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γ-ray performance of a 1242 cm^3 $LaCl_3$:Ce scintillation spectrometer

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Abstract

We present the results of γ-ray measurements on a large $4'' \times 6''$ $LaCl_3$:Ce crystal, characterized using radioactive sources over the range 14–3220 keV. The response of the crystal was found to be largely linear over the upper 90% of its dynamic range—the deviations at lower energies can be attributed to the shortcomings of the photomultiplier tube (PMT) and is in fact a generic problem when using high-gain PMTs with lanthanum halide scintillators. At 662 keV, a measured energy resolution of 4.1% FWHM was recorded at room temperature. In comparison to earlier generations of $LaCl_3$ detectors, the internal background spectrum shows significantly less X-ray, β, γ-ray and α contamination. The integral background count rate in the energy region 20 keV–3 MeV was determined to be 1.8 cm^{-3} s^{-1}, of which most (1.5 cm^{-3} s^{-1}) can be attributed to the β continuum from ^{138}La and only $\sim$0.04 cm^{-3} s^{-1} to the α complex from ^{227}Ac and daughters. Using these data, we have determined the light output of α particles relative to the equivalent energy electrons (or alternately γ-rays) to be, $\alpha/\gamma = 0.35 \pm 0.02$.

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Keywords: Lanthanum chloride; Scintillator detectors; γ-ray spectroscopy

1. Introduction

The recently discovered lanthanum halide (LaX_3:Ce) scintillators [1] could potentially revolutionize spectroscopic systems because of their excellent energy resolutions and proportionality of response when compared with traditional scintillating materials. This is especially true for those applications, where large-volume scintillators still provide the most sensitive means for γ-ray detection, such as remote sensing and homeland security. The compound is relatively easy to grow, since it melts congruently at 860 °C and, therefore, can be grown by a number of melt-based techniques, such as Bridgeman or Czochralski. However, until recently, the production of crystals with volumes above $\sim$50 cm^3 had been plagued by growth problems, largely related to cracking. This arises because the lanthanum halides are highly anisotropic materials with hexagonal structures. As such, the thermal conductivities and thermal expansion coefficients differ for the main crystallographic axes [2]. This leads to the introduction of very high stresses into the lattice during the cooling process. Fortunately, recent advances in crystal growth technology have now made large-volume lanthanum halide crystals commercially available [3]. In this paper, we report the results obtained with the first extremely large single-crystal $LaCl_3$ scintillation detector grown using the new technology.

2. Experimental

Ce-doped $LaCl_3$ scintillators have a fast light output decay (26 ns) and an emission spectrum with a maximum at

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350 nm. The light yield is typically 50,000 photons MeV^{-1} or 130% relative to NaI(Tl). The material density is similar to NaI at 3.8 $g\,cm^{-3}$. FWHM energy resolutions of 3.2% and 4.2% at 662 keV (^{137}Cs line) have been reported previously for smaller, ~1 and 12.8 cm^{-3} crystals, respectively [1,4].

The $LaCl_3$:Ce scintillator described here was grown from melt by Saint Gobain, processed and packaged in a hermetic Al can to prevent hydration. The cerium fraction is 10%. The crystal is a right-circular cylinder, nominally 4 in in diameter and 6 in high. The actual dimensions are 10.2 cm × 15.2 cm (1242 cm^3). To detect as much scintillation light as possible, the crystal is covered on all sides, except the read-out side, with a reflective material. The crystal is viewed through a quartz window coupled to a Photonis XP3540B, 10-stage 5 in photomultiplier tube (PMT). The PMT is enclosed in a μ-metal shield to minimize the effects of magnetic fields. The PMT base also contains a unity gain preamplifier whose output is shaped and amplified by an Ortec 671 spectroscopy amplifier. The output of the amplifier, in turn, is digitized by an Amptek 8000 A multichannel analyzer (MCA). The conversion gain of the ADC was set to 8192 channels. Detector bias is provided by a Canberra 3106D high-voltage supply unit. A PMT bias of 870 V and an amplifier shaping time of 0.5 μs were chosen so as to give an optimum spectral resolution of 4.1% keV FWHM at 662 keV and a dynamic range of 2 MeV. Spectral measurements were carried out using a set of calibrated γ-ray reference sources (^{241}Am, ^{133}Ba, ^{109}Cd, ^{57}Co, ^{60}Co, ^{137}Cs, ^{152}Eu, ^{203}Hg, ^{22}Na and ^{88}Y) placed coaxially 25 cm in front of the detector, along the principal forward detection axis. The aluminum can is 0.5 mm thick, which imposes a ~20 keV low-energy threshold (Fig. 1).

3. Results

Experience with smaller crystals, has shown that when using PMTs, there is a trade between dynamic range, linearity (proportionality of response) and resolution. This arises as follows. The very high photon yields of the scintillator coupled with its fast emission time, means that standard PMTs easily saturate when operated at nominal anode to cathode biases of ~1000 V—purely due to their very high gains. This can result in gross non-linearities being introduced into spectra. Reducing the PMT bias may reduce these non-linearities and extend the dynamic range, but at the expense of a deteriorated energy resolution. Therefore, a trade is required between dynamic range, linearity and resolution. This is illustrated in Fig. 2 in which we show the linearity curves (i.e., ADC channel versus line energy) for two detector biases: (a) optimized for a dynamic range appropriate for the remote sensing applications, i.e., the nuclear transition region (0–8 MeV) and (b) for a dynamic range appropriate for standard laboratory spectroscopy, i.e., the range covered by radioactive calibration sources (~0–2 MeV). The biases for these two dynamic ranges are 700 and 870 V, respectively. Two things are worth noting. The first is that 700 V is manufacturers minimum recommended bias, so biases less than this value are not viable, and secondly, for biases above 870 V, the light output from lanthanum halides saturate the PMT—so biases above this value are again not viable. From Fig. 2, we can clearly see that the larger deviations in linearity occur at the lower bias (i.e., larger dynamic range). In the inset, we have attempted to linearize the energy response by dividing by the energy. We have normalized both functions to unity above 1 MeV for comparison. From the graph, it is apparent that the

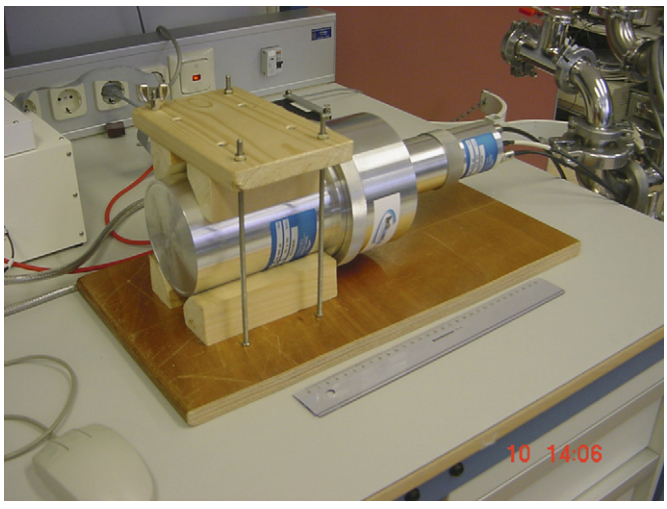

Fig. 1. Photograph of the 4″ × 6″ $LaCl_3$:Ce scintillation detector in its test jig.

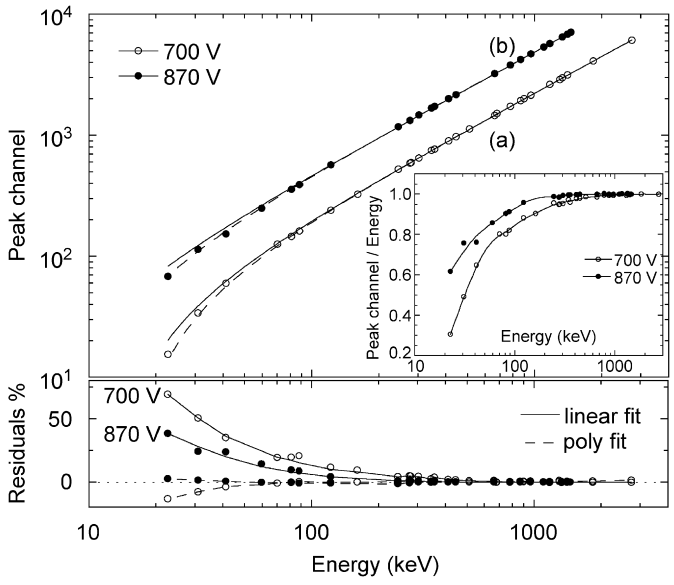

Fig. 2. The detector proportionality of response or linearity curves plotted for two biases: (a) optimized for remote sensing applications, i.e., a dynamic range of 8 MeV (V_b = 700 V) and (b) optimized to 2 MeV for general laboratory operation with standard radiation calibration sources (V_b = 870 V). The panel below shows the corresponding residuals, i.e., the percentage deviation from a best fit (measured–calculated /calculated × 100). Two cases are given. The first is for a straightforward linear fit (solid line) and the second for a quadratic polynomial fit (dashed line).