Chapter 4 Scintillation Detectors
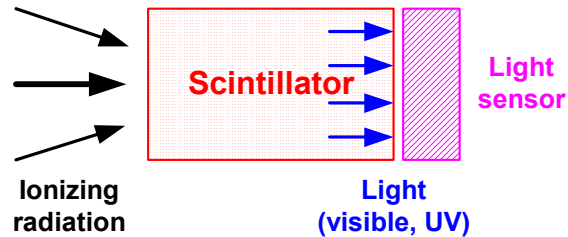
4.1. Basic principle of the scintillator

Fig. 4.1. Principle of the scintillation detectors.

Scintillates are one of the oldest types of radiation detector because measurements could be made with photographic film. Images could be collected or intensity measurements could be made. Measurements were also made with the human eye observing the brightness of frequency of flashes in the scintillator. Nowadays the light output is converted into voltage pulses that are processed in the same way as pulses from proportional counters, semiconductor detectors etc. The whole point of scintillation detectors is that we want to produce a large light output in the visible range.

There are two commonly used types of scintillators, inorganic crystals and organic scintillators. The scintillation mechanism is different for these two types.
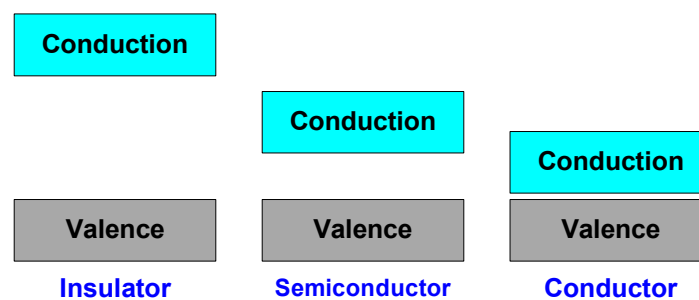
4.2. Inorganic Scintillators

Fig. 4.2. Band structure for electron energies in solids.

The scintillation mechanism depends on the structure of the crystal lattice. In a pure inorganic crystal lattice such as NaI, electrons are only allowed to occupy selected energy bands. The forbidden band or band gap is the range of energies in which electrons can never be found in the pure crystal.

In the pure crystal, absorption of energy can elevate electrons from the valence band to the conduction band leaving a gap in the valence band. However, the return of an electron to the

valence band with the emission of a photon is an inefficient process. Few photons are released per decay, the energy is emitted by other mechanisms. In addition, band gap widths in pure crystals are such that the resulting emitted photon is too high to lie within the visible range.

Small amounts of impurities are therefore added to the crystal. Tl is added to NaI in trace amounts. The impurities are called **activators**, they create special sites in the lattice at which the band gap structure, the energy structure, is modified. The energy structure of the overall crystal is not changed, just the energy structure at the activator sites.

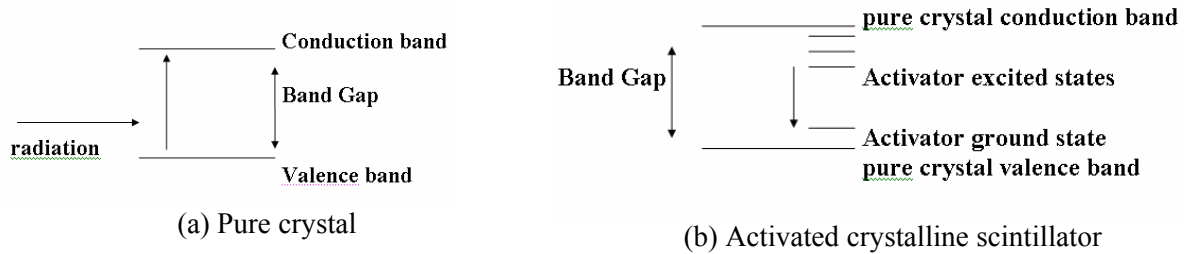(a) Pure crystal

(b) Activated crystalline scintillator

Fig. 4.3. Energy band structure of an inorganic scintillator.

At the few activator sites within the sample, the energy structure is modified. Energy states are created within what would be the forbidden band in the pure crystal. The electron can de-excite through these levels back to the valence band.

The energy levels created by the activator's presence within the crystal are narrower than in the pure crystal.

➡ The photons emitted by the transitions of electrons from upper to lower states will be lower in energy than in the pure crystal: The emission spectrum is shifted to longer wavelengths and will not be influenced by the optical absorption band of the bulk crystal.

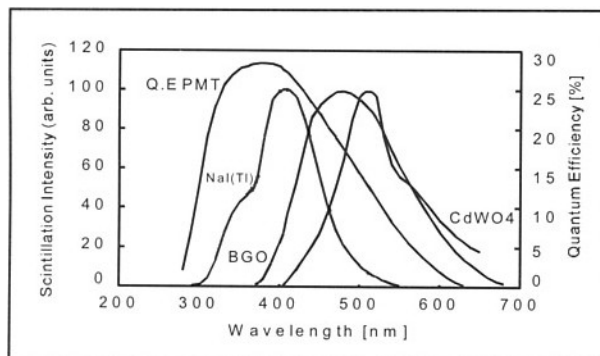➡ The photons are emitted in the visible range.

Fig. 4.4. The emission spectra of several common scintillators.

A charged particle (e.g. a photoelectron) passing through the crystal will create a large number of electron-hole pairs.

- The positive hole quickly drifts to the location of an activator site and the site is ionized. The activator sites are preferentially ionized because the ionization energy of the activator is less than a typical lattice site.
- The electron elevated to the conduction band is free to migrate through the crystal and will do so until it encounters an ionized activation site.
- The electron drops into the impurity site, creates a neutral impurity configuration with its own set of excited states.

- Excited configuration $\xrightarrow{\text{transition}}$ ground state

- De-excitation occurs quickly with a high probability of photon emission.
- The activator is chosen so that the photon is visible.
- The typical half-life of the activator excited states are 10^{-7} seconds with the migration of the electrons through the crystal taking a much shorter time. The timing of the light output is therefore dependent on the half-life of the state.

Types of inorganic scintillators:

Alkali halide: NaI(Tl), CsI(Tl), CsI(Na), LiI(Ei)

Other slow Inorganics: BGO, $CdWO_4$, ZnS(Ag)

Cerium-Activated Fast Inorganics: GSO, YAP, YAG, LSO, LuAP, **LaBr$_3$**

	NaI(Tl)	CsI(Tl)	BaF$_2$	BGO	LSO:Ce	GSO:Ce	YAP:Ce	LuAP:Ce
Emission peak (nm)	410	565/420	310/220	480	420	440	360	365
Light yield (ph/keV)	38	65	11/1.5	8.2	25	9	18	12
Decay time								
Slow (ns)	230	680/3000	600	300		400		
Fast (ns)			0.8		40	60	27	17
Density (g/cm^3)	3.7	4.5	4.9	7.1	7.4	6.7	5.4	8.4
Chemical composition				Bi$_4$Ge$_3$O$_{12}$	Lu$_2$SiO$_5$	Gd$_2$SiO$_5$	YAlO$_3$	LuAlO$_3$
$1/\mu$ (cm) at 140 keV	0.41	0.28	0.29	0.086	0.11	0.16	0.7	0.1
$1/\mu$ (cm) at 511 keV	3.1	2.4	2.3	1.1	1.2	1.5	2.2	1.1
μ_{ph}/μ (%) at 511 keV	18	22	19	44	34	26	4.4	32

The total attenuation and absorption coefficients, μ and μ_{ph}, respectively, were calculated with XCOM (Berger et al., 1999) without including the coherent scattering. The detector materials are: NaI(Tl) and CsI(Tl)—thallium-doped sodium/caesium iodide, respectively; BGO—bismuth germinate; LSO:Ce and GSO:Ce—cerium-doped lutetium/gadolinium oxyorthosilicate, respectively; YAP:Ce and LuAP:Ce—yttrium/lutetium aluminium perovskite, respectively.

4.3. Organic Scintillators

The scintillation mechanism in organic materials is quite different from the mechanism in inorganic crystals. In inorganic scintillators, e.g. NaI, CsI the scintillation arises because of the structure of the crystal lattice.

The fluorescence mechanism in organic materials arises from transitions in the energy levels of a **single** molecule and therefore the fluorescence can be observed independently of the physical state. For example anthracene is observed to fluoresce as
- a polycrystalline material
- a vapor
- part of a mixed solution

Practical organic scintillators are organic molecules which have symmetry properties associated with the electron structure.
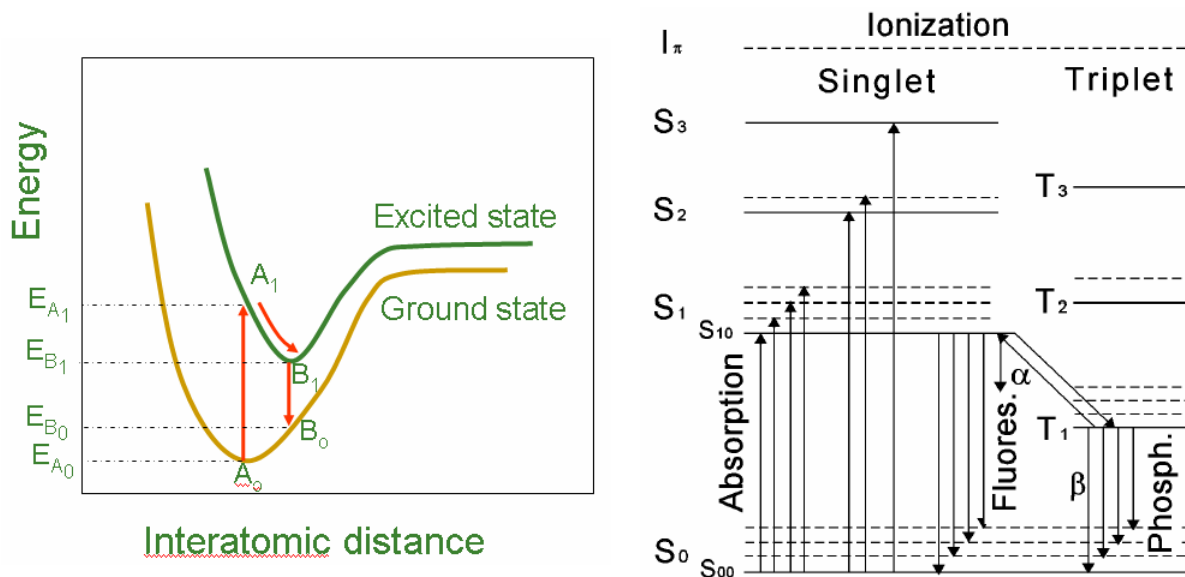

Fig. 4.5. Energy levels of organic molecules.

Energy from a charged particle is absorbed and excites the electron into a variety of excited states - the singlet states (spin = 0) are labeled S_1, S_2, S_3 in Fig. 4.5. For organic scintillators the spacing between S_0 and S_1 is 3 to 4 eV, the spacing between the upper states is much smaller.

Each of the S levels is subdivided into a series of levels with much finer structure (corresponding to the vibrational states of the molecule). The typical spacing is 0.15 eV. The 2nd subscript denotes the fine structure level.

Spacing between S states is 3-4 eV, spacing in vibrational structure is 0.15 eV

Scintillation Mechanism

- At room temperature, average energy is approximately 0.025 eV so all molecules are in the S_{00} state.

- When the charged particle passes through, kinetic energy is absorbed by the molecules, and electrons are excited to the upper levels.

- The higher states S_2, S_3, de-excite quickly (picoseconds) to S_1 state through radiationless transitions (internal conversion).

- States such as S_{11}, S_{12} that have extra vibrational energy and are not in thermal equilibrium with neighboring molecules, quickly lose energy.

➡ After negligibly short time a population of excited molecules in S_{10} state is produced as the net effect of the excitation process.

Scintillation light, prompt fluorescence, is emitted in transitions between S_{10} and the ground state. The prompt fluorescence intensity at time t following excitation is described by

$$I = I_0 e^{-t/\tau}$$

where τ is the fluorescence decay time for the S_{10} level.

In most organic scintillators, τ is the order of a few nanoseconds therefore organic scintillators are fast.

The lifetime for the T_1 state is much longer than the S_1 state. T_1 is populated by a transition called an intersystem crossing. The lifetime of the T_1 state can be ms.

$T_1 \rightarrow S_0$ transitions give rise to phosphorescence (delayed light emission).
T_1 lies below S_0, therefore the wavelength of the emitted phosphorescence is longer than the wavelength of the fluorescent light. The phosphorescent light can be discriminated from the scintillation light on the basis of timing and wavelength.

The energy level scheme explains why organic scintillators can be transparent to their own fluorescence emission. **All fluorescence emissions (except $S_{10} \rightarrow S_{00}$) have a lower energy than the minimum required for absorption.** There is little overlap between emission and absorption spectra, therefore the emitted light mostly passes straight on through the scintillation medium.

Types of organic scintillators:
    Pure organic crystals: Anthracene, Stilbene
    Liquid organic solutions: by dissolving an organic scintillator in a solvent
    Plastic scintillators: dissolving & polymerizing

4.4. Photomultiplier Tubes (PMT) and Photodiodes

In order to provide a useful measurement device, an electrical signal has to be formed from the scintillation light. Two main types of device are used to do this; the photomultiplier tube and the photodiode.

A. Photomultiplier Tube (PMT)

Photomultiplier tubes are extremely good at converting light into an electrical signal; electrical pulses can be obtained from a few hundred visible photons.

Photocathode
Photomultiplier tubes are vacuum tubes in which the first major component is a photocathode. A light photon may interact in the photocathode to eject a low-energy electron into the vacuum. This process can be thought to occur in three steps

1) absorption of the photon and energy transfer to the electron in the photocathode material
2) the migration of the photoelectron to the surface of the photocathode
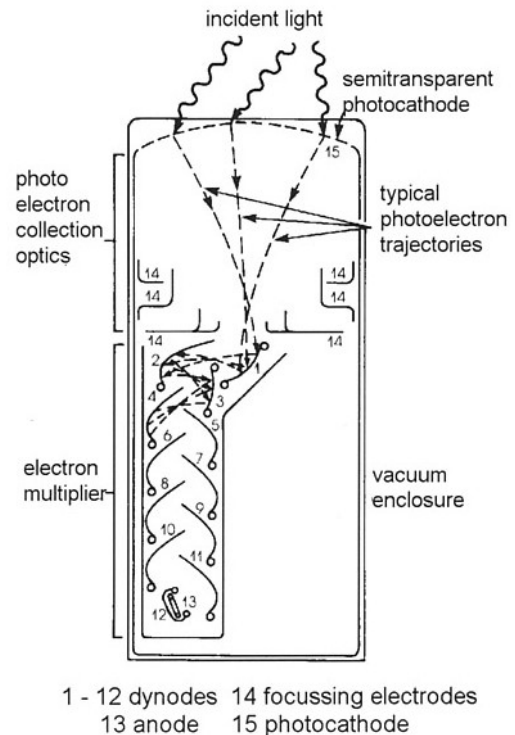3) escape of the electron from the photocathode surface.

Fig. 4.6. Basic elements of a PMT.

The energy available to be transferred from a scintillation photon to an electron is, for blue scintillation light, approximately 3 eV. However, some of this energy is lost in electron-electron collisions as the electron migrates to the surface and then the potential barrier at the surface vacuum interface (the work function) must be overcome in order for the electron to enter the vacuum. There are therefore energy limitations on the system posed by the potential barrier at the surface/vacuum interface and the interactions in the material.

Some limitations are placed on the materials that can be used for the photocathode. Typically metals have a work function of 3 ~ 4 eV, semiconductors 1.5 ~ 2 eV.

In addition, the thickness of the material that can generate photoelectrons into the vacuum is limited, because there is a rate of energy loss as the electron migrates to the surface. For metals this energy loss rate is relatively high and an electron will have its energy drop below the potential barrier after traveling only a few nanometers. In semiconductors the electron can travel 20 – 30 nanometers, before its energy drops below the barrier. In either case only a thin layer of material contributes photoelectrons into the vacuum. The cathodes are sometimes designed so that the light is incident on one side, while the electrons are ejected on the other. The photocathode is therefore extremely thin, because of the problems of electron interaction in the material, and is therefore semi-transparent to visible light. This means that a large proportion (more than half) of the light passes straight through without interacting with the photocathode.

Even if the work function is low, the necessary thinness of the photocathode means that they can only convert, at best, a small proportion of the photons.

The problems of work function and electron interactions in the material mean, in addition, that all photocathodes have an energy cut-off with respect to the frequency of the light that can generate photoelectrons into the vacuum. Some light frequencies cannot create electrons with enough energy to overcome the potential barrier. The energy cut-off is usually in the infra-red or near infra-red part of the spectrum.

The sensitivity of a photocathode is usually quoted in terms of the quantum efficiency. The quantum efficiency of the photocathode is defined as the probability for the conversion of light to an electrical signal and is defined as

Quantum efficiency = number of photoelectrons emitted/ number of incident photons

The quantum efficiency is a strong function of wavelength of the incident light, and an effort is made to match the spectral response of the photocathode to the emission spectrum of the scintillator in use. The average quantum efficiency over the emission spectrum of a typical scintillator is about 15 to 20 percent while the peak quantum efficiency is 25 ~ 30 %. The standard for quotation is the number of photoelectrons per keV energy loss by fast electrons in a NaI(Tl) crystal. For the peak quantum efficiency, about 8 ~ 10 photoelectrons are produced per keV energy loss. Therefore, the average energy loss required to create a single photoelectron (or one basic information carrier) is ~ 100 eV, which is much bigger than the values in gas-filled or semiconductor detectors. Presently, multialkali (based on Na_2KSb) and bialkali (based on K_2CsSb) materials are usually used for the photocathode.
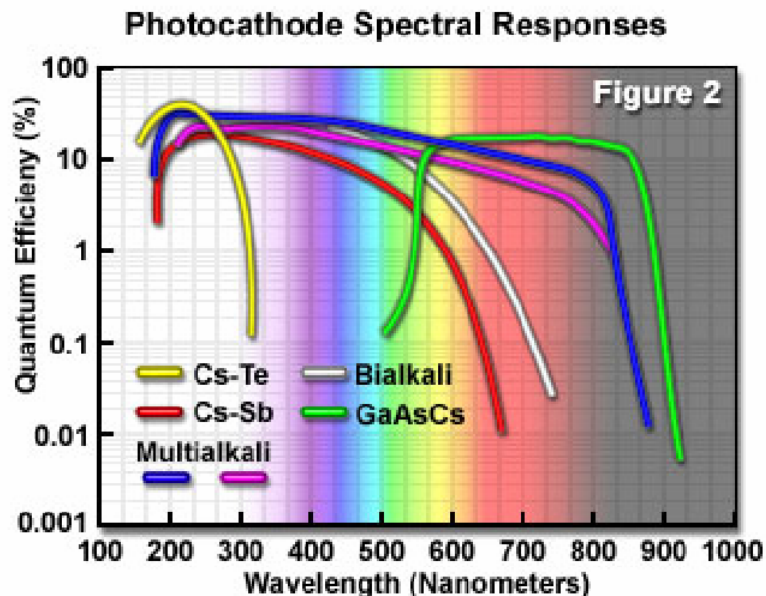

Fig. 4.7. The spectral sensitivities of photocathode materials.

Electron Multiplication

A PMT takes the electrical signal from the photocathode and amplifies through a dynode chain by the process of electron multiplication.

Electrons are ejected from the photocathode into the vacuum with an energy of ≈ 1 eV and are accelerated by a voltage of a few hundred volts toward an electrode. The accelerated electron has an energy of a few hundred electron volts upon arrival at the electrode; this deposition of kinetic energy can result in the re-emission of secondary electrons. It typically takes 2-3 eV to excite an electron in the dynode, so 100 V can theoretically create ≈ 30 electrons.

However, there is the same problem as in the photocathode; electrons must be near the surface to be emitted, otherwise they lose too much energy through collisions and are unable to overcome the potential barrier at the surface. Only a small proportion of the electrons excited up to the conduction band reaches the surface and creates secondary electrons.

The overall multiplication factor for a single dynode is given by

δ = number of secondary electrons emitted / primary incident electron
δ is typically 4 ~ 6 for voltages of a few hundred volts.

Multiple Stage Multiplication

Photomultiplier tubes employ dynode chains of several stages of multiplication to create electron gains of the order of 10^6.

Electrons are created in the photocathode, and accelerated toward the first dyode, where secondary electrons are emitted. The secondary electrons are accelerated by the electric field toward a second dynode, where they create secondary electrons, and so on. This can be repeated many times.

The overall gain for the PM tube $= \alpha \delta^N$

Where N is the number of multiplication stages, α is the fraction of electrons collected by the multiplier structure, and δ is the multiplication factor for a single electrode.

For typical tubes δ is around 5 and α approaches unity. 10 stages of multiplication result in an overall tube gain of 5^{10} or 10^7.

Statistics of Electron Multiplication

The emission of secondary electrons from the dynodes is a statistical process, so the multiplication factor δ is not a fixed constant but varies from event to event around a mean value. Therefore pulses are amplified overall down the dynode chain by a mean value which has some statistical variation.

We measure a final mean voltage that has statistical spread, i.e. we measure not just a single energy, but a mean energy with a spread. The dynode chain therefore affects the energy resolution of the system.

We can assume a simple model where the production of secondary electrons follows a Poisson distribution about an average yield.

A single photoelectron incident on the first dynode results in a mean number of secondary electrons of $\delta \pm \sqrt{\delta}$. The relative variance is thus $1/\delta$.

The process is compounded over N identical stages of the PM tube, and the mean number of electrons collected at the anode is given by δ^N.

The relative variance in this number is $1/\delta + 1/\delta^2 + 1/\delta^3 + \ldots + 1/\delta^N \cong 1/(\delta-1)$

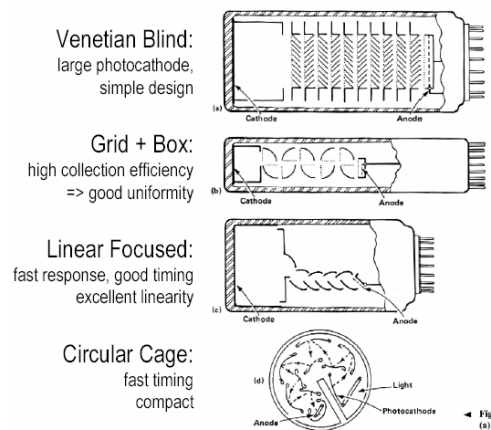The variance in the dynode chain adds to the poor observed energy resolution of scintillation detectors.

Fig. 4.8. Different configurations of PMTs.

B. Photodiodes

An alternative way to detect the scintillation light is the use of a silicon photodiode. This is a semiconductor device which consists of a thin layer of silicon in which the light is absorbed after which free charge carriers (electrons and holes) are created. Electron and holes are collected at the anode and cathode of the diode. Most frequently used are PIN diodes operated in reverse bias mode.

Photodiodes have potential advantages: higher quantum efficiency and compact size. However, electronic noise is a major problem due to the small signal amplitude.

References

1. G.F. Knoll, Radiation Detection and Measurement - 3rd edition (Chapters 16 to 18), John Wiley & Sons, 1999.