# UNIT-IV

## Nuclear Detectors

## Scintillation detector

## 5.3 Scintillation Detectors

Weakly interacting radiation gives poor detection efficiency in gas detectors, largely because of the low density of material in the sensitive volume. A condensed medium, such as a scintillator, offers an improvement of at least three orders of magnitude in this respect. The term luminescence is used as a general description of this process.

A scintillation detector consists of :

- the scintillator material (organic or inorganic). This produces optical photons whenever an ionization event has occurred (sections 5.3.1 and 5.3.2). The scintillator is enclosed in a light-tight protective can and coupled to either a light guide or the entrance window of a photomultiplier. This coupling is made with a thin layer of fluid (often silicone oil or grease) which transmits the photons with minimum absorption.
- the reflector. The emission of scintillation light is isotropic. Since $4\pi$ collection is not generally available, an efficient detector relies on the recovery of photons which would otherwise miss the photomultiplier.
- a light guide. This is sometimes required when the scintillator itself is not immediately coupled to the photomultiplier tube. Situations in which a light guide is used include: (1) when measurements need to be made in a magnetic field: (2) when the size of the scintillator is larger than the size of the photomultiplier (although this causes a large loss of light).
- the photomultiplier tube (PMT). This consists of an evacuated housing (generally glass or quartz) into which the photons are admitted through a window backed by a photocathode material. The window material must have very low absorption, and the photocathode material a very high absorption, towards the incident scintillation light. Electron emission from the cathode is focused onto a series of dynodes held at an increasingly high positive potential with respect to the cathode.
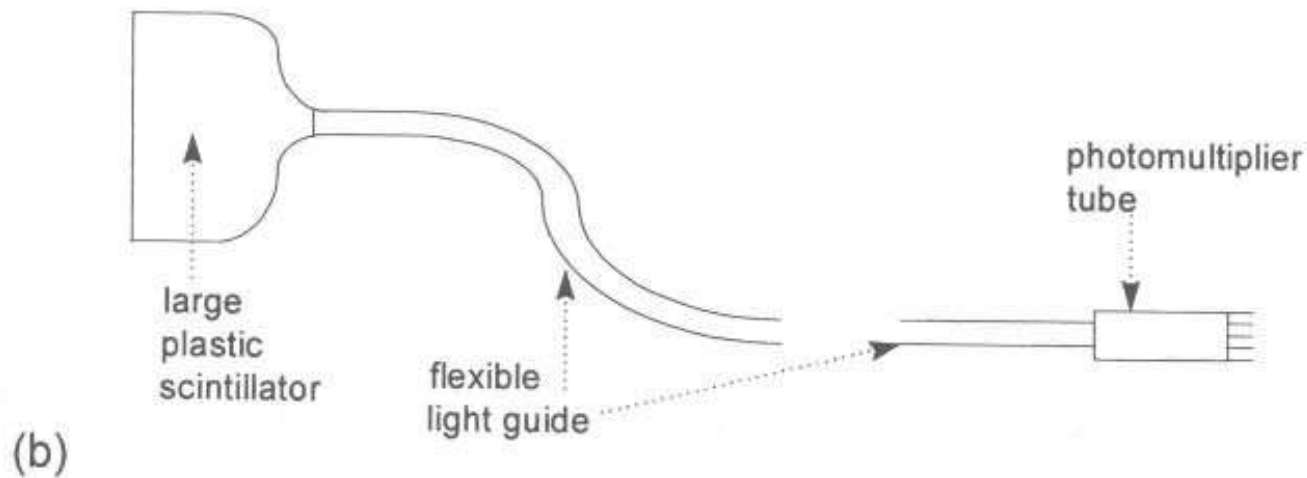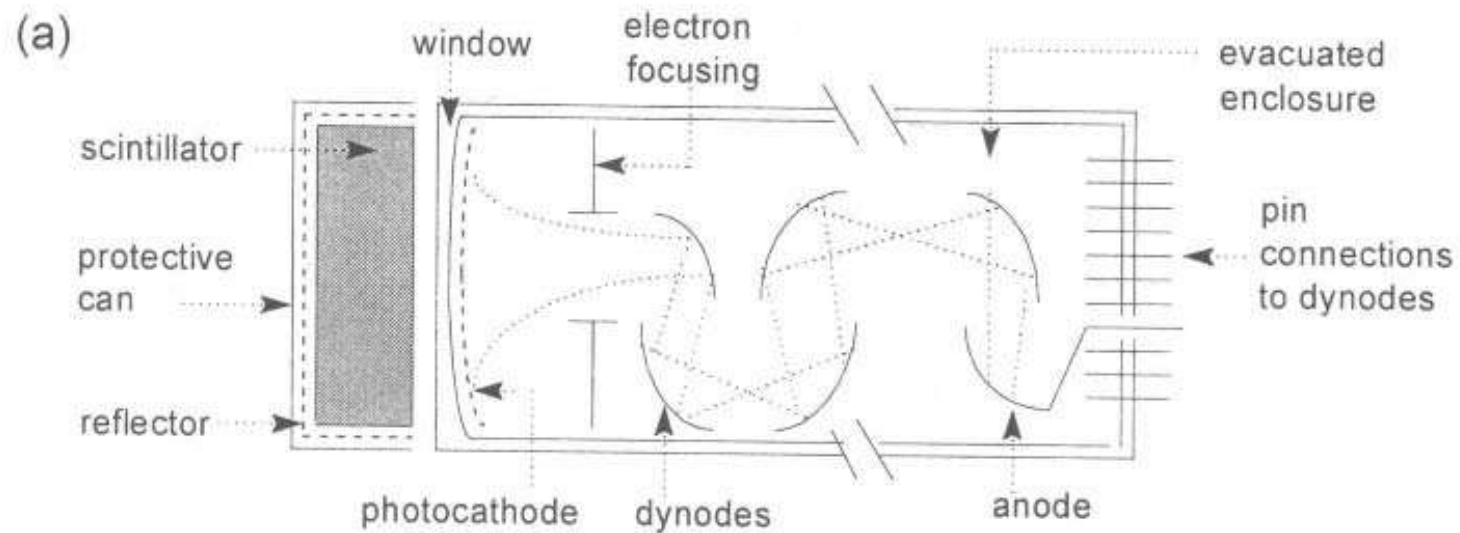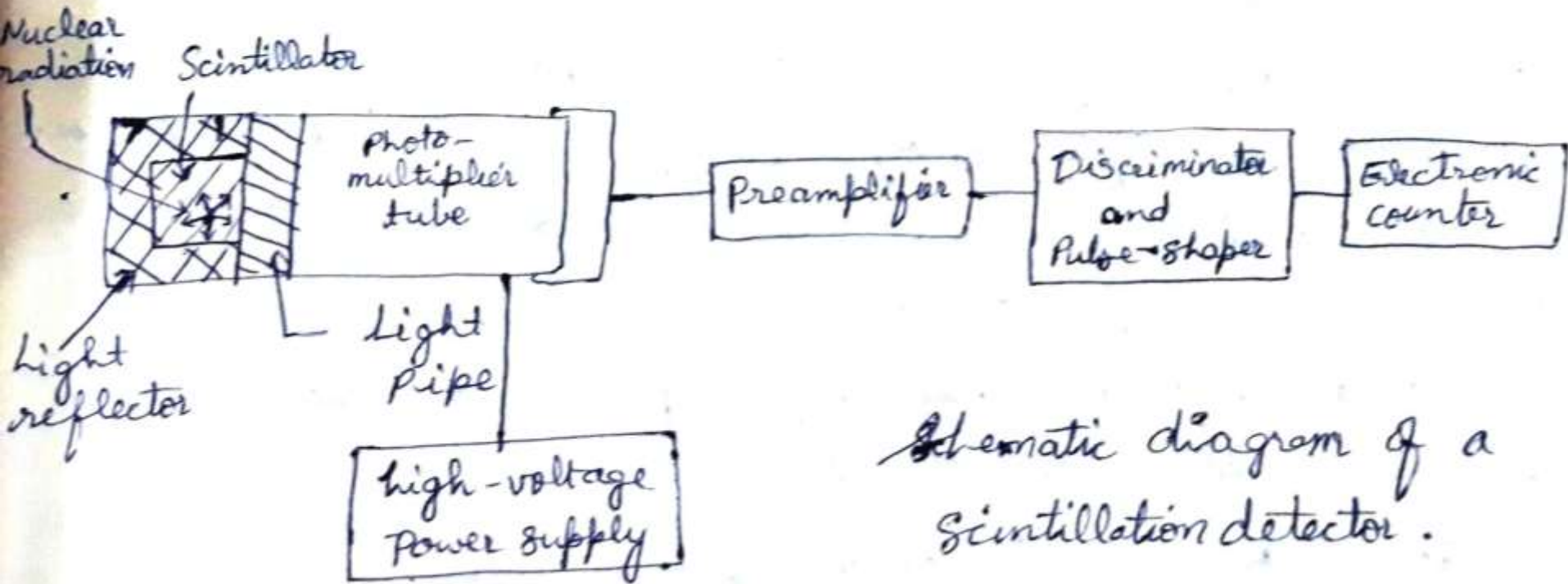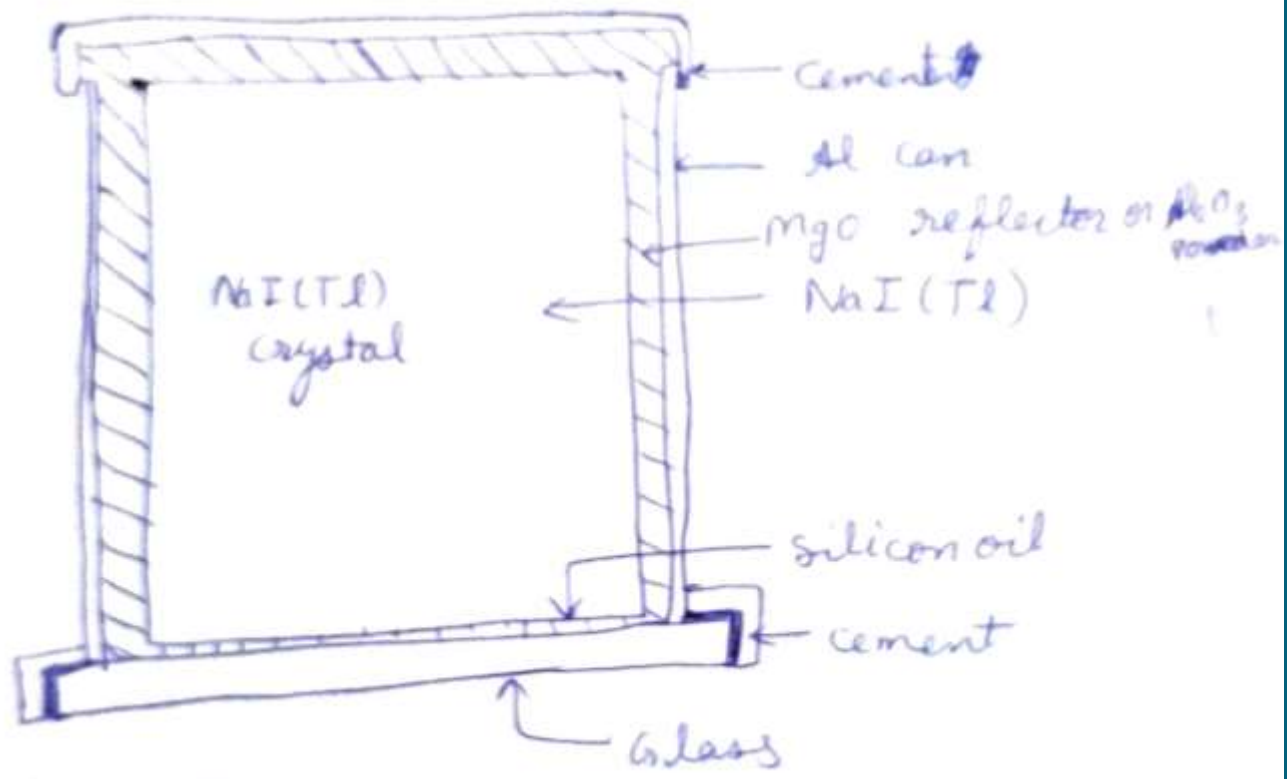
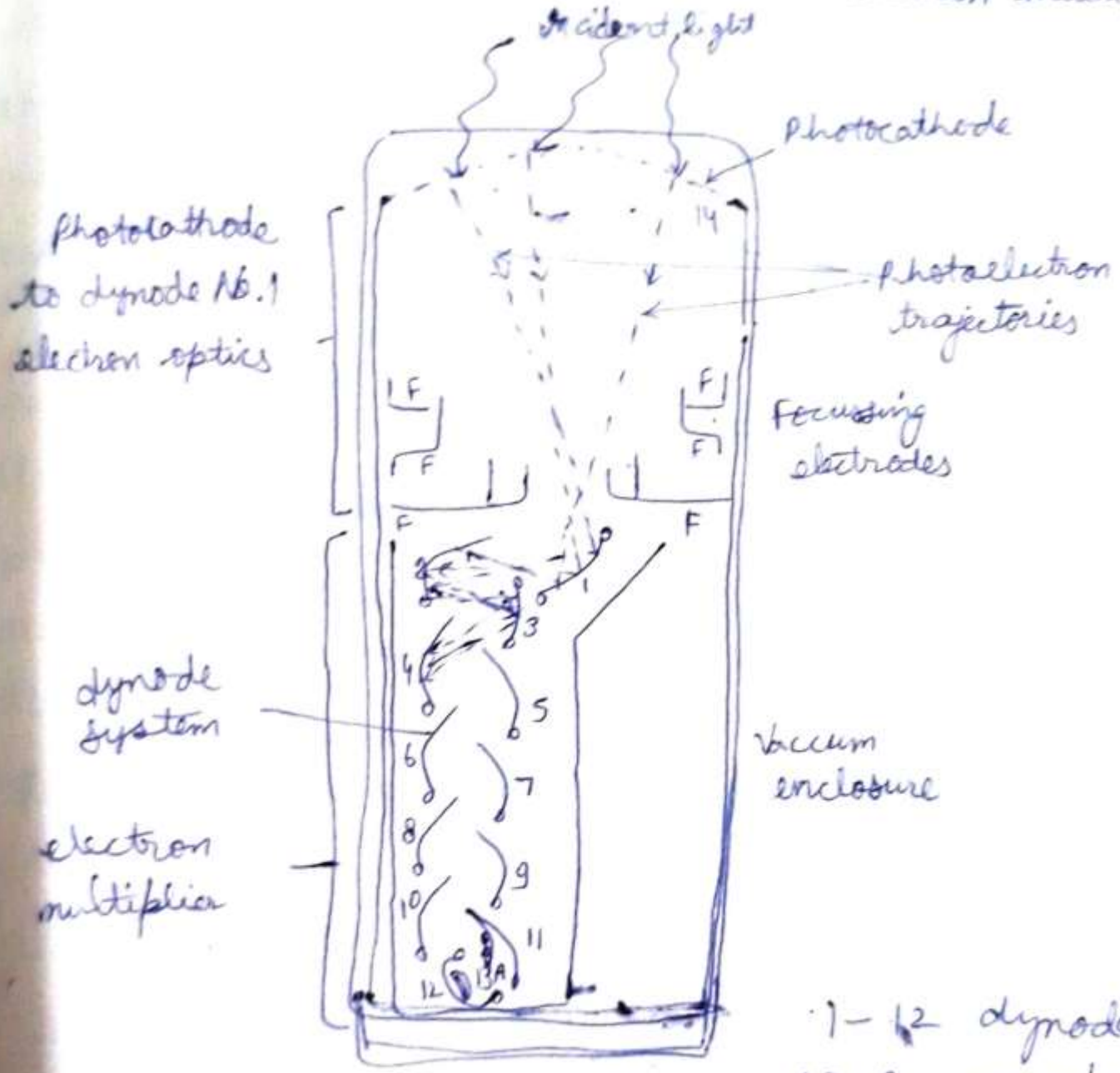Fig.(5.14) Schematic diagrams of scintillator systems. (a) direct coupling to a PMT (b) a light-pipe for use in high magnetic or radiation fields.

General System for the Scintillation detector:
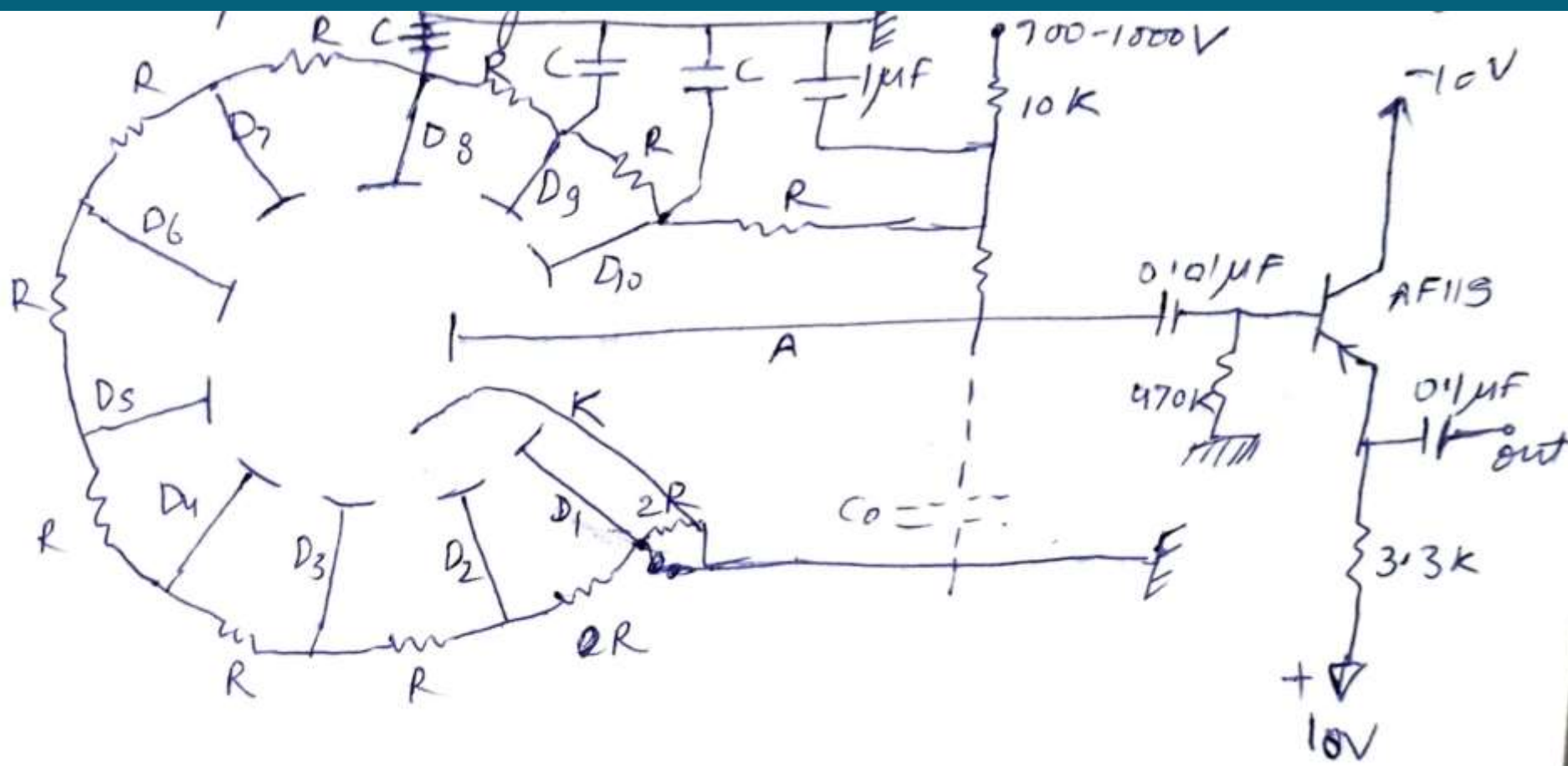
Nuclear radiation    Scintillator



Light reflector

Light Pipe

photo-multiplier tube

high-voltage Power supply

Preamplifier

Discriminator and Pulse-shaper

Electronic counter

Schematic diagram of a scintillation detector.

Let the energy of incident nuclear radiation be $E_n(eV)$, it loses part of its energy in scintillator. Let $f_n$ be the fraction of this energy dissipated in the scintillator.

This energy $(f_n E_n)$ given to the phosphor is used up in ionisation, excitation and also in dissociation of phosphor molecules.

If this energy, $f_n E_n$, is converted into light energy by the scintillator with efficiency $\epsilon_1$, then $\epsilon_1 f_n E_n$ is the amount of light energy produced in the scintillator.

Now, let $f_p$ is the fraction of photons reaching the photocathode as, some photons are absorbed, then $f_p \epsilon_1 f_n E_n$ amount of light energy reaches the photocathode.

Let $\alpha$ be the conversion efficiency of photocathode then, it will convert (emit) $\alpha f_p \epsilon_1 f_n E_n$ number of photoelectrons per ev energy of light which strikes it.

If $f_d$ is the fraction of photoelectrons collected

by the ~~first~~ dynode, and if the electron multiplication factor is M, then ~~if the~~ every dynode will produce M times new electrons and will be collected by next dynode. If the no. of dynodes are n, then, total number of electrons reaching the final anode will be

$$N_o = \alpha \, f_d \, f_p \, \varepsilon_1 \, f_n \, E_n \, M'^n$$

Charge collected at the ~~surfo~~ plates of capacitor will be $Q = Ne$

$$= e M^n \alpha \, f_d \, f_p \, \varepsilon_1 \, f_n E_n$$

$$V = \frac{Q}{C} \qquad or \quad V \alpha \bar{E}_n$$

Pulse height in a Scintillation detector is $\alpha$ to the initial energy of the incident particle.

## 5.3.1 Light production mechanism in inorganic scintillators

Inorganic scintillators are crystalline materials and they therefore possess long-range order. Deposition of energy results in the excitation of an electron from the valence to the conduction band. The first stage of the process is always the elevation of an electron from the valence band. This produces either an electron-hole pair if the deposited energy is greater than the band gap energy, $E_g$, or an exciton if it is less than $E_g$ (see also Section 6.10 and [6]). Thus if :

- $E > E_g$ : a mobile electron and its less mobile hole is produced. These can diffuse independently of each other throughout the crystal.
- $E < E_g$ : the coupled electron-hole system can transport excitation energy (the exciton), but not charge, through the crystal.

An inorganic scintillator relies on the presence of localized trapping centres in the crystal which produce energy states within the band gap. It is the population of these states, followed by their de-excitation by photon emission, that results in the production of light which is used for radiation detection. There are two essential requirements on the emitted photon energy :

- it must be different from the band-gap energy in order to prevent self-absorption within the crystal, and
- it must be in the visible or near ultra-violet part of the spectrum for convenient conversion and amplification by a photomultiplier or photodiode.

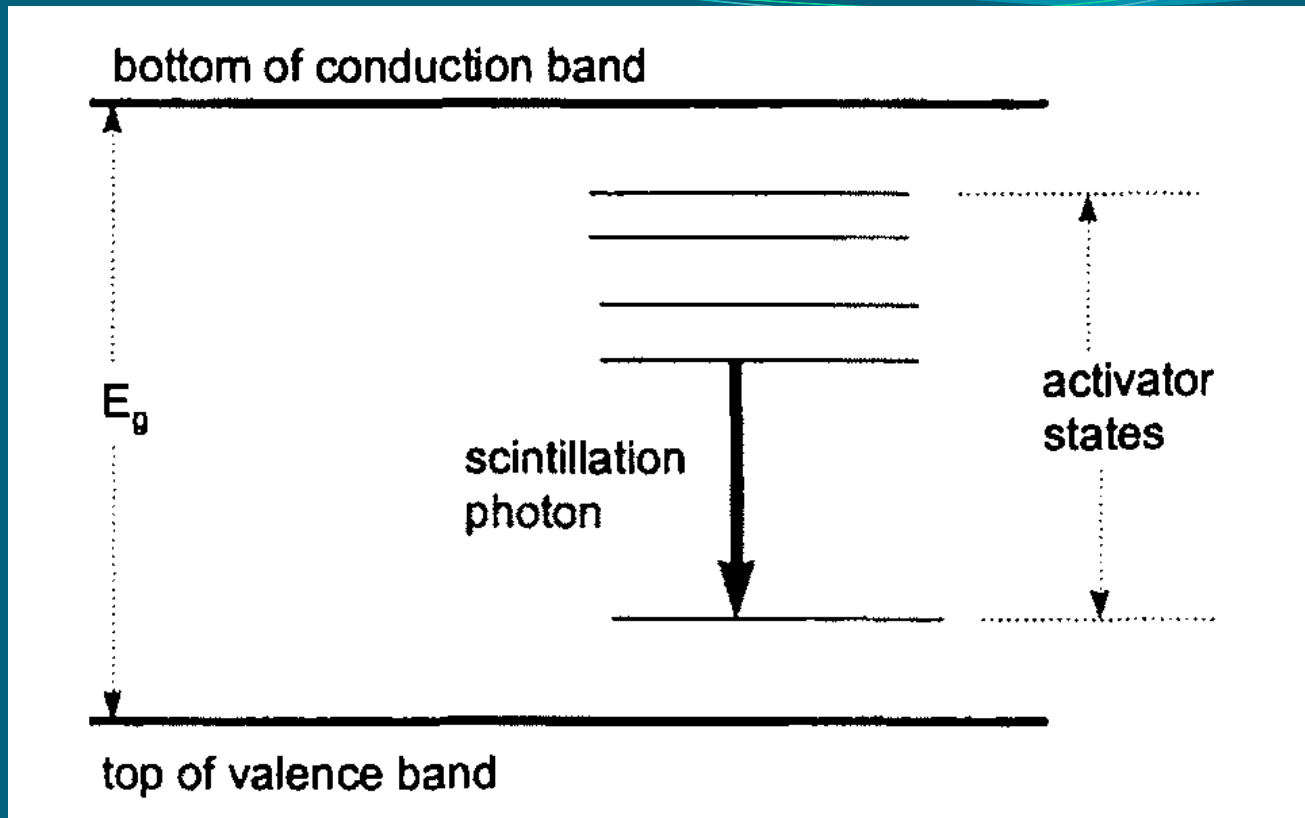Fig.(5.15) Schematic diagram of activator states within the band gap of an inorganic scintillator. Deactivation of any of the higher excited states to the first excited state will take place rapidly ($<10^{-10}$s). Deactivation of the first excited state to the ground state takes place with a lifetime $\sim 10^{-7}$s. It is this lifetime which determines the response (decay) time of the scintillator.

Trapping centres may be produced in a variety of ways. The mechanism of scintillation depends slightly on the type and concentration of these centres. However, there is still no full understanding of how impurity concentration or crystal temperature, for example, determines efficiency, luminescence decay time or pulse-shape.

Inorganic scintillation materials generally fall into two broad groups :

- crystals that are without additives. This category includes crystals which may contain strains or dislocations, or which may have trace quantities of impurity atoms. Caesium fluoride, CsF, and barium fluoride, $BaF_2$, are two such examples. They have the fastest decay times of the most widely used inorganic phosphors. There is still no evidence to confirm that a perfect crystal, without strain or trace impurities, will not scintillate.
- an otherwise perfect crystal in which there are added impurities (called activators), e.g. sodium iodide which has been doped with thallium, NaI(Tl). The activator is usually a large atom (Tl with Z=69) compared with the host material, Na (11) and I (53).  An exception is CsI(Na).

Table (5.1)   Characteristics of some widely-used inorganic scintillators, [6].   Note that: (1) BGO has the highest density,   (2) $BaF_2$ has the shortest decay time and therefore the best timing resolution, (3) CsI(Tl) has the highest photon yield,  (4) to a large extent, NaI(Tl) combines all of these advantages. The disadvantage with NaI(Tl) is that it is hygroscopic and it therefore needs canning (i.e. hermetic sealing) to prevent moisture absorption.

| | Na(Tl) | CsI(Tl) | $Bi_4Ge_3O_{12}$ (BGO) | $BaF_2$ |
|---|---|---|---|---|
| Density (g cm$^{-3}$) | 3.67 | 4.51 | 7.13 | 4.89 |
| Principal decay time ($\mu$s) | 0.23 | 1.0 | 0.35 | 0.62 (slow) 0.0006 (fast) |
| Maximum emission (nm) | 415 | 540 | 505 | 310 (slow) 220  (fast) |
| Yield (photons/MeV) | $3.8 \times 10^4$ | $5.2 \times 10^4$ | $8.2 \times 10^3$ | $10^4$ |

A qualitative description of the inorganic scintillation process can be made with reference to Fig.(5.15) as follows :

- When the deposited energy is greater than $E_g$, an electron can be transferred from the valence to the conduction band. The electron can diffuse through the conduction band with relatively high mobility, but the positively charged hole diffuses through the valence band with a much lower mobility. In the event that geminate recombination takes place, it is highly likely that the resulting photon will have an energy close to $E_g$ and be absorbed within the crystal.
- The presence of the activator (or other type of defect) modifies the band structure in that region of the crystal. This creates localized energy states, within the band gap which the electron can occupy. Thus, the initially-created hole state can diffuse through the crystal and become trapped at the activator site. This site will remain ionized, *i.e.* it has a positive charge, until it traps the electron which has been migrating through the crystal with high mobility.
- Upon trapping, the electron is likely to occupy first an excited activator state from where it rapidly de-excites to the ground state. As it does so, it emits a photon whose energy is smaller than the host lattice band gap $E_g$. This photon forms the scintillation light which emerges from the crystal. The time between the initial energy deposition and the emission of the photon, is determined by the migration time of the hole, and the lifetime of the activator states. Both of these are considerably longer than the migration time of the electron.

- The de-excitation of the activator states is governed by quantum mechanical selection rules. Depending on these rules, different host-activator systems give rise to prompt (fluorescence) photon emission or delayed (phosphorescence) emission. Certain combinations have selection rules which prohibit photon emission entirely. In these cases the energy can be dissipated through phonon emission when energy flows into the host lattice as low grade heat. This is referred to as a quenching process.

Some alkali halide systems are able to trap electrons in metastable inter-band states. This is the basis of the (integrating) thermoluminescence dosimeter (see section 7.13).

### 5.3.2 Light production mechanism in organic scintillators

The performance of an organic scintillator is virtually independent of its state of aggregation. It can be in a crystalline, liquid, polymer or amorphous state so long as it is transparent to the emitted photons. The only requirement is that the light emission spectrum of the material does not overlap with its absorption spectrum.

By comparison with its inorganic counterpart, an organic scintillator is distinguished by :

- a lower density,
- a lower atomic number, and hence a lower efficiency for photons interacting primarily by the photoelectric effect ($\propto Z^{3.5}$),
- a better time resolution,
- a worse energy resolution.

The scintillation process in organic materials involves transitions between electronic, vibrational and rotational states. A precise description requires accurate knowledge of the wavefunctions of the initial and final states involved. Although these are not easily determined for polyatomic molecules, the underlying principles of a scintillator can be illustrated using the picture of a diatomic system.

Apart from the crystalline materials – anthracene and stilbene are the most common – there is no long range order in organic scintillator materials. Deposition of energy results in the excitation of a molecular system, represented in Fig. (5.16), from the ground to one of several excited electronic states.
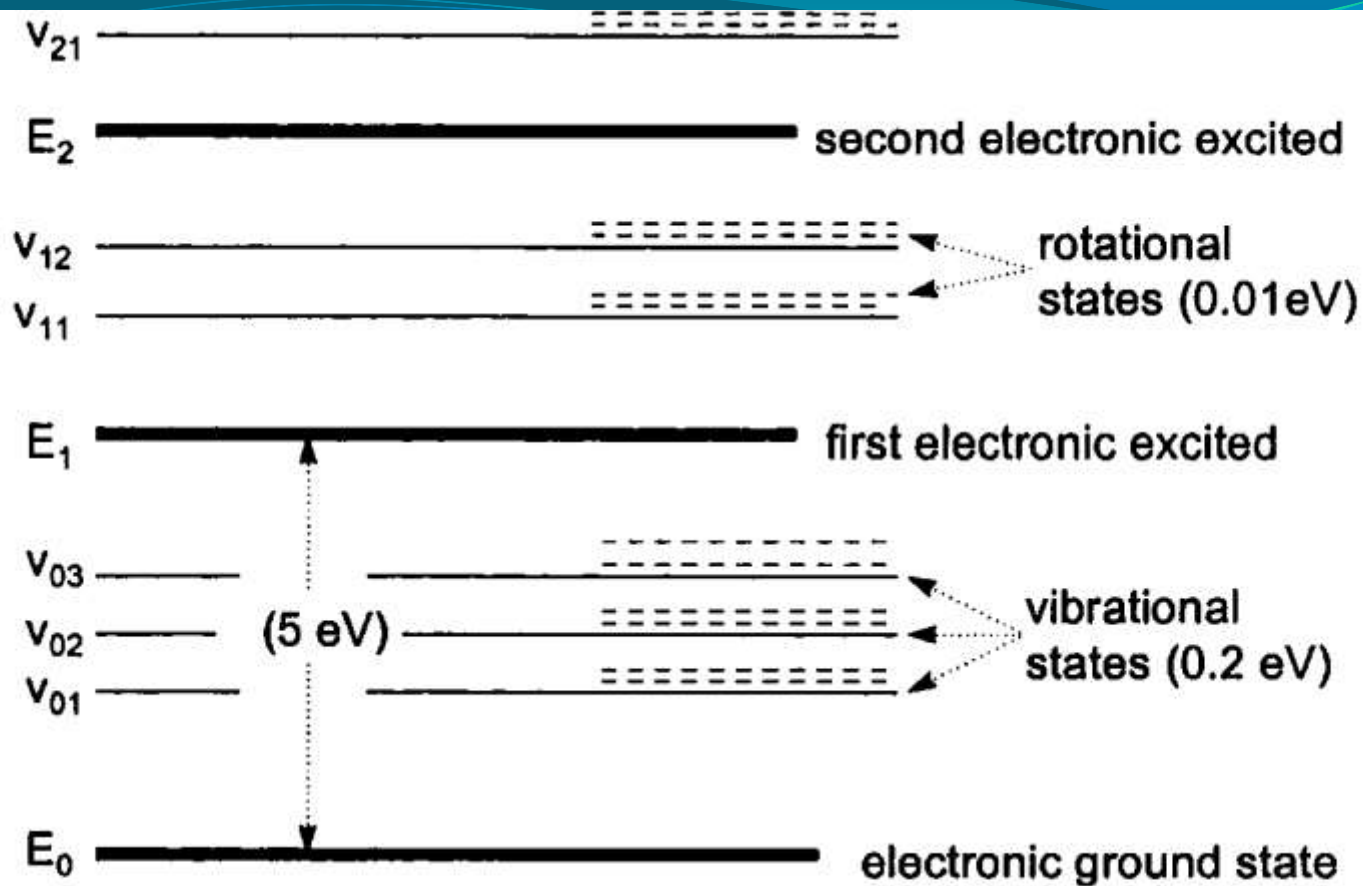
Fig.(5.16) A schematic diagram of the molecular energy levels of an organic system. Electronic ($E_0, E_1, E_2$..) vibrational ($v_{00}, v_{01}, v_{02}$ ..) and rotational states are separated by approximately 5, 0.2 and 0.01 eV respectively (values decrease with energy). In this simple scheme the ground vibrational states, $v_{00}, v_{10}$, etc. are assumed to lie at the same energies as their electronic levels, $E_0$ , $E_1$, etc.

De-excitation is determined by one or more of the following possibilities :

- if there is enough deposited energy to ionize a molecule, the electron is unlikely to be sufficiently mobile to escape the residual molecular ion. Geminate recombination is then most likely to follow.
- when the energy deposited is sufficient to populate a higher electronic state, there is rapid decay ($\sim 10^{-12}$s) of the system to the first excited electronic state. This occurs through radiation-less transitions as excitation energy flows into vibrational and rotational states. Thermal equilibrium is eventually reached with neighbouring molecules.
- the decay of the first excited electronic state can take place to the various vibrational states of the ground electronic state (*i.e.* $v_{10}$ to .... $v_{02}$, $v_{01}$ or $v_{00}$) with the emission of fluorescence light.

The Franck-Condon principle states simply that the time required to make a transition is very short compared to the vibrational periods of the molecule. This means that the inter-nuclear separation remains constant during the transition. In Fig.(5.17) the vertical line represents the transition between $v_{00}$ and $v_{12}$. This is the most probable outcome since the wavefunction overlap is larger than in the case of $v_{00} \rightarrow v_{10}$ or $v_{00} \rightarrow v_{11}$. Transitions to higher electronic states, which are also possible, quickly relax to the ground vibrational level $v_{10}$ of electronic state $E_1$. In doing this, the excitation energy is dissipated non-radiatively.
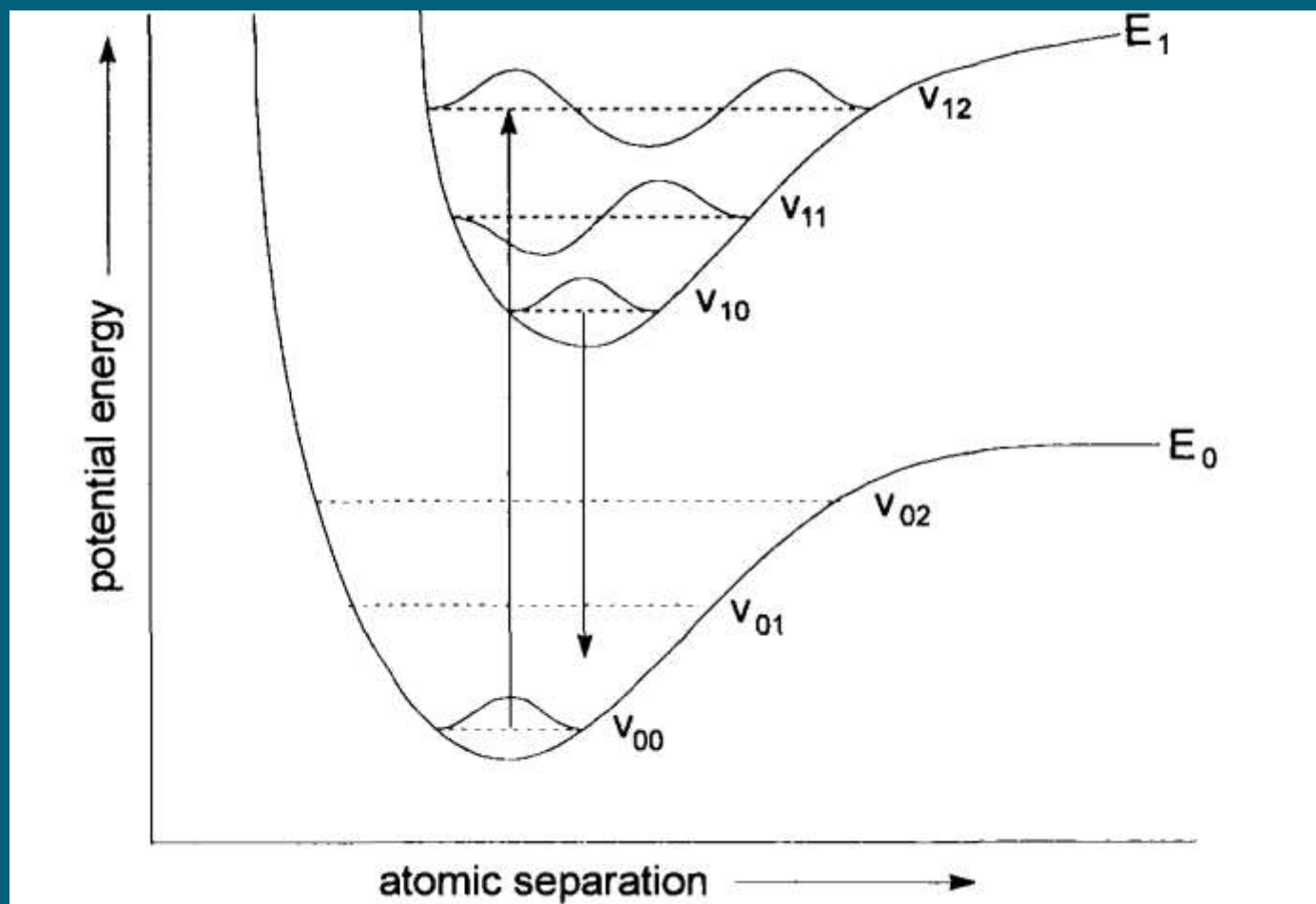
Fig.(5.17)    Application  of the Franck-Condon principle to the organic scintillation process, [7].

The final de-excitation from state $v_{10}$ to the ground state means that the photon emission has a lower energy than the energy initially absorbed.
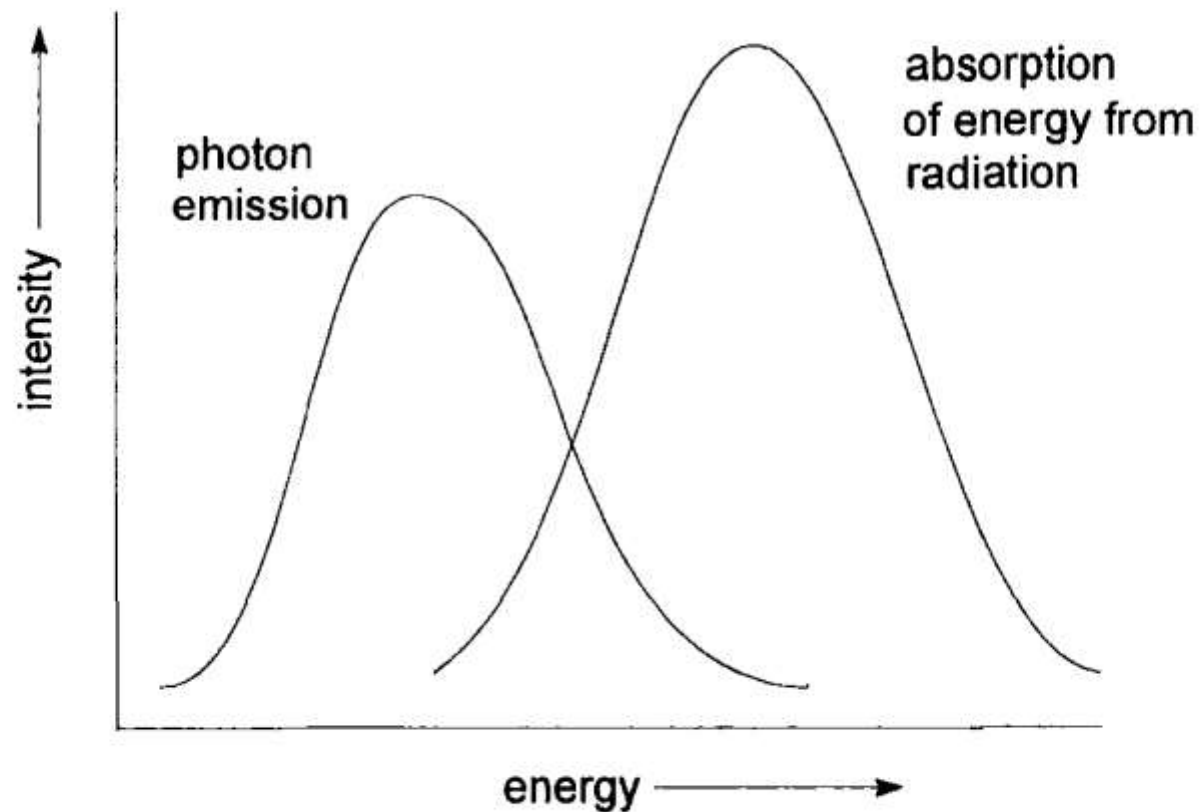


Fig.(5.18)   Schematic spectra of energy absorbed and radiated from an organic scintillator.

Figs.(5.16) and (5.17) show that many electronic and vibrational levels are involved in the organic scintillation process. These lead to the typical spectra of absorbed and emitted energy in Fig.(5.18). Non-radiative (quenching) processes compete with fluorescence in the de-excitation of the scintillator molecules, and account for the smaller integrated area of the photon emission (fluorescence) spectrum.

An important feature of Fig.(5.18) is the area of overlap between the two spectra. Photons emitted in this region are likely to be absorbed in their passage out of the scintillator. As a consequence, the light output depends on the size and shape of the scintillator material. A thin scintillator has a greater overall efficiency than one in which the optical path length is large. This problem can be partly overcome by the introduction of a second (solute) molecule chosen because :

- its absorption spectrum matches the emission spectrum of the primary (solvent) molecule, and
- its emission spectrum is shifted to even lower energies.

Additionally, there is requirement to match the emission spectrum to the absorption spectrum of the photocathode. Sometimes a second solute molecule is added for this purpose. This is called a wavelength shifter.

Table (5.2) Characteristics of some organic scintillators (data taken from [6]). In a comparison with comparable data for inorganic scintillators in Table (5.1), note :
(1) the much shorter decay constants compared with inorganic scintillators,
(2) that the efficiency of organic scintillators is always compared to that for anthracene,
(3) that the densities are not very different from unity,
(4) that discrimination between different types of radiation can be made in some cases by pulse shape discrimination (stilbene, NE 213) and in others by loading the scintillator with the appropriate elements (Gd for neutrons and Sn for photons).

| Type | scintillator | density ($g\ cm^{-3}$) | light output (%) | main decay constant (ns) | maximum emission (nm) | main application |
|---|---|---|---|---|---|---|
| crystal | anthracene | 1.25 | 100 | 30 | 447 | $\gamma, \alpha, \beta$, fast n |
| crystal | stilbene | 1.16 | 50 | 4.5 | 410 | $\gamma$, fast n |
| plastic | pilot U | 1.032 | 67 | 1.36 | 391 | $\gamma$, fast n |
| plastic | NE 115 | | 35 | 225 | 385 | |
| plastic | NE 102 | 1.032 | 65 | 2.4 | 423 | $\gamma, \alpha, \beta$, fast n |
| liquid | NE 213 | 0.874 | 78 | 3.7 | 425 | fast n |
| liquid | NE 224 | 0.877 | 80 | 2.6 | 425 | $\gamma$, fast n |
| liquid | NE 226 | 1.61 | 20 | 3.3 | 430 | $\gamma$ only |
| Gd loaded liquid | NE 313 | 0.88 | 62 | 4.0 | 425 | n only |
| Sn loaded liquid | NE 316 | 0.93 | 35 | 4.0 | 425 | $\gamma$, X-rays |

Certain scintillator molecules have triplet (s = 1) as well as singlet (s = 0) levels which are the result of electronic configurations having $\pi$-bonding orbitals and generally longer lifetimes. Excitation energy can flow from singlet to triplet states and produce a much slower (delayed) fluorescence.

### 5.3.3   Efficiency of scintillation detectors

Factors which affect the overall efficiency, and both the time– and energy– resolution of a scintillation detector are :

- the intrinsic efficiency of the scintillation process in the conversion of energy absorbed from the radiation into optical photons,
- the efficiency of the transportation process of the above photons to the photocathode,
- the efficiency of multiplying the number of electrons released from the photocathode into a measurable charge pulse at the anode.

## (a) Intrinsic efficiency (organic)

The high efficiency of crystalline anthracene makes it the frequently used standard against which all other organic scintillators are compared. For the same reasons, inorganic scintillators are usually compared against the performance of sodium iodide crystals.
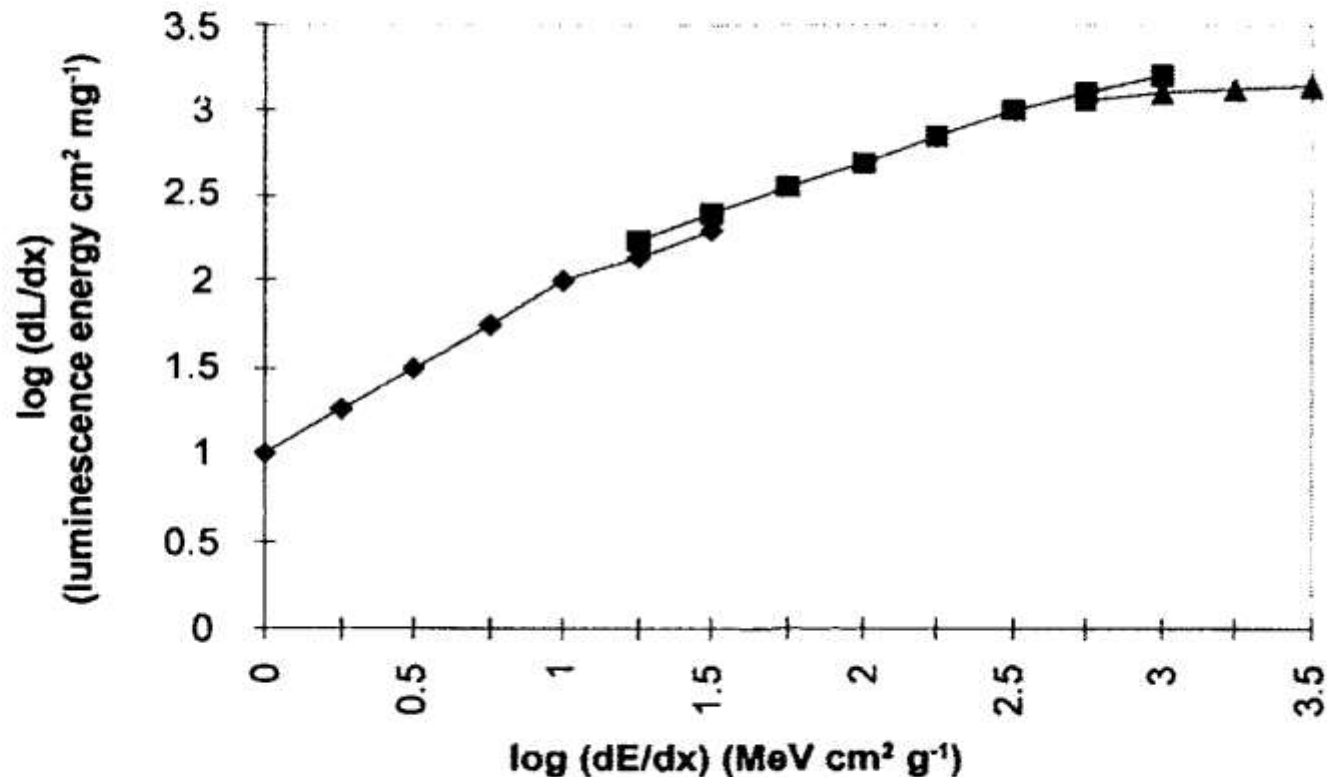


Fig.(5.19)   Specific Luminescence energy versus particle stopping power for an anthracene scintillator crystal. ◆ electrons (600 – 5 keV) :  ■ protons (20 – 1 MeV) :  ▲ α-particles (20 – 1 MeV). The response to photons is determined by the response to electrons. Data from [2].

From a practical point of view, the light output of the scintillator must depend primarily on the efficiency of energy deposition by the incoming radiation. To a first approximation therefore,

$$\text{light output} = \text{constant} \times \frac{dE}{dx}$$

where the proportionality constant depends on the scintillation mechanism. This is therefore different for organic and inorganic materials.

As Fig.(5.19) shows, this dependence is not linear except at low stopping powers, and shows a tendency to saturate at high stopping powers. This non-linearity is due to the various quenching mechanisms (a de-excitation process which does not result in the emission of photons). These reduce the efficiency of light emission and can take place either in close proximity to, or some distance away from, the original site of energy deposition.

A number of semiempirical formulae have been used to relate the specific photon production rate (dL/dx) to the charged particle stopping power (dE/dx) [2], [8]. Light output is expressed as the amount of luminescence energy emitted per unit length of a particle track, dL/dx. The most widely used formula is :

$$\frac{dL}{dx} = \frac{A(dE/dx)}{1 + B(dE/dx)} \qquad (5.13)$$

where A and B are fitted parameters.

There is now widespread use of organic scintillators in plastic form. These comprise a solvent with a primary solute together with a secondary solute as wavelength shifter. The actual molecules and their concentrations used in the plastic can be varied in order to alter the characteristics shown in Table (5.2). Relative efficiencies of light output – as measured by pulse amplitude for a given radiation flux – also vary as a consequence of different solute concentrations, Fig.(5.20).
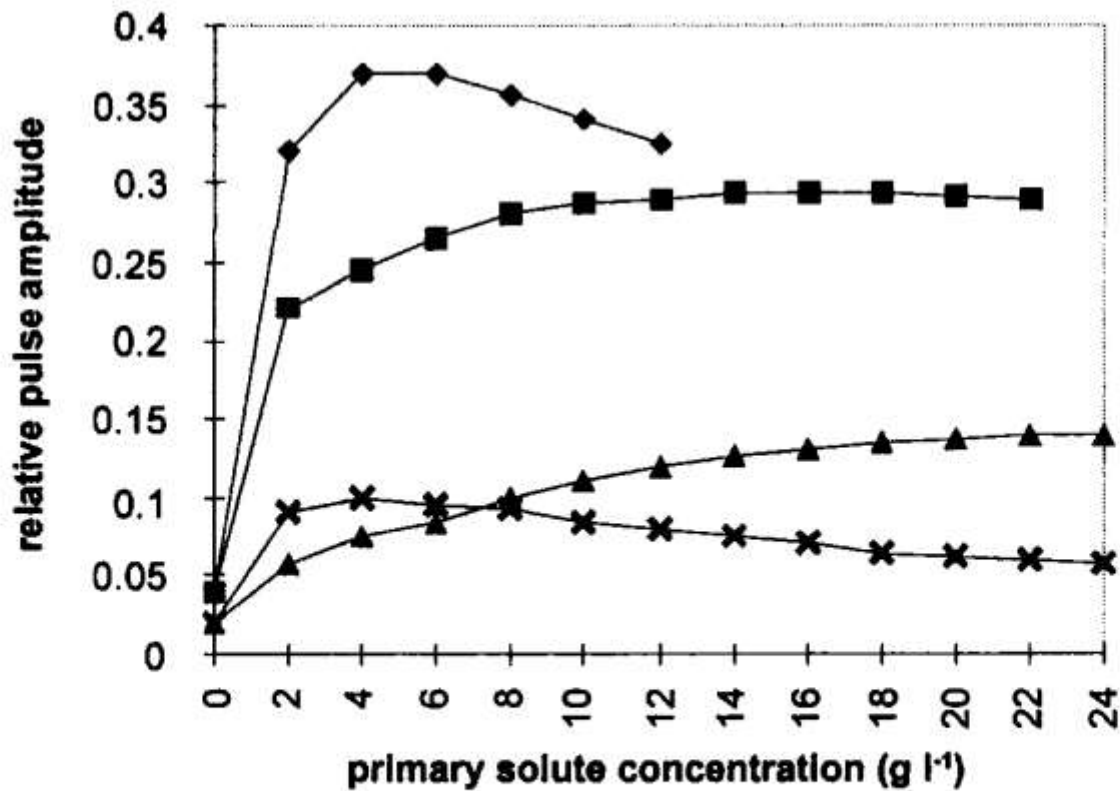
Fig.(5.20) Relative pulse amplitude in a plastic scintillator comprising a polystyrene solvent and different primary solutes at the given concentrations (g litre⁻¹). ◆ 1,1,4,4-tetraphenyl-1-3-butadiene (TPB) : ■ p-terphenyl : ▲ anthracene : ✖ stilbene, [2].

## (b)    *Intrinsic efficiency (inorganic)*

A semiempirical relation between specific light output and stopping power, similar to Eq.(5.13), is also appropriate for inorganic materials. In this case, however, the detection process is initiated by the excitation of a valence electron into the conduction band. Production of usable light from this de-excitation depends on the presence of thallium (or other) activator centres.

The influence of thallium ion concentration on the response of NaI(TI) scintillators to energetic particles at high stopping powers is shown in Fig.(5.21) [9]. For deuterons and $\alpha$-particles there is an optimum thallium mole fraction of $\sim 0.0015$. Note the ordinate scaling factors on pulse amplitude in Fig.(5.21). When the $\alpha$-particle data are expressed in terms of stopping power in Fig.(5.22), the specific luminescence is seen to be almost independent of stopping power but strongly dependent on thallium concentration.
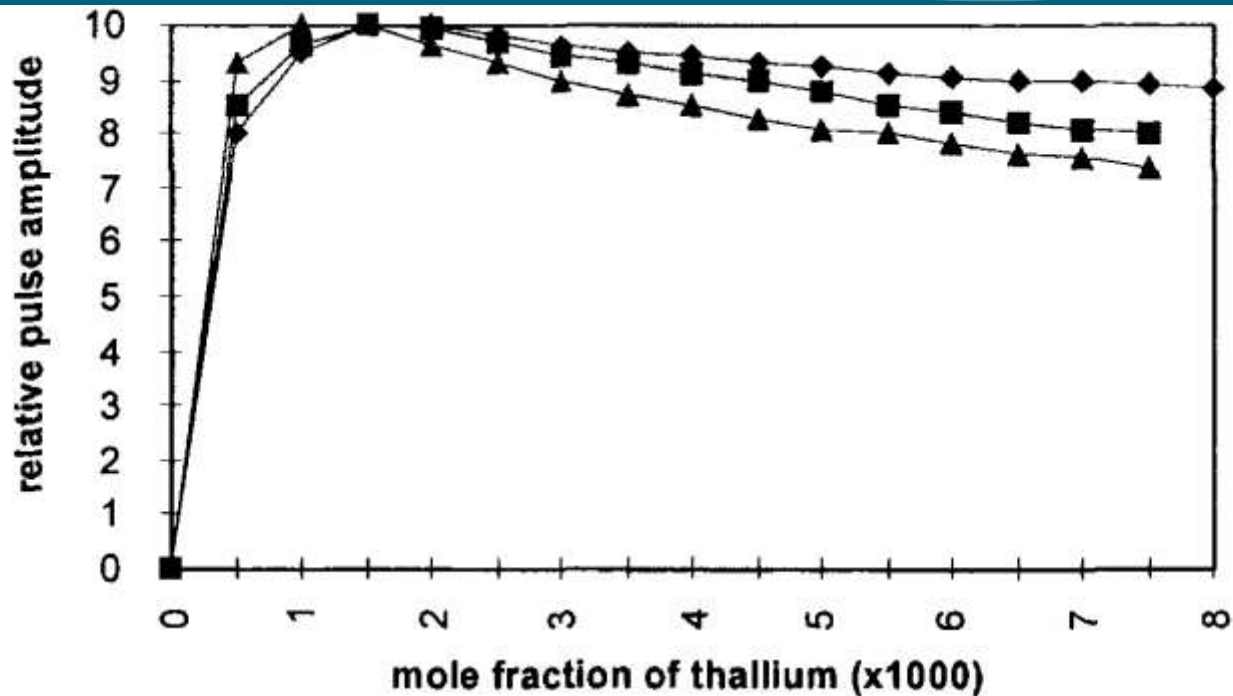
Fig.(5.21)  Relative pulse amplitude from α-particle and deuteron detection in a NaI(Tl) crystal scintillation detector versus thallium concentration (mole fraction × 1000), [9].
◆  polonium α-particles (5.3 MeV) (× 3.33) :      ■ 23 MeV α-particles (× 0.573) :
▲ 11.5 MeV deuterons (× 0.861).

## (c) Additional factors affecting scintillation detector efficiency

An early value for the absolute efficiency (*i.e.* the conversion of absorbed energy into usable photons) of a NaI(Tl) crystal was $14.1 \pm 0.7\%$ [11], with later values in the order of 12% [12]. Similar values for the absolute efficiency of crystalline anthracene vary between 1 and 10% [13].

In addition to the importance of thallium concentration in NaI(Tl), and solute concentration in organic scintillators, there are other practical factors which affect the efficiency of scintillation detectors. The most important of these are :

- The isotropic emission of scintillation photons. Detection of these photons is ideally made over $4\pi$. However, they are usually extracted through only one face of the crystal using a diffuse reflecting material on all other surfaces (*e.g.* MgO paint). This introduces a further reduction in efficiency.

- The window material between scintillator and photocathode, and the material of the photocathode itself. This window must be transparent to the scintillation photons to enable the maximum photon flux to reach the photocathode. From the photocathode electrons are drawn towards the first dynode of the photomultiplier tube by an applied voltage. Each photocathode material has a wavelength-dependent conversion efficiency. The spectral efficiency of the photocathode, which should be matched to the spectrum of photons emerging from the crystal, is quantified using :

$$\text{quantum efficiency (QE)} = \frac{\text{number of emitted photoelectrons}}{\text{number of incident photons}}$$

Quantum efficiency at wavelength $\lambda$ is related to radiant sensitivity, $R$, by :

$$QE(\%) = \frac{124}{\lambda(nm)} \times R(mA / W)$$

Most photocathodes are of the alkali-antimonide type and consist of various atomic combinations, such as Sb-K-Cs, Sb-Na$_2$-K-Cs, etc., which determine the range of spectral sensitivity.

From the wavelengths of maximum emission of the scintillators in Tables (5.1) and (5.2), the most appropriate photocathode materials can be selected. These have highest sensitivity – in the uv for Pilot U and BaF$_2$ – in the blue for NaI(Tl) and anthracene and – in the red for CsI(Tl) and BGO.

- Electron multiplication along the dynode chain. The gain factor of each stage of the photomultiplier is frequently called $\delta$. In such a case, $\delta$ electrons are produced by the first dynode for each electron which is incident from the photocathode. For further multiplication at n dynodes, the overall electron gain should therefore be $\delta^n$. However, $\delta$ is not a linear function of inter-dynode voltage because space charge effects suppress the gain at the later dynodes. The mean gain of a system of $n$ dynode stages, Fig.(5.14), is therefore more appropriately given by:

$$\overline{G_n} = f\ A\ V^n\ exp\left(-n\frac{V}{V_{max}}\right)$$

(5.14)

- In Eq.(5.14), $f$ is the efficiency of collecting onto one dynode all the electrons emitted from the previous dynode and $A$ is the efficiency of secondary electron emission from the dynode surface. $V$ is the applied voltage difference between

stages and $V_{max}$ is the value of $V$ at which secondary electron emission is a maximum. This formula implies that the voltage per stage is constant. In practice, focusing and pulse shaping requirements make the inter-dynode voltages near the photocathode and the anode different from those in the rest of the chain.
- Other systematic factors affecting the efficiency in solid scintillators are the homogeneity of the activator or solute within the primary material, together with its size, shape, purity and surface condition.

Attempts are sometimes made to relate the energy deposited in the scintillator to the final charge produced at the end of the electron multiplication stage. This is quite inappropriate, since the factors which contribute to the loss of efficiency in this conversion process relate only to a particular system. As a consequence, they are rarely quantified.

## 5.3.4 Energy resolution of scintillation detectors

Combined statistical and systematic variations in the processes involved in a scintillation detector lead inevitably to a consideration of energy resolution. The variation in the pulse output when an energy $\overline{E}$ is deposited in the scintillator can be expressed [14] as :

$$\Gamma^2\left(\overline{E}\right) = \Gamma_c^2\left(\overline{E}\right) + \frac{5.56 \times \overline{G}}{\overline{np}\left(\overline{G}-1\right)} \tag{5.15}$$

In Eq.(5.15), $\Gamma\left(\overline{E}\right)$ is the Full Width Half Maximum of the response of the detector when energy $\overline{E}$ is deposited, and $\Gamma_c\left(\overline{E}\right)$ is the contribution to the observed peak due to the intrinsic resolution of the scintillator alone. The mean number of photons per scintillation event is $\overline{n}$, while $\overline{p}$ is the mean probability that a photon which hits the photocathode produces an electron which arrives at the first dynode. The mean gain per dynode stage is $\overline{G}$.

Table (5.3)    Best estimates of intrinsic and overall energy resolution with approximate energy dependence for "standard" scintillation detectors.

| Crystal | approximate energy dependence | $\dfrac{\Gamma_c(\bar{E})}{\bar{E}}$ | $\dfrac{\Gamma(\bar{E})}{\bar{E}}$ | reference |
|---|---|---|---|---|
| NaI(Tl) | $E^{-1/2}$ at 662 keV | 4% at 662 keV | 6 - 10 % | [15] |
| anthracene | $E^{-1}$ at $\infty$ energy | 9% at $\infty$ energy | 15 - 25% | [16] |

Purely statistical contributions lead to a simple relationship between the width of a photopeak and the energy deposited in the scintillator. In most practical situations, statistical broadening dominates over other contributions [6]. On this assumption, we can define the energy resolution $R$ as :

$$R \propto \frac{\Gamma(\bar{E})}{\bar{E}} = \text{constant} \times \frac{\sqrt{\bar{E}}}{\bar{E}} = \text{constant} \times \frac{1}{\sqrt{\bar{E}}} \qquad (5.16)$$