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Performance of large lanthanum bromide scintillators

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Abstract

Cerium-doped lanthanum bromide, $LaBr_3$(Ce), scintillator possesses several outstanding properties that make it an attractive choice for security, medical, and geophysical radiation detection applications. Among these properties are good density (5.1 g/cc), excellent energy resolution (~3% full-width at half-maximum (FWHM) at $E_\gamma = 662$ keV), brightness (>65,000 photon/MeV), and speed, (τ_d<20 ns). Unfortunately, the development of many prospective devices using this scintillator has been hampered by the lack of large crystals (>100 cc). The anisotropic thermal expansion exhibited by this material makes it difficult to grow large ingots due to the build up of internal stresses as it cools, causing fracturing. Recently, Saint–Gobain Crystals has achieved successful growths of large unfractured ingots, from which large detectors have been assembled (>150 cc). The outstanding properties seen in small pieces are retained up to at least 155 cc (the largest assembled into a single detector thus far). A cylindrical $LaBr_3$(Ce = 5%) crystal with dimensions of diameter = 51 mm, and length = 76 mm achieves energy resolution of 3.1% FWHM at 662 keV, and brightness of 165% of NaI with good uniformity throughout the crystal. Scintillation light yield and energy resolution have been examined as a function of crystal size and γ-ray energy. Spatial mapping of a large crystal was examined and shown to be uniform. Large crystals enable accurate measurements of the intrinsic γ-ray background from ^{138}La (0.09% nat. abun., γ-ray emission at 1436 and 789 keV). This background is shown to scale appropriately in size with theoretical calculations.
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1. Introduction

Cerium-doped lanthanum bromide scintillator, $LaBr_3$:Ce, has some remarkable properties that qualify it as a superior choice for some applications in the security, medical imaging, and geophysics fields. Notably, it has a very high scintillation light yield (>65,000 photons/MeV), fast emission (decay time, τ_d, ~16 ns), good density (5.1 g/cc), and the best energy resolution among scintillators (~3% FWHM at 662 keV). Since its invention by Delft and Bern Universities in 2001 [1], the crystal has only been available in small sizes. The reason for this has been primarily due to the crystal's strong anisotropic properties. Since it exhibits anisotropy in thermal expansion, heat transfer, and mechanical strength [2], large ingots are prone to cracking during the cool down after growth. Large single crystals are necessary in applications that wish to take advantage of $LaBr_3$'s excellent energy resolution, yet be efficient at capturing γ-rays. Recently, Saint–Gobain Crystals has developed a growth process that regularly succeeds in producing large uncracked ingots of this material. Saint–Gobain markets $LaBr_3$:5%Ce under the trade name BrilLanCe 380®. All further references to this material in this report will assume the 5% Ce composition. A concern exists that large crystals may have degraded performance in comparison to smaller ones due to material non-uniformity [3] or optical self–absorption of the emitted scintillation light. The purpose of this report will be to present the most recent data comparing the performance of small crystals to the newly available large ones. The parameters investigated are overall light output and energy resolution, energy linearity, and volumetric uniformity. Another quantity under investigation is the crystal's response to the decay of

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^{138}La, which has a natural abundance of 0.090%. The larger the crystal, the greater the background of the characteristic γ-rays at 1436 and 789 keV. Large crystals present an opportunity to quantify this background and compare it to expected values.

2. Progress in growth of large uncracked ingots

Uncracked, single-piece ingots have achieved the size of 130 mm diameter by 105 mm long. The largest crystal cut and tested from such ingots has been a cylinder 51 × 76 mm (diameter × length). However, nothing precludes making a detector from the full size of the ingot, and plans are to do that in the near future. Fig. 1 shows Saint–Gobain's progress in growing ever-larger crystals by plotting the largest unfractured crystal capability versus date (calendar year). Note the dramatic upswing in the graph coming at the close of 2005. At this time, new furnaces came on-line, which were designed to reduce the internal mechanical stresses induced by the anisotropy during growth. The current furnace design is capable of being scaled up to grow 200 mm diameter ingots.

3. Light output and energy resolution as a function of size

For this report, 37 crystals of the sizes listed in the Table 1 were tested for relative light output and energy resolution. Cylinder dimensions are listed as diameter × length. Where available, several crystals of the same size, but from different growths were tested.

3.1. Test system configuration

The crystals listed in Table 1 were wrapped with Teflon reflector and hermetically sealed in aluminum or titanium housings with sapphire windows on one end. One face of each crystal was coupled to the window with clear silicone rubber. For the cuboidal crystals one of the 4 × 4 mm faces was coupled to the window, and one of the circular faces was coupled for the cylindrical crystals. This package was then optically coupled to a photomultiplier tube (PMT) and multichannel analyzer (MCA) system. The PMT used was a Hamamatsu R1306 (8 stage) operated at +800 V with a modified voltage divider scheme. It was modified in a similar fashion to [4], where the signal was picked off at the sixth dynode with extra capacitance added to the penultimate dynode. This was done because $LaBr_3$:Ce puts out a very large number of photons in a very short period of time, which produces very high instantaneous currents in the PMT. This can momentarily cause the voltage to droop between dynodes, leading to non-linearities in energy response, especially at high γ-ray energies (>1 MeV). With the modified voltage divider, the current is extracted before reaching full gain and capacitance is added to provide needed voltage support. The PMT output was fed to a fast preamplifier and then to the MCA (Aptek model S5008, bi-polar shaping, 1 μs shaping time, 11-bit digitization).

Table 1
Sizes of $LaBr_3$:Ce crystal tested

Dimension (mm)	Shape	Volume	Quantity
4 × 4 × 5	Cuboid	0.08	6
4 × 4 × 15	Cuboid	0.24	4
4 × 4 × 30	Cuboid	0.48	12
13 × 13	Cylinder	1.7	4
19 × 19	Cylinder	5.4	3
25 × 25	Cylinder	12.3	4
38 × 38	Cylinder	43.1	2
41 × 76	Cylinder	100	1
51 × 76	Cylinder	155	1

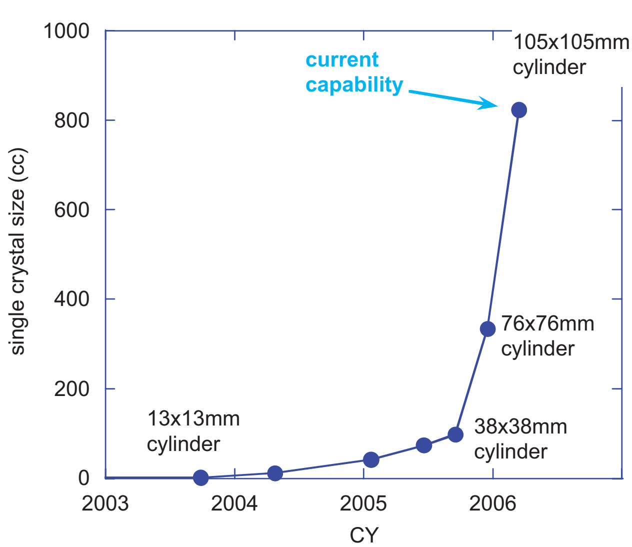

Fig. 1. Progress in growing large $LaBr_3$ crystals.

3.2. Light output variation with size

Light output and energy resolution for the 662 keV γ-ray photopeak are shown in Fig. 2a. The γ-ray source was placed for end-on illumination. The symbols show the mean of quantities listed in Table 1, and the error bars denote the minimum and maximum of the crystals tested. Note that although there is some variance, there is no noticeable degradation with increasing size among crystals with similar aspect ratios. The normalized light output of 1 is approximately equal to 165% of the light output of NaI, or about 65,000 photons/MeV. One may note that the longer cuboids (i.e. pixels) are worse performers than shorter ones. Most of this difference can be attributed to light loss at the reflector boundary. Simulations with the DETECT97 optical transport software [5,6] predict 11% less photons escaping a 4 × 4 × 30 mm pixel versus a 4 × 4 × 5 mm pixel. This is due to the average photon requiring more interactions with the (imperfect) reflector in the longer crystals before it

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