Design of a Lanthanum Bromide Detector for Time-of-Flight PET

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Abstract—Recent improvements in the growth and packaging of lanthanum bromide (LaBr₃), in addition to its superb intrinsic properties of high light output, excellent energy resolution, and fast decay time, make it a viable detection material for a positron emission tomography (PET) scanner based on time-of-flight (TOF). We have utilized theoretical simulations and experimental measurements to investigate the design and performance of pixelated LaBr₃ Anger-logic detectors suitable for use in a TOF PET scanner. Our results indicate that excellent energy resolution can be obtained from individual as well as multicrystal arrays of LaBr₃ in a 4 mm \times 4 mm \times 30 mm geometry. Measured energy resolutions (at 511 keV) of 4.1% for a single crystal and an average of 5.1% for an array of 100 crystals have been achieved with our best samples. Both simulations and experimental measurements of an Anger-logic based detector consisting of the LaBr₃ crystal array coupled to a continuous light guide and seven photomultiplier tubes (PMTs), have resulted in the ability to clearly discriminate 511 keV interactions in each crystal. We have measured coincidence time resolutions for both 0.5% and 5.0% cerium-doped LaBr₃ and found that the higher level of Ce-doping yielded superior results with little to no degradation in light output or energy resolution. The time resolution for a single 5.0% Ce-doped LaBr₃ crystal (4 mm \times 4 mm \times 30 mm) coupled directly to a PMT was measured to be 275 ps full-width at half-maximum (FWHM). With an array of 100 crystals coupled to a light guide and seven PMT cluster an average time resolution of 290 ps FWHM was obtained by summing the signals from the PMT cluster. Ultimately, two 5.0% Ce-doped LaBr₃ Anger-logic detectors placed in coincidence yielded a time resolution of 313 ps

Index Terms—Anger-logic, gamma-ray detectors, lanthanum detectors, positron emission tomography, scintillation detectors, 3-D PET, time resolution, time-of-flight.

I. INTRODUCTION

THE incorporation of time-of-flight (TOF) has long been recognized for its potential to improve positron emission tomography (PET) scanner performance [1]–[3]. One of the major challenges in TOF PET is finding an ideal scintillator.

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In recent years, there has been resurgence in the search for an ideal scintillator (i.e., one having high light output, high stopping power, fast decay time, and good linearity) [4]–[6]. Out of the considerable research and development of inorganic scintillators [7]–[12], the possibility for TOF PET has recently been proposed for both LSO [13] and lanthanum scintillators [14].

Of particular interest to us is the lanthanum bromide (LaBr $_3$) scintillator. The high light output (\sim 61 000 photons per MeV), fast decay time (\sim 35 ns), and excellent energy resolution (2.8% at 662 keV) of LaBr $_3$ [11] make it a viable detection material for a PET scanner based on TOF. The high light output allows the use of long narrow crystals (i.e., 4 mm \times 4 mm \times 30 mm), thus maintaining high sensitivity and spatial resolution in a scanner. Excellent energy resolution allows for improved rejection of scattered events and reduction in random coincidences, which are limiting factors of image quality in three-dimensional (3-D) PET. The fast decay and excellent coincidence time resolution will reduce dead time as well as random coincidences. Here, we will evaluate the design and performance of a pixelated LaBr $_3$ detector suitable for use in a whole-body 3-D PET scanner incorporating TOF.

II. DETECTOR DESIGN

The proposed LaBr $_3$ detector design is based on previous designs of pixelated GSO and NaI(Tl) detectors used in 3-D PET [15], [16]. The detector consists of an array of individual 4 mm \times 4 mm \times 30 mm LaBr $_3$ crystals (hermetically sealed and packed in a reflective powder) coupled to a continuous light guide and array of photomultiplier tubes (PMTs). Fig. 1 illustrates the arrangement of the pixelated Anger-logic detector.

The scintillation light from the crystals is distributed to multiple PMTs, via the light guide, to allow the photon interaction position and energy to be determined. In this arrangement, the PMTs are packed in a hexagonal array and the light guide thickness is chosen such that the light spread from a crystal positioned directly above the PMT center is limited to a cluster of seven PMTs. The crystal dimensions are chosen to maintain good spatial resolution and detection efficiency in a potential 3-D PET scanner. A singles detection efficiency of $\sim\!51\%$ is obtained with 30 mm thick LaBr₃ ($\mu=0.47~\rm cm^{-1})$ for events with energies above 400 keV.

III. MEASUREMENTS WITH SINGLE LaBr₃ CRYSTALS

Several single crystal samples of LaBr $_3$ were evaluated in order to characterize energy resolution, coincidence time resolution and light output in a 4 mm \times 4 mm \times 30 mm geometry

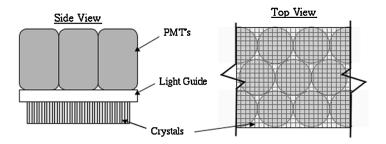


Fig. 1. Two views of a pixelated Anger-logic detector illustrating the arrangement of the PMTs, light guide, and scintillator crystals. Note the hexagonal PMT arrangement in the top view.

as well as differences in varying cerium concentration. LaBr₃ samples (each encapsulated and packed in reflective powder) were obtained from Radiation Monitoring Devices, Inc. (RMD) and Saint-Gobain Crystals and Detectors (Saint-Gobain). Three small LaBr₃ samples with Ce concentrations of 0.5%, 5.0%, and 10% were obtained from RMD with varying geometries (each less than $\sim 1~{\rm cm}^3$ in volume). The two samples obtained from Saint-Gobain were each polished and in a 4 mm \times 4 mm \times 30 mm geometry with Ce concentrations of 0.5% and 5.0%.

Each of the Saint-Gobain samples were tested with the 4 mm \times 4 mm cross section optically coupled to the center of the PMT photocathode. Fig. 2(a) and (b) show measured pulse shapes obtained by digitizing the PMT anode output for 511 keV energy deposition in four LaBr $_3$ samples and one LYSO sample included for comparison.

The pulse shapes indicate that both rise and decay times improve (i.e., reduce) with increasing Ce concentration while the total light output measured for the 0.5% and 5.0% Saint-Gobain crystals remains the same. Additionally, the measured light output for the 4 mm \times 4 mm \times 30 mm LaBr $_3$ crystals was approximately twice that of a 4 mm \times 6 mm \times 25 mm LYSO crystal, with the 5.0% Ce crystal having faster rise and decay times. The improvement is more significant from 0.5% to 5.0% Ce [see Fig. 2(b)] than 5.0% to 10.0% Ce [see Fig. 2(a)]. Our simulations indicate that the difference in sample geometry largely accounts for the 1/3 reduction in light output between the small (<1 cm 3) samples and 4 mm \times 4 mm \times 30 mm samples (see Table I).

The energy resolution of each sample was measured with the crystals optically coupled to the center of the PMT photocathode and excited by 511 keV photons from a ⁶⁸Ge source. The signals from the PMT anode were input directly into a charge integrating analog-to-digital converter (ADC) with an integration time of 120 ns. Fig. 3 shows the energy spectrum generated for the Saint-Gobain 5.0% Ce LaBr₃ sample. The energy spectrum was calibrated by exciting the crystal with 662 keV photons from a ¹³⁷Cs source.

Additionally, the coincidence time resolution for each $LaBr_3$ sample was measured. This measurement was accomplished by irradiating a BC-418 plastic scintillator (coupled to a XP2020Q PMT) and each $LaBr_3$ sample coupled to a 50 mm diameter XP20Y0 PMT (a fast, eight-stage version of the XP2020Q without a quartz window) with 511 keV positron annihilation photon pairs from a 68 Ge source. The signal from the plastic-PMT detector was processed by an Ortec 934 CFD

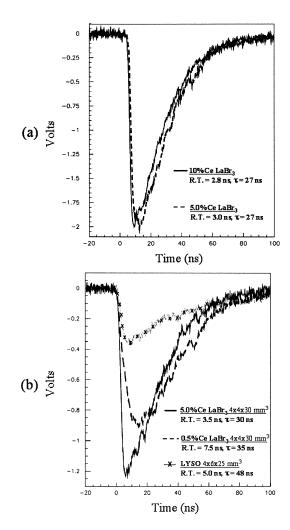


Fig. 2. Single pulse shapes measured at 511 keV for (a) two RMD LaBr₃ samples (10% and 5.0% Ce, each \sim 1 cm³ in volume) and (b) two Saint-Gobain samples (5.0% and 0.5% Ce, each 4 mm \times 4 mm \times 30 mm) and one LYSO sample of similar geometry for comparison. The 10–90 rise time (RT) and decay time (τ) are given for each pulse. The pulse shapes were measured with the crystals directly coupled to a Photonis XP2020Q PMT (tube rise time \sim 1.5 ns).

and formed the "start" channel of the timing circuit. The LaBr₃-PMT detector was processed by a LeCroy 825 Z rise time–compensated discriminator and delay forming the "stop" channel. The time difference was digitized with a time-to-digital converter (TDC) having 45 ps per bin resolution. Fig. 4 shows the coincidence timing resolution spectrum obtained for the Saint-Gobain 5.0% Ce LaBr₃ crystal.

LaBr ₃ Manufacturer (size)	%Ce	ΔΕ/Ε (at 511 keV)	Time Resolution (FWHM)	Rise Time	Decay Time	Relative Light Output
RMD Inc. (<1 cm³)	0.5	3.5%		7.0 ns	33 ns	1.50
RMD Inc. (<1 cm ³)	5.0	3.7%	250 ps	3.0 ns	27 ns	1.56
RMD Inc. (<1 cm ³)	10.0	4.7%	245 ps	2.8 ns	27 ns	1.46
Saint-Gobain (4x4x30 mm³)	0.5	4.1%	350 ps	7.5 ns	35 ns	1.00
Saint-Gobain (4x4x30 mm³)	5.0	4.5%	275 ps	3.5 ns	30 ns	1.00

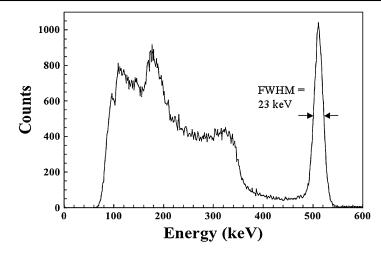


Fig. 3. Energy spectrum measured at 511 keV with the Saint-Gobain 5.0% Ce LaBr₃ (4 mm \times 4 mm \times 30 mm) crystal optically coupled to the center of the PMT photocathode. For this pixel, an energy resolution of 23 keV FWHM (Δ E/E = 4.5%) was obtained by fitting the photopeak with a Gaussian distribution.

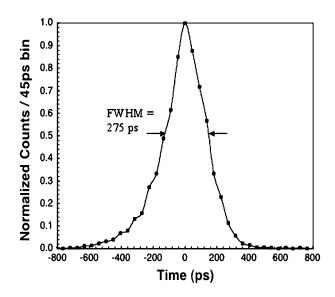


Fig. 4. Timing resolution spectrum measured with the Saint-Gobain 5.0% Ce LaBr $_3$ (4 mm \times 4 mm \times 30 mm) crystal optically coupled to the center of a XP20Y0 PMT photocathode in coincidence with a BC-418 plastic scintillator coupled to a XP2020Q. A time resolution of 275 ps FWHM was obtained. A time resolution of 235 ps FWHM was measured for two BC-418 plastic scintillators in coincidence.

Results from the energy and time resolution measurements for each of the samples are shown in Table I (values for energy resolution $\pm 0.2\%$ and time resolution ± 5 ps). Additionally, the rise and decay time as well as the samples relative light output (normalized to the Saint-Gobain 0.5% Ce sample) are listed in the table. Excellent energy resolution is measured for both

small RMD samples and the full size Saint-Gobain crystals. In general, samples with higher Ce concentration (having faster rise and decay times with approximately equal light output) had better time resolution with a greater improvement between the 0.5% and 5.0% Ce (4 mm × 4 mm × 30 mm) samples as compared the 5.0% and 10.0% Ce samples. The time resolution measured for the 5.0% Ce sample from RMD is consistent with previous measurements reported by Shah *et al.* [17]. Degradation in time resolution from 250 ps to 275 ps FWHM is measured between the small 5.0% Ce RMD sample and the 5.0% Ce Saint-Gobain sample. This degradation is consistent with findings based on different sample geometries in other fast scintillators [13], [18].

IV. MEASUREMENTS WITH A LaBr₃ ANGER-LOGIC DETECTOR

A. Methods

We have received several multicrystal $(10 \times 10 \text{ and } 5 \times 5)$ arrays of 0.5% and 5.0% Ce LaBr₃ from Saint-Gobain. Each array consisted of individually cut and polished 4 mm \times 4 mm \times 30 mm crystals (with 0.3 mm spacing) optically coupled to a 3 mm glass window, packed in a reflective powder and hermetically sealed in an aluminum housing.

In order to test energy resolution, timing resolution, and relative light output of each crystal in the array as well as the ability to discriminate 511 keV events in each crystal with a 7 PMT Anger-logic detector, a data acquisition system utilizing both a TDC and ADC was implemented. Fig. 5 illustrates a block diagram layout of the acquisition system.

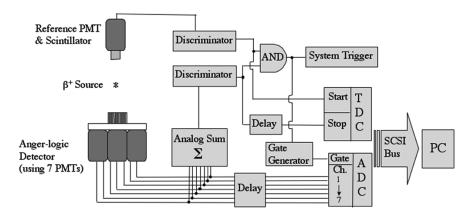


Fig. 5. Block diagram showing the electronics architecture used to acquire data. A coarse coincidence gate of 30 ns (set in the AND logic unit) is used as the system trigger. Following a triggered event, the amount of charge integrated in each ADC channel and the time difference recorded by the TDC are processed via PC software. Processing included correction of ADC offsets and PMT gains, implementation of a weighted centroid algorithm to determine the interaction location, and position and energy gating to generate time and energy spectra for individual crystals.

Each crystal array was tested in an Anger-logic arrangement consisting of the LaBr₃ crystal array optically coupled to a 1.6 cm thick light guide (UVT Lucite) and cluster of seven PMTs (hexagonally arranged). While both 39 mm and 50 mm PMTs were considered in the detector design, the measurements presented hereafter represent the results obtained with a 7 PMT cluster of 50 mm diameter Photonis XP20Y0 PMTs. It was found that these PMTs provided coincidence time resolution performance close to that of the XP2020Q at a much reduced cost.

B. Results

Two-dimensional flood maps of photon interaction positions in the crystal array were generated by implementing a weighted centroid algorithm on the ADC data obtained from each of the seven PMTs following events from a 511 keV source. For crystal identification purposes this was done without requiring the coincidence with the reference detector shown in Fig. 5. Fig. 6(a) shows the two-dimensional (2-D) flood map for events having a total energy >450 keV in a 5.0% Ce LaBr₃ array with 100 crystals. Fig. 6(b) shows a histogram for a center row of crystals in the 2-D flood. The crystal discrimination for this array was typical of all the crystal arrays received.

Coincidence time resolution measurements with the Anger-logic detector were made in a similar fashion to those described in Section III (i.e., with the plastic-PMT detector forming the "start" and LaBr₃ detector forming the "stop"). Here, however, the seven PMT signals were summed before being processed by the discriminator (see Fig. 5). We found that summing the seven PMT signals before processing in the discriminator yielded better time resolution than processing a single PMT with the largest pulse (i.e., the central PMT). This is illustrated in Fig. 7, showing the time resolution measured with a single 5.0% Ce LaBr₃ (4 mm \times 4 mm \times 30 mm) crystal in three different arrangements. In position 1 [shown for reference, see Fig. 7(a)] the crystal is directly coupled to the PMT and a time resolution of 275 ps FWHM is measured (as reported in Section III). With the crystal coupled to a light guide and positioned above the center of a seven PMT cluster [position 2, Fig. 7(a)] the time resolution degrades to 350 ps FWHM when

using only the signal from the central PMT in the timing circuit. The degradation is largely due to the fact that only a fraction of the total light out of the crystal is processed by any single PMT in the cluster (i.e., about 70% of the total light is incident upon the central PMT with the crystal in position 2). However, the time resolution is improved to 300 ps FWHM when all seven PMT signals are summed. This improvement is more dramatic (465 ps to 310 ps) when the crystal is positioned above the edge of the central PMT (position 3). The difference in time resolution measured for the crystal coupled directly to the PMT and in the Anger-logic arrangement (275 ps compared to 300 ps) is primarily due to light loss in the Anger-logic arrangement (i.e., coupling interfaces, gaps between PMTs, etc.) and differences in PMT responses (i.e., transit times, transit time spreads, etc.). While it is recognized that the coincidence time resolution may be improved by correcting for PMT transit time differences, the measurements presented here do not incorporate a correction.

In order to obtain coincidence time resolution for crystals in the arrays, position, and energy gates were implemented in software prior to generating a histogram of time differences between the reference detector (plastic-XP2020Q) and the LaBr₃ Anger-logic detector. The location of the position gate for each crystal was determined from the 2-D flood map [see Fig. 6(a)]. Additionally, an energy gate was placed on the events in a crystal such that the total energy deposited in the crystal was > 450 keV. Fig. 8 shows the coincidence time resolution measured for crystals in the 0.5% and 5.0% Ce LaBr₃ arrays as a function of crystal position (in the X- and Y-direction) in the Anger-logic detector. Consistent time resolution results are obtained for both crystal arrays with a \sim 170 ps improvement measured for the 5.0% Ce array. An average coincidence time resolution of 308±18 ps is measured for all the crystals in the 5.0% Ce array with little variation in resolution over the center region of the seven PMT cluster. The average time resolution for crystals in each of the arrays is given in Table II.

The energy resolution and relative light output were measured for each crystal in the arrays by similarly applying position gating. Excellent energy resolution was obtained for crystals in 0.5% and 5.0% Ce arrays with little degradation from the measurements with single crystals coupled directly to a PMT.

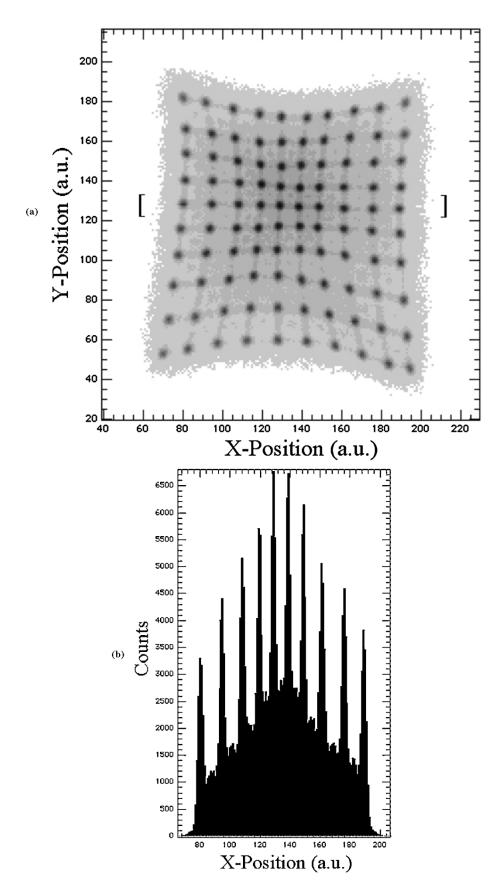


Fig. 6. (a) 2-D flood map of interaction positions for events >450 keV in a 5.0% Ce LaBr₃ array with 100 crystals coupled to a 1.6 cm light guide and cluster of 7 XP20Y0 PMTs. (b) Histogram of events for the fifth row of crystals from the top summed over 4 mm in the Y-direction [see brackets in (a)]. Note that the shape of the distribution in (b) (i.e., increased counts in the center of the distribution) is due to the geometric efficiency of the crystal array when irradiated by a point source at \sim 30 cm distance.

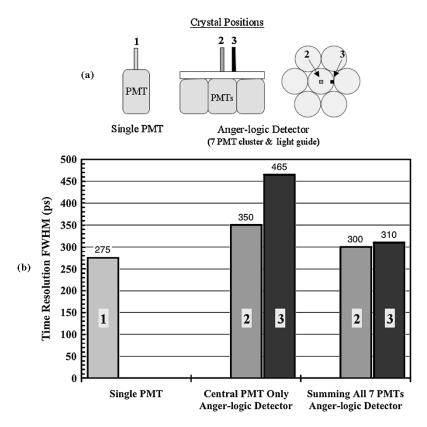


Fig. 7. (a) PMT and crystal arrangement for the three measured positions. In the Anger-logic arrangement both side and top views of the detector are shown. (b) Coincidence time resolution for a single 5.0% Ce LaBr₃ (4 mm \times 4 mm \times 30 mm) crystal coupled to a single PMT (labeled 1) and array of PMTs using the central and sum of seven PMTs (for crystal positions 2 and 3). Note the improvement for both position 2 and 3 with all 7 PMT's summed.

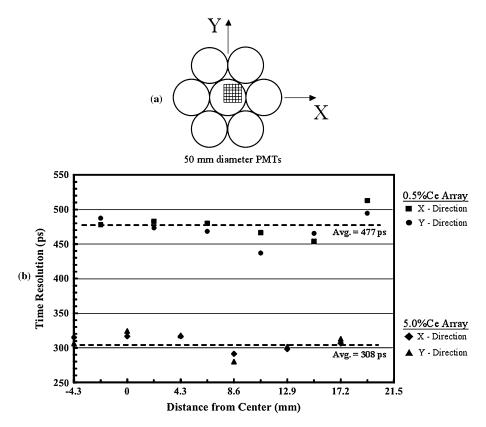


Fig. 8. (a) Arrangement of the seven PMT cluster and crystal array relative to the X- and Y-coordinate axis. (b) Coincidence time resolution for crystals in the 0.5% and 5.0% Ce arrays at various positions above the seven PMT cluster. Each point represents the average time resolution for two crystals at locations along each coordinate direction. The dashed lines indicate the average time resolution for the crystals in each array.

Array	% Ce	# Crystals	Average ΔE/E at 511 keV	Relative Light Output	$\sigma_{\!\scriptscriptstyle m LO}$	Average Time Resolution (FWHM)
1	0.5	100	5.31% <u>+</u> 0.44%	1.25	3%	477 <u>+</u> 23 ps
2	0.5	100	5.41% <u>+</u> 0.45%	1.19	3%	481 <u>+</u> 29 ps
3	0.5	100	5.20% ± 0.40%	1.29	2%	469 <u>+</u> 20 ps
4	0.5	100	5.47% ± 0.45%	1.37	2%	455 <u>+</u> 19 ps
5	5.0	25	5.65% ± 0.59%	1.31	4%	308 <u>+</u> 18 ps
6	5.0	25	5.42% <u>+</u> 0.42%	1.30	6%	300 <u>+</u> 13 ps
7	5.0	100	5.08% ± 0.68%	1.37	3%	290 <u>+</u> 10 ps

 $\begin{array}{c} \text{TABLE} \quad \text{II} \\ \text{Results With LaBr}_3 \quad \text{Crystal Arrays} \end{array}$

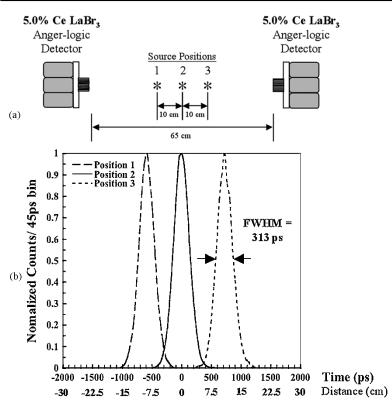


Fig. 9. (a) Arrangement of the two Anger-logic detectors and the three source positions. (b) Three measured time spectra for the source positions in (a). A time resolution of approximately 313 ps FWHM was obtained for each of the spectra. Note that the time scale is also given in units of distance and thus the time peaks correspond to an uncertainty in position of about 4.7 cm FWHM.

An average $\Delta E/E \sim 5.4\%$ was obtained for crystals in the arrays (see Table II) with little variation between crystals. The relative light output in Table II represents the average for the crystals in the array and is given relative to the single 4 mm \times 4 mm \times 30 mm samples tested in Section III. Light output equal to or better than the single crystal samples is obtained with little variation (i.e., low standard deviation in light output, σ_{LO}), thus indicating crystals in the arrays are well polished and packed in reflective powder.

V. COINCIDENCE TIME RESOLUTION FOR TWO LaBr₃ ANGER-LOGIC DETECTORS

The time resolution for two LaBr₃ Anger-logic detectors (array numbers 5 and 6, see Table II) placed in coincidence

was measured similarly to the timing measurements described in Section IV; however, the reference plastic-PMT detector was replaced with a second LaBr₃ Anger-logic detector. Each detector consisted of the LaBr₃ crystal array coupled to a light guide and cluster of seven PMTs (each a 50 mm diameter XP20Y0). The electronics shown in Fig. 5 for a single LaBr₃ detector was duplicated for the second detector allowing the interaction position and energy to be determined in the crystal arrays of both detectors.

Position and energy gates were placed on individual crystals in each detector allowing coincidence time spectra to be generated for pairs of crystals (i.e., events which deposit >450 keV in a single crystal in each detector). The two detectors were separated by 65 cm and irradiated with positron annihilation photon

pairs from a 68 Ge source. Fig. 9(a) illustrates the arrangement of the two Anger-logic detectors (each consisting of a 25 crystal 5.0% Ce LaBr₃ array) and the three source positions separated by 10 cm for which time spectra were measured. The coincidence time spectra measured at each source position for one pair of crystals are shown in Fig. 9(b).

A time resolution of 313 ps FWHM is obtained for each of the three spectra. This corresponds to an uncertainty in position $\Delta x=4.7$ cm (obtained by using $\Delta x=c*\Delta t/2$, where c is the speed of light and Δt is the time resolution). Incorporating this time resolution into a TOF PET scanner is expected to significantly improve image signal-to-noise ratio and noise equivalent count-rate (NEC) [3], [14]. A similar measurement was performed with two 0.5% Ce LaBr₃ crystal arrays and a time resolution of 605 ps FWHM was measured. Additionally, a measurement with two LYSO crystal arrays yielded a time resolution of 640 ps FWHM.

VI. CONCLUSION

Our results show that excellent energy and time resolution is obtained with LaBr $_3$ crystals in a 4 mm \times 4 mm \times 30 mm geometry suitable for use in a whole body 3-D PET scanner. Samples with higher Ce concentration had faster rise and decay times (with approximately equal light output) and subsequently yielded superior coincidence time resolution. However, the improvement from 0.5% Ce to 5.0% Ce was more significant than the improvement from 5.0% Ce to 10.0% Ce.

Excellent crystal discrimination was achieved with Anger-logic detectors consisting of multicrystal arrays of 4 mm \times 4 mm \times 30 mm LaBr $_3$ coupled to a light guide and cluster of seven PMTs. The crystals in the arrays exhibited uniformity in light output and energy resolution. By summing the seven PMT signals, excellent time resolution is maintained over the central region of the cluster. A time resolution of 313 ps FWHM was obtained for two 5.0% Ce LaBr $_3$ Anger-logic detectors in coincidence.

By expanding the basic detector design to a pixelated LaBr₃ Anger-logic detector with a continuous light guide and hexagonal arrangement of PMTs, our results indicate excellent performance can be attained. As we have shown with previous NaI(Tl) and GSO detector designs, a dynamically chosen local cluster of seven PMTs can be utilized to determine the energy and position for each event. Using this method we have also achieved excellent and uniform time resolution.

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