9.3 Ionization in Gas Discharges: Experiment and Modeling

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9.3.1 Introduction

Electron impact ionization is the dominant source of electrons and ions in many electrical discharges in gases. This includes the ionization of atoms and molecules in excited states such as metastable states, as well as ionization of the ground state. Other processes resulting in the production of electrons in gas discharges, which we will not consider, are associative and Penning ionization, chemionization, photoionization, collisional and associative detachment, and collisional ionization by heavy particles. In Sections 9.3.2 and 9.3.3 we discuss the measurement and calculation of ionization coefficients in gases, while in Section 9.3.4 we discuss the
production of electrons by ionization in various forms of discharges. In order to save space we will generally cite only those recent references containing enough review of the topic so as to serve as a starting point for a literature search.

9.3.2 Measurement of Ionization Coefficients in Gases

The probability of electron impact ionization by electrons moving in a moderate or high density gas is customarily described by either a Townsend ionization coefficient \( \alpha \), the number of ionization events per electron per unit distance in the direction of electron drift, or by an ionization rate coefficient \( k \), the number of ionization events per unit time per electron per atom or molecule (Cherrington, 1979). Most measurements of ionization coefficients have been made using the parallel plane electrode system in which electrons are emitted photoelectrically from the cathode and drift through the gas under the action of a uniform electric field. Measurements are made of the current in the external circuit and/or of the light emitted by atoms or molecules excited by the electrons. The measurements may be steady-state or may follow the time dependence of the current or light output. Recent theory (Tagashira et al., 1977) has shown the importance of a careful distinction among various experimental techniques.

9.3.2.1 Spatial Growth Experiments

The spatial growth or steady-state Townsend experiments discussed in this subsection are conducted on a time scale which is long compared to the electron transit time across the apparatus. In the conventional Townsend experiments the growth of current is measured as the electrode separation is increased while keeping the gas density and electric field gas density ratio \( E/\rho \) constant. In the absence of secondary processes, the current measurements are fitted to an exponential of the form

\[
i = i_0 \exp (\alpha d),
\]

in which \( d \) is the electrode spacing and \( i_0 \) is the current leaving the cathode. Equation (9.3-1) describes the variation with distance of the current resulting from an electron avalanche in which the multiplication factor is \( \exp (\alpha d) \). For the conditions of these experiments keeping the \( E/\rho \) value constant keeps the distribution of electron energies constant.

When the emission of electrons from the cathode due to the arrival of photons, ions or metastables is significant or when electron attachment occurs the expression for the current ratio becomes more complicated (Dutton, 1978). In order to remove the first-order dependence of the Townsend coefficient on gas density \( \rho \) it is customary to express the results of experiment in terms of the ratio \( \alpha /\rho \). The quantity \( \alpha /\rho \) can be shown to be a function of \( E/\rho \). The results of numerous experiments have been summarized by Dutton (1975) and by Gallagher et al. (1983). The points of Fig. 9.3-1 show representative experimental values of \( \alpha /\rho \) for \( N_2 \) (Haydon and Williams, 1976). The curves show the results of recent theoretical calculations of \( \alpha /\rho \) which will be discussed later.
Fig. 9.3-1. Calculated and experimental ionization coefficients for N$_2$. Calculations: – – – neglecting new electron; — — temporal growth; ——— spatial growth. Experiment from Haydon and Williams (1976)

A problem with the conventional Townsend measurements is that it is sometimes difficult to separate the effects of secondary processes such as the release of electrons from the cathode by ions, metastables or photons from the growth of current caused by electron impact ionization. Therefore the more recent experiments make use of measurements of the current multiplication on a time scale short compared to ion transient times or metastable diffusion times (Haydon and Williams, 1976). Alternatively, measurements are sometimes made of the spatial variation of the light output (Bhattacharya, 1979). Occasionally measurements have been made of the growth of current at fixed electrode separation and gas density as the electric field is increased (Dutton, 1978).

9.3.2.2 Temporal Growth Experiments

The temporal growth or pulsed Townsend measurements are usually made on the time scale of 10 to 20 electron ionization collisions and are of the transient current flowing in the external circuit, the total light output, or of the time required for the electron current to grow to some critical value. Thus, the measurement is of a spatial integral of the electron distribution when the electrons are far from electrodes and is independent of the spatial gradients which may be of significance in other experiments (Tagashira et al., 1977).
For dc electric fields, this technique has been applied to a few gases (Raether, 1964; Dutton, 1978), to high \( E/\rho \) measurements (Byszewski et al., 1982), and to attaching gases (Fromhold, 1964). The experimental data are fitted to the function

\[ i(t) = i_0 \exp(k_i \rho t), \]  

(9.3-2)

where \( k_i \rho \) is determined from the exponential portion of a plot of \( i(t) \) versus the time \( t \). \( k_i \) is a function of \( E/\rho \) (Raether, 1964). We have not shown data from such experiments in Fig. 9.3-1 because of difficulties in the interpretation of the data at high \( E/\rho \) (Byszewski et al., 1982).

9.3.2.3 Temporal and Spatial Growth

The most difficult and, in principle, the most informative experimental measurements of ionization in gases are those in which measurements are made of both the temporal and spatial dependence of the electron density as the electrons drift and ionize under the action of a uniform electric field. The temporal distribution at a given point in space, i.e., the time-of-flight (arrival) distribution, or the spatial distribution of the electrons at a given time is generally determined by the light emitted by short-lived excited atoms or molecules (Fletcher, 1982). Thus far, these techniques have been used more for the determination of electron transport coefficients than for ionization coefficients.

9.3.2.4 High-Frequency Electric Fields

Most experimental measurements of ionization by electrons in the presence of high-frequency electric fields, e.g., microwave fields, do not fit into the previous categories since the dominant electron loss process is diffusion to the walls with its associated density gradients (Cottingham and Buchsbaum, 1963; MacDonald, 1966). However, in some cases (Felsenthal, 1966) the time scale is short enough so that the diffusive loss of electrons is not important.

9.3.2.5 Non-Equilibrium Ionization

In this section non-equilibrium ionization refers to the time and/or spatial variations in the apparent ionization coefficients which occur at short times and/or at very high values of \( E/\rho \). For example, experiment shows that the onset of ionization occurs some distance from the cathode when the electrons are emitted with an energy below the ionization energy (Folkhard and Haydon, 1973; Hayashi, 1980). In addition, experiments at very high \( E/\rho \) have shown that the apparent \( a \) coefficients are not independent of position in the steady-state Townsend experiment (Folkhard and Haydon, 1973; Dutton, 1978). This phenomenon is believed to be the result of the fact that some of the electrons gain energy from the electric field more rapidly than they lose energy in collisions with the atoms and molecules, i.e. they become "runaway" electrons. This effect will be discussed further in Section 9.3.3.3.
9.3.3 Theory of Ionization Coefficients in Gases

The theory of ionization by electrons in moderate and high-pressure gases is reviewed in many texts (Dutton, 1978). We will divide this discussion into theories primarily concerned with the calculation of ionization coefficients and the associated electron transport coefficients, i.e., solutions of the Boltzmann equation, and theories concerned with the statistical aspects of ionization and the associated electrical breakdown, i.e., statistical theories and numerical simulation.

9.3.3.1 Application of the Boltzmann Equation

The application of the Boltzmann equation to the calculation of electron impact ionization and transport coefficients has received considerable attention during the past decade (Huxley and Crompton, 1974; Tagashira et al., 1977; Lin et al., 1979; Kumar et al., 1980). Ionization coefficient data have played a key role in the determination of electron collision cross sections at the higher electron energies using solutions of the two-term spherical harmonic or Lorentz approximation to the Boltzmann equation (Pitchford and Phelps, 1982). The two-term approximation has also been used to predict ionization coefficients for various gas mixtures of technological interest (Aleksandrov et al., 1981; Sakai et al., 1979; Pfau and Winkler, 1978; Kline et al., 1979; Itoh et al., 1980; Cacciatore et al., 1982).

Important recent advances are the theoretical treatments of the effects of spatial density gradients (Tagashira et al., 1977; Kumar et al., 1980), electric field gradients (Alexandrov and Konchakov, 1981), anisotropic electron scattering (Haddad et al., 1981), large values of the ratio of the cross sections for inelastic collisions to the cross section for elastic scattering (Reid et al., 1980; Pitchford et al., 1981) and the production of new electrons in the ionization process (Wilhelm and Winkler, 1980; Yoshida et al., 1983). An example of the error resulting from calculations omitting the effects of spatial gradients and of the new electrons is shown by the dashed curve of Fig. 9.3-1 (Yoshida et al., 1983), which is much higher than the more accurate calculations shown by the lower two curves. The solid curve shows a calculation of $\alpha/\rho$ which includes spatial growth (Taniguchi, 1978), while the intermediate chain curve shows the $k/W$ values appropriate to the temporal growth experiments (Yoshida et al., 1983). Here $W$ is the calculated electron drift velocity. One important conclusion of recent investigations is that one expects a reasonably high degree of accuracy for ionization and other transport coefficients calculated using the two-term approximation under many conditions of practical importance. Investigations to define these conditions more generally are continuing (Allis, 1982; Braglia et al., 1982).

9.3.3.2 Statistical and Simulation Theories

Although they have received far less attention than the Boltzmann equation approach, the statistical and simulation theories of the growth of ionization in gases provide very useful models of early stages of discharges (Dutton, 1978; Hayashi,
1976; Tseng and Kunhardt, 1983). In addition, simulation theories using the Monte Carlo technique provide a very general method which is easily adapted to new problems. However, the high cost of Monte Carlo solutions for steady-state problems usually limits their use to testing the validity of the Boltzmann equation (Reid, 1979; Pitchford et al., 1981; Braglia et al., 1982).

9.3.3.3 Non-Equilibrium Theory

Both the simulation and Boltzmann approaches have been applied to the modeling of the non-equilibrium behavior of electrons. Of particular recent interest has been the modeling of the cathode fall regions of glow discharges (Boeuf and Marode, 1982). The onset of runaway behavior has been investigated by Ecker and Müller (1961) and others (Bhasavanich and Parker, 1977; Hayashi, 1976; Kunhardt and Byszewski, 1980). Non-equilibrium in the time domain has been reviewed recently by Wilhelm and Winkler (1979).

9.3.4 Ionization in Various Discharge Forms

In this section we will review studies of the role of ionization in various forms of discharges. A number of more extensive reviews are available (Pfau et al., 1969; Bekefi, 1976; Hirsh and Oskam, 1978; Cherrington, 1979).

9.3.4.1 Externally Maintained Discharges

Interest in externally maintained discharges has been high because of their use for electric discharge excited lasers (Bekefi, 1976; Velikov et al., 1977). Numerous papers have modeled the ionization by high-energy electrons (Bogdanov et al., 1982), the distribution in energy of the secondary and plasma electrons (Konovalov and Son, 1981; Bretange et al., 1981) and the criteria for stability against ionization waves, etc. (Nighan, 1976).

9.3.4.2 The Positive Column

The status of experiments and theories for the positive column of discharges in the rare gases has been thoroughly reviewed (Francis, 1956; Pfau et al., 1969; Cherrington, 1979; Ingold, 1979). Very detailed models of the relevant ionization and excitation processes, including the ionization of metastable states (Hyman, 1979; Armentrout et al., 1981), metastable-metastable collisional ionization and the effects of electron-electron collision, have been developed (Lighthardt and Keijser, 1980; Cernogora et al., 1981; Dothan and Kagan, 1982; Winkler et al., 1983). Theory and experiment concerned with the role of ionization in the striations found in the positive column have been reviewed by Garscadden (1978). Discussions of the balance between ionization and loss of electrons by diffusion include the transition
from ambipolar to free diffusion as the electron density decreases (Muller and Phelps, 1980), the effects of attachment (Maccullum, 1970; Mikhalev and Selin, 1974), and the role of excited states in ionization and recombination (Fujimoto, 1979; Devos et al., 1979).

9.3.4.3 Electrode Effects

Only a few references present recent experimental work on ionization in the cathode fall of glow discharges, while there has been much progress on the theory (Baksht and Yur'ev, 1979; Emeleus, 1981; Boeuf and Marode, 1982). Very little work has been done recently on the anode of glow discharges (Francis, 1956). A great deal of progress has been made in understanding the cathode regions of arcs (Holmes, 1978; Hantsche, 1979), as well as the anode (Kimblin, 1974; Chen and Pfender, 1980). Ionization in the hollow cathode has been the subject of a number of recent investigations (Helm, 1979; Reshnov, 1981).

9.3.4.4 Corona and Streamers

The understanding of coronas and streamers occurring during electrical breakdown is progressing rapidly as the result of the application of improved experimental and theoretical techniques (Goldman and Goldman, 1978; Gallimberti, 1978; Craggs, 1979; Marode et al., 1979; Yoshida et al., 1979; Sigmond, 1980). In particular, detailed models of streamer development have led to reasonable agreement between experiment and theory. The effect of the non-equilibrium caused by high electric fields at the head of the streamer on the streamer growth is currently being investigated (Abbas and Bayle, 1981; Byszewski and Rheinhold, 1982; Tzeng and Kunhardt, 1983).

9.3.4.5 Arcs and Lightning

Most models of the arc column, such as found in lightning, assume that the electron, positive ion and neutral atom densities are in local thermodynamic equilibrium (LTE) at their common temperature and so do not consider the details of the ionization process (Finkenburg and Maeker, 1956). In the rare gases significant departures from LTE occur (Uhlenbush, 1974; Gleizes et al., 1982) and models of these arcs usually use some form of the collisional-radiative model of ionization and recombination (Biberan et al., 1979; Vriens and Smeets, 1980). See Sections 9.1 and 9.2. At the higher relative gas densities it may be necessary to take into account the effects of the excitation and deexcitation of highly excited atoms by neutral atoms (Devos et al., 1979; Bacri et al., 1982).
References

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