



ELSEVIER

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

A.I. Popov ^{a,*}, S.A. Chernov ^b, L.E. Trinkler ^b

^a CIRIL, CEA-CNRS, Rue Claude Bloch, BP-5133, F-14040 Caen Cedex, France

^b Institute of Solid State Physics, University of Latvia, 8 Kengaraga iela, LV-1063 Riga, Latvia

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 Tl^0 centres. Stimulation spectra of photostimulated luminescence of CsI–Tl are also analyzed.

1. Introduction

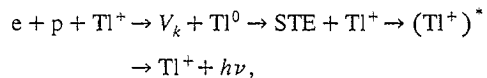
For many years, CsI–Tl is one of the widely used scintillation crystals for the detection of all kinds of ionizing particles [1–12]. However, the mismatch of its emission spectrum to the spectral sensitivity of the common photomultiplier have prevented its more widespread use. The recent development 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 Tl^0 centre and a V_k 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'skya 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 Nagimiyi et al. [17]. The

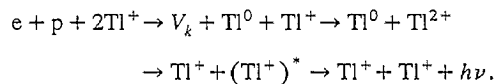
other component in the luminescence spectra peaking at 400–420 nm has been associated with I^- vacancies [18] or F centres [11,19]. It should be noted that F centre absorption band peaks at 800 nm [20], therefore F centre luminescence should be observed in infrared spectral region. Systematic study of 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)



(2) electron recombination mechanism (ERL)



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

* Corresponding author. E-mail: popov@ganac4.in2p3.fr.

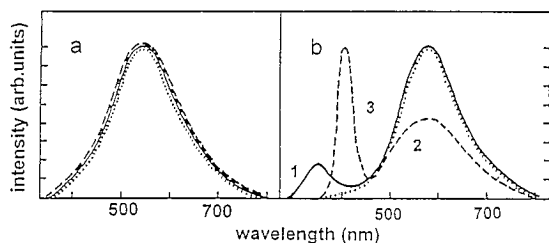


Fig. 1. Photoluminescence of CsI-Tl excited by photons with wavelength 200 nm (1), 225 nm (2) and 300 nm (3) at 300 K (a) and 80 K (b).

the band-to-band excitation (200 nm) causes the luminescence of STE in regular lattice (350 nm) and STE near Tl^+ ion (550 nm), whereas excitation in the A-absorption band (300 nm) rises the appearance of 550 and 405 nm bands of comparable intensity. Excitation in the D-absorption band (225 nm), creating an exciton near Tl^+ ion is the reason of 550 nm luminescence band and a very weak band in the 400 nm region. At 300 K the contribution of 400 nm band in the luminescence spectrum increases, compared with 80 K; luminescence spectra remain the same for all types of excitation. Thus we can conclude that the 400 nm luminescence arising under ionizing radiation is determined by the Tl^+ luminescence.

Results of kinetics measurements under electron pulse irradiation (energy 270 keV, duration 10 ns, excitation density 10 MW/cm^2) at 300 K are shown in Fig. 2. As follows from Fig. 2, the temporal distribution of scintillation light can be represented as a rising component and a decaying exponential. The value of the decay time constant, 780 ns, corresponds closely to that observed in the kinetics of the luminescence under A-band excitation, where the same components of the decay time for both bands at $T > 180 \text{ K}$ were found [23]. Thus we get another reason for interpretation of the 400 nm band as the emission of Tl^+ centre.

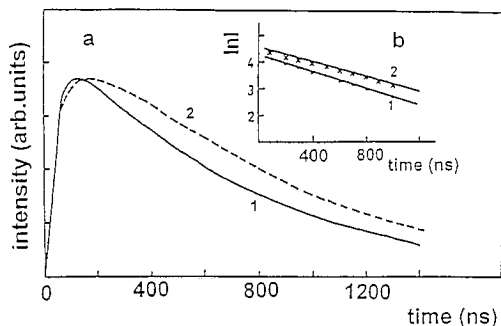


Fig. 2. CsI-Tl luminescence rise and decay in 550 nm band (1) and 400 nm band (2) under electron pulse at 300 K (a); fitting of the luminescence decay in coordinates $\ln I$ versus time (b).

The excitation of Tl^+ 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 Tl^+ and STE near Tl^+ . At high temperatures ($T > 180 \text{ K}$) the energy transfer from STE near Tl^+ to Tl^+ causes the excitation of thallium center. 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 τ longer than rise time corresponding to V_k centre migration toward Tl^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_{kA} or Tl^{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 $10^9 \text{ centres/cm}^{-3}$) [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-Tl 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 Tl^+ ion. The excitation created in such a way can dissociate giving electron-hole pair. The electron is cap-

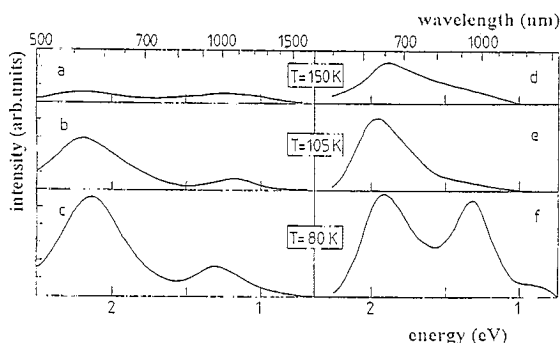


Fig. 3. Photostimulation spectra of CsI-Tl crystal after irradiation in the D-absorption band ($\lambda = 225 \text{ nm}$) at 80 K recording the 405 nm luminescence band (a, b, c) and the 550 nm band (d, e, f). The temperatures of the measurements are marked on the figure. The intensity scale of (e)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

tured by the nearest Tl^+ ion forming the Tl^0 centre, while the hole is self-trapped near the Tl^0 centre, or is captured by the nearest Tl^+ ion, forming the V_{kA} centre. The V_{kA} centres of such type have been recently observed in CsI–Tl crystal [16]. In such a way the excitation in the D-absorption band provides the formation of the spatially correlated pairs of Tl^0-V_k or Tl^0-V_{kA} defects. It should be mentioned that irradiation of CsI–Tl 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 Tl^0-V_k and Tl^0-V_{kA} 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 V_k 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–Tl, it is necessary to remember the recent study of the PSL in the RbI–Tl crystal [26]. The photostimulation of RbI–Tl 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–Tl crystal. This fact enables to ascribe the photostimulation bands in CsI–Tl to the optical transitions in Tl^0 centres. The thermal behavior of photostimulation bands in CsI–Tl allows to suggest that the 1400 and 950 nm bands belong to Tl^0 centres perturbed by the close V_k 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 Tl^0 centres.

The last figure shows the preliminary result of the study of the luminescence yield behavior as a function of

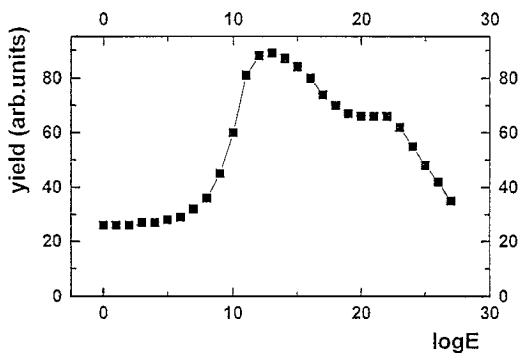


Fig. 4. Luminescence yield of 550 nm luminescence as a function of relative excitation density upon irradiation of CsI–Tl by 10 keV electrons.

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 Tl^+ 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 Tl 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 Tl 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.

References

- [1] R. Hofstadter, *Nucleonics* 6 (1950) 70.
- [2] R.S. Storey, W. Jack and A. Ward, *Proc. Phys. Soc.* 72 (1958) 1.
- [3] G. Hrehuss, *Nucl. Instr. and Meth.* 8 (1960) 344.
- [4] J.S. Robertson and J.G. Lynch, *Proc. Phys. Soc.* 77 (1961) 751.
- [5] F.E. Senftle, P. Martinez and B.P. Aleksna, *Rev. Sci. Instr.* 33 (1962) 819.
- [6] R. Gwin and R.B. Murray, *Phys. Rev.* 131 (1963) 501; and 131 (1963) 508.
- [7] S. Keszthelyi-Landori and G. Hrehuss, *Nucl. Instr. and Meth.* 68 (1969) 9.
- [8] C. Crannell, R.J. Kurz and W. Vienmann, *Nucl. Instr. and Meth.* 115 (1974) 253.
- [9] B.C. Grabmaier, *IEEE Trans. Nucl. Sci.* NS31 (1984) 372.
- [10] H. Grassmann, E. Lorenz and H.-G. Moser, *Nucl. Instr. and Meth.* 228 (1985) 323.
- [11] P. Schotanus, R. Kamermans and P. Dorenbos, *IEEE Trans. Nucl. Sci.* NS37 (1990) 177.
- [12] T. Masuda, M. Tomizawa, Y. Nagashima, N. Nakamoto and Y. Tagishi, *Nucl. Instr. and Meth. A* 322 (1992) 135.
- [13] S. Masunaga, I. Morita and M. Ishiguro, *J. Phys. Soc. Japan* 21 (1968) 638.
- [14] V.B. Gutan, L.M. Shamovskii, A.A. Dunina and B.S. Gorobets, *Opt. Spectrosc.* 37 (1974) 407.
- [15] L.P. Smol'skaya and T.A. Kolesnikova, *Opt. Spectrosc.* 47 (1979) 292.

- [16] J.-M. Spaeth, W. Meise and K.S. Song, *J. Phys.: Condens. Matter* 6 (1994) 3999.
- [17] V. Nagirnyi, A. Stolovich, S. Zazubovich, V. Zepelin, E. Mihokova, M. Nikl, G.P. Pazzi and L. Salvini, *J. Phys.: Condens. Matter* 7 (1995) 3637.
- [18] J.B. Birks, *The Theory and Practice of Scintillation Counting* (Pergamon, New York, 1964).
- [19] J.D. Valentine, W.M. Moses, S.E. Derenzo, D.K. Wehe and G.F. Knoll, *Nucl. Instr. and Meth. A* 325 (1993) 147.
- [20] P. Avakian and A. Smakula, *Phys. Rev.* 120 (1960) 2007.
- [21] V.S. Sivasankar and P.W.M. Jacobs, *Philos. Mag. B* 51 (1985) 479.
- [22] S.A. Chernov, A.I. Popov and L.E. Trinkler, in: *Proc. Intern. Conf. Inorganic Scintillators and Their Applications* (Delft, 1995), eds. P. Dorenbos and C.W.E. van Eijk, pp. 439–441.
- [23] M. Nikl, J. Hlinka, E. Mihokova, K. Polak, P. Fabeni and G.P. Pazzi, *Philos. Mag. B* 67 (1993) 627.
- [24] S.A. Chernov, A.I. Popov and L.E. Trinkler, *Opt. Spectrosc.* 80 (1996) 592.
- [25] A. Kalnin, I. Plavina, A.I. Popov and A. Tale, *J. Phys.: Condens. Matter* 3 (1991) 1265.
- [26] M. Thoms, H. von Seggern and A. Winnacker, *J. Appl. Phys.* 76 (1994) 1800.
- [27] S. Chernov and V. Gavrilov, *Radiat. Eff. Def. Solids* 135 (1995) 333.