

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

M.A. Vincenti, R.M. Montereali, E. Nichelatti, V. Nigro, M. Piccinini, M.

Koenig, P. Mabey, G. Rigon, H.J. Dabrowski, Y. Benkadoum, et al.

▶ To cite this version:

M.A. Vincenti, R.M. Montereali, E. Nichelatti, V. Nigro, M. Piccinini, et al.. Optical characterization of lithium fluoride thin-film imaging detectors for monochromatic hard X-rays. Journal of Instrumentation, 2023, 18 (04), pp.C04012. 10.1088/1748-0221/18/04/C04012. hal-04287326

HAL Id: hal-04287326 https://hal.science/hal-04287326

Submitted on 15 Nov 2023

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers. L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution 4.0 International License

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

3 4 5	M.A. Vincenti ^{a,1} , R.M. Montereali ^a , E. Nichelatti ^b , V. Nigro ^a , M. Piccinini ^a , M. Koenig ^c , P. Mabey ^d , G. Rigon ^c , H.J. Dabrowski ^c , Y. Benkadoum ^c , P. Mercere ^e , P. Da Silva ^e , T. Pikuz ^f , N. Ozaki ^g , S. Makarov ^h , S. Pikuz ^h , B. Albertazzi ^c
6	^a ENEA C.R. Frascati, Fusion and Technologies for Nuclear Safety and Security Dept., Via E. Fermi
7	45, 00044 Frascati (RM), Italy
8	^b ENEA C.R. Casaccia, Fusion and Technologies for Nuclear Safety and Security Dept., Via
9	Anguillarese 301, 00123 S. Maria di Galeria (RM), Italy
10	^c LULI-CNRS, Ecole Polytechnique, CEA, Universitè Paris-Saclay, F-91128 Palaiseau Cedex, France
11	^d Department of Physics, Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany
12	^e SOLEIL synchrotron, L'Orme des Merisiers, Départementale 128, 91190 Saint Aubin
13	^f Institute for Open and Transdisciplinary Research Initiatives, Osaka University, Suita, Osaka 565-
14	0871 Japan
15	^g Graduate School of Engineering, Osaka University, Suita, Osaka 565-0871 Japan
16	^h Joint Institute for High Temperature RAS, Moscov 125412, Russia
17	E-mail: aurora.vincenti@enea.it

ABSTRACT: Lithium fluoride (LiF) crystals and thin films have been successfully investigated as 18 19 X-ray imaging detectors based on optical reading of visible photoluminescence emitted by stable 20 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 evap-21 22 oration 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 mi-23 croscopy. For all the film thicknesses, the photoluminescence response linearly depends on the 24 irradiation dose in the investigated dose range. The lowest detected dose, delivered to the thinnest 25 LiF film, only 0.5 µm thick, is estimated 13 Gy. Edge-enhancement imaging experiments, con-26 27 ducted 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 $(0.38 \pm 0.05) \,\mu\text{m}$, which is 28 comparable to the microscope one. This very high spatial resolution in LiF film radiation detectors 29 based on colour centres photoluminescence is combined with the availability of a wide field of 30 31 view on large areas.

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

- 33
- 34 ¹ Corresponding author.

35 **Contents**

36	1. Introduction	1
37	2. Materials and methods	1
38	3. Results and discussion	2
39	4. Conclusions	3
40		

41

42 **1. Introduction**

High spatial resolution X-ray diagnostic techniques such as micro-radiography, X-ray micros-43 44 copy, 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 45 is due to the limited characteristics of the employed detectors in terms of spatial resolution, dy-46 47 namic 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 detec-48 tors [4-8] based on optical reading of visible photoluminescence (PL) emitted by stable radiation-49 induced F_2 and F_3^+ colour centres (CCs) [9]. These aggregate CCs (two electrons bound to two 50 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 simultaneously emit broad PL bands peaked at 678 and 541 nm (red and green spectral regions), 53 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 intrinsic spatial resolution over a large field of view, wide dynamic range and simplicity of use as they 56 are insensitive to ambient light. They can be considered "sustainable" as they do not need chem-57 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.

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/cm³ - 12 J/cm³).

65

66 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 μ m, 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 (2×2) mm². 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 calculated and they resulted to be in a range between 13 and 4.5×10^3 Gy (34 mJ/cm³ – 12 J/cm³). 75 Edge-enhancement X-ray imaging experiments, aimed to evaluate the spatial resolution of LiF 76 film detectors, were conducted placing an Au mesh (400 lpi, wire thickness of $12 \,\mu\text{m}$) in front of 77 them, at a fixed distance of 15 mm. The X-ray beam energy and the irradiation dose were 7 keV 78 and about 4×10^3 Gy, respectively. The PL emitted by irradiated areas under blue light illumination 79 was carefully investigated by using a Nikon Eclipse 80i optical microscope operating in 80 fluorescence mode, equipped with an excitation source consisting in a 100 W mercury lamp 81 optically-filtered in the blue spectral range, which simultaneously excited the PL of F_2 and F_3^+ 82 CCs, and an s-CMOS camera (Andor Neo, 16 bit, cooled at -30°C) as 2D imaging detector. For 83 each irradiated spot, a spectrally integrated PL intensity profile was obtained by acquiring the 84 85 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. 86

87

88 3. Results and discussion

Figure 1 (a) shows the PL response of LiF film detectors grown on Si(100) substrate together with 89 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 μ m) 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 J/cm³) – acquired by a conventional fluorescence 92 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 (spots) on the LiF film detector irradiated at the highest doses, while the dashed white rectangles 95 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 investigated dose range. It can be observed that, at the same irradiation dose, the PL intensity 98 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 exhibit PL when optically excited. Even the lowest dose, estimated 13 Gy (34 mJ/cm³), delivered 101 to the thinnest LiF film $(0.5 \,\mu\text{m})$ was detected and it is comparable with that obtained in [11] for 102 103 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 104 105 white line shown in the inset, which reports the fluorescence image of the Au mesh stored in the 1.8 μ m thick LiF film irradiated with a dose of 3.75×10^3 Gy (objective magnification = 20×, N.A. 106 107 = 0.75). Starting from the PL intensity profile, an Au mesh pitch of about 41 μ m was estimated. 108 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 $100 \times (N.A. = 0.90)$ (see the inset 109 of Fig. 1 (c)). The PL intensity profile measured along the luminous spots, highlighted with a 110 white line in the inset, is reported in Fig. 1 (c). Figure 1 (d) shows the PL intensity profile within 111 the region marked with a dashed rectangle in Fig. 1 (c), together with the Gaussian best fit (dashed 112 line) of the highest peak of the diffraction pattern. From the best-fit procedure, a Half Width at 113 114 Half 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 115 microscope resolution – this latter is about 367 and 460 nm at the emission wavelengths of F_3^+ 116 and F_2 CCs, respectively. These imaging tests demonstrate that LiF film detectors grown on 117 118 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 μ m). 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 μ m 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 thicknesses were deposited by thermal evaporation on Si(100) substrates. After irradiation with 7 150 keV X-rays at different doses in the range between 13 and 4.5×10^3 Gy, their visible PL response, 151 152 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 153 that encourages the use of LiF film detectors even at clinical doses. The evaluated high spatial 154 resolution of $(0.38 \pm 0.05) \,\mu$ m, together with a large field of view, make LiF film promising as 155 imaging detectors in X-ray diagnostic techniques. 156

157 Acknowledgments

Research carried out within the TecHea (Technologies for Health) project, funded by ENEA, Italy
and partly supported by KAKENHI (grant no. 21K03499) from the Japan Society for the
Promotion of Science (JSPS). JIHT RAS team was supported by MSHE RF (grant No. 075-152021-1352).

162

163 **References**

- 164 [1] M. Endrizzi, X-ray phase-contrast imaging, Nucl. Inst. Methods Phys. Res., A 878 (2018) 88-98.
- [2] S.C. Mayo, A.W. Stevenson and S.W. Wilkins, *In-line Phase-Contrast X-ray Imaging and Tomography for Material Science*, *Materials* 2012, 5, 937-965.
- [3] A. Faenov, T. Pikuz, P. Mabey, B. Albertazzi, T. Michel, G. Rigon, S. Pikuz, A. Buzmakov, S.
 Makarov, N. Ozaki et al., Advanced high resolution X-ray diagnostic for HEDP experiments, Sci.
 Rep. 8, 16407 (2018).
- [4] G. Baldacchini, F. Bonfigli, A. Faenov, F. Flora, R. M. Montereali, A. Pace, T. Pikuz, L. Reale, *Lithium Fluoride as a Novel X-Ray Image Detector for Biological μ-World Capture*, *J. Nanoscience and Nanotechnology 3,6 (2003) 483-486.*
- [5] F. Bonfigli, A. Faenov, F. Flora, M. Francucci, P. Gaudio, A. Lai, S. Martellucci, R.M. Montereali,
 T. Pikuz, L. Reale, M. Richetta, M.A. Vincenti, G. Baldacchini, *High-Resolution Water-Window X- Ray Imaging of In Vivo Cells and Their Products Using LiF Crystal Detectors*, *Microscopy Research and Techniques* 71 (2008) 35-41.
- [6] S.H. Bateni, F. Bonfigli, A. Cecilia, T. Baumbach, D. Pelliccia, F. Somma, M.A. Vincenti and R.M.
 Montereali, *Optical characterisation of lithium fluoride detectors for broadband X-ray imaging*, *Nucl. Inst. Methods Phys. Res. A 720 (2013) 109-112.*
- [7] P. Mabey, B. Albertazzi, Th. Michel, G. Rigon, S. Makarov, N. Ozaki, T. Matsuoka, S. Pikuz, T.
 Pikuz and M.Koenig. *Characterization of high spatial resolution lithium fluoride X-ray detectors*, *Rev. Sci. Instrum. 90, 063702 (2019).*
- [8] F. Flora, S. Almaviva, G. Baldacchini, D. Batani, S. Bollanti, F. Bonfigli, P. Di Lazzaro, M.
 Francucci, P. Gaudio, A. Lai, T. Letardi, L. Mezi, R.M. Montereali, D. Murra, E. Nichelatti, A. Poma,
 A. Scafati, L. Palladino, T. Pikuz, A. Reale, L. Reale, M. Richetta, A. Ritucci, A. Rydzy, M.A.
 Vincenti and P. Zuppella, X-ray imaging of bio/medical samples using laser-plasma-based X-ray
 sources and LiF detector, Journal of Instrumentation, JNST 14 (2019) C10006.
- [9] J. Nahum, Optical Properties and Mechanism of Formation of Some F-aggregate centers in LiF,
 Phys. Rev. 158 (1967) 814.
- 190[10]M.A. Vincenti, M. Leoncini, S. Libera, A. Ampollini, A. Mancini, E. Nichelatti, V. Nigro, L. Picardi,191M. Piccinini, C. Ronsivalle, A. Rufoloni, R.M. Montereali, Enhanced photoluminescence of F_2 and192 F_3^+ colour centres in lithium fluoride film-based detectors for proton beams, Opt Mat 119 (2021)193111376.
- [11] T. Kurobori and A. Matoba, Development of accurate two-dimensional dose-imaging detectors using atomic-scale color centers in Ag-activated phosphate glass and LiF thin films, Jpn J Appl Phys 53, 02BD14 (2014).