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Radiation-induced photoluminescent colour centres in lithium fluoride films for the detection of monochromatic hard X-rays

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Abstract: Optically-transparent LiF film radiation imaging 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, were investigated by fluorescence microscopy to evaluate their visible photoluminescence response of X-ray induced colour centres and their spatial resolution. By edge-enhancement imaging experiments, performed by irradiating LiF film detectors with an Au mesh in front of them, a spatial resolution of ~0.3 µm was estimated. This high spatial resolution, combined with the large field of view and the wide dynamic range offered by LiF detectors, encourages their use in X-ray imaging and for high-energy density experiments in synchrotron facilities.

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

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Contents

1 Introduction

Contact X-ray imaging techniques take great advantages from high spatial resolution of the selected radiation detectors, which should be also characterised by good sensitivity, high dynamic range and response linearity. Passive solid-state imaging detectors based on thermally evaporated lithium fluoride (LiF) thin films were initially proposed for soft X-ray contact microscopy [1] and then extended at higher X-ray energies using polychromatic sources, like X-ray tubes [2] and synchrotrons [3]. These detectors are based on optical reading of visible photoluminescence (PL) emitted by radiation-induced F_2 and F_3^+ colour centres (CCs) (two electrons bound to two and three close anionic vacancies, respectively), stable at room temperature. The F_2 and F_3^+ CCs have almost overlapped absorption bands peaked at $\sim$ 450 nm; under optical excitation in the blue spectral interval, they simultaneously emit broad PL bands peaked at 678 and 541 nm, respectively, which can be directly read in non-destructive way by fluorescence microscopy. The colouration depth is limited by the thickness of LiF films, which allows obtaining well-contrasted two-dimensional (2D) imaging even for high-energy X-rays [2, 3], whose attenuation lengths are much higher. Additionally, the limited film thickness allows reducing the contribution to the PL signal due to secondary processes, such as the diffusion of F-centres and the photoelectron cascade of electrons, that occurs in LiF crystal irradiated with hard X-rays [4]. However, a poorer detector sensitivity is expected due to the limited volume of the irradiated material. As reported in our previous works [5, 6], LiF film-based detectors are characterised by a PL response (the spectrally-integrated PL intensity due to both F_2 and F_3^+ CCs) linearly proportional to the irradiation dose for all the three film thicknesses, in the investigated dose range. This behaviour is particularly interesting in radiation imaging and dosimetry. In this paper we investigate the spatial resolution of LiF film detectors of increasing thickness, confirming a great improvement compared to that of LiF crystal detectors, which was found to lie in the range 0.8–1.3 µm in experiments with XFEL sources at photon energies of 7–10 keV and in high energy density (HED) experiments [4, 7–9].

2 Materials and methods

Optically-transparent polycrystalline LiF films (of nominal thickness 0.5, 1.1 and 1.8 µm) were grown by thermal evaporation on Si(100) substrates at ENEA C.R. Frascati [10]. LiF powder (Merck Suprapur, 99.99% pure), heated in a tantalum crucible at about 850 °C, was used as starting material. The deposition processes were performed at a pressure inside the vacuum chamber below 1 mPa prior to evaporation, while keeping the substrate temperature at 300 °C. The deposition rate, fixed at the

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nominal value of 1 nm/s, and the film thickness were monitored in situ by an INFICON quartz-crystal oscillator. They were irradiated at several doses from 13 to $4{\times}10^3$ Gy with monochromatic 7 keV X-ray beams at the METROLOGIE beamline of the SOLEIL synchrotron facility, to study their visible PL response. At this energy, the attenuation length of X-rays in LiF is about 220 μm, much higher than the thickness of the investigated samples, so that the colouration can be assumed as homogeneous along the film depth. To study spatial resolution, edge-enhancement X-ray imaging experiments were carried out placing an Au test grid (400 lpi, wire thickness 12 μm) in front of the LiF detectors, at a fixed distance of 15 mm. The irradiations were performed at a dose of about $4{\times}10^3$ Gy. The PL intensity emitted by the irradiated areas was 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 interval, as polychromatic illumination source used to simultaneously excite the visible 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.

3 Results and discussion

Figure 1(a) shows the images of the Au test grid recorded in the three LiF films of different thicknesses. In the enlarged views of the grid wire (figure 1(b)), the diffraction patterns with a few maxima are clearly seen. The plot in figure 1(c) shows that up to 4 diffraction fringes are well resolved with the distance between 3rd and 4th peak of ~ 0.8 μm. It is noticeable that the intensity profiles in the three LiF samples are similar.

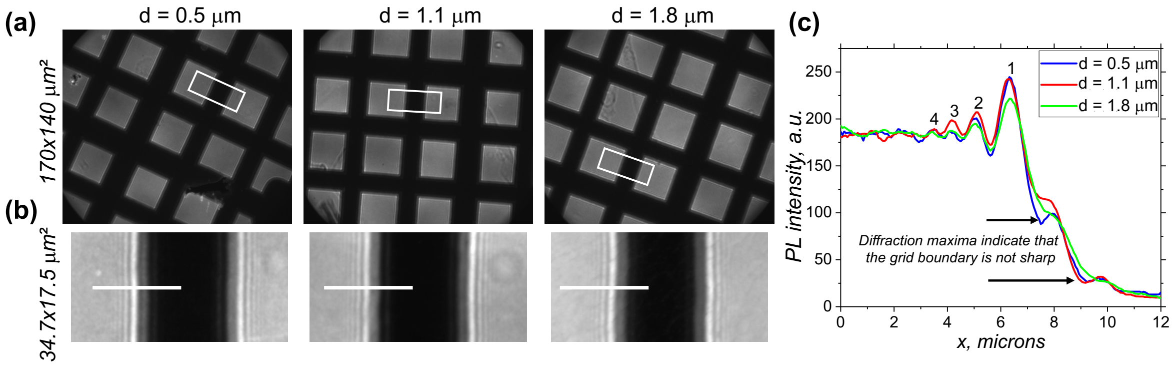

Figure 1. (a) Fluorescence images of the Au test grid recorded on LiF films with thickness d = 0.5, 1.1 and 1.8 μm (on Si(100) substrates) observed with 100× objective. (b) Enlarged images of the grid wire enclosed in the white rectangles. (c) Comparison of PL intensity profiles taken along the white lines in (b) for each film thickness.

To evaluate the achieved spatial resolution (due to the LiF detector and microscope objective) for the photon energy of 7 keV, the diffraction pattern of the 0.5 μm-thick LiF film (blue curve in figure 1(c)) was compared with a model. Simulations were performed using the open-source software package WavePropagator [11]. 2D synthetic diffraction patterns were calculated for different spatial resolutions in the range 0.05–0.4 μm and compared with the experimental ones (figure 2). The best agreement was obtained for a spatial resolution of 0.3 μm (panel (c)). The amplitudes of the diffraction peaks, their profiles and number are in good agreement with the experimental ones. The obtained value corresponds to the optical resolution of the read-out system. Therefore, this value is an upper

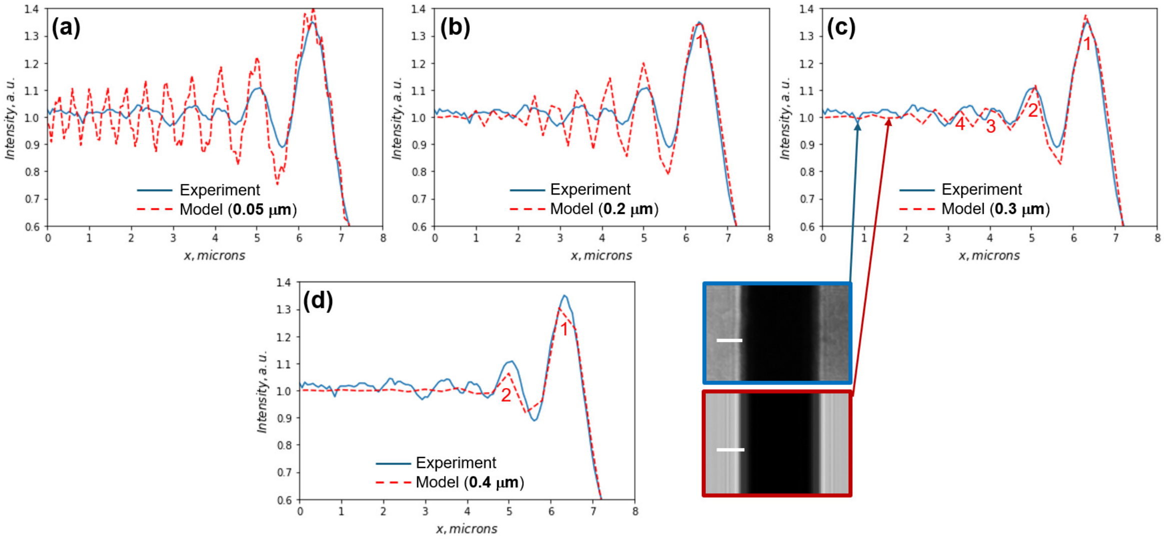

Figure 2. Comparison of the experimental PL intensity profile for the 0.5-µm thick LiF film (left PL image (b) and plot in blue (c) in figure 1) with the simulations for different sizes of model spatial resolution: (a) 0.05 µm, (b) 0.2 µm, (c) 0.3 µm and (d) 0.4 µm. Experimental and simulation images are also shown in the right-below couple of images.

estimate of the LiF detector resolution, whose actual resolving power could be even better [12]. As example, this was demonstrated in [13, 14], where image acquisition of soft X-ray microradiographies stored in LiF films, performed by a scanning near-field optical microscope (SNOM), allowed obtaining a spatial resolution in the range between 50 and 75 nm.

4 Conclusions

The visible PL response of radiation-induced CCs in optically-transparent polycrystalline LiF films of increasing thicknesses, grown by thermal evaporation on Si(100) substrates, irradiated with 7 keV X-rays at different doses, were investigated by using a fluorescence microscope. Their spatial resolution was carefully tested in edge-enhancement imaging experiments performed at the same X-ray energy. It was found that it does not depend on the film thickness in the investigated thickness range and read-out conditions. The obtained value (~ 0.3 µm), although much better than that found for LiF crystals (~ 0.8 µm, as reported in ref. [8], as example), is quite larger than the hypothetical intrinsic spatial resolution of these detectors, which should be in principle comparable with the CC atomic-scale size. Indeed, it is limited by the spatial resolution of the read-out system and by the size of the secondary electron cloud generated by X-ray radiation, which depends on the photon energy [4]. Nonetheless, this latter contribution is minimized by using LiF thin films, with which it is possible to achieve a better spatial resolution using, for example, a SNOM as readout system [13, 14]. These properties, in combination with a wide dynamic range and a large field of view, make LiF films promising as imaging detectors in X-ray diagnostic methods [13] and for conducting HED experiments using modern synchrotron sources [8, 9].

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