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Photoluminescent colour centres in lithium fluoride film imaging detectors for monochromatic hard X-rays

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Summary. — Passive solid-state detectors based on the optical reading of the visible photoluminescence (PL) emitted by radiation-induced F_2 and F_3^+ colour centres in lithium fluoride (LiF) have been successfully tested for X-ray imaging. They are characterized by high spatial resolution, wide dynamic range, large field of view, non-destructive readout capability, and simplicity of use. Optically-transparent polycrystalline LiF films of increasing thickness (0.5 and 1.1 μm) grown by thermal evaporation on glass and Si(100) substrates were irradiated with monochromatic 7 keV X-rays at several doses from 13 to 1.4×10^3 Gy at the SOLEIL synchrotron facility. For all the LiF films, the spectrally-integrated visible PL signal intensity was found to depend linearly on the irradiation dose, with films grown on Si(100) substrates exhibiting up to a 50% higher response compared to those grown on glass. The minimum dose of 13 Gy was detected, despite the low thickness of the irradiated films. A spatial resolution of (0.54 ± 0.02) μm was obtained in edge-enhancement imaging experiments conducted by placing an Au mesh in front of the LiF film detectors.

1. – Introduction

X-ray diagnostic techniques such as diffraction and phase-contrast imaging, microradiography and X-ray microscopy are extensively used in many fields of application ranging from biomedicine to material science [1,2]. Despite significant achievements made in the past decades, the field of X-ray diagnostics is still looking for high-performance solid-state X-ray detectors characterized by high spatial resolution, wide dynamic range,

large field of view and non-destructive readout capability. In recent years, lithium fluoride (LiF) crystals and thin films have been successfully investigated as X-ray imaging detectors [3, 4] based on optical reading of visible photoluminescence (PL) emitted by radiation-induced F_2 and F_3^+ colour centres (CCs), which are stable at room temperature [5]. These aggregate CCs, consisting of two electrons bound to two and three close anionic vacancies, respectively, possess almost overlapped absorption bands peaked at about 450 nm (blue spectral region). Under blue light excitation, they emit broad PL bands peaked at 678 and 541 nm (red and green spectral regions), respectively, which can be read in non-destructive way by using, for instance, fluorescence microscopy. The optical reading experimental conditions have to be chosen in order to avoid photobleaching effect of radiation-induced F_2 and F_3^+ CCs. Passive solid-state radiation detectors based on LiF are characterized by high intrinsic spatial resolution over a large field of view, wide dynamic range, and simplicity of use. They are insensitive to ambient light and do not need chemical development after irradiation. The non-destructive readout capability allows for multiple measurements of the F_2 and F_3^+ PL without signal loss. Moreover, LiF detectors are reusable after proper thermal annealing treatments. In this work we present and discuss experimental results concerning radiation imaging detectors based on LiF thin films grown on glass and Si(100) substrates irradiated at several doses with monochromatic 7 keV X-rays.

2. – Materials and methods

Optically-transparent polycrystalline LiF thin films, of circular shape with a diameter of 10 mm and nominal thickness 0.5 and 1.1 μm, were grown by thermal evaporation on glass and Si(100) substrates at ENEA C.R. Frascati [10]. They were irradiated at several doses in the range from 13 to 1.4×10^3 Gy with monochromatic 7 keV X-rays at the METROLOGIE beamline of the SOLEIL synchrotron facility (Paris, France). To perform the irradiations, the LiF detectors were mounted on a xy-motorized sample holder, and the X-ray beam transverse area was reduced to a square of size $(2 \times 2)\,\mathrm{mm}^2$ by means of two mutually perpendicular shutters. Each LiF detector was irradiated in five distinct zones of its surface (spots), each with a different irradiation dose. To estimate the detectors' spatial resolution, edge-enhancement X-ray imaging experiments were conducted by placing an Au mesh (400 lines per inch, wire thickness of 12 μm) between the 7 keV X-ray source and the detectors at a fixed distance of 15 mm from them. The irradiations were performed with a dose of about 4×10^3 Gy. The visible PL emitted by the CCs formed in the irradiated areas was carefully investigated by using a Nikon Eclipse 80i optical microscope operating in fluorescence mode. The microscope is equipped with a 100 W mercury lamp optically filtered in the blue spectral region (420–460 nm), and an s-CMOS camera (Andor Neo, 16 bit, cooled at $-30\,^\circ$C) as 2D imaging detector, acquiring the F_2 and F_3^+ photoluminescence at wavelenghts higher then 515 nm by using an emission band pass optical filter. The microscope software (Nikon NIS-Elements) was used to process the acquired fluorescence images and estimate the corresponding spectrally-integrated PL intensities.

3. – Results and discussion

Figure 1 shows the PL response, *i.e.*, the spectrally-integrated F_2 and F_3^+ PL intensity as a function of irradiation dose, of LiF film detectors grown on glass (a) and Si(100) (b) substrates together with its linear best fit for each film thickness.

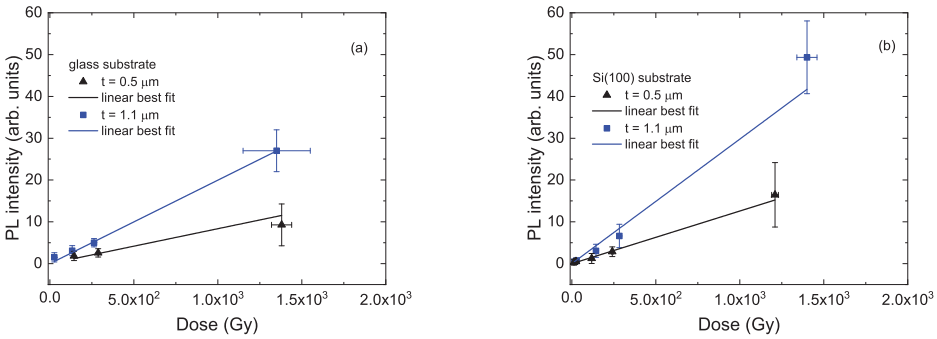

Fig. 1. – PL response of LiF film detectors grown on glass (a) and Si(100) substrates irradiated with 7 keV X-rays, together with its linear best fit for each film thickness t.

In both graphs, the same scale and range for the x and y axes was used to facilitate the comparison of the PL responses. Both for LiF film detectors grown on glass and Si(100) substrates, the PL response linearly depends on the irradiation dose in the investigated dose range, for each film thickness. The X-ray attenuation length at the energy of 7 keV is estimated to be 220 μm, which is much higher than the LiF film thickness of the investigated samples. This means that the coloration of the LiF films grown on glass and Si(100) substrates can be assumed to be homogeneous. For this reason, at the same irradiation dose, the observed difference in the PL intensities of LiF films grown on the same substrate is essentially ascribed to the different film thickness. As expected, the PL signal increases with the increasing the film thickness. For both substrates, moving from the thinnest 0.5 μm thick film to the one of thickness 1.1 μm, the PL signal is more than doubled within the experimental errors. At the same irradiation dose, the PL response of the LiF film detectors grown on Si(100) is higher than that of those deposited on glass in the same deposition run (*i.e.*, with same film thickness). The ratio between the slopes of the best-fit straight lines in fig. 1(b) to those in (a) is about 1.5 for both thicknesses. This value corresponds to an enhancement of about 50% of the PL signal intensity, mainly ascribable to the reflectivity of the silicon substrate in the visible spectral range, where the emission bands of the F_2 and F_3^+ CCs are located [6]. The lowest detected dose, delivered to the thinnest (t = 0.5 μm) LiF film detector grown on Si(100), was about 13 Gy, a value comparable to the minimum of 10 Gy reported in [7]. As far as the performances of the LiF film detectors in terms of field of view and spatial resolution are concerned, fig. 2 reports the fluorescence image of the Au mesh stored in the 1.1 μm thick LiF film grown on Si(100) acquired at magnification 20× (a) and 100× (b). Figure 2(c) reports the PL intensity profile measured along the white line drawn in fig. 2(b), while fig. 2(d) shows the PL intensity profile of a right portion (from 62 to 70 μm) of the second fluorescent spot in fig. 2(c), together with the Gaussian best fit (dashed line) of the highest peak of the diffraction pattern. The Half Width at Half Maximum of the Gaussian function, equal to $(0.54 \pm 0.02)\,\mu$m, can be considered as an evaluation of the spatial resolution of these LiF film detectors. This value, comparable to the diffraction-limited resolution of the microscope, confirms the submicrometric spatial resolution of the LiF film detectors.

4. – Conclusions

Passive solid-state X-ray detectors based on optically transparent polycrystalline LiF thin films of different thicknesses were deposited by thermal evaporation on glass and

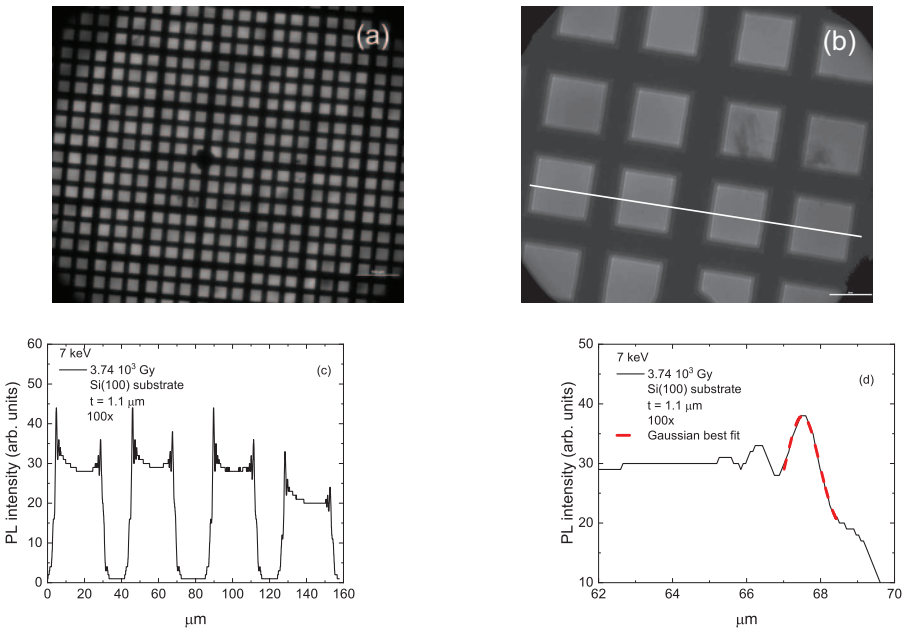

Fig. 2. – Fluorescence images of the Au mesh stored in the 1.1 μm thick LiF film grown on Si(100) irradiated with 7 keV X-rays at a dose of 3.74×10^3 Gy, acquired using an objective magnification of 20× (a) (image field size $(0.83 \times 0.70)\,\mathrm{mm}^2$) and 100×; (b) (image field size $0.17 \times 0.14\,\mathrm{mm}^2$); (c) PL intensity profile measured along the white line in fig. 2(b); (d) PL intensity profile of a right portion (from 62 to 70 μm) of the second fluorescent spot in fig. 2(c) together with the Gaussian best fit (dashed line) of the highest peak of the diffraction pattern.

Si(100) substrates. After irradiation with 7 keV X-rays at different doses in the range from 13 to 1.4×10^3 Gy, their visible PL response, investigated by fluorescence microscopy, has been found to follow a linear behaviour as a function of the irradiation dose. The lowest detected dose was of 13 Gy, despite of their limited thickness. This result suggests the usability of LiF film detectors even at clinical doses. The assessed high spatial resolution of $(0.54 \pm 0.02)\,\mu$m, combined with a wide field of view, renders LiF films promising for use as imaging detectors in X-ray diagnostic techniques.

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