YSO, LSO, GSO and LGSO. A Study of Energy Resolution and Nonproportionality.

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Abstract

We have studied nonproportionality and intrinsic energy resolution of cerium doped YSO, GSO, LSO and LGSO crystals. While LSO and YSO have similar light output, GSO and LGSO have ca. 70% and 20% lower light output than LSO, respectively. YSO, as a compound containing fairly light elements, was expected to be proportional at light output vs. energy scale, like YAP:Ce. Surprisingly it is almost the same nonproportional as LSO and GSO. Nonproportionality of YSO is followed by large values of intrinsic energy resolution. The comparison of nonproportionality of YSO – LSO and YAP – LuAP pairs indicates that high proportionality of scintillator is connected with the structure of the crystal and *not* with the presence of light elements. To our knowledge, this is the first study of nonproportionality and intrinsic resolution for LGSO.

I. INTRODUCTION

Growing interest in the development of new scintillation detectors in the last decade has prompted efforts to better understand the limitations of achievable energy resolution. Number of works was addressed to study nonproportionality of scintillator response [1-10]. The largest nonproportionality effect was observed for LSO crystal [1,10] reflected also in the large contribution of scintillator to the measured energy resolution (i.e. intrinsic resolution). On the contrary, YAP crystal showed nearly proportional light output versus energy, that yielded energy resolution of 4.3% for 662 keV γ-rays from a ^{137}Cs source and extremely low contribution of intrinsic resolution of 1.3% [11]. Some other scintillators based on light elements as ZnSe(Te) [12] and K_2LaCl_5:Ce [13] exhibited both a good proportionality and a low intrinsic resolution.

These results raised the question whether a good proportionality and low intrinsic resolution are the effect of light elements in the crystals or this effect is rather associated with the structure of crystals.

A fact that those three crystals LSO, GSO and YSO have the same chemical composition and crystal structure and differ only by atomic number of cations suggests making a direct comparison of nonproportionality and intrinsic resolution for all of them. LSO (Lu_2SiO_5:Ce) was discovered as a scintillator by C.L. Melcher [14]. YSO (Y_2SiO_5:Ce and/or Tb) is known as commercial phosphor. GSO (Gd_2SiO_5:Ce) was first introduced by Takagi [15], LGSO ($Lu_{1-x}Gd_xSiO_5$) was first introduced by S. Yamamoto [16] and then studied by Pichler *et.al* [17]. Non-proportionality of LSO and GSO were reported in [1,10]. Data on intrinsic resolution versus energy for LSO and GSO are known from the study with avalanche photodiodes [18,19]. YSO, because of the low atomic number of yttrium could be a key crystal to get an answer for the question given above. In the study the newest LGSO crystal was tested as an example of mixed compound.

Experimentally, all studies were done by measuring both energy resolution and a number of photoelectrons for a number of γ-rays from different radioactive sources. That allowed determining proportionality curves versus energy and calculation of contribution of photoelectron statistic to the measured

Table 1.

Samples of oxyorthosilicates used in this study

Sample	size, mm	Ce content in melt, %	Ce content in crystal, ppm[b]	polishing	manufacturer
YSO (2)	ϕ60x10	0.2	420±90	ground	CTI PET Systems Inc., Knoxville, TN, USA
YSO (1)	3x3x20	0.2	420±90	manual	CTI
LSO(1)	3x3x20	0.2	520±40	manual	Ramet Ltd, Moscow, Russia
LSO(2)	ϕ60x10 half disk	0.2	100±40	ground	CTI
GSO	10x10x5	unknown	unknown	manual	Lebedev Institute, Moscow, Russia
LGSO, 10%Gd[a]	2x2x7	0.5[a]	420±140	chemical	Hitachi

a – Gd content in crystal $Lu_{2(1-x-y)}Gd_{2x}Ce_{2y}SiO_5$ was measured as x = 0.037, y = 0.0025 nominally ; b – parts per million atoms of cation(s)

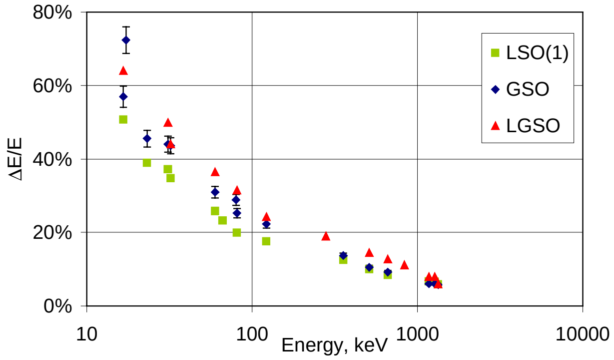

Figure 1. Energy resolution vs. energy for LSO(1), GSO and LGSO

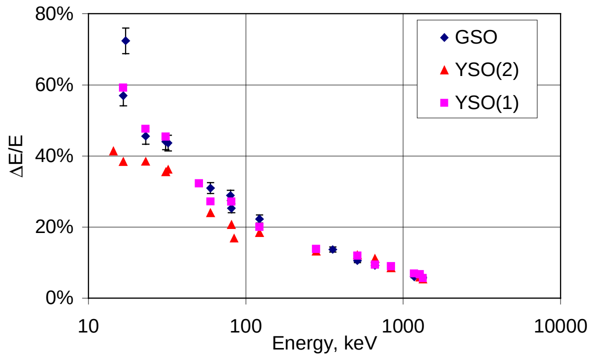

Figure 2. Energy resolution vs. energy for GSO and YSO (1) & (2).

energy resolution. X-ray fluorescence (XRF) method let us measure Ce doping of the tested samples and, in the case of YSO, to show a number of impurities in the crystal.

II. Outline of the Problem

The energy resolution, $\Delta E/E$, of the full energy peak measured with a scintillator coupled to a photomultiplier can be written:

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

where Δ_{sc} is the intrinsic resolution of the crystal, δ_p is the transfer resolution and DN/N is the photomultiplier resolution.

The intrinsic resolution of the 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 the non-proportional response of the scintillator.

The transfer resolution is described by the variance associated with the probability that a photon from the scintillator results in the arrival of photoelectron at the first dynode and then is fully multiplied by the PMT. The transfer resolution depends on the quality of the optical coupling of the crystal and PMT, homogeneity of the quantum efficiency of the photocathode and efficiency of photoelectron collection at the first dynode. In the modern scintillation detectors the transfer resolution is negligible compared to the other components of the energy resolution.

The photomultiplier resolution represents the statistical accuracy of the signal from the PMT:

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

where N_{phe} is the number of photoelectrons and ε is the variance of the electron multiplier gain. ε is equal to 0.09 for XP2020Q photomultiplier, 0.15 for XP3212 and 0.19 for XP3312. For a more detailed discussion of energy resolution, see Refs [1] and [10].

The photomultiplier resolution is the only quantity that can be determined experimentally in eq. (1), based on the measured number of photoelectrons.

III. Experimental Setup

The list of studied samples is given in Table 1. LSO and YSO were tested in small and large size. We have used Philips 51 mm XP2020Q and XP3212 and 76 mm XP3312 photomultipliers (PMT), custom preamplifier and Tennelec TC244 spectroscopy amplifier. The shaping time for all spectra was 1 µs. Quantum efficiency of XP3312 PMT and XP3212 at maximum of emission spectrum of LSO (420 nm) is 30% and 26% respectively and for XP2020Q 22%. Two 51 mm PMTs were checked to cross test the energy resolution measurements. Small size samples were wrapped in PTFE tape and on a largest area side a window was cut in PTFE and crystal was

Table 2
Scintillating properties of oxyorthosilicates for 511 keV and 662 keV

Sample	$\Delta E/E$, [%] for 511keV	Δ_{sc},[%] for 511keV	$\Delta E/E$, [%] for 662keV	Δ_{sc},[%] for 662keV	photoelectron yield, [phe/MeV] for 662keV
YSO(1)	11.9±0.6	11.0±0.7	9.4±0.5	8.5 ±0.6	6230±190[c]
YSO(2)	12.1±0.7	11.3±0.8	11.1±0.5	10.4 ±0.7	6950±210[b]
LSO(1)	10.0±0.6	8.9±0.6	8.4±0.5	7.4±0.5	5730±170[a]
LSO(2)	13.5±0.7	12.8±0.8	11.9±0.6	11.3±0.7	6860±210[b]
GSO	10.5±0.6	6.2±0.3	9.2±0.5	5.3±1.0	1630±50[a]
LGSO	14.5±0.8	13.7±0.8	12.4±0.6	11.6±0.7	4600±180[a] 5040±150[c]

a – XP2020Q, b – XP3312, c – XP3212

glued to PMT with silicone oil. Large samples were glued to PMT and covered with 3 layers of PTFE tape. Radioactive sources used are ^{55}Fe, ^{57}Co, ^{133}Ba, ^{137}Cs, ^{241}Am, ^{170}Tm, ^{203}Hg, ^{22}Na, ^{54}Mn, ^{60}Co. Photoelectron yields were obtained by comparing the position of the full energy peak with that of single photoelectron. XP3312 PMT was used for YSO(2) and LSO(2) samples; XP3212 PMT was used for YSO(1) and LGSO; XP2020Q was used for LSO(1), GSO and LGSO. Ce and Gd content was measured by X-ray fluorescence method described in [20]. Since the apparatus did not allowed excitation of K lines of Lu, germanium secondary target was used to excite L lines of Lu.

IV. Results

Figure 1 shows overall energy resolution Δ*E*/*E* (full width at half maximum) vs. energy for LSO(1), GSO and LGSO samples and Figure 2 for GSO and YSO (1) and (2). GSO data serve as a benchmark for other data on Figures 1-6. Energy resolution of LSO, GSO and YSO is comparable, somewhat worse is observed for LGSO. However this is the newest crystal and its technology yet cannot be sufficiently good. Note a good energy resolution of 8.4% measured with LSO(1) for 662 keV γ-rays from a ^{137}Cs source, see Table 2. Comparable energy resolution of GSO, crystal showing more than three times lower light output, has to be associated with the lowest intrinsic resolution.

Measured number of photoelectrons allowed calculating photomultiplier contribution to the energy resolution, following eq. (2) and than intrinsic resolution. Note that for all the studied crystal, except GSO, high photoelectron numbers were observed (see Table 2), particularly determined with XP3312 PMT. Photoelectron numbers close to 7000 phe/MeV were measured for large LSO and YSO crystals due to high quantum efficiency of PMT.

Intrinsic energy resolution for LSO(1), GSO and LGSO is shown in Figure 3. and for GSO and YSO (1) & (2) in Figure 4. LSO(1) and YSO show $1/E^{1/2}$ type of dependence for Δ_{sc} (*E* is γ energy), while GSO shows more step-like trace decreasing with energy down to tens of keV, approximately flat up to ca. 300 keV and decreasing again. Table 2 shows values of energy resolution, intrinsic energy resolution and photoelectron yield for 511 and 662 keV. Energy 511 keV is of interest for PET application and 662 keV is used as standard reference. Note that among the measured samples the best intrinsic energy resolution is observed for GSO, for which the crystal growth technology is the most developed.

Figure 5 shows proportionality of the light output measured as percentage of the photoelectron yield per MeV of the value at 662 keV for LSO(1), GSO and LGSO and Figure 6 for GSO and YSO (1) and (2). Curves for LSO and GSO are comparable to those reported in [10]. Note almost the same nonproportionality of LSO and YSO. GSO exhibits the most

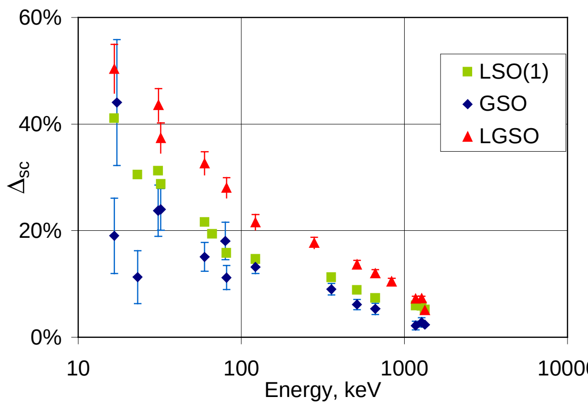

Figure 3. Intrinsic energy resolution for LSO(1), GSO and LGSO.

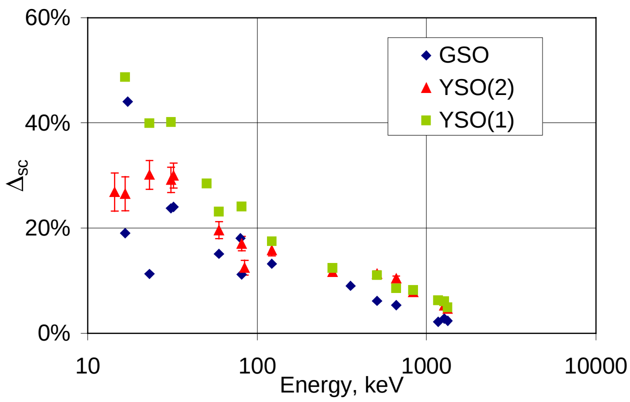

Figure 4. Intrinsic energy resolution for GSO and YSO (1) & (2).

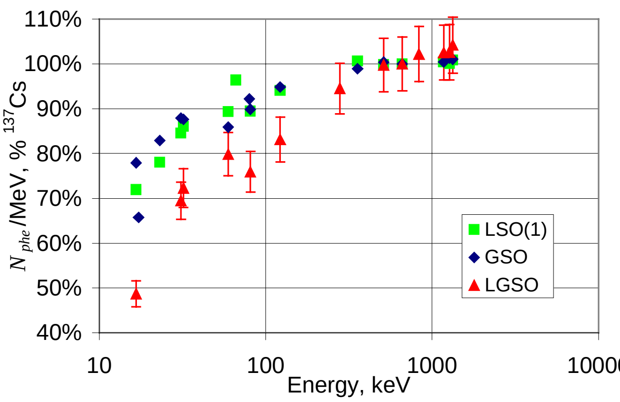

Figure 5. Nonproportionality of the light output for LSO, GSO and LGSO.

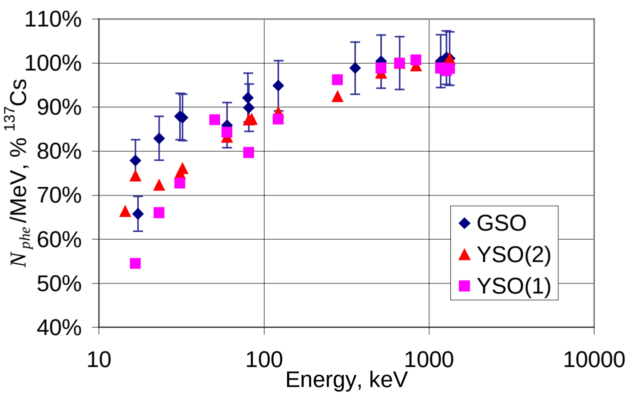

Figure 6. Nonproportionality of the light output for GSO and YSO (1) and (2).

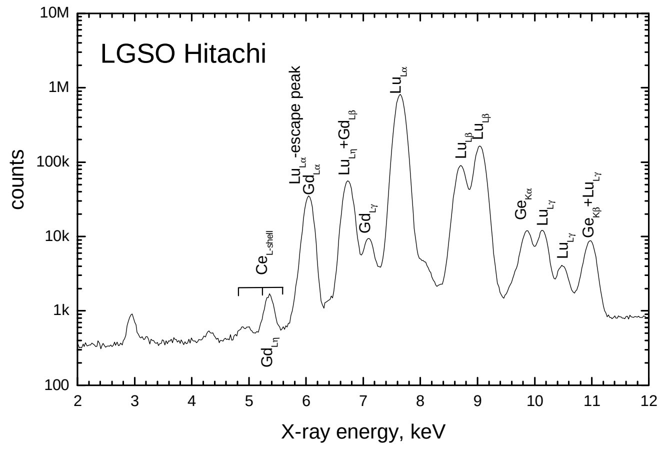

Figure 7. XRF spectra of LGSO:Ce

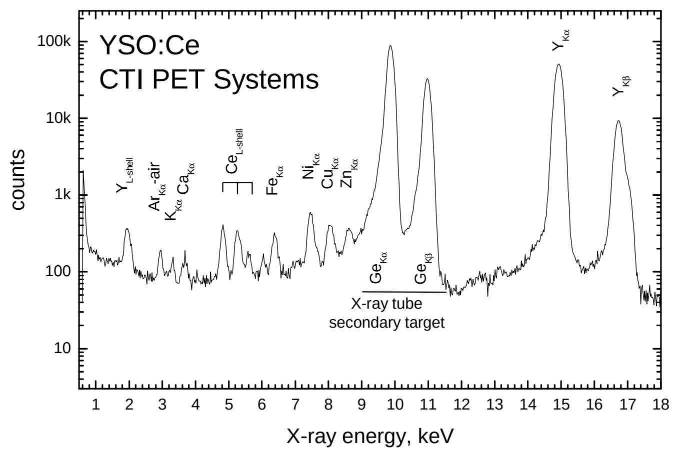

Figure 8. XRF spectra of YSO:Ce

proportional curve while LGSO the least. It suggests that all the measurements of non-proportionality and intrinsic resolution carried out at the present stage of development may be affected by the technology of crystals production.

In order to measure concentration on Ce and Gd in orthosilicates X-ray fluorescence spectra were measured. Figure 7 shows XRF spectra of LGSO and Figure 8 shows XRF spectra of YSO. The spectrum of LSO(2) from CTI can be seen in [20] in Figure 1. In Figure 8 K_α and K_β lines of Y are excited by bremstrahlung. YSO and LSO(2) crystals were grown by the same method at the same location. Spectrum of YSO reveals significant content of metal impurities Ni, Cu, and Zn. Their concentration is at the same order as Ce. In LSO spectra that area is obscured by intense L lines of Lu. Since the technology is the same we can suspect their presence also in LSO.

We report the first measurement of real concentration of Gd and Ce in LGSO ($Lu_{2(1-x-y)}Gd_{2x}Ce_{2y}SiO_5$) by Hitachi Chemical Co. for which $x = 0.037$ in comparison to $x = 0.1$ in the melt. The content of cerium was $y = 420\times10^{-6}$. The content of cerium in $Y_{2(1-x)}Ce_{2x}SiO_5$ was on the level of $x = 420\times10^{-6}$ while in $Lu_{2(1-x)}Ce_{2x}SiO_5$ it was $x = 100\times10^{-6}$ that is much lower then expected from usual distribution coefficient of Ce in LSO (0.18 by number of atoms) [21].

V. CONCLUSIONS.

A comparable nonproportionality is observed for LSO and YSO. YSO has even larger intrinsic resolution than LSO. The exceptional proportionality and low intrinsic resolution observed for YAP crystal [1] is not caused by the fact that crystal is based on yttrium but rather because of its electronic properties and structure. It can be promising observation for LuAP, a scintillator having the same crystal structure as YAP. At present, still nonproportional response is observed for LuAP, however, it can be associated with a large self-absorption of light in that crystal [1, 22, 23]. On the contrary, self-absorption is not observed for YSO, and other orthosilicates. Therefore large nonproportionality and poor energy resolution of YSO is not associated with the atomic number of elements in the crystal, but rather with its structure. We suggest that vice versa is also true, namely that proportionality and low values of energy resolution are connected with crystal structure and electronic properties of scintillators.

The nonproportionality curve for LGSO drops down faster with lower energies than the same curves for LSO and GSO. This property is reflected in higher values of $\Delta E/E$ and Δ_{sc} for LGSO in the whole measured energy scale. LGSO is a new crystal without established technology; therefore its proportionality and Δ_{sc} curve is the worst of all orthosilicates. YSO contains important amount of metal impurities and the same holds probably also for LSO. Impurities presence can cause lowering of crystal quality and hence energy resolution and should be avoided in future crystal growth. One of the possible consequences may be opening of additional channel of energy losses in orthosilicate crystals.

VI. ACKNOWLEDGEMENTS

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