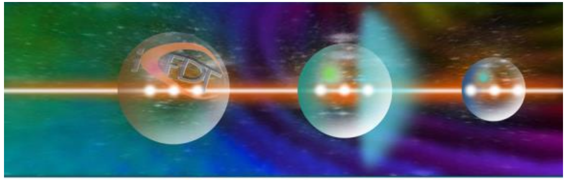
Italian National Agency for New Technologies, Energy and Sustainable Economic Development



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

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Outline

- *Introduction*

  Lithium fluoride: material properties and main colour centres;

  Thermally-evaporated LiF film-based detectors;

  LiF radiation imaging detectors: peculiarities.

- *LiF film-based detectors irradiated with 7 keV X-rays*

  Photoluminescence response of irradiated LiF films grown on glass and Si(100) substrates;

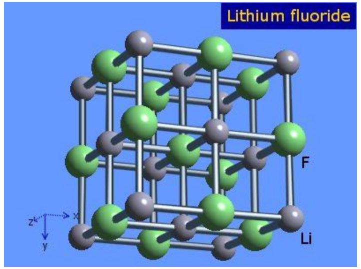
  Substrate-enhanced photoluminescence of radiation-induced colour centres;

  Edge-enhancement X-ray imaging experiments.

- *Conclusions and future perspectives*

Lithium fluoride

Properties and colour centres:



- fcc ionic crystal;
- hard;
- almost non-hygroscopic;
- optically transparent from 120 nm to 7μm (band gap ~ 14 eV);
- irradiation by ionising radiations (X rays, γ rays, neutrons, protons etc.) gives rise to stable formation at room temperature (RT) of primary and aggregate colour centres (CCs) characterized by wide tunability and high emission quantum efficiency, even at RT;
- LiF is a nearly tissue-equivalent material (Z_{eff} = 8.1, $Z_{eff\ water}$ = 7.5)

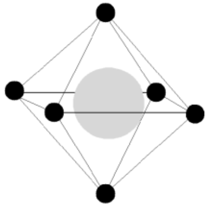
Main applications:

- solid state tuneable lasers (Vis, NIR);
- miniaturized light sources;
- radiation detectors;
- dosemeters.

Nearest neighbour distance (Å)	2.013
Melting point (°C)	848.2
Density (g/cm^3 a RT)	2.639
Molecular weight	25.939
Refractive index at 640 nm, RT	1.3912
Solubility (g/100 g H_2O a RT)	0.134
Hardness (Knoop)	102

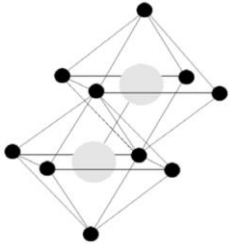
Main physical LiF parameters

Main colour centres in LiF



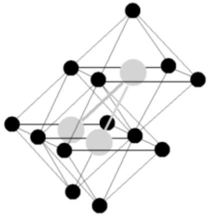
F

F centre is an anion vacancy occupied by an electron.



F_2

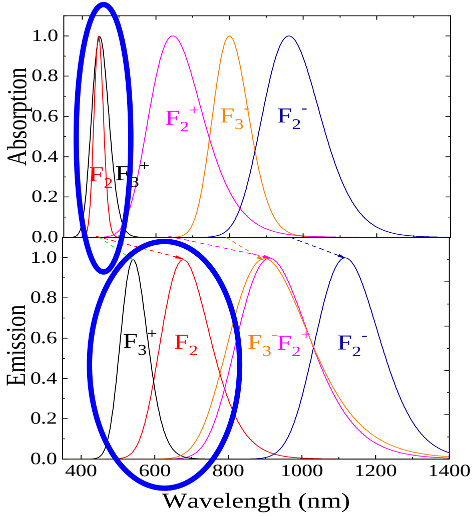
F_2 electronic defect consists of two nearest-neighbour F centres along a <100> direction of the cubic lattice.



F_3

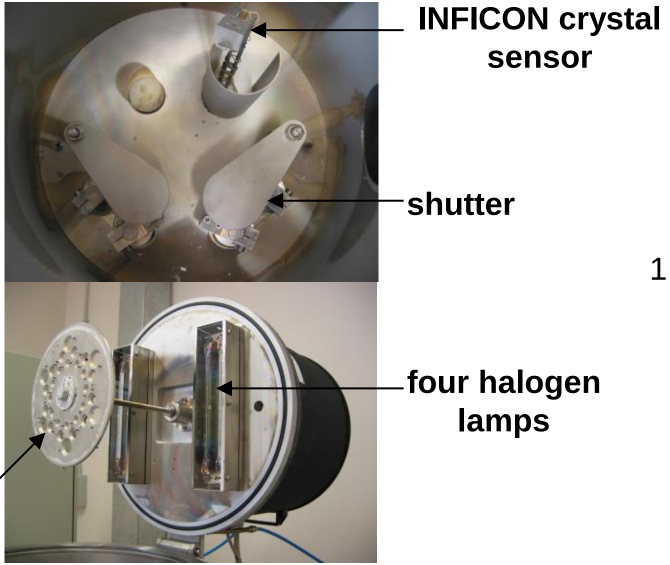
F_3 centre consists of three F centres in nearest-neighbour sites in the (111) plane.

Center	E_a (eV, nm)	E_e (eV, nm)	$FWHW_a$ (eV)	$FWHW_e$ (eV)
F	5.00, 248	-	0.76	
F_2	2.79, 444	1.83, 678	0.16	0.36
F_3^+	2.77, 448	2.29, 541	0.29	0.31

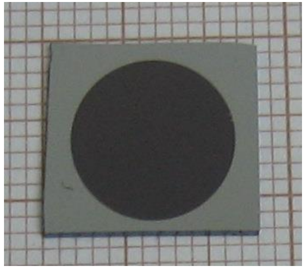


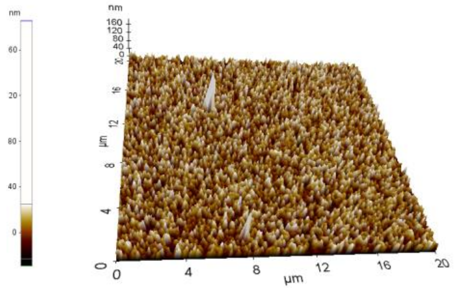
Thermally-evaporated LiF thin films

Optically-transparent polycrystalline LiF films can be grown by thermal evaporation on different substrates, in controlled conditions, tailoring the appropriate geometry, size and thickness.



GP20 deposition system, ENEA C.R. Frascati.





1 µm thick LiF film grown on Si(100) substrate and its 3D AFM image

Controlled deposition parameters

- ✓ pressure $<$ 1 mPa;
- ✓ deposition rate: 0.5 ÷ 2 nm/s;
- ✓ film thickness: up to few µm
- ✓ substrate temperature: 30 ÷ 350 °C
- ✓ nature of substrate: glass, silica, LiF crystals, Si, plastic and metal layers, etc.

LiF radiation imaging detectors

They are based on **optical reading** of **F_2** and **F_3^+ photoluminescence**.

Main features:

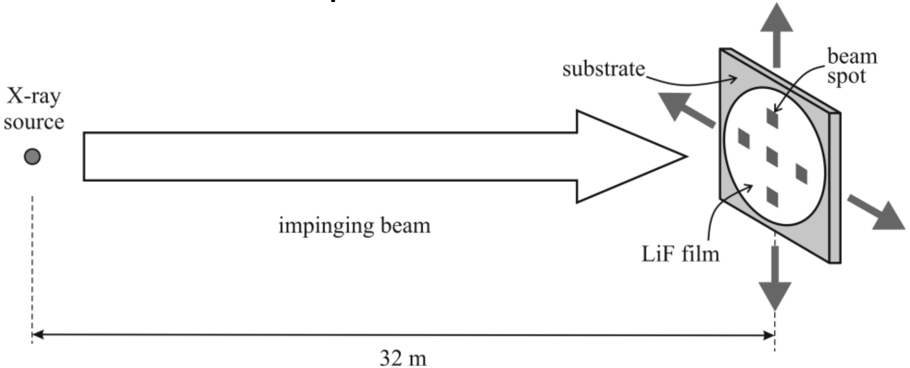
- ✓ **multi-purpose** (X-rays, protons, neutrons, electrons, etc.)
- ✓ **easy handling** (insensitive to light, no development needs)
- ✓ **efficient optical readout process** (Vis spectral range)
- ✓ **fast evaluation time** (seconds)
- ✓ **wide dynamic range** (> 10^5)
- ✓ **high spatial resolution** (intrinsic < 2 nm, standard < 250 nm)
- ✓ **large field of view** (> 1 cm^2)
- ✓ **PL signal stability** (signal stability at RT, multiple evaluations without signal loss)
- ✓ **reusability** (after thermal annealing process).

X-rays irradiation conditions and samples

Detectors:

- ✓ LiF thin films on glass and Si(100) substrates
- ✓ thickness = 0.5, 1.1 and 1.8 μm
- ✓ substrate temperature = 300 °C

METROLOGIE beamline

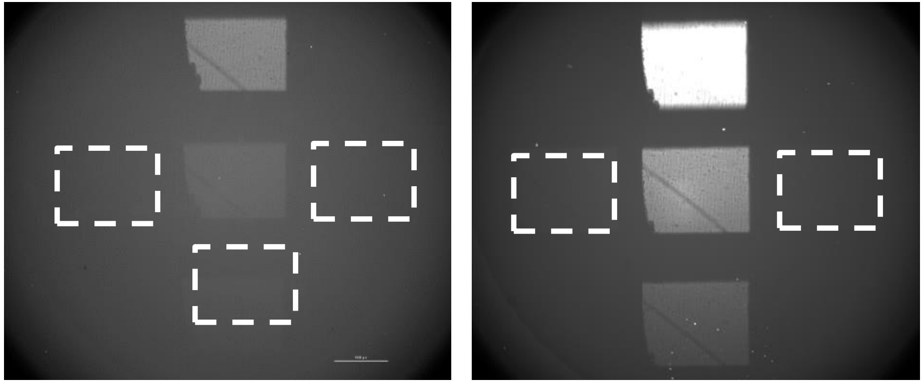


Scheme of the experimental set-up

- ➢ X-ray beam energy = 7 keV
- ➢ Beam transverse area ~ (2×2) mm^2
- ➢ Dose range = $(13 \div 4.5\times10^3)$ Gy
- ➢ X-ray depth of attenuation in LiF ~ 220 μm

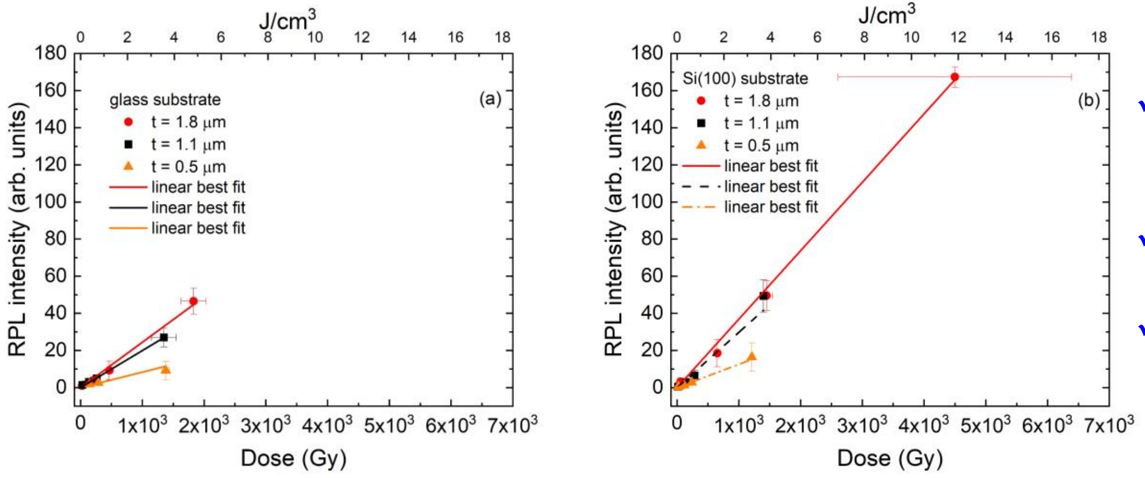
Dose (Gy)
26.9 ± 0.5
51.7 ± 0.3
208 ± 9
467 ± 30
$(1.83 \pm 0.20)\times10^3$

Dose (Gy)
83.3 ± 0.3
156.8 ± 0.7
652 ± 25
$(1.45 \pm 0.10)\times10^3$
$(4.50 \pm 1.89)\times10^3$



Fluorescence images of the thickest LiF films (t = 1.8 μm) grown on glass (left) and Si(100) (right) substrates irradiated with monochromatic 7 keV X-rays at five doses.

Spectrally-integrated PL vs Dose



PL response vs. Dose of LiF film detectors grown on glass (a) and Si(100) (b) substrates irradiated with monochromatic 7 keV X-rays, together with their linear best fit.

- ✓ The PL response of LiF film detectors linearly depends on the irradiation dose in the investigated dose range;
- ✓ At the same irradiation dose, the PL intensity increases with the film thickness;
- ✓ Lowest detected dose = 13 Gy, delivered to the thinnest LiF film;
- ✓ The ratios of the slopes of the best-fit straight lines for the films grown on Si(100) to those on glass in the same deposition run is ~ 1.5. This corresponds to a PL enhancement of about 50%, due to the reflectivity of the silicon substrate in the visible spectral range, where the absorption and emission bands of the F_2 and F_3^+ CCs are located.

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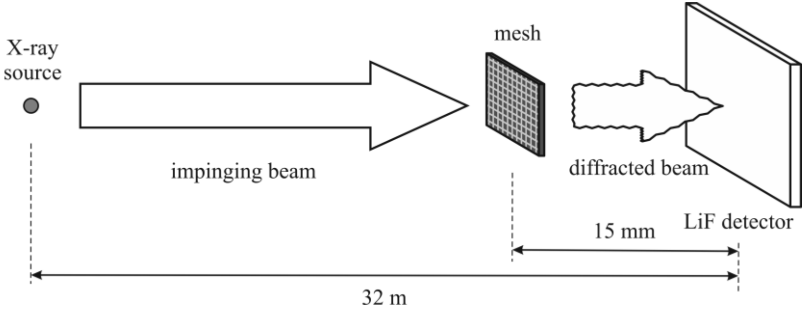
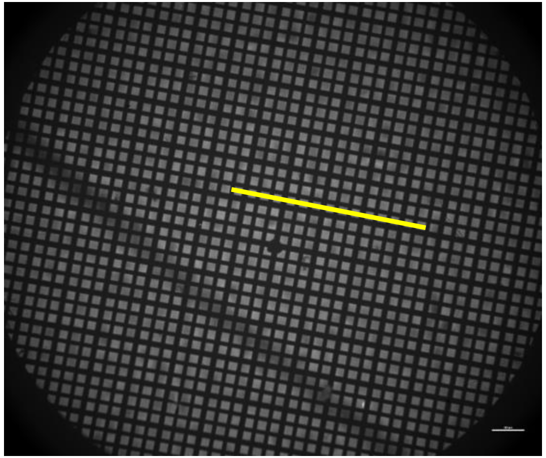
Edge-enhancement X-ray imaging experiments

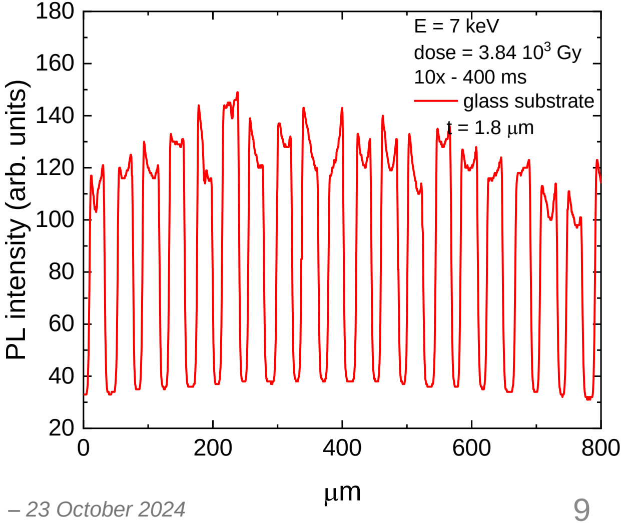


Scheme of the experimental set-up

METROLOGIE beamline

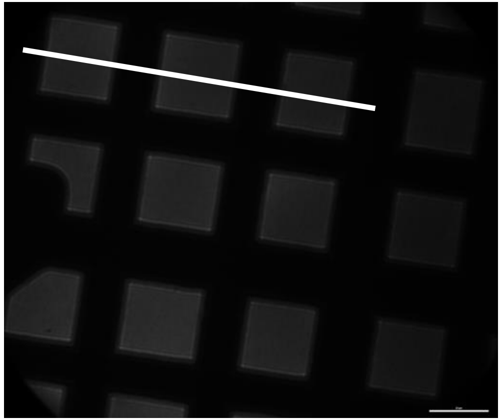


Fluorescence image of the test mesh stored in the LiF film grown on glass, thickness 1.8 µm, dose ~ 4×10^3 Gy (objective magnification 10x, bar size 100 µm).

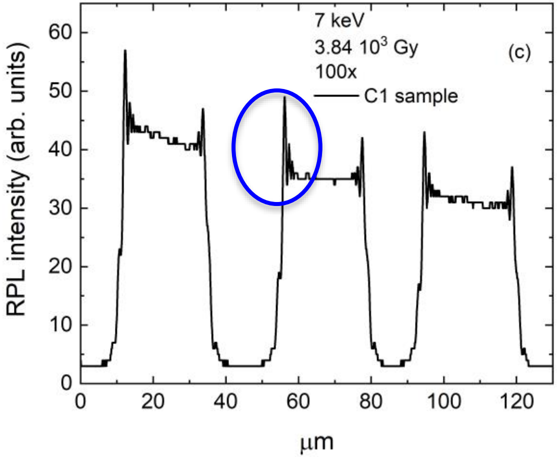


Mesh pitch ~ 41 µm

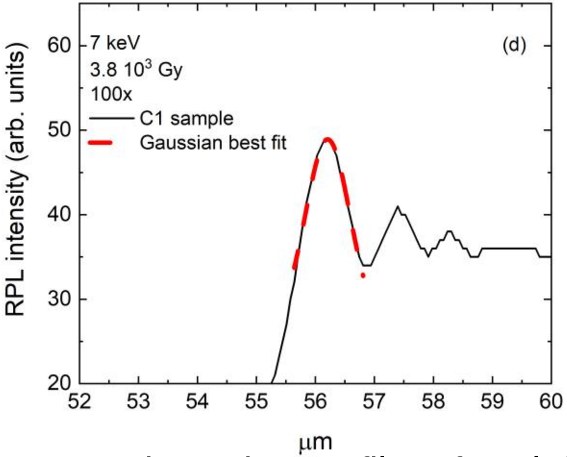
Spatial resolution of LiF detectors



Fluorescence image of the Au mesh stored in the 1.8 µm thick LiF film grown on glass irradiated with 7 keV X-rays, dose = 3.8×10^3 Gy (objective magnification 100×, bar size = 20 µm)



PL intensity profile measured along the white line



PL intensity profile of a left portion of the second fluorescent spot together with the Gaussian best fit (red dashed line) of the highest peak of the diffraction pattern.

Half Width at Half Maximum = (0.38 ± 0.05) µm

Conclusions and future perspectives

- ✓ The PL response of LiF film-based detectors of increasing thicknesses, irradiated with 7 keV X-rays at different doses, was measured using a fluorescence microscope.
- ✓ The PL response shows a linear behavior in the investigated dose range ($13 \div 4.5\times10^3$ Gy) both for LiF films grown on glass and Si(100) substrate.
- ✓ The lowest detected dose was of 13 Gy.
- ✓ A substrate-enhanced PL response amplified by 50% was obtained for LiF film detectors grown on Si(100) with respect to those deposited on glass in the same deposition run.
- ✓ In edge-enhancement imaging experiments, a submicrometric (< 0.5 µm) spatial resolution was obtained on a large field of view (> 1 cm^2).
- ✓ Further experiments with monochromatic X-rays at energies of several keV are under way to study the LiF film sensitivity and their RPL dose response and improve the reproducibility of the observed behavior by a careful control of the film growth conditions.

Thanks for your attention!

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