



Damage creation in lithium fluoride by Swift heavy ions in the electronic energy near the maximum of the Bragg's peak

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ABSTRACT

Single crystal samples of LiF were irradiated at room temperature with different energy of 0.7- 4.1 MeV/u Pb⁺⁵³ ions in fluence range from 10⁹ and 4.10¹² ions/cm². are characterized by optical absorption and photoluminescence (PL) techniques were used for characterisation of the irradiated samples. The damage is dominated by the creation of simple defects (F centers) and aggregated defects (F_n centers). Their concentration increase of a function of fluence and energy. On the other hand two very intense PL bands observed at around 545 and 665 nm which correspond to the emission of F₃⁺ and F₂ centers respectively and increase linearly in the same fluence range.

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KEYWORDS

LiF;
Swift heavy;
Ion irradiation;
Optical absorption;
PL.

INTRODUCTION

Defects in alkali halides under various types of irradiations e.g. neutrons^[10], photons^[10], electrons^[10], and ions^[10] are studied extensively during the past few decades. Lithium fluoride is an ionic crystal and an insulator with high band gap (14 eV), its transmission spectrum extends from about 190 nm to 1200 nm. The LiF is an important crystal used for a number of high technology, optical and electro-optical applications, like radiation dosimetry, detectors for ionizing radiations. The optical properties of this material can be modified by point and extended defects, created due to different types of particle irradiation. The defect centers induced by ion irradiation in LiF are mainly F center, and more complex defects F-aggregate centers such as F₂, F₃ and F₄. These centers have been identified from various experimental techniques like optical absorption lu-

minescence, small angle X diffusion and electron spin resonance^[10].

EXPERIMENTAL PROCEDURE

Pure monocrystal of LiF, Their thickness is 0.5 – 1.5 mm and always exceeds the ion penetration depth, were cleaved along the (100) plane. Samples are irradiated at GANIL (Caen, France) on the medium energy beam line SME. The LiF were exposed to ²⁰⁸Pb ions of 936 MeV energy at room temperature under normal incidence, and the fluence extended from 10⁹ to 4. 10¹² ions.cm⁻² and the flux was about 4.10⁸ ions cm⁻²s⁻¹ on a 1 cm² surface. Thin aluminum foils of different thicknesses (12, 24, and 30µm) were placed in front of each sample in order to modify the initial energy of the ions (and consequently the range R, and (dE/dx)_e). The main irradiation parameters were deduced from TRIM

TABLE 1 : Irradiation parameters of Pb in LiF crystals.

| Ion | E(MeV) 208 Pb ⁺⁵³ | (dE/dx) _e (keV/nm) | (dE/dx) _n (keV/nm) | Range (μm) | E/R (keV/nm) | Fluence Ions/cm ² |
|-----|------------------------------------|-------------------------------|-------------------------------|------------|--------------|--------------------------------------|
| | 840 | 27 | 0.05 | 40 | 21 | 10 ⁹ - 4.10 ¹² |
| | 544 | 27.5 | 0.075 | 29 | 18.7 | 10 ⁹ - 4.10 ¹² |

TABLE 2 : Absorption bands of various electron color centers in LiF.

| Center | Absorption | | Emission | |
|-----------------------------|-------------|-----------------|-------------|-----------------|
| | Energy (eV) | Wavelength (nm) | Energy (eV) | Wavelength (nm) |
| F | 5.00 | 245 | | |
| F ₃ | 3.92, 3.32 | 315, 375 | | |
| F ₂ | 2.79 | 444 | 1.85 | 670 |
| F ₃ ⁺ | 2.77 | 448 | 2.35 | 528 |
| F ₂ ⁻ | 1.29 | 960 | 1.17 | 1060 |
| F ₄ | 2.39 2.30 | 515, 545 | | |
| F ₂ ⁺ | 1.97 | 630 | 1.39 | 910 |
| F ₂ ⁻ | 1.29 | 960 | 1.1 | 1120 |

2006^[12] code calculations, are listed in TABLE 1.

Optical absorption and photoluminescence measurements are carried out using a SHIMDZU 1700 spectrometer and Perkin Elmer LS50B Luminescence Spectrometer respectively. The absorption spectra are recorded in the wavelength 190 - 1100 nm, the luminescences are analyzed, in the 200-800 nm range. With optical absorption spectroscopy technique it is possible to obtain information about the generated defects in the crystal by ion irradiation, and to calculate the concentration of defects. The concentration of defects can be determined by the Smakula formula, modified by Dexter^[13]:

$$N (\text{cm}^{-3}) = A \cdot \frac{n}{(n^2 + 2)^2} \frac{W}{f} \alpha_{\text{max}} \quad (1)$$

Where f is the oscillator strength of the optical transition, n the refractive index, α_{max} the absorption coefficient measured at the maximum of the band peak and W the band's full-width at half maximum (FWHM). A is a constant, which takes $0.87 \times 10^{17} \text{ eV}^{-1} \text{ cm}^{-2}$ for Gaussian bands. The absorption coefficient α_{max} was determined from the optical spectra of each simple using the relation:

$$\alpha_{\text{max}} = 2.304 \frac{\text{OD}}{R} \quad (2)$$

where: OD represents the optical density at band maximum and R is the ion range.

In order to compare the evolution of F and F₂ ag-

gregate centers per track as a function of the fluence at the different energies, the number per single track of F center can be determined by:

$n^s = \frac{n_F}{\Phi}$ ^[14], where n_F is given in cm⁻². Since the different bands in the 300–600 nm region strongly overlap and it is difficult to analyze individual contributions separately due to the unknown oscillator strength of the different centers. The creation of all F₂ centers has been estimated from the parameter a^s which is deduced from the integrated absorption:

$$S = \int_{300}^{600} \text{OD} \, d\lambda \quad (3)$$

normalised by the ion fluence $a^s = \frac{S}{\Phi}$ (4)

Where: a^s the integral absorption per ion in units of nm³.

The ions having an energy of the order of hundreds MeV create in alkali halides various types of defects. In LiF, defects corresponding to the different absorption and emission peaks are well known, the main ones are summarized in TABLE 2 according to refs.3,25, and 28.

The detection of these centers is relatively easy, the only difficulty in the case of center, F₃⁺, resides in the fact that the absorption band is coincident with the center F₂⁻, always more intense: the F₃⁺ center must always be observed by their emission.

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RESULTS AND DISCUSSION

Optical absorption spectroscopy

Low ion fluence

Figure 1 and Figure 2 shows the optical absorption spectra obtained with LiF crystals irradiated with 840 MeV and 544 MeV Pb ions at low fluence. We observed no modification of the position of the absorption band during irradiation. The spectra show predominant absorption bands around 245 nm. This band corresponds to F-center (halogen-ion vacancy with a trapped electron). A threshold of creating F_2 centers is observed for the energy 544 MeV and a fluence above $4 \cdot 10^{10}$ ions/cm².



For fluencies higher than 10^{10} ions/cm² and the energy 840 MeV, one can clearly observe the creating of F_2 and F_3 centers. can be modalized by the reaction:

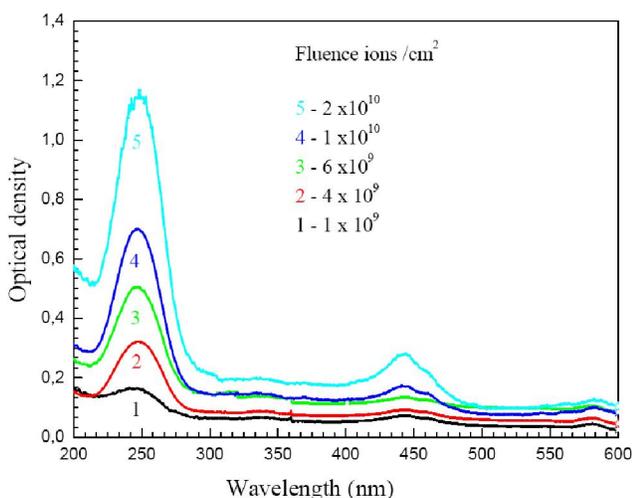
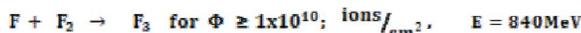


Figure 1 : Optical absorption spectra of LiF irradiated at low - fluence With Pb ions 840-MeV of various fluencies

High ion fluence

Figure 3 and Figure 4 shows the optical absorption spectrum for different fluencies of 840 MeV and 544 MeV lead ion irradiation of LiF crystal. One sees quite clearly the absorption bands shows two predominant bands centered at 245 and 445 nm corresponding to F and F_2 centers (two electrons trapped in two neighboring anion vacancies) respectively. Also at higher fluence, of $2 \cdot 10^{11}$ ions/cm² the spectra become more complex due to track overlapping, F_3 -center bands centered at

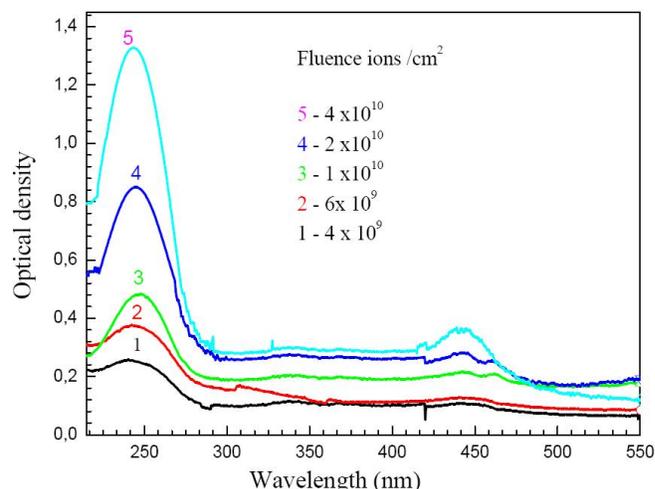


Figure 2 : Optical absorption spectra of LiF irradiated at low - fluence With Pb ions 544-MeV of various fluencies.

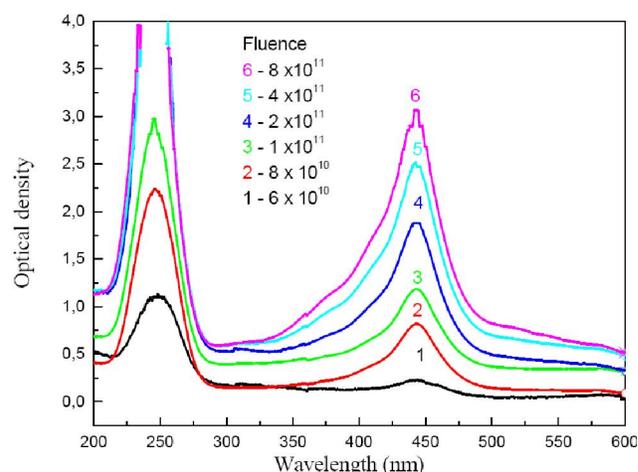


Figure 3 : Optical absorption spectra of LiF irradiated at high - fluence With Pb ions 840-MeV of various fluencies.

315 and 375 nm, and two weak bands (F_4 – center) centered at 515 and 545 nm. But the absorption band around 445 nm is a result of the overlapped band corresponding to the F_2 and F_3^+ (two electrons trapped in a three neighboring anion vacancies).

For a quantitative comparison, the saturation of optical density of the band F-center for a higher fluence 4×10^{11} ions/cm² and energy 5440 MeV. for energy 840 MeV the saturation of the optical density is reached at a value of 1×10^{11} ions/cm².

The dependences of n_f^s and a^s on the irradiation fluence and with different energies are displayed in Figure 5 and Figure 6. In all cases the number of n_f^s and per ion as a function of the fluence take the same evolution. At energies 4.1 MeV/u and 2.6 MeV/u the num-

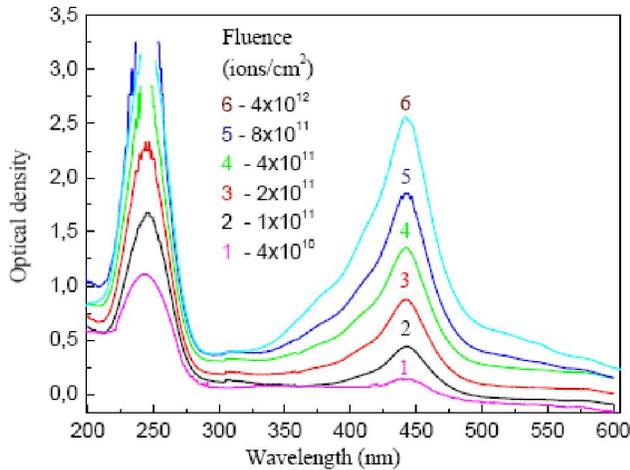


Figure 4 : Optical absorption spectra of LiF irradiated at high-fluence with Pb ions 544-MeV of various fluences.

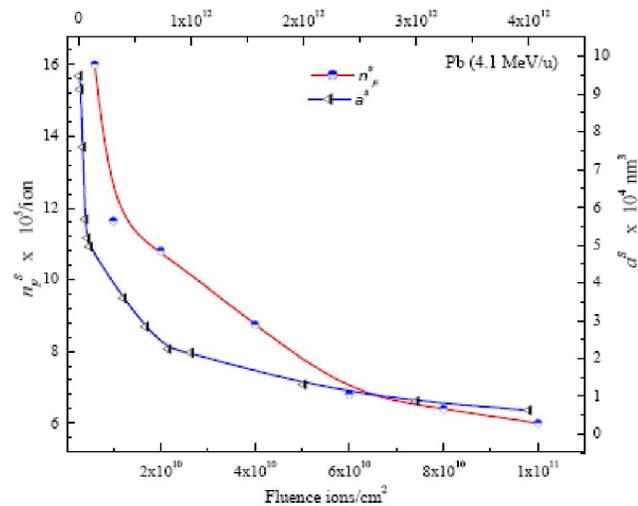


Figure 5 : Concentration of n_F^s and aggregates (a^s) per ion track as a function of fluence for LiF crystals irradiated with Pb ions at energy 840 MeV.

ber a^s of aggregate centers color centers produced per incident ion decreases with fluence. However, the n_F^s color centers per ion increases in the first stage of the fluence, reaches a maximum and finally decreases at higher fluences. These maximum appears at fluence 6×10^{10} ions.cm⁻². The number of color centers produced per ion track decreases at higher fluences gives an indication that the recombination processes of electron and hole centers become dominant in this regime.

Comparing the results of LiF to those obtained earlier for LiF irradiated of Ni ion at 170 MeV and with Xe ion 300 MeV^[15], we observe very similar effects. Also the evolution of n_F^s and a^s as a function of fluence

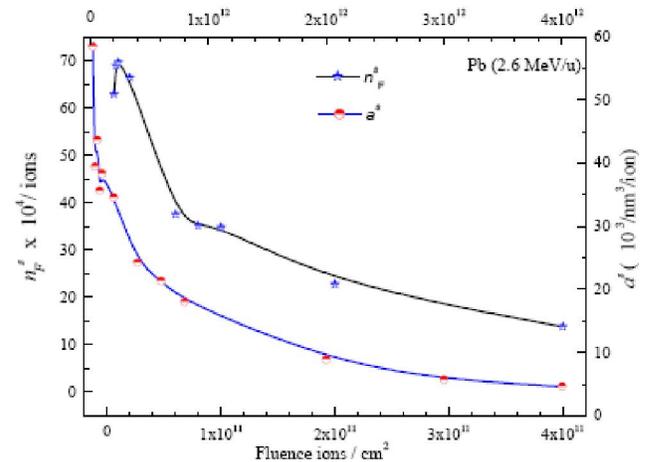


Figure 6 : Concentration of n_F^s and aggregates (a^s) per ion track as a function of fluence for LiF crystals irradiated with Pb ions at energy 544 MeV.

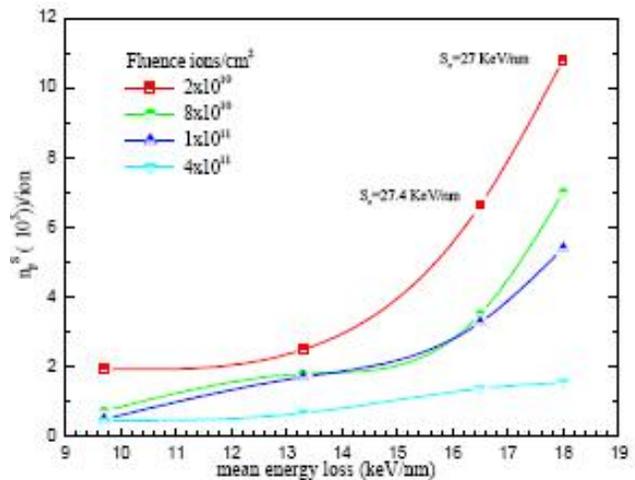


Figure 7 : Number of F centers n_F^s per ion as a function of the mean energy loss at high-fluence radiation (fluence are expressed in ions/cm²).

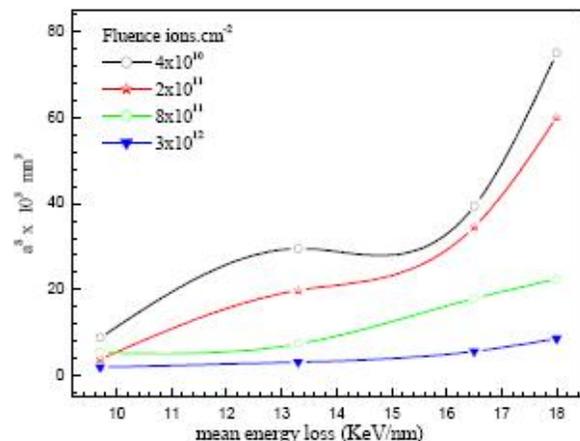


Figure 8 : Number of F_n aggregate centers a^s per ion as a function of the mean energy loss at high-fluence.

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is similar with MgF_2 [15].

In Figure 7 and Figure 8, displays F center and F_n aggregate centers per track as a function of the mean energy loss at high ion fluence. The concentration of F-center (n_f^+) and aggregates (a^s) per ion track propor-

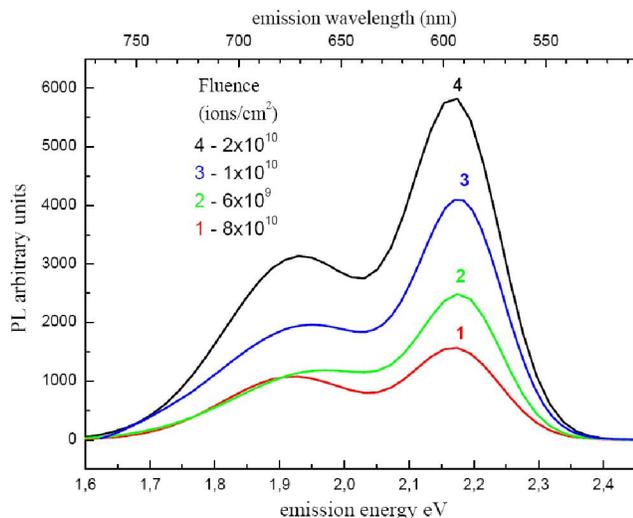


Figure 9 : Luminescence emission of F_2 and F_3^+ centers in LiF crystals irradiated with 840 MeV Pb ions of various fluences. (4.1 MeV/u Se= 27 keV/nm R=40 μ m).

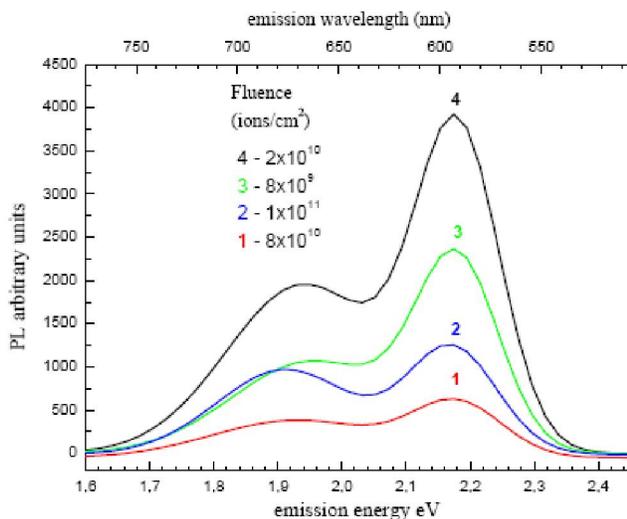


Figure 10 : Luminescence emission of F_2 and F_3^+ centers in LiF crystals irradiated with 544 MeV, Pb ions of various fluences (2.6 MeV/u Se= 27.4 keV/nm R=29 μ m).

tional to the mean energy loss, and inverse proportional to the fluence.

It should be mentioned that the number of and per ion continues to increase with incident energy. The energy 4.1 MeV/u ($S_e \sim 27$ KeV/nm) and, 2.6 MeV/u ($S_e \sim 27.4$ KeV/nm) those values situated on right and left of the maximum to Bragg's peak.

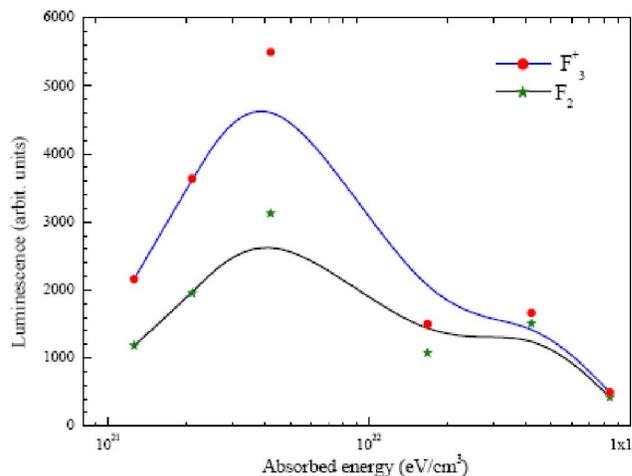


Figure 11 : Luminescence emission for F_3^+ and F_2 centers depending on the absorbed energy for LiF irradiated with Pb ions at 840 MeV.

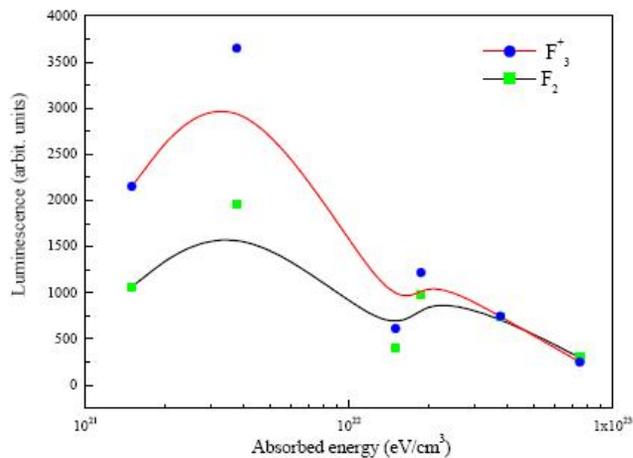


Figure 12 : Luminescence emission for F_3^+ and F_2 centers depending on the absorbed energy for LiF irradiated with Pb ions at 544 MeV

Photoluminescence measurements

In addition to optical absorption, luminescence is another technique useful tool to examine defects created by ion irradiation. It is possible to separate the presence of defects whose optical absorption bands overlap. In our case with detect the F_3^+ and F_2 centers. The emission bands of F_3^+ and F_2 centers are well resolved contrary to their absorption bands. The excitation with a 445 nm wavelength-photon induces emission bands centred at about 553 nm and 680 nm assigned to F_3^+ and F_2 centers respectively [16,17]. Figure 9 and Figure 10 shows photoluminescence spectra of SHI irradiated samples at RT of LiF single crystal irradiated with 840

MeV and 544 MeV Pb ions at different fluence.

As mentioned above the spectra show two emission bands with maxima at about 2.17 eV (570 nm) and 1.95 eV (640 nm) due to the luminescence of F_3^+ and F_2 centers^[18,19,3,19] respectively.

The peak intensities of F_3^+ , and F_2 , color centers emissions, obtained from the two Gaussian peaks, were plotted as a function of fluence for both energies the curves shown in Figure 11 and Figure 12. The intensities of both bands increase of the absorption energy and followed by an exponential decrease.

The PL intensity is sensitive to the damage created by SHI. Initially, strong PL intensity indicates dominant radiative transitions. As the concentration of color centers increases the rate of radiative transitions will increase and hence the luminescence intensity will also increase. At higher fluences the sample becomes defect rich material, which affects the radiative transitions. In the presence of defect rich material, the radiative transition rate will decrease, thus, decreasing the integrated PL intensity from the sample 13.

CONCLUSIONS

This paper reports on the damage produced in single crystals of LiF by irradiation with lead ions at different energies in the range of several hundred MeV. Single defects such as F -centers are produced in a large halo of 9–22 nm around the ion trajectory and track radii increases with the mean energy loss. Photoluminescence spectroscopy reveals that F centers aggregates (F_3^+ , F_2) increase linearly with increasing fluence mean energy loss. Increase of PL intensity of the irradiated samples indicates increase of high energy induced radiation defects in the samples.

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