Investigation of the Radiation Sensitivity of the Modified SrCl$_2$–Tl$^+$ Crystals

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If the SrCl$_2$–Tl$^+$ crystals get irradiated with ionizing radiation at a temperature of 80–150 K, the low-temperature color centers appear in them, having surplus electric charge relative to the lattice as a result of capturing electrons and holes by the impurity–vacancy dipoles. If the crystal is irradiated at temperatures above 150 K, when the anionic vacancies can migrate, the high-temperature color centers emerge, which are electrically neutral relative to the lattice. Hence, the coloration of crystal is more effective at high temperatures. However, if the crystal irradiated at high temperatures is cooled down to 80 K, then discolored and re-irradiated with ionizing radiation, both types of color centers are effectively generated in it. Namely, SrCl$_2$–Tl$^+$ crystals have "radiation memory" and, as a result of such modifications, the effectiveness of crystal coloration at low temperatures increases several times. The parameters of radiation sensitivity of the SrCl$_2$–Tl$^+$ crystals were calculated in a one-dimensional model. The limit concentrations of the color centers as a function of the concentration of the Tl$^+$-impurity in the crystal were defined. The calculation results are in good agreement with the experimental data, which confirms the validity of using such a one-dimensional model for predicting the radiation properties of real crystals and searching for ways to increase the effectiveness of crystal coloration.

topics: crystal, color centers, model

1. Introduction

SrCl$_2$ crystals are widely used in photolithography, laser technology, and dosimetry as model objects for theoretical research of ionic crystals [1–6]. The "pure" crystals, as is known [7], do not get colored under irradiation, i.e., they are radiation-resistant. When the crystals are being doped with non-isovalent impurities (alkaline metals, thallium, or oxygen), the latter substitute the host ions, whereas the anionic vacancy compensates for excessive (relative to the lattice) charge — thus forming the impurity–vacancy defects [8, 9].

If the SrCl$_2$–Tl$^+$ crystals with such defects get irradiated with ionizing radiation, color centers appear as a result of their capture of electrons and holes. They have different structures at different temperatures [10–13]. For practical application, it is necessary to achieve the maximum concentration of color centers under these conditions. The mere increase of the impurity concentration in the crystal might result in the creation of unwanted sets of color centers. The alternative way of increasing the sensitivity of the crystal consists in performing actions that include heating, cooling, coloring, and decoloring — performed in a certain order. These procedures — executed in a certain sequence —

significantly enhance the radiation sensitivity of the doped crystals [14, 15]. In order to measure the effectiveness of the proposed modification, the concentrations of color centers for various crystal processing methods were calculated using the ion chain model.

Structurally, this paper begins with generalizations of the experimental results and information on the structure of defects in the SrCl$_2$–Tl$^+$ crystals, as well as with the description of the mechanism of color center generation under the influence of ionizing irradiation. Next, based on the results of the experiment, the radiation sensitivity of the SrCl$_2$–Tl$^+$ crystals in the ion chain model is calculated. Finally, a discussion on the obtained results and findings is presented.

2. Point defects structure in the SrCl$_2$–Tl$^+$ crystals before and after their irradiation at different temperatures

The SrCl$_2$–Tl$^+$ crystals were grown in a sealed quartz ampoule using the modified Stockbarger method. Thallium ingress into the crystal, as well as its concentration, was monitored by measuring the thermo-stimulated depolarization (TSD) currents.
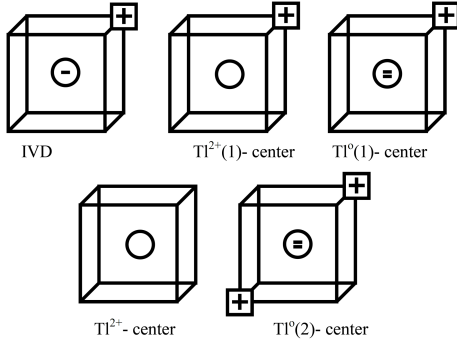
Fig. 1. IVD and color centers models in the SrCl$_2$–Tl$^+$ crystals.

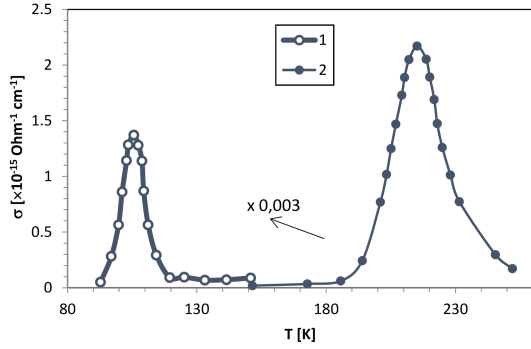Fig. 2. Currents of thermally stimulated depolarization of the SrCl$_2$–Tl$^+$ crystals (at electric field strength $E_p = 5000$ V/cm and heating rate $\beta = 0.1$ K/s).

The measured samples were presented in the form of plates cut out in the plane (100) of the crystal (cleavage plane is (111)). The irradiation conditions of the samples, the methods for measuring the induced absorption spectra and TSD currents are described in our previous paper [8].

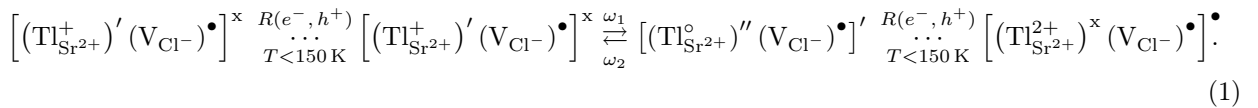As follows from the experimental studies [10–13], Tl$^+$-ions enter the crystal lattice as the substitutional ions. The charge of the Tl$^+$-ion, which is surplus relative to the lattice, is compensated by the anionic chlorine vacancy located in the first coordination sphere. The available data suggest that the following defects can be formed in the SrCl$_2$–Tl$^+$ crystals:

⊖ — Tl$^+$-ion in place of Sr^{2+} $\left(\mathrm{Tl}^+_{\mathrm{Sr}^{2+}}\right)'$,

⊜ — Tl$^\circ$ atom in place of Sr^{2+} $\left(\mathrm{Tl}^\circ_{\mathrm{Sr}^{2+}}\right)''$,

◯ — Tl^{2+}-ion in place of Sr^{2+} $\left(\mathrm{Tl}^{2+}_{\mathrm{Sr}^{2+}}\right)^{\mathrm{x}}$,

⊞ — the chlorine vacancy $(\mathrm{V}_{\mathrm{Cl}^-})^\bullet$,

⊖⊞ — impurity–vacancy dipole (IVD) $\left[\left(\mathrm{Tl}^+_{\mathrm{Sr}^{2+}}\right)'(\mathrm{V}_{\mathrm{Cl}^-})^\bullet\right]^{\mathrm{x}}$,

⊜⊞ — Tl$^\circ$(1)-center $\left[\left(\mathrm{Tl}^\circ_{\mathrm{Sr}^{2+}}\right)''(\mathrm{V}_{\mathrm{Cl}^-})^\bullet\right]'$,

◯⊞ — Tl^{2+}(1)-center $\left[\left(\mathrm{Tl}^{2+}_{\mathrm{Sr}^{2+}}\right)^{\mathrm{x}}(\mathrm{V}_{\mathrm{Cl}^-})^\bullet\right]$,

⊜⊞⊞ — Tl$^\circ$(2)-center $\left[\left(\mathrm{Tl}^\circ_{\mathrm{Sr}^{2+}}\right)''(\mathrm{V}_{\mathrm{Cl}^-})(\mathrm{V}_{\mathrm{Cl}^-})^\bullet\right]^{\mathrm{x}}$,

⊖⊞⊞ — Tl$^+$(2)-center $\left[\left(\mathrm{Tl}^+_{\mathrm{Sr}^{2+}}\right)'(\mathrm{V}_{\mathrm{Cl}^-})^\bullet(\mathrm{V}_{\mathrm{Cl}^-})^\bullet\right]$.

Color centers models are shown in Fig. 1.

IVD can be detected while studying TSD currents. As is known from the investigation of TSD currents in the SrCl$_2$–Tl$^+$ crystals [9], at temperatures $T < 107$ K, anionic vacancies are located in the vicinity of thallium ions and are "frozen" in the crystal lattice. At $T > 110$ K, anionic vacancies are only able to perform rotational motion in the vicinity of the impurity. At $T \simeq 215$ K, thermal dissociation of dipoles is observed, i.e., anionic vacancy detaches from the Tl$^+$-ion. Temperature dependence of the dipole relaxation currents of the SrCl$_2$–Tl$^+$ crystals is shown in Fig. 2.

Under low-temperature irradiation of the SrCl$_2$–Tl$^+$ crystals, as a result of localization of charge carriers on the $\left[\left(\mathrm{Tl}^+_{\mathrm{Sr}^{2+}}\right)'(\mathrm{V}_{\mathrm{Cl}^-})^\bullet\right]^{\mathrm{x}}$ impurity–vacancy dipoles, color centers are generated, i.e.,

$$\left[\left(\mathrm{Tl}^+_{\mathrm{Sr}^{2+}}\right)'(\mathrm{V}_{\mathrm{Cl}^-})^\bullet\right]^{\mathrm{x}} \overset{R(e^-,h^+)}{\underset{T<150\,\mathrm{K}}{\cdots}} \left[\left(\mathrm{Tl}^+_{\mathrm{Sr}^{2+}}\right)'(\mathrm{V}_{\mathrm{Cl}^-})^\bullet\right]^{\mathrm{x}} \overset{\omega_1}{\underset{\omega_2}{\rightleftarrows}} \left[\left(\mathrm{Tl}^\circ_{\mathrm{Sr}^{2+}}\right)''(\mathrm{V}_{\mathrm{Cl}^-})^\bullet\right]' \overset{R(e^-,h^+)}{\underset{T<150\,\mathrm{K}}{\cdots}} \left[\left(\mathrm{Tl}^{2+}_{\mathrm{Sr}^{2+}}\right)^{\mathrm{x}}(\mathrm{V}_{\mathrm{Cl}^-})^\bullet\right]^\bullet.$$

(1)

Here, $R(e^-,h^+)$ stands for the electron–hole pair (e^-,h^+) generated by radiation; ω_1 is the probability of capturing the charge carriers (e^-,h^+) by the IVD pairs and, accordingly, of the emergence of the $\{[(\mathrm{Tl}^\circ_{\mathrm{Sr}^{2+}})''(\mathrm{V}_{\mathrm{Cl}^-})^\bullet]' - [(\mathrm{Tl}^{2+}_{\mathrm{Sr}^{2+}})^{\mathrm{x}}(\mathrm{V}_{\mathrm{Cl}^-})^\bullet]^\bullet\}$-pairs of color centers; ω_2 is the probability of capturing the charge carriers (e^-,h^+) by the pairs of $\{[(\mathrm{Tl}^\circ_{\mathrm{Sr}^{2+}})''(\mathrm{V}_{\mathrm{Cl}^-})^\bullet]' - [(\mathrm{Tl}^{2+}_{\mathrm{Sr}^{2+}})^{\mathrm{x}}(\mathrm{V}_{\mathrm{Cl}^-})^\bullet]^\bullet\}$ color centers and, accordingly, of the restoration of the impurity–vacancy dipoles.

Figure 3 shows the induced absorption spectra of the SrCl$_2$–Tl$^+$ crystals irradiated by X-rays at different temperatures.

According to (1), radiation destruction of each of the dipole pairs causes the emergence of $\{[(\mathrm{Tl}^\circ_{\mathrm{Sr}^{2+}})''(\mathrm{V}_{\mathrm{Cl}^-})^\bullet]' - [(\mathrm{Tl}^{2+}_{\mathrm{Sr}^{2+}})^{\mathrm{x}}(\mathrm{V}_{\mathrm{Cl}^-})^\bullet]^\bullet\}$-pair of color centers, each having surplus electric charge relative to the lattice. This disrupts the thermodynamic equilibrium of the crystal lattice and increases the potential energy. In contrast,
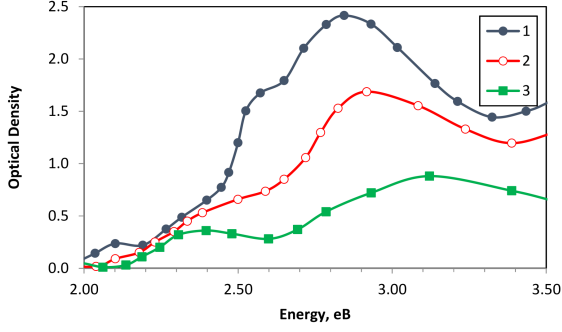
Fig. 3. Induced absorption spectra of the $SrCl_2-$ Tl^+ crystals: after being irradiated at 80 K — line 3; after being irradiated at 200 K — line 1; after being irradiated at 200 K, decolored and re-irradiated at 80 K — line 2.
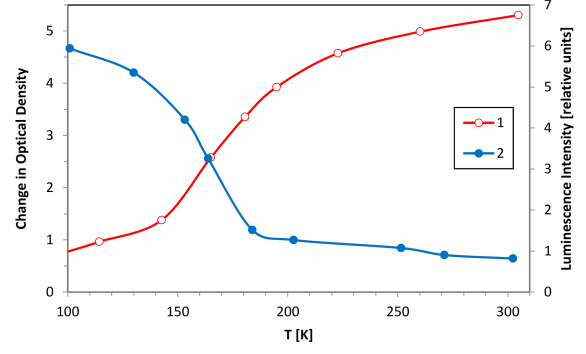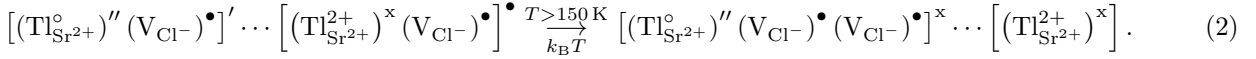

Fig. 4. Temperature dependence of the optical absorption of color centers (line 1) and the intensity of X-ray stimulated luminescence (line 2) of the $SrCl_2-$ Tl^+ crystals.

the localization of electrons and holes on the $\{[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl^-})^{\bullet}]' - [(Tl^{2+}_{Sr^{2+}})^x(V_{Cl^-})^{\bullet}]^{\bullet}\}$-pairs of color centers leads to the emergence of the impurity–vacancy dipoles which are electrically neutral relative to the lattice.
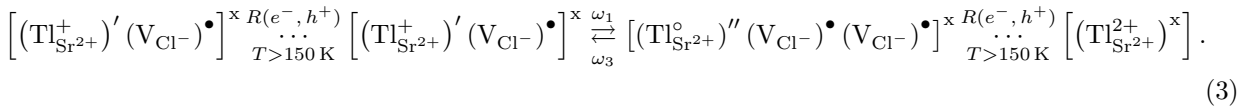
When heating up the crystals that have been colored at low temperatures, the low-temperature $\{[(Tl^{2+}_{Sr^{2+}})''(V_{Cl^-})^{\bullet}]' - [(Tl^{2+}_{Sr^{2+}})^x(V^-_{Cl})^{\bullet}]^{\bullet}\}$-pairs of color centers are transformed into new high-temperature pairs of color centers

$$\left[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl^-})^{\bullet}\right]' \cdots \left[(Tl^{2+}_{Sr^{2+}})^x(V_{Cl^-})^{\bullet}\right]^{\bullet} \xrightarrow[k_B T]{T > 150\,K} \left[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl^-})^{\bullet}(V_{Cl^-})^{\bullet}\right]^x \cdots \left[(Tl^{2+}_{Sr^{2+}})^x\right]. \tag{2}$$

This is caused by the detachment of anionic vacancy from the $(Tl^{2+}_{Sr^{2+}})^x$ substitutional ion, which is electrically neutral relative to the lattice, followed by the vacancy capture by $[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl^-})^{\bullet}]'$-center, which is negatively charged relative to the lattice. This means that there occurs a transition from a volumetric mechanism to a localized mechanism of electric charge compensation. Since the new high-temperature $[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl^-})^{\bullet}(V_{Cl^-})^{\bullet}]^x$

and $(Tl^{2+}_{Sr^{2+}})^x$ color centers are electrically neutral relative to the lattice, the process of their formation is accompanied by a decrease in the intensity of X-ray stimulated luminescence and an increase in the efficiency of color center generation (Fig. 4).

In fact, $\{[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl^-})^{\bullet}(V_{Cl^-})^{\bullet}]^x - [(Tl^{2+}_{Sr^{2+}})^x]\}$-pairs of color centers can also be generated by irradiating the crystals at $T > 150$ K,
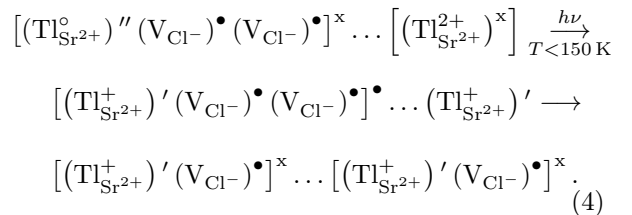
$$\left[(Tl^+_{Sr^{2+}})'(V_{Cl^-})^{\bullet}\right]^x \underset{T>150\,K}{\overset{R(e^-,\,h^+)}{\cdots}} \left[(Tl^+_{Sr^{2+}})'(V_{Cl^-})^{\bullet}\right]^x \underset{\omega_3}{\overset{\omega_1}{\rightleftarrows}} \left[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl^-})^{\bullet}(V_{Cl^-})^{\bullet}\right]^x \underset{T>150\,K}{\overset{R(e^-,\,h^+)}{\cdots}} \left[(Tl^{2+}_{Sr^{2+}})^x\right]. \tag{3}$$

Here, ω_3 stands for the probability of destruction of $\{[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl^-})^{\bullet}(V_{Cl^-})^{\bullet}]^x - [(Tl^{2+}_{Sr^{2+}})^x]\}$-pairs of color centers during the disintegration of the electron–hole pair in the ion chain.

3. Calculating radiation sensitivity of the crystals after re-irradiation

High-temperature $[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl^-})^{\bullet}(V_{Cl^-})^{\bullet}]^x$ and $\left[(Tl^{2+}_{Sr^{2+}})^x\right]$ color centers can be destructed by illuminating the crystal with the light in $[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl^-})^{\bullet}(V_{Cl^-})^{\bullet}]^x$-absorption bands. If

the crystal is discolored at $T > 150$ K, then these centers are completely destructed, and impurity–vacancy dipoles are restored. Therefore,

$$\left[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl^-})^{\bullet}(V_{Cl^-})^{\bullet}\right]^x \cdots \left[(Tl^{2+}_{Sr^{2+}})^x\right] \xrightarrow[T<150\,K]{h\nu}$$

$$\left[(Tl^+_{Sr^{2+}})'(V_{Cl^-})^{\bullet}(V_{Cl^-})^{\bullet}\right]^{\bullet} \cdots (Tl^+_{Sr^{2+}})' \longrightarrow$$

$$\left[(Tl^+_{Sr^{2+}})'(V_{Cl^-})^{\bullet}\right]^x \cdots \left[(Tl^+_{Sr^{2+}})'(V_{Cl^-})^{\bullet}\right]^x. \tag{4}$$

If photobleaching is carried out at temperatures $T < 150$ K, then this will lead to the appearance of new defects, i.e.,

$$\left[\left(\mathrm{Tl}^{\circ}_{\mathrm{Sr}^{2+}}\right)''\left(\mathrm{V}_{\mathrm{Cl}-}\right)^{\bullet}\left(\mathrm{V}_{\mathrm{Cl}-}\right)^{\bullet}\right]^{\mathrm{x}}\ldots\left[\left(\mathrm{Tl}^{2+}_{\mathrm{Sr}^{2+}}\right)^{\mathrm{x}}\right]\xrightarrow[T<150\,\mathrm{K}]{h\nu}\left[\left(\mathrm{Tl}^{+}_{\mathrm{Sr}^{2+}}\right)'\left(\mathrm{V}_{\mathrm{Cl}-}\right)^{\bullet}\left(\mathrm{V}_{\mathrm{Cl}-}\right)^{\bullet}\right]^{\bullet}\ldots\left(\mathrm{Tl}^{+}_{\mathrm{Sr}^{2+}}\right)'. \qquad (5)$$

Such defects are absent in newly grown crystals, which have only impurity–vacancy dipoles. As opposed to dipoles, $[(\mathrm{Tl}^{+}_{\mathrm{Sr}^{2+}})'(\mathrm{V}_{\mathrm{Cl}-})^{\bullet}(\mathrm{V}_{\mathrm{Cl}-})^{\bullet}]^{\bullet}$ and $(\mathrm{Tl}^{+}_{\mathrm{Sr}^{2+}})'$-defects have an effective electric charge relative to the lattice and substantially alter the properties of the crystal — in particular, its radiation sensitivity. During repeated low-temperature irradiation of discolored crystals, in addition to the processes of capturing the electrons and holes by electrically neutral dipoles (see (1)), the processes of capturing the electrons and holes by the created charged defects proceed in parallel. This leads to the generation of "high-temperature" color centers in the crystal by X-ray irradiation at low temperatures,

$$\left[\left(\mathrm{Tl}^{+}_{\mathrm{Sr}^{2+}}\right)'\left(\mathrm{V}_{\mathrm{Cl}-}\right)^{\bullet}\left(\mathrm{V}_{\mathrm{Cl}-}\right)^{\bullet}\right]^{\bullet}\underset{T<150\,\mathrm{K}}{\overset{R(e^-,\,h^+)}{\cdots}}\left(\mathrm{Tl}^{+}_{\mathrm{Sr}^{2+}}\right)'\underset{\omega_5}{\overset{\omega_4}{\rightleftarrows}}\left[\left(\mathrm{Tl}^{\circ}_{\mathrm{Sr}^{2+}}\right)''\left(\mathrm{V}_{\mathrm{Cl}-}\right)^{\bullet}\left(\mathrm{V}_{\mathrm{Cl}-}\right)^{\bullet}\right]^{\mathrm{x}}\underset{T<150\,\mathrm{K}}{\overset{R(e^-,\,h^+)}{\cdots}}\left(\mathrm{Tl}^{2+}_{\mathrm{Sr}^{2+}}\right)^{\mathrm{x}},$$
$$(6)$$

where ω_4 is the probability of generation of $\{[(\mathrm{Tl}^{\circ}_{\mathrm{Sr}^{2+}})''(\mathrm{V}_{\mathrm{Cl}-})^{\bullet}(\mathrm{V}_{\mathrm{Cl}-})^{\bullet}]^{\mathrm{x}}-[(\mathrm{Tl}^{2+}_{\mathrm{Sr}^{2+}})^{\mathrm{x}}]\}$-pairs of color centers during the dissolution of electron–hole pairs; ω_5 is the probability of bleaching of $\{[(\mathrm{Tl}^{\circ}_{\mathrm{Sr}^{2+}})''(\mathrm{V}_{\mathrm{Cl}-})^{\bullet}(\mathrm{V}_{\mathrm{Cl}-})^{\bullet}]^{\mathrm{x}}-[(\mathrm{Tl}^{2+}_{\mathrm{Sr}^{2+}})^{\mathrm{x}}]\}$-pairs of color centers during the dissolution of electron–hole pairs ($\omega_5=\omega_3$).

Resulting from the reaction (6), the $(\mathrm{Tl}^{2+}_{\mathrm{Sr}^{2+}})^{\mathrm{x}}$- and $[(\mathrm{Tl}^{\circ}_{\mathrm{Sr}^{2+}})''(\mathrm{V}_{\mathrm{Cl}-})^{\bullet}(\mathrm{V}_{\mathrm{Cl}-})^{\bullet}]^{\mathrm{x}}$ color centers emerge, which are electrically neutral relative to the lattice. The efficiency of destructing such centers by the electron–hole pairs is lower than for $[(\mathrm{Tl}^{\circ}_{\mathrm{Sr}^{2+}})''(\mathrm{V}_{\mathrm{Cl}-})^{\bullet}]'$ and $[(\mathrm{Tl}^{2+}_{\mathrm{Sr}^{2+}})^{\mathrm{x}}(\mathrm{V}_{\mathrm{Cl}-})^{\bullet}]^{\bullet}$ charged centers, which leads to the increase in radiation sensitivity of the crystal at low temperatures.

In a one-dimensional model, the real crystal can be represented as an ion chain containing the host ions and impurity–vacancy dipoles. Based on this model, it is possible to calculate the radiation sensitivity parameters of crystals after low-temperature irradiation and re-irradiation of previously discolored crystals.

In so doing, we proceeded from the following. Ionizing radiation generates electron–hole pairs in the ion chain. An electron–hole pair can appear with equal probability at any of the nodes of the ion chain. The distance between the electron and the hole depends on the excitation energy of the electron. The position of the hole remains unchanged, which is due to its large effective mass. Depending on the relative positions of the electron, the hole, and IVD, the electron and the hole can be captured by the dipole pair to form color centers, or they can recombine with each other. The contribution of each of these processes depends on the distribution of the electric potential along the ion chain.

The calculations were performed as follows. The number of host ions contained in the ion chain is denoted by n. The chain's length is $l=na$, where a is the parameter of the lattice. The average distance between impurities in the crystal lattice is determined by their concentration. The impurity ions concentration varied from 0.01 to 0.5 mol.%.

Calculations of the radiation sensitivity of the $\mathrm{SrCl_2}$–Tl^+ crystals under low-temperature irradiation at 80 K were performed as follows. The irradiation creates the electron–hole pair. Note that in the ion chain, there are $N=n^2$ ways of arrangement of the electron and the hole. Each of them was taken into account during the calculation of the electron–hole pair relaxation. The electron–hole pairs can be localized: (i) on the impurity–vacancy dipoles, thus forming a pair of color centers (n_1); (ii) on the same IVD (n_2); (iii) they can annihilate on one of the nodes of the ion chain (n_3). In the present case, $n_1+n_2+n_3=N$. By finding n_1, n_2, and n_3, one can calculate the probability ω_1 of color center generation and the probability ω_2 of their radiation decay as follows

$$\omega_1=\frac{n_1}{N},\quad \omega_2=\frac{n_2}{N}. \qquad (7)$$

At the stage of crystal color saturation, a dynamic equilibrium is established between the processes of generation of color centers and their destruction.

The calculation results are shown in Tables I–III. Table I presents the results of calculating the parameters of radiation sensitivity of the $\mathrm{SrCl_2}$–Tl^+ crystals. Probabilities of generation of color centers are ω_1, ω_4, and probabilities of their radiation decay during the breakdown of the electron–hole pairs in the ion chain fragment are ω_2, ω_3, and ω_5; c is molar concentration of thallium ions in the crystal; l is the average distance between impurity ions; a is the parameter of the ion chain; c_0 is dipole pairs concentration in the crystal lattice before its irradiation, i.e., $c_0=\frac{1}{2}c$; c_1 is the concentration of $\{[(\mathrm{Tl}^{\circ}_{\mathrm{Sr}^{2+}})''(\mathrm{V}_{\mathrm{Cl}-})^{\bullet}]'-[(\mathrm{Tl}^{2+}_{\mathrm{Sr}^{2+}})^{\mathrm{x}}(\mathrm{V}_{\mathrm{Cl}-})^{\bullet}]^{\bullet}\}$-pairs of color centers after low-temperature irradiation of the samples ($T=80$ K) at the stage of color

saturation of the crystal; c_2 is the concentration of $\{[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl-})^{\bullet}(V_{Cl-})^{\bullet}]^x - (Tl^{2+}_{Sr^{2+}})^x\}$-pairs of color centers after irradiation of the crystal at $T > 150$ K at the stage of color saturation of the crystal, c_3 is the concentration of $\{[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl-})^{\bullet}(V_{Cl-})^{\bullet}]^x - (Tl^{2+}_{Sr^{2+}})^x\}$-pairs of color centers at the stage of color saturation after SrCl$_2$–Tl$^+$ crystals have been irradiated at $T > 150$ K, cooled down to 80 K, discolored and re-irradiated at this temperature.

Limit concentrations of color centers are calculated by the following formulas

$$c_1 = \frac{\omega_1}{\omega_1 + \omega_2} c_0, \quad n_1 = \frac{\omega_1}{\omega_1 + \omega_2} n_0, \qquad (8)$$

$$c_2 = \frac{\omega_1}{\omega_1 + \omega_3} c_0, \quad n_2 = \frac{\omega_1}{\omega_1 + \omega_3} n_0, \qquad (9)$$

$$c_3 = \frac{\omega_4}{\omega_4 + \omega_5} c_2, \quad n_3 = \frac{\omega_4}{\omega_4 + \omega_5} n_2, \qquad (10)$$

$$c_3 = \frac{\omega_4}{\omega_4 + \omega_5} \frac{\omega_1}{\omega_1 + \omega_3} c_0,$$

$$n_3 = \frac{\omega_4}{\omega_4 + \omega_5} \frac{\omega_1}{\omega_1 + \omega_3} n_0, \qquad (11)$$

where n_0 is the IVD concentration in the crystal before the ionizing irradiation.

4. Discussion of results

As evidenced by the data in Table I, at low-temperature irradiation of the crystal, the probability ω_1 of generation of low-temperature color centers is lower than the probability ω_2 of their radiation decay. That is why the concentration c_1 of the $\{[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl-})^{\bullet}]' - [(Tl^{2+}_{Sr^{2+}})^x(V_{Cl-})^{\bullet}]^{\bullet}\}$-pairs of low-temperature color centers at the stage of color saturation is considerably lower than the concentration c_0 of impurity–vacancy dipole pairs before irradiation (Table II). Thus, if the impurity concentration is 0.1 mol.%, then only 16% of the impurity–vacancy dipoles are transformed into low-temperature color centers. However, if the crystal is irradiated at $T > 150$ K, the probability ω_3 of disintegration of $[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl-})^{\bullet}(V_{Cl-})^{\bullet}]^x$ and $[(Tl^{2+}_{Sr^{2+}})^x]$ high-temperature electroneutral centers is lower than the probability ω_2 of disintegration of low-temperature centers that are charged relative to the lattice. As a result, the concentration c_2 in the crystal of high-temperature color centers at temperatures $T > 150$ K is several times higher than the concentration c_1 of the low-temperature color centers at $T < 150$ K. Thus, if the impurity concentration constitutes 0.1 mol.%, then 43 % of the impurity–vacancy dipoles are transformed into high-temperature color centers.

The concentration of color centers at $T < 150$ K can be increased by performing the following actions. (i) Irradiating the SrCl$_2$–Tl$^+$ crystal by X-rays at temperatures higher than 150 K. As a result, about 40% of the impurity–vacancy dipoles are converted to high-temperature color centers. (ii) Cooling the crystal down to the temperature of liquid nitrogen and optically discoloring it. This makes $\{[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl-})^{\bullet}(V_{Cl-})^{\bullet}]^x - [(Tl^{2+}_{Sr^{2+}})^x]\}$ electroneutral pairs of color centers convert to $[(Tl^+_{Sr^{2+}})'(V_{Cl-})^{\bullet}(V_{Cl-})^{\bullet}]^{\bullet}$ and $(Tl^+_{Sr^{2+}})'$ charged defects, which are stable at low temperatures. (iii) If the crystal is re-irradiated at liquid-nitrogen temperatures, these defects — which are charged relative to the lattice — capture electrons and holes more effectively (ω_4) compared to electroneutral dipoles (ω_1). Nearly 80–90% of these defects (c_3/c_2) are transformed back into color centers $\{[(Tl^{\circ}_{Sr^{2+}})''(V_{Cl-})^{\bullet}(V_{Cl-})^{\bullet}]^x - [(Tl^{2+}_{Sr^{2+}})^x]\}$.

As a result, at low temperatures, in addition to the "regular" charged low-temperature color centers, high-temperature electroneutral centers are also generated, and this significantly increases the radiation sensitivity of the crystal. Thus, if the impurity concentration is 0.1 mol.%, then 16% of the impurity–vacancy dipoles turn into low-temperature color centers whose concentration constitutes 9% of the concentration of impurity–vacancy dipoles in the newly grown crystal, whereas 84% of the charged defects will turn into high-temperature centers, the concentration of which will be 36% of the concentration of impurity–vacancy dipoles in the newly grown crystal. The total concentration of color centers at liquid-nitrogen temperatures will be 45%, while at direct low-temperature irradiation it will be only 16%.

TABLE I

Probabilities of generation and bleaching of color centers in the SrCl$_2$–Tl$^+$ crystals.

c [mol.%]	l	ω_1	ω_2	ω_3	ω_4	ω_5
0.50	$6a$	0.100	0.40	0.150	0.40	0.100
0.10	$10a$	0.069	0.37	0.088	0.37	0.069
0.01	$21a$	0.032	0.36	0.039	0.36	0.032

TABLE II

Limit concentrations of color centers.

c [mol.%]	l	$c_1 : c_0$	$c_2 : c_0$	$c_3 : c_2$	$c_3 : c_0$
0.50	$6a$	0.20	0.40	0.80	0.32
0.10	$10a$	0.16	0.43	0.84	0.36
0.01	$21a$	0.082	0.45	0.92	0.41

TABLE III

Concentrations of color center pairs in the crystal at the stage of color saturation.

c [mol.%]	l	n_1 [cm^{-3}]	n_2 [cm^{-3}]	n_3 [cm^{-3}]	$n_3 : n_1$
0.50	$6a$	6.5×10^{18}	13×10^{18}	10.4×10^{18}	1.60
0.10	$10a$	1.1×10^{18}	2.8×10^{18}	2.3×10^{18}	2.25
0.01	$21a$	5.33×10^{16}	2.9×10^{17}	2.7×10^{17}	5.02

5. Conclusions

This paper presents the results of theoretical calculations of the parameters of radiation sensitivity of the $SrCl_2$–Tl^+ crystals, namely, the probabilities of capturing electrons and holes by the impurity–vacancy defects as well as the limit concentrations of centers at the stage of crystal color saturation in the model of a one-dimensional ion chain. The obtained results satisfactorily correlate with the experimental studies.

It is shown that by manipulating the crystal, the concentration of color centers can be increased several times. So, if the impurity concentration is 0.1 mol.%, then under regular irradiation the number of color centers is 1.1×10^{18} cm^{-3}, whereas after the manipulations, their number increases to 3.1×10^{18} cm^{-3}. The enhancement of the radiation sensitivity of the modified thallium-containing $SrCl_2$–Tl^+ crystals can improve the performance characteristics of the devices operating on their basis.

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