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AVALANCHE SPARK BREAKDOWN IN Ne-Br_2

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Abstract: The properties of an avalanche spark in Ne-Br_2 mixtures and parallel plate geometry are discussed. Intense spectral emission of the Br_2 1g ($^3\Pi_{1g}$) → → 2u ($^3\Delta_{2u}$) bands, which persists a few μs during afterglow, is observed. Evidence is obtained also of imprisonment of the NeI resonance radiation. The influence of various processes on the current buildup is considered, and the observed high rate of current growth is attributed primarily to Penning ionization of bromine and to U. V. radiation emitted by Br'_2 molecules. With discharging of the plates the initially negative characteristic changes its sign, and the plates may be discharged to voltages as low as 25 — 30 V; it is suggested that delayed photons are responsible for the observed effect.

During the first microsecond after the onset of the spark breakdown the entire cathode surface is covered by a space charge zone. With prolongation of the pulse the cathode spot contracts both axially and radially, and a normal glow discharge is established within 20 — 50 μs according to the circuit parameters, gas composition, and the initial voltage. The contraction mechanism is briefly discussed.

1. Introduction

It has been established that the breakdown in a uniform field and at values of $p \cdot d < \sim 200$ torr · cm, which sets in after the appearance of an initiatory electron, is due to primary and secondary processes of the Townsend — Rogowsky type[1, 2]. Up to currents of 10^{-7}—10^{-6} A the voltage remains essentially constant and the gap field practically uniform[3]. At higher currents the field becomes distorted due to piling up of positive ions in the vicinity of the cathode, and the rate of current growth becomes accelerated[2, 4]. This stage of breakdown, which, at currents of 10^{-4} — 10^{-3} A, leads to the establishment of a glow discharge is designated as an avalanche spark breakdown[2].

There is little information on that stage of breakdown. The theories proposed[4, 5] are based on rather simplifying assumptions. Experimental data concern mainly the rate of current buildup. They show that the acceleration of the discharge sets in at currents of > 10^{-5} A in argon[6] and at > 10^{-4} A in air[7].

It is generally accepted that the characteristic of an avalanche spark breakdown is negative[2]. This view is supported by measurements of the V-A characteristic of the breakdown in hydrogen in which the gap current has been restricted by a saturated diode[1]. Closely related to the V-A characteristic is the cathode space charge zone. Bandel observed that the discharge in a space charge distorted field is more or less uniformly spread over the electrodes[7]. However, there is very little direct information about changes in space charge structure and current density during the formation of the normal glow discharge.

The present paper is concerned with breakdown phenomena between parallel plates in Ne-Br_2 mixtures. Mainly that stage of breakdown is investigated after which the gap current strength has attained values exceeding ~ 10^{-4} A. It is shown that the dynamic characteristic of a spark breakdown

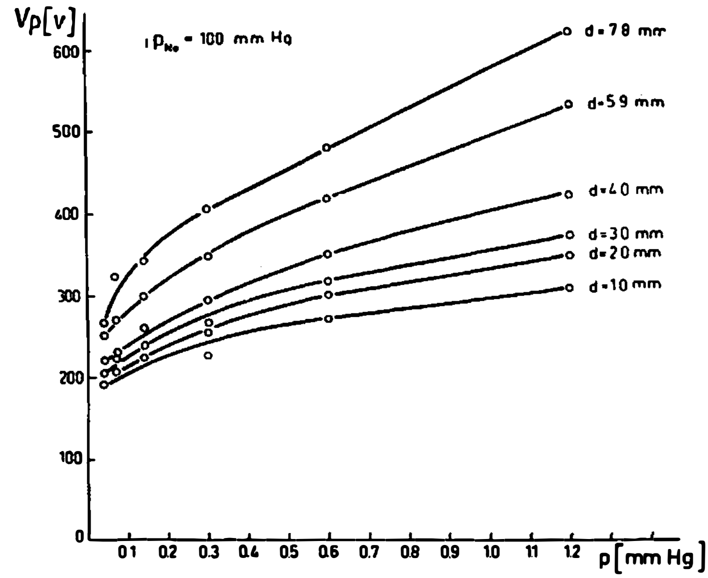


Fig. 1 — Minimum breakdown voltage as a function of the partial pressure of bromine for various gap widths *d*.

is not necessarily only negative, and that the electrodes may be discharged to unexpectedly low voltages, due to the action of delayed photons. Spectroscopic results give evidence of the presence of delayed photons in the spark gap.

The results on current densities at the cathode of the avalanche spark are also reported. They indicate that the axial and radial contraction of the cathode space charge zone during the spark breakdown are essential to the establishment of a normal glow discharge.

2. *Experimental arrangement and results*

Design and operating conditions of the discharge tube. The design of the discharge tube and the filling technique were developed by Srdoč[8] and cowonkers. A cylindrical pyrex tube contained two parallel stainless-steel plates, supported by tungsten wire leads melted in the glass envelope. The interelectrode capacitance in most experiments was of the order of 1 pF and the separation between the plates about 3 mm. The plates were charged up by a stabilized voltage supply via a series resistance R and the breakdown was triggered by a weak source of ionizing radiation.

The discharge tube was filled with spectroscopically pure gases: neon at a pressure of 100 mm Hg and bromine at partial pressures ranging from 0.07 — 2 mm Hg. As shown in Fig. 1 the minimum breakdown potential V_p depended upon the partial pressure of bromine and upon the interelectrode distance d. It is seen that V_p was largely affected by the variation of bromine concentration, while the slope of the curves only slowly increased with increasing d. In fact, at larger values of d the reduced minimum breakdown field strength E_s/p asymptotically approached a certain limiting value (Fig. 2).

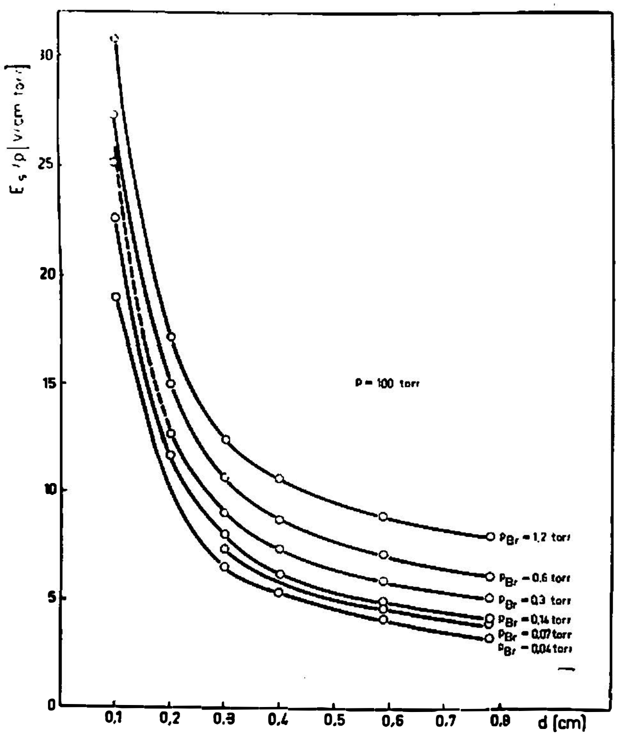


Fig. 2 — Reduced minimum breakdown field strength as a function of the interelectrode distance with the partial pressure of bromine as a parameter.

In order to drastically reduce the probability of streamer formation, further experiments described in this paper were performed using discharge tubes with $d = 3$ mm. It is safe to assume that with $p \cdot d \sim 30$ torr · cm the breakdown develops solely by avalanches.

Another point concerns the influence of the series resistance and of the gas composition on the evolution of the discharge. It has been found that below a critical value R_c, which is characteristic of a given gas composition and geometry (where $R_c \sim 300$ kΩ), the breakdown is followed by the establishment of a stationary discharge. With R slightly exceeding R_c and at low bromine concentrations, $p_{Br} < \sim 0.3$ mm Hg, intermittency of the discharge appears, the frequency and the form of oscillations depending on R, on the concentration of bromine and on the charging voltage.

At sufficiently high concentrations of bromine and with $R > R_c$ the plates are discharged by a single pulse, the discharge becoming unstable shortly after the appearance of a comparatively bright light pulse. After the interruption of the discharge the interelectrode space remains insensitive to ionizing radiation until the space charges formed are dispersed and the plates recharged above V_p.

Further considerations are limited to the single pulse regime, with a series resistance of 100 MΩ and bromine concentrations of > 0.5 %. In order to examine the transition into the glow discharge, an additional external variable capacitor C_1 was connected in parallel with the plates and discharged through a variable resistance R_1. The repetition frequency of the breakdown was maintained low, ~ 1 cycle per second, in order to allow sufficient time for complete recovery of the discharge gap.

Spectral emission. The light emitted from the discharge was resolved by means of a medium quartz and a glass spectrograph. Integrating over a large number of light pulses and using fast emulsions it was possible to record some of the most intense NeI lines: the yellow 5852 Å line and three lines in the red at 6143, 6402 and 6506 Å, respectively. In the ultraviolet a quasi-continuum extending from about 2850 — 3200 Å was observed. The fact that the continuum could be detected even through the glass envelope of the discharge tube indicated that the emission in the ultraviolet was rather intense. Using a discharge tube provided with a quartz window the weakly resolved band structure of excited Br_2 molecules, centred around 2900 Å was identified. These bands originate from 1g ($^3\Pi_{1g}$) → 2u ($^3\Delta_{2u}$) transitions, involving seven bands which overlap[9].

Current buildup and time resolved emission in the visible and U. V. The temporal variation of the NeI 5852 Å line intensity and of the peak at 2900 Å in the Br_2-band spectrum was measured with a 6256 B photomultiplier mounted on a Tektronix 551 osciloscope. The other beam of the scope displayed the variation of voltage with time.

The current through the discharge tube could not be measured directly and therefore was derived from voltage oscillograms according to

$$V(t_1) - V(t_2) = \frac{1}{C} \int_{t_1}^{t_2} [I_g(t) - I_e(t)] \, dt \,, \tag{1}$$

where $V(t_1)$ and $V(t_2)$ are the voltages between the plates at instants t_1 and t_2, respectively. C is the total capacitance of the discharge tube plus the measuring probe. $I_g(t)$ is the current in the gap and $I_e(t)$ is the current strength in the external circuit; being comparatively very small, $I_e(t)$ was neglected.

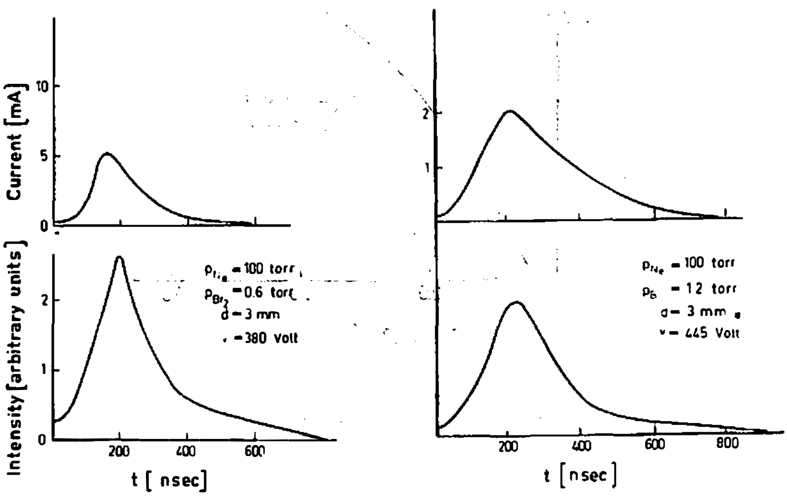


Fig. 3 — The buildup and the decay of the gap current compared with the variation of light intensity at a) 0.6 % Br_2 and 380 V, and b) 1.2 % Br_2 and 445 V.

The upper curves in Figs. 3a and 3b show the change of current with time, as compared with the variation of the NeI 5852 Å line intensity. As seen, the NeI line intensity curves in Figs. 3a and 3b only approximately follow the voltage derivatives. The maximum at 0.6 % bromine lags about 50 ns behind the current peak while at 1.2 % bromine concentration the two peaks almost coincide. In both cases the light intensity persists for ~ 200 ns after the current has dropped to zero.

Furthermore, the comparison of the results in Figs. 3a and 3b shows that the buildup of the spark current is slower at higher bromine concentration, obviously due to the increased probability of attachment. An increase of the initial voltage V_i has the opposite effect; it brings about a certain increase of the rate of current growth.

The 2900 Å bromine band decays much slower than the NeI line. It was detected 1.5 — 2 μs after the NeI line intensity dropped to zero. The Br'_2 afterglow can be due neither to electron impact nor to thermal excitation. For the electron concentration is vanishing already at the instant when the

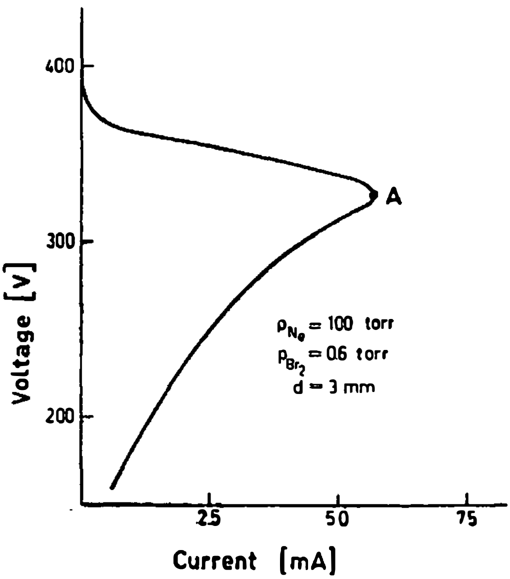


Fig. 4 — The *V-I* characteristic of a spark breakdown in a Ne-Br_2 mixture containing 0.6 % bromine; V_i = 380 V.

gap current approaches zero. On the other hand, the gas temperature certainly does not attain values needed for excitation of the 1g ($^3\Pi_{1g}$) level of the bromine molecule at actual current densities and short pulse durations. The observed high intensity ob Br'_2-bands and their formation during afterglow is tentatively attributed to recombination processes.

V-I characteristic. A *V-I* characteristic of the avalanche spark in a Ne-Br_2 mixture containing 0.6 % Br is shown in Fig. 4. It was obtained from a V oscillogram and from the derived value of $I_g(t)$, Eq. (1). The characteristic changes its sign at $V \sim V_p$. It is remarkable that the plates are discharged to a rather low residual voltage V_f. Fig. 5 shows plots of V_i/V_f for gas mixtures containing 0.6 and 1.2 % Br, the individual points in the V_i/V_f plots representing the average of 10 to 15 measurements. The dotted lines denote the minimum breakdown voltage V_f, which differs only little from the voltage at which the instability of a normal glow discharge at the corresponding pressure and gas composition sets in. As discussed later, these results clearly indicate that the delayed photons present in the gap may bring about the prolongation of the discharge even if the condition for the breakdown is no longer fulfilled.

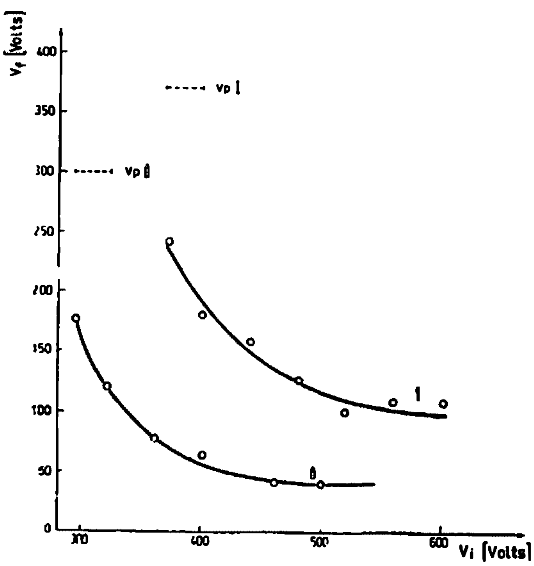


Fig. 5 — Plots of V_f versus V_i. The lower curve for p_{Br} = 0.6 torr, the upper curve for p_{Br} = 1.2 torr.

Space charge formation. It has been observed that an avalanche spark, lasting < 1 μs, completely fills the interelectrode space, and unlike in a stationary glow discharge, at the same gas composition, pressure and mean current strength the luminous zones completely cover the plates. At lower bromine concentrations and higher voltages, the luminous zone in front of the cathode has a slightly concave shape (Fig. 6).

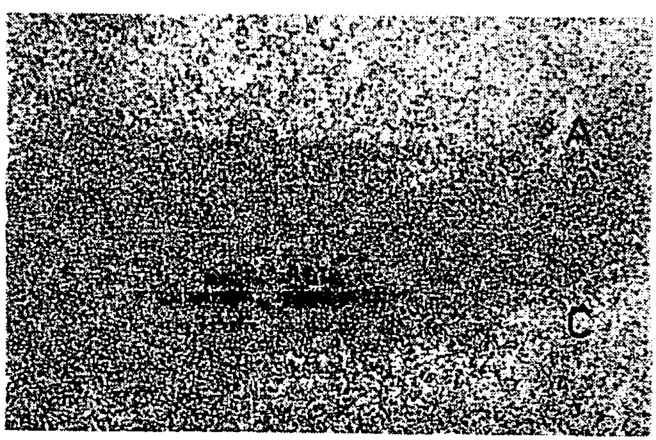


Fig. 6 — Photograph of the discharge V_i = 440 V, p_{Br} = 0.6 torr, (magnification 6×).

Quantitative information on the axial intensity distribution of light emitted from the discharge is obtained from stigmatic photographs in the visible. Plots of photographic densities versus interelectrode distance are shown in Fig. 7a (0.6 % bromine) and Fig. 7b (1.2 % bromine). As seen, the maximum in front of the cathode falls off much steeper towards the anode at higher bromine concentrations (Fig. 7b), clearly indicating the quenching influence of bromine. The small peaks between the anode and cathode space charge zone are due only to the graininess of the emulsion.

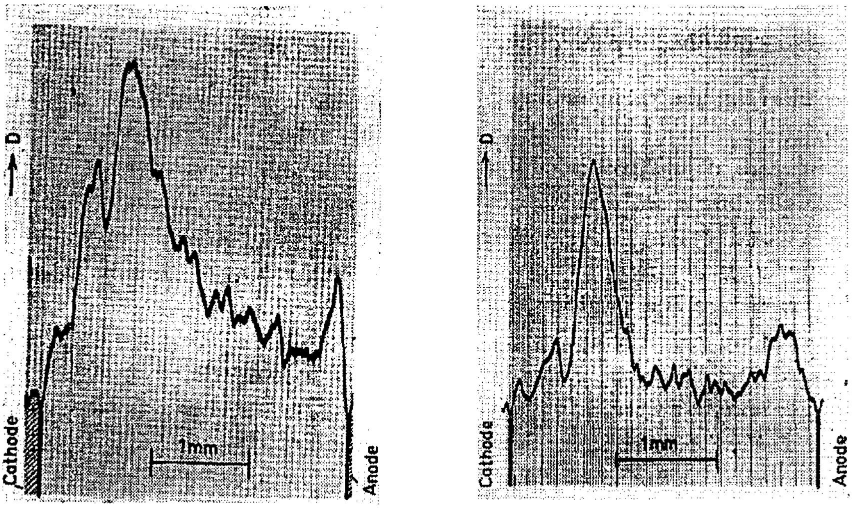
The luminous maxima in both cases are at a distance of 0.8 mm from the cathode. This value is by about one order of magnitude larger than the distance of the negative glow from the cathode in a stationary glow discharge under the same conditions. Prolonging, however, the current pulse one observes that the cathode spot, probably gradually, contracts.

Because of the low luminosity of the event it was found difficult to obtain time resolved photographs, and the contraction rate of the cathode spot is thus not yet well known. Very approximate estimates based on visual observation show that by adjusting the time constant of the external circuit to values ranging between 20 and 50 μs the contraction is ended by that time, provided the discharge voltage is maintained during the entire interval of time at/or slightly above the minimum breakdown voltage V_p. The contraction rate is increased by raising the discharge voltage and/or by lowering the bromine concentration. The contraction of the cathode spot is accompanied by a contraction of other regions of the discharge.

3. Discussion

Classification of the discharge. The experimental data give evidence that during the investigated stage of breakdown the current strength and consequently the space charge density considerably exceed the corresponding values, characteristic of a stationary self-sustained Townsend discharge. On the other hand, the current density in the cathode space charge zone is initially much lower than that in a normal glow discharge; it increases with the duration of the breakdown, thus indicating that the stability conditions in the space charge zone are not fulfilled. Because of these properties the observed transient stage in parallel plate halogen counters may be classified as a spark breakdown, and since filamentary structures are fully absent it represents an avalanche spark.

The rate of current growth and the processes in the discharge. It is found that as long as the voltage between the plates is sufficiently high the rate of current growth is very fast. Afterwards, with discharging of the plates there is a range, preceding point *A* (Fig. 4), of approximately linear current growth. In this range equation[10] $V_p - V = ki$ applies.



Figs. 7a and 7b — Plots of plotographic densities versus interelectrode distance at p_{Br} = 0.6 torr and 1.2 torr, respectively.

A direct comparison of the present results with those obtained for pure neon[11] is not possible because of the different modes of triggering the breakdown. Nevertheless, oscilloscopic measurements allow to conclude that the initial rate of current growth in Ne-Br_2 mixtures is faster than in pure gases[11] or in air[7]. This is attributed to very efficient ionization processes and to the observed intense excitation of Br'_2 bands.

Bromine in halogen counters is readily ionized by second order collisions with neon metastables[12]. From the point of view of the adiabatic criterion[13] the process

$$Br_2 + Ne^* \rightarrow Br^+ + Br + Ne + e \tag{2}$$

appears to be more likely than

$$Br_2 + Ne^* \rightarrow Br_2^+ + Ne + e \ . \tag{3}$$

However, both processes are estimated to be very fast since the internal energy change is transferred to the electron as kinetic energy[13]. With the cross section for the resonant energy transfer $Q \sim 10^{-4}$ cm^2, the mean lifetime of metastables,

$$\tau = [Q \cdot N_{Br} \cdot V_{rel}]^{-1}, \tag{4}$$

is of the order of $2 \cdot 10^{-7}$. The same considerations are expected to apply to the excited molecular species Ne'_2 formed by three body collisions[11] and also to the excited levels of neon atom whose effective lifetime would be otherwise considerably prolonged by imprisonment of resonance radiation.

The electron emission at the cathode. The secondary emission of electrons due to the action of metastables is probably negligible, not only because of the low rate of diffusion compared with the short duration of the spark breakdown, but also because of the processes in Eqs. (2) and (3).

The secondary emission coefficient γ_i for bromine ions is expected to be small or even negligible[15], and probably neon ions are mainly responsible for the γ_i part of the total secondary emission coefficient ω.

The present results show that Ne^+ is predominantly formed at a distance of 0.4 — 1 mm from the cathode. Taking into account that the average mobility of Ne ions[16] as well as that of bromine ions[17] in neon is $\sim$ 6.5 cm^2/Vs, the initial, i.e. the maximum drift velocity of ions is only $\sim 7 \cdot 10^3$ cm s^{-1}. Even assuming that the drift velocity of ions is little affected by the decrease of the field strength during the discharge, the time necessary for the drift of the bulk of ions to the cathode lasts at least an order of magnitude longer than the entire interval of current growth. Thus it appears safe to conclude that secondary electron emission during the first microsecond of the spark breakdown is mainly due to the photoeffect.

The threshold of the photoeffect for the discharge tubes used was found to be $\sim$ 4.3 eV. Consequently, the secondary emission of electrons is due not only to resonance radiation of Ne' and nonresonance vacuum U. V. radiation of excited Ne'_2 molecules but also to photons emitted by the spontaneous decay of excited Br'_2 molecules. The NeI resonance and the nonresonance vacuum U. V. radiation in pure neon[11] do not bring about so high rates of current growth as observed in $Ne\text{-}Br_2$ mixtures. On the other hand the Br_2 1g $(^3\Pi_{1g}) \rightarrow$ 2u $(^3\Delta_{2u})$ bands are not reabsorbed and very intense. This suggests that the large temporal growth constant in $Ne\text{-}Br_2$ is mainly due to the U. V. radiation of excited bromine molecules.

Delayed photons and their effects. The evidence of imprisoned NeI resonance radiation is somewhat less direct than that of the comparatively slowly decaying Br'_2 bands. The NeI 5852 Å line is due to the transition 3p [1/2] $\rightarrow$ $\rightarrow$ 3s' [1/2]°. The upper, 3p' [1/2]° level, may be populated in the avalanche spark by three processes:

1) direct electron impact excitation of Ne atoms in the ground state;
2) collisions of Ne metastables with electrons; and
3) cascading from higher lying levels.

Electron-ion recombination processes during afterglow appear to be of little importance since the electrons disappear quickly, swept away by the residual field. The first two processes last only during the passage of current and consequently, it is the third process responsible for the NeI emission after cessation of the current flow.

As may be concluded from the relative intensities of the NeI lines[18], cascading involves mainly the 3d, 4d and 5d resonance levels, and particularly the 3d $[1\,{}^1/_2]^\circ$ level. All these levels are imprisoned, and according to Holstein[19]-Biberman's[20] theory, the imprisonment of resonance radiation, under the actual conditions and with the estimated value of transition probability of $\sim 10^8$, should last about 5 μs. The observed much faster decay of the NeI line (Figs. 3a and 3b) is therefore attributed to the depopulation of resonance levels by the Penning process, as discussed above. The apparently small difference in the rate of quenching of excited NeI levels at 0.6 and 1.2 % Br is very likely due to the inaccuracy in the evaluation of the current strength according to Eq. (1), and to the error in estimates concerning the slowly decaying tails.

The prolongation of the growth of NeI line intensity (Fig. 3a) after V has dropped below V_p and the fact that the plates are discharged to very low voltages may originate only from the effect of delayed photons. Photoelectrons liberated by delayed photons from the cathode are accelerated in the residual field maintaining the process of multiplication even when the criterion for the avalanche growth in electronegative gases[2]

$$\frac{\omega \cdot \alpha}{\alpha - a} [\exp (\alpha - a) \cdot d] > 1 \tag{5}$$

is no longer fulfilled.

As seen from the above equation, the ionization coefficient must exceed the coefficient of attachment, a, during the period of current growth. In the investigated range of E/p the ionization coefficient in neon increases[21] and the attachment coefficient in bromine decreases with increasing E[21, 22]. Consequently, with the discharging of the plates, i.e. with lowering of the mean value of E/p, the difference $(\alpha - a)$ will decrease and belov E_s the condition given by Eq. (6) is no longer fulfilled; in the absence of delayed U. V. photons the discharge below V_p would become immediately unstable.

According to the above discussion and in agreement with the fact that an increase of V_i and of $C\,(V_i - V_p)$ will increase the yield of excited Br'_2 and NeI species and thus also the density of delayed photons, it is expected that an increase of V_i will bring about the attainment of lower values of V_f; indeed this is observed in Fig. 5. Unfortunately, the complexity of the ion density distribution, the rather large number of interactions involved and the lack of relevant data make it premature to give a quantitative interpretation of the rates of current change and of the V-I characteristic.

Establishment of the glow discharge regime. Due to the low mobility of ions the growth of avalanches brings about a space charge distortion of the gap field; initially the highest ion density is attained in the vicinity of the anode. As a result of the increased local field strength the excitation and ionization rates between the cathode and the positive space charge zone are accelerated, thus further increasing the space charge density and shifting its maximum towards the cathode. In that way the development of the spark breakdown and the establishment of the normal cathode fall are closely related to the propagation of an ionization wave towards the cathode.

Comparison of light intensity oscillograms with the integral light emission (Figs. 7a and 7b) indicates that the velocity of ionization waves is rather large, of the order of 10^6 cm/s. Incidentally, this value is close to that found for *r* waves, observed in the positive column of a low current discharge in neon[23]

As seen in Fig. 6. the axial contraction is almost terminated while the space charge still covers the entire cathode surface; there is perceptible enhanced emission from the central part of the cathode, but it takes further 10 and more μs until the radial contraction is ended. With prolongation of the discharge the peripheral parts of the space charge become bent upwards, decreasing in brightness, until the discharge concentrates on a small contracted cathode spot.

The radial contraction might be due to the higher density of photons near the axis of symmetry, and thus to a more copious secondary emission of electrons from the central region of the cathode, which brings about further local intensification of current growth.

Very little is known about the mechanism of cathode spot contraction. It is hoped that the work in course will yield more information on the phenomena in the cathode space charge and thus contribute to a better understanding of the correlation between contraction and the stability conditions in a normal glow discharge.

Acknowledgement

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ISPITIVANJE PROBOJA LAVINSKE ISKRE U Ne-Br_2

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Sadržaj

Opisana su svojstva Townsendove iskre u smjesi Ne-Br_2 među paralelnim pločastim elektrodama. Utvrđena je intenzivna spektralna emisija Br_2 1g ($^3\Pi_{1g}$) $\rightarrow$ 2u ($^3\Delta_{2u}$) vrpci, koja traje nekoliko μs i nakon prestanka izbijanja. Na osnovu spektralne emisije nekih tripletnih linija NeI zaključuje se da je rezonantno zračenje atoma neona kratkotrajno zarobljeno.

Razmatra se utjecaj raznih procesa na porast jakosti struje i relativno velika brzina porasta tumači prvenstveno Penningovom ionizacijom broma, te sekundarnom emisijom elektrona uslijed U. V. zračenja uzbuđenih Br_2-molekula.

Izbijanjem ploča negativna *V-I* karakteristika mijenja predznak, a ploče se izbijaju na vrlo niske napone od 25 — 30 V. Pokazuje se da pojava može nastati uslijed djelovanja tzv. zakašnjelih fotona.

Za vrijeme prve mikrosekunde nakon početka razvoja iskre čitava katodna površina je prekrivena prostornim nabojem. S produženjem trajanja impulsa katodni prostorni naboj kontrahira aksijalno i radijalno, te se normalno tinjavo izbijanje uspostavlja nakon 20 — 50 μs, već prema parametrima kruga, sastavu plina i početnom naponu. Ukratko se diskutira mehanizam kontrakcije.