Growth and characterization of Ce-doped YAG and LuAG fibers

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Abstract:

Undoped and Ce-doped $Lu_3Al_5O_{12}$ (LuAG) and $Y_3Al_5O_{12}$ (YAG) single crystal fibers were grown by the micro-pulling down technique (μ-PD) with a purpose to fit the design of new dual-redaout calorimeter planned to operate in future experiences of high energy physics. Fibers up to 20 cm in length and 1mm in diameter were grown along [111] direction. Based on the measurements of the attenuation length along the fibers, the growth conditions to improve the fibers quality were selected. Our results showed that the grown fibers have the capability to be used for future detectors.

Keywords: YAG, LuAG, Fibers, Scintillation, Growth, μ-PD

1. Introduction:

Currently various scintillator materials have attracted much interest due to their importance for research at high energy experiments, space exploration, nuclear medicine and high technology industrial applications[1,2,3]. Each application has its own requirements and very often even well-known composition or technique needs to be newly investigated or adapted. Different single crystal materials such as garnets $Y_3Al_5O_{12}$ (YAG) [4], $Lu_3Al_5O_{12}$ (LuAG) [5,6], oxyorthosilicates Lu_2SiO_5, $Lu_{2-x}Y_xSiO_5$ (LSO,LYSO)[7,8,9] and Y_2SiO_5 (YSO) [10] have been intensively studied. Among various inorganic crystalline materials, we choose Ce^{+3} doped YAG and LuAG compositions for their optical and radiation hardness properties. Our previous study showed the improvement possibilities of undoped and Ce-doped LuAG single crystal fibers [5,6]. The attenuation length results were improved by the selection of the best growth pulling rate, heat control and activator concentration. The post growth annealing allowed improving the attenuation length up to length of 1 m.

The calorimeter granularity might be improved by 1 mm fibers diameter of LuAG or YAG with comparable physical properties. However the similarity, the features of the growth methods especially for doped materials play an important role for the final result. Basically, activator or admixtures are more concentrated at the periphery of the fibers. The surface layer impact of thinner fibers might reduce the attenuation length, that is the reason why we investigate the growth of 1mm diameter fibers.

Ce-doped $Y_3Al_5O_{12}$ (YAG:Ce) fiber was reported in the literature as a fast oxide scintillator [11,12]. The density of YAG is 4.56 g/cm^3 and its effective atomic number Z is 35.The emission spectrum at room temperature is peaked around 540 nm. Because of the cost of lutetium oxide, YAG is more cheaper than LuAG, so the YAG:Ce is a promising candidate to substitute LuAG:Ce composition.

In this paper, we report the scintillation properties of LuAG:Ce and YAG:Ce garnet single crystal fibers of 1mm diameter grown by μ-PD technique.

2. Experimental details

The Ce–doped YAG and LuAG single crystal fibers were pulled from the inductively heated crucible with an opening from the bottom of the capillary die. The Micro-pulling down technique growth from melt was provided in the inert Ar atmosphere gas at pulling rate 0.3 mm/min. Fibers length up to 20 cm were obtained, the diameter is about 1 mm and they were seeded grown along <111> crystallographic direction. The starting raw materials were provided from LuAG:Ce and YAG:Ce crackle crystals previously grown by Czochralski method. They were re-melted in the Ir crucible and crystallized into fibers by pulling the melt through the hole of diameter 0.4 mm. To grow the fibers of the required diameter, the capillary die (shaper) was 1 mm in diameter.

The fiber luminescence has been measured using a 420 nm LED (Thorlabs). The light emitted from the LED was transported to the sample through a 2 mm optical fiber bundle with a focusing system composed by a fiber collimator and a 30 mm lens (all from Thorlabs). The light emitted by the sample was collected from one of the sample end sing a VIS-NIR optical fiber (st-opt-8019, Andor) connected to a Shamrock 500i monochromator and to a Newton iCCD.

3. Results

3.1. Crystal growth and optimizing of conditions

The geometrical configuration (shape) of the die is one of the most important parameters for precise fiber shape control. Because of the wettability of YAG and LuAG melt [5], the choice of the modified capillary die of the crucible is essential for a stable fiber diameter growth. So the regulation of the diameter of the fiber is related to the combining of the power applied to the crucible and the pulling rate during the growth process. These growth parameters (power generator and pulling rate) were kept constant to pull fibers with stable meniscus height (h=150µm) and, as a consequence, a stable fiber diameter is grown (Figure 1).The as grown Ce-YAG fiber showed high transparency with yellow color and constant diameter. The Ce-LuAG is also transparent with green color and homogeneous diameter.

Whatever the host composition, macroscopic defects were not observed in the grown transparent fibers. Quite often, in most of the cases, the fiber diameter variation is less than 1.1% (Figure 2), which confirms the stability of the process and the good selected growth parameters.

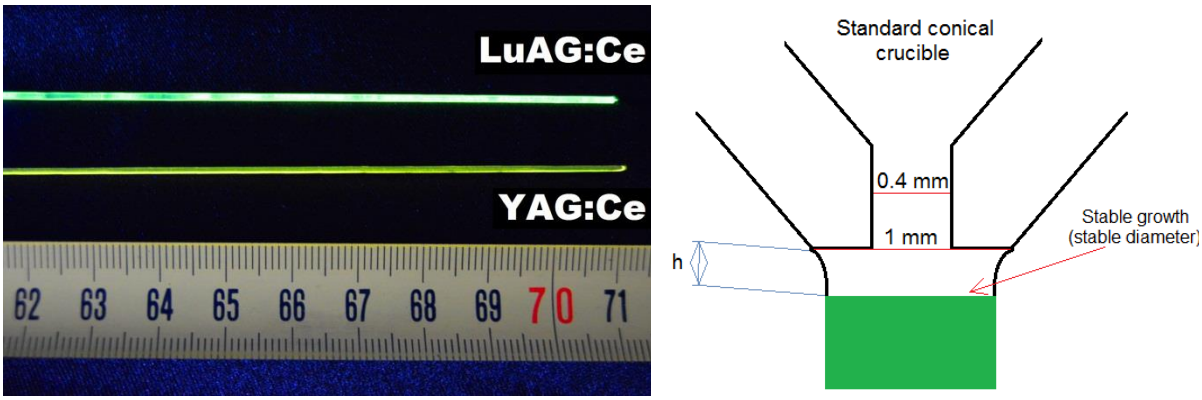

Figure. 1. Grown fibers (left) and crucible design for growing fiber under a stationary stable regime (right).

Depending on the activator and the host characteristics, the distribution of activator content in the host medium can be quite different. The Ce concentration in YAG and LuAG almost never exceeds 1 at.% [13,14]. Besides doping level, the activator can also present variation of spatial distribution within the grown fiber from the melt. This is due to the segregation process (the concentration of the dopant/impurity is different in the liquid and solid phase). The segregation phenomenon is different for atomic-rough and facet growth. The predominance of one of these two processes over the other is highly related to the transverse

thermal gradient. The atomic-rough growth process is the most favored one. It requires a growth interface as flat as possible to minimize the transverse gradient. In both cases, during the growth of Ce-doped YAG and LuAG we didn't observed inclusions or secondary phases such $YAlO_3$ and $LuAlO_3$. But quite often we noted for high Ce (>1%) dopant concentration in LuAG fibers, transversal non-uniformity Ce dopant causing core cladding region in the grown fiber. The Ce concentration will segregate in the periphery of the fiber creating a Ce gradient concentration which strongly affects the optical properties. An another hand this phenomenon is limited in the case of Ce-doped YAG. But by playing by the growth parameters especially pulling rate, it is possible to surmount such problem. Our observation showed that YAG crystal accepts Ce dopant more easily than LuAG in a good agreement with the difference in lattice parameters between YAG and LuAG. This is principally due to difference of the Ce^{3+} size (114.3 pm) which is greater than Y^{3+} (101.9 pm) and considerably larger than the size of Lu^{3+}(97.7 pm) [15] .

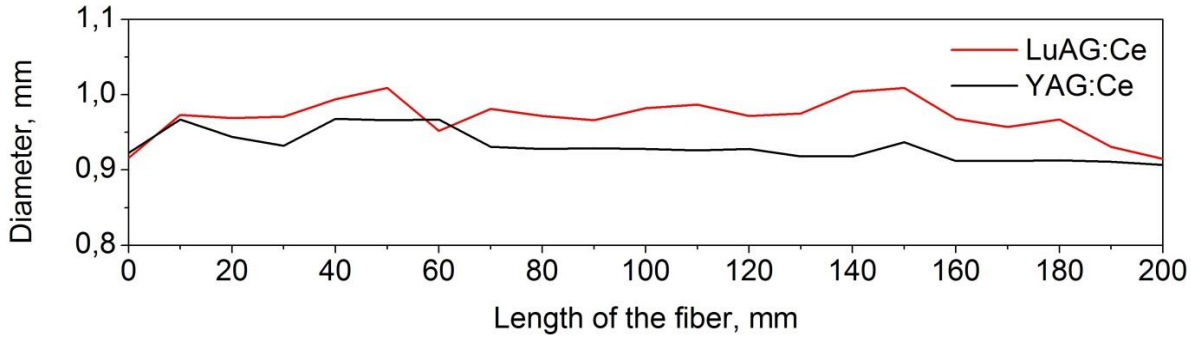

Figure 2. Variation of diameter along the fibers.

3.2. Light propagation

We measured the attenuation property of the fiber. For each interacting position, the photoluminescence spectrum is recorded and the set of measurements is shown in Figure 3 and Figure 4. The change of the spectra shape as a function of the excitation position can be seen. When the interaction position moved away from one end of the fiber, the intensity of luminescence decreased and the emission bands shifted to longer wavelengths. According to the literature [16,17] there are two kinds of light propagation losses: self-absorption and reflection losses in the crystals. The decrease of the intensity of the signal at longer distances between the PMT and the source is related to the both reasons (Figure 3, left). At the normalized

spectra a shift of the luminescence band to the longer wavelength zone due to the reabsorption in the fiber when the interaction position was moved away from one end of the fiber (Figure 3, right). Figure 4 show the emission spectra at excitation position of LuAG:Ce crystal fibers grown with 0.3 mm/min pulling rate. Analogously to the YAG:Ce, in LuAG:Ce there is a redshift of the peaks related to the self-absorption in the material at longer distance between the part of the fiber with original event and the PMT.

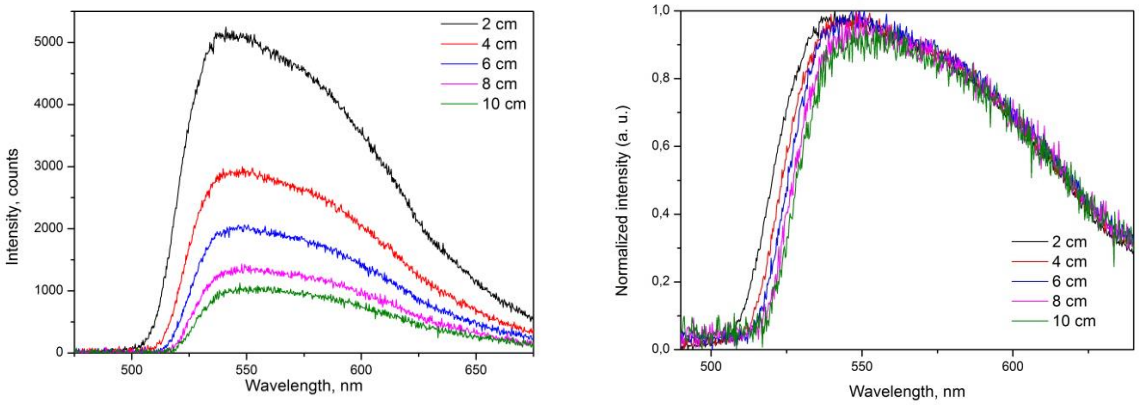

Figure 3. Emission spectra of YAG:Ce crystal fiber grown at pulling rate 0.3 mm/min.

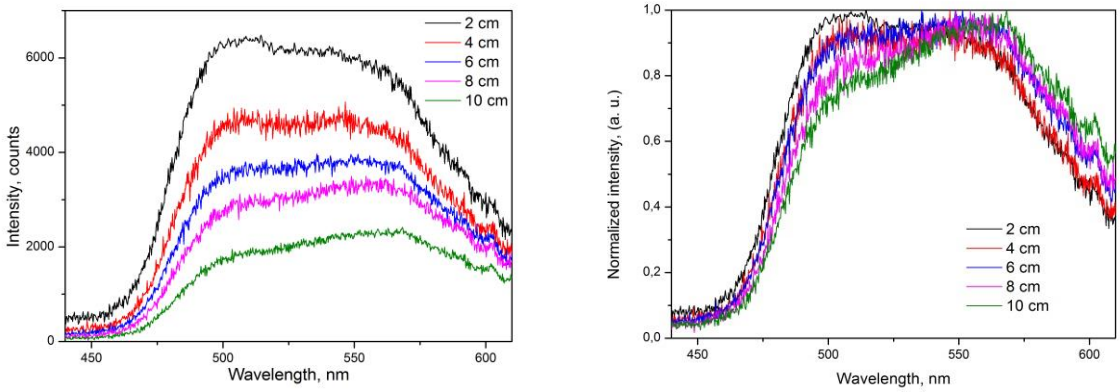

Figure 4. Emission spectra of LuAG:Ce crystal fiber grown at pulling rate 0.3 mm/min,

3. Conclusion:

The results presented in this paper demonstrate that Ce^{3+} doped YAG and LuAG single crystals fibers of 1 mm in diameter grown by micro-pulling down technology present a good light propagation along the whole fiber length. We obtained homogeneous transparent fibers, with stable diameter and high optical grade. The shape of the crucible capillary die allowed a stable diameter growth by taking in account the wetting angle between the Ir capillary die and the melt under specific growth conditions.

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