Electrical breakdown in nitrogen at low pressure – physical processes and statistics

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Abstract. The results of investigation of the electrical breakdown in nitrogen, obtained in combined approach based on measuring of the current-voltage characteristic and statistical analysis of the breakdown time delay are presented in this report. Measurement of the current-voltage characteristics with additional monitoring of spatial and temporal distribution of the emission from discharge provides information concerned on development of different regime of low-pressure gas discharge and on processes of the electrical breakdown and discharge maintenance. Also, two new distributions of the statistical time delay of electrical breakdown in nitrogen, the Gaussian and Gauss-exponential ones, are presented. Distributions are theoretically founded on binomial distribution for the occurrence of initiating electrons and described by using analytical and numerical models. Moreover, the correlation coefficient between the statistical and formative time delay of electrical breakdown in nitrogen is determined. Starting from bivariate normal (Gaussian) distribution of two random variables, the analytical distribution of the electrical breakdown time delay is theoretically founded on correlation of the dependent statistical and formative time delay.

1. Introduction
The process of transition from non-conducting to conducting states of gas is known as the electrical breakdown. The research of the gas-phase and surface processes which lead to the gas breakdown has been attracting the attention of scientific community for at least one century. Starting with Paschen measurements of dependences of the static breakdown voltage \( U_s \) on \( pd \) values (pressure × interelectrode gap), it was taken further with Townsend’s theory of the electrical breakdown [1]. This theory was based on the secondary electron emission from the cathode surface and charged particle multiplications in the gas phase. Recently, modifications of Townsend's theory were published in [2] for Ar. These authors developed models of electron, ion, fast atom, excited atoms and photon transport and kinetics and applied them to explain measured data of secondary electron yield in argon for various clean and dirty metal surfaces. The determination of the secondary electron yield from measured \( U_s (pd) \) dependencies (Paschen curves) in nitrogen for the cathode surfaces made of different metals was published in [3].

The model of oscillatory and negative differential resistance behavior of low-current \( H_2 \) discharge between parallel-plane electrodes was developed in [4]. The nonequilibrium model that includes the effects of space-charge distortion of the electric field on the secondary electron emission is used to obtain the negative differential resistance. Spatial distributions of light emission from \( N_2 \) discharge at very wide \( E/N \) range were presented in [5]. Measured distributions of characteristic bands were connected with electron excitation near the anode at lower \( E/N \). Observed emission near the cathode at
high $E/N$ was postulated to be result of excitation by fast molecules. An analysis of time- and space-resolved development of different regimes of low-pressure DC discharge in argon, based on ICCD recordings of the discharge structure, was presented in [6].

Another area of investigation of the electrical breakdown of gases is represented by statistically based measurements of the electrical breakdown time delay $t_s$, the time that elapses from the application of voltage greater then the static breakdown voltage $U_s$ to the breakdown. It comprises the statistical $t_s$ and the formative time delay $t_f$ [1]. $t_s$ is the time which elapses from the application of sufficient voltage to the appearance of a free electron initiating breakdown. $t_f$ is the time interval from this moment to the collapse of applied voltage and occurrence of self-sustained current [1]. The stochastic nature of breakdown time delay was experimentally proven by Zuber [7] and the exponential distribution was theoretically derived by von Laue [8]. Kiselev [9] derived mathematically the exponential distribution of $t_s$ based on binomial distribution of electrons occurrence in interelectrode gap. In recent papers [10,11] the model of the time delay based on the convolution of independent $t_s$ and $t_f$ was presented. Also, the measured distribution of the statistical time delay at high electron yield is non-adequately ascribed to the formative time delay.

The results of the investigation of the electrical breakdown in nitrogen at low pressure based on measuring of the current-voltage ($I\!-\!V$) characteristic and statistical analysis of the breakdown time delay are presented in this report. The influence of the cathode surface conditions on $I\!-\!V$ characteristic is examined through monitoring of axial emission profiles and 2D images of the discharge spatial structure. In statistical part, two new distributions of the statistical time delay are presented [12]. Also, the analytical distribution of the electrical breakdown time delay obtained by the correlation theory of dependent $t_s$ and $t_f$, and the determination of the correlation coefficient in nitrogen are described [13].

2. Experimental details
The measurements presented in this report were carried out on a gas tube made of borosilicate glass (8245, Schott technical glass) equipped with planar copper electrode. The diameter of the electrode was $D=2.2$ cm and the interelectrode gaps $d=0.6$ and 0.8 cm. The tube was filled with research grade nitrogen at a pressure of 2.66 mbar (Matheson Co. with oxygen impurities of below 1 ppm). The static breakdown voltage was $U_s \approx 267$ V at $d=0.6$ cm and $U_s \approx 316$ V at $d=0.8$ cm.

The measurement of current-voltage characteristic at $d=0.8$ cm was made by applying a voltage pulse to a DC voltage that maintains the discharge at very low current ($1-2 \, \mu A$). The voltage and current signals were monitored by digital oscilloscope with appropriate probes. Development of discharge structure is traced down in time using a fast ICCD camera (Andor, iStar DH720) [6].

The time delay measurements were carried out at $d=0.6$ cm by applying rectangular (step) pulses to the discharge tube, at the glow current $I_g = 0.1 \, mA$, glow time $t_g = 1s$ and at different relaxation times $\tau$. The measurements were performed using an electronic system [12] which reached the voltage rise time and resolution limit of 0.18 $\mu s$.

3. Current-Voltage characteristics and spatial development of the dc discharge
The secondary electron emission from the cathode surface is one of the main processes governing the electrical breakdown and dc discharges maintenance. The secondary emission is induced by positive ions, photons and metastable atoms and molecules and it is usually described as an effective yield per ion $\gamma_{eff}$. The aim of this work is to explain the non-standard current-voltage characteristic (with positive slope in low-current regime) that we obtained (figure 1a). As we will show, the cause of this kind of behavior was in the inhomogenous cathode surface that was severely conditioned by the discharge that operated in constricted glow regime for a long period of time. The $I\!-\!V$ characteristic deviations from the usual dependences [6,14] and a positive slope appears in the transition to normal glow instead of negative slope (dashed line). At the same time axial emission profiles (figure 1b), taken at the axes of the discharge, are in agreement with the standard behavior of low-pressure discharges [6,14], starting from low current (Townsend) mode with exponential distribution of intensity (1-3), through formation of the negative glow (4-7) with the development of the cathode fall.
It should be noted that positive slopes in Townsend regime were predicted for secondary yields that are independent of ion energy [15].

Figure 1. a) The current-voltage characteristic of N₂ discharge. b) Axial profiles of emission intensity in numbered points of I-V characteristic (▲ - position of maximum intensity).

2D images of the discharge spatial structure (figure 2) clearly show that at lower currents (labels 1-5), discharge operates only within conditioned area of the cathode – limited circular area positioned close to the center of the cathode. From points 1 to 5, discharge goes from Townsend to glow-like discharge, while remaining within the same limited area. As the current-voltage characteristic gets negative slope, the discharge gradually spreads to the remaining cathode area.

Figure 2. 2D images of discharge spatial structure. The cathode is on the left side.

Further on, the discharge resembles a typical low-pressure glow discharge behavior, except the profile is radially distorted – curved towards the cathode. Practically, due to the conditioning of the cathode, central area has different secondary electron yield than the remaining surface. This leads to the complex behavior of the discharge – it starts in the central area where, presumably, $\gamma_{\text{eff}}$ is higher. The discharge will spread to the area with different $\gamma_{\text{eff}}$ only when current density increases enough for the charged particles, diffused outside the conditioned area, induce sufficient space charge effects to ignite the discharge at the given voltage.

Furthermore, these results are particularly useful for studies of discharges with cathodes of different materials where the same kind of complex behavior may be expected.
4. New distributions of the statistical time delay

In this section the experimentally obtained Gaussian and Gauss-exponential distributions of the statistical time delay will be presented. They are theoretically founded on the binomial distribution for the occurrence of initiating electrons in the interelectrode space [12]. It is shown that transitions from binomial to Gaussian, Gauss-exponential and exponential distribution are caused by the value of the effective electron yield \( YP \) (the electron yield \( Y \) multiplied by the probability of breakdown \( P \)) [12].

The exponential distribution of the statistical time delay was mathematically strictly derived from the probability of occurrence of electrons during the time interval \( t_s \) [9]. The probability of occurrence of an effective electron in the subinterval \( t_s/m \) is \( YPt_s/m \) and for non-occurrence is \( 1 - YPt_s/m \). According to the statistical theory, the probability of breakdown \( W \) in the time interval \( t \) is equal to the sum of probabilities of occurrence of \( j \) electrons in the interelectrode space, where \( j \) takes all the whole number values in the interval \([k-m]\):

\[
W = \sum_{j=k}^{m} C^j_n \left( \frac{YPt_s}{m} \right)^j \left( 1 - \frac{YPt_s}{m} \right)^{m-j},
\]

where \( C^j_n \) are the binomial coefficients. Generally, the sum (1) represents Gauss-exponential distribution of waiting time \( t_s \) for the occurrence of the initial electrons. In the derivation of exponential cumulative distribution [9], it is implicitly assumed that \( YPt_s/m = p \) is close to zero. On the other hand, if \( m \to \infty \) and if neither \( p \) nor \( 1-p \) is too close to zero, Gaussian distribution is obtained as a limiting case of binomial distribution for appearance of electrons or of Gauss-exponential for \( t_s \).

\[ \text{Figure 3.} \] Experimental Gaussian \( f_G(t_s) \), Gauss-exponential \( f_{GE}(t_s) \) and exponential \( f_E(t_s) \) density distributions fitted by numerical differentiation of equation (1) (solid line).

In figure 3, it is demonstrated how the sum (1) of binomial distributions for the electron occurrence goes to Gauss-exponential distribution \( f_{GE}(t_s) \) and approaches two limiting cases of exponential \( f_E(t_s) \) and Gaussian distribution \( f_G(t_s) \) for the statistical time delay. The experimental distributions were obtained on basis of about 1000 measurements in series at the voltage 10% higher then \( U_s \). The statistic time delay values were obtained from measured \( t_d \) values, with \( t_f \) subtracted, at \( \tau = 10ms, 1s \) and 3ms of the afterglow. The methods for determination of \( t_d \) values are described in [16]. In the case of Gauss-exponential distribution all methods give approximately the same results \( t_f = 1.34 ms \), while in the case of exponential and Gaussian distribution only minimum \( t_d \) values can be used, giving for the formative time \( t_f = 1.35 ms \) and \( t_f = 1.27 ms \), respectively.

The experimental density distributions \( f_{GE}(t_s), f_G(t_s) \) and \( f_E(t_s) \) in figure 3 are fitted by numerical differentiation of relation (1). The fitting parameters for Gauss-exponential distribution \( f_{GE}(t_s) \) are as follows: \( k = 3, m = 82, p = 0.008 - 0.16 \) and \( YP = 2.63 \times 10^3 s^{-1} \). In the present case the breakdown
probability is calculated to be approximately equal $P=0.2$ [16], giving the value of electron yield $Y\approx 1.3 \times 10^4 \text{s}^{-1}$. When $p$ is close to zero, the exponential distribution $f_p(t_p)$ at $\tau=1$ is fitted with parameters: $k=1$, $m=36$, $p=0.005-0.115$ and $YP=36 \text{s}^{-1}$. Taking $P=0.2$ into account the electron yield $Y=180 \text{s}^{-1}$ is obtained. On the other hand, when $p$ and $1-p$ are not too close to zero, the Gaussian distribution $f_c(t_c)$ at $t=3\text{ms}$ (figure 3) is fitted with parameters: $k=747$, $m=2587$, $p=0.2535-0.32$ and $YP=1.51 \times 10^7 - 9.6 \times 10^5 \text{s}^{-1}$, which give the electron yield $Y=\bar{Y}+\sigma_y=(6.2 \pm 1.4) \times 10^7 \text{s}^{-1}$. The presented results show that in the early afterglow, with a very high $Y$, the statistical time delay has a Gaussian distribution and this conclusion will be a starting point for the derivation of Gaussian distributions of the electrical breakdown and formative time delay.

5. Theory of correlation of the statistical and formative time delay

In a mathematical sense, the dependence of two continuous random variables $X$ and $Y$ implies that one variable, say $Y$, either increases or decreases as $X$ changes. Two measures of their dependence are the covariance and the correlation coefficient $\rho(X,Y)$. If $X$ and $Y$ are independent continuous variables, then their covariance and correlation coefficient are equal to zero, but the opposite statement is true only in the special case of bivariate normal distribution. It can be shown that the correlation coefficient satisfies the inequality $-1 \leq \rho \leq 1$. Thus, a positive correlation coefficient indicates that $Y$ increases as $X$ increases, a negative $\rho$ implies that $Y$ decreases as $X$ increases and $\rho=0$ implies zero covariance and no correlation. The characteristic case of the joint density function of two continuous random variables is the earlier mentioned bivariate normal distribution, sometimes also called a bivariate Gaussian distribution. If we regard the statistical and formative time delay as continuous random variables $t_s$ and $t_f$, their bivariate Gaussian density distribution $f(t_s,t_f)$ is given by [17]:

$$f(t_s,t_f)=\frac{1}{2\pi\sigma_s\sigma_f\sqrt{1-\rho^2}}\exp\left[\frac{1}{2(1-\rho^2)}\left(\frac{(t_s-\bar{t}_s)^2}{\sigma_s^2} - 2\rho\frac{(t_s-\bar{t}_s)(t_f-\bar{t}_f)}{\sigma_s\sigma_f} + \frac{(t_f-\bar{t}_f)^2}{\sigma_f^2}\right)\right], \tag{2}$$

with the marginal Gaussian distribution of the formative and statistical time, $f_f(t_f)$ and $f_s(t_s)$, which can be obtained by integration of equation (2) for all $t_s$ and $t_f$, respectively. $\bar{t}_s$ and $\bar{t}_f$ denote the mean values of marginal distribution of $t_s$ and $t_f$, $\sigma_s$ and $\sigma_f$ are their standard deviations and $\rho$ is the correlation coefficient of $t_s$ and $t_f$. Taking into account the above-mentioned dependence between $t_s$ and $t_f$, the density distribution of the breakdown time delay, $t_d = t_s + t_f$ is calculated by [17]:

$$f(t_d)=\int_0^\infty f(t_s,t_d-t_s)dt_s, \tag{3}$$

which in the case of the bivariate Gaussian distribution $f(t_s,t_f)$ (equation (2)) gives [17]:

$$f(t_d)=\frac{1}{\sqrt{2\pi(\sigma_d^2+2\rho\sigma_s\sigma_f+\sigma_f^2)}}\exp\left[-\frac{1}{2}\frac{[t_d-(\bar{t}_d+\bar{t}_f)]^2}{\sigma_d^2+2\rho\sigma_s\sigma_f+\sigma_f^2}\right]. \tag{4}$$

Equation (4) implies that the breakdown time delay $t_d$ has a Gaussian density distribution with the mean value $\bar{t}_d=\bar{t}_f+\bar{t}_s$ and the standard deviation $\sigma_d=(\sigma_s^2+2\rho\sigma_s\sigma_f+\sigma_f^2)^{1/2}$, from which the correlation coefficient $\rho$ can be determined.

In order to check earlier hypothesis of the independence of the statistical and formative time delay [10,11] and to demonstrate determination of the correlation coefficient, we have performed new measurements of the breakdown time delay. The dependence of the mean values of the breakdown time delay $\bar{t}_d$ on the relaxation time $\tau$ (the well known 'memory curve' [18]), obtained at the voltage 50% higher then $U_c$, is presented in figure 4. In the first region (I in figure 4), below $\tau=3\text{ms}$, the $t_d$ standard deviation is nearly equal to $t_f$ one ($\sigma_d \approx \sigma_f$), since $\sigma_s$ can be neglected. In the second region (II in figure 4) the contributions of $\sigma_f$ and $\sigma_s$ to $\sigma_d$ are of comparable value, and the
correlation coefficient between $t_s$ and $t_f$ will be determined. In the region III of the memory curve, $\sigma_n$ dominates ($\sigma_{sd} \approx \sigma_n$) and the characteristic distributions of the statistical time delay appear (Gaussian, Gauss-exponential and exponential). The good estimation for the formative time delay in the region III is $t_{f,\text{ms}}$, which represents the minimal value of the time delay obtained in the series of 200 measurements. The $\sigma_n(\tau)=1/Y(\tau)$ ($P \approx 1$ at a given voltage) dependence to about 10ms of the region III is approximately linear in semilog scale confirming that a secondary electron yield in the interelectrode space is determined by the nitrogen ions decaying by diffusion [16]. After that, the dominant emission mechanism of electrons initiating breakdown is the surface recombination of nitrogen atoms remained from the preceding glow [18].

Figure 4. The memory curve, the formative and statistical time delay and their standard deviations as a function of relaxation times.

In order to determine the correlation coefficient between $t_s$ and $t_f$, the $\sigma_{sd}(\tau)$ dependence in the first 7 ms of the memory curve is presented in figure 5. The experimental points below 3 ms refer to the formative time delay and their linear extrapolation to longer $\tau$ gives an estimate of $\sigma_{sd}$ in the regions II and III. Similarly, the extrapolation of the last two experimental points in figure 5 gives the estimation of $\sigma_n$ in the regions I and II. The experimental point at $\tau=9.4\mu s$ with $std(\mu s)=43.0\approx\sigma_{sd}$ was selected to determine $\rho$ (marked with arrow in figure 5). The extrapolated values of $t_s$ and $t_f$ dispersions at this $\tau$ are $\sigma_{sd}=0.25\mu s$ and $\sigma_{sd}=0.27\mu s$. From the dispersion of the density distribution given by equation (4), $\rho$ can be expressed in form:

$$\rho = \frac{\sigma_{st}^2 - (\sigma_{st}^2 + \sigma_{tt}^2)}{2\sigma_{st}\sigma_{tt}},$$

which gives the value of the correlation coefficient $\rho=0.37$; that means that the statistical and formative time delay are medium correlated. The correlation coefficient, determined on the finite sample size ($n=200$), represents the estimation of the population correlation coefficient $\rho$ between $t_s$ and $t_f$. In order to determine the confidence level of $\rho$ we have applied the standard statistical test for the correlation coefficient on large sample which results in the probability greater then 99% that the population correlation coefficient is $\rho>0$. Thus, we can reject the hypothesis that $t_s$ and $t_f$ are uncorrelated, independent variables that is widely exploited in series of papers [10,11].

It is clear from the above analysis that neither the statistical and formative time delay are independent variables nor the density distribution of $t_d$ can be calculated like their convolution [10-11]. This statement will be tested through analysis of the experimental Gaussian density distribution $f_{sd}(t_d)$.
presented in figure 6a, obtained at \( \tau = 4.9 \text{ms} \). It is fitted by theoretical density (equation (4)) with the parameters \( \bar{t}_d \mu = 118.3 \mu s \), \( \rho = 0.37 \), \( \sigma_o = 0.25 \mu s \), \( \sigma_d = 0.27 \mu s \) (the solid line in figure 6a). The agreement between experimental and theoretical distributions is obvious and confirms our statements that \( t_s \) and \( t_f \) are the dependent, correlated variables. Also, the experimental density is fitted by an analytical Gaussian distribution (the dashed line in figure 6a):

\[
f_c(t_d) = \frac{1}{\sqrt{2\pi} \sigma_d} \exp \left( -\frac{(t_d - \bar{t}_d)^2}{2\sigma_d^2} \right),
\]

with the parameters \( \bar{t}_d = 118.295 \mu s \) and \( \sigma_d = 0.431 \mu s \).

**Figure 6.** The Gaussian density distributions of: (a) the breakdown time \( t_d \) (solid line – fit based on equation (4), dashed line – the analytical expression (6)); (b) the formative time \( t_f \) (dashed line – the analytical expression (7)); (c) the statistical time \( t_s \) (dashed line – the analytical expression (8)).

Going to shorter relaxation times \( \tau \), the statistical time delay and its dispersion become negligible compared to the formative time dispersion due to the high level of residual ionization, and the experimental Gaussian density distribution \( f_G(t_f) \) of the formative time at \( \tau = 2.7 \text{ms} \) is obtained (figure 6b). It is fitted by an analytical Gaussian distribution (the dashed line):

\[
f_c(t_f) = \frac{1}{\sqrt{2\pi} \sigma_f} \exp \left( -\frac{(t_f - \bar{t}_f)^2}{2\sigma_f^2} \right),
\]

with the parameters \( \bar{t}_f = 110.5 \mu s \) and \( \sigma_f = 0.30 \mu s \). The \( f_c(t_f) \) density in figure 6b can be fitted by the theoretical distribution (equation (4)) with neglecting terms with \( \sigma_s \) and assumptions \( t_s << t_f \) and \( t_d \approx t_f \). Moreover, the \( t_f \) Gaussian distribution (figure 6b) is obtained for \( Y_{\text{eff}} \approx 10^9 \text{s}^{-1} \), similarly to the \( t_f \) Gaussian distributions in neon [19] obtained for \( Y_{\text{eff}} \approx 10^9 \text{s}^{-1} \) (or \( \sigma_s \approx 1/Y_{\text{eff}} \approx n_s \)). On the other hand, for \( \tau \approx 6 \text{ms} \), the formative time standard deviation can be neglected, i.e. \( \sigma_d \approx \sigma_s \) and the experimental distribution functions describe the statistical time delay [12]. The Gaussian density function \( f_c(t_f) \) obtained at \( \tau = 6 \text{ms} \) from the measured time delay data by subtracting \( t_f \approx t_{d,\text{min}} = 120 \mu s \) is presented in figure 6c. Also, it is fitted by an analytical Gaussian distribution of the form:

\[
f_c(t_s) = \frac{1}{\sqrt{2\pi} \sigma_s} \exp \left( -\frac{(t_s - \bar{t}_s)^2}{2\sigma_s^2} \right),
\]

with the fitting parameters \( \bar{t}_s = 3.8 \mu s \) and \( \sigma_s = 1.25 \mu s \) (the dashed line). From relation \( \bar{t}_s = \kappa \cdot \sigma_s \) [18], \( \kappa \approx 3 \) is obtained, which confirms that the Gaussian distribution in nitrogen (figure 6a) refers to the
statistical time delay. Also, it was shown [12] that the statistical time delay in nitrogen has a Gaussian density distribution for \( Y_{st} \approx 10^8 s^{-1} \) which is now confirmed in the case of the \( t_f \) distribution presented in figure 6c. As in case of \( t_f \) distribution, the \( f_{st}(t_f) \) density in figure 6c can be fitted by the theoretical distribution (equation (4)) with neglecting terms with \( \sigma_y \) and the assumption \( t_f \approx t_f \approx t_{d,\min} \).

6. Conclusion

The observed deviation of \( I-U \) characteristic is related to the changes of the secondary electron yield with the cathode surface conditioning. 2D discharge images clearly show significant change of the discharge structure and development of non-linear phenomena caused by space charge effects. The experimentally obtained new distributions of statistical time delay (Gauss and Gauss-exponential) based on binomial distribution for electron occurrence were presented. The experimental density distribution function of breakdown time delay \( t_d \) was modeled by the theoretical distribution of the sum of two random variables, \( t_s \) and \( t_f \), based on the bivariate Gaussian density distribution of the dependent statistical and formative time delay. The value \( \rho=0.37 \) of the correlation coefficient between \( t_s \) and \( t_f \) was determined from the measured values of the standard deviations. In the limiting cases, the Gaussian density distribution functions of the formative and statistical time delay were experimentally obtained and successfully modeled.

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