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Temperature dependences of $LaBr_3$(Ce), $LaCl_3$(Ce) and NaI(Tl) scintillators

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Abstract

The temperature dependence of light output, energy resolution and decay time constants of the light pulses of NaI(Tl), $LaCl_3$(Ce) ($LaCl_3$) and $LaBr_3$(Ce) ($LaBr_3$) crystals were measured over the temperature range of −30 to 60 °C. In the study of the light output, the number of photoelectrons produced by the scintillators in the XP2020 photomultiplier was measured and corrected for by the temperature dependence of the quantum efficiency determined for 360 and 420 nm, respectively. It showed a high stability of the light output of $LaBr_3$ of about 0.01%/°C and a comparable uniformity of $LaCl_3$ at a long peaking time of 12 μs. The well-known thermal instability of NaI(Tl) was confirmed at a short peaking time of 2 μs. However, a much better stability of NaI(Tl) at low temperatures was observed for a long peaking time. The study of the decay of light pulses from $LaCl_3$ and $LaBr_3$ crystals confirmed earlier measurements, while NaI(Tl) showed a complex behavior at different temperatures. At low temperatures a strong contribution of a slow component of up to 60% of the total light was observed, while at elevated temperatures a well-known initial slow decay was replaced by a delayed maximum and the slow component became insignificant. The results of the study of energy resolution seem to be correlated with the variation of both the light output and a dependence of the decay time constants of the light pulses at changing temperature. This is particularly interesting in the case of NaI(Tl), where different dependencies of the energy resolution as a function of temperature for different peaking times in the spectroscopy amplifier were found. Tests of the XP2020 PMT itself showed that the thermal instability of the gain of the dynode structure of about−0.4%/°C is a dominating effect. The opposite effect on an increasing quantum efficiency, partly compensating for the gain instability, was observed above 10 °C for the longer wavelength of 420 nm.

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1. Introduction

The temperature dependence of the light output of scintillators is the major obstacle in the isotope identification by the hand-held isotope identifiers and in the radiation portal monitors used in border monitoring. The required temperature range of −30 °C to 50 °C creates serious technical problem to compensate for a change of the light output of the scintillators in gamma spectrometers. At present, NaI(Tl) crystals are mainly used for this purpose. The realization of instruments with new crystals, such as $LaCl_3$ and $LaBr_3$ [1–4], characterized by a superior energy resolution, are proposed, particularly, in isotope identifiers [5].

The temperature dependences of the most widely used NaI(Tl) crystal were studied since many years [6–8]. It showed a maximum light output at about 20–30 °C and then a reduced light output to about 70% at −40 °C and to about 95% at 60 °C [9]. Moreover, it was also published that the effective decay time constant of the NaI(Tl) light pulse exhibits a strong dependence at low temperatures, growing up to about 700 ns at −40 °C [9]. The recent

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studies showed a correlation of the light output measured by the digital spectrometer and the decay time constant and this principle was used to stabilize the energy spectra [10]. In contrast, due to a full integration of the light pulse by means of a gated integrator amplifier, the photomultiplier (PMT) anode signal showed a linear decrease of the pulse height with rising temperature [8], which allows a simple compensation.

According to Saint-Gobain, the thermal stability of the new $LaCl_3$ and $LaBr_3$ crystals is much better [11,12]. The light output of a $LaCl_3$ crystal, at a long shaping time constant of 16 μs, is stable within $\pm 5\%$ for the temperature range of -50 to 100 °C. However, at a short shaping time constant of 1 μs, the light output is decreasing linearly down to about 40% at –50 °C [11]. Both measurements were done at varying temperatures of the crystal, while the temperature of the PMT was kept constant.

The temperature performance of the $LaBr_3$ crystal is much better. Again, according to Saint-Gobain, the light output is stable within $\pm 2\%$ in the temperature range of -50–100 °C, independently of the shaping time constant [12]. This confirms a superior performance of $LaBr_3$ crystals for border monitoring instrumentation.

The thermal behavior of commonly used scintillation detectors depends also on the stability of PMT's, gain and quantum efficiency (QE) of the photocathode [13]. The spectral sensitivity characteristic of photocathodes does not vary much with temperature. The most prominent variation is usually observed at wavelengths close to the photoemission threshold. A typical value of about $\pm 5\%$ for the temperature range of -40 °C to 60 °C was reported in Ref. [13] for 405 nm wavelengths. At shorter wavelengths particularly for the $LaCl_3$ and $LaBr_3$ crystals, exhibiting the peak emission at about 360 nm, the variation is very small.

Dynode secondary emission also depends on temperature affecting the gain of the PMT. The temperature coefficient of gain is usually negative and depends not only upon the composition of the dynodes but also upon the photocathode process and, to some extent, the structure of the multiplier. For CuBe and AgMg dynodes a typical coefficient is about $-0.1\%/°C$ for PMTs equipped with bialkali photocathodes [13].

The aim of this work was to study the temperature dependent behaviors of NaI(Tl), $LaCl_3$ and $LaBr_3$ crystals. The measured quantities covered the light output of the crystal, its energy resolution and the decay time constants of the light pulses. The tests of the light output and energy resolution were carried out with the crystals coupled to the very well-known Photonis XP2020 PMT. The temperature of the whole detector, including PMT and its base, was varied in the range of -30 to 60 °C. The crystals were irradiated by 662 keV γ-rays from a ^{137}Cs source and the peak position and its energy resolution were recorded. To exclude the variation of the PMT gain shift, the temperature stability of the single-photoelectron peak was measured, which allowed determining the thermal dependence of the photoelectron number. Further, the variation of the light output and the relative thermal stability of the photocathode QE was estimated measuring the photoelectron number for light pulses from two light-emitting diodes (LED) providing light with defined wavelengths of 360 and 420 nm, respectively.

In parallel to the above-mentioned measurements, the light pulse shapes of the studied crystals were measured at different temperatures. The crystals coupled to a very fast Hamamatsu R5320 PMT [14], were studied by means of recording the waveforms taken from the PMT anode by a Tektronix Digital Oscilloscope type TDS5034B. The analysis of the light pulse shapes allowed a better understanding of the temperature dependence of the light output and energy resolution.

2. Experimental details

All the studies were carried out on ∅25 mm × 25 mm $LaCl_3$ and $LaBr_3$ crystals delivered by Saint-Gobain and on a ∅25 mm × 30 mm NaI(Tl) from Amcrys-H. The nominal Ce doping of the $LaCl_3$ and $LaBr_3$ crystals were equal to 10% and 5%, respectively, selected by Saint-Gobain to get the optimal performance of the crystals. The crystals were coupled to the Photonis XP2020 PMT, no 25377, equipped with a standard bialkali photocathode and CuBe dynodes. The XP2020 was characterized by a blue sensitivity of 10.4 μA/lmF and a low dark noise of about 240 cps. The latter quantity was considered to be of importance, to avoid a high counting rate of single photoelectrons at elevated temperatures. The anode signal of the PMT was sent to an Ortec 113 scintillation preamplifier and then to a Tennelec 244 spectroscopy amplifier working with 2 and 12 μs peaking times, respectively. It allowed for the inspection of the thermal instability of the light output and energy resolution depending on the amount of integrated light of the scintillators. The bipolar shaping in the spectroscopy amplifier was used in all measurements, particularly important for recording single-photoelectron spectra at elevated temperatures. A PC-based multichannel analyzer Tukan 8 K [15] recorded the energy spectra. Peak positions and their full-width at half-maximum (FWHM) were obtained from a Gaussian fit, which included a separation of double peaks, if necessary. To control the stability of the PMT, the position of the single-photoelectron peak was recorded during the measurements of $LaBr_3$. Its low afterglow does not disturb the single-photoelectron spectrum measured with the crystal coupled to the PMT [16].

The whole detector, including the PMT and its base, was inserted to the climatic exposure test chamber which provides a computer-controlled thermal environment. The temperature in the test chamber was varied in the range of -30 up to 60 °C in steps of 10 °C/h. The temperature of the crystal was additionally monitored by a temperature sensor and its equilibrium was checked by observing the stability of the 662 keV peak near the end of