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## Charge transfer processes in CsI:Tl using near-UV light

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### ABSTRACT

This paper studies charge transfer processes in CsI:Tl crystals by analyzing the bulk photo-conductivity spectra, the temperature behavior of the bulk photo-conductivity current and the shape and intensity of the activator emission pulse excited by an electron pulse beam and/or laser pulse emission at temperatures between 80 and 400 K. The Tl concentration in CsI:Tl crystals varies from  $10^{-3}$ – $10^{-1}$  mass%. It has been determined that near-UV light induces a bulk conductivity in CsI:Tl crystals only when the Tl concentration is greater than  $3 \times 10^{-3}$  mass%. A mechanism is proposed to explain the charge transfer processes with photons whose energy is approximately half the width of the CsI band gap. Near-UV light causes charge transfer from  $I^-$  to  $Tl^+$  ions, forming  $Tl^0$  centers in the  $6p^2P_{1/2}$  ground and  $6p^2P_{3/2}$  excited states. The electron, assisted by phonons, leaves the  $Tl^0$  center from either the  $6p^2P_{1/2}$  or  $6p^2P_{3/2}$  states and overcomes the 0.13 or 0.30 eV energy barrier, respectively, and subsequently populates the activator conduction sub-bands, which are found inside the band gap of CsI:Tl. The formation of activator sub-bands is possible only above the threshold Tl concentration, i.e., above  $3 \times 10^{-3}$  mass%.

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### 1. Introduction

It is well known that CsI:Tl crystals exposed to daylight at room temperature emit visible light at approximately 2.3 eV [1,2]. However, CsI:Tl crystals exposed to near-UV light (3.0–3.7 eV) produce a persistent afterglow that temporarily blocks or distorts the scintillation response. Should the crystal contain either carbonate or borate impurities, it becomes colored under this light [3,4]. To eliminate the (persistent) afterglow, the crystal has to remain in a dark place for some time, and it has to be thermally treated at  $T > 340$  K to eliminate the color centers. In spite of the extensive studies of CsI:Tl luminescence excited in activator absorption bands [5–10], it is not clear how the photons, which are unable to excite electron transitions even in the lowest energy absorption band (A-band) of  $Tl^+$  centers, give rise to the mobile charge carriers. Indeed, previous studies [5–11] explained the 2.3 eV luminescence using the radiative annihilation of  $Tl^+$  perturbed excitons, but the charge transfer processes under near-UV light responsible for the 2.3 eV luminescence do not fit the framework of the exciton model for  $Tl^+$  center emission. Earlier, in our paper [12], we suggested a model based on which the 2.3 eV emission arises due to a  $Tl^0$ - $V_k$  tunnel recombination process that does not give rise to  $Tl^+$  perturbed excitons. We believe that the model can

explain the formation of radiative states for  $Tl^+$  centers at band-to-band ( $E > 6.0$  eV), for the  $Tl^+$  absorption band ( $3.7$  eV  $< E_{ex} < 5.3$  eV) and for near-UV light ( $3.0 < E < 3.7$  eV) excitations.

The aim of this paper is to investigate the charge transfer processes in CsI:Tl crystals and explain why photons with a comparatively low energy are able to delocalize the charge carriers.

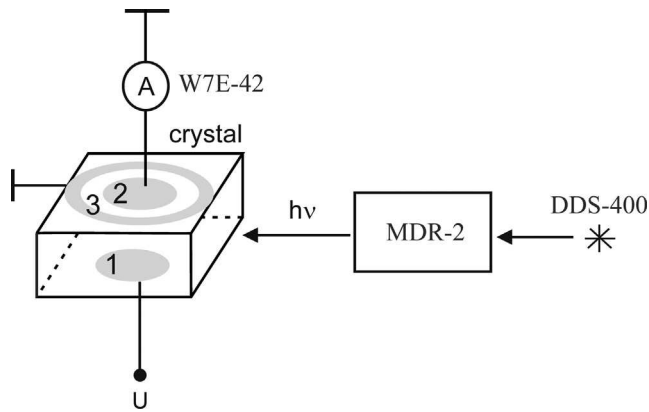
### 2. Experiment

CsI:Tl bulks were grown using the Stockbarger technique in evacuated quartz ampoules. Samples with dimensions of  $10 \times 8 \times 2$  mm were cut from the bulks. The Tl concentration in the samples was chemically determined and varied from  $2 \times 10^{-5}$  to  $2 \times 10^{-1}$  mass%.

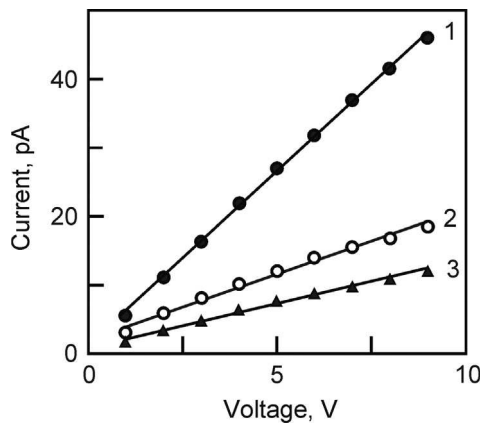
Photoluminescence was excited in the samples by  $N_2$  laser pulses ( $\lambda = 337.1$  nm;  $t_{1/2} = 5$  ns;  $Q = 1$  mJ) and measured with an optical spectrometer consisting of an MDR-3 monochromator, an FEU-106 photomultiplier and a GDS-2204 digital oscilloscope with a time resolution of 7 ns. A bulk photocurrent was induced in the samples using the light of a DDS-400 deuterium lamp through an MDR-2 monochromator and subsequently measured with a W7E-42 ammeter (Fig. 1). The samples were screened against surface currents. The measurements were carried out at temperatures in the range of 78–400 K. A two-cascade excitation technique employing

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**Fig. 1.** Scheme of an experimental setup for measuring photoconductivity excitation spectra. 1,2 - electrodes, 3-screen.



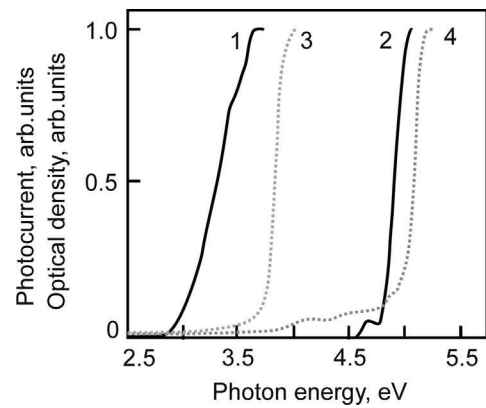
**Fig. 2.** Volt-ampere characteristics for CsI:4 × 10<sup>-2</sup>%Tl under a mercury arc without a filter (1), with a FS-7 filter ( $\Delta\lambda=320\text{--}450\text{ nm}$ ) (2), and with a BS-7 filter ( $\lambda > 350\text{ nm}$ ) (3).

an electron pulse ( $E_e=0.25\text{ MeV}$ ,  $t_{1/2}=15\text{ ns}$ ,  $W=0.1\text{ J/cm}^2$ ) followed, after a  $9\text{ }\mu\text{s}$  delay, by a laser pulse ( $\text{YAlO}_3\text{-Nd}^{3+}$ ,  $\lambda=1080\text{ nm}$ ,  $t_{1/2}=30\text{ ns}$ ) was also used in studying the charge transfer processes.

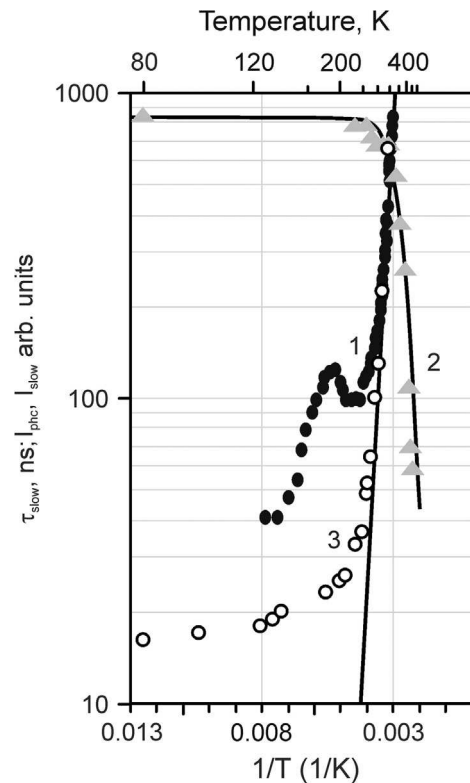
### 3. Results and discussion

#### 3.1. Photo-induced conductivity

We experimentally determined that UV light with a constant intensity induces a bulk photocurrent with a linear volt-ampere characteristic in CsI:Tl crystals (Fig. 2), which means that CsI:Tl crystals have an intrinsic photoeffect. A CsI:4 × 10<sup>-2</sup>% Tl sample becomes photoconductive at photon excitation energies in the range of 3.0–3.7 eV, whereas CsI:4 × 10<sup>-4</sup>% Tl becomes photoconductive in the range of 4.8–5.1 eV (Fig. 3, curves 1,3). Note that the energies of 3.7 and 5.1 eV are points from which the A-absorption band and the band-to-band absorption, respectively, begin to increase. Both samples abruptly lose their photoconductivity at photon excitation energies greater than 3.7 and 5.1 eV, respectively, because the photons cannot enter the region between electrodes 1 and 2 (Fig. 1) to induce the photocurrent because of a dramatic increase in the sample's optical density. If the widths of both samples are equal the absorption depth of the high-energy photons, their photoconductivity will not be lost beyond 3.7 and 5.1 eV, respectively. A comparison of the photocurrent excitation spectra of the CsI:Tl samples with different Tl concentrations ranging from 2 × 10<sup>-5</sup>% to 2 × 10<sup>-1</sup>% shows that photons with



**Fig. 3.** Photoconductivity excitation spectra (1,2) and optical absorption spectra (3,4) for CsI:4 × 10<sup>-2</sup>%Tl (1,3) and CsI:4 × 10<sup>-4</sup>%Tl (2,4).



**Fig. 4.** Photocurrent (1), decay constant (2) and photo-luminescence intensity at 2.3 eV (3) vs. reciprocal temperature for CsI:4 × 10<sup>-2</sup>%Tl at  $\lambda_{\text{exc}}=337.1\text{ nm}$ .

energies of approximately half of the band-gap width induce a photocurrent in CsI:Tl crystals only when the Tl concentration exceeds its 3 × 10<sup>-3</sup>% threshold. These experimental results suggest that there must be an impurity conduction sub-band in the band gap of the CsI:Tl crystal-dielectric. Because this impurity sub-band is caused by the activator, it will be hereinafter referred to as the activator conduction sub-band.

As shown in Fig. 4, the photocurrent in CsI:4 × 10<sup>-2</sup>%Tl crystals increases erratically with temperature. The photocurrent increases for temperatures up to 140 K, decreases slightly at approximately 200 K and subsequently increases again with temperature. We believe that the increasing photocurrent at low temperature (up to 140 K) is due to the thermal release of electrons from the Tl<sup>0</sup> centers. Other evidence of such a release includes the thermal glow peak at temperatures between 120 and 130 K [10,13–15] and

a post-irradiation increase of the scintillation pulse with a thermal activation energy of 0.13 eV [16]. The photocurrent decrease at 200 K can be explained as follows: anionic vacancies, being mobile at temperatures over 200 K, are captured by  $Tl^0$  centers, giving rise to  $Tl^0V_a^+$  color centers (deep traps for electrons [13]), which are responsible for the reduction of the photocurrent. Over 295 K, the temperature dependence of the photocurrent  $I_{phc}(T)$  becomes linear in Arrhenius coordinates. The thermal activation energy estimated by the slope of  $I_{phc}(T)$  amounts to  $E_a = 0.32 \pm 0.01$  eV. The origin of the increase in the photocurrent at temperatures over 295 K (Fig. 4a) will be explained below.

### 3.2. Photo-induced luminescence

#### 3.2.1. Luminescence under excitation by near-UV light

A luminescence spectrum excited by laser pulses at 3.68 eV ( $\lambda = 337.1$  nm) is shown in Fig. 5a. Ranging from 3.0 to 1.6 eV, this spectrum has the same shape as that of the scintillations caused by the recombination of  $\{Tl^0, V_k\}$  pairs [17]. The decay kinetics of the photoluminescence pulse at temperatures in the range of 78–280 K have two exponential stages: fast ( $\tau_{fast} \leq 10$  ns) and slow ( $\tau_{slow} > 600$  ns). The fast stage is not observed at  $T \geq 295$  K. Fig. 5b shows an oscillogram of the photoluminescence pulse at 295 K using a semi-log scale. As shown in Fig. 4b, the constant exponential decay parameter  $\tau_{slow}$  decreases with temperature, whereas the photoluminescence pulse intensity  $I_{slow}$  increases with temperature, demonstrating a strong correlation with temperature in the range of 295–400 K.

We now examine the spectral kinetic properties of the photoluminescence. The authors of [18] and [19] investigated the excitation spectra of the X-ray-colored crystals KCl(Tl) and RbI(Tl), respectively, and came to the conclusion that  $Tl^+$  center luminescence is excited in the  $Tl^0$  center absorption band as a result of the  $6^2P_{1/2} \rightarrow 6^2P_{3/2}$  transition. The diagram of the levels of

isolated Tl atoms and  $Tl^0$  centers proposed by [18] is shown in Fig. 6. In an isolated Tl atom, the transition  $6p^2P_{1/2} \rightarrow 6p^2P_{3/2}$  is forbidden, whereas the transitions  $6^2P_{1/2} \rightarrow 7s^2S$  and  $6^2P_{3/2} \rightarrow 7s^2S$  are allowed.  $Tl^0$  centers can be in the  $\varphi$ ,  $\psi$ ,  $\chi$  and  $\Sigma$  states. States such as  $\varphi$ ,  $\psi$ , and  $\chi$  mainly correspond to the 6p state, whereas  $\Sigma$  corresponds to the 7s state. Although the  $6^2P_{1/2} \rightarrow 6^2P_{3/2}$  transition is forbidden in an isolated Tl atom, it is partly allowed in  $Tl^0$  centers because of the mixing of  $\varphi$ ,  $\psi$  and  $\Sigma$  states resulting from the electron-phonon interaction. This is precisely the transition that causes the absorption band at 1.32 eV in RbI(Tl) [19]. According to the literature, the  $6^2P_{1/2} \rightarrow 6^2P_{3/2}$  transition produces a photocurrent in RbI(Tl) [19] and IR emissions in KCl(Tl) [18]. This emission peaks at 0.8 eV (1550 nm) with a decay constant of approximately 1.5  $\mu$ s, which slightly varies at temperatures in the range of 80–250 K. Because the spectral-kinetics properties of the  $Tl^0$  centers are almost unaffected by the lattice of any alkali halide crystals, the  $6^2P_{1/2} \rightarrow 6^2P_{3/2}$  transition in the  $Tl^0$  center is expected to produce the photocurrent and IR emission in CsI(Tl). A similar IR luminescence, peaking at approximately 0.77 eV (1600 nm), was measured by the authors of [20] when they exposed CsI(Tl) to proton irradiation at an energy of 4 MeV, which supports our idea. We measured the photocurrent in a CsI:  $4 \times 10^{-2}$ %Tl crystal exposed to photons with energies from 3.0 to 3.7 eV. Therefore, the photocurrent does not arise in the Cs conduction band because the band-to-band transition requires energies greater than 5.0 eV. We also experimentally demonstrated [12] that the  $6^2P_{1/2} \rightarrow 6^2P_{3/2}$  transition is responsible for the 1.33 eV absorption band in CsI(Tl) crystals exposed to electron pulses. Because  $Tl^0$  centers are short lived, we used the pulse cascade technique to excite  $6^2P_{1/2} \rightarrow 6^2P_{3/2}$  transitions in CsI(Tl).

#### 3.2.2. Luminescence under two cascade excitation

Fig. 7 shows the kinetic curves of the optical density in the  $Tl^0$  center band (Fig. 7a) and the luminescence at 2.3 eV (Fig. 7b) from when CsI(Tl) was irradiated in two cascades: first by an electron pulse ( $E_e = 0.25$  MeV,  $t_{1/2} = 15$  ns,  $W = 0.1$  J/cm<sup>2</sup>) and subsequently by a laser pulse (YAIO<sub>3</sub>-Nd<sup>3+</sup>,  $\lambda = 1080$  nm ( $E = 1.15$  eV),  $t_{1/2} = 30$  ns) after a 9  $\mu$ s delay. The insert in Fig. 7 shows a fragment of the short-lived absorption spectrum. Arrow 1 points to the maximum of the band that corresponds to the  $6^2P_{1/2} \rightarrow 6^2P_{3/2}$  transition, and arrow 2 points to the region where the laser pulse causes this transition. One can observe in Fig. 6a that the 1080 nm laser pulse causes an abrupt reduction in the optical density (Fig. 7a) and an abrupt increase in the luminescence intensity,

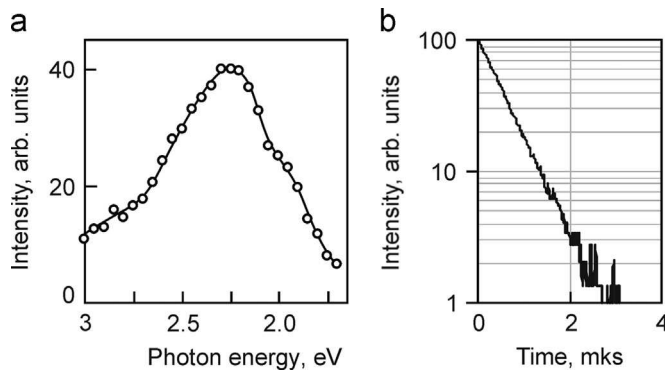


Fig. 5. Photoluminescence pulse spectrum (a) and photoluminescence pulse decay oscillogram (b) for CsI:  $4 \times 10^{-2}$  mass%Tl at 295 K.

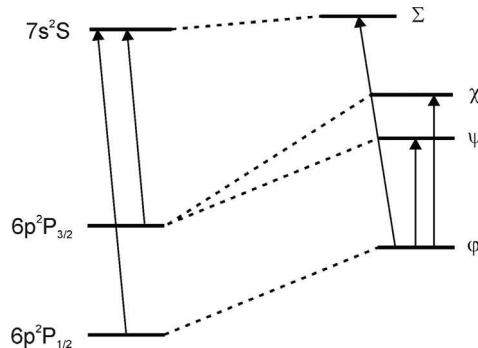


Fig. 6. Energy levels of Tl atoms (left) and  $Tl^0$  centers (right) [18].

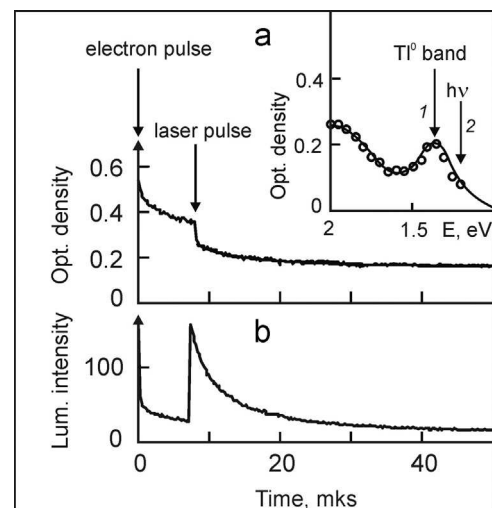


Fig. 7. Kinetic curves for the optical density at 1.33 eV (a) and the luminescence at 2.3 eV (b) under cascade pulse irradiation of CsI(Tl) at  $T = 80$  K.

which is approximately fourfold higher than the initial intensity of the cathodoluminescence (Fig. 7b).

According to [12], the cathodoluminescence at approximately 2.3 eV is caused by the radiative recombination of  $Tl^0$  and  $V_k$  centers, where  $V_k$  localizes in the nearest halide surroundings of  $Tl^0$ , forming a short interspace pair  $[Tl^0, V_k]$ . At 80 K, the electron pulse irradiation gives rise to short and long interspace pairs  $[Tl^0, V_k]$ , with the number of the former accounting for approximately 30% of the total  $Tl^0$  and  $V_k$  centers, having little influence on each other when they form long interspace pairs, can be considered as isolated centers. In such pairs,  $V_k$  can be found either in an area free from  $Tl^+$  or near  $Tl^+$  ions.  $V_k$  near  $Tl^+$  ions is denoted as either  $V_{kA}$  or  $[Tl^+V_k]$ . We believe that the sudden increase in the photoluminescence intensity at 2.3 eV was due to an immediate formation of a substantial number of short interspace pairs  $[Tl^0, V_k]$ . This can be explained as follows: when the crystal is exposed to the laser pulse, electrons escape from the  $Tl^0$  through the  $6^2P_{1/2} \rightarrow 6^2P_{3/2}$  transition and travel along an activator conduction sub-band until they are captured by  $[Tl^+V_k]$  centers, which change into short interspace pairs  $[Tl^0, V_k]$  responsible for the 2.3 eV emission. Thus, we believe that the  $\chi$  sub-level of the  $6^2P_{3/2}$  state of the  $Tl^0$  centers is responsible for the conduction state of CsI:Tl.

### 3.3. A model of photo-thermo-induced processes with charge transfer in CsI:Tl

The experimental results presented in the previous sections can be interpreted using the electron transition model shown in Fig. 8. The levels of the  $6^2P_{1/2}$  ground and  $6^2P_{3/2}$  excited states of the  $Tl^0$  center are denoted by  $\phi$ ,  $\psi$ , and  $\chi$ , respectively. Because the photocurrent in CsI:4 × 10<sup>-2</sup>%Tl is caused by photons with energies of approximately 3.0 eV, we believe that the activator conduction sub-band is found inside the band gap of the CsI crystal. The erratic behavior of the photocurrent in the CsI:Tl crystals is most likely due to  $Tl^0$  centers either in the  $6^2P_{1/2}$  or  $6^2P_{3/2}$  state from which an electron escapes into an activator sub-band by thermal assistance.  $Tl^0$  centers in the  $6^2P_{1/2}$  and  $6^2P_{3/2}$  states are responsible for the increase in photocurrent from 90 K to 140 K and over 295 K, respectively. As shown in Fig. 4, the temperature-dependence curves of the photocurrent and photoluminescence concur at temperature over 295 K, with the luminescence decay constant being inverse to them. We explain this behavior as follows: When the crystal is exposed to the near-UV light, an electron from the valence band occupies the  $\psi$  sub-level of the  $Tl^0$

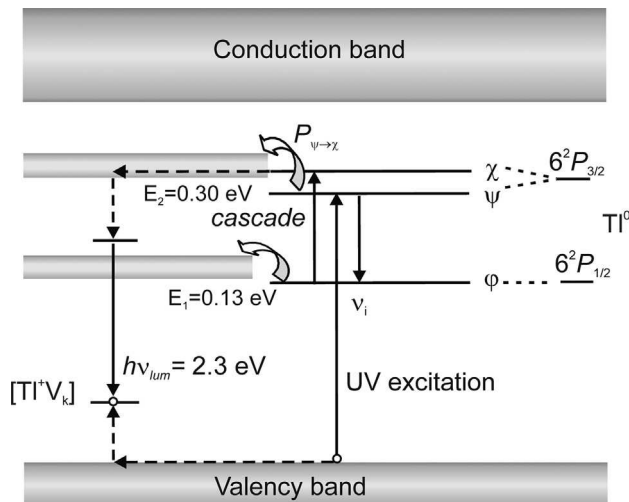


Fig. 8. Diagram of photo-induced electron transitions in CsI:Tl crystals.

**Table 1**  
Parameters of inter-level transitions for  $Tl^0$  centers in CsI:Tl crystals.

$\nu_i$	$\omega, c^{-1}$	$E_a, eV$
$1.2 \times 10^6$	$2.4 \times 10^{10}$	$0.3 \pm 0.01$

center in the  $6^2P_{3/2}$  state (“UV excitation”, straight arrow in Fig. 8), whereas a hole is captured by an  $Tl^+$  ion, forming a  $[Tl^+V_k]$  center. The probability of the spontaneous radiative transition of the electron from the  $\psi$  sub-level of the  $6^2P_{3/2}$  state to the  $\phi$  level of the  $6^2P_{1/2}$  state (the  $\psi \rightarrow \phi$  transition is shown using a straight arrow) and the probability of its thermally assisted non-radiative transition from the  $\psi$  to  $\chi$  sub-level of the  $Tl^0$  center (the  $\psi \rightarrow \chi$  transition is shown using a bent arrow) determines its lifetime at the  $\psi$  sub-level. An electron can occupy the  $\chi$  sub-level either from a 5p level  $I^-$  ion assisted by a photon with an excitation energy of approximately 3.0 eV or from the  $\phi$  level of a  $Tl^0$  center assisted by the optical cascade excitation (a bent arrow labeled “cascade”). The electron occupying the  $\chi$  sub-level becomes mobile until a  $[Tl^+V_k]$  center captures it, forming a short interspace pair  $[Tl^0V_k]$ , which is responsible for the 2.3 eV emission.

The decrease in the number of electrons in the  $\psi$  sub-level can be expressed as

$$-dn_\psi = n(\nu_i + P_{\psi \rightarrow \chi})dt \quad (1)$$

where  $\nu_i$  is the probability of spontaneous radiative transitions,  $P_{\psi \rightarrow \chi} = \omega \cdot \exp(-E_a/kT)$  is the probability of thermally assisted non-radiative electron transitions to the conduction sub-band,  $\omega$  is the frequency factor and  $E_a$  is the thermal activation energy.

Having integrated (1), we obtain

$$n_\psi(t) = n_{\psi 0} \exp(-(\nu_i + P_{\psi \rightarrow \chi})t) \quad (2)$$

where  $n_{\psi 0}$  is the number of electrons in the  $\psi$  sub-level immediately following the depletion of the laser pulse at  $t=0$ .

The temperature dependencies of the luminescence decay constant at 1.6  $\mu m$  and 2.3 eV are obtained from (2) as follows:

$$\tau(T) = \frac{1}{\nu_i + \omega \cdot \exp(-E_a/kT)} \quad (3)$$

The increases in the photocurrent and photoluminescence intensities at temperatures over 295 K are both expressed as

$$I(T) = A \cdot \exp(-E_a/kT) \quad (4)$$

where  $A = 10^{-4}$  is a proportionality factor.

The curves calculated using Eqs. (3) and (4) are shown as solid lines in Fig. 4, and the numerical values of the fitting parameters for these curves are given in Table 1.

## 4. Conclusion

The charge transfer processes in CsI:Tl crystals are responsible for the exhibited bulk conductivity and recombination luminescence. Near-UV radiation facilitates a charge transfer process from  $I^-$  to  $Tl^+$  ions, forming  $Tl^0$  centers in the  $6^2P_{1/2}$  ground and  $6^2P_{3/2}$  excited states. The electron, assisted by phonons, leaves the  $Tl^0$  center from either the  $6^2P_{1/2}$  or  $6^2P_{3/2}$  states and overcomes either the 0.13 or 0.32 eV energy barrier, respectively, and populates the activator conduction sub-bands, which are found inside the band gap of the CsI:Tl crystal. The formation of the activator sub-bands is only possible when the Tl concentration is greater than  $3 \times 10^{-3}\%$ .

## Acknowledgments

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