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Color center accumulation in LiF crystals under irradiation with MeV ions: Optical spectroscopy and modeling

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ABSTRACT

Lithium fluoride crystals irradiated at room temperature with energetic ^{14}N and ^{84}Kr ions were studied by absorption and luminescence spectroscopy. Dependence of the average F-center concentration on the fluence was found to be different for the lighter and the heavier projectiles. In particular, the maximal concentration of the F centers reached a notably higher value in the case of irradiation with ^{14}N than that for ^{84}Kr ions. This result is compared with our model of track overlapping and defect accumulation.

The luminescence studies allowed distinguishing F_3^+ and F_2 centers, having very similar absorption spectra. The luminescence corresponding to both centers was observed starting from the minimal fluencies. It means that the aggregation processes take place in single tracks in agreement with our model, which predicts the coagulation radius of about five lattice parameters. Anion vacancies v_a^+, having higher mobility than F centers, should play an important role in the aggregation.

1. Introduction

Radiation damage in LiF crystals, including the effects of swift ions, was a subject of intensive studies since the development of ion accelerators in the 70ths of the last century [1–4]. Nevertheless, the nascent experimental abilities reveal some new aspects of the ion-irradiation effects, such as processes at high fluence [5,6] and creation of nano-defects [7,8].

Ion induced electronic excitations in ionic crystals (electron-hole pairs and self-trapped excitons), create Frenkel pairs in the anion sub-lattice. Although both neutral (F and H) and charged ($v_a^+ + X_{int}^-$) pairs can be generated, only the F centers remain stable at room temperature, whereas the interstitials (H and X_{int}^-) and the anion vacancies v_a^+ are mobile and can interact with other defects, in particular forming complex color centers and aggregates [9,10–15]. High-fluence irradiation increases the defect concentration and enforces their interaction, which leads to a sub-linear fluence dependence of the F-center concentration. Furthermore the intensive creation and growth of the aggregate centers can even decrease the concentration of the F single centers at high fluences. This effect can take place under various irradiations, including electrons, γ-rays and neutrons [16]. However the ion irradiation gives an important advantage for this study, since it is possible to employ the optical spectroscopy measurements of color center concentrations even at high dose (absorbed energy) due to the limited irradiation depth [2,5].

In our previous paper [9] we studied the peculiarities of F_2 and F_3^+ center creation in LiF irradiated with 150 MeV Kr ions. Having the very close absorption maximum around 445 nm, these centers can be distinguished only by luminescence measurements [17,18], which were applied in the present study. The luminescence measurement combined with absorption spectroscopy was aimed to analyze the electron color center creation and aggregation in LiF irradiated with the ^{14}N and ^{84}Kr ions, having different energy loss. The experimental results were compared with a model of track overlapping and defect accumulation.

2. Irradiation

High quality LiF crystals grown from the melt in the inert atmosphere (Optical institute, St. Petersburg) were used for the experiments. LiF platelets of $10 \times 10\ mm^2$ and the thickness of about 1 mm, cleaved along the (001) plane, were irradiated at the cyclotron DC-60 (Astana, Kazakhstan) with N and Kr ions (Table 1). The thickness of the samples for all irradiations was larger than the ion range [19]. The irradiations were performed at room temperature at fluences Φ from 10^{10} up to 10^{15} ions/cm^2 at a constant beam

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Table 1
The ion parameters [19].

Ion	Energy E_{ion}, MeV	Range R, µm	E_{ion}/R, keV/nm
$^{14}N^{2+}$	14	8.65	1.62
$^{14}N^{3+}$	23	14.13	1.63
$^{84}Kr^{14+}$	150	17.76	8.45

current density of 10 nA/cm^2, corresponding to the fluxes of 3.12×10^{10} (N^{+2}), 2.08×10^{10} (N^{+3}) and 4.46×10^9 ions/(cm^2 × s) (Kr^{+14}). The sample holder was cooled with room temperature water and the crystal heating during irradiation was negligible.

For both ions the energy loss does not exceed the threshold of the track core formation [2]. However it is interesting to note that the high-fluence irradiation of LiF crystals either with N or Kr ions corresponds to the absorbed energy per atom much higher than the forbidden gap of the material (14.6 eV) or the binding energy.

3. Absorption and luminescence measurements

Absorption spectra of the irradiated LiF crystals were measured with a double beam UV–VIS spectrometer in the spectral range from 190 to 800 nm (6.5–1.6 eV). The spectra for LiF irradiated with 14 MeV N and 150 MeV Kr ions are presented in the Fig. 1a and b. The absorption of the crystals irradiated with 23 MeV N ions is similar to that of 14 MeV N ions. The integral absorptions

$$A_F = \int_{4.13}^{5.90} D(\varepsilon)\, d\varepsilon, \qquad A_{Fn} = \int_{1.77}^{4.13} D(\varepsilon)\, d\varepsilon \tag{1}$$

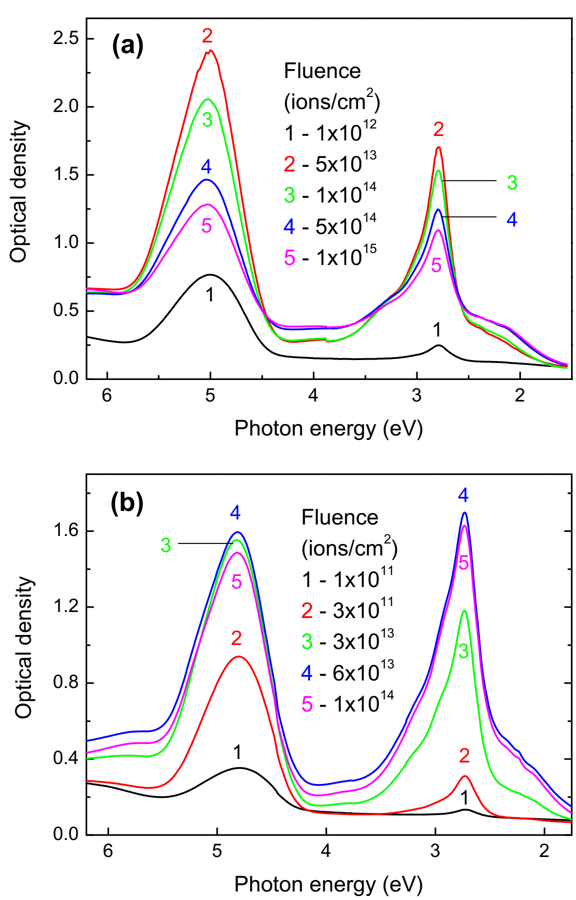

Fig. 1. Absorption spectra of LiF crystals irradiated with (a) 14 MeV N ions and (b) 150 MeV Kr ions.

can be used for the estimation of the relative concentration of color centers at high fluences where the absorption of various color centers overlaps.

The number of F centers per surface area n_F can be extracted from the spectra by means of the Smakula-Dexter formula [2,20].

$$n_F\left[\mathrm{cm}^{-2}\right] = 9.48 \times 10^{15} D_F \tag{2}$$

where D_F is the optical density at the absorption peak of F centers (248 nm). The average volume concentration of F centers can be estimated as

$$\langle N_F \rangle = n_F / R \tag{3}$$

It is interesting to plot this value versus the average absorbed energy

$$\langle E_{abs} \rangle = E_{ion}\Phi / R \tag{4}$$

for different ions (Fig. 2). It turns out than $\langle N_F \rangle$ is not straightforward determined by the average absorbed energy, but depends on it in a complex manner peculiar to different projectiles. We will discuss the most prominent features of this dependence.

In the case of N ion irradiation the absorption increases up to saturation of F centers at about 3×10^{13} ions/cm^2, and at higher fluence a strong decrease of both F and F_n centers takes place (Figs. 1 and 2). The decrease of the integral absorption for F centers A_F from the saturation fluence up to $\sim 10^{15}$ ions/cm^2 reaches 30%, whereas the value of A_{Fn} remains approximately the same taking into account the increase of the absorption around the F_2 and F_3^+ centers (Fig. 1a), and the ratio A_{Fn}/A_F increases from 0.5 (at $\Phi = 3 \times 10^{13}$ ions/cm^2) to 0.78 (at 10^{15} ions/cm^2). Such behavior demonstrates the aggregation of F centers above the fluence of $\sim 3 \times 10^{13}$ ions/cm^2. The decrease of F centers is connected with F_n center and some aggregate center formation.

The absorption of LiF irradiated with 150 MeV Kr ions is more prominent for F_n centers (Fig. 1b). The F centers saturate at the fluence of 10^{12} ions/cm^2 which is lower than that for LiF irradiated with N ions. The magnitude of $A_{Fn}/A_F \approx 0.8$ remains almost constant in the whole fluence range up to 10^{14} ions/cm^2. No remarkable decrease of F and F_n centers at high fluences takes place (Figs. 1b and 2).

Luminescence emission and excitation spectra were obtained using a CM 2203 spectrophotometer. The irradiated LiF crystals were excited with monochromatic light of 445 nm, and the emission spectra were measured (Fig. 3). In all irradiated LiF crystals we observed the emission bands with the maxima at 540 and

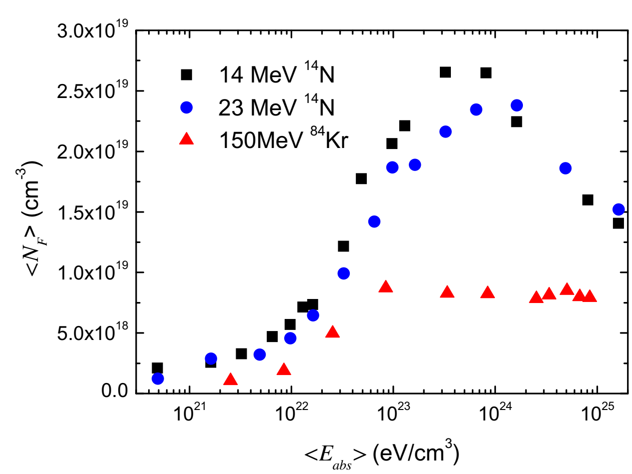

Fig. 2. Dependence of the average F-center volume concentration on the average absorbed energy for LiF irradiated with N and Kr ions.