

Time-Lag Analysis of the Townsend Discharge in Argon with Activated Caesium Electrodes*

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A study of the time-lag in a photoelectric, gas amplified discharge for parallel electrodes as a function of gas pressure, plate separation, and amplification has demonstrated that diffusion of metastable argon atoms is the source of the lag and that these atoms striking the activated caesium cathode are highly efficient in liberating secondary electrons. The Townsend ionization coefficients, α and γ , have been determined in the usual manner over an extended range of values of E/p_0 . Analysis of the time-lag function has made possible the separation of the number of

secondary electrons released at the cathode into those due to positive ions and those produced by metastable argon atoms. A factor representing the fraction of electron energy gained in the field which is used to excite atoms to the metastable level has been determined as a function of E/p_0 . The coefficient of diffusion of metastable argon atoms in argon has been computed and shown to be that expected for a metastable atom having a diameter effectively 1.74 that of the normal atom.

THE TOWNSEND DISCHARGE

ONLY in recent years has the Townsend discharge been studied with suitable gas purity to provide acceptable data on the primary ionization coefficient α . Secondary coefficients, often lumped into a single coefficient, γ , have been determined for a number of cases in which normally the cathode is a pure metal. But interpretation of γ is usually not precise since analysis of the ionization data can neither distinguish between proposed mechanisms nor separate the total γ into its parts when more than one secondary process is operative. The case of argon gas with activated caesium electrodes has been considered by W. S. Huxford¹ who shows that the most important considerations for relatively low pressures are secondary electron emission by positive ions and the action of metastable atoms in ionizing impurities or in releasing electrons from the cathode. Since separation of these factors is impossible by analysis of the static Townsend discharge some new means of investigation is necessary.

Such a new approach to the study of the Townsend discharge is suggested by the time of action expected for each of the several mechanisms. The release of a photoelectron has been shown by Lawrence and Beams² to occur in less than 10^{-8}

second after the arrival of the exciting photon. The time of transit for electrons is very short but for positive ions is long enough to be easily detected. This time is determined by the positive ion mobility and has been observed by numerous investigators³⁻⁵ using frequency response methods. But an even greater time-lag is evidenced in commercial types of gas photo-tubes.^{5, 6, 7} Many suggestions have been made as to the probable cause of this lag, but delayed secondary emission by positive ions⁸ and the natural delay time involved in the diffusion of metastable atoms are probably the most important of the proposed hypotheses. Certain impurity gases are known to destroy metastable states of argon; Campbell and Rivlin⁹ have shown that the introduction of hydrogen decreases the amount of time-lag in the discharge. Similarly, a significant decrease in this lag by the introduction of argon into neon has been obtained by Kruithof⁵ and taken as a strong argument in favor of the metastable atom hypothesis.

If metastable atoms are responsible for the large time-lag, the lag should increase with gas pressure and with length of the discharge path,

³ A. Roggendorf, *Physik. Zeits.* **36**, 660 (1935).

⁴ A. M. Skellett, *J. App. Phys.* **9**, 631 (1938).

⁵ A. A. Kruithof, *Philips Tech. Rev.* **4**, 48 (1939).

⁶ N. R. Campbell, H. R. Noble, and L. G. Stoodley, *Phys. Soc. Proc.* **48**, 589 (1936).

⁷ F. Schroter and G. Lubszynski, *Physik. Zeits.* **31**, 897 (1930).

⁸ K. H. Kingdon and H. E. Thompson, *Physics* (now *J. App. Phys.*) **1**, 343 (1931).

⁹ N. R. Campbell and R. S. Rivlin, *Phys. Soc. Proc.* **49**, 12 (1937).

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¹ W. S. Huxford, *Phys. Rev.* **55**, 754 (1939).

² E. O. Lawrence and J. W. Beams, *Phys. Rev.* **32**, 478 (1928).

predictions which are contrary to those based on the hypothesis of delayed action of positive ions. Tests of the variation of time-lag with pressure have been unconvincing. Campbell, Noble, and Stoodley⁶ have reported no appreciable variation with length of the discharge path. Their experiment, however, was handicapped by a narrow tube and small electrodes, a combination which does not afford a critical test. Therefore it was felt that a more thorough examination of the contrasting predictions was necessary.

In order to investigate the time-lag phenomenon more carefully, a tube with parallel plates was constructed in which gas pressure and plate separation could be varied. A critical investigation was undertaken with a new method recently developed in this laboratory¹⁰ and particularly suited to direct measurement of the lagging current response to a periodically interrupted light beam. Combining the results of this study and the measured static coefficients for argon for the same tube, it was possible to separate the secondary mechanisms, and thus to obtain a complete analysis of the factors which contribute to gas-amplification in tubes having compound photo-cathodes.

THEORY OF ACTION OF METASTABLE ATOMS IN THE DISCHARGE

Results of the experiment show that the diffusion of metastable atoms to the cathode is the source of the large time-lags observed; further, that the release of secondary electrons at the cathode by metastable atoms contributes substantially to the current amplification. An analysis of these effects is presented below for use in evaluation of the experimental data.

Amplification of the photoelectric current is accomplished by the ionization of the gas together with secondary effects at the cathode. Of these, the release of γ_p secondary electrons by each positive ion impact at the cathode is assumed instantaneous. A delayed emission is caused when γ_m secondary electrons per positive ion are released by *metastable atoms* diffusing to the cathode. We set

$$\gamma = \gamma_p + \gamma_m, \quad (1)$$

¹⁰ W. S. Huxford and R. W. Engstrom, Rev. Sci. Inst. **8**, 385 (1937).

where γ is defined by the amplification equation upon which analysis of the Townsend discharge is based:

$$n_a = \frac{n_0 e^{\alpha(d-d_0)}}{1 - \gamma(e^{\alpha(d-d_0)} - 1)}. \quad (2)$$

Here n_0 is the number of photoelectrons emitted per second, n_a is the equilibrium electron current collected at the anode, α is the number of electrons produced by each electron in traveling 1 cm in the direction of the electric field, d is the plate separation, and d_0 is the distance an electron moves in the direction of the field before gaining sufficient energy for ionization of an argon atom.

In order to obtain a more direct insight into the part played by metastable atoms as symbolized by the term γ_m , consider α_m to represent the number of metastable atoms produced per cm per electron. Since the number of positive ions created per cm per electron is α , the ratio of the number of metastable atoms produced per cm to the number of positive ions produced in the same cm is a constant, α_m/α . For simplification one may approximate the critical distance for onset of excitation of the metastable level by the corresponding distance for ionization, d_0 . (Actually the ionization energy is 15.7 electron volts; the excitation energies, 11.5 and 11.7 electron volts.) It follows then that the ratio of the total number of metastable atoms created per second throughout the entire discharge to the number of positive ions created per second in the same discharge is again as above, α_m/α . However, of all the metastable atoms created in the discharge, only a fraction, f , determined by geometrical considerations, diffuse to the cathode. If each of those reaching the cathode liberates an average of ϵ electrons, the number of secondary electrons emitted from the cathode due to metastable atoms, per positive ion created, is

$$\gamma_m = \epsilon f \alpha_m / \alpha. \quad (3)$$

The ionization equation may now be written

$$n_a = n_0 e^{\alpha(d-d_0)} / [1 - (\gamma_p + \epsilon f \alpha_m / \alpha)(e^{\alpha(d-d_0)} - 1)]. \quad (4)$$

This equation gives the equilibrium current for the case we have considered, but it does not indicate the build-up time of the discharge.

In order to introduce the element of time it is necessary to consider first the probable delay

time in metastable atoms diffusing to the cathode and second the effect this delay has on the measured current function. For those metastable atoms reaching the cathode by diffusion, it may be shown that the probability of arrival at time t per unit time is closely approximated by the form

$$P(t) = (1/\tau_0)e^{-t/\tau_0}, \quad t \geq 0 \\ = 0, \quad t < 0. \quad (5)$$

τ_0 is taken as the average interval between creation and arrival for those metastable atoms reaching the cathode, $t=0$ denoting the time of creation of the metastable atoms in question.

In order to determine the average arrival time of metastable atoms at the cathode it is necessary to set up the differential equation for the diffusion of the metastable atoms. If infinite plane parallel electrodes are assumed, this equation is

$$\partial m(x, t)/\partial t = D\partial^2 m(x, t)/\partial x^2, \quad (6)$$

where D is the coefficient of diffusion and $m(x, t)$ is the linear density of metastable atoms at a distance x from the cathode, at time t . We assume the spacial distribution of metastable atoms when created to be

$$m(x, 0) = 0, \quad x < x_m, \\ m(x, 0) \propto e^{\alpha(x-x_m)}, \quad x_m \leq x \leq d, \quad (7)$$

where x_m is the distance which an electron must traverse in the direction of the field to obtain the energy of excitation of the metastable state. A further assumption is that any metastable atom contacting either electrode loses its metastable energy. With these conditions Eq. (6) may be solved for $m(x, t)$ thus giving the distribution of metastable atoms at any time after creation. Of these, the number arriving per second at the cathode is

$$D[\partial m(x, t)/\partial x]_{x=0}.$$

The average arrival time may be obtained as follows:

$$\tau_0 = \frac{\int_0^\infty t D[\partial m(x, t)/\partial x]_{x=0} dt}{\int_0^\infty D[\partial m(x, t)/\partial x]_{x=0} dt} \quad (8)$$

Carrying out this operation, we find that τ_0 may

be expressed as a series involving constants of the discharge:

$$\tau_0 = (d^2/12D) \left\{ 1 + (2/15)\alpha d + (1/180)\alpha^2 d^2 + \dots \right. \\ \left. + (x_m/d) [2 - (2/5)\alpha d - (1/45)\alpha^2 d^2 + \dots] \right. \\ \left. + (x_m^2/d^2) [-1 + (2/5)\alpha d + (1/36)\alpha^2 d^2 + \dots] \right. \\ \left. + \dots \right\}. \quad (9)$$

Knowing the probable time of delay in the arrival of metastable atoms in form [Eq. (5)] and in magnitude [Eq. (9)], we can now calculate its retarding effect on the measured current. Thus, it may be shown¹¹ that the form of the

¹¹ In order to do this and retain generality so that the method may be applicable to similar discharge problems, we proceed as follows: Let R represent the number of secondary electrons immediately produced at the cathode as a result of one primary electron leaving the cathode (R includes also the secondary emission by positive ions whose transit time is assumed to be negligible). Let L represent the delayed production of electrons per electron leaving the cathode (including those produced at the cathode by impact of metastable atoms whose diffusion time is the source of the observed time-lag). In the present case it follows that

$$R = \gamma_p(e^{\alpha(d-d_0)} - 1), \\ L = \gamma_m(e^{\alpha(d-d_0)} - 1).$$

If $n(t)$ represents the number of electrons leaving the cathode at time t , it must follow that

$$n(t) = n_0(t) + Rn(t) + L \int_0^\infty P(\tau)n(t-\tau)d\tau, \quad (A)$$

where τ represents the time variable of integration. A fundamental assumption of this equation is that the effect produced by an individual electron leaving the cathode is in no way a function of the immediate or previous current in the discharge. In this experiment the use of low current densities insured the absence of appreciable space charge. Small changes in emissivity of the cathode were found when the bombarding current was varied, as indicated in a later section. Equation A is supplemented, of course, by the normalization requirement:

$$\int_0^\infty P(t)dt = 1.$$

Consider two cases corresponding to the experimental sequence in measuring the lagging current: the *rise*, where the light comes on and the current increases from zero to equilibrium; the *fall*, beginning with cessation of illumination and including the dying away of the current. Case I: The *Rise*. Take $n_0(t) = 0, t < 0; n_0(t) = 1, t \geq 0$. It follows that $n(t) \equiv 0, t < 0$. Therefore, indicating the *rise* solution by subscript r , one obtains

$$(1-R)n_r(t) - L \int_0^t P(\tau)n_r(t-\tau)d\tau = 1, \quad t > 0, \quad (B)$$

the general integral equation for the rising current. Case II: The *Fall*. Add the stipulation, $n_0(t) = 0, t > T$, where T is the duration of the illumination. It follows for $t > T$, subscript f denoting the *fall* solution, that

$$(1-R)n_f(t-T) - L \int_0^{t-T} P(\tau)n_f(t-T-\tau)d\tau \\ - L \int_{t-T}^t P(\tau)n_r(t-\tau)d\tau = 0, \quad (C)$$

where $n_r(t-T)$ has been written so that $n_f(0)$ corresponds

lagging current response to a constant photo-current initiated at time $t=0$ is

$$i_{\text{rise}}(t) = 1 - (1 - i_a)e^{-(i_a/\tau_0)t}, \quad (10)$$

where the maximum equilibrium current is referred to as unity. Further, if the photoelectric source is cut off at time $t=T$ ($t'=0$), when the rising current lacked saturation by an amount i_c , the falling characteristic may be represented by the equation:

$$i_{\text{fall}}(t') = (1 - i_b - i_c)e^{-(i_b/\tau_0)t'}. \quad (11)$$

In these equations, i_a and i_b represent the fraction of nonlagging current for the *rise* and for the *fall* characteristic, respectively; it can be shown that $i_a = i_b$. Eqs. (10) and (11) may now be used in the analysis and interpretation of the time lag data.

Since it is experimentally possible to measure the exponential decay constant, τ ($\tau = \tau_0/i_a$), and the nonlagging fraction of the current i_a , one may evaluate the fundamental time constant, τ_0 . The variation of τ_0 may be studied as a function of distance and pressure as predicted by Eq. (9) wherein D varies inversely as the pressure. One may also determine D_0 , the coefficient of diffusion of metastable argon atoms in normal argon for standard temperature and pressure.

Having α from static ionization measurements, we can calculate γ for each time-lag curve since the current amplification is known to be

$$A = n_a/n_0 = e^{\alpha(d-d_0)} / [1 - \gamma(e^{\alpha(d-d_0)} - 1)]. \quad (12)$$

to the beginning of the *fall* current at $t=T$. Throughout this discussion $n_r(t)$ is considered to maintain its form independent of the condition imposed by interruption of the light. By direct substitution in Eq. (C) and comparison with Eq. (B) it is easily shown that

$$n_f(t') \equiv n_r(t'+T) - n_r(t'), \quad t' > 0, \quad (D)$$

where $t' = t - T$. Therefore it is only necessary to solve Eq. B for $n_r(t)$ and obtain $n_f(t')$ by Eq. (D). When T is sufficiently large, as may or may not be the case, the solution for the *fall* is simply the *rise* solution inverted with the maximum value as zero. Using $P(t)$ of Eq. (5) in integral Eq. (B), one obtains:

$$n_r(t) = [1/(1-R-L)] [1 - (L/(1-R)) \times \exp\{-[(1-R-L)/(1-R)]t/\tau_0\}]. \quad (E)$$

From Eq. (D):

$$n_f(t') = \frac{L}{1-R-L} \times \left[\frac{1 - \exp\{-[(1-R-L)/(1-R)]T/\tau_0\}}{1-R} \right] \times \exp\{-[(1-R-L)/(1-R)]t'/\tau_0\}. \quad (F)$$

Eqs. (E) and (F) represent the form of the time-lagging current for the *rise* and for the *fall*, respectively. From these two it is only necessary to make the proper interpretations of symbols to obtain forms (10) and (11).

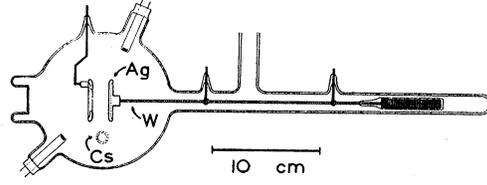


FIG. 1. The experimental tube, side view, showing arrangement for adjusting the plate separation of the plane parallel electrodes. A side arm is indicated which contained encapsulated metallic caesium for activation of the electrodes.

In addition, having determined the amount of nonlagging current, one may find γ_p from the fact that the instantaneous amplification is independent of the metastable atom contribution in the term γ_m . Thus,

$$i_a A = \frac{e^{\alpha(d-d_0)}}{1 - \gamma_p(e^{\alpha(d-d_0)} - 1)}. \quad (13)$$

Having both γ and γ_p , one may now determine $\gamma_m = \gamma - \gamma_p$. Furthermore, since f may be calculated from the geometry of the tube and a knowledge of the particular values of α , d , and d_0 , one has a measure of $\epsilon \cdot \alpha_m$ [Eq. (3)], and, since ϵ is independent of the field strength, a function proportional to α_m . However, it is more convenient to discuss a number proportional to the fractional electron energy being delivered to excitation of metastable states. Therefore, write

$$\epsilon \cdot (\alpha_m/E) = (\alpha/E)(\gamma_m/f), \quad (14)$$

where E is the magnitude of the electric field. Comparing this function with the maximum excitation possible if all the energy available to the electron in the field were used for this purpose [$(\alpha_m/E)_{\text{max}} = 1/V_m$, where V_m is the critical energy of excitation (11.5 and 11.7 electron volts for argon)], we can obtain an estimate at least of the lower limit of ϵ .

Thus, an analysis of the time-lag current function, while significant as a means of testing the metastable atom theory of the lag, also allows a separation of the secondary emission into positive ion and metastable atom sources, provides a measure of the fraction of electron energy used to excite atoms to the metastable state, and permits an estimate of the efficiency of liberation of electrons at the cathode by metastable atom contact.

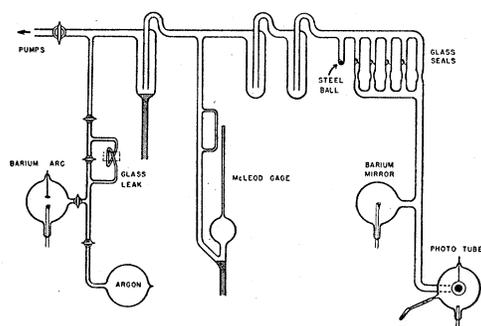


FIG. 2. The vacuum system including the experimental photo-tube, end view.

EXPERIMENTAL PROCEDURE

The essential parts of the tube are the two parallel electrodes. These were made of cast silver, 4 cm in diameter, turned into shape with an approximate Rogowski contour,¹² polished to plane surfaces and supported by tungsten rods as shown in Fig. 1. A circular hole 2 cm in diameter was cut in the anode and across it a mesh was woven flush with the flat plate. The mesh was made of 0.005-inch silver wire spaced at 1-mm intervals; its transparency to light was about 80 percent. The cathode was mounted axially on a tungsten rod supported by nickel eyelets. At the other end of the tungsten rod an iron bar was welded and enveloped in glass. This arrangement provided external magnetic control of the position of the cathode, separation of the plates being measured within an error of approximately 0.003 cm. The plates were parallel to within an error of 0.5° of arc. For plate separations up to a maximum of 1.5 cm the spacing of electrodes and side walls was such as to prevent wall charges from distorting the electric field in the region of the discharge. Light from an incandescent filament was admitted through the anode mesh to the cathode by means of a plane Corex window in one end of the Pyrex envelope. Tungsten filaments were provided for degassing the metal parts by electron bombardment.

The vacuum system of Fig. 2 shows the arrangement used for admitting argon to the photo-tube. Spiral tungsten filaments were used to hold pieces of barium metal in both the arc chamber and in the barium mirror tube. The arc discharge

in argon was controlled by carefully adjusting the temperature of the hot tungsten-barium cathode and was run many hours in the purification of the gas. Satisfactory purity of the argon was judged not only by the absence of impurity lines in the spectrum but also by the observation of no change in photo-sensitivity of the caesium surface when the gas entered the photo-tube. After the gas had been slowly admitted to the tube chamber and the pressure measured, one of the series of glass seals was closed by means of a small flame so that photo-tube and barium mirror were completely isolated from the rest of the system. When the measurements at one pressure were completed the fore-system was baked and evacuated, one of the inner seals broken by means of a magnetically controlled steel ball, and the entire purification and filling process repeated. If small amounts of gas impurities were released in closing the seals they were effectively removed by the barium mirror surface in the side tube, since no change of photo-sensitivity was observed during the series of measurements at four different pressures.¹³ The processing of the caesium-oxide surface was carried out prior to admission of argon. The cathode had the usual sensitivity of commercial photo-tubes with a threshold at about 10,000A.

MEASURING CIRCUITS

The method used in measuring the time variation in the photo-tube has been described in a previous paper.¹⁰ The circuit shown in Fig. 3, with switches in the *solid line* positions, is an improved arrangement having a current sensitivity some ten times that of the earlier circuit. Light from a tungsten filament entering the photo-tube was interrupted by means of a 45° sector in a disk driven by synchronous motor M_1 . Instantaneous values of current were measured by potentiometer, P , by a null method using a highly insulated electrometer tube, T_3 . The limit of accuracy of the readings was 0.0001 volt, corresponding to a current sensitivity of 5×10^{-10}

¹² W. Rogowski and H. Rengier, *Archiv f. Elektrot.* **16**, 73 (1926).

¹³ Experience shows that great care must be exercised in degassing the metal parts and in applying a flame in sealing off the photo-tube if contaminations arising from chemically active gases and vapors are to be avoided. We are indebted to Professor Robert J. Cashman for valuable suggestions concerning vacuum technique and use of the barium mirror surface for removing impurity gases and vapors.

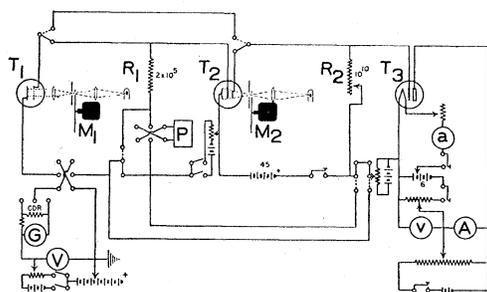


FIG. 3. Electrical circuit. T_1 , the experimental tube; T_2 , photoelectric triode; T_3 , a three-element electrometer tube; M_1 and M_2 , synchronous motors; P , potentiometer. The time-lag measuring circuit is shown with switches in the *solid line* position; the ionization measuring circuit with switches in the *dotted line* position.

amp. The arrangement shown with the switches in the *dotted line* positions, eliminating the photoelectric triode, T_2 , from the circuit, was used for measuring static ionization currents.

IONIZATION COEFFICIENTS

The usual method for obtaining the Townsend ionization coefficients requires a knowledge of the current as a function of plate separation at constant field. However, fatigue effects common to the type of surface used made it expedient to measure current-voltage characteristics at a number of plate separations and from these to obtain current-distance data by interpolation. Effects of back diffusion of electrons, of change of photo-sensitivity with field, and of variation of scattered light with plate separation were accurately measured and corrections suitably applied to the data. In these measurements the density of the initiating photo-current was of the order of 1×10^{-10} ampere cm^{-2} ; smaller currents were not feasible because of the magnitude of the thermionic emission at room temperature.

Data were obtained for four pressures, p_0 (reduced to a temperature of 0°C) = 0.0983, 0.1933, 0.407 and 0.815 mm of Hg. In Fig. 4, the results are shown in the form α/p_0 plotted as a function of E/p_0 , with the curve of Kruithof and Penning¹⁴ for argon dotted in for comparison. Two tendencies are observed: for successively lower pressures the data are lower; for the higher pressures the points are high with reference to the Kruithof and Penning curve. Accuracy of

¹⁴ A. A. Kruithof and F. M. Penning, *Physica* **3**, 515 (1936).

analysis decreases with decrease in pressure; for the lowest pressure the inaccuracy is so large that the points are of little value; (this is again apparent in the trend of the secondary coefficient, γ). However it is believed that the downward trend of the points for lower pressures was the unfortunate result of using an open mesh which undoubtedly distorted the field besides allowing penetration of electrons and possibly ionization behind the anode. In general the trend of the data agrees with that of Kruithof and Penning. It is possible that the apparently larger α/p_0 values are the result of ionization of caesium vapor which may be present in the gas. For very large values of E/p_0 there is no tendency to reach a maximum value of α/p_0 as was observed by Huxford.¹

Values of the secondary coefficient γ obtained from the ionization analysis are shown in Fig. 5 (solid points). The order of magnitude agrees very well with that reported by Huxford¹ for similar cathodes. The data for the three highest pressures are fairly consistent but those for the lowest show a divergence, as anticipated. The solid line has been drawn disregarding the points for the lowest pressure used.

TIME-LAG DATA

It was not possible to measure the trend of the current with time for currents of the low order of magnitude used in the measurement of the ionization coefficients. However, the maximum

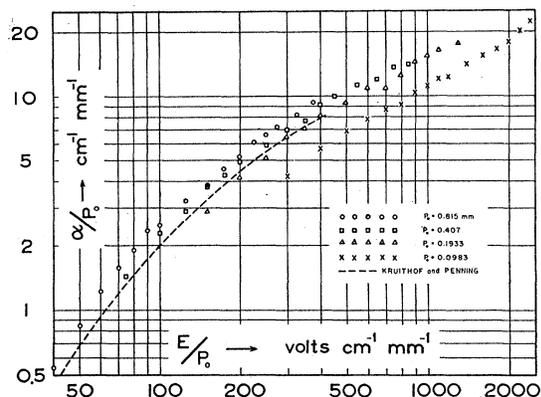


FIG. 4. Results for the first Townsend coefficient, for argon, compared with those of Kruithof and Penning. The errors were so large in the determinations at the lowest pressure (crosses) that these values are disregarded in the analysis.

gas amplified current densities used never exceeded 1×10^{-7} ampere cm^{-2} . When currents of the order 10 to 100 times this were used serious distortion of the time-lag function resulted as judged by the asymmetry of rising and falling current functions. Even for the currents used there was some evidence of asymmetry, but in general the distortion was negligible.

Figure 6 shows a characteristic time-lag curve at a pressure of 0.815 mm of Hg as compared with the current observed for a discharge in vacuum. Since the photoelectric response is instantaneous² the curve for a vacuum represents quite accurately the illumination function. From the time-lag currents plotted on a logarithmic current scale, Fig. 7, we see that the currents are indeed exponential as predicted. Measurement of the slope allowed a determination of the time constant τ . The values of i_a , i_b , and i_c were also obtained from the logarithmic plot as indicated.

Table I shows the tabulated constants for the curves analyzed. A comparison of $(\tau)_r$ and $(\tau)_f$ as well as i_a and i_b shows fairly good agreement between the rising and falling characteristics, confirming the assumption that the effects produced by an individual electron leaving the cathode are independent of the immediate or previous discharge current. However, for the lowest pressure, besides the regular time-lag attributable to metastable atoms, a very much longer time-lag was found. This lagging current had a time constant of the order 0.01 sec. and amounted in some cases to a few percent of the

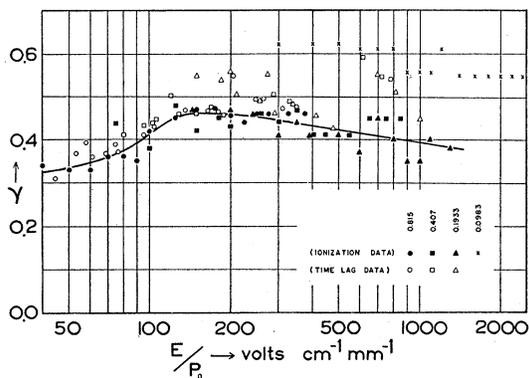


FIG. 5. Values of the second Townsend coefficient, γ vs. E/p_0 for the ionization data (solid points), and for the time-lag data (open points). The points for the lowest pressure (crosses) were disregarded in drawing the line through the solid points.

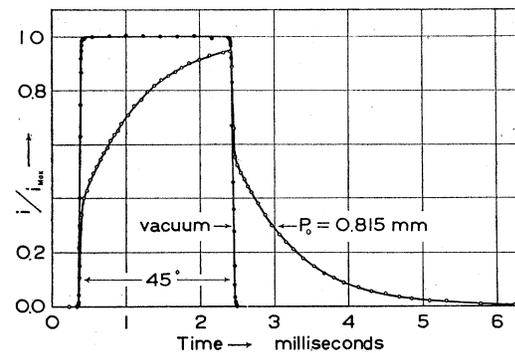


FIG. 6. Samples of the time-lag curves as measured, i/i_{\max} vs. time in milliseconds, for vacuum and for argon with $p_0=0.815$ mm of Hg, $d=0.399$ cm, $V=57$ volts, current amplification, 29.6. The period of illumination was 45° a.c. cycle as indicated.

total current. The probable cause is a slow change in the cathode surface with bombarding current. In all cases where this lag appeared it was subtracted from the main data which could then be satisfactorily analyzed in terms of the action of metastable atoms.

The values of τ show a very decided increase with current amplification, but the fundamental constant, τ_0 , shows very little trend. Values of τ_0 for the rise and for the fall are given in Table I for the separate corresponding values of i_a and i_b . The data for the falling current are more accurate since measurement of the time constant was direct and did not involve inverting the curve about the maximum value.

Since the variation of τ_0 with amplification is small and uncertain, it was disregarded and an average value, obtained by weighting according to accuracy of measurement at a given distance and pressure, was used. The variation of these average time constants with pressure and distance furnishes the major confirmation of the metastable atom theory of the time-lag. A weighting equivalent to that of the time constants applied to the averaging of the constants of Eq. (9) yielded values of $(\alpha \cdot d)_{av} = 1.25$, and $(x_m/d)_{av} = 0.20$. Substituting these average values in Eq. (9) and dropping insignificant terms, we approximate

$$\tau_0 = 1.36d^2/12D = k \cdot p_0 \cdot d^2, \quad (15)$$

where

$$k = 1.36/12 \cdot D_1 \quad (16)$$

and D_1 is numerically equal to the coefficient of

TABLE I. Tabulation of constants for the time-lag curves.

p_0	d	V	A	i_a	i_b	i_c	$(\tau)_r$	$(\tau)_f$	$(\tau_0)_r$	$(\tau_0)_f$
0.815	0.210	25.0	1.64	0.940	0.950	0.000	0.137	0.201	0.129	0.191
		35.0	2.57	0.877	0.896	0.000	0.124	0.141	0.109	0.126
		45.0	4.90	0.826	0.836	0.000	0.152	0.159	0.125	0.133
		53.0	10.1	0.63	0.692	0.000	0.137	0.170	0.086	0.118
		59.0	38.2	0.31	0.408	0.000	0.269	0.244	0.083	0.099
	0.399	35.0	2.32	0.875	0.877	0.001	0.402	0.419	0.352	0.367
		45.0	4.45	0.762	0.787	0.001	0.426	0.444	0.325	0.349
		53.0	10.7	0.585	0.624	0.009	0.551	0.559	0.322	0.348
		57.0	25.4	0.361	0.382	0.048	0.800	0.815	0.289	0.311
	0.715	30.0	1.44	0.955	0.917	0.009	1.27*	1.19	1.21*	1.09
		43.0	2.73	0.819	0.807	0.037	1.20*	1.20	0.98*	0.97
		55.0	7.20	0.624	0.598	0.099	1.40	1.51	0.873	0.903
		60.0	16.3	0.421	0.422	0.220	2.16*	2.18	0.910*	0.920
	1.107	45.0	2.08	0.851	0.833	0.064	2.46	2.41	2.09	2.00
		55.0	3.74	0.728	0.717	0.130	2.84*	2.88	2.06*	2.06
		62.0	7.00	0.594	0.585	0.210	3.18*	3.28	1.89*	1.92
		68.0	19.5	0.360	0.348	0.412	5.24*	5.19	1.88*	1.80
	1.604	45.0	1.53	0.897	0.855	0.091	—	4.20	—	3.59
		58.0	2.57	0.780	0.760	0.151	5.8	4.43	4.5	3.36
		68.0	5.10	0.627	0.633	0.252	5.6	5.60	3.5	3.54
		75.0	13.2	0.420	0.434	0.429	7.9	7.30	3.3	3.16
0.407	0.221	35.0	2.31	0.948	0.958	0.000	0.24	0.155	0.23*	0.148
		55.0	6.31	0.67	0.87	0.000	0.105	0.131	0.070	0.114
		65.0	12.1	0.55	0.83	0.000	0.167	0.103	0.092	0.085
		70.0	20.2	0.37	0.85	0.000	0.193	0.211	0.071	0.179
	0.450	30.0	1.78	0.907	0.923	0.000	0.232	0.272	0.210	0.251
		45.0	4.00	0.824	0.845	0.000	0.274	0.298	0.226	0.252
		53.0	7.21	0.718	0.798	0.001	0.328	0.324	0.236	0.258
		60.0	15.9	0.519	0.664	0.002	0.382	0.360	0.198	0.228
	0.822	40.0	2.71	0.850	0.854	0.009	0.769	0.780	0.652	0.665
		50.0	5.12	0.732	0.762	0.021	0.835	0.867	0.610	0.660
		55.0	8.65	0.628	0.646	0.047	1.00	0.990	0.628	0.640
		62.0	35.4	0.328	0.347	0.242	2.1	1.74	0.67	0.604
	1.157	30.0	1.42	0.916	0.921	0.022	1.6	1.37	1.5	1.26
		40.0	2.29	0.842	0.861	0.037	1.5	1.40	1.3	1.21
		50.0	4.24	0.748	0.755	0.068	1.60	1.47	1.20	1.11
		60.0	13.4	0.514	0.514	0.202	2.5	2.08	1.0	1.07
	1.530	35.0	1.52	0.908	0.908	0.037	2.4	2.68	2.2	2.43
		50.0	3.14	0.778	0.780	0.100	2.7	2.54	2.1	1.98
		60.0	7.42	0.601	0.604	0.205	3.4	3.06	2.0	1.85
		65.0	16.6	0.435	0.442	0.332	4.7	4.04	2.0	1.78
0.1933	0.194	25.0	1.47	0.924	0.948	0.000	0.230	0.201	0.212	0.190
		40.0	2.31	0.89	0.86	0.000	0.161	0.167	0.143	0.144
		65.0	4.69	0.67	0.81	0.000	0.147	0.175	0.098	0.142
		80.0	8.72	0.61	0.75	0.000	0.208	0.314	0.127	0.236
	0.443	40.0	2.41	0.889	0.901	0.000	0.224	0.167	0.199	0.150
		60.0	4.85	0.75	0.85	0.000	0.165	0.191	0.124	0.162
		70.0	7.28	0.65	0.81	0.000	0.145	0.226	0.094	0.183
		85.0	19.5	0.60	0.70	0.000	0.195	0.258	0.117	0.180
	0.762	40.0	2.54	0.869	0.892	0.000	0.282	0.296	0.245	0.264
		50.0	3.84	0.820	0.853	0.000	0.268	0.322	0.220	0.274
		60.0	6.48	0.754	0.815	0.001	0.370	0.330	0.278	0.269
		70.0	12.2	0.66	0.748	0.002	0.402	0.334	0.265	0.250
	1.167	35.0	1.97	0.917	0.920	0.001	0.390	0.665	0.358	0.612
		45.0	3.18	0.885	0.870	0.006	0.725	0.649	0.640	0.565
		55.0	5.64	0.808	0.819	0.004	0.550	0.716	0.444	0.586
		65.0	14.1	0.64	0.672	0.040	0.960	0.885	0.61	0.595
	1.556	35.0	1.74	0.912	0.920	0.018	1.31	1.13	1.19	1.04
		45.0	2.89	0.857	0.869	0.020	1.07	1.04	0.92	0.90
		55.0	5.64	0.77	0.791	0.048	1.33	1.15	1.02	0.91
		62.0	11.4	0.66	0.65	0.11	1.93	1.38	1.27	0.90

EXPLANATION OF SYMBOLS: p_0 = pressure of argon in mm of Hg reduced to 0°C. d = plate separation in cm.

V = potential in volts applied to the electrodes.

A = current amplification = ratio of the current collected to the initiating photo-current (corrections having been made to account for back diffusion and change in emission with field, so that A may be inserted directly in Eqs. (12) and (13)).

i_a = nonlagging fraction of the current as measured in the rise of the time-lag current function.

i_b = nonlagging fraction of the current as measured in the fall.

i_c = ratio of the difference between the equilibrium current and the current at the end of the light period, to the equilibrium current.

$(\tau)_r$ = 1/eth life of the exponential current for the rise in milliseconds.

$(\tau)_f$ = 1/eth life of the exponential current for the fall in milliseconds.

$(\tau_0)_r$ = fundamental time constant obtained from the rise, in milliseconds, = $i_a \cdot (\tau)_r$, = average time of arrival of metastable atoms at the cathode [see Eq. (10)].

$(\tau_0)_f$ = fundamental time constant obtained from the fall, in milliseconds, = $i_b(\tau)_f$ [see Eq. (11)].

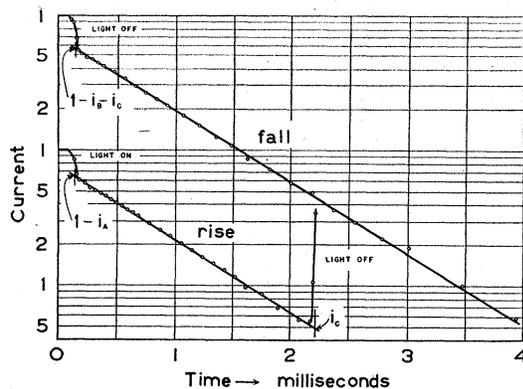


FIG. 7. The time-lag curve of Fig. 6 for argon is here plotted on a logarithmic current scale. The slope of the line determines the time constant. Intercepts of the lines with those lines indicating the time of light on and light off are measures of i_a , i_b , and i_c .

diffusion of metastable argon atoms in argon at 1 mm Hg reduced pressure.

Preliminary graphing indicated that at the lower pressures there is an apparent elongation of the discharge region. This suggests that metastable atoms may diffuse back and forth through the mesh thus allowing more diffusion space than was assumed. Since the transparency of the mesh was about 80 percent, this apparent elongation of the discharge may amount to several free paths. Therefore one may write

$$\tau_0 = k \cdot p_0(d + c\lambda)^2, \quad (17)$$

where λ is the mean free path of a metastable argon atom and c is a constant. Since $\lambda = \lambda_1/p_0$ where λ_1 is numerically the mean free path at 1 mm Hg,

$$\tau_0 = k \cdot p_0(d + c\lambda_1/p_0)^2; \quad (18)$$

and by algebraic manipulation,

$$\tau_0^{1/2} p_0^{1/2} = k^{1/2}(p_0 d + c\lambda_1). \quad (19)$$

If $\tau_0^{1/2} p_0^{1/2}$ is plotted against $p_0 \cdot d$ a straight line of slope $k^{1/2}$ should result regardless of the correction due to the mesh. In addition, by measuring the negative intercept on the $p_0 \cdot d$ axis, the term $c\lambda_1$ may be evaluated.

The averaged data are portrayed graphically as suggested by Eq. (19) in Fig. 8, showing satisfactory conformity to the predicted relationship. Since the data cover a considerable range of pressures and distances we may say that, qualitatively, the theory of release of electrons from the cathode by metastable atoms is confirmed.

Again it may be noted that while there are slight differences in the points obtained from the *rise* and from the *fall*, the agreement is satisfactory.

The data were evaluated by a least squares method to obtain the best values for the slope of the line and for the intercept on the p_0d axis. The following are the values so determined:

$$k^{\frac{1}{2}} = 0.0410 \text{ mm}^{\frac{1}{2}} \text{ sec.}^{\frac{1}{2}} \text{ mm}^{-1} \text{ cm}^{-1},$$

$$c\lambda_1 = 0.047 \text{ mm cm.}$$

Since for argon λ_1 is very nearly 0.0076 mm (of Hg) cm, c is approximately 6 free paths, a reasonable figure. Computing by use of Eq. (16), we find the coefficient of diffusion of metastable argon atoms in argon for $p_0 = 1$ mm Hg at room temperature to be $D_1 = 67.5 \text{ cm}^2 \text{ sec.}^{-1} \text{ mm}$.

From the experimental law,

$$D = D_0(T/273)^n(760/p) = D_0(T/273)^{n-1}(760/p_0)$$

and taking $n = 1.75$,¹⁵ we obtain: $D_0 = 0.084 \text{ cm}^2 \text{ sec.}^{-1}$ at standard temperature and pressure.

There are no experimental values for the coefficient of self-diffusion of argon atoms or for excited argon atoms in argon. However, D_0 may be obtained with fair accuracy from the expression involving the coefficient of viscosity, η , and the density of the gas, ρ , both of which are known. Thus,¹⁶ $D_0 = 1.336\eta/\rho$. Taking the value $\eta = 2196 \times 10^{-7} \text{ dyne sec. cm}^{-2}$,¹⁷ we obtain $D_0 = 0.157$ for argon atoms in argon.

However, we are dealing with excited atoms diffusing in a gas composed of normal atoms. Undoubtedly the kinetic cross section of the excited atoms is larger than that of the normal atoms. Just how much, it is difficult to predict. Coulliette¹⁸ concluded, in an experiment with metastable mercury atoms diffusing into mercury vapor and subsequently striking a nickel hemisphere where secondary electrons were ejected, that the effective diameter of the excited mercury atom is 1.5 normal.

The normal argon state is a $3p^6\ ^1S$ configuration; the excited metastable levels are $3p^5(^2P_{1/2})4s, J=2$; $3p^5(^2P_{3/2})4s, J=0$. The electron involved in each case has moved from a $3p$ to a

$4s$ level. Since the average distance of the electron from the nucleus increases as the square of the principal quantum number, the average diameter would be increased by a factor $\frac{1}{9}^{\frac{1}{2}} = 1.78$. This of course, does not necessarily approximate the kinetic diameter but it gives strong evidence that the diameter is larger.

According to Jeans¹⁹ the coefficient of inter-diffusion varies inversely as the square of the arithmetical mean of the diameters. If we assume the difference in diameters as the explanation of the discrepancy in the experimental and calculated values above, we obtain for the ratio of the excited atom diameter to the normal atom diameter the value 1.74. This very close agreement of the metastable atom diameter as observed and predicted, while fortuitous in view of the various assumptions involved, is significant in that the correction is the right order of magnitude and thus lends strong support to the whole theory of action of metastable atoms in the discharge.

EVALUATION OF SECONDARY MECHANISMS

From the values of α calculated from the ionization data at each pressure and the measured amplification factor, A , γ may be computed for each of the time-lag curves by means of Eq. (12). For low amplifications, the errors were quite large; hence only results for amplifications greater than 2.5 are shown in Fig. 5 (open

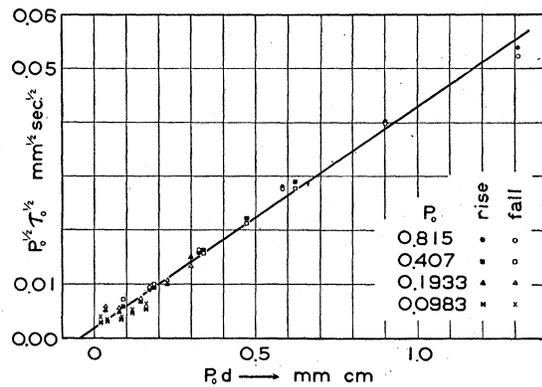


FIG. 8. Experimental verification of Eq. (19) [$\tau_0^{\frac{1}{2}} p_0^{\frac{1}{2}} = k^{\frac{1}{2}}(p_0 d + c\lambda_1)$] which is based on considerations of the diffusion time of metastable argon atoms and their subsequent release of electrons at the cathode.

¹⁵ Int. Crit. Tab. 5, p. 62.

¹⁶ L. B. Loeb, *The Kinetic Theory of Gases* (McGraw-Hill, second edition, 1934), p. 273.

¹⁷ E. H. Kennard, *Kinetic Theory of Gases*, (McGraw-Hill, first edition, 1938), p. 149.

¹⁸ J. H. Coulliette, *Phys. Rev.* **32**, 636 (1928).

¹⁹ J. H. Jeans, *The Dynamic Theory of Gases* (Cambridge University Press, second edition, 1921), p. 332.

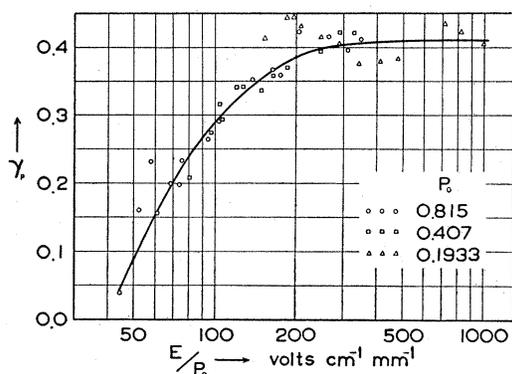


FIG. 9. γ_p , the number of secondary electrons emitted per positive argon ion at the activated caesium cathode, vs. E/p_0 . These values were obtained from the time-lag data by eliminating the contribution of the metastable atoms to the Townsend γ .

points). The trend of γ values so obtained is somewhat higher than that for the ionization data, again indicating increased surface sensitivity for larger bombarding currents.²⁰

Values of γ_p , the secondary coefficient for positive ions may now be determined by the use of Eq. (16). These values, corrected for back diffusion to yield the total emission, are plotted in Fig. 9. It is interesting to note that while the γ values showed no appreciable decrease for low E/p_0 , the values of γ_p do show this trend with a tendency to level out at higher E/p_0 values. The difference between γ and γ_p gives an indication of the contribution of metastable atoms to the discharge. This is seen to be most important in the lower range of E/p_0 .

Now, following Eq. (14), it is possible to calculate $\epsilon\alpha_m/E$ which is proportional to the fractional energy being used in the excitation of metastable atoms. The curve, Fig. 10, showing $\epsilon\alpha_m/E$ as a function of E/p_0 , was drawn to fit the more reliable points. By comparing this curve with the energy balance curves of Penning²¹ and with the maximum possible value of $\alpha_m/E = 1/V_m = 0.086$, it has been determined that the *minimum efficiency* for the production of secondary electrons by metastable argon atoms from an activated caesium surface is $\epsilon = 0.4$ secondary electron per metastable atom.

²⁰ Increased gas amplification and photo-sensitivity of the cathode with rise of current has previously been noted. See reference 1, p. 756.

²¹ F. M. Penning, *Physica* **5**, 286 (1938).

OTHER EVIDENCE

Time-lag curves obtained from measurements made with the caesium-activated, mesh-centered electrode as cathode show that the order of the time-delay was the same as for the solid cathode, but the fraction of lag current was less. Analysis shows nearly the same γ_p but considerably smaller γ_m . One possible explanation is that the surfaces differ in activation, but the obvious interpretation is that many of the metastable atoms pass through the mesh cathode and fail to contribute to the discharge.

In connection with work already reported on ionization coefficients for argon with a pure barium cathode,²² time-lag measurements were also taken. There was no measurable lag of the order found in the present experiment with caesium. At a pressure of 1.51 mm Hg, a plate separation of 2.96 cm, an applied potential of 186 volts, and a gas amplification of 18.9, the 1/eth life of the barely observable lag was approximately 7×10^{-5} second which agrees very closely with the time of transit of positive ions. Since metastable atoms certainly are present, the obvious conclusion is that the efficiency of release of electrons by metastable atoms, ϵ , is very small for pure barium.

After the main data had been taken with the barium tube, the surface was contaminated with hydrogen and an attempt was made to produce an activated surface having a higher sensitivity. Although the increased photo-sensitivity was not of the magnitude found in the case of the caesium surface, there was considerable increase in photo-emission and the surface no doubt had a complex structure. Time-lag measurements were again taken and at a pressure of 0.219 mm, a plate separation of 0.24 cm, a potential of 238 volts and an amplification of about 49, the 1/eth life was 0.17 millisecond, the fraction of the lagging current being about 62 percent. Calculating, we find $\tau_0 = 0.067$ millisecond, which is in good agreement with the value 0.076 millisecond, computed from the assumption of metastable atom action. It would seem that activation of the barium surface greatly increased the efficiency of release of electrons by metastable atoms. Undoubtedly many of the conflicting results of

²² R. W. Engstrom, *Phys. Rev.* **55**, 239(A) (1939).

previous investigations of the efficiency of metastable atoms in releasing electrons are due to wide variations in surface contamination of the electrodes used.

Interesting in this connection is the experiment of Skellett⁴ in which the frequency response of photo-currents was measured in an argon-filled spherical tube having a carbon cathode on the walls. Skellett reported no time-lag that could not be explained by mobility of positive ions. Low efficiency of the carbon to release of electrons by metastable atoms no doubt accounts for the nonappearance of the long time-lag.

IONIZATION OF IMPURITIES

Before concluding, we must consider the other possible effect of metastable atoms. As shown by Penning,²³ gas impurities with an ionization potential below the metastable excitation potential of the main gas may be ionized by collisions with metastable atoms. The rate of disappearance of metastable atoms would in this case be proportional to the impurity present.

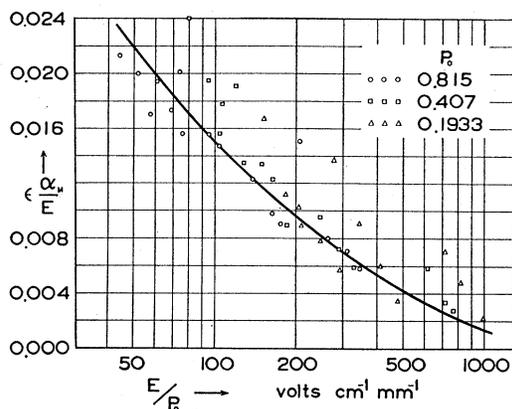


FIG. 10. $\epsilon \cdot \alpha_m / E$ vs. E/p_0 where ϵ is the number of electrons liberated from the cathode per impacting metastable atom and α_m is the number of metastable atoms produced per electron in traveling one cm in the direction of the electric field. $\epsilon \cdot \alpha_m / E$ is proportional to the fractional energy dissipation of the electrons in exciting atoms to the metastable state. It is estimated that ϵ is greater than 0.4.

²³ F. M. Penning, *Zeits. f. Physik* **46**, 335 (1928).

A study of Fig. 8 shows a slight tendency of the data toward a negative curvature. This is what would be expected if there were ionization of caesium vapor by metastable atoms. When a least square evaluation of the curvature tendency was made it was found that the pressure of caesium necessary to account for the result is slightly more than 10 times the vapor pressure of free caesium at room temperature, 1.7×10^{-6} mm of Hg (if every collision of a metastable argon atom with a caesium atom resulted in ionization). Nevertheless, although ionization of impurities may occur, it cannot account for the entire action of the metastable atoms, and the evidence suggests that the Penning effect is of minor importance in this experiment. Moreover, the curvature (Fig. 8) may also be explained on the assumption that there is a small but finite probability of a metastable atom losing its energy in a collision with a normal argon atom; one destructive collision in about 10^4 would suffice. Of course any loss of metastable atoms by collision of the second kind with electrons would be negligible since the current density is so small.

CONCLUSION

In the Townsend discharge for argon gas and an activated caesium cathode, the important auxiliary mechanisms are the secondary electron emission by positive ions and the liberation of cathode electrons by the impact of metastable argon atoms. The diffusion time of the latter remarkably efficient agent is the source of the long time-lag observed in this type of discharge.

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