Energy Resolution and Temperature Dependence of Ce:GAGG Coupled to 3 mm × 3 mm Silicon Photomultipliers

B. Seitz, N. Campos Rivera, and A. G. Stewart

Abstract—Scintillators are a critical component of sensor systems for the detection of ionizing radiation. Such systems have a diverse portfolio of applications from medical imaging, well logging in oil exploration, and detection systems for the prevention of the illicit movement of nuclear materials. The rare earth element cerium is an ideal dopant for a variety of host scintillating materials due to the fast 5d\textsubscript{1} → 4f radiative transition of Ce\textsuperscript{3+}. Cerium-doped gadolinium aluminium gallium garnet (Ce:GAGG) is a relatively new single crystal scintillator with several interesting properties. These include high light yield, an emission peak well-matched to silicon sensors, and low intrinsic energy resolution. Moreover, the material has high density and is nonhygroscopic.

In this paper, we review the properties of cerium-doped GAGG and report energy-resolution (ER) measurements over the temperature range −10°C to +50°C for 3 × 3 × 30 mm\textsuperscript{3} Ce:GAGG crystals optically coupled to a silicon photomultiplier (SiPM) sensor with a 3 mm × 3 mm active area. In addition, the linearity of the scintillator-SiPM response as a function of gamma energy is reported.

Index Terms—Cerium-doped gadolinium aluminium gallium garnet (Ce:GAGG), gamma spectroscopy, positron emission tomography (PET), scintillation detection, silicon photomultiplier.

I. INTRODUCTION

CERIUM-DOPED Gd\textsubscript{3}Al\textsubscript{2}Ga\textsubscript{3}O\textsubscript{12} (Ce:GAGG) is a relatively new single crystal scintillator with several properties that make it interesting for applications, such as gamma spectroscopy [1], [2]; alpha particle detection [3], [4]; and nuclear medicine [5]–[9]. The material was first reported in 2011 and single crystals can be grown by the Czochralski (Cz) method [10], [11]. It is the brightest of the oxide scintillators with a light yield (LY) of 46 000 photons/MeV and an emission peak at 530 nm from the 5d\textsubscript{1} → 4f radiative transition.

GAGG has no intrinsic radioactivity and is nonhygroscopic. The crystal has been studied as a suitable scintillator for the block detectors used in positron emission tomography (PET) and single-photon emission tomography (SPECT) scanners. The crystal is mechanically stable and crystals with dimensions as small as 0.4 mm × 0.4 mm × 5 mm have been reported for use in ultra-high resolution block detectors [12].

II. REVIEW OF PROPERTIES

The usual figure-of-merit values from recent studies of Ce:GAGG are summarized in Table I. The energy resolutions quoted in the table are the measured values for 662 keV gamma photons. The majority of these studies have used short crystals, either 1 or 5 mm in length, in order to minimise self-absorption and photon refraction effects in the crystal. These effects have been shown to deteriorate the energy and time resolution in longer crystals [13], [33]. All of the samples detailed in the table were grown by the Cz method except for the first one which was grown by the micropull-down method (μPD).

A. Light Yield

The LY, typically measured in response to 662 keV gamma photons, has been reported to be as high as 50 600 photons per MeV [15] and has been found to decrease with sample thickness [15], [21] and Ce dopant concentration [14]–[17]. The dependence on sample thickness is the result of the loss of photons due to self-absorption and scattering while the reduction in LY with increasing Ce content is attributed to Ce aggregate centres or crystal defects resulting from localized concentrations of Ce. The LY of Ce:GAGG is approximately 43% higher than for cerium-doped lutetium-yttrium orthosilicate (Ce:LYSO), the established standard scintillator for PET. A ceramic version of the Ce:GAGG scintillator, produced by sintering crystalline nano-micrograins into a bulk ceramic, has also been reported. The ceramic version of the scintillator is reported to have a LY of 70 000 ph/MeV [18].

B. Intrinsic Energy Resolution

The ER of a photopeak or full-energy peak is defined as the full-width half maximum (FWHM) of the peak divided by the mean value and when measured by coupling a scintillator to a detector, such as a photomultiplier tube (PMT), can be written as

\[
\frac{\Delta E}{E} = \left(\frac{\delta_{sc}}{E}\right)^2 + \left(\frac{\delta_p}{E}\right)^2 + \left(\frac{\delta_{st}}{E}\right)^2
\]

where \(\delta_{sc}\) is the intrinsic energy resolution of the crystal, \(\delta_p\) is the transfer resolution, \(\delta_{st}\) is the statistical contribution, defined
by the number of detected photons and the detector noise, and \( \Delta E/E \) is the energy resolution of the scintillation-sensor system [19]. The main contribution to the intrinsic resolution of the crystal is the nonproportionality in the number of scintillation photons generated as a function of gamma energy. The nonproportionality of Ce:GAGG has been studied by a number of groups and is on the order of 20% over the energy range 32 to 662 keV. The intrinsic ER of single crystal Ce:GAGG at a gamma energy of 662 keV is reported as 5.2 \( \pm \) 0.1% [1]. Compared with Ce:LYSO, the energy resolution of Ce:GAGG is a factor of 1.6 lower and similar to that of thallium-doped cesium iodide (Tl:CsI).

C. Decay Time

The decay of the scintillation light pulse is reported to have two components: a fast component of the order of 60–130 ns and a slow component of the order of several hundred nanoseconds (260–530 ns). In the lower range of the fast component, the decay time is comparable to the radiative lifetime of Ce\(^{3+}\) [20]. The decay time as a function of Ce concentration has been studied and found to decrease with increasing concentration [14]. The pulse decay time in the transparent ceramic version of Ce:GAGG is reported to be 165 ns [18].

III. EXPERIMENTAL

A. Cerium-Doped Gd\(_3\)Al\(_2\)Ga\(_3\)O\(_{12}\) (Ce:GAGG)

The properties of Ce:GAGG samples used in this study are summarized in Table II. For comparison, the properties of thallium-doped sodium iodide (Tl:NaI) and the oxide scintillators Bi\(_4\)Ge\(_3\)O\(_{12}\) (BGO) and Ce:LYSO are also given. Tl:NaI is one of the most widely used scintillating materials while BGO was the material of choice for early positron emission tomography (PET) scanners. Modern PET systems use Ce:LYSO. The crystals used in this study have dimensions 3 \( \times \) 3 \( \times \) 30 mm\(^3\) and were supplied by Furukawa Co. Ltd., Japan. All faces of the crystals had been polished and five faces were coated in a white reflective material. A photograph of the four crystals is shown in Fig. 1.\(^{1}\)

\(^{1}\)The Ce:GAGG crystal parameters in Table II are taken from the supplied specification sheet from Furukawa.

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### TABLE I

**Review of Properties of Ce:GAGG**

<table>
<thead>
<tr>
<th>Crystal Size (mm)</th>
<th>Ce (%)</th>
<th>LY (ph/MeV)</th>
<th>ER (@ 662keV) (%)</th>
<th>Decay (ns)</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 ( \times ) 3 ( \times ) 3</td>
<td>0.2</td>
<td>42,000</td>
<td>8.3</td>
<td>53.7</td>
<td>[20]</td>
</tr>
<tr>
<td>5 ( \times ) 5</td>
<td>1</td>
<td>46,000</td>
<td>4.9</td>
<td>88</td>
<td>[11]</td>
</tr>
<tr>
<td>3 ( \times ) 3 ( \times ) 5</td>
<td>1</td>
<td>46,000</td>
<td>7.8</td>
<td>92</td>
<td>[14]</td>
</tr>
<tr>
<td>5 ( \times ) 5 ( \times ) 10</td>
<td>1</td>
<td>47,900</td>
<td>6.8( \pm )0.2</td>
<td>-</td>
<td>[21]</td>
</tr>
<tr>
<td>10 ( \times ) 10 ( \times ) 5</td>
<td>-</td>
<td>33,000</td>
<td>6.1</td>
<td>127( \pm )6</td>
<td>[1]</td>
</tr>
<tr>
<td>5 ( \times ) 5 ( \times ) 5</td>
<td>1</td>
<td>50,600</td>
<td>5.5</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>5 ( \times ) 5 ( \times ) 10</td>
<td>1</td>
<td>41,100</td>
<td>7.3</td>
<td>-</td>
<td>[15]</td>
</tr>
</tbody>
</table>

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### TABLE II

**Properties of Scintillating Materials**

<table>
<thead>
<tr>
<th>Material</th>
<th>Ce:GAGG</th>
<th>Ce:LYSO</th>
<th>BGO</th>
<th>Tl:NaI</th>
</tr>
</thead>
<tbody>
<tr>
<td>Light Yield (photons/MeV)</td>
<td>46,000</td>
<td>32,000</td>
<td>8,000</td>
<td>40,000</td>
</tr>
<tr>
<td>Decay Time [ns]</td>
<td>90</td>
<td>41</td>
<td>300</td>
<td>230</td>
</tr>
<tr>
<td>Peak Emission [nm]</td>
<td>520</td>
<td>420</td>
<td>480</td>
<td>415</td>
</tr>
<tr>
<td>Density [g/cm(^3)]</td>
<td>6.6</td>
<td>7.1</td>
<td>7.13</td>
<td>3.67</td>
</tr>
<tr>
<td>Intrinsic ER [%] (662keV)</td>
<td>5.2</td>
<td>8</td>
<td>12</td>
<td>6.6</td>
</tr>
<tr>
<td>Z(_{eff})</td>
<td>54</td>
<td>66</td>
<td>75</td>
<td>51</td>
</tr>
</tbody>
</table>

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B. Silicon Photomultiplier Sensor

The silicon photomultiplier sensor consists of a 2-D array of microcells [22]–[24]. Each microcell consists of a photon counting or Geiger-mode avalanche photodiode (GAPD) in series with a passive quenching element. For each detected photon, the microcell emits a pulse of current and is considered to be a digital device. However, since all of the microcells are connected in parallel to a single output, the summed output forms an analog output in which the total emitted charge is proportional to the number of incident photons detected.

The SiPM is an ideal sensor for the optical read-out of scintillating materials and its compact size and form factor allow for the design of highly granular detectors with one-to-one coupling between the scintillator and the sensor. Additional benefits of the SiPM sensor include ease of operation, inherent immunity to interference by strong magnetic fields, and a low operating voltage. Another key feature is its compatibility with modern semiconductor manufacturing processes (CMOS), which allows the sensors and processing electronics to be combined on a single chip.

C. Experimental Setup

To provide a dark, temperature-controlled environment, the scintillator-SiPM detector, amplifier, and radioactive source were placed inside a Heraeus Votsch 4004 environmental chamber. The chamber was used to vary the temperature of the scintillator-SiPM detector between \(-10\) and \(+50^\circ\)C. Optical coupling between the uncoated scintillator facet and the SiPM sensor was achieved using Dow Corning 20–057 (\(n = 1.48\)) optical coupling compound [5].
The SiPM used in this study was a 3 mm × 3 mm MicroFC sensor from SensL [26]. While the MicroFC is not the best spectral match to Ce:GAGG, the peak optical response of the MicroFC occurs at 420 nm compared with the emission peak of 520 nm for Ce:GAGG, the device has significantly lower noise performance (dark rate) than similar devices with peak sensitivity in the green [27]. The MicroFM has a peak response of 500 nm and is therefore a better spectral match to Ce:GAGG. The MicroFC consists of 4774 microcells and has a fill factor of 64%. The typical characteristics of this sensor are summarized in Table III.

The SiPM signal was amplified using a high-bandwidth amplifier from Mini-Circuits. The amplified signals were displayed on a 1 GHz LeCroy (LC574AL) oscilloscope. The detected signals were captured and transferred from the oscilloscope to a PC for further processing. The amplitude of each signal pulse was taken as a measure of the energy deposited by the gamma photon inside the crystal. A minimum of 10,000 pulses were used to generate the pulse height spectrum for each gamma energy.

### IV. EXPERIMENTAL RESULTS

#### A. IV Characteristics

Fig. 2 shows the reverse bias portion of the IV characteristic of a 3 mm × 3 mm MicroFC sensor recorded at 20 °C and −10°C. The IV characteristic was used to determine the breakdown voltage of the GAPD junction [28]. The breakdown voltage is defined as the bias voltage corresponding to the peak in the second derivative of the IV curve [29]. The IV characteristic was recorded at several temperatures between −10 and 20°C and the breakdown voltage determined at each temperature. Fig. 3 shows the breakdown voltage as a function of temperature. The breakdown voltage decreases monotonically with decreasing temperature and has a temperature coefficient of 20 mV/°C.

#### B. Detector Response Linearity

As described before, a silicon photomultiplier consists of a limited number of single-photon sensitive microcells. The detection of a photon, assuming a uniform spatial distribution, is a statistical process based on the probability that the photon is absorbed within the sensitive volume of a microcell and the probability that the photo-generated electron or hole initiate an avalanche breakdown of the GAPD [30]. In addition, the microcells have a finite recovery time during which the microcells can be considered insensitive to photons. The dynamic range of an SiPM is therefore a function of the number of microcells and the sensor photon detection efficiency (PDE). For an instantaneous light pulse, the number of detected photons can be approximated by the expression

\[
N_d = N_{MC} \cdot \left( 1 - \exp \left( -\frac{\eta \cdot N_{ph}}{N_{MC}} \right) \right)
\]

where \( N_d \) is the number of detected photons, \( N_{ph} \) is the number of incident photons, \( \eta \) is the SiPM PDE, and \( N_{MC} \) is the total number of microcells [31]. This expression gives an approximately linear response when the number of detected photons \( (\eta \times N_{ph}) \) is much less than the total number of microcells \( (N_{MC}) \). However, the response begins to saturate as the number of detected photons approaches the number of microcells. Hence there is a tradeoff between the geometry, the dimensions, and number of microcells for a given area, the PDE, and the dynamic range. A more complex model of the response of an
Fig. 4. Photopeak mean height as a function of gamma energy for Ce:GAGG optically coupled to the SiPM sensor at 20°C. The data show the mean pulse height at SiPM bias values of 2 V (green squares), 3 V (black diamonds), 4 V (red circles), and 5 V (blue squares) over the breakdown voltage. The data at 4 and 5 V above the breakdown voltage are fitted with an exponential saturation curve (equation (2)) and a linear model (excluding the 662 keV data point) to show the deviation from linearity at higher energies.

SiPM sensor that takes into account the effects of the recovery time, afterpulsing, and crosstalk has also been developed [32].

Fig. 4 shows the photopeak mean height as a function of gamma energy at 2, 3, 4, and 5 V above the SiPM breakdown voltage. The measurements were recorded at 20 °C using $^{241}$Am, $^{133}$Ba, and $^{137}$Cs sources. At 2 and 3 V above the breakdown voltage, the response shows good linearity with increasing gamma energy. At 4 and 5 V above the breakdown voltage, the data sets are fitted with an exponential saturation model. In addition, the gamma energies between 60 keV and 356 keV are fitted with a linear model which is extrapolated to 680 keV to show the deviation from linearity. At 5 V above the breakdown voltage and at 662 keV, the response of the detector is approximately 6.5% below that expected from a linear trend.

C. Detector Pulse Decay Time

Fig. 5 shows the decay time of the detector pulse from 662 keV gamma photons at 20 °C. The pulse shape was averaged for 2000 pulses using the oscilloscope and transferred to a PC for analysis. The decay time was obtained by fitting the pulse with a single exponential with a decay constant $\tau$ of 191 ns.

V. ENERGY RESOLUTION

Fig. 6 shows the pulse height spectrum for the scintillator-SiPM detector in response to 662 keV gamma photons ($^{137}$Cs) at 20 °C. The spectrum is modeled as the sum of two Gaussian distributions—one fitted to the Compton edge of the spectrum and one fitted to the photopeak. The energy resolution is defined as the FWHM divided by the centroid of the Gaussian fit to the photopeak. The 662 keV photopeak in Fig. 6 has an energy resolution of 10.2 ± 0.5%. After correcting for the saturation effect, described in section IV B, the energy resolution is 10.5%. Extrapolating the ER dependence on the crystal length, given in [21], to 30 mm gives an energy resolution of approximately 10.3% in good agreement with our result.

A. Bias Dependence

The energy resolution as a function of SiPM bias is shown in Fig. 7. The figure shows the energy resolution for 662 eV gamma photons as a function of SiPM bias recorded at 20 °C. Both the uncorrected and corrected energy resolution values are displayed. The corrected values were determined by measuring the SiPM response, as described in section IV B, at each bias value. At high bias values, the figure shows that the energy resolution (corrected) begins to saturate at about 4 V above the breakdown voltage.
The SiPM PDE, gain, and noise (dark rate, crosstalk, and afterpulsing) are all functions of the applied bias, and the relationship between these terms and the bias determines the bias dependence of the energy resolution. For relatively large optical signals, such as those for high LY scintillators, the bias dependence of the PDE largely determines the response of the energy resolution. As a function of the applied bias, the PDE initially increases rapidly, resulting in a reduction in the energy resolution as the photopeak mean increases faster than the variance. As the bias dependence of the PDE begins to level off, the energy resolution also begins to saturate assuming that the SiPM is operating in the linear region of its response.

### B. Temperature Dependence

The temperature dependence of the energy resolution measured at 662 keV and at a constant overbias of 4 V above the breakdown voltage is shown in Fig. 8. A second Ce:GAGG crystal to that used for the room temperature measurements was used to characterize the temperature dependence of the crystal. The energy resolution values are uncorrected for the effect of saturation of the SiPM response, and the y errorbars are calculated from the one sigma confidence intervals from the FWHM and centroid parameters of the Gaussian fit. At 662 keV, the main contribution to the energy resolution, (1) comes from the statistical and transfer terms assuming an intrinsic energy resolution for Ce:GAGG of 5.2%.

Fig. 9 shows the 662 keV photopeak mean, measured at both a constant overbias (4 V above the breakdown voltage) and a constant bias, as a function of temperature. At a fixed bias, the photopeak mean decreases approximately linearly with increasing temperature. This is a direct result of the temperature dependence of the breakdown voltage of the SiPM and the resulting reduction in the sensor overbias. As the sensor overbias is reduced, the PDE and gain of the sensor decrease and, hence, the photopeak mean decreases. For a fixed overbias, the photopeak mean is relatively constant over the temperature range $-10\,^\circ\text{C}$ to $30\,^\circ\text{C}$. Above $30\,^\circ\text{C}$, the photopeak mean was observed to decrease by 10.6% between $30\,^\circ\text{C}$ and $50\,^\circ\text{C}$. This temperature dependence is similar to that reported in [25]. This reduction is likely to be the result of thermal quenching of the radiative transition in Ce$^{3+}$.

### VI. DISCUSSION

Cerium-doped GAGG has a high light yield and excellent intrinsic energy resolution that make it a promising scintillator material for a range of applications including medical imaging. With minimal optimization, an energy resolution of 10.5% was achieved when optically coupled to a 3 mm $\times$ 3 mm SiPM sensor at room temperature. Below 30 $^\circ\text{C}$, the light output of the scintillator is relatively constant while above 30 $^\circ\text{C}$, the thermal quenching of the Ce$^{3+}$ transition was observed.

While the light yield and intrinsic energy resolution of Ce:GAGG is superior to Ce:LYSO, the longer decay time and
lower atomic number remains a drawback for the adoption of the Ce:GAGG in modern PET scanners. The lower atomic number results in reduced gamma detection efficiency and, hence, a longer scan time would be required to generate the required statistics compared with Ce:LYSO crystals with similar axial dimensions. The longer decay time increases the probability of pile-up effects.

VII. CONCLUSION

Cerium-doped GAGG is a promising scintillating crystal for a number of applications, including medical-imaging modalities such as PET and SPECT. The crystal has the highest LY of the oxide scintillators, low intrinsic energy resolution, and relatively fast timing properties. A room temperature energy resolution, corrected for the effect of saturation, of 10.5% was recorded for a long 3 × 3 × 30 mm3 crystal coupled to a 3 × 3 mm3 SiPM sensor. The measured energy resolution reported here is higher than that previously reported for short Ce:GAGG crystals (see Table I) and is likely to be due to self-absorption, crystal impurities, and surface reflection effects which increase with crystal length and result in photon transmission losses. The energy resolution is constant over the temperature range −10 to 50 °C while the mean of the photopeak was observed to decrease at temperatures greater than 30 °C. The reduction in the number of detected photons above 30 °C is consistent with thermal quenching of the Ce3+ transition.

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REFERENCES