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1 Optical characterization of lithium fluoride thin-film 2 imaging detectors for monochromatic hard X-rays

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18 **ABSTRACT:** Lithium fluoride (LiF) crystals and thin films have been successfully investigated as
19 X-ray imaging detectors based on optical reading of visible photoluminescence emitted by stable
20 radiation-induced F₂ and F₃⁺ colour centres. In this work, the visible photoluminescence response
21 of optically-transparent LiF film detectors of three different thicknesses, grown by thermal evap-
22 oration on Si(100) substrates and irradiated with monochromatic 7 keV X-rays at several doses
23 in the range between 13 and 4.5×10³ Gy, was carefully investigated by fluorescence optical mi-
24 croscopy. For all the film thicknesses, the photoluminescence response linearly depends on the
25 irradiation dose in the investigated dose range. The lowest detected dose, delivered to the thinnest
26 LiF film, only 0.5 μm thick, is estimated 13 Gy. Edge-enhancement imaging experiments, con-
27 ducted by irradiating LiF film detectors at the same energy placing an Au mesh in front of them
28 at a distance of 15 mm, allowed estimating a spatial resolution of (0.38 ± 0.05) μm, which is
29 comparable to the microscope one. This very high spatial resolution in LiF film radiation detectors
30 based on colour centres photoluminescence is combined with the availability of a wide field of
31 view on large areas.

32 **KEYWORDS:** lithium fluoride; colour centres; thin films; hard X-rays; imaging detectors.

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42 **1. Introduction**

43 High spatial resolution X-ray diagnostic techniques such as micro-radiography, X-ray micros-
44 copy, diffraction and phase-contrast imaging have important applications in various experimental
45 fields ranging from biology to material science [1, 2]. A barrier to the uptake of these techniques
46 is due to the limited characteristics of the employed detectors in terms of spatial resolution, dy-
47 namic range, field of view and non-destructive readout capabilities [3]. In the last decades, lithium
48 fluoride (LiF) crystals and thin films have been successfully investigated as X-ray imaging detec-
49 tors [4-8] based on optical reading of visible photoluminescence (PL) emitted by stable radiation-
50 induced F_2 and F_3^+ colour centres (CCs) [9]. These aggregate CCs (two electrons bound to two
51 and three close anionic vacancies, respectively) possess almost overlapped absorption bands
52 peaked at about 450 nm (blue spectral region); under optical pumping with blue light, they sim-
53 ultaneously emit broad PL bands peaked at 678 and 541 nm (red and green spectral regions),
54 respectively, which can be read in non-destructive way by using fluorescence microscopy. Passive
55 solid-state radiation detectors based on PL reading of CCs in LiF are characterized by high intrin-
56 sic spatial resolution over a large field of view, wide dynamic range and simplicity of use as they
57 are insensitive to ambient light. They can be considered “sustainable” as they do not need chem-
58 ical development after irradiation and are reusable if subjected to proper thermal annealing pro-
59 cesses. The non-destructive readout capability and the long term stability against fading of the
60 latent images stored in irradiated LiF are other important features of these detectors.

61 In this work we present and discuss some experimental results on LiF thin films of three different
62 thicknesses that were thermally evaporated on Si(100) substrates and irradiated by
63 monochromatic X-rays of energy 7 keV at several doses in the range between 13 and 4.5×10^3 Gy
64 ($34 \text{ mJ/cm}^3 - 12 \text{ J/cm}^3$).

66 **2. Materials and methods**

67 Radiation imaging detectors based on optically-transparent polycrystalline LiF thin films, of
68 circular shape, with a diameter of 10 mm and nominal thickness 0.5, 1.1 and 1.8 μm , were grown
69 on Si(100) substrates by thermal evaporation at ENEA C.R. Frascati [10]. They were irradiated
70 at several doses with a monochromatic 7 keV X-ray beam at the METROLOGIE beamline of the
71 SOLEIL synchrotron facility (Paris, France), in order to study their PL response. By means of
72 two mutually perpendicular shutters, the X-ray beam area was reduced to a square of size (2×2)
73 mm^2 . Starting from the measurements of incident photon flux on the LiF films, performed before

74 and after each irradiation by using a photodiode, the values of the irradiation doses were
75 calculated and they resulted to be in a range between 13 and 4.5×10^3 Gy ($34 \text{ mJ/cm}^3 - 12 \text{ J/cm}^3$).
76 Edge-enhancement X-ray imaging experiments, aimed to evaluate the spatial resolution of LiF
77 film detectors, were conducted placing an Au mesh (400 lpi, wire thickness of $12 \mu\text{m}$) in front of
78 them, at a fixed distance of 15 mm. The X-ray beam energy and the irradiation dose were 7 keV
79 and about 4×10^3 Gy, respectively. The PL emitted by irradiated areas under blue light illumination
80 was carefully investigated by using a Nikon Eclipse 80i optical microscope operating in
81 fluorescence mode, equipped with an excitation source consisting in a 100 W mercury lamp
82 optically-filtered in the blue spectral range, which simultaneously excited the PL of F_2 and F_3^+
83 CCs, and an s-CMOS camera (Andor Neo, 16 bit, cooled at -30°C) as 2D imaging detector. For
84 each irradiated spot, a spectrally integrated PL intensity profile was obtained by acquiring the
85 fluorescence image with the microscope software. Then, the net PL signal was obtained by
86 subtracting the minimum PL intensity (background noise) from the maximum intensity.
87

88 3. Results and discussion

89 Figure 1 (a) shows the PL response of LiF film detectors grown on Si(100) substrate together with
90 its linear best fit for each film thickness. In the inset of Fig.1 (a) the fluorescence image of the
91 thickest LiF film ($1.8 \mu\text{m}$) irradiated with 7 keV X-rays at five doses: 4.5×10^3 , 1.45×10^3 , 652,
92 157 and 83 Gy (12 , 3.8 , 1.7 , 0.4 and 0.2 J/cm^3) – acquired by a conventional fluorescence
93 microscope Nikon Eclipse 80-i (objective magnification $2\times$, numerical aperture (N.A.) = 0.10,
94 binning 2×2 , exposure time 2s) is reported as an example. It clearly shows three of the five areas
95 (spots) on the LiF film detector irradiated at the highest doses, while the dashed white rectangles
96 highlight the lower-dose irradiated spots, which are not visible with the naked eye. For each film
97 thickness, the PL response of LiF film detectors linearly depends on the irradiation dose in the
98 investigated dose range. It can be observed that, at the same irradiation dose, the PL intensity
99 increases with the film thickness. This can be attributed to the corresponding increase in the
100 volume of irradiated LiF, which in turn leads to a higher number of radiation-induced CCs that
101 exhibit PL when optically excited. Even the lowest dose, estimated 13 Gy (34 mJ/cm^3), delivered
102 to the thinnest LiF film ($0.5 \mu\text{m}$) was detected and it is comparable with that obtained in [11] for
103 a $1 \mu\text{m}$ thick film, despite the smaller thickness of the radiation-sensitive material layer.
104 Figure 1 (b) shows the PL intensity profile measured along the luminous spots highlighted by the
105 white line shown in the inset, which reports the fluorescence image of the Au mesh stored in the
106 $1.8 \mu\text{m}$ thick LiF film irradiated with a dose of 3.75×10^3 Gy (objective magnification = $20\times$, N.A.
107 = 0.75). Starting from the PL intensity profile, an Au mesh pitch of about $41 \mu\text{m}$ was estimated.
108 In order to evaluate the spatial resolution of the LiF detectors, the fluorescence image of the same
109 sample was also acquired by using an objective magnification of $100\times$ (N.A. = 0.90) (see the inset
110 of Fig. 1 (c)). The PL intensity profile measured along the luminous spots, highlighted with a
111 white line in the inset, is reported in Fig. 1 (c). Figure 1 (d) shows the PL intensity profile within
112 the region marked with a dashed rectangle in Fig. 1 (c), together with the Gaussian best fit (dashed
113 line) of the highest peak of the diffraction pattern. From the best-fit procedure, a Half Width at
114 Half Maximum of the Gaussian function of $(0.38 \pm 0.05) \mu\text{m}$ was obtained. This value can be
115 considered an evaluation of the spatial resolution of the LiF detector, which is comparable to the
116 microscope resolution – this latter is about 367 and 460 nm at the emission wavelengths of F_3^+
117 and F_2 CCs, respectively. These imaging tests demonstrate that LiF film detectors grown on
118 Si(100) offer large field of view in combination with high spatial resolution.
119
120

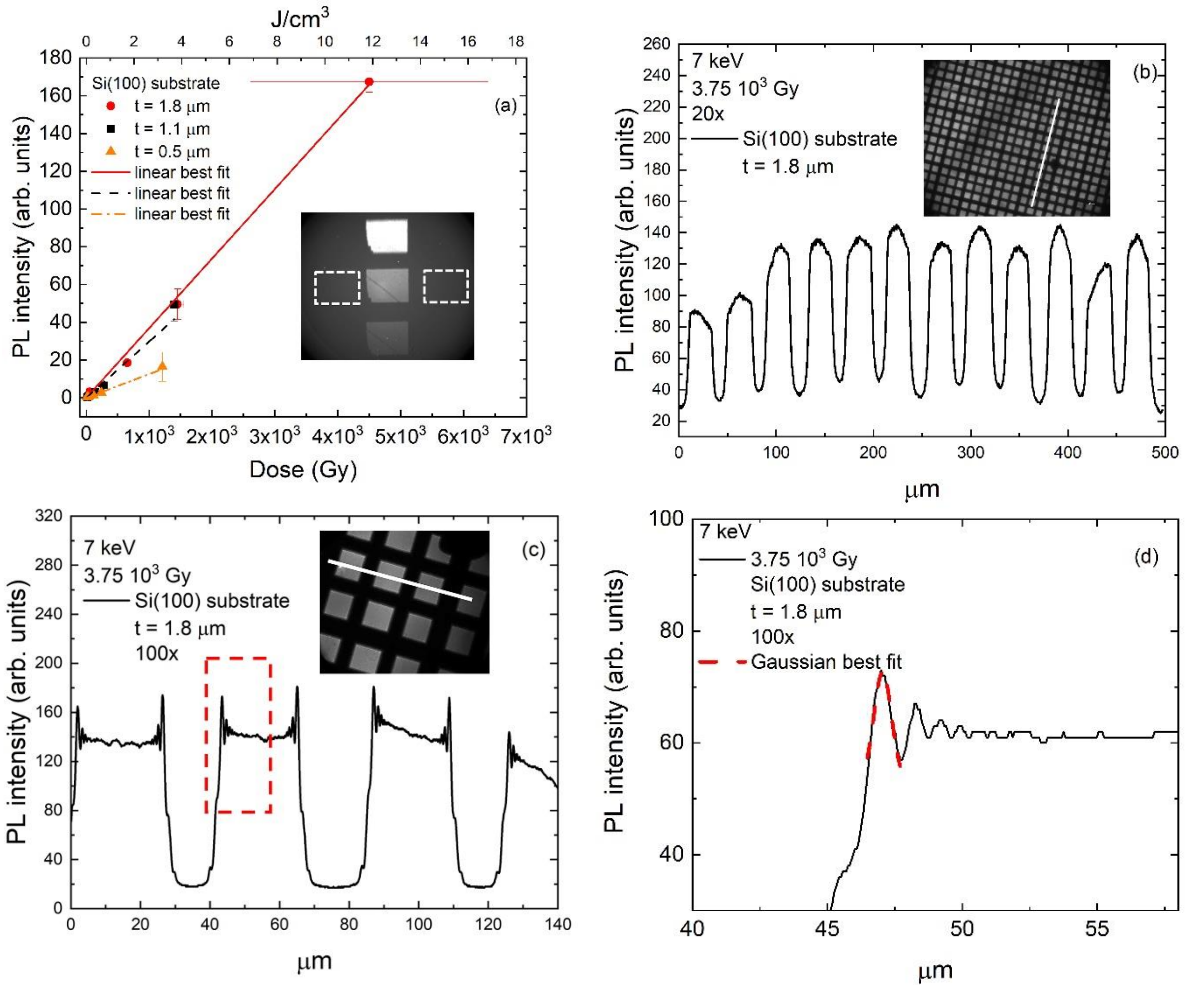


Figure 1. (a) PL response vs. dose of LiF film detectors, irradiated with 7 keV X-rays, together with its linear best fit for each film thickness (0.5, 1.1 and 1.8 μm). Inset of Fig.1 (a): fluorescence image of the thickest LiF film at five doses, image field size (1.67×1.41) cm². (b) PL intensity profile 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 3.75×10³ Gy, acquired by the optical microscope with a 20× objective, image field size (0.83×0.70) mm². (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) mm². (d) PL intensity profile within the region marked with a dashed rectangle in Fig. 1 (c), together with the Gaussian best fit (dashed line) of the highest peak of the diffraction pattern.

147

148 4. Conclusions

149 Passive X-ray imaging detectors based on polycrystalline LiF thin films of three different
 150 thicknesses were deposited by thermal evaporation on Si(100) substrates. After irradiation with 7
 151 keV X-rays at different doses in the range between 13 and 4.5×10³ Gy, their visible PL response,
 152 carefully investigated by fluorescence microscopy, has been found to follow a linear behaviour
 153 as a function of the irradiation dose. The lowest dose of 13 Gy was successfully detected, a fact
 154 that encourages the use of LiF film detectors even at clinical doses. The evaluated high spatial
 155 resolution of (0.38 ± 0.05) μm, together with a large field of view, make LiF film promising as
 156 imaging detectors in X-ray diagnostic techniques.

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162

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