Pulsed cathodoluminescence and $\gamma$-luminescence of scintillation crystals

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Abstract. The spectra and decay time of pulsed cathodoluminescence (PCL) of a scintillating crystals excited by the electron beam is compared to the spectra and decay time of the luminescence of the same crystals initiated by $\gamma$-rays (GL). It is shown that spectra and decay time of PCL and GL are identical within the experimental errors. The explanation of these results is based on taking into account the physical processes within the crystal media under the irradiation by high-energy particles. The results of this study confirm that the PCL method may be used for the rapid analysis of the luminescent properties of scintillators.

1. Introduction
Scintillation crystals for gamma-photon detectors are used in a broad spectrum of applications, such as high-energy physics, nuclear physics, medical imaging, well logging, etc. The positron emission tomography (PET) is one of the important domain where the scintillation crystals are widely used. At present, scintillation crystals are tested by the method of $\gamma$-luminescence using radioactive sources. This approach has problems related both to the safe use and storage of radioactive preparations and the necessity of detecting optical radiation of low intensity. Particularly, for the decay time measurement it is necessary to use the single photon counting technique (see, for example, [1]). In our publication we present the results of the studies based on PCL method [2, 3] using a high-power ($\sim 10$ MW) short-time (2 ns) pulsed electron beam with an average particle energy of $\sim 150$ keV. The comparison with the results of GL measurements are also presented.

2. Experimental setup
The experimental setup consists of several autonomous units including a source of luminescence excitation (1), which comprises a pulsed electron accelerator of RADAN-Expert type with an analytical chamber. The unit for PCL spectra measurements (2), consisting of spectrograph OS-1S with multichannel photodetector with LX511 CCD array [4]. The spectral resolution not worse then 2 nm, wavelength range is 400–700 nm. The measured spectra are corrected according to a spectral sensitivity curve of optical system.
Figure 1. Block diagram of the setup for measuring the PCL: (1) PCL excitation unit, (2) multichannel photodetector with OS-1S spectrograph, (3) optical fiber strand, (4) personal computer, (5) monochromator, (6) photomultiplier and (7) digital oscilloscope.

Figure 2. Spectra of YAG:Ce$^{3+}$,Tb$^{3+}$. Solid curve is a GL spectrum, dashed curve is a low-resolution PCL spectrum and dot-dashed curve is a standard PCL spectrum.

Two optical fiber strands (3) are used to transmit the PCL radiation to the unit for the spectra measurements and unit for the decay time measurement. This decision allows to decouple measurement unit from the high-voltage nanosecond luminescence excitation unit.

Block of luminescence excitation is synchronized via electro-optical synchronization line with the spectra measurement unit and personal computer (4).

A block for PCL decay time measurements (5–7) is consisted of prism monochromator DMR-4 (5), photomultiplier FEU-62 (6) and digital oscilloscope Tektronix TDS-210B (7) with passband of 300 MHz. The time resolution of the setup is $\sim$ 15 ns.

For PCL and GL spectra recording the same unit was used. Gamma luminescence was induced by the $^{22}$Na radioactive source with total activity of 900 kBq. This isotope has two decay channels, with 90% cases of positron emission and 10% cases of $\gamma$-quanta with the energy about 1.3 MeV. In the cases of positron emission, the positron immediately annihilates with electron inside the source, with the emission of two 511 KeV $\gamma$-quanta, the energy common in PET applications.

The GL spectra are measured using the spectrograph of the CLAVI setup. Since the $\gamma$-quanta from the radioactive source induce the low intensity luminescence in scintillation crystal, the entrance slit of the spectrograph is set to a maximum width of 5 mm (which led to a decrease in the spectral resolution to 40 nm) and the spectrum is obtained by averaging over 300–400 runs.

For the correct comparison of the PCL spectrum measured at a 2-nm resolution and the $\gamma$-luminescence spectrum measured at a 40-nm resolution, we have performed convolution of the PCL spectrum of 40-nm-wide rectangular step function. The decay times of the GL had been taken from the literature [1, 6–8].

3. Experimental results
In figure 2 the comparison of the PCL and GL spectra of a YAG:Ce$^{3+}$,Tb$^{3+}$ is shown. A solid curve is the GL spectrum, the dashed curve is a low-resolution PCL spectrum after the
Figure 3. Spectra of LFS-3. Solid curve is a GL spectrum, dashed curve is a low-resolution PCL spectrum.

Figure 4. Spectra of CsI:Tl. Solid curve is a GL spectrum, dashed curve is a low-resolution PCL spectrum.

convolution with 40-nm-wide rectangular function and the dot-dashed curve is a standard PCL spectrum. The low-resolution PCL spectra is a standard PCL (with ∼2 nm resolution) spectra after the convolution with rectangular function of 40 nm width. It is also seen from figure 2 that spectrum of GL and low-resolution PCL spectrum of YAG:Ce³⁺,Tb³⁺ are identical within the experimental error. The comparison of GL spectrum (solid curve) and low-resolution PCL spectrum (dashed curve) of LFS-3 crystal [9] is presented on figure 3 and spectra of CsI:Tl crystal are presented on figure 4. You can see from figures 2 and 3 that the PCL spectra and GL spectra are also identical in case of CsI:Tl and LFS-3 crystals.

In our experiment the decay times of PCL are measured for Lu₂SiO₅ (LSO) and Lu₁.₈Y₀.₂SiO₅ (LYSO) crystals. PCL decay times of all crystals were equal to 45 ± 7 ns. In the literature the GL decay times for LSO [6, 7] and LYSO [8] crystals are published. The table 1 shows the comparison between PCL and GL decay times of these crystals. “Dop.” in table 1 implies the type of dopant and “Conc.” implies the concentration of dopant. The decay times of PCL and GL of LSO and LYSO crystals are equal within the experimental errors.

4. Discussion

In case of interaction of high energy particles, γ-quanta or electrons, with the crystal media the dominant process is the atom ionization with the emission of characteristic x-ray photons. The impact on luminescence spectra of Compton scattering and electron-positron pairs production in case of γ-quanta or bremsstrahlung [2, 10, 11] in case of electrons is relatively small. Thus, despite of difference between the primary processes (involving initial particles), the secondary processes are always characterized by the ionization and excitation of medium by secondary electrons and the absorption of x-ray photons in the volume of the sample.

The luminescence results from processes involving electrons with energies on the order of a bandgap width [2], thus we should not expect differences in the PCL and GL γ-luminescence spectra.

This result shows that Pulsed Cathodo Luminescence technique can be used for rapid analysis of luminescent properties of crystals for detection of γ-radiation.

The PCL technique have two reasonable advantages: intensity of PCL is much higher (4–5
Table 1. Comparison of PCL and GL decay times for the scintillators of similar type. Errors are dominant by systematic.

| Crystal type | Pulsed cathodoluminescence | Chemical formula | Dop. conc., % | Decay time, ns | γ-luminescence | Chemical formula | Dop. conc., % | Rad. source | Decay time, ns |
|--------------|----------------------------|------------------|---------------|---------------|----------------|------------------|---------------|-------------|---------------|
| LSO          | Lu₂SiO₅                    | Ce³⁺             | 0.15          | 45 ± 7        | Lu₂SiO₅        | Ce³⁺             | 0.05          | ¹³⁷Cs       | 40            |
|              |                            | Ce³⁺             | 0.15          | 45 ± 7        |                | Ce³⁺             | 0.22          | ¹³⁷Ce       | 46–47         |
|              |                            |                  |               |               |                | Ce³⁺             | —             | ¹⁵¹Ce       | 40–50         |
|              |                            | Ce³⁺             | 0.15          | 45 ± 7        |                |                  | —             | ¹⁰⁹Cd       | 99            |
|              |                            | Sc³⁺             | 0.6           |               |                |                  | —             | ⁵⁷Co        | 50            |
| LYSO         | Lu₁₁.₈Y₆.₄SiO₅             | Ce³⁺             | 0.075         | 45 ± 7        | Lu₁₁.₈Y₆.₄SiO₅ | Ce³⁺             | —             | ¹³⁷Cs       | 50.1          |

orders of magnitude) then the GL intensity and there is no problems concerning with storage and usage of radioactive sources.

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