



Luminescence response of CsI:Na to electron pulse irradiation

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HIGHLIGHTS

- ▶ Processes of the energy transfer to emission centers in CsI:Na is explained.
- ▶ Mathematical description of the mechanism of energy transfer to activator emission centers is given.
- ▶ V_{kA} thermal dissociation is the main factor in forming the scintillation pulse in CsI:Na.
- ▶ Activator emission centre in CsI:Na is Na-bound two halide exciton.

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ABSTRACT

This paper investigates the influence of activator concentration and temperature on the scintillation process in CsI:Na crystal which was excited by a pulsed electron beam ($E_{ex} = 0.25$ MeV, $t_{1/2} = 15$ ns, $W = 0.003 \dots 0.16$ J/cm²) at temperature within 77–300 K. It has been established that the transition of Na-bound two halide exciton from singlet state causes 3 eV emission of CsI:Na. The capturing of V_k by Na^+ ion determines the scintillation pulse shape. Thermal liberation of V_k from V_{kA} centers results in an inertial rise of the emission pulse in lightly activated CsI:Na and in a mono-exponential decay of the emission pulse in heavily activated CsI:Na. The value of thermal dissociation energy of V_{kA} centers is 0.24 ± 0.01 eV, which was obtained from the temperature dependences of the emission decay time constant for CsI:2.8·10⁻³%Na and of the rise time constant for CsI:2.0·10⁻⁴%Na.

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1. Introduction

This paper continues the series of our investigation of the scintillation process in activated alkali halide crystals by means of the time resolved spectroscopy (Trefilova et al., 2010; Yakovlev et al., 2009, 2011), which can be successfully applied for studying both thermally activated and tunnel processes. The obtained results instigated us to suggest the scintillation mechanisms for CsI:Tl and CsI:Na. The cause of the scintillation in CsI:Tl is the tunnel recombination in the short interspace pairs $\{Tl^0, V_k\}$ (Yakovlev et al., 2009). In CsI:Na the same tunnel recombination in the short interspace pairs $\{Na^0, V_k\}$ gives rise to the Na-bound two halide singlet excitons, and their radiative annihilation results in the scintillations (Yakovlev et al., 2011).

Abduvaliev et al. (1975), Aluker et al. (1987), Gutan et al. (1974), Popov et al. (1997), Spaeth et al. (1994), Zazubovich (2001) have suggested various mechanisms of energy transfer for CsI:Tl but they fail to fit the kinetics of the rise and decay of a scintillation pulse in

a wide temperature interval for the crystals with different activator concentration. Our model of the mechanism of scintillation processes in CsI:Tl appear to overcome the faults of its predecessors. The aim of this paper is to explain the influence of activator concentration and temperature on the yield of the scintillations and the shape of scintillation pulse for CsI:Na.

2. Materials and methods

Two samples of CsI:Na containing 2.8·10⁻³%Na and 2.0·10⁻⁴%Na were investigated under pulsed electron beam generated by an electron accelerator GIN-600. Na concentration was chemically determined in mass%. The electron beam had the following parameters: average electron energy $E_e = 0.25$ MeV, pulse duration $t_{1/2} = 15$ ns and power density 0.003...0.16 J/cm². The scheme and the procedure for the optical measurements with a time resolution are the same as those described by Ueta et al. (1969). The luminescence pulse and transparency restoration after the electron pulse depletion were registered with an optical spectrometer consisting of a monochromator MDR-3, photomultipliers FEU-83, FEU-106 and a digital oscilloscope GDS-2204. A pulsed lamp INP-5 × 75 was used as a source of probe light.

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3. Results

3.1. Transient absorption and recombination emission kinetics of CsI:Na at 80 K

The transient absorption spectra of CsI:2.8·10⁻³%Na and CsI:2.0·10⁻⁴%Na shown in Fig. 1 consist of the bands related to Na⁰, V_k centers (Mori et al., 1980; Silder et al., 1973) and self-trapped excitons (STE) (Gafiatulina et al., 1998). However, we observed V_k and Na⁰ bands at 3.02 eV and 1.72 eV, respectively, right after an electron pulse depletion in CsI:2.8·10⁻³%Na, whereas we could see the same bands in the spectra of CsI:2.0·10⁻⁴%Na only after 5 μs when STE absorption had decayed (curve 3 in Fig. 1b). V_k centers capture electrons, giving rise to STE in CsI, whereas in CsI:Na not only V_k centers but Na⁺ ions capture electrons, forming Na⁰ centers. Being a deep electron trap, Na⁺ ion significantly reduces the probability of STE formation, and therefore the optical density of CsI:2.0·10⁻⁴%Na at 1.72 eV is about 15 times weaker than that of CsI:2.8·10⁻³%Na.

Aluker et al. (1987) experimentally proved that an electron pulse irradiation gives rise to V_k and Na⁰ color centers in CsI:Na, and their recombination causes the scintillations. We measured the emission spectra of the both crystals at various probe delays and detected three bands at 4.2, 3.7 and 3.0 eV. Fig. 2 illustrates the spectra for CsI:2.8·10⁻³%Na at 80 K. Nishimura et al. (1995) related the bands at 4.2 and 3.7 eV to the radiative annihilation of singlet STE and triplet STE in defect-free areas of CsI lattice, respectively. As for the band at 3.0 eV, it arises due to Na and fits a Gaussian with a maximum 3.0 eV and FWHM = 0.44 ± 0.02 eV.

The emission of self-trapped excitons at 4.3 eV and 3.7 eV decays exponentially with time constants 10 ns and 0.85 μs at 80 K, respectively. The emission at 3.0 eV has a complex decay because its kinetics varies throughout nano-, micro- and millisecond regions. The kinetic curves of this emission are presented for CsI:2.8·10⁻³%Na and CsI:2.0·10⁻⁴%Na in log scale in Fig. 3.

It has been found that the emission decay kinetics of CsI:2.8·10⁻³%Na and CsI:2.0·10⁻⁴%Na within 1·10⁻⁸–5·10⁻² s fits the sum of four components: three exponential and a hyperbolic one:

$$I(t) = \sum_{i=1}^3 I_i \cdot e^{-t/\tau_i} + I_0 \cdot (1 + \alpha \cdot t)^{-\beta} \quad (1)$$

where I_i is intensity peak for i -component, $\tau_1 \leq 10$ ns, $\tau_2 = 1.0$ μs, $\tau_3 = 3.0$ μs, $\beta = 0.55$.

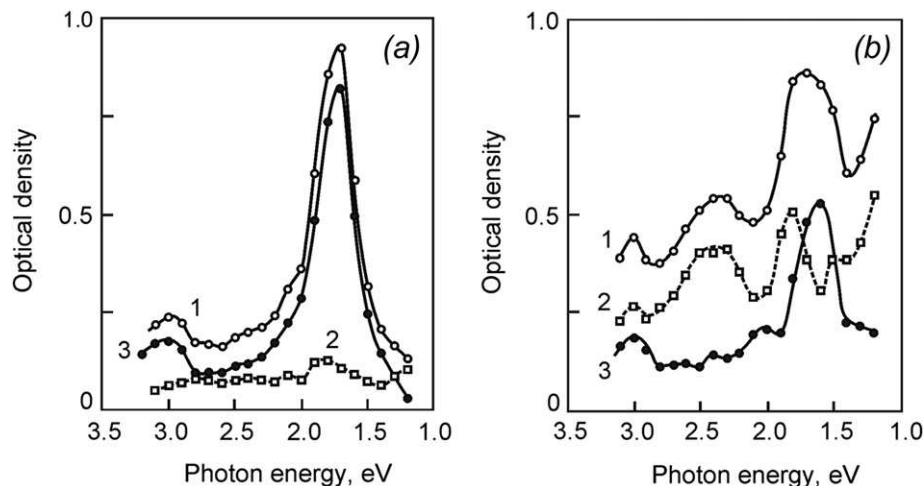


Fig. 1. Absorption spectra of CsI:2.8·10⁻³%Na (a) and CsI:2.0·10⁻⁴%Na (b) at 80 K: 1 – right after pulse electron irradiation, 2 – at 5 μs delay; 3 – the spectrum of short-living absorption.

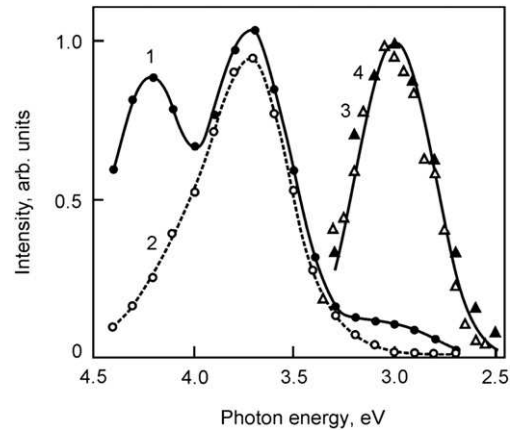


Fig. 2. Cathode-luminescence spectra of CsI:2.8·10⁻³%Na at 80 K: 1 – right after pulse electron irradiation, 2 – at 100 ns delay; 3 – at 5 μs delay, 4 – at 1 ms delay.

3.2. Emission decay kinetics of CsI:Na at different temperatures

T = 80–130 K. We have found that the parameters of emission decay kinetics are the same for CsI:2.8·10⁻³%Na and CsI:2.0·10⁻⁴%Na. However, the temperature has a different effect on the parameters of exponential and hyperbolic components. The time constants of exponential components (exponential factor in (1)) up to 150 K (Fig. 4 curve 1,2) and intensity peak I_i (pre-exponential factor in (1)) remain the same. As for the hyperbolic component, at temperature over 90 K its index β springs from 0.55 to 2, owing to the temperature which sets V_k centers in motion (Silder et al., 1973).

T = 130–300 K. The crystals CsI:2.8·10⁻³%Na and CsI:2.0·10⁻⁴%Na considerably differ in decay kinetics. The comparison of our results with literature data shows that the parameters of the hyperbolic decay component for CsI:2.0·10⁻⁴%Na and for CsI:Na (Aluker et al., 1987) have similar temperature behavior.

CsI:2.0·10⁻⁴%Na. At temperature over 90 K, the slow decay component of the emission for CsI:2.0·10⁻⁴%Na fits the following hyperbolic function:

$$I(t) = I_0 \cdot (1 + \alpha \cdot t)^{-2} \quad (2)$$

According to Aluker et al. (1987) the bimolecular decay kinetics (2) is caused by the recombination of mobile V_k with Na⁰. At

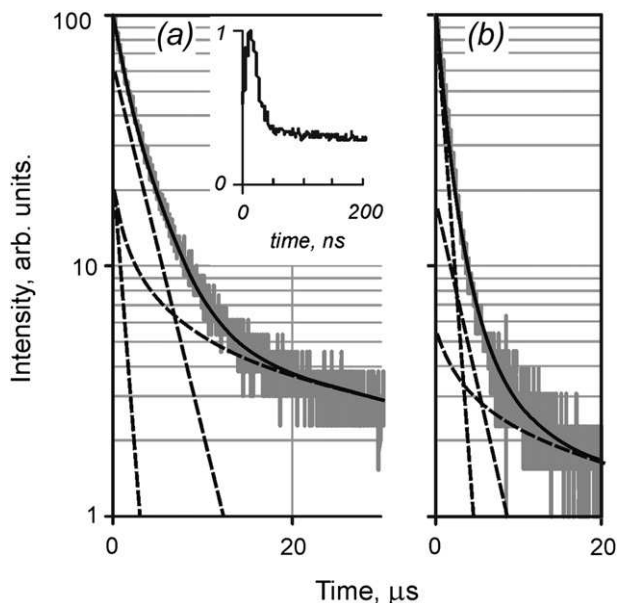


Fig. 3. Pulse decay curves of cathode-luminescence at 2.9 eV for CsI:2.8·10⁻³%Na (a) and for CsI:2.0·10⁻⁴%Na (b) at T = 80 K. An oscillogram inserted in Fig. 3a shows the fast decay component with τ₁ ≤ 10 ns. Fitting decay components are in dashed lines and their sum in a solid one.

temperature over 140 K the 3.0 eV emission pulse rise becomes inertial, exceeding the duration of the excitation pulse. [Abduvaliev et al. \(1975\)](#) defined the emission pulse constant rise τ_r as the time of the pulse intensity rise from 0.1 to 0.9 of its maximum. The higher the temperature the faster the emission pulse reaches its maximum and the faster its hyperbolic component decays. This behavior is illustrated by the oscillogram family in a log–log scale in [Fig. 5a](#). Temperature dependencies of kinetics parameters α(T) and τ_r(T) are shown in [Fig. 4a](#) (curve 3,4). The values of the thermal activation energy estimated by the slope angle of α(T) and τ_r(T) vs. 1/T are E_a = 0.2 ± 0.01 eV and E_a = 0.13 ± 0.01 eV, respectively.

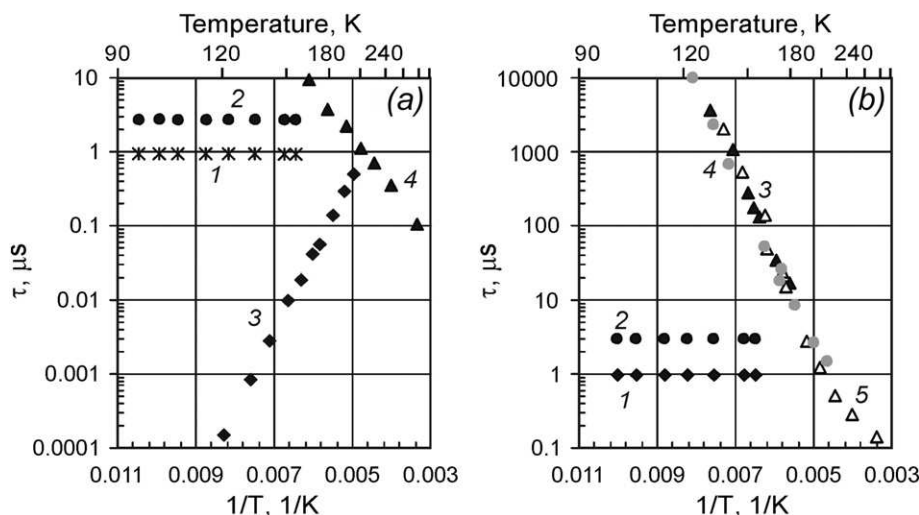


Fig. 4. (a) Kinetics parameters of an emission pulse at 2.9 eV vs. reciprocal temperature for CsI:2·10⁻⁴%Na: 1 (asterisks) – exponential decay time τ₂; 2 (black dots) – exponential decay time τ₃; 3 (back rhombuses) – hyperbola parameter α; 4 (black triangles) exponential – rise time τ_r. (b) Kinetics parameters of an emission pulse at 2.9 eV vs. reciprocal temperature for CsI:2.8·10⁻³%Na: 1 (black rhombuses) – exponential decay time τ₂; 2 (black dots) – exponential decay time τ₃; 3 (black triangles) – exponential decay time τ_d for W₁ = 1.6 mJ/cm²; 4 (grey dots) – exponential decay time τ_d for W₂ = 12.8 mJ/cm². For CsI:2·10⁻⁴%Na: 5 (white triangles) – exponential rise time τ_r of V_k centers which fits the expression (6).

The same values of activation energy were demonstrated by [Silder et al. \(1973\)](#) and [Abduvaliev et al. \(1975\)](#), respectively.

CsI:2.8·10⁻³%Na. emission pulse curves for various temperatures become straight in a log scale ([Fig. 5b](#)) because the emission pulse decays exponentially, which leads us to believe that a monomolecular process gives rise to the decay. However, according to the law of formalistic kinetics ([Atkins and de Paula, 2010](#)) the exponential decay may be caused also by a bimolecular process (so called reaction of pseudo first order), and in this case the time constant of exponential decay τ_d is inversely to the initial concentration of Na⁰ and V_k centers originated by an irradiation pulse. We found that the density (W₁ = 1.6 mJ/cm² and W₂ = 12.8 mJ/cm²) of the electron beam does not affect the temperature dependence of cathode luminescence time decay ([Fig. 4b](#) curve 3,4). It proves that only a monomolecular process with E_a = 0.24 ± 0.02 eV is responsible for the emission decay. A further analysis reveals that the emission decay kinetics has not only the above mentioned exponential component but a hyperbolic one, which takes less than 10% of the initial intensity. The sum of the exponential and hyperbolic functions I(t) = I₁·e^{-t/τ} + I₂·(1 + α·t)⁻² fits the decay kinetics of CsI:2.8·10⁻³%Na ([Fig. 5b](#)).

4. Discussion

4.1. The origin of exponential components with decay constants 1.0 and 3.0 μs at temperature within 80–150 K

According to [Aluker et al. \(1986\)](#), Na⁰ and V_k centers arise in CsI:Na under electron pulse irradiation, and their recombination causes 3.0 eV emission, which has been verified by our experiments. We found that the spectrum of 3.0 eV emission fits a Gaussian and remains unchanged at different time delays. The exponential decay time constants of this emission are not affected by Na concentration and temperature from 80 to 150 K. We also detected four emission decay components: three exponential (τ₁ ≤ 10 ns, τ₂ = 1.0 μs, τ₃ = 3.0 μs) and a hyperbolic one. The presence of the fast exponential component with τ ≤ 10 ns in the decay kinetics proves the radiative transition of the emission centers to be allowed by the selection rules. Thus, the three slow

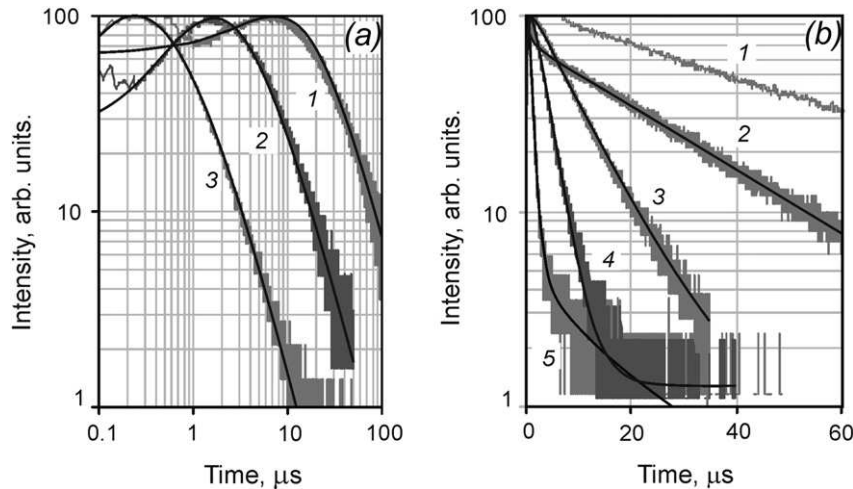


Fig. 5. Pulse decay curves of cathode-luminescence at 2.9 eV for CsI:2.0 · 10⁻⁴%Na (a) and CsI:2.8 · 10⁻³%Na (b) at different temperature: a) 1–176 K; 2–208 K; 3–295 K; b) 1–160 K; 2–172 K; 3–183 K; 4–200; 5–238 K. Fitting curves are shown by solid line.

components (two exponential and a hyperbolic) testify the presence of various channels through which the emission centers are populated.

It is worth noting that both Na⁰ and V_k centers at 80 K are immobile in the lattice and the only possible mechanism of their radiative recombination can be a tunnel one. [Huntley \(2006\)](#) theoretically analyzed the tunneling of electrons from traps to the centers of recombination and showed that the tunnel luminescence decay kinetics fits well the hyperbolic law with the index >1 or <1, depending on the trap concentration. His theory gives good explanation for the origin of the hyperbolic component with the index 0.55 in the decay kinetics of CsI:Na cathode-luminescence. As for both the microsecond exponential decay components, like in CsI:Tl they are caused by electron tunneling in short interspace pairs {Na⁰, V_k} which can be of two types $r_1 = \sqrt{2}/2a$ (type I) and $r_2 = \sqrt{6}/2a$ (type II) where a – lattice constant ([Fig. 6](#)). Thus, it is safe to assume that the emission band with maximum at 3.0 eV of

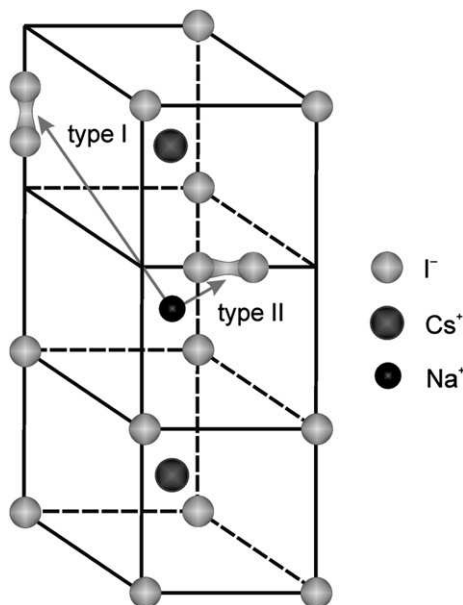


Fig. 6. Two types of short interspace pairs {Na⁰, V_k} in CsI lattice.

CsI:Na is caused by the radiative annihilation singlet excitons localized near Na⁺ ions. Such excitons arise either through the direct relaxation of low energy electron excitations or through the electron tunneling in all kinds of interspace pairs {Na⁰, V_k}. The former is responsible for the fast exponential decay component ($\tau \leq 10$ ns) and the latter for two exponential components ($\tau = 1.0$ and $3.0 \mu\text{s}$) as well as a hyperbolic one ($\beta \leq 1$).

4.2. The effect of Na concentration and temperature on recombination process

It is known that the temperature higher 90 K sets V_k centers in motion ([Silder et al., 1973](#)) which leads to the formation of short interspace pairs {Na⁰, V_k} in the crystal. The 3.0 eV emission decay kinetics significantly depends on Na concentration. Although the emission decay of CsI:2.8 · 10⁻³%Na and CsI:2.0 · 10⁻⁴%Na fits the kinetics of the first and the second orders, respectively, the inertial rise of CsI:2.0 · 10⁻⁴%Na and the exponential decay of CsI:2.8 · 10⁻³%Na have almost the same duration at any fixed temperature within 160–295 K ([Fig. 5](#)), for instance at 162 K ([Fig. 7](#)).

Now we shall endeavor to explain the origin of slow emission pulse rise which duration is much longer than the lifetime of short interspace pairs {Na⁰, V_k} ([Fig. 5a](#)). [Schoemaker \(1973\)](#) and [Galaganov et al. \(1987\)](#) believe that light cations in alkali halide crystals trap V_k centers with formation of V_{kA} centers. Therefore being a light cation for CsI, Na⁺ traps V_k center, delaying Na⁰-V_k recombination which can occur after thermal liberation of V_k from V_{kA} centers. This can be an explanation of the slow emission pulse rise for CsI:2.0 · 10⁻⁴%Na.

[Aluker et al. \(1987\)](#) analyzed the recombination emission with the expression (2) for the kinetics of the second order, believing that the initial concentration of two reagents ($N_{t=0}(\text{Na}^0) = N_{t=0}(\text{V}_k) = N_0$) is equal. The kinetics parameters of I_0 and α are expressed as follows:

$$I_0 = (k_2 \cdot N_0)^2; \quad \alpha = k_2 \cdot N_0 \quad (3)$$

where k_2 – reaction rate constant of the second order, N_0 – concentration of Na⁰ and V_k centers.

We find that such approximation is too simplistic, because not all V_k centers are mobile right after irradiation. Some number of V_k centers is caught by Na⁺ ions, resulting in V_{kA} centers. These Na-bound V_k centers can get mobile only after their thermal

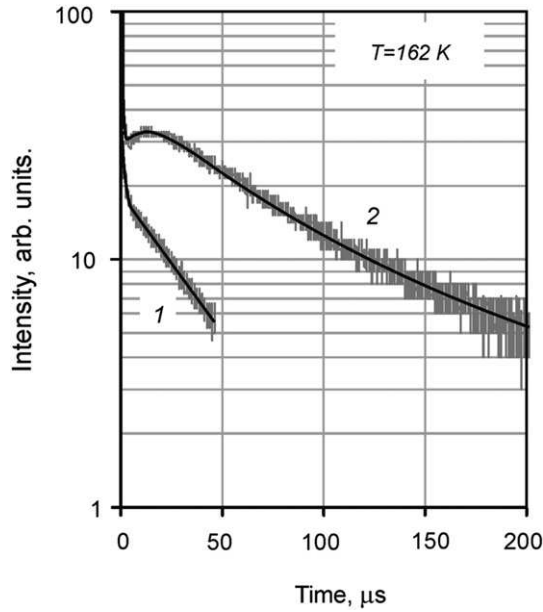


Fig. 7. Oscillograms of scintillations for $2.8 \cdot 10^{-3}\%Na$ (1) and $CsI:2.0 \cdot 10^{-4}\%Na$ (2) Fitting curves for (1) and (2) were calculated by the expressions (7) and (6), respectively.

liberation from V_{kA} . Once temperature is higher the temperature at which V_{kA} centers start dissociating, the number of mobile V_k centers varies with time as follows:

$$Na^0(0) = V_k(0) + V_{kA}(0) \quad (4)$$

$$V_k(t) = V_k(0) + V_{kA}(0) \cdot (1 - e^{-pt}) \quad (5)$$

where $p = 1/\tau_{rise} = \omega \cdot \exp(-E_a/kT)$ is probability of V_{kA} thermal dissociation, ω is frequency factor, E_a is thermal activation energy for liberation of V_k from V_{kA} centers. It is worth noticing that V_{kA} thermal dissociation process is monomolecular. Using the expressions (4) and (5) in (2) we obtained the expression (6), which fits the luminescence kinetics:

$$I(t) = \frac{I_1 + I_2 \cdot (1 - e^{-t/\tau})}{(1 + \alpha \cdot (I_1 + I_2 \cdot (1 - e^{-t/\tau}) \cdot t))^2} \quad (6)$$

Fitting curves calculated by the expression (6) are shown in solid lines in Fig. 5a.

We found that the rise time constant τ_{rise} of V_k centers for $CsI:2.0 \cdot 10^{-4}\%Na$ (Fig. 4b curve 5) and the emission decay time τ_d for $CsI:2.8 \cdot 10^{-3}\%Na$ (Fig. 4b curve 3,4) are almost equal within 130–295 K. The emission decay kinetics of $CsI:2.8 \cdot 10^{-3}\%Na$ is caused by a monomolecular process which was experimentally proved in 3.2. Thus, we have arrived to the following conclusion that the same process of V_{kA} thermal dissociation is responsible for an inertial rise of emission pulse for $CsI:2.0 \cdot 10^{-4}\%Na$ and mono-exponential decay of emission pulse for $CsI:2.8 \cdot 10^{-3}\%Na$.

According to Aluker et al. (1986) the emission decay kinetics of $CsI:2.0 \cdot 10^{-4}\%Na$ caused by the recombination reaction of V_k with Na^0 with a rate proportional to the product of concentrations of the reagents. The rate of recombination reaction in $CsI:2.8 \cdot 10^{-3}\%Na$ is expected to be higher than the rate of monomolecular process of V_{kA} dissociation. Thus, the activator emission arises right after V_k liberation, and therefore the emission decay kinetics of $CsI:2.8 \cdot 10^{-3}\%Na$ is determined by the rate of V_{kA} center dissociation.

$$I(t) = -\frac{dV_{kA}}{dt} = -\frac{d}{dt} [V_{kA}(0) \cdot e^{-pt}] = V_{kA} \cdot p \cdot e^{-pt} = I_0 \cdot e^{-t/\tau} \quad (7)$$

5. Conclusion

The detailed study of the influence of Na concentration and temperature on the scintillation process in $CsI:Na$ can be summarized in the following way. We have established that the transition of Na-bound two halide exciton from singlet state causes 3 eV emission of $CsI:Na$. The capturing of V_k by Na^+ ion determines the scintillation pulse shape. Thermal liberation of V_k from V_{kA} centers results in an inertial rise of the emission pulse in lightly activated $CsI:Na$ and in a mono-exponential decay of the emission pulse in heavily activated $CsI:Na$. The value of thermal dissociation energy of V_{kA} centers is 0.24 ± 0.01 eV, which was obtained from the temperature dependences of the emission decay time constant for $CsI:2.8 \cdot 10^{-3}\%Na$ and of the rise time constant for $CsI:2.0 \cdot 10^{-4}\%Na$.

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