Fast inorganic scintillators – status and outlook

Rainer W. Novotny

2nd Physics Institute, Justus-Liebig-University, Heinrich-Buff-Ring 16, 35392 Giessen, Germany

r.novotny@exp2.physik.uni-giessen.de

Abstract. Scintillation detectors based on inorganic materials represent one of the most widely applied instrumentation techniques in the fields of nuclear and high-energy physics as well as medical or industrial applications. Driven by experimental requirements the research has focused onto a faster response, shorter decay times and higher compactness implementing high-Z ions. The discovery of the fast core-valence luminescence in BaF_2, the allowed electric dipole transitions in Ce^{3+}, and the short radiation lengths of BGO and $PbWO_4$ have set important milestones. However, excellent time resolution is affected as well by the density of photoelectrons produced at the early stage of the signal generation and the integral light output. The paper will give a detailed overview of the present status on fast inorganic scintillators, their performance and the theoretical limitations on achievable energy and time resolutions. The results are illustrated by various applications in research. The state of the art scintillator material will be discussed based on new materials such as $LaBr_3$ or $LaCl_3$, which were doped with rare earth ions such as Ce^{3+}-centers to reach decay times between 20 and 40ns, respectively.

1. Introduction

For decades, scintillation detectors based on inorganic materials have become one of the most widely applied instrumentation techniques in physics – in particular in the fields of nuclear and high-energy physics. Their discovery and development are strongly correlated with the experimental needs in basic research and technology in physics. Visual counting of the discovered X-rays or natural radioactivity became possible with $BaPt(CN)_4$, $CaWO_4$ or ZnS at the end of the 19th century. The first construction of scintillation detectors, made possible by the development of the photomultiplier tube [1], started with the discovery of activated and pure alkali halide crystals. NaI(Tl) and CsI(Tl), introduced by Hofstadter [2,3], provide since more than 60 years efficient photon and particle detection. The continuous increase of the energy range of the probes to be detected directed crystal development towards faster response, shorter decay times and higher compactness implementing high-Z ions. In particular, the discovery of the fast core-valence luminescence in BaF_2 [4,5], the allowed electric dipole transitions in Ce^{3+} and the short radiation lengths of BGO [6] or $PbWO_4$ [7] set important milestones in the last decades of the 20th century to find or even engineer the ideal scintillator.

In particular, the evolution in nuclear, medium and high-energy physics illustrates the impact on the detector development by asking for complex detector systems to cope with multi-particle production and high photon multiplicities. In high-energy physics detector technology had to be adapted in a similar way.

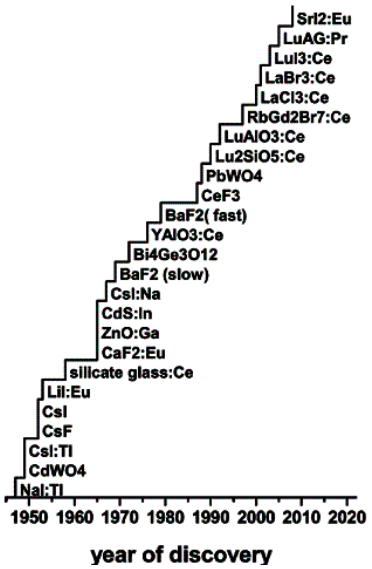

Figure 1. Development of inorganic scintillators starting with the availability of electronic photo sensors.

2. Basic properties of inorganic scintillators

The overall luminescence yield is one of the most selective criteria for the selection of a scintillator material. This holds in particular for nuclear structure studies or medical imaging when low energetic photons have to be detected with high resolution. A phenomenological approach leads to the following estimate of the primary light yield Y of scintillation photons per deposited energy given in MeV:

$$\mathbf{Y} = \frac{\mathbf{E}_\gamma}{\beta \cdot \mathbf{E}_g} \cdot \mathbf{S} \cdot \mathbf{Q} \tag{1}$$

where $\beta \cdot E_g$ is the mean energy necessary for the formation of one electron-hole pair in a medium with a forbidden zone of width E_g and E_γ is the absorbed energy. S describes the efficiency of the energy transfer to the luminescent center and Q the quantum yield of the intra-center luminescence. Assuming S and Q to be close to 100% the light yield will depend inverse proportionally on the energy E_g.

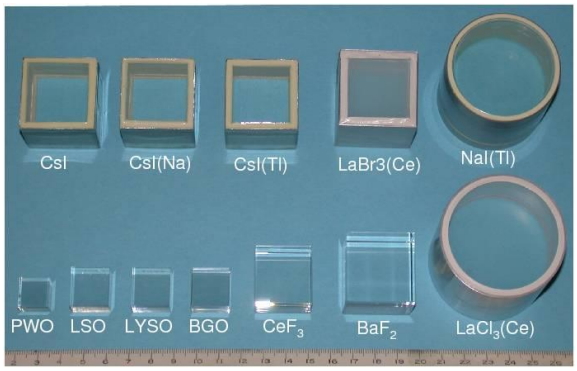

Figure 2. Comparison of different cubic scintillator materials shown with dimensions of $1.5X_o$. The picture is taken from [8].

However, the energy resolution finally achieved with the complete detector is a complex interplay between statistical fluctuations within the above formula, the light collection efficiency and the performance and matching of the photo sensor. The different mechanisms of interaction for neutral and charged particles within the detector medium and the completeness of energy absorption have to be included on top. Due to high count rates, coincidence requirements or particle identification via

time-of-flight technique time resolution is another criteria for scintillator selection. Additional factors including the rise and decay time of the scintillation process, fluctuations of the photon transit time from the emission point within the crystal to the photo sensor and fluctuation within the photo detector affect the timing performance. Therefore, most of the present applications focus on the search for bright and fast scintillators limiting the variety of optional luminescence centers either intrinsic or created by dopants.

The detection of electromagnetic (EM) probes relies on the measurement of the energy and impact location, both with high resolution. In particular, the reconstruction of the invariant mass of neutral mesons, which decay preferentially into photon pairs, requires the simultaneous measurement of energy and the relative angle between both photon directions. Besides a full geometrical coverage with high efficiency the absorption of the total photon energy in the active material is mandatory. The asymptotic cross sections of the elementary processes determine the radiation length X_o depending on the composition of the material. The subsequently created pairs of electrons and positrons, which undergo bremsstrahlung at energies above a material specific critical value, close the circle by generating again high-energy photons. In this process, leptons loose their energy on a logarithmic scale and lead to a lateral spread of the secondary products due to multiple scattering, which is quantified geometrically by the Molière-radius (R_M). The interplay of these processes leads to the formation of the EM shower. The present accelerator technology in medium- and high-energy physics aiming for higher beam energies and luminosities has forced the scintillator development towards extremely compact, fast and radiation hard crystals (see figure 2).

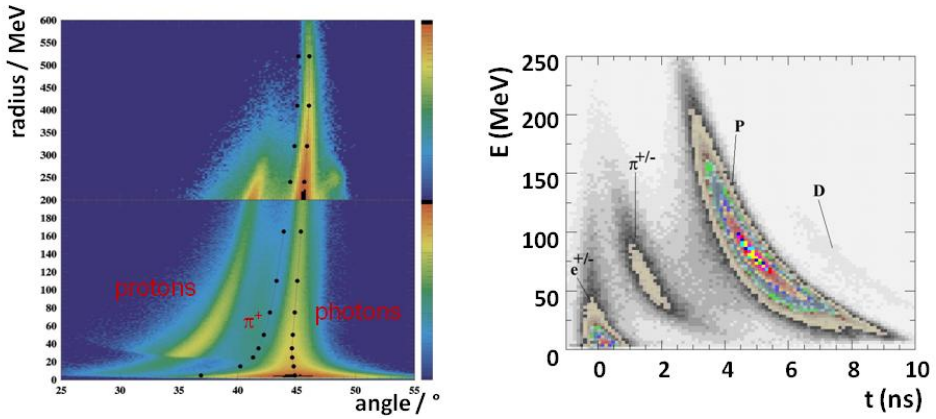

Figure 3. Both correlations illustrate the particle identification power of BaF_2 crystals exploiting the different line shape characterized by the specific ratio of the fast and slow scintillation component (left) and the correlation between measured energy and time of flight based on a time resolution $\sigma <$ 100ps. In case of the left figure, the lower part has been scaled by a factor and the black dots determine the ridge and the left valley of the photon response.

3. Present search for fast scintillators

In the 1980`s a new opportunity has been observed, the core-valence luminescence in BaF_2 which showed extremely fast rise and slow times, respectively, at a level below 1ns. Since the specific band structure - showing an energy distance between the core and valence band being smaller than the usual band gap between valence and conduction band - suppressed Auger transitions leading to a temperature independent so-called cross luminescence with a moderate light yield compared to the dominant luminescence based on self-trapped excitons [9]. In addition, both mechanisms showed a strong sensitivity to the ionization density of the absorbed particle providing particle identification via pulse shape analysis in addition to the excellent time-of-flight resolution. Figure 3 illustrates both techniques. Since the ratio of the fast and slow scintillation component remains nearly constant for a given particle species, the correlation of both separately measured and calibrated components can be

expressed by polar coordinates as plotted in the left part of figure 3. Unfortunately, only a few ionic crystals shows such a highly sensitive behavior.

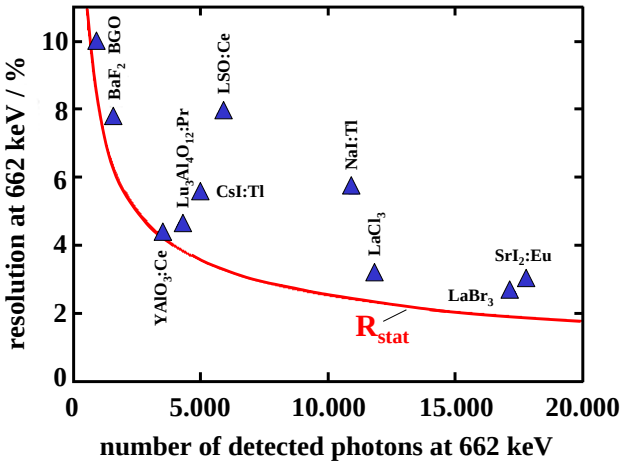

Figure 4. Energy Resolution (FWHM/E) measured at 662keV γ-energy with different scintillator crystals as a function of the number of detected photons in a photomultiplier tube. The red curve shows the limit expected from photon statistics [11].

About one decade ago [10] the search for bright scintillators with small band gap succeeded in growing large volume rare earth halides such as $LaBr_3$ or $LaCl_3$. In order to accomplish fast luminescence for high-rate studies or good timing resolutions these halides were doped with rare earth ions. Primarily Ce^{3+}-centers with an allowed 5f-4d transition provide decay times τ between 20 and 40ns depending on the host structure. Implementing Pr^{3+}-ions instead can further reduce τ by nearly a factor of two.

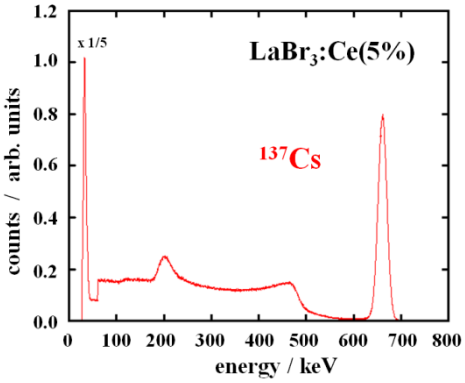

Figure 5. Response of a 19x19mm^2 $LaBr_3$:Ce crystal to a ^{137}Cs source. The part below 60keV is scaled down by a factor of 5 to display the simultaneous detection of 32keV Ba K_α X-rays[11].

Exploiting the fundamental limits given by Equation (1) the achievable energy resolution should be limited only by Poisson statistics of the luminescence yield. Figure 4 illustrates the presently achieved energy resolutions (FWHM) with respect to the theoretical limit and figure 5 shows the response function to γ-rays of 662keV energy (^{137}Cs) measured with a small $LaBr_3$:Ce crystal. However, besides the imperfection of the crystal, of light collection or of the sensitivity of the photo sensor a not yet fully understood mechanism of a non-proportional response at energies below 100keV appears to have a strong impact. This holds in particular for scintillators like LSO, NaI(Tl) or CsI(Tl), respectively. The recently developed halides have started to replace the traditional materials like NaI(Tl) or CsI(Tl)

in a wide field of application for γ-cameras, radiation protection or homeland security. Unfortunately, most of these materials are hygroscopic and therefore, have to be canned and consequently impose limits on a dense packing of multi detector systems.

In the past years Ce-doped silicate based inorganic scintillator materials were developed and heavily used for medical applications such as SPECT and PET tomography. Mass production of small samples but large quantities and the principal capability to grow also large size crystals was established for oxyorthosilicates such as Lu_2SiO_5 (LSO) and $Lu_{2(1-x)}Y_{2x}SiO_5$ (LYSO), both doped with cerium. The latter material combines high density (ρ = 7.4g/cm^3), fast response (decay time τ ~40ns at420nm) and in case of high energy applications a compact EM-shower containment (X_o=1.14cm, R_M = 2.3cm). In addition, high light yields comparable to NaI(Tl) were achieved for optimized Ce-concentrations [12].

4. Outlook

The engineering of future inorganic scintillation detectors and concepts has already started partly promoted by a series of new technologies in material sciences. The recently developed method of *micro pulling down* offers the consideration of optical fibers based on inorganic scintillators. Diameters between 0.3mm and 2.0mm, respectively, can be grown up to a typical length of 100cm. Production has been started successfully with doped or un-doped LuAG, YAG or LYSO samples (see figures 6 and 7). These fibers offer all advantages of inorganic materials such as high light yield, radiation hardness and fast response and provide intrinsically excellent position information. The currently developed Silicon photomultiplier, pixel arrays of avalanche diodes operated in the Geiger-mode, offer a perfectly matched readout with high quantum efficiency. First attempts for small animal PETS or position sensitive monitor detectors even for photon beams are under investigations.

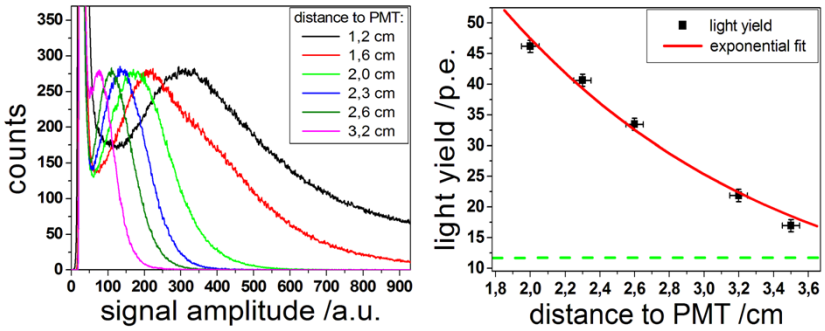

Figure 6. Response of a LYSO:Ce fiber (Ø=0.3mm, length L=100mm) to a ^{241}Am α-source as a function of the source distance to the photo sensor. The fibers still show a strong light attenuation due to imperfections [14].

For the first time, crystals have been proposed to construct a homogeneous calorimeter combining the EM and hadronic part, which would eliminate disturbing dead materials and would drastically improve the energy resolution in particular for hadronic jets measurement. The concept is based on the dual readout method to measure both Cherenkov and scintillation light simultaneously. Due to the extremely large volume of the calorimeter, the crystal material must be dense and UV-transparent. Present concepts either follow a bulk scintillator approach based on materials like BGO, rare earth doped PbF_2 or doped PWO emitting green scintillation light with slow decay time. The fast Cherenkov component can be either separated from the scintillation light either by the difference in wavelength or decay time similar to phoswich technology [8]. Alternatively, densely packed Ce-doped and un-doped scintillation fibers based on LuAG would allow a direct readout of both light components. In addition, the fibers could be integrated into an absorber matrix operating as a sampling calorimeter.

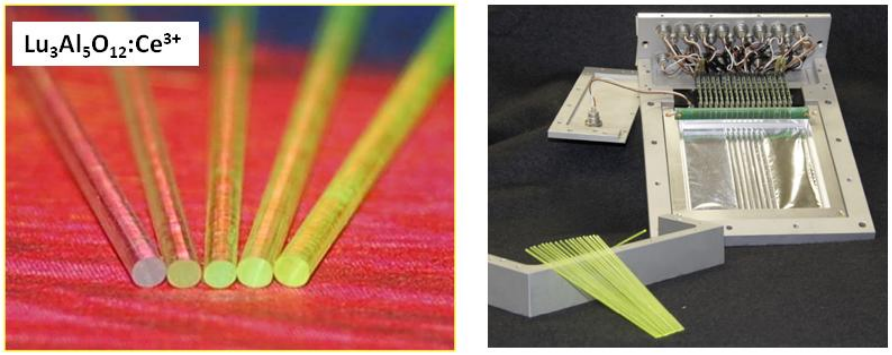

Figure 7. Inorganic fibres made of LuAG:Ce (left) and a first detector prototype used as 2D position sensitive monitoring detector[15].

In contrast to all previous investigations and applications of inorganic scintillators the study of the luminescence of scintillating nano-particles made of doped insulators has opened up a completely new field. Similar to the unexpected luminescence features of the quantum dots (Q-dots), made of semiconductors, the relaxation of energy, non-proportionality, surface effects and quantum and dielectric confinement can completely deviate from our understanding of a well known scintillator crystal. In spite of the tradition going back more than 60 years, inorganic scintillators are still a very vivid and expanding field of research and application.

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