Prospective scintillators for low-energy BSE detectors

Authors: Ondřej Lalinský (1), Petr Schauer (1), Miroslav Kučera (2), Zuzana Lučeničová (2), Martin Hanuš (2)

1. Department of Electron Microscopy, Institute of Scientific Instruments of the CAS, Brno, CZECH REPUBLIC 2. Division of Magnetooptics, Institute of Physics, Charles University, Prague, CZECH REPUBLIC

DOI: 10.1002/EMC2016.0763

Corresponding email: xodr@isibrno.cz

Keywords: Scintillator, cathodoluminescence, GAGG:Ce, epitaxial layer, low-energy electron, BSE detector

Cerium activated bulk single crystals of yttrium aluminium garnet (YAG:Ce) $Ce_x:Y_{3-x}Al_5O_{12}$ are widely used as scintillators for the detection of backscattered electrons (BSE). In the electron microscopy research of nanomaterials, biomaterials or semiconductors, low energy (units of keV) electron beam imaging is often necessary. Because BSE detectors are mostly non-accelerating or low-accelerating, electrons with approximately the same energy as primary beam (PB) have to be detected. However, commonly used YAG:Ce single crystal strongly loses its light yield (LY) with the decrease of the PB energy [1]. As possible available alternatives for this application, bulk single crystals of YAIO₃:Ce (YAP:Ce) and CRY018 can be predicted. However, similar LY sink can be expected also with these scintillators.

There are two main reasons, why this occurs. Firstly, slower electrons don't have enough energy to pass through the relatively thick conductive layer on the scintillator surface. Therefore, thinner conductive layer has to be used. Secondly, commonly available scintillators suffer from structural defects that are created mostly due to surface damage (as a result of its grinding, polishing, purification or contamination) or already during the own bulk single crystal growth.

The influence of all of these defects on cathodoluminescence (CL) properties can be eliminated by the scintillators in form of thin single crystalline films because, as shown previously [2], the concentration of these defects decreases with the decreasing temperature of the crystal growth. Therefore, single crystalline epitaxial films have attracted a lot of attention recently because the growth temperature of these films is about a half (1000 °C) of the bulk ones (2000 °C). Moreover, appropriate doping of the garnet structure can suppress the influence of the defects on the CL properties.

For the purpose of this work, bulk single crystals of YAG:Ce, YAP:Ce and CRY018 were studied. Results were compared with those of promising cerium activated single crystalline films of gadolinium aluminium gallium garnet (GAGG:Ce) $Ce_x:Gd_{3-x}Ga_yAl_{5-y}O_{12}$. These films were grown by the isothermal dipping liquid phase epitaxy onto YGG substrates from lead-free BaO-B₂O₃-BaF₂ flux [3]. These specimens were coated with conductive layers of different composition and different thicknesses. Properties of these layers are in the table shown in Table 1.

The specimens were excited by an electron beam with energy in range from 0.8 to 10 keV using a specialized CL apparatus [4]. In this energy range, CL LY of YAG:Ce were measured for coating layers of different composition and different thicknesses (Fig. 1). Moreover, CL spectra (Fig. 2) and CL intensity decays (Fig. 3) have been measured for all presented specimens.

It was shown, that the coating layer with thickness of only units of nm has to be used to allow low-energy BSE penetrating the layer without significant losses. Moreover, it was shown, that the GAGG:Ce film with balanced Ga content shows excellent scintillation properties where the effect of unwanted structural defects was suppressed, the spectrally corrected CL LY value exceeded 160 % of the commercially available bulk YAG:Ce single crystal, and CL decay was dominated by a fast component with 50 ns decay time which is close to that of Ce^{3+} (5d-4f) photoluminescence decay. Thanks to these excellent CL properties at PB energy of 10 keV, GAGG:Ce single crystalline films are new prospective scintillators suitable for low-energy BSE detectors. This research is in progress, therefore other results at different PB energies will be presented at the conference.

Acknowledgements: The research was supported by the Technology Agency of the Czech Republic (TE01020118), by Czech Science Foundation (projects GA16-05631S, GA16-15569S) and by Ministry of Education, Youth and Sports of the Czech Republic (project LO1212) and by European Commission (project CZ.1.05/2.1.00/01.0017). The authors thank the company CRYTUR, s.r.o., for supplying with the specimens of bulk single crystals.

References:

[1] G. F. J. Garlick, Brit. J. Appl. Phys. 13 (1962) 541-547.

[2] M. Nikl, E. Mihokova, J. Pejchal, A. Vedda, Y. Zorenko, K. Nejezchleb, Phys. Status Solidi B 242 (2005) R119-R121.

[3] M. Kucera, K. Nitsch, M. Kubova, N. Solovieva, M. Nikl, J.A. Mares, IEEE Trans. Nucl. Sci. 55 (2008) 1201.
[4] J. Bok, P. Schauer, Rev. Sci. Instrum. 82 (2011) 113109.

Film	Atomic number	Atomic weight	Density (g/cm³)	Thickness (nm)	Trans. of el. of 700 eV (%)
Aluminium (Al)	13	26.98	2.7	3.8	63.2
				5.0	48.0
				10.0	15.1
				15.0	1.3
Scandium (Sc)	21	44.95	2.99	3.0	69.0
				5.0	45.1
Nickel (Ni)	28	58.69	8.91	1.0	74.8
				2.0	50.2
Indium Tin	24.21*	55.11*	7.16	4.0	6.24
Oxide (ITO)				7.0	0.0
*weighted values	;				

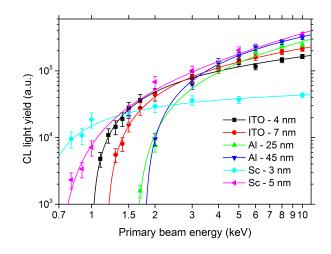
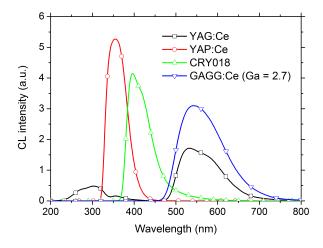


Table 1: Properties of commonly used elements for coating layers for scintillators. Data in last column were simulated by the Monte Carlo method that used the elastic single scattering model with the Mott cross-sections and the Bethe slowing-down approximation.

Fig. 1: The cathodoluminescence (CL) light yield of the YAG:Ce single crystal scintillator coated with layers of different composition and different thicknesses as a function of the primary beam energy.



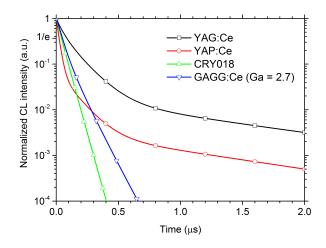


Fig. 2: Cathodoluminescence (CL) spectra of Ce0.02:Gd3Ga2.7Al2.3O12 (GAGG:Ce) compared with commercially available YAG:Ce, YAP:Ce and CRY018. Spectra were corrected for the apparatus transmissivity and detector spectral sensitivity. Primary beam energy was 10 keV.

Fig. 3: Cathodoluminescence (CL) intensity decays of Ce0.02:Gd3Ga2.7Al2.3O12 (GAGG:Ce) compared with commercially available YAG:Ce, YAP:Ce and CRY018. Excitation time was 50 ns. Primary beam energy was 10 keV.