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SCINTILLATORS FOR CRYOGENIC APPLICATIONS: STATE-OF-ART

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The latest results on the low-temperature characterisation of scintillation materials selected for rare event search are reviewed. The temperature dependences of the light output and decay time are analysed on the basis of the current understanding of the underlying physical processes that control the emission of light in solids. It is shown that scintillation properties of the materials under study are adequate for the purpose of cryogenic experiments searching for rare events. From the analysis of the performance characteristics it is concluded that currently $ZnWO_4$ is the most suitable scintillation target for dark matter search.

Key words: cryogenic scintillators, light yield, decay time, tungstates, molybdates, sapphire, bismuth germanate, zinc selenide.

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I. INTRODUCTION

During the past decade a new generation of hybrid cryogenic phonon-scintillation detectors (CPSD), combining powerful background discrimination with remarkable energy threshold and resolution, has been developed and introduced into experiments [1–3]. The demonstration of successful operation of CPSD opened exciting possibilities for rare-event searches. It is now widely agreed that CPSDs are ideal to for reaching the sensitivity levels required by future experiments searching for dark matter [4] and double-beta decay [5, 6]. This in turn gave a strong impetus to the studies of scintillation properties of materials at cryogenic temperatures. There has been a swift increase in research activities and the development of scintillator materials for cryogenic rare event search experiments that resulted in a number of publications [7–20] as well as designated workshops (CryoScint [21] and RPScint [22]).

There is number of selection criteria for the cryogenic scintillators to be satisfied in order to meet requirements of cryogenic rare event search experiments. A high scintillation yield in the milli Kelvin temperature range and low intrinsic radioactivity are the most important parameters that define the ultimate sensitivity of the detector. Other important physical properties are chemical stability of surface and thermodynamic characteristics of lattice (low specific heat and high Debye temperature). Finally, given the prospects of future experiments having a target mass of 100 kg and more, the cost of the material can not be excessive.

In this paper we discuss the latest progress achieved in research and development of scintillation materials for these applications, both in the material preparation and in the understanding of the scintillation mechanisms. The results obtained through studying of temperature dependences of scintillation characteristics of the materials selected so far for rare event searches will be presented. Systematic studies of these properties are shown to be a highly-effective method for understanding the origins of the basic performance limits of known materials. The advancing of this knowledge should lead to new or improved compounds for a new generation of cryogenic scintillation detectors.

II. EXPERIMENT

The measurements of scintillation characteristics of materials over a wide range of temperature were carried out using the multiphoton counting technique (MPC). A detailed description of the method is given elsewhere [23, 24]. The measurements were carried out in a ^{4}He-flow cryostat using an ^{241}Am α-source (5.5 MeV). For compatibility of the experimental results we measured samples of the same dimensions ($5 \times 5 \times 1$ mm^3) and maintained a fixed geometry of the experiment. Provided these conditions are fulfilled, the accuracy of the measurements is assessed to be within 10%. The error on the relative measurements of the light output for different materials is ca. 30%, as it includes additionally the errors of the measurements of luminescence spectra and the spectral sensitivity of PMT.

The experimental samples discussed in this study were obtained from different sources. $CaWO_4$, $CaMoO_4$ and $ZnWO_4$ are the off-cuts of Czochralski-grown crystals used for the production of cryogenic scintillation detectors. They where supplied by SRC Carat, Lviv, Ukraine ($CaWO_4$ and $CaMoO_4$) and the Institute for Scintillation Materials, Kharkiv, Ukraine ($ZnWO_4$). The Institute for Scintillation Materials also provided the samples of $ZnMoO_4$ and $MgWO_4$ and supplied commercial samples of $Bi_4Ge_3O_{12}$, ZnSe and Al2O$_3$-Ti. The samples of $CdMoO_4$ and $CdWO_4$ where received from Hilger Crystals (Margate, UK).

III. RESULTS AND DISCUSSION

$CaWO_4$ is currently actively used in a cryogenic experiment searching for dark matter [3, 25]. The first studies of the temperature variation of scintillation properties of $CaWO_4$ as well as many other materials were done over the 350–9 K range using MPC [23]. Since the temperature dependant processes affecting the emission of solids (energy transport and thermal quenching) appear not to change significantly with temperature below ∼10 K it is expected that the results obtained at this temperature can provide instructive information on the major scintillation characteristics at millikelvin temperatures. Although quite consistent with the fundamentals of solid state physics, this intuitive notion needed experimental proof that has been eventually delivered when scintillation properties of $CaWO_4$ were investigated down to 0.020 K [11].

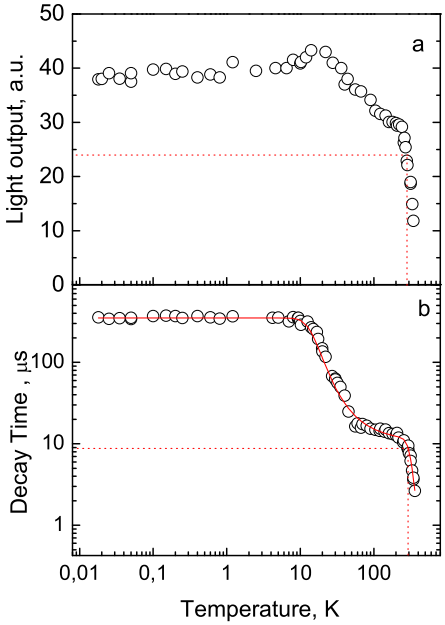

Fig. 1. (a) The temperature dependence of the scintillation light output of $CaWO_4$ measured at excitation with 5.5 MeV α-particles from ^{241}Am. (b) The temperature dependence of the long scintillation decay time constant of $CaWO_4$. The solid curve displays the result of the best fit to the experimental data using the three-level model (Eq. 2). Parameters of the fit are: $k_1 = 3.0{\cdot}10^3$ s^{-1}, $k_2 = 1.1{\cdot}10^5$ s^{-1}, D=4.4 meV, $K = 8.6 \cdot 10^9$ s^{-1}, $\Delta E = 320$ meV. The dotted lines indicate the value of scintillation parameters at $T = 295$ K.

Figure 1a shows the variation of scintillation light output of $CaWO_4$ over the 0.020–350 K temperature range. At high temperatures (> 250 K) the light output is dominated by thermal quenching: as the probability of non-radiative decays increases strongly with temperature, the emission intensity decreases. From 20 to 250 K only a small reduction in the light output is observed. The enhancement observed at ∼ 20 K can be assigned to a contribution from the radiative recombination of carriers captured by shallow traps. In the region between 0.02 and 10 K the light output remains constant within the error limits. This is the most important practical finding of this study, as it provides an explicit demonstration of the constancy of the light yield in the operating temperature range of CPSDs.

The temperature dependence of the decay time constant of $CaWO_4$ is shown in figure 1b. The scintillation decay curve is usually presented as a sum of two or three exponential components as following:

$$f(t) = y_0 + \sum_i A_i \exp\left(-t/\tau_i\right) \qquad (1)$$

where y_0 is background, A_i and τ_i are amplitude and decay time constant.

The decay kinetic of $CaWO_4$ and its temperature dependence was analysed within the framework of a simple three-level model of the emission centre with one metastable level. Using the differential equations that describe the dynamics of population of the levels in the case of thermal equilibrium we derived the following expression for the temperature dependence of the decay time constant:

$$\frac{1}{\tau} = \frac{k_1 + k_2 \exp\left(-D/kT\right)}{1 + \exp\left(-D/kT\right)} + K \exp\left(-\Delta E/kT\right) \qquad (2)$$

where k_1 and k_2 are the probabilities of radiative decay from levels 1 and 2 separated by an energy gap D, K is the probability for the non-radiative decay rate and ΔE is the energy barrier of the non-radiative quenching process. As can be seen from Fig. 1b, Eq. (2) gives an adequate fit for the experimental results. Thus, the model offers a sensible approximation, allowing to describe the features of the decay kinetics and to obtain the parameters of the relaxed excited state of the emission centre in $CaWO_4$.

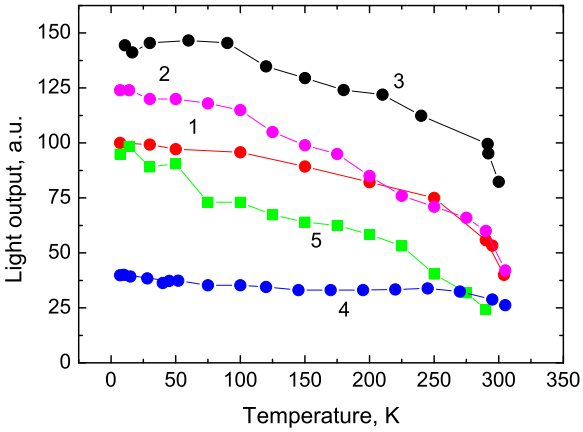

Fig. 2. Temperature dependence of the scintillation light output in $CaWO_4$ (1), $ZnWO_4$ (2), $CdWO_4$ (3), $MgWO_4$ (4) and $CaMoO_4$ (5), measured at excitation with 5.5 MeV α-particles from ^{241}Am.

The variation of scintillation light yield of several tungstate and molybdate crystals in the temperature range 8–310 K is displayed in figure 2. The relative light yield was derived from pulse height spectra by correcting for the spectral sensitivity of the photodetector. Table 1 summarises the results of our studies of the main scintillation properties of these as well as other potentially interesting materials. The measured dependences show a very similar trend in all crystals under study. In tungstates, at first, the light yield rapidly increases as the temperature is lowered to $T\sim$ 250–280 K. Below this temperature the light yield exhibits moderate changes and at $T < 100$ K it flattens.

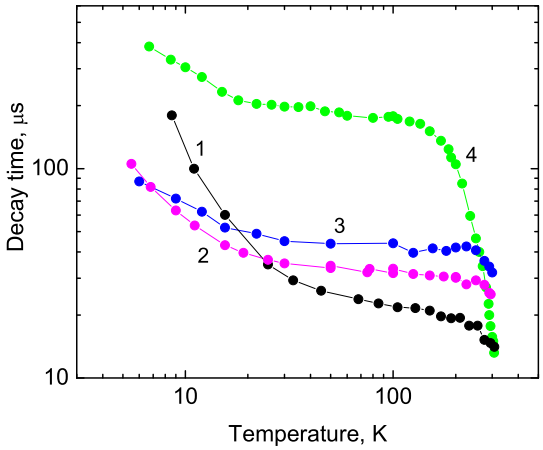

Fig. 3. Temperature dependence of the long scintillation decay time constant of $CdWO_4$ (1), $ZnWO_4$ (2), $MgWO_4$ (3) and $CaMoO_4$ (4) measured at excitation with 5.5 MeV α-particles from ^{241}Am.

It should be noted that at low temperatures $ZnWO_4$ and $CdWO_4$ exhibit superior scintillation efficiency in comparison with calcium tungstate. The relative light output of optimised samples is about 120 and 145 % with respect to $CaWO_4$ (see Table 1). The molybdates exhibit qualitatively the same features but the temperature of the thermal quenching is considerably lower. $CaMoO_4$ is the only compound exhibiting a practically useful light yield at room temperature, about 55 % that of $CaWO_4$ [26].

Figure 3 shows the scintillation decay time constant of wolframite tungstates and $CaMoO_4$ measured over the 8–310 K temperature range. The long scintillation decay time constant changes by more than an order of magnitude with cooling to cryogenic temperatures. In the high temperature range the kinetics of the radiative decay is dominated by thermal quenching and the scintillation decay constant rapidly increases with cooling. Below the roll-off temperature the non-radiative processes are less efficient and the increase of the decay time constant is less pronounced. The character of the scintillation kinetics changes abruptly below 20 K: it demonstrates considerable slowing of the decay. As is demonstrated for $CaWO_4$, this is due to the fine energy structure of the emitting centre in tungstates and molybdates that constitutes a metastable level a few meV below the emitting one. The scintillation decay time constants of the crystals studied at temperatures of 295 K and 9 K are listed in Table 1.

Another classical example of an intrinsic scintillator is $Bi_4Ge_3O_{12}$ that has been extensively examined at room temperatures. Recent studies of Moszynski et al [27] indicated a significant (factor of three) increase of the light yield at $T = 77$ K. We measured the temperature dependence of the light output and the decay time of $Bi_4Ge_3O_{12}$ from room temperature down to 6 K [15]. The variation of the scintillation light output with temperature demonstrates the features that are common to intrinsic scintillators. As the temperature decreases to 100 K the scintillation light output of the crystal increases steadily while at lower temperature, however, the scintillation response remains virtually constant (see fig. 4a). The total light output of the scintillator increases by a factor of $\sim$ 3.5 from $T = 295$ K to 6 K. The estimate based on the absolute room temperature value of scintillation efficiency [27] gives a light yield of $Bi_4Ge_3O_{12}$ at 6 K equal to 23700±2600 ph/MeV, which is 150% of $CaWO_4$ (see table 1).

Crystal	Density, g/cm^3	Emission peak, nm	Decay time*, μs (α-particle excitation)		Light output, (relative to $CaWO_4$ at 9 K)**	
			295 K	9 K	295 K	9 K
$CaWO_4$	6.06	420	9	360	55	100
$MgWO_4$	5.66	480	36	90	30	40
$ZnWO_4$	7.87	490	24	110	60	120
$CdWO_4$	7.90	480	14	180	80	145
$CaMoO_4$	4.35	540	16	380	30	95
$CdMoO_4$	60.7	550	—	460	—-	80
$Bi_4Ge_3O_{12}$	7.13	480	0.43	138	45	150
Al_2O_3-Ti	3.98	290; 430; 740	0.15; . . . ; 3	. . .	15	30
ZnSe	5.42	640	—	8	—	120

* — the long decay time constant;

** — light output in units proportional to the number of emitted photons

Table 1. Scintillation properties of crystals for cryogenic rare event searches.

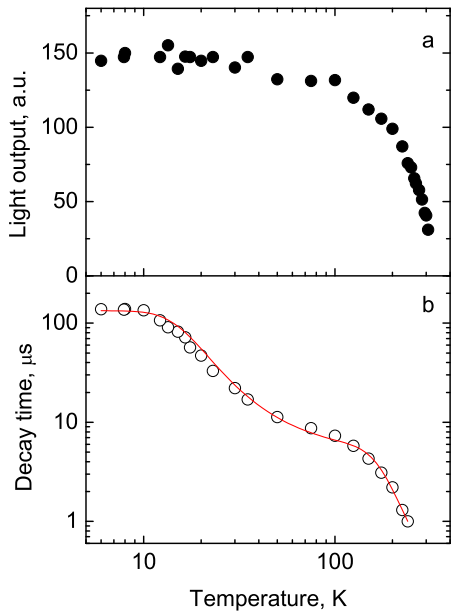

Fig. 4. (a) Temperature dependence of the scintillation light output of $Bi_4Ge_3O_{12}$. (b) Temperature dependence of the long scintillation decay time constant of $Bi_4Ge_3O_{12}$. The solid curve displays the result of the best fit to the experimental data using the three-level model (Eq. 2). The parameters of the fit are: $k_1 = 7.5 \cdot 10^3$ s^{-1}, $k_2 = 4.5 \pm 0.3 \cdot 10^5$ s^{-1}, $D = 6.4 \pm 0.5$ meV, $K = 1.3 \pm 0.2 \cdot 10^8$ s^{-1}, $\Delta E = 105 \pm 4$ meV.

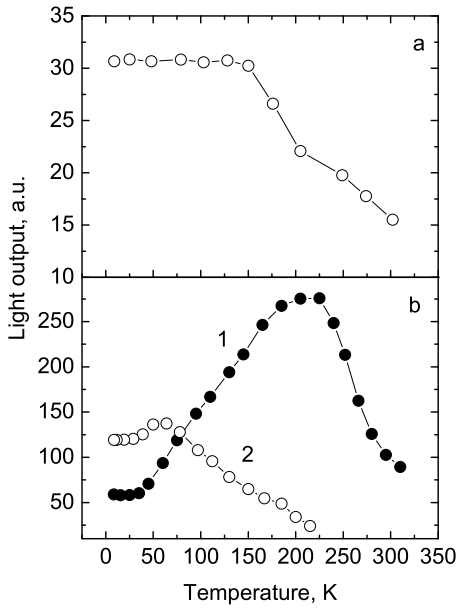

Fig. 5. Temperature dependence of the scintillation light output in Al_2O_3–Ti (700 ppm). (b) Temperature dependence of the scintillation light output in ZnSe–Te (1) and pure ZnSe (2).

Figure 4b shows the variation of the long decay time constant with temperature. The character of the temperature dependence of the decay time constant observed for $Bi_4Ge_3O_{12}$ is typical for the three-level model of the emitting centre constituting a ground and two excited levels of which the lower one is metastable. The luminescence of $Bi_4Ge_3O_{12}$ is attributed to the radiative transition within Bi^{3+}, and given the electron configuration of the excited state of Bi^{3+}, it can be understood by referring to the model discussed for $CaWO_4$. The emission at low temperatures originates from the $^3P_0$ metastable level, while at higher temperature transitions occurs mainly between the excited $^3P_1$ and the ground $^1S_0$ states. The model predicts the existence of the low-temperature plateau in the $\tau = f(T)$ dependence and the gradual decrease of the decay time constant with temperature, caused by establishing a thermal equilibrium between the two emitting levels. The shortening of the decay time constant τ with increasing temperature T can be understood as the result of a thermally-activated re-population of excited states and their non-radiative de-excitation.

Al_2O_3–Ti and ZnSe are interesting materials to study different scenarios of interaction with dark matter and double beta decay. The temperature change of the light output in Al_2O_3–Ti is shown in fig. 5a. The light output increases gradually with cooling and remains constant below the roll-off temperature which is 150 K. The temperature changes are controlled by the processes of thermal quenching. The total increase of the light output of Ti-doped Al_2O_3 at cooling to 9 K is by a factor of two [16, 28]. The nominal concentration of titanium was found to be between 50 and 100 ppm; for this concentration the low-temperature light output of the Al_2O_3–Ti-doped sapphire is assessed to be 30% of $CaWO_4$.

We also measured the temperature dependences of the light output in Te-doped and pure ZnSe. As can be seen from fig. 5b there is a marked dissimilarity in the dependences. This indicates differences in the mechanisms that control the scintillation process in pure and doped crystals at different temperatures. In doped crystals, the scintillation efficiency is governed to a major extent by the efficiency of the energy transfer from the host to the emitting centres. Thus, the light output of ZnSe–Te first increases with cooling (stage of thermal quenching) then it decreases below $T = 225$ K and, finally, below $T = 50$ K it remains constant. The decrease of the light output of ZnSe–Te below the critical temperature of 225 K suggests that with cooling the probability of the energy transfer reduces, illustrating hereby a characteristic feature of doped scintillators. In contrast, pure ZnSe exhibits a temperature dependence of the light output which is typical for self-activated scintillators. According to fig. 5b the light output of ZnSe at low temperature is very close to what ZnSe-Te exhibits at room temperature. Based upon the room temperature value of the absolute scintillation yield of ZnSe–Te, 28300 ph/MeV [29] we estimated that the relative light yield of ZnSe at 9 K is in excess of $CaWO_4$ (see Table 1). The results of our low-temperature characterisation of zinc selenide evidenced that this material has very good prospects for cryogenic applications. This has been recently confirmed by successful test result of CPSD with ZnSe scintillator [30,31].

IV. CONCLUSION

In this work we summarised the low-temperature scintillation properties for a number of scintillation materials suitable for application in cryogenic rare event search. Taking into account the high light output of $ZnWO_4$ at cryogenic temperatures it is suggested that this material is most promising for dark matter searches [32]. Furthermore, zinc tungstate, which has been produced as a commercial scintillator for a while, has reached maturity and the material composition favours low intrinsic radioactivity. It is also expected that $CaWO_4$ as well as $CaMoO_4$ could be the constituent parts of a multi-target absorber in a future experiments.

Low-temperature scintillation studies were carried out for a selected group of other promising compounds. It is demonstrated that $Bi_4Ge_3O_{12}$, ZnSe and Ti-doped Al_2O_3 are very appealing materials for cryogenic applications due to their high scintillation output at low temperatures. The feasibility to use these materials as CPSDs opens a possibility to explore many interesting scenarios of rare-event interactions.

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[1] A. Alessandrello *et al.*, Phys. Lett. B **420**, 109 (1998).
[2] C. Bobin *et al.*, Nucl. Instrum. Methods Phys. Res. A **386**, 453 (1997).
[3] P. Meunier *et al.*, Appl. Phys. Lett. **75**, 133 (1999).
[4] H. Kraus *et al.*, Nucl. Phys. B- Proc. Suppl. **173**, 168 (2007).
[5] L. Gironi *et al.*, Opt. Mater. **31**, 1388 (2009).
[6] J. W. Arnaboldi *et al.*, arXiv:1005.1239v1 [nucl-ex].
[7] V. B. Mikhailik, H. Kraus, J. Imber, D. Wahl, Nucl. Instrum. Methods Phys. Res. A **566**, 522 (2006).
[8] S. Pirro *et al.*, Phys. At. Nucl. **69**, 2109 (2006).
[9] H. Kraus, V. B. Mikhailik, D. Wahl, Radiat. Meas. **42**, 921 (2007).
[10] F. A. Danevich *et al.*, Phys. Status Solidi A **205**, 335 (2008).
[11] V. B. Mikhailik, H. Kraus, S. Henry, A. J. B. Tolhurst, Phys. Rev. B **75**, 184308 (2007).
[12] A. N. Annenkov *et al.*, Nucl. Instrum. Methods Phys. Res. A **584**, 334 (2008).
[13] L. L. Nagornaya *et al.*, IEEE Trans. Nucl. Sci. **56**, 2513 (2009).
[14] P. C. F. Di Stefano *et al.*, J. Low Temp. Phys. **151**, 902 (2008).
[15] J. Gironnet *et al.*, Nucl. Instrum. Methods Phys. Res. A **594**, 358 (2008).
[16] M. Luca *et al.*, Nucl. Instrum. Methods Phys. Res. A **606**, 545 (2009).
[17] H. Kraus *et al.*, Phys. Lett. B **610**, 37 (2005).
[18] I. Bavykina *et al.*, Opt. Mater. **31**, 1382 (2009).
[19] L. L. Nagornaya *et al.*, IEEE Trans. Nucl. Sci. **56**, 994 (2009).
[20] F. A. Danevich *et al.*, Nucl. Instrum. Methods Phys. Res. A **608**, 107 (2009).
[21] P. C. F. Di Stefano, F. Petricca, Opt. Mater. **31**, 1381 (2009).
[22] Proceedings of the 1st Int. Workshop "Radiopure Scintillators for EURECA" (RPScint'2008) arXiv:0903.1539v2 [nucl-ex].
[23] H. Kraus, V. B. Mikhailik, D. Wahl, Nucl. Instrum. Methods Phys. Res. A **553**, 522 (2005).
[24] V. B. Mikhailik, H. Kraus in Proceedings of the 1st Int. Workshop "Radiopure Scintillators for EURECA" (RPScint'2008) arXiv:0903.1539v2 [nucl-ex] p. 64.
[25] G. Angloher *et al.*, Astropart. Phys. **31**, 270 (2009).
[26] V. B. Mikhailik, S. Henry, H. Kraus, I. Solskii, Nucl. Instrum. Methods Phys. Res. A **583** 350 (2007).
[27] M. Moszynski *et al.*, IEEE Trans. Nucl. Sci. **51**, 1074 (2004).
[28] V. B. Mikhailik *et al.*, Nucl. Instrum. Methods Phys. Res. A **546**, 523 (2005).
[29] M. Balcerzyk *et al.*, Nucl. Instrum. Methods Phys. Res. A **482**, 720 (2002).
[30] I. Dafinei *et al.*, IEEE Trans. Nucl. Sci. **57**, 1470 (2010).
[31] C. Arnaboldi *et al.*, arXiv:1006.2721v2.
[32] H. Kraus *et al.*, Nucl. Instrum. Methods Phys. Res. A **600**, 594 (2009).

СУЧАСНИЙ СТАН СПРАВ У ДІЛЯНЦІ СЦИНТИЛЯТОРІВ ДЛЯ КРІОГЕННОГО ЗАСТОСУВАННЯ

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У статті узагальнено результати останніх досліджень низькотемпературних властивостей сцинтиляторів, придатних для застосування у кріогенних експериментах з пошуку рідкісних подій. На основі сучасних уявлень про природу фізичних процесів, що контролюють випромінювання світла у твердих тілах, проаналізовано температурні залежності світловиходу та константи загасання люмінесценції ряду сцинтиляторів. Показано, що сцинтиляційні властивості досліджених кристалів задовольняють вимоги кріогенних експериментів з пошуку рідкісних подій. Із порівняльного аналізу ключових властивостей зроблено висновок, що $ZnWO_4$ є найпридатнішим сцинтилятором для пошуку темної матерії.