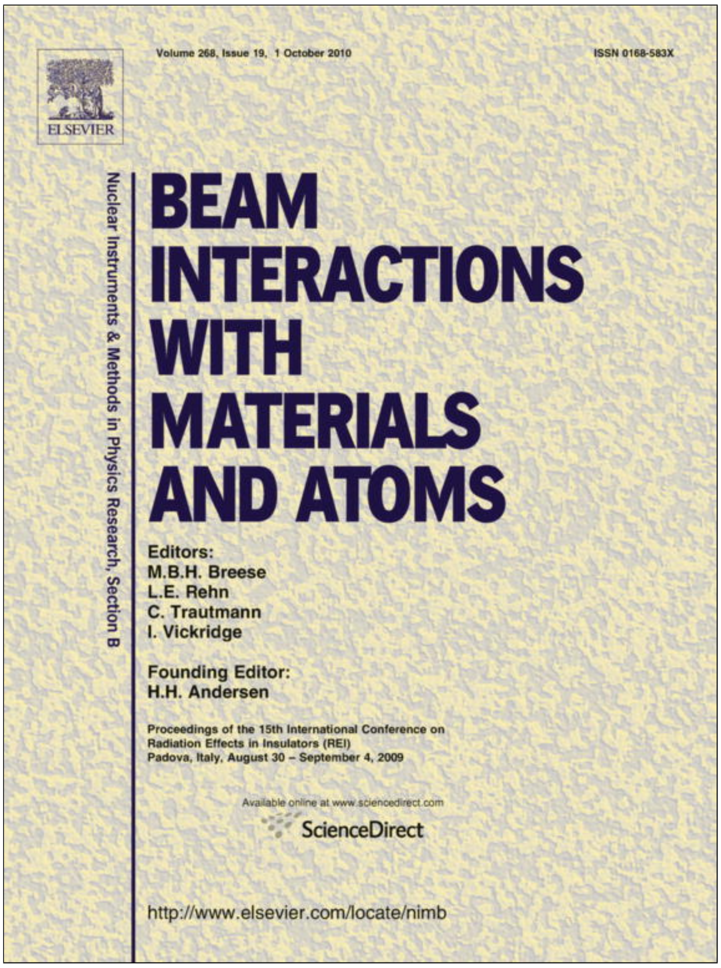

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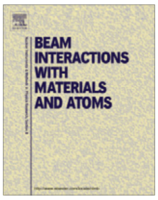

Color center creation in LiF crystals irradiated with Xe, Kr and N ions: Dependence on fluence and beam current density

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ABSTRACT

Single LiF crystals were irradiated with Xe (195 MeV), Kr (117 MeV), and N (18 MeV) ions. Using absorption spectroscopy, color center creation was analyzed as a function of the ion energy loss, fluence, and flux. The concentration of single F centers and F_2 centers versus fluence and flux exhibits a nonlinear evolution with saturation at higher fluences. For LiF irradiated with N ions at high fluence, the concentration of F centers is proportional to the cube root of the flux indicating the strong interaction of primary hole centers. Macroscopic hillocks were observed in all irradiated LiF crystals by atomic force microscopy.

1. Introduction

Heavy ion induced damage in LiF and other alkali halides 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) [3–5]. Therefore, the defect creation strongly depends on the energy loss of the ions.

The aim of this study is investigate peculiarities of damage creation in LiF crystals irradiated with heavy ions at high beam current (flux). Color centers were studied by absorption spectroscopy in the spectral range of 6–1.5 eV (200–850 nm), and atomic force microscopy (AFM) was used for macro-defect studies on the irradiated surface.

2. Ion irradiation

LiF crystals (GOI, St. Petersburg, Russia) cleaved along one of the (1 0 0) plane with the thickness of 1 mm were irradiated at the ion cyclotron accelerator DC-60 (Astana, Kazakhstan). All experiments were carried out at room temperature with normal beam incidence. The irradiation for 195 MeV Xe ions ranged from 6×10^{10} to 10^{13} ions/cm^2 with a flux (φ) from 6.24×10^9 to 10^{10} ions/cm^2 s, for 117 MeV Kr ions from 6×10^{10} to 4×10^{11} ions/cm^2 with a flux 10^{10} ions/cm^2 s, and for 18 MeV N ions from 2.4×10^{11} to 10^{13} ions/cm^2 with a flux from 3×10^9 to 3×10^{11} ions/cm^2 s. The irradiation parameters are presented in Table 1 [6]. The flux φ depends on the effective charge of the ion (k) and the beam current density i_{beam} (nA/cm^2) and is equal to $\varphi = 6.24 \times 10^9$ $i_{\text{beam}} \times k^{-1}$. The ion range was in all cases smaller than the sample thickness. For all ions and at this range of energy the electronic energy loss was much larger than the nuclear energy loss which can be neglected [1].

3. Results and discussion

3.1. Optical absorption and the concentration of color centers

Absorption spectra were measured in the range 1.5–6 eV which corresponds to the absorption of the main electron color centers in LiF crystals. Stable at room temperature hole centers (V_3) have the absorption maximum in VUV spectral region (10.8 eV) and are not discussed in this study [7]. In the absorption spectra, F and F_2 centers are dominating (Fig. 1). The absorption at $h\nu \leqslant 4$ eV depends on the energy loss and the fluence, as well on the beam current density [1–3]. We studied color center creation via fluence and flux (beam current). At low fluences ($\Phi < 10^{10}$ ions/cm^2) and for lower energy loss ($(dE/dx)_e < 10$ keV/nm) only F and F_2 centers are created with the absorption maximum at 4.95 eV (250 nm) and 2.79 eV (445 nm), respectively. At higher fluences ($\Phi > 10^{11}$ ions/cm^2) and for higher energy losses ($(dE/dx)_e > 10$ keV/nm) and higher beam current density ($i_{\text{beam}} > 50$ nA/cm^2) besides F_2 centers also F_3, F_4 and other aggregate centers are created [2]. The increase of the beam current leads to a higher excitation level during irradiation and an enhancement of the efficiency of color center creation takes place, especially for F_n and F center aggregates [2]. The ratio of the concentration of F to F_n centers depends on several factors:

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Table 1
Irradiation parameters [6].

E_{ion} (MeV)	R (μm)	Effective charge (k)	$(dE/dx)_e$ (keV/nm)	$(dE/dx)_n$ (keV/nm)
Xe, 195	17.6	+20	18.85	0.052
Kr, 117	15.3	+13	12.11	0.026
N, 18	11	+2	1.65	0.0013

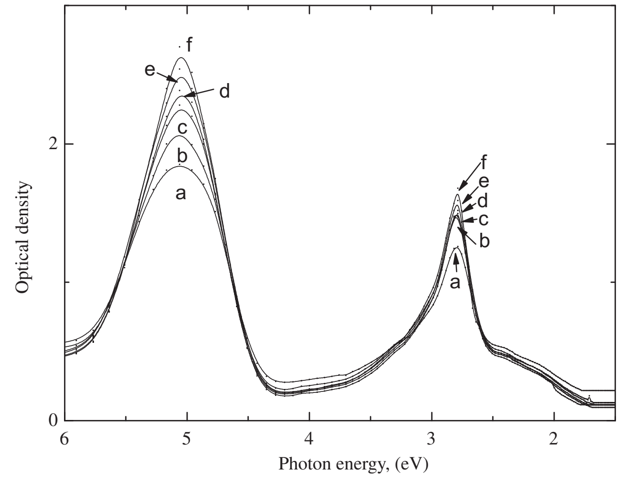

Fig. 1. Absorption spectra for LiF crystals irradiated with 195 MeV Xe ions at a fluence $\Phi = 9.5 \times 10^{12}$ ions/cm^2 with various beam current density [nA/cm^2]: a, 20; b, 35; c, 50; d, 65; e, 80; f, 100.

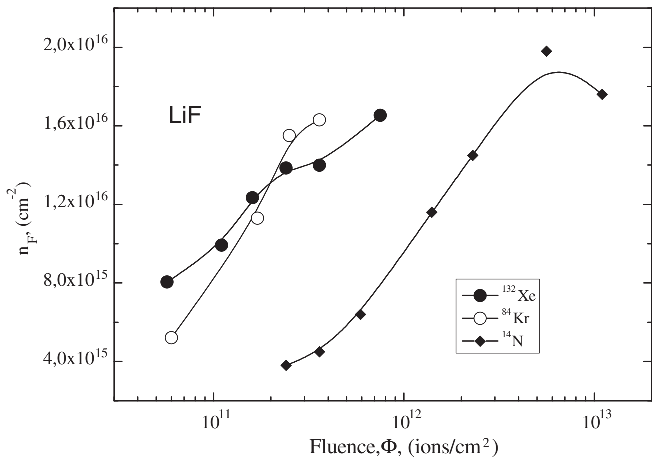

Fig. 2. Concentration of F centers (n_F, cm^{-2}) via fluence (Φ) in LiF crystals irradiated with ^{14}N, ^{84}Kr, and ^{132}Xe-ions at a constant beam current density of 10 nA/cm^2.

(1) the energy loss of the ion, (2) the fluence, and (3) the beam current (flux).

It is difficult to estimate the exact concentration of F_n centers due to the overlapping of various F_n centers and F center aggregates [1,2]. Therefore, we used an approximation and estimated the whole concentration of various F_n centers using the optical density of F_2 centers (the absorption of F_2 centers is dominating in the spectral region $h\nu \leqslant 4$ eV) [1].

The efficiency of color center creation was estimated from the concentration of F and F_2 centers, the number of F centers in a single track, and the energy to create an F center ΔE_F.

The concentration of created F and F_2 centers was estimated from the optical density at the absorption maxima [1,2] according to $n_F[\text{cm}^{-2}] = 9.48 \times 10^{15} \times D_{opt}(F)$ and $n_2[\text{cm}^{-2}] = 4.421 \times 10^{15} \times D_{opt}(F_2)$. The average volume concentration of F and F_2 centers can be estimated as $N_F = n_F/R$ and $N_2 = n_2/R$, where R is the ion range.

The number of F centers in a single ion track n^s is equal to $n^s = n_F/\Phi$. From n^s, we can estimate the average energy for F center creation $\Delta E_F = E_{ion}/n^s = E_{ion} \times \Phi/n_F$. The value of ΔE_F characterizes the efficiency of color center creation under various irradiation conditions [1–3].

3.2. Efficiency of color center creation via fluence

We analyzed the efficiency of color center creation versus fluence and beam current density. In Fig. 1 the absorption spectra are presented for LiF irradiated with Xe ions at a constant fluence ($\Phi = 9.5 \times 10^{12}$ ions/cm^2) with various beam current. We observed only a small increase of the absorption for the F and F_n centers by increasing i_{beam} from 20 to 100 nA/cm^2. The influence of the beam current density was much smaller than in the study of Lushchik et al. for LiF irradiated with 5 and 10 MeV Au ions [2]. This, probably, can be explained by the higher energy and higher $(dE/dx)_e$ for Xe ions which leads to a saturation of color centers at $\Phi = 9.5 \times 10^{12}$ ions/cm^2.

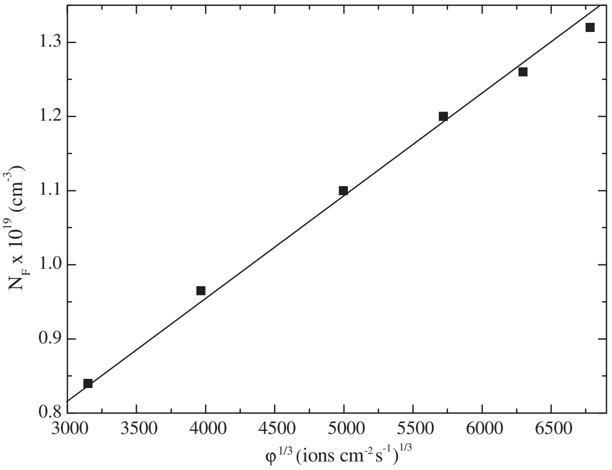

Fig. 3. In LiF irradiated with 18 MeV N ions at a constant fluence of 9.5×10^{12} ions/cm^2 the volume concentration of F centers N_F is proportional to $\varphi^{1/3}$.

Table 2
Color centers in ion irradiated LiF crystals at a constant beam current density of 10 nA/cm^2.

Ion	Φ (ions/cm^2)	E_{abs} (eV/cm^2)	$n_F \times 10^{15}$ (cm^{-2})	n_2/n_F	$n^s \times 10^4$	ΔE_F (keV)
^{132}Xe	5.71×10^{10}	1.10×10^{19}	8.1	0.17	14	1.4
	7.51×10^{11}	1.51×10^{19}	16.5	0.46	2.2	8.8
^{84}Kr	6.1×10^{10}	7.10×10^{18}	5.2	0.11	8.7	1.3
	3.61×10^{11}	4.21×10^{19}	16.3	0.15	4.5	2.6
^{14}N	2.41×10^{12}	4.30×10^{19}	3.8	0.14	1.5	1.1
	10^{13}	1.81×10^{20}	17.6	0.47	0.1	11.3

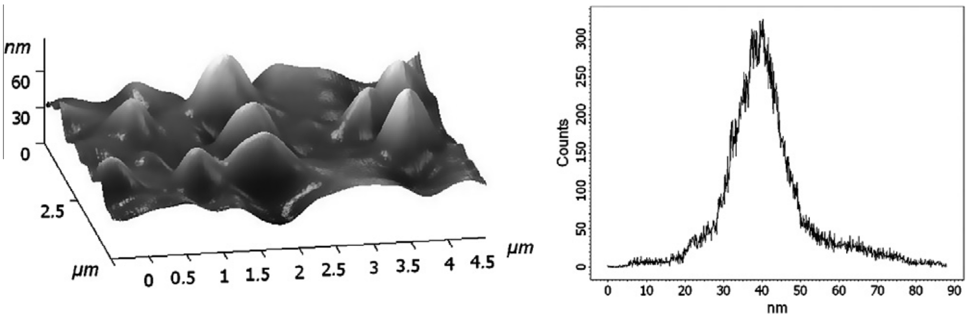

Fig. 4. Macro-defects on the surface of irradiated LiF crystals: surface hillocks in LiF crystals irradiated with 195 MeV Xe ions ($\Phi = 5.7 \times 10^{10}$ ions/cm^2) in the AFM. The height of the hillocks is $h = 39 \pm 5$ nm.

We analyzed the color center creation via fluence for LiF irradiated with Xe, Kr, and N ions at a constant beam current of 10 nA/cm^2 (Fig. 2) and Table 2). For all ions the concentration of F the value n$_F$ increases with the fluence and at higher fluences saturates (Fig. 2). For LiF irradiated with N ions at fluences above 5×10^{12} ions/cm^2, the concentration of F centers decreases which demonstrates the coagulation of single F centers (transformation of single F centers into complex F centers, $F \rightarrow F_n$). Such decrease of the concentration of single F centers is typical for irradiation with high absorbed energy where the volume concentration of F centers is about 10^{19} cm^{-3} [1,2]. In the case of LiF irradiated with N ions, N$_F$ is about 2×10^{19} cm^{-3} (Table 2).

The energy ΔE_F for LiF irradiated with various ions decreases with the fluence (Table 2). This demonstrates the decrease of the efficiency of color center creation by recombination losses of electron and hole centers at high absorbed energy where the concentration of color centers is high and the mean distance between the centers decreases. Note, that for similar absorbed energy E_{abs} the value of ΔE_{abs} is approximately the same (Table 2).

The ratio n_2/n_F depends on the energy loss of the ion and increases with the fluence where neighbor tracks overlap (Table 2). These results are in agreement with former investigations [1–3].

From the dependence $n_F = f(\Phi)$, we estimate the track radius r_F for single F centers according to Thevenard et al. [8]. The track radius r_F describes a cylinder around the ion path where single F centers are dominating. At higher fluences track overlapping takes place and at $r \geqslant r_F$ complex F$_n$ centers are dominating due to interaction of single F centers ($F + F \rightarrow F_2$, $F + F_2 \rightarrow F_3$, etc.). The track radius r_F increase with the energy loss of the ions and for LiF irradiated with N, Kr, and Xe ions are equal to 4, 12, and 17 nm, respectively. These results are in a good agreement with former investigations [1,3].

3.3. Dependence of F center concentration on the flux

We analyzed the kinetic of F center creation via the flux (φ) for a constant fluence of $\Phi = 9.5\text{x}10^{12}$ ions/cm^2. For LiF irradiated with N ions, and we found the volume concentration of F centers depends on $\varphi^{1/3}$, i.e., $N_F \sim \varphi^{1/3}$ (Fig. 3). Such dependence was first observed and analyzed for LiF irradiated with Au ions [9]. At high fluences and high beam current (flux) 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) [6,9]. At high excitation density the concentration of H centers is high and interaction of two H centers can create 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. The di-halide molecules can further form gaseous fluorine filled bubbles (nX_2) which are complementary hole center products to intrinsic colloids [2,9].

3.4. Macro-defects on the surface of irradiated LiF crystals

As mentioned above, the ion track morphology depends on the energy loss of the ions [1–3]. If $(dE/dx)_e$ exceeds a critical value of about 10 keV/nm, track core damage is created [1,10]. We analyzed the macroscopic core damage in our irradiated LiF crystals using track etching and atomic force microscopy.

Using the standard etch solution of hydrofluoric and glacial acetic acid (see [10]), we observed pyramidal etch pits in LiF irradiated with 117 MeV Kr and 195 MeV Xe ions. This demonstrates the presence of core damage in our irradiated samples.

We also used AFM macroscopic damage studies on the irradiate LiF crystal surface. In all irradiated LiF crystals we observed hillocks. In Fig. 4 the scan in the AFM is shown for LiF irradiated with Xe ions. The height of the hillocks depend on the energy loss of the ion and are equal to 13, 24, and 39 nm for N, Kr, and Xe ions, respectively [11]. Nevertheless, the mechanism of core damage creation and the microstructure of the macro-defects is an open question.

4. Conclusions

The ion induced color centers in irradiated LiF crystals depend on the energy loss and fluence, as well on the ion beam current (flux). For all ions the efficiency of color center creation at a constant fluence increases with increasing the beam current (flux). 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 [8]. We observed also track etching and surface hillocks in irradiated LiF crystals.

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