Pramāṇa, Vol. 8, No. 3, 1977, pp. 292–301. © Printed in India.

Validity of diffusion theory in radio frequency breakdown in molecular gases in longitudinal magnetic field

S N SEN and D C JANA
Department of Physics, North Bengal University, Darjeeling 734430

MS received 27 July 1976

Abstract. The breakdown of a gas excited by a radio frequency voltage of frequency 5·6 MHz has been studied in a cylindrical discharge tube 7·2 cm long and 2·9 cm in dia and fitted with two internal electrodes at a distance of 2·5 cm in hydrogen, oxygen and air within the pressure range of a few microns to 2 torr in the presence of a longitudinal magnetic field varying from zero to 800 G. Experimental results indicate that the breakdown is diffusion controlled and the values of (α/P) at different E/P values calculations obtained by Brown as well as by Kihara's theory have been compared with (α/P) values obtained in the literature. It is concluded that the diffusion theory is also valid when the frequency of the exciting voltage is scaled down to radio frequency provided the collision frequency is much higher than the exciting frequency. The change of diffusion length in the presence of longitudinal magnetic field has been obtained from measured E/P values and comparison with theoretical values indicates that there is quantitative agreement for small (H/P) values where H is the magnetic field. The calculated values of pressure at which the breakdown voltage shows a minimum in the presence of magnetic field is in very good agreement with experimental values. It is concluded that in the presence of magnetic field also the loss of electrons takes place predominantly by the process of diffusion.

Keywords. Breakdown of gas; diffusion; radiofrequency.

1. Introduction

The study of the breakdown of a gas excited by high frequency electromagnetic field has shown that the breakdown voltage depends upon the pressure of the gas, the dimension of the discharge tube and the frequency of excitation. The dominant factors responsible for the loss of electrons are diffusion and mobility and if the gas is electron attaching, the loss also takes place by electron attachment. It has been observed that when the pressure of the gas is of the order of a few millitorr and the length of the discharge tube is large as compared with the mean free path of the electrons in the gas, both the mobility and diffusion are the dominant factors by which electrons are removed. On the other hand, when the gas pressure is high and the exciting frequency of the applied voltage lies in the micro wave region, the electrons are lost mainly by diffusion. The theoretical method of calculating the breakdown voltage of a gas excited by high frequency voltages at high pressure has been developed by Herlin and Brown (1948) where the dominant factor for electron removal process has been assumed to be diffusion. Starting from a molecular model, Kihara (1952) developed a theoretical method to calculate the breakdown voltage of a gas under high frequency excitation taking into consideration the loss due to mobility and diffusion. In a series of papers from this laboratory (Sen and Ghosh 1963; Sen and Bhattacharjee 1965, 1966, 1967) the experimental results have indicated that when the pressure is of the order of a few millitorr and the frequency of excitation of the order of a few MHz, the major electron removal processes are diffusion and mobility.

To test the limitations of the diffusion theory, it is proposed here to undertake some experiments on the breakdown voltages of gases where the frequency of the exciting voltage is of the order of a few MHz and the pressure of the gas is of the order of a few torr. To study the effect of attachment, breakdown measurements have been made in some electron attaching gases such as air and oxygen. The object of the present investigation is to find out whether the loss mechanism remains the same when the frequency of the exciting voltage is scaled down from microwave to radio frequencies keeping the pressure in the range of a few torr.

The breakdown of a gas excited by a radio frequency field in presence of a magnetic field has been studied previously by Lax *et al* (1950) who performed experiments on the breakdown voltage of helium containing a small admixture of mercury vapour and obtained breakdown curves for different values of the pressure. Ferritti and Veronesi (1955) performed experiments for frequencies ranging from 10 to 30 MHz in air, the magnetic field varying from 0–600 G and observed a lowering of breakdown voltage in the presence of magnetic field. Sen and Bhattacherjee (1969) performed experiments in the case of air, hydrogen, oxygen and carbon dioxide in the presence of a magnetic field from 300–1800 G.

Brown (1956) has explained the change of breakdown voltage observed in presence of magnetic field by assuming that the diffusion length in the presence of a magnetic field is altered according to the equation

$$\Lambda_H^2 = \Lambda^2\left[1+\frac{\omega_B^2}{\nu_C^2}\right]$$

where Λ and Λ_H are respectively the diffusion lengths in the absence and in the presence of magnetic field. ω_B is the electron cyclotron frequency and ν_C is the collision frequency. To make a further test of diffusion theory in the presence of a magnetic field, it is also proposed to verify the above equation of Brown from the experimental results obtained in the present set of experiments. The results are expected to prove the validity of the diffusion theory in the presence and absence of the applied magnetic field.

2. Experimental arrangement

The method of measurement of breakdown voltage was the same as was used earlier (Sen and Ghosh 1963). The discharge tube of 7·2 cm long, cylindrical, and fitted with two internal electrodes with a separation distance of 2·5 cm and discharge tube was 2·9 cm in dia. The radio frequency voltage was supplied from a tuned grid tuned plate oscillator, the frequency of the oscillator being variable from 3·5–11 MHz and the output of the oscillator could be continuously varied from 0–550 volts. The r.m.s. output voltage was measured with a vacuum tube voltmeter, The pressure of the gas was measured with a calibrated Mcleod gauge. The magnetic field was provided by an electromagnet, the lines of force were parallel to the length of the discharge tube which was placed entirely within the polepieces of the electromagnet. The magnetic field was measured with a calibrated fluxmeter. Keeping the magnetic field at a constant value, the pressure was varied and the breakdown voltage measured for various values of gas pressure. The experiments were repeated and the results were found to be reproducible within ± 1%.

Pure and dry air was passed through phosphorus pentoxide to remove traces of water vapour. Hydrogen was prepared by electrolysing warm concentrated solution of barium hydroxide in a hard glass U-tube fitted with nickel electrodes in which hydrogen gas was liberated at the cathode. The gas was dried by passing it over broken pieces of potassium hydroxide and then over purified phosphorus pentoxide. Pure oxygen was evolved at the anode in the electrolysis of barium hydroxide solution and was passed through pure concentrated sulphuric acid before collection in the discharge tube.

3. Results and discussion

The breakdown voltages for hydrogen, oxygen and air have been plotted for different values of pressure (0·1 to 2·4 torr) with and without magnetic field (110 g to 795 g) in figures 1, 2 and 3 respectively. It is observed that the breakdown voltage is always smaller in the presence of the magnetic field than in its absence for all values of pressure and the pressure at which the breakdown voltage becomes

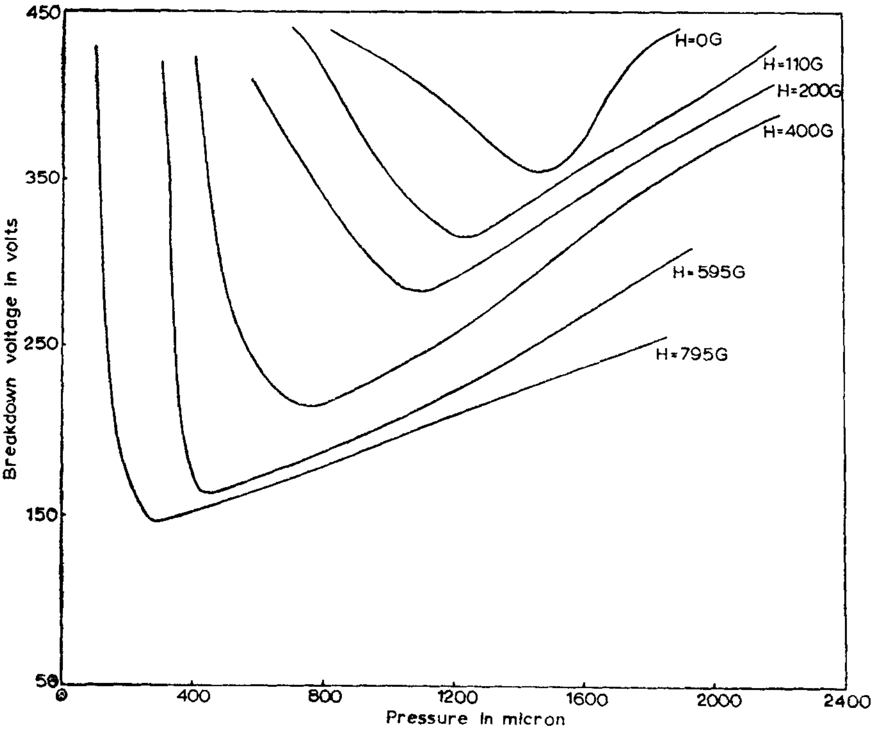

Figure 1. Variation of breakdown voltage with pressure: hydrogen.

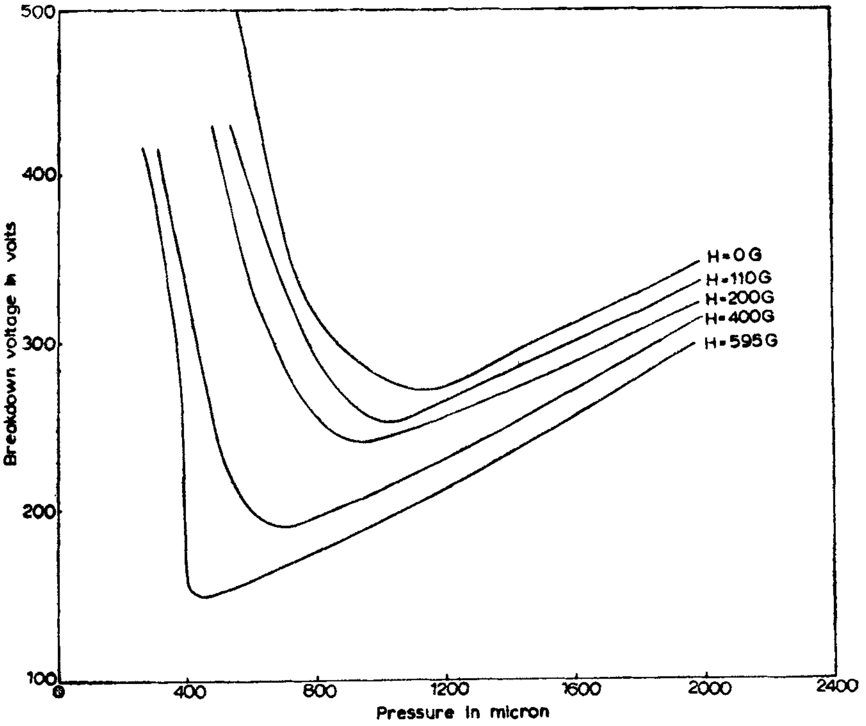

Figure 2. Variation of breakdown voltage with pressure: oxygen

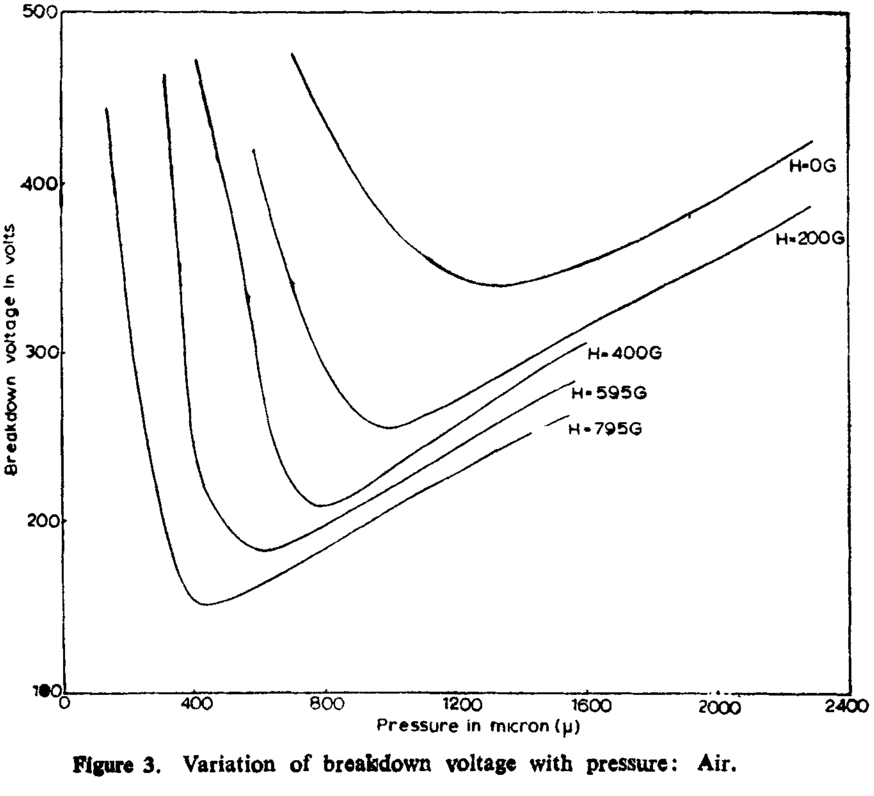

Figure 3. Variation of breakdown voltage with pressure: Air.

minimum always shifts to lower pressure with the increase of the magnetic field. To provide a meaningful interpretation, it is proposed to examine the results in the light of the prevalent theories (Brown 1959; Kihara 1952). In order to determine which process is predominant in electron removal under the present experimental set up the following points have been considered:

(i) According to Brown for the diffusion theory to be valid the dimensions of the discharge tube must be small compared to wavelength of the exciting power. As the wavelength is of the order of 51·2 cm and the length of the discharge tube is 7·2 cm and 2·6 cm dia. this condition is satisfied.

(ii) The maximum mean free path of the gases used here is 0·6 cm at a pressure of 0·1 torr (Townsend 1947) which is much smaller than either the length or radius of the tube.

(iii) The amplitude of electron oscillation when calculated from the equation

$$x = \frac{eE_0}{m\omega\,[\omega^2 + \nu^2]^{1/2}}$$

where E_0, the field intensity, is 0·02 cm at a pressure of 1 torr and will be smaller at higher pressures.

(iv) The collision frequency is v_r/λ_e where v_r is the random velocity and λ_e is the mean free path and is of the order of 10^9 collisions/sec and is much greater than the exciting frequency even at a pressure of 1 torr.

Under the above conditions, it is apparent that the electrons make many oscillations of small amplitude, because the motion is restricted by collisions and the cloud of electrons appear stationary (there being no drift motion), spreading outwards only by diffusion. Hence loss due to drift can be neglected. New charged particles are formed due to ionizing collisions and loss due to diffusion predominates. In case of electron attaching gases, the loss due to attachments should also be taken into consideration.

As stated above, under the present experimental set up and range of pressure investigated the electron suffers many collisions per oscillation of the field. Brown pointed out that as pressure increases, mean free path decreases and the energy gain per mean free path is proportional to mean free path at constant E. In order to cause breakdown, the field must increase in inverse proportion with the mean free path or in direct proportion with pressure. Thus at high pressure where the electrons make many collisions per oscillation their behaviour is much the same as in the case of d.c. field. The value of (α/P) where α is the ionization coefficient can then be calculated from the experimental values of E/P from the Townsend's relation

$$\alpha/P = A_0 \exp\,[-B_0/(E/P)] \tag{1}$$

where A_0 and B_0 are the values of constants for a particular gas.

Kihara (1952) has treated the phenomenon of electrical discharge by adopting a proper molecular model for collision processes. Assuming a model for the cross section of the molecule for elastic, exciting and ionization collisions with a Maxwellian distribution of electron velocities which is nearly valid for the case of molecular gases studied here, he has deduced that

$$\frac{\alpha}{P} = \left(\frac{N}{P}\right)\frac{\sigma}{C_i}\left(\frac{3\lambda}{\rho}\right)^{1/2} \exp\left[\frac{-\,mC_i^2\left(\frac{N}{P}\right)(3\lambda\rho)^{1/2}}{2e\cdot(E/P)}\right] \tag{2}$$

where σ is a molecular constant equivalent to collision cross section, λ is another constant having the dimension of cm^3 S^{-1}, N is the number density of the gas atom, K is the Boltzman constant, ρ is another molecular constant having the dimension of cms. The values of these molecular constants have been provided by Kihara (1952).

The values of (α/P) have been calculated from eq. (1) using the experimental values of (E/P) obtained in the present investigation for hydrogen. The values of (α/P) have also been calculated from eq. (2) for corresponding values of (E/P) using the numerical values of the constants given by Kihara. The results for hydrogen have been plotted in figure 4 and for purposes of comparison, the published experimental values of (α/P) from literature are also plotted in the figure. In the case of electron attaching gases such as air and oxygen, the loss due to attachment is also taken into consideration and the ionization coefficient (α/P) have been calculated from the expression

$$\frac{\alpha}{P} = \frac{\alpha_a}{P} + A_0 \exp\left(-\frac{B_0}{E/P}\right). \tag{3}$$

Similarly from Kihara's theory it can be shown that when attachment is taken into consideration

$$\nu - \nu_a = N \cdot \frac{3\sigma}{C_i} \frac{KT_e}{m} \exp\left[-\frac{mC_i^2}{2KT_e}\right]$$

and hence

$$\frac{\alpha}{P} = \frac{\alpha_a}{P} + \frac{N}{P} \cdot \left(\frac{\sigma}{C_i}\right)\left(\frac{3\lambda}{\rho}\right)^{1/2} \exp\left[\frac{-mC_i^2\left(\frac{N}{P}\right)(3\lambda\rho)^{1/2}}{2e \cdot E/P}\right]. \tag{4}$$

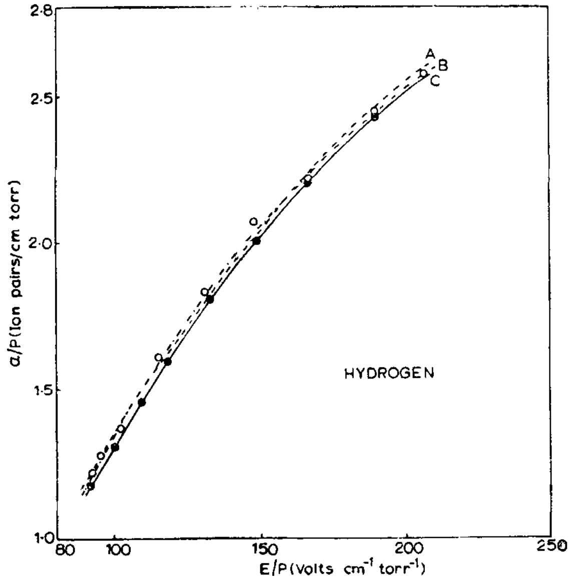

Figure 4. Variation of (α/P) with E/P for hydrogen A: Kihara; B: Brown; C: Literature values.

The values of (α_e/P) for air and oxygen for different (E/P) values have been obtained from (Brown 1959) and (α/P) values have been calculated from eq. (3) using experimental values of (E/P) obtained and also from eq. (4) and then plotted in figures 5 and 6 for oxygen and air respectively together with values obtained from literature.

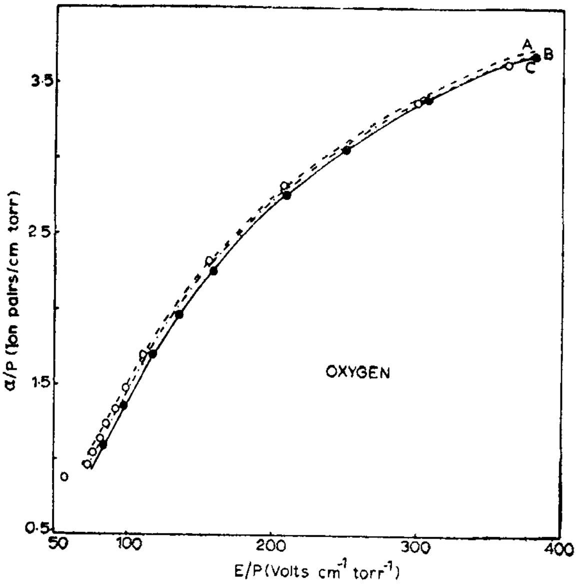

Figure 5. Variation of (α/P) with (E/P) for oxygen: A: Kihara; B: Brown; C: Literature values.

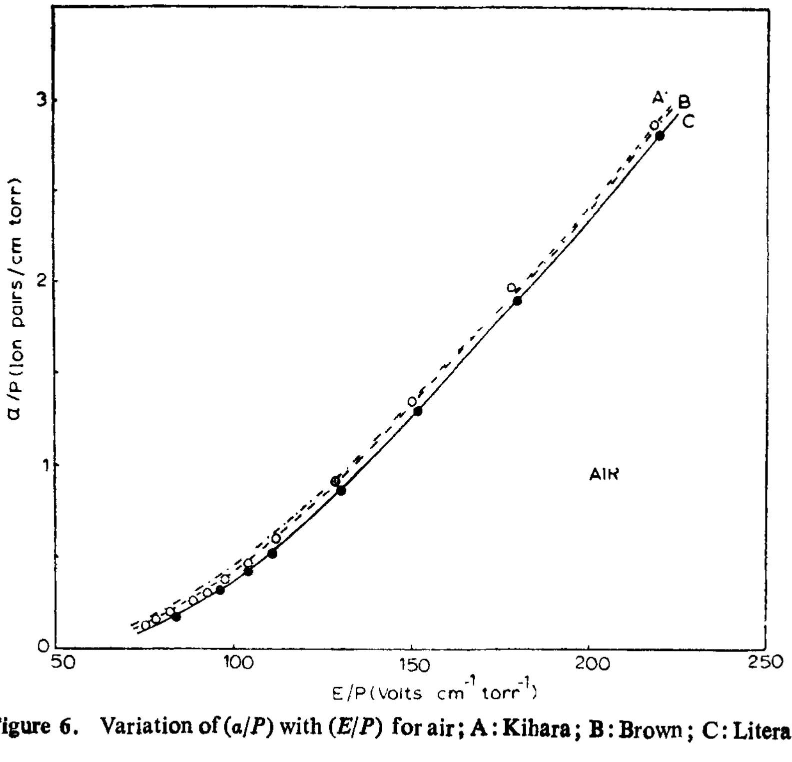

Figure 6. Variation of (α/P) with (E/P) for air; A: Kihara; B: Brown; C: Literature values.

It is thus evident that in the case of all the gases studied here the values of (α/P) calculated from breakdown voltage are in fairly good agreement with the values of (α/P) obtained from literature for (E/P) values studied here. Further the results calculated from Brown's expression are in better agreement than those calculated from Kihara's theory. This may be due to uncertainties in the values of molecular constants introduced by Kihara. We can further conclude that under the present experimental conditions and where the electrons make a large number of collision per oscillation, diffusion is the dominating factor for the loss of electrons and the breakdown process is identical with the d.c. breakdown mechanism.

4. Effect of magnetic field

In the above discussion we have concluded that under the present experimental set up and for the values of pressure and the frequency of the applied radio frequency field used, diffusion is the main electron removal process. However, the effect of an external magnetic field is to modify the breakdown mechanism to the same extent as the process of diffusion. As the diffusion perpendicular to the magnetic field is reduced, the breakdown field will show a reduction in value. The mean square displacement travelled by an electron is proportional to diffusion constant and Brown has shown that the effective diffusion length Λ_H appropriate to infinite parallel plate is given by

$$\Lambda_H = \Lambda\left[1+\frac{\omega_B{}^2}{\nu_C{}^2}\right]^{1/2} \tag{5}$$

where ω_B is the cyclotron frequency $= (eH)/m$ and ν_C is the collision frequency at the pressure considered. In a recent communication (Sen and Jana 1976) we have measured the collision frequency of the electrons in hydrogen, oxygen and air by the radio frequency conductivity method and the value at a pressure of 1 torr for hydrogen is $1{\cdot}74 \times 10^9$, for oxygen $3{\cdot}58 \times 10^9$ and for air $3{\cdot}222 \times 10^9$. In order to verify whether Brown's expression for the modified diffusion length is valid the values of Λ_H/Λ have been calculated for each gas separately for different values of H/P from 50–500 G torr^{-1}. To see whether these are consistent with the experimental values, Λ_H/Λ has also been calculated from values of E and E_H obtained experimentally. It has been shown that the discharge is diffusion controlled and the breakdown criteria is given by

$\nu/D = 1/\Lambda^2$ or $(\alpha\mu E)/D = 1/\Lambda^2$ where μ is the mobility as $\mu/D = e/(KT_e)$, where T_e is the electron temperature

$$\left(\frac{\alpha}{P}\right)\frac{eEP}{KT_e} = \frac{1}{\Lambda^2}.$$

From Townsend's equation $\alpha/P = A_0 \exp(-B_0/(E/P))$ and

$$\frac{KT_e}{e} = \frac{L}{\sqrt{R}}\cdot\frac{E}{P} = r\cdot\left(\frac{E}{P}\right) \text{ Von Engel (1955)}$$

where $r = L/\sqrt{R}$ and L is the mean free path of the electron in the gas at a pressure of 1 torr and $R = 2m/M$ where m is the mass of the electron and M is the mass of the ion.

Hence

$$A_0 \exp\left(-\frac{B_0 P}{E}\right) \cdot \frac{P^2}{r} = \frac{1}{\Lambda^2}$$

or

$$E/P = \frac{B_0}{\log\,[A_0 P^2 \Lambda^2/r]} \,. \tag{6}$$

When magnetic field is present if E_H is the breakdown field for the same value of P

$$E_H/P = \frac{B_0}{\log\,[A_0 P^2 \Lambda_H{}^2/r]} \tag{7}$$

Hence

$$\frac{\Lambda_H}{\Lambda} = \left[\exp \frac{B_0 P\,(E - E_H)}{E E_H}\right]^{1/2} . \tag{8}$$

From the experimental values of E and E_H values of Λ_H/Λ for all the three gases have been obtained from eq. (8) and entered in table 1 for H/P varying between 50 and 500 gauss torr^{-1}.

From a comparison of the theoretical and experimental values, it is evident that the values are more or less consistent with one another and lends additiona support to the assumption that the loss of electrons under the present experimental set up is governed mainly by diffusion.

We further note that maximising eq. (6) with respect to pressure, the pressure at which the breakdown voltage becomes a minimum, is given by

$$P_{\min} = \frac{2E_{\min}}{B_0} \tag{9}$$

and in the presence of magnetic field

$$\frac{2\,(E_H)_{\min}}{(P_H)_{\min}\left[1 + \dfrac{\omega_B{}^2}{C^2\,(P_H)^2_{\min}}\right]} = B_0 \tag{10}$$

where $(P_H)_{\min}$ is the pressure at which the breakdown voltage becomes $(E_H)_{\min}$ and C is the collision frequency at a pressure of 1 torr, from eqs (9) and (10);

Table 1. Theoretical and experimental values of Λ_H/Λ for different (H/P) values

Hydrogen			Oxygen			Air		
(H/P) Gauss torr^{-1}	$\frac{\Lambda_H}{\Lambda}$ (Theory)	$\frac{\Lambda_H}{\Lambda}$ (Expt.)	H/P Gausst torr^{-1}	$\frac{\Lambda_H}{\Lambda}$ (Theory)	$\frac{\Lambda_H}{\Lambda}$ (Expt.)	H/P Gauss torr^{-1}	$\frac{\Lambda H}{\Lambda}$ (Theory)	$\frac{\Lambda H}{\Lambda}$ (Expt).
78·57	1·008	1·038	61·1	1·002	1·031	90·90	1·116	1·260
89·80	1·010	1·140	111	1·006	1·053	111·1	1·169	1·167
181·8	1·042	1·212	160·6	1·008	1·060	200	1·481	1·382
250·0	1·077	1·293	222	1·014	1·108	250	1·682	1·793
350·0	1·186	1·301	285·7	1·024	1·079	333·3	2·077	1·892
533·3	1·317	1·342	333·3	1·034	1·134	371·8	2·263	1·982

Table 2. Theoretical and experimental values of $(P_H)_{\min}$ from equation (11).

Magnetic field (Gauss)	Hydrogen		Oxygen		Air	
	$(P_H)_{\min}$ (calc) torr	$(P_H)_{\min}$ Expt. torr	$(P_H)_{\min}$ Calc. torr	$(P_H)_{\min}$ Expt. torr	$(P_H)_{\min}$ Calc. torr	$(P_H)_{\min}$ Expt. torr
110	1·268	1·25	1·062	1·05	..	..
200	1·0792	1·09	0·9899	0·95	0·9661	0·99
400	Indeterminate	0·75	0·6534	0·66	0·6972	0·73
595	..	..	Indeterminate	0·45	Indeterminate	0·60

$$(P_H)_{\min} = \frac{P_{\min}\dfrac{(E_H)_{\min}}{E_{\min}} \pm \left(P^2_{\min}\dfrac{(E_H)^2_{\min}}{E^2_{\min}} - 4\omega_B{}^2/c^2\right)^{1/2}}{2}. \tag{11}$$

The values of $(P_H)_{\min}$ thus calculated for the three gases for different values of the magnetic field are given in table 2.

5. Conclusions

It is thus concluded that when the frequency of excitation is much smaller than the collision frequency, the major factor responsible for electron removal is the process of diffusion and this is also the dominating factor when the magnetic field is applied. The mechanism of breakdown becomes almost identical with d.c. breakdown of gases and the experimental results are in agreement with theoretical values calculated on the basis of these assumptions. It is further noted that eq. (11) becomes invalid for values of magnetic field greater than 400 gauss which shows that deductions are valid for low values of magnetic field, and also corroborated by the values of Λ_H/Λ as shown in table 1.

References

Brown S C 1956 *Handbuch Phys.* (Berlin: Springer Verlag)
Brown S C 1959 *Basic Data in Plasma Phys.* (New York: Technology Press)
Ferritti L and Veronesi P 1955 *Nuovo Cimento* **2** 639
Herlin M A and Brown S C 1948 *Phys. Rev.* **74** 291
Kihara T 1952 *Rev. Mod. Phys.* **24** 43
Lax B, Allis W P and Brown S C 1950 *J. Appl. Phys.* **21** 1297
Sen S N and Ghosh A K 1963 *Can. J. Phys.* **41** 1443
Sen S N and Bhattacharjee B 1965 *Can. J. Phys.* **43** 1543
Sen S N and Bhattacharjee B 1966 *Can. J. Phys.* **44** 3270
Sen S N and Bhattacharjee B 1967 *J. Phys. Soc. Jpn.* **22** 1477
Sen S N and Bhattacherjee 1969 *Br. J. Appl. Phys.* (*J. Phys. D*) **2** 1739
Sen S N and Jana D C 1976 *Proc. Nat. Symp. Atomic and Molecular Collision Processes*, Indian Assn. for Cultivation of Science, Calcutta
Townsend J S 1947 *Electrons in Gases* (London: Hutchinson)
Von Engel E 1955 *Ionised gases* (Oxford: Clarendon Press)