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Nuclear Instruments and Methods in Physics Research A 486 (2002) 474–481

NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A

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Concentration dependence of the light yield and energy resolution of NaI:Tl and CsI:Tl crystals excited by gamma, soft X-rays and alpha particles

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Abstract

Based on the analysis of light yield dependence on activator concentration for NaI:Tl and CsI:Tl excited by γ-rays, soft X-rays and α-particles, an explanation of the effect of energy resolution enhancement with the rise of Tl content has been proposed. Based on the concept regarding the electron track structure, we proposed an alternative explanation of the intrinsic resolution value. The concept does not take into account the non-proportional response to electrons of different energies and is based on the statistic fluctuation of scintillation photon number formed outside and inside the regions of higher ionization density.

PACS: 29.30.Kv; 72.20.Jv; 78.60.Ya; 78.70.−g

Keywords: Scintillator; Activator concentration; Non-proportionality; Light yield; Intrinsic energy resolution

1. Introduction

The concentration dependence of the light output (L) is a very important characteristic of scintillation systems since the activator content (C) is the only substantively controlled technological parameter which also defines properties of scintillators. From the practical point of view, the knowledge of the $L(C)$ dependence is necessary for the determination of optimal C values. The analysis of known dependences for the standard energy 662 keV allows to conclude that for both NaI:Tl [1,2] and CsI:Tl [3], these dependences are, in general, similar and essentially depend on the growth process, first of all on melt-mixing conditions [1], pulling rate [1,4] and growth atmosphere [5]. The problem of an optimal activator content is not solved entirely, in particular for CsI:Tl scintillators [3].

Knowledge of the $L(C)$ dependence only for 662 keV is insufficient from the scientific point of view. Therefore, similar dependencies for excitation by protons, α-particles [6] and even fission fragments [7] are used for the analysis; however, there are very few papers on this subject. Concerning the low-energy X-rays, we know only two papers [8,9] where $L(C)$ for 5.9 and 60 keV have been studied. The overall conclusion from these results [10] is of great interest: curve $L(C)$ for NaI:Tl is invariable in the range of 10–662 keV and varies appreciably for 5.9 keV. To understand the scintillation pulse formation process more exactly, it is important to know similar dependencies of the

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0168-9002/02/$ - see front matter
PII: S0168-9002(02)00756-8

energy resolution (R). However, data on $R(C)$ became available only recently [2,11].

The problem concerning the character of $L(C)$ dependence for different energies is linked with the non-proportionality (NP) of response to electrons of different energies, since it is possible to compensate NP of response partly and to achieve a linear response in the range of 5.9–60 keV [12] by increasing C. If the concept of NP with regard to intrinsic scintillator property [13] is refuted [14] completely or partly, for example, for crystals with a quite high content of luminescence centers, we will have the only alternative to explain the $R(C)$ dependence character and minimum values of the intrinsic resolution (R_i) starting from the data concerning $L(C)$ for different energies and ionization density. In the work presented here, probably, the first attempt was made to explain the intrinsic resolution of NaI:Tl and CsI:Tl based on an alternative approach which coincides with Gwin and Murray [15] in a sense but does not relate to NP of response [16].

2. Basic model concepts

Introduced theoretically in Ref. [17] to explain the $R(E_\gamma)$ dependence, a model concept of scintillators with micro- and macro-inhomogeneities was used to analyze light output and energy resolution data. Following the model, let us imagine a crystal consisting of two parts: scintillating and non-scintillating regions. A typical region size is designated as q, with $q \ll l$, where l is the range of an ionizing particle of energy E (for a 1 MeV electron $l \sim 1$ mm in NaI). The number of micro-inhomogeneities (MI) in the track of particle is $m = l/q$: let us introduce the probability p of track and scintillation part intersection, and $(1-p)$, the same for parts with $L \ll L_{max}$. As is shown in Ref. [17], the change of light yield for a crystal with MI, $\Delta L = (L_{max} - L)$, can be written as:

$$\Delta L = (1-p)L_{max} \quad (1)$$

and the intrinsic resolution can be expressed by:

$$R_i^2 = 5.56(1-p)/mp. \quad (2)$$

Physically, $(1-p)$ is the quota of volume occupied by non-scintillating regions. The model [17] should be supplemented with the concept of the probable origin of MI to connect it with the $L(C)$ and $R(C)$ dependences. The reason of MI formation could be the decay of a solid solution. According to the data of electron microscopy, in alkali halide crystals doped with Tl^+ [9,18] and Na^+ [19,20], the spinodal decay of solid solution was observed, namely some parts of the crystal will be enriched with an activator due to the depletion of the others without a defined boundary between different regions of the crystal. MI is understood as parts enriched in a dopant, the decoration character of which considerably differs from that typical for the main volume of the crystal [19]. The activator is mainly represented as aggregated centers in the enriched regions; therefore, the light output is reduced considerably while the afterglow is distinctly increased [9]. Obviously, the parameter $(1-p)$ linearly depends on C in a first approximation.

As is seen from Fig. 1, the response to particles with $l \gg q$ and to those with $l \ll q$ is different. For electron with $l \gg q$ MI, volume should be subtracted from the luminescent track volume. Another situation is for particles with $l \ll q$: this main part will be recorded with the maximum light yield; while a small part of them being in the MI will be of no importance for the full absorption peak. Since $L \neq 0$ even for non-activated NaI, these last events will be recorded with quite low output corresponding to the noise region or valley in the amplitude spectrum. Peak-to-valley ratio becomes worse and the registration efficiency (ε) of particles decreases; ε will be reduced in proportion to $(1-p)$.

Apart from the model concept concerning MI of activator origin, we also use the concept of dE/dx micro-inhomogeneity in electron tracks, in particular, the concept of regions of higher ionization density. Schemes of 1 MeV electron track fragment [21] are shown in Fig. 1. Circles and hatching indicate the regions where dE/dx is extremely high. The particle forms separate electron–hole (e–h) pairs between the circles. More explanations are given in Section 6.

3. Experiment

Experimental details and all the necessary references concerning the crystal growth and

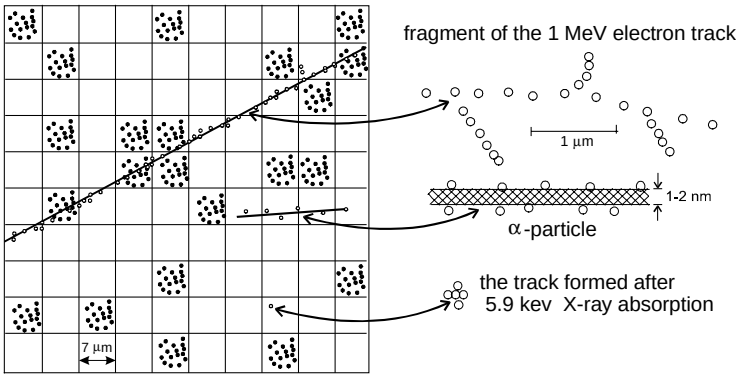

Fig. 1. The model of crystal with micro-inhomogeneity (left). The calculated structure of track fragment for 1 Mev electron and 4 MeV α-particle (right). An explanation is given in the text.

detector-making are given in Ref. [1–4]. In this paper, we added measurements of the $L(C)$ dependences for CsI:Tl at the excitation by 5.15 MeV α-particles and 5.9 keV X-rays to the data published previously.

In addition to the data concerning crystals grown in vacuum by Stockbarger technique (crystals of the VC-type) [1,2] or in inert atmosphere by modified Kyropulos technique (KC-type) [3], we represent data for crystals grown in hermetic quartz ampules in reaction gas atmosphere (at partial oxygen pressure $\sim 3 \times 10^4$ Pa, OC-type). Although NaI:Tl and CsI:Tl of OC-type are grown under purer conditions, the growth process of these crystals is similar to that in the dry air atmosphere in many aspects and has been practiced so far [11].

4. NaI:Tl Scintillator

According to Panova and Kudin et al. [1,2], $L(C)$ and $R(C)$ dependences for excitation by 5.9 and 662 keV quanta and 5.15 MeV, α-particles are shown in Fig. 2 for crystals of VC-type (curves 1–4). $L(C)$ for 662 keV energy (curve 1) is characterized by the presence of a plateau in the interval $0.022 \leqslant C \leqslant 0.073$ mol% of Tl. A further increase of C up to 0.7% results in the decrease of L up to 37.5% from the maximum value. Data scattering in the region of concentration quenching of L is much higher than that in the region of plateau. Within this scattering the character of L-quenching is approximated by a straight line. Curve 4

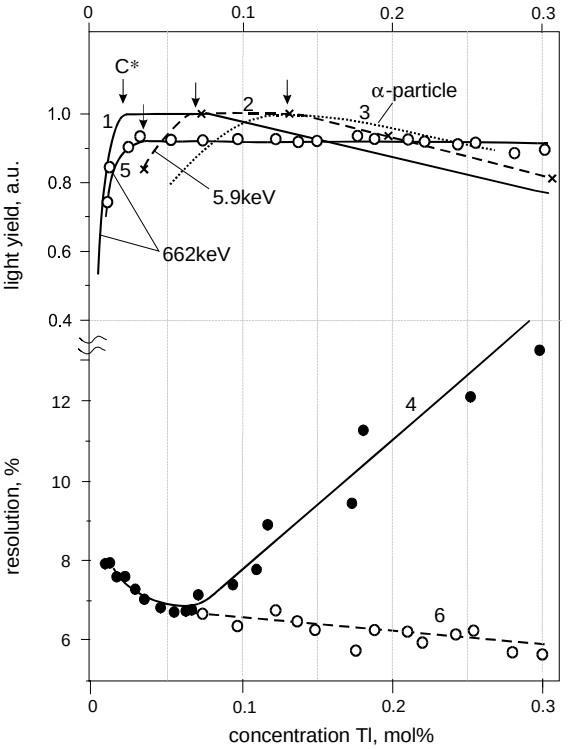

Fig. 2. The dependencies $L(C)$ (top) and $R(C)$ (bottom) for NaI:Tl of VC- (curves 1–4) and OC-type (5,6). Excitation by 662 keV γ (1,4–6), 5.9 keV X-rays (2) and 5.15 MeV α (3). The initial point C^* of the L-plateau is, respectively, 0.022 (1), 0.073 · (2), 0.13 (3) and 0.035 (5) mol% of Tl.

shows the $R(C)$ dependence for the energy 662 keV, it is antisymbate to curve 1. R values are minimal for the detectors with the maximum light yield. At the same time, the $R(C)$ dependence

exhibits some peculiarities. First, the values of R within the plateau of L are unstable, they decrease gradually and the best spectrometric characteristics of the detectors are reached at the end of the plateau. Secondly, the reduction of R in the interval of concentration quenching is not defined by the decrease of L. For example, at $C = 0.3\%$, when the light output is reduced by 23%, R reaches 14%. If R was determined by only the change in L, it would have only increased from 6.3% to 6.8%. We succeeded in defining the change of R with the rise of C in the limited range of concentrations for VC crystals; the resolution becomes worser at $C > 0.3\%$ that it is impossible to define it correctly. High values of R in the quenching interval testify to the MI formation in crystals of the VC-type.

Let us analyze the data shown in Fig. 2 using the MI model. According to curve 1 $\Delta L = 0.77$ for $C = 0.3\%$; then $(1-p) = 0.23$. Curve 5 indicates that $R_i = 0.14$; then, according to Eq. (2), $m \approx 85$, the average size of MI is $q \approx 7\,\mu m$. This corresponds exactly to the data of electron microscopy (4–9 μm) in crystals with higher Tl content which have regions where the decoration character differs from the main volume [8,18].

The $L(C)$ dependence for 5.9 keV X-rays is represented by curve 2 in Fig. 2 and differs from that described above. Maximum values of the light output will be achieved at $C \geqslant 0.073\%$. The plateau interval reaches 0.016%, then there is a quenching one. Thus, the plateau of curve 2 overlaps the quenching interval of curve 1. For 5.9 keV X-rays, the probability of resonance interaction with the electrons of L_1-iodine shell is very high. According to the cascade model of the energy exchange [22], a photoelectron with 0.713 keV energy ($l \approx 7$ nm), an Auger-electron of 3.445 keV ($l \approx 60$ nm) and two quanta with $E = 0.871$ keV are formed in this case. As it can be seen from the crystal model of Fig. 1, only the 5.9 keV X-rays absorbed in scintillating parts will be recorded in the full absorption peak. The events in the regions of MI will be recorded with quite a low yield. When increasing $C > 0.1\%$, the position of the full absorption peak shifts insignificantly in the amplitude spectrum but worsening of ε and peak-to-valley ratio are distinctly seen.

The dependence of the observed NP of response of NaI:Tl on Tl concentration is the consequence of the fact that the curve 2 in Fig. 2 reaches its maximum values at only $C \geqslant 0.73\%$ [10]. According to Zagariy and Vyday [12], the NP of L/E between 5.9 and 59.6 keV for $C = 0.03\%$ reaches 8% but for crystals with $C = 0.1\%$, it decreases up to 2% when the light output is maximum, as shown in curve 2 (see also Fig. 4). This fact allowed to predict the character of the dependence $R(C)$ [10]: with the rise of C, the resolution for $E = 662$ keV will decrease within the L_{662} plateau.

Experimentally, the enhancement of R was observed for the first time in OC crystals [2] where the concentration quenching interval was shifted to a higher C, i.e. L plateau was broadened up to 0.3% (see Fig. 2, curve 5). The peculiarity of this crystal was the lower light output ($L_{max} = 0.92$) because of the harmful effect of oxide compounds, oxygen as O_2^- is assumed to have a negative influence on the luminescence of Tl^+ [9,23]. The principle of negative influence of dipole isovalent impurity center on luminescence was considered in Ref. [24] based on the example of OH^- ions. Additional non-radiative recombination channel in OC crystals induces the shift of the initial point of $L(C)$ plateau up to $C = 0.035\%$. Despite lower L values, $R(C)$ for OC crystals differs fundamentally from VC ones. R_i (curve 6) have a tendency to decrease in the whole interval of the extended plateau. MI was not revealed in these crystals by electron microscopy analysis using gold decoration technique.

$l = 23\,\mu m$ in NaI for 5.15 MeV α-particle, it is by a negligible margin more than q. For VC crystals, the dependence $L_\alpha(C)$ is represented by the curve 3 normalized to 1.0 although the specific yield L/E is lower in this case (best value of α/γ-ratio is 0.7 [25]). When the parameter $(1-p)$ is low, the majority of α-particles will be recorded with the maximum light yield; with the increase of $(1-p)$ a part of them will intersect MI regions. In this connection, L_α will not change significantly but the resolution will dramatically drop because of the asymmetric broadening of the full absorption peak at a low energy. Thus, for VC crystals with $C = 0.13\%$, the resolution is $\sim 3.4\%$, and $\sim 15\%$ for $C = 0.3\%$. The light output being

measured for α-particles and soft X-rays will drop when Tl^+ concentration in the scintillating regions decreases due to Tl depletion. According to Kudin et al. [26] this could only occur at $C > 0.13\%$. The experiments carried out show this effect for VC crystals (see curves 2 and 3, Fig. 2).

The data given in this paper permit to conclude that the model considered adequately describes the properties of NaI:Tl crystals in the interval of concentration quenching. Thus, with the increase of the activator content, NaI:Tl crystals will have a better resolution. From a practical point of view, this effect is used only in the production of 1 mm thick X-ray detectors. It is connected with a negative peculiarity of the material—the luminescence spectrum is partly overlapped by Tl^+ absorption, $(Tl^+)_2$ dimers and complex centers, the number of which depends on C. The light collection coefficient τ strongly depends on C and detector size because of the reabsorption; therefore, Tl-concentration increase for big-size scintillators is unwanted.

5. CsI:Tl scintillator

Stoke's shifting of the activator luminescence in CsI:Tl crystal is wide; luminescence and Tl^+ absorption do not overlap significantly. The fact that the afterglow of CsI:Tl contrary to NaI:Tl has a non-activator origin, and even has a tendency to decrease with the rise of C according to Zakharin et al. [27], is another circumstance favorable to the application of these crystals. So the investigation of the dependence $R(C)$ at the higher thallium content interval is of great interest. For a long time, such attempts were not made because of the existing concept about the concentration quenching at $C > 0.03\%$ [28,29]. Later, it became obvious that the character of the $L(C)$ dependence significantly depends on the photoreceiver-type [30] and the interest in crystals with a higher C was roused again.

Many scientists studied the influence of C on the light output of CsI:Tl by excitation with γ-quanta and α-particles, the majority of known dependences was summarized in Ref. [3]. In spite of the fact that known dependences were mainly obtained for crystals grown by Bridgman–Stockbarger technique, they differ essentially from each other, this concerns dependences obtained using "blue" PMT. At the same time, curves obtained using "red" photoreceiver are qualitatively similar [3,11,30]. This is the reason why we consider only these results. Data concerning the character of the dependence $R(C)$ for CsI:Tl have become available only recently [3,11].

Fig. 3 represents the dependence $L(C)$ for 662 keV γ-quanta obtained using PMT R669 ($\lambda_{max} \approx 600$ nm). According to curve 1, the plateau of this dependence for KC-crystals is in the interval $0.08\% < C < 0.3\%$. Curve 3 shows the dependence of α-yield on C and is normalized to 1 as in Fig. 2. The character of curve 3 and the value $C \approx 0.2\%$ corresponding to the initial point of the plateau $L_\alpha(C)$ coincide with the known data [11,28,30]. Curves 1 and 3 do not show the concentration quenching in the considered C interval.

The well-defined dependence $L(C)$ for energy 5.9 keV was obtained only for blue PMT R1307 and is not considered in the present paper. We could only remark that the position of curves for 5.9, 662 keV and 5.15 MeV corresponds to that of NaI:Tl shown in Fig. 2. $L_{5.9}$ reaches its maximum approximately between the initial points of the plateau for 662 keV and 5.15 MeV. A clear photopeak starting at $C = 0.1\%$ was obtained for "red" PMT, so we can estimate the initial point of the plateau as 0.13% for 5.9 keV X-rays (curve 2).

No concentration quenching of light output is also revealed in CsI:Tl of OC type (curves 5 and 6, Fig. 3). The same situation holds for these crystals as for NaI:Tl crystals of OC-type: maximum values L are a bit lower and the initial point of the plateau is shifted as compared to KC and VC crystals. Curve 6 in Fig. 3 was considered in Ref. [11] and was obtained using silicon PIN photodiode. A big difference in the characters of curves 5 and 6 is connected not with the type of "red" photoreceiver [3,30] but with a high or low content of cesium oxide compound in the crystals. The threshold indicator when oxides exert a negative influence over Tl^+ luminescence is $CsIO_3$ which is easy to reveal in crystals using IR-spectroscopy [9]. Non-radiative energy losses in the track of α-particle are so high that the difference in the light

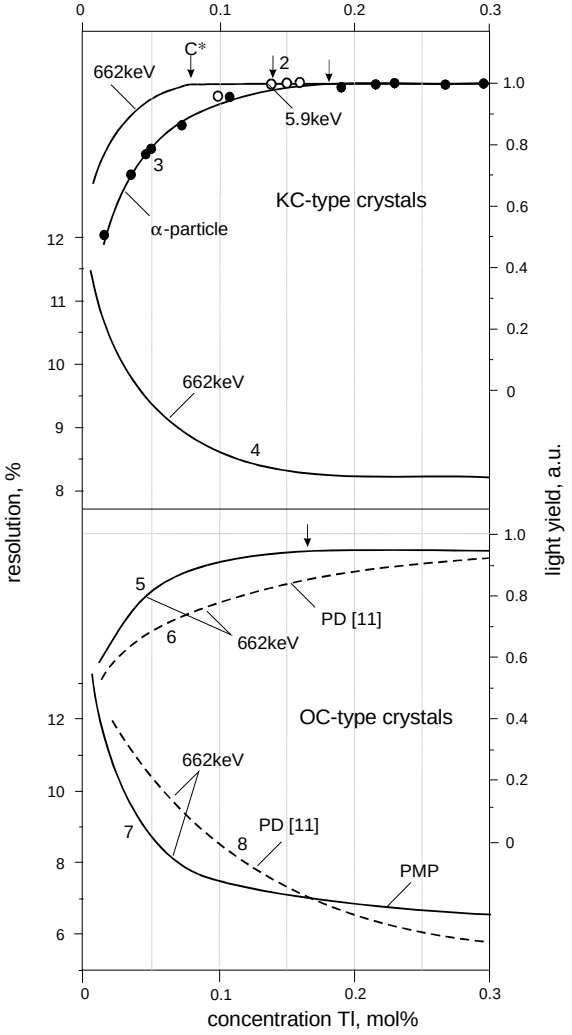

Fig. 3. The dependencies $L(C)$ (curves 1–3,5,6) and $R(C)$ (4,7,8) for CsI:Tl of KC- (top) and OC-type (bottom). PMTs Hamamatsu R669 are used for curves 1–5 and 7, silicon photodiodes [11] for 6 and 8. Excitation by 662 keV γ (1,4–8), 5.9 keV X-rays (2) and 5.15 MeV α (3). The initial point C^* of the L-plateau is, respectively, 0.08 (1), $\sim$0.14 (2) and 0.18 (3) mol% of Tl.

output between our OC and KC crystals could not be revealed.

The non-proportionality of L/E between 5.9 and 60 keV for CsI:Tl as for NaI:Tl could be reduced by increasing C. Fig. 4 represents NP curves for two crystals with different C. It is obvious that NP is almost not observed for $C = 0.2\%$ (curve 4) contrary to $C = 0.05\%$ (curve 3).

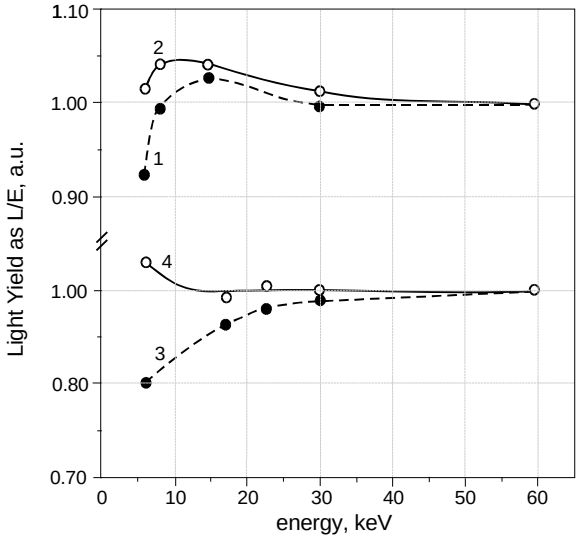

Fig. 4. Light yield per unit excitation energy of X-rays for NaI:Tl (1,2) [12] and CsI:Tl (3,4). Concentrations of Tl are 0.03 (1), 0.1 · (2); 0.05 · (3) and 0.2 (4) mol%.

Logically, an improvement of R must be expected with the rise of C. In fact, as can be seen from curves 4 and 7 in Fig. 3, R decreases in the whole C region as well as in the interval of L plateau. The same result for $R(C)$ dependence in CsI:Tl was reported in Ref. [11] when recording the signal with a silicon photodiode (curve 8, Fig. 3). But $L(C)$ curve in the work of reference did not have a well-defined plateau (curve 6). Our results are of great value because the improvement of the resolution is observed in the concentration interval where the light yield is constant and has its maximum values.

6. Discussion

The resolution and NP of response both for NaI:Tl and CsI:Tl could be corrected by increasing C. But this is not a principal way out since the value R in the plateau L decreases by $\sim$1%. Our explanation of this effect based on $L(C)$-dependence-type for different energies and ionization densities is of great interest. In our opinion, the correction of NP of response for the energy 5.9 keV can be explained by $L(C)$ dependence character only, when drawing attention to the fact

that $L(C)$ dependence for the low-energy electrons will be similar to that for α-particles. To achieve maximum values of L_α, it is necessary to increase C [6,15], this means that average values of dE/dx in the track formed after absorption of a quantum with $E = 5.9$ keV are also increased.

The ionization density in electron track is not distributed uniformly and is concentrated in the region of high ionization. A track fragment of a 1 MeV electron is represented in Fig. 1 according to the calculation of Paretzke [21]. When the energy of the electron is <0.5 keV, its track will shorten strongly [14,31]. A 1 MeV electron creates $\sim 10^3$ δ-electrons in NaI and 80% of them have an energy from 100 to 500 eV [32]. The total energy of the δ-electrons is ~ 320 keV.

Let us apply the MI model to the electron track structure, divide the track into cells is shown in Fig. 1 and substitute $m = 700$, that corresponds to the number of the regions with high dE/dx for track of the 662 keV electron, in Eq. (2). The best value of R_i is 0.05 for NaI:Tl [33]. Then $p \approx 0.7$ can be calculated. In other words, to obtain a "bad" resolution ($\sim 5\%$) the light yield should be 70% of L_{max} by fixed m set by the track structure. It should be noticed that this value coincides with the best values of α/γ-ratio for NaI:Tl and CsI:Tl [25]. In this connection, the resolution will be lower than 5% for the scintillator with $\alpha/\gamma \approx 1$. In spite of the fact that this estimation arbitrarily interprets the meaning of p, it is relevant for $m \approx 10^3$ and indicates the probable origin of fluctuations of a total number of scintillation photons (N_{ph}). Let us explain R_i value in a rigorous way.

The primary 1 MeV electron spends 32% of its total energy for the creation of δ-electrons, the rest for the creation of low-energy e−h pairs. The majority of δ-electrons create higher ionization regions in their formation point. Let us separate from the total number of photons N_{ph} the contributions N_1 (quantity of photons formed by the primary electron through separate e−h pairs), and N_2 (number of photons connected with δ-electrons). Table 1 shows the sequence of the calculation. When calculating N_1 we considered the following: (1) average energy of e−h pair formation E_{ave} (material constant does not depend on the energy, for NaI $E_{ave} = 20$ eV [7,34]); (2) migration losses using a coefficient $\eta_m = 0.8$; (3) light collection coefficient $\tau = 0.7$, a typical value for a NaI:Tl detector [33]. For N_2, we introduced the coefficient $\eta_\delta = 0.85$ like the α/γ ratio. $\eta_\delta = 0.85$ means that e−h pairs convert into photons with an efficiency from 1 to 0.7 (as bottom limit) in the regions with high dE/dx.

As can be seen from Table 1, a quite acceptable value ($\sim 4\%$) was obtained for R_i. The main assumption consists of the fact that N_1 and N_2 are statistically independent since they are formed by different processes. The advantage of our approach, where NP of response will not be taken into account, is that it is based on the investigation of the dependences $L(C)$ and $R(C)$. This approach allows to explain the decrease of R_i with the growth of C, namely by the $\eta_\delta(C)$ coefficient dependence which must be similar to the α/γ-ratio dependence [25]. For lower C, we calculated $R_i = 4.81\%$ when $\eta_\delta = 0.5$, and $R_i = 4.47\%$ for $\eta_\delta = 0.7$.

We do not take NP of response into account because it is not considered to be a fundamental property of the crystal. We suppose that it is a real observed physical phenomenon connected with the light collection peculiarities near the crystal surface [14]. To explain NP of response in the energy interval 5.9–662 keV, careful calculations of τ are necessary; the use of a point light source is not obligatory.

Table 1
Statistical calculation of the intrinsic resolution of NaI:Tl detector for 662 keV gammas

	Energy quota (eV)	N_{e-h}	N_{ph} with account of			Account of $\tau = 0.7$	$2.36/\sqrt{N}$ (%)
			$\eta_m = 0.8$	$\eta_\delta = 0.85$	Experiment		
N_1	450 000	22 500	18 000		38 000 MeV^{-1}	12 600	2.1
N_2	212 000	10 600	8480	7208		5045	3.32
Σ	662 000	32 000	26 400	25 208	25 156	17 645	$(\Sigma R_i^2)^{1/2} = 3.93$

7. Conclusions

Based on the analysis of $L(C)$ dependences for different energies and ionization densities, the frequently observed concentration quenching of the light output was explained by the formation of MI in the lattice of NaI:Tl crystal due to the spinodal decay of solid solution. We showed that the concentration quenching region can be significantly shifted to the region of high thallium content; the effect of resolution enhancement with the rise of activator concentration is well-defined in such crystals. This effect is accompanied by an increase of the light yield for 5.9 keV X-rays. The specific yield L/E for quite high activator concentrations does not practically depend on the energy in the range of 5.9–60 keV, both for NaI:Tl and CsI:Tl crystals. This circumstance can explain the concentration dependence of the resolution in the region of light output constancy.

Starting from the concept of complicated electron track structure, we proposed an alternative explanation of the value of the intrinsic detector resolution based on the statistic fluctuation of a number of photons formed outside and inside the regions of higher ionization density. The number of the latter depends on the activator concentration even in the plateau interval of $L(C)$ standard dependence for a 662 keV energy.

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