

# Color centers in heavily irradiated CsI(Tl) crystals

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## Abstract

The absorption and luminescence properties of CsI(Tl) crystals colored by irradiation are studied by the method of the time-resolved spectroscopy. The scheme of the electron transitions in CsI(Tl) crystal is suggested to explain the appearance of the color centers under exposure to the near-UV light. It is established that either of the two types activator color centers holds the charge carrier with opposite sign. The model of the hole  $Tl^{2+}v_c^-$  activator color center is suggested. According to the model the positive charge of  $Tl^{2+}$  ion is compensated by the negative charge of a close cation vacancy  $v_c^-$ . The color center emission reveals in the cathode-luminescence spectrum of the colored CsI(Tl) crystal. The high-dose irradiation of CsI(Tl) crystal results in the reduction of the decay time of the near-thallium self-trapped excitons (STE) emission. The decay kinetics of  $Tl^{2+}v_c^-$  emission contains the time components typical for the decay kinetics of near-thallium STE emission. The reason of the observed effects is the energy transfer from the near-thallium STE excitons to the color centers via the inductive-resonant mechanism.

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## 1. Introduction

The ability of CsI(Tl) crystals for coloration under high-energy radiation or exposure to the light of the visible and near-ultraviolet range was noted in earlier papers devoted to the study of luminescence of these crystals [1,2]. The authors of the mentioned works established the fundamental properties of the color center luminescence excited in the additional absorption bands. The luminescence spectrum of color centers with a common maximum at 1.94 eV was found to be non-elementary and related to at least two types of centers oriented along the fourth-order symmetry axes [2].

The use of CsI(Tl) crystals as scintillation counters has shown that radiation-induced color centers cause changes of the detector operating characters, a shift of the scintillation spectrum to the red region [3] and the light yield decrease [3–5]. As it has turned out, the degradation of the light yield is related not only to the deterioration of the crystal transparency due to the absorption induced by

irradiation [6,7], but also to the decrease of the energy yield of the activator emission [8–10]. According to the existing conceptions, this effect is explained by the competition of color centers with activator  $Tl^+$  centers in the capture of the electron excitations. Taking into consideration the fact that the quantity of the radiation-induced color centers does not exceed a thousandth of the activator center number [11], the mechanisms of the competition between the mentioned centers should be regarded as extremely effective. The origin of the color centers is not completely understood and undoubtedly should be studied further. The irradiation effect on the absorption characteristic and the luminescence spectral-kinetic parameters of CsI(Tl) crystals with different content of thallium and the radiation-induced structure defects has been studied in the paper.

## 2. Experiment

The samples of CsI(Tl) crystals with the thallium content of  $2.6 \times 10^{-3}$ – $2.6 \times 10^{-1}$  mol% were grown at the Institute for Scintillation Materials of NAS of Ukraine by seeding in

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the inert atmosphere. To increase the efficiency of coloration [7], some of CsI(Tl) crystals were doped by the carbonate ions  $\text{CO}_3^{2-}$ . Their concentration in the crystal was  $5 \times 10^{-4}$  mol%. The samples were colored by  $\gamma$ -rays from a  $^{60}\text{Co}$  source as well as by the filtered light of a mercury arc (UFS-2 filter, 365.4 nm emission line). The absorption spectra were measured by a spectrophotometer SF-26. The luminescence of the samples cut out in the form of a rectangular plate with the dimensions of  $1 \times 8 \times 8 \text{ mm}^3$  was excited with pulses of accelerated electrons ( $E_e = 0.25 \text{ MeV}$ ,  $j = 8 \text{ A/sm}^2$ ,  $t_{1/2} = 15 \text{ ns}$ ). The spectral-kinetic study of the cathode luminescence was carried out by a pulse optical spectrometer [12] with monochromator MDR-3, photomultiplier FEU-106 and oscilloscope S8-12 in the spectral range of 1.1–5 eV with a time resolution of 10 ns [12].

### 3. Results and discussion

#### 3.1. Optical absorption

The absorption spectra of CsI(Tl) sample measured at 295 K before and after its irradiation and thermal treatment are shown in Fig. 1. The  $\text{TI}^+$  center absorption can be seen in the spectrum of the newly grown crystal (Fig. 1, curve 1) as the low-energy edge of A-band caused by the electronic transitions between the states of  $\text{TI}^+$  ion disturbed by its environment [13].  $\gamma$ -irradiation

( $^{60}\text{Co}$ ,  $1 \times 10^6 \text{ Gy}$ ) of the crystal results in the appearance of the intense coloration in UV, visible and near-IR spectral region (Fig. 1, curve 2). According to Refs. [11,14], the observed coloration is caused by the formation of two types of color centers labeled as (i) and (ii). The maxima of their absorption bands are marked in Fig. 1 by the arrows. The (ii) type centers are not photo-stable and destroyed under illumination in 2.88 eV band [14]. On the contrary, the centers of (i) type are optically stable, but they disappear at heating of the crystal up to 420 K [11,15].

All additional absorption bands disappear on heating up to 800 K. However, comparing the spectra in Fig. 1 (curves 1 and 3), one can see that the spectrum does not restore completely even after keeping the crystal at this temperature for an hour. A significant increase of the absorption intensity on the long-wavelength decline of the  $\text{TI}^+$  center band is observed in the spectrum of the thermally discolored crystal. Like for undoped alkali halides crystals [16], we can explain the “long-wave shift” of the  $\text{TI}^+$  center absorption edge in the spectra of irradiated crystals by the influence of intrinsic defects such as vacancies of cationic and anionic sublattices  $v_c^-$  and  $v_a^+$ , respectively, on the levels of the valence electrons and on the p state levels of  $\text{TI}^+$  ions. The activator  $p(\text{TI}^+)$  level is situated in the band-gap. The lowering of  $p(\text{TI}^+)$  level in the complex  $\text{TI}^+v_a^+$  and the rise of the energy levels of the valency electrons in the complex  $\text{TI}^+v_c^-$  should be expected at the radiation creation of a vacancy in the nearest environment of  $\text{TI}^+$  ion.  $\text{TI}^+v_a^+$  and  $\text{TI}^+v_c^-$  activator complexes induced by irradiation cause the appearance of the allowed transitions with the photon absorption from the longer wavelengths region in comparison with a non-irradiated CsI(Tl) crystal. The optical transitions resulting in charge transfer are shown by arrows in the diagram of the energy levels (Fig. 2). In our opinion, the formation of color centers in CsI(Tl) crystal under exposure to the daylight is due to such transitions.

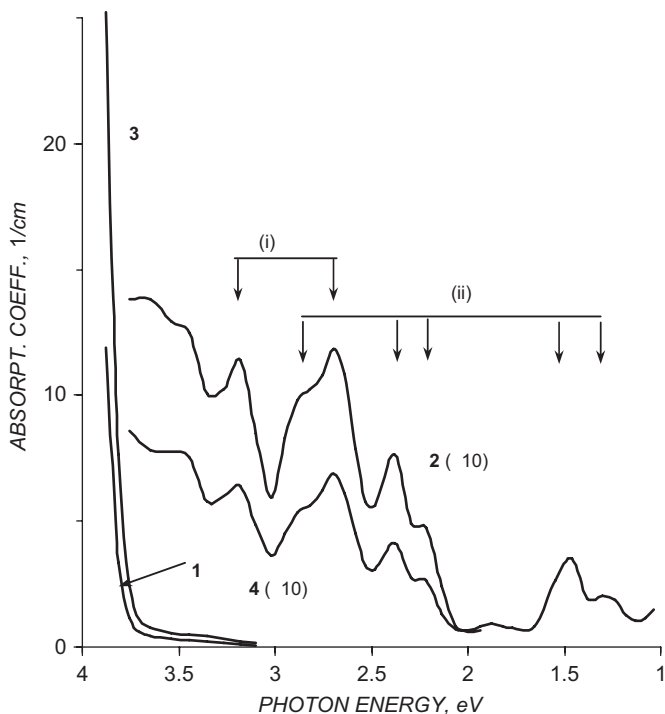


Fig. 1. Absorption spectra of CsI:0.08 mol% Tl crystal measured before (1) and after (2–4) a series of successive operations: 2— $\gamma$ -irradiation ( $^{60}\text{Co}$ ,  $1 \times 10^6 \text{ Gy}$ ); 3—thermal decoloration during an hour at 800 K; 4—coloration by 365 nm ultraviolet light during an hour. The spectra were measured at 295 K.

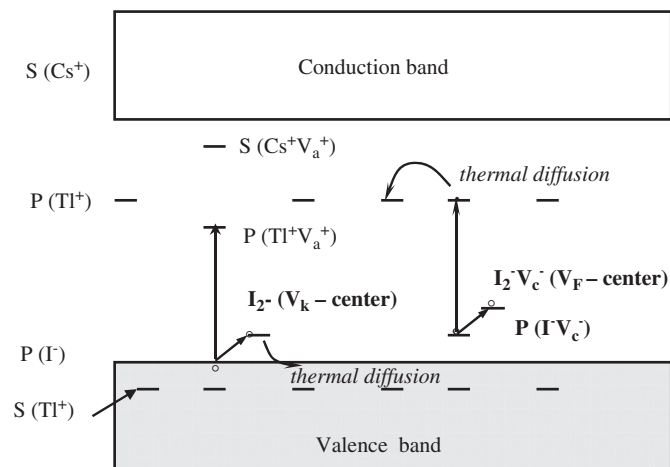


Fig. 2. Band diagram of CsI(Tl) crystal containing single anion vacancies ( $v_a^+$ ) and cation vacancies ( $v_c^-$ ). The arrows denote the allowed electron transitions with the absorption of photons from the near-ultraviolet region of the spectrum.

The studied CsI(Tl) sample after preliminary  $\gamma$ -irradiation was discolored by a high-temperature annealing. After that the sample was exposed to UV-light ( $\lambda = 365.4$  nm) for 1 h. Its absorption spectrum is shown by curve 4 in Fig. 1. It comes from the comparison of the induced absorption spectra (curves 2 and 4 in Fig. 1) that in terms of the spectral composition the light-induced coloration is the same as the one caused by  $\gamma$ -irradiation. The intensities of the absorption bands in both spectra are also comparable.

The inertial growth of the coloration intensity after cessation of irradiation of CsI(Tl) crystal is an unusual property for the phenomenon of the radiation coloration [1]. A short-term (about 1 min) exposure to UV-light of the CsI(Tl) sample preliminary colored by daylight at room temperature causes the change of its color. We monitored on the optical density change for 1 h (Fig. 3). The dependence of the optical density on time was measured for bands with 2.69 and 2.88 eV maxima related to (i) and (ii) centers, respectively. Kinetic curves of optical density have two stages. The first stage of the fast rise is the same for the centers of both types. The second slow stage lasts for hours. The reduction of the number of (ii) centers on the second stage is accompanied by the synchronous growth of the number of (i) centers. As mentioned above, the main effect of UV-light should be considered as the

recharge of the defects existing in the crystal. Therefore, the inverse dependences (curves 1 and 2 in Fig. 3) can be considered as an important argument in favor of the conclusion that (i) and (ii) centers hold the charge carriers of the opposite sign.

The possibility of the formation of  $Tl^0$ -centers stable at 295 K in  $\gamma$ -irradiated CsI(Tl) crystal is grounded as in Ref. [14]. The increase of the thermal stability of  $Tl^0$ -centers is caused by anion vacancies generated under irradiation. The model of (ii) center is regarded to be a complex  $Tl^0V_a^+$ . The following arguments can be considered in favor of the conclusion:

- (A) It is shown in work [17] that the ionizing radiation does not cause the increase of the Tl diffusion constant in KBr, KCl and CsI crystals. It means that the aggregation of activator ions under irradiation does not occur. Neither dimers  $Tl_2^{2+}$  [18,19] nor trimers  $Tl_3^{3+}$ , as it was supposed in Ref. [20], but single thallium ion is the structure unit of the color centers. The evidence of such conclusion is the linear dependence of the optical density at 2.55 eV on Tl concentration in CsI(Tl) samples irradiated with the same dose of  $5.5 \times 10^5$  Gy. As Tl concentration increases from  $2.6 \times 10^{-3}$  to  $2.6 \times 10^{-1}$  mol%, the number of color centers increases by linear, rather than quadratic or cubic law (Fig. 4).
- (B) Like the absorption spectrum of  $Tl^0$  center perturbed by anion vacancy in KCl(Tl) crystal [21] or by  $V_k$  center in CsI(Tl) crystal [22], the absorption spectrum of (ii) type center contains the infrared bands with maxima at 1.48 and 1.28 eV (Fig. 1). Authors [21,22] ascribe these infrared bands to transitions  $6^2P_{1/2} \rightarrow 6^2P_{3/2}$ , which are forbidden in free  $Tl^0$  atom, but partially allowed in a crystal over lowering of the  $Tl^0$

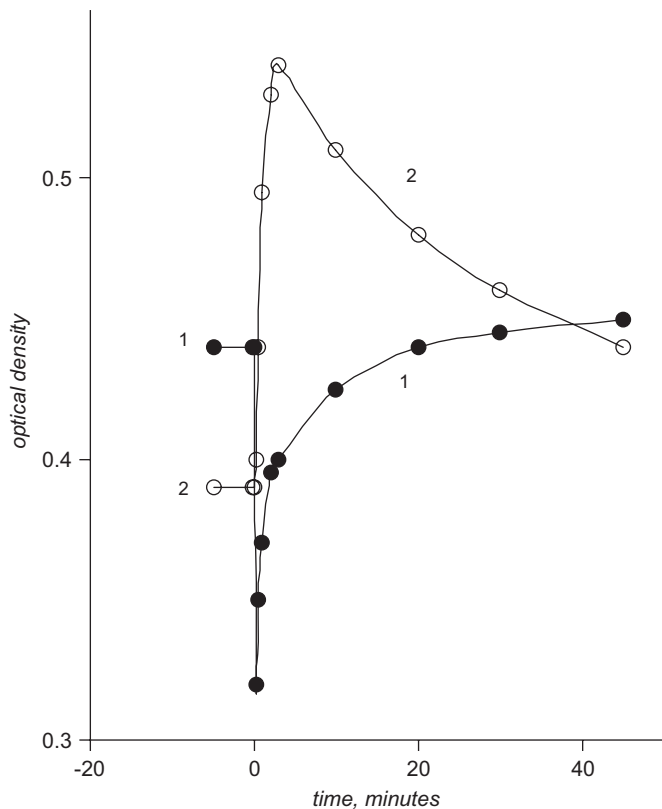


Fig. 3. Kinetic curves of the optical density change at 2.69 eV ((i)-type centers) (1) and at 2.88 eV ((ii)-type centers) (2) after exposure to filtered (365.4 nm) mercury lamp emission ( $I = 2 \times 10^{16}$  photon  $cm^{-2} s^{-1}$ ;  $t = 1$  min).

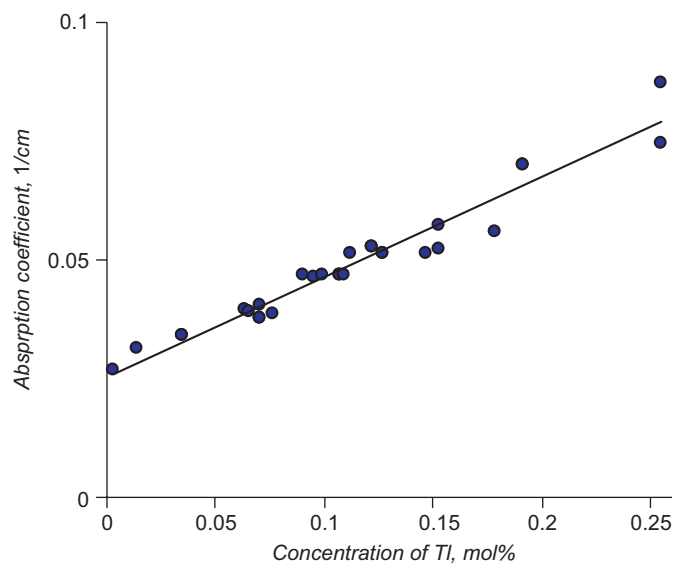


Fig. 4. The concentration dependence of the absorption coefficient at 550 nm for CsI(Tl) crystals irradiated with a dose of  $5.5 \times 10^5$  Gy ( $^{60}Co$ ).

center symmetry in a field of the effectively charged close defect.

As noticed above, either of the two types of color centers hold the charge carrier of the opposite sign. Therefore the structure of (i) center is logically regarded to be a  $\text{TI}^{2+}\text{v}_c^-$  complex antimorphic to (ii) center. The excessive positive charge of  $\text{TI}^{2+}$  ion is compensated by a negatively charged cation vacancy in  $\text{TI}^{2+}\text{v}_c^-$  center. The optical properties of  $\text{TI}^{2+}$  hole centers in alkali halide crystals with NaCl-type structure are thoroughly investigated (see Ref. [24] and references therein). Among the features of these centers, the authors denote that the bond between  $\text{TI}^+$  ion and its halide environment is predominantly covalent. It means that maxima of absorption bands in the spectra of these crystals slightly depend on the kind of cation. We believe, that the closeness of maxima of (i) center bands in the CsI(Tl) spectrum and  $\text{TI}^{2+}$  center bands in the spectra of KI(Tl) and RbI(Tl) (Table 1) are not spontaneous but caused by the structure of electronic levels in  $\text{TI}^{2+}\text{I}^-$  molecular ion.

Based on the interpretation of (i) and (ii) centers as complexes ( $\text{TI}^{2+}\text{v}_c^-$ ) and ( $\text{TI}^0\text{v}_a^+$ ) we can explain the dependences in Fig. 3 as follows. As it is shown in the scheme (Fig. 2), the effect of UV light can cause the formation of electronic excitations mobile at room temperature: the holes of the valency band and the holes localized on cation vacancies as well as electrons of the thallium P sub-band  $\text{TI}^+\text{v}_c^-\text{e}^-$ . According to Ref. [23], holes in CsI(Tl) crystal are localized on cation vacancies in the form of molecular anions with the structure of  $(\text{I}_2^-)_{\text{aa}}\text{v}_c^-$  (or  $\text{V}_F$  center) and  $(\text{I}_3^-)_{\text{aca}}$  (or  $\text{V}_2$ -center) responsible for the peaks of the thermo-stimulated luminescence at 210 and 340 K, respectively. In our opinion, the presence of two types of the hole color centers with different thermal stability results in the appearance of fast and slow stages in the change of the number of (i) and (ii) centers after UV-irradiation. The less stable  $\text{V}_F$ -centers define course of the fast stage whereas the more stable  $\text{V}_2$ -centers define course of the slow stage.

Fast stage (in seconds) can be described by the reactions with participation of electrons and the two-halide hole centers:

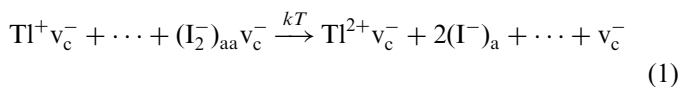
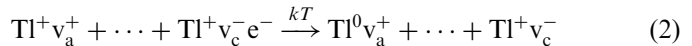
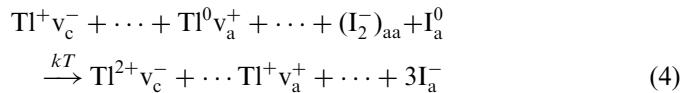
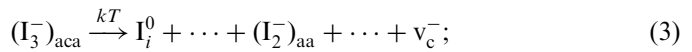


Table 1  
The maxima of the  $\text{TI}^{2+}$ -center absorption bands

Crystal	Type of center	Maxima of absorption band	References
KI(Tl)	$\text{TI}^{2+}$	eV nm 3.43; 2.80 361; 443	(see Ref. [24] and references therein)
RbI(Tl)	$\text{TI}^{2+}$	eV nm 3.25; 2.70 380; 460	
CsI(Tl)	(i)	eV nm 3.18; 2.70 390; 460	



Slow stage (in minutes) can be described by the thermal dissociation of  $\text{V}_2$  centers:



### 3.2. Luminescence

The luminescence spectra of the non-irradiated and  $\gamma$ -colored CsI(Tl) samples excited with the pulsed electron beam at 80 K are shown in Fig. 5. The spectrum of the non-irradiated crystal (Fig. 5a) in the visible region is well fitted by two Gaussians with maxima at 2.55 and 2.25 eV and FWHM = 0.35 eV. According to Ref. [13], these two bands are related to the radiative annihilation of near-activator two-halide excitons of two morphologies, which differ in the mutual position of both  $\text{TI}^+$  ion and the two-halide exciton nucleus. In accordance with Ref. [13], the “weak

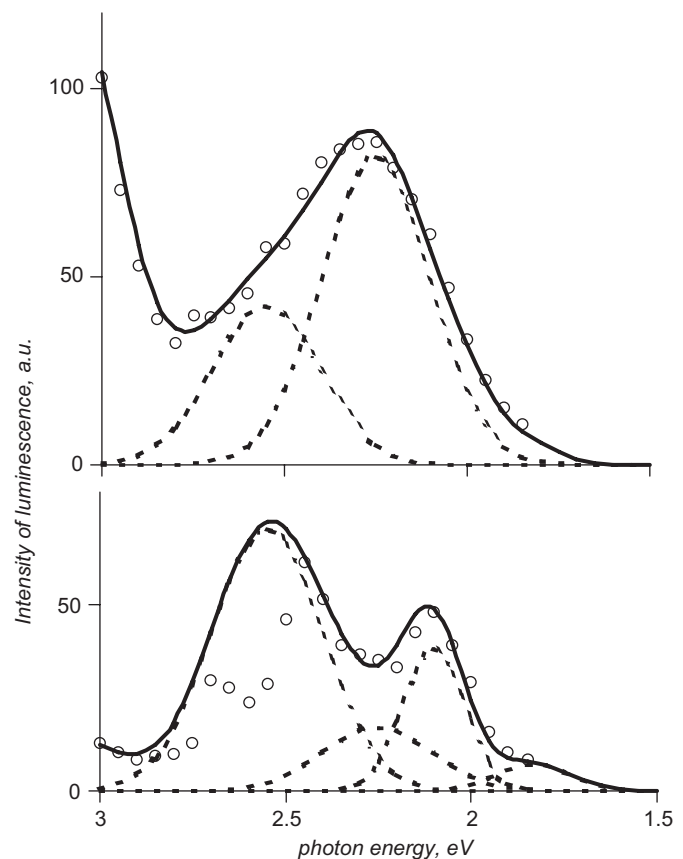


Fig. 5. Spectra of the cathode luminescence excited with a pulse electron beam at 80 K in CsI(Tl) samples: (a) non-irradiated and (b) colored by  $\gamma$ -radiation ( $^{60}\text{Co}$ ,  $1 \times 10^6$  Gy). Dots—experiment, dash line—fitted by Gaussians of the bands related to the emission of STE and color centers, solid lines—the best fit of experiment by the Gaussians.

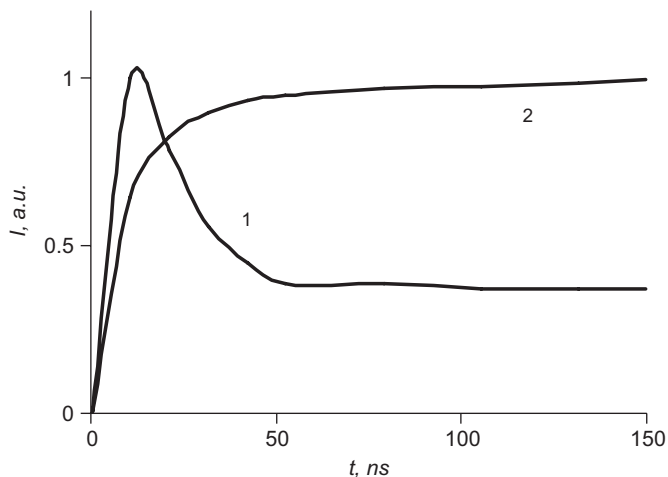


Fig. 6. Kinetic curves of the cathode luminescence intensity change at 2.25 eV (1) and 2.55 eV (2) for CsI(Tl) crystal excited by a pulse electron beam at 80 K.

off-center” and “strong off-center” excitons are responsible correspondingly for 2.55 eV band and 2.25 eV band having a bigger Stokes shift. The pulses of cathode luminescence registered at 2.25 and 2.55 eV are shown in the nanosecond time scale by curves 1 and 2 in Fig. 6. The rise of the 2.55 eV emission intensity while partial decrease of the 2.25 eV emission is quite uncommon. The inverse dependencies of the 2.25 and 2.55 eV emission intensity can be explained by the transformation of some amount of “strong off-center” excitons into “weak off-center” ones. Such transformation can be explained, for example, by reorientation of two-halide nuclei of “strong off-center” exciton by a 90° angle turn. The coloration of CsI(Tl) crystal by  $\gamma$ -irradiation causes a drastic change of the cathode luminescence spectra. The comparison of the spectra (a) and (b) in Fig. 5 allows to reveal the changes caused by color centers:

- The cathode luminescence intensity in the short-wave region of the spectrum decreases almost by one order.
- The intensity ratio of 2.25 and 2.55 eV bands changes because of almost the total disappearance of the nanosecond component in the decay kinetics of 2.25 eV emission.
- The new emission band with  $E_m = 2.1$  eV and  $\text{FWHM} = 0.2 \pm 0.01$  eV appears in the cathode luminescence spectra.

A band with the same parameters has already been observed in the spectra of the photoluminescence of the colored crystals [14]. This band is ascribed to (i) type color centers. Besides the mentioned changes, the emission spectrum contour is distorted by a narrow dip at 2.59 eV. This energy corresponds to the maximum of excitation spectrum for 2.1 eV emission related to (i) centers [11,14]. The appearance of the dip in the cathode luminescence spectrum of the colored crystal could reasonably be

explained by re-absorption of  $\text{Tl}^+$  center luminescence by (i) type color centers. However, such an explanation is unacceptable for the following reason. Penetration of 250 keV electrons into CsI(Tl) crystal does not exceed 200  $\mu\text{m}$ . The estimation based on the data of the absorption coefficient dispersion (Fig. 1) shows that the optical density of a layer with 200  $\mu\text{m}$  thickness for 2.59 eV emission is insignificant and equals to about 0.02. It means that the decrease of the transmitted light intensity as a result of re-absorption does not exceed 4%. It should be also noted that the contour of the dip corresponds exactly to the excitation spectrum of 2.1 eV emission, but not to the absorption spectrum of the irradiated crystal.

Thus we believe that the cause of the discussed effects is the energy transfer from the near-activator excitons to the (i) color centers by a more effective resonant mechanism (see, e.g. Ref. [25]). Concentration of  $\text{Tl}^+$  ions in the investigated crystal is estimated as  $10^{19} \text{ cm}^{-3}$ . In this case the average distance between a  $\text{Tl}^+$  center and an activator color center is about 20 nm, which is significantly less than the wavelength of emission. Such ratio is one of the criteria for the possibility of the inductive-resonant energy transfer from the excited emission center (donor D) to the non-excited one (acceptor A) [26,27]. The second important criterion is an equality of the intrinsic frequencies of these two systems. The equality results in the dependence of the probability of the non-radiative energy transfer on the overlapping degree of the emission spectrum of the donor and absorption spectrum of the acceptor. According to quantum-mechanics theory, the constant of non-radiative energy transfer turns out to be proportional to the overlap integral [26]:

$$K_{\text{DA}} = \int_0^{\infty} F_{\text{D}}(v)\varepsilon(v) \frac{dv}{v^4} \quad (5)$$

where  $F_{\text{D}}(v)$  is the emission spectrum of the donor,  $\varepsilon(v)$  the absorption spectrum of the acceptor.

Thus, the origin of the dip (see Fig. 5) could be explained due to the inductive-resonant energy transfer from the donor (“weak off-center” exciton responsible for 2.55 eV wide emission band) to the acceptor ((i) color center responsible for 2.59 eV narrow absorption peak).

The same conclusion can be drawn comparing the cathode luminescence decay curves measured in the microsecond time scale for the non-irradiated and colored samples of CsI(Tl) (Fig. 7a, b). The luminescence of the non-colored sample registered in 2.25 and 2.55 eV bands (curves 1 and 2 in Fig. 7a, respectively) decays exponentially with time constants 12 and 3.6  $\mu\text{s}$ , respectively. Such constants correspond to the life times of the “strong off-center” and “weak off-center” self-trapped excitons (STE) at 80 K [13]. Comparing curves 1 and 2 with curves 1' and 2', respectively (Fig. 7a), one can see that coloration of the crystal reduces duration of near-thallium STE emission, especially at initial stages. The cathode luminescence decay curve registered at 2.1 eV (Fig. 7b, curve 3) is well fitted by three exponents with time constants of 1.0, 3.0 and 10.0  $\mu\text{s}$ .

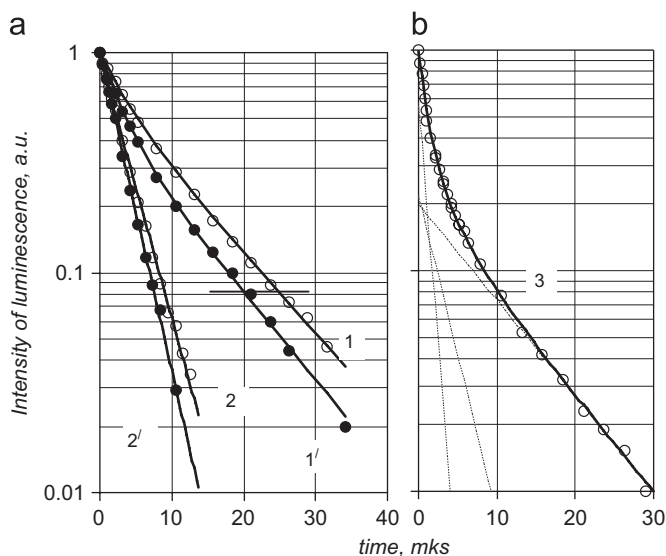


Fig. 7. Curves of the cathode luminescence decay of CsI(Tl) crystal excited by an pulse electron beam at 80 K: (a) at 2.25 eV (1, 1') and at 2.55 eV (2, 2'); (b) at 2.1 eV. 1,2—non-irradiated crystal; 1',2',3— $\gamma$ -irradiated crystal.

It should be noted, that two time constants of 3.0 and 10.0  $\mu$ s coincide, respectively, with the time constants of 2.55 and 2.25 eV emission observed in the colored crystal. In our opinion, there are two experimental facts, which prove the resonant energy transfer from the excited activator  $\text{Tl}^+$  centers (donor) to the color centers (acceptor). The first one is the decay time reduction of the near-activator exciton emission. The second one is the presence of the components typical for  $\text{Tl}^+$  luminescence in the decay kinetics of the (i) center emission. The significant contribution of the component with  $\tau = 1.0 \mu$ s in the decay kinetics of 2.1 eV emission, can be explained by a high probability of the direct excitation of (i) centers with the electron beam and by the presence of the fast component in the decay kinetics of the “strong off-center” STE emission. (Fig. 6, curve 1). The value  $\tau = 1.0 \mu$ s apparently corresponds to the intrinsic frequency of transitions in (i) center with emission of 2.1 eV photons.

#### 4. Conclusion

The study results can be generalized as follows:

(1) The “long-wave shift” of the  $\text{Tl}^+$  center absorption edge was found under the investigation of the influence of the  $\gamma$ -irradiation and the high temperature annealing on the optical absorption of CsI(Tl) crystals. This effect is related to the influence of the anion and cation vacancies induced by irradiation on the energy levels of ions situated in their nearest environment. The scheme of the electron transitions is suggested for the first time to explain the effect of the coloration of CsI(Tl) under exposure to the daylight.

- (2) Either of (i) and (ii) color centers holds the charge carrier of the opposite sign. The model of (i) center as  $\text{Tl}^{2+}v_c^-$  complex is suggested. The excessive positive charge of  $\text{Tl}^{2+}$  ion belonging to  $\text{Tl}^{2+}v_c^-$  center is compensated by the negative charge of a nearby cation vacancy.
- (3) The appearance of the dip in the spectrum of the near-activator STE emission, the decay time reduction of the “strong” and “weak” off-center exciton emission, the appearance of the intensive (i) center emission have been discovered under study of the color centers influence on the spectra and on the decay kinetics of the emission at 80 K. The decay kinetics of (i) centers contains the time components typical for the decay kinetics of near-thallium STE emission. In our opinion, these experimental facts prove that there is an effective non-radiative energy transfer from STE to (i) color centers by the inductive-resonant mechanism in colored CsI(Tl) crystal.

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