LUMINESCENCE AND THE SCINTILLATION COUNTER

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CURRAN

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by

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PREFACE

THE scintillation method of investigating atomic and nuclear radiations has developed since 1945 at an almost unprecedented rate. Within some six years it has become predominant among the electrical methods of counting and analysing. It has been applied in practically every kind of investigation previously tackled with the help of the Geiger-Müller or proportional counter or pulse ionization chamber and in most instances with very considerable advantages to the investigator. In parallel with this entry into established fields we have seen the new method successfully employed in circumstances where the others prove relatively useless. Its flexibility is indeed astonishing when one considers that several rather complex processes enter into its operation.

In these circumstances it seemed desirable that a fairly complete guide to the subject should be written. The treatment has not digressed far from the main theme at any point but topics such as secondary emission, photoelectricity and luminescence, which were not hitherto very closely involved in the electrical methods of counting, have received some detailed consideration, particularly in those aspects which seem to have some bearing on present or possible future forms of scintillation devices. It is hoped that the collection of such topics within the one volume will ease the task of many workers. The writer will be satisfied if he has, at one and the same time, managed to produce a book useful to those using or proposing to use the technique as incidental to their main studies and to those intending to develop further its inherent possibilities. The former in particular deserve sympathy in their exposure to the flood of papers on counters and related topics. In short it is hoped that the book manages to present the various parts of a rather complicated subject in their proper perspective.

It seems to the writer that no very useful purpose is served by discussing in detail the almost unlimited number of particular applications of the scintillation method. Hence the chapter on applications deals with the more basic problems such as gamma, neutron and meson spectroscopy. The same remarks apply to the treatment of circuits. Those circuits which seem well adapted to the new technique have been discussed. It is thought that crystals, electronic units, *etc* which are available commercially are adequately described in the advertisements appearing regularly in some of the journals cited in the references and the sources are not given in detail in the text.

The author would like to record his debt to those who have collaborated with him since his first experience with the scintillation method in 1944. In addition he is indebted to Dr J. M. VALENTINE for much assistance in several directions and especially in proof-reading. He is glad to express his appreciation of the help and facilities accorded by Professor E. O. LAWRENCE, of the University of California, and Professor P. I. DEE, F.R.S., of the University of Glasgow, in whose laboratories his own research was performed. The author is pleased to record his debt to his wife (J. E. STROTHERS) for much assistance throughout and especially for her work on the indexes.

He would like to record his thanks to the many authors and publishers of papers for permission to reproduce original material, relevant sources being given in reference lists and indicated appropriately in the text.

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21*st April* 1953

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GENERAL IDEA OF THE SCINTILLATION COUNTER

HISTORICAL

IN ONE SENSE the scintillation counter is not new and may be said to antedate the proportional tube and Geiger-Müller counter. The light flashes emitted when heavily ionizing particles such as α-rays and protons struck a thin screen of a suitable material, for example zinc sulphide, were visible to the human eye and could be counted at a slow rate. This very simple arrangement, in which the skill of the observer played a dominant part, was used in many experiments of great fundamental importance to the development of atomic and nuclear physics. It became practically obsolete as a method of counting with the introduction of the pulse ionization chamber and associated electronic amplifier. Feeding into some type of scaling and registering circuit the chamber proved to be a very sensitive and versatile tool. Parallel developments of the Geiger-Müller counter brought this instrument to a high pitch of performance and it became a reliable consistent detector, particularly for β-particles and electromagnetic radiations.

This was the situation prior to 1939, but during the 1939–45 war unusual needs arose in the field of instrumentation and these needs frequently highlighted certain defects in the performance of the orthodox instruments. For example, the microphony, fragility and sensitivity to the presence of magnetic fields became major objections to the use of pulse ionization chambers and limited their use to certain types of investigation. Furthermore, considerable experience and knowledge were required from the operator. In this situation the first simple form of the modern scintillation counter was introduced by CURRAN and BAKER in 1944. Expressed very briefly, the counter was a revival of the older scintillation technique with the important difference that the eye of the observer was replaced by a photomultiplier capable of delivering large pulses of current of short duration, one per scintillation, and these could be registered directly by almost any electronic device currently in use for such purposes. KALLMANN independently discovered this method of counting at a later date and he applied it to both particles and quanta, using mainly single crystals of various luminescent materials to produce the scintillations.

This new technique of counting constituted a major advance, since the instrument with its commercially available photomultiplier was simple in design, free from microphony and fragility of any kind, and it could count at rates exceeding those possible with any other counting device. Moreover, the work of KALLMANN soon established clearly that the device could be used proportionally and from the pulse amplitudes much knowledge of the nature and energy of the radiations could be deduced. The instrument possessed within itself the major advantage of the proportional tube counter and at the time of writing it has superseded in promise, if not in actual performance, the gas-filled proportional counter, except in in the field of analysis of radiations of relatively low energy.

Among the really striking advances which followed the general introduction of the new instrument into atomic and nuclear physics we must stress particularly the achievements made possible by its application to the detection and analysis of γ-radiation.

At present there are scarcely any types of radiation to the studies of which the scintillation detector has failed to make a notable contribution. More will be said in later chapters concerning these advances, but as one of the more interesting cases we would refer to the successful application of the method to the examination of neutron sources. Here both crystalline and liquid scintillators are being exploited.

MAIN FEATURES OF THE SCINTILLATION DETECTOR

The scintillation counter involves in its operation a number of fundamental physical processes. In its elementary form it takes the shape indicated in *Figure 1*, but, as we shall see in the succeeding chapters, there are many variations and complications of the detector system. A source *S* emits radiation which falls on the luminescent material *L*. This may in practice take the form of a thin layer of microcrystals of say activated zinc sulphide, or it may be a single large crystal of an inorganic substance such as sodium iodide (activated perhaps with thallium) or of an organic crystal such as anthracene. It may be a vessel containing a liquid such as a solution of terphenyl in xylene. A multiplier *M* with a photosensitive cathode *P* is placed so as to collect the maximum amount of light radiated by *L*. If *M* and *L* cannot be put in good optical contact some form of light guide such as a rod of quartz may be required. The cathode *P* may be a semi-transparent layer of suitable photosensitive material, deposited on the end face of the tube *M*, or it may be a layer on a solid electrode supported within the envelope. The electrode structure of *M* takes a variety of shapes and we choose for illustration the 'venetian-blind' type which is favoured in some designs. We show separate side-connections to the dynodes in *Figure 1*, but normally they are brought out at the end with the anode lead. The photoelectrons from *P* are accelerated by the applied field to the first dynode and on striking this they release secondary electrons which in turn are drawn to the second dynode and so on till a pulse of secondary electrons is finally collected at the anode *O*. We thus have a magnified avalanche of electrons at *O* for each group of one or more

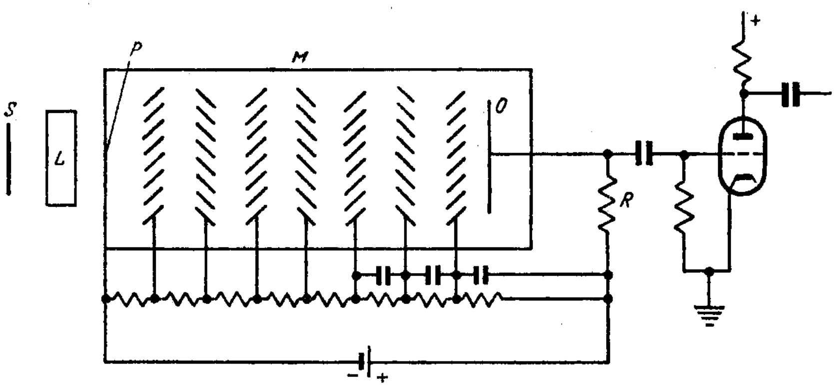

Figure 1. Diagrammatic representation of scintillation detector

photoelectrons released at *P*. In the most favoured design of *M* the electrodes are solid and arranged in an approximately circular array. The secondary electrons are emitted and drawn away from the same surface of the dynode as is bombarded by the incident electrons. The electrical fields are shaped so that the slower secondaries can be directed successfully and indeed roughly focused on the succeeding dynode. The dynode surface is so processed that about three to five slow secondary electrons are emitted by each incident fast electron. This ratio of secondary electrons to primary electrons is known as the multiplication factor per stage, and generally between ten and fourteen stages are incorporated in commercial tubes. A voltage developed across the final collector resistor *R*, by virtue of a current (steady or pulse) passing through it, may be further amplified if necessary by an external electronic amplifier as indicated. Since the multiplier is itself a high-gain, low-noise, high-passband amplifier the design of any associated external circuit is a matter of considerable difficulty unless some of the advantages afforded by the tube itself are to be sacrificed. For this reason it is usually desirable to have a minimum of external amplification, which means that tubes with high stable gain per stage and with as many stages as required should be chosen if available.

We show an a.c. amplifier attached but different devices are employed according to the nature of the problem. Thus it is frequently possible that a measurement of the output current with a current meter of some kind is all that is necessary or again, if current pulses through *R* are being examined and they are of sufficient magnitude, they may be applied directly to the deflecting system of a cathode ray tube. A great variety of methods of display and registration exists and the actual choice depends on the nature of the particular experiment.

PHYSICAL PROCESSES INVOLVED IN SCINTILLATION COUNTING

We can somewhat arbitrarily break down the process of scintillation counting into three main parts:

(1) The kinetic energy of the radiation incident upon the luminescent material must be converted as efficiently as possible into energy of excitation and ionization of the atoms or molecules of the scintillator.

(2) The de-ionization and de-excitation of the molecules should result in the emission of fluorescent radiation and this radiation should be transmitted as freely as possible through the luminescent material. The radiation should be of such wavelengths as to match well with the known spectral characteristic curve of the photomultiplier.

(3) Assuming the spectral distribution curve of the fluorescent radiation and the spectral sensitivity curve of the tube are well matched, the photosensitivity of the cathode of the tube should be high so that the maximum emission of photoelectrons is secured. These photoelectrons in turn must be efficiently collected and directed on to the first dynode. Finally the multiplication of these in number should be as high as is consistent with stable and uniform performance of the multiplier itself.

We can see that there are many factors involved in the process of detecting particles or quanta with the scintillation counter and the specification is by no means set forth completely in the above;

for example, for any application to spectrometry we must add the stipulation that the number of fluorescent quanta emitted (and the fraction reaching the cathode) should be proportional if possible to the energy expended by the radiation incident upon the material. Further, the number of photoelectrons released by the fluorescent quanta should in turn be directly proportional to the expended energy. This second stipulation imposes fairly stringent standards on the uniformity of response of the photocathode over its whole useful area, while the first involves uniformity in response of the luminescent material through the volume exposed to the primary radiation. Provided these two main conditions are satisfied, and to a large extent in practice they are, the scintillation detector can be used to produce an impulsive charge at the final collector of magnitude proportional to the energy dissipated by an ionizing particle within the scintillator. Measurement of the amplitude of the output pulses thus can yield information on the energy of a homogeneous radiation. The detector is then employed as a spectrometer.

It is instructive at this point to illustrate the mode of action by considering a particular case, say the detection of the α-particles of polonium by the scintillations produced in zinc sulphide, silver activated, ZnS (Ag). If the tube is one of 11 stages and the inter-stage gain at say 100 V per stage is 4, an overall gain M of 4^{11} or roughly 4×10^6 is obtained. Now consider the conversion efficiency, C, of the kinetic energy E of the α-particles (for polonium $E = 5$ MeV approximately) into fluorescent radiation. For a good sample of ZnS (Ag) the efficiency is about 20 per cent or a total energy per α-ray of about 1 MeV is converted into photons within the phosphor. Next the transmission factor T of the ZnS (Ag) preparation for its own radiations must be taken into account and in addition the geometry of the apparatus will be such that only a fraction G of the emitted photons will reach a useful part of the photocathode. For a small screen of ZnS (Ag) mounted directly on a semi-transparent photocathode and covered with a good reflecting material such as very thin highly polished aluminium the geometry factor G will usually lie between 0·5 and 1·0. This same range will cover the possible values of T. Consider next the average value of the photosensitivity P of a suitable multiplier. This is of the order of 40 μamp/lumen for a good photocathode and is equivalent to about 10 electrons per 100 quanta for light of wavelength 4000 Å (energy 3 eV). Let us take $P = 0{\cdot}1$ electrons per photon (energy W) in our example. Finally the collection efficiency factor F for the photoelectrons ranges in practice from about 40 to 100 per cent according to the design of the tube, the presence of a magnetic field, the first stage voltage and so on. With these considerations in mind we can write down an expression for the charge (in electron charges) collected at the final anode, *viz*

$$Q = (CE/W)\ TGPFM \quad \ldots\ldots (1.1)$$

Adopting the least favourable values discussed above

$$Q = \tfrac{1}{3}(0{\cdot}2 \times 5 \times 10^6) \times 0{\cdot}5 \times 0{\cdot}5 \times 0{\cdot}1 \times 0{\cdot}4 \times 4 \times 10^6$$

in electron charges or approximately 2×10^{-9} coulombs per pulse. If this charge is stored momentarily in a capacity of 20 $\mu\mu$F at the anode, its voltage alters by 100 V. It is obvious that a large pulse corresponding to each α-particle can be observed easily and its amplitude is proportional to the energy of the incident particles.

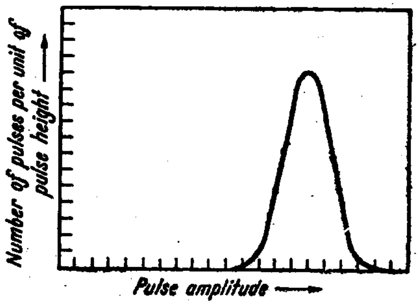

Figure 2. Intensity distribution of alpha rays as function of pulse amplitude

The distribution of the pulses thus takes a form as shown schematically in *Figure 2* and we have a method of analysing the energy spectrum of such radiations. *Figure 2* refers to a source of mono-energetic α-rays.

The example is interesting in showing that many factors influence the performance of the scintillation counter. Each of these factors will be considered in detail in the subsequent chapters. Much has been learned about them in the rapid and fruitful research which has been pursued during the last six or so years. Not only has this research greatly advanced our understanding of the instrument and led to greater skill and ingenuity in its application, but it has also resulted in fresh and profitable attacks on the nature of some of the fundamental physical processes involved, such as the mechanism of fluorescence. The instrument, in forms both simple and complex, has been applied in many fields with great success, and as a tool for research in nuclear and atomic physics it will undoubtedly prove of very great value.

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For general information on various forms of counters, other than the scintillation type, the following texts are available:

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RADIATIONS AND THEIR INTERACTION WITH MATTER

ELEMENTARY PARTICLES

We propose to discuss very briefly the nature of the particles which will feature very prominently in our later discussions of the scintillation counter.

The electron

The particle is of very small rest mass m_0 (9×10^{-28} g) and it carries an invariable negative charge—the electronic charge equal to $1{\cdot}59 \times 10^{-19}$ C. The mass of an electron travelling at velocity V is given by

$$m = m_0/(1 - V^2/c^2)^{\frac{1}{2}}$$

The term β-particle is generally applied to the electrons which are emitted by many radioactive elements, so-called β-emitters. These electrons form a continuous or primary β-ray spectrum with energy extending to a well defined upper energy limit.

The positron

Particles with the same rest mass and charge as the electron are known as positrons when the sign of the charge is reversed. They were first observed in cosmic radiation but later were shown to be emitted by many artificially radioactive substances.

The positive ions

On the Bohr picture of the atom a positively charged nucleus is surrounded by a number of electrons with total negative charge exactly neutralizing the positive charge on the nucleus. Thus atoms and molecules in their normal state are neutral. When one (or more) of the satellite electrons is stripped from an atom it possesses a positive charge and is known as a singly (or multiply) ionized atom. A few cases are of special interest to us. An atom of hydrogen lacking its electron is known as a proton, and heavy hydrogen or deuterium gives the deuteron, while tritium without