Nuclear Instruments and Methods in Physics Research B 326 (2014) 307–310

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Modeling of defect accumulation in lithium fluoride crystals under irradiation with swift ions

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ARTICLE INFO

Article history:
Received 21 June 2013
Received in revised form 22 October 2013
Accepted 23 October 2013
Available online 23 January 2014

Keywords:
Ion irradiation
Color centers
Defect accumulation
Absorbed energy

ABSTRACT

In many materials electronic excitations created around the trajectories of swift ions result in defect creation. Experimental observations often yield information on integral damage effects. The presented approach suggests a theoretical model to correlate integral damage results with microscopic effect produced by overlapping of individual single ion tracks. The model is applied to ion-beam induced defects in LiF crystals. Two aspects are treated separately viz. the ion-deposited energy distribution for a given fluence and the material response to the absorbed energy. The first problem is treated within the framework of stochastic superposition of ion tracks, taking into account the radial distribution of the energy transfer of a single ion. For lithium fluoride the creation of color centers is considered as the materials response. The dependence of the defect concentration on the absorbed energy is included in order to obtain the integral defect production.

1. Introduction

In various materials, in particular in insulators swift ions produce radiation damage around their trajectories [1–3]. Tracks of individual ions can be analysed by microscopic methods such as transmission electron microscopy (TEM), scanning force microscopy (SFM) or by other direct techniques analysing non-overlapping ions such as small angle X-ray spectroscopy (SAXS) [4–6]. However, many other techniques provide only integral values of the irradiated surface and track size information is deduced indirectly from fluence series. These indirect techniques include optical absorption spectroscopy, luminescence measurements, X-ray and Raman analysis, electron and nuclear magnetic spectrometry, phosphorus afterglow measurements [7] and many others.

A simple model considering track overlapping effects was first suggested for elastic collision cascades [8] and later applied for swift ions [9–12]. This model assumes that an experimentally measured integral value, which characterizes the damage production, is proportional to the irradiated area A_i of the sample surface according to

$$A_i = A(1 - \exp(-a\Phi)) \quad (1)$$

where A is the area of the sample surface, Φ is the ion fluence, and a is the effective cross-section of the track [9,10]. Extensions of this model took into account some threshold, when double or multiple hit processes are required for damage production [8,11,12]. This model considers a homogeneous damage within the track cross section and as such does not take into account the radial energy distribution around the projectile trajectory. It also ignores specific damage accumulation processes during multiple track overlapping. In this approach it is also difficult to distinguish damage saturation at high fluences (according to Eq. (1)) from saturation given by the material response, which would likewise take place for some uniform high-dose irradiations. Here we propose an alternative approach, which considers track overlapping processes in a more realistic manner by including radial form factors.

2. Accumulation of absorbed energy

We can select an arbitrary point on the material surface. Assuming an equiprobability surface distribution of a single ion impact within the circle of radius R around the selected point, the distribution function for the distance r between this point and the ion ingress is given by

$$f(r) = \frac{2r}{R^2}, \quad 0 \leqslant r \leqslant R \quad (2)$$

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http://dx.doi.org/10.1016/j.nimb.2013.10.033

The radius R should be large enough to provide the negligible cut-off of the absorbed energy. For a given radial absorbed energy distribution $g(r)$ it means

$$2\pi \int_0^R g(r) r dr = dE/dx \tag{3}$$

where dE/dx is the projectile stopping power.

Thus the function

$$f(g) = f(r(g))\left|\frac{dr}{dg}\right| \tag{4}$$

gives the distribution of the absorbed energy in the selected point, provided by the single projectile in the circle $0 \leqslant r \leqslant R$. In the case of n projectiles, the distribution function of the accumulative absorbed energy

$$G = \sum_{i=1}^{n} g_i \tag{5}$$

is given by the convolution of the functions (4)

$$F_2 = f * f = \int f(G-g) f(g) dg F_n = \underbrace{f * f * \ldots * f}_{n} \equiv f^{*n}$$

Note, that all these functions are normalized

$$\int F_n(G) dG = 1 \tag{7}$$

and according to the central limit theorem converge to the normal distributions as $n \to \infty$.

The number of ions n is Poisson-distributed [13]

$$P_n(N) = \frac{N^n \exp(-N)}{n!} \tag{8}$$

where $N = \langle n \rangle$ is the average number of ions in the area of πR^2. For a given fluence Φ it is

$$N = \pi R^2 \Phi \tag{9}$$

Thus the distribution function of the total absorbed energy is given by the series [14]

$$F(G) = \sum_{n=1}^{\infty} F_n(G) p_n(N) \tag{10}$$

where $F_1 \equiv f$.

Since $n = 0$ does not provide the absorbed energy,

$$\int F(G) dG = 1 - p_0(N) = 1 - \exp(-N) \tag{11}$$

Eq. (11) is concordant with the single-impact model [8,9] with $a = \pi R^2$ in the Eq. (1) and gives the surface fraction, which is hit by at least one ion. Deducting sequentially the surface fractions with $n = 1, 2, 3 \ldots$ according to Eq. (8), one can obtain the area irradiated with at least $2, 3, 4 \ldots$ ions correspondingly, and finally obtain the multiple hit model, suggested by Gibbons [8,11,12]. Such a model is suitable in the case of a sharp cut-off threshold such as open pores have in a track-etch polymer membrane [1], but it does not describe the correct situation in the case of gradual damage accumulation from the subsequent projectiles, when the distribution function $F(G)$ given by the Eq. (10) has to be considered.

The average absorbed energy, provided by a single ion, is given by:

$$\langle g \rangle = \int g f(g) dg = \int_0^R f(r) g(r) dr = \frac{dE/dx}{\pi R^2} \tag{12}$$

where Eq. (3) is taken into account. Assuming independent ion impacts, the average total absorbed energy can be deduced by using Eqs. 5, 9, and 12:

$$\langle G \rangle = \int G F(G) dG = \langle g \rangle \sum_{n=1}^{\infty} n p_n(N) = \langle g \rangle N = \frac{dE}{dx} \Phi \tag{13}$$

If the radiation damage would be linearly proportional to the absorbed energy G, the average damage would grow linearly with increasing fluence and no information about the track structure could be extracted from the fluence dependence. However the material response to irradiation $\eta(G)$ is non-linear in many materials. For instance the simplest sub-linear model based on the idea of material response saturation in the vicinities of projectile trajectories corresponds to the Eq. (11). Generally the measured value $\langle \eta(G) \rangle$ can be found as:

$$\langle \eta(G) \rangle = \int \eta(G) F(G) dG \tag{14}$$

The response function could be either sub-linear if one considered point defect accumulation or super-linear for complex defects or amorphization [15]. In the following we focus on ion-beam induced defects in lithium fluoride and in particular on accumulation of single color centers (F centers) into more complex color centers (F_2, F_3 centers) and aggregates.

3. Response function of lithium fluoride crystals

Radiation-induced defects in alkali halides have been studied over decades. The peculiar situation of ionic crystals is the fact that the exciton mechanism is a very efficient process for defect creation [2,16]. Defects due to elastic collisions are only important for slow ions of few hundred keV per nucleon [17]. We consider here the absorbed energy dependence of the defect production, but ignore non-linear kinetic effects of the energy density and the irradiation intensity, which could violate the reciprocity principle [18].

We will consider the response function of lithium fluoride $\eta(G)$ as the electron color center concentration, so that $\langle \eta(G) \rangle$ can be measured by absorption spectroscopy. At room temperature, the ionizing irradiation mainly produces F centers (absorption maximum at 248 nm) and F_2/F_3^+ centers (450 nm) [19,20]. Due to their high activation energy (above 1 eV), F centers do not migrate but remain stable at room temperature and can only interact if by chance a defect is created in the close vicinity. The response dependence on the absorbed energy can be extracted from uniform LiF irradiations with electrons, X-rays, or neutrons [21,22]. Another way is the development of a suitable defect accumulation model [23,24].

Let us consider volume V, which includes the volumes V_1 and V_x occupied by single F centers and complex (F_2, F_3, etc.) centers. We introduce the coefficient α so that the energy VdG, absorbed in the volume V, produces the F centers, filling the volume αVdG. The coefficient α should take into account the effect of recombination and can be expressed as:

$$\alpha = \frac{\Delta V}{\Delta E_F} \tag{15}$$

where ΔE_F is the creation energy for a stable F center. The actual value of ΔE_F depends on the F-center concentration due to their recombination with interstitials and therefore depends on the applied fluence. However in this simple model we will not consider this effect. ΔV is the minimal volume per F center preventing coagulation, so that V_1 contains $V_1/\Delta V$ of F centers. Creation of a new F center within the volume ΔV already containing the F center leads to aggregation of a single F-center into F_2-centers: