Time-resolved luminescence of CsI–Tl crystals excited by pulsed electron beam

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Abstract

Results of the time-resolved measurements of CsI–Tl crystal luminescence under electron pulse excitation at room temperature are reported. The rise and decay times of both 400 and 500 nm emission bands have been measured. The results obtained show that the main mechanism of the luminescence excitation is hole recombination luminescence which is due to self-trapped hole migration toward TlO centres. Stimulation spectra of photostimulated luminescence of CsI–Tl are also analyzed.

1. Introduction

For many years, CsI–Tl is one of the widely used scintillation crystals for the detection of all kinds of ionizing particles [1–12]. However, the mismatch of its emission spectrum to the spectral sensitivity of the common photomultiplier have prevented its more widespread use. The recent development of the large area silicon photodiode detectors well matched to the emission spectrum of CsI–Tl has caused the renaissance of its applications. It was also stimulated a revival of an interest to the studies of the scintillation processes in CsI–Tl crystals.

The spectrum of its luminescence has been reported to extend from 300 to 800 nm, having up to four emission bands, of which the most of prevalent peaks were at about 550–560 nm. Although this luminescence band has been studied intensively for a long time there is still controversial discussion about the origin of this band. It was assigned as being a Tl+ intra-centre transition [13]. The donor–acceptor recombination between a TlO centre and a Vc centre has been considered by Gutan et al. [14], whereas the mechanism of recombination luminescence of exciton captured at Tl+ ion has been suggested by Smol’skaya and Kolesnikova [15]. Recently Spaeth et al. [16] have demonstrated direct experimental evidence by ODEPR method that the X-ray induced luminescence at 560 nm is explained as a recombination luminescence of a kind of Tl-bound exciton. The model explaining the 550 nm band as a luminescence of the self-trapped exciton located near Tl+ ion has been proposed by Nagimyl et al. [17]. The other component in the luminescence spectra peaking at 400–420 nm has been associated with I− vacancies [18] or F centres [11,19]. It should be noted that F centre absorption band peaks at 800 nm [20], therefore F centre luminescence should be observed in infrared spectral region. Systematic study of steady-state emission properties has been made by Sivasankar and Jacobs [21], who ascribed both these emission bands to transitions within isolated Tl+ ions.

The excitation of scintillation in doped alkali-halide crystals is determined by two processes [22]:

1. Hole recombination mechanism (HRL)

\[
e + p + \text{Tl}^+ \rightarrow V_k + \text{Tl}^0 \rightarrow \text{STE} + \text{Tl}^+ \rightarrow (\text{Tl}^+)^
\]

\[
\rightarrow \text{Tl}^+ + h\nu.
\]

2. Electron recombination mechanism (ERL)

\[
e + p + 2\text{Tl}^+ \rightarrow V_k + \text{Tl}^0 + \text{Tl}^+ \rightarrow \text{Tl}^0 + \text{Tl}^2+
\]

\[
\rightarrow \text{Tl}^+ + (\text{Tl}^+)^
\]

\[
\rightarrow \text{Tl}^+ + \text{Tl}^+ + h\nu.
\]

In order to evaluate the contribution of these processes in CsI–Tl scintillations an attempt was made to elucidate the origin of 400 nm luminescence band rising under ionizing radiation. Spectra of photo- and thermo-stimulated luminescence (PSL and TSL) as well as kinetics of rise and decay of luminescence under electron pulse excitation were measured.

2. Results and discussion

Steady-state emission spectra were measured under excitation by photons of different energies (Fig. 1). At 80 K

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The excitation of Ti$^+$ centres under ionizing irradiation can occur due to: (i) electron recombination mechanism; or (ii) thermoactivated energy transfer between two minima of the adiabatic potential energy surface (APES), connecting Ti$^+$ and STE near Ti$^+$. At high temperatures ($T > 180$ K) the energy transfer from STE near Ti$^+$ to Ti$^+$ causes the excitation of thallium centre. This was proved by the measurements of the thermal evolution of 400 and 550 nm for both PSL and TSL processes [22,24].

It is known that the rise time activation energy of the 550 nm emission corresponds closely to the activation energy of $V_k$ centres in CsI [19]. As follows from Fig. 2 the rise time of the 400 nm band is slightly longer than that of the 550 nm one. We can propose that the appearance of such component with $\tau$ longer than rise time corresponding to $V_k$ centre migration toward Ti$^0$ centre maybe connected with some shallow electron traps. Then the small ERL component can be ascribed to an electron release from such shallow trap and its recombination with $V_{k,a}$ or Ti$^{2+}$ centre.

In order to clarify the origin of the above-mentioned electron traps, we measure the PSL stimulation spectra. The high sensitivity (up to $10^9$ centres/cm$^{-2}$) [25] of the PSL allows to obtain an important information about optical properties of the defect centres participating in the recombination processes. It is especially important for the CsI–Ti crystals, which has well detected absorption bands only of self-trapped holes ($V_k$ centres) and $F$ centres. Fig. 3 shows the photostimulation spectra of the 550 and 400 nm luminescence bands after irradiation in the D-absorption band (225 nm) at 80 K. Explaining results the obtained it should be taken into account that the D-absorption band corresponds to the excitation of the halide ion located near the Ti$^+$ ion. The excitation created in such a way can dissociate giving electron–hole pair. The electron is cap-

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**Fig. 1.** Photoluminescence of CsI–Ti excited by photons with wavelength 200 nm (1), 225 nm (2) and 300 nm (3) at 300 K (a) and 80 K (b).

**Fig. 2.** CsI–Ti luminescence rise and decay in 550 nm band (1) and 400 nm band (2) under electron pulse at 300 K (a); fitting of the luminescence decay in coordinates $\ln I$ versus time (b).

**Fig. 3.** Photostimulation spectra of CsI–Ti crystal after irradiation in the D-absorption band ($\lambda = 225$ nm) at 80 K recording the 405 nm luminescence band (a, b, c) and the 550 nm band (d, e, f). The temperatures of the measurements are marked on the figure. The intensity scale of (e) and (f) is 20 times larger than that of (a, b, c, d) figures.

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tured by the nearest Ti⁺ ion forming the Ti⁰ centre, while the hole is self-trapped near the Ti⁰ centre, or is captured by the nearest Ti⁺ ions, forming the Vₓ₋ₓ centre. The Vₓ₋ₓ centres of such type have been recently observed in CsI–Ti crystal [16]. In such a way the excitation in the D-absorption band provides the formation of the spatially correlated pairs of Ti⁰–Vₓ or Ti⁰–Vₓ₋ₓ defects. It should be mentioned that irradiation of CsI–Ti crystal by X-rays provides the TSL glow curves similar to that under excitation in the D-absorption band. This fact proves that ionizing radiation produces also the spatially correlated Ti⁰–Vₓ and Ti⁰–Vₓ₋ₓ pairs. Fig. 3 shows that at 80 K the photostimulation spectra are similar for both luminescence bands and involve a weak 1400 nm band and two composed bands with maxima at 950 and 600 nm. The heating of the crystal irradiated at 80 K on 10–15 K (that is up to temperature of Vₓ centre delocalization – 90 K), or the irradiation of the crystal at that temperature leads to the almost complete disappearance of 1400 and 950 nm stimulation bands. The main part of the short wavelength stimulation band at 600 nm disappears after passing the 125 K glow peak. At higher temperatures the stimulation spectra consist of several weak bands, their origin will be discussed elsewhere. Interpreting the PSL stimulation spectrum of CsI–Ti, it is necessary to remember the recent study of the PSL in the Rbl–Ti crystal [26]. The photostimulation of Rbl–Ti at 130 K involves 3 bands besides the F-band and the position of the long wavelength bands precisely coincides with our data on CsI–Ti crystal. This fact enables to ascribe the photostimulation bands in CsI–Ti to the optical transitions in Ti⁰ centres. The thermal behavior of photo-stimulation bands in CsI–Ti allows to suggest that the 1400 and 950 nm bands belong to Ti⁰ centres perturbed by the close Vₓ centres. Taking into account the thermal stability of the stimulation bands we can suggest that 600 nm band is likely to correspond to the unperturbed Ti⁰ centres.

The last figure shows the preliminary result of the study of the luminescence yield behavior as a function of excitation density upon 10 keV electron irradiation (Fig. 4). The increase of scintillation efficiency for low values of excitation density can be explained by the increasing of fraction of electron-hole recombination at Ti⁺ sites. The prominent decline in scintillation efficiency for high values of excitation density is in a qualitative agreement with the results of Gwin and Murray [6], where it was concluded that such decline is not due to saturation of activator centres. They have reported that a narrower emission near 330 nm together with a broad Ti emission centered at 550 nm has been observed upon 1.4 MeV protons and 8.7 MeV alpha particles irradiations [6]. Furthermore it was established that the intensity of the 330 nm band relative to the common Ti emission band is the greatest upon excitation by radiation of high dE/dx. The probable origin of the 330 nm band has been discussed in Ref. [27], where a fast luminescence in the spectral region of 300–340 nm in pure and doped CsI crystals has been observed. It was found that luminescence rise time is less than 50 ps and the decay time is 1 ns at room temperature. It has been suggested that the emission center is formed as a result of the interaction of several electronic excitations. It is clear that more experiments are needed to elucidate both phenomena: the decline of 550 nm band for high dE/dx values and the origin of the fast luminescence in the spectral region 300–340 nm.

Fig. 4. Luminescence yield of 550 nm luminescence as a function of relative excitation density upon irradiation of CsI–Ti by 10 keV electrons.

References


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