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Scintillation Properties of SrF_2 and SrF_2–Ce^{3+} Crystals

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Abstract—This Letter presents the results of measuring scintillation properties of pure SrF_2 crystals and crystals activated by various concentrations of Ce^{3+} ions. The light yield of these materials is compared to that of the known scintillators NaI–Tl and CaF_2–Eu^{2+}. Strontium fluoride crystals activated with Ce^{3+} ions are found to be characterized by high light yield and to be promising materials for use in scintillation detectors employed for γ-ray well logging.

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Scintillators are currently widely applied in different fields of physics, medicine, and geology. They are widely used in sensors in airports and freight terminals. Because of this, the demand for scintillation materials is growing, which makes it necessary to search for new materials with better characteristics. The highest light yield is observed for the crystals of iodides, bromides, and chlorides activated with cerium and europium ions [1, 2]. The drawbacks of these materials consist in high cost and hygroscopicity. Furthermore, the scintillation decay time in materials activated with europium is ~1 μs and these scintillators are characterized by low temperature stability of light yield, which limits their use (e.g., in geology) [3].

For most applications, it is sufficient that the light yield be comparable to that of a NaI–Tl scintillator, but scintillators have a shorter decay time and are not hygroscopic. In this connection, alkaline earth fluoride crystals activated with Ce^{3+}, Pr^{3+}, and Eu^{2+} ions may be rather promising materials. The ultimate light yield for these materials is approximately 50000 photons/MeV [1]. The maximum light yield was attained in CaF_2–Eu^{2+} crystals: 18000–24000 photons/MeV. A lower light yield (10000 photons/MeV) was observed in BaF_2 and BaF_2–Ce^{3+} crystals [2]. The results of studying the scintillation properties of SrF_2 and SrF_2–Ce^{3+} crystals are first presented in this Letter.

SrF_2 and SrF_2–Ce^{3+} crystals were grown with the Stockbarger technique in vacuum. Cadmium fluoride was added to remove traces of oxygen from the charge [4]. The resulting samples contain no oxygen impurity, which can be seen from the absence of characteristic bands in the absorption spectrum of the crystals [5]. SrF_2–Ce^{3+} with activator concentration varied from 0.03 to 3 mol % were obtained.

The X-ray luminescence (XRL) spectra were recorded after excitation with a Pd anode X-ray tube; the tube voltage and current were 35 kV and 0.8 mA, respectively. The spectra were recorded in the photon-counting mode; an FEU-39a and VM-4 monochromator were used.

The amplitude spectra of the pulses were measured using an FEU-39a, a preamplifier, and an Ortec 570 spectroscopy amplifier. The pulse shaping time of the spectroscopy amplifier was set equal to 10 μs in order to register as much scintillation light as possible. The ^{137}Cs calibration source was used for excitation. The sample under study was wrapped in four layers of UV-reflecting Teflon PTFE tape and placed directly on the FEU window. To ensure the best optical contact, glycerol lubricant was applied between the FEU window and the scintillator.

The scintillation decay time of Ce^{3+} in SrF_2–Ce^{3+} crystals was measured after the excitation with a ^{137}Cs calibration source. The decay curves were recorded with a Rigol DS-1202CA oscilloscope.

Figure 1 shows the XRL spectra of SrF_2, NaI–Tl, SrF_2–0.3 mol % Ce^{3+}, and CaF_2–0.1 mol % Eu^{2+} crystals. In the spectrum of SrF_2, a broad band with the maximum at 280 nm is associated with luminescence of excitons. In the strontium fluoride crystal activated by cerium ions, luminescence of excitons is quenched and disappears at activator concentrations above 1 mol %. The most intense bands in the XRL spectrum of the SrF_2–Ce^{3+} crystal with maxima at 310 and 325 nm are associated with the $5d$–$4f$ luminescence of Ce^{3+} ions. The light yield in these samples can be estimated by comparing the areas below the luminescence spectra of the samples. The inset in Fig. 1 shows the concentration dependence of the light yield of crystals activated with different concentrations of cerium ions (0.03–3 mol %) standardized to the light

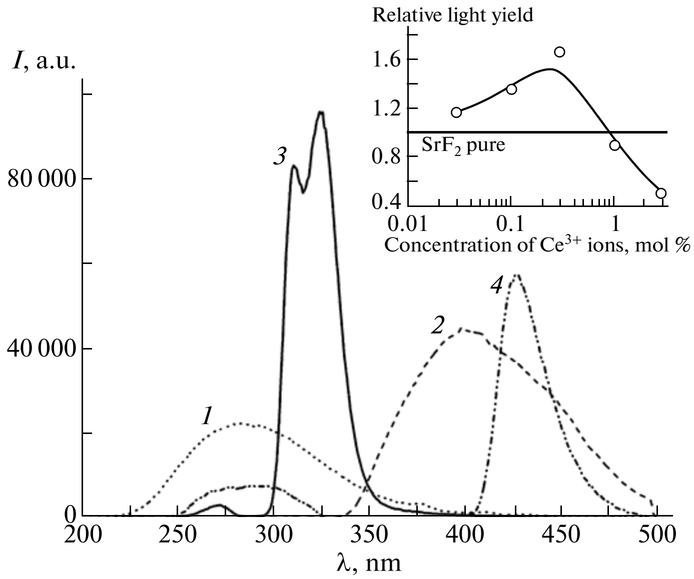

Fig. 1. Luminescence spectra of SrF_2 (curve *1*), NaI–Tl (curve *2*), SrF_2–0.3 mol % Ce^{3+} (curve *3*), and CaF_2–0.1 mol % Eu^{2+} (curve *4*) crystals excited by X-rays. The inset shows the light yield for excitation by X-rays as a function of concentration of Ce^{3+} ions in SrF_2 crystals. The light yield of pure SrF_2 crystal is taken as 1.

yield of pure SrF_2 crystal. The maximum light yield is observed in the crystals activated with 0.3 mol % of Ce^{3+} ions.

The light yield of luminescence in SrF_2 and SrF_2–Ce^{3+} crystals excited by X-rays was compared with the yield of luminescence in NaI–Tl crystals. The light yield of the NaI–Tl crystal is 43000 photons/MeV; hence, the light yield of the rest of the crystals under study excited by X-rays can be determined. The resulting data are listed in the table. The estimated light yield for CaF_2–0.1 mol % Eu^{2+} crystal is 21500 photons/MeV, which is in close agreement with the published data [2]. For SrF_2, the light yield is 20640 photons/MeV; for SrF_2 crystals activated with 0.3 and 1 mol % Ce^{3+} ions, it is ~34000 and 18500 photons/MeV, respectively.

Figure 2a shows the amplitude spectra of pulses in SrF_2, SrF_2–0.3 mol % Ce^{3+}, and NaI–Tl crystals. The channels of amplitude distributions are plotted along the *X* axis. The inset shows the dependence of the light yield measured based on the position of the total absorption peak on concentration of Ce^{3+} ions in SrF_2. The light yield of pure SrF_2 crystal is taken as unity. The energy resolution of the total absorption peak for NaI–Tl is 6.7%; for SrF_2, it is 10%; and for the SrF_2–0.3 mol % Ce^{3+}, it is 9.3%. The light yield of pure SrF_2 crystal is 42% of NaI–Tl, which is close to the values obtained from the XRL spectra (see table). The light yield for SrF_2 crystals activated with 0.3 mol % Ce^{3+} ions is equal to 32% of the yield for NaI–Tl, which is considerably lower than the values obtained from the XRL spectra. These data are listed in the table below. It should be mentioned that the table lists values that have not been corrected for the spectral sensitivity of the registration channel. However, the spectral sensitivity of a S20 photocathode (FEU-39a) at 400 nm is known [6] to be higher than at 280–330 nm.

Figure 3 shows the curve of scintillation decay in SrF_2–0.3 mol % Ce^{3+} crystal. An integrating oscillo-

Light yield of SrF_2, SrF_2–Ce^{3+}, NaI–Tl, and CaF_2–0.1 mol % Eu^{2+} crystals measured for excitation by X- and γ-rays

Scintillator	Light yield derived from X-ray luminescence spectra		Light yield derived from the amplitude spectra of pulses	
	rel. units	photons/MeV	rel. units	photons/MeV
NaI–Tl	1	43000	1	43000
CaF_2–0.1 mol % Eu	0.5	21500	0.44	18920
SrF_2	0.48	20640	0.42	18060
SrF_2–0.3 mol % Ce^{3+}	0.79	33970	0.32	13760
SrF_2–1 mol % Ce^{3+}	0.43	18490	0.2	8600

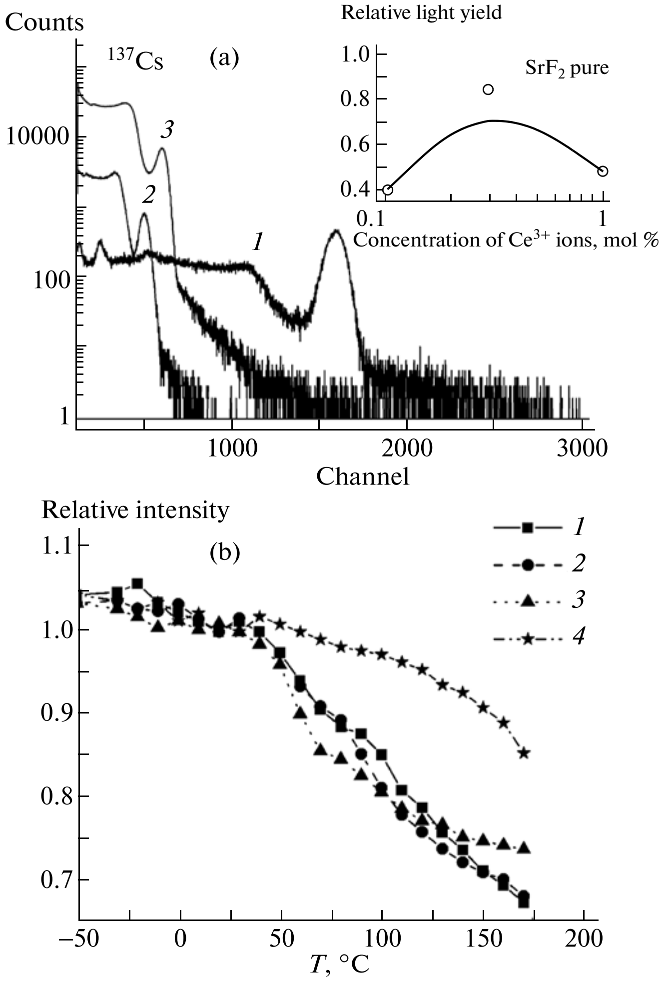

Fig. 2. Amplitude spectra of the pulses of NaI–Tl (curve *1*), SrF_2–0.3 mol % Ce^{3+} (curve *2*), and SrF_2 (curve *3*) crystals after excitation with the ^{137}Cs gamma source (662 keV) (a) and temperature dependence of the light yield of SrF_2 crystals activated with 0.01 mol % Ce^{3+} (curve *1*), 0.1 mol % Ce^{3+} (curve *2*), 0.3 mol % Ce^{3+} (curve *3*), and 1 mol % Ce^{3+} (curve *4*) (b). The inset shows the light yield of SrF_2–Ce^{3+} crystals as function of concentration.

scope input impedance of 2.6 kΩ was used to detect the slow-decay components of luminescence of Ce^{3+} ions. The time resolution of the recording system for the input impedance of 2.6 kΩ is ~2.8 μs. Thus, the first component in luminescence decay (2.8 μs) is the integrated short component, which is equal to 130 ns at an input resistance of 50 Ω. It can be seen from Fig. 3 that the contribution of slow components to luminescence of SrF_2–0.3 mol % Ce^{3+} crystal can reach 50%.

Hence, the difference in light yields derived from the XRL spectra and the amplitude spectra of the pulses is connected with the fact that luminescence of cerium ions contains intense slow components (Fig. 3)

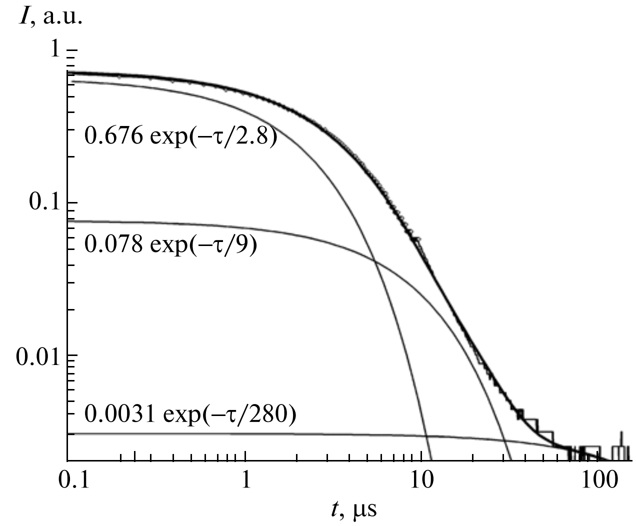

Fig. 3. Scintillation decay curve of SrF_2 crystal measured after the excitation with the ^{137}Cs gamma source (energy 662 keV). The exponential components to which the total curve is deconvoluted are also shown.

contributing to the light yield. When measuring the amplitude spectra of the pulses, the pulse shaping time is 10 μs, which is why most light is not detected. When measuring the XRL spectra, the integration time is ~1 s.

The practical results of the study are as follows. The temperature stability of the light yield (Fig. 2b) was determined according to the procedure described in [7]. In the temperature range from –50 to 50°C, the light yield of SrF_2–Ce^{3+} crystals is temperature-independent. At a temperature above 50°C, the light yield decreases with temperature. For the crystals activated with 0.01 mol % Ce^{3+} (Fig. 2b, curve *1*) and 0.1 mol % Ce^{3+} (Fig. 2b, curve *2*), the light yield at temperature 170°C decreases by 30%; for the samples with concentration 0.3 mol % Ce^{3+}, by 25% (Fig. 2b, curve *3*). In crystals containing 1 mol % Ce^{3+} impurity, the light yield decreases by 15% at 170°C (Fig. 2b, curve *4*). Thus, significant temperature stability of the light yield in the temperature range from –50 to 170°C is demonstrated by SrF_2–0.3 mol % Ce^{3+} and SrF_2–1 mol % Ce^{3+} crystals, which makes SrF_2–Ce^{3+} crystals promising materials for use as scintillators in sensors for γ-ray well logging.

Being characterized by a light yield comparable to that of NaI–Tl, SrF_2 crystals possess higher temperature stability of light yield, density (4.18 g/cm^3) as compared to NaI–Tl (3.67 g/cm^3) and are nonhygroscopic. The potential light yield of SrF_2–Ce^{3+} crystals may reach 34000 photons/MeV. Thus, SrF_2 and SrF_2–Ce^{3+} are promising crystals for scintillation applications.

In terms of their parameters (relatively high light yield, nonhygroscopicity, temperature stability of the light yield), SrF_2–0.3 mol % Ce^{3+} crystals are a promising material for scintillation sensors used in γ-ray well logging.

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REFERENCES

1. P. Dorenbos, IEEE Trans. Nucl. Sci. **57**, 1162 (2010).
2. S. E. Derenzo, http://scintillator.lbl.gov.
3. D. V. Ellis and J. M. Singer, *Well Logging for Earth Scientists*, 2nd ed. (Springer, Dordrecht, 2007).
4. E. Radzhabov, J. Phys.: Condens. Matter **13**, 10955 (2001).
5. A. V. Egranov and E. Radzhabov, *Spectroscopy of Oxygen and Hydrogen Impurity in Alkali-Halide Crystals* (Nauka, Novosibirsk, 1992) [in Russian].
6. S. O. Flyckt, *Photomultiplier Tubes* (Photonis, Brive, France, 2002).
7. R. Shendrik and E. Radzhabov, IEEE Trans. Nucl. Sci. **57**, 1295 (2010).

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