



# Scintillation properties of CsI:Na thin films from viewpoint of nanoparticle formation

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## Abstract

We have investigated scintillation properties of Na-doped CsI (CsI:Na) thin films grown by vacuum deposition. The Na-related luminescence, which is the dominant scintillation of CsI:Na, is activated by heat treatment and associated with formation of NaI nanoparticles. It is found that the Na-related luminescence, which is remarkably degraded in atmosphere, the so-called hygroscopic property, is reversibly recovered in vacuum, while the luminescence due to the self-trapped exciton of CsI hardly decreases in ambient conditions. This finding indicates that the hygroscopic property of CsI:Na results from degradation of NaI nanoparticles embedded in the CsI:Na thin film and that the recovery in vacuum is due to recrystallization. This is a novel proposal to explain the high hygroscopicity of CsI:Na. Furthermore, it is demonstrated from the temperature dependence of the luminescence properties that the energy transfer from the host material of CsI to the NaI nanoparticles is mediated through thermally activated hopping of the self-trapped exciton of CsI.

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## 1. Introduction

Na-doped CsI (CsI:Na) crystals have been widely used for high-efficiency scintillators; however, the scintillation mechanism and properties

are still under debate. Recently, we found that scintillation of CsI:Na thin films prepared by vacuum deposition is activated by the formation of NaI nanoparticles caused by heat treatment [1,2]. This is a key finding for the investigation of the scintillation properties of CsI:Na. It is noted that in previous models the origin of the scintillation has been a self-trapped exciton (STE) of CsI perturbed by a Na<sup>+</sup> ion [3–6]. Since such a

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perturbed STE is usually unstable at temperatures higher than  $\sim 100$  K [7], we argue that the perturbed STE cannot be the origin of efficient scintillation at room temperature. Thus, the scintillation mechanism is now controversial. Furthermore, it is well known that a CsI:Na scintillator is highly hygroscopic, i.e., the scintillation activity is remarkably degraded in the atmosphere. This fact is mysterious because the host material CsI and a related scintillator material of CsI:Tl are slightly hygroscopic. Until now, there has been no explanation for the hygroscopic nature of CsI:Na.

In the present work, we have investigated scintillation properties of CsI:Na thin films prepared by vacuum deposition from the following viewpoints taking account of the nanoparticle formation: (1) the reason for the highly hygroscopic property of CsI:Na and (2) the excitation energy transfer from the host material of CsI to the nanoparticles. Since surfaces are sensitive to ambience and nanoparticle formation, we have adapted thin films as a sample. We have found that Na-related luminescence, which is remarkably degraded in atmosphere, is reversibly recovered in vacuum, while the luminescence due to the STE of CsI hardly changes. This fact will be connected with the degradation of nanoparticles of NaI, one of the most hygroscopic alkali halides, in an ambient atmosphere and the recrystallization in vacuum. Based on temperature dependence of the luminescence properties, we propose a model for the excitation-energy transfer mechanism to the nanoparticles via hopping of the STE of CsI.

## 2. Experimental methods

Samples of CsI:Na thin films were grown on a quartz substrate at room temperature using a conventional vacuum deposition method in high vacuum ( $\sim 1 \times 10^{-4}$  Pa). The source material was a CsI:Na crystal prepared from powders of CsI and NaI by the Bridgman method; the doping concentration of NaI was 0.1 mol%. Powders of CsI:Na, obtained by crushing the bulk crystal, were heated in a crucible for the deposition, where the growth rate was 0.5 nm/s. The film thickness

was fixed at 1.0  $\mu\text{m}$ . Heat treatment of the thin film was performed at 250 °C in high vacuum ( $1 \times 10^{-4}$  Pa) for 2 h. The crystal quality of the samples was characterized by measuring X-ray diffraction patterns. It was confirmed that the thin films are preferentially oriented along the  $\langle 100 \rangle$  axis. The X-ray diffraction patterns were not changed by heat treatment, i.e., the crystal quality of the host material of CsI was not modified [1]. Surface morphology was measured with an atomic force microscope (AFM) (Digital Instruments, NanoScope III). X-ray-excitation luminescence (scintillation) spectra were observed with a prism monochromator (Carl Zeiss, M4QIII), where the spectral resolution was 10 nm at 400 nm, and a  $\text{CuK}\alpha$  line was used for excitation. During measurements of temperature dependence of luminescence, samples were held in a cryostat cooled by liquid nitrogen.

## 3. Results and discussion

At first we discuss heat treatment effects on luminescence properties and surface morphology. Fig. 1 shows X-ray-excitation luminescence spectra of the CsI:Na thin films at room temperature before and after the heat treatment. Moreover, insets show AFM images with scan area of  $700 \times 700 \text{ nm}^2$ . In the thin film before the heat treatment, the luminescence band peaking at 4.1 eV is dominant. In our previous study of a pure CsI crystal [8], we reported that there exist two types of STEs, one with an on-center and one with an off-center configuration, and that the luminescence from the on-center STE at 4.1 eV is the origin of the scintillation at room temperature. Thus, the 4.1 eV band observed in the CsI:Na thin film is attributed to this on-center STE. It is evident from Fig. 1 that the Na-related band peaking at 3.0 eV, which is known to be the dominant scintillation feature in a CsI:Na crystal, is activated by the heat treatment. From the AFM images we can see that the heat treatment causes formation of nanoparticles with various sizes from  $\sim 40$  to  $\sim 80$  nm. Thus, the activation of the scintillation is closely related to the nanoparticle formation. The reason for the nanoparticle

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