Experimental Report

Proposal title: XEOL and µLaue measurements of YAG:Ce phosphors
Proposal number: 20171182
Beamline: BM32
Shifts:12 (implementation of XEOL)
Date(s) of experiment: 2/3 Jul. 2018, 25/26 Jun 2018, 16/16 Jul 2018
Date of report: 15 feb.2020

- Objective & expected results (less than 10 lines): **-**

Combination of XEOL and µLaue has been demonstrated with several phosphors exhibiting different structural properties and emission efficiencies with a beam lower than 1 µm to provide a good spatial resolution. Similar counting times can be chosen for these two techniques and it allows performing simultaneous structural and optical mappings of samples. We realized only line-scans, but 2D mappings are possible, only the time requirement for a given area has to be checked.

- Results and the conclusions of the study (main part): **-**

To measure the XEOL signal in parallel to µLaue acquisition, we developed a new setup on the diffractometer to record the light emission in the framework of the Bottom-UP project of CEA called LumiX. The emitted photons are collected by an off-axis parabolic Al-mirror (X-rays are going through the mirror) that are then focalized by a second parabolic mirror to the entrance of an optic fiber going to a QEPRO Oceanview spectrometer. This light is then dispersed by a diffraction grating (HC1-GE) on a back-illuminated pixels CCD (SCMOS) camera cooled with a Peltier device. During the experiment, we have a shutter mechanism to select the acquisition time and to acquire the background. The entire system has been optimized to measure wavelength in the visible range. As an illustration of the technique, we measured four "phosphor" samples called A,B,C,D accordingly to Fig. 1. They will not be detailed in this report for confidentiality purpose (size, composition…) and the results presented here must be correlated to other characterization techniques. Grains are dispersed by hand on a glass standard microscope slide that is glued on a metallic holder

Results Sample A. We performed 20 µm line-scan with 51 points crossing an aggregation of crystals with 20 s counting time per points for both techniques. The beam size was 1.2 µm vertically and 700 nm horizontally.

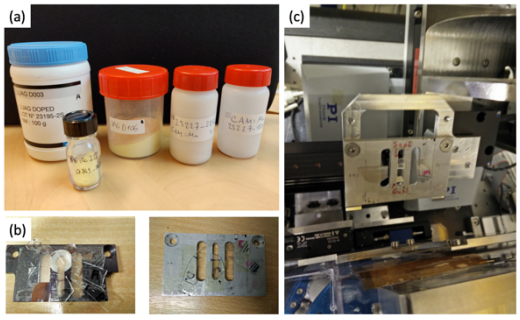


Fig. 1. (a) Sample descriptions, (b) metallic holders showing glass microscope slides with dispersed powers,
(c) the holder on the setup.

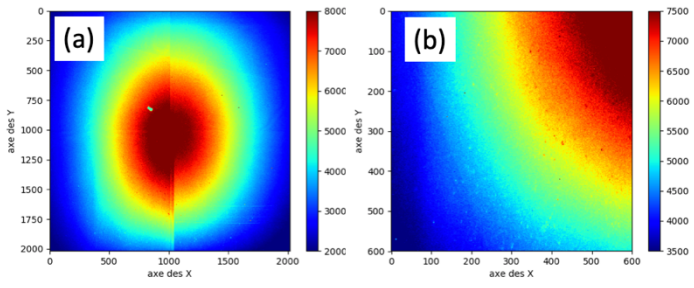
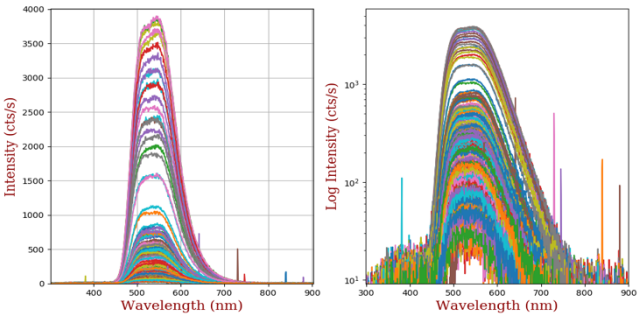


Fig. 2. (a) Raw 2D detector view: at this time (first experiment), the four SCMOS quadrants were not similar. This point is now corrected (b) zoom on the left-bottom quadrant showing individual Bragg peaks.

A representative µLaue pattern of the 4-quadrants SCMOS 2D detector is shown in Fig. 2 (a-b). The high intensity at the centre of the detector indicates a large diffusion mainly coming from the glass substrate, and to a less extent, from a quite large number of grains in the

sample. Indeed, all the intensities corresponding to the small spots observed in Fig. 2 (b) are summed and contribute to the formation of the image.



A direct improvement of this measurement can be obtained by optimizing the substrate on which grains are dispersed and by mastering the density of dispersed grains.

Fig. 3. XEOL of 51 spectra recorded during a 20 µm line-scan through a grain aggregate (counting time 20 s).

The XEOL signal is centred at about 540 nm. The intensity seems to be directly proportional to the volume of the grains that interacted with the beam (X-ray absorption mechanism). We can note that the general shape of the profile does not depend too much on the density. The sample seems to be quite homogeneous.

Results. Sample B. In a similar way, we performed for sample B 100 µm line-scan with 101 pts crossing aggregated crystals with 40 s counting time for both techniques. Beam size: 1.2 µm vertically and 540 nm horizontally. Fig. 4 shows a representative µLaue pattern. This measurement is very similar to what has been observed for sample A, counting rates are similar, and general conclusions are identical, stressing the necessity to isolated crystal under the beam for further investigations. In the right part of Fig. 4, we can also note that Bragg peaks have different shapes: some of them are sharp, some more diffuse and some are strongly elongated. It indicates different strain states and presence of defects in the individual grains.

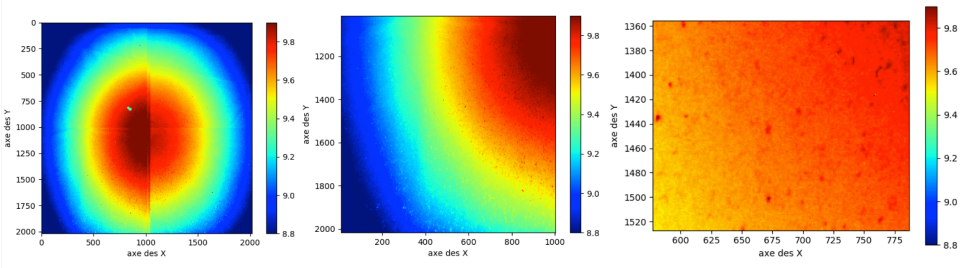


Fig. 4. (left) Raw 2D detector view of a diffraction pattern of sample B: (middle and right) zoom at different magnification.

As shown in Fig. 5, the XEOL signal is centred at about 565 nm (*i.e.* slightly larger than Sample A). It seems that the shape of the signal, in particular the broadening, is changing as a function of the intensity (*i.e.* number of grains or analysed volume). The peak is also less symmetric with a larger tail at longer wavelength. This point could be studied in more details.

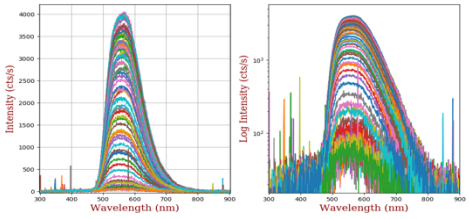


Fig. 5. XEOL of 101 spectra recorded during a 100 µm line-scan through a grain aggregate (counting time 40 s).

Results Sample C. We performed 20 µm line-scan with 51 pts crossing an aggregation of crystals with 20 s counting time for both techniques. Beam size: 1.2 µm vertically and 700 nm horizontally. We performed video (snapshot given in Fig. 6(demonstrating that scanning a small number of grains was possible. Large grains have clearly subgrains and distortions.The XEOL features for this sample is very different than for samples A and B. The emission is peaked at very sharp positions (to be indexed and compared to materials composition) that are superposed to broader contributions. The scanning shows also very clearly that the sample

homogeneity is not good because the relative intensities are changing as a function of the position.

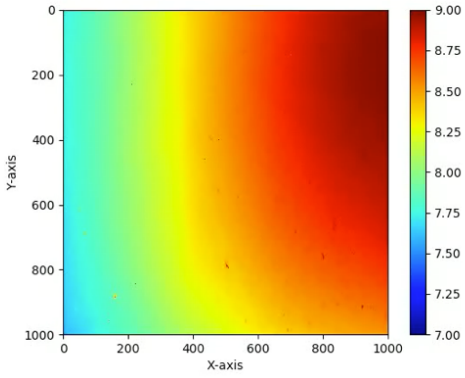


Fig. 6. Video of the bottom left detector sector. 20 µm line-scan, 51 points and 20 s counting time.

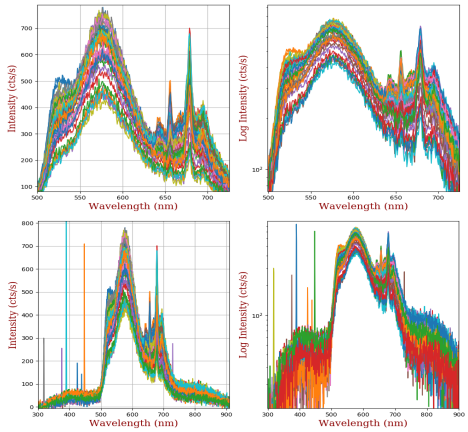


Fig. 7. XEOL of 51 spectra recorded during a 20 µm line-scan through a grain aggregate (counting time 20 s). Zooms in the lower part. Individual noise peaks came from radiation shielding, this problem has been solved.

Results Sample D. For these measurements, the beam size was significantly smaller (0.8 µm vertically and 400 nm horizontally) to demonstrate the possibility to isolate single grains.
This is illustrated in Fig. 8 obtained for 60 s counting. As for the other samples, we can see the signature of different crystallinity from the observation of peak shapes. The measured intensity was very low and for XEOL, it was mandatory to select big aggregated grains. Data correspond to 1 mm scanning, 51 points and 60 s counting.

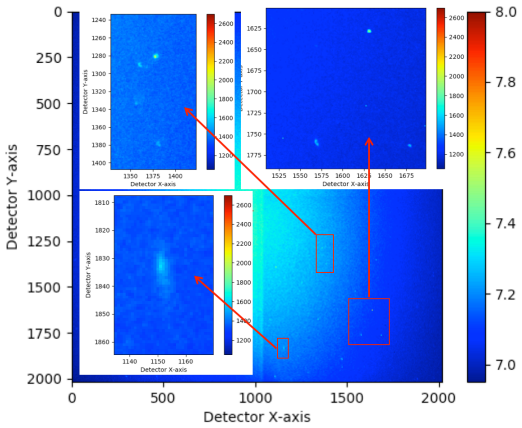


Fig. 8. Example of the µLaue pattern for sample D. 60 s counting.

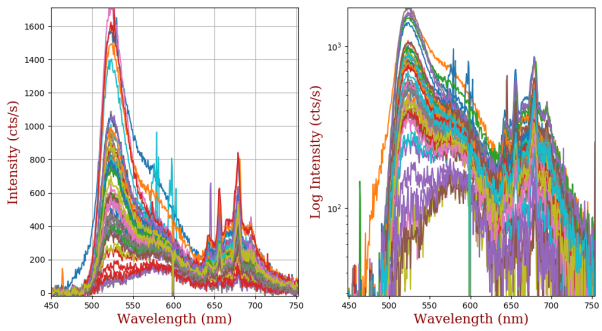


Fig. 9. XEOL of 51 spectra recorded during a 1 mm line-scan through a large aggregated grain (counting time 60 s).

The emission efficiency of sample D was lower than those of sample C, but similar features are observed with sharp emission peaks centered at characteristic wavelengths (noticeably red emission) and a lack of spatial homogeneity (see Fig. 9).

- Justification and comments about the use of beam time (5 lines max.): **-**

Very first experiments using this technique. Equipment has been designed, acquisition developed just before the EBS shutdown.
Evolution of the data file format with EBS. New work has to be developed to automatize the analysis procedure, especially with LaueTools analysis (writing notebooks). Very good XEOL results with respect to other ESRF beamline.

- Publication(s): **-**

No yet published.