

FIG. 2. Secondary electron yields for Ar<sup>+</sup> and Ar<sup>0</sup> on Mo.

The variation of the yield of secondary electrons with kinetic energy of the incident neutral particles is shown as curve (N) of Fig. 2. On the same figure we show as curve (I) the results for the variation of  $\gamma_i$ with energy. There is a divergence in the energy dependence of  $\gamma_i$  and  $\gamma_n$  above the "threshold" for kinetic ejection. This divergence is clearly apparent

by comparison of the plots (I) and (S), where S is obtained by adding  $\gamma_n$  to  $\gamma_\tau$ ;  $\gamma_\tau$  is computed from (I) as  $\gamma_{\pi} \simeq 0.074$ . It has previously been assumed<sup>2,4</sup> that for E > 1 keV  $\gamma_{\tau}$  is independent of energy, i.e.,  $\gamma_i(E)$  $=\gamma_{\pi}+\gamma_{k}(E)$ , where  $\gamma_{k}(E)$  is the electron yield due to the kinetic energy (E) of the incident ion. Arifov et al.<sup>2</sup> cite their results for Ar<sup>+</sup> and Ar<sup>0</sup> on Mo as evidence in support of this assumption. However, our results shown in Fig. 2 give slopes for the ascending part of the curves as

$$d\gamma_i/dE = 0.06/\text{keV}$$
 and  $d\gamma_n/dE = 0.04/\text{keV}$ .

It has been suggested that this observed divergence shows there is a dependence of potential ejection efficiency on energy, i.e., for  $Ar^+ \gamma_{\pi}$  increases with E. Comparative measurements of  $\gamma_i$  and  $\gamma_n$  for He should then exhibit an inversion of the behavior shown here for Ar since  $\gamma_{\pi}(E)$  for He<sup>+</sup> is considered to be a decreasing function of energy.<sup>5</sup> Our measurements to date on the He<sup>+</sup>, He<sup>0</sup> system have not clearly shown such expected behavior.

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## Net Frequency of Ionization in Oxygen\*

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A new microwave method for determining r, the net frequency of ionization in a gas, has been developed and applied to oxygen. The determination is made from measurements of the formative time of a pulsed microwave discharge as a function of the time between pulses and from a knowledge of the rate of decay of the electron density in the afterglow of the discharge. Microwaves of 3.2-cm wavelength were used and the pressure of the oxygen was varied from 5 to 20 mm of Hg. Values of  $\nu/p$  were determined for values of E/pfrom 36 to 62 V cm<sup>-1</sup> (mm Hg)<sup>-1</sup>. The results from the microwave experiment agree well with dc data.

#### 1. INTRODUCTION

CEVERAL experimental methods have been de-**D** veloped to measure the net frequency of ionization,  $\nu$ , in a gas<sup>1,2</sup> in which a knowledge of the electron diffusion coefficient either at low electron densities or throughout a range from low to high electron densities is required. If the ambient electron density is maintained at a relatively high value throughout the experiment the diffusion losses are by ambipolar diffusion and under certain conditions can be made negligible compared with

other losses. In the present experiment a pulsed microwave source is used to produce the electrons. The net frequency of ionization is determined from measurements of the formative time of a "steady state" pulsed microwave discharge as a function of the time between pulses and from a knowledge of the rate of decay of the electron density in the afterglow of a microwave discharge. The term "steady state" implies that the electron density repeats its cycle with each incident microwave pulse. The formative time  $\tau$  is the time taken for the electron density to build up from some initial value  $n_0$ , that is present at the arrival of the microwave pulse to some convenient larger value  $n_b$ . For simplicity the upper value is taken to be that which produces a large attenuation and a large reflection of the incident microwave power used to produce the electron density.

<sup>&</sup>lt;sup>4</sup> N. N. Petrov, Bull. Acad. Sci. U.S.S.R. 24, 673 (1960). <sup>5</sup> H. D. Hagstrum, Phys. Rev. 104, 672 (1956).

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New Jersey. <sup>1</sup> M. A. Herlin and S. C. Brown, Phys. Rev. 74, 291 (1948).

<sup>&</sup>lt;sup>2</sup> M. P. Madan, E. I. Gordon, S. J. Buchsbaum, and S. C. Brown,

Phys. Rev. 106, 839 (1957).

The solution to the rate of change of electron density, n, equation during the formative time of the discharge is given by<sup>8</sup>

$$\ln \frac{(n_b/n_0)}{p} = \frac{\nu}{p} - \frac{Dp}{(p\Lambda)^2},$$
 (1)

where p is the gas pressure, D is the electron diffusion coefficient, and  $\Lambda$  is the characteristic diffusion length of the discharge chamber.

The solution of the rate-of-change equation of the electron density during the decay period (time between pulses) is given by

$$\ln \frac{n(T')}{n_f} = -\int_0^{T'} \beta(T) dT, \qquad (2)$$

where T=0 occurs at the end of the microwave pulse (i.e., the beginning of the decay period),  $n_f$  is the final electron density that is attained at time T=0, and  $\beta(T)$  is a general decay constant which includes recombination, attachment and diffusion. After the initial decay period during which the recombination process may play an important role, the value of  $\beta$  should be constant with time and is given by

$$\beta/p = \nu_a/p + D'p/(p\Lambda)^2, \qquad (3)$$

where  $\nu_a$  is the frequency of electron attachment to neutral molecules and D' is the electron diffusion coefficient during the decay period. From the experimental work of Biondi<sup>4</sup> and Sexton et al.<sup>5</sup> on the rate of decay of the electron density in the microwave afterglow of oxygen, we can obtain a value for  $\beta$ . Specifically, Sexton *et al.* gives an average value of  $\beta/p$  of 69.1 (sec mm Hg)<sup>-1</sup> for a pressure range of 5 to 20 mm Hg, providing the diffusion loss is negligible.

#### 2. METHOD OF DETERMINING v/p

Let T' be the time between pulses, then  $n(T') = n_0$ . Substituting this into Eq. (2) and combining the results with Eq. (1) we obtain the relation

$$\ln\left(\frac{n_b}{n_f}\right) + \int_0^{T'} \beta(T) dT = p\tau \left[\frac{\nu}{p} - \frac{Dp}{(p\Lambda)^2}\right].$$
(4)

Differentiating Eq. (4) with respect to time T', we obtain

$$\frac{d}{dT'}\ln\left(\frac{n_b}{n_f}\right) + \beta(T') = p\frac{d\tau}{dT'}\left[\frac{\nu}{p} - \frac{Dp}{(p\Lambda)^2}\right] + p\tau\frac{d}{dT'}\left[\frac{\nu}{p} - \frac{Dp}{(p\Lambda)^2}\right].$$
 (5)

If we assume that the electron density always attains the same equilibrium value  $n_f$  for a given power level (i.e., assuming that small changes in the formative time due to changes in the repetition rate do not affect the equilibrium density), then the first term on the left is zero. Further consideration on this point is given in the Appendix. The last term on the right-hand side of Eq. (5) is controlled solely by the net electron production parameters. By selecting the discharge conditions so that the electron density remains at a relatively large value, then the parameter D will be the ambipolar diffusion coefficient and should remain constant with time. Hence, Eq. (5) reduces to

$$\frac{\nu}{p} = \frac{\beta}{p} \frac{1}{d\tau/dT} + \frac{Dp}{(p\Lambda)^2}.$$
 (6)

By selecting a suitable chamber geometry and gas pressure range so that the diffusion term is negligible, Eq. (6) reduces to

$$\frac{\nu}{p} = \frac{\beta}{p} \frac{1}{d\tau/dT} = \frac{69.1}{d\tau/dT} \text{ (sec mm Hg)}^{-1}.$$
(7)

The value of  $d\tau/dT$  may be obtained from a series of plots of the formative time of the discharge vs the reciprocal of the repetition rate of the microwave pulses at a constant energy per pulse. From a knowledge of the length and shape of the microwave pulse and the repetition rate, the microwave power can be expressed in terms of an effective electric  $E_e$ . One can then obtain a plot of the net frequency of ionization  $\nu/p$  as a function of  $E_e/p$ . The effective electric field  $E_e$  is given by the relation<sup>1</sup>

$$E_s^2 = E_{\rm rms}^2 \frac{\nu_c^2}{\nu_c^2 + \omega^2},$$

where  $E_{\rm rms}$  is the root mean square of the applied microwave electric field of angular frequency  $\omega$ . The value of  $3.5 \times 10^{9} p$  has been used for  $\nu_{c}$ .<sup>6</sup>

For comparison purposes the value of  $\nu/p$  may be determined from existing data evaluated under dc electric field conditions,<sup>6</sup> that is,  $\nu = (\alpha - \eta)v_d$ , where  $\alpha$  is the first Townsend ionization coefficient,  $\eta$  is the attachment coefficient, and  $v_d$  is the electron drift velocity. The values of  $(\alpha - \eta)$  obtained by Harrison and Geballe<sup>7</sup> and the values of  $v_d$  obtained by Bröse<sup>8</sup> have been used to determine the comparison value of  $\nu/p$ .

From the results of Sexton et al.<sup>5</sup> the diffusion losses in the afterglow may be neglected providing the value of  $p\Lambda$  is greater than 1.5 cm mm Hg. The discharge chamber in this experiment consists of a length of standard X-band waveguide whose effective character-

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<sup>&</sup>lt;sup>8</sup> J. M. Anderson and L. Goldstein, Phys. Rev. 100, 1037 (1955).

 <sup>&</sup>lt;sup>4</sup> M. A. Biondi, Phys. Rev. 84, 1072 (1951).
<sup>6</sup> M. C. Sexton, M. J. Mulcahy, and J. J. Lennon, in *Proceedings* of the Fourth International Conference on Ionization Phenomena in Gases, Uppsala, 1959 (North-Holland Publishing Company, Amsterdam, 1960), Vol. I.

<sup>&</sup>lt;sup>6</sup>S. C. Brown, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1956), Vol. 22, p. 531. <sup>7</sup>M. A. Harrison and R. Geballe, Phys. Rev. 91, 1 (1953). <sup>8</sup>H. L. Bröse, Phil. Mag. 50, 536 (1925).

istic diffusion length is about 0.16 cm or greater.<sup>9</sup> This requires that the gas pressure in the present experiment be 10 mm Hg or greater in order for the diffusion losses in the afterglow to be neglected.

#### 3. APPARATUS

The microwave technique used to produce the discharges is to set up standing waves in the discharge region and to locate the nodes of the standing waves at regions in the system, where discharges can take place with abnormally low fields such as at the microwave window, so that unwanted discharges may be avoided. The discharge chamber is 88 mm long. It is sealed off at one end with a flat brass plate and at the other end with a microwave window. The gas inlet is located in the center of the length of the chamber and on one of the broad faces of the waveguide.

The gas-handling system was designed for another experiment and it was not possible to bake out the discharge chamber. However, a gas discharge was maintained in the discharge chamber for long periods of time before the final data was obtained. Results obtained over a period of several months agreed very well. The justification for using such a scheme is that the results of this experiment were to be applied to a system which could not be baked out. Furthermore, it is felt that since the gas being examined was oxygen, any small quantities of impurities would have little effect. The gas was obtained from a standard gas cylinder with a listed gas purity of 99.5%. (The question of gas purity has been considered by Prasad and Craggs<sup>10</sup> in their measurements of the net ionization coefficient of oxygen.) The discharge chamber was flushed out every 3-4 min during the experiment to avoid the possible accumulation of gas impurities due to by-products from the gas discharge. The system includes a solid CO2 and acetone trap in the gas flow line,



FIG. 1. Schematic diagram of the microwave circuit.



FIG. 2. Formative time of the discharge vs the time between pulses.

and a liquid nitrogen trap near the discharge chamber to remove the water vapor from the gas.

A schematic of the microwave circuit is shown in Fig. 1. The 3.19-cm pulsed power is obtained from a modified APS-4 radar unit.

### 4. MEASUREMENT OF THE FORMATIVE TIME

The required data to determine the value of  $d\tau/dT$ were obtained by measuring the formative time of the discharge as a function of power at repetition rates of 250 to 450 pps (pulses per second), in steps of 50 pps, and at gas pressures of 4.9, 9.8, 15.1 and 19.6 mm Hg (corrected to a temperature of 22°C).

A typical set of experimental results of the formative time of the discharge as a function of the time between pulses, for a constant value of  $E_e/p$  is shown in Fig. 2 from which it can be seen that the slope  $d\tau/dT$  is very uniform and easily evaluated. The completed set of results of  $\nu/p$  vs  $E_e/p$  is shown in Fig. 3; the expected error on the points not marked with error lines is less than 10%. The value of  $\nu/p$  obtained from the dc parameter is plotted on the same graph for comparison purposes.

#### 5. CONCLUSIONS

The experimental results from the microwave experiment agree very well with the results obtained using dc

 <sup>&</sup>lt;sup>9</sup> P. M. Platzman and E. H. Solt, Phys. Rev. 119, 1143 (1960).
<sup>10</sup> A. N. Prasad and J. D. Craggs, Proc. Phys. Soc. (London) 77, 335 (1961).



FIG. 3. Net frequency of ionization  $\nu/p$  vs  $E_e/p$ .

techniques and certain correspondence equations. The described method can be applied to any gas whose primary electron mechanism, in the afterglow of a microwave discharge, is either by diffusion and/or electron attachment; the requirement being that the electron decay rate be exponential and that the decay rate be known.

Plans are being made to repeat this experiment using a microwave cavity which will enable the electron decay rate to be measured in the same experiment.

#### APPENDIX

# Error Involved by Neglecting the Term $(d/dT) \ln(n_b/n_f)$

An estimate of the maximum error involved may be obtained from energy considerations. It is observed experimentally that very little of the incident power is absorbed during the formation of the discharge. We will assume that a portion X of the incident power, P, goes into the production of electrons when the electron density exceeds  $n_b$ . It is also assumed that the final electron density  $n_f$  is determined by the absorbed power and is not limited by recombination, electron attachment or diffusion; therefore these calculations place an upper limit on the error involved. Let L be the length of the microwave pulse, then the value of  $n_f$  is given by

$$n_f = XP(L-\tau)/12.57 \text{ eV}_s$$

where the value 12.57 eV is the ionization energy of molecular oxygen. Evaluating for the required term yields  $(d/dT) \ln(n_b/n_f) = d\tau/dT/(L-\tau)$ . Including this term in the calculations for  $\nu/p$ , neglecting the diffusion losses, we obtain

$$\frac{\nu}{p} = \frac{\beta/p}{d\tau/dT} - \frac{1}{p(L-\tau)}.$$

The term  $1/p(L-\tau)$  represents the result of retaining the expression  $(d/dT) \ln(n_b/n_f)$ . For small values of  $\tau$ and for a pressure of 5 mm of Hg,  $1/p(L-\tau)=10^5$  (sec mm Hg)<sup>-1</sup> compared to the minimum experimental value 10<sup>6</sup> (sec mm Hg)<sup>-1</sup>. Therefore, the maximum error is about 10% at the low values of  $\nu/p$ , and the error decreases as the value of  $\nu/p$  increases. The shape of the light pulses from the discharge region is very flat topped which suggests that the electron density rapidly attains an equilibrium value soon after the discharge is formed. Therefore, small variations in the formative time, due to varying the repetition rate, should have a negligible effect.

## Error Involved by Neglecting the Diffusion Losses during the Formation Period

In order for this diffusion loss to be negligible it is necessary that

$$\frac{Dp}{(p\Lambda)^2} \ll \frac{\beta/p}{d\tau/dT} = \frac{69.1}{d\tau/dT} \text{ (sec mm Hg)}^{-1}.$$

Assuming that D is the ambipolar diffusion coefficient, and using the method given by Kelly and Margenau<sup>11</sup> to evaluate D, yields the value  $Dp=6\times10^4$  mm Hg cm<sup>2</sup>/sec. At a gas pressure of 10 mm Hg the maximum error involved by neglecting the diffusion term is about 4%.

## Increase in the Effective Electric Field due to the Presence of a High Electron Density

Equation (1) is obtained under the assumption that the value of  $\nu/p$  remains constant throughout the formation of the discharge. However, due to the presence of the electrons the effective electric field, and hence the value of  $\nu/p$ , increases as the electron density increases. The effect is partly offset by the reduction in the electric field due to a portion of the incident power being reflected. Calculations indicate that the net effect is a 3% increase of the electric field at the electron density value of  $n_b$ . This small increase has been ignored.

#### Accuracy of Measurements

The recorded data on Fig. 3 are the arithmetic average of three or four sets of results. The calculations for the error in the value of  $\nu/p$  are based on the variation of the formative time of the discharge due to the accuracy with which the microwave power meter and the gas pressure gauge can be read. The listed accuracy of the pressure gauge is 0.25 mm Hg.

<sup>11</sup> D. Kelly and H. Margenau, J. Appl. Phys. 31, 1617 (1960).