

SCINTILLATION SPECTROMETER STATISTICS

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1. INTRODUCTION

It was realized early in the development of scintillation counters that the size of an observed pulse is closely proportional to the energy of the registered particle (KALLMANN, 1949; HOYT, 1949; JORDAN and BELL, 1949). From this fact has evolved the familiar technique of scintillation spectrometry which is today one of the most powerful tools of experimental and applied physics.

Whenever the method shall be used—whether in a measurement of particle energy, investigation of luminescence characteristics, coincidence work, or mere particle detection—it is well to inquire beforehand into the expected response of the spectrometer. The formation of a pulse is indeed sufficiently complicated

to leave room for many disturbing side effects. It starts with the loss of energy to the phosphor by some charged particle (Compton electron, recoil proton, incident α -particle, etc.). Most of the resulting excitation is quickly degraded into heat; the remaining part (20% in the most efficient known phosphors) is stored by fluorescence centres or metastable energy levels and eventually reappears in the form of light. Some of the emitted quanta are reabsorbed in the phosphor, others trapped within its boundaries by total reflexion; the rest (a fraction e , say) escape into the optical system which guides them towards the photocathode of the electron multiplier tube. On the way there, some more are lost by absorption and reflexion, and of the fraction f which arrive at the cathode only a few (a fraction p ; about one in a dozen) release a photoelectron. Again, some photoelectrons (up to 50%) are lost between the cathode and the first secondary emission dynode through imperfect electron-optical focusing; the remaining ones (a fraction c , say) finally initiate the secondary emission avalanches whose last "daughter generations" accumulate on the multiplier anode and add up to the observed output pulse. Fig. 1 symbolizes this sequence of steps in a "block diagram."

Every stage in this process, and hence also the size of the output pulse, is governed by a law of chance and varies from one particle to the next. Consequently, the scintillation spectrometer renders an incident "monochromatic line" in the form of a broad pulse-height distribution which we shall call, for convenience, the "instrumental line." The experimenter should know both shape and width of the latter, and allow for them properly in the design and use of his spectrometer; failure to do so may lead to serious error (cf. the discrepancies between various results listed 1953 by ROBERTS).

In this article the pulse formation, instrumental width and line shape in a scintillation spectrometer are discussed with particular attention to the technical details and requirements of experimental work. Although many authors (WOODWARD, 1948; MORTON and MITCHELL, 1948, 1949; HOYT, 1949; MORTON, 1949; SAUTER, 1949; SEITZ and MUELLER, 1950; SCHARDT and BERNSTEIN, 1951; MAIGNAN, BLANC and DETOBUF, 1952; SWANK and BUCK, 1952; GARLICK and WRIGHT, 1952; WRIGHT, 1952; ROBERTS, 1953; CAMPBELL and BOYLE, 1953; GODLOVE and WADEY, 1954) have dealt with isolated features of this subject no comprehensive treatment has yet been published. The present survey is based largely on unpublished work by the author and aims at closing this gap by providing a firm statistical basis for design and development.

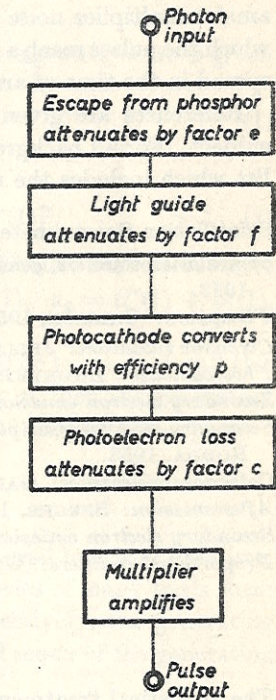


Fig. 1. Symbolical block scheme of pulse formation.

Most of the problems pertaining to the mere counting of scintillations or to fast coincidence experiments have been fully discussed in the literature and will not be touched here. Adequate information is available on the questions of absolute counting efficiency for γ -rays (SEITZ and MUELLER, 1950; MAIGNAN, BLANC and DETOEUF, 1952; MAEDER and WINTERSTEIGER, 1952a, b, c; MAEDER, MÜLLER and WINTERSTEIGER, 1954; LIDÉN and STARFELT, 1954) and soft electrons (BUTT, 1953), the detection of small, slow scintillations amidst multiplier noise (HERBERT, 1952), the statistical spread in the time at which the pulses reach a given height (POST and SCHIFF, 1950), and the statistical spread in the time of arrival of the photons at the cathode (POST, 1952b).

References are given only to papers which have a direct bearing on the subject. For all background information the reader is referred to the following list which includes the most useful and up-to-date sources as of March, 1954:

Scintillation Spectrometer Statistics—

Scintillation counters, general: HANLE, 1951; BIRKS, 1953; CURRAN, 1953; KREBS, 1953.

Phosphors: GARLICK, 1952.

Čerenkov radiation: JELLEY, 1953.

Photocathodes: ZWORYKIN and RAMBERG, 1949; SOMMER, 1951.

Secondary electron emission: MCKAY, 1948; BRUINING, 1954.

Secondary electron multipliers: FENGSTROM, 1947; MORTON, 1949; SCHAEFFLI, 1951; RODDA, 1953.

Cathodoluminescence: GARLICK, 1950.

Afteremission: SEEGER, 1953.

Secondary electron emission by ions: MASSEY and BURHOP, 1952.

Proportional counters: WILKINSON, 1950; WEST, 1953.

2. GENERATING FUNCTIONS

The statistical treatment of the pulse formation becomes very simple through the use of probability generating functions. They were introduced by LAPLACE in 1812 but did not find much favour with his followers (cf. PRICE, 1928); during the last few years they were, however, revived and applied successfully to the statistical problems of counting (KOSTEN, 1943; RAMAKRISHNAN and MATHEWS, 1953), neutron density in a reactor (COURANT and WALLACE, 1947; FRISCH and LITTLER, 1954), gas amplification (FRISCH, 1948), radioactive chains (JORGENSEN, 1948), scintillation counters (WOODWARD, 1948; MORTON, 1949; SAUTER, 1949; SEITZ and MUELLER, 1950; POST and SCHIFF, 1950; MAIGNAN, BLANC and DETOEUF, 1952) and nucleon cascades (JÁNOSSY, 1950).

Unfortunately, there exists no comprehensive monograph on generating functions. Various interesting theorems are scattered throughout the papers quoted above and many textbooks (e.g. CZUBER, 1914; FOWLER, 1929, 1936; KAMKE, 1932; USPENSKY, 1937; AITKEN, 1944; CRAMÉR, 1946; ARLEY and BUCH, 1950). The elementary theory, which we shall need here, was summed up and exemplified by FRISCH (1948) and JORGENSEN (1948). All essential formulae are recapitulated in the following three paragraphs; the proofs are

so simple that the reader will have no difficulty to supply them himself, should he have no access to the papers by FRISCH or JORGENSEN.

If a statistical event, such as the release of several secondary electrons by one primary, yields integral scores $s = 0, 1, 2, \dots$ whose respective probabilities of appearance in one trial are p_0, p_1, p_2, \dots its generating function is defined by means of an auxiliary variable x as

$$G(x) = \sum_s p_s x^s.$$

By definition we must have $G(1) = 1$; the average, or expected, score is given by

$$\bar{s} = G'(1), \quad (1)$$

and the variance (squared standard deviation) by

$$\sigma^2 = \bar{s}^2 - \bar{s}^2 = G''(1) + G'(1) - [G'(1)]^2. \quad (2)$$

The primes denote differentiation with respect to x . Similar expressions give the moments $\alpha_k = \bar{s}^k$ about the origin: $\alpha_1 = G'(1)$, $\alpha_2 = G''(1) + G'(1)$, $\alpha_3 = G'''(1) + 3G''(1) + G'(1)$ and so forth.

As an example consider an event which scores either 0 or 1; we shall call this a "head-tail" event. Let "tails" come with probability p , then the generating function is $(1 - p) + px$, the average score p , and the variance $p(1 - p)$.

3. ALTERNATIVE EVENTS

If an event is of such a composite nature that in each trial one, and only one, of several basic events A, B, \dots happens, and if in a series of many trials these alternatives occur with frequencies q_A, q_B, \dots respectively, the generating function for the composite event is simply the weighted mean of the generating functions $A(x), B(x), \dots$ for the alternatives:

$$G(x) = q_A A(x) + q_B B(x) + \dots \quad (3)$$

The score expected in one trial and its variance can then be calculated by applying (1) and (2).

For instance, let all the alternatives be of the head-tail type, with respective probabilities p_1, p_2, \dots for "tails." Their generating functions $(1 - p_i) + p_i x$ combine into another linear function of the form $(1 - p) + px$, where now $p = \sum q_i p_i$; thus the alternatives combine into another head-tail event of probability p for "tails."

This sounds quite trivial but has important applications. A photon incident on the photocathode can either release a photoelectron ("tail") or not ("head"). However, the photoelectric sensitivity of the cathode varies with photon energy and is not, in general, the same at all points of its surface.* Hence the

* Of course we are concerned here with actual local inhomogeneities, such as exhibited clearly in the experiments of RAYCHMAN (1939), MARSHALL, COLTMAN and BENNETT (1948), GODLOVE and WADSWORTH (1954) and LINDEN (1954), and not with the inhomogeneity simulated by the uneven efficacy of photoelectron collection from different cathode regions.

photoelectric conversion of incident polychromatic light is an event of the composite head-tail type which, as the example shows, is described completely by the average conversion efficiency, or effective cathode sensitivity, p .

The same holds *mutatis mutandis* for the light collection efficiency. Here "tail" means that a photon, after escape from the scintillator, actually arrives at the cathode. The chance of arrival will depend on the number of reflexions which the photon has to undergo, and on other similar factors, but its average value (called f in Fig. 1) suffices for the statistical description of light collection.

Of course, these statements must be taken *cum grano salis*. Since the light collection efficiency depends on the geometry of the ray paths, its average f must depend on the angular distribution with which the light emerges from the phosphor, for it is this distribution which determines the weight factors q_i in (3). Thus a change of crystal shape, or mere roughening of its surface, can alter f . Especially in Čerenkov counters (e.g. MARSHALL, 1952; BASSI and BIANCHI, 1952) or shower counters (HOFSTADTER, 1953) this effect could be quite marked on account of the highly directional light emission. Similarly, the illumination of the cathode by the scintillations will not in general be uniform, so that a simple rotation of the multiplier can alter p if the cathode is inhomogeneous. For analogous reasons we must expect changes of p as well as of f when the particles under observation are made to impinge on different regions of the phosphor. The strong dependence of both p and f on the luminescence spectrum will be obvious.

In brief, p and f are by no means constants of the given light collector and multiplier, but depend on the luminescence spectrum, shape and optical behaviour of the phosphor as well as on the point of origin of the scintillations within the phosphor. This is of particular importance when comparative measurements on different phosphors are made in a standard setup. Their results will be free from gross uncertainties only if (a) f is almost 100% so that it cannot change appreciably when the phosphors are exchanged, (b) the cathode is illuminated very uniformly and always over the same area, and (c) its spectral response is accurately corrected for.

4. SIMULTANEOUS EVENTS

Let two independent events with generating functions $A(x)$ and $B(x)$ happen at the same time, and add their scores. The generating function for the combined score is easily shown to be

$$G(x) = A(x)B(x).$$

From (1) and (2) we find that $\bar{s} = \bar{s}_A + \bar{s}_B$ and $\sigma^2 = \sigma_A^2 + \sigma_B^2$. Induction immediately gives for the expected total score of an arbitrary number of simultaneous events

$$\bar{s} = \sum \bar{s}_i, \quad (4)$$

and for the variance

$$\sigma^2 = \sum \sigma_i^2. \quad (5)$$

As an example, take Z simultaneous head-tail events with the same probability p for "tails." The generating function for the total score is $[(1-p) + px]^Z$ and has the coefficients

$$p_s = \binom{Z}{s} (1-p)^{Z-s} p^s. \quad (6)$$

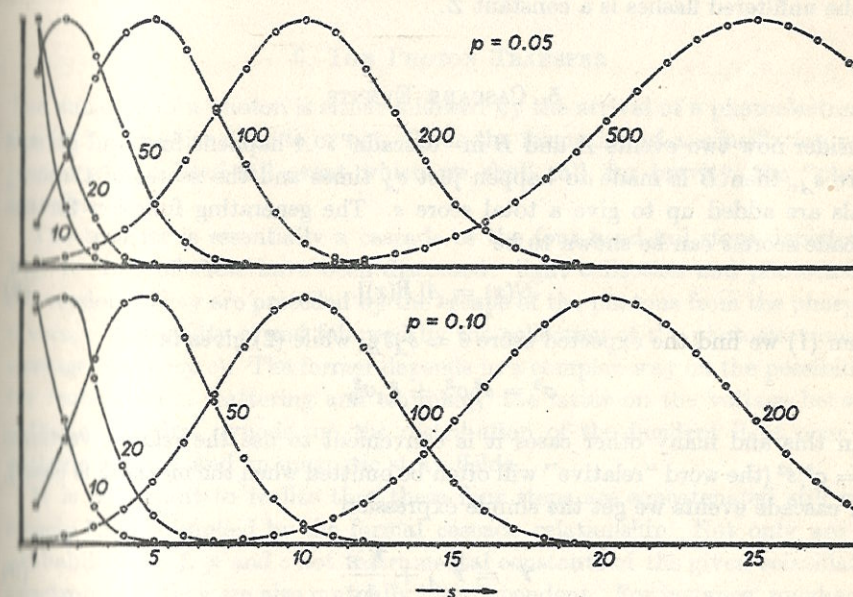


Fig. 2. Binomial distribution for $p = 5\%$ and $p = 10\%$ and several representative values of Z . The distributions are given by the little circles; the connecting smooth curves have only been drawn in to facilitate inspection. Normalization is to unit maximum height, not to unit area.

So the score is distributed binomially; it has an average pZ and a variance $p(1-p)Z$, as found from (4) and (5). We recall that for large pZ , and s near pZ , (6) is approximated closely by the well-known deMoivre gaussian

$$p_s = \frac{1}{\sigma\sqrt{2\pi}} e^{-\frac{(s-pZ)^2}{2\sigma^2}} \quad (7)$$

where again $\sigma^2 = p(1-p)Z$.

In Fig. 2 the distribution (6) is plotted for several values of p and Z which seem representative for our case. These curves give directly the probability distribution of the number of photoelectrons released from a photocathode of effective sensitivity p by a group of Z incident photons. They show clearly that the rather small sensitivity of the cathodes available at present substantially limits the resolving power of the scintillation spectrometer.

Note also how large the chances are that a group of photons releases no photoelectrons at all. According to (6) this chance is $p_0 = (1-p)^Z$ and will be negligible only for $Z > 100$ or so.

We meet the binomial distribution in yet another context. Multipliers are often tested with short light flashes from some standard lamp that have been attenuated in a dark filter. It is then assumed that the number of photons in the attenuated flashes is constant. This is incorrect; the numbers are distributed binomially and therefore have a variance $p(1-p)Z$, if we call the filter transmission p and make the simplifying assumption that the number of photons in the unfiltered flashes is a constant Z .

5. CASCADE EVENTS

Consider now two events A and B in "cascade": A happens first and gives a score s_A , then B is made to happen just s_A times and the scores of these s_A trials are added up to give a total score s . The generating function for the cascade score s can be shown to be

$$G(x) = A[B(x)]. \quad (8)$$

From (1) we find the expected score $\bar{s} = \bar{s}_A \bar{s}_B$, while (2) gives first

$$\sigma^2 = \bar{s}_B^2 \sigma_A^2 + \bar{s}_A \sigma_B^2.$$

In this and many other cases it is convenient to use the *relative* variance $\mathcal{V} = \sigma^2/\bar{s}^2$ (the word "relative" will often be omitted when the meaning is clear). For cascade events we get the simple expression

$$\mathcal{V} = \mathcal{V}_A + \frac{\mathcal{V}_B}{\bar{s}_A}. \quad (9)$$

Induction readily yields for a cascade of n events:

$$\bar{s} = \bar{s}_1 \bar{s}_2 \dots \bar{s}_n, \quad (10)$$

$$\mathcal{V} = \mathcal{V}_1 + \frac{\mathcal{V}_2}{\bar{s}_1} + \dots + \frac{\mathcal{V}_n}{\bar{s}_1 \bar{s}_2 \dots \bar{s}_{n-1}}. \quad (11)$$

An elementary derivation of the important formula (11) can be found in the appendix to a paper by SHOCKLEY and PIERCE (1938).

Once more head-tail events are a convenient example. Let there be but two of them, with probabilities f and p for "tails." With (8) comes $G(x) = (1 - fp) + fpx$, hence the cascade behaves like a single head-tail event of probability fp for "tails." This applies in particular to the cascade of light collection and photoelectric conversion.

Another example is furnished by attenuated light flashes when the number Z of photons in the unfiltered flashes is subject to statistical fluctuations. Calling s the number of photons in an attenuated flash, we have of course $\bar{s} = p\bar{Z}$, whereas from (9)

$$\mathcal{V}_s = \mathcal{V}_Z + \frac{p(1-p)}{p^2 \bar{Z}} = \left(\mathcal{V}_Z - \frac{1}{\bar{Z}} \right) + \frac{1}{p \bar{Z}}.$$

If Z is distributed normally (i.e. according to Poisson's law) only the last term on the right remains; if the distribution is of a different character, this term will outweigh the parenthesis provided the transmission p is very low. Since $1/p\bar{Z} = 1/\bar{s}$ is precisely the relative variance of s which would result if s was normally distributed, we can say that a strong filter always delivers flashes whose sizes are distributed with normal variance.

6. THE PHOTON TRANSFER

The emission of a photon is either followed by the arrival of a photoelectron at the first multiplier dynode or not. Thus the formation of a scintillation pulse begins with a head-tail event which we shall call, for brevity, the "photon transfer."

The transfer is essentially a cascade of the four head-tail steps depicted in Fig. 1. Two of these have been discussed: light collection and photoelectric conversion. They are preceded by the escape of the photons from the phosphor (average probability e) and followed by the collection of the photoelectrons (of average efficiency c). The former depends in a complex way on the possibilities for reabsorption, scattering and trapping; the latter on the voltage between cathode and first dynode, on the distribution of the incident light over the cathode surface, and on magnetic stray fields.

It is important to realize that these four steps are concatenated still more closely than is implied by the formal cascade relationship. Not only are the probabilities e , f , p and c not instrumental constants of the given scintillation spectrometer, they are also mutually interdependent. For instance, roughening the phosphor will increase e but may at the same time reduce f if it permits the escape of light into directions which are unfavourable to safe collection. Further interdependencies will be caused by all chromatic dispersions, local inhomogeneities and optical anisotropies existing in the given setup. This will be taken up in more detail in § 11; until then we assume that there is a *constant* probability $efpc = T$ for the successful transfer of a photon from the phosphor onto the first dynode, without inquiring about possible statistical consequences of any interrelations.

Exact calculation of T is of course impossible under these circumstances. For design considerations and estimates of T the literature provides useful material on trapping (SHURCLIFF and JONES, 1949; GILLETTE, 1950), light collector optics (MARSHALL, COLTMAN and BENNETT, 1948; SCHARDT and BERNSTEIN, 1951; GARWIN, 1952; GRAVES and DAVIS, 1953) and the optical transmission of light guide materials (TIMMERHAUS, GILLER, DUFFIELD and DRICKAMER, 1950). Reliable values for the *absolute* yield of photocathodes do not exist except for the EMI cathode (BUTT, 1953; his Fig. 2). In connexion with cathodes it should be remembered that the chromatic response differs considerably among tubes of the same type (see SCHAEFFER and BAUMGARTNER, 1951a, ENGSTROM, STODENHEIMER and GLOVER, 1952), that the response curve shifts towards the ultraviolet on cooling (KELLEY and GOODRICH, 1950;

SANGSTER, 1951; SCHAETTI and BAUMGARTNER, 1951a, and observations by the author on EMI tubes), and that illumination enhances the sensitivity in the red (SCHAETTI and BAUMGARTNER, 1952; SCHAETTI, BAUMGARTNER and FLURY, 1953). For cathode behaviour under heavy duty see the references given in § 13.7.

7. THE SECONDARY ELECTRON CASCADE

The photoelectrons which arrive at the first dynode are subsequently multiplied in the familiar secondary emission avalanches. We shall deal with the multiplication of a whole group of electrons later and consider first the formation of an avalanche by one single photoelectron.

7.1 General

Throughout the n stages the multiplication proceeds formally according to the law that the incidence of one electron on a dynode (say the i -th) entails the incidence of some secondaries (say m_i) on the next dynode. We shall call \bar{m}_i the gain, and the corresponding relative variance \mathcal{V}_i the gain variance, of dynode number i . Then the number G of secondaries arriving at the multiplier anode due to the incidence of one electron at the first dynode is simply the final score of a cascade event of n stages which is easily treated with the formulae developed in § 5. The gain of the whole multiplier (tube gain) is

$$\bar{G} = \bar{m}_1 \bar{m}_2 \dots \bar{m}_n \quad (12)$$

and the (relative) tube gain variance

$$\mathcal{V}_G = \mathcal{V}_1 + \frac{\mathcal{V}_2}{\bar{m}_1} + \dots + \frac{\mathcal{V}_n}{\bar{m}_1 \bar{m}_2 \dots \bar{m}_{n-1}}. \quad (13)$$

If all stages operate at the same gain \bar{m} and the same gain variance \mathcal{V} (13) becomes

$$\mathcal{V}_G = \mathcal{V} \frac{\bar{m}^n - 1}{(\bar{m} - 1)\bar{m}^{n-1}}$$

or, on account of $\bar{m}^n \gg 1$,

$$\mathcal{V}_G = \mathcal{V} \frac{\bar{m}}{\bar{m} - 1}. \quad (14)$$

Note that this does not depend on the number of stages, and that the tube gain variance is only $\bar{m}/(\bar{m} - 1)$ times the gain variance of the first stage.

In practice, one always runs the first dynode at a somewhat higher voltage in order to increase the photoelectron collection efficiency c . For this case one obtains from (9) and (14) in obvious notation

$$\mathcal{V}_G = \mathcal{V}_1 + \mathcal{V} \frac{m}{\bar{m}_1(\bar{m} - 1)}. \quad (15)$$

If it is further assumed that the stage gain has normal variance, i.e. $\mathcal{V}_i = 1/\bar{m}_i$, as for a Poisson distribution, (14) and (15) reduce to

$$\mathcal{V}_G = \frac{1}{\bar{m} - 1} \psi \text{ resp. } \psi \mathcal{V}_G = \frac{\bar{m}}{\bar{m}_1(\bar{m} - 1)}. \quad (16a, 16b)$$

Eq. (16a) was first obtained by MORTON (1949).

7.2 The stage gain variance

The assumption of normal stage gain variance is, however, as questionable as it is tempting. Indeed, it is known from studies of secondary emission that the number of secondaries released by one primary has supranormal variance even at fairly low primary energies. This has been demonstrated for BaO + SrO surfaces (HAYNER, 1935; ZIEGLER, 1936), metallic Ni and Be (KURRELMAYER and HAYNER, 1937), and was also reported for Be-Cu alloys (MAYNE, 1952). At primary energies of 15 kev or more, very strong departures from normality are observed with almost any substance (MCMULLAN, 1953).

The reason for this behaviour must be sought in the fact (reviewed, for example, by GARLICK, 1950) that the primary on penetrating into the emitter can liberate secondaries all along its range, while only those liberated sufficiently close to the surface are able to escape into the vacuum. Furthermore, in making a secondary the primary loses energy (10–15 ev, according to recent (1952) results of JACOBS, MARTIN and BRAND) and therefore range; hence the liberation of one secondary near the surface is likely to be accompanied by the liberation of more secondaries near the surface. These effects collaborate to favour the escape of secondaries in little groups so that the distribution of the number of secondaries due to one primary has a long tail and corresponding supranormal variance. When the primary energy is increased this tail becomes more prominent; at high energies (near and beyond the maximum emission yield) the distribution will be further broadened by the appearance of tertiaries.

At very low primary energies, on the other hand, all secondaries are liberated so close to the surface that they have almost equal chances of escape. One would here expect a certain analogy between secondary emission and ionization straggling (FANO, 1947): since the liberation of a secondary consumes a comparatively large fraction of the primary's energy it prejudices the liberation of others, and then the number of secondaries must be distributed with subnormal variance. Indeed, if for energetic reasons at most one secondary can be produced the variance is that for a head-tail event, i.e. $\delta(1 - \delta)$, which is smaller than the normal variance, i.e. δ (by δ we denote as usual the secondary emission ratio).

A correct description of secondary emission phenomena requires of course the use of wave mechanics so that this picture is certainly oversimplified; it nevertheless deserves some confidence in view of the remarkable success of classical concepts in this field (cf. the theory of secondary emission by JONEER, 1952).

An experimental example (calculated from measurements by ZIEGLER, 1936) is given in Fig. 3. Up to a secondary emission ratio of $\delta = 1.5$ the variance in the number of secondaries is a little less than normal ($\sigma^2 < \delta$), then it increases approximately like $\delta + 0.11\delta^2$; in other words, the relative variance first is less than $1/\delta$ but later increases like $1/\delta + 0.11\delta$. At high energies, when the maximum yield is approached, the departure from normality becomes still stronger. Certainly this isolated instance does not imply that the secondary emission variance increases all that rapidly for every emitter, nor that the transition from subnormal to supranormal variance always occurs at a δ as low

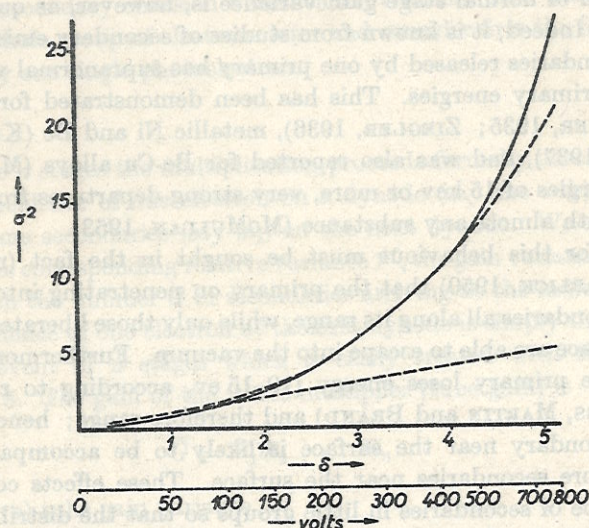


Fig. 3. Secondary emission variance for activated BaO + SrO (calculated from data given by ZIEGLER, 1936). Broken straight line: $\sigma^2 = \delta$ (normal variance), broken curve: $\sigma^2 = \delta + 0.11\delta^2$.

as 1.5; but it qualitatively bears out the above conclusions, namely that such a transition does in fact occur and that at high primary energies the variance becomes very high (the latter point is confirmed well by the results of KURREL-MAYER and HAYNER, 1937, and of McMULLAN, 1953).

Returning to the stage gain, we must anticipate that there are always some secondaries which do not hit the sensitive area of their target dynode; in non-focusing multipliers the loss of secondaries is actually quite considerable (30% in the venetian blind type manufactured by EMI, according to SOMMER and TURK, 1950). Hence the stage gain is certainly smaller than the δ at which the dynodes operate; the stage gain variance, however, should not differ much from the relative secondary emission variance of the dynodes.

7.3 The tube gain variance

Thus the formulae (16) are based on a rather unrealistic assumption. Whether they can be applied will depend entirely on the secondary emission variance

which the usual dynode materials display under the operating conditions prevailing in scintillation spectrometric work. It is of some importance to settle this question by reference to experimental results because (16a) has been used frequently and uncritically.

The tube gain variance can be measured directly by permitting a very weak flux of electrons to fall on the first multiplier dynode. All observed output

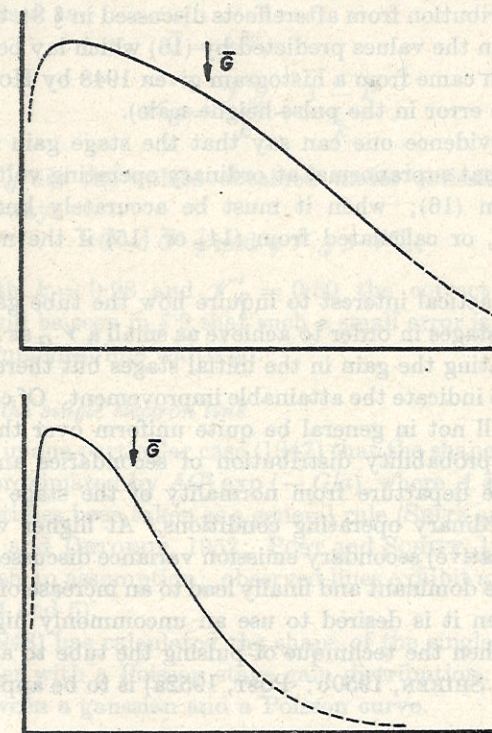


Fig. 4. Single electron lines (counts per pulse height interval vs. pulse height) in a 12-stage particle-counting multiplier with Be-Cu dynodes (from bias curves given by ALLEN, 1947). Voltages were 500 V on the first stage and 375 V on all others. The arrows indicate the mean pulse height, i.e. the tube gain.

- (a) Immediately after dynode activation.
- (b) After numerous exposures to air.

pulses are then due to single electron avalanches; hence the observed pulse-height spectrum represents the probability distribution of G and yields \mathcal{V}_G through a simple computation.

Fig. 4 shows two typical "single electron lines" obtained in this way by ALLEN (1947) in a particle-counting multiplier with Be-Cu dynodes. Right after activation of the surfaces \mathcal{V}_G was 0.44; after numerous exposures to air it had increased to 0.51. The stage gain was 3.85 in the former case and 3.70 in the latter (and slightly more for the first stage). It follows that here the tube gain variance is more sensitive to the history of the dynodes than the stage gain, and

that the value predicted by (16) is definitely too small (less than 0.35 in either case).

In photomultipliers one can produce initial single electrons by exposing the cathode to a weak flux of light. From data thus obtained by several workers with various tubes operated under normal conditions (with stage gains of 4-5) the author has calculated values of the tube gain variance ranging from 0.15 to 0.55. These figures are not entirely representative for \mathcal{V}_G because they comprise the unknown contribution from aftereffects discussed in § 8; they were however always greater than the values predicted by (16) which lay between 0.2 and 0.3 (the only exception came from a histogram given 1948 by HOYT which looks as if there was a zero error in the pulse-height scale).

Collecting the evidence one can say that the stage gain variance of usual dynodes is somewhat supranormal at ordinary operating voltages. \mathcal{V}_G cannot be calculated from (16); when it must be accurately known it should be measured directly, or calculated from (14) or (15) if the necessary data are available.

It is of some practical interest to inquire how the tube gain should be distributed over the stages in order to achieve as small a \mathcal{V}_G as possible. Eq. (13) suggests concentrating the gain in the initial stages but there exists no experimental evidence to indicate the attainable improvement. Of course the response of the dynodes will not in general be quite uniform over their surfaces; this will broaden the probability distribution of secondaries and may be partly responsible for the departure from normality of the stage gain variance, as observed under ordinary operating conditions. At higher voltages, however, the increase of (relative) secondary emission variance discussed in the preceding section will become dominant and finally lead to an increase of \mathcal{V}_G . This should be considered when it is desired to use an uncommonly high voltage on the first dynode, or when the technique of pulsing the tube to a very high overall voltage (POST and SHIREN, 1950b; POST, 1952a) is to be applied.

7.4 Interruptions of the cascade

There is an appreciable chance that no secondaries are released when a primary hits a dynode. For instance, if the stage gain is 4 and exhibits a Poisson distribution, the probability for such a blank is $e^{-4} = 0.018$. So the cascade sometimes does not start, or it breaks down at a later stage as WOODWARD has discussed in detail (1948). An important consequence is that the detection efficiency of the multiplier for the photoelectrons is not strictly 100%.

Corresponding to the photoelectron blanks the probability distribution of G has an infinitely sharp spike at $G = 0$ which is, of course, not observed in the simple experiment described in the preceding section. Hence the tube gain variance calculated directly from the experimental single electron line is in error. As will be shown briefly now, the true value lies slightly higher.

The omission of blanks results in a wrong normalization of the single electron line to unit area: all ordinates p_G are divided by $p_1 + p_2 + \dots$ instead of by $p_0 + p_1 + p_2 + \dots$. The error is compensated for by multiplying the first

and second moments of the observed line with

$$\frac{p_1 + p_2 + \dots}{p_0 + p_1 + p_2 + \dots} = k$$

(the photoelectron detection efficiency of the multiplier). The true values of mean and variance then follow immediately from the corrected moments and are readily expressed as

$$\bar{G} = k\bar{G}', \quad (17)$$

$$\mathcal{V}_G = \frac{\mathcal{V}_G'}{k} + \frac{1-k}{k} \quad (18)$$

where \bar{G}' and \mathcal{V}_G' are the values obtained under omission of blanks. One recognizes that always

$$\bar{G} < \bar{G}' \text{ and } \mathcal{V}_G > \mathcal{V}_G';$$

for instance, with $k = 0.98$ and $\mathcal{V}_G' = 0.50$ the correct variance becomes $\mathcal{V}_G = 0.53$. It will be seen in § 9 that such a small error is negligible as far as questions of instrumental line width go.

7.5 The shape of the single electron line

ENGSTROM found in one particular case (1947) that the shape of a single electron line is closely approximated by $AG^2 \exp(-G/a)$, where A and a are constants, and this has sometimes been taken as a general rule (SEITZ and MUELLER, 1950; MAIGNAN, BLANC and DETOEUF, 1952; POST and SCHIFF, 1950). The evidence speaks against such an assumption; observed lines exhibit a variety of different shapes (cf. Figs. 4 and 5).

WOODWARD (1948) has calculated the shape of the single electron line in an idealized multiplier with a Poisson stage gain distribution; he finds that it is intermediate between a gaussian and a Poisson curve.

7.6 The duration of the single electron pulse

A figure of great practical interest is the length Δt of the time interval during which the last daughter generation of a single electron avalanche arrives at the multiplier anode. In focusing tubes Δt is very short indeed because secondary emission is an instantaneous phenomenon.* So all members of the first avalanche generation are born simultaneously, then travel together to the second dynode where they produce the next generation, also simultaneously, and so on until the last generation arrives at the anode. The times required for the transit between dynodes add up to a total of perhaps $t = 20 \text{ m}\mu \text{ sec.}$ in a typical case; still, the last generation hits the anode almost *en bloc*, with only a small straggling in the times of arrival of its members which is due to slight differences in total path length (and therefore total transit time) of different genealogical branches of the avalanche.

* This point has recently (1952) been doubted by LAW, but his experiments are certainly in error. (cf. POST (1952a), and DIEMER and JONKER (1953).)

In non-focusing dynode structures (such as the venetian blind and box types) the electron trajectories are not neatly bunched, and many secondaries are released at points where the electrostatic fields are very weak; thus both the differences between the total path lengths and the differences between the dynode-to-dynode transit times are much greater and lead to a fairly large Δt . In any case Δt depends of course on the linear dimensions of the dynode structure and on the applied voltages.

The values of Δt in the more important tube types when operated under normal conditions are summarized below:

- (i) *RCA and Mazda tubes of the 931A type* (circular RAYCHMAN structure, see RAYCHMAN and SNYDER, 1940). SARD has calculated (1946) the probability distribution of the total transit time of a genealogical branch and finds that it is gaussian with a half-width of 0.6 μ sec. The pulse observed in the external recording circuit should be just a little longer because the electrons induce charges at the anode before they have actually reached it. POST and SHIREN (1950b) and LUNDBY (1950) found experimentally that $\Delta t < 1 \mu$ sec.
- (ii) *Straight Raychman structures* (see ZWORYKIN and RAYCHMAN, 1939) such as used in most particle-counting multipliers (e.g. ALLEN, 1947; BAXTER, 1949) and manufactured by SCHAEFFER (cf. 1952, 1953b): ca. 10 μ sec. (MEYER, BALDINGER and HUBER, 1950).
- (iii) *The RCA 4646* (L-16 structure by RAYCHMAN, see STONE, 1949): $\Delta t < 5 \mu$ sec. (MORTON, 1952; GREENBLATT, GREEN, DAVISON and MORTON, 1952).
- (iv) *EMI tubes* (LALLEMAND's venetian blind structure, 1948): 30 μ sec. was reported by OWEN (1951); data by ALLEN and ENGELDER (1951) suggest 10 μ sec. at very high tube voltages.
- (v) *DuMont tubes* (box structure by LARSEN and SALIGER, 1940): no data available yet; 10 μ sec. or so may be expected.

One should guard against the belief that these figures set lower limits to the resolving time of coincidence experiments with the various multiplier types. By proper techniques one can undercut Δt considerably; BENEDETTI and RICHINGS (1952), for example, have made it likely that intervals of 0.1 μ sec. can be measured with 1P21 tubes (see also POST, 1952b). To be sure, such refinements calls for tubes of outstanding quality which are entirely free from aftereffects (see §§ 8 and 13.5).

7.7 Space charge effects

Since Δt is so small the current represented by the passage of a single electron avalanche through the multiplier can be fairly large at the later stages; e.g. in a RCA 5819 operating at a gain of 5.10^6 the anode current during a single electron pulse is about 1 mA. The corresponding space charges between the dynodes could be dense enough to limit the size of the avalanche by shielding off the electrostatic fields and causing loss of secondaries. In this situation our formalism does not apply any more because the secondary emission "scores"

are prevented from adding up; so it becomes necessary to investigate this matter further.

At what magnitudes of current the space charges begin to exert limiting actions in a given multiplier will depend very much on its electron-optical properties and on the applied voltages. Danger values for the anode current, at which the departure from linearity of the response characteristic reaches 3% under normal operating conditions, are listed below for the more important tube types:

- (i) *931A and similar tubes*: 10 mA (ENGSTROM, 1947).
- (ii) *Straight Raychman tubes*: 25 mA (SCHAEFFER, 1951).
- (iii) *RCA 4646*: 200 mA (GREENBLATT *et al.* 1952).
- (iv) *EMI tubes of the new design* which has a flat last dynode and a grid-like anode between this dynode and its predecessor: 2 mA (author). *EMI tubes of the old design*: 0.1 mA (concluded from RAFFLE and ROBBINS, 1952).
- (v) *DuMont tubes*: 20 mA (extrapolated from LINDEN, 1953).

The space-charge saturated currents are 5–10 times stronger. The critical values have been given for the anode current because of purely practical reasons; it is not implied that the limiting charges appear in front of the anode. In fact, in circular Raychman structures the limitations occur (ENGSTROM, 1947; POST, 1952a) between the last but one and the last, in the RCA 4646 between (GREENBLATT *et al.* 1952) the 14th and 15th, and in the new EMI 6262 between the 12th and 13th dynodes.

Comparison of the anode currents from a single electron avalanche in the various multiplier types with the corresponding danger values then shows that single electron pulses are practically never affected by space charge. If in an exceptional case limitations should occur they will be only slight. The statistical consequences of this eventuality are immediately clear: by opposing larger avalanches, weak space charges tend to compress the long tail of the single electron line and thereby decrease both \bar{G} and \mathcal{V}_G a little.

Another effect of space charges is that they lengthen Δt (MORTON, 1949). Their action during scintillation pulses will be discussed in § 13.4.

8. AFTEREFFECTS

The performance of photomultipliers is complicated by the fact that the formation of a single electron avalanche is always accompanied by a number of parasitic phenomena which lead to the delayed emission of electrons from the photocathode and from the dynodes. There we have the PAETOW effect (1939), due to the illumination of the cathode by incident light, and of the dynodes by light generated during secondary emission; the TANAKA effect (1940), due to the electron bombardment of the dynodes; the MALTER effect (1936) and its variants (see TREY, 1943), that is, field emission induced by positive surface charges on cathode or dynodes; then the possibilities that positive ions produced in the residual gas slowly drift back to the preceding electrode and release

secondary electrons on arriving there ("ion feedback"), that negative ions do the same in the opposite direction, and finally, that quanta from the de-excitation of the residual gas and quanta generated during secondary emission by cathodoluminescence travel back to the cathode and release fresh photoelectrons ("light feedback").

All the unwanted latecomers liberated by these various mechanisms then give rise to spurious pulses whose sizes will be widely different according to their starting points in the multiplier, and whose delays with respect to the initial pulse cover a very wide range too. The time lag of ion feedback and of negative ion multiplication is of the order 1 μ sec., corresponding to the time-of-flight of the ions; light feedback is delayed by about 20 μ sec. for an analogous reason; but the other aftereffects can liberate electrons with any delay up to 10 μ sec. or more. Such afterpulses are extremely annoying in coincidence experiments, or when very fast scalers are used.

The aftereffects associated with a single electron avalanche, though mentioned by GODFREY, HARRISON and KEUFFEL (1951), DAVISON (1952) and LANTER and CORWIN (1952, contains photographs), have not yet been investigated systematically. All relevant evidence comes from observations on large scintillation pulses; see § 13.5 on "Satellites." Ion feedback between first dynode and cathode is quite common; Paetow effect is well known to occur on the cathode. Feedback of light or ions from the back end of the multiplier structure to the cathode is prevented in carefully designed tubes by appropriate internal shields. Afteremission from, and ion feedback between, the dynodes seems to be relatively unimportant. In several EMI tubes of medium quality the author has observed occasional noise pulses of exceptional height (corresponding to more than 50 photoelectrons) and rather long duration (0.2–0.3 μ sec.); the cause may be Malter effect on the cathode, but these events are too rare to warrant detailed consideration here.

The residual gas present in the multiplier envelope is not only responsible for the occurrence of ion feedback; it also takes part in all genuine afteremission processes, their cause being essentially a phase readjustment (or "chemisorption, intermediate between mere physical readorption of gas and a true chemical reaction) in the uppermost layer of the emitting surfaces (see SEEGER, 1953). Thus the tube vacuum is a factor of great importance. The outgassing procedures employed by the manufacturers at present are not really adequate; all tubes exhibit afterpulsing to a considerable extent, variations from tube to tube are great, and the gas is often absorbed and desorbed at random during an experiment. Tubes with a bad vacuum are unsuitable for most precision work; they are easily spotted by the strong dark current which they exhibit, frequently together with a high gain, under standard testing conditions. Sometimes such a specimen can be cured by exposing the cathode to broad daylight for a few seconds with the full voltage on (private communication from B. RIDLEY; verified by the author; see also POST, 1952a); this brutal treatment presumably heats the dynodes up to just the right gettering point.

The statistical consequences of afterpulsing are as complex as its causes;

we therefore attempt only a qualitative orientation. The most prominent afterpulses obviously come from the cathode, so we neglect for simplicity all dynode effects and assume that at the cathode the release of one genuine photoelectron is followed by the release of A afterelectrons. The collecting time constant at the multiplier anode (total anode-to-earth capacity times the leak resistor) shall be so large that initial and delayed pulses are added up; the total number of collected electrons will be called M . Then we have a case of simultaneous events in the sense of § 4. The tube gain including aftereffects is clearly

$$\bar{M} = (1 + \bar{A})\bar{G}. \quad (19)$$

To obtain the tube gain variance we must add (see eq. 5) the absolute variances of the genuine and the afterpulse; a short calculation yields in terms of relative variances:

$$\mathcal{V}_M = \frac{\mathcal{V}_G}{1 + \bar{A}} + \mathcal{V}_A \frac{\bar{A}^2}{(1 + \bar{A})^2}. \quad (20)$$

This result shows somewhat surprisingly that \mathcal{V}_M need not be greater than \mathcal{V}_G . The reason is, of course, that the fluctuations in the genuine and in the after-pulse can to some extent cancel each other. Numerically, \bar{A} will be small whereas values of \mathcal{V}_A as large as 2 seem quite possible. Under these circumstances \mathcal{V}_M is not much different from \mathcal{V}_G ,

and can be both smaller or greater. Thus one cannot predict in which way aftereffects will deform the single electron line.

\bar{A} increases rapidly with the voltage applied between cathode and first dynode; in fact the feedback through ions soon becomes regenerative ($\bar{A} > 1$) and causes a continuous discharge (ENGSTROM, 1947; POST and SHIREN, 1960b; POST, 1952a). This is very unfortunate, because it makes it impossible with any but the very best tubes to operate the first dynode at a voltage high enough for complete photoelectron collection. Similar troubles tend to arise when the tube gain is made variable by an adjustable overall tube voltage across a potentiometer chain with fixedappings which supply the dynode

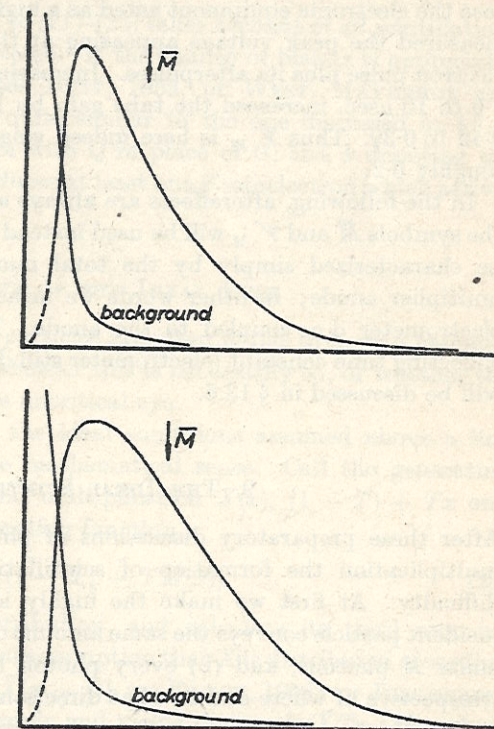


Fig. 5. Single electron lines in an EMI 6262 tube of fair quality. Voltages were 250 V on first stage well as on anode, 125 V on all other stages. Tube gain ca. $4 \cdot 10^8$. Note the higher background pulses in (b).

- (a) Collecting time constant 1.5 μ sec.
(b) Collecting time constant 10 μ sec.

potentials. In such an arrangement it is best to avoid the danger of breakdown by replacing the resistor across the first stage with a little neon bulb (ROBERTS, 1953; cf. also HEMPSON, 1953).

In counting experiments aftereffects can often be rendered innocuous by the introduction of a deadtime. A short collecting time constant sometimes suppresses them in spectrometric work. Fig. 5 provides an illustration. In this case the electronic equipment acted as a high-bandwidth valve voltmeter which measured the peak voltage appearing at the multiplier anode due to a single electron pulse plus its afterpulses. Increasing the collecting time constant from 1.5 to 10 μ sec. increased the tube gain by 18% and reduced its variance from 0.42 to 0.39. Thus \mathcal{V}_M is here indeed smaller than \mathcal{V}_G , and \bar{A} seems to be roughly 0.2.

In the following, aftereffects are always assumed to be present and therefore the symbols \bar{M} and \mathcal{V}_M will be used instead of \bar{G} and \mathcal{V}_G . The output pulse will be characterized simply by the total number of electrons delivered at the multiplier anode; in other words we measure the pulse height with an ideal electrometer d.c. coupled to the anode. The more realistic case of a small collecting time constant (electrometer still d.c. coupled, but with a leak resistor) will be discussed in § 13.6.

9. THE IDEAL SCINTILLATION LINE

After these preparatory discussions of photon transfer and of photoelectron multiplication the formation of scintillation pulses can be treated without difficulty. At first we make the highly idealized assumptions that (a) each incident particle conveys the same amount of energy to the phosphor which then emits X photons, and (b) every photon has the same chance T of transfer irrespective of where and in what direction it is emitted.

The events emission, transfer and multiplication form obviously a cascade of three. Call Q the total number of electrons arriving at the anode due to one scintillation; from (10) follows the mean pulse height

$$\bar{Q} = \bar{X} T \bar{M}, \quad (21)$$

and from (11) the scintillation line variance

$$\mathcal{V}_Q = \mathcal{V}_X + \frac{T(1-T)}{T^2 \bar{X}} + \frac{\mathcal{V}_M}{\bar{X} T} = \left(\mathcal{V}_X - \frac{1}{\bar{X}} \right) + \frac{1 + \mathcal{V}_M}{\bar{X} T}. \quad (22)$$

A formula equivalent to (22) has been given 1952 by GARLICK and WRIGHT; they substitute however $\mathcal{V}_M = 1/(\bar{m} - 1)$ and this should not be done for the reasons outlined in § 7.3. It is best to regard \mathcal{V}_M as an instrumental parameter which must be adjusted for every particular case, although it is seen that a few hundredths of difference in \mathcal{V}_M do not make an appreciable difference to \mathcal{V}_Q .

If the luminescence mechanism in the phosphor is such that $\mathcal{V}_X = 1/\bar{X}$

("normal scintillation variance") the parenthesis in (22) drops out and

$$\mathcal{V}_Q = \frac{1 + \mathcal{V}_M}{\bar{X} T}. \quad (23)$$

This formula* holds also if a light source plus filter is used to simulate scintillations (see § 5).

Note that the formalism leading to (21)–(23) takes account of *all* scintillations including those which are not detected. If the chance of blanks is appreciable (very small $\bar{X} T$; for examples see BUTT, 1953, or WEST, MEYERHOF and HOFSTADTER, 1951) the case is quite similar to the one discussed in § 7.4; equations (17) and (18) apply here with Q in place of G , and k denoting the probability that a scintillation produces at least one photoelectron which arrives at the first dynode.

10. THE SHAPE OF THE IDEAL LINE

Scintillation lines from monoergic particles are often found to have a gaussian shape. There arises the question whether this is necessarily so, or whether the lines merely appear gaussian to the uncritical eye.

It is readily shown that under the ideal conditions assumed above a line cannot strictly be gaussian in the mathematical sense. Call the generating functions for emission, transfer and multiplication $X(x)$, $(1 - T) + Tx$ and $M(x)$, respectively, set up the generating function

$$Q(x) = X[(1 - T) + TM(x)] \quad (24)$$

for the resultant pulse-height distribution, and calculate its third moment about the mean. With the additional assumption that X is distributed according to a Poisson law (then $X(x) = \exp[\bar{X}(x - 1)]$, see FRISCH, 1948; or JORGENSEN, 1948) the algebra becomes quite simple and yields the result $\bar{X} T \alpha_3(M)$, where $\alpha_3(M)$ denotes the third moment of the single electron line about the origin. Since this cannot vanish the scintillation line must be skew and so is certainly not gaussian.

Still, the skewness will be small, and the line very nearly gaussian in the vicinity of its peak, provided the mean number $\bar{X} T$ of effective photoelectrons is large. To see this, we first remember that the Poisson distribution of X becomes gaussian for high \bar{X} . Next, if $\bar{X} T$ is large too, the distribution of the number of photoelectrons for a fixed number Z of photons will be the deMoivre gaussian (7) with p replaced by T . And lastly, a fixed number s of photoelectrons must always give a gaussian pulse-height distribution of relative variance \mathcal{V}_M/s on account of the central limit theorem of statistics. So the shape of the scintillation line results through "smearing out" one gaussian with two

* When a secondary electron multiplier is employed as a current amplifier the shot noise in the anode current is just $1 + \mathcal{V}_G$ times the shot noise in the input current (ZIEGLER, 1936; ZWORYKIN, MORTON and MALTER, 1936; SHOCKLEY and PIERCE, 1938; WILLIAMS, 1938; HARTMANN and RUTHER, 1941). There is an obvious analogy between this theorem and eq. (23), but the two cases should not be confused: in scintillation work the multiplier amplifies a charge, not a current.

other gaussians. As is well known, this operation leads again to a gaussian; hence the observed line shape. A slight amount of skewness is created in our case by the fact that the two smearing gaussians are not really of constant width over the whole smearing interval. For the same reason we must not expect that the *maximum* of the pulse-height distribution coincides with its *mean*. A more refined discussion of this point shows indeed that the maximum is displaced by an amount of the order $\bar{M}/2$ towards the right of the mean; in other words there is a relative peak shift of the order $+1/(2\bar{X}\bar{T})$. Both skewness and peak shift will obviously be imperceptible if $\bar{X}\bar{T}$ is sufficiently large.

Thus lines of high $\bar{X}\bar{T}$ observed under sufficiently ideal conditions exhibit to all intents and purposes a gaussian shape. What happens at low $\bar{X}\bar{T}$ becomes clear on inspection of Fig. 2 (then the central limit theorem does not apply either). Under non-ideal conditions a line need not be gaussian at all; see § 14.

A useful relation exists between the relative full width of a gaussian line at half-maximum (usually called the "half-width" η) and the line variance \mathcal{V}_Q . The absolute width being $2\sqrt{2\ln 2}$ times the standard deviation, one has $\eta^2 = 5.56\mathcal{V}_Q$. A numerical example: 1 Mev-electrons in stilbene, transfer efficiency 8% and tube gain variance 0.40; assuming a yield of 1 photon per 75 ev we have $\mathcal{V}_Q = 0.0013$, $\eta = 0.085$ and the absolute half-width of the line is 85 kev.

11. VARYING TRANSFER EFFICIENCY

Successive scintillations never occur at exactly the same points inside the phosphor and the photons therefore have different chances of escape even from a perfect crystal, as was first pointed out by BELCHER (1950b). Speaking less specifically, we must expect the transfer efficiency to vary from one scintillation to the next. This fact is of great practical importance because it will unavoidably lead to line broadening; so we must now drop the ideal assumption (b) of § 9.

The correct treatment requires that the transfer efficiencies be averaged in the sense of § 3, not over many scintillations, but over the photons of each individual scintillation. The individual averages T_i thus obtained will occur with respective frequencies q_i in a series of many scintillations; the distribution of output pulse heights is then the outcome of alternative events and has the generating function

$$Q(x) = \sum_i q_i X[(1 - T_i) + T_i M(x)] \quad (25)$$

whose members are, of course, all of the form (24).

It follows immediately that $\bar{Q} = \bar{X}\bar{T}\bar{M}$, where $\bar{T} = \sum q_i T_i$. To get the line variance one first has to calculate $Q''(x)$ and then finds from (2) after a few simplifications

$$\mathcal{V}_Q = \mathcal{V}_T + (1 + \mathcal{V}_T) \left(\mathcal{V}_X - \frac{1}{\bar{X}} \right) + \frac{1 + \mathcal{V}_M}{\bar{X}\bar{T}} \quad (26)$$

where \mathcal{V}_T denotes the relative variance of the distribution of the T_i 's. In the case of normal scintillation variance (26) reduces to

$$\mathcal{V}_Q = \mathcal{V}_T + \frac{1 + \mathcal{V}_M}{\bar{X}\bar{T}}. \quad (27)$$

An experimental test of this relation for a given spectrometer would be to determine \mathcal{V}_Q at different particle energies and to plot the measured values

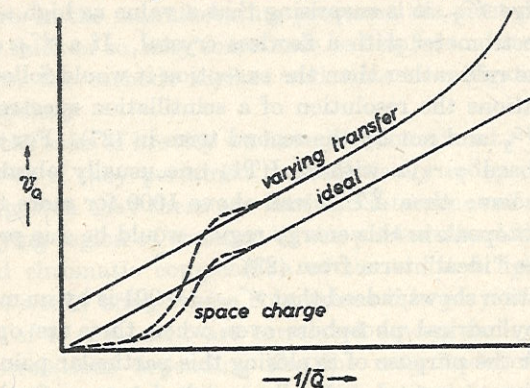


Fig. 6. Scintillation line variance and its dependence on pulse height under different conditions.

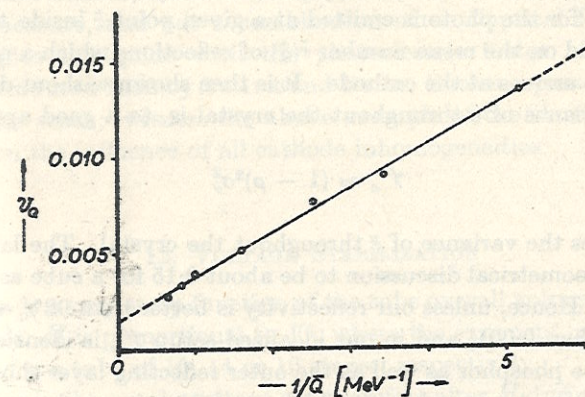


Fig. 7. Scintillation line variance for monoenergetic electrons incident on a cylindrical anthracene crystal of 13 mm thickness (calculated from data by HOPKINS, 1951).

against $1/\bar{X}\bar{T}$ (which is of course proportional to the reciprocal mean pulse height). In the ideal case one obtains a straight line through the origin, as predicted by (23), in the present case a curve lying higher than that straight line by \mathcal{V}_T . Fig. 6 depicts the situation.

Only one set of such data has been published (HOPKINS, 1951). They are reproduced in Fig. 7; the points lie on a straight line which, if extrapolated to infinite pulse height, indicates $\mathcal{V}_T = 0.0015$. Similar measurements have been made by SCHARDT and BERNSTEIN (1951) with γ -rays in NaI, but unfortunately

their drawings do not lend themselves to the re-evaluation necessary for comparison with (27). Some additional information comes from the work of GARLICK and WRIGHT (1952, their Fig. 4). These authors investigated the response of several different phosphors to the same monoenergetic α -particles and compared the line variances; it appears from their results that in a crystal with flaws \mathcal{V}_T can be greater than 0.01.

While it seems quite plausible that optical imperfections of the phosphor should cause a large \mathcal{V}_T , it is surprising that a value as high as 0.0015 should be found in a spectrometer with a flawless crystal. If a \mathcal{V}_T of this order of magnitude was the rule rather than the exception it would follow that in many important applications the resolution of a scintillation spectrometer is determined solely by \mathcal{V}_T and not by the second term in (27). For example, in the spectroscopy of hard γ -rays with NaI(Tl) one usually obtains at least one photoelectron per kev; thus $\bar{X}\bar{T}$ is well above 1000 for more than 1 Mev and the width of a photopeak in this energy region would be due predominantly to \mathcal{V}_T and not to the "ideal" term from (23).

A rough calculation shows indeed that $\mathcal{V}_T = 0.001$ is by no means impossible with cubical or cylindrical phosphors even when they are optically perfect. Let us assume, for the purpose of exploring this particular point, that we have a perfectly transparent crystal, optically matched to a perfectly homogeneous photocathode, and with its front and side faces processed in such a way that they behave like mirrors of (isotropic) reflectivity ρ . The average escape probability $e(\xi)$ for the photons emitted at a given point ξ inside the phosphor depends on ρ and on the mean number $\bar{r}(\xi)$ of reflections which a photon has to undergo until it arrives at the cathode. It is then shown without difficulty that the relative variance of e throughout the crystal is, to a good approximation,

$$\mathcal{V}_e \approx (1 - \rho)^2 \sigma_{\bar{r}}^2$$

where $\sigma_{\bar{r}}^2$ denotes the variance of \bar{r} throughout the crystal. The latter is found from a simple geometrical discussion to be about 0.15 for a cube as well as for a short cylinder. Hence, unless our reflectivity is better than 92% we shall have a \mathcal{V}_e of more than 0.001, and in our idealized setup \mathcal{V}_e is identical with \mathcal{V}_T . The shape of the phosphor as well as the outer reflecting layer should therefore be chosen with great care whenever high resolution is a primary design consideration. Substantial improvements over the somewhat meagre optical characteristics of the usual cubes and cylinders are certainly not impossible; even a simple shape like a truncated cone may be preferable. Similarly, ordinary aluminium foil wrapped loosely around the crystal will hardly be an adequate reflector. This important aspect of scintillation spectrometry deserves a great deal more attention.

Equally disturbing are all cathode inhomogeneities. It is readily seen that point-to-point variations of the effective cathode sensitivity of the order $\pm 10\%$ can give rise to a \mathcal{V}_T of 0.001. The work of SCHARDT and BERNSTEIN (1951, their Fig. 3) points in this direction; these authors measured the width of

photopeaks from monochromatic γ -rays, using the same crystal and light collector arrangement with several multipliers, and their $\log \mathcal{V}_Q$ vs. $\log Q$ diagrams suggest a contribution to \mathcal{V}_T by the cathode of the order 0.001. Unfortunately this much seems to be unavoidable with present-day photocathodes, unless one can afford to select a reasonably uniform one from a large sample of tubes. The sensitivity contours measured by GODLOVE and WADEY (1954) on fifteen RCA 5819 cathodes demonstrate how low the general standard of homogeneity is; improvements in the manufacturing processes are most desirable. A short lightguide, 1 cm thick or so, may sometimes be helpful to distribute the photons more evenly over the cathode surface and thus to mitigate the influence of non-uniform response.

Although for the sake of brevity we have just considered \mathcal{V}_e and the cathode inhomogeneities as if they pertained to statistically independent processes it must not be forgotten that there are intrinsic interconnexions between e , f , p and c . The varying region of emission of the photons (and the varying spatial distribution and chromatic composition of the emitted light as well, if the scintillations are weak) brings these connexions into play with the result that the fluctuations of e , f , p and c are correlated among each other, extent and sense of the correlations depending like the magnitude of the fluctuations wholly on the qualities of the given setup. It is quite impossible to disentangle these circumstances mathematically. This is the reason why, in the present treatment, the variance of the overall parameter T was chosen as the relevant fluctuation measure, and not a combination of an "optical factor of merit" for crystal plus lightguide with the photoelectric factor of merit for the multiplier introduced 1952 by SWANK and BUCK. Such a separation of variables does not make sense; witness the above example of the short lightguide and its reaction on the influence of all cathode inhomogeneities.

12. VOLTAGE STABILIZATION

The tube gain is a very steep function of the tube overall voltage V . As a rough but useful rule, \bar{M} is proportional to V^α , where the exponent α takes the value 6.5, 10 or 12 for a tube of 10, 14 or 16 stages, respectively.

Fluctuations in the supply voltage can therefore alter \bar{M} during an experiment and smear out the observed line. Such fluctuations will be slow enough to leave \bar{M} unaffected during any one scintillation; on the other hand we assume them to be sufficiently fast for many changes of \bar{M} to occur during the time in which the pulse-height spectrum is taken down. Then we have another case of alternative events: the scintillations are detected with different multiplier gains \bar{M}_i , which occur in the long run with given frequencies q_i .

A generating function is easily set up in analogy to (25) and all calculations are quite similar to those of the preceding paragraph. First we obtain $\bar{Q} = \bar{X}\bar{T}\bar{M}$, where $M = \sum q_i \bar{M}_i$. Then we introduce the voltage fluctuations explicitly. From $\bar{M} \sim V^\alpha$ we get to a good approximation $\bar{M}_i = \bar{M}(1 + \alpha \Delta V_i / \bar{V})$ and

find by means of this relation that

$$\mathcal{V}_Q = \alpha^2 \mathcal{V}_V + (1 + \alpha^2 \mathcal{V}_V) \left(\mathcal{V}_X - \frac{1}{\bar{X}} \right) + \frac{1 + \alpha^2 \mathcal{V}_V + \mathcal{V}_M}{\bar{X} T}. \quad (28)$$

Here \mathcal{V}_M is, of course, the tube gain variance as observed during a time which is long compared with the voltage fluctuation period. For $\alpha^2 \mathcal{V}_V \ll 1$ and normal scintillation variance,

$$\mathcal{V}_Q = \alpha^2 \mathcal{V}_V + \frac{1 + \mathcal{V}_M}{\bar{X} T}. \quad (29)$$

When variations of transfer efficiency are present as well as voltage fluctuations, "interference" terms appear and make the formula for \mathcal{V}_Q rather cumbersome. Under the same approximations as in (29) we have, however,

$$\mathcal{V}_Q = \mathcal{V}_T + \alpha^2 \mathcal{V}_V + \frac{1 + \mathcal{V}_M}{\bar{X} T}. \quad (30)$$

As a numerical example take a feedback-regulated electronic supply which derives its reference voltage from a good neon stabilizer so that,* allowing for temperature effects and resistor drifts, the h.t. output is stable to within ± 2 parts in 1000. Then we can safely estimate that \mathcal{V}_V is 10^{-6} , and if a 14-stage tube is used we have $\alpha^2 \mathcal{V}_V = 10^{-4}$. Thus the first term in (29) will be negligible compared with the second unless $\bar{X} T > 3000$ or so, and a look at (30) shows that in any case the voltage fluctuations can have a noticeable effect only if \mathcal{V}_T is smaller than about 0.0005. As T will not be constant to such a degree except under very good conditions we conclude that the stabilization of the example is adequate for most practical work.

The whole analysis does not, of course, refer to very slow long-term voltage drifts; these only cause a corresponding slow drift in the values of \bar{Q} measured at different times whereas \mathcal{V}_Q remains unaltered.

13. OTHER SOURCES OF LINE WIDTH

Besides the almost unavoidable variations of transfer efficiency and the rather less important supply voltage fluctuations there are a good many more factors which can increase the instrumental width beyond the ideal value predicted by (23). We are now going to consider these in turn. The discussion will be simplified by the fact that most of those further mechanisms possess some essential feature which renders them equivalent to one or the other of the circumstances that have been dealt with already.

13.1 Non-normal scintillation variance

The number of photons in a scintillation can only be distributed according to a Poisson law if the initial excitation energy of the phosphor is allocated *randomly* to the available luminescence centres. If the excitation of one centre favours

or prejudices the excitation of neighbouring centres \mathcal{V}_X must be greater or smaller than $1/\bar{X}$, respectively, in exact analogy to the behaviour of ionization straggling in gases (FANO, 1947). This aspect of luminescence has apparently not been investigated (SYMON has treated 1948 the somewhat related topic of ionization straggling in crystals).

There are indications, however, that in some phosphors the emission of light is connected with cooperative processes. It is well known that in NaI(Tl) two Tl-ions collaborate to emit one quantum (SEITZ, 1938). In anthracene it has been noticed that the photons are emitted in little groups so that the pulses show a very ragged front edge when viewed with an oscilloscope (POST and SHIREN, 1950a; for photographs see SCHMID and BALDINGER, 1950); one has the impression that the decay of the luminescence is not a random process like the decay of a radioactive source. Scheelite seems to behave in a similar way (HOLMES, 1949, quoted from Fig. 30 given by CARLICK, 1952). Perhaps the afterglow of many phosphors (see BELCHER, 1950a on NaI(Tl), MEYER and MAIER 1953 on anthracene and stilbene) should also be examined in this light.

Although these phenomena occur in the *de*-excitation of the centres and may not affect \mathcal{V}_X , their mere existence suggests that cooperative mechanisms could play a part in the excitation of centres, too, and then the necessary condition for X to be distributed according to a Poisson law is clearly violated. One should therefore not discard the possibility that the luminescence process itself contributes to the instrumental line width *via* a non-normal scintillation variance \mathcal{V}_X .

13.2 Inhomogeneous luminescence yield

Closer examination of many a seemingly perfect crystal reveals that its luminescence efficiency varies considerably from one region to another. Striking examples, due most certainly to inhomogeneous concentration of the activating ions, are to be found in the work of BERNSTEIN and SCHARDT (1952) with LiI(Tl, Ag, Sn) and of LAWSON and FRAUENFELDER (1953) with NaI(Tl). In organic crystals no such variations of luminescence yield have been reported as yet, but there is no reason why they should be impossible. The author has found that anthracene crystals grown from solution were 30% more efficient than clear crystals grown from the melt of the same, extremely pure material. Probably this should be attributed to the presence of a much greater number of lattice defects (which act as quenching traps) in the specimens grown from the melt. As the degree of lattice perfection can vary appreciably throughout a larger crystal one may anticipate concomitant variations of the degree of quenching. Thus in organic crystals variations of luminescence yield are likely to occur too.

An increase in light output from the phosphor has obviously the same effect as an increase in cathode sensitivity; hence varying luminescence yield is statistically equivalent to varying transfer efficiency. The resultant line broadening may be serious if the variations are of the order of several per cent and spread through the whole crystal.

* Up-to-date report on voltage stabilization: BENSON, 1952, 1953b. Behaviour of glow tubes as voltage standards: BENSON, 1953a and further references given there.

13.3 Interaction, edge and scattering effects

It has been assumed until now that each of the observed particles leaves the same amount of energy in the phosphor. In this section we consider briefly the various circumstances which can invalidate this assumption. The result is in each case that the number of photons produced by one particle fluctuates more than indicated by the genuine scintillation variance; thus there will be line broadening due to an apparent \mathcal{V}_X .

There is first of all the finite size of the phosphor which, in complete analogy to the proportional counter, causes edge, escape and combination effects (for the latter see JASTRAM, WHALEN and ZINKE, 1952). A typical example, recently (1953) discussed by CAMPBELL and BOYLE, is the escape from the crystal of bremsstrahlung emitted by fast electrons. This is quite appreciable at higher energies and seriously limits the spectrometry of electron pairs created by hard γ -rays. For example, a pair created by 18 Mev-gammas in NaI loses on the average 20% of its energy in form of bremsstrahlung quanta, most of which are hard enough to escape from the crystal unless the latter is very large (G. M. GRIFFITHS, private communication).

Another possibility is that the incident particles, though monochromatic, are detected *via* an interaction process which yields a continuous distribution of secondary particles in the phosphor. The obvious case is Compton scattering. The resulting pulse spectrum can here be predicted accurately with the help of the extensive data calculated by MAEDER, MÜLLER and WINTERSTEIGER (1954) and LIDÉN and STARFELT (1954), which allow one to take full account of almost every edge and escape effect as well as of the influence of Compton quanta originating in shields, collimator, lightguide and multiplier envelope. Into the same class belongs, for example, the effect of δ -rays emerging from the track of a fast primary particle; here the different response of the phosphor to the two particles will play a rôle.

Finally, the incident particles may be backscattered from the crystal face. This will be of importance only with soft electrons and phosphors of high atomic number. For an organic phosphor it follows from data by SCHONLAND (1923, 1925)* that the backscattering at electron energies below 100 keV amounts to about 10%, independent of energy (see TAYLOR *et al.* 1951, and BUTT, 1952, 1953). Moreover, most of the backscattered electrons lose the greater part of their initial energy to the phosphor so that they give scintillations almost as high as if they had been stopped completely; thus the backscatter will be hardly perceptible because at these low energies all scintillation lines are rather broad anyway (e.g., 50 keV electrons in anthracene: $\bar{X} \approx 1000$, ideal line variance at least 0.01). It seems that scintillation spectrometry of soft electrons is definitely limited much less by backscattering than by the large instrumental width, provided one uses an organic phosphor: in NaI the backscatter would amount to about 25%, in scheelite to 40%.

13.4 Space charge effects

We must come back to space charges at this point because their action is not quite the same in the case of scintillation pulses as it was for single electron avalanches.

The scintillation pulse is a succession of many single electron avalanches which are more or less disconnected owing to their extremely short duration. If there are but few of them they will be separated altogether and so cannot be in each other's way. The sequence emission-transfer-multiplication is then truly a cascade event even though space charges may be affecting the multiplication of each individual photoelectron. In the latter case the situation requires only that we put the space-charge limited values of \bar{M} and \mathcal{V}_M (see § 7.7) into all our formulae.

A limiting action of another type will, however, occur when the constituent avalanches of a pulse overlap and impede each other's formation. Here the cascade formalism becomes useless, but it is readily seen what happens to the line width. At not too dense overlap only a few of the avalanches are affected by space charge and remain smaller than they would otherwise become; this causes firstly a decrease in \bar{M} and hence in \bar{Q} , but at the same time the peak of the single electron line assumes a more rounded shape so that \mathcal{V}_M and with it \mathcal{V}_Q must increase: the scintillation line broadens a little. Not so when the current through the multiplier is high enough to reach saturation. Then all statistical fluctuations are more or less ironed out. The line width is determined only by the number of widely spaced photoelectrons from the tail of the scintillation which are multiplied normally, and by the shot effect in the saturation current: the line will be much narrower than expected from the observed \bar{Q} . Of course, the resolution does not benefit from this effect because of the accompanying non-linear compression of the spectrum. Qualitatively, this dependence of \mathcal{V}_Q on the space-charge limited \bar{Q} is indicated in Fig. 6.

The most prominent consequence of the limitations is the familiar non-linearity of response to increasing particle energies. In the end complete saturation is attained, which obeys the usual $V^{3/2}$ law (ENGSTROM, 1947; N.N., 1952). It seems reasonable to assume that the onset of the limitations, too, obeys such a law; then the limiting action can be lifted by applying higher voltages on and after the limiting dynode because the stage gain increases much slower than $V^{3/2}$ (for an interesting example see Post, 1952a). The non-linear response to scintillations, light flashes and steady photocurrents has been used to study space-charge effects, and to measure the 3% danger values quoted in § 7.7. It seems a little doubtful if the latter are quite representative of the space charge actions on single electron avalanches because allowance should be made for the increase of Δt ; probably the true danger values lie a good deal higher for this case.

13.5 Satellites

As readily seen with the help of a fast oscilloscope, scintillation pulses are usually followed within 0.2 to 2 μ sec. by a ragged bump, termed a "satellite"

* Other reliable backscattering data: WAGNER, 1930; CHYLINSKI, 1932; BRAND, 1936; PALLUEL, 1947; BOTHE, 1949; BURTT, 1949; SUZOR and CHARPAK, 1952; SELIGER, 1952.

by GODFREY, HARRISON and KEUFFEL who first reported on the phenomenon (1951, 1952). The grossly fluctuating size and shape of these afterpulses, their delay together with its voltage dependence, and the different mean delays found in geometrically different multiplier types suggest that they are caused predominantly by ion feedback between cathode and first dynode.

Another afteremission mechanism which contributes to the formation of satellites is that which causes also the increase, familiar to every experimenter, of the background counting rate concomitant with any exposure of a photo-multiplier to scintillations or steady light. The latter has been carefully investigated by SCHAEFFI and BAUMGARTNER (1952, 1953), SCHAEFFI, BAUMGARTNER and FLURY (1953) and SCHAEFFI (1953a) in several tubes which had the same dynode structure but different cathodes, and their results suggest that it is due to Paetow effect (see § 8) on the photocathode. As this is a phenomenon of long persistence which dies out completely only after an hour or so, one would not foresee its direct participation in the formation of satellites, initial quanta and following Paetow electrons being separated by too long a time interval. However, DAVISON (1952) and MUELLER, BEST, JACKSON and SINGLETARY (1952), working with very fast oscilloscopes, were able to resolve the constituent single electron pulses in a satellite and found that besides a group with a delay pertinent to ion feedback there is another group, weaker in intensity, starting immediately after the genuine pulse and thinning out gradually over a few microseconds so that its later part merges with the ion feedback group. When the cathode is exposed to intense rectangular light flashes this group appears as a fairly strong aftercurrent of some 10 μ sec. duration (MUELLER *et al.*), which has often been observed in multipliers serving as photocells in flying-spot scanners (see Fig. 12 of SCHAEFFI, 1953b). Within the limits of our present understanding of both afteremission and the properties of photo-electric surfaces the afterelectrons responsible for this pulse group are most likely to originate during rapid phase rearrangements in the upper cathode layer, and thus should not be called "delayed photoelectrons" (DAVISON) but "early Paetow electrons." This view is corroborated by the fact that cathodes which exhibit a weak long-term Paetow effect (such as introduced by SCHAEFFI 1953a) also exhibit a weaker fast aftercurrent under pulsed-light operation (see again Fig. 12 of SCHAEFFI, 1953b), which leaves little doubt on the common origin of the two phenomena. On the other hand, the hypothesis that the afterelectrons are emitted from traps that lie between the valence and conduction bands of the cathode (as conjectured by SCHAEFFI, 1953a) could not easily explain the long-term emission, and is at variance with the fact that this long-term emission is considerably more intense in gassy multipliers (as observed by the writer with several EMI 6262 tubes of widely different quality).

Thus the main causes of the satellites are ion feedback to, and Paetow effect at, the photocathode. Afteremission from the dynodes seems negligible, as demonstrated by the photographs of oscilloscope traces given by LANTER and CORWIN (1952) and MUELLER *et al.* (1952). No further significant information is available; in particular the influence of temperature has not yet been

studied in sufficient detail. A factor of practical importance is the possibility that one afterpulse begets another. This regeneration can lead to very long delays: small satellites were found by MUELLER (1952) 10 μ sec., and by LANTER and CORWIN (1952) as much as 40 μ sec., after the genuine pulse.

To account statistically for the presence of aftereffects by always using \bar{M} and \mathcal{V}_M instead of \bar{G} and \mathcal{V}_G (see § 8) is the correct procedure only if the satellites are made up of independent contributions from each genuine photoelectron; otherwise the pulse formation is not a true cascade event. As far as ion feedback is concerned we may be sure of additivity. Paetow effect is another matter; its intensity could possibly depend in a non-linear way on cathode illumination. The exact statistical treatment then becomes very complex and will not be reproduced here because it indicates only an imperceptible line broadening under ordinary conditions. The possibility of a non-linear Paetow effect should however be kept in mind as it would lead to a faint non-linearity of multiplier response.

Many workers have noticed that the mean intensity of afterpulsing (the \bar{A} of § 8) often changes appreciably from one minute to the next. This behaviour cannot affect \mathcal{V}_M to a marked extent, as follows from the discussion of eq. (20), but it will influence the tube gain according to (19). Thus we can regard it as equivalent to fluctuations of the multiplier supply voltage. The only difference from the considerations of § 12 is that we have now $\bar{M}_i = \bar{M}[1 + \Delta\bar{A}_i/(1 + \bar{A})]$, as is readily deduced from (19), and therefore end up with

$$\mathcal{V}_A \frac{\bar{A}^2}{(1 + \bar{A})^2}$$

replacing $\alpha^2\mathcal{V}_V$ in formulae (28)–(30). Numerically, we may have $\bar{A} = 0.2$ as for the tube of Fig. 5, and a \mathcal{V}_A of 0.04 seems a plausible possibility; then the above equivalent $\alpha^2\mathcal{V}_V$ is about 0.001. This is serious enough and speaks quite definitely against the use of gassy tubes in precision spectrometry.

13.6 The collecting time constant

In the foregoing, the collecting time constant at the multiplier anode was always assumed to be effectively infinite. A short collecting constant is often employed on purpose: in the interest of recording speed, to prevent pile-up of pulses, or to suppress the satellites. This has some consequences of considerable practical importance.

Disregarding aftereffects for the moment, we note that a short collecting time constant partly differentiates the pulse. The ensuing ballistic loss of pulse height will be called the "collection deficit"; for a fast scintillation it is identical with the combined height of all those single electron avalanches which arrive at the anode so late that they fall on the descending back of the output (voltage) pulse. We can therefore represent it by an increase in the probability k that a photoelectron is not detected which (see § 7.4) entails a decrease of \bar{M} as well as an increase of \mathcal{V}_M . But that is not all; since the number of latecoming photoelectrons fluctuates from one pulse to the next the collection deficit also has a

statistical character. The consequence is, as a little calculation similar to that in § 15 shows, that the apparent increase in \mathcal{V}_M is greater than the increase predicted by (18) for a constant k .

In practice, the collecting time constant will always be several times longer than the decay time of the phosphor. Then the collection deficit is no serious matter. SWANK and BUCK (1952) have calculated the apparent increase in \mathcal{V}_M (δ in their notation) for phosphors with a truly random, exponential decay, and find that it amounts to at most a few per cent. But the point must be made that this increase depends necessarily on the statistical character of the luminescence decay. If there is no true randomness of de-excitation the collection deficit will fluctuate rather erratically from one pulse to the next and cause \mathcal{V}_M to increase appreciably. In other words, with phosphors such as scheelite a short collecting time constant leads to line broadening.

Turning now to satellites, one realizes that a short time constant discriminates against them if the scintillation itself is very fast. For example, a collecting time constant of 2 $\mu\text{sec.}$ or less gives such a steep back to pulses from an organic phosphor that no satellite can climb up as high as the genuine peak. Not so with slower phosphors. For example, pulses from NaI(Tl) collected with a time constant of 2 $\mu\text{sec.}$ reach maximum height about 0.6 $\mu\text{sec.}$ after the beginning of emission so that here the satellites pile right on top of the genuine pulse.

It will depend mainly on the performance of the electronic recording equipment if this discrimination leads to noticeable changes in the observed pulse spectrum. Suppose we are using a valve voltmeter which always measures the maximum height of an incoming voltage pulse, whether that has one or several peaks (e.g. wide-band amplifier plus pulse lengthening stage plus slow kicksorter). Then NaI pulses will be processed as if the collection deficit was subject to a little extra fluctuation, owing to the irregularity of ion feedback, whereas pulses from an organic phosphor, divorced from their satellites as they are, will simply appear subject to the tube gain variance \mathcal{V}_G instead of \mathcal{V}_M . In either case the relative line width remains practically the same; there is, however, an obvious shift in the mean pulse height which has often been overlooked and led to considerable errors. For instance, in comparing the scintillation efficiency of organic substances with an NaI standard a long time constant *must* be used, otherwise the satellites are not as efficiently suppressed in the NaI pulses as in the specimen pulses and falsify the comparison in favour of the NaI.

Suppose, on the other hand, that we have a valve electrometer which measures the total area under an incoming voltage pulse, e.g. a wide-band amplifier plus integrating stage plus kicksorter. The integration will compensate rigorously for all effects of the preceding differentiation, provided the integrating time constant is very long. If it is comparable to the collecting time constant, we must, however, expect all lines to broaden as if there was a fluctuating collection deficit, the shape of the pulses being highly irregular due to the satellites.

It emerges from these qualitative considerations that a *steady* level of after-pulsing is not really annoying as far as the instrumental width is concerned.

13.7 High counting rates

At very high counting rates the photocathode, being a semiconductor (SCHAETTI, 1951a, b, 1953), can charge up so strongly that the voltage between cathode and first dynode is appreciably lowered. The ensuing reduction of photoelectron collection efficiency causes both a decrease in pulse height and an increase in line width, as every reduction of T will do. Moreover, if the potential partially recovers during the irregular intervals between successive scintillations, a varying transfer efficiency may be simulated. The latter would not arise from mere cathode fatigue, and the dynode fatigue usually observed under heavy duty (ENGSTROM, 1947; MARSHALL, COLTMAN and HUNTER, 1948; HILLERT, 1951) will not affect the line width at all (except for possible fatigue drifts in \mathcal{V}_G).

13.8 The electronic equipment

It is not commonly realized that the electronic apparatus employed to measure the pulses is a copious source of small parasitic statistical fluctuations too. Amplifier noise is no problem to scintillation spectrometry because the pulses delivered by a multiplier are always high enough. But there is the production of pulse tails in the amplifier, due to short coupling time constants; the instability of amplifier gain, due to drifts in valve characteristics, flicker effect and changing resistor values; then output hum, also from cathode follower stages; and if the setup includes a discriminator, the well-known instability of its bias level. All these factors are statistically equivalent to fluctuations of the multiplier supply voltage and can be assessed as such. The effects are usually small but it is well to keep them in mind when experiments of high precision are intended.

14. THE SHAPE OF NON-IDEAL LINES

All the non-ideal conditions discussed in the foregoing give rise to alternative events: some parameter fluctuates from one scintillation to the next, instead of remaining constant as in the ideal case, and the overall pulse-height distribution results from the superposition of many ideal lines, one for each possible parameter value. It is quite obvious that the line shape need not be gaussian under such circumstances, the superposition process being determined by the probability distribution of the fluctuating parameter which can have almost any form. For example, for a crystal with one large crack the possible values of T could cluster about two main values (bimodal distribution for T) so that the observed line could have two peaks. Another, almost trivial, example is the shape of a Compton edge. A gaussian shape will only be observed if the parameter distribution itself is a narrow gaussian; e.g. with a crystal whose luminescence yield varies in a random way over very small domains. Note that only in such a case it is legitimate to regard \mathcal{V}_G as a measure of the full line width at half-maximum or *vice versa*.

15. NATURAL AND INSTRUMENTAL WIDTH

Frequently, the particles under observation are not strictly monochromatic but possess an energy spread which is entirely analogous to the natural width of a spectral line. We briefly consider how this natural width combines with the instrumental width of the scintillation spectrometer.

Let a particle of energy E produce on the average $\bar{X}(E)$ photons, with a variance $\mathcal{V}_X(E)$. Energy E contributes to the overall distribution of emitted photons with a weight equal to the frequency of its occurrence in the given distribution of particle energies, thus the case is again one of alternative events. The calculation of the mean number \bar{Y} of photons released by the whole natural line, and of its variance \mathcal{V}_Y , is quite straightforward if it is assumed that both $\bar{X}(E)$ and $\sigma_X^2(E)$ are proportional to E ; this means linearity of phosphor response, and energy independence of the departure from normality of the scintillation variance. One readily obtains

$$\bar{Y} = \bar{X}(\bar{E}) \quad (31)$$

$$\mathcal{V}_Y = \mathcal{V}_E + \mathcal{V}_X(\bar{E}) \quad (32)$$

where \mathcal{V}_E is the relative variance of the natural line.

The combination of natural and instrumental width is then effected by substituting (31) and (32) for \bar{X} and \mathcal{V}_X in all the formulae for the monoergic case. In particular, one finds from (22)

$$\mathcal{V}_Q = \mathcal{V}_E + \frac{1 + \mathcal{V}_M}{\bar{X}(\bar{E}) \cdot T} \quad (33)$$

for the line variance observed under ideal conditions with a phosphor of normal scintillation variance. The line shape is of course not necessarily gaussian; cf. the remarks made in the preceding paragraph.

16. COMPARISON WITH THE PROPORTIONAL COUNTER

The pulse formation in a proportional counter is a simple cascade event. First an ionizing particle creates X ion pairs, and each of these is subsequently multiplied in the familiar gas amplification process with a gas gain M , subject to a gas gain variance \mathcal{V}_M . In the end Q electrons arrive at the central wire. From § 5 it is clear that

$$Q = \bar{X} \bar{M} \psi \text{ and } \psi \mathcal{V}_Q = \mathcal{V}_X + \frac{\mathcal{V}_M}{\bar{X}}.$$

One realizes that there is a close formal analogy with the scintillation spectrometer, the only difference being that now there is no transfer efficiency smaller than 1 since none of the initial ion pairs is lost to the multiplication.

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Hence the formulae developed in the foregoing can be adapted to the proportional counter case by simply setting $T = 1$. In particular we have

$$\mathcal{V}_Q = \frac{1 + \mathcal{V}_M}{\bar{X}} \quad (34)$$

if the ionization straggling is normal; as FANO (1947) has shown \mathcal{V}_X differs in general somewhat from $1/\bar{X}$, but (34) is nevertheless a sufficiently good approximation for the purpose of the following rough comparison.

We may ask first how large the gas gain variance is. FRISCH (1948; cf. WILKINSON, 1950; or WEST, 1953) calculated a value slightly smaller than 1, starting from somewhat pessimistic assumptions; the experimental investigation of a single electron line by CURRAN, COCKROFT and ANGUS (1949) yielded very nearly $\frac{2}{3}$. We note that this is not greatly different from the usual value of 0.4–0.5 for the tube gain variance of a photomultiplier. There is still another similarity between the two instruments. In any gas an ionizing particle spends about 30 ev of its energy to produce one ion pair; on the other hand, in a good phosphor with a scintillation efficiency of 10% again about 30 ev are required for the production of one photon ($h\nu$ at 4000 Å is nearly 3 ev). Thus there is not only a formal, but also a close numerical analogy between (34) and (23). One sees that a monochromatic line observed in an ideal scintillation spectrometer with a transfer efficiency of 10% has about ten times the variance which it would have in a proportional counter; or: the line width is about thrice that which the same particles would give in a proportional counter.

This result exposes once more the present main weakness of the scintillation spectrometer—its poor resolution. The comparison is actually optimistic because it rests on highly idealized assumptions. Indeed, to obtain no more than three times the proportional counter width we must use a specially shaped crystal of excellent quality in conjunction with a photocathode of much better homogeneity than is commercially available at this time.

On the other hand, we have assumed a luminescence yield and a transfer efficiency of only 10%. There is no fundamental reason why we should not be able to do much better in the future. Photoelectric processes with a quantum efficiency of 50% have been reported (HEIJNE, SCHAGEN and BRUINING, 1954); their study may lead to the discovery of more efficient photocathodes. Some day, it is to be hoped, our understanding of energy transport and quenching phenomena will have advanced far enough to enable us to “design” phosphors of desirable characteristics, including high yield. With such improvements the scintillation counter might become a spectrometric tool of much greater power than is commonly realized.

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NOTES ADDED IN PROOF

Several of the above conclusions have been reached independently by G. T. WRIGHT (*J. Sci. Instr.*, in the press). He also discusses the influence of dynode inhomogeneities on the instrumental width. This topic has not been treated in the present article because the electron-optical properties of usual multipliers are such that the varying energy of the photoelectrons and their varying direction of emission suffice to distribute the photoelectrons ejected from a given cathode point over a rather large area of the first dynode (ca. 0.5 cm^2 in the RCA 5819 and EMI tubes). Thus all inhomogeneities of the first dynode—unless exceptionally prominent—are automatically concealed, and affect only the tube gain variance (see p. 68).

The strong cathodoluminescence of MgO was recently shown by K. P. MEYER and A. MAIER (1954), *Helv. Phys. Acta*, **27**, 57) to cause considerable light feedback on single electron avalanches in tubes with Ag-Mg dynodes.

G. T. WRIGHT and G. F. J. GARLICK (1954, *Brit. J. Appl. Phys.*, **5**, 13; their Figs. 6 and 7) have published data similar to those of HOPKINS reproduced on p. 77. In some instances they found non-ideal contributions to the instrumental width which are much too large (up to 0.017) to be explained only by varying transfer efficiency.

3

MESON PHENOMENA AND THE
MESON THEORY

R. H. Dalitz

I. INTRODUCTION

The meson theory was first proposed by YUKAWA in 1935 in order to account for the qualitative properties of nuclear forces. A meson field interacting directly with the nucleons was introduced in analogy with the Maxwell field, which gives rise to electromagnetic forces between charged particles, and the nuclear forces between nucleons were considered to result from the virtual interchange of the quanta of the meson field between the nucleons. The short range of nuclear forces was then a consequence of the massiveness of the meson quanta, according to the Uncertainty Principle, the range then known corresponding to a mass of 200–300 electron masses. The strength of the nuclear forces required this interaction between nucleon and meson field to be very strong and their charge exchange character further implied the existence of charged quanta of the meson field.

It is now known that the π -meson, first identified by POWELL and OCCHIALINI in 1947, is the particle which fits the qualitative requirements of YUKAWA's proposal, as stated above. The π -meson is the meson which is produced most copiously and directly in the high energy collisions of cosmic rays. The development of particle accelerators in recent years has enabled the laboratory production of π -mesons by collisions of nucleons or photons with nuclei. Many of the quantitative properties of the π -mesons have now become well established as a result of laboratory experiments with these π -meson beams and the beam intensities available have even permitted the detailed study of the scattering of mesons by protons, both in angular distribution and as function of energy.

Since YUKAWA's original proposal, a very considerable literature on meson theories has accumulated, many modifications of the detailed meson theory having been proposed and developed, keeping to the spirit of YUKAWA's interpretation of nuclear forces. Theories have been discussed for various possibilities of meson spin and parity and for various forms of the meson-nucleon interaction. Calculations for many meson processes have been made with these theories, using a variety of approximate methods. Although these investigations have played a significant role in the development of our understanding of meson theories, it is now clear that most of these calculations are not appropriate for the case of physical interest, the π -meson.

In this article, the aim is to collect together our empirical knowledge of those detailed properties of the π -mesons obtainable from the analysis of the experimental data now available, and then to discuss to what extent these necessary