Discharge Spread in Geiger Counters

I. With Self-Quenching Gases

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Despite the work of Korff, Stever, and others, many aspects of Geiger counter behavior are as yet unexplained. In particular, the extent to which the photons produced in the counter discharge are of importance, and the manner in which they are absorbed in the counter gas, seem to have been insufficiently studied. The experiments described here were made with single wire, divided cathode, counters of various types. The results show that photon absorption seems to be less complete than is often thought, but that when such absorption is fairly small, it may be almost entirely manifest as a cathode, not a gas, effect, as shown by experiments with a new type of divided cathode counter in which the cathode effect is virtually eliminated.

I. INTRODUCTION

THE differences in the behavior of proportional counters containing simple and complex gases[1,2] have been attributed mostly to the greater importance of photo-emission of electrons from the counter cathode in the former case. The experiments of Rose and Korff[2] support this hypothesis. In the case of non-proportional (i.e., Geiger-Müller) counters the experimental work does not seem to have shown either the absence or presence of the cathode photo-emission with any certainty.

Stever,[3] Wilkening and Kanne,[4] Nawijn,[5] and others have investigated the effects of various barriers, e.g., glass beads sealed on to the anode wire, on the spread of the discharge along the counter tube, following earlier similar work by Greiner[6] on hydrogen and by Curran and Strothers[7] and Swiss workers[8] on various gas mixtures. It is easy to conclude from these and other experiments that the counter discharge spreads much less in the self-quenching type of counter, containing a complex (polyatomic) gas, than in the non-self-quenching (simple gas) type containing, for example, argon or hydrogen. It has even been stated[9] (see the Discussion below) that there was no evidence, in self-quenching counters, for the presence of photons at appreciable distances from the wire, although most authors do not take this extreme view. It was considered worth while to investigate the discharge spread by photo-ionization in counters more thoroughly, with particular emphasis on such simple gases as argon and hydrogen. The first part of the work was on self-quenching mixtures, and the results are reported here since it is hoped they may be of interest to users of counters containing such mixtures. It was thought necessary, for completeness, to mention the work with non-self-quenching gases in the present paper.

II. APPARATUS

Divided cathode, single anode, counters of the type used for other experiments by Stever,[3] Ramsey and Hudspeth,[10] and others were employed for some of the experiments. They consisted of either 6 (Fig. 1) or 12 cathode assemblies, the copper cathodes being 2 inches or 1 inch long, respectively, 0.75 inch internal diameter, spaced $\frac{1}{8}$ inch apart. The anode wires were of 5-mil tungsten. The cathodes were given various chemical treatments (see below); the pumping

[1] S. A. Korff and R. D. Present, Phys. Rev. **65**, 274 (1944).
[2] M. E. Rose and S. A. Korff, Phys. Rev. **59**, 850 (1941).
[3] H. G. Stever, Phys. Rev. **61**, 38 (1942).
[4] M. H. Wilkening and W. R. Kanne, Phys. Rev. **62**, 534 (1942).
[5] A. Nawijn, *Het Gasontladings Mechanisme Van Den Geiger-Müller Teller* (Drukkerij Hoogland-Delft, 1943), pp. 52, 53.
[6] E. Greiner, Zeits. f. Physik **81**, 543 (1933).
[7] S. G. Curran and J. E. Strothers, Proc. Camb. Phil. Soc. **35**, 654 (1939).
[8] F. Alder, E. Baldinger, P. Huber, and F. Metzger, Helv. Phys. Acta **20**, 73 (1947), and references therein to earlier work.
[9] P. Weisz, Phys. Rev. **62**, 477 (1942).
[10] W. E. Ramsey and E. C. Hudspeth, Phys. Rev. **61**, 95 (1942).

technique was not good, but was of the simple nature described and discussed, for example, by Rochester and Janossy[11] and found to be quite satisfactory for the production of reliable self-quenching counters.

Two other types of counter were used. In the first of these (Fig. 2) which contained 4 copper cathodes (above dimensions), the arrangement was as shown. The second new type of counter had 6 cathodes (above dimensions) spaced 0.5 inch apart as in the counter of Fig. 2, with a window (as in Fig. 2) attached to the front of each cathode except the first; it is shown schematically in Fig. 3. Experiments with such window-cathode counters, designed to eliminate the cathode effect by collimating the photon beams to the neighborhood of the wire, do not appear to have been previously described. The device, due to Wilkening and Kanne,[4] of inserting a Bakelite ring in the cathode is only superficially similar. Collimation is not perfect in cylinders 1 and 2 of Fig. 3, since the discharge region in 2 is visible at the cathode of 1. Great care was taken in the lining-up of all the cylinders in the multi-cathode counters.

The method of experiment was to irradiate an extreme cathode by carefully collimated gamma-rays from radium, and to determine the extent of discharge spread by counting coincidences between the irradiated cathode and each of the other cathodes in turn. The voltage on intervening cathodes was reduced to a value just below the starting potential, to avoid effects attributable to spurious charges and to prevent diffusion of electrons along the counter. Many check experiments to assess the importance of these and other effects were made.

The coincidence circuit is shown in Fig. 4. The mixer valves are preceded by SP.41's. The scaler and counting circuit will be described elsewhere. The amplification given by the pulse

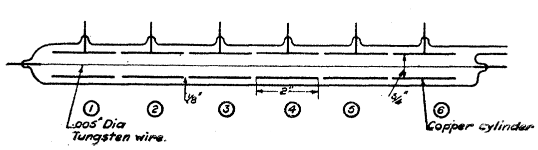

FIG. 1. 6-cylinder counter.

[11] G. D. Rochester and L. Janossy, Phys. Rev. **63**, 52 (1943).

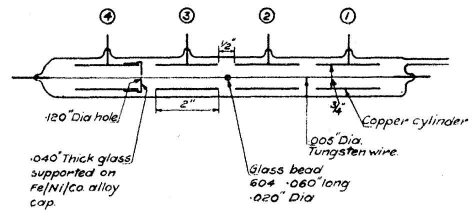

FIG. 2. 4-cylinder counter.

reversing stages of Fig. 4 was valuable since the pulse size, especially from the 1-inch cathodes, was fairly small. When coincidences with non-self-quenching gases were to be counted, the quenching circuit of Fig. 5 was used; it is similar to that of Neher and Harper.[12] It was, of course, also necessary in our case to arrange for independent control of the voltage on each cathode. Careful tests were made to ensure that the coincidence circuits were symmetrical and that both channels were identical in sensitivity.

III. PRELIMINARY TESTS

In order to calculate the counting rate resulting from chance coincidences (cosmic rays, etc.) or those arising from the small amount of radiation passing sideways through the lead collimator block, it was necessary to find the resolving time of the circuit of Fig. 4. The method used by many previous authors was adopted here. Two small separate counters ($2\frac{1}{2}$-inch cathodes) were set up at a distance apart (4 inches) roughly equal to the length of the divided cathode counters. By taking individual counts (N_1N_2), using radium gamma-rays, and the chance coincidence counts in the same conditions of irradiation, etc., the resolving time (τ) may be found from the expression

$$C = 2N_1N_2\tau + K.$$

N_1 and N_2 included the counter background in each case. With the circuit as used for self-quenched counters (Fig. 4), the resolving time with 0.001 μf condensers was about 30 μsec., and with 0.002 μf condensers, about 40 μsec. This could easily have been reduced but was adequate for the present work (see below). The true cosmic-ray coincidence count K may be eliminated by using two sets of data.

[12] H. V. Neher and W. W. Harper, Phys. Rev. **49**, 940 (1936).

It was easy to show that circuit pick-up effects were absent, by reducing the operating voltage of one counter just below its starting potential. This was checked also with two separate counters.

A typical experiment with the two separate counters (A and B) gave the following results:

(a) with no gamma-ray irradiation:

$N_1=52$/min., $N_2=81$/min., $C=0.21$/min.

(b) with gamma-ray irradiation:

$N_1=1238$/min., $N_2=676$/min., $C=0.94$/min.

From this particular experiment τ can be calculated and found to be about 26 /μsec. The effect of the stray irradiation of the cathodes, other than that deliberately irradiated, i.e., the importance of imperfect collimation, will be assessed below, using the values of resolving time deduced above. It would have been undesirable to reduce the resolving time to a value comparable with or less than the time for discharge spread, which would be very small (10^{-8}–10^{-9} sec.) for photon-induced spread. Hill and Dunworth[13] have measured the velocity for discharge spread by electron movement, etc., and find it to be ~10 cm/μsec.

IV. EXPERIMENTAL RESULTS WITH DIVIDED CATHODE COUNTERS

Typical results with a 6-cathode counter (Fig. 1) are shown in Fig. 6 for various self-quenching gases. Even for a case of little discharge spread (1 cm Hg ethyl ether/10 cm Hg argon) there were still 16 discharges spreading per 5 min. over 2 inches of counter with 3000 discharges per 5 min. in the irradiated counter. The curves of Figs. 6 and 7 should be corrected (before absorption coefficients are deduced) for the lack of perfect collimation, but the lack of knowledge of the effective size of the cylinder of radiation in the initiating cylinder makes this a questionable procedure. The results are therefore meant to be comparative only; further data in collimated tubes will be obtained.

The counter conditions for these curves were as follows:—

The operating voltage was generally about 40 volts above the starting voltage. The extent of the stray irradiation from the collimator may be judged by the following representative results, taken with 1 cm Hg ethyl ether/10 cm argon. The counts on the first and third cylinders (2.25 inches apart, i.e., one cathode length+two gap lengths) individually were 499 and 35/minute, and the coincidence rate was 13/minute. The chance coincidence rate, calculated as above from a knowledge of resolving time was 0.02/minute, with a true coincidence count caused by cosmic rays of about 0.2/min. The spurious coincidence rate is therefore negligible even for the most highly quenching mixtures except at the lowest counts (~1/min.). That this is a conservative estimate is shown by the repeated coincidence counts of about 0.5/min. that were obtained for the two extreme cathodes in the tube, despite the presence of the stray irradiation from the second to the last cathode (typical variations in the latter being from 35/min. to 25/min. for the second and last cathodes, respectively, with the first cathode counting at ~500/minute). The discharge invariably spread from the first to the second cylinder (gap $\frac{1}{8}$ inch), see Fig. 1, since field penetration would be appreciable; coincidence readings were therefore only useful from the third cylinder outwards.

Check results were taken also with the 12-cylinder counter, with 1-inch cathodes in order

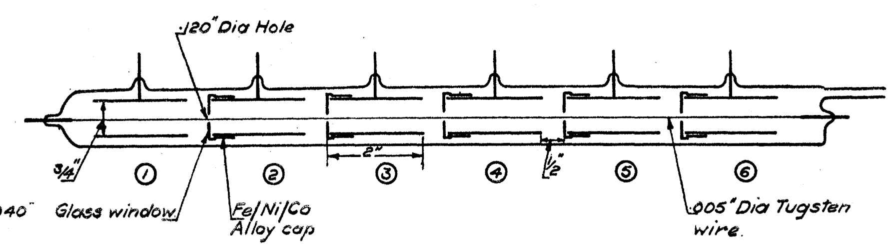

FIG. 3. 6-cylinder window counter.

[13] J. M. Hill and J. V. Dunworth, Nature **158**, 833 (1946).

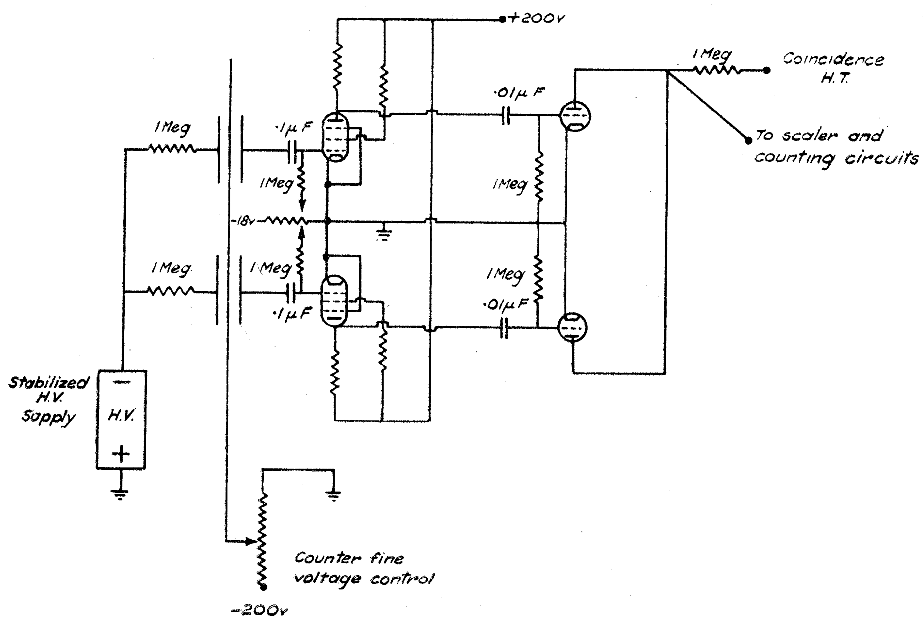

FIG. 4. 2-channel reversing stage and coincidence stage.

to obtain more accurate data for the highly quenched mixtures. Results are shown in Fig. 7 (counter operating data as in Table I).

Experiments made with hydrogen and argon showed no detectable quenching for the full counter length. The results were in accordance with expectations and are only mentioned here for completeness.

Having thus demonstrated the existence of appreciable discharge spread in counters which worked most satisfactorily in self-quenching circuits and showed excellent plateaus (flat to within ±5 percent for 200 volts) it seemed necessary to show the relative importance of cathode (photo-emission) and gas (photo-ionization) effects. Nawijn[14] considered that the discharge spread found in a beaded counter only at high voltages was due entirely to discharge spread over the bead and not to the cathode effect, since at sufficiently low operating voltages no spreading was observed. The supposition was that discharge spread caused by cathode photo-emission could theoretically have taken place at low voltages and its absence at such voltages excluded the possibility of its occurrence at higher voltages. This reasoning seems specious; in our opinion (see Fig. 9) discharge spread attributable to cathode effect is merely less likely at lower than at higher voltages. Before describing special counters made for this purpose it is of interest to quote further experimental results.

The curves of Fig. 8 show the effect on discharge spreading with two types of cathode treatment (i) heavy oxidation obtained by heating in air, and (ii) chemical cleaning with acid and washing with distilled water immediately prior to the pumping and filling. It was found that the clean cathodes gave more spreading (see Discussion, below). (Note (Fig. 8) s.v. ≡starting voltage and w.v.≡working voltage.)

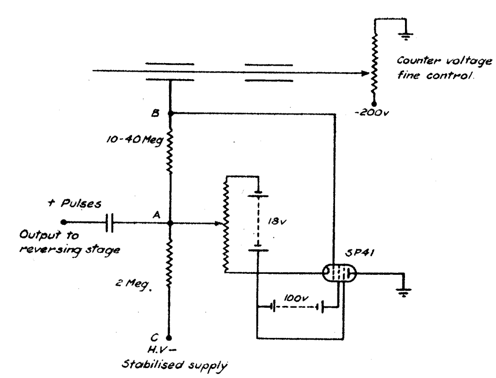

FIG. 5. External quenching circuit.

[14] See reference 5, p. 51.

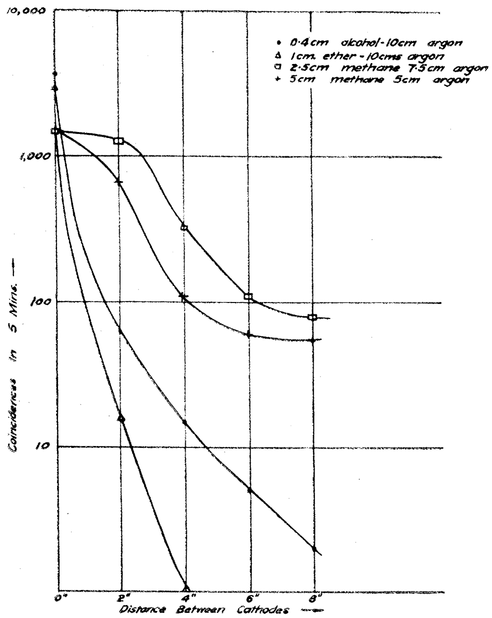

FIG. 6. Spread of discharge in 6-cylinder counter.

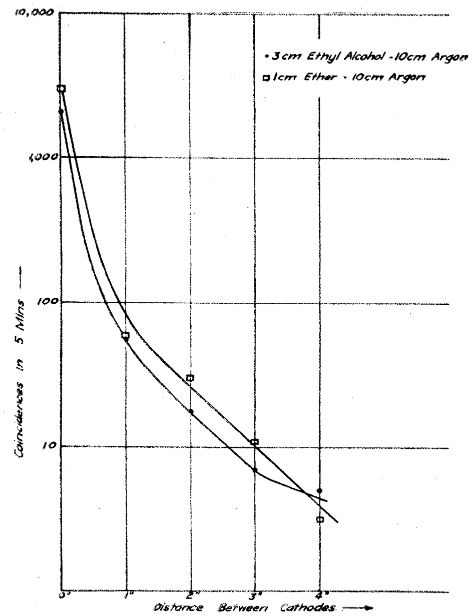

FIG. 7. Spread of discharge in 12-cylinder counter.

The variation of discharge spread (between the first and third cylinders) with counter voltage and pulse size is shown in Figs. 9, 10, and 11. (1 cm Hg ethyl alcohol vapor/10 cm Hg argon.) These are representative curves; the shape of the pulse size/voltage curve has been discussed by Nawijn,[14] and the first part only of it by Korff and Present.[1] Further and more detailed experiments on the significance of these results will be performed.

V. EXPERIMENTAL RESULTS WITH SPECIAL COUNTERS

Some experiments with the 4-cathode counter of Fig. 2 were made with the intention of assessing the relative importance of gas and cathode effects. The cylinders are numbered in Fig. 2, and the mean results are tabulated for brevity (Table II). The irradiated cathode is marked with an asterisk.

It was found, by taking plateau characteristics with cylinder 4, that the presence of the glass window seemed to have no effect on the counter's properties.

Tests with argon and hydrogen (5 cm Hg pressure) showed 100 percent spreading even with cylinders 3 and 4.

The results of Table II may be summarized briefly by stating that the spread of discharge for the simple divided cathode counters is very largely due to emission at the cathodes due to photon bombardment. The discharge spread with the window counter is much less than with the beaded counter (cylinders 2 and 3 of Fig. 2). If the discharge spread with the beaded counter was due entirely to cathode effect, the latter being unaffected by the presence of a bead, then the sum of counts for cylinders 2 and 3, and 3 and 4 (Fig. 2) should equal the number for 1 and 2. That this is not so, in general, is probably due to the above assumptions failing; that they do so to varying degrees in different gas mixtures (Table II) is interesting and, we intend, will be studied further. The effect of a possible amount of electron diffusion between cylinders (Fig. 2) was not ignored; this effect was, we consider,

TABLE I. Counter conditions for measurements with divided cathode counters.

Gas mixture	Starting voltage approx.
0.4 cm Hg alcohol/10 cm Hg argon	750
1 cm Hg ether/10 cm Hg argon	1130
2.5 cm Hg methane/7.5 cm Hg argon	1370
5 cm Hg methane/5 cm Hg argon	1500
3 cm Hg alcohol/10 cm Hg argon	1320

satisfactorily eliminated in the other divided cylinder counters (Figs. 1 and 3).

To check the effect of windows further and to obtain more accurate results on discharge spread through photo-ionization, the counter with 5 windows and 1 normal cathode was used. The alignment of the 5 windows was carried out with great care, and checked before the wire was inserted and the tube sealed at the ends. Results with a mixture of 0.5 cm Hg ethyl alcohol/10 cm Hg argon (starting voltage 760), which was known to give relatively poor quenching, gave at 800 volts 15 coincidences in 5 min. between cylinders 1 and 2 counting at 751 and 44/min., respectively. Between cylinders 1 and 3 (756 and 49/min.) there were no coincidences in 5 min., while there would have been about 50–60 coincidences in that time with the cathode effect operative (Figs. 6 and 7). It therefore seemed safe to conclude that the discharge spread in the latter figures was due almost entirely to the cathode effect in self-quenching gases. As a check, some tests were made with a mixture of 2.5 cm Hg methane/7.5 cm Hg argon, since this had given the largest amount of discharge spread in the previous experiments (Fig. 6). The starting voltage was 1300, and at 1340 volts there were 1402 coincidences in 5 minutes between cylinders 1 and 2, counting at 798 and 317/min., respectively. Between cylinders 1 and 3 there were 3 coincidences in 5 minutes, showing again that gas quenching is most efficient, with cylinders 1 and 3 counting at 789 and 42/min. Absorption curves for methane (10 cm Hg) and with ethyl alcohol/argon (0.4 cm Hg/10 cm Hg) taken with the counter of Fig. 3 are shown in Fig. 12, and these curves should represent the absorption of photo-ionizing radiations from the irradiated end cylinder. It is possible to deduce the true absorption coefficient for the operative photons from Fig. 12 by allowing (inverse square law) for the lack of perfect collimation. Thus the curve shown in Fig. 12 (crosses) is derived, and it is seen that the observed absorption effect cannot be due to the inverse square fall-off of radiation. It should be emphasized that the absorption even in methane is so large that the coefficient cannot accurately be derived from Fig. 12. Further experiments will be performed with another counter tube incorporating a collimation chamber.

VI. DISCUSSION OF RESULTS

It has, apparently, generally been supposed that discharge spreading in self-quenching counters by virtue of photo-ionization in the gas, or by liberation of electrons from the counter cathode due to the incidence on it of photons, is negligible or indeed absent. For example, Collie and Roaf[15] used a double counter built for β-ray spectrographic work and reported that the number of coincidences (12/min.) barely exceeded the number (10/min.) obtained with a separate pair of counters. They conclude, since the passage of electrons between counters might account in their case for this difference, that the photo-effect at the cathode is much too small to have any influence on discharge maintenance.

Stever[3] carried out exhaustive tests on divided counters of various types but did not use a window counter (such as that shown in Fig. 3).

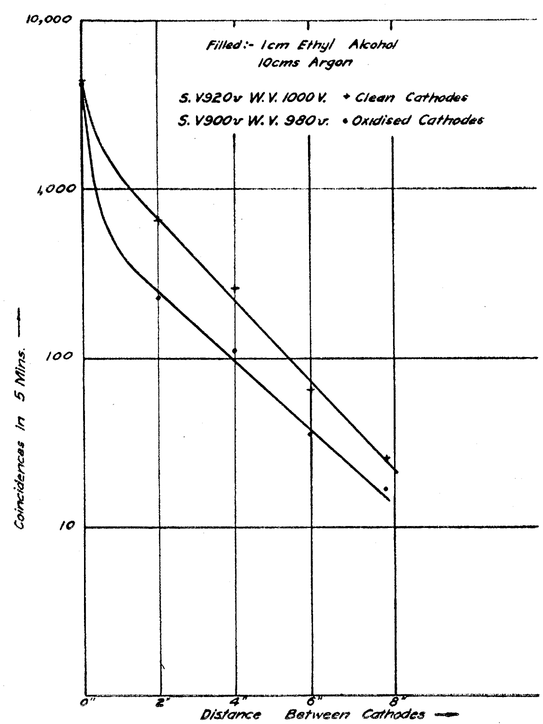

FIG. 8. Effect of cathode treatment on discharge spreading in 6-cylinder counter.

[15] C. H. Collie and D. Roaf, Proc. Phys. Soc. 52, 186 (1940).

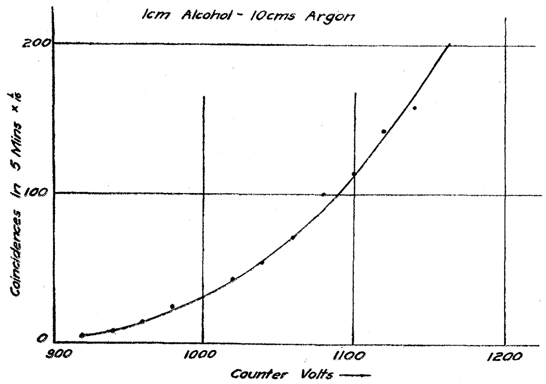

FIG. 9. Coincidence rate as function of counter voltage.

He found no spreading with a $\frac{1}{8}$-inch diameter bead, $\frac{1}{4}$ inch long, on the wire, but he apparently used the NO_2 treatment of the copper cathodes which is known to produce a surface of high work function. With a smaller bead at a lower counter pressure the discharge could be made to spread; it is not certain, however, that even then the discharge spread was due purely to photo-ionization in the gas, since a cathode mechanism might have become operative with the lower photon absorption given by the lower gas pressure.

Wilkening and Kanne[4] have published (as have Ramsey[16] and others) similar results and Korff and Present,[1] discussing their results in the light of the comprehensive and masterly theory of Korff on fast counter action, stress also that fast counters showed 100 percent localization. This is misleading, and indeed Korff and Present state that with, for example, a 50 percent/50 percent argon/methane mixture there is appreciable photo-effect. Rose and Korff[2] have, in a well-known paper, shown the presence of a cathode effect in certain fast gas mixtures used in proportional counters. However, such a counter would work in a self-quenching arrangement, and it therefore seems that the existence of an appreciable cathode effect is not a serious objection in a fast non-proportional counter. We believe this to be reasonable since the essential part of the fast counter mechanism is that first explained by Korff,[1] namely, the absence of secondary emission from bombardment of the cathode by the positive-ion sheath in such a counter; this seems to be quite explicable on qualitative but sound physical grounds.

Weisz[8] stated that no evidence has been found for the presence of photons at appreciable distances from the wire. This may be the case, with cathodes of very high work function, but Weisz's statement appears to us to be incorrect, since no qualification is made.

Korff[1] appears to dismiss rather summarily the presence of photons incapable of absorption in the complex constituent in, say, an alcohol/argon counter. The present experiments certainly seem to support that hypothesis since the discharge spread is almost zero (Fig. 6) for quite short, but appreciable ($\sim$10 cm), distances in suitable gas mixtures.

Objections have been made that the presence of appreciable photo-emission at the cathode is inconsistent with Simpson's[17] pulse-reversal method of shortening dead times by attracting the positive-ion sheath into the wire, since the continued emission of electrons would prevent the production of a short clearing time. It is

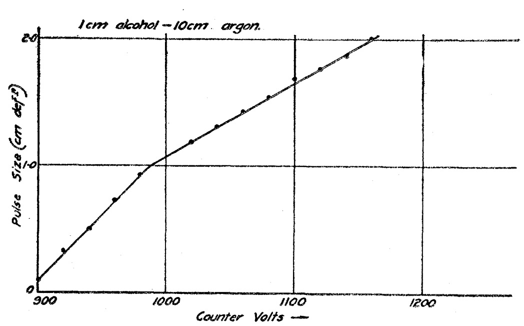

FIG. 10. Variation of pulse size with counter voltage.

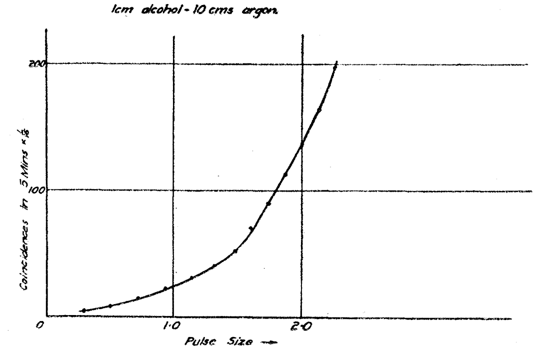

FIG. 11. Variation of coincidence rate with pulse size.

[16] W. E. Ramsey, Phys. Rev. 61, 96 (1942).

[17] J. A. Simpson, Phys. Rev. 66, 39 (1944).

TABLE II. Measurements on the importance of gas and cathode effects.

Gas filling 10 cm Hg argon with	Starting voltage	Working voltage	Counting rates with cathode (per min.) 1	2	3	4	Coincidence rate (per min.) $\frac{1}{2}$	$\frac{2}{3}$	$\frac{3}{4}$
0.4 cm Hg ethyl alcohol	approx. 840	860	48	596*	—	—	81	—	—
		900	57	633	—	—	174	—	—
		860	—	52	702*	—	—	73	—
		900	—	48	707*	—	—	155	—
		860	—	—	710*	55	—	—	3
		900	—	—	707*	51	—	—	34
1.0 cm Hg ethyl alcohol	approx. 980	1000	59	803*	—	—	42	—	—
		1120	96	852*	—	—	145	—	—
		1000	—	46	899*	—	—	17	—
		1120	—	122	923*	—	—	70	—
		1000	—	—	928*	50	—	—	1
		1120	—	—	1038*	65	—	—	5
10 cm Hg methane (no argon)	2080	2140	73	716*	—	—	213	—	—
			—	36	586*	—	—	82	—
			—	—	586*	31	—	—	7

clear, however, that the existence of the reverse field entirely removes this objection and Simpson[17] indicates that photo-emission could only arise at the wire in the period during which the latter is made negative. The present experiments obviously, for that and other reasons, are not invalidated by Simpson's work.

It seemed surprising that the work function of our copper cathodes could be so high as to make them respond only to radiations absorbable in the fast counter fillings. The continuous absorption in ethyl alcohol vapor and in methane begins roughly at about 1700 and 1450A,[1] respectively, and if photons of longer wave-lengths were present in the counter discharge (so that there would be little or no absorption of them in the gas filling) then presumably the fall-off in spreading with distance (Figs. 6 and 7) would be much less than is found and would then be governed by an inverse-square law. The presence of appreciable absorption seems to suggest that the cathode work function is at least 8.5 volts

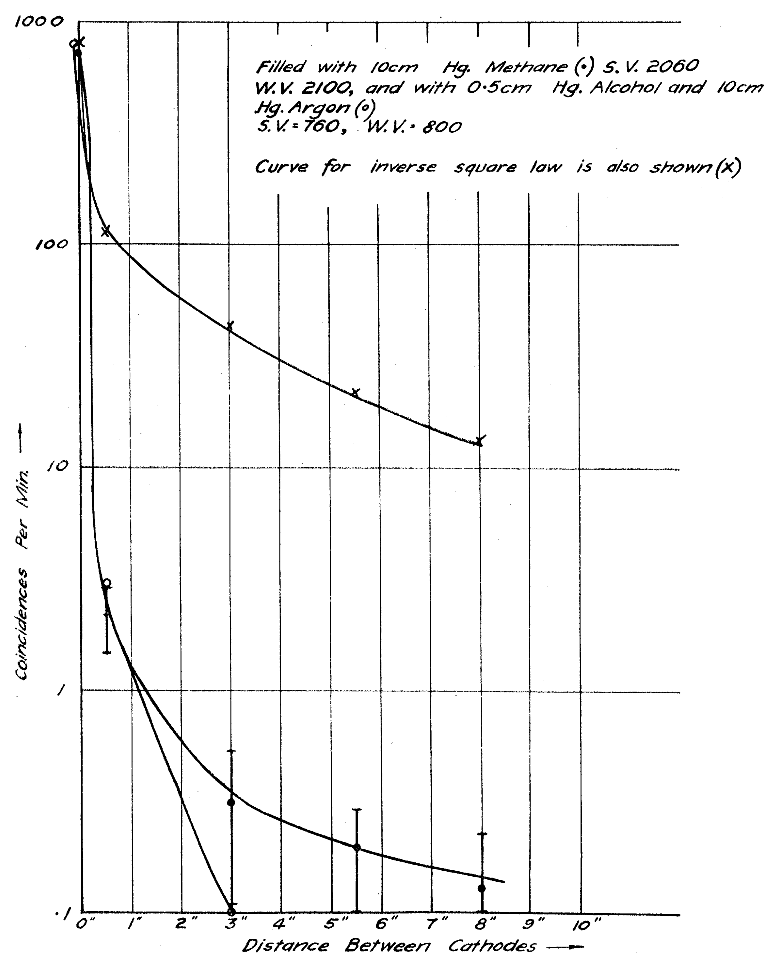

FIG. 12. Absorption curves for methane taken with the six-cylinder counter of Fig. 3.

(corresponding to $\lambda_{max}=1450A$). Apparently little data exist on the work function of oxidized copper; the work function of clean copper is 4.4 v[18] and, by analogy with the work on oxidation of Pt and Ag[19] which produced almost a doubling of the work function, a value for copper of 8.5 volts seems possible and reasonable.

Attempts made to show the difference between fairly clean and oxidized copper by a rough vacuum technique which was, however, as good as that often used in the preparation of satisfactory counters,[10] were made, and the results showed that copper cleaned as well as possible in our apparatus had apparently a higher work function and was therefore probably better oxidized in the physical, but not chemical sense, than the grossly oxidized cathodes we prepared. It is intended to carry out further experiments with a greatly improved vacuum technique, but this deficiency of knowledge does not seriously affect the main conclusions of this paper, i.e., that counters working very satisfactorily in normal self-quenching arrangements show considerable discharge spread which is mainly due to the cathode photo-emission effect. The results of Figs. 6, 7, and 8 will therefore not be accurately reproducible; the data of Fig. 12 should have more fundamental significance.

[18] A. L. Hughes and L. A. Du Bridge, *Photoelectric Phenomena* (McGraw-Hill Book Company, Inc., New York, 1932), p. 75.

[19] J. H. de Boer, *Electron Emission and Adsorption Phenomena* (Cambridge University Press, Teddington, England, 1935), pp. 148–151.

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The Ionization and Dissociation of Formic Acid Monomer by Electron Impact*

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In HCOOH vapor in a mass spectrometer, low velocity electrons have produced the ionized molecule, one negative and twelve positive fragment ions. For the negative ion and six positive ions the mode of production has not been established. The other fragment ions originate in the reactions requiring the least dissociation energy for the production of the corresponding radicals, although for one of these a more energetic reaction seems preferred at high electron energies. One of the ions of doubtful origin also probably originates in a reaction involving more than the minimum degree of dissociation. The first appearance potential of four ions with established reactions, of one ion of doubtful origin, and probably of another of doubtful origin, as well as the second appearance potential of CO_2^+, all indicate that the corresponding reactions involve no more than 1.0 volt excess energy. Both HCO^+ and $COOH^+$ utilize the trivalency of O^+ in their structure. $HCOOH^+$, $COOH^+$, and HCO^+ seem to have a second appearance potential close to the first. To explain this, and to make the electron impact and spectroscopic ionization potentials of the molecule compatible, it is assumed that a *trans*-form of HCOOH exists. Estimates of its abundance relative to the known *cis*-form give for the energy difference between the two forms the probable limits 1.2–3.0 kcal/mole.

I. INTRODUCTION

IN the electron-impact studies of the more than forty molecules already reported in the literature,†† the most useful information has been obtained by application of the mass spectrometer to the determination of appearance

* The data on which this paper is based were obtained prior to November, 1941; but circumstances beyond our control have delayed preparation of the paper. A preliminary presentation of data was made at a recent A.P.S. meeting (see Phys. Rev. **71**, 139(A), 1947).

** This work is a summary of Part II of a thesis presented to the Faculty of Princeton University in partial fulfillment of the requirements for the degree of Doctor of Philosophy.

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†† See. H. D. Smyth, Rev. Mod. Phys. **3**, 347 (1931) for a review of work to that date, and J. A. Hipple, J. App. Phys. **13**, 551 (1942) for more recent references.