Radiation Measurements 90 (2016) 188–191

Contents lists available at ScienceDirect

Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeas

Lithium fluoride colour centres-based imaging detectors for proton beam characterization at high doses

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HIGHLIGHTS

- Photoluminescence of colour centres in LiF can be used for 2-D proton beam imaging.
- Photoluminescence response of colour centres in LiF films is linear over several orders of magnitude of proton dose range.
- Principal Component Analysis was applied to optical absorption spectra to highlight defect formation with dose in LiF.
- LiF crystals and thin films are promising for proton beam characterization/dosimetry by photoluminescence at high doses.

ARTICLE INFO

Article history:
Received 22 October 2015
Received in revised form
23 December 2015
Accepted 30 December 2015
Available online 4 January 2016

Keywords:
Colour centres
Photoluminescence
Proton beams
Thin films
Lithium fluoride

ABSTRACT

Proton beams of 7 MeV energy, produced by a linear accelerator, were used to irradiate LiF crystals and thin films thermally evaporated on glass substrates in the dose range from 10^3 to 4×10^6 Gy, inducing the formation of stable photoluminescent colour centres (mainly F_2 and F_3^+), emitting in the visible spectral range. Using a conventional fluorescence microscope, the transversal proton beam intensity was mapped by acquiring the photoluminescence image of the irradiated spots. Image analysis allowed measuring the integrated photoluminescence intensity as a function of the irradiation dose: a linear optical response was obtained up to different maximum dose values, after which a quenching was observed, depending on the nature of the samples (crystals or films). The colour centres formation was investigated by optical absorption spectroscopy at room temperature and the Principal Component Analysis was applied to the absorption spectra of irradiated LiF crystals. In samples irradiated at highest doses, it allowed clearly identifying the formation of more complex aggregate defects, which appears strictly related to the observed photoluminescence quenching effect.

1. Introduction

Colour centres (CCs) in lithium fluoride (LiF) crystals and thin films found application in optically pumped solid-state lasers (Basiev et al., 1988) and miniaturized light-emitting devices (Montereali, 2002), due to the high efficiency of their photoluminescence (PL) process. Recently, LiF crystals and thin films have been also proposed as novel solid-state soft x-ray and neutron imaging detectors (Baldacchini, 2005; Matsubayashi et al., 2010), based on the optical reading of the visible PL of the radiation-induced F_2 and F_3^+ (two electrons bound to two and three close anion vacancies, respectively) CCs, stable at room temperature (RT) (Montereali, 2002; Perez et al., 1990). On the other hand, ion beams of different energies are widely investigated for applications ranging from material modifications (Abu-Hassan and Townsend, 1986; Perez et al., 1990) to radiobiology and radiotherapy. With the aim to use PL of point defects in LiF for proton beam monitoring and dosimetry, we recently started the investigation of the optical properties of stable CCs induced by proton beams in LiF crystals and thin films (Piccinini et al., 2014a,2014b,2015).

Proton beams of 7 MeV energy, produced by the injector of a proton therapy linear accelerator, under development at ENEA Frascati (Ronsivalle et al., 2011), were used to irradiate in air RT LiF crystals and polycrystalline thin films grown by thermal evaporation on glass substrates (Montereali, 2002). The irradiation dose range was from 10^3 to 4×10^6 Gy. Proton irradiation induced the

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http://dx.doi.org/10.1016/j.radmeas.2015.12.045

stable formation of primary (F) and aggregate (mainly F_2 and F_3^+) CCs. These laser-active defects possess almost overlapping absorption bands peaked around 450 nm, called M band (Nahum and Wiegand, 1967), and, under light excitation in this spectral range, they emit broad PL bands peaked at 678 nm and 541 nm for F_2 and F_3^+ CCs, respectively (Abu-Hassan and Townsend, 1986; Baldacchini et al., 2000).

Using a conventional fluorescence microscope, the transversal proton beam intensity, stored in the exposed LiF samples, was mapped by acquiring the photoluminescence image of the irradiated spots and image analysis allowed measuring the integrated photoluminescence intensity as a function of the irradiation dose. CCs formation was also investigated by optical absorption spectroscopy and the Principal Component Analysis (PCA) was applied to the absorption spectra of irradiated crystals.

2. Materials and methods

Exposed samples were $10 \times 10 \times 1$ mm^3 LiF crystals polished on both faces, commercially available (MacroOptica Ltd.), and polycrystalline LiF thin films, about 1 μm thick, grown by thermal evaporation on glass substrates (Montereali et al., 1991) kept at a constant temperature of 300 °C during the deposition process, at an evaporation rate of 1 nm/s in a vacuum chamber at a pressure below 1 mPa, at the Solid State Lasers Laboratory in ENEA Frascati. The starting material consists of LiF microcrystalline powder (Merck Suprapur, 99.99% pure), heated at about 800 °C in a water-cooled tantalum crucible.

Proton beams of 7 MeV energy were produced by the PL7 model LINAC by ACCSYS-HITACHI. At the output of the machine beamline a 50 μm thick kapton window was placed, which reduced the impinging protons energy to 6.7 MeV. LiF samples were irradiated in air at RT. The beam diameter was about 3 mm with an average current of 1 μA in 60 μs-long pulses at a repetition frequency of 50 Hz. Different doses were delivered to each LiF sample by varying the total number of beam pulses.

The PL images of proton irradiated spots were acquired by a fluorescence microscope Nikon Eclipse 80-i C1, equipped with a Hg lamp and a 4x objective. The blue emission of the Hg lamp, peaking at 434 nm, was selected in order to simultaneously excite the PL of the F_3^+ and F_2 CCs and an Andor Neo s-CMOS camera was used to acquire the PL images with an 11-bit dynamic range. As the PL intensities of the coloured spots were different, depending on the used proton beam fluences, the exposure time of the camera was adjusted to exploit as much as possible all the dynamic range. After selecting the irradiated spot area in the image, the integrated intensity of the F_3^+ and F_2 CCs PL signal was measured by using the "integrated density" function of ImageJ software, the measured integrated PL signal was then normalised to an exposure time of 1 s. For each dose value only one sample was irradiated and the corresponding measured PL signal was determined with an uncertainty less than 2%, due to the good signal-to-noise ratio of the images.

Optical absorption spectra were acquired at RT on the irradiated areas of LiF crystals by a Perkin–Elmer Lambda 950 spectrophotometer with 1 nm resolution in the spectral range from 200 to 1200 nm.

3. Results and discussion

Fig. 1 shows the visible PL image, acquired with the fluorescence microscope, of the two-dimensional intensity distribution of the 7 MeV proton beam stored by stable CCs formation in a LiF crystal irradiated at a dose of 10^4 Gy. It shows LiF detectors are able to store information about the proton beam intensity with a high spatial resolution and revealing even subtle intensity differences. The 11-bit camera used to acquire the image is likely to be adequate to show with an optimal greyscale accuracy all the information stored in LiF.

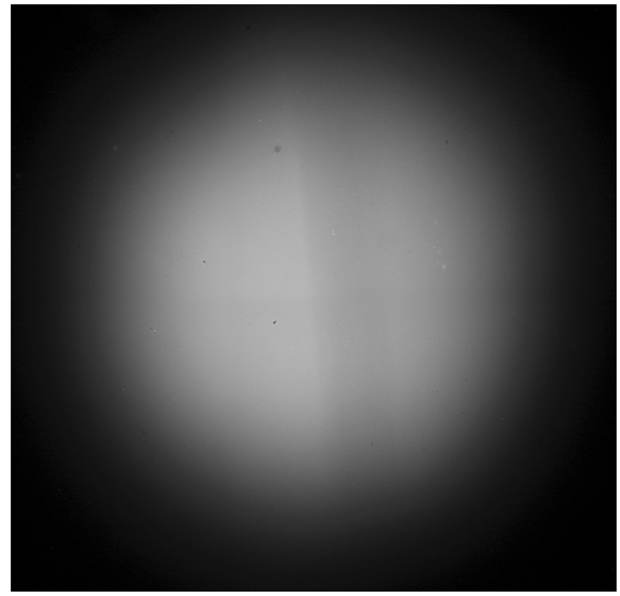

Fig. 1. Photoluminescence image of the 7 MeV proton beam transversal section stored by colour centres in a LiF crystal at a dose of 10^4 Gy.

Using the optical images acquired at the fluorescence microscope, the integrated PL signal was obtained by image analysis with ImageJ software. Fig. 2 shows the integrated CCs PL signal as a function of the dose of 7 MeV protons in LiF crystals and thin films. In LiF crystals, it increases with dose and shows a linear behaviour up to 2.5×10^4 Gy, while at higher doses saturation effects take place. Also in LiF films the PL signal is proportional to the dose values, but the linear optical response reaches the highest dose value at 5×10^5 Gy, after which it turns into sub-linear. Fig. 2 shows also the linear fit of both PL curves highlighting the linear PL response ranges. According to Fig. 2, in the dose interval where both crystals and films show a linear optical response, the

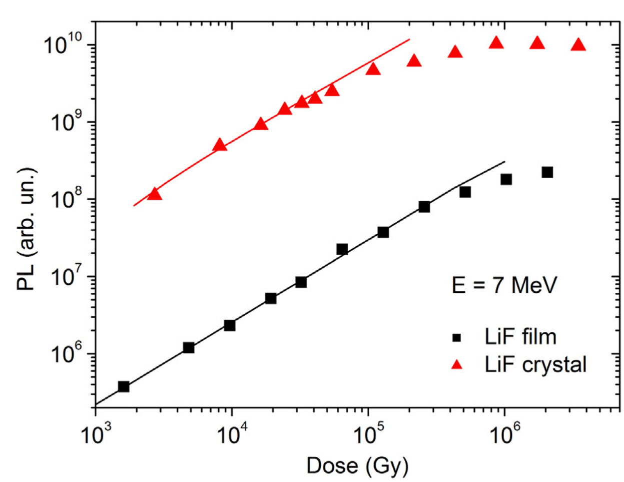

Fig. 2. Integrated visible photoluminescence signal of F_2 and F_3^+ colour centres as a function of the 7 MeV proton dose in colored LiF crystals and 1 μm thick films grown on a glass substrate. The solid lines show the best fit of both PL curves highlighting the linear PL response ranges.