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Ce^{3+}-doped crystalline garnet films – scintillation characterization using α–particle excitation

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ABSTRACT

Scintillating properties of Ce^{3+}-doped (Lu,Y) aluminum garnet single crystalline films (SCF) were investigated. Thin SCF films of thickness between 1 and 30 μm were grown by a liquid phase epitaxy (LPE) method in various fluxes. The α-particle excitation (mainly 5.4857 MeV line of ^{241}Am) of pulse height spectra is used to measure scintillation response of SCF, especially peak of those α-rays which are totally absorbed in the films. Detailed studies and evaluation of scintillation measurements of large sets of Ce^{3+}-doped SCF (Lu,Y) aluminum garnets showed that at present time (i) YAG:Ce SCF have comparable scintillation properties as YAG:Ce single crystals, especially their N_{phels} photoelectron yields are the same while (ii) scintillation properties of LuAG:Ce SCF do not reach those of LuAG:Ce single crystal.

1. Introduction/scope

Ce^{3+}-doped (Lu,Y) aluminum garnet crystals belong to fast, with high or medium light yield, mechanically hard and chemically stable (non-hygroscopic) single crystal scintillators (Autrata et al., 1983; Moszynski et al., 1994; Zorenko, 2005; Nikl et al., 2009, 2000). Especially, $Lu_3Al_5O_{12}$:Ce crystal (LuAG:Ce) belongs to heavy scintillators due to its high density (6.67 g/cm^3) and high $Z_{eff} = 59$ (Nikl et al., 2009). New development in X-ray imaging applications of μm or even sub-μm resolution (Koch et al., 1998; Kucera et al., 2008; Nikl et al., 2009) needs very thin single crystalline plates or epitaxial layers or films of thickness a few tenths μm and down to 10 μm. Due to good mechanical and chemical properties of (Lu,Y) aluminum garnet single crystals (SC) it is possible to prepare SC plates of 30 μm thin but thinner plates must be coupled (glued) with glass base (Crytur Ltd, 2009).

Besides very thin SC plates from Czochralski grown crystals the another possibility to obtain very thin single crystalline layers (or films – SCF) is offered by a liquid phase epitaxial technology (LPE) (Robertson and van Tool, 1984; Koch et al., 1998; Kucera et al., 2008). The LPE technology allows to grow (Lu,Y) garnet films with much lower content of antisite defects compared with SC plates (Babin et al., 2007). SCF films are grown at much lower temperatures (around 1000 °C) than those of garnet SC where their melting points are around 1900 °C. Now, the LPE technology is being developed further and, different PbO-, BaO- and MoO_3-based fluxes are used (Kucera et al., 2008; Mares et al., 2007a; Prusa et al., 2009). The latest development also shows that other dopants can be used as e.g. Bi^{3+} (Zorenko et al., 2009).

This paper has two main goals: (i) presentation and description of the way of measurements and characterization of scintillation properties of SCF (Lu,Y) garnet films using α- particle excitation and (ii) to summarize and evaluate scintillation response of large sets of LuAG:Ce and YAG:Ce SCF films grown by LPE technology under different conditions, especially an influence of the flux composition (Kucera et al., 2008; Prusa et al., 2008).

2. Experimental

Scintillation characteristics and measurements of thin SCF of (Lu, Y) aluminum garnets are not possible using γ-ray lines excitation due to their small linear attenuation absorption (D'Ambrosio et al., 2000; Mares and D'Ambrosio, 2007). Energy of γ-ray lines is mainly deposited in the substrates (undoped LuAG or YAG SC) on which SCF are grown. The only possibility to measure scintillation response of the films (N_{phels} photoelectron yield, FWHM or N_{phels} yield time development (Mares et al., 2007a,b)) is to use that kind of excitation which fully deposit the energy in the films itself (Shimizu

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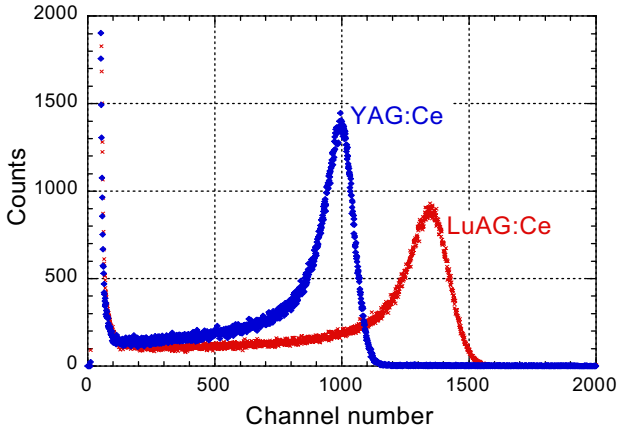

Fig. 1. Pulse height spectra of LuAG:Ce and YAG:Ce reference single crystals excited using α-particle excitation of 5.4857 MeV of ^{241}Am.

et al., 2005). Measured set-up consists of a HPMT (Hybrid Photo-Multiplier Tube), shaping amplifier ORTEC 672, MCB 927™ buffer and PC. This set-up is used in γ-ray excited measurements of the pulse height spectra (Mares et al., 2004). But for scintillation measurements of SCF we must use the α-particle excitation source, especially the energy line 5.4857 MeV of ^{241}Am. This α-particle source is put directly on surface of the films (Prusa et al., 2008, 2009).

The α-particle excited pulsed height spectra of Czochralski grown LuAG:Ce and YAG:Ce single crystals (Crytur Ltd, 2009) are presented in Fig. 1 (these spectra are used as the reference ones to those of measured SCF films). (Lu,Y)AG:Ce SCF have penetration range of α-particles between 9 and 13 μm (Prusa et al., 2009) and thickness of some of measured films is below the penetration range of α-particles. Fig. 2 presents pulse height spectrum of LuAG:Ce SCF of 5.2 μm thickness. This spectrum was decomposed into three Gaussians: (i) two clearly seen G2 and G3 peaks and (ii) less intense G1 one. G3 peak at channel number ~608 is due to total absorption of α-particles inside SCF while G2 one is due to α-rays moving perpendicularly to surface of SCF through (5.2 μm thickness) into substrate. Wide G3 peak is probably due to UV scintillation emission of the substrate.

Majority of measurements of LuAG:Ce SCF was carried out using optical cut-off filter ~480 nm and one example is shown in Fig. 3. An effect of optical filter influences shape of α-particle peaks compared if no filter is used because: (i) it decreases pulse height spectra

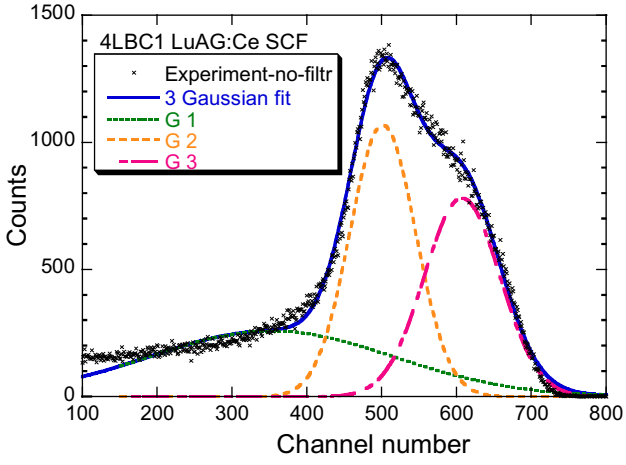

Fig. 2. Pulse height spectrum of LuAG:Ce SCF of 5.2 μm thickness prepared using BaO-based flux and its decomposition into three Gaussians (measured by 5.4857 MeV of ^{241}Am and 0.5 μs shaping time).

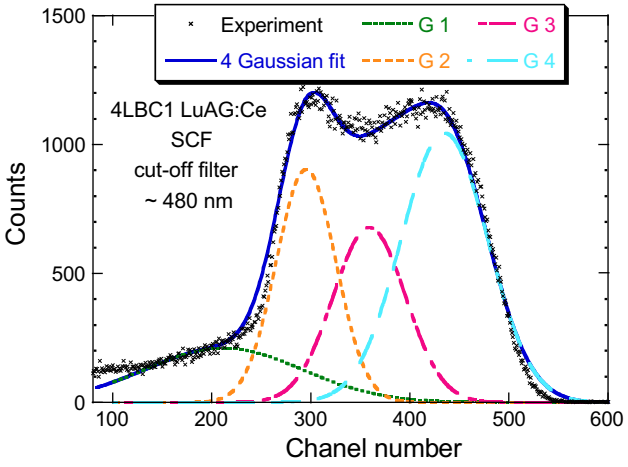

Fig. 3. Pulse height spectrum of LuAG:Ce SCF of 5.2 μm thickness prepared using BaO based flux measured with optical cut-off filter 480 nm and its decomposition into four Gaussians (measured by 5.4857 MeV of ^{241}Am and 0.5 μs shaping time).

intensity of about 25% and (ii) we only measure Ce^{3+} scintillation (emission) and not the UV one of the substrate (its intensity is much less compared that without filter use). The above given reasons (i) and (ii) allows more precise measurements of Ce^{3+} scintillation response (peak of penetrating α-particles) of (Lu,Y) aluminum garnet SCF. Pulse height spectrum in Fig. 3 was decomposed into four Gaussians G1–G4. Peaks G2 and G4 are due to α-particles moving either perpendicularly to film surface or in those directions where penetrate in films, respectively. G3 peak is due to those α-particles which move in directions between those of G2 and G4 peaks while G1 peak is probably due to weak rest of UV substrate emission. Now, we measure and compare properties of Ce^{3+}-doped (Lu,Y) aluminum garnet SCF using optical cut-off filter.

3. Results of measurements

In this paper we summarize our long term research of Ce^{3+}-doped SCF of (Lu,Y)AG (Mares et al., 2007a; Prusa et al., 2008, 2009). A few tens of both YAG:Ce and LuAG:Ce SCF samples were investigated. Majority of them was grown by us (Kucera and Nitsch) but some others were from Department of Electronics, Ivan Franko National University from Lviv, Ukraine (Zorenko, 2005; Zorenko et al., 2009). All SCF samples were grown by the LPE method at different temperatures (around 1000 °C) in three various fluxes: (i) $PbO–B_2O_3$, (ii) $BaO–B_2O_3$ and (iii) $MoO_3–Li_2MoO_4$. Parameters and basic scintillation properties of the best SCF or some others are given in Tables 1 and 2.

Now, the LPE technology allows to prepare well defined and good quality YAG:Ce SCF. Their N_{phels}/MeV photoelectron yields and FWHM (at 5.4857 MeV of ^{241}Am) are the same or even higher than those of YAG:Ce reference single crystal (see Table 1). The only deviation is higher time difference of N_{phels} yield in the shaping time

Table 1
Parameters and scintillation properties of YAG:Ce SCF and comparison with those of YAG:Ce single crystal measured at 1 μs shaping time under ^{241}Am 5.4857 MeV excitation (SC – single crystal, samples in rows 3 to 6 are SCF).

Sample	Thickness [μm]	N_{phels} per MeV (phels)	FWHM (%)	N_{phels} change 0.5–10 μs (%)	Flux based on
YAG:Ce SC	1 mm	349	11	~ 10	CZ grown
4C2	8	317.6	9.6	60	PbO
5YBC1	4.4	402.5	10.8	26.5	BaO
1MC13	3.5	372.2	9.5	47.5	MoO_3
N42-7	58	374.1	17.6	a few	PbO

Table 2
Parameters and scintillation properties of LuAG:Ce SCF and comparison with those of LuAG:Ce single crystal measured at 1 μs shaping time under 5.4857 MeV excitation of ^{241}Am (SC – single crystal, samples in rows 3 to 6 are SCF).

Sample	Thick-ness μm	N_{phels} per MeV (phels)	FWHM (%)	N_{phels} change 0.5–10 μs (%)	Flux based on
LuAG:Ce SC	1 mm	471	10.7	76	CZ grown
4LBC3	6	236	16.3 (^{239}Pu)	65.7	BaO
6Lu1	14.6	77	21	~ 5	PbO
LY6	17	228	10.5	20.8	PbO
LY16	27	152.6	17.6	6.8	PbO

range 0.5–10 μs. Some measured samples exhibit various defects or morphology deviations. Detailed YAG:Ce measurements of SCF (Prusa et al., 2009) showed that the best samples were grown using BaO-based flux.

Table 2 presents parameters and scintillation properties of Ce^{3+}-doped LuAG SCFs. Here, their scintillation properties do not reach those of LuAG:Ce single crystal. SCF N_{phels}/MeV yields of SCF are about one half of that of LuAG:Ce crystal. We measured two groups of LuAG:Ce SCF, one of them was prepared by us the second one is from Lviv (Zorenko, 2005). Some of LuAG:Ce SCF exhibit large N_{phels} time increase between 0.5 and 10 μs (around 70%) but thicker SCF have much less time difference (see Table 2). Again, the highest N_{phels} yield exhibit LuAG:Ce SCF grown from BaO-based flux.

4. Conclusions

Detailed research of large sets of Ce^{3+}-doped (Lu,Y)AG SCF was carried out using α-particle excitation. Scintillation properties of SCFs were evaluated from pulse height spectral bands arising from α-particles depositing their energy in the films (their paths are equal or larger than the penetration range). YAG:Ce SCF grown in different fluxes and conditions reached comparable scintillation properties, compared with their single crystal analogue. Further development is necessary however, in case of LuAG:Ce SCFs where their scintillation N_{phels}/MeV yield is in average about half of that of LuAG:Ce crystal.

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References

Autrata, R., Schauer, P., Kvapil, J., Kvapil, J., 1983. Cathodoluminescent efficiency of $Y_3Al_5O_{12}$ and $YalO_3$ single crystals in dependence on Ce^{3+} and other dopant concentrations. Cryst. Res. Technol. 18, 907–913.
Babin, V., Gorbenko, V., Makhov, A., Mares, J.A., Nikl, M., Zazubovich, S., Zorenko, Yu, 2007. Luminescence characteristics of Pb^{2+} centres in undoped and Ce^{3+}-doped $Lu_3Al_5O_{12}$ single-crystalline films and $Pb^{2+} \rightarrow Ce^{3+}$ energy transfer. J. Lumin. 127, 384–390.
Crytur Ltd., 2009. Booklets of Crytur or see www.crytur.com, Palackeho 175, 511 19 Turnov, Czech Republic.
D'Ambrosio, C., De Notaristefani, F., Leutz, H., Puertolas, D., Rosso, E., 2000. X-Ray detection with a scintillation YAP-window hybrid photomultiplier tube. IEEE. Trans. Nucl. Sci. 47, 6–10.
Koch, A., Raven, C., Spadne, P., Snigirev, A., 1998. X-ray imaging with submicrometer resolution employing transparent luminescent screens. J. Opt. Soc. Am. A. 15, 1940–1951.
Kucera, M., Nitsch, K., Kubova, M., Solovieva, N., Nikl, M., Mares, J.A., 2008. Ce-doped YAG and LuAG epitaxial films for scintillation detectors. IEEE. Trans. Nucl. Sci. 55, 1201–1205.
Mares, J.A., D'Ambrosio, C., 2007. Hybrid photomultipliers – their properties and application in scintillation studies. Opt. Mat. 30, 22–25.
Mares, J.A., Beitlerova, A., Nikl, M., Solovieva, N., D'Ambrosio, C., Blazek, K., Maly, P., Nejezchleb, K., De Notaristefani, F., 2004. Scintillation response of Ce-doped or intrinsic scintillating crystals in the range up to 1 MeV. Radiat. Meas. 38, 353–357.
Mares, J.A., Beitlerova, A., Nikl, M., Solovieva, N., Nitsch, K., Kucera, M., Kubova, M., Gorbenko, V., Zorenko, Yu, 2007a. Scintillation and optical properties of YAG: Ce films grown by liquid phase epitaxy. Radiat. Meas. 42, 533–536.
Mares, J.A., Beitlerova, A., Nikl, M., Vedda, A., D'Ambrosio, C., Blazek, K., Nejezchleb, K., 2007b. Time development of scintillating response in Ce-or Pr-doped crystals. Phys. Stat. Sol. (c) 4, 996–999.
Moszynski, M., Ludziejewski, T., Wolski, D., Klamra, W., Norlin, L.O., 1994. Properties of YAG: Ce scintillator. NIM Phys. Res. A 345, 461–467.
Nikl, M., Mihokova, A., Mares, J.A., Vedda, A., Martini, M., Nejezchleb, K., Blazek, K., 2000. Traps and timing characteristics of LuAG: Ce^{3+} scintillator. Phys. Stat. Sol. (a) 181, R10–R12.
Nikl, M., Tous, J., Mares, J.A., Prusa, P., Mihokova, E., Blazek, K., Vedda, A., Zorenko, Yu., Gorbenko, V., Babin, V., 2009. $Lu_3Al_5O_{12}$-based materials for high 2D-resoultion scintillation detectors. Proc. SPIE, Vol. 7310_7, pp. 731008-1–10.
Prusa, P., Cechak, T., Mares, J.A., Nikl, M., Beitlerova, A., Solovieva, N., Zorenko, Yu., Gorbenko, V.I., Tous, J., Blazek, K., 2008. The α-particle excited scintillation response of the liquid phase epitaxy grown LuAG: Ce thin films. Appl. Phys. Lett. 92, 041903-1-3.
Prusa, P., Nikl, M., Mares, J.A., Kucera, M., Nitsch, K., Beitlerova, A., 2009. The α-particle excited scintillation response of YAG: Ce thin films grown by liquid phase epitaxy. Phys. Stat. Sol. A 206, 1494–1500.
Robertson, J.M., van Tool, M.V., 1984. Cathodoluminescent garnet layers. Thin Solid Films 114, 221–240.
Shimizu, S., Suzuji, M., Koizumi, Y., Ishibashi, H., Kubota, S., 2005. Light output and decay curve of GSO: Ce under electron, proton, alpha particle and fission fragment excitations. NIM Phys. Res. A 537, 57–60.
Zorenko, Yu, 2005. Luminescence of isoelectronic impurities and antisite defects in garnets. Phys. Stat. Sol. 2, 375–379.
Zorenko, Yu., Mares, J.A., Kucerkova, R., Gorbenko, V., Savchyn, V., Voznyak, T., Nikl, M., Beitlerova, A., Jurek, K., 2009. Optical, luminescence and scintillation characteristics of Bi-doped LuAG and YAG single crystalline films. J. Phys. D: Appl. Phys. 42 (7), 075501.