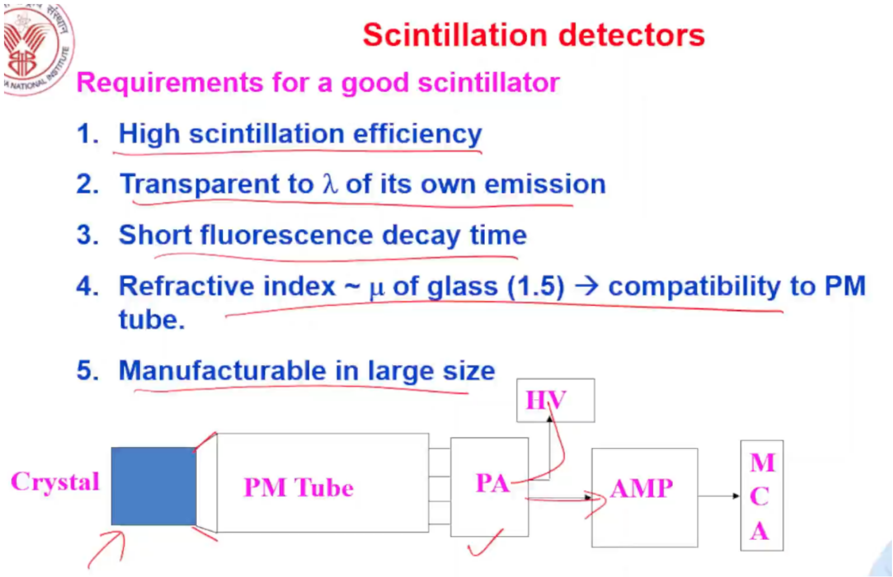
Scintillation detectors

B.S.Tomar

Homi Bhabha National Institute

Lecture-10, module-1

Hello everyone. In the previous lecture, I discussed the gas filled counters like ionization chambers, proportional counters and Geiger-Meyer counters. Now, these gas filled counters you will notice that the density of gases is low and particularly for gamma rays, they are not very suitable. So now I will discuss the detectors which are is solid state, solid state type detectors. They can be emitting light or they can be semiconducting type. So, in the first lecture I will take the scintillation detectors.



Scintillation detectors

Requirements for a good scintillator

1. High scintillation efficiency
2. Transparent to λ of its own emission
3. Short fluorescence decay time
4. Refractive index ~ μ of glass (1.5) → compatibility to PM tube.
5. Manufacturable in large size

As the name itself implies, when the radiation falls on these materials, certain materials are luminescent materials, they fluoresce. So, they are basically the fluorescing materials and when radiation falls on them, they produce light. And then that light can be used to in a photomultiplier tube to generate the electrons. That is the type of detectors we will be discussing in this lecture. So, scintillation detectors are those detectors which when radiation falls on them produce light.

Now, there are certain requirements for a good scintillation material.

First thing is that the scintillator should have high scintillation efficiency. Like if you have a 1 MeV of the gamma ray, then how much of the energy of the gamma ray is utilized in producing light. So, that is an important point. Higher the scintillation efficiency, higher is the detector efficiency, the resolution becomes higher. Then once this light is produced in the crystal, it should be taken out of the crystal and so that the PMT can generate an electron signal from here. So, this should be transparent to the wavelength of its own emission.

The second important property is the decay, the fluorescence decay should be fast so that all the light is generated in a very short span of time. Otherwise, if it is a phosphor material, phosphorescence, then the light will be produced very slowly and we may not be able to integrate the charge that is being collected. So, short fluorescence decay time, typically few nanoseconds to few microseconds is the order of time for fluorescence decay.

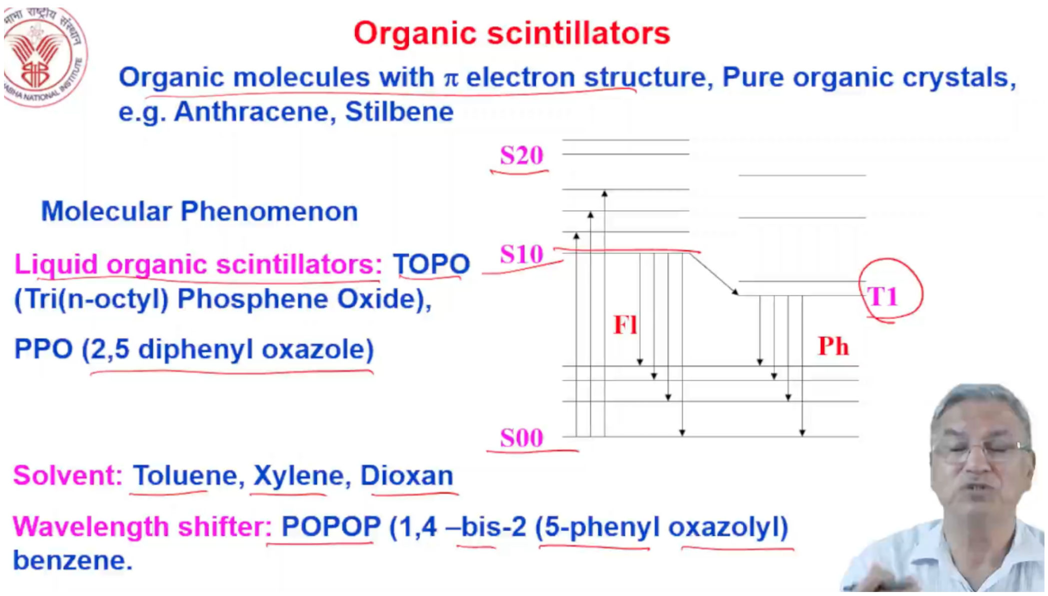
Thirdly, to make it compatible with the PMT, we require that the refractive index of the crystal should be compatible, same order as glass because the PMT has a window made of glass. Sometimes you have the UV radiation being emitted, so you require the UV compatible transparent PMT windows. So, in general for normal PMT, the glass window is used and so refractive index of the crystal should be of the order of 1.5.

And lastly, it should be possible to manufacture these crystals in large sizes.

So, all of them are important requirements for a material to be used as a scintillator. So, the block diagram of the detector scintillator is, you have a crystal. The crystal could be 1 inch by 1 inch, 2 inch by 2 inch. There are some crystals which are available in 3 inches by 3 inch or even 5 inches by 5 inches. So, it depends what type of application you have in mind. Then you have an optically coupled crystal with the PMT. We have a 1 inch by 1 inch crystal and we have 2 inch by 2 inch PMT tube. You have to have some optical coupling. So, there are optically coupling materials.

Then you have the preamplifier. PMT will have a breeder circuit. Apply the high voltage to the anode of the PMT. And then the preamplifier signal is amplified using a signal amplifier. And the amplifier output analog signal you can take to multichannel analyzers or it can go even to a timer scale. So, this is a block diagram of a scintillation detector. The heart of the detector lies in the crystal which has the above properties.

Now, the scintillators can be of two types. One could be the organic scintillator or it could be inorganic scintillators.



Organic scintillators

Organic molecules with π electron structure, Pure organic crystals, e.g. Anthracene, Stilbene

Molecular Phenomenon

Liquid organic scintillators: TOPO (Tri(n-octyl) Phosphene Oxide),

PPO (2,5 diphenyl oxazole)

Solvent: Toluene, Xylene, Dioxan

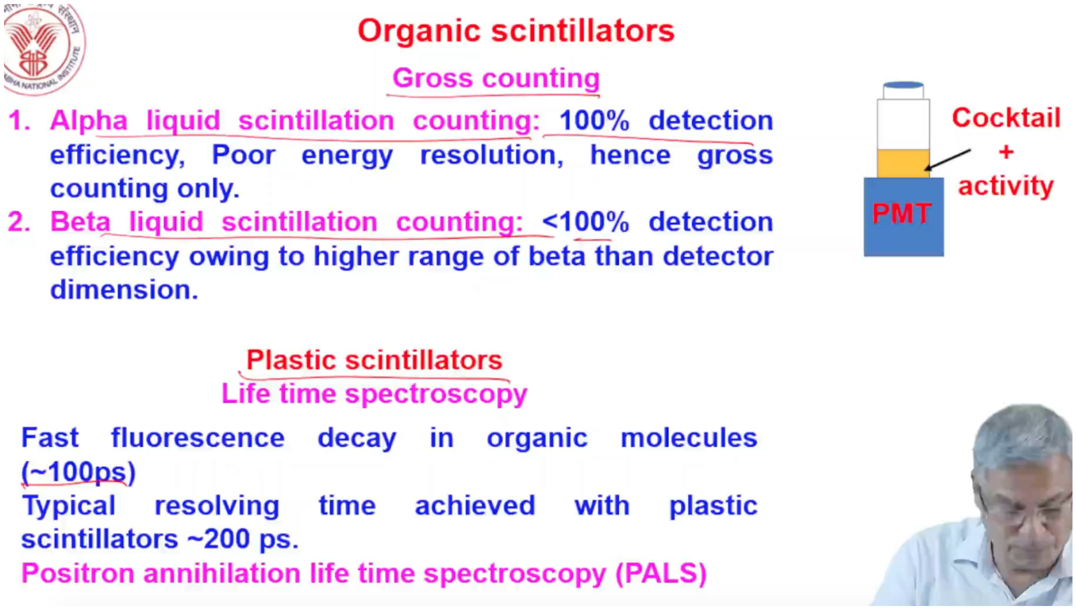
Wavelength shifter: POPOP (1,4 –bis-2 (5-phenyl oxazolyl) benzene.

Organic scintillators are having their own advantages and their own disadvantages. Advantage is that they have a molecular structure. So, each molecule is fluorescent unlike inorganic scintillators where the entire crystal lattice generates the light. So, organic molecules with pi-electron structure like pure organic crystals anthracene, stibine and so on. So, each molecule when the radiation falls on it when it gets excited and the excited molecule can lead to fluorescence. So, this is the typical diagram for a fluorescence molecule, the Singlet ground state and excited states.

You can also have a triplet state which will undergo phosphorescence and the singlet excited state will undergo fluorescence. So, there are several molecules which will undergo fluorescence when they are ionized or they are excited. But the typical ones which are used in detection of radiations are called the liquid organic scintillators like TOPO, this is the tri-n-octyl-phosphene oxide or PPO, 2,5-dipynyl-oxazole. Basically, you have a pi-electronic structure in molecules, conjugated double bonds that will give the fluorescent nature to the molecule. So, you have each organic molecule acting as a fluorescent.

Now, these organic molecules, they have to be dissolved in some solvent, so you can use toluene, xylene and dioxane. So, for organic solution, you can use toluene, xylene or aqueous soluble, you can use dioxane. And many a times, the fluorescence of these molecules may not be compatible with the PMT, photomultiplier tube. So, you use some other reagents called wavelength shifter like POPOP, 1,4-bis-2-phenyl-oxazolyl benzene. So, these molecules take the radiation from the fluorescence molecule, get excited and redshift the light that is emitted.

So, you have a redshift, then that the light that is emitted from these POPOP molecules will be compatible with the PMT. So, these organic molecules are very good for detecting the alpha particles or beta particles of low energy, but they have a low photo fraction because they are mostly hydrocarbon material, carbon, hydrogen, oxygen. And so, the photo fraction, the photo electric effect which require a high Z material that is very small. So, there are some applications wherever they are useful, but particularly for gamma counting, they are not very useful.



Organic scintillators

Gross counting

1. Alpha liquid scintillation counting: 100% detection efficiency, Poor energy resolution, hence gross counting only.
2. Beta liquid scintillation counting: <100% detection efficiency owing to higher range of beta than detector dimension.

Plastic scintillators

Life time spectroscopy

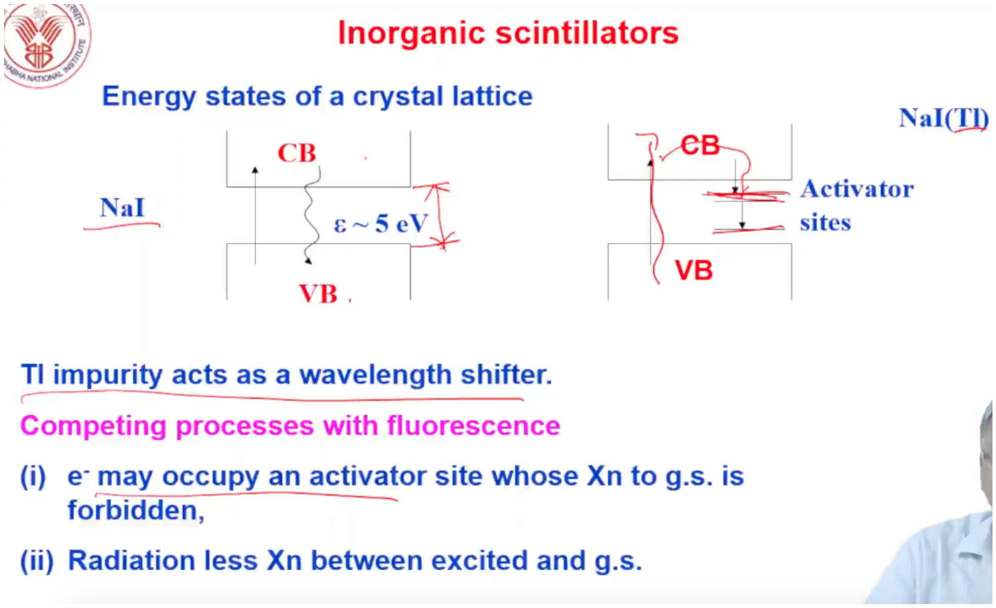
Fast fluorescence decay in organic molecules (~100ps)

Typical resolving time achieved with plastic scintillators ~200 ps.

Positron annihilation life time spectroscopy (PALS)

So, organic scintillators are mostly used for gross counting of alpha and beta. So, alpha liquid scintillation counting, alpha you have, for example, you have radium or you have plutonium, very small quantity microliters of solution can be dissolved in an organic solvent. So, here we have a vial, this 30 ml vial and then you have this yellow is the cocktail, cocktail of like TOPO and PPO and POPOP and you dissolve the activity in this organic solvent. So, you have toluene or xylene, in that you have this activity and then so now this sample, active sample is a part of the detector material itself and then you have to put it the over the PMT and the PMT will take the light signal which you can go to the subsequent circuitry. So, here the advantage is that this sample is dissolved in detector material. Every time you want to count a new sample, you take a new detector.

So, the efficiency is 100% because all radiations are being stopped in the detector material. For beta counting, efficiency is close to 100%, but since the range of beta is more, some part of beta may get lost. But mostly, the scintillation counters are used for alpha beta counting. There are a class of materials which you can polymerize a fluorescent material. So, it's a solid material, but it's still low Z, they're called plastic scintillators. So, these plastic scintillators are used for gamma ray spectroscopy, but for timing, not for energy spectroscopy. So, organic molecules have very short fluorescence decay time of the order of picoseconds. And because of that, they find applications wherever you are doing lifetime spectroscopy. So, lifetime spectroscopy where you use like in positron, annihilation spectroscopy, you are detecting the time between the two events, it's called birth and decay of a positron or a positronium atom. But there you require very fast detectors which will generate a signal within 100s of picoseconds. For both purposes, these organic scintillator-based detectors are very useful, but they do not have higher atomic number. So, for gamma counting, they are not very useful. For timing purposes, yes, they can be utilized.



Inorganic scintillators

Energy states of a crystal lattice

Tl impurity acts as a wavelength shifter.

Competing processes with fluorescence

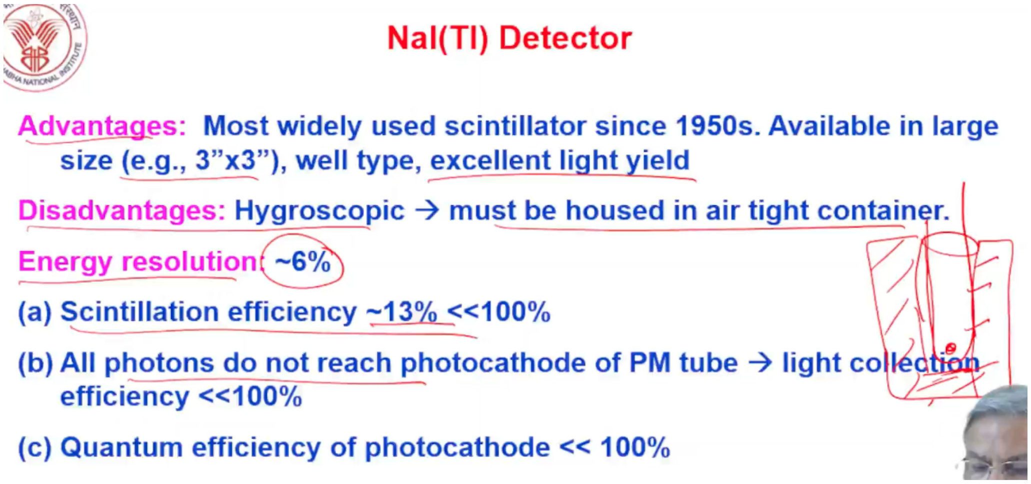
(i) e^- may occupy an activator site whose Xn to g.s. is forbidden,

(ii) Radiation less Xn between excited and g.s.

Now, I come to the workhorse of gamma counting, that is, the inorganic scintillators, particularly the sodium iodide doped with thallium. So, inorganic scintillators are now based on their band structure. So, you have any crystal, a single crystal of sodium iodide, a single crystal, we have the valence band and the conduction band. So, it is an insulating material, that the band gap, the difference between the conduction band and valence band is of 4-5 eV, which falls in the insulating range. Sodium iodide as such is a very poor scintillator, does not emit fluorescence with high yield. And secondly, even if it is emitting fluorescence, that is in the ultraviolet region. So, sodium iodide as such is not used for counting of gamma rays.

Instead, if you dope this sodium iodide with a small quantity 0.1% of thallium, which is as an impurity, then this thallium impurity generates activator sites in between the valence band and conduction band. And now, when the radiation is exciting this valence band electron to conduction band, these electrons can get trapped in the activator excited states. And these activator sites then undergo fluorescence to come to ground state of activator site and thereby emit light, which is compatible to the photomultiplier tubes.

So, by adding 0.1% of thallium to a sodium iodide crystal, you can have a fluorescence material of high fluorescence yield and compatible wavelengths to the PMT. So, the thallium impurity acts as a wavelength shifter like POPOP in the organic scintillators. Even then, the fluorescence intensity is not very high because there are competing processes to fluorescence. For example, the electron when it is generated in the valance band, which is going to conduction band, this may occupy activator site, and all these activator sites may not undergo fluorescence emission to ground state. So, those interactions which give radiations less transitions, are not useful for detection. So, the radiation less transition between excited to ground state and the trapping of this electron in the activator sites, most transition to ground state is forbidden, they will lower the fluorescence intensity of this crystals.



NaI(Tl) Detector

Advantages: Most widely used scintillator since 1950s. Available in large size (e.g., 3”x3”), well type, excellent light yield

Disadvantages: Hygroscopic → must be housed in air tight container.

Energy resolution: ~6%

(a) Scintillation efficiency ~13% <<100%

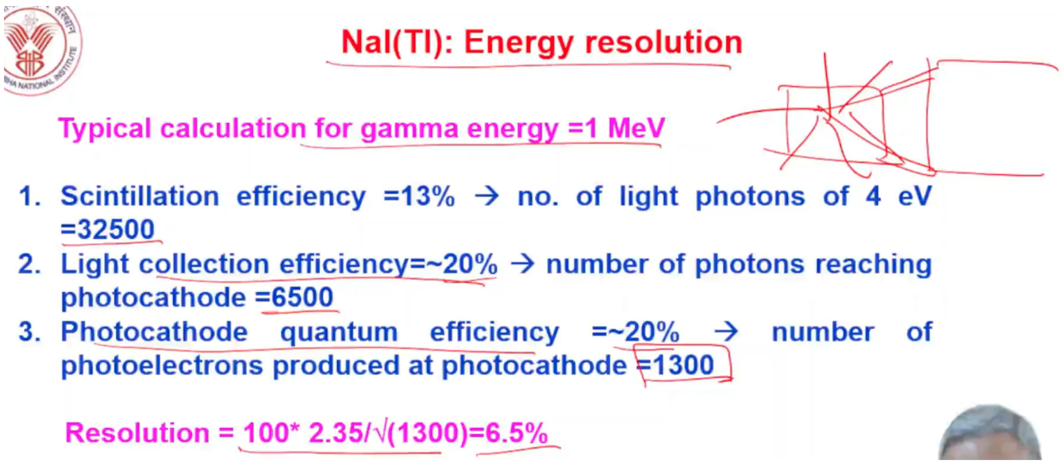
(b) All photons do not reach photocathode of PM tube → light collection efficiency <<100%

(c) Quantum efficiency of photocathode << 100%

So, NaI(Tl), even if this doesn't have very high fluorescence efficiency, but it still it is the work horse of gamma counting. So, advantages are that is most widely used scintillator since 1950s, is available in large sizes, 3 inch by 3 inch, even 5 inch by 5 inch sizes available. You can machine it in such a way you can make a well type.

So, you can have a detector, you know. So, you have a well, you can put the sample in the counting tube, you can put in this well. So, you have a well type sodium iodide and the tube, so the source is here, so it is almost giving you 100% geometric efficiency. So, counting tubes can be inserted in the well of the NaI(Tl). Excellent light yield though it is not very high, but you see 13% in the efficiency, but still in the parlance of scintillation detectors it is called excellent. Only disadvantage it has got, it is hygroscopic in nature. So, when you are making a detector, you have to house it in an airtight container to hermetically seal it in the aluminum case and also so you should make sure that moisture does not get inside.

The energy resolution is not very good, it is about 6%, we will discuss more on this. The scintillation efficiency is one of the best that you have among the different scintillators, though it is much less than 100%, still the 13% is considered to be pretty good. Now, why this resolution is so poor? We expected about 0.1% or 0.2%, but it is 6%. The reason is that all the photons that are produced in the crystal do not reach the photo cathode of the PMT. So, the light collection efficiency is very, very low. Similarly, the photo cathode has its own quantum efficiency is much less than 100%. So, first of all, the crystal itself has got very low quantum efficiency yield, then all the light that is produced does not reach the photo cathode of the PMT and the photo cathode does not have a good quantum efficiency. So, because of these three factors, this NaI(Tl) or for that matter, any scintillator does not have a good energy resolution.



NaI(Tl): Energy resolution

Typical calculation for gamma energy =1 MeV

1. Scintillation efficiency =13% → no. of light photons of 4 eV =32500
2. Light collection efficiency=~20% → number of photons reaching photocathode =6500
3. Photocathode quantum efficiency =~20% → number of photoelectrons produced at photocathode =1300

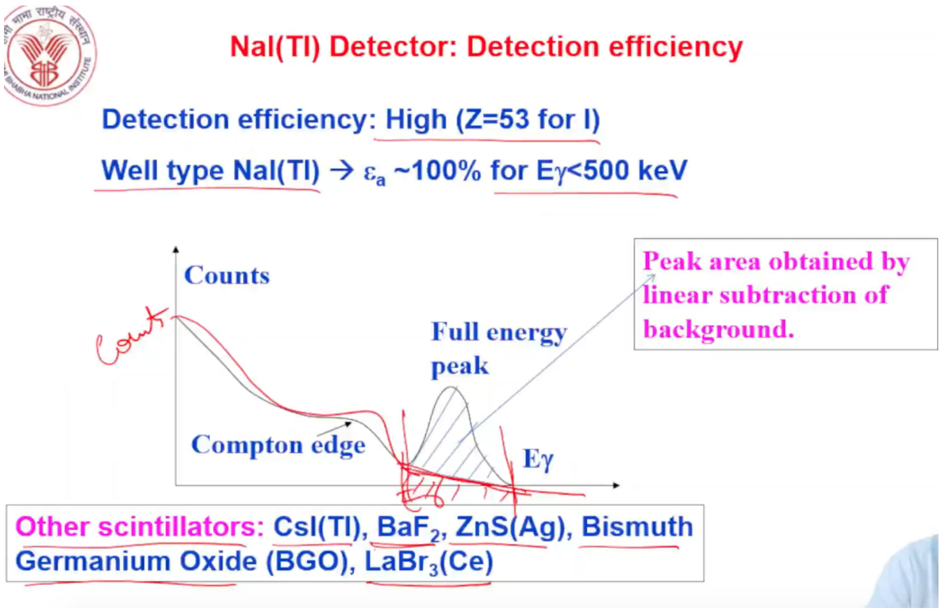
Resolution = 100* 2.35/√(1300)=6.5%

So, let us do an exercise here to compute the energy resolution of NaI(Tl). Why this resolution is so bad? So, typically, let us see for a 1 MeV gamma ray. So, out of the 1 MeV gamma ray, the scintillation efficiency is 13% means what? 13% of this 1 MeV is utilized in generating photons. And so, you can see, so 1 MeV means 100 keV, so 130 KeV energy of the gamma ray is utilized in producing scintillation. And if you have 4 eV as the energy of each photon, then you can calculate how many photons will be produced.

So, you will be having 32500 photons of 4 eV, 32500×4 will be 130000 eV. So, that is how 130 keV, you can calculate 13% of 1000 keV is 130 keV and 130 keV divided by 4 eV will be 32500. Now, this many photons are produced in the crystal, all of them are not going to photo cathode, only 20% of them may reach because we have a crystal here and you have PMT only on one side, and the light can be produced in all directions. So, you will have very small fraction reaching the photo cathode.

So, that is let us say 20%. So, out of 32,500, only 6500 reach the photo cathode and then photo cathode has its own quantum efficiency of say 20%. So, out of 6500, only 20%, 1300 photo electrons will be produced at the photo cathode. So, these are the primary ion pairs like charge carriers, which will determine the energy resolution of detector. Because after this now the subsequent multiplication of electrons in the photo multiplier tube, you will get 10^7 10 10^8 electrons at the anode, they will not increase the resolution. Any multiplication will not improve the resolution, only the primary ion pairs or electrons will determine the resolution.

So, the resolution you can see (100 ×2.35)/√n that is n is 1300. So, it becomes 6.5%. So, I hope this explains the reason for the poor energy resolution of scintillation detectors. Even then, you will find that this NaI(Tl) are widely used in counting of gamma ray radioactive samples. Resolution is poor, 6 to 7%, but the efficiency is very high.



NaI(Tl) Detector: Detection efficiency

Detection efficiency: High (Z=53 for I)

Well type NaI(Tl) → ε_a ~100% for Eγ<500 keV

Peak area obtained by linear subtraction of background.

Other scintillators: CsI(Tl), BaF_2, ZnS(Ag), Bismuth Germanium Oxide (BGO), $LaBr_3$(Ce)

So, one of the positive points of the NaI(Tl) detector is the detection efficiency is very high. Why it is so? Because of the atomic number of iodine that is 53. So, high atomic number means the photoelectric effect is more. So, because of that, the majority of the interaction will be by photoelectric effect, though Compton scattering will also take place, but because of the high Z of the iodine photo fraction is very high.

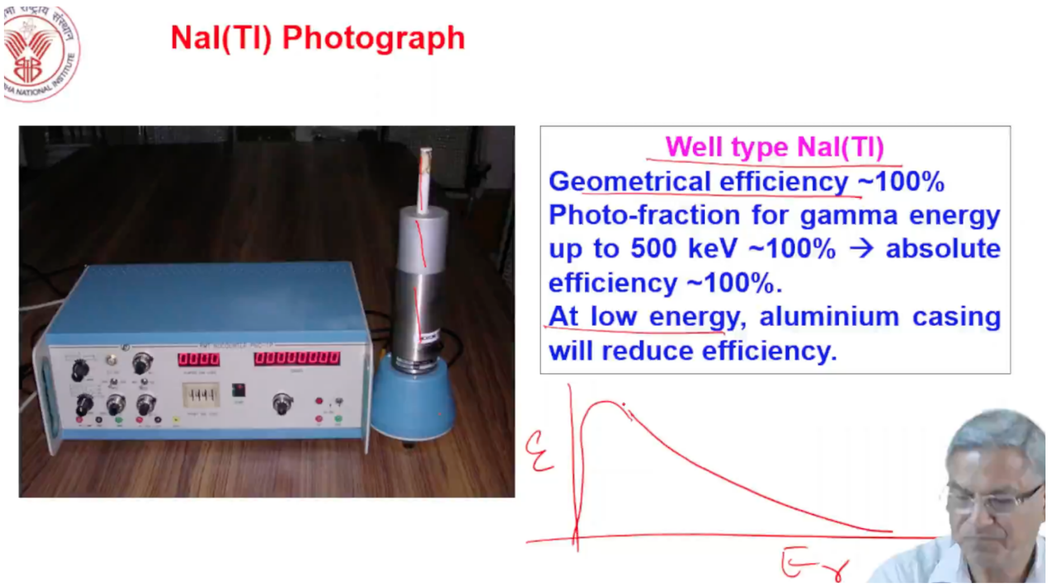
So, typically NaI(Tl) detectors are in fact available also in well type. Just now I explained what is the well type NaI(Tl) and if you put the sample in the well, then you get almost 100% efficiency because geometrical efficiency is now 100%. So, for gamma ray energy less than 500 keV, the efficiencies are of the order of 100%. For example, you have 100 keV, 200 keV gamma ray emitting source, the efficiency is close to 100%.

So, this is the typical gamma spectrum as a function of E_γ, you have the counts and you have this Compton edge. So, you have the Compton edge and this is called the full energy peak. This full energy peak can be due to photoelectric effect or multiple Compton scattering. So, this is a broad peak because of the poor resolution and so it is sitting on a background. So, we can take the area of this peak by what you call as the linear subtraction of the Compton background.

Compton background is linear. So, you can make a trapezium and subtract, take the gross area, subtract the background and get the net peak area of the gamma ray peak. And then this peak area you can divide by time to get the count rate, counts per second. And then subsequently you correct for the efficiency and the abundance of the gamma ray to get the absolute activity, disintegration per second.

So, while NaI(Tl) is the workhorse for gamma counting when you do not need high resolution, but there are other scintillators like CsI(Tl), BaF2. BaF2 emits UV radiation. So, you have to use UV compatible PMT, but it has got fast timings. The NaI(Tl) has got a fluorescent decay time of 230 microseconds, whereas BaF2 has got in 200 picoseconds. ZnS(Ag), Zinc sulfide is not a single crystal. It is a polycrystalline material and doped with silver, silver is like wavelength shifter. So, for alpha counting, we have very thin layer of zinc sulfide powder on a Perspex plate and coupled with the PMT, you can use for alpha counting.

Bismuth germanium oxide, BGO, it's a good scintillator material. High Z of the bismuth makes it very high efficiency, but resolution is worse than NaI(Tl). And recently, one new detector has come in the market, lanthanum bromide detector, which is doped with cerium. So, lanthanum bromide detectors are another class of detectors which we will be discussing very shortly.

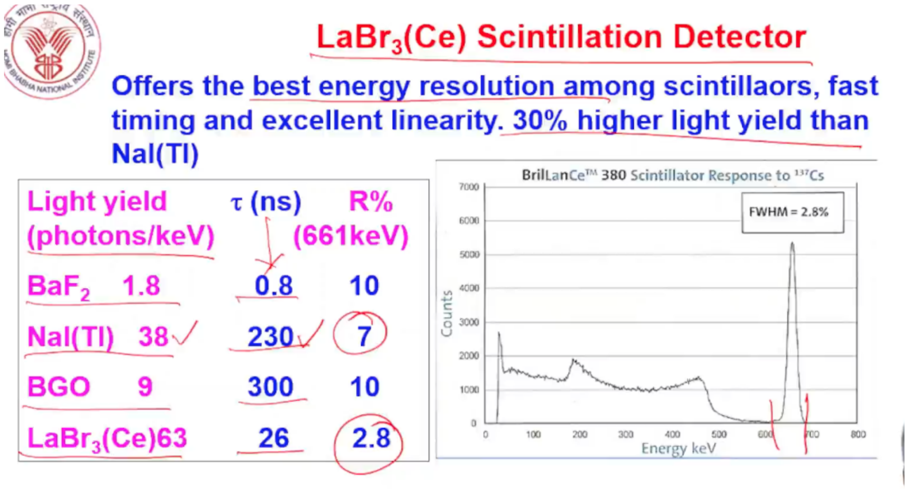


NaI(Tl) Photograph

Well type NaI(Tl)
Geometrical efficiency ~100%
Photo-fraction for gamma energy up to 500 keV ~100% → absolute efficiency ~100%.
At low energy, aluminium casing will reduce efficiency.

So, commercially you will find a NaI(Tl) detector well type. So, you can see the pencil source. Half of it is going inside the detector well and this is the photomultiplier tube. This is the breeder circuit and you have the unit, timer scalar, which will be giving you the counts. It will not give you the gamma ray spectrum. You want to get the gamma spectrum, then you can connect this with a multichannel analyzer. So, the well type NaI(Tl) is very widely used in the laboratories. It has got the 100% geometric efficiency and so for gamma energy of 500 keV or low, it has got the even absolute efficiency as 100%. And at lower energy is let us say 50 keV and below, because the aluminum casing will reduce the gamma ray efficiency for the low energy gamma rays. So, efficiency will become low for low energy. So, typically you will find that the efficiency as a function of energy of the gamma ray, it should have been going down like this, but at low energy again will fall down because of the aluminum window. So, this is how the efficiency for detection of gamma ray will change with the energy of the gamma ray.

So, almost up to 500 keV you will find efficiency is close to 100%. Now, this is the advancement which has taken place in the last one decade or so, more than just 15 years. So, there are always research going on to develop new detector materials which are having better performance than the existing systems. And in this series, a new crystal $LaBr_3$(Ce), lanthanum bromide doped with cerium, has been developed. In fact, in the technical university, Delft, Netherlands, have developed this crystal. This detector offers the best energy resolution among the scintillators, not all, but among the scintillation detectors.



$LaBr_3$(Ce) Scintillation Detector

Offers the best energy resolution among scintillaors, fast timing and excellent linearity. 30% higher light yield than NaI(Tl)

Light yield (photons/keV)	τ (ns)	R% (661keV)
BaF_2 1.8	0.8	10
NaI(Tl) 38✓	230✓	7
BGO 9	300	10
$LaBr_3$(Ce)63	26	2.8

So, like since the 1950s, NaI(Tl) has been ruling for the gamma counting, for gross gamma counting and even for pure gamma sources. But this detector was developed and it has got 30% higher yield than NaI(Tl). So, when we say 30% higher fluorescence yield means, NaI(Tl) was having 13% fluorescent yield, lanthanum bromide has got 16%. So, because of high yield, you will find the resolution is better because more photons are produced for same energy. So, the light yield, photons per keV, NaI(Tl), 38 photons per keV, $LaBr_3$(Ce), 63 photons per keV, compare this with BaF_2 and BGO.

So, if the number of photons per keV are low, resolution is going to be worse. The resolving time, the fluorescence decay time, how fast the fluorescence is decaying, I was telling that this for NaI(Tl) is 230 nanoseconds, for $LaBr_3$(Ce) it is 26 nanoseconds, BaF_2 it is 0.8 nanoseconds, BGO 300 nanoseconds. So, it is better than NaI(Tl) in terms of the fluorescence decay time as well as the number of photons per keV. So, the resolution we can see, NaI(Tl) 7%, lanthanum bromide 2.8%. So, you can see here, this is a typical resolution of a lanthanum bromide detector, 2.8%. So, this has in fact, almost revolutionized the gamma spectrometry with scintillators. We will discuss subsequently that the semiconductor detectors are much better in terms of resolution. But this lanthanum bromide detector, not only in terms of resolution, but efficiency wise also, is better than sodium iodide thallium.

Because of that, now more and more scientists are using LaBr3(Ce). The cost of LaBr3(Ce) is much higher than NaI(Tl) still, cost of production is not that high. And only one drawback is that ^{138}La, which is a long lived isotope of lanthanum, lanthanum-139 is the stable isotope. But because of the presence of ^{138}La, which has got very long half life, which is available in natural lanthanum, and it is emitting a high energy gamma ray. Because of that, if you have a lanthanum bromide detector, you will have a background peak in the gamma spectrum. So, though that background peak will be very, very small, if you count for a long time, you will find a peak will appear somewhere in the high energy region.

So, that is the only drawback. But other than that, lanthanum bromide is going to score all points with respect to sodium iodide and other scintillation detectors. So, I have tried to explain the fundamental principles of scintillation detectors, both organic as well as inorganic. Organic scintillators, good for alpha beta counting in liquid solution, where you get higher 100% efficiency, but not good in terms of energy resolution because of the hydrocarbon nature of the organic molecules. Whereas the inorganic scintillators, they are having rugged high Z materials, sodium iodide, BGO, lanthanum bromide, because of that they have high photo fraction, and hence the resolution is also good, efficiency is also good. But now subsequent lecture, we'll discuss the semiconductors, germanium, silicon, which though have low Z because of that the efficiency is not very high, but you will have excellent energy resolution and they are ideal for gamma rays. So, I'll stop here. Thank you very much. Thank you.