Electron color center creation in LiF irradiated with Kr ions

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First irradiation experiments on the cyclotron ion accelerator DC-60 are presented. LiF crystals were irradiated with 147 MeV Kr ions in the fluence \( \Phi \) range \( 8 \times 10^{10} \)–\( 3 \times 10^{12} \) ions/cm\(^2\) at a beam current 20 nA/cm\(^2\) (corresponding to a flux \( \phi \approx 9 \times 10^9 \) ions/cm\(^2\) s). The main electron micro-defects (F-type color centers) were studied by absorption spectroscopy in the spectral range 200–650 nm. After a linear increase with the fluence, single F centers saturate at \( \Phi \approx 10^{12} \) ions/cm\(^2\) and at higher fluences decrease, whereas the concentration of complex centers have a strong increase. The efficiency of F center creation at higher fluences decreases due to the strong recombination losses of primary Frenkel pairs.

1 Introduction

For a long time LiF crystals have been used as optical windows for the UV and VUV spectral region and today LiF is a standard personal thermo-luminescence dosimeter for \( \gamma \)-rays, electrons, and thermal neutrons. Therefore electron–hole and exciton processes responsible for storage of the absorbed radiation energy have been studied [1].

The damage creation in alkali halides under heavy ion irradiation is similar to the conventional irradiation with X-rays or fast electrons and occurs by the exciton mechanism [2]. In alkali halide crystals, particularly in LiF, the exciton mechanism of defects creation is much more efficient than the impact one by atomic collisions [1]. Created during the decay of self-trapped excitons, Frenkel pairs consist of an anion vacancy with a captured electron (F center) and a halogen interstitial molecule (X\(_2^+\)) in the regular lattice (H center). The further transformation of these defects depends on the irradiation temperature and dose. Particularly, at room temperature F centers are stable, while the mobile H centers are transformed into V\(_3^+\) centers [3, 4]. The V\(_3^+\) center is a three-halogen molecule (X\(_3^+\)) in the anion sub-lattice with the absorption maximum in the VUV spectral region (114 nm) [4] and will be not discussed here.

Radiation defects in alkali halides under heavy ion irradiation strongly depend on the energy loss, irradiation temperature and fluence (dose). Irradiation with ions having an energy loss above a critical threshold of 6–10 keV/nm leads to a complex track structure with nanosize aggregates in the central part of the track (core) and color centers in a larger cylindrical defect zone of several tens of nanometers (halo) [5–7]. At higher fluences where neighbor tracks overlap more complex defects and defect aggregates (metal colloids, halogen molecular clusters) are created. There are several studies on defect creation under Kr ion irradiation in LiF crystals. In Ref. [7] etching along the ion tracks was investigated. In Ref. [8] the change of microhardness in LiF irradiated with 210 MeV Kr ions was compared with that by irradiation with 5 MeV electrons. The authors found that heavy ion induced microhardness change in LiF crystals is stronger than that by electron irradiation at a comparable absorbed dose. In Refs. [9, 10] the microhardness change was investigated along the ion track. It was found that the magnitude of microhardness change follows the energy loss reaching the highest value at the Bragg maximum.

Taking into account the former results of color center creation under heavy ion irradiation in LiF crystals [5–10], the main goal of our study is to investigate the creation of single and complex F centers under irradiation with 147 MeV \(^{84}\)Kr ions at high fluences.

2 Experimental results

2.1 Irradiation with Kr ions

For the experiments high quality LiF crystals were used grown from the melt in vacuum (Optical Institute (GOI), St. Petersburg, Russia). Platelets with the size of \( 10 \times 10 \) mm\(^2\) and the thickness of about 1 mm were irradiated at room temperature...
Kr ions irradiation parameters in LiF crystals [11].

<table>
<thead>
<tr>
<th>Energy (MeV) range,</th>
<th>Electronic loss</th>
<th>Nuclear loss</th>
</tr>
</thead>
<tbody>
<tr>
<td>R (μm)</td>
<td>(dE/dx)_e (keV/nm)</td>
<td>(dE/dx)_n (keV/nm)</td>
</tr>
<tr>
<td>147</td>
<td>17.5</td>
<td>12.1</td>
</tr>
</tbody>
</table>

The absorption spectra of LiF irradiated with 147 MeV Kr ions at the DC-60 cyclotron accelerator (Astana). The fluence (Φ) was varied in the range from \(8 \times 10^{10}\) to \(3 \times 10^{12}\) ions/cm\(^2\) with the beam current density of 20 nA/cm\(^2\) corresponding to the flux of about \(9 \times 10^9\) ions/cm\(^2\) s. Online temperature measurements show that at such flux magnitude the macroscopic heating of the samples did not exceed \(30^\circ\)C. The thickness for all samples was larger than the ion range.

The irradiation parameters for \(^{84}\)Kr ions in LiF crystals are presented in (Table 1).

For 147 MeV Kr ions the interaction with the LiF target occurs over electronic excitations and the collision processes (nuclear energy loss) can be neglected [6, 11].

### 2.2 Results

Optical measurements were carried out at room temperature at the spectrophotometer СФ-103 in the spectral region of 200–650 nm. The most significant peaks in the absorption spectrum are \(F\) and \(F_2\) centers, with absorption maxima at 4.94 eV (250 nm) and 2.79 eV (445 nm), respectively [6]. From the absorption spectra of irradiated LiF crystals (Fig. 1) the concentration of \(F\) and \(F_2\) centers (cm\(^{-2}\)) was estimated by the Smakula–Dexter formula [5, 6]:

\[n_F [\text{cm}^{-2}] = 9.48 \times 10^{15} D_{opt}(F),\]  
\[n_{F_2} [\text{cm}^{-2}] = 4.42 \times 10^{15} D_{opt}(F_2),\]

where \(D_{opt}(F)\) and \(D_{opt}(F_2)\) are the optical density at the absorption maxima for \(F\) and \(F_2\) centers, respectively. The average volume concentration can be estimated according to

\[N_F = n_F / R,\]

where \(R\) is the ion range. The average distance between the ion tracks (\(d_{tr} = 2r\)) in LiF crystals was estimated according to \(d_{tr} = 2(\pi\Phi)^{-1/2}\) [6].

Figure 1 presents the absorption spectra of LiF irradiated with Kr ions. The absorption band of \(F\) centers (4.94 eV) dominates in all irradiated spectra. An additional maximum at 2.79 eV is present, corresponding to \(F_2\) centers. At higher fluences additional complex \(F_n\) centers are created: \(F_3\) (with the absorption maxima at 3.91 and 3.28 eV) and \(F_4\) (with the absorption maxima at 2.39 and 2.30 eV). The concentration of these centers increases nonlinearly with the fluence (Fig. 2). The main parameters of \(F\) and \(F_2\) centers in irradiated LiF crystals are presented in Table 2. There are also data of the concentration of single \(F\) centers per track (\(n^s\)), and the average energy to create an \(F\) center (\(\Delta E_F\)). The magnitude \(n^s\) and (\(\Delta E_F\)) where estimated according to Ref. [6]:

\[n^s = \frac{n_F}{\Phi},\]

\[\Delta E_F = \frac{E_{ion} \times \Phi}{n^s}.\]

Figure 2 shows the evolution of \(n_F\) as a function of the ion fluence. The concentration of \(F\) centers in the initial stage increases linearly and reaches saturation at a fluence of \(\Phi \approx 10^{12}\) ions/cm\(^2\) and at higher fluences decreases. The decrease of \(n_F\) is accompanied with an increase of the concentration of \(F_n\) centers and the concentration of \(F_2\) centers is not more dominating. Therefore, we used the integral absorption as a parameter for the concentration of all \(F_n\) centers (\(n = 2, 3, 4\)) [6]. The integral absorption (i.e., the area under the absorption curve) in the spectral range from 4 to 2 eV (Fig. 1) versus the fluence is shown in Fig. 2. At the fluence of \(\Phi\) about \(10^{12}\) ions/cm\(^2\) saturation takes place both for single \(F\) centers and complex \(F_n\) centers (Fig. 2). At higher fluences the decrease of \(n_F\) is accompanied with a
Table 2 The main parameters of color centers in LiF crystals irradiated with 147 MeV $^{84}$Kr ions.

<table>
<thead>
<tr>
<th>$\Phi \times 10^{13}$ (ions/cm$^2$)</th>
<th>$n_F \times 10^{15}$ (cm$^{-2}$)</th>
<th>$n_{F2} \times 10^{15}$ (cm$^{-2}$)</th>
<th>$n_F/n_F$</th>
<th>$n^* \times 10^3$</th>
<th>$\Delta E_F$ (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.8</td>
<td>7.25</td>
<td>0.59</td>
<td>0.081</td>
<td>90.6</td>
<td>1.62</td>
</tr>
<tr>
<td>2</td>
<td>13.0</td>
<td>1.13</td>
<td>0.087</td>
<td>65.0</td>
<td>2.26</td>
</tr>
<tr>
<td>4</td>
<td>17.8</td>
<td>1.67</td>
<td>0.094</td>
<td>44.5</td>
<td>3.30</td>
</tr>
<tr>
<td>6</td>
<td>19.9</td>
<td>2.06</td>
<td>0.104</td>
<td>33.2</td>
<td>4.42</td>
</tr>
<tr>
<td>8</td>
<td>22.4</td>
<td>2.98</td>
<td>0.133</td>
<td>28.0</td>
<td>5.25</td>
</tr>
<tr>
<td>10</td>
<td>24.0</td>
<td>2.90</td>
<td>0.120</td>
<td>24.0</td>
<td>6.13</td>
</tr>
<tr>
<td>20</td>
<td>23.4</td>
<td>5.02</td>
<td>0.216</td>
<td>11.7</td>
<td>12.6</td>
</tr>
<tr>
<td>30</td>
<td>21.2</td>
<td>5.32</td>
<td>0.25</td>
<td>7.1</td>
<td>20.7</td>
</tr>
</tbody>
</table>

strong increase of the concentration of complex $F_n$ centers (Fig. 2).

From the dependence $n_F = f(\Phi)$ we can estimate the track radius around the ionic path according to Ref. [12]:

$$n_F = n_{F2}(1 - \exp(-\pi r_F^2 \Phi)),$$

(5)

where $r_F$ is a radius of the cylindrical area where single $F$ centers are dominating. The model supposed that at $r < r_F$ single $F$ centers are dominating whereas at $r > r_F$ complex $F_n$ centers and $F$ centers aggregates are created [7]. When two neighbor tracks overlap ($r < r_F$), single $F$ centers are getting insignificant. From $n_F = f(\Phi)$ (Fig. 2) and Eq. (5) the estimated track radius is $r_F = 11.4$ nm.

3 Discussion and conclusions By irradiation of LiF with 147 MeV Kr ions $F$ centers versus fluence have an initial linear stage (Fig. 2). At higher fluences the dependence $n_F = f(\Phi)$ becomes nonlinear with the saturation of single $F$ centers at $\Phi \approx 10^{12}$ ions/cm$^2$ (Fig. 2). At the fluence of $10^{12}$ ions/cm$^2$ the mean distance between the ion impacts (tracks) $d_t$ is about 11 nm comparable with track radius $r_p = 11.4$ nm (Eq. 5) which illustrates the track overlapping. By track overlapping the increase of the concentration of $F_n$ centers occurs faster than in the initial stage at $\Phi < 10^{12}$ ions/cm$^2$. The increase of $F_n$ centers after saturation of single $F$ centers is in a good agreement with the theory of aggregation processes in alkali halides [13]. Such processes occur at a volume concentration of single $F$ centers around $10^{19}$ cm$^{-3}$. At such high volume concentration single $F$ centers interact with formation of more complex defects ($F_n$ centers, $F$ center aggregates ($n_F$). Li colloids [13]. In our experiments at the saturation fluence the volume concentration $N_F = n_F/R \approx 1.4 \times 10^{19}$ cm$^{-3}$ stay in excellent agreement with the theory [13].

At high fluences where $F$ centers aggregate and a strong increase of the $F_n$ center concentration takes place, also primary $H$ centers can form fluorine molecules and their aggregate. Such aggregation was observed in LiF irradiated with low energy Au ions [14]. We suppose that similar primary $H$ center coagulation takes place also in LiF irradiated with Kr ions at high fluences.

The color center parameters ($n_F$, $n_{F2}$, $n^*$, and $\Delta E_F$) (Table 2) are in a good agreement with former results [2, 5, 6]. With increasing fluence the number of single $F$ centers per track ($n^*$) decreases and the average energy to create an $F$ center ($\Delta E_F$) increases. This demonstrates the strong recombination losses (annihilation of primary Frenkel pairs) during ion irradiation at higher fluences.

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References