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

R.M. Montereali¹, S. Almaviva¹, F. Bonfigli¹, E. Castagna², F. Sarto², M.A. Vincenti¹, V. Violante²

¹ENEA, Physical Technologies and New Materials Dept., C.R. Frascati, Via E. Fermi 45, 00044 Frascati, Rome (Italy)
²Associazione Euratom-ENEA sulla Fusione, C.R. Frascati, Via E. Fermi, 45, 00044 Frascati (RM), Italy

email: montereali@frascati.enea.it
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
We recently proposed an innovative \textit{film-like soft-hard X-ray imaging detector} based on \textit{photoluminescence (PL)} of radiation-induced \textit{active color centers} in \textit{Lithium Fluoride (LiF) thin layers}, with

- \textbf{High spatial resolution} \hspace{1cm} \textbf{Large field of view}
- \textbf{Wide dynamic range} \hspace{1cm} \textbf{Efficient photoluminescence readout process}
- \textbf{Easy handling: no development needs and no sensitivity to visible light}
- \textbf{Compatible with permanent protective layers and different substrates}

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

\textbf{Outline}

\textbf{Introduction}

- Lithium Fluoride: material properties
- Primary and aggregate electronic defects in LiF

\textbf{Experimental}

- LiF films: growth and characterization
- X-ray irradiation and characterization of LiF crystals and films

\textbf{Results}

- Primary and aggregate color centers vs irradiation dose in LiF crystals
- X-ray imaging applications in LiF films: examples

\textbf{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-hygrosopic**;
- **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, $\gamma$ 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.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nearest neighbour distance (Å)</td>
<td>2.013</td>
</tr>
<tr>
<td>Melting point (℃)</td>
<td>848.2</td>
</tr>
<tr>
<td>Density (g/cm³)</td>
<td>2.640</td>
</tr>
<tr>
<td>Molecular weight</td>
<td>25.939</td>
</tr>
<tr>
<td>Refractive index @ 640 nm, RT</td>
<td>1.3912</td>
</tr>
<tr>
<td>Solubility (g/100g H₂O @ 25℃)</td>
<td>0.134</td>
</tr>
<tr>
<td>Hardness (Knoop 600 g indenter)</td>
<td>102</td>
</tr>
<tr>
<td>Transmission range (μm)</td>
<td>0.12 - 7</td>
</tr>
</tbody>
</table>
**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.
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
- the deposition parameters: \( T_s, t, R \)

Deposition parameters:

- Pressure < 10⁻⁶ mbar
- Evaporation rate \( R = 0.5 - 2 \text{ nm/s} \)
- Total film thickness \( t = 0.2 - 6 \mu\text{m} \)
- Substrate temperature \( T_s = 30 - 350°C \)

θ-20 diffraction patterns of LiF films grown on glass at \( T_s = 30°C (LT) \) and \( 300°C (HT) \) with two different \( t \).
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.
RT photoluminescence spectra of colored LiF crystals vs dose

Argon laser excitation at $\lambda_p = 458$ nm

Energy (eV)

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

Cu-K$_{\alpha}$ = 8.042 keV
Ni-K$_{\alpha}$ = 8.333 keV
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$_2$SO$_4$ 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.

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 $K_{\alpha}$ 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.