# An Investigation of the Properties of Proportional Counters. I\*

M. E. Rose and S. A. Korff

Bartol Research Foundation of the Franklin Institute, Swarthmore, Pennsylvania

(Received April 2, 1941)

A theoretical and experimental investigation of the amplification properties of proportional counters has been made. On the basis of a discharge mechanism in which the amplification is due to electron avalanches the theory has been developed with the following primary assumptions: (1) All secondaries are produced by electron impact with no photon emission in the counter gas and thus no photoelectric emission at the cathode; (2) fluctuations in energy loss and specific ionization are neglected. The analysis involves the determination of the average number of ionizing collisions per unit distance (average energy) as a function of position in the counter, and the spatial extent of the ionization region near the wire anode. The former quantity is calculated explicitly in terms of the constants of the counter and the latter is shown to be simply related to the threshold voltage for proportional amplification, and is most suitably determined from the measurements. The gas fillers used in the measurements of the amplification factor were, for the most part, methane-argon mixtures of various relative concentrations and total pressures. In addition, other polyatomic mixtures, illuminating gas, BF<sub>3</sub>-A and ether-A were used. Comparison of theory and experiment gives quite satisfactory agreement in all cases but two: large A concentration (50 percent or more at a total pressure of 10 cm Hg) and low total pressure (5 cm, or less, with a

### I. INTRODUCTION

A LTHOUGH counters are extremely useful instruments and have been used extensively as sensitive detectors for all types of radiations, the investigation and clarification of the physical phenomena taking place in the counter has until recently been neglected.<sup>1</sup> In the case of counters operated in the sub-threshold region our understanding of the discharge mechanism, of the procedures to be followed and of the limits within which useful results may be obtained has been mixture containing 90 percent CH<sub>4</sub>). In the anomalous cases the measured amplification factor rises extremely rapidly and such mixtures are therefore somewhat undesirable for stability reasons. This unstable behavior is characteristic of gas mixtures of the monatomic and/or diatomic type. An explanation of the difference in amplification properties of the polyatomic and simpler type gases is proposed. It is shown that while photon emission in the ultraviolet and subsequent contributions to the avalanche by photoelectric emission at the cathode is to be expected for the simpler gases, polyatomic gases should effectively quench such photon emission by virtue of greater energy loss of the slower electrons in exciting molecular vibrations and rotations. Moreover, appreciable emission of ultraviolet light by polyatomic molecules under electron bombardment is not to be expected. This explanation receives confirmation from cathode tests which were performed: measurements of pulse size, or amplification factor, for solid and perforated, oxidized and non-oxidized Cu cathodes. Further support of these arguments is found in experiments of other investigators: energy loss and mean free path measurements, electron bombardment of gases in a photo-cell. Finally, specific recommendations as to desirable pressure and concentration in the CH4-A mixture are given.

particularly limited with the result that the use of these counters has been somewhat restricted. Aside from the intrinsic interest in the subthreshold discharge phenomena, a more thorough understanding of them and the way in which they affect counter behavior, or even a complete set of empirical rules regarding counter properties, would contribute to a more efficient use and a wider application of these counters to new types of measurements.

From previous investigations<sup>2</sup> it has been established that, for voltages well below the Geiger threshold, and over most of the subthreshold region, the pulse size on the central system will be proportional to the initial ionization produced by the primary particles (discriminatory property of the proportional counter).

<sup>\*</sup> This is a report of a preliminary survey of proportional counter properties dealing with the questions of discharge mechanism and the effect of the nature of the gas on counter amplification. A subsequent paper will provide more complete data on amplification and other proportional properties.

<sup>&</sup>lt;sup>1</sup>A discussion of the mechanism of the discharge in Geiger-Mueller counters has been given by C. G. Montgomery and D. D. Montgomery, Phys. Rev. 57, 1030 (1940). Attention may be called to the definition of the Geiger threshold adopted by these authors, which we follow, as that voltage above which all pulse sizes are independent of the initial ionization produced in the counter.

<sup>&</sup>lt;sup>2</sup> H. Geiger and O. Klemperer, Zeits. f. Physik **49**, 753 (1928); S. A. Korff, Rev. Mod. Phys. **11**, 211 (1939); Rev. Sci. Inst. **12**, 94 (1941); G. Brubaker and E. Pollard, Rev. Sci. Inst. **8**, 255 (1937); C. G. Montgomery and D. D. Montgomery, Rev. Sci. Inst. **11**, 237 (1940).

In addition, in using the proportional counter one is interested in the following two questions: First, the more qualitative question as to what type of gas fillers should be used and secondly, the quantitative dependence of counter properties on gas pressure, relative concentration in the case of a mixture and on geometry (cathode, anode sizes). Here we shall be mainly interested in the first of these questions although some results are presented (Section IV) to show the effect of the other variables. In the following section the theoretical treatment of the discharge process is presented.

### II. THEORETICAL

The properties of the proportional counter are most suitably defined by the characteristic curve or the dependence on counter voltage of the pulse size produced by amplification of the initial ionization. By virtue of the proportionality property we may define the amplification factor in a general way as the ratio of charge collected by the central wire anode to the amount of charge formed by the primary particle. In general, it is desirable that the counter have a large amplification factor so as to minimize the necessity for vacuum tube amplification; or, when portability and weight are important considerations, as in cosmic-ray balloon flights, it is economical to use counters with large amplification so as to allow the use of smaller voltage sources. On the other hand, counters whose amplification does not vary too sensitively with voltage would be more desirable for the minimization of errors due to voltage fluctuations.

The analysis of the proportional discharge requires first the postulation of some mechanism whereby the primary ionization is amplified. This primary ionization, whether it is direct ionization by charged particles,  $\gamma$ -ray secondaries or due to recoil nuclei produced by neutrons, will take place in the immediate vicinity of the path of the primary particle. This also applies to the ionization produced by such secondary electrons as are formed with energies above the ionization potential of the counter gas. Excluding the very rare case that the primary particle passes very near the wire (within a diameter, say) the initial energy of the secondary electrons is rapidly expended in various types of inelastic impacts and there will be no further ionization until the electrons are in the immediate vicinity of the wire where, by virtue of the strong field there prevailing, their energy becomes sufficiently great and increases sufficiently rapidly to produce sustained ionization. If the distance from the wire at which this ionization starts is several mean free paths a large multiplication of the charge density will take place by the successive and cumulative ionizations. In short, an electron avalanche is formed.

This discharge mechanism obviously implies the assumption that all secondaries are produced by electron impact and that all of them reach the wire. Explicitly we assume that:

1. (a) The emission of ultraviolet light and the consequent photoelectric effect at the (Cu) cathode is negligible. This is the most interesting assumption since evidence to be presented below (Sections IV and V) indicates that photoeffects are indeed negligible in polyatomic gases but not in simpler gases, monatomic and diatomic. (b) Secondary electron emission by positive ions at the cathode is neglected. This assumption should be well justified below the Geiger threshold where no effects of positive ion space charge are evident.

2. Recombination and electron attachment to neutral molecules can be neglected. Recombination will be very unimportant under the conditions of high field and low pressure usual in counters. Electron attachment will be appreciable for only a few gases ( $O_2$ , air and certain other gases not usually used in counters).<sup>3</sup>

Even in this picture the amplification process is of such complexity that it is necessary to make a further assumption :

3. We neglect fluctuations in energy loss and in specific ionization. While it is difficult to justify this assumption on *a priori* grounds the comparison of theory and experiment (Section IV) would seem to indicate that fluctuations do not play an important role.

We now consider a single electron arising from the initial ionization and calculate the amplification factor as the number arriving at the wire. Denoting the number of electrons at a distance r

<sup>&</sup>lt;sup>3</sup> An effect on pulse size of the distance of the primary particles from the wire has been noted by Brubaker and Pollard (reference 2), and this is attributed to electron capture. However, under usual conditions of operation the primary particles are not very well collimated or are scattered by the counter walls so that no capture effects should be observed. In any case the capture introduces only a time lag ( $\sim 10^{-6}$  sec.), because of the small mobility of the negative ions formed, and the time constant of the counter wire system can always be made longer than this.

from the center of the wire by N(r) we have for the increase of N(r) when r decreases by dr,

$$-dN = N(r)dr \int_{0}^{\epsilon_{m}} \frac{n(\epsilon, r)}{l(\epsilon)} d\epsilon.$$
 (1)

Here we denote by  $\epsilon$  the excess of the electron energy over the (average) ionization potential of the gas (or gas mixture) in the counter. For the sake of brevity we shall refer to the excess energy  $\epsilon$  as the *energy*, whereas the actual energy is unitalicized. In (1)  $n(\epsilon, r)d\epsilon$  is the fraction of the total number of electrons at r with *energy* between  $\epsilon$  and  $\epsilon + d\epsilon$ , that is, the *energy* distribution normalized to unity.  $l(\epsilon)$  is the mean free path for ionization and  $\epsilon_m$  is the maximum *energy* of the electrons at r. The integral in (1) is thus simply the average number of ionizing collisions per unit distance which we shall denote by  $\kappa(r)$ , so that we have from (1)

$$N(r) = \exp \int_{r}^{r_0} \kappa(r) dr.$$
 (2)

In (2) we have introduced  $r_0$  as the distance from the center of the wire at which the avalanche-producing ionization starts.

The amplification factor A is then given by  $N(r_1)$  where  $r_1$  is the radius of the wire:

$$A = N(r_1) = \exp \int_{r_1}^{r_0} \kappa(r) dr.$$
 (3)

It is seen, as may have been anticipated, that the main task of the theory is the calculation of the average number of ionizing collisions per unit distance  $\kappa(r)$  and the spatial extent of the ionization region  $r_0$ . Considering the former quantity it is clear that in principle a knowledge of the *energy* distribution at each point r is involved although all that is eventually required is the average value of a known function of the *energy*, viz.:

$$\kappa(r) = (1/l(\epsilon))_{\text{Av}}.$$
 (4)

In order to specify the *energy* dependence of the mean free path we recognize the fact that the electrons attain an energy of the order of the ionization potential ( $\sim 15$  volts) at a point very near the wire and that the energy gain in the region of ionization is sufficiently slow so that the electron energy at the wire is only a few times the ionization potential. The maximum energy may, in fact, be computed (cf. Eq. (10) below), and is generally 30–50 volts for counter voltages of as much as 1000 volts, the inelastic collisions being almost sufficient to overcome the effect of the accelerating field. Therefore the energy region pertinent to our considerations is such that the ionization cross section increases linearly with energy.<sup>4</sup> We write for the cross section

$$\sigma(\epsilon) = a\epsilon, \tag{5}$$

where a is a constant depending on the nature of the gas (cf. Table I below); and for the mean free path

$$l(\epsilon) = 1/a N_m \epsilon, \qquad (5a)$$

where  $N_m$  is the number of molecules (or atoms) per unit volume (see also Eq. (15)). Thus we have

$$\kappa(r) = a N_m \epsilon_{Av} \tag{6}$$

and we are concerned with the determination of the average energy,  $\epsilon_{AV}$ .

In order to obtain the average *energy* we first consider the manner in which the *energy* distribution arises. The distribution, according to the present scheme (no fluctuations), arises from the fact that electrons at r have different points of origin r' and the *energy* at r is given by the average *energy* gain in traversing the distance from r' to r. The average *energy* gain is determined by<sup>5</sup>

$$-d\epsilon/dr = (cV_0/r) - \gamma(\epsilon).$$
(7)

Equation (7) determines  $\epsilon$  for those electrons at r which have started from r', as a function of r and r' subject to the initial condition

$$\epsilon(r=r')=0. \tag{7a}$$

Here  $\gamma(\epsilon)$  is the energy loss per unit distance due to all energetically possible inelastic processes,  $V_0$  is the voltage across the counter and 1/c

<sup>&</sup>lt;sup>4</sup> Cf. for instance, K. T. Compton and I. Langmuir, Rev. Mod. Phys. 2, 127 (1930).

<sup>&</sup>lt;sup>5</sup> The average *energy* determined from the integration of (7) is, of course, to be distinguished from the true average *energy* referred to above. The former  $\epsilon(r, r')$  is the average *energy* of an electron at r when it has started from a given point r', while the true average is  $\epsilon(r, r')$  averaged over all points of formation with the appropriate weight factor, viz. the number of electronic charge so that all energies are measured in the same units as the counter voltage.)

 $=\log(r_2/r_1)$  so that  $\frac{1}{2}c$  is the capacity per unit length of the counter, with  $r_2$  the cylinder (cathode) radius.

According to the above, the number of electrons at r having an energy between  $\epsilon$  and  $\epsilon + d\epsilon$ is just the number which were formed at a distance between r' and r' + dr' where  $dr' = (dr'/d\epsilon)d\epsilon$  and r' is given in terms of  $\epsilon$  and r by (7). Thus the energy distribution at r depends on the distribution at r' and is accordingly given by the following integral equation:

$$N(r)n(\epsilon, r)d\epsilon = dr'N(r')\int_{0}^{\epsilon_{m'}} \frac{n(\epsilon', r')}{l(\epsilon')}d\epsilon'.$$
 (8)

Here  $\epsilon_{m'}$  is the maximum energy at r', and we have introduced the actual number of electrons with energy between  $\epsilon$  and  $\epsilon + d\epsilon$ , namely  $N(r)n(\epsilon, r)d\epsilon$ .

An exact solution of this integral equation, and the determination of the distribution function itself, appears to be out of the question. Instead we proceed directly to the average energy which may be obtained by the following approximate method. First we put the average energy in evidence by noting that  $n(\epsilon, r)$  has the dimensions of  $(energy)^{-1}$  and hence can be written quite generally as

$$n(\epsilon, r) = (1/\epsilon_{Av})\varphi(\epsilon/\epsilon_{Av}), \qquad (9)$$

where  $\varphi$  is a dimensionless function of the indicated dimensionless argument and the dependence on r is contained in  $\epsilon_{Av}(r)$ . Then the essential approximation consists in the fulfillment of the integral equation only for those energies for which the number of electrons is greatest. Since most of

TABLE I. Rate of increase of the ionization cross section with energy.

Gas	$a(10^{17} \text{ CM}^2/\text{VOLT})$	Reference
A	1.81	*
Ne	0.14	*
He	0.11	*
H,	0.46	t
$\overline{O_2}$	0.66	÷
N <sub>2</sub>	0.70	ŧ
NÖ	0.74	ŧ
CO	0.83	÷
$C_2H_2$	1.91	÷
CH₄	1.24	**

\* P. T. Smith, Phys. Rev. 36, 1293 (1930).
 † J. T. Tate and P. T. Smith, Phys. Rev. 39, 270 (1932).
 \*\* A. Ll. Hughes and E. Klein, Phys. Rev. 23, 450 (1924).

the electrons at r, more than half, were formed in the last mean free path and since the more remote the point of origin (i.e., the greater the energy) the fewer the number of corresponding electrons, the energy distribution will be monotonically decreasing with the most important energy region at  $\epsilon = 0$ . Thus we may set r' = r,  $\epsilon = 0$ and  $d\epsilon/dr' = -d\epsilon/dr = cV_0/r$ . Then the average energy is obtained at once from (8) with the help of (5a), (6) and (9). We find

$$\epsilon_{Av}(r) = \left(\frac{\varphi(0)}{aN_m} \frac{c V_0}{r}\right)^{\frac{1}{2}}.$$
 (10)

Thus we see that the average *energy*, and the width of the energy distribution which will be proportional to  $\epsilon_{Av}$ , both increase as the electrons approach the wire, as was to be expected. From (6) the average number of ionizing collisions per unit distance follows immediately.6

$$\kappa(r) = (\varphi(0)aN_m c V_0/r)^{\frac{1}{2}}.$$
 (11)

$$\int_0^\infty \varphi(x) dx = 1$$

and from the definition of x,

$$\int_0^\infty x\varphi(x)dx=1.$$

Various functional forms for  $\varphi$  may be assumed and after Various functional forms for  $\varphi$  may be assumed and after the above conditions have been imposed  $\varphi(0)$  may be calculated. For a great variety of functions we find  $\varphi(0)$ is between  $\frac{1}{2}$  and 2 with by far the great majority of the values lying near unity. Accordingly, in the numerical applications to follow we have taken  $\varphi(0)=1$ . It may be noted that the dependence of the average number of ionizing collisions per unit distance on r is a direct con-sequence of the cylindrical geometry. The result (11) may at once be generalized for any geometry by replacing  $cV_0/r$  by the electric field  $\mathcal{E}$ . (There will also be a slight geometry dependence of  $\varphi(0)$  but this can be neglected in first approximation.) In the case of plane parallel electrodes we may compare our result with the measurements of the first Townsend ionization coefficient, generally denoted in the literature by  $\alpha$ . For field strengths comparable to those near the counter wire and for the usual counter pressures (~100 mm Hg) the ratio  $\mathcal{E}/p$  is of order 1000 volts/mm. For such values of  $\mathcal{E}/p$  the observations show that  $\alpha/p \equiv \kappa/p$  is proportional to  $(\mathcal{E}/p)^{\frac{1}{2}}$  in exact conformity with the result predicted by (11). Fairly good quantitative agreement is also found. Cf. L. B. Loeb, *Fundamental* Processes of Electrical Discharge in Gases (John Wiley and Sons, 1939), p. 360.

<sup>&</sup>lt;sup>6</sup> The quantity  $\varphi(0)$  which occurs in these results, and which is essentially the number of slowest electrons, cannot be evaluated uniquely without knowing the actual form of the distribution function. However the function  $\varphi$ is sufficiently restricted so that  $\varphi(0)$  cannot, in general, vary over wide limits. In addition to the requirement that  $\varphi$  shall be a monotonically decreasing function of its argument, there are two further conditions to be imposed on this function. With  $x = \epsilon/\epsilon_{AV}$  we have from the normalization condition,

The final step in the calculation is the consideration of  $r_0$ . According to the way in which this quantity was introduced (see Eq. (2)) it is the distance from the wire at which the avalanche starts, and at this point one may expect fluctuations to play a more important role. Therefore we may not expect any reasonably accurate calculation of  $r_0$  in terms of the physical constants of the counter without a much more refined and greatly complicated theory. However, it may be shown that  $r_0$  is related in a simple way to another constant of the counter which could be directly measured and which is of considerably greater interest in defining the properties of the proportional counter. To see this we introduce a critical energy  $\epsilon_0$  which is the average energy at the point where ionization starts. According to (10)  $\epsilon_0$  is given by

$$\epsilon_0 = \left(\frac{\varphi(0)}{aN_m} \frac{cV_0}{r_0}\right)^{\frac{1}{2}}.$$
 (12)

The critical energy  $\epsilon_0$  for the onset of the avalanche will be determined by the nature of the gas (ionization potentials and cross section) but should not depend on the counter voltage or on geometrical factors. It then follows from (12) that  $r_0$  increases linearly with counter voltage. In particular there will be some counter voltage for which ionization does not begin until the electrons reach the wire, i.e.,  $r_0 = r_1$ . At this voltage, the threshold voltage for the proportional region, the amplification factor becomes unity and below this voltage the counter acts as an ionization chamber. Denoting the proportional threshold by  $V_t$  we have

$$r_0 = r_1 V_0 / V_t.$$
 (13)

Thus  $r_0$  may be defined in terms of the directly measurable voltages and wire radius.

With (13) and (11) the final result for the amplification factor follows readily from (3):

$$A = \exp 2(a N_m c r_1 \varphi(0) V_0)^{\frac{1}{2}} [(V_0 / V_t)^{\frac{1}{2}} - 1] \quad (14)$$

in which, as was mentioned above,  $\varphi(0)$  is to be set equal to unity and  $V_t$  is to be determined from the measurements. The quantity a in (14), that is, the rate of change of the ionization cross section for energies just above the ionization potential, is given in Table I for those gases for which measurements exist. For the quantity  $aN_m$  in the case of a mixture of gases, we obviously have

$$aN_m = \sum_i a_i N_{m\,i},\tag{15}$$

where  $N_{mi}$  is the particle density of molecules of the *i*th constituent of the mixture.

### III. Apparatus

The amplification factor for the several counters used in the experiments was determined by measuring the counter voltage corresponding to a previously determined pulse size. The pulse sizes were measured by using a variable-gain amplifier and a thyratron recorder. The amplifier consisted of a conventional resistance-coupled circuit, employing one or alternatively three stages, with tubes of type 6J7. The gain of each stage could be controlled. For any given bias setting a certain minimum pulse size was necessary to record. Calibration of the gain settings was accomplished by the use of a simple pulse-generator. This consisted of a resistance potential divider across a battery, equipped with a key and capacitatively coupled to the input.

The potential applied to the counter was produced by a modified Street-Johnson type of voltage regulator. The counter voltage remained independent of the fluctuations in the a.c. line, and could be controlled and kept stable in the range between 200 and 1600 volts. The output voltage was read on a microammeter in series with a standard resistance and was believed to be accurate to within 2 percent.

The various counters were attached to a manifold connected to a vacuum system. Gassupply bulbs were attached so that the kind and amount of each gas could be changed at will, and the pressure in the system and in all the counters could be read on a mercury manometer. Many of the runs were repeated in random order to insure reproducibility.

In each case the maximum pulse size due to Po  $\alpha$ -particles passing down the length of the counter was measured. Therefore the initial ionization is that due to  $\alpha$ -particles with a residual range equal to the length of the sensitive part of the counter. This ionization may then be obtained from the integrated Bragg curve,<sup>7</sup> re-

<sup>&</sup>lt;sup>7</sup> For specific ionization data see, for example, M. G. Holloway and M. S. Livingston, Phys. Rev. 54, 18 (1938).



FIG. 1. Total ionization of alpha-particles in air at  $0^{\circ}$ C and 76 cm Hg as a function of residual range.

produced in Fig. 1 for convenience, and for gases other than air the conversion factors as given by Gurney<sup>8</sup> were used. The amplification factor is then A = CP/en, where C is the capacity of the central system, P the pulse size, n the initial ionization and e the electronic charge.

### IV. COMPARISON OF THEORY AND EXPERIMENT

In addition to providing a test of the theory the present measurements were designed to furnish a first orientation with regard to proportional counter properties. In fact the limitations as to measurable pulse size of the amplifier used allowed measurements to be made only for the higher voltages (roughly the upper half of the proportional region); since this is the region of large amplification, it is, to be sure, the more important region from a practical point of view. However, the proportional threshold voltages were therefore not measured directly but were obtained by fitting the theoretical curves at one experimental point.<sup>9</sup>

For the most part the measurements were carried out with methane-argon mixtures which were found to be a suitable gas filler for the counters. We have investigated the effect of the relative concentration of the constituents of this mixture and the results are shown in Fig. 2. The relative concentrations were 100, 90, 75 and 50 percent methane. In each case the highest voltage measured marks approximately the limit of the proportional region. With the amplification factor plotted on a logarithmic scale the theoretical curves over the measured range of voltages are, in appearance, virtually straight lines. For the three methane-rich mixtures the agreement between theory and experiment is seen to be very satisfactory, the deviations of the measured points lying within the experimental error. The threshold voltages as determined by fitting at each experimental point showed an extreme variation of only 5 percent or less. These threshold voltages, which with the maximum voltages measured give the approximate breadth of the proportional region, are listed in the second column of Table II.

The results for the 50 percent concentration are anomalous in the sense that it is not possible to fit a theoretical curve to the observed points. The measured amplification factor rises too rapidly with voltage. This case requires special consideration which is reserved for the next section.

Figure 3 shows the experimental points for the amplification factor and the corresponding theoretical curves for mixtures of 90 percent methane and 10 percent argon for total pressures of 5, 10, 20, 30 and 40 cm. Again, in the case of all but the lowest pressure good agreement between theory and experiment is obtained. The threshold voltage as a function of pressure is given in the last column of Table II. It is interesting to note that these four threshold voltages show a linear increase with pressure so that the amplification factor (from Eq. (14)) decreases essentially exponentially with increasing pressure. The same sort of anomalous behavior as noted before is found for the lowest pressure (cf. Section V).

The experimental points and theoretical amplification-voltage curves for some widely used mixtures are shown in Fig. 4. The  $CH_4$ - $H_2$ mixture was actually illuminating gas whose heterogeneous composition was known—except

<sup>&</sup>lt;sup>8</sup> R. W. Gurney, Proc. Roy. Soc. A107, 332 (1925). Although no measurements exist for  $CH_4$ , used in the experiments described below, the specific ionization may be taken equal to that in Ne, with very little error, because of the similarity of electron structure.

<sup>&</sup>lt;sup>9</sup> Measurements down to the ionization chamber region of voltage would require a more elaborate amplifier or the use of a balanced pliotron circuit with a counter carefully shielded by guard rings. Using the latter apparatus such measurements are now being conducted in this laboratory. In addition to providing a complete exploration of the proportional region a direct measurement of the proportional threshold as well as the amplification factor is then possible.



FIG. 2. The amplification factor (logarithmic scale) plotted against counter voltage for various relative concentrations in CH<sub>4</sub>-A mixtures at 10-cm pressure. The points are experimental and the full curves are theoretical with one point adjusted (see Eq. (14)). The numbers affixed to the curves give the relative concentration of CH<sub>4</sub> in percent. The dimensions of the counter were: wire diameter 0.075 mm; Cu cylinder, diameter 1 cm and length 3 cm.

for a small amount of  $CO_2$  the mixture is composed of gases whose ionization at low energies has been measured (Table I). The BF<sub>3</sub>-A counter was of the type used in neutron measurements. (In obtaining the theoretical curves for this and the A-ether counter the ionization in BF3 and in ether was obtained by fitting at a second point. The ionization coefficient a obtained for ether was then in agreement—within 5 percent—with the value to be expected with the assumption of additivity of cross sections and from the data listed in Table I for H<sub>2</sub>, O<sub>2</sub> and CO, viz. a = 6.2 $\times 10^{-17}$  cm<sup>2</sup>/volt). The main point of interest in connection with these measurements is that in each case the mixtures contained a polyatomic constituent of rather large concentration: 31 percent  $CH_4$  in the illuminating gas, 47 percent ether in the ether-A mixture and 82 percent BF<sub>3</sub> in the BF<sub>3</sub>-A mixture. The total pressures were 3.5, 17 and 11 cm, respectively. The threshold voltages for the three counters were determined to be 450, 470 and 955 volts, respectively.

Finally, the effect of geometry, which is of course mainly dependent on wire diameter, was investigated. As is to be expected, and as is predicted by the theoretical formula (Eq. (14)), the amplification factor increases as the wire diameter is made smaller. The counters were filled with the 75 percent CH<sub>4</sub>-A mixture at a total pressure of 10 cm. The results for the wire diameter of 0.075 mm have already been given, Fig. 2. Decreasing the wire diameter to 0.025 mm shifts the amplification curve by about 100 volts

in the direction of lower voltages. This corresponds to an increase in the amplification by a factor of 5. Increasing the wire diameter to 0.125 mm shifts the amplification curve towards higher voltages by about 40 volts, as compared to the curve of Fig. 2. This is equivalent to a reduction in the amplification factor by 50 percent. As in the case of the intermediate wire size, the measurements for the 0.025 and 0.125-mm wires may be satisfactorily fitted by a theoretical curve. Thus, with the exception of the anomalous cases noted above, the theory is in agreement with the data for concentration, pressure and geometry dependence.

## V. Comparison of Discharge in Polyatomic and Simple Gases

The anomalously rapid rise of the amplification factor, at least in the case of the mixture with relatively large argon concentration, is not surprising. It had been previously observed that the amplification in counters filled with A, A-O<sub>2</sub>, A-H<sub>2</sub>, Ne-H<sub>2</sub> or H<sub>2</sub> will increase by a factor of as

 TABLE II. Threshold voltages for proportional counter discharge.

Pressure 10 cm Hg Conc. CH4 (%)	Vt VOLTS	0.90 CH4-0.10A Total Pressure cm Hg	Vt Volts
100 90 75	430 350 270	40 30 20 10	1090 840 610 350



much as 100 when the counter voltage is increased by only 2–5 volts.<sup>10</sup> On the other hand such mixtures as CCl<sub>4</sub>-A, BF<sub>3</sub>-A and illuminating gas were observed to give a much more slowly rising amplification. We have also used A and H<sub>2</sub> and for each gas have verified the extremely rapid rise of the amplification with voltage. In no case can the theory (based on the assumptions of Section II) account for this behavior.

This rather striking difference in the amplification of counters filled with simple gases and with polyatomic gases may be explained in terms of a fundamental difference in the discharge mechanism in the two cases. In view of the agreement obtained between theory and experiment (Section IV) the discharge mechanism postulated in Section II may be regarded as correct for polyatomic gases. For the simpler gases it is most probable that the first assumption regarding the absence of photoelectric emission no longer holds. Indeed, it is easier to account for the presence of photons capable of emitting photoelectrons than it is to explain their absence. Since the electrons attain sufficient energy to ionize they must also cause excitation of electronic levels with the subsequent emission of ultraviolet radiation. The emission of photoelectrons at the cathode then initiates a new avalanche and the process may be repeated several times. The

rapid rise of the amplification is then readily understandable. To account for the presumable absence of such photons in the mixtures relatively rich in a polyatomic constituent we recognize the fact that over the greatest part of the path of the electrons their energy is insufficient for electronic excitation in the gas molecules. By far most of the inelastic collisions may therefore excite only vibrational and rotational energy so that, except near the wire, only infra-red radiation could be emitted. In the case of polyatomic molecules, as contrasted to the simpler molecules, the much greater number of degrees of freedom for the molecular motions may be expected to enhance the excitation of low energy vibrations. Moreover, because of the great number of collisions taking place in the "infra-red" region of the counter, even a small increase in the probability of vibrational excitation would have an important effect. Hence, in a gas with fairly large concentration of polyatomic molecules, the energy of the electrons would rise more slowly than in the simpler gases and the spatial extent of the "ultraviolet" region, near the wire, would become considerably smaller. This would then mean a smaller intensity of the higher energy photons.

In addition, emission of ultraviolet radiation from the polyatomic molecules excited by electron bombardment would not be expected as a general rule. For methane, as an example, no

<sup>&</sup>lt;sup>10</sup> S. A. Korff, Rev. Mod. Phys. 11, 213 (1939).



FIG. 4. The amplification factor (logarithmic scale) against counter voltage for various poly-atomic gas mixtures. Note change in voltage scale for each curve. The scale for the amplification factor in the case of CH4-H2 is on the left, the scale for the other two mixtures is on the right. Points are experimental, curves theoretical. Counter dimensions:  $CH_4$ - $H_2$  and  $BF_3$ -A counters, wire diameter 0.075 mm, cylinder diameter 1.8 cm and length 20 cm; ether-A counter, same as in Fig. 2.

absorption in the ultraviolet is obtained until 1600A (at 10-cm pressure) and then the appearance of the absorption bands indicates predissociation of the molecule.<sup>11</sup> In general it may be expected that the absorption of radiation of 7-8 volts or more, or bombardment with electrons of such energies, would lead to dissociation rather than light emission. Thus, the binding energy of the H<sub>2</sub> molecule in methane is 3.7 volts and the binding energy of the H atom is 4.5 volts so that with 1600A radiation dissociation could take place leaving some 3 or 4 volts as kinetic energy of the products.<sup>12</sup> Thus the methane in CH4-A mixtures, and presumably other polyatomic molecules, would appear effectively to quench the emission of ultraviolet by providing a more probable mode of energy loss for the lower energy electrons and by failing to contribute to such emission even when the excitation of electronic levels is energetically possible.

As a test of the above hypothesis we have performed the following experiments. Three counters were simultaneously attached to the manifold so that they could be filled with the same gas at exactly the same pressure. The counters themselves were identical in all but one respect: One had a non-oxidized Cu cathode as usual, another had a similarly clean cathode made of Cu screening so that the cathode area was reduced by nearly 50 percent and the third was constructed of the same screening but was oxidized to CuO. The counters were otherwise the same as those used with the CH<sub>4</sub>-A mixtures, (Figs. 2 and 3). The amplification curves with hydrogen in the counters are shown in Fig. 5. It is seen that at the higher voltages there is a very definite cathode effect. The reduction of pulse sizes by oxidation of the cathode (2nd and 3rd counters) shows that the very small change in electric field due to the substitution of screening for solid Cu is not responsible for the reduction in pulse size upon perforation of the cathode (1st and 2nd counters). These results are most plausibly interpreted in terms of avalanche production by photoelectrons.<sup>13</sup> At the lower voltages inappreciable photon emission is indicated as could be expected from intensity considerations (much smaller number of electrons in the initial avalanche). With argon in the counters similar results were obtained. When the same experiments were repeated with only methane in the counters the results shown in Fig. 6 were found. Here it is seen that even at the highest voltages

<sup>&</sup>lt;sup>11</sup> Cf. for instance, A. Rose, Zeits. f. Physik **81**, 751 (1933). <sup>12</sup> L. G. Smith, Phys. Rev. **51**, 263 (1937).

<sup>&</sup>lt;sup>13</sup> Unfortunately the comparison of pulse sizes with Cu and CuO cathodes cannot be checked with photoelectric data. Such measurements as exist show that from the photoelectric thresholds to 1800A Cu gives a greater yield of photoelectrons, R. Fleischmann, Ann. d. Physik 5, 73 (1930). However, the cathode tests with H<sub>2</sub> may be taken as an indication that this is also true at 1000A.

there is no evidence for a cathode effect. A similar absence of cathode effect was obtained with the other polyatomic gas mixtures (Fig. 4). On the other hand the two CH<sub>4</sub>-A mixtures whose amplification factor rose too rapidly, Figs. 2 and 3, showed a cathode effect only slightly less pronounced than that shown in Fig. 5 for H<sub>2</sub>. These results therefore justify the assumption (1) on which the theory was based and form a strong argument for the essential correctness of the proposed hypothesis as to the role of photons in simple gases as contrasted with the "quenching" action of the polyatomic gases.

Further support for the arguments presented in the above may be obtained from other experiments. Thus, Bailey and Duncanson<sup>14</sup> have measured energy losses and mean free paths for slow electrons in various gases and have found that certain polyatomic gases of the hydrocarbon type and triatomic gases (NH<sub>3</sub>, H<sub>2</sub>O vapor and  $CO_2$ ) have smaller mean free paths and larger fractional energy losses at less than 1 volt, where only infra-red may be excited, than at higher energies. On the other hand, inert gases and several of the diatomic variety were shown to have the largest energy losses and smallest mean free paths at the higher energies where visible and ultraviolet light may be excited. More direct experiments, from the present point of view, were carried out by Glocker<sup>15</sup> who found that electrons



FIG. 5. Pulse size, or amplification factor (linear scale, arbitrary units), against counter voltage for the three cathodes described in the figure. Counter was filled with  $H_2$  at 7.3-cm pressure. Counter dimensions as in Fig. 2.



FIG. 6. Pulse size, or amplification factor (linear scale, arbitrary units), against counter voltage for the three cathodes described. Counter was filled with  $CH_4$  at 7.3 cm pressure. Counter dimensions as in Fig. 2.

with energies up to 17 volts bombarding methane in a photoelectric cell gave no measurable photoelectric currents. However, with hydrogen in the photo-cell photoelectric currents were obtained for electron energies above 13 volts which is slightly larger than the energy of the first excited state in  $H_2$ .

These experiments have shown that for effective photon quenching and consequent stability not only must the relative concentration of the polyatomic gas be greater than a certain critical value but in addition the absolute amount of this gas must not be too small. While the existence of a critical concentration is easily understood in the light of the foregoing qualitative considerations, the existence of a critical pressure could be understood only on the basis of a more quantitative treatment of the photon emission and photoelectric effect in the counter. However, this pressure effect may be accepted as an empirical fact to be used as a guide in the determination of the most suitable conditions for counter operation.<sup>16</sup> Thus, if it is desired to operate the counter so as to obtain large and stable amplification, the gas mixture should contain polyatomic as well as simpler type constituents with pressure and concentration conditions near the critical limits. With CH<sub>4</sub>-A mixtures, for example, a relative concentration of 75 percent CH<sub>4</sub> at a total pressure of 10 cm would be close to optimum conditions.

<sup>&</sup>lt;sup>14</sup> V. A. Bailey and W. E. Duncanson, Phil. Mag. 10, 145 (1930).

<sup>&</sup>lt;sup>15</sup>G. Glocker, Proc. Nat. Acad. Sci. 11, 74 (1925).

<sup>&</sup>lt;sup>16</sup> Cf. S. C. Curran and J. E. Strothers, Proc. Camb. Phil. Soc. **35**, 654 (1939) where a similar effect is observed for counters filled with argon-alcohol mixtures above the Geiger threshold.