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Effects of packaging $SrI_2(Eu)$ scintillator crystals

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ABSTRACT

Recent renewed emphasis placed on gamma-ray detectors for national security purposes has motivated researchers to identify and develop new scintillator materials capable of high energy resolution and growable to large sizes. We have discovered that $SrI_2(Eu)$ has many desirable properties for gamma-ray detection and spectroscopy, including high light yield of $\sim$90,000 photons/MeV and excellent light yield proportionality. We have measured $<2.7\%$ FWHM at 662 keV with small detectors (<1 cm^3) in direct contact with a photomultiplier tube, and $\sim$3% resolution at 662 keV is obtained for 1 in.^{3} crystals. Due to the hygroscopic nature of $SrI_2(Eu)$, similar to NaI(Tl), proper packaging is required for field use. This work describes a systematic study performed to determine the key factors in the packaging process to optimize performance. These factors include proper polishing of the surface, the geometry of the crystal, reflector materials and windows. A technique based on use of a collimated ^{137}Cs source was developed to examine light collection uniformity. Employing this technique, we found that when the crystal is packaged properly, the variation in the pulse height at 662 keV from events near the bottom of the crystal compared to those near the top of the crystal could be reduced to $<1\%$. This paper describes the design and engineering of our detector package in order to improve energy resolution of 1 in.^{3}-scale $SrI_2(Eu)$ crystals.

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1. Introduction

There has been a lot of excitement in the gamma-ray detection community about new scintillator materials that are starting to compete with some of the best room temperature semiconductor materials. One of these is $SrI_2(Eu)$, which offers an effective Z slightly better than that of $LaBr_3(Ce)$, a high light yield of $\sim$90,000 photons/MeV, and excellent light yield proportionality, surpassing that of $LaBr_3(Ce)$[1–4]. We routinely obtain spectroscopic performance with this material of $<2.8\%$ FWHM at 662 keV. Like many other halide materials, $SrI_2(Eu)$ is hygroscopic. It will decompose in air, but is stable indefinitely when stored in an environment with low water content, such as anhydrous mineral oil. We have observed that when left in ambient conditions, $SrI_2(Eu)$ develops a whitish yellow layer on its surface that is optically absorptive and thus degrades light collection efficiency. Thus, it is important to ensure that the surface is free of this layer using optical polishing and packaging to achieve the best possible light collection.

For field use, $SrI_2(Eu)$ requires a hermetic enclosure. Although packaging of hygroscopic scintillators such as NaI(Tl) is not new, we took the opportunity to explore certain aspects such as crystal geometry and modern Teflon-based reflectors in our packaging design. In this paper we will describe factors that we found to be most important in achieving good energy resolution with packaged $SrI_2(Eu)$ detectors.

2. Study of crystal surface polish and geometry

An important aspect of achieving good energy resolution with a scintillator detector is careful consideration of the optical properties of the crystal. The high light yield of $SrI_2(Eu)$ and excellent light yield proportionality suggest that an energy resolution on the order of 2.3% at 662 keV may be attainable. However, this assumes homogeneous crystal response and uniform light collection, independent of where in the crystal the scintillation photons are produced. There are a few optical effects which impact the uniformity of light collection: (1) optical absorption at the surface, which is a loss mechanism, (2) absorption of light due to impurities in the bulk of the crystal, another loss mechanism, and (3) optical absorption and re-emission in the bulk by Eu^{2+}, known as

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light-trapping, which has no deleterious effect alone, other than lengthening the decay time. The optical absorption at the surface, thought to be due to entrained iodine which can result from even tiny amounts of decomposition, results in non-uniform light collection efficiency and therefore resolution degradation. Additionally, depending on the purity of the crystal growth feedstock, absorptive impurities such as trace amounts of graphitic carbon can degrade uniformity via non-uniform bulk absorption. Finally, we have found that light trapping within the crystal, coupled with optical absorption at the surface, leads to nonuniformity as a function of the variation in optical pathlengths. Therefore, to achieve the best energy resolution possible, the surfaces must be clean, the crystal growth feedstock must be high purity, and the average photon pathlength should be reduced, by use of the best geometries and optical design of the scintillator detector.

The best surface finish we have achieved so far is by coating the crystal with mineral oil and polishing it with a polishing cloth and 3 μm polycrystalline diamond. Fig. 1 shows an example of a gamma ray spectrum obtained from a ~1 in.^{3} crystal after polishing. This was obtained with a Radiation Monitoring Devices (RMD) SrI_2(5% Eu) crystal. The high energy tailing we observed remained a mystery for some time until the technique described in the next section was developed to examine this effect.

2.1. Collimation study to investigate light collection nonuniformity

In order to investigate high energy tailing observed in some large SrI_2(Eu) crystals, a technique was developed to examine the spatial response of a crystal coupled to a PMT. In this experiment, a Hamamatsu R6231-100 super bialkali PMT was employed. We used 2 lead bricks to form a slit for collimating ^{137}Cs gamma-rays.

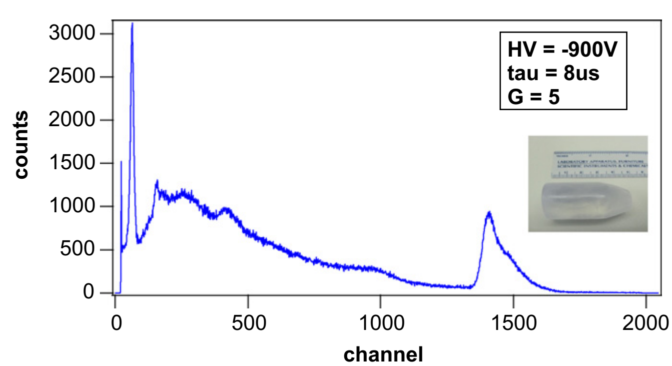

Fig. 1. ^{137}Cs spectrum obtained with an RMD detector doped with 5% Eu (vol. = 19.7 cm^3). Reduced high energy tailing and improved light collection was observed after polishing.

Then, a crystal was scanned along the *z*-axis (see Fig. 2a). Pulse height spectra taken at each of these planar positions, acquired with polished detector 52, are shown in Fig. 2b. The data shows a clear trend of increasing photopeak position, and thus increased light collection, as the irradiation distance from the PMT window (*z*) decreases. The overall peak shift between position 1 and position 4 is ~10%. This trend is believed to be caused by a combination of light trapping in the crystal, as well as light losses at the surfaces of the crystal. Another trend we typically observe is poorer resolution as the gamma interaction occurs closer to the PMT window. Although the exact cause of this is under investigation, we believe this to be due to greater path length variation that the light travels in the crystal, the closer the event occurs to the PMT window. Light emitted isotropically near the bottom of the crystal will result in about half of the light directed toward the PMT and the other half directed away. If the light that travels away from the PMT gets reflected at the back plane of the crystal, then it travels at least 2 × the length of the crystal, resulting in higher probability of interaction with the surface and bulk impurities, which may exhibit some optical absorption. However, the other half of the light directed toward the PMT will travel very little in the crystal, and thus will have low light loss due to surface and bulk interactions. The broadening of the distribution of average photon pathlengths is expected to broaden the photopeak, and this effect is expected to be less pronounced the further away the event occurs from the PMT window. The combination of these effects gives us a good understanding of the high energy tailing observed in Fig. 1, which can be generally described as light collection nonuniformity.

2.2. Detector geometry study

It has been well established that the geometry of a scintillator detector affects light collection efficiency at the PMT photocathode. In general, it has been determined that a tapered cylinder is most favorable for efficient collection of scintillation photons [5]. Thus, it was decided to cut and taper the detector tested in Fig. 1. A pulse-height spectrum with 662 keV excitation of this newly shaped crystal along with its dimensions is displayed in Fig. 3. The high energy tailing we saw previously in the larger crystal (Fig. 1) was greatly reduced and the resolution was improved to 4.49% at 662 keV. We also observed substantially better light collection, such that light collection efficiency increased by a factor of 2.52.

To characterize the potential of SrI_2(Eu), we selected a 5% Eu doped SrI_2(Eu) crystal, grown by RMD, that exhibited good light yield as a large boule, and cut a small ~1 cm^3 cone from it. A spectrum demonstrating 2.68% energy resolution FWHM at 662 keV acquired with this crystal is shown in Fig. 4. We estimated the light yield of this detector to be 103,000 photons/MeV. So far,

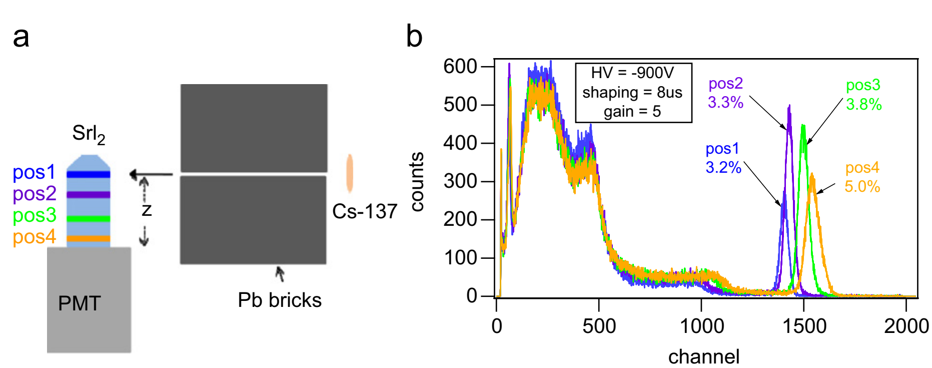

Fig. 2. Source collimation experiment, where (a) provides a schematic of the experimental setup. (b) Pulse height spectra acquired with the same RMD detector as tested previously. We see a clear trend in which the photopeak centroid position increases with decreasing scan distance from the PMT window, while the resolution (percent value indicated below position) degrades. The peak shift between position 1 and position 4 is ~10%.