GSI-Preprint-98-32
Juli 1998

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TRACKS IN LITHIUM FLUORIDE**

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(Accepted for publication in Nucl. Instr. and Meth. B)

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Scanning Force Microscopy of Heavy-Ion Tracks in Lithium Fluoride

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Abstract.

Single-crystal samples of lithium fluoride are irradiated normal to the (100) lattice plane with nickel, gold, bismuth, and uranium ions with kinetic energies of several tens of MeV to some GeV. The ion-induced damage on the surface is inspected by scanning force microscopy (SFM) under ambient conditions. SFM reveals circular damage zones, consisting of small hillocks. The mean diameter of the track cross sections depends linearly on the ion-energy loss and increases from about 20 nm for Ni ions to almost 70 nm for U projectiles. The hillocks exhibit average heights of 0.5-2 nm.

PACS: 07.79.Lh, 61.72.Ff, 61.80.Jh, 78.55.Fv

1 Introduction

Irradiation-induced damage in alkali halide crystals, caused by electrons, neutrons, X- and gamma rays, and recently also by energetic heavy ions has been studied intensively by numerous groups. A considerable part of the effort focused on lithium fluoride (for a review of early work see for example [1,2]). For the sake of brevity, a few remarks may suffice here to illustrate the wide spectrum of aspects. It is worthwhile mentioning that among all ionic crystals LiF has the most pronounced ionic binding character and the highest binding energy. It is a suitable window material for light in the vacuum ultraviolet regime. Laser emission in the infrared is feasible due to the formation of colour centers [3]. Etching of radiation damage in LiF has been reported already in the fifties [4,5]. LiF represents a radiation dosimeter of high sensitivity, characterized by longtime-storage of the absorbed energy without significant fading and by an efficient release of the stored energy via luminescence [6]. In addition, it allows high-dose measurements via absorption spectroscopy. X-ray diffraction and density measurements revealed that radiation induced damage in LiF consisted of Frenkel defects [7,8]. In LiF, the primary Frenkel defects are F- and H-centers (H-centers being stable only at $T \leq 60$ K) which are transformed into more complex defects at higher irradiation temperatures and higher doses. At high doses, defect aggregates like F-center clusters, alkali colloids, and concentrations of diatomic halogen molecules are formed [4,9]. Techniques such as nuclear magnetic resonance and electron spin resonance (ESR) proved the occurrence of metal aggregates [1]. Furthermore, by means of ESR and absorption spectroscopy, polaron states (self-trapped holes) were found for the first time in LiF [10].

Recently, we reported first results of scanning force microscopy (SFM) of LiF single crystals after exposure to gold ions with a kinetic energy of about 2.2 GeV [11]. This present paper contains the extension of our SFM measurements on LiF samples irradiated with different heavy-ion species with energies of several tens of MeV to several GeV. This work complements studies of ion-irradiated LiF performed with optical spectroscopy, small-angle X-ray scattering, chemical etching, and surface profilometry [12-21].

As far as the application of SFM to LiF is concerned, it should be mentioned that the surface of undamaged LiF crystals has been imaged with atomic resolution by several groups [22-26]. We would also like to emphasize that SFM has become a widely used tool for the characterization of damage caused by heavy-ion impact [27-42].

2. Experimental

2.1 Sample preparation and ion irradiation

Samples with a thickness of about 0.5 mm were cleaved from a lithium fluoride single-crystal of high purity. Most of the samples were irradiated with Au, Bi, and U ions at the linear accelerator (UNILAC) of GSI. The projectiles had a specific energy of 11.4 MeV/u and for Bi also 4.1 MeV/u. Further, some irradiations were performed with Ni ions of 1.4 MeV/u at the GSI-High-Charge Injector (HLI). For a list of ion parameters see Table 1. All crystal platelets were irradiated normal to the (100) lattice plane and at room temperature, with fluences in the range of 1×10^{10} - 3×10^{11} ions/cm². During the irradiation with Ni-ions the outgassing of fluorine from the target was registered. We conclude that this occurs also for higher energy losses.

2.2 Scanning force microscopy measurements

The LiF surfaces that had been exposed to the ion beam were imaged by scanning force microscopy under ambient conditions. As for previous measurements [11], the microscope was operated with a commercial bar-shaped Si₃N₄ cantilever at loading forces of 5-100 nN. In all cases, topographic and lateral-force images were acquired simultaneously.

The Figs. 1a and 1b show a topographic and a lateral-force image ($2 \times 2 \mu\text{m}^2$) of a LiF single-crystal surface irradiated with Bi ions of 11.4 MeV/u at a dose of 1×10^{10} ions/cm². The micrographs were acquired simultaneously by fast scanning from top to bottom. A magnified section ($840 \times 840 \text{ nm}^2$) of Fig. 1b is depicted in Fig. 1c. It should be emphasized that the

contrast of a lateral-force image represents a convolution of friction and of the derivative of the surface profile (i.e. the slope) with respect to the direction of fast scanning [43,44].

3. Results

As we reported in the recent past, Au ion projectiles caused hillocks on the LiF surface [11]. It was found now that hillocks are created already by the impact of Ni-ions with a significantly lower energy loss. We preferably extracted the hillock diameters from lateral-force micrographs, since they provide a higher contrast than the related topographic images. For this purpose, line profiles were drawn through the track cross sections normal to the direction of fast scanning. As an example, Fig. 1d shows the (inverted) lateral-force profile along the straight line from A to B in Fig 1c. We define the diameter of an individual track cross section by the distance between a left (F_l) and a right foot (F_r) that mark the transition from the contour of the track to the horizontal baseline representing the undamaged crystal surface [37,11]. For each ion species, the number of tracks was plotted as a function of the diameter. Such a histogram is displayed for Bi-ion tracks (4.1 MeV/u) in Fig. 2. Mean hillock diameters were extracted by fitting a Gaussian curve to each of these distributions. In Fig. 3, the mean diameters are plotted as a function the electronic energy loss, the vertical error bars representing $\pm 1\sigma$. The energy loss values have been calculated by means of the TRIM code [45]. Due to uncertainties inherent to the code, error bars of about $\pm 15\%$ are taken into account.

Another track parameter of interest is the mean height of the hillocks. Applying the same procedure as for the evaluation of the diameters, the heights were extracted from topographic images. All mean diameters and heights are listed in Table 1.

The creation of individual hillocks, each by the impact of a single energetic heavy ion, has been found by SFM for example also for LiNbO₃ [39] and for mica [41]. Similar to LiF, the heights of the hillocks were of the order of 1-2 nm for both materials. We would like to add that mica develops significantly larger track diameters on the surface exposed to the ion beam than

in the interior as shows the comparison with track images acquired after removing a thin surface layer by cleavage. Presently, it is not known whether this size effect occurs also in LiF.

4. Conclusions

Our earlier SFM measurements of hillocks induced by Au ion impact on the surface of LiF single-crystals [11] were extended to Ni, Bi, and U ions. In particular, we succeeded in imaging also the impact zones of the light Ni ions with an electronic energy loss of about 9.7 keV/nm. The mean diameter of the tracks occurring on the original LiF surface as a function of energy loss shows a linear increase, similar to ion tracks in the bulk of materials like mica [27,28,37], GeS [46], and others. Remarkably, the diameter (extracted via absorption spectroscopy [19]) of the cylindrical ion track region in which single F-centers prevail, is of the same order of magnitude as the hillock diameters.

The LiF hillocks have mean heights of 0.5 to 2 nm, however the presently available results do not yet allow a conclusion about the functional dependence on the energy loss. We suppose that besides the crucial role of surface parameters, structural changes in the bulk participate in the hillock formation. For example, the exposure to X-rays caused a volume expansion and a simultaneous density decrease, resulting from F-center creation [7,8]. Recently, the occurrence of a step-like out-of-plane swelling under ion irradiation was found [21]. These observations do not straightforwardly provide a quantitative or qualitative understanding of the mechanisms underlying the creation of hillocks with heights of 0.5-2 nm. The observation of fluorine outgassing during Ni-ion irradiation gives rise to the assumption that also an enrichment of fluorine molecules in the surface region should be taken into account.

Future SFM studies of ion irradiated LiF crystals, including measurements in an ultrahigh-vacuum chamber flanged to our beam line of the GSI-High-Charge Injector, intend to improve the data of the mean diameter and height of the hillocks as a function of the ion energy loss. Conceivably, SFM can provide access also to ion track parameters inside the bulk.

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Figure captions

Fig. 1. a) Topographic and b) lateral-force image ($2 \times 2 \mu\text{m}^2$) of a LiF single-crystal surface irradiated with 1×10^{10} Bi ions/ cm^2 of 11.4 MeV/u. c) Magnified section ($840 \times 840 \text{ nm}^2$) of the area shown in Fig. 1b. d) Inverted lateral-force profile along the straight line AB in c).

Fig. 2. Histogram of hillock diameters (4.1 MeV/u Bi ions).

Fig. 3. Mean hillock diameters vs electronic energy loss.

Table 1. Irradiation of lithium fluoride single-crystals: Ion parameters and track characteristics

Ion	Fluence	Kinetic energy	Energy loss ¹⁾	Hillock diameter ²⁾	Hillock height ²⁾
	[ions/cm ²]	[Mev/u]	[kev/nm]	[nm]	[nm]
Ni	3x10 ¹¹	1.4	9.7	19.9 (4.5)	1.0 (0.4)
Au	5x10 ¹⁰	11.4	24.0	55.3 (17.5)	1.1 (0.5)
Bi	1x10 ¹⁰	11.4	25.4	62.6 (10.0)	0.6 (0.3)
Bi	1x10 ¹⁰	4.1	28.4	62.6 (10.4)	1.0 (0.4)
U	5x10 ¹⁰	11.4	28.9	68.3 (10.3)	2.0 (0.5)

1) The values represent the electronic energy loss within the first few microns of the ion trajectory in LiF.

2) The error corresponds to σ

Fig. 1a

Fig. 1b

Fig. 1c

Fig. 1d

Fig. 2

Fig.3