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Optical characterization of lithium fluoride thin-film imaging detectors for monochromatic hard X-rays

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Abstract: Lithium fluoride (LiF) crystals and thin films have been successfully investigated as X-ray imaging detectors based on optical reading of visible photoluminescence emitted by stable radiation-induced F_2 and F_3^+ colour centres. In this work, the visible photoluminescence response of optically-transparent LiF film detectors of three different thicknesses, grown by thermal evaporation on Si(100) substrates and irradiated with monochromatic 7 keV X-rays at several doses in the range between 13 and 4.5×10^3 Gy, was carefully investigated by fluorescence optical microscopy. For all the film thicknesses, the photoluminescence response linearly depends on the irradiation dose in the investigated dose range. The lowest detected dose, delivered to the thinnest LiF film, only 0.5 μm thick, is estimated 13 Gy. Edge-enhancement imaging experiments, conducted by irradiating LiF film detectors at the same energy placing an Au mesh in front of them at a distance of 15 mm, allowed estimating a spatial resolution of (0.38 ± 0.05) μm, which is comparable to the microscope one. This very high spatial resolution in LiF film radiation detectors based on colour centres photoluminescence is combined with the availability of a wide field of view on large areas.

Keywords: Materials for solid-state detectors; Solid state detectors; X-ray detectors

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Contents

1 Introduction

High spatial resolution X-ray diagnostic techniques such as micro-radiography, X-ray microscopy, diffraction and phase-contrast imaging have important applications in various experimental fields ranging from biology to material science [1, 2]. A barrier to the uptake of these techniques is due to the limited characteristics of the employed detectors in terms of spatial resolution, dynamic range, field of view and non-destructive readout capabilities [3]. In the last decades, lithium fluoride (LiF) crystals and thin films have been successfully investigated as X-ray imaging detectors [4–8] based on optical reading of visible photoluminescence (PL) emitted by stable radiation-induced F_2 and F_3^+ colour centres (CCs) [9]. These aggregate CCs (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 optical pumping with blue light, they simultaneously 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 fluorescence microscopy. Passive solid-state radiation detectors based on PL reading of CCs in LiF are characterized by high intrinsic spatial resolution over a large field of view, wide dynamic range and simplicity of use as they are insensitive to ambient light. They can be considered “sustainable” as they do not need chemical development after irradiation and are reusable if subjected to proper thermal annealing processes. The non-destructive readout capability and the long term stability against fading of the latent images stored in irradiated LiF are other important features of these detectors.

In this work we present and discuss some experimental results on LiF thin films of three different thicknesses that were thermally evaporated on Si(100) substrates and irradiated by monochromatic X-rays of energy 7 keV at several doses in the range between 13 and 4.5×10^3 Gy (34 mJ/cm^3–12 J/cm^3).

2 Materials and methods

Radiation imaging detectors based on optically-transparent polycrystalline LiF thin films, of circular shape, with a diameter of 10 mm and nominal thickness 0.5, 1.1 and 1.8 µm, were grown on Si(100) substrates by thermal evaporation at ENEA C.R. Frascati [10]. They were irradiated at several doses with a monochromatic 7 keV X-ray beam at the METROLOGIE beamline of the SOLEIL synchrotron facility (Paris, France), in order to study their PL response. By means of two mutually perpendicular shutters, the X-ray beam area was reduced to a square of size $(2 \times 2)\,\mathrm{mm}^2$. Starting from the measurements of incident photon flux on the LiF films, performed before and after each irradiation by using a photodiode, the values of the irradiation doses were calculated and they resulted to be in a range between 13 and 4.5×10^3 Gy ($34\,\mathrm{mJ/cm}^3$–$12\,\mathrm{J/cm}^3$). Edge-enhancement X-ray imaging experiments, aimed to evaluate the spatial resolution of LiF film detectors, were conducted placing an Au mesh (400 lpi, thickness of 12 µm) in front of them, at a fixed distance of 15 mm. The X-ray beam energy and the irradiation dose were 7 keV and about 4×10^3 Gy, respectively. The PL emitted by irradiated areas under blue light illumination was carefully investigated by using a Nikon Eclipse 80i optical microscope operating in fluorescence mode, equipped with an excitation source consisting in a 100 W mercury lamp optically-filtered in the blue spectral range, which simultaneously excited the PL of F_2 and F_3^+ CCs, and an s-CMOS camera (Andor Neo, 16 bit, cooled at −30°C) as 2D imaging detector. For each irradiated spot, a spectrally integrated PL intensity profile was obtained by acquiring the fluorescence image with the microscope software. Then, the net PL signal was obtained by subtracting the minimum PL intensity (background noise) from the maximum intensity.

3 Results and discussion

Figure 1(a) shows the PL response of LiF film detectors grown on Si(100) substrate together with its linear best fit for each film thickness. In the inset of figure 1(a) the fluorescence image of the thickest LiF film (1.8 µm) irradiated with 7 keV X-rays at five doses: 4.5×10^3, 1.45×10^3, 652, 157 and 83 Gy (12, 3.8, 1.7, 0.4 and $0.2\,\mathrm{J/cm}^3$) — acquired by a conventional fluorescence microscope Nikon Eclipse 80-i (objective magnification 2×, numerical aperture (N.A.) = 0.10, binning 2×2, exposure time 2 s) is reported as an example. It clearly shows three of the five areas (spots) on the LiF film detector irradiated at the highest doses, while the dashed white rectangles highlight the lower-dose irradiated spots, which are not visible with the naked eye. For each film thickness, the PL response of LiF film detectors linearly depends on the irradiation dose in the investigated dose range. It can be observed that, at the same irradiation dose, the PL intensity increases with the film thickness. This can be attributed to the corresponding increase in the volume of irradiated LiF, which in turn leads to a higher number of radiation-induced CCs that exhibit PL when optically excited. Even the lowest dose, estimated 13 Gy ($34\,\mathrm{mJ/cm}^3$), delivered to the thinnest LiF film (0.5 µm) was detected and it is comparable with that obtained in [11] for a 1 µm thick film, despite the smaller thickness of the radiation-sensitive material layer.

Figure 1(b) shows the PL intensity profile measured along the luminous spots highlighted by the white line shown in the inset, which reports the fluorescence image of the Au mesh stored in the 1.8 µm thick LiF film irradiated with a dose of 3.75×10^3 Gy (objective magnification = 20×, N.A. = 0.75). Starting from the PL intensity profile, an Au mesh pitch of about 41 µm was estimated. In order to evaluate the spatial resolution of the LiF detectors, the fluorescence image of the same sample was also acquired by using an objective magnification of 100× (N.A. = 0.90) (see the inset of figure 1(c)). The PL intensity profile measured along the luminous spots, highlighted with a white line in the inset, is reported in figure 1(c). Figure 1(d) shows the PL intensity profile within the region marked with a dashed rectangle in figure 1(c), together with the Gaussian best fit (dashed line) of the highest peak of the diffraction pattern. From the best-fit procedure, a Half Width at Half

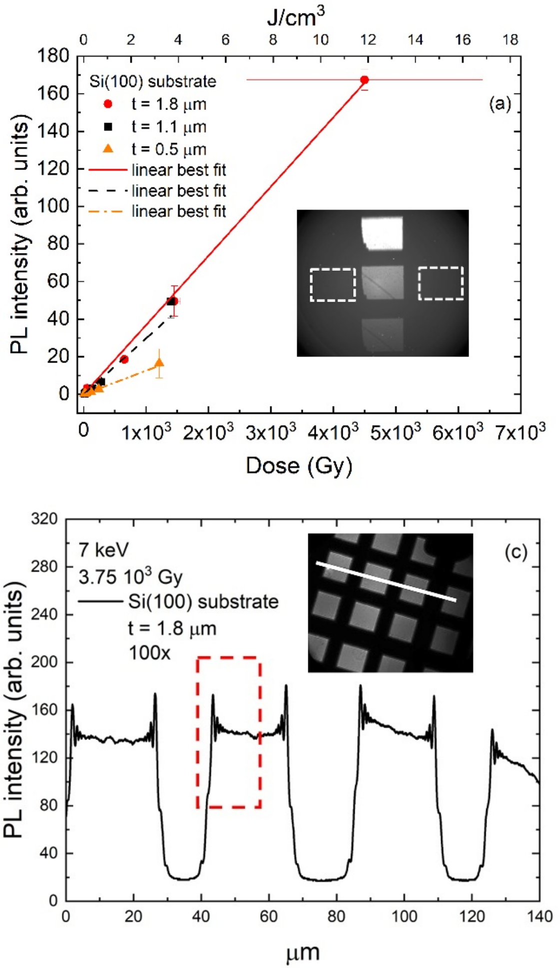

Figure 1. (a) PL response vs. dose of LiF film detectors, irradiated with 7 keV X-rays, together with its linear best fit for each film thickness (0.5, 1.1 and 1.8 μm). Inset of figure 1(a): fluorescence image of the thickest LiF film at five doses, image field size (1.67×1.41) cm^2. (b) PL intensity profile measured along the luminous spots highlighted with a white line in the inset, which reports the fluorescence image of the Au mesh as stored in the 1.8 μm thick LiF film irradiated with a dose of 3.75×10^3 Gy, acquired by the optical microscope with a 20× objective, image field size (0.83×0.70) mm^2. (c) PL intensity profile measured along the luminous spots highlighted with a white line in the inset, which reports the fluorescence image of the same sample acquired with a 100× objective, image field size (0.17×0.14) mm^2. (d) PL intensity profile within the region marked with a dashed rectangle in figure 1(c), together with the Gaussian best fit (dashed line) of the highest peak of the diffraction pattern.

Maximum of the Gaussian function of (0.38 ± 0.05) μm was obtained. This value can be considered an evaluation of the spatial resolution of the LiF detector, which is comparable to the microscope resolution — this latter is about 367 and 460 nm at the emission wavelengths of F_3^+ and F_2 CCs, respectively. These imaging tests demonstrate that LiF film detectors grown on Si(100) offer large field of view in combination with high spatial resolution.

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4 Conclusions

Passive X-ray imaging detectors based on polycrystalline LiF thin films of three different thicknesses were deposited by thermal evaporation on Si(100) substrates. After irradiation with 7 keV X-rays at different doses in the range between 13 and 4.5×10^3 Gy, their visible PL response, carefully investigated by fluorescence microscopy, has been found to follow a linear behaviour as a function of the irradiation dose. The lowest dose of 13 Gy was successfully detected, a fact that encourages the use of LiF film detectors even at clinical doses. The evaluated high spatial resolution of (0.38 ± 0.05) µm, together with a large field of view, make LiF films promising as imaging detectors in X-ray diagnostic techniques.

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