Studies of LSO:Tb radio-luminescence properties using white beam hard X-ray synchrotron irradiation

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A radio-luminescence set-up was installed at the synchrotron light source ANKA to characterise scintillators under the high X-ray photon flux density of white beam synchrotron radiation. The system allows for investigating the radio-luminescence spectrum of the material under study as well as analysing *in situ* changes of its scintillation behaviour (e.g. under heat load and/or intensive ionising radiation). In this work we applied the radio-luminescence set-up for investigating the radiation damage effects on the luminescence properties of a new kind of thin single crystal scintillator for high resolution X-ray imaging based on a layer of modified LSO (Lu_2SiO_5) grown by LPE (Liquid Phase Epitaxy) on a dedicated substrate within the framework of an EC project ($SCIN^{TAX}$)[1].

Keywords: radio-luminescence; white beam synchrotron radiation; LSO

Introduction

Third generation synchrotron light sources offer new possibilities for different X-ray imaging techniques thanks to their high brilliance and partial spatial coherence. These techniques (e.g. microtomography with absorption or phase contrast and holotomography) demand highly efficient X-ray detectors with a spatial resolution in the micrometer and even submicrometer range [1, 2, 3, 4]. In addition, microtomography calls for fast and on-line working detectors, due to the necessity to collect fast and efficient several hundred images for a single tomogram. X-ray energies are typically between 6-30 keV for absorption radiography and up to 60 keV in phase contrast mode [5, 6, 7]. A successful approach to achieve X-ray imaging with submicrometer resolution is given by the combination of a transparent luminescent screen (single crystal scintillator) with diffraction limited microscope optics to magnify the luminescence image onto a CCD (Charge Coupled Device) camera. At

present, the best spatial resolution measurements of 0.8 and 0.5 μm were achieved by A. Koch et al. by using a commercially available 5 μm YAG:Ce^{3+} (Ce doped $Y_3Al_5O_{12}$) scintillating single crystal (on the top of an undoped YAG substrate) and a thin LAG ($Lu_3Al_5O_{12}$) scintillator grown by LPE on a YAG substrate and doped by Tb^{3+} or Eu^{3+} [5, 6, 8, 9]. To further increase the spatial resolution and the DQE (Detective Quantum Efficiency) of an X-ray imaging system, the research should be focused on scintillating thin layers with a high atomic number, high density, high light yield (amount of X-ray photon energy converted into visible light photons) and outstanding optical surface quality [7]. At the present stage, LSO (Lu_2SiO_5) single crystal represents one of the best candidates to develop a high performance X-ray imaging system, thanks to its density (7.4 g/cm^3) and effective atomic number (65.2) [10, 11]. At 20 keV, the LSO absorption efficiency is 2.2 times higher than the YAG efficiency and 1.34 times higher than the LAG efficiency. At 60 keV it is 11 times higher than the YAG efficiency and 1.37 times more than the LAG efficiency [7]. Currently, a LSO:Ce scintillator is used in the Hamamatsu AA50 Super high-resolution X-ray detector where it is glued on a carbon plate [12]. In such a configuration the best achievable resolution is > 1 μm due to the impossibility to reduce by mechanical polishing the LSO thickness below 10 μm.

Within the framework of an EC project ($SCIN^{TAX}$ - 6^{th} framework program) we have developed a new thin single crystal scintillator for high resolution X-ray imaging based on a layer of modified LSO grown by LPE on a dedicated substrate with thickness ranging between a few microns up to 100 μm [13].

One of the most important parameters to be analysed during the characterisation of a new scintillating material is represented by its radiation resistance, which regards mainly the changes and the performance instabilities due to the induced absorption resulting from material irradiation and colour centre creation. While this parameter has been considered mainly in the research on scintillators for high-energy physics, it should be noted that it has importance also in medical imaging techniques and in the case of industrial flaw detection or synchrotron beam diagnostics applications as well [18]. To characterise the newly developed scintillators under the high X-ray photon flux density of white beam synchrotron radiation, we designed a radio-luminescence set-up where the scintillator is excited by the synchrotron radiation. In this work we report the results of the investigation performed on a 12 μm thick LSO:Tb film, an 8 μm LSO:Tb,Sm film and a 500 μm substrate. Further details on the substrate are accessible via [13] (a patent application is pending).

Experimental set-up

The synchrotron light source ANKA (*Angstroemquelle Karlsruhe*) is located at the Research Centre Karlsruhe (*Forschungszentrum Karlsruhe / K.I.T.*) and is operated with ring electron energy of 2.5 GeV and beam currents of 180-80 mA. The dipole bending magnets are working with 1.5 T magnetic field and the resulting critical energy is E_c=6.2 keV [19]. The synchrotron light produced by ANKA is used for a wide range of analytical methods as well as for microfabrication techniques. At present 13 beamlines are operative and offering competences which include the areas X-ray spectroscopy, x-ray diffraction and imaging, infrared spectroscopy and x-ray lithography.

The radio-luminescence measurements described in this work were performed at the TopoTomo beamline, whose characteristic features are the first slit system in the front end (6 m distance to the source), no optical component inside the 30 m long beam

path and a second in-vacuum slit system followed by one Be window directly in front of the experiment (cf. Figure 1). The available energy spectrum ranges between 1.5 up to 50 keV and the total flux at sample position is in the order of 10^{16} ph/s (5 mm x 10mm). Further details can be found in [14, 15, 16, 21].

The radio-luminescence set-up is composed of a *Jobin Yvon CP140-104* spectrograph working in the (250-800) nm range, coupled to a high resolution cooled CCD camera, type *PCO4000* positioned in the focal plane of the spectrograph. The rather simple layout results in a robust unit which can be easily installed at different light sources and aligned within a short period of time. Due to the camera used, fast data acquisition is possible, allowing one to follow changes in the emitted spectra *in situ* with a time resolution down to the millisecond scale.

The radio-luminescence measurements were performed by exciting the scintillating light with a synchrotron beam size equal to 4 x 4 mm^2. The CCD detector was integrated for 25 ms and 11 images were acquired for every spectrum in order to optimise the SNR. The calibration of the CCD pixel position in terms of wavelength was performed by using the known laser wavelengths of 632 nm (He-Ne laser) and 532 nm wavelengths (green laser). During the overall measurements the the ANKA ring current was monitored in order to compare the spectra recorded at different times in a quantitative manner. In addition, preliminarily to the radiation damage tests we have estimated an error on the measurements equal to 1 %.

Results and discussion

In Figure 2 are reported the radio-luminescence spectra of the investigated samples: the LSO:Tb^{3+} spectrum is characterised by emission lines typical of Tb^{3+} ($4f^8$) transitions $^5D_4 \rightarrow {}^7F_J$ (J=3, 4, 5, 6) and they are qualitatively similar to those observed in [22] in float-zone grown single crystals. The absence of the Tb^{3+} blue emission lines corresponding to the transition $^5D_3 \rightarrow {}^7F_J$ (J=3, 4, 5, 6) is consistent with the high Tb^{3+} concentration used to dope the LSO crystal films (above 10 % [13]): the blue emission is optimum for 0.1 % Tb^{3+}, but is absent for concentrations exceeding 1%, due to the occurrence of the cross relaxation $(^5D_3 \rightarrow {}^7F_J) \rightarrow (^7F_6 \rightarrow {}^7F_0)$ process.

The emission spectrum of the codoped LSO:Tb^{3+}, Sm^{3+} is the overlap of the Tb^{3+} scintillation spectrum with the Sm^{3+} emission peaks between 560 nm - 620 nm. A point to be stressed is that such a peak could not be detected by using conventional laboratory X-ray sources whose total flux is around 10^6 ph/s. On the contrary, it is detectable at the higher synchrotron flux of up to 10^{16} ph/s.

Particularly interesting is the result concerning the substrate: its radio-luminescence spectrum is completely free from any parasitic component. Very often the substrates used to deposit the scintillating films emit undesired luminescence and such effect could deteriorate the spatial resolution performance reachable with the later scintillating film. The observed result confirms the suitability of the substrate choice also from the radio-luminescence point of view.

To investigate the effect induced by white beam X-ray irradiation the samples were irradiated with cumulative irradiations of 0, 0.5, 1, 1.8 and 3.2 hours. During each irradiation test a beam stopper was positioned between the scintillator and the entrance slit of the spectrograph to protect the spectrograph's optics. In addition, every irradiation was performed in the dark in order to avoid possible process induced by the light of the experimental hutch. The radiation damage tests were performed with a beam size equal to 8 x 8 mm^2 on the sample.

Results showed that the exposition to X-ray radiation induces an increase of the Tb^{3+} prompt luminescence in all the investigated samples. An example of such behaviour is

reported in Figure 3 which shows the radio-luminescence spectra of the LSO:Tb^{3+}, Sm^{3+} recorded after cumulative X-ray irradiations.
To better estimate the radiation induced increase of the LSO radio-luminescence we have calculated the integral luminescence for each spectrum and we have plotted it vs. irradiation time in Figure 4 for all the investigated samples. The LSO:Tb luminescence increases of about 6 % after a total irradiation time of 2 hours. For the LSO:Tb^{3+} we calculated an increase of the radio-luminescence integral of about 3.6 % after 3.2 s. In addition, the observed effect is saturated for the LSO:Tb^{3+}after 2 hours of irradiation. On the contrary for the Sm co-doped sample it is still increasing after 3.2 hours of irradiation.
Considering the absence of persistence components in the scintillation decay of the analysed samples a possible explanation to the observed behaviour could reside in some kind of competition effects between prompt recombination of free carriers and trapping at defects (traps). After prolonged irradiation, traps could become completely filled, so that they cannot compete anymore with prompt recombination. The consequence could be the observed increase of the radio-luminescence intensity under the irradiation with white beam X-rays.

Conclusions

Summarising our work, we have investigated the radiation induced effects on the radio-luminescence properties of a new scintillating materials based on the LPE growth of LSO:Tb^{3+} thin films on suitable substrate. A substrate and a codoped LSO: Tb^{3+},Sm^{3+} were considered as well. The recorded spectra are characterised by the main electronic transition lines of rare earth ions and the substrate is free from parasitic luminescence. Under the irradiation with white beam X-ray synchrotron radiation we observed and increase of the radio-luminescence intensity which is slower in case of the co-doped LSO: Tb^{3+},Sm^{3}. The most reliable explanation of the observed behaviour could be some kind of radiation induced increase of the Tb^{3+} emission efficiency primed by the filling process of the competing traps during the interaction of the X-rays with the scintillating material.

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Figure captions

Figure 1. Layout of the TopoTomo beamline at the ANKA light source [16].

Figure 2. Radio-luminescence spectra of LSO:Tb^{3+},Sm^{3+}, LSO:Tb^{3+} specimens and of the substrate.

Figure 3. Radio-luminescence spectra of LSO:Tb^{3+},Sm^{3+} after cumulative X-ray irradiations

Figure 4. LSO:Tb^{3+},Sm^{3+} and LSO:Tb^{3+} radio-luminescence integral vs. irradiation time.

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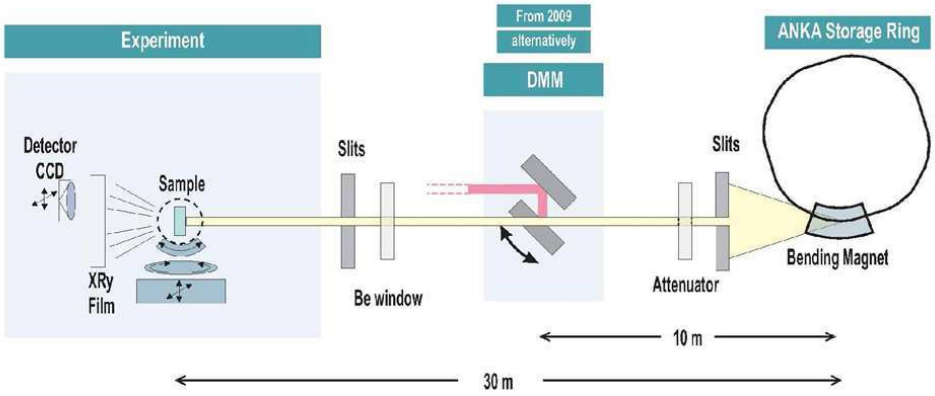

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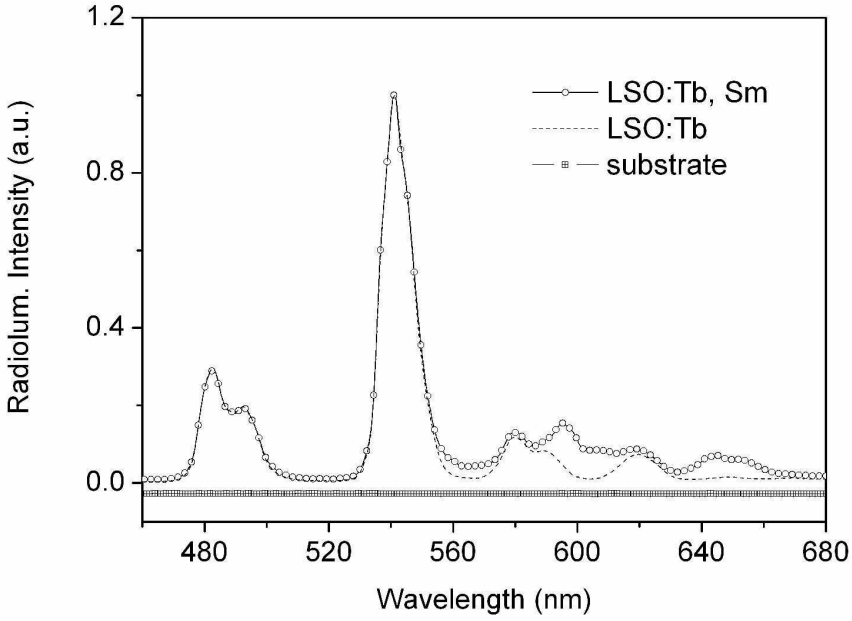

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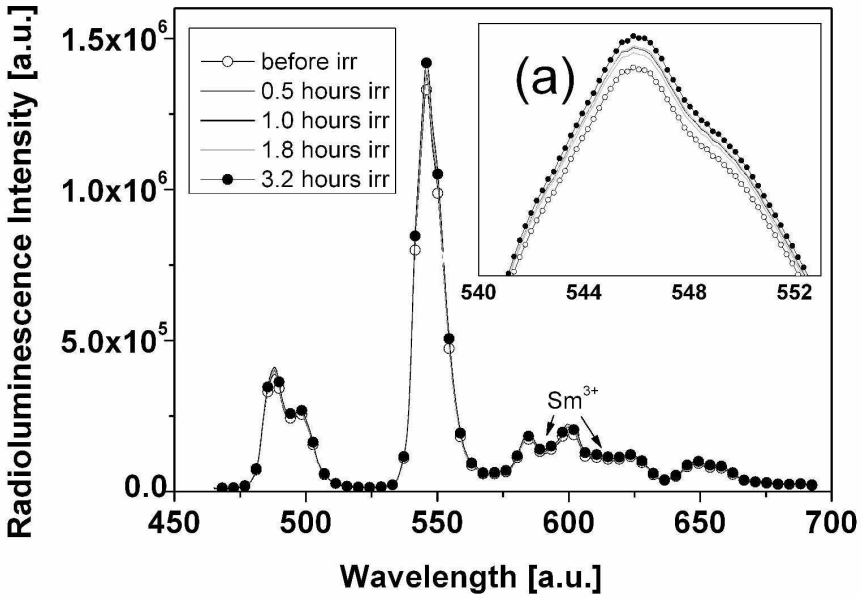

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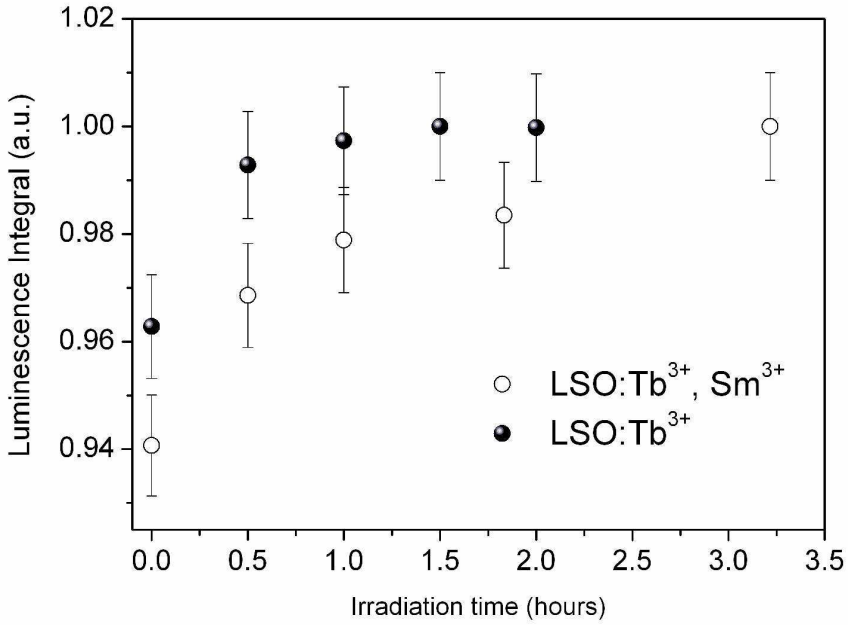

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