $\mu N$ reproduce Raju’s recommendations and provide complementary data between 20 and 50 Td. Please note that in this $(E/N)$-range tabulated $\mu N$-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 $((\alpha_{eff}/N)$ for $(E/N) \leq 50$ Td from SParX match to Magboltz results. The two methods also agree on $(E/N)_{crit}$. However, the minimum of $((\alpha_{eff}/N)$ 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 $(\alpha_{eff}/N)$ and thus $(E/N)_{crit}$ cannot 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 $(\alpha_{eff}/N)$-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 $\nu_{eff}$, and the diffusion time constant $\tau_D$. 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 $T$, $\nu_{eff}$, $\tau_D$ 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 \( \nu_{\text{eff}}/N \) and \( \nu \text{TD} \). 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 \( \text{Ar, N}_2 \) and \( \text{CO}_2 \)

Electron swarm parameters have been measured in \( \text{Ar, N}_2 \) and \( \text{CO}_2 \). Tabulated data of our measurements are provided, which might later be compared with 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 \( \text{N}_2 \) and \( \text{CO}_2 \). In applications, where electron diffusion is important, anisotropic diffusion should be used for \( \text{Ar} \), but isotropic diffusion should be preferred for \( \text{N}_2 \) above 80 Td. For \( \text{CO}_2 \) experimental data of longitudinal diffusion should be preferred for \( \text{N}_2 \) above 80 Td. For \( \text{CO}_2 \), the parameter range of this study. Because of evident discrepancies between hitherto recommended and new experimental values one would advise critical assessment and revision of recommendations for ionization of \( \text{N}_2 \) below 170 Td, and for attachment and ionization of \( \text{CO}_2 \) below 120 Td, in order to improve the input data for plasma models.

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References

[21] Anzai K et al 2012 Cross section data sets for electron collisions with \( \text{H}_2 \), \( \text{O}_2 \), \( \text{CO} \), \( \text{CO}_2 \), \( \text{N}_2 \) and \( \text{H}_2\text{O} \) Eur. Phys. J. D 66 36