Energy Resolution and Nonproportionality of ZnSe:Te scintillator studied by large area avalanche photodiodes and photomultipliers.[1]

M. Balcerzyk[a,*], W. Klamra[b], M. Moszyński[a], M. Kapusta[a], and M. Szawlowski[c]

[a]*Soltan Institute for Nuclear Studies, 05-400 Otwock, Poland*
[b]*Royal Institute of Technology, Department of Physics, Frescati S-104 05 Stockholm, Sweden*
[c]*Advanced Photonix, Inc. 1240 Avenida Acaso, Camarillo, CA 93012, USA,*

Abstract:

The ZnSe:Te scintillator has been studied by means of a photomultiplier with bialkali photocathode, Large Area Avalanche Photodiode (LAAPD) and PIN photodiode. The light output was determined as 28300±1700 photons per MeV. The crystals showed a high proportionality for light output versus energy dependence, as observed for gamma radiation. The energy spectrum for the 662 keV gamma line recorded using the system of LAAPD gave an energy resolution of 5.4%. This results in intrinsic energy resolutuion of 3.3%. We have observed also the increase of the values of *intrinsic* energy resolution with lowered shaping time constant and anomalously high influence of Te on proportionality curve and on energy spectra.

Keywords: ZnSe doped with Te, nonproportionality, X-ray detector, energy resolution

[*] Corresponding Author: Marcin.Balcerzyk@ipj.gov.pl, tel. ++48 22 7180547, fax ++ 48 22 7793481

[1]This work was supported in part by Polish Committee for Scientific Research (KBN) grants 8 T11E 025 15 and 8T 10C 005 15 and Swedish Institute.

1. Introduction

Growing interest in the development of new scintillation detectors has prompted efforts to better understand the limitations of achievable energy resolution in γ-ray spectroscopy [1-3]. It is known that the energy resolution is controlled by a statistical spread of photoelectrons or electron-hole pairs (e-h) produced by scintillating light in photomultipliers or photodiodes, respectively [2]. Excess in the measured energy resolution is frequently called the intrinsic resolution of the scintillator [2]. The intrinsic resolution is connected with many effects such as inhomogeneities in the scintillator, causing local variations of the light output, non-uniform reflectivity of the reflecting layer covering of the crystal, as well as, the non-proportional response of the scintillator [1].

Recently, special attention has been paid to the non-proportionality of the light output of scintillators versus the energy of γ-rays [1,3-11] and its influence on the measured energy resolution [7-10]. This effect is expected to be of the great importance for large volume crystals when a full-energy peak is mostly a result of multiple Compton scattering [2]. In case of small crystals, when a full-energy peak is mainly due to photoelectric absorption, the expected effect should be much weaker [8]. The study of a number of new Ce doped crystals showed, however, evident correlation between the intrinsic resolution and non-proportionality, even for small volume crystals of fraction of 1 cm^3 [10]. A small YAP:Ce ($YAlO_3$:Ce) crystal exhibited an exceptionally good energy resolution of 4.35±0.11% for 662 keV γ-rays from a ^{137}Cs source and its intrinsic resolution of 1.3±0.5% was determined in Ref. [11]. The relative light output of the YAP was constant within ±2% for energies between 14.4 keV and 1.275 MeV.

The origin of the non-proportionality of scintillators is inadequately understood. There are indications that scintillators containing light elements, as YAP:Ce, are better in this respect. However, a comparative study of LSO, GSO and YSO showed the same dependence of non-proportionality, independently of a chemical composition of the crystal [12]. The aim of the present work was to study the energy resolution and non-proportionality of ZnSe:Te crystal. According to the earlier works [13] it exhibits a very high light output of 80000 ph/MeV. Thus, we have expected a high accuracy of evaluation of the intrinsic resolution. Both, ZnSe:Te and YAP:Ce crystals contain only light elements thus, so the aim is to check the non-proportionality, overall and intrinsic energy resolution of ZnSe:Te.

Previously the increase of the values of intrinsic energy resolution with the lowering shaping time constant was observed for CsI(Tl) [9]. The light pulse of CsI(Tl) has two exponential components of scintillation pulse shape (0.679 μs – 91% of light and 3.34 μs – 9% of light) [14]. We wanted to test that dependency for a system with only one exponential component of scintillation pulses, for which decay time constant is in the range of standard commercial spectroscopy amplifiers.

The first report on use of ZnSe as a X-ray detector was published by Ryzhikov *et al* [15]. The first use of tellurium doped ZnSe as scintillator was reported by Vakulenko *et al.*[16]. The luminescence mechanism was reported by Ryzhikov *et al.* [17]. A more detailed bibliography can be found in Ref. [18]. ZnSe is a low-density crystal (5.42 g/cm^3). The emission wavelength of ZnSe:Te from ISC Kharkov, Ukraine is 610 nm. It is not suitable for photomultiplier (PMT) detection, therefore we decided to check ZnSe:Te properties by green extended bialkali PMT and Large Area Avalanche Photodiodes (LAAPD). ZnSe:Te has refractive index n = 2.6 [19] that neither well matches refractive index of glass window of PMT nor n of sili-

con at that wavelength (n = 3.87 [20] and reflectance R = 0.34 [21]). Ryzhikov *et al.* reported light output L of ZnSe:Te as high as 80000 photons per MeV (ph/MeV) at X-ray excitation [12]. Schotanous *et al.* [22] reported only 1300 photoelectrons per MeV (phe/MeV) measured with Hamamatsu R1017 PMT, having 5% QE at 610 nm and 12000 e-h pairs per MeV with photodiode, both at 12 μs shaping time. It is worthy to add that zinc has K absorption line at 9.65 keV and selenium at 12.66 keV, namely both energies are below 20 keV, usual cutoff of energy for 70 kV peak voltages X-ray tube used in dental radiography [23]. Thus it can be applied for this purpose.

2 Experimental setup

The crystals of ZnSe:0.2%Te were delivered from STC Institute for Single Crystals, Kharkov, Ukraine in two samples (1) 10×10×1 mm and (2) ∅10×5 mm. They were characterized by the producer as containing only fast component of the luminescence (i.e. 1 - 3μs decay time constant). The samples were of deep yellow and deep brown colors respectively. Sample (1) was polished on both large surfaces and sample (2) on one round surface, other being grounded only. The crystals were glued onto PMT, PIN photodiode or LAAPD with silicone grease of unknown refractive index – estimated around n = 1.5.

For light output and energy resolution measurements a 51 mm diameter ADIT B51D01 PMT with extended bialkali photocathode was used. ADIT PMT has a quantum efficiency (QE) of 25% at 400 nm, 10% at 550 nm, 4% at 610 nm and 2% at 640 nm. 16 mm diameter windowless large area avalanche photodiode (LAAPD) from Advanced Photonix, Inc. was used. It is characterized by a QE of 90% for 610 nm and 640 nm. Detailed technical characteristics of LAAPD can be found in Table 1 and elsewhere [24]. We have also used 10×10 mm Hamamatsu silicon PIN diode S3590-08 with photosensitivity of 0.42 A/W at 610 nm [25]. The rest of experimental setup consisted of a charge sensitive preamplifier EG&G ORTEC preamplifier model 142AH and Tennelec Spectroscopy Amplifier model 244 or EG&G ORTEC 672 Spectroscopy Amplifier. PC-based multichannel analyzer Tukan, produced at SINS, was used to record energy spectra.

3 Results

3.1. Light output

To determine the light output of tested crystals the number of photoelectrons per energy unit (phe/MeV) was measured with PMT and the number of e-h pairs ($N_{e\text{-}h}$, e-h/MeV) was measured with LAAPD. With PMT, it was done by comparing the position of the 662 keV full energy peak from a ^{137}Cs source with that of the single photoelectron. The photoelectron number for sample (1) was measured as 1280±40 phe/MeV at 10μs shaping time. We have folded the light emission spectrum at 610 nm as published by the manufacturer with standard PMT QE curve. The calculation yields the light output of 29000±2900 photons/MeV (ph/MeV).

The number of e-h pairs produced by the ZnSe:Te crystal in LAAPD was measured by comparing the position of the 662 keV γ-peak detected in the scintillator with that of 16.6 keV X-rays from a ^{93}Mo source detected directly by the LAAPD. The measurements were carried out at the LAAPD gain of 50 and shaping time constant of the amplifier of 10 μs. The measure-

ment with LAAPD with QE of 90% at 610 nm yielded 26400±1500 e-h pairs/MeV, that is 29300±3000 ph/MeV.

Similar method was used for measurement with PIN diode. This was performed at 12 μs shaping time and yielded 23900±1200 e-h pairs /MeV for sample 1 (1 mm thick) and 19700±1000 e-h pairs /MeV for sample 2 (5 mm thick). To obtain a photon yield one has to apply the same procedure as described in [28]. The quantum yield of the photodiode must be corrected for the light reflected from diode surface. The corrected value of QE is 90% compared to 75% before correction. The resulting photon yields are 26500±2700 ph/MeV and 21900±2200 ph/MeV respectively. Manufacturer quotes self-absorption constant for scintillation light as 0.15 cm^{-1}. The lower light output for thicker sample suggests that scintillation light undergoes not only self-absorption but that it also travels a much longer path than the crystal thickness, because of internal reflections and reflection from photodiode surface.

Having three results from different measurements for light output from ZnSe:Te we conclude that it is 28300±1700 ph/MeV with the long time component of unknown, but considerable contribution.

Note consistent results of the light outputs given in photons/MeV for PMT and LAAPD with refractive indices not matched. However, the measured light output of ZnSe:Te is much lower than the 80000 ph/MeV quoted by Ryzhikov *et al.* [12]. In contrast, the photoelectron number measured with PMT is very close to that reported by Schotanous *et al.* in [22]. A very approximate inspection of the light pulse shape done by single photon method seems to show that the light pulse consist of a dominating decay component of 2.7±0.3 μs and a much longer one with a decay well above 10 μs. It is not integrated out by the longest shaping times of 10 μs in the spectroscopy amplifier. It may suggest that the total light output of ZnSe:Te can be very easily much larger than that reported in this work.

Proportionality curve defined as photoelectron yield measured at specific energy of excitation relative to the photoelectron yield at 662 keV is shown in Fig. 1. ZnSe:Te is a quite proportional scintillator down to 85% at 5.9 keV. Note a dip in proportionality curve at 30 keV, close to the K absorption line of Te (31.8 keV). The same unusual dip is observed for CaF_2:Eu at 48.5 keV [26]. The value of the dip (more then 5% in proportionality for ZnSe:Te) cannot be accounted by simple additional absorption of dopant of the heavy element.

We have observed a shoulder (Fig. 2) in the energy spectrum of ^{57}Co source (122.06 keV line) which can be attributed, by energy difference, to Te escape peak. The $K_{\alpha 1}$, $K_{\alpha 2}$, $K_{\beta 1}$ emission lines of Te are at 27.4723; 27.2017; 30.9957 keV respectively. The relative intensity of this shoulder suggests that Te dopant interacts with fast electrons generated by γ-rays more intensively than it is expected just from its relative abundance. This and above observations are in good agreement.

3.2. Energy resolution

Fig. 3 shows the energy spectra of the 662 keV γ-rays from a ^{137}Cs source measured for ZnSe:Te crystal (sample 1) coupled to the Hamamatsu PIN photodiode S3590-08 at 12 μs shaping time (a), the ADIT B51D01 PMT (b), and to the LAAPD (c) at 10 μs shaping time constant in the spectroscopy amplifier. The energy resolution of 10.5±0.6% and 9.5±0.5% observed with PMT and PIN photodiode respectively is rather poor, while the same spectrum measured with LAAPD light readout shows an excellent energy resolution of 5.4±0.3%. In the

case of the measurement with PMT, a large statistical error of photoelectrons in PMT and poor separation of the full energy peak from a Compton spectrum limits the observed energy resolution. The latter effect is a result of the low efficiency for a full energy peak in the tested crystal. It is associated with the light elements (Zn, Se) of the crystal and its small volume. In contrast, about 20 times larger number of e-h pairs obtained with LAAPD than photoelectrons for PMT measurement, reduces dramatically the statistical error and improves the energy resolution. The good energy resolution, as observed at the long shaping time constant, is limited by the noise of LAAPD associated with its dark current. It is of particular importance at low γ-rays energies, since the noise contribution is inversely proportional to the energy. A more precise study could be done with a cooled LAAPD reducing the noise contribution of LAAPD at long shaping time constants. For PIN photodiode a large contribution of noise at 12 μs worsens the energy resolution.

The energy resolution, ΔE/E, measured with a scintillator coupled to a photomultiplier can be quantitatively represented as [1]:

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

where: δ_{sc} is the intrinsic resolution of the crystal and ΔN/N the photoelectron statistical contribution.

The contribution of photoelectron statistics can be written:

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

where N is the number of photoelectrons and ε is the variance of the electron multiplier gain, equal to 0.15 for the ADIT B51D01 PMT, as was determined analyzing the width of the single photoelectron peak.

In the case of LAAPD, the energy resolution is further affected by the noise contribution of the diode and preamplifier [7]:

$$(\Delta E/E)^2 = (\delta_{sc})^2 + (\Delta N/N)^2 + (\Delta_{noise}/N)^2 \qquad (3)$$

where $(\Delta_{noise}/N)^2$ is the noise contribution and the statistical contribution of e-h pairs. According to the following equation, it can be expressed as:

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

where F is the excess noise factor of APD equal to 2.16 at gain 100.

The overall energy resolution and the statistical contribution to the energy resolution for ZnSe:Te (sample 1) is shown in Fig. 4 as a function of energy, as measured with ADIT PMT at 10 μs shaping time. It confirms that the energy resolution measured with PMT light readout is mainly described by the statistical error of photoelectrons. No doubt that emission spectrum of the ZnSe:Te crystal with peak value at 610 nm weakly fit to PMTs even with a green extended bialkali photocathode.

The contribution of the intrinsic energy resolution can only be calculated based on the measurement with the LAAPD. The overall energy resolution for 662 keV γ line of 5.4±0.3% allows calculating the intrinsic resolution of 3.3±0.7%. It shows that a good proportionality of the ZnSe:Te light output, presented in sect. 3.1, is reflected also in a low intrinsic resolution. It is better presented in section 3.3 where the intrinsic resolution was measured versus shaping time constant in the amplifier.

3.3. Shaping time dependencies

For the first time the peculiarity of intrinsic energy resolution dependence on shaping time of the spectroscopy amplifier (i.e. the fraction of integrated light) was observed by Moszyński *et al.* [9] for CsI(Tl) for measurement with LAAPD and by Fiorini et al. [27] for CsI(Tl) coupled to silicon drift chamber. Variation of δ_{sc} for 662 keV γ excitation for ZnSe:Te is shown in Fig. 5 for measurement with LAAPD at gain 100. The contribution of noise, statistics and overall resolution are also shown. Note that the dependence on e-h pair (or photoelectron) statistics and LAAPD noise contribution are accounted by their respective contribution to the overall energy resolution. The noise contribution expressed in the percent fraction is a function of the absolute noise contribution and the number of e-h pairs integrated by different shaping time constants shown in Fig. 5(a) (see eq. (3)). Thus it shows a flat distribution on Fig. 5 (b). It is worthy to add that the effect of the intrinsic resolution dependency on a fraction of integrated light was also observed for CsI(Tl) crystal measured with XP2020Q PMT. It excludes hypothesis that it is connected with the unidentified properties of silicon photosensors. The reason for this dependence is presently not understood.

4 Discussion

The performed study showed that ZnSe:Te crystal is a scintillator with a high light output of 29000±2000 ph/MeV, as measured with 10 μs shaping time constant. This quantity is, however, much lower than 80000 ph/MeV quoted in Ref. [12]. The difference is attributed to the long component of the light pulse with the decay time constant much longer than 10 μs, which was only partially integrated in the amplifier. The high light output of ZnSe:Te is not employable in the measurements with PMTs, since at peak emission of 610 nm, the low quantum efficiency of PMTs limits its application. The long decay time constant of about 3 μs of the primary light pulse requires also the use of a long shaping time constants of about 10 μs in the amplifier, which seriously limits the counting rate capability of the detector. The best energy resolution of 5.4±0.3% was measured with the avalanche photodiode in spite of the fact that the long shaping time constant is far from that optimal one for the noise contribution.

ZnSe:Te is a scintillator with a good proportionality of the light output. It is followed by an excellent energy resolution and a low intrinsic resolution, as measured for 662 keV γ-line. No doubt that a further improvement can be expected with a better technology of crystal production. It confirms that the good intrinsic resolution of scintillators is due to good proportionality of the crystals.

References

[1] P. Dorenbos, J.T.M. de Haas, C.W.E. van Eijk, IEEE Trans. Nucl. Sci. NS-42(1995)2190.

[2] G.F. Knoll, „Radiation Detction and Measurements“, Third Edition, John Willey and Son, New York, 2000.

[3] P. Dorenbos, J.T.M. de Haas, C.W.E. van Eijk, C.L. Melcher, J.S. Schweitzer, IEEE Trans. on Nucl. Sci. 41(1994)735.

[4] P. Dorenbos, J.T.M. de Haas, C.W.E. van Eijk, 1994 IEEE Nucl. Sci Symp. Norfolk, USA, Conference Records, p. 304.

[5] J.D. Valentine, B.D. Rooney, Nucl. Instr. and Meth. A353(1994)51.

[6] B.D. Rooney, J.D. Valentine, IEEE Trans. Nucl. Sci. 44(1997)509.

[7] M. Moszyński, M. Kapusta, D. Wolski, M. Szawlowski, W. Klamra, IEEE Trans. Nucl. Sci. 45 (1998) 472.

[8] J. Valentine, B. D. Rooney, J. Li, IEEE Trans. Nucl. Sci. 45(1998)512.

[9] M. Moszyński, M. Kapusta, J. Zalipska, M. Balcerzyk, D. Wolski, M. Szawlowski, W. Klamra, IEEE Trans. Nucl. Sci. 46(1999)243.

[10] M. Balcerzyk, M. Moszyński, M. Kapusta, Proceedings of The Fifth International Conference on Inorganic Scintillators and Their Applications SCINT99 August 16-20, 1999 Moscow State University, 119899 Moscow, Russia, p.167-173.

[11] M. Kapusta, M. Balcerzyk, M. Moszyński, J. Pawelke, Nucl. Instr. Meth. A421 (1999) 610.

[12] M. Balcerzyk, M. Moszyński, M. Kapusta, D. Wolski, J. Pawelke, C.L. Melcher, IEEE Trans. Nucl. Sci., 47 (2000)1324-8.

[13] V.D. Ryzhikov, Yu. A. Borodenko, S.N. Galkin, L.P. Galchinetskii, E.A. Danshin, K. A. Katrunov, E.M. Selegnev, V.I. Silin, Proceedings of The International Conference on Inorganic Scintillators and Their Applications SCINT95 August 28 – September 1, 1995 Delft, The Netherlands (1996) 465.

[14] J.D. Valentine, D.K. Wehe, G.F. Knoll, C.E. Moss, IEEE Trans. Nucl. Sci. 40 (1993) 1267.

[15] V.D. Ryzhikov, O.S. Shapiro, S.M. Ignatov, V.I. Silin, Pribory i Tekhnika Eksperimenta, **29** (1986) 155.

[16] O.V. Vakulenko, A.N. Veretennikov, V.D. Ryzhikov, V.V. Chepelev, Zhurnal Tekhnicheskoi Fiziki **58** (1988) 632.

[17] V.D. Ryzhikov, V.I. Silin, N.G. Starzhinsky, Nuclear Tracks & Radiation Measurements **21** (1993) 53.

[18] http://www.ipj.gov.pl/~balcerzm/dowziecia/OVID_ZnSe_Te.htm (27 papers).

[19] STC Institute for Single Crystals, Kharkov, Ukraine Product Catalog. See also http://mail.isc.kharkov.com/index.html.

[20] C.M. Herzinger, B. Johs, W.A. McGahan,. J.A. Woollam. W. Paulson, J. App. Phys. **83** (1998) 3323. See also D. E. Aspnes, A. A. Studna, Phys. Rev. B **27** (1983) 985

[21] K. Navratil, J. Sik, J. Humlicek, S. Nespurek Optical Materials **12** (1999) 105

[22] P. Schotanus, P. Dorenbos V.D. Ryzhikov IEEE Transactions Nuclear Science, vol.39, no.4, Aug. 1992, pp.546-50.

[23] C. Fröjdh , J. Andersson , R. Bates , M. Heuken, R. Irsigler, C.S. Petersson ,V. O'Shea, K. Smith, H. Stamatakis, U. Welander *Nucl. Instr. Meth* **A434** (1999) 18;
see also: http://www.ipj.gov.pl/~balcerzm/dowziecia/X-ray_dental_source_spectrum.gif

[24] Advanced Photonix Inc. LAAPD catalogue and http://www.advancedphotonix.com/windowle.htm.

[25] Hamamatsu catalog and http://www.hpk.co.jp/hp2e/products/ssd/PDE/CeraPINE.htm
[26] D.W. Aitken, B.L. Beron, G. Yenicay, H.R. Zulliger, IEEE Trans. Nucl. Sci. **NS-14** (1967) 468.
[27] C. Fiorini, A. Longoni, F. Perotti, C. Labanti, P. Lechner, L. Struder IEEE Trans. Nucl. Sci. 44 (1997) 2553.
[28] M. Moszyński, M.Kapusta, M. Mayhugh, D. Wolski, S.O. Flyck IEEE Trans. Nucl. Sci. 44 (1997) 1052-61.

Figure captions:

Fig. 1 Proportionality of light output of ZnSe:Te in units of light output at 662 keV

Fig. 2 Energy spectra of γ-rays from a ^{57}Co source measured with ZnSe:Te crystal (sample 1) coupled to the LAAPD at 6 μs shaping time constant in the spectroscopy amplifier

Fig. 3 Energy spectra of 662 keV γ-rays from a ^{137}Cs source measured with ZnSe:Te crystal (sample 1) coupled to Hamamatsu PIN photodiode S3590-08 at 12 μs shaping time (a), the ADIT B51D01 PMT (b), and to the LAAPD (c) at 10 μs shaping time constant in the spectroscopy amplifier

Fig. 4 Energy resolution of ZnSe:Te for sample (1) measured with PMT

Fig. 5 (a) Light yield of ZnSe:Te in units of the number of e-h pairs at 662 keV vs. shaping time. (b) Energy resolution $\Delta E/E$; intrinsic energy resolution, δ_{sc}; statistical contribution to energy resolution $\Delta N/N$, and noise contribution to energy resolution Δ_{noise}/N of ZnSe:Te at 662 keV versus shaping time.

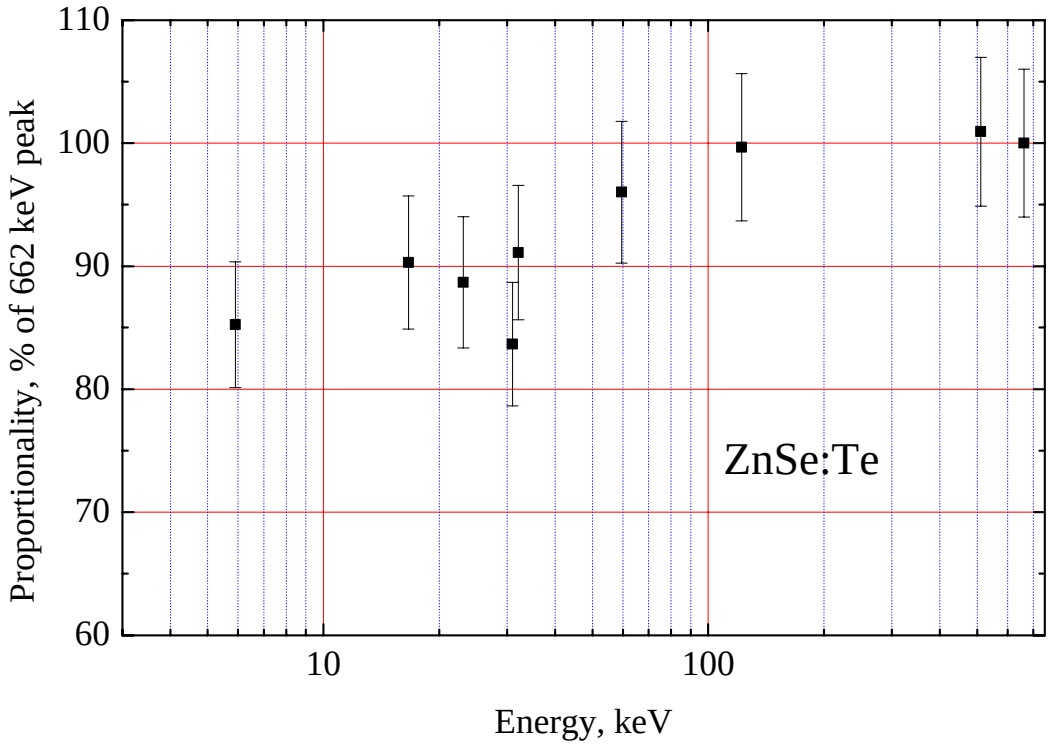

Fig. 1 Proportionality of light output of ZnSe:Te in units of light output at 662 keV

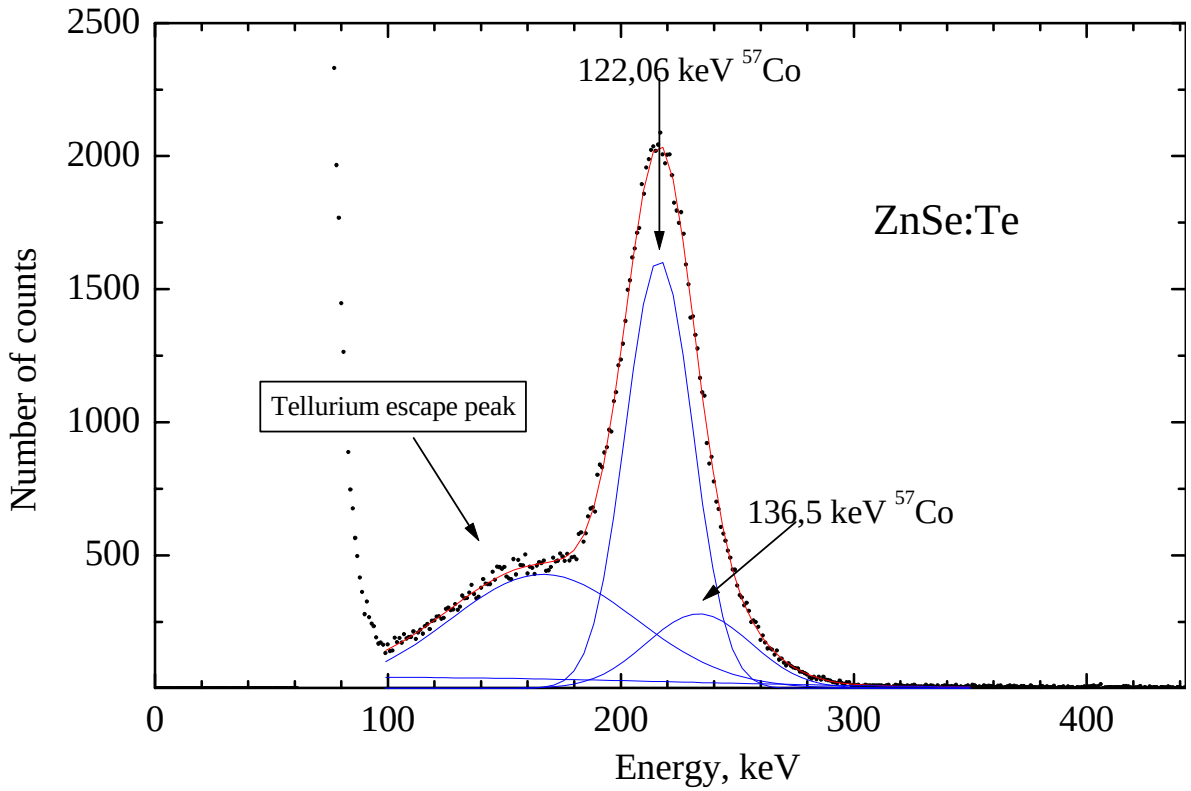

Fig. 2 Energy spectra of γ-rays from a ^{57}Co source measured with ZnSe:Te crystal (sample 1) coupled to the LAAPD at 6 μs shaping time constant in the spectroscopy amplifier

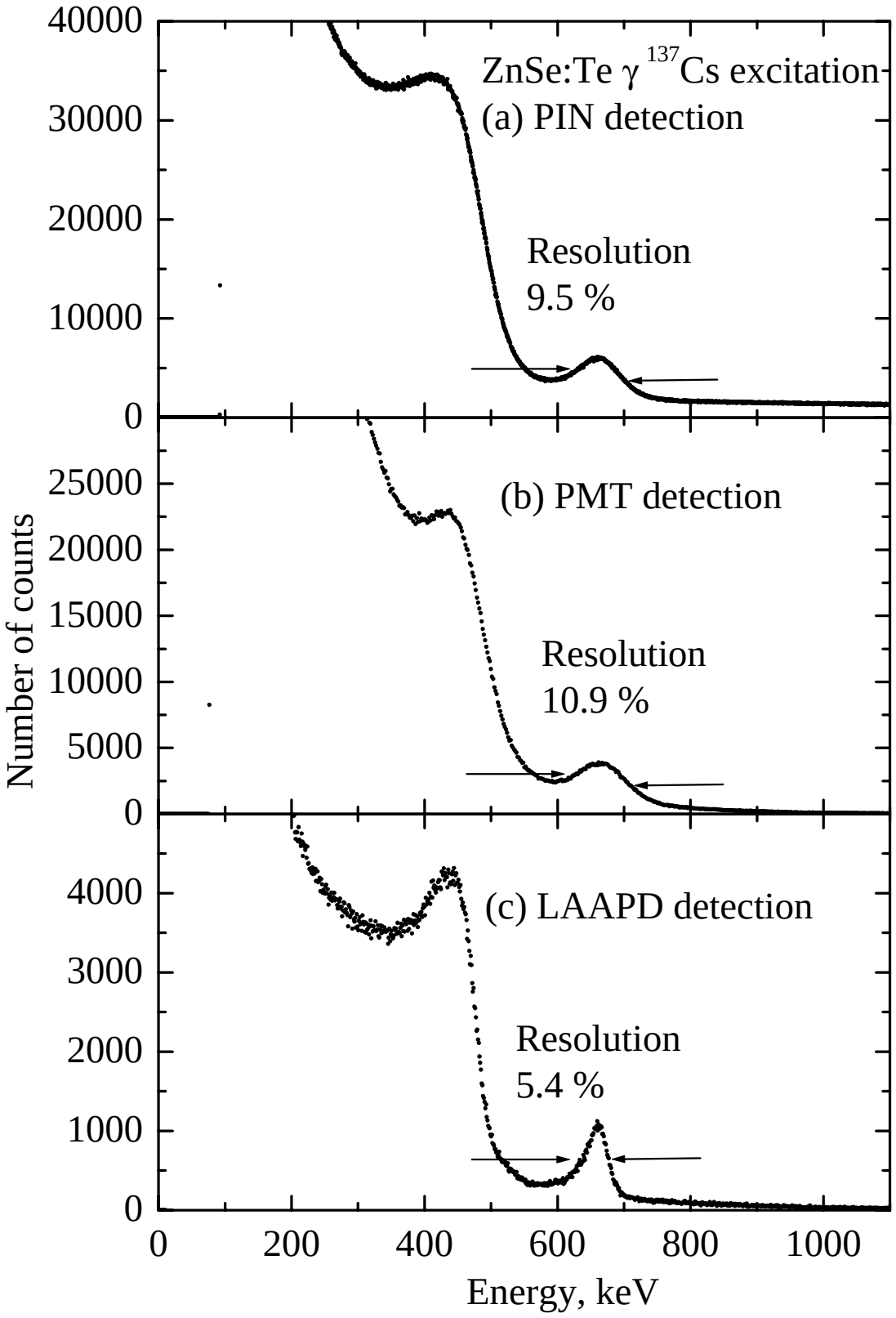

Fig. 3 Energy spectra of 662 keV γ-rays from a ^{137}Cs source measured with ZnSe:Te crystal (sample 1) coupled to Hamamatsu PIN photodiode S3590-08 at 12 μs shaping time (a), the ADIT B51D01 PMT (b), and to the LAAPD (c) at 10 μs shaping time constant in the spectroscopy amplifier

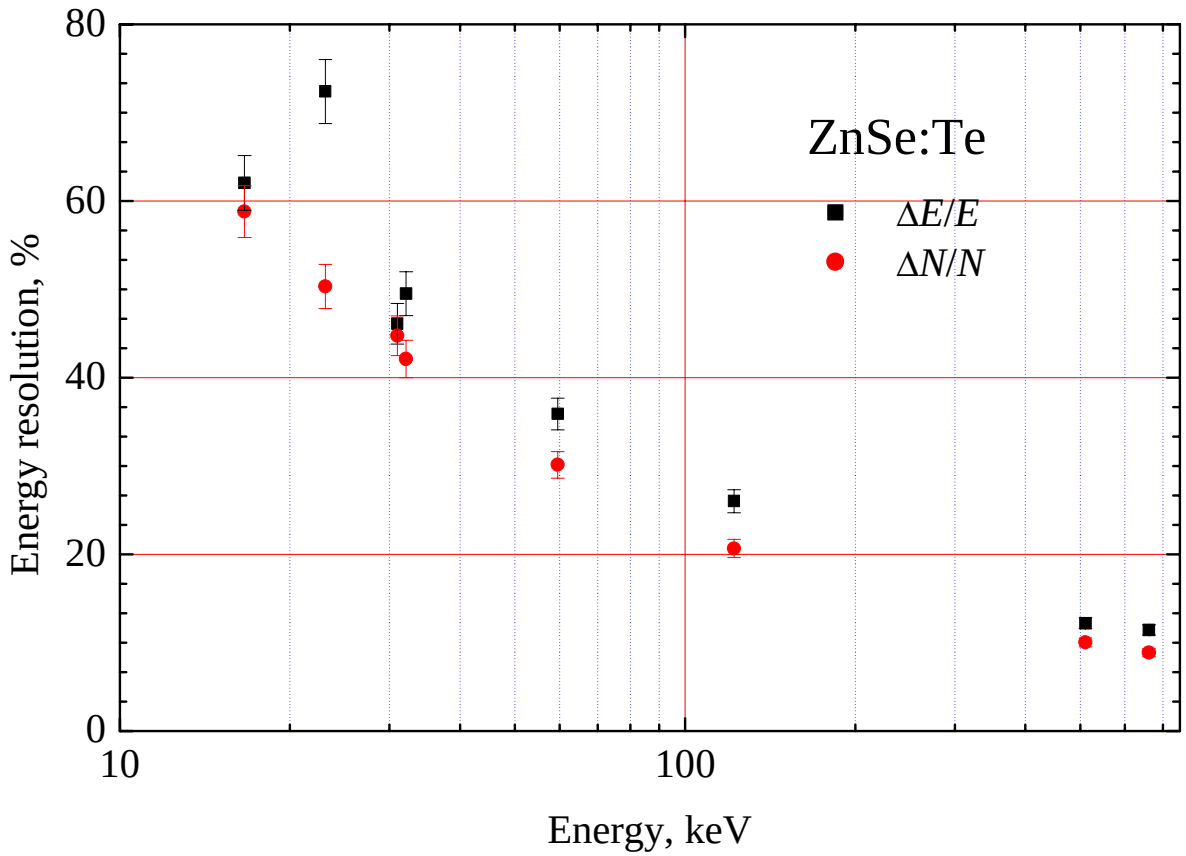

Fig. 4 Energy resolution of ZnSe:Te for sample (1) measured with PMT

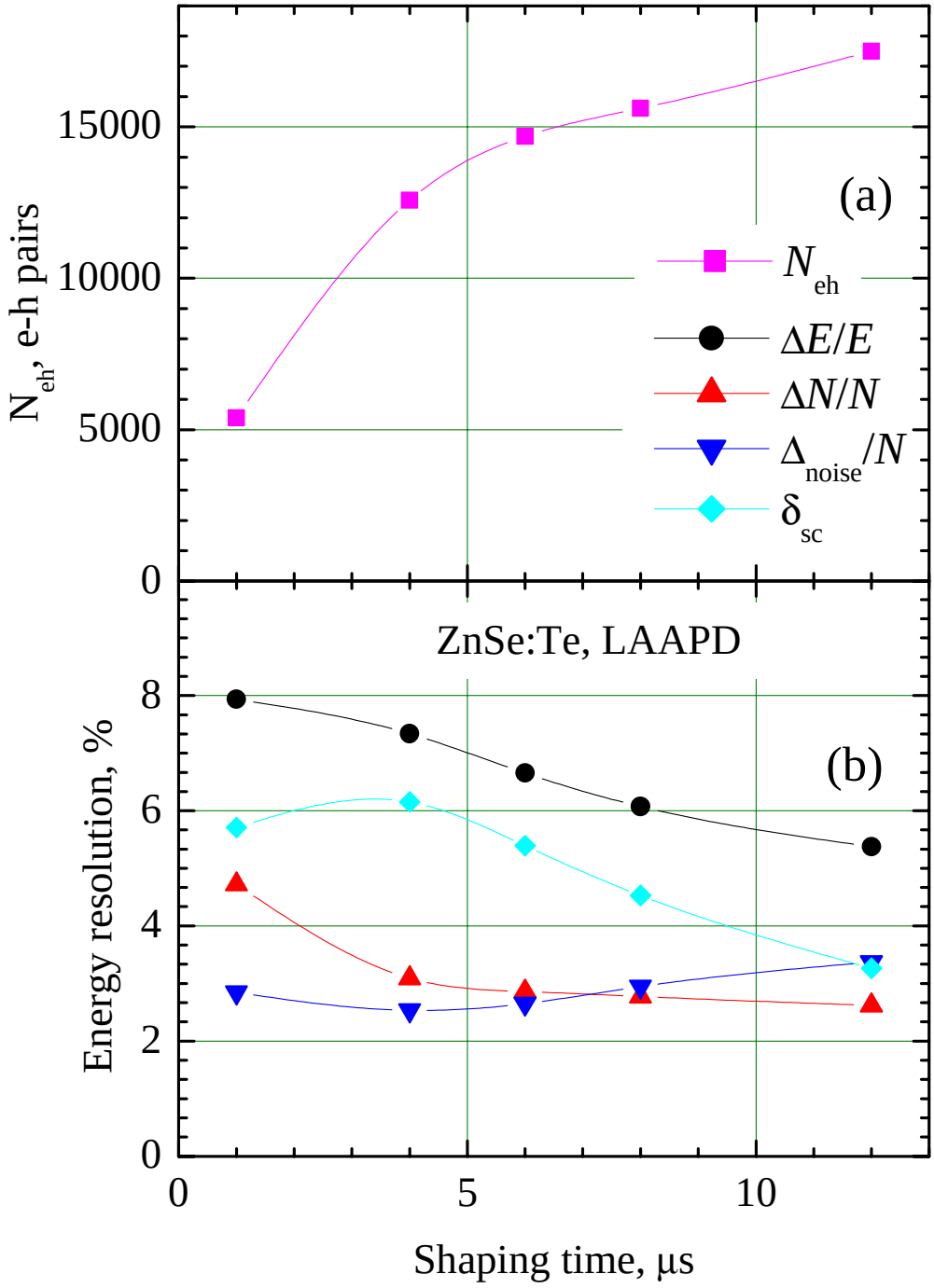

Fig. 5 (a) Light yield of ZnSe:Te in units of the number of e-h pairs at 662 keV vs. shaping time. (b) Energy resolution $\Delta E/E$; intrinsic energy resolution, δ_{sc}; statistical contribution to energy resolution $\Delta N/N$, and noise contribution to energy resolution Δ_{noise}/N of ZnSe:Te at 662 keV versus shaping time. The solid lines are only for eye guiding.

Table 1 Properties of API LAAPD used in this study

Model	630-70-73-500
Enhancement	UV
Serial No.	113-3-1
Active area diameter	16 mm
Window	None
Q.E. at 610 nm	90%
Gain (M)/Bias Voltage	200/2468V (23 deg. C)
Dark current at M=200	226 nA (23 deg. C)
Capacitance (M>20)	110 pF
Step signal rise time	11 ns