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Energy resolution of scintillation detectors

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ABSTRACT

According to current knowledge, the non-proportionality of the light yield of scintillators appears to be a fundamental limitation of energy resolution. A good energy resolution is of great importance for most applications of scintillation detectors. Thus, its limitations are discussed below; which arise from the non-proportional response of scintillators to gamma rays and electrons, being of crucial importance to the intrinsic energy resolution of crystals. The important influence of Landau fluctuations and the scattering of secondary electrons (δ-rays) on intrinsic resolution is pointed out here. The study on undoped NaI and CsI at liquid nitrogen temperature with a light readout by avalanche photodiodes strongly suggests that the non-proportionality of many crystals is not their intrinsic property and may be improved by selective co-doping. Finally, several observations that have been collected in the last 15 years on the influence of the slow components of light pulses on energy resolution suggest that more complex processes are taking place in the scintillators. This was observed with CsI(Tl), CsI(Na), ZnSe(Te), and undoped NaI at liquid nitrogen temperature and, finally, for NaI(Tl) at temperatures reduced below 0 °C. A common conclusion of these observations is that the highest energy resolution, and particularly intrinsic resolution measured with the scintillators, characterized by two or more components of the light pulse decay, is obtainable when the spectrometry equipment integrates the whole light of the components. In contrast, the slow components observed in many other crystals degrade the intrinsic resolution. In the limiting case, afterglow could also be considered as a very slow component that spoils the energy resolution. The aim of this work is to summarize all of the above observations by looking for their origin.

1. Introduction

γ-Ray spectrometry with scintillation detectors is one of the most important methods in research on and different applications of nuclear science. It covers, for example, a basic study of high energy physics and nuclear physics, environmental studies, nuclear medicine and, recently, homeland security equipment. The great importance of scintillation detectors is associated with their high detection efficiency for nuclear radiation, their ability to measure energy spectra, the possibility of working with a very high counting rate up to 10^7 counts/s, and achievable best time resolution in coincidence or time-of-flight experiments. Their ability to detect a wide assortment of radiations, including γ and X-rays, charged particles and neutrons, their great variety in size and constitution of scintillators make them the best choice in different applications [1].

For γ-ray spectrometry, the following properties of scintillation materials are essential [2–4]:

- High density of the material and a high atomic number of the major element which will assure high detection efficiency of the γ-rays and a high photofraction.
- High light output responsible for high statistical accuracy of the delivered signal.
- Fast decay time of the light pulse reflecting the decay time of the fluorescence components of the crystal, and allowing for high counting rate measurements.
- Low degradation of scintillator energy resolution, which is associated mainly with its non-proportionality characteristics.

The first three properties are straightforward, as they are described by the basic properties of a scintillator. The energy resolution achieved with different crystals sounds the most intriguing. It is a function of the light output, but it is also affected by the internal properties of the scintillator.

Intense experimental and theoretical studies have shown that the energy resolution of scintillators does not only depend on the

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light output of a crystal, but is also limited by the non-proportional response of scintillators [2–70]. The non-proportional response to numerous secondary γ and X-ray quanta as well as secondary electrons produced in the absorption process of γ–rays in the crystal all affect the energy resolution. However, several observations collected in the last 20 years on the influence of slow components of the light pulses on energy resolution suggest that more complex processes take place in scintillators [71]. These experiments were done with CsI(Tl) [72,73], ZnSe(Te) [74], and undoped NaI at liquid nitrogen temperature [34], CsI(Na) [75] and, finally, for NaI(Tl) at temperatures reduced below 0 °C [76]. A common conclusion of these observations is that in the case of scintillators showing two components, or more, of the light pulse decay, the best energy resolution, and particularly the lowest degradation of intrinsic resolution, is obtainable when the spectrometry equipment integrates the whole scintillation light [71].

The most recent studies on scintillation decay times for different energy depositions [77–79], in different inorganic scintillators by a simple experimental method as presented in [78,79], have, for the first time, allowed measurement, separately, of the fast and slow component non-proportionalities of NaI(Tl) [78]. Two scintillation decay modes in NaI(Tl) (the intense fast component of 225 ns and the weak slow component of about 1 μs) have opposite non-proportionality characteristics versus the deposited energy. While the fast component of non-proportionality closely follows the overall non-proportionality of the NaI(Tl) crystal, its slow components exhibit reduction of light output per energy unit at low energies, well known from oxides and Ce-doped scintillators. Similar observations made for CsI(Tl) and CsI(Na) [79].

In contrast, some other crystals, e.g. LuAG:Pr [60] or CsI(In) [80,81], and different samples of undoped NaI at liquid nitrogen temperature [82] showed a deterioration of the energy resolution correlated with the intensity of the slow components. Moreover, other crystals, e.g. LSO, seem to show a particularly poor energy resolution exceeding that expected from the non-proportionality [35]. In this study, the correlation of the non-proportionality and intrinsic resolution of LSO crystals with their thermoluminescence-integrated intensity and then with their afterglow was explored. Further studies were carried out with LGSO:Ce [36] and LSO:Ce,Ca [38] crystals.

Good energy resolution is of great importance for most applications of scintillation detectors. Thus, the limitations are discussed below. These limitations arise from the non-proportional response of scintillators to gamma rays and electrons, as this is of crucial importance to the intrinsic energy resolution of the crystals. The important influence of Landau fluctuations and the scattering of secondary electrons (δ-rays) on intrinsic resolution is pointed out here. Research on undoped NaI and CsI at liquid nitrogen temperature with a light readout by avalanche photodiodes strongly suggests that the non-proportionality of many crystals is not an intrinsic limitation and may be improved by selective co-doping. Finally, the influence of the slow components of light pulses on energy resolution and non-proportionality is discussed.

2. Outline of the problem

The detection process of γ-rays in a scintillation detector can be described by a chain of subsequent processes which introduce uncertainty in the measured energy as a result of γ-rays absorbed in the detector. These processes can be identified as: 1) γ-ray absorption and light generation in the crystal, 2) light collection at the photocathode, 3) photoelectron production at the photocathode, 4) photoelectron collection at the first dynode, and 5) multiplication by the PMT dynodes [2,3].

The energy resolution, $\Delta E/E$, of the full energy peak measured with a scintillator coupled to a photomultiplier (PMT) or avalanche photodiode (APD) can be written as [2,30]:

$$(\Delta E/E)^2 = (\delta_{sc})^2 + (\delta_p)^2 + (\delta_{st})^2 + (\delta_n)^2 \quad (1)$$

where δ_{sc} is the intrinsic resolution of the crystal, δ_p is the transfer resolution, δ_{st} is the statistical contribution of PMT or the photodiode and δ_n is the dark noise contribution connected with the detector's current and the noise of the electronics (negligible in the case of the PMT readout). The intrinsic resolution of a crystal is mainly associated with the non-proportional response of the scintillator [2,3,30]. However, an experimentally determined intrinsic resolution is also affected by many other effects, such as inhomogeneities in the scintillator causing local variations in the light output and the non-uniform reflectivity of the reflecting cover of the crystal.

The statistical uncertainty of the signal from the PMT, corresponding to processes 3–5, can be described as:

$$\delta_{st} = 2.35 \times 1/N^{1/2} \times (1+\varepsilon)^{1/2} \quad (2)$$

where N is the number of photoelectrons and ε is the variance of the electron multiplier gain, which is typically 0.1–0.2 for modern PMTs [2,3,30].

The PMT contribution can be determined experimentally based on the measured number of photoelectrons and it depends on the light output of the crystal being studied, the quantum efficiency of the photocathode and the efficiency of photoelectron collection at the first dynode and gain variation of the PMT.

The transfer component (processes 2–3) is described by variance associated with the probability that a photon from the scintillator results in the arrival of a photoelectron at the first dynode and then is fully multiplied by the PMT. The transfer component depends on the quality of the optical coupling of the crystal and PMT, the homogeneity of the quantum efficiency of the photocathode and the efficiency of photoelectron collection at the first dynode. In modern scintillation detectors, the transfer component is negligible when compared to the other components of energy resolution [25,30].

Since the intrinsic resolution of the crystal (process 1) is mainly connected with the non-proportional response of the scintillator [2–70], the process of γ-ray absorption in the crystal must be considered.

A full-energy peak after gamma energy absorption results from electrons produced in photoelectric absorption followed by emission and subsequent absorption of a cascade of X-rays and Auger electrons, and electrons generated by Compton scattering and terminated by photoelectric absorption. In the end, the amount of light produced corresponding to full energy deposition in the crystal of γ-quanta consists of contributions due to numerous secondary electrons that have a variety of energies. In the low energy region and in small volume crystals, photoelectric absorption dominates and the spread in the amount of light is due to different contributions from the X-ray and the Auger electron cascade. At high energies, mainly in large volume crystals, Compton scattering is largely responsible for secondary electrons of different energies [30].

Another source of spread in total light produced occurs when a given electron does not lose its energy in a unique manner in the crystal but produces further energetic electrons, known as δ-rays. In the low energy region, numerous low energy electrons, typically with energy below 10 keV [9], will affect energy resolution. In the high energy region, scattered electrons have a higher energy and may more significantly influence the spread in the total light produced.

Although the intrinsic resolution of the scintillators appears to be mainly correlated with the non-proportional light response, in

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