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Efficiency of light collection in the combined detector NaI(Tl)//CsI(Na)

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A combined NaI(Tl)//CsI(Na) detector is an assembly of two scintillators, one of which is the "fast" NaI(Tl) (τ_f=230 ns), and the other is the "slow" CsI(Na) (τ_s=630 ns). Both crystals are cylindrical with a diameter of 19 cm. The NaI(Tl) scintillator has a thickness of 0.35 cm, while CsI(Na) has a thickness of 4 cm. For this scintillator configuration, a light collection method based on the dielectric permeability dependence of NaI(Tl) on the concentration of the activating impurity has been tested. The results of measurements of detectors using this principle of light collection indicate that the energy resolution of the 59.54 keV gamma line is in the range of 11-12%, and the non-uniformity of the light yield distribution over the surface of the input window is approximately 3%. This principle of light collection requires polished crystal surfaces and the use of an external light reflector with a diffusive light reflection characteristic. The polished state of the scintillator surfaces is a necessary condition for ensuring the stability of the characteristics of combined detectors during their operation.
Keywords: combined detector, phoswich detectors, non-uniformity of light yield distribution, grinding

Ефективність світлозбирання в комбінованому детекторі NaI(Tl)//CsI(Na). *Д.І.Зосим, І.К.Кириченко*

Комбінований детектор NaI(Tl)//CsI(Na) являє собою збірку двох сцинтиляторів, один із яких «швидкий» NaI(Tl) (τ_f=230 нс), а другий - «повільний» CsI(Na) (τ_s=630 нс). Обидва кристали виготовлені у формі циліндра діаметром 19 см. Товщина сцинтилятора NaI(Tl) становить 0.35см, а CsI(Na) - 4см. Для такої конфігурації сцинтиляторів випробувано спосіб світлозбирання, заснований на залежності діелектричної проникності NaI(Tl) від величини концентрації активаторної домішки. Результати вимірювань детекторів з реалізованим принципом світлозбирання показують, що енергетичне розділення гамма-лінії 59.54кэВ становить 11-12%, а неоднорідність розподілу світловиходу по поверхні вхідного вікна ~3%. Даний принцип світлозбирання узгоджується з полірованим станом поверхонь кристалів і використанням зовнішнього світловідбивача з дифузним характером відбивання світла. Полірований стан поверхонь сцинтиляторів є необхідною умовою забезпечення стабільності характеристик комбінованих детекторів в процесі їх експлуатації.

1. Introduction

The method of a combined detector, also known in literature as a phoswich detector, was developed in the early 1950s was first used by the HEAO1/A4 telescope in an astrophysical experiment [1, 2]. This type of detector has the advantages of both high photopeak efficiency and background event compensation compared to a single scintillator. The traditional configuration of a phoswich detector involves a combination of NaI(Tl) and CsI(Na) crystals, where NaI serves as the sensing volume and CsI acts as the anti-coincidence. Currently, this advanced technology is widely used in high-energy astrophysics for investigating objects in the hard X-ray and soft gamma-ray ranges. The application of phoswich detectors is particularly critical in scenarios requiring a large detection area, fast response times, and low background, such as the PDS (Proportional Counter Array) [3] on board BeppoSAX or experiments involving the synchronization of high-energy X-ray bursts [4] on the Rossi X-ray Timing Explorer. Phoswich detectors with NaI//CsI configuration are used in equipping modern and future space missions, including a telescope with modulation of hard X-ray radiation [5] and a multi-range cosmic monitor for astronomical variable objects (French-Chinese SVOM mission) [6,7]. The average amplitude resolution of the 59.54 keV line (R_{60}) for NaI//CsI phoswich detectors is typically in the range of R_{60} = 15 to 17%. Some authors note the degradation of the average R_{60} value over time. For instance, the authors of [8] observed an increase in R60 from 14.8% before launch to 15.4% in orbit. Based on the analysis of publications, it appears that R_{60} values of 14-15% for a phoswich detector in the NaI//CsI configuration are practically achievable limits. Therefore, the question of determining the mechanism for limiting the energy resolution of soft gamma radiation by a combined detector is relevant. The obtained result will contribute to the creation of conditions for light collection in a combined detector, ensuring an energy resolution of the 59.54 keV γ-line of less than 14%, a non-uniformity of light yield distribution over its input surface of no more than 4%, and the stability of the results obtained over a long period of operation of the detectors.

2. Experimental

The combined (phoswich) detector NaI(Tl)//CsI(Na) (hereinafter referred to as PhD) is a combination of a “fast” (τ_f=230 ns) NaI(Tl) scintillator in the form of a disk with a diameter of 19 cm and a thickness of 0.35 cm, and a “slow” (τ_s=630 ns) CsI(Na) scintillator, also in the form of a cylinder with a diameter of 19 cm and a thickness of 4 cm. One of the bases of the CsI(Na) cylinder is narrowed to the diameter of the photocathode of the photomultiplier. Both scintillators are carefully polished and bonded using optical gel, wrapped in several layers of polytetrafluoroethylene (ePTFE) foam [9] and placed in an aluminum alloy container. The entrance window is made of a beryllium plate (Be) with a thickness of 0.15 cm, which allows the transmission of more than 90% of 20 keV X-ray photons, meeting the requirements of sealing and vibration. The Be plate is positioned above the NaI(Tl) scintillator and seals the aluminum container. The light exit window made of one-centimeter-thick quartz glass is in optical contact with the CsI(Na) crystal, and seals the aluminum container from the opposite side. The scintillation light from NaI and CsI is detected by a common photomultiplier (PMT) Photonis XP3530 with a photocathode diameter of 11.4 cm.

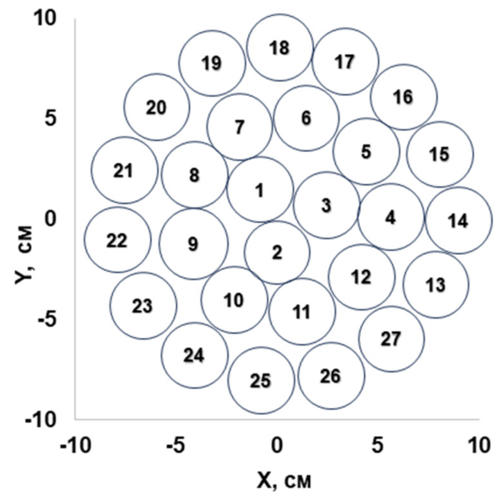

Fig. 1. Scheme of measuring the uniformity of the light yield distribution of the NaI(Tl)//CsI(Na) scintillation detector over its entrance surface

NaI(Tl) and CsI(Na) crystals were grown by an automated method of pulling on a seed in a conical crucible [10]. Using the method of directed dissolution in water, the crystal was cut along the growth axis; from the cut layer, samples were made in the form of a rectangular parallelepiped with a length of 4.5 cm and a cross-section of 1.1 cm×2.3 cm for qualitative and quantitative analysis of impurity composition. The content of uncontrolled impurity molecular anions in the crystals does not exceed 3×10^{-5} mol%. Cylinders made of NaI(Tl) and CsI(Na) crystals for scintillation detectors were turned on a lathe. After machining, the products were ground and polished.

The distribution of pulse amplitudes formed by the excitation of scintillators with gamma radiation was recorded by a Sugan spectrometer [11]. To determine the non-uniformity of the light yield distribution of PhD (Δ) over the surface of the detector entrance window, measurements were taken at 27 points distributed on it as shown in Fig. 1. In these measurements, a collimated radionuclide ^{241}Am (E=59.54 keV)

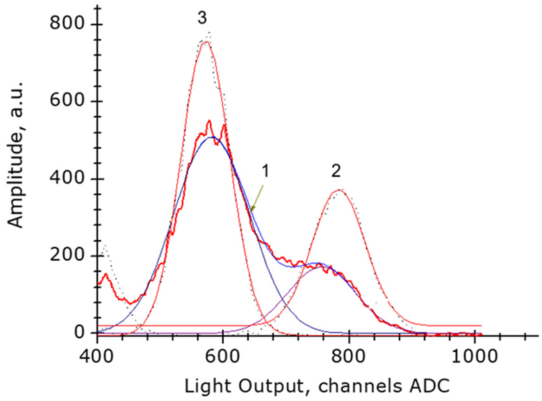

Fig. 2. Distribution of pulses by amplitudes recorded by the combined NaI(Tl)//CsI(Na) detector: 1 – non-collimated radiation source (^{241}Am) placed at a distance of 100 cm above the entrance window; 2 – collimated source of γ-photons placed directly on the entrance window at the center of the PhD (positions 1-3 in Fig. 1); 3 – collimated source of γ-radiation located directly on the entrance window in any position from 13 to 27 in Fig. 1.

was used. The collimator is made of lead one centimeter thick. The aperture of the collimator with a diameter of 0.5 cm is positioned one centimeter away from the surface of the entrance window. The total light yield and the spatial resolution of the PhD were measured using an uncollimated radionuclide ^{241}Am placed above the entrance window at a distance of 100 cm (SM position).

The quantity Δ was calculated using the relation: $\Delta = (V_{max} - V_{min})/V_{ave} \times 100\%$, where V_{max}, V_{min}, V_{ave} are the maximum, minimum, and average values of the light yield in the data array obtained during the scanning of the detector.

3. Discussion and results

If the activator is distributed uniformly over the volume of the NaI(Tl) disk, then the typical distribution of pulse amplitudes registered by the PhD during measurements in the SM position consists of two Gaussians (Fig. 2.1). One of them is more or less pronounced and corresponds to low light yield (V_s) (Fig. 2.3); the second is poorly pronounced and corresponds to high light yield (V_b) (Fig. 2.2). This fact indicates that the energy separation of the 59.54 keV PhD line is in the range of R=35-37%.

What is the mechanism of amplitude distribution formation (Fig. 2.1)? The formation of amplitude distribution (Figure 2.1) is described as follows: when a collimated source of γ-photons is positioned at the center of the detector entrance surface (positions 1-3 in Figure 1), the recorded amplitude distribution coincides with the Gaussian V_b. If the collimated source of γ-radiation is placed in any position from 13 to 27 (Figure 1), the amplitude distribution corresponds to the V_s one. We assume that the mechanism of formation of the amplitude spectrum (Fig. 2.1) is associated with the uneven distribution of pulses over the integration time (τ=1 μs). Specifically, the "direct" light pulses predominantly arrive first, forming the V_b distribution, while pulses from the peripheral part of the detector mainly arrive at the end, generating the V_s distribution.

An additional contribution to the formation of the two peaks is attributed to the dependence of the photocurrent of the photomultiplier tube (PMT) photocathode on the angle of light incidence. Thus, the difference in the time of arrival of scintillation radiation at the PMT photocathode may prevent the achievement of energy resolution $R_{60} \leq 14\%$ for the combined NaI(Tl)//CsI(Na) detector with a diameter of 19 cm when excited by γ-radiation with E_γ=59.54 keV.

Solving the problem of minimizing R and Δ of a scintillation detector involves choosing one of two options: either by aligning the arrival times of light pulses from the central (τ_b) and peripheral (τ_s) parts of the scintillator to the photocathode of the photomultiplier tube (PMT), or by "removing" one of its components V_s or V_b from the amplitude distribution. Achieving $\tau_s = \tau_b$ is possible by changing the crystal geometry, which is excluded in this experiment. As for excluding one of the amplitude spectrum components (Figure 2.1), the simplest way is to do it with "direct" light (V_b). By adjusting the optical and geometric characteristics of the reflector located above the NaI(Tl) scintillator, it is possible to obtain a normal amplitude spectrum of pulses, resulting in R_{60} values ranging from 22% to 19% (measured in the SM mode). In order to completely "avoid" pulses of direct scintillation light, the experiment was set up so that no scintillations were excited in the central part of the photomultiplier, on a certain plane (S_x). Results from measurements in the SM mode showed that with the ratio S_x/S_d = 0.512÷0.574, where S_d is the area of the PhD entrance window of a standard ePTFE reflector, the energy resolution at 59.54 keV is R ~12%. However, the drawback of this solution is that Δ is 100%. Therefore, "removing" one of

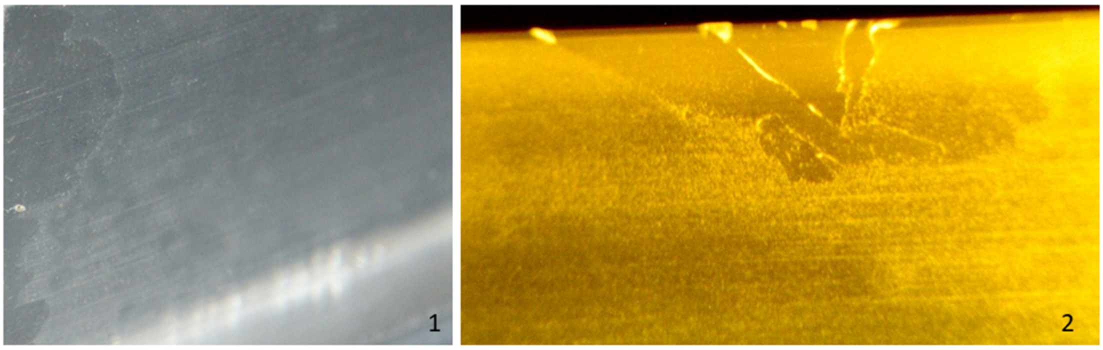

Fig. 3. Photographs of scintillator surface regions affected by uncontrolled changes in their state

the components of the amplitude spectrum is also not a solution to the problem.

We consider matting (or grinding) of scintillator surfaces, as a method of regulating light collection to be inappropriate. First, this method increases the effective path of scintillation light rays in the PhD. Secondly, the mechanical treatment of the crystal with abrasives leads to an increase in its free surface and, as a result, an increase in the free surface energy (F_s). The crystal gets rid of the created imbalance by activating various mechanisms. As a result, the state of the surface of the crystals changes uncontrollably. Fig. 3 shows some fragments of the spontaneous change of state of the surfaces of partially matted scintillators.

Figure 3.1 shows a section of the crystal surface that, when partially matted, remained polished, but over time, mostly spontaneously, became covered with roughness. Whether the surface remains polished (in the partially polished zone shown in Fig. 3.1) or whether roughness spontaneously develops is determined by the mechanisms of movement of mobile dislocations. Therefore, there is a probability that in certain areas of the crystal, local stresses induced by dislocation clustering may reach the crystal elastic limit. As a result, cracks appear in these areas, leading to the crystal fracture. This case is illustrated in Fig. 3.2 showing a fragment of the crystal surface where artificially induced roughness has partially disappeared, and cracks have emerged. In general, the thermodynamic imbalance of any of the crystals (NaI or CsI), being caused by physicochemical factors at the stage of their processing, serves as a mechanism for the instability of the characteristics of combined detectors.

An acceptable solution for creating the optimal light-collection geometry in a PhD may be related to the geometric parameters of the growth process of single-crystalline boules. It involves the following. It is known that one of the parameters of the crystal growth from melt is the shape of the crystallization front [10]. In our case, it has the geometry of a rotating paraboloid [10]. At the initial stage of growth, after the growing crystal reaches the required diameter, an activator is introduced. The activator dopant, in turn, decorates the crystallization front. Measurements of the activator distribution (C_{Tl}) in a direction perpendicular to the crystal growth axis confirm this.

In the NaI-TlI system, the lattice constant (d) of NaI(Tl) varies with the change in the Tl concentration (C_{Tl}) according to Vegard's rule. The change in the lattice parameter d leads to a variation in the dielectric susceptibility (χ) of the crystal. For alkali metal halides, the $\chi(d)$ function has the form [12]: $\chi = (\frac{d}{d_0})^3$, where the constant $d = 6\times10^{-8}$ cm. The refractive index (n) is expressed through the parameter χ as: $n = \sqrt{1+4\pi\chi}$. Thus, it can be concluded that the introduction of the activator in NaI results in the formation of an interface between NaI and NaI(Tl). Consequently, at a certain distance from the seed crystal, a NaI(Tl) scintillator exists in the form of a rotating paraboloid (*effect of geometry transformation of the scintillator!*). Such geometry of the interface allows for directing the scintillation light rays parallel to the axis of the detector. This assumption can be justified by Fig. 4.

The curve formed when a paraboloid of rotation intersects with a plane parallel to the axis of growth of the boule is a parabola. Fig. 4: AC is a tangent at point B to a section of the parabola; BE is normal to AC. Given the task condition: $\angle DBC = \alpha$. $\angle OBA = \angle DBC$ as complementary to $\pi/2$ angles of incidence ($\angle OBE$) and reflection ($\angle EBD$). Therefore,

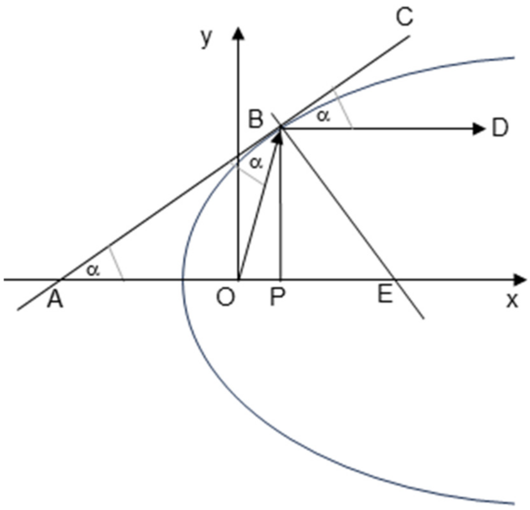

Fig. 4. Geometric interpretation of solving the problem of minimizing R and Δ in the PhD

$\angle OBA=\alpha$. Then ΔOAB is isosceles and $OB=OA$. Since and $OB=\sqrt{x^2+y^2}$, the equality will take the form: $AO=AP-OP=\frac{y}{dy/dx}-x$ or $ydx-\left(x+\sqrt{x^2+y^2}\right)dy=0$. The resulting equation is homogeneous with respect to x and y. By substituting $x = yq$, we turn it into an equation: $ydq=\sqrt{q^2+1}dy$ or $\frac{dq}{\sqrt{q^2+1}}=\frac{dy}{y}$. Having integrated this equation, we have $q+\sqrt{q^2+1}=\frac{\mathrm{y}}{\mathrm{C}}$. By eliminating irrationality and returning to the variables x and y in the general integral, we can write as $y^2=2C[x+\frac{C}{2}]$ describing a family of parabolas whose axis of symmetry coincides with the axis OX.

The parameter of the parabola is denoted as $r=C$, and its vertex is located to the left of the origin at a distance of $C/2$ from it. With a mirror diameter of melt D and a front depth h being constant, the integration constant is given by: $C=\frac{D^2}{8h}$ C, and finally, the equation of the parabola takes the form: $y^2=\frac{D^2}{4h}\,(x+\frac{D^2}{16h})$.

Thanks to the nature of light propagation n the crystal in the amplitude spectrum, the phenomenon of "merging" of pulses is observed from the region corresponding to the distribution V_s with pulses forming the distribution V_b, thereby condition is satisfied $\tau_s \approx \tau_b$. This light collection principle in NaI(Tl), through the creation of a boundary between two media, has been identified as the principle of a *virtual phokon* (VF). The "shifting" effect of V_s towards V_b is well pronounced in the measurements of the PhD, for which the NaI(Tl) scintillator is initially prepared from the region of stable crystal growth (uniform distribution of the activator throughout its volume). Subsequently, each successive element of NaI(Tl) is manufactured closer and closer to the seed. In the experiments for the detection of 59.54 keV γ-radiation using the NaI(Tl)//CsI(Na) detector with VF-type light collection geometry, the best values for the parameters R and Δ are R=11.3% and Δ=±1.3%. The generalized results are as follows: in 11 experimental PhD samples from different boules, in 7 of them, obtained R values are grouped in the range of 11.3-14.4%, and Δ in the range of 2.6-3.7%. The obtained R=11.3% is close to the value of 10.6% in the $LaBr_3$:Ce scintillator, which is a constituent element of the LaBr3: Ce//NaI(Tl) phoswitch detector with a diameter of 10.16 cm. In this detector, $LaBr_3$:Ce with a thickness of 0.6 cm acts as an active element, while NaI(Tl) with a thickness of 4 cm acts as an anti-coincidence detector [13].

The method of light collection, VF, can be implemented in a scintillator exclusively during the crystal growth stage. The ability to control the geometry of the crystallization front provides additional opportunities during its implementation. However, certain limitations are expected for the application of such a detector engineering method, as the geometry of the crystallization front affects the structural perfection of single crystals. Thus, the principle of VF light collection requires more detailed investigation. As a consequence of implementing the VF light collection mechanism in the PhD, acceptable values of R and Δ are achieved with polished crystal surfaces, ensuring the stability of detector characteristics. Measurements of R and Δ after performance tests of the PhD, involving mechanical vibrations at several g (g is acceleration due to gravity), heating up to 370K, and cooling down to temperatures of 248K, showed that their values vary within ±0.1÷0.2%, which does not exceed the measurement error.

4. Conclusions

A mechanism limiting the energy resolution of soft γ-radiation in a combined NaI(Tl)//CsI(Na) detector with a uniform distribution of activator throughout the volume of the NaI(Tl) scintillator has been identified. This mechanism arises from the time difference in the response of pulses from the central and peripheral parts of the detector. Implementing a virtual *fokon* light collection method in the NaI(Tl) scintillator allows overcoming this limitation when detecting with the combined NaI(Tl)//CsI(Na) detector for the 59.54 keV γ-line, resulting in energy resolution and light yield distribution non-uniformity values of 11.3% and 2.6%, respectively. A distinctive feature of the virtual *fokon* method in scintillation technology is that it can only be used at the growth stage of single crystal boules and requires caution regarding its potential influence on the structural perfection of single crystals. The virtual *fokon* method is compatible with the polished state of crystal surfaces in combination with a diffusely reflecting light reflector. Polished crystal surfaces are a necessary condition for ensuring the stability of detector characteristics during their operation.

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