Optical performance test and energy resolution analysis on LaBr₃(Ce) crystal

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Abstract. The paper used ¹³⁷Cs and ⁶⁰Co γ sources to measure and analyze the effects of different voltage, gain, count time of photomultiplier tube and different distance of sources on the energy resolution of LaBr₃(Ce) crystal; the relationship curves were obtained quantitatively. The experiment values are in good agreement with MCNP simulation results. The results show that the energy resolution of LaBr₃(Ce) is negatively correlated with voltage and gain; the gain is more influential. The energy resolution of LaBr₃(Ce) is positively correlated with detection distance and count time; the detection distance is more influential. The paper used picosecond X-ray generated by pulsed power accelerator “Qiangguang-1” to excite the LaBr₃(Ce) crystal and measure its fluorescence decay time and emission spectrum. The results are consistent with the available experimental data of other scholars and verify the excellent time response of LaBr₃(Ce). With the increase in X-ray energy, the peak emission wavelength shifts towards infrared direction, which can better match the spectral response of photocathode.

1. Introduction

Lanthanum bromide (LaBr₃) crystal, especially doped with cerium (Ce³⁺), is one of the most popular inorganic scintillators in recent years. LaBr₃(Ce) crystal has excellent energy and time response, good spatial resolution, good machinability, and stable chemical properties [1-2]. Its energy resolution for 0.662MeV photons (3%) is superior to that of traditional inorganic scintillator NaI(Tl) (7%) and new inorganic scintillator LuYAP(Ce) (8.2%) [3-4]. The photon yield of its single crystal (63 photon/KeV) is also higher than that of NaI(Tl) (38 photon/KeV) [5]. Compared with HPGe, it has unique capability to perform gamma-gamma coincidence measurement in high counting rate [6].

LaBr₃(Ce) scintillator is favored by researchers because of its listed traits. Vedia V et al. evaluated the time and energy response of LaBr₃(Ce) crystals with two geometric shapes, and simulated the energy resolution of the truncated cone type was 3.3% and that of tapered type was 3.8% [7]. Xie X C et al. obtained that the detection efficiency of larger LaBr₃ crystal is higher than that of smaller one by Monte Carlo method [8]. Fraile L M et al. compared the time and energy resolutions of LaBr₃(Ce+Sr) and LaBr₃(5%Ce), and found that adding 0.5% Sr²⁺ to LaBr₃(5%Ce) increased the energy resolution from 3.5% to 3.3%; but deteriorated the time response [9]. Deng Z H et al. obtained the spontaneous background spectra of LaBr₃ by using three different radioactive sources (⁶⁰Co, ¹³⁷Cs, ¹⁵²Eu) [10].

In recent years, the studies have focused on the applications of LaBr₃ in large volume, high energy, high dose, neutron detection and other aspects; all of these broaden the application scope of LaBr₃. In order to protect LaBr₃ crystal structure, Yi Y C et al. researched that the fluence rate linear response
limit of γ-ray is about $2.0 \times 10^{18}$ MeV/(cm$^2$·s) and of X-ray is about $1.8 \times 10^{20}$ MeV/(cm$^2$·s) [11-12]. Dhibar M et al. designed a large square bar of LaBr$_3$(Ce) detector array, and measured its energy and time resolution under high-energy γ-ray [13]. Quarati F G A et al. found that reducing the number of active electrodes help to maintain the energy resolution and avoid the signal saturation of photomultiplier [14]. Gosta G et al. researched that the response function of large volume LaBr$_3$(Ce) and the linearity of coupled photomultiplier under high-energy γ-ray [15]. Based on some specific neutron induction peaks in the inelastic interaction between neutron and crystal, Liu C et al. put forward that LaBr$_3$(Ce) detector can be applied to neutron detection directly without TOF [16].

So far, the researches of factors affecting the LaBr$_3$(Ce) detection performance in the actual use are insufficient. However, different back-end electronic conditions can affect the detection. Therefore, the paper used $^{137}$Cs and $^{60}$Co γ sources to measure and analyze the effects of different voltage, gain, count time of photomultiplier tube and different distance of sources on the energy resolution of LaBr$_3$(Ce) scintillator. The experiments and simulations verified each other.

2. Monte Carlo simulation scheme
Monte Carlo method uses random number to calculate states of the particles, then counts all the reactions to get the result. The calculation errors are along with the results [17]. MCNP program is a particle transport computing software based on the Monte Carlo method. MCNP is developed and supported by the Eolus team at Los Alamos National Laboratory (LANL) for neutron, photon, electron, or coupled neutron/photon/electron transport. MCNP5, released in 2003, is rewritten in ANSI standard Fortran 90. In the paper, MCNP5 program is used to simulate the calculation of photon energy emitted from LaBr$_3$(Ce) crystal.

2.1. Selection of simulation parameters
The LaBr$_3$(Ce) crystal was used in the experiment, with density of 5.26 g/cm$^3$, cerium dopant concentration of 0.3%, radius of 1.27 cm, and height of 2.54 cm. The LaBr$_3$(Ce) crystal is produced by Kehaijijing company (www.khjjtech.com). The crystal parameters above and the experimental encapsulation structure are used in our simulation model. The material parameters of the encapsulation structure are shown in Table 1. These parameters are written in MCNP’s material card, which is an essential part of MCNP program.

| Material             | Reflector | Optical couplant | Windowpane | Case | Shielding Layer |
|----------------------|-----------|------------------|------------|------|-----------------|
| Density g/cm$^3$     | 3.58      | 0.98             | 2.3        | 2.7  | 11.34           |

2.2. Gaussian broadening calculation coefficient of card FT$_n$
The MCNP5 program provides the F8 counting card to calculate the distribution of pulse energy, and the FT$_n$ card to broaden the pulse amplitude distribution by gaussian. FT$_n$ is special treatment card. We use one of its functions, which is called gaussian energy broadening (GEB). We used the measured Full Width Half Maximum (FWHM) of the full-energy peak and used the following formula (1) to determine the simulation factors a, b and c:

$$FWHM = a + b\sqrt{E} + cE^2$$  

Where a, b, and c specify the weight width of the FWHM observed in the radiation detector, and are obtained from experimental data [18].

2.3. Establishment of MCNP model
We have established an idealized model by MCNP5. We can deem that the whole simulation process is an undisturbed situation relatively. Figure 1 and Figure 2 are the Monte Carlo simulation model of LaBr$_3$(Ce) detector, which show the encapsulation structure and materials by MCNP Visual Editor.
Three effects of LaBr₃(Ce) crystal interacting with γ-ray (Photoelectric effect, Compton scattering, Electron pair effect) produce secondary electrons. Secondary electrons energize the crystal, producing electron-hole pairs. As an electron transitions from the conduction band back into the valence band, the system emits a photon with an energy equal to the gap width. Because the fluorescence fluctuates, a single-energy γ-ray produces a continuous energy distribution spectrum.

Figure 1. The longitudinal section of the model.

Figure 2. The cross-section of the model.

3. Measurement of LaBr₃(Ce) energy resolution under the effects of multiple factors
We used the energy resolution measurement platform (Figure 3) for the experiments in our laboratory.

Figure 3. The energy resolution measurement platform.
Encapsulated LaBr₃(Ce) crystal contacts the photomultiplier tube (PMT HAMAMATSU, CR105, Japan) tightly. The pulse signals of PMT are amplified and processed in amplifier and the multichannel...
analyzer (MCA Canberra, France). The crystal, radioactive sources, and PMT are shielded by lead brick to ensure the darkness of the experimental environment.

3.1. Effect of changing HV on energy resolution

We measured the energy spectrum of LaBr$_3$(Ce) scintillator excited by $^{137}$Cs $\gamma$-ray first, and calibrated the channels with $^{60}$Co energy spectrum measured simultaneously. The energy resolution is the ratio of FWHM to peak channel.

After a number of experiments, we choose the stable operating voltage range 467V–475V of the measurement platform as the experiment HV; and we take 2V as a gradient. As shown in Figure 4, at 0.662MeV, the experiment values are in good agreement with the simulation results. The low energy region doesn’t fit well, because the simulation doesn’t account the characteristic X-ray peak at 32KeV, which is emitted by conversion electron during $\gamma$ transition, or by orbital electron capture during $\beta$ decay. At the same time, we find that the experiment values were higher than the simulation values on both sides of the full-energy peak. The phenomenon is due to the existence of dark counts during the experiments; the dark counts are ignored in the simulation.

The relationship between channel and HV is shown in Table 2. The channel of the full-energy peak increases with the raise of the operating voltage of the photomultiplier. The full-energy peaks of LaBr$_3$(Ce) scintillator exited by $^{60}$Co $\gamma$-ray at 1.172MeV and 1.332MeV are compared with the corresponding channels too. The relationship reflects the availability of calibration system. As can be seen from Table 2, the energy resolution increases with the raise of HV. The main reason is that the raise of the dynode voltage increases the number of electrons captured by the anode, and the statistical performance of the data becomes better. We find that the energy resolution is linearly negatively correlated with HV in measurement range. The experiment and the simulation are consistent. The linear relationship between energy resolution and HV is: $Y = -0.041X + 22.98$.

![Figure 4](image-url)

**Figure 4.** Energy spectrums at different voltages.

**Table 2.** Effect of changing HV on energy resolution.

| HV (V) | Counts | Channel | Energy resolution (%) |
|--------|--------|---------|-----------------------|
|        |        | 0.662MeV | 1.172MeV | 1.332MeV | Simulation | Experiment |
| 467    | 1746   | 404      | 687      | 770      | 3.88       | 3.86       |
| 469    | 1728   | 415      | 706      | 790      | 3.76       | 3.75       |
| 471    | 1704   | 432      | 730      | 816      | 3.59       | 3.60       |
| 473    | 1669   | 442      | 749      | 837      | 3.58       | 3.59       |
| 475    | 1644   | 457      | 773      | 863      | 3.52       | 3.53       |
3.2. Effect of changing Gain on energy resolution
We used the spectral calculation software GenieTM2000 to analyze the energy resolution of LaBr₃(Ce) with different gains. The shape of the energy spectrum is similar to above. As can be seen from Table 3, when the rough-tuning is set at 2, the counts are 1107. With the increase in fine-tuning gain, the counts decrease by 11.5%, 22.1%, 34.1% and 43.0% respectively. For every 0.2 increase in fine-tuning gain, the counting rate decreases by 10%. Similar to the effect of the HV, the energy resolution increases with the raise of gain. Within the experimental range, when the gain goes up by 1, the energy resolution increases by 0.305% approximately, which is far greater than the effect of HV. The paper analyzes the correlation between energy resolution and gain, and obtains the fitting curve. The slope obtained in the experiment is -0.305, while the slope obtained in the simulation is -0.28. The difference between the experiment and simulation may be caused by the experimental error. The results show that the raise of gain reduces the counting rate with the energy resolution increasing. Therefore, it is critical to find appropriate gain to balance the energy resolution and counting rate to meet specific detection needs.

Table 3. Effect of changing gain on energy resolution.

| Gain (Rough/Fine) | Counts | Channel 0.662MeV | Channel 1.172MeV | Channel 1.332MeV | Energy resolution (%) |
|-------------------|--------|-------------------|-------------------|-------------------|-----------------------|
| 2 / 1.0           | 1107   | 255               | 432               | 482               | 3.67                  | 3.69                  |
| 2 / 1.2           | 980    | 306               | 518               | 579               | 3.63                  | 3.61                  |
| 2 / 1.4           | 862    | 359               | 605               | 675               | 3.56                  | 3.57                  |
| 2 / 1.6           | 730    | 408               | 691               | 771               | 3.53                  | 3.52                  |
| 2 / 1.8           | 631    | 460               | 778               | 869               | 3.44                  | 3.43                  |

3.3. Effect of changing detection distance on energy resolution
We studied the effect of the distance between the crystal and radioactive sources on energy resolution by setting HV of 475V, rough-tuning gain of 2, fine-tuning gain of 1.8, and count time of 500 seconds. During the study, the radioactive sources were placed 0cm, 2cm, 4cm and 6cm away from the crystal. The distance was measured by vernier caliper.

Table 4. Effect of changing detection distance on energy resolution.

| Distance (cm) | Counts | Channel 0.662MeV | Channel 1.172MeV | Channel 1.332MeV | Energy resolution (%) |
|---------------|--------|-------------------|-------------------|-------------------|-----------------------|
| 0             | 1644   | 458               | 774               | 864               | 3.52                  | 3.53                  |
| 2             | 629    | 431               | 730               | 815               | 3.74                  | 3.71                  |
| 4             | 285    | 433               | 732               | 819               | 3.55                  | 3.56                  |
| 6             | 182    | 443               | 749               | 837               | 3.78                  | 3.78                  |

The energy spectrums are shown in Figure 5. It can be seen from Table 4 that the counts decrease with the increase in detection distance; the experiment values are in good agreement with the simulation results. When the radioactive sources and LaBr₃(Ce) crystal are attached, the counts are 1644. With the increase in detection distance, the counts decrease by 61.7%, 82.7%, and 88.9% respectively. When the detection distance is 0cm, almost all the light emitted by LaBr₃(Ce) crystal can be caught by photocathode of photomultiplier. With the increase in detection distance, the radiation emitted by the radioactive source will diffuse towards the solid angle of space, and will lose some of its intensity. The increase in detection distance results in worse energy resolution and lower detection efficiency. It can also be seen from Figure 5 that the increase in detection distance increases the counting ratio of the noise, which is mainly gathered in low energy region. It is due to the counts of low energy scattered Compton photons produced by experimental apparatus interacting with γ-ray. The energy resolution is fitted with detection distance in Figure 6(c). It can be seen that the relationship between the energy resolution and
detection distance is positively correlated. For every 1cm increase in detection distance, the energy resolution increases by 0.3% approximately. It can be concluded from correlation analysis that the effect of detection distance on the energy resolution of LaBr₃(Ce) is great. In actual detection, we should pay attention to the placement of scintillator, and the location of radioactive object.

![Figure 5](image_url)

**Figure 5.** Energy spectrums at different detection distances.

3.4. Effect of changing count time on energy resolution

We studied the effect of the count time of PMT on the LaBr₃(Ce) crystal energy resolution by setting HV of 475V, rough-tuning gain of 2, fine-tuning gain of 1.8, and detection distance of 0cm. The time interval is 400s, and the total count time is 2000s.

**Table 5.** Effect of changing count time on energy resolution.

| Count time (s) | Counts | Channel 0.662MeV | Channel 1.172MeV | Channel 1.332MeV | Energy resolution (%) Simulation | Energy resolution (%) Experiment |
|---------------|--------|-------------------|-------------------|-------------------|----------------------------------|----------------------------------|
| 400           | 1353   | 443               | 752               | 839               | 3.59                             | 3.59                             |
| 800           | 2520   | 443               | 752               | 839               | 3.58                             | 3.62                             |
| 1200          | 4118   | 443               | 752               | 840               | 3.59                             | 3.60                             |
| 1600          | 5331   | 444               | 752               | 840               | 3.63                             | 3.63                             |
| 2000          | 6630   | 444               | 752               | 840               | 3.60                             | 3.60                             |

The experiment values are consistent with the simulation results. With the increase in count time, it is obvious that the counts will increase. But the location of the full-energy peak at 0.662MeV, 1.172MeV and 1.332MeV γ-ray does not change. The increase in counts also includes the dark counts, so the energy resolution will be slightly different. As can be seen from Table 5, the increase in count time has little impact on energy resolution; and the resolution maintains at 3.60% approximately. The energy
resolution is fitted with count time; the fitting formula is \( Y = 7.5 \times 10^{-6} X + 3.599 \). The low slope indicates that the energy resolution of LaBr\(_3\)(Ce) crystal is stable over a long time range. Therefore, we don’t have to sacrifice long time to increase the counts in actual detection process.

In addition, figure 6(a) shows the relationship between the energy resolution and the HV, figure 6(b) shows the relationship between the energy resolution and the gain.

4. Measurement on fluorescence decay time and emission spectrum of LaBr\(_3\)(Ce)

The pulsed power accelerator called “Qiangguang-I” can provide high fluence rate pulsed X-ray radiation field. High voltage pulse is produced by a linear transformer driver. After twice pulse compression, great electrical current pulse is collected on the load. High energy electron beam was produced then. The bremsstrahlung X-ray is generated by the collision of high energy electron beam and tantalum targets. The average X-ray energy is approximately 1 MeV [19].
Because the luminous efficiency of LaBr$_3$(Ce) is as high as 67000 Photons/MeV, we used phototube without dynodes for photoelectric conversion. In order to prevent the phototube from being exposed to X-ray directly and to shield the crystal better, we designed the experiment device shown in Figure 7. The crystal and phototube were placed in a shielded darkroom made of lead brick. The outside of the lead brick is covered with black cloth for further shading.

The interior is shown in Figure 7. X-rays from the accelerator are focused on a convex lens at the entrance, then reflect off the mirror and hit the LaBr$_3$(Ce) crystal; The crystal is coupled to the phototube with silicon oil. The power cord and signal cord of the phototube are drawn from the opposite side of the light inlet.

Picosecond X-rays and ultra-fast phototube make the fluorescence generated by the crystal the slowest signal in the measurement process, thus avoiding the oscilloscope signal shape reflecting the X-ray pulse. Compared with single-photon method and other measurement methods which require start-stop signal consistency, direct measurement uses fewer electronic devices, which is conducive to improving measurement efficiency and reducing electronic noise.

Response curve was obtained by the monitoring detector. We redraw the fluorescence decay curve with experiment data, and performed an exponential fitting to the curve. As shown in Figure 8, the measured LaBr$_3$(Ce) fluorescence decay time is 21.85ns. The result is similar to those measured by other scholars [1][20]; the excellent time response of LaBr$_3$(Ce) is verified.

**Figure 8.** The fluorescence decay curve of LaBr$_3$(Ce).

**Figure 9.** The emission spectrum of LaBr$_3$(Ce).

We used NOVA-EX refrigerated fiber optic spectrometer to measure the emission spectrums of LaBr$_3$(Ce) under X-ray. The X-ray energy was set at 20KeV, 40KeV and 80KeV. The measurement was completed in the dark room. The spectrums are shown in Figure 9. When the crystal is excited by higher energy X-ray, the peak emission wavelength is around 380nm, which is consistent with the ideal truth value [1][20]. We found the fluorescence spectrum of LaBr$_3$(Ce) crystals has an infrared shift tendency with the increase in X-ray energy, which can better match the response spectrum of the photomultiplier, improve the quantum efficiency, and thus optimize the detection performance of the LaBr$_3$(Ce) crystal.

5. Summary

In this paper, we find that the energy resolution of LaBr$_3$(Ce) crystal is negatively correlated with the voltage and gain of PMT; the effect of gain is more significant. The energy resolution of LaBr$_3$(Ce) is positively correlated with detection distance and count time, the effect of distance is more significant. The experiment values are in good agreement with MCNP simulation results. To some extent, increasing the operating voltage of the photomultiplier can improve the energy resolution of LaBr$_3$(Ce) scintillator. In the actual detection, we should pay attention to the placement of the scintillator, but don’t have to sacrifice long time to increase the counts. The LaBr$_3$(Ce) fluorescence decay time we measured is
21.85ns, the result reflects the excellent time response of the crystal. We find the peak emission wavelength of LaBr$_3$(Ce) is around 380nm. The fluorescence spectrum of LaBr$_3$(Ce) crystals has an infrared shift tendency with the increase in X-ray energy, which can better match the response spectrum of the photomultiplier. Then the next work will research the factors which can affect the time resolution of LaBr$_3$(Ce) crystal.

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