LITHIUM FLUORIDE X-RAY IMAGING FILM DETECTORS FOR CONDENSED MATTER NUCLEAR MEASUREMENTS

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Lithium Fluoride, LiF, is a radiation sensitive alkali halide material well known as dosimeter and as active medium in light-emitting devices and lasers. Point defects can be produced in LiF crystals and films [1] by different kinds of radiation. Some of these electronic defects, known as colour centres, are optically active, with broad absorption and emission bands in the visible spectral range. Novel thin-film imaging detectors for soft X-rays, based on photoluminescence from aggregate colour centres in LiF, have been proposed [2] and are currently under development [3], successfully extending their operation also in the hard X-ray region, up to 10 keV [4]. Recently their use was proposed and preliminarily tested to obtain the image of radiation emitted from a nickel film hydride loaded by electrolysis, under light coupling with an He-Ne laser [5]. Among the main peculiarities of LiF-film based X-ray imaging detectors, there are an intrinsic high spatial resolution, a large field of view and a wide dynamic range. Moreover, they are easy to handle, as insensitive to visible light and no development process is needed. After exposure to X-rays, the latent images stored in the LiF thin layers are read by advanced optical fluorescence microscopes, with typical spatial resolution below 300nm. These detectors can be applied in photonics, biology, material science, and in the characterization of intense X-ray sources. They allow great versatility, as they can be grown in the form of thin films by well-assessed physical deposition techniques.

[1] R.M. Montereali, "Point Defects in Thin Insulating Films of Lithium Fluoride for Optical Microsystems, in Handbook of Thin Film Materials", H.S.Nalwa ed., Vol.3: Ferroelectric and Dielectric Thin Films, Academic Press, Ch.7, pages 399-431, (2002).
[2] G. Baldacchini, F. Bonfigli, A. Faenov, F. Flora, R.M. Montereali, A. Pace, T. Pikuz, and L.Reale, "Lithium Fluoride as a Novel X-Ray Image Detector for Biological μ-World Capture", J. Nanoscience and Nanotechnology 3, 6, pages 483-486, (2003).
[3] G. Baldacchini, S. Bollanti, F. Bonfigli, F. Flora, P. Di Lazzaro, A. Lai, T. Marolo, R.M. Montereali, D. Murra, A. Faenov, T. Pikuz, E. Nichelatti, G. Tomassetti, A. Reale, L. Reale, A. Ritucci, T. Limongi, L. Palladino, M. Francucci, S. Martellucci and G. Petrocelli, "A Novel Soft X-Ray Submicron Imaging Detector Based on Point Defects in LiF", Review Scientific Instruments 76, pages 113104:1-12, (2005) (Also in the Virtual Journal of Biological Physics Research 10(11), December 1, 2005).
[4] S. Almaviva, F. Bonfigli, I. Franzini, A. Lai, R.M. Montereali, D. Pelliccia, A. Cedola, S. Lagomarsino, "Hard X-Ray Contact Microscopy with 250 nm Spatial Resolution using a LiF Film Detector and a Tabletop Microsource", Appl. Phys. Lett. 89, pages 54102-54104 (2006).
[5] R.M. Montereali, S. Almaviva, E. Castagna, T. Marolo, F. Sarto, C. Sibilia, M.A. Vincenti and V. Violante, "A Novel LiF-based Detector for X-Ray Imaging in Hydrogen Loaded Ni Films under Laser Irradiation", in Condensed Matter Nuclear Sciences, Proc.12th Int. Conf. On Cold Fusion, Yokohama, Japan 27 Nov – 2 dec 2005, A. Takahashi, K. Ota and Y. Iwamura, eds., World Scientific, pages 251-255, (2006) ISBN 981-256-901-4.

Lithium Fluoride X-Ray Imaging Film Detectors for Condensed Matter Nuclear Measurements

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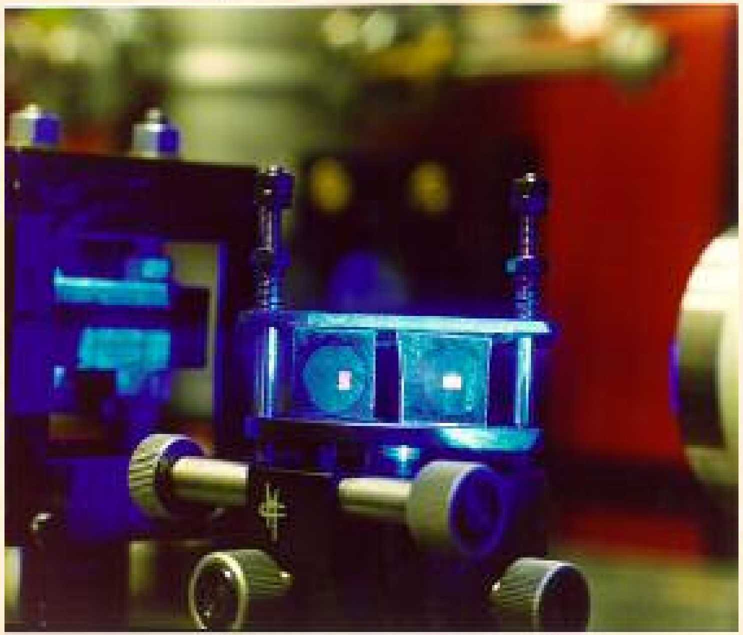
http://www.frascati.enea.it/fis/lac/solidstate/



Solid-state **green**-**red** light emitters based on LiF films thermally evaporated on **silicon** (left) and glass (right) substrates

Introduction and motivation

We recently proposed an innovative

film-like soft-hard X-ray imaging detector based on **photoluminescence (PL)** of radiation-induced **active color centers** in **Lithium Fluoride (LiF) thin layers**, with

- **High spatial resolution**
- **Large field of view**
- **Wide dynamic range**
- **Efficient photoluminescence readout process**
- **Easy handling: no development needs and no sensitivity to visible light**
- **Compatible with permanent protective layers and different substrates**

It is currently under further development in soft-hard X-rays for imaging applications in **biology, photonics, material science, characterization of intense X-ray sources…**

Outline

- **Introduction**
  - Lithium Fluoride: material properties
  - Primary and aggregate electronic defects in LiF
- **Experimental**
  - LiF films: growth and characterization
  - X-ray irradiation and characterization of LiF crystals and films
- **Results**
  - Primary and aggregate color centers vs irradiation dose in LiF crystals
  - X-ray imaging applications in LiF films: examples
- **Future perspectives**

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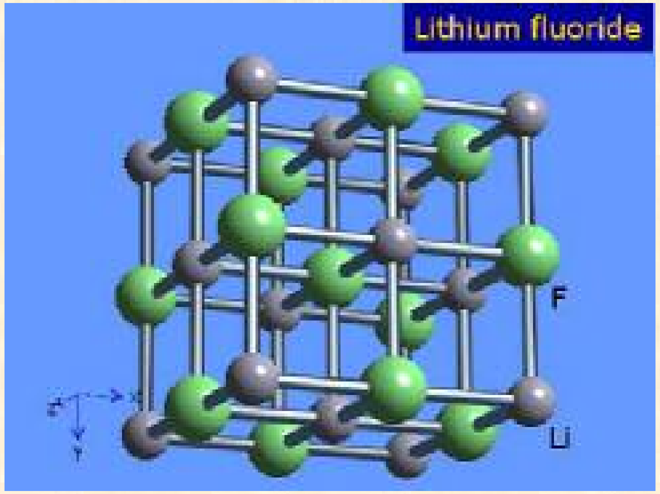
Lithium Fluoride (LiF)

Color Centers (CCs): point defects in insulating materials

Alkali Halides (AH): ionic crystals with fcc structure, optically transparent from near UV to IR.



LiF stands apart because

- it is almost **non-hygroscopic;**
- **polycrystalline LiF films** can be grown by thermal evaporation on different substrates;
- it can host **CCs stable at RT;**
- it can host **laser active CCs tunable in a broad wavelength range in the visible and near IR.**

It can be colored only by **ionizing radiation**, like **elementary particles** and **ions**, as well as **photons**, such us **EUV light, X-rays**, γ rays and even intense ultra-short laser pulses.

Nearest neighbour distance (Å)	2.013
Melting point (°C)	848.2
Density (g/cm^3)	2.640
Molecular weight	25.939
Refractive index @ 640 nm, RT	1.3912
Solubility (g/100g H_2O @ 25°C)	0.134
Hardness (Knoop 600 g indenter)	102
Transmission range (μm)	0.12 - 7

Irradiation of LiF gives rise to **stable formation** of **primary and aggregate CCs**, which generally coexist with often overlapping absorption bands.

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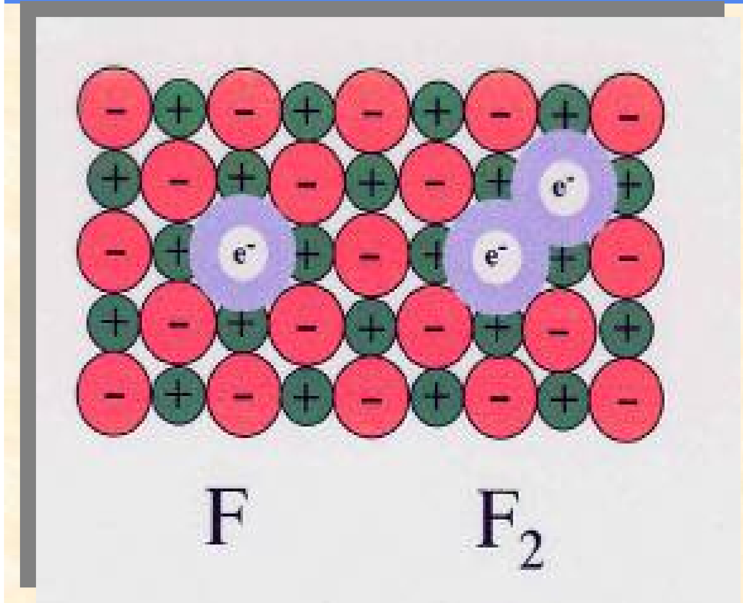
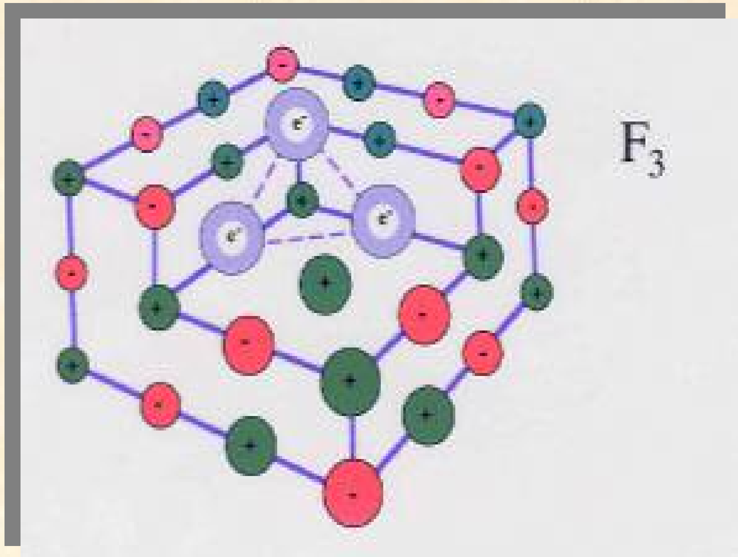
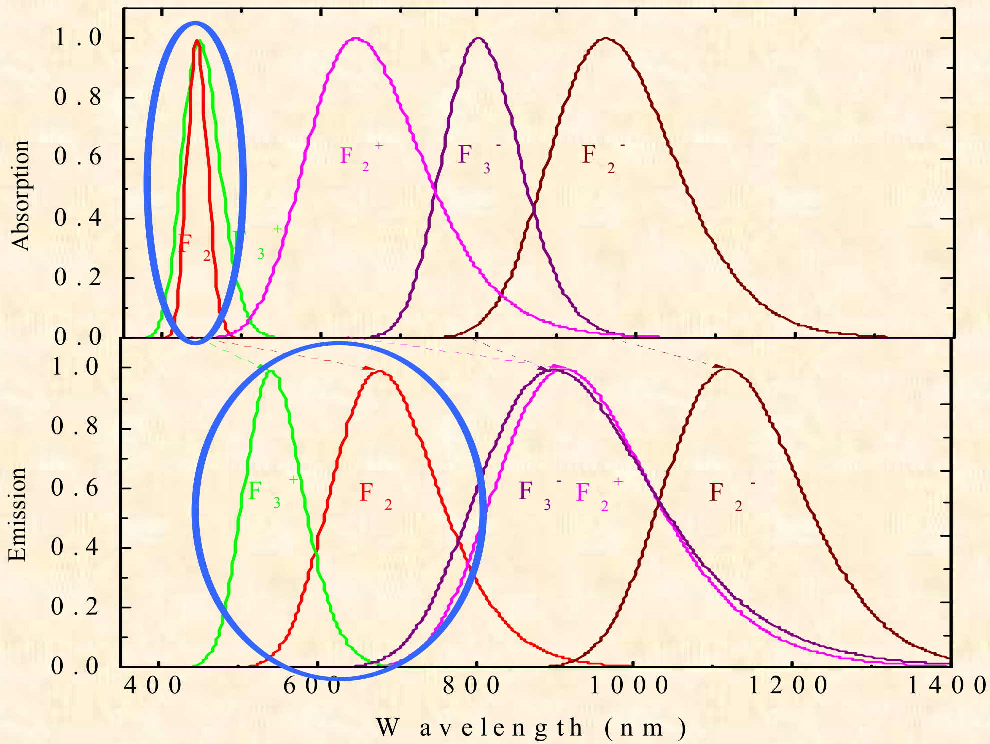
Laser active color centers in LiF at RT







F center is an anion vacancy occupied by an electron; it is not an optically active centers in LiF.

F_2 and **F_3^+ centers** are optically active F-aggregates consisting in two electrons bound to two and three close anion vacancies, respectively.

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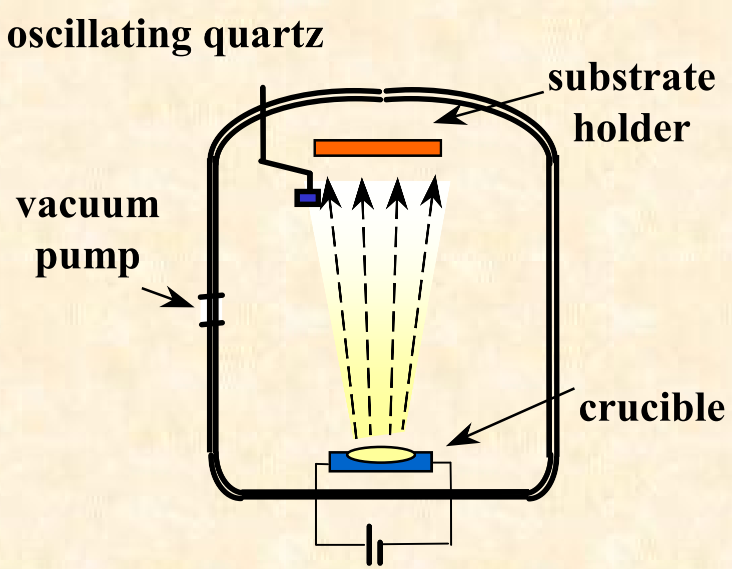
LiF film deposition by thermal evaporation

Polycrystalline films are grown by thermal evaporation on **amorphous** (glass, silica, silica on silicon, …) and **crystalline** (LiF single crystals, NaF, MgF_2, silicon, …) **substrates**. The **structural, morphological and optical properties of the films** are strongly dependent on

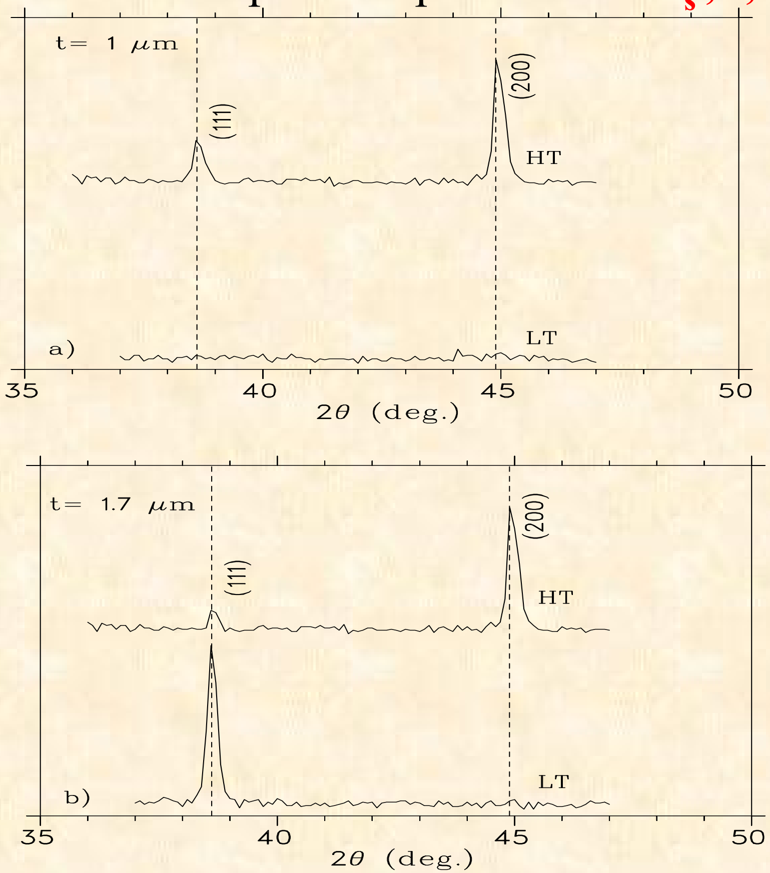
➔ the nature of the substrate

➔ the deposition parameters: T_s, t, R



Deposition parameters:

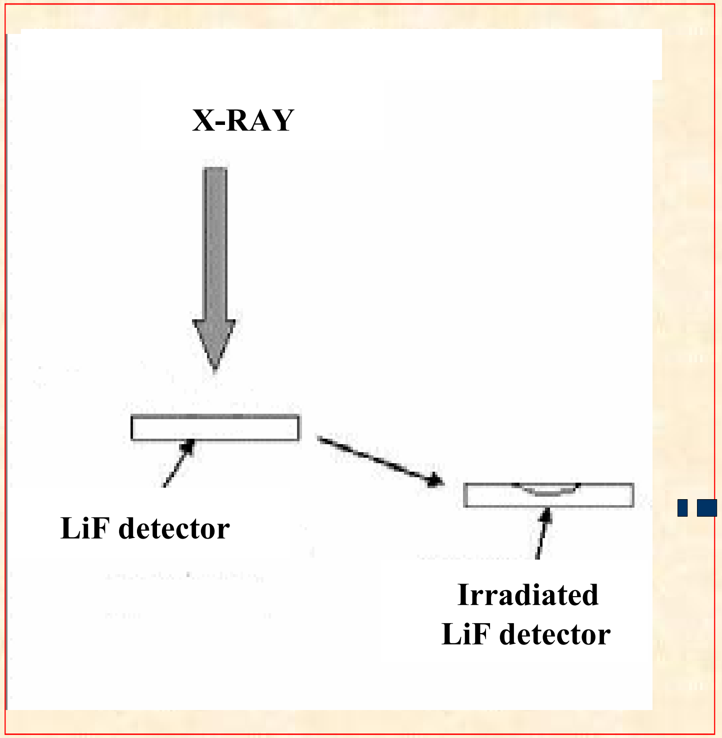
- Pressure < 10^{-6} mbar
- Evaporation rate **R** = 0.5-2 nm/s
- Total film thickness **t** = 0.2 - 6 µm
- Substrate temperature **Ts**=30-350°C



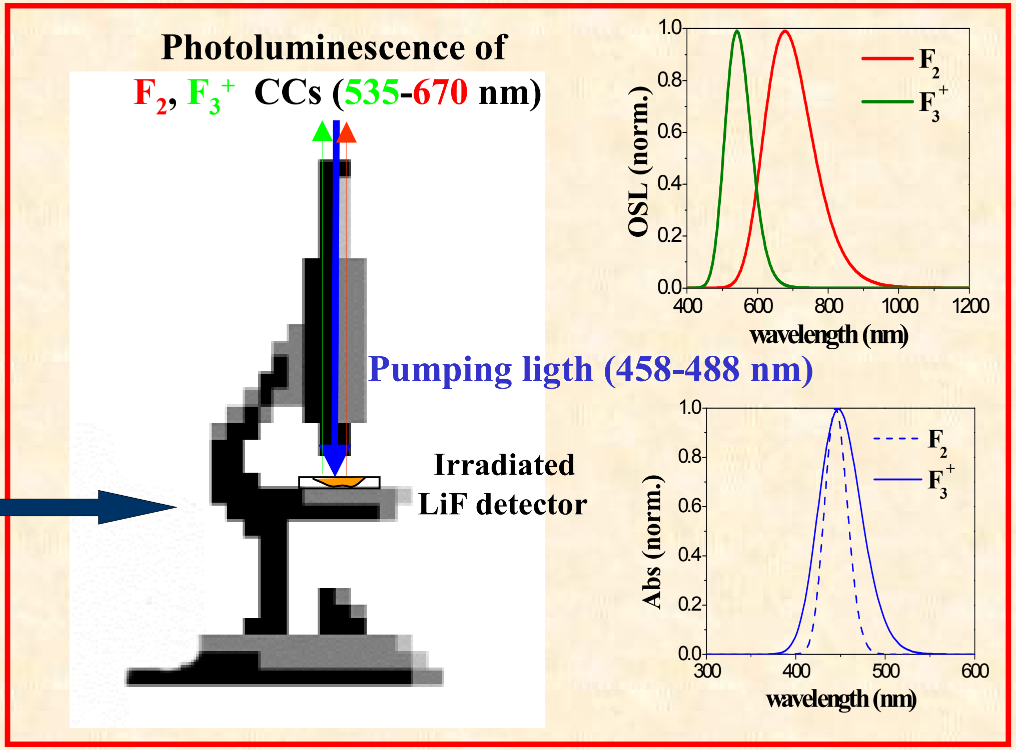
θ-2θ diffraction patterns of LiF films grown on glass at T_s=30°C(LT) and 300°C(HT) with two different t.

LiF-based X-ray imaging detector: optical readout technique

Irradiation process



Readout process: photoluminescence (PL)



Permanent fluorescent patterns based on **F_2** and **F_3^+ defects in LiF** can be produced by using several X-ray sources in different configurations (contact mode, direct writing, projection mode, etc.)

The permanent photoluminescent patterns, stored in the irradiated LiF samples, are observed by using **optical microscopes in fluorescence mode**. Irradiation with **blue** light excites the **visible photoluminescence of the F_2 and F_3^+ defects** locally created in the areas previously exposed to the X-ray beam.

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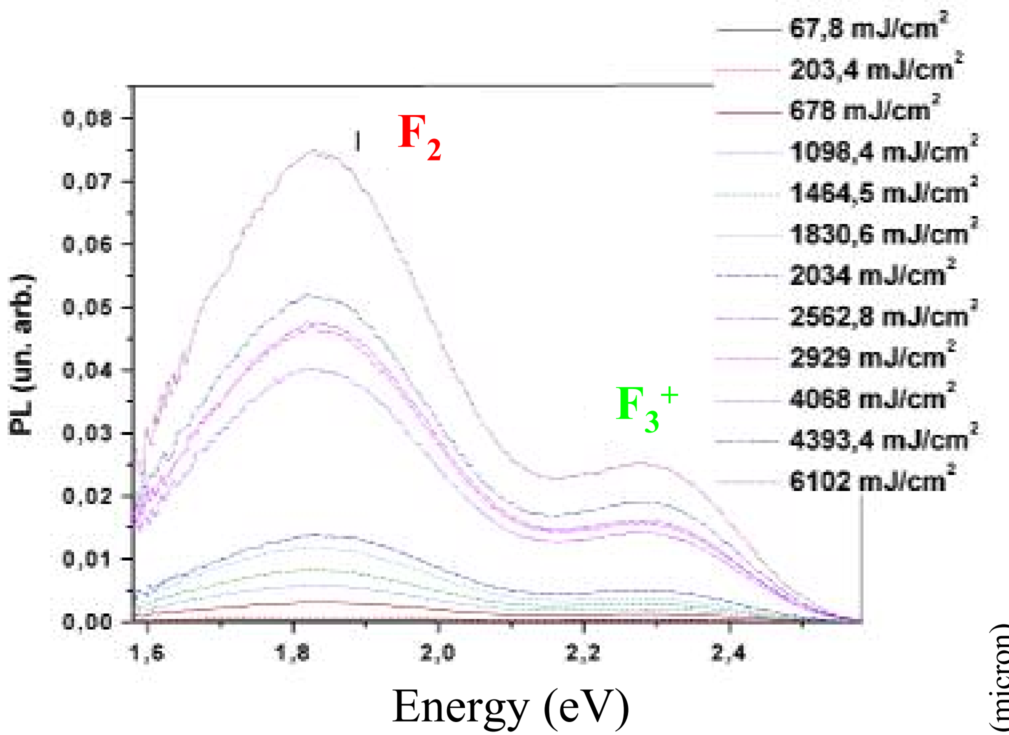
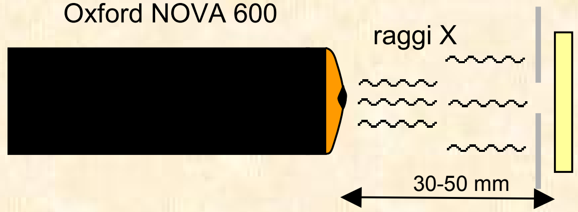
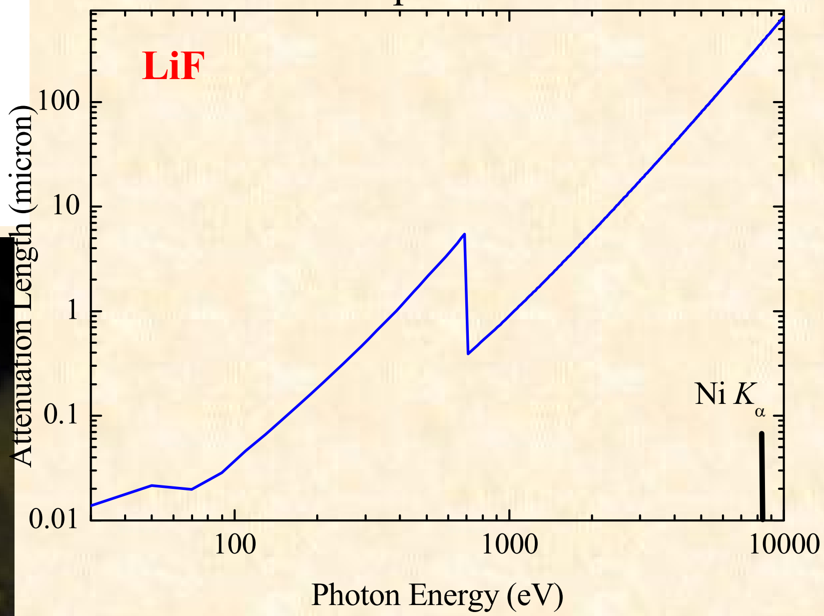
RT photoluminescence spectra of colored LiF crystals vs dose

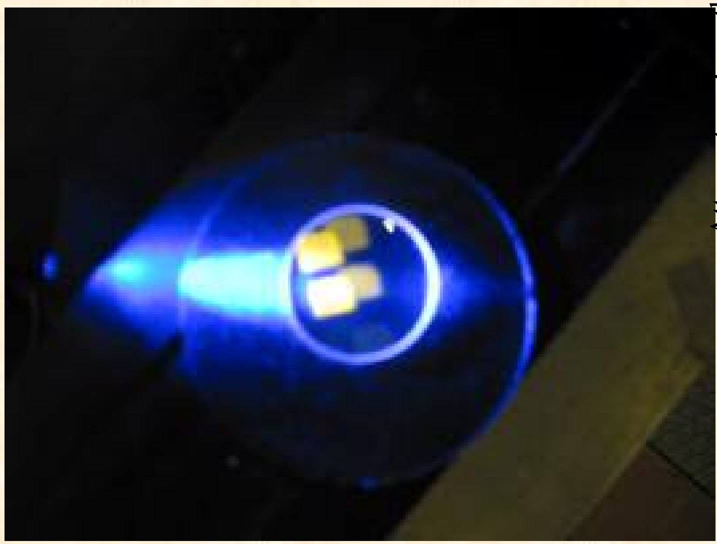




Typical irradiation parameters:
Target voltage: 20 - 30 kV
Target current: 0.5 – 2 mA
Exposure time: 1 – 90 min
Flux: 6×10^{11} photon/s x sr



Argon laser excitation at λ_p = 458 nm



Cu-K_α = 8.042 keV

Ni-K_α = 8.333 keV

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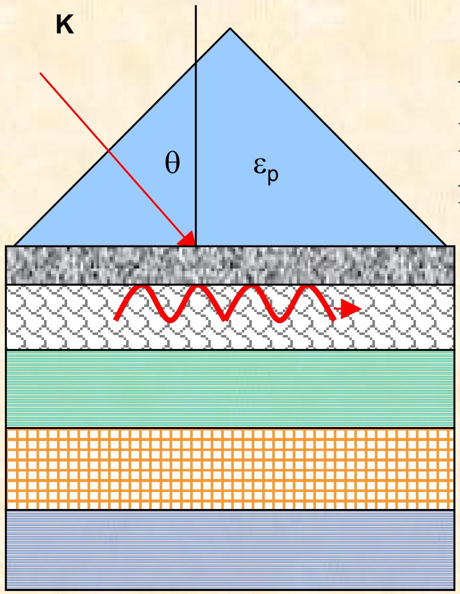
The experiment

Surface plasmons (**polaritons**) are quantum of plasma oscillations created by the collective oscillation of electrons on a solid surface. They may be generated by mechanisms able to produce charge separation between Fermi level electrons and a background of positive charges (i.e. lattice atoms).



Sputtered **Ni film** previously loaded with hydrogen by electrolysis with 1 M Li_2SO_4 electrolyte in light water (40 minutes, current ranging 10 to 30 mA).

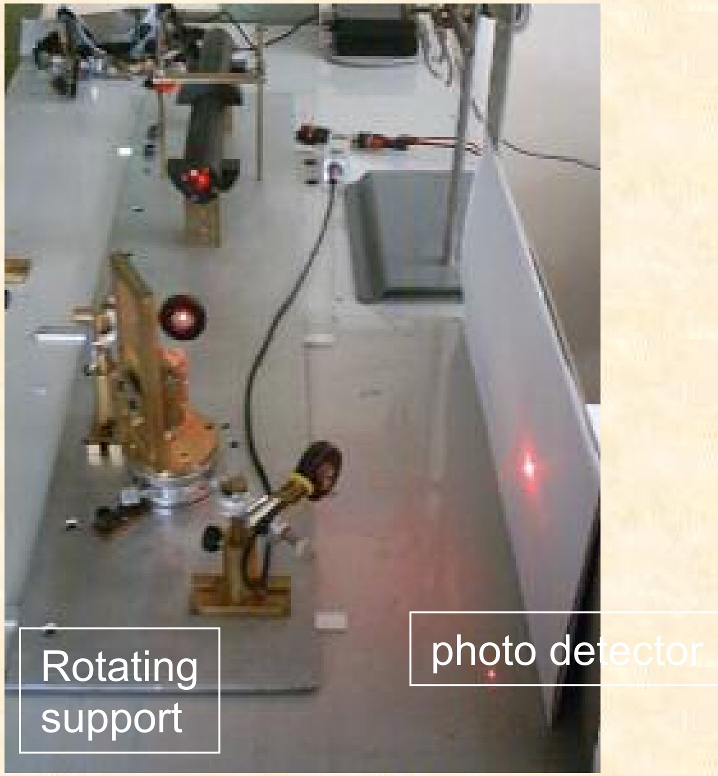
45 nm thick **Ni film**, on

1 mm thick **polyethylene** substrate

LiF film (t=1.9 μm) on

1 mm thick **glass** substrate

The **LiF film** detector, consisting in a LiF film thermally evaporated on glass, has been mounted in close contact with the back-side of the hydride Ni sample, positioned on a rotating support at the selected reflectance minimum angle under a **c.w. He-Ne laser** (**632.8 nm**, 5 mW), coupled in the metallic layer trough a glass cylindrical lens placed on the Ni surface for an irradiation time of 3h.



R.M.Montereali, S.Almaviva, E.Castagna, T.Marolo, F.Sarto, C.Sibilia, M.A.Vincenti and V.Violante, in Condensed Matter Nuclear Sciences, Proc.12th Int. Conf. On Cold Fusion, Yokohama, Japan 27 Nov – 2 dec 2005, A. Takahashi, K. Ota and Y. Iwamura, eds., 2006, World Scientific, p.251-255.

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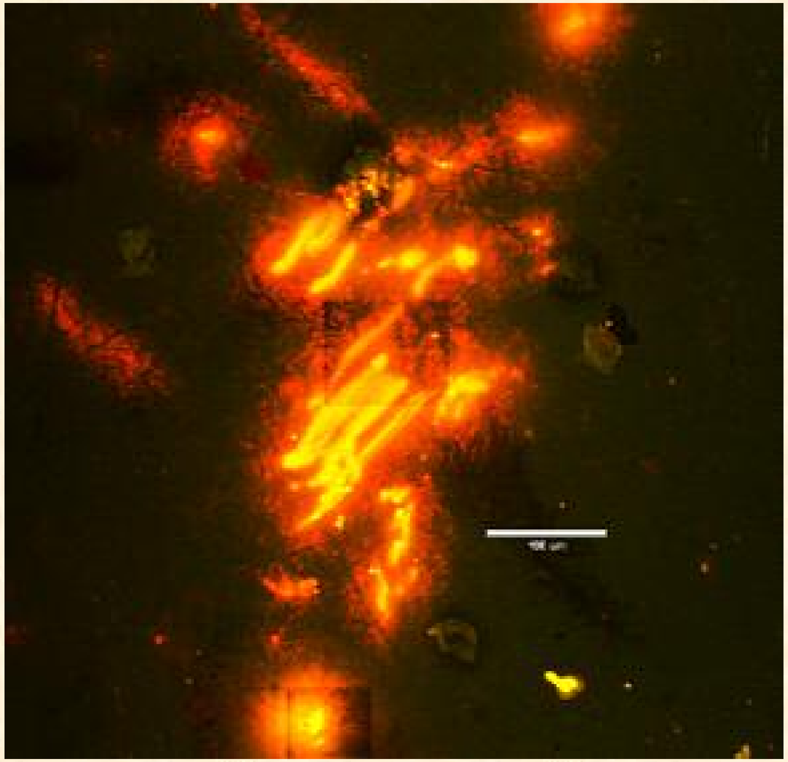
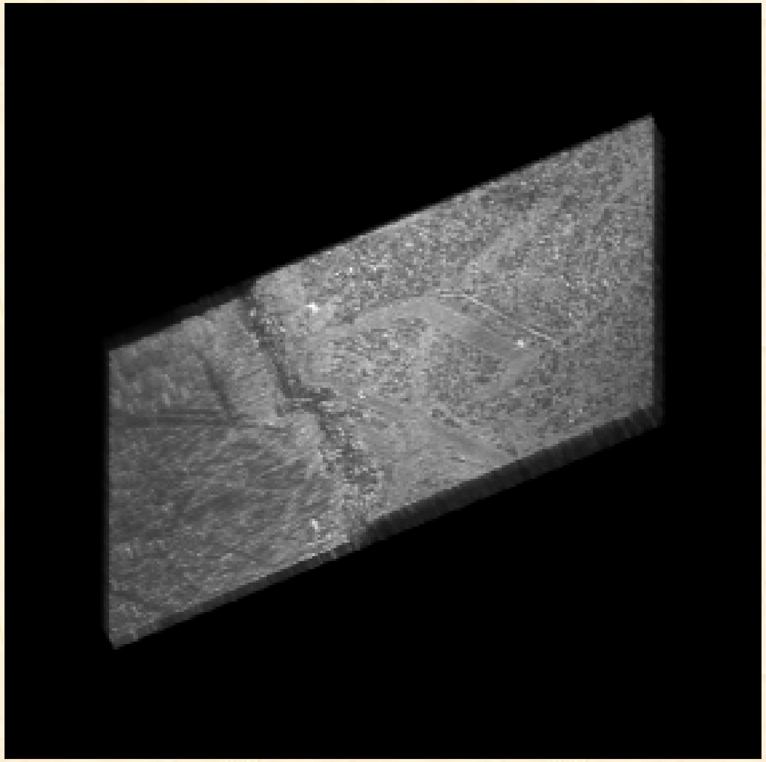
CLSM investigation on exposed and blank LiF films

The coupled e.m. wave can produce coherent oscillations of the Fermi-level electrons in the metal Ni lattice, as its frequency is quasi-resonant with electronic plasma one. The excitation could produce local intense electric field, and X-ray emission at energies below the Ni K_α edge can take place.



2-D confocal image in fluorescence mode of the exposed LiF film on glass. **Several light-emitting spots, closely grouped, with typical spatial dimension from tens to hundreds of micrometers,** are detected.

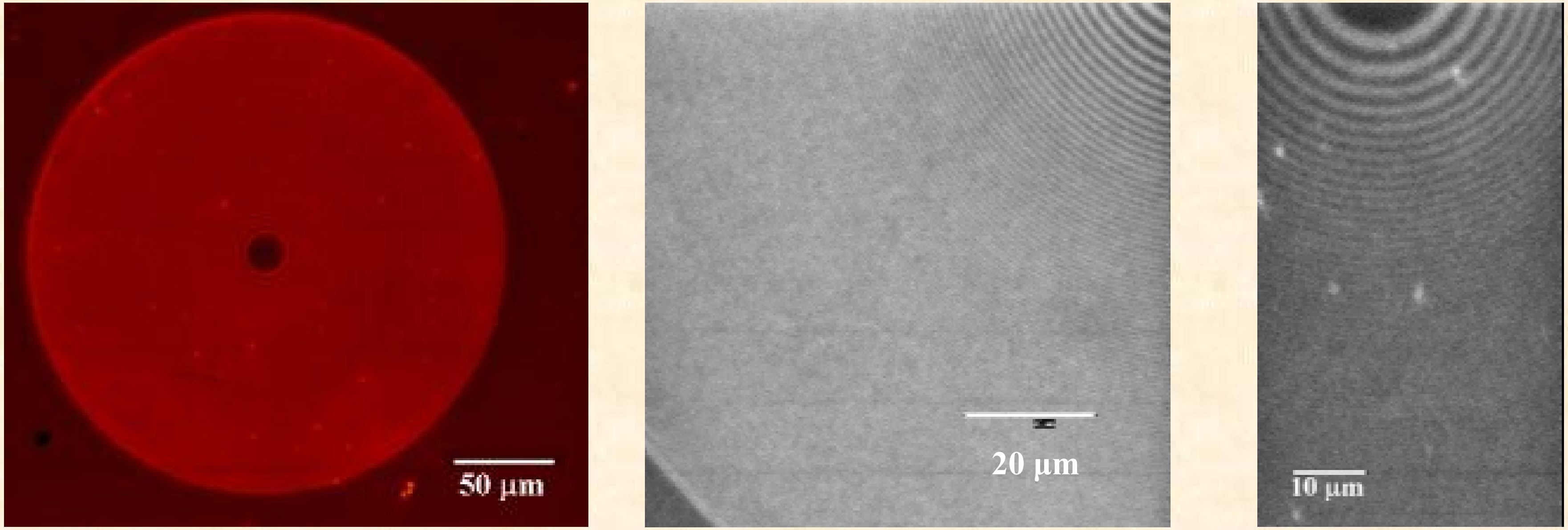


3-D confocal image (60x) in reflection mode of a LiF film on glass (212x212 μm^2).

Conclusions

- Promising results in **X-ray imaging** have been obtained for **hard X-rays** (8 keV)
- **Efficient formation of stable color centers** in **LiF** crystals has been obtained.
- **Intense broad visible photoluminescence at RT** has been measured.
- **X-ray micro-radiography and microscopy images on LiF** crystals and films have been obtained with a sub-micrometric spatial resolution.

The main features of these LiF films based **X-ray imaging detectors** are promising for many applications, including radiation detection in NFCM.



Zone plate X-ray micro-radiography confocal images on a 1.4 µm thick LiF film grown on a glass substrate irradiated by OXFORD microfocus.

S.Almaviva, F.Bonfigli, I.Franzini, A.Lai, R.M.Montereali, D.Pelliccia, A.Cedola, S.Lagomarsino, Appl. Phys. Lett. 89(2006)54102-4