# SCINTILLATION COUNTERS

Scintillation counters employ a phenomenon known as luminescence: a particle passing through a medium excites or ionizes atoms along its path, and, upon deexciting, these atoms emit photons. Such a medium, whether solid, liquid, or gaseous, which has the property of emitting luminous light, is called a scintillator.

#### HISTORY OF INSTRUMENTS

Sir William Crookes built the first scintillation counter in 1903, employing a microscope for visual observation of light flashes from a zinc sulfide screen. The physiological limit of this tiring procedure is a counting rate of about one per second (1). Nevertheless, this setup was used in 1909 by Geiger and Marsden (2) to study the scattering of  $\alpha$  particles and resulted in Rutherford's discovery of the atomic nucleus (3). Visual scintillation counters were used up to the 1930s. In 1941 Krebs (4) invented the first electrical counter for scintillation events in the form of a Geiger-Müller tube sensitive to luminous photons. This device had a response time of about  $10^{-4}$  s, which is typical for gas counters. The application of the scintillation method greatly increased in the 1950s after the development of the photomultiplier in 1944 by Curran and Baker (5), which enabled the automation of the tedious manual counting and at the same time allowed much higher

counting rates. Another advantage over the previously used zinc sulfide screen came in 1947 and 1948 with the discovery of luminescent materials that are transparent to their own scintillation light, such as naphthalene by Kallmann (6), an-thracene by Bell (7), and thallium-activated crystalline so-dium iodide by Hofstadter (8). It was found that, due to their transparency, these materials can be used as scintillators in large volumes to detect even the only weakly interacting  $\gamma$  rays. Moreover, naphthalene and anthracene exhibit pulse decay times on the order of  $10^{-8}$  s and hence, in conjunction with a photomultiplier, allowed the construction of detectors that were much faster than any gas-filled counter.

### **APPLICATIONS**

Today, scintillation counters are widely used for the detection of  $\alpha$  and  $\beta$  particles,  $\gamma$  rays, and neutrons in many fields, such as in high-energy physics research, environmental radiation protection, dosimetry, and medicine. They can be operated (1) as pulse counters to measure the activity of a radioactive source or the dose received by food items or biological tissue, (2) as spectrometers to determine the energy of the incident particles mentioned, and (3) as timing devices for the investigation of cosmic rays or for lifetime measurements on unstable particles. Examples of applications are monitoring the exhalation of the radioactive noble gas radon from soils and rocks and examining beam profiles in accelerators. Scintillating materials are also used as screen coatings—for example, in television picture tubes.

When compared to other counter types, scintillation counters show the highest counting efficiency, because of their large volumes and their high absorption. Scintillation counters are also much faster than gas counters and semiconductor counters and can handle counting rates of up to  $10^8 \text{ s}^{-1}$ . However, semiconductor counters show better energy resolution (below 1%) than scintillation counters (on the order of 10%). The performance characteristics of scintillation counters are discussed in detail later on.

A detailed treatment of the applications of scintillators as well as the physical processes therein is contained in the standard work by Birks (1). The usage of scintillation counters in high-energy physics experiments such as coincidence measurements is described in Ref. 9. A comprehensive introduction into the detection of nuclear radiation with various detector types can be found in Refs. 10 and 11.

#### STRUCTURE OF A COUNTING SYSTEM

A modern scintillation counter (as shown in Fig. 1) consists of three main parts: a scintillator, a photomultiplier, and signal-processing electronics.

In the scintillator the energy of the incident particle is converted into light pulses. This scintillation light travels through the scintillator volume and eventually hits the photocathode of the photomultiplier, where it releases photoelectrons. In order to increase the number of photons incident on the photocathode, the scintillator can be covered with a reflecting layer.

In the photomultiplier, a system of electrodes, called *dy*nodes, accelerates the electrons towards the anode by means of a high voltage. Upon impact on a dynode, each electron can release additional electrons, a process called *secondary*  emission. Hence, the number of electrons in the pulse is multiplied between subsequent dynodes, leading to a large current at the anode. Since the photomultiplier can easily be destroyed if this current is too large, both the scintillator and the photomultiplier have to be screened from ambient light (see Fig. 1). Good optical contact has to be made between them in order to minimize optical losses due to total reflection. A light-guiding structure can also be inserted if the geometry of the application requires the scintillator and the photomultiplier to be at some distance from each other. The anode pulse of the photomultiplier is fed into a pulse amplifier. In order to minimize background noise, the connection between the photomultiplier and the amplifier has to be kept as short as possible. Therefore, the amplifier is usually contained in a housing, which is directly connected to the photomultiplier socket. In most cases, this housing also contains a voltage divider circuit, which distributes the high voltage to the dynodes of the photomultiplier.

The final constituent of a complete detection system is the signal-processing and data acquisition electronics. The design of the signal-processing electronics depends on the type of application. For measurements of the particle energy, the amplifier output signal is connected to a pulse discriminator/digital counter or a multichannel pulse height analyzer. In measurements requiring a low background, two or more photomultipliers are coupled to the same scintillator. Their pulse amplifiers are connected to a coincidence circuit, which passes a signal to a counter only if the pulses delivered by different photomultipliers arrive within the resolving time of the circuit. In this way, random, spurious pulses generated independently in the photomultipliers can be excluded. In many cases, a digital counter or a multichannel pulse height analyzer is connected to a digital data storage system, for example a computer, where the counts are stored to memory.

The physical processes leading to scintillation are dependent on the scintillator material. The nature of these processes also determines important performance characteristics of scintillation counters, such as the *response time*, the *sensitivity*, the *proportionality*, and the *energy resolution*. This is described in the following sections. A more detailed treatment of these aspects can be found in Refs. 1 and 12.

### SCINTILLATOR MATERIALS

Scintillators can be classified into inorganic crystals, glasses and gases, and organic compounds.

The most commonly used inorganic materials are alkali halides such as NaI, CsI, or KI. These are doped with about 0.1% of thallium, which acts as a center for the emission of luminescent photons. Thallium-activated sodium iodide, NaI(Tl), is the most commonly used scintillator; it can be produced in large crystals. Zinc sulfide crystals doped with silver, ZnS(Ag), are used as a scintillating coating. Silicate glasses scintillate when doped with lithium or cerium.

Among the organic substances are anthracene and stilbene crystals, and terphenyl dissolved in liquids (for example xylene or toluene) or incorporated in plastics (for example polystyrene).

Finally, the noble gases helium, neon, argon, krypton, and xenon can be used as scintillators.

The wavelength of maximum intensity of the emitted scintillation light varies among different scintillator materials





and is typically between 300 nm and 500 nm for inorganic crystals and organic compounds, and in the ultraviolet region for noble gases.

# **PROCESSES IN SCINTILLATORS**

Producing a scintillation event in one of the materials mentioned involves two steps: the interaction of an incident particle and the subsequent emission of luminescent light.

In general, a part of the energy of the incident particle is absorbed, causing excitation or ionization of scintillator molecules. Luminescent light is emitted during transitions of excited electrons to lower energy levels. The interaction depends on the type of the particle, and the emission process varies between different scintillator materials. Incident particles can be either primary or secondary, examples of the latter being photoelectrons and Compton electrons produced by a  $\gamma$  ray, and secondaries of neutrons. In addition to luminescent light, Čerenkov radiation can be emitted by a charged particle traversing matter at a speed greater than the speed of light in the respective medium. However, this effect will not be treated in this article.

The energy loss of charged particles in matter, as given by the Bethe-Bloch formula, and the probability for photoelectric absorption, Compton scattering, and pair production by  $\gamma$  rays all increase with increasing density  $\rho$  and atomic number Z of the scintillator. Therefore, materials with high values of  $\rho$  and Z show high absorption of incident particles per unit volume. In this respect, inorganic crystals (3.0 g cm<sup>-3</sup> <  $\rho$  < 7.5 g cm<sup>-3</sup>; e.g. Z = 53 for iodine) have advantages over organic compounds (0.8 g cm<sup>-3</sup> <  $\rho$  < 1.2 g cm<sup>-3</sup>;  $Z \leq 6$ ) and noble gases ( $\rho \leq 10^{-3}$  g cm<sup>-3</sup> at atmospheric pressure and room temperature). However, because of their low absorption efficiency for  $\gamma$  rays, organic scintillators are predominantly used for the measurement of charged particles at low  $\gamma$  background counting rates.

Charged particles usually deposit all their energy, especially in solid or liquid scintillators, because the range of  $\alpha$ particles and electrons in these scintillators is in the order of micrometers and millimeters, respectively. In contrast,  $\gamma$  rays attenuate exponentially by a factor of two over several centimeters and more, and it is therefore possible that they leave only a fraction of their energy in the scintillator. This circumstance is reflected in characteristic features in the spectra observed with scintillation counters suitable for detecting  $\gamma$  rays. This will be briefly explained in the following (with high Z). If the detector is dense (with high Z) and large enough, some  $\gamma$  rays can be totally absorbed in the scintillator due to the photoelectric effect. It is also possible that they undergo a number of Compton scattering processes before leaving the scintillator, which implies that only the energy of the Compton electrons is transformed into scintillation light. Finally, a  $\gamma$  ray with an energy of more than two electron rest masses (1.022 MeV) can produce an electron-positron pair. The positron is likely to recombine with electrons in the scintillator, producing two 0.511 MeV photons, one or both of which can leave the scintillator.

Figure 2 shows a typical count spectrum of monoenergetic  $\gamma$  radiation measured with a scintillation counter. The peak denoted by  $E_{\gamma}$  contains events where  $\gamma$  rays deposit their total energy in the scintillator, which includes photoelectric absorption and positron annihilation without the escape of an annihilation photon. The broad spectrum at energies below  $E_{\gamma}$  is the Compton distribution, resulting from scattering processes involving variable amounts of energy transfer. Within this distribution, there are two distinct peaks at  $E_{\gamma} - 0.511$ MeV and  $E_{\gamma}$  – 1.022 MeV, which are due to the escape of one or two annihilation photons. It should be pointed out that both the Compton distribution and the single and double escape peaks decrease in height with increasing absorption (that is, with increasing atomic number, density and volume of the scintillator). Hence, a  $\gamma$  spectrum of many different energies is easier to interpret when measured with a large and highly absorbing detector.



**Figure 2.** Energy spectrum of  $\gamma$  rays obtained with a scintillation counter.

Neutrons can be detected with scintillation counters by means of secondary reaction products. In (hydrogen-containing) organic scintillators, for example, fast neutrons can produce recoil protons in a collision with hydrogen nuclei (*hydrogen knock-on* reaction). Boron trifluoride (BF<sub>3</sub>) counters can detect slow neutrons via nuclear capture according to <sup>10</sup>B +  $n \rightarrow {}^{7}\text{Li} + \alpha + 2.78$  MeV (11). Europium-activated lithium iodide [LiI(Eu)] crystals have also been successfully used for the  ${}^{6}\text{Li}(n,\alpha){}^{3}\text{H}$  reaction.

It is possible to distinguish between two types of luminescent emission, depending on how energy is transferred from the excited particle to the luminous photon. A process is called *phosphorescence* if it involves deexcitation of scintillator molecules from relatively long-lived, metastable energy levels. If this is not the case, the process is termed *fluorescence*. Historically, these two variants were distinguished by the typical *relaxation time*  $\tau_r$  of the pulses, which characterizes the decrease of the scintillation intensity I(t) (number of luminous photons per unit of time t). This decrease can be approximated by

$$I(t) = I(t=0)\exp(-t/\tau_{\rm r}) \tag{1}$$

These relaxation times are on the order of  $10^{-6}$  s to 1 s for phosphorescence and  $10^{-7}$  s to  $10^{-9}$  s for fluorescence. For further details about processes in scintillators, see Ref. 1.

# **PERFORMANCE CHARACTERISTICS**

The type of emission process depends on the scintillator material. Since a typical photomultiplier spreads a pulse by about  $10^{-9}$  s, the choice of scintillator determines the response time of the whole detector. Organic scintillators and noble gases, for example, show fluorescence, and are therefore predominantly used for applications requiring a fast response. In inorganic crystals, the energy deposited by the incident particle has to be transferred thermally to the luminescent centers (thallium or silver atoms). This leads to longer relaxation times, or, if these luminescent centers have metastable energy levels, to phosphorescence. The scintillation processes in inorganic and organic scintillators and their influence on the response time are described in detail later on.

The sensitivity S of a scintillation counter can be written (see Refs. 12 and 13) as the product

$$S = \eta_{\rm s} T_{\rm s} f_1 s_\lambda f_2 \eta_{\rm c} G \tag{2}$$

of (1) the quantum yield or conversion efficiency  $\eta_s$  (the ratio  $E_{\rm s}/E_{\rm p}$  of the energy  $E_{\rm s}$  of the emitted luminescent photons to the energy  $E_{\rm p}$  deposited in the scintillator by the incident particle), (2) the transparency  $T_s$  of the scintillator to its own scintillation light (dependent on the distance traveled through the scintillator volume), (3) a factor  $f_1$  representing the optical loss due to reflection and absorption in the windows of the scintillator and the photomultiplier as well as the optical contact between them, (4) the maximum spectral sensi*tivity*  $s_{\lambda}$  of the photocathode (the ratio of the photoelectron current to the power of luminescent photons), (5) a factor  $f_2$ for the spectral match of the photocathode material to the wavelength of the scintillation light, (6) the collection efficiency  $\eta_c$  of the first photomultiplier dynode for photoelectrons, and finally (7) the photomultiplier gain G. All quantities mentioned above are dimensionless, except for the spectral sensitivity  $s_{\lambda}$ , which has units of milliamperes per watt. The overall sensitivity *S* can thus be expressed as the current at the photomultiplier output per unit energy flux of the incident particle current. It has units of mA/(MeV  $s^{-1}$ ).

The output pulse of a scintillation counter shows proportionality to the energy  $E_p$  of the incident particle only if this energy is completely deposited in the scintillator and if the conversion efficiency  $\eta_s$  is constant. In this case, the pulse charge released in the photomultiplier from the last dynode to the anode is determined by the sensitivity S of the counter according to

$$Q = SE_{\rm p} \tag{3}$$

It is this charge Q that is measured with the signal-processing electronics and then multiplied by the counter sensitivity S (usually obtained from calibration) in order to obtain  $E_{\rm p}$ . However, in some circumstances [as in measuring  $\gamma$  rays with a small NaI(Tl) crystal or  $\alpha$  particles with a scintillation chamber, as will be discussed later], only a variable fraction of  $E_{\rm p}$  is deposited in the scintillator. In this case, the charge collected at the photomultiplier anode does not reflect  $E_{\rm p}$ , but only the fraction deposited.

The energy resolution describes the uncertainty occurring in the energy measurement described above, and hence is a measure of the ability of the counter to distinguish two different particles even if their energies  $E_{p}$  are very close together. It can be obtained from a measured pulse height or particle energy spectrum as the ratio of the full width of the corresponding peak at half maximum height to the energy  $E_{\rm p}$ . The spread of a pulse height or energy peak is due to two factors: (1) statistical fluctuations in the numbers  $N_{\gamma}$ ,  $N_{p}$ , and  $N_{s}$  of released luminous photons, photoelectrons, and secondary electrons, and (2) variations in the counter sensitivity S. Since the relative statistical fluctuations are proportional to  $1/\sqrt{N}$ , and since  $N_{
m p} < N_{
m \gamma}$  and  $N_{
m p} < N_{
m s}$ , the largest fluctuations are introduced during the release of photoelectrons. Furthermore, the light collection (and hence the transparency  $T_{s}$ ) is not uniform over the scintillator volume, and the photomultiplier gain G fluctuates due to instabilities in the high voltage (14), both of which affect the counter sensitivity. For the detection of 1 MeV  $\gamma$ -rays with a NaI(Tl) crystal, for example, the energy resolution is in the order of 7%.

# DETECTOR TYPES AND ACCESSORIES

This section will deal with the most common types of scintillation counters, such as inorganic and organic scintillation counters and crystal, liquid, and gas scintillation counters, as well as scintillation chambers. In addition, details on photomultipliers and optical contacts are presented. Since the early visual and Geiger-Müller scintillation counters have no relevant applications anymore, they will not be described here. A detailed treatment of these types can be found in Refs. 1 and 15.

#### **Inorganic Scintillators**

Inorganic scintillators can be classified into three main groups: crystals, glasses, and gases (including liquefied noble gases). The most commonly used are the crystal scintillators, which are mainly alkali halides. Inorganic scintillators are generally characterized by high quantum yield (and therefore

Scintillator	Light Output [% of NaI(Tl) for Bialkali PMT]	Wavelength of Maximum Emission (nm)	Decay Time (ns)	$\begin{array}{c} \text{Density} \\ (\textbf{g} \cdot \textbf{cm}^{-3}) \end{array}$
NaI(Tl)	100	415	230	3.67
CsI(Na)	80-85	430	630	4.51
CsI(Tl)	45 - 50	530	1000	4.51
CsI(pure)	4-8	310	$\sim 8$	4.51
BGO	10-20	505	300	7.13
$BaF_2$	20	310	630	4.88
$CaF_2(Eu)$	50	435	940	3.19
CsF	5	390	5	4.11
ZnS(Ag)	130	450	70	4.09
LiI(Eu)	30 - 35	470	1400	4.08
GSO(Ce)	20	440	60	6.71
Ar	4	250	20	$1.8 imes10^{-3}$
Xe	$\sim 20$	330	20	$5.9 imes10^{-3}$

Table 1. Physical Properties of Some Common Inorganic Scintillators

good resolution and sensitivity), high density, and high Z (and therefore high efficiencies). See Table 1 for the physical properties of some common inorganic scintillators. They are therefore ideal as high-energy-electron and  $\gamma$ -ray detectors, and they are used as such in medical imaging applications,  $\gamma$ -ray spectrometry, and radiation dosimetry and as general-purpose radiation detectors for nuclear and particle physics research.

Scintillation Mechanism. Details of the scintillation mechanism for inorganic crystal scintillators vary from one material to another, but the general principle is similar. The scintillation mechanism depends on excitation and deexcitation of electrons between energy states determined by the crystal lattice of the material. The large number of atoms in the crystal leads to energy *bands* (consisting of large numbers of very closely spaced energy levels). Most electrons are in the valence band (ground state). A charged particle passing through the material will excite some of the electrons to the *conduc*tion band (excited state). The electron may emit a scintillation photon and return to the valence band or lose its energy to nonradiative thermal motion. In a pure crystal the scintillation photon can be readily absorbed, since it corresponds to the energy difference between the valence and conduction bands and there are many electrons in the valence band. Absorption leads again to an excited electron that can either emit a photon or lose its energy to nonradiative thermal motion. At each stage further energy is lost to the nonradiative process, so generally pure crystals are not efficient scintillators.

This problem is solved by adding small amounts of impurities, called *activators*, to the crystals. The activators modify the energy band structure of the crystal by introducing activator ground states that are at a higher energy than the crystal valence band and excited states that are at a lower energy than the conduction band; see Fig. 3. When an electron is excited from the valence band to the conduction band, a hole (vacancy) is formed in the valence band. The electron can move freely through the crystal, whilst the hole can do the same. The hole will quickly drift to a site in the crystal lattice occupied by an activator atom. An electron from the ground state of the activator atom will drop down to the valence band of the crystal, releasing thermal energy. The excited electron (in the conduction band) can now drop down to the excited activator state, also releasing thermal energy. It then emits a scintillation photon as it deexcites to the activator ground state. This has two important consequences. First, the energy of the scintillation photon is less than that required to excite a valence electron (most electrons) to the conduction band, so it is not readily absorbed. The scintillator is therefore transparent to the photons it emits. Second, the energy difference between the ground and excited activator states corresponds to photons in the visible range that are better matched to the spectral response of photomultiplier tubes. The energy difference between the valence and conduction bands is higher, corresponding to ultraviolet (UV) photons.

Under certain conditions pure (or intrinsic) crystals can, however, be used as efficient scintillators. The difference in the emission and absorption spectra for scintillation photons is attributed to the Franck–Condon principle, which states that any transition between electron energy states happens very quickly compared to atomic or ionic vibrations. This is important because the minimum energy for the conduction band occurs at a larger interionic separation than for the valence band. When an electron is excited to the conduction band, the interionic separation initially remains the same. This means the electron is left with excess energy. It loses it as thermal energy to the crystal lattice while the interionic separation increases. It then emits a scintillation photon and deexcites to the upper levels of the valence band. The energy of this scintillation photon is slightly less than the energy re-



**Figure 3.** Energy band structure of an activated inorganic scintillator. The presence of activator sites introduces new energy levels. Transitions between these levels lead to photons that are not readily absorbed by the scintillator.

quired to excite an electron from the valence band to the conduction band. Pure crystals are efficient only if the number of electrons in the higher valence states is small, and this is usually achieved by cooling the crystals. At room temperature electrons can be found in the higher valence state due to thermal motion and cause reabsorption of the scintillation photons. As long as the pure crystals are cooled, their quantum yield is high and pulse decay time short (16).

In some cases the excited electron is left in a state that cannot decay directly to the ground state (forbidden transition). The electron must obtain some extra energy (usually from thermal excitation) and move to a higher state from which it can decay to the ground state. This process is slow, and the emission is known as phosphorescence.

Crystal Scintillators. By far the most common general-purpose  $\gamma$ -ray detectors are the alkali halide crystals. Sodium iodide crystals with 0.1% thallium as an activator, NaI(Tl), are readily available commercially and are the standard detector used for comparisons with other scintillators. The notable features of NaI(Tl) are its high quantum yield, good energy resolution, and linear response over a large energy range of incident radiation. Unfortunately, it is also hygroscopic, which requires the crystal to be hermetically sealed, usually together with the photomultiplier tube. If not sealed, the crystal quickly deteriorates due to the absorption of moisture in the air. It also has a relatively long scintillation decay time of 230 ns, which may be a problem for very high counting rates, and a phosphorescence component with decay time of 0.15 s. In most applications the phosphorescence is not important, since each phosphorescence photon is detected as a single event giving rise to a single photoelectron. Such a small signal can be ignored by using a discriminator set to the appropriate level. NaI is also fragile and can be easily damaged by mechanical or thermal shock.

For low-energy  $\gamma$  rays the efficiency of a scintillation detector can be increased by using a *well crystal*. This is a circular cylinder with a well, or cavity, along the axis of the cylinder. The source or sample to be measured is placed inside the well, and the scintillator almost completely surrounds it. The counting efficiency is therefore close to 100%.

Cesium iodide is also a popular, readily available scintillator, activated with either sodium or thallium, CsI(Na) and CsI(Tl). Both types are slightly denser than NaI(Tl), resulting in a larger absorption coefficient. They are only slightly hygroscopic and not as susceptible to shock and vibration. A useful property of CsI(Tl) is its variable decay time for different incident particles (17). This allows *pulse shape discrimination* between heavy charged particles such as  $\alpha$  particles and protons on the one hand and electrons and  $\gamma$  rays on the other. However, the emission spectrum is at a much longer wavelength than NaI(Tl) and is poorly matched to the spectral response of most photocathodes. The response is greatly improved by using photodiodes (semiconductor devices) that are sensitive to these longer wavelengths.

Lithium iodide activated with europium, LiI(Eu), and enriched with <sup>6</sup>Li is often used for the detection of slow neutrons. A slow neutron interacts with <sup>6</sup>Li in the crystal and produces tritium and an  $\alpha$  particle. The total energy of the reaction products is fixed, and so the response is characterized by a well-defined peak. However, this response also corresponds to that for electrons or  $\gamma$ -rays of energy 4.1 MeV. Therefore, any electron and  $\gamma$ -ray background at these energies is important when making neutron measurements. Lithium iodide is also highly hygroscopic and must be sealed.

A high-density pure crystal scintillator that is commonly used is bismuth germanate (BGO),  $Bi_4Ge_3O_{12}$ . Its high density is an advantage in applications where a compact high-efficiency counter is required. It is also mechanically and chemically robust and therefore easy to handle. The scintillation emission is characterized by a fast component with a decay time of 60 ns and a dominant slower component of 300 ns making its timing characteristics similar to NaI(Tl). The quantum yield, however, is only about 10% to 20% of that of NaI(Tl), and so the resolution is much poorer. This means it is appropriate for counting applications, such as PET positron emission tomography) scanners, but unsuitable for spectrometry.

One of the first scintillators used was zinc sulfide activated by silver, ZnS(Ag). Unlike other crystal scintillators, it can only be produced as a polycrystalline powder rather than a single crystal. It is opaque to its own scintillation light, so it is used as a thin layer or screen between 20  $\mu$ m and 60  $\mu$ m thick. It is primarily used for counting very low-energy electrons,  $\alpha$  particles, or other highly ionizing charged particles. Its quantum yield is comparable to that of NaI(Tl), making it very efficient. It is sometimes used to coat layers of plastic, or some other material high in hydrogen, in order to detect recoil protons due to incident fast neutrons (16). If the plastic contains <sup>6</sup>Li or <sup>10</sup>B, it can also detect slow neutrons.

**Glass Scintillators.** Most glass scintillators are made from silicate glasses and activated by cerium. The decay time for the scintillation emission is typically between 50 ns and 100 ns. They can be produced cheaply and easily in many different shapes and sizes, and they are mechanically and chemically very robust, making them suitable for extreme environments such as high temperatures or the presence of corrosive chemicals. Their quantum yield is only about 10% of that of NaI(Tl), so they are not suitable for spectrometry. Glass scintillators containing lithium are widely used as fast-neutron detectors. Their short response time means they are suitable in neutron time-of-flight spectroscopy.

Gas Scintillators. Gases can also be made to scintillate and serve as useful detectors for certain applications. The most efficient scintillating gases are the noble gases and nitrogen (18). Several different processes give rise to the scintillation light, of which the most efficient is the formation of excited molecules that radiate when returning directly to the ground state. For the noble gases the emission spectrum is usually in the UV and forms a continuum. To detect such photons requires the use of special UV-sensitive photocathodes, photodiodes, or wavelength shifters, such as a small amount of nitrogen gas. The quantum yield is quite low compared to that of NaI(Tl), but the decay time for the emission is generally short, in the order of 20 ns or less. The quantum yield also depends on the purity of the gas, as any impurities can reduce the light output. This is because the impurities absorb the excitation and ionization energy without emitting scintillation photons. Such a process is termed quenching. The response of gas scintillators is also very linear with energy over a large range. At low pressures ( $<10^5$  Pa = 1 atm) they have very low efficiencies for  $\gamma$  rays, so that they are used almost

Table 2.	Physical	<b>Properties</b>	of Some	Common	Organic	Scintillators

Scintillator (Solvent–Primary Solute– Secondary Solute)	Light Output [% of NaI(Tl) for Bialkali PMT]	Wavelength of Maximum Emission (nm)	Decay Time (ns)	Density $(g \cdot cm^{-3})$
Crystals				
Anthracene	40	440	30	1.25
trans-Stilbene	20	410	6	1.16
Plastics				
Polystyrene-tetraphenylbutadiene	14	450	5	$\sim 1.0$
Polyvinyltoluene- <i>p</i> -terphenyl- <i>p</i> , <i>p</i> '-diphenylstilbene	19	380	3	$\sim 1.0$
$\label{eq:polyconstraint} Polyvinyl to luene-p-terphenyl-tetraphenyl but adiene$	18	445	4	$\sim 1.0$
Liquids				
Toluene-PPO	20	365	3.8	0.88
Toluene-p-terphenyl-POPOP	24	425	3.5	0.88
Toluene-PPO-POPOP	30	425	3.7	0.88

exclusively as heavy-ion detectors, most notably for fission fragments. In most practical applications the gas is used at high pressure, increasing the efficiency of the detector.

Gas scintillators are also used in a hybrid detector called the *scintillating proportional detector* (19). A large electric field between two electrodes accelerates any electrons produced from ionizing interactions in the gas. As these electrons move towards the anode, they collide with neutral gas molecules, exciting them. The gas molecules deexcite, emitting scintillation photons. This is a source of secondary scintillation. The quantum yield is greatly increased, approaching or even exceeding that of NaI(Tl), but the drift of the electrons is slow and the secondary scintillation photons are emitted a few microseconds after the prompt scintillation photons and last for a few microseconds, resulting in a relatively slow response.

The noble gases also scintillate when condensed cryogenically to a liquid or solid. In fact, their quantum yield is then comparable to that of NaI(Tl). Liquid argon (LAr) ionization chambers have been used for many years, but only recently has the need for fast detectors prompted their use as scintillators because of their fast scintillation response. Liquid xenon and krypton detectors, which have high Z, can also be used as  $\gamma$ -ray detectors. Being liquid, they can be produced in any shape and size, which is also advantageous.

# **Organic Scintillators**

Organic scintillators can be classified into three types: crystal, liquid, and plastic. Compared to the inorganic scintillators, they have lower densities and Z values, resulting in lower detection efficiencies, especially for  $\gamma$  rays. The quantum yield is also lower, resulting in poorer energy resolution. (See Table 2 for the physical properties of some common organic scintillators.) They do, however, have several advantages. They are very cheap, can be produced in many shapes and sizes, and are mechanically robust. They are also characterized by extremely short response times, whence their use in fast timing applications and at high event rates. Addition of boron or gadolinium produces scintillators that can be used for neutron detection and spectrometry.

Scintillation Mechanism. The luminescence process in organic scintillators is due to energy transitions in individual molecules, rather than the lattice structure of the material as in inorganic crystals. Any particular molecule can therefore be observed to scintillate, regardless of its physical state (solid, liquid, or gas). Almost all organic scintillators contain at least one benzene ring (or benzene-ring derivative) and it is this structure that is responsible for the scintillation.

Benzene is an organic molecule that contains six carbon atoms arranged in a closed ring. Each carbon atom forms four bonds with its four valence electrons. Three of these bonds are  $\sigma$  bonds that are close to the carbon atom and hence strong bonds. The fourth electron forms a  $\pi$  bond that is further away and only weakly bound. This electron is therefore easily excited or ionized and is responsible for the scintillation mechanism. The  $\pi$  electron is usually found in the ground state, but it can be excited into any of the higher energy states. The structure of these electronic states is shown in Fig. 4. It consists of the basic electronic states  $S_0$  (the ground state),  $S_1$  (first excited state),  $S_2$  (second excited state), and so



**Figure 4.** Energy levels of an organic molecule with  $\pi$  electron structure. Electron transitions are indicated for fluorescence and phosphorescence.

on. The typical difference in energy between  $S_0$  and  $S_1$  is about 3 eV or 4 eV. These states are further split into substates with slightly different energies due to vibration of the molecule, the difference between adjacent states being about 0.15 eV. They are denoted by  $S_{00}$ ,  $S_{01}$ ,  $S_{10}$ ,  $S_{11}$ , and so on. These states are actually split even further by different molecular rotational energy substates, but the difference of such energy levels is much smaller and can be ignored when considering scintillation. Together these energy levels form the singlet states (spin 0) for the  $\pi$  electron. There is a similar set of states called the triplet states (spin 1) that the  $\pi$  electron can also occupy. These are indicated as  $T_1$ ,  $T_2$ ,  $T_3$ , and their splitting has not been included in Fig. 4 for simplicity.

Three types of luminescence have been identified for organic scintillators that differ in either their emission spectrum or their decay time. They are fluorescence, phosphorescence, and delayed fluorescence. The most important process that is responsible for most of the scintillation light emission is fluorescence.

The spacing of the vibrational and rotational substates means that at room temperature nearly all the molecules of a scintillator are in the  $S_{00}$  state. A charged particle incident on the scintillator can either excite such a molecule to any of the higher energy level states or ionize it. An excited molecule quickly ( $\sim 10^{-12}$  s) loses energy through internal nonradiative processes until it reaches one of the  $S_1$  substates and then thermally deexcites to the  $S_{10}$  state. The molecule then emits scintillation light (fluorescence) as it deexcites from the  $S_{10}$ state to one of the  $S_0$  substates. This process occurs on a time scale of nanoseconds and determines the decay time of fluorescence. Since the higher vibrational substates of the ground state are not occupied, the emission and absorption spectra overlap only slightly and the scintillator is transparent to its own emission. An ionized molecule, on the other hand, usually recombines with the electron and ends up in one of the excited states. In most cases this is a triplet state.

In some cases the excited  $S_{10}$  state can undergo a nonradiative transition to the  $T_1$  triplet state. This is called *inter-system crossing*. The lifetime of the  $T_1$  state is characteristically much longer  $(10^{-4} \text{ s})$  than that of the  $S_1$  state. The molecule deexcites to an  $S_0$  state by emitting light that is usually of slightly longer wavelength than that emitted during fluorescence, due to the difference in energy between the  $T_1$  and  $S_1$ states. This emission is also much slower and is called phosphorescence. Whilst in the  $T_1$  state, a molecule may absorb some energy and be excited back to the  $S_1$  state. This molecule may then deexcite to a  $S_0$  state, just as in normal fluorescence. The emission spectrum is the same as in fluorescence, but the process is delayed ( $\sim 10^{-6} \text{ s}$ ) due to the time spent in the  $T_1$  state. Such emission is called *delayed fluorescence*.

**Organic Crystals.** There are many organic crystal scintillators, but only three are widely used: anthracence, trans-stilbene, and quaterphenyl. Anthracene has the highest scintillation efficiency of all organic scintillators, yet still only about half that of NaI(Tl). The fluorescence of both trans-stilbene and quaterphenyl has a decay time that depends on the incident particle. Fast fluorescence is produced by  $\gamma$  rays, slower fluorescence by neutrons, and even slower fluorescence by  $\alpha$ particles. Pulse shape discrimination can therefore be used to identify the incident radiation and perhaps reduce or eliminate any background. Since organic scintillators contain a large amount of hydrogen, they are efficient fast-neutron detectors when used in pulse shape discrimination mode. Organic crystal scintillators are, however, fragile and difficult to obtain in large sizes. In addition, the quantum yield is known to depend on the direction of the incident particle with respect to the crystal axis (20). The difference in response with direction can be as much as 20% to 30%, which severely degrades the energy resolution of such detectors.

Liquid Organic Scintillators. Liquid organic scintillators form a very versatile and useful category of detectors. The liquid state allows easy addition of other materials to improve and optimize the efficiency of such detectors. Addition of boron or gadolinium allows the detection of slow neutrons. To enhance the probability of photoelectric conversion of  $\gamma$  rays, lead or tin is added, which results in a very efficient and fast scintillator (21). Low-energy  $\gamma$  rays or  $\beta$  particles can be easily detected by dissolving or incorporating the sample in the liquid organic scintillator. This reduces losses due to absorption by the scintillator outer covering or the source itself, resulting in very high detection efficiencies. In addition, further fluorescent compounds can be added to shift the scintillation light spectrum to longer wavelengths in order to match better the spectral response of most photocathodes.

The simplest type of liquid organic scintillator consists of a fluorescent material, called the primary solute, dissolved in the solvent that makes up the bulk of the scintillator. Common solvents include toluene, xylene, and 1,4-dioxan. These compounds fluoresce when exposed to ionizing radiation; however, the quantum yield is low, and self-absorption can be a problem for large detectors. The addition of a primary solute has several effects that increase the efficiency of the scintillator. Firstly, the primary solute is a much more efficient scintillator, so that energy is transferred from molecules in the solvent (most energy loss occurs in the solvent, since it constitutes the bulk of the material) to molecules of the solute. This can occur by one of the following means: radiative transfer (scintillation light emission by solvent and absorption by solute), nonradiative dipole-dipole exchange, or collision exchange via diffusion of excited solvent molecules. Secondly, scintillation emission from most solvents is usually at short wavelengths that are not easily detected by most common photocathodes. The solute, therefore, also acts as a wavelength shifter with an absorption spectrum well matched to the emission of the solvent and an emission spectrum well matched to the spectral response of most photocathodes. This wavelength shift also eliminates the problem of self-absorption by the solvent. In some cases it is necessary or desirable to add a secondary solute to act purely as a wavelength shifter, absorbing the scintillation light of the primary solute and emitting at slightly longer wavelengths. This achieves an optimum match with the spectral response of the photocathode and reduces self-absorption by the primary solute if the dimensions of the detector are large. Popular primary solutes include PPO (2,5-diphenyloxazole), PBD (2-phenyl-5-(4-biphenylyl)-1,3,4-oxadiazole), and PTP (para-terphenyl), a common secondary solute is POPOP [1,4-di-(2-5-phenyloxazolyl)benzene].

The quantum yield of liquid organic scintillators is affected by the presence of impurities. Such impurities, especially dissolved oxygen, can serve as quenching agents significantly re-

ducing the quantum yield of the scintillator. Dissolved oxygen is generally removed by bubbling nitrogen through the liquid and kept out by sealing the liquid in an airtight container.

A popular use of liquid scintillators is as anticoincidence shields to reduce unwanted background events. Such shields are effective at detecting high-energy cosmic rays or high-energy  $\gamma$  rays that may mimic a real event. When an event is detected by the anticoincidence shields, any signal in the main detector is vetoed. Liquid scintillators can be of any shape or size and hence provide complete coverage at a very low cost.

Liquid organic scintillators are also routinely used to measure the activity of biological or chemical samples containing <sup>14</sup>C or <sup>3</sup>H. The low-energy electrons from such sources are most easily measured by dissolving the sample in the liquid scintillator. This may introduce problems such as low solubility of the sample, quenching by the sample, and reactions with the solvent or solute. However, stable solutions for most cases can be obtained by the appropriate choice of solvent and solute (22). Once the solution of liquid scintillator and sample has been prepared, it is placed in a light-tight enclosure and counted by a photomultiplier tube.

**Plastic Scintillators.** Organic plastic scintillators can be thought of as solid solutions, and hence they are very similar to organic liquid scintillators. They are prepared by using a solvent that can be polymerized (such as polystyrene or polyvinyltoluene), a primary solute, and perhaps a secondary solute. The solutes that make efficient scintillators for liquid solutions are also efficient scintillators in solid (plastic) form, so that PPO, PBD, and PTP are used. POPOP is a common secondary solute used as a wavelength shifter. For plastics the quantum yield of the scintillator can also depend on the method of preparation as well as its constituents (23). In addition, transfer of energy from the solvent to the solute is not as efficient as for solutions (solvent molecules cannot diffuse), and therefore greater concentrations of solute must be used to obtain comparable quantum yields.

Due to their mechanical strength, plastic scintillators are easy to handle and can be cut to any shape and size. They are relatively cheap and therefore practical in large-volume coincidence or anticoincidence counters. They are used extensively in nuclear physics, for lifetime and time-of-flight measurements, because of their fast response. Although, like the other organic scintillators, they can be used to detect both fast (scattering from protons) and slow (loading with boron or gadolinium) neutrons they do not have such good pulse shape discrimination characteristics.

Plastic detectors can be produced as very thin film scintillators with thicknesses down to 10  $\mu$ m (24). This allows the detection of low-energy highly ionizing particles such as heavy ions without complete loss of energy. Such detectors are known as *transmission detectors*.

## Scintillation Chambers or Lucas Cells

A *Lucas cell*, named after its inventor (25), is a scintillation chamber used for the measurement of  $\alpha$  radiation in gases. The cell is a cylinder of volume 0.1 L to 2 L, with a ZnS(Ag) coating on the internal surfaces. One end of the cylinder is a



Model 110A

**Figure 5.** Lucas cell. This type of counter is used for measuring  $\alpha$  radiation in gases.

quartz window, while at the opposite end two connectors are mounted for the exchange of gas samples (see Fig. 5).

Because of their high counting efficiency (about 75% for the measurement of 6 MeV  $\alpha$  particles in a standard 150 mL cell), Lucas cells allow the measurement of gas samples with small volumes and activities as low as 1 Bq m<sup>-3</sup>. The ZnS(Ag) scintillator material has its maximum emission at a wavelength of  $\lambda = 450$  nm, with a bandwidth of about 55 nm. Because of its high density and atomic number ( $\rho = 4.1$  g cm<sup>-3</sup>, Z = 23), it has a high conversion effciency of 150 eV/keV  $\leq \eta_{\rm s} \leq 200$  eV/keV. In order to ensure a maximum yield of luminous photons, the coating thickness has to be larger than the range of the  $\alpha$  particle, which, for ZnS, is about 15  $\mu$ m at energies around 6 MeV. The only disadvantage of Lucas cells is that ZnS(Ag), as a phosphorescent material, has a long relaxation time  $\tau_{\rm r} \approx 20 \ \mu$ s.

The efficiency of Lucas cells depends on their geometry and size. This is because the distance which the  $\alpha$  particle has to travel to reach a cylinder wall depends on the location of the  $\alpha$  decay within the cell and the direction of emission of the  $\alpha$  particle. The efficiency of the cell is equal to the fraction of decay events where the distance between the decay location and the cylinder wall for a given direction is less than the range of the  $\alpha$  particle in the gas (26). This circumstance also implies that, even for monoenergetic  $\alpha$  particles, the amount of energy deposited in the scintillator ranges from zero (the particle does not reach the cylinder wall) to  $E_p$  (the particle is emitted at a location directly adjacent to a wall). Therefore, Lucas cells are unsuitable for energy measurements.

Lucas cells are extensively used to detect radon, a naturally occuring radioactive noble gas. They can monitor the radon concentration in air either as a passive device with radon diffusing into the sensitive volume (27), or actively with air being continuously forced through the cell (28) or periodically exchanged (29). Radon is produced in the decay of radium in soils, rocks, and building materials. From there it enters residential buildings or workplaces such as mines, and, together with its progeny, forms a significant source of the human radioactive dose. In addition to surveys of the radon concentrations in the human environment, Lucas cells have also been employed to investigate the migration of radon in the earth's crust. Radon is released into the pore space of the rock preferentially under mechanical stress, and is then transported to the surface along fractures and faults. Thus, field studies have demonstrated a correlation of the radon exhalation from the ground with periodic stresses caused by tidal forces as well as with transient stresses before and during seismic events. Due to the latter fact, a sudden increase in the radon exhalation can be utilized as an earthquake precursor. In contrast, a constant regional anomaly of the radon exhalation can indicate the presence of an underground uranium ore body.

#### Photomultipliers

The photomultiplier is an essential constituent of every modern scintillation counter, because it enables the scintillation light pulse to be converted into an electrical signal, which can be electronically processed for further purposes.

A photomultiplier, as shown in Fig. 6, is a vacuum tube with a window for the entry of light at the top and a socket with electrical connectors at the base. The inside of the window is coated with an either opaque or semitransparent photoemissive layer, the photocathode. Photons incident on the photocathode release up to five photoelectrons, which are collected by an electron-optical system and focused onto the first of several electrodes. In the case of a photomultiplier, these electrodes are called *dynodes*. Since a high voltage ( $\approx 100$  V to 150 V typically) exists between each pair of subsequent dynodes, the electrons are accelerated from one dynode to the next. Upon impact on each dynode, an electron causes the emission of further secondary electrons, thus amplifying the pulse from one stage to another. The secondary electron yield depends on the impact energy of the primary electrons and therefore on the accelerating high voltage. Electrons emitted from the last dynode are collected on the anode, which forms the output stage of the photomultiplier. The transit time of a pulse from the photocathode to the anode is in the order of 50 ns, but the pulse is spread by less than 1 ns during its passage. During operation the photomultiplier has to be shielded



**Figure 6.** Internal structure of a photomultiplier. (Courtesy of Photonis, France)

from ambient light in order to avoid its destruction due to an excessive anode current. Furthermore, the electrostatic focusing of the dynodes and hence the pulse shape can be adversely affected by external magnetic fields. Therefore, the light shield can also serve as a magnetic shield when made of Mumetal (9).

Photomultiplier tubes vary mainly with regard to the number of dynodes and the photocathode material. Tubes with ten or more dynode stages are common. The wavelength  $\lambda_m$  of maximum photocathode sensitivity should be within the spectral range of the scintillation light. It depends on the photocathode material and ranges from 200 nm to about 450 nm. At wavelengths above 600 nm, the photon energy is below 2 eV and is hence lower than the energy  $E_a$  that is needed to release a photoelectron from the surface of a typical photoemissive material (2 eV  $\leq E_a \leq 4$  eV). The sensitivity at short wavelengths is limited by the absorption of photons in the window glass. The cutoff wavelength is about 350 nm for normal glass, but can be extended to 160 nm by using a quartz window (14). The maximum sensitivity  $s_{\lambda}$  is typically 80 mA/ W. With h being Planck's constant, c the speed of light, and ethe electron charge, this corresponds to photocathode quantum efficiency

$$\eta_{\rm c} = s_{\lambda} \frac{hc}{\lambda_{\rm m} e} \tag{4}$$

of about 25%.

Two further important characteristics of photomultipliers are the gain and the dark current. The gain is the ratio of the amplified anode current to the initial photoelectron current. It depends on the number and design of the dynodes and also on the high voltage applied. In order to ensure a constant gain, the high voltage should be well stabilized. The gain is typically very high (between  $10^5$  and  $10^8$ ), so that single photons incident on the photocathode can be registered. The dark current is mainly the anode current at zero photocathode illumination. It consists of very short pulses ( $\approx 1$  ns, the transit time spread), which are due to the thermally activated emission of electrons from the photocathode. The dark current is therefore dependent on the operating temperature. Typical values range from 1 nA to 100 nA, depending on the high voltage.

Since the gain is determined by the high voltage, particular attention has to be paid in designing a suitable high-voltage divider circuit. The voltage distribution along the divider can vary widely, depending on the desired pulse shape. Figure 7 shows a simple divider circuit supplying a high voltage to the dynodes  $d_1$  to  $d_{10}$  as well as to the cathode c and anode a of a photomultiplier tube.

The anode has to be positive with regard to the photocathode in order to accelerate the photoelectrons. This can be achieved by either (1) connecting the high voltage to the anode and grounding the photocathode, or (2) vice versa. The second polarity option has the disadvantage of a high voltage across the glass housing, which can induce spurious pulses due to electroluminescence in the glass (14). However, the anode can be connected directly to the input of the pulse amplifier. This is not the case in the first option (as shown in Fig. 7), where a high-voltage capacitor  $C_{\rm HV}$  has to be inserted in order to protect the pulse amplifier from the high dc voltage while still allowing the signal pulse to pass.



**Figure 7.** A simple high voltage divider circuit used with a photomultiplier. Photoelectrons generated at the photocathode c are accelerated toward the anode by a high voltage  $U_{\rm H}$ .

The gain and therefore the signal current  $I_a$  at the anode of the photomultiplier is adjusted via the high voltage  $U_{\rm H}$ . If A is the maximum count rate and Q is the charge per pulse [see Eq. (3)], then

$$I_a = QA \tag{5}$$

 $I_a$  should be much higher than the dark current.

The electrons transiting between the dynodes cause a mean current across the divider resistors  $R_d$  in the opposite direction. Hence, the divider voltage is reduced during a pulse passage, giving rise to variations in the photomultiplier gain. To minimize the effect of a variable mean anode current on the gain, these resistors are chosen so that the divider current  $I_d \geq 100 I_a$ .

Furthermore, a pulse current across the last dynodes with a decay time equal to the relaxation time  $\tau_r$  of the scintillation pulse causes a sudden reduction of the voltage  $U_d$  across the corresponding divider resistor  $R_d$  of about  $\Delta U_d = QR_d/\tau_r$ , leading to gain fluctuations. These fluctuations can be attenuated by using *reservoir capacitors* ( $C_r$  in Fig. 7). During a pulse these capacitors instantaneously supply enough charge to keep the interdynodal voltage almost constant. They are subsequently recharged with a current through the corresponding divider resistor  $R_d$  with a time constant  $\tau_d = R_dC_r$ . If  $\tau_d \gg$  $\tau_r$ , the pulse current across  $R_d$  is spread out in time and has a much lower peak amplitude of about  $\Delta U_d = Q/C_r$  (9).

Similar considerations apply for the shape of the signal pulse at the photomultiplier output. This stage consists of an effective anode resistance  $R_a$  and a capacitance  $C_a$ , forming an RC circuit with a time constant  $\tau_a = R_a C_a$ . Here  $C_a$  is usually not a physical capacitor, but the stray capacitance of the anode to ground (and hence drawn in dashed lines in Fig. 7). Note that  $R_a$  and  $C_a$  include resistances and capacitances at both the photomultiplier output and the input of the following pulse amplifier (22). Two cases can be distinguished. If (1)  $\tau_a \ll \tau_r$ , the pulse charge is quickly passed to ground through  $R_a$  and the duration  $au_o$  and height  $\Delta U_o$  of the output pulse are determined by the relaxation time  $\tau_{\rm r}$ , with  $\tau_{\rm o} = \tau_{\rm r}$  and  $\Delta U_{\rm o} =$  $QR_a/\tau_r$ . This mode of operation is called the *current mode*. It ensures a minimum response time and is therefore used in applications requiring fast timing. In this mode, the thermally activated pulses of the dark current can produce quite large output pulses because of their short duration. This background can be effectively eliminated in coincidence measurements, where two photomultipliers are coupled to the same scintillator, and a pulse is counted only if it occurs in

both photomultipliers within a very small time window. If (2)  $\tau_a \gg \tau_r$ , the whole pulse charge is integrated on  $C_a$  before it can flow to ground through  $R_a$ . Then  $\tau_o = \tau_a$ , and the pulse height  $\Delta U_o = Q/C_a$  is proportional to the charge Q and independent of  $\tau_r$ . Hence, if the sensitivity of the counter is a constant, the energy  $E_p$  of the incident particle can be calculated from  $\Delta U_o$  [see Eq. (3)]. This mode is called the *voltage mode* and is suitable for the measurement of particle energy spectra (30).

Further details about pulse amplifier and high-voltage divider circuits can be found in Ref. 31.

### **Optical Contact**

When transiting from the scintillator to the photocathode of the photomultiplier, the luminous photons can be lost from the light pulse due to reflection from the window surfaces (see Fig. 1). These optical losses can be avoided by coupling the scintillator and the photomultiplier windows with a transparent optical grease or oil, which should have a refractive index that is in between that of the scintillator (1.45 to 1.85) and that of the glass of the photomultiplier window ( $\approx$  1.5).

Some applications require the photomultiplier to be mounted at some distance from the scintillator, for example, because of geometrical constraints in the experiment or because of the presence of strong magnetic fields near the scintillator. In these cases, the scintillator and the photomultiplier are connected with a light guide made of Perspex or Plexiglas or a similar transparent material. Light guides are also used in order to couple scintillators and photocathodes of different sizes and shapes.

## PRESENT STATE OF TECHNOLOGY AND OUTLOOK

The widespread use of scintillators has ensured that advances and improvements in scintillator technology continue. Research into new scintillating material with higher quantum yield, better-matched spectral response, improved radiation hardness (deterioration of response with exposure to radiation), faster response, or a combination of these properties is actively pursued. In addition, new methods for using scintillating materials are being developed.

The new generation of nuclear and particle physics experiments require beams of even higher energy and higher fluxes. This means the new detectors must be radiation-hard, very fast, and dense (to provide compact detectors). A very likely candidate for such a scintillator material is barium fluoride  $BaF_2$  (32). It has a fast (subnanosecond, 0.6 ns to 0.79 ns) decay component and a slower (100 ns to 1000 ns) component. These components have different emission spectra (195 nm and 220 nm for the fast component and 310 nm for the slow component) and a large difference in decay constants, so that it is very simple to select the fast component only. A disadvantage is that the fast emission is in the UV, requiring special photomultiplier windows and photocathodes that are sensitive at these lower wavelengths but insensitive to higher (> 300 nm) wavelengths. Attempts to decrease the relative contribution of the slow component include doping the  $BaF_2$ with lanthanum. This has proved to be successful.

Other possible candidates for fast, dense, radiation-hard scintillators include cerium fluoride (CeF<sub>3</sub>) (33) and gadolinium orthosilicate doped with cerium [GSO(Ce)] (34). They both show excellent radiation hardness, good quantum yield, and a good spectral match with most existing photocathodes. Their main disadvantage is their relatively long decay times: 20 ns to 35 ns for CeF<sub>3</sub>, and even longer for GSO(Ce), depending on the doping concentration of Ce. Along with lutetium orthosilicate doped with cerium [LSO(Ce)], these new scintillating materials have already been used in positron emission tomography (PET).

Advances have also been made with organic scintillators. Scintillators that are radiation-hard and have a higher quantum yield and better-matched spectral response have been developed. Radiation hardness depends on how the optical transmission properties of the base are affected by radiation and also on damage to fluor molecules. Radiation-tolerant bases have been developed by some manufacturers (35), and the effect of radiation damage to the fluor molecules can be minimized by increasing their concentration. New polystyrene bases have also been produced and tested regarding radiation hardness, quantum yield, and stability of response with time (36). To produce scintillators with better spectral response and longer attenuation length, a new class of fluors with a large Stokes shift (difference in the absorption and emission spectra) have been developed, such as 1-phynyl-3mesityl-2-pyrazoline (PMP) and 3-hydroxyflavone (3HF). They enable the construction of larger detectors and longer scintillating fibers.

A recent application of scintillators is in scintillating fiber *trackers* (37). Such detectors consist of a large number ( $10^5$  to 10<sup>6</sup>) of scintillating fibers that can be either glass, plastic, or capillaries filled with liquid scintillator. The fibers can be square, circular, or hexagonal in cross section and are typically 20  $\mu$ m to 50  $\mu$ m across and up to several meters in length. The scintillating fibers provide fast and efficient charged-particle tracking with position resolution down to 20  $\mu$ m combined with the flexibility of instrumenting large and unusually shaped volumes. Such trackers have been used in many particle and nuclear physics experiments. Glass fibers doped with cerium oxide were the first to be implemented and produced fast and efficient trackers. However, the short attenuation length of such fibers limits their use in larger detectors. Plastic scintillators and capillaries with liquid scintillator show more promise for such applications, although a disadvantage of plastic scintillators is crosstalk between nearby fibers. The light output of such huge arrays of fibers can be detected using several different types of photosensors. If the event rate is low, image intensifiers and charge-coupled devices (CCDs) can be used. Otherwise, multianode photomultipliers, avalanche photodiodes, or visible-light photon counters (VLPCs) must be used. The last are the preferred choice, due to their spectral response, high quantum yield (70% to 80%), high gain, gain stability, and low noise level. Although for the present scintillating fiber trackers are used only in research detectors, it is only a matter of time before their excellent resolution, speed, and efficiency are made available in medical imaging (PET detectors) and other areas.

An alternative to scintillating fibers is *scintillating pads* (38). Such pads are rectangular pieces of organic plastic scintillator of area 1 cm  $\times$  2 cm and about 3 mm thick. They are coupled to photodetectors by wavelength-shifting fibers. They do not have the position resolution of scintillating fibers, but can be effectively used for triggering and tracking over large volumes.

A further method of using scintillators is in *scintillation* proximity assay (PSA). This is a technique used to trace biological molecules and analyze biomolecular interactions. Small beads of scintillator about 5  $\mu$ m across are chemically coated to allow molecules such as antibodies or proteins to become attached to the bead. These molecules are labeled with a radioactive tracer such as tritium. The beads and molecules are placed in aqueous solution. When the molecule becomes attached to the scintillating bead, the radiation from the tracer causes the scintillator to emit light that can be detected by a photomultiplier or other light-sensitive device. The tracer is chosen to emit low-energy radiation, so that a bead will only emit scintillation light if the tracer molecule is very close. The scintillating beads are usually made from cerium-doped glass or some organic plastic scintillator.

Current research and development of new scintillating materials and scintillation techniques will ensure that scintillators will continue to be an important part of radiation and particle detection. New devices based on semiconductor technology have not replaced but rather enhanced the use of scintillators by providing alternative and more sensitive photosensors.

# **BIBLIOGRAPHY**

- 1. J. B. Birks, *The Theory and Practice of Scintillation Counting*, Oxford: Pergamon Press, 1964.
- 2. H. Geiger and E. Marsden, On a diffuse reflection of the  $\alpha$ -particles, Proc. Roy. Soc., A82: 495–500, 1909.
- 3. E. Rutherford, The scattering of  $\alpha$  and  $\beta$  particles by matter and the structure of the atom, *Phil. Mag. VI*, **21**: 669–688, 1911.
- A. Krebs, Ein Demonstrationsversuch zur Emanationsdiffusion, Ann. Physik, 39: 330–332, 1941.
- S. C. Curran and W. R. Baker, Photoelectric alpha-particle-detector, *Rev. Sci. Instrum.*, 19: 116, 1948.
- 6. H. Kallmann, Natur und Technik, July 1947.
- P. R. Bell, The use of anthracene as a scintillation counter, *Phys. Rev.*, **73**: 1405–1406, 1948.
- 8. R. Hofstadter, Alkali halide scintillation counters, *Phys. Rev.*, **74**: 100–101, 1948.
- Yu. K. Akimov, Scintillation Counters in High Energy Physics, New York: Academic Press, 1965.
- H. Frauenfelder and E. M. Henley, Subatomic Physics, 2nd ed., Englewood Cliffs, NJ: Prentice-Hall, 1991.
- 11. S. E. Hunt, Nuclear Physics for Engineers and Scientists, New York: Wiley, 1987.

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- 12. E. Fenyves and O. Haiman, *The Physical Principles of Nuclear Radiation Measurements*, New York: Academic Press, 1969.
- W. J. Price, Nuclear Radiation Detection, New York: McGraw-Hill, 1964.
- J. Chariton and C. A. Lea, Some experiments concerning the counting of scintillation produced by alpha particles, *Proc. Roy.* Soc., A122: 304-352, 1929.
- G. F. Knoll, Radiation Detection and Measurement, 2nd ed., New York: Wiley, 1979.
- 16. W. H. Tait, Radiation Detection, London: Butterworths, 1980.
- C. J. Crannell, R. J. Kurz, and W. Viehmann, Nucl. Instrum. and Meth., A115: 253–261, 1974.
- 18. M. Mutterer, Nucl. Instrum. Meth., A196: 73-81, 1982.
- 19. A. J. P. L. Policarpo, Space Sci. Instrum., 3: 77, 1977.
- D. B. Oliver and G. F. Knoll, *IEEE Trans. Nucl. Sci.*, NS-15: 122–126, 1968.
- Z. H. Cho, I. Ahn, and C. M. Tsai, *IEEE Trans. Nucl. Sci.*, NS-21: 218–226, 1974.
- 22. B. W. Fox, Techniques of Sample Preparation for Liquid Scintillation Counting, Amsterdam: North-Holland, 1976.
- 23. M. G. Schorr and F. L. Torney, Phys. Rev., 80: 474, 1950.
- 24. G. Bendiscioli et al., Nucl. Instrum. Meth., A206: 471-476, 1983.
- M. F. Lucas, Improved low-level alpha scintillation counter for radon, *Rev. Sci. Instrum.*, 28: 680–684, 1957.
- M. Lenzen and H. J. Neugebauer, A theoretical investigation in the Lucas cell, Nucl. Instrum. Meth. Phys. Res. A, 368: 479–483, 1996.
- 27. H. Woith et al., On the feasibility of monitoring radon in soil gas and groundwater as a precursor to earthquakes, in J. Zschau and O. Ergünay (eds.), *Turkish–German Earthquake Research Project*, Kiel, Germany: Kiel University, 1989, pp. 112–129.
- J. W. Thomas and R. J. Countess, Continuous radon monitor, Health Phys., 36: 734–738, 1979.
- M. Lenzen and H. J. Neugebauer, An automatic radon sensor for borehole measurements, *Rev. Sci. Instrum.*, 68 (7): 2898–2903, 1997.
- W. E. Burcham and M. Jobes, Nuclear and Particle Physics, Harlow, Essex: Longman Scientific & Technical, 1994.
- 31. E. Kowalski, Nuclear Electronics, Berlin: Springer-Verlag, 1970.
- F. Sauli (ed.), Instrumentation in High Energy Physics, Singapore: World Scientific, 1992.
- 33. D. F. Anderson, Nucl. Instrum. Meth., A287: 606-612, 1990.
- M. Kobayashi et al., A beam test on a fast EM-calorimeter of gadolinium silicate, GSO(CE), Nucl. Instrum. Meth., A306: 139– 144, 1991.
- C. Zorn, Progress in the design of a radiation-hard plastic scintillator, *IEEE Trans. Nucl. Sci.*, NS-38: 194–199, 1991.
- T. Hasegawa et al., Tests of new polystyrene-based scintillators, Nucl. Instrum. Meth., A311: 498-504, 1992.
- R. C. Ruchti, The use of scintillating fibers for charged-particle tracking, Annu. Rev. Nucl. Part. Sci., 46: 281–319, 1996.
- D. Adams et al., Scintillating pad detectors, *IEEE Trans. Nucl. Sci.*, NS-44 (3): 455–459, 1997.

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SCINTILLATION COUNTERS. See Photomultipliers. SCOPES. See Oscilloscopes. SCR. See Thyristor types. SCREENING. See Burn-in and screening.