# Absolute Light Yield Measurements on $SrF_2$ and $BaF_2$ Doped With Rare Earth Ions

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***Abstract*—Results of absolute light yield measurements on strontium and barium fluoride doped with $PrF_3$ and $CeF_3$ are presented and compared with light yield of well-known scintillators s(NaI-Tl and CsI-Tl). For pure $SrF_2$ crystal, we obtain the value of about 29,200 photons/MeV.**

***Index Terms*—Absolute light yield, fluorides, light output, scintillator, strontium fluoride.**

## I. INTRODUCTION

MEASUREMENT of the absolute light yield of a scintillator is complicated especially in ultra-violet and far ultra-violet region of the spectra. Many different factors such as quantum efficiency of photomultiplier tube (PMT), optical transmission of PMT window, and optical coupling compounds should be taken into account.

Methods of absolute light yield measurements were reported in literature [1]–[7]. However, fluoride scintillators ($CaF_2$, $SrF_2$, and $BaF_2$) are rarely investigated contrary to such wide used scintillators as sodium and cesium iodides doped with thallium ions (NaI-Tl and CsI-Tl), which have been measured in a greater amount of papers. The first spread fluoride scintillator was $CaF_2$-Eu, which absolute light yield was evaluated as 24,000 photons/MeV [1]. The second one was barium fluoride crystal. It is a relative dense material, its decay of scintillation can be resolved into two components: the fastest decay time about 0.8 ns and slower one about 600 ns. Absolute light yield of $BaF_2$ and $BaF_2$-Ce crystals was reported by [2], [8]–[10]. Typical one of pure $BaF_2$ (decay constant 600 ns) was about 9500-10000 photons/MeV [2], [10]. Light yield of fast luminescence was evaluated about 1500 photons/MeV [2], [8]. Also light yield of $BaF_2$ doped with $Ce^{3+}$ was about 13000 photons/MeV [9].

Absolute light yield of $SrF_2$, $SrF_2$-Ce, and $SrF_2$-Pr crystals has not been measured yet. Nevertheless, preliminary evaluations give light yield of pure $SrF_2$ about 30,000 photons/MeV [11].

Fluoride crystals doped with $Pr^{3+}$ and $Ce^{3+}$ ions are prospective scintillators for well-logging applications [11]–[13] that has stimulated interest in study of the crystals. The aim of this paper was to measure the absolute light yield of pure and $Ce^{3+}/Pr^{3+}$ doped $SrF_2$ and $BaF_2$ crystals. Measurements were made for the samples of pure and doped with $Pr^{3+}$ and $Ce^{3+}$ ions in concentrations of 0.01, 0.1, 0.3, and 1 mol.%. The given concentrations of impurities were chosen because crystals doped with higher than 1 mol.% concentrations of $Pr^{3+}/Ce^{3+}$ ions showed dramatically low light yields [14], [15]. Light yield of well-known scintillators of CsI-Tl and NaI-Tl were also measured. Photoelectron yield is determined from comparison full-energy peak of scintillator with single electron response. The measured numbers of photoelectrons were converted into photon number using the quantum efficiency of the PMT as specified by the manufacturer.

## II. EXPERIMENTAL METHODOLOGY

Crystals of CsI-Tl ($10 \times 10 \times 10\,mm^3$) and NaI-Tl ($40 \times 40 \times 40\,mm^3$) were given by "Crystal", Usol'e-Sibirskoe, Russia. Fluoride crystals were grown using the Stockbarger method in graphite crucible in vacuum with 1% of $CdF_2$ added as a scavenger for oxygen containing impurities [20]. No fluoride crystals contained oxygen that was controlled by absence of oxygen centers luminescence in visible spectral region under UV-excitation. Oxygen ions form $RE^{3+}$-$O^{2-}$ and $O^{2-}$-$V_a^+$ dipoles in fluorides, where $RE^{3+}$ is rare-earth ion and $V_a^+$–anion vacancy. If a crystal contains oxygen defects, strong emission in the blue-green spectral range will appear [21], [22].

The pure crystals show a large optical transmission without absorption bands caused by unwanted impurities. The presence of rare earth ions in the crystals is proved by absorption spectra measured with a Perkin-Elmer Lambda 950 spectrophotometer. Bands in UV spectral region in the $Pr^{3+}$ and $Ce^{3+}$ doped crystals attribute to 4f-5d transitions in the rare-earth ions (Fig. 1). Absorption coefficient depends on concentration of the activator ions.

Luminescence of investigated fluoride crystals lies in UV spectral region. Therefore, photomultiplier with quartz windows is required. The most measurements of absolute light yield of the fluorides were carried out with PMT Photonis 2020Q. A photomultiplier tube Enterprises 9814QSB with comparable characteristics is used in our measurements. The quantum efficiency of photocathode is specified by manufacturer and given in Fig. 2(b). The PMT demonstrates smaller decrease of the sensitivity in 250-300 nm spectral region than a PMT 2020Q. The PMT was operated with a CSN638C1

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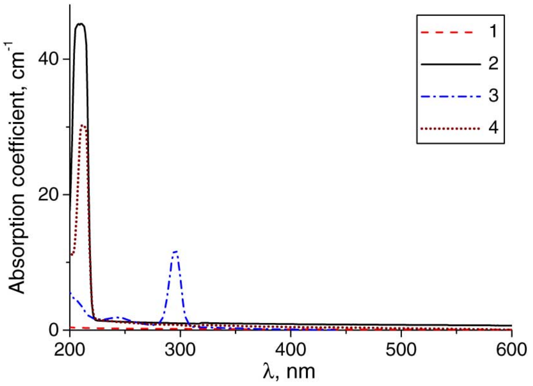
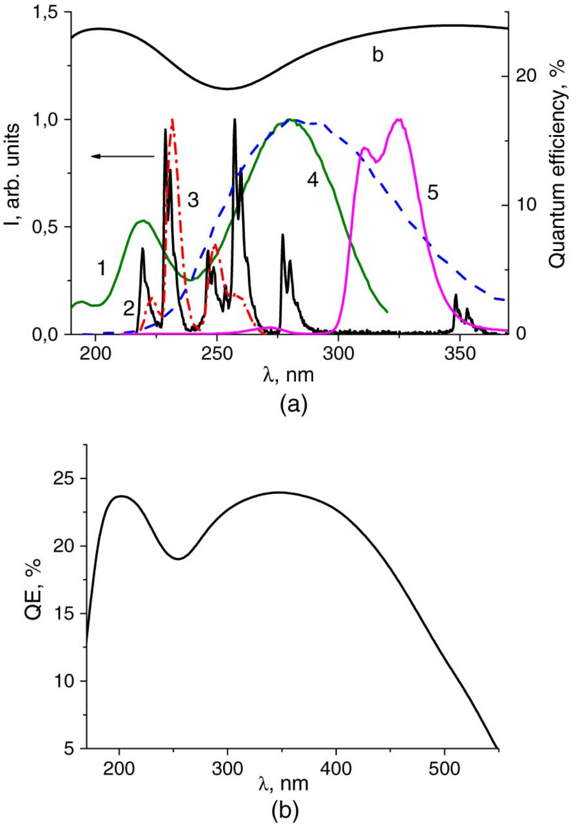


Fig. 1. Absorption spectra of pure SrF$_2$ (curve 1), BaF$_2$-0.1 mol.% Pr$^{3+}$ (curve 2), SrF$_2$-0.01 mol.% Ce$^{3+}$ (curve 3), and SrF$_2$-0.05 mol.% Pr$^{3+}$ (curve 4) crystals.



(a)

(b)

Fig. 2. Normalized emission spectra of pure BaF$_2$ (curve 1), BaF$_2$-Pr$^{3+}$ (curve 2), SrF$_2$-Pr$^{3+}$ (curve 3), pure SrF$_2$ (curve 4), and SrF$_2$-Ce$^{3+}$ (curve 5) shown in the inset (a) are compared with quantum efficiency of PMT 9814QSB (curve b and inset b).

negative polarity voltage chain [23]. The focusing system of 46 mm active diameter is assumed 100% photoelectron collection efficiency in the center of the photocathode. Measurements of photoelectron number are made three times to calculate the errors of the measured numerical quantities.

A sample is irradiated with gamma rays from a monoenergetic $\gamma$-ray source of $^{137}$Cs (E $=$ 662 KeV). A homemade preamplifier and an Ortec 570 amplifier are used to obtain the single electron and crystal pulse height spectra. $^{137}$Cs pulse height spectrum of samples is determined for shaping times of 10 $\mu$s. The coarse gain control is used to switch between the single photoelectron pulse height spectrum and the $^{137}$Cs pulse height spectrum. The gain scale on the amplifier is linear. The samples are coated with PTFE tape to maximize the light collection efficiency and optically coupled to the window of PMT using glycerin grease.

X-ray excited luminescence is performed using X-ray tube with Pd anode operating at 35 kV and 0.8 mA. The spectra are recorded at photon-counting regime using PMT FEU-39A and vacuum grating monochromator VM-4.

## III. RESULTS AND DISCUSSION

### A. *X-ray Excited Emission*

Luminescence spectra of the crystals are given in Fig. 2. In the spectrum of SrF$_2$ a wide band at 280 nm is attributed to self-trapped exciton (STE) emission (Fig. 2, curve 4) [24]. In the luminescence spectrum of pure BaF$_2$ wide band corresponded to STE at 270-280 nm and a band peaked at 220 nm related to core-valence transitions [25] are observed (Fig. 2, curve 1).

In Ce$^{3+}$-doped fluorides STE luminescence is quenched. The most intense bands in x-ray luminescence spectra of SrF$_2$-Ce$^{3+}$ crystals at 310 and 325 nm correspond to 5d-4f emission of Ce$^{3+}$ ions (Fig. 2, curve 5). In Pr-doped crystals several bands observed in 220–360 nm spectral region are attributed to 5d-4f transitions in Pr$^{3+}$ ions [26].

### B. *Photoelectron Yield Measurements*

The photoelectron yield ($Y_{phe}$) is calculated by comparing the 662 keV photopeak in the pulse height spectrum and the average pulse height in the single electron pulse height spectrum of the PMT (Table II, column 5). Fig. 3 shows pulse height spectra of pure and rare-earth doped fluorides, NaI-Tl and CsI-Tl crystals. The photopeaks corresponding to the $^{137}$Cs energy photon are seen in each curve in Fig. 3.

Crystals CsI-Tl and NaI-Tl were used as testing scintillators with well-known photoelectron light yields ($Y_{phe}$). Photoelectron yield of the NaI-Tl crystal (8,500 photoelectrons/MeV) compares with the value of about 8,900 photoelectrons/MeV reported by M. Moszynski *et al.* [19]. For $Y_{phe}$ of the CsI-Tl, we obtain a value of about 4,900 photoelectrons/MeV which is close to the yield 4,400 photoelectrons/MeV reported in [6].

Photoelectron yield of the pure BaF$_2$ (1,930 photoelectrons/MeV) is comparable to the value 2,110 photoelectrons/MeV given by Dorenbos *et al.* [2]. The largest one among the pure SrF$_2$ samples is about 6,010 photoelectrons/MeV in the polished crystal. The cleaved sample shows about 30% lower photoelectron yield.

After rare-earth doping, photoelectron yields of the fluoride crystals are decreased. We obtain the largest yield about 2,200 photoelectrons/MeV among Pr-doped SrF$_2$ in the crystal doped 0.15 mol.% of the activator. The lowest yield (220 photoelectrons/MeV) is measured in the crystal doped with 1 mol.% of Pr$^{3+}$ ions.

Pr-doped BaF$_2$ crystals demonstrate lower $Y_{phe}$ than the ones measured in SrF$_2$-Pr crystals. Photoelectron yield of BaF$_2$-0.15 mol.% Pr$^{3+}$ small crystal (10 × 2 mm) is about

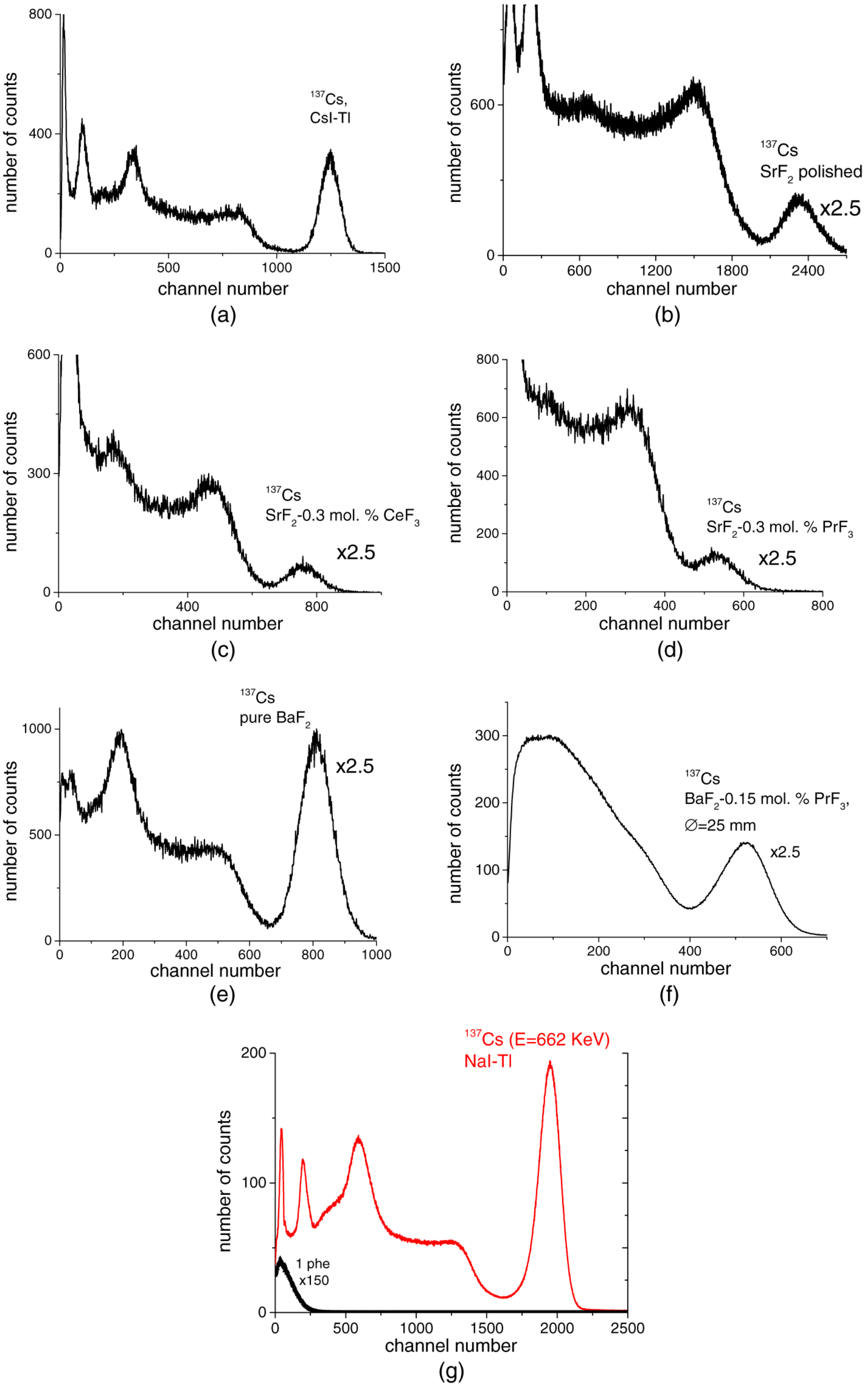


Fig. 3. A comparison of the energy spectra of $\gamma$-rays from a $^{137}$Cs source measured with different scintillators ((a) CsI-Tl; (b) pure $\mathrm{SrF_2}$; (c) $\mathrm{SrF_2}$-0.3 mol.% $\mathrm{Ce^{3+}}$; (d) $\mathrm{SrF_2}$-0.3 mol.% $\mathrm{Pr^{3+}}$; (e) pure $\mathrm{BaF_2}$; (f) $\mathrm{BaF_2}$-0.15 mol.% $\mathrm{Pr^{3+}}$; (g) NaI-Tl and single electron peak).

1,500 photoelectrons/MeV. Large crystal doped with the same concentration of the activator shows a lower yield.

### C. Photon Yield Measurements

To calculate the photon yield per MeV from the photoelectron yield/MeV, a correction must be made for the transmittance of the optical coupling compound, reflectivity losses, and the quantum efficiency of the employed PMT. To obtain the absolute photon yield $Y_{ph}$ we use the equation [27]:

$$Y_{ph} = Y_{phe}\frac{1 - R_{eff}}{0.98 \cdot QE_{eff}} \tag{1}$$

where $QE_{eff}$ is the integral quantum efficiency given in Table II, $R_{eff}$ is the PMT effective reflectivity, and $Y_{phe}$ is photoelectron yield.

TABLE I
TESTED CRYSTALS

| Sample | $\rho$ [g/cm$^3$] | Temperature response, %/°C | Refractive index @ emission max[a] | Cleavage plane | Size [mm] | Manufacturer |
|---|---|---|---|---|---|---|
| NaI-Tl | 3.67 | -0.3 [17] | 1.83 | <100> | cylindrical, Ø40x40 | ”Crystal”, Usol’e-Sibirskoe, Russia |
| CsI-Tl | 4.51 | -0,5 [18] | 1.79 | none | cubic, 10x10 | ”Crystal”, Usol’e-Sibirskoe, Russia |
| $BaF_2$ | 4.88 | -0.8 [1] | 1.50 | <111> | cubic, 25x25 | IGC SB RAS, Irkutsk, Russia |
| $BaF_2$-$Pr^{3+}$ | 4.88 | -0.25 [1] | 1.51 | <111> | cylindrical, Ø10x2<br>cylindrical, Ø25x25 | IGC SB RAS |
| $SrF_2$ | 4.18 | -0.7 [1] | 1.46 | <111> | cubic, 10x10 | IGC SB RAS |
| $SrF_2$ [b] | 4.18 | | 1.46 | <111> | cylindrical, Ø10x10 | IGC SB RAS |
| $SrF_2$-$Ce^{3+}$ | 4.18 | | 1.46 | <111> | cylindrical, Ø10x10 | IGC SB RAS |
| $SrF_2$-$Pr^{3+}$ | 4.18 | -0.08 [2] | 1.50 | <111> | cylindrical, Ø10x2 | IGC SB RAS |

[a] Data were given by [16]
[b] Cleaved sample

TABLE II
COMPILATION OF INTEGRAL QUANTUM EFFICIENCIES, WAVELENGTH OF LUMINESCENCE MAXIMA, PHOTOELECTRON ($Y_{phe}$) AND ABSOLUTE PHOTON ($Y_{ph}$) YIELDS, AND FULL WIDTH AT HALF MAXIMUM (FWHM) AT 662 KEV GAMMA-RAY ENERGY ($^{137}$CS SOURCE) OF TESTED SCINTILLATORS IN COMPARISON WITH LITERATURE PHOTOELECTRON YIELD DATA

| Scintillator | Primary decay time [ns] | Wavelength of emission max [nm] | Integral quantum efficiency $QE_{eff}$ | Photoelectron yield $Y_{phe}$ [photoelectrons/MeV] | Reference data $Y_{phe}$ [photoelectrons/MeV] | Photon yield $Y_{ph}$ [ph/MeV] | FWHM[a] @ 662 KeV [%] |
|---|---|---|---|---|---|---|---|
| NaI-Tl | 250 | 415 | 0.21 | 8500±600 | 8900±200 [19] | 38000±2700 | 8 |
| CsI-Tl | 1000 | 540 | 0.07 | 4900±390 | 4400±130 [6] | 55700±4400 | 7.1 |
| $BaF_2$ | 600 | 280 | 0.21 | 1930±120 | 2110±70 [5] | 9400±600 | 13 |
| $BaF_2$-0.15 mol.% $Pr^{3+}$ | 21 | 228; 257 | 0.20 | 1230±80 | | 6300±400 | 23 |
| $BaF_2$-0.15 mol.% $Pr^{3+}$ | 21 | 228; 257 | 0.20 | 1500±100 | | 7700±500 | 19 |
| $SrF_2$ | 1000 | 285 | 0.21 | 6010±420 | | 29200±2000 | 10 |
| $SrF_2$ [b] | 1000 | 285 | 0.21 | 4020±280 | | 19500±1400 | 13 |
| $SrF_2$-0.3 mol.% $Ce^{3+}$ | 130[c] | 310; 325 | 0.23 | 2100±150 | | 9300±700 | 13 |
| $SrF_2$-0.15 mol.% $Pr^{3+}$ | 25 | 232; 250 | 0.19 | 2200±150 | | 11800±800 | |
| $SrF_2$-0.3 mol.% $Pr^{3+}$ | 25 | 232; 250 | 0.19 | 1300±90 | | 7000±500 | 20 |
| $SrF_2$-1 mol.% $Pr^{3+}$ | 24 | 232; 250 | 0.19 | 220±20 | | 1200±100 | |

[a] Full width at half maximum
[b] Cleaved sample
[c] The crystal has long decays [3]

In considering absolute light yield, the integral quantum efficiency is an important characteristic that has to be calculated taking account emission spectrum of a scintillator. Integral quantum efficiency values are given in Table II. Almost all values of quantum efficiency lie in 0.19-0.23 interval. Emission of CsI-Tl crystal is observed in green spectral region, where sensitivity of the PMT is dropped. Therefore, integral quantum efficiency for CsI-Tl crystal is 0.07.

The light yield of the scintillator should be corrected for the reflectivity of the photocathode. A part of the light which is reflected from the photocathode is lost in quantum efficiency measurement procedure. In the measurement with the crystal having full optical contact due to grease the reflected light can be collected back into the photocathode. Therefore, the effective quantum efficiency is increased. The largest effective reflectivity for PMT 9814QSB is observed in green spectral range. The reflectivity in CsI-Tl luminescence region is about 21%. For NaI-Tl the reflectivity is given 8%. In UV spectral region no light is reflected from photocathode and $R_{eff}$ is less than 1%. The parameters of effective reflectivity were specified by the PMT manufacturer.

Absolute light yields of the tested samples (NaI-Tl, CsI-Tl, and $BaF_2$) are similar to known results [2], [3], [28].

The largest light yield 29200 photons/MeV in fluoride crystals is found for pure $SrF_2$ crystals (Table II, column 7) that is compared with our earlier results [11].

The fluoride crystals doped with trivalent rare-earth ions show lower light yields than ones in the pure crystals (probably, due to ineffective energy transfer mechanism). The most favorable energy transfer in fluoride crystals is a resonance STE transfer [9], [14]. This mechanism takes place at room temperature only in the crystals doped with $Ce^{3+}$ ions [26]. Drawback of exciton transfer is that following luminescence is slower than the one in case of electron-hole energy transfer. However, decay time constant becomes shorter with increasing of $Ce^{3+}$ ions concentration due to extension of probability of resonance STE to Ce-ion energy transfer [11].

The shortest decay time component of the Ce-luminescence under gamma-ray excitation equals to 130 ns in $SrF_2$-0.3 mol.% $Ce^{3+}$ and it becomes longer with decrease of Ce concentration [11]. Decay time of $Ce^{3+}$ ions luminescence under optical excitation is about 30 ns [9], [29], [30]. Under vacuum ultraviolet excitation at exciton and higher energies regions the decay of Ce-doped fluorides became nonexponential [9], [31]. Observed decay constant (130 ns) can be ascribed to resonance energy transition in the nearest pairs of exciton and cerium ion [14], [29]. Electron-hole energy transfer was also found in Ce-doped fluoride crystals. Delayed energy transfer, when electron or hole is captured by trap instead of direct recombination in Ce-ions, is dominant [11]. In Ce-doped crystals eventual light yield could reach 35,000 photons/MeV, but a large part of emitted light is not registered in pulse height measurements due to presence of long time components (about tens and hundreds of microseconds) in $Ce^{3+}$ ions emission [11].

Primary luminescence mechanism in $Pr^{3+}$ doped crystals consists of consecutive capture of electron forming $Pr^{2+}$ center and then hole with following recombination. Apart from this fast electron-hole recombination, delayed energy transfer process involving hole traps also takes place in Pr-doped crystals. To sum up, at room temperatures two complete processes are found. However, efficiency of the second process is higher in the fluoride crystals [13]. Therefore, the light yield of Pr-doped crystals is lower than one of the pure samples.

## IV. CONCLUSION

In the present paper results of measurements of absolute light yields on $SrF_2$, $BaF_2$ doped with various concentrations of $Ce^{3+}$ and $Pr^{3+}$ ions are reported. Pure strontium fluoride crystals demonstrate the highest light yield among fluorides. Crystals doped with $Ce^{3+}/Pr^{3+}$ ions have lower light yield than the pure samples.

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