

SCINTILLATION COUNTERS

by

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South Africa

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EDITOR'S PREFACE

THE aim of these monographs is to report upon research carried out in electronics and applied physics. Work in these fields continues to expand rapidly, and it is recognised that the collation and dissemination of information in a usable form is of the greatest importance to all those actively engaged in them. The monographs will be written by specialists in their own subjects, and the time required for publication will be kept to a minimum in order that these accounts of new work may be made quickly and widely available.

Wherever it is practical the monographs will be kept short in length to enable all those interested in electronics to find the essentials necessary for their work in a condensed and concentrated form.

D. W. FREY

AUTHOR'S PREFACE

THE scintillation counter is an instrument of great versatility and interest. In its original visual form it played a major role in the development of *classical* nuclear physics, from the identification of the α -particle and the discovery of the atomic nucleus up to the COCKCROFT-WALTON experiments which initiated the "machine age" of modern physics. The scintillation counter then passed through a *dark age* of 15 years, during which it was relegated to the museum and the Honours laboratory, while the Geiger counter and the new electronics dominated the scene. The advent of the photo-multiplier tube brought about the *renaissance* of the scintillation counter, and as in the historical analogue new modes of expression, new materials, were developed to replace those inherited from the classical period. Zinc sulphide, the classical phosphor, was joined by naphthalene, anthracene, stilbene and the other organic crystals, by the iodides and tungstates, and by the plastic and liquid phosphors. With these materials a host of new experimental techniques became available; milli-microsecond resolving times, β - and γ -spectrometry of milli-microcurie activities, efficient X-ray, γ -ray and neutron detection, simple heavy particle spectrometry, observation of the life-times of mesons, positrons and nuclear isomers, and detection of the neutral π -mesons, to name some of the recent applications of the scintillation counter. The history of the instrument since 1949 is largely a history of experimental nuclear physics since that date. Already the scintillation counter has regained the place, previously abdicated by its visual ancestor, as the most important and versatile instrument in nuclear physics research.

In this monograph an attempt has been made to integrate and to

review critically these extensive developments of the scintillation counter. After a brief historical introduction (1), the numerous factors influencing the performance of this deceptively "simple" instrument are enumerated (2). Photo-multiplier characteristics are then considered and compared (3), and this is followed by a discussion of the pulse-height and time resolution, two of the major properties of the instrument (4). The inorganic phosphors are treated in the following chapter (5). A detailed discussion (6) follows of the photo-fluorescence and scintillation characteristics of the organic crystalline phosphors. An account is given of the research leading to the author's new "photon transfer" theory of fluorescence. This theory was developed while the monograph was being prepared, and its publication here precedes many of the papers on which it is based. In (7) the properties of the plastic and liquid phosphors are described and interpreted in terms of the "photon exchange" theory. The final chapter (8) is a comprehensive review of the major applications of the instrument to date.

Apart from the *nuclear physicist* and those applying similar techniques in related fields, it is hoped that this monograph will be of interest to those in other sciences. The new experimental and theoretical data on molecular fluorescence represent a radical modification of the technical data in the previous literature. They offer a distinct possibility to the *organic, physical and theoretical chemist* of achieving a correlation between the spectral properties of the aromatic compounds and their π -electron structure, and they suggest new fields of investigation for the *solid-state and molecular physicist*.

Since the text was completed a further development into the *biological and medical sciences* has been suggested by the author. Fluorescent organic compounds are present in many important biological systems. These include *inter alia* the chlorophylls, xanthophylls and carotenes in plants, and the sterols, hemoglobins, vitamins, hormones and carcinogens in animals. A general physical theory of the role of these fluorescent compounds in different biological processes, including carcinogenesis and carcinolysis, has been proposed, and it will be published shortly. Experiments are in progress in conjunction with workers in many different sciences to verify, extend or modify this theory. The basis of this new venture in *biophysics* is the work described in Chapters 6 and 7 of the present monograph.

In conclusion I wish to thank Mr. D. W. FRY and the publishers and printers for the miraculous speed of publication, and finally my wife without whose unfailing encouragement and invaluable assistance this monograph might never have been published.

J. B. BIRKS

CONTENTS

	PAGE
EDITOR'S PREFACE	
AUTHOR'S PREFACE	v
1 INTRODUCTION	1
1.1 The Detection of Atomic and Nuclear Radiations	1
1.2 The Visual Scintillation Counter	2
1.3 The Photo-multiplier Scintillation Counter	5
2 PRINCIPLES OF THE SCINTILLATION COUNTER	8
2.1 Introduction	8
2.2 Absorption of the Incident Radiation	9
2.3 Photon Emission from the Phosphor	13
2.4 Transit to the Photo-cathode	14
2.5 Conversion into Photo-electrons	15
2.6 Electron Multiplication	18
2.7 Overall Characteristics	20
3 THE PHOTO-MULTIPLIER TUBE	21
3.1 The Circular Electrostatically-focused Photo-multiplier	21
3.2 The Longitudinal Electrostatic Photo-multiplier	30
3.3 Comparison of the Photo-multiplier Tubes	34
3.4 Light Collection	36
4 PULSE HEIGHT AND TIME RESOLUTION	39
4.1 Pulse Height Resolution	39
4.2 Time Resolution	42
4.3 Satellite After-pulses	45

	PAGE
5 INORGANIC PHOSPHORS	47
5.1 The Luminescence Process	47
5.2 Zinc Sulphide and Similar Phosphors	50
5.3 Sodium Iodide	52
5.4 Other Alkali Halide Phosphors	55
5.5 The Tungstate Phosphors	57
5.6 Other Inorganic Phosphors	60
6 ORGANIC CRYSTALLINE PHOSPHORS	62
6.1 The Photo-luminescence Process	62
6.2 General Properties	78
6.3 Anthracene	79
6.4 Other Organic Crystals	84
6.5 The Scintillation Process	91
7 ORGANIC PLASTIC AND SOLUTION PHOSPHORS	102
7.1 Phosphor-plastic Mixtures	102
7.2 Organic Solution Phosphors	103
8 APPLICATIONS OF SCINTILLATION COUNTERS	111
8.1 The Detection of Heavy Particles	111
8.2 The Spectrometry of Heavy Particles and Electrons	114
8.3 The Detection of γ -radiation	120
8.4 The Spectrometry of γ - and X-radiations	121
(a) Single-crystal Methods	121
(b) Coincidence Methods	125
8.5 Studies of Short-lived Nuclear Isomers	129
8.6 Studies of Mesons and Positrons	132
8.7 Miscellaneous Applications	136
8.8 Conclusion	137
BIBLIOGRAPHY	138
INDEX	143

INTRODUCTION

1.1 THE DETECTION OF ATOMIC AND NUCLEAR RADIATIONS

WHEN a charged atomic or nuclear particle passes through matter, it causes excitation and ionisation of the molecules of the material. This ionisation is the basis of all the major instruments used for the detection and measurement of such particles. Similar instruments are also used for the detection of uncharged radiations (γ -rays, X-rays, neutrons) which can produce secondary ionising particles in their passage through matter.

The various types of detection instrument differ in the material within which the ionisation is produced, and in the manner by which it is observed. If the particle passes through a gas contained in a chamber across which a relatively small electric field is applied, the primary ions produced can be collected by the field, and the ionisation current or charge measured. This is the principle of the *ionisation chamber*. If, in a similar system, the applied field is increased, so that the primary ions formed by the particle are accelerated and produce secondary ions, which are also accelerated and produce further secondary ion multiplication before collection at the electrodes, the secondary ionisation pulse is, under suitable conditions, proportional to the primary ionisation. Such an instrument is known as a *proportional counter*. If the applied field is further increased, so that the production of a single ion-pair in the chamber is sufficient to cause a large ionisation current pulse to pass, whose magnitude is independent of the primary ionisation, the instrument is known as a *Geiger-Müller counter*. In a few liquids and crystals, the application of an electric field of suitable magnitude across the material results in a similar acceleration and collection of primary or secondary ions formed by the passage of an ionising particle. Such instruments are known as *liquid counters* and *crystal counters* respectively.

When the medium through which the ionising particle passes is a

gas, which is supersaturated to a suitable degree with vapour, due to prior adiabatic expansion and cooling, the vapour condenses in small droplets on the ions formed by the particle. This effect, discovered by C. T. R. WILSON, is the basis of the *cloud chamber* instrument, used for observing the tracks of ionising particles.

When the material through which the ionising particles passes is a photographic emulsion—a suspension of silver bromide grains in gelatine—it produces a photo-chemical reaction similar to that of light, which results in a darkening of the grains along the path of the particle, on development of the emulsion. This effect is used in the *nuclear emulsion* technique for the detection and measurement of individual ionising radiations.

When the particle impinges on certain liquid or solid materials known as phosphors, which possess the property of luminescence, part of the energy dissipated in molecular excitation and ionisation is re-emitted as visible or ultra-violet photons. The observation and measurement of the light “flashes” or scintillations produced in phosphors by individual ionising particles is the basis of the *scintillation counter* instrument. Initially the scintillations were only observed visually, but in the modern instrument they are observed and converted into voltage pulses by a photo-multiplier. In a related type of instrument, the *scintillation Geiger counter* at present under development, the scintillations are used to trigger a photo-sensitive Geiger-Müller tube.

1.2 THE VISUAL SCINTILLATION COUNTER

α -particles, which are the most intensely ionising of the radiations from radio-active materials, produce individual scintillations on an activated zinc sulphide screen sufficiently bright to be observed visually through a microscope in a darkened room. A visual scintillation counter of this type is capable of detecting and counting all α -particles incident on the phosphor screen within the field of view. Such visual observation is extremely tedious by present laboratory standards, and the counting rate is limited physiologically to about 60 scintillations per minute. Nevertheless visual scintillation counting played a very important part in nuclear research from its introduction by CROOKES and REGENER in 1908, up to the 1930's. A detailed account of this work has been given by RUTHERFORD, CHADWICK and ELLIS (1930).

The visual scintillation counter was used in the experiments which established the nature and charge of the α -particle. REGENER determined the number of α -particles emitted per second through a given solid angle by a known mass of radium or polonium, by counting the scintillations produced on a diamond screen in the focal plane of his observing microscope. He hence computed the total number of α -particles emitted per second by one gram of the radioactive substance. He then observed the total charge collected in a condenser from a known α -particle flux, and thus found the mean

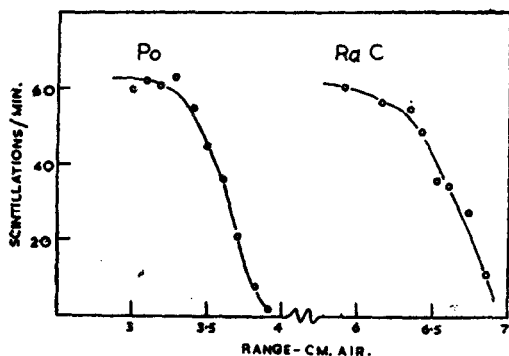


Fig. 1. Scintillation counting rate *vs* range of α -particles from Po and RaC charge on each α -particle. REGENER's value of $2e = 2 \times 4.79 \times 10^{-10}$ e.s.u. was in excellent agreement with the value of the electronic charge obtained by MILLIKAN's oil-drop experiments.

The scintillation method was used extensively for measurements of the range, absorption and straggling of α -rays. Typical curves of counting rate *vs* air range obtained for the α -particles from Po and RaC are shown in fig. 1. In the historic α -particle scattering experiments, which led to the discovery of the atomic nucleus, the relative numbers of α -particles deflected through a given angle on their passage through a thin foil, were determined by visual scintillation counting.

The visual counter is insensitive to β -rays and γ -rays, because of the weak ionisation which they produce in the thin phosphor screen. It can be used however for the detection of other particles, such as protons, whose ionisation is comparable with that of the α -particle. Thus in RUTHERFORD's discovery in 1919 of artificial nuclear disintegration, when nitrogen nuclei were bombarded with α -particles, visual scintillation counting was used to detect and measure

the range, and hence estimate the energy, of the protons produced in the reaction.

COCKCROFT and WALTON (1932) studied the disintegration of lithium nuclei by artificially accelerated protons. They used the visual scintillation technique for the detection and the measurement of the range and energy of the pair of α -particles which are the products of the $\text{Li}_3^7(p, 2\alpha)$ reaction. They employed a coincidence counting method with two observers and two ZnS screens, to show that the α -particles were of equal energy and travelled in opposite

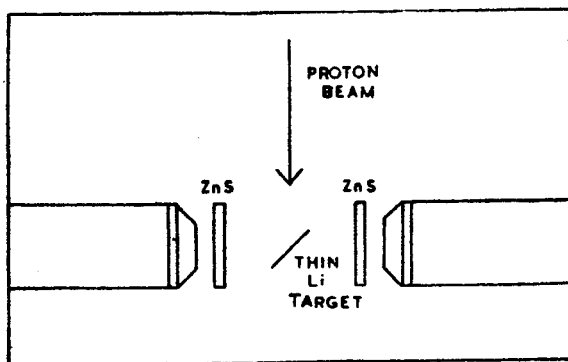


Fig. 2. Observation of $\text{Li}_3^7(p, 2\alpha)$ reaction (COCKCROFT and WALTON 1932)

directions from the target (fig. 2). The net energy E produced in the reaction was found to be equivalent to the difference in mass m between the initial and product nuclei, in agreement with EINSTEIN's relation $E = mc^2$. It is of interest to note that COCKCROFT and WALTON also used the ionisation chamber, amplifier and oscillograph method, developed by WYNN WILLIAMS and WARD, to detect the α -particles and verify their results. A new era of electronic instrumentation in nuclear physics was starting, alongside the development of machines for the acceleration of particles to high energies. By 1933, visual scintillation counting was obsolescent. Considering the major experiments performed during its useful life of 25 years, the visual scintillation counter with its zinc sulphide screen and microscope was probably the most valuable and at the same time the simplest of all nuclear instruments.

CHARITON and LEA (1929) made a comprehensive study of the visual scintillation counting process. Initially they used artificial light flashes, and they found that the minimum radiation of green light ($\lambda = 5060 \text{ \AA}$) visually perceptible is 17 quanta for a trained

observer. A rather higher minimum value of 30 quanta was obtained for α -particle scintillations observed directly with the naked eye, due to the random nature in time and position of the scintillations. The effect of the duration of the light flash on its perceptibility was investigated, and it was found that for flashes of 4×10^{-5} sec. up to 10^{-2} sec. duration, only the energy in the flash is important, and not the duration. The radiant efficiency (the percentage of incident α -particle energy converted into luminescent radiation) and the luminous efficiency (the radiant efficiency evaluated as green light, i.e. corrected for the scotopic luminosity curve of the eye) of several zinc sulphide preparations were determined, by comparison with filtered and diminished light from a standard source of known candle-power and colour temperature. High values of about 25% and 12% respectively were obtained, with some variation between different preparations.

In a typical visual scintillation counter, with a microscope of numerical aperture 0.45 and magnification 50, the faintest scintillations that could be detected were those produced by α -particles with a velocity of 2.7×10^8 cm/sec. and corresponded to 300 quanta entering the eye.

In the 1930's with the development of gas ionisation counters, in which the passage of an ionising particle produces an electrical pulse, and with the design of electronic circuits for amplifying, measuring and counting these pulses, the visual scintillation counter became obsolete. With the aid of Geiger-Müller and proportional counters, and their associated electronic circuits, it became possible to detect not only α -particles but all the other less ionising radiations which give no visible scintillations, and to count these at rates of up to 10^6 /min. Resolving times of less than 10 microsec. were obtainable, so that coincidence measurements between related ionising events could be made with this resolution.

1.3 THE PHOTO-MULTIPLIER SCINTILLATION COUNTER

Since 1947 interest has revived in scintillation counters. This is primarily due to the development of efficient photo-multiplier tubes of high gain, which are sensitive to very small light intensities. By replacing the microscope and observer of the original visual scintillation counter by a photo-multiplier, the scintillations from the phosphor are converted into amplified electrical pulses at the multiplier

output. These pulses can be fed into similar electronic circuits to those used with gas ionisation chambers.

BLAU and DREYFUS (1945) used a zinc sulphide screen and photo-multiplier. They showed that the output current of the photo-multiplier increased as an α -particle source was brought near the screen, and that this could be used as a measure of the radiation intensity. CURRAN and BAKER (1948), SHERE (1947), and COLTMAN and MARSHALL (1947) showed that the output pulses produced by the individual scintillations in such a system could be used for the efficient counting of the incident α -particles.

A major advance in the new technique was made by KALLMANN (1947) who found that the scintillations produced in large transparent blocks of naphthalene by β -rays and γ -rays could be detected with a photo-multiplier. DEUTSCH (1948) showed that such screens, several centimetres thick, gave a detection efficiency of 20% for 1.2 MeV γ -rays, and even higher for lower energy radiations. BELL (1948) discovered that anthracene was a more suitable phosphor and that it gave scintillation pulses about 5 times the amplitude of those from naphthalene. He also found that fast neutrons could be detected by the scintillations caused by recoil protons within the phosphor. HOFSTADTER (1948) discovered that sodium iodide crystals, activated with thallium, gave even larger pulses than anthracene, and that due to the high photo-electric absorption of the iodine constituent, could be used for γ -ray spectroscopy of very weak sources. He also suggested the use of lithium iodide for the detection of thermal neutrons, using the $\text{Li}_6(n, \alpha)\text{H}_1^3$ reaction within the phosphor. COLLINS (1948) measured the fluorescence decay periods of naphthalene and anthracene and found them to be of the order of 10^{-8} sec. These results indicated that the instrument had a potential resolving time of this order of magnitude. REYNOLDS, HARRISON and SALVINI (1950) reported that the scintillation pulses from certain organic solutions were comparable in magnitude with those from anthracene. Thus large volumes of transparent liquid phosphors could be used as γ -ray counters of very high efficiency.

This very brief survey of the principal developments to date indicates some of the potentialities of the photo-multiplier scintillation counter. It may be used for

- (a) The detection and energy measurement of ionising particles (electrons, mesons, protons, deuterons, α -particles, heavy ions) with a detection efficiency of 100%, under suitable conditions.

(b) The detection and spectroscopy of γ -ray and X-ray quanta with much higher detection efficiencies than a gas counter, due to the increased absorbing power of the phosphor compared with the gas and its containing walls.

(c) The detection and energy measurement of fast neutrons by the scintillations from proton recoils within an organic crystal or liquid phosphor.

(d) The detection of thermal neutrons by the scintillations produced by nuclear reactions within the phosphor.

(e) The measurement of decay times, or of intervals between related ionising events of the order of 10^{-9} sec. This represents an improvement by a factor of over 1000 compared with the resolving times of gas counters.

(f) The spectroscopy of radiations from very short-lived isomeric nuclei.

(g) The counting of radiation fluxes of the order of 10^6 /sec.

The chief disadvantages of the instrument compared with the gas counter are

(a) Its relatively low sensitivity to low-energy radiations.

(b) Its relatively poor energy resolution.

(c) The non-linearity of the energy response of organic phosphors to the more intensely ionising radiations.

Judging from the many important applications of the photo-multiplier scintillation counter to date, it will probably prove as valuable an instrument in nuclear physics as its visual ancestor.

PRINCIPLES OF THE SCINTILLATION COUNTER

2.1 INTRODUCTION

The general arrangement of the photo-multiplier scintillation counter, and the basic processes involved in the detection and measurement of an ionising particle are shown diagrammatically in fig. 3. The incident particle impinges on the phosphor where it

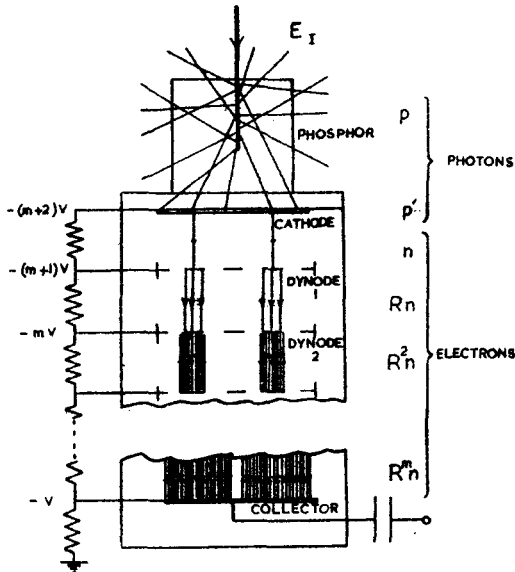


Fig. 3. Schematic diagram of photo-multiplier scintillation counter

dissipates its energy in the ionisation and excitation of the molecules. A fraction of this energy is converted into photons which are radiated in all directions. Some of these photons fall on the photo-cathode of the multiplier tube, and eject a number of photo-electrons. These photo-electrons are accelerated by the potential applied between the

cathode and the first electrode (or dynode) of the tube. On striking the first dynode, each photo-electron ejects several further electrons by secondary emission. This electron multiplication process is repeated at subsequent dynodes, each of which is at a higher potential than the preceding one. Finally after a multiplication of about 10^6 to 10^9 the electron avalanche arrives at the collector plate. It produces a voltage pulse in the output condenser which is applied to an external pulse-recording circuit. Thus the initial energy of a single ionising particle is transformed into a single voltage pulse, which may be used for detection and measurement. The whole system is enclosed in a light-tight box, to eliminate effects other than those due to incident ionising radiation.

The operation of the scintillation counter can be divided into five distinct, consecutive stages—

- (a) The absorption of the incident radiation by the phosphor.
- (b) The luminescent conversion of the energy dissipated in the phosphor, and the emission of photons.
- (c) The transit of the emitted photons to the cathode of the photo-multiplier.
- (d) The absorption of the photons at the cathode, and the emission of the photo-electrons.
- (e) The electron multiplication process.

At each stage various factors enter which affect the relation of the output pulse to the incident radiation, and hence influence the characteristics of the instrument. These factors will be enumerated in this chapter, prior to a more detailed discussion.

2.2 ABSORPTION OF THE INCIDENT RADIATION

A charged particle incident on the phosphor dissipates its energy continuously in its passage through the material. The energy E_i of the incident particle may be completely absorbed in the phosphor, or the particle may emerge, having dissipated only a fraction A of its energy. The amount $E_i A$ expended depends on the dimensions of the phosphor compared with the maximum range r of the particle in the material. In the simplest case of a heavy particle incident normally on a phosphor, thickness d , and travelling through it in a straight line, $A = 1$ if $d \geq r$. The range r depends on the

nature and energy of the incident particle, and on the stopping power of the phosphor, which is a function of its density and the atomic number and mass of its constituent elements. These factors will be considered when the properties of individual phosphors are discussed.

Uncharged radiations—X- and γ -ray quanta, and neutrons—do not ionise directly. They can, however, transfer the whole or more usually part of their energy to individual charged particles within the phosphor, and the ionisation produced by these secondary particles gives rise to the scintillations used to detect the primary radiation. When the primary radiation is charged there is a potential 100% detection efficiency, since every incident particle dissipates the whole or part of its energy within the phosphor. When it is uncharged, only a fraction f of the incident quanta or neutrons produce secondary ionising particles within the phosphor, and the detection efficiency is correspondingly reduced. For a beam of uncharged radiation incident normally on a phosphor, thickness d

$$f = 1 - e^{-kd} \quad (1)$$

where k is the absorption coefficient of the phosphor for the radiation.

There are three alternative processes by which X- or γ -ray quanta can transfer energy to electrons within the phosphor—

- (a) Photo-electric absorption.
- (b) Compton scattering.
- (c) Pair production.

k is made up additively of the absorption coefficients k_{Ph} , k_C and k_{PP} corresponding to these three processes, so that

$$k = k_{Ph} + k_C + k_{PP} \quad (2)$$

The magnitudes of these three absorption coefficients depend on the energy of the incident radiation, on the density ρ of the phosphor and on the atomic number and mass of its constituent elements. At low energies k_{Ph} is the largest component, but it decreases rapidly as the energy is increased. With heavy elements however it may be still appreciable at energies of a few MeV. k_C remains nearly constant from 0 to about 0.5 MeV and then decreases with increasing energy. k_{PP} is zero at energies below 1.02 MeV, and then increases steadily with increasing energy. Typical absorption coefficient curves for a

light element Al and a heavy element Pb are shown in fig. 4. The relevant values of k for the various phosphors will be discussed later.

The energy transferred by the incident quantum to the secondary electron depends on which of the three absorption processes is operative. In photo-electric absorption, the whole of the incident quantum energy E_I is transferred to the electron and its accompanying X-ray, which is usually also absorbed in the phosphor. In

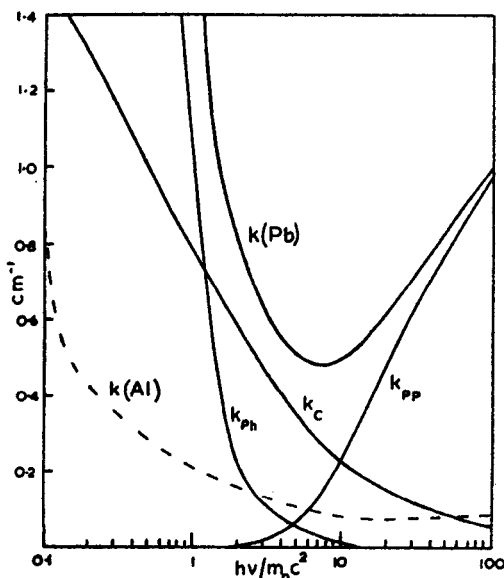


Fig. 4. γ -ray absorption coefficients of lead (—) and aluminium (-----)

Compton scattering, the electrons produced by quanta of energy E_I have a continuous energy distribution from zero up to a maximum E_C , where

$$E_C = \frac{E_I}{1 + m_0c^2/2E_I} \quad (3)$$

and $m_0c^2 (= 0.51 \text{ MeV})$ is the rest-energy of the electron. The remainder of the incident energy, in the form of the scattered quantum, normally escapes from the phosphor. In pair-production absorption of quanta of energy E_I , $2m_0c^2 (= 1.02 \text{ MeV})$ of the energy is expended in the creation of the mass of an electron-positron pair, and these two particles share the residual energy,

$$E_{PP} = E_I - 2m_0c^2 \quad (4)$$