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Ionization processes in helium at pressures near to atmospheric

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Abstract. Determinations of primary α and secondary ω ionization coefficients in helium at pressures p_0 near to atmospheric and low values of E/p_0 (E is the electric field) show that $\alpha/p_0 = f(E/p_0)$ and $\omega/\alpha = \phi(E/p_0)$ for $30 < p_0 d < 730$ cm torr. The results obtained for ω/α and its dependence on E/p_0 are shown to be adequately accounted for on the basis of the assumption that the predominant secondary ionization process in helium at high pressures is the destruction of metastable states in the gas with the consequent production of non-resonance photons which liberate secondary electrons at the cathode.

1. Introduction

In recent years there has been considerable interest in the determination of ionization coefficients in helium (Davies, Llewellyn Jones and Morgan 1962, Chanin and Rork 1964, Dutton, Llewellyn Jones and Rees 1965). One of the main reasons for this interest is that helium is, atomically, a relatively simple gas about which a great deal of knowledge is available; this enables accurate theoretical computations (Dunlop 1949, Abdelnabi and Massey 1953, Heylen and Lewis 1963) to be made for comparison with the experimental data. The main difficulty in the experimental determination of ionization coefficients in helium is the extreme dependence of the coefficients on the purity of the gas. This is particularly so at low values of the parameter E/p_0 (E is the electric field, p_0 the gas pressure at 0 °c) and values of p_0 near to atmospheric, where, as shown recently (Dutton *et al.* 1965, hereafter referred to as I), a change in purity as low as about 1 part in 10^6 can lead to changes in α of about 10%. In I an apparatus was described for the determination of ionization coefficients in this range of low E/p_0 (about 4 v cm^{-1} torr^{-1}); the results showed that by using the techniques there described it was possible to obtain consistent values of the coefficients. The values of α/p_0 lay between the values obtained at low pressures by Townsend and McCallum (1928) and by Chanin and Rork (1964). The differences in the values of α/p_0 in the different investigations may well be due to differences in the purity of the gas samples used, but since the experiments were carried out at different pressures the possibility also exists that there may be departures from similarity which appear as a pressure effect. It is thus of interest to investigate further the question of similarity: in the present paper values of the ionization coefficients over a range of values of p_0 for a given value of E/p_0 are reported. The information which can be obtained concerning the predominant secondary ionization process from this investigation of similarity and the previously observed (I) dependence of the secondary ionization coefficient ω/α on E/p_0 is also discussed.

2. Investigation of similarity

In the previous work (I) measurements of the ionization current I as a function of the gap separation d for constant values of p_0 and E/p_0 were found to be in agreement with the generalized Townsend relationship

$$I = \frac{CI_c \exp\{(\alpha/p_0)(p_0 d)\}}{1 - (\omega/\alpha)[\exp\{(\alpha/p_0)(p_0 d)\} - 1]} \tag{1}$$

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where I_c is the current measured at a value of E/p_0 which is sufficiently low for no ionization to occur so that $I_c = I_0/C$, where C is a constant and I_0 is the initial externally generated current from the cathode. These previous results showed that within the experimental error the values of α/p_0 and ω/α were independent of d for given values of E/p_0 and p_0.

In the present work, possible dependence of the coefficients α/p_0 and ω/α on pressure was first investigated by measuring I as a function of d at a given value of E/p_0 for a series of different gas pressures in the range $355 < p_0 < 560$ torr. A value of $E/p_0 = 4$ v cm^{-1} torr^{-1} was chosen since this gave the largest range of pressure, at a given value of E/p_0, for which reliable determinations of α/p_0 and ω/α could be made with the present apparatus. The gas sample used was that corresponding to the highest purity attainable with this apparatus (see I), although it is not claimed that it is completely pure. The results are shown in figure 1, from which it can be seen that both α/p_0 and ω/α are independent of the gas pressure over the range investigated.

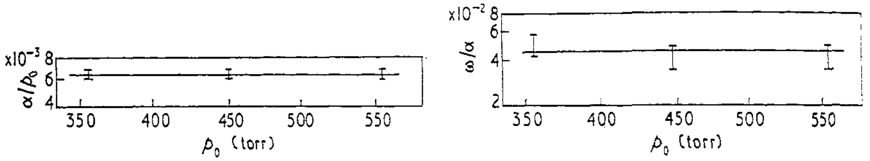

Figure 1. Values of α/p_0 and ω/α for $E/p_0 = 4$ v cm^{-1} torr^{-1} and various gas pressures.

In order to investigate further whether α/p_0 and ω/α were dependent on gas pressure, measurements were also made of I/I_c as a function of p_0 over the range $128 < p_0 < 560$ torr using a fixed electrode separation of 1·1 cm at $E/p_0 = 3{\cdot}7$ v cm^{-1} torr^{-1}. The results are shown in figure 2. It can be seen that the experimental values are in good agreement with the curve computed from equation (1) using constant values of the coefficients α/p_0 and ω/α. Furthermore, the value of α/p_0 so obtained is in agreement, to within the experimental

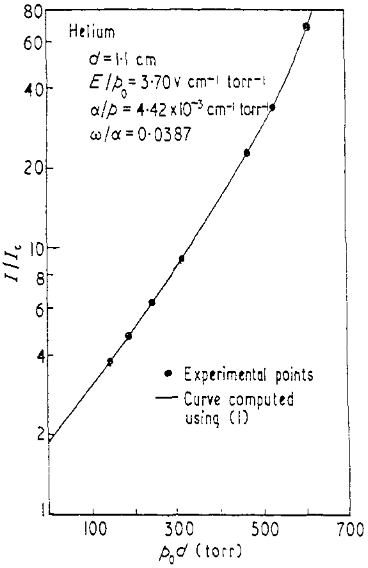

Figure 2. Spatial growth of ionization with pressure at constant values of d and E/p_0.

error, with that obtained at the same value of E/p_0 from the curve of I/I_c as a function of d at constant pressure (see I). The value of ω/α is about half that obtained from the corresponding $(I/I_c, d)$ curve; the reason for this was that, as indicated by the detailed measurement of I_c, the cathode was in a different surface state in the two experiments, and as is shown in §3.4 the process of secondary ionization is cathode dependent. These results at a constant value of d strongly suggest that the coefficients, particularly α/p_0, are independent of the pressure over a wider range of pressure than that already established by the results given in figure 1.

It seems unlikely therefore that the differences between the various values of α/p_0 obtained by different observers at low values of E/p_0 in helium result from a pressure effect arising from the operation of processes which violate similarity, but rather that the reason for the differences lies in the purity of the gas sample. This is currently being further investigated by measurement of the ionization coefficients of gas samples from different sources and subjected to different treatments.

3. Secondary ionization processes

It is now well known that there are a large number of possible secondary ionization processes in helium (Phelps 1960, Davidson 1962). It is of interest to consider whether any information can be obtained about the predominant secondary ionization process in helium at high pressures from the results given in the previous section and from the previously reported (I) variation of ω/α with E/p_0.

It was shown in I that a marked increase in the value of ω/α occurred with decreasing values of E/p_0. This type of variation is characteristic of secondary emission due to the incidence of excited states and photons (produced from the decay of excited states) on the cathode and arises because the number of excitations per ionizing collision in helium increases as E/p_0 diminishes for low values of the mean electron energy (< 6 ev). The variation cannot be explained on the basis of secondary ionization due to atomic or molecular ion action at the cathode, which gives quite a different dependence of ω/α on E/p_0. Further evidence that the production of excited states in the gas by electron–atom collisions is likely to play an important role in secondary ionization in helium may be obtained from consideration of the energy balance.

3.1. *Electron energy balance equation*

In the steady state the average energy E received by the electrons in travelling 1 cm in the direction of the electric field E at 1 torr pressure must be balanced by the average energy lost by the electrons in elastic and inelastic collisions. This condition is expressed in helium by the following equation:

$$E = \alpha V_i + \left(\frac{\epsilon}{150m}\right)^{1/2} \frac{\epsilon\lambda Q_e}{W_-} + \sum_n (\epsilon_x)_n \tag{2}$$

where the terms on the right-hand side represent, respectively, the electron energy loss by ionization, elastic collisions and excitations of electronic levels.

The loss of energy due to ionizing collisions was calculated using the experimental values of α/p_0 given in I and the ionization potential V_i of the helium atom. The following data were used in the computation of elastic losses: Townsend's (1947) experimental values of ϵ, the mean electron energy, and Townsend and Bailey's (1923) values of λ, the fraction of the mean electron energy lost per elastic collision; values for the total elastic cross section Q_e were taken from Phelps, Fundingsland and Brown (1951) and values of the electron drift velocity W_- from Phelps, Pack and Frost (1960). The ionization (ϵ_i) and elastic (ϵ_e) losses were thus computed and equation (2) was then used to determine the total excitation loss $\sum_n (\epsilon_x)_n$ as a function of E/p_0. The results are given in the table where values of E/p_0, ϵ_i, ϵ_e and $\sum_n (\epsilon_x)_n$ are in units of v cm^{-1} torr^{-1}.

It is clear from the table that about 70% of the energy loss in this region is due to excitation and this, together with the experimentally observed variation of ω/α with E/p_0, suggests that the predominant processes of secondary ionization in helium at high pressure

Energy lost in elastic collisions, excitation of electronic levels and ionization

E/p_0	α/p_0	ϵ_i	ϵ_e	$\Sigma\,(\epsilon_x)_n$
3·0	0·00130	0·0320	0·887	2·08
3·5	0·00326	0·0802	0·818	2·60
4·0	0·00649	0·160	0·772	3·07
4·5	0·0111	0·273	0·741	3·49
5·0	0·0169	0·416	0·722	3·86

arise from the production of excited states. It is thus of interest to see whether it is possible to compute values for ω/α on this basis for comparison with the experimental data. In order to do this it is necessary to discuss the excitation processes in detail.

3.2. *Excitation processes*

The only excited states which are of importance to the production of secondary electrons at the cathode are the lowest 2^1P resonance state (21·2 ev) and the two metastable 2^3S and 2^1S states (19·8 ev and 20·6 ev). Non-resonance photons liberated in transitions from one excited state to another do not have enough energy to produce a significant photoelectric current at the cathode.

Corrigan and von Engel (1958) calculated the ratio of the number θ_m of atoms in metastable levels to the number θ_r in radiating levels produced by one electron in moving 1 cm in the direction of the electric field, and found that for low values of E/p_0 in the range 3–5 v cm^{-1} torr^{-1}, the production of metastable atoms far exceeded that of the resonance states. For example, at $E/p_0 = 5$ v cm^{-1} torr^{-1} the ratio θ_m/θ_r was about 10, whereas for a value of $E/p_0 = 3$ v cm^{-1} torr^{-1} the value of θ_m/θ_r was as large as about 30. Furthermore it has been concluded by Phelps (1960) that at high pressures a large fraction ($\sim$40%) of the atoms in the 2^1P resonance state are de-excited to the lower 2^1S metastable state with the emission of infra-red quanta. It is therefore considered that excitations to the metastable states play the dominant role in the mechanism of secondary ionization in helium at low values of E/p_0.

To proceed further it is necessary to investigate whether the metastable states produced in the gas at high pressures diffuse to the electrodes, or whether most of the metastable states are destroyed in the gas by collision with ground state atoms to yield non-resonance radiation.

3.3. *Production of non-resonance radiation from metastable states*

If a metastable atom is generated at a distance x from the cathode, then the average time it would take to diffuse to the cathode is of the order x^2/D where D is the diffusion coefficient of the particular metastable state considered. In the present experiments the influence of secondary ionization on the growth of ionization current became experimentally significant at electrode separations from about 0·8 to 1·3 cm. Most of the metastable states were produced near the anode, so that an approximate estimate of the average time taken by a metastable atom to reach the cathode was made by taking a value of 1 cm for x.

By using Phelps' (1955) values for the diffusion coefficient, it is found that at the gas pressures ($\sim$300 torr) used in the present experiments both the singlet and triplet metastable states required times of the order of one second to reach the cathode from their point of production, whereas the average cross section for volume destruction of these states (Phelps 1955) leads to a value for their lifetime of about 10^{-5} sec. It is thus concluded that the metastable states were almost wholly destroyed in the body of the gas near to the region in which they were produced.

The destruction of singlet metastable states results in the production of non-resonance radiation only (Phelps 1955), whereas the destruction of triplet metastable states can result in the production of stable molecular metastable states (Phelps 1955), as well as giving rise to the production of non-resonance radiation (Colli and Facchini 1954). However, if the predominant secondary process were the liberation of electrons at the cathode by metastable molecules, then the spatial growth of ionization with d would be given by an expression (Davidson 1958) in which ω/α is a function of d: experimentally, however, ω/α was independent of d to within the experimental error, and thus molecular metastable action at the cathode did not appear to play a dominant role in these conditions.

On the basis of the above considerations, therefore, it seems likely that in helium at the high pressures used in the present investigation the secondary ionization was predominantly due to the electronic excitation of atoms to the metastable states. These states were then largely destroyed in the gas in times of the order of 10^{-4} sec with the consequent production of non-resonance radiation. Such radiation cannot be absorbed in the gas and travels unimpeded to the electrodes with the velocity of light.

It is now of interest to compute theoretically the magnitude of the secondary ionization coefficient in helium on the basis of this assumption and to compare the computed values with the experimental values.

3.4. *Calculation of secondary ionization coefficient*

When the metastable helium atoms are destroyed in the gas near to the region in which they are originally produced with the production of non-resonance photons, a coefficient θ_n may be defined such that $\theta_n dx$ is the number of non-resonance photons due to transition from the nth metastable state produced on the average by one electron in moving a distance dx in the direction of the field.

Only two metastable atomic states exist in helium, $2^3\mathrm{S}$ (19·8 ev) and $2^1\mathrm{S}$ (20·6 ev), so that

$$\sum_n (\epsilon_x)_n = \sum_n \theta_n V_n = \theta_t(19{\cdot}8) + \theta_s(20{\cdot}6) \tag{3}$$

where t and s refer to transitions from triplet and singlet states respectively. As the two metastable states are high-energy states, and differ in energy by only 0·8 ev, it is considered that no significant error will be introduced by writing equation (3) as $\sum_n (\epsilon_x)_n = \theta_m(20)$ v, where θ_m is the total number of non-resonance photons due to transitions from both metastable states. Since $\sum_n (\epsilon_x)_n$ has already been computed as a function of E/p_0, values for θ_m/p_0 in cm^{-1} torr^{-1} may be readily obtained from

$$\theta_m/p_0 = \sum_n (\epsilon_x)_n/20. \tag{4}$$

If, in the steady state, $(\delta/\alpha)'$ represents the number of electrons emitted at the cathode by non-resonance photons per primary ionization, then

$$\left(\frac{\delta}{\alpha}\right)' = \frac{kg\theta_m/p_0}{\alpha/p_0} \tag{5}$$

where k is the number of electrons per photon released by the non-resonance photons at the cathode and g is a geometrical factor which depends upon the angle subtended at the cathode by the head of the electron avalanche. Since the photons are non-resonance photons, absorption of the photons can be neglected.

Equation (5) has to be corrected to take into account back-diffusion of electrons into the cathode at high pressure in helium. An experimental investigation (which will be reported elsewhere) showed that the Thomson expression for back-diffusion was in good agreement with the experimental results in helium at high pressure so that the expression for the secondary ionization coefficient δ/α is

$$\frac{\delta}{\alpha} = \frac{1}{1 + u/(6\pi)^{1/2}W_-} \, \frac{kg\theta_m/p_0}{\alpha/p_0} \tag{6}$$

where u is the most probable velocity with which the electrons leave the cathode, and W_- is the drift velocity of the electrons. The following data were then used to compute the magnitude of δ/α.

The work function of the cathode surface, measured by a retarding potential method, was 4·52 ev so that the value of u was $1{\cdot}4 \times 10^8$ cm sec^{-1}. Values of W_- in helium were taken from the experimental data of Phelps, Pack and Frost (1960), and the value of g was estimated

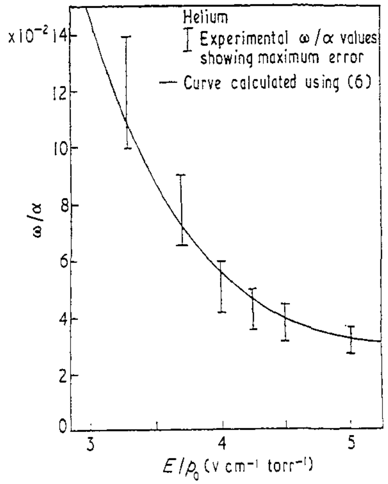

Figure 3. Comparison of experimental and theoretical values of ω/α.

from the geometry of the system to be about $\frac{1}{6}$. The measured values of α/p_0 are given in the table and θ_m/p_0 was found using equation (6) together with the computed values of the total excitation loss (see table). Stebbings (1957) obtained a value for k of 0·2 electrons per photon for helium resonance photons from the 2^1P state striking a gold surface, so that the assumption of a value of 0·15 electrons per photon for the less energetic non-resonance photons from the metastable states striking a copper surface was not unreasonable. The values of δ/α computed using the above data in equation (6) are shown in figure 3, where they are compared with the experimental values. It can be seen that good agreement exists between the calculated and experimental values.

3.5. *Conclusions*

It is concluded from figure 3 that the destruction of metastable states in the gas, with the consequent production of non-resonance photons which liberate secondary electrons at the cathode, can adequately account for the observed dependence of ω/α on E/p_0 at low values of E/p_0.

Furthermore, for the pressure range near to atmospheric investigated in the present work, the rate of destruction of metastable states in the gas is sufficiently high for the value of ω/α in the steady state to be independent of pressure at a constant value of E/p_0. Moreover, it follows from equation (6) that since the values of θ_m/p_0, α/p_0 and W_- are functions of E/p_0 only, the value of the secondary ionization coefficient should be independent of the value of the electrode separation. These conclusions are consistent with the experimental results obtained in the present investigation which showed that the secondary ionization coefficient obeyed the similarity relationship $\omega/\alpha = \phi(E/p_0)$.

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