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Branching transport model of NaI(Tl) alkali-halide scintillator

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Abstract

We measure the time dependence of the scintillator light-emission pulses in NaI(Tl) crystals at different temperatures, after activation by gamma rays. We confirm that there are two main nonexponential components to the time decay and find that their amplitude ratio shows Arrhenius temperature dependence. We explain these nonexponential components as arising from two competing mechanisms of carrier transport to the Tl activation levels. The total light output of the NaI(Tl) detectors shows a linear temperature dependence explained by our model.
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1. Introduction

Doped alkali-halide scintillation crystals, developed 50 years ago, continue to be the workhorses of outdoor gamma spectroscopy because of their size, cost, and performance. Recently, there has been a growing interest in NaI(Tl) detectors for use in Homeland Security applications [1,2]. However, a well known, problematic feature of NaI(Tl) detectors is the temperature dependence of their light yield. The core of this problem stems from the temperature dependence of the shape and amplitude of the light pulses emitted from the scintillator for a given energy of the incident ionizing particle. Although multiple exponential components describing the shape of the light pulse in time have been reported [3], the common understanding for an NaI(Tl) light pulse assumes a single dominant exponential component with a temperature-dependent decay-time constant [4–8] based on the effective three-level activator model described in Ref. [9]. This is inconsistent with the well-known fact that below 60 °C, the current pulses cannot be fitted well with a single exponent, even at a fixed temperature. Until now, it has been also generally accepted that the temperature behavior of the total light output of NaI(Tl) is nonlinear and has a broad maximum below room temperature [3–6].

Recent experimental results obtained by using a novel method and device demonstrated that this experimentally observed nonlinear temperature behavior arises because of a convolution between the temperature-dependent shape of the light pulse and the pulse response of the shaping circuitry [10]. It was shown experimentally that when a gated integrator is used, the whole light output shows approximately linear temperature dependence over a wide temperature range (−30 to +60 °C) [10–13]. In the same works, two main components of the shape of the light pulses, with a temperature redistribution between their amplitudes, were found. Importantly, it was shown in Refs. [11–13] that the slow component is negligible above room temperature, but it produces up to 40% of the total light at −20 °C and lasts several microseconds. It was also shown that at high temperatures, only one almost exponential decay component exists, consistent with Ref. [6].

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Several earlier precise measurements [16,17] are consistent with our results showing linear temperature dependence of the total light output, at different temperature intervals, and using different integration times. It is important to note that in those experiments the photomultiplier tube (PMT) was isolated from the crystal and held at a constant temperature. In such way only the temperature dependence caused by intrinsic processes originating from the crystal was measured. We believe that the deviation from approximately linear temperature dependence observed in Ref. [17] below $T = -10\,^{\circ}\mathrm{C}$ is due to not long enough integration time.

Recent precise measurements [18,19] provide data for two different shapers with 2 and 12 μs picking time. Those experiments showed again, that when a longer integrating time is used, the thermal stability of NaI(Tl) is better, and the nonlinearity of the temperature behavior of the total light output is different for different shapers. The authors in Ref. [18] did not recover a linear temperature dependence of the total light output at the whole temperature interval, but as they explained, the reason was they used relatively small integration time of 12 μs, in comparison with the gated integrator used in Ref. [11]. In the same article [18] the authors made detailed and precise measurements of the temperature stability of the single-photoelectron peak in order to exclude the temperature variation of the used PMT [18]. The results of their tests show that the thermal variation of the gain of the dynode structure is almost linear and it is approximately $-0.4\%/^{\circ}\mathrm{C}$. It is consistent with our data where using gated integrator, similar standard Photonis PMT, and almost the same temperature interval, we reported [10–13] approximately $-0.7\%/^{\circ}\mathrm{C}$ linear temperature dependence of the total light output. We interpreted this $-0.7\%/^{\circ}\mathrm{C}$ linear temperature dependence as a combination of the linear PMT's temperature variation reported in Ref. [14] and an approximately linear temperature behavior of the total light output from the scintillator.

Experimentally measured in Ref. [18] current pulses of NaI(Tl) confirmed the presence of different components of the time shape of the light pulse of NaI(Tl) as well as the existence of temperature redistribution between them. However, in Ref. [18], the authors recognized more than two different exponential components of the time shape of the light pulse at almost the whole temperature interval.

We measured precisely the time dependence of NaI(Tl) current pulses at different temperatures. We show that rather than one or more exponential decay components, there are two dominant *nonexponential* light components, corresponding to two competing mechanisms of carrier transport to the Tl activation levels. We model the transport and reproduce the experimental data with nonlinear rate equations. The data may also be fitted with two exponential decay components using fixed fast and slow time constants and an Arrhenius temperature-dependent redistribution between the amplitudes, but the accuracy is approximately 5%.

2. Experimental results

We placed a standard 2-in. × 2-in. Bicron NaI(Tl) detector in an environmental chamber. The chamber temperature changed at a rate of $2\,^{\circ}\mathrm{C/h}$, and each set of measurements at a given temperature was taken after an 8-h hold time to allow good thermal equilibration. We digitized the PMT current pulses with a 14-bit digital scope, manufactured by Gage Applied Sciences Inc., connected via a $50\,\Omega$ resistor directly to the anode of the PMT for each temperature. The temperature coefficient of the PMT [14] cannot change the shape of the light pulse. The area of the pulses was selected to correspond to an excitation around 662 keV. We normalized all pulses to the unit area to study the effect on pulse shape alone. In Fig. 1, we show the time dependence of the emitted light output on a log scale, where the two components are resolved and the temperature-dependent redistribution between the amplitudes of the slow and fast components is readily seen. Note that the emission spectrum of this doped inorganic scintillator shows that the emitted light is at a single wavelength, characteristic of a transition between a single excited state of the activator and its ground state. Thus the two components of the light pulse do not represent decays from different energy levels, but rather, they represent two different pathways for the secondary electrons and holes to the activator levels. We normalized the area of the current pulses to remove the temperature dependence arising from a competition between the scintillation transitions and the nonradiative phonon-assisted transitions. This competition takes place at the excited Tl level, where the electrons and holes recombine [3]. The nonradiative, phonon-assisted transitions decrease the total amount of the collected light, but they play this role after the secondary carriers, electrons and holes, are recombined at the $(Tl^{+})^{*}$ level. The nonradiative, phonon-assisted transitions do not significantly change the time shape of the current pulse. This can readily be understood at very higher temperatures where only one exponential component of the current pulses exists [6] and the time shape of the pulse is almost temperature independent. On the other hand the nonradiative, phonon-assisted transitions are responsible for the strong temperature dependence of the peak position, and this dependence can be observed at arbitrarily higher temperatures. At such high temperatures, the shape of the current pulse does not change with temperature, but the area of the pulse and the corresponding peak position for a given energy of the incident particle becomes less and less with increasing temperature.

In our measurements of the light pulses from NaI(Tl), we found an Arrhenius dependence of the ratio between the amplitudes of the two dominant components with which we fit the data. We found a similar dependence in CsI(Tl) using the numerical data published in Ref. [15], as shown in Fig. 2. We found that

$$\frac{Q_1}{Q_2} \propto e^{(-\Delta E/kT)} \tag{1}$$