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Invited Papers
DISCHARGES AT EXTREMELY HIGH VALUES OF E/n AND LOW CURRENTS

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Introduction
Our interest in electrical discharges and breakdown at very high ratios of the electric field to gas density E/n in spatially uniform electric fields originally arose from the opportunity the investigation of such phenomena offers for the development and testing of models of the motion of electrons at sufficiently large E/n that departures from the local equilibrium or hydrodynamic models are important. When the interpretation of our experiments indicated that collisions of fast ions and fast neutrals with the gas are important, we were faced with the challenge of developing and testing quantitative experiments and models characterizing the behavior of these species. The understanding of the motion of electrons, ions, and their products at high E/n in a spatially uniform electric field is fundamental to the understanding and modeling of their behavior in the nonuniform and time varying electric fields of the cathode fall, low pressure rf discharges, Tokamak type device startup, and the high field regions of semiconductors.

Electrical discharges and breakdown at very high E/n have been investigated since the appropriate vacuum systems became available [1,2]. Interest in the topic relative to breakdown and discharges at the higher pressures later declined as evidenced by the short discussions in more recent texts [3]. This paper begins with a review of representative published experiments and models [3-30]. A summary is then given of some recent work carried out in collaboration with B.M. Jelenković [31,32], L.C. Pitchford [32], D.A. Scott [33], and V.T. Gilya [34]. The E/n of these experiments cover the range from E/n for which there is a local equilibrium between the electron energy gain from the electric field and the loss in collisions to extremely high values E/n for which the electron motion approaches free fall under the force of the electric field.

Breakdown and discharge I-V characteristics
The early experimental determinations of electrical breakdown at low gas pressures showed the large increase in breakdown voltage with decreasing gas pressure resulting in what is known as the left-hand side of the Paschen curve [1,2]. This behavior is illustrated in Fig. 1 by our measurements in Ar, D2, and N2 of breakdown voltage Vb vs the product of the gas density n and the electrode separation d [31]. Penning and Addink [4] have shown that the simple minima found for these gases becomes a multiple minima curve for Penning mixtures and becomes a multiple valued curve of Vb vs nd for He. The validity of the Paschen law scaling, according to which Vb is a unique function of nd, was found to be a function of electrode design in the experiments of Pokrovskii-Sobolev and Klyverfeld [5] and of McClure [6] in N2 and D2. Surveys of the breakdown voltages for various gases at low nd values were made by Guseva [7] and by Schönhuber [8]. The latter also made a

![Figure 1. Breakdown voltage Vb vs product of gas density and electrode separation for Ar, N2, and D2 [31]. The electrode separation was 39 mm for Ar and N2 and either 20 mm (a) or 39 mm (g) for D2. The electrode material was sintered graphite.](image-url)
careful study of electrode design. The effects of reflection of electrons at the anode on the breakdown potential were investigated by Dikidzhi and Klyarfel'd [9], McClure [6] and Bhattacharjee and Parker [10].

The voltage-current characteristics of low pressure, low current discharges in parallel plane electrode geometry have been investigated primarily by Klyarfel'd and coworkers [11,12]. As illustrated in Fig. 2 by our measurements [31] in H₂ and D₂, they found that the discharge voltage is independent of discharge current to within experimental uncertainties for total current densities of less than about 3 x 10⁻⁴ A/m². This behavior is consistent with calculations which show that for H₂⁺ ions in H₂, space charge distortion of the electric field can be neglected for total discharge current densities jₓ < 10⁻³ A/m² at E/n > 300 Td, where 1 Td = 10⁻²¹ Vm². For H₂⁺ ions in H₂ and Ar⁺ in Ar the calculated current density limits are 10⁻⁴ A/m² at E/n > 300 Td. The absence of variation of voltage with current is also consistent with calculations which show that multistep excitation and ionization and gas heating effects should be negligible for these jₓ/n and nd values. Because of these simple discharge conditions, the models of electron and ion motion can be kept simple and are expected to yield definitive predictions for comparison with experiment.

Operation of these low pressure discharges at significantly higher current densities has been investigated by Klyarfel'd and others [11-14]. A discussion of these results is beyond the scope of this paper.

![Figure 2](image_url)

**Figure 2.** Discharge voltage Vₐ vs total current jₓ for discharges in D₂ and H₂ at the pressures indicated. The electrode diameter was 78 mm and their separations were as indicated [31].

**Discharge and breakdown models**

Quantitative theoretical models of the behavior of electrons and ions at very high E/n are rather limited. Approximate solutions of the electron Boltzmann equation for sufficiently high E/n that electron energy losses can be neglected were obtained by Stuart and Gerjuoy [15]. Energy losses were added by Gurvich [16], while momentum losses and anisotropic scattering were considered by Müller and coworkers [17]. More recently, the limit of small electron energy loss has been considered by Briggs and Yu [18], Friedland and Kagan [19], and Lagushanko and Maya [20]. The velocity moment method used by Müller and coworkers has been applied more recently by Ingold [21] and by Friedland and Kagan [19]. Iterative solutions for the full nonequilibrium Boltzmann equation have been carried out by Pitchford et al. [22], but only for isotropically scattered electrons. Monte Carlo techniques have been used for simulation of the electron motion during breakdown at very high E/n by Grasnov and McClure [23], by Parker and coworkers [24], and by Lauer, Yu and Cox [25]. Folkhard and Haydon [26], Hayashi [27], Tagashira and coworkers [28], and Ul'yanov and Chulkov [29] applied the Monte Carlo technique to the lower E/n range of interest here.

Reisman [30] has considered the transition in the electron motion from equilibrium to runaway. The models of Grasnov and McClure [23], of Parker and coworkers [24] and of Lauer, Yu and Cox [25] considered the contributions to breakdown of backscattered electrons from the anode and of secondary electrons released from the cathode by ion bombardment.

Among the more useful theoretical results for our purposes are the calculations of the spatial dependence of the apparent ionization coefficients for H₂ by Müller [17]. Figure 3 shows the results of Müller for E/n of 2.26 and 28.2 kTd. One reason for the utility of these calculations is that the cross sections for the dissociative excitation of H₂ [35] have very nearly the same energy dependence as the ionization cross section. This means that Müller's predictions for ionization of H₂ can be scaled to predict the magnitude and spatial dependence of the Blaser series emission from high E/n discharges in H₂ and D₂.
Figure 3. Calculated apparent ionization coefficients for electrons in H₂. The curves are from Müller [17], while the points are from our single beam model of electron motion [32].

A second source of detailed calculations of the behavior of electrons at very high E/n is the Monte Carlo models of Parker et al [10, 24]. The points of Fig. 4 show the values of the current multiplication factor $M(x)$ calculated by Pace and Parker [24] and by Bhasavanich and Parker [10], while the curves show values calculated using various approximations to the solution of the electron Boltzmann equation [32].

Figure 4. Calculated and measured electron current multiplication $M(x)$ vs available electron energy for Ar. The solid points are from Monte Carlo calculations by Parker et al. [10, 24], while the open points show the results of modification to remove backscattering effects [31, 32]. The curves are the predictions of various electron beam models [32]. The uppermost dashed line is from experiment [36].

Discharge diagnostics

Diagnostics available for use with these high E/n discharges include the measurement of the spatial and temporal variation of the light emission, the spectral distribution of the emission, the energy and charge-to-mass distributions of the ions arriving at the cathode, and the energy distribution of the electrons arriving at the anode. Although these techniques have been applied to discharges at lower E/n and/or at higher current densities [14], the only published diagnostic studies we have found for the low current, very high E/n discharges are the photographs of Ne discharges [12, 13]. Techniques which may possibly be applicable include laser induced fluorescence and laser absorption.

In this paper we will present representative results obtained using steady-state [31] and transient [33, 34] measurements of the light emission from high E/n, low j₀/n discharges in Ar and H₂ at low and. Most of the data discussed are for Ar since it has proven easier to interpret. The data for D₂ have been only partially analyzed at this time. Examples of measurements by Jelenković and Phelps [31] of the steady-state spatial distribution of emission at 813.5 nm from discharges in Ar are shown in Fig. 5 for 272 Td and 42,600 Td, respectively. These data were obtained by scanning a photomultiplier and optical slit system past the discharge. The relative intensity measurements are made absolute by normalization to published measurements at overlapping lower E/n. The results are expressed in terms of an apparent excitation coefficient, i.e., the local value of the probability of production of an excited state per unit distance in the field direction times the local electron current density normalized to the total current density.

Steady-state measurements in Ar

Following previous work on the spatial distribution of emission [37, 38], the spatial distribution of intensity shown in Fig. 5 is interpreted as evidence for an exponential growth of electron density according to the conventional Townsend avalanche model [1, 2]. The ionization coefficient derived from the slope of the straight line through the points agrees well
Figure 5. Apparent excitation coefficients for 811.5 nm emission from Ar discharges. The triangular points are for 272 kTd and an Ar density of 2.7 x 10<sup>21</sup> m<sup>-3</sup>, while the solid circles are for 42.6 kTd and 1.64 x 10<sup>21</sup> m<sup>-3</sup>. The line through the triangles indicates an exponential growth of ionization.

with previous results of measurements of current growth vs electrode separation [36]. This result is consistent with our expectation that the electron energy distribution is in equilibrium with the applied E/n and that previous measurements and calculations of electron excitation rate coefficients are applicable to our experiment. We therefore normalize the measured intensities to the emission coefficient data of Tachibana [39]. This normalization is done at the anode, since it is expected that the ion current at the anode is zero and the electron current is equal to the total or measured current.

Figure 6 shows a comparison of the experimental apparent excitation coefficient data for E/n = 42.7 kTd from Fig. 5 with the predictions of models of the excitation of the 811.5 nm line by electrons and by fast neutral atoms. The dashed line was calculated using the measured electron excitation cross section [40] and the energy-balance, single-beam model [32]. The maximum in the apparent excitation coefficient occurs when the electron energy is at the peak of the excitation cross section. The flatter portion at greater distances from the cathode is an approximate calculation [32] of the excitation due to low energy electrons produced by ionization. Cascading effects and the leakage of 810.4 nm emission through the interference filter are not included in this curve and could increase the electron excitation by as much as a factor of five at distances greater than a few mm from the cathode. The discrepancy between these predictions and experiment is at least an order of magnitude over most of the gap.

A much closer fit to experiment is obtained by considering the excitation due to fast neutrals [31]. This process has been observed by Kempter, Veith, and Zehnle [41] at 900 eV. We have assumed that the energy dependence for excitation of the 811.5 nm line is the same as that observed [41] for the weaker 750 nm line. The fast Ar atoms are produced in charge transfer collisions between Ar<sup>+</sup> and the thermal Ar gas atoms. For the E/n of Fig. 6 the calculated [42,43] one dimensional "temperature" of the Ar<sup>+</sup> and, therefore, of the fast Ar is 106 eV. According to this model of ion and fast atom behavior in the discharge [31], there are about six fast Ar atoms per ion at positions near the cathode. The agreement between the measured and calculated spatial dependence is noticeably less satisfactory near the cathode when ionization of Ar by fast Ar is omitted from the model. The cross section for excitation of the 811.5 nm line by fast Ar<sup>+</sup> is unknown, but estimates based on the corresponding 614.3 nm line for Ne [44] suggest that it is about an order of magnitude smaller than for excitation by fast Ar of the same energy. The discrepancy near the anode is at least in part due to the omission from the model of excitation by backscattered electrons.
Transient measurements in Ar

In order to aid in the separation of the contributions of electrons, fast ions, and fast neutrals to the observed excitation in Ar, Scott and Phelps [33] have made a series of measurements of the time dependent emission from prebreakdown discharges. In these experiments the graphite cathode was replaced by a semitransparent photocathode [45], which was illuminated with the output of a quadrupled YAG laser at 266 nm to produce a peak current of about 10 mA with < 8 ns width (FWHM). A grid was added at a plane 10 mm in front of the cathode and, in the experiments to be described, was maintained at the original equipotential.

The gas density was adjusted to be low enough such that the charge in the second avalanche was significantly less than that of the initial avalanche. At E/n less than 1000 Td the resultant cathode current and spatially dependent 811.5 nm emission waveforms were consistent with the published waveforms observed [46] for lower E/n when the dominant secondary emission process was photoemission from the cathode. The observations at very high E/n are represented by the waveforms for E/n = 18.6 kTd shown in Fig. 7. The upper trace is the current measured in the cathode to grid circuit and to a first approximation represents the rate of arrival of charge at the region near the cathode. The middle and lowest traces are the signals from the photomultiplier when placed so as to observe 811.5 nm emission from a narrow region midway between the grid and the cathode and from a point 5 mm from the anode, respectively.

Points to be emphasized in connection with the current trace are: the virtual absence of an electron pulse due to the limited time resolution of the digitizer and the presence of oscillations in the true waveform resulting from lead inductance; the rising ion current resulting from the initial, spatially increasing ionization produced by the electron avalanche; a decrease in the ion current at the ion transit time calculated from the ion mobility [42] using the measured charge transfer cross section [43]; and the small magnitude of the charge in the second avalanche indicating a small number of secondary electrons relative to the number of photoelectrons. The 811.5 nm emission waveform from near the cathode is consistent with the following: a small excitation peak

![Figure 7. Prebreakdown current and 811.5 emission from Ar for E/n = 18.6 kTd. The electrode separation was 39 mm and the gas density was 1.6 x 10^24 m^-3. The upper trace is the cathode current, while the lower traces are the 811.5 emission at positions 5 mm from the cathode and anode, respectively. The Ar^- transit time is from the anode to the middle of the grid-cathode gap.](image-url)
caused by electrons in the first avalanche; a delay in the rise of the emission such as would be caused by the time required to build up the fast atom flux; the emission from the point 5 mm from the cathode peaks earlier in time relative to that of the maximum in the arrival of ions at that point; and the area under the delayed emission peak is much larger than that under the initial electron avalanche peak. The latter point is evidence for excitation by ions or fast atoms. An incomplete analysis of the details of the waveform indicates that the early peak in the emission is the result of excitation by fast atoms rather than ions. The 811.5 nm emission waveform from near the anode shows: an initial peak expected for significant excitation by primary or backscattered electrons; a rapid decrease in the delayed emission as expected when the ions or fast atoms move toward the cathode; and a small delayed component to the emission suggestive of excitation by electrons, ions, or atoms from the second avalanche.

Observations of the 750.3 nm emission waveform at midgap show a much larger prompt electron peak relative to the delayed fast atom peak than does the 811.5 nm waveform. This change results from the fact that the cross section for excitation of the 750.3 nm line by electrons has a typical allowed character in that it decreases slowly with energy at above 100 eV [40] so that the probability of excitation by high energy electrons is much higher than for the 811.5 nm line. In addition, the cross section for excitation of the 750.3 nm line by fast Ar is about a factor of 6 less than that for the 811.5 nm line [41].

From these waveforms and from the steady-state data it is concluded that while at the lower E/n the excitation of Ar can be explained by electron impact, at the very high E/n the excitation of the 811.5 nm line is dominated by collisions with fast Ar atoms formed in charge transfer collisions between Ar⁺ and thermal Ar.

**Results for N₂**

The results presented for Ar are typical of those obtained for N₂ and D₂ in that at the lower E/n the data are consistent with the conventional electron avalanche models [1], while at the highest E/n the emission observed for at least some of the bands and lines cannot be accounted for by electron excitation. In particular, for N₂ Jelaznović and Phelps [31] propose that at the higher E/n the excitation of the 670 nm band and 337.1 nm band of the 1st and 2nd positive systems is primarily by fast N₂, while the excitation of the 391.4 nm band of the 1st negative system is by electrons. Some of the N₂ results are shown in Fig. 8, where the solid curves show the measured apparent excitation coefficients for the C³Πu state and for the 391.4 nm band. The lower chain curve shows that the calculated contribution of electron excitation to the C³Πu signal is much too small to explain the experimental result. It is proposed [31] that the excitation of the C³Πu is caused by collisions of fast N₂ with thermal N₂, where the fast N₂ is produced in charge transfer collisions of N₂⁺ with ground state molecules.

Unfortunately, only relative excitation cross section data for this process are available [47]. The short dashed curve shows that one can obtain good agreement with the spatial dependence of the 1st negative band emission at 391.4 nm using simple electron beam models and published excitation cross sections [48]. The absolute magnitude of the experimental curve is somewhat uncertain because of the weak 391.4 nm signal at the lower E/n [31]. Observations of current and emission waveforms for pulsed prebreakdown discharges in N₂ are being made by Gylis and Phelps [34]. They find that at all E/n the 391.4 nm emission waveforms can be entirely accounted for by excitation by primary and

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**Figure 8.** Apparent excitation coefficients vs distance from the cathode for the C³Πu state and 391.4 nm band of N₂. The lower chain curve and the dashed curve show the respective calculated excitation coefficients, while the solid curves show the coefficients derived from experiment.
secondary avalanche electrons. The emission from the C$^3$N$_{2}$ and B$^3$I$_{2}^+$ states, however, correlates with the electron transit time only at the lower $E/n$. At the higher $E/n$ most of the emission from these bands is delayed as expected for excitation by either fast ions or neutrals and is therefore consistent with the proposed excitation by fast $N_2$ [31].

Summary

The processes occurring in the gas phase of steady-state electrical discharges at high electric fields and low gas densities are beginning to be understood quantitatively through the application of optical diagnostics, such as the measurement of absolute emission intensities. Observations of prebreakdown transients under single avalanche conditions have demonstrated the importance of collisions involving fast atomic species. Present indications are that the fast atoms and molecules are more important sources of excitation and ionization than are fast ions, although further tests of this hypothesis are desirable. The transient experiments define the conditions under which the emission from various spectral lines and bands are satisfactory diagnostics of the behavior of the electrons and of the fast neutrals. In particular, the 391.4 nm band of $N_2$ and, to a lesser extent, the 750.3 nm line of Ar are excited by electrons under both moderate and very high $E/n$ conditions. On the other hand the 1st and 2nd positive bands of $N_2$ and the 811.5 nm line of Ar are excited efficiently by fast neutrals at very high $E/n$.

Future work with very high $E/n$ discharges should include: direct measurements of electron current gain for comparison with models; transient emission and current waveform measurements in other gases such as $D_2$; extension of the limited current growth measurements under breakdown conditions [31]; and quantitative evaluation of the contribution of electrons released from the electrode surfaces to breakdown and steady-state discharges. It would be particularly desirable to extend the pioneering measurements of ionization coefficients for ions of Townsend and Llewellyn-Jones [49] to other gases and to higher $E/n$.

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References


