Peculiarity of Energy Transfer in Irradiated CsI(Tl) Crystal

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Abstract – The role of radiation defects in the light yield degradation of detectors CsI(Tl) has been investigated. The spectral kinetic study of the luminescence of CsI(Tl) crystals colored by γ-radiation (^{60}Co, 10^6 Gy) allowed to determine that two types of the induced by irradiation centers absorb the luminescence of Tl^+ centers responsible for the scintillations. The re-absorption of the Tl^+ emission leads to appearance of the long-wave luminescence caused by the color centers of the first type. The luminescence of the other type centers is quenched at room temperature. These centers not only deteriorate transparency to the activator luminescence, but also function as quenching centers to which the non-radiative energy transfer from Tl^+centers is realized.

1. Introduction

It is known the scintillation characteristics deterioration of the CsI(Tl) detector subjected to the high-dosed irradiation is caused by two factors: the transparency loss of CsI(Tl) crystal to the activator luminescence and scintillation efficiency reduction [1–3]. Since the spectrum of the induced absorption strongly overlaps the luminescence spectrum of the Tl^+ centers responsible for the scintillations, transparency deterioration of the irradiated crystal contributes substantially to the light yield degradation. As the radiation dose does not exceed $1\cdot10^5$ Gy, the light yield deterioration is determined only by the loss of the transparency as a result of the color center absorption, but the scintillation efficiency keeps safe. It means the energy transfer to the luminescence centers in the non-irradiated and irradiated with the said above dose CsI(Tl) crystal is realized over the same mechanizm. However at the higher radiation dose ($5\cdot10^5$ Gy) the mechanism of the energy transfer is subjected to change.

At present time there are different conceptions in what way the radiation defects take part in forming the light pulses. According to [3] the radiation defects block the V_k centers diffusion to both Tl^+ and Tl^0 centers. In the theoretical model of the effective energy transfer to the radiation defects [2], the scintillation efficiency deterioration is explained by the non-radiative decay of excitons on the vacancy clusters formed as a result of irradiation. It is authors' opinion of this model that the number of the vacancy clusters termed by them as vacancy pores is by 4–5 orders lower than the number of Tl^+ centers. Results of [4] show that the best scintillation parameters are achieved as activator concentration is varied in the region of $1\cdot10^{18} – 3\cdot10^{19}$ cm^{-3}.

Formation of the same color centers is caused not only irradiation with ionizing radiation but also illumination of the crystal with the laser emission [6]. The quantity of this centers generated with the laser emission can run up to $1\cdot10^{16}$ cm^{-3} as activator concentration equals $1\cdot10^{18}$ cm^{-3}.

Using absorption methods for investigation of the radiation damage of CsI(Tl) crystals with different impurities the authors of the present work found out that maximum concentration of the color centers at the same irradiation dose is observed for CsI(Tl) crystal containing additionally carbonate ions. According to the formula of Smakula it was estimated the maximum number of color centers formed in CsI(Tl) crystal under irradiation with the dose of $1\cdot10^6$ Gy. It turned out, that their concentration does not exceed $5\cdot10^{16}$ cm^{-3}. Making a comparison one can see that the concentration of Tl^+ centers in the crystal with optimal activator concentration at least is by 1.5–3 orders higher than concentration of the color centers.

Therefore hypothesis that the radiation defects compete in capture of electron excitations a priori makes us assume that decay of excitations can occur on the radiation defects in that case, if their capture section significantly exceeds those of Tl^+ ions.

On the other hand, considerable overlapping of spectra of Tl^+ luminescence and bands of induced absorption evidence that radiation defects can participate in forming scintillations otherwise. It may be not competition of radiation defects with Tl^+ ions in captures of electron excitations, but radiative or non-radiative energy transfer from excited Tl^+ centers to the color centers caused with irradiation.

The aim of the present work is to study the process of the energy transfer from Tl^+ centers to the color centers in the strongly irradiated CsI(Tl) crystal.

2. Experimental procedure

The spectral measurements were performed with the CsI(Tl) crystal containing additionally CO_3^{2-}ions, because of these ions stimulate radiation defects formation. The content of activator and carbonate ions in the crystal amounts to $8\cdot10^{-2}$ mol % and $5\cdot10^{-4}$ mol %, respectively. The luminescence was excited with the light of a deuterium lamp through an MDR-2 mono-

chromator as well as by the radiation of a ^{241}Am source and analysed by the DMR-23 monochromator. The absorption spectra were recorded using the specially designed set-up based on SF-4 unit. Spectral-kinetic measuremens were carried out under electron pulses (with parameters: energy of 240 KeV, the pulse duration of 10ns and the excitation density of ~6 mJ/cm^2). Crystal was irradiated at 295K by γ-ray from a ^{60}Co source with a dose of $1\cdot10^6$ Gy. Measurements were carried out in the temperature interval of 80–295K.

3. Results and discussion

The efficiency of the radiation defect formation in CsI(Tl) crystals depends not only on the activator concentration, but also on the impurity presence in the lattice [1]. Irrespective of the impurity ions nature the identical bands are observed in the spectrum of induced absorption. Fig. 1 shows the typical absorption spectrum of the irradiated CsI(Tl) crystal. Many of the absorption bands are strongly ovelapped with the spectrum of Tl^+ center luminescence. According to [6] the defects formed in the CsI(Tl) under irradiation have the activator nature. The bands with the maxima of 390 nm and 465 nm are caused by the first (i) type color centers. The additional thermal annealing of the crystal at 373°C destroys the defects, responsible for these centers. The color centers of second (ii) type are steady to this annealing. The bands with the maxima of 430 nm, 520 nm, 560 nm in the absorption spectrum are connected with them. The temperature decrease from 295 K to 80 K leads to the shift of the absorption band maxima (see Fig. 2 and Table).

The spectral composition of the luminescence excited at 80 K in (i) and (ii) absorption bands depends on wavelength of the exciting light (Fig. 2). The spectrum of luminescence excited in the band of 394 nm caused by (i) centers consists of two well-divided bands with the maxima of 591 nm and 675 nm. Only one band with the maximum of 591 nm is observed in the luminescence spectrum under excitation in the (i) center band with the maximum of 480 nm.

The luminescence of 591 nm and 675 nm is excited in the absorption area of Tl^+ ions (220–300 nm) as well as in the area of the induced absorption of 320–540 nm. The most intensive 394 nm and 480 nm bands of the excitation spectra coincide with the absorption bands of the (i) centers.

Table. The maxima position of the bands in the absorption spectra measured at 295 K and 80 K (in brackets) for the irradiated CsI(Tl) crystal

Type of absorption center	Position of maximum, nm		
(i)	390 (394)	465 (480)	
(ii)	430 (438)	520 (532)	560 (565)

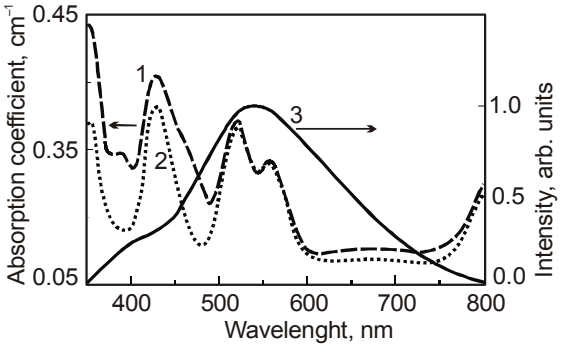

Fig. 1. Absorption spectra of the irradiated (^{60}Co, $1\cdot10^6$ Gy) CsI(Tl) crystal measured before (1) after (2) additionally annealing during 6 hours at T=373 K. The radioluminescence spectrum non-irradiated CsI(Tl) crystal (3)

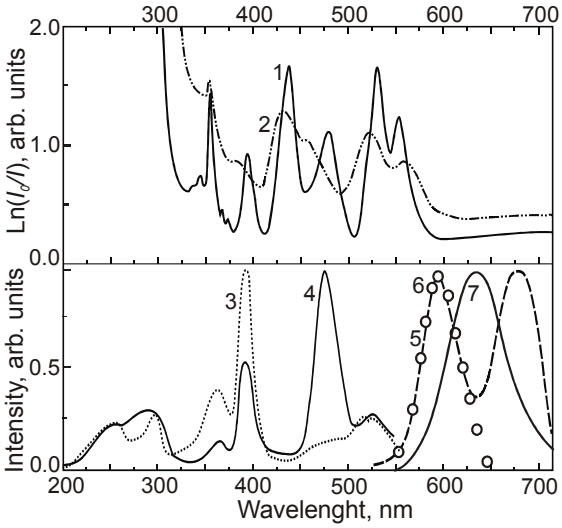

Fig. 2. Spectra of the absorption (1,2), excitation (3,4) for λ_{lum} = 675 nm (3), 591 nm (4) and luminescence for λ_{exc} = 394 nm (5), 488 nm (6), 530 nm (7). Measurements are carried out at 295 K (1) and 80 K (2–7) before (1–6) and after (7) additionally annealing at 373°K of the irradiated CsI(Tl) crystal

The study of the (ii) center luminescence was carried out for the irradiated sample subjected to the additional thermal treatment at 373 K. As result of the annealing the (i) type centers were destroyed, therefore the bands of 394 nm and 480 nm were absent in the absorption spectrum (Fig. 1, curve 2). The luminescence spectrum of the (ii) color centers excited in 530 nm region consists of one band with the maximum of 630 nm, which is well described by Gaussian. With the rise of temperature the intensity of the 630 nm luminescence decreases. The temperature dependence of this luminescence yield is straightened in the coordinates *{Ln((Io/I)-1) –1/T}*. It testifies to the intra-center nature of quenching. The activation energy of quenching for the (ii) center luminescence is equal to 0.053 eV. As it can be seen in the Fig. 3 the emission of these centers is quenched at the temperature above 225 K.

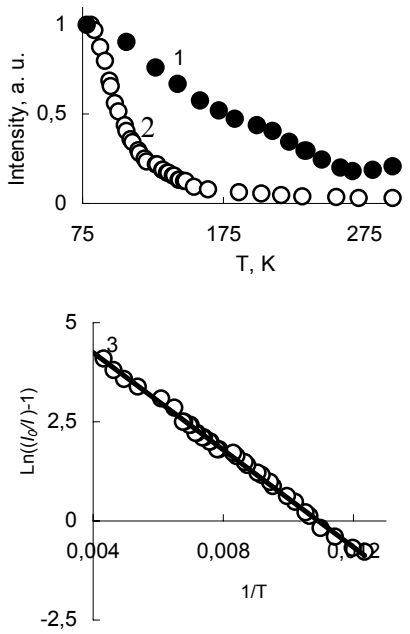

Fig. 3. The temperature dependences of the photoluminescence intensity λ_{lum}=590 nm (1), 630 nm (2,3) in the coordinates of {*I(T) – T*} (1,2) and {*Ln((Io/I)-1) –1/T*} (3). Measurements are carried out before (1) and after (2,3) aditionally thermal treatment at 373°K of the irradiated CsI(Tl) crystal λ_{exc} = 480 nm (1), 530 nm (2,3)

The temperature dependence of the intensity for the luminescence excited in the 480 nm band is shown in Fig. 3. Curve is described this dependence is more complicated. It is remarkable that the (i) type centers luminescence is not quenched at the room temperature.

The Tl^+ centers luminescence revealed in the radioluminescence spectrum of the irradiated CsI(Tl) crystal is distorted by the absorption of both type centers induced with irradiation (Fig. 4).

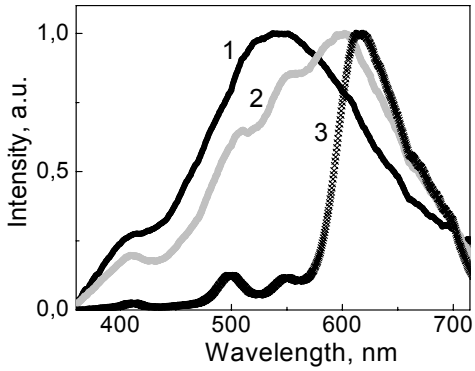

Fig. 4. Radio-luminescence spectra of CsI(Tl) crystal measured at T=295 K before (1) and after (2,3) irradiation. The thickness of samples cut out of the same irradiated crystal equals to 30 mm (2) and 0,3 mm (3)

The radioluminescence spectra measured for the irradiated by the dose of $1 \cdot 10^6$ Gy samples with thickness 0,3 mm and 30 mm have maxima at 600 nm and 620 nm, respectively, whereas one can see maximum at 540 nm in the spectrum of non-irradiated crystal. Thus, the radioluminescence spectrum is shifted to the long-wave area then farther then more the thickness of the irradiated crystal as well as then more the radiation dose [7]. The long-wave shift is accompanied by the intensity decreasing in the short-wave area. At 430 nm, 520 nm, 560 nm is observed cavities, which by their position coincide with the maxima of the most intensive (ii) center absorption bands.

The Tl^+ centers luminescence re-absorption with the color centers of both types leads to their excitation. Since the (i) color centers luminescence at room temperature is not quenched, it can be revealed beside the Tl^+ center emission in the total radio-luminescence spectrum of the irradiated crystal. The radiative energy transfer from Tl^+ centers to the (i) type color centers should increase duration of the scintillations. It is proved by the results of the authors [3]. They observed increasing the scintillation duration after irradiation of a CsI (Tl) crystal with the dose of $5 \cdot 10^5$ Gy.

But as the (ii) type color centers luminescence are quenched at room temperature, the radiation energy transfer to these centers should not influence on duration of the scintillations.

In order to decrease the re-absorption effect, the study of the irradiation influence on the decay kinetics of the scintillation pulses was carried out on the small-size sample. The decay curves of the luminescence excited by electron impulses is well described by the exponents with a decay constant of 833 ns and 770 ns obtained, respectively, for the irradiated and non-irradiated crystal sample (Fig. 5).

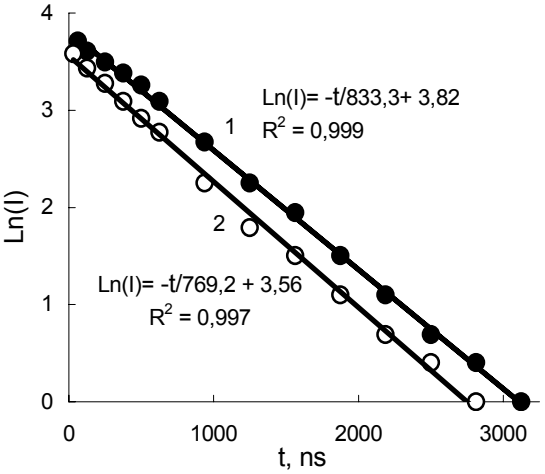

Fig. 4. The pulse decay of the cathodoluminescence excited in the crystal CsI(Tl) before (1) and after (2) its irradiation with the dose of $1 \cdot 10^6$ Gy

Shortening the luminescence duration of a CsI(Tl) crystal containing both Tl^+ centers and the radiation deffects can be explained by the non-radiative energy transfer from the Tl^+ centers to the (ii) type centers. Since at room temperature (ii) centers are represented quenching centers, the resonant energy transfer to these centers inevitably leads to the scintillation efficiency reduction of the irradiated CsI(Tl) crystal.

The analogical effect of the scintillation efficiency decrease as a result of the resonance energy transfer from the excited molecules of the ground substance to radiation defects, wich are quenching centers have been revealed in the organic scintillators [7].

The centers of (ii) type are described to a color center $Tl^0V_a^+$, where the electron is captured with Tl^+ ion, disturbed by an anion vacancy V_a^+ [6]. The distance R between the Tl^+ ion and the nearest $Tl^0V_a^+$ color center corresponds to one between the close Tl^+ ions. As shown by our estimation it equals to about 11 lattice parameters in the investigated sample. Since in this case one can observe shortening scintillation pulses, the obtained value R is smaller then the distance of transfer R_0. The rise of the irradiation dose leads to increasing the radiation defect concentration. Hence the probability of getting these defects to the track of ionizing particle becomes higher. That is why the scintillation efficiency reduction is occurred only under high-dose irradiation. The value of this characteristic dose can be altered in dependence on the Tl^+ concentration and presence of some impurities in the lattice [1].

4. Conclusion

The scintillation parameters deterioration of the irradiated CsI(Tl) crystals is connected with the energy transfer processes from Tl^+ centers, responsible for the scintillation, to the radiation deffects of two types, one of which is the quenching center.

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