Radiation Measurements 90 (2016) 132–135

Contents lists available at ScienceDirect

Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeas

# Luminescent properties of LuAG:Yb and YAG:Yb single crystalline films grown by Liquid Phase Epitaxy method

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## HIGHLIGHTS

- Single crystalline films of Yb doped LuAG and YAG garnets were grown by LPE method.
- $Yb^{3+}$ luminescence of LuAG:Yb and YAG:Yb film were studied using synchrotron radiation.
- Basic parameters of $Yb^{3+}$ charge transfer luminescence in LuAG and YAG were determined.

## ARTICLE INFO

*Article history:*
Received 26 October 2015
Received in revised form
24 December 2015
Accepted 26 December 2015
Available online 30 December 2015

*Keywords:*
Luminescence
Liquid phase epitaxy
Single crystalline film
YAG and LuAG garnets
Yb dopant

## ABSTRACT

In this work, investigation of the spectroscopic parameters of the luminescence of $Yb^{3+}$ ions in single crystalline films of $Lu_3Al_5O_{12}$ and $Y_3Al_5O_{12}$ garnets was performed using the synchrotron radiation excitation with the energy in the range of $Yb^{3+}$ charge transitions (CT), exciton range and the onset of interband transitions of these garnets. The basic spectroscopic parameters of the $Yb^{3+}$ CT luminescence in LuAG and YAG hosts were determined and summarized with taking into account the differences in the band gap structure of these garnets.



## 1. Introduction

Single crystals (SC) of $Y_3Al_5O_{12}$(YAG) and $Lu_3Al_5O_{12}$ (LuAG) garnets doped with $Yb^{3+}$ are being considered as promising scintillators for application in the neutrino physics (Kamenskikh et al., 2003; Nikl et al., 2006). For these reasons, the luminescent properties of YAG:Yb and LuAG:Yb SCs were the subject of detailed studies in (Kamenskikh et al., 2003; Nikl et al., 2006) using the synchrotron radiation excitation. Single crystalline films (SCF) of YAG:Yb with a thickness up to 200 μm are also known as thin film disk laser media (Malinowski et al., 2001). However, by now there are no data on obtaining LuAG:Yb SCF and investigation of their luminescent properties. Systematic analysis of the features of luminescent properties of LuAG:Yb and YAG:Yb SCFs in comparison with their SC analogues is also absent. At the same time, these features can be caused by the low-temperature (<1000 °C) growth of these SCFs by the Liquid Phase Epitaxy (LPE) method as distinct from the high-temperature synthesis of SC of Yb-doped garnets from melt (Ashurov et al., 1977; Zorenko et al., 2014). Particularly, the LPE method allows one to obtain Yb doped SCF of LuAG or YAG garnet without $Lu_{Al}$, $Y_{Al}$ and $Yb_{Al}$ antisite defects and low concentration of vacancy type defects (Zorenko et al., 2005; Zorenko et al.,

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http://dx.doi.org/10.1016/j.radmeas.2015.12.032
1350-4487/

2014). For these reasons, these SCFs are excellent model objects for investigation of the basic luminescence behaviour of garnet matrixes and luminescence dopants in them (Zorenko et al., 2005; Zorenko et al., 2007a, 2007b).

In this work, we have performed the investigation of the spectroscopic parameters of the luminescence of $Yb^{3+}$ ions in SCFs of $Lu_3Al_5O_{12}$ and $Y_3Al_5O_{12}$ garnets using the synchrotron radiation excitation with the energy in the exciton range and the onset of interband transitions of the mentioned garnets.

## 2. Growth of LuAG:Yb and YAG:Yb SCFs and experimental technique

The two sets of LuAG:Yb and YAG:Yb SCFs were grown by the LPE method from melt-solutions based on the $PbO–B_2O_3$ flux onto YAG substrates with (111) orientations. Concentration of $Yb_2O_3$ activating oxide ions in the melt solution at the growth of LuAG and YAG SCFs is varied in the 1.0–4.0 at. % and 1.5–15 at. % ranges, respectively. The composition of SCF samples was determined using a JEOL JSM-820 electronic microscope, equipped by an EDX microanalyzer with IXRF 500 and LN2 Eumex detectors. From the analysis of the content of YAG:Yb and LuAG:Yb SCF samples we have also found that the Yb segregation coefficients in the mentioned concentration ranges of $Yb_2O_3$ dopant at LPE growth of these SCFs onto YAG substrates are close to 1.05–0.95 and 1.0–0.85, respectively.

The XRD measurements (spectrometer DRON 4, $Cu_{K\alpha}$ X-ray source) were used for characterization of the structural perfection of LuAG:Yb and YAG:Yb SCF samples (Fig. 1a). All the LuAG:Yb and YAG:Yb SCFs under study show single crystalline structure and good structural quality due to the same wideness of the peaks as in the case of YAG substrate. From the respective XRD patterns of LuAG:Yb SCF we have also calculated the misfit between the lattice constant of LuAG:Yb SCF and YAG substrate $\mathbf{\Delta a = (a_{SCF}–a_{sub})/a_{sub}}$*100% which is equal to 0.82% (Fig. 1).

For investigation of the optical properties of LuAG:Yb and YAG:Yb garnets, the two high-quality samples were selected: the LuAG:Yb (1%) T10 SCF with a thickness of 14.4 μm and YAG:Yb (1.5%) B13 SCF with a thickness of 28 μm. The comparative analysis of reflectivity and luminescent properties of selected LuAG:Yb and YAG:Yb SCF samples has been carried out at 300 and 8 K under excitation by pulsed (0.126 ns) synchrotron radiation with an energy of 3–25 eV at the Superlumi station in HASYLAB, DESY. Due to the different bunch mode of synchrotron radiation at investigation of LuAG:Yb and YAG:Yb SCF samples, the time intervals for registration of the $Yb^{3+}$ luminescence in these SCFs between SR pulses were different and equal to 0–200 ns and 0–100 ns, respectively. Namely, the luminescence excitation spectra were registered in the integral (2.2–200 ns), fast (2.2–42 ns) and slow (100–170 ns) intervals for LuAG:Yb.

SCF and in the integral (1.2–100 ns), fast (2.2–12 ns) and slow (60–100 ns) intervals for YAG:Yb SCF (see legends of Figs. 3 and 4, respectively). The excitation spectra and were corrected on the sensitivity of monochromator and detector units but emission spectra were not corrected.

Excellent structural and optical properties of the SCF samples under study can be also confirmed by the reflectivity spectra of LuAG:Yb SCF at 300 and 8 K (Fig. 1b). Specifically, the exciton peak at 7.925 eV is clearly resolved in the reflectivity spectra from the free grown surface of this LuAG:Yb SCF. The observation of so well resolved exciton peak is fully impossible in the case of investigation of the reflectivity of LuAG or YAG crystals due to the imperfections of sample surface introduced by mechanical polishing and significantly larger concentration of intrinsic defects (Kolobanov et al., 2006; 2007).

## 3. Luminescent properties of LuAG:Yb and YAG:Yb SCFs

The characteristic $Yb^{3+}$ charge transfer (CT) luminescence is observed in the emission spectra of LuAG:Yb and YAG:Yb SCFs in the two bands peaked at 344, 492 nm and 342, 495 nm, respectively, under excitation in the ranges of respective absorption bands (195–230 nm), in the exciton range or in the range of interband transitions of YAG and LuAG garnet hosts (Fig. 2). The well known emission band at 1030 nm is also observed in the luminescence spectra of LuAG:Yb and YAG:Yb SCFs in the IR range and caused by the $^2F_{5/2} \rightarrow {}^2F_{7/2}$ 4f-4f transitions of $Yb^{3+}$ ions.

The excitation spectra of the $Yb^{3+}$ CT luminescence in LuAG:Yb SCF (Fig. 3) and YAG:Yb SCF (Fig. 4) SCF contain the dominant CT absorption bands peaked approximately at 5.85 eV in LuAG:Yb and 5.91 eV in YAG:Yb as well as the bands in the exciton range peaked correspondingly at 7.3 eV and 7.01 eV in LuAG:Yb and YAG:Yb SCFs, respectively, which are caused by creation of the excitons bound with $Yb^{3+}$ ions. It is interesting to note that the band peaked at 5.62 eV is also well resolved in the excitation spectra of the $Yb^{3+}$ luminescence in LuAG:Yb SCF (Fig. 3). This band is especially dominated in the spectra of slow component of emission (Fig. 3). The determination of the origin of this band needs more detailed investigations. The increase of the CT luminescence intensity in the above 15 eV range in the excitation spectra of LuAG:Yb and YAG:Yb SCFs (Figs. 3 and 4) is caused by the multiplication of electron-hole pairs in these garnets.

The decay kinetics of the CT luminescence in LuAG:Yb (Fig. 5a) and YAG:Yb (Fig. 5b) SCFs shows strong dependence of the decay

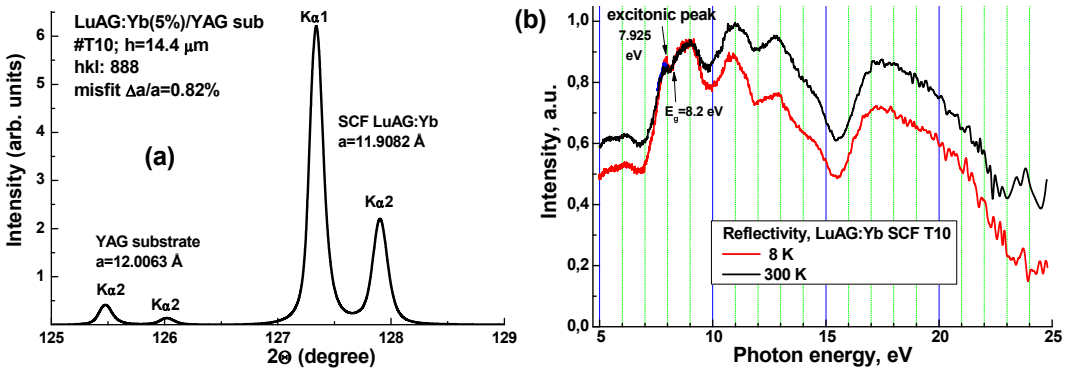


**Fig. 1.** (a) - XRD pattern of (888) plane of LuAG:Yb SCF sample with a thickness of 14.4 μm grown onto YAG substrate with (111) orientation. (b) – reflectivity spectra of LuAG:Yb SCF at 8 and 300 K.