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ZnTe cryogenic scintillator

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A B S T R A C T

The X-ray luminescence and scintillation properties of undoped ZnTe crystal were investigated as function of temperature down to T < 10 K. The luminescence of ZnTe is quenched at room temperature, but below 150 K the crystal exhibits emission at 575 nm with characteristic recombination decay kinetics. The emission is attributed to the radiative annihilation of electrons captured at the shallow levels of impurities or defects with holes trapped by Zn vacancies. The temperature dependence of scintillation light output of ZnTe was studied. It was found that at excitation with α-particles the light output of undoped ZnTe is 117 ± 20% of a reference $CaWO_4$ crystal measured under the same experimental conditions. It is concluded that undoped zinc telluride is a promising scintillator for cryogenic application, particularly for the cryogenic search for neutrinoless double beta decay (0νDBD) of ^{130}Te.

1. Introduction

Zinc chalcohenides ZnX (X = S, Se and Te) activated by isovalent dopants represent a group of semiconducting scintillation materials, known for their very high light yield. ZnS-Ag, discovered at the beginning of the last century [1], remains one of the most efficient one among the scintillators, exhibiting a light yield of 75,000 photons/MeV [2]. Single crystalline ZnSe-Te, introduced into common use for scintillation detection in the early 1990s [3] exhibits similar light yield [4]. Recently a new oxygen-doped ZnTe-O phosphor was developed for high-resolution X-ray imaging, exhibiting higher light yield than CsI-Tl, fast decay (~1 μs) and low afterglow [5]. The high efficiency of the conversion of X-ray radiation into emission in the visible wavelength range is a distinctive feature common to activated zinc chalcohenide scintillators. The main factors that facilitate the enhanced light yield can be attributed to the specific properties of scintillating semiconductors i.e. lower band gap compared with dielectric scintillators and higher mobility of free carriers. Indeed, the light yield per unit of absorbed energy is inversely proportional to the value of energy gap of a material E_g [6,7], so the decrease of this parameter directly translates into higher light yield. Moreover, the high mobility of free electrons and holes in semiconductors facilitates their prompt escape from the region of the ionisation track, in which high excitation density increases the probability of non-radiative quenching [8–10]. In addition, this is beneficial for the transfer of excitation energy to the activators. Thus, due to their high light yield, activated zinc chalcohenide scintillators are used in various applications that require detection of high energy photons and particles [11–13].

During recent years there has been continuous interest in developing techniques for particle detection at low temperature. This is driven by the particle physics community and in particular by searching for rare events such as neutrinoless double beta decay (0νDBD) [14] and interaction of dark matter particles with target nuclei [15]. Pursuing the aim of reaching the ultimate sensitivity some of these experiments use cryogenic phonon-scintillation detectors that feature both high energy resolution and event discrimination capability [16–18]. The detectors have proven their advantages in strongly reducing contributions from radioactive background in the region of interest explored by experiments searching for rare events [19–21]. Therefore, there is currently steady interest in furthering this technique and finding new scintillation materials with high light yield at cryogenic temperatures is an important part of this activity.

Because of their excellent scintillation properties, zinc chalcohenides attracted instant attention. The scintillation properties of ZnSe were investigated down to a temperature of 10 K and it has been suggested that this material has great potential for application at

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cryogenic temperatures, specifically for the experimental detection of 0νDBD in ^{82}Se [22]. This finding instigated an extensive research programme, aiming to produce enriched Zn^{82}Se scintillators and to develop background-free cryogenic phonon-scintillation detectors [23]. The latest results from the prototype of such an experiment demonstrated the excellent background rejection capability of this detection approach, which can translate into unprecedented sensitivity of future 0νDBD searches [24,25].

ZnTe is another representative of the zinc chalcohenide family that may offer a unique opportunity for experimental searches of 0νDBD. ^{130}Te is considered to be a very promising nucleus for this due to its high natural abundance (33.8%) and high transition energy (Q = 2528 keV) [26]. It is worthwhile to remark that the best limit on the half-life of ^{130}Te ($> 4.0\times10^{24}$ years) is currently set by a cryogenic experiment that uses TeO_2 crystals as phonon detectors [27]. Further improvement of sensitivity requires better background discrimination, which can be achieved by detecting, in addition to the phonon signal, also the scintillation response of the crystals. However, this option is difficult to implement for TeO_2 because it is a very poor scintillator [28,29]. Thus, if ZnTe is proven to be a good cryogenic scintillator, it could make a strong contribution to the existing cryogenic searches for 0νDBD in ^{130}Te. Up to now, no data on scintillation properties of ZnTe crystals have been reported. This finding motivated us to investigate the feasibility of ZnTe for application as cryogenic scintillator. In this paper we present results on X-ray luminescence, scintillation light output and decay time characterisation of undoped ZnTe down to 6 K.

2. Experiment

The sample of undoped ZnTe of $5\times5\times2$ mm^3 volume, used in this study, was produced in the Institute of Scintillation Materials (Kharkiv, Ukraine), using the high-pressure Bridgman method. The sample was placed into a closed-cycle He cryostat, equipped with a DE-202A cryocooler (Advanced Research Systems) and Cryocon 32 (Cryogenic Control Systems Inc.) temperature regulator. The emission was excited by a URS-55A X-ray tube with a Cu-anticathode tube operating at 55 kV and 10 mA. The emission spectra were recorded using an automated spectrograph M266 and CCD-camera equipped with a Hamamatsu S7030-1006S sensor, sensitive over a 200–1100 nm wavelength range.

For measurements of scintillation characteristics the crystal was placed in a helium constant flow cryostat (Oxford Instruments) and excited by α-particles from an ^{241}Am source. Scintillations were detected by a multi-alkali photomultiplier model 9124A (Electron Tube Enterprises, Ruislip, UK). For the measurements and data analysis we used the multi-photon counting techniques described in detail elsewhere [22,30].

3. Results and discussion

Undoped ZnTe exhibits no measurable scintillations at room temperature; for that to appear the crystal needs to be cooled to below 150 K. Fig. 1 shows the pulse height spectrum of scintillations, emitted by the crystal held at a temperature of 6 K and irradiated with α-particles, emitted by a ^{241}Am source. The spectrum features a broad band with Gaussian shape, attributed to the energy deposited by α-particles (~5.5 MeV).

The position of the peak in the pulse height spectrum is proportional to the scintillation light output of the sample under investigation. Therefore, changes of the light response with temperature can be monitored by measuring the peak position as a function of temperature. Fig. 2 shows this temperature dependence of the light output for the ZnTe crystal under test. With cooling the crystal the peak shifts towards higher amplitudes, indicating a steady rise of the scintillation light output. This trend is observed until the temperature reaches 30 K when scintillation light output remains fairly constant. This type of dependence is controlled predominantly by the processes of temperature

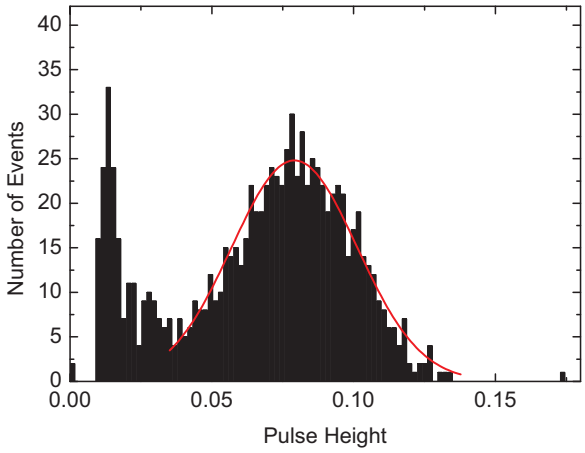

Fig. 1. Pulse height spectrum of scintillations excited by α-particles from ^{241}Am in ZnTe at T = 6 K. The spectrum is fitted by Gaussian (red). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

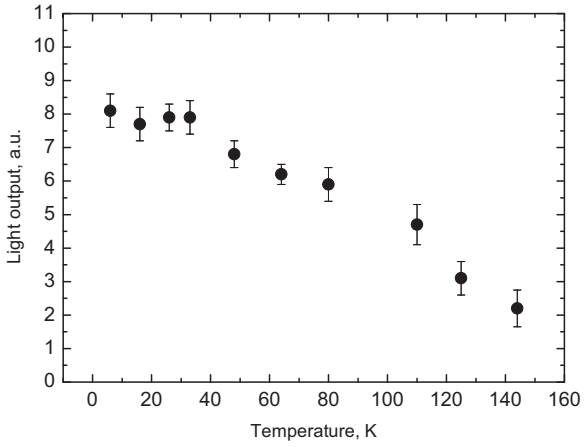

Fig. 2. Temperature dependence of light output of ZnTe. Excitation by α-particles from ^{241}Am (5.5 MeV).

quenching in which the captured carriers can be thermally released from the emission centres and then undergo non-radiative decay. It is worth noting that this type of temperature dependence is characteristic for undoped ZnSe [22] as well as other intrinsic scintillators [7].

Fig. 3 shows the scintillation decay curve of undoped ZnTe measured at 6 K. The scintillation curve exhibits a complex non-exponential decay that is characteristic for recombination lumines-

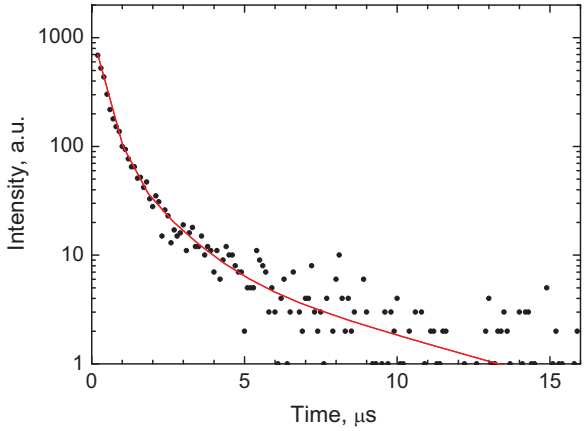

Fig. 3. Scintillation decay curve of ZnTe measured at T = 6 K. Excitation by α-particles from ^{241}Am (5.5 MeV). The line shows the best fit to the experimental data using three exponentials with $\tau_1 = 0.3$, $\tau_2 = 1.2$ and $\tau_3 = 5.6$ μs.

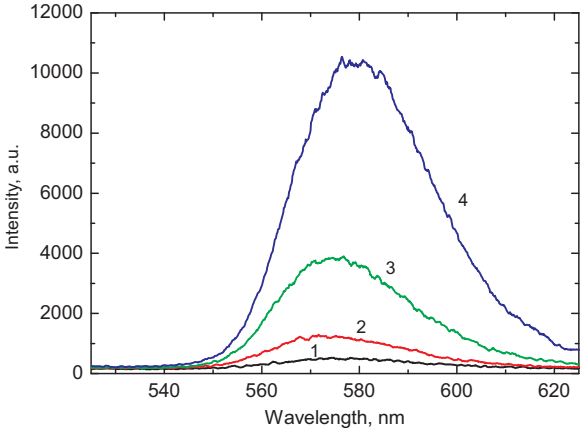

Fig. 4. X-ray luminescence spectra of undoped ZnTe monitored at T = 120 K (1) 80 K (2), 40 K (3) and 10 K (4).

cence. Despite the limited agreement of the fit to an exponential function, for the sake of quantitative representation, the recombination decay curves observed in semiconductors are often approximated by the sum of several exponential functions $f(t) = \sum_i A_i \exp(-t/\tau_i)$, where A_i and τ_i are amplitudes and decay time constants [31]. Therefore, the scintillation decay curve of undoped ZnTe displayed in Fig. 3 was fitted using three exponential functions with decay time constants 0.3(89%) 1.2(9%) and 5.6 μs(2%), respectively (the values in the brackets represent relative intensities of components). The shape of the scintillation curves is largely unaffected by temperature changes, consistent with the previous observations made for ZnS and ZnSe [2,22]. This indicates that temperature changes of the emission in these materials are mostly due to the thermally activated release of charge carriers followed by their trapping by non-radiative centres [31].

The X-ray luminescence spectra of the undoped ZnTe crystal, measured when cooled to 10 K are displayed in Fig. 4. The luminescence band with the maximum at 575 nm (2.15 eV) starts to appear when the temperature reduces to below 140 K. Further cooling of the sample leads to a gradual increase of the emission intensity which is consistent with the measurements of scintillations. The nature of the intrinsic emission of undoped ZnTe has been studied in details for decades. The current, common opinion is that the sharp emission lines observed at low temperatures just below the band gap ($E_g = 2.4$ eV) are due to free excitons and excitons bound to shallow traps. The broad luminescence bands observed in the 2.1–2.3 eV region are attributed to the radiative recombination of donor-acceptor pairs localised at impurities or defects [32–35]. Similar to other semiconductors, the physical processes that involve transitions of carriers across the band gap in ZnTe are strongly influenced by intrinsic defects and impurities. They form different centres with energy levels within the band gap of the crystal that in turn may drastically affect the radiative decay. It has been shown that the cation sublattice of zinc chalcohenides has lower stability and hence higher propensity for defect formation [36]. Indeed, due to a higher evaporation rate of zinc at elevated temperature the most common type of native defect in ZnTe is a Zn vacancy (V_{Zn}) [37]. A Zn vacancy can act as double acceptor with ionisation energies of 0.05 and 0.14 eV [32]. At high-energy excitation, electrons are promoted to the conduction band, creating holes in the valence band of the crystal. The holes are then trapped by Zn vacancies while thermalised electrons can be captured by shallow levels of the charge-compensating sites associated with either donor type impurities or intrinsic defects. The holes recombine with the electrons from these sites, resulting in a broad emission band with characteristic bimolecular decay kinetics. This type of emission complexes is regarded as efficient centres of radiative recombination in zinc chalcohenides and responsible for their high scintillation light yield [12].

In order to assess the prospects for application of ZnTe as cryogenic scintillator we estimated the light output of the crystal under investigation in comparison with a sample of our reference $CaWO_4$ scintillator. Assuming identical light collection, the relative light output R can be expressed as a ratio of the measured light outputs of the two crystals L corrected for the difference in emission-weighted detector efficiency $s_\lambda(\lambda)$:

$$R = \frac{L_{ZnTe}}{L_{CaWO4}} \times \frac{\varepsilon_{\lambda,\ CaWO4}}{\varepsilon_{\lambda,\ ZnTe}}$$

The emission-weighted detector efficiency $s_\lambda(\lambda)$ calculated from the known spectral sensitivity of the photomultiplier $\varepsilon(\lambda)$ and the spectral response of scintillators $s(\lambda)$ is 0.24 for $CaWO_4$ and 0.05 for ZnTe. The light output of the reference $CaWO_4$ measured in the same experimental setup at 6 K is found to be 4.1 times higher than that of ZnTe. From this we can estimate that the relative light output of ZnTe at 6 K is equal to $(1/4.1) \times (0.24/0.05) \times 100\% = 117 \pm 20\%$ of $CaWO_4$. The error of this evaluation stems from the uncertainty in the calculation of the emission-weighted detector efficiency and the position of the peak that corresponds to α-events in ZnTe (see Fig. 1). It is important to accentuate that this estimate is made for scintillations excited by alpha particles; it does not hold for the excitation with gamma rays due to the significant difference of quenching factors in the two scintillators under test [38]. (The amount of light produced by heavy particles in dielectric scintillators is lower than that produced by electrons of the same energy, while for scintillating semiconductors the opposite is true, i.e. much more light is produced by these particles.) Nonetheless, the finding evidences that due to the high scintillation light yield at low temperatures ZnTe is a promising cryogenic scintillator.

4. Conclusion

In this study for the first time, we carried out measurements of X-ray luminescence and scintillation properties of undoped ZnTe below 10 K. Due to significant thermal quenching, scintillations are detected in the crystal only at T < 150 K. The emission of the crystal is attributed to the radiative recombination of holes trapped by Zn vacancies and electrons captured at the shallow levels of impurities or defects. The scintillation efficiency increases with further cooling. It is found that at α-particle excitation undoped ZnTe exhibits fairly competitive light output when compared with our $CaWO_4$ reference scintillator. Such a value is very encouraging for a first test of the scintillator. Improvement can normally be expected through subjecting the material production to optimization processes. This finding underpins potential applications of ZnTe as a cryogenic scintillation detector.

Based on the results of this study it is interesting to consider applications of ZnTe as conventional scintillation detectors at the temperature of liquid nitrogen (T = 77 K). The rationale behind this is fairly straightforward: at this temperature the scintillator exhibits a reasonably short decay time constant and sufficient scintillation response to particle excitation. A practical implementation of this idea poses no real technical challenge since photomultipliers and Si-based photodetectors are proven to operate reliably and efficiently at this temperature. Furthermore, latest advances in optimization of light collection efficiency permits achieving high light yield and compatible energy resolution in geometries without optical coupling, which is an important constraint in the case of cryogenic scintillation detectors [39]. Thus, a detection module with a ZnTe scintillator, operating at the temperature of liquid nitrogen, may offer an attractive option for experiments searching for 0νDBD of ^{130}Te. It should be noted that the idea of a 0νDBD experiment with scintillation detectors cooled to modest temperatures is currently actively investigated [40,41].

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