Studies of scintillation light nonproportionality of ZnSe(Te), CsI(Tl) and YAP(Ce) crystals using heavy ions

W. Klamra, M. Balcerzyk, M. Kapusta, A. Kerek, M. Moszynski, L.-O. Norlin, D. Novák, G. Possnert

Department of Physics, Royal Institute of Technology, SCFAB, 106 91 Stockholm, Sweden
Soltan Institute for Nuclear Studies, 05-400 Otwock-Swierk, Poland
Ångström Laboratory, 751 21 Uppsala, Sweden

Received 30 August 2001; accepted 11 September 2001

Abstract

The scintillation light yield for ZnSe(Te), CsI(Tl) and YAP(Ce) crystals have been studied with alpha particles, $^{12}$C and $^{81}$Br in the energy region 2.8–42.2 MeV. A nonproportional behavior was observed, mostly pronounced for alpha particles on YAP(Ce). The results are understood in terms of δ-rays effect.

PACS: 29.40.Mc

Keywords: Heavy ions; Scintillation detectors; ZnSe(Te); CsI(Tl); YAP(Ce)

1. Introduction

An ideal behavior of a scintillator would be characterized by a proportionality between the energy deposited in the crystal and the number of scintillation photons. However, this is often not the case, since most of the known scintillators indicate a nonproportionality and this effect have been observed not only for gamma radiation [1], but even for charge particles [2]. In that respect BaF$_2$ and YAP(Ce) represent here an interesting case. Both crystals exhibit a proportional behavior as observed for gamma radiation, however, not for heavy ions [3]. Since the nonproportionality is mostly pronounced in the low-energy region, it makes the mentioned crystals less suitable in low energetic heavy ion spectrometry. For these reasons search for crystals which does not exhibit this kind of behavior is found well motivated. This problem is of a particular importance for an accelerator like CRYRING in Stockholm, which is a storage ring for low energetic highly charged heavy ions. In addition, the Ultra-High Vacuum (UHV) conditions in the CRYRING put special requirements on the detector material.

Previously, a roughly proportional gamma response have been observed for ZnSe(Te) [4]. The aim of the present studies was to examine if this proportionality is valid for low energetic heavy ions as well. Thus, the question is if ZnSe(Te) possibly represents a case different to that of BaF$_2$ and YAP(Ce). For comparison...
measurements for CsI(Tl) and YAP(Ce) were performed as well. The ZnSe(Te) crystal is a scintillator characterized by very long time constants, but also very high light output [5]. For most of the known scintillators the energy gap is roughly in the region 5–10 eV, as for example CsI(Tl) — 6.4 eV, NaI(Tl) — 5.9 eV and BaF₂ — 10.8 eV. ZnSe(Te) is here an exception, with a very low-energy gap of 2.6 eV. The density is 5.42 g/cm³, while the effective Z-value is 30. The latter makes the crystal less suitable for high-energy gamma detection. Since the emission band is in the wavelength region 560–750 nm with a maximum around 610 nm, the use of PIN or avalanche photodiodes is preferred.

2. Experimental procedure

The studies were performed using a polished 10 × 10 × 1 mm ZnSe(Te) crystal. The crystal was delivered by STC Institute for Single Crystal, Kharkov, Ukraine.

In the first series of measurements the crystal was mounted on a diam. 16 mm large area avalanche photodiode (LAAPD) from Advanced Photonix, Inc, alternatively Hamamatsu S3590-08 PIN photodiode. Alpha and gamma spectra from radioactive ²⁴¹Am and ¹³⁷Cs sources were recorded, respectively. The spectra were collected in a PC-based MCA. The alpha and gamma spectra for both LAAPD and PIN photodiode are shown in Fig. 1.

The experiments with heavy ions were performed at the Tandem Laboratory in Uppsala, Sweden, using alpha and ⁸¹Br beams from the tandem accelerator. The alpha beam energies were 3–15 MeV and 13–50 MeV for ⁸¹Br. The ZnSe(Te) crystal was mounted on S3590-08 Hamamatsu PIN photodiode and placed in a scattering chamber at an angle of 20° relative to the beam direction. Symmetrical, but at —20° angle a

![Fig. 1. ²⁴¹Am alpha and ¹³⁷Cs gamma spectra recorded by ZnSe(Te) crystal mounted on LAAPD and PIN photodiode, respectively.](attachment:image-url)
A 13 × 13 × 1 mm CsI(Tl) crystal was positioned, mounted on a 5 mm thick plexi light guide and Hamamatsu S1723-06 PIN photodiode. 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 $^{81}$Br, respectively. Some of the collected spectra are shown in Fig. 2.

Measurements for YAP(Ce) were performed by means of exactly the same experimental procedure as in Ref. [3]. Thus, a crystal of diameter 30 × 5 mm was mounted as a vacuum window in the tandem accelerator scattering chamber at an angle of 45° relative to the beam direction and coupled to a XP2020 QUR PM-tube from Photoni. Beams of 5–15 MeV alpha and 10–30 MeV $^{12}$C ions were directed on the detector due to Rutherford scattering on a gold foil. These resulted in energies of 4.9–14.8 and 9.2–28.6 MeV for alpha and $^{12}$C, respectively. Examples of spectra for YAP(Ce) are presented in Fig. 3.

3. Results and discussion

From the alpha and gamma spectra, as measured by means of LAAPD and PIN photodiodes for $^{241}$Am (5.48 MeV) and $^{137}$Cs (661.6 keV), the ratios $\alpha/\gamma$ were determined. Sysoeva et al. [6] found the $\alpha/\gamma$ values for ZnSe(Te) to be clearly depended on the time constant of the amplifier. Therefore, the PIN photodiode spectra were recorded for time constants from 2 to 12 μs, resulting in values between 0.74 and 0.69, respectively. In the case of LAAPD a value of 0.60 is obtained, as measured at 12 μs time constant. The $\alpha/\gamma$ ratios obtained in the present study are generally below values reported in Ref. [6],

![Fig. 2. Heavy ion spectra with ZnSe(Te) and CsI(Tl).](image)
however, a slight decrease with the time constant is observed.

The results from the in-beam measurements for ZnSe(Te), CsI(Tl) and YAP(Ce) are plotted in terms of light output per energy versus energy, as shown in Figs. 4–6, respectively.

The data for the ZnSe(Te) crystal exhibit a slightly nonproportional behavior, in particular for the alpha beam. This is manifested by a slight down bend curve at lower energies. The nonproportionality for CsI(Tl) is also visible and while for alpha particles the relative light output is increasing with energy, the behavior for $^{81}$Br is just the opposite. The alpha beam data for YAP(Ce) show similar effect as for the two other crystals, however more pronounced. On the other hand, the YAP(Ce) data for the $^{12}$C beam strongly reminds results for $^{16}$O beam in Ref. [3]. For ZnSe(Te) a very slight nonproportionality have been observed for gamma radiation, as reported by Balcerzyk et al. [4]. The scintillation response of CsI(Tl) to alpha particles have been studied previously [7], but not for low energetic $^{81}$Br, as obtained in present experiments. Common for the measurements of all three crystals are the alpha beam results. A way to compare the departure from proportionality in a more quantitative manner is suggested by means of ratios for light yield/energy values at the experimental points at 5.9 and 14.9 MeV alpha beam energies. The value at 5.9 MeV for YAP(Ce) has been extracted by interpolation between other available experimental data points. The obtained ratio quantities are 93%, 86% and 67% for ZnSe(Te), CsI(Tl) and YAP(Ce), respectively. The above procedure to compare the light yield/energy ratio values is motivated by similar shape of the curves in

Fig. 3. Heavy ion spectra with YAP(Ce).

Fig. 4. Light yield/energy versus energy for ZnSe(Te).
Figs. 4–6, as well as by the fact that the data points at the highest alpha beam energies strongly show indication for approaching the region of proportionality. One general conclusion from the comparison of the obtained values is that the nonproportionality for YAP(Ce) at the studied beam energies is much stronger than in the case of ZnSe(Te) and CsI(Tl).

According to Murray et al. [2] the nonproportional scintillation response is attributed to the nonproportionality in the electron response. In fact, results from electron response measurements by means of Compton Coincidence Technique obtained by Mengesha et al. [1] for a number of scintillators exhibit a significant deviation from proportionality (up to about 40%), mostly pronounced in the low-energy region. CsI(Tl) represents a typical case in that respect, contrary to YAP(Ce). The latter shows a highly proportional dependence, but despite that the relative light yield per energy for heavy ions have been found as sharply increasing with energy, Fig. 6 and Ref. [3]. The electron response for ZnSe(Te) 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. [8]. These δ-rays most likely result in rather low energetic electrons.

In a very simplified approach the total light yield $L_{\text{tot}}$ may be presented as sum of two components:

$$L_{\text{tot}} = L_p + L_\delta$$

where subscripts $p$ and $\delta$ represents primary and δ-rays, respectively.

The light yield due to primary particles depends on the energy gap of the material, i.e. is a function of $1/E_{\text{gap}}$, see Ref. [9]. On the other hand, the production of δ-rays have been found to be dependent on $E/A$ of the incident particles [8]. Since the energy gap for ZnSe(Te) is as low as 2.6 eV, the component $L_p$ is expected to be very large and thus much dominant compared to the second term $L_\delta$. In fact, a high light output of $28300 \pm 1700$ photons/MeV as measured for gamma rays is reported by Balcerzyk et al. [4]. The slight deviation from proportionality in Fig. 4 for alpha beam on ZnSe(Te) may most likely be accounted to contribution from the low energetic δ-electrons. This may also be an indication for a nonproportionality in the electron response. The electron response for CsI(Tl) is highly nonproportional [1], mostly in the low energetic 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. This is thus suggested as a possible explanation for data in Fig. 5. YAP(Ce) represent slightly similar case, however, the electron response was found here to be highly proportional [1]. In addition, the energy gap is much higher, 8.1 eV [10]. This possibly makes the effect of δ-rays much stronger. Hence, one may arrive to a more general remark based on the presently known response curves for scintillation crystals. The observed nonproportionality is governed by the nonproportionality in the electron response, but this in turn is suggested to be most likely a general property for most of the crystals. However, the effect may be less pronounced if the light yield due to primary particles is much dominant. Further studies on other crystals may prove if the suggestion above is correct.

In conclusion, the studied scintillation response of ZnSe(Te), CsI(Tl) and YAP(Ce) for low energetic alpha, $^{12}$C and $^{81}$Br ions revealed different results. The nonproportionality for YAP(Ce) is much more pronounced compared to the two other crystals, in particular for lighter ions. The results may most likely be understood in terms of nonproportionality in the electron response and the effect of δ-rays.

Acknowledgements

One of the authors (W.K.) would like to acknowledge financial support from the Swedish Institute.

References