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Solid state detectors based on point defects in lithium fluoride for advanced proton beam diagnostics

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ABSTRACT

Proton beams of 3 and 7 MeV energies, produced by a linear accelerator, were used to irradiate lithium fluoride crystals and thermally evaporated LiF thin films in the fluence range of 10^{11}–10^{15} protons/cm^2. The irradiation induces the formation of stable colour centres, mainly the primary F centre and the aggregate F_2 and F_3^+ defects. By optical pumping in the blue spectral region, the F_2 and F_3^+ centres emit broad photoluminescence bands in the visible spectral range. By conventional fluorescence microscopy, the integrated photoluminescence intensity was carefully measured in LiF crystals and thin films as a function of the irradiation fluence: a linear optical response was obtained in a large range of fluence, which is dependent on the used LiF samples and the selected beam energy. It was possible to record the transversal proton beam intensity profile by acquiring the photoluminescence image of the irradiated spots on LiF films by a standard optical microscope. Using LiF films grown on silicon substrates irradiated in a particular geometry, the same optical reading microscopy technique allowed one to measure the distribution of colour centres photoluminescence along the depth and direct imaging the Bragg peak position, which gives a rough estimation of the initial proton beam energy.

1. Introduction

Point defects in insulating materials are widely used for radiation detectors and dosimetry [1]. Among them, colour centres (CCs) in lithium fluoride, LiF, are well known for applications in light-emitting miniaturised devices [2–5] and tuneable solid-state lasers [6]. Various kinds of ionising radiations generate stable primary and aggregate defects in LiF crystals [7] and thin films [8]. The simplest defect that can be created is the so-called F centre, which is formed when a negatively charged ion is dislocated in the crystal lattice, leaving an electron trapped in a positively charged vacancy and its photoluminescence was not unambiguously observed up to now. Neutral and charged aggregates of F centres have characteristic broad absorption bands located in the visible spectral region and several of them exhibit intense Stokes-shifted photoluminescence (PL) at room temperature (RT) by optical pumping in the same spectral interval [6,7]. In particular, the laser-active F_3^+ and F_2 CCs (two electrons bound to three and two anion vacancies, respectively) have the peculiarity to possess almost overlapped absorption bands, at around 450 nm, generally called M band [9]. Under blue light excitation in this spectral range, they simultaneously emit green (F_3^+) and red (F_2) luminescence, peaking at about 540 and 680 nm, respectively [9,10].

In the last years, photoluminescence of these radiation-induced CCs in LiF crystals and thin films was proposed [11] for the development of novel high-spatial resolution solid-state soft X-ray imaging detectors, successfully tested also at different photon energies [12]. Their optical emission properties under light excitation were investigated in pure and doped LiF materials in different forms [13–17] for application in radiation dosimetry.

On the other hand, ion beams of different energies are also widely investigated for applications ranging from material modifications [12,18,19] to radiobiology and radiotherapy. The use of low penetrating elementary particles is particularly interesting in LiF because it makes possible to prepare thin coloured layers to depths of a few micrometres with high concentration of defects located close to the surface of the crystalline material [3,8,20,21]. The energy is delivered via a complex mixture of processes, which are generally separated out as those involving electronic excitation and, at low energies, nuclear stopping regime.

Very recently we started the investigation of the optical absorption and emission properties of CCs induced in LiF crystals and thin films by low energy (3 MeV) protons [22]. In this paper we present experimental results about the stable formation of CCs in LiF crystals and thin films, grown by thermal evaporation, irradiated by 3 and 7 MeV protons in a large interval of beam fluences.

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By conventional optical fluorescence microscopy, the integrated F_3^+ and F_2 PL intensity was carefully measured in LiF crystals as well as in thin films as a function of the irradiation fluence. The formation of the aggregate F_3^+ and F_2 electronic defects was investigated also by photoluminescence spectroscopy. Using LiF films grown on silicon substrates irradiated in a particular geometry, the CCs fluorescence microscopy distribution provides a direct imaging of the Bragg peak position, which gives a rough estimation of the initial proton beam energy.

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 [23] and Si(100) substrates [24] kept at a constant temperature of 300 °C during the deposition process, performed in a vacuum chamber at a pressure below 1 mPa, at the Solid State Lasers Laboratory in ENEA C.R 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. The evaporation rate, monitored in situ by an INFICON quartz oscillator, was automatically controlled at a fixed value of 1 nm/s during the growth.

Proton beams of 3 and 7 MeV energy were produced by a linear accelerator (PL7 model by ACCSYS-HITACHI) working as the injector of the prototype of a protontherapy linac under development at ENEA C.R. Frascati [25]. A 50 μm thick kapton window was placed at the output of the machine beamline. The LiF samples were irradiated in air at a distance of 10 mm from this exit window.

During proton irradiation at RT, LiF crystals and LiF films on glass were fixed on an aluminium mask with a 3 mm pin-hole, in order to irradiate them on circular spots with the highest and most uniform transversal intensity distribution of the proton beam. The average beam current was 1 μA in 60 μs-long pulses at a repetition frequency of 50 Hz. The irradiation fluence covered the range of 10^{11}–10^{15} protons/cm^2 by varying the total number of pulses delivered to different LiF samples.

The integrated intensity of the F_3^+ and F_2 CCs PL signal was measured by a fluorescence microscope Nikon Eclipse 80-i C1, equipped with a Hg lamp and a 4× objective (N.A.=0.10). The blue emission of the Hg lamp, peaking at 434 nm, was selected in

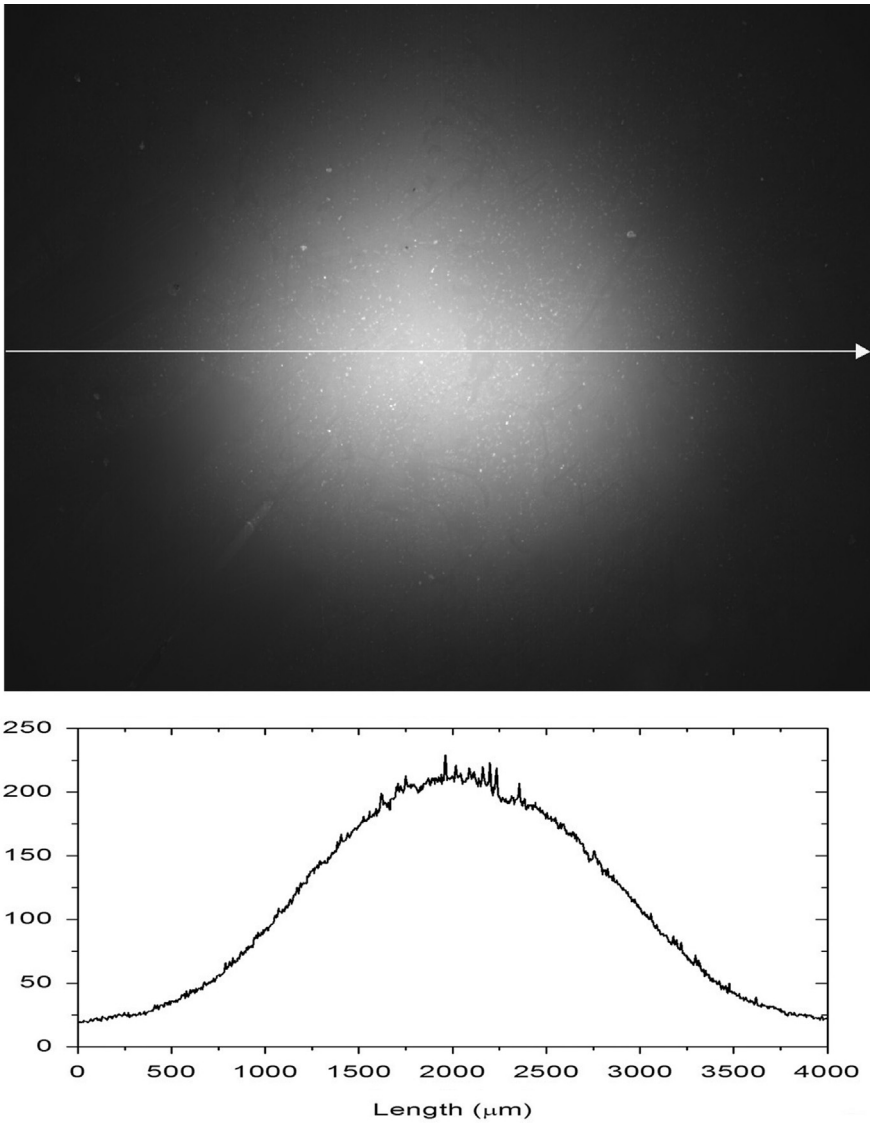

Fig. 1. (Top) Photoluminescence image of the 3 MeV proton beam transversal section stored by colour centres in a 1 μm thick LiF film grown on a glass substrate at a fluence of 5.0×10^{12} protons/cm^2. (Bottom) Intensity profile along the arrow shown in the upper image.