Temperature dependences of LaBr₃(Ce), LaCl₃(Ce) and NaI(Tl) scintillators


Soltan Institute for Nuclear Studies, PL 05-400 Otwock-Swierk, Poland
Target Systemelectronic GmbH, Koebner Str. 99, D-42651 Solingen, Germany

Received 26 May 2006; received in revised form 12 June 2006; accepted 15 June 2006
Available online 10 July 2006

Abstract

The temperature dependence of light output, energy resolution and decay time constants of the light pulses of NaI(Tl), LaCl₃(Ce) (LaCl₃) and LaBr₃(Ce) (LaBr₃) 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 LaBr₃ of about 0.01%/°C and a comparable uniformity of LaCl₃ 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 LaCl₃ and LaBr₃ 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.

PACS: 29.40.Mc; 85.60.Ha

Keywords: NaI(Tl); LaCl₃ and LaBr₃ scintillators; Thermal instability of light output; Decay time constants of light pulses; Energy resolution; Photomultiplier thermal instability

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 LaCl₃ and LaBr₃ [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
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 LaCl₃ and LaBr₃ crystals is much better [11,12]. The light output of a LaCl₃ crystal, at a long shaping time constant of 16 µs, is stable within ±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 LaBr₃ crystal is much better. Again, according to Saint-Gobain, the light output is stable within ±2% in the temperature range of −50–100 °C, independently of the shaping time constant [12]. This confirms a superior performance of LaBr₃ 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 ±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 LaCl₃ and LaBr₃ 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), LaCl₃ and LaBr₃ 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 ¹³⁷Cs 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 ø25 mm × 25 mm LaCl₃ and LaBr₃ crystals delivered by Saint-Gobain and on a ø25 mm × 30 mm NaI(Tl) from Amcry-H. The nominal Ce doping of the LaCl₃ and LaBr₃ 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/lnF 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 LaBr₃. 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
each 10°C period. To avoid an influence of the PMT base on its gain, the same type of resistors was used in the voltage divider, characterized by the same temperature coefficient.

The temperature dependence of the QE was determined by the measurement of the number of photoelectrons due to the light pulses produced by the LED working in constant temperature and driven by a high-stability pulser. Two LEDs, RLT360-05-10 and RLT420-3-30, with a narrow peak emission of about ±10 nm at 360 and 420 nm wavelengths, were used. The light was transmitted to the PMT by an optical fiber. The selected LEDs fit well to the peak emission of LaBr₃ and LaCl₃ in the case of RLT360 LED and to NaI(Tl) in the case of RLT420 LED.

The light pulse decay time constants of the studied crystals were measured independently by means of recording the waves taken from the PMT anode by a Tektronix Digital Phosphor Oscilloscope type TDS5034B. In those measurements a very fast Hamamatsu R5320 PMT, characterized by the width of the single-photoelectron pulse of 750 ps FWHM was used. The detector was irradiated by γ-rays from a ¹³⁷Cs source. To quantify the decay curves, they were fitted mainly with the sum of two exponential terms on the linear background.

3. Thermal dependency of the light output

In the first part of the study the thermal dependence of the light output of the selected scintillators was measured. To exclude the influence of the instability of the PMT, the position of the single-photoelectron peak was measured versus temperature and the number of photoelectrons produced by the crystals was determined. Moreover, the inspection of the relative QE was carried out at 360 and 420 nm wavelengths to make a further correction for a variation of QE versus temperature. The selected wavelengths correspond approximately to the peaks of the emission spectra of LaCl₃ and LaBr₃ crystals, and that of NaI(Tl) equal to 350 nm [11], 370 nm [12] and 420 nm [9], respectively.

The determination of the position of the single-photoelectron peak allowed measuring the variation of the PMT gain shift and the thermal dependence of the photoelectron number produced in the photocathode by the tested scintillator. The photoelectron number was measured by comparing the position of the 662 keV peak of γ-rays from a ¹³⁷Cs source detected in the crystal to that of the single-photoelectron peak [17].

Due to a low afterglow of the LaBr₃ crystal [16], the single-photoelectron peak positions were measured during the stability test of the crystal done with a ¹³⁷Cs source, changing only the gain settings of the TC244 amplifier. Then, they were applied later on for the remaining crystals.

3.1. Thermal instability of the XP2020 photomultiplier

Fig. 1 presents spectra of the single photoelectrons measured at −20 and 40°C temperatures. A large decrease of the peak position is seen at the elevated temperature. Fig. 2 shows the dependence of the single-photoelectron peak position on PMT temperature, reflecting the variation of the PMT gain. Below 20°C, an instability corresponding to about −0.1%/°C is observed, while above, it is equal to about −0.4%/°C.

Fig. 3 presents the relative QE versus temperature, normalized to 1 at 20°C. At the short wavelength of 360 nm, a weak decrease of the QE with increasing temperatures is observed, corresponding to about −0.02%/°C. At the longer wavelength of 420 nm, the QE is increasing for temperatures above 10°C with a slope of about 0.2%/°C. It shows a very good stability of the QE at short wavelengths and a stronger sensitivity to temperatures above 10°C at longer wavelengths. It confirms the general statements given in Ref. [13].
Fig. 4 presents the temperature variation of the PMT response to the light of 360 and 420 nm wavelengths, respectively, including that of the QE. The study of the XP2020 PMT showed a dominating effect of the instability of the PMT gain, because of a negative temperature coefficient of the secondary emission of the dynodes. The QE is affected mainly at elevated temperatures and at the longer wavelength of 420 nm. For the shorter wavelength of 360 nm, corresponding to the peak emission of LaCl₃ and LaBr₃ crystals, the QE instability of approximately 0.02% is practically negligible. At low temperatures, below 0 °C, the stability of the XP2020 is very good, corresponding to about 0.1%/°C centigrade. At elevated temperatures the PMT gain is negative and more significant, however, the PMT sensitivity is partly compensated, at 420 nm, by the QE positive temperature dependence (see Fig. 4). The PMT sensitivity is defined here as the product of the dynode structure gain multiplied by the QE.

3.2. The LaBr₃ crystal

In the case of the LaBr₃ study the measurements were done only with a short peaking time of 2 μs. This crystal is known not to inhere any slow components of the light pulse [2], confirmed also below in the study of the light pulse decay. Fig. 5 presents the energy spectra of γ-rays from a ¹³⁷Cs source measured at −20 and 40 °C. Note a large shift of the peak position, while the energy resolution was only weakly affected by the increased temperature. The energy resolutions were found by the fit of a Gaussian curve to the peaks.

Fig. 6 shows the shift of the 662 keV peak versus temperature plotted together with that of the single-photon peak, following Fig. 2. Both curves are shown in relative scale, normalized to 1 at 20 °C temperature. It exhibits a common curve within the experimental errors.
The measured positions of the PMT single-photoelectron peak and that of the 662 keV \(\gamma\)-peak allows determining the number of photoelectrons produced by LaBr\(_3\) for each temperature of the studied detector. It is plotted in Fig. 7 together with the points corrected for the relative QE, following data of Fig. 3 for 360 nm. It reflects the variation of the LaBr\(_3\) light output with temperature.

The fitted horizontal line to the experimental points of the photoelectron numbers demonstrates an almost negligible slope versus temperature of about 0.002\%/°C. The same points corrected for the relative QE exhibit somewhat larger instability of the light output, corresponding to the slope of about 0.01\%/°C, still negligible in comparison to the instability of the PMT gain.

As presented in Fig. 7, the high stability of the light output of LaBr\(_3\) is significantly better than that reported by Saint-Gobain [12] and measured with the PMT kept at a constant temperature. It could be an effect of the more dedicated experiment carried out in the present work, addressed to a narrower range of temperatures.

### 3.3. The LaCl\(_3\) crystal

The temperature dependence of the LaCl\(_3\) was measured with two peaking times of 2 and 12 \(\mu\)s. According to Saint-Gobain and earlier works [1,11], a longer shaping time in the spectroscopy amplifiers improves the thermal stability. For the measurements of the photoelectron number at 2 \(\mu\)s peaking time, the single-photoelectron peak positions determined in the LaBr\(_3\) tests were used (see Fig. 1). For the 12 \(\mu\)s peaking time, the single-photoelectron position was measured precisely at room temperature and then its ratio to that measured at 2 \(\mu\)s was calculated and further used as the correction factor for the whole range of temperatures. It was done to bypass a problem of a larger noise of the PMT at higher temperatures, introducing a distortion of the single-photoelectron spectra for 12 \(\mu\)s peaking time.

Fig. 8 presents the temperature dependences of the LaCl\(_3\) light at the output of the PMT for two tested peaking times. It is interesting that, contrary to the large instability of LaCl\(_3\) crystals reported by Saint-Gobain at short shaping time constants, the 662 keV peak position in the LaCl\(_3\), at the PMT output, showed a quite good stability of ±3\% in the temperature range between −30 and 50 °C. This indicates that the variation of the PMT gain, reflected in Fig. 2 by the position of the single-photoelectron peak, compensates the instability of the light output. It is not the case for the large peaking time, where the temperature variation of the PMT gain is the dominating factor.

Fig. 9 presents the number of photoelectrons measured for both peaking times versus temperature and those corrected for the relative QE. The data of the QE curve measured for 360 nm was used, as this wavelength is close to the peak emission of the LaCl\(_3\). The straight horizontal
line represents the fit of the linear function to the distribution of the experimental points at 12 μs peaking time.

The general trends of both curves follow those of the Saint-Gobain data [11]. However, the LaCl₃ crystal, at long peaking time, exhibits a much better stability of the number of photoelectrons with or without taking the relative QE into consideration. The latter quantity corresponds to about 0.01%/°C, as was observed for the LaBr₃ crystal (see Fig. 7). It is important to note that the photoelectron number measured at 2 μs peaking time approaches that at long peaking time for the temperatures above room temperature.

3.4. The NaI(Tl) crystal

The temperature dependence of the NaI(Tl) was measured using two peaking times of 2 and 12 μs. The following measurements of the light pulse shapes were carried out at different temperature (see Section 5.3) exhibiting a considerable contribution of slow components at low temperatures.

Fig. 10 presents the temperature dependence of the NaI(Tl) light at the output of the PMT for two selected peaking times.

The lower curve, measured at 2 μs peaking time, seems to follow the temperature dependence attributed to the NaI(Tl) crystal itself and presented in Ref. [9]. However, it was shown that at temperatures above 20 °C the decreasing PMT signal is mainly affected by the PMT gain instability (see Fig. 2). For a more precise discussion, the dependence of the number of photoelectrons should be analyzed.

The curve for the 12 μs peaking time follows approximately the linear reduction of the anode signal reported by Ianakiev et al. [8], however, in the temperature range above −10 °C. It suggests that the integration time of the light pulse of 12 μs peaking time used in the present studies is too short compared to that of the gated integrator in the Ianakiev et al. [8] experiment.

Fig. 11(a) presents the dependences of the number of photoelectrons on the temperature for both peaking times. The reference single-photoelectron peak positions measured during tests of LaBr₃ and shown in Fig 2 were used for the 2 μs peaking time, while for the 12 μs peaking time the procedure described above for the LaCl₃ was applied.

Fig. 11(b) shows the light output variation with temperature, expressed in relative units. It was obtained correcting the measured number of photoelectrons for the relative QE presented in Fig. 3. For the correction, the data from the test carried out with the LED at 420 nm were used, as that fitted well to the peak emission of the NaI(Tl). The correction for the PMT relative QE is of importance, particularly for temperatures above 0 °C, where the QE is increasing with temperature.

The curves corrected for the relative QE are, in fact, more important, as they reflect the dependence of the light output on temperature. The curve determined for the 2 μs peaking time follows well that presented in Ref. [9] and generally accepted in the literature as the dependence of the NaI(Tl) light output on temperature. However, the same curve measured at 12 μs differs significantly, as it does not exhibit the reduced light output at lower temperatures. No doubt, that in discussing the temperature dependency of the total light of NaI(Tl), one has to use the curve...
measured with a long peaking time (see the light pulse shape discussed in Section 5.3).

4. Thermal variation of energy resolution

The inspection of the peak drifts of the studied crystals versus temperature allowed simultaneous recording of the energy resolution for 662 keV γ-rays from a $^{137}$Cs source. The results of the study are discussed below, taking into account the contribution of the statistical uncertainty of the photoelectron number and that due to the intrinsic resolution of the scintillators.

The energy resolution, $\Delta E/E$, of the full-energy peak measured with a scintillator coupled to a PMT can be written \[^{[18]}\]

$$\left( \frac{\Delta E}{E} \right)^2 = (\delta_{sc})^2 + (\delta_p)^2 + (\delta_{st})^2,$$

(1)

where $\delta_{sc}$ is the intrinsic resolution of the scintillator, $\delta_p$ is the transfer resolution and $\delta_{st}$ is the PMT contribution to the resolution.

The statistical uncertainty of the signal from the PMT is described as

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

(2)

where $N$ is the number of photoelectrons and $\varepsilon$ is the variance of the electron multiplier gain, typically 0.1–0.2 for modern PMTs \[^{[19,20]}\].

The transfer component is described by the 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 dynode chain. The transfer component depends on the quality of the optical coupling of the scintillator to the PMT input window, the homogeneity of the QE of the photocathode and the efficiency of photoelectron collection at the first dynode. In modern scintillation detectors, the transfer component is negligible compared to the other components of the energy resolution \[^{[19,20]}\].

The intrinsic resolution of the scintillator is connected mainly with the non-proportional response of the scintillator \[^{[18–20]}\]. However, the experimentally determined intrinsic resolution is affected also by many effects such as inhomogeneities in the scintillator causing local variations in the light output and non-uniform reflectivity of the reflecting cover of the crystal.

4.1. The LaBr$_3$ crystal

Fig. 12 presents the dependence of energy resolution of the 662 keV peak versus temperature measured at 2 $\mu$s peaking time. The energy resolution was determined fitting a Gaussian curve to the measured peak.

The energy resolution is almost independent of temperature up to 20–30 °C and then it is increasing slightly with temperature. It differs from that given in the Saint-Gobain data sheet \[^{[12]}\] reporting an improvement of energy resolution by about 5% with a rising temperature in the same temperature range. The present measurement was done varying the temperature of the whole detector, including PMT. Thus, one cannot exclude that the energy resolution may be affected also by the PMT. It is better reflected in the rising intrinsic resolution at elevated temperatures (see bottom curve in Fig. 12). The intrinsic resolution of the crystal was calculated here by correcting the measured energy resolution for the contribution of the photoelectron statistic \[^{[19,20]}\], following Eqs. (1) and (2).

On the other hand, in the Saint Gobain experiment an 18" long light guide was used, which reduced the collected light on PMT photocathode and could affect energy resolution \[^{[21]}\].

4.2. The LaCl$_3$ crystal

Fig. 13 presents the dependence of energy resolution of the 662 keV peak on temperature, as measured with 2 and 12 $\mu$s peaking time with the XP2020. The full points present the measured energy resolution at 12 $\mu$s peaking time and the open points that measured for 2 $\mu$s peaking time.
12 µs peaking times. The energy resolution was determined fitting a Gaussian curve to the measured peaks. It showed a somewhat poorer energy resolution measured with the XP2020, in comparison to the previous tests of the same crystal coupled to the XP3212 PMT [3], where an energy resolution of 4.2% at room temperature was reported. It is probably an effect of a lower photoelectron number.

It is not clear that comparable or somewhat better energy resolutions are observed for 2 µs peaking time than those measured with 12 µs peaking time, while the photoelectron number was lower, particularly at temperatures below 20 °C (see Fig. 9). It is confirmed by the bottom curves in Fig. 13, which represent the intrinsic resolution of the crystal. The systematically larger contribution of the scintillator is observed for 12 µs peaking time, for the whole range of temperatures. It is in contrary to the previous measurements with other scintillators exhibiting slow components of light pulses [22,23,25] and coupled to avalanche photodiodes as photodetectors.

Since the measurements were incessantly running alternating the shaping time constant in the spectroscopy amplifier at each temperature, any systematical experimental errors should be excluded. Thus, the comparison of energy resolution, measured with 2 and 12 µs peaking times, was repeated for the same LaCl₃ and LaBr₃ crystals coupled to the same XP2020 PMT at room temperature, close to 20 °C. The results of the measurements confirmed the poorer energy resolution measured with long peaking time, observed also for the LaBr₃ crystal, which light pulse does not exhibit slow components [2,12]. It suggests that a certain distortion of the energy resolution is introduced by PMTs at long shaping time constants, not observed before with scintillators showing poorer energy resolution, as below for the NaI(Tl). Probably, special efforts are needed to improve a decoupling of the last dynodes in the voltage divider of the PMT, particularly working with the ORTEC 113 scintillation preamplifier. The estimated contribution of this effect is equal to 1.3 ± 0.3%, a mean value calculation based on the data presented in Fig. 13, and those found from several measurements carried out with the LaCl₃ and LaBr₃ crystals at room temperature. No doubt that it is a negligible contribution in the measurements with NaI(Tl).

For 2 µs peaking time a continuous improvement of energy resolution of LaCl₃ with increasing temperature seems to suggest a different mechanism affecting energy resolution at low temperatures, than that in the case of LaBr₃ (see Fig. 12). The postulated contribution of the PMT to the deterioration of LaBr₃ energy resolution at elevated temperatures is negligible in the present experiment. The intrinsic resolution of LaCl₃ (Fig. 13) showed almost the same slope as observed for the measured energy resolution. This suggests that it is the dominating effect responsible for the deterioration of the energy resolution at low temperatures.

4.3. The NaI(Tl) crystal

Fig. 14(a) presents the dependences of energy resolution on temperature in the NaI(Tl), as measured for the 662 keV γ-peak. It shows two separate curves for both peaking times. For 2 µs peaking time, the energy resolution is significantly distorted at low temperatures, below 20 °C. At the long shaping time the best energy resolution was measured around 0 °C.

It is well known that the energy resolution of the 662 keV peak in NaI(Tl) is mainly determined by the intrinsic resolution of the crystal [18,19]. Therefore, it is expected that the varying number of photoelectrons with temperature affects the energy resolution just slightly.

It is confirmed in Fig. 14(b), showing the intrinsic resolution determined for both peaking times, using Eq. (1). The measured energy resolution was corrected for the statistical contribution calculated from the number of photoelectrons, following Eq. (2). The contribution of the transfer resolution for the XP2020 PMT was neglected, according to Ref. [19]. Both curves presenting intrinsic resolution follow the temperature dependences of the measured energy resolution. The observed different curves for short and long peaking times and a variation of the intrinsic resolution with temperature can be correlated with
the two component decay of light pulses at low temperatures (see Section 5.3).

Earlier, a significant improvement of the energy resolution and particularly intrinsic resolution were reported for several crystals, as undoped NaI [22], CsI(Tl) [23,25,26] and ZnSe(Te) [24], obtained by the integration of the total light due to the two components of the light pulses by the long shaping in the spectroscopy amplifier.

Thus, the observed variation of the intrinsic resolution, presented in Fig. 15, seems to reflect a different contribution of the fast and slow components of the light pulses varying with the temperature and integrated differently by both peaking times. Again, it indicates a correlation between the above results and the study of the light pulse shape of NaI(Tl) versus temperature.

5. Thermal dependences of the light pulse shape

The observed different characteristics of light output and energy resolution of the crystals measured with different peaking times strongly suggested an investigation of the light pulse shape at different temperatures. The crystal, coupled to the Hamamatsu R5320 PMT, was inserted to the climatic exposure test chamber and was irradiated by γ-rays from a $^{137}$Cs source. The temperature was varied from −30 to 60 °C, as previously. The anode signal was recorded by a Tektronix Digital Oscilloscope type TDS5034B.

5.1. LaBr$_3$ crystal

Fig. 15 presents a set of the light pulses of LaBr$_3$ crystal as observed by the digital scope at different temperatures. It was also repeated using different ranges of the time base in the scope, up to 200 ns per division, for a more precise inspection of possible slow components. Note a background increases under the pulses at elevated temperatures reflecting a DC component of the dark current of the PMT. Moreover, afterpulses, delayed by about 150 ns, are seen on the pulses, with the intensity of about 1% in relation to the peak of the measured light pulse.

The observed decay times of the light pulses, seen in Fig. 15, are slightly varied with temperature, but they are also affected by a different contribution of the background. To quantify the decay time constant of the observed light pulses, a single exponential function on the constant background was fitted to the measured light pulses,

$$ y = A \exp(-t/\tau) + y_0, $$

where $\tau$ is the decay time constant of the observed light pulses.

Fig. 16 presents the dependence of the decay time constant on the temperature. The decay time constant is varying between $18.1 \pm 0.5$ ns at −30 °C and $22.0 \pm 0.7$ ns at 60 °C. Fits did not indicate significant slow components.

5.2. The LaCl$_3$ crystal

Fig. 17 presents the light pulse shapes from the LaCl$_3$ crystal measured at different temperatures. The measurement method was the same as described above for the LaBr$_3$ crystal. To quantify the decay curves, they were fitted with the sum of two exponential terms on the constant background,

$$ y = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2) + y_0, $$

where $\tau_1$ and $\tau_2$ are the decay time constants of the fast and slow components of the light pulses, respectively.

Fig. 17(a) shows a week variation of the primary decay time of the light pulse on temperature. It is presented better in Fig. 18(a) showing the decay time constant of the fast component versus temperature. The decay time constant is ranging between $21.9 \pm 0.7$ ns at −20 °C and $29.5 \pm 0.5$ ns at 60 °C, which agree well with those measured by Saint-Gobain [21]. It follows very qualitatively the growing light output measured at 2 µs peaking time.
Fig. 17(b) presents the selected light pulse shapes in the time range of 2 μs to enhance better the contribution of the slow component.

Fig. 18 summarizes the light pulse decay analysis. The upper panel shows the dependence of the fast component on the temperature, the middle one that of the slow component and the bottom panel exhibits the relative intensities of both the components. The presented data are characteristic for the LaCl₃ crystal doped with 10% of Ce, used by Saint-Gobain in the produced crystals.

5.3. The NaI(Tl) crystal

Fig. 19 presents the light pulse shapes from the NaI(Tl) crystal measured at different temperatures. The measurement method was the same as described above for the LaBr₃ and LaCl₃ crystals. To quantify the decay curves, they were fitted with the sum of two or three exponential terms on the constant background, as it was done for the LaCl₃ (see Eq. (4)). The fitting range did not include the initial slow decay observed at elevated temperatures.

Note a steady increase of the effective decay time with decreasing temperature as it was reported earlier in Ref. [7]. However, a more detailed examination of the light pulse decay at temperatures below 10 °C seems to suggest a different process of the light emission, reflected in the fast initial decay. It is seen well in Fig. 20(a) and (b), which present the selected light pulses at elevated and low temperatures, respectively. It shows clearly a different mechanism of the light emission at various temperatures. Again, a good agreement of the light pulse shapes at different temperatures with those measured at Saint-Gobain has to be pointed out [21].

First, a very well-known slow initial decay of the NaI(Tl) light pulse observed at room temperature [27], is replaced, at higher temperatures, by a delayed maximum suggesting a much slower energy transfer to the Tl emission centers. In contrast, the slow initial decay of the light pulses is not observed any more at temperatures below 0 °C and it is replaced by a much faster initial decay. Moreover, the intensity of the slow component is stronger, which is well correlated with the measured higher photoelectron number at low temperatures for the 12 μs peaking time.
Fig. 21(a) and (b) present the dependence of the decay time constants of the fast and slow components on temperature as determined in the present experiment. At low temperatures, below 0°C, another fastest component was found in the fit (see Fig. 21(a)).

The decay time constants of the fast and slow components are increasing with the reduced temperature, which was reported in the previous measurements of the effective decay time constant of NaI(Tl) [7].

The estimated intensities of all the three components are shown in Fig. 22. The intensity of the fast component at elevated temperatures was corrected for the initial slow slope of the light pulse. It is interesting that the intensity of the fast component drops down below 0°C temperature on behalf of an increasing intensity of the slow one. Moreover, simultaneously the third fastest component is observed at those temperatures with an increasing intensity at lower temperatures.

A different behavior of the decay time constant of the light pulses above and below room temperature was interpreted by Schweitzer and Ziehl [7] within a model based on a single temperature-dependent decay time constant, which describes the lifetime of the activator level [6]. An additional function, \( f(T) \), that represents the probability of occupation of the activation centers via only one process has been introduced to explain the non-linear behavior of the light output. However, according to Ianakiev et al. [8], the model does not take into account the second process of multi-phonon dissociation followed by binary diffusion to the activator centers.

The two-component light decay of NaI(Tl) was observed earlier in several papers [27–29]. However, because of a weak contribution of the slow component at room temperature it was not taken into account in general considerations.
Ianakiev et al. [8] assumes that the NaI(Tl) light pulse consists of two components with the decay time constants of 230 ns and 1 µs, respectively, and with a temperature-dependent redistribution between their intensities. It leads to about 95% intensity of the fast component at room temperature and only 60% at −20 °C.

In the case of the slow component, the Ianakiev assumption [8], that the decay time constant of about 1 µs is independent of temperature is contradictory to the results presented in Fig. 21.

6. Discussion and conclusions

The performed study confirmed an excellent thermal stability of the light output of LaBr3 over a wide temperature range and that of LaCl3, measured with a long shaping time constant. A similar effect was observed for NaI(Tl). 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. Both LaCl3 and NaI(Tl) crystals exhibit slow components, particularly observed at low temperatures. It suggests a temperature redistribution of the light decaying with different time constants, which affects the thermal stability at short peaking time. It is well observed in Fig. 22 for NaI(Tl) showing relative intensities of the fast and slow components versus temperature.

The same effect seems to affect the energy resolution measured at different temperatures. It was shown earlier that the energy resolution measured for the 662 keV γ-rays with a CsI(Tl) crystal is significantly improved when integrating the total light by a long shaping time constant up to 12 µs. Therefore, a full integration of the light from different scintillators, exhibiting two components of the decay of the light pulse, improves the thermal stability of energy resolution. This seems to be confirmed by the poorer intrinsic resolution of NaI(Tl) and LaCl3 when measured with a short peaking time at low temperatures and exhibiting an increased contribution of slow components at reduced temperatures.

LaBr3 is particularly important in this respect because of a single decay time constant of the light pulse and in fact a negligible variation of the light output and energy resolution with temperature.

The study of the decay of light pulses from LaCl3 and LaBr3 crystals confirmed earlier measurements, while NaI(Tl) showed a complex behavior at different temperatures. At low temperatures a strong contribution of a slow component with up to 60% of the total light was observed, while at an elevated temperature well-known initial slow decay was replaced by a delayed maximum and the slow component became insignificant. It suggests the need of a further study of the light emission mechanism in NaI(Tl) versus temperature.

The results of tests of the XP2020 PMT showed that the thermal variation of the gain of the dynode structure of about −0.4%/°C is the dominating effect. A positive instability of the QE was observed above 10 °C for longer wavelengths of 420 nm, thereby partly compensating the negative gain shift up to at least 50 °C.

Acknowledgments

This work was supported in part by the Polish Committee for Scientific Research, Grant no. 8 T10C 002 20. The Authors are very much indebted to Drs Esso Flyckt (ex-Photonis), Tadeusz Kozlowski (Soltan Institute) and Csaba Rozsa (Saint-Gobain) for a very valuable discussion on the paper.

References