Development of Lithium Lanthanide Borate Scintillator For Neutron Scattering Applications

Grant No. DE-FG02-03ER46045

Final Report 2003-2004 Project Period

Program Goal - 1-mm Resolution Detectors for Single Crystal/Disordered Materials Diffractometers

Two approaches to designing 1-mm resolution detectors were evaluated during the first program year—an Anger-type detector and a Silicon-well type detector.

Task 1 - 1-mm Resolution Detector Employing LGB Scintillator for a Multi-Anode Anger Cameras

All of the prior neutron detectors constructed for neutron scattering applications were composed of evenly-sized particles of LGB scintillator (350-700 microns) uniformly distributed in an equal weight of scintillating plastic matrix. This detector design provided signal uniformity for instruments that do not require 1-mm resolution and had good pulse height resolution and gamma discrimination. At the suggestion of Dr. J. Schelten of Forschungszentrum, Jülich, we have constructed several neutron detectors in which the ground LGB crystals varied over an order of magnitude in size and were closely packed in a minimum amount of plastic scintillator binder. The purpose of this "maximum density" configuration is to provide adequate "micro-uniformity", necessary for the 1-mm resolution required in a neutron Anger camera.

We have been collaborating with neutron scattering instrumentation scientists in both Germany and Japan to ensure that the development and testing of the maximum density LGB detectors is completed in the context of advanced Anger cameras for neutron scattering instruments. The Anger-camera approach uses a centroid weighting algorithm to fix the point of neutron interaction. The resolution produced by this calculation is directly related to the light output of the scintillator employed and the size of the initial pixel. The Anger camera weighting scheme generally produces a factor-of-ten improvement in resolution compared to the pixel dimension. Present Anger-camera systems employ Li-glass scintillator and 25 or 50 mm pixels, which yields a resolution of 2.5 and 5 mm, respectively. To increase the resolution of the next generation of Anger-camera detectors, both groups have chosen to use Hamamatsu H8500 multianode phototubes with a pixel-size of approximately 5 mm and the increased light output of LGB scintillator. This combination should easily provide the required 1-mm resolution in two dimensions. We have supplied both the German and Japanese research programs with 50-mm square
samples of LGB bound with plastic scintillator to use in a single multi-anode PMT module. The enclosed reports describe the progress achieved in these collaborations.

The LGB crystals for our maximum-density detectors were ground and sorted into the 63-micron to 710-micron size range. Figure 1 shows the pulse-height response to thermal neutron capture of a sample made with 0.12 gm cm⁻² of ⁶Li, natural Gd, and ¹¹B LGB. The crystals were bound together at maximum density with 0.16 gm cm⁻² of plastic scintillator. Samples a) and b) in the section on gamma sensitivity below provide the gamma sensitivity data for these maximum-density Anger camera detector sheets.

Program Goal Summary

The summary of our work with the Japanese neutron scattering scientists is that multianode photomultiplier tubes in conjunction with LGB scintillator sheets will easily satisfy the 1-mm spatial resolution requirements of single crystal diffractometers at the SNS. A similar conclusion was previously reached by the Jülich group. Further work will be required to reduce the gamma sensitivity of neutron Anger cameras utilizing these methods. However, our previous work with sandwich gamma veto layers will be directly applicable to the solution of this problem.

Task 2 – A 1-mm Resolution Photodiode Detector Employing LGB Scintillator

The past two decades has seen increasing usage of miniaturized radiation detectors. Due to technical constraints, such miniaturization has primarily been only applied to gamma and charged particle detection with great success on very small scales. Inspired by the preliminary work of Cedric Allier under the direction of Carel van Eijk at Delft University, we set out to investigate application of the latest electronic technology to the miniaturization of neutron detectors. Our primary goal was to provide the means of fabricating a two-dimensional neutron detector for crystal diffractometry instruments with a 1 mm² resolution. The conclusion of our investigation was that such a device with 1 mm² pixels is presently not achievable. However, we determined that other non-miniaturized techniques, such as Anger camera methods, will meet the desired parameters.

Detector Considerations

The development and construction of a large pixilated neutron detector is a process that must be considered in two independent parts. The first part is the actual particle detection device which is usually a diode. This part has been easy to apply to gamma and charged particle detection due to the ability of those particles to directly interact with the diode material and make a signal. For this reason, most miniaturized work in radiation detection has taken place with gamma and charged particle detectors. Because of this, large pixilated diode arrays have been constructed using present fabrication technology.
The second area of consideration is that of signal processing. The complexity and size of the electronics is dependent upon the nature of the signal processing needed to be performed. Most systems require some sort of signal amplification. The most commonly utilized simple signal processing system integrates a number of pulses into a single signal. More complicated systems may time stamp and record the pulse height of each event.

Crystal Diffractometer Detectors

Three years ago Cedric Allier at the Delft University concluded work on a preliminary pixilated neutron detector. This detector married segmented neutron scintillators to respective photodiodes in a two-dimensional array. This was accomplished by bonding a 500 micron wafer to a second wafer containing a silicon on insulator (SOI) layer which was included in the bond. Next, the second wafer was polished till it had a thickness of 50 microns and photodiodes were fabricated on its surface. Then, Deep Reactive Ion Etching was used to etch 700 by 700 micron wells from the exposed side of the 500 micron wafer to the SOI layer. One well correlated with one photodiode. These wells were then filled with a neutron detecting scintillator such as LGB.

Figure 1 abc.
Schematic cross-sections of: a) unprocessed SOI wafer; b) diodes processing; c) final sensor.

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This design, although very involved, has its advantages:

- Each photodiode is associated with its own neutron scintillator which is contained in the well above the diode.
- This manner of fabrication allowed the construction of low noise diodes.
- The photodiode array was contained on a single chip.

This design was very preliminary and meant to prove the concept of deep well etching with photodiodes. It has drawbacks in several areas:

- There were no associated miniaturized signal processing or amplification electronics developed.
- Approximately a 250 micron wall thickness between wells. This large thickness significantly cut down on the active area of the detector.
- Large sloped undercut (30 – 40 microns) towards the bottom of the wells. This is due to the large depth of the etching and is an obstacle to making the wall thickness thinner.

Our initial plan was to improve upon Dr. Allier’s detector design by creating a similar deep-etched photodiode array that could be made on a large scale for single crystal diffractometry instruments. This meant building a device with etched wells having thin walls and negligible undercut. We also looked into developing microelectronic processors for each photodiode. The desired resolution for this detector was 1 mm². Working units of these prototypes would eventually tiled to a surface to cover an area of approximately 10 m².

After some investigation, it soon became apparent that deep etching is a recently developed and specialized field. Only a very few universities and companies have the capability to make the deep etches required to produce narrow wells. Etches on the scale of what was done for Allier’s device are produced at a very high price with little hope for making significant improvements on the wall thickness and undercut problems. With these circumstances coupled together, it was decided that the deep etching route was an inadequate direction to go.

Photodiode Arrays

As deep etching became an increasingly unattractive means of fabrication, another alternative was investigated. It involved replacing Allier’s 500-micron wafer that was etched through with a fabricated foil or plastic grid having thin walls. With this grid bonded to a wafer of silicon, the same procedures could now be performed as with Allier’s device excluding the etching process. After bonding, the first procedure is to polish the silicon wafer to 50 or 10 microns with the grid providing structure. If this step is successful without the bonded assembly failing apart, the diodes may now be fabricated on the side of the wafer opposite the grid. This process involves heating the structure at temperatures of around 1100°C. Such temperatures would melt any grid.
Another approach would be to fabricate the diodes on the silicon wafer on the side that the grid would be placed, and then bond on the grid. This technique suffers from electrical contact problems. As the grid is placed over the side of the wafer on which the diodes are fabricated, electrical leads from each diode must be channeled to the edge of the device for pickup. The closer-to-the-edge, higher density of leads significantly cuts down on the active area of the diodes. There is also the problem of routing the leads onto the backside of the device.

Dr. Schelten of Forschungszentrum, Jülich suggested an alternative scheme that is very simple in its fabrication. The idea was to place p-type strips the width of a pixel on one side of the silicon and on the other side to have strips running in a perpendicular direction but of n-type of material. Leads would be connected to one end of each of the strips and a pixel would be located by a signal coincidence in two perpendicular strips. The advantage of this design is the reduced number of electrical leads. On one side of the photodiode array would be bonded a foil or plastic grid which would be filled with a neutron scintillator. This design suffers from capacitance problems due to the larger area of the strips compared to a pixel which will affect the time resolution. The electronic engineering staff at Brigham Young University estimated that at least $10,000–15,000 would have to be spent on the development of this design with no guarantee of it working.

Electronics

For practicality, a miniaturized array of detectors requires miniaturized electronics. There are three obstacles in the way of electronic signal processing for the photodiode array due to such miniaturization:

- Connecting the photodiode electrical contacts with the processing electronics.
- The processing electronics require a larger volume than the detectors
- The expense in developing a processing chip.

Each of these points will be discussed independently.

The density of contacts for 1 x 1 mm photodiodes is too high to allow for wire contacts. Alternatives include bump bonding and conductive rubber mats. Bump bonding is a 35 year old technology which has found increasing use as contact densities have increased in many areas of electronics. Many specialized detection chips used in high energy physics employ this technology. As for the photodiode array, the contact size, although too small to use wire contacts, is too large to justify the expense of bump bonding. In addition to the expense to set up, there is a failure rate among a percentage of bump bonded contacts. Conductive rubber mats are said to work well for contacts within the dimensions of the photodiode array. However, since rubber mats require contacts on directly opposite sides of the mat, it is unsure a processing chip can be made in such a fashion.
As the photodiodes are shrunk in size, the electronics must decrease in size to fit in the same area as the detector. With the current state of technology, the extent of this miniaturizing of the electronics has limitations. Furthermore, as the miniaturization continues before the limits are reached, the expense continues to increase. In a project to design a photodiode array for use in PET imaging, Gruber et al. decided to use relatively large 3 x 3 mm pixels due to the complexity and expense of the electronics when considering the size of the pixels.

Finally, processing chips are expensive to design. They require a number of specialized designers with technical expertise, software, time, special materials and facilities.

Due to all the above cited constraints for designing a practical photodiode array with 1 mm² resolution and its associated electronics, it was decided to abandon this project.

After extensive discussion with instrument scientists concerning the optimum technique to achieve 1-mm resolution in two dimensions, it was decided to replace the photodiode technique with an Anger camera based upon multi-anode photomultiplier tubes.

**Program Goal – Powder Diffractometer Detectors**

**Task 1 – Improve Crossed-Fiber Readout Systems:**

We have developed a viable alternative to crossed-fiber readout systems employing LGB microcrystals in tubular geometry, read out by a single fiber. The enclosed preprint describes the results. This work was presented at the Tokyo Third International Workshop on Position-Sensitive Neutron Detectors and will be published in Nuclear Instruments and Methods A. See attached paper.

It is our opinion that we have achieved both program goals set for this grant. The techniques described above would also permit the design of detectors that would allow SNS to meet all of its goals for detector performance, if they were implemented.

**Program Goal – Determine Absolute Gamma Sensitivity of Neutron Detector Materials**

We have measured the absolute gamma sensitivity of six contemporary neutron detectors at 1.17, 1.33, and 6.13 MeV gamma energy. These detectors are normally fabricated with neutron-sensitive isotopes, such as °Li and °He. For this study, some of the detectors were fabricated with neutron-insensitive isotopes, °Li and °He) eliminating the possibility of neutron signal contaminating the gamma signal and yielding an “absolute” gamma sensitivity for each detector at the three energies listed above.
The detectors are as follows:

a) Lithium gadolinium borate (LGB) microcrystals made with $^7$Li, natural Gd, and $^{11}$B. A layer of 0.165 gm cm$^{-2}$ LGB was bound together with 0.032 gm cm$^{-2}$ of plastic scintillator. 0.375 gm cm$^{-2}$ of plastic scintillator was placed downstream from the LGB to serve as a veto detector for Compton-scattered electrons.

b) A sample similar to (a) above but made with $^6$Li, natural Gd, and $^{11}$B.

c) A sample with $^6$Li, natural Gd, and $^{11}$B LGB mixed uniformly with plastic scintillator. The sample contained 0.165 gm cm$^{-2}$ of LGB and 0.384 gm cm$^{-2}$ of plastic scintillator.

d) A 1-mm-thick sheet of GS-30 glass scintillator made with separated $^7$Li.

e) A commercial ZnS/LiF scintillator matrix made with highly purified $^7$Li, to avoid neutron sensitivity. The material was the standard 0.4 mm thick.

f) A 1.27 cm diameter proportional counter filled with $^3$He. An identical counter filled with $^3$He was used to obtain the response function to thermal neutron absorption. The $^4$He counter yielded only the response to incident gamma rays.

The Brigham Young University Van de Graaf accelerator provided 1.6 MeV protons to induce the $^{16}$F(p,$\gamma$) reaction in a teflon foil. This reaction yields 6.13 MeV gamma rays at a rate of 2x10$^9$ per hour from the excited $^{16}$O produced as a result of the (p,$\gamma$) reaction. The absolute rate of gamma production was measured with a calibrated NaI scintillator detector. The scintillator detector samples were optically sealed to a five-inch phototube placed approximately 50 cm from the target.

An absolutely calibrated $^{60}$Co source was obtained from Isotope Products Laboratory to provide 1.17 MeV and 1.33 MeV gamma rays for the low energy measurements.

The scintillator detector signals were analyzed for pulse shape to discriminate as much as possible against gamma generated events. The results shown in Table I therefore represent the achievable gamma insensitivity of these neutron detectors.

($S_{1.33}$ signifies the sensitivity to an equal number of 1.33-MeV and 1.17-MeV gamma rays from $^{60}$Co.)
Table I
Absolute Gamma Sensitivity of Contemporary Neutron Detectors

<table>
<thead>
<tr>
<th>Detector</th>
<th>$S_{6.13}$</th>
<th>$S_{1.33}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>a) 7Li LGB</td>
<td>$2.0 \pm 0.4 \times 10^{-5}$</td>
<td>$0.06 \pm 0.17 \times 10^{-5}$</td>
</tr>
<tr>
<td>b) 8Li LGB</td>
<td>$28 \pm 2 \times 10^{-5}$</td>
<td>$0.09 \pm 0.05 \times 10^{-5}$</td>
</tr>
<tr>
<td>c) Homogeneous 6Li</td>
<td>$42 \pm 2 \times 10^{-5}$</td>
<td>$0.20 \pm 0.05 \times 10^{-5}$</td>
</tr>
<tr>
<td>d) 7Li Glass</td>
<td>$223 \pm 2 \times 10^{-5}$</td>
<td>$0.11 \pm 0.04 \times 10^{-5}$</td>
</tr>
<tr>
<td>e) ZnS/LiF</td>
<td>$-0.03 \pm 0.01 \times 10^{-5}$</td>
<td>$0.0085 \pm 0.0045 \times 10^{-5}$</td>
</tr>
<tr>
<td>f) 4He</td>
<td>$49 \pm 2 \times 10^{-5}$</td>
<td>$6.4 \pm 0.1 \times 10^{-5}$</td>
</tr>
</tbody>
</table>

Table II lists the absolute gamma sensitivity of the 1.27-cm diameter 4He-filled proportional counter. “Bias Level” refers to the ratio between the lower-level discriminator setting and the channel number at the peak of the neutron-capture distribution. A setting of 22% corresponds to the minimum in the distribution just below the plateau. The upper level discriminator was set to include all capture events in a matched 3He proportional counter.

Table II
Sensitivity of 4He-Filled Proportional Counter to 6.13 MeV Gammas

<table>
<thead>
<tr>
<th>Bias Level</th>
<th>Absolute Gamma Sensitivity</th>
</tr>
</thead>
<tbody>
<tr>
<td>22%</td>
<td>$49 \pm 2 \times 10^{-5}$</td>
</tr>
<tr>
<td>30%</td>
<td>$5.4 \pm 0.4 \times 10^{-5}$</td>
</tr>
<tr>
<td>50%</td>
<td>$-0.2 \pm 0.3 \times 10^{-5}$</td>
</tr>
</tbody>
</table>

The following recommendations are based upon these results:

1) The gamma sensitivity of commercial 3He proportional counters is unacceptably high when the bias is set near the lower end of the plateau region. A bias setting at 50% of the peak pulse height is required to obtain acceptable gamma sensitivity. These requirements will have only a modest effect upon neutron detection efficiency but will increase the sensitivity to gain changes. The gamma sensitivity can be significantly reduced by proper choice of shielding materials, such that high-energy gamma background (>2 MeV) is avoided.

2) A gamma sensitivity of $10^{-5}$ is considered to be marginally acceptable at many neutron scattering instruments. For this reason, it is vital in general to avoid all instrumental materials, exposed to neutrons, that would yield high energy capture-gamma rays. This is true for all detectors except ZnS/LiF.
Li-6 borate at maximum density with veto layer; Cf source

Early/late plot

Lithium capture histogram

FIGURE 1
Development of position-sensitive neutron detector based on scintillator

K. Sakai\textsuperscript{a,b,\ast}, K. Hirot\textsuperscript{b}, T. Adachi\textsuperscript{b}, K. Ikeda\textsuperscript{b}, T. Morishima\textsuperscript{b}, H.M. Shimizu\textsuperscript{b}, M. Furusaka\textsuperscript{c}, S. Sato\textsuperscript{c}, Y. Kiyana\textsuperscript{g}, N. Sakamoto\textsuperscript{d}, T. Sakuma\textsuperscript{d}, T. Oku\textsuperscript{c}, J. Suzuki\textsuperscript{e}, K. Littrell\textsuperscript{f}, C.-K. Loong\textsuperscript{f}, J.B. Czirr\textsuperscript{g}, T.K. McKnight\textsuperscript{g}

\textsuperscript{a}Department of Physics, Tokyo Institute of Technology, 2-12-1 O-okayama, Meguro-ku, Tokyo 152-8551, Japan
\textsuperscript{b}RIKEN, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan
\textsuperscript{c}High Energy Accelerator Research Organization, 1-1 Oho, Tsukuba, Ibaraki 305-0801, Japan
\textsuperscript{d}Hokkaido University, Kita 8, Nishi 5, Kita-ku, Sapporo, Hokkaido 060-8628, Japan
\textsuperscript{e}Japan Atomic Energy Research Institute, 2-4 Shirakata-shi, Tokai, Ibaraki 319-1195, Japan
\textsuperscript{f}Intense Pulsed Neutron Source, Argonne National Laboratory, 9700 S. Cat\textsuperscript{e} Ave, Argonne, IL 60439-4814, USA
\textsuperscript{g}Photogenics Inc, 515 East 1860 South, Provo, UT 84606, USA

Abstract

For evaluating neutron optical devices, we have developed the two-dimensional position-sensitive neutron detectors (PSND) of the crossing fibre array type. In parallel, we have studied the PSNDs of the direct coupling type with flat panel photomultiplier tubes for responding to various spatial resolutions with high detection efficiency. This paper views and reports on the recent status of development of PSNDs based on neutron scintillators in neutron optics group. © 2004 Elsevier B.V. All rights reserved.

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Keywords: UCN; Position sensitive neutron detector; WLS crossing fibers; Anger camera

1. Introduction

Designs of neutron detectors strongly depend on requirements from neutron spectrometers. Especially, in neutron scattering experiments with a high intense pulsed neutron source and neutron optical devices such as a lens and prism, neutron detectors require high capabilities such as a large sensitive area, high spatial resolution, high counting rate, high detection efficiency, good temporal resolution for a time-of-flight method, and so on. For example, in typical instruments of Japanese Spallation Neutron Source, position-sensitive neutron detectors (PSND) having various pixel sizes from 0.25 to 1000 mm\textsuperscript{2} and total sensitive areas from 0.04 to 40 m\textsuperscript{2} are required. In such a situation, it is important to develop PSNDs based on not only gas but also scintillator [1]. A PSND

*Corresponding author. Tel.: +81-3-5734-2716; fax: +81-3-5734-2731.
E-mail address: sakai@ap.nucl.ap.titech.ac.jp (K. Sakai).
of the crossing fiber array type has an advantage for achieving a large sensitive area with high spatial resolution. So we have developed it for evaluating neutron optical devices [2–7]. In parallel, we have also studied the PSND of the direct coupling type with multianode photomultiplier tubes (MAPMT) for responding to various spatial resolutions with a high neutron counting rate and detection efficiency. This paper views and reports on the recent status of development of PSNDs based on neutron scintillators in neutron optics (NOP) group.

2. PSND of crossing fiber array type

Fig. 1 shows a PSND (PDI) having a 50 × 50 mm² sensitive area. The PDI consists of a 0.4 mm-thick ZnS/6LiF scintillator plate and two wavelength shifting (WLS) fiber arrays optically coupled onto the upper and lower side of the plate. Each array consists of 128 fibers with cross-sections of 0.4 × 0.4 mm². The direction of fiber array X is orthogonal to that of array Y for determining the neutron position. The neutron entering the scintillator generates charged particles by the 6Li (n, α)t reaction and photons are caused by the charged particles. The photons having a large incident angle with respect to the fiber surface enter and are absorbed in the fiber. The fiber isotropically radiates photons with shifted wavelengths. The photons transmitting through the fiber are detected by four MAPMTs having 64 anodes and one last dynode (Hamamatsu: H7546). The output signals of the anodes are amplified and individually analyzed by ADC modules mounted in the VME crate. The X and Y are decided by selecting the maximum values of anode outputs. A preliminary test of the PDI has been carried out with the pulsed neutron source at Hokkaido University and KEK [6,7]. Fig. 1(b) shows a neutron image obtained by using a Cd mask of Chinese character. As a result, a neutron counting rate of 10 kHz, a neutron efficiency of 6% at 4 Å, and a pixel resolution of 0.4 × 0.4 mm² were obtained [7].

Fig. 2 shows the developed PSND (PD2) having a 128 × 128 mm² sensitive area and a 0.5 × 0.5 mm² pixel size for a focusing small-angle scattering instrument. Each fiber array of the PD2 consists of 256 fibers with cross-sections of 0.5 × 0.5 mm². The fibers are connected to four 64ch-MAPMTs by each fiber array. For a PSND having a large sensitive area such as the PD2, we
studied a dynode coding as a "practical" coding technique. In this coding, at one end of the fiber array $X$, every 64 fibers are connected to the 64-ch-MAPMT as Belt $X$. While, anode outputs having the same anode number are picked up from four MAPMTs and bundled as Stripe $X$. The Belt $X$ can be decided by selecting the maximum value of dynode outputs, and the Stripe $X$ by that of anode outputs. The $X$ direction of neutron position is decided by the combination of Belt $X$ and Stripe $X$. The $Y$ direction is also determined by the same process. This coding has the advantage that the data correcting process for a spatial uniformity becomes simple because a relation of a fiber and an anode output is one-to-one correspondence. By applying the coding to the PD2, readout channels could be reduced from 512 to 136. Already we have observed a neutron image by the PD2 with Cd mask [6].

Although the PSND with crossing fibers has an advantage for a large sensitive area with high spatial resolution, some problems such as a low efficiency and low counting rate have remained yet. They are caused by poor light yields reaching to anodes. So we are studying scintillator materials and MAPMTs for improving capability of our PSNDs.

3. Application of PSNDs of crossing fibers

We have carried out measurements of neutron diffractions with our PSNDs to demonstrate their usefulness for neutron scattering experiments [6,7]. One of our experiments was performed using a pulsed neutron beam from the QUIP beam line at Argonne's facility. A distance between the neutron source and a sample is about 10.5 m. Thermal neutrons diffracted from a sample were measured by the PD1 mounted at distance of 0.18 m from the sample position, and perpendicularly with respect to the incident beam direction.

Figs. 3–5 show the results of diffraction measurement of a quartz single crystal ($\alpha$-SiO$_2$). Fig. 3 represents the TOF spectrum of the PD1 plotted every 40 µs. Fig. 4 represents a series of two-dimensional neutron images observed by the PD1. In Figs. 4 and 5, one channel corresponds to 0.4 mm. Each image in Fig. 4 is obtained by integrating every 0.4 ms between 4.66 and 6.26 ms of the TOF spectrum. The integration region is painted by diagonal lines in Fig. 3. Each region of neutron wavelength of (a), (b), (c) and (d) in Fig. 4 is 1.76–1.90, 1.91–2.05, 2.06–2.20, and 2.21–2.35 Å. We can see that many Laue spots from the quartz crystal appear and disappear as the region of neutron wavelength changes. Fig. 5 shows one-dimensional spatial distribution as a function of position $X$ within an open square in Fig. 4(b). The distribution in Fig. 5 is obtained by integrating with respect to the $Y$-direction. In Fig. 5, some peaks of Laue spots having about
Fig. 4. Two-dimensional neutron images diffracted a quartz single crystal: Each region of neutron wavelength of (a), (b), (c) and (d) is 1.76-1.90, 1.91-2.05, 2.06-2.20, and 2.21-2.35 Å.

2.5 mm FWHM are observed clearly because of the high resolution capability of the PD1. We notice the peaks can be discriminated from backgrounds efficiently by selecting proper spatial and temporal region because the peaks of diffractions depend on spatial and temporal structure strongly in comparison with backgrounds. These results suggest the PD1 becomes a powerful tool for neutron scattering experiments with a pulsed source.

4. PSND of the direct coupling type

In order to respond to various requirements from instruments, we have also studied PSNDs of the direct coupling type with a MAPMT. They are superior to the PSND with crossing fibers on the number of photons reaching to MAPMT. So we can use transparent scintillators such as a ⁶Li glass and ⁶LiGdBO whose primary light yields are less than that of a ZnS/¹²LiF scintillator. A transparent property is very useful for a high detection efficiency and discrimination of neutron events.
For high spatial resolution less than 1 mm², we have developed the PSND based on Anger camera technique [8]. In Fig. 6, it consists of a transparent neutron scintillator plate, light guide and MAPMT coupled onto lower side of the plate. A neutron entering the scintillator generates charged particles by the neutron reaction and photons are caused by these particles. The photons dispersing through the light guide are detected by several anodes of a MAPMT. The signals from anodes and a last dynode are analyzed by ADC modules. A neutron event is discriminated with the neutron reaction peak in the pulse height spectrum of the dynode output. A neutron position is determined by analysing the center of light yield with several anode outputs. As MAPMT, we consider a flat panel photomultiplier tube (FPPMT; Hamamatsu; H8500) which has 64 anodes, a 5.8 x 5.8 mm² anode size, 6 mm interval D between anode centers, and 49 x 49 mm² effective area. As the spatial resolution of the PSND is proportional to $D/\sqrt{N}$, where $N$ is photoelectrons reaching to anodes, this FPPMT is suitable to obtain a high resolution because of its small $D$. According to the estimation with a 6 mm $D$ and a primary light yield of $^6\text{Li}$ glass, its spatial resolution will become less than 0.5 x 0.5 mm².

Fig. 7(a) shows a fabricated PSND (PD3) consisting of a $^6\text{Li}$GdBO scintillator of a 50 x 50 x 1 mm³ size, a light guide of 5 mm thickness and H8500. Fig. 7(b) represents the preliminary result of a x ray image by using the PD3 and a slit of 1 mm diameter. The spatial resolution is not so good yet and ghost peaks also appear. They are caused by the electric noises, poor adjustments of gains of anode outputs, and small detection efficiency of photons at the edge of each anode pixel. The improvement of the PD3 is now in progress [9].

For spatial resolutions between 1 and 10 mm², we consider the digital Anger system based on a modified Anger camera. In the system, photons caused by a neutron reaction disperse through a light guide and reach to several anodes of a FPPMT. The anodes detecting the photons are controlled to be less than four by adjusting a thickness of a light guide. A neutron event is discriminated with a neutron reaction peak of dynode outputs, but a neutron position is decided by only the digital signals of "on/off" from anode outputs. For example, in Fig. 6, we can decide neutron hit positions of A, B and C by detecting the digital signals of "on" from the anodes of $(X2, Y2)$, $(X5, Y3, Y4)$ and $(X3, X4, Y5, Y6)$. This system enables to be a spatial resolution of a quarter of one anode size and very simplified data taking system without analyzing the center of light yield. We have a plan to fabricate the PSND with a FPPMT having 256 anodes and a 3 x 3 mm² anode size for obtaining a spatial resolution of 1.5 x 1.5 mm².

For spatial resolution between 10 and 100 mm², we consider a PSND consisting of a FPPMT and divided scintillator pixels, where each pixel is connected to each anode through a light guide tube in parallel. Although its concept is similar to the PSND developed by NOP group in Ref. [10], we use a light guide tube that its cross-section at the anode side is shrinking in comparison with that at the scintillator side. Photons caused by a neutron reaction are detected by each anode of FPPMT after passing through the guide tube. A signal of neutron event is discriminated with a neutron reaction peak of each anode output in parallel. This PSND has advantages of a miniaturization and cost performance because of using one anode of FPPMT as a single anode PMT. For realizing this PSND, the development of light...
guide tubes having good property on light transmission is now in progress in our group.

5. Conclusion

Recent status of the development of PSNDs based on neutron scintillators in NOP group was viewed and reported. For obtaining a large sensitive area with a spatial resolution less than 1 mm², PSNDs of crossing fiber array type have been studied. The PSNDs having a 50 × 50 and 128 × 128 mm² sensitive area with less than 0.5 × 0.5 mm² pixel size have been developed, and their usefulness are demonstrated by measuring neutron diffractions. In parallel, PSNDs of the direct coupling type with a FPPMT have been studied. For a spatial resolution less than 1 mm² with high detection efficiency, a PSND based on Anger Camera has been developed and the preliminary result of x ray image was reported. For spatial resolutions between 1 and 100 mm², the digital Anger system and a PSND based on pixel divided scintillators were discussed. Although the sensitivity of γ rays and stability should be investigated, we can show the various possibilities of the PSNDs based on scintillators through our works.

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References

LGB Scintillators for Anger-type Neutron Detectors

J. Schelten and R. Engels

Forschungszentrum Jülich GmbH, D52428 Jülich Germany

The scintillator is the most critical component in any Anger-type neutron detector and thus, scintillator properties seriously influence the quality of these detectors. The (1) neutron detection efficiency, (2) spatial resolution, (3) insensitivity to high energy gammas; and, (4) uniformity of the detector response are all determined essentially by the scintillator properties. In almost all existing scintillator detectors, commercially available Li-glass scintillator is used. LGB is the only attractive alternative, for the following reasons:

(1) Detectors supplied by Photogenics were composed of LGB scintillator uniformly distributed in a polymer matrix. These detectors were designed and fabricated in such a way that the neutron absorption was the same as for a 1-mm glass scintillator. Despite an always observed broad pulse height distribution, a lower level discriminator could be set such that all neutron absorption events and none of the electronic noise signals are recorded.

(2) The spatial resolution of an Anger-type detector is proportional to the square root of the light output per neutron absorption event. Because of the large LGB light pulses, which are about four times larger than in Li glass, LGB scintillators were of particular interest for Anger-type cameras.

(3) The gamma insensitivity of neutron detectors always plays a critical role because a low gamma sensitivity improves the signal-to-noise ratio of every scattering experiment at both continuous and pulsed neutron sources. With LGB scintillators, a pronounced lower gamma sensitivity was measured which dramatically helped to reduce the background in scattering experiments.

(4) The composite LGB scintillator material consisting of single crystalline LGB grains and polymer binder is certainly less homogeneous than a glass scintillator. One expects local composition fluctuations due to variations of the average size, of the size distribution and of the volume fraction of grains which result in locally varying absorption probabilities of neutrons and in locally varying heights of light pulses generated by neutrons. Such annoying effects were observed with two-dimensional neutron detectors using such LGBG scintillators instead of glass scintillators. The composition fluctuations could be detected more directly by X-Ray Fine Focus Absorption measurements. The absorption diagrams clearly showed fluctuations with a strength which increased with decreasing scintillator volume sampled in the
absorption experiment. For volumes below 1mm$^3$ the fluctuations became really dramatic. Systematic studies were planned in order to correlate the neutron light fluctuations with the X-ray absorption fluctuations. However, such experiments have not been started yet.

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“LBG-Anger.doc”
An improved detector for powder diffractometers

J. Bart Czirr and Thomas K. McKnight
Photogenics, Inc., 515 East 1860 South, Provo, UT, 84606, USA
Email Address: photogenicsczirr@aol.com

Abstract

We present a new pixel design for powder diffractometers located at pulsed neutron sources. Small crystals of lithium gadolinium borate scintillator are enclosed in a tubular housing, with a single co-axial wave-length-shifting fiber used to collect and transmit the scintillator light to a pair of photomultiplier tubes. In the best case, 44 photoelectrons are detected from a neutron capture in $^6$Li event, when the light from both ends of the fiber is summed. Two techniques for discriminating against gamma background are described and quantified.

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Keywords: Scintillator, Neutron detector, Powder diffractometer

1. Introduction

We have designed and tested a new pixel configuration for powder diffractometers located at pulsed neutron scattering facilities. Detectors incorporating this pixel design are intended to satisfy the published detector requirements for POW-GEN3, the proposed powder diffractometer for the Spallation Neutron Source (SNS) at Oak Ridge, Tennessee[1]. These requirements are listed in Table 1.

Table 1
POW-GEN3 diffractometer specifications

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pixel size</td>
<td>0.5 cm wide by 4-to-40-cm long</td>
</tr>
<tr>
<td>Neutron capture efficiency</td>
<td>50% at 0.5 Å</td>
</tr>
<tr>
<td>Time Resolution</td>
<td>1 microsecond</td>
</tr>
<tr>
<td>Gamma detection efficiency</td>
<td>$&lt;10^{-6}$</td>
</tr>
</tbody>
</table>

Our design is centered around the new inorganic scintillator $\text{Li}_6\text{GdB}_5\text{O}_{12}:\text{Ce}$, Lithium Gadolinium Borate(LGB). The characteristics of this material are as follows: 50 cm$^3$, water-white, single-crystal boules grown by Czochralski method[2], 8 isotopic permutations with Li, B, Gd (even or odd isotopes) constituents, ($\text{Li}$-6, $\text{B}$-10, odd Gd) = high $\sigma_n$, ($\text{Li}$-7, $\text{B}$-11, even Gd) = low $\sigma_n$.

The scintillation efficiency of Lithium Gadolinium Borate is comparable to a variety of well-known neutron and gamma scintillators, as show in Table 2.
Table 2
Measured yield for inorganic scintillators.

<table>
<thead>
<tr>
<th>Scintillator</th>
<th>Photons/MeV (electrons)</th>
<th>Photoelectrons/Neutron</th>
</tr>
</thead>
<tbody>
<tr>
<td>YSO</td>
<td>30,000</td>
<td>--</td>
</tr>
<tr>
<td>LSO</td>
<td>26,000</td>
<td>--</td>
</tr>
<tr>
<td>LGB</td>
<td>25,000</td>
<td>8,300</td>
</tr>
<tr>
<td>GSO</td>
<td>8,000</td>
<td>--</td>
</tr>
<tr>
<td>Li glass</td>
<td>3,700</td>
<td>1,140</td>
</tr>
</tbody>
</table>

The characteristics of LGB's light emission are: emission wavelength; 370-470 nm, index of refraction; 1.64 (very low), principal decay time; 200 nsec. The high scintillation efficiency depends upon efficient excitation energy transfer to the 4% Ce$^{3+}$ ions. This transfer is greatly enhanced by the half-filled f-shell of Gd$^{3+}$. This also produces optical transparency in Gd$^{3+}$ compounds. For neutron scattering detectors, the highly exothermic $^6$Li reaction must be employed. To optimize the light output of the scintillator, the LGB molecule must be fabricated with the $^{160}$Gd and $^{11}$B isotopes, increasing the cost of the new scintillator.

We have searched for alternatives to Lithium Gadolinium Borate among related compounds and not found any material with a comparable scintillation efficiency. Table 3 lists the results.

Table 3
Alternative scintillators

<table>
<thead>
<tr>
<th>Scintillator</th>
<th>Light Output</th>
<th>Form</th>
</tr>
</thead>
<tbody>
<tr>
<td>Li$_6$Y$_5$B$<em>3$O$</em>{12}$:Ce</td>
<td>20% of LGB light output</td>
<td>20% of LGB light output</td>
</tr>
<tr>
<td>Li$_3$Y$_2$B$_3$O$_9$:Ce</td>
<td>No crystalline form</td>
<td>No crystalline form</td>
</tr>
<tr>
<td>Li$_3$Y$_2$B$_3$O$_9$:Ce</td>
<td>No crystalline form</td>
<td>No crystalline form</td>
</tr>
<tr>
<td>Li$_3$La$_2$B$_3$O$_9$:Ce</td>
<td>No measurable light</td>
<td>No measurable light</td>
</tr>
<tr>
<td>Li$_3$Ce$_2$B$_3$O$_9$</td>
<td>No measurable light</td>
<td>No measurable light</td>
</tr>
<tr>
<td>Li$_6$HoB$_3$O$_9$:Ce</td>
<td>No measurable light</td>
<td>No measurable light</td>
</tr>
<tr>
<td>Li$_6$BiB$_3$O$_9$:Ce</td>
<td>No crystalline form</td>
<td>No crystalline form</td>
</tr>
<tr>
<td>Li$_6$ThB$_3$O$_9$:Ce</td>
<td>No crystalline form</td>
<td>No crystalline form</td>
</tr>
</tbody>
</table>

The conclusion is that separated, even isotopes of Gd are required for neutron wavelengths greater than 0.5 Å.

2. Readout

In most applications of LGB, we have ground large crystals into sub-mm size and incorporated these microcrystals in polymerized plastic or plastic scintillator. An example is shown in the figure of a GEM module at ISIS (Figure 1). Each pixel is read
out by 100 end-on fibers distributed to 2 phototubes. A neutron captured in the $^6\text{Li}$ results in approximately 25 photoelectrons in each PMT.

Scientists at ORNL have built modules consisting of flat plates of ZnS/LiF scintillator viewed by wave-length-shifting fibers (WLSF) lying parallel to the surface. We attempted to duplicate this geometry using an LGB/plastic matrix and a single WLSF. The advantage of the transparent LGB material is the peaked response upon $^6\text{Li}$ capture. However, in flat geometry, the variations in solid angle subtended by the single fiber are large enough to obscure the peak.

We then turned to a tubular geometry to minimize the variations in solid angle and thereby retain the peaked response function. We tested this technique using 3-mm and 5-mm inside diameter x10 cm long Al tubes with a co-axial 1-mm-diameter BCF-91A WLSF. The tubes were filled with mineral oil and close-packed LGB microcrystals. Figure 2 shows the pulse-height response with 3-mm tubing, 250 to 350 micron LGB crystals, and with both ends of a 1-m-long fiber fastened to an ETL-9266B PMT. The detector was irradiated by a $^{252}\text{Cf}$ source moderated with paraffin blocks. The PMT, amplifier and pulse height analyzer system was calibrated with a small Li-glass scintillator that exhibits no intrinsic broadening with neutron capture. This calibration yields the number of photoelectrons corresponding to the peak channel number, directly from the Gaussian width. The peak in Figure 2 corresponds to 44 photoelectrons from the sum of both ends of the single fiber.

Figure 3 shows the response of a 5-mm tube with a 1-mm fiber, and filled with 350 micron to 500 micron LGB in mineral oil. The peak corresponds to 27 photoelectrons. Figure 4 shows the pulse height response for the 3-mm tube, with only one end of the fiber on the PMT. The peak corresponds to 23 photoelectrons. For all of these figures, a small moderator-out background has been subtracted from the moderator-in data. The sharp rise near threshold is due to 2.2 MeV gammas interacting in the borate scintillator.

3. Gamma Rejection

We have developed two techniques to reduce the gamma sensitivity of the tubular pixels to an acceptable level.

The first technique relies on the forward-peaked angular distribution of Compton-scattered electrons for MeV gamma rays. Figure 5 shows the angular distribution of electrons generated by Compton scattering of 2-MeV gamma rays. The electron energy also drops off rapidly with angle. To take advantage of this effect, we placed a sheet of plastic scintillator downstream from a thin sheet of LGB/plastic matrix. Figure 6 shows the distribution of events with a simple pulse-shape-discrimination analysis. The fast-decaying electron events are in the top island, and the slow-decaying captured-neutron events are in the bottom island. A $^{238}\text{Pu}/^{13}\text{C}$ neutron, gamma source was used with paraffin moderator. The gamma rays are of predominantly 6 MeV energy. The plastic
scintillator veto detector was 1.4 mm thick. Figure 7 shows the response to $^{226}$Ra gamma rays.

The second gamma rejection technique again relies on pulse-shape discrimination to separate electron and neutron events. We mixed small crystals of LGB with ground yttrium orthosilicate (YSO) scintillator and incorporated them in polyvinyl toluene plastic sheets. The decay time of YSO is 70 ns, so the gamma background events are easily separated from the slower LGB neutron-capture events. Figure 8 shows the resulting separation with pulse-shape discrimination applied. The LGB used for Figure 9 was made with $^7$Li to eliminate low-energy neutron capture. Ambient gamma background produced the isolated island. The lack of interference with the neutron capture events is very encouraging.

4. Conclusions

Based upon these results, we come to the following conclusions:

The tubular design with transparent LGB as the neutron sensitive scintillator is just barely adequate at this stage of development. Either or both of the described gamma rejection techniques will be necessary to reach the insensitivity to background required at powder diffractometer instruments. Weaker or less transparent scintillators will not produce a peaked response function, even in this ideal tubular geometry. No existing scintillator will be adequate with a flat scintillator, viewed by WLSF parallel to the surface. The added cost for scintillator material in tubular geometry is 4% greater than flat geometry, at equal neutron detection efficiency. A single WLSF in tubular geometry can replace 100 end-on fibers in flat geometry, for the same pixel size. There is no penalty in extending the pixel length with WLSF.

References


Figure 1. GEM module at ISIS.

Figure 2. Response of tubular pixel to neutron capture in LGB. 3-mm ID tube with 250 to 350 micron LGB. Both ends of WLSF attached to PMT.

Figure 3. Response of tubular pixel to neutron capture in LGB. 5-mm ID tube with 350 to 500 micron LGB. Both ends of WLSF attached to PMT.
Figure 4. Response of tubular pixel to neutron capture in LGB. 3-mm ID tube with 250 to 350 micron LGB. One end of WLSF attached to PMT.

Figure 5. Number of electrons per unit solid angle vs. angle, relative to the direction of incident 2.04-MeV gamma rays.

Figure 6. Pu/C-13 neutron/gamma source. LGB plus PVT binder, with 0.142 gm/cm² BC408 backing. Note the excellent separation between neutron and gamma induced events.
Figure 7. Ra-226 gamma source. LGB plus PVT binder, with 0.142 gm/cm² BC408 backing. Note the absence of a neutron capture peak.

Figure 8. Pu/C-13 Source 2:1 YSO:LGB PVT Binder/Li-6 LGB 50,000 Events

Figure 9. Background 2:1 YSO:LGB PVT Binder/Li-7 LGB 50,000 Events