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Radiative and non-radiative processes of Ag^- centres doped in NaCl single crystals

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Abstract

Luminescence properties including decay kinetics have been investigated for Ag^- centres doped in NaCl single crystals in the temperature range from 7 to 250 K. When the C absorption band peaking at 4.5 eV is photo-excited at low temperature, the A' and C' luminescence bands are observed at 2.85 and 4.12 eV, respectively. As the temperature increases, the A' luminescence grows, with compensation for the decrease of the luminescence intensity of the C' band, reflecting a thermally activated non-radiative process from the $^1\text{T}_{1u}$ to $^3\text{T}_{1u}$ states. With increasing temperature above 130 K, the A' luminescence intensity also decreases. Since the decay time of the A' luminescence is almost constant at 10 μs above 80 K, the decrease of the A' luminescence intensity is not attributed to a non-radiative process from the $^3\text{T}_{1u}$ state but arises from the quenching of the non-radiative relaxation from the $^1\text{T}_{1u}$ to $^3\text{T}_{1u}$ states. The temperature dependence of the luminescence intensities of the A' and C' bands is discussed on the basis of a simple configuration coordinate model, which consists of the $^1\text{T}_{1u}$, $^3\text{T}_{1u}$ and $^1\text{A}_{1g}$ adiabatic potential energy surfaces intersecting at each other.

1. Introduction

Anionic Ag^- ions doped in alkali halide crystals belong to a family of TI^+ -type impurity centres [1–8]. The ions have the electronic configuration ns^2 in the ground state and an $a_{1g}t_{1u}$ configuration in the first excited state [9–11]. The ions doped in alkali halides exhibit three absorption bands called A, B and C in the order of increasing energy below the band edge of host crystals. The A, B and C absorption bands are attributed to the intraionic transitions as follows.

A band: ${}^1A_{1g} \rightarrow {}^3T_{1u}$, which is allowed owing to the spin-orbit interaction.

B band: ${}^1A_{1g} \rightarrow {}^3T_{2u} + {}^3E_u$, which is partially allowed owing to noncubic phonons.

C band: ${}^1A_{1g} \rightarrow {}^1T_{1u}$, which is a dipole allowed transition.

In addition to these states, the ${}^3A_{1u}$ state exists below the ${}^3T_{1u}$ state.

When these absorption bands are photo-excited, the luminescence bands due to the radiative transition from their relaxed states are observed with Stokes shifts [9–11]. In the case of the cationic Tl^+ -type centres such as Tl^+ , In^+ and Ga^+ , the luminescence properties are remarkably complex, especially under the excitation on the C absorption band. Even under the excitation on the lowest A absorption band, the cationic Tl^+ -type centres often exhibit two luminescence bands, so-called A'_T and A'_X . The two luminescence bands arise from two kinds of minima coexisting on the adiabatic potential energy surface (APES) of the ${}^3T_{1u}$ excited state. Several investigations have been carried out for the non-radiative process between two kinds of minima on the ${}^3T_{1u}$ and ${}^3A_{1u}$ APESs [12–15].

On the other hand, unlike the case of the cationic Tl^+ -type centres, the Ag^- centres doped in alkali halides exhibit no two A' luminescence band. As compared with the cationic Tl^+ -type centres, the luminescence processes under the excitation on the C band are comparatively simple. In the standard case for the Ag^- centres [16–18], the C band excitation exhibits mainly the C' luminescence due to the radiative transition from the ${}^1T_{1u}$ APES at low temperature. As the temperature increases, the C' luminescence intensity decreases and the A' luminescence intensity increases complementary with the decrease of the C' luminescence intensity. In the higher temperature range, only the A' luminescence band is observed and finally the A' luminescence itself diminishes and disappears. The temperature variation of these luminescence intensities is probably due to the non-radiative processes between the ${}^1T_{1u}$ and ${}^3T_{1u}$ APESs, and/or the ${}^3T_{1u}$ and ${}^1A_{1g}$ APESs, and is subject to the static process following the thermal equilibrium on the ${}^1T_{1u}$ and ${}^3T_{1u}$ APESs.

In this paper, we have investigated the temperature dependence of the A' and C' luminescence intensities and of the decay time in $NaCl:Ag^-$ single crystals. From the experimental results, the non-radiative processes among the ${}^1T_{1u}$, ${}^3T_{1u}$ and ${}^1A_{1g}$ APESs in the space of configuration coordinates are discussed. This study would provide information on the dynamical non-radiative processes at the intersection points between the APESs consisting of the triplet and singlet states.

2. Experimental method

Single crystals of $NaCl$ containing Ag^+ ions were grown by the Kyropoulos method under a N_2 gas atmosphere. The conversion from Ag^+ to Ag^- ions was achieved through electrolytic colouration or irradiation with x-rays.

The sample was mounted on a copper cold stage connected to a cryogenic refrigerator. The temperature of the sample was monitored with an accuracy of 1.0 K with a $AuFe:chromel$ thermocouple attached to the cold stage.

In order to measure the luminescence and excitation spectra, light emitted from a 2 kW Xe lamp was made monochromatic by a grating monochromator (JASCO: CT25CS) and was used as an exciting light source. The luminescence from the sample was passed through another monochromator (Spex 270M) and was detected with a photomultiplier (Hamamatsu: R-955). All the spectra were corrected for the spectral distribution of the excitation energy, dispersion of the monochromator and spectral sensitivity of the detector.

In the measurements of the temporal behaviour of the luminescence, second harmonic output from an OPO laser system (Spectra-Physics: MOPO-HF) with a pulse duration of ~ 10 ns and a repetition rate of 10 Hz was employed. Temporal profiles of the luminescence

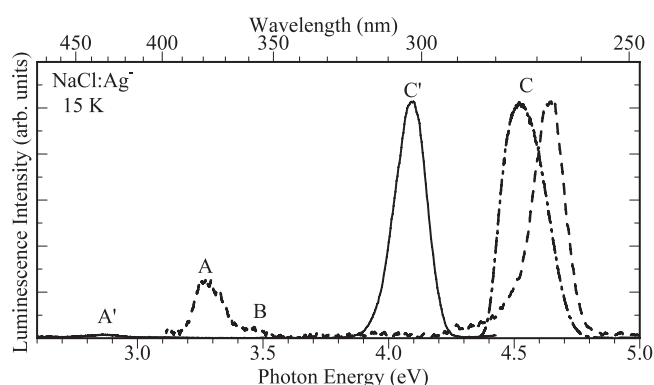


Figure 1. Luminescence and excitation spectra in NaCl:Ag⁻ crystals at 15 K. The solid line is the luminescence spectrum under excitation on the C absorption band. Broken and dash-dotted lines are the excitation spectra for the A' and C' luminescence bands, respectively. All maxima of the luminescence intensity are normalized.

intensity were detected with a streak camera system (Hamamatsu: C2909) through a grating monochromator (Oriel: 77480).

3. Experimental results

In general, the A absorption band of Ag⁻ centres doped in alkali halides has a much smaller intensity than the C absorption band, because of the weak spin-orbit interaction [2, 3]. Thus, the A absorption band cannot be observed except in samples with extremely high concentration of impurity. Figure 1 shows schematically the luminescence and excitation spectra of the Ag⁻ centres doped in NaCl single crystals at 15 K. Solid, broken and dash-dotted lines are the luminescence spectrum and the excitation spectra for the A' and C' luminescence, respectively. Under the excitation on the C absorption band peaking at 4.5 eV, both the A' and C' luminescence bands are observed at 2.85 and 4.12 eV, respectively. In the excitation spectra for the A' luminescence, the A and B bands are found near 3.27 and 3.48 eV, respectively. The excitation spectrum for the C' luminescence exhibits a remarkable response around 4.5 eV, whose shape is almost equal to that of the C absorption band. These values are coincident with those reported in the previous paper [16].

Under the excitation on the C band, the C' luminescence band decreases and the A' luminescence band increases with increasing temperature. Figure 2 shows the temperature dependence of the luminescence intensities of the A' and C' bands. The vertical axis is a logarithmic scale. The luminescence intensity of the A' band is smaller than that of the C' band at low temperature. Above 30 K, the luminescence intensity of the C' band decreases and that of the A' band increases with compensation for the decrease of the C' luminescence intensity. This fact implies that the thermally activated non-radiative relaxation from the ¹T_{1u} to ³T_{1u} APESs occurs.

As the temperature increases from 130 K, the luminescence intensity of the A' band decreases. The decrease of the A' luminescence intensity is not attributed to the non-radiative relaxation from the ³T_{1u} APES, because the decay time constant of the A' luminescence does not change in this temperature range, as mentioned below. That is to say, the non-radiative relaxation from the ³T_{1u} APES should bring about the reduction of the decay time of the A' luminescence. Therefore, the decrease of the A' luminescence intensity arises from the

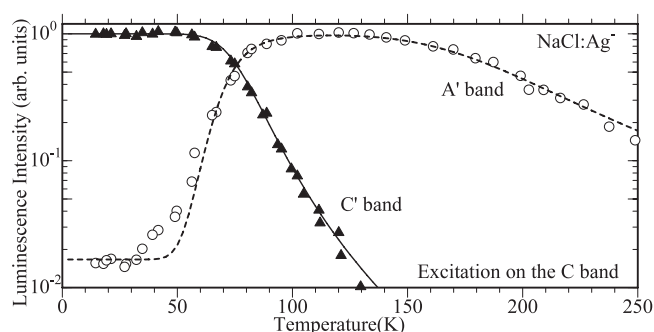


Figure 2. Logarithmic plots of the luminescence intensities of the A' (open circles) and C' (filled triangles) bands as a function of the temperature. Maxima of the A' and C' luminescence intensities are normalized to unity. The solid and broken curves represent the best-fitted results of equations (6) and (7) for the C' and A' luminescence intensities, respectively.

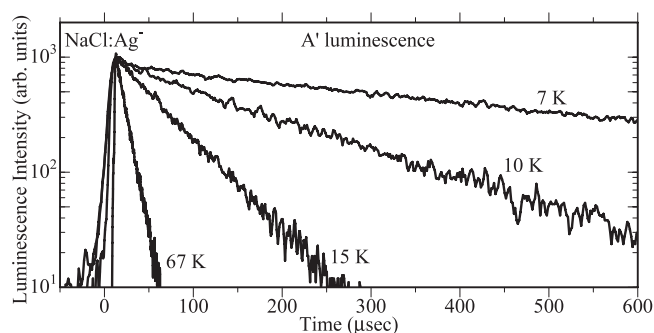


Figure 3. Decay profiles of the A' luminescence in $\text{NaCl}:\text{Ag}^-$ crystals at various temperatures. All maxima of the luminescence intensity are normalized.

quenching of the non-radiative relaxation from the $^1T_{1u}$ to $^3T_{1u}$ APESs. This idea leads to the occurrence of another thermally activated non-radiative process from the $^1T_{1u}$ APES. This process is thought to be the non-radiative transition from the $^1T_{1u}$ APES to the $^1A_{1g}$ APES, which corresponds to the ground state.

Figure 3 shows the temporal behaviours of the A' luminescence under excitation on the C absorption band at various temperatures. The A' luminescence exhibits a single exponential decay at any temperature. In case of the cationic Tl^+ -type centres, a fast decay component with the decay time of a few nanoseconds is usually observed in the temporal behaviour of the A' luminescence [11, 14]. In $\text{NaCl}:\text{Ag}^-$, such a fast decay component could not be confirmed in the A' luminescence by our detecting system.

As the temperature increases from 7 K, the decay time of the A' luminescence becomes fast. Figure 4 shows the temperature dependence of the decay time constant of the A' luminescence. The decay time constant at 7 K is about $530 \mu\text{s}$. As seen in figure 4, the decay time exhibits a rapid decrease with increasing temperature. Above 80 K, the decay time has the constant value of about $10 \mu\text{s}$. The decay time constant of the A' luminescence is not different between the excitation of the A and C absorption bands. This similarity indicates that the radiative process from the $^3T_{1u}$ APES is dominated not by the relaxation process to the minimum point on the APES but the dynamical process after the relaxation to the minimum.

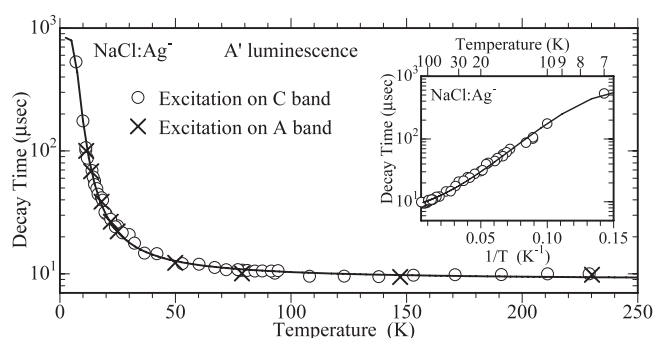


Figure 4. Temperature dependence of the decay time constant of the A' luminescence in NaCl:Ag⁻ crystals. The vertical axis of the figure is the logarithmic scale. The open circles and crosses correspond to the decay times under the excitation on the C and A absorption bands, respectively. The solid curve represents the best-fitted result of the equation (9) for the experimental results. Inset is the logarithmic plot of the decay time as a function of the reciprocal temperature.

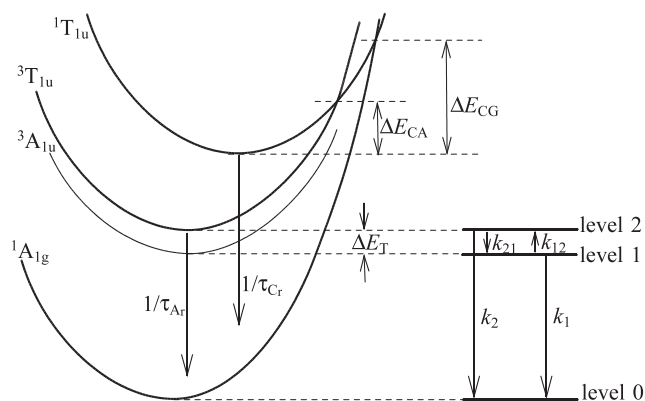


Figure 5. Configuration coordinate diagram for the A' and C' luminescence processes in NaCl:Ag⁻ crystals. On the right in the figure is the three-level model.

4. Discussion

As mentioned in the previous section, the temperature dependence of the A' and C' luminescence intensities is governed by the non-radiative processes among the ¹T_{1u}, ³T_{1u} and ¹A_{1g} APESs. On the other hand, it has been known that the A' luminescence of the TI⁺-type centres is attributed to the radiative transition from the ³T_{1u} APES and the ³A_{1u} APES located below the ³T_{1u} APES [11, 13–15]. The ³T_{1u} and ³A_{1u} APESs are connected by a thermally activated non-radiative process, which plays an important role in the decay kinetics of the A' luminescence.

In order to explain the temperature dependence, we introduce a configuration coordinate model consisting of the ¹T_{1u}, ³T_{1u}, ³A_{1u} and ¹A_{1g} APESs, which is illustrated in figure 5. The ³A_{1u} APES is located at the lower energy of the ³T_{1u} APES by ΔE_T. The ¹T_{1u} APES intersects the ³T_{1u} and ¹A_{1g} APESs at points away from the minimum point. The heights of the cross points to the ³T_{1u} and ¹A_{1g} APESs from the minimum point of the ¹T_{1u} APES are ΔE_{CA} and ΔE_{CG}, respectively.

4.1. Temperature dependence of the luminescence intensity

First, we discuss the temperature dependence of the A' and C' luminescence intensities on the basis of the APESs as shown in figure 5. The A' luminescence is attributed to the radiative transitions from the ${}^3T_{1u}$ and ${}^3A_{1u}$ APESs, which are connected by a thermally activated non-radiative process. The temperature dependence of the decay time of the A' luminescence indicates no non-radiative process and the high luminescence efficiency from the ${}^3T_{1u}$ and ${}^3A_{1u}$ APESs. Now, we consider that the A' luminescence arises from one state: the ${}^3T_{1u}$ APES.

When the C absorption band is photo-excited, the population is created on the ${}^1T_{1u}$ APES. The population on the ${}^1T_{1u}$ APES disappears with time through the non-radiative and radiative processes. The population on the ${}^3T_{1u}$ APES is generated in terms of the non-radiative transition from the ${}^1T_{1u}$ to ${}^3T_{1u}$ APESs. Therefore, the populations $N_C(t)$ and $N_A(t)$ on the ${}^1T_{1u}$ and ${}^3T_{1u}$ APESs obey the following respective rate equations:

$$\begin{aligned}\frac{dN_C(t)}{dt} &= -\frac{N_C(t)}{\tau_C}, \\ \frac{dN_A(t)}{dt} &= -\frac{N_A(t)}{\tau_A} + \frac{N_C(t)}{\tau_{CA}},\end{aligned}\quad (1)$$

where τ_C^{-1} and τ_A^{-1} are the decay rates from the ${}^1T_{1u}$ and ${}^3T_{1u}$ APESs, respectively. Since the non-radiative transition from the ${}^3T_{1u}$ APES is not considered, τ_A^{-1} is equal to the radiative transition probability from the ${}^3T_{1u}$ APES. τ_{CA}^{-1} is the non-radiative transition probability from the ${}^1T_{1u}$ to ${}^3T_{1u}$ APESs. By solving the system of differential equations (1) with respect to $N_C(t)$ or $N_A(t)$, we get

$$N_C(t) = N_0 \exp\left(-\frac{t}{\tau_C}\right), \quad (2)$$

$$N_A(t) = \frac{N_0}{\tau_{CA}} \left(\frac{1}{\tau_A} - \frac{1}{\tau_C}\right)^{-1} \left\{ \exp\left(-\frac{t}{\tau_C}\right) - \exp\left(-\frac{t}{\tau_A}\right) \right\}, \quad (3)$$

where N_0 is the initial population on the ${}^1T_{1u}$ APES at $t = 0$.

Let us now consider the decay rate τ_C^{-1} and the non-radiative transition probability τ_{CA}^{-1} . As seen in figure 2, the A' luminescence is observed even at low temperature. This fact indicates that the non-radiative processes from the ${}^1T_{1u}$ to ${}^3T_{1u}$ APESs consist of a temperature-independent process such as the tunnelling process and a thermally activated process. When the non-radiative transition probability due to the tunnelling process is τ_t^{-1} , the non-radiative transition probability τ_{CA}^{-1} from the ${}^1T_{1u}$ to ${}^3T_{1u}$ APESs is given by

$$\tau_{CA}^{-1} = \tau_t^{-1} + \nu_{CA} \exp\left(\frac{-\Delta E_{CA}}{k_B T}\right), \quad (4)$$

where ν_{CA} is the frequency factor for the thermally non-radiative process to the ${}^3T_{1u}$ APES. The thermally non-radiative process from the ${}^1T_{1u}$ APES has two pathways: towards the ${}^3T_{1u}$ and ${}^1A_{1g}$ APESs. The activation energies due to the thermally activated processes are ΔE_{CA} and ΔE_{CG} , respectively. Thus, the decay rate τ_C^{-1} of the ${}^1T_{1u}$ APES is given by

$$\tau_C^{-1} = \tau_{CA}^{-1} + \tau_{C_r}^{-1} + \nu_{CG} \exp\left(\frac{-\Delta E_{CG}}{k_B T}\right), \quad (5)$$

where $\tau_{C_r}^{-1}$ is the radiative transition probability from the ${}^1T_{1u}$ APES and ν_{CG} is the frequency factor for the thermally non-radiative process to the ${}^1A_{1g}$ APES.

From equation (2) and the decay rate (5), the temperature dependence of the C' luminescence intensity $I_C(T)$ is obtained as follows:

$$I_C(T) = \tau_{C'}^{-1} \int_0^{\infty} N_C(t) dt = \frac{\tau_{C'}^{-1}}{\tau_t^{-1} + \tau_{C'}^{-1} + \nu_{CA} \exp\left(\frac{-\Delta E_{CA}}{k_B T}\right) + \nu_{CG} \exp\left(\frac{-\Delta E_{CG}}{k_B T}\right)}, \quad (6)$$

where the initial population at $t = 0$ is unity. From equation (3) and the non-radiative transition probability (4), the temperature dependence of the A' luminescence intensity $I_A(T)$ is given as follows:

$$I_A(T) \propto \frac{\tau_t^{-1} + \nu_{CA} \exp\left(\frac{-\Delta E_{CA}}{k_B T}\right)}{\tau_t^{-1} + \tau_{C'}^{-1} + \nu_{CA} \exp\left(\frac{-\Delta E_{CA}}{k_B T}\right) + \nu_{CG} \exp\left(\frac{-\Delta E_{CG}}{k_B T}\right)}. \quad (7)$$

We fit equations (6) and (7) to the experimental results. In the present study, the decay time constant of the C' luminescence could not be measured, because the decay time of the C' luminescence is much shorter than the pulse duration of the exciting laser. In general, the C' luminescence of the Ag⁻ centre has the decay time constant of about 1 ns, reflecting the dipole allowed transition [17]. Thus, we take $\tau_{C'} = 1.0 \times 10^{-9}$ s. There remain the five fitting parameters: ΔE_{CA} , ΔE_{CG} , ν_{CA} , ν_{CG} and τ_t^{-1} . For the best fitting results, which are shown by the solid and broken lines in figure 2, respectively, the following set of parameters is used:

$$\begin{aligned} \Delta E_{CA} &= 65 \text{ meV}, & \nu_{CA} &= 2.0 \times 10^{13} \text{ s}^{-1}, \\ \Delta E_{CG} &= 185 \text{ meV}, & \nu_{CG} &= 3.0 \times 10^{16} \text{ s}^{-1}, \\ \tau_t^{-1} &= 1.5 \times 10^7 \text{ s}^{-1}. \end{aligned}$$

The activated energy from the $^1T_{1u}$ to $^3T_{1u}$ APESs has been reported for RbBr:Ag⁻ and CsBr:Ag⁻, whose values are 10.7 and 20.4 meV, respectively [17]. These activated energies are smaller than that of 65 meV in NaCl:Ag⁻. The activated energies for RbBr:Ag⁻ and CsBr:Ag⁻ have been estimated only from the increase of the A' luminescence intensity with increasing temperature.

The frequency factor ν_{CG} is three orders of magnitude larger than ν_{CA} . The frequency factor might correspond to the transfer probability at the cross point between two APESs. The larger value of ν_{CG} than ν_{CA} implies that the transference at the cross point from the $^1T_{1u}$ to $^1A_{1g}$ APESs occurs promptly in comparison with that from the $^1T_{1u}$ to $^3T_{1u}$ APESs. This fact may be derived from the difference between the singlet–singlet transfer and the singlet–triplet transfer.

In the temperature region between 30 and 50 K, there is a slight difference between the experimental data of the A' luminescence intensity and the fitting result calculated by equation (7). This difference might suggest the existence of another non-radiative transition process with small thermally activated energy, which is not taken into account in the model shown in figure 5.

4.2. Temperature dependence of the decay time

Next, we consider the kinetics of the luminescence decay of the A' band in NaCl:Ag⁻ crystals. In the Tl⁺-type centres, the temperature dependence of the decay time constant of the A' luminescence has been extensively explained in terms of a three-level model consisting of two excited states $^3T_{1u}$ (level 2) and $^3A_{1u}$ (level 1) and one ground state $^1A_{1g}$ (level 0) [11–14]. The three-level model is shown on the right in figure 5.

According to the three-level model, the A' luminescence takes place from two excited levels to the $^1A_{1g}$ ground state of level 0 with decay rates k_1 ($=\tau_1^{-1}$) and k_2 ($=\tau_2^{-1}$). The

two excited levels are connected with the thermally activated non-radiative transitions, whose probabilities between them are k_{12} and k_{21} . These non-radiative transition probabilities are related by

$$k_{12} = gk_{21} \exp\left(-\frac{\Delta E_T}{k_B T}\right), \quad (8)$$

where g is the ratio of the degeneracy multiplicities of levels 2 and 1. In this case, $g = 2$ is used. Usually, an appropriate approximation $k_{12}, k_{21} \gg k_2$ and $k_2 \gg k_1$ is made. This means that the A' luminescence mainly occurs from level 1 at low temperature and from level 2 at higher temperatures. Under these conditions, the temperature dependence of the decay time constant can be expressed as [12, 13, 19]

$$\tau = \frac{1 + g \exp\left(-\frac{\Delta E_T}{k_B T}\right)}{k_1 + gk_2 \exp\left(-\frac{\Delta E_T}{k_B T}\right)}. \quad (9)$$

The best fitted result for the experimental results is shown by the solid line in figure 4. The set of parameters obtained from the best fitting is as follows:

$$\Delta E_T = 3.7 \text{ meV}, \quad k_1 = 1.2 \times 10^3 \text{ s}^{-1}, \quad k_2 = 1.7 \times 10^5 \text{ s}^{-1}.$$

The separation energy of 3.7 meV is much smaller than those in KCl:TI^+ and KBr:TI^+ , whose values are 55.6 and 60 meV, respectively [13, 14]. The value in NaCl:Ag^- is almost equal to those of the Ag^- and In^+ centres doped in alkali halides [19, 20]. The separation energy between the ${}^3\text{T}_{1u}$ and ${}^3\text{A}_{1g}$ states depends on the strength of the spin-orbit interaction in impurity ions. The larger the atomic number is, the stronger the spin-orbit interaction is. As the atomic number of the In^+ and Ag^- ions is smaller than that of the TI^+ ion, the strength of the spin-orbit interaction in the In^+ and Ag^- centres is weaker than that of the TI^+ centre. The In^+ ion has an outermost $5s^2$ configuration the same as the Ag^- ion and a similar atomic number to the Ag^- ion. Therefore, it is reasonable that the separation energies of the In^+ and Ag^- centres are smaller than that of the TI^+ centre.

The strength of the spin-orbit interaction influences the decay rate k_2 corresponding to the radiative transition probability from the ${}^3\text{T}_{1u}$ state. The decay rate k_1 in NaCl:Ag^- , which corresponds to the radiative transition probability from the ${}^3\text{A}_{1u}$ state, is a similar value to those of the TI^+ and In^+ centres doped in alkali halides [13, 14, 20]. On the other hand, the decay rate k_2 in NaCl:Ag^- is smaller than those of the TI^+ centres and is similar to those of the In^+ centres, because the ${}^3\text{T}_{1u}$ state has the allowed component through the mixing by the spin-orbit interaction. The parameters obtained above reflect the weak spin-orbit interaction system in NaCl:Ag^- .

5. Conclusions

We have measured the temperature dependence of the A' and C' luminescence intensities and the decay time constant of the A' luminescence in NaCl:Ag^- single crystals. At low temperature, the A' and C' luminescence bands are observed at 2.85 and 4.12 eV, respectively. As the temperature increases, the C' luminescence intensity decreases and the A' luminescence grows with compensation for the decrease of the luminescence intensity of the C' band. This temperature dependence implies the existence of the thermally activated non-radiative process from the ${}^1\text{T}_{1u}$ to ${}^3\text{T}_{1u}$ APESs. With increasing temperature above 130 K, the A' luminescence also becomes weak. Then, the decay time of the A' luminescence is almost constant at 10 μs . The fact indicates that the decrease of the A' luminescence intensity above 130 K is attributed

to the reduction of the non-radiative process from the $^1T_{1u}$ to $^3T_{1u}$ APESs. The temperature dependence of the luminescence intensities of the A' and C' bands can be explained by using the rate equations formulated on the basis of the simple configuration coordinate model consisting of the $^1T_{1u}$, $^3T_{1u}$, $^3A_{1u}$ and $^1A_{1g}$ APESs.

The decay time constant of the A' luminescence exhibits a rapid decrease in the temperature range from 7 to 80 K. The three-level model consisting of the $^3T_{1u}$, $^3A_{1u}$ and $^1A_{1g}$ states is adopted to explain the temperature dependence of the decay time of the A' luminescence. The three-level model gives a good agreement with the experimental results.

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