

Energy Resolution of Scintillation Detectors with Large Area Avalanche Photodiodes and Photomultipliers Light Readout

M. Moszyński¹⁾, IEEE, Member, M. Kapusta^{1, 2)}, D. Wolski¹⁾,
M. Szawlowski³⁾, IEEE, Member, W. Klamra⁴⁾,

¹⁾ Soltan Institute for Nuclear Studies, PL 05-400 Świerk-Otwock, Poland,

²⁾ University of Warsaw, Institute of Experimental Physics, Hoża 69, PL 00-681 Warsaw, Poland,

³⁾ Advanced Photonix, Inc. 1240 Avenida Acaso, Camarillo, CA 93012, USA,

⁴⁾ Royal Institute of Technology, Department of Physics, Frescati, S-104 05 Stockholm, Sweden,

Abstract

Energy resolution of small NaI(Tl), CsI(Tl), BGO, GSO, YAP and LSO crystals was studied using 16 mm diameter large area avalanche photodiodes (LAAPD) and a 52 mm diameter photomultiplier. The best result of 4.8% for the 662 keV γ -rays from a 137Cs source was obtained for a 9 mm in diameter by 9 mm high CsI(Tl) scintillator coupled to an LAAPD. Measured number of primary electron-hole pairs produced in the LAAPD and photoelectrons in the photomultiplier, as well as, noise contribution of the LAAPD, allow for quantitative discussion of the results. It has been shown that the energy resolutions measured with LAAPDs are comparable, or significantly better (at certain emission wavelengths), than those obtained with the photomultiplier. We observed that at energies, above 100 keV, the energy resolution measured with majority of crystals and the LAAPDs was weakly affected by photodiode noise contribution. The advantages and limitations of the LAAPDs in energy spectrometry with scintillation detectors are also discussed here.

I. INTRODUCTION

A growing interest in applications of avalanche photodiodes in physics experiments [1] and nuclear medicine [2] prompted us to study energy resolution obtainable with different scintillators in γ -ray spectroscopy. This was stimulated by the excellent energy resolution obtained in the previous study [3] using a YAP crystal coupled to a beveled-edge LAAPD produced recently by Advanced Photonix, Inc. (API) [4,5]. The energy resolution of 5.9% measured for the 662 keV γ -rays from a 137 Cs source was better than that observed with the Philips XP2020Q photomultiplier. The study confirmed high quantum efficiency and a low noise of the LAAPDs. It is worth adding that the LAAPDs allowed getting also a very good time resolution of 570 ps, as measured with an LSO crystal [6], for γ -rays from a 60 Co source [7].

The aim of this work was to study energy resolution of various scintillators coupled to LAAPDs and the XP2020Q PMT. Since the energy spectrometry with the PMT light readout is well known, the data on the figures are presented for LAAPDs only. The energy resolution is defined here as the full width at half maximum (FWHM) of the peak in the pulse height spectrum divided by its mean position. Measured numbers of electron-hole pairs (e-h) and photoelectrons determined for the LAAPDs and PMT, respectively, as well as noise contribution of the LAAPD

make possible a quantitative discussion of the observed energy resolution. In conclusions, the advantages and limitation of the LAAPD in energy spectrometry with scintillation detectors are discussed.

II. EXPERIMENTAL DETAILS

All the measurements were done with small NaI(Tl), CsI(Tl), BGO, GSO:Ce, YAP:Ce and LSO:Ce crystals. The crystals were wrapped with several layers of white Teflon tape, except for the NaI(Tl) crystal, which had been assembled by the manufacturer. An overview of all the crystals used is given in Table 1.

Table 1
Tested scintillators

Crystal	Ce conc. [mol%]	Size [mm]	Manu- facturer	Surface finish
NaI(TI)	-	Ø10x10 Amcrys-H		ground
CsI(TI)	-	Ø9x9 Bicron		ground
BGO	•	Ø9x9	Ø9x9 Bicron	
GSO		10x10x5	Russia	ground
LSO	0.22%	4x5x14.5	Russia	polished
YAP	0.56	10x10x5	Preciosa	ground

The crystals were fitted to the XP2020Q PMT [8] and to three 16 mm diameter LAAPDs produced by API [5]. The XP2020Q PMT no. 40978, with the quantum efficiency of 25.6% at 400 nm, operated with a type C voltage chain, as recommended by the manufacturer [8]. The main characteristics of the diodes under tests are listed in Table 2. These blue enhanced LAAPDs feature a high gain of 140 (at about 2600 V bias voltage) and a low dark current below 300 nA. High quantum efficiency of about 70% at 400 nm is another excellent feature of these LAAPDs, which is invaluable when working with fast scintillators.

In our experiments, signal from LAAPDs was fed to an Ortec 142AH preamplifier and then to a TC244 spectroscopy amplifier. For all the crystals, an optimization of the signal shaping time constant was carried out. This operation is particularly important for such slow scintillators as CsI(Tl) or BGO

Table 2
The main parameters of the tested LAAPDs

Model	630-70-73-510				
Serial No.	103-1-3 103-1-6		105-23-3		
Diameter	16 mm				
Window	none				
Q.E. at 400 nm	69 % 68%		77 %		
Gain at 2600 V	132 140		130		
Dark current	269.4 nA 219.4 nA 110.0 nA		110.0 nA		
Capacitance	110 pF				
. Rise time	14.7 ns 16.2 ns 11 ns				

In the measurements with the PMT, the signal collected across 1 $M\Omega$ was passed to an Ortec 113 scintillation preamplifier and then to a Tennelec TC244 spectroscopy amplifier.

III. RESULTS

A. Number of Electron-Hole Pairs and Photoelectrons

The number of electron-hole pairs in the LAAPDs was measured by comparing the 662 keV full energy peak position from a ¹³⁷Cs source detected in the scintillator to that of the 16.6 keV X-rays from a ⁹³Mo source detected directly by the LAAPD [3]. The number of photoelectrons in the XP2020Q was determined by a standard method [9] comparing the 662 keV peak position from a ¹³⁷Cs source with that of the single photoelectrons. All the measurements were carried out at 3 μs shaping time constant in the amplifier.

Table 3
Number of e-h pairs and photoelectrons
measured with LAAPD*) and XP2020Q PMT

Crystal	LAAPD [e-h/MeV]	XP2020Q [phe/MeV]	Hamamatsu PD [e-h/MeV]
CsI(TI)	37000±1100	3840±110	36800±740
NaI(TI)	26900±790	8900±260	•
BGO	5300±155	980±40	4750±140
GSO	6670±195	1800±53	4770±140
LSO	21000±615	5490±160	18300±360
YAP	11500±340	4270±125	immeasurable

Serial No 105-23-3

Table 3 summarizes the results of the measurements carried out for all tested crystals with one of the LAAPD (Ser. No 105-23-3) and XP2020Q PMT. For comparison, in the last column the numbers of e-h pairs measured with the Hamamatsu pin photodiodes (S3590-03 and S2744-03) for the tested crystals are

listed too, according to Ref. [9]. All the photoelectron numbers compare well with those reported recently in Ref. [9]. In contrast, a high number of e-h pairs was measured with the GSO and LSO crystals, and it was significantly higher than that observed with the Hamamatsu pin photodiodes [9]. Note also that in case of the YAP crystal its emission at 365 nm was immeasurable with the Hamamatsu devices [9]. All that confirmes a high quantum efficiency of the studied LAAPDs in the blue region of light.

B. Energy Resolution Study

The main part of the energy resolution study was carried out for the 662 keV γ-rays from a ¹³⁷Cs source. To get the best possible energy resolution, the gain of the LAAPD and the shaping time constant on the amplifier were optimized for each scintillator. The LAAPD gain of 100 was chosen finally to get the lowest possible contribution of the noise. The shaping time constant was optimized depending on the decay time of the light pulse. It was equal to 0.25 μs for LSO, GSO and YAP, which feature a fast decay time constant below 50 ns, and as much as 6 μs for CsI(Tl) crystal. In the measurements with PMT a shaping time constant of 1 to 3 μs were chosen.

TUKAN PC-based multichannel analyzer was used to record all the energy spectra. The FWHM and position of the full-energy peak corrected for the background were determined by a computer program. To increase the accuracy of the measured quantities all measurements were made three times and the mean values were used for a further discussion.

Fig. 1 presents the energy spectrum of γ -rays from a 137 Cs source measured with the CsI(Tl) crystal. Note the excellent energy resolution of 4.8%, one of the best ever observed with scintillation detectors. Energy threshold is below KX-ray peak of 31 keV reflecting a large dynamic range of measured energies. The same test done with the XP2020Q PMT showed the energy resolution of 6.6%.

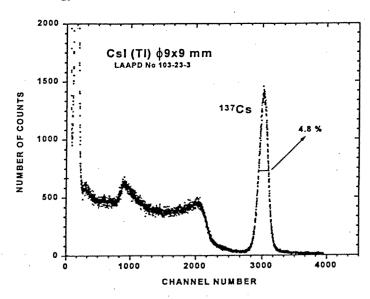


Figure 1: Energy spectrum of γ -rays from a ¹³⁷Cs source measured with a Csl(Tl) crystal coupled to a LAAPD.

Fig. 2 presents energy spectra observed with some other crystals.

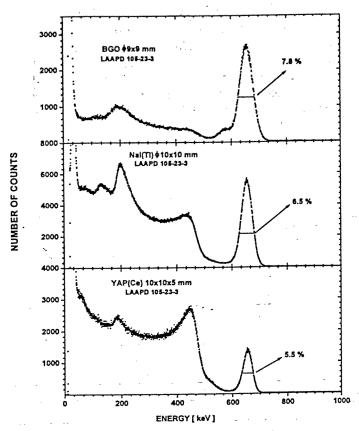


Figure 2: Energy spectra of γ-rays from a ¹³⁷Cs source measured with NaI(Tl), BGO and YAP crystals.

In Table 4, the results of the determined energy resolution for all the studied crystals are presented, as measured with LAAPD and PMT, respectively. The shaping time constants used for each test are listed too. Proper choice of shaping time constant was particularly important for the measurements carried out with the LAAPD.

Table 4 Energy resolution for 662 keV γ -rays from a 137 Cs source measured with the LAAPD and XP2020Q PMT

	LAAPD		PMT	
Crystal	τ [μς]	ΔΕ/E [%]	· τ [μs]	ΔΕ/E [%]
CsI(Tl)	6	4.8±0.14	3	6.5±0.19
NaI(TI)	1	6.5±0.19	1 ~	6.6±0.19
BGO	0.75	7.8±0.23	1	10.2±0.20
GSO	0.25	7.4±0.22	1	8.5±0.25
LSO	0.25	10.6±0.31	1	10.0±0.29
YAP	0.25	5.5±0.16	1.	5.7±0.17

It is worth noting that energy resolution measured with the

LAAPD light readout is better than those observed with the PMT for all tested crystals, except for NaI(Tl) and LSO. These crystals, however, are known to exhibit a large contribution of an intrinsic resolution [10]. A better energy resolution is particularly evident for the crystals with a long wavelength emission as CsI(Tl) and BGO, but it is also pronounced for GSO and YAP with the peak emission at 420 nm and 365 nm, respectively. Energy resolutions listed in the Table 4, particularly those measured with the LAAPD belong to the best published elsewhere.

Energy resolution, $\Delta E/E$, measured with the PMT light readout can be presented quantitatively as:

$$(\Delta E/E)^2 = (\delta_{sc})^2 + (\Delta N/N)^2 \tag{1}$$

where δ_{sc} represents the intrinsic resolution of the crystal and $\Delta N/N$ the photoelectron statistic contribution.

The intrinsic resolution of a crystal is connected with many effects such as inhomogeneities in the scintillator causing local variations of the light output, non-uniform reflectivity of the reflecting covering of the crystal, as well as recently reported the non-proportional response of the scintillator [10]. The contribution of the photoelectron statistics can be calculated as:

$$\Delta N/N = 2.36 \times 1/N^{1/2} \times (1+\epsilon)^{1/2} \tag{2}$$

where N is the number of photoelectrons and ε is the variance of the electron multiplier gain, typically equal to 0.1 for the XP2020Q PMT [10].

Table 5
Energy resolution and its components for the XP2020Q PMT

Crystal	Phe number ^{a)}	ΔΕ/Ε [%]	ΔN/N. [%]	δ⊭ [%]	δ _{sc} [10] [%]
CsI(TI)	2540±74	6.6±0.19	4.91 ±0.07	4.4 ±0.35	3.9±0.4
Nal(Tl)	5890±172	6.6±0.19	3.22 ±0.05	5.8 ±0.25	5.7±0.2
BGO	650±26	10.2±0.20	9.72 ±0.13	3.1 ±1.7	4.2±0.6
GSO	1190±35	8.5±0.25	7.17 ±0.10	4.6 ±0.60	4.2±0.5
LSO	3630±106	10.0±0.29	4.11 ±0.06	9.1 ±0.35	6.6±0.4
YAP	2830±56	5.7±0.17	4.66 ±0.05	3.3 ±0.37	3.5±0.5

a) for 662 keV and the optimal shaping, see Table 4.

Based on the eq. (1) one can estimate contribution of the intrinsic resolution of the crystals as the geometrical difference of the measured energy resolution and that from the photoelectron statistics. Thus, to discuss quantitatively the energy resolution measured with the PMT it is sufficient to know the number of photoelectrons and to calculate the contribution of photoelectron statistics according to eq. (2). Results of the analysis are presented in Table 5.

The last two columns of Table 5 contain the intrinsic energy resolution of scintillators calculated according to this study and the same quantity as listed in Ref. [10], respectively. Note a good agreement, except for LSO. The LSO crystals, however, are known to show energy resolution limited by the quality of the crystals [3,11]. The data reported in Ref. [10] are exceptional in this respect.

In case of avalanche photodiodes, the energy resolution, $\Delta E/E$, of the full energy peak measured with a scintillator coupled to an APD can be expressed as:

$$(\Delta E/E)^{2} = (\delta_{sc})^{2} + (\Delta N/N_{e-h})^{2} + (\Delta_{noise}/N_{e-h})^{2}$$
 (3)

where $\Delta N/N_{e-h}$ represents the e-h pair statistic contribution and Δ_{noise}/N_{e-h} the noise contribution.

The statistical accuracy of the signal from an APD is affected by the excess noise factor, F, reflecting the statistical fluctuation of the APD gain, as follows:

$$\Delta N/N_{eh} = 2.36 (F/N_{eh})^{1/2}$$
 (4)

where N_{e-h} is a number of primary electron-hole pairs.

In good beveled-edge APDs the excess noise factor of 2 to 2.5 is observed [12] depending on the APD gain:

$$F = k_{eff}M + (2 - 1/M)(1 - k_{eff})$$
 (5)

where k_{eff} is a weighted average ratio of the electron and hole ionization rates equal to about 0.0017 [12], while M is the APD gain.

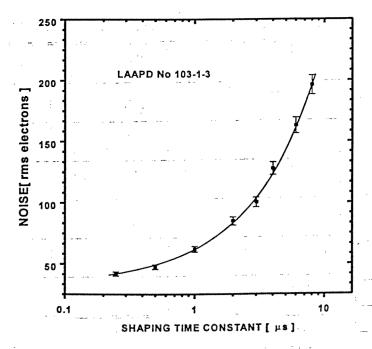


Figure 3: Noise of the LAAPD system versus shaping time constant of the spectroscopy amplifier.

The noise contribution was measured by a standard method of the test pulser hooked up to a preamplifier. The charge injected by the pulser, expressed in the number of e-h pairs, was calibrated using response from a measured scintillator to the 122 keV γ -peak from a ⁵⁷Co source. Fig. 3 shows noise contribution

of an LAAPD versus shaping time constant of the spectroscopy amplifier measured at the LAAPD gain of 100. Note a very low noise of just 42 rms electrons at 0.25 µs shaping.

The results of the analysis of the energy resolution measured with the LAAPD light readout are summarised in Table 6. To evaluate statistical contribution of e-h pairs, according to eq (4), the excess noise factor, F, of 2.1 was assumed. Noise contribution, based on the curve in Fig. 3 and expressed as a percentage fraction in FWHM for 662 keV energy, was calculated, as follows:

$$\Delta_{\text{noise}}/N_{\text{e-h}} = 2.36 \,\sigma_{\text{noise}}/N_{\text{e-h}} \tag{6}$$

where σ_{noise} is noise expressed in rms electrons (Fig. 3).

Table 6
Energy resolution and its components for LAAPD light readout

	·				
Crystal	e-h number ^{a)}	ΔΕ/E [%]	ΔN/N _{e-h} [%]	$\Delta_{\text{noise}}/N_{\text{e-h}}$ [%] ^{b)} .	δ₅c [%]
CsI(TI)	27800 ±813	4.8±0.14	2.05 ±0.03	1.38 ±0.04	4.1±0.19
NaI(TI)	17150 ±500	6.5±0.19	2.61 ±0.04	0.94 ±0.027	5.9±0.24
BGO	3310 ±100	7.8±0.23	5.94 ±0.10	4.1 ±0.12	3.0±1.2
GSO	3910 ±114	7.4±0.22	5.47 ±0.08	2.54 ±0.074	4.3±0.56
LSO	13900 ±406	10.6±0.3	2.90 ±0.87	0.87 ±0.025	10.2±0.5
YAP	7654 ±223	5.5±0.16	3.9 ±0.06	1.30 ±0.038	3.7±0.33

a) for 662 keV, measured with the optimal shaping, see Table 4

Table 6 seems to explain a better energy resolution observed with APD than that measured with PMT. The contribution of e-h pair statistic is up to about factor of 2 lower than those found for PMT (see Table 5). This is due to high quantum efficiency and the low excess noise factor of LAAPDs. The latter is a big advantage of beveled-edge APDs. Noise contribution is low and, except for the BGO, has a week influence on the measured energy resolution. The result of the analysis is further justified by the same intrinsic resolution of scintillators found with LAAPDs as those for PMT (see Table 5). That confirms also high accuracy of all the measurements.

The study shows also that energy resolution is strongly limited by the intrinsic energy resolution of the crystals for scintillators with a high light output, such as CsI(TI), NaI(TI) and LSO. In this respect, a good energy resolution of the YAP crystal is due to one of the lowest contribution of the intrinsic resolution [3,10,13].

The experiments presented above were carried out for 662 keV γ-rays from a ¹³⁷Cs source. Consequently, the study characterizes scintillation detectors with LAAPD light readout

b) for 662 keV and optimal shaping.

mainly in the high-energy γ -rays domain.

Fig. 4 shows the energy resolution of various crystals measured in 30-1200 keV energy range of γ -rays. The slope of the curves for energies below 100 keV is steeper suggesting a larger importance of a noise contribution. It is particularly evident for the CsI(Tl) crystal. Its best energy resolution measured for energies above 100 keV is getting significantly poorer for low energies, and it is starting to be even somewhat worse than thet measured with the NaI(Tl) crystal. Note, that the measurements with the CsI(Tl) were carried out with a 6 μ s shaping, while for the NaI(Tl) a 1 μ s shaping was used. Thus, the absolute noise contribution was lower by a factor of about 2.5 (see Fig. 3) in measurements with NaI(Tl).

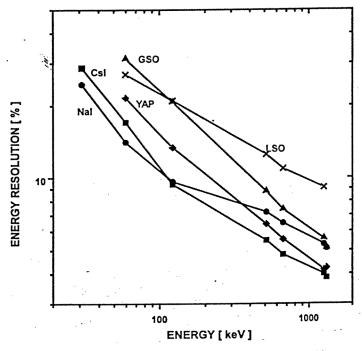


Figure 4: Energy resolution versus γ -ray energy measured with some crystals.

This effect is even more pronounced in Fig. 5 which presents the dependency of the energy resolution on the γ -ray energy measured with the NaI(Tl) crystal coupled to an LAAPD. The thin continuous lines reflect noise contribution and a statistical spread in the signal. Fig. 5 shows that for energies above 100 keV noise contribution (61 rms electrons) has negligible influence on the measured energy resolution. This is in fact mainly controlled by the intrinsic energy resolution of the NaI(Tl) estimated to be equal to 5.9% for 662 keV γ -rays (see Tables 5 and 6), well in agreement with Ref. [10]. In the low energy region, particularly for the 31 keV X-ray peak from a 137 Cs source, the energy resolution is strongly affected by the noise.

Fig. 6 presents similar dependencies for the YAP crystal. In spite of the fact that the e-h number for the YAP is much lower than that measured for NaI(Tl) (see Table 3), the noise contribution for high energies is also almost negligible. This is due to the 0.25 µs optimal shaping. It assures the lowest noise

contribution of 41 rms electrons (see Fig. 3), and a full charge collection for the crystal with the decay time constant of 26 ns [13]. This is an advantage of the new fast Ce doped scintillators in the work with avalanche photodiodes. Finally, a very good energy resolution obtained with the YAP crystal for energies above 500 keV is due to a low intrinsic resolution of this crystal (see Tables 5 and 6, and Refs. [3], [10] and [13]).

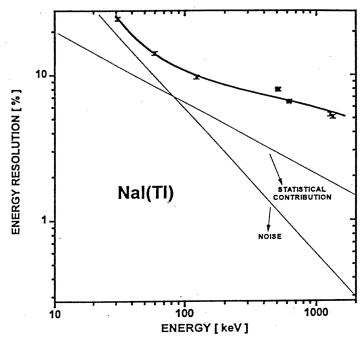


Figure 5: Energy resolution versus γ -ray energy measured with a NaI(Tl) crystal. The thin continuos lines reflect noise contribution and statistical spread of the signal.

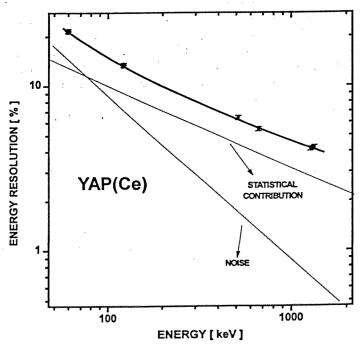


Figure 6: Energy resolution versus γ-ray energy measured with a YAP crystal. The thin continuos lines reflect noise contribution and statistical spread of the signal.

Although the presented data on energy resolution were based on the measurements carried out mainly with the LAAPD ser. no 105-23-3, similar results were obtained for the remaining LAAPDs studied in the course of this work.

IV. CONCLUSIONS

The presented study has shown excellent performances of the new large area avalanche photodiodes developed recently by Advanced Photonix, Inc., in application to scintillation detection. This is reflected by a significantly better, or comparable, energy resolution obtained with LAAPDs for a number of different scintillators, to those observed with the XP2020Q photomultiplier. A high number of e-h pairs obtained with various scintillators allows to get a better statistical accuracy of the detected light and almost negligible contribution of the noise for γ -rays with energy above 100 keV. For lower energies, the noise contribution affects the resolution and makes its somewhat worse than that observed with PMTs.

Due to high quantum efficiency of LAAPD, number of e-h pairs is up to 2.5 times higher than the photoelectron number observed with the XP2020Q PMT. The beveled-edge structure of the studied LAAPDs assures also a low estimated excess noise factor of 2.1.

A low noise level of just 41 rms electrons at 0.25 µs shaping allows to get a good energy resolution also with new fast Ce doped scintillators having a lower light output than the classical CsI(Tl) and NaI(Tl) crystals.

The analysis of the energy resolution measured with the PMT and LAAPD light readout allows calculating the intrinsic resolution of the tested crystals. It shows a good agreement for both PMT and LAAPD, as well as, when compared to the recent data reported in Ref. [10]. It is worth noting that for the scintillators with a high light output such as CsI(Tl), NaI(Tl) and LSO the intrinsic resolution is the main limiting factor for the energy resolution. The results of the analysis confirmed also a good accuracy of all the measurements. In addition, the estimation of the noise excess factor for the LAAPD at 2.1 turned out to be very well fitted with the values reported in [12].

V. ACKNOWLEDGMENTS

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VI. REFERENCES

[1] I. Holl, E. Lorenz, S. Natkaniec, D. Renker, C. Schmeltzer, B. Schwartz, "Some studies of

- Avalanche Photodiode Readout of Fast Scintillators", IEEE Trans. on Nucl. Sci. 42(1995)351.
- C. Schmelz, S.M. Bradbury, I. Holl, E. Lorenz, D. Renker, S. Ziegler, "Feasibility Study of an Avalanche Photodiode Readout for a High Resolution PET with nsec Time Resolution", IEEE Trans. on Nucl. Sci. 42(1995)1080.

[2]

- [3] M. Moszyński, M. Kapusta, D. Wolski, M. Szawlowski, W. Klamra, "Blue Enhanced Large Area Avalanche Photodiodes in Scintillation Detection with LSO, YAP and LuAP Crystals", presented at IEEE Nucl. Sci. Symp. 1996, Anaheim, USA, IEEE Trans. on Nucl. Sci. 44(1997)436.
- [4] M.J. Szawlowski, S. Zhang, A. De Cecco, M. Madden, M. Lindberg and E. Gramsch, "Performances of Large Area Avalanche Photodiodes", IEEE Nucl. Sci. Symp. 1992, Conference Records, p. 239.
- J.C. Boisvert, J.T. Montroy, L. Jostad, B. Zhou, M. Szawlowski, "Improved Large-Area Avalanche Photodiodes for Scintillation Detection in Calorimetry", presented at IEEE Nucl. Sci. Symp. 1996, Anaheim, USA, Conference Records, p. 16.
 C.L. Melcher and J.S. Schweitzer, "Cerium-doped
- [6] C.L. Melcher and J.S. Schweitzer, "Cerium-doped Lutetium Oxyorthosilicate: A Fast, Efficient New Scintillator", IEEE Trans. on Nucl. Sci. 39(1992)502.
 [7] M. Moszyński, T. Ludziejewski, D. Wolski, W. Klamra, M. Szawlowski, M. Kapusta, "Subnanosecond Timing
- with Large Area Avalanche Photodiodes and LSO Scintillator", IEEE Trans. on Nucl. Sci.43(1996)1298.

 [8] Philips Photonics, Photomultiplier Catalog, 1994.
- [9] M. Moszyński, M. Kapusta, M. Mayhugh, D. Wolski, S.O. Flyckt, "Absolute Light Output of Scintillators", presented at IEEE Nucl. Sci. Symp. 1996, Anaheim, USA, IEEE Trans. on Nucl. Sci. 44(1997)1052.
- [10] P. Dorenbos, J.T.M. de Haas, C.W.E. van Ejik, "Non-proportionality in the Scintillation Response and the Energy Resolution Obtainable with Scintillation Crystals", IEEE Trans. on Nucl. Sci. 42(1995)2190.
- [11] T. Ludziejewski, K. Moszyńska, M. Moszyński, D. Wolski, W. Klamra, L.O. Norlin, E. Devitsin, V. Kozlov, "Advantages and Limitations of LSO scintillator in Nuclear Physics Experiments", IEEE Trans. on Nucl. Sci. 42(1995)328.
- [12] Advanced Photonix, Inc. Application Note, "Noise Characteristics of Advanced Photonix Avalanche Photodiodes",
- [13] M. Moszyński, M. Kapusta, D. Wolski, W. Klamra and B. Cederwall, "Properties of the YAP:Ce scintillator" Nucl. Instr. and Meth., in press.