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Nuclear Instruments and Methods in Physics Research A 548 (2005) 446–454
NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A
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Scintillation properties of cerium-doped gadolinium silicate with γ-rays from ^{137}Cs

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Received 25 October 2004; received in revised form 9 February 2005; accepted 1 April 2005
Available online 16 June 2005

Abstract

The temperature dependence of the scintillation time profile of a cerium-doped gadolinium silicate (GSO:Ce) crystal has been measured with 662 keV γ-rays from ^{137}Cs between 303 and 464 K. A scintillation mechanism has been assumed where electrons and holes created by radiation are recombined at Ce^{3+} color centers and scintillation light is emitted by the de-excitation of excited Ce^{3+} ions. The pulse shapes have been fitted with a function derived based on this mechanism. The functional form of the pulse shapes was consistent with the data. The obtained decay time constants for the fast and slow components of the de-excitation of excited Ce^{3+} ions have been explained using radiative decay time constants and non-radiative decay time constants. The radiative decay time constant has been evaluated to be 52.0 ± 1.1 ns and the activation energy has been evaluated to be 0.246 ± 0.036 eV for the fast component using the function of the temperature dependence of the non-radiative decay time constant.

PACS: 29.40.Mc; 07.85.Fv

Keywords: Gadolinium silicate; GSO; Scintillation detector; Scintillation mechanism

1. Introduction

Thallium-doped sodium iodide (NaI:Tl) single crystals have been used to detect X-rays and γ-rays for a long time. However, these crystals must be covered to protect against humidity because of the deliquescence of NaI:Tl. Cerium-doped gadolinium silicate (GSO:Ce) has been studied as a scintillation crystal over the past ten years because of (a) the non-deliquescence of GSO:Ce, (b) the light output of GSO:Ce, which is 20% larger than that of NaI:Tl, and (c) the fact that it has almost the same emission wavelength as NaI:Tl [1].

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0168-9002/$ - see front matter
doi:10.1016/j.nima.2005.04.059

The scintillation properties of GSO:Ce have been investigated in various works. The dependence on the density of cerium ions of the light output and decay time constants have been investigated by Melcher et al. [2]. They concluded that a density of cerium of around 0.5 mol% is good for detecting γ-rays. The temperature dependence of the light output of GSO:Ce has been studied by many groups. A temperature range between around 243 and 453 K was considered in the investigation by Melcher et al. [3]. In Refs. [4–6], temperatures below around 333 K were considered. The temperature dependence of light output at temperatures higher than 373 K have not been sufficiently investigated. A study of whole pulse shapes, especially a study of the rising part of the pulse shape, is important to determine the scintillation mechanism. However, no work has attempted to accurately explain pulse shapes, including the rising part of the pulse shape. Therefore, in this work measurements of whole pulse shapes, including the rising part, were taken at temperatures between 303 and 464 K using a digitizing oscilloscope with a sampling rate of 1 GHz. These pulse shapes were analyzed to determine a functional form of the whole pulse shape. γ-rays were used as incident radiation in this work as GSO:Ce crystals are expected to be used as γ-ray detectors in particle and nuclear physics and for medical uses, for example, in positron emission tomography.

The experimental method and the analytic method are reported in Section 2. A model of the scintillation mechanism is described in Section 3, assuming recombination of electron–hole pairs produced by radiation. A functional form of the pulse shape is also obtained in Section 3. The scintillation time profiles are fitted with the pulse shape function obtained in Section 3. The results are shown in Section 4. Decay time constants for fast and slow components of the de-excitation of excited Ce^{3+} ions, obtained from the results of fitting, are evaluated using radiative decay time constants and non-radiative decay time constants. The temperature dependence of the non-radiative decay time constants and the activation energies are obtained. Finally, we conclude the work in Section 5.

2. Experiment and analysis

A GSO:Ce crystal with a cerium concentration of 0.5 mol% was used. The size of the crystal was 1 in. in diameter and it was 1 in. long. The crystal was set in an optical dewar (VPF-700, Janis Research Company), as shown in Fig. 1. Two heaters and two thermocouples were attached near the crystal to control the temperature of the crystal. A photomultiplier tube (PMT) (H3171-04, Hamamatsu) was used to detect the light emitted from the GSO:Ce crystal. A voltage of −1700 V was applied to the PMT. A light guide made from aluminum foil was attached to the GSO:Ce crystal to guide the scintillation light from the crystal to the PMT effectively. The optical dewar was covered with a black curtain to block off the background light. As incident radiation, γ-rays of 662 keV from ^{137}Cs were used. As shown in Fig. 1(b), two lead blocks were put near the ^{137}Cs γ-ray source to prevent γ-rays from hitting the window of the PMT directly and inducing Cherenkov light in it.

Fig. 2 shows the data acquisition hardware. Signals from the PMT were sent to a digitizing oscilloscope, TDS620A (Tektronix). The trigger threshold of the TDS620A was set to −180 mV, for which photo-peak data could be taken effectively. The sampling rate of the TDS620A was set to 1 GHz for digitizing signals. Then, the signal data consisted of a voltage value every 1 ns. LabVIEW, working on a PXI (PCI eXtensions for Instrumentation) controller (PXI-8156B, National Instruments), was used as data acquisition software.

Data were taken at the crystal temperatures of 303, 323, 343, 364, 384, 404, 424, 444 and 464 K, with an accuracy of around 1 K. The pressure inside the optical dewar was set to around 2 Pa during the experiment for thermal insulation. 5000 signals were accumulated at each temperature.

The data have been analyzed on a UNIX workstation. First, noise reduction has been carried out as follows. Noise signals were rejected by using the trigger threshold of the TDS620A when taking the data. However, the data included many signals with very narrow widths around 10 ns. The origin of these signals was Cherenkov