Energy Resolution of Ce:GAGG and Pr:LuAG Scintillators coupled to 3mm × 3mm Silicon Photomultipliers

A. G. Stewart, B. Seitz, K. O’Neill, Member, IEEE, L. Wall, Senior Member, IEEE and J. C. Jackson, Senior Member, IEEE

Abstract—Silicon Photomultipliers (SiPM) have shown great promise as a suitable replacement for conventional vacuum based Photomultiplier Tubes (PMT). Progress in recent years has been vast with SiPM sensors pushing the boundaries in energy and timing resolution as well as photon detection efficiency and active surface area. In this study we report the energy resolution and linearity of two novel scintillator crystals, Cerium doped Gd₃Al₂Ga₂O₁₂ (Ce:GAGG) and Praseodymium doped Lu₃Al₂O₁₂ (Pr:LuAG), readout by three different types of 3 × 3mm² silicon photomultiplier sensors produced by SensL (MicroFM, MicroFB, MicroFC). Such a scintillator-SiPM architecture has applications in pre-clinical and clinical medical imaging modalities such as Positron Emission Tomography (PET), N- on-P (M series), P-on-N (B series) and a low-noise P-on-N (C series) versions of the SiPM sensor platform have been coupled to the crystals and the response characterized over a range of gamma energies and detector bias values. A saturation-corrected energy resolution of 11.7% was achieved for a MicroFM device coupled to a 3 × 3 × 30 mm³ Ce:GAGG crystal at 20°C. An energy resolution of 16.1% was obtained for a MicroFB device optically coupled to a 3 × 3 × 30 mm³ Pr:LuAG crystal at 20°C.

Index Terms—Silicon Photomultiplier, Geiger Mode Photodiodes, Silicon, Ce:GAGG, Pr:LuAG, Gamma Spectroscopy, Scintillation Detection, Positron Emission Tomography, PET, SPECT.

I. INTRODUCTION

THE detection of low-level light using arrays of Geiger-mode Avalanche Photodiodes (GAPD), commonly known as a Silicon Photomultiplier (SiPM), has received considerable interest from both academic research and industry alike [1]–[5]. In recent years there has been a significant increase in the use of SiPM sensors in a large variety of applications together with continuous optimization of the design and operating parameters by a variety of manufacturers. SiPM sensors provide a compact, single-photon sensitive, detection platform which, due to its production process, offers a cost effective alternative to conventional photon detection solutions. In addition, SiPM sensors have a low operating voltage and are inherently insensitive to magnetic fields [6]. These features have meant there has been significant research effort in investigating SiPM sensors for a variety of medical imaging applications either in combination with a high magnetic field (e.g. PET-MRI) or where a compact design is desired [7], [8], [24]. Modern medical imaging techniques such as PET require both fast, high-density scintillator materials together with compact, spectrally matched, high sensitivity soft photon sensors for optical read-out. Scintillator materials such as Lutetium Yttrium Orthosilicate (LYSO) are an established standard but are a poor spectral match to SiPM sensors with a conventional N-on-P structure. To overcome this spectral mismatch either scintillators with a peak emission in the green region of the spectrum are required or the peak detection efficiency of silicon photomultipliers needs to be shifted to shorter wavelengths. To an extent both of these goals have been achieved. The peak detection efficiency of SiPM sensors can be shifted to shorter (near-UV/blue) wavelengths by inverting the structure and forming microcells with P-on-N junctions while a new scintillator material, Cerium doped Gd₃Al₂Ga₂O₁₂ (Ce:GAGG), with a peak emission at 520nm has recently been reported. In this study we report the energy resolution of two relatively novel inorganic scintillator materials, with emission peaks at 310nm (Pr:LuAG) and 520nm (Ce:GAGG), optically coupled to both conventional green sensitive N-on-P (M series) and near-UV sensitive P-on-N SiPM sensors (B & C series).

II. EXPERIMENTAL

A. Scintillating Crystals for Medical Imaging

The scintillating crystal plays a key role in determining the performance of the PET detector system. The material choice, surface treatment and reflector type all influence

<table>
<thead>
<tr>
<th>TABLE I</th>
<th>PROPERTIES OF SCINTILLATING MATERIALS</th>
</tr>
</thead>
<tbody>
<tr>
<td>CeGAGG</td>
<td>Pr:LuAG</td>
</tr>
<tr>
<td>Light Yield (photons/MeV)</td>
<td>46,000</td>
</tr>
<tr>
<td>Decay Time (ns)</td>
<td>&lt;25</td>
</tr>
<tr>
<td>Peak Emission (nm)</td>
<td>520</td>
</tr>
<tr>
<td>Density (g/cm³)</td>
<td>6.6</td>
</tr>
<tr>
<td>Intrinsic Energy Resolution (%)</td>
<td>5.2</td>
</tr>
<tr>
<td>Effective Atomic Number (Z_eff)</td>
<td>54</td>
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</tbody>
</table>
the timing, energy and spatial resolution of the scintillator-sensor detector [9]. While cerium doped lutetium-yttrium orthosilicate, Lu$_2$(1−$x$)Y$_2$SiO$_5$, (LYSO) is the most widely used scintillator in PET systems, new materials that exceed the performance of LYSO in terms of light-yield, timing or intrinsic energy resolution are under development. Two such materials are Cerium doped Gd$_3$Al$_2$Ga$_5$O$_{12}$ (Ce:GAGG) and Praseodymium doped Lu$_3$Al$_5$O$_{12}$ (Pr:LuAG). The properties of both Ce:GAGG and Pr:LuAG are summarized in table I. For comparison the properties of Ce:LYSO are also reported.  

1) Cerium doped Gd$_3$Al$_2$Ga$_5$O$_{12}$ (Ce:GAGG): Growth of Ce:GAGG crystals by the Czochralski method and its scintillation properties were first reported in 2011 [10], [11]. The crystal has an emission peak in the 520-530nm region and is therefore well matched to the peak spectral sensitivity of SiPM detectors with an N-on-P structure. The light yield of Ce:GAGG has been reported as 33,100 photons/MeV [12] and 46,000 photons/MeV [13]. The crystal has a decay time of the order of 90ns and a density of 6.63g/cm$^3$. While Ce:GAGG is a promising scintillator for PET, it is unsuitable for MRI-compatible PET detector systems since gadolinium is paramagnetic.

2) Praseodymium doped Lu$_3$Al$_5$O$_{12}$ (Pr:LuAG): The properties of single-crystal Pr-doped Lu$_3$Al$_5$O$_{12}$ are also well-suited to medical imaging applications [14]. Crystals can be grown by the Czochralski method and have a density similar to Ce:GAGG. The light yield per MeV is approximately half that of Ce:GAGG at around 19,000 photons/MeV [15]. Pr:LuAG has an extremely fast decay time which makes this material of particular interest to body-area specific scanners such as Position Emission Mammography (PEM) scanners [16] and time-of-flight (TOF) PET applications. The crystal is also non-hygrosopic. This gives Pr:LuAG a significant advantage over Lanthanum Bromide (Ce:LaBr$_3$) which also has a fast decay time and a closer spectral match to the P-on-N structure [17]. However, Ce:LaBr$_3$ is hygroscopic and therefore requires hermetic encapsulation. Ce:LaBr$_3$ has a density of 5.06 g/cm$^3$ which is lower than that of Pr:LuAG. Pr:LuAG has a peak emission wavelength of 310nm and is a poor match to the P-on-N structure. Further details on the respective merits of P-on-N and N-on-P structures can be found in [18], [19].

In this study we assess the properties of three different SiPM devices from SensL; an N-on-P (M series) structure; a P-on-N (B series) structure and; a low-noise P-on-N (C series) structure [20]. Both the N-on-P and P-on-N processes are fabricated in a CMOS foundry. Each of the SiPM detectors consist of 4774 microcells and have a fill factor (FF) of 64%. The microcell active area is 35$\mu$m $\times$ 35$\mu$m which is lower than that of Pr:LuAG. Pr:LuAG has a peak emission wavelength of 310nm and is a poor match to the absorption spectra of both PMT and SiPM sensors.

### B. Silicon Photomultiplier Technology

Silicon photomultiplier sensors consist of a 2D array of microcells and are an ideal candidate for the optical read-out of scintillator materials for modern medical imaging techniques such as PET. Each microcell is composed of a Geiger mode Avalanche Photodiode (GAPD) in series with a passive quench resistor. The microcells are connected in parallel and have a single common output. The devices are compact, scalable and have a low material budget.

N-on-P devices typically have a peak spectral response around 500-520nm with a sharp cut-off towards shorter wavelengths. Hence this structure is a poor match to the emission spectrum of PET scintillators such as Ce:LYSO. Ce:LYSO has an emission peak at 420nm. The poor response of the N-on-P structure at short wavelengths (near-UV/blue) is mainly due to the fact that short wavelength photons have a very short penetration depth in silicon. As a result, photons in this region of the spectrum have a high probability of being absorbed in the top n$^+$ layer where the recombination rate is high and the minority carrier holes initiate the impact ionization avalanche process. Since the Avalanche Initiation Probability (AIP) of electrons, P$_e$, is greater than that of holes (P$_h$), an inverse or P-on-N structure optimizes the response at shorter wavelengths. Further details on the respective merits of P-on-N and N-on-P structures can be found in [18], [19].

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### C. Experimental Setup

The SiPM detectors and scintillating crystals were aligned using a rigid mechanical holder. The cylindrical holder has a hole for the crystal and a recess for the detector package [21]. Both the crystal and detector can be readily exchanged.

The SiPM detector bias voltage was supplied using a Keithley 2410 source-measure unit (SMU). The SMU is controlled by a LabView program for recording the current-voltage (IV) characteristics. The SiPM, crystal, source and amplifier were
housed in a Heraeus Votsch HT 4004 environmental chamber. The chamber provided a dark, temperature controlled environment for the study. The chamber temperature was monitored using a thermocouple placed in close proximity to the SiPM sensor.

Figure 1 shows a photograph of the long $3 \times 3 \times 30 \text{mm}^3$ Ce:GAGG and Pr:LuAG crystals. The crystal dimensions were chosen to cover the entire optical surface area of the SiPM detectors and match the typical crystal depth (25mm) used in modern PET scanners. The crystal faces were polished and 5 of the faces are covered in a white reflective coating by the manufacturer. The SiPM detectors were packaged with the silicon surface and wire bonds protected with an epoxy fill. Optical coupling between the epoxy layer covering the silicon and crystal surface was achieved using an optical grease. Radioactive sources were placed on top of the holder such that the predominant direction of interaction was along the long axis of the scintillator. The SiPM signal was amplified using a high bandwidth amplifier from MiniCircuits with a voltage gain of 20dB. The amplified signals were displayed on a 1GHz LeCroy oscilloscope. The oscilloscope signals were captured and transferred to a PC for further processing. The amplitude of each pulse was taken as a measure of the energy deposited inside the scintillation crystal. A minimum of 10,000 pulses was used to derive the pulse height spectrum at each gamma energy.

III. EXPERIMENTAL RESULTS

A. IV Characteristics

Figure 2 shows the IV characteristics of the three different sensor types at 20°C. The ultra-low noise performance of the C-series device is evident from the post breakdown dark current. The MicroFC device has over an order of magnitude less dark current than the MicroFB and MicroFM devices. The reduction in dark rate has been achieved through defect reduction techniques in the fabrication process. Figure 3 compares the typical microcell gain of the MicroFC and MicroFB devices. The figure shows that the gain of these two devices is similar. In the figure a fit to the IV characteristic for the MicroFB device is also shown. The pre-breakdown device leakage current is modeled using a two term exponential fit while the post-breakdown current is modeled with a parabolic fit. The quadratic dependence of the dark current on the overbias $(V - V_{br})$ is expected since the SiPM dark current is a function of both the dark rate and the gain, $G$, of the microcells. Both the dark rate and gain of the microcells have been shown to have a linear dependence on over-bias [22]. In a perfectly uniform device with identical microcells the dark current $(I_{dark})$ is given by the following expression

$$I_{dark}(V^2) = DR(V) \times G(V) \times q$$ (1)

where $DR(V)$ is the SiPM dark rate or average number of dark pulses per second, $G(V)$ is the gain of the microcells and $q$ is the electronic charge. The parabolic growth of the dark current is maintained up to about 5.4V above the breakdown voltage after which the current deviates from the model by more than 10%. The faster growth of the current than that predicted by the parabolic fit is a result of correlated noise effects such as optical cross-talk and after-pulsing [23].

The IV characteristics were measured at 4 different temperatures between -10°C and 20°C and the breakdown voltage determined at each temperature. Figure 4 shows the breakdown voltage for each of the SiPM devices as a function of temperature. The temperature dependence of the breakdown voltage of the MicroFM and MicroFB devices were found to be similar at 23mV/°C and 24mV/°C respectively. The breakdown voltage temperature coefficient of the MicroFC device was found to have a lower value of 20mV/°C.

B. Linearity of Response

1) Ce:GAGG: The linearity of the detector response as a function of the incident gamma energy $(E_{\gamma})$ is a critical performance parameter for any scintillator-sensor combination. This is especially true for high light yield scintillators such as Ce:GAGG when coupled to silicon photomultiplier sensors whose dynamic range is limited by the number of microcells [24]. Figure 5 shows the mean pulse height of the photopeak as a function of gamma energy for three $10\mu$Ci isotopes: $^{241}$Am, $^{133}$Ba and $^{137}$Cs. For each SiPM device the photopeak mean was recorded at an over-bias of 4V. The data points for
Fig. 4. SiPM breakdown voltage as a function of temperature. The breakdown voltage of the MicroFM, MicroFC and MicroFB devices have temperature coefficients of 23mV/°C, 20mV/°C and 24mV/°C respectively.

Fig. 5. Photopeak mean height as a function of gamma energy for each SiPM device when coupled to a Ce:GAGG crystal. A detector bias of 4V over the breakdown voltage was used for each device. The data for the microFM device (blue squares) is fitted with an exponential saturation curve (solid blue curve) and a linear model (excluding the 662keV data point). The MicroFC (black circles) and MicroFB devices (red diamonds) are fitted with a linear model between 59.5keV (241Am) and 356keV (133Ba).

the microFM device are fitted with an exponential saturation expression of the form,

$$P(E_\gamma) = M \cdot (1 - \exp(-N \cdot E_\gamma))$$

where $P(E_\gamma)$ is the photo-peak pulse height, $E_\gamma$ is the gamma energy in keV and $M$ and $N$ are constants. In addition, the gamma energies between 59.5keV (241Am) and 356keV (133Ba) are also fitted with a linear model which is extrapolated to 670keV. The figure shows that the microFM device, which is the best match to the Ce:GAGG emission spectrum, shows the largest saturation effect. At 662keV the deviation from linearity is 13.4%, 6.2% and 0.5% for the microFM, microFC and microFB devices respectively.

2) Pr:LuAG: Figure 6 shows the photopeak mean as a function of gamma energy for the microFC device optically coupled to a Pr:LuAG crystal. Given the smaller light yield and spectral mismatch between the detector response and the emission of the Pr:LuAG crystal, the number of photons detected by the SiPM is much less that the total number of microcells and hence the device operates in the linear region of its dynamic range.

Fig. 6. Mean pulse height of the photopeak as a function of gamma energy for the microFC device optically coupled to a Pr:LuAG crystal. The detector bias was 28.7V or 4V above the breakdown voltage. The detector response shows excellent linearity with a gradient of 1.45±0.04 mV/keV.

C. Energy Resolution

1) Ce:GAGG: The energy resolution of the scintillator-photodetector system impacts the image quality in PET through the rejection of scattered gamma photons. In this study the energy resolution is defined as the Full Width Half
Maximum (FWHM) divided by the centroid of a Gaussian fit to the photo-peak. The fit to the photopeak and Compton edge was modeled as the sum of two Gaussians. Figure 7 shows the pulse height spectrum from the microFM (N-on-P) device optically coupled to a Ce:GAGG crystal in response to 662keV gamma photons from a 137Cs source. The SiPM bias was 31.4V or 4V above the breakdown voltage. The background corrected 662keV photo-peak has an energy resolution (FWHM) of 10.4%. After correcting for the saturation shown in figure 5 the energy resolution is 11.7%. This result is comparable to that reported for shorter (5mm) Ce:GAGG crystals coupled to SiPM sensors from different manufacturers [24].

Figure 8 shows the pulse height spectrum for the low-noise microFC (P-on-N) device optically coupled to the Ce:GAGG crystal. At a detector bias of 28.7V or 4V above the breakdown voltage the 662keV photopeak has an energy resolution of 10.9%. After correcting for saturation the energy resolution is 11.6%.

2) Pr:LuAG: Due to the spectral mismatch between the emission from the Pr:LuAG crystal and the spectral response of the microFM (N-on-P) detector, it was difficult to obtain a photopeak from the 137Cs source for reasonable detector bias values. Increasing the bias on the detector improved the signal pulse but it remained difficult to resolve the photopeak from the background spectrum.

Figure 9 shows the pulse height spectrum from the MicroFC (P-on-N) device optically coupled to a Pr:LuAG crystal at a detector bias of 4V above the breakdown voltage. The background corrected 662keV photopeak has an energy resolution of 16.7%. This relatively poor energy resolution is a result of the spectral mismatch between the emission peak of the crystal (310nm) and the peak sensitivity of the detector (420nm).

D. Bias Dependence

1) Ce:GAGG: Figure 10 shows the bias dependence of the 662keV photopeak for the microFB device coupled to a Ce:GAGG crystal between 2 and 8V above the breakdown voltage. The bias dependence is modeled with a quadratic fit.

<table>
<thead>
<tr>
<th>SiPM Structure</th>
<th>Crystal</th>
<th>Uncorrected</th>
<th>Corrected</th>
</tr>
</thead>
<tbody>
<tr>
<td>MicroFM (N-on-P)</td>
<td>Ce:GAGG</td>
<td>10.4%</td>
<td>11.7%</td>
</tr>
<tr>
<td>MicroFM (N-on-P)</td>
<td>Pr:LuAG</td>
<td>-%</td>
<td>-%</td>
</tr>
<tr>
<td>MicroFB (P-on-N)</td>
<td>Ce:GAGG</td>
<td>10.6%</td>
<td>10.65%</td>
</tr>
<tr>
<td>MicroFB (P-on-N)</td>
<td>Pr:LuAG</td>
<td>16.1%</td>
<td>16.1%</td>
</tr>
<tr>
<td>MicroFC (P-on-N)</td>
<td>Ce:GAGG</td>
<td>10.9%</td>
<td>11.6%</td>
</tr>
<tr>
<td>MicroFC (P-on-N)</td>
<td>Pr:LuAG</td>
<td>16.7%</td>
<td>16.7%</td>
</tr>
</tbody>
</table>

IV. DISCUSSION

The conventional N-on-P structure of silicon photomultipliers is a poor spectral match to established scintillators such as Ce:LYSO as well as contemporary crystals such as Ce:LaBr3 and Pr:LuAG. This presents a challenge when optimizing designs for detector systems for PET scanners. The spectral match has been improved by shifting the peak response of SiPM sensors to shorter wavelengths (420nm) by inverting the junction structure and this has been shown to improve the energy resolution of Ce:LYSO-SiPM detectors [25]. With minimal optimization, energy resolution values of 16-17% have been achieved for 30mm long Pr:LuAG crystals coupled to microFB and microFC devices.

A new high light yield scintillator crystal, Ce:GAGG, with an emission spectrum well-matched to silicon photomultipliers is now commercially available. Excellent energy resolution values of the order of 10-12% were achieved for 30mm long Ce:GAGG crystals coupled to the three different types of detectors. The best energy resolution was achieved for the microFB device reflecting the fact that for high light yield crystals, saturation of the SiPM limits the energy resolution performance that can be realized. These energy resolution values are equivalent to the resolution of LYSO-PMT block detectors used in commercial PET scanners. Table III summarizes the uncorrected and saturation-corrected energy resolution values obtained for each crystal-SiPM combination.
V. Conclusion

Silicon Photomultipliers show great promise for time-of-flight PET and are beginning to be integrated into commercial systems. The adoption of silicon photomultiplier technology has the potential to significantly reduce the cost of scanners and hence increase access to patients, enhance image quality and improve quantitative assessment of the metabolic status of tumors. In addition, the compact form of the detection platform allows 1:1 coupling between crystal and sensor and facilitates the development of high-resolution targeted scanners for specific regions or organs such as the brain, prostate or breast. In this study we have shown that an energy resolution of the order 10-12% is readily achievable for long (30mm) Ce:GAGG crystals optically coupled to SiPM sensors.

The crystal Pr:LuAG is also of interest for TOF PET application due to its extremely short decay time (< 25ns). However, with an emission peak at 310nm, the crystal is a poor spectral match to P-on-N SiPM sensors. The PDE also plays a role in determining the Coincidence Time Resolution (CTR) of the scintillator-sensor combination and as a result the CTR of Pr:LuAG has been found to be significantly worse than Ce:LYSO [26]. Nevertheless, we achieved an energy resolution of 16.1% for Pr:LuAG optically coupled to a near-UV optimised microFB device.

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