Considerable data are available on ionization and attachment coefficients in certain gases. A good summary on these coefficients has been documented by Prasad and Craggs\(^1\). The early work can conveniently be divided into two groups. One involving low $E/p$'s (volt cm\(^{-1}\) torr\(^{-1}\)) and high pressures (of the order of hundreds of torr) and the second involving high $E/p$'s and low pressures (a few torr). (The use of $N$ in place of $p$ is explained in chapter 4). A variety of techniques have been used for the measurement of the swarm coefficients $\alpha$, $\gamma$, $\chi$ and $\delta$. For example, Loeb\(^2\), Prasad and Craggs\(^1\), McDaniel\(^3\) and Raether\(^4\) have given a detailed treatment of these techniques. In this chapter, only a brief description of the methods used for the measurement of ionization and attachment coefficients is given along with a brief review of the results obtained by earlier workers.

Detailed discussions of the earlier investigations on gases of particular interest to this thesis are presented in chapters 5 to 8.
3.2 **Review of previous experimental results**

This section contains the results of some of the earlier works on the measurement of swarm coefficients and sparking potentials in oxygen, nitrogen, dry air, artificial air, ammonia, water vapour and humid air.

3.2.1 **Results in oxygen**

(a) **Primary ionization and attachment coefficients**

Considerable amount of data are available on the primary ionization and attachment coefficients in oxygen in addition to the one summarized by Prasad and Craggs\(^5\). Some of the earlier workers who obtained valuable information on these coefficients are as follows: \(\text{Masch}^6\) over the range \(60 \leq \varepsilon/p \leq 500\); \(\text{Harrison and Geballe}^7\) and Prasad and Craggs\(^5\) in the range \(25 \leq \varepsilon/p \leq 70\); \(\text{Freely and Fisher}^8\) in the range \(32 \leq \varepsilon/p_0 \leq 100\); \(\text{Dutton et al}^9\) for \(\varepsilon/p \leq 43\); \(\text{Betts and Davies}^{10}\) in the range \(50 \leq \varepsilon/p_0 \leq 250\); \(\text{Wagner}^{11}\) in the range \(30 \leq \varepsilon/p \leq 50\); \(\text{Price and Moruzzi}^{12,13}\) in the range \(25 \leq \varepsilon/p_{20} \leq 116\); and \(\text{Blair and Whittington}^{14}\) in the range \(36 \leq \varepsilon/p_{20} \leq 70\). Only \(\text{Schlunbohm}^{15}\) has covered a very wide range of \(\varepsilon/p\) (upto 2500). \(\text{Sukhum et al}^{16}\) have investigated for \(\varepsilon/p \leq 80\); only for attachment and detachment, employing the usual experimental techniques. The disagreement of the results of \(\varepsilon/p\) of some of the
above workers especially at the lowest values of $E/p$ is accounted for by Dutton et al$^9$. Results of Masch$^6$ on $\alpha/p$ are probably not reliable, particularly in the lower ranges of $E/p$ for the attachment processes were ignored and experiments were done in mercury contaminated atmosphere. The coefficient $\alpha/p$ is, in general, found to increase with $E/p$. The results pertaining to the coefficient, $\eta/p$ (Fig. 3.1), show a large variation and some of the difficulties that give rise to these variations are accounted for by Lucas et al$^{17}$ and the effect of detachment is taken care of by considering 'effective attachment coefficient' e.g. by Price et al$^{13}$. The general variation of this coefficient with $E/p$ is found to increase at a rapid rate in the beginning, reaches a maximum and then falls off less rapidly. Further discussion on these coefficients is given in chapter 5.

(b) Detachment coefficients

Investigations of Frommhold$^{18}$, Prasad$^{19}$, Wagner$^{11}$, Price et al$^{13}$ and O'Neill and Craggs$^{20}$ show electron detachment in the ionization current growth phenomena in oxygen at very low values of $E/p$. Lucas et al$^{17}$ suggest that the electron attachment is via a three-body process and detachment may occur from $O^-$, $O_2^-$ and $O_3^-$, the products of attachment and ion-molecule reactions, with $O_2^-$ having the lowest theoretical onset energy and $O_3^-$ the highest depending upon
the $E/p$. However, electron detachment is negligibly small for $E/p \geq 50$ (Sukhum et al.\textsuperscript{16}) and of no significance.

(c) \textit{Secondary ionization coefficient}

Though secondary ionization coefficient, $\gamma$, depends on the nature of the cathode, its variation with $E/p$ gives an apparent indication of the type of secondary process that is predominative. In general, $\gamma$ is an increasing function of $E/p$, usually on account of the increase in energy of the ions involved in the emission of electrons from the cathode. At low $E/p$'s, on the other hand, there is sometimes (vide fig. 3.2) a marked increase of $\gamma$ with decreasing $E/p$ indicating that electron emission due to the incidence of photons at the cathode is important.

(d) \textit{Sparking potential measurements}

Betts and Davies\textsuperscript{10} report breakdown voltage measurements in the region around the Paschen minimum. Their experiments further show that Paschen's law is obeyed in spectroscopically pure oxygen and the Paschen curve for values of $p_d$ corresponding to minimum breakdown voltage, the curve is made up of two linear parts with different slopes but in oxygen, contaminated with phosphorous pentoxide, deviations from Paschen's law are
observed agreeing with the observations of Frike\textsuperscript{21}. Betts and Davies\textsuperscript{10} explain the transition with one slope to the other of the Paschen curve to the right of minimum breakdown voltage in terms of the onset of appreciable $O^-$ production at that $E/p$ value. No such transition is evidenced in the Paschen curves in the work of Betts and Davies\textsuperscript{10} for impure oxygen agreeing with the results of Schonhuber\textsuperscript{22}. Spreads of a few per cent in the measured values of the breakdown voltages have been reported by Debitetto and Fisher\textsuperscript{23} and Freely and Fisher\textsuperscript{8} but not by Dutton et al\textsuperscript{9}.

3.2.2 Results in nitrogen

(a) Primary ionization coefficient

The values of $\alpha/p$ of Lutton et al\textsuperscript{24}, Harrison\textsuperscript{25} and Heylen\textsuperscript{26} obtained in better gas purity and controlled experimental conditions in nitrogen are lower than those of Ayres\textsuperscript{27}, Masch\textsuperscript{6}, Posin\textsuperscript{28} and Bowls\textsuperscript{29} in $E/p_o \leq 90$. This may possibly be due to preferential ionization of the apparent mercury vapour present in the gas under test. Comparatively more reliable data on this coefficient over the range $100 \leq E/p_o \leq 510$ in pure nitrogen and nitrogen containing radioactive impurities are found in the investigations of Jones\textsuperscript{30,31}. Jone's values of $\alpha/p$ in nitrogen with radioactive impurities are lower than those in pure
nitrogen and was attributed to the preferential ionization of long-lived metastables of nitrogen produced by the decay radiations of the radioactive impurities. Tagashira and Lucas\textsuperscript{32} determined the values of \( \zeta / p \) using both Townsend and Lucas equations over \( 54 \leq E/p_0 \leq 99 \), so selected in order to achieve swarm conditions without electrode effects and non-equilibrium regions. The values of \( \zeta / p \), obtained by Lucas equation are found to have differences with those of Masch\textsuperscript{6}, Bowls\textsuperscript{29} and Harrison\textsuperscript{33}. The more recent results reported by Daniel and Harris\textsuperscript{34} in the range \( 28 \leq E/p_{20} \leq 52 \) establishing the dependence of \( \zeta / p \) only on \( E/p \) are in good agreement with those of Dutton et al\textsuperscript{24}, Cookson et al\textsuperscript{35} and Debitetto and Fisher\textsuperscript{36}, with same impurity content of the gas. Determination of the values of \( \zeta / p \) over the range of \( 30 \leq E/p_0 \leq 450 \) by Kontoleon et al\textsuperscript{37} for estimating the electron energy distribution closely agree with those of Masch\textsuperscript{6}, Tagashira and Lucas\textsuperscript{32}, Bowls\textsuperscript{29}, Jones\textsuperscript{30,31} and Posin\textsuperscript{28}. The latest results (to the knowledge of the author) on this coefficient by Maller and Naidu\textsuperscript{38} covering a range of \( 100 \leq E/p_0 \leq 1000 \) are in excellent agreement with those of Polkard and Haydon\textsuperscript{39} but higher than those of Bagnall and Haydon\textsuperscript{40} in crossed electric and magnetic fields. It appears that experimental parameters impose a significant limitation on the accuracy of \( \zeta / p \) values because of the onset of non-equilibrium ionization.
(b) \textit{Secondary ionization coefficient}

Danial et al\textsuperscript{41} report that \( \gamma \) is independent of \( p \), and increases with increasing \( E/p \) in the range \( 28 \leq E/p_{20} \leq 50 \) and conclude that the observed variation is due to photoionization from the cathode that predominates the secondary ionization process. Jones\textsuperscript{30} gives the values of \( \gamma \) over the range \( 100 \leq E/p_0 \leq 470 \) that are derived from the experimentally determined values of \( \alpha/p \) and sparking potentials \( V_s \). The observed variation of \( \gamma \) with \( E/p \) is explained quantitatively by assuming resonance neutralization of an ion to occur as it approaches the metal surface of the cathode; for sufficiently high approach velocities Auger de-excitation follows. Jones\textsuperscript{31} also reports that there is no significant difference in the values of secondary co-efficients for the pure and contaminated (with radioactive impurities) nitrogen. Danial and Harris\textsuperscript{34} give both the experimentally determined and theoretically calculated values for \( \gamma \) in the same range of \( E/p \) covered by Danial et al\textsuperscript{41} and arrive at the same conclusions. They also report that the cathode processes are significant which was confirmed by the fact that a spark between clean electrodes lead to a considerable reduction in the values of \( \gamma \). The secondary coefficients are found to be pressure-dependent at low values of \( E/p \) \( (100 \leq E/p_0 \leq 300) \) and no such dependence was observed at higher values of \( E/p \).
(300 \leq E/p_0 \leq 1000) by Maller and Naidu\textsuperscript{38}. The large amount of scatter in the values of $\gamma$ is attributed to the effect of non-equilibrium and equilibrium ionization processes. No other data on $\gamma$ except that due to Maller and Naidu\textsuperscript{38}, seem to have been reported in the literature, at the higher values of $E/p$.

(c) **Sparking potential measurements**

The investigations of Jones\textsuperscript{30,31}, Parish and Tedford\textsuperscript{42}, Daniel et al\textsuperscript{43} and Daniel and Harris\textsuperscript{34} show that Paschen's law holds in nitrogen. In the region of Paschen minimum the breakdown voltage is strongly dependent on gas purity and electrode material and for this reason the individual results in this region show the greatest differences. At high values of breakdown field strength, deviations from Paschen’s law are observed (vide fig. 3.3) and ascribed to field emission at cathode.

3.2.3 **Results in dry air and artificial air**

(a) **Primary ionization and attachment coefficients**

Investigations by Sanders\textsuperscript{44}, Masch\textsuperscript{6} and Townsend\textsuperscript{45} are said to have been carried out in air contaminated with mercury vapour and neglecting electron attachment in obtaining $\alpha/p$ values over a range $20 \leq E/p \leq 1000$. These results are thoroughly discussed by Loeb\textsuperscript{2}. Several
investigations have been carried out later in dry air employing high vacuum techniques and different degrees of gas purities over the range $25 \leq E/p \leq 65$ by many workers and by Raja Rao and Govinda Raju\textsuperscript{46} over a wide range of $100 \leq E/p \leq 1000$. Calculated values of $\alpha/p$ are reported by Narayana Swamy and Harrison\textsuperscript{47} in artificial air ($79\%$ of $N_2 + 21\% O_2$) over a range of $40 \leq E/p \leq 450$ and appear to be the only data reported in the literature in this gas (to the authors knowledge).

Fig. 3.4 shows the values of the primary ionization coefficient, $\alpha/p$, in low ranges of $E/p$ in dry air obtained by Llewellyn Jones and Parker\textsuperscript{48} ($39 \leq E/p \leq 45$), Harrison and Geballe\textsuperscript{7} ($25 \leq E/p \leq 65$) Prasad\textsuperscript{49} ($25 \leq E/p \leq 45$), Dutton et al\textsuperscript{50,51} ($35 \leq E/p \leq 40$), Frommhold\textsuperscript{52} ($35 \leq E/p \leq 80$) and Ryzko\textsuperscript{53} ($30 \leq E/p \leq 60$). All the above workers employed the well known Townsend steady state method i.e., measuring the gas amplified current using plane parallel electrodes, except the latter two, who used dynamic avalanche method. The latter method is as follows: the current due to electron avalanches resulting from the release of short pulses of photoelectrons of duration 30 to 40 n sec. at the cathode of a uniform field gap is amplified and displayed on a C.R.O. using wideband and low noise amplifiers. Although the data of Llewellyn Jones and Parker\textsuperscript{48} were obtained
with considerable care the results are in error due to neglect of attachment being closely \( \frac{(\alpha - \eta)}{p} \). As far as the attachment coefficient is concerned, the data shown in fig. 3.5 over the range \( 37 \leq \frac{E}{p} \leq 47 \) (Ryzko)\(^{53}\) is the accurate available and is well supported by mathematical analysis. In all the above investigations generally no pressure dependance of \( \frac{\eta}{p} \) is observed. At low values of \( \frac{E}{p} \) \( (2 \leq \frac{E}{p} \leq 36) \) Bradbury\(^{54}\), Kuffel\(^{55}\) and Chatterton and Griggs\(^{56}\) measured \( \frac{\eta}{p} \) using Bradbury electron filter apparatus using radio frequencies and the results are shown in fig. 3.6. Chatterton particularly studied certain possible sources of error, neglected partly or wholly by other workers, viz., (1) diffusion losses of electrons in the discharge, (2) collection of negative ions at the r.f. grids and (3) current multiplication at the grids, including ionization, secondary emission of electrons and electron detachment. Consequently the results of Bradbury\(^{54}\) and Kuffel\(^{55}\) cannot probably be considered reliable over the whole range because of the neglect of the above sources of error.

(b) **Detachment coefficients**

Detachment of electrons from negative ions has been reported by Frommhold\(^{52}\), Ryzko and Astrom\(^{57}\) and
Lutton and Morris. For example, the former in his experiments on the growth of single electron avalanches in oxygen and air at low pressure (\( \leq 32 \) torr) detected a delayed electron component which could be attributed to the detachment of electrons from negative ions. In fact he has given a relationship between \( E/p \) and the product of detachment frequency and pressure \( (\tau p) \) over the range \( 35 \leq E/p \leq 80 \). All the observations are only qualitative and no quantitative information about the detachment is available in air.

(c) **Secondary ionization coefficient**

The experimental data of Dutton et al.\(^{50,51}\) and Prasad\(^{49}\) under steady-state conditions, show that the cathode surface has a marked influence on the secondary ionization even at higher values of pd (2300 torr-cm). The secondary ionization coefficient, \( \gamma \), is found to depend on \( E/p \) although minor variations are observed over the pressure range \( 400 \leq p \leq 1000 \) torr indicating that the destruction of excited molecules in collisions of the second kind does not appear to be an important process in air. Further a general increase of \( \gamma \) with \( E/p \) is observed. The only data available on this coefficient in dry air over a wide range of \( E/p \) is that of Raja Rao and Govinda Raju\(^{46}\). No values of this coefficient appear
to have been reported so far in the literature in artificial air.

(d) **Sparking potentials measurement**

Although the sparking potential data of Dutton et al.\textsuperscript{50,51} and Prasad\textsuperscript{49} show no pressure dependence or deviation from Paschen's law, investigations of Farish and Tedford\textsuperscript{59}, Daniel et al.\textsuperscript{60} and Raja Rao and Govinda Raju\textsuperscript{46}, using plane parallel electrode systems, show that Paschen's law does not hold for air. Detailed discussion of data is however, presented in chapter 6.

3.2.4 **Results in ammonia**

(a) **Primary ionization and attachment coefficients**

Values of $\alpha/p$ in ammonia over a range of $30 \leq E/p_{20} \leq 1000$ are reported by Risbud and Mudra\textsuperscript{61} (this appears to be the only work over a wide range of $E/p$). The variation of $\alpha/p$ with $E/p$ is rapid for low values and less rapid for high values of $E/p$. Parr and Moruzzi\textsuperscript{62} report that attachment proceeds via the dissociative attachment process only for $E/p_{20} \geq 9$ and is found to increase rapidly with $E/p$ until $E/p_{20} \approx 30$ where it then reaches a plateau at $7.4 \times 10^{-19}$ cm$^2$. The values of $\eta/p$ of Parr and Moruzzi\textsuperscript{62} closely agree only with those of
Bailey and Duncanson\textsuperscript{63} not with the results of Bradbury\textsuperscript{64}. The values of Risbud and Naidu\textsuperscript{61} over $20 \leq \frac{E}{p_20} \leq 40$ are lower than those of Parr and \textsuperscript{62} and Bailey and Duncanson\textsuperscript{63} and higher than those of Bradbury\textsuperscript{64}. All these investigations reveal that the attachment arises mainly through dissociative processes.

(b)) \textbf{Secondary ionization coefficient}

No values of $\gamma$ in ammonia appear to have been reported in the literature so far (to the knowledge of the author), possibly due to the heavy attachment in the large majority of the above mentioned investigations.

(c) \textbf{Spark potential measurements}

Apparently no measurements of breakdown potentials in ammonia are reported so far in the literature.

\subsection*{3.2.5 Results in water vapour}

(a) \textbf{Primary ionization and attachment coefficients}

There is significant amount of published work on the attachment properties of water vapour but not on the values of ionization coefficients. Craggs and McDowell\textsuperscript{65} reviews the work of Townsend\textsuperscript{45} ($50 \leq \frac{E}{p} \leq 100$) and Bailey and Duncanson\textsuperscript{63} ($12 \leq \frac{E}{p} \leq 32$), where electron
attachment is neglected by the former and ionization by the latter who used diffusion techniques. The values of $\alpha/p$ of Ryzko $^{53}$ (25 $\leq E/p \leq$ 60) and Prasad and Craggs $^{66}$ (30 $\leq E/p \leq$ 50, using dynamic avalanche method) are higher than those in dry air or humid air (see chapters 6 and 8). The recent data of Risbud and Naidu $^{61}$ (20 $\leq E/p_20 \leq$ 400) on $\alpha/p$ agree only to $\pm$ 3% with that of Prasad and Craggs $^{66}$ and Ryzko $^{53}$.

The results of Ryzko $^{53}$ on $\eta/p$ are not in good agreement with the data of Bailey and Duncanson $^{63}$, Bradbury $^{64}$, Prasad and Craggs $^{66}$ and Crompton et al $^{67}$ but they do appear to merge with that of Kuffel $^{55}$. The data of Risbud and Naidu $^{61}$ are in good agreement with those of Crompton et al $^{67}$ and they also report that $\eta/p$ is independent of gas pressure suggesting that attachment arises in this gas through dissociative attachment process.

(b) **Secondary ionization coefficient**

No measurements on $\gamma$ in water vapour are reported by any of the above investigators. There is no upcurving in the observations of Prasad and Craggs $^{66}$, in the log(I,d) plots even at a value of E/p = 50 at 3 cm gap. The work of Hackam $^{68}$ appears to be the only available data in the range 50 $\leq E/p_0 \leq$ 67 on this coefficient calculated using Ryzko's $^{53}$ $\alpha/p$ and indicate a rapid fall of $\gamma$ with E/p.
(c) **Sparking potential measurements**

The only data on the breakdown voltages in water vapour appear to be that of Prasad and Craggs\(^{66}\) over a range of the spark parameter \(10 \leq pd \leq 50\) and Hackam\(^{68}\) over the range \(10 \leq p_d \leq 300\). Hackam reports that the minimum breakdown voltage is about 2550 V with nickel and 2000 V with stainless steel and occurs over the \(p_d\) range 17-23 torr-cm.at. a gap of about 0.5 cm (see sec. 7.3.1) both higher than those of Prasad and Craggs\(^{66}\) with platinum electrodes. The minimum sparking potential in water vapour appears to be higher than that reported in most of the common gases (Meek and Craggs\(^{69}\)). Hackam\(^{68}\) concludes that addition of water vapour to other gases in an enclosed system, either from surface desorption or from external leaks, would not result in lowering the holding voltage of the device.

3.2.6 **Results in humid air**

(a) **Primary ionization and attachment coefficients**

\(\xi/p\) and \(\eta/p\) in humid air for various concentrations of water vapour are reported by Prasad and Craggs\(^{66}\) (30 \(\leq E/p \leq 45\)) and Ryzko\(^{70}\) (25 \(\leq E/p \leq 55\)), the latter using dynamic avalanche method. Kuffel\(^{55}\) also reports values of \(\eta/p\) for 21 torr of water vapour in a
total pressure of 760 torr. The value of $\alpha / p$ in humid air at any $E/p$ is larger than in dry air and depends on the ratio of the partial pressure of water vapour ($p_w$) to the total pressure ($p$) of humid air, and $\alpha / p$ increases with higher concentrations of water vapour. At a constant ratio of $p_w / p$, a relative increase in $\alpha / p$ is higher in the lower range of $E/p$. Typical $\alpha / p$ values are also reported in humid air (room air without drying) by Raja Rao\textsuperscript{71} over a wide range of $E/p$ ($50 \leq E/p \leq 1000$) at a relative humidity of 56% along with the values in dry air, the former being found to be slightly higher than the latter.

In the above investigations\textsuperscript{66,70,55} $\eta / p$ increases approximately linearly with $E/p$. It also increases markedly with higher concentrations of water vapour in air. Kuffel reports that the value of $\eta / p$ in humid air is nearly twice the corresponding value in dry air at certain values of $E/p$ (e.g. $E/p = 26$). Ryzko's investigations in humid air reveal hardly any three body processes in the range $25 \leq E/p \leq 55$, as suggested by Dutton et al\textsuperscript{72} in dry air.

(b) **Secondary ionization coefficient**

Secondary processes are cathode processes and are evidenced by the calculations of Prasad and Craggs\textsuperscript{66}
(using Townsend's steady state method) which also show that $\gamma$ increases generally with $E/p$ (in the range $30 \leq E/p \leq 45$). These values of $\gamma$ are mainly calculated from sparking potential data. For no up-curving was observed in all the $(\log I,d)$ plots. Meek gives a preliminary interpretation of the low secondary coefficients in terms of photoabsorption, a mechanism suggested earlier by Köhramann.

(c) **Sparking potential measurements**

The results of Köhramann and Prasad and Craggs show a large increase in sparking potential of air with increase in humidity at any given pd. This increase in sparking potential is clearly due to pronounced increase in the values of $\eta/p$ and also due to very low values of $\gamma$ as indicated by both the relatively poor photoelectric yields and by the absence of up-curving in the $(\log I,d)$ plots.
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3.2 Variation of $\omega/\alpha'$ with $E/\text{Po}$ for oxygen $\uparrow, \bullet, \circ$ (Dutton, Evans and Morgan, 1967); $\times$ (Debitetto and Fisher, 1958), $\Delta, \ldots, \ldots$ (Prasad and Craggs, 1961); $\ldots, \ldots$ (Freely and Fisher, 1964)
FIG - 3 Breakdown voltages in nitrogen at high nd

- Palm (1934)
- Finkelmann (1937)
- Howell (1939)
- Zeier (1932)
- Orgler (1900)
- Kolev (1973)
FIG. 3.4 VALUES OF $\alpha/p$ IN DRY AIR AS A FUNCTION OF $E/\sigma$

- TRUE VALUES OF $\alpha/p$, PRASAD (1959, 1960)
- APPARENT VALUES OF $\alpha/p$, PRASAD (1959, 1960)
- VALUES OF $\alpha/p$ OBTAINED BY HARRISON AND Geballe (1953)
- VALUES OF $\alpha/p$ OBTAINED BY Llewellyn Jones AND Parker (1952)
- VALUES OF $\alpha/p$ OBTAINED BY SANDERS (1932 1933)
- VALUES OF $\alpha/p$ OBTAINED BY FROMMELD (1964)
**Fig. 3.5** $\eta/p$ as a function of $E/P$ in dry air
A - RIZKO; B - FROMMOLD; C - PRASAD

**Fig. 3.6** $\eta/p$ as a function of $E/P$ in dry air
A - BRADBURY
B - KUFFEL (290°K)
C - PRASAD
D - GEBALLE AND HARRISON
E - CHATTERTON