Nuclear Instruments and Methods in Physics Research A 629 (2011) 157–169

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

High energy gamma-ray spectroscopy with $LaBr_3$ scintillation detectors

F.G.A. Quarati [a,b,*], Alan Owens [a], P. Dorenbos [b], J.T.M. de Haas [b], G. Benzoni [c], N. Blasi [c], C. Boiano [c], S. Brambilla [c], F. Camera [c,d], R. Alba [e], G. Bellia [e], C. Maiolino [e], D. Santonocito [e], M. Ahmed [f], N. Brown [f], S. Stave [f], H.R. Weller [f], Y.K. Wu [g]

[a] SRE-PA, ESA/ESTEC, Keplerlaan 1, 2200AG Noordwijk, The Netherlands
[b] Delft University of Technology, I.R.I., Mekelweg 15, 2629 JB Delft, The Netherlands
[c] Istituto Nazionale Fisica Nucleare sez. di Milano, Via Celoria 16, 20133 Milano, Italy
[d] Università di Milano, Dipartimento di Fisica, Via Celoria 16, 20133 Milano, Italy
[e] Laboratori Nazionali del Sud, Istituto Nazionale Fisica Nucleare, via S. Sofia 62, 95125 Catania, Italy
[f] Triangle Universities Nuclear Laboratory (TUNL), Box 90308, Durham, NC 27708-0308, USA
[g] The Duke Free Electron Laser Laboratory (DFELL), Box 90319, Durham, NC 27708-0319, USA

ARTICLE INFO

Article history:
Received 11 July 2010
Received in revised form
12 October 2010
Accepted 5 November 2010
Available online 18 November 2010

Keywords:
Lanthanum halide
$LaBr_3$
Brillance
Scintillation detector
Gamma ray spectroscopy
BepiColombo
Mercury
Remote sensing

ABSTRACT

Lanthanum bromide scintillation detectors produce very high light outputs (~60,000 ph/MeV) within a very short decay time (typically ~20 ns) which means that high instantaneous currents can be generated in the photocathode and dynode chain of the photomultiplier tube (PMT) used for the scintillation readout. The net result is that signal saturation can occur long before the recommended PMT biasing conditions can be reached.

In search of an optimized light readout system for $LaBr_3$, we have tested and compared two different PMT configurations for detection of gamma-rays up to 15 MeV. This range was chosen as being appropriate for gamma-ray remote sensing and medium energy nuclear physics applications. The experiments were conducted at two facilities: the Laboratori Nazionali del Sud (LNS) in Catania, Italy [1] and the High Intensity Gamma-ray Source (HIγS) at Triangle University Nuclear Laboratory, in Durham, North Carolina, USA [2].

The PMT configurations we have tested are (1) a standard dynode chain operated under-biased; (2) a 4-stage reduced chain operated at nominal inter-dynode bias.

The results are that shortening the number of active stages, as in configuration (2), has advantages in preserving energy resolution and avoiding PMT saturation over a large energy range.

However, the use of an under-biased PMT, configuration (1), can still be considered a satisfactory solution, at least in the case of PMTs manufactured by Photonis.

The results of this study will be used in support of the Mercury Gamma-ray and Neutron Spectrometer (MGNS) on board of BepiColombo, the joint ESA/JAXA mission to Mercury, scheduled for launch in 2014.

1. Introduction: lanthanum bromide for space exploration

Lanthanum bromide scintillation detectors, discovered at Delft University of Technology and the University of Bern [3], owe their development into large volume detectors to a European Space Agency (ESA) project for their application in space exploration. As part of the development, a comprehensive assessment of their radiation tolerance was undertaken [4,5], after which it was decided to fly a $LaBr_3$ scintillation detector on the joint ESA/JAXA mission to Mercury: BepiColombo. For the mission, a 3 in. × 3 in. $LaBr_3$ gamma-ray spectrometer will remotely sense Mercury by detecting gamma-ray lines emanating from natural and activated surface elements. More information on this particular application of $LaBr_3$ spectrometers can be found in Refs. [6–8]. The energy range of the instrument needs to extent over the nuclear transition region (150 keV–10 MeV) in order to successfully investigate the chemical composition of Mercury's shallow surface.

We have tested our detectors up to 15 MeV, exceeding the instrument's dynamic range by nearly a factor of two which is standard pre-flight calibration practice to avoid uncertainty encountered at the end of the detection range. Specifically, if we could demonstrate the operation of $LaBr_3$ spectrometers with a proportional response at nearly twice the required energy range, we gain a safe margin to avoid any deterioration in response due to instrument aging during the cruise and subsequent operation in space.

We also need to guarantee the ability to quickly and effectively calibrate the spectrometer once in orbit around Mercury. For $LaBr_3$

---

[*] Corresponding author at: SRE-PA, ESA/ESTEC, Keplerlaan 1, 2200AG Noordwijk, The Netherlands. Tel.: +31 71 565 8530; fax: +31 (0)71 565 5985.
E-mail addresses: F.G.A.Quarati@tudelft.nl, fquarati@rssd.esa.int (F.G.A. Quarati).

0168-9002/$ - see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.nima.2010.11.035

we can use its self-activity as a calibration source, in particular its ^{138}La line at 1470 keV [9]. This line offers a very reliable calibration point because of its peculiar shape. However, we can effectively use this line to calibrate the instrument's entire dynamic range only if the response is linear. Experiments have shown that $LaBr_3$ spectrometers potentially suffer from photomultiplier tube (PMT) saturation [10–12], which could substantially affect the ease to which this calibration can be carried out.

In this article, we first examine the problem of signal saturation occurring in $LaBr_3$/PMT assemblies and review possible solutions. We then report on the measurements and results of high energy gamma-ray detection and conclude by assessing operating limits and further implementations.

The work is mainly focused on testing 3 in. × 3 in. $LaBr_3$ detectors, although results obtained with a smaller 2 in. × 2 in. $LaBr_3$ detector are also reported.

The main conclusion of this study is that $LaBr_3$ material is entirely suitable for inclusion, not only on the BepiColombo mission to Mercury, but also to the Jovian system as recently proposed by the ESA/NASA Laplace mission.

2. PMT signal saturation with $LaBr_3$ and possible solutions

2.1. Origin and evidence of PMT saturation

Among scintillation detectors, $LaBr_3$ offers superior energy resolution mainly because of its high light output of $\sim$60,000 ph/MeV. Its scintillation process is also very fast, with $1/e$ decay time of 16 ns [10,13]. Light output and decay time directly contribute to the current flowing into the PMT, photocathode and dynode chain, which forms the detection signal.

For scintillator/PMT assemblies the anode peak current I_a can be approximately expressed as

$$I_a = \frac{\text{Light output} \times \text{Quantum efficiency} \times e \times \text{Gain}}{\text{Decay time}} \tag{1}$$

where e is the electron charge. With reference to the literature, e.g. Refs. [14,15], I_a can be evaluated for the most common scintillation detectors. This is summarised in Table 1, clearly showing that $LaBr_3$ surpasses all other scintillation detectors in terms of photocathode and anode peak currents. Actually, below a certain scintillation decay time, it is a combination of scintillation decay and PMT transit time spread which determines I_a intensity [16]. A typical value of transit time spread for box and grid PMTs can be taken to be 10 ns [17]. This value summed with the scintillation decay time has been used for a second evaluation of I_a for the "faster" scintillators with decay time <50 ns. The two values of I_a reported represent the lower and upper evaluation limits.

$LaBr_3$ represents a bright and fast source of light pulses which has not been available before with other scintillation detectors, so that new techniques and tailored PMTs are needed to optimize spectroscopic performance.

Till date, efforts in improving scintillator performances have concentrated more on increasing the photocathode quantum efficiency (QE), see Hamamatsu "Ultra Bialkali" [18–20] and Photonis "Clarity" [21,22]. An independent overview for these new PMTs is available from Mirzoyan et al. [23]. Recently, photocathodes with QE > 40% have been advertised by both Hamamatsu and Photonis. However, beside its benefit for energy resolution through enhanced statistics, an increased QE also increases the current flowing in the photocathode and the dynode chain, which exacerbates saturation problems.

In proton measurements at the Kernfysisch Versneller Instituut in Groningen, The Netherlands [24], we obtained evidence of saturation, as shown in Fig. 1. Two different setups have been used to detect protons with energies from 8 to 90 MeV. The first setup is a standard $LaBr_3$/PMT assembly; in the second we have introduced a neutral density filter between crystal and photocathode to attenuate the light yield by a factor of 15. The proton energy range has been achieved using calibrated layers of degraders, i.e. aluminium plates that reduce the primary beam energy, 90 MeV, by a known amount; a similar technique was used for the proton irradiations [4]. Because the energy degradation process is stochastic, the intrinsic beam energy spread increases after degradation affecting the energy resolution evaluation. At the highest energy, 90 MeV, we have measured a FWHM of 1.1% with the attenuated setup and 0.8% with the unattenuated setup. The latter value is corrected for the non-proportional response, having carried out its evaluation in the energy scale, while the higher value measured with the attenuated setup is consistent with the loss of photons in the filter. Both values could be affected by the intrinsic beam energy spread.

As seen in Fig. 1, a linear proportionality is only achieved with the attenuated setup, meaning that the non-proportional response observed when collecting all the scintillation light is due to saturation of either the photocathode, the dynode chain, or both.

For the unattenuated setup we have included three gamma-ray lines in Fig. 1 after correcting for the difference in ionization of protons and gamma-rays in $LaBr_3$. In a parallel experiment, we have estimated the proton ionization to be 0.76 ± 0.03 times less than that of gamma-rays. The three gamma-ray lines are the 511 keV annihilation line, the 1470 keV from $LaBr_3$ self-activity, and the 2.27 MeV decay line from proton activated Al. We cannot report the same gamma-ray lines for the attenuated setup because of the degraded energy resolution, caused by light attenuation. The data acquired with the unattenuated configuration clearly show that

Table 1

Comparison of photocathode and anode peak currents generated in the most common scintillation detectors for 1 MeV absorbed gamma-ray. The anode peak currents (I_a) are evaluated assuming a PMT gain of 2.5×10^5, typical for 8-stage PMTs.

	Wavelength max emission (nm)	Light yield (phe/keV)	Decay time (ns)	Typical photocathode QE (%)	Photocathode current per absorbed MeV (μA)	Anode peak current (I_a) per absorbed MeV (mA)
NaI:Tl	415	38	250	30	0.0073	1.82
CsI:Tl	550	54	1000	7	0.0006	0.15
CsI:Na	420	41	630	30	0.0031	0.77
CsI[a]	315	2	16	17	0.0034	0.85 (0.52)
BGO	480	10	300	21	0.0011	0.28
BaF_2 (fast)[a]	220	1.8	0.8	1	0.0036	0.9 (0.07)
BaF_2 (slow)	310	10	630	15	0.0004	0.09
$LaCl_3$:Ce[a]	350	49	28	27	0.0745	18.6 (13.7)
$LaBr_3$:Ce[a]	380	63	16	30	0.1890	47.3 (29.1)
LYSO[a]	420	32	41	29	0.0368	9.21 (7.4)

[a] For these scintillation crystals, we report in brackets the anode peak current value evaluated using PMT transit time plus scintillation decay time.