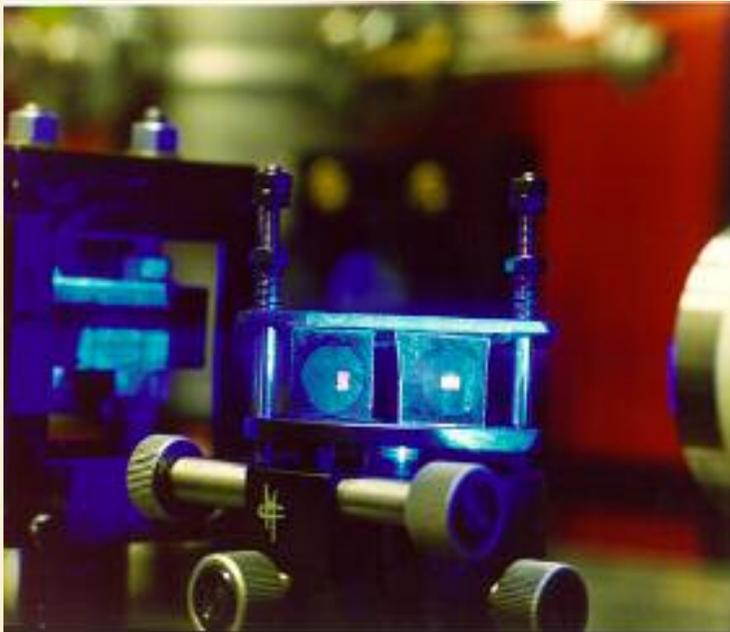


Lithium Fluoride X-Ray Imaging Film Detectors for Condensed Matter Nuclear Measurements

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

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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**

Lithium Fluoride (LiF)

Color Centers (CCs): point defects in insulating materials

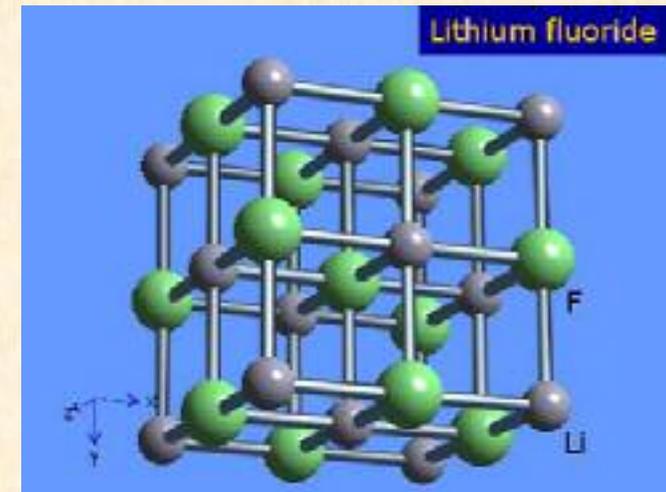
Alkali Halides (AH): ionic crystals with fcc structure, optically transparent from near UV to IR.

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.**

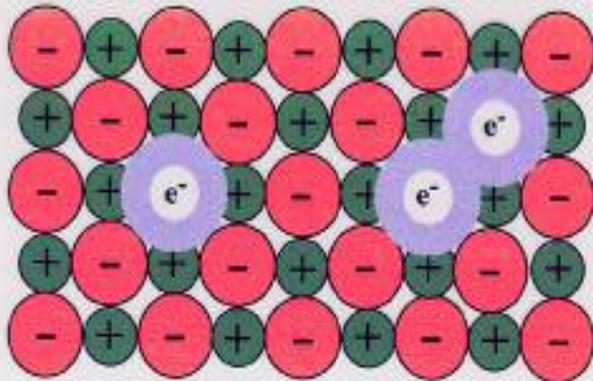
It can be colored only by **ionizing radiation**, like **elementary particles** and **ions**, as well as **photons**, such as **EUV light, X-rays**, γ rays and even intense ultra-short laser pulses.

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

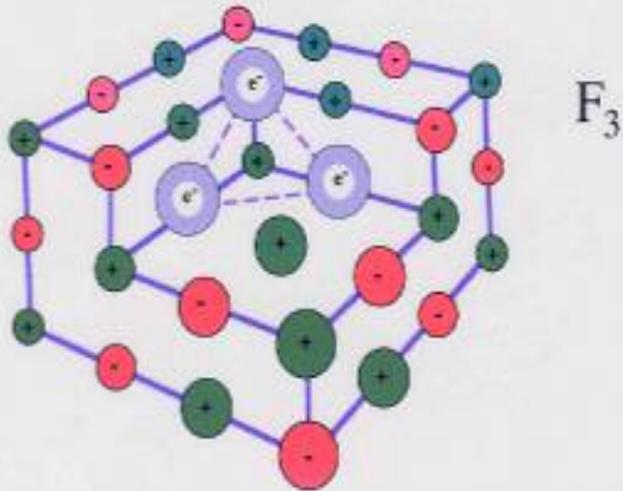


Nearest neighbour distance (Å)	2.013
Melting point (°C)	848.2
Density (g/cm ³)	2.640
Molecular weight	25.939
Refractive index @ 640 nm, RT	1.3912
Solubility (g/100g H ₂ O @ 25°C)	0.134
Hardness (Knoop 600 g indenter)	102
Transmission range (μm)	0.12 - 7

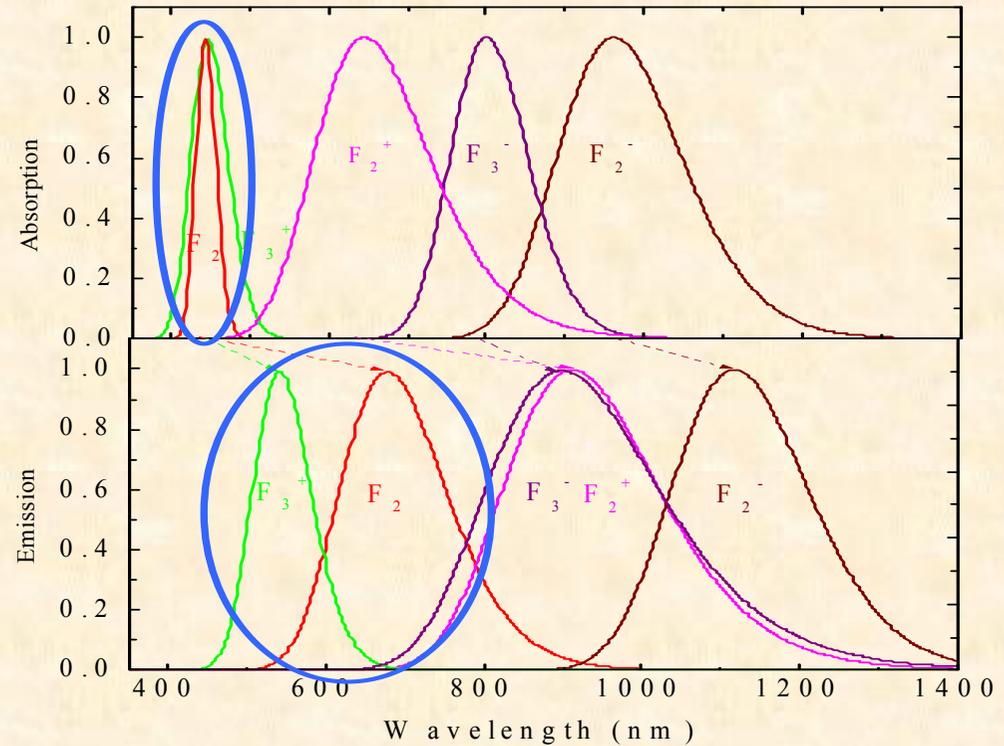
Laser active color centers in LiF at RT



F F₂



F₃



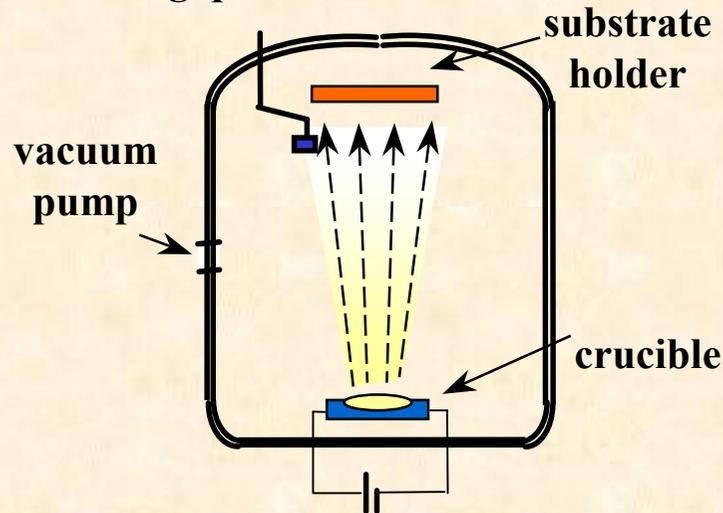
F center is an anion vacancy occupied by an electron; it is not an optically active center in LiF.

F₂ and **F₃⁺ centers** are optically active F-aggregates consisting in two electrons bound to two and three close anion vacancies, respectively.

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, MgF₂, silicon, ...) **substrates**. The **structural, morphological and optical properties of the films** are strongly dependent on
→ the **nature of the substrate**

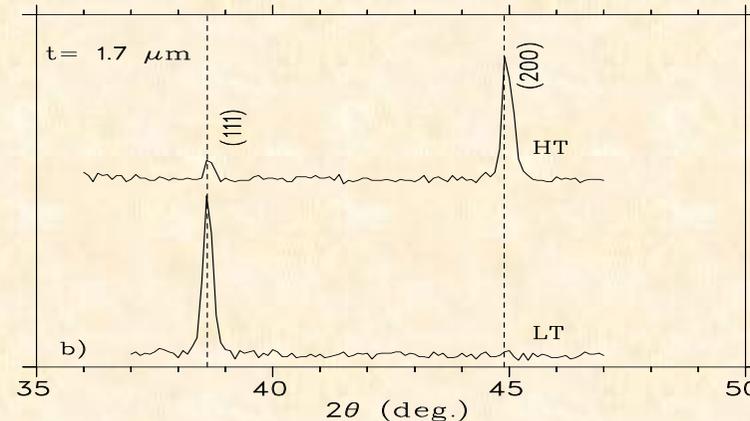
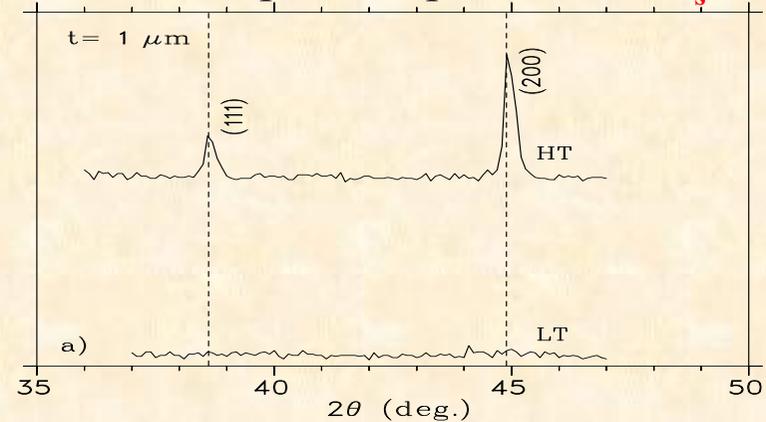
oscillating quartz



→ the deposition parameters: **T_s, t, R**

Deposition parameters:

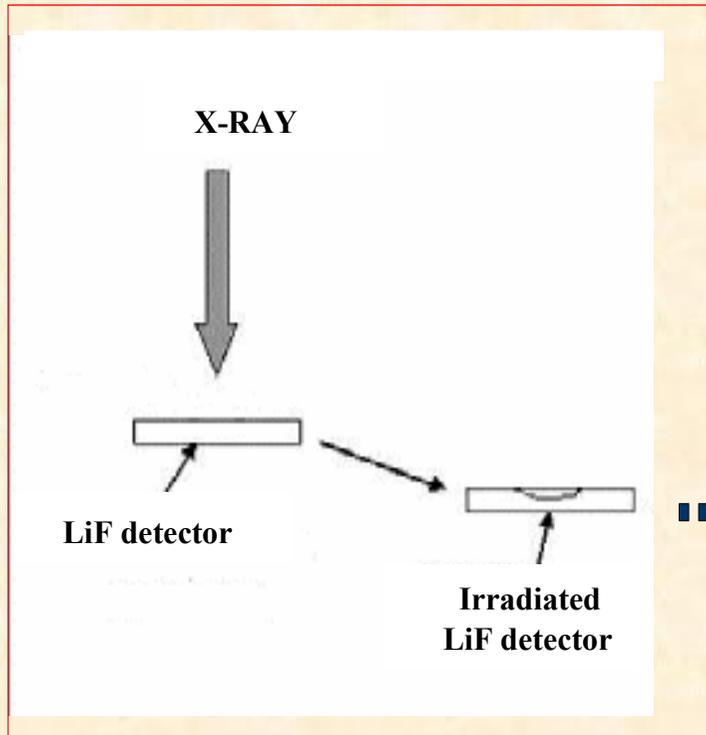
- Pressure < 10⁻⁶ mbar
- Evaporation rate **R** = 0.5-2 nm/s
- Total film thickness **t** = 0.2 - 6 μm
- Substrate temperature **T_s** = 30-350°C



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

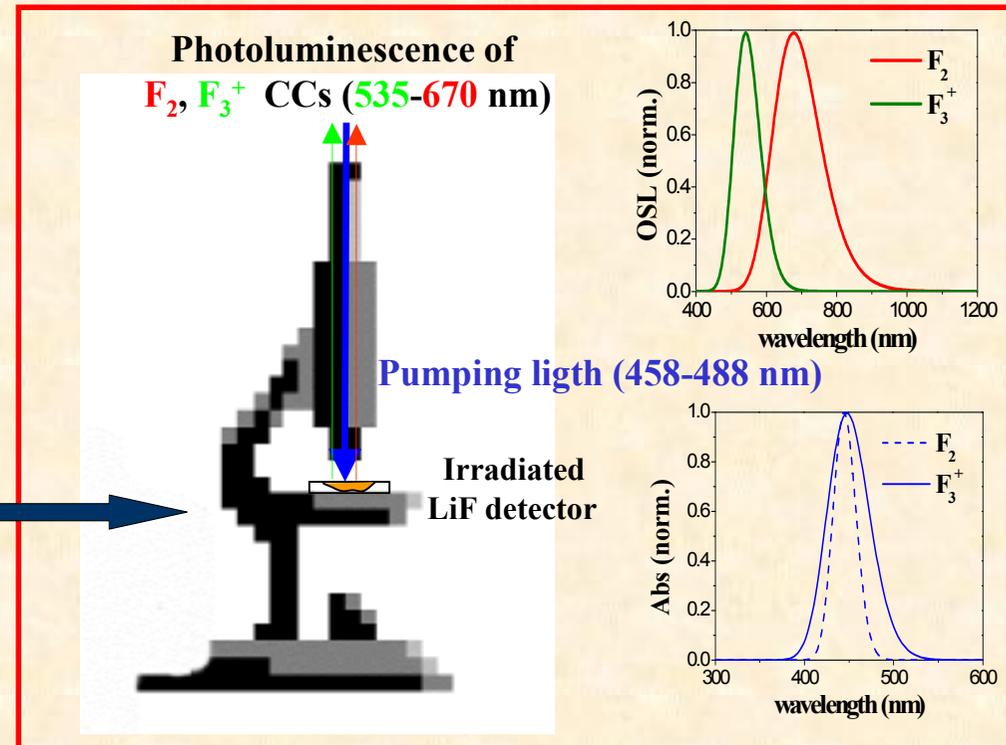
LiF-based X-ray imaging detector: optical readout technique

Irradiation process



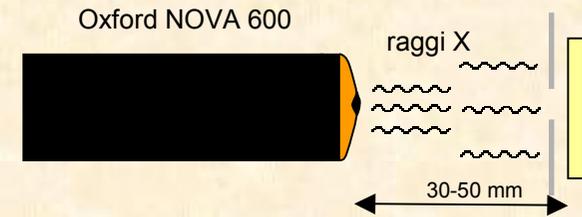
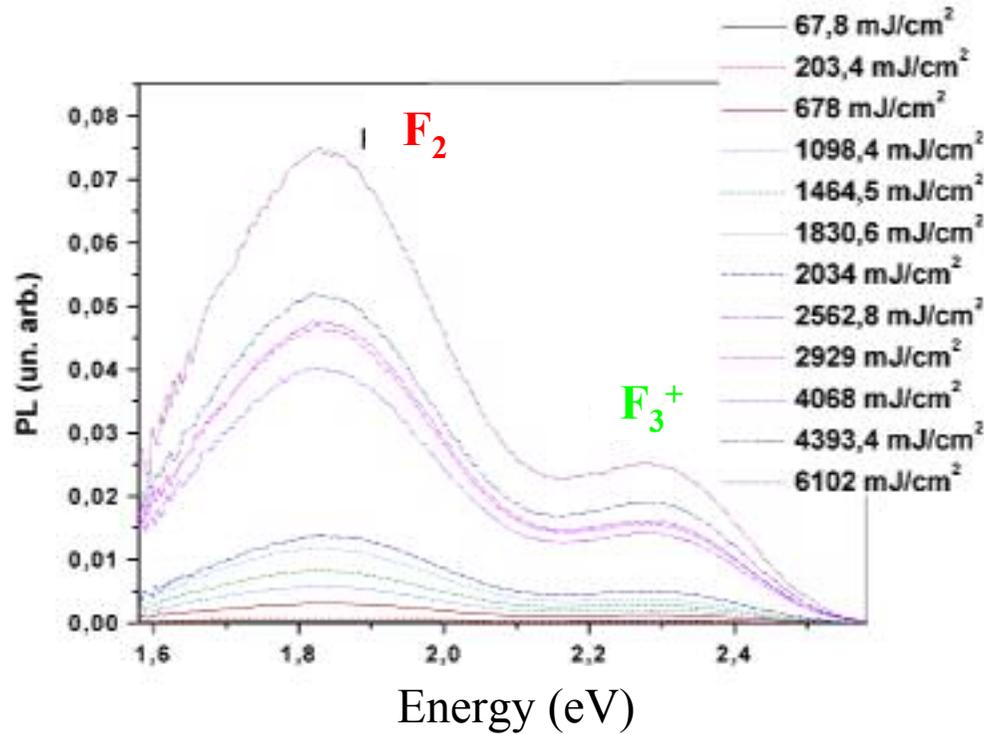
Permanent fluorescent patterns based on F_2 and F_3^+ defects in LiF can be produced by using several X-ray sources in different configurations (contact mode, direct writing, projection mode, etc.)

Readout process: photoluminescence (PL)

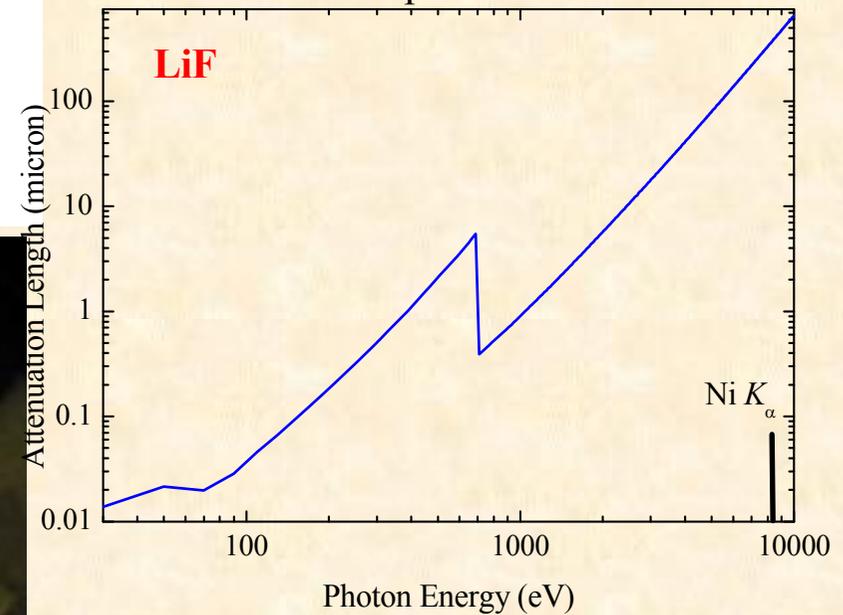


The permanent photoluminescent patterns, stored in the irradiated LiF samples, are observed by using **optical microscopes in fluorescence mode**. Irradiation with **blue** light excites the **visible photoluminescence of the F_2 and F_3^+ defects** locally created in the areas previously exposed to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

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



Cu-K_α = 8.042 keV

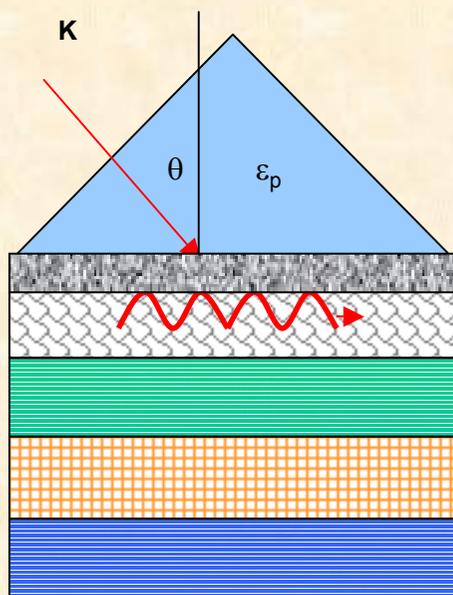
Ni-K_α = 8.333 keV

Argon laser
 excitation at
 $\lambda_p = 458 \text{ nm}$



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).

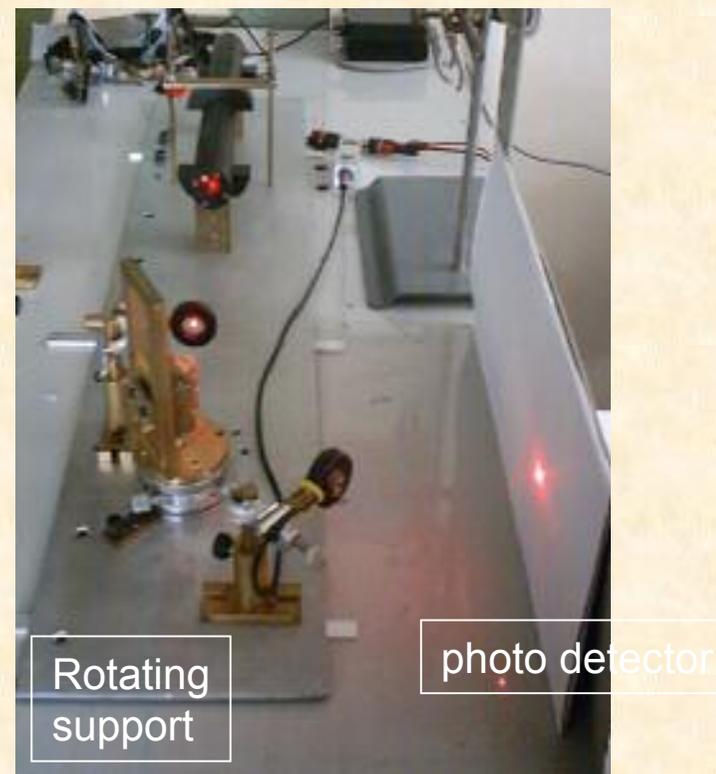


Sputtered Ni film previously loaded with hydrogen by electrolysis with 1 M Li_2SO_4 electrolyte in light water (40 minutes, current ranging 10 to 30 mA).

45 nm thick Ni film, on
1 mm thick polyethylene substrate

LiF film ($t=1.9 \mu\text{m}$) on
1 mm thick glass substrate

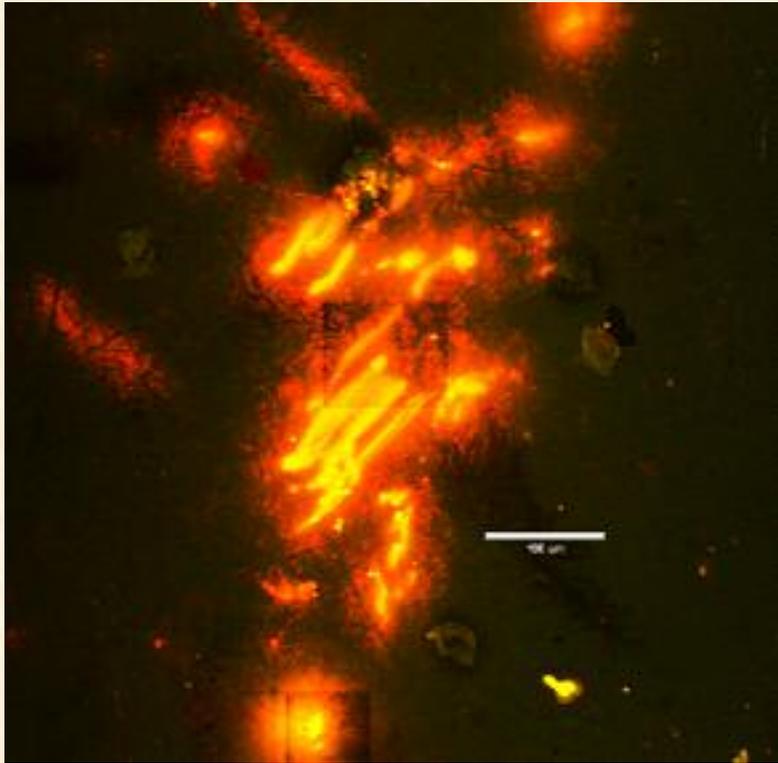
The LiF film detector, consisting in a LiF film thermally evaporated on glass, has been mounted in close contact with the back-side of the hydride Ni sample, positioned on a rotating support at the selected reflectance minimum angle under a c.w. He-Ne laser (632.8 nm, 5 mW), coupled in the metallic layer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



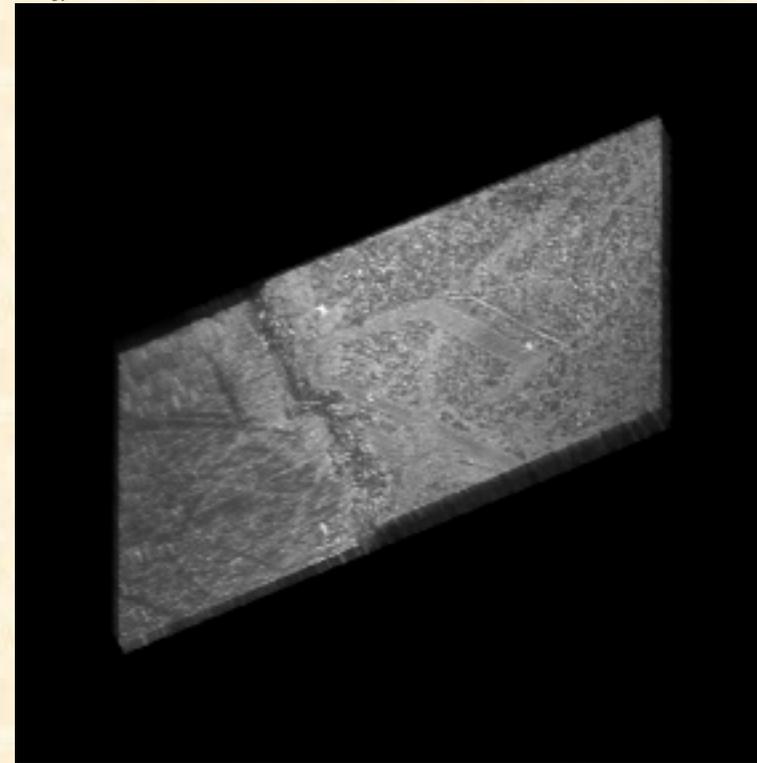
R.M.Montereali, S.Almaviva, E.Castagna, T.Marolo, F.Sarto, C.Sibilia, M.A.Vincenti and V.Violante, in *Condensed Matter Nuclear Sciences, Proc.12th Int. Conf. On Cold Fusion, Yokohama, Japan 27 Nov – 2 dec 2005*, A. Takahashi, K. Ota and Y. Iwamura, eds., 2006, World Scientific, p.251-255.

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.

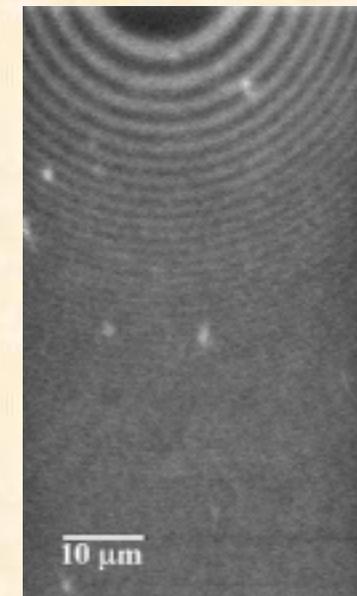
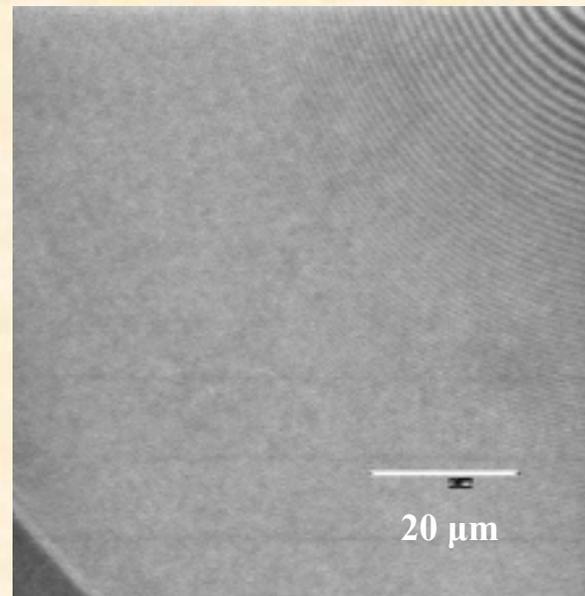
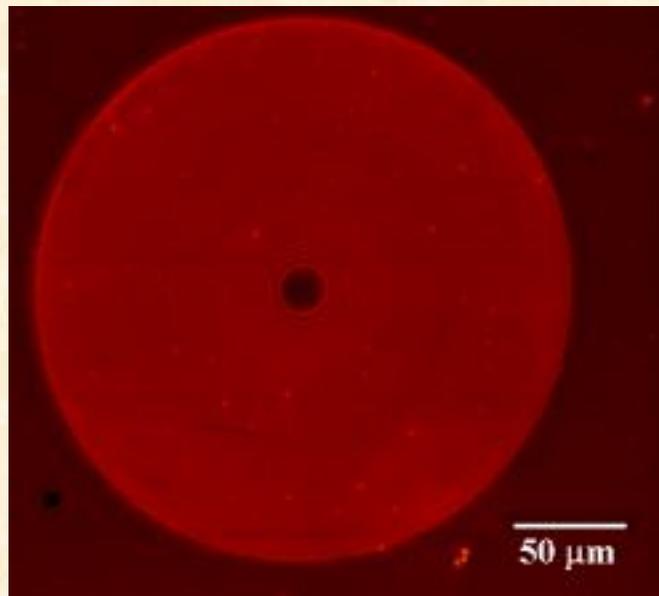


3-D confocal image (60x) in reflection mode of a LiF film on glass ($212 \times 212 \mu\text{m}^2$).

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