

# Dependence of Scintillation Characteristics in the CsI(Tl) Crystal on $Tl^+$ Concentrations Under Electron and Alpha Particles Excitations

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**Abstract**—This paper reports the emission spectra, pulse heights, and decay curves of CsI(Tl) crystals with a  $Tl^+$  concentration ranging from  $10^{-6}$  to  $10^{-2}$  mol under electron and alpha particle excitations. Larger pulse heights from the crystals with  $Tl^+$  concentration of more than  $10^{-3}$  mol were observed for a higher ionization density of alpha particle excitation compared with those of electron excitation. This enhancement may be explained in terms of the higher scintillation efficiency for higher  $Tl^+$  concentration. Decay curves under electron and alpha particle excitations were compared with the decay curve under pulsed UV excitation. The observed decay curves were explained in terms of the sum of the three energy transfer processes to create excited states ( $Tl^+$ )\*: the prompt process, the  $V_k$  diffusion process to  $Tl^0$ , and the electron release process from  $Tl^0$  to  $Tl^{++}$ . Here,  $Tl^0$  and  $Tl^{++}$  were produced through  $(Tl^+ + \text{electron}) \rightarrow Tl^0$  and  $[Tl^+ + \text{hole (or } V_k)] \rightarrow Tl^{++}$ .

**Index Terms**—Cesium iodide, crystals, delay time, scintillation.

## I. INTRODUCTION

SOME works on the basic physical behavior of the scintillation in CsI (Tl) have been summarized by Murray and Meyer [1] and Gwin and Murray [2], while general features of scintillation processes in inorganic crystals were described by Birks in 1964 [3]. More recently, Derenzo and Weber have modeled the scintillation process of CsI(Tl) and CsI(Na) using the first principles of the quantum chemistry cluster calculation and high-performance computers [4]. However, there are not so many studies on the scintillation process in CsI(Tl) and CsI(Na) as on NaI(Tl), and they are still under discussion [4]–[6].

On the other hand, in recent years a considerable number of papers have been published using CsI(Tl) scintillators as a particle identification detector for heavy ionizing particles using a pulse shape difference for several ionization densities. The use of the inorganic scintillator for identification of high light particles had been overcome by the solid-state detectors. However, there has been revival of interest, particularly in the field of heavy ion physics [7].

The large number of detectors used in these experiments, essential to discriminate the light particles, requires the

use of a simple and rather cheap detection system. CsI(Tl) scintillators have been chosen to be the basic scintillators in several detection devices. The  $Tl^+$  concentration of the CsI(Tl) scintillator reported in references for particle identification has been usually available commercially and is nominally around  $10^{-3}$  mol [7]–[9]. However, this concentration was determined considering the maximum pulse height for gamma-ray excitation nearly half a century ago [3], and this concentration may be different for particles with different ionization densities other than electrons.

To obtain a better performance of the radiation detector, more studies on the dependence of the Tl concentration ( $[Tl^+]$ ) in the scintillation characteristics are necessary. Furthermore, the comprehension of the scintillation mechanism is required. The effect of  $[Tl^+]$  ranging from  $6 \times 10^{-5}$  to  $3 \times 10^{-2}$  mol on the scintillation characteristics has been studied by Schotonaus *et al.* [10]. In this paper, we report the measurement results for emission spectra, pulse heights, and decay curves of the CsI(Tl), doped with different  $[Tl^+]$ , under electron and alpha particles excitations. The  $[Tl^+]$ , varied from  $10^{-6}$  to  $10^{-2}$  mol. Measured decay curves were compared with those of pulsed UV excitation.

## II. EXPERIMENTAL METHODS

CsI crystals with  $[Tl^+]$  of  $10^{-6}$ ,  $10^{-5}$ ,  $10^{-4}$ ,  $10^{-3}$  and  $10^{-2}$  mol used in this work were grown in accordance with the vertical Bridgman method using a quartz crucible in vacuum atmosphere. The starting procedure used with a purity of 99.99% was obtained from Metal Gesellschaft K. K., Germany. Before starting the crystal growth, the powder was purified by evacuating and heating in the quartz crucible at about 150 °C, for 2 h, to remove mainly residual water. After the purification process, the quartz crucible was filled with argon gas and sealed. Crystals around 100 mm long were obtained with a growth rate of 2 mm/h.

The  $[Tl^+]$  was nominal, referring to the  $[Tl^+]$  before the crystal growth. The CsI crystals with  $[Tl^+]$  of  $10^{-6}$ ,  $10^{-5}$ ,  $10^{-4}$ ,  $10^{-3}$  and  $10^{-2}$  mol were cut into three pieces of about 30 mm long, obtaining samples of the first, middle, and last part of the crystal.

In this work, only the middle parts of the crystals were used. A typical sample size of about 20 mm diameter and 30 mm length was used for measurements of the scintillation emission, efficiency, and decay curves. As an auxiliary experiment, the variation of the  $[Tl^+]$  along these samples was measured. The

Manuscript received October 26, 2000; revised February 16, 2001. This work was supported by FAPESP-SP.

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Publisher Item Identifier S 0018-9499(01)06982-9.

[Tl<sup>+</sup>] was determined using an Atomic Absorption Spectrometer (AA-1475, Varian). The variation of the [Tl<sup>+</sup>] was found to be less than 10%.

The emission spectra of these crystals were measured with a monochromator (JASCL FP55A) having a wavelength resolution of 20 nm. For the electron excitation of the crystals, 511-keV annihilation gamma-rays from a <sup>22</sup>Na source were used. Furthermore, 5.4-MeV alpha particles from an <sup>241</sup>Am source were also used to excite the crystal near surface. The signal from the monochromator was detected with a UV-sensitive quartz photomultiplier (Hamamatsu Photonics R 1668). The spectral response of the detection system was not corrected.

The scintillation pulse heights were measured using a glass window RCA8875 photomultiplier. The output signals from the photomultiplier were shaped with an RC network, having an RC clipping time constant of about 5  $\mu$ s.

Measurements of the decay curves were carried out under excitation of alpha particles from an <sup>241</sup>Am source and electrons produced by 662-keV gamma-rays from a <sup>137</sup>Cs source. The scintillation photons were detected using a quartz window photomultiplier (Hamamatsu Photonics R3235). The output signal from the plate of the photomultiplier was shaped with a resistance capacitance (RC) network having RC clipping time constant of about 5 ns and fed directly to a Tektronix 784A oscilloscope. The decay data were decomposed and analyzed with the least squares fitting technique.

To study the scintillation mechanism, the decay curves under electrons and alpha particles excitations were compared with the decay curve under the pulsed UV excitation. The pulsed UV excitation was performed using a nanosecond flash lamp system (LS-100, Photon Technology International, Inc). This flash lamp used nitrogen as the plasma gas, and the excitation pulse had a duration of about 3 ns, with an after-pulse as shown in Fig. 3. The width of the after pulse was about 4000 ns. The wide range of excitation wavelength (294–407 nm) of this N<sub>2</sub> flash lamp made it possible to excite the scintillators at various UV wavelengths. Monochromators were used to select the excitation and emission wavelengths. The bandpass was 2 nm for the excitation measurements and 8 nm for the emission measurements. The emitted light was detected at a right angle to the direction of the incident light. Decay was measured at room temperature.

### III. RESULTS AND DISCUSSION

#### A. Emission Spectra

Fig. 1 shows the emission spectra of the CsI crystals with [Tl<sup>+</sup>] from 10<sup>-6</sup> to 10<sup>-2</sup> mol, measured under the excitations of electrons (a) generated by 511-keV annihilation gamma-rays from a <sup>22</sup>Na source and 5.4-MeV alpha particles and (b) from an <sup>241</sup>Am source. Similar emission spectra for the [Tl<sup>+</sup>] ranging from 6  $\times$  10<sup>-5</sup> to 3  $\times$  10<sup>-2</sup> mol are reported in [10].

It is clearly seen from Fig. 1(a) that the intensity of the 305–320 nm band, which corresponds to the intrinsic luminescence from pure CsI, decreases with increasing [Tl<sup>+</sup>]. The intensity of the 480-nm band, originated from Tl<sup>+</sup>, increases with increasing [Tl<sup>+</sup>]. The intensity of the 400-nm band,

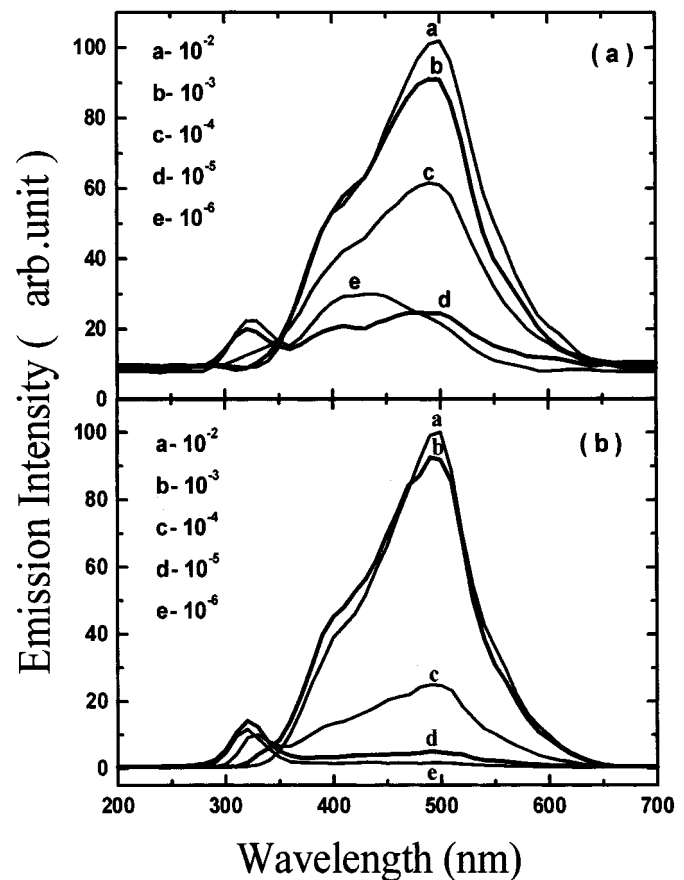


Fig. 1. Measured emission spectra of the CsI(Tl) crystal with [Tl<sup>+</sup>] of 10<sup>-6</sup>, 10<sup>-5</sup>, 10<sup>-4</sup>, 10<sup>-3</sup>, and 10<sup>-2</sup> mol under (a) gamma-rays and (b) alpha particles excitations.

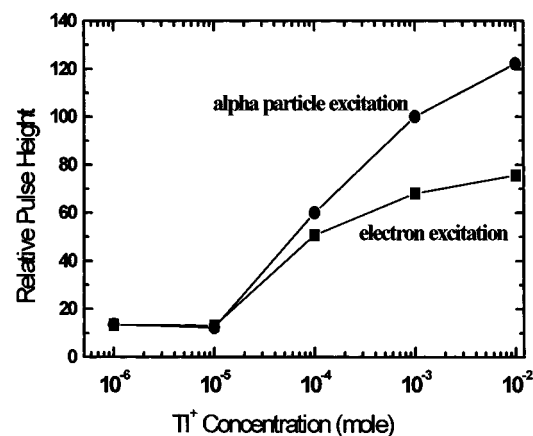


Fig. 2. Relative pulse height against [Tl<sup>+</sup>] under electron and alpha particle excitations. Pulse heights were normalized at the lowest [Tl<sup>+</sup>] = 10<sup>-6</sup> mol.

originated from crystal defects, is increased for the lowest Tl<sup>+</sup> concentration of 10<sup>-6</sup> mol for electron excitation.

#### B. Scintillation Efficiency

Fig. 2 shows the relative pulse height as a function of [Tl<sup>+</sup>] under electron and alpha particle excitations. The pulse height increases with increasing [Tl<sup>+</sup>] for both excitations. The larger pulse heights of [Tl<sup>+</sup>], more than 10<sup>-3</sup> mol, are observed for

higher ionization density of alpha particles excitation compared with those of electron excitation. The scintillation efficiency was showed to be dependent on  $[Tl^+]$ . These results are in agreement with the behavior of the  $[Tl^+]$  ranging from  $6 \times 10^{-5}$  to  $3 \times 10^{-2}$  mol in [10] and also with the work in [11], [16], and [12], but in sharp contrast to the results obtained by Gwin and Murray [2]. Therefore, the increase in scintillation efficiency is expected for high  $dE/dx$  particles in larger  $[Tl^+]$ , providing the optical transmission of the samples does not decrease.

Murray and Meyer proposed a phenomenological model of the scintillation process by taking into account the saturation process by taking into account the saturation concept of the available activator centers [1]. This mode predicts that the scintillation efficiency depends on  $[Tl^+]$ . Murray and Meyer [1] estimated the diffusion length of the energy carriers to be about  $20 \times 10^{-8}$  cm, which is at best a rough value. It should be significant, however, as to order of magnitude; in particular, the diffusion length is quite small and indicates that the energy transport occurs in a very limited region. Here, it is noted that the number of  $Tl^+$  activators for  $[Tl^+]$  of  $10^{-3}$  mol is apparently lower for electron excitation than that of excited states for alpha particle excitation. Thus, all possible activator sites have been filled; i.e., a depletion of available activator sites by a particle of high specific-energy loss,  $dE/dx$ . Therefore, the increase in scintillation efficiency is expected for high  $dE/dx$  particles for larger  $[Tl^+]$ .

We believe that the saturation mechanism of available activator centers is partly responsible for the decrease in scintillation efficiency at the high  $dE/dx$  region together with the concept of ionization quenching and a nonradiative process for the excited states  $(Tl^+)^*$  in the high  $dE/dx$  regions described in Section III-B. Recently, Kubota *et al.* [13] published a paper on the test of scintillation models for thallium-activated alkali halides, proposed by Birks [3], [14] and Murray and Meyer [1].

### C. Scintillation Decay Curves

Fig. 3 shows the measured decay curves of the crystals with different  $[Tl^+]$  ranging from  $10^{-6}$  to  $10^{-2}$  mol under electrons and alpha particle excitations.

A rising part was observed in its decay curves under electron excitation. The rising time decreased with increasing  $[Tl^+]$ . This is in disagreement with the data in [10], where the rising time was independent of the  $[Tl^+]$ . On the other hand, after the peak intensity, the light intensity decreased to 1/10 of the peak intensity within  $5 \mu s$ , and its decay feature does not strongly depend on  $[Tl^+]$ . This feature is in agreement with the results of [10].

For alpha-particle excitation, the decay feature of the crystals with  $[Tl^+]$  of less than  $10^{-4}$  mol is similar to those of electron excitation. However, for  $[Tl^+]$  of  $10^{-3}$  and  $10^{-2}$  mol, the decay curves lack the rising part and show a faster decay compared with those of electron excitation for the time range of less than 1000 ns. Those two decay curves in the time range from 200 to about 1500 ns show a similar decay feature to that of UV excitation, as shown in Fig. 3. However, it is noted that after about 2000 ns, the decay feature of those two decay curves is similar to those of electron excitation. A typical decay curve of the crystal with  $[Tl^+]$  of  $10^{-3}$  mol under pulsed UV excitation is

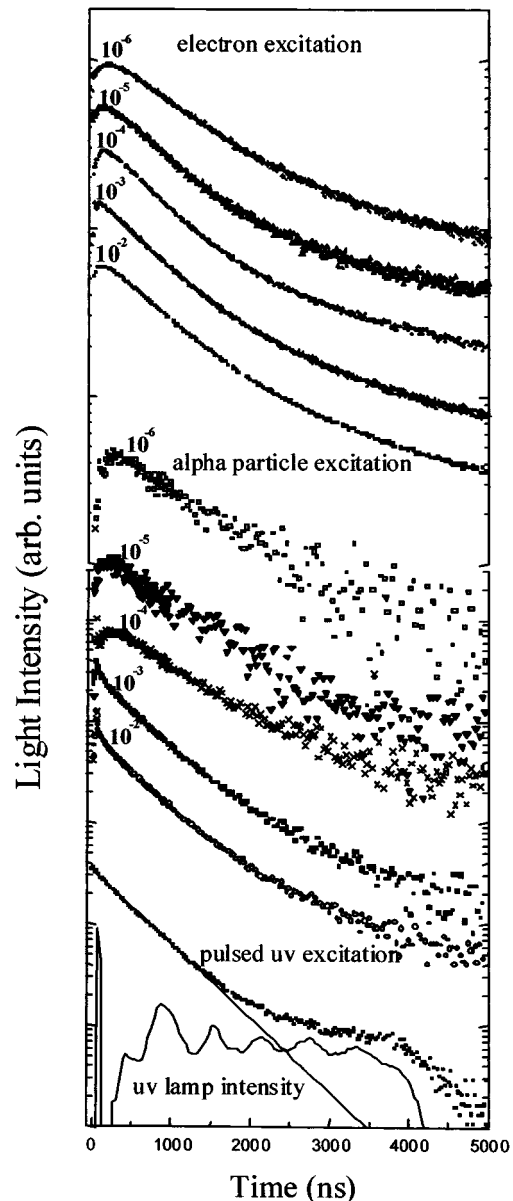


Fig. 3. Decay curves of the CsI crystals with  $10^{-6}$ ,  $10^{-5}$ ,  $10^{-4}$ ,  $10^{-3}$ , and  $10^{-2}$  mol under electron and alpha particle excitations. The decay curve of the CsI crystal with  $10^{-3}$  mol under pulsed UV excitation is also shown together with the time profile of the pulsed UV lamp. The solid line represents the fitted decay curve for an exponential function for the time range up to 1500 ns.

shown in Fig. 3, together with a time profile of the UV excitation pulse. As shown in this figure, the decay curve under UV excitation can be well fitted by an exponential function with a decay time of  $575 \pm 5$  ns for the time range to 1500 ns. A deviation of the data points from the fitted line after 1500 ns is explained in terms of the effect due to the excitation under the after pulse, which persists to 4000 ns. All crystals under pulsed UV excitation presented similar decay curves. Table I summarizes the fitted decay times. We believe that this decay time corresponds to that of excited state  $(Tl^+)^*$ .

### D. Energy Transfer Processes

It is well known that the excitation of CsI(Tl) at room temperature by ionizing radiation results in emission that is char-

TABLE I  
 FITTED DECAY TIMES  $\tau_V$ ,  $\tau_r$ , AND  $\tau$  FOR CsI(Tl) CRYSTALS WITH  $[Tl^+]$  OF  $10^{-2}$ ,  $10^{-3}$ ,  $10^{-4}$ ,  $10^{-5}$ , AND  $10^{-6}$  mol AND THE LIGHT INTENSITY RATIOS FOR ELECTRON, ALPHA PARTICLE AND PULSED UV EXCITATIONS. THE DECAY TIMES  $\tau$  WERE FIXED AS BEING THE SAME VALUES OBTAINED FROM FITTING FOR THE PULSED UV EXCITATION

Tl <sup>+</sup> Concentration (mol)	Fitted Decay Time (ns)		Intensity Ratio (%)	
<b>Electron Excitation</b>				
	$\tau_V$	$\tau_r$		
$10^{-6}$	140 ± 10	2100 ± 140	$I_i:I_{ii}:I_{iii} = 23:25:52$	
$10^{-5}$	140 ± 5	2700 ± 110	$I_i:I_{ii}:I_{iii} = 35:20:45$	
$10^{-4}$	100 ± 5	2800 ± 100	$I_i:I_{ii}:I_{iii} = 32:29:39$	
$10^{-3}$	80 ± 5	2400 ± 100	$I_i:I_{ii}:I_{iii} = 33:27:40$	
$10^{-2}$	70 ± 10	2000 ± 180	$I_i:I_{ii}:I_{iii} = 32:17:51$	
<b>Alpha particle Excitation</b>				
	$\tau_f$	$\tau_V$	$\tau_r$	
$10^{-6}$	—	130 ± 30	1900 ± 400	$I_i:I_{ii}:I_{iii} = 57:21:22$
$10^{-5}$	—	110 ± 10	2000 ± 200	$I_i:I_{ii}:I_{iii} = 45:22:33$
$10^{-4}$	—	120 ± 10	2000 ± 200	$I_i:I_{ii}:I_{iii} = 46:24:30$
$10^{-3}$	160 ± 50		2200 ± 700	$I_f:(I_i+I_{ii}):I_{iii}=4:78:18$
$10^{-2}$	130 ± 40		2300 ± 700	$I_f:(I_i+I_{ii}):I_{iii}=5:77:18$
<b>Pulsed uv Excitation</b>				
	$\tau$			
$10^{-6}$	580 ± 20			
$10^{-5}$	565 ± 5			
$10^{-4}$	570 ± 5			
$10^{-3}$	585 ± 5			
$10^{-2}$	580 ± 5			

acteristic of the Tl<sup>+</sup> ion. Since the Tl concentration in a typical CsI(Tl) scintillator is about  $10^{-3}$  mol, it is clear that the energy of the incident charged particles is predominantly deposited in the host crystal lattice. Therefore, some of the energy initially deposited in the host crystal must be transferred to Tl<sup>+</sup> ions to create excited states (Tl<sup>+</sup>)<sup>\*</sup>. In this paper, we consider three energy-transfer processes that convert the Tl<sup>+</sup> ions to (Tl<sup>+</sup>)<sup>\*</sup>, following the same considerations used for NaI(Tl) by Kubota *et al.* [15].

- 1) The first process is the prompt creation of an excited state (Tl<sup>+</sup>)<sup>\*</sup> by some mechanisms, including nonthermal trapping of holes and electrons at a Tl<sup>+</sup>, and exciton capture. The scintillation decay time is determined by the lifetime  $\tau$  of (Tl<sup>+</sup>)<sup>\*</sup>.
- 2) The second process is the prompt capture of electron to form Tl<sup>0</sup>, followed by diffusion of self-trapped holes ( $V_k$  centers) at Tl<sup>0</sup> to form (Tl<sup>+</sup>)<sup>\*</sup>. The scintillation decay will be composed of the  $V_k$  diffusion time  $\tau_V$  and  $\tau$ .
- 3) The third process is the prompt capture of electron to form Tl<sup>0</sup>, followed by diffusion of  $V_k$  center to some Tl<sup>+</sup> ion and capture to form Tl<sup>++</sup>. Then at a later time, electrons are thermally excited and released from the Tl<sup>0</sup> traps and subsequently retrapped or captured at Tl<sup>++</sup> to form (Tl<sup>+</sup>)<sup>\*</sup>. The scintillation decay time will be composed of  $\tau$  and the lifetime  $\tau_r$  for thermal release of electron from Tl<sup>0</sup>.

Process 1) simply gives rise to an exponentially decaying luminescence intensity with lifetime of  $\tau$

$$I_i(t)\alpha(Tl^+)^* = [(Tl^+)_0^*/\tau] \exp(-t/\tau). \quad (1)$$

We consider the creation of (Tl<sup>+</sup>)<sup>\*</sup> by process 2). The equation describing process 2) is

$$d(Tl^+)^*/dt = -[d(Tl^+)^*/\tau] + dV_k(t)/dt \quad (2)$$

where  $V_k(t)$  is the  $V_k$  concentration as a function of time.

The  $V_k$  diffusion time is defined as the time necessary to reach 2/3 of the maximum Tl<sup>++</sup> concentration [15]. Here, we consider for practicality  $V_k(t)$  as an exponential function with the  $V_k$  diffusion time of  $\tau_V$ . Then we have

$$I_{ii}(t)\alpha(Tl^+)^* = (Tl^{++})_0 [\exp(-t/\tau) - \exp(-t/\tau_V)] / (\tau - \tau_V). \quad (3)$$

Due to the difference observed in decay times of a CsI(Tl) crystal under electron excitation and pulsed UV excitation, the electron release process 3) was also introduced.

Combining processes 1), 2), and 3), we have

$$I(t) = I_i(t) + I_{ii}(t) + I_{iii}(t) \quad (4)$$

where

$$I_i(t) + I_{ii}(t) = A \exp(-t/\tau) - B \exp(-t/\tau_\nu)$$

and

$$I_{iii}(t) = C[\exp(-t/\tau_r) - \exp(-t/\tau)].$$

Here

$$A = [(TI^+)_0^*/\tau] + [(TI^{++})_0/(\tau - \tau_\nu)]$$

$$B = (TI^{++})_0/(\tau - \tau_\nu) \text{ and } C = (TI^0)_0/(\tau_r - \tau).$$

The measured decay curves with the rising part were fitted to (4). In this fitting,  $\tau$  in (4) is fixed to the fitted values  $\tau$  obtained under pulsed UV excitation. Table I summarizes the fitted  $V_k$  diffusion time  $\tau_\nu$ , electron release time  $\tau_r$ , and the light intensity ratio of the  $i$ th component. It is to be noted here that the fitted  $V_k$  diffusion time  $\tau_\nu$  slightly depends on  $[TI^+]$ ; i.e.,  $\tau_\nu$  decreases with increasing  $[TI^+]$ . This result is in contrast to the case of NaI(Tl), where  $\tau_\nu$  does not strongly depend on  $[TI^+]$  ranging from  $10^{-4}$  mol to  $2.2 \times 10^{-3}$  mol, i.e.,  $50 \pm 5$  ns. The electron release time is found to be  $2000 \sim 3000$  ns, longer than that of  $390 \pm 40$  ns in NaI(Tl).

The decay curves without the rising part under alpha particle excitation of the crystals with  $[TI^+]$  of  $10^{-3}$  and  $10^{-2}$  mol are fitted to the form

$$\sum_{i=1}^3 A_i \exp(-t/\tau_i) \quad (5)$$

where  $A_i$  and  $\tau_i$  are the amplitude and the decay time of the  $i$ th component. Table I summarizes the fitted  $\tau_i$  of the  $i$ th component.

Due to the high density excitation of alpha particles in a CsI crystal, it is supposed that prompt process 1) should dominate in the scintillation. We believe that the dominant second decay component is due to the emission from prompt process 1). Thus, in this fitting,  $\tau_2$  in (5) is fixed to the fitted values  $\tau$  obtained under pulsed UV excitation.

A faster decay component of  $\tau_1 = \tau_f = 130 \sim 160$  ns is observed in addition to the dominant second component. This indicates the presence of some nonradiative processes of  $(TI^+)^*$  due to interaction with excited atoms under high-specific-energy loss  $dE/dx$ . This nonradiative process may be partly responsible for the decrease in the scintillation efficiency for high  $dE/dx$ . Since the fitted  $\tau_3$  is the same order of the  $\tau_r$  obtained, we interpreted that this third component is due to the emission from electron release process 3).

Fig. 4 shows the typical results of decay curves and their fitted curves of CsI crystals with  $[TI^+]$  of  $10^{-3}$  mol (no rising part) and  $10^{-5}$  mol (rising part) under electrons excitations compared to that of alpha particle and pulsed UV excitations. A striking difference between the decay curves for electron excitation and those for alpha and pulsed UV excitations is observed for  $[TI^+]$  of more than  $10^{-3}$  mol, while for  $[TI^+]$  of less than  $10^{-4}$  mol, the decay feature is similar for electron and alpha excitations.

This difference is explained by taking into account the relative importance of the scintillation photons emitted from prompt

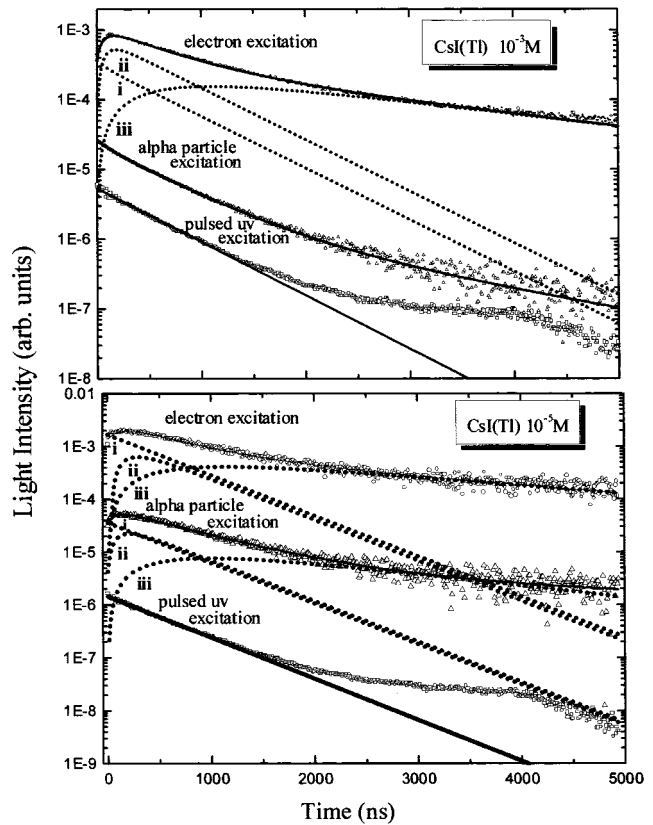


Fig. 4. Decay curves and their fitted curves of CsI crystals with  $[TI^+]$  of  $10^{-3}$  mol (no rising part) and  $10^{-5}$  mol (rising part) under electrons excitation compared with those of alpha particle and pulsed UV excitations.

process 1) and  $V_k$  diffusion process 2). Similar results were found for NaI(Tl) crystals by Kubota *et al.* [15].

#### IV. CONCLUSION

From experiments on the emission spectra, pulse heights, and decay curves from CsI(Tl) crystals with different Tl concentrations under electron and alpha particle and pulsed UV excitations, the following conclusions were drawn.

- 1) The higher pulse height is obtained by increasing  $[TI^+]$  for particles of high ionization density.
- 2) The lifetime of  $(TI^+)^*$  is measured to be  $575 \pm 5$  ns by using pulsed UV excitation.
- 3) The rising part of the decay curve from CsI(Tl) crystals under electron excitation and from lower  $[TI^+]$  crystals under alpha excitation is interpreted in terms of the sum of scintillation photons from prompt process 1) and  $V_k$  diffusion process 2).
- 4) The decay curve in CsI(Tl) crystals under electron excitation is composed of the two components with the decay times of  $\tau = 575 \pm 5$  ns and  $\tau_r = 2000 \sim 3000$  ns. The former component is due to the sum of scintillation photons from prompt processes 1) and the  $V_k$  diffusion process 2), and the latter is due to electron release process 3).
- 5) The faster decay component with the decay time of  $\tau_f = 130 \sim 160$  ns is observed in crystals with high  $[TI^+]$  under alpha particle excitation in addition to the dominant

scintillation component with decay time of  $575 \pm 5$  ns. This component is interpreted in term of the emission from some nonradiative processes of  $(\text{Tl}^+)^*$  due to the interaction with excited atoms under a high-specific-energy loss,  $dE/dx$ .

Further experimental data of the light output and optical transmission in large crystals with high  $[\text{Tl}^+]$ , on the order of  $10^{-2}$  mol, are recommended to develop detectors for heavy ion physics.

#### ACKNOWLEDGMENT

F. E. Costa would like to thank Rikkyo University for the research fellowship and facilities offered.

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