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Slow components and afterglow in PWO crystal scintillations

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Abstract

The origin of the slow component and of the afterglow in PWO scintillation is discussed. It is shown that their presence depends strongly on the growth conditions and originates from specific impurities and host point-structure defects. The influence of the scintillation kinetics parameters on the detection properties of PWO crystals is discussed. © 1998 Elsevier Science B.V. All rights reserved.

1. Introduction

Lead tungstate crystal (PWO) is one of the prospective scintillating materials to be used in the new generation of particle physics experiments at the large hadron collider (LHC) and at other facilities. It is a suitable γ-quanta detector with a satisfactory energy response in a wide energy range [1], and allows a time resolution better than 130 ps [2]. Lead tungstate crystal has been under investigation since the 1940s but it has only been under develop-

ment as a scintillator for high-energy physics applications for the last four years [3]. Significant progress in the improvement and understanding of PWO scintillation properties has been achieved [4–6], but a production technology that delivers scintillating crystals with homogeneous parameters acceptable for users is still not well established. The main source of the technological difficulties is the greater sensitivity of the crystals produced to the variations in the growth conditions. This is due to nontrivial scintillation mechanisms [7] caused by the combination of regular and irregular radiating centres. The relative contribution of different types of centre to the scintillation light yield varies with particular changes in technology, and is very sensitive to the purity of the raw materials. The influence

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of the most dangerous impurities on the scintillation properties of PWO crystals is described in Refs. [8–10]. Another reason is the structural peculiarities of lead tungstate crystals. Recently, it was shown [8] that in the PbO–WO$_3$ system a new stable compound close to the pure sheelite crystal structure [11,12] exists. It has physical properties close to the scheelite type of PbWO$_4$ crystal but has a lead deficiency. The competition of these two crystal phases during the growing process creates defects in the final crystals. The investigation of PWO crystal series, grown by different producers and obtained from specified raw materials, has shown that the influence of some impurity ions and host matrix point-structure defects dominates in the changes of the PWO scintillation parameters from crystal to crystal or even along the crystal ingot. A joint analysis of the influence of point structure defects in the matrix and of specific impurities on the afterglow and radiation damage for scintillating PWO crystal are given in Ref. [13]. In this paper we describe their influence on the scintillation kinetics of PWO crystals. The influence of the scintillation kinetics parameters on the PWO detection properties will also be discussed.

2. Samples and experimental methods

All crystals analysed were grown from platinum crucibles in an atmosphere close to that of air in composition, using the Czochralski technique. More details on the crystal growth are given elsewhere [14]. To clarify the role of the Mo impurity on the PWO scintillation properties, the crystals doped with Mo or obtained from oxides specially cleaned of Mo and some other impurities, were grown in separate crucibles using a step-by-step crystallization. The samples investigated in the present work were prepared from the top parts of the ingots. Other parts of the ingots were used to prepare scintillating elements for the beam tests of CMS PWO R and D at CERN. Equipment and techniques used for spectroscopic and scintillation parameter measurements are described elsewhere [5]. The spectroscopic and scintillation parameter measurements, as well as Thermo Stimulated Luminescence (TSL) spectra in the 80–300 K temperature region, have been investigated. The crystals were activated at liquid nitrogen temperature with a hydrogen lamp equipped with a sapphire window. The temperature increase rate during the recording of the TSL spectra integral was 0.5 K/s. The crystal structures of the samples were studied according to the method described in Ref. [11,12]. It was determined that they are sheelite-type crystals.

3. Luminescence and scintillation kinetics of PWO crystals

3.1. Slow component sources in PWO scintillations

The scintillation kinetics of PWO crystals is characterized by three main stages. Their contribution to the total kinetics depends on the crystal-growth conditions but can be stabilized. A fast component with typical decay time $\tau = 2$–4 ns is emitted. It is due to the green and blue luminescence region [14] and usually accounts for half or more of the scintillation light. The medium-fast components with $\tau = 10$–20 and 60–100 ns, which contribute to the scintillations for about 50% or less, are mainly connected with green radiating centres. Finally, slow components with $\tau = 100$–1000 ns appear due to various reasons discussed below. Sometimes their contribution can vary up to some tens of per cent. An afterglow with components in the microsecond region also exists in the PWO scintillations. Its level depends strongly on the technological peculiarities and is well characterized by the α parameter introduced in Ref. [15].

The scintillation in PWO crystals is produced by the contribution of regular WO_4^{2-} and by two types of $(WO_3 + F) + Pb^{2+}$ groups [4,7]. The last radiating centres are defect centres in pure sheelite crystals but in sheelite-type crystals they also exist thanks to peculiarities in the crystal structure. The regular centres emit in the blue light range ($\lambda_{max} = 420$ nm) and the WO_3-based centres provide two overlapping green ($\lambda_{max} = 500$ nm) luminescence bands [14] under photoexcitation. Under ionizing radiation, excited scintillations appear as the superposition of at least these three

luminescence bands overlapping to give a broad luminescence band with a maximum near 500 nm. Thus, the scintillation kinetics of PWO is also the superposition of the contribution of at least three radiating centres.

Let us consider the possible sources of slow components and afterglow appearing in PWO scintillations:

1. PWO is not an exception among the crystals of the tungstate family, its intrinsic luminescence bands are strongly quenched by multiphonon interaction. Their brightness increases more than one order of magnitude and their decay times τ of kinetics come in the microsecond region with decreasing temperature [16,17]. The scintillation kinetics depends on the temperature according to the tendencies described above [4]. On the other hand, the Stocks shifts of the luminescence bands detected in PWO are more that $1 \times 10^4 \, cm^{-1}$ and lead to a strong quenching of the luminescence bands at room temperature. Owing to the quenching, the luminescence kinetics comes to the nanosecond region. Nevertheless, one has to take into account the slow components because the mechanism of quenching is casual. However, our measurements of the green and blue luminescence band kinetics under photoexcitation on a 1000 ns scale have shown that the relative amplitude of the components with τ in the range of some hundreds of nanoseconds to microseconds is smaller than 5×10^{-4} at least. However, even if the amplitude of the slow components is rather small, the number of the photons emitted through it in luminescence kinetics can, nevertheless, reach up to some per cent of the total luminescence light. This amount of the natural slow component's contribution appearing a priori is a minimal level of the slow component's presence in scintillations.

2. PWO is a member of the large tungstate family which contains a set of slow scintillators such as $CdWO_4$ and $ZnWO_4$ as well as crystalophosphors. They have luminescence bands located in the blue–green region with decay times of scintillation or luminescence kinetics in the microsecond region. There is also a class of molybdate compounds that are isostructural to wolframate

crystals. Some of them like $PbMoO_4$ have green luminescence under photoexcitation but do not scintillate [18]. It is obvious that divalent ions like Cd, Zn, Mg, Ca or Mo^{6+} which create the host matrices of the sheelite or wolframite types can influence the scintillation properties of PWO crystals through their isomorphic incorporation in the host. They can suppress existing radiating centres or create new ones with slow scintillations, or provide afterglow. However, among the mentioned impurities, only molybdenum impurity is usually present in the raw material for PWO crystal growth and as determined, is the most influencing doping even at the level of concentration of few ppm. Molybdenum ion is very close chemically to the tungsten ion and is rather hard to separate at the raw material production level. Moreover, the Mo impurity creates, in $PbWO_4$ crystals, specific electron capturing defects [19], the influence of which on the PWO scintillations is not yet investigated.

3. The PWO crystal is very sensitive to its crystal growth conditions. For the Czochralski method, the creation of defects in the ingot depends strongly on the parameters of the equilibrium between the melt, the already grown part of the crystal, and the isolated atmosphere of the oven chamber. Nonoptimal crystal growth conditions or step-by-step crystallization lead to dramatic changes of the stoichiometric ratio and, as a consequence, some specific defects will appear in the crystals. They influence most of the PWO physical parameters including scintillation.

3.2. Experimental data

Fig. 1a and Fig. 1b illustrates typical scintillation kinetics of PWO crystals grown at first crystallization from cleaned (a) raw materials and (b) doped with Mo (2.5 w% in the melt). The light yield of the doped crystal is three times less than that of the undoped one and the scintillation kinetics display a slow component and a afterglow contribution which are quite noticeable. The sample demonstrates an absence or suppression of the regular fast component in scintillation kinetics. Such a slow component and the afterglow are also
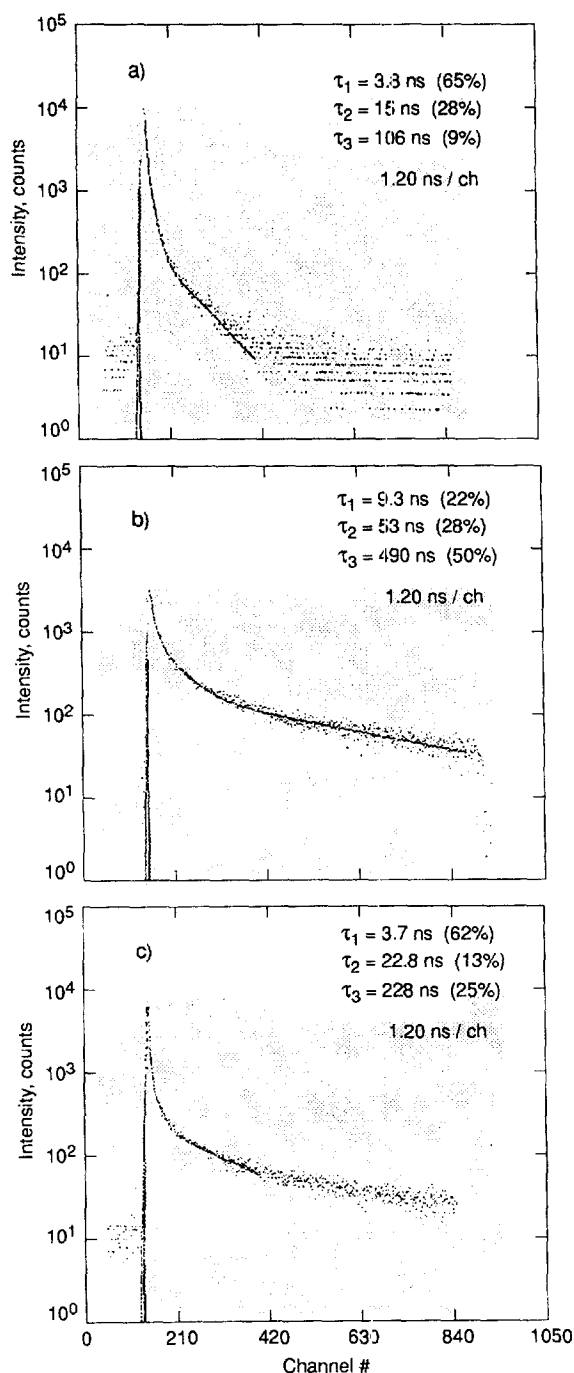
Fig. 1. Scintillation kinetics of the PWO crystals grown from cleaned raw materials (a); doped with Mo (2.5% by weight) (b), and containing Mo with (c) concentration 90 ppm. $T = 300$ K.

clearly identified in the scintillation kinetics of the crystals that contain an Mo amount of just a few tens of ppm, Fig. 1c.

On the other hand, crystals obtained from purified raw materials but grown in nonoptimized conditions after various recrystallizations also present the appearance of slow components. Fig. 2a gives the changes of the scintillation kinetics parameters with the number of the crystallization increase. The light yield of the samples (Fig. 2b) correlates with the increase of the slow-component contribution in the scintillation kinetics. It indicates that an additional source of excitations, which is transmitting energy to the radiating centres relatively slowly, appears in the crystals. The contribution of this source is increased with the increase of the crystallization number. Spectroscopic properties of these crystals also change with the number of the crystallization. Fig. 3 shows the spectra of the green luminescence excitation at room temperature of the crystals obtained in the first (a) and in the last crystallization (b) of the investigated series. In the last case the spectrum is shifted to the long-wave region. A new excitation band overlapping with the luminescence excitation band of WO_3-based centres with maximum around $\lambda = 312$ nm appears in the spectrum. Probably, a real maximum of this band is shifted in the short-wave region and a detected maximum appears due to the influence of fast growing absorption at the fundamental edge. Its relative intensity grows as the crystallization number increases and is correlated with the mentioned scintillation parameters. The relative change of the 312 nm excitation band intensity with the number of the crystallization is shown in Fig. 4.

TSL spectra also depend on the purity of the raw materials and the number of the crystallization. The intensity of the TSL increases with the number of the crystallization and becomes the most intensive at the temperature 120 K. However, it has practically the same shape and intensity in the 200–280 K region. TSL data are presented in Fig. 5. The analysis of the two PWO characteristic peaks that appear near 120 and 250 K has been made to determine the frequency factor S, the activation energy E, and the time of release from the traps. These data are presented in Fig. 6.
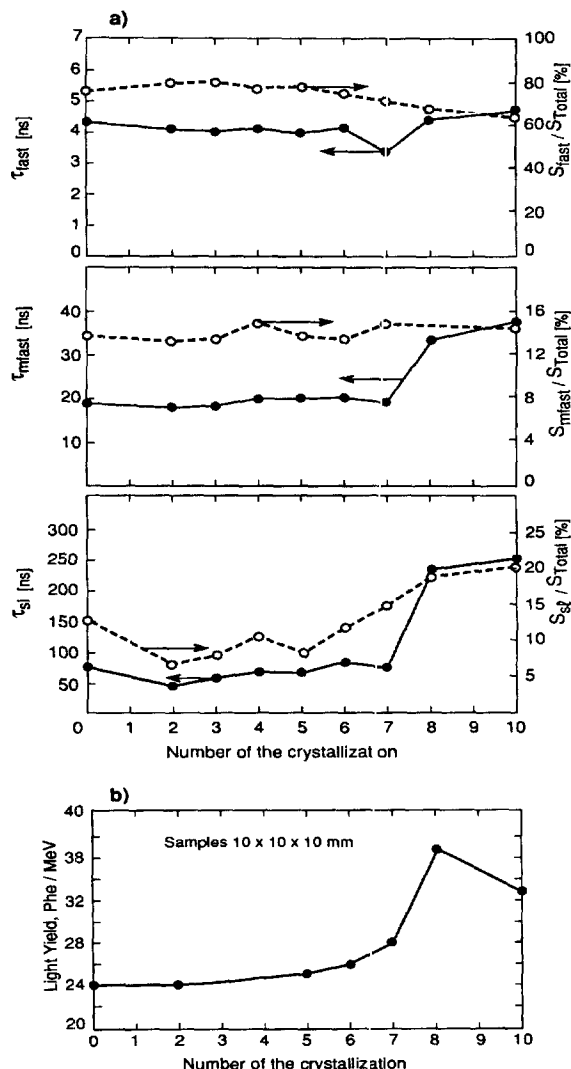
Fig. 2. Parameters of the scintillation kinetics τ and contribution of the different scintillation components to the total kinetics $S_{(sl,\ mfast)} / S_{Total}$ (a) and light yield (b) with the number of the crystallization. $T = 300$ K.

Fig. 3. Excitation spectra of the green luminescence of PWO crystals of first (a) and last crystallization (b). $T = 300$ K.
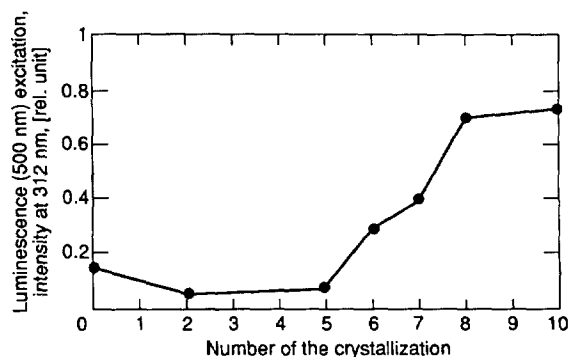
Fig. 4. Relative change of excitation intensity at 312 nm for the green luminescence with the number of the crystallization. $T = 300$ K.

No clear correlation has been found between the slow components' appearance in PWO crystals and the changes of the TSL spectra in the 80–300 K region. Following the common tendencies it may be stated that:

1. The defects which generate TSL bands in the 80–300 K temperature region are not connected with the centres causing slow components in

scintillations. They provide just an afterglow, the fraction of which grows up with the increase in the crystallization number. Defects have a rather small frequency factor S and the parameters τ of their release are more than tens of microseconds.

2. The total intensity of the TSL spectra increases with the increase of the recrystallization number. The main changes are due to intensity growth of the 120 K peak.

3. Additional doping with ions which make an isomorphic change to Pb or W ions leads to considerable changes in the TSL spectra. The
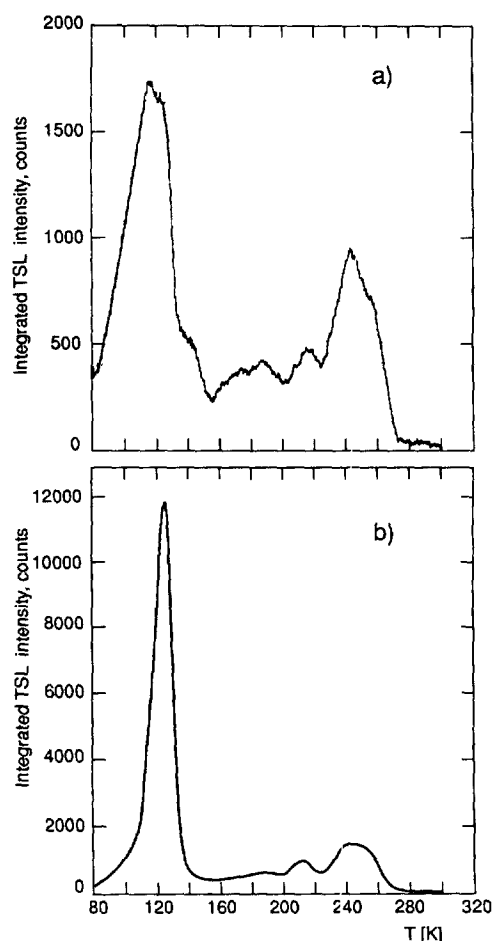
Fig. 5. Integrated thermostimulated luminescence spectra of PWO crystals obtained from the first (a) and last (b) crystallization samples.

intensity of the existing bands are redistributed but total intensity is at the level of undoped crystals.

3.3. Discussion

Let us consider the influence of Mo impurity on the scintillation kinetics and afterglow in PWO. Mo impurity creates in $PbWO_4$ crystals strong electron capturing centres $MoO_4^{2-} + e \rightarrow MoO_4^{3-}$ [19]. At small concentrations these centres exhibit a strong peak in the TSL in the region 200–300 K. With a small content in the melt the distribution

coefficient of the Mo in the crystal is close to 1, thus the MoO_4^{3-} defects' concentration will be of the same order at different crystallizations. This is exactly detected in the TSL spectra in the 200–300 K region for crystals of different crystallizations. With Mo concentration increase this peak intensity falls but new peak appears near 160 K. On the other hand, the MoO_4^{2-} complex has a lower energy of the 2e state than one of WO_4^{2-}. So both types of radiating centres, but mainly blue luminescence emitting ones, are quenched by MoO_4^{2-} groups. Thus, the influence of Mo impurity is twofold. At small concentration it generates afterglow in the seconds region. With the increase in Mo concentration, afterglow due to Mo comes to the same tens of microseconds region, and quenching of the scintillations becomes sufficient. Owing to preferential quenching of the regular groups' luminescence, the fraction of the fast components in Mo-doped crystals is rather less. Additional Mo content leads to full scintillation suppression.

The creation of defects in both PWO structures is of another kind and has a common peculiarity. Stoichiometric variations appear due to a Pb leakage from the melt which is compensated by an oxygen deficiency. A correlated leakage of the Pb and O also exists. In sheelite-type crystals Pb leakage generates cation vacancies V_{Pb} and, consequently, some types of centres based on O^- holes appear. Oxygen leakage also generates group centres $(WO_3 + F) + Pb^2$ in addition to the structurally existing WO_3. The correlated leakage of the Pb and O also generates WO_3 groups but of another kind than the ones mentioned above. They are probably $(WO_3 + V_{Pb} + V_O) + Pb^{2+}$. The energy level structures of the centres listed above are, of course, different. For instance, one can have lower metastable excited energy states such that conditions for the sensitization of the $(WO_3 + F) + Pb^{2+}$ green luminescence appear. Nevertheless, we have no direct confirmation that the $(WO_3 + V_{Pb} + V_O) + Pb^{2+}$ centre causes the appearance of the new excitation band at $\lambda = 312$ nm in the excitation of the green luminescence, but the amount of these centres should increase with the repetition of the crystallization. This trend is correlated with the increase of the relative intensity of the 312 nm band in excitation
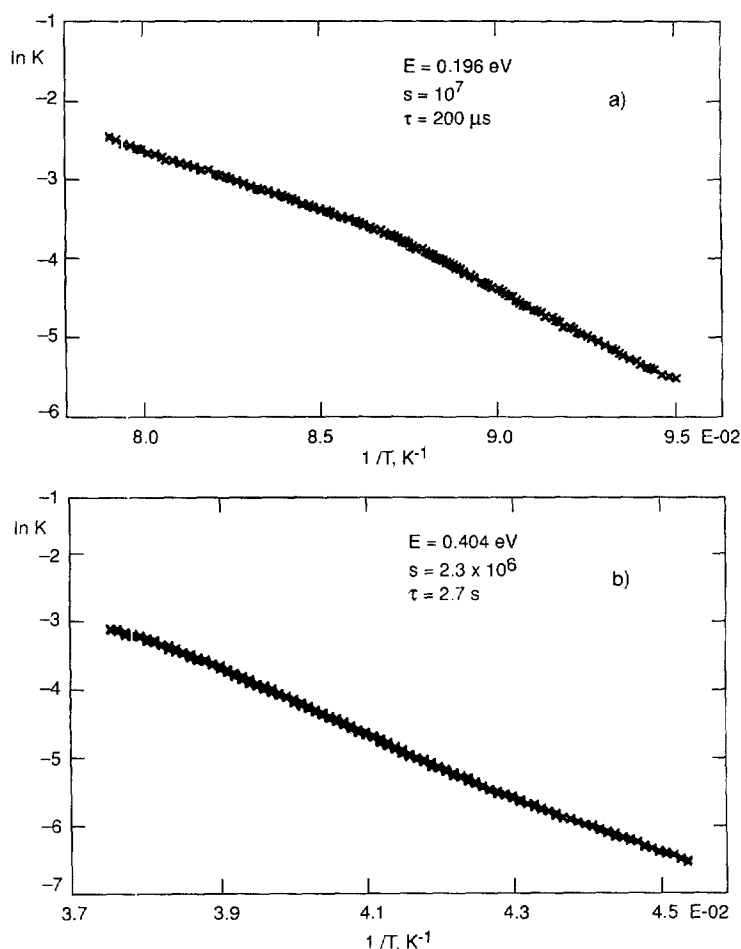
Fig. 6. Presentation of the 120 K (a) and 250 K (b) bands in the ln scale versus $1/T$.

spectra. Thus, we infer that this. or such a type of the point structure defect centres which appears with lead leakage from the melt during the crystal growth, sensitizes the green luminescence through multipole Coulomb interaction or tunnelling. Anyway, we have registered such a phenomena for crystals obtained from different raw material, so we concluded that it is not caused by the accumulation of the separated impurities. This hypothesis needs further confirmation and more detailed calculations of the point defects' energy level structure in lead tungstate crystal.

The crystal features quoted indicate that an accumulation of the different defects in PWO crystals

happens with the increasing number of the crystallization. Between them those which are associated with a peak at 120 K give an afterglow with $\tau = 200$ µs. Other bands in the TSL sited between the 120 and 250 K bands grow slower with the increase of the crystallization number and do not contribute significantly to afterglow.

4. Influence of PWO scintillation kinetics on PWO detection properties

One way to increase the light yield of PWO scintillators is to decrease the temperature in the

Table 1
Parameters of the samples investigated

Sample	λ_{max} of the radioluminescence (nm)	Light yield at 291 K (with PM (XP2262) gate 1000 ns) (Ph.e./MeV)	Decay times at 291 K [(ns) (%)]		
1	450	40.6	6.4 (61)	27.5 (30)	139 (9)
2	430	44.6	5.6 (58)	24.8 (31)	124 (11)
3	500	42.6	5.57 (58)	26 (32)	141 (10)
4	500	38.4	4.3 (63)	42 (15)	408 (22)

detecting units. It seems that the optimal operational temperature for PWO is in the region 273–283 K. However, the scintillation kinetics has an influence on the performance of the PWO-based detectors through temperature dependence of the light yield of the components with different decay times. The following samples with the parameters quoted in Table 1 were chosen for the investigation of temperature dependence of the light emission within a set of specific time gates (30, 50, 100, 300, 1000 ns) for several crystals. Results are displayed in Table 2.

Samples 1–3 have dimensions $2 \times 2 \times 2$ cm^3, all sides polished; sample 4 was $1 \times 1 \times 1$ cm^3, two sides polished. According to their luminescence difference they form four groups: 'aqua-blue fast' (1); 'blue fast' (2); 'green fast' (3); 'green slow' (4). Crystals used for the sample preparation were grown at the different stages of the PWO technology optimization. Crystals 1–2 were crystals of pure sheelite type and crystals 3–4 were of sheelite-like type obtained from the first and last crystallizations. This distribution covers all types of crystals under investigation.

The scintillation kinetics of the samples in terms of the kinetics integral at different temperatures are plotted in Fig. 7a–Fig. 7h. This kind of kinetics presentation is convenient to show the amount of light emitted in the different time gates mentioned above. The extrapolated time, when 99.9% of light is emitted, is also indicated.

Samples 1–3, which are 'fast', show normal behaviour of the light yield with a decrease in temperature. The relative amount of light registered within short gates (30, 100 ns) decreases in agreement with Ref. [4], but the absolute amount of light within the same gates as well as total light yield is increased.

The light yield temperature dependence is $-2 \pm 0.2\%/C$ [20]. Sample 4 which is 'green slow' demonstrates, on the contrary, an opposite trend in the relative amount of light change, although the absolute amount of light emitted is increased. Also this sample shows a smaller light yield temperature dependence which is $-1.5 \pm 0.2\%/C$. These features of the slow crystal are due to the opposite tendency acting on the green luminescence light yield: on one hand, the decrease in temperature leads to the light yield growth but the probability of green luminescence sensitizing described above decreases with temperature. Thus, the mentioned peculiarity of the 'green slow' crystals indicates that they have to be clearly separated from the fast crystals at the time of certification and delivery.

5. Conclusions

The origin of the slow components in scintillations and afterglow in PWO crystals have been investigated.

It is determined that accumulation of the different defects in PWO crystal happens with the increasing number of crystallization. Among these defects which are associated with a 120 K peak in TSL give an afterglow with decay time about 200 μs. Another defect type, associated with the point structure defects appearing with the lead leakage from the melt during the crystal growth, is responsible for the slow components in PWO scintillations.

A Mo impurity at the level of some tens of ppm creates in PWO crystals strong electron capturing centres which afterglow in seconds region. Mo impurities lead to the quenching of the scintillations of

Table 2

| Crystal	Temp. (K)	Gate time (ns)	Integral	Ph.e./MeV	Light fraction in gate (%)
		30.0	71 352.4	31.0	73.0
		50.0	80 080.6	34.8	82.0
		100.0	87 650.2	38.1	89.7
	288.8	300.0	93 255.0	40.5	95.5
		1000.0	96 517.4	42.0	98.8
1		2364.7	97 589.7	42.4	99.9
		30.0	83 360.6	36.0	68.5
		50.0	95 805.6	41.4	78.7
		100.0	107 339.3	46.4	88.2
	279	300.0	116 012.4	50.1	95.3
		1000.0	120 391.4	52.0	98.9
		2231.6	121 633.8	52.5	99.9
		30.0	53 487.7	32.0	70.4
		50.0	60 299.7	36.1	79.3
		100.0	66 315.4	39.7	87.2
	290	300.0	71 123.3	42.6	93.5
		1000.0	74 398.2	44.5	97.9
2		2996.5	75 951.1	45.5	99.9
		30.0	61 403.6	36.7	67.3
		50.0	71 145.7	42.5	78.0
		100.0	80 189.3	47.9	87.9
	278.6	300.0	87 367.8	52.2	95.7
		1000.0	90 743.7	54.2	99.4
		1609.3	91 164.8	54.4	99.9
		30.0	48 003.1	31.4	72.1
		50.0	54 220.3	35.4	81.5
		100.0	59 601.5	39.0	89.6
	290	300.0	63 701.8	41.6	95.7
		1000.0	66 024.1	43.2	99.2
3		1850.9	66 470.9	43.4	99.9
		30.0	56 180.4	36.7	67.7
		50.0	64 940.6	42.4	78.3
		100.0	73 012.2	47.7	88.0
	278.7	300.0	79 405.5	51.9	95.7
		1000.0	82 637.5	54.0	99.6
		1363.5	82 857.1	54.2	99.9
		30.0	30 020.8	21.8	54.1
		50.0	33 164.9	24.0	59.8
		100.0	37 525.9	27.2	67.7
	289.2	300.0	44 519.1	32.3	80.3
		1000.0	52 557.1	38.1	94.8
4		3101.2	55 406.3	40.1	99.9
		30.0	35 355.7	25.8	54.1
		50.0	39 364.2	28.7	60.3
		100.0	44 832.4	32.7	68.6
	277.5	300.0	53 685.0	39.2	82.2
		1000.0	63 110.1	46.1	96.6
		2441.0	65 249.4	47.6	99.9
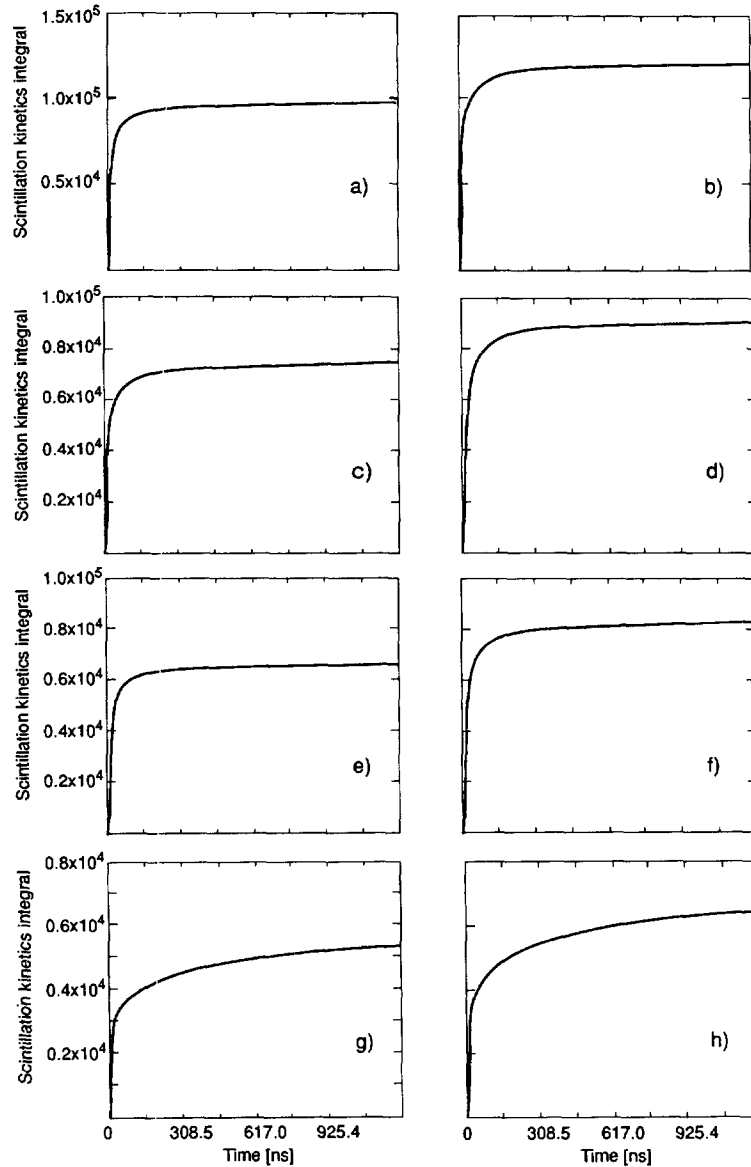
Fig. 7. Integral of the scintillation kinetics of the crystals mentioned in Table 1 at different temperatures, a-1 (288.8 K), b-1 (279 K); c-2 (290 K), d-2 (278.6 K); e-3 (290 K), f-3 (278.7 K); g-4 (289.2 K), h-4 (277.5 K).

the regular tungstate groups, decreasing the fraction of the fast components in the scintillations.

Crystals which have a sufficient level of slow components in scintillations have a smaller light yield temperature dependence relative to the fast scintillating crystals. For the construction of high-precision electromagnetic calorimeter such

crystals have to be clearly separated from the fast crystals.

Acknowledgements

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