HEAVY ION INDUCED ELECTRON COLOR CENTERS AND SURFACE DEFECTS OF LIF CRYSTALS

A. Dauletbekova\textsuperscript{1}, M. Zdorovets\textsuperscript{2}, A. Akilbekov\textsuperscript{1}, I. Nyshanbaeva\textsuperscript{1}, Zh. Zhanuzak\textsuperscript{2}, D. Akilbekova\textsuperscript{3}

\textsuperscript{1}L.N. Gumilyov Eurasian National University, 5 Munaitpassov str., 010008, Astana, the Republic of Kazakhstan
\textsuperscript{2}Institute of Nuclear Physics, accelerator DC-60, 2/1, Abylaikhan av., 010008, Astana, the Republic of Kazakhstan
\textsuperscript{3}Rice University, Houston, TX, USA

Abstract

The irradiation experiments at the ion accelerator DC-60 (Astan, Kazakhstan) and high current electron impulse accelerator RADAN-220 (Tomsk, Russia) are presented. Single LiF crystals were irradiated with Xe (195 MeV), Kr (117 MeV, 147 MeV), and N (18 MeV) ions. Using absorption and thermoactivation spectroscopy, color center creation was analyzed as a function of the ion energy loss, fluence, and current beam (flux). Surface macro-defects were examined in all irradiated LiF crystals by atomic force microscopy.

Keywords: lithium fluoride, track, fluence, hillock, scanning high resolution microscopy

1. Introduction

Heavy ion induced damage in LiF can be described by a nanosize core region with defect aggregates and a larger halo of several tens of nanometers with color centers [1 – 3]. The defect creation in alkali halides is determined by electronic excitations (excitons, electrons and holes) [3, 4]. Therefore, the defect creation strongly depends on the energy loss of the ions, fluence, flux and temperature.

Last achievements in atomic force microscopy (AFM) has allowed for allocating new direction in solid state physics related to the research of structural surface damages caused by heavy high – energy ions and their relation with radiation damages in the bulk. High level of electronic losses of energy, typical for such irradiation, leads to specific structural failures: structural phase transformations, formation of latent tracks in bulk and formation of macro-defects on the surface of target [5-8].

In this paper, features of damage creation in LiF crystals irradiated with heavy ions at high beam current (flux) are discussed. Color centers were examined by absorption spectroscopy in the spectral range of 6 – 1.5 eV (200 – 850 nm) and thermoactivation spectroscopy (300-750K). Atomic force microscopy (AFM) was used for macro-defect studies on the irradiated surface.

2. Experimental methods

LiF crystals (GOI, St. Petersburg, Russia) cleaved along one of the (100) plane with the thickness of 1 mm were irradiated at the ion cyclotron accelerator DC-60 (Astan, Kazakhstan) and high current electron impulse accelerator RADAN -220 (Tomsk, Russia) with pulse duration - 20ns. All experiments were carried out at room temperature with normal beam incidence. Type and energy values of used ions, fluence $\Phi$, are shown in Table 1 as well as path length $R$, values of electron $(dE/dx)_e$ and nuclear $(dE/dx)_n$, energy losses calculated by SRIM-2008 software [9].

AFM and scanning electron microscope (SEM) images of defects were obtained using JSM-5200 (Physics Technical Institute, Almaty, Kazakhstan) and JSM-7500F SEM (Institute of Nuclear Physics, Astana). Ion irradiated LiF crystal
was heated at a constant rate of $\beta = 2.86 \text{ Ks}^{-1}$ to necessary temperature and rapidly ($\sim 10$ s) cooled down to room temperature (RT) and the absorption spectrum was measured at RT.

| Table 1. Beam characteristics and ion irradiation parameters in LiF [9]. |
|-----------------------------|----------------|----------------|----------------|
| Ion                        | N              | Kr             | Kr             | Xe             |
| $E$, MeV/nucleon           | 18             | 117            | 147            | 195            |
| $(dE/dx)_c$, keV/nm        | 1.65           | 12.11          | 12.1           | 18.85          |
| $(dE/dx)_n$, keV/nm        | 0.0013         | 0.026          | 0.021          | 0.052          |
| Effective charge           | $+2$           | $+13$          | $+14$          | $+20$          |
| Range $R$, $\mu$m          | 11.0           | 15.3           | 17.5           | 17.6           |

3. Experimental results and discussions

Figures 1 shows absorption spectra of LiF crystals irradiated with Kr (147 meV) ions with different flux. All measurements were carried out at RT.

![Image](image1.png)

Figure 1. The absorption spectra of a LiF crystal irradiated with Kr (147 MeV) ions with fluence $\Phi = 6 \times 10^{12}$ ions/cm$^2$. Ion beam current (flux) equals to $1.78 \times 10^{10}$ions/cm$^2$ (blue curve), $7.13 \times 10^{10}$ions/cm$^2$ (red curve) and $8 \times 10^{10}$ions/cm$^2$ (black curve).

With increasing of flux, which is proportional to ion current, increase of single F centers and complex centers as well are noticed. Figure 2 shows absorption spectra of those crystals after the heat treatment at 315 K and 295 K. During the treatment $n_F/n_T$ increased to 0.94 for 330 K, 1.33 for 513 K and 1.59 for 593 K, this ratio was calculated using ratio of optical absorption in the absorption bands peaks of $F_2$ and $F$ centers. That is in process of heat treatment overall number of single and complex F centers are decreasing. However, number of complex F centers increases due to aggregation process of mobile F centers and due to decrease of recombination processes with hole aggregates. Presence of hole aggregates along[10] can lead to the dependence of volume concentration of F centers on $\Phi$ i.e. $N_F = \Phi^{0.5}$ for LiF irradiated with N and Xe ions (Figure 3). Such dependence was first observed and analyzed for LiF irradiated with Au ions [8]. At high fluences and high beam current the interaction of primary Frenkel pairs ($F - H$) is different from that at low excitation density. The mobile part of the Frenkel pairs is the $H$ center (a halide molecule $X_2^-$ $\equiv X^+ X^0$ replacing an anion in the lattice) [10, 11]. At high excitation density (high concentration of H centers), interaction of two $H$ centers formed a halogen molecule ($H + H \rightarrow X_2$). The di-halide molecules ($X_2$) play a crucial role at high fluence irradiation. Such molecules can not recombine with $F$ centers and an enhancement of the F centers takes place.

After ion irradiation all crystals exhibit appearance of macro-defects on a surface – hillocks [5, 7]. Number of hillocks per unit surface area is in compliance with ion fluence with 10% error. Peaks are obtained from topographic micrograph analysis. Mean values of the peak height are estimated using Gaussian fit for each height-frequency histogram and shown in Table 2.
Figure 2. The absorption spectra of a LiF crystal irradiated with 147 MeV Kr ions with fluence $\Phi = 6 \times 10^{12}$ ions/cm$^2$ at 300K. The spectra are measured at RT after the heating at 300K or additional preheating to 513 K, 593 K

Figure 3. In LiF irradiated with 18 MeV N ions at a constant fluence of $9.5 \times 10^{12}$ ions/cm$^2$ the volume concentration of F centers $N_f$ is proportional to $\Phi^{0.3}$.

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<th>Table 2. Hillocks heights for LiF irradiated with Kr, Xe, N ions</th>
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<td>Hillocks heights, $h$, nm</td>
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Hillocks height increases with increasing of energy losses. Mean roughness of nonexposed surface is 2 - 5 nm. LiF crystals exposed with electron impulses have similar defects on a surface (Figure 4).

Figure 4. AFM image of LiF surface irradiated with electron impulses with current density 300A/cm$^2$

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<th>Table 3. Hillocks heights for LiF irradiated with electron impulses</th>
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<td>Current density, A/cm$^2$</td>
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<td>$\Phi$, $10^{13}$ cm$^{-2}$</td>
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<td>Hillocks height, $h$, nm</td>
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Processes lead to the formation of hillocks exposed with ion and in our case with electron impulses irradiation are rather complex. One can assume contribution of defects in the bulk, in particular, appearance of fluorine $F_2^0$ molecules on the surface, as well as $Li^+ F_i^-$ interstitial defects formed at aggregation of H centers in the reaction shown below:

$$X^- + X^- \rightarrow \left( X^- \right)_{\text{ag}} + K^- + X^+ \ [11],$$

where $\lambda$ - halogen, $\lambda^-$ caton caused by irradiation and which form $Li^+ F_i^-$ structures serve as material for completing hillocks construction. This is indirectly verified by SEM energy dispersion analysis of the surface showing 75% of fluorine concentration on the irradiated surface.

4. Conclusions

For all ions the efficiency of color centers creation at a constant fluence and at room temperature increases with increasing of the flux. At heat treatment aggregation of F centers caused by single F centers motion and decrease of recombination processes due to aggregation of H centers is observed. At high fluence the volume concentration of F centers depends on the flux as $\varphi^{1/3}$ which can be explained by interaction of primary H centers with fluorine molecule formation. Hillocks on the surface of the irradiated LiF crystals were also observed.

References