

Quenching factor for alpha particles in ZnSe scintillating bolometers

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Abstract. In the framework of the CUPID-0 experiment, a numbers of ZnSe single crystals were produced and subjected to different thermal treatments, and later tested as cryogenic scintillating bolometers. We have found that a specific thermal treatment (24 hours under argon atmosphere at 900 °C) has a strong impact on some properties of ZnSe crystals (amplitude of signal, light yield, specific resistivity) and most interestingly, changes the quenching factor for alpha particles from values > 1 to values < 1. Thus such thermal treatment opens the possibility to modify this experimental parameter for a various applications.

1. Introduction

Cryogenic scintillating bolometers are a promising technology to explore diverse rare nuclear processes like rare alpha decay, rare or forbidden beta decays, neutrinoless double beta decay, which have characteristic decay times larger than 10^{14} y. The main feature of scintillating bolometers is the simultaneous heat release and light emission caused an interacting particle in the crystal. Read-out of both these signals allows for an active discrimination of the type of interacting particle since the amount of emitted light depends on the nature of the particle. Besides excellent particle discrimination, the bolometers also possess a very good energy resolution and nearly 100% detection efficiency.

Observations including ^{209}Bi alpha decay ($T_{1/2} = 1.9 \times 10^{19}$ y) [1], ^{180}W alpha decay (1.8×10^{18} y) [2], ^{151}Eu alpha decay (4.6×10^{18} y) [3] and ^{148}Sm alpha decay (4.6×10^{15} y) [4] are good examples to power for this technique. ZnSe scintillating bolometers are the fundamental technology of the CUPID-0 experiment which is searching for neutrinoless double beta decay of ^{82}Se .

Zinc selenide, especially when doped with Te or Al, has a long history of use [5] particularly as X-ray and gamma-ray detectors [6]. However, ZnSe crystals have an interesting feature: alpha particles produce more light than electrons at the same deposited energy in the detector, i.e. Quenching Factor (QF) is greater than unity. To our knowledge no other scintillator with a QF for alphas greater than 1 has ever been reported in literature. Such anomalous behavior of scintillation cannot be easily accommodated within the theoretical framework used to describe the scintillation properties of materials. Indeed, heavy particles usually have a QF smaller than one and it is explained on the basis of a saturation effect due to the high ionization density that characterizes the interaction of heavy



particle in matter [7]. Although this feature is still not understood it is very important for particle discrimination. When the light yield is a strong function of the particle type, the effective particle discrimination can be achieved.

The results reported here reflect ongoing R&D activities within the CUPID-0 experiment to improve the ZnSe scintillating bolometers performance. We have investigated methods to reduce after-growth thermal stress which can trigger cracking of the crystals. We have found that the anomalous QF for alpha particles in ZnSe crystals depends on certain type of defects and it can be adjusted by thermal treatment.

2. Scintillating bolometers and experimental details

The working principle of bolometers is based on measuring the temperature rise induced by particle interactions in a crystal absorber that is operated at mK-temperatures. The interaction in the scintillating crystal produces ionization and excitation, triggering a series of processes that lead to the conversion of largest part of the deposited energy into phonons (i.e. lattice elementary excitations). Some fraction of the energy may be avoiding phonon conversion, for example populating long-lived excited states of defects or other *hidden* energy channels. In scintillating bolometers, some energy is transferred to luminescence centers and the emitted photons escape the crystal. In this case, for each particle interaction two signals are recorded: a phonon signal produced in the scintillating crystal and a light signal read-out by an adjacent Light Detector (LD). The ratio between the two (light-to-heat) depends on the particle Light Yield (LY) that is defined here as the fraction of the total energy deposited in the crystal, which is converted into the light. Betas and gammas show the same LY, which is typically different from the LY of alpha particles. Consequently, the simultaneous read-out of heat and light signal allows for particle discrimination.

The temperature rise induced by the particle interaction in the scintillating crystals and by the scintillation photons in the LD are read out by means of Germanium Neutron Transmutation Doped thermistors (Ge-NTD) [8]. The Ge-NTDs are coupled to their respective absorbers using a small amount of epoxy resin. In order to monitor the drift of thermal gain of the ZnSe bolometer and LD, a heavily doped silicon resistor was glued on the respective detectors to inject calibrated heat pulses. Temperature drifts can thus be corrected off-line on the basis of the measured thermal gain variation [9,10]. The crystal and the LD are housed in a copper structure similar to the one described in [11] and kept in position by PTFE tips. The ZnSe crystals are surrounded by a reflecting foil (3M VM2002) to increase the overall light collection efficiency; however they are not in thermal contact with the foil. A high purity Ge wafer ($\varnothing 44.5 \times 0.2$ mm) is used as a LD, more details can be found in [12].

Ge-NTD thermistors transduce the energy deposits in the crystal and the light detector into voltage signals which are amplified and fed into an 18-bit ADC. The entire waveform of each voltage signal is amplified and continuously sampled and digitized by an ADC. At the ADC input, a low pass Bessel filter is used [13]. The trigger of the ZnSe crystal was software generated, while the LD is independently acquired in coincidence with the former. The waveforms are filtered with the optimal filter technique [13,14] to improve the signal-to-noise performance and the amplitude (V_{heat}), taken as the maximum of the filtered pulse, is determined.

The ZnSe bolometers were energy calibrated with external gamma sources (^{40}K and ^{232}Th) placed outside the cryostat. Gamma lines between 911.2 keV and 2614.5 keV were used to determine the amplitude (V_{heat}) to energy (E) conversion function of the heat signal, attributing to each identified peak the nominal energy of the gamma line as if all the energy was converted into heat. Since it is performed using the electrons produced by gamma rays, it is commonly indicated as “electron equivalent” (keVee). The energy dependence of amplitude was parameterized with a second order polynomial with zero intercept in $\log(V_{\text{heat}})$.

In order to compare the detector response to alpha and beta/gamma particles the ZnSe crystals were exposed in all runs to an external low-intensity smeared alpha ^{238}U source, which provides alpha particles in range from 0 to 4.5 MeV.

A weak ^{55}Fe source was permanently facing the LD, on the side opposite to the ZnSe crystal scintillator, to allow for a direct energy calibration of the LD. The signal amplitude from the light energy was assumed to have a linear dependence in the range of 0 – 35 keV. An energy resolution (FWHM) of 590 eV was evaluated for the X-ray doublet of 5.9 and 6.5 keV. The ^{55}Fe calibration provides a method to measure and compare the LY of different type of particles, but it cannot be used for an absolute evaluation of the amount of scintillation light emitted since the light amplitude is not corrected for the photon collection efficiency.

3. Crystals and thermal treatment

For our studies we have used three different ZnSe crystals whose characteristics are reported in table 1. The crystals have been produced by the high pressure Bridgman technique in a graphite crucible under argon atmosphere in the Institute for Scintillation Materials of NASU (Kharkov, Ukraine). Two of the crystals are nominally undoped, while the third one is doped by Al at the level of 10 ppm. Their initial color variation is caused by slightly different initial stoichiometry – crystals #1 and #2 were produced from ZnSe powder, while crystal #3 was produced from Chemical Vapor Deposition (CVD) smashed pieces - and the thermal conditions during crystal growth, that have also a final impact on stoichiometry of the melt [15], as well as on defect types and amount.

Table 1. Characteristics of the three different used ZnSe crystals.

	#1	#2	#3
Dimensions, mm	Ø43×27	Ø43×44	Ø45×50
Mass, g	250	329	430
Initial charge	ZnSe powder	ZnSe powder	CVD pieces
Doping	undoped	undoped	Al (10 ppm)

For the thermal treatment the crystals were placed in a graphite crucible, which then was placed in a quartz ampule. The quartz container was put under a low pressure of argon (99.93% purity) and placed in a vertical oven, which was heated up to 900°C (100°C/h). The annealing was done at 900°C for 24 hours followed by cooling down at a rate of 100°C/h. After this annealing, for all three crystals were undertaken a cryogenic test as scintillating bolometers.

4. Effect of thermal treatment on detector performance

The cryogenic tests have been performed operating the ZnSe crystals as scintillating bolometers at about 15 mK, in a low temperature refrigerator installed underground in Hall C of Laboratori Nazionali del Gran Sasso (LNGS), Italy. In all runs the ZnSe bolometers worked properly but they cooled more slowly than all other crystals ever tested.

The thermal treatment (TT) significantly reduced the signal amplitude, computed as the maximum of the filtered pulse, both in the heat and the light channel for all crystals under investigation. Table 2 gives a summary on the recorded signal amplitude before and after TT.

Table 2. Impact of thermal treatment on the signal’s amplitude in the heat and the light channel of ZnSe scintillating bolometers. Values of the signal amplitudes are expressed in $\mu\text{V}/\text{MeV}$.

	#1		#2		#3	
	heat	light	heat	light	heat	light
Before TT	6.5	1700	6.5	400	50	980
After TT	1.5	250	5.0	210	13	310

Such a significant decrease in signal amplitude is about five times in case of the ZnSe #1 and is about three times in case of the ZnSe #3 crystal cannot be explained by the different mounting of the thermistors before and after TT. The observation suggests changes of the crystal defect structure and the appearance of a large amount of high mobility charge carriers. Last conclusion is supported by

measurements of the specific resistivity of the ZnSe material. In case of the ZnSe #3, the crystal's specific resistivity decreased from 10^{10} Ohm·cm to 10^7 Ohm·cm after TT. Since the material resistivity is proportional to the inverse product of the number of defects and the mobility of charge carriers, the TT drastically affects on both parameters. At the same time, the TT decreases also the number of luminescence centers or changes their nature, which results in a reduction of the light signals by a factor between 2 and 5 (see table 2).

After TT the heat capacity of the crystals increased, indicating an increase in the crystal lattice distortion due to introduction of new defects and/or new types of defects. Evidence for changes of the crystal structure is also derived from the light and thermal signal pulse-shape.

The time response of the LD is extremely slow – the light pulse has a rise time of the order of a few ms and decay time of the order of hundreds ms. However, at low temperature, also the light emission from scintillators is characterized by a long time constants, with different values for alphas or beta/gammas [16]. This is true for ZnSe crystals and thus a discrimination based on a pulse shape analysis is achievable. The shape of heat pulses also appears to be different for alpha and beta/gamma events [17]. As can be seen from figure 1, the TT changed the kinetics of luminescence; the radiative recombination of luminescent centers is demonstrating redistribution among different components of scintillation signal.

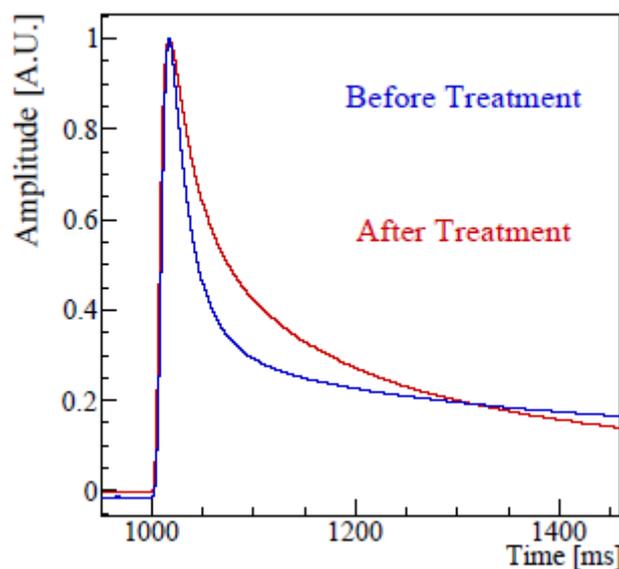


Figure 1. Impact of thermal treatment on the pulse-shape of the light signal for the ZnSe #2 (Color online).

Alpha vs. beta/gamma discrimination in scintillating bolometers may be visualized using light vs. heat scatter plot. In such a plot, alphas and betas/gammas populate separate bands by virtue of their differing light to heat ratio.

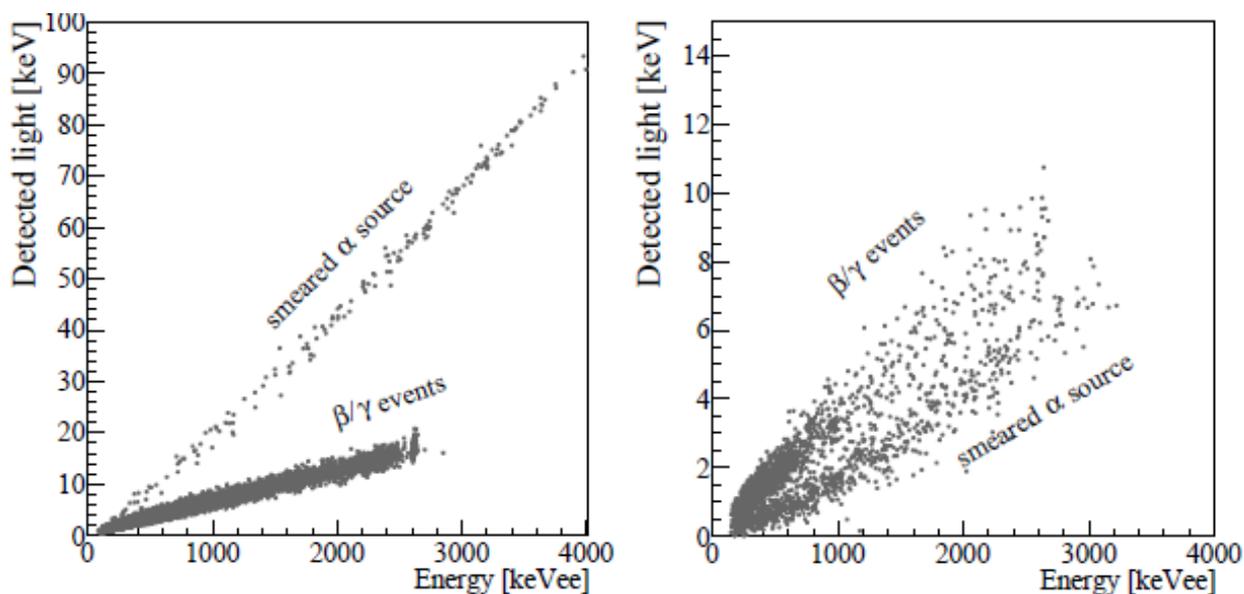


Figure 2. Light vs. Heat scatter plots corresponding to calibration measurements (external ^{232}Th gamma source and smeared ^{238}U alpha source) acquired with a ZnSe #3 before (left) and after (right) thermal treatment. The significant decrease in LY both for alphas and betas/gammas particles and the inversion of the QF for alphas is apparent.

As one can see from figure 2, the LY for alphas and betas/gammas is inverted. The LY for betas/gammas changed from 6.4 ± 0.1 keV/MeV for the “as-grown” ZnSe crystal to a value of 3.2 ± 0.5 keV/MeV for the ZnSe crystal after TT. While the LY for alpha particles decreased more significantly from 29.70 ± 0.17 keV/MeV for the “as-grown” crystal to 2.2 ± 0.2 keV/MeV for the thermally treated ZnSe crystals. It should be stressed, that the LY is changed not only for external alpha particles of calibration source, that reacts with a thin layer of the crystal surfaces, but leads to the significant reduction of the LY for gammas in 2-3 times, which interacts with the entire volume of the crystal. Based on this we can conclude that TT changes the bulk properties of the ZnSe crystals. Such a situation where the characteristic LY values for different type of particles become very close is unacceptable for experiments that rely on the this parameter for particle discrimination.

5. Discussion and summary

The optoelectronic properties of A^2B^6 semiconductors, as e.g. ZnSe, are mainly determined by their defects structure caused by deviation of stoichiometry and the presence of impurities. Despite the huge number of articles dedicated to study the luminescence centers in ZnSe crystals, the interpretation of its luminescence spectrum is still contradictory [18,19,20].

Since all crystals used in these measurements were produced from high purity initial materials (99.9995% grade) we can exclude the presence of significant amount of impurities that can be responsible for the formation of non-radiative recombination centers, defects, and its complexes. But, recalling that the main emission band of ZnSe peaks at 630 nm, the induced light is supposed to be due to the radiative recombination process that occurs within $\{\text{Zn}_i\text{-V}_{\text{Zn}}\}$ complex [21]. In terms of this model the electron recombines with the hole captured by the V_{Zn} center. The annealing of ZnSe crystal at high temperature destroys such complexes due to the high mobility of Zn atoms diffusing out of the crystal volume. But single defects of V_{Zn} are thermally unstable, and in order to compensate charge can forms a thermally more stable and immobile $\{\text{V}_{\text{Zn}}\text{-I}_{\text{Se}}\}$ complex. However, this situation looks similar to the annealing of ZnSe crystals in Te or Se vapors, i.e. introduction of isovalent impurities, when quenching of luminescence is interpreted by the formation of recombination center, which cause the strong photo-induced bleaching.

The reduction of ZnSe specific resistivity after thermal treatment indicates appearance of large amount of interstitial Zn, which is the origin of charge carriers, namely electrons. The same effect is observed after annealing of ZnSe crystals in Zn vapors. This fact of the specific resistivity reduction also demonstrates the thermal instability of the $\{Zn_i-V_{Zn}\}$ defects complex which was destroyed by annealing at 900°C. Based on our observations, we conclude that the $\{Zn_i-V_{Zn}\}$ defects complex is responsible for the fast radiative recombination process, while formed after thermal treatment defects complex, most probably the $\{V_{Zn}-I_{se}\}$, demonstrate quenching of luminescence and enlarge kinetics of radiative recombination process. The details on the nature of defects, which are responsible for the luminescence in ZnSe crystals, definitely needs further investigation.

Moreover, it should be mentioned, that scintillating bolometers are suitable tool in order to study fundamental properties of materials at low temperatures. Indeed, this technique is able to evaluate which part of the deposited energy is converted into emitted light, and which is dissipated in the crystal in form of heat.

We have demonstrated that the anomalous QF for alpha particles in ZnSe crystals is due to a certain type of defect in crystal structure and can be adjusted for a various applications via dedicated thermal treatment. Finally, the “as-grown” ZnSe crystals without any kind of thermal treatment are used for the CUPID-0 experiment to avoid a deterioration of their scintillating and bolometric properties.

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