Characterization and Applications of New High Quality LuAG:Ce and LYSO:Ce fibers

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Abstract. Inorganic scintillating fibers have a large potential as highly granular detector components in hadron and particle physics as well as in medical applications. With the micro-pulling-down-method a fast and cost efficient technique was developed over the last decades to grow such fibers from the melt. This paper will present the recent development of the quality of LuAG:Ce and LYSO:Ce fibers in terms of light output and light attenuation inside the fiber. For this purpose different steps in optimizing the fiber growth will be compared with the achieved fiber performance. In addition the response of the fibers to low energetic gamma rays will be studied and different readout concepts for the fibers will be discussed and compared.

1. Introduction

Inorganic scintillating fibers show significant advantages compared to organic fibers, like a higher light yield and a more effective interaction with electromagnetic probes due to the high Z elements contained in the fiber material. In addition one expects a higher radiation resistance, which is especially important in highly radioactive environments like the inner or forward parts of high energy physics detectors. These advantages in combination with the high granularity of the fibers open up a wide field of applications. In high energy physics the combination of doped and un-doped fibers will allow the design of compact hadron sampling calorimeters exploiting the dual-readout concept [1], [2]. In addition, the sub millimeter position resolution given directly by the fiber size can be exploited for beam monitoring and in combination with the good efficiency for γ-rays for medical applications like PET-scanners as well [3], [4].

2. Production Technology and Scintillator Materials

2.1. The micro-pulling-down method

Conventional methods for crystal growth like the Czochralski or Bridgeman method usually provide large crystal ingots. To produce highly granular detectors mechanical treatments like cutting and polishing impose a large workload. This can be avoided by producing crystals as fibers directly from the melt, like with the edge defined film-fed growth, the laser heated pedestal growth (LHPG) or the micro-pulling-down (μ-PD) method is needed. The μ-PD-method has been established over the last decades to be one of the best methods for the growth of single
crystal fibers with a diameter below 1 or 2 mm, respectively, and with a length up to several tens of centimeters [5], [6]. Fig. 1 shows a schematic view of the fiber growth with the micro pulling down method. The crystal melt is heated up in a crucible by an external radiofrequency heater. The fiber is pulled down on a small seed through an specially shaped opening on the bottom of the crucible. During this process several critical parameters, like the temperature gradient, and the speed of pulling down have a strong influence on the fiber quality.

2.2. Properties of LuAG:Ce and LYSO:Ce
For the fiber production the materials LuAG:Ce and LYSO:Ce have been chosen. Table 1 summarizes the most relevant physical and scintillation properties of the two materials. It shows that both fulfill the requirements for high energy physics detectors. However, LYSO:Ce appears most favorable due to the short radiation length and due to the high light yield. On the other hand, LuAG:Ce is much easier to grow due to the lower melting point.

|                         | LuAG:Ce          | LYSO:Ce         |
|-------------------------|------------------|-----------------|
| density                 | 6.7 g/cm³        | 7.1 g/cm³       |
| $Z_{\text{eff}}$        | 63               | 66              |
| radiation length        | 1.41 cm          | 1.2 cm          |
| decay time              | (50-60) ns       | 45 ns           |
| $\lambda_{\text{max, emission}}$ | 530 nm          | 420 nm          |
| refractive index        | 1.85 @ 530 nm    | 1.82 @ 420 nm   |
| light yield             | 12000 - 25000 ph/MeV* | 26000 - 32000 ph/MeV* |
| melting point           | 1980°C           | 2100°C          |

3. Characterization - setup and procedure
The readout of the fibers is performed with silicon photomultipliers (SiPMs). Sensors with a pixel size of 25 µm (S12572-025C) and 50 µm (S12572-050C), respectively, from Hamamatsu [7] have been used. The measurements confirmed, that for fibers with 2 mm diameter the 50 µm version has a sufficient numbers of pixels to detect the total light yield with a significantly increased photon detection efficiency compared to the 25 µm version. The signals of the SiPMs are amplified by a special preamplifier (photonic AMP-0611, AMP-0604) and digitalized with commercial NIM and CAMAC electronics using a charge integrating ADC. The fiber is coupled at one or both ends to the SiPMs using optical grease. Fig. 2 shows the experimental setup with the movable collimated source on the top of the fiber.
3.1. Response to alpha particles
For the characterization of the light yield and the attenuation coefficient, alpha particles from an collimated $^{241}$Am source with a mean effective energy of 4.0 MeV were used. The main advantage of alpha particles compared to gamma sources is, that they deposit locally their complete energy even in very thin fibers. One obtains a narrowly peaked response, which can be fitted with a Gaussian distribution. Fig. 3 shows the response to an $\alpha$-source for a $\otimes$ 2 mm fiber with increasing distance between source and SiPM. The highest signal amplitude is observed at closest distance to the SiPM. The attenuation of the light yield $LY$ as a function of the distance $d$ between source and PMT can be described by the following relation: $LY(d) = LY_0 \cdot e^{-\mu d} = LY_0 \cdot e^{-\frac{d}{\lambda}}$ with $LY_0$ as initial light yield, $\mu$ as attenuation coefficient and $\lambda$ as attenuation length, respectively.

4. Characterization and optimization of light yield and light attenuation of LuAG:Ce fibers
To optimize the light yield at reduced light attenuation several production runs have been performed.

4.1. Influence of the speed of pulling down and other growth parameters
In a first optimization run the influence of different growth parameters was investigated, like the speed of pulling down, the orientation of the seed and the sensitivity of the fiber quality to the distance to the seed. Table 2 shows some of the properties of the investigated fibers, while the corresponding attenuation curves are shown in fig. 4. The results do not indicate a significant influence of the seed orientation but favor a slow pulling speed, since it provides the crystal lattice more time to relax leading to less bubbles and cracks. In addition, fibers samples cut close to the seed show smaller attenuation coefficients.

| fiber | $\mu$ cm$^{-1}$ | orientation of seed | dist. to seed | speed of pulling down |
|-------|-----------------|---------------------|---------------|-----------------------|
| # 1   | 0.69            | $< 100 >$           | 11.0          | high                  |
| # 2   | 0.77            | $< 100 >$           | 22.5          | high                  |
| # 3   | 0.13 - 0.4      | $< 100 >$           | 2.5           | avg.                  |
| # 4   | 0.46            | $< 111 >$           | 22.5          | low                   |
| # 5   | 0.68            | $< 100 >$           | 22.5          | low                   |
4.2. Influence of the position within the grown fiber

The $\mu$-PD method allows the growth of fibers even longer than 1 m in length. To study the variation of the fiber quality over the full fiber length, a fiber with a diameter of 1 mm and a length $> 60$ cm was grown and cut into several 10 cm long samples, starting 0.5 cm apart from the seed. Fig. 5 shows the attenuation curves for 4 of these samples indicating a large variation. Attenuation coefficients of $0.38 \text{ cm}^{-1}$ and $0.32 \text{ cm}^{-1}$, respectively, are measured for the first two samples. In contrast, the last two ones, cut further apart from the seed, show significantly larger values of $0.92 \text{ cm}^{-1}$ or even $1.14 \text{ cm}^{-1}$. The significantly decreased fiber quality can be addressed to a variation of the Ce-concentration inside the fiber. In the beginning of the pulling process, less Ce might be implemented into the LuAG-matrix located close to the seed while Ce-Ions could become more abundant in the crucible at a later stage leading to a higher Ce-concentration causing more surface defects, cracks and bubbles [9], affecting the light attenuation. This hypothesis is supported by the attenuation corrected absolute light yield. For the first two samples a corrected light yield of 88 - 110 photons can be obtained in contrast to 121 and 149 photons for the second pair of fibers. This increase is expected for a higher Ce-concentration as described for bulk LuAG:Ce crystals [12]. The results are in agreement with a visual optical inspection (see Fig. 6). Sample 1378.1, which was cut close to the seed, appears in light color and shows almost no cracks. In contrast, sample 1378.6 has a darker color, indicating a higher Ce-concentration and showing many small cracks.

4.3. Influence of the Ce-concentration

To investigate directly the influence of the Ce-concentration, samples were grown with different Ce-concentrations. Fig. 7 shows the obtained attenuation curves for LuAG:Ce fibers with 1 mm and 2 mm diameter, respectively. The correlation of the light yield and the attenuation coefficient with the Ce-concentration is shown in fig. 8 for fibers with two different diameters. As
expected, a lower Ce-concentration decreases the attenuation coefficient but to the disadvantage of a reduced light yield, as known for LuAG:Ce bulk crystals.

4.4. Visual inspection of the fiber quality
The optimization discussed in this chapter can also be confirmed in a visual inspection of the different fiber samples shown in fig. 9. It becomes clearly visible, that a lower pulling speed and a reduced Ce-concentration avoid significantly visible cracks and bubbles along the fiber causing primarily strong intrinsic attenuation.

5. Characterization and optimization of light yield and light attenuation of LYSO:Ce fibers
So far only a few LYSO:Ce fiber samples with a diameter between 0.35 mm and 0.65 mm and a length of up to 10 cm have been produced and investigated. All fibers investigated so far have a
Y-content of 6% and a Ce-concentration of around 0.05%. While the first fibers had a quite high attenuation coefficient of up to 1.8 cm$^{-1}$, it was possible to reduce the attenuation coefficient down to $\mu = (0.68 \pm 0.02)$ cm$^{-1}$ for the last production run. The light yield measured for the four available samples from this run is very homogeneous. Fig. 10 shows the light yield as a function of the source position for a typical LYSO:Ce fiber of the latest production run. Up to now LYSO:Ce fibers show a very promising performance but compared to the actual fiber quality of LuAG:Ce there is still a lot of space for improvements. Due to this, the optimization of LYSO:Ce fibers will start soon. For this process, a lot of the experiences made with LuAG:Ce can be used.

6. Coincidence readout of both fiber ends
For the developed high quality fibers made of LuAG:Ce, a readout at both fiber ends is possible, even for 23 cm long fibers. Fig 11 shows the recorded signal distribution at both SiPMs placing a $^{241}$Am source either close to SiPM 1 or in the middle of a 23 cm long fiber. The correlation between the signals of both SiPMs is shown for both cases in addition. Even after a distance of 23 cm, a signal well separated from the background can be observed. Fig. 12 (left) shows the obtained attenuation curves for both 50 $\mu$m SiPMs and the behavior of the sum of both signals. In the latter case, one would expect a symmetric function for an homogeneous fiber. The asymmetry indicates an increased attenuation near SiPM 2. Presenting the logarithm of the ratio of the two amplitudes $A_1$ and $A_2$, respectively, one can directly determine the attenuation coefficient according to the relation $\ln(A_1/A_2) = \ln([LY_0e^{-\mu x}]/[LY_0e^{-\mu (L-x)}]) = -2\mu x + \mu L$, which predicts a linear function for a constant value of $\mu$, depending on the distance $x$ to SiPM 1. Fig. 12 (right) indicates instead three regions of different attenuation coefficients and confirms the increase of the attenuation coefficient close to SiPM 2. Nevertheless, knowing the attenuation characteristics of the fiber, the logarithm of the amplitude ratio can be used to reconstruct the point of interaction within the fiber. For the coincidence readout, a time resolution of $\sigma \sim 800$ ps was achieved for a typical light output of 120 photons seen by the 50 $\mu$m SiPM.

7. Response to low energetic gamma sources
An important aspect is the response of the fibers to low energetic gamma rays, in particular for medical applications. In case of the detection of the 511 keV $\gamma$-rays of a $^{22}$Na source in coincidence with an external scintillation detector, the energy spectra show clearly visible indications of the full energy photo peak (see fig. 13 (left)). This supports the perspectives for the use in medical applications, like PET scanners. The overall response of 511 keV $\gamma$-rays differs significantly from the spectrum obtained using a $^{60}$Co source. The right part of fig. 13 illustrates the measurement using the 662 keV $\gamma$-rays emitted from a $^{137}$Cs source. For a distance of 1 cm between $\gamma$-source and SiPM, a light yield of 270 $\pm$ 25 ph/MeV has been determined with a 50 $\mu$m SiPM for $\odot$ 2 mm fiber wrapped with teflon.
Figure 11. Response of both SiPMs to a $^{241}$Am source placed close to SiPM 1 (left) and in the center of a 23 cm long LuAG:Ce fiber (right), respectively. In addition, the correlation between the signals of both SiPMs is shown for both cases (lower row).

Figure 12. Attenuation curves individually for both SiPMs and the shape of the sum of both signals (left). Dependence of the logarithm of the ratio of the signal amplitudes of both SiPMs as a function of the source location relative to SiPM1 (right).

Figure 13. Response of a $\odot$ 2 mm LuAG:Ce fiber to a $^{22}$Na and a $^{60}$Co source (left) and to a $^{137}$Cs source (right).

8. Conclusion and Outlook
Over the last years, there was a significant improvement of the quality and homogeneity of LuAG:Ce fibers. Fig. 14 (left) shows the attenuation curves for different LuAG:Ce fiber productions from 2013 and 2014 in comparison to a typical curve for LYSO:Ce fibers and to rectangular LYSO:Ce rods cut from a large conventional crystal. The illustration shows, that with an attenuation coefficient of 0.05 cm$^{-1}$ the quality of the best fibers from the most recent production run comes close to the reference value of 0.02 cm$^{-1}$ obtained with the rectangular rods. In addition the right part of fig. 14 summarizes the development of the attenuation coefficient over the last years, which documents an improvement by more than a factor 20. Based on the experience with the growing parameters from the LuAG:Ce production, the optimization of LYSO:Ce fibers can be pushed forward.

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Figure 14. Comparison of the attenuation curves of different fiber production runs of rectangular LYSO:Ce rods cut from a large crystal and polished (left). Development of the fiber quality over the last years (right).

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