# ON PROPORTIONALITY OF THE LIGHT OUTPUT OF SEMICONDUCTOR SCINTILLATORS UNDER IRRADIATION BY ALPHA-PARTICLES AND HEAVY IONS

V. Ryzhikov<sup>a</sup>, W. Klamra<sup>b</sup>, N. Starzhinskiy<sup>a</sup>, L. Gal'chinetskii<sup>a</sup>, V. Silin<sup>a</sup>, L. Lisetski<sup>a</sup>, E. Dan-

shin<sup>a</sup>, M. Balcerzyk<sup>c</sup>, M. Moszyński<sup>c</sup>, M. Kapusta<sup>c</sup>, M. Szawlowski<sup>d</sup>, K. Katrunov<sup>a</sup>, V. Chernikov<sup>a</sup>, V. Tarasov<sup>a</sup>

"STC "Institute for Single Crystals", 60, Lenin Ave., 61001, Kharkov, Ukraine;

bRoyal Institute of Technology, Department of Physics, Frescati S-104 05 Stockholm, Sweden;

cSoltan Institute for Nuclear Studies, 05-400 Otwock, Poland;

dAdvanced Photonix, Inc. 1240 Avenida Acaso, Camarillo, CA 93012, USA;

E-mail: nstar747@isc.kharkov.com

In the energy range E =2.8...42.2 MeV, the light output S was measured for ZnSe-based scintillators under irradiation with  $\alpha$ -particles and heavy ions <sup>81</sup>Br. Under such irradiation, the proportionality  $S(E_{\alpha,l})$  is observed up to energies  $E_{\alpha} \le 7$  MeV. Substantial deviation from linearity in the region of higher energies is explained by different contribution to S from both primary and  $\delta$ -electrons. Increases were also observed in the values of intrinsic energy resolution with shorter shaping time constants.

PACS: 29.27.Fa

#### 1. INTRODUCTION

For most purposes in radiation physics and technology, scintillator detectors should show proportionality between the energy deposited in the crystal and the number of scintillation photons. However, most of the known scintillators show non-proportionality not only for  $\gamma$ -radiation [1], but also for charged particles [2]. The origin of such non-proportionality has not been fully understood. There are indications that scintillators containing light elements, such as YAP (Ce), are better in this respect [1-3]. However, a comparative study of LSO, GSO and YSO proved this non-proportionality to be independent from the chemical composition of the crystal [3,4]. The aim of the present work was to study the non-proportion-

ality and energy resolution of new scintillators of small effective atomic number A -ZnSe(IVD) crystals (IVD – isovalent dopant Te, O or Cd). Our studies were carried out using irradiation by  $\alpha$ -particles, heavy ions <sup>81</sup>Br and  $\gamma$ -quanta in the energy range E =2.8...42.2 MeV.

## 2. EXPERIMENTAL PROCEDURES

ZnSe(IVD) crystals were grown as described earlier [5] in STC "Institute for Single Crystals", Kharkov, Ukraine. From the single crystals, plates were cut, of dimensions 10x10 mm<sup>2</sup> and thickness 0.8...1.0 mm. In parallel with ZnSe(IVD), similar measurements were carried out for CsI(Tl) crystals. Comparative characteristics of ZnSe(IVD) and CsI(Tl) are given in Table.

Characteristics of scintillators produced at the STC "Institute for Single Crystals"

Crystal	$\lambda_{max}$ , nm	τ, μs	α, cm <sup>-1</sup>	A	S, rel.un.	T <sub>max</sub> , K
ZnSe(IVD)	600640	2150	0,050,15	33	≤150	400450
CsI(Tl)	550	0,633,4	≤ 0.05	54	100	350400

Designations:  $\lambda_{max}$  – maximum position in the radioluminesce spectra,  $\tau$  - decay time,  $\alpha$  - scintillation light absorption coefficient, A – effective atomic number, S – relative light output,  $T_{max}$  – maximum operation temperature.

For light output and energy resolution measurements, we used a 16 mm diameter windowless large area avalanche photodiode (APD) from Advanced Photonix, Inc. It is characterized by quantum efficiency (QE) of 90% for 610 nm and 640 nm. We have also used 10×10 mm Hamamatsu silicon PIN photodiode (PD) S3590-08. The experimental setup included a charge sensitive EG&G ORTEC preamplifier (Model 142AH) and Tennelec Spectroscopy Amplifier (Model 244) or EG&G ORTEC 672 Spectroscopy Amplifier. A PC-based multichannel analyzer Tukan, produced at SINS, was used to record energy spectra.

The experiments with heavy ions were performed at the Tandem Laboratory in Uppsala, Sweden, using alpha and <sup>81</sup>Br beams from the tandem accelerator. The alpha beam energies were 3...15 MeV and 13...50 MeV for <sup>81</sup>Br. The ZnSe(IVD) crystal was mounted on S3590-08

Hamamatsu PIN PD and placed in a scattering chamber at an angle of 20° relative to the beam direction. Symmetrically, but at ~20° angle, a 13x13x1 mm CsI(Tl) crystal was positioned, mounted on a 5 mm thick plexi light guide and Hamamatsu S1723-06 PIN PD. A gold foil, placed in the scattering chamber, was used for Rutherford scattering of the incident beam. Thus, the corresponding energies of the scattered beam were 2.8...14.9 and 10...42.2 MeV for alpha and <sup>81</sup>Br, respectively.

## 3. RESULTS AND DISCUSSION

Our studies have shown that, except some specifically mentioned cases, the type of IVD does not noticeably affect the energy dependence of the light output of ZnSe-based scintillators.

To determine the light output of ZnSe(IVD) crys-

tals, the number of e-h pairs (N<sub>e-h</sub>, e-h/MeV) was measured with APD. The number of e-h pairs produced by the ZnSe(IVD) crystals in APD 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 93Mo source detected directly by the APD. The measurements were carried out at the APD gain of 50 and shaping time constant of the amplifier of 10 µs. The measurement with APD with QE of 90% at 610 nm yielded 26400±1500 e-h pairs/MeV, that is 29300±3000 ph/MeV. This quantity is, however, much lower than 80000 ph/MeV quoted in Ref. [4]. 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 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 manufacturer quotes self-absorption constant for scintillation light as 0.15 cm<sup>-1</sup>. The lower light output for thicker samples 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 the photodiode surface. A very approximate inspection of the light pulse shape done by single photon method indicates that the light pulse consists of a dominating decay component of 2.7±0.3 µs for O- and Cd-doped ZnSe crystals, and a much longer one with decay times well above 10 us for Te-doped ones. 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(IVD) can easily be much larger than that reported in this work.

The proportionality curve, defined as the photoelectron yield measured at a specific energy of excitation relative to the photoelectron yield at 662 keV, is shown in Fig.1. For example, ZnSe(Te) is a rather proportional scintillator down to 85% at 5.9 keV. Note a dip in the proportionality curve at 30 keV, close to the K absorption line of Te (31.8 keV). The same unusual dip is observed for CaF<sub>2</sub>(Eu) at 48.5 keV [7]. The value of the dip (more then 5% in proportionality for ZnSe(Te)) cannot be accounted by simple additional absorption of the heavy element dopant. We have observed a shoulder (Fig.2) in the energy spectrum of <sup>57</sup>Co source (122.06 keV line), which can be attributed, by energy difference, to the 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 y-rays more intensively than it could be expected just from its relative abundance. This and above observations are in good agreement. For O- and Cd-doped ZnSe crystals, these features were not observed.

The results from the in-beam measurements for ZnSe(IVD) and CsI(Tl) are plotted in terms of light output per energy versus energy, as shown in Figs.3 and 4. The data for the ZnSe(IVD) crystal exhibit a slightly non-proportional behavior, in particular for the alpha beam. This is manifested by a slightly concave curve at lower energies. The non-proportionality for CsI(Tl) is also ob-

served, with the relative light output increasing with energy for  $\alpha$ -particles and decreasing for <sup>81</sup>Br. The obtained ratio quantities are 93% and 86% for ZnSe(IVD) and CsI(Tl), respectively.

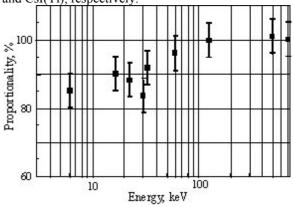


Fig.1. Proportionality of light output of ZnSe(Te) in units of light output at 662 keV

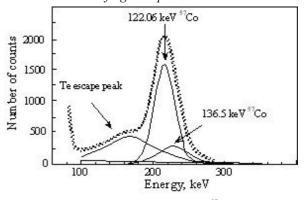


Fig.2. Energy spectra of  $\gamma$ -rays from a <sup>57</sup>Co source measured with ZnSe(Te) crystal coupled to the APD at 6  $\mu$ s shaping time constant in the spectroscopy amplifier

The above procedure of comparing the light yield/energy ratio values is motivated by similar shapes of the curves in Fig.3 and 4, as well as by the fact that the data point at the highest alpha beam energies, which strongly indicates approaching the region of proportionality. One general conclusion is that the non-proportionality for CsI(Tl) at the studied beam energies is much stronger than in the case of ZnSe(IVD).

According to Murray et al. [2] the non-proportional scintillation response is attributed to the non-proportionality in the electron response. In fact, results from electron response measurements by means of Compton Coincidence Technique obtained by Mengesha et al. [1] show that many scintillators exhibit significant deviations from proportionality (up to about 40%),

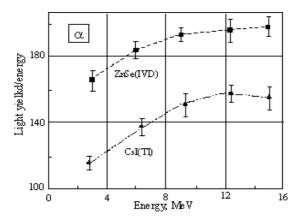


Fig. 3. Light yield/energy versus energy for ZnSe(Te) and CsI(Tl) for the alpha beam excitation

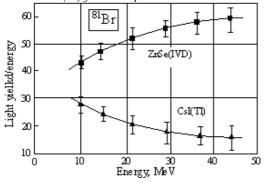


Fig. 4. Light yield/energy versus energy for ZnSe(Te) and CsI(Tl) for the 81 Br beam excitation

mostly pronounced in the low-energy region. CsI(Tl) represents a typical case in that respect. The electron response for ZnSe(IVD) has not been studied so far. An alternative explanation concerns the effect of  $\delta$ -rays produced by the primary particles, as suggested by Murray et al. [2]. These δ-rays most likely result in rather low energetic electrons. In a very simplified approach the total light yield L<sub>tot</sub> may be presented as the sum of two components:  $L_{tot} = L_p + L_{\delta_n}$ where subscripts p and  $\delta$  represents primary and  $\delta$ -rays, respectively. The light yield due to primary particles depends on the energy gap of the material, i.e. is a function of  $1/E_{gap}$ ; see Ref. [6]. On the other hand, the production of δ-rays has been found to be dependent on E/A (A – atomic number) of the incident particles [7]. Since the energy gap for ZnSe(Te) is as low as 2.6 eV, the component Lp is expected to be very large and thus much dominant compared to the second term  $L_{\delta}$ . The slight deviation from proportionality in Fig.3 for alpha beam on ZnSe(Te) may most likely be attributed to a contribution from the low-energy δ-electrons. This may also be an indication for a non-proportionality in the electron response. The electron response

for CsI(Tl) is highly non-proportional [1], mostly in the low-energy region, and the energy gap for this crystal is relatively high, 6.4 eV, making the component  $L_p$  less strong compared to the second term.

### 4. CONCLUSION

ZnSe (IVD) scintillators have a good proportionality of the light output with energy, and this property is also accompanied by high energy resolution. This confirms the assumption that high intrinsic resolution of scintillators is due to good proportionality of the crystals.

#### REFERENCES

- W.Mengesha and J.D.Valentine. A Technique for Measuring Scintillator Electron Energy Resolution using the Compton Coincidence Technique // Proceedings of the Fifth International Conference on Inorganic Scintillators and Their Applications "SCINT99", Moscow State University, Russia. 1999, p.173.
- R.B.Murray and A.Meyer. Scintillation Response of Activated Inorganic Crystals to Various Charged Particles // Physical Review. 1961, v. 122, N 3, p. 815-826.
- M.Balcerzyk, W.Klamra, M.Moszynski, M.Kapusta, and M.Szawlowski. Nonproportionality and temporal response of ZnSe:Te scintillator studied by large area avalanche photodiodes and photomultipliers // Proceedings of the Fifth International Conference on Inorganic Scintillators and Their Applications "SCINT99", Moscow State University, Russia. 1999, p.571.
- W.Klamra, M.Balcerzyk, M.Kapusta, A.Kerek, M.Moszynski, L.-O.Norlin, D.Novak, G.Possnert. Studies of scintillation light non-proportionality of ZnSe(Te), CsI(Tl) and YAP(Ce) crystals using heavy ions // Nucl. Instr. and Meth. 2002, v. A. 484, p. 327-332.
- L.V.Atroschenko, S.F.Burachas, L.P.Gal'chinetskii, B.V.Grinev, V.D.Ryzhikov, N.G.Starzhinskiy. Crystals of scintillators and detectors of ionizing radiation on their base. Kiev: "Naukova dumka", 1998, p. 310.
- 6. A.Lempicki, A.Wojtowicz, E.Berman. Fundamental limits of scintillator performance // *Nucl. Instr. and Meth.* 1993, v. A. 333, N 2,3, p. 304-311.
- 7. R.B.Murray, A.Meyer. Effect of delta rays on the response of inorganic scintillators to heavy particles // *IRE Trans. Nucl. Sci.* 1962, v. NS-9, N 3, p. 33-35.

## О ПРОПОРЦИОНАЛЬНОСТИ СВЕТОВОГО ВЫХОДА ПОЛУПРОВОДНИКОВОГО СЦИНТИЛЛЯТОРА ПРИ ОБЛУЧЕНИИ α-ЧАСТИЦАМИ И ТЯЖЕЛЫМИ ИОНАМИ

В. Рыжиков, В. Кламра, Н. Стажинский, Л. Гальчинецкий, В. Силин, Л. Лисецкий, Е. Даншин, М. Бальцержик, М. Мажинский, М. Капуста, М. Цевловский, К. Катрунов, В. Черников, В. Тарасов

Измерен световой выход S для сцинтилляторов, основанных на ZnSe, при облучении  $\alpha$ -частицами и тяжелыми ионами <sup>81</sup>Br в диапазоне энергий E=2.8...42.2 MэB. При облучении наблюдалась пропорциональность S(E<sub>α,i</sub>) до энергий E<sub>α</sub> ≤7 МэB. Существенное отклонение от линейности в области более высоких энергий объясняется различным вкладом в S как первичных, так и и  $\delta$ -электронов. Заметное влияние на кривую пропорциональности и спектры энергии оказывало легирование Te.

ПРО ПРОПОРЦІЙНІСТЬ СВІТЛОВОГО ВИХОДУ НАПІВПРОВІДНИКОВОГО СЦИНТИЛЯТОРА

#### ПРИ ОПРОМІНЕННІ α-ЧАСТКАМИ І ВАЖКИМИ ІОНАМИ

В. Рижиков, В. Кламра, М. Стажинський, Л. Гальчинецький, В. Силин, Л. Лисецький, Є. Даншин, М. Бальцержик, М. Мажинський, М. Капуста, М. Цевловський, К. Катрунов, В. Черніков, В. Тарасов

Виміряно світловий вихід S для сцинтиляторів, заснованих на ZnSe, при опроміненні  $\alpha$ -частками і важкими іонами <sup>81</sup>Br у діапазоні енергій E =2.8...42.2 MeB. При опроміненні спостерігалася пропорційність S(E  $\alpha$ , і) до енергій  $\alpha$  =27 MeB. Істотне відхилення від лінійності в області більш високих енергій пояснюється різним внеском у S як первинних, так і  $\alpha$ -електронів. Помітний вплив на криву пропорційності і спектри енергії робило легування Te.