ELSEVIER

Spectrochimica Acta Part B 62 (2007) 631 – 635

SPECTROCHIMICA
ACTA
PART B

www.elsevier.com/locate/sab

Soft X-ray imaging by optically stimulated luminescence from color centers in lithium fluoride ☆

F. Bonfigli [a,*], S. Almaviva [a], G. Baldacchini [a], S. Bollanti [a], F. Flora [a], A. Lai [a], R.M. Montereali [a], E. Nichelatti [b], G. Tomassetti [c], A. Ritucci [c], L. Reale [c], A. Ya. Faenov [d], T.A. Pikuz [d], R. Larciprete [e], L. Gregoratti [f], M. Kiskinova [f]

[a] *ENEA, C.R. Frascati, Via E. Fermi, 45, 00044 Frascati (Rome), Italy*
[b] *ENEA, C.R. Casaccia, Via Anguillarese 301, 00060 S.Maria di Galeria (Rome), Italy*
[c] *Università de L'Aquila e INFN, Dip. di Fisica, Coppito, L'Aquila, Italy*
[d] *MISDC of VNIIFTRI Mendeleevo, Moscow region, 141570, Russia*
[e] *ISC-CNR, Sezione Montelibretti, Via Salaria, Km. 29.3, 00016 Monterotondo Scalo (Rome), Italy*
[f] *Sincrotrone Trieste, S. S. 14, Km. 163.5, 34012 Basovizza (TS), Italy*

Received 21 November 2005; accepted 20 February 2007
Available online 2 March 2007

Abstract

An innovative X-ray imaging detector based on Optically Stimulated Luminescence from color centers in lithium fluoride is presented. Regular photoluminescent patterns produced on LiF samples by different intense X-ray sources, like synchrotrons, laser plasma sources and a capillary discharge laser have been investigated by a Confocal Laser Scanning Microscope. The use of a LiF-based imaging plate for X-ray microscopy is also discussed showing microradiographies of small animals.

Keywords: Lithium fluoride; Color Center; Photoluminescence; Soft X-ray; Imaging detector

---

1. Introduction

The recent scientific and technological interest for soft X-ray applications in different fields makes still more attractive the development of novel detectors. Among radiation sensitive materials exhibiting active optical properties, lithium fluoride (LiF), in the form of bulk and film, is a very promising candidate for the fabrication of miniaturized light sources for innovative photonic devices. Significant results have been obtained in the manufacture and characterization of integrated active optical devices based on color centers (CCs) in LiF, mainly produced by using low-energy electron-beam lithography, like active waveguides [1], microcavities [2], point light sources [3,4] and optical memories [5]. The aggregate CCs are characterized by broad absorption and emission bands in the visible spectral range. Particular attention has been devoted to F_3^+ and F_2 defects, which consist of two electrons bound to two and three close anion vacancies, respectively. These centers have almost overlapping absorption bands (M band) centered at about 450 nm [6] and, therefore, can be simultaneously excited with a single laser pump wavelength. On the other hand, they exhibit two different broad emission bands in the green (F_3^+) and red (F_2) spectral ranges.

The formation of CCs induces an increase in the refractive index of the colored volume with respect to that of an uncolored one, in the same spectral interval where the defects photoemissions are located. So it is possible to create an active waveguiding region, or other devices, that exhibit a periodic modification of the refractive index and of the gain, such as a Distributed Feedback

---

☆ This paper was presented at the "18th International Congress on X-ray Optics and Microanalysis" (ICXOM-18) held in Frascati, Rome (Italy), 25-30 September 2005, and is published in the Special Issue of Spectrochimica Acta Part B, dedicated to that conference".

* Corresponding author. Tel.: +39 06 9400 5567; fax: +39 06 9400 5400.
E-mail address: bonfigli@frascati.enea.it (F. Bonfigli).

0584-8547/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.sab.2007.02.016

(DFB) laser. Very recently simultaneous fabrication of laser-active F_2 and F_3^+ color centers in LiF and permanent periodic gratings with fringe spacings as fine as sub-micron size has been obtained by two interfering infrared femtosecond laser pulses [7]. As far as the techniques are concerned, the increasing demand for low-dimensionality photonic devices imposes the utilization of advanced lithographic systems for producing optical structures with submicrometric spatial resolution.

In LiF stable CCs production is possible only by ionizing radiation, like neutral and charged particles (neutrons, electrons and ions), gamma and X-rays, and UV light. By using electron beam irradiation, the spatial resolution of the colored volume is limited by beam enlargement due to charge effects in insulating materials and also by lateral spreading of the electrons due to scattering processes in the interaction volume. On the other hand, highly penetrating radiation, like gamma and X-rays, are not suitable to color thin layers with controlled depth and in producing luminescent patterns with high spatial resolution (the resolution is limited by the photoelectrons blurring, as well known in the proximity X-ray lithography experiments [8]). The low penetration, the short wavelength and neutrality of soft X-rays ($0.3 < h\nu < 8$ keV) and EUV light ($20 < h\nu < 300$ eV) [8] make both types of electromagnetic radiation particularly attractive for localized surface coloration [9] of LiF single crystals and thin films.

Regular photoluminescent patterns based on the stable generation of CCs in LiF samples obtained by different irradiation techniques and by several soft X-ray sources will be presented. The simplicity and the efficiency of the Optically Stimulated Luminescence (OSL) reading technique allows for an extremely large field of view without limiting the final detectable resolution if advanced optical microscopes (like Confocal Laser Scanning Microscope "CLSM" or Scanning Near Field Optical Microscope "SNOM") are used. Luminescent structures on LiF have been investigated by a CLSM, achieving high spatial resolution images.

Recently, the idea of using a LiF film as imaging plate for X-ray microscopy and micro-radiography based on OSL from CCs has been proposed [10] and developed [11] and an example will be presented and discussed.

2. Experimental details on irradiation sources

For testing LiF crystals and films as an imaging detector, as well as for photonic device developments, permanent photoluminescent structures were generated by several intense soft X-rays sources using different lithographic irradiation techniques.

A laser plasma source, obtained by focusing a powerful excimer laser beam on a solid target and developed at C.R. ENEA Frascati [12], has been used to irradiate LiF material by masking the incoming radiation [13]. The X-ray emission of this source covers the spectral interval of 0.8–60 nm (1.5 keV–20 eV), that is the full EUV region and part of the soft X-rays region. The emission spectral distribution can be adjusted by changing the laser parameters and the target material. The typical repetition rate is 1–10 Hz and the pulse duration is 10, 30 or 120 ns depending on the characteristic of the selected excimer laser. As the plasma source emits X-rays in a 2π solid angle following roughly a $\cos(\theta)$ angular distribution, it is possible to irradiate the whole surface of the sample placed in its proximity. A significant advantage of this technique is that a large area, up to 4×4 cm^2 for a typical distance of LiF from the source of 10 cm, can be uniformly irradiated in a single shot.

Direct writing of fluorescent patterns on LiF films has been obtained by using a focused X-ray beam provided by the ELETTRA synchrotron facility of Trieste (ESCA Microscopy beamline) [14]. Photoluminescent patterns of CCs in LiF have been obtained by scanning the LiF specimen with respect to the X-ray microprobe [15]. The X-ray source is optimized for the energy range (400–700) eV and can be spatially limited by a pinhole ≈ 80 μm wide, or can be demagnified to a microspot with a diameter of ≈ 100 nm by using a zone plate focusing optic, which has the advantage to keep the beam size constant, independently from the chosen energy. In the first operation mode the photon flux is $\approx 5 \times 10^{14}$ photons/cm^2 s and rises to 2×10^{18} photons/cm^2 s in the microprobe configuration, high enough to allow local creation of CCs using short exposure times between 1 and 100 ms. The LiF specimens were mounted on the ESCA positioning and scanning stage, equipped with stepper (accuracy 1 μm) and piezo-driven (accuracy 10 nm) motors, controlled by a computer program. By moving the LiF specimens

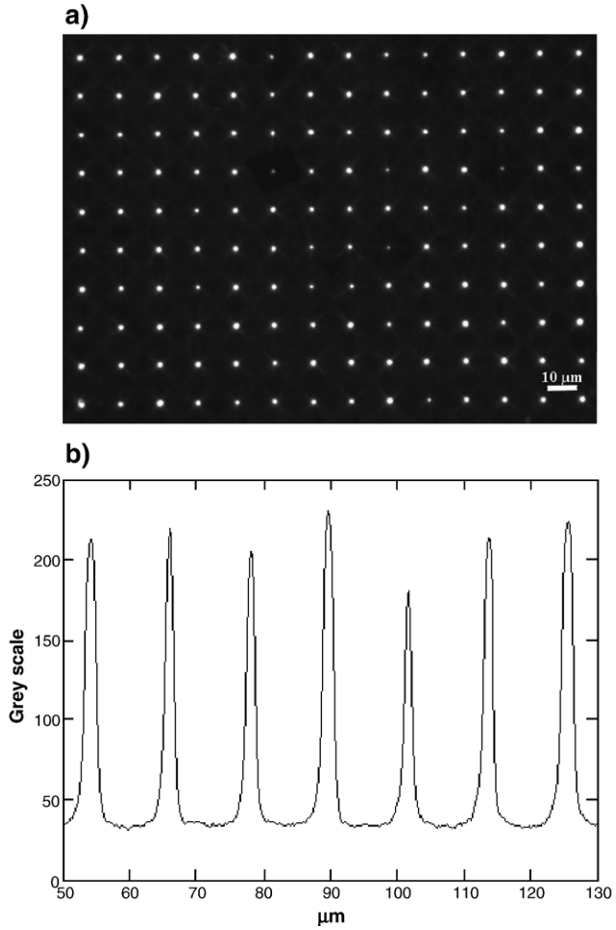

Fig. 1. a) CLSM optical image in fluorescence mode of a periodic photoluminescent structures at the surface of a LiF crystal, subsequent to irradiation by a laser plasma soft X-rays by masking the LiF sample. b) Gray-level scan of an irradiated zone.